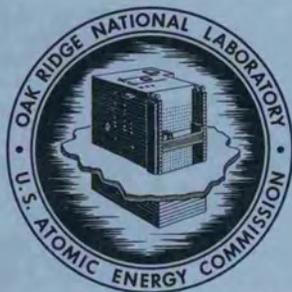


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TECHNICAL PROGRESS REVIEW

ISOTOPES AND RADIATION TECHNOLOGY

VOLUME 8, NO. 2

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Isotopes and Radiation Technology

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DRAFT
TECHNICAL PROGRESS REVIEW
ISOTOPES AND RADIATION TECHNOLOGY
VOLUME 8, NO. 2

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I. ISOTOPE PRODUCTION AND DEVELOPMENT (1)

THE U. S. RADIOISOTOPE AND RADIATION PROCESSING INDUSTRIES--1969 (2)

(DTIE: Please pick up supplementary keywords at end of article)

Editor's Note: The USAEC Division of Industrial Participation prepares a yearly summary of the status of the nuclear industry.¹ The section of this report dealing with the radioisotope and radiation processing industries is presented here because it provides a good summary of those industrial concerns which are active in this field. The editors thank Ernest B. Tremmel, Director of the Division of Industrial Participation, for permission to use this section and Robert J. Berte, also of the Division of Industrial Participation, for his help in preparation of this material for use in this publication.

Abstract: The status of the commercial U. S. radioisotope and radiation processing industries in 1969 is reviewed, and commercial suppliers of the following types of materials and services are listed: radioisotopes, radiochemicals and radiopharmaceuticals, radioisotope sources, radiation equipment, radioisotopic power sources, and radiation processing.

(Introduction)

The commercial radioisotope industry in the United States continued to show a healthy growth in 1969. The market increase in industrial interest in process radiation noted last year continued in 1969.

Because of the many facets of industry's participation in these fields, the discussion is divided into the following sections: radiochemicals and radiopharmaceuticals; radiation-equipment suppliers (including radiography and teletherapy units, radioisotope gages, and self-illuminating materials); radioisotopic power; radiation processing; significant rule changes in the radioisotopes area; and the AEC Advisory Committee on Isotopes and Radiation Development.

Radiochemicals and Radiopharmaceuticals (3)

During the year, the AEC's Division of Isotopes Development conducted an informal survey of the industry and has provided the following market predictions and growth estimates.

The domestic market for radiochemicals and radiopharmaceuticals has increased during the past year by about 25% over 1968. It is

estimated that the radioisotope industry's growth will continue at this rate for the next several years.

Market Predictions
(Estimated Sales in Millions of Dollars)

	<u>1968</u>	<u>1969</u>	<u>1970</u>
Basic Radioisotopes	8	10	13
Radiochemicals	16	20	25
Radiopharmaceuticals	25	31	39
Sealed Sources	<u>4</u>	<u>5</u>	<u>6</u>
Total	53	66	83

The estimated private sales of radiochemicals include the value of AEC-supplied radioisotopes purchased by private suppliers for further processing or repackaging and distribution.

Radioisotopes of commercial interest fall into four classes: those found in nature, such as ^{226}Ra ; those deliberately produced in nuclear reactors, such as ^{60}Co and ^{14}C ; those unavoidably produced in nuclear reactors through the fission process, such as ^{90}Sr and ^{137}Cs ; and those produced in accelerators, such as ^{22}Na and ^{123}I . A radiochemical is a chemical compound prepared with a radioactive element. A radiopharmaceutical is a radiochemical which has been prepared to meet requirements for use in the human body.

The market for radioisotopes ^{should} continue to expand at the rate of 25 % annually as a result of new developments and increasing requirements for applications in areas such as radiation processing, medicine, and isotopic fueled power systems. Industry continues to make significant increases in its capacity to produce, process, and distribute isotopes. While the packaging and distribution of radioisotopes is performed to a great extent by private industry, the AEC is still one of the principal producers and distributors.

As in previous reports, the radioisotope industry is divided into four categories: producers, processors, source fabricators, and equipment manufacturers.

PRODUCERS (4)

A producer is defined as one who can perform target irradiation and may also perform target preparations and separation of the desired radioisotope for ultimate distribution. Radioisotopes are obtained from cyclotron bombardments, reactor irradiations, and by separation from high-level fission product waste streams.

Cyclotron Radioisotopes (5)

→ In 1955, the AEC discontinued processing and distributing cyclotron-produced radioisotopes. Except for target preparation and irradiation services provided by Oak Ridge National Laboratory or by the University of Pittsburgh, the industry supplies cyclotron radioisotopes.

Cyclotrons can generate a wide variety of radioisotopes which cannot be produced in reactors. Many have unique properties which make them especially useful or necessary for certain applications, in spite of their relatively high cost. For example, the use of ^{123}I (with a half-life of 13 hours) instead of ^{131}I , for in-vivo thyroid up-take studies reduces radiation exposure to patients substantially because it emits no beta radiation. It is also very promising for brain scans and cancer research. Because of the special interest in cyclotron-produced ^{123}I , by the medical profession and with the encouragement of the private processors, the Commission makes it available on a special order basis as a processed target, similar to ^{84}Rb and ^{57}Co . Other cyclotron-produced radioisotopes of interest include: ^{22}Na , ^{57}Co , ^{84}Rb , ^{11}C , ^{13}N , ^{15}O , ^{55}Fe , ^{199}Hg , ^{49}Cr , ^{61}Cu , ^{62}Zn , and ^{73}Se .

Because of the very short half life of the radioisotopes desired for medical applications and the difficulty in transporting usable quantities over great distances from the cyclotron, there is a need for commercial cyclotron facilities near large population centers to provide medical researchers and nuclear medicine departments with the necessary isotopes on a timely basis.

Small cyclotrons, designed for medical research and isotope production, are now in use or planned at the following locations:

- Washington University Medical Center, St. Louis, Missouri
(first commercially installed unit)
- Massachusetts General Hospital, Boston, Massachusetts
- Sloan-Kettering Institute, New York, New York
- Argonne Cancer Research Hospital, Chicago, Illinois
- New England Nuclear Corporation, Boston, Massachusetts
- Mt. Sinai Hospital, Miami Beach, Florida
- University of California, Los Angeles, California

The Cyclotron Corporation, Berkeley, California, and Allis-Chalmers, Milwaukee, Wisconsin, are the only producers of small cyclotrons for medical research and isotope production. Allis-Chalmers supplied the Washington University Medical Center and the Massachusetts General Hospital machines. The Cyclotron Corporation supplied the Argonne Cancer Research Hospital, Sloan Kettering Institute, and New England Nuclear Corporation machines, and is supplying the unit ordered for the Mt. Sinai Hospital.

The New England Nuclear and Mt. Sinai Hospital installations were privately financed. The others were funded by the AEC and National Institutes of Health. It has been estimated that a market for some 20 such facilities may develop for medical radioisotope production by 1975.

The present market for cyclotron-produced radioisotopes is estimated at about \$3 million per year. While the U.S. has been the principal supplier, cyclotron radioisotopes are also marketed by the United Kingdom, South Africa, and the USSR.

The principal industrial suppliers of cyclotron radioisotopes are listed in Table 1.

TABLE 1

Principal Industrial Suppliers of
Cyclotron Radioisotopes

Abbott Laboratories, North Chicago, Illinois
 Cambridge Nuclear Corporation, Cambridge, Mass.
 Hastings Radiochemical Works, Houston, Texas
 International Chemical and Nuclear Corporation,
 City of Industry, California
 Isotope Products Lab, Los Angeles, California
 New England Nuclear Corporation, Boston, Mass.
 Nuclear Consultants, Division of Mallinckrodt
 Chemical Company, St. Louis, Missouri

Reactor Radioisotopes. (5)

→As indicated previously, the AEC is still a principal domestic producer and distributor of radioisotopes, including those produced in reactors. However, the industry is offering an increasing number of products and higher specific activity material than in past years.

Commercial reactor irradiation services, for production of radioisotopes in targets supplied by customers, are offered by the following facilities: (1) General Electric Co. (Pleasanton, California), (2) Union Carbide Corporation, (Sterling Forest, New York), and (3) Babcock & Wilcox, (Lynchburg, Virginia). Neutron Products, Inc. (Dickerson, Maryland) has been producing ^{60}Co in the Big Rock Point nuclear power reactor owned by Consumers Power Company.

Up to 1965, the Commission had withdrawn, on an ad hoc basis, from producing and distributing selected radioisotopes. In March 1965, the AEC adopted and published in the Federal Register formal policies and procedures for the transfer of commercial AEC radioisotope production and distribution activities to private industry. These policies and procedures provide for orderly AEC withdrawal from this area of activity.

As of December 1967, the AEC had withdrawn from routine production of a total of 38 reactor-produced isotopes, which are listed in Table 2.

Table 3 lists AEC sales of reactor produced radioisotopes.

TABLE 3

AEC Sales of Reactor Produced Radioisotopes

<u>Fiscal Years</u>	<u>Million Dollars</u>
1963	\$1.2
1964	1.3
1965	1.9
1966	2.3
1967	1.8
1968	1.8
1969	1.4

During the year, the Commission made several public announcements regarding prices, availability, production, and distribution policies for certain isotopes it produces. The significant items are summarized briefly.

In July 1969, the Commission offered to make available on loan, under contract, a high specific activity grade of ^{60}Co for heat source applications research and development of interest to AEC.

In July 1969, AEC made available for sale high purity concentrations of ^{233}U and ^{242}Pu for tracer analytical and alpha standards for instrument calibration. ^{234}U is being made available for the manufacture of in-core fission chambers for nuclear power reactor flux monitoring.

Table 2

WITHDRAWN RADIOISOTOPES

<u>Radioisotope</u>	<u>Effective date of withdrawal</u>
1. Antimony 124	May 1, 1966
2. Antimony 125	April 18, 1965
3. Arsenic 76	May 1, 1966
4. Arsenic 77	May 1, 1966
5. Bromine 82	May 1, 1966
6. Cadmium 109	May 1, 1966
7. Cadmium 115m	May 1, 1966
8. Cadmium 115	May 1, 1966
9. Calcium 45	April 18, 1965
10. Cerium 141	June 23, 1964
11. Cesium 134	June 23, 1964
12. Chromium 51	June 23, 1964
13. Cobalt 58	June 23, 1964
14. Cobalt 60 - greater than 45 Ci/g	May 2, 1961
15. Cobalt 60 - less than 45 Ci/g	March 14, 1968
16. Copper 64	May 1, 1966
17. Gold 198	May 1, 1966
18. Gold 199	May 1, 1966
19. Iodine 125	October 1, 1963
20. Iodine 131	October 1, 1963
21. Iridium 192, metallic sources	February 1, 1964 - May 1, 1965
22. Iron 55	June 23, 1964
23. Iron 59	April 18, 1965
24. Lanthanum 140	May 1, 1966
25. Mercury 197	May 1, 1966
26. Mercury 203	May 1, 1966
27. Molybdenum 99	May 1, 1966
28. Mo-99 - Tc99m generator	July and August 1966
29. Phosphorus 32	May 1, 1966
30. Potassium 42	May 1, 1966
31. Selenium 75	April 18, 1965
32. Silver 110	May 1, 1966
33. Sodium 24	May 1, 1966
34. Strontium 85	October 15, 1964
35. Strontium 87m	April 30, 1967
36. Sulfur 35	May 1, 1966
37. Tin 113	April 18, 1965
38. Zinc 65	April 18, 1965

The AEC's Savannah River Operations Office, in June 1969, announced a market evaluation program to develop potential uses for ^{252}Cf . The unusual characteristics of ^{252}Cf , i.e., its emission of a large number of neutrons through spontaneous fission and its reasonable half-life of 2.6 years, distinguish it from all other commonly available radioisotopes. These characteristics, along with other data and applications, have been set forth in a brochure² and distributed to interested groups which are invited to evaluate the feasibility of many proposed applications and to estimate market potentials. Test sources of ^{252}Cf will be loaned for this purpose.

To date, agreements to loan ^{252}Cf for neutron well-logging have been reached with Texaco, Inc. and Schlumberger Technology, Inc. ^{252}Cf sources have also been loaned to M. D. Anderson Hospital for cancer therapy, to the U.S. Geological Survey for mineral exploration studies, and to the U.S. Bureau of Mines for evaluation in monitoring the sulfur content of coal.

The AEC increased its price for ^{210}Po , effective October 1969, from \$15 to \$80 per curie in order to recover production costs. These costs increased as a result of a decrease in demand, due to the cancellation of the SNAP-29 program to develop a radioisotope thermoelectric generator for space flight missions.

Reactor-produced radioisotopes are used in a large number of applications: to name a few, medicine, agriculture, crime detection, industrial gauges, and space. As indicated in the introduction, the market is estimated to grow at about 25% annually for the next several years.

It is estimated³ that between 3 and 4 million patients are benefiting each year from medical procedures using radioisotopes. About 98% of these procedures are diagnostic. About two U.S. hospitals and over five U.S. doctors and medical organizations in the U.S. routinely use some 30 different radioisotopes in diagnoses and treatment of many diseases and disorders and in research on them. About 8 million individual diagnostic or therapeutic treatments are administered annually. About 50% of the 8 million employ ^{60}Co (primarily for therapy), 33% use ^{131}I (for diagnosis and therapy), and 7% use $^{99\text{m}}\text{Tc}$ (for diagnosis). At least two diagnostic procedures are being performed daily using $^{99\text{m}}\text{Tc}$. The remaining 10% of these procedures use a large variety of other radioisotopes.

Some of the medical applications include: ^{60}Co to treat cancer by radiation; ^{131}I to diagnose thyroid disorders, treat hyperthyroidism, diagnose liver and kidney disorders and make function tests of these organs, locate brain tumors and make blood volume and cardiac output studies;

^{14}C is used to study metabolic diseases; ^{68}Ga , ^{85}Sr , $^{87\text{m}}\text{Sr}$, ^{18}F , and ^{47}Ca are used for bone scanning in suspected metabolic disorders and malignancies; ^{51}Cr to measure cell mass; ^{74}As and ^{197}Hg to locate brain tumors; ^{90}Y for pituitary gland therapy; and ^{22}Na and ^{24}Na are used to measure exchangeable sodium and diagnose circulatory disorders.

Fission Product Radioisotopes (5)

↪ In addition to cyclotron and reactor-produced radioisotopes, fission product radioisotopes are separated from high level waste streams from AEC production facilities. The radioisotopes from fission products of principal interest are ^{90}Sr , ^{137}Cs , and ^{147}Pm . At present, fission product isotopes are obtained from waste streams in the Richland chemical separations plant and are distributed through the Oak Ridge National Laboratory, Isotopes Sales Department. These fission products will also be available in large quantities in the future, when their production is warranted by market demands, from commercial firms operating chemical processing plants, such as Nuclear Fuel Services, Inc., Allied Chemical Corp., General Electric Company, and Atlantic Richfield Company. A computer projection of isotope production by power reactors through 1990 was prepared by Battelle Northwest Laboratories.⁴ It is expected that the expanding uses of these isotopes may eventually create a source of revenue for utilities from the by-products resulting from reprocessing of fuel from reactor operation.

Fission product radioisotopes are used in a number of industrial applications, as well as for space, power, and heat purposes. Promethium-147 and ^{90}Sr are used in radioisotope power generators. Strontium-90 is also employed in industrial thickness gauges and in the treatment of certain eye disorders and in medical blood irradiators. Promethium-147 is also used to activate luminous sources. Cesium-137 is used as a radiation source for both industrial and medical uses.

AEC revenue for the distribution of the above three radioisotopes is given in Table 4 for fiscal years 1965 through 1969.

Table 4. AEC Sales of ^{137}Cs , ^{147}Pm , and ^{90}Sr

	Revenue, dollars				
	1965	1966	1967	1968	1969
^{137}Cs	192,000	27,000	60,000	83,000	42,000
^{147}Pm	28,000	12,000	55,000	56,000	45,000
^{90}Sr	3,000	65,000	59,000	188,000	169,000
Total	\$223,000	\$104,000	\$174,000	\$327,000	\$256,000

In July 1969, the AEC cancelled its 1968 proposal to increase ^{137}Cs prices. Due to a rapidly increasing sales rate for ^{137}Cs and a reduction in production and distribution costs resulting from process improvements, the present prices, which have been in effect since late 1965, will be retained. The Commission will reexamine its ^{137}Cs price schedule again in about two years.

PROCESSORS (4)

A processor is defined as one who can prepare irradiation targets and (subsequent to irradiation in an accelerator or nuclear reactor) can separate and prepare the desired radiochemicals or radiopharmaceuticals for ultimate distribution.

A number of changes in the radioisotope industry have taken place since the last report.

International Chemical & Nuclear Corporation of Burbank, California, acquired the radiochemical operations of Tracerlab, Division of LFE, in late 1968, as well as its film badge service and line of radiation instruments and detectors. Capintec, Inc., of Mt. Vernon, New York, entered the radiochemical processing and distribution business and also introduced a new radioisotope calibrator. The Mallinckrodt Chemical Works of St. Louis, Missouri, and the Daiichi Chemical Drug Company of Japan formed a joint corporation to manufacture and sell radiopharmaceuticals for diagnostic applications.

The principal commercial firms in this area are listed in Table 5.

SOURCE FABRICATION (4)

There are approximately 30 commercial firms producing alpha, beta, gamma, and neutron sources. The current volume of business in radioactive sources is estimated at about \$4 million per year, and is based on information furnished by several of the principal suppliers. The principal commercial firms supplying radioactive sources are listed in Table 6.

TABLE 5

Principal Processors of
Radiochemicals and Radiopharmaceuticals

	<u>Radio- Chemicals</u>	<u>Radio- Pharmaceuticals</u>
Abbott Laboratories, North Chicago, Ill.		X
Atomic Corp. of America, Sun Valley, Calif.	X	
Bio-Nuclear, Div. of Bio-Assay Labs, Houston, Texas	X	X
Bio-Rad Laboratories, Richmond, Calif.	X	
Calbiochem, Los Angeles, Calif.	X	
Cambridge Nuclear Corp., Cambridge, Mass.	X	X
Capintec, Inc., Mt. Vernon, N. Y.	X	
General Electric Co., Pleasanton, Calif.	X	
General Nuclear, Inc., Houston, Tex.	X	
Hastings Radiochemical Works, Houston, Tex.	X	X
International Chemical & Nuclear Corp., Burbank, Calif.	X	
Nuclear Science Div., Pittsburgh, Pa.	X	
Isotopes, Inc., Westwood, N. J., and Baltimore, Md., (Subsidiary of Teledyne)	X	
New England Nuclear Corp., Boston, Mass.	X	X
Amersham-Searle, Chicago, Illinois (G. D. Searle Company)	X	X
Nuclear Consultants Corp., St. Louis, Mo. (Div. of Mallinckrodt Chemical Works)	X	X
Nuclear Equipment Chemical Corp., Farmingdale, N.Y.	X	X
Picker-Hoechst, Bedford, Mass.	X	
Schwartz Bio-Research Inc., Orangeburg, N. Y. (Sub. of Becton, Dickinson & Co.)	X	
E. R. Squibb & Sons, New Brunswick, N. J.		X
Union Carbide Corp., Tuxedo, N. Y.	X	

TABLE 6

Principal Suppliers of Radioactive Sources

American Nuclear Corp., Oak Ridge, Tennessee
 Atomchem Corp., Farmingdale, N. Y.
 Bio-Nuclear, Div. of Bio-Assay Labs, Houston, Texas
 The Budd Co., Phoenixville, Pa.
 Canrad Precision Industries, New York, N. Y.
 Cumberland Research Corp., Port Norris, N. J.
 Gamma Industries, Inc., Baton Rouge, La.
 Gamma Process Company, New York, N. Y.
 General Electric Co., Pleasanton, Calif.
 General Nuclear, Inc., Houston, Texas
 Hastings Radiochemical Works, Houston, Texas
 International Chemical & Nuclear Corp., Burbank, Calif.
 Industrial Reactor Labs., Plainsboro, N. J.
 Isotope Products Lab., Los Angeles, Calif.
 Isotopes, Inc., Palo Alto, California
 Lockheed Georgia Company, Marietta, Ga.
 3M Company, St. Paul, Minn.
 Monsanto Research Corp., Dayton, Ohio
 New England Nuclear, Boston, Mass.
 Amersham-Searle, Chicago, Ill.
 Nuclear Materials & Equipment Corp., Apollo, Pa.
 Picker Nuclear, White Plains, N. Y.
 Radionics Corp., Norristown, Pa.
 Radium Chemical Corp., New York, N. Y.
 Technical Operations, Inc., Burlington, Mass.

Equipment Manufacturers (3)

There are six principal areas of interest for the use of radiation equipment. These are radiographic and teletherapy units, ~~gauges~~, irradiators, isotopic power devices, and self-illuminating devices,

Radiographic Units (4)

The annual volume of sales of radiographic units is about \$2 million and the industry growth rate is about 20% per year. These units, which utilize radioisotopes such as ^{192}Ir , ^{60}Co , ^{137}Cs , and ^{170}Tm , are manufactured entirely by industry. The commercial firms active in this field are listed in Table 7.

TABLE 7

Principal Suppliers of Radiographic Units

The Budd Company, Phoenixville, Pennsylvania
 Gamma Industries, Inc., Baton Rouge, Louisiana
 Nuclear Materials & Equipment Corp., Apollo, Pa.
 Technical Operations, Inc., Burlington, Mass.

six thousand

There are approximately [^] radiographers working under AEC licenses and it is estimated that some 10 to 15 thousand persons are engaged in industrial radiographic operations using radioisotopes. Seasonal variations in this type of work decrease the number of persons by 50 % or more during the different periods of the year.

Teletherapy Units (4)

There are eight commercial suppliers of teletherapy units. Four of the eight suppliers (General Electric, North American Philips Company, Siemens America, and Schick X-Ray Company) are distributors of foreign-made teletherapy units. The firms active in this field are listed in Table 8.

TABLE 8

Principal Suppliers of
Radioisotope Teletherapy Units

W. F. and John Barnes Company, Automated Machine Div.
 of Babcock & Wilcox, Rockford, Illinois
 General Electric Company, Milwaukee, Wisconsin
 Laboratories for Electronics, Waltham, Massachusetts
 (Tracerlab)
 North American Philips Company, Inc., New York, N. Y.
 Picker X-Ray Company, Cleveland, Ohio
 Schick X-Ray Company, Chicago, Illinois
 Siemens America, Inc., New York, New York
 Westinghouse Electric Corporation, Baltimore, Maryland

The IAEA published in 1968 a new "Directory of High Energy Radiotherapy Centres" ⁵ which lists radioisotope and other high energy teletherapy installations throughout the world. Some 1,578 radiotherapy centers in 62 countries are listed, with details on 2,074 treatment units.

The world total of 2,074 units includes 1,676 ⁶⁰Co units and 140 ¹³⁷Cs units. . Since 1959, the annual increase in cobalt units has been nearly constant at about 160 to 170 per year. The average new unit being installed in the U.S. contains 3 to 4 thousand curies of high-specific activity ⁶⁰Co, with some containing up to 8 to 10 thousand curies. There are some 258 nonisotopic teletherapy units reported including betatrons, linear accelerators, Van de Graaff machines, and high-energy resonant transformer x-ray generators.

Radioisotope Gauges (4)

There are currently approximately 30 firms active in the radioisotope gauge field. This type of gauge combines a radioisotope (alpha, beta, gamma, or neutron) source of radiation and appropriate radiation detection elements and control equipment, which permit measurement and control of various processes. These include flow rate, thickness, level, density, and activation analysis systems. The current market for radioisotope gauges is estimated at about \$30 million and is expected to grow at about 20% annually.

The principal suppliers of industrial radioisotope gauges and gauging systems are listed in Table 9.

Table 10 gives shipments of radioisotope gauges, as reported by the Bureau of the Census, and estimates of sales for 1968, 1969, and 1970 made by the Division of Industrial Participation and industry sources.

^{AEC} As in the case of radiation detection and measurement instruments, the shipments of radioisotope gauges reported by the Census Bureau are considerably lower than estimated sales for the years 1968, 1969, and 1970. The difference (allowing for annual growth) would be found if the smaller gauging firms were solicited.

Some examples of radioisotope gauge applications include: measuring and automatically adjusting the thickness of steel sheet, paper, tire cords, textiles, plastics, and other materials; controlling sugar, fat, and meat content in food packaging; measuring soil density and moisture content; measuring snow density for water run-off forecasting; detecting gold; and measuring the density of ocean bottom sediments.

TABLE 9

Principal Suppliers of Industrial
Radioisotope Gauges and Gauging Systems

Ball Corporation, Boulder, Colorado
 Bendix Corporation, Sheffield Division, Dayton, Ohio
 Brun Sensor Systems Corporation, Columbus, Ohio
 Columbia Scientific Instrument Industries, Austin, Texas
 Control Services Company, Pacheco, California
 Densitronics Div., of Aerospace Controls, Los Angeles, Calif.
 Electronic Automation Systems, Grand Island, New York
 General Electric Company, Lynn, Massachusetts
 General Means Instrument, Covina, California
 General Nucleonics Division of Tyco, Pomona, California
 Industrial Nucleonics, Inc., Columbus, Ohio
 Industrionics Div. of Avionic Products Eng. Corp.,
 Succasunna, New Jersey
 IN-VAL-CO, formerly Instruments, Inc.,
 Tulsa, Oklahoma
 Kay-Ray, Inc., Palatine, Illinois
 Measurex Corporation, Santa Clara, California
 Nuclear Research Corporation, Southampton, Pa.
 Nucleonics Development Company, Monrovia, California
 Ohmart Corporation, Cincinnati, Ohio
 Panametrics Division of Esterline-Angus, Waltham, Mass.
 Robertshaw Controls Company, Anaheim, California
 Seaman Nuclear Corporation, Milwaukee, Wisconsin
 Texas-Nuclear Division of Nuclear-Chicago, Austin, Texas
 Tracerlab, Division of LFE, Inc., Waltham, Massachusetts
 Troxler Electronic Laboratories, Inc., Raleigh, N. C.
 Twin Cities Testing Corporation, North Tonawanda, N. Y.
 Unit Process Assemblies, Woodside, New York

TABLE 10

Shipments and Sales of Radioisotope Gauges

<u>Census Bureau Reported Shipments</u>		<u>Estimated Sales</u>	
<u>Year</u>	<u>\$ in Millions</u>	<u>Year</u>	<u>\$ in Millions</u>
1957	\$ 6.0	1968	\$30
1958	3.9	1969	36
1959	8.9	1970	43
1960	10.3		
1961	11.0		
1962	9.6		
1963	9.9		
1964	9.7		
1965	13.1		
1966	15.2		
1967	14.1		

Self-Illuminating Materials and Devices (4)

Beta-emitting radioisotopes, such as ^{85}Kr , tritium, and ^{147}Pm , are used in the preparation of self-illuminating materials and devices. Tritium is the most frequently used radioisotope. The growing use and interest in self-illuminating materials is reflected in the sales of tritium by the AEC given below.

<u>Fiscal Year</u>	<u>Kilocuries</u>	<u>AEC Sales</u>
1963	60	\$ 95,000
1964	112	125,000
1965	118	128,000
1966	171	196,000
1967	179	200,000
1968	179	199,000
1969	308	259,590

The firms active in this field are listed in Table 11.

TABLE 11

Principal Commercial Suppliers of
Self-Illuminating Materials and Devices

American Atomics, Tucson, Arizona
 Canrad Precision Industries, Inc., New York, New York
 Luminous Products Corporation, Boston, Mass.
 (Subsidiary of New England Nuclear Corp.)
 3M Company, St. Paul, Minnesota
 Radium Chemical Company, New York, New York
 U.S. Radium Corporation, Morristown, New Jersey

Radioisotopic Power (3)

It is possible to utilize the heat released, when isotopes decay, for various specialized applications requiring power. Growth in this area has been much slower than anticipated a few years ago, primarily because space and military requirements have not developed or have had missions and funding reduced. Reliable forecasts of isotopes requirements have been

difficult to make, due to the uncertainty of users as to availability and costs of isotopes, and the uncertainty of the producer about requirements.

An AEC report⁶; published in May 1968, contains much useful information on the development of large-scale uses of radioisotopes for electrical power, thermal energy, and process radiation applications.

It is expected that the expanding nuclear power economy should contribute significantly to the production of isotopes, such as ^{238}Pu , ^{244}Cm , and ^{90}Sr , as a by-product of reactor operation and chemical reprocessing operations. As mentioned previously, some isotopes may be produced in power reactors through the irradiation of target materials, i.e., ^{60}Co and ^{170}Tm . To encourage the demand for irradiation services to produce such isotopes, the Commission has proposed legislation that would amend the Atomic Energy Act of 1954 to clarify the authority of the AEC to enter into long-term arrangements to provide for toll irradiation in AEC facilities of materials for foreign and domestic customers.

Radioisotopic power applications fall into the following general categories:

Space electric power applications include weather, communications, and navigation satellites; space exploratory power systems, such as that for the Apollo Lunar Surface Experiment; and space mission requirements. left on the moon by Apollo-12;

Terrestrial power applications include remote weather stations, navigation buoys and beacons, for power on off-shore oil rigs, and undersea power systems.

Thermal applications include spacecraft and swim-suit heaters; waste water reprocessing systems for manned space missions; and energy sources for cardiac pacemakers, heart pumps, and small undersea propulsion systems. For example, two 15-thermal-watt heaters were placed on the moon during the first manned lunar landing, to warm the Seismic Experiment Package during the lunar night. (Apollo-11)

The General Electric Company was selected to design and develop a ^{238}Pu powered waste disposal and water recovery system for a prolonged manned space mission. This development work is expected to take approximately three years.

Isotopic propulsion systems are primarily for auxiliary spacecraft propulsion.

MARKETS FOR RADIOISOTOPE POWER (4)

thousand/

The Federal Government continues to be the principal market for these devices. While forecasts of isotope requirements are given in WASH-1095 (Ref. 6), the dollar value of power-producing devices cannot really be estimated yet. Substantial commercial markets for applications, such as direct broadcast satellites, marine and remote land power systems, could develop eventually. The cardiac pacemaker and the artificial heart appear to be among the most promising future markets. It is estimated that there could be a need for about 10 cardiac pacemakers per year in the future. These units produce 150-200 electrical microwatts of power and do not require large quantities of ^{238}Pu . The need for artificial heart implantation operations has been estimated as high as 100 thousand patients or more per year and could use all foreseen supplies of ^{238}Pu , if this device is eventually successfully developed.

The following companies are active in the isotopic power generation field:

Hittman Associates, Columbia, Md.
 Aerojet-General Corporation, San Ramon, California
 Atomics International Div., North American Rockwell Corp.,
 Canoga Park, California
 Donald W. Douglas Laboratories, Richland, Washington
 General Electric Company, Missiles & Space Division,
 Valley Forge, Pennsylvania
 Gulf General Atomic, Incorporated, San Diego, California
 3M Company, St. Paul, Minnesota
 Nuclear Materials & Equipment Corp., (NUMEC), Apollo, Pa.
 Sanders Nuclear Corporation, Nashua, New Hampshire
 Teledyne, Isotopes Division, Westwood, N. J., and
 Baltimore, Maryland
 Westinghouse Electric Corp., Pittsburgh, Pennsylvania

It is expected that other firms will enter this field as the market develops.

NUMEC has delivered a strontium-fueled 15-watt radioisotope-thermoelectric-generator to the U.S. Navy to be operated at an ocean depth of 13,500 feet. Also, a NUMEC developed ^{238}Pu powered pacemaker was successfully implanted in a dog in June 1969. → Seven additional NUMEC units are planned for implantation by the end of 1969.

(URIPS)

The Navy ordered four 1-watt Aerojet-General Undersea Radioisotope Power Supplies for installation in the Atlantic Underwater Test Evaluation Center and to power oceanographic data-collection systems in the Pacific.

General Electric was selected for development work on a multi-hundred watt thermoelectric power system that is ultimately expected to produce a modular, lightweight, versatile system applicable to a number of potential space missions.

Radiation Processing (3)

RADIATION PROCESSING APPLICATIONS AND INDUSTRIAL INTEREST (4)

Dr. Seaborg, Chairman, U.S. Atomic Energy Commission, in his remarks at the dedication of Radiation Machinery Corporation's new headquarters and facility at Hanover, New Jersey, in July 1969, reviewed the current status of radiation processing. Excerpts from Dr. Seaborg's remarks follow:

The commercial use of ionizing radiation to improve the properties of existing products and catalyze the synthesis of new materials has been studied for nearly 20 years, but only in recent years has this potential become a reality. The interaction of gamma rays and high-energy electrons with matter can catalyze chemical reactions, change the molecular or lattice structure of solids, and destroy micro-organisms. Each of these types of radiation effects has been used by industrial firms to manufacture or process a number of products, and many companies are conducting experimental programs to apply the beneficial effects of radiation to other production operations.

Commercial radiation processing has grown rapidly from virtually nothing a decade ago to an annual volume of irradiated products estimated at more than \$200 million. Radiation is used by industry to improve the properties of transistors and diodes, to cross-link polyethylene and other polymers for electrical insulation and packaging film, to initiate the synthesis of

ethyl bromide, and to sterilize surgical sutures and other medical supplies. These radiation applications result in products now sold commercially, and there is an even broader spectrum of AEC and industrial research and development on other irradiated products that may soon appear on the market.

Most commercial radiation processes currently use electron beam irradiation. This type of radiation, produced by machines of various kinds, is ideal when sheet or film is to be treated or when only surface treatment is required, since electrons have relatively poor penetrating power. In recent years, however, a number of radiation products and processes have been commercialized or have entered the pilot plant stage for which isotopic radiation sources have proved to have technical or economic advantage.

The most promising radiation processing applications in use or under development are listed below:

- ¶ Curing of surface coatings by machine irradiation of special lacquers and paints. The Moleculon Research Corporation; Boise Cascade Corporation, using the Electro cure process under license to the Ford Motor Company; Radiation Dynamics, Inc., using the Dynacote Process; and PPG Industries are the principal firms carrying out development work in this area.
- ¶ Irradiation of wood plastic combinations to produce superior qualities in strength, appearance, and surface properties has been underway for some time by American Novawood Corporation and Lockheed-Georgia Company. Atlantic Richfield Company, Gamma Process Company, and Radiation Machinery Corporation have recently entered this field. All of the above firms use ^{60}Co .

As an illustration of the current market for these products, the American Novawood Corporation was awarded a \$1.2 million contract to supply Gammapar parquet flooring and Novawood guard rails and panels for the new Kansas City International Airport terminal buildings, and have also a contract with the U.S. Army Mobility Equipment Research & Development Center, Fort Belvoir, Virginia, for the development and prototype fabrication of a new materials handling system made from plastic-cellulose composites.

④ Crosslinking of polyethylene films to produce special wire coatings, insulating materials, heat shrinkable packaging film, and bags of improved physical properties. The major companies in this area are Raychem, W. R. Grace (Cryovac Division), Moleculon Research Corporation, Midwest Irradiation Center, Inc., High Voltage Engineering Corporation, and International Telephone & Telegraph.

④ Preparation of specialty copolymers for use as battery separators for Department of Defense requirements.

④ Synthesis of new graft copolymer fibers with improved "crease resistance" and "soil resistance" properties by Deering-Milliken Corporation.

④ Synthesis of ethyl bromide by Dow Chemical Company.

④ The controlled degradation of Polyox, a polyethylene oxide polymer, by Union Carbide Corporation.

Two new applications of radiation processing have been demonstrated. Concrete polymer materials, prepared similarly to wood plastics, have greatly increased strength and resistance to freezing and corrosion. Radiation-induced emulsion polymerization has been used to prepare a vinyl acetate paint latex which has been evaluated by potential users and found to have desirable properties in latex paint formulations.

FIRMS ASSOCIATED WITH RADIATION PROCESSING (4)

In January 1968, the Business and Defense Services Administration of the Department of Commerce released a new "Guide to Services, Suppliers, and Research on Food Irradiation."⁷ The guide lists commercial firms in all areas of radiation processing, including design, engineering, and construction of facilities; design and fabrication of irradiation equipment; radiation source suppliers; encapsulators; and electron machine manufacturers. Because of the additional companies that have been identified in the above categories, lists of firms associated with radiation processing in all categories are shown in the following tabulations.

(A)

Principal Firms Supplying Electron Accelerator Equipment, and/or Services (S)
or Finished Products (P).

Accelerators, Inc.
Austin, Texas (A)

Applied Radiation Corporation
Walnut Creek, California (A,S)

Columbia Research Corporation
Gaithersburg, Maryland (S)

Electra Process Industries, Inc.
Spokane, Washington (S)

General Electric Company
Milwaukee, Wisconsin (A)

High Voltage Engineering Corporation
Burlington, Massachusetts (A)

Midwest Irradiation Center, Inc.
Rockford, Illinois (S)

Moleculon Research Corporation
Cambridge, Massachusetts (S)

Radiation Dynamics, Inc.
Westbury, New York (A,S)

Radiation Polymer Corporation
Plainfield, Illinois
(Sub. of PPG Industries) (S)

Raychem
Menlo Park, California (P)

Raycon, Inc.
South Windsor, Connecticut (S)

Texas Nuclear Corporation
Austin, Texas
(Sub. of G. D. Searle & Company) (A)

Varian Associates
Palo Alto, California (A,S)

Westinghouse Electric Corporation
Baltimore, Maryland (A)

Commercial Irradiation Processing Facilities

The American Novawood Corp., Lynchburg, Va.
 Electra-Process Industries, Inc., Spokane, Wash.
 Ethicon, Somerville, N. J., (Div. of Johnson & Johnson)
 Gamma Process Co., New York, New York
 Lockheed-Georgia Co., Dawsonville, Ga.
 Midwest Irradiation Center, Inc., Rockford, Ill.
 Moleculon Research Corp., Cambridge, Mass.
 Neutron Products, Inc., Dickerson, Maryland
 Nuclear Materials & Equipment Corp., Apollo, Pa.
 Radiation Dynamics, Inc., Westbury, L. I., N. Y.
 Radiation Industries, Inc., Lodi, New Jersey
 Radiation Machinery Corp., Parsippany, New Jersey
 Radiation Technology, Inc., New Brunswick, N. J.
 Raycon, Inc., South Windsor, Conn.
 Raychem, Redwood City, California

Architectural Design, Engineering and Construction

The American Novawood Corp., Lynchburg, Va.
 American Nuclear Corp., Oak Ridge, Tennessee
 Atomchem Corp., Plainview, New York
 Biggs and Hill, Inc., New York, New York
 Burns & Roe, Inc., Oradell, N. J.
 Commonwealth Associates, Inc., Jackson, Mich.
 A. Epstein and Sons, Inc., Chicago, Ill.
 Food Industries Research & Engineering, Yakima, Wash.
 Gamma Process Company, New York, New York
 General Electric Co., Pleasanton, California
 Holmes & Narver, Inc., Los Angeles, California
 Isotopes, Inc., Westwood, New Jersey
 Lockheed-Georgia Co., Dawsonville, Georgia
 Chas. T. Main, Inc., Boston, Massachusetts
 Nuclear Materials & Equipment Corp., Apollo, Pa.
 Nuclear Technology Corp., White Plains, N. Y.
 Neutron Products, Inc., Dickerson, Maryland
 Parson-Jurden Corp., New York, New York
 (Subsidiary of Ralph M. Parsons Co.)
 Radiation Dynamics, Inc., Westbury, L. I., N. Y.
 Radiation Industries, Inc., Lodi, New Jersey
 Radiation Machinery Corp., Parsippany, New Jersey
 Stearns-Roger Corp., Denver, Colorado
 Stone & Webster Engineering Corp., Garden City, L. I., N. Y.

Irradiation Equipment, Design and Fabrication

The American Novawood Corporation, Lynchburg, Va.
 American Nuclear Corporation, Oak Ridge, Tennessee
 Applied Radiation Corporation, Walnut Creek, California
 Atomchem Corporation, Plainview, New York
 Gamma Process Company, New York, New York
 Isotopes, Inc., Westwood, New Jersey
 Lockheed-Georgia Company, Dawsonville, Georgia
 National Lead Company, Wilmington, Delaware
 Neutron Products, Inc., Dickerson, Maryland
 Nuclear Materials & Equipment Corporation, Apollo, Pa.
 Nuclear Technology Corporation, White Plains, New York
 Radiation Industries, Inc., Lodi, New Jersey
 Stearns-Roger Corporation, Denver, Colorado

Radiation Processing Source Suppliers and Encapsulators

American Nuclear Corporation, Oak Ridge, Tennessee
 Atomchem Corporation, Plainview, New York
 Gamma Industries, Baton Rouge, Louisiana
 Gamma Process Company, New York, New York
 General Electric Company, Pleasanton, California
 Industrial Reactor Labs., Plainsboro, New Jersey
 Lockheed-Georgia Company, Dawsonville, Georgia
 Neutron Products, Inc., Dickerson, Maryland
 Nuclear Materials & Equipment Corporation, Apollo, Pa.
 Radiation Machinery Corporation, Parsippany, New Jersey

IRRADIATION PROCESSING OF FOOD (4)

The possibility of radiation preservation of foods was recognized in general terms in the early 1900's. Research in the mid-1940's revealed the potential of the process, as well as determining some of the problems. In the early 1950's, the AEC was looking for ways to use radioactive products and began a limited investigation of possibilities in food protection. In August 1953, the Army also began modest research (since greatly expanded) in food preservation by irradiation.

By 1959, the Army had decided to concentrate its attention on several meat items (ham, pork, chicken, and beef), with the objective of developing them for combat rations through high-dose radiation (sterilization). Responsibility for low-dose (pasteurization) research was assumed by the AEC, in the belief that this process would be of more direct benefit to civilian consumers. The goal in each program was to bring the

techniques to the point of technical and economic feasibility so that private industry could apply them commercially. Under this program, AEC was to develop the technical aspects of low-dose irradiation using the wholesomeness data developed through the Army's sterilization program.

The Food and Drug Administration (FDA), which has the responsibility for deciding whether a particular radiation-processed food is fit for human consumption, requires that extensive wholesomeness data accompany each application for clearance of a food.

Pasteurization requires only about one-tenth as much radiation dosage as sterilization. Foods that have been proven safe for consumption after radiation sterilization are generally assumed to be safe if lesser pasteurizing doses are administered. In recent years, advances in science have put an additional burden on a petitioner, such as AEC, and many previous agreements regarding irradiated food petitions were negated and either the petitions were withdrawn or new phases of research had to be undertaken.

Between 1963 and early 1966, the FDA had issued approvals to the AEC, the Army, and private firms for radiation preservation of bacon, wheat and wheat products, and white potatoes. In 1968, the FDA withdrew its approval of bacon and decided not to grant the Army's petition for approval of radiation sterilized canned smoked ham for public consumption, pending further animal feeding tests. They felt that additional information was necessary to assure there were no long-term bad effects on the food products. At present, wheat and wheat products and white potatoes are the only foods approved by FDA for irradiation processing.

Early in the year, the AEC's program for the radiation preservation of foods was reoriented, following the FDA decision not to approve ham and due to budget restrictions. AEC decided that its future petitions to FDA should be supported from results of two year animal feeding studies using foods irradiated at doses required for a specific food. The re-orientation required temporarily dropping any new food irradiation work in the AEC's Divisions of Biology and Medicine and Isotopes Development. The AEC is continuing work on key contracts that are gathering data required to support and process petitions to the FDA on strawberries, papayas, and east coast fin fish.

The principal irradiators which have supported the U.S. food irradiation effort are listed in Table 12.

TABLE 12

Irradiators Supporting the Food-Irradiation Effort

<u>Irradiator</u>	<u>Description</u>	<u>Location</u>	<u>Purpose</u>
<u>Research</u>			
Research	30,000-40,000 curies of ⁶⁰ Co. pool type	Massachusetts Institute of Technology University of California (Davis) University of Washington University of Hawaii	On-site research
Research	Refueled with 100,000 curies of Cs-137	University of Florida	On-site research
On-Ship No. 3	36,000 curies of ⁶⁰ Co. 18-ton portable unit	University of Nebraska	On-site research
Portable Cesium No. 1	170,000 curies of ¹³⁷ Cs, 18-ton portable unit	Was operated for AEC by Isotopes, Inc., Westwood, N. J., presently undergoing modification at BNL.	Demonstration to and use by industry
<u>Pilot Plant</u>			
Marine Products Development Irradiator (MPDI)	250,000 curies of ⁶⁰ Co. 1,000 lb/hr. at pasteurizing dose	Gloucester, Massachusetts	Seafood irradiation
Mobile Gamma Irradiator (MGI)	100,000 curies of ⁶⁰ Co. 1,000 lb/hr. at pasteurizing dose	Davis, California	Fruit irradiation
Grain Products Irradiator (GPI)	25,000 curies of ⁶⁰ Co. 5,000 lb/hr. bulk or 2,800 lb/hr. packaged	Savannah, Georgia	Grain products disinfestation
Hawaii Development Irradiator (HDI)	225,000 curies of ⁶⁰ Co. 4,000 lb/hr. at pasteurizing dose	Honolulu, Hawaii	Tropical fruits

The Mobile Gamma Irradiator, Hawaii Development Irradiator, and the Marine Products Development Irradiator are those facilities supporting the "key" contracts presently being continued for the collection of data demonstrating on a semi-commercial basis the technical effect of the radiation process for strawberries, papayas, and east coast fin fish.

The Defense Department contracted for irradiation processing of 130,000 pounds of wheat flour in the Neutron Products, Inc. newly installed dry package irradiator, the only procurement of irradiated food this year.

Significant Rule Changes in Radioisotope Area (3)

The Division of Materials Licensing booklet, "How to Get a License to Use Radioisotopes,"⁸ explains, in non-technical terms, the objectives of the regulatory program, how it functions, and the safety responsibilities of persons who use radioisotopes.

Since the last report, the AEC has taken a number of significant steps in its regulatory program and has amended, or proposed amendments of, several of its regulations concerning radioactive materials. The most important items are summarized below.

In April 1969, the AEC amended Parts 30, 31, and 32 of its regulations to exempt from licensing requirements the use of small quantities of tritium, ^{60}Co , ^{63}Ni , ^{85}Kr , ^{137}Cs , and ^{147}Pm in electron tubes.

The AEC also amended its regulations in April to provide a new class exemption from licensing for radioisotopes in smoke detectors and other gas and aerosol detectors designed to protect life or property from fire and airborne hazards.

In June, the Commission established a new class exemption from licensing for the use of products containing tritium, ^{85}Kr , and ^{147}Pm in self-luminous items, and incorporated safety standards for their manufacture in its regulations. Excluded from the class exemption are toys, adornments, and frivolous uses.

In July the AEC proposed amendments to Part 40 of its regulations in response to a petition from the National Lead Company and in September, the approved amendments were published in the Federal Register. The changes modify the labeling requirements for uranium counterweights used in aircraft and clarify the requirements for repairs on them.

AEC Advisory Committee on Isotopes and Radiation Development (3)

The present AEC Advisory Committee on Isotopes and Radiation Development was established in 1958. This committee grew out of the Advisory

Committee on Isotope Production and Distribution, which was formed in 1946 and terminated in 1958 when the Commission's regulatory and licensing activities were separated from the operating activities.

The Advisory Committee on Isotopes and Radiation Development reviews AEC policies and the potential development of new applications of isotopes and radiation and makes appropriate recommendations for the Commission's consideration.

The composition of this committee has been gradually changed to bring in representatives from various segments of the isotope industry. The present membership is given below:

Chairman - John W. Landis, Gulf General Atomic, Inc.
AEC Member - Dr. Nathaniel F. Barr, Division of Biology and Medicine
Mr. Robert E. Kettner, Nuclear Assurance Corporation
Dr. Milton Burton, University of Notre Dame
Dr. Bernard Fries, Chevron Research Company
Dr. David E. Harmer, Dow Chemical Company
Dr. Merrill Eisenbud, Environmental Protection Administration
of New York
Dr. Ira Lon Morgan, University of Texas
Mr. Lyle E. Packard, Packard Instrument Company, Inc.
Dr. A. J. Restaino, Atlas Chemical Industries, Inc.
Dr. Seymour Rothchild, New England Nuclear Corporation

(RHL)

Supplementary Keywords: production, radioisotope; production, reactor; fission product, intentional; reactor by-product recovery; production, accelerator; source preparation; source, radioisotope; industry; industry, nuclear energy; industry, radioisotope; radiography; measurement and control system; luminescence; therapy, external; energy source; regulation; process radiation

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1967 (\$0.30).

THE UNIVERSITY OF MISSOURI RESEARCH REACTOR FACILITY (2)

By A. H. Emmons^{*}, E. L. Cox^{**}, and D. M. Alger^{***}

Supplementary Keywords: reactor; facility description; irradiator design; hot laboratory; cobalt-60.

Abstract: ^{The} Research Reactor Facility at the University of Missouri in Columbia is described. This flux trap reactor is designed to operate at 10 MW power but is currently operating at 5 MW. The reactor is cooled and moderated by light water and uses beryllium and graphite as reflectors. The laboratories and other facilities related to the reactor operation are also described.

(Introduction)

In the latter part of the 1950's, the University of Missouri recognized the potential of a nuclear reactor in university research activities. In the early stages of planning, the University decided it should provide a reactor of relatively high power and of unique design. This planning resulted in the University of Missouri Research Reactor Facility^(RRF), located in the Research Park south of the main campus of the University of Missouri in Columbia. The function of this facility^{is} to provide extensive nuclear science research facilities for education and for fundamental and applied research in the life, physical, and engineering sciences.

This nuclear research center is equipped to provide complete radiation science research service to the University. Facilities available include: a 10-MW nuclear reactor, 17 scientific

*Director of the Research Reactor Facility located in Columbia, Missouri.

**Associate Director of the Research Reactor Facility.

***Reactor Supervisor of the Research Reactor Facility.

laboratories, a cobalt irradiation facility, a hot cell facility, a neutron radiography facility, two counting rooms, two animal rooms, photographic darkroom, machine shop, electronics shop, and x-ray room.

Research^{is} initiated by experimenters from the University's campuses at Columbia, Kansas City, Rolla and St. Louis, representing many diverse scientific disciplines. In addition to its University-wide research support programs, the RRF assists other educational, industrial, and governmental programs.

Construction of the facility began in July, 1963, and^{was} completed in the spring of 1966. The reactor was designed for 10-MW operation; however, initial installation of cooling equipment permitted only 5-MW operations. The reactor first attained criticality in October, 1966. A power level of 5-MW was reached in June, 1967, and the reactor has been operating at this power since then. The current operating schedule is 24 hr/day, 5 days per week. Additional cooling equipment is now being installed, and reactor power will be increased to 10 MW in December, 1970.

The RRF^{overall} is administered as a part of the University system and is not associated with any single campus or department. Operation is controlled by the Facility Director, who is responsible to the Vice-President for Academic Affairs. The Director is aided in his duties by his senior staff consisting of the Associate Director, Assistant to the Director, Reactor Supervisor, and Manager of the Nuclear Science Group. The facility is staffed by thirty-six^{persons; the staff includes} professional chemists, nuclear engineers, reactor operators, and shop people, many of whom are also working on advanced degrees at the University. A number of the senior staff also hold academic positions in the University.

The Director is also assisted by a Reactor Advisory Committee consisting of professors and officials of the University as appointed by the President of the University. The Committee's responsibility^{is} to provide periodic safety review of reactor and experimental operations. The Committee membership represents expertise in many disciplines of science and engineering.

The primary purpose of the RRF is to provide outstanding, well-managed, safe radiation research services to faculty and students of the University.

For further details on the facility and for information regarding a possible use, please contact the Director, Research Reactor Facility, Columbia, Missouri, 65201.

Research Reactor Facility (3)

The RRF building is 143 ft wide by 230 ft long (Fig. 1). The grade level floor plan is shown in Fig. 2.

The reactor containment^{structure,} located near the center of the facility, is four stories high and is surrounded at grade level by laboratories. The reactor building has five operational levels, one of which is below ground. The containment walls are poured concrete, and containment integrity is maintained at all times the reactor is operating. Entry and exit is permitted through an interlocked double door passage at grade level. In addition to the reactor and associated shielding,

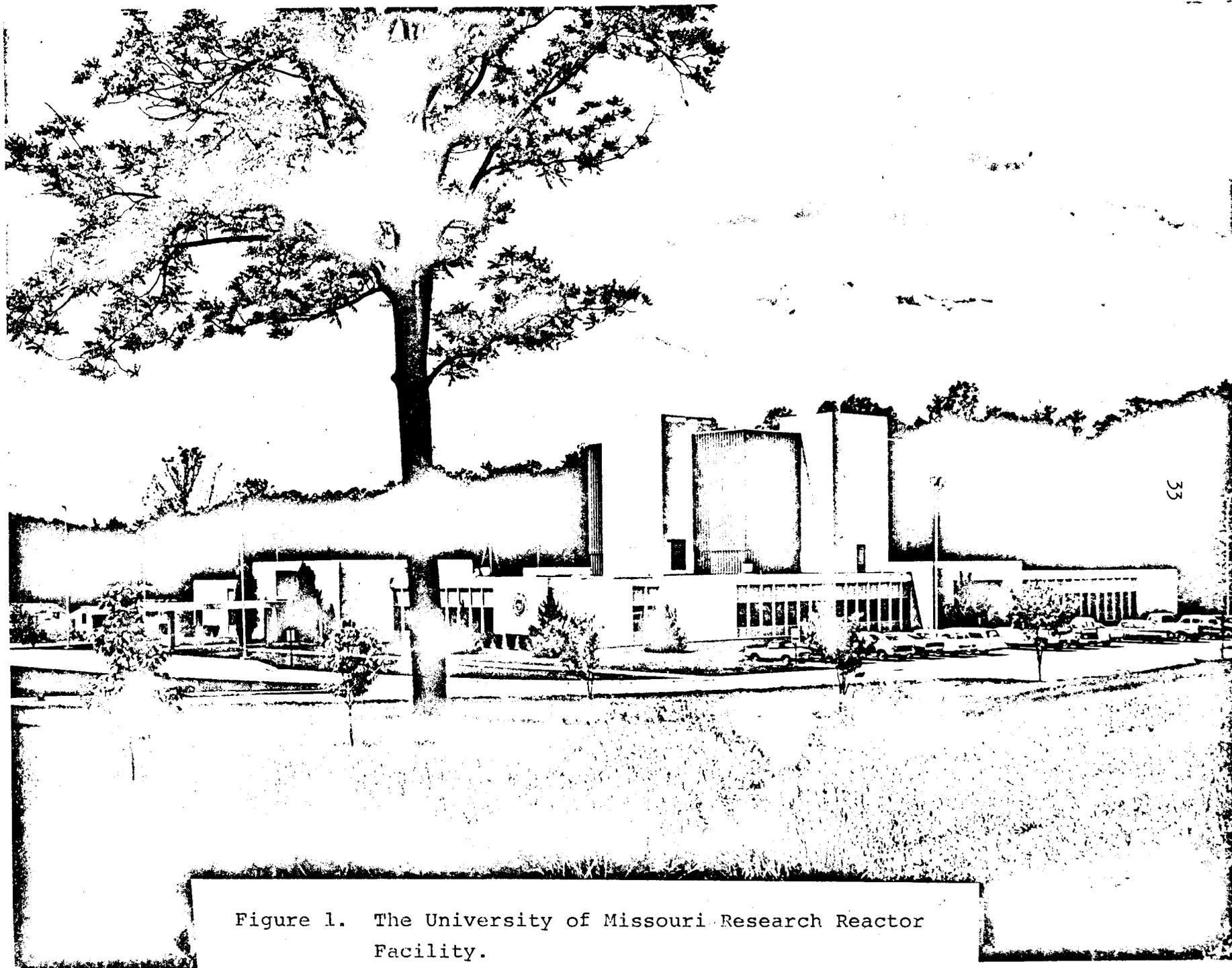


Figure 1. The University of Missouri Research Reactor Facility.

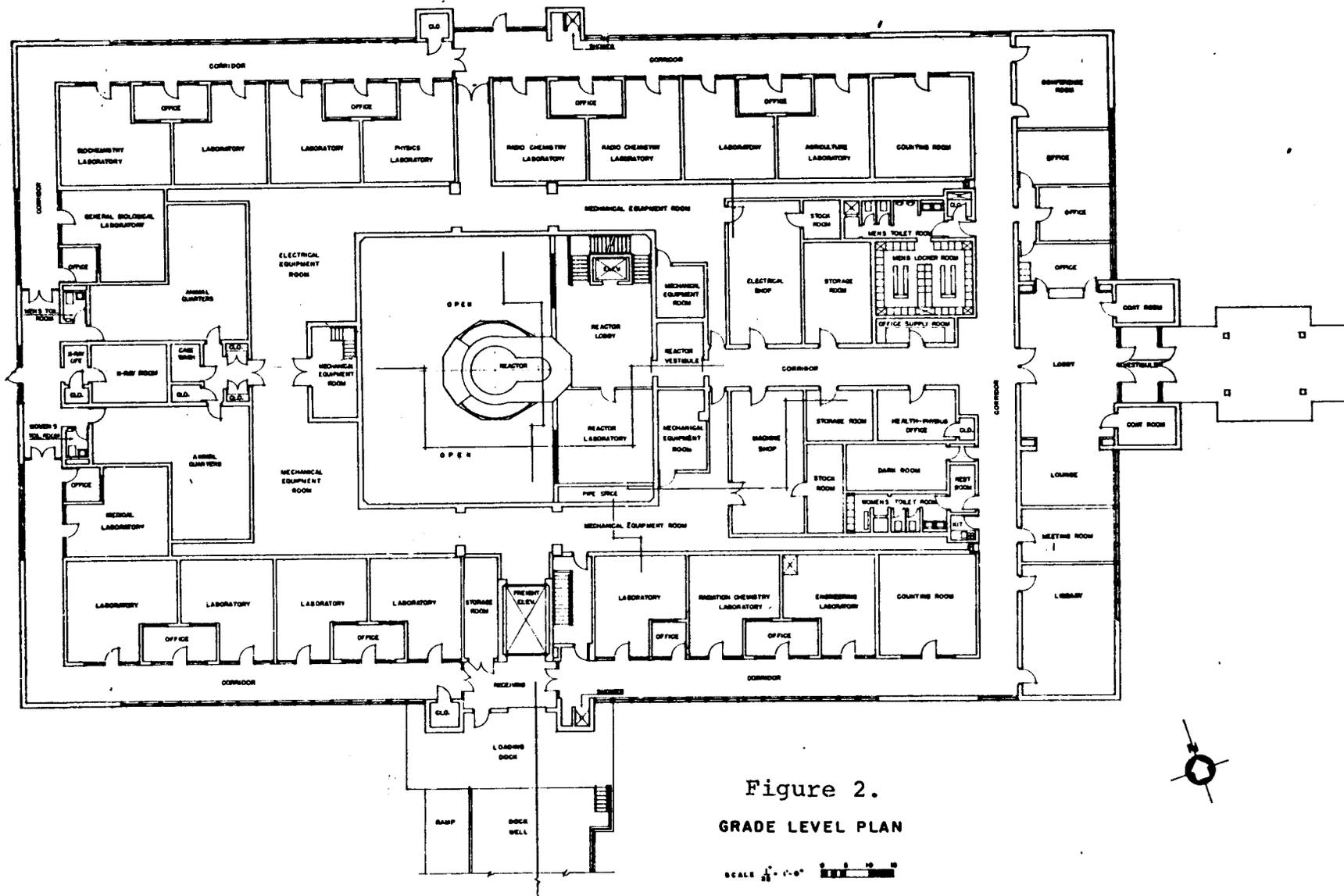


Figure 2.
GRADE LEVEL PLAN

SCALE 1/4" = 1'-0"

the containment building houses the reactor control room, offices, a laboratory for reactor operations, and reactor users' experimental equipment.

The reactor control room on the third floor allows the reactor operator a direct view of the reactor pool area (Fig. 3). The reactor operator may observe experimental activities on the beam port floor by direct observation (Fig. 4) or he may observe the activity by a remote TV camera system with the monitor in the reactor control room. An observer on the beam port floor has ^{the} a view of the reactor shown in Fig 5.

Spent-fuel storage is provided in the reactor pool. The fuel storage racks are placed near a removable portion of the biological shield in such a manner that the spent fuel may be used as the source for a gamma irradiation facility. Recessed storage ports are provided in the west concrete wall on the beam port floor for radioactive materials that are removed from the reactor. These materials are allowed to decay to a level permitting disposal or reuse.

Laboratories (3)

The laboratory building surrounds the reactor containment building and consists of seventeen research laboratories and adjacent offices. The laboratories are arranged on the periphery of the reactor building and enclose a central mechanical equipment area. Each chemistry laboratory is provided with a fume hood which discharges through particulate filters to a central exhaust system and then to the building exhaust stack.

A pneumatic-tube system provides rapid transport of samples into the reactor and returns them to certain laboratories. Four pneumatic-tube

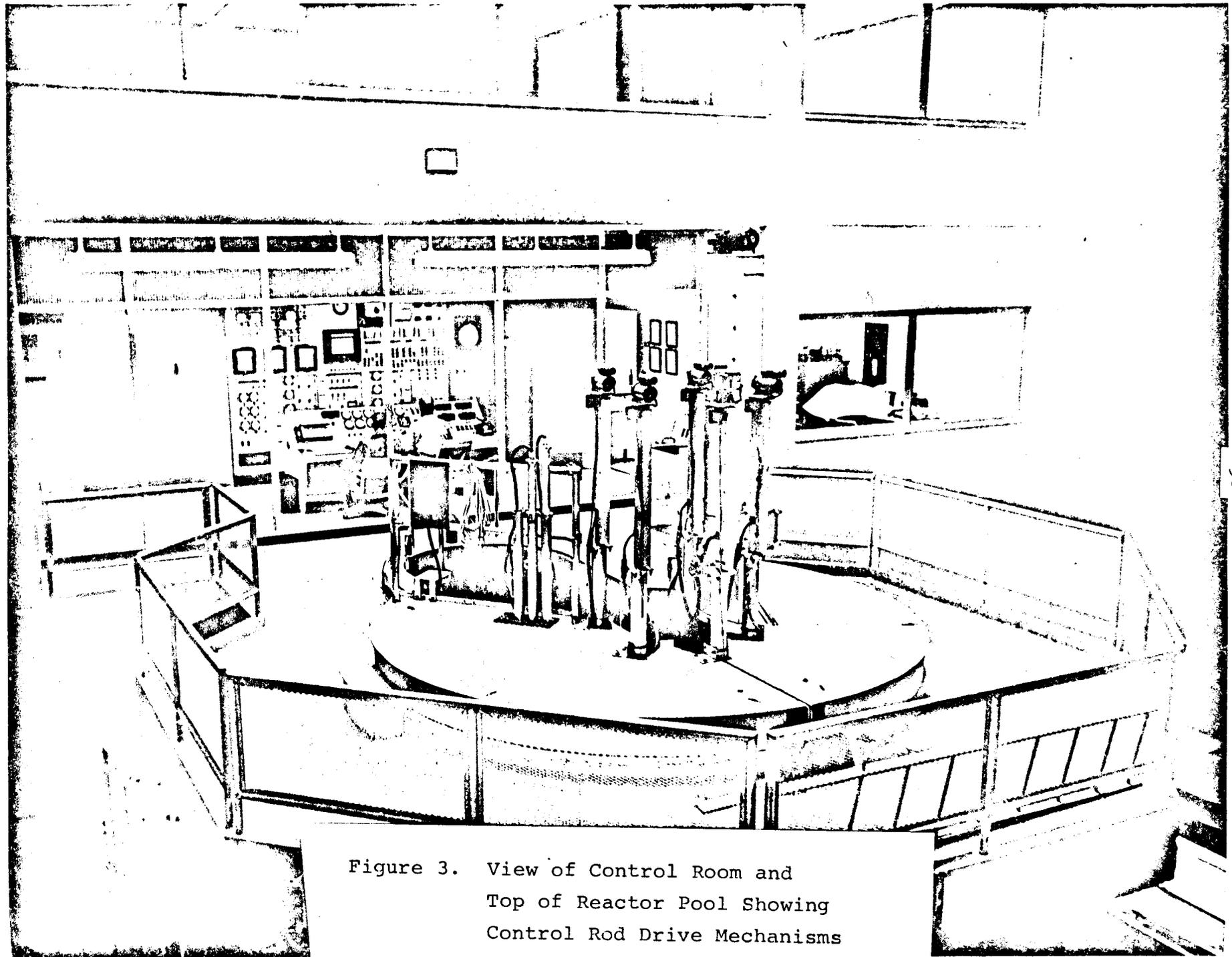


Figure 3. View of Control Room and
Top of Reactor Pool Showing
Control Rod Drive Mechanisms

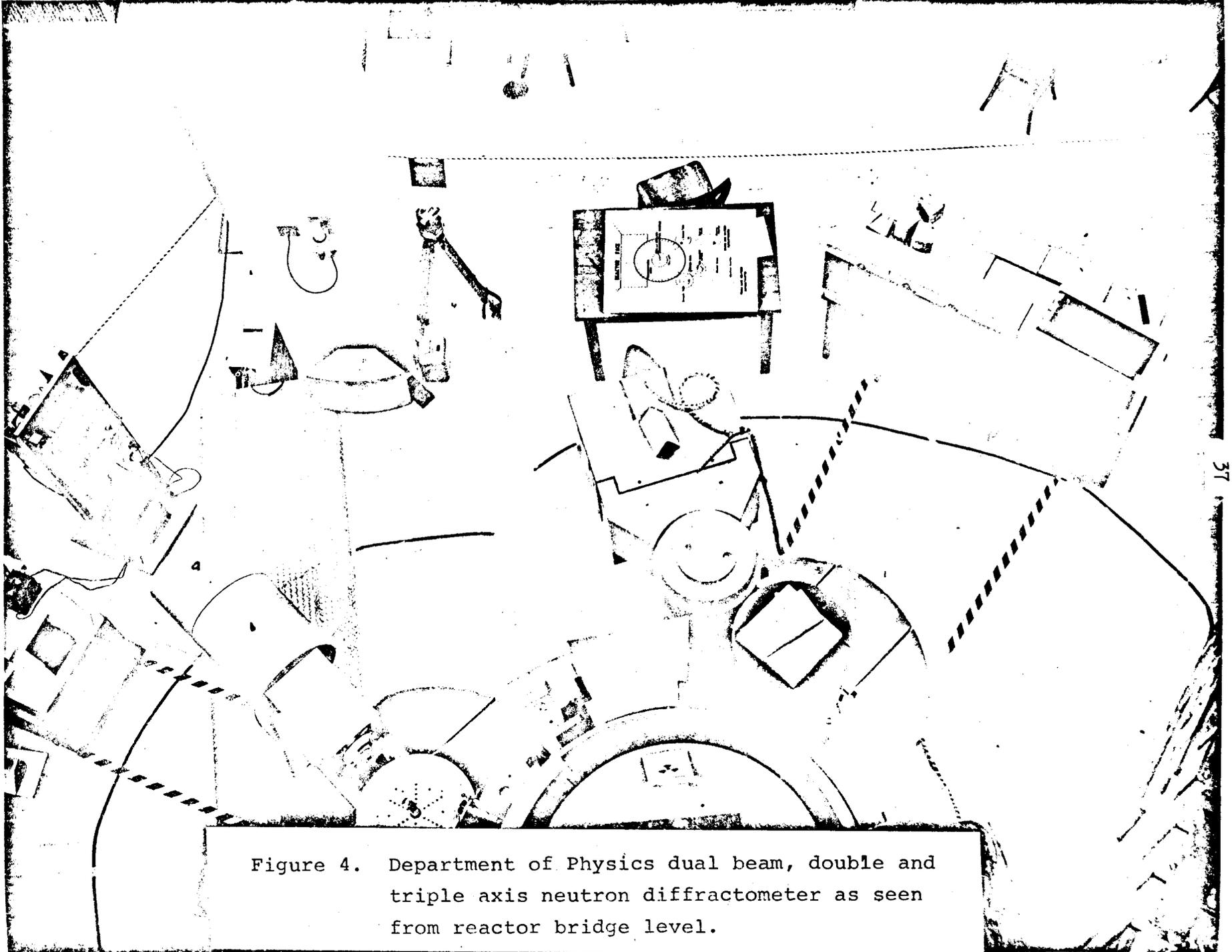


Figure 4. Department of Physics dual beam, double and triple axis neutron diffractometer as seen from reactor bridge level.

