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THE COVER

*The first photograph of functioning genes.* *See page 51*

# 1968
# at the
# Oak Ridge
# National Laboratory

# FOREWORD

This our 25th year has been a good one, even though, because of the general reduction of scientific budgets, ORNL has had to tighten its belt a bit. In this Annual Report we have chosen to present fairly coherent essays on a relatively few accomplishments, rather than trying to cover many activities somewhat superficially. Much that was interesting and important at ORNL during 1968 has therefore had to be left out.

Among the activities that we could not include were most of those concerned with the environment, both those related directly to nuclear energy and those that are aimed more broadly at the environmental impact of technology. If one includes our work on the medical implications of physical insults to the biosphere, as well as our extensive work in ecology, waste disposal, and water purification, then ORNL's concern with the environment approaches eight percent of all that we do.

The environment is only one of the newer problems around which our society is beginning to mobilize. Insofar as such innately diffuse issues have scientific components, it seems reasonable to expect that large scientific institutions such as ORNL will be able to contribute to their resolution. In this we possess an important advantage. When government tackles big, diffuse problems such as the environment, it is hampered by a fragmentation of approach. This is a reflection of the

*Edited by Arthur H. Snell and assembled with the assistance of the Graphic Arts, the Photography, and the Technical Publications Departments of the ORNL Technical Information Division.*

inevitable fragmentation of government bureaucracy: each office or bureau or agency usually has responsibility for only a piece of the problem. A place like ORNL, with its interdisciplinary style, can reintegrate the separate fragments *at the working level.* I would therefore expect that in future annual reports we shall see ORNL increasingly involved with great sociotechnological problems on which our interdisciplinary, reintegrative power can be brought to bear.

But the perceptible shift in public concern toward this new class of problems and even, perhaps, a growing sense of disenchantment with science among many of our articulate younger people must not becloud our continuing main purpose: to master the science and technology of nuclear processes for the betterment of man. During 1968 the Laboratory has made important progress both in fission and in fusion; in exploration of the heavy elements; in radioisotope applications, particularly to medicine; in applications of cheap nuclear energy to heavy, energy-intensive chemical processes; and, supporting all this, in basic research in most of the physical and biological sciences. We expect ORNL-developed science and technology – intellectually challenging and relevant and humane – to continue to contribute importantly to our society's progress and well-being.

Alvin M. Weinberg

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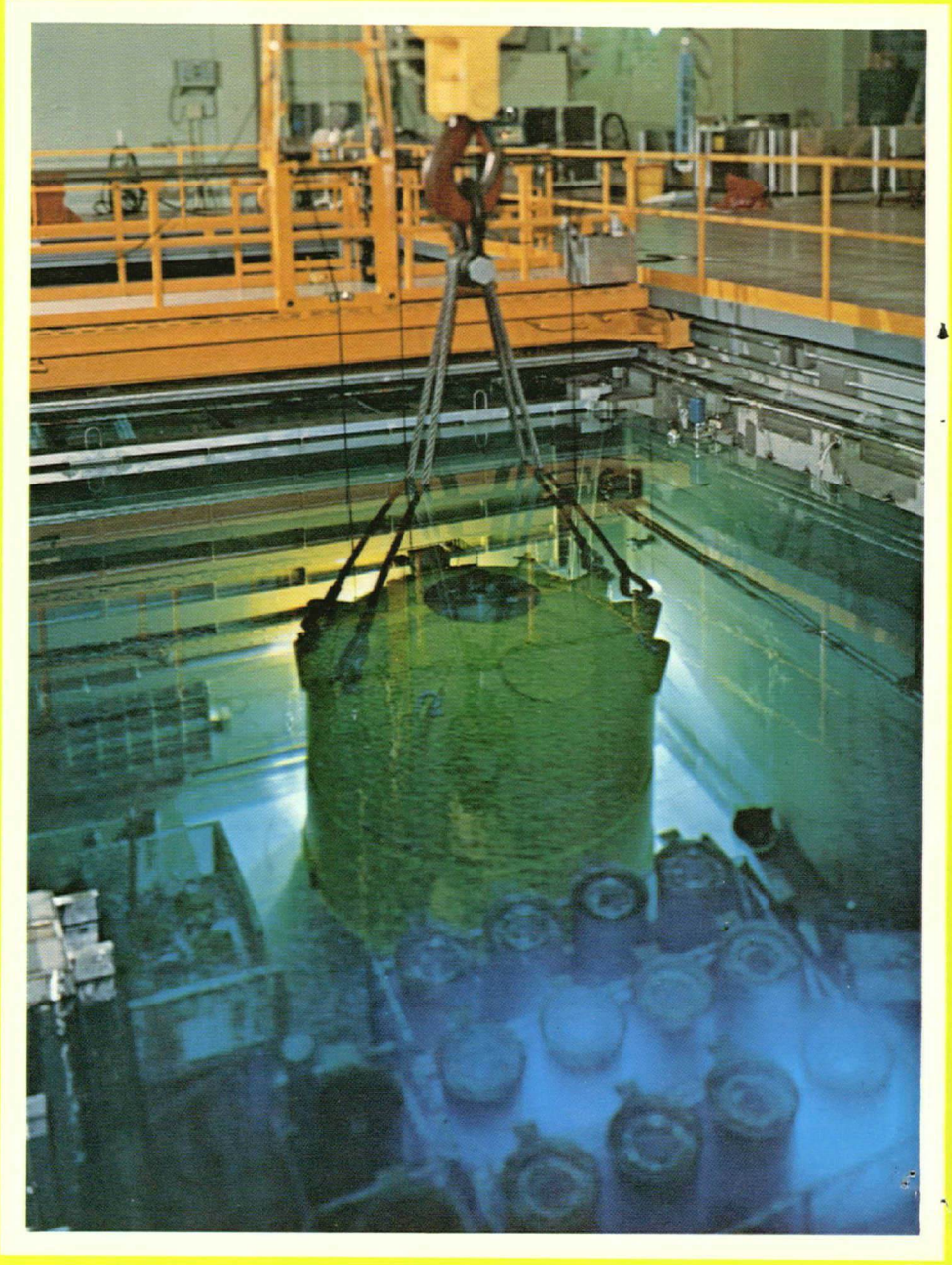
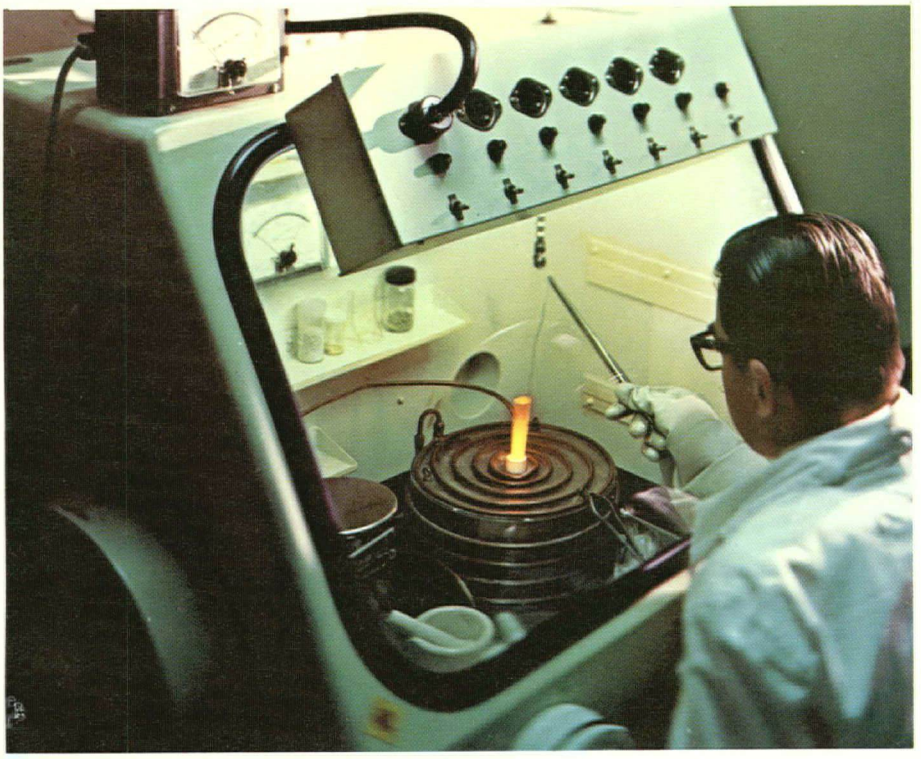
# Physical Sciences



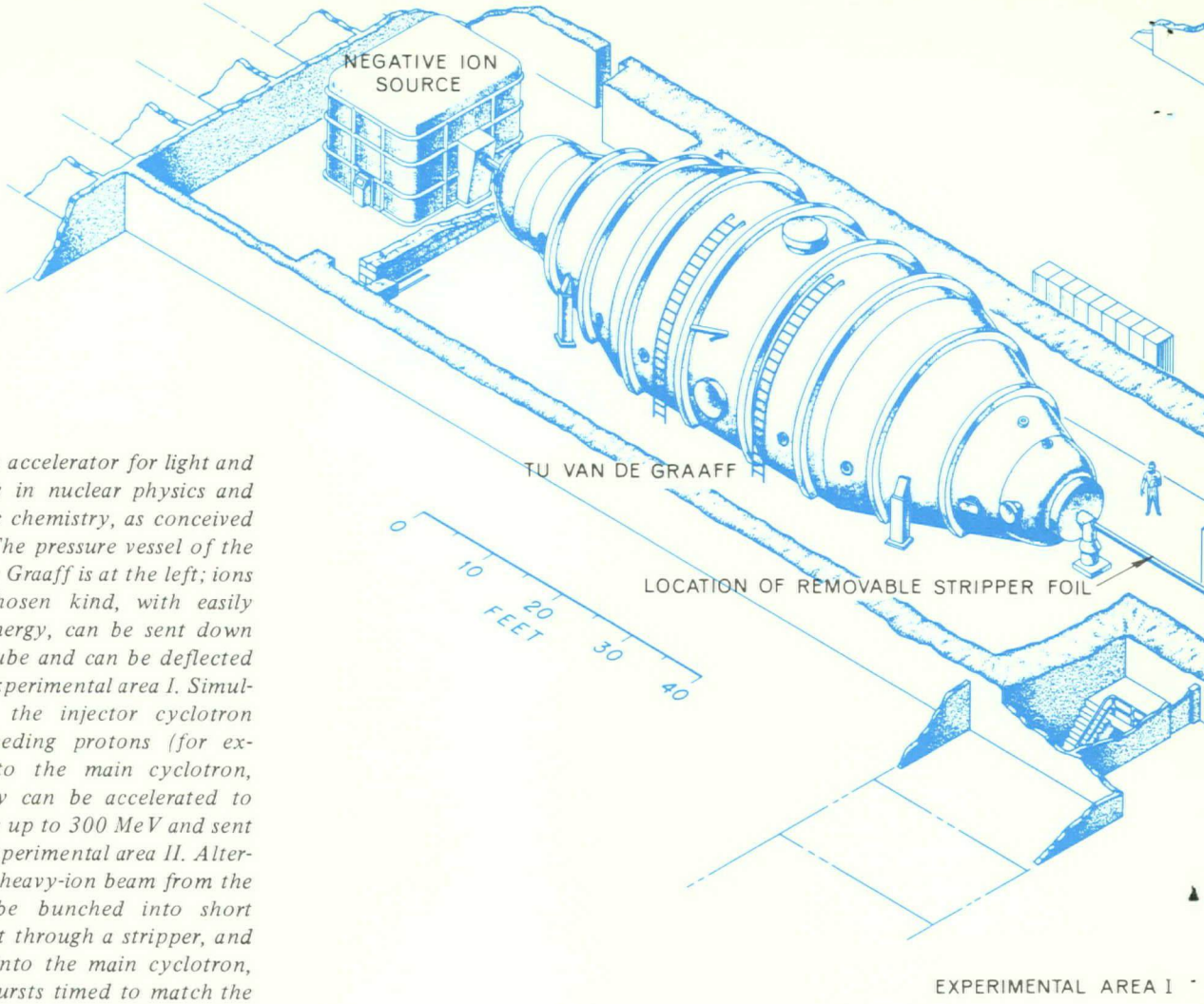
*Transfer vessel (center) and spent fuel elements (lower right) in the swimming pool of the High Flux Isotope Reactor.*

# The Transuranic Elements ..... Research

Laboratory work on the transuranic elements in 1968 made strong progress both in research concepts and in actual production. Contributions were made to the active, worldwide theoretical discussion about the possible existence of nuclear stability a jump or two beyond the heaviest elements now known, that is, at atomic numbers 114 and 126, as compared with 103 for the heaviest element up to 1968 – lawrencium. Maybe nature has never been able to make these elements in the processes of stellar evolution because of the intervening group of unstable elements, but if man could make them by bombarding heavy nuclei with other heavy nuclei, we could see if the theoreticians are right and land in a brand-new region of great interest – a chemist's leap into the beyond, conceptually akin to the leap to the moon.



*Single crystals containing transuranium isotopes are grown in special contained furnaces. These crystals are used for spectroscopic and magnetic studies.*

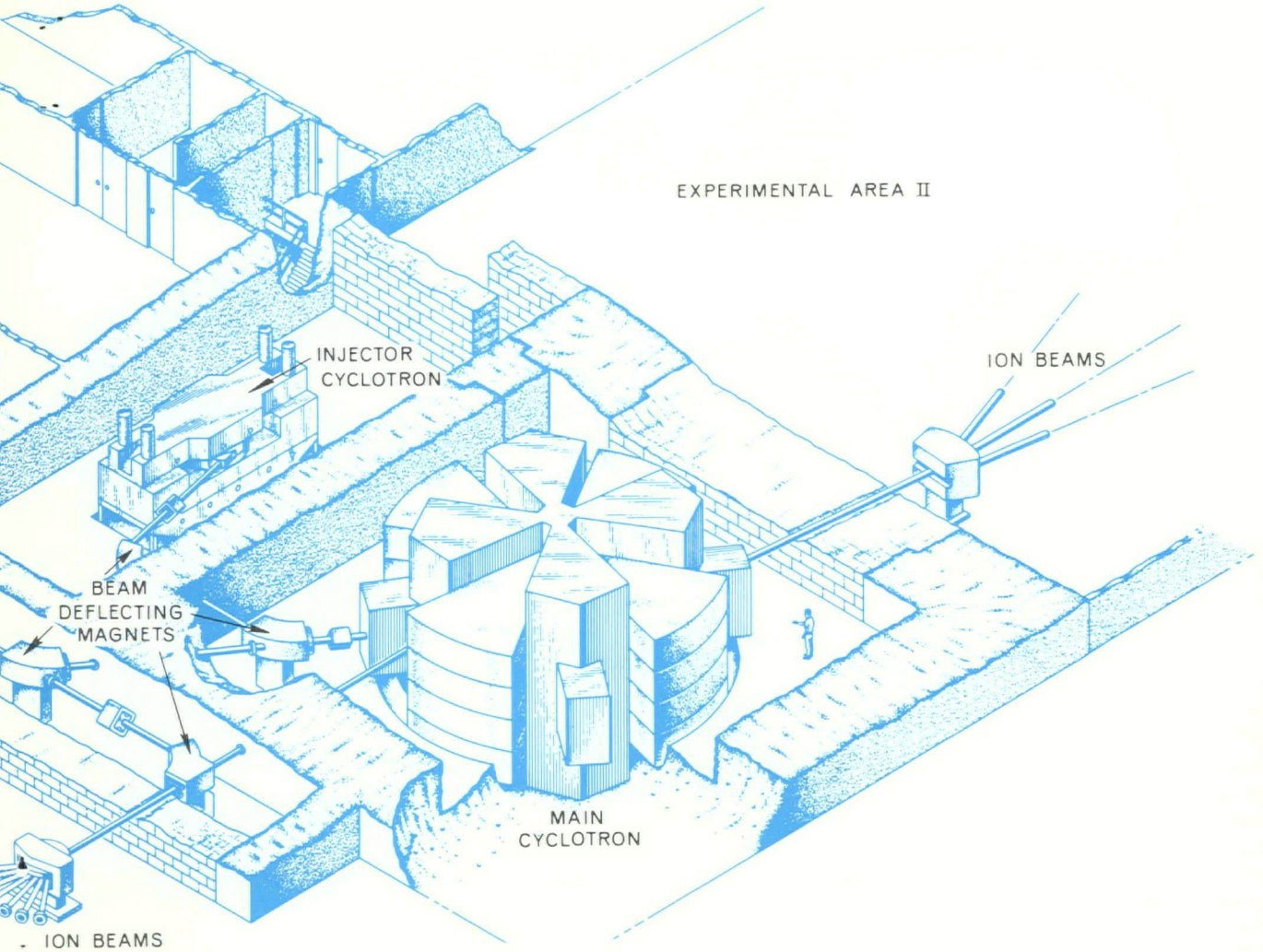



*The dream accelerator for light and heavy ions in nuclear physics and transuranic chemistry, as conceived in 1968. The pressure vessel of the TU Van de Graaff is at the left; ions of any chosen kind, with easily variable energy, can be sent down its beam tube and can be deflected into the experimental area I. Simultaneously, the injector cyclotron can be feeding protons (for example) into the main cyclotron, where they can be accelerated to any energy up to 300 MeV and sent into the experimental area II. Alternatively, a heavy-ion beam from the TU can be bunched into short bursts, sent through a stripper, and deflected into the main cyclotron, with the bursts timed to match the radio frequency of the cyclotron. After further acceleration, they can then be sent into either of the two experimental areas. Under this mode of operation, the injector cyclotron is free for measurements in activation analysis.*

## The Heavy-Ion Accelerator Concept

The major development in transuranic element research at ORNL was the conception of an accelerator capable in principle of synthesizing these superheavy elements. High energies are required, because both the projectile and the target nuclei are highly positively charged, and one must overcome the consequent Coulomb repulsion in order to get the nuclei to interact. The required energy is estimated at 7.5 MeV per atomic mass unit of the projectile (assuming the target is of about equal weight), and this estimate includes an effect due to distortion of the two nuclei as they approach one another. In a head-on collision, they will distort into shapes like the halves of a hamburger bun, and Laboratory theoreticians have found that if you don't take the distortion into account, your accelerator will be too small.

These energies can be reached in apparatus of reasonable size only if you strip several electrons from the atoms under acceleration; that is, you use multiply charged ions. Staff members of our Physics, Chemistry, and Solid State Divisions have had experience in the generation and handling of beams of multiply charged ions of bromine and iodine in connection with their work on channeling in crystals (see a later section of this report), using the tandem Van de Graaff. Their experience and knowledge, combined with familiarity with both Van de Graaffs and




cyclotrons, led to the invention of an accelerator concept that would be capable of accelerating ions of lead and uranium sufficiently for nuclear interaction with targets of equally heavy elements. In fact, as we shall see, it goes far in fulfilling the accelerator needs of the Laboratory for a long time to come.

To see how the accelerator complex works, let's start with the TU tandem. This would be one of the largest tandem Van de Graaff accelerators built; the pressure tank is 80 ft long and 25 ft in diameter, and the insulated terminal rides at steady, controllable positive voltages as high as 16 MV (guaranteed) to 20 MV (hoped for). Sulfur hexafluoride gas at a pressure of 100 psi prevents this enormous voltage from sparking to the tank. Evacuated, insulating acceleration tubes extend from the central terminal to either end. In conventional tandem usage, the source of negative ions ($H^-$, $Br^-$, $I^-$, for example) is turned on, and the ions are pulled to the central terminal through the first tube. Inside the terminal they pass through a thin foil (a "stripper") where they lose at least two electrons, thereby becoming positive ions; they then enter the second tube and are again accelerated, this time toward ground potential, and they emerge from the TU with energies corresponding to two to ten times the voltage on the central terminal depending on their charge.

For elements that don't give large yields of negative ions in an ion source, the alternative exists of placing a positive ion source inside the terminal. Let's take the extreme example and follow the acceleration of uranium. A uranium source will put out a beam of mixed charges, from $U^{+}$ to perhaps $U^{12+}$ or higher. Of these, $U^{9+}$ gives a good combination of high charge state with sufficient yield, so the $U^{9+}$ component will be selected and sent down the accelerator tube. On reaching ground, the ions will have an energy of $9 \times 16 = 144$ MeV. They then pass through a stripper, and they are going so fast that many more electrons are peeled from the ions; in fact, about 15% of them will end up in the $U^{36+}$ state. The $U^{36+}$ component is selected by a deflecting magnet and sent through an ion-optical transport system, to be injected into a separated-sector cyclotron. This is a cyclotron about 36 ft in diameter (1500 tons) with a hollow center and wide gaps between its four magnet sections. The cyclotron further accelerates the $U^{36+}$ ions to 1600 MeV, which is about 7.5 MeV per atomic mass unit, and this is sufficient for nuclear interaction with any kind of target nucleus.

Clearly the TU tandem need not always be used as the cyclotron injector; it can accelerate light and medium-weight ions for nuclear structure studies of the kinds that require ion beams of the quality historically associated with Van de Graaff accelerators. As such, it will fulfill all the functions covered in a proposal that the Laboratory submitted to the Atomic Energy Commission in 1967. Also, the cyclotron need not be confined to acceleration of heavy ions. If a small, relatively inexpensive injector cyclotron is provided, as indicated in the sketch, protons, deuterons, and helium ions can be fed into the large cyclotron, and it will accelerate them to 280 MeV for the protons and equivalently high energies for the deuterons and helium ions. Of special interest is the acceleration of $^{3}He^{2+}$ ions by the small cyclotron alone, because the Analytical Chemistry Division can use them for activation analysis of several light elements. There would be time available for such use whenever the big machines are accelerating heavy ions.

In summary, the accelerator complex would fulfill the Chemistry Division's need for heavy ions for transuranic synthesis, the Physics Division's need for a modern Van de Graaff accelerator, the Electronuclear Division's need for protons and light ions several times as energetic as those provided by the Oak Ridge Isochronous Cyclotron (ORIC), and the Analytical Chemistry Division's need for activation analysis for light elements. In combination with other ORNL facilities such as the Transuranium Laboratory, the High Flux Isotope Reactor, the Transuranium Processing Plant, and the electromagnetic isotope separators, it would enable Oak Ridge to make a wide, massive attack on the whole problem of the transuranics with facilities not likely to be duplicated elsewhere.

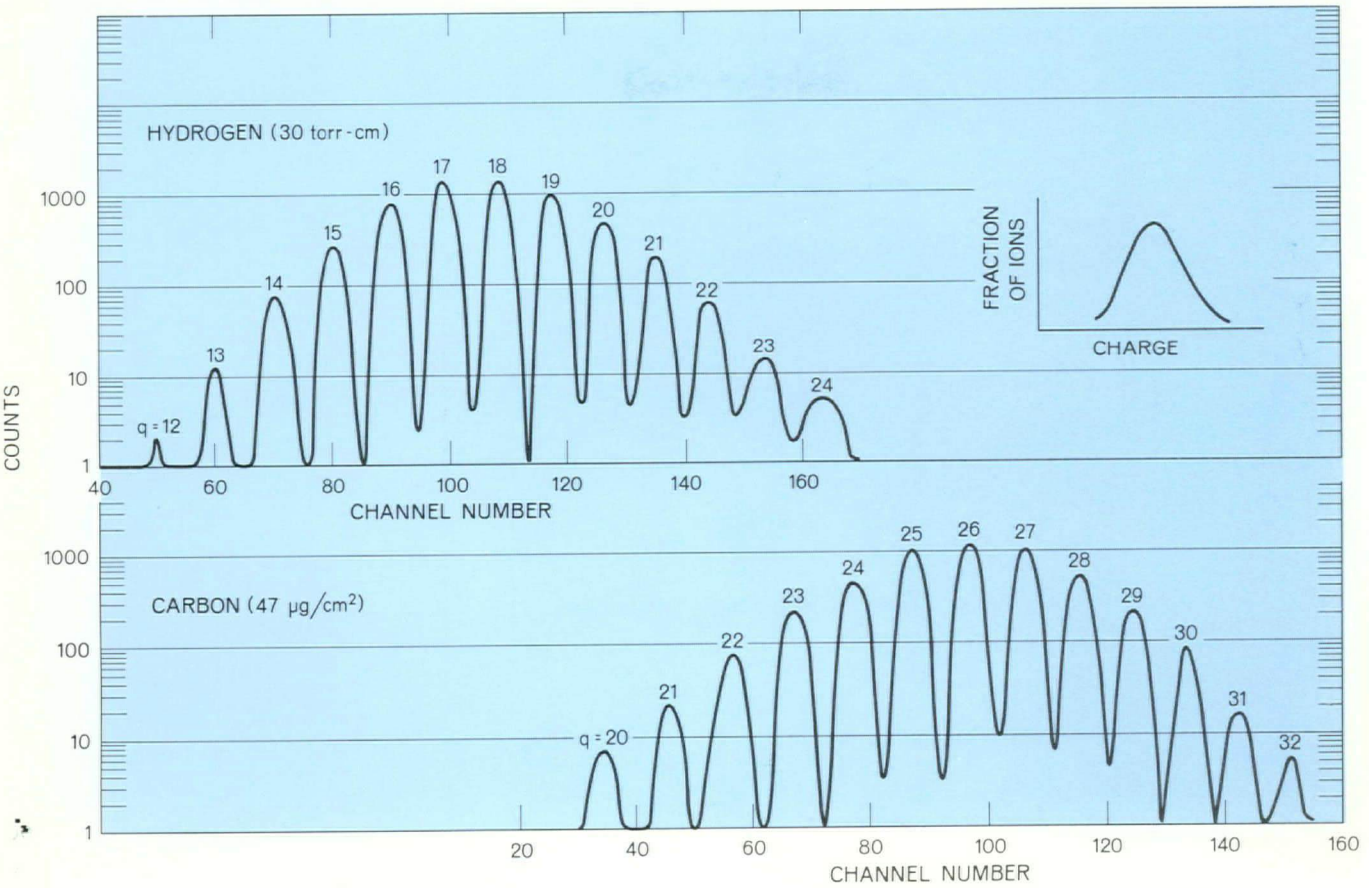
## Beams of Multiply Charged Ions

How can we say so confidently, as we did in the foregoing, that the multiply charged ions can be produced and that the design of the

elaborate and expensive accelerator complex lies on firm ground? The whole injector-stripper-booster concept rests on two considerations: (*a*) that ions in high charge states can actually be produced and (*b*) that they can be produced without an intolerable loss of beam.

Experiments in the Van de Graaff Laboratory have provided the answer. The charge spectra of beams of ions as heavy as iodine have been measured, and a good "feel" for the subject has been established. The sample charge spectra shown here were taken using the small tandem Van de Graaff accelerator. Iodine ions were accelerated to 110 MeV and then shot through hydrogen gas (upper curve) or a very thin carbon foil (lower curve). After that they were sent through a magnetic field, where those in the various charge states were separated for measurement. The hydrogen gas stripper produced ions of charge 18 most abundantly, while the carbon foil produced charge 26 most abundantly. Furthermore, as the inset shows, the charge spectrum is rather concentrated; its full width at half maximum is about six charge states. In the contemplated accelerator, therefore, a beam of ions in a single charge state will be spread into a number of charge states when it passes through a stripper, but if you select the most abundant one for injection into the cyclotron, you will still have about one-sixth of your original beam. This is sufficient to make a very practical acceleration system.

*Actual experimental curves for charge states observed for 110-MeV $^{127}I$ ions passing through hydrogen gas and a carbon foil. Solid strippers clearly produce higher charge states, which in turn produce higher ion energies in a tandem accelerator.*



## Carbon and Oxygen Beams in the ORIC

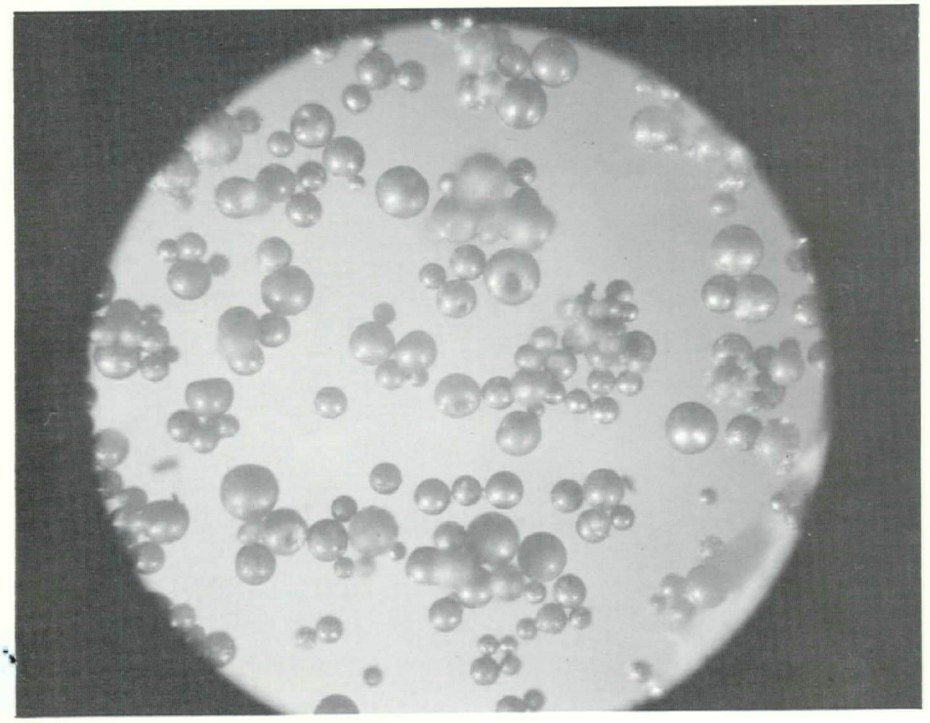
While the men in the Van de Graaff Laboratory were studying the multiply charged ion beams, the men in the Cyclotron Laboratory were actually putting them to use. The ORIC is capable of accelerating ions to interesting energies if the ions have a charge-to-mass ratio of $^1/_3$ or $^1/_4$. With the ion sources that have been developed for use within the cyclotron, this limits the operation to rather light elements, for example, $C^{4+}$ (charge 4, mass 12), $N^{4+}$ (charge 4, mass 14), $O^{4+}$ (charge 4, mass 16), and to some extent $Ne^{5+}$ (charge 5, mass 20). By modifications to the ion source, mainly in the direction of forcing more power through the arc discharge within the source, beam currents of these ions were increased from the nanoampere level to the microampere level. The ions have an energy of 118 MeV for $C^{4+}$, 103 MeV for $N^{4+}$, 90 MeV for $O^{4+}$, and 112 MeV for $Ne^{5+}$. They were immediately put to research use by the chemists engaged in transuranic experimentation.

*Swan Lake and the cyclotron building.*

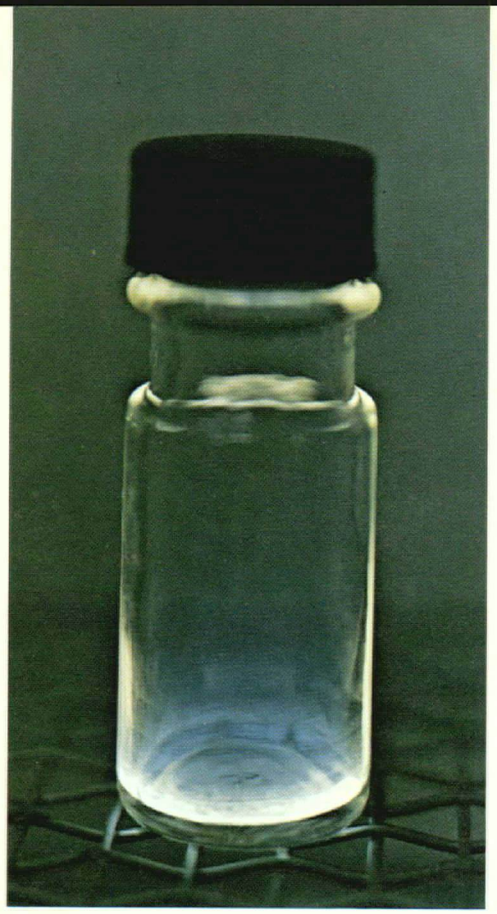
# The Transuranic Elements .....Production

## The Production of Transuranic Elements

The High Flux Isotope Reactor (HFIR) and the Transuranium Processing Plant (TRU) have been in full-scale operation for over two years. Both facilities have performed extremely well. The HFIR has been on stream at full power more than 95% of the time during the last six months, and uninterrupted operation in the TRU during processing campaigns has made it possible to isolate substantially larger fractions of short-lived nuclides before they decay. Within the last year, approximately 100 shipments of TRU products have been made, including various isotopes of americium, curium, berkelium, californium, einsteinium, and fermium. As in the past, most of this material was sent to research scientists at AEC laboratories throughout the United States. Included in these shipments were about 1 mg of berkelium-249, 4 mg of californium-252,



*Actinide oxides in the form of pressed pellets of aluminum powder and the oxide are loaded into HFIR targets. Oxide particles of the proper size are required to produce pressed pellets with a continuous aluminum phase having high thermal conductivity. This photomicrograph shows 40- to 177-μ-diam $AmO_2$-$CmO_2$ microspheres which are used in HFIR targets. The microspheres were made behind concrete shielding by the sol-gel method.*



*Contained in this ½-oz bottle is 26 μg of einsteinium-253. This quantity is so small that only a small stain can be seen when viewed in ordinary light. This photograph was taken with the radiation of the einsteinium as the light source and required a 10-min exposure with sensitive film. The einsteinium is all at the very bottom of the bottle, but light transmitted through the glass also illuminates the sides of the bottle.*

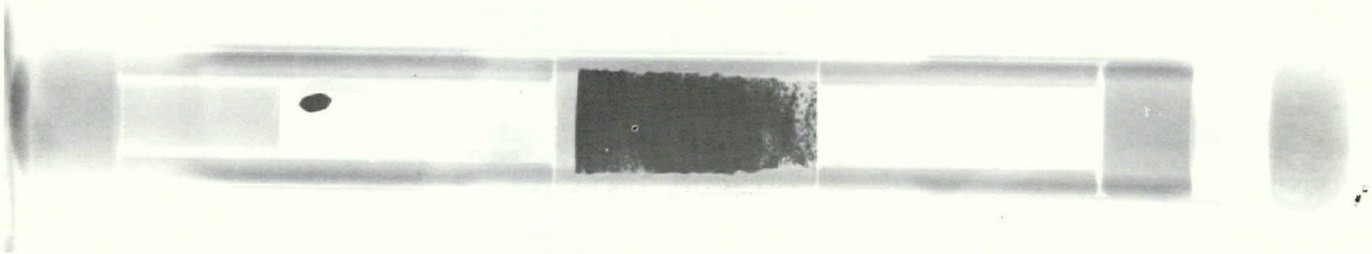
42 μg of einsteinium-253, and $7 \times 10^7$ atoms of fermium-257. Although fermium-257 has been detected previously in irradiated material from nuclear reactors, this very tiny amount (~0.00000003 μg) represents the first significant quantity produced by this method. Knowledge of its nuclear properties is of great importance, since no heavier nuclides have yet been detected in any nuclear reactor irradiations.

