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THE PREPARATION OF THIN, UNIFORM SOURCES FOR A BETA-RAY SPECTROMETER

R. L. Blanchard
Bernd Kahn
R. D. Birkhoff

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THE PREPARATION OF THIN, UNIFORM SOURCES FOR A BETA-RAY SPECTROMETER

Richard L. Blanchard
Bernd Kahn
R. D. Birkhoff

Date Issued

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I. Introduction

In the middle and late 1930's it was believed that the Fermi theory of beta decay would have to be modified because experimentally determined Beta spectra showed a larger number of low energy electrons than could be accounted for by the theory. However, in 1939, Tyler[1] showed by using varying thicknesses of source and backing materials that the thinner these materials, the nearer the Fermi plot approached a straight line. In 1951, Tiamuty[2] demonstrated the same effect in the spectrum of promethium-147. Also, Flammersfeld,[3] using a RaE source, showed it was absolutely essential to use very thin sources and source backing materials to obtain accurate spectra at low energies.

Degrading of the energies of beta particles produced by elastic and inelastic scattering in the source and source mounting has a noticeable effect upon the conversion line spectra, as well as producing a widening in the conversion line on the low energy side with a resulting loss of energy resolution. Frequently, two or three lines will be superimposed as one poorly resolved line. Therefore, it was the purpose of this work to devise a simple, fast, and relatively

---

1 Tyler, A. W., Phys. Rev. 56, 125 (1939).

2 Tiamuty, S. I., Phys. Rev. 81, 461 (1951).

3 Flammersfeld, A., Zeits. f. Physik 112, 727 (1939).

inexpensive method of preparing sources for the beta spectrometer.

A study was made of the radiocolloidal properties of radionuclides as applied to the production of depositions by adsorption and precipitation, and of electrodeposition of source material upon very thin, inert metallic films. The isotopes that were prepared as sources from carrier-free or very nearly carrier-free solutions were obtained from the Isotopes Division of the Oak Ridge National Laboratory and are listed in Table I.[4] The sources prepared by each method were autoradiographed and their uniformity determined by a densitometer. Also, two sources prepared by electrodeposition were analyzed in a beta spectrometer as an additional measure of the source uniformity and thickness.

Sources prepared by adsorption and electrodeposition proved to be quite uniform, having a variation in uniformity, as measured by autoradiography, of only a few percent. In the beta spectrometer the momentum resolution of the conversion lines of Hg-199 varied from 0.25% at 144 kev to 0.28% at 35 kev, indicating again that the source and source mounting were very thin and uniform. The source thickness was calculated to be about 6 $\mu g/cm^2$ thick, giving a total thickness of source mount, vaporized gold conducting film and source of 60 to 70 $\mu g/cm^2$.

---

[4] Catalogue and Price Lists of Radioisotopes, Special Material and Services, Oak Ridge National Laboratory, Current Issue.

Table I. Isotopes Used

Isotope	Half Life	Chemical Form	Specific Activity Millicuries/gram
Cerium-144	282 d.	$CeCl_3$	CF*
Chromium-51	27.8 d.	$CrCl_3$	25,000
Cobalt-60	5.27 y.	$CoCl_2$	10,000
Gold-198	2.69 d.	$AuCl_3$	15,000
Gold-199	3.15 d.	$AuCl_3$	CF
Indium-114	49.0 d.	InCl	50
Iridium-192	74.4 d.	Na_2IrCl_6	1,000
Iron-59	45.1 d.	$FeCl_3$	500
Mercury-203	45.0 d.	$Hg(NO_3)_2$	50
Phosphorous-32	14.3 d.	H_3PO_4	CF
Rubidium-86	19.5 d.	RbCl	100
Ruthenium-106	1.0 y.	$RuCl_3$	CF
Scandium-46	85.0 d.	$ScCl_3$	5,000
Selenium-75	127 d.	H_2SeO_3	5,000
Silver-110	270 d.	$AgNo_3$	100
Strontium-89	53.0 d.	$SrCl_2$	CF
Thallium-204	4.0 y.	$TlNO_3$	50
Yttrium-91	61.0 d.	YCl_3	CF

* CF - Carrier-Free

II. A Survey of Source Preparation Methods

A. Source Mountings

There are a large number of materials which can be employed as source mountings.[5] A few of the most common ones will be described briefly.

(1) Zapon Films. Zapon lacquer is diluted in anyl acetate and a drop or two of the solution is placed on a surface of distilled water. The drops are spread by surface tension, forming a thin film over the surface of the water. After the film has dried, it may be removed by a circular frame of wire or by the source ring itself. Films made in this way may range in thickness from several $\mu g/cm^2$ to several mg/cm^2. Zapon films are not as strong as films of nylon or formvar.

(2) Nylon Films. Nylon, in the form of chips or sheets, is dissolved in isobutyl alcohol by careful boiling. The concentrated solution is then decanted from the undissolved nylon and films prepared as described above for zapon films. For best results, the solution should be used within about an hour. In this way, films of about 10 $\mu g/cm^2$ can be made. Thicker films are obtained by using several films together, thus eliminating the effect of microscopic pinholes which may be present in the individual layers.

---

[5] Siegbahn, K., BETA- AND GAMMA-RAY SPECTROSCOPY, Interscience Publishers, Inc., New York, 1955, pp. 261-263.

(3) Formvar Films. Formvar films were used as the source mountings in this work and will be described in Chapter IV.

(4) Aluminum Foils. Aluminum foils of 0.2 mg/cm^2 can be purchased commercially and have the added advantage that no precautions are necessary in order to prevent charging of the sample. These foils are also quite resistant in nitric acid solutions and in very dilute hydrochloric acid and sulfuric acid solutions.

(5) Rubber Hydrochloride Films. Two-way stretched rubber hydrochloride films of 0.5 mg/cm^2 and 0.75 mg/cm^2 thickness are commercially obtained. They may be mounted on source rings as described above for zapon films.

B. Source Preparations

Numerous methods have been employed to prepare beta sources, and a careful consideration of the advantages and disadvantages of the methods of deposition available must be made for optimum results. The rate of deposition, the percent of the activity deposited, the equipment at hand, the cost of the operation, the uniformity and thickness of the source, and the amount of activity needed must all be taken into account. Also, there are limitations attached to each radionuclide because of its characteristic chemical and physical properties, which must be considered before a successful source can be made.

A few of the most common methods of source preparation will be discussed briefly.

(1) The evaporation to dryness of radioactive solutions is used

because it is easy, rapid, and is not limited to any specific group of nuclides. In general, the sources prepared in this manner are non-uniform and relatively thick.

(2) The spreading of the source as a powder over the source backing is another common method. To keep the activity from falling off, it is either spread on cellophane tape[6] or spread on film and covered with another piece of film.[7,8] This method also produces thick and nonuniform sources. Also, the beta particles will lose energy in passing through the film covering the source, resulting in added distortions to the spectrum.

(3) Sources can be prepared by direct irradiation. For example, metallic foils are placed in a reactor and left until sufficient activity has been produced. Because of heat and radiation damage, the use of organic films as backing materials is restricted. The difficulty in using an irradiated metal foil directly as the source is that impurities in the metal will also become active and may lead to erroneous interpretations of the spectrum.

(4) Sources have been deposited electromagnetically.[9] The

---

[6] Emmerich, W. S. and Kurbatov, J. D., Phys. Rev. 83, 40 (1951).

[7] Kondaiah, E., Phys. Rev. 79, 891 (1950).

[8] Scoville, C. L., Fultz, S. C., and Pool, M. L., Phys. Rev. 85, 1046 (1952).

[9] Bergstrom, I., Phys. Rev. 82, 112 (1951).

radionuclides are passed into a magnetic field, generally from a heated filament, and are bent into an arc with a radius depending upon the mass of the nuclide. After passing through an arc, usually of 180°, the nuclides are deposited according to their mass. By this method, the radionuclide is purified at the same time the source is being produced. The source backing, which is used as the target in the mass spectrograph, must be a metal generally having considerable thickness in order to withstand the bombardment. Sources prepared by this method can be very thin.

(5) Another method which produces very thin sources is that of vacuum thermal volatilization, which has been discussed in great detail by Holland.[10] With this method, sources may be prepared which are very thin and uniform. However, only a small fraction of the initial activity placed on the filament reaches the source backing, the remainder depositing within the evaporating apparatus. To attain a source of sufficient activity, the initial activity on the filament must be quite large. Pate and Yaffe[11] have improved these conditions by distilling the radionuclides in relatively volatile forms from a crucible onto the film backing, which is placed over the top of the crucible. At present, they have been most successful with compounds containing large organic groupings, such as aromatic compounds and especially

---

[10] Holland, L., VACUUM DEPOSITION OF THIN FILMS, John Wiley and Sons, Inc., New York (1956).

[11] Pate, B. D. and Yaffe, L., Can. J. Chem. 34, 265 (1956).

chelate compounds.

(6) Electrodeposition has also been applied as a method of source preparation.[12] A metal foil, usually having a thickness in the order of mgs/cm^2, is used as the cathode and source backing. These sources are usually quite uniform but have considerable backing thickness.

Table II lists the methods described above, along with the nuclides to which they were applied. Solvent evaporation, the spreading of the nuclides on the source backing and pile irradiation have been omitted because they are so numerous.

There have also been methods of preparation devised which are suited to special radionuclides. For instance, ThC'D has been deposited on a thin aluminum leaf by alpha recoil in a vacuum from a source of thorium (B+C+C').[13] Mathur has produced a source of Xe-127 on a 0.1 mil. aluminum foil by the glow discharge method.[14] A source of Cs-138 has been prepared by placing a wire in a closed cylindrical vessel of Xe-138, which decays to Cs-138. One-third of the cesium

---

[12] Broyles, C. D., Thomas, D. C., and Haynes, S. K., Phys. Rev. 89, 715 (1953).

[13] Martin, D. G. E., Richardson, H. O. W., and Yun-Kueihsu, Proc. Phys. Soc. 60, 446 (1948).

[14] Mathur, H. B., Phys. Rev. 97, 707 (1955).

TABLE II
METHODS OF SOURCE PREPARATION

H A-41																	He
Li	Be											B	C A-54	N	O	F	Ne
Na	Mg											Al	Si	P E-23 T-62 A-70	S T-51	Cl	A
K T-44,58	Ca	Sc T-5	Ti Em-68	V	Cr	Mn	Fe E-26,43	Co E-2, 11 43,64 T-51 A-63	Ni E-8 T-51	Cu A-14,27 T-38,50 E-66	Zn E-25,43 T-52	Ga	Ge	As T-67 Em-68	Se	Br Em-68	Kr Em-3, 68
Rb T-30,42 Em-68	Sr E-4 Em-31 T-62	Y Em-31 T-62	Zr	Cb	Mo Em-68	Tc	Ru E-24,40 Em-31	Rh E-13	Pd	Ag E-13,57 A-59 T-72	Cd E-13 T-20	In E-35	Sn E-9,13,56	Sb E-13,56 T-39	Te E-13	I Em-68 A-70	Xe Em-3,68 T-45
Cs T-9 Em-46,68	Ba Em-31 A-70	La Em-31	Hf T-10 Em-68	Ta	W	Re E-19,53 T-53	Os	Ir T-49	Pt	Au T-9 E-32	Hg T-20 Em-68	Tl Em-46 E-71	Pb A-18 E-29 T-61 Em-68	Bi E-1,29,56 A-14,18 T-16	Po E-7,55	At A-36 E-36 T-36	Rn
Fr	Ra E-69	Ac E-7, 15, 69															
				Ce Em-31	Pr	Nd	Pm T-21,54 E-22 Em-31	Sm Em-34	Eu Em-34	Gd	Tb	Dy	Ho	Er Em-46 T-49	Tm Em-46	Yb Em-46	Lu
				Th E-15, 17 37, 69 A-18	Pa E-6, 28 A-12	U E-7, 17 33,37 T-17, 33	Np E-7, 17, 37 60	Pu T-17,33,47 E-17,33,37 48, 60	Am E-37 T-47	Cm E-37,65 T-47	Bk	Cf					

T - THERMAL VACUUM EVAPORATION
E - ELECTRODEPOSITION
Em - ELECTROMAGNETIC
A - ADSORPTION

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2. Ballentine, R. and Burford, D. D., Anal. Chem. 26, 1031 (1954).

3. Bergstrom, I., Arkiv f. Fysik 5, 191 (1952).

4. Bisi, A., Terrani, S., and Zappa, L., Nuovo Cimento (10) 2, 1297 (1955).

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10. Burford, A. O., Perkins, J. F., and Haynes, S. K., Phys. Rev. 99, 3 (1955).

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65. Smith, W. G. and Hollander, J. M., Phys. Rev. 101, 746 (1956).

66. Steigman, J., Phys. Rev. 53, 771 (1938).

67. Stoker, P. H. and Ong Ping Hok, Physica 19, 279 (1953).

68. Thulin, S., Arkiv f. Fysik 9, 107, 137 (1955).

69. Toedt, F., Zeit. f. Phys. Chem. 113, 329 (1924).

70. Tompkins, P. C. and Bizzell, O. M., Ind. Eng. Chem. 42, 1469 (1950).

71. Van Cleve, A. and McDonough, F. D., Nucleonics 12, 53 (1954).

72. Wright, B. T., Phys. Rev. 71, 839 (1947).

ions collected on the wire, which was then used as the source.[15] A 149 $\mu g/cm^2$ Lu-176 source has been prepared by suspending Lu_2O_3 powder in amyl acetate in which was dissolved Durofix to act as an adhesive.[16] Rb-88 has been electrostatically deposited from a fission gas mixture onto an aluminum foil 0.15 mg/cm^2.[17] By utilizing an ultrasonic vibrator, a very uniform 15 $\mu g/cm^2$ source of C-14 has been deposited.[18] Another interesting method has been described by Haworth et. al.[19] for the preparation of a Co-60 source. A solution of stearic acid and benzene was spread on the surface of a water solution of the active isotope (Co-60). The benzene then evaporated, leaving a monolayer of stearic acid, which acts as an adsorbant for the activity. The deposition rate can be increased by increasing the temperature, the concentration of the divalent ion, and the pH of the solution. The monolayer is then removed on a thin backing and is ready for use. The number of monolayers acquired by this method is unlimited and can be built up if a more active source is needed.

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[15] Bunker, M. E., Duffield, R. B., Mize, J. P., and Starner, J. W., Phys. Rev. 103, 1417 (1956).

[16] Dixon, D., McNair, A., and Curran, S. C., Phil. Mag. 45, 683 (1954).

[17] Thulin, S., Arkiv for Fysik 4, 363 (1952).

[18] Wu, C. S., and Feldman, L., A.E.C.D. 2420 (1948).

[19] Haworth, D. T., Koch, J. R., Martinovich, R. J., and Surak, J. G., Nucleonics 13, No. 11, 112 (1955).

III. Autoradiography

A measure of the uniformity of the beta sources was obtained by autoradiography, a technique in which an ionization-sensitive emulsion is placed near or in contact with a specimen in order to determine the location and intensity of its radioactive deposition.[20] If sufficient energy is adsorbed from the radiation by the emulsion, its halide grains are rendered developable. The degree to which the image corresponds to the radioactive specimen depends, in varying amount, upon the specimen thickness, the emulsion thickness, the width of the separating gap, and the range of the radioactive particles.[21] From geometrical considerations, it is evident that the size of the image will always exceed that of the original specimen.

Autoradiographs can be divided into two categories: In high resolution autoradiography the tracks of the individual particles are studied, while in contrast autoradiography (which was used here), emulsion densities become high enough to allow densitometric measurements. An Ansco densitometer, having a slit of 0.13 mm^2 in area, was employed to measure the variations in uniformity of the photographic

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[20] Norris, W. P. and Woodruff, L. A., ANNUAL REVIEW OF NUCLEAR SCIENCE, Annual Reviews, Inc., Stanford, California, 1955, pp. 297-323.

[21] Yagoda, H., RADIOACTIVE MEASUREMENTS WITH NUCLEAR EMULSIONS, John Wiley and Sons, Inc., New York, 1949, pp. 17-20.

emulsions. In order to determine the relation of the densitometer reading to film darkening, the densitometer was read with a number of sheets of paper of uniform density placed between the photo cell and the light source. Each additional sheet of paper increased the darkening proportionately. The values obtained are shown graphically in Fig. 1. Twenty readings were taken of each autoradiograph and the two extreme readings were used. The percent variation in the uniformity of the emulsion was then calculated from the graph in Fig. 1.

All sources were autoradiographed with Dupont-502 film from the Dupont-552 industrial pack, which is sensitive from 30 milliroentgens to 10,000 milliroentgens and is energy dependent. That is, excessive darkening of the film occurs for beta energies between 0 and 300 kev, whereas for higher energies the rate of darkening becomes almost constant.