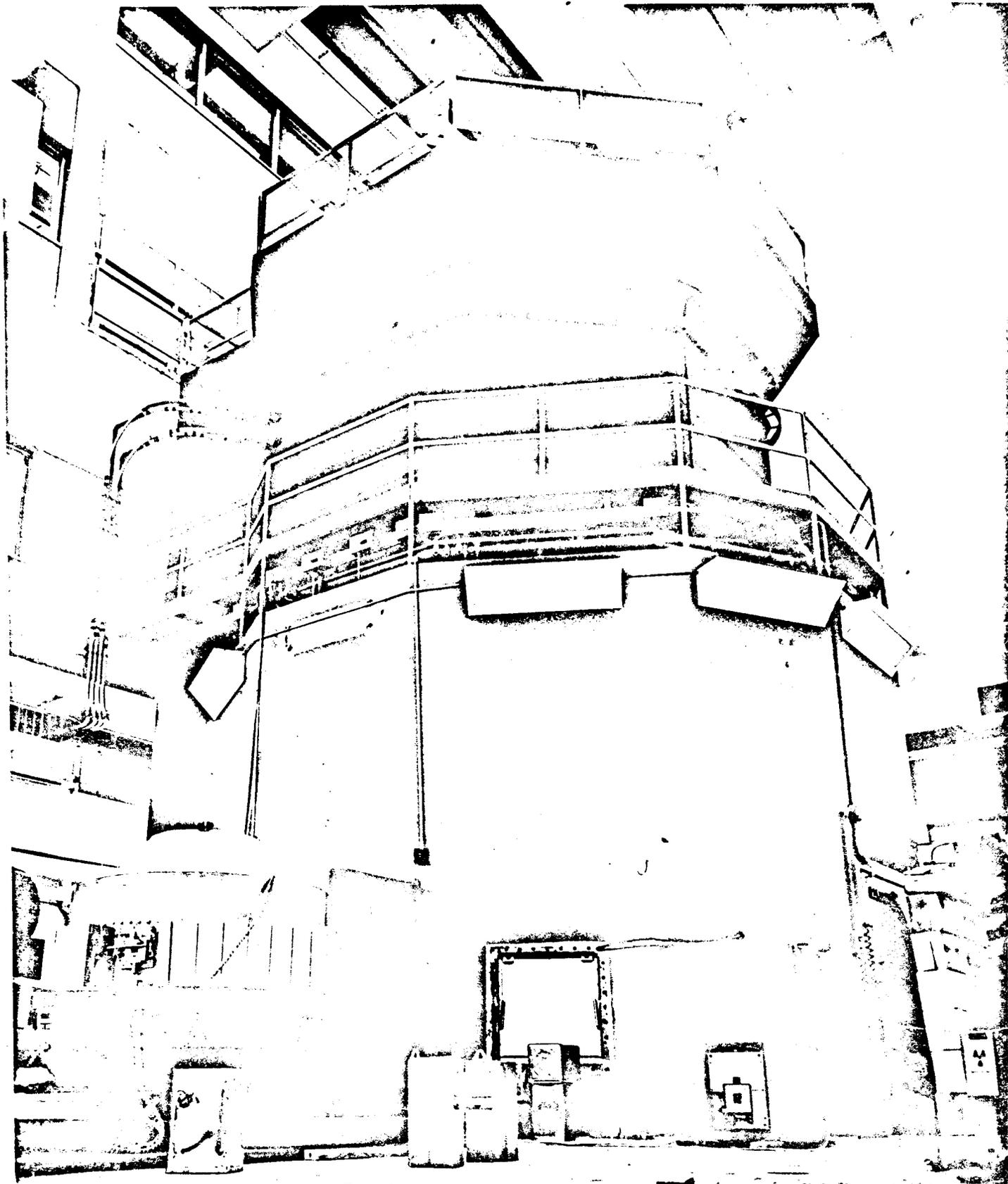


Figure 5. Reactor biological shielding as viewed from beam port floor level.

positions in the reactor service seven laboratories by means of switching stations. One laboratory send-receive station is shown in Fig. 6.

Two radiation detection and measurement laboratories (counting rooms) are located in the laboratory building. In addition to other special counting equipment, such as a liquid scintillation counter and a low background beta counter, there is a high-resolution gamma-ray spectrometer system consisting of a 45-cm³ coaxial Ge(Li) detector coupled, through a preamplifier, to a Packard 4096 multi-channel analyzer system. The readout equipment includes display oscilloscope, IBM 029 card punch, Franklin high-speed printer, tally punched paper tape, magnetic tape with tag word identification and omnigraphic analog plotter. Auxiliary electronic equipment includes two linear amplifiers and base line restorers to provide separate front-end electronics for NaI(Tl) and Ge(Li) detectors, and a precision pulser and stabilizer to enhance the system stability. Also available is a high resolution Si(Li) x-ray detector for use with the 4096-multichannel analyzer. The system is capable of resolution of 2.5 keV (FWHM for the 1.33 MeV ⁶⁰Co), with a peak height-to-Compton ratio of approximately 26:1. Computerized, routine, multi-element activation analysis methods are being developed and evaluated.

Research Reactor (3)

The Research Reactor is a flux trap reactor fueled with uranium highly enriched in ²³⁵U. Other descriptive features of the reactor are: pressurized (60 psi) tank type, light water moderated and cooled, beryllium and graphite reflected, reflector controlled, and forced convection cooling systems.

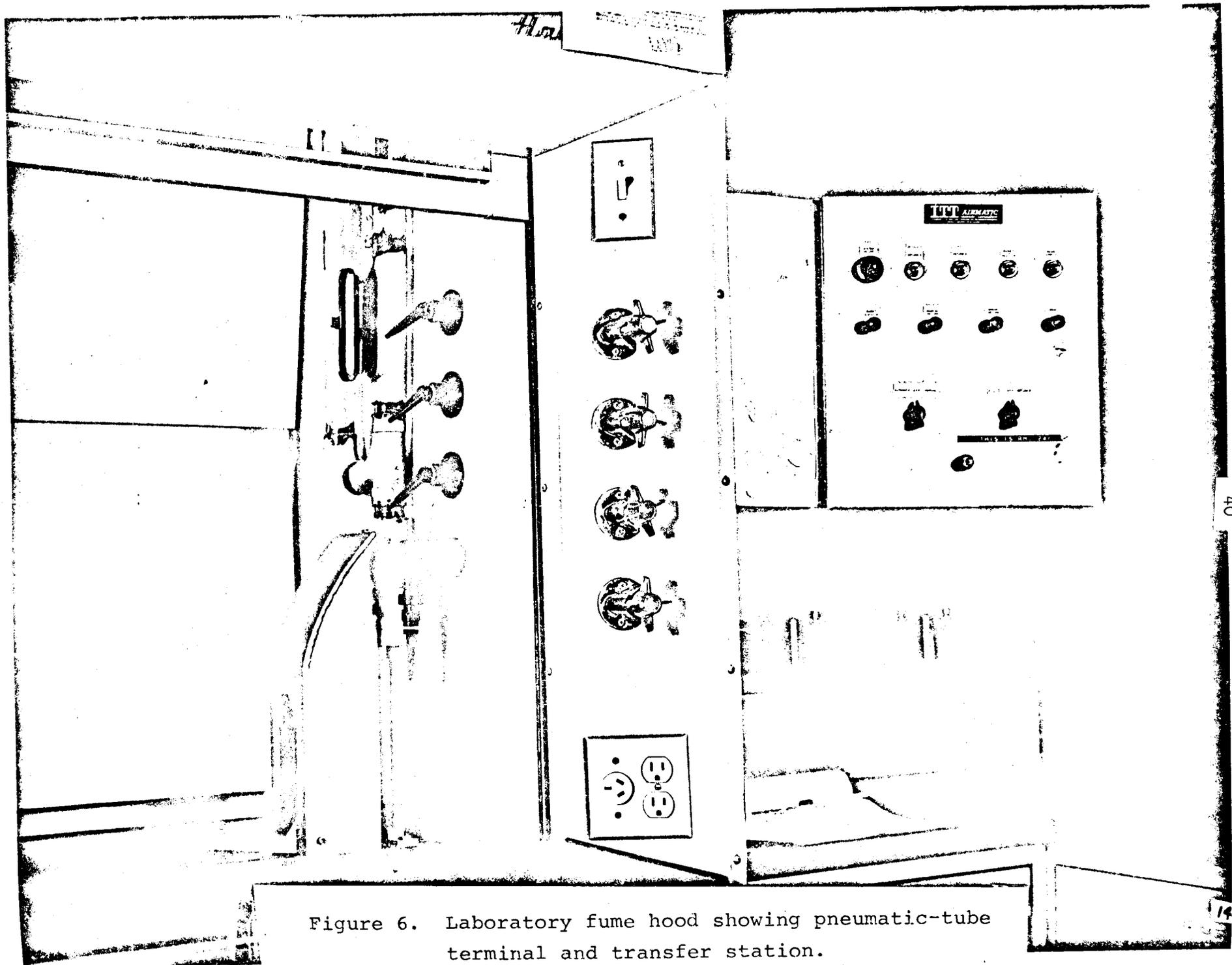


Figure 6. Laboratory fume hood showing pneumatic-tube terminal and transfer station.

The reactor is shown in operation in Fig. 7. The reactor core is an annular cylinder 12 in. in outside diameter and 24 in. high. The core consists of eight plate-type fuel elements (Fig. 8). The center of the core is unpressurized and contains the flux trap. Surrounding the core ^{are} the beryllium and then the graphite. The control shroud consists of five independently movable blades in a water gap between the core and the beryllium reflector.

Since the prime purpose of the University of Missouri reactor is research, many experimental facilities are available. The flux-trap provides the highest thermal flux. A hollow aluminum center-hole canister holds samples in the flux trap.

The graphite reflector section forms an irradiation facility. Its 12 modified reflector elements contain ^{sample} holes for irradiation. Four of the reflector elements are used as ^{irradiation} positions for the pneumatic-tube system. This system is a standard 1.5-in. diameter vacuum-operated system, which can transport irradiation containers from the laboratories of the Laboratory Building into the Reactor Containment Building through the biological shield and to the reactor positions.

The reactor has a thermal column for experiments with thermal neutrons. This thermal column is presently being used for neutron radiography (Fig. 9). Figure 10 shows a neutron radiograph taken without collimation. A collimator being constructed is expected to significantly enhance resolution. At present all radiographs are taken using a transfer technique with indium foil.

The reactor has three 4-in. diameter and three 6-in.

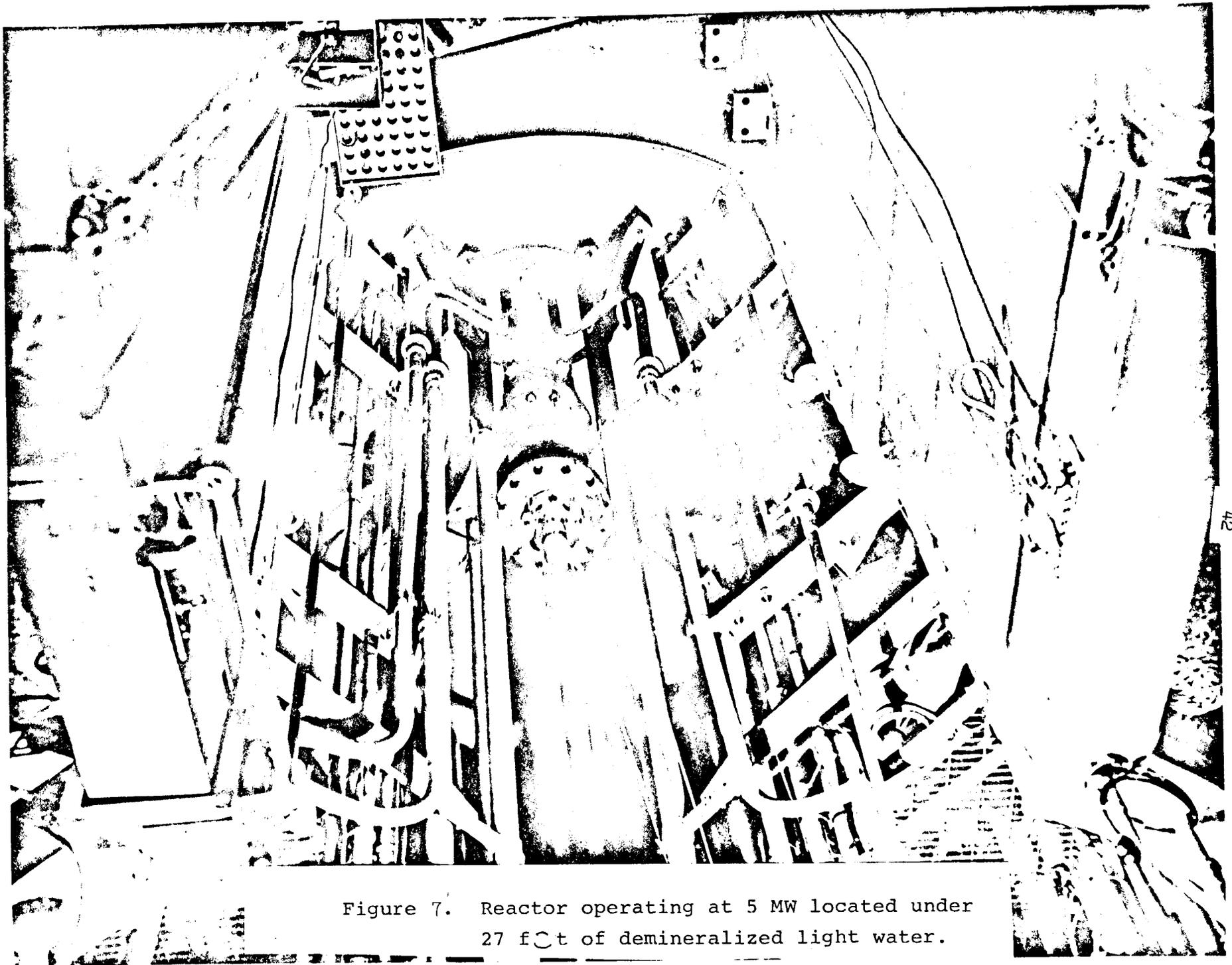


Figure 7. Reactor operating at 5 MW located under 27 fct of demineralized light water.

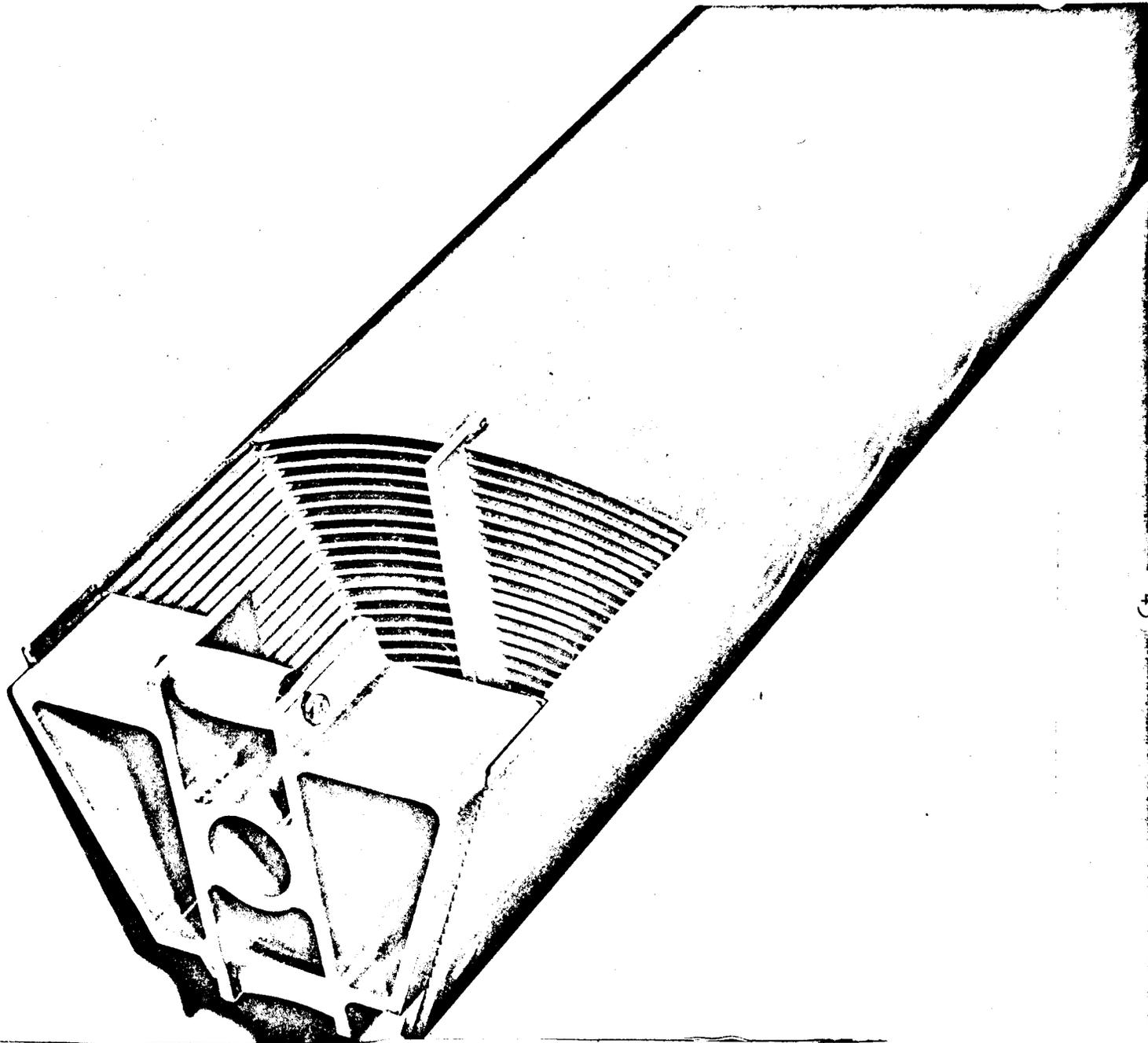


Figure 8. An end view of one of eight fuel elements. Each element contains 650 grams ^{235}U . The active core length is 24 inches.

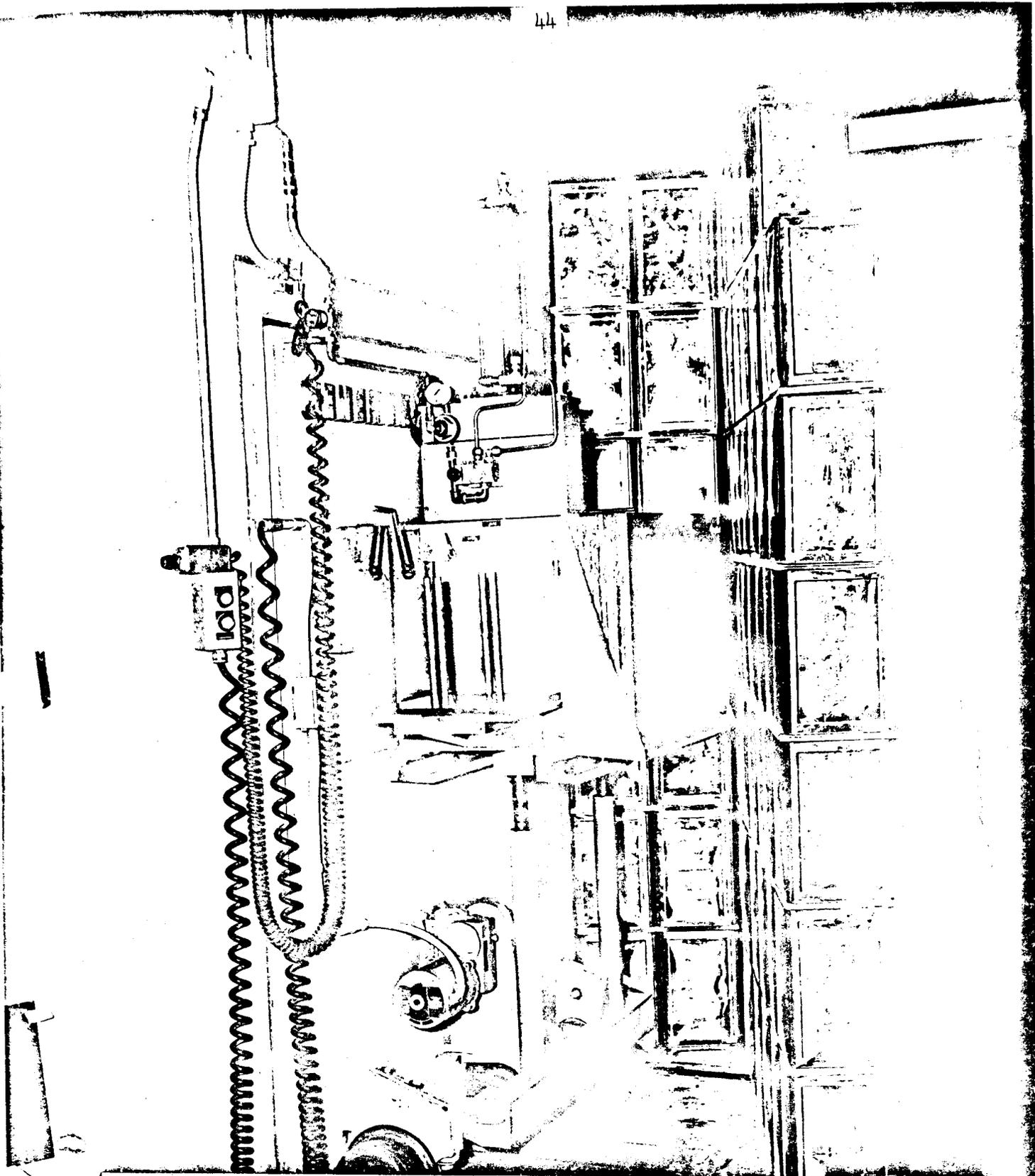


Figure 9. The neutron radiography facility is installed in the reactor thermal column. Cans of borated water provide supplemental shielding to adjacent experiments.

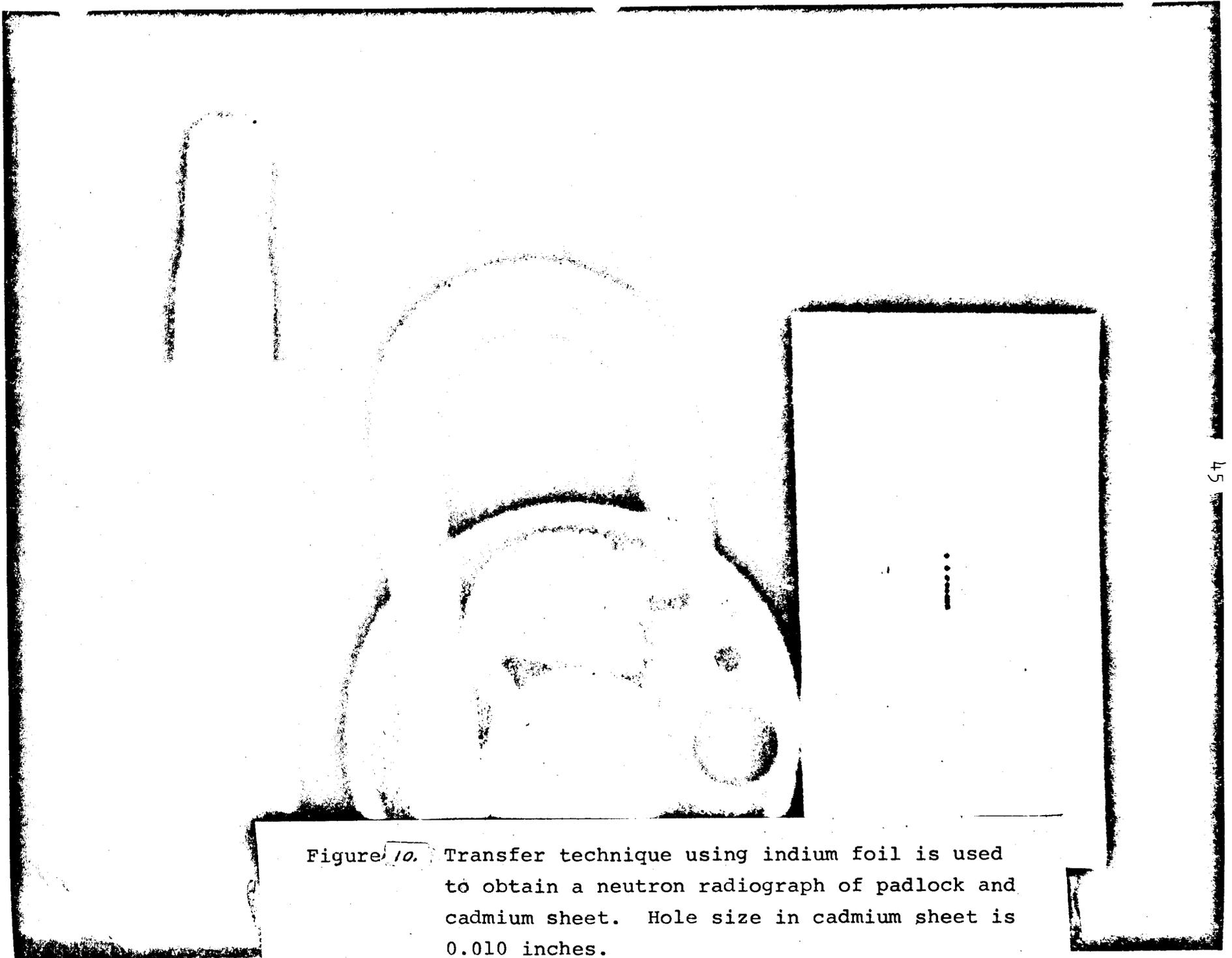


Figure 10. Transfer technique using indium foil is used to obtain a neutron radiograph of padlock and cadmium sheet. Hole size in cadmium sheet is 0.010 inches.

diameter beam tubes. These tubes provide collimated neutron beams from the reactor core to experimental facilities located around the outside of the biological shield of the reactor. All six beam tubes penetrate the graphite reflector. Two of the 4-in. tubes also penetrate the beryllium reflector to provide a larger ratio of fast to thermal neutrons. To reduce the fast neutron and gamma-ray background, one 4-in. and one 6-in. beam tube do not view the core directly, but terminate below the fuel level of the core. When a port is not being used as an experimental facility, a shield plug is installed and the port is flooded with water to complete the reactor shield.

Table 1 presents pertinent characteristics of the University of Missouri Research Reactor, and Table 2 provides neutron flux data on experimental positions in the facility. For reference to the experimental facilities, a plan view of the reactor core region is shown in Fig. 11.

TABLE 1
SUMMARY OF PERTINENT REACTOR DATA

POWER

Initial power capability, MW	5
Eventual power with modification, MW	10
Average power density at 5 MW, kW/liter	151
Average power density at 10 MW, kW/liter	303

REACTOR CORE

Geometry	Annular
Inner fuel radius, in.	2.66
Outer fuel radius, in.	5.90
Active fuel height, in.	24.0
Active volume of core, liters	33.0
Fuel loading for 5 MW operation, kg ²³⁵ U	5.2
Number of fuel elements	8
Fuel plates per element	24
Cladding of fuel plates	Aluminum
Coolant gap thickness between plates, in.	0.080
Fuel plate thickness, in.	0.050
Fuel thickness, in.	0.020
Clad thickness, in.	0.015

CORE COOLANT

Total flow rate @ 5 MW operation, gpm	1800
Coolant pressure, psia	65
Inlet temperature, °F	140
Outlet temperature, °F	157
Demineralizer flow, gpm	50

POOL COOLANT

Flow rate @ 5 MW operation, gpm	400 to 600
Mixed pool temperature, °F	100
Pool inlet temperature, °F	99
Pool outlet temperature, °F	105
Demineralizer flow, gpm	50

ASSORTED CHARACTERISTICS

Reactivity requirements ($\Delta k/k$) @ 5 MW	0.0973
Rod worth (Δk)	0.259
Core lifetime, MW days	560
U-235 consumed, Kgm	0.7

REFLECTORS

Inner reflector	Beryllium
Inner reflector thickness, in.	2.71
Outer reflector	Graphite
Outer reflector thickness, in.	8.89
Height, in.	36
Coolant	Pool water

CONTROL RODS

Location	Outside vessel
Type	Curved plate
Material	Boral
Clad	Aluminum
Overall thickness, in.	0.200

TABLE 2
 NEUTRON FLUX AVAILABLE IN THE UNIVERSITY
 OF MISSOURI RESEARCH REACTOR AT 5 MW

<u>POSITION</u>	<u>THERMAL FLUX, n/cm²·sec</u>	
← Flux-trap	3.7 x 10 ¹⁴ max.	2.3 x 10 ¹⁴ av.
← 6-in. Beam Ports		
1) Port B	5.3 x 10 ¹³	
2) Port D	3.0 x 10 ¹³	
3) Port E	5.0 x 10 ¹³	
← 4-in. Beam Ports		
1) Port A	7.5 x 10 ¹³	
2) Port C	3.0 x 10 ¹³	
3) Port F	7.5 x 10 ¹³	
← Pneumatic Tube System		
1) Tube G-1	4.8 x 10 ¹³	
2) Tube G-2	3.9 x 10 ¹³	
3) Tube J-1	6.2 x 10 ¹³	
4) Tube J-2	4.3 x 10 ¹³	
← Reflector Positions	<u>MAX.</u>	<u>AV.</u>
1) Position H-1	5.0 x 10 ¹³	2.7 x 10 ¹³
2) Position I-1	5.0 x 10 ¹³	2.7 x 10 ¹³
3) Position H-2	2.0 x 10 ¹³	1.3 x 10 ¹³
4) Position I-2	2.0 x 10 ¹³	1.3 x 10 ¹³
5) Position G-3	8.0 x 10 ¹²	5.8 x 10 ¹²
6) Position H-3	8.0 x 10 ¹²	5.8 x 10 ¹²
7) Position I-3	8.0 x 10 ¹²	5.8 x 10 ¹²
8) Position J-3	8.0 x 10 ¹²	5.8 x 10 ¹²
← Thermal Column	Variable thermal flux from 10 ¹² to 10 ⁸	

The data in Table 2 are for 5-MW power levels of operation. At 10-MW, the flux in each position will approximately double.

In addition to the positions shown in Table 2, a sector of the reactor pool is available for the irradiation of large items at flux levels up to 10¹² n/cm²·sec.

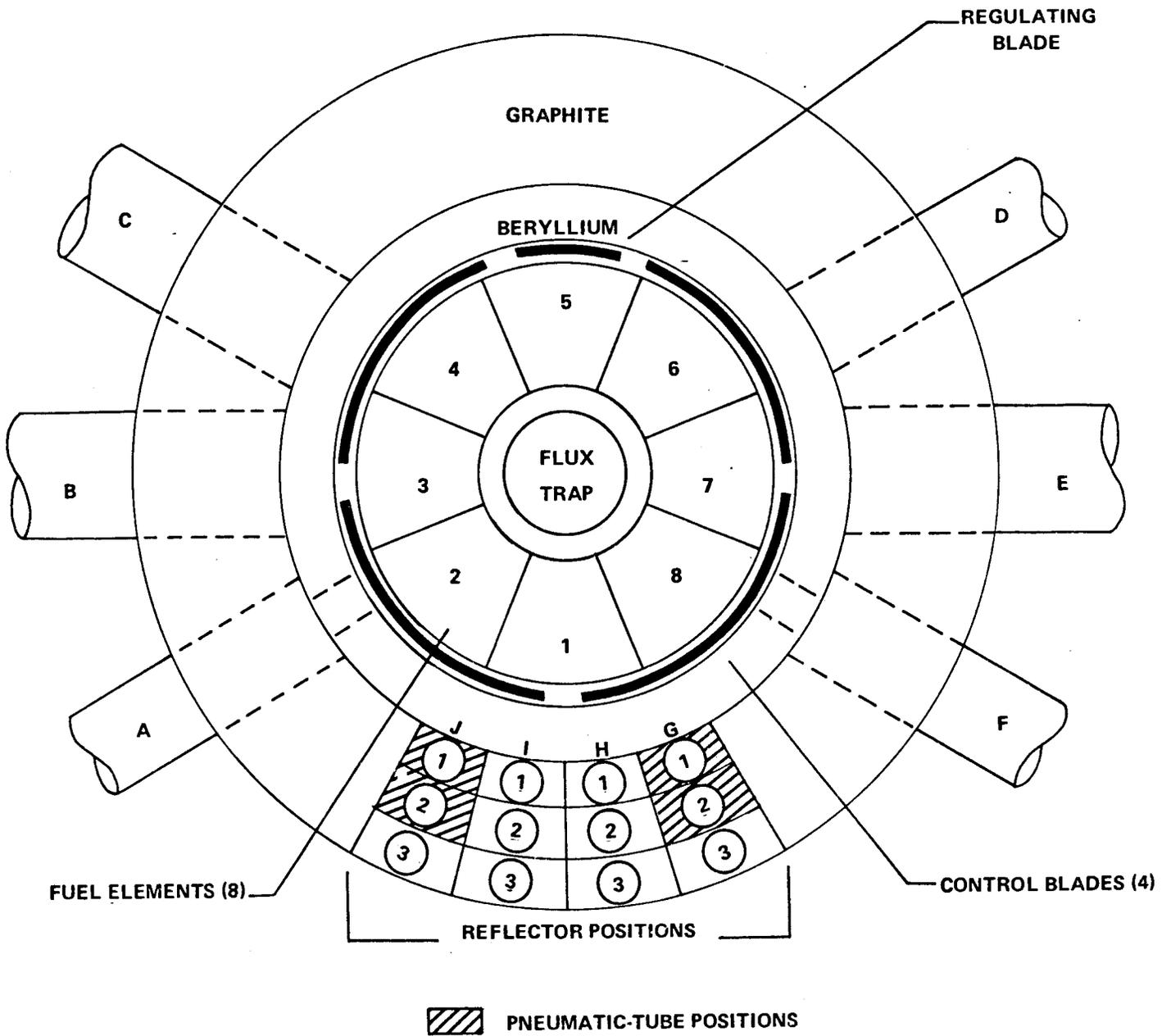


Fig.11. Flux Trap, Beamport, and Reflector Experimental Facilities

Irradiation Facility (3)

Completing the major experimental facilities is the ^{60}Co irradiation facility (Fig. 12) and associated hot cell. Provision for 12 kilocuries of ^{60}Co are made in a 16-ft deep pool of water in the basement of the laboratory building. By means of an elevator system, the source may be used for irradiations in air or water. The source, now amounting to approximately 4 kilocuries consists of ten stainless steel clad cylinders of cobalt equally spaced circumferentially on a diameter of 5.5 in.

The hot cell is designed to accommodate multi-curie levels of radioactive materials. The cell is equipped with leaded glass shielding windows and two sets of master slave manipulators.

(RHL)

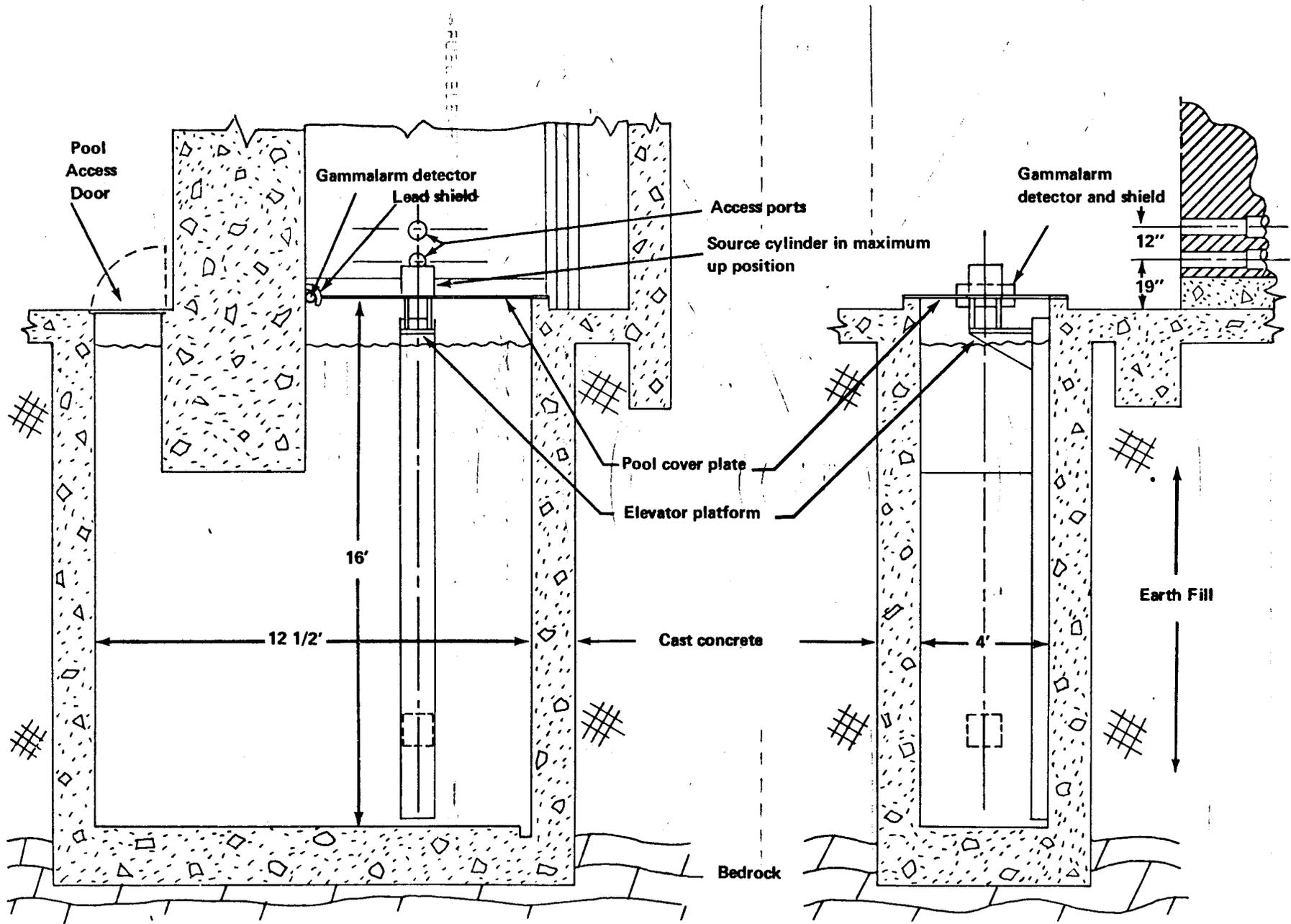
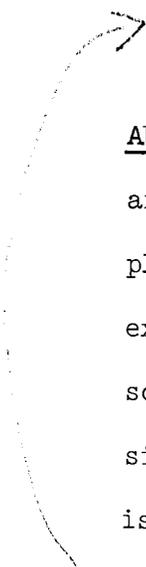


Fig. 12. ⁶⁰COBALT IN-AIR IRRADIATION FACILITY

NUCLEAR APPLICATIONS IN HIGHWAY RESEARCH (2)

Supplemental Keywords: (LUMINESCENCE / INDUSTRY / POLLUTION / INDUSTRY,
STONE, CLAY AND GLASS / POLYMERIZATION / MEASUREMENT, DENSITY/ X-RAY
FLUORESCENCE / MEASUREMENT, THICKNESS / INDUSTRY, TRANSPORTATION / COMPARISON /
BACKSCATTER / CONFERENCE)



Abstract: Several devices and/or techniques involving highway-related research are described: (1) portable nuclear gage for measuring cement content of plastic concrete; (2) x-ray fluorescence systems for analyzing lead in automobile exhaust and calcium in raw cement mix; (3) a dual-type gage for gamma-ray backscatter measurement of soil density; and (4) a self-luminous overhead traffic sign activated by ^{85}Kr . Polymer-impregnated concrete for highway applications is also discussed.

Editors' Note.

On January 12-16, 1970, the Highway Research Board held their 49th annual meeting in Washington, D. C. One of the sessions entitled "Nuclear Applications" included several papers related to the use of isotopes and radiation in highway programs. The six items below are edited versions of papers given at that session.

PORTABLE NUCLEAR GAGE FOR MEASURING CEMENT CONTENT
OF PLASTIC CONCRETE (2)

P. F. Berry*

(i.e., between delivery and pouring)

Rapid, on-site, analysis of the cement content of wet concrete is an outstanding requirement in civil engineering. A variety of methods, some nucleonic, are available to determine such parameters as water content, bulk density, and the degree of air entrainment, and such measurements can provide valuable advance information on the concrete strength. Suitably rapid and representative methods for the determination of the cement content, however, have not so far been developed.

The instrumentation for such analyses should be portable, rugged and relatively easy to operate. The measurement should be capable of an accuracy of 0.3 sacks/cu.yd. or better, in a measurement time of less than 1 minute, and should require the minimum of sample preparation.

Method of Analysis (3)

Quantitative analysis of cement in concrete is made possible by virtue of its relatively unique and substantially constant chemical composition. Of the two more common nuclear methods of analysis,

→ Activation analysis is not considered suitable for on-site application. X-ray fluorescence techniques, using radioisotope sources of excitation were found to be impractical to apply in concrete analysis on account of the extreme heterogeneity^{of the samples} over the depth of penetration of the fluorescence x rays — i.e., a few hundred microns.

*Texas Nuclear Corp., Austin, TX.

On the other hand, the method considered here depends in principle on the relatively high atomic number and specific gravity of cement as compared to the rest of the concrete matrix. This contrast can be measured most readily by means of radiation absorption and scatter techniques employing low energy γ or X-radiation. Though non-specific, in the true analytical sense, these techniques are capable of high accuracy for essentially binary mixtures and furthermore offer some latitude in the choice of radiation energy in order to reduce the effects of sample heterogeneity.

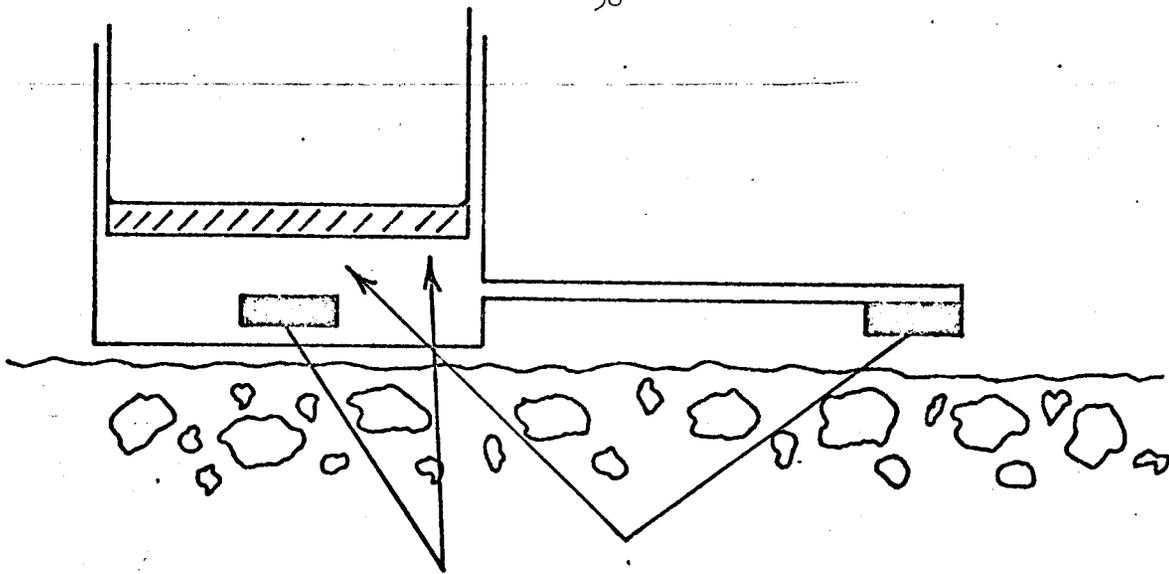
>The radiation emerging from the sample may be measured in either the transmission mode or in the back-scattered mode, as illustrated in Fig. 1.

Preliminary Studies (3)

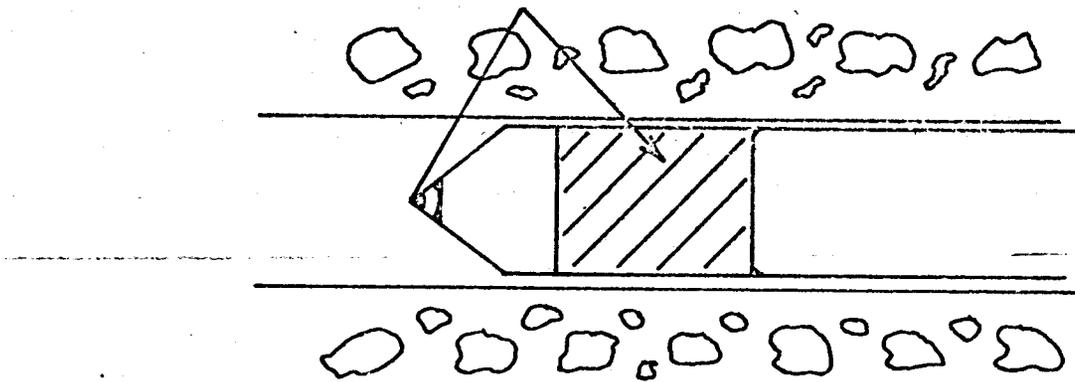
During the investigation, each of the geometrical arrangements of source and detector (shown in Fig. 1) was investigated with a view to establishing the sensitivity to changes in cement content and also the influence of aggregate size and composition, sample heterogeneity, and other sample variables such as water content and bulk density.

The detectors were NaI(Tl) scintillation detectors with crystals of the appropriate dimensions; a variety of X- and low energy gamma-ray sources including ^{109}Cd (22 keV), ^{241}Am (60 keV), and ^{57}Co (122 keV) were used. A higher energy γ -ray source, ^{137}Cs , was also used for bulk-density measurement.

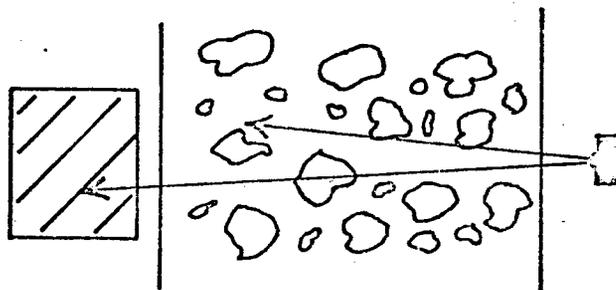
Measurements were carried out on actual concrete mixes in both the wet and cured state. The latter samples, annular in shape to accommodate the depth-type backscatter gauge, were prepared mainly for reference purposes and to



SURFACE BACKSCATTER GEOMETRY



DEPTH BACKSCATTER GEOMETRY



TRANSMISSION GEOMETRY

Figure 1. Geometrical Arrangements Employing Low Energy Radiation Scatter and Transmission for Cement Analysis.

evaluate further gauge improvements.

Most of the experimentation has involved the backscatter techniques since they afford greater simplicity in gauge design. The same probe contains both the source and detector and thus there are no problems of physical displacement of one relative to the other. Variability of sample mass and shape is also less of a problem, since it can be arranged to be effectively infinite to the scattered radiation.

In view of the far superior performance of the depth-type backscatter probe over the surface-type geometry (which is sensitive to changes in surface conditions --- e. g., surface free-water seepage and sample compaction), the depth-type configuration was adopted.

Studies With the Depth Probe (3)

The physical configuration of the depth probe consists of a 1-inch-diameter by 1-inch-thick NaI(Tl) crystal scintillation detector and a 2-mm-diameter spherical 14-mCi ^{241}Am source mounted on a conical radiation shield and axially separated from the crystal at a distance of about 2 inches. The source and detector assembly are encased in a 0.020-inch-wall stainless steel tube. The initial backscatter measurements (Fig. 2) were made on simple concrete mixes to which no air entrainment resin was added and of a constant water content of about 7%. A siliceous aggregate was used in the usual proportion of 3 parts coarse (maximum size 1.5 inches) to 2 parts fine aggregate.

Each data point represents the average of 5 discrete point-to-point measurements made throughout the sample, and the error bars represent the standard deviation in this average deduced from measurements on 6 remixes of the same sample. The error reflects the coarse aggregate and normal heterogeneity effects, and runs about ± 0.1 sack/cu.yd. Clearly one would not be so naive as to assume that this is an achievable

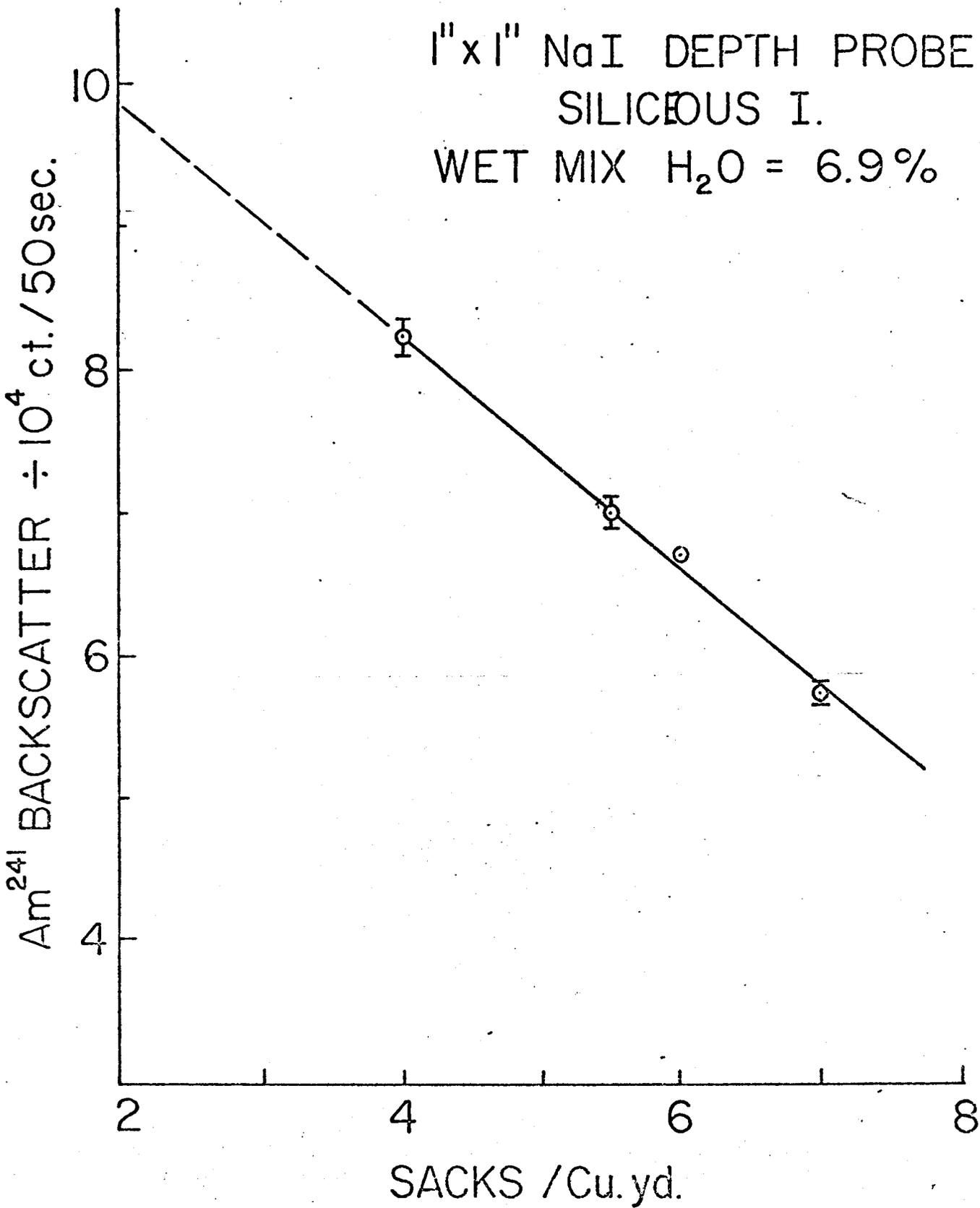


Figure 2. Am²⁴¹ Backscatter Depth Probe Response.

accuracy in practice. Apart from the problem of variations in aggregate composition, the major variables encountered in practice are water content and bulk density, the latter variation being induced deliberately by air entrainment resin or naturally as a result of variations in water content, cement content, air entrapment, and other sources.

Effects of Variations in Water Content (3)

The effects of variations in water content and bulk density were next investigated (Fig. 3).

Although it may appear that variable water content has a significant effect on the ^{241}Am backscatter intensity, the effect is only partially attributable to water content per se. Taking the results in Figs. 3a and 3b together, the principal effect appears to arise from an accompanying density change (as measured by a standard surface-type gamma scatter gage).

Comparison of Theory and Experiment (3)

The theoretical gage performance in terms of the backscattered ^{241}Am intensity as a function of cement content, water content, and bulk density has been calculated and compared with experiment (Fig. 4)

The Am backscatter intensity has been plotted as a function only of bulk density in each cement content group. The calculated response curves have been normalized as a group to the experimental points, and agreement is excellent. In particular, the correlation with bulk density and relative independence of water content per se should be noted.

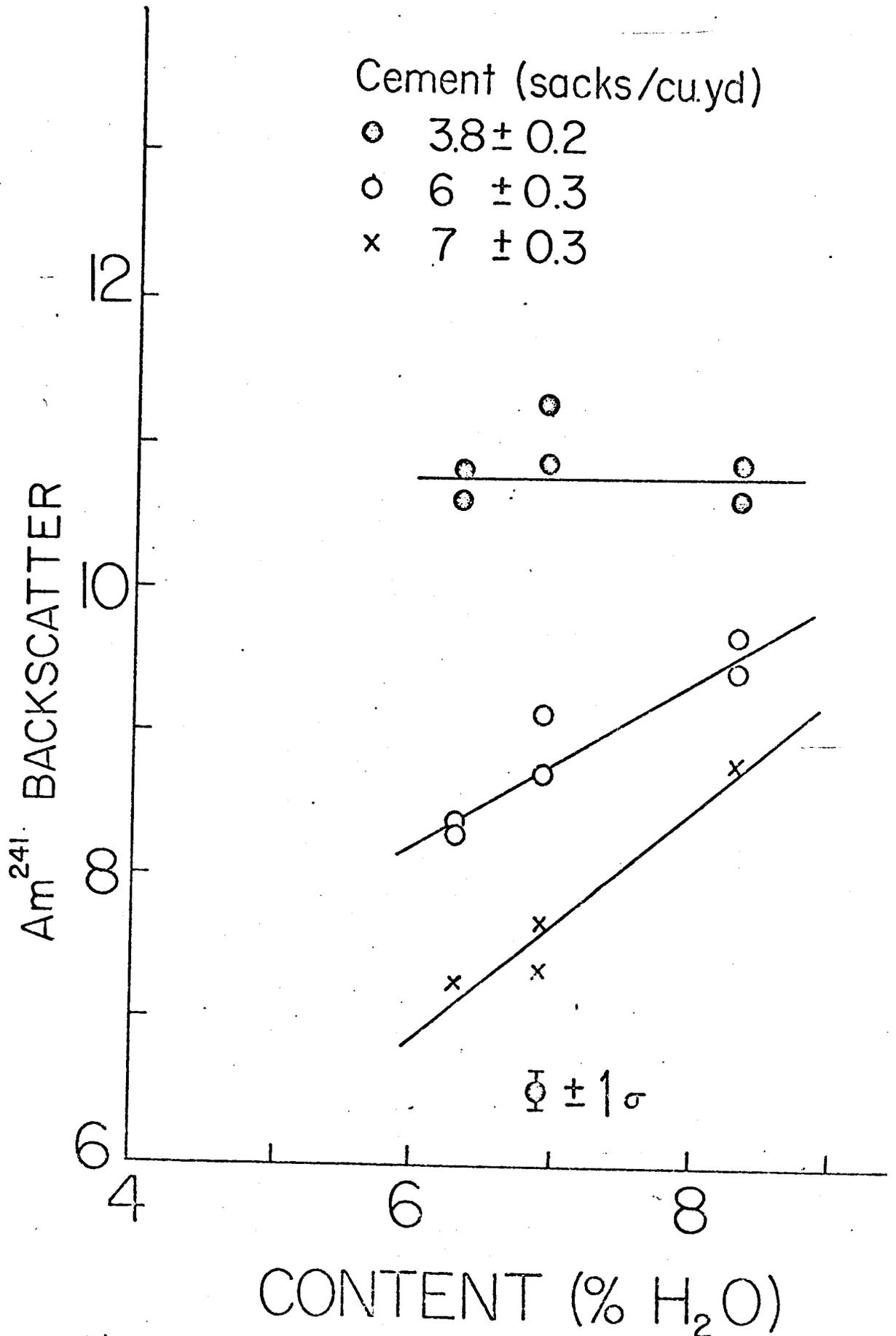


Figure 3a. Water Content Effect on Backscattered Intensity

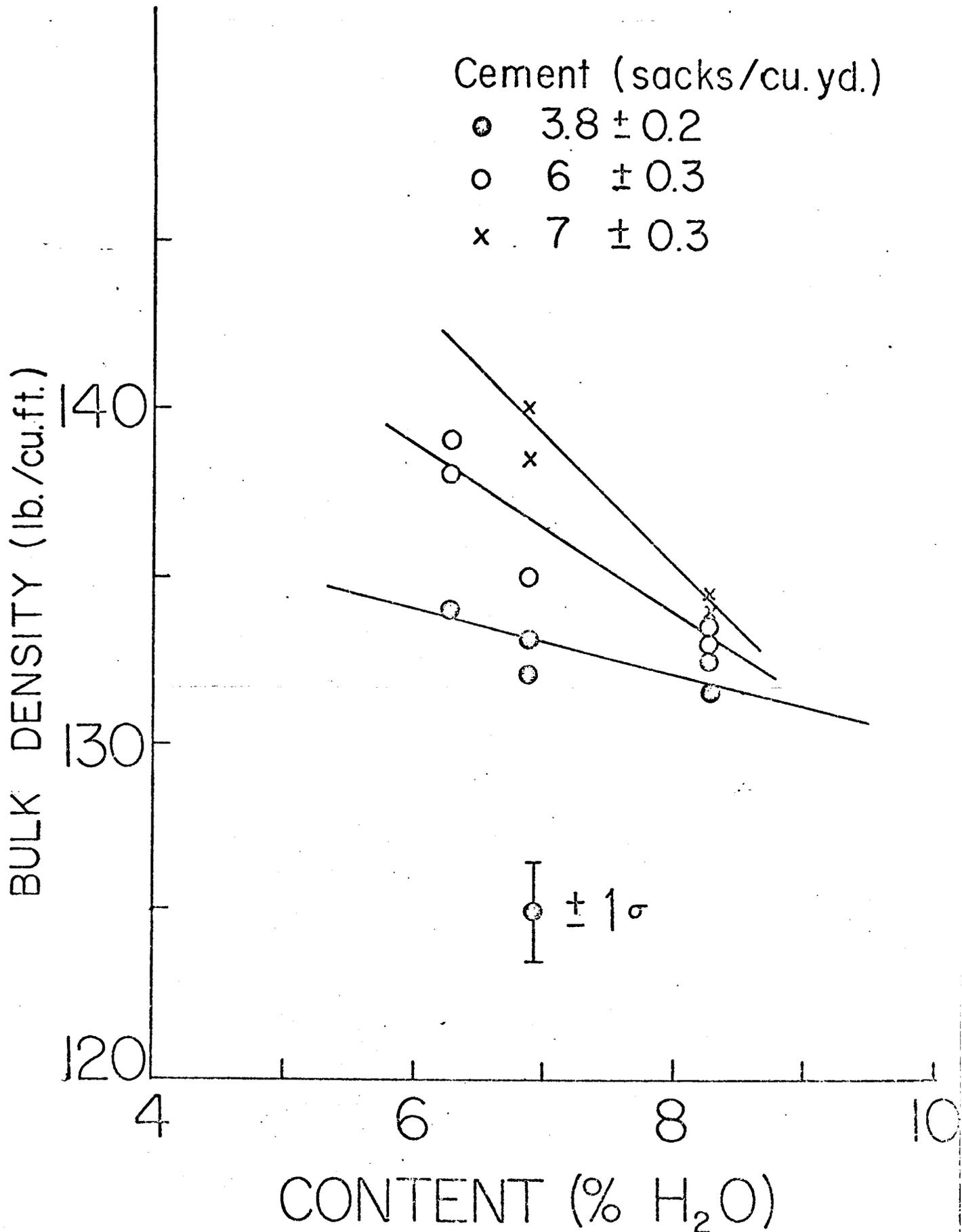


Figure 3b. Water Content Effect on Bulk Density

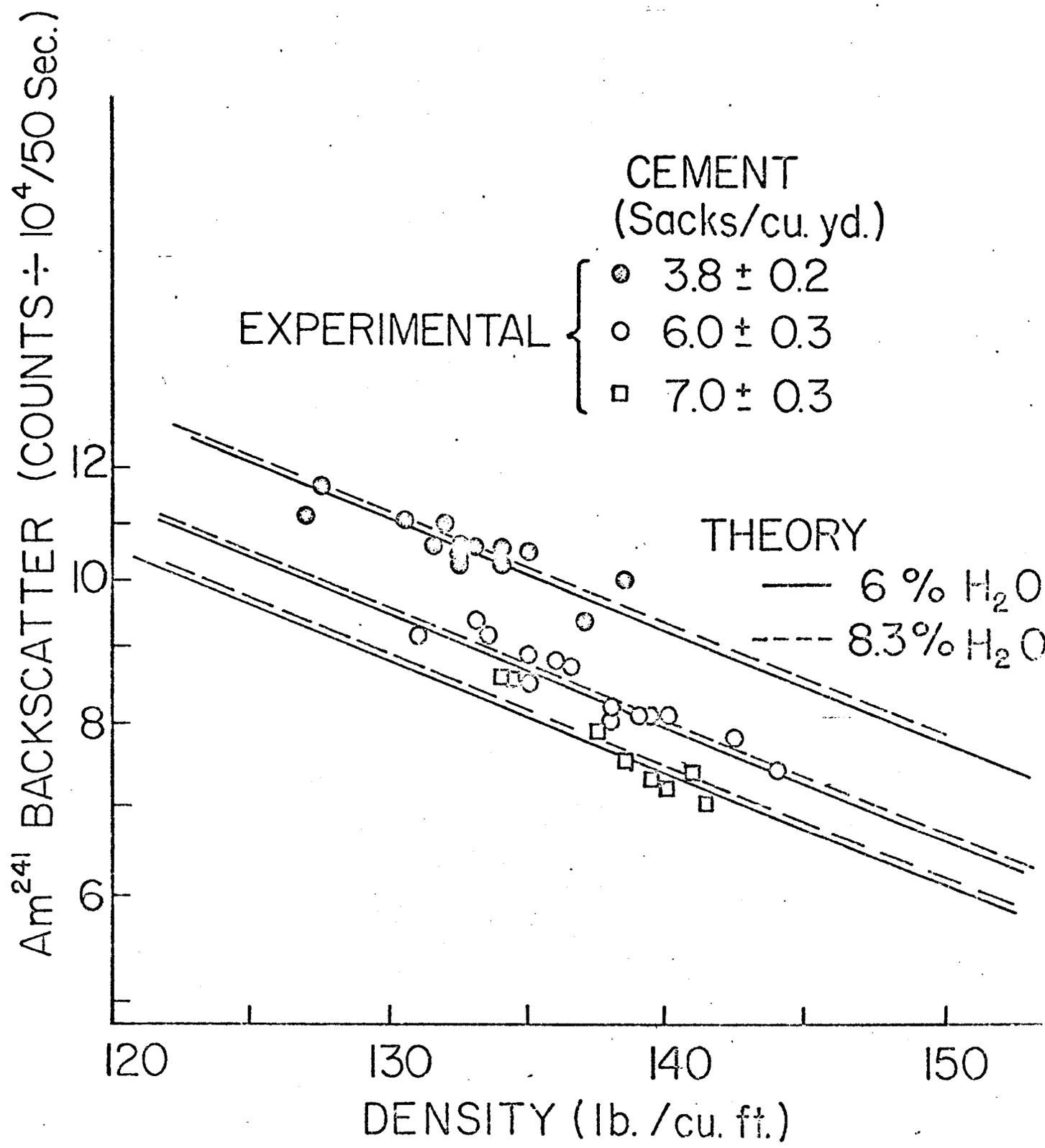


Figure 4. Comparison of Theory and Experiment

The results of the preliminary studies indicate that for a particular type of aggregate, two measurements—an ^{241}Am backscatter measurement and a measurement of bulk density---will suffice to determine the cement content. Other sample variables such as aggregate size, water content, and air entrainment are automatically compensated for. Differences in type of aggregate (i.e., chemical composition) clearly dictate a separate calibration in each case. In this respect, our experience so far has been limited to only a few different types of aggregate, mainly of the silicious type. For these aggregate mixes, a normalization procedure based on intensity measurements on a sample of the raw aggregate was sufficient. Since a sample of the aggregate is usually made available for the purpose of other tests, calibration for each aggregate mix is a practical possibility.

Instrument Design and Performance (3)

Design (4)

The instrument comprises essentially two parts; the submersible probe unit which contains the radiation detector and sources and the electronic unit which provides the detector high voltage, pulse amplification, and count-rate display.

The main features of the probe layout are shown in Fig. 5. The density-measurement feature is provided within the probe and consists of a ^{137}Cs γ -ray source located about 10 in. behind, and shielded from, the same detector which is used to detect the ^{241}Am scattered radiation. The assembly is hermetically encapsulated in an aluminum housing that can be mounted within the hand-held accessory (Fig. 5) or within the sample-containing device (Fig. 6).

The latter arrangement is recommended for the best accuracy for several reasons. In the first place, although the ^{241}Am intensity is not affected by the volume of sample presented to the gauge (provided the radial dimension is more than 3 inches or so) the ^{137}Cs intensity is affected

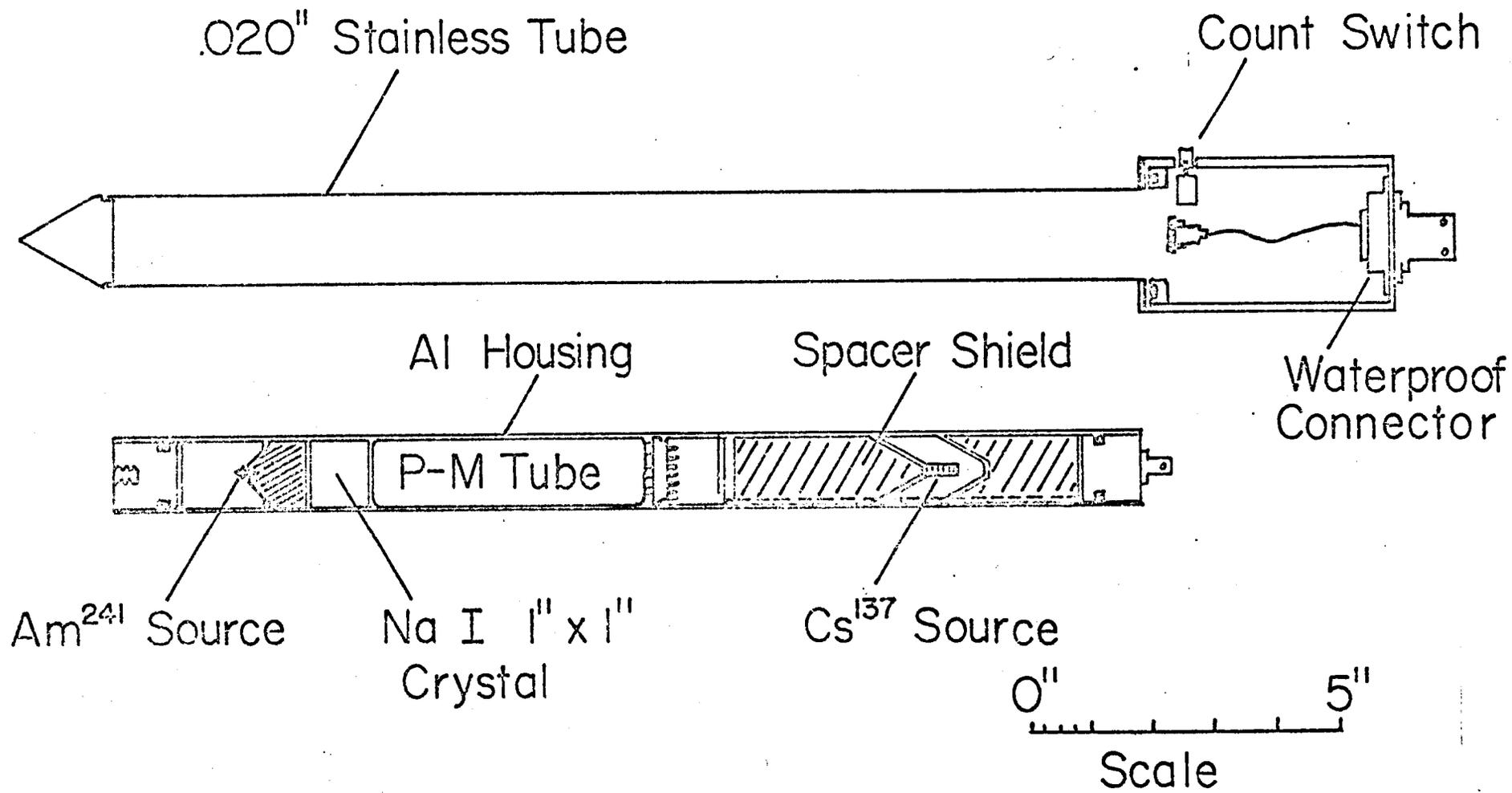


Figure 5. Probe Layout.

