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Transuranic Elements and The High-Flux Isotope Reactor

A high-flux isotope reactor, designed and built at Oak Ridge National Laboratory, will produce 1 g/yr of Cf^{252} and appreciable amounts of other transuranic isotopes. Along with the reactor, a complex processing and extraction plant and a laboratory for investigating these elements make a unique facility for transuranic research.

by Alvin Weinberg

A TRANSURANIC-ELEMENTS research and production facility is being completed at Oak Ridge National Laboratory. Its three principal components are a High Flux Isotope Reactor (HFIR), an elaborate radiochemical processing plant called TRU and a transuranic research laboratory. Built over the past eight years at a cost of more than $24.5 million, this facility provides scientists from many different institutions with tools to advance understanding of the chemistry and nuclear physics of the transuranics and also to advance the technology of high-flux reactors.

Because generation of transuranic elements occurs by neutron bombardment, the need for high fluxes of neutrons is crucial. ORNL has used flux trapping to obtain 3.5×10^{15} thermal neutrons/cm^{2}sec in an appreciable volume without burning the reactor fuel at such a rate as to require too frequent refueling. Elements containing Pu^{242} are irradiated in the high-flux volume of HFIR; then transuranics are extracted in TRU and studied in the transuranic research laboratory. By 1971 we expect to produce 1 g/yr of Cf^{252}, 0.1 g/yr of Bk^{249} and Cf^{249}, 10 mg/yr of Es^{253} and 1 mg/yr of Es^{254}.

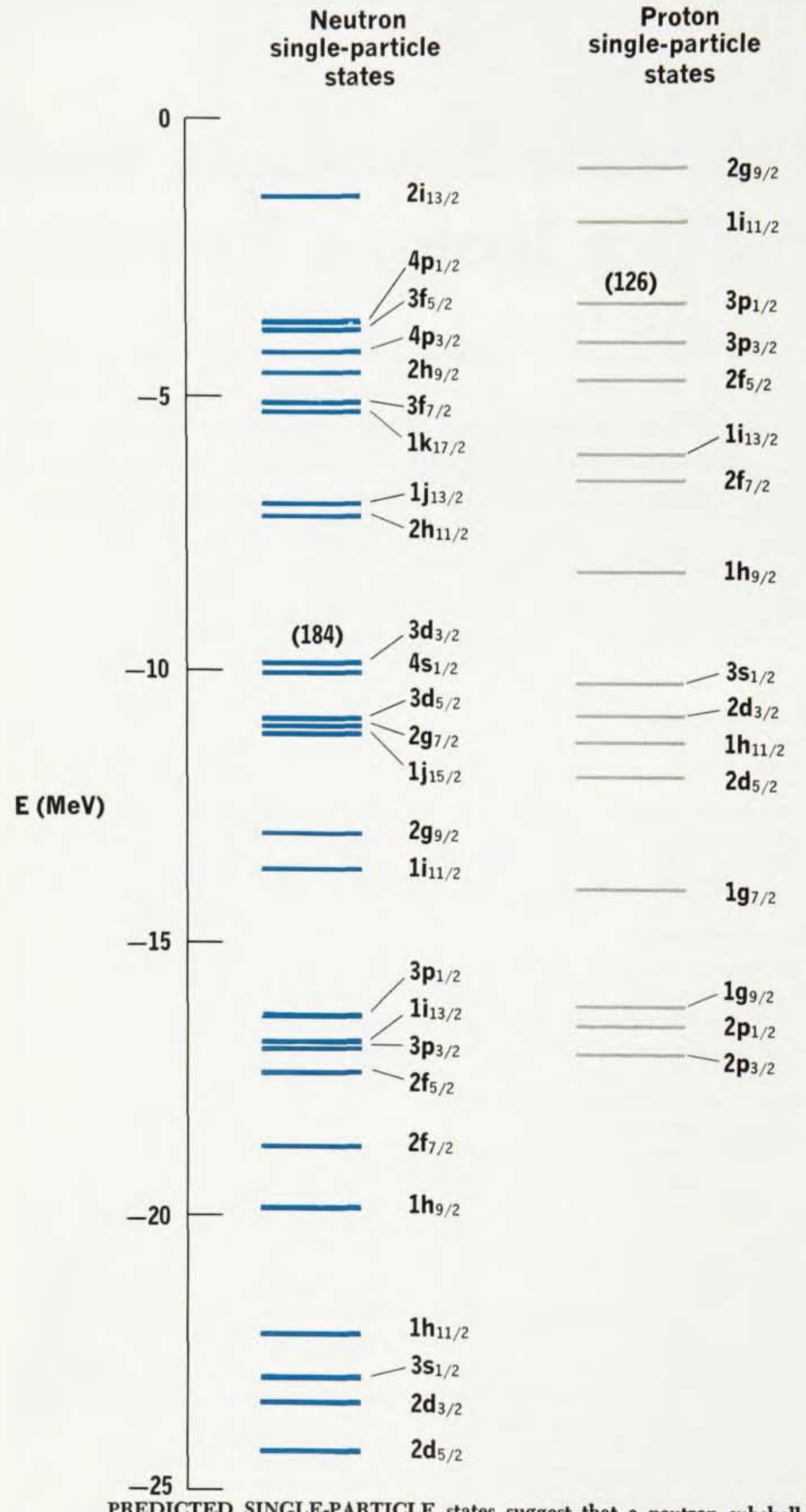
Transuranic vistas

In nuclear structure the transuranic elements are unique: First, together with isotopes of uranium, they are the only elements which undergo slow-neutron fission; second, they are the only highly deformed nuclei that generally decay by alpha emission.