The film was separated from the source by 1 mg/cm^2 of polystyrene to prevent contamination of the film and subsequently the photographic solutions. The exposure time was based on the counting rate of the source relative to a previous source for which autoradiographs had been made, with corrections for the energy dependency of the film. The exposure time was long enough to show the contrasts due to irregularities in the radioactive source specimen, but not so long that the film was blackened to the extent of obliterating all detail.

After the film had been properly exposed, it was developed for three minutes in Dupont x-ray film developer, rinsed thoroughly in water, fixed in Dupont x-ray film hypo for twenty minutes, and washed in circulating water for one hour.

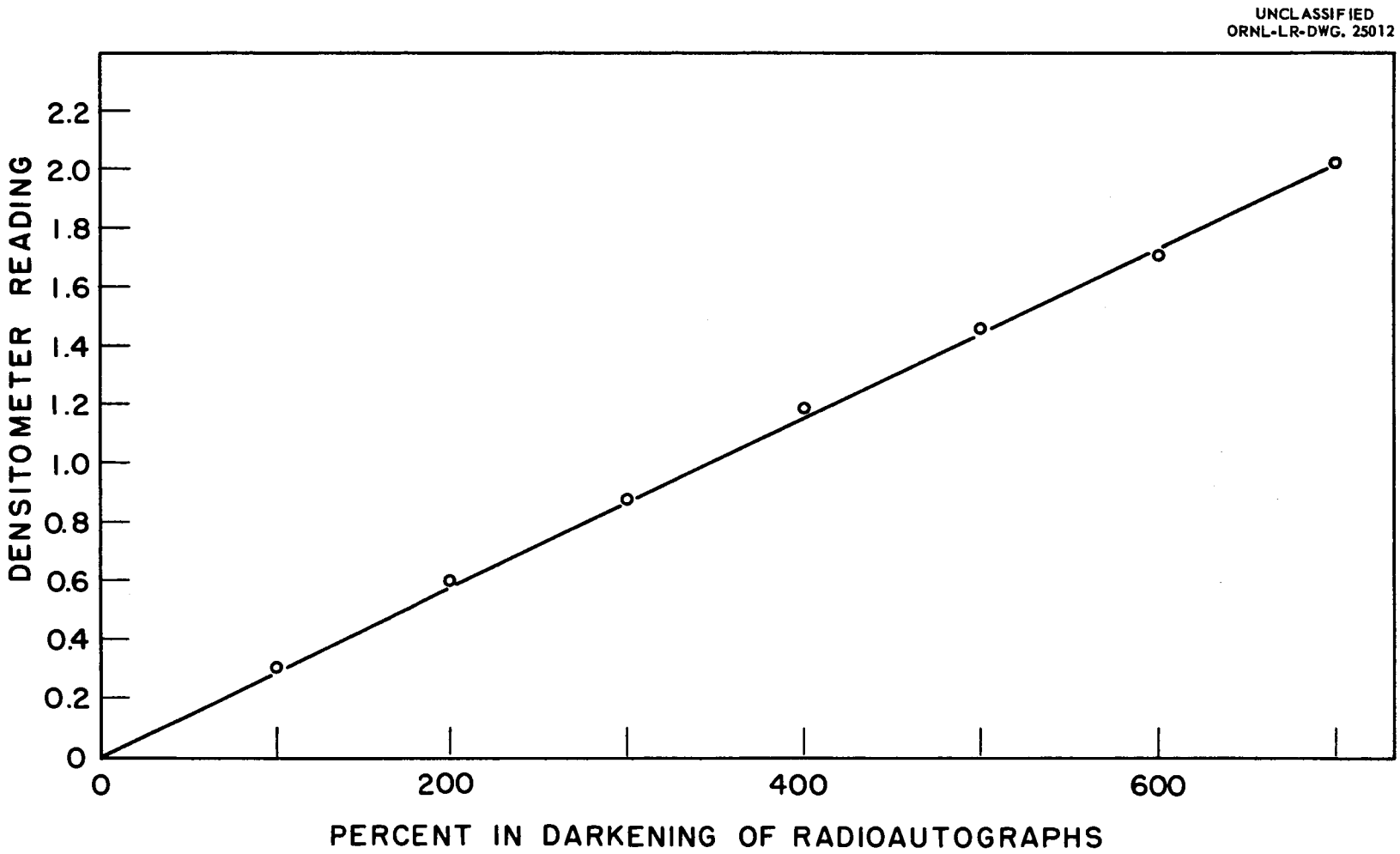

Fig. 1. The Calibration of the Densitometer.

IV. The Source Mounting

It has been shown that back scattering from an infinitely thick source mounting is strongly dependent upon the atomic number of the mounting, increasing from 17% for beryllium to 80% for lead.[22] Hence, it is evident that the source mountings should be made from materials having low atomic weight, such as aluminum or organic compounds.

Formvar film was chosen as the source mounting because of its strength, the ease with which it may be made into very thin films, and its homogeneity even at thickness of 2 $\mu g/cm^2$.[23] Polystyrene was also employed where resistance to acids was needed, since formvar is acted upon quite readily by acids. To test the extent of acid resistance, a film was left in 1.0 M solutions of nitric acid, hydrochloric acid, and sulphuric acid for twenty-four hours without physical change being observed.

A 0.5% formvar solution was prepared with ethylene dichloride as the solvent, and a 1.0% polystyrene solution was prepared in benzene. Care was taken to keep the evaporation of these solutions at a minimum since any evaporation would concentrate the solutions and produce thicker films.

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[22] Siegbahn, K., BETA- AND GAMMA-RAY SPECTROSCOPY, Interscience Publishers, Inc., New York, 1955, p. 260.

[23] Ibid., pp. 261-62.

The films were prepared as described by Siegbahn.[24] A glass plate, previously cleaned and free of all dust particles, was dipped into the formvar solution, let dry, and then dipped into the polystyrene solution and let dry again. The glass plate was then submerged into a small vat of clean, distilled water. The thin film was floated off the glass plate onto the surface of the water with the aid of forceps. Source rings were coated on one side with rubber cement and carefully placed on the film. The source film, now attached to the ring, was trimmed and allowed to dry. The thickness of the films were governed by the concentrations of the two solutions. Films were measured and weighed and found to have a surface density of 35 $\mu g/cm^2$.

When a beta source decays, a positive charge remains on the source; hence, it must be grounded to prevent the slowing down of beta particles emitted, subsequently. Hence, a metallic film was thermally vaporized in a vacuum upon the formvar film to render the source conducting. The metal to be evaporated was placed either on a tungsten filament or in a tantalum boat, and the formvar film was mounted above the filament. A bell jar was placed over the arrangement and evacuated to about 75 microns with a Welch-1397 fore pump determined by a Stokes McLeod pressure gage. When the pressure reached this point, two Distillation Products-MC-275 oil diffusion pumps were added to the system, and remained pumping throughout the remainder of the process.

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[24] Siegbahn, K., BETA- AND GAMMA-RAY SPECTROSCOPY, Interscience Publishers, Inc., New York, 1955, pp. 261-62.

When the pressure reached about 10^{-4} mm of mercury, determined by a General Electric ion gage, the filament was heated and the metal was vaporized. The current was increased slowly until the metal melted and wetted the filament, at which time the current was increased further until vaporization began, the only limitation being the amount of heat the formvar film could withstand. Usually at the beginning of the heating process, the pressure in the bell jar increased. When this happened, the heating process was discontinued until the pressure reached its previous value. Gold, aluminum, magnesium, or copper were vaporized onto the source backings. Assuming that the metal was emitted isotropically when a tungsten wire filament was used, the thickness of the vaporized metal was given by the equation $W_1 - W_2 \big/ 4\pi R^2$, where W_1 and W_2 were the weights of the filament and metal before and after vaporization respectively, and R was the distance from the filament to the source mounting. The thickness of the vaporized metallic films, calculated in this manner, were about 30 $\mu g/cm^2$. These sources were used in a beta-spectrometer designed for a disc source 0.6 mm in diameter. An annular ring source was used, with the beta rays passing into the spectrometer through a 0.6 mm diameter hole coaxial with ring source and spaced from it a distance equal to the radius of the ring. This source geometry offered no advantages over a disc source and was employed merely because the spectrometer was set up to accommodate a source of this type in another application. The dimensions of the ring were calculated to be 1.33 cm in outside diameter and 0.6 mm in radial width. The thin, metallic conducting films were vaporized onto the

source backing film through an aluminum foil mask cut to the above dimensions.

V. The Preparation of Sources by Solvent Evaporation

The evaporation method of source preparation, in general, yields nonuniform deposits. However, this method may be used as a standard with which to compare the other methods of source preparation.

An autoradiograph prepared from an evaporated cerium-144 source is shown in Fig. 2. The activity was distributed in a nonuniform ring created by surface tension during the evaporation, and by the hydrophobic characteristic of the source mounting.[25] The variation in uniformity of the autoradiograph was about 400%.

In an attempt to eliminate these effects, insulin was used. Insulin tends to decrease the surface tension and render the surface of the film hydrophilic. Hence, the source spreads readily, covering a larger area and becoming more evenly distributed when evaporated.

The evaporation with the aid of insulin was performed by three methods. First, 0.025 ml of ruthenium-106 solution was mixed with 0.025 ml of insulin (40 units/cc). This was transferred to a source mounting and evaporated to dryness (see Fig. 2, No. 1). Second, a drop of insulin was placed on a source mounting and then removed, leaving only a wet spot of insulin, after which 0.025 ml of carrier-free ruthenium-106 solution was placed on the wetted mounting and evaporated

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[25] Pate, B. D. and Yaffe, L., Can. J. Chem. 34, 266 (1956).

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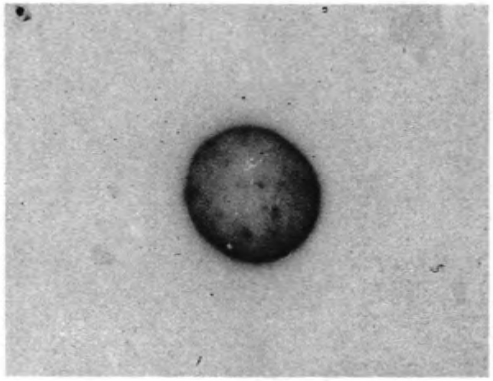

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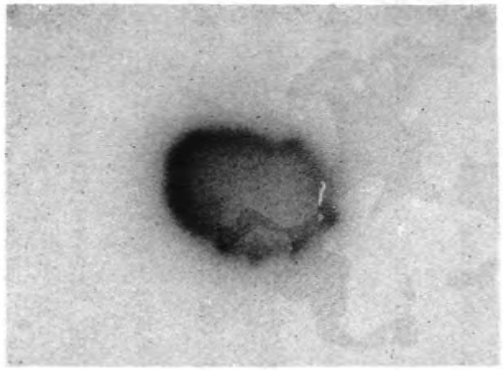

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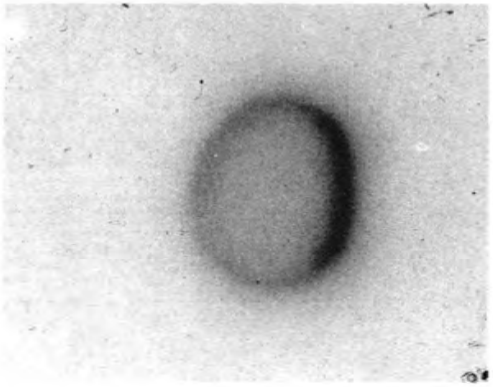

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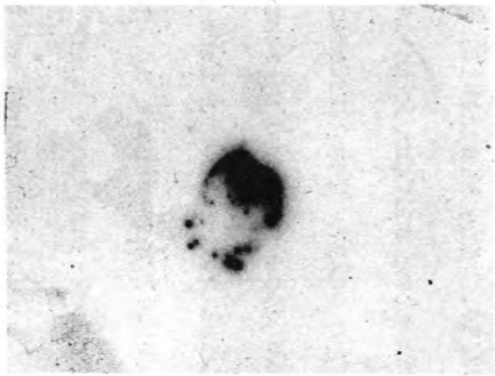

1. EVAPORATION OF RUTHENIUM – 106 WITH INSULIN BY MIXING

2. EVAPORATION OF RUTHENIUM – 106 WITH INSULIN BY WETTING

3. EVAPORATION OF RUTHENIUM – 106 BY PARTIAL EVAPORATION

4. EVAPORATION OF CERIUM – 144

Fig. 2. Autoradiographs of Depositions by Evaporation.

to dryness (see Fig. 2, No. 2). Third, a drop of insulin was placed on the source mounting and evaporated nearly to dryness. Then 0.025 ml of carrier-free ruthenium-106 solution was added and taken to complete dryness (see Fig. 2, No. 3). The ring still exists to some extent, although the over-all source is more uniform with less clumping than the source prepared by evaporation only. The uniformity variation is about 140%. Therefore, the insulin does render the source more uniform. It is difficult to distinguish which of the three methods listed above give the best results since all three have about the same characteristics. Possibly number one is somewhat more uniform since the variation in uniformity is about 10% less than that of the other two.

VI. Source Preparation by the Radiocolloid Method

Under certain conditions, radioelements act as true colloids, in which the particles are non-ionic and not in solution, and are distributed throughout the solution as small discrete particles. This occurs in very dilute radioactive solutions and when macro-amounts of the compound would be insoluble. Because of this similarity, radioelements under these conditions have been termed "radiocolloidal." To explain the nature and origin of these formations, two theories have been developed.[26] One theory is that these radioelements are true colloids. Colloidal solutions contain particles which are larger than molecules but too small to be seen with a microscope.[27] These particles are insoluble, non-ionic, and charged so that other particles are repelled and no growth of these particles takes place. The other theory states that the radioelements are adsorbed by impurities in the solution, thus appearing to be colloids. Neither theory has been completely verified and it is believed that an acceptance of both actions is necessary to explain radiocolloids.

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[26] Schweitzer, G. K. and Jackson, M., J. Chem. Ed. 29, 518 (1952).

[27] Glasstone, S., ELEMENTS OF PHYSICAL CHEMISTRY, D. Van Nostrand Company, Inc., New York, pp. 557-558 (1946).

When radionuclides act as colloids they can be adsorbed, centrifuged, filtered, and will settle by the force of gravity, even though the solubility product is not exceeded. Hence, radionuclides of elements forming insoluble sulfides or hydroxides were prepared in solutions and permitted to settle onto a source mount as a method of source preparation.

A. The Precipitation Method

First, 0.025 ml of a carrier-free or high specific activity solution, 0.1 M in hydrochloric or nitric acid, was placed upon a source mount which in turn was placed in a desiccator. The desiccator was then evacuated and ammonia gas was admitted to form an insoluble hydroxide. After one hour, the source mount was removed and the solution drawn off with a micro pipette. Then 0.025 ml of distilled water was placed on the mount in the same area as before and placed again in the desiccator. Again the desiccator was evacuated and ammonia gas admitted. After another hour, the source mount was removed and the solution drawn off as before. The source was then counted with a G-M counter and compared with the counting rates obtained from the sources prepared by evaporation (Chapter V). For 100% deposition, the counting rates should be approximately equal for high energy beta sources, and greater by precipitation for low-energy radionuclides. Source preparation by this method was conducted for the carrier-free radionuclides listed in Table III, along with the per cent of deposition.

The same procedure was followed for carrier-free or high specific

Table III. The Percent Deposited by Radiocolloid Precipitation

Radionuclide	Deposited in NH_3 Atmosphere	Deposited in H_2S Atmosphere
Phosphorous-32	9.5%	
Scandium-46	20.4	
Selenium-57	1.4	36.7%
Cobalt-60	1.5	12.1
Yttrium-91	32.4	
Ruthenium-106	11.2	34.0
Silver-110	65.6	50.0
Indium-114	1.4	13.4
Cerium-144	28.2	
Iridium-192	4.4	3.0
Mercury-203	27.1	82.0

radionuclides in a sulfide atmosphere produced by hydrogen sulfide in an attempt to form an insoluble sulfide. These radionuclides with the per cent deposited are also lited in Table III.

It may be noted that as a general rule, the per cent deposited was not very high. By the hydroxide precipitation, silver-110 was considerably higher than any of the others, while the depositions of scandium-46, ytterium-91, cerium-144, and mercury-203 were appreciable.

The length of time that the solutions remained in the chemical atmospheres had some effect upon the amount of activity deposited. Cerium-144 sources were prepared with reacting times of 10 minutes, 30 minutes, 1 hour, 2.5 hours, and 6 hours to determine this effect. The results are shown in Fig. 3. The per cent deposited was a maximum at 2.5 hours and remained approximately constant thereafter.