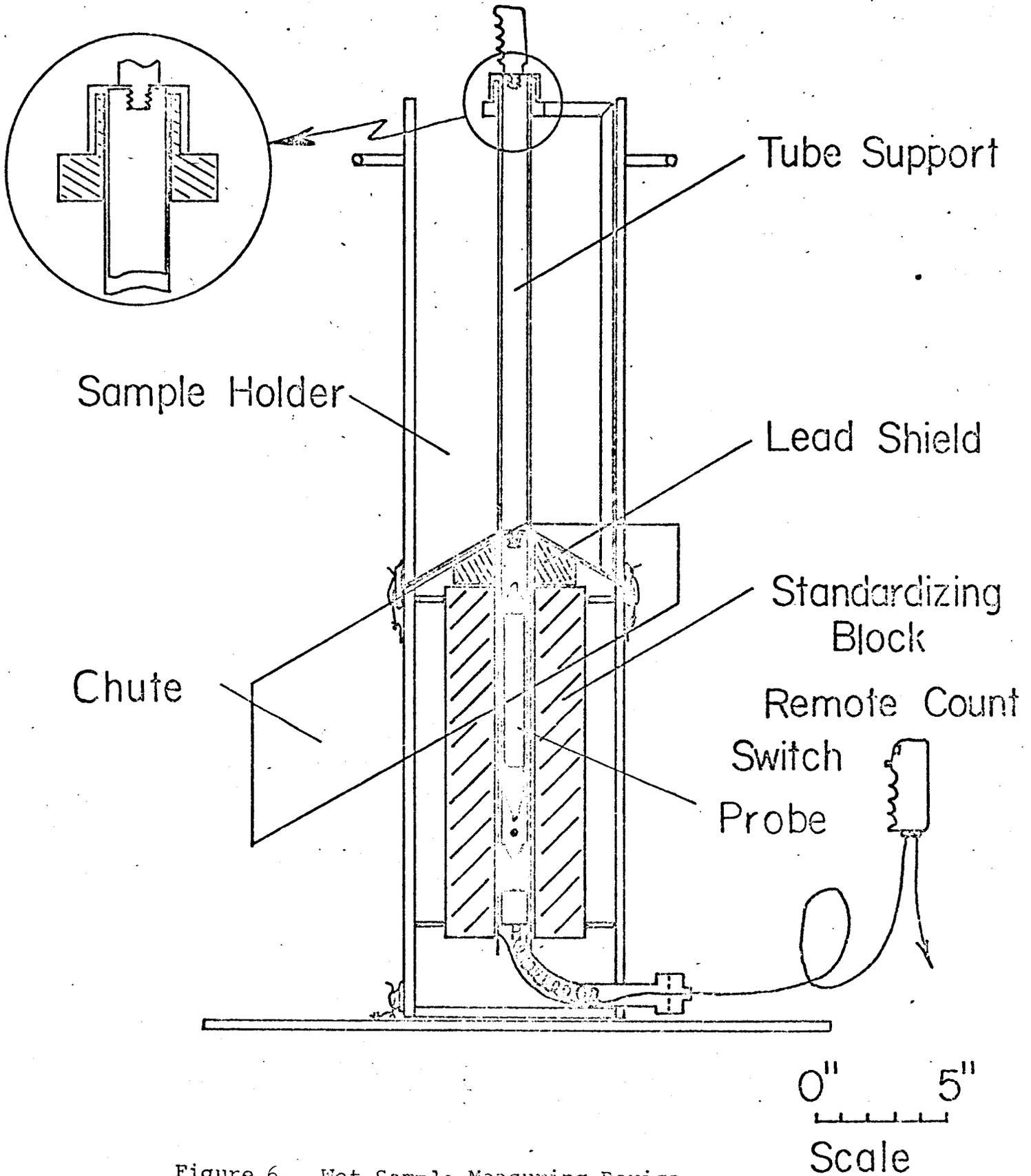


Figure 6. Wet Sample Measuring Device

unless the volume is considerable (i.e., at least 6 inches around the probe). In the sample-containing device, the critical sample dimensions are held constant. A second feature of the containing device, which promotes good accuracy, is the built-in standardization block in which the probe is located before each measurement. (The lead shield which divides the measuring compartment from the standardizing compartment suppresses the nonrepresentative contribution of the ^{137}Cs intensity to the measured intensity until the probe is drawn up into the final position, when both intensities are recorded. At the intermediate positions, the ^{241}Am measurement is relatively unaffected by the ^{137}Cs source.

The backscattered radiation spectra are shown in Fig. 7 . The electronic unit employs a single channel analyzer which is set on the ^{241}Am backscatter peak. Pulses above the upper level of the analyzer window are registered in a separate scaler contained within the unit. Either channel of information can be displayed at the end of the count by the throw of a switch.

Preliminary Results (4)

Some preliminary results with cured concrete annular blocks, using the dual-source probe, are shown in Fig. 8 . These show the same pattern of behavior as those recorded earlier and establish the effectiveness of the density compensation.)

→ The estimated accuracy of these determinations is better than 0.3 sacks/cu.yd. The counting time in each measurement position was 20 seconds and the overall measurement time per sample was about 2 minutes.

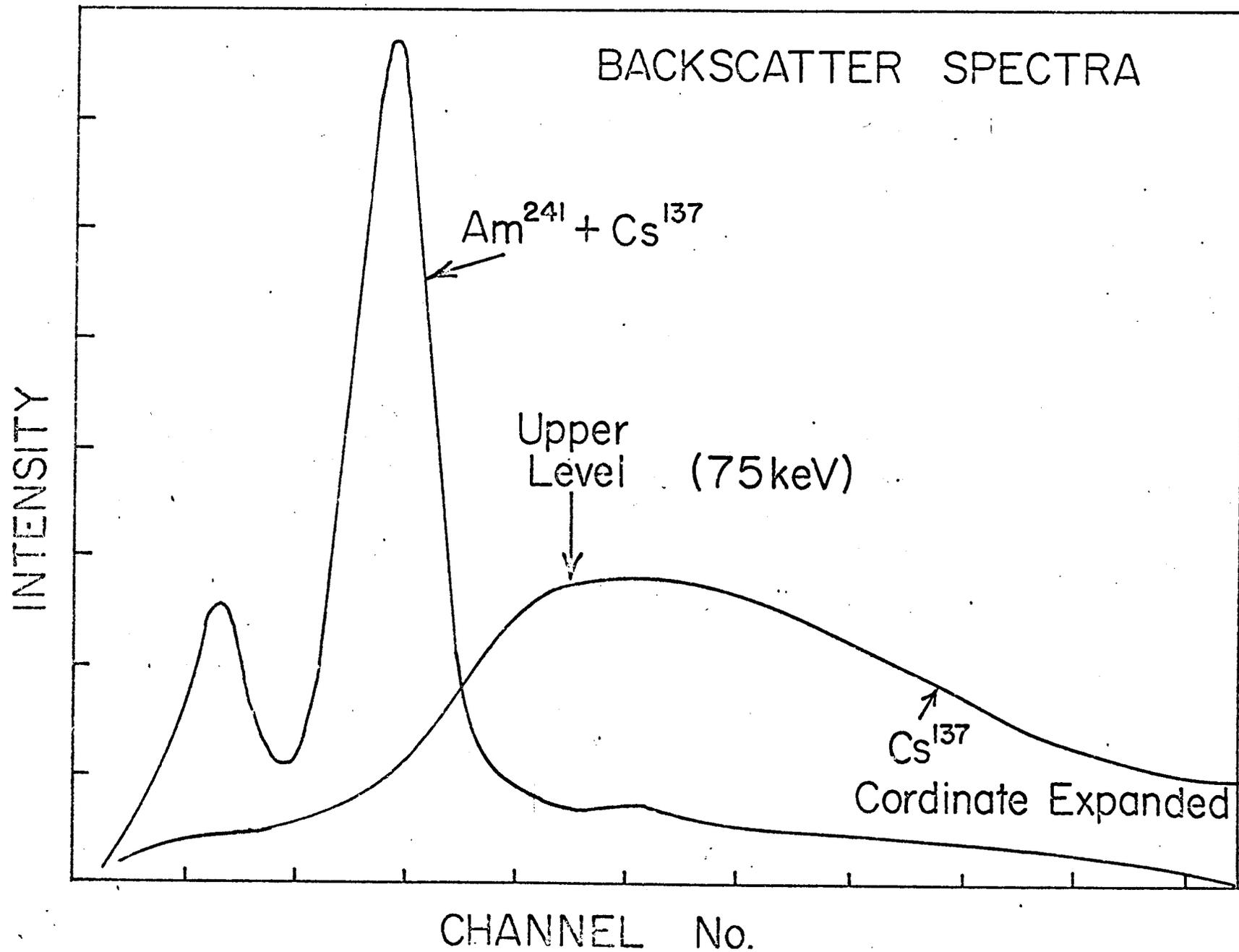


Figure 7. Backscatter Spectra

SILICEOUS I.
CURED SAMPLES

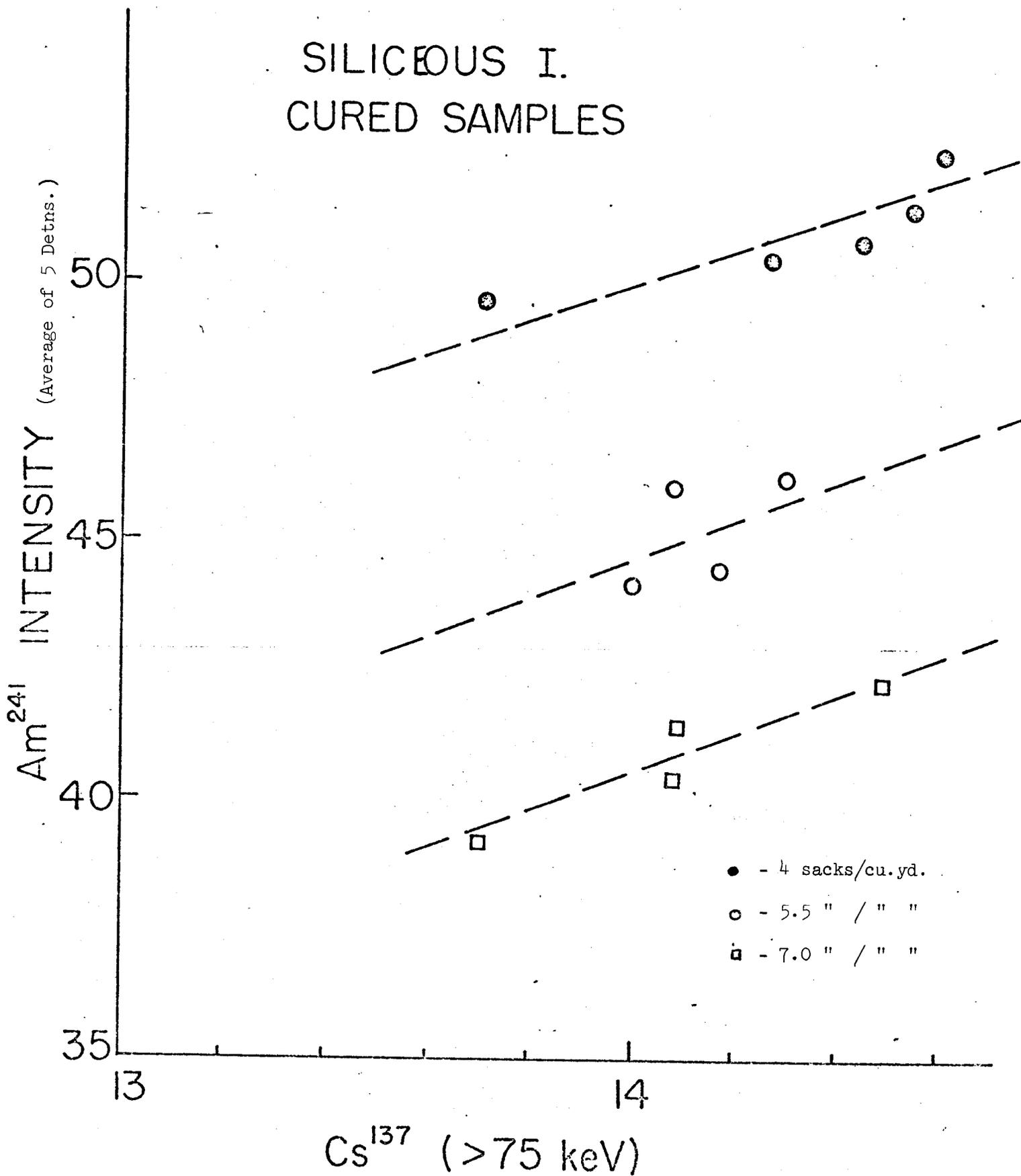


Figure 8. Typical Calibration Nomograph

Conclusion

The results obtained so far are certainly encouraging. Some improvements in the electronics are currently underway to provide better field operation—for example, to minimize the effects of electronic drift and gain variations. Further work on a wider variation of aggregate types has still to be carried out and field tests have to be initiated. We expect the instrument to be available for these tests during early 1970.

POLYMER-IMPREGNATED CONCRETE FOR HIGHWAY APPLICATIONS (2)

R. G. Pike*

When hardened portland cement concrete is impregnated with monomer and the monomer then polymerized by radiation, strengths almost four times the strength of the regular concrete can be obtained. This technique, originally suggested by the U.S. Bureau of Reclamation and jointly developed with AEC's Brookhaven National Laboratory (BNL), is being investigated further by the U.S. Bureau of Public Roads (BPR) and BNL for possible use in highway applications.

Preparation of Polymer Concrete (3)

In the preparation of the polymer concrete, regular concrete, made in the usual way with portland cement, water, and aggregates, is aged for 28 days (or in some cases for a shorter period), dried at 150°C, evacuated, saturated with methyl methacrylate monomer, then irradiated for about 8 hours with ^{60}Co at a dose rate of about 5×10^5 rad/hr to polymerize the monomer.

Properties of Polymer Concrete (3)

The engineering properties of the "Polymer Concrete" of major interest are listed in Table I,^{1/} which shows a comparison of the values obtained on regular (control) concrete and similar impregnated concrete. Preliminary estimates indicate that the cost per pound of a product (for example concrete pipe) made with polymer concrete should be about double that of conventional concrete products; however, if full advantage can be taken of the increased strength of the polymer concrete by appropriate reduction in unit thickness, the cost of the finished product could be significantly less than that of the conventional product.

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Applications of Polymer Concrete (3)

One of the ^{serious} / problems in the highway field is the deterioration of bridge decks and reinforcing steel, caused largely by the action of deicing salts under conditions of freezing and thawing. The polymer treatment greatly reduces the water permeability and water absorption of the concrete, which, in turn, is undoubtedly responsible for the greatly increased resistance

to freezing and thawing (Table 1). Because

of these greatly improved properties of the polymer concrete, BPR and BNL are investigating the feasibility of making precast polymer concrete deck slabs and concrete pavements in-place.

Preliminary tests have shown that the surface of concrete impregnated with polymer is much more resistant to abrasion than regular concrete. Such polymer concrete would help to preserve the nonskid properties of decks and pavement and should be of particular interest where studded tires are in common use. More research is needed, however, to develop practical methods of impregnating the surface of existing pavements.

A new safety feature recently developed for highways is the break-away signpost and lightpost. However, if the weight of the poles could be reduced significantly, their efficiency would be greatly increased, particularly when struck at lower speeds by the lighter cars. The BPR-BNL team has recently developed a lightweight concrete (about 50 lbs/cu. ft.) with a compressive strength greater than 2400 psi, and preliminary designs indicate that lightpoles made with this material may be feasible and better than those made with conventional materials.

For further details on the preparation technique, and for results with other monomers, the reader should consult references (1) and (2).

1/ Concrete Polymer Materials, First Topical Report, BNL Report 50134(T-509), Brookhaven National Laboratory, Upton, Long Island, New York. Also, U.S.B.R. general report No. 41, U.S. Bureau of Reclamation, Denver, Colorado, December 1968.

2/ Polymerization Makes Tougher Concrete, J. T. Dikeou, L. E. Kukacka, J. E. Backstrom, and M. Steinberg, ACI Journal, Proceedings V. 66, No. 10, October 1969, pp. 8-9-39.

Table 1

Summary of Properties of Concrete-Polymer Material

	Concrete control specimen (type II cement)	Concrete with up to 6.7 wt % loading of polymethyl methacrylate Co ⁶⁰ gamma radiation polymerized
Compressive strength, psi	5,267	20,255
Tensile strength, psi	416	1,627
Modulus of elasticity, psi	3.5×10^6	6.3×10^6
Modulus of rupture, psi	739	2,637
Flexural modulus of elasticity, psi	4.3×10^6	6.2×10^6
Coefficient of expansion, in./in.-°F	4.02×10^{-6}	5.36×10^{-6}
Thermal conductivity at 73°F (23°C), Btu/ft-hr-°F	1.332	1.306
Water permeability, ft/yr	6.2×10^{-4}	0
Water absorption, %	5.3	0.29
Freeze-thaw durability		
Number of cycles	590	2,420
% wt loss	26.5	0.5
Hardness-impact ("L" hammer)	32.0	55.3
Corrosion by 15% HCl (84-day exposure), % wt loss	10.4	3.6
Corrosion by sulfates (300-day exposure), % expansion	0.144	0
Corrosion by distilled water	severe attack	no attack

PORTABLE RADIOISOTOPE X-RAY ANALYZERS---TECHNIQUES AND APPLICATIONS (2)

J. R. Rhodes*

X-ray fluorescence analysis is a sensitive and widely applied technique that until recently has been thought of as a purely laboratory tool. However, in the last few years, small radioisotope sealed sources have been developed that can replace the X-ray tube and its high voltage power supply. Also, simplified methods of X-ray energy selection, based on X-ray filters, have been found adequate to isolate the characteristic X-rays of the element to be determined. As a result, it has been practicable to develop portable, lightweight, inexpensive X-ray fluorescence analyzers for use in field and industrial conditions.

Such instruments have been available since 1965 and manufactured in the U.S.A. since about 1968. Their main uses have been in mining for rapid assay of ore samples in the field, in field laboratories, and even on board ships. They have also found use in industry for alloy analysis in scrap sorting and identification of alloy types, and for checking the concentration of electroplating bath solutions. In these applications the advantages of rapid, in-situ measurements are significant. Decisions that would otherwise have awaited a laboratory report can be made on the spot; the number of samples that have to be sent back to the laboratory can be reduced by an order of magnitude; shipments of large quantities of the wrong alloy can be avoided and so on.

The portable analyzers at present on the market are general-purpose instruments priced in the range \$3000 to \$5000. This is only a small fraction of the price of conventional laboratory equipment.

X-ray analysis is thus available to people who have never before considered using it.

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As these people become more and more aware of the existence of such instruments, a wide range of new applications should be discovered (some of which ought to be large enough to justify development of special-purpose portable analyzers in an even lower price range).

Description of the Instrument (3)

The basic arrangement of probe and electronic unit has been described. The probe is a thin sodium iodide crystal and includes a source and shield, filters, and sample holder. The arrangement of source, sample and detector is compact giving a very high efficiency of X-ray collection. An important characteristic of this geometry is the existence of a count rate distance plateau, such that if the sample is at a certain distance changes in that distance hardly affect the count rate. Surfaces with a roughness of up to about 1/16" can be measured without significant geometrical errors.

Applications (3)

Applications to measurement of coating thickness and to elemental analysis have been / described previously.² These included determination of calcium in cement raw mix, vanadium, molybdenum and chromium in steel, and tin and lead in copper alloys. Table 1 shows the main features of many of the commercially available portable x-ray analyzers.

Four of the instruments use 2" dia. sodium iodide detectors and three of these use a scaler output in preference to the ratemeter. The usual sample diameter measured is 1-1/2 to 2". Columbia Scientific Research Institute
The one exception to this is the / probe which can measure significantly smaller samples.

1. Portable Radioisotope X-Ray Fluorescence Analyzer, Isotop. Radiat. Technol. 5(4): 280-4 (Summer 1968).
2. J. R. Rhodes, X-Ray Analysis Using Radioisotope Sources, USAEC Report ORO-3224-14, Texas Nuclear Corp., 1968.

Table 1. RELEVANT PROPERTIES OF SOME COMMERCIALY AVAILABLE

PORTABLE X-RAY ANALYZERS

MAKER	PROBE	FILTER PAIRS	S C A	OUTPUT	BATTERY LIFE (HRS)	WEIGHT (lb)		SAMPLE DIAM.
						PROBE	READ-OUT	
HILGER & WATTS	NaI (TI) 2"	1	NO	R/M	100	3 1/2	15	1 1/2 TO 2"
EXCO ELECTRONICS	NaI (TI) 2"	1	YES	SCALER (10 ⁶)	10	3 1/2	14	1 1/2 TO 2"
NUCLEAR ENTERPRISES	NaI (TI) 2"	1	YES	SCALER OR R/M	10	3 1/2	10	1 1/2 TO 2"
NUCLEAR CHICAGO	NaI (TI) 2"	1	YES	SCALER (6.4 X 10 ⁵)	50	3	12	1 1/2 TO 2"
COLUMBIA SCIENTIFIC INDUSTRIES	NaI (TI) 1 1/2"	1	YES	SCALER (10 ⁶)	50	2	10	3/4 TO 1 1/2"
PANAMETRICS	PROP. OR SCINT.	1	YES	SCALER (6.4 X 10 ⁵)	50	4	14	1 1/2 TO 2"
TELSEC	TWIN PROP COUNTER	6	NO	R/M DIFFERENTIAL	20	20	13	1 1/2 TO 2"

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EXPERIMENTAL SELF-LUMINOUS OVERHEAD TYPE TRAFFIC SIGN (2)

Harry H. Dooley* and Jack Snyder**

The applications of self-luminous products in outdoor informational and directional signs have been a subject of interest to traffic engineers for many years. Engineers for the Bureau of Public Roads recognized certain advantages of self-luminous overhead traffic signs over ten years ago. However, prior to the availability of enriched ^{85}Kr , little use had been made of radioisotope-excited light sources in highway sign applications---primarily because of brightness limitations of the then available self-luminous sources.

Advantages of Self-luminous Sources (3)

The use of self-luminous sources offers many advantages over ordinary light sources: (1) A complete independence of electrical power makes them worthy of consideration for use in locations where electricity is unavailable or costly to install; (2) Since they do not require electricity, they are not subject to power failures or interruptions; (3) They do not "burn out," so that maintenance is virtually eliminated; (4) There is no need for replacement of bulbs, batteries, cables or other accessories; (5) Because they need no wires or connections to secondary sources, signs containing self-luminous sources are simple to install.

 ^{85}Kr Light Sources (3)

American Atomic Corporation (AAC) undertook in 1963, and has continued, a company-sponsored research program to develop ^{85}Kr self-luminous sources, known as I-LITES, which would have sufficient brightness for use in highway traffic signs. Since ^{85}Kr decreases in intensity 50% every 10.6 years, self-luminous sources containing this radioisotope should have a long enough useful life to be of interest, provided the initial brightness level was sufficiently high in the optimum range.

Luminance data obtained over a four-year period showed that a near-linear

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** Arizona Highway Dept.,

relationship exists between the brightness of the I-LITE and the ^{85}Kr content of the source. To construct light sources in the desired brightness range of 10-20 foot-lamberts, however, it was necessary to obtain ^{85}Kr gas which had been "enriched" or increased in specific activity to a level at least 5-6 times that of the byproduct gas available from the uranium fission process.

The Division of Isotopes Development, USAEC, has been extremely interested in seeking uses for this byproduct and, in 1968, through the pilot-plant enrichment facility at the Isotopes Development Center, Oak Ridge National Laboratory, Tennessee, provided the Arizona Highway Department (AHD) and AAC sufficiently enriched ^{85}Kr for fabrication of an experimental self-luminous overhead traffic sign.

Design---Basic Concepts (3)

Safety is the primary factor affecting design, fabrication, installation, and use of such a device. Any product of this type is subject to regulatory authority--- in this instance, the Arizona Atomic Energy Commission, which has licensed the AHD for use of the sign at the specified location.

The I-LITE source whose components were selected on the basis of offering maximum integrity was designed to withstand severe environmental conditions. The stability of this configuration is supported by over 4 years of usage in a variety of applications. The phosphor used for this experimental sign has a spectral emission curve which is similar to the sensitivity curve for the eye of an average observer.

The legibility and brightness factors are now based largely on Michigan State Highway Report No. R581,¹ which indicates that: (1) legibility decreases with increasing brightness so that 10-20 foot-lamberts is optimum; (2) contrast is important and is an optimum at 10/1; (3) legibility under high contrast and low background is not reduced drastically, even at brightnesses as low as 1.0 foot-lambert; and (4) various letters have differing legibility requirements. They note further that white or light letters on a dark background are somewhat better than dark letters on a white background.

Although Allen's report refers to photometric measurements made on electrically illuminated signs, extrapolation to self-luminous performance appears very reasonable, particularly in that their sign simulator was very much like that being considered here. Further, their brightness measurements were made using the same type of instrument (Pritchard Photometer) used in our laboratory.

A point of departure between these referenced tests and the experimental self-luminous sign is in the display arrangement. For several reasons, AAC has used a modular approach to illumination of letters and signals as opposed to lighting the entire stroke width and height:

1. It minimizes the amount of radioisotope and, hence, minimizes costs while not significantly affecting legibility.
2. It offers resistance to accidental or mischievous breakage of the light modules.
3. It provides economies in construction, assembly and in replacement.

Previous visual tests at AAC have shown that the dark spacings between the modular lights do not affect legibility in the range of 100 to 1,000 feet, since the human eye will "group" closely spaced lights. Experiments on spacing requirements for this intended purpose have shown that the merging effect is retained up to about 3 inches separation for 1-in.-diameter sources. Moreover, a psychological effect due to vision experience will define groupings when the separation becomes a possible problem.

Construction (3)

The direction sign is 6 feet high, 5 feet wide and has an overall weight of 750 pounds. It is attached to a supporting pole capable of sustaining a threefold increase in weight, and faces in a westerly direction. The bottom of the sign is 17-1/2 feet from the ground. The word "ONLY" has a height of 10 inches with letter strokes of 2 inches. The arrow is approximately 34-1/2 inches high with a 6-inch stroke. The 66 I-LITES are 1 inch in visual diameter and each contains

1 curie of enriched ^{85}Kr . Each light is located approximately 5 inches from the face of the sign and is doubly encapsulated within a metal cylindrical shell which contains a light pipe. Further internal protection is afforded through the use of metal shielding around each light, thus increasing the resistance to destructive acts or accidents.

Performance (3)

On July 8, 1969, the first functional, self-luminous, overhead traffic-control sign in the United States was installed in Phoenix, Arizona by the AHD. The nominal brightness level was 10 foot-lamberts. The radiation profile measurements made at that time at ground level in the immediate vicinity of the sign were slightly above background. Nighttime visibility under ambient lighting conditions of the area is in excess of 500 feet (Fig. 1).

The sign is observed monthly from ground level by AAC personnel, with periodic checks scheduled at the surface of the sign. In December, 1969, such an inspection of the sign surface was made. All I-LITES were functional and no major changes in structural integrity were found. Random photometric measurements on 27 of the lights, using portable equipment, showed an average luminous decrease of approximately 4% per year. This is in substantial agreement with luminous decay data obtained using I-LITES having a nominal brightness of 1 foot-lambert.

Future Usage (3)

Since the construction of the experimental sign, prototype I-LITE sources have been fabricated at brightness levels exceeding 50 foot-lamberts---i.e., 5 times those in the experimental sign. Widespread usage of this sign-illumination technique, however, depends largely upon the availability and cost of ^{85}Kr . The availability of this reactor byproduct gas for reprocessing and enrichment is increasing rapidly and is expected to reach megacurie quantities during the next ten years. This supply is expected also to reduce significantly the present price which, from an economic standpoint, will enhance markedly its

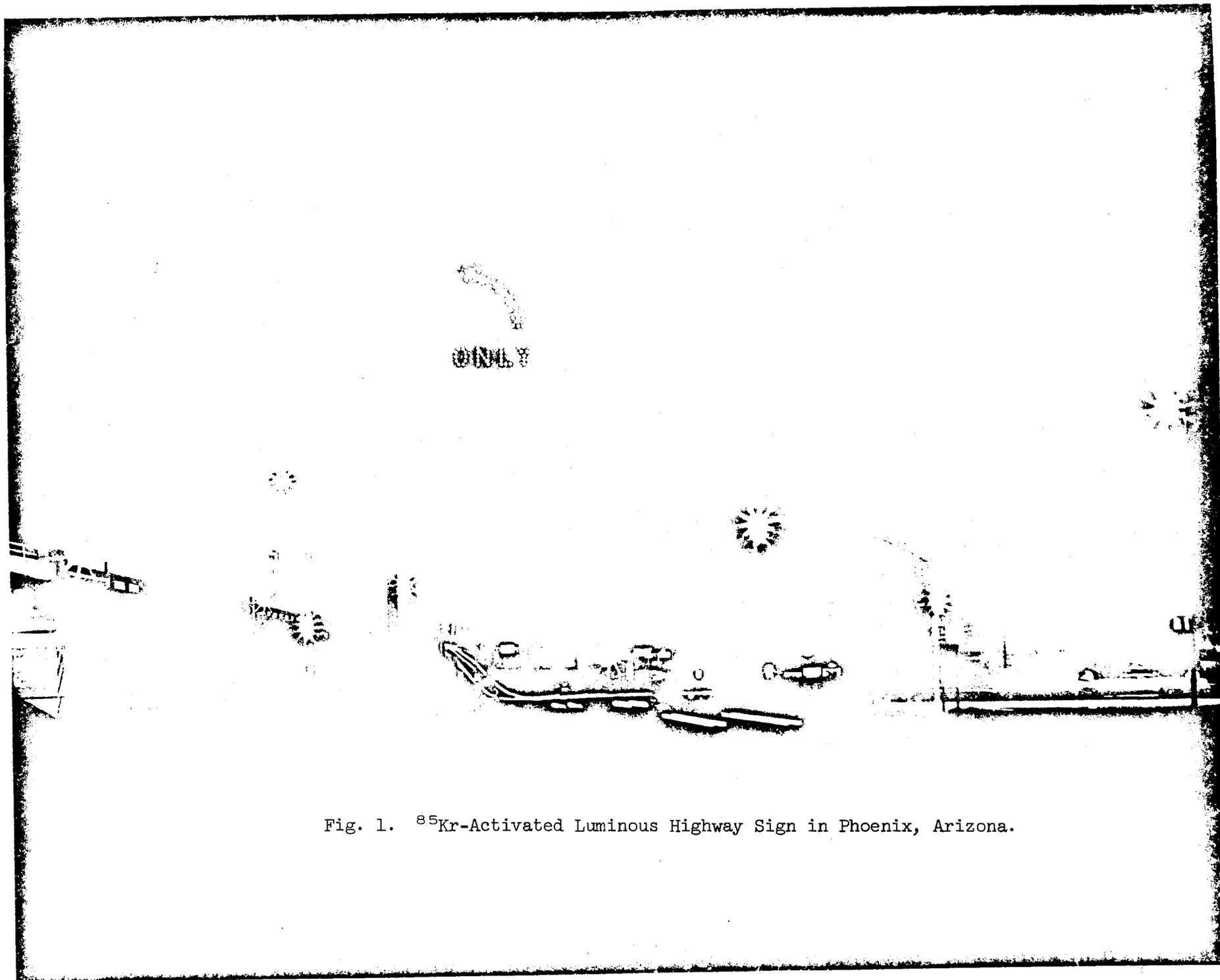


Fig. 1. ^{85}Kr -Activated Luminous Highway Sign in Phoenix, Arizona.

consideration for highway-sign illumination. In addition, the commercial utilization of ^{85}Kr in this type of application will help to resolve some of the problems which arise in connection with the ultimate disposition of the radioactive byproduct.

Through continuing evaluation of this experimental unit, data will be obtained by following the effect of ^{85}Kr on the emission spectra and brightness of various phosphors used in light sources at various environmental temperatures and the effect of the radioisotope on the brightness of the light sources as they increase in age. All of this information is needed in order for those involved in highway research, development, and application engineering programs to evaluate the prospective uses of ^{85}Kr -excited light sources in highway signs.

AUTOMOBILE EXHAUST ANALYSIS FOR LEAD USING A NUCLEAR TECHNIQUE (2)

P. L. Sciaraffa* and C. A. Ziegler**

The main source of atmospheric lead contamination is automobile exhaust. Recently, increasing interest has been shown in a convenient method of measuring the level of lead in the atmosphere. Because of this interest, an investigation was made of the feasibility of sampling the atmosphere with a Millipore filter and then using a radioisotope-excited x-ray-fluorescence analyzer to determine the amount of lead collected.

The purpose of this study was threefold: (1) to show the feasibility of collecting samples on a Millipore filter and doing elemental analysis by x-ray fluorescence, (2) to establish working calibration charts for the instrument, and (3) to determine the minimum quantity of lead detectable with the Panalyzer-4000.***

Instrument Design and Operation (3)

The instrument consists of a probe and an electronic console. The probe contains a radioactive source which is constructed so as to emit radiation only in the direction of the sample, i.e., the particles collected on the Millipore filter. Radiation striking the sample causes some of its atoms to emit x rays of an energy characteristic of the emitting element. These characteristic x rays pass through the window of an x-ray detector; the detector can resolve x-ray energies, producing as an output a voltage pulse. Voltage pulses from the detector are introduced into the electronic console where they are sorted and counted for a preset measurement time and the number of pulses is displayed digitally on the console. This number is then used in conjunction with a calibration chart to obtain the amount of the measured element in the sample. Typical measurement times are of the order of one minute.

In the design of the probe, provision has been made for the use of the difference-

* Millipore Corp., Bedford, MA.

**Panametrics, Inc., Waltham, MA.

***Manufactured by Panametrics, Inc., Waltham, MA.

filter technique to cancel out background radiation. Whether or not filters are required depends on the application and the accuracy desired. That is, the instrument is designed so that it will either count up (i.e., add), or count down (i.e., subtract). Thus, it can be used in applications requiring (a) no filter, (b) a single filter, or (c) balanced-difference filters. In either of the first two cases, it is only necessary to "count up" for a determination. In the latter case, the first of the difference filters is selected so that it transmits both the background and the desired x-ray signal, and the second so that it transmits the background preferentially. The filters are balanced so that they transmit equal background intensities.

Sample Collection (3)

The x-ray fluorescence technique is feasible where airborne lead concentrations are such that a sufficient sample can be collected in a reasonable time. Typically, in urban areas, air sampling times of 6 to 12 hours provide enough lead sample to allow measurement.

The Millipore filter acts as a two-dimensional screen; it readily removes and retains on its surface all airborne particles larger than the specified pore size. Using 47-mm, type AA filters (0.8- μ pore size) and the standard Millipore aerosol holder, samples were collected in various locations and for various lengths of time.

Analyses (3)

The samples were analyzed by x-ray fluorescence using the Panalyzer-4000. The advantages of this radioisotope method are that wet chemistry is not required and rapid results are obtained using low-cost, portable equipment. The procedure, using the previously described instrument, is to "count up" with the first filter and then to "count down" for the same length of time with the second filter, thus subtracting the background and leaving only the desired signal counted and displayed. It should be noted that because this technique is nondestructive; several elements can be determined from one sample. Also, after determination by x-ray fluorescence, the sample is available for other tests.

Since the instrument depends on x-radiation originating in the atoms of the sample, it is desirable that the particles be on a surface, as is true of a sample collected on a screen filter. Radiation from particles within the matrix of a depth filter runs the risk of not reaching the detector, and thus of not being counted.

A ^{109}Cd source excited the L-lines of lead, and a 64-second counting time was used. Both qualitative and quantitative analyses are possible. A number of elements were detected qualitatively, most notably bromine and lead. Since ethylene dibromide is added to gasoline to remove lead from automobile engines, the lead in the exhaust is expelled mostly as lead bromide, PbBr_2 .

Calibrations (3)

Calibration charts (showing counts recorded versus amount of element sought) were established by collecting samples, obtaining the fluorescence count on the instrument, and having the samples analyzed for lead by independent laboratories using atomic absorption. However, it was soon obvious that the bromine fluorescence overlapped a portion of the fluorescence due to lead. To get around the problem of the bromine, two methods were tried and both gave consistent results. The first, and simpler, method is to make the assumption that the lead/bromine ratio is constant; this assumption can be tested, for purposes of establishing a calibration chart, by chemical analysis. The second is to use the filters. This method excludes all the fluorescence due to bromine; it also excludes part of the fluorescence due to lead. The result is a count due only to lead.

Unfortunately, this technique also results in decreased sensitivity. Thus, while it is possible to detect 5 micrograms of lead without the filters on this instrument, the minimum detectable amount of lead, with the filters in place, is more like 15-20 micrograms.

MINIMIZING ERRORS IN GAMMA-RAY BACKSCATTER DENSITY GAGES:

EXISTING GAGES AND NEW DESIGN CONCEPTS (2)

F. H. McDougall and W. L. Dunn*

Gamma-ray backscatter-type soil density gages have been in existence for a decade, but have been plagued by calibration and accuracy problems. Before these devices can gain more widespread acceptance, their accuracy must be improved. A project sponsored by the Highway Research Board has been undertaken to evaluate new design concepts for improving these nuclear devices. Two prototype systems were designed to accomplish the project's objectives---one similar in design to existing commercial gages and another utilizing gamma-ray spectrometry, a more sophisticated approach.

Gage Errors (3)

A dual-gage principle involving the simultaneous solution of calibration models for the two gages was developed by Gardner and Roberts and has been used to define and evaluate three gage errors: composition, measurement of counting rate, and surface roughness. The dual-gage principle was designed to minimize composition error, which it does successfully. The other two errors can be minimized by an optimization procedure which evaluates gage performance through the use of a quality factor which, in turn, is a function of the three gage errors and the sample volume measured. The quality factor is studied as various gage design parameters are varied. Work is also continuing in search of a mathematical model which would be used to eliminate the surface-roughness error, probably the most serious of the three.

Gage Systems (3)

One of the prototype gage systems uses a Geiger-Mueller tube detector and is concerned with the variation of the following gage parameters: source energy, source collimation length, source--detector separation, and detector efficiency spectrum. Results of initial testing with the system are promising and indicate

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that the three gage errors can each be reduced to about one or less simultaneously for a proper combination of the design parameters. The best dual-gage system in the initial study uses a ^{60}Co source, a source collimation length of $5/8$ inches, and both a Harshaw G 17-6 G-M tube and an identical tube except for a 10 mg/cm^2 platinum coating on its inner wall at source—detector separations of four and ten inches, respectively.

The new design factors of the gamma-ray spectrometry prototype system permit varying source and detector collimation, angle of source and detector incidence into the soil, and energy regions being detected. These new design factors are used in conjunction with the existing basic parameters: source energy and intensity, source—detector separation, and detector type. The prototype utilizes the spectrometric capabilities of a NaI(Tl) crystal detector and a multichannel analyzer system. Mathematical modeling appears to be a promising tool for further reduction of the surface-effect error.

Results (3)

Preliminary results indicate that significant advantages are realized in combining the various gage design factors in an optimum manner. Considerable improvement over existing commercial gages is noted.

POTENTIAL INDUSTRIAL APPLICATIONS OF THE ^{57}Fe MÖSSBAUER EFFECT*

J. H. Terrell**

Supplementary keywords: industry; industry, primary metal; metallurgy, ferrous; physical property; Mössbauer effect; absorptiometry; spectrometry; review, technical; backscatter; theory, applied; comparison; atomic structure; molecular structure; crystal; differentiation; carbon; chromium; zinc; cobalt-57; iron-57.

Abstract. The application of the Mössbauer-effect technique to several industrially significant problems is discussed; iron-bearing materials are emphasized. A review is given of past results and of current efforts to conduct Mössbauer-effect measurements that use the 2% naturally abundant ^{57}Fe isotope and that do not require sample preparation, that is, backscatter rather than transmission measurements. Detection schemes in the backscatter geometry that use 6.3-keV internal-conversion x rays, 14.4-keV gamma rays, or 8-keV internal-conversion electrons are compared. Potential applications of the backscatter technique to metallurgical problems are summarized; they include studies of types, thermomechanical history, magnetic properties, corrosion products, surface-hardening processes, and metallic coatings. Preliminary results obtained with the backscatter method on a steel coupon in axial tension are included. Other industrial applications that are accomplished best with transmission geometry are also summarized.

(Introduction)

Since the discovery of the Mössbauer effect in 1957, it has been possible to probe the microstructure of materials in new and meaningful ways. The complex instrumentation and the specialized sample preparation needed have made Mössbauer-effect measurement almost exclusively a laboratory technique. Minimal attention

* Work supported jointly by Naval Air Systems Command and Sanders Associates, Inc.
Formerly

**Mithras Division, Sanders Associates, Inc., Cambridge, Mass.; present address: Keystone Computer Associates, Inc., Belmont, Mass.

has been paid to the possibility of making practical use of the effect for non-destructive testing and other applications.

In the earth's composition, iron is one of the most abundant elements. It constitutes ~5 wt % of the continental crust and ~7 wt % of the entire crust. Furthermore, it is so widely distributed that few natural solid materials are without it. Among the principal elements of the earth's crust, iron is exceptional in that it commonly exists in two valence forms and passes relatively easily from one to the other as the result of oxidation or reduction. The ^{57}Fe isotope is 2% naturally abundant. It appears that enough ^{57}Fe Mössbauer-effect data have been accumulated to permit serious consideration of its potential industrial applications. The discussion that follows is based on work^{done} at Sanders Associates and at other laboratories. Some of the results are preliminary and exploratory.

Principle of the Mössbauer Effect (3)

The Mössbauer effect is the recoilless emission and reabsorption of a low-energy gamma ray by a nucleus embedded in a solid. Figure 1 depicts a gamma-emitting nucleus embedded in an atom that is in free space. On an energy scale, the energy of the gamma ray as it leaves the nucleus is designated by E_0 , and its energy after it leaves the nucleus is denoted ^{by} $E_0 - R$, R being the recoil energy of the nucleus. Thus, the energy of the gamma after it leaves a nucleus that can recoil is diminished by the imprecisely defined amount R , as shown in Fig. 1.

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later.

When a gamma ray is emitted from a nucleus embedded in a crystalline solid, E_0 is well defined, because emission occurs without recoil and R is therefore negligible.

The principle of resonant absorption is illustrated at the bottom of Fig. 1 by the superimposed emission and absorption lines. The equation indicates that R can be made very, very small if the mass of the solid, M , is large enough. This result follows from the conservation of energy and momentum. Thus, it can be considered that R is reduced by an amount inversely proportional to M . This recoilless emission and reabsorption of a low-energy gamma ray by a nucleus in a solid is the

Mössbauer effect. The last equation on Fig. 1 implies that E_0 may be varied by Doppler-shifting either the source or the absorber at relative velocity, V , with respect to the other; the magnitude of V is of the order of millimeters per second.

The hardness, dimensional stability, strain aging, and stress-corrosion cracking of iron alloys have been determined by observing the metallurgical phase changes involved. X-ray and electron diffraction, resistometry, and internal-friction techniques have traditionally been used to follow the phase changes. Transmission Mössbauer-effect measurements have recently been used to study some of these properties, but, as in the x-ray- and electron-diffraction methods, sample preparation is needed to obtain useful results. However, in the backscatter Mössbauer method, sample preparation is not necessary. The specimen need not be "touched" except by the Mössbauer gamma rays. Therefore, backscatter measurements, as compared with transmission measurements, give a more realistic picture of metallurgical processes that occur in and near the surface of iron alloys, inasmuch as sample preparation can introduce unwanted---and sometimes unknown---effects.

Instrumentation (3)

A block diagram of the instrumentation for transmission Mössbauer spectrometry is given in Fig. 2. The radioactive source is driven back and forth by a velocity transducer; the gamma rays from the source pass through a thin sample and impinge on a detector. It is convenient to display the number of counts detected versus the velocity of the transducer as an X-Y plot; a multichannel scaler performs this function. The top spectrum contains a single inverted peak, which is indicative of ^{57}Fe nuclei that are ^{present} / in cubic environments. The middle spectrum shows two inverted peaks, which are characteristic of ^{57}Fe nuclei _{that are} present in a noncubic environment. The bottom spectrum shows six inverted peaks, which indicate that the ^{57}Fe is in a magnetic state. The spectrums are called "fingerprints," because they indicate unique features about the physical state of the iron embedded in the solid. In addition, each spectrum gives information on the chemical state of the iron, which is measured by the shift of the center of gravity of the spectrum away from

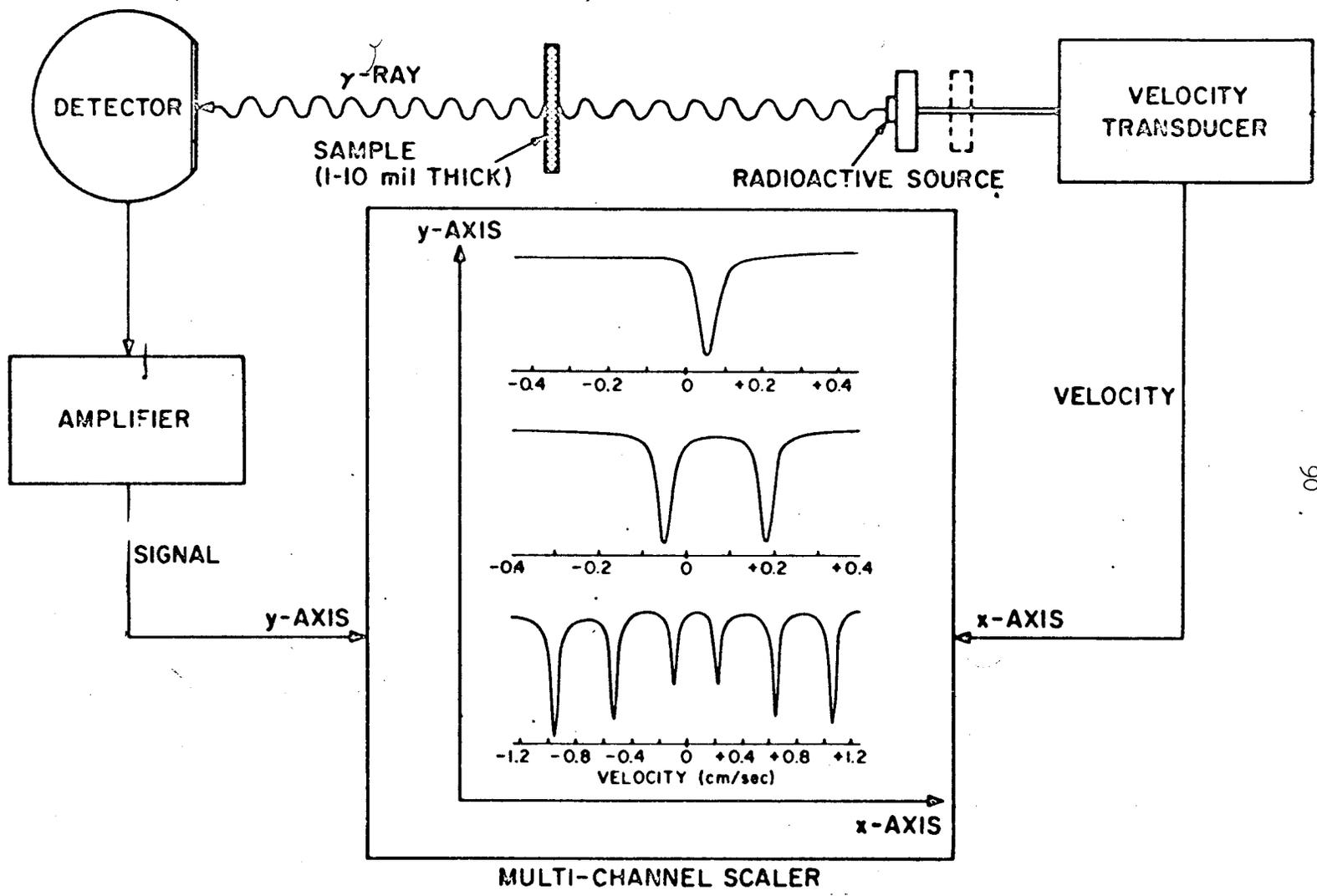


Fig. 2. Block diagram of instrumentation for transmission Mössbauer spectrometry.

the 0-velocity point. Note that the transmission geometry requires thin samples (1 to 10 mils thick).

Figure 3 shows the ^{configuration} of a backscatter (reflection) Mössbauer spectrometer. In the backscatter Mössbauer spectrometer, the reflected 6-keV internal-conversion x rays, 14-keV gamma rays, or 8-keV internal-conversion electrons are counted as velocity of an emitter (⁵⁷Co) relative to that of an absorber is varied. The Mössbauer spectrometer is a "spectrometer," because the precise energy of emitted or absorbed radiation depends on the chemical environment, local electric-field gradient, and local magnetic field at the emitting or absorbing nucleus. The velocity of the emitted relative to that of ^{the} / absorbed serves to bring them into coincidence via a Doppler shift, a necessary task when emitter and absorber are not identical. Thin samples, which require special preparation, are not needed, and the backscatter produced by gamma rays impinging on a semi-infinitely thick solid are detected as indicated. For naturally abundant ⁵⁷Fe, it is convenient to count the internal-conversion 6-keV x rays that are emitted following Mössbauer-effect events in the solid. Again, the display as an X-Y plot is useful. The six-peak fingerprint spectrum obtained by use of the backscatter Mössbauer spectrometer indicates that the material is magnetic; the distance between the outermost peaks shows that it is a ferritic steel. In the backscatter spectrum the peaks are not inverted as in the transmission spectrum.

Transmission Measurements (3)

In certain applications of the Mössbauer effect, the backscatter technique has no advantage over the transmission technique. Table 1 gives some of these applications.

Typical transmission Mössbauer-effect spectrums (Fig. 4) illustrate that various phases of steel give characteristic peaks. The data from cast iron (Fig. 5) show how cementite (Fe_3C) is readily distinguished from ferrite (under separated six-peak spectrum).

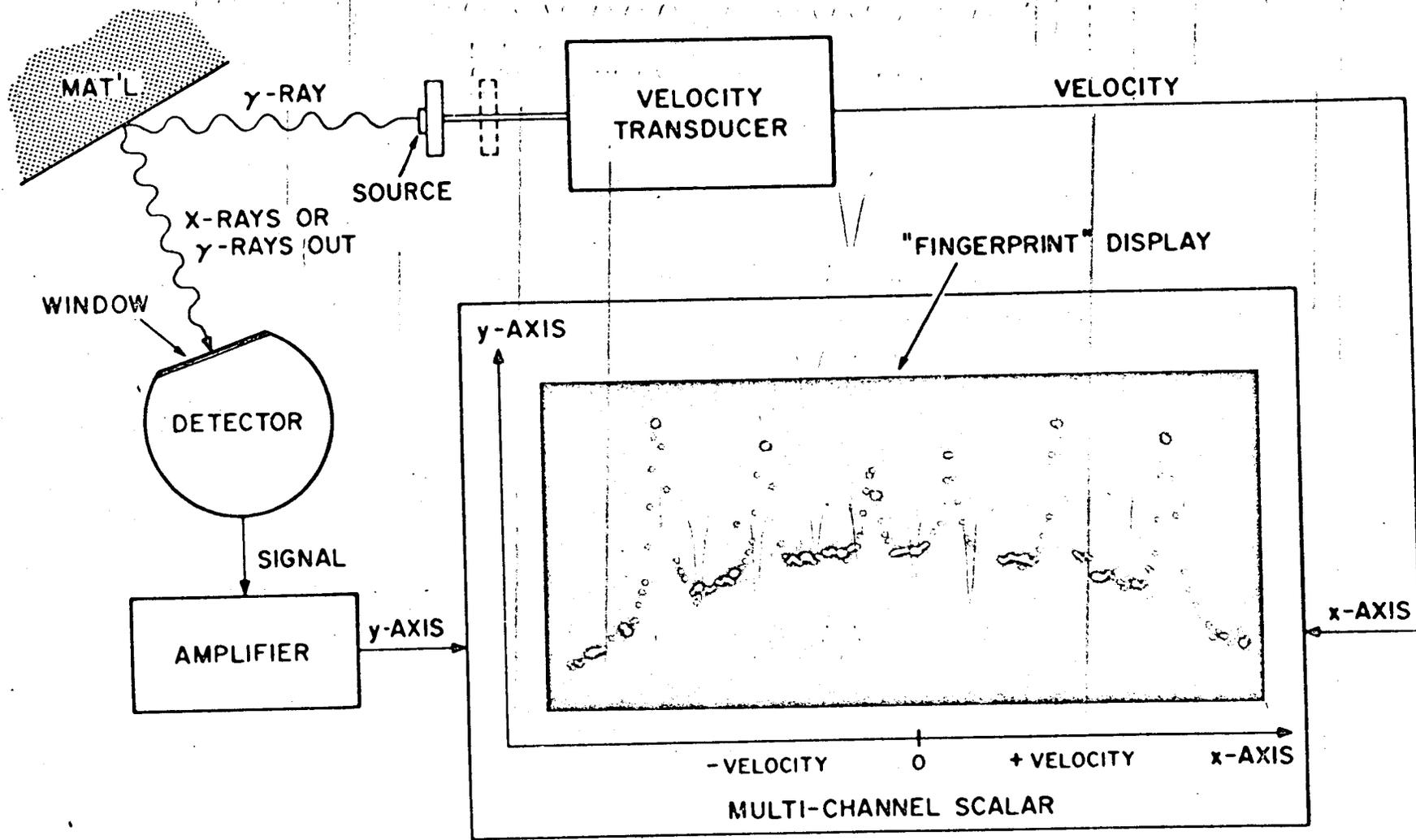


Fig. 3. Block diagram of instrumentation for backscatter (reflection) Mössbauer spectrometry.

Table 1. APPLICATIONS OF TRANSMISSION MÖSSBAUER-EFFECT MEASUREMENTS
(BACKSCATTER MEASUREMENTS HAVE NO ADVANTAGE)

Measurement	Application
Iron in cement	Monitoring the reaction that goes to completion after the mixing of cement and water, i.e., hydration of iron-containing clinker components. Relating the Fe^{2+} : Fe^{3+} ratio to color of white and buff cements.
Iron in coal	Characterizing nonmineral iron, e.g., the ionic state of the organic iron. (Identification of minerals present usually does not account quantitatively for existence of elements such as Na, Mg, Al, Si, K, Ca, Ti, and Fe. In this work, a hitherto-unknown Mössbauer spectrum was found.)
Iron in slag	Identifying forms of iron present in slag from engine pipes. (Amorphous nature of material makes x-ray-diffraction measurements inaccurate.)
Vibration	In biomechanics, a small ^{57}Co source was attached to an ear tympanum, and the velocity of the tympanum was determined, since the frequency remained fixed. Amplitudes of several angstroms were measured with an error of 1 to 2 Å.

Backscatter Measurements (3)

Some applications in which backscatter Mössbauer-effect measurements are considered to be potentially suitable are given in Table 2. Discussions of specific applications follow.

The suitability of the Mössbauer-effect method for measuring the volume fraction of retained austenite in steel specimens has been demonstrated by Chow and Bogner;¹ for this measurement, the backscatter technique is accurate and requires no sample preparation.

The backscatter spectrum of tool steel (Fig. 6) is characterized by the structure in the central portion of the spectrum. With increase in the chromium content

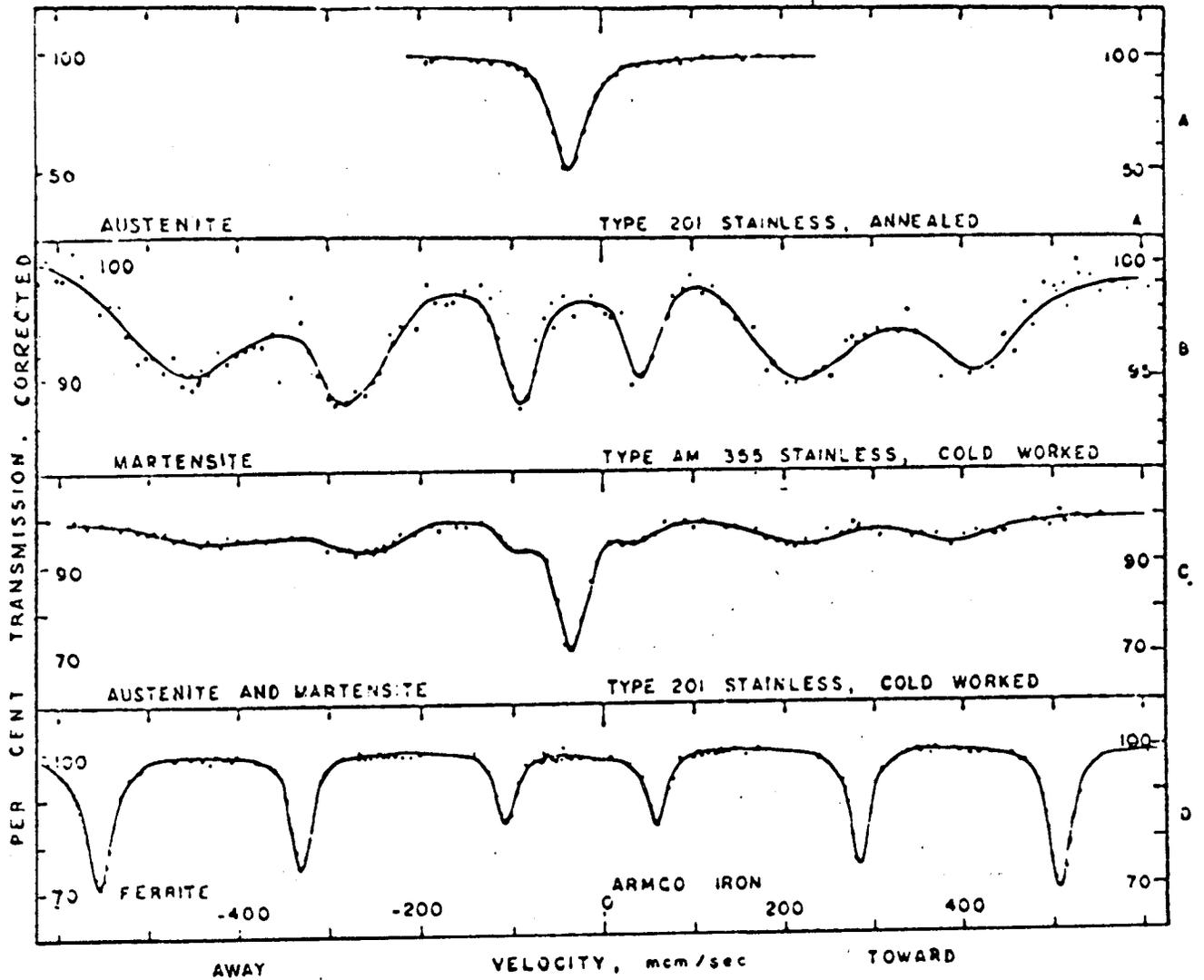


Fig. 4. Transmission Mössbauer-effect spectrums of various types of stainless steel and iron.

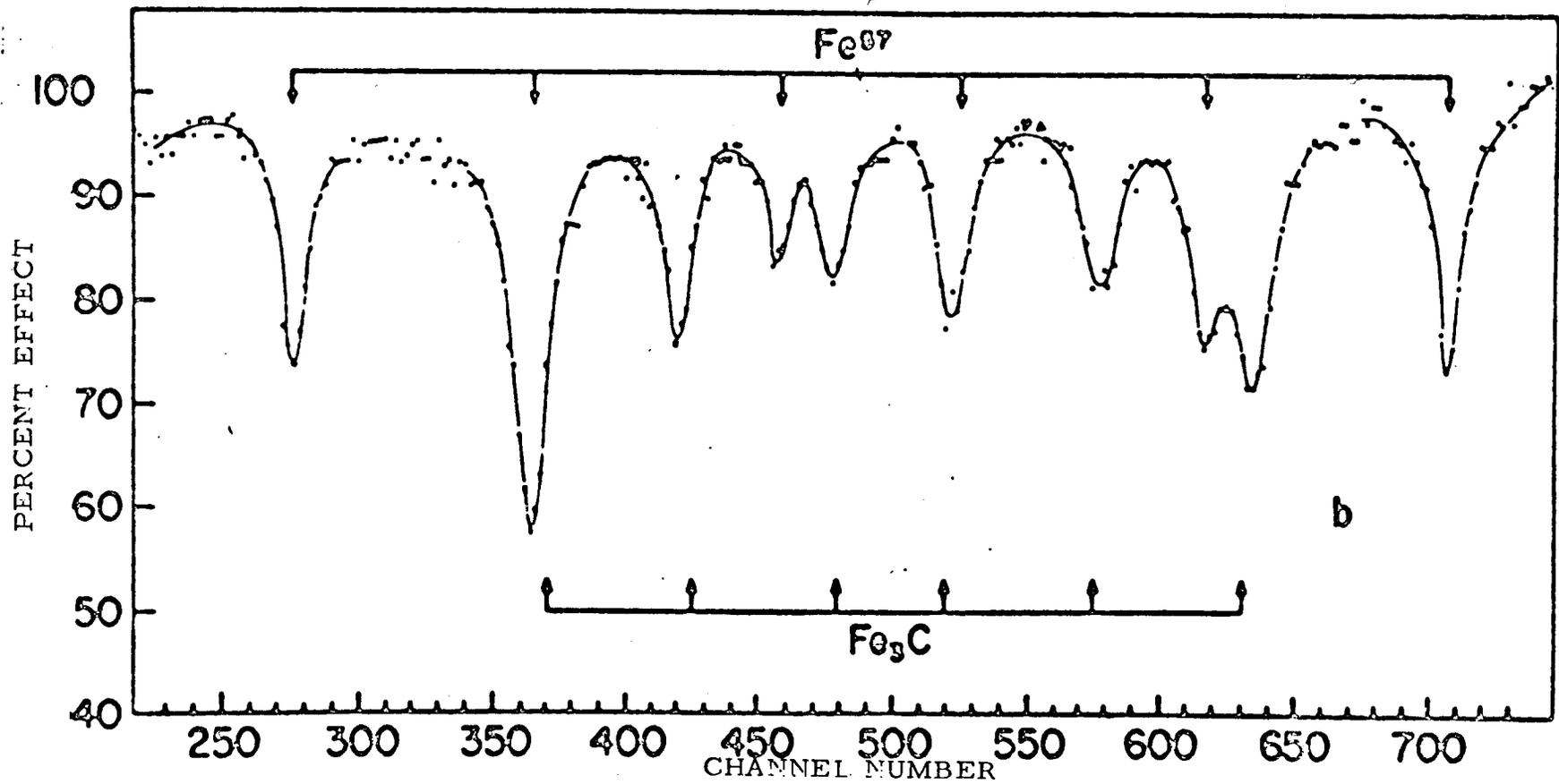


Fig. 5. Mössbauer-effect data from cast iron.

Table 2. APPLICABILITY OF BACKSCATTER MÖSSBAUER-EFFECT TECHNIQUE: WITH COMPARISON Δ OTHER TECHNIQUES

Application	Backscatter Mössbauer-Effect Technique	X-ray Diffraction	Radioisotope X-ray Fluorescence and X-ray Spectrography	Magnetic Technique (e.g., Saturation Magnetization)
Stained austenite	Yes. Presence of Fe ₃ C can ⁶ give error. No sample preparation needed.	Possible but only with special sample preparation. Error same as for Mössbauer effect	Not possible	Gross effect and not quantitative
Carbon content in martensite and austenite	Yes. Error not exactly ^{11, 12} known. No sample preparation needed	Yes. Sample preparation needed, since texture can affect results	Not possible	Yes. Not accurate
Identification of steel types (ferrite, martensite, austenite, cementite)	Yes. No sample ^{7, 8, 9, 14} preparation needed	Yes, but preferred orientation and broad lines make sample preparation necessary	Not possible	Questionable
Precipitation due to aging and heat treatment	Yes. No sample ^{4, 5, 10, 13} preparation needed	Not possible in martensite with high carbon content; possible in austenite but sample preparation required	Not possible	Not possible
Quality control of nonferrous coatings on steel	Yes. Down to depths of ¹⁵ 2-mil-bonding properties	Not possible	Could be used to give coating depth but no structural information	Not possible
Dry chemical analysis (e.g., ores, corrosion products on steel)	Yes	Not possible except with powder patterns	Specific elements monitored but no identification of chemical compounds possible	No
Detection of relative motion, velocity, and frequency of vibrations	Yes. Amplitudes of 10 ⁶ Å have been measured ²	No	No	No
Monitor ductile-brittle transition in stainless steels	In research stage; ³ looks promising	No	No	No
Marking of parts with ⁵⁷ Fe isotope for lation studies or monitor corrosion formation	Yes, unique	No	Possible if a radioactive isotope with useful half-life is used	No

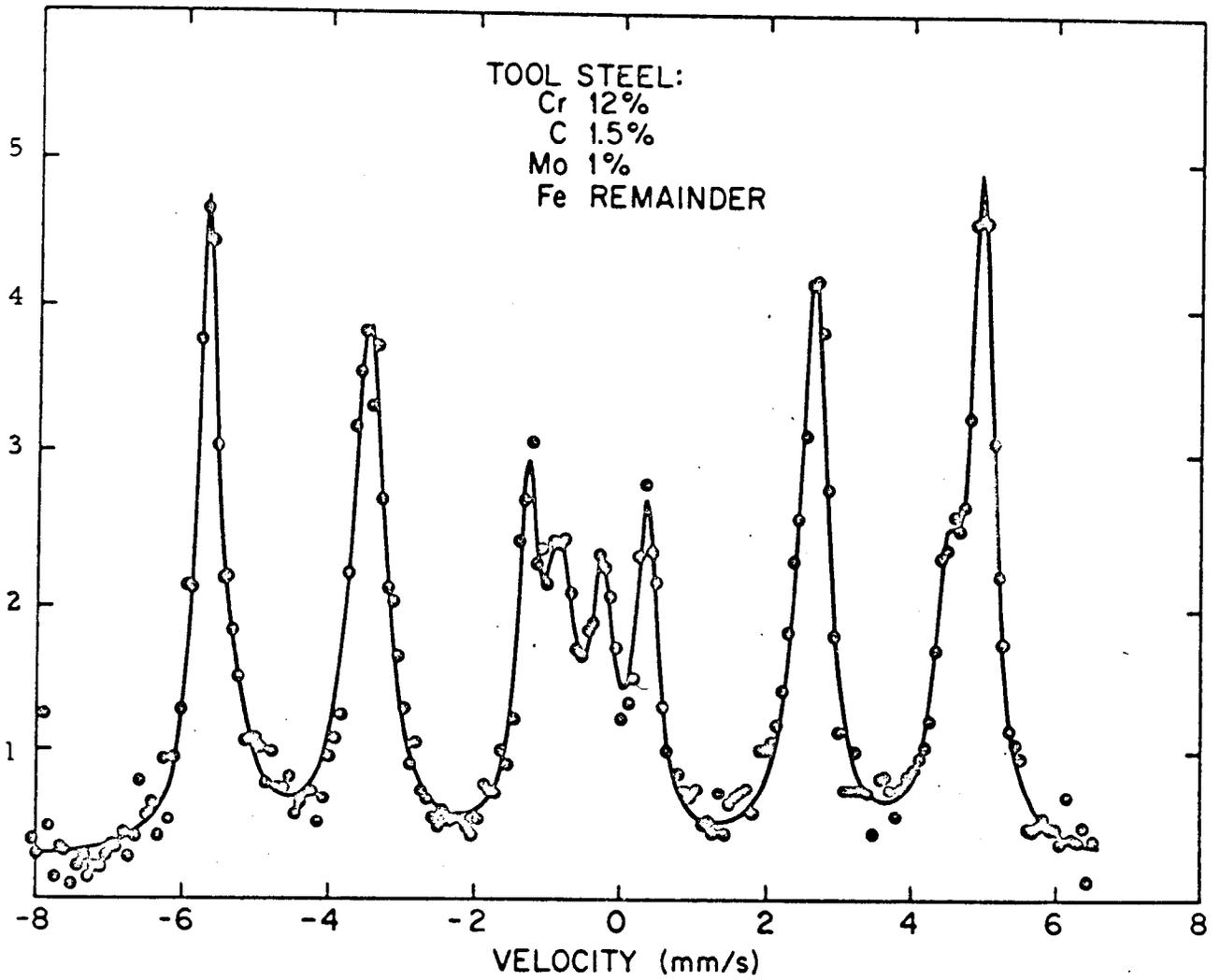


Fig. 6. Backscatter Mössbauer spectrum of tool steel (Fe, 85.5%; Cr, 12%; C, 1.5%; Mo, 1%).

of such a steel, the velocity shift of the centroid of the spectrum (isomer shift) varies (Fig. 7). In Fe-Cr alloys, the maximum of the isomer shift occurs at about the same composition as the alloy softening. The separation between the outermost peaks (hyperfine field) in the six-peak spectrum of a martensite steel varies with carbon content (Fig. 8); the solid line is the x-ray-diffraction result. Carbon concentrations <0.5 wt % approach the limit of resolution.

Figure 9 shows the backscatter Mössbauer-effect spectrum of as-machined 52100 bearing steel and also the spectrum of the steel after it was given a normal heat treatment. The appearance of the central peak for the heat-treated sample permits measurement of the effect of the heat treatment. This peak is characteristic of many heat-treated martensite steels and can be related to their magnetic backscatter spectrum of a Simonds-81 hysteresis sample (Fig. 10) properties acquired as a result of the heat treatment. The / also illustrates the heat-treatment effect. The amplitude of the additional peak in the center of the spectrum is related directly to the squareness of the hysteresis loop of this material. Thus, the amplitude of the peak can indicate the mechanical integrity and the magnetic quality of steels.

The backscatter Mössbauer effect can be used to detect compound formation on a steel surface without sample preparation (Fig. 11). The spectrum at the top of Fig. 11 is of a rusted steel plate. After the rust was removed in two successive steps, the next two spectrums were obtained. The double-peaked pattern was identified as being typical of β -ferrioxyhydroxide, whose Mössbauer-effect parameters are given in Table 3. These parameters of iron oxides and iron oxyhydroxides can be described uniquely by the values of their chemical (or isomer) shifts, quadrupole splitting (spectrum between double peaks), and internal magnetic or hyperfine field (i.e., if there is a six-peak pattern). The measurement of values for these parameters proved that the "rust" actually was β -ferrioxyhydroxide. Such data can be used in the same way that ^{data from} ASTM cards are used to analyze x-ray-diffraction patterns of powders.