The main-line effort for producing transplutonium elements consists in fabricating, irradiating, and processing HFIR target rods containing plutonium-242 or americium and curium. The original target rods irradiated in the HFIR contained plutonium-242, but recently rods containing americium and curium were fabricated and are now being irradiated. This is an important step toward increased production of the transplutonium elements, since fewer neutrons and hence shorter irradiation times are required to produce the heavier nuclides. Curium recycled from irradiated plutonium-242 targets is especially valuable, since it contains significant quantities of the heavier isotopes: curium-246, -247, and -248. Gram quantities of americium and curium must be handled behind protective shielding; therefore procedures which can be used in cells are required. Sol-gel techniques developed as part of the nuclear fuel cycle program were modified and used to make controlled sizes of mixed americium-curium oxide particles. These oxides were mixed with aluminum powder, pressed into pellets, and fabricated into HFIR target rods in equipment contained in three of the TRU cells. Eight rods have been fabricated, of which six are now being irradiated in the HFIR.

Facilities in the HFIR-TRU complex are frequently used for special projects with heavier transplutonium elements. Several capsules of californium-252 have been prepared for use as neutron sources; special separations are made periodically to isolate isotopically pure einsteinium-253 from aged californium product and to isolate californium-249 from aged berkelium; and a number of small targets containing

*Samples for special HFIR irradiations are fabricated as oxides dispersed in pressed aluminum pellets which are then welded into aluminum "rabbits" about ⅜ in. in diameter by 2.5 in. long. This is an enlarged x-ray photograph of a "rabbit" containing 800 μg of californium-252.*

*Intricate operations with this minute amount of californium, which must be kept behind several feet of concrete, are difficult. In order to simplify handling, the californium was prepared for irradiation by adding californium nitrate solution to 1 g of cerium hydroxide sol, which was formed into microspheres and then calcined to the oxide. The mixed oxide microspheres were added to aluminum powder and pressed into the pellet shown in the center of the x ray.*



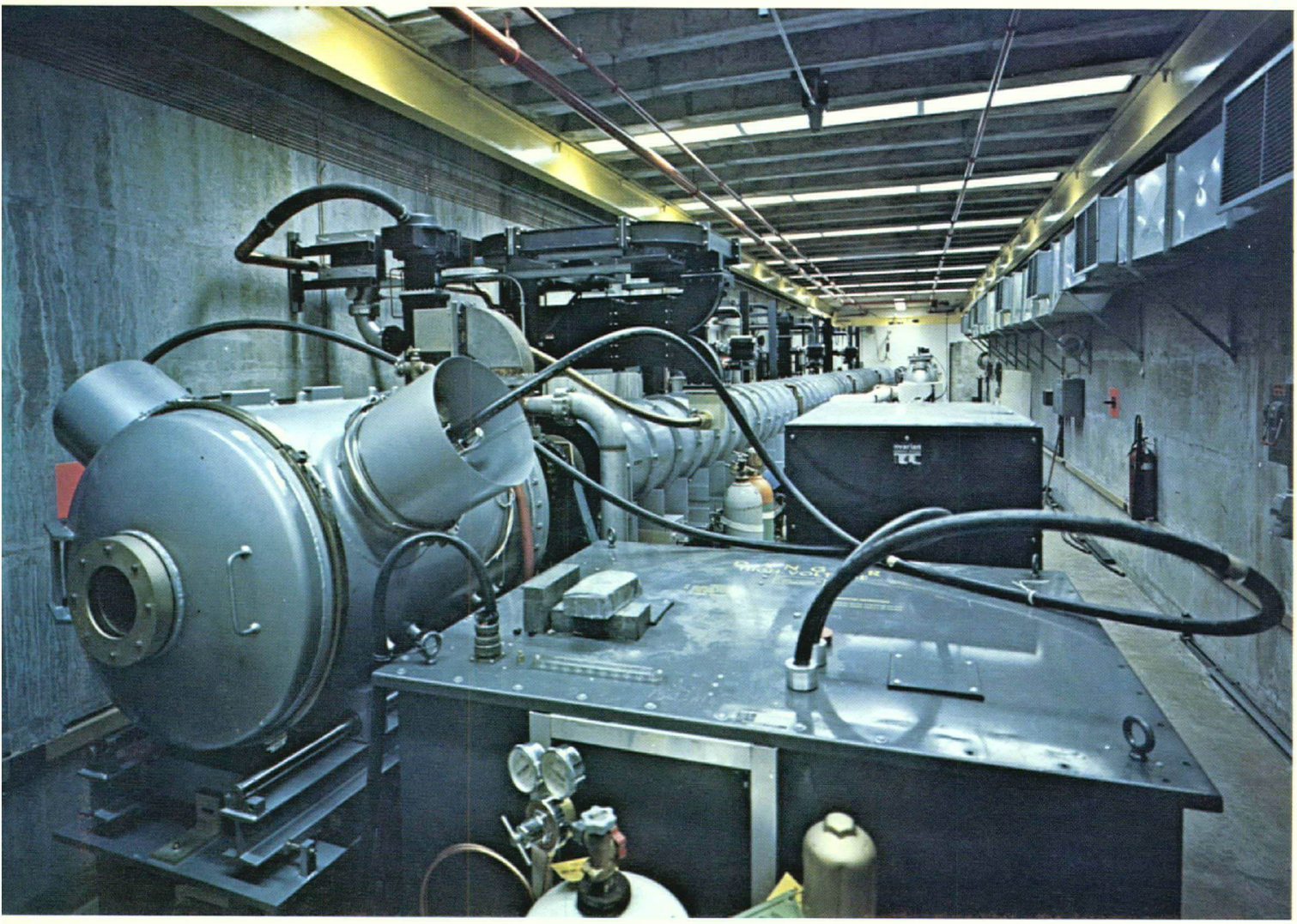
# The Oak Ridge Electron Linear Accelerator

A powerful new physical instrument came into operation at the Laboratory in the summer of 1968 when the Oak Ridge Electron Linear Accelerator (ORELA) produced its first beam. Although by the end of the year the accelerator had not completely passed its acceptance tests, this is a fitting time to describe what the ORELA is and what it can do.

The main purpose of the ORELA is directed to the measurement of neutron cross sections, using the time-of-flight technique. As such, it is a thousand times better than the old reactor-chopper method. The neutrons are generated in short, intense bursts when clumps of high-energy electrons are made to strike a tantalum target. The neutrons travel over a "flight path" and arrive at a detector at a measured later time that depends on their energy. When a sample to be measured is interposed, it affects neutrons of different energies in different ways, and thus the interaction cross sections are measured as a function of neutron energy. A great variety of cross-section measurements can be made, given this basic facility.

## The Accelerator Proper

This is the prime mover in the extensive ORELA complex. Bursts of electrons are pulsed from a hot-cathode gun into the end of a 75-ft-long evacuated tube. The tube is a "diaphragm-loaded waveguide"; that is, it conducts an electromagnetic wave from one end to the other, and the wave front catches up the electrons, forcing them toward the velocity of light. The electromagnetic wave is fed by powerful klystrons located nearby in the next room with connecting rectangular waveguides. A typical bunch of electrons is caught on one of the wave fronts and "surfboards" down to the end of the tube through holes in the diaphragms. It is kept in line by a magnetic field produced by coils around the tube, and it is also steered by additional coils. When the electrons reach the water-cooled tantalum target, they have an energy of 140 MeV. The peak current is about 15 A, and the length of the electron



*View of 75-ft-long linear electron accelerator at Oak Ridge. Bursts of electrons ejected from the electron "gun" in the foreground (large cylinder) travel down a linear waveguide and are boosted in energy by "surfboarding" electromagnetic waves. Klystrons required to drive the waves are beyond the wall at the left, and the connecting waveguide can be seen above the accelerating tube. At the far end of the accelerator the electrons are injected into a tantalum target housed in a heavily shielded circular room. When the tantalum is hit by the electrons, it emits neutrons, which enter several flight paths and are sorted in energy by timed detection circuitry according to their time of flight to the point of measurement.*

pulse can be varied from a microsecond down to 2 nsec. At a pulse repetition rate of 1000 per second, the time-averaged beam power can reach 50 kW.

Each pulse of electrons generates about $10^{13}$ high-energy x rays (bremsstrahlung) and $10^{11}$ neutrons when it strikes the tantalum. The neutrons are emitted in all directions, but the x rays go mainly forward. There is so much energy in these x rays that they would destroy the concrete shielding of the target room; so a barrier of water-cooled lead is interposed between the tantalum target and the back wall.

The ORNL electron accelerator is one of the most powerful machines of its kind. The shortness of the 2-nsec pulses is an unusual advantage because it increases the resolution of the neutron spectroscopy. The accelerator poses severe safety problems, because a single x-ray burst would be fatal to a man standing in the target room. Interlocks and a lock-and-key switching system have been provided to prevent accidents.

## The Building

The building that houses the ORELA is to a large extent a part of the apparatus. The accelerator and experimental facilities are underground and buried under a hill of earth for shielding purposes. The concrete target room in which the neutrons are produced is 12 ft in diameter and 10 ft high. The equivalent of 16 ft of concrete is required for shielding. The target room can be evacuated to a pressure of less than 0.01 atm. The air is pumped out to reduce the backgrounds in the experiments due to scattered neutrons and to eliminate the production of radioactive and noxious gases. The air is exhausted through a 50-ft stack, and it is carefully monitored for radioactivity.

Eleven evacuable neutron flight tubes (with multiple flight stations on each flight tube) radiate from the target room. The detectors for the experiments are located in the various "flight stations," which are all underground. The distances of the "flight stations" from the target vary from 15 to 250 ft, with a 600-ft station planned. The signals from the detectors, which may contain pulse-height information as well as timing information, are fed back to the laboratories in cable trays insulated from ground. To prevent "ground loops," these cable trays are connected at only one point to a grounding mat consisting of ½-in.-diam rods 1 ft apart buried under the accelerator area.

While primary emphasis is placed upon the neutron cross-section spectroscopy, nevertheless the electrons themselves can also be used directly for nuclear structure studies. For this reason, an "electron room" is provided, but initially this room will be used for flight stations for four time-of-flight neutron experiments.

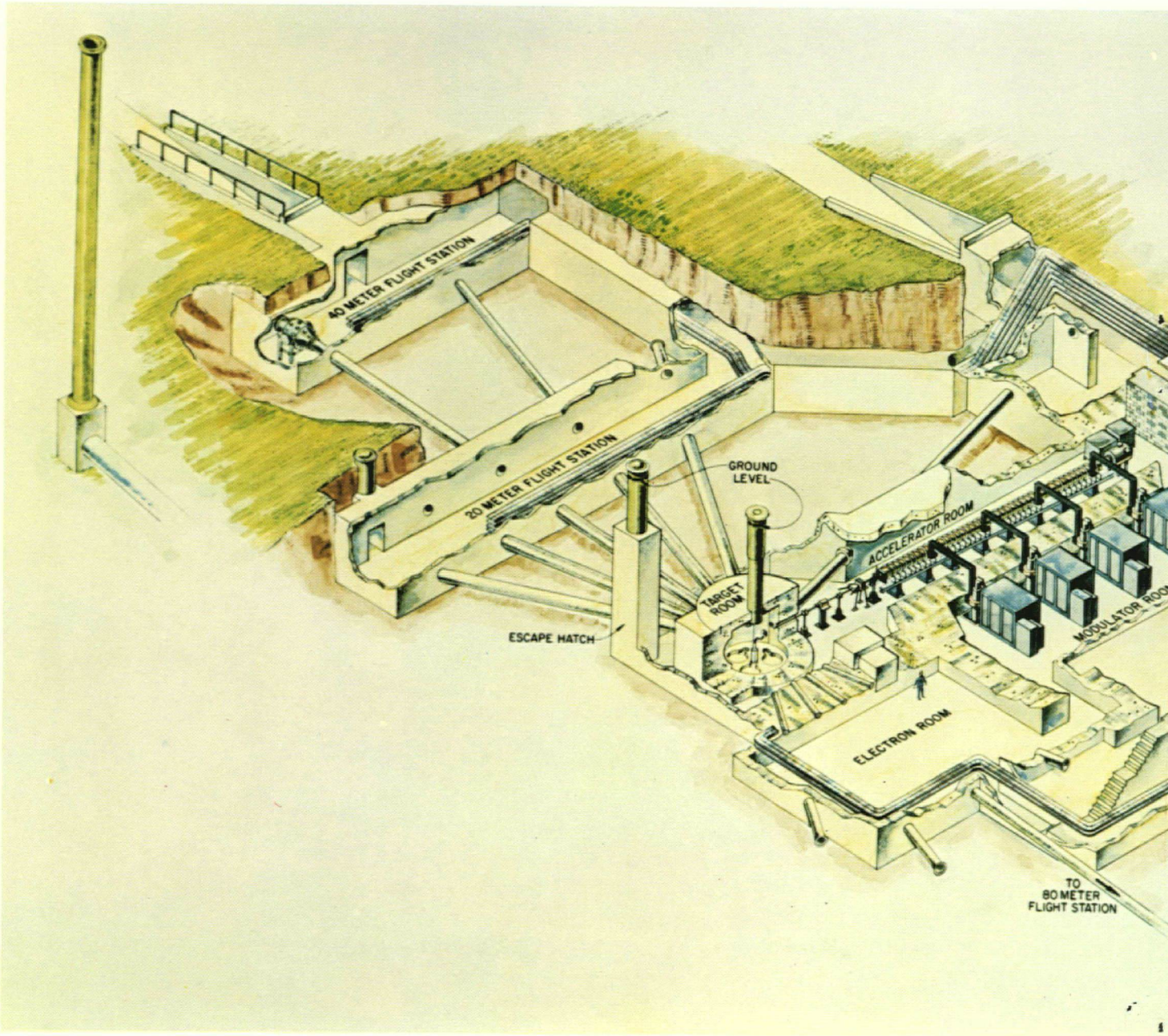
The offices and laboratories are the only part of the facility located above ground. Ten laboratories which contain the needed electronic equipment surround the central data room. The computers for data acquisition and the control console for the accelerator are located in this room. The building has been occupied by research staff members since March 1968.

## Data Acquisition Equipment

Since many experiments can be in operation simultaneously, the ORELA will be capable of producing more experimental data than all other existing ORNL facilities combined. To process and store these vast amounts of data a unique system has been designed and put into operation. It consists of two small digital computers with two fixed-head disks with 400,000 and 800,000 memory capacity for channel and program storage designed specifically for rapid semirandom access of individual channels. Each experimental event is tested to determine whether it is to be ignored or stored into a channel address whose contents are then incremented. Several experimenters will be able to accumulate data simultaneously on each computer, provided their combined data rates are not over ~5000 events/sec and the total number of channels required is somewhat less than the number of words on the disks.

## Experiments

Neutron cross-section experiments of all types are planned for the ORELA by physicists in the Neutron Physics and Physics Divisions. Measurements of the fission and capture cross sections and the ratio of the capture to the fission cross section at neutron energies up to hundreds of kilovolts are of vital concern to the Atomic Energy Commission's liquid-metal fast breeder reactor program. Neutron capture cross sections of many nuclides in the energy range from 1 to 500 kV are



plutonium-244, curium-248, californium-252, or einsteinium-253 have been fabricated and irradiated. These irradiation capsules are prepared by mixing the oxide of the element with aluminum powder and pressing a single pellet which is then welded into a small aluminum container (~3/8 in. in diameter by 2½ in. long) called a "rabbit." An irradiation facility has been installed in the center of the HFIR flux trap in which a "rabbit" can be inserted and removed hydraulically while the reactor is in operation. Thus it is possible to irradiate these special samples at a thermal neutron flux of about $2.5 \times 10^{15}$ neutrons $cm^{-2}$ $sec^{-1}$ for periods varying from a few seconds to several days. Data gathered from such irradiations have been useful for determining neutron capture cross sections. The einsteinium-253 irradiations were made to produce einsteinium-254*m* (39 h half-life) for studies at Argonne National Laboratory. Since requests for einsteinium have exceeded the present main-line production capacity, "rabbits" containing 2.7 mg of californium-252 have been fabricated and are being irradiated to produce additional einsteinium.

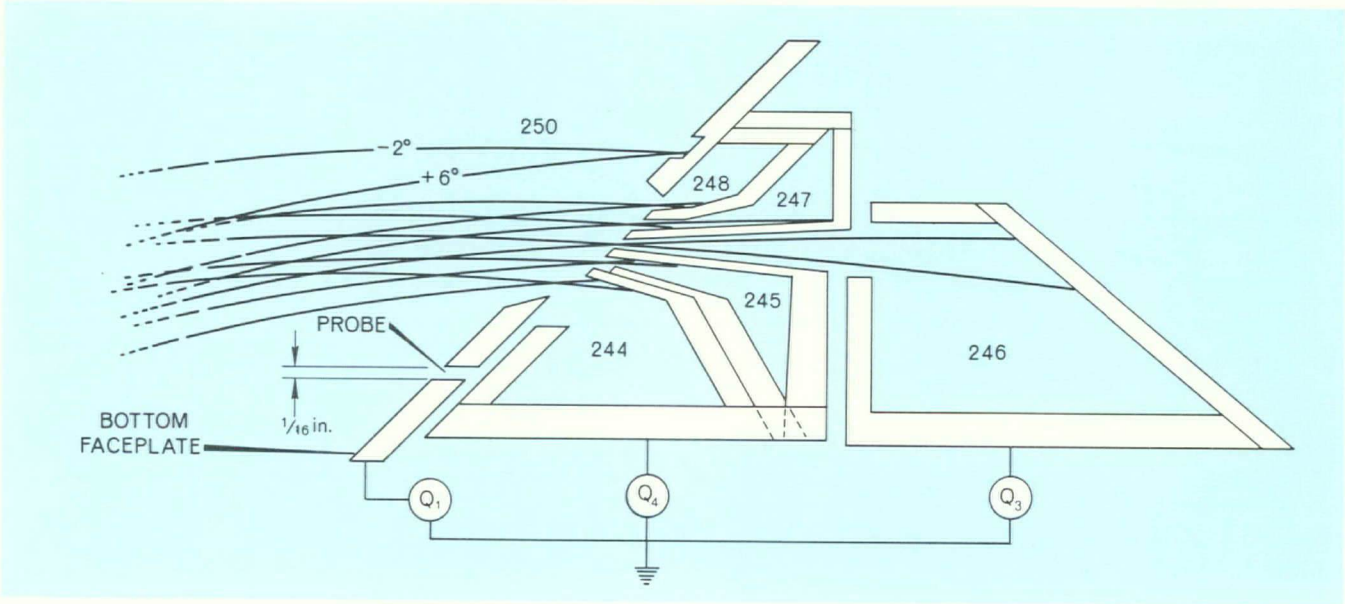
## Electromagnetic Separation of Curium Isotopes

The Oak Ridge National Laboratory electromagnetic isotope separators provided the first substantial quantities of enriched curium isotopes in 1968. A separator normally used to enrich plutonium and uranium isotopes was used to process 3.48 g of the intensely radioactive curium, which emits alpha, neutron, and gamma radiation. Products of the isotopic composition shown below were obtained:

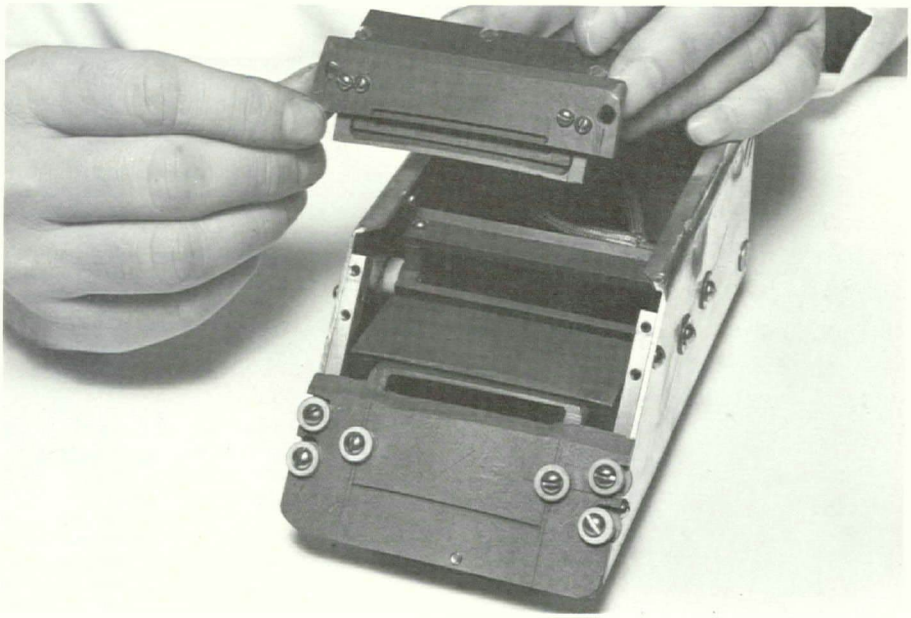
| Isotope | Amount (mg) | Isotopic Composition (%) |
|---|---|---|
| $^{245}Cm$ | 8 | 21–76 |
| $^{246}Cm$ | 15 | 13–96 |
| $^{247}Cm$ | 1.25 | 19–21 |
| $^{248}Cm$ | 0.9 | 30 |
| $^{250}Cm$ | 0.37 | 5.5 (ppm) |

The major component of all feeds was curium-244, but one feed contained 10% of curium-246, and another 4% of curium-245. This variation in feed composition caused the wide range in product composition shown.

Miniaturized, readily repairable equipment had to be developed to handle the small quantities of ionized feed and to collect the isotopes, in a separator usually used to process hundred-gram quantities. The receiver




*Curium receiver.*

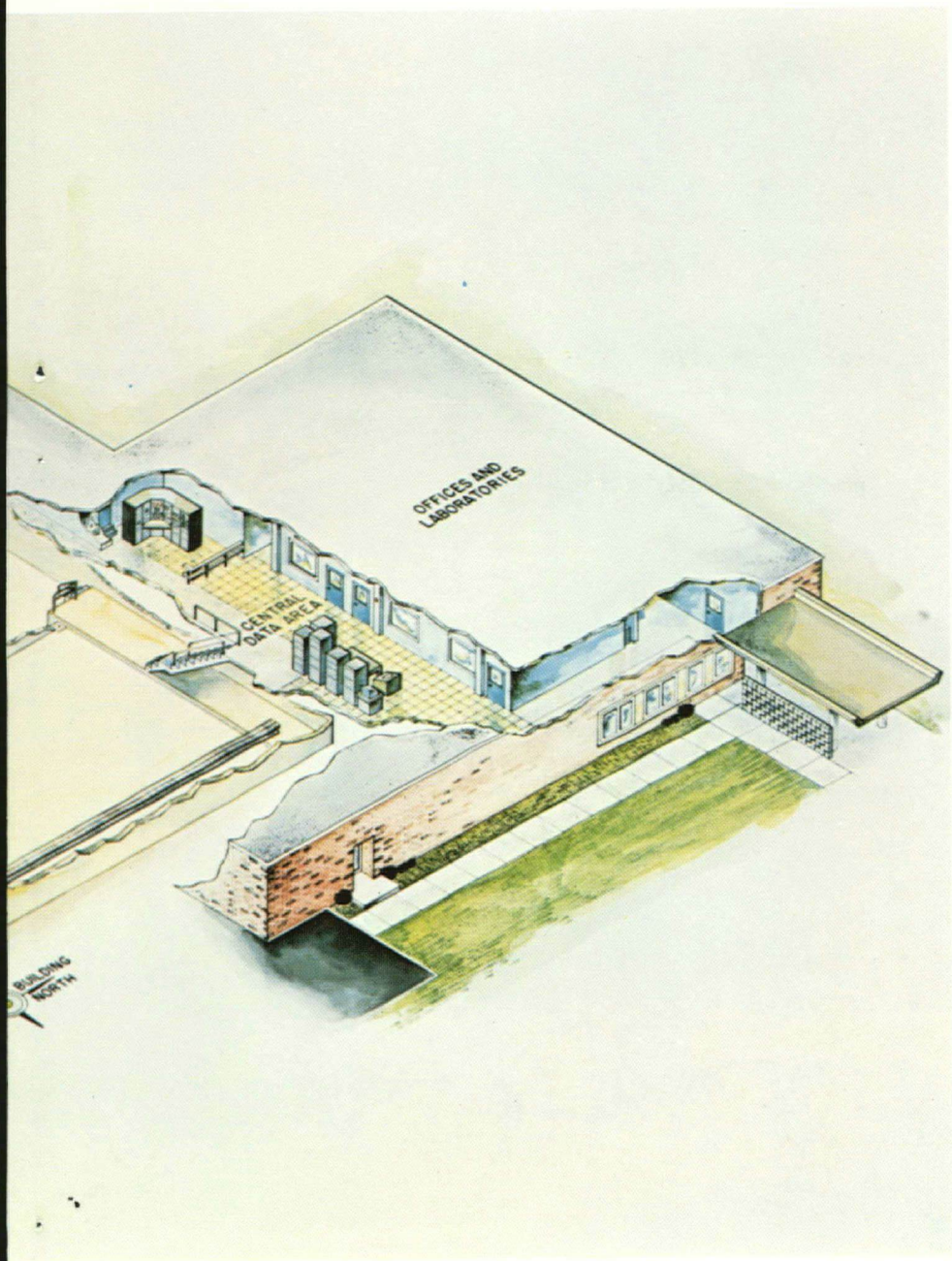


*Curium isotope receiver assembly.*

shown in the photograph was used to collect the separated ionized beams of curium isotopes as illustrated in the schematic drawing. It was removed from the separator through a vacuum lock to shorten the time required to recover a vacuum in the calutron tank.

The enriched curium isotopes are to be used in determinations of their neutron cross sections with the high precision needed to define the production scheme of transplutonium elements of higher atomic number, such as californium-252. In addition, the longer-lived isotopes, curium-245 (9320 years), curium-246 (5480 years), curium-247 (16.7 million years), and curium-248 (470,000 years), can be used as accelerator targets and in experiments that would be affected by the more intense radioactivity of curium-244 (18.1 years).

of interest for nucleosynthesis and also are of importance for reactors. The spectra of the gamma rays emitted when neutrons are captured in individual resonances yield information on the mechanism of neutron capture and on nuclear structure as well as information needed for designing radiation shields. Experiments with aligned fissile nuclei and polarized neutrons and polarized fissile nuclei will enable us to better understand the complicated fission process.




*Layout of Oak Ridge Electron Linear Accelerator. The accelerator is located in an underground room shown near the center of the drawing. Electrons travel along the accelerator from right to left, and the tubes radiating from the target room carry the neutrons to the various experimental stations situated 20, 40, and 80 m away. The "electron room" shown in the illustration is currently used to provide additional flight stations for neutron experiments. Eventually, when provisions are made to deflect the electron beam into the room, it will be used for experiments involving direct interactions of electrons with nuclei.*

# Mathematics and Computation

## About Prime Numbers . . .

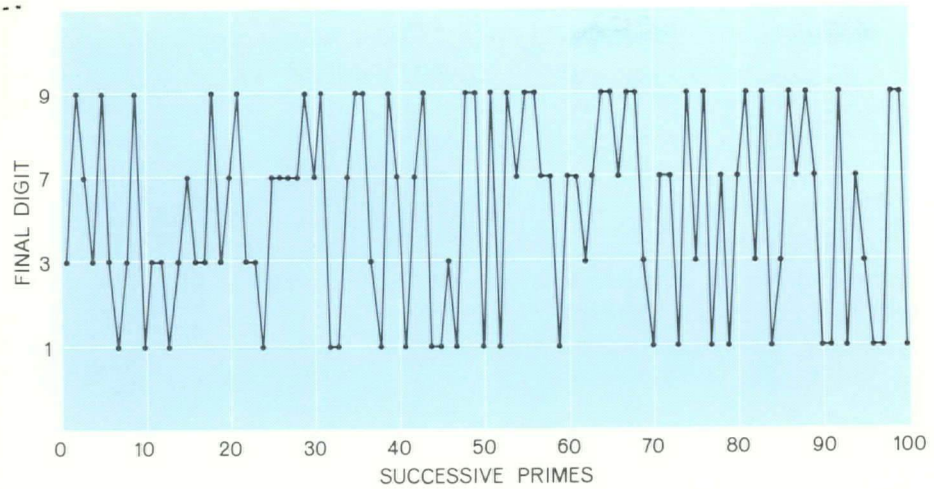
Everybody knows that a prime number is a number evenly divisible only by itself and 1, that all prime numbers are odd except 2, and that the only prime number with 5 as a last digit is 5 itself. This means that all prime numbers above 6 end in 1, 3, 7, or 9.

The number of prime numbers is infinite; indeed, since Euclid's time, it has been known that there is no largest prime. Every natural number can be expressed as a product of primes; for example, $360 = 2^3 \times 3^2 \times 5$. But there is no known formula for the $n$th prime number, nor is there any recursive relation between successive primes. There is no known formula for the number of primes less than a given value $X$, although a so-called prime number theorem gives an asymptotic result. A famous conjecture by Goldbach states that every even integer can be expressed as a sum of two primes, but the conjecture has not been proved, although it has been verified to large values of integers.

Can any systematics be found in the primes that end respectively in 1, 3, 7, and 9? It has been said that "since Euler's time, a general law has been discovered . . . which, applied to our particular case, asserts that there are infinitely many primes of each kind (viz. those ending in 1, 3, 7, or 9), and what is more, that each kind is equally probable." However, an analytical proof of this implied randomness in the occurrence of the four kinds of primes is formidable.

## Enter the Computer, Probability Theory, and the Markov Chain

It's easy for a computer to test the primeness of a number. Given a number ending in 1, 3, 7, or 9, the computer is asked to divide by the smaller primes, one after the other, and if no whole-number multiplicity is found, the original number is a prime. In this way, prime numbers have been located up to $10^{10}$ or more. The largest prime number so far found is alleged to be $2^{11213} - 1$.




*Transitions between final digits of 100 successive primes larger than 100 billion.*

The straightforward way to test as to whether prime numbers ending in 1, 3, 7, and 9 occur in equal probabilities $\frac{1}{4}$, $\frac{1}{4}$, $\frac{1}{4}$, $\frac{1}{4}$ would seem to be simply to take a batch of the numbers and just count the final digits. The trouble is that you'll never have a random sample, because any such batch must come from the lower population of the infinitude; the complete set of primes, if you could get it, would give you the exact information.

This lack of known randomness can be softened somewhat if one looks not at the numbers themselves, but at how they succeed one another. Statisticians speak of "imbedding a Markov chain," which for present purposes means that attention is focused on the succession. Consider the figure, which shows the final digits of the 100 primes larger than 100 billion. At the left, we see that the first prime in the sample ends in a 3, the next in a 9, the next in a 7, then back to 3, and so on toward the right. We could count up these transitions and find how many times a 3 is followed by a 9, or a 7 by a 1, and so on, and derive the relative frequencies of the *transitions*. It's convenient to arrange the probabilities in a table, but let's not bother with a sample as small as a mere 100. Members of the Mathematics Division have done it for the first 788,000 primes, and their results are like this:

| Final digit of the $n$th prime \ Final digit of the $(n+1)$st prime | 1 | 3 | 7 | 9 |
|---|---|---|---|---|
| 1 | 0.17116 | 0.31016 | 0.31984 | 0.19884 |
| 3 | 0.23235 | 0.15732 | 0.29202 | 0.31831 |
| 7 | 0.25639 | 0.27458 | 0.15734 | 0.31169 |
| 9 | 0.33989 | 0.25797 | 0.23139 | 0.17057 |

Table of relative frequencies in the first 788,000 primes that a prime number with a given terminal digit 1, 3, 7, or 9 be followed by a next prime ending respectively in 1, 3, 7, or 9.

The table means, for example, that if some $n$th prime ends in a 1, then in our sample there is a probability of 0.17116 that the next prime will also end in a 1, a probability 0.31016 that it will end in a 3, a probability 0.31984 that it will end in a 7, and so on. The probabilities in the rows add up to unity, because every $n$th prime *must* be followed by a prime that ends in 1, 3, 7, *or* 9.

If there were true randomness in how the primes succeed one another, all of the numbers in the table would be close to 0.25000, but a glance shows a remarkable thing: it is relatively *improbable* that a terminal 1 follows a terminal 1, or a terminal 3 a terminal 3, or a 7 a 7, or a 9 a 9. In other words, the terms in the downward diagonal are uniformly low. What is more, when the mathematicians looked only at the first 200,000 primes, they found that the tendency is stronger.

### But True Randomness Comes . . .

The question then arises, can the lowness of the diagonal numbers in the table be itself an accident derived from the smallness of the samples, or in other words and to take an example, how reliable is the figure 0.15732 as an indicator of the relative frequency of the 3-3 transitions *of the infinitude*? Here the statisticians apply the Markov chain theory as a test of the reliability of the numbers. The test consists in considering the table to be a matrix and then multiplying it by itself several times to see how quickly the numbers flatten out to 0.2500 all over. Well, they flatten out pretty well after eight self-multiplications. This is considered pretty early as these things go, and it makes the statisticians feel that the numbers are pretty reliable. But still, they feel in their bones that if they could count *all* the primes, the transition probabilities would all be 0.2500. The only way out of the dilemma appears to be that the sample of 788,000 is not only too small, but it's *much* too small. If it were not merely a hundred times as large, but a hundred hundred times as large, then maybe you'd see a flat distribution.

Meanwhile, what do you experimentalists say when you generalize from about ten observations?