Highly deformed nuclei have been of special interest since Gösta Nilsson suggested in 1955 that the levels in such nuclei group into basic states (characterized by different values of the quantum number K) upon each of which is built an elaborate fine structure of different rotational states. These states can be seen in Coulomb excitation and to some extent in beta decay; however, with the high precision, lithium-drifted germanium counters the decay schemes deduced from alpha particle spectra are very reliable and relatively easy to obtain. Moreover, selection rules in alpha decay reveal rotational spectra very clearly so that heavy elements afford an additional and unique degree of freedom to the nuclear-structure physicist. Except for a few rare exceptions like Sm there are no other highly deformed nuclei that emit alpha particles. The consequences of the Nilsson model are therefore best studied in the alpha-decaying transuranics.

A second more speculative interest

The author is a long-standing contributor to reactor theory and design. He got his PhD from Chicago in 1939 and then joined Eugene Wigner's wartime group on reactor design. Later he and Wigner wrote the well known book *The Physical Theory of Chain Reactors.* In 1945 he went to Oak Ridge, and in 1955 he was made its director. He is also known for his contributions to discussions of public policy and science.



PREDICTED SINGLE-PARTICLE states suggest that a neutron subshell closes at N = 184 and a proton subshell closes at Z = 126 or 114. —FIG. 1

of the ultraheavy elements arises from the possible stability of element $_{126}X^{310}_{184}$ or $_{114}X^{298}_{184}$. These remarkable possibilities were first raised by William D. Myers and Wladyslaw J. Swiatecki in 1965. Myers and Swiatecki predicted, using a new empirical mass formula, that these elements may be stable against spontaneous fission. The order of filling neutron and proton states in a spherically symmetric potential (whose parameters are based on experimental data obtained from lower mass regions) is shown in figure 1. This level scheme suggests that a neutron subshell $3d_{3/2}$ closes at $N = 184$ and that proton subshells should close at either $Z = 126$ or $Z = 114$. The filling of the subshell at 114 leads to a nucleus of mass 298 closer to the beta stability line, but since the resulting nucleus is probably not spherical, the energy levels in figure 1 may be less reliable for $N = 114$. Thus the nuclei $_{126}X^{310}_{184}$ or $_{114}X^{298}_{184}$ ought to be doubly magic. Consequently, these nuclei ought to be relatively stable against spontaneous fission. Myers and Swiatecki predict the fission barrier height for $_{126}X^{310}_{184}$ will be about 9 MeV, which corresponds to a fission half-life of about 10^8 sec. Cheuk-Yin Wong has examined theoretically the stability of these superheavy elements against alpha and beta decay and estimates the half-life for beta decay to be 10^9 sec, the half-life for alpha decay to be of the order of 10^4 sec and probably considerably longer. The superheavy elements might be produced by bombarding Pa with Br ions of around 450-MeV energy

$$_{91}Pa^{231} + {}_{35}Br^{81} \rightarrow {}_{126}X^{310}_{184} + 2n - 315 \text{ MeV}$$

or Cf with Ni ions at around 350 MeV

$$_{98}Cf^{249} + {}_{28}Ni^{64} \rightarrow {}_{126}X^{310}_{184} + 3n - 276 \text{ MeV.}$$

If such startling reactions could be made to occur, then the entire series of elements between $_{103}Lw$ and $_{126}X^{310}_{184}$ would presumably become accessible through successive alpha decays provided they were not overwhelmed by fission. Thus, nuclear-structure physicists would have some two dozen new elements on which to try their nuclear systematics and their various models.

For inorganic chemists the transuranic elements have presented a major challenge since 1944 when Glenn T. Seaborg pointed out that the actinide-series elements were analogs of the rare earths with the electrons going into the 5f shell. The question was then raised: What happens when the 5f shell is completed? Will element 104 be an analog of Hf, the first element past the 4f rare earths, as suggested by Georgii N. Flerov and his coworkers at Dubna, or will something entirely different happen? To answer these basic questions in organic chemistry, we need enough very heavy elements—like elements 105 and 106—to be able to perform chemistry.

Nuclear engineers can be expected to find the transuranics of great interest. The role of Pu^{239} is well known, but it is not so well known, that Cm^{240} ($T_{1/2}$ = 163 days) and Cm^{244} ($T_{1/2}$ = 18.1 years) are extremely useful as heat sources for satellite power plants and, perhaps eventually, for cardiac pacemakers. It is remarkable that Cm^{242}, which was discovered in 1944 and remained a laboratory curiosity for many years, is now produced in multigram quantities for use as a heat source. In a recent speech Seaborg predicted that we may eventually use tons of Cm^{244} as a convenient source of heat.