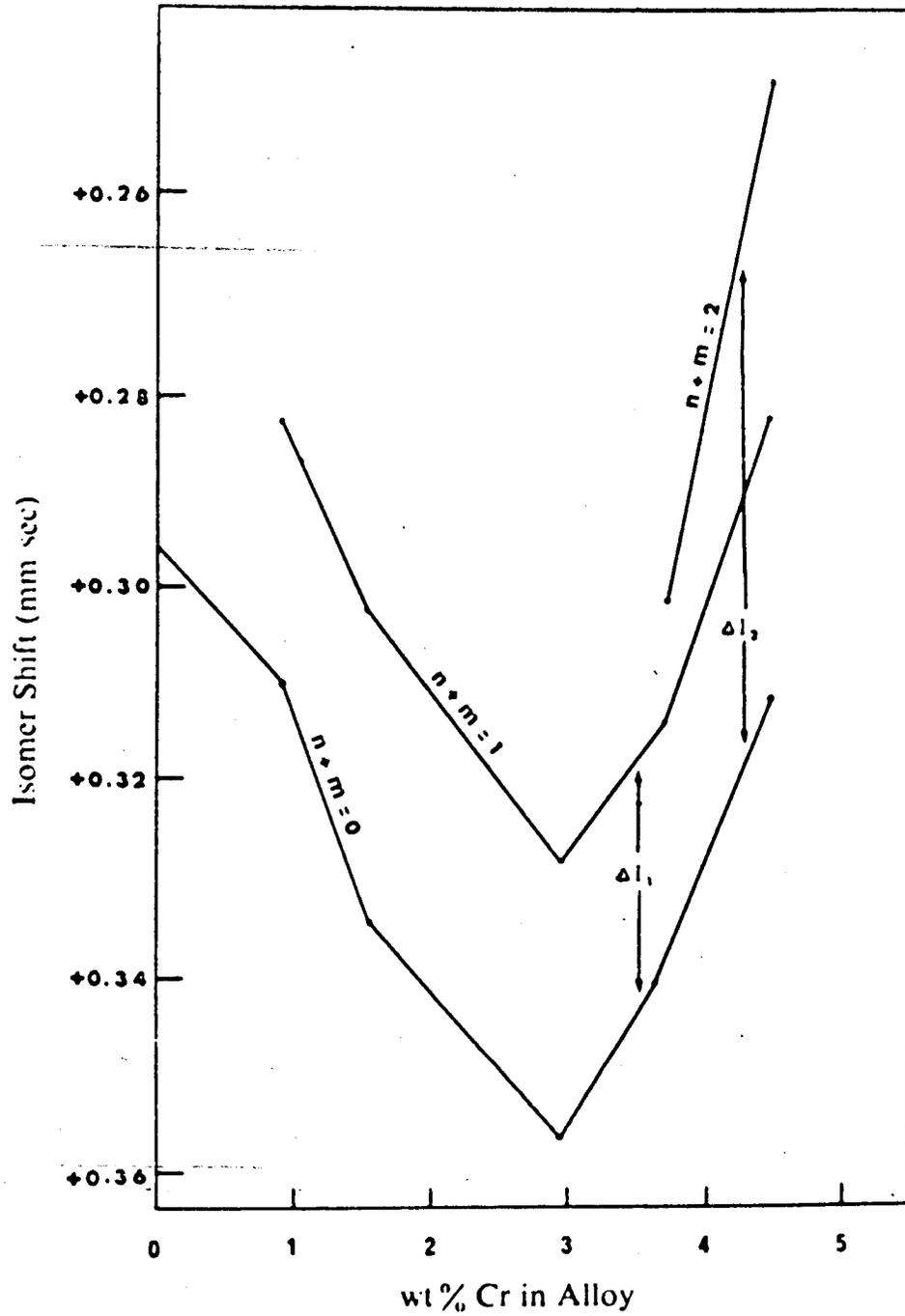


Fig. 7. Isomer shift observed in Fe-Cr alloys as a function of composition and number of chromium neighbors (data are plotted relative to the centroid of absorption spectrum of sodium nitroprusside at 300°K). the

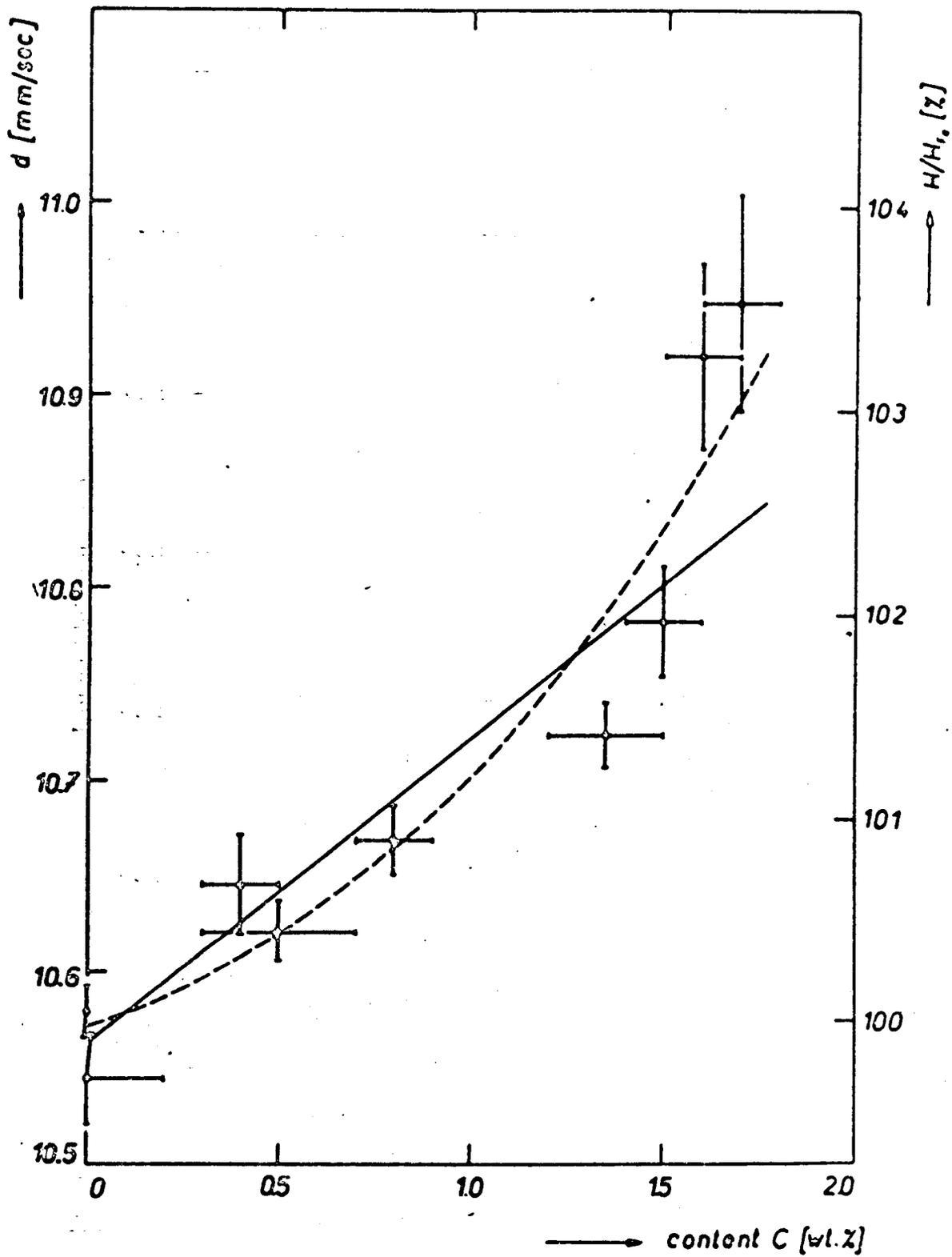


Fig. 8. Hyperfine field in martensite versus carbon content.

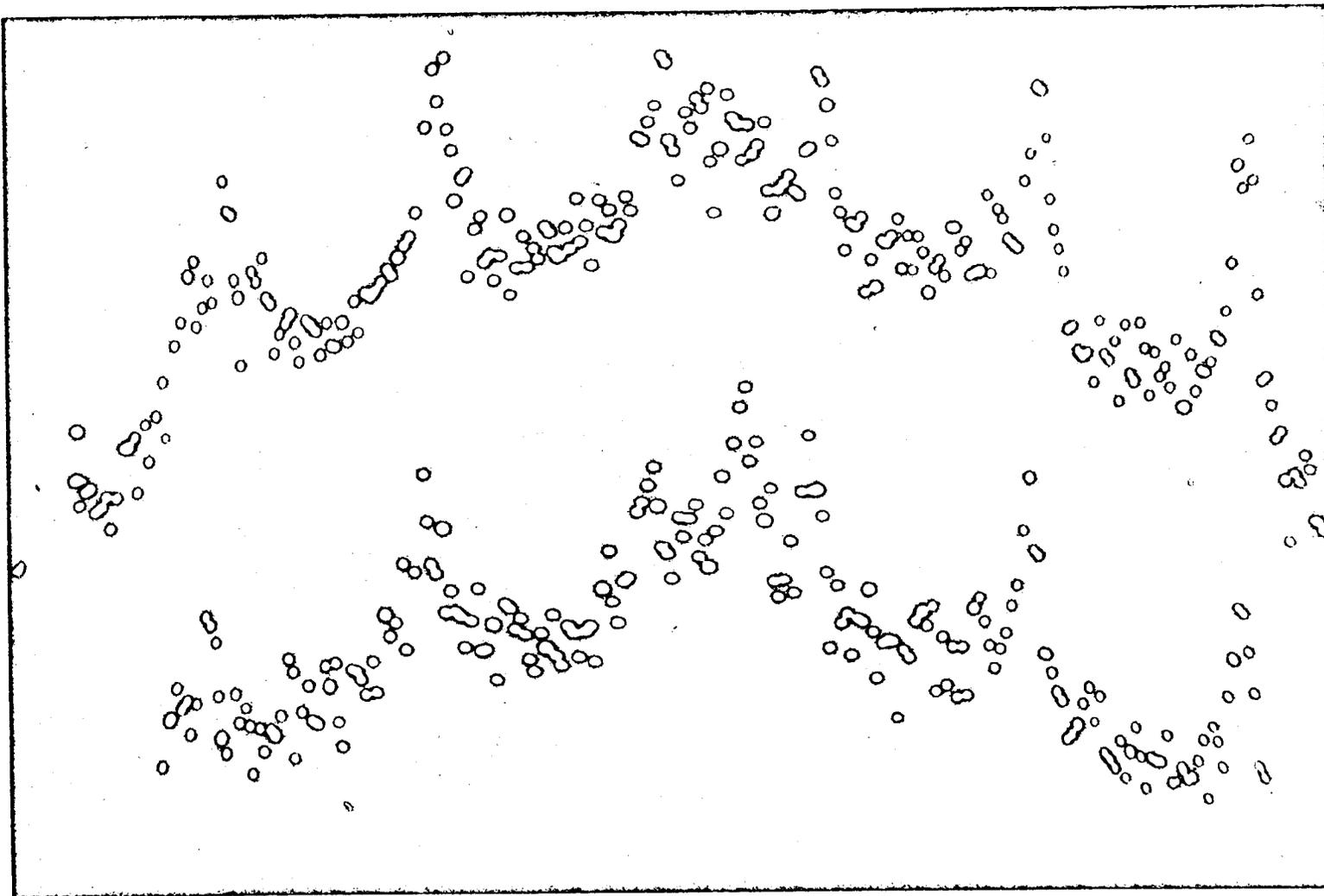


Fig. 9. "Inner-race" of 52100 bearing steel as-machined (upper) and after normal heat treatment (lower).

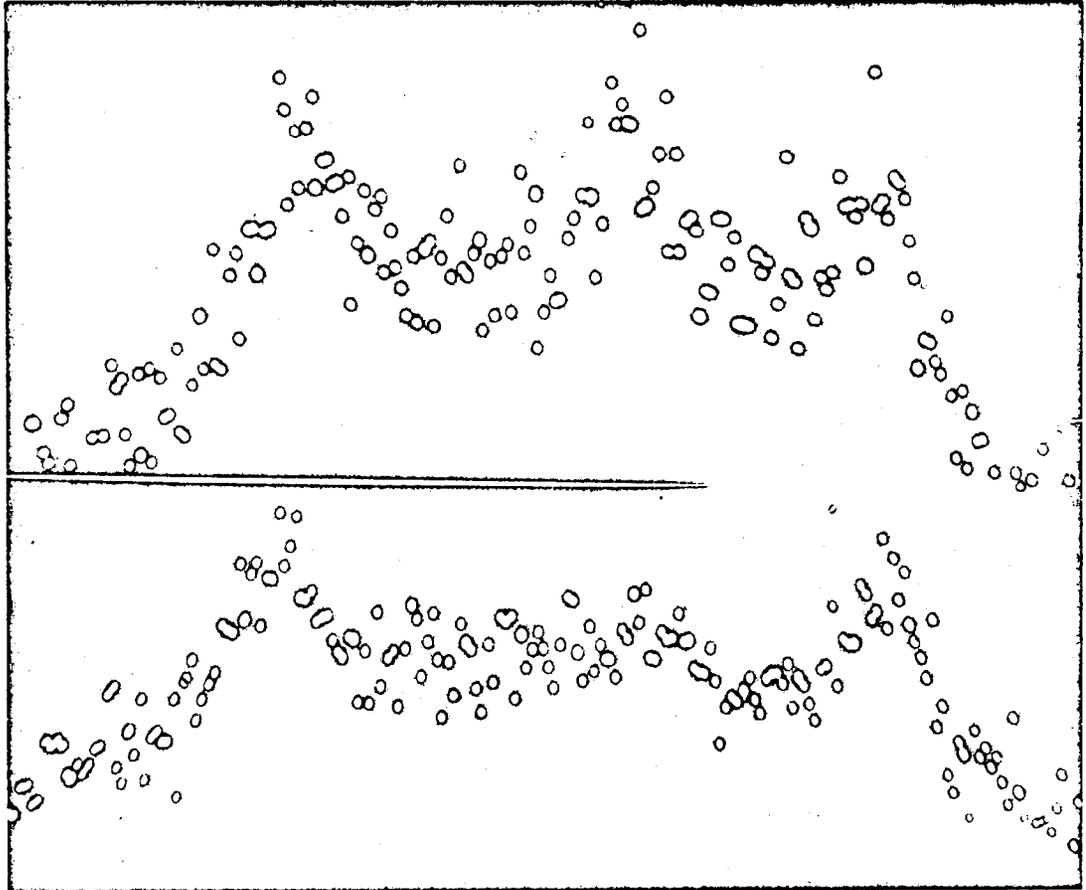


Fig. 10. Central portion of backscatter Mössbauer spectrums of Simonds-81 hysteresis material as-received (lower) and after heat treatment (upper).

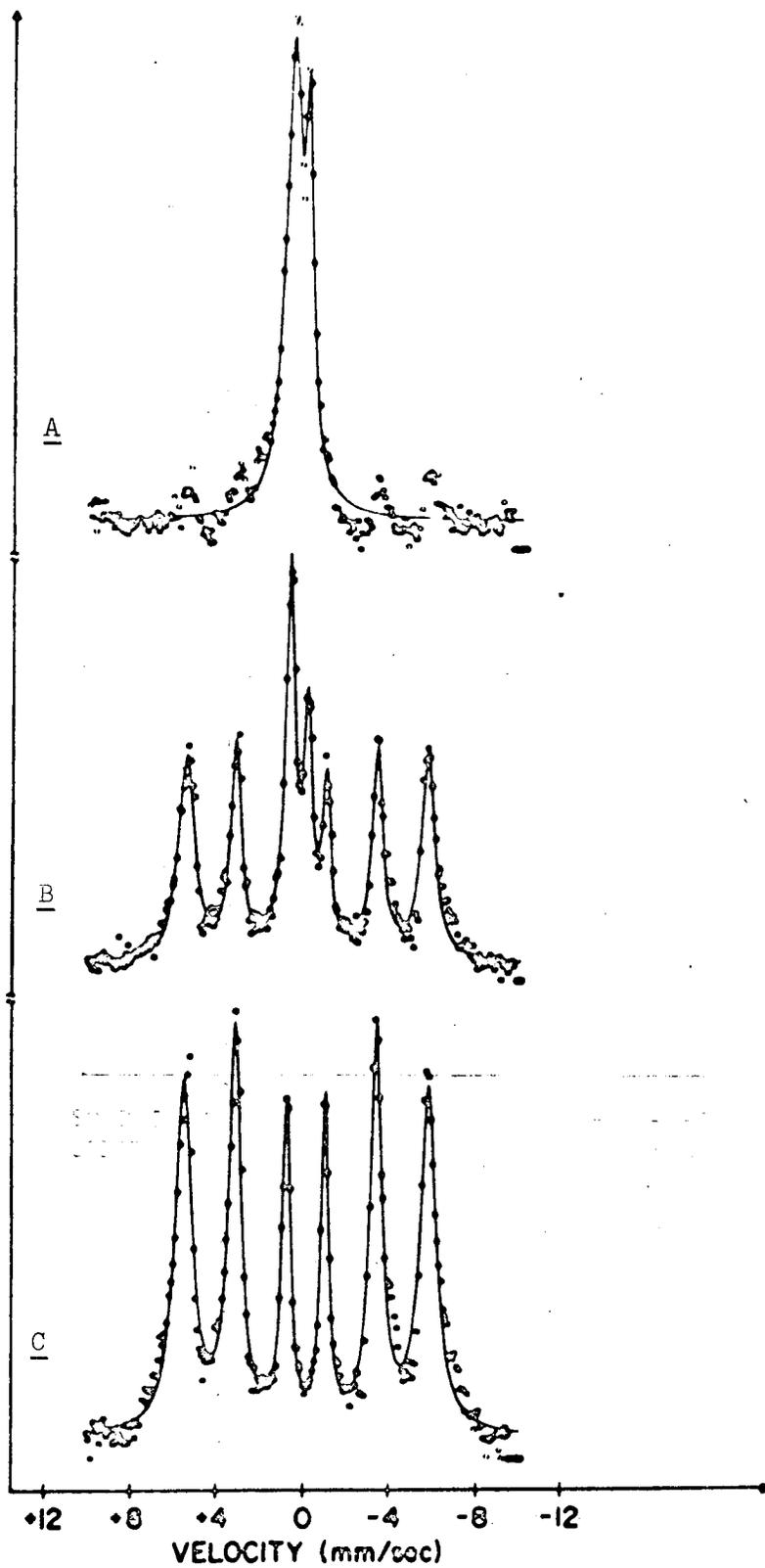


Fig. 11. Backscatter Mössbauer spectrums of a rusted 1/8-in.-thick steel plate (A, original condition; B, after rust was scraped off; C, after a 1-mm-thick layer was removed from the surface).

Table 3. MÖSSBAUER PARAMETERS OF IRON OXIDES
AND IRON OXYHYDROXIDES AT 300°K

Compound	Chemical Shift, ^a mm/sec	Quadrupole Split, mm/sec	Internal Magnetic Field, koe	Reference
FeO	1.37	0.3	0	62S02
α -Fe ₂ O ₃	0.61	0.20	517	63C08
γ -Fe ₂ O ₃	0.67	0.10	505	61B04
α -FeOOH	0.70 ^c	0 ± 0.1	364 · 37	65R02
β -FeOOH	0.63	0.65	0	This Work
γ -FeOOH	0.65	0.55	0	This Work
β -FeOOH ^b	0.62 ^c	0 · 0.1	533 · 51	65R02

^aChemical shift relative to sodium nitroprusside standard.

^bValues are for 77°K.

^cCorrected, by adding 0.17 mm/sec, to sodium nitroprusside standard.

The Mössbauer effect is indicative of metallic coating characteristics. Figure 12 shows the results obtained on zinc-coated turbine blades. The familiar six-peak spectrum (A) was derived essentially from the base material. Increasing the zinc thickness to 1.2 mils produced the fingerprint (B); the six peaks have disappeared, leaving instead a single peak in the center of the spectrum. This peak indicates that the iron either occupies centers of cubic symmetry or is randomly situated in the lattice. The only region of the sample that could be responsible for this behavior is the diffusion-bonded intermetallic zone. Thus, the backscatter Mössbauer technique can provide, nondestructively, metallurgical information about the intermetallic region without sample preparation.

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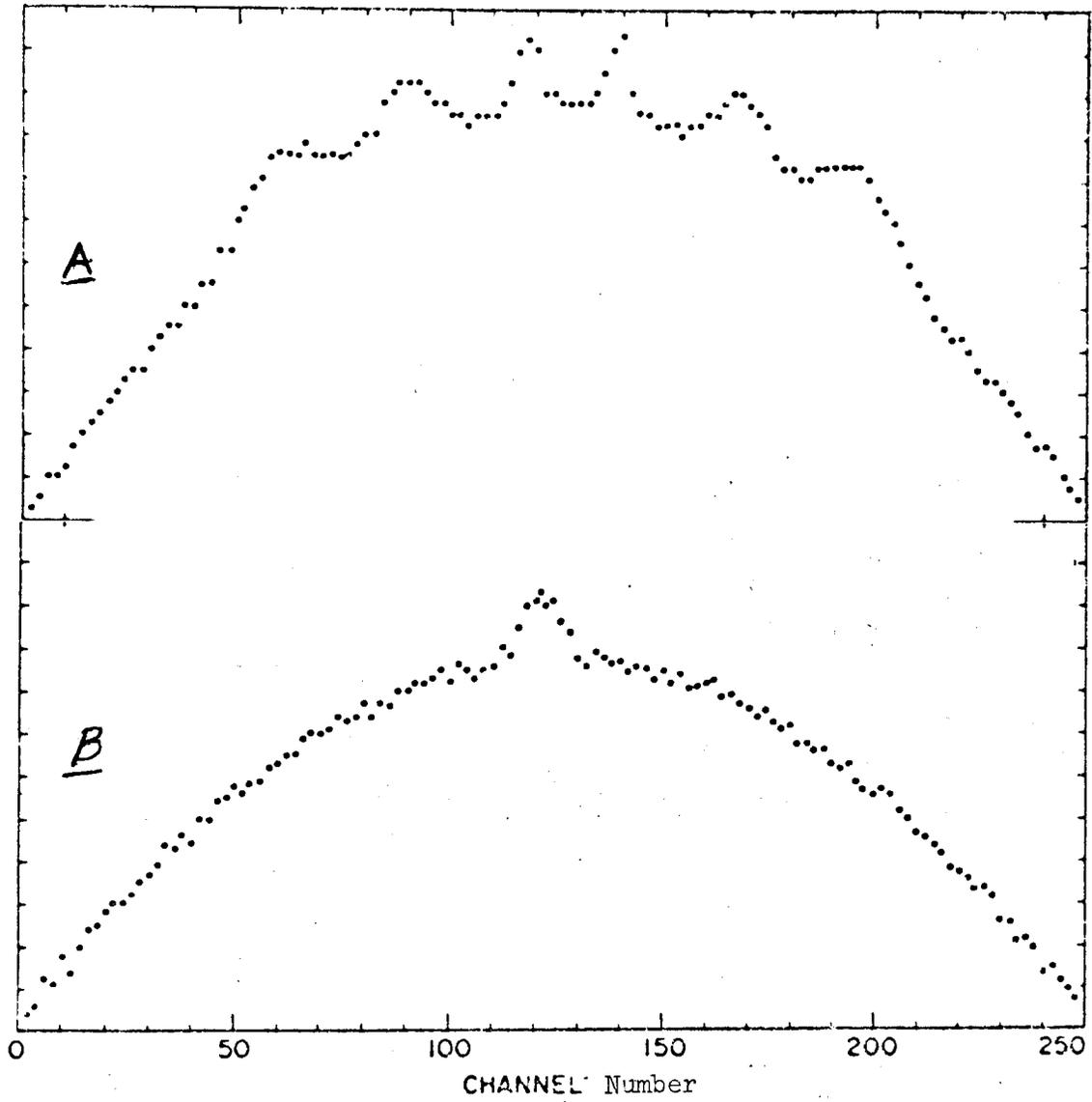


Fig. 12. Backscatter Mössbauer spectrums of zinc-coated turbine blades (A, 0.2-mil coating; B, 1.2-mil coating).

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(DTIE: These reference sheets will be replaced.)

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NEUTRON ACTIVATION ANALYSIS IN OCEANOGRAPHY:

INTERNATIONAL SYMPOSIUM

(2)

Supplementary Keywords: activation, neutron; source, bremsstrahlung; reactor; generator; chemistry, analytical; chemistry, inorganic; biology; ecology; geology; geophysics; oceanology; sedimentology; industry; industry, nuclear energy; tracer; material tracing; water and waste treatment; safety; radio-chemistry; radiolysis; separation, physical process; chromatography; spectrometry; luminescence; secondary radiation effect; solvent extraction; sorption; ion exchange; precipitation; counting; conference; bibliography; data handling; facility description; pollution; comparison; sea; worldwide; Belgium; France; Germany; England; Italy; Monaco; Norway; semiconductor; fallout.

Abstract. The NATO Subcommittee on Oceanographic Research sponsored the International Symposium on the Application of Neutron Activation Analysis in Oceanography held at the Royal Institute of Natural Sciences of Belgium, Brussels, June 17--22, 1968.* Thirteen participants from eight countries presented 12 papers. The symposium is summarized briefly below; the papers presented are listed at the end of the summary.

(Introduction)

In his inaugural speech, the chairman of the symposium, Professor A. Capart, Royal Institute of Natural Sciences of Belgium, pointed out that the characterization of oceanic regions no longer depends on their the laborious measurement of Δ temperature and salinity. The determinants are likely the presence or the absence of certain oligo elements. In the scope of oceanographic research, he believes that there are three important problems without sufficient data: the energetic exchange between "air and sea," the oceans' productivity of nutrients, and sea pollution. As an oceanographer, he expects neutron activation analysis to provide a fingerprint of each oceanic mass

* NATO Subcommittee on Oceanographic Research, "International Symposium on the Application of Neutron Activation Analysis in Oceanography," Institut Royal des Sciences Naturelles de Belgique, Bruxelles, Juin, 1968.

and to answer previously unanswered questions.

The papers presented reiterate the need for great care in taking and preserving samples so as to ensure that the results of their analysis will indicate correctly their initial compositions. Neutron activation analysis is shown to be sufficiently sensitive to measure the ocean's trace elements without prior concentration of samples. The technique, used together with neutron generators and on-line computers, permits the handling of the great quantity of results needed to study the innumerable phenomena involved in the interaction between life and chemical elements in the sea---an interaction that starts with the radiant energy of the sun and goes to the bottom of the ocean. Concurrent studies of natural and artificial radioisotopes and of stable elements and their radioactive counterparts are shown to be highly valuable in these fundamental investigations in oceanography.

Inorganic Chemistry of the Ocean (3)

In ^{the} ₁ single paper on the subject, Goldberg¹ assigned ^{three} ₁ characteristics to the inorganic chemical reactions in the ocean: (1) the reactants are in micromolar or submicromolar concentrations; (2) the time constants of the reaction rates are in units of thousands, tens of thousands, or hundreds of thousands of years or more; and (3) the reactions occur at the interfaces between geologic domains --- the sediment-hydrosphere, the hydrosphere-biosphere, and the hydrosphere-atmosphere. He discussed how these reactions may be understood from knowledge of the concentrations and speciations of the reactants, from mineralogical and chemical studies of the products, and from the properties of the reaction sites.

Activation Analysis of Seawater, Marine Organisms, and Marine Sediments (3)

Because activation analysis is not a sea-going technique, samples from the sea must be preserved for a certain period of time. The problems of proper sampling and of pretreatment before the irradiation were discussed by Fukai.² He considered pretreatment procedures, such as phase separations and chemical separations, from a general point of view and discussed briefly separations of organically bound fractions of trace elements and chemical-reaction rates of trace elements in

extremely small concentrations in seawater. He emphasized the need to cross-check the results of various methods of analysis and suggested that the neutron activation technique may serve as a reference and permit the adoption of procedures that are free of reagent blanks.

The rates of introduction of radioactivity into the marine environment must be so adjusted as to ensure that the rates of return of radioactivity to man, and the consequential human radiation dose rates, do not exceed those deemed acceptable. Preston and Dutton³ used neutron activation analysis in studies of marine radioactive pollution. They studied the cycling of trace elements in ^{United Kingdom} coastal and shelf waters already contaminated by radioactive effluents. Their study has particular relevance in predicting the environmental consequences of the use of nuclear explosives or the contamination resulting from waste-disposal operations. They reviewed the use of neutron activation and atomic absorption spectrophotometry for the determination of traces of selected metallic elements in multicompartiment concentration processes typical of United Kingdom coastal waters. They compared the behavior of stable and radioactive nuclides and their change with distance from disposal area and with time.

Over a 3-year period, Høgdahl⁴ studied the distribution of lanthanides in seawater. He presented the results of some 40 to 50 analyses, discussed the results in relation to seawater masses, and gave a tentative explanation of the observed distributions.

At their Institute, in the Activation Laboratory built for the purpose of determining trace and oligo elements in seawater and marine organisms, Peeters and Lacroix⁵ analyzed samples from the North and Mediterranean Sea, brought back from their expedition in August 1967 onboard the oceanographic vessel "The Charcot." Without using chemical separation, they were able to determine more than 20 elements. They ^{presented} _A gamma-ray spectra of ^{four} _A samples, including surface- and deep-water samples. They discussed the problems of sampling without contamination and of obtaining quartz sufficiently pure for use as irradiation

capsules. With the objective of "fingerprinting" each type of sea or ocean and correlating its types of flora and fauna with the composition of their environment, they showed how the results of their first experiments justified their plan to acquire a multiprogramming computer and a neutron generator.

The marine biosphere must be included in any comprehensive study of the oceans' trace elements, because biological assimilation is an important mechanism that affects the concentrations and behaviors of many trace elements in the marine environment. In a more broad-based study, Robertson, Rancitelli, and Perkins⁶ measured the concentrations of numerous trace elements in seawater, marine organisms, and pelagic sediments. They counted neutron-activated samples directly on either a multi-dimensional gamma-ray spectrometer or a large-volume high-resolution $\text{Ge}(\text{Li})$ diode spectrometer after optimized decay periods. They thus measured the concentrations of the trace elements Sr, Cs, U, Co, Sb, Zn, Fe, and Sc in seawater samples and their distribution over a wide geographical area of the Pacific and Atlantic Oceans. The concentrations of Zn, Co, Ag, Cs, Sb, and Sc were measured in a large number of surface, midwater, and benthic marine organisms from various trophic levels. The additional elements Na, K, Rb, Br, Fe, Se, and Hg were also measured in tissue samples from silver salmon and Alaskan king salmon. The elements Mn, Fe, Cu, Zn, Co, La, Sc, and Sb were measured in pelagic sediments and in manganese nodules by directly counting neutron-activated samples on a multi-dimensional gamma-ray spectrometer. With a $\text{Ge}(\text{Li})$ diode detector and properly selected irradiation and decay periods, they also measured the additional elements Al, V, Dy, Ba, Cr, Ce, Sm, Yb, Lu, Hf, and Th in sediment samples by direct counting. The direct-counting methods greatly simplified the multielement analysis of samples from the marine environment by eliminating the uncertainties associated with many complex chemical preconcentration and separation procedures. Also, the inherent risk of contamination of the samples during such procedures was eliminated. The behavior of several artificial radioelements entering the marine environment as fallout, relative to their stable element counterparts, was

determined and points out the importance of chemical or physical form on the behavior of an element in the ocean. Robertson ⁷ described methods for extracting trace radionuclides from large volumes of seawater to depths as great as 1600 meters. also.

Studies of the marine biosphere were described by Merlini ⁷. She pointed out that the interest in the amounts and distribution of stable elements in aquatic organisms has increased with the recognition that such information is important as a guide and measure of the levels that radioisotopes, introduced as contaminants, might attain at the equilibrium state. She described the determination by neutron activation analysis of ^{five} elements (Co, Cs, Mn, Sc, and Zn) over a period of ^{one} year in fishes, macrophytes (water plants), molluscs, and the water of Lake Maggiore, north Italy. She compared her results with results reported in the literature for the same elements in seawater and marine organisms. She concluded that activation analysis is an excellent tool for obtaining information that is especially useful to biologists, who are faced with many badgering problems related to radiocontamination of environments, especially aquatic.

The usefulness of neutron activation analysis in the different branches of radioecology was discussed by Feldt.⁸ He presented results of determinations of six elements (Co, Cu, Fe, Mn, Mo, and Zn) in deep-sea organisms. The difficulties of sampling and storage were also considered. The facilities of the new Activation Laboratory of the German Board of Fisheries Research were described.

Activation analysis techniques used by Boudin and associates⁹ in geology and glaciology are also useful in oceanography. Boudin described the advantages of the techniques, based on detection of short-half-life nuclides, for the determination of lutetium, zirconium, and hafnium in oceanographic samples. Low contamination levels were achieved by the storage and irradiation of samples in the frozen state and the use of specially tested clean material in dust-free laboratories. The activation analysis results were compared with those obtained by atomic absorption and isotope dilution.

No copy of the paper presented by Chesselet¹⁰ was made available for inclusion with the symposium papers, nor was an abstract.

Potential of New Facilities and Computer Techniques in Oceanography (3)

Two papers described the potential of new facilities and computer techniques in oceanography but did not give results of the analysis of oceanographic samples.

The activation analysis laboratory of the EURATOM Research Center at Ispra, Italy, was described by Girardi,¹¹ who discussed the irradiation facilities, radiochemical techniques, and counting techniques and computer data handling. The irradiation system is composed of ^{six} irradiation channels, a fast pneumatic system, and a 30-position storage and transfer rack. The advantages of the system in oceanographic applications ^{were} stressed. The radiochemical techniques include the use of "inorganic ion separators" (IIS), which have potential for simple and fast radiochemistry, especially elimination of sodium. The counting system consists of two complete gamma spectrometers with various detectors, including a Ge(Li) semiconductor, two pneumatic sample changers, and different read-out units. Useful computer programs were outlined briefly.

Computer programs were described by Pagden¹² that use published nuclear data and spectrometric criteria in calculations which enable a user to know what radioisotopes may possibly be present in a sample. The programs are based on the observation of gamma-ray spectra of various activation products; the spectra are detected with Ge(Li) detectors.

(HPR)

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3. A. Preston and J. W. R. Dutton, The Application of Neutron Activation Analysis to the Study of Trace Elements in United Kingdom Coastal Waters, Fisheries Radiobiological Laboratory, Ministry of Agriculture, Fisheries, and Food, Lowestoft, Suffolk, England.

4. O. Høgdahl, Neutron Activation Analysis of Lanthanides in the Hydrosphere, Central Institute for Industrial Research, Blindern, Oslo, Norway.
5. E. Peeters and R. Lacroix, A Fine Structure Analysis of Trace Elements in Seawater Samples: Multiparametric Spectrometry after Thermic Neutron Activation, Laboratory of Oceanographic Physics, Royal Institute of Natural Sciences of Belgium, Brussels, Belgium.
6. D. E. Robertson, L. A. Rancitelli, and R. W. Perkins, Multielement Analysis of Seawater, Marine Organisms, and Sediments by Neutron Activation Without Chemical Separations, Battelle Memorial Institute, Pacific Northwest Laboratory, Richland, Wash.
7. M. Merlini, Trace Element Analysis of Aquatic Organisms, Biology Service, EURATOM Joint Research Center, Ispra (Varese), Italy.
8. W. Feldt, Application of the Neutron Activation Analysis in Radioecology, German Federal Board of Fisheries Research, Isotope Laboratory, Hamburg, Germany.
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10. R. Chesselet, Application of Neutron Activation Analysis to the Study of the Low-Level Composition of Marine Aerosols, Center for Radioactivities, National Center for Scientific Research, Gif-sur-Yvette, France.
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12. I. M. H. Pagden, Some Programs for the Resolution of Gamma Ray Spectra into Their Components, Bedford Institute of Oceanography, Canada.

THE USE OF IONIZATION IN THE AIR FOR LIGHTNING PROTECTION (2)

G. Berio*

Supplementary Keywords: ionization of gas; Am-241

Abstract: The E.F. (electricite froide) lightning rod, which uses eight ^{241}Am sources to promote the ionization of air is described. This rod has an effective radius of 250 meters and is being used extensively in Europe to protect commercial installations from lightning.

(Introduction)

The two classical methods for the protection of buildings and other structures from being harmed by lightning are the Franklin lightning rod and the Faraday Cage. The Faraday cage, however, is not practical for modern buildings.

The Franklin lightning rod, developed by Benjamin Franklin in the eighteenth century, is based on the principle of that a grounded point high in the air will cause ionization of the air and thus provide a track through which the lightning will discharge.

A Liechtenstein firm, General Protection Establishment, has recently improved the Franklin lightning rod, using a theory developed by Szilard¹ that air ionized by radiation would provide better protection than a standard Franklin rod. This rod is marketed under the name E.F. lightning rod. E.F. stands for "electricite froide," which means cold or low-energy, high-voltage electricity.**

Description of the E.F. Rod (3)

A picture of the E.F. rod is shown in Fig. 1 and a sketch² is shown in Fig. 2. The basic parts of the device are a cylindrical rod having a

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** A recent Euratom report³ lists 8 European firms that make radioactive lightning rods and states that there are between 5 and 10 thousand such rods installed in Europe.

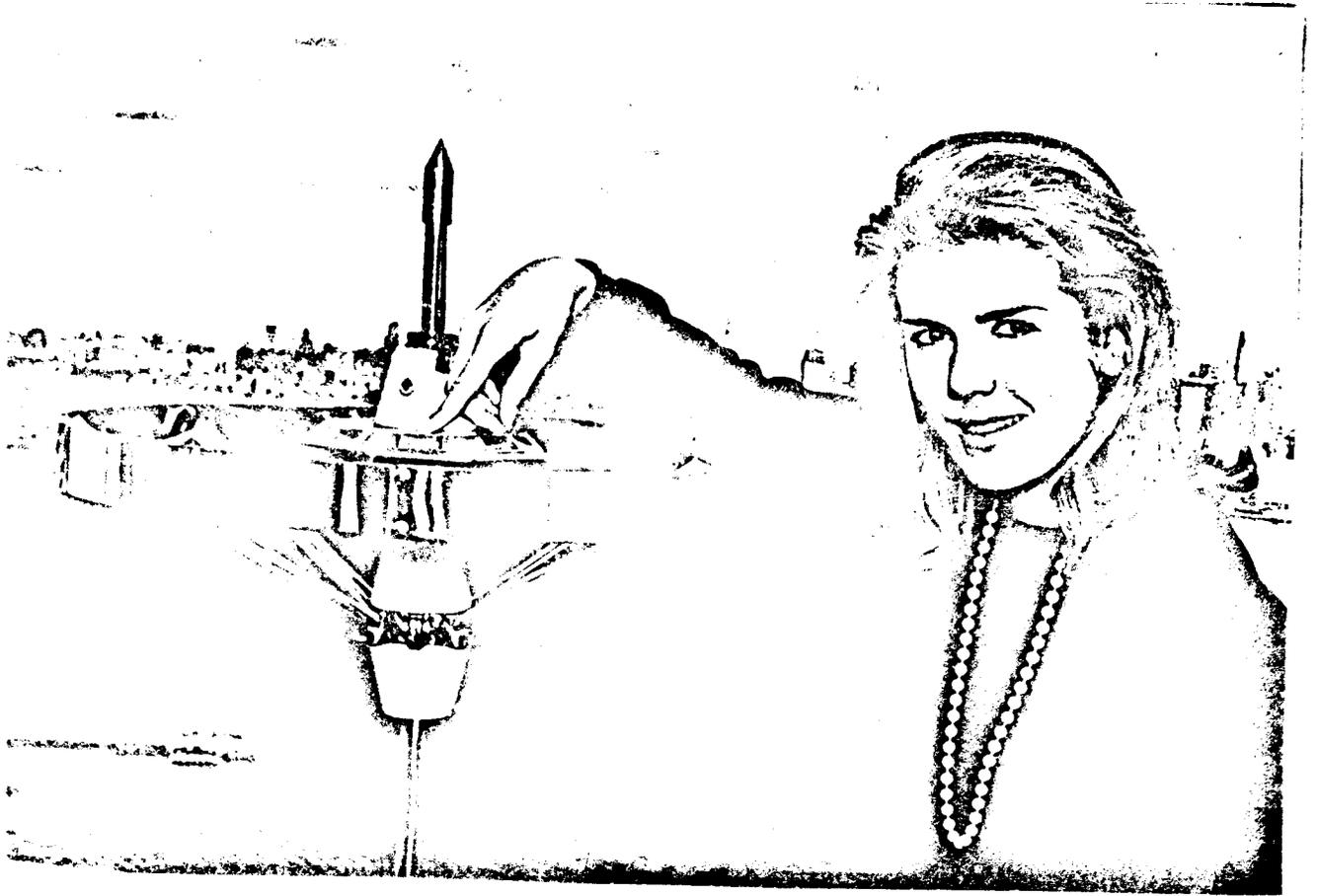


Fig. 1. Photograph of E.F. lightning rod.

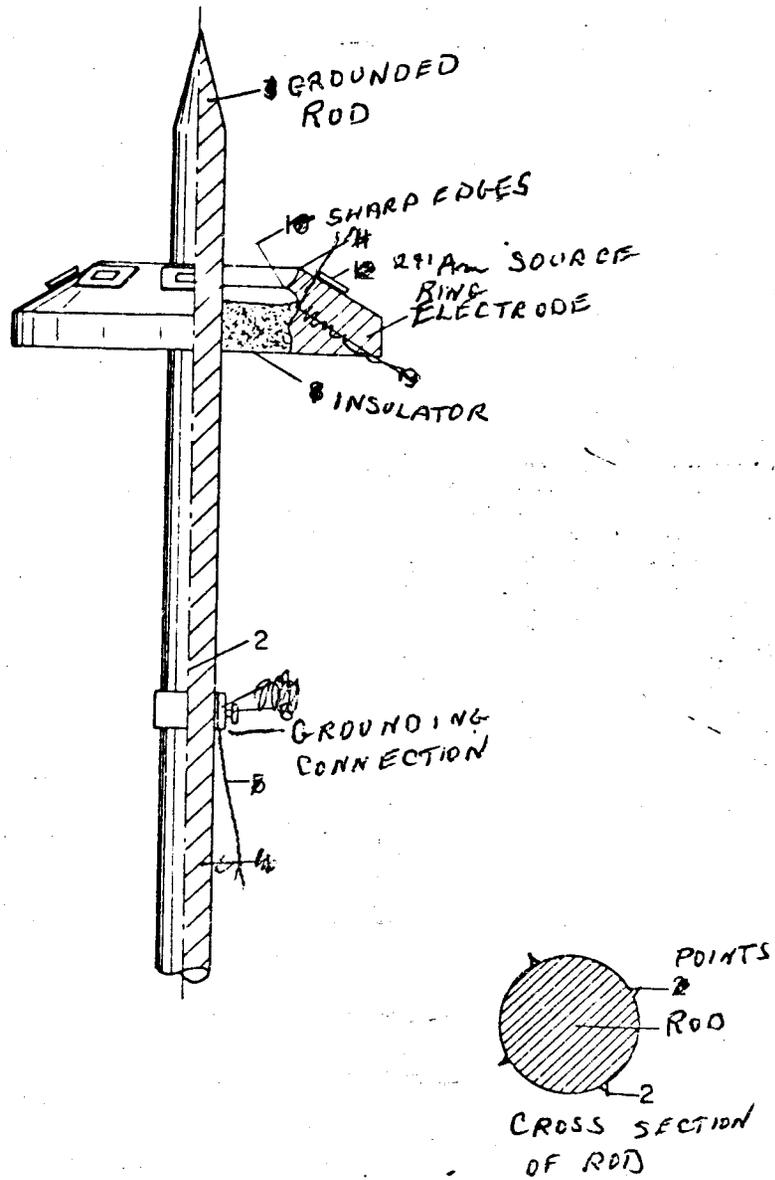


Fig. 2. Schematic diagram of E.F. lightning rod.

sharp point at the top and several ridges running longitudinally down the rod, an annular electrode having sharp edges directed toward the rod, an insulator which separates the rod and the electrode, and eight ^{241}Am sources in the electrode.

When the ^{device} is in operation, the rod is grounded and the electrode assumes the potential of the atmosphere. Under thunderstorm conditions, the potential gradient between the rod and the electrode reaches high enough values that there will be electrical discharges between the sharp edges of the electrode and the ridges of the rod. This discharge is, of course, aided by the ionization of the air caused by the alpha particles emitted by the radioactive sources. The electrical discharge causes additional ionization of the gases, the net effect being to create an ionized cloud around the unit that will provide a path for the lightning to the rod where it will be grounded.

At the present time, eight ^{241}Am sources are provided on each electrode. This ensures protection to the area within a radius of 250 meters.

Tests and Use of the System (3)

The ionizing effect of an excellent Franklin installation was compared with that of an E.F. installation during periods of stormy weather in 1964. The measurements were taken on the Concord skyscraper (64 meters) in Toulon, France. It was observed that under all trial conditions, the E.F. installation, grounded on the same ground plate as the Franklin unit, at the same gradient values, gave an approximate rate of ionization 10^4 times the Franklin unit.

In regions such as Italy and the Spanish Peninsula—examples of European countries most affected by lightning—entire units of telecommunication systems, networks of electricity production and distribution, and fuel and gas systems, as well as a large number of important industries such as FIAT, have adopted the E.F. system for lightning protection.

In the nuclear field, the Pierrelatte Plant for isotopic separation in France has been protected since 1963 by 25 E.F. installations. The plant is situated in an area particularly prone to lightning. The French Commissariat à l'Énergie

Atomique has adopted the E.F. system to assure the protection of its plants at Fontenay-aux-Roses and Chinon. The French Electric Power Authority has also chosen this type of protection for its nuclear plant at St. Laurent des Eaux. The Italian government assures the protection of its nuclear plants of Saluggia and Garigliano with the E.F. system, as Euratom has done for its Ispra plant.

An installation of one unit on the Acropolis in Athens is shown in Fig. 3, and Fig. 4 shows an E.F. lightning rod on a monument in Santander, Spain.

(RHL)

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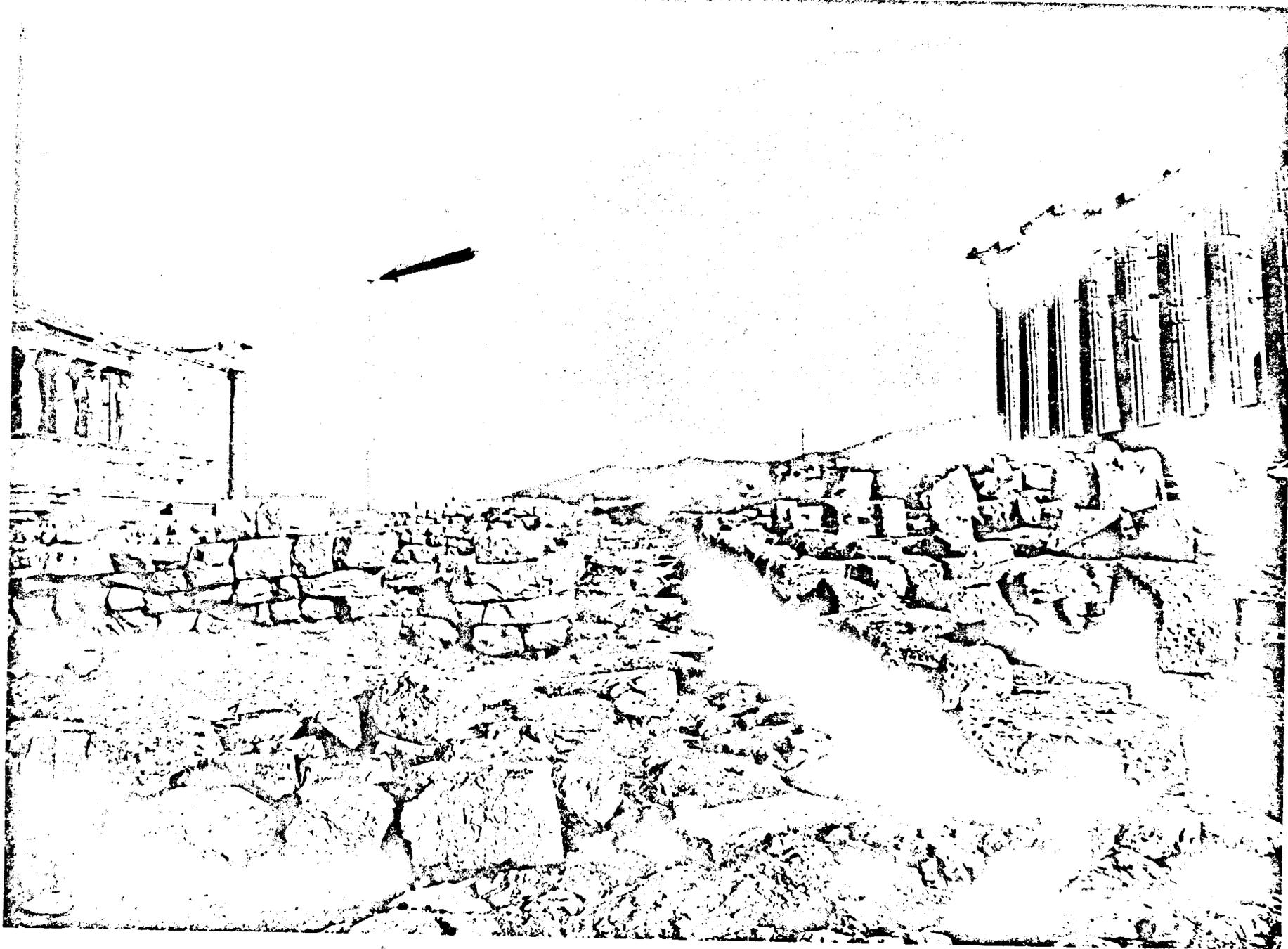


Fig. 3. E.F. protection installed on the Acropolis, Athens, Greece.

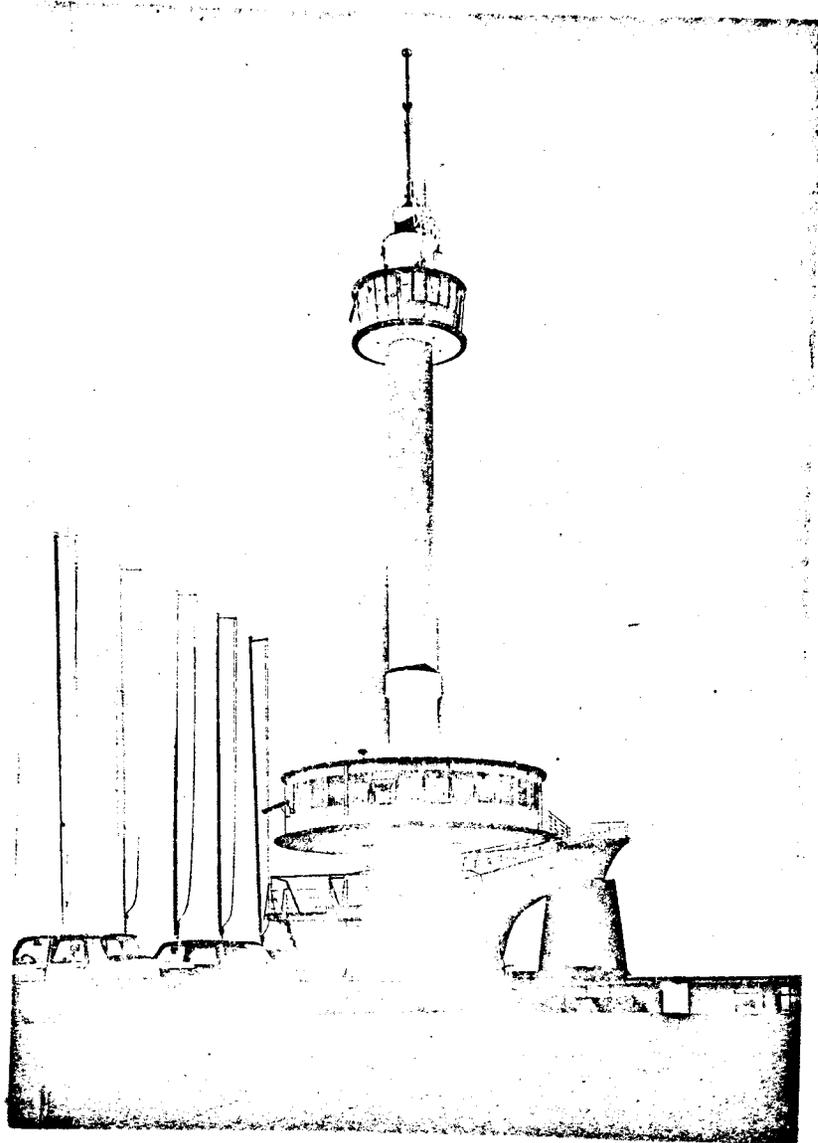


Fig. 4. E.F. lightning rod on monument at Santander, Spain.

WATER PERMEABILITY OF COATING FILMS USING A
RADIOISOTOPE TRACER TECHNIQUE*

Supplementary key words: Measurement, other; physical property; coating; hydrogen-3.

Abstract. A radioisotopic tracer technique was developed for determining the moisture permeability of protective coatings. In tests with epoxy and alkyd resins, the method was more sensitive and rapid and results were more precise than by the ASTM method for measuring film penetrability to moisture.

(Introduction)

Corrosion of metal costs U.S. military and civilian establishments more than \$10¹⁰ annually.¹ Metal corrosion is electrochemical in nature,²⁻⁶ and water must be present for corrosion to occur. Painting is the method most widely used for isolating metallic surfaces from water or water vapor in order to prevent or decrease corrosion. However, because water or other corrosive materials may penetrate paint and reach the substrate, a study of the mechanism of water vapor permeation through paint films should help in understanding the limitations of such films, aid in improving them, and thus contribute to corrosion control.

Unfortunately, studies⁷⁻¹² on the permeability of paints and other coatings to moisture have not given satisfactory data, largely because of the difficulty of measuring the amount of water vapor diffusing through a film. The quantity is usually too minute to be measured accurately by chemical means, and the diffused vapor is not in a form that is readily

*Extracts from Technical Report R-674 (April 1970) by Eddy S. Matsui, Naval Civil Engineering Laboratory, Port Hueneme, Calif. 93041.

determinable by ordinary analytical methods. For example, the American Society for Testing Materials (ASTM) standard method D1653-62T involves determination of the weight lost when a quantity of water passes through a sample film under a certain set of conditions, but the effect on diffusion of variations in temperature and pressure, for example, are not accounted for. At least a week is required for a readily measurable quantity of water to pass a membrane, and even then accuracy is low.

Accordingly, a study was undertaken by the Naval Civil Engineering Laboratory (NCEL) to develop a new and improved method for measuring water vapor diffusion through paint films, using tritiated water as a tracer. In the NCEL tests, the effects on diffusion rate of various parameters---film thickness, vapor pressure temperature, film composition, film pigment type and concentration, and form of moisture (liquid or vapor)---were investigated. Detailed data on these tests are reported in the original report, but only the methodology used is reviewed here.

Theory (3)

The passage of water vapor through an organic membrane such as a resin-based paint is not a simple diffusion process but occurs^{7,13} in three stages:

1. Condensation of the vapor on and solution into one side of the film
2. Diffusion through the film
3. Evaporation from the other side

The permeability, P_r (g.cm/cm².hr.cm Hg), is thus the product of the vapor diffusion coefficient, D (cm²/hr), through the membrane and its solubility, S (g/cm³.cm Hg), in the membrane:

$$P_r = DS \quad (1)$$

The driving force for water-vapor diffusion through a membrane is the

vapor-pressure difference between its two faces, and the amount of water vapor, Q , diffusing through a given membrane at a given temperature depends on the effective area, A , and thickness, d , of the membrane, the vapor-pressure differential, Δp , between the faces of the membrane, and the time, t , during which the diffusion occurs. The one-dimensional steady-state equation^{7,14} is, from Fick's law,

$$Q = \frac{P_r}{d} A t \Delta p \quad (2)$$

The permeability, which characterizes the water-vapor-transmission resistance of the membrane under consideration, depends on the nature of both membrane and the permeating vapor. Incidentally, this equation is completely analogous to the basic heat-conduction equation,

$$Q = \frac{K}{d} A t \Delta T$$

where K is the conductivity and ΔT the temperature differential.

Method (3)

Apparatus (4)

A diffusion cell (Fig. 1), designed at NCFEL, was constructed of Plexiglas and Pyrex tubing. Each half of the cell consisted of an exposure chamber and a water mantle, which maintained the desired temperature in the chamber and in the sample membrane during experiments. The membrane was placed between two fine-copper-gauze supports, which protected it from any damage due to a sudden change in pressure between the two chambers. The two halves of the cell were sealed by an O ring and a rubber gasket. A thermocouple probe on the lower chamber allowed direct monitoring of the membrane temperature. When the cell was assembled, the exposed membrane area was 20 cm².

A vacuum system (Fig. 2) was provided to maintain a known, constant

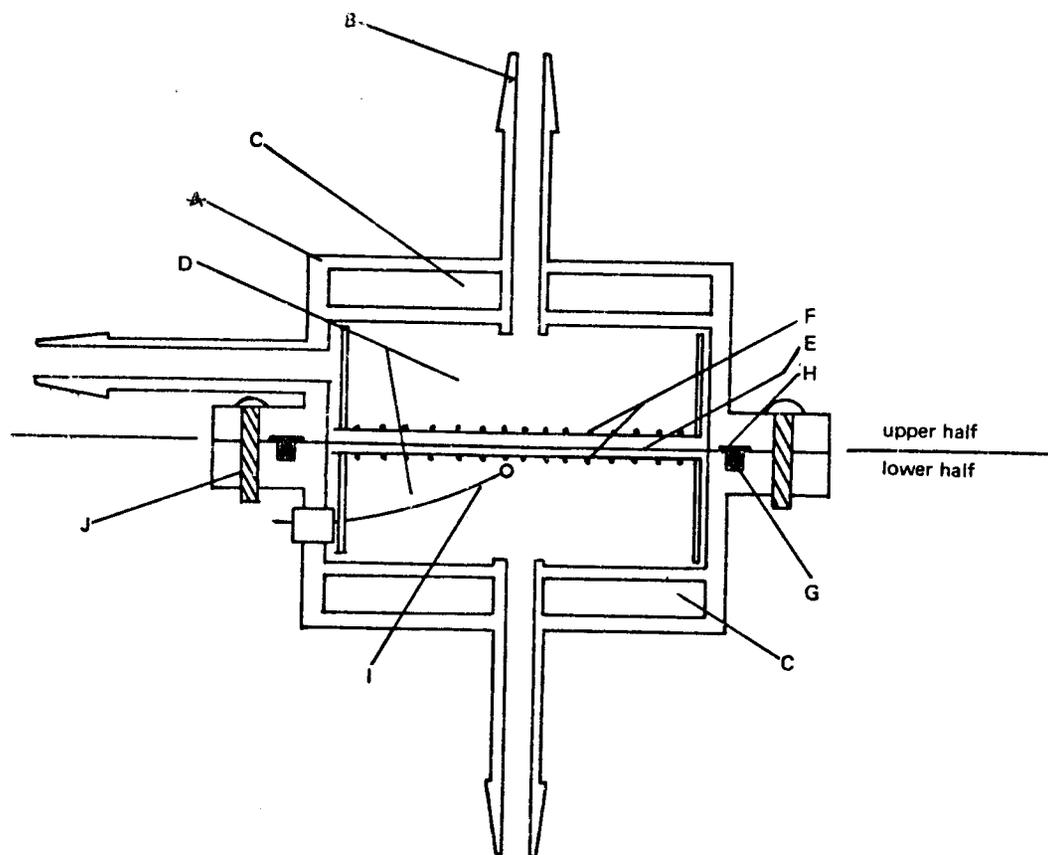


Fig. 1. Cell used in membrane-permeability tests. (A) Plexiglas wall. (E) Pyrex tube. (C) water mantles. (D) Exposure chambers. (F) Sample membrane. (F) copper gauzes. (G) O ring. (H) Rubber gasket. (I) Thermocouple probe. (J) Screw.

water-vapor pressure on the membrane surface so that Eq. 2 would be valid.

The system was fabricated from high-vacuum-grade Pyrex. A vessel containing tritiated water (approximately 15 mCi/ml) was connected through a T-shaped stopcock to the lower half of the diffusion cell and to one arm of a mercury manometer. The other arm of the manometer was connected to the upper half of the diffusion cell. The manometer thus provided direct reading of vapor-pressure differential between the two faces of the membrane. The manometer was equipped with a temperature-regulating device connected to the heater for the tritiated water to control the vapor pressure in the

system. A Nichrome wire coil prevented condensation of tritiated water vapor in the Pyrex tubing by maintaining the temperature above the dew point at all times. The diffusion cell, the manometer, or other element could be disconnected from the assembly by the use of stopcocks. The entire system was evacuated prior to the experiment.

Procedure (4)

The membrane studied was placed between the two halves of the diffusion cell.

Tritiated water vapor was admitted to the manometer through the T-shaped stopcock. When the desired pressure had been reached, as indicated by the manometer, the vapor stream was switched to the lower half of the diffusion cell. This time was taken as the starting time of the experiment, i.e., $t = 0$. The stopcock in a line leading to a receiving tube, which was immersed in coolant, was then opened to condense and freeze out any tritiated water vapor that passed through the membrane. Freezing of the receiving tube ensured a constant pressure differential between the two faces of the membrane, since any vapor that passed through the membrane would be condensed and frozen.

After a predetermined length of time---between 1 and 2 hr---the tritiated-water vessel was isolated from the rest of the system by turning the T-shaped stopcock, and this time was recorded as the end of the experiment. A vessel of absolute alcohol was warmed to release alcohol vapor and into the upper half of the diffusion cell and purge it of any residual tritiated water vapor, the vapor being carried to and condensed in the receiving tube.

The receiving tube containing the frozen-out tritiated water was removed from the vacuum system, and the activity was counted, using a liquid

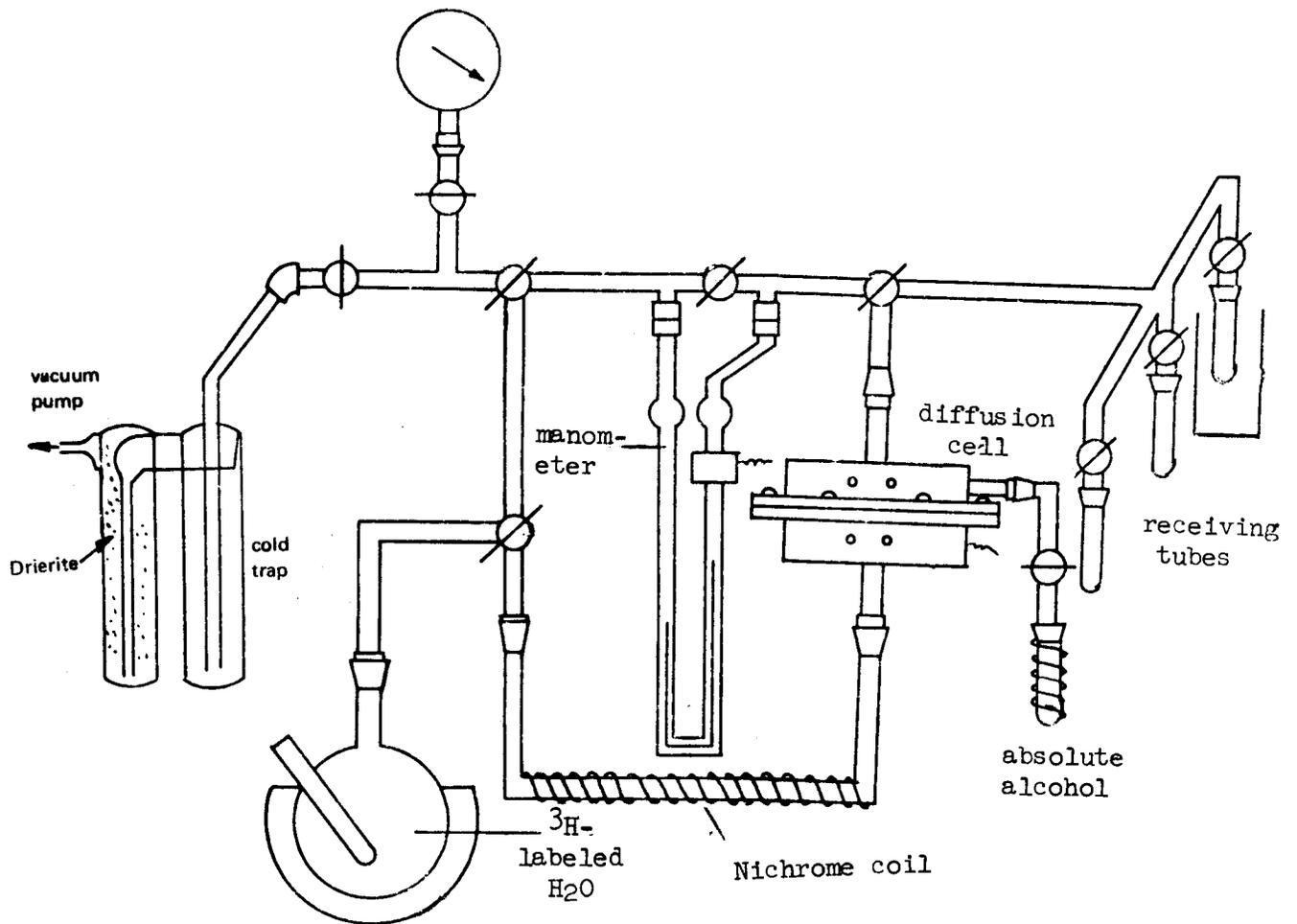


Fig. 2. Apparatus system for testing vapor diffusion through a membrane.

scintillation spectrometer. The amount of vapor that diffused through the membrane was used to calculate the diffusion rate, R , in $\text{mg}/\text{cm}^2\cdot\text{hr}$, and the permeability constant for the membrane in $\text{mg}\cdot\text{mm}/\text{cm}^2\cdot\text{hr}$:

$$R = \frac{N 100}{a A t k}$$

$$P_r = \frac{N d 100}{a k A t \Delta p}$$

where N is the activity in counts per minute, a the specific activity of the tritiated water in counts per minute per milligram, and ϵ_k the efficiency of the scintillation counter in percent; the other symbols have the same meanings as in the earlier equations, with A in square centimeters, d in millimeters, t in hours, and p in centimeters of mercury.

In order to decrease the random error of counting to less than 1% at the 95% probability level, each sample was counted to register 40×10^4 or more counts per measurement. A minimum of five condensates was collected for analysis after a steady rate of diffusion had been attained. The counting efficiency of the liquid scintillation counter was determined by applying the channel-ratio method described by E. T. Buch.¹⁴

The results were processed by computer techniques. The spectrometer was loaded with up to 100 samples, and each sample was counted.

The number of counts from each channel was automatically put on punch cards, which were fed into the computer along with other essential data. The computer printed out in tabular form the counting rate, channel ratio, counting efficiency, diffusion rate, and the permeability constant of each sample.

Comparison of Radiotracer and ASTM Methods (3)

The water-vapor-diffusion rate obtained by the radiotracer method was compared with that obtained with the same films using ASTM method D1653-62T. The ASTM method uses a two-part permeability device---a shallow flanged cup and a flat ring matching the flange on the cup. Water is poured into the cup, and the film under test is held tightly between the cup and the ring by clamps.

The assembled cup was placed in a desiccator over P2O₅ and weighed every 24 hr for 1 week. The desiccator was in a well-ventilated room where

the temperature was maintained between 70 and 85°F. The rate of water-vapor passage through the film was determined from the weight-loss rate.

Since the amount of moisture passing through a film is affected by the film thickness, and since it is very difficult if not impossible to produce films of exactly the same thickness, the films used in the ASTM test were first used in the radiotracer experiments.

The coefficient of variation (a measure of the relative dispersion about the sample mean) values determined by the ASTM method were considerably larger than those determined by the radiotracer method (Table 1).

Table 1. COMPARISON OF WATER-VAPOR-DIFFUSION RATE THROUGH ALKYD FILM AS DETERMINED BY THE RADIOTRACER AND ASTM METHODS

Alkyd Film Thickness, ^a mils	Diffusion Rate, ^b $\mu\text{g}/\text{cm}^2\cdot\text{hr}$		Standard Deviation		Coefficient of Variation, %	
	Radiotracer Method	ASTM Method	Radiotracer Method	ASTM Method	Radiotracer Method	ASTM Method
0.9	358.30 ± 17.94	380.38 ± 41.91	14.43	33.71	4.02	8.86
1.9	197.33 ± 6.55	196.32 ± 20.35	5.27	16.37	2.67	8.32
2.7	151.69 ± 14.15	136.80 ± 14.57	11.38	11.72	7.50	8.56
3.5	102.58 ± 4.83	132.48 ± 98.26	3.89	79.03	3.79	59.66 ^c

^a Unpigmented alkyd film (TT-R-266, Type II).

^b At 95% confidence level.

^c Had not attained a steady state of flow after 3 days.

The greater precision of the radiotracer method was shown by the results of the F test---a statistical measure used to compare the precision of two sets of measurements by taking the ratio of two variances, i.e., the variance obtained in the radiotracer method divided by that obtained in the ASTM method. When the ratio of the variances exceeds the critical value of F, which is obtained from a statistical table, there is considered to be a significant difference in precision between the two systems.

In the NCEL experiments, the critical F values at the 55 and 99.9% levels were 3.37 and 5.30, respectively. The F ratio obtained, 19.78, greatly exceeded these two critical values. This indicated that the NCEL radiotracer method was considerably more precise than the ASTM.

The sensitivity of the measuring system used in the radiotracer method can be increased to measure as low as 1 μg or less, where the analytical balance used in the ASTM method is accurate to only $\pm 100 \mu\text{g}$.

Results are obtained more rapidly by the radiotracer method. Once a steady state of flow has been attained, the diffusion rate can be measured in 1 to 2 hr, compared to a week for the ASTM method.

The radiotracer method permits separate control of various parameters, while the ASTM method does not. With the radiotracer apparatus, precisely monitorable vapor pressure differential and temperature provide the means for examining separately the effects of vapor pressure and temperature on film permeability. With the ASTM method, temperature cannot be monitored without affecting the vapor pressure.

Further details of the sample preparation, the computer program, and the reliability of the counting system are given in ref. 15.