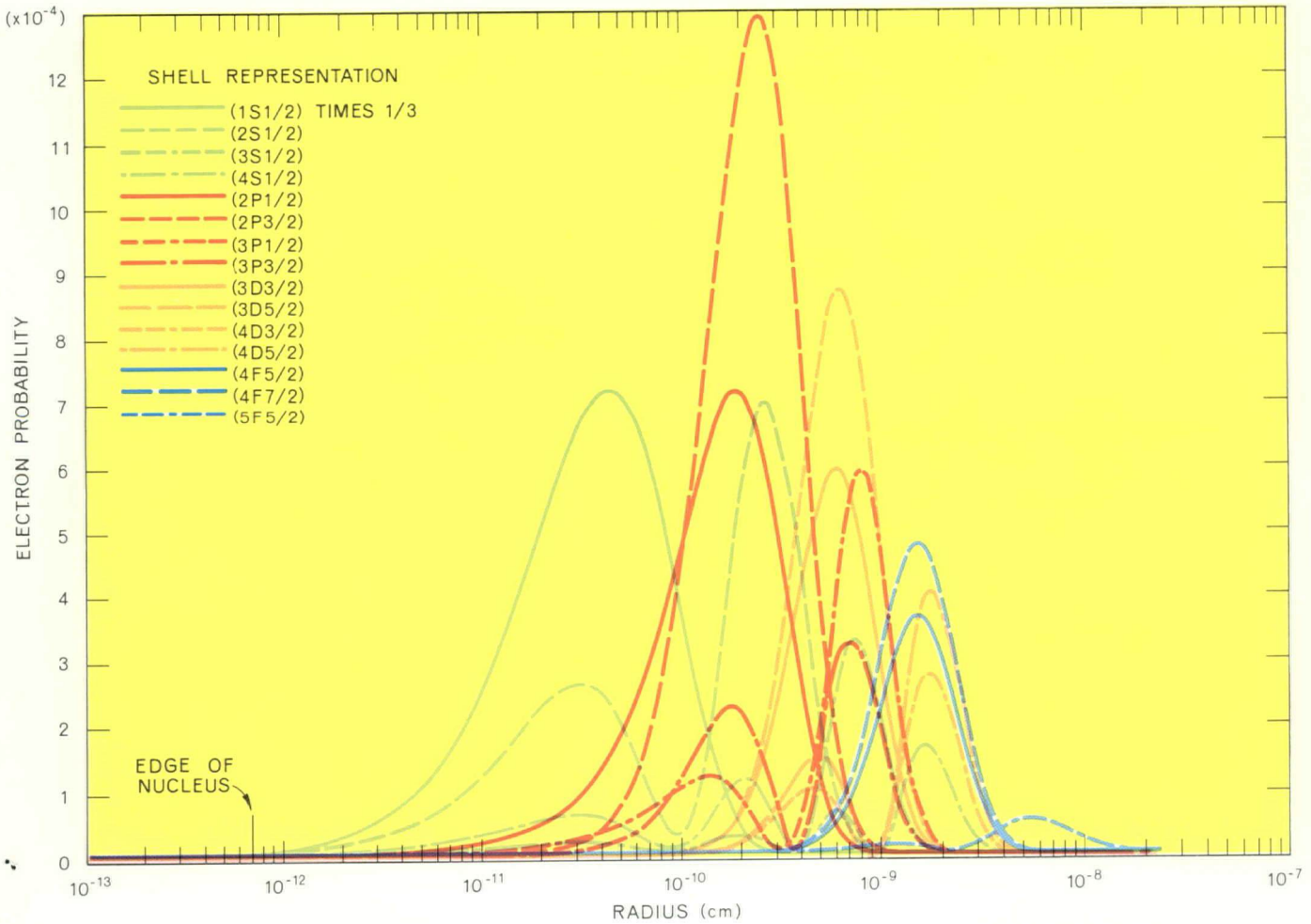
## Atomic Wave Functions

There has been much speculation about the possibility that certain superheavy elements ($Z = 114$ and $Z = 126$) may exist, or can be made to exist, either stably or with relatively long half-lives. It is likely that the radioactive decay of unstable isotopes of such elements will give the most sensitive clue as to their existence. The radioactive decay in turn would probably involve the emission of characteristic x rays. These x rays might be used to identify the elements, whether they are produced by heavy-ion bombardment, or if they occur in minerals, or even if they are products of the explosions of supernovae. Some of the Laboratory's scientists have recently calculated the characteristic x-ray energies of these elements using a computer program that generates atomic wave functions.

There are many other reasons for computing atomic wave functions. Chemical properties of unobservable elements or elements observed in small quantities, such as transuranic elements, can be predicted. This could help in their identification in the laboratory. Effects of extremely high pressure can be estimated. Comparison of calculated and experimental x-ray energies may lead to an improvement in our knowledge of nuclear size and charge distribution. The wave functions assist in calculations of the interactions of radiation and fast-moving particles with atoms and accordingly give radiation dosage information of interest in health physics. One could go on and on.

Since almost all of the physical and chemical properties of atoms depend on the number and arrangement of the atomic electrons, there has been since the beginning of the century a great and continuing interest in the calculation of the electronic orbits. However, the early notion of calculating the precise positions and velocities of the atomic electrons lost importance when Heisenberg announced the Uncertainty Principle in 1927. That principle tells us that the exact arrangement of electrons at a specific time is unknown, and unknowable. What we can hope to know is the *probability* that an electron may be in a certain place or, equivalently, what fraction of its time the electron spends there.

*Results of atomic wave function calculations for uranium. The probability of finding an electron within a spherical layer $4 \times 10^{-14}$ cm thick is plotted as a function of distance from the center of the nucleus. The various maxima represent the electron shells of the uranium atom, each shell being associated with a set of quantum numbers, as keyed in at the upper left in the language of the atomic physicists.*



These probabilities can be readily calculated from other quantities called atomic wave functions. The atomic wave functions in turn can in principle be calculated quite accurately for all atoms by using the Dirac equation, with a number of refinements for small effects such as the effect of the size of the nucleus, already tiny compared with the volume occupied by the cloud of electrons. The application of the Dirac equation to complex atoms, however, is not at all straightforward. The reason is that the energy of each electron is an all-important parameter, and the trouble arises when you try to calculate the potential energy of your electron at a given location. The potential energy is determined by the interactions of the electron's charge not only with the positively charged nucleus but also with all the other negatively charged electrons. But their wave functions in turn depend on their interaction with your electron, so you get into a feedback tangle typical of a multibody problem.

The approximation method worked out nearly 40 years ago consisted in guessing at a wave function for each electron, calculating the feedback interaction between a chosen electron and the other electrons, and then shifting the chosen electron's wave function in a manner favorable to overall consistency. This has to be done for each electron in turn, each shift requiring a reevaluation of the total picture. Hopefully, the required shifts become smaller and smaller until consistency is reached. The procedure, called the Hartree method of the self-consistent field, has had several embellishments since its invention, but for about three decades the calculations had the reputation of being the most laborious in physics.

Even with the most modern computers the problem is still difficult, and special methods have to be developed to find just the right potentials which lead to electron densities which in turn lead back to the same potentials. One of the most advanced methods is now called the Relativistic Hartree-Fock-Slater Self-Consistent Field method, and a computer program has been written to supply ORNL scientists with wave functions and electron densities based on this method.

A special feature of this program is that the effects of nearby atoms may be taken into account, at least approximately. Since much research involves atoms in company with other atoms, the nearby atoms will slightly change the wave functions of the atom under study. This effect is approximately accounted for by the use of a suggestion due to E. Wigner and F. Seitz and known as the Wigner-Seitz boundary condition. Special methods for atoms arranged in a crystal lattice structure are also being developed.

Modern numerical techniques permit the solution of very heavy atoms in 10 min or less. A computation of the uranium atom, requiring a total of 134,478 solutions of the Dirac equation, was completed in 5.9 min. Once calculated, the solutions are stored in a magnetic disk memory, from which they can be retrieved in a matter of seconds. The solution for one atom can also be used as a starting point for calculating a nearby atom, thereby reducing the calculation time for most atoms to less than 3 min.

*A glimpse of the computer facilities at ORNL, with cathode-ray display output.*

The table below shows the results calculated by this procedure for the characteristic x rays of superheavy elements. Values for gold and uranium were calculated to test the computer program.

| | X Ray | Energy (keV) Calculation | Experiment |
|---|---|---|---|
| Gold, $Z = 79$ | $K\alpha_1$ | 68.749 | 68.804 |
| | $K\alpha_2$ | 66.915 | 66.990 |
| Uranium, $Z = 92$ | $K\alpha_1$ | 98.526 | 98.439 |
| | $K\alpha_2$ | 94.703 | 94.665 |
| $Z = 114$ | $K\alpha_1$ | 172.560 | |
| | $K\alpha_2$ | 160.562 | |
| $Z = 126$ | $K\alpha_1$ | 233.952 | |
| | $K\alpha_2$ | 211.362 | |

Thus, if the physicists aim their balloon-mounted x-ray spectrometer at the Crab Nebula and register two x-ray lines at energies of 234 and 211 keV, they can be pretty sure that element 126 was made in the stellar explosion of AD 1054 and that it has lasted since that time.

# Chemical Reactions Between Individual Molecules

For the exploitation as well as for the understanding of the physical world, it is at least as important to know about the changes that may be undergone by a particular kind of matter as it is to know about its properties in its original state. For example, the energy in the nucleus of an atom of uranium-235 or that in the outer electrons of a carbon atom in coal is made available for useful work only by changing the particular kind of matter to another – by fission of the uranium, a nuclear change; or by burning of the carbon, a chemical change. Furthermore, the largest useful function of chemistry, the production of new materials with desirable properties (drugs, dyes, fibers, foods), consists in the purposeful carrying out of chemical reactions to convert available substances into desired new ones.

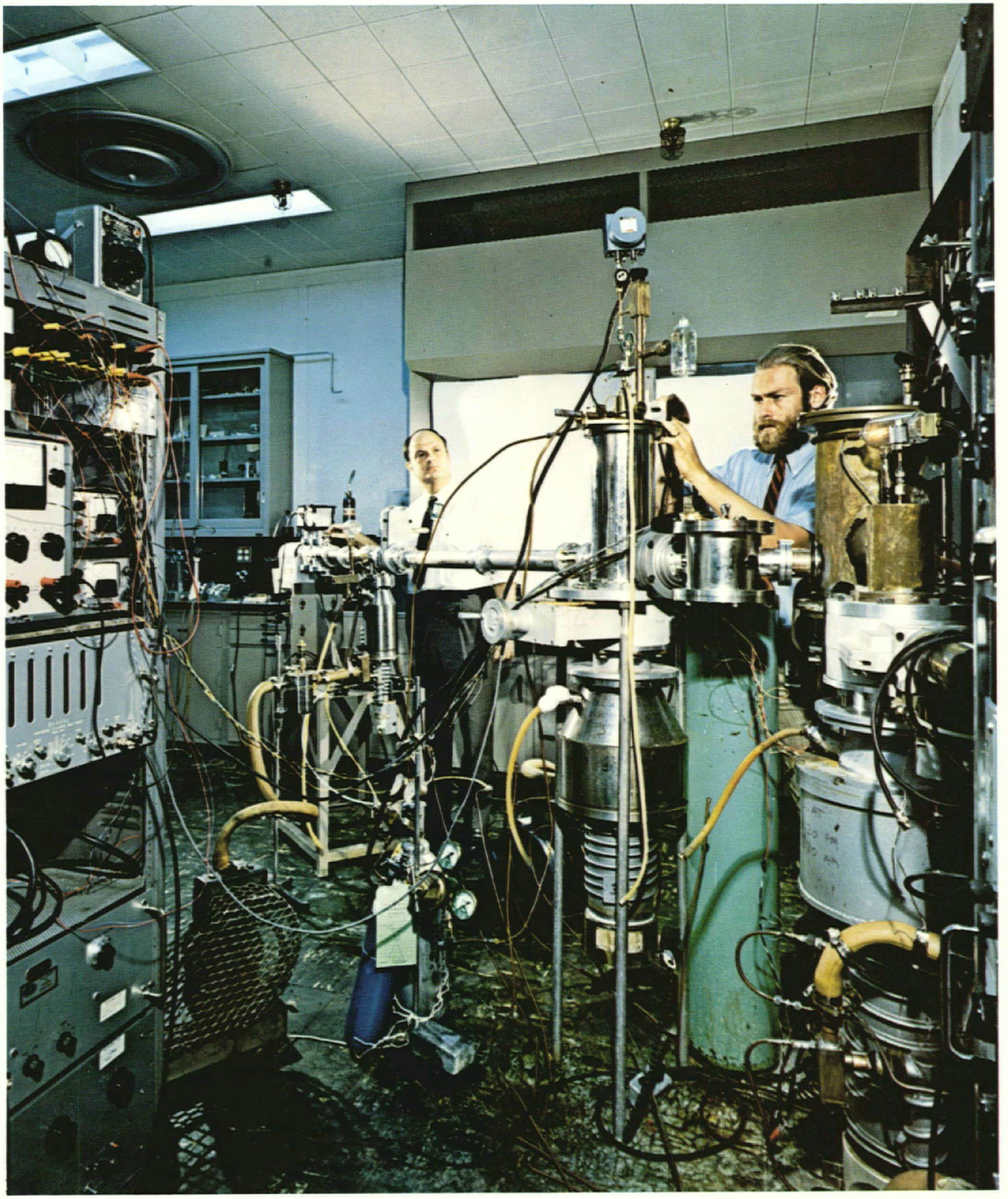
In the study of how a chemical reaction occurs, it turns out that the most revealing information is obtained from the rate of the reaction under various conditions. By studying how temperature affects the rate, for example, it has been concluded that there is an energy barrier that keeps reactions from going as fast as the molecules can get together. Although the average energy of a molecule (or of a beaker full of molecules) increases just in proportion to temperature, the fraction of very high energy molecules increases much faster. Indeed, the fraction, $f$, having energy greater than some arbitrary figure, $E^*$, increases exponentially with temperature, $T$:

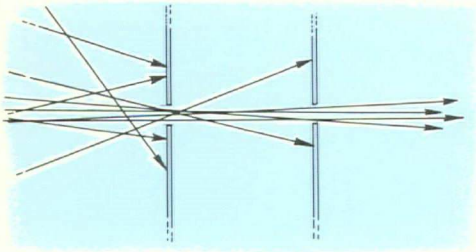
$$f = e^{-E^*/RT},$$

where $e$ is 2.718, the base of natural logarithms, and $R$ is another constant. Since the rates of most chemical reactions follow the same law with respect to temperature, it can be concluded that only those molecules with energy above some value $E^*$ are able to react. By measuring the rate at more than one temperature, the value of $E^*$ for the particular reaction can be calculated. Typically, it is found to be from 10 to 100 times the average molecular energy at room temperature.

A whole branch of chemistry, chemical kinetics, has grown up around the measurement and interpretation of the rates of reactions conducted in bulk, that is, in solutions of the reacting substances or in gaseous mixtures. There is a disadvantage to studying reactions in bulk,

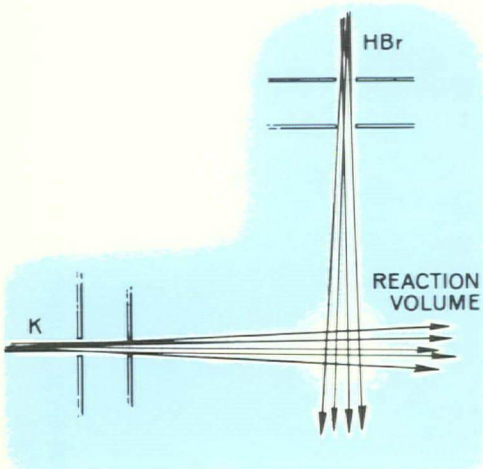
*Apparatus for the study of individual collisions between atoms and molecules (Chemistry Division).*





however: the details of the individual molecular collisions that lead to reactions are obscured by the chaotic nature of a sample of molecules, either liquid or gaseous. Only average properties of the whole sample can be measured, either before or after reaction, and instead of knowing the velocities and orientations of individual pairs of colliding molecules, one knows only the average energy of all of them, as measured by the temperature.

A way to circumvent the molecular chaos has been known in principle since 1912, when Dunoyer described the production of beams of atoms or molecules by allowing them to leak out into a vacuum through a narrow slit. If the pressure behind the slit was low enough (typically about 1/10,000 of atmospheric pressure), then the molecules would come out without bumping into each other after exiting, and would therefore stream away in straight lines from the slit, like rays of light. If a second, parallel slit was interposed in the vacuum some distance from the first, the molecules which traversed it formed a ribbon-shaped beam in which all the molecules were going in the same direction, without collisions. Such a beam proved useful for experimentation on isolated atoms or molecules. For instance, a magnetic or electric field could be placed across the beam, and any deflection observed could be attributed to magnetic (or electric) properties of the individual atoms or molecules. To be sure, the observation of a gross deflection of the beam represented an average of the deflections of many millions of individuals, but because they were isolated from each other by virtue of being in free flight, there were no interactions between different molecules to take into account as there would be in a gross sample. It was as if each molecule was being experimented upon separately.




It was apparent for a long time that molecular beams could in principle be used to study chemical reactions in an almost ideal way. If two molecular beams, one of one substance and one of another capable of reacting with the first, were aimed so as to intersect, then there would be collisions in the region of intersection and not elsewhere. Some of the collisions would be elastic, like those of billiard balls, and would merely scatter the colliding molecules out of their respective beams. Some collisions would result in reactions, and the fragments leaving such collisions would differ chemically from the original molecules. The feature that would be so valuable about this way of studying a reaction is that the product molecules would be observed without having undergone any but the one collision that produced the reaction, and would therefore be in just the state in which the reaction left them. "State" in this context means not only their velocities but also the degree to which they are vibrating internally and rotating. Furthermore, the directions which the fragments would take would not be disturbed, so that one could really see every detail of the collisions.

Although the molecular beam approach to chemical kinetics was thus obviously a most desirable one, a basic difficulty prevented its realization for many years. Since a molecular beam is formed by the effusion of an initially dilute gas into a vacuum, the number of molecules arriving in this beam at some distance from the source is very small, and only the

most sensitive means are adequate to measure it. Trying to measure the scattered products of a chemical reaction is even worse, since only a small fraction of those molecules undergoing some sort of collision succeeds in reacting. So, although certain physical experiments of great interest were done with molecular beams between 1920 and 1939, the successful study of a chemical reaction in this way remained an elusive hope.

Sensitive detectors were available for only one class of substances, the alkali metals, and the key to eventual success was found in the choice of one reactant from this class. Chemists at ORNL realized that interesting chemical reactions could be found that involved these metals and were optimistic that they could somehow employ the sensitive surface ionization gage to measure not only the one reactant, say potassium, for which it was already used in physics experiments, but also a product of some interesting reaction, say potassium bromide from the reaction

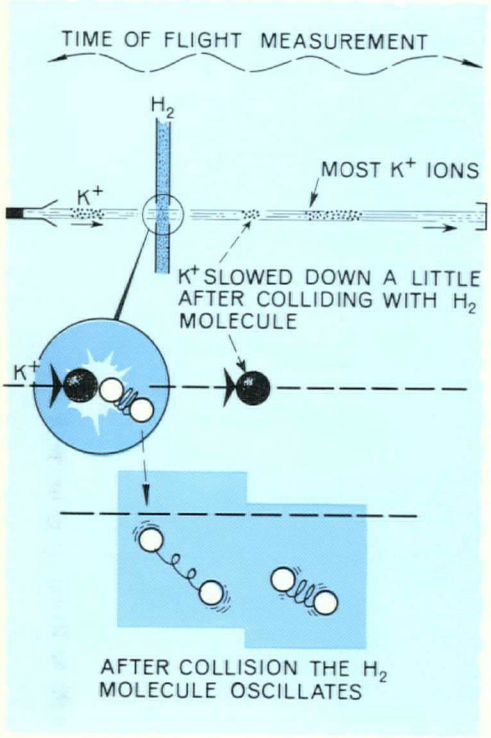
$$K + HBr \rightarrow KBr + H .$$

In the beginning, the ORNL workers planned to use neutron activation analysis for bromine in conjunction with the surface ionization gage to distinguish between K, which would be elastically scattered into every angle, and KBr, the reaction product; but they discovered a modification of the surface ionization gage that could distinguish between K and KBr. With this in hand, the experiment was a success.

Other laboratories took up this kind of research, using the Oak Ridge technique, and their work together with ours has made our knowledge of the reactions of alkali metals with halogen compounds more detailed than that about any other group of chemical reactions. The limitation to alkali metals is of course a severe one, and great interest attached to the possible use of a mass spectrometer for detection of products, since it could ionize (with an electron beam) any molecule and distinguish it by the mass of the resulting ion. The difficulty was the inherent inefficiency of the ionization step. However, gradual improvement in such ionization sources and in the general performance of ion counting circuitry began to make the mass spectrometer marginally feasible for detecting molecular beams, and the ORNL group performed the first reaction study done by this means. They chose the simplest, and therefore the most interesting, reaction of all, the exchange of a deuterium atom for a hydrogen in hydrogen gas:

$$D + H_2 \rightarrow HD + H .$$

Here the greatest difficulty proved to be the distinguishing of HD molecules coming from reactions from those present as impurity in the $D_2$ gas used to make D atoms, and it prevented as complete success as in the alkali-metal experiments; but the results showed clearly that the technique was feasible, thus opening up almost any reaction (of sufficient velocity) for study in crossed molecular beams.



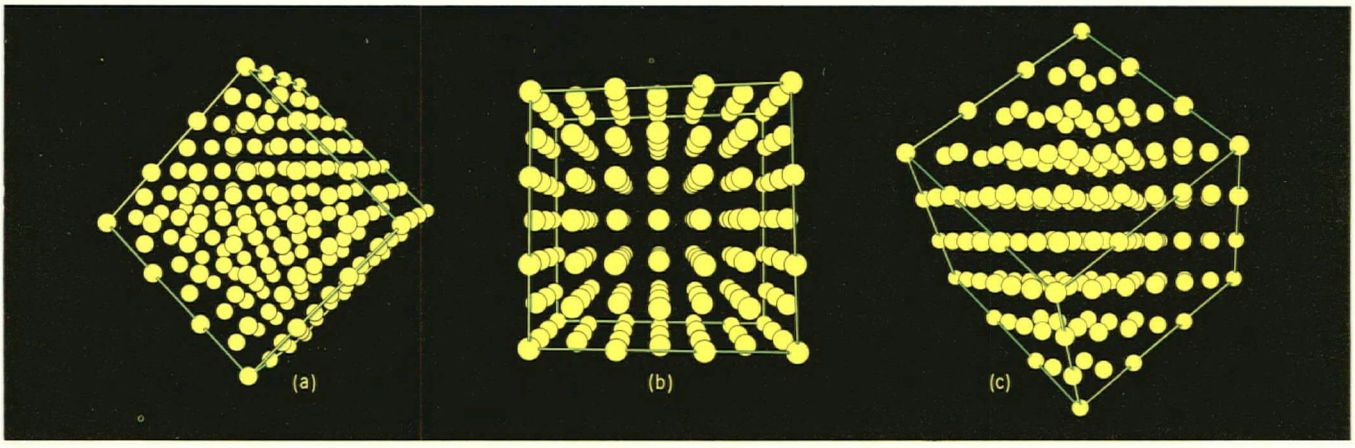

A powerful feature of the beam technique is that it lends itself to energy selection of reactants and energy measurement of products. This is because the molecules are spatially isolated and can be operated on by velocity selectors and other devices before they get to or after they leave the reaction zone. Men in the Chemistry Division have recently been using this ability to explore a simpler molecular process than chemical reaction, namely, the excitation in collision of molecular vibrations. They use a beam of alkali-metal ions for ease of production and detection and are studying collisions between it and hydrogen molecules to see the details of energy transfer from a fast potassium ion to vibrations of the $H_2$ molecule. They study the energy transfer not by looking directly at the excited $H_2$ molecules (it would be difficult to see the vibrational excitation directly) but by measuring the loss in velocity by the K ions after collision. The $K^+$ ions are produced in a source, collimated into a beam, accelerated to the desired velocity, allowed to traverse (through fine slits) a short region containing the scattering gas ($H_2$), and detected some distance farther down an extension of the vacuum system. The velocity before collision is set by the accelerating voltage used, and the velocity after collision is measured by a technique borrowed from nuclear physics, time-of-flight spectroscopy. The beam of ions is chopped into short pulses, and the time required for one ion of a pulse to reach the detector several feet away is measured. Most of the transit times recorded correspond simply to the original velocity of the ions, because most ions fail to make a detectable collision with the $H_2$ gas. A small number are delayed by making elastic (billiard ball) collisions, and a fraction of these are delayed somewhat differently because some energy is transferred to vibrations of the $H_2$ molecule, a process which alters the elastic kick given to the $K^+$ ion in the collision. This shift in the time of arrival can be definitely resolved, and it changes in an interpretable way with changes in the original energy (velocity) of the $K^+$ ions. For instance, at a high enough energy the $H_2$ molecule flies into two atoms, and the onset of this process can be observed.

The chemical reactions which sometimes follow nuclear decay, because the product of the decay is given an energetic kick by the departing beta particle (for example), involve energies in the range which these new experiments cover, and one is therefore in a region of considerable direct interest to AEC programs. Not all of the studies that have been made with molecular beams have had so direct a relevance, but because they have advanced basic understanding in an important part of chemistry the chemists have pursued them. Other laboratories would eventually have undertaken such research if the Oak Ridge chemists had kept more narrowly to strict programmatic interests, but the success of this research demonstrates the advantage of sometimes undertaking difficult and risky ventures at a laboratory such as ORNL.

# Ion Crystallography

The regular arrangement of the atoms of crystalline substances is revealed by its influence on a variety of phenomena, including the transmission and reflection of visible light and of x rays. Studies of crystals by means of these radiations are termed *optical* and *x-ray crystallography* respectively. It was suggested nearly 60 years ago by Stark that the internal regularity of crystals, then being first disclosed by x-ray studies, might also influence the motion of energetic ions, but his proposal lay forgotten for half a century. In the interim, the discovery of fission and the development of nuclear reactors directed the attention of materials scientists to the effects of irradiation, particularly by fast neutrons and fission fragments, on the properties of solids. A proper understanding of these effects required detailed knowledge of the way in which energetic atoms and ions move in solids, these being either foreign atoms such as fission fragments and particles from an accelerator, or atoms of the solid itself, displaced from their proper positions (*lattice sites*) by collisions of fast neutrons or of other particles. Theoretical studies of the slowing down of energetic atoms in solids were initiated in the Solid State Division more than ten years ago, as a foundation for understanding radiation effects in solids. The original objective was the calculation of the *range* of a moderately energetic atom in a solid, that is, the distance which the atom would travel in slowing down to rest by collisions with the atoms of the solid. Experimental data of accuracy adequate for testing the theory were just becoming available when these calculations were begun.

In 1961 a new approach to the study of energetic atom ranges was initiated at ORNL, employing a large digital computer to *simulate numerically* the many collisions made by such an atom as it slows down to rest and to calculate the distance which it travels in doing so. A machine can be caused to carry out such calculations for a large number of moving atoms (projectiles), differing slightly from one another in their initial positions or directions of motion, thus rather realistically simulating the results of an experiment. The original calculations were performed for amorphous solids, in which the atoms are not arranged in a regular manner, but they were soon extended to crystals of simple metals, each atom of the target being located at its proper lattice site. These calculations showed that atoms entering the target crystal in



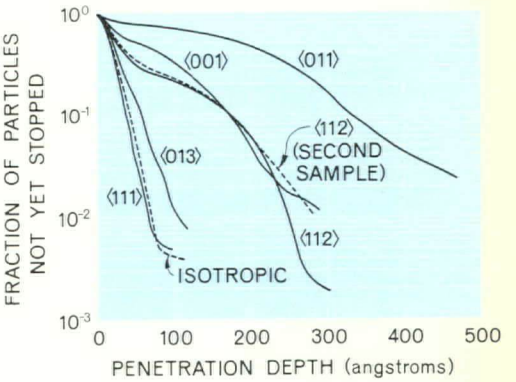

*These sketches show a face-centered cubic crystal as viewed from various vantage points. In* a, *the viewing direction is more or less randomly chosen and displays rather little symmetry. In* b, *the viewpoint is selected to show the* ⟨*001*⟩ axial channel: *note how the atoms appear to lie in* rows. *In* c, *a* planar channel *is displayed: the atoms now lie in closely packed but widely spaced sheets.*

certain directions penetrate exceptionally deeply into it. The directions of greatest penetration were found to be associated with *channels* in the target crystal, that is, with regions of the crystal symmetrically surrounded by several long straight rows of closely packed atoms. A projectile is constrained to move in a channel by making a large number of glancing collisions with the rows of atoms constituting the walls of the channel. In each collision the moving atom is deflected only slightly, the cooperative effect of many collisions with adjoining target atoms serving to steer the projectile away from the row of atoms and back toward the channel axis. Since only a very small energy loss occurs at each collision, the channeled projectile is able to penetrate much more deeply into the target than can an unchanneled particle. This phenomenon, now termed *axial channeling,* was first reported at a small conference of specialists held at ORNL in the autumn of 1962. Inspired by the ORNL calculations, experimentalists at the Canadian Chalk River Laboratories very quickly confirmed the predictions, thus initiating the field of *ion crystallography,* the study of crystals by means of energetic ion beams.

The original ORNL calculations referred to 1- to 10-keV copper atoms slowing down in copper crystals, while the first Chalk River experiments involved 40-keV krypton ions injected into crystals of aluminum. Experiments with proton beams, first performed at Harwell in England, and extended there and elsewhere, showed that channeling effects were not limited to these low energies but could be observed above 1 MeV as well. Workers in several laboratories have extended the observations to many different ions and to a variety of different crystals. Although axial channels are the main ones observed at low energies, *planar channels* become significant as the energy of the projectile is raised. These are regions of a crystal bounded by a pair of closely packed sheets of atoms. The projectile is constrained to move back and forth between the bounding planes, but its motion parallel to the planes is uncontrolled.

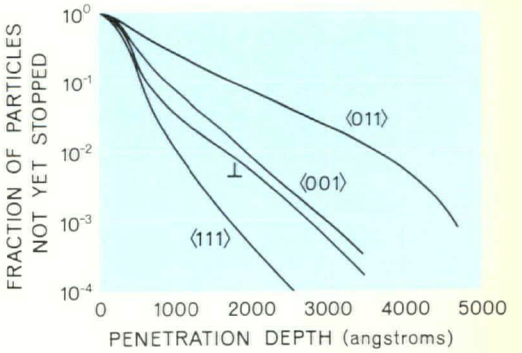
Channeling effects can be observed only if the angle between the incident particle beam and the controlling axis or plane is less than a *critical angle.* Critical angles decrease with increasing velocity of the incident beam but are approximately 0.5 to 2° in typical experiments.

Because of these rather small angles, successful channeling experiments require that the orientation of the target crystal be accurately known and that the direction of the incident beam be carefully aligned. Projectiles traveling in either axial or planar channels are prevented from approaching very closely to the atoms of the target. Consequently, those phenomena connected with close approaches between two atoms will be greatly influenced by crystal effects. The yields of nuclear reactions between the projectile and target are greatly reduced when the projectiles are channeled. This was first observed for the 405-keV resonance of the $^{27}Al(p,\gamma)^{28}Si$ reaction, where the yield of gamma radiation was reduced to 20% of the normal value when the protons were well channeled. Similarly, the yields of proton-excited characteristic x radiation are reduced when the protons are channeled in monocrystalline targets. The wide-angle scattering of energetic light ions by atoms is often termed Rutherford scattering. Because Rutherford scattering requires a close encounter between two atoms, it is strongly influenced by channeling: When the incident ion is directed into a channel of the target crystal, the Rutherford scattering yield is strongly reduced. This is probably the most important of these phenomena, since it permits a readily available beam of protons to be used as a probe for the study of crystals. Finally, the phenomenon complementary to channeling has also been observed. If a particle emitted from a lattice site is directed toward a nearby neighboring atom, it will be strongly deflected. Consequently, particles emitted in such directions cannot reach a suitably located detector and are said to be *blocked.* The emitted particles may be alpha particles from the decay of a radioactive lattice atom, for example, or energetic protons undergoing Rutherford scattering at lattice atoms. Both channeling and blocking effects can be used in ion crystallographic studies. The latter offer interesting opportunities for the examination of radioactive crystals, especially those of substances containing transuranic elements.