Transuranium program

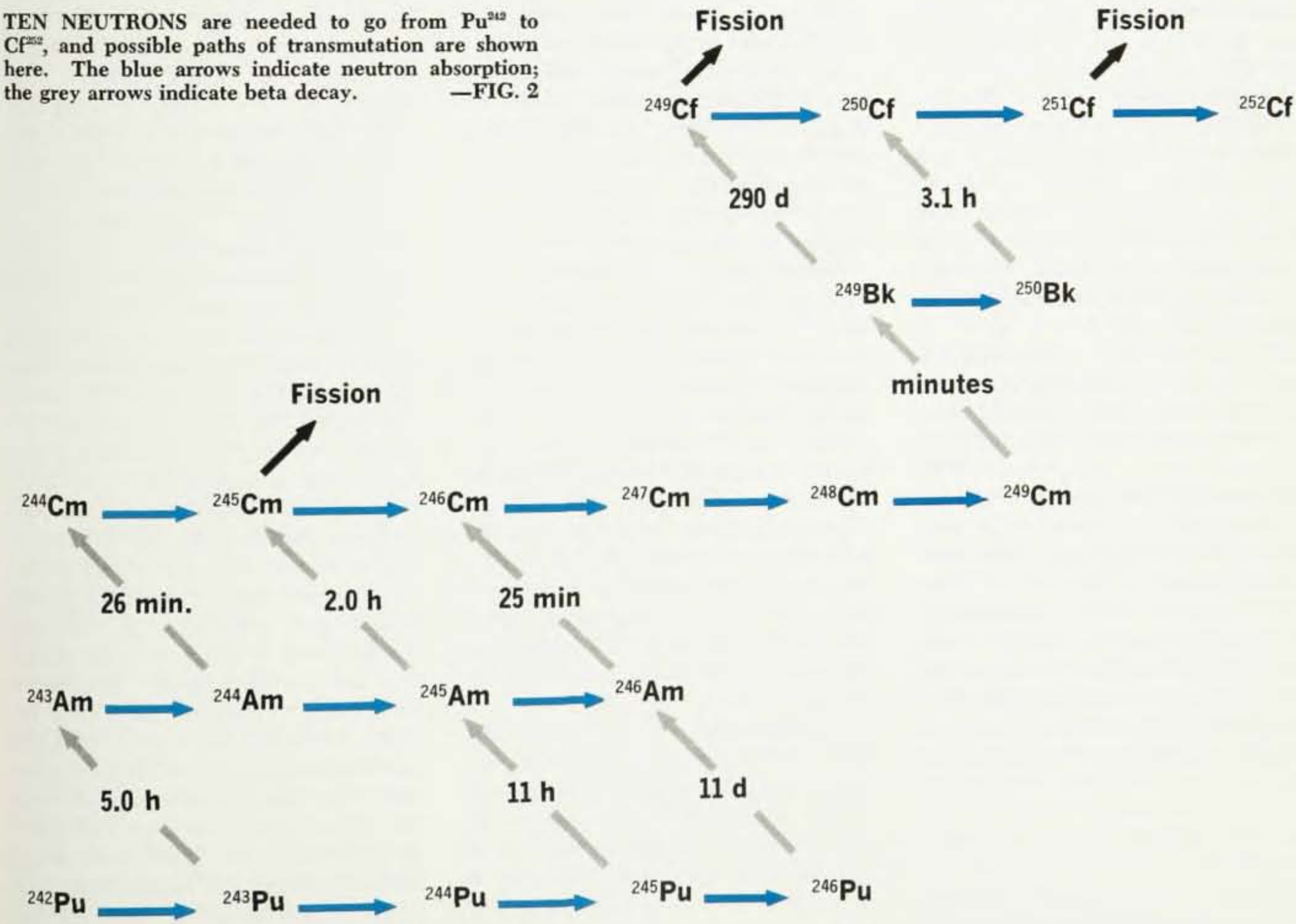
Because of the many interests in heavy transuranic elements, and particularly the advantage of doing transuranic inorganic chemistry on a macroscopic scale, the Atomic Energy Commission in 1958 decided to produce large quantities of these elements for use by researchers throughout the United States. It was most logical to aim for the isotope Cf^{252} since this is the heaviest transuranic with a conveniently long half-life—2.6 years. Starting with massive targets of Cf^{249} or Cf^{252} one could bombard with, say, C^{13} or N^{15} ions in a linac or an isochronous cyclotron and produce elements 104, 105 and possibly even heavier elements.