Editors' note: A related article appeared in I & RT 8(1),
"A Radiochemical Study of the Diffusion of Ions through Protective Films" by A. L. Glass and J. Smith.

(MG)

DTIE: A NEW SET OF REFERENCES WILL BE SENT TO YOU SHORTLY

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ALPHA-PARTICLE DENSITOMETER FOR MEASURING THE DENSITY OF THE MARTIAN
ATMOSPHERE (2)

Supplementary Keywords: Source, alpha; measurement, density; measurement and control system; space; americium-241; polonium-210; curium-242; x-ray fluorescence.

Abstract. A feasibility study* indicated that an alpha-particle densitometer may be used to measure the density of the Martian atmosphere, and construction of a prototype instrument is recommended.

(Text)

It is expected that, within the next decade, instruments will be landed on Mars which will send back vertical atmospheric density profiles during the entry descent and then diurnal and seasonal surface atmospheric density variations during the rest of the instrument's life. Direct measurement of density by determining the amount of interaction of the atmosphere with radiation from radioisotopes offers great promise. The interaction of matter with such radiation depends only on the amount of matter involved and not on its pressure and temperature, except as these parameters affect the amount present. However, if pressure, temperature, and average molecular weight are known, internal checks can be made.

During the period July 1968 - May 1969, a theoretical study was made of an alpha-particle densitometer, and properties of system components were determined. In the system considered (Fig. 1), an alpha-particle source would be separated from a detector by a fixed distance, D , and the particles would pass through the atmosphere under study to reach the detector. Those reaching the detector would generate a signal, which would be analyzed to give the density of the gas involved. Prior to use, the analyzer output would be measured for several known gas

*Frederick A. Hanser and Bach Sellers, Feasibility Study of Alpha-particle Densitometers for Measuring Planetary Atmosphere Density, report NASA CR-66825, June 1969 (154 pp).

densities to give calibration curves. Thereafter, the density of a gas of the same composition could be measured.

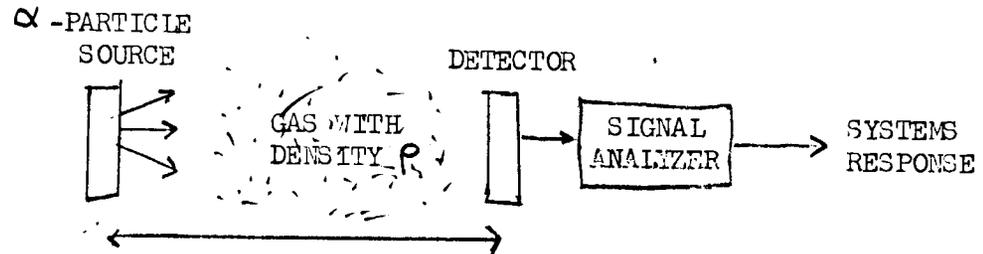


Fig. 1. Basic design of alpha-particle densitometer for determining gas density

Two variants of the system are (1) the thin-source system, which uses a monoenergetic alpha source and measures the average energy loss; and (2) the thick-source system, which uses a broad-spectrum source and measures the count rate above a fixed threshold. For either system, with a gas mixture of CO_2 , N_2 , and Ar---the components thought to be significant in the Martian atmosphere---the density, ρ , is given by

$$\rho_{\text{gas}} = \rho_{\text{CO}_2} + \rho_{\text{N}_2} + 0.67\rho_{\text{Ar}}$$

The thin-source system gives greater accuracy at high densities, high alpha source energies, and large source-detector separations. The thick-source system can be made to give a nearly constant fractional density error with accuracy at low densities of about 0.5×10^{-6} g/ml. Since composition errors of a few percent are likely to be present, the thick-source system is preferable to the thin. Maximum ground level density of the Martian atmosphere is estimated at about 40×10^{-6} g/cm³, and systems will be designed for densities up to 80×10^{-6} g/cm³, i.e., with a safety factor of 2, to allow for possible landing in a depression or for actual densities somewhat greater than estimated.

One requirement for the system is that it must be able to stand complete decontamination prior to launching to avoid contamination of Mars with Earth organisms. Further, a Martian lander will be in flight from Earth to Mars for about 6 months,

and if there is no shield en route between the detector and source, the detector will be continuously irradiated during this time. Since at least several months on the Martian surface will be necessary, the detector should have a lifetime of more than a year, or 3×10^7 sec.

Alpha particles lose energy nearly continuously rather than being completely absorbed, like x rays. The chief difficulty with a particle system is that the response is somewhat dependent on composition, but for the expected composition of the Martian atmosphere, this may not be too critical. The system can be designed so that CO_2 and N_2 produce nearly the same response, and argon can be measured by other methods and its effect compensated for. The argon component can be obtained by measuring the attenuation of the 3.3-keV neptunium M x rays from a ^{241}Am source, which would double the preliminary estimates of 0.75 lb and 200 to 500 mW power. Use with an x-ray attenuation system would allow the values of $\rho_{\text{CO}_2} + \rho_{\text{N}_2}$ and ρ_{Ar} to be determined separately. When no composition information is available, the alpha-particle system alone will measure the density of the Martian atmosphere to $\pm 8\%$. If the composition is known, the error may be decreased to $\pm 2\%$.

Alpha sources considered were ^{241}Am , ^{210}Po , and ^{242}Cm for thin sources and only the first two for thick sources.

The report shows particle-density-measuring-system designs, including evaluations as to which of these designs is best suited for use on a Martian lander, and recommends that a prototype instrument be constructed.

(Martha Gerrard)

Filler for Isotope Technology section

EVALUATION OF A HIGH-SPECIFIC-ACTIVITY ^{60}Co GAMMA SOURCE USED IN A
MONOCHROMATOR

Under contract to the USAEC Division of Isotopes Development, Texas Research Foundation
A & M/has installed a 40 thousand Ci ^{60}Co source and used it in various studies on resonance fluorescence Compton scattering. Several M.S. theses have been written and papers presented at the American Physical Society using the data obtained. Report ORO-662, by John A. McIntyre (May 25, 1969) reviews the program briefly.

(MG)

POTATO SPROUT INHIBITION BY RADIATION. PART 1 (2)

By F. E. McKinney

Supplementary Keywords: (FOOD PRESERVATION / POTATO AND ONION / REVIEW, TECHNICAL / BIBLIOGRAPHY / INDUSTRY, FOOD / INDUSTRY)

On July 8, 1964, the United States became the third country to officially approve the use of ionizing radiation to inhibit sprouting of potatoes. According to the International Atomic Energy Agency,¹ the first country to clear this treatment was the USSR on March 14, 1958; Canada approved on Nov. 9, 1960. Since the USA action, Israel (July 5, 1967) and Spain² have given full approval. France, West Germany, and the Netherlands all have temporary marketing approvals.³

The popular press has covered the subject well, and the technical literature on radiation inhibition of sprouting is extensive. Radiation does inhibit sprouting (Fig. 1). However, even 5 years after the first Food and Drug Administration (FDA) approval, the process has not been commercialized in the USA. There have been some military procurements,⁴ but the housewife cannot buy irradiated potatoes at the supermarket. Interest in irradiation of potatoes remains high; 26 countries have research programs⁵ on this application of radiation. Hence, it may now be appropriate to review the literature on radiation inhibition of sprouting in potatoes. Three noteworthy reviews⁶⁻⁸ exist, but no one of them has attempted any real measure of completeness. Freund⁷ explains why: "An attempt to correlate all potato irradiation research and put it on a common basis is handicapped by the subjective nature of many of the findings and by the almost total absence of a common basis for reporting." His point is well-taken, and, thus, in this review, comparison and evaluation will necessarily be subordinate to reporting.

This is the first article in a planned three-part series for Isotopes and Radiation Technology. This part will provide introductory material, report on

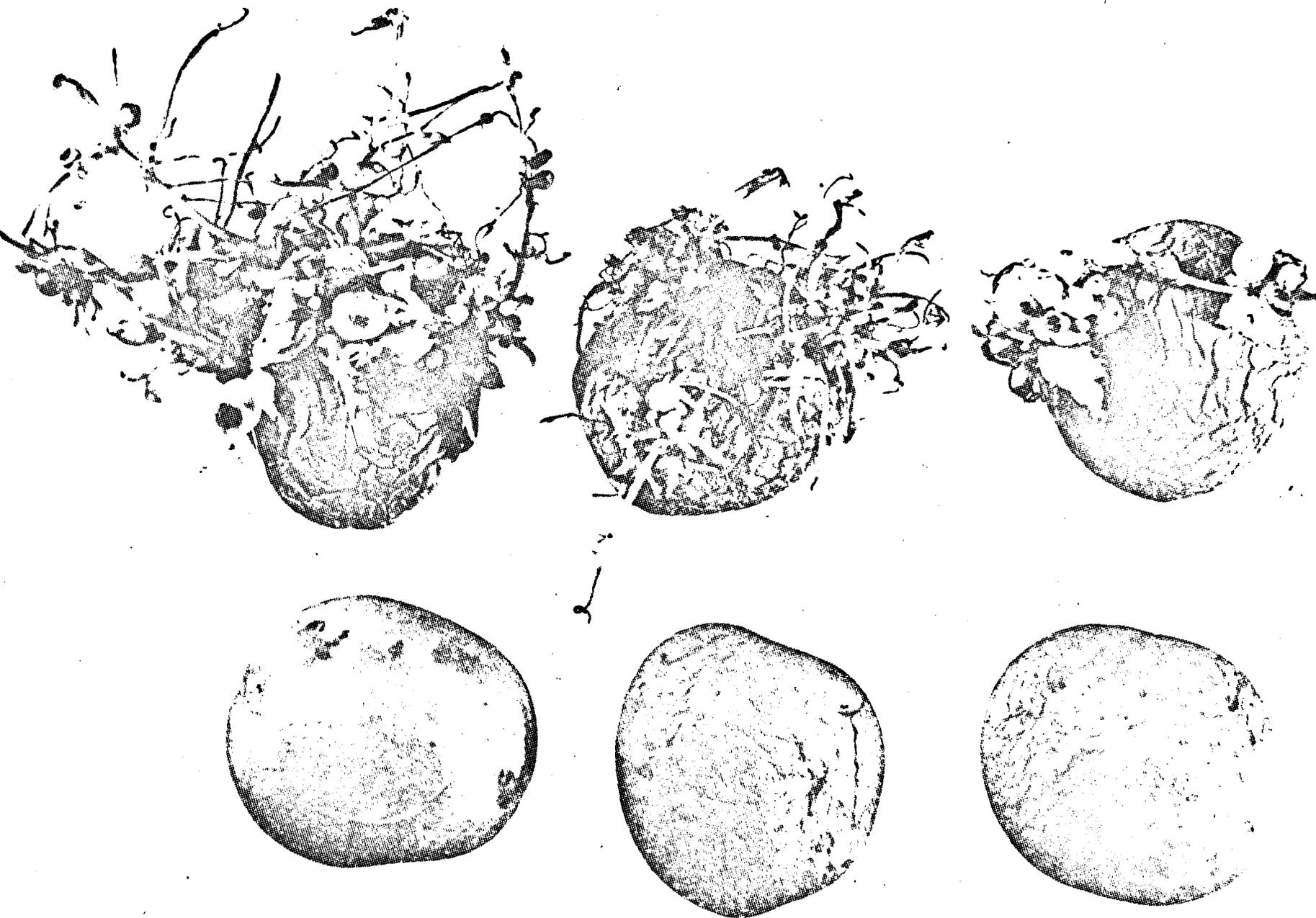


Fig. 1. Typical result of radiation sprout inhibition. Doses ranged from 0 (upper left) to 106 kr (lower left).

legislation, cover the history of the early research into the problem, treat in detail results on sprouting, compare the effects of radiation with those of chemical inhibitors, and describe the sprout / inhibition mechanism. The second article will discuss the effects of radiation on the chemical components, cooking properties, and microbial resistance of the potato. The third article will describe economics, irradiator designs, growth improvement of seed potatoes, feeding studies, and shipping studies. The combined series is intended for later publication as a single report.

Potato History and Production (3)

History (4)

The so-called common Irish potato was first found being cultivated by natives in the South American highlands. In the second half of the 16th century, Spanish explorers introduced the potato into Europe. From Europe, the potato was spread throughout the English-speaking colonies of the world. Most stories of the introduction of the potato into England from Virginia or into Virginia from Spain are legend.⁹

Production (4)

Peak acreage and peak per capita consumption in the United States occurred in 1910. More than 3.6×10^6 acres / were planted in potatoes and individuals ate 200 lb/year. Today, production is nearly three orders of magnitude greater than in 1910, but potato acreage has declined to about one-third the 1910 figure. Per capita consumption is ~135 lb now; about one-third of this is as commercially processed potatoes of some form.

Potatoes are grown in every state and in one place or another throughout the whole year, but 72% of the crop is grown in 26 states and is harvested in October through December. Table 1 shows the distribution of growers and harvesting dates. The Russet Burbank variety is grown most often and accounts for ~35% of the crop. It is a superior baking potato, and processors think it preferable for frozen and dehydrated products. The Kennebec variety is considered best for potato

Table 1. SEASONAL AND REGIONAL U. S. POTATO PRODUCTION---1962 (Ref. 7)

Crop	Harvest	Major producing states	Percent of yearly crop	Varieties
Winter	Jan-March	Fla., Calif.	1.6%	White Rose, Red Pontiac, Red Lasoda, Norland
Spring:				
Early	April-May 15	Fla., Tex.	1.3%	Sebago, Red Pontiac, Red Lasoda
Late	May 16-June 30	Calif., Ariz., N.C., Ala.	8.1%	Cobbler, Kennebec, Sebago, Red Pontiac, Red Lasoda, Norland, White Rose
Summer:				
Early	July 1-Aug 15	Tex., Del., Va., Calif.	4.8%	Sebago, Katahdin, Kennebec, White Rose, Cobbler, Red Pontiac, Red Lasoda, Norland
Late	Aug 16-Sept 30	N.J., N.Y. (L.I. only), Wash., Ore., Wis., Ida., Colo., Calif.	12.6%	Early Gem, Russet Burbank, Red Pontiac, Red Lasoda, Norland, Red McClure, White Rose, Katahdin, Kennebec, Cobbler
Fall	Oct-Dec	Me., Ida., Minn., N.D., N.Y. (inc. L.I.), Colo.	71.6%	Katahdin, Russet Burbank, Red Pontiac, Kennebec, Sebago, Red Lasoda, Norland, Red McClure, Cobbler

chips. Other important varieties* grown in the United States are Katahdin, Red Pontiac, Irish Cobbler, and Sebago.

Potato Storage (3)

Potatoes are usually stored^{*} in one of three ways: (1) Common storage is in large warehouses cooled only by admission of outside winter air; humidity is not controlled. In this type of storage, the temperature varies from 4°C in winter to 20°C in mid-summer. (2) Refrigerated storage for processors of frozen or dehydrated products is at 4 to 10°C and 90% relative humidity (R.H.). (3) Controlled storage for potato chip manufacturers is at 21°C and 95% R.H.; the higher temperature prevents conversion of starch to sugar---an undesirable change in chipping potatoes.

A low relative humidity allows potatoes to yield their moisture to the air and to become soft and flabby; too high a relative humidity encourages microbial growth and rot. A very low temperature could cause freeze damage to cells and a permanent sweet taste in the potato; too high a temperature allows early sprouting.

Under common storage conditions, potatoes harvested in the fall start sprouting in the spring when the weather warms up. Potatoes can be stored at 4°C for up to 8 months before weight losses due to sprouting and general decay prevent their use. Because of the high storage temperatures needed to prevent sugar formation, potatoes stored by chip manufacturers spoil relatively soon, and potato chippers must purchase early potatoes from the next crop at higher prices and transport them to the manufacturing plant. Unlike other potato processors, who locate close to the source of supply, manufacturers of potato chips and potato snack foods locate close to the market because of the need to keep a fresh product on the market and because of the excessively frangible nature of the product.

Losses Due to Sprouting (3)

Estimates vary widely on the extent of sprouting and whether control is actually necessary, although most observers agree that sprouting control would be

*There are almost as many varieties of potatoes as there are varieties of roses; this list includes those varieties most often grown commercially. The index to the entire series of three articles will include all varieties encountered in preparing this review.---FEM.

convenient. One report¹⁰ states most of the sprouting occurs in the consumers' homes and that this loss cannot be assessed. Another report¹¹ states that in 1965 at least 5% of USA production was lost because of sprouting, and Powers¹² uses this figure to estimate that annual losses due to sprouting probably amount to between \$75 x 10⁶ and \$100 x 10⁶. On the other hand, Bradley¹³ argues that disease and rot cause more damage than sprouting and suggests that doses sufficiently high to kill microorganisms be used on potatoes. Freund⁷ points out that the extent of the sprouting problem depends on the growing area, storage requirements, and end use of the potatoes. Table stock is no good once it begins to sprout; indeed, sprouts contain the toxic alkaloid solanine. Bradley¹³ notes that over 80% of the Pacific Northwest crop goes to processors, and consequently there is little need there for sprout control.

Freund⁷ and Smith¹⁴ both estimate that sprouting inhibitors, whether radiation or chemical, could be used on 40 x 10⁶ to 50 x 10⁶ lb of potatoes annually.

Legislation (3)

In the United States, irradiation of food comes under the jurisdiction of the Food and Drug Administration (FDA), because radiation is defined by government regulations as a food additive. Clearances must therefore be obtained from FDA to market irradiated food. The use of radiation to inhibit sprouting of potatoes¹⁵ was approved by the FDA on July 8, 1964. (The order was dated June 30, and this date is sometimes cited¹⁰ as the approval date, but the order specifically states that it is effective as of the date of its publication in the Federal Register--- i.e., July 8.) The order approved the use of gamma rays from ⁶⁰Co to provide doses of 5 to 10 krad to potatoes. On October 10, 1964, use of ¹³⁷Cs to provide the same dose was approved.¹⁶ The approved maximum dose was later increased to 15 krad to accommodate designs of commercial irradiators.¹⁷ To process potatoes in economically feasible quantities would require the use of large pallet boxes, and to obtain at least an 8-krad dose in the center of the boxes would require a 15-krad dose to the outside.

Early Research (3)

Although the Russians cite very early work¹³ in the use of radiation to inhibit sprouting in potatoes, modern work is generally agreed to have begun with Sparrow and Christensen¹⁹ in 1954. Most of the early work grew out of mutation and radiation-effect studies. In 1950, Sparrow and Christensen²⁰ reported that potatoes irradiated* to 4800 r from an x-ray machine produced only 4% as much crop as control potatoes, and Hagberg and Nybom²¹ reported in 1954 that after a 10- or 20-kr dose from an x-ray machine sprouts already growing on potatoes died and did not reappear even when the potato was planted.

The research of Sparrow and Christensen¹⁹ was based on work with the Katahdin variety. Doses of 1.25, 5, 10, 20, 80, and 106 kr from a ⁶⁰Co source were used. Sprouting was reduced by the two smaller doses and was completely inhibited by doses of 20 kr or more. Potatoes given the 20-kr dose were still firm after 18 months of storage, and a taste panel found no undesirable tastes in any of the irradiated samples.

Shortly thereafter, Hannan,^{23,24} Brownell and Nehemias,²⁵ and Mikaelson and co-workers²⁶ all reported similar results. Hannan gave one-half a potato a 25-krep dose of 2-MeV cathode rays and compared that half with the untreated half; sprouting was inhibited, but some radiation damage occurred. Brownell's results²⁵ closely paralleled those of Sparrow and Christensen; a 20-krep dose inhibited sprouting. The Norwegian researchers²⁶ found no sprouting of potatoes stored 8 months after receiving doses from 5 to 20 krep. Control potatoes had a 9% weight loss, compared

*Throughout this review, the dose units used by the researchers have been retained. Conversion to a consistent unit is not justified because of the limitations on dosimetry, statistics, and other factors involved in dose measurement. In any case, the differences are relatively minor. The reader, therefore, will encounter r, kr, krep, and krad---whatever the researchers cited as dose units.---FEM

with a 4% loss of the irradiated potatoes.

The 1955 Brookhaven Conference (3)

At Brookhaven National Laboratory on May 25, 1955, the USAEC convened its researchers and contractors with members of the potato-growing and processing industry for the first large-scale meeting on this new application of radiation; almost 140 people attended.²⁷ The conference proceedings were not published, but a verbatim transcript²⁸ was made. Smith¹⁴ reviewed current problems and methods of storage. In the northern states, potatoes harvested in the fall keep until May or June because of low ambient temperatures. Smith acknowledged that temperatures of less than 45°F (7.2°C) lead to excessive sugar levels. High sugar level is undesirable in potatoes to be processed, especially for french fries and potato chips, because caramelization of the sugar causes the product to be dark brown. Chemical sprout inhibitors have been used; these include the methyl ester of naphthalene acetic acid, tetrachloronitrobenzene, maleic hydrazide, and chloro-isopropyl phenylcarbamate. Application costs of chemical inhibitors range from 4¢ to 7¢ per bushel.

Sparrow and Schairer²⁹ essentially reiterated the earlier report.¹⁹ Four different types of radiation were used: x rays from a machine, gamma rays from ⁶⁰Co, fast electrons from an accelerator, and fast neutrons from reactors. A 4800-r dose of x rays had about the same effect as 1200 rep of fast neutrons---a type of radiation not likely to be used commercially. Fast electrons even at 20 kr did not produce good results, presumably because even a little dirt or a flap of tissue over the eye would shield the sprout mechanism. Gamma rays and x rays gave the best results and an 8- to 10-kr dose was effective.

Sawyer, Dallyn, and Cotter³⁰ irradiated Green Mountain potatoes with the ⁶⁰Co source in the Brookhaven Gamma Garden; 10-kr or larger doses inhibited sprouting at any temperature. Potatoes stored at 50°C (10°C) were inhibited by a 7.5-kr dose. Katahdin variety potatoes were also tested, 1 year later, with similar results. These researchers pointed out that sprouting is a severe problem with potatoes grown on Long Island, because the temperature at harvest is high enough to break dormancy and to cause almost immediate sprouting.

Long Island potatoes are infested by the Golden Nematode, and Fassuliotis and Sparrow³¹ attempted to control this pest with irradiation. The LD₅₀ for the nematode was apparently 10 kr. This dose is also the usual one for sprout inhibition, so perhaps a solution to the nematode problem would be adoption of a radiation sprout inhibition program.

Effects of radiation on cooking quality, wholesomeness, and biochemistry of potatoes reported at the BNL conference will be reviewed in later articles. During a discussion period, Nielsen³² pointed out that one source of difference in response of different varieties of potatoes to radiation could easily be the variations in skin thicknesses of the varieties.

The papers comparing different sources of radiation and the presentations of various irradiator designs will be covered later. Rogers³³ made the point that ionization in the cell structure causes the desired effect and that the potato does not differentiate between radiation sources. For commercial treatment of bulk quantities, penetrating radiation will obviously be necessary. If only the skin and eyes of the potato are treated, electron-beam irradiation may be satisfactory. Most accelerators produce a small, narrow beam, however, which would have to be widened to treat quantities of potatoes. The usual method of widening the beam reduces the energy of the beam. Hatch and Regan³⁴ noted that the use of fission products, especially ¹³⁷Cs, might be stimulated by this possible application. Ballantine³⁵ posed a crucial question: would irradiation inhibition of sprouting cause the housewife to pay 2¢ more or 2¢ less per pound for potatoes? There were various replies from the attendees at this late discussion, but the general opinion was that the housewife would pay less.

Results of Sprouting Studies (3)

Radiation inhibition of potato sprouting has been studied in nearly all the major countries of the world. Although most of the literature^{covered} in this review is from the United States, results from Canada and 22 other countries are presented. Because the reader would surely find the repetition of "such-and-such variety irradiated at x dose and y dose rate, stored at whatever temperature and relative humidity for so long a time" seemingly endless, the results of the experiments are presented in the text, but the experimental conditions are presented in a capsule form:

Variety	Dose	T, °C	R.H.	Δt	t	Ref.
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where Variety is the variety of potato used in the experiment; Dose is the dose specified by the experimenter (presented in the units---krep, kr, or krad---he uses); T is the storage temperature in °C (converted from °F, if necessary); R.H. is relative humidity; Δt is the time between harvest and irradiation; t is the length of storage time; and Ref. is the reference where full details may be found. Not all these data are given in all cases, of course.

United States (4)

After Sparrow and Christensen¹⁹ and Brownell and co-workers²⁵ reported the results of their early studies, many others investigated this use of radiation. Sawyer and Dallyn³⁶ established that a 10-kr dose inhibited sprouting at several relatively high storage temperatures (Table 2). In a study³⁷ of both gamma radiation and chemical sprout inhibitors,

Green Mountain	1.25 to 40 kr	4.4 to 21.1°	30-40%	-	6 months	36
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both ⁶⁰Co and an accelerator were used. A 10-kr dose completely inhibited sprouting. Lower temperatures gave longer storage times. Varietal differences

Green Mountain Katahdin	2.5 to 12 kr	4.4, 10, 21.1°	-	-	10 months	37
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Table 2 Sprouting on Irradiated Potatoes after 6 Months of Storage
(From Ref. 36)

Storage temperature, °F(°C)	Average sprout length, inches					
	Control	Irradiation dose, kr				
		1.25	5	10	20	40
40(4.4)	1/8	1/8	None	None	None	None
50(10)	12	6	1	None	None	None
70(21.1)	30	30	30	None	None	None

Variety	Dose	T, °C	B.H.	Δ t	t	Ref.
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were investigated in four experiments by Sawyer and Dallyn,³⁸ with the results shown in Table 3 and Fig. 2. An early review of Pedersen³⁹ covered 32 refer-

Green Mountain Katahdin Cobbler Kennebec Saco Russet Burbank	5 to 20 kr	10 and 21.1°	-	-	8 months	38
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ences. Heiligman⁴⁰ noted that at high doses, some softening and shriveling occurred. A 5-krep dose retarded but did not inhibit sprouting. The 10-krep dose was sufficient to inhibit sprouting at 13°C, but some small sprouts did appear on the lot stored at 22°C. There were

Sebago Russet Rural	5 to 200 krep	13 and 22°	85-90%	-	-	40
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no sprouts on any lot irradiated at ^{doses of} 15-krep or greater. The weight loss from potatoes treated at doses larger than that needed for sprout inhibition was greater than that from controls; potatoes irradiated at doses at or near the minimum for sprout inhibition also showed higher weight losses than the control potatoes.

The group under Brownell at Michigan had made early studies⁴¹ with Russet Burbank variety potatoes. Tests conducted after 5 months showed no

Russet Burbank	7 to 28 krep	10°	50%	-	5 months	41
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sprouting in the irradiated potatoes. A subsequent report⁴² summarized the experiments. Again, 10 krep seemed to be the best dose. After 5 months, lots that had received this dose were 86 to 98% useable. Those that received doses

Russet Burbank Russet Rural Sebago Katahdin	5 to 200 krep	1.7 to 26.7°	66-90%	-	5 months	42
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greater than 5 krep did not sprout. Weight loss increased with increase in storage temperature; because of sprouting, controls lost more than did irradiated potatoes. Weight loss decreased with increase in ^{weight} relative humidity; again, controls lost more _^ than did irradiated potatoes.

Table 3 Loss of Potatoes After 8 Months of / ^{Storage} (From Ref. 38)

Irradiation dose, kr	Katahdin		Green Mountain		Kennebec		Russet Burbank	
	Sprouting*	Shrinkage**	Sprouting	Shrinkage	Sprouting	Shrinkage	Sprouting	Shrinkage
0	31.4	10.0	120.4	8.8	38.3	12.2	62.0	12.8
5	10.6	7.5	32.5	13.4	30.5	12.2	37.2	12.8
7.5	2.5	9.9	13.6	10.5	26.2	12.2	35.1	10.0
10	0.8	8.2	3.2	10.8	10.5	11.8	22.8	8.1
12.5	0.5	4.4	-	9.1	3.5	12.0	6.0	6.1
15	0.1	6.8	-	8.3	0.6	9.9	0.1	5.4

* Grams of sprouts per kilogram of potato
 ** Percent of original weight

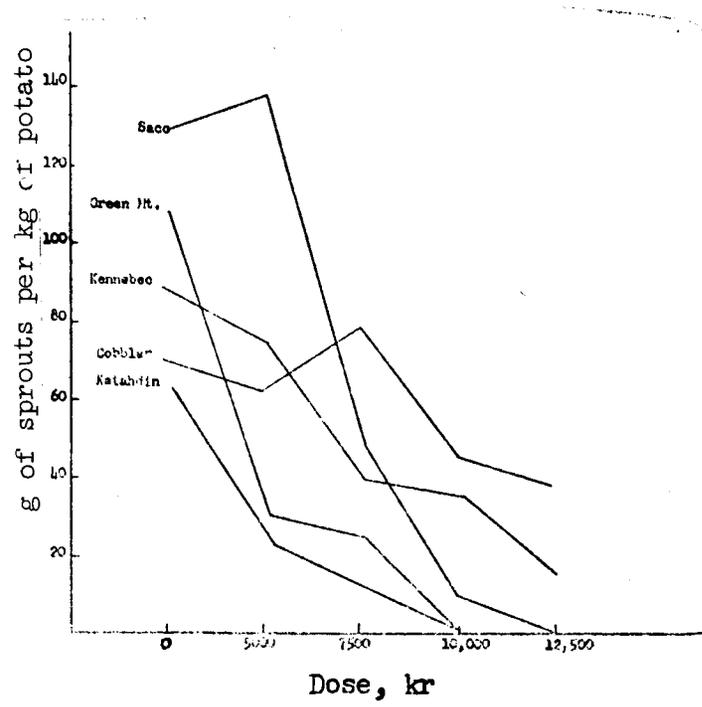


Figure 2 Variety--dose relationship for potatoes stored at 50°F (10°C) for 8 months

Variety	Dose	T, °C	R.H.	Δt	t	Ref.
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In another experiment, the weight loss of Sebago potatoes increased as the irradiation dose increased, a result which indicates that the sprout inhibition mechanism is simply one of mechanical damage rather than of cell destruction and reduced metabolism. When sprouts were irradiated, their growth ceased. When the potatoes were irradiated and the sprouts were shielded, sprout growth was not affected. The optimum conditions, reported by Brownell and associates,⁴³ are a dose of 10 krep, T of 7.2°C, and 85 to 90% R.H.

Russet Burbank Russet Rural Sebago Katahdin	5 to 200 krep	1.7 to 26.7°	60-90%	-	5 months	43
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Ellis and co-workers⁴⁴ undertook to determine the optimum dose, curing schedule, time of irradiation, and storage temperature that are required to preserve four varieties for 18 months. The curing schedule was storage at

White Sebago Red Pontiac Katahdin Russet Burbank	5 to 15 krep	5, 8.3, 12.8°	-	1, 2, or 3 months after harvest	2 years	44
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21°C and high R.H. for 7 days; potatoes were cured before irradiation, or not cured. cured after irradiation, cured both before and after irradiation, / Dose and curing schedule were investigated at 5 and 12.8°C in one phase of the study, and optimum time to irradiate (with storage at 8.3°) in another. Ellis and associates intended to irradiate potatoes immediately at three stages--- after harvest, at the midpoint of dormancy, and at the end of dormancy; however, the four varieties were obtain from four different locations in the United States at four relatively different harvest times. Therefore, three arbitrary irradiation times were chosen: Dec. 4, Jan. 3, and Jan 30.

Variety	Dose	T, °C	R.H.	Δt	t	Ref.
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or White Sebago, a 5-krep dose prevented sprouting at 5°C, but a 7.5-krep dose was necessary at 12.8°C. At the lower storage temperature, curing both before and after irradiation gave the best results, i.e., the lowest weight loss. At the higher temperature, curing before irradiation was sufficient. At 17 months, the doubly-cured potatoes stored at the lower temperature still appeared normal. There was little observable effect of the length of time before irradiating, although the potatoes irradiated Jan. 30 had a slightly greater weight loss than the potatoes irradiated earlier.

The Red Pontiac variety required a 7.5-krep dose at the lower temperature and a 12.5-krep dose at the higher temperature to prevent sprouting. No curing effect was noted; no time-of-irradiation effect was observed. Appearance of the 7.5-krep-treated potatoes after 17 months at 5°C was excellent; however, decay was excessive in potatoes stored at 12.8°C for 10 months.

The Katahdin variety, like the Sebago variety, required a 7.5-krep dose to prevent sprouting when stored at 12.8°C, although a 5-krep dose inhibited sprouting at 5°C. Time of irradiation after harvest did not affect the results; neither did curing.

A 5-krep dose inhibited sprouting of Russet Burbank potatoes stored at 5°C and markedly reduced sprouting of those stored at 12.8°C. At the lower temperature, potatoes cured before irradiation had less weight loss than the other potatoes, but this effect was not noted at the higher storage temperature. This variety stores well, and the appearance of some irradiated potatoes stored at 5°C was still excellent slightly over 2^{years} after harvest. Time of irradiation had some significance; potatoes irradiated Jan. 4 after harvest on Oct. 17 lost weight more slowly than did the other potatoes.

Ellis and associates⁴⁴ conclude that, for all the varieties except Red Pontiac, storage at 12.8°C results in the greatest increase in shelf life

Variety	Dose	T, °C	R.H.	Δ t	t	Ref.
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(Table 4). For all four varieties, a dose increase was required to inhibit sprouting at the higher storage temperature.

Emerson and co-workers⁴⁵⁻⁴⁷ studied the effects of radiation on many

Arenac	5 and 7.5 krads	21.1°	35%	-	6 weeks	45
Ontario					55 days	46
Sebago						

fruits and vegetables. Under conditions designed to accelerate sprouting^{of} potatoes, a 7.5-krad or greater was needed to inhibit sprouting. The controls showed a weight loss of 24.8% for Arenac and 23% for Ontario after 55 days; the 7.5-krad-treated potatoes lost 16.3 and 20.5%, respectively.

A number of experiments were conducted on Idaho-grown Russet Burbank potatoes by Sparks and Iritani.⁴⁸ Potatoes were irradiated in 10-lb sacks

Russet Burbank	5 to 80 krads	common, 3.3, 10, and 21°	-	2-wk intervals up to 34 weeks	1 year	48
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lowered between spent fuel elements from reactors. In 5 years of experiments, no potato tested immediately after irradiation showed any induced radio-activity whatsoever. Table 5 gives results of early experiments, and Fig. 3 summarizes the loss data at all doses and temperatures. Any potato with excessive sprouting, withering, or shriveling was discarded. Sparks and Iritani justify using the mean of the dose data in Fig. 3 by pointing out that time of irradiation had a much greater effect than did dose variation. At 3.3°C, controls lost less weight than did irradiated potatoes; but at higher temperatures, controls were the only potatoes that were completely discarded after 333 days of storage. In the 1958-59 experiments, minimum dose was studied; no data were collected for more than 6 months. No treatment

Table 4 Storage-life increase of potatoes caused by radiation (From Ref. 44)

Variety	Storage temperature, °F(°C)	Storage life from harvest, days		Extension, days	Extension, percent
		Controls	Irradiated		
White Sebago	55(12.8)	90	325	235	261
	47(8.3)	170	385	215	126
	41(5)	330	415	85	26
Katahdin	55(12.8)	115	365	250	217
	47(8.3)	225	410	185	82
	41(5)	410	495	85	21
Red Pontiac	55(12.8)	130	325	195	150
	47(8.3)	175	450	275	157
	41(5)	300	625	325	108
Russet Burbank	55(12.8)	140	335	195	139
	47(8.3)	210	410	200	95
	41(5)	400	650	250	63

Table 5 Loss Data on Russet Burbank Potatoes 312 Days After Harvest
After Being Kept in Common Storage* (From Ref. 48)

Days after harvest	Percent Loss						Mean
	Controls	Irradiation, krep					
		5	10	20	40	80	
1	100	49	52	65	100	100	73
14	100	75	87	59	100	100	84
28	64	9	20	26	52	78	37
42	59	17	21	12	28	71	30
56	61	16	20	56	27	58	35
70	62	18	12	17	13	31	18
84	59	11	20	15	14	22	16
98	61	11	18	26	14	20	18
112	64	10	14	14	14	23	15
126	61	10	9	19	16	10	13
140	57	9	9	14	13	14	12
154	58	15	11	5	13	21	13
168	57	15	8	7	19	15	13
182	56	14	13	4	16	15	12
196	56	4	10	8	12	23	12
210	54	9	5	7	10	29	12
224	53	9	7	8	17	37	16
238	53	10	21	20	24	33	20
	Mean 63	18	20	21	28	30	

* Cooled by ambient air only.

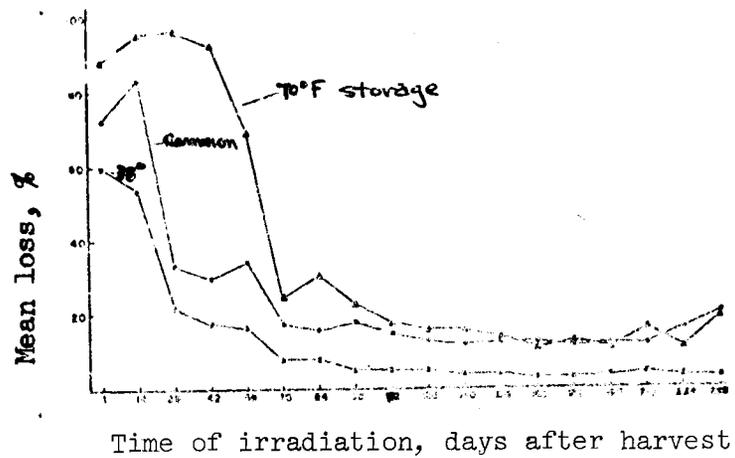


Figure 3 Loss data on irradiated Russet Burbank potatoes under three storage conditions, 312 days after harvest (from Ref. 48).

Variety	Dose	T, °C	R.H.	Δt	t	Ref.
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of less than 5 krep prevented sprouting, and 10 krep was concluded to be an optimum dose. Storage at 10°C gave better results than storage at higher temperatures. Results of the 1959-60 experiments confirmed that a long delay after harvest before irradiation gives best shelf life. A constant storage temperature (10°C) gave better results than common storage. There was little dose effect among potatoes stored at low temperatures, but 5- or 10-krep treated potatoes stored better than other potatoes at intermediate storage temperatures, and potatoes given a 10-krep dose kept best at 21°C.

The review of Freund⁷ has already been mentioned. He concluded that a 7- to 10-krad dose should inhibit sprouting under any commercially practical storage conditions; his later addendum report⁴⁹ did not differ. Freund summarizes known varietal differences (Table 6). Although researchers have

Table 6. Sprout Inhibition Doses Required for Commercial Varieties of Potatoes

Variety	Dose, krads
Katahdin	5 to 10
Russet Burbank	5 to 15
Red Pontiac	7.5 to 12.5
Sebago	5 to 10
Kennebec	8 to 12.5
Irish Cobbler	8 to 12.5

investigated storage periods of up to 2 years, Freund points out that anyone storing potatoes for more than 1 year is essentially competing with himself during ^{the} second year. His review also contains a summary of the commercially significant qualities of these six most important varieties.

The most recent results¹³ in the United States come from the demonstration tour of the Portable Cesium Irradiator.^{50, 51} Prosser Packers, Inc., Prosser,

Russet Burbank	6.2-8.5, 7.6-10.8, 9.7-11.9 krads	common, 2.2, 7.2 ⁰	-	2 days	182 days 211 days	13
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Variety	Dose	T, °C	R.H.	Δt	t	Ref.
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Washington, and Rogers Walla Walla, Inc., Walla Walla, Washington, evaluated irradiated potatoes and reported no sprouting 182 days and 211 days, respectively, after irradiation in any lots regardless of storage conditions (Figs. 4 and 5). Irradiated potatoes were shipped to Honolulu from Seattle; no sprouting occurred. Potatoes sent to Chicago by railroad also showed no sprouting.

Canada (4)

Of the Western nations, Canada has come closest to commercializing the radiation treatment of potatoes. Its research has been detailed; a demonstration program in 1961-62 was very successful, and a commercial plant to irradiate potatoes and other products was opened. Unfortunately, the plant shut down after 1 year, because the potato crop failed and there were no potatoes to be irradiated. Typical sprout inhibition results are shown in Fig. 6.

Results of the first large-scale experiments were reported⁵² in 1960.

Sprouting results were covered by Parks.⁵³ Controls of both varieties

Katahdin Netted Gem (Russet Burbank)	8 to 15 krep	4.4 and 20°	-	-	11 months	52
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stored at the higher temperature were well sprouted at 4.5 months of storage; control potatoes at the lower temperature showed short sprouts at 7 months. No irradiated potatoes had sprouted after 11 months of storage, although Netted Gem potatoes were in better condition than the Katahdin potatoes. Throughout the experiment, control potatoes at the lower temperature showed only slightly more weight loss than did the irradiated potatoes. At the higher storage temperature, control potatoes were discarded after 8 months; irradiated potatoes showed only 9% weight loss at that time. These results clearly justified further study of the problem. Errington and MacQueen⁵⁴



Figure 4 Storage of Washington State potatoes after 180 days at 45°F (7.2°C) and 95% R.H. (From Ref. 13, courtesy of L. B. Bradley, Office of Nuclear Energy Development, Washington State).



Figure 5 Storage of Washington State potatoes after 211 days under common storage conditions (From Ref. 13, courtesy of L. B. Bradley, Office of Nuclear Energy Development, Washington State).



Figure 6

Canadian
Katahdin potatoes treated as part of the pilot scale demonstration
program. Doses range up to 7.7 krad. These potatoes were stored
5.5 months at -12.8°C. (Photo courtesy of Atomic Energy of Canada Ltd.)

Variety	Dose	T, °C	Δt	t	Ref.
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gave a popular-level presentation in which they stated that later research had shown that Katahdin and Russet Burbank varieties can be stored 8 months at 12.8°C after an 8-krad dose. They also reported a further experiment which showed that an 8-krad dose inhibits sprouting in each of 10 varieties of potatoes for at least 8 months under the heated storage conditions necessary in Canada during the winter. This article⁵⁴ and a subsequent article⁵⁵ gave preliminary details on the pilot scale potato irradiation demonstration program.

The pilot scale program⁵⁶ was carried out in Canada's four eastern provinces in 1961-62; 26 members of industry cooperated. A mobile irradiator with an 18-kCi ⁶⁰Co source was used; an 8-krad dose could be applied to 2200 lb of potatoes per hour. Each industrial participant provided his own potatoes,

Katahdin Kennebec Cherokee Sebago Netted Gem Irish Cobbler	8 krads	up to 26.7°	-	-	up to 10 months	56
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which were cured before irradiation and were stored with controls under identical conditions after irradiation. The irradiated potatoes could be sold to the public, in whichever form the participants chose, i.e., processed or whole. Kennebec, one of the six varieties tested, is a good chipping variety that is grown in three of the provinces. Sebago is the variety grown mainly on Prince Edward Island. Storage was in pallet boxes, bins, barrels, and bags. Eight of the 26 participants stored potatoes at high temperatures to minimize sugar content; the other participants placed potatoes in common storage. Sprouting was completely controlled in all cases. A field run was made in which special care was taken with handling to show that where there was no mechanical injury, curing potatoes before irradiation was unnecessary. MacQueen,⁵⁷ reporting on the entire Canadian food irradiation program to an international symposium in 1964, stated that irradiation did not affect stored potatoes as much as differences in varieties and storage conditions. He also said that in Canada special labeling of marketed irradiated

Variety	Dose	T, °C	R.H.	Δt	t	Ref.
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potatoes is not required; this practice was thought to be fair, because the use of chemical sprout inhibitors also does not require special labeling. Irradiation can best be used as a sprout inhibition treatment when potatoes are cured, irradiated, and stored all in the same 4-ft³ pallet box. Use of a pallet box greatly reduces injuries caused by excessive handling, offers greater flexibility in storage, and makes temperature and humidity control easier.

Australia (4)

Wills⁵⁸ irradiated Tasmanian potatoes in 1961 and 1962, using spent fuel

Up-to-Date Kennebec Sebago Sequoia	5 to 16 krads	7.2 and 20°	-	10 weeks	6 months	58
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elements the first year and ⁶⁰Co the second year. A 5.5-krad dose inhibited sprouting of the Up-to-Date variety for 33 weeks; a 10-krad dose inhibited sprouting of Kennebec and Sebago varieties for 25 weeks. In 1962, the potatoes had begun sprouting by the time they were irradiated, but a 5-krad dose stopped this sprouting. Wills notes that these doses are lower than those reported elsewhere.

Belgium (4)

Results from the Institut National pour l'Amelioration des Conservees de Legumes show that radiation inhibits sprouting of the Bintje variety.⁵⁹ However, the results were not consistent, probably because of nonuniformity of dose. The IRMA irradiator⁶⁰ with a ¹³⁷Cs source was used to provide a 10-krad dose to potatoes that had been cured for 6 weeks at 8°C.

Colombia (4)

Potato production in Colombia is ~1.2 x 10⁹ lb annually; spoilage ruins

Tosarrena Pardo Pastusa	5 to 15 krads	10°	-	-	8 months	61
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~20%. In this experiment, control potatoes had to be discarded, but potatoes⁶¹ irradiated at 7.5-krad or greater doses were in good condition.

Variety	Dose	T, °C	R.H.	Δt	t	Ref.
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Denmark (4)

Truelsen⁶² reported two experiments that had to be terminated when both

Bintje	6 to 12 krads	5, 10, 15°	-	1, 2, 4, and 6 months	8 months	62
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irradiated and control potatoes became discolored. Although slightly higher doses were required as time between harvest and irradiation increased, 8 krads was an adequate dose in each case.

The literature review by Skou⁶ (49 references) states that all doses greater than 2 krads have some inhibiting effect, but doses greater than 8 krads are required to completely inhibit sprouting.

Egypt (4)

Abdel-al⁶³ reported that the crop harvested in May and June sometimes starts sprouting immediately because of the high temperatures in the Near East. All potatoes in this experiment had begun sprouting before they were

Alpha	8 to 14 krads	common (30-35°)	-	-	6 months	63
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irradiated. After 6 months, the weight of sprouts per 100 g of potatoes was 9.25 g for the unirradiated controls but only 0.14 g for the 14-krad-treated potatoes.

England (4)

In England and Wales, losses to sprouting and enhanced water loss were probably ~30 kilotons/year/ ⁱⁿ the early 1950's, according to Burton and Hannan.⁶⁴

Dr. McIntosh	2 to 35 krads	10°	78%	5 days, 55 days, 6 months	15 months	64
Arran Consul Craig's Defiance Golder Wonder Home Guard	8.5 krads	10°	78%	3 to 4.5 months	15 months	64

Variety	Dose	T, °C	R.H.	Δt	t	Ref.
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Extensive tests were undertaken, particularly with the Dr. McIntosh variety. Burton and Hannan concluded that a 3.5-krad dose gives a commercially acceptable result; this experiment and results have not been duplicated by others. Some sprouting occurred at all doses less than 8.5 krads, but the sprouts were very small---2 mm long. Larger doses killed existing sprouts and completely prevented additional sprouting. With varieties other than Dr. McIntosh, the 8.5-krad dose did not kill sprouts already well started. Burton and deJong⁶⁵ state that inhibition varies widely with the variety and state of the potato and with the kind of storage. A 10-krad dose of $\frac{500\text{-keV}}$ electrons caused no inhibition, but 11 krads from $\frac{200\text{-keV}}$ x rays gave complete suppression. Gamma radiation is effective at 10-krad or smaller doses. The net result of irradiation---inhibition of sprouting---does not prolong storage indefinitely.

France (4)

Vidal⁶⁶ has stated that a 5- to 6-krad dose is satisfactory for short-term storage, but a 7.5- to 10-krad dose is necessary for storage of 12 months or

Saskia Bintje	5 to 15 krads	-	-	-	12 months	67
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Bintje Kerpondy	-	10-12°	high	-	1.5 years	68
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more. In a later experiment, Vidal⁶⁷ claimed that 7.5 krads was almost completely effective, but he recommended 10 krads for industrial use. Sandret and Michiels⁶⁸ achieved lengthy storage under ideal conditions. Losses in irradiated potatoes are usually ~ 20% less than that of controls.

Germany, East (4)

Buhr and co-workers⁶⁹ irradiated 19 varieties of potatoes and found that

See Table 7	4 to 16 krads	4 to 18°	-	-	15 months	69
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an 8-krad dose gave complete sprout inhibition. Table 7 presents an evaluation

Variety	Dose	T, °C	R.H.	Δt	t	Ref.
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of the potatoes after 15 months of storage; the dose used was 16 krads.

Germany, West (4)

In one of his ^{early} / food irradiation articles, Kuprianoff⁷⁰ mentions radiation sprout inhibition. Patzold and Kolb,⁷¹ early German researchers,

Vera Bona Ackersegen	0.25 to 16 kr	2 to 14°	-	-	6 months	71
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found that a

^ 4-kr dose inhibited but did not stop sprouting in the Bona variety; 8-kr and 16-kr doses stopped sprouting entirely, even after it had begun. On Vera potatoes, weight loss ^{from} / unirradiated controls was 16.5%, but potatoes receiving an 8-kr dose only lost 0.6%. For the Bona potatoes, the losses were 6.6% for controls and 0.4% for irradiated specimens. For the Ackersegen variety, controls lost 5.1% and irradiated potatoes lost 0.3%. Patzold and Weiss⁷² stated that an 8-kr dose of gamma radiation from ⁶⁰Co gave lasting sprout inhibition and reduction of weight loss.

Gantzer and Heilinger⁷³ used an x-ray machine to inhibit sprouting in potatoes, and Berger and Wolf⁷⁴ suppressed sprouting with radiation from a Betatron. Hansen and Grunewald⁷⁵ compared inhibiting doses for 14 varieties

Corona Feldeslohn	4 kr	-	-	-	-	73
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Heida	1 to 60 kr	-	-	-	30 weeks	74
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(Table 8).

of potatoes. ^ More recent results from Scheid and Heilinger⁷⁶ confirmed the finding by Hansen and Grunewald that the Maritta variety is most difficult to prevent from sprouting.

See Table 8	4 to 15 kr	15°	-	-	11 weeks	75
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Maritta	3.2 kr	20°	-	-	10 days	76
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Table 7. Evaluation of 19 potato varieties after a 16-krad dose and storage for 15 months (Ref. 69).

Variety	Appearance	Variety	Appearance
Pirat	very good	Ora	satisfactory
Amsel	very good	Sperber	satisfactory
Li. 4987/54	good to very good	Sagitta	satisfactory
Gunosa	good	Kastor	poor
Stieglitz	good	Apollo	poor
Meise	good	Spekula	poor
Drossel	good	Pollux	poor
Fink	good	Schwalbe	very poor, rotten
Gerlinde	satisfactory	Spatz	very poor, rotten
Rotkehlchen	satisfactory		

Table 8. Sprouting results on 14 potato varieties stored 11 weeks at 15°C (Ref. 75).

Variety	Percent sprouted at dose indicated			
	4 kr	8 kr	12 kr	15 kr
Anco	100	30	-	-
Arensa	5	-	-	-
Bona	5	-	-	-
Carmen	50	10	-	-
Cosima	30	-	-	-
Datura	90	40	10	-
Delos	100	20	-	-
Feldeslohn	60	-	-	-
Grata	70	-	-	-
Heida	70	20	-	-
Heiko	80	10	-	-
Isola	70	5	-	-
Lori	30	-	-	-
Maritta	100	80	30	5

Variety	Dose	T, °C	R.H.	Δt	t	Ref.
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India (4)

Mathur⁷⁷ concluded that a 6-krad dose was optimum for Gola potatoes and

Gola Up-to-Date	3 to 9 krads	-	-	2 weeks	4 months	77
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a 9-krad dose for Up-to-Date potatoes. Mathur and Lewis⁷⁸ established a 6-krad dose as optimum for the Phulwa variety. Mathur has also studied the effect of

Phulwa	6 krads	12° and 21 to 35°	85-90% 57-90%	-	10 months. 5 months	78
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time after harvest before irradiation,^{and} he concluded that the nearer the potato is to the end of its dormancy period, the greater is the effect of radiation. Recently, Mathur⁷⁹ published results of studies with Up-to-Date seed potatoes. He attempted to extend their storage life by using a chemical treatment with correct time-of-irradiation and a dose that produced an effect that could be reversed with chemicals. Mathur established five periods of development

Up-to-Date	3.5 krads	9°	85-90%	up to 16 weeks	up to 44 weeks	79
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during storage of potatoes: (1) predormancy maximum (time of harvest), (2) predormancy minimum (3.5 to 4.5 weeks after harvest), (3) dormancy (8.5 to 9.5 weeks), (4) presprouting minimum (13.5 to 14.5 weeks), and (5) sprouting (15.5 to 16.5 weeks). He concluded that a 3.5-krad dose given at the predormancy minimum stage and followed by a 50-minute dip in a 500-ppm solution of the methyl ester of indolyl-3-acetic acid^{would} result in a 44-week storage life for Up-to-Date seed potatoes (Table 9).

Dharkar⁸⁰ obtained a storage life of up to 6.5 months for Indian potatoes.

Phulwa Up-to-Date	6 and 9 krads	room T 10-12°	-	-	1.5 months 6.5 months	80
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Dharkar and Sreenivasan,⁸¹ reviewing India's food irradiation program at an international conference, stated that a 10-krad dose inhibits sprouting for 32 weeks at room temperature; unirradiated controls sprout at 8 weeks.

Table 9. Combined effect of radiation and chemical treatment on
Up-to-Date seed potatoes stored at 9°C and 85-90% R.H. (Ref. 79).

Developmental stage	Irradiation treatment	Post-irradiation IAA treatment ^a	Mean storage life, weeks ^b
Predormancy maximum	None	None	17.0
	3.5 krads	None	12.5
	3.5 krads	Yes	22.0
Predormancy minimum	3.5 krads	None	25.5
	3.5 krads	Yes	44.0
Dormancy	3.5 krads	None	23.0
	3.5 krads	Yes	31.5
Presprouting minimum	3.5 krads	None	19.0
	3.5 krads	Yes	24.5

a

A 50-minute dip in a 500-ppm solution of the methyl ester of indolyl-3-acetic acid.

b

Including the pre-irradiation period.

Variety	Dose	T, °C	R.H.	Δt	t	Ref.
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Israel (4)

A preliminary report⁸² of Israeli experiments on radiation inhibition of potato sprouting appeared in 1966, and full details were presented at two international conferences later that same year.⁸³⁻⁸⁴ Because there are two crops each year for Mid-East countries, storage for more than 6 months is not needed. However, storage at ambient temperatures, which are high, leads to rapid sprouting. Most potatoes are stored at 4°C in relatively expensive refrigerated warehouses. The Up-to-Date variety accounts for 70% of Israel's crop.

Up-to-Date	6 to 14 krads	ambient, 4, 8, 14°	-	8 weeks	up to 12 months	84
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At 24-week storage, controls at all temperatures were sprouting; 4 weeks after withdrawal from storage, all controls were 100% sprouted. The 10-krad dose reduced initial sprouting to 5 and 15% at 4 and 14°C, respectively, with almost no growth of sprouts during the ensuing 4 weeks. Under ambient conditions, the 10- and 14-krad-treated lots were in good condition and were organoleptically acceptable after 24 weeks.

Italy (4)

The Italian potato crop is $\sim 4 \times 10^6$ tons/year; per capita consumption is ~ 50 kg/year (110 lb/year). Denti⁸⁵ has reviewed the possibilities for irradiation preservation of potatoes, but no work has been reported.

Japan (4)

Ogata and co-workers⁸⁶ found that irradiation at the beginning of dormancy

Irish Cobbler	3 to 12 kr	room T	-	-	-	86
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did not prevent sprouting, although sprouts on potatoes irradiated at 7 and 12 kr did not grow further. The 12-kr treatment prevented sprouting on potatoes irradiated at the end of dormancy. A review⁸⁷ of Japanese food irradiation studies recommended a 7-krad dose at the beginning of storage for the best results.

Variety	Dose	T, °C	R.H.	Δt	t	Ref.
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New Zealand (4)

Unsatisfactory results were reported by McNaughton,⁸⁸ who suggested that

Ilham Hardy Chippewa	7 to 20 krep	21.1°	-	6 months	10 weeks	88
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excessive time between harvest and irradiation was the cause.

Norway (4)

Mikaelsen and co-workers²⁶ did some of the earliest work on radiation inhibition of sprouting of potatoes. Later, they compared the effect of the chemical Fusarex, a tetrachloronitrobenzene compound, with radiation doses from a reactor.⁸⁹ Sprouts had already appeared on some potatoes; these died

-	5 to 40 krads	4 to 6°	-	-	12 months	89
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immediately after 10-krad or greater doses. Potatoes irradiated at doses of more than 5 krads showed only a few primordial sprouts after 12 months.

Kerr's Pink	5 to 15 krep	7° avg. 14° avg.	-	-	15 months	90
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Mikaelsen and associates⁹⁰ found that a 5-krep dose was not as effective at high temperatures as a larger dose, but 5 krep did give satisfactory results at low temperatures. After 12 months of storage, 15-krep-treated potatoes lost the least weight; after 15 months, these were the only potatoes still suitable for taste-panel testing, although they were very sweet.

Pakistan (4)

Farooqi and co-workers⁹¹ reported that controls lost 31.2% weight after

Ultimas Holland	2 to 10 krads	28.3° avg.	69% avg.	15 days	210 days	91
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150 days, but 10-krad-treated potatoes lost only 14.3%. After 210 days, only two potatoes of the 10-krad lot had to be discarded---a loss of 0.1%.

Variety	Dose	T, °C	R.H.	Δt	t	Ref.
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Portugal (4)

The main variety grown in Portugal is Arran Banner; farmers grow it because of its high yield, but it keeps poorly. Preliminary experiments⁹² with this

Arran Banner Condea Anco Claudia Arran Consul Desiree	10 krad	-	-	-	4 months	92
	variety and others showed that a 10-krad dose stops sprouting after it has begun. In a later experiment with Arran Banner,					
Arran Banner	5 to 15 krads	18-20°	70-80%	-	-	92

a 5-krad dose allowed only slight sprouting; higher doses completely inhibited sprouting.

Variety	Dose	T, °C	R.H.	Δt	t	Ref.
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South Korea (4)

Park and associates⁹³ reported that, ^{sprouting of} 7.5-krad-treated potatoes was inhibited at a low storage temperature, but a 9-krad dose was required to inhibit sprouting at a high storage temperature

Irish Cobbler Saco Tachibana Wai'ba Kennebec Shimabara	4.5 to 9 krads	low high	-	1 wk	-	93
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Spain (4)

In a general review of food irradiation projects, Fernandez y Gonzalez⁹⁴ reported satisfactory sprout inhibition. Garcia de Mateos Lopez and associated⁹⁵

Urgenta	5 krads	19 to 31°	60-80%	-	150 days	94
Alava Gineke	5 to 15 krads	10 to 25°	40-87%	-	9 months	95

chose a rapid-sprouting variety to thoroughly test irradiation. A dose rate of 520 krad/hr was used. Of the controls, 80% sprouted after 20 days and 100% after 3 months. Doses greater than 5 krads totally inhibited sprouting during the 9 months of the experiment. The dose recommended for industrial practice was 8 krads.

Sweden (4)

Early results of Jaarma⁹⁶ showed the effectiveness of the method. Later,

Eva Bintje President	16 krads	5°	-	-	-	96
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Jaarma⁹⁷ reported a study of several varieties; different dose rates from a ⁶⁰Co source and from an x-ray machine were used. Inhibition

Bintje Early Puritan Eva President Primula Ulster Chieftain	10 to 20 krads	-	-	1-month intervals	-	97
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of sprouting is most pronounced if the potatoes are irradiated immediately after harvest, although there were considerable varietal differences. Larger doses are needed as time after harvest

Variety	Dose	T, °C	R.H.	Δt	t	Ref.
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increases. The results obtained with the x-ray machine are considered to be preliminary, because the machine could not be used in any practical application. A dose of 15 krads with soft x-rays did satisfactorily inhibit sprouting, however. USSR (4)

At a USSR national conference in 1957, Metlitskii and co-workers⁹⁸ reported that a 10-krad dose inhibits sprouting in the Lorkh variety, but other varieties require only 8 krads. After 1 year, total loss and wastage was only 10%. Rakitin and Krylov⁹⁹ reported results of studies using an

Berlikhingen	10 or 40 krads	-	-	-	8 months	99
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x-ray machine. The 10-krad-treated potatoes did not sprout and had a weight loss of only 11.8%; control potatoes were well-sprouted and had a total weight loss of 72.0%.

In 1959, Rubin and associates⁸ published a comprehensive review of the use of ionizing radiation to inhibit sprouting of potatoes; they cite 113 references, of which 59 are to Russian work. Although 10-krad-treated potatoes will not sprout, this is not necessarily the optimum dose. The minimum dose necessary to maintain resistance to microorganisms while preserving the nutritional value should be used. Lorkh potatoes irradiated at 8 kr had weak shoots after 12

Lorkh Berlikhingen Priekul'skii Epron Seyanets 9729/Ukhtomskii Rannyaya roza Moskovskii Peredovik	2 to 10 kr	-	-	various	12 months	8
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months of storage; complete inhibition was obtained with 10 kr. The effect of radiation is greater at different stages of storage; potatoes irradiated Dec. 1 and planted April 27 sprouted 36 days later, but potatoes from the same harvest that were not irradiated until April 15 and planted April 27 did not

Variety	Dose	T, °C	R.H.	t	t	Ref.
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sprout until 53 days later. Maximum sensitivity to radiation does not occur at the same time in all varieties. The Moskovskii potatoes showed greatest sensitivity during, rather than at the end of, dormancy. Epron and Peredovik varieties were more sensitive during dormancy, but Priekul'skii and Berlikhingen were more sensitive when the eyes sprouted. Some varieties are more sensitive than other varieties regardless of stage of dormancy. The Ranryaya roza, Moskovskii, Peredovik, and Lorkh varieties require lower doses to inhibit sprouting than do the Epron or Berlikhingen varieties.

Rubin and Metlitskii¹⁰⁰ stated that the recommended dose is 8 kr, but Krushchev and associates¹⁰¹ stated that 10 krads is the usual dose. Krushchev and associates also pointed out that the dose needed can vary with the time of use of the potatoes. Potatoes to be used in April should be given a 3-krad dose in October; potatoes to be used in June should be given a larger dose in November.

Krushchev and co-workers¹⁰¹ cite Metlitskii's work¹⁸ as being the world's first on radiation sprout inhibition of potatoes, but Rubin and associates⁸ imply that Metlitskii's work involved only onions. However, Metlitskii was a co-author of both papers; the confusion may be in the translations.

Work with ⁶⁰Co and with 620-MeV protons was reported by Shaidorov and co-workers.¹⁰² The inhibitory effect was stronger when radiation was directed onto the eyes than onto the whole tuber; the effect of protons was much stronger than that of gamma rays.

Lorkh	5 to 15 krads	-	-	2 weeks, 5 months, 7 months	13 months	103
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Metlitskii and others¹⁰³ found that irradiation 2 weeks after harvest gave excellent storage results; some of the potatoes used in their experiment were eaten as long as 13 months after irradiation.

Variety	Dose	T, °C	R.H.	At	t	Ref.
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Venezuela (4)

Solanas and Darder¹⁰⁴⁻¹⁰⁶ found that a 5-krad dose is sufficient to inhibit

Katahdin	1.25 to 20 krads	10-22°	60-95%	-	8 months	106
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sprouting for short-term storage. These authors point out that determination of the optimum dose is important, because the cost of treatment depends on the dose required to inhibit sprouting.

Other Countries (4)

Poland and the Union of South Africa are both reported to have programs on potato irradiation.^{5,10} However, no actual references to work in either country were found in the available literature. A 1963 Polish book¹⁰⁷ on food irradiation, which contains several pages on sprout inhibition of potatoes, cites 30 references on potato irradiation--but not one to Polish work.

South American countries other than Colombia and Venezuela are known to be studying potato sprout inhibition,¹⁰⁸ with the general result given that an 8- to 10-krad dose will inhibit sprouting for 8 months.

Summary of Results (4)

The results obtained by the researchers cited above would indicate that, in general, the best results of irradiation of potatoes are obtained when top quality potatoes are dug, cured for a period of 2 to 10 weeks, and irradiated to a dose of 8- to 10-krads. If possible, the potatoes should be cured, irradiated, and stored all in the same container---to minimize handling damage and handling costs. Where care is taken to avoid cuts and bruises during harvest, curing may not be required. The temperature of storage is governed more by the intended use of the potatoes than by any other criterion, but the lower the temperature the longer the storage that can be expected.

Results cited in the above sections that are particularly at odds with this summary would include Wills,⁵⁸ Burton and Hannan,⁶⁴ Ogata et al.,⁸⁶ and Jaarma.⁹⁷ Wills⁵⁸ admits that good results were obtained at doses lower than those usually used, but no explanation for this variation is given. The other three references are all to early work in this subject, when differing results could be expected.

Effect of Dose Rate (3)

Several researchers have observed that a dose delivered at a high rate is more effective than the same dose delivered at a low rate. Jaarma⁹⁷ reported that in three of six potato varieties a dose rate of 600 krad/hr was more effective than a dose rate of 100 rad/hr. Krushchev and associates¹⁰¹ reported on potatoes given a total of 5 krads at dose rates of 40 and 630 rad/min and then planted. The potatoes irradiated at the lower rate sprouted in 28 days, but the potatoes irradiated at the higher rate did not sprout for 53 days.

Kahan and Temkin-Gorodeiski⁸⁴ used two different dose rates---19 and 76 krads/hr---but did not discern any rate effect. This dose rate variation is smaller than the variation in the two previous works^{97, 101} (1:4 compared with 1:6000 and 1:16). Mathur⁷⁷ found a high rate to be more effective than a low rate at a dose rate ratio of 1:12. Although the total doses used by Scheid and Heilinger⁷⁸ were not high enough to effectively inhibit sprouting, they did record a dose rate difference; 45 r/min was more effective than 4 r/min.

Except for a mention in Freund's review,⁷ no dose-rate effect was reported by experimenters in the United States or Canada.

Chemical Sprout Inhibitors (3)

Maleic hydrazide (MH or MH-30) and isopropyl N-(3-chlorophenyl)carbamate---also referred to as chloro-isopropyl carbamate and abbreviated in both cases as CIPC---are the two chemicals most widely used/for sprout inhibition of potatoes in the United States. Fusarex, a tetrachloronitrobenzene compound, is used in Norway. In the USSR, CIPC and a dust that contains 3.5% α -naphthyl methyl acetate are used. In Spain, MH, CIPC, and derivatives of 2,4-D acid are used. Canada has authorized the use of both MH and CIPC. On the other hand, since 1956 France has expressly forbidden the use of chemical inhibitors.

Sawyer and Dallyn³⁷ investigated tetrachloronitrobenzene, isopropyl n-phenyl carbamate, CIPC, MH-30, and the methyl ester of α -naphthlene acetic acid (MENA). At that time they concluded that CIPC and MENA were the most promising chemical inhibitors. However, they apparently applied MH-30 as a gas after harvest; it is

usually applied as a spray on the foliage before harvest, with good results.

All these chemicals inhibit sprouting in stored potatoes to some degree. Both MH-30 and CIPC are effective for 1 year on potatoes stored at 5°C (10°C). According to Freund,⁷ both prevent potatoes in common storage from sprouting/ until the following June. Mikaelson and co-workers⁹⁰ reported a 28% weight loss/for Fusarex-treated potatoes after 1 year at 7°C.

Advantages and Disadvantages (4)

The Russians¹⁰¹ state that most chemical preparations have a specific odor, and they characterize dusting as extremely laborious and unpleasant. Freund⁷ states that chemical inhibitors have several advantages, not the least of which is that the time of application is not critical during storage of potatoes. Maleic hydrazide is applied in the field, which lessens storage requirements considerably; it does not interfere with wound healing; and, because MH-30 translocates to the interior of the potato, there is no problem of compliance with limits on external residues. This compound is used extensively by farmers who grow potatoes under contract to chip manufacturers and know in advance that long-term storage will be required. One disadvantage of MH-30 is that it must be applied at exactly the right time; early application may reduce crop yield and late application may give incomplete sprout inhibition. Uniformity of treatment is important and frequently is difficult to obtain; for 24 hr after application, the atmosphere must be free of rain or heavy dew. Because its application must precede harvest, MH-30 may be used needlessly sometimes by a farmer whose crop is bought for immediate use; this is an economic disadvantage of MH-30.

Application of CIPC to potatoes in bulk storage is simple. The warehouse must be made airtight. The chemical is then applied for 1 hr, often by a mist generator made from a wartime smoke screen generator.¹⁰⁹ The building is left sealed for 48 hr; fans keep the mist circulating. The residue level of CIPC is regulated. (CIPC is not allowed on table stock potatoes in Canada.) This compound can increase rot if applied to new potatoes; evidence is contradictory but indicates that potatoes treated with CIPC may sprout internally, which renders them unfit for use.

Comparison of Chemicals and Radiation (3)

Few direct comparisons of chemical sprout inhibition with radiation inhibition exist in the literature. Mikaelson and associates²⁸ reported a 3.6% weight loss in potatoes treated with Fusarex and a 4.5% weight loss in 10-kr-treated potatoes after 8 months; neither group sprouted. Later, Mikaelson and Roer⁸⁹ reported that Fusarex-treated potatoes started sprouting before irradiated potatoes. After 12 months of storage, both Fusarex-treated and control potatoes looked unsuitable for food, but irradiated potatoes were still acceptable. When planted, the Fusarex-treated potatoes sprouted 1 week later than did the controls; potatoes irradiated at more than 5 kr did not germinate. These Norwegian researchers⁹⁰ have indicated that Fusarex applied at double^A the recommended strength gave sprouting protection equal only to about a 5-kr dose of gamma radiation.