*A theoretical machine calculation which shows the influence of the direction of incident 5-keV copper atoms on their penetration into a copper crystal. The most "open" channel is designated ⟨011⟩.*

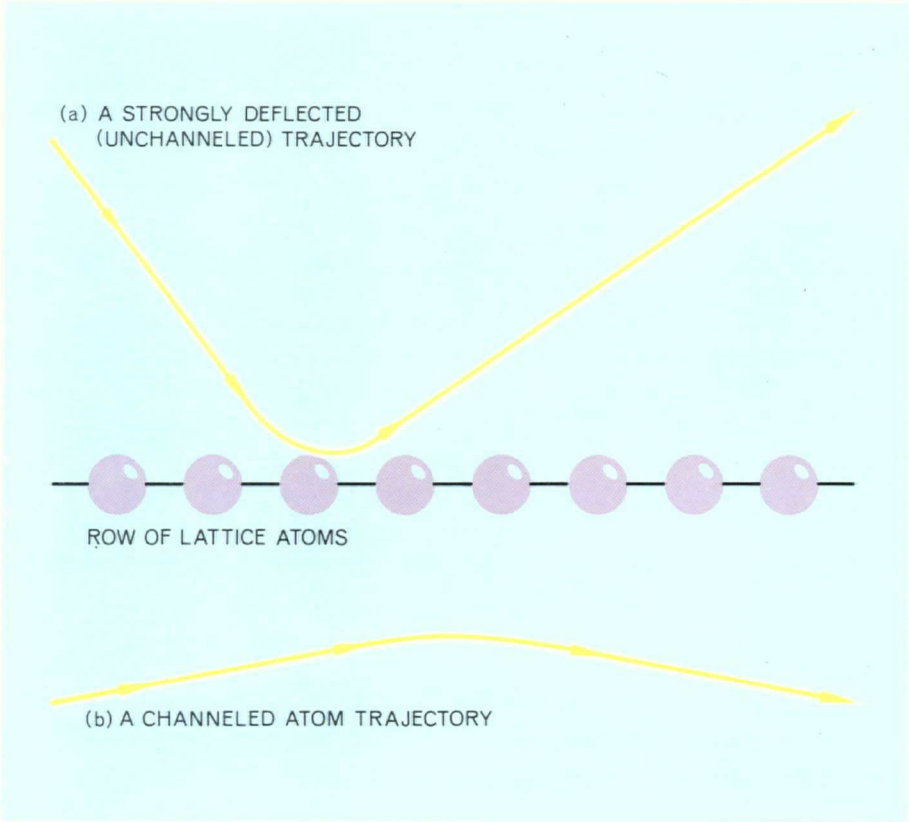
A number of studies of ion crystallography are under way, in part as a cooperative effort of personnel in the Solid State, Chemistry, and Physics Divisions. In one series of experiments, observations are being made of the energy lost by a beam of particles in passing through planar channels in thin gold crystals. Under the conditions of these experiments, the energy losses occur almost exclusively through excitation and ionization of the atoms of the target crystal. When viewed with a small-angle detector the emergent beams display an unexpectedly rich spectrum, consisting of several groups of particles, each with a discrete energy. The groups observed and their populations depend on the nature and energy of the incident ions and on the angular relationships between the incident beam, the crystal axes, and the detector. Essentially similar observations have been made using 15- to 60-MeV iodine ions and 3-MeV helium ions (alpha particles) from the tandem Van de Graaff accelerator. A quantitative explanation of these observations has been given in terms of a model in which the ions oscillate back and forth between the planar channel walls, the wavelength of their oscillation decreasing as its amplitude increases, and in which the energy lost by the ion increases with increasing amplitude of oscillation. The explanation of the group



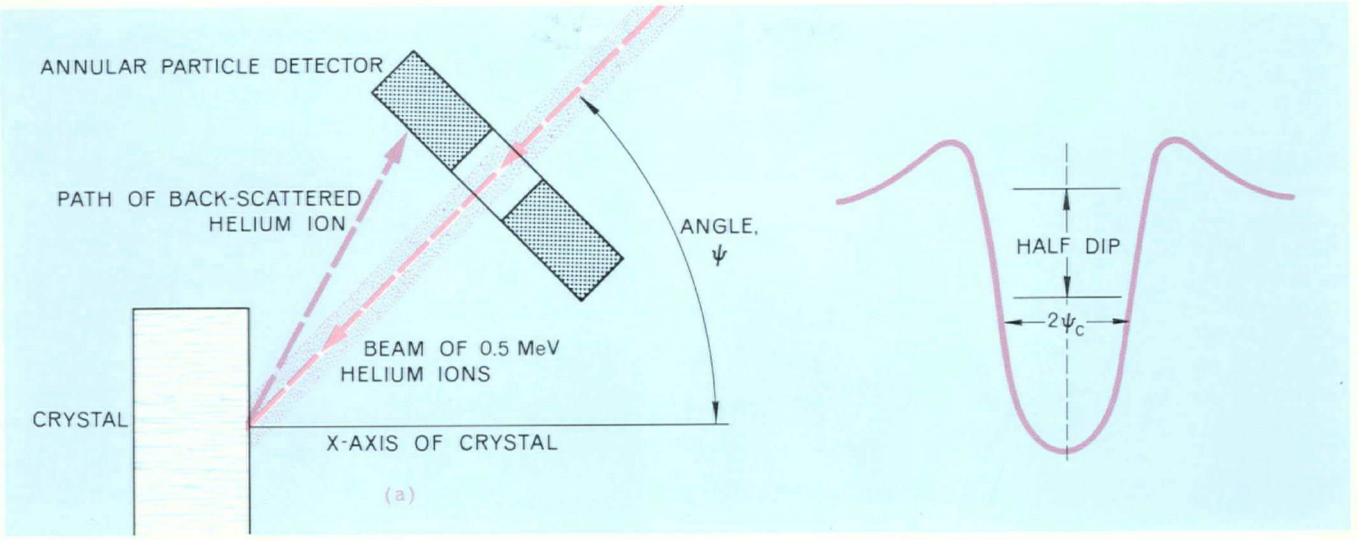

*The first experimental demonstration of axial channeling was performed at the Canadian Chalk River Nuclear Laboratories in early 1963, using 40-keV krypton ions incident on a crystal of aluminum. Note the similarity of these curves to the theoretical ones above.*

structure, as first shown by a group of ORNL investigators, rests on the fact that the detector receives only those ions which emerge from the target in a particular direction. Thus most particles are lost, and only those are detected which perform an integral (or half-integral) number of oscillations in traversing the thickness of the crystal. This explanation opened up the exciting possibility of obtaining for the first time direct experimental information about the nature of the collision between a projectile and a target atom of the crystal. Since the wavelength of the oscillation and the energy loss depend upon the amplitude of the oscillation, it is possible, within limits, to map out the force between two atoms and the probability of electronic excitation as a function of their distance of closest approach.

Another experiment in this program concerns the influence of crystal orientation on the amount of radiation damage produced in a thin gold crystal by a beam of energetic iodine ions drawn from the ORNL tandem Van de Graaff accelerator. When one of these ions, which may be regarded as artificial fission fragments, makes a sufficiently close encounter with a lattice atom, it will initiate a cascade of displaced atoms and produce damage to the crystal observable in the electron microscope as a small black spot. In the experiments performed at the Laboratory, gold crystals about $0.22\,\mu m$ thick were irradiated in various crystal directions with 60-MeV iodine ions, and the number of black spots produced was counted in an electron microscope. It was found that well-channeled ions produce as little as $^1/_{15}$ the damage produced by

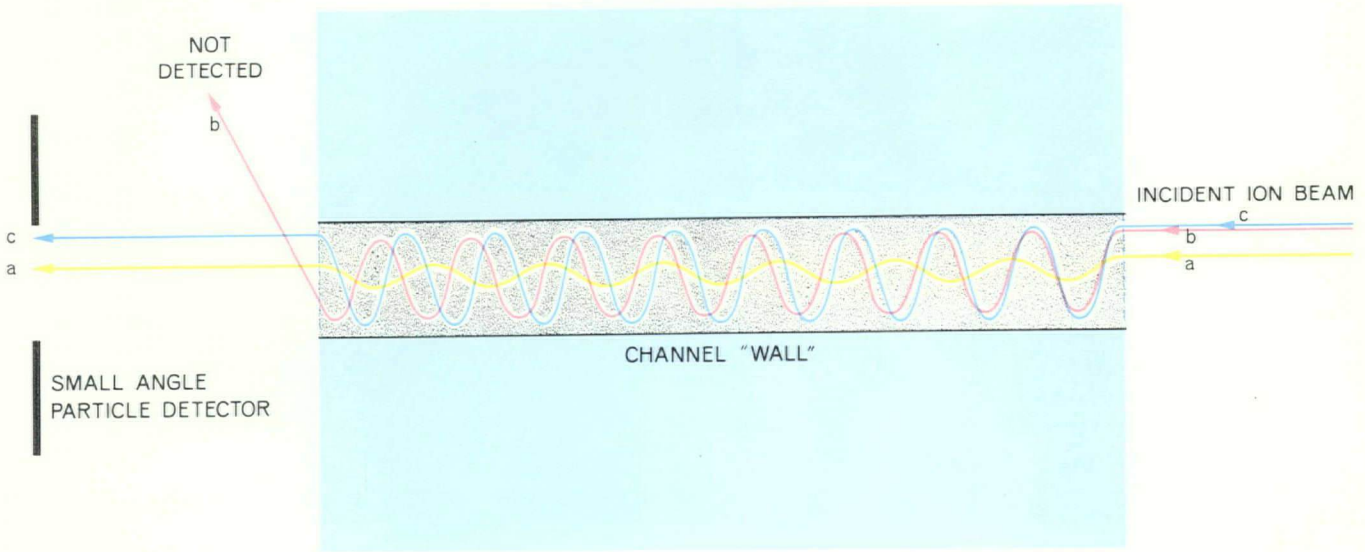



*A row of lattice atoms in a crystal. The size of each atom represents the minimum distance to which an energetic atom can approach the lattice atom. The minimum distance is achieved only in a head-on collision, in which the incident particle is strongly deflected, as at* a. *A channeled atom, on the other hand, is steered by interaction with many lattice atoms in the row and is prevented from making such close approaches, as at* b.



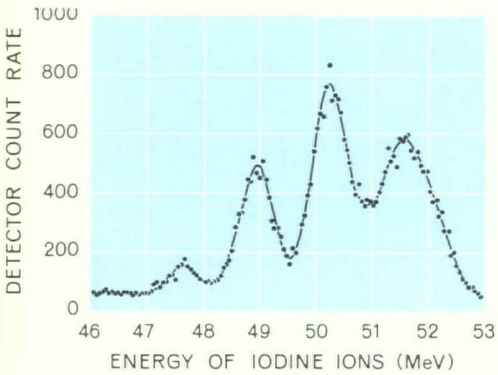

*The wide-angle Rutherford scattering of 0.5-MeV helium ions from a single crystal of zinc oxide is influenced by axial channeling. The experimental arrangement is shown at* a. *The detector has a hole drilled through its center to permit the beam of ions to reach the target crystal. Ions backscattered in the range 170 to 190° are detected. The yield of ions, normalized to a random direction, varies with the angle of incidence of the beam, being at a minimum when the beam coincides with the* x *axis of the crystal. The half-width of the dip in yield, measured to be 1.48°, represents the* critical channeling angle.

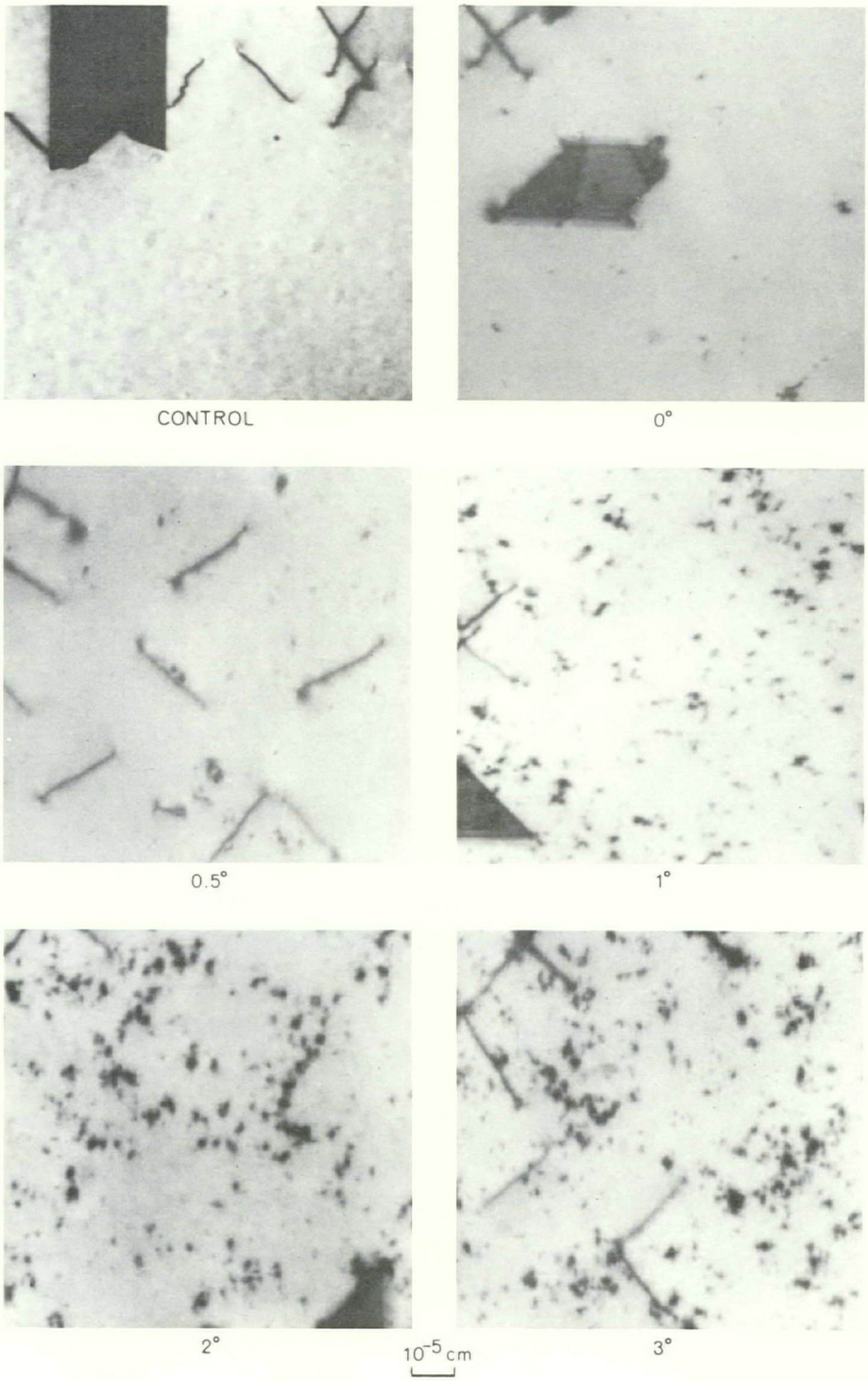
*Three typical trajectories of ions passing through a planar channel are represented by* a, b, *and* c. *Trajectory* a *makes seven oscillations while trajectory* c, *of larger amplitude, makes nine. Both of these emerge in a direction that will reach the collinear detector. Trajectory* b, *on the other hand, makes about* $8\frac{3}{4}$ *oscillations and emerges from the crystal at an angle to the channel walls. It misses the very small acceptance angle of the detector. Particles with the energy loss corresponding to this amplitude will not be found in the detected beam.*



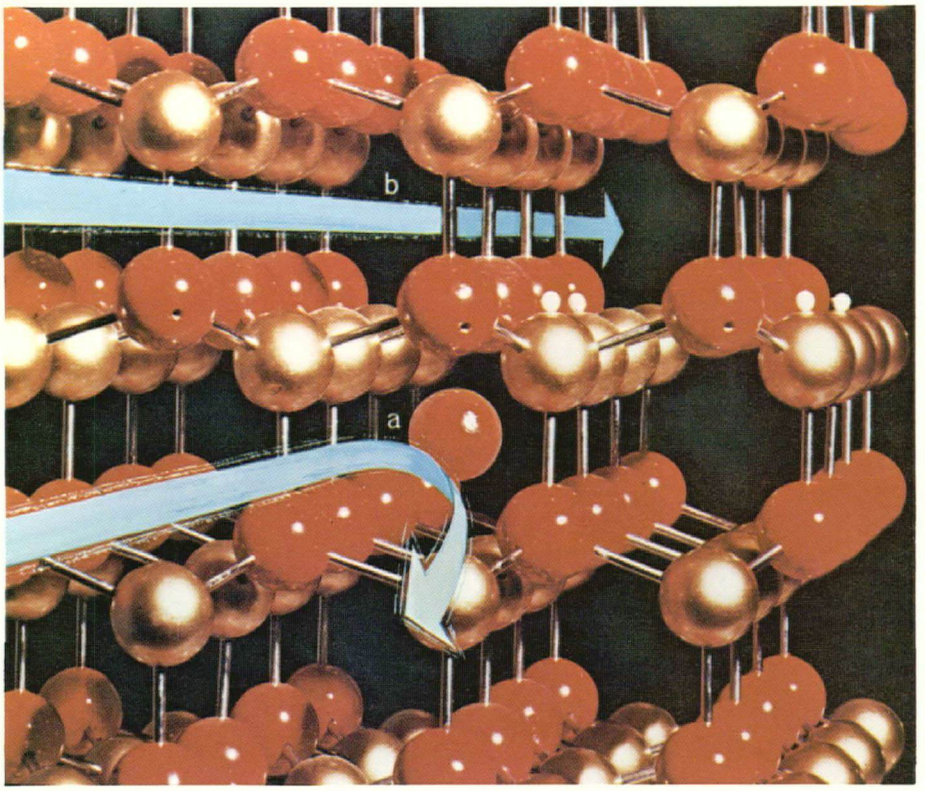
unchanneled ions, the amount of damage increasing as the angle made by the incident ions with the channel axis increases. The results of the experiments have been accounted for in some detail by extensive machine calculations, which take into account not only the lattice sites on which the target atoms are located but also that they are vibrating about these sites as a result of the temperature of the sample.

*This graph shows a typical energy loss spectrum, observed in this example for 60-MeV iodine ions passing through a 0.7-μm-thick gold crystal and traversing a {111} planar channel. The group of particles at the right-hand end of the spectrum pass through the central part of the channel and experience the smallest energy loss. The other groups, making respectively six, seven, and eight oscillations in passing through the crystal, have larger amplitudes and consequently lose more energy.*




*These are electron microscope photographs of 0.22-μm-thick gold crystals which were irradiated with 51-MeV iodine ions. The radiation damage produced in each sample is visible as small dark spots. The "control" specimen is typical of unirradiated crystals. The large dark areas and the heavy dark lines are various kinds of defects grown into the crystals and play no role in interpreting the radiation damage results. The other specimens were oriented so that the incident beam made the specified angle with the ⟨011⟩ axial channel. As the incident beam makes increasing angles with the channel axis, the amount of damage – the density of dark spots – increases in a regular manner.*




*This figure shows the arrangement of the atoms in a crystal of zinc oxide. When exposed to zinc vapor, these crystals absorb extra zinc atoms and become highly colored. If the extra zinc is located in an interstitial position, it will block a channeled helium ion, as at* a, *and increase the yield of Rutherford backscattered ions. If the extra zinc occupies a normal lattice site, the channeled ions will travel on unimpeded, as at* b.

The possibility of using ion crystallography to obtain unique information about the target crystal is shown by experiments in progress at ORNL on samples of zinc oxide containing excess zinc. The location of the extra zinc atoms in these highly colored crystals is not known with certainty. They may be located in *interstitial* positions, that is, not on ordinary lattice sites, or they may be located on ordinary zinc sites and be accompanied by some vacant oxygen sites. The Rutherford scattering of 0.5- to 2-MeV helium ions is being used in an attempt to settle this question. If the extra zinc atoms are located interstitially, they will effectively block some of the crystal channels and increase the yield of Rutherford-scattered helium ions. If they are located on ordinary lattice sites, however, they will not interfere with the channeling of the incident ions. The experimental results thus far are consistent with the latter interpretation. Similar techniques have been employed at other laboratories in studies of the crystallography of important impurities in semiconductors. They offer considerable promise for study of the details of radiation damage in solids, as well.

In the half-dozen years since its discovery at ORNL, ion crystallography has developed into a useful tool for the study of crystals and the interactions between energetic atoms. It has technological applications in the development and manufacture of improved semiconductor devices. It appears to offer a useful adjunct to more traditional methods of studying crystals, especially in areas where its unique powers can be brought to bear. These include the study of surfaces and thin films, of impurities and defects, especially near surfaces, and of radioactive substances.

# Fast Breeding and the "Alpha" of Plutonium

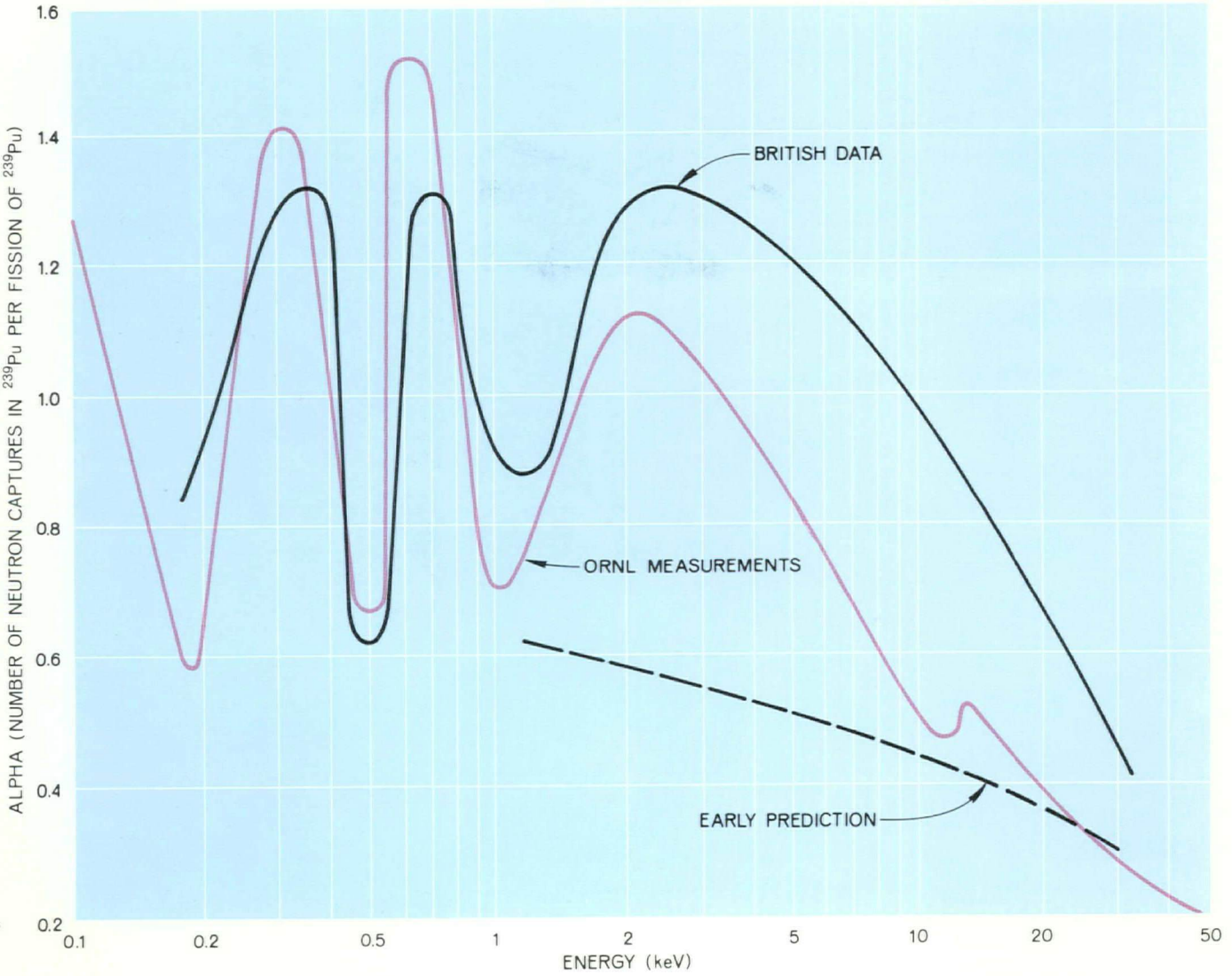
The Laboratory was able in 1968 to resolve a fundamental and even dramatic situation that appeared in some measure to threaten the Atomic Energy Commission's great program of fast breeding. Let us recall that in an ordinary nuclear reactor the rare isotope uranium-235 is burned and that extra neutrons are absorbed in the abundant uranium-238 to make (through intermediate steps) plutonium-239. Plutonium-239 is as good a fuel as uranium-235, so in fact you have "bred" some new fuel to replace the uranium-235 burned; if you are careful, you can breed more atoms of plutonium-239 than you burned of uranium-235.

The breeding process is a marginal business, and all factors have to be optimized. One of them is the energy of the neutrons; the nuclear cross sections are such that more plutonium-239 will be made if the neutrons are in the energy range of up to about 20,000 eV, as compared with 0.03 eV such as they have in the plutonium-producing Hanford reactors, which don't give a breeding gain. Thus the term "fast breeding" refers to the fact that the *neutrons* are energetic and does not mean that the breeding is fast. It isn't.

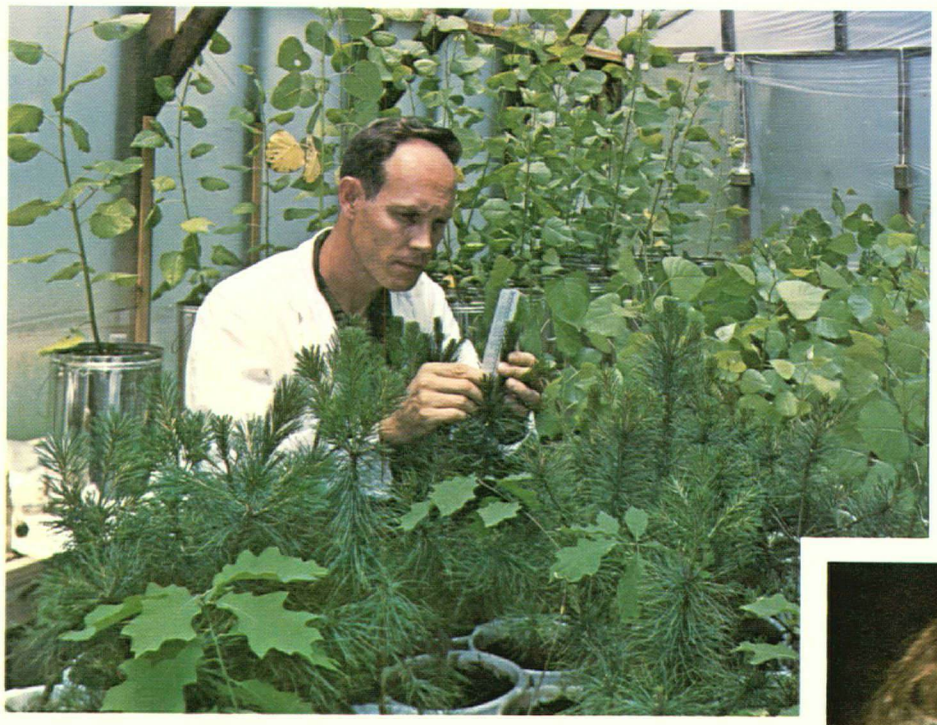
As the plutonium content builds up in the fuel elements of the reactor, neutrons, of course, interact with it. The main thing that happens is that the neutrons cause the plutonium to fission. This isn't bad, because the neutrons released in the fission contribute to the chain reaction and generate more plutonium. A lesser thing that can happen is that the neutrons merely stick with the plutonium, converting plutonium-239 to plutonium-240. If this happens, no contribution is made to the chain reaction, and a loss is suffered from the store of plutonium-239. The probability of this "parasitic capture" is expressed in a number traditionally called "alpha." Alpha varies with neutron energy, and a high value of alpha is bad for breeding.

Alpha can be measured in independent physical apparatus, but the measurements are not easy, especially in the kilovolt range of neutron energy. Early estimates placed alpha at about 0.6 of a parasitic capture per fission (see graph), but about two years ago some measurements in the United Kingdom caused a flurry of concern because they indicated a value about twice as high. This would have made breeding difficult.

An independent series of measurements were then undertaken at ORNL, and as the graph shows, they fell in between the old estimates and the British values. The situation eased. Breeding will still be difficult, but solid information exists upon which the breeding cycle can now be designed.

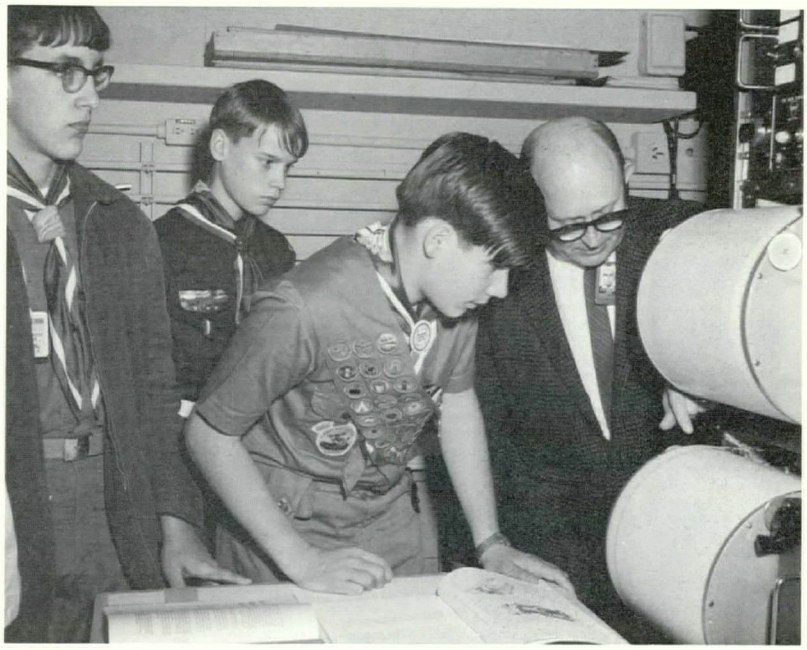


# LABORATORY LIFE.....

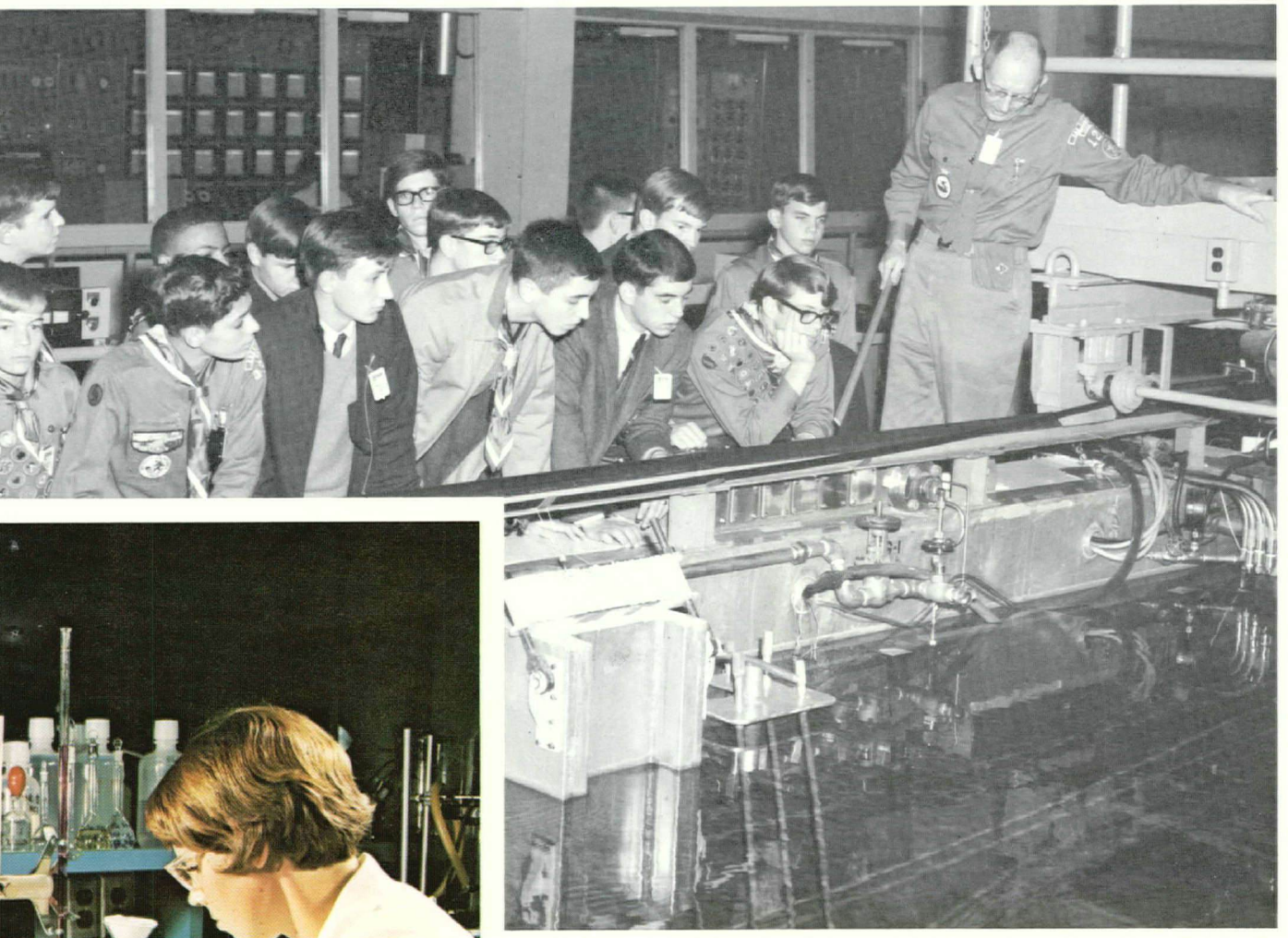


*The Health Physics Division's ecology greenhouse.*

*Summer student trainees in analytical chemistry.*



*Any earth-shaking discoveries today? Visiting Boy Scouts inspect the Health Physics Division's seismograph.*



*The Laboratory's visitors numbered 28,255 in 1968, including 1712 from 69 different countries. Here, visiting Boy Scouts inspect the Research Reactor.*

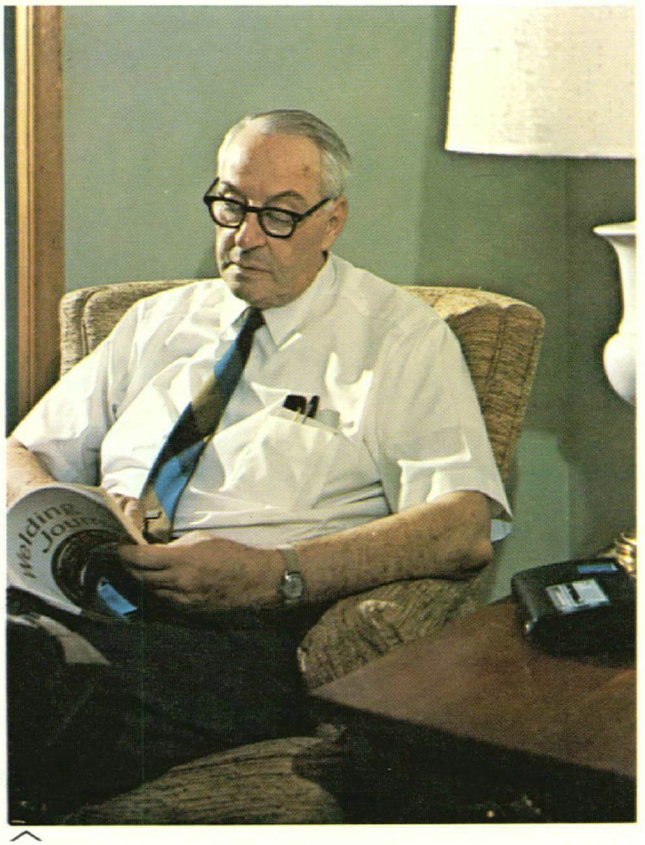
# AWARDS

*Alexander Hollaender, consultant to ORNL and a past Director of the Biology Division, was awarded the Finsen Medal of the International Congress of Photobiology. The award is named in honor of Niels Finsen, a Nobelist of 1903 in medicine and physiology, who had made early studies of the effects of sunlight upon living matter. The citation accompanying the Finsen Medal award mentioned Dr. Hollaender's early contributions in photobiology, particularly in radiation genetics. These investigations were made many years ago, but their impact was strong and had great influence on the development of the subject of radiation-induced mutations in microbes, with consequent genetic effects. The Finsen Medal joins several other national and international distinctions that have come to Dr. Hollaender.*

