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Remarks by
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MASS PRODUCTION AND PRACTICAL APPLICATIONS OF ACTINIDE ELEMENTS

INTRODUCTION

I am indeed honored to be the introductory speaker at this five-day symposium on macroscopic studies of the actinides. Since almost my entire scientific career has been devoted to the actinides, and since I spent most of that career in this geographic area, it is a great temptation to be nostalgic and reminiscent about the "good old days." However, nostalgia has had its place during the several anniversaries we have celebrated recently commemorating such events as the first weighing of plutonium and the first sustained nuclear fission reaction.

Research on the actinides remains one of the most dynamic areas of scientific endeavor, and one has to look only at the breadth and depth of this symposium to be convinced of this. I will try not to infringe on the contents of the many excellent papers to follow, and perhaps my remarks should therefore be considered an overview of the symposium.

Programs underway for the large-scale production of numerous isotopes of the synthetic actinide elements, together with unexpected practical applications of a number of these isotopes, point to an exciting future in the field of large-scale manufacture and exploitation of these elements. The magnitude of these possibilities is an aspect of the transuranium field that was not foreseen

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even as recently as a few years ago. I shall attempt in this discussion to describe and forecast some of these unexpected and intriguing possibilities.

A modern version of the periodic table (Figure 1) is helpful in explaining the status of actinide synthesis. Each of the eleven synthetic actinide elements (atomic numbers 93 to 103, inclusive) was discovered as the result of its first production and identification in tracer quantities. In the case of the last three actinide elements, mendelevium (number 101), nobelium (number 102) and lawrencium (number 103), the discovery experiments were performed with almost incredibly small amounts -- on a "one atom at a time" basis. This will continue to be the case for the elements beyond lawrencium, the "trans-actinide" elements.

Very exciting -- at least to me -- has been the recent experimental work on the tracer chemistry in aqueous solution of the elements mendelevium, nobelium, and lawrencium. This type of work had not been possible previously with nobelium and lawrencium. A remarkable series of experiments with these elements, using them on a "one atom at a time" basis in chemical reactions, has been performed. E. K. Hulet, R. W. Lougheed, J. D. Brady, R. E. Stone, and M. S. Coops at Livermore, and J. Maly and B. B. Cunningham at Berkeley have observed and studied the (II) as well as the (III) oxidation state of mendelevium. A. Ghiorso, J. Maly, T. Sikkeland, and R. J. Silva at Berkeley have found a very stable (II) oxidation state, as well as a quite unstable (III) oxidation state, of nobelium. And perhaps most exciting of all, A. Ghiorso, M. Nurmia, T. Sikkeland, and R. J. Silva have found, in experiments at Berkeley, that lawrencium exhibits a stable (III) oxidation state. Silva will report at this symposium on these chemical properties of nobelium and lawrencium, derived from classical experiments performed with incredible speed. In the absence of any known longer-lived isotopes of these elements, they managed to determine these definitive chemical characteristics by using as tracers the 3-minute nobelium-255 and the 35-second lawrencium-256! These results are particularly gratifying to me because, in the case of all three of these elements, they support predictions based on the actinide concept. Thus we can feel confident that predictions being made of the chemical properties of the trans-actinide elements now stand on a firmer base.

The discovery of the synthetic actinide elements was followed, for the elements with atomic numbers 93 to 99, inclusive, by their isolation in macroscopic (weighable) quantities. The first isolation of einsteinium (element 99) in large enough quantities to be visible was accomplished just recently by Cunningham and co-workers at Berkeley. In each case this phase of the element's history followed a number of years after its discovery. And following this winning of the element in macroscopic quantities, it has been obtained in ever increasing amounts. Plutonium, of course, has been produced in by far the largest quantity, while the production of and the prospects for the production of the transplutonium elements decreases in a general way with increasing atomic number.

Although the half-lives of the isotopes of these elements suggest that einsteinium is the highest one that will be available in very useful quantities, it is interesting to note that the recent discovery of relatively long-lived fermium-257 (half-life, 95 days) and mendelevium-258 (half-life, 54 days) may make it possible to extend the work with macroscopic quantities to elements 100 and 101. This does not seem to be a likely possibility for trans-mendelevium elements (elements with atomic numbers beyond 101).

This discussion will not be concerned with the production and applications of plutonium-239, which has such well known utility as a source of energy in nuclear weapons and nuclear power reactors. It will be concerned, rather, with (1) the production potentialities and (2) the practical applications of a number of other transuranium isotopes.

With respect to the first of these topics, the present indications are that these other isotopes can be and will be produced in quantities that are amazing, at least to those who recall their birth in extremely small amounts - sometimes as little as a few atoms. We are talking about quantities measured in tons for two of these isotopes, plutonium-238 and curium-244, and in kilograms or hundreds of grams for heavier isotopes up to californium.