During the Canadian pilot-scale demonstration program, internal sprouting was observed in potatoes treated with an unidentified chemical inhibitor.⁵⁶ More specifically, Gardner and MacQueen¹¹⁰ noted internal sprouting in Kennebec potatoes to be used for chips, that had been treated with CIPC. Also during the pilot scale program,⁵⁶ shrivelling and softening / of both MH-30—treated and irradiated potatoes occurred toward the end of storage at high temperatures. However, this softening occurred only in the surface layer of irradiated potatoes; it was uniform throughout the MH-30—treated potatoes.

In Belgian work,⁵⁹ potatoes treated with CIPC were compared with the irradiated potatoes. Although numerical results were not given, evidently the irradiated potatoes were sufficiently better to warrant continuation of the work.

A specific study was carried out by Kwiat¹¹¹ to compare irradiation, CIPC, and MH-30 for treatment of Kennebec, Pontiac, and Russet Burbank potatoes. Although many factors were evaluated in this study, the degrees of sprouting after 1 year at 55°F (12.8°C) and 75% R.H. show that CIPC and irradiation (7.5- to 15-krad dose) were both superior to MH-30. For lower storage temperatures, the effects of the treatments differed little; in fact, the control potatoes were still acceptable for some process uses 8 months after harvest. Irradiated Pontiac potatoes were

marginally better than CIPC-treated Pontiacs, but the reverse was true for Kennebec potatoes. The Russet Burbank potatoes were not chemically treated; only irradiated and control potatoes were compared.

In the results given by Bradley,¹³ irradiated potatoes had not begun to sprout 211 days after irradiation. He pointed out that control potatoes treated in January with a chemical---presumably CIPC---had also not sprouted under the excellent storage conditions used [45°F (7.2°C) and 95% R.H.].

How Radiation Inhibits Sprouting (3)

Exactly how radiation inhibits sprouting of potatoes was largely ignored by early researchers. Indeed, the literature on radiation inhibition of sprouting is remarkable for this very obvious lack of information. The proceedings²⁸ of the Brookhaven Conference contain no mention of the mechanism.

Metlitsky and associates⁹⁸ state that inhibition results from a combination of factors: change in state of cellular colloids; alkalization of cellular sap in meristematic tissue; and suppression of nucleic acid synthesis, oxidative enzyme activity, and respiration. Brownell and co-workers⁴² suggest that irradiation disturbs the dividing mechanism or the metabolic systems of the sprout cells without affecting the metabolic processes of the cells in the potato itself.

Extensive Russian work, reviewed by Rubin and colleagues,⁸ at first concentrated on the differences in respiration between irradiated and control potatoes; nothing directly affecting the sprouting mechanism was discovered. In cognizance of the extreme sensitivity of nucleic acid synthesis to radiation, the work was reoriented. These researchers thus established that potatoes lose the ability to sprout because irradiation inhibits nucleic acid synthesis in the apical meristems and alters the oxidation-reduction equilibrium. These changes are manifested by shifts in the pH of the interior of the cells and in the isoelectric point of the plasmatic colloids.

Previously (in 1958), Rubin and Metlitsky¹⁰⁰ had reported that retardation of germination in potatoes is governed by a combination of changes: (1) changes in the state of the cell colloids; (2) shift of isoelectric zone of albumens

and nucleoproteins to the acid side; (3) retardation of synthesis of nucleic acids in the buds; and (4) suppression of activity of a number of oxidizing enzymes in the mitochondria.

Although the Russian work was still not widely known, Jaarma⁹⁷ stated in 1960 that sprout inhibition is the result of a disturbance of the phosphorylating mechanism. In 1963, Mathur¹¹² cited radiation inhibition of the endogenous indolyl-3-acetic acid synthesizing system, which he found could be reversed by dipping potatoes in a 1000-ppm solution of indolyl-3-acetic acid.

Freund, in his review,⁷ stated simply that the basic cause of sprout inhibition by radiation is the decreased activity of vital nucleic acids. He cited Rubin and Metlitsky¹⁰⁰ to support this statement.

In the 1967 review by Metlitsky, Rogachev, and Krushchev¹¹³ of all Russian food irradiation work, the exact connection between potato respiration and sprout inhibition was given. Energy is stored in the cell in special phosphorus compounds, such as adenosine triphosphoric acid (ATP), by oxidative phosphorylation. Nucleic acid synthesis is suppressed by disrupting ATP synthesis; radiation breaks the chain between respiration and phosphorylation. The energy for nucleic acid synthesis is thus dispersed, and the potato cannot sprout.

Summary (3)

This first article in a planned three-article series on sprout inhibition in potatoes by radiation has given the early history of research into this phenomenon and summarized international results accumulated in the 16 years since Sparrow and Christensen¹⁹ first aroused interest in this new way to extend the storage life of one of the world's most important food crops. A brief comparison to chemical inhibitors on the basis of effectiveness has been given, and the explanation that is now accepted for the phenomenon has also been presented.

When potatoes are irradiated, more than just the sprouting mechanism is affected. The vitamin, sugar, and starch contents are changed, and cell structure is altered. The potato resistance to microorganisms may also be affected. All these modifications bear on subsequent uses of irradiated potatoes. Cooking

properties, e.g., suitability for chip manufacture, are of concern, as are the usual subjective organoleptic qualities. The second article in this series, planned for a subsequent issue of Isotopes and Radiation Technology, will summarize these radiation effects on the potato.

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For 8(2): Filler to follow Potato irradiation paper

POTATO-IRRADIATION SYMPOSIUM IN BRUSSELS (2)

A symposium sponsored by several organizations, including the International Atomic Energy Agency, on irradiation of potatoes to prevent sprouting during storage was held in Brussels on October 1-2, 1969. Some fifty representatives of government and industry participated. Three different panels discussed the subject from the standpoints of scientific and technological problems, economics, and wholesomeness of the product. Problems still to be solved include determination of varieties best suited to radiation processing, radiation-induced changes in biochemical composition of the tubers, and the culinary and nutritional properties of the irradiation product. Costs were estimated to be somewhat greater than for chemical treatment, but, at the same time, irradiation was considered more effective and convenient and more readily adaptable to automation. The consensus was that inhibition of sprouting by use of ^{60}Co or ^{137}Cs gamma radiation is entirely without danger to the consumer.

A subsequent meeting on Nov. 3 of experts in the field outlined a tentative program to be followed in the European Community in further potato-irradiation methodology and testing. (MG)

STUDY OF IRRADIATED PASTEURIZED FISHERY PRODUCTS* (2)

L. J. Ronsivalli, F. J. King, V. G. Ampola, and J. A. Holston**

Supplementary Keywords: food preservation; seafood; modification, material; food evaluation, subjective; industry, food; industry; bibliography

are given irradiated and nonirradiated
Abstract. The results of a study of the maximum shelf life of commercial grade cod fillets, prime quality haddock fillets, and prime quality pink and brown shrimp. The increase in shelf life provided by irradiation is inversely proportional to the storage temperature. Gas chromatography was used to study the effects of radiation or storage, or both, on the chemical composition of cod and haddock fillets and to assess the similarity in chemical composition between irradiated and nonirradiated fish of both species.

(Introduction)

This article briefly summarizes research on radiation pasteurization of seafood conducted by the Bureau of Commercial Fisheries, Technological Laboratory, Gloucester, Mass., for the USAEC from Oct. 1, 1960 to Sept. 30, 1967. Detailed results are given for the research conducted from Oct. 1, 1967 to Sept. 30, 1968. The early work included the determination of the feasibility of radiopasteurizing various commercially important marine species; more recent work has been aimed at acquiring data needed to obtain approval from the Food and Drug Administration (FDA) to market irradiated seafood.

Results of the Past Work (3)

Radiopasteurization of haddock, cod, pollock, ocean perch, and soft-shell clams is technologically feasible. Lobster, mackerel, scallops, and blue crab are, to some extent, also amenable to radiopasteurization. Radiation

* L. J. Ronsivalli, F. J. King, V. G. Ampola, and J. A. Holston, Study of Irradiated Pasteurized Fishery Products, USAEC Report TID-25312, Bureau of Commercial Fisheries, April 1970.

** Bureau of Commercial Fisheries, Technological Laboratory, Gloucester, Mass.

doses in the 150- to 450-krad range doubled or tripled the refrigerated shelf life of these products. Storage temperature has some effect; irradiated fish had a shelf life of 1 month or greater at 33 to 35°F (0.5 to 1.7°C) but only 2 weeks at 42°F (5.5°C). For irradiation, as well as ^{for} any preservation technique, the better the pretreatment quality of the product, the longer the shelf life.

Irradiation also extends the shelf life of fish that has ^{already been stored} in ice for a substantial portion of its normal shelf life. Nonirradiated fish stored at sea for 7 days in ice can be kept no more than 7 additional days ^{on shore,} but if they are _{when the boat is unloaded,} irradiated ^{the fish can be kept up to 2 or 3 times as long.}

The organoleptic acceptability of radiopasteurized haddock and cod fillets has been demonstrated by tests with large groups of people, including panels of several hundred soldiers at a military post and of up to one hundred people at scientific meetings.¹ Radiation effect on amino acids and B vitamins in seafood is relatively insignificant ^(m) — ^{effects of} certainly no greater than the _{seasonal} variations.^{2,3}

Many ^{packaging} of the FDA-approved materials for irradiated food were found to be suitable for packaging radiopasteurized fish; best results are obtained with materials having excellent gas-barrier properties. All the packaging materials tested are very good ^{barriers to} bacteria.⁴ Clams, which are susceptible to oxidative browning, and ocean perch, which are susceptible to rancidity, store better when vacuum packaged.

Gas chromatographic and classical wet-chemistry methods were used to follow ^{compounds} changes in the volatile _{from nonirradiated and} irradiated fish. Radiation caused immediate increases in carbonyl concentrations in the presence of oxygen, but the volumes of these compounds did not exceed those _{volumes} reached as a function of storage only. Production of sulfides

was inhibited by irradiation. No aberrant compounds were found in volatile substances from irradiated fish.¹

Irradiation at pasteurization levels reduces the microbial population 2 to 3 log cycles . as determined by total plate count.⁵

By the time irradiated fish become organoleptically unacceptable, total plate count may be as high as 10^8 or 10^9 microorganisms/g, which is relatively high; nonirradiated fish may have a total plate count of 10^6 to 10^7 microorganisms/g at spoilage. The difference may result from differences in the biochemical activities between common spoilage organisms and organisms that survive irradiation.

← The composition of microflora in fish was altered significantly not only by irradiation but also by packaging the fish in hermetically sealed containers.^{6,7} These two techniques appear to complement each other, because a lower radiation energy is required to preserve vacuum-packed fish than is needed for air-packed fish.

Data¹ have been obtained on heat penetration of haddock fillets during cooking and on heat inactivation of the toxin of Clostridium botulinum Type E. Results indicate that normal cooking readily destroys the toxin. Sodium nitrite and sodium tripolyphosphate can be absorbed by fish fillets⁸ in amounts sufficient to inhibit growth of Cl. botulinum Type E.

Maximum Shelf Life Study (3)

Evidence is needed that radiopasteurized fish undergo spoilage just as nonirradiated fish do, even though the former do take longer to spoil. Data are also needed for assessing the potential hazard of Cl. botulinum Type E. Radiation destroys the common fish spoilage microorganisms and thereby extends product shelf life; this effect might create a situation favorable for the growth and toxin production of Cl. botulinum. Spoilage must occur before any possible botulinum toxin development. A program has been initiated to determine the maximum shelf life of nonirradiated and irradiated haddock and cod fillets, because these data have direct bearing on spoilage and Cl. botulinum hazards.

Some research into this problem was reported previously.⁹ In addition to haddock and cod fillets, pink shrimp and brown shrimp were also investigated.

Maximum shelf life is defined as the period, after in days, Δ which a product stored at a particular temperature is rejected unanimously on the basis of odor alone by an untrained panel of at least 20 consumers. The term "X-value" is used to denote maximum shelf life. An

expert panel also determined maximum shelf life. A comparison of the two panels' results, Δ should reveal any radiation-induced alteration in seafood that might bias an untrained panel. No such bias was found.

Cod (4)

Experiments were completed with irradiated and nonirradiated commercial quality fillets cut from 2- to 4-day old cod, Δ that were caught and handled under ordinary conditions. cellophane-wrapped fillets Δ were separated into four lots for irradiation at doses of 0, 75, 100, and 200 krad. Each lot was subdivided for storage at 38, 42, 48, and 55°F (3.3, 5.5, 8.9, and 12.8°C). Samples were presented to the expert panel in loosely capped 1000-ml beakers. Two or more samples

of known ages were included in each test. After each lot was rejected by the expert panel, it was submitted to the consumer panel (personnel of the Bureau of Commercial Fisheries, Technological Laboratory). The untrained panel was also presented with good quality samples to check the accuracy of the response of the untrained panelists. All samples eventually spoiled, of course. Results of the experiments (Table 1) show that nonirradiated fish become unacceptable within a relatively narrow range of time at each storage temperature. However, irradiated samples differ from each other, especially at the lower temperatures and higher doses. This finding indicates that the extra-long shelf-life extensions occur at temperatures at which botulinum toxin cannot be produced (<38°F). Since irradiation is most effective at lower storage temperatures, commercial handlers will be financially motivated to maintain low temperatures. The only unusual data are the relatively short shelf-life extensions obtained at 38°F (3.3°C) in Experiment No. 1 (Table 1).

Haddock (4)

One X-value experiment was completed on prime quality skinless haddock fillets. The results (Table 2) are similar to the cod data.

Shrimp (4)

Pink shrimp and brown shrimp, caught off Florida,

were packaged in polyethylene-coated polyester pouches, with headspace volume minimized by the packager's pushing out the air before heat-sealing the pouch. At irradiation, the brown shrimp were 2 days and the pink shrimp 4 days out of the water.

Table 1. MAXIMUM SHELF LIFE OF IRRADIATED AND NONIRRADIATED COMMERCIAL-QUALITY COD FILLETS

Dose, krad	Exp.	X value, days				
		Storage temperature, °F				
		33°	38°	42°	48°	55°
0	1		11	8	7	4
	2	14	11	7	7	3
	3	12	11	7	5	4
75	1		11	11	10	7
	2	23	18	9	8	7
	3	40	27	9	11	6
100	1		15	11	11	9
	2	30	21	15	10	8
	3	49	30	15	12	6
200	1		30	23	15	10
	2	45	32	22	14	8
	3	50	29	22	13	9

Table 2. Maximum shelf life of prime quality skinless haddock fillets.

Dose, krad	X value, days			
	Storage temperature, °F			
	38°	44°	46°	50°
0	14	7	7	4
50	19	14	9	9
100	24	15	16	10
200	35	18	17	15

All samples developed the characteristic spoilage odors of shrimp; brown shrimp became discolored, and nonirradiated brown shrimp developed black spots. The brown shrimp had longer X-values than did the pink shrimp (Tables 3 and 4); however, the X-values of the two species were almost the same at the 200-krad dose. As with the cod and haddock fillets, the effectiveness of radiation as a preservative was greater at lower temperatures.

Toxin Outbreak (4)

Proper evaluation of the safety of radiopasteurized fish requires information on toxin outbreak times as well as information on X-values. The minimum time required for production of toxin in seafood inoculated with Cl. botulinum and stored under conditions similar to those used for X-value determinations is necessary. The inoculation level should be sufficiently higher than the normal incidence level to provide a safety factor. ^{Massachusetts Institute of Technology} Microbiologists at / conducted a survey¹⁰ which showed that the incident level of Cl. botulinum Type E is very low--- 0.17 microorganisms/g. A safety factor of 1000 would mean that the product would spoil recognizably ⁱⁿ well in advance of toxin production in a sample inoculated with 170 microorganisms/g.

Radiation Effect on Chemical Composition of Cod and Haddock (3)

Considerable progress has been made in developing the technology of radiation preservation of seafood, but there is a need to establish that consumption¹¹ of irradiated / seafood is safe. Some animal feeding studies have been completed and more are planned; however, basic information on the chemical effects of ionizing radiation on seafood is needed to evaluate these studies and to plan further studies. Most of the knowledge about effects of radiation on organic compounds ^{simple systems} has been gained from studies of / of known components.¹² Some model system

Table 3. Maximum shelf life of irradiated and nonirradiated brown shrimp.

Dose, krad	X-value, days			
	Storage temperature, °F			
	38°	42°	48°	55°
0	15	13	6	3
50	27	15	8	9
100	35	24	10	8
200	30	27	10	10

Table 4. Maximum shelf life of irradiated and nonirradiated pink shrimp.

Dose, krad	X-value, days				
	Storage temperature, °F				
	33°	37°	42°	48°	55°
0	17	10	10	5	2
50	27	20	12	6	4
100	28	25	12	10	6
200	44	33	26	13	7

studies have been oriented towards foods,¹³ and some work has been done on irradiated foods.

The rationale of the experimental procedures used in this study is that ionizing radiation has the potential to induce oxidative reactions in muscle and to fragment a wide variety of chemical compounds

in situ. Chemical or physical methods are used to extract chemical components from fish fillets; the extracts are then analyzed by gas chromatography. Nonvolatile extractable components are converted to volatile derivatives, which are also analyzed by gas chromatography.

Packaging, irradiation, and storage procedures used in this study were based on requirements for the petition to FDA for approval to market irradiated seafood. Two "sterilizing" doses---2.8 and 5.6 Mrads---were used for comparison, even though product temperature at irradiation was between ambient¹⁵ and cryogenic,¹⁶ Raw samples were analyzed, rather than cooked samples, mainly to eliminate variables caused by the variety of heat treatments that might affect the results.^{2, 3, 17, 18}

Preparation of test samples (4)

Skinless fillets^{were} cut from 1-day-old fish purchased from a dealer in Gloucester, Mass., and were halved and packaged in heat-sealed polyethylene bags, with at least one nape section from one fillet and one tail section from another fillet, in each bag. Cod and haddock were irradiated in the Marine Products Devel-

opment Irradiator to ^adose of 0.2 Mrad, given in 82 minutes. The irradiated samples were stored at 40°F (4.4°C) until evaluation. Doses

of 2.8 Mrads and 5.6 Mrads were given to cod and haddock.

Analysis of neutral pH sample (4)

Gas-liquid chromatographic analysis of volatile components has been used to characterize the chemical effects of radiation on seafoods. Mendelsohn et al.¹⁹ showed that the number and quantity of volatile components detected are proportional to the radiation dose.

An exact amount of small fillet pieces ^{was} placed in a glass flask. Because neither acid nor alkali is added to this sample, it is called the neutral pH sample. The glassware and flask design are described by Mebritt et al.²⁰ Use of this system in high vacuum distillation of fish samples is covered by Mendelsohn et al.²¹ (The full details of sample preparation and gas chromatographic conditions are given in the report from which this summary was prepared.)

The general effect of radiation was to increase the number and size of peaks in the ^{chromatograms.} _{chromatograms} These increases were clearly evident in the chromatograms of a control sample and the 0.2-Mrad sample (Fig. 1) for cod. Further increases are evident in the ^{chromatograms} of samples irradiated to higher doses; also, there is evidence that some of the volatile products of low-dose irradiation are fragmented again _{at higher doses} into smaller, more volatile molecules. Along with results from previous studies,^{9, 19, 22} these results demonstrate the desirability of reducing product temperature during irradiation, especially at sterilizing doses.

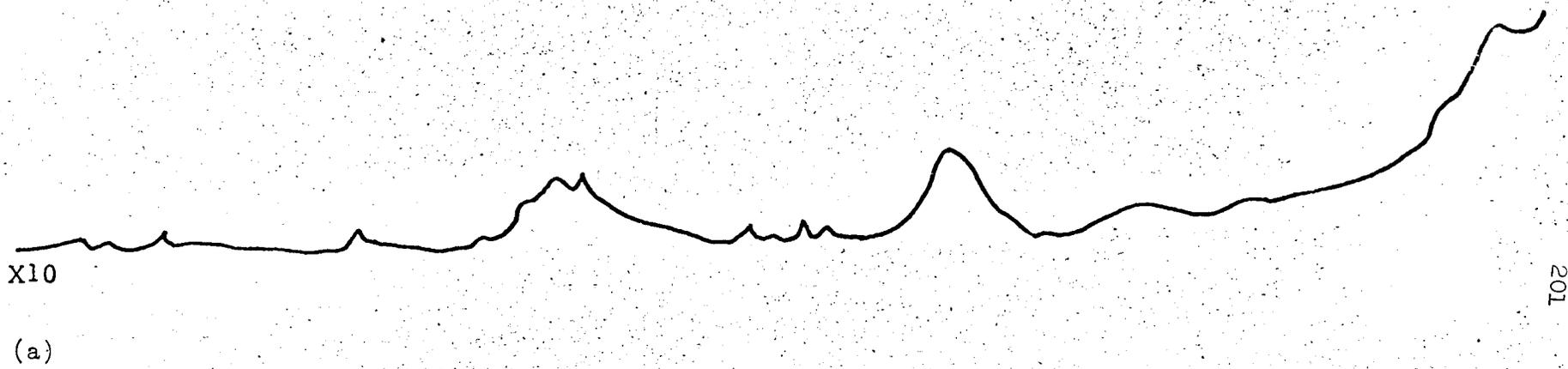
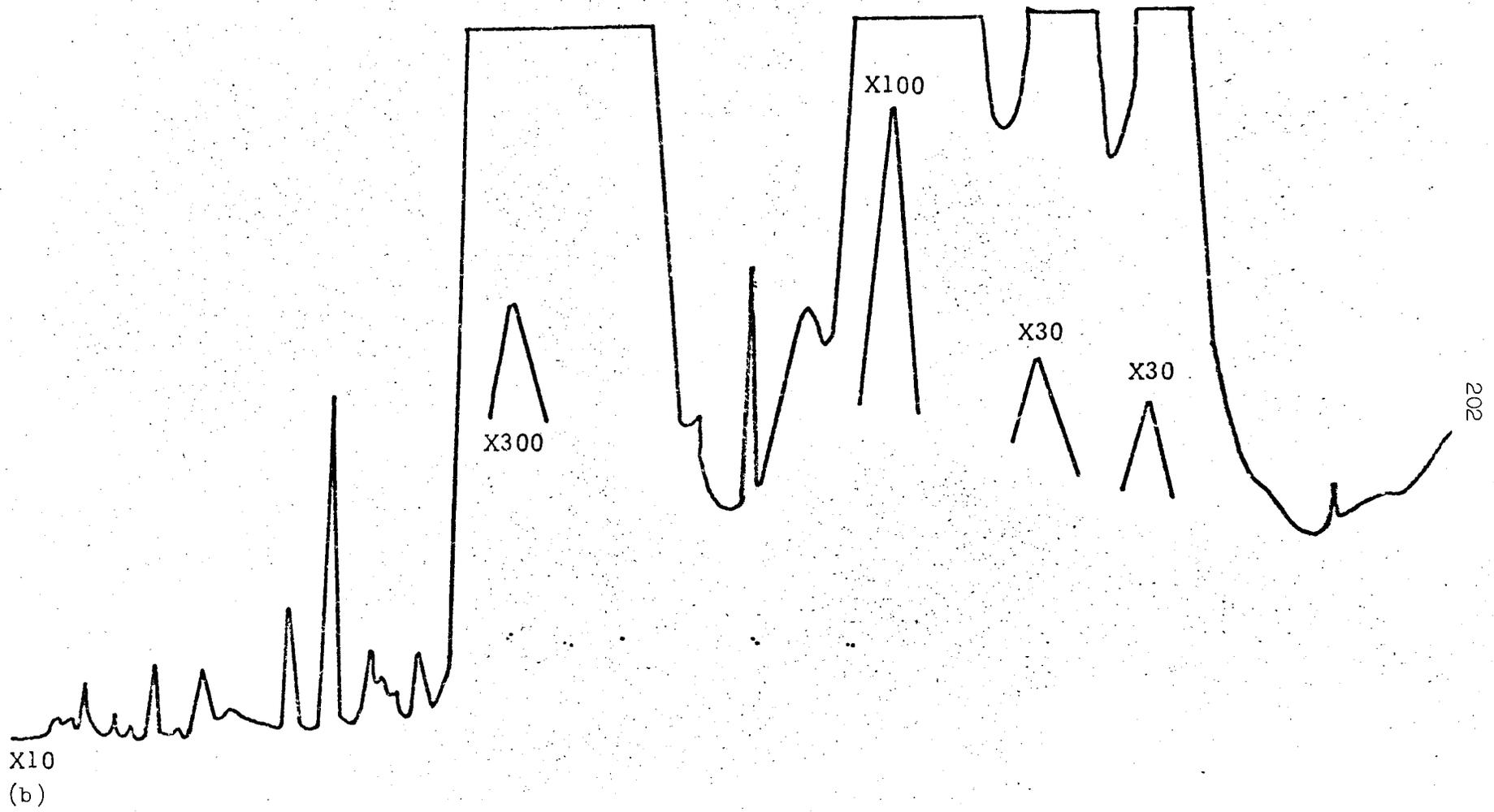


Figure 1. Gas chromatogram of a headspace gas sample from (a) nonirradiated cod muscle and (b) 0.2-Mrad irradiated cod muscle. (1/2-scale drawing)



Likewise,
 /the effect of storing samples at 40°F (4.4°C) was to increase
 the size and number of peaks on the chromatograms; these effects were
 generally additive to the radiation effects (Fig. 2).

Analysis of alkaline pH sample (4)

Because radiation increases the number and quantity of neutral volatile
 components, other compounds such as basic polar compounds (e.g., amines) might
 also be produced or changed during irradiation. However,
 polar compounds in fresh ground fish muscle (pH ~6) might be / insufficiently
 volatile to be detected. Accordingly, the pH of the sample was
 adjusted to 12 by addition of 6 M sodium hydroxide. Making the sample
 alkaline prevented the release of acidic and neutral components and limited
 the collection of volatile condensate to the basic components.

The number and concentrations of basic compounds detected tend to increase
 with increases in radiation dose and in duration of storage (Fig. 3), but the increases do not
 appear to be additive, although in specific components they may be.
 The differences between nonirradiated samples and samples irradiated at high
 doses are greater than differences between nonirradiated and low-dose
 irradiated samples. This result is expected, since there is no evidence of
 organoleptic changes at the lower dose. In the samples irradiated at sterilizing
 doses, several compounds appeared that were either absent or present in very low
 concentrations in the nonirradiated or low-dose irradiated samples. These com-
 pounds may be useful as indicators of radiation treatment, but identification
 and quantification are necessary before conclusions can be made.

Analysis of water-methanol extract (4)

The Bligh-Dyer technique¹⁴ has been used to extract lipids from foodstuffs;
 also, it has the potential to extract nonlipid components from tissue residue.
 Predetermined quantities of water, methanol, and chloroform are used to form

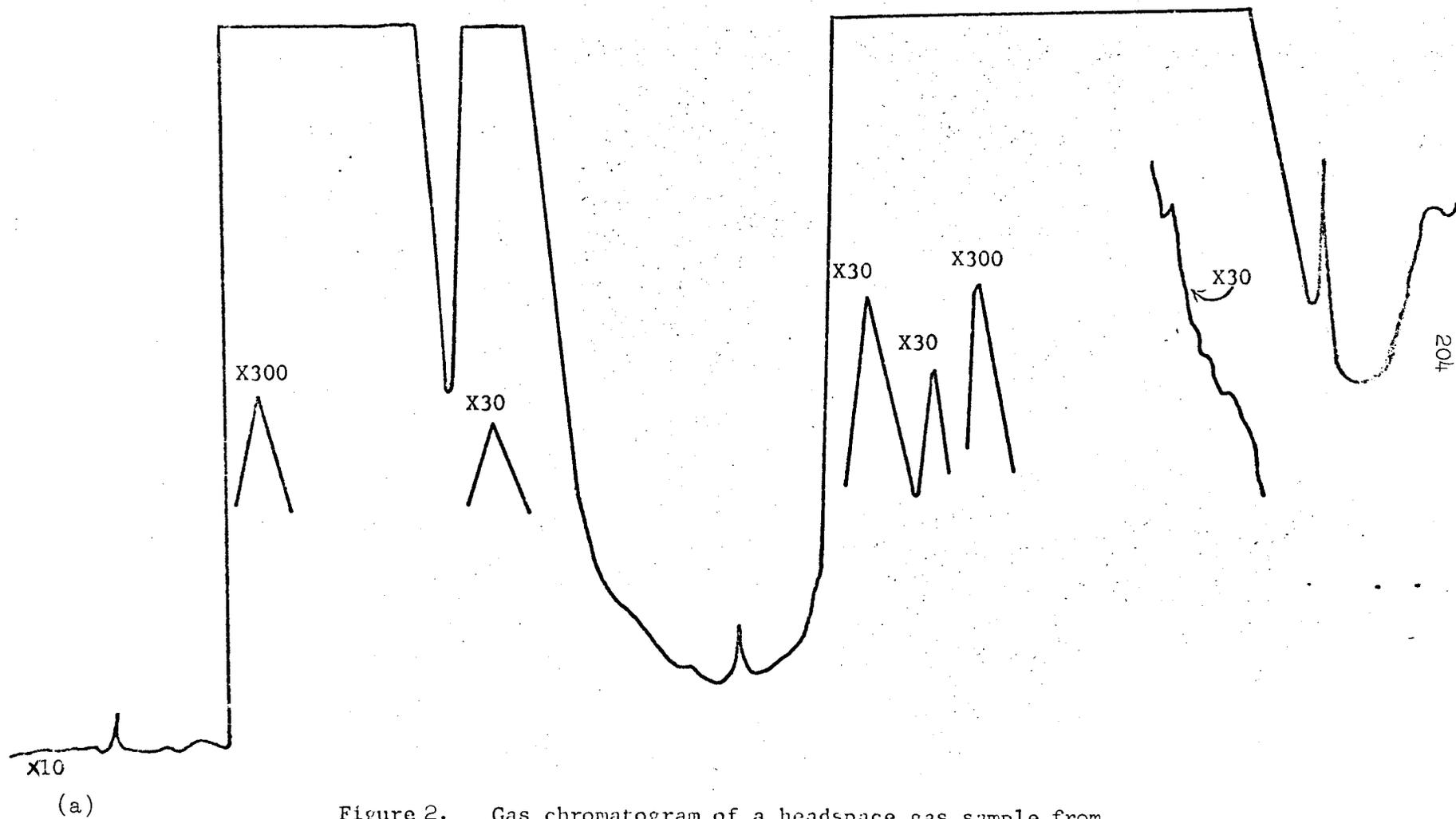
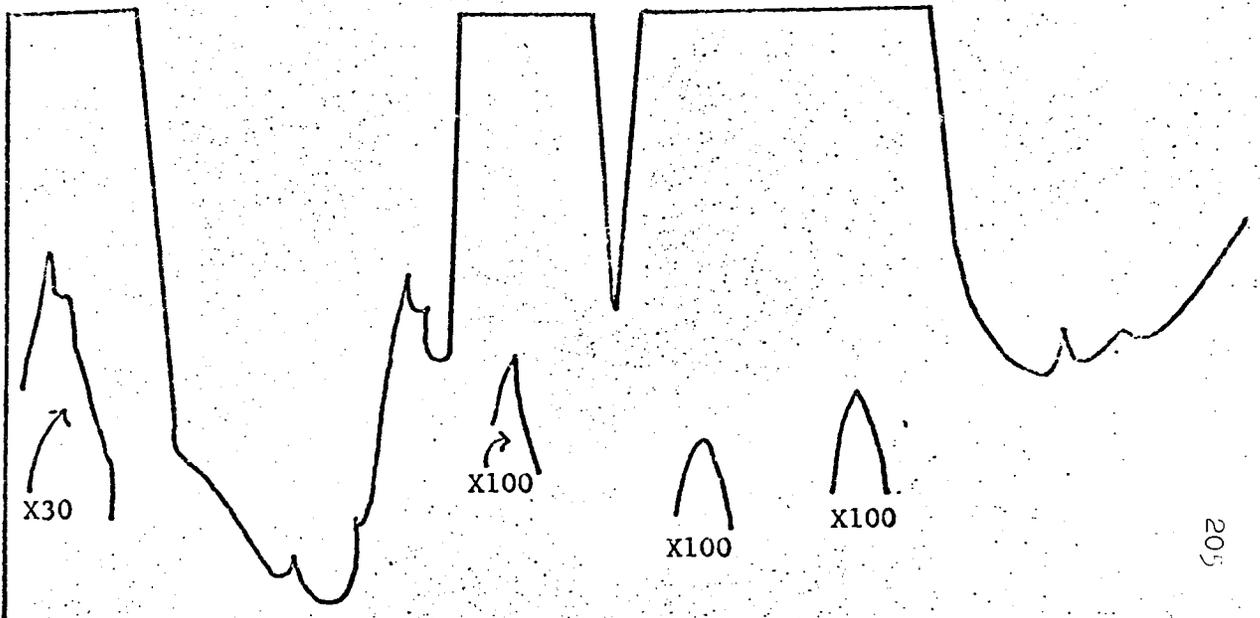
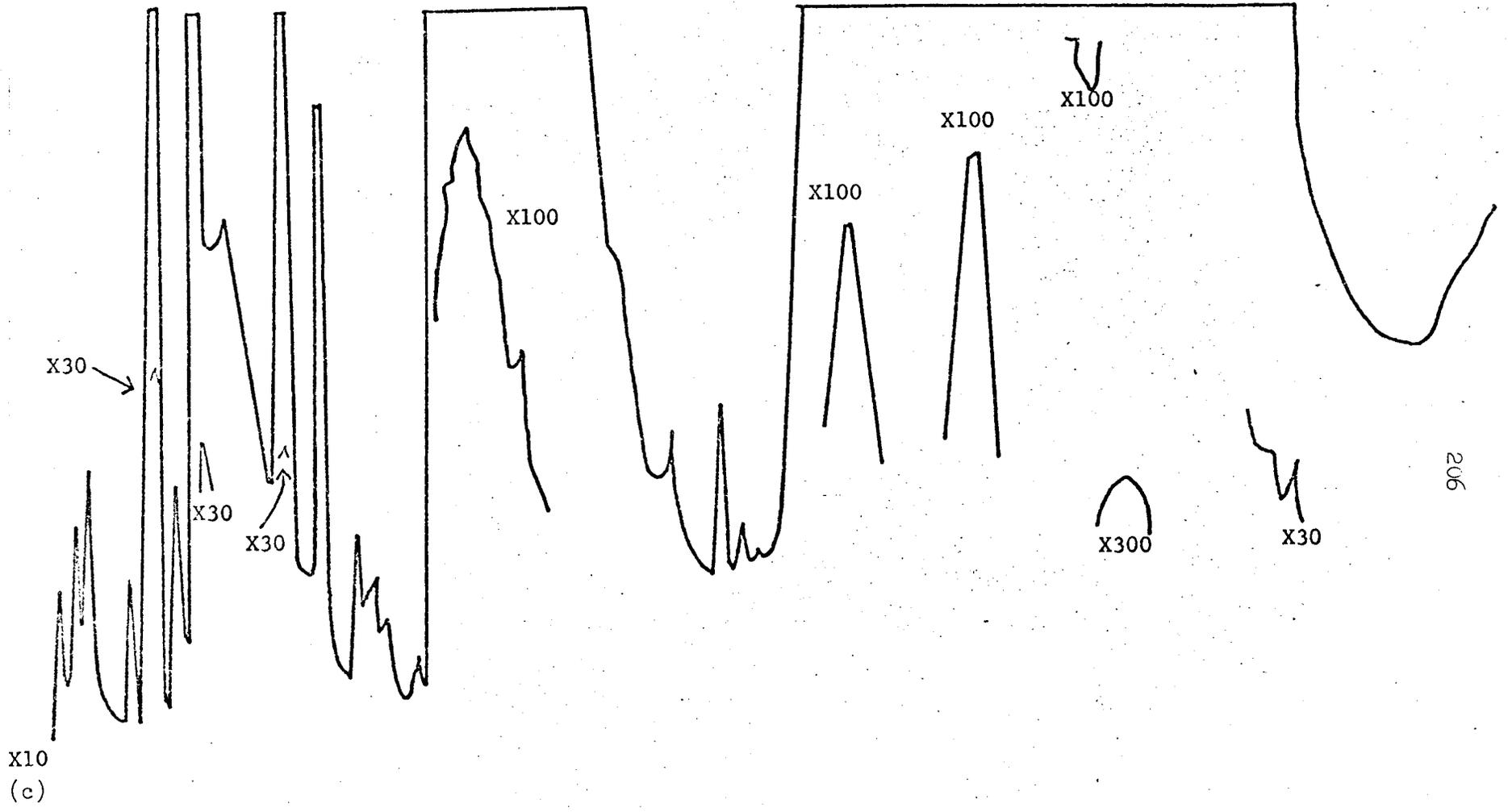


Figure 2. Gas chromatogram of a headspace gas sample from
 (a) nonirradiated cod muscle stored 10 days at 40°F (4.4°C);
 (b) 0.2-Mrad irradiated cod muscle stored 10 days at 40°F (4.4°C); and
 (c) 0.2-Mrad irradiated cod muscle stored 29 days at 40°F (4.4°C).
 (1/2-scale drawing)

X10
(b)





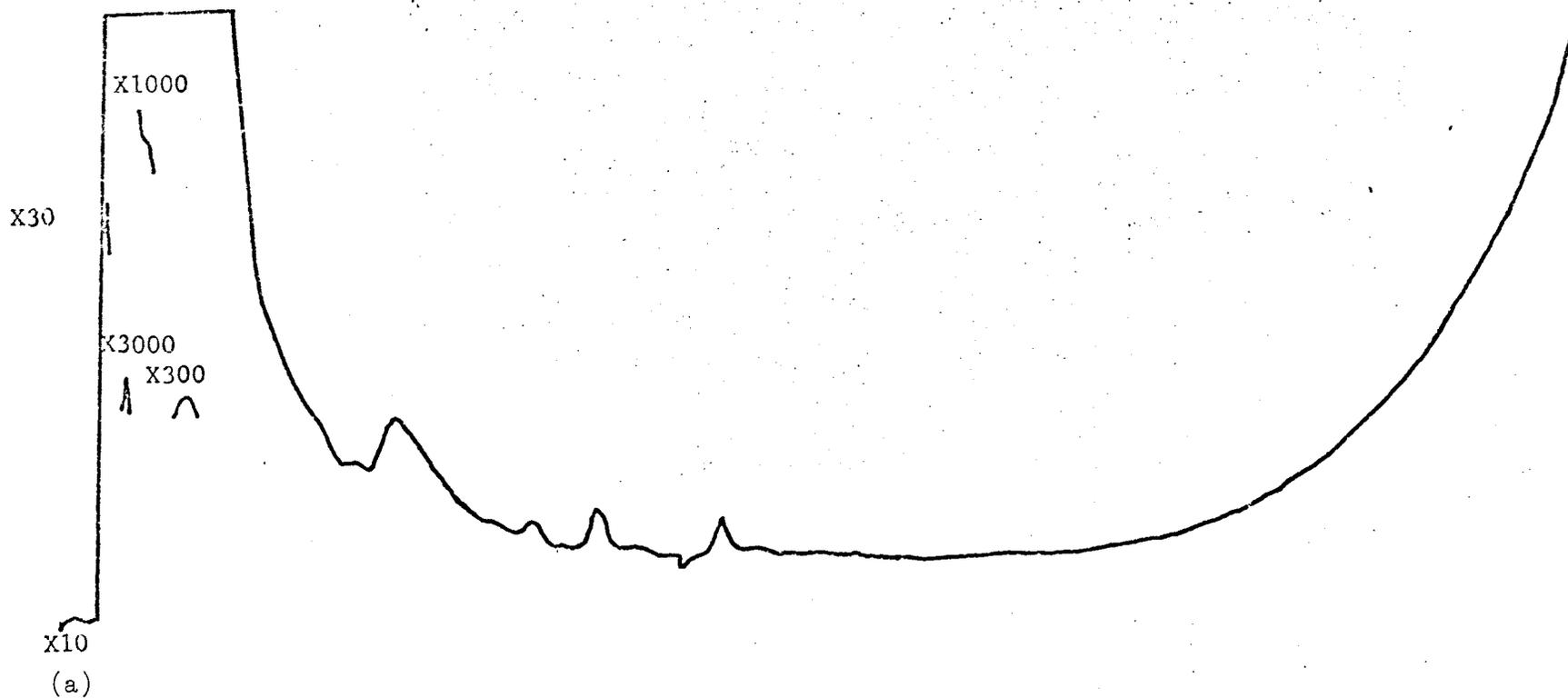
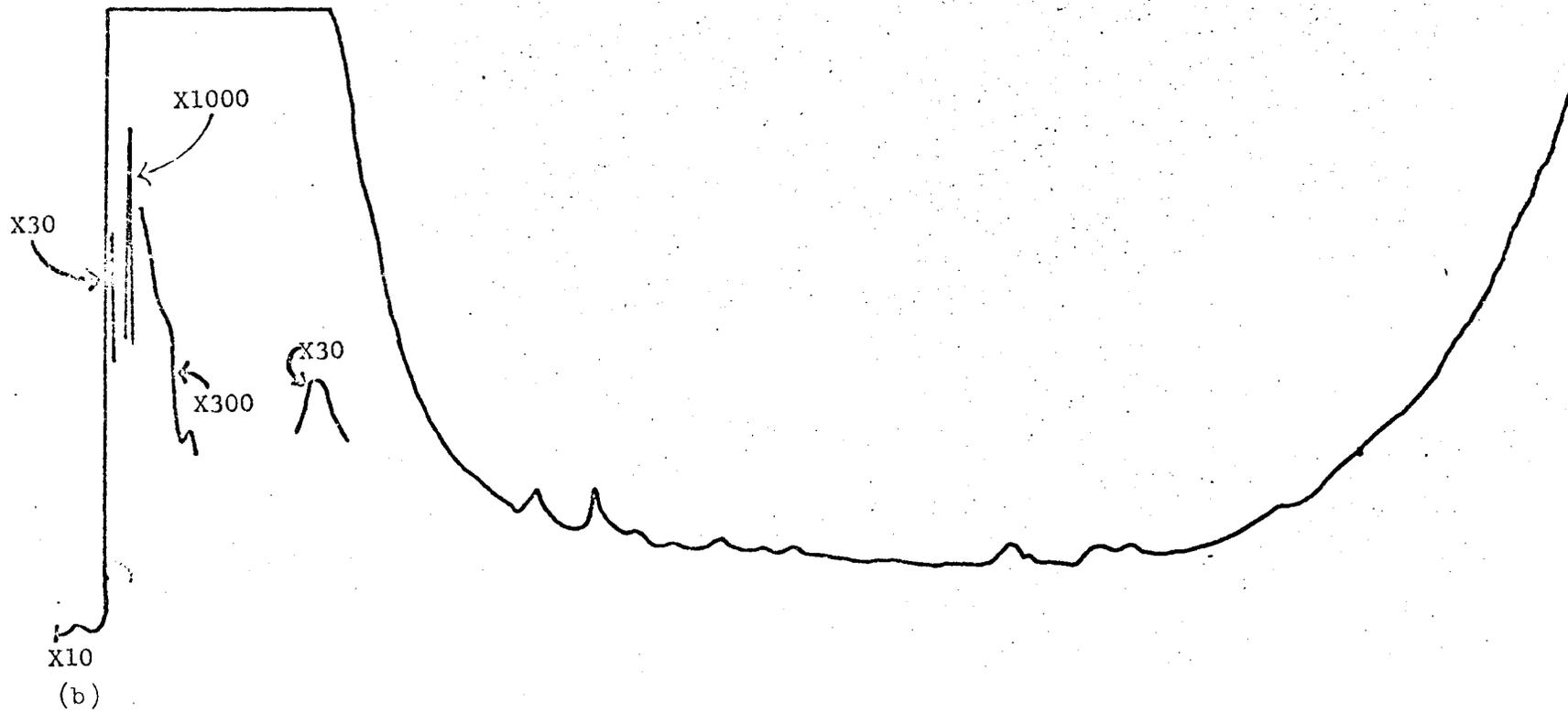
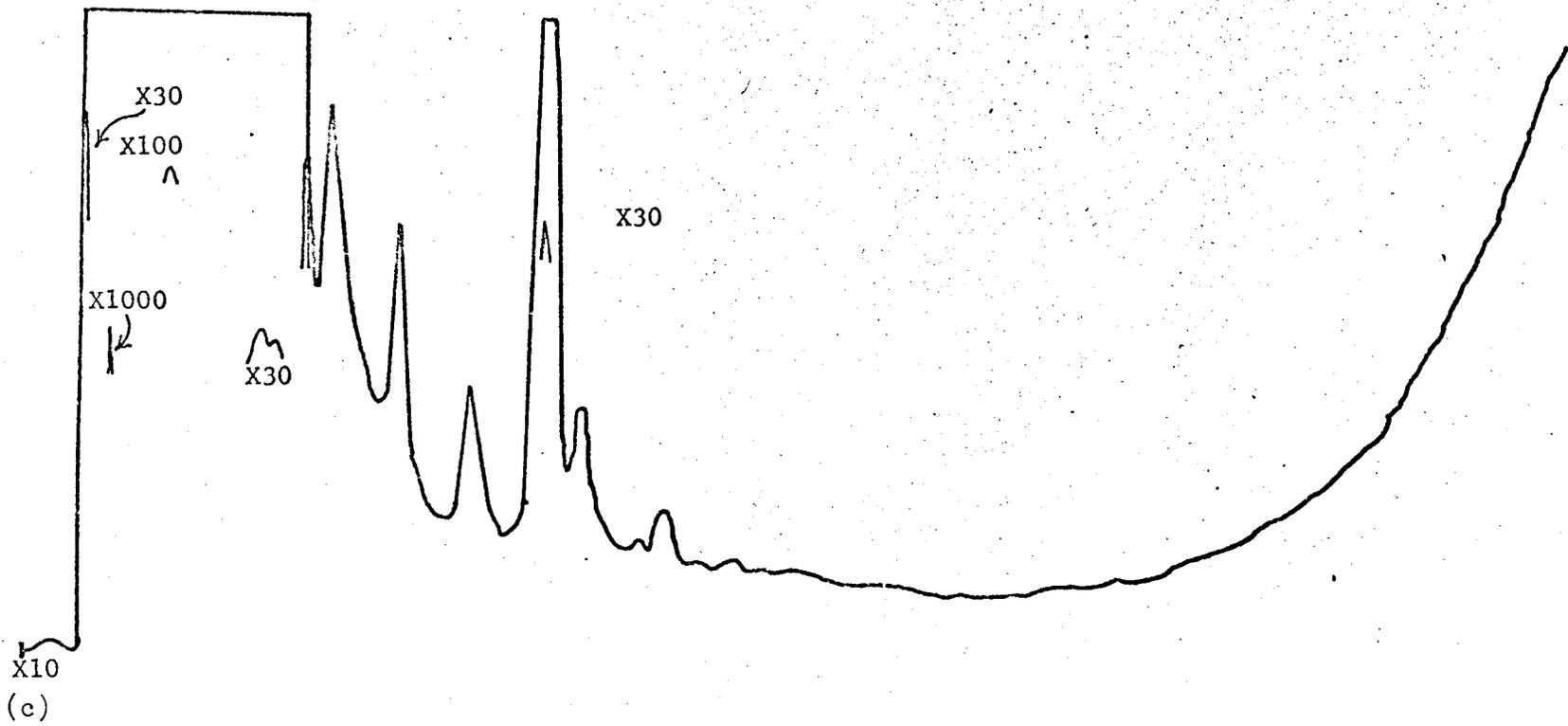


Figure 3. Gas chromatogram of an alkaline headspace gas sample from (a) nonirradiated haddock muscle; (b) 0.2-Mrad irradiated haddock muscle stored 15 days at 40°F (4.4°C); and (c) 5.6-Mrad irradiated haddock muscle stored 32 days at 40°F (4.4°C). (3/8-scale drawing)





two liquid phases and a tissue residue that is mostly denatured protein. Normally the chloroform phase is used to determine total lipids or certain classes of lipids, and the water-methanol phase is discarded with the tissue residue. However, the water-methanol phase probably contains several classes of nonlipid extractives, and the tissue residue might yield components to different or more drastic extraction methods. Accordingly, the water-methanol phase was studied. The compounds responsible for the appearance of the gas chromatograms obtained from the fish samples are evidently soluble in the water-methanol phase of a Bligh-Dyer¹⁴ extract, extractable from this phase into n-pentane, and less volatile than n-pentane itself. (The full report contains the details of preparation of water-methanol phase, n-pentane extraction, and microdistillation.)

Chromatograms from both cod and haddock showed peaks whose size increased with dose in the range 0 to 2.8 Mrad; however, many peaks from the 5.6-Mrad sample were smaller than the corresponding peaks from the 2.8-Mrad sample (Fig. 4). Because highly volatile compounds were supposedly eliminated from the samples by the experimental methods used, the results support the hypothesis that some volatile products caused by low-dose radiation can be fragmented into smaller molecules by subsequent higher doses of radiation.

Effects of storage at 40°F (4.4°C) were similar for both species. Storage increased the number of peaks and some of the peaks seemed to reduce the size of/the peaks (Fig. 5). Decreases in amounts of certain chemical components are to be expected, since the system (i.e., fish fillets) is dynamic and maintains a continuing chemistry. Further experiments, based on other extraction techniques and other chromatographic methods would be useful, that are to determine the chemical effects of radiation on sugars, amino acids, nucleotides, and lipids in fish muscle.

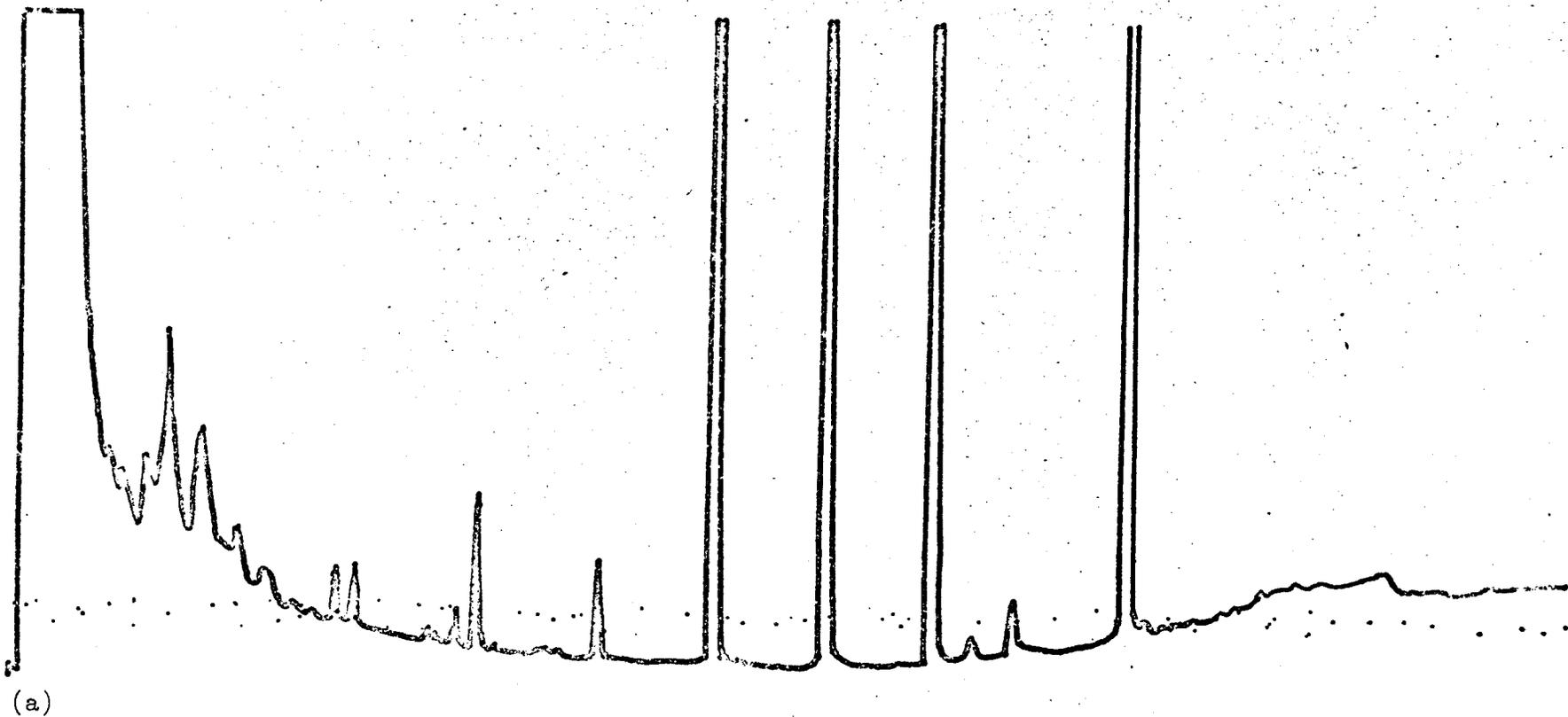
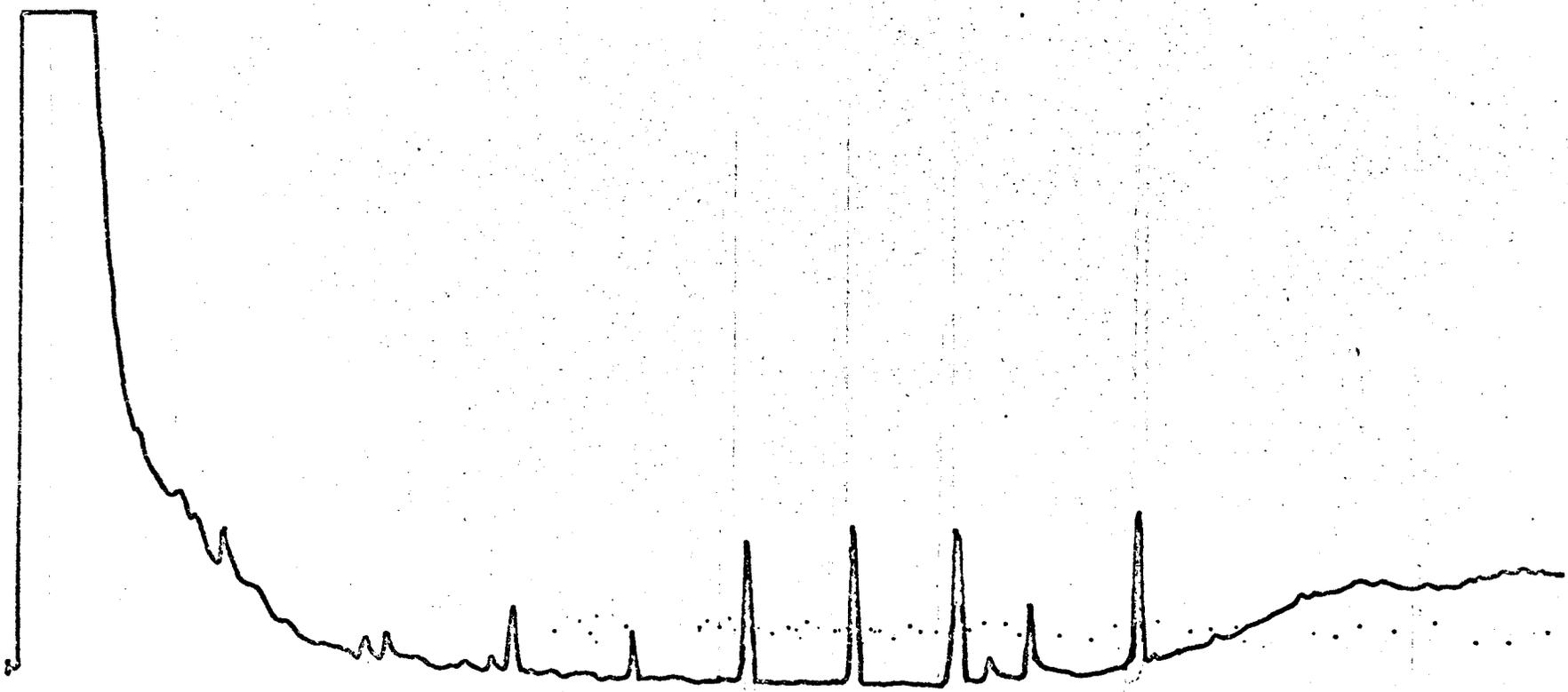


Figure 4. Gas chromatogram of components from a water-methanol extract of
(a) 2.8-Mrad irradiated cod muscle; (b) 5.6-Mrad irradiated cod muscle .
(1/2-scale drawing)



(b)

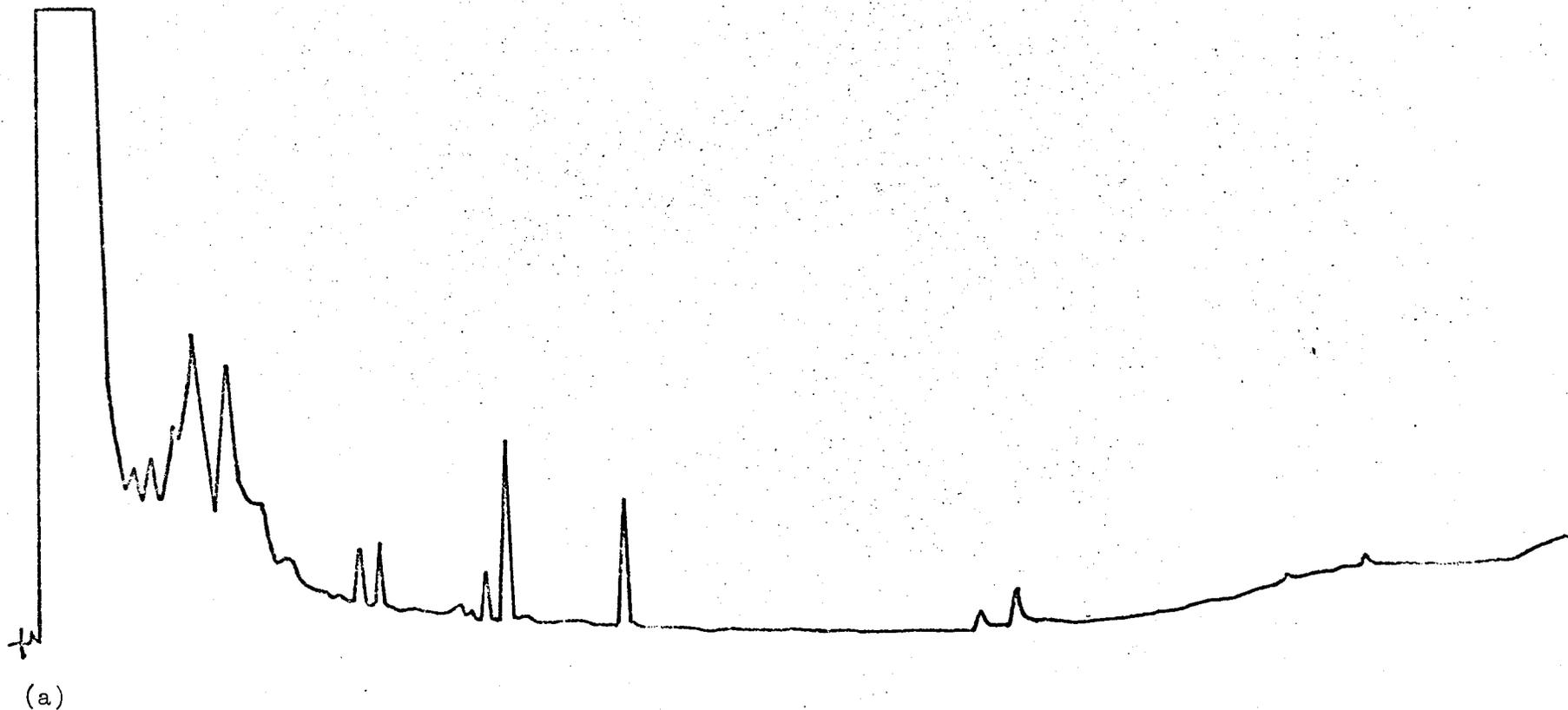


Fig. 5. Gas chromatogram of components from a water-methanol extract of
(a) nonirradiated cod muscle stored 10 days at 40°F (4.4°C);
(b) 0.2-Mrad irradiated cod muscle stored 29 days at 40°F (4.4°C).
(1/2-scale drawing)



(b)

Analysis of free amino acids (4)

Previous analyses of free amino acids in seafood have been undertaken to develop a chemical index of quality in fresh or spoiling unfrozen seafoods.²³⁻³⁵ Free amino acids may contribute to the flavor or acceptability of seafoods³⁶⁻³⁸ and also may affect lipid oxidation during seafood storage.³⁹⁻⁴¹ Autolytic cleavage of muscle protein, as well as microbial activity, can contribute to the formation of free amino acids during storage of seafoods.⁴²⁻⁴⁴ Pasteurizing doses of radiation seem to have little effect on total amino acid composition of seafoods,^{2-3,45} but the effect of radiation on free amino acids in situ has received very little attention. Model-system studies⁴⁶⁻⁴⁷ have shown that radiation can fragment amino acid molecules; therefore, irradiation might be expected to decrease free amino acid content of seafood. Study of a shellfish species² has suggested an increase, however.

Nape portions of fillets were used to prepare samples for analysis of free amino acids. (Full details are given in the report.) The final extract was loaded onto an ion exchange column and the amino acids were eluted with 1 M NH₄OH. Volatile amino acid esters were then prepared for gas chromatographic analysis. Each sample was chromatographed first on a column that contained a polar substrate (adipate) to separate most amino acid derivatives and then on a column that contained nonpolar substrate (Ovid) to separate derivatives of tryptophane, arginine, histidine, and cystine.

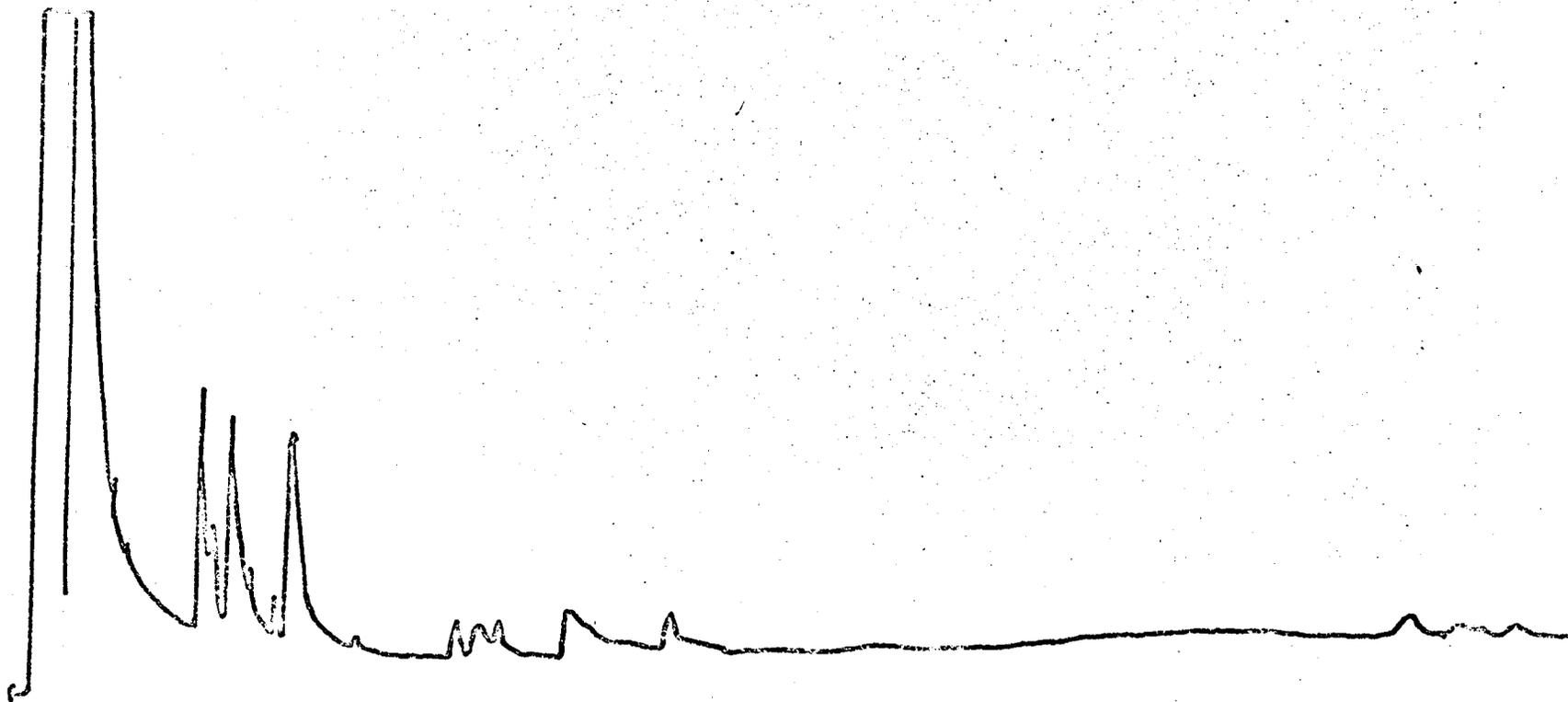
The general effect of radiation was to increase the size or number of peaks. The effect was dose-dependent; the higher doses gave much larger increases in both size and number of peaks (Fig. 6). This finding agrees with that of the earlier shellfish study.² Although destruction of some amino acid molecules may conceivably occur in situ during irradiation, results on all seafoods studied so far--- clam,² cod, and haddock---indicate a net increase in free amino acid concentrations.



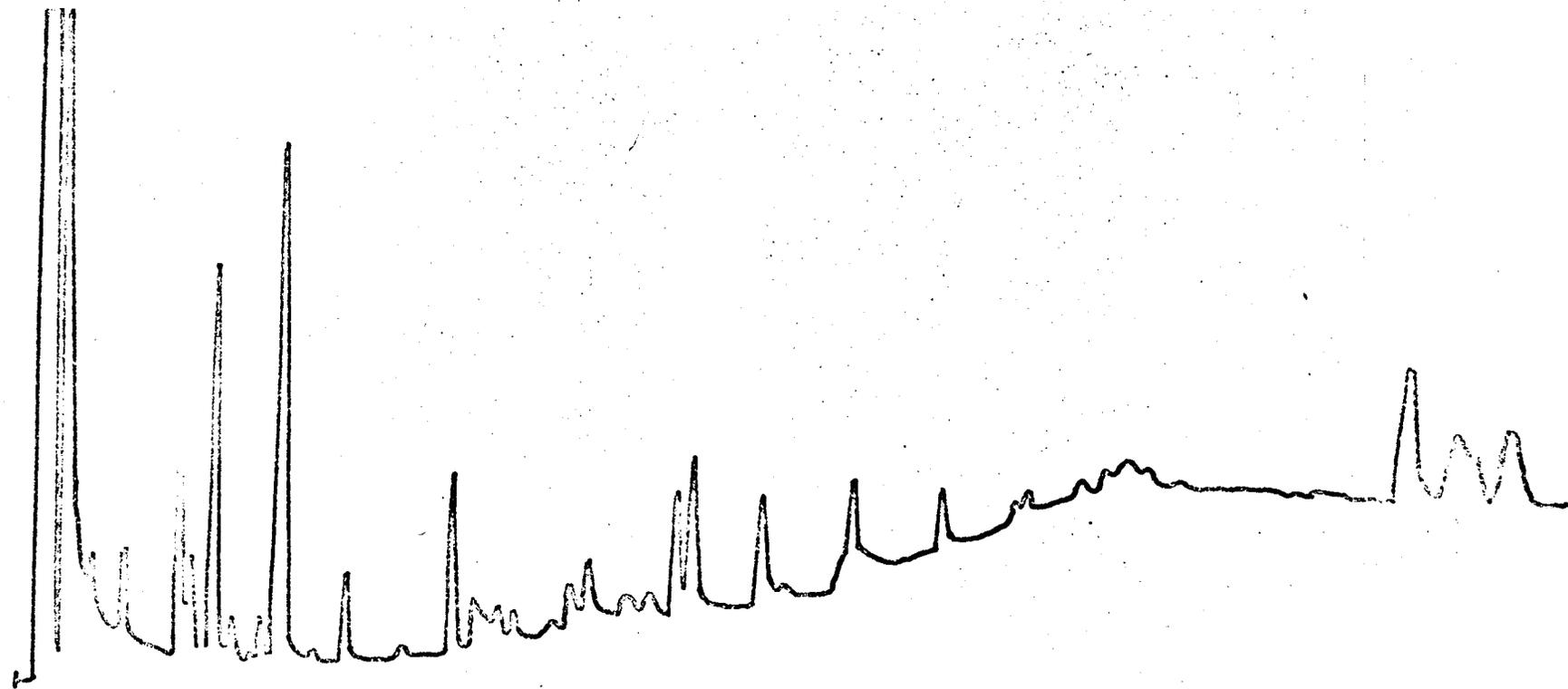
Figure 6. Gas chromatographic determination of free amino acids (Ovid column) from (a) nonirradiated cod muscle; (b) 0.2-Mrad irradiated cod muscle; (c) 2.8-Mrad irradiated cod muscle; (d) 5.6-Mrad irradiated cod muscle. (13/32-scale drawing)



(b)



(c)



(d)

The effects of storing the samples at 40°F (4.4°C) can be explained by considering the relative importance of microbial spoilage chromatograms from enzymatic spoilage. In nonirradiated samples, where microbial activity is dominant, changes in size of peaks (Fig. 7) were much greater than changes in peaks of chromatograms from irradiated samples. In chromatograms of the 0.2-Mrad-treated samples, size of peaks increased during the first half of the storage period ---enzymatic activity---and decreased during the second half of the storage period---microbial activity (Fig. 8). In chromatograms of 2.8-Mrad and 5.6-Mrad samples, there was a general increase in size of the peaks, presumably due to enzymatic activity (Fig. 9). Enzyme activity evidently is the cause of observed storage effects in samples irradiated to eliminate ^{all} microbial spoilage activity.