Autoradiographs were prepared from sources prepared by this method and appear in Fig. 4. Number one is an autoradiograph of a cerium-144 source prepared as an insoluble hydroxide in an ammonium atmosphere. Number two is an autoradiograph of a mercury-203 source prepared as an insoluble sulfide in a hydrogen sulfide atmosphere. These two autoradiographs indicate a somewhat non-uniform sample. The density variance, measured with the densitometer, of the autoradiographs 1 and 2 are 40% and 30%, respectively. It is possible that this non-uniformity is due to the radioactive particles settling unevenly on the source mount or because of unclean surfaces which were too thin to clean properly.

To determine the adherence of the activity to the source mounting, the source was held upside down while being tapped quite hard with no

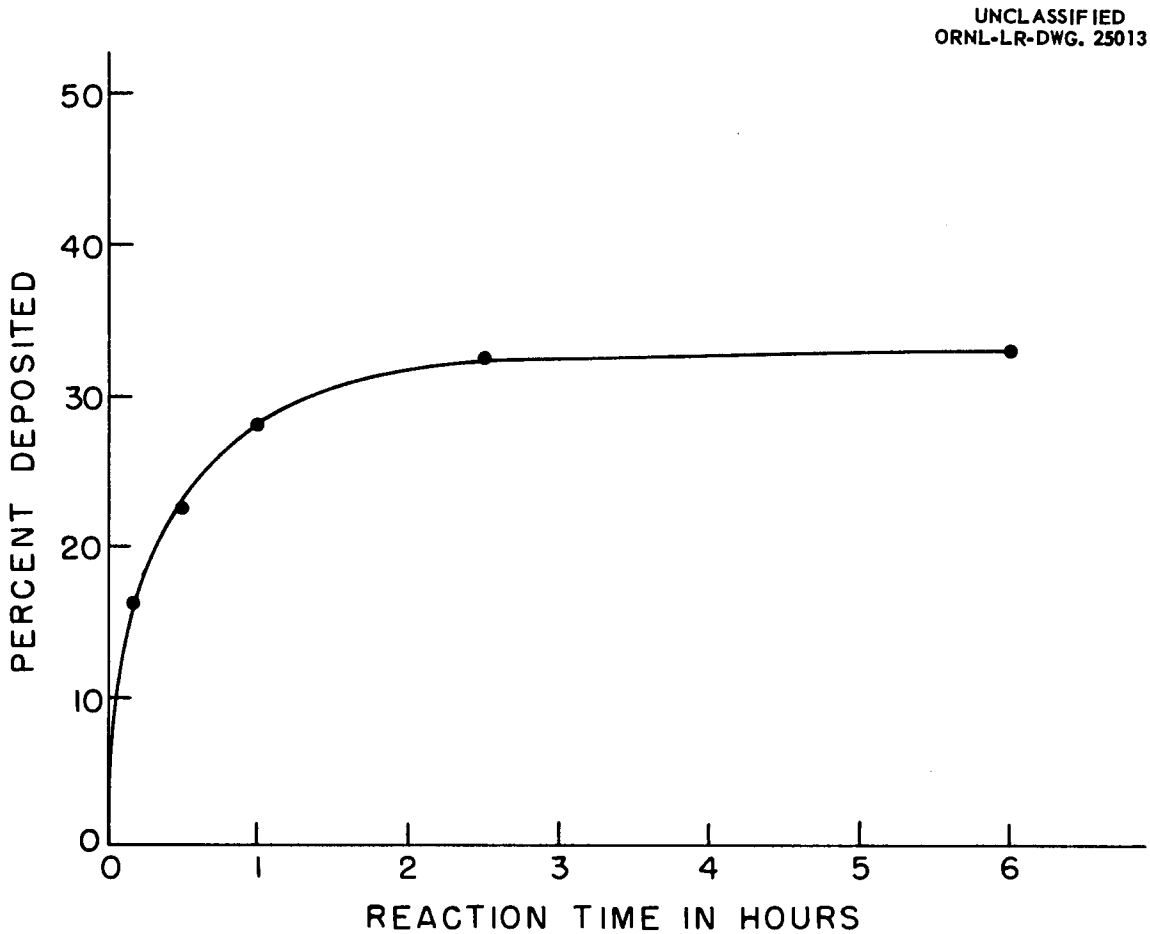

Fig. 3. Deposition Rate of Ce-144 by Radiocolloid Precipitation.

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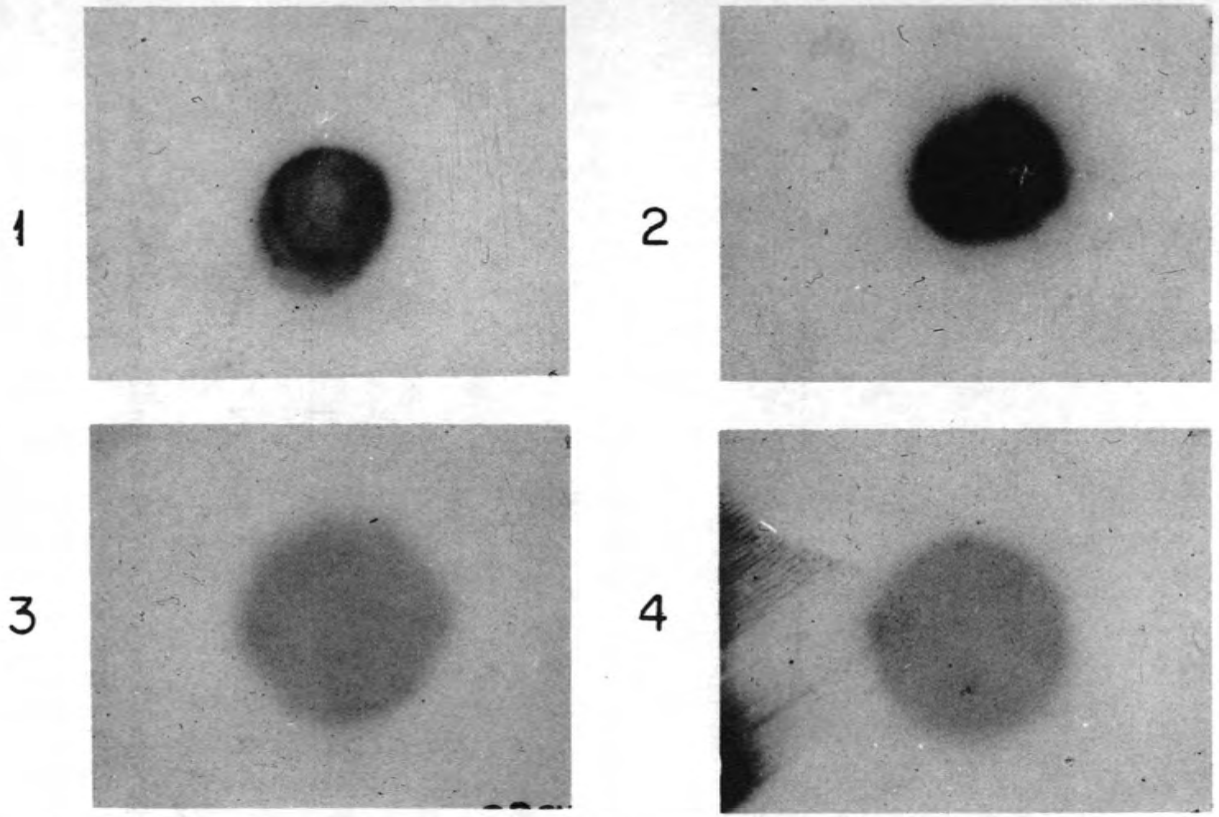

1. CERIUM-144 DEPOSITED ON FORMVAR IN A NH_4OH ATMOSPHERE

2. MERCURY-203 DEPOSITED ON FORMVAR IN A H_2S ATMOSPHERE

3. MERCURY-203 DEPOSITED ON GOLD BY REDUCTION IN 0.1 *M* HNO_3

4. SILVER-110 DEPOSITED ON GOLD BY REDUCTION IN 0.1 *M* HNO_3

Fig. 4. Autoradiographs of Sources Prepared by Radiocolloid Methods.

apparent loss of activity. The source was next introduced into a strong jet of air blowing horizontally across it, which produced a 2.0% loss in activity. The source was then rubbed quite firmly with tissue producing a 10% loss in activity. The adherence of the activity of this source was 87% greater than that of a source prepared by solvent evaporation.

B. The Reduction Method

Many radionuclides may be reduced to their metal state by another metal in solution and will then be adsorbed onto the reducing metal.[28] The metal vaporized onto the source mounting, Chapter IV, was used as the reducing metal, and the reduced radionuclides were then adsorbed onto the residual metal and not upon the glass container, since the reaction took place near the film. Since the metal is slowly dissolving, its surface will be continually cleaned, and this should eliminate the effects of an unclean surface found in previous studies of adsorption on metal surfaces.[29] The greatest difficulty in this method arises from the fact that the metal is dissolving, so that the reaction must be halted before the metal is completely dissolved.

Preliminary experiments were conducted with the radionuclides and

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[28] Bonner, N. A. and Wahl, A. C., RADIOACTIVITY APPLIED TO CHEMISTRY, John Wiley and Sons, Inc., New York, pp. 169-171 (1951).

[29] Hensley, J. W., Long, A. O., and Willard, J. E., Ind. Eng. Chemistry 41, 1419 (1949).

the reducing metals listed in Table IV. The metals were cut into strips and inserted into lusteroid tubes containing a radioactive 0.1 M acid solution which had been previously counted in a sodium iodide crystal, well-type scintillation counter. Lusteroid tubes were used instead of glass to minimize the adsorption on the tube walls. The metal strips were immersed in the active solution for 12 hours with occasional stirring. At the end of this time the metal strips were removed, washed, and counted along with the lusteroid tubes containing the supernatant solution. The solution was then removed, and the lusteroid test tubes were washed with water and counted. Table IV indicates that magnesium is the better reducing metal as would be expected since it has the greater positive potential. For this reason gold is a poor reducing agent and was used only as a blank, although silver-110 and mercury-203 deposited to a great extent upon the gold. Erbacher et al.[30,31] also found this to be true in the cases of Pb, Bi, Th, and Ag adsorbed on Au, Pb, and copper and concluded it was due to surface adsorption which follows the Freundlich isotherm; i.e., the log of the adsorption is proportional to the log of the concentration.

The same procedure was conducted a second time with silver-110, iridium-192, selenium-75, and ruthenium-106 except that this time sulfurous acid was added as a reducing agent to aid with the reducing

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[30] Erbacher, O., Herr, W., and Wiedemann, M., Z. Naturforsch. 3a, 637 (1948).

[31] Erbacher, O., Z. Physik. Chem. Abit. A, 182, 243 (1938).

Table IV. The Percent Deposited by Reduction and Radiocolloid Adsorption Occurring in 12 Hours

Isotope	Steel	Magnesium	Aluminum	Gold	Copper
Cobalt-60					
Adsorbed on Metal	0.1%	67.4%	0.2%	0.2%	0.2%
Adsorbed on Tube	0.2	2.8	0.0	0.1	0.1
Chromium-51					
Adsorbed on Metal	0.3	37.4	0.3	0.6	4.3
Adsorbed on Tube	0.1	3.6	0.0	0.3	13.2
Indium-114					
Adsorbed on Metal	2.4	31.3	0.3	3.1	23.2
Adsorbed on Tube	1.0	10.3	0.3	5.6	35.1
Iridium-192					
Adsorbed on Metal	1.0	69.1	1.4	0.6	4.3
Adsorbed on Tube	0.4	2.7	0.1	0.2	0.6
Mercury-203					
Adsorbed on Metal	65.0	22.5	44.9	99.9	64.3
Adsorbed on Tube	6.0	4.2	4.2	1.4	4.1
Ruthenium-106					
Adsorbed on Metal	9.2	72.5	48.1	8.5	36.3
Adsorbed on Tube	10.7	0.8	1.2	5.5	14.5
Selenium-75					
Adsorbed on Metal	42.1	75.4	42.8	1.1	15.1
Adsorbed on Tube	6.6	5.3	15.3	1.1	4.7
Silver-110					
Adsorbed on Metal	18.4	64.9	72.4	87.2	92.0
Adsorbed on Tube	36.5	11.5	11.2	4.5	2.2

process. The precipitate of elemental sulfur which resulted was separated and counted. The same metals were used except for magnesium, which was dissolved when the sulfurous acid was added. The sulfurous acid seemed to have varied effects since it increases the deposition in some instances and decreases the deposition in others, as shown in Table V. Also, considerable activity was carried down with the precipitate when the aluminum and copper metals were used.

To determine the effect of the acid concentration upon adsorption, the same radionuclides were repeated with magnesium metal in a 0.01 M nitric acid solution. The adsorption onto the magnesium metal was practically the same as in 0.10 M nitric acid, but the adsorption onto the walls of the lusteroid tubes, in most cases, increased. Therefore, better conditions are produced in 0.10 M acid solutions.

In the case of volatile nuclides, the metals with the adsorbed activities were exposed to decreased pressures and increased temperatures to see how well the activity on the metal would remain. The results are listed in Table VI. The mercury-203 volatilized quite readily from all metals except copper. The activities of all other radionuclides remained practically unchanged.

As a result of the data accumulated in the preliminary work, the following conditions were chosen for source preparation: (1) silver-110 on gold film, (2) mercury-203 on gold film, (3) ruthenium-106 on magnesium film, (4) silver-110 on aluminum film, and (5) silver-110 on copper film. A glass column, coated with "Desicote" to diminish adsorption on the glass walls, and of the correct diameter to fit the

Table V. The Percent Deposited by Reduction with Sulfurous Acid and Radiocolloid Adsorption Occurring in 12 Hours

Isotope	Steel	Magnesium	Aluminum	Gold	Copper
Silver-110					
Adsorbed on Metal	79.9%		52.7%	96.3%	93.6%
Adsorbed on Tube	4.4		6.8	1.2	1.6
ppt. Activity	2.6	Mg dissolved	4.5	0.3	1.3
Iridium-192					
Adsorbed on Metal	0.8		0.9	2.1	0.6
Adsorbed on Tube	0.1		0.1	0.0	0.1
ppt. Activity	2.4		2.1	2.6	2.4
Selenium-75					
Adsorbed on Metal	73.8		16.6	1.8	27.0
Adsorbed on Tube	3.9		52.5	0.1	22.9
ppt. Activity	1.1		8.4	0.4	39.6
Ruthenium-106					
Adsorbed on Metal	6.2		24.0	1.7	1.0
Adsorbed on Tube	0.4		27.0	1.1	0.3
ppt. Activity	1.0		26.7	1.2	2.4

Table VI. The Effects of Pressure and Temperature on the Adherence of Mercury-203 to the Source Backing

	Percent Adhering		
Metal	Pressure of 27.5 Inches of Hg	100° C	300° C
Steel	97%	65%	9%
Magnesium	65%	36%	9%
Aluminum	53%	23%	9%
Gold	100%	12%	3%
Copper	97%	92%	60%

source ring, was clamped to the source mounting and made water tight by a rubber gasket between the glass and the metal source ring. A 0.1 M acid radioactive solution was poured into the column and a platinum stirring rod coated with wax to eliminate adsorption was inserted for continual stirring. The reaction was continued until the maximum amount of activity was adsorbed or until the reaction had to be halted for fear of the metal surface dissolving completely. The deposits created by adsorption could not be seen.

The data obtained by this method are presented graphically in Fig. 5 as the per cent of the activity deposited versus time. The results of the adsorption onto the thin foils correspond to that of the preliminary results quite closely. However, the adsorption was more rapid, presumably because of the continual stirring. From the total activity deposited, only 1.4% was deposited upon the stirrer and 2.3% upon the glass column.

The sources produced by this method were autoradiographed, and those of the sources prepared by the adsorption of mercury-203 onto gold film and of silver-110 onto gold film appear in Fig. 4. The autoradiographs appear to be very uniform with a density variation of only approximately 1.5%.