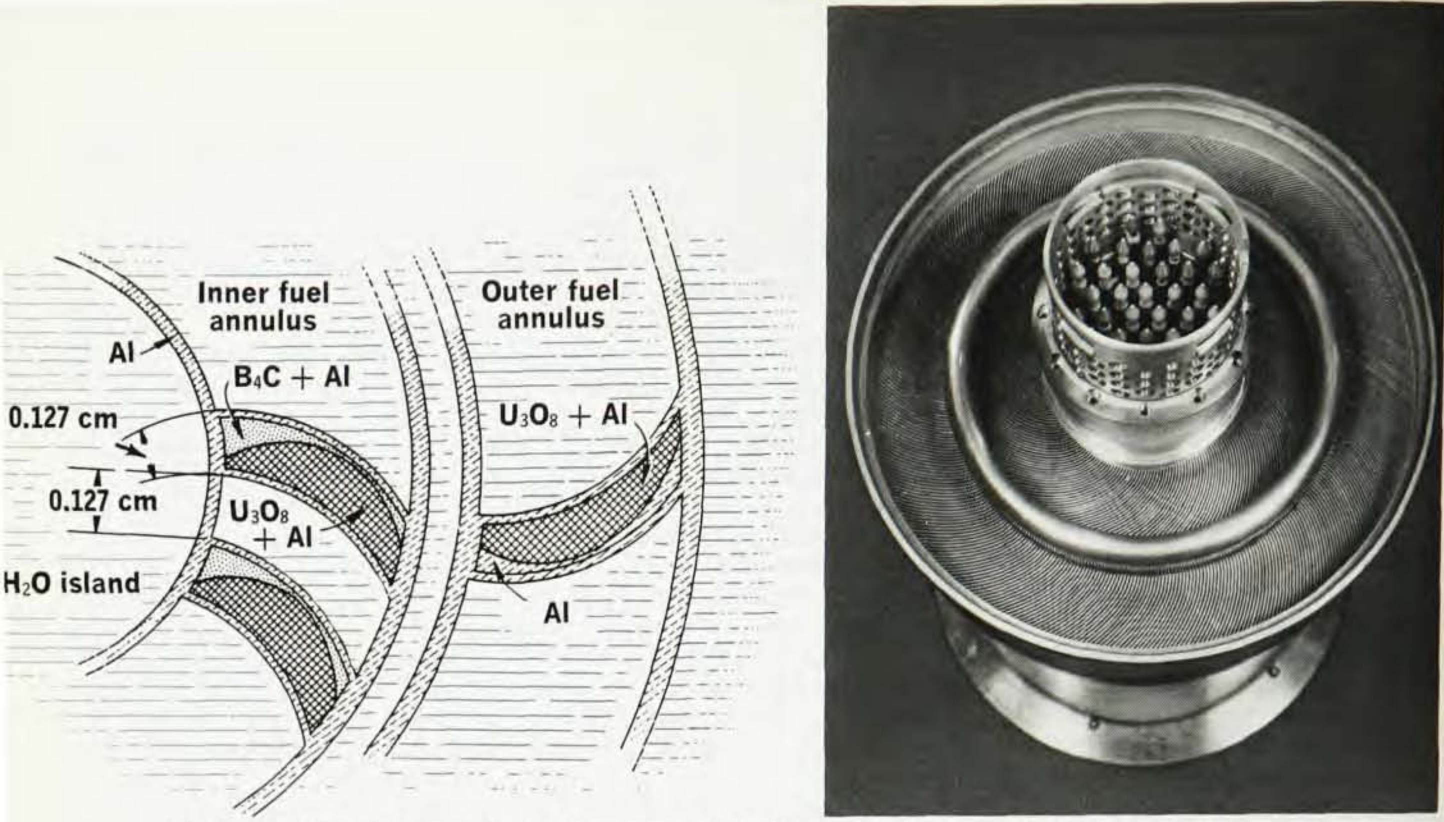
The most readily available isotope from which to produce Cf^{252} is Pu^{242} ($T_{1/2} = 3.8 \times 10^5$ years); it is produced in large quantities in the Savannah River reactor by irradiating Pu^{239}. During the neutron irradiation about 27% of the Pu^{239} is transformed into Pu^{240}, the remainder being eventually destroyed by fission; the Pu^{240}, being nonfissile, goes to Pu^{241}, 29% of which on further neutron bombardment converts to Pu^{242}.

To go from Pu^{242} to Cf^{252} requires 10 neutrons (figure 2); hence, the rate of production of Cf^{252} would at first be proportional to ϕ^{10}, ϕ being the neutron flux. As the irradiation continues, some of the intermediates burn out, and in consequence the dependence of the production rate upon flux is less than the tenth power. The rate of production of Cf^{252} in the HFIR over a one-year period is roughly proportional to the third power of the neutron flux.

But even with the weaker depen-

TEN NEUTRONS are needed to go from Pu^{242} to Cf^{252}, and possible paths of transmutation are shown here. The blue arrows indicate neutron absorption; the grey arrows indicate beta decay. —FIG. 2





FUEL ELEMENT of the HFIR is shown at right with target rods in the central trap. To reduce "hot spots" the fuel elements are graded as shown in the schematic of the fuel-element cross section shown at left. —FIG. 3

dence on neutron flux, there is strong incentive to irradiate Pu^{242} with the highest possible flux. Thus it was decided in 1960 to build a reactor, the High Flux Isotope Reactor (HFIR), that would achieve a thermal flux approaching 5×10^{15} neutrons/cm^2 sec, specifically for the production of 1 g of Cf^{252} per year. Water was chosen as the moderator and coolant, and U^{235}–Al as the fuel element material largely because this combination had performed so well in the many predecessors of the HFIR.

Although the HFIR is intended primarily as a source of slow neutrons for producing transuranic elements, several experimental beam tubes also have been built into it. Four beam holes look directly at the reflector region of the reactor; two of these can be converted into a through hole. The slow-neutron current at the exit of these tubes is around 10^{10} neutrons/sec. HFIR should therefore provide unexcelled, though limited, facilities for neutron diffraction. In addition, there are 38 holes in the reflector, in several of which the neutron flux is around 10^{15}.

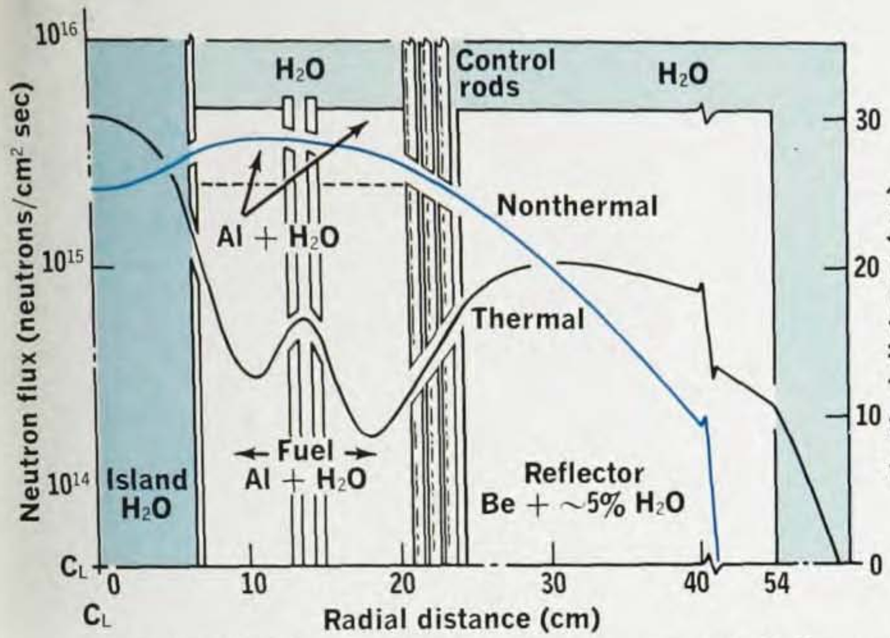
The HFIR

To achieve a flux approaching 5×10^{15} neutrons/cm^2 sec in a heterogeneous, solid-fuel-element, water-moderated reactor obviously poses major engineering problems. For at this flux the U^{235} fuel in the reactor is burned to $1/e$ of its initial value in only 3 1/2 days. Thus the reactor would be largely a refueling machine unless some means were achieved to separate the very high flux, where the Pu^{242} is to be cooked, from the U^{235}, which supplies the neutrons.