How irradiation causes an increase in free amino acids is not yet clearly understood. Three hypotheses invoked to explain this effect are: (1) Proteolytic enzymes in situ, ^{perhaps} of bacterial origin, may increase activity because of localized increases in thermal energy. (2) Peptide bands in situ may be broken by direct radiation hits. (3) Tissue structure may break down ^{thus} and _{all} increase the amount of extractable amino acid.

The radiation-induced increase in free amino acid content of seafoods may beneficially affect their nutritive value. Although seafoods are thought to be excellent sources of protein, their dietary value depends on the efficiency with which they are digested and utilized, as well as ^{on} their amino acid composition. Consequently, a nutritional study of protein utilization from irradiated seafood would be worthwhile.

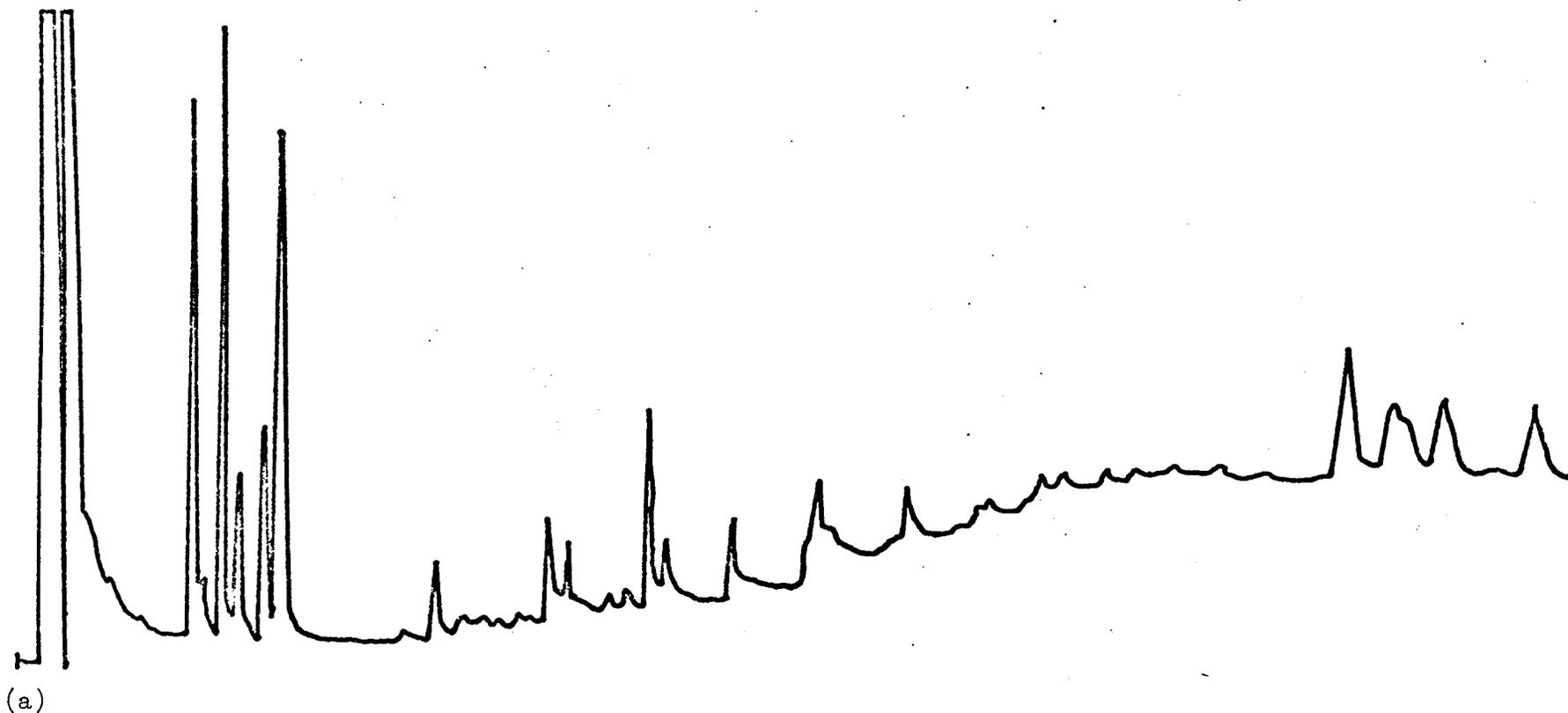
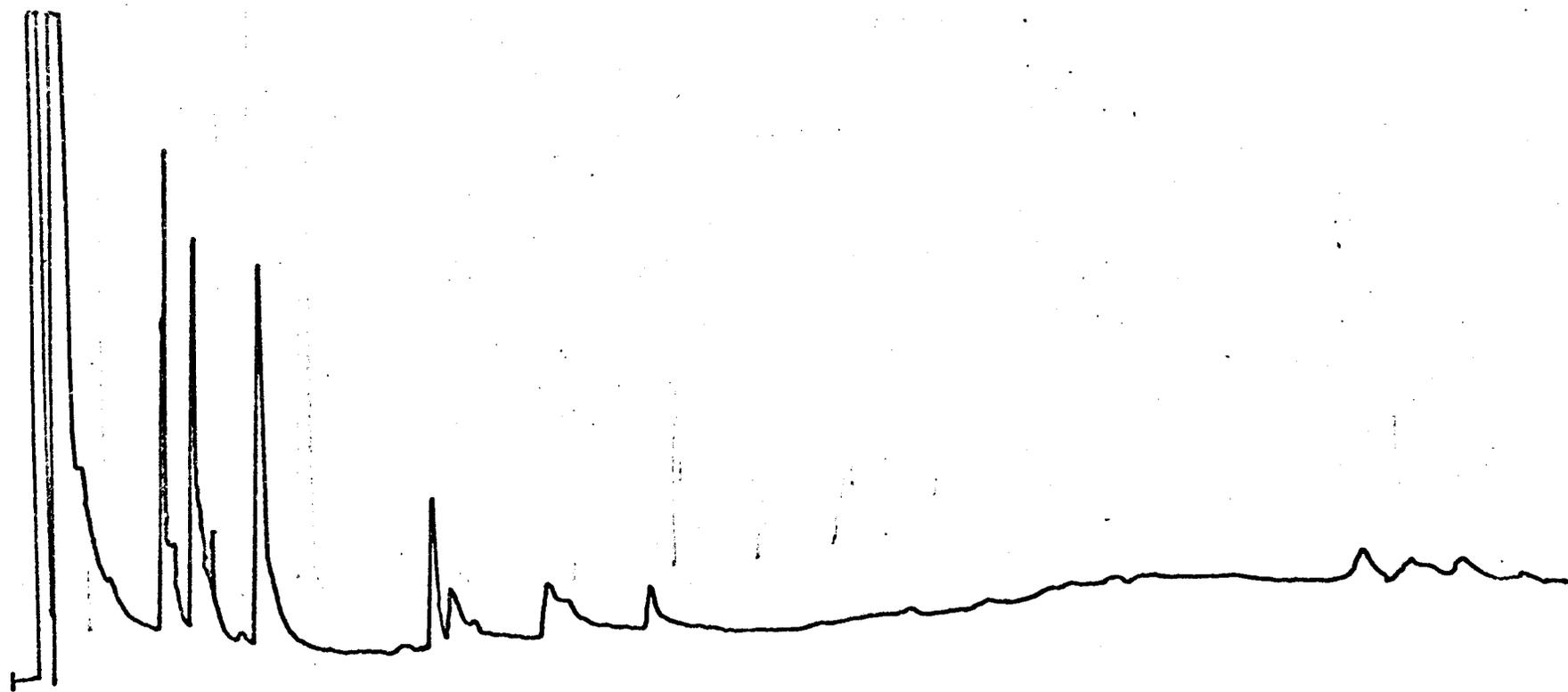
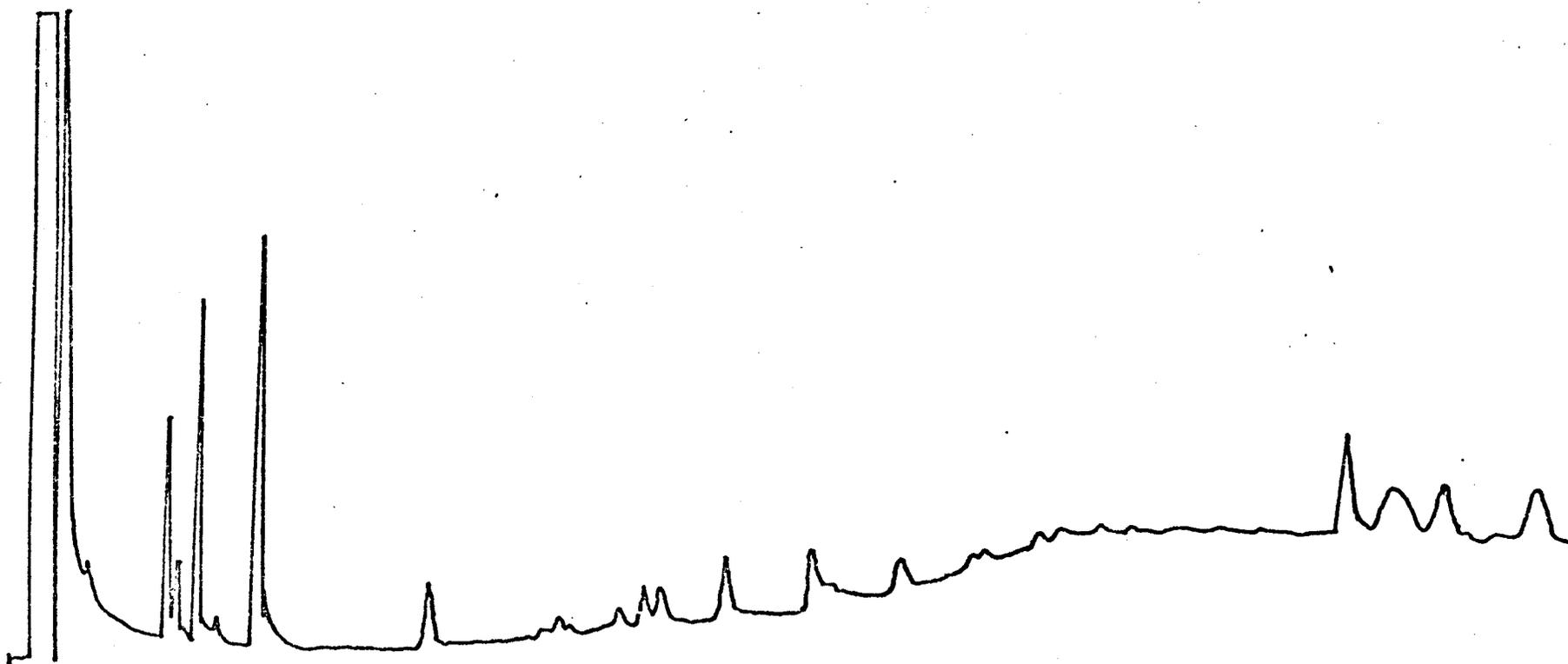


Figure 7. Gas chromatographic determination of free amino acids (Ovid column) from nonirradiated cod muscle stored (a) 10 days at 40°F (4.4°C); (b) 15 days at 40°F (4.4°C). (13/32-scale drawing)

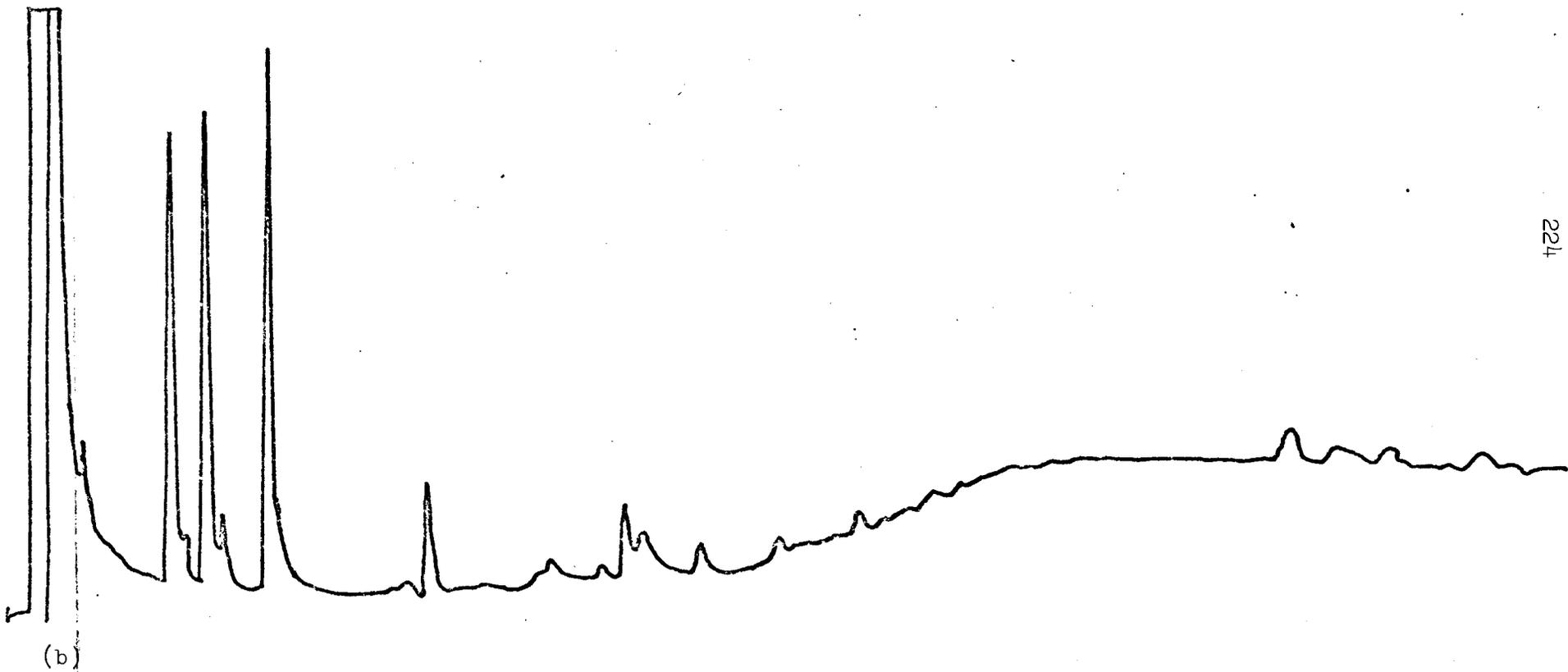


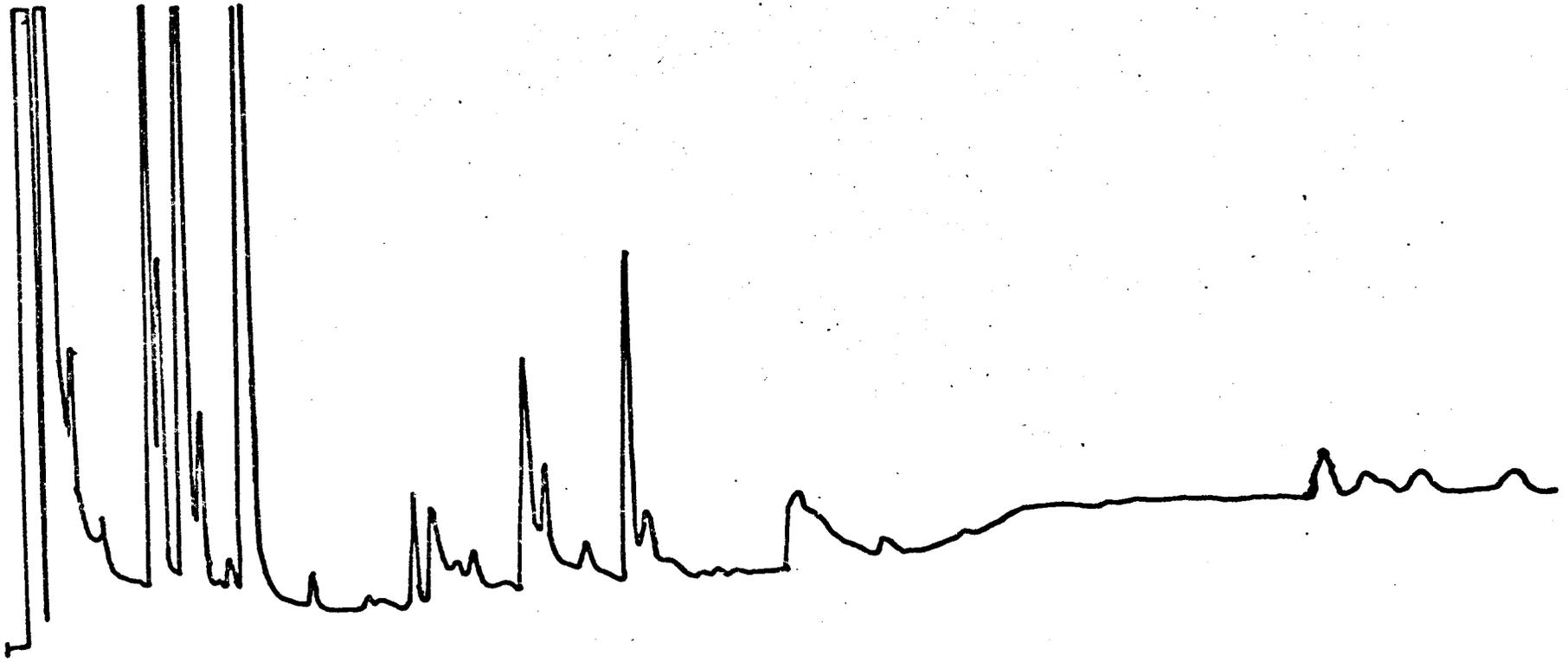
(b)



(a)

Figure 8. Gas chromatographic determination of free amino acids (Ovid column) from 0.2-Mrad irradiated cod muscle stored (a) 10 days at 40°F (4.4°C) (b) 29 days at 40°F (4.4°C). (13/32-scale drawing)





(b)

Analysis of fatty acids (4)

Commercial preservation treatments can cause lipid deterioration in situ and reduce organoleptic acceptability of foodstuffs.⁴⁸ More attention so far has been given to fatty species of seafood than to lean species such as cod and haddock; however, the influence of lipid deterioration on quality of cod and haddock has been documented.⁴⁹ Deterioration of seafood lipid is the result of the reactivity of its characteristic fatty acids.

The primary radiation effects on fatty acids are oxidative; many radiolytic products are formed.⁵⁰ Lipid^{-containing} materials, including mackerel oil, beef fat, pork fat, and synthetic triglycerides, have been irradiated by a ⁶⁰Co source; their volatile products^{have been} determined quantitatively.⁵¹ Nawar⁵¹ has suggested that the type and relative amounts of the volatile products can be predicted if the nature of the irradiated lipid, the radiation dose, and the irradiation temperature are known.⁵¹⁻⁵⁵ These studies did not include synthetic phospholipids or lean species of fish, whose lipid content is 70 to 80% phospholipid.^{49, 56}

The compounds responsible for ^{the} chromatographic peaks are being identified by mass spectrometry. Results indicate that these peaks are not caused unintentionally by artifacts introduced during the lengthy extraction, purification, and derivatization procedures. Some of these amino acids have not been identified previously in seafoods.

In this study, fatty acid composition of irradiated cod and haddock muscle has been determined. Lipids were extracted by the Bligh-

Dyer procedure.¹⁴ The ^{total} amount of lipids extracted was estimated to be 0.5 to 0.6 g ^{of} per 100 g _A cod flesh or 0.6 to 0.7 g ^{per} 100 g _A haddock flesh. After further treatment, the extracts were analyzed ^{of} by gas/^{-liquid} chromatography; ^{polar, moderately polar, and nonpolar} stationary liquid phases were used.

Methyl esters of fatty ^{acids} were identified by comparing chromatographic data the data from ^{from National Institutes of Health} with _A Hormel Institute standards or _A standard ^{mixtures} or by log-plot and separation factor procedures.⁵⁶⁻⁵⁹ The relative amount of each ester present was determined by calculating the ratio of its peak area to the sum of all the ^(Tables 5 to 8) peak areas. Results/^{are} presented in tabular ^{rather than chromatographic} form because rapid identification procedures were used. Conventional shorthand is used to describe the compounds (see footnote, Table 7), which are listed according to structural similarity.

Most of the compounds have been identified previously in nonirradiated cod or haddock muscle.⁶⁰⁻⁶² ^{Compounds associated with peaks} _A labeled RS20:4, RS20:5, and RS22:6 have not yet been identified. (These designations are used to indicate that the peaks were present only in the radiation sterilized samples and that they appeared shortly after 20:4w6, 20:5w3, and 22:6w3, respectively.) The small peak ^{24X} that appeared in some chromatograms may have been observed⁶⁰ previously, but ^{the compound associated with it also} _A remains unidentified. Some of the results are in general agreement with the hypothesis of Nawar⁵¹ that the radiation effect will be quantitative decreases in fatty acid concentrations. The appearance of three compounds was not predicted, probably because Nawar⁵¹ measured volatile components obtained directly from irradiated samples.

RESULTS OF
Table 5. /FATTY ACID ANALYSIS OF LIPID EXTRACTS FROM COD FILLETS USING AN EGSS-X
CHROMATOGRAPHIC COLUMN

Fatty acid*	Percent area under peak**								
	Nonirradiated sample		0.2-Mrad sample			2.8-Mrad sample		5.6-Mrad sample	
	Storage, days		Storage, days			Storage, days		Storage, days	
	0	10	0	10	29	0	29	0	29
14:0	0.52	0.80	0.63	0.48	0.91	0.97	1.29	1.12	0.73
15:0	0.26	0.12	0.33	0.25	0.53	0.32	0.54	0.42	0.43
16:0	15.28	14.84	9.50	14.30	13.80	17.63	16.79	10.76	20.70
17:0	--	--	--	--	--	--	--	--	--
18:0	3.19	2.80	3.75	4.88	3.44	3.51	2.97	7.58	4.33
16:1w7i	1.17	1.36	1.25	1.14	1.41	1.34	1.63	1.87	1.15
17:1w9	--	--	--	--	--	--	--	--	--
18:1w9	9.11	8.30	9.22	10.84	9.07	10.66	11.09	15.23	9.30
20:1w9i	1.73	1.68	1.61	1.15	1.96	2.21	1.63	3.80	2.53
22:1w11i	0.50	0.31		0.57	0.46	1.15	0.45	1.0	1.01
24:1i	0.19	--	--	0.37	0.88	0.16	traces	traces	traces
16:2w4	--	--	--	--	--	--	--	--	--
18:2w6	0.82	0.43	0.67	0.83	1.06	1.11	1.09	1.79	0.79
20:2w6	--	--	--	--	--	--	--	--	--
18:3w6i	0.29	0.36	0.37	0.28	0.34	0.35	0.58	0.73	0.32
20:3w6	--	--	--	--	--	--	--	--	--
18:4w3	0.16	traces	0.24	0.30	0.25	traces	0.28	0.41	0.21
20:4w6	2.38	2.80	3.76	4.81	2.92	2.49	2.87	1.47	3.74
RS20:4	--	--	--	--	--	0.48	0.51	0.14	0.33
20:4w3	0.37	0.24	0.27	0.24	2.26	0.42	traces	0.16	traces
20:5w3	12.95	18.15	20.95	19.35	20.73	11.87	17.33	10.44	13.0
RS20:5	--	--	--	--	--	2.67	3.0	3.63	3.96
21:5w2	0.34	traces	0.53	traces	traces	0.14	traces	0.43	traces
22:5w6	0.42	traces	0.96	0.81	0.95	0.35	traces	0.27	0.25
22:5w3	1.20	1.86	1.85	1.86	3.15	1.18	0.23	1.21	1.78
22:6w3	49.13	45.92	44.07	37.53	35.85	34.87	35.12	28.85	25.41
RS22:6	--	--	--	--	--	6.58	3.14	8.98	7.22
24X	--	--	--	--	--	traces	--	--	--

* Determinations based on relative retention times and peak areas of methyl ester derivatives. Identifications are described in conventional shorthand. For example, 18:3w6i has 18 carbon atoms, ^{and} 3 methylene interrupted double bonds; the terminal methyl group, w, is six carbon atoms from the ultimate double bond; and i signifies that other isomers of 18:3 (such as w3 or w9) may be present but not resolved chromatographically. The RS indicates that the compound occurred only in irradiated samples.

**A dashed line indicates no positive evidence for presence of this compound in the sample.

Table 6. RESULTS OF FATTY ACID ANALYSIS OF LIPID EXTRACTS FROM COD FILLETS USING AN EGSS-Y CHROMATOGRAPHIC COLUMN

Fatty acid*	Percent area under peak**									
	Nonirradiated sample		0.2-Mrad sample			2.8-Mrad sample		5.6-Mrad sample		
	Storage, days		Storage, days			Storage, days		Storage, days		
	0	10	0	10	29	0	29	0	29	
14:0	0.96	1.27	1.05	0.84	1.35	1.81	2.04	1.73	1.10	
15:0	0.48	0.43	0.40	0.45	0.83	0.54	0.94	0.60	0.47	
16:0	29.46	26.10	26.22	29.25	25.92	34.0	29.24	30.0	29.68	
17:0	0.37	0.51	0.38	0.35	2.94	0.44	0.34	0.40	1.39	
18:0	5.60	5.49	6.03	7.45	5.96	5.96	4.89	6.85	6.44	
16:1w7i	7.54	3.76	3.58	3.81	5.58	1.09	5.03	4.52	6.17	
17:1w9	0.38	0.13	0.16	0.22	3.81	0.52	0.42	0.31	1.01	
18:1w9	14.05	14.70	14.37	16.88	13.83	15.0	16.36	16.14	14.60	
20:1w9i	2.56	2.73	2.05	1.26	1.70	2.20	1.53	2.31	2.39	
22:1w	1.17	--	--	--	--	--	--	--	--	
24:1i	--	--	--	--	--	--	--	--	--	
16:2w4	0.95	0.13	--	0.11	3.0	0.32	0.28	0.20	0.94	
18:2w6	1.55	1.08	1.36	1.29	2.98	1.77	1.51	1.56	1.20	
20:2w6	0.12	traces	0.10	0.17	traces	0.16	0.18	0.13	0.09	
18:3w6i	0.31	0.19	0.43	0.31	0.57	0.36	0.39	0.34	0.15	
20:3w6	0.12	0.11	--	0.17	traces	--	traces	--	traces	
18:4w3	--	--	--	--	--	--	--	0.35	0.23	
20:4w6	2.50	2.46	3.12	4.01	1.77	1.79	1.78	1.62	1.11	
RS20:4	--	--	--	--	--	0.47	0.60	0.80	0.43	
20:4w3	--	1.01	0.77	0.55	0.38	0.52	0.63	1.13	0.50	
20:5w3	12.39	13.92	15.82	14.64	11.01	8.72	10.93	7.44	8.60	
RS:20:5	--	--	--	--	--	--	2.25	2.81	3.18	
21:5w2	0.61	0.41	1.88	0.45	0.45	0.25	0.50	0.32	0.13	
22:5w6	0.64	0.29	0.27	0.37	0.49	0.24	0.41	0.47	0.33	
22:5w3	1.21	1.16	1.08	0.67	1.30	0.67	0.89	0.84	1.25	
22:6w3	13.46	23.30	19.97	16.07	14.66	16.00	15.13	13.95	14.20	
RS22:6	--	--	--	--	--	3.54	3.06	4.40	4.58	
24X	1.87	--	0.52	0.52	1.03	0.84	0.78	0.90	0.54	

* See footnote, Table 5.

**See footnote, Table 5.

Table 7. RESULTS OF FATTY ACID ANALYSIS OF LIPID EXTRACTS FROM HADDOCK FILLETS USING AN EGSS-X CHROMATOGRAPHIC COLUMN

Fatty acid*	Percent area under peak**									
	Nonirradiated sample		0.2-Mrad sample			2.8-Mrad sample		5.6-Mrad sample		
	Storage, days		Storage, days			Storage, days		Storage, days		
	0	15	0	15	32	0	32	0	32	
14:0	0.51	0.51	0.72	0.57	0.71	0.71	0.90	0.87	0.86	
15:0	0.27	0.26	0.65	0.51	0.64	0.39	0.76	0.42	0.74	
16:0	13.68	14.77	16.60	15.76	16.55	17.36	20.80	20.75	20.05	
17:0	--	--	--	--	--	--	--	--	--	
18:0	3.95	3.94	3.65	3.82	3.64	4.50	4.43	4.58	4.63	
16:1w7i	0.87	1.32	1.00	1.23	1.43	0.97	1.13	1.50	1.69	
17:1w9	--	--	--	--	--	--	--	--	--	
18:1w9	8.48	8.61	10.04	9.66	9.42	9.76	11.46	12.66	12.13	
20:1w9i	1.23	1.19	2.23	1.82	2.53	2.02	2.00	2.86	1.19	
22:1w11i	0.40	0.51	0.64	0.86	0.37	0.10	0.86	1.09	0.83	
24:1i	0.77	--	--	0.96	1.0	0.33	0.33	0.26	0.78	
16:2w4	--	--	--	--	--	--	--	--	--	
18:2w6	0.66	0.66	0.57	0.61	0.62	1.07	1.14	1.08	1.64	
20:2w6	--	--	--	--	--	--	--	--	--	
18:3w6i	0.35	0.12	0.47	0.38	0.39	0.50	0.24	0.59	0.30	
20:3w6	--	--	--	--	--	--	--	--	--	
18:4w3	0.24	0.11	0.50	0.67	0.28	0.40	0.13	0.63	0.17	
20:4w6	3.68	4.25	2.66	4.20	5.57	2.89	2.68	2.24	3.40	
RS20:4	--	--	--	--	--	0.40	0.19	0.62	0.92	
20:4w3	0.62	0.42	0.79	0.42	0.35	0.85	0.10	0.50	0.26	
20:5w3	20.41	20.53	20.94	23.07	30.21	16.41	16.87	12.18	15.18	
RS20:5	--	--	--	--	--	3.07	2.29	3.78	5.57	
21:5w2	0.36	0.57	--	0.30	0.29	0.29	0.10	0.09	0.14	
22:5w6	1.06	1.06	0.78	0.73	0.74	0.72	0.18	0.29	0.43	
22:5w3	2.70	2.63	1.59	1.75	3.45	2.06	2.10	1.01	1.45	
22:6w3	39.75	37.90	35.51	32.62	21.80	30.89	24.48	26.01	21.33	
RS22:6	--	--	--	--	--	4.29	5.22	5.98	6.31	
24X	--	--	--	--	--	--	--	--	--	

*See footnote, Table 5.

**See footnote, Table 5.

Table 8. RESULTS OF FATTY ACID ANALYSIS OF LIPID EXTRACTS FROM HADDOCK FILLETS USING AN EGSS-Y CHROMATOGRAPHIC COLUMN

Fatty acid*	Percent area under peak**									
	Nonirradiated sample		0.2-Mrad sample			2.8-Mrad sample		5.6-Mrad sample		
	Storage, days		Storage, days			Storage, days		Storage, days		
	0	15	0	15	32	0	32	0	32	
14:0	1.36	0.90	1.01	1.06	1.06	1.10	1.21	1.56	1.10	
15:0	0.53	0.47	0.44	0.45	0.83	0.47	0.37	0.63	1.01	
16:0	29.30	28.78	44.94	31.75	31.01	35.76	34.33	31.28	31.98	
17:0	1.27	1.43	1.36	1.49	0.95	1.02	0.91	1.15	0.97	
18:0	9.48	5.44	5.45	4.83	5.23	6.75	6.27	6.58	5.18	
16:1w7i	3.03	10.0	3.55	5.18	5.66	4.77	4.35	6.02	5.47	
17:1w9	0.94	0.79	1.58	1.16	1.12	1.05	0.78	0.74	1.45	
18:1w9	13.59	11.76	12.36	16.98	14.29	13.19	14.59	17.01	15.66	
20:1w9i	1.08	1.04	1.74	1.53	0.69	1.57	1.53	2.50	0.89	
22:1w11i	--	--	--	--	--	--	--	--	--	
24:1i	--	--	--	--	--	--	--	--	--	
16:2w4	0.78	0.75	1.13	0.86	0.60	1.00	0.55	0.68	0.81	
18:2w6	1.68	1.04	2.08	1.44	1.19	1.21	1.50	1.67	1.95	
20:2w6	0.14	0.11	0.24	0.11	0.09	0.17	0.16	0.23	0.06	
18:3w6i	0.31	0.32	1.01	0.26	0.32	0.33	0.30	0.10	0.30	
20:3w6	0.04	--	--	0.08	0.06	0.09	0.07	0.10	0.07	
18:4w3	0.65	0.24	1.01	0.27	0.57	0.30	0.29	0.41	0.25	
20:4w6	2.58	2.75	1.54	2.54	4.09	1.58	1.63	1.41	1.88	
RS20:4	--	--	--	--	--	0.39	0.44	0.52	0.85	
20:4w3	0.21	0.19	0.28	0.27	0.29	0.35	0.34	0.58	0.55	
20:5w3	13.15	13.40	8.72	13.45	18.60	10.28	9.04	7.39	9.13	
RS20:5	--	--	--	--	--	3.09	2.70	2.70	4.18	
21:5w2	0.65	0.52	0.34	0.27	0.51	0.22	0.27	0.23	0.38	
22:5w6	0.33	0.34	0.38	0.24	0.28	0.21	0.20	0.25	0.21	
22:5w3	1.14	1.18	0.41	1.02	1.50	0.67	0.75	0.75	0.72	
22:6w3	16.11	17.95	10.48	14.43	10.14	11.84	14.07	11.22	10.18	
RS22:6	--	--	--	--	--	2.09	2.80	3.53	4.25	
24X	1.09	0.58	0.54	0.33	0.32	0.50	0.55	0.49	0.57	

*See footnote, Table 5.

**See footnote, Table 5.

Four of the fatty acids identified (16:0, 18:1w9, 20:5, and 22:6w3) are predominant components of cod and haddock lipid samples as well as representatives of their major classes of lipids.⁶⁰ Phospholipids, which are well dispersed in muscle tissues since they are included in membrane structures and are involved in the physiological functions of muscle tissue, are richer in polyenes (such as 20:5w3 and 22:6w3) and saturated fatty acids (such as 16:0) than are triglycerides. On the other hand, triglycerides are rich in monoenes (such as 18:1w9) and are not intimately dispersed in muscle tissue. Consequently, a consideration of the location of a fatty acid molecule in situ and its chemical reactivity in vitro may aid in interpreting the results. For example, the decreases in 20:5w3 and 22:6w3 are not surprising, because these acids are concentrated in the lecithins, cephalins, and sterols and are highly reactive. Larger increases in the amount of 16:0 obtained from the irradiated sample might be expected because of its relative chemical inertness except that phospholipids contain about twice as much 16:0 as ^{do} nonphospholipids. Similarly, recovery of 18:1w9 is not surprising in view of its high concentration in triglycerides and its chemical reactivity.

The interpretation of the effect of storage is based on the potential for microbial or autolytic enzymatic activity in the samples during storage. In general, storage seems to have little effect on fatty acid composition. Lipid hydrolysis is assumed to occur, because lipids of non-irradiated fish muscle are ^{known to} deesterify during chilled storage; freezing

treatments, which cause disorganization of muscle tissue, can accelerate this reaction.⁶¹ The effect of radiation on the disorganization process in muscle tissue or on the rate of lipid hydrolysis has not been studied extensively, but there are indications that nonesterified fatty acids can be produced.⁶³ Present results indicate that, ^{even} if nonesterified fatty acids are formed during storage, they are not subsequently oxidized to any appreciable extent.

Nawar et al.⁵³ have suggested that the effect of radiation on lipids might become the basis for a test to determine whether or not seafood has been irradiated. Two obvious requirements are that the change caused by radiation be measurable and that the degree of this change be unaffected by storage of the seafood either before or after irradiation. Determination of polyunsaturated fatty acids or possibly the three new components might fulfill these requirements, but more study is needed to establish the reliability and practicality of any suggested method.

General Discussion (3)

Irradiated and nonirradiated cod and haddock fillets have been chemically analyzed for evidence of (1) the effect of radiation (using unstored samples), ⁽²⁾ the effect of storage (using samples given identical irradiation treatments), ⁽³⁾ and similarities or differences between the two species. The effect of radiation dose is shown by the fact that gas chromatograms of 0.2-Mrad-treated samples were more similar to ^{chromatograms} of untreated samples than to chromatograms of samples given 2.8 or 5.6 Mrads. The observed changes in the chromatograms were usually continuous with dose, except for some of the volatile components. ^{The} similarities between cod and haddock chromatograms ^{the} far outweighed ^{the} differences, which is not surprising in view of the phylogenetic

and ecological similarities of these two species. This comparison is based on only one lot of each species, however, and seasonal variations between species can be expected. The various effects of storage have been explained in terms of microbial ^{or} enzymatic spoilage; consideration of these results indicates that an after-the-fact irradiation test is possible but needs more work.

The available evidence suggests that further study is needed of the effects of radiation on tissue structure or function. The observed decreases in polyunsaturated fatty acids and increases in free amino acids support the hypothesis⁶⁴⁻⁶⁷ that radiation alters the structure of cellular membranes. ^{Conceivably,} a simple colorimetric test based on altered membrane permeability could be developed to determine whether or not seafood has been irradiated.

The gas chromatographic results support the original rationale that radiation fragments a wide variety of chemical compounds in situ into several types of smaller molecules. Further work to determine which volatile components are most responsive to radiation might lead to another test to tell whether or not seafood has been irradiated. The chemical pyrolysis test of MacGee⁶⁸ warrants study because it is sensitive, reliable, and simple. Some of these proposals for future study may be undertaken with Bureau of Commercial Fisheries support. The Bureau is already supporting work to identify components by mass spectrometry; the results will be published separately. (FEM)

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A report of a symposium in Munich, 1969, on the
UTILIZATION OF LARGE RADIATION SOURCES AND ACCELERATORS IN
INDUSTRIAL PROCESSING*

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Abstract. A brief review of the Symposium on the Utilization of Large Radiation Sources and Accelerators in Industrial Processing held in Munich, Federal Republic of Germany, August 18-22, 1969, is presented. The main topics covered are chemical synthesis, polymerization and modification of polymers, dosimetry, and engineering.

Supplementary Keywords: (CONFERENCE / SYNTHESIS / PLANT SCALE / PILOT-PLANT SCALE / SOURCE, BETA / SOURCE, GAMMA / ACCELERATOR / DOSIMETRY / RADIOLYSIS / POLYMERIZATION / MODIFICATION, MATERIAL / GRAFTING / MODIFICATION, FIBER / COATING / CROSS-LINKING / ADDITIVE / SOURCE PREPARATION / INDUSTRY, STONE, CLAY, AND GLASS / INDUSTRY / INDUSTRY, TEXTILE / INDUSTRY, PLASTICS / INDUSTRY, CHEMICAL / INDUSTRY, WOOD / INDUSTRY, RUBBER / COBALT-60 / STRONTIUM-90)

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(Introduction)

Recent advances in radiation chemistry and radiation processing have stimulated research and development programs in the chemical and plastics industries. Successful commercial processes for polymerization have been developed through the application of gamma or electron-beam irradiation to initiate chemical changes. Following two previous symposiums in Warsaw 1959 (Ref. 1) and in Salzburg 1963 (Ref. 2), the current symposium,³ sponsored by the International Atomic Energy, highlights recent activities in fundamental research, process development, engineering, and operations in the use of large radiation sources and accelerators. One hundred eighty-six participants from 29 countries and 5 international organizations attended the symposium. There were nine sessions for the presentation and discussion of 50 papers. The four main topics were:

- Chemical synthesis
- Polymerization and modification of polymers
- Dosimetry
- Engineering

Extensive activities were and particularly noticeable relative to polymerization and modification of polymers, included grafting, surface curing, textile treatment, composite materials, and the vulcanization of elastomers. The trend in radiation processing is toward diversification to further exploit the technology. Nevertheless, the symposium included many papers that merged theory and practice.

Chemical Synthesis (3)← Pilot plants (4)

Pilot plants for radiation-induced chemical processes were described and both ^{60}Co and accelerators have been used as the source of radiation for initiating reactions in flow systems.

The use of an accelerator for the gas-phase synthesis of hydrogen chloride was reported by Konkov.⁴

Danno⁵ described the chlorination of dichloroethane at the Takasaki Radiation Chemistry Research Establishment, Japan, using ^{60}Co sources arranged in two different configurations— inner-source and outer-source types. The effect of flow rates and dose rates on the product distribution of polychloroethanes has been studied and compared with the results of batch studies.

Rösinger⁶ reported the sulf oxidation of normal paraffins. The reaction rate was slow at low dose rates, and was appreciably affected by the mass transfer rate of gaseous reactants. The use of polychloromethanes and acetic anhydride to increase the rate of sulf oxidation was reported by Asinger and Saus.⁷

A performance analysis of a tubular reactor and a continuous-flow stirred tank reactor has been made at the Brookhaven National Laboratory, USA.⁸ Irradiation of aerated Fricke dosimeter solution showed the effect of absorbed dose on conversion. In large conversions, the rate of radial mixing in the once-through tubular reactor and the degree of segregation in the stirred reactor caused deviations from either plug-flow or complete mixing in the reaction system. These findings provide the basis for a rational approach to reactor design in radiation processes.

← Radiation chemistry (4)

Swallow⁹ reviewed the use of pulse-radiolysis techniques to study active species, including hydrated electrons, anion complexes of halogen atoms, excited states, and free radicals of aromatic compounds. Williams¹⁰ discussed ion-recombination processes and the phenomena of trapped electrons in the glassy state or crystalline structure.

The radiation-induced addition reactions involving the ethylenic double bond in chloroolefins and the reaction of 2-propanol with maleic acid were the other topics on chemical synthesis.

Polymerization and Modification of Polymers (3)

The fundamentals of radiation-induced copolymerization were reviewed by Hummel¹¹ who described copolymer systems containing ethylene, fluoromethylene, isobutene, and sulfur dioxide. Williams¹⁰ discussed the role of free ions on ionic polymerization and the effect of extreme dryness in such a system.

← Solid-state polymerization (4)

The solid-state polymerization of trioxane, which has entered the stage of pilot-plant studies, was reviewed by Hayashi,¹² who also discussed the topographic structures of polyoxymethylenes. Tabata and co-workers illustrated the features of the nucleation process in the solid-state polymerization of itaconic esters. Through a measurement of the lifetime of positronium they observed the effect of nucleation on the induction period of a typical solid-state polymerization reaction. Kiss¹⁴

reported that the polymerization of acrylonitrile in the channel complex of urea was another example of solid-state polymerization leading to the formation of oriented crystalline polymers.

← Graft polymerization (4)

Machi and Silverman¹⁵ discussed the grafting of styrene to polyethylene and also to ethylene-propylene copolymer. Nonuniform viscosity of the amorphous region in polyethylene influenced the covalent grafting rate. The occlusion of homopolymers of styrene in the polyethylene matrix was studied by solvent extraction.

Seguchi and Tamura¹⁶ conducted electron-spin-resonance studies of methyl methacrylate with polyethylene. Both alkyl and allyl radicals initiated grafting reactions, the allyl radicals were more reactive.

Laizier and Wajs¹⁷ reported the improvement in hydrophilic properties and gas permeability of silicones grafting with N-vinylpyrrolidone. Although on resulting from

← Modification of textiles (4)

In the radiation modification of textiles Hoffman¹⁸ explained the kinetics of grafting reactions and stressed the role of the diffusion of monomer molecules into the matrix of fibres. The current status of the technology for durable press was reviewed and the improvement of wetting properties of synthetic fibres by grafting with hydrophilic monomers was discussed with respect to surface energy.

Blin et al.¹⁹ reported the vapour phase grafting of acrylic acid to polyester fibres for the improvement of hydrophilic properties.

← Surface curing (4)

Electron-beam curing of surface coatings has become a commercial success very recently. Blin and Gausse²⁰ reported the effect of dosage on the gel formation of unsaturated polyesters, acrylic esters, and epoxy acrylates. The effect of oxygen on the properties of cured films was also investigated.

The use of polyester-polyethylene film along with unsaturated polyesters for the surface treatment of fiberboard was reported by Miettinen²¹ and the economics of this process was compared with that of the peroxide catalyzed process discussed by Rotkirch.²²

Economics of aluminum or steel coil coating was the subject of Morgenstern's paper²³ which / the adaptability of electron-beam curing to high-speed production lines. A drastic reduction of operating cost is the merit of radiation curing, which could be of real significance if much higher line speeds (approximately 1000 ft per min) are considered. illustrated

← Vulcanization of elastomers (4)

Balestic²⁴ reviewed the dose requirement in the cross-linking of natural and synthetic elastomers, with special emphasis on the vulcanization of elastomers based on polyfluoroethylenes.

The properties of the films obtained by radiation-induced vulcanization of natural rubber latex were discussed in a paper by Laizier et al.²⁵ Additives, including carbon tetrachloride, divinylbenzene, etc., were used to reduce the dosage requirement for the vulcanization of natural rubber and polychloroprene.

The effect of irradiation on the cross-linking of elastomers with allyl, acrylic, and vinyl compounds has been extensively investigated by Balestic²⁶ and his group. The mechanical properties of such combinations were evaluated to test their applicability as binders for explosives.

← Composite materials including wood-plastic combinations (4)

The impregnation of concrete with ~6% vinyl monomers followed by gamma-irradiation is a current research program of Brookhaven National Laboratory, USA. Manowitz²⁷ reported on recent developments relative to and the results of corrosion tests / the handling of hot brine that may be encountered in water desalination plants. Monomers that can be cross-linked are particularly promising for this purpose.

A premixing technique followed by radiation curing was used by the Union Industrial Research Institute,²⁸ Republic of China, to make composites from red mud, sisal, and unsaturated polyester-styrene mixtures. Sufficient grafting of the polymer to the inorganic components was observed by X-ray diffraction examination of the composites. The impact strength of the composites was correlated to the effect of process variables by means of response surface technology.

Laizier et al.²⁹ reported on impregnation systems containing a ternary mixture of monomer-solvent-water for the treatment of wood. By varying the composition of the system it is possible to obtain different degrees of treatment. A comonomer system with vinyl carbazole was also found to be effective in modifying the cellular structure of wood. The grafting of polymer to wood was investigated by means of electron micrograph, hydrolysis, delignification, ^{on} and histochemical methods.

Dalton³⁰ reported the UKAEA's activities on the product applications of wood-plastics based on an acrylonitrile-styrene (60:40) mixture. The durability of these products was shown during their use in carton and pattern production. Research activities in India and Austria were reported by Jayaraman³¹ and Proksch,³² respectively.

← Pilot plants (4)

Stahel and Stannett³³ used a recirculating-flow reactor to study the polymerization of vinyl monomer systems. The reaction zone was confined to a tubular section in the circulation loop, and, by varying flow-rates in the system, the reactor can be used to study polymerization under conditions ranging from plug-flow to completely mixing. They investigated ionic homogeneous polymerization of styrene and also α -methylstyrene. Emulsion polymerization ^{of} styrene and vinyl chloride was also studied.³⁴

Pilot-plant studies on the vulcanization of natural rubber latex were reported by Icre.³⁵ With an accelerator it has been possible to process two tons per hour of latex in a continuous-flow tubular reactor system.

Dosimetry (3)

The distribution of flux during irradiation can influence localized rates of chemical changes as well as product informity. The current practice of dosimetry and the comparison of various methods were reviewed by Holm³⁶. In fluid systems the effects of flow characteristics and degree of mixing were discussed in a paper by Hill and his co-workers.⁸

The importance of dose-depth distributions in electron-beam irradiation has been shown in the cross-linking of polyethylene insulation used in cables and wires. The approach through the backscattering effect as revealed by Wiesner³⁷ was to circumvent and overcome the difficulties associated with dose-distribution problems. A theoretical analysis of electron beams from both line sources and plane-parallel sources was performed to correlate the angles of scattering, geometry, and structure of targets. By using Goudsmit and Sanderson's multiple scattering theory, Kniedler and Silverman³⁸ lumped successive collisions into one multiple collision and proposed four computer programs for the solution of boundary crossing and angular distribution of scattering problems.

On the measurement side, a radiochromic dye system incorporated into films, paper, or gels was described by McLaughlin and Hussmann.³⁹ After irradiation, the dyes form stable excited-state molecules to sustain color stability. The image contrast and resolution of dye films were reproducible for dosimetric purposes over a wide range of electron and gamma-ray energies. Dvornik⁴⁰ reported the use of an ethanol-chlorobenzene dosimeter for electron-beam dose rates up to 10^3 rad/sec.

Engineering (3)

Only the papers referring to the design of radiation sources and accelerators are described here since pilot-plant and reactor-design development has been summarized under the chemical synthesis and polymerization sections.

Electron accelerator design activities in the USSR were reported by Abramyan⁴¹ who summarized the various models from the compact 0.5 MeV transportable assembly to machines of 1-3 MeV with a power output between 10-30 kW. A high-intensity proton accelerator of 4 to 5 MeV was also mentioned. In Konkov's⁴² work, the magnetic focusing principle has been used to concentrate the beam. These accelerators were used in the cross-linking of polyethylene, grain disinfestation, etc.

The fabrication of beta sources from strontium silicate beads for process irradiation was reported by Case⁴³. Oak Ridge National Laboratory, USA, has designed thin-walled capsules that can provide approximately 30% of the energy emitted from ^{90}Sr . of This laboratory

Christen⁴³ described the design for housing a ^{60}Co source on roof shieldings to facilitate the transfer of source elements and also a stack conveying unit for the handling of bulk materials.

Miscellaneous (3)

Pastuska⁴⁴ described the radiation-induced reduction of chromium salt from the hexavalent to the trivalent state, in the presence of glutaraldehyde in the tanning of hides, and Cramer et al.⁴⁵ reported improvement in wet strength of paper by grafting.

Compton et al.⁴⁶ investigated the effect of radiation on the oxidation, settling rate, and sedimentation of solids in waste waters and sewage sludge.

Silverman⁴⁷ summarized the general aspect of this symposium.

(REG)

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RADIATION PROCESSING OF POULTRY WASTES

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Supplementary Key Words:
(POLLUTION/SOURCE, GAMMA/AGRICULTURE/FACILITY DESCRIPTION/COBALT-60)

Abstract. A system containing a 50-thousand-Ci ^{60}Co source for radiation processing of poultry litter is described.

The outstanding success of the U. S. Poultry industry in producing highly efficient farms for the production of meat and eggs in enormous quantities has resulted in some areas having serious waste handling problems. The problem of disposing ^{of} poultry wastes is further accentuated by the tendency of the industry to concentrate in localized areas, such as Sullivan County, New York, and the Atlanta, Georgia, area. Because of the increasing pollution from poultry wastes, the Federal Government has awarded Cornell University a large grant to study the problem.

The inefficiency of poultry in utilizing the nutrients in feed has long been recognized and numerous studies have been carried out to evaluate the reuse of poultry wastes (litter) in feeding studies for poultry, swine, sheep, and cattle.¹⁻⁵ The performance of animals fed poultry wastes with various bases has generally been good, and in some cases, superior to that of control animals, indicating that they were able to derive considerable feed value from the material. Treatment of the litter used in these studies varied from none to, at most, thermal drying. In most of these studies, roughage and other feed materials were added in various amounts. The major finding was that, as the poultry waste content of the feed increased, the palatability and intake decreased. Other problems mentioned by these investigators were dustiness of dry litter, unpalatability

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of wet litter, ^{the} presence of foreign objects such as metal, and the possibility of chemical residues and disease organisms.

These findings suggested that many of the problems might be eliminated if radiation treatment of the poultry wastes could be combined with a limitation of the amount of waste in the reconstituted feed. Research studies on irradiation of sewage sponsored by the U.S.A.E.C. and other organizations have shown that irradiation increases sedimentation, enhances oxidation of chemicals, destroys bacteria, and eliminates odors.⁶⁻¹² The Sanitary District of Greater Chicago has incorporated these findings into a design study of an irradiation facility for processing about 2 million gal of sewage sludge per day. The use of about 4 million Ci of ⁶⁰Co or ¹³⁷Cs should decrease the cost¹³ of sludge disposal from \$57 to about \$16 per ton.

American Nuclear Corporation, Oak Ridge, Tenn., has developed an irradiator¹⁴ for sewage and animal wastes which is almost 100% efficient in utilization of the gamma energy from ⁶⁰Co or ¹³⁷Cs. The flow-through design uses several passes by the centrally located sources to ensure uniform dosage to the liquid. The total dose to the liquid is determined by the activity of the sources and the flow rate through the irradiator.

A unique feature of the design is the provision for removal of the solids from the inner walls of the irradiator, which decrease the radiation dose to the liquid and impede flow.

The first system using this irradiator is on order by Agritomics, Inc., Washington, D. C., for treatment of poultry wastes (Fig. 1). The radiation source is 50 thousand Ci of ⁶⁰Co, and the system is designed for a capacity of 60 tons of aqueous slurry per day containing 17 to 18%

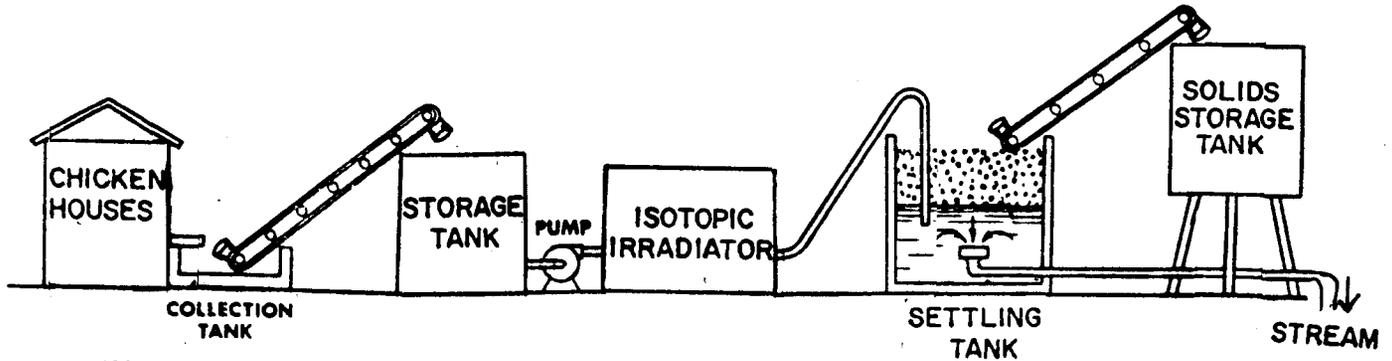


Fig. 1. Poultry-waste treatment system using radioisotopic irradiator.

solids. The slurry will receive a radiation dose of 120 / thousand rads, which is sufficient to kill most of the bacteria and will enhance rapid separation of the solids. This dosage is about 15 times as great as that planned by the city of Chicago for treating sewage sludge. Adequate provision has been made for adjusting the flow rate to obtain the desired level of pasteurization and for compensation as the 6000 decays.

The significance of irradiation of poultry wastes is that an obnoxious and troublesome by-product of the poultry industry is converted to a safe and valuable product. The granular, odor-free product may be blended back into feed at the 10 to 15% level to obtain an excellent feed with full nutritional values for poultry, cattle, swine, sheep, etc. The product also may be used directly as feed for commercial fish and oyster farming and is potentially useful as a fertilizer for vegetable farming. (MG)

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^{133}Xe PULMONARY FUNCTION ANALYZER

A. S. Blum*

Supplementary Keywords: medicine; diagnosis; tracer; xenon-133; ventilation

Abstract. A mobile system is described for the automatic/separate measurement of the function of each lung. External measurement of the gamma radiation of ^{133}Xe eliminates the need to seal catheters within the bronchial tree. Lung volume data are computed and displayed digitally in liters. A volumetric maneuver that calibrates the entire system, including the patient, makes this computation possible. The radioactive gas, supplied in a tank of compressed air, is metered and dispensed by an electrically operated valve. Radioactivity in exhaled air is removed by a trap; this minimizes the problem of handling radioactivity and avoids environmental contamination.

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(Introduction)

The hope of arresting many lung diseases, such as emphysema and bronchogenic carcinoma, rests upon the ability to detect lung pathology before so much function is lost that the disease is clinically manifest. In a study of 2000 subjects, Hutchinson¹ found that spirometric measurement of Vital Capacity could be predicted on the basis of height and age. A reduction of this volume below predicted levels accompanied, and often preceded, clinical manifestation of lung disease. His work was published in 1846, but a reduction in spirometric ventilation values below predicted levels is still recognized as a sensitive indicator of lung pathology.

If ^{is measured} the function of each lung separately, ^{is achieved,} even greater sensitivity ^{data} by virtue of asymmetry, before the sum of the two lungs deviates significantly from predicted values.

There is another vital requirement for quantitative pulmonary function data from each lung. When a surgeon is contemplating removal of a lung, or ^a portion thereof, he must know if there will be enough function in the remaining lung tissue to support life.

Pulmonary Function Analysis (3)

④ The total volume of gas in the lungs at full inspiration is the Total Lung Capacity (TLC), which cannot be measured directly. The maximum amount of gas that can be moved from

full inspiration to full expiration is the Vital Capacity (VC) which

is easily measured by blowing into a spirometer. The amount of gas remaining in the lungs at full expiration is the Residual Volume (RV), which cannot be measured directly; gas dilution techniques and a gas analyzer are required. The sum of VC and RV equals

TLC. One of the most useful indicators of airway obstruction is the Forced Expiratory Volume One Second (FEV_1), the volume of gas that can be exhaled in 1 sec from full inspiration with maximum effort. This is measured by a spirometer connected to the patient mouthpiece and recording the sum of the ventilation of both lungs.

To get these data for each lung separately requires the technique of bronchospirometry. With ^{the patient under} local anesthesia, a four-lumen catheter is inserted into the trachea. One branch is sealed in the left main bronchus with an inflated cuff; another branch is sealed in the trachea and receives gas from the right lung. Gas from each lung flows to a separate spirometer and gas analyzer. Because of the small catheter lumens, no information on flow rate (FEV) is obtainable; ^{and} because of the stress to the patient, ^{meaningful} capacity values in liters are not obtained. Instead, relative volume on each side is multiplied by total volumes found by previously performed conventional spirometry. Although bronchospirometry was introduced² in

the 1930's, very few centers perform the procedure, even for preoperative evaluation.

Radioactive Gas Technique (3)

Knipping³ and associates first used radioactive gas to study pulmonary function at the University Clinic in Cologne in 1953. Multiple collimated detectors were placed against the chest to record the varying amounts of radiation in different regions of the lungs as the gas was inhaled and exhaled. Investigators in Montreal,⁴ London,⁵ and Malmo⁶ expanded on these ventilation techniques and introduced perfusion measurements as well.

When the relatively insoluble gas is injected intravenously, it is released in the first gas space it reaches. The amount of radioactivity in a lung region is then directly proportional to the pulmonary perfusion of that region.

All of the published techniques convert detector impulses into analog signals with ratemeters. The signal is recorded and subsequently converted to digital form for computation. These studies have resulted in a much greater understanding of man's pulmonary physiology in both normal and disease states.)

However the equipment is cumbersome, the data processing laborious, and the results are in relative terms, not liters, because externally

measured radiation levels cannot be converted to ^{gas volumes.} / An excellent review of the subject is presented by Miorner⁶ along with the most practical clinical technique for relative regional ventilation perfusion measurements.

These measurements can be made simultaneously, safely, and automatically by a new pulmonary function analyzer using the gamma-emitting, inert gas isotope ^{133}Xe . No catheters are used, only a mouthpiece and noseclip. Values ^{in liters} for each lung's VC and RV and FEV₁ expressed as percent of that lung's Total Capacity can be displayed in digital form without any calculations by the operator. After an ^{intravenous} injection of the gas in solution, the fraction of the total pulmonary blood supply perfusing one lung can also be displayed.

The new ^{133}Xe technique has different objectives:

- (1) Quantitative volumetric data in liters for each lung separately,
- (2) Data presentation in readily interpretable format (classical spirometric values) for comparison with predicted norms, and
- (3) Simplicity of gas handling with automatic computation.

Description of the Instrument (3)

The ^{133}Xe pulmonary function analyzer is a completely self-contained system ^(Fig. 1) for the automatic collection and display of information on the ventilation and perfusion of each lung. The subject is seated (Fig. 2)

with his back against a

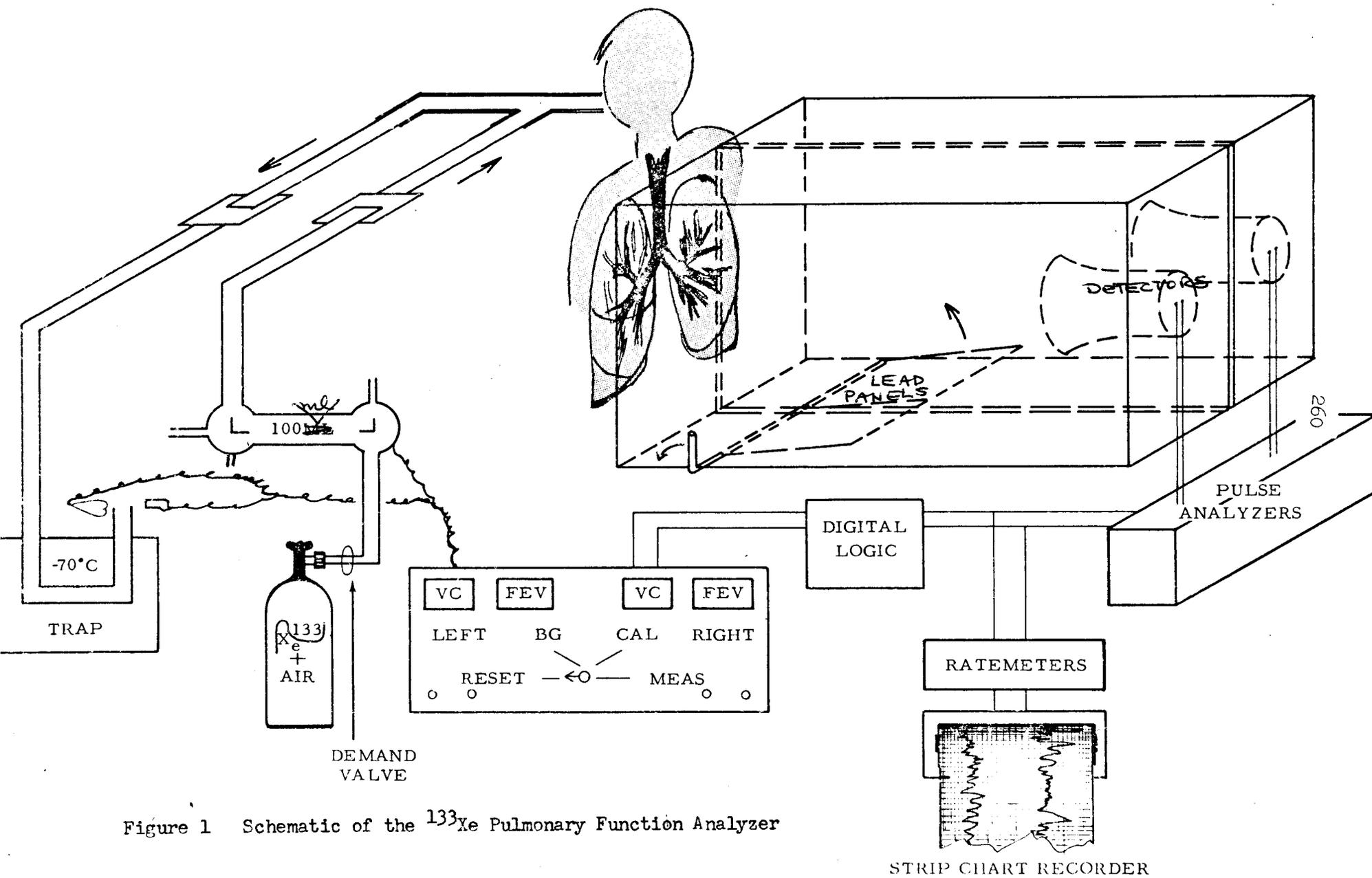


Figure 1 Schematic of the ^{133}Xe Pulmonary Function Analyzer

double box, his spine along a central partition. The mouthpiece and valve are suspended in front of the patient. They connect by hoses to a tank of ^{133}Xe -compressed air mixture and an exhaust trap (Fig.3). On one side of the unit are strip-chart recorders for analog display of the counting rates over each lung. A selector switch and "start count" push-button are the only operating controls. Numerical data are calculated automatically and displayed by the usual Nixie tubes.

In keeping with the dictum primum non nocere,* a special mouthpiece has been designed so that the portion of the mouthpiece where incoming air mixes with exhaled air can be replaced after each patient, to eliminate the danger of cross contamination between patients. This problem has too often been ignored in the design of pulmonary function apparatus.

The inert gas ^{133}Xe is either injected intravenously in saline solution for perfusion studies or inhaled in an air mixture from a pressurized tank for ventilation studies. Activated charcoal at -70°C adsorbs xenon from exhaled air.

A NaI crystal scintillation detector, centered over each lung, measures radiation from the xenon gas in that lung. The crystals are located at the back of the double box, one on each side of the central vertical partition. The walls and central vertical partition are lead-lined so that each crystal is at the rear of an elongate lead chamber, closed at the open end by a single lung. The detector is far enough from the chest (18 in.) that sensitivity is relatively uniform from apex to base and front to back.

*"First of all, do no harm."---Ed.



Figure 2 Test subject with back to detectors and nurse holding mouthpiece of pulmonary function analyzer. Digital output can be seen on lower right section of analyzer.

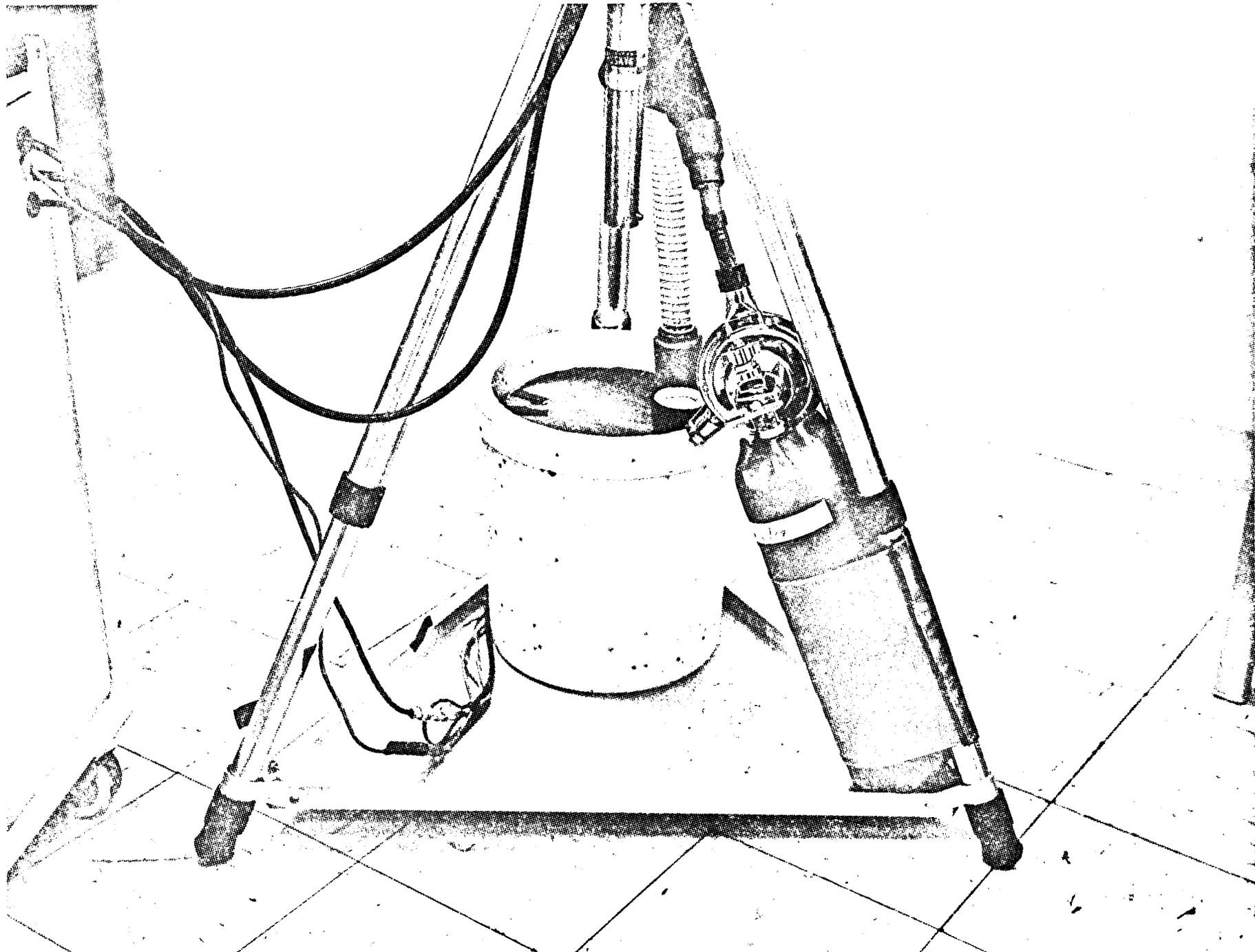


Figure 3 Gas handling system and ^{133}Xe trap for pulmonary function analyzer.

The lead partition and the spine effectively separate the radiation from the two lungs. The amount of "cross talk" was tested in a patient whose right lung had been removed (Fig. 4). Essentially all the volume measured was in the remaining lung.