*At ORNL, Dr. Hollaender is recognized as the founder of the Biology Division, and his foresight is apparent in the Division's pervading interest in the fundamental processes underlying cellular biology. He and Mrs. Hollaender have long taken a leading part in the artistic activities of Oak Ridge.*

*Robert M. Nicklow of the Solid State Division was grante the Sidhu Award in 1968. The Sidhu Award is give annually to a young scientist who has made an outstandin contribution to the field of diffraction. It honors th memory of Dr. Surain Sidhu, one of the founders of th Pittsburgh Diffraction Conference, who made numerou contributions to the fields of x-ray diffraction and neutro diffraction.*

*Dr. Nicklow received both undergraduate and graduat training at the Georgia Institute of Technology, and earne his Ph.D. in physics in 1964. He has been at ORNL sinc December 1963 and has specialized in a study of th vibrations set up among the atoms of a crystal lattice whe slow neutrons bounce against individual atoms. The ene gies of such vibrations are studied by looking at the energ lost by the scattered neutrons. Dr. Nicklow's particula interest has centered recently on the effects introduced b the presence of substitutional impurities in the lattice. A the Pittsburgh Diffraction Conference on the occasion o the Award, he delivered an address entitled "Investigatio of Lattice Dynamics by Coherent Inelastic Neutron Scatte ing."*

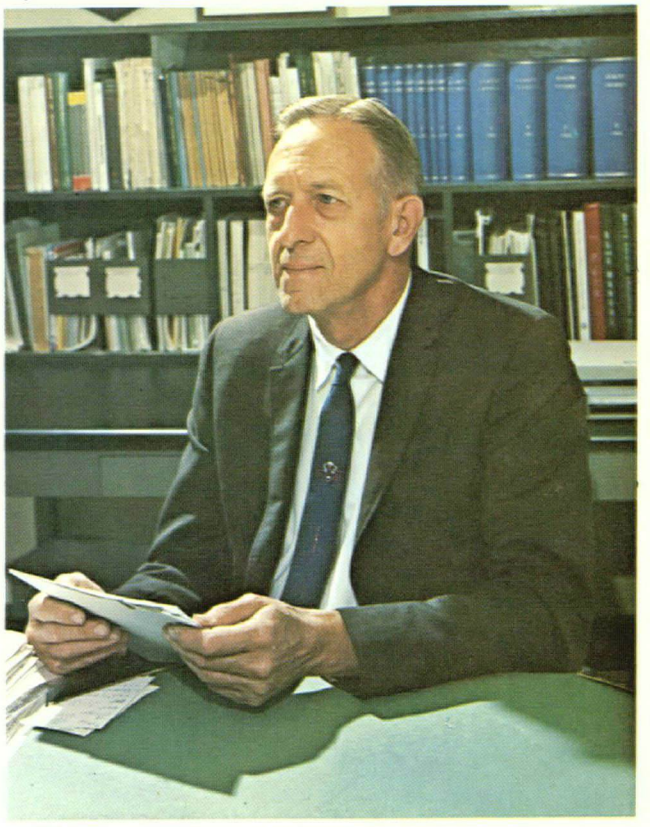


*Edward C. Miller, ORNL Superintendent of Inspection Engineering, received the 1968 Samuel Wylie Miller Memorial Award (the names are coincidental), one of the highest honors of the American Welding Society, given annually since 1927 to individuals who have "contributed conspicuously to the advancement of the art of welding and cutting." Mr. Miller has served the Welding Society for some ten years as Director, as Vice President, as President in 1967–68, and as chairman of its Technical, Standardization, and Educational Activities Committees.*

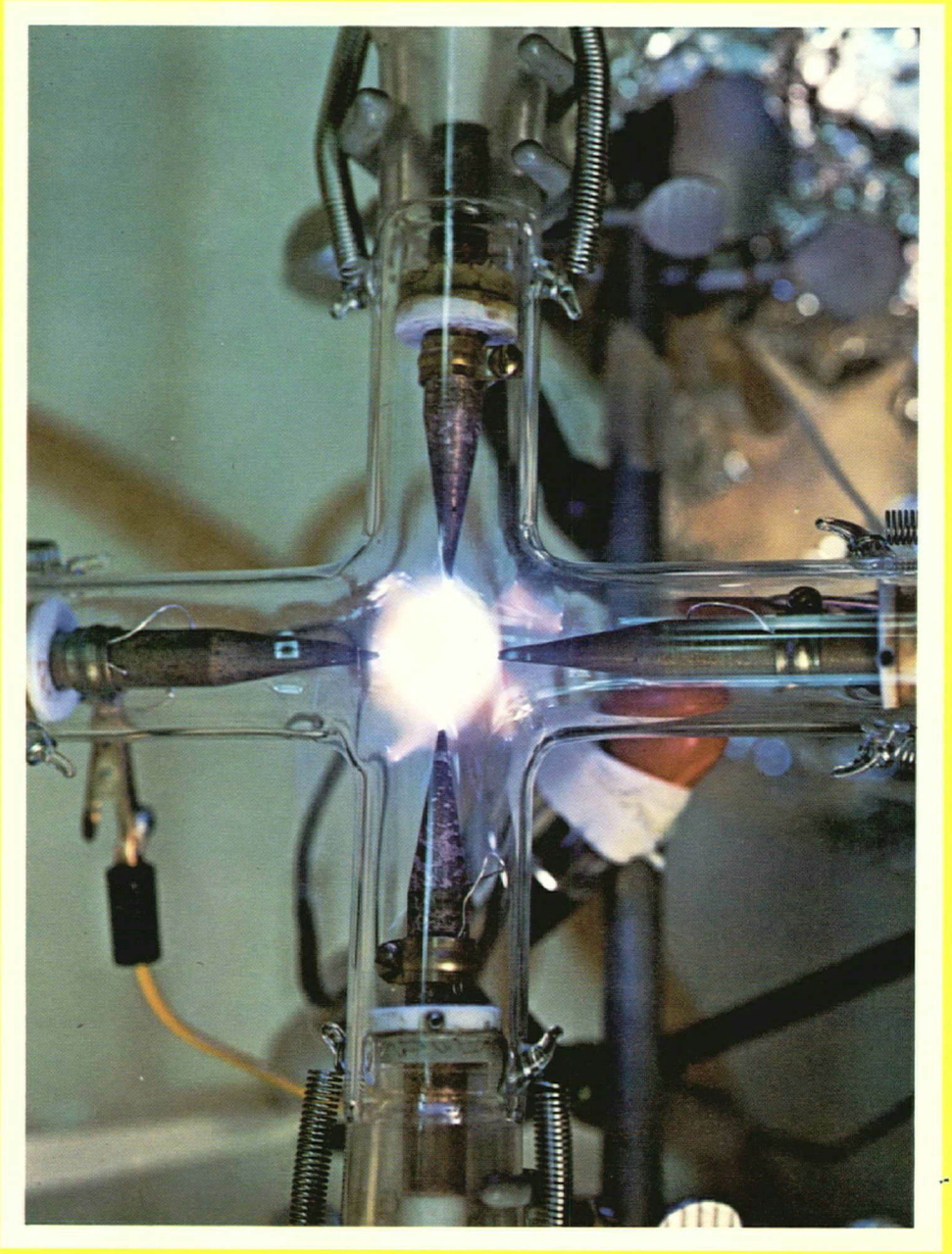
*Mr. Miller joined ORNL in 1948, from teaching assignments in the Metallurgical Engineering Departments of Purdue and Wayne State Universities. He holds degrees in metallurgical engineering from the University of Missouri School of Mines and from the University of Idaho. His present responsibilities as head of Inspection Engineering at ORNL date from 1955. He is the U.S. Delegate to the International Standards Organization group which is writing an International Reactor Vessel Code, and has been U.S. representative to several Assemblies of the International Institute of Welding and of the World Metallurgical Congress. In 1962, he was one of a five-man team representing the American Welding Society and the U.S. State Department on an exchange tour of the welding industry of the U.S.S.R. He is a member of the ASME Boiler and Pressure Vessel Code's Main Committee and of its Nuclear Vessel Subcommittee.*

*Karl Z. Morgan was honored recently as the first recipient of the new Distinguished Service Award of the Western Pennsylvania Chapter of the Health Physics Society. The accompanying citation reads in part, "in recognition of his outstanding leadership and scientific contributions in establishing and promoting the art and science of radiation protection as an important profession in the nuclear age."*

*Dr. Morgan joined the Metallurgical Laboratory at the University of Chicago in 1943, and became a member of the original Health Physics Group headed by Dr. E. O. Wollan. Later that same year he came to the Clinton Laboratories, and has served as Director of the Health Physics Division since that time. Under his directorship, health physics matured markedly as a science, and the Health Physics Division at ORNL has established an outstanding reputation for research and education, as well as for its service and vigilance in protecting the Laboratory's population against latent hazards. Dr. Morgan organized the Health Physics Society and served as its first president; he also established the journal* Health Physics *and has served as its editor since its inception. In 1962 he was the recipient of the first meritorious Gold Medal Award of the Royal Academy of Science of Sweden for work in radiation protection. He is now the first president of the International Radiation Protection Association, and has for years been active in the establishment of dose criteria in the framework of international committees.*



# Materials Research



# Voids in Irradiated Metals

A development of probably far-reaching importance in the materials science aspect of reactor engineering came to light last year. It occurred upon examination of structural metals that had been irradiated in reactors for unprecedentedly large "fluences," that is, for large integrated fast-neutron doses. Samples became available that had been irradiated in the Experimental Breeder Reactor and the High Flux Isotope Reactor to fluences of $10^{23}$ neutrons/cm$^2$. The most revealing information from these samples came as a result of an elegant exercise in electron microscopy.

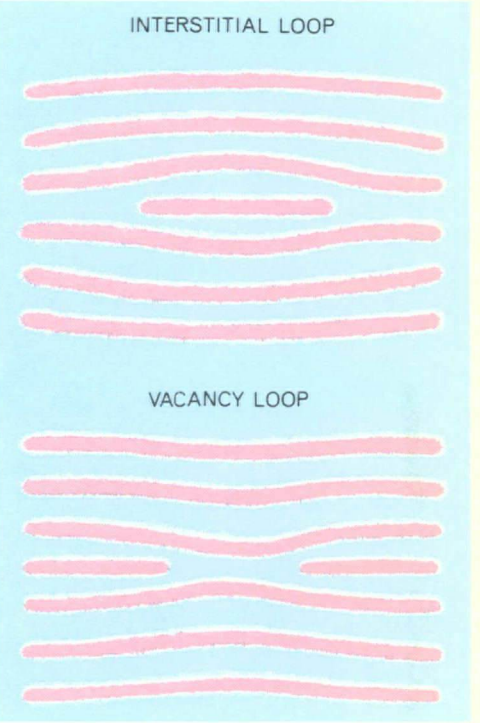
## Damage Centers: Clustering and Coalescence

Since the early days of reactor development, the displacement of individual atoms from their lattice sites in solid materials has been recognized as an immediate result of fast-neutron collisions. Such atoms lodge in new locations which are usually not lattice positions and are called "interstitials." The holes that they leave behind are called "vacancies." The electron microscope shows what happens next: in unannealed metals, like gathers like; the interstitials cluster into flat aggregates between lattice planes, and the vacancies do likewise. The sizes and distributions of the defect clusters appear to depend upon the material and the irradiation conditions, but the clustering of defects is a common occurrence in neutron-irradiated materials.

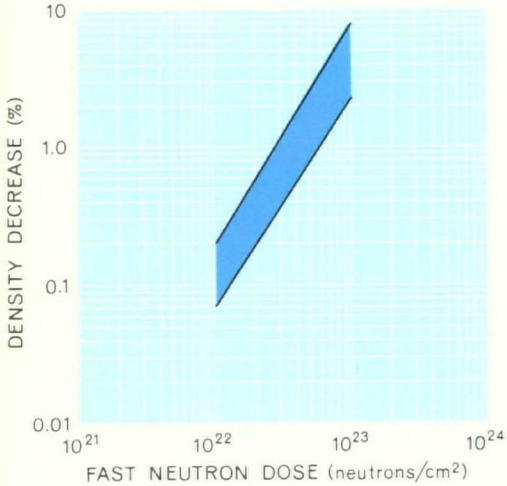
The 1968 activities at Oak Ridge centered about a study of the effect upon these clusters of the temperature of the sample during irradiation, with the surprising result that some new and unexpected defect configurations have been found at the higher temperatures and fluences. A major effort is now devoted to the definition and characterization of radiation damage processes in the intermediate- to high-temperature range.

## Voids

The most significant change in the damage structure occurs between about 0.3 and 0.5 times the absolute melting temperature; in this range




*When solid materials are irradiated, atoms are displaced by fast-neutron collisions. At low irradiation temperatures these interstitial atoms collect into platelets between close-packed planes of the crystal lattice (upper diagram). The vacant lattice sites also collect on a close-packed plane (lower diagram). In these diagrams you are looking sidewise at lattice planes. If you looked downward on the planes of the upper diagram, you would see the island of interstitials surrounded by a region of lower density, where the lattice planes are strained apart; that is, you would see a loop (interstitial dislocation loop). In a similar view the vacancies in the lower diagram would also be seen as a loop (vacancy dislocation loop) where the adjacent planes collapse inward.*




*Swelling of irradiated materials occurs when vacancies collect into voids. The graph shows data obtained at several laboratories in the United States and Great Britain for stainless steel irradiated at temperatures between 350 and 600°C. The swelling depends slightly on the temperature within this range, but the results fall within the scatter band shown. The swelling is clearly not directly proportional to the fast-neutron fluence (which would give a 45° slope on the graph) but rather varies as a higher power of the fluence.*

the vacancies aggregate into three-dimensional voids in addition to the two-dimensional dislocation loops observed at lower temperatures. This results in an appreciable expansion or swelling of the material. At low temperatures the contraction around a vacancy loop largely compensates for the expansion around an interstitial loop, and little overall volume change occurs. This is not the case if the vacancies collect into voids, and volume changes of several percent have already been observed. Design fluences for some components of liquid-metal fast breeder reactors are in excess of $10^{23}$ neutrons/cm$^2$; at this fluence the swelling may exceed 10%.

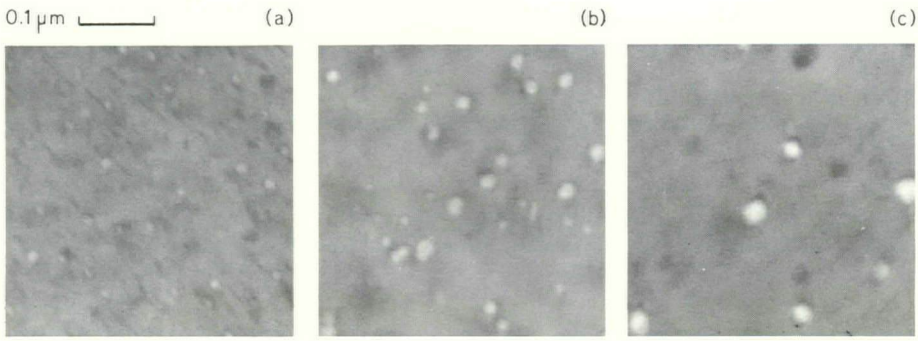
Within the temperature range in which swelling occurs, if the irradiation temperature is raised the average void size increases and the concentration decreases. These factors largely compensate each other, so that the swelling is only weakly dependent on irradiation temperature.

There are strong differences in the susceptibilities of different materials to void formation. Stainless steel is one of the most resistant observed to date. In general, higher fluences are necessary to produce observable voids in alloys than in pure metals. For example, the voids formed in 99.9999% aluminum by a fluence of $4 \times 10^{20}$ neutrons/cm$^2$ are comparable in size and concentration with those formed in stainless steel by a fluence of approximately $1 \times 10^{22}$ neutrons/cm$^2$. In addition, no voids are visible in aluminum of only 99% purity after irradiation to $4 \times 10^{20}$ neutrons/cm$^2$.

The distribution of voids in the high-purity aluminum is also inhomogeneous. ORNL studies of aluminum suggest strongly that impurities in the aluminum influence the amount and nature of radiation damage. The function of certain impurities, particularly dissolved gases, is now under extensive investigation.

## . . . And Transmutations

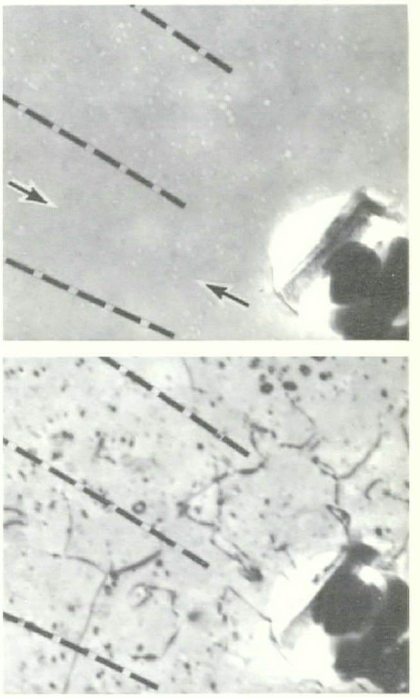
In addition to displacing atoms, a neutron may be captured by a nucleus, transmuting the nucleus to a different chemical species. Cross sections for transmutation reactions are generally small, and this possibility has received little attention in the past. However, the development of higher-flux reactors has again focused attention on the problem, especially for permanent components.




*An increase in the irradiation temperature of stainless steel decreases the void concentration but increases the average void size. The changes are depicted in these transmission electron micrographs, in which the voids appear as white geometrical spots.* (a) *$0.8 \times 10^{22}$ neutrons/cm$^2$ at 370°C;* (b) *$1.4 \times 10^{22}$ neutrons/cm$^2$ at 430°C;* (c) *$0.9 \times 10^{22}$ neutrons/cm$^2$ at 470°C.*

Aluminum is particularly vulnerable to damage by the formation of transmutation products, since it has relatively high cross sections for the formation of silicon by a thermal-neutron reaction and of hydrogen and helium by fast-neutron reactions. Exposure of aluminum to $10^{22}$ thermal neutrons/cm$^2$ will produce about 0.5% silicon, and exposure to $10^{22}$ fast neutrons/cm$^2$ will produce about $10^{-4}$ atom fraction of hydrogen and $6 \times 10^{-6}$ atom fraction of helium.

Electron microscopy of aluminum alloy pieces taken from the High Flux Isotope Reactor and the Oak Ridge Research Reactor has demonstrated that, in addition to displacement damage, solid as well as gaseous precipitates develop. The mechanical properties of this material reflect the microstructure. Strengthening is greatest at low test temperatures, at which voids and precipitate particles are most closely spaced. It decreases continuously with increasing test temperature, eventually reaching the value for unirradiated material. The ductility in a tensile test goes through a minimum at a test temperature of about 250°C, where void annealing occurs during the test. Most rapid annealing occurs along grain boundaries and may produce soft regions in the material where deformation is localized, leading to premature failure. Although the strength is recovered by annealing, ductility is not, possibly because the gas bubbles grow under stress to form crack nuclei.



0.5 μm

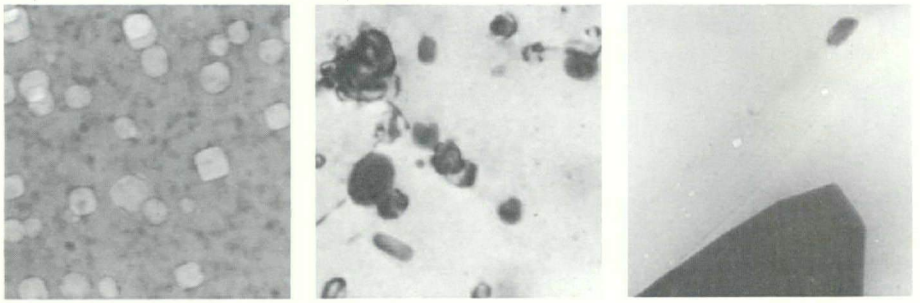
*Voids and dislocation loops are distributed inhomogeneously in aluminum of 99.9999% purity exposed to $4 \times 10^{20}$ neutrons/cm$^2$ at 50°C. The upper electron micrograph was taken with the specimen oriented to show voids; the lower one was taken of the same area with the specimen oriented to show dislocation loops. In the upper figure a higher concentration of smaller voids is present around the precipitate particle in the lower right-hand corner than in areas away from it; in addition, the band running between the arrows is relatively free of voids. In the lower figure, larger dislocation loops are present in the channels.*

## Where Do We Stand?

These effects within structural materials impose new constraints upon the design of advanced reactors. More research will be needed to define the materials and the temperature ranges that can be used in locations where dimensional stability, strength, and ductility are going to be important.

*Electron micrographs of grade 1100 aluminum (99%) irradiated in the ORR to $2.8 \times 10^{22}$ thermal neutrons/cm$^2$ and $1.5 \times 10^{22}$ fast neutrons/cm$^2$ at 45°C. The as-irradiated material (left) shows cubic voids and also tiny, faint dark spots, believed to be particles of silicon formed by a nuclear transmutation reaction. If the aluminum is annealed at 315°C for 20 h (center), the voids shrink and disappear, and the precipitate particles coarsen. These particles have been identified as silicon by means of electron diffraction. Further annealing of the material at 455°C for 20 h (right) results in additional coarsening of the precipitate particles and in the development of small gas bubbles, mainly at grain boundaries and at precipitate-matrix interfaces.*

0.1 μm 0.1 μm 0.5 μm



# Biological Sciences

*Matchstick lichens,* Cladonia cristatella, *growing on the surface of a log.* ×4.

# Mutagenesis

## The Radiation and Chemical Mutagenesis Programs

The Biology Division has for years been a center for the study of radiation-induced hereditary changes or mutations. The hereditary units, the genes, are arranged in linear order in the chromosome. Radiation can either change individual genes (gene mutation) or break chromosomes.

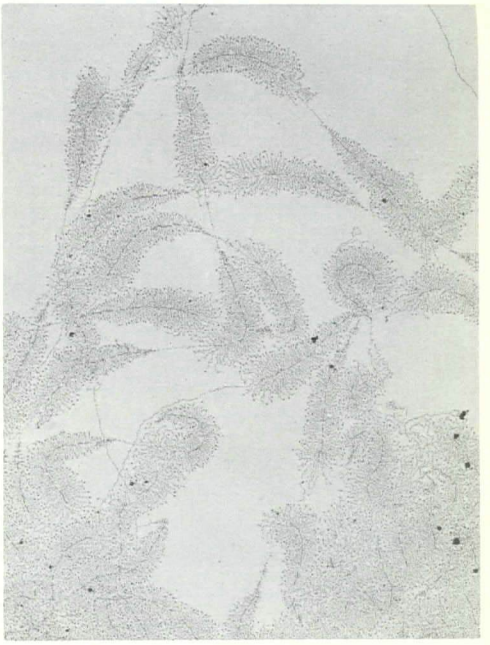
Mutations can cause cell death and other harmful effects in somatic (body) cells of the irradiated individual. If chromosome breakage or gene mutations occur in the reproductive cells, harmful effects or even death may result in the next or later generations. Such potential damage to later generations has been a major factor in limiting radiation levels when large numbers of people would be exposed.

The mouse genetics program in the Biology Division has concentrated on obtaining accurate mutation rates for ionizing radiation and in finding out how these rates are influenced by various factors. These studies have furnished important data which have been used in setting maximum permissible limits for radiation exposures.

Radiation is not, however, the only man-made cause of mutations. Some of the increasingly large number of chemicals that man is adding to his environment are also mutagenic. Like radiation, such chemicals can be of great benefit to mankind provided they are used in such a way and at such levels that the beneficial effects sufficiently outweigh any possible harmful effects. The Food and Drug Administration requires that chemicals be tested for toxicity, developmental effects, and cancer induction on various experimental animals. No requirement exists for testing for genetic effects that could be transmitted to future generations. Partly this is because techniques for carrying out such tests have not been sufficiently developed, and one of the Biology Division's goals is to develop such tests.

Various investigators in the Division have been comparing spontaneous, radiation-induced, and chemically induced mutations to learn more about the mechanism of mutation induction. Because some of these chemicals react with deoxyribonucleic acid in highly specific ways and produce only particular types of genetic alterations, they offer exciting new tools for obtaining fundamental information on the mechanisms of mutagenesis. Under the joint sponsorship of the AEC and

*This is the first electron microscope photograph of active genes with known function. Each of the fern-like sections is a gene. The central stem of the fern is the gene itself, consisting of a deoxyribonucleic acid strand which acts as a template for the gene product. The fronds are successively longer pieces of ribonucleic acid which are being synthesized along the length of the gene. As each ribonucleic acid molecule is made, it is immediately coated with protein in a manner that prevents it from extending its full length. Presumably, identical genes repeat many times along the same deoxyribonucleic acid molecule. These genes, which were isolated from a salamander oocyte, carry the code for ribosomal ribonucleic acid – a major component of small particles called ribosomes, which are essential elements of the protein-synthesizing machinery of all living cells.*



*Chromosomes of a wallaby, a primitive Australian mammal. The Biology Division recently acquired a colony of these animals from Australia to study the breakage of chromosomes by radiation and chemicals. Because of their small number and large size these chromosomes are more suitable for quantitative study of chromosome breakage than the chromosomes of higher mammals, including man, which are more numerous and smaller.*

*Wallaby chromosomes from cultured blood cells irradiated outside the body. There is one rearranged chromosome (double arrow) consisting of two broken chromosomes joined end to end, with the resulting fragment nearby (arrow).*

the National Cancer Institute, studies are also in progress on the relation between carcinogenesis and mutagenesis. The Biology Division has also started investigating chemical mutagenesis in the reproductive cells of mice under joint sponsorship of the AEC and the National Institute of General Medical Sciences. Finally, the question has been raised whether irradiation of foods for preservation might convert some substances in the foods to mutagens, and the Division has initiated investigations of this matter.

The result has been the development within the Biology Division of multiple and yet coordinated programs on chemical mutagenesis designed to expand the knowledge and refine and develop techniques needed to answer such questions. Some of the major advances in these programs during the past year are summarized here.

## Mutagenicity of Chemical Compounds

Two of the major questions which must be answered if a meaningful way is to be found to test chemicals for production of mutations are these: (1) Is there a single test organism that can be depended upon and (2) do substances belonging to the same class of chemical compounds act the same way? If the answers were affirmative, the problem of testing would be simplified. One might be able to use a single test organism and to select a single compound from each of the different classes of compounds and adopt the techniques developed for radiation mutagenesis studies. Tests on mutations in the reproductive cells of the mouse during the last year, as well as other tests for mutagenicity, show that neither question can be answered completely affirmatively. Some of the compounds which are potent mutagens in microorganisms and in mammalian tissue culture did not show any mutagenicity in the mouse test. Strains of mice differ in their response to chemical mutagens, and compounds belonging to the same chemical class differ in their action. For example, the mutagens and alkylating agents methyl methanesulfonate (MMS) and ethyl methanesulfonate (EMS) induced a high frequency of dominant lethal mutations in females of one strain, whereas they induced only a low frequency in another. Similar differences do not seem to occur when male mice are treated. The three closely related alkylating agents MMS, EMS, and PMS (propyl methanesulfonate) act very much alike in that late stages in sperm formation are more sensitive to induction of dominant lethal mutations. IMS (isopropyl methanesulfonate), however, appears to have its largest effect on earlier stages. The complexities revealed by these experiments emphasize the importance of going beyond screening tests of mutagens to investigating the basic mechanisms of mutagenesis.

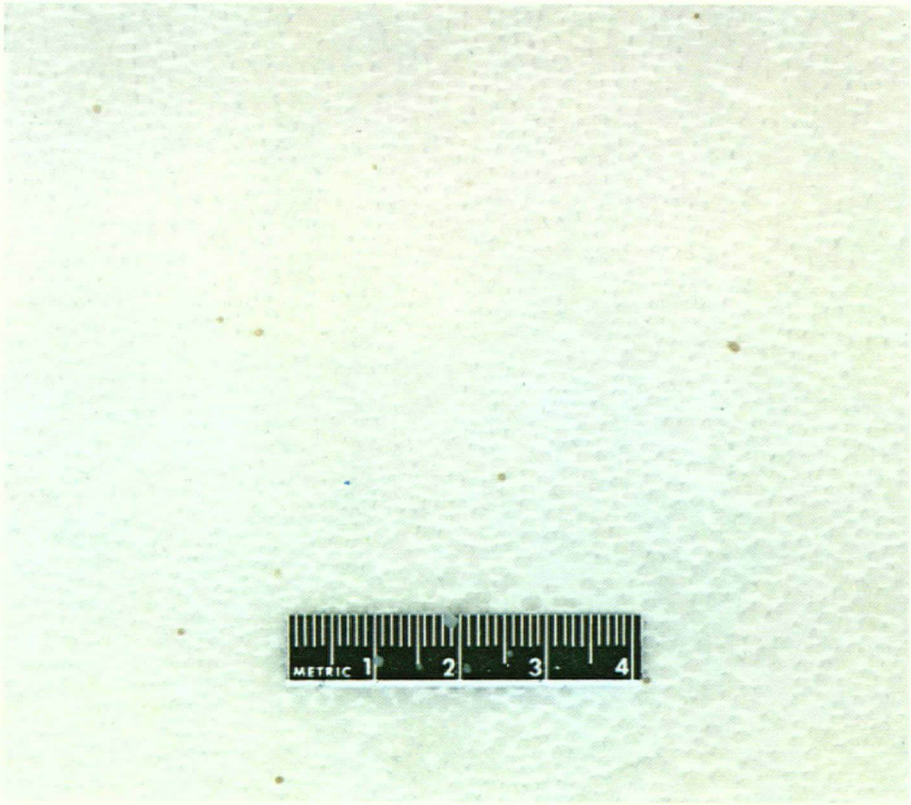
Another major question is, what happens to compounds when they are taken into the body? Are some potential mutagens destroyed before they can reach the germ cells? Are some nonmutagenic compounds converted in the body to mutagenic metabolites? Two approaches to answer these questions have been developed within the Division during the last year. In the first, radioactively labeled compounds are injected

*Examination of stained animal tissues mounted as lantern slides. A variety of structural alterations, ranging from malignant tumors to damaged cells and organs, can be readily identified in these slides. For example, the slide at the extreme left of the second row from the bottom shows the structural changes in the walls of an artery and a vein from an aging animal.*

into the mouse, and their distribution to the tissues and organs, especially the reproductive organs, is followed over the course of time. The reactions of the compounds with the major macromolecular cell constituents, the nucleic acids and proteins, are also followed. Studies with MMS and EMS labeled in the alkyl group have shown that they are distributed throughout the body quite rapidly and alkylate both the nucleic acids and proteins of the reproductive organs. MMS is broken down and excreted less rapidly than EMS, alkylates the tissue proteins and nucleic acids more effectively, and produces more mutations. Thus some of the differences between compounds and between strains of mice may result from differences in the way in which compounds are handled by the body. The use of such radioactive labeling is obviously desirable whenever a pure compound is being studied.

## The Host-Mediated Assay Technique ("Moldy Mouse")

The detection of mutagenic activity is made more difficult by the fact that it may be one or more of the metabolic breakdown products that exhibit mutagenic activity, rather than the compound itself. To cope with this problem the Division is working with what is called a




*Mutant colonies of the bread mold,* Neurospora. *The mutant colonies are small purplish-red objects scattered in a background of normal white colonies. The mutant colonies can be counted and picked out for further testing.*

host-mediated assay. This assay was developed originally by investigators at the Food and Drug Administration.

In the host-mediated assay, microorganisms are injected into the peritoneal cavity of a mouse or other mammal during the treatment with a chemical mutagen. There these microorganisms are exposed not only to the original compound but also to its metabolic breakdown products. The microorganisms are then removed after several hours' or days' incubation in the mammal and assayed for the presence of mutations. In the Biology Division we are trying to use different mammals as hosts and to study mutagenesis in specific organs such as the testes and liver. We are also making use of an exceptionally informative assay system developed with the bread mold *Neurospora.* Previous work in the Division has made this test system one of the best assays for the induction of mutation.

The spores of this mold can be treated with mutagens and then put into growth medium. Each spore grows into a small round white ball of mold. Mutations in two specific genes block the synthesis of adenine in such a way that a purple intermediate compound accumulates. The mutants can then be detected as purple balls or colonies in among the white ones. The method also allows a subtle distinction between different kinds of mutations, such as between those that are a change in the gene itself and those that are a loss of a section of chromosome including not only the gene in question but several others as well. Thus this test system can supply much valuable information about the kind of mutational process that has occurred.

The *Neurospora* assay system has been injected into both mice and rats to develop new host-mediated assay systems during the past year. Preliminary work with these systems has proved quite successful. Spores of the mold have been injected into the body cavity or organs and recovered in viable condition some hours or days later. Mutations have been induced in these spores when known mutagens were injected. The preliminary work to validate the system is being completed, and it will then be ready to use as a test system for compounds with unknown or suspected mutagenic activity.

This new host-mediated assay system has the advantage that it is a quicker, easier test to use than the usual breeding experiments with mice and that it should be usable even in mammals that are not suitable for direct genetic work. It is clear, however, that it cannot replace direct work with mouse reproductive cells, since there are undoubtedly features of the reproductive cells that cannot be mimicked in an introduced microorganism.