Flux trapping. The HFIR therefore embodies the flux-trap principle, an idea first used to some extent in the Materials Testing Reactor and then exploited in detailed design by Richard M. Stephenson and Milan F. Osredkar. In the flux-trap reactor the densely packed fuel region (figure 3) surrounds a water-filled hole in which the target rods sit. Neutrons from the fuel region enter the hole, are moderated there, and since the lifetime in the water hold or flux trap is much longer than it is in the fuel region, the neutron flux builds up in the flux trap (figure 4).

Construction. In the HFIR 9.4 kg of U^{235} occupy a cylindrical annular region with an inner diameter of 14 cm and an outer diameter of 42 cm. The average flux in the fuel at full operating power of 100 MW is about 3×10^{14}, whereas flux in the flux trap when unoccupied reaches a maximum of 5×10^{15} neutrons/cm^2-sec. Of course when the flux trap is filled with the target rods of Cm^{244} or Pu^{242}, the average slow neutron flux in the target rods at full power is only about 2×10^{15}. At the average flux in the fuel region of 3×10^{14}, about 1.3% of the fuel is burned each day; this causes the reactor to lose 0.4% reactivity per day. The cycle time of the reactor is about 22 days, which is quite tolerable even though a complete fuel loading costs around \$75 000.

The aluminum fuel plates containing the U^{235} extend radially outward from the edge of the flux trap. The plates are shaped like involutes, so they fill space compactly and uniformly. The fuel region is surrounded by a beryllium reflector. The flux near the central trap and near the Be reflector is higher than it is at the middle of the annular fuel region. Hence, the fission density and, therefore, the heat production tend to peak near the water trap and the Be reflector. To flatten the power generation (and thus remove hot spots that would limit the maximum flux achieveable in the reactor) the uranium is graded through the width of each fuel plate, being less concentrated at the flux-trap edge and reflector, where the flux is high, and more concentrated at the middle, where the flux is low. To so tailor the



FLUX TRAPPING occurs when neutrons from annular fuel elements are moderated in a central region and build up there in flux. —FIG. 4

uranium concentration, the fabricators of the HFIR fuel elements sandwiched a tapered thickness of U–Al cermet with a suitably tapered aluminum plate between the aluminum cladding (figure 3). Furthermore, to lengthen the time between fuel changes and reduce the reactivity control requirements, the heavily absorbing B^{10} was incorporated in the fuel elements. Thus, as the U^{235} is burned, so is the B^{10}, thereby compensating partly for the loss in reactivity caused by the burning of the U^{235}.

The entire fuel assembly and reflector is contained within a stout pressure vessel, 2.5 m in diameter and about 6.7 m high. 60 500 liters of water, pressurized to 4200 kg/m², pass through the reactor core each minute. The maximum heat flux in the fuel region is 6.6×10^6 W/m². The reactor in its pressure vessel is located at the bottom of a 12-m deep pool of water. The water serves to shield the reactor during operation and allows workers to maintain the reactor during shutdowns by using longhandled tools.

HFIR is controlled by two neutron-absorbing metallic cylinders, located between the outer fuel annulus and a beryllium reflector, that regulate the flow of neutrons between the fuel and the reflector. The two cylinders move in opposite directions so that the control absorbers are always disposed symmetrically on either side of the median plane of the reactor. Thus, the fuel burns out much more evenly than is customary in a compact water-moderated reactor where the control rods usually enter only from the top. The control plates are graded: a relatively "transparent" section of aluminum followed by a "grey" section of tantalum–aluminum and then by a "black" section of europium-oxide dispersed in aluminum.