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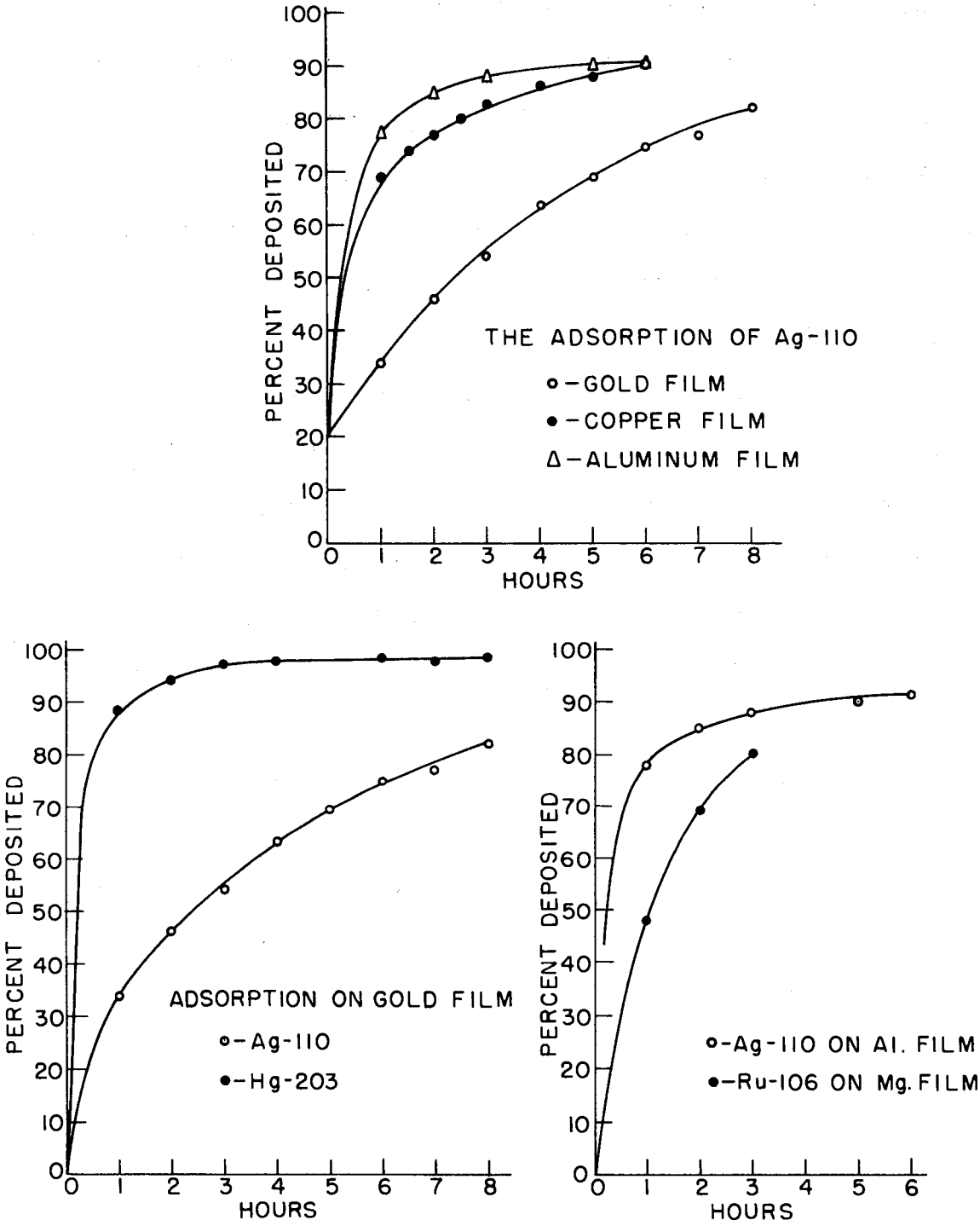

Fig. 5. Radiocolloid Adsorption on Various Metallic Films.

VII. Sources Prepared by Electrodeposition

Since the source mounting to be used in a spectrometer must already have a metallic coating, it is possible that this coating may also serve as a cathode in an electroplating process. The films employed here were very thin, 30 micrograms/cm^2, but sufficiently thick to render them conducting.

Gold was chosen as the best metal to use as the cathode because it is chemically inert and easily vaporized. The vaporization was conducted as before, except that a small tab of gold was vaporized onto the film backing which led from the gold ring, deposited just inside the source ring, to the outer edge of the source ring shown in Fig. 6. By attaching an electrical contact to the gold tab on the outer edge of the source ring, a current flowed to the inner gold deposit, which was insulated from the metal source ring by the formvar-polystyrene film. Therefore, when the platinum stirring rod was attached to a positive terminal of a battery and the metal film on the source mount to the negative terminal, a current flowed through the solution.

There are a number of different electrolytic cell designs described in the literature.[32-34] The cell which was designed and

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[32] Lee, D. and Cook G. B., AERE-C/R-430, 12 (1950).

[33] Caldwell, P. A. and Graves, J. D., Nucleonics 13, No. 12, 49 (1955).

[34] Broyles, C. D., Thomas, D. A., and Haynes, S. K., Phys. Rev. 89, 715 (1953).

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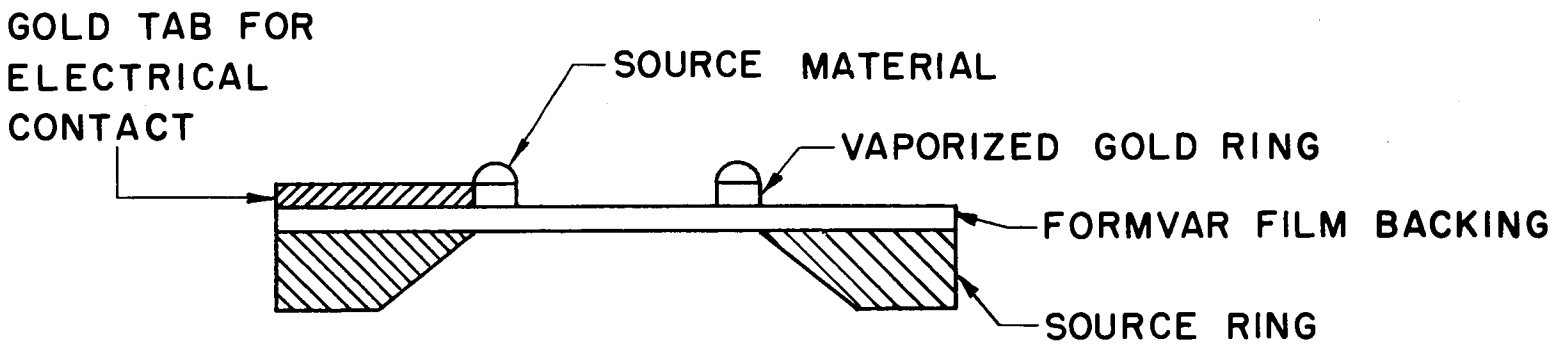

Fig. 6. The Cross-Sectional View of the Source and Source Mounting.

used in this work is shown in Fig. 7 and Fig. 8. Figure 7 shows the component parts of the cell and Fig. 8 shows the cell assembled. The source ring and mounting fit into an annular groove in the base shown at the left in Fig. 7. A rubber gasket, shown to the left of the glass column, is then inserted over the metal source ring and a thin metal lead makes contact with the thin strip of vaporized gold on the outer edge of the source ring. The lead, shown directly in front of the glass column, passes through the groove in the base and makes contact with a negative external potential. A glass column, 7/8 cm by 5 cm, and previously coated with "Desicote" to reduce adsorption on the glass walls, is placed on the rubber washer. The glass column is fitted into a groove in the lucite top shown at the right and is clamped tight by three bolts and wing nuts. When the glass column is clamped into place, both the column and the source ring are pressed into the rubber washer to make the connection water tight. The rubber washer is rigid enough so that it will not spread out onto the gold surfaced area as the column is clamped tight. All components of the cell fit so that only the inner surface of the gold foil is exposed to the solution. A platinum stirring rod, shown directly above the three bolts in Fig. 7 is inserted through a hole in the top of the cell and connected to a positive potential. When the deposition is completed, the stirring rod is removed and the solution is pipetted out through the hole in the top of the cell. The emf was provided by a simple series circuit including a voltmeter, a milliammeter, a 1000 ohms "Helipot" resistor, and a 6 volt dry cell. The same cell was used in the source preparation by the radiocolloid method, Chapter VI, although in this case, no

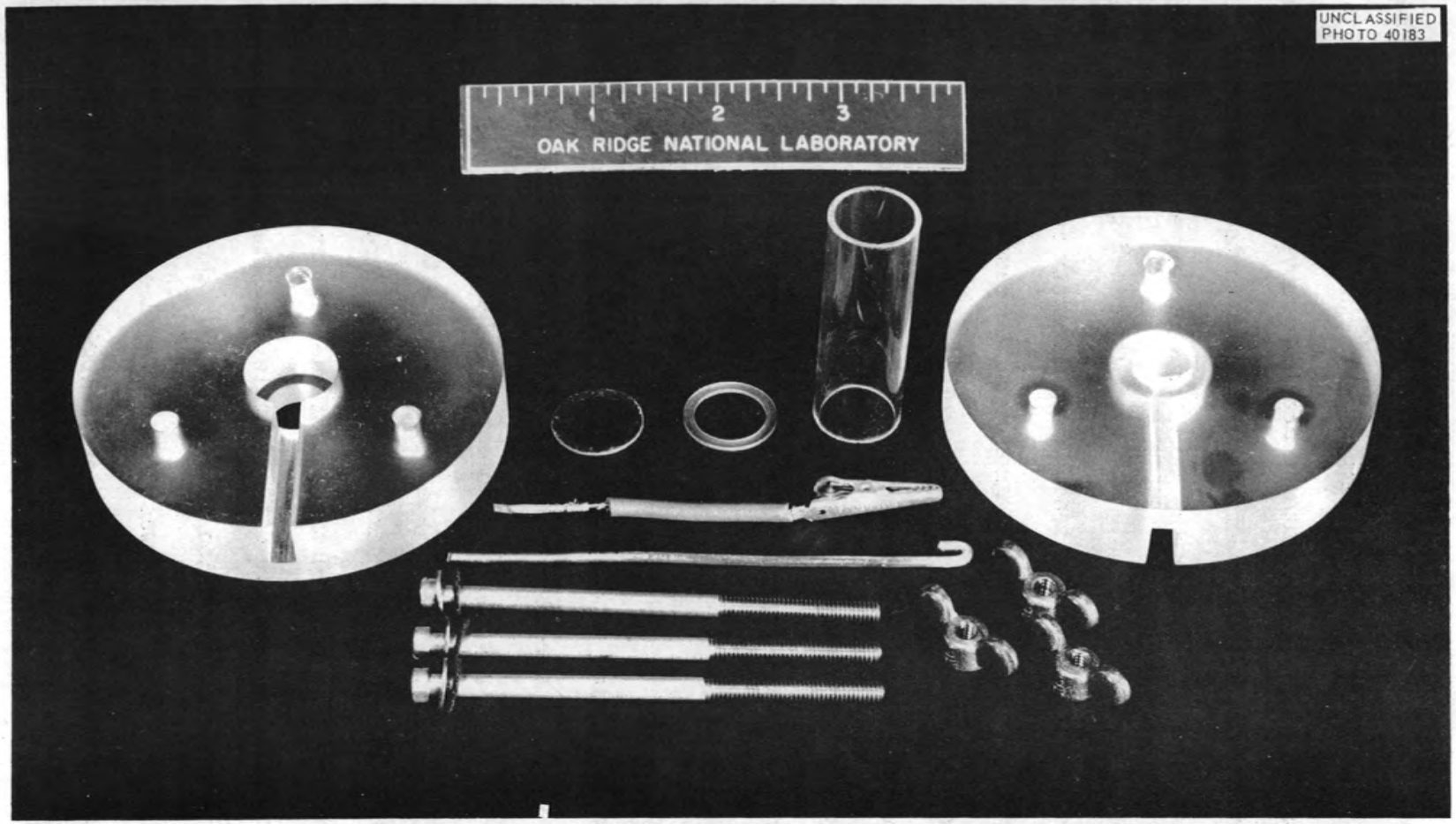

Fig. 7. The Component Parts of the Electrolytic Cell.

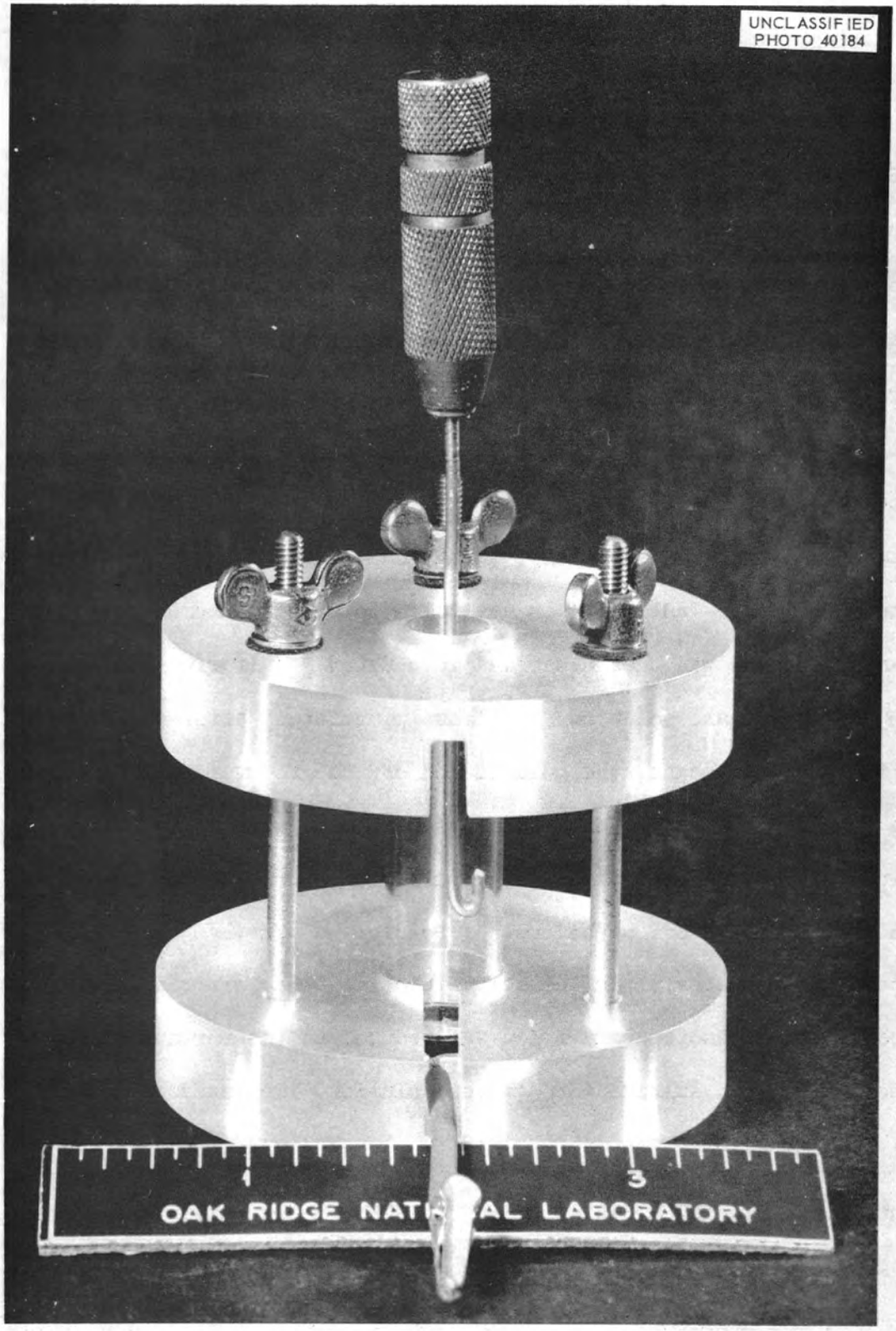

Fig. 8. The Electrolytic Cell Assembled.

electrical current or contacts were needed.

By using an external applied emf and the electrolytic cell described, ruthenium-106, silver-110, mercury-203, cobalt-60, indium-114, chromium-51, gold-198, and iron-59 were electrodeposited onto the gold coated source mountings from the solutions obtained from the Isotopes Division after first being made 0.1 M acid. These data are presented in Table VII. The percentage of the activity deposited on the source mountings was rather low in the cases of cobalt-60, indium-114, chromium-51, and iron-59. These four radionuclides were repeated using prepared electrolytes that had previously produced good results. An active InCl solution was taken to dryness and heated to form an oxide, and was then dissolved in a 0.025 M sulfuric acid and 0.35 M sodium citrate solution.[35] Almost immediately after the deposition process began, the gold flaked off the formvar film, even in such a low acid concentration. Attempts were also made to deposite chromium-51 as a sulfate in a 0.025 M sulfuric acid solution, but again the gold film flaked off as before.[36] Possibly a platinum cathode would have been more successful in the sulfuric acid. The ferric solution obtained from the Isotopes Division was reduced to the ferrous state by passing hydrogen sulfide through the solution. The excess hydrogen

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[35] Mantell, C. L., INDUSTRIAL ELECTROCHEMISTRY, McGraw-Hill Book Company, Inc., New York, p. 200 (1940).