## Mutagenesis in Cultured Mammalian Cells

Another system for studying mutation, which employs cultured mammalian cells, has been developed during the year and has great promise. A number of investigators have attempted to develop adequate systems for studying mutation in mammalian cells in tissue culture. None of these, however, proved satisfactory for studying induced mutation. Within the past year, systems for this purpose have been developed independently and simultaneously in the Biology Division and at the University of Colorado.

The system developed at Oak Ridge employs Chinese hamster cells. The cells are treated with a mutagen and spread out on a selective medium that allows only mutant cells to multiply. Each mutant cell forms a small colony of cells that can be readily seen. Selective methods were developed for mutations concerned with the cell's resistance to the drug 8-azaguanine and with the cell's ability to synthesize the amino acid L-glutamine.

Mutations have been induced with a number of mutagens whose action is already well known in microorganisms. In this way, it will be possible to find out whether hamster cells mutate by the same mechanisms as do microorganisms. If so, then the large body of data on mutation induction in microorganisms can be carried over to mammalian cells. Now that the initial difficulties have been overcome with hamster cells, it should be possible to extend the method to other mammalian cells, including human cells. Thus direct tests for mutations should become possible with human cells.

*Giant cell formed after irradiation of a mutant strain of the bacterium* Escherichia coli *(approximately 50 μ from breast to tail feathers).*

## Mutagenesis and Carcinogenesis

There are a number of theories about how chemicals induce cancer. One of these, which has had considerable popularity but is by no means

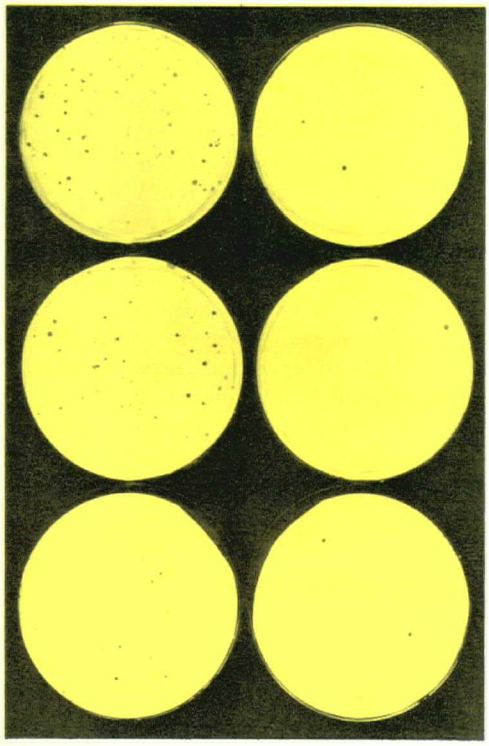
universally accepted, is that chemicals cause mutations that convert normal cells into cancer cells. On this view any carcinogenic chemical should be a mutagen and vice versa. In fact, however, there are carcinogens which have not produced mutations, as well as mutagens that have not produced cancer, in the usual test systems. The tests for mutation induction are usually done in microorganisms or in insects, while the tests for cancer induction are done in mammals. It is known that some substances that are not carcinogenic themselves can be converted to carcinogens in the mammalian body. The same conversions might not occur in microorganisms or insects. Consequently, the tests for mutagenesis might be made with the unchanged noncarcinogenic compound instead of the true ultimate carcinogen. Indeed, such a case has been found with the bread mold system. Aminofluorene has been shown by work done elsewhere to be converted to acetyl-*N*-hydroxyacetylfluorene in the mouse, and it is this compound rather than aminofluorene that is carcinogenic. Likewise, aminofluorene does not cause mutations in the bread mold, which cannot make the conversion, but acetyl-*N*-hydroxyacetylfluorene is mutagenic. How many other cases of compounds that produce cancer but not mutations are really of this sort is not known.

With cultured mammalian cells, it is in principle possible to test for mutagenesis and carcinogenesis in the same system. Such double tests have not yet been performed, but a number of carcinogens have been shown to be mutagenic.

Another difficulty with the cancer-by-mutation hypothesis is the existence of potent mutagens which differ drastically in their carcinogenic activity. We now know that gene mutations result from a wide variety of genetic alterations and that the spectrum of genetic alteration varies with different mutagens. A characterization at the molecular level of the genetic alterations in the mutations produced by different carcinogens could provide some understanding as to whether carcinogenic activity is correlated with the production of a particular type of genetic alteration.

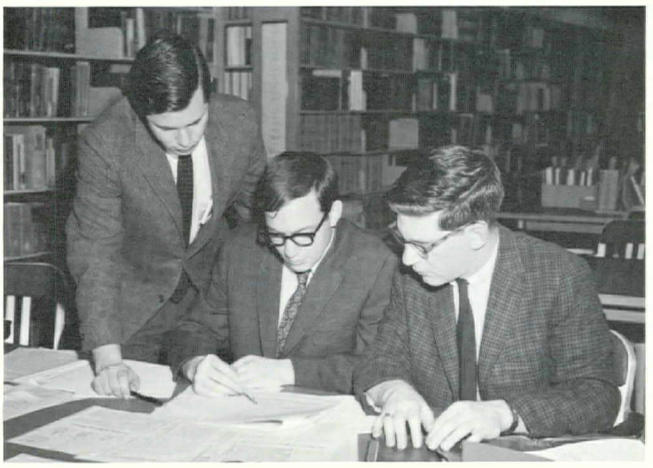
Methyl methanesulfonate (MMS) and *N*-methyl-*N'*-nitro-*N*-nitrosoguanidine (MNNG) are examples of compounds which are potent mutagens but differ in their carcinogenic activity. MMS and MNNG are both strong mutagens in microorganisms. MNNG is a potent carcinogen, whereas MMS has very weak carcinogenic activity. Characterization of the gene mutations produced by these two compounds showed that a high proportion of the gene mutations induced by MMS led to metabolically inactive gene products, whereas a high proportion of the MNNG-induced gene mutations produced gene products with altered function. How many additional compounds will show this same correlation is unknown. As a working hypothesis it seems likely that compounds that induce a high percentage of mutants in which the specific gene product has an altered function are strong carcinogens, whereas compounds that induce a high percentage of mutants in which the specific gene product is nonfunctional have only a limited carcinogenic activity.

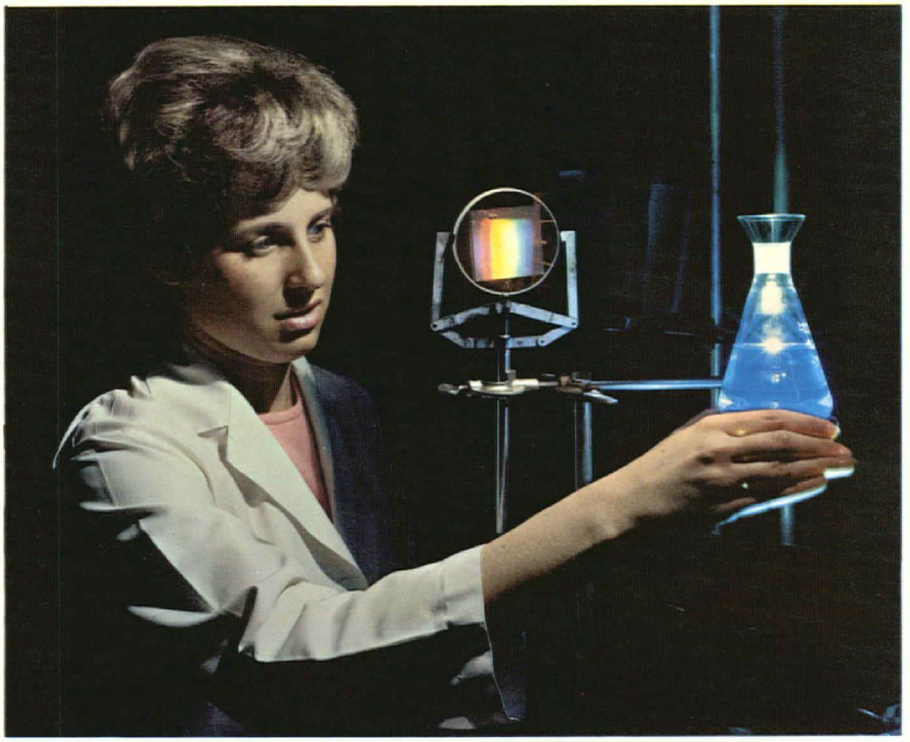
*Mutant colonies of cultured mammalian cells. Each dark spot on the surface of the six dishes is a mutant colony. The culture medium is such that only mutant colonies can grow. The cells in the dishes on the left were treated with different concentrations of the mutagen ethyl methanesulfonate; those on the right were untreated.*



# LABORATORY LIFE.....

*(Right) A student assistant fills the liquid-nitrogen trap of the Electronuclear Division's polarized-proton source while the engineer observes the plasma through a window. (Below) Visiting students from the Massachusetts Institute of Technology.*





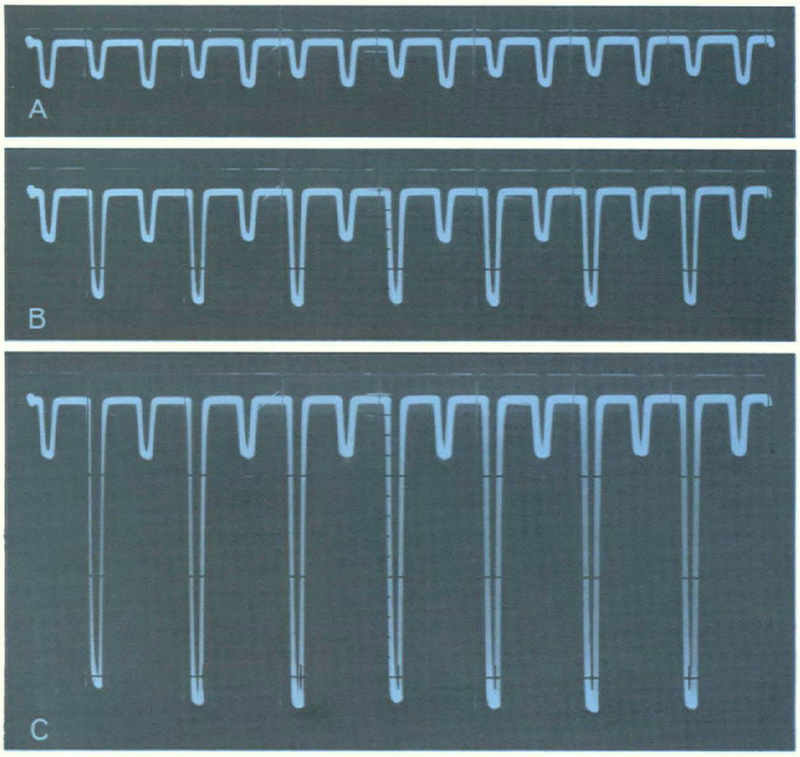
*A graduate student from the University of Tennessee working in the Health Physics Division. She holds a flask of water of extremely high purity used in measurements of the dielectric behavior of water in the vacuum ultraviolet region. Light from the diffraction grating shown strikes the water at a particular angle, and the reflectance of the water is measured. From these data are obtained the dielectric constants of the water and their dependence on the wavelength of the light.*

# Molecular Anatomy (MAN) Program

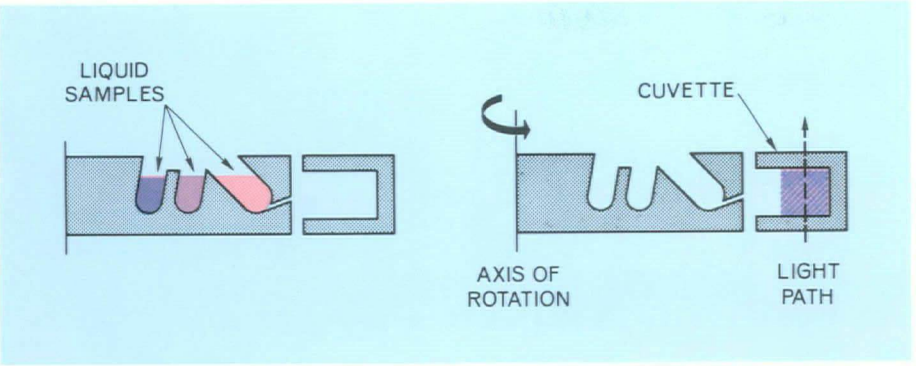
## The GeMSAEC System for Rapid Biomedical Assay

It is a commonplace to say that success brings new problems, but that is what has happened in the Molecular Anatomy Program. The zonal centrifuge and other high-resolution separation methods produce a flood of biomedical samples, each of which must be analyzed for one or more constituents. Can analytical methods now be accelerated to match?

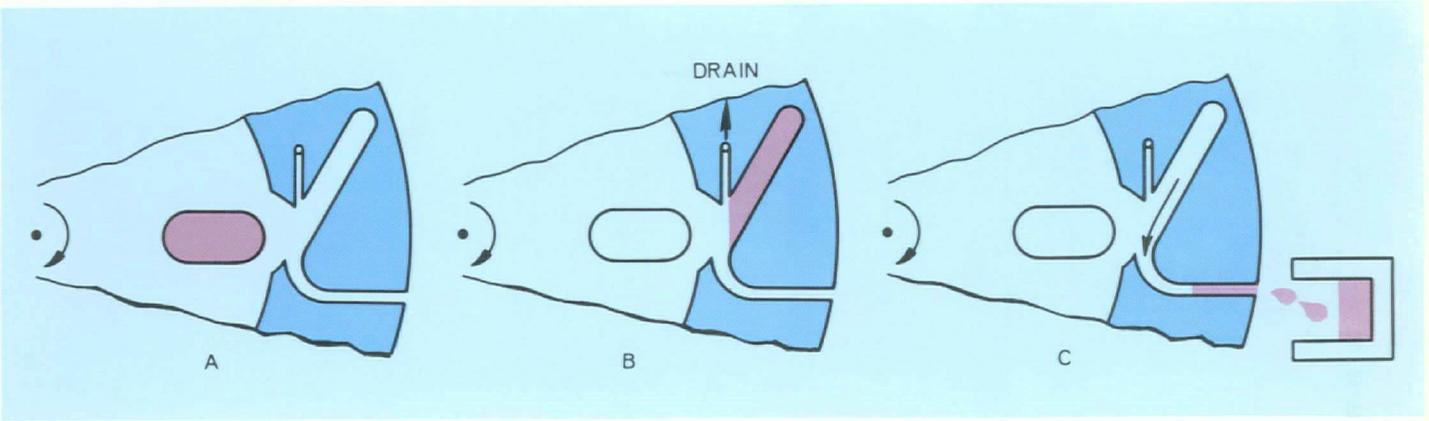
Actually this is a local aspect of a much more general problem. More and more analytical tests are demanded of the clinical laboratories of hospitals and other biomedical service groups. The rate of increase is about 15% per year, and this is a sufficient rate of exponentiation to




*Determination of the enzyme lactic dehydrogenase in the GeMSAEC fast analyzer. The oscilloscope patterns at various times (A = zero, B = 15, C = 30 min) after the start of rotation are shown. This was a test run, to assess reproducibility, and the same amount of enzyme was placed in each cuvette; consequently, the pattern is uniform. In an analysis of unknowns, the pattern would be jagged, because the reactions in the various cuvettes would proceed at different rates.*




*Principle of the GeMSAEC system. The cross section of a rotor is shown, in which liquid samples are placed in pockets (left) in a Teflon disk. When rotation starts, the liquids are mixed together in the cuvette, which spins with the rotor. The absorbance of light of a selected wavelength is used to measure the concentration of the product of the reaction.*

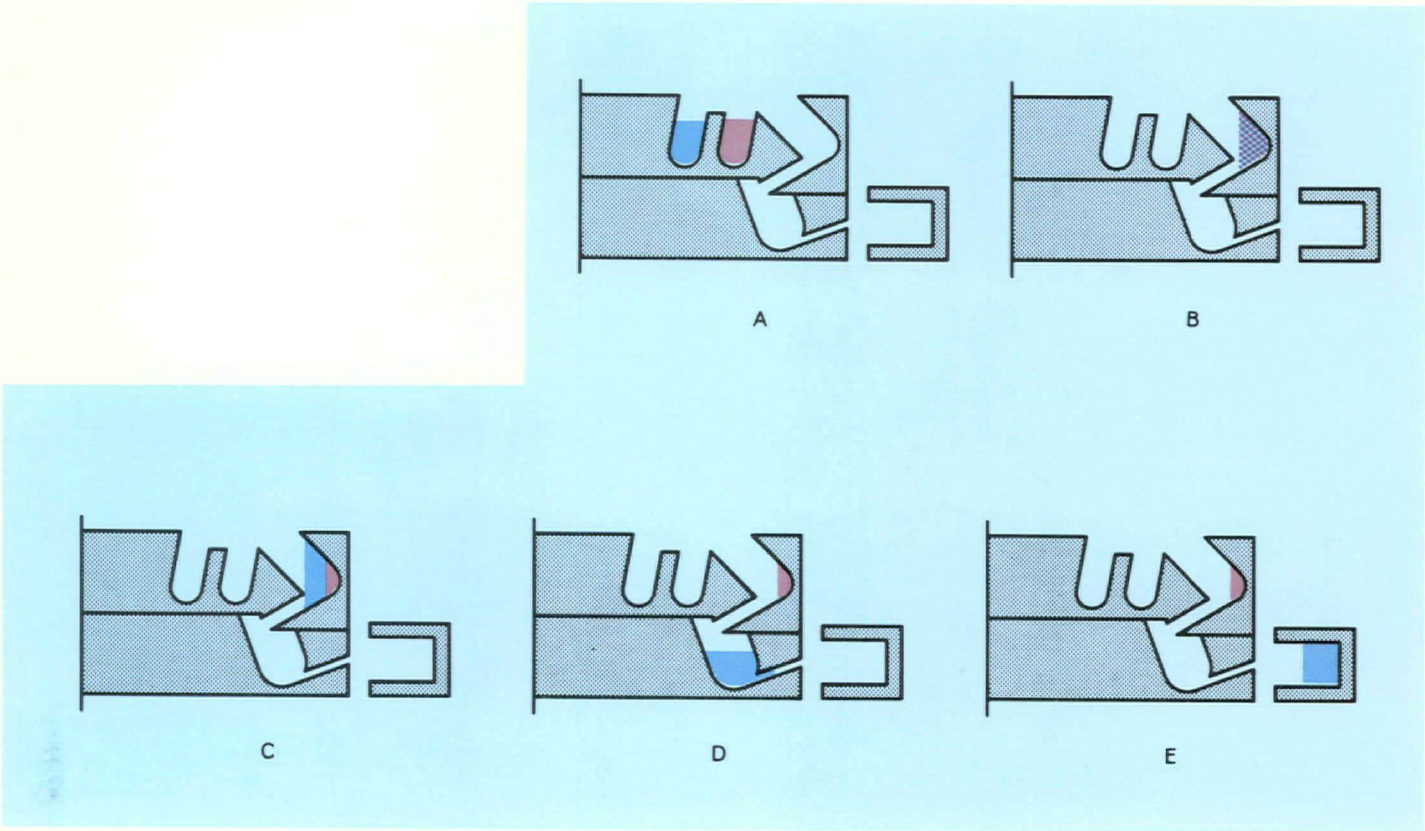



*Automatic pipetting with the GeMSAEC system. Here you are looking down on the rotor, and an unmeasured but oversufficient amount of liquid is contained in a pocket* (A). *With rotation* (B) *the liquid transfers to a slanted radial recess, and it fills it to a volume determined by the overflow. A jerk in the rotation* (C) *then transfers the measured quantity to the cuvette. If the cuvettes are arranged so as not to spill when stopped, then a second measured quantity can be added by changing Teflon disks.*

create severe problems, because manpower training, equipment development, and construction of new laboratories have not kept pace with demands.

In 1968 at least a partial answer to the problem was developed in the MAN Program. It uses centrifugal techniques whereby a number of samples can be analyzed simultaneously, or in parallel. The system has been called GeMSAEC after the two sponsoring governmental agencies, the National Institute of General Medical Sciences and the AEC.

The principle of the method can be seen in the first of the adjacent drawings. A Teflon disk about 5 in. in diameter and an inch thick has a number of recesses shaped into it, and the drawing is a cross section of one sector. In all, there may be 15 such sectors with similar recesses accurately molded into the disk. The liquid samples to be analyzed, together with one or more of the required reagents, are loaded into the pockets of the Teflon. Then the disk is set in a rotor and made to spin at a moderate speed (2000 rpm is typical). The centrifugal force overcomes the capillarity of the narrow radial channel, forcing all these samples in each sector out into the respective cuvettes, which are also rotating with the disk. The cuvettes are equipped with glass windows at top and bottom, and as they spin they pass successively over a light beam coming from a monochromator and going to a photomultiplier. The chemical reaction affects the opacity of the samples of liquid in the cuvettes, and this opacity for light of a chosen wavelength is the measure of the analysis. The signal from the photomultiplier is displayed on an oscilloscope with sweep synchronized with the rotor, so that 15 peaks or dips appear, each corresponding to a cuvette and showing by its height or




*Sedimentation and decantation. In* A *and* B, *two samples are transferred and mixed. In* C, *a precipitate centrifuges out; in* D *the liquid drains off as the rotor is brought to a stop. In the drawing, it is assumed that the sediment is hard-packed; if it is not, the pocket containing it will be shaped differently. In* E, *transfer is made to the cuvette upon restarting the rotation.*

depth the measure of the light transmittance of the sample. Once the rotor is loaded, it takes only a few seconds to get the analyses of all 15 samples, and the system would seem to lend itself to rapid recording and digital processing of the data in a small computer. Alternatively, measurement or photography of the oscilloscope trace can of course also be used.

This basic system is capable of various embellishments and elaborations. For example, to mix the liquids in the cuvettes, the rotor can be quickly slowed or accelerated; this action produces a small eddy in each cuvette. The assembly can be made self-pipetting by a scheme such as shown in the second drawing, where an overflow level determines the volume of liquid to be transferred to the cuvette, and transfer is accomplished by braking. The application of air pressure or suction can also be used to transfer liquid from one recess to another in more elaborately designed rotors, and bubbling air through the reaction mixture can be used for mixing. The scheme in the third drawing shows how a supernatant can be withdrawn from a precipitate and then be transferred to a cuvette for examination. In principle, many more than 15 sample holders and cuvettes can be built into a rotor, and it is clear too that one is not necessarily limited to room-temperature analyses. Furthermore, since the light beam can be very small, the system lends itself to the analysis of small samples – down to 0.02 ml, although the total reaction mixture volume customarily used is about 0.4 ml. Flushing and washing can be accomplished by introducing water at the axis of the rotor.

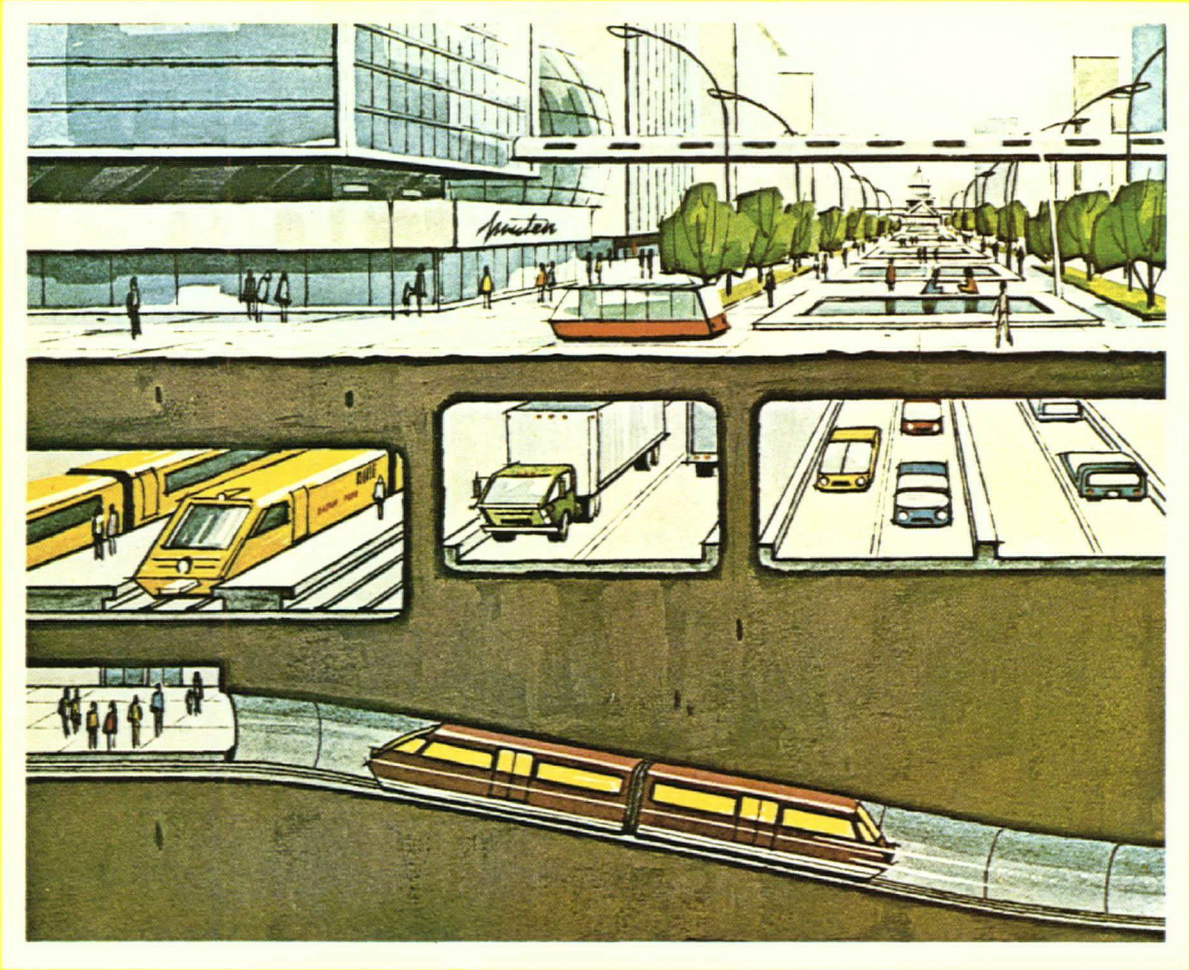
*The GeMSAEC system in the laboratory. The spinning rotor is seen at the center, with the photomultiplier tube mounted horizontally just above it. Two of the multiple-sample Teflon disks rest on the table. At left the oscilloscope displays 15 dips, each of which by its downward length measures the light transmission through one of the 15 cuvettes in the whirling rotor.*

A test of the usefulness of the GeMSAEC system for the analysis of protein in blood serum was made. Fifteen samples of serum were obtained from the Health Division and analyzed by the biuret reaction – a color-change method involving tartrate and copper sulfate. The results were compared with standard methods and found to be in good agreement. The GeMSAEC, however, proved to be faster, cheaper, and more accurate, and it used smaller samples.

For the determination of enzymes, a rate-of-reaction method is used. The unknowns and the appropriate reagents are placed in the pockets of the Teflon disk, and when the spin starts, all samples get an even start as the reactants transfer to the cuvettes. The enzymes get to work, and opacity changes with time over several minutes. The *rate* of decrease or increase in the transmitted light is accordingly the measure of the amount of enzyme present in each cuvette. For many reactions, sufficiently good data may be obtained in the first few seconds of spinning, even though the reactions may still be far from completion.

We can expect the GeMSAEC analytical system to spread quickly to determination of several more substances of clinical interest, such as serum cholesterol, uric acid, and blood glucose. Enzymatic assays that are due for development in the near future are those for creatine phosphokinase and the transaminases. If fluorescence measurements are substituted for simple light absorption, other classes of substances are expected to be adaptable to assay by GeMSAEC. The biomedical manufacturing industry is keeping a sharp eye on the developments at Oak Ridge.

# Civil Defense



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# Civil Defense Research

A wide examination of the problems of civil defense in the nuclear age has taken the Oak Ridge group into an engaging variety of topics in engineering and social behavior under conditions of stress. We briefly mention several of these in the following paragraphs.

## The Technology of Tunneling

Occasionally, and in especially happy circumstances, a common technology can serve both to protect life in wartime and to enhance life in times of peace. An opportunity of this kind offers itself in the matter of cheap excavation of tunnels. If tunnels could be dug at greater speed and lower costs they could both provide shelter for an urban population during nuclear attack and increase the beauty and convenience of city streets during normal times by making it possible to place utilities beneath the surface. System changes and repairs could then be made without surface disruption.

The Civil Defense Research Project is undertaking a study to help the Department of Housing and Urban Development in establishing a research program that would lead to faster and cheaper methods of tunneling. This study includes investigation of both well-established and new technologies. A hard-rock tunnel, for example, is conventionally constructed by the "drill, blast, and muck" method. While this cyclical approach is applicable to a wide variety of geologic conditions with a modest outlay for equipment, it is slow, and costly in terms of labor. Ten-foot-diameter tunnels have cost from $200 to $1400 per linear foot, depending on the type of rock.

On the other hand, several new and promising methods for breaking rock have appeared. One of them uses the eroding power of water moving at speeds of up to 1000 m/sec. A tunneling system can be conceived which would combine multiple revolving high-speed erosion jets playing upon the working face, with hydraulic transport of the muck that they break loose. The available data for high-velocity water jets are limited to stationary jets perpendicular to the rock face. In order to evaluate the use of high-pressure water jets for an automated system of tunneling, data are needed for traversing jets in systems where the

specific energy required for cutting rock can be measured under a variety of adjustments and parameters. One such system cuts grooves (kerfs) with the jets and breaks off large quantities of rock between adjacent slots by the sidewise thrust of the jet. Plans have been made to test jets with three diameters at 6000 psi, using a Halliburton pumper rig and a modified turntable while varying jet standoff distance, jet inclination angle (two planes), jet transverse velocity, kerf spacing, and the nature of the rock.

## Electromagnetic Pulse

Fallout, blast, and fire are effects of nuclear weapons which have received considerable publicity since the detonation of the first atomic bomb. An effect which has received virtually no publicity but could also have serious and widespread consequences is electromagnetic pulse (EMP).

Just what is EMP? It is an intense burst of radio-frequency noise whose ultimate origin is from Compton electrons produced by the gamma-ray output of a nuclear weapon. It has been established that EMP is completely harmless to living things. People and animals are unaffected by it. Sensitive electric and electronic equipment, however, may be damaged by EMP under certain conditions. Its effects on large systems are as yet little understood, but data do exist for individual pieces of equipment.

Both theoretical and experimental studies on EMP were undertaken by the Civil Defense Research Project during the past year. A theoretical analysis of pickup by aboveground transmission lines, such as electric and telephone lines, was made to determine the voltage and current delivered to equipment resulting from the signal picked up by the wire. To check certain aspects of the theoretical calculations through experiment, an EMP simulator was developed and assembled. The simulator is a device for reproducing in a small volume typical fields produced by nuclear weapons. A small-scale model of a section of transmission line was built and the measured voltage compared with that predicted for the model in the theoretical calculations. Two other independent experiments were performed using the simulator, one involving a mouse and one with AM and FM radios. There were no effects whatsoever from EMP on the mouse, confirming the harmlessness of EMP to life. Nor was the AM radio affected. On the FM radio, however, the circuitry nearest the antenna was damaged as a result of EMP.

## Civil-Defense Behavior in Time of Crisis

In times of international crisis, does the American public take individual civil defense measures to ensure its personal safety? If so, what correlation, if any, exists between a person's level of education and his behavior in the direction of civil defense? An analysis of six public

opinion surveys conducted during the United States–Soviet confrontation over Berlin in 1961 yielded the following conclusions:

1. Regardless of the level of international tension, the lower the level of education, the higher the expectation of war.

2. Regardless of the level of international tension, the higher the level of education, the greater the tendency to engage in personal civil-defense-oriented behavior.

3. During the early stages of a crisis, expectation of war is positively correlated with civil defense behavior for high-school- and college-educated groups, but the correlation is higher for grade-school-educated groups during the later stages of the crisis.

*Turntable rig used for "kerfing" measurements and tests in the cutting of rock by a high-speed water jet. Two blocks of sandstone are mounted on the turntable, and the nozzle of the jet (being adjusted) is directed downward. The high-pressure water enters through the pipe at top left, coming from a truck-mounted commercial pump used in well technology. The grooves in the block facing the viewer were cut in a previous test. They are about an inch deep, and each was cut in a single sweep requiring only about 1 sec for the passage of the block under the nozzle.*

4. The civil defense behavior of people with high school and college educations changes much more quickly in response to increasing levels of tension than that of people with a grade school education, but these differences are not due to differential perceptions of the chances of war.

5. Changes in civil defense behavior and expectations of war are in the same direction and of the same magnitude regardless of the group education level.

6. Changes in civil defense behavior and expectations of war induced by increases in international tension are more enduring, the higher the educational level of the group.

## Management of Incomes in Postattack Period

A disastrous nuclear attack on a country would have many severe economic effects. The nation's ability to recover from these effects would depend mainly on the rebuilding of the complex of social structures that support, facilitate, and coordinate the production and distribution of goods and services. The most efficient use of all available resources would speed the recovery. In a society that has wealth and income differentials, this optimization would necessitate some method of limiting the consumption of nonessentials.

One extreme manner of eliminating wasteful consumption is confiscating excess wealth and centrally distributing essential goods. Such a program would be very difficult to administer, however, and many economists feel that it would harm the overall economy by eliminating work incentives.

The present official policy for a postattack economy would use monetary and fiscal controls to prop up and regulate the financial sectors, and rationing and price controls to allocate consumer goods and services. These plans would be satisfactory for a relatively mild attack but not for one of devastating proportions. Under such emergency conditions a fiscal-control and rationing system would probably fail because it is complex and relies on either voluntary cooperation or vigorous enforcement. The optimal system for allocating resources must effectively control consumer spending while permitting incentive wages.

A dual-account system, under which all financial transactions would be made with checks or credit cards, would allocate all incomes between disposable and blocked accounts. Funds in the disposable account would be immediately available for expenditure on essential goods and savings. The other funds would be blocked from being spent until the emergency was over. These blocked-account funds would generally represent a form of forced savings with interest paid, thus permitting incentive incomes to be realized while preventing important differences in per capita command over essential goods.