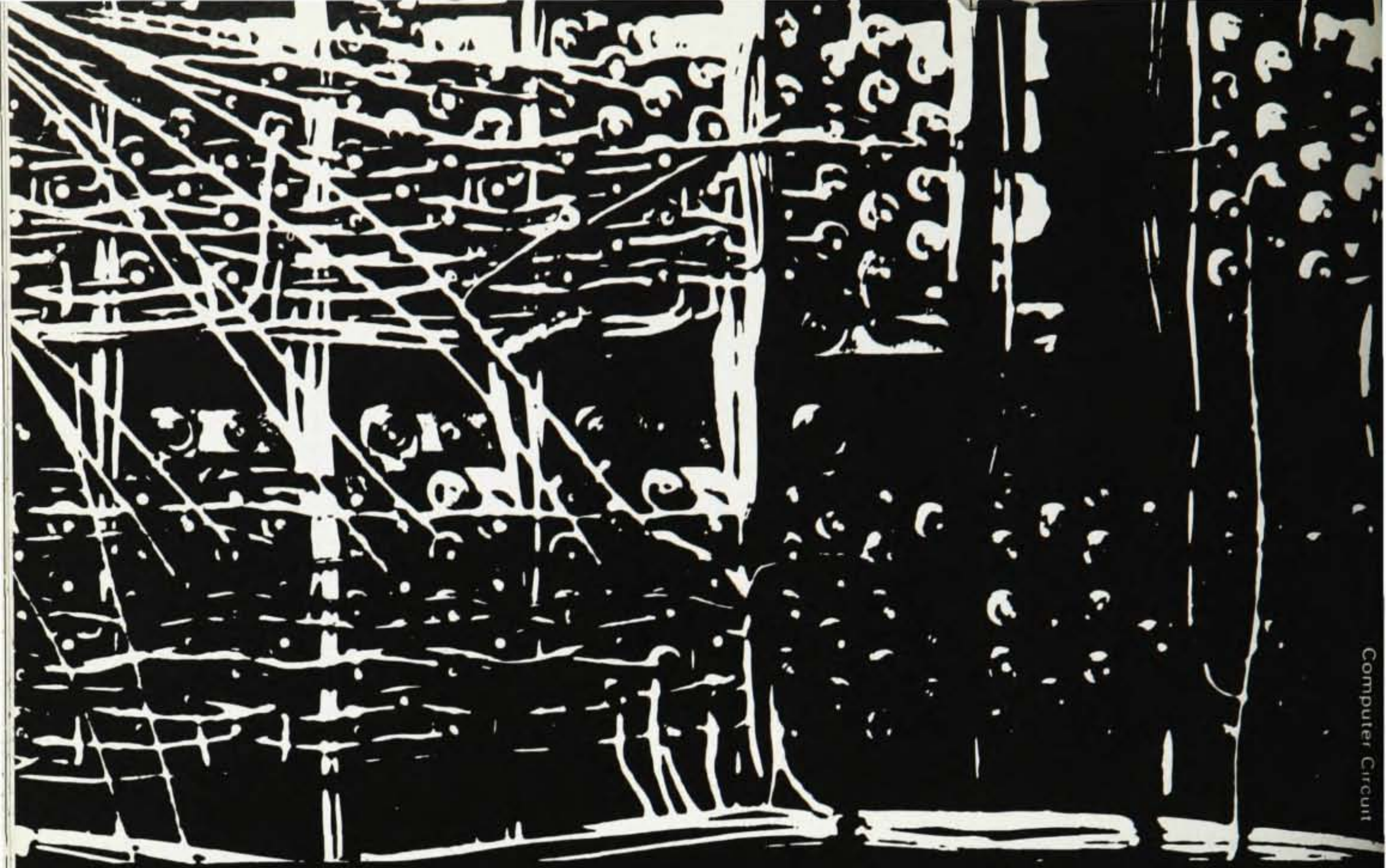
Control of HFIR is dominated by the buildup of the heavily absorbing Xe^{135}, with its absorption cross section of 3×10^6 barns. During operation the Xe is maintained at a low concentration by burnup in the intense neutron flux. Should the reactor be inadvertently scrammed, the Xe^{135} produced by the decay of its precursor 6.7-hour I^{135} builds up until the reactivity is reduced by about 40% after 12 hours; approximately 90% of the Xe^{135} decays in another 24 hours.

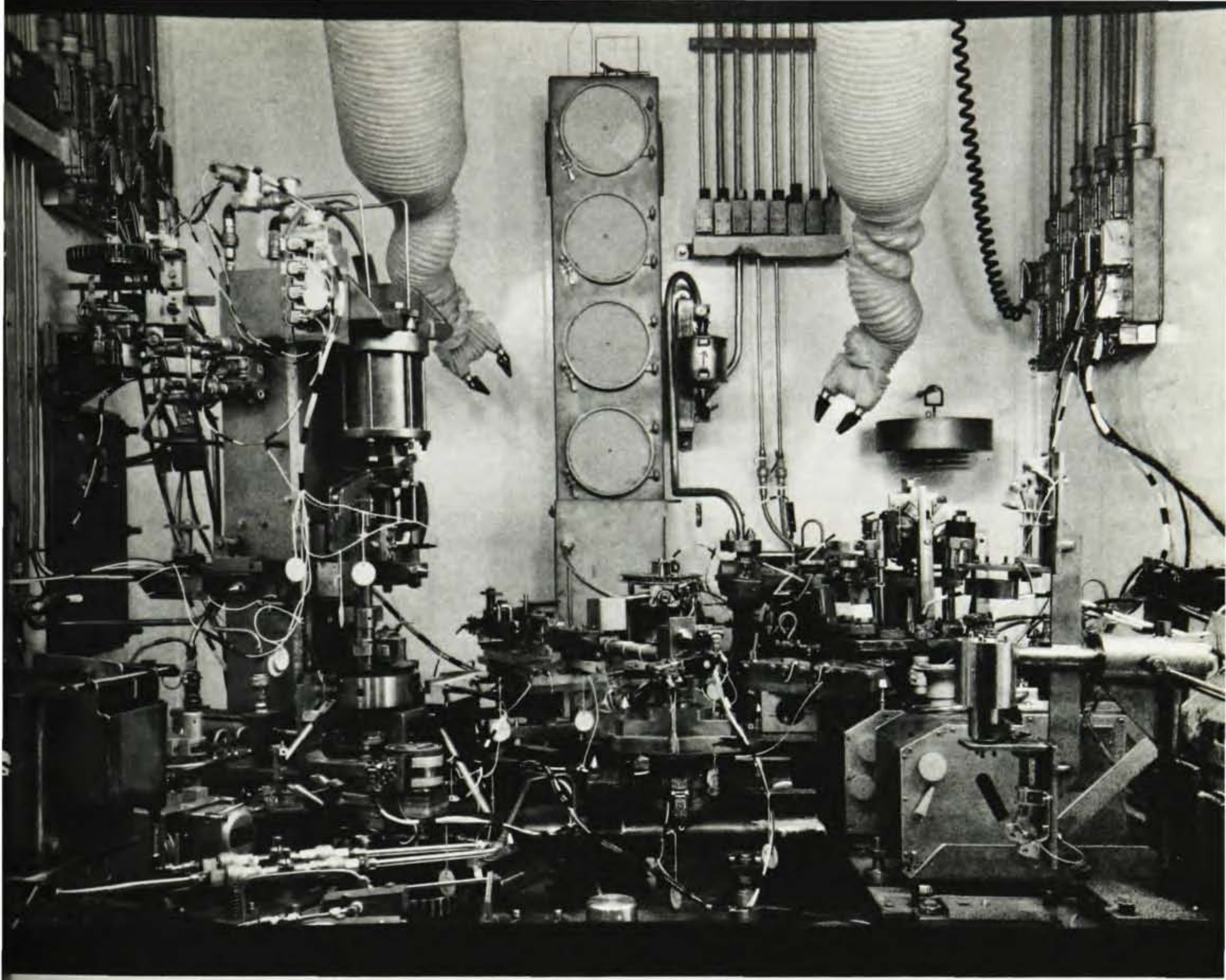
To override xenon at all times after a shutdown is out of the question since this would require a dangerous number of critical masses in the reactor. Moreover, since the reactor consists of only two large fuel elements, one cannot replace a few xenon-poisoned fuel elements with new fuel elements, as is often done in research reactors, and start up the reactor again. One must be able to start the reactor up very quickly after a shutdown—say within 10 to 15 minutes so as not to "lose" a fuel loading. The danger of losing a fuel element is particularly acute toward the end of a fuel cycle when the fuel has already been depleted, and the reactor possesses relatively little margin of reactivity.