[36] Blum, W. and Hogaboom, G. B., PRINCIPLES OF ELECTROPLATING AND ELECTROFORMING, McGraw-Hill Book Company, Inc., New York, pp. 288-307 (1930).

Table VII. Electrodeposition by an Applied EMF

Radionuclide	Chemical Form	Solution	Volts	Milli-Amperes	Time in Hours	Percent Deposited
Ru-106	$RuCl_3$	0.1 M - HCl	3.8	55	3.5	62.1
Silver-110	$AgNo_3$	0.1 M - HNO_3	2.8	20	3.0	92.2
Hg-203	$Hg(NO_3)_2$	0.1 M - HNO_3	3.0	20	3.0	94.4
Co-60	$CoCl_2$	0.1 M - HCl	2.9	55	4.0	1.4
In-114	InCl	0.1 M - HCl	3.4	50	3.0	33.1
Cr-51	$CrCl_3$	0.1 M - HCl	2.8	40	4.5	14.6
Au-198	$AuCl_3$	0.1 M - HCl + HNO_3	4.0	40	4.0	95.4
Fe-59	$FeCl_3$	0.1 M - HCl	3.2	55	3.0	1.9
Co-60	Co complex	Prepared Electrolyte	4.2	14	4.0	91.1
Fe-59	$FeCl_2$	Prepared Electrolyte	5.0	40	5.0	94.8

sulfide was boiled off by placing the ferrous solution in a stoppered flask with an outlet leading into a sodium bicarbonate solution, which prevented the ferrous ions from being oxidized back to the ferric state when boiled at atmospheric pressure. One-half gram of calcium chloride was added to the 6 ml electrolyte and the ion was then deposited.[37] For the active cobalt chloride solution, a complex was formed with ammonium peroxydisulphate by refluxing for 45 minutes. The solution was allowed to cool and ammonium hydroxide was added very slowly to neutralize the solution and then allowed to stand for 24 hours. The solution was then ready to be used as the electrolyte.[38] The results of the second iron-57 and cobalt-60 depositions are given in Table VII.

Autoradiographs were made of all sources prepared by electrodeposition. The uniformity of the autoradiographs varied from 3.6% to 8.8%. The autoradiographs prepared from a mercury-203 source and a ruthenium-106 source appear in Fig. 9. The variance in uniformity of the mercury-203 and ruthenium-106 sources appearing in Fig. 9 is 6.2% and 5.5%, respectively.

A. Internal Electrolysis

It has been demonstrated that metals can be electrodeposited without

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[37] Blum, W. and Hogaboom, G. B., PRINCIPLES OF ELECTROPLATING AND ELECTROFORMING, McGraw-Hill Book Company, Inc., New York, pp. 282-286 (1930).

[38] Caldwell, P. S. and Graves, J. D., Nucleonics 13, No. 12, 49 (1955).

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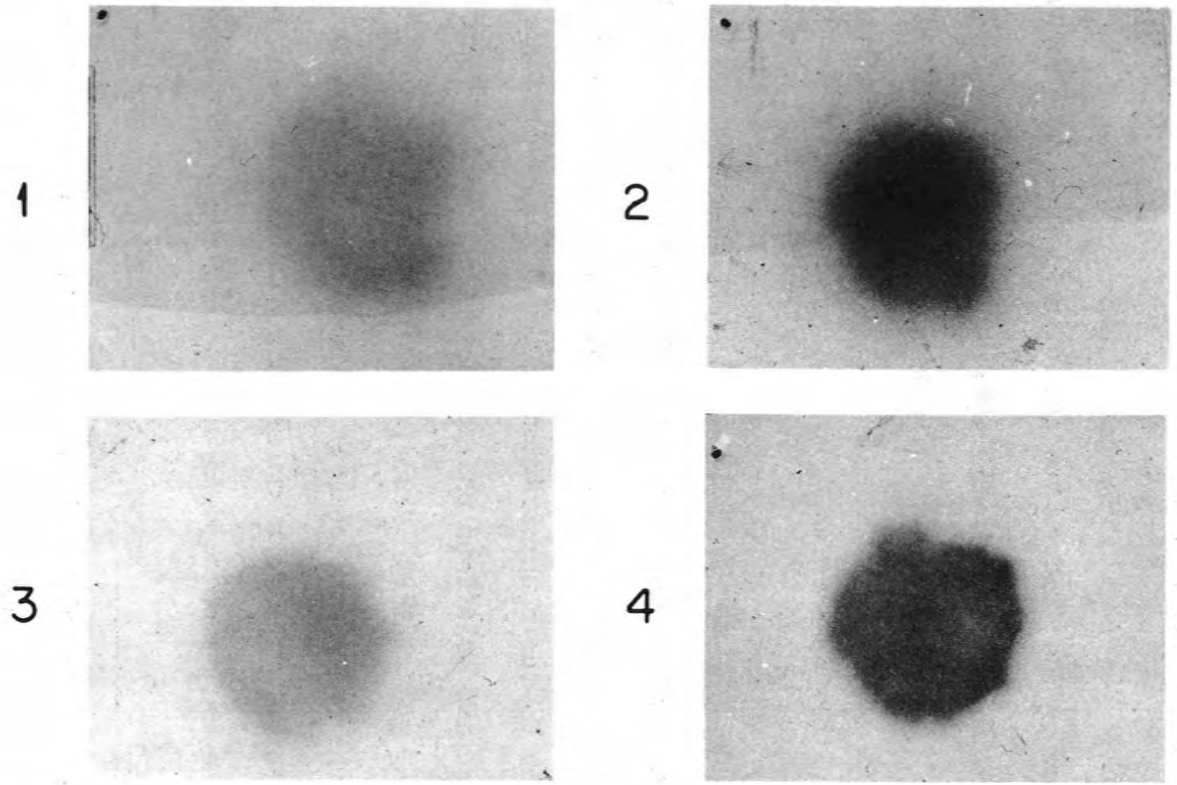

1. MERCURY-203 ELECTRODEPOSITED ON GOLD; 3 v, 20 ma IN 0.1 *M* HNO_3

2. RUTHENIUM-106 ELECTRODEPOSITED ON GOLD; 4 v, 55 ma IN 0.1 *M* HCl

3. INDIUM-114 INTERNAL ELECTRODEPOSITED ON GOLD IN 0.1 *M* HCl

4. GOLD-198 INTERNAL ELECTRODEPOSITED ON GOLD IN 0.1 *M* HCl

Fig. 9. Autoradiographs of Sources Prepared by Electrodeposition.

an external source of electrical energy by using electrodes with a sufficiently great difference in potential.[39] When these are immersed in an ionic solution, a current flows and deposition on the cathode can occur. The conditions at an inert cathode are exactly the same as if an external voltage was applied.

The concentration of the electrolyte has considerable effect upon the emf as can be shown by the Nernst equation,[40]

$$E = E^{o} - \frac{RT}{nF} \ln C_{M} , \qquad (1)$$

where E^{o} is the standard emf, R is a gas constant equal to 8.314 absolute joules, T the absolute temperature, F is equal to 96,500 coulombs, n is the valence change, and C_M the concentration of the depositable ion. In dealing with radioactive solutions the concentration reached factors in the order of 10^{-7} or 10^{-8} molar. Hence, the minus sign in Eq. (1) becomes positive and the emf of the cell is increased by an appreciable factor. However, at very low concentrations this equation does not hold true.[41] The emf of the cell also depends upon temperature, pressure,

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[39] Lingane, J. J., ELECTROANALYTICAL CHEMISTRY, Interscience Publishers, Inc., New York, pp. 333-337 (1953).

[40] Glasstone, S., ELEMENTS OF PHYSICAL CHEMISTRY, D. Van Nostrand Company, Inc., New York, pp. 442-453 (1946).

[41] Rogers, L. B., Record of Chem. Progress 16, 197 (1955).

and the type of anode material. In fact the potential can be controlled by the appropriate choice of an anode. The total emf of the cell is the sum of the potential differences at each electrode with the added effect of concentration. When a magnesium anode is employed and when the electrodes are connected, the concentration of the magnesium ions increases and the concentration of radioactive ion decreases, creating a decrease in the emf of the cell. Taking i to be the current and R the resistance of the cell, and M^{++} the ion to be deposited, the following relation holds as the entire emf is dissipated as iR drop through the cell:

$$iR = E^{o}_{Mg,Mg^{++}} + E^{o}_{M^{++},M} - \frac{0.059}{2} \log_{10} \frac{C_{Mg}}{C_{M}} \quad . \qquad (2)$$

However, in the case of radioactive solutions, the ionic concentration C_M is so small that other ions in solution (in these experiments, hydrogen) carry the burden of the current.

If the potential of the anode is smaller than the potential needed to reduce hydrogen at the cathode, the emf and current will decrease to practically zero. However, if the anode potential is great enough to reduce the hydrogen ion, the decline in emf will be halted by the reduction of the hydrogen ion at the cathode and the final current will be greater than zero.

Because of the many outside factors effected the emf during electrolysis, the emf of the cell was not calculated from theory, but

was determined experimentally by the potentiometer method.[42] Equation (3) was derived and used to obtain the emf of the cell from the experimental data. Resistances R_1 and R_2 are determined experimentally; E_1 is a known standard cell and E_2 is the emf of the unknown cell.

$$E_2 = E_1 \frac{R_2}{R_1} . \qquad (3)$$

The emf of the magnesium cell was measured with electrolytes of 0.10 M and 0.05 M hydrochloric and nitric acids. The data obtained are presented graphically in Fig. 10. The emf begins high, declines rather rapidly, and becomes constant after about one hour. It may also be noted that the emf varies with the concentration, being a little higher in the 0.1 M solutions than in the 0.05 M solutions.

Since the emf was great enough to reduce hydrogen ions, the current would not reach zero for the reason given above. Hence, in highly dilute solutions, in which the negative term of the Nernst equation becomes positive, thus decreasing the critical potential, this generated emf becomes of sufficient magnitude to deposite many radioactive nuclides. This presents a simple method of source preparation.

By using this principle of internal electrolysis, electrodeposition was performed in the electrolytic cell shown in Figs. 7 and 8. A

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[42] Smith, A. W., ELECTRICAL MEASUREMENTS IN THEORY AND APPLICATION, McGraw-Hill Book Company, Inc., New York, pp. 130-133 (1948).

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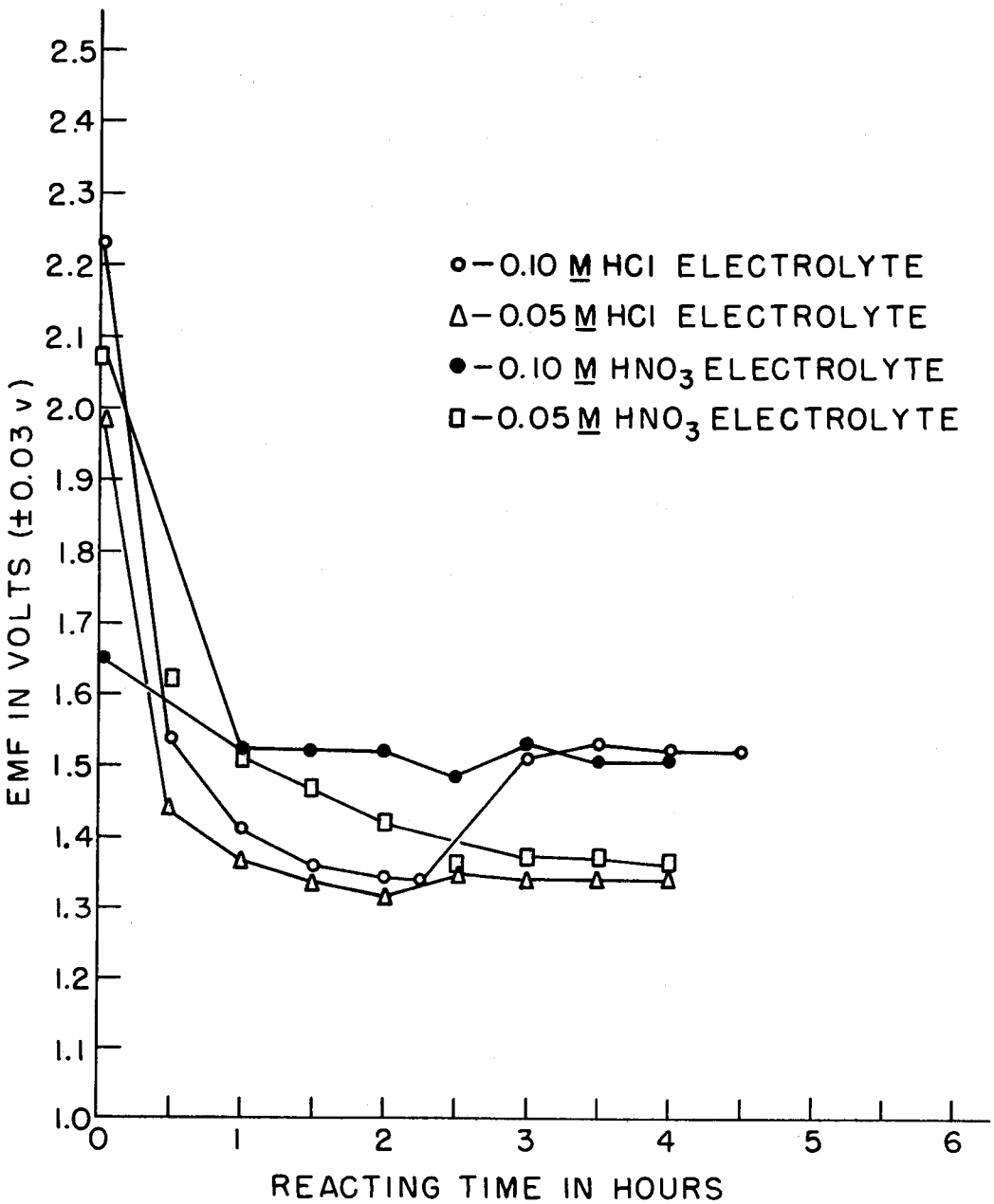

Fig. 10. The EMF of the Magnesium Cell.

magnesium anode inserted at the top was immersed in the electrolyte and connected externally to the lead passing from the gold film cathode at the base. A platinum stirring rod, coated with wax to prevent adsorption, was also inserted through the top and immersed in the electrolyte. The radionuclides deposited by this method are listed in Table VIII. In the beginning the electrolytes were made 0.01 M acid, to reduce the dissolving rate of the magnesium, but the uniformity was so poor (about 30% variation in the uniformity of the autoradiographs) that the acid concentration was increased to 0.10 M which increased the uniformity considerably. In every case the depositions were almost complete and the autoradiographs prepared were highly uniform, the per cent variation being about 5%, as shown by the autoradiographs prepared from sources of indium-114 and gold-198 in Fig. 9. The variation in the uniformity of these two autoradiographs is 5% and 4%, respectively.

Figure 11 compares the two types of electrodeposition. It may be seen that in every case the per cent deposited was generally greater for internal electrolysis than for the electrolysis with an applied emf, although the rate of deposition was not as great in the former method. After two hours of electrodepositing, the electrodeposition by applied emf was practically at its maximum in every case, whereas that of internal electrolysis took approximately three hours and sometimes longer to reach the point of maximum deposition.