If it appears at times that my optimism is beginning to carry me away, let me note right here that the chemistry of plutonium was initially studied with picogram quantities of this element as a tracer, that the first weighing of the element involved a few micrograms, and that we are now predicting without many qualms the production within a few years of plutonium in power reactors in quantities of hundreds of metric tons. The scale-up in quantities thus amounts to about 10^{20}, a number more meaningful to astrophysicists than to chemists!

Starting with the isotope plutonium-239, the sequence of thermal or resonance neutron absorption in nuclear reactors leads to such typical transmutation products as plutonium-242, americium-243, and curium-244. Plutonium-242 can be produced in quantity by special irradiations of plutonium-239 in large reactors such as the AEC's production reactors, and this method has been used up to now to obtain the target material for successive irradiations. The future extensive operation of nuclear power reactors will produce isotopes such as americium-243 and curium-244 in massive quantities, and amounts of other higher isotopes which diminish in quantity with increasing mass number will be formed by neutron irradiation in other reactors. Figure 2 shows the main line of buildup of these isotopes. A projection of the quantities of these isotopes which will be available in future years is given in Figure 3.

With respect to the second topic, the practical applications, there is much more to be expected from work with the actinides than increasing fundamental knowledge, as important as this is. Among the numerous practical applications which can be visualized in the near- and long-term future are the use of plutonium-238 and curium-244 as energy sources in long-lived, compact power units, plutonium-238 to power medical prosthetic devices such as cardiac pacemakers or even artificial hearts, americium-241 to provide radiation for industrial measurement of thickness, density and distance, and californium-252 as a portable point neutron source in neutron radiography, neutron activation analysis, hydrology, and medical therapy.

THE ACTINIDES AS HEAT SOURCES

An excellent indicator of the increased availability of actinide elements is their application in energy conversion systems as sources of thermal power. This technology is only a little more than ten years old, and much less than that if we measure only the period of time from the first practical application.

If an alpha particle emitting isotope is considered as a heat source, we are making use only of the five or so million electron volts of kinetic energy possessed by each helium ion liberated from the parent in the decay process, as compared with forty times this energy liberated in the fission of a heavy atom of roughly the same mass number. Early workers in nuclear physics were inclined to dismiss

the potential for nuclear energy because they did not know about fission, and the energy release rate of an alpha or beta particle emitter did not appear to be too attractive, considering the effort required to obtain from nature macroscopic quantities of such isotopes as polonium-210 or radium-226.

With the advent of the nuclear reactor and our consequent ability to manufacture heavy actinide elements by transmutation from the more stable lighter actinide elements of nature, we now find that there is real merit in considering how to apply their unusual properties to the solution of real problems.

Consider for a moment a typical transuranium alpha particle emitter such as plutonium-238, whose production I shall describe a little later. Its alpha decay half-life of about 90 years is equivalent to an initial energy emission rate of about a half a watt per gram of pure isotope. Now if the decay is integrated over a ten-year period, assuming this to be a reasonable useful life for its application as a heat source, we find that one gram of this isotope has liberated about 42,000 watt-hours of energy (Figure 4). To accomplish this same release of energy by the combustion of a hydrocarbon, we would have had to consume 3,300 grams of butane, for example, and would have had to supply 12,000 grams of oxygen to enable the combustion to proceed. Thus, there is a weight ratio of at least 15,000 between the nuclear and conventional fuels and this would be much larger if my calculations were based on air rather than oxygen and did not assume exact stoichiometry.

Now, obviously, one is not going to use radioisotope heat sources to supplant the butane torch of the home workshop or the electric hot plate or range in the kitchen. Radioisotopes are still expensive, compared to chemical fuels. In the example I just used, one gram of plutonium-238 can be taken to be worth about $1,000 (a cost that will certainly drop in the future). The combined cost of its thermal equivalent in compressed butane and oxygen would be of the order of $10 for the ten-year use period. Of course, the plutonium isotope is not being used very effectively from an economic viewpoint in this example, because only about 7% of its potential alpha particle decay energy would be expended in that first ten-year period, and I have not given any dollar credit to the undecayed fraction remaining. If I prorate the cost accordingly, then the isotopic heat source costs only about $70.

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The most effective comparison, though, is the relative size of these two energy sources. The one gram of plutonium-238, in the form of the oxide, occupies just about one-tenth of a cubic centimeter, while it would be necessary to cart around at least two very large compressed gas cylinders to supply the butane and oxygen.

ENERGY CONVERSION SYSTEMS

Weight, volume, and cost are not the only factors which determine if a radioisotope heat source is practical. Very few requirements have appeared so far for a supply of thermal energy per se. Most applications need electrical power or mechanical power, and thus a means for converting the thermal power of the radioisotope to the other types of power must be provided, and this leads to further technical and economic complexities. The development of energy conversion systems for radioisotope heat sources has been the responsibility of the AEC, although other Federal agencies have participated heavily also - particularly in prototype testing. Some private development of isotopic power supplies has also occurred in recent years.