The impulses from the two detectors pass through pulse height analyzers, tuned to the 81-keV gamma energy peak to reduce scatter and background. The selected pulses then pass into ratemeters whose outputs go to a two-channel strip chart recorder, which provides analog recordings of radiation intensity versus time as gas goes into and out of each lung. However, the use of ratemeters causes information loss. The impulses are simultaneously fed to a digital processing unit which integrates the pulses received ^{over} Δ fixed time intervals for optimum counting statistics. It performs ^{the} Δ discrete mathematical operations indicated by the multi-position selector switch.)

In contrast to the graphic analog data, these data are precise, and the results of the automatic computations appear as numbers on the control panel.

Results displayed are: Vital Capacity of left lung, in liters
 Vital Capacity of right lung, in liters

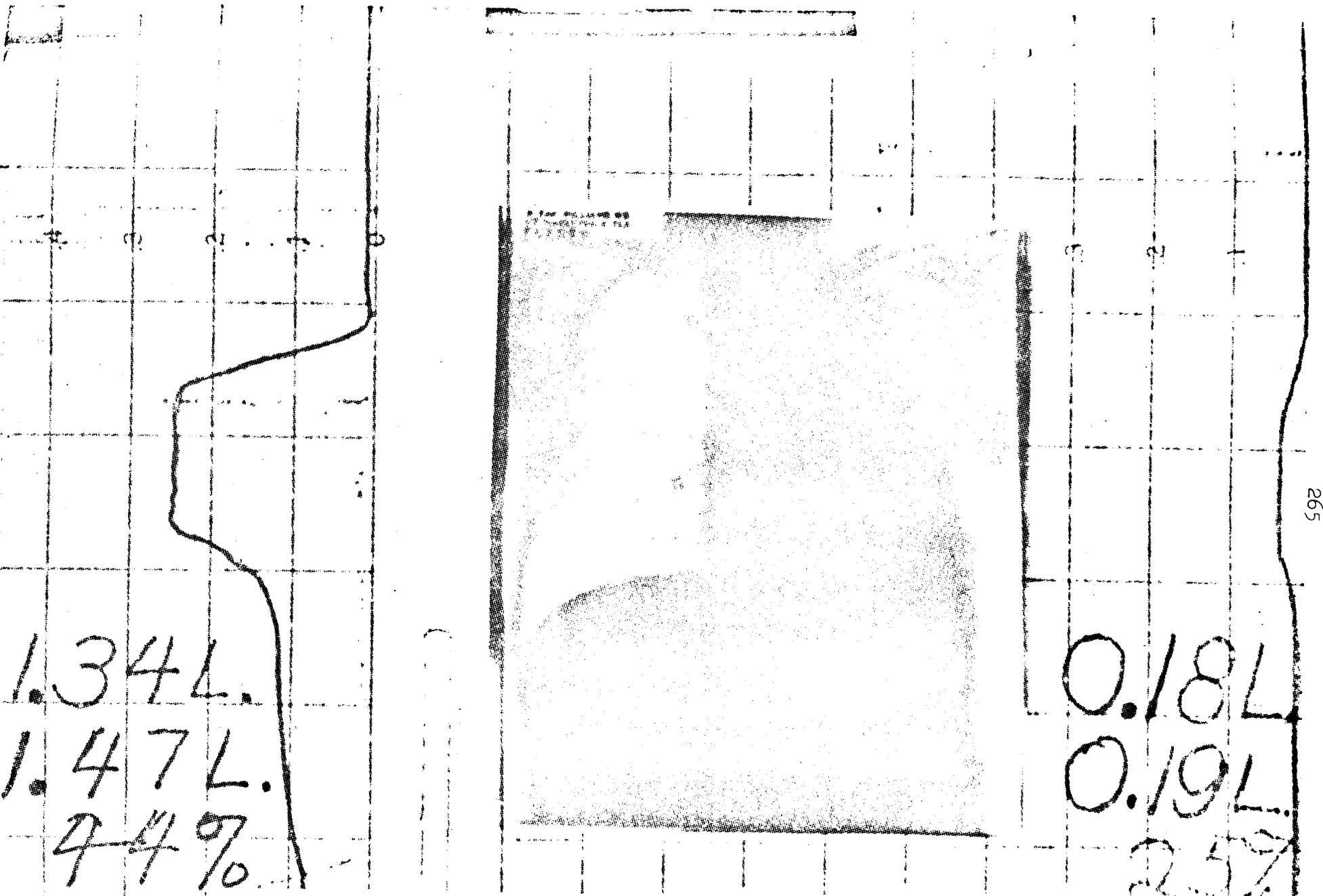


Figure 4 Duplicate readings of Vital Capacity in a patient with right lung removed. Essentially all the ventilation is seen to be on the unoperated side, showing how well the instrument separates data from each lung.

(FEV₁ of left lung)/total left lung capacity

(FEV₁ of right lung)/total right lung capacity

Residual Volume of left lung, in liters*

Residual Volume of right lung, in liters*

System Calibration (4)

To convert radiation intensity (counts per unit time) to liters of gas, the system (which includes the patient's chest) is first calibrated by introducing a known volume^(0.1 liter) of the gas mixture into the subject's lungs. This is followed by room air to full inspiration. While^{the subject is} breathholding, the sum of counts from both detectors is stored during a 4-sec counting interval. The sum of xenon activity will be equivalent to 0.1 liters; the fraction in each lung need not be known if detector sensitivities are equal. The instrument now has inscribed in its memory the information of the counting rate of 0.1 liter of that gas mixture in that chest. From this information, absolute volumes in liters can be derived from counting rates while various ventilation maneuvers are performed.

*The instrument now in operation does not include the digital logic to compute these terms automatically. They are calculated manually from the chart and the VC, since calibration in liters is derived from VC in the equation for RV. - ASB.

Ventilation Determination (3)

The patient sits in position with the gas mouthpiece and noseclip in place; the valve provides room air, and exhaust air goes through the filter trap. The selector switch is moved from "reset" to "background," and the start count button is pressed. An indicator light remains on for 4 sec while the background count from each detector is stored in a memory. In every subsequent operation, these background counts will be subtracted from the total counts, so that the net counts used in the calculations will represent xenon entering the system.

After the indicator light signals that background counting is completed, the operator moves the selector switch to "calibrate" while the patient is exhaling; and the patient is instructed to take a full breath and hold it for 4 sec. The automatic valve connecting the xenon^(w)-compressed air tank to the inlet of the mouthpiece includes a pipe of 0.1-liter capacity, which has been filled previously with the radioactive gas mixture at atmospheric pressure. The valve is electrically switched, introducing the gas in the pipe into the fresh air stream being inhaled by the patient. The

gas is flushed into the lungs by the following fresh air so that none remains in trachea or mouth. While^{the subject is} breathholding, the radiation from the 0.1 liter of gas is counted for 4 sec. The counting is started by depressing the start-count button. When the signal light indicates completion of counting, the patient is instructed to breathe normally while the calibration gas is washed out.

The operation switch is turned to the fourth position "measure," while the patient is instructed to exhale completely, take a full breath, and hold for 4 sec. The automatic valve connects the mouthpiece to the ^(m) xenon-air mixture at atmospheric pressure so that the entire inspiratory vital capacity is filled with the same composition gas used for calibration. While^{the subject is} breathholding, the radiation is counted for 4 sec. The valve disconnects the xenon source automatically when the measure count is completed.

When the signal light^{again} indicates completion of counting, the patient is instructed to^{exhale} forcefully and completely. ← Exhalation triggers first a 0.5-sec timer, then a 1-sec count is automatically made. The measurement gives the gas remaining in each lung an average of 1 sec after the start of exhalation.

The patient now inhales fresh air to maximum inspiration and breathholds again for 4 sec, while another

count is made. This count reflects the volume of radioactive gas remaining in the lungs after complete exhalation, i.e., RV. The patient breathes fresh air normally until all remaining xenon accumulates in the exhaust-line trap. Washout curves are recorded, but no further digital measurements are taken of ventilation. Typical ventilation data are presented in Table 1.

Perfusion Determination (3)

Perfusion measurements are made after an intravenous injection of xenon gas in a saline solution. An estimated 95% of the dissolved gas is released to the air space in a single passage through the pulmonary alveolus. Since most of the gas is lost before blood reaches the left ventricle, gas spaces perfused by the bronchial circulation will not contain xenon, and xenon in each lung will be proportional to perfusion by the pulmonary circulation alone.

Measurement of pulmonary perfusion of each lung may be done at any stage of the respiratory cycle, but ^{the} counting interval should not be started until visual inspection of the rate-meter curves shows that the injection bolus has passed through the heart.

← ~~_____~~ A single 4-sec counting interval is used without calibration, since only fractional perfusion on each side is determined.

Table 1 Comparison of measured and predicted Vital Capacities and Residual Volumes for nine asymptomatic control subjects

Patient	VC _L , liters	VC _P , liters	Sum, liters	Predicted, liters	RV _L , liters	RV _R , liters	Sum, liters	Predicted, liters
P. W.	1.18	1.50	2.68	2.70	0.472	0.947	1.42	1.70
P. B.	1.77	1.91	3.68	3.55	0.664	1.10	1.76	1.51
L. K.	2.66	2.95	5.61	4.22	1.38	2.21	3.59	2.14
P. M.	2.47	2.50	4.97	4.36	0.967	1.25	2.22	1.85
L. W.	1.84	2.40	4.24	4.10	1.16	2.10	3.26	2.03
D. B.	2.31	2.14	4.45	4.07	0.735	1.07	1.80	2.06
E. N.	1.48	1.76	3.24	3.68	1.18	1.41	2.59	2.98
R. E.	1.89	2.89	4.78	4.65	0.576	1.17	1.75	1.97
J. P.	1.68	1.59	3.27	4.36	1.05	1.59	2.64	2.13

Derivation of Equations (3)

Within the data processing unit, the following values may be stored electronically, representing counts collected in fixed time intervals:

- A = full vital capacity breath
- B = background
- C = calibration
- D = count after 1 sec
- E = residue after complete exhalation
- P = count after injection

Subscripts L and R denote left and right, respectively, of chamber (i.e., lungs).

Vital Capacity (4)

$C_L + C_R$ = sum of radiation counts from both lungs after inhaling 0.1 liter of gas mixture.

$C_L + C_R - B_L - B_R$ = net counts due to 0.1 liter of gas in lungs after subtracting for background.

A - B = net counts due to full vital capacity of gas, at same concentration as the 0.1 liter.

$$\text{Then, } VC = A - B \quad (1)$$

$$\text{and } 0.1 \text{ liter} = C_L + C_R - B_L - B_R. \quad (2)$$

Dividing (1) by (2), and solving for VC, gives

$$VC = 0.1 \left[\frac{A - B}{C_L + C_R - B_L - B_R} \right]. \quad (3)$$

These values are all measured separately; therefore,

$$VC_L = 0.1 \left[\frac{A_L - B_L}{C_L + C_R - B_L - B_R} \right] \quad (4)$$

$$\text{and} \quad VC_R = 0.1 \left[\frac{A_R - F_P}{C_L \quad C_R \quad B_L \quad B_R} \right] \quad (5)$$

Residual Volume (4)

The gas is assumed to be mixed in the entire lung capacity after breath-holding at full inspiration; data obtained with helium indicates this is true. The net count after inhalation of full vital capacity was $A - B$. After mixing with the total lung capacity, the radioactivity is present in the same amount but diluted by the residual volume. Therefore, $A - B$ represents total lung capacity. When a vital capacity is exhaled, the net count remaining, $E - B$, is proportional to the residual volume; and the difference, $A - B - (E - B)$, is proportional to VC.

$$RV = E - B \quad (6)$$

$$VC = A - B - (E - B) = A - E \quad (7)$$

Dividing (6) by (7) and solving for RV,

$$RV = \frac{VC (E - B)}{A - E} \quad (8)$$

Since VC is in liters in equation (3), RV is in liters. Adding the specific notation for each lung,

$$RV_L = \frac{VC_L (E_L - B_L)}{A_L - E_L} \quad (9)$$

and

$$RV_R = \frac{VC_R (E_R - B_R)}{A_R - E_R} \quad (10)$$

Forced Expiratory Volume Ratio (4)

If forced expiration could be interrupted after 1 sec and the chest counted for 4 sec, the net count $D - B$ representing gas remaining in lung after 1 sec would be obtained. The ratio $(D - B)/(A - B)$ would be the fraction of the total lung capacity remaining, and $[1 - (D - B)/(A - B)]$ would be the fraction of the total lung capacity expired in 1 sec, i.e., FEV_1/TLC .

However, the chest is counted for only 1 sec, and the result is multiplied

by 4:

$$\frac{\text{FEV}_1}{\text{TLC}} = 1 - \frac{4D - B}{A - B} \quad (11)$$

Because expiration cannot easily be arrested at 1 sec while counting, the actual count is made around the 1-sec mark, i.e., from 0.5 to 1.5 sec after start of expiration. The results will deviate from the true 1-sec value to the extent that the rate deviates from a straight line during this period.

For the separate lungs, the equation becomes:

$$\frac{(\text{FEV}_1)_L}{(\text{TLC})_L} = 1 - \frac{4D_L - B_L}{A_L - B_L} \quad (12)$$

and

$$\frac{(\text{FEV})_R}{(\text{TLC})_R} = 1 - \frac{4D_R - B_R}{A_R - B_R} \quad (13)$$

Perfusion (4)

The equations for perfusion ratio in both lungs are similarly derived:

$$\frac{\text{Perfusion}_L}{\text{Total Perfusion}} = \frac{P_L - B_L}{P_L + P_R - B_L - B_R} \quad (14)$$

and

$$\frac{\text{Perfusion}_R}{\text{Total Perfusion}} = \frac{P_R - B_R}{P_L + P_R - B_L - B_R} \quad (15)$$

Statistics (3)

The radiation emitted by a radionuclide obeys the laws

of a true random phenomenon. Therefore, the standard deviation of a measurement equals the square root of the number of counts measured. The greater the number of counts collected, the greater the precision.

All the reported studies with xenon have involved first feeding the pulses to a ratemeter for conversion to analog form. The varying voltage output is then fed to strip-chart recorders, magnetic tape records, or both. The stored data, now in analog form, must be read out and converted back to digital before calculations can be made. Information is lost at every step, and each step adds to the cost and time of the operation. By adding all the counts received during the measurement interval in a scaler, the maximum available information is obtained, and the calculations are started directly with digital data.

The precision of measurement can be improved by increasing the sensitivity of the detectors, the time intervals, or the amount of ^{133}Xe used. The calibration volume of 0.1 liters is only 2% of the volume of gas used to determine vital capacity. This is the limiting point statistically. The calibration only affects the absolute values; it has no influence on measurement of symmetry. The calibration volume could easily be increased to 1 liter in normal adults to give either greater precision or to lower radiation doses by a factor of 10. The small volume was chosen so that the test could

be used in children and patients with severe lung disease and also could be introduced at any point of the respiratory cycle. There is evidence that the first bolus of air entering from FRC does not go to the bases. Taking the calibration volume close to tidal levels will probably give more valid results, especially in patients with severe lung disease; however, taking the calibration bolus at various levels of the respiratory cycle opens the way to more detailed study of pulmonary pathology.

Detector sensitivity can be increased by the use of large cross-section scintillation crystals. Large diameter, general-purpose scintillation detectors are quite expensive because the crystal must be thick enough (5 cm) to absorb high energy radiation. An instrument designed for the low energy of xenon can use a crystal less than 1 cm thick, and large diameter detectors are then economically feasible.

Radiation Exposure (3)

Lassen⁷ estimated the radiation exposure from inhalation of 1 mCi of ^{133}Xe per liter for 1 minute or injection of 5 mCi intravenously to be: tracheal mucosa, 96.8 mrad; lung, 17.5 mrad; and gonads, 1.1 mrad. To use his exposure data requires a conversion factor. In this work, a maximum concentration of 0.33 mCi/liter with estimated 6 sec breathing (4 sec breathholding) gives a conversion factor of

$$\frac{6 \text{ sec}}{60 \text{ sec}} \times \frac{0.33 \text{ mCi/liter}}{1 \text{ mCi/liter}} = 0.033.$$

Therefore, the ^{133}Xe pulmonary function analyzer gives doses of 3.2 mrad to tracheal mucosa, 0.58 mrad to lung, and 0.036 mrad to gonads. In the perfusion studies, a dose of 0.05 mCi is adequate; $\frac{0.05 \text{ mCi}}{5 \text{ mCi}} = 0.01$. Therefore, the perfusion studies results in doses of 0.968 mrad to tracheal mucosa, 0.17 mrad to lung, and 0.011 mrad to gonads.

According to the International Commission on Radiological Protection (ICRP), the highest dose allowed for personnel in radiological work for most organs is 300 mrad each week of their working lives. The usual dose for a diagnostic chest x ray given in a medical facility is 26 mrad to the lungs. The dose from an x-ray machine of the mobile van survey type is 220 mrad to the lungs. Therefore, the ^{133}Xe pulmonary function analyzer is a very low-dose diagnostic device.

Gas Handling System (3)

One of the obstacles to widespread application of xenon in clinical diagnosis is the same property which makes it so

valuable. No matter by what route it is administered, the gas is exhaled to the air within minutes. As long as the room air is rapidly replaced, there are no hazards to personnel or to the accuracy of measurement. Large air-collecting bags or hoods exhausting outside the building are most often used. These limit the convenience and mobility of the procedures.

With the pulmonary function analyzer, xenon for ventilation is automatically kept within the closed system. The gas has a bio-

logical half-life of ~1 minute, although its physical half-life is 5.27 days. It emits a 0.345-MeV beta ray and 0.081- and 0.031-MeV gamma and x rays. The gas

is provided in a small cylinder containing 150 liters of air and less than 0.1 mliter (50 mCi) of ^{133}Xe compressed to 2000 psi. The fittings and demand valve are commercial scuba gear. No tools are required to connect the tank to the system. The apparatus has a sufficient range of sensitivity that a tank may be used for at least 2 weeks with acceptable statistical accuracy.

The demand valve feeds gas at atmospheric pressure to the system. The negative pressure required to open the valve is so slight that it does not affect the normal subject's vital capacity as compared to conventional spirometry (Fig.5). This is not true for emphysema patients. The sum of their differential vital capacities is much less than that found

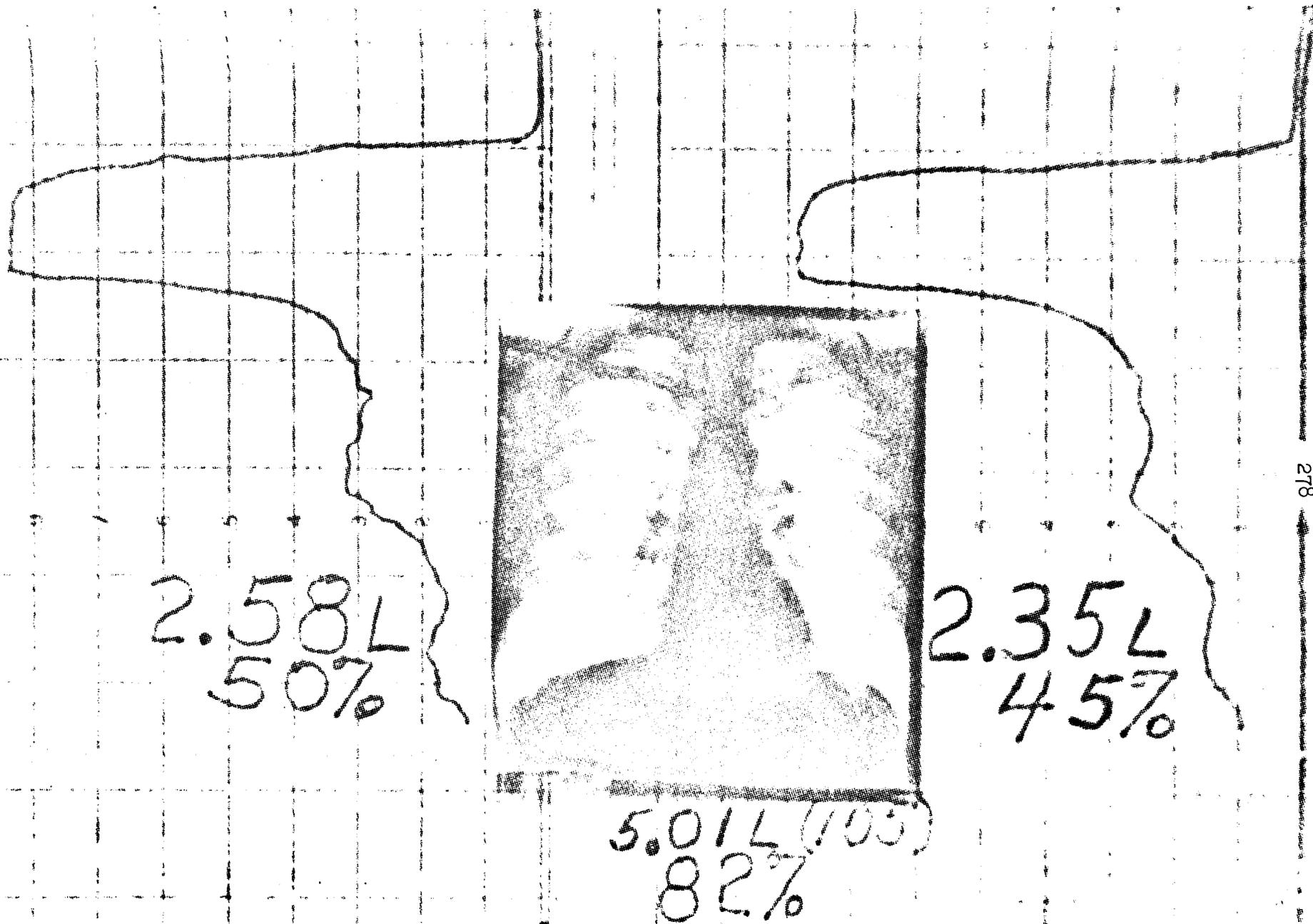


Fig. 5. Result of ventilation study of a normal individual. Digital and analog data are given on each side of the chest x ray. Upper figure is VC and lower figure is FEV₁; FEV₁ is given as percent of VC. Conventional spirometry data are shown below the x ray; VC in liters is followed by percent predicted in parentheses.

(Fig. 6).
 by spirometry / For these cases, a bag reservoir, provided in the gas inlet line, may be filled in advance. The bag

is not used for screening tests, making them sensitive to pathology although not registering a true vital capacity. When pathology is detected, the test can be repeated with the filled bag, giving true vital capacity.

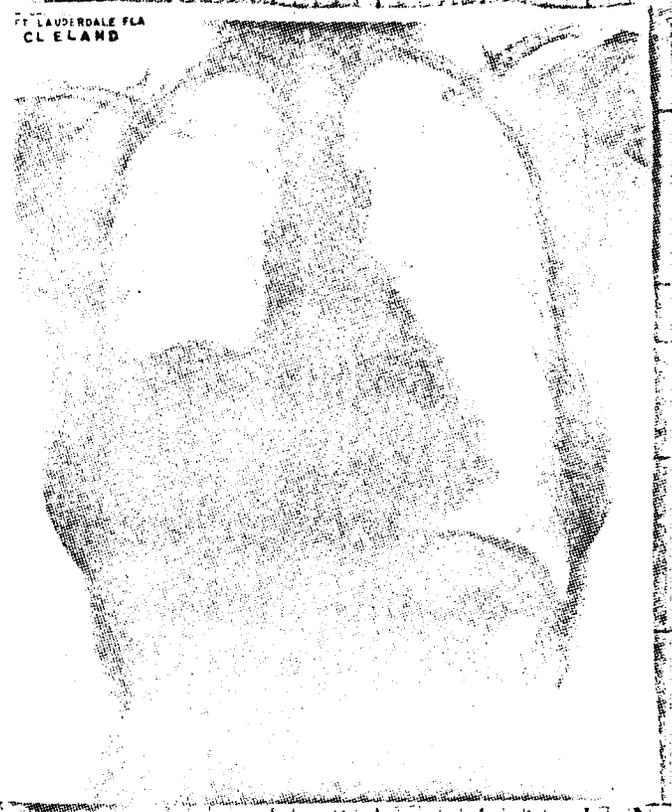
Xenon in saline for perfusion is dispensed from a sealed capsule directly into a syringe for injection. Exact dose is not critical, since only relative/
 perfusion on each side is measured.
Xenon Trap (4)

A specially designed xenon trap has been incorporated into the apparatus which passes all the patient's exhaled air but removes and retains all the xenon in a shielded container. Since this is a closed system, it may be used anywhere, even at bedside, without fear of contamination.

The trap consists of a set of baffles which conduct the exhaled air through four absorbers in series:

- (1) Indicating soda-lime granules (sodium hydroxide, calcium oxide) absorb CO_2 . At Dry Ice temperature, water is also removed.
- (2) Two activated charcoal filters adsorb xenon at -70°C .

0.44L
0.45L
32%



1.09L
1.08L
39%

2.45L (8%)
68%

Figure 6 Sensitivity of the ¹³³Xe analyzer. Patient had two-thirds of right lung removed. Conventional spirometry indicated VC was 86% of predicted value; pulmonary function analyzer correctly shows that only one-third as much function remains in right lung.

(3) Soda-lime granules, that perform no active function, protect the charcoal from CO_2 which might back-diffuse into the trap from the exit and saturate the charcoal, exhausting its adsorptive potential for xenon.

Low temperature is maintained by Dry Ice or mechanical refrigeration. Absorption of CO_2 by soda-lime is an irreversible reaction. Saturation is indicated by a color change visible through the transparent trap. Adsorption of xenon by activated charcoal at low temperature is a reversible phenomenon, and A ^{xenon} may be desorbed by warming A ^{the filters} to room temperature. Even if the trap is warmed, xenon is 4.5 times as heavy as air, and the entrance and exit A ^{of the trap} are at the top. In the absence of a current of air, the heavy xenon tends to remain in the trap. Unidirectional valves at entrance and exit prevent inhalation of exhaled air. Since these A ^{valves} have rubber flaps which seal off the air passages, the trap is closed when not in use. It would always be cold when in use.

The xenon may be pulled from the warmed trap and reused or discarded. However,

the isotope decays so rapidly and it is so easily shielded that the trap need not be ^{emptied.} / The trap holds 1 lb of charcoal, which has an enormous capacity--- more than 1,000 Ci. At 0.001 Ci/test, there

is no problem of saturation of the charcoal with xenon.

However, the large amounts of CO_2 and water in the breath could easily saturate the charcoal. This is prevented

by the preliminary passage through soda-lime.

Safety Test (4)

A 1.5-liter syringe was filled with the gas mixture and counted. A trap with a single valve was purged with gas mixture and the same syringe filled as rapidly as an FEV maneuver after passing thru the trap: 99.7% remained in the trap. The trap was kept at room temperature, 5 days; approximately 20% of activity was lost when compared to a sealed standard of ^{133}Xe . The trap was moved each day, and the exit valve had been removed to allow diffusion.

Regional Function Technique (3)

Lead panels may be interposed between patient and detector to shield the bases of the lungs. The panels in the current instrument (Fig. 1) are operated like a furnace damper. When the lever is raised, the lead panels shield the lower halves of the fields of view.

As soon as the 4-sec count interval is completed and while the subject is still holding his breath, the lever may be raised and lowered. Then the patient can exhale for the FEV₁ maneuver in the conventional manner. The recorder curves (Fig. 7), which had been indicating VC level, dip momentarily to lower levels indicating the fraction of

Mr. A. KAPLAN
2/26/70
72" 44 ♂
FEV₁ LEFT .53 RIGHT .56
VC 1.85 2.12

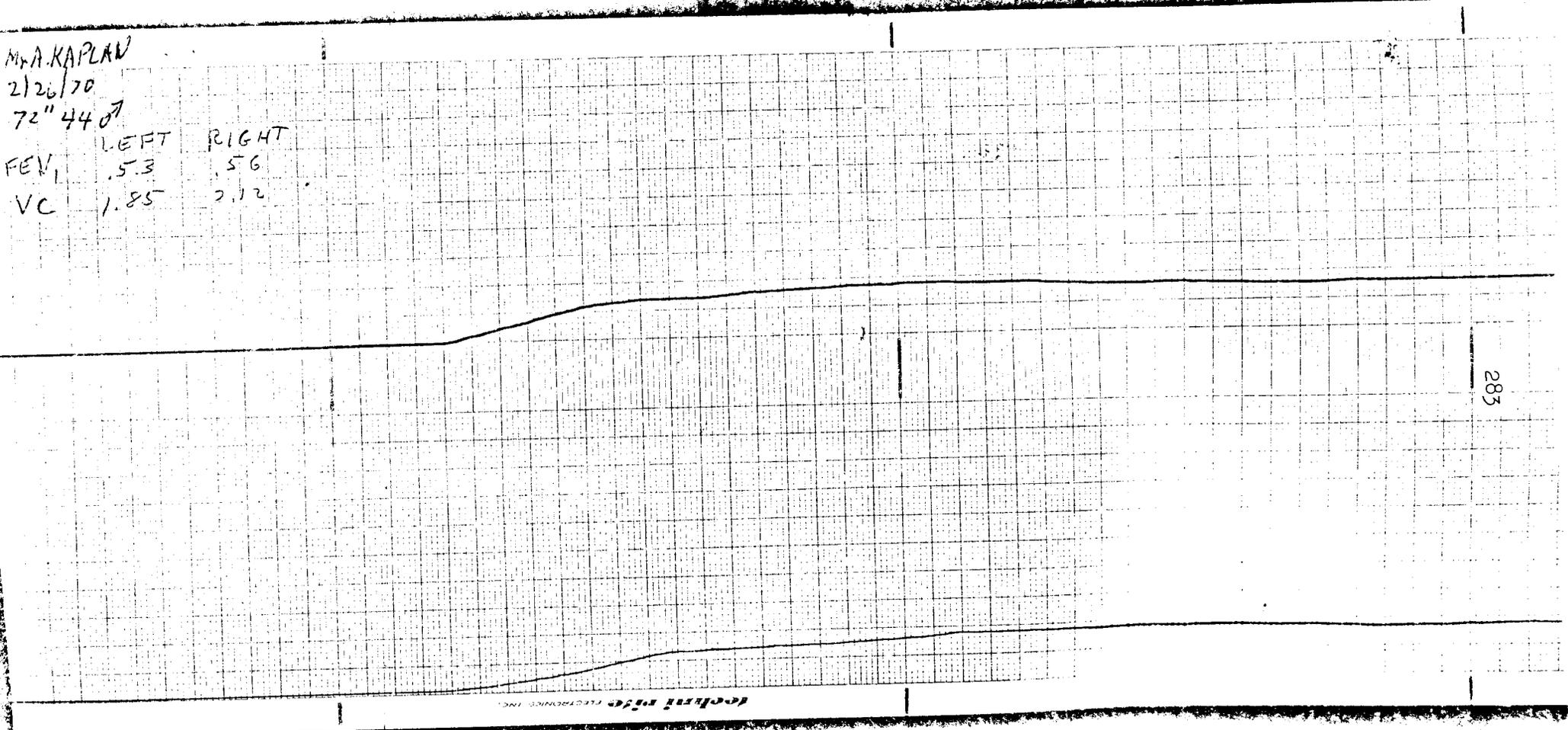


Figure 7 Recorder curves showing use of regional function technique (lead shield over base of lungs)

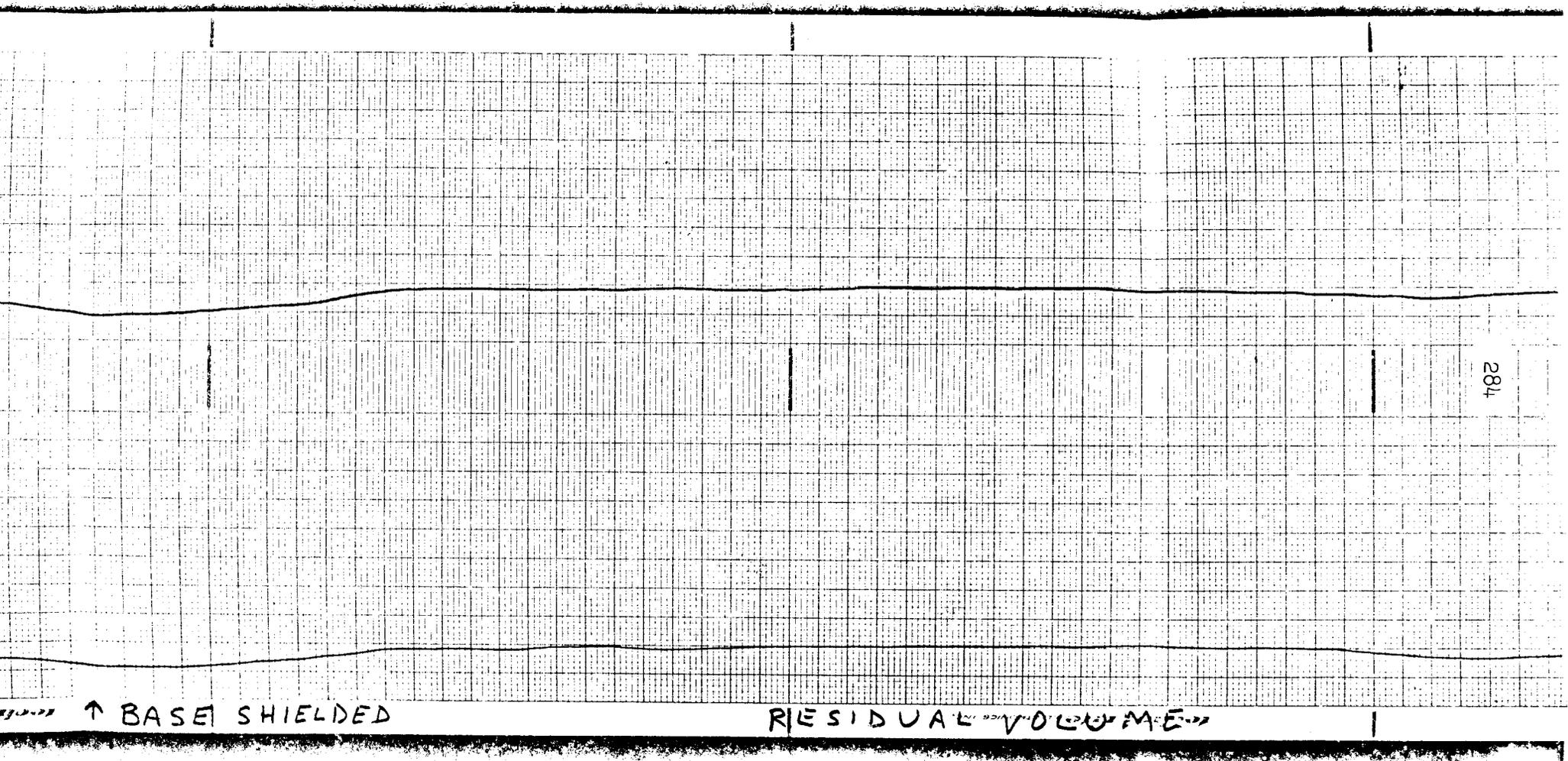


Fig. 7 (continued, pt. 1)

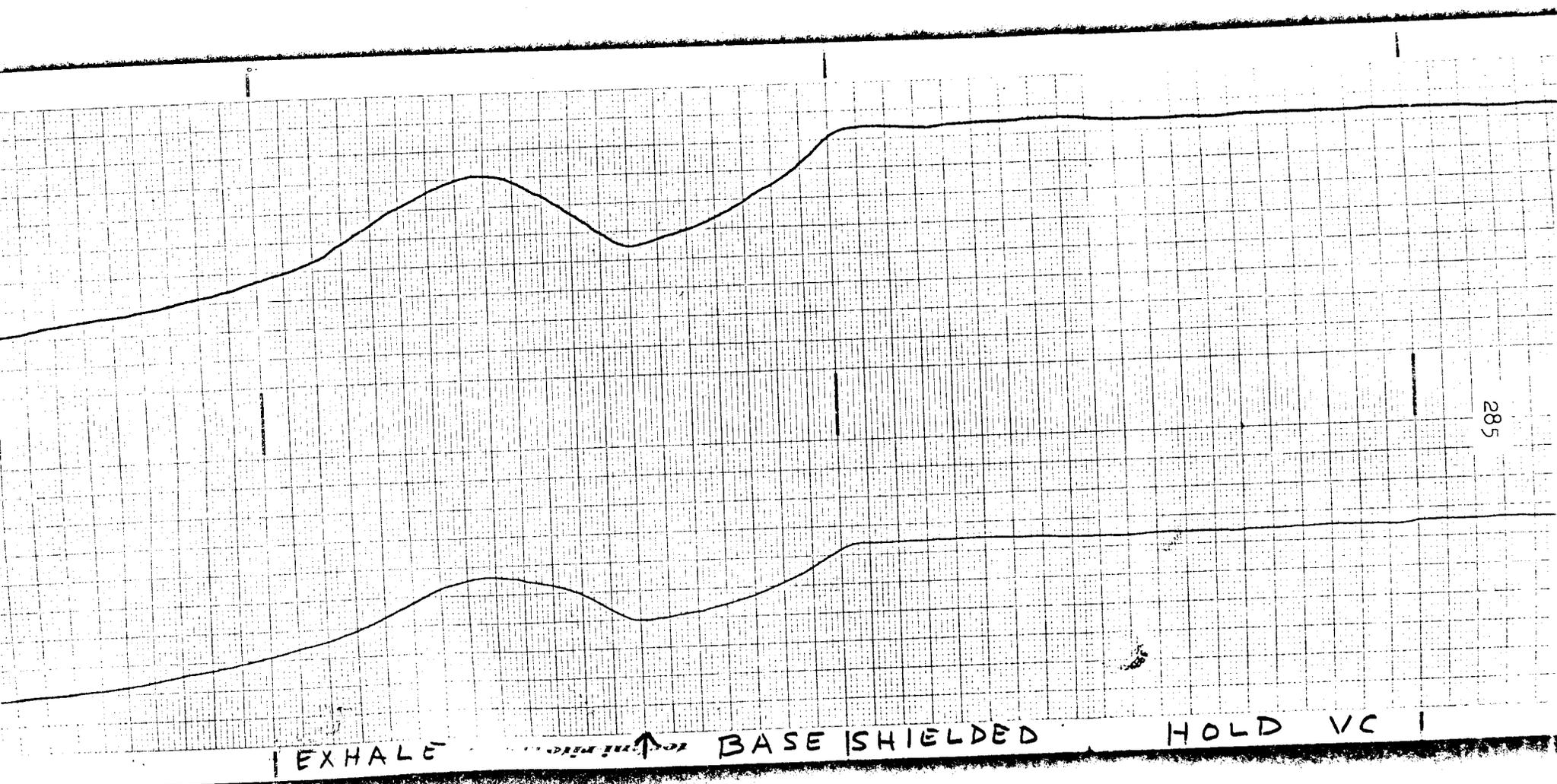


Fig. 7 continued (third page)

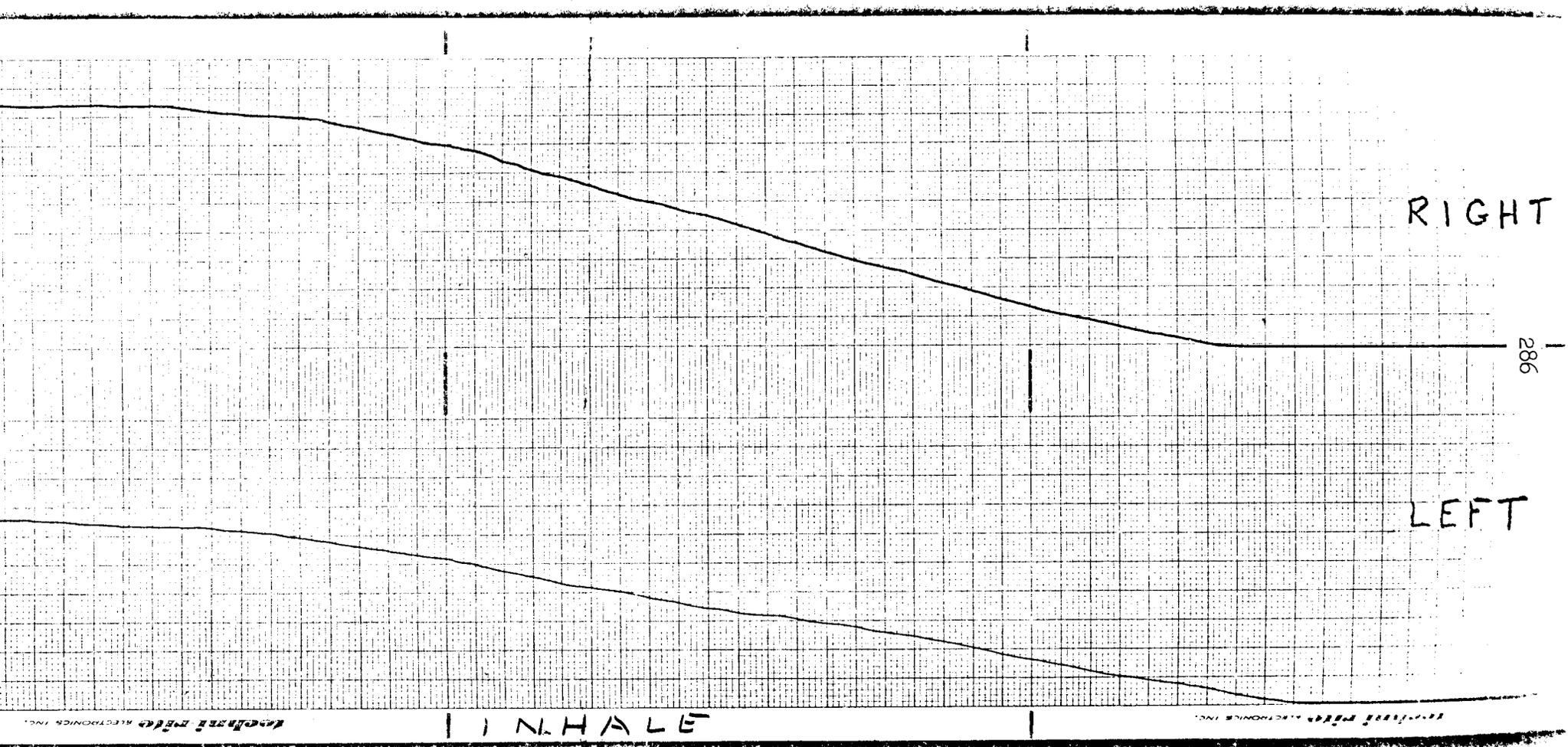


Fig. 7 continued (end)

gas in the upper halves. This ^{technique} can be repeated after exhalation, indicating the relative distribution of residual gas. These extra manipulations do not interfere with the quantitative data collection and offer a simple technique for localizing pathology to upper or lower lung.

Summary (3)

The ^{133}Xe pulmonary function analyzer is a self-contained system that provides digital output of lung volume data in liters. It incorporates several outstanding design features not available in other lung function analyzers, one of which is the digital output and another of which is the trapping system that makes the system so safe. Digital Products Corp., Fort Lauderdale, Fla., is now manufacturing and retailing the complete system. This company also made the digital data processing unit for the prototype system. (FEM)

Acknowledgements (3)

The author thanks Dr. W. S. Williams for the clinical material as well as his advice and encouragement and Mr. L. Kelman of Digital Products Corp. for his major contribution to the design of the data processing and control circuitry. This research was supported in part by the Friedman Fund.

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SURVEY OF THE USE OF RADIONUCLIDES IN MEDICINE*

Phase II

(2)

Summarized by Martha Gerrard

Supplementary Keywords: diagnosis; tracer; data compilation; phosphorus-32; chromium-51; cobalt-57; cobalt-60; strontium-85; strontium-87m; technetium-99m; iodine-125; iodine-131; mercury-197; mercury-203; gold-198

Phase I** of this study was a national survey of all licensed and registered medical users of radionuclides. In Phase II, methods of diagnosis using radioisotopes were surveyed with particular emphasis on measures that minimize patient exposure. The persons contacted in Phase II were selected from those who in the Phase I study reported having carried out one or more of some 23 radiopharmaceutical procedures in 1966. These 23 procedures (Table 1) accounted for more than 95% of the total number of patients reported in Phase I as receiving radiopharmaceuticals for diagnostic purposes. Unlike the Phase I study, therapy practices were not included in the Phase II questionnaire.

In 1969, questionnaires were mailed to 879 physicians, 85.2% of whom responded. Of these, 12.9% were no longer doing the particular procedure, and in 21.6% of the cases the radionuclide had been changed. A change of this magnitude in the 3 years between 1966 and 1969 indicates the fast pace of technical development in the practice of nuclear medicine.

At least 75% of the physicians who perform any of the 23 specified procedures do so in hospitals (Table 2). The only procedures used by a significant fraction of the physicians (more than 10%) not engaged in hospital practice (i.e., private or group practice) include thyroid uptake and scanning, the two vitamin B₁₂

*Summary of report BRH/DMRE 70-1, prepared by Stanford Research Inst., Menlo Park, Calif., for Radioactive Materials Branch, Division of Medical Radiation Exposure, U. S. Dept. of Health, Education, and Welfare, Public Health Service, Bureau of Radiological Health, Rockville, Md. 20852.

**Report MORP-68-10, summarized in Isotop. Radiat. Technol. 6: 415-23 (1969).

Please change isotopes to usual style, i.e., ^{198}Au , 57, etc.

Table 1. Radionuclides currently used in procedures sampled

Procedure Sampled	Number of Responding Physicians Using Radionuclide in 1969															Total	
	Au-198	Co-57	Co-60	Cr-51	I-125	I-131	I-125/131	Hg-197	Hg-203	P-32	Sr-85	Sr-87m	Tc-99m	In-113m	F-18		H-3
P-32 Blood Volume	0	0	0	4	0	8	0	0	0	2	0	0	0	0	0	0	14
Cr-51 Blood Volume	0	0	0	28	5	3	1	0	0	0	0	0	0	0	0	0	37
Co-57 Vitamin B-12	0	38	1	0	0	0	0	0	0	0	0	0	0	0	0	0	39
Co-60 Vitamin B-12	0	17	17	0	0	0	0	0	0	0	0	0	0	0	0	0	34
I-125/131 Blood Volume	0	0	0	0	8	27	0	0	0	0	0	0	0	0	0	0	35
I-125/131 Fat Absorption	0	0	0	0	0	37	3	0	0	0	0	0	0	0	0	1	41
I-125/131 Kidney Function	0	0	0	0	1	41	0	1	0	0	0	0	0	0	0	0	43
I-131 Thyroid Function	0	0	0	0	0	38	0	0	0	0	0	0	0	0	0	0	38
Tc-99m Brain Scan	0	0	0	0	0	0	0	0	0	0	0	0	39	0	0	0	39
Tc-99m Liver Scan	6	0	0	0	0	3	0	0	0	0	0	0	27	0	0	0	36
Sr-85 Bone Scan	0	0	0	0	0	0	0	0	0	0	43	0	0	0	1	0	44
Sr-87m Bone Scan	0	0	0	0	0	0	0	0	0	7	6	0	0	0	0	0	13
I-131 Thyroid Scan	0	0	0	0	1	36	0	0	0	0	0	0	1	0	0	0	38
I-131 Lung Scan	0	0	0	0	0	36	0	0	0	0	0	0	4	1	0	0	41
I-125/131 Placenta Scan	0	0	0	1	2	32	1	0	0	0	0	0	5	0	0	0	41
I-131 Liver Scan	11	0	0	0	0	26	0	0	0	0	0	0	4	0	0	0	41
I-131 Kidney Scan	0	0	0	0	1	15	0	6	8	0	0	0	0	0	0	0	30
Hg-197 Kidney Scan	0	0	0	0	1	1	0	27	9	0	0	0	1	0	0	0	39
Au-198 Liver Scan	30	0	0	0	0	2	0	0	0	0	0	0	5	0	0	0	37
Hg-203 Kidney Scan	0	0	0	0	0	0	0	15	16	0	0	0	0	0	0	0	31
Hg-203 Brain Scan	0	0	0	0	0	1	0	0	5	0	0	0	30	0	0	0	36
Hg-197 Brain Scan	0	0	0	0	0	0	0	8	1	0	0	0	30	0	0	0	39
Cr-51 Placenta Scan	0	0	0	9	0	0	0	0	0	0	0	0	3	0	0	0	12
Total	47	55	18	42	19	306	5	57	39	2	50	6	149	1	1	1	798

Table 2. Place of practice

Procedure	Percent of Physicians Doing Procedure		
	Hospital Practice	Individual Office Practice	Group Practice Outside Hospital
Cr-51 Blood Volume	94.7	--	5.3
Co-57 Vitamin B-12	87.8	9.1	3.2
Co-60 Vitamin B-12	89.4	--	10.6
I-125/131 Blood Volume	92.0	5.1	2.9
I-125/131 Fat Absorption	96.2	--	3.8
I-125/131 Kidney Function	93.6	5.9	0.4
I-131 Thyroid Function	81.2	15.4	3.4
Tc-99m Brain Scan	97.5	1.9	0.6
Tc-99m Liver Scan	100.0	--	--
Sr-85 Bone Scan	92.9	3.5	3.6
I-131 Thyroid Scan	76.4	16.3	7.3
I-131 Lung Scan	100.0	--	--
I-125/131 Placenta Scan	100.0	--	--
I-131 Liver Scan	87.9	2.7	9.4
I-131 Kidney Scan	96.6	--	3.5
Hg-197 Kidney Scan	98.9	--	1.1
Au-198 Liver Scan	86.4	7.5	6.1
Hg-203 Kidney Scan	83.2	13.1	3.7
Cr-51 Placenta Scan	100.0	--	--

malabsorption procedures, the two most widely used liver scanning procedures, and the most widely used kidney scanning procedure.

The only radionuclide widely procured by the physicians in the form of a generator is ^{99m}Tc . More than half the physicians procure unit doses of radiopharmaceuticals for the vitamin B_{12} malabsorption, the $^{125}/^{131}\text{I}$ fat malabsorption, the ^{131}I -iodide thyroid uptake and scanning, and the $^{125}/^{131}\text{I}$ human serum albumin blood volume and placental localization procedures. The majority of the physicians performing the remaining procedures under consideration procure the radiopharmaceutical in the form of multidose lots.

In all but three procedures, at least half the practitioners rely on the suppliers for quality control of the radiopharmaceutical. These three procedures use radionuclides that most of the practitioners procure as generator-produced material. Those practitioners who test the as-received material usually conduct an activity assay. In general, more of the physicians who procure a radionuclide in multidose lots conduct quality control tests than do those who procure it in unit doses.

In all the scanning procedures except liver scanning with sulfur colloid tagged with ^{99m}Tc , more than 75% of the physicians reported no radiopharmaceutical modification. Tagging was also indicated as the modification made by about a third of the physicians performing placental localization with ^{51}Cr -labeled red blood cells. Most of the other physicians conducting modifications perform only dilutions. The majority of the physicians who use modified radiopharmaceuticals conduct some testing before administration, and almost all conduct at least an activity assay.

Although the range of values of average administered activity was wide for most procedures, the average value (which is weighted to represent all physicians performing a procedure) compared favorably with the values reported in well-known references (Table 3).

More than two thirds of the practitioners of three of the procedures using

Table 3 . Administered radiopharmaceutical dosages

Procedure	Administered Activity per Patient, μCi		
	Average of All Practitioners	Range of Values Reported by Sample of Practitioners	
		Minimum	Maximum
CR-51 Blood Volume	27.33	3.00	50.00
Co-57 Vitamin B-12	0.50	0.10	1.40
I-125/131 Blood Volume	5.50	0.50	22.50
I-125/131 Fat Absorption	44.44	15.00	100.00
I-125/131 Kidney Function	45.01	3.00	150.00
I-131 Thyroid Function	36.86	2.50	100.00
Tc-99m Brain Scan	7,936.59	600.00	15,000.00
Tc-99m Liver Scan	2,971.61	700.00	8,000.00
Sr-85 Bone Scan	104.97	50.00	300.00
I-131 Thyroid Scan	56.86	10.00	150.00
I-131 Lung Scan	260.44	100.00	350.00
I-125/131 Placenta Scan	7.07	1.50	15.00
I-131 Liver Scan	157.71	55.00	400.00
Hg-197 Kidney Scan	144.42	45.00	300.00
Am-198 Liver Scan	174.97	60.00	500.00
Hg-203 Kidney Scan	116.58	70.00	280.00
Cr-51 Placenta Scan	23.94	5.00	50.00

radioactive-iodine-tagged materials reported that they administered iodine-blocking agents to prevent accumulation of radioactive iodine in the thyroid (Table 4).

The practice of administering agents to accelerate the elimination of radionuclides from the patient is not widespread. It was reported (Table 5) as being used by more than 25% of the practitioners in the cases of only two procedures, and these are procedures that use accelerated elimination to improve diagnostic effectiveness.

Almost all the practitioners calibrate their radiation detection and measuring instruments with some frequency---generally speaking, with each use or at least daily. The fraction of those who calibrate at increasing intervals decreases with the length of the interval.

The forms in which the radionuclides were reported as being used are:

Procedure	Radionuclide	Form	Procedure	Radionuclide	Form
Blood volume	P-32	Sodium phosphate	Liver scanning	Tc-99m	Colloid
	Cr-51	Sodium chromate		I-131	Rose bengal
	I-125/131	Albumin		Au-198	Colloid
Vitamin B-12 malabsorption	Co-57	Cyanocobalamine	Bone scanning	Sr-85	Nitrate or chloride
	Co-60	Cyanocobalamine		Sr-87m	Nitrate or chloride
Fat malabsorption	I-125/131	Labeled fats and fatty acids	Lung scanning	I-131	Albumin macroaggregates
			Placenta scanning	I-125/131	Albumin
Kidney function	I-125/131	Sodium iodohippurate	Kidney scanning	Cr-51	Sodium chromate
Thyroid function	I-131	Sodium iodide	Kidney scanning	I-131	Renal labeled compounds
Thyroid scanning	I-131	Sodium iodide		Hg-197	Labeled mercurials
Brain scanning	Tc-99m	Pertechnetate		Hg-203	Labeled mercurials
	Hg-203	Labeled mercurials			
	Hg-197	Labeled mercurials			

The report itself gives, in tabular form, detailed breakdown of the data arranged according to numerous schemes.

Table 4. Blocking agent practices

Procedure	Percent of Physicians Doing Procedure						
	Blocking		Blocking Agents Used				
	No	Yes	Iodides	Per chlorates	Mercurial Diuretics	Vitamin B ₁₂ Other	
Cr-51 Blood Volume	84.0	16.0	16.0	--	--	--	--
Co-57 Vitamin B-12	89.0	11.0	--	--	--	11.0	--
Co-60 Vitamin B-12	82.7	17.3	← Insufficient Data →				
I-125/131 Blood Volume	88.5	11.5	11.4	--	--	--	0.1
I-125/131 Fat Absorption	32.2	67.8	67.8	--	--	--	--
I-125/131 Kidney Function	95.9	4.1	4.1	--	--	--	--
I-131 Thyroid Function	98.3	1.7	--	--	--	--	1.7
Tc-99m Brain Scan	35.1	64.9	18.9	37.4	2.7	--	5.9
Tc-99m Liver Scan	95.8	4.2	1.4	2.8	--	--	--
Sr-85 Bone Scan	98.9	1.1	--	--	--	--	1.1
I-131 Thyroid Scan	100	--	--	--	--	--	--
I-131 Lung Scan	16.9	83.1	82.0	0.3	--	--	0.8
I-125/131 Placenta Scan	9.8	90.2	90.2	--	--	--	--
I-131 Liver Scan	72.6	27.4	27.4	--	--	--	--
I-131 Kidney Scan	55.9	44.1	← Insufficient Data →				
Hg-197 Kidney Scan	87.6	12.4	--	--	12.4	--	--
Au-198 Liver Scan	90.3	9.7	4.9	4.8	--	--	--
Hg-203 Kidney Scan	86.9	13.1	--	--	13.1	--	--
Cr-51 Placenta Scan	92.2	7.8	7.8	--	--	--	--

Table 5. Accelerated elimination practices

Procedure	Percent of Physicians Doing Procedure	
	Accelerated Elimination	
	No	Yes
Cr-51 Blood Volume	84	16
Co-57 Vitamin B-12	74	26
Co-60 Vitamin B-12	86	14
I-125/131 Blood Volume	92	8
I-125/131 Fat Absorption	99	1
I-125/131 Kidney Function	95	5
I-131 Thyroid Function	100	--
Tc-99m Brain Scan	98	2
Tc-99m Liver Scan	100	--
Sr-85 Bone Scan	72	28
I-131 Thyroid Scan	88	12
I-131 Lung Scan	99	1
I-125/131 Placenta Scan	97	3
I-131 Liver Scan	100	--
I-131 Kidney Scan	100	--
Hg-197 Kidney Scan	96	4
Au-198 Liver Scan	100	--
Hg-203 Kidney Scan	92	8
Cr-51 Placenta Scan	100	--

AVAILABILITY OF ISOTOPES AND SERVICES (2)

Additions to ORNL Stable-Isotope Inventory (3)

Recent additions to the Oak Ridge National Laboratory (ORNL) enriched—
stable-isotope inventory include the following:

Isotope	Purity, %	Amount Available, g
^{33}S	86.8	0.09
^{58}Ni	99.8	30.5
^{61}Ni	92.9	2.4
^{61}Ni	90.4	3.3
^{62}Ni	96.1	4.7
^{112}Sn	79.6	3.8
^{136}Ce	21.7	1.4
^{154}Gd	66.5	6.7
^{166}Er	96.2	27.7
^{198}Hg	85.3	0.22
^{198}Hg	71.5	0.72
^{18}O	99.0	2.3
^{18}O	97.3	14.3

Isotopes Sales Agents

(3)

New England Nuclear Corp. has announced that it has been appointed the commercial sales agent for Union Carbide's short-lived radioisotopes. The materials involved are ^{24}Na , ^{42}K , ^{82}Br , ^{64}Cu , ^{99}Mo , ^{131}Ba , ^{131}Cs , ^{198}Au , $^{114\text{m}}\text{In}$, ^{197}Hg , and ^{169}Yb .

Mallinckrodt Chemical has closed its Sterling Forest and Flushing (New York) facilities and opened a radiopharmaceutical production and distribution center in Carlstadt, N.J. The center produces technetium products, including $^{99\text{m}}\text{Tc}$ generators, and supplies these and other short-lived isotopes.

New Services in Radioisotope Field (3)

E. A. Burrill & Associates (701 Welch Road, Suite 324, Palo Alto, Calif. 94304) is a new consulting organization specializing in adapting the marketing, technology, and application of ionizing energy and nuclear radiation to a broad scope of problem areas in research and industry. Among the fields of particular interest are processing with ionizing radiation, nondestructive testing, chemical analysis with nuclear radiation, environmental-quality monitoring and control, ^{and} ion implantation.

Suntac Nuclear Corporation (1528 Walnut St., Philadelphia, Pa. 19102) is a joint venture of the NUS Corporation and Catalytic, Inc. which provides services to various nuclear installations, including irradiation facilities and radioactive facilities of various types. Both parent companies have a long background of work in this field.

Brochure on Radiopharmaceutical Specifications

Detailed specifications for 116 labeled compounds for radiopharmaceutical use are given in a brochure, Spécification Radiopharmaceutique, issued by Département des Radioéléments, C.E.N./Saclay, B.P. No. 2, 91, Gif-sur Yvette, France. It lists artificial radioisotope-labeled materials for medical usage, both diagnostic and therapeutic, as well as radioactive preparations used in in vitro tests. Physical, chemical, and biological properties are included, together with methods used in quality control and testing. Thirty radioisotopes are involved. The brochure is supplied free upon request.

Shielding Container for Small Volumes
of Radioactive Solutions (3)

Shielding containers for radioisotope solutions, made of lead and having high-density lead-glass windows are being marketed by Nuclear Associates, Westbury, N.Y. The Gamma Vue Shields completely protect a person handling the radioisotope, for example, a physician inserting a syringe into a vial inside the shield, while allowing full view of the contents.

AEC ACTIVITIES (2)

New Contracts Awarded (3)

Among the contracts recently awarded by the USAEC's Division of Isotopes Development are to Washington State University, Pullman, for study of radioisotopic techniques/in stream pollution resulting from paper mill additives and to investigate polymer grafting to inorganic substrates with radiation techniques; to the Commerce Dept., Boulder, Colo., for a cloud buoyance meter; to the Naval Oceanographic Office, Washington, evaluating for various nuclear oceanographic techniques; to Hughes Research Labs., Malibu, Calif., for research leading to production of a cadmium telluride gamma-ray detector and spectrometer; to Texas Nuclear, Austin, to develop a method for field analysis of wet-concrete quality; to Panametrics for application of instruments based on ^{85}Kr to determination of air pollution from automobile exhaust; and to Zoller Dental Clinic of the University of Chicago for determining requirements for a radioisotopic radiation-backscatter gage for tooth-enamel-density measurements.

Funds for Radiation Processing of Food (3)

The Joint Committee on Atomic Energy has recommended increases in the USAEC's FY 1971 requests for food-pasteurization-research funds. The recommendation is that the \$150,000 and \$130,000 requested for the Division of Isotopes Development and the Division of Biology and Medicine programs, respectively, be increased to \$270,000 each, subject to availability of funds. In addition, the House Committee on Armed Services has recommended \$1,825,000 for the Army's food sterilization program. The Army had not sought FY 1971 funds for the program, objecting to the estimated \$2.7 million cost of obtaining food approvals from the Food and Drug Administration. The Committee feels that the recommended funding level would ensure U.S. leadership in the field and preserve the highly qualified scientific staff executing the program.

Radioisotope Production Technology Seminar (3)

Approximately 120 representatives of government and industry met in Oak Ridge, Tenn., on June 2-3 to discuss radioisotope production technology developments. Formal papers, besides those on general subjects, covered accelerator production of radionuclides, neutron products, fission product recovery, biomedical isotopes, ^{252}Cf , beta sources, and shipping casks.

Nuclide Price Changes (3)

According to the Federal Register 35(103): 8300 (May 27, 1970), the USAEC proposes to reduce the price for ^{241}Am to \$150 a gram based on its full cost of production and distribution. Also proposed is a change in the base charge for 80% ^{238}Pu and the establishment of a base charge for 90% ^{238}Pu containing not more than 0.3 part of ^{236}Pu per million.

GENERAL (2)

Progress in Radioisotope-powered Pacemakers (3)

The successful implantation of a radioisotope-powered pacemaker in a human patient in May has been reported from France. The UKAEA reports successful implantations in two dogs, one in February and the other in March, both dogs still doing well. The French and the British pacemakers are both fueled with ^{238}Pu , and the nuclear batteries have a design life of more than 10 years.

 ^{128}Xe Produced from ^{127}I (3)

An economical process for making ^{128}Xe from ^{127}I has been developed by Douglas United Nuclear. The ^{127}I is irradiated to form ^{128}I , which decays to ^{128}Xe . About 200 ml of ^{128}Xe has been produced for Argonne National Laboratory, who is using it in fuel-element testing.

Five More ^{90}Sr Generators Ordered (3)

Teledyne Isotopes has an order from the Navy for five 25-w ^{90}Sr generators, to be used in remote land-based or marine weather stations. These bring to 21 the number of such sources in use.

Radioisotope Packaging Guide (3)

In a report by R. D. Seagren, "ORNL Isotopes Guide for the Packaging of Radioactive Materials for Transport" (ORNL-TM-2769, March 1970), packaging regulations are condensed for convenient use. The guide provides the necessary information for packaging, labeling, and marking routine shipments from ORNL, and supplements will be issued as regulations change.

Meeting on Neutron Sources (3)

The American Nuclear Society will hold a National Topical Meeting on neutron sources and applications in Augusta, Ga., on April 18-21, 1971. The meeting will be cosponsored by the local section and the Isotopes and Radiation Division of the ANS together with the Savannah River Operations Office

of the U.S Atomic Energy Commission. The program will deal with the production of neutron sources, their unique qualities, and their uses in industry, medical and forensic sciences, and exploratory and conservation areas.

Neutron Products Delivers One-Millionth Curie (3)

A milestone in radioisotope production and sales was reached May 12, 1970 when Neutron Products, Inc. delivered a quantity of ^{60}Co from its Dickerson, Maryland facility to ARCO Chemical Company, A Division of Atlantic Richfield Company. Included in the delivery was the one-millionth curie of ^{60}Co produced in a power reactor for public sale.

Neutron Products, Inc. reached agreement in 1966 with Consumers Power Company, a large utility company serving Michigan, for production of ^{60}Co in available space in the core of the Big Rock Point power reactor, Charlevoix, Michigan. Under J. A. Ransohoff, Neutron Products had decided that there was sufficient market for ^{60}Co to justify production by a private company; until Neutron Products entered the field, all ^{commercial} production---worldwide---of radiation-processing-grade ^{60}Co was by sovereign governments. One other company was already producing teletherapy-grade ^{60}Co .

At ceremonies marking the occasion of the one-millionth-curie sale, J. H. Campbell, president of Consumers Power Company, stressed the aggressiveness and practicality of ^{the} Neutron Products, Inc. venture. Consumers Power was not particularly interested in ^{60}Co production and had a ^{number of} / elaborate programs with the USAEC and General Electric Company which resulted in some rather difficult production conditions. To help alleviate these, Neutron Products built its own processing facilities to handle the ^{60}Co produced. Campbell cited the joint program as an example of how a small group of relatively uninfluential individuals with a good idea can be accommodated within large American industry. He added that Consumers Power is satisfied with both technology and economics and plans to continue producing ^{60}Co in its power reactors as a routine matter.

Also included in the ceremonies was a tour of the new materials irradiator of Neutron Products. This operating prototype, placed into operation early in 1969, is able to accommodate many different sizes and shapes of products to be irradiated. M. Turkanis, marketing director, pointed out that the system has provided trouble-free service under the high demand placed on it since early 1970. Neutron Products may soon offer facilities based on this design at a fixed price with guaranteed performance.

The one-millinth curie^{that} was purchased by ARCO Chemical Company will be used to process a wood-plastic composite marketed by ARCO as Permagrain. Other uses of ^{60}Co include sterilization of medical supplies, synthesis of certain chemicals, modification of polymers, treatment of cancer victims, and pasteurization and disinfestation of food. Ransohoff emphasized the size of the ^{60}Co market by pointing to the orders Neutron Products has on hand for delivery of 4.5 MCi of ^{60}Co during the next three years.

(FEM)

CALIFORNIUM-252 TO EXPLORE OCEAN BOTTOM FOR MINERALS (3)

Under a contract with the National Science Foundation, researchers of the University of Georgia will use a 197- μg ^{252}Cf neutron source to evaluate the possibility of using activation analysis for underwater mineral exploration. Dr. John Noakes will lead the group, which will use the Westinghouse Deepstar 4000 submersible in tests off the California coast in June 1970. Laboratory tests have established limits of detection for minerals in the presence of seawater and under various thicknesses of sediment. In the undersea tests, scuba divers will place and mark specific samples, which the Deepstar system will then analyze by the neutron activation method. Data will appear onboard as spectra and will also be stored on magnetic tape for later detailed study. In addition to the ^{252}Cf neutron source, a compact neutron accelerator will be used in the tests.

Nuclear Electronics (3)

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Springer-Verlag, New York, 1970 (402 pp., 337 figs., 918 refs.; hardback, \$26.40).

Supplementary keywords: book review; textbook; review, technical; bibliography; theory, applied; instrument; design; measurement and control system; industry; industry, electronics; biology; physics; radiochemistry; counting; luminescence; Moessbauer effect; ionization of gas; data handling; semiconductor; crystal.

A close relation exists between electronics and radiation measurements in nuclear physics. This relation is evident from the large number of electronic measuring instruments used in pure or applied nuclear research, radiation protection, and the application of radioisotopes. Nuclear electronics has enjoyed such rapid growth that heretofore no summary of the subject has been available. To provide such a summary, Prof. Kowalski supplemented the script of his lectures with a literature review and thus ^{contributed} / Nuclear Electronics, which he says could be called a monograph with the character of a textbook. It is the only complete up-to-date monograph that deals with the specialized electronic circuits and instruments used in nuclear radiation metrology. It covers the literature through 1968. Its systematic structure reflects the author's effort to ensure the continuance of its value as a reference work for years to come. The book is intended primarily for the experimental physicist but is also useful in the training of electronic engineers and as a reference for technicians. For conciseness, the general concepts of electronics are taken for granted, and certain basic knowledge is assumed.

The object of Nuclear Electronics is to emphasize the part of signal processing that is characteristic of nuclear physics. Only pulse techniques deal adequately with the quantum character of nuclear processes. In awareness of their importance, the author has devoted his descriptions only to circuits for pulse signals. A detector of the nuclear process gives rise to a current or

voltage pulse. The pulse is correlated with the time instant of the nuclear process and carries most of its information in a continuous manner. The representation of data by a continuously variable parameter is an analog representation.

Because it is very difficult to process analog signals, they are converted as quickly as possible to digital signals, which are then processed simply.

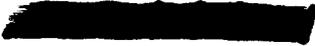
Electronic apparatus for nuclear research usually consists of a detector, analog part, analog-to-digital converter (ADC), and digital part for signal processing. Professor Kowalski represents the array as



The arrangement of his book follows this array. He describes various radiation detectors, preamplifiers, and auxiliary circuits (Ch. 2). The analog operation of the detector signals is discussed, especially the complex problem of pulse forming and amplification (Ch. 3). The various types of ADC's for conversion of pulse amplitude, pulse shape, or time interval between two pulses are described (Chs. 4 and 5). A discussion is also included of the digital circuits found in data-processing assemblies and computers, industrial measurement and control systems, and nuclear measuring equipment (Ch. 6). Scaler assemblies and analyzers for nuclear research are reviewed (Ch. 7). A discussion of Laplace transform calculus and of noise is appended (Ch. 8).

The book should be extremely useful to those working in the field of radiation biophysics, nuclear physics, electronics, photochemistry, or radiochemistry.

(HPR)


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