Table VIII. Electrodeposition by Internal Electrolyses

Radionuclide	Chemical Form	Electrolyte	Time in Hours	% Deposited
Ruthenium-106	$RuCl_3$	0.01 M - HCl	8.0	95.7
Silver-110	$AgNO^3$	0.01 M - HNO_3	5.0	98.7
Mercury-203	$Hg(NO_3)^2$	0.1 M - HNO_3	8.0	97.3
Cobalt-60	$CoCl_2$	0.1 M - HCl	8.0	90.1
Indium-114	InCl	0.1 M - HCl	3.0	98.6
Iron-59	$FeCl_3$	0.1 M - HCl	6.0	99.2
Chromium-51	$CrCl_3$	0.1 M - HCl	5.0	97.3
Gold-198	$AuCl_3$	0.1 M - HCl	5.5	97.8

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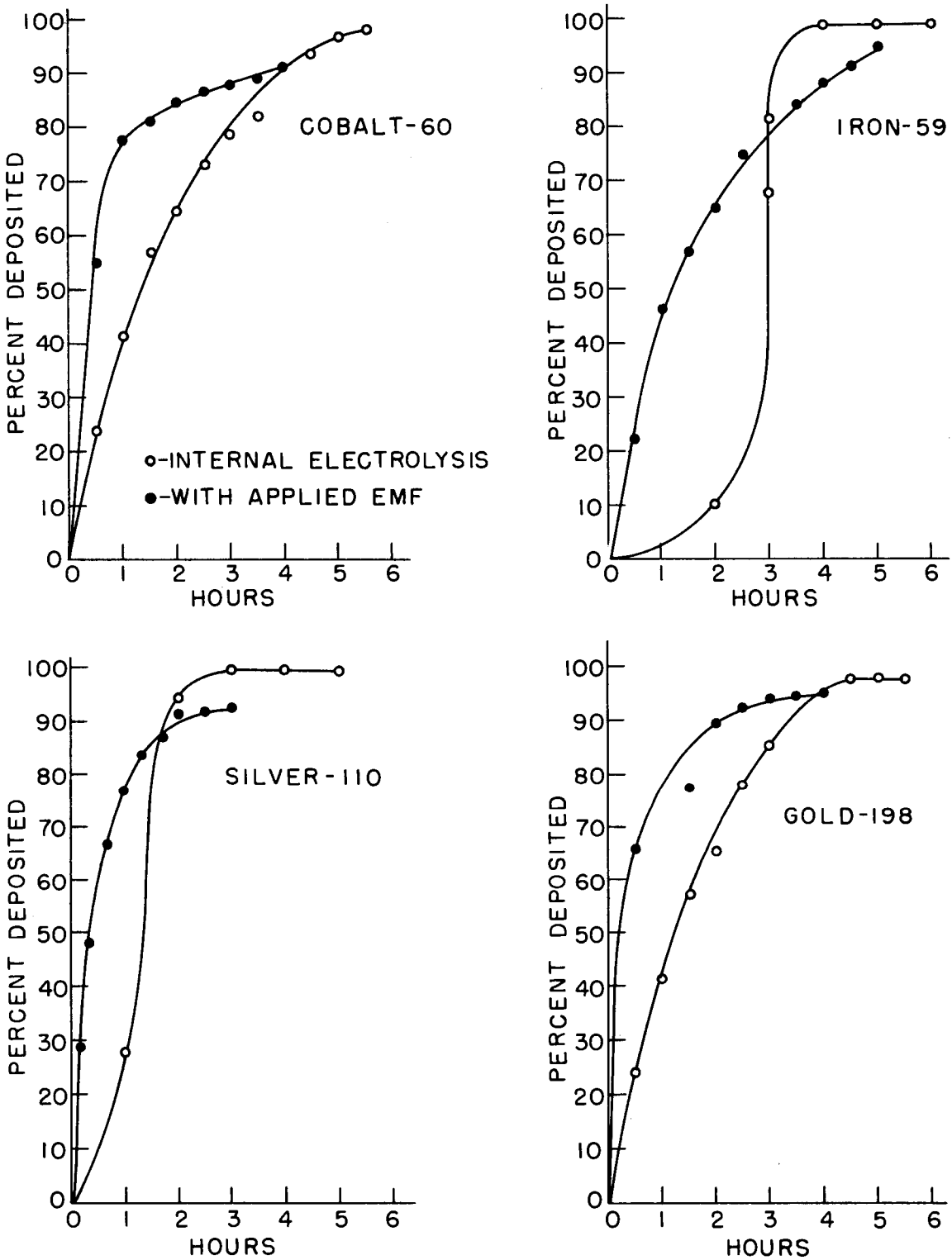

Fig. 11. Applied EMF Compared with Internal Electrolysis.

VIII. The Preparation of A Source for the Beta Spectrometer

Autoradiography provides a useful method of measuring the relative uniformity and activity of a source. However, a more sensitive method consists in placing the source in a beta-spectrometer and noting the change in the resolution of the internal conversion lines as the electron energy decreases. The effect of backscattering from the source can be evaluated because, for thin source mountings, the increase in backscattering is proportional to the increase in thickness of the backing material.

Gold-199 was chosen as the nuclide to be inspected in the beta spectrometer. The gold-199 was produced by irradiating one gram of very pure platinum metal in the Oak Ridge National Laboratory Low Intensity Testing Reactor for a 64-hour period. The nuclear reaction which took place in the reactor was

$$Pt^{198} + n \rightarrow Pt^{199} + \gamma .$$

The platinum-199, having a half-life of 30 minutes, decayed into gold-199. To separate the gold from the platinum, the irradiated metal was dissolved in aqua regia, taken to dryness, and the nitrate destroyed with 6 M hydrochloric acid. The gold-199 was extracted from the 6 M hydrochloric acid solution into ethyl acetate. The ethyl acetate was washed twice with 6 M hydrochloric acid, and then evaporated to dryness. The gold was dissolved in 10 ml of 0.5 M aqua regia. The radioactive gold

solution was analyzed with a gamma spectrometer, and 2% of the activity was found to be gold-198, probably produced from gold-197 that was initially present in the platinum. By making an ethyl acetate extraction on a second platinum sample before irradiation, the gold-198 present after irradiation was reduced to 0.7%. For this second sample, the platinum was dissolved in aqua regia, evaporated to dryness and the nitrate destroyed with hydrochloric acid. The gold was extracted from the hydrochloric acid solution with ethyl acetate and the platinum, remaining in the aqueous phase, was reduced back to the metallic form with magnesium.

From the previous studies, three methods were considered the best available means of source preparation. These were reduction followed by radiocolloidal adsorption, electrodeposition by an applied emf, and internal electrolysis. Because of the solubility of the gold film in the 0.5 M aqua regia solution, the method had to produce a fast rate of deposition. The method which produced the most rapid deposition as well as a high percentage of deposition was electrodeposition by an applied emf. Hence, this method was used.

The electrolytic cell was assembled and 6 ml of the gold-199 was pipetted into the cell. The electrodeposition was continued for 15 minutes at 4 volts and 50 milliamperes per square centimeter. At the conclusion of this time, 20% of the initial activity (1 mc/ml) was deposited on the thin gold film. The short electrodepositing time was a necessity, for the gold film dissolved if immersed in the aqua regia solution for any additional time. At the end of the deposition, the

solution was removed by a pipette, the electrolytic cell dissambled, and the source carefully washed with distilled water. The ring source was still quite transparent after the deposition was complete.

The activity of the gold source was 1.2 mc, measured in a calibrated ionization chamber, and, as stated above, about 0.7% of this activity was due to the presence of gold-198. Hence, the amount of inactive gold-197 present, which determined the density of the source, was calculated and found to be 1.4 micrograms. Since the ring shaped source area was 0.24 cm^2, the source density was calculated to be 6 $\mu g/cm^2$.

IX. The Beta Spectrometer

The beta spectrometer used in this work has been described previously.[43] The spectrometer shown in Figs. 12 and 13 is of the homogeneous magnetic field solenoidal type, and is designed to attain resolutions of about 0.2%. Resolution is defined here as the full momentum width at half maximum intensity, divided by the momentum of the monoenergetic electrons used. The main solenoid coil consists of 639 turns of 1/2 inch copper tubing wound in three layers on a brass cylindrical tank 28 inches in diameter and 10 feet long, and built up to a true cylinder with asbestos cement. Interwound with this tubing is an identical number of turns of No. 12 magnetic wire to serve in stabilizing the magnetic field. Near each end of the solenoid is a correcting coil of 28 turns which makes the magnetic field uniform within 0.25% over a volume which will completely contain all of the electron trajectories. Built horizontally along the supporting wooden frame of the spectrometer are four groups of 18 strands each of No. 12 copper magnet wire passing a current of 1.2 amperes to compensate for the vertical component of the magnetic field of the earth. The solenoid was aligned with the horizontal component of the earth's field.

Four access ports, four inches in diameter, are located near each

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[43] Birkhoff, R. D., Smith, A. W., Hubbell, H. H., and Cheka, J. S., Rev. Sci. Instr. 26, No. 10, 959 (1955).

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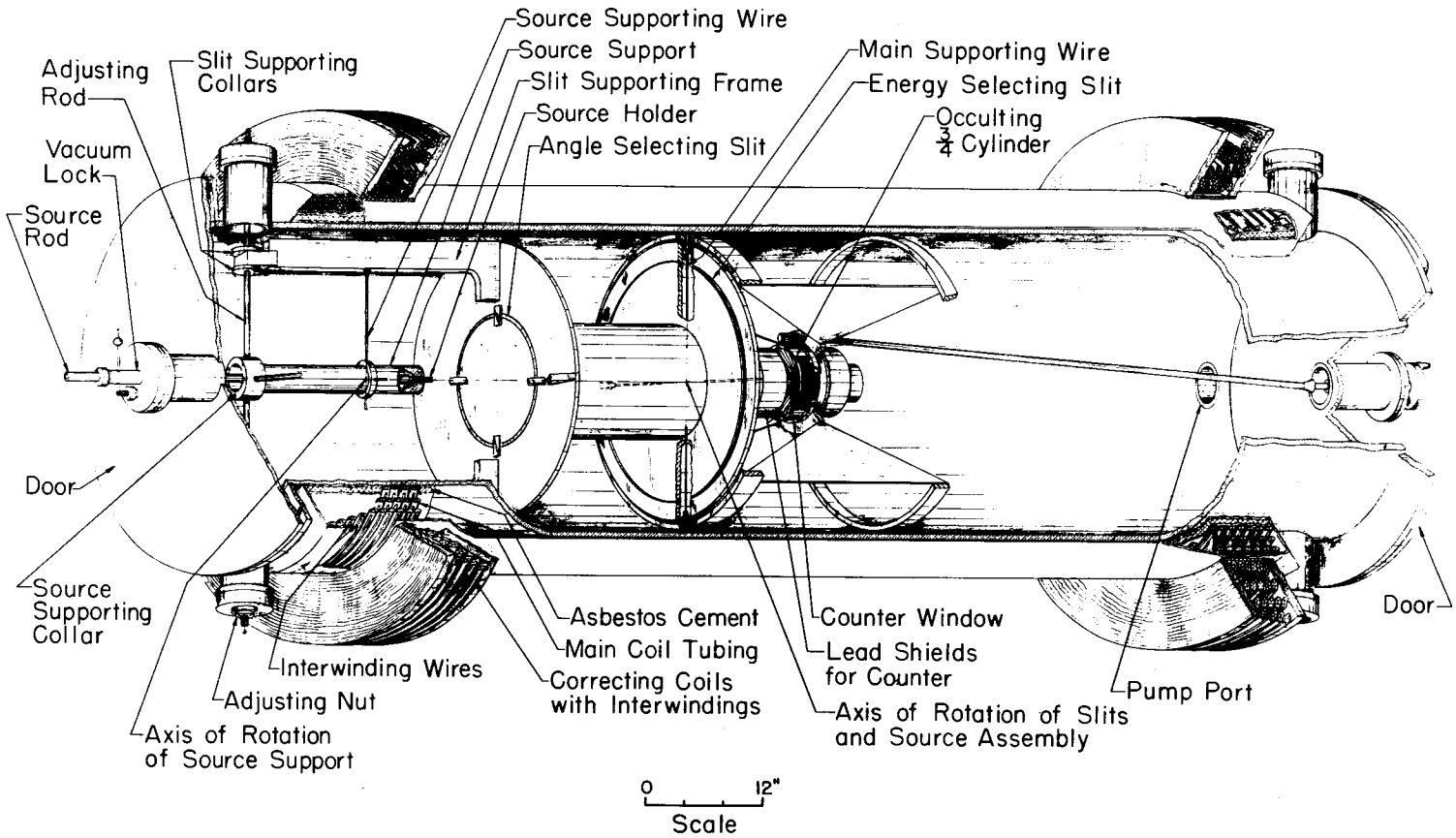

Fig. 12. A Cut-Away View of the Beta Spectrometer.

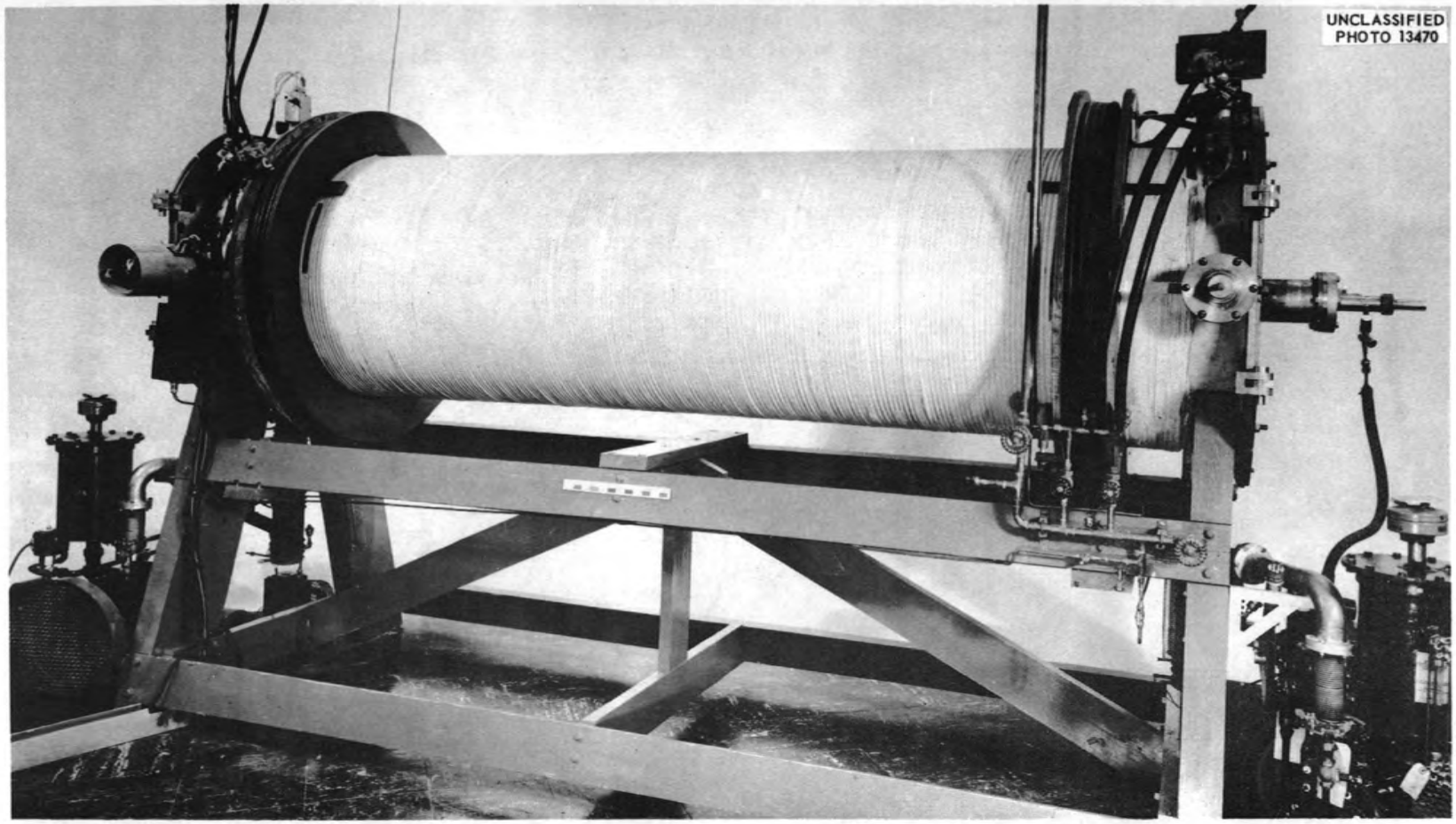

Fig. 13. An External View of the Beta Spectrometer.

end of the tank. They are in a radial position about the tank, spaced 90° apart.

Sources are placed in a thin-walled cylindrical aluminum holder which is screwed to the end of a long source rod which, in turn, is inserted through a vacuum lock in the end of the spectrometer, Fig. 12. The axial adjustment of the source with respect to the slits is made by screwing the source rod through threads in the right end of the source support. The radial adjustments of the source with respect to the slits are made by rotating the adjusting rods and the adjusting nuts together. This screws the rods into the source support collar, thus pivoting the source about the point indicated in Fig. 12. The beta particles leave the ring source and converge on a hole in the end of the source holder. The particles pass through the hole and through two annular slits coaxial with the solenoid. The first slit passes only electrons leaving the hole which have trajectories which make an angle of about 45° with the instrument axis. The second slit accepts only those electrons which have passed the first slit and whose energy is within narrow limits. Each trajectory forms a helix having a diameter of 30 cm; thus the envelope of all helices has a diameter of 60 cm. The trajectories return to the axis of the solenoid and enter a Geiger counter mounted in a cylindrical lead shield which has an annular conical aperture. There is a continual flow of gas through the counter which is controlled by a Cartesian manostat.