The danger is aggravated by another fission-product poison, Sm^{149}, with a yield of about 1%, and a cross section of about 40 000 barns. Stable Sm^{149} is the daughter of 2.3-day Pm^{149}. Thus, during a shutdown, the Sm^{149} can grow by decay of Pm^{149} and can reduce the reactivity by as much as 6%—very much more than its steady-state loss, which is about equal to its fission yield—since the Pm^{149}, having a relatively small absorption cross section, accumulates during operation of the reactor. As the Sm^{149} does not decay, a fuel element might thereby be permanently poisoned and rendered unusable. The phenomenon of "killing a fuel element by Sm^{149} poisoning" has been observed in HFIR fuel operated at 100 MW.

Operation. The HFIR is the most compact of the modern generation of water-moderated, enriched-uranium reactors. At 100 000 kW its power density reaches a peak of 4 megawatts/liter (MW/l). It therefore represents the culmination of a line of water-moderated research reactors that began with the Materials Testing Reactor (40 000 kW, peak power density = 0.8 MW/l) and includes the Engineering Test Reactor (175 000 kW, peak power density = 1.2 MW/l) and the various swimming-pool reactors.

Construction of HFIR began during June 1961, and the reactor first became critical on 25 Aug. 1965. Power was increased in several rather uneventful steps, and HFIR first achieved 100 MW on 9 Sept. 1966. At this power the slow neutron flux in the trap, filled with a dummy aluminum target, reached 3.5×10^{15} neutrons/cm² sec, which we believe represents the highest slow-neutron flux ever reached in a research reactor, although somewhat higher fluxes have been reached in special loadings in the Savannah River production reactors. (The SM reactor in the Soviet Union


Computer Circuit



FUEL FABRICATION CELL in the elaborate radiochemical plant (TRU) in which the target rods are processed. All processing steps must be done remotely. —FIG. 5

is reputed to be capable of achieving fluxes higher than 10^{15} neutrons/cm^2 sec; but we do not know the exact status of this reactor. Its operation at 50 MW was reported at the 1964 Geneva Atoms for Peace Conference.)

Altogether, the operation of HFIR has been extremely smooth. We were pleasantly surprised, though a little puzzled, that the fuel lasted as long as 22 days at 100 MW, this being somewhat longer than the limit of the estimated fuel life (18 days). But, except for a few very minor and easily remedied difficulties, the reactor has behaved exactly as planned.

The technology of HFIR should be widely useful in research reactors. For with the flux-trap principle, as exploited in HFIR, one gains a factor of at least 4 in maximum flux per unit of power. Hence many university research reactors, which now operate at 1 MW and yield a maximum flux of 2×10^{13} neutrons/cm^{2}sec might, by using the flux-trap arrangement of HFIR, achieve a flux of close to 10^{14} neutrons/cm^{2}sec without a corresponding increase in power.

Transuranic production and research

The Pu^{242} cooked in the flux trap is contained as PuO_2–Al cermet. The cermet is fashioned into pellets that are contained in long, thin, aluminum rods. Thirty-one rods comprise a complete target assembly that sits in the central flux trap. The target assembly generates 0.5 MW of heat. At intervals of about a year (the exact interval is determined by the radiation damage to the target assembly) each target rod is reprocessed in a very elaborate radio-chemical plant, the TRU, located next to the HFIR. After the rods are dissolved in strong HCl, the transuranic elements are separated from fission products and aluminum by extraction into an organic solvent, a mixture of a tertiary amine and diethyl benzene. The Am and Cm are isolated from the heavier transuranics by extraction of the heavier elements with di-2-ethylphenyl phosphonic acid and are returned to the HFIR target to be irradiated again.