# LABORATORY LIFE.....

*Storage and retrieval of data by computer – Nuclear Safety Information Center.*

*Youth Opportunity Program.*

*Visiting students – Career Day.*

# Nuclear Energy

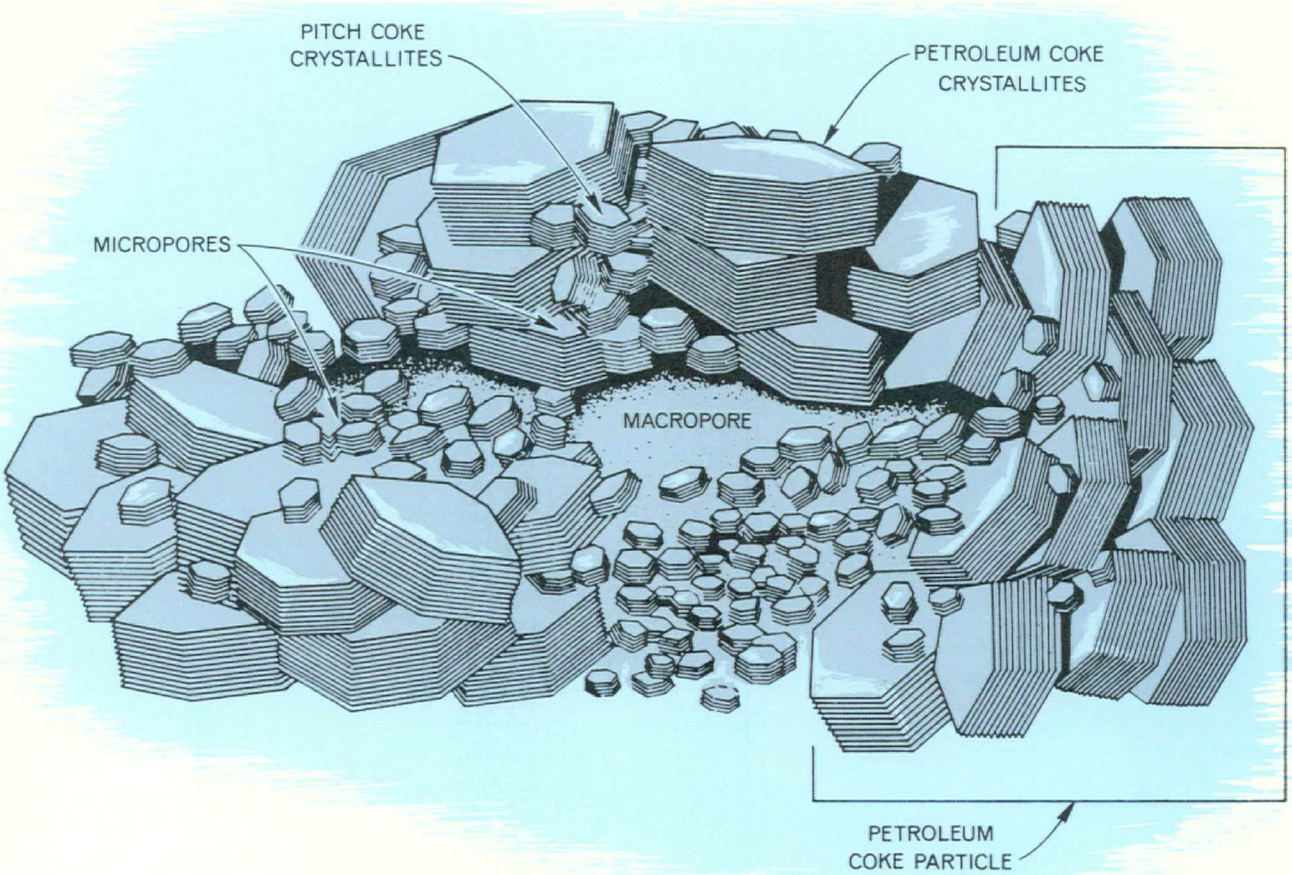
# Molten-Salt Reactor Program

The Molten-Salt Reactor Program is exploring one of the most promising paths to the long-range, low-cost source of energy which is a primary goal of the Atomic Energy Commission.

The pace of nuclear power plant construction is evidence that the harnessing of fission for production of electricity is today an economic success. But continued cheap nuclear power in the face of inevitably rising ore costs depends on the development of reactors that can more fully use the nuclear fuel that is mined – today's reactors use only 1 to 2%. The key to better fuel utilization is use of excess neutrons from the fission chain reaction to convert nonfissile isotopes in the fuel to fissile nuclides. Some conversion of uranium-238 to fissile plutonium-239 occurs in all uranium-fueled reactors, the amount depending on the size and composition of the reactor core. Some reactors also include thorium, which can be converted to fissile uranium-233. The goal of the Atomic Energy Commission is to develop reactors that can use neutrons so efficiently that practically all of the uranium-238 or thorium can be converted. Such reactors are called "breeders," since they produce more fissile material than they consume. Because they would waste almost none of the mined fuel, breeders could use quite expensive ore without incurring excessive feed costs. Thus breeders conceivably could open the way to cheap power for centuries to come.

Design of an economical breeder reactor is not easy, however. The nuclear physics of uranium-238 and plutonium-239 are such that a reactor operating on this cycle can breed only if the neutrons are caused to react before they slow down, and this requires a large inventory of fissile material. An alternative route to breeding, requiring lower inventories, is the thorium–uranium-233 cycle in a reactor using slow ("thermal") neutrons. Although the yield of neutrons from uranium-233 is great enough to make breeding with thermal neutrons possible in principle, the feasibility of a thermal breeder hinges on keeping nonproductive absorptions of neutrons unusually low. The necessity of frequent processing to remove the "ashes" (fission products) from the core then tends to make the cost of power from a thermal breeder too expensive.

For some years it has been the conviction of the Oak Ridge National Laboratory that the requirements of a thermal breeder reactor can be




*An improved understanding of different graphite structures and fast-neutron damage mechanisms at high temperatures has come out of studies aimed at improved graphites for the proposed molten-salt breeder reactors. The way now appears to be open to longer-lived graphite, which will minimize replacement costs in a power reactor.*

met in a practical and economical manner by the use of fluid fuels, specifically, molten fluoride salts. This conclusion is well founded on a wealth of research and development experience beginning with the Aircraft Nuclear Propulsion program of the 1950's. A mixture of the fluorides of lithium, beryllium, uranium, and thorium is a stable, noncorrosive, low-vapor-pressure liquid at the incandescent temperatures of modern steam power plants. There is very little nonproductive capture of neutrons in the primary constituents of this salt. Furthermore, neutron absorption in fission products could be minimized by rapid on-site processing of the liquid fuel of a molten-salt reactor. Studies show that molten-salt reactors should perform well in terms of utilizing nuclear fuel supplies and that the cost of power from them should be quite competitive, mainly because the high costs of fabricating, transporting, and dissolving fuel elements are eliminated. The objective of the Molten-Salt Reactor Program at ORNL, therefore, is to develop the designs and the technology necessary for the molten-salt power reactor to emerge as an economically viable reality.

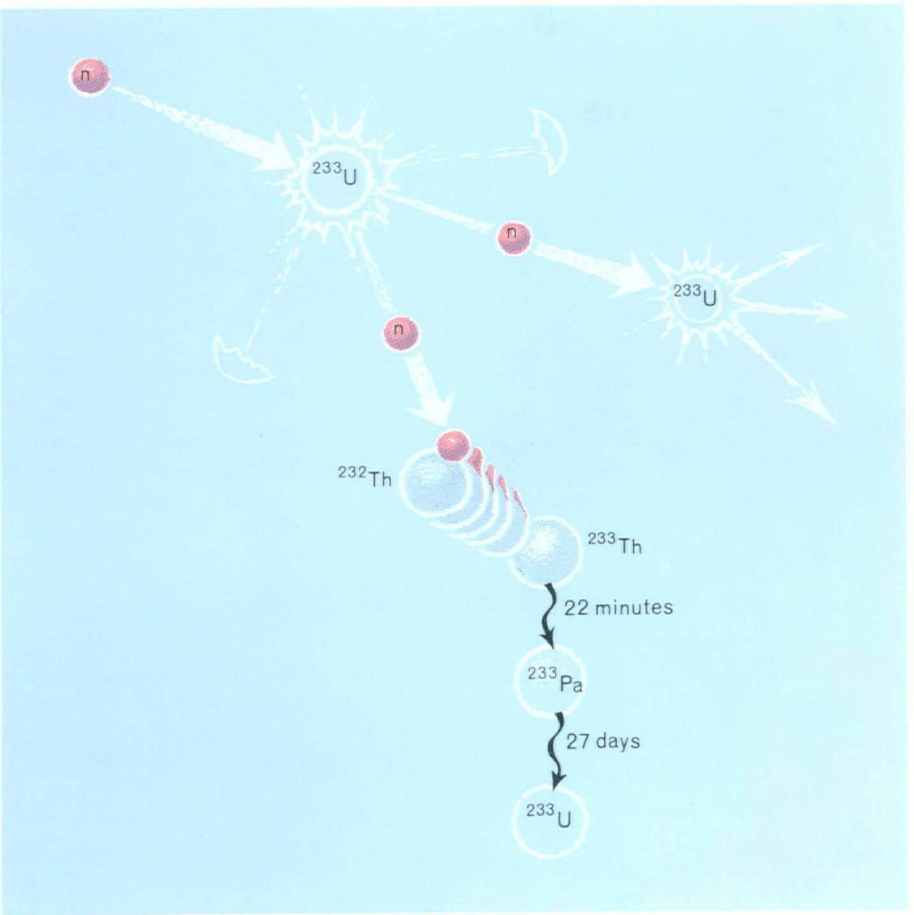
A major step in the program has been the Molten-Salt Reactor Experiment, a small 8000-kW reactor incorporating many of the features of the proposed power reactors. Built in 1962–64, the MSRE began nuclear operation in June 1965 with uranium-235 as the fissile material. By March 1968 the MSRE had run for more than 9000 equivalent

full-power hours at a temperature of 1200°F. Over the preceding 15 months the reactor had been critical 80% of the time, and for the last six months the fuel was in the core continuously and the reactor was critical 97% of the time. The salt had proved quite stable, corrosion was practically nil, key equipment had performed reliably, and maintenance of the radioactive systems had been shown to be quite feasible. In short, the MSRE had convincingly demonstrated the reliability and practicality of the molten-salt reactor concept.

Although the MSRE had accomplished its primary purpose, a new major objective had been established – experimental operation with uranium-233. From this would come direct confirmation of the data and design procedures used to predict the performance of future molten-salt power reactors.

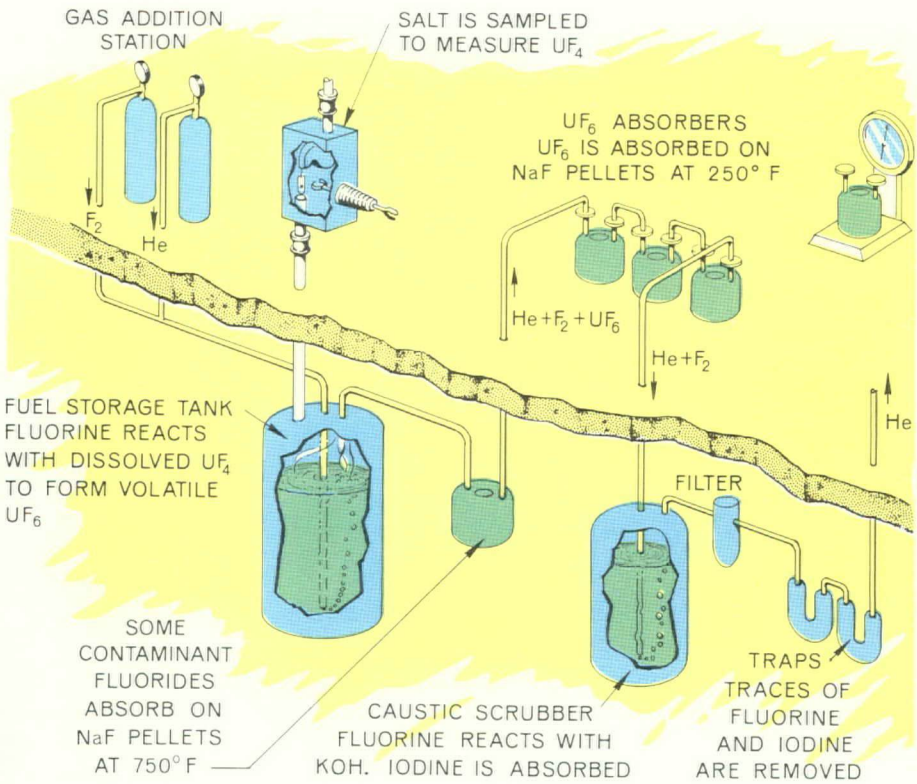
After the March shutdown, the uranium originally in the fuel salt was very efficiently stripped by the fluoride volatility process in equipment located in the reactor building. This was in itself a significant accomplishment, for it demonstrated, on practically a full scale, the only chemical process essential to economical operation of large molten-salt converter reactors.

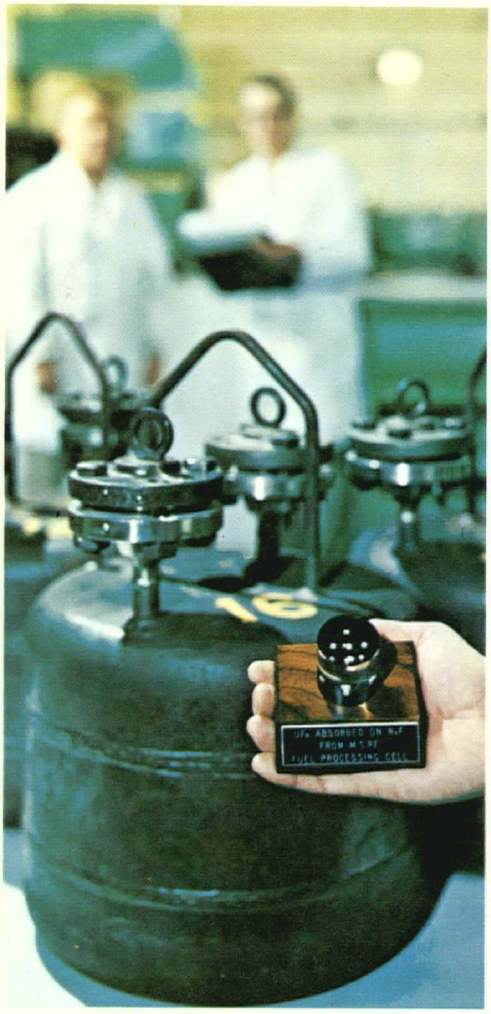
The uranium-233 for reloading the MSRE was prepared in the form of $UF_4$-LiF (73-27 mole %) eutectic salt in the Thorium-Uranium




*The key to energy production from thorium is the process depicted here. Neutrons and heat are produced by the fission of uranium-233. Neutrons not needed to sustain the fission chain reaction are absorbed in thorium, which is not itself fissionable. This initiates a natural process of radioactive decay that results in uranium-233, which is then available as fuel to produce more heat.*

*Recovery of uranium from the MSRE salt was accomplished in a cell adjacent to the reactor, using the simple equipment depicted schematically here. The fluoride volatility process, which will be part of the processing scheme for future molten-salt breeder reactors, very efficiently removed 218 kg of uranium from 5000 kg of salt in only 47 h of fluorination.*



*Excellent decontamination of the uranium was achieved as it was stripped from the MSRE salt and absorbed on sodium fluoride pellets. Absorber pots, each containing 12 to 14 kg of uranium, fresh from a tank of salt radiating over 2,000,000 millirems/h, read less than 2 millirems/h at contact.*

*The batch of uranium-233 used for refueling the MSRE contained so much uranium-232 (225 ppm) and its radioactive daughters that its use in basic research or conventional fabrication of reactor fuel elements was practically prohibited. The relatively simple operations of fueling a molten-salt reactor could be done remotely, however. Equipment set up in the Thorium-Uranium Recycle Facility at ORNL was used to convert uranium-233 oxide into $UF_4$-LiF eutectic salt. Here a can of this salt containing 7 kg of uranium-233 has been transported to the MSRE and is being lowered into a heated tank. The enriching salt will melt and drain into the stripped carrier salt as the first step in the startup of the MSRE with uranium-233.*

*Twenty-six years after he had discovered uranium-233, AEC Chairman Glenn T. Seaborg started up the world's first reactor to operate on this man-made isotope. Seated on Dr. Seaborg's right at the MSRE console is ORNL staff member Dr. R. W. Stoughton, a codiscoverer of uranium-233.*

Recycle Facility at ORNL. The loading operation consisted in melting 14 liters of this eutectic containing 35 kg of uranium-233 into the 1600 liters of stripped carrier salt. After the required amount of uranium had been added, AEC Chairman Glenn Seaborg took the power up to 100 kW in a ceremony marking the world's first power operation with this man-made isotope, around which the thermal breeder concept revolves. By the year's end, experiments had shown that uranium-233, in an environment very close to that of the proposed molten-salt power reactors, behaved as predicted.

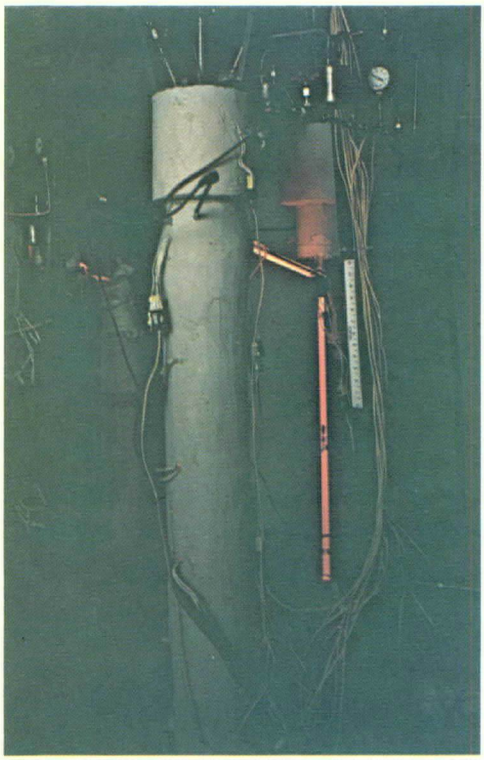
Besides the successes with the MSRE, 1968 also saw significant strides in other development efforts for the molten-salt breeder, notably toward improved graphites for its moderator and in methods of processing molten salt containing both fissile and fertile material.

A better understanding of different graphite microstructures and fast-neutron damage mechanisms at high temperatures has come out of recent studies. In addition, exposure of special grades of graphite to extremely high fluences of neutrons in the High Flux Isotope Reactor at ORNL gave encouraging results during 1968, pointing the way to the long-lived graphite needed to minimize replacement costs in a molten-salt power reactor. New methods to reduce gas permeability of the graphite were also proved during 1968.

In harnessing the molten-salt reactor to produce electricity, it will be necessary to use an intermediate heat transfer medium to transport heat from the fuel salt to the steam. Screening of possible media showed that

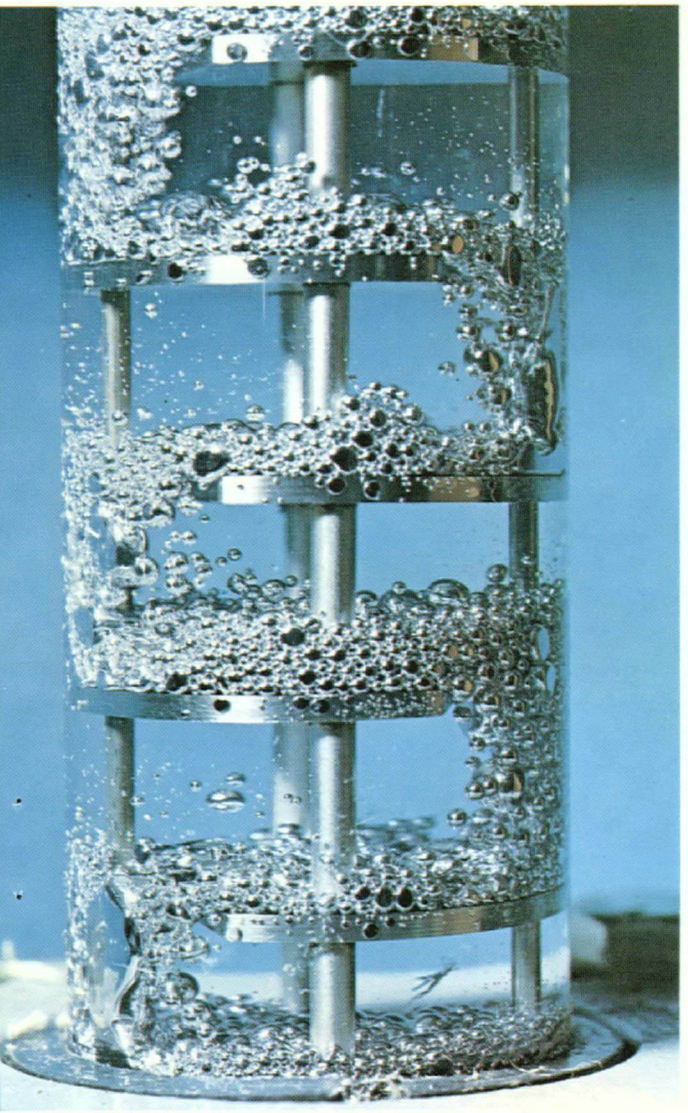
*Graphite permeability to gaseous fission products can be cut by a factor of a thousand or more when surface-connected pores are sealed with pyrolytic graphite deposited by a quick pressure-cycling method developed at ORNL. This hollow graphite cylinder was sealed by pulsing with butadiene gas at 800°C. A thin slice was then soaked with mercury, which filled the porous region in the interior and caused it to show up light in this x-ray photograph. The excellent surface sealing shown here will help in minimizing neutron absorption by gaseous fission products in future molten-salt reactors.*

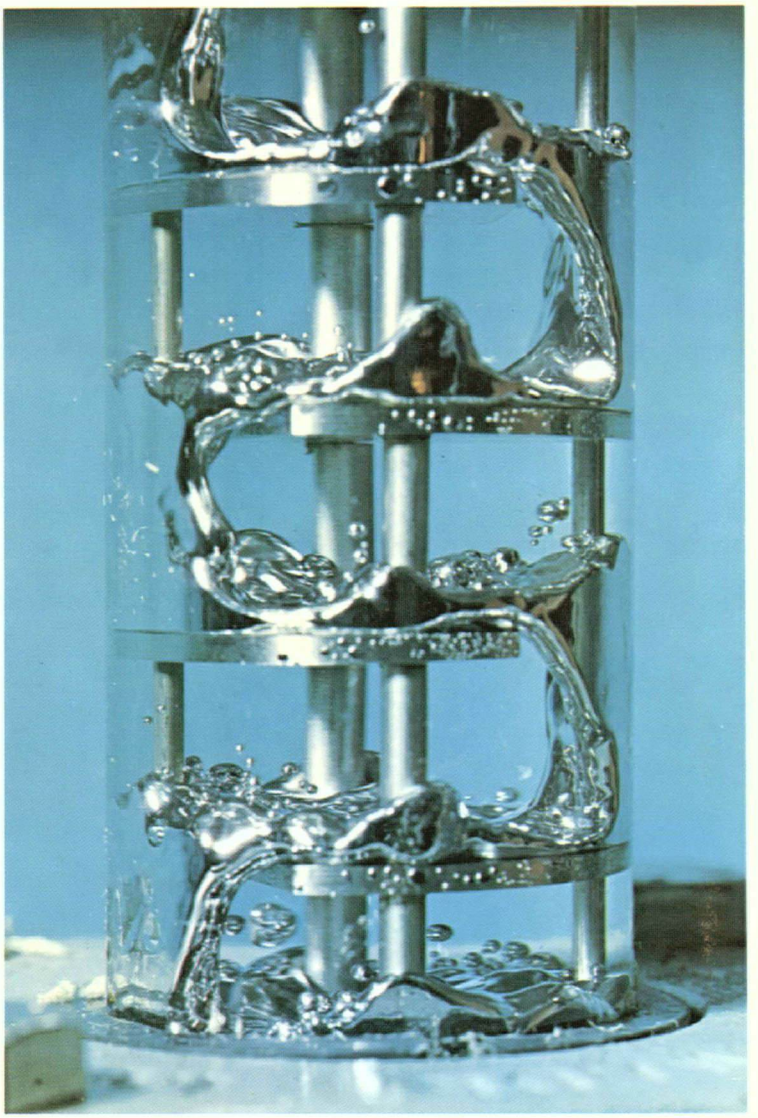
*A red glow marks the "cool" (1000–1200°F) leg of a test loop in which a molten salt circulates by natural convection. Materials for high-temperature, low-pressure molten-salt power reactors are tested by exposure for thousands of hours in loops such as these.*



sodium fluoroborate, an inexpensive, readily available material, looked most promising. During 1968, engineering-scale development loops were operated for thousands of hours with fluoroborate salts. Their operation provided valuable experience in handling this material.

Rapid separation of protactinium from the fuel salt is essential to the attainment of a positive breeding gain in the simple single-fluid molten-salt reactors toward which we are working. In 1968, careful measurements of solubilities and distribution coefficients in fluoride-salt–liquid-bismuth systems not only confirmed earlier evidence that reductive extraction of protactinium is chemically feasible but indicated that the necessary equipment can be of reasonable size. Based on these encouraging results, development work moved ahead toward a separations system that could be connected directly to a breeder reactor salt system, to function as a "kidney" for selectively removing certain constituents from the salt. The extraction process, while similar in some

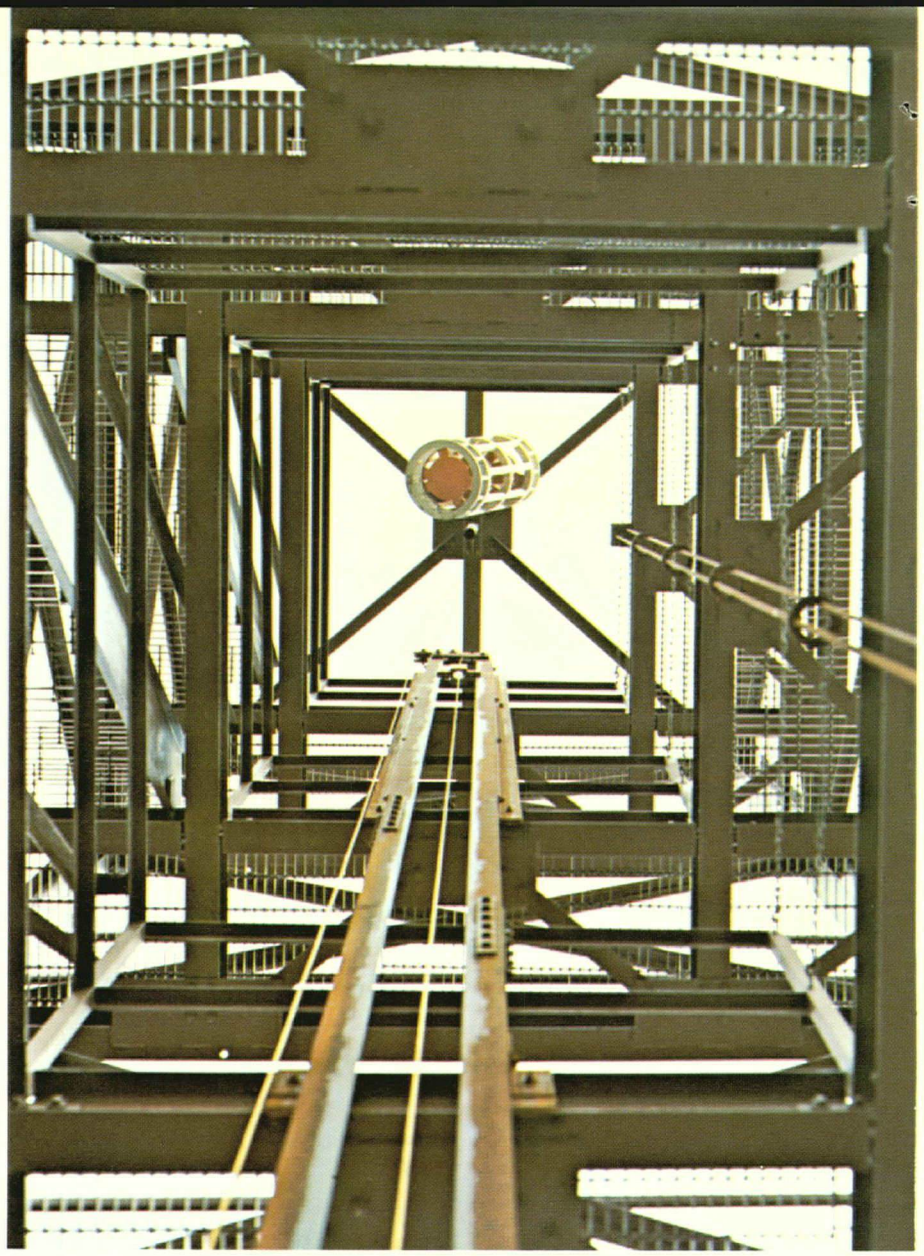




*The efficient contacting of reactor salt with molten bismuth is an essential operation in the processing of molten-salt breeder reactors, and this has led to studies at ORNL in the relatively unexplored field of two-fluid systems with a large difference in specific gravities. These photographs illustrate a peculiarity of such systems that has been discovered. At low flow rates the metallic phase is well dispersed, as shown in the photograph on the left, but at high rates it forms a continuous stream, drastically reducing the efficiency with which a dissolved component can be exchanged.*

respects to the widely used solvent extraction process in aqueous systems, is different in that it involves a reduction of the uranium, protactinium, and thorium to the metallic state and exploits the differences in reduction potential to effect differences in distribution between a molten bismuth phase and the molten salt. Novel components on which development was started include electrolysis cells with molten bismuth anode and cathode and liquid-liquid contactors for fluids differing widely in density.

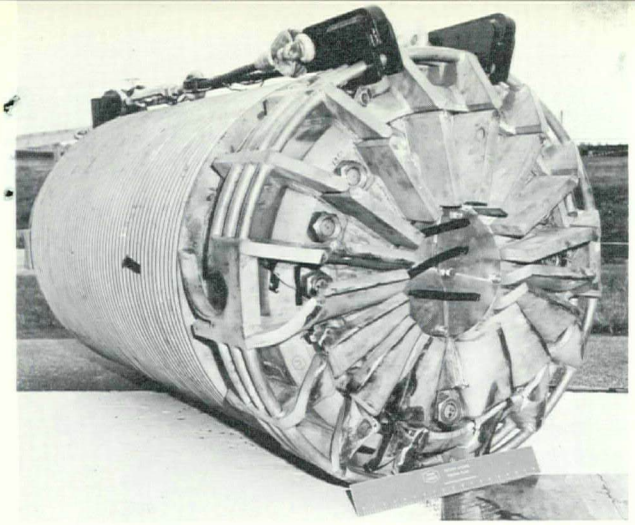
The solubility and distribution data also showed that it would be practical to separate plutonium by the reductive extraction process. This means that it would be feasible to start molten-salt breeders with plutonium and process the salt to remove the unwanted plutonium after uranium-233 has bred in. Studies of the economics show that this mode of startup will be economical for the molten-salt reactor and will be of benefit to the entire power industry, which anticipates a surplus of plutonium for the next two decades.



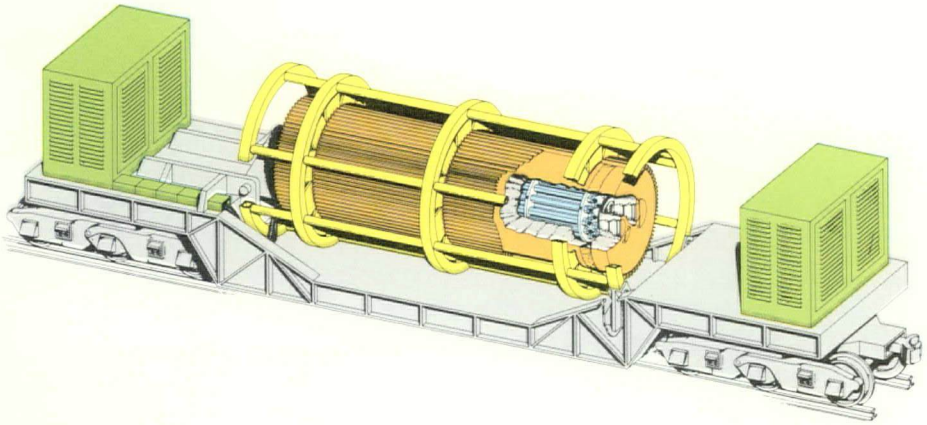
*Heads up! You are looking upward at a scale model of a fuel shipping cask (orange-colored cylinder) equipped with an energy-absorbing crash frame (light gray) in position at the top of a 30-ft tower prior to its being released to fall onto an unyielding surface. The simulated cask suffered no significant damage when it came to rest.*

# Shipping of Nuclear Fuels and Wastes

In the nuclear-powered age, concentrated quantities of radioactive materials will be constantly on the move between reactors, processing plants, and disposal plants. How can safety be provided during transport equal to the safety within the plants and in the disposal sites? Concern and foresight are needed to forestall disastrous accidents. What happens in a railroad wreck? What if a truck goes off a bridge into the bed of a stream that eventually supplies a city with water?



*The photograph shows the result of another drop test carried out by the Nuclear Safety Program. The cask in this case was designed for shipping either liquid or solid radioactive materials and was made with thick walls of depleted uranium for radiation shielding. It weighed 28,000 lb and was equipped with fins to cushion an impact by crumpling. It was dropped closure end downward from 30 ft onto a concrete surface, but showed no leakage of its content despite an internal pressure of 165 psi.*



*This conceptual drawing illustrates a fuel shipping system which could readily dissipate 200 kW of heat emanated by 18 bundles of fast breeder reactor fuel element tubes. The cask is 7 ft in diameter and 24 ft long, with thick steel walls which provide both radiation shielding and containment. The cask is filled with sodium to provide efficient heat transport from the fuel throughout the fuel cavity. The boxlike structures at the ends of the car portray portions of a cooling system, which maintains the cask contents at low temperature under normal conditions. The cask is cradled within an energy-absorbing steel crash shield (cage) to reduce impact loads in case of an accident. The temperature of 20-day-cooled fuel elements will not exceed 900°F, even if accidental loss of coolant occurs.*

Staff members in the Nuclear Safety Program have undertaken to find out how to forestall spillage of this kind. They have, in fact, assembled a "Guide" that provides information on all important aspects of shipping cask design such as shielding, heat removal, structural design, and fabrication. In the case of nuclear fuel, the Guide provides criteria for spatial separation or internal poisoning so as to prevent a chain reaction in the event of a change of shape or in case separated vessels are thrown together so as to make an overcritical amount. The Guide provides design information not previously available and builds extensively on tests and good engineering practice in cask design and fabrication both in the United States and abroad. It is written to conform to the appropriate United States regulations and to provide safe transport for large quantities.