The size of aperture in the source holder has a direct bearing upon the transmission and the resolution of the instrument. The narrower

the aperture, the higher the resolution and the lower the transmission. Hence, to determine the best size aperture for which both transmission and resolution are acceptable, the L_1 internal conversion line of the 159 kev gamma of Hg-199 was investigated with source holder aperture diameters of 1.4 mm, 1.0 mm, and 0.6 mm. The results are shown in Fig. 14. The transmission with the three source holder openings were 1,215 counts/minute, 600 counts/minute, and 200 counts/minute, respectively, and the resolution with the three source holder openings were 0.31%, 0.25%, and 0.22%, respectively. The best resolution with respect to good transmission for this source was with the 1.0 mm aperture diameter. The instrument was initially designed for the 0.6 mm diameter of source holder aperture, although, since the internal conversion lines were well defined with the larger opening there was no need to sacrifice the added transmission by using the smaller aperture.

The beta spectrometer was operated entirely from the console shown in Fig. 15. The panel on the left controls and monitors the pressure. The second panel from the left includes the generator voltage, the field voltage, the cooling water temperature, the current monitor, and the generator switch. The third panel contains a Rubicon high precision, Type B potentiometer that adjusts the current through the solenoid. The next panel contains the counter and scaler, the next contains an ammeter that monitors the fast fluctuating correcting current, and the last panel contains the Streeter-Amet Counter.

The current to the solenoid is supplied by a 20 kw motor generator capable of delivering 160 amperes at 125 volts. The current in the

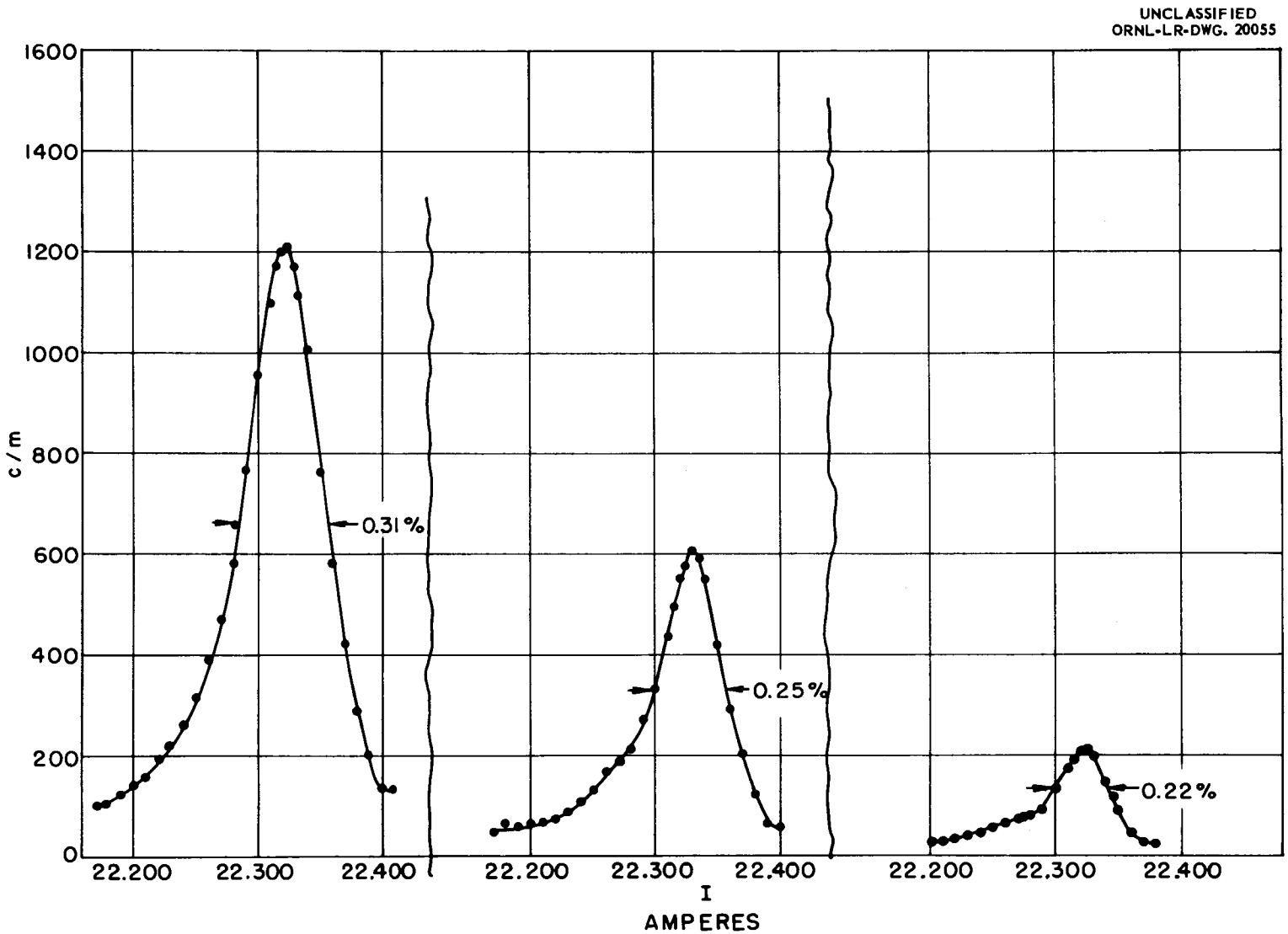

Fig. 14. Transmission and Resolution of the Spectrometer as a Function of Source Holder Aperture.

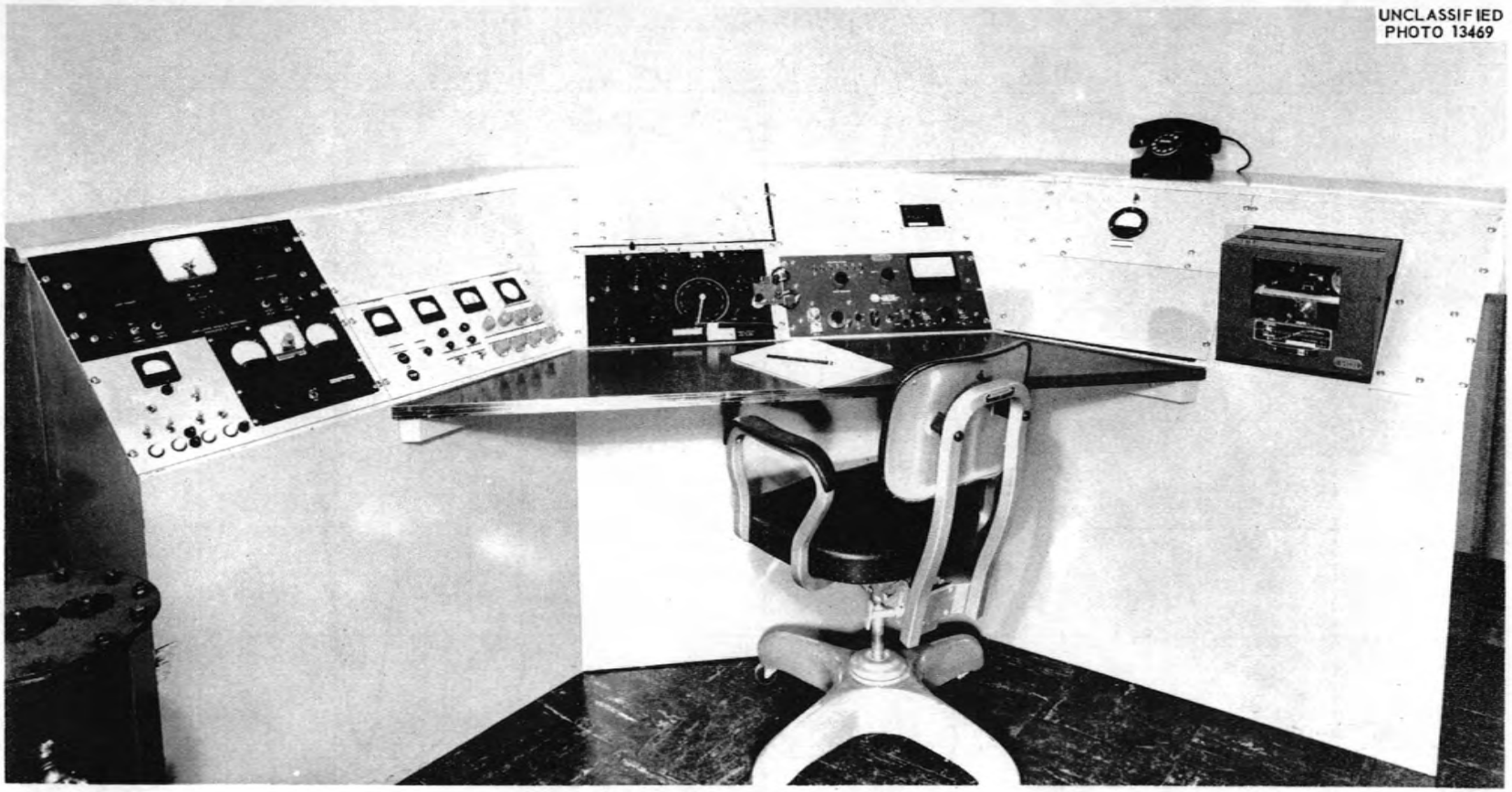

Fig. 15. The Beta Spectrometer Console.

solenoid is held constant by a General Electric induction voltage regulator which is driven by a Brown reversible motor. This system can hold the solenoid current constant within 0.005% at any current from a minimum of about 10 amperes, limited by the residual magnetism of the generator field, to the full output of the generator. Also, to correct for rapid fluctuations in the solenoid current, a special correcting circuit is used.[44]

The calibration of the energy setting of the instrument as a function of current was particularly simple because of the lack of iron in the magnet. If the solenoid current is denoted by i in amperes, and the kinetic energy of the transmitted electrons is T in kev, then

$$T = \sqrt{Ki^2 + (m_o c^2)^2} - m_o c^2 \quad . \tag{4}$$

The constant K was determined experimentally from the well known K conversion line of Au^{198} and found to be 3.34962×10^{-4}.

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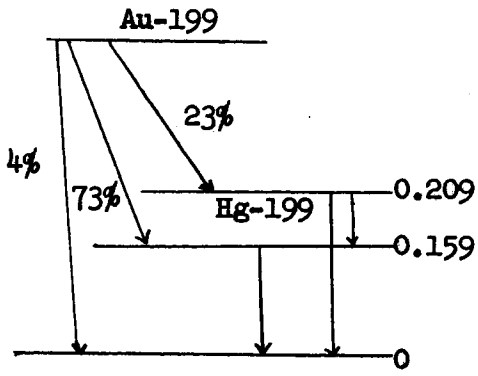
[44] Birkhoff, R. D., Smith, A. W., Hubbell, H. H., and Cheka, J. S., Rev. Sci. Instr. 26, No. 10, 960-961 (1955).

X. Results

Gold-199 decays by the emission of three groups of beta particles to excited states of mercury-199, which in turn decay by three gamma rays to the stable state. The decay scheme is shown in Fig. 16. The three beta energies are 0.460 Mev, 0.297 Mev, and 0.250 Mev.[45]

Fig. 16
The Decay Scheme of Gold-199



The internal conversion lines of the 50 kev, 159 kev, and 209 kev gamma rays were scanned, and all of the peaks that were resolved are shown in Figs. 17, 18, and 19, respectively. The momentum resolutions

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[45] Bell, R. E., Graham, R. L., and Patch, H. E., Can. J. Phys. 30, 35 (1952).

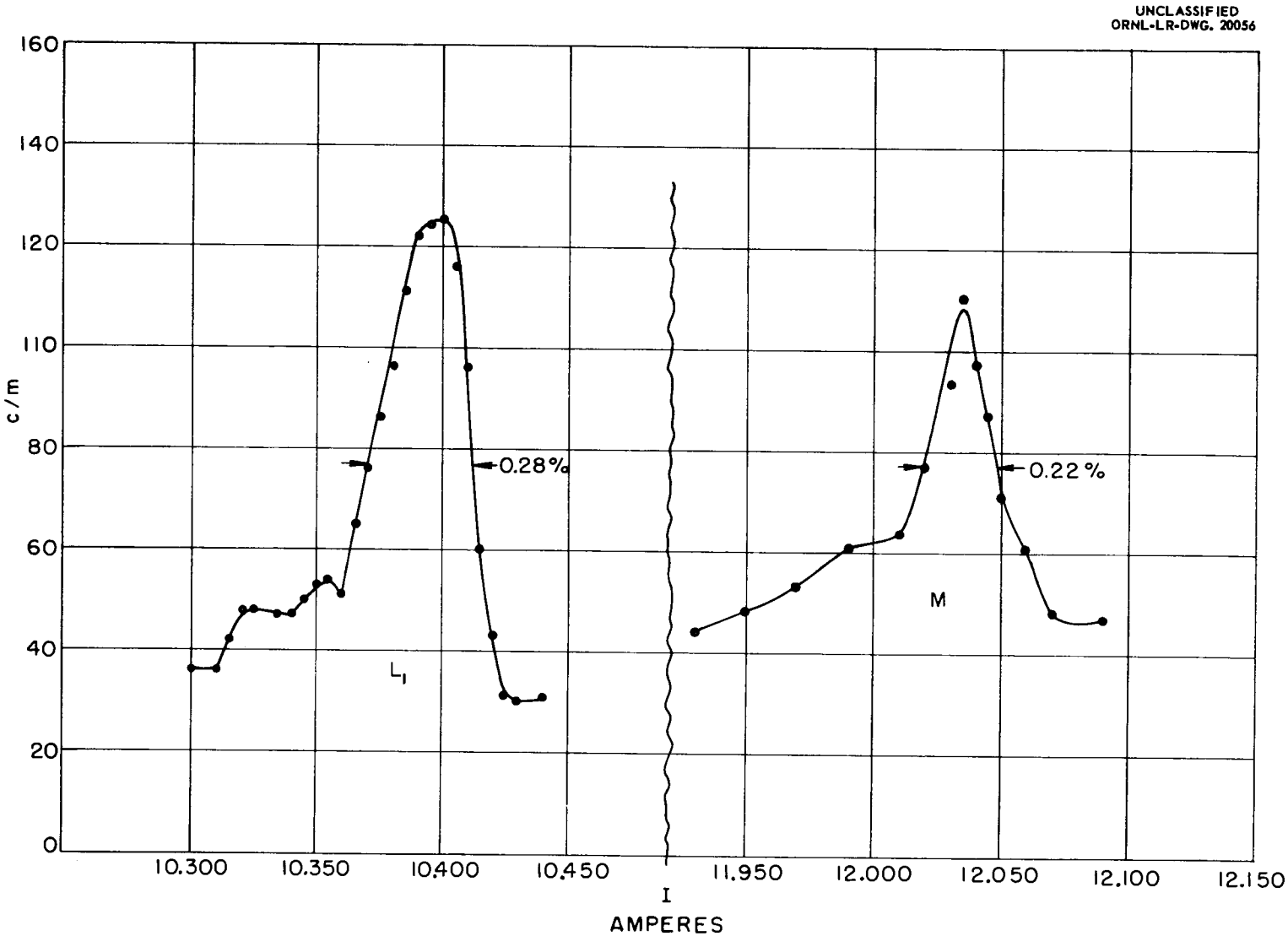

Fig. 17. The Internal Conversion Lines of the 50-Kev Gamma Ray of Mercury-199.