Berkelium is then oxidized and separated by solvent extraction, and then the Cf, Es, and Fm are separated from each other by chromatographic elution in ion-exchange columns. To increase production of heavy elements, it is necessary to recycle the target material. The TRU facility therefore includes a complete remotely operated metallurgical processing line. Pu, Am, and Cm are converted into oxide and then mixed with aluminum powder and compacted and refabricated into new target rods.

All of these radiochemical and metallurgical steps must be done remotely. The almost fantastic intricacy of the various steps can perhaps best be judged from a picture of the pelletizing and final fabrication cells in the TRU facility (figure 5). Several of the isotopes handled in TRU, notably Cf^{252} and its contaminant Cf^{254}, decay by spontaneous fission with rather short half-life—66 years for Cf^{252}, 60 days for Cf^{254}. Thus one gram of production grade Cf^{252} with its Cf^{254} contaminant emits as many fast neutrons as does a water-boiler reactor operating at several hundred watts!

Though this intense neutron emission from the product californium makes it an unequalled "point" neutron source, it also makes it difficult to handle. The concrete shielding in the cells of TRU therefore contain an aggregate that is rich in water and is a good neutron shield.

The TRU facility has already completed several runs and has produced about 2 milligrams of Cf^{252} and 30 micrograms of Bk. The production rate will increase gradually until, by 1971, we expect to produce 1 g/yr of Cf^{252}, 0.1 g/yr of Bk^{249}, 10 mg/yr of Es^{253}, and 1 mg/yr of Es^{254}.

The transuranics produced at ORNL will be distributed to qualified research workers throughout the world. Since some of the interesting species are rather short-lived, ORNL has itself embarked on a major investigation of the chemical and physical properties of the heavy transuranics. This work will be carried out in the newly completed Transuranium Research Laboratory. This laboratory contains the usual glove boxes and lead shielded box needed to work with intensely active and dangerous alpha emitters. In addition, it will contain a concrete-shielded-box facility that has enough neutron shielding to allow a researcher to work with up to 10 mg of Cf^{252}.

When the research is fully in progress, we expect as many as 30 physicists and chemists to participate. Many of these scientists are expected to come from universities, so that the Transuranium Research Laboratory will have much the same flavor as any very large and unique facility to which many visitors come to carry on their research.

Beyond HFIR

The HFIR and the TRU represent tour de forces of engineering that I believe will influence the design of future reactor and radiochemical processing plants. As for reactor technology, the HFIR represents what appears to me to be close to the ultimate in solid-fueled, water-moderated reactors. Its average power density is about three times higher than the power density of other research reactors and exceeds the power density of water-moderated power reactors by an even higher factor. I have already alluded to the flux-trap principle as a means of locally increasing the flux without raising the power very much; also of significance is the idea of a single, large, annular fuel element. Many new research reactors do embody one or both of these principles; for example, the newly authorized Franco-German reactor in Grenoble uses a single, large, annular fuel element, and the Argonne Advanced Research Reactor will use the HFIR fuel element.

What about moving beyond 5×10^{15} say to 10^{16} or even 5×10^{16} neutrons/cm²sec? I have already pointed out that this is very difficult in a compact, enriched-solid-fuel reactor because the fuel is burned so fast that the reactor becomes a refueling machine. Nevertheless, three paths seem available to reach the goal of 10^{16} or higher. Perhaps the simplest though most expensive way is to generate enormous amounts of power, of the order of a million killowats, in a heavy-water reactor containing relatively little U^{235}. Richard D. Cheverton of ORNL has estimated that a larger HFIR-type reactor using D_2O as moderator and H_2O in the flux trap and operating at 750 MW might produce an unperturbed flux of 3×10^{16}. Such a reactor would of course require better fuel elements than are now available.

A second method, much under discussion now, is to generate fast neutrons by means of a very intense high-energy accelerator and "flux trap" the neutrons in a large heavy-water bath surrounding the target. This possibility is under active consideration in Canada. In one version of the Intense Neutron Generator (ING) the accelerator is a separated orbit cyclotron—in another, a huge linac. In either case the beam power is around 65 000 kW, and the cost is very high—\$125 million.

A third way, and one that gets around the problem of rapid fuel recycle, is to use a solution of uranium salt instead of solid fuel plates. One can visualize a graphite sphere of perhaps 60 cm diameter surrounded by an annulus of say 5 cm thickness and by an outer graphite reflector. Through the annulus would be pumped a molten salt of $U^{235}F_4$–Li^7F–BeF_2. William K. Ergen has estimated that fluxes in excess of 10^{16} could be achieved in the central graphite core of such a reactor with a power output of about 10^6 kW. Since the fuel is molten, refueling is very simple. However, one must deal with an intensely radioactive, and difficult to maintain, circulating system. Nevertheless, we have brought into operation at ORNL, at just about the same time as HFIR, a 7.25-MW molten-salt reactor moderated with graphite. It remains to be seen if such a system will be applied to research reactors.

The transuranic elements provide many interesting vistas in physics, in chemistry and in engineering. I suggest that the technology of producing the transuranic elements—a technology represented by HFIR and TRU—may itself lead to unexpected new applications. One must remember that the technology of research reactors of 15 years ago, represented by the MTR, gave birth to the very successful technology of water-moderated power reactors. So I think that attempts to push to even higher fluxes and larger quantities of transuranic elements, our Canadian friends' ventures into electrical accelerator–reactors or our own ventures into molten-salt reactors, may open even broader and as yet unsuspected technological and scientific vistas.

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The success of the project is due to contributions from many people. The paper is based on an invited talk at the Southeastern Section APS meeting in Nashville last December. □