Within the Chemical Technology Division, safe transport is viewed as part of the larger problem of fuel-element reprocessing. The fuel elements of the Atomic Energy Commission's future liquid-metal fast breeder reactors, for example, may consist of bundles of 200 or more fuel tubes, each a quarter of an inch in diameter and nearly 20 ft long. The radioactive afterheat that a group of them will generate while being shipped from reactor to reprocessing plant requires that a 200-kW cooling plant go along with them. They must, of course, be inside a radiation shield, probably made of steel 2 ft thick. This great cylinder is then mounted in a cage that will crumple under impact, thereby absorbing the energy of a collision. Drop tests have shown the adequacy of the design. With this kind of foresight, spillage should be very, very rare.

# Desalting

The Department of the Interior's Office of Saline Water supports an extensive program of desalination studies, evaporator component development, and evaporator plant design at ORNL. The AEC supports closely related work on nuclear power plants to provide low-cost energy for desalting and on the further development of the new concept of an agroindustrial complex.

In 1968 ORNL developed for the Office of Saline Water a complete conceptual design of a 250,000,000-gal/day vertical-tube evaporator plant with multistage flash feed heating. This efficient process incorporated technological advances which reduced the estimated cost of distilled water to levels that would make it attractive for agricultural use in many locations.

In studying nuclear energy sources for desalting plants, ORNL engineers investigated the savings that might be achieved by improvements in light-water reactor technology. They found that the use of metallic uranium fuel could result in substantial improvements in the fuel cycle of pressurized water reactors. With clad metal fuel, reactors could operate not only at low temperatures to provide process heat for water-only plants but also at higher temperatures for electric power and water production in a dual-purpose plant. The major advantage of the metal fuel is that high conversion ratios – greater than 0.9 – can be achieved. Coincident with the ORNL work, irradiation tests at the Pacific Northwest Laboratories showed that hollow fuel rods of uranium metal did not exhibit the severe irradiation swelling which had been a serious drawback of the previously used uranium metal fuels. Stability against irradiation swelling would allow the fuel to be used to high burnups, and metallic uranium would give good neutron economy. These advantages would allow commercial light-water reactors to achieve lower fuel-cycle costs and improved utilization of fuel resources. If further work substantiates the promising results of the work to date, particularly if the metal-water reaction that occurs when the clad is inadvertently penetrated can be controlled, light-water reactors may produce energy at costs in the low range once thought to be attainable only with advanced reactors.

The application of low-cost energy and desalted seawater to food production and industrial processes in an agroindustrial complex was

*A photograph taken by a member of the ORNL visiting team that inspected irrigation methods in India. This is a shallow-well pumping rig. The ox walks in a circle, carrying the long pole. This turns the sprocket wheel in the center of the picture, which engages pegs on a wheel attached to the shaft. As the shaft rotates, the buckets pick up water.*

reported last year. This idea received enthusiastic response all over the world and was the subject of one session of the International Atomic Energy Agency's international meeting at Madrid, Spain, in the latter part of 1968. At that meeting many countries indicated great interest in the promising results of the initial ORNL studies and reported the start of investigations into the applicability of this concept to their own needs. Investigation of the specific problems and opportunities of each particular site is necessary to determine whether the large investment required for an agroindustrial complex makes economic sense. ORNL is participating in more detailed studies of specific sites in the Middle East, India, and Puerto Rico.

The most comprehensive of the agroindustrial studies is that for the Middle East. Late in 1967 the U.S. Senate adopted Senate Resolution 155, sponsored by Howard Baker (Tennessee), calling on the administration to consider the application of large-scale nuclear desalting plants as a means of supporting a stable and durable peace in the Middle East. Pursuant to this resolution and in cooperation with the AEC and the U.S. Departments of State and Interior, in June 1968 the Laboratory embarked on a year-long study of the potential of nuclear-powered agroindustrial complexes for the area. Initial findings indicate that nuclear dual-purpose electric-power–desalting plants combined with modern intensive agriculture may offer substantial economic benefits to the countries in the Middle East. A field trip to the United Arab Republic and the other Arab countries by ORNL staff members and representatives of the U.S. Atomic Energy Commission and the International Atomic Energy Agency provided information which generally confirmed the assumptions used as a basis for the preliminary findings; however, to obtain more definitive results will require actual test data for the specific sites being considered. Cooperative studies with the nations concerned are being initiated as a means of obtaining such data.

Ancillary studies including market analyses to assist in selecting optimum product mixes; the development of information on food processing and other synergistic industries; the consideration of managerial, financial, and operational aspects; and the evaluation of the socioeconomic benefits of the selected complexes are also under way.

Laboratory representatives visited India to discuss possible applications of the agroindustrial complex at locations in that country. Following up the ideas generated in these discussions, the Indians studied applications for specific sites. A special type of agroindustrial complex for India has been suggested by P. R. Stout, working at Oak Ridge on leave of absence from the University of California. This complex would not involve desalting but would use electricity from nuclear power plants to pump irrigation water from the large aquifer which underlies most of the Indo-Gangetic Plain. In this way the immensely fertile plains of the Ganges could grow two or three crops per year instead of producing just one crop during the monsoon season, as at present.

The AEC and the Interior Department entered into an agreement with the government of Puerto Rico for a study of the feasibility of locating a nuclear energy center on the island. Of the three regions to be considered, the south side is already suffering a water shortage and will particularly be considered as a site for a desalting plant. The Puerto Rican complex will supply water mainly for municipal and industrial use rather than irrigation and will place emphasis on the low-cost electricity and industrial production aspects of the complex.

ORNL's hyperfiltration studies, which are a part of the OSW water research program here, were expanded in 1968 to include a project for the Federal Water Pollution Control Agency. Under this agreement, arrangements were made to subject municipal sewage to filtration through membranes formed dynamically from inherent constituents of the sewage itself, as well as from added substances shown in laboratory studies to form salt-rejecting layers. Accordingly, filtration equipment is in operation at the Oak Ridge secondary sewage plant.

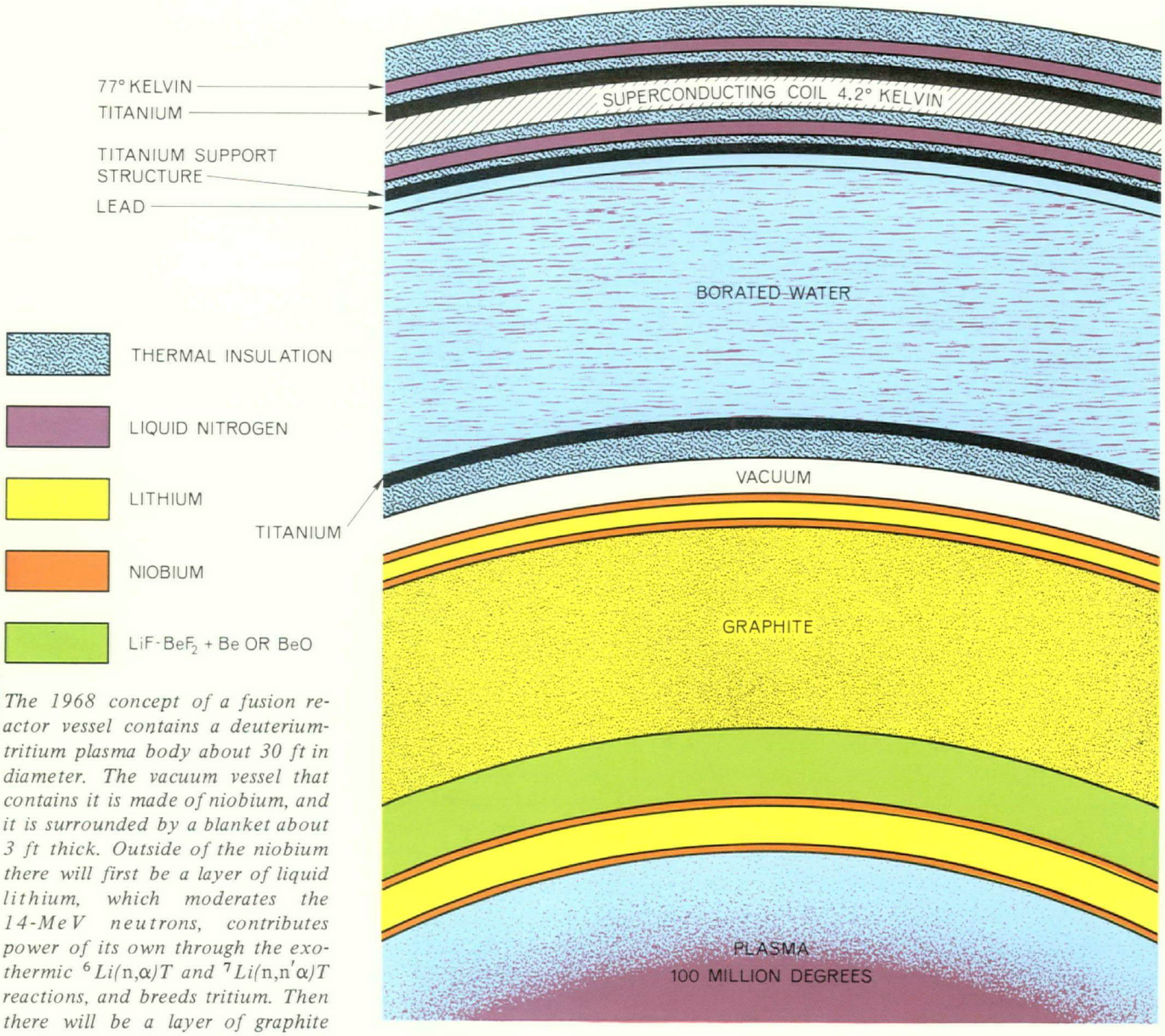
# Fusion

## Engineering Feasibility of Controlled Nuclear Fusion

To obtain useful power from the nuclear fusion of light nuclei requires the solution of problems in two broad areas. The plasma physics problems are mainly those of adequate plasma confinement and stability but also those of initial starting and fueling of a going reactor. In the second group are nuclear engineering problems which seek solutions to the moderation of the 14-MeV neutrons produced through fusion, the regeneration of the tritium fuel with a favorable breeding ratio, the practical development of complex, sturdy, but cheap magnetic confinement systems, and the development of a vacuum wall resistant to radiation damage and sputtering, yet having high heat transfer properties. All of these major tasks must be done cheaply enough to provide central-plant electric power competitive with fission breeders.

The traditional attractions of fusion power remain: a plentiful neutron supply and resulting short doubling times in the breeding of tritium, minimal hazards, and small afterheat, hence low radioactivity and few disposal problems. Cost reductions through possible siting advantages inside cities and through the use of low-grade steam for heating have not been adequately evaluated.

New studies during the year, principally at ORNL, sought answers to these engineering problems and the effects of these economic criteria on the plasma problems. One of the primary costs will be that associated with the production of the magnetic field which confines the energetic reacting plasma. To be economical these fields must be produced by large superconducting coils; the study showed further that present technology in size and field strengths for these coils is now adequate to serve long-range fusion needs. The blanket – moderator of the neutrons and breeder of tritium fuel – can be made cheaply from lithium; the large excesses of neutrons give doubling times for tritium breeding of months instead of years. The reactor would be large in size (30 ft in diameter) and in power rating (10,000,000 kW of electricity); this reduces the cost per installed kilowatt to about $15.00. Large sizes fortunately also ease some present stringent demands on plasma confinement properties, but they restrict these power sources to central power plants. The possibility of direct energy conversion seems remote, and the steam cycle must be used.




*The 1968 concept of a fusion reactor vessel contains a deuterium-tritium plasma body about 30 ft in diameter. The vacuum vessel that contains it is made of niobium, and it is surrounded by a blanket about 3 ft thick. Outside of the niobium there will first be a layer of liquid lithium, which moderates the 14-MeV neutrons, contributes power of its own through the exothermic* $^{6}Li(n,\alpha)T$ *and* $^{7}Li(n,n'\alpha)T$ *reactions, and breeds tritium. Then there will be a layer of graphite moderator, cooled again by liquid lithium. Outside of this (after thermal insulation) will be a layer of borated water, and then the thermally shielded superconducting coil structure starts, with the magnetic stresses supported by titanium walls. Heat removed from the blanket would drive a steam cycle for electric power generation. Whether the reactor would be cylindrical or toroidal is not yet known.*

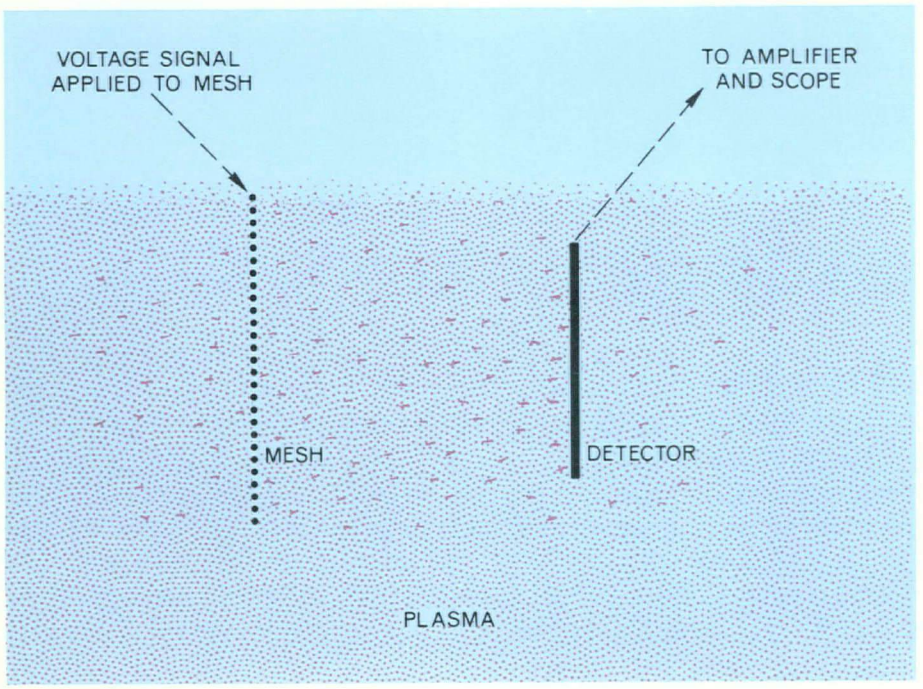
In summary, fusion looks remarkably good as far as engineering, breeding, and safety are concerned. We must now turn our attention back to the problems of plasma stability in large sizes and densities, and we must devote increasing concern to the integrity of the vacuum-vessel wall under the great throughput of 14-MeV neutrons. The threat is the voids that arise in the metals, as pictured in the *Materials* section of this booklet. Do the voids pose the ultimate threat to the economy of both fission and fusion?

## Computer Simulation of Plasmas

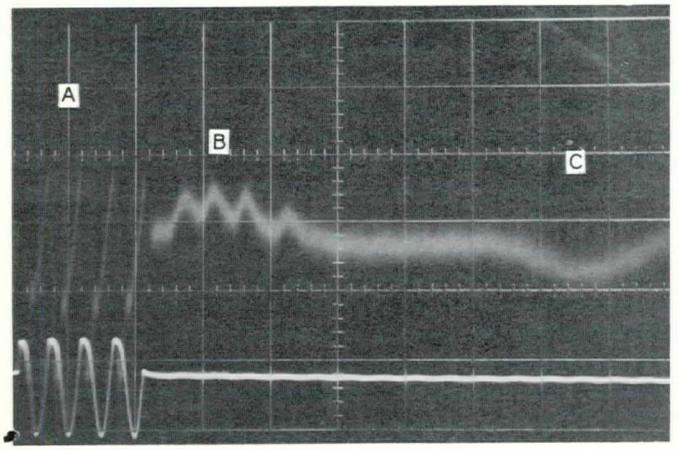
Computers are finding increasing use in plasma physics in a way somewhat akin to the "Monte Carlo" methods that have been used in neutron physics for many years. You start with a sample of perhaps 10,000 or 50,000 conceptual particles in the plasma, give the computer their locations and instantaneous motions, apply some kind of stimulus that you are investigating, and have the computer solve for the paths of

*A study of the properties of pseudowaves (ion bursts) by means of* (a *and* b) *an actual experiment and* (c) *a computer simulation "experiment." In the actual experiment* (a) *an oscillating voltage applied to a mesh immersed in a plasma generates bursts of free-flying ions that give a wavelike signal on the detector. The lower trace in the oscilloscope pattern* (b) *shows the time-varying voltage placed on the grid. Signal* B *in the upper trace is the pseudowave signal detected by an ion-sensitive probe. (Signal* A *is a directly coupled signal, and signal* C *is an ion acoustic wave that has suffered dispersion and Landau damping.) In the computer simulation* (c), *ions with uniform velocities but with 20 different phases are directed upward toward a sinusoidally varying potential region* XY, *and their trajectories are numerically calculated. A majority of the ions are resonantly trapped, accelerated, and then ejected upward in bursts, bunched in velocity and phase. The basic properties of pseudowaves are such as shown in* c.

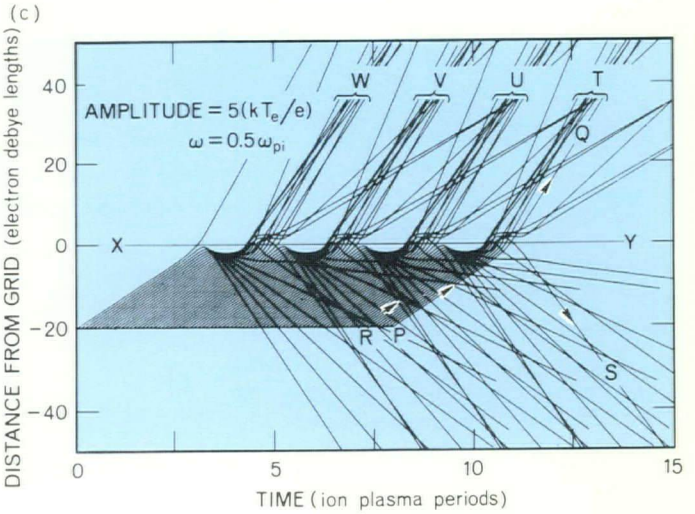
(a)




(b)




(c)



the particles one by one under the restrictions of plasma equations and finally display the cumulated result. The display can be by slow-motion movies, in which a microsecond is slowed to as many seconds as you wish. The results are often quite illuminating; waves grow and subside, turbulent surgings and reflections are seen, and sometimes gobs of plasma move out of the main body.

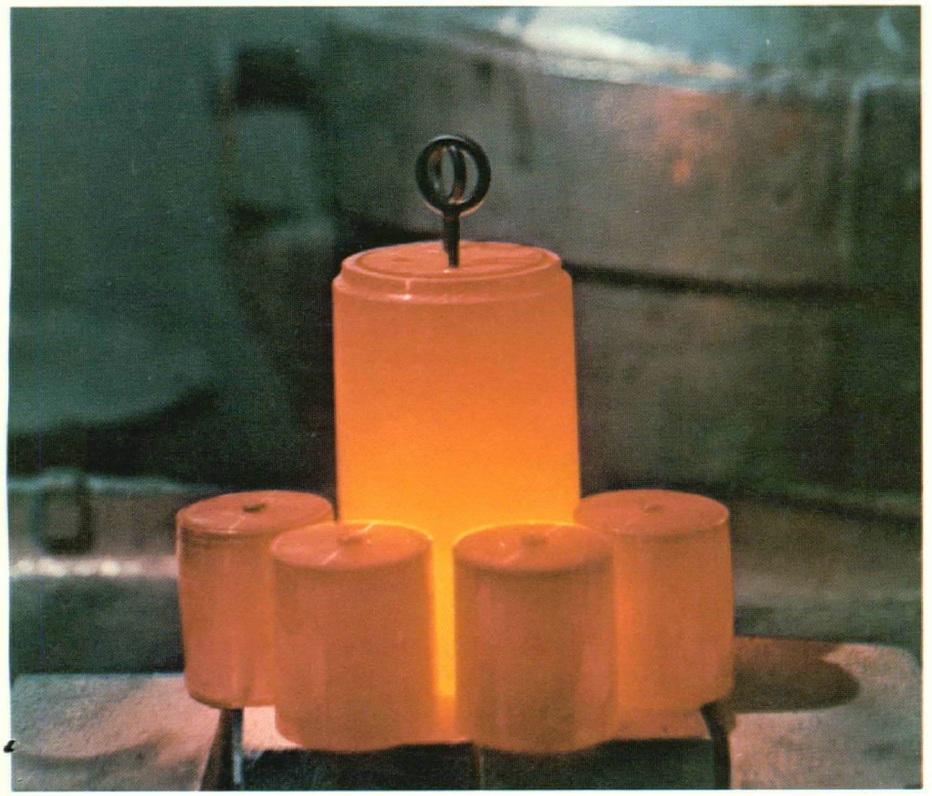
Does the result correspond to reality? It does if the "experimenter" has been smart enough to include in his equations all the items that correspond to nature. The experimenter knows of this limitation, but nevertheless he can learn things because the motions are often far too complex for him to imagine and because he can study the effects of this or that modification to the stimulus; that is, he can develop a "feel" for the plasma response.

A simple example of computer simulation of plasma arose in the Thermonuclear Division last year. An experiment (*a*) had four sine waves of voltage applied to a wire mesh in a plasma in a vacuum vessel, and a few inches away was a metal detector plate connected to an amplifier and an oscilloscope. The second picture (*b*) shows what was seen on the scope. The lower trace shows the voltage applied on the mesh, and the upper trace shows what arrived at the detector. First, at *A* we see a large detector signal caused by direct capacitive coupling between mesh and detector. It has nothing to do with the plasma. Second, at *B* four wiggles later appear, somewhat resembling the impulses that had been applied to the mesh. Later still, at *C* a signal appears on the detector corresponding to a plasma wave transmitted from mesh to detector.

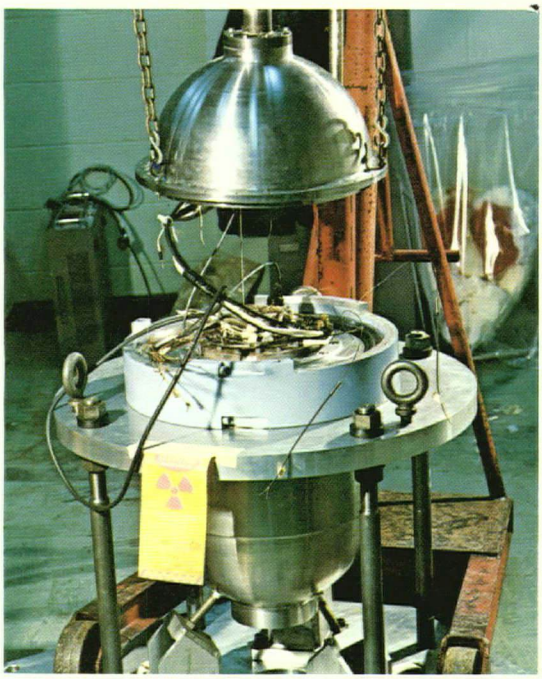
What is happening at *B*? Let us ask the computer if it can give us understanding. First we replace the mesh with a bodiless mathematical line, shown as *XY* on the computer output plot (*c*). We will let the voltage wiggles appear on this line. Then, plotting distance vertically, we start a positive ion at *P* and let it move toward *XY*. With time on the abscissa, this motion is shown as a straight line sloping upward. The particle arrives at *XY* when the voltage is slightly negative, so it is accelerated a little and proceeds on to *Q*. The increased speed is shown by the steeper slope of the portion *PQ* of the ion's path. Later another particle is started, say from *R*. It arrives at *XY* when *XY* is positive. It therefore fails to pass *XY* and is reflected to *S*. Thus you start maybe a hundred particles at various times relative to the four voltage signals on *XY*, you let the computer plot their paths, and you look at the total result. What you see is that many of the paths have a tendency to group into the bunches *T*, *U*, *V*, and *W*, and in those groups *the paths have approximately the same slope.* This lets the cat out of the bag, because equal slopes mean equal velocities. What was happening in the real experiment was that ions emerged from the holes in the mesh whenever the voltage on the mesh was appropriate, and flew in four successive bursts to the detector. Since the velocities were uniform, each burst reproduced on the detector, after a flight time, approximately the voltage impulse applied to the mesh. The wiggles at *B* may have looked like waves caused by coherent plasma motion, but the plasma physicists recognized them as *pseudowaves.* They were due to free-flying ions.

# Strontium Power

Radioisotopes recognized by the general public are few: people have heard of uranium-235 and the radioelement plutonium, and more than likely they have also heard of strontium-90 – probably unfavorably. Castigated in news stories, editorials, and modern folk songs, strontium-90 serves as the focal point for comment on the menacing aspects of nuclear energy. Yet, like our other sometimes dangerous servants, fire and electricity, strontium-90 has its benign side: its radiations can be absorbed and converted into heat or electrical energy, or absorption can be minimized and the radiation used to effect changes in materials. Utilization of fission products has been one of the most important parts of our isotopes program since 1948. Two fission products – cesium-137 and strontium-90 – have received most of our attention because they are



*Strontium power "sources" glowing with the heat of radioactivity. These sources were fabricated for two current AEC-contracted projects for the development of improved isotopic power generators. The larger source is for SNAP-23, being developed by Westinghouse for terrestrial uses; it contains 167,000 curies of strontium-90, generating 1130 W of heat. The smaller sources each contain 31,600 curies and generate 215 W; they are intended for SNAP-21 generators, being developed by the 3M Corporation for undersea applications.*



*Assembly of a SNAP-21 radioactive power source.*

the major long-lived components of nuclear fuel process waste. In the past several years, especial emphasis has been given to utilization of strontium-90.

The element strontium has been largely ignored since its discovery in samples of Scottish lead ore about 200 years ago. Used only to give a red color to fireworks and in a few bearing metal alloys, strontium has been studied primarily by comparison with its alkaline-earth cousins, calcium and barium. The radioisotope strontium-90 has a half-life of 28 years, and the first curie amounts were separated at ORNL as a by-product of barium-140 separations from uranium fuel slugs discharged from the old ORNL Graphite Reactor. Barium and strontium were coprecipitated with lead sulfate, the lead was removed by electrodeposition, and the barium-strontium separation was made by concentrated $HNO_3$ and HCl-ether precipitations. The radiostrontium fraction was mostly 51-day strontium-89 with a small percentage of strontium-90. The strontium-89 was allowed to decay, and the strontium was chemically purified by chromatographic ion exchange, a new procedure in those days. For about ten years, most of the strontium-90 used throughout the world came from this source. Although the amounts were minute compared with present production, manifold uses for strontium-90 in all kinds of measuring devices developed. In all of these devices, the 2.27-MeV beta particle from 64.4-h yttrium-90, daughter of strontium-90, interacted with materials in various ways to measure film or coating thickness, vapor densities, cigarette firmness, the filling of containers, carbon-hydrogen ratios, sediment thickness – in hundreds of devices to provide information for electronic systems and automatic controls.

Although the use of strontium-90 in instruments was important, it meant little to us in achieving our goal of utilizing fission product waste. Most instrument sources contain only millicurie amounts of strontium-90; we could get a year's supply by rinsing our processing

equipment. Furthermore, other radioisotopes can be and are used in instruments. What we needed were uses that required megacuries – a thousand million times as much. This would have to be for heat, power, and industrial beta irradiators.

Opportunity knocked when the Space Age dawned. For a totally independent heat-power source, a nuclear reactor or "the other nuclear power" from the decay of radioactivity was needed. An AEC-multiagency program labeled SNAP (Systems for Nuclear Auxiliary Power) became the main avenue for the development of isotopic power. SNAP-1, which died aborning, was to use fission product cerium-144 to boil mercury to operate a tiny turbogenerator. As it turned out, fission products never did get into space because of questions about safety and strong competition from solar cells and chemical systems. The several isotopic power generators that did get into space (fueled with plutonium-238) have been performing steadily for many years.

The early efforts on isotopic power for space did lead to terrestrial applications such as power for remote weather stations. The first of these, an automatic weather station using thermoelectric power, was built in 1961 by the Martin Company, and the Laboratory's Isotopes Division prepared a 17,500-curie, 110-W $^{90}SrTiO_3$ source and installed it in the unit. The weather station was placed on Axel Heiberg Island in the Arctic, where it performed perfectly for three years, radioing weather data every few hours. It was closed down because of a minor electronic malfunction not related to the strontium-90 power source. Members of the Isotopes Division performed a postoperational destructive examination on the doubly encapsulated source and found all components to be in excellent condition. Up to the end of 1967, 36 sources were fabricated, containing a total of 445,000 curies of strontium-90, generating 3130 W of heat, thermoelectrically converted to 125 W of electricity. These sources were used by the U.S. Navy, Coast Guard, and Weather Bureau and for the British RIPPLE generators. The uses were for fixed and floating weather stations, buoys, sonic beacons, submarine cable repeaters, and aircraft ground radio beacons. The largest units put out 60 W of electric power.

In 1968, strontium power, now pretty well proved in terrestrial uses, passed another milestone. Commercial equipment manufacturers designed fairly standardized power generators to be made in larger lots; now we could fabricate 10 or 15 units of the same design instead of having to do custom fabrication on each source. This allowed some manufacturing economies to be made, although the scale is still too low to allow the cost reductions necessary to make strontium power widely competitive.

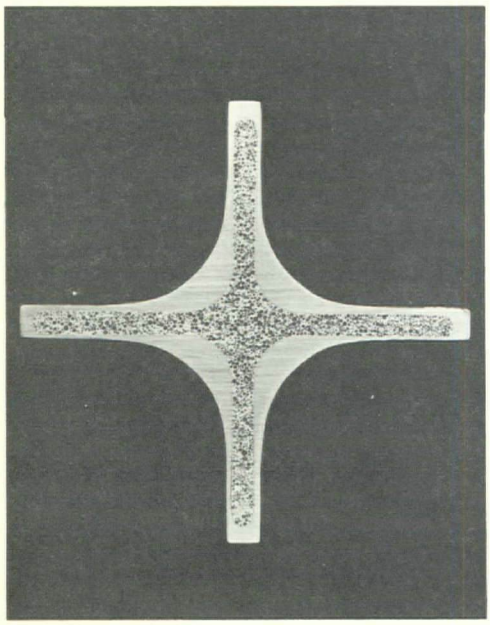
There is a constant effort to improve the output and efficiency of thermoelectric devices. In 1968 and the first two months of 1969, 15 sources of various kinds were fabricated, containing 1,400,000 curies of strontium-90, with a total heat output of 9480 W; converted by improved thermoelectric units, this would represent about 570 W of electricity.

Isotopic power is expensive, but it occupies a unique position in providing heat or electricity under very difficult environmental con-

ditions. At present, strontium-90 thermoelectric units furnish power at 3¢ per watt-hour over a ten-year life. Such a cost would be absurd when compared with house electricity; however, in a more appropriate comparison with dry-cell batteries at ~6¢ per watt-hour, which also have a much shorter useful life, it is seen to be not so unreasonable. There is still much room for improvement in isotopic power costs. In systems large enough to generate a kilowatt of electricity, heat-engine–generator systems can be used to improve efficiency by a factor of 2 or 3 over thermoelectric devices, thus markedly lowering unit costs.

Direct (or indirect, through heat exchangers) heating is also needed for small areas in difficult environments, for example, warming electronic equipment, chemical batteries, or diesel fuel in Arctic locations, or underseas. Tiny sources of a few microwatts can be used for spot heating of electronic components; kilowatt units large enough to heat a small insulated room in winter are quite feasible; specially insulated large sources rise to temperatures as high as 1600°C and could serve as small furnaces. Radioactivity is of course a factor in all applications. Even though strontium-90 and yttrium-90 emit only beta radiation, which is largely internally absorbed in large sources, the bremsstrahlung generated makes shielding necessary; for example, a 10-W strontium-90 source requires about 3.75 in. of lead to reduce the radiation field to 10 mR/h at 1 m.

*Cross section of one type of strontium-90 radiation source configuration showing strontium silicate minibeads in an aluminum matrix. 6×.*



That brings us to the other general technique for using strontium power, which technically is a mirror image of the problem just discussed. The energetic beta particles emitted by yttrium-90 make it potentially useful for process irradiation, supplementing electron generators and complementing the large-scale gamma irradiation sources, cobalt-60 and cesium-137. Strontium-90 is the only long-lived energetic pure beta emitter available in very large amounts. We have therefore concentrated our efforts on the problem of safely containing strontium-90 in a source that will allow a large fraction of the beta radiation to penetrate the covering. Source material was prepared by making the silicate compound of strontium-90, which was fused in glasslike spheres 0.1 to 0.4 mm in diameter, called "minibeads." Stainless steel source containers in a variety of shapes, such as thin blades, plaques, and annular cylinders, were filled with minibeads, placed in a vacuum furnace, and invested with molten aluminum, forming a strontium silicate bead–aluminum matrix. A test irradiation source made up of blade-shaped components will be assembled for use in experimental beta irradiation of packaged beef at Brookhaven, primarily for surface pasteurization to improve shelf life. We are studying the use of similar sources in promoting high-pressure industrial oxidation processes, in the destruction of organic pollutants in waste water and air streams, and in the polymerization of plastic films and fibers.

We have found that strontium-90 can be tamed and made useful, when treated with due respect. The megacurie amounts now being separated and used for many applications indicate that this is a good direction to go, not only for waste by-product utilization but also for waste disposal. Work during the past several years has clearly shown that strontium power can be made to serve man.

# THE RECORD OF ACCOMPLISHMENT - 1968

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| BOOKS AND BOOK ARTICLES | 61 | 62 | 23 | 146 |
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