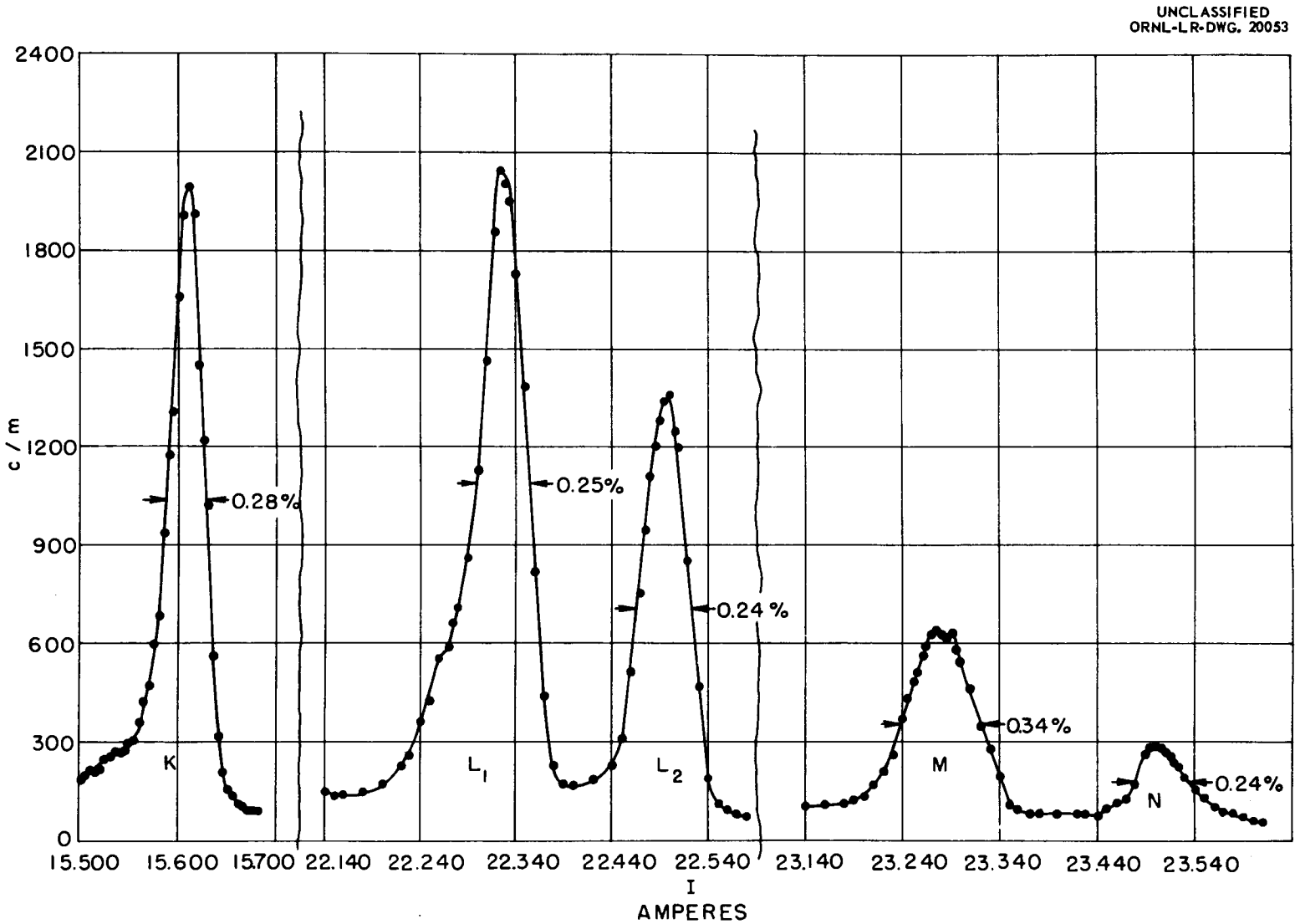

Fig. 18. The Internal Conversion Lines of the 159-Kev Gamma Ray of Mercury-199.

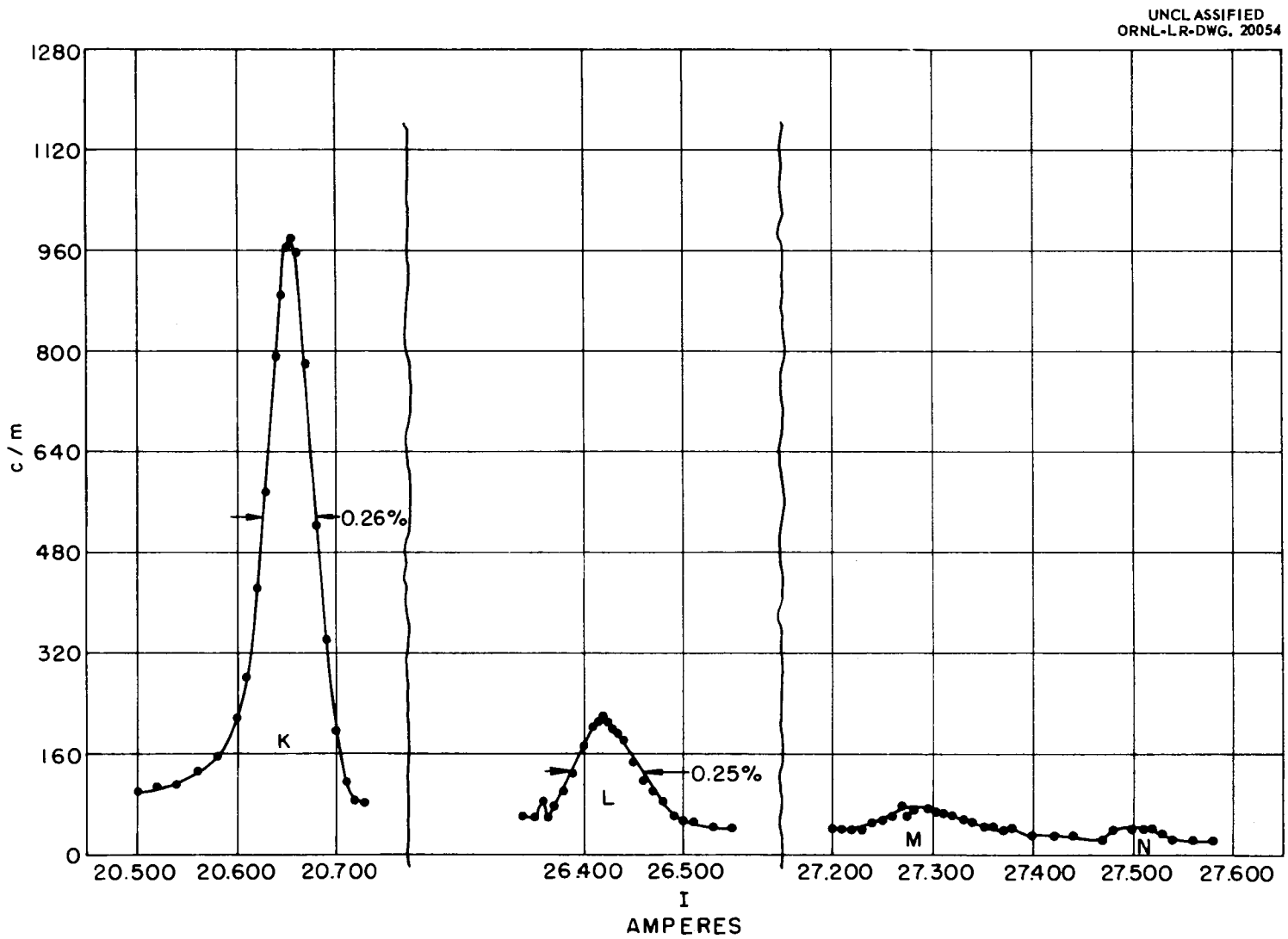

Fig. 19. The Internal Conversion Lines of the 208-Kev Gamma Ray of Mercury-199.

are shown on each peak at one-half maximum intensity, indicated by the arrows. The 159 gamma ray was the most intense, producing well defined conversion lines except for the M line which has a resolution of 0.34%, indicating that several lines were superimposed as one line. Only the L_1 line and the M line of the 50 kev gamma ray were found because it was the least intense gamma ray of the three, and also because the transmission of the spectrometer, due to the counter window, becomes very small at these low energies. The two lumps at the lower energy side of the L_1 line is thought to be within the statistical variation expected at the low counting rates. The momentum resolution was 0.24% and 0.25% for the higher energy conversion lines and decreasing with energy to 0.28% resolution for the lower energy conversion line of about 35 kev.

All of the conversion lines were corrected for transmission, except those of the 50 kev gamma ray (since the transmission was so very low in the low energy region), and the data obtained is listed in Table IX. The binding energy[46,47] was added to each conversion line energy and an average gamma energy obtained. The results agreed quite well with previous values.

The internal conversion line ratios are given in Table X along with previous values. Most of these values checked reasonably well.

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[46] Fine, S. and Hendee, C. F., Nucleonics 13, No. 3, 37 (1955).

[47] Compton, A. H. and Allison, S. K., X-RAYS IN THEORY AND EXPERIMENT, D. Van Nostrand Company, Inc., New York, pp. 789-790 (1935).

Table IX. Conversion Line and Gamma Energies of Hg-199

Con. Line	Con.Line Energy (kev)	γ Energy (kev)	Average γ Energy (kev)	Previous Results	Relative Intensity
L_1	35.1	50.0		50.0[49], 50.6[50]	
M	46.1	49.2	49.6	49.8[51]	
K	74.9	158.0			91
L_1	144.2	159.1		157.5[50]	100
L_{11}	146.5	158.8		158.5[51]	52
M	155.5	158.6		158.5[49]	29
N	157.8	158.1	158.5		9
K	125.6	208.7		208.1[50]	73
L	193.5	208.4		208.3[51]	12
M	204.0	207.1		208.5[49]	2
N	206.8	207.1	207.8		1

[49] Hill, R. D., Phys. Rev. 79, 413 (1950).

[50] de-Shalit, A., Huber, O., and Schneider, H., Helv. Phys. Acta 25, 279 (1952).

[51] Cork, J. M., LeBlanc, J. M., Stumpf, F. B., and Nester, W. H., Phys. Rev. 86, 415 (1952).

Table X. Internal Conversion Line Ratios of Hg-199

γ Energy	Ratio	Values Obtained	Previous Values
159 kev	$K/L_1/L_{11}/M/N$	1.00/1.10/0.57/0.38/0.13	
159 kev	K/L	0.60	0.56[51], 0.37[49], 0.67[50]
159 kev	L/M	4.40	3.60[51]
159 kev	L_{11}/L_1	0.52	0.55[49]
208 kev	K/L/M/N	1.00/0.17/0.06/0.01	
208 kev	K/L	6.05	5.65[49], 5.0[50], 4.5[51]
208 kev	L/M	3.00	

The continuous beta spectrum was corrected for decay and was plotted in Fig. 20 as counts/minute divided by the current, which is proportional to the momentum, versus the current. The spectrum has a double hump, due to the complex beta spectra. The loss of transmission of the spectrometer window at low energies can be noted by the sharp decrease in counting rate at the low energy side of the continuous spectrum which makes it quite difficult to work at energies much below 50 kev.

The minimum source thickness based on the total gold-198 content present (see page 57) was 6 $\mu g/cm^2$. The maximum thickness of the source, assuming that the conversion electron peak width at half-height does not increase more than 0.03%, an amount of the experimental error, between the energies given, is 80 $\mu g/cm^2$, as calculated by the Landau straggling theory.[48]

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[48] Landau, L., J. Phys. U.S.S.R. 8, 201 (1949).

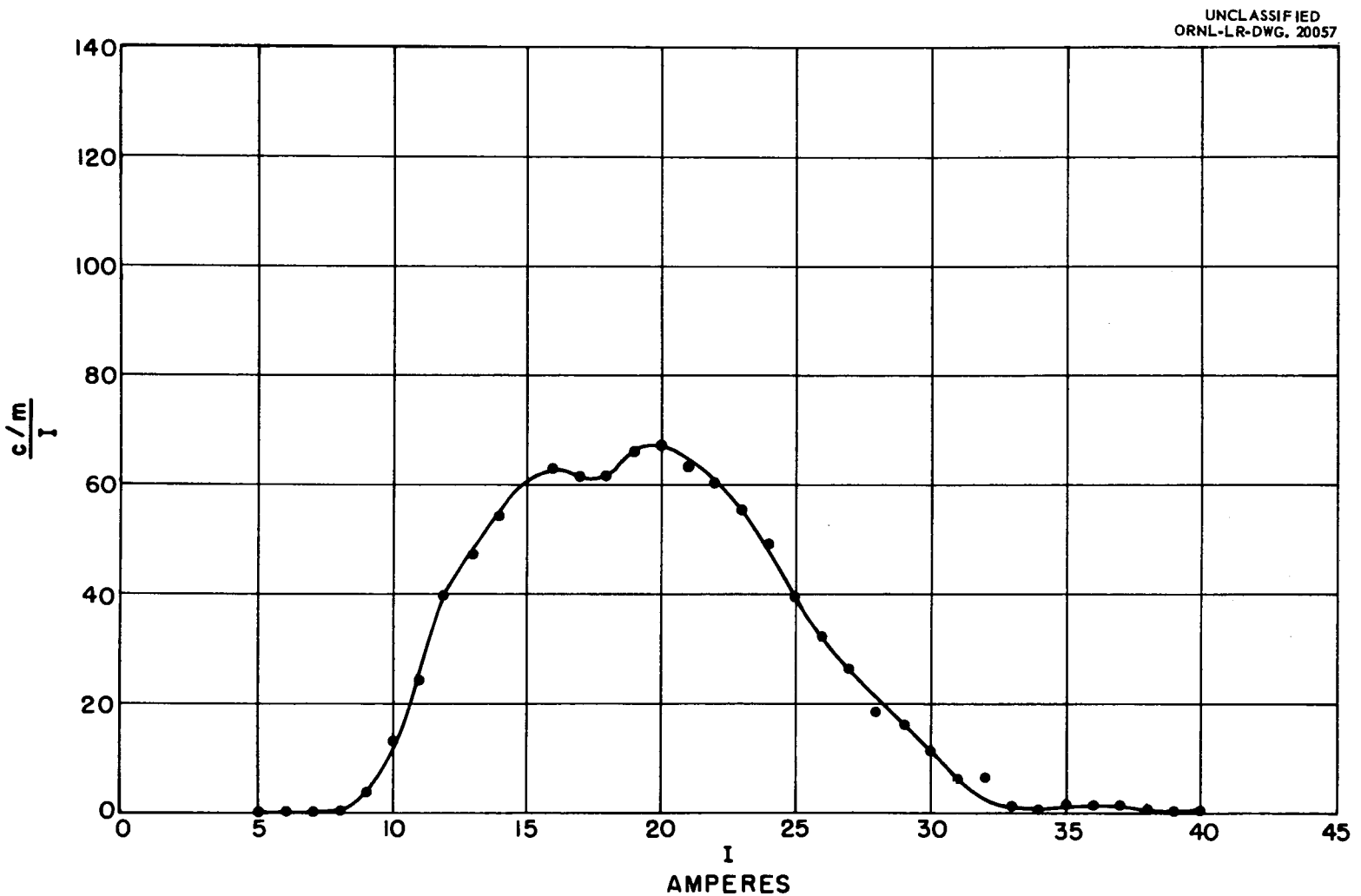

Fig. 20. The Continuous Beta Spectrum of Gold-199.

XI. Summary and Conclusions

Four methods of source preparation were investigated and the quality of the sources produced by each method was determined by autoradiography. Two methods were devised by taking advantage of the colloidal properties of radionuclides. The first radiocolloidal method (the formation of hydroxides and sulfides) produces depositions as high as 82% of the total activity, although, in general, the fractions deposited were much lower. Their uniformity, measured by a densitometer, was 30% to 40%. The second radiocolloidal method (reduction followed by adsorption) produced better results, giving depositions of 90% and greater for many radionuclides and having variations in uniformity of about 1.5%, the highest uniformity obtained. Two methods of electrodeposition were also investigated as possible methods of source preparation. Electrodeposition by an applied emf produced sources having variations in uniformity of 3% to 8% while producing depositions which were nearly complete in almost every case. The sources prepared by internal electrolysis varied in uniformity by about 5% and produced depositions which were from 1% to 8% more complete than the first method of electrodeposition, although about 25% slower. Hence, radiocolloidal adsorption after reduction produced the highest uniformity, but the two methods by electrodeposition were much faster and, in general, produced a higher percentage of deposition. The radiocolloidal method by precipitation should not be omitted from consideration because of the low

percentage of activity deposited, for in many cases the activity deposited would be sufficient, and the uniformity produced would be considerably better than that of solution evaporation.

The two gold-199 sources prepared by electrodeposition with an applied emf and investigated in the beta spectrometer produced internal conversion lines that were symmetrical and well defined. The momentum resolution of the internal conversion lines varied from 0.25% at 194 kev to 0.28% at 35 kev. Since the thickness of the source and source backing becomes much more critical at lower energies, this is an indication that the sources were very thin and uniform. The resolution was increased to 0.22% by using a smaller source holder opening, although, by doing so, the transmission was decreased by about 70%.

The energy of the three gamma rays were calculated by adding the internal conversion line energies and the binding energies, and the values of 49.6 kev, 158.5 kev, and 207.8 kev were obtained. Both these values and the internal conversion line ratios agreed quite well with previous values.

The gold-198 present (0.7% of the total activity), which decays into mercury-198 by beta emission, did not contribute enough activity to affect the beta spectrum. Also, since the conversion electrons of mercury-198 are much more energetic than those of mercury-199, they did not affect the conversion line spectrum.

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