Three general types of conversion systems have been given the most attention:

- Dynamic
- Thermoelectric
- Thermionic

Dynamic Energy Conversion - Under "dynamic systems" I include schemes which make use of thermodynamic cycles, such as the Rankine cycle, the Brayton cycle, and the Stirling cycle (Figure 5). Engines operating on these cycles have the characteristic of requiring the repetitive use in a closed system of a heated fluid which may or may not change phase during the cycle. Working fluids that have been considered include water, mercury, and inert gases, depending on the cycle. Their output is mechanical power, but by coupling them to a rotating generator we can obtain electrical power readily. These cycles are all in common usage in systems where fossil fuels provide the source of energy, but in this case the working fluid is often employed only once in a so-called "open" cycle. Conversion efficiency is determined by the temperature extremes of the cycle and

can range as high as 50% or so thermodynamically, although this figure would be difficult to achieve in practice.

Thermoelectric Conversion - Thermoelectric conversion (Figure 6) is a more recent innovation and makes use of the Seebeck effect in doped semi-conductors. It has the distinct advantage of requiring no moving parts to convert heat directly to electricity but it, too, has a thermodynamic limitation to conversion efficiency, with actual efficiencies ranging from 5 to 10%, depending on the semi-conductors used and the temperature extremes achieved.

Thermionic Conversion - Thermionic conversion, the principle used in the vacuum diode in which a heated filament emits sufficient electrons to provide a useful current and voltage, also can be employed with thermal energy sources derived from radioisotopes (Figure 7). No moving parts are involved and the efficiencies attainable are projected to be about twice those now available from thermoelectric conversion.

DEVELOPMENT OF PRACTICAL SYSTEMS

All three modes of energy conversion for radioisotopes face the same general development problem: in order to maximize the conversion efficiency and hence reduce the inventory of radioisotope required for a given power output, the heat source and conversion equipment must operate at the highest possible temperature. As chemists, you will not be surprised that most of the effort involved in developing radioisotope power supplies has been concerned with inventing, modifying and testing high-temperature materials, including the chemical form of the radioisotope, its encapsulant, and the materials for construction of the conversion equipment.

The development effort on radioisotope power supplies, now running at about $35 million per year in the AEC's budget, can be justified only because there are near-term and long-range applications for these devices which cannot be met by batteries or other chemical conversion processes. What are some of these applications which we find to be so exciting technically and so important as to warrant the investment being made? Let me categorize them by the nature of the energy requirement.

THERMAL APPLICATIONS

First, there are indeed some applications which need only thermal energy. Consider the case of a space satellite or probe containing an electronics package which has to operate in an environment whose temperature approaches absolute zero, but which functions optimally at a higher temperature. Perhaps the spacecraft is on a mission away from the sun where the incident solar energy is insufficient to warm it, or has landed on the moon, where there is no solar heating for 14 earth days at a time. Some thermal energy is available from resistive heating of the electronic components, but this may be insufficient. If solar cells are the source of electrical power for the space vehicle, it is almost immoral to go to the expense and technical difficulty of making electrical power and then to degrade it to thermal power for the sole purpose of providing additional heating for the spacecraft. A small radioisotope heat source can provide the compensating thermal energy with 100% reliability, and suffers only from the fact that it cannot be turned on and off. In a manned spacecraft, a continuous and reliable source of heat may be required for long-duration missions, not only for temperature conditioning but for such purposes as water recycle and recovery. A joint AEC-Air Force development program for an isotopically heated water recovery system is well down the development path and looks very attractive in its economic and technical aspects (Figure 8).

Another purely thermal application is more down-to-earth. You may have seen some publicity about the joint program of the AEC and the Navy to develop a heater for the suits of deep-sea divers and aquanauts. The first prototype has been completed and is under test (Figure 9). The heating unit consists of 750 grams of plutonium-238, which produce 420 watts of thermal power to heat a fluid which circulates through the veins of the suit. A development problem still to be solved in this application is reduction of the ionizing radiation level from the heat source. Although plutonium-238 is primarily an alpha particle emitter, it also decays by spontaneous fission in a small fraction of the total decay events and gamma radiation is emitted as part of both the alpha and spontaneous fission decay processes. The background level of gamma rays and the energetic neutrons from fission cannot be ignored. Other contributors to the radiation level are neutrons resulting from alpha-n reactions on light element impurities in the chemical form of the isotope and gamma radiation from the decay of plutonium-236, a nuclear impurity.

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ELECTRICAL POWER SUPPLIES

The next category of use for radioisotope heat sources is the electrical power supply. I am quite sure that you are all familiar by this time with one or more of the so-called SNAP (Systems for Nuclear Auxiliary Power) devices which have already been used in every environment of the earth. Some of these power supplies use fission products as heat sources, primarily because they are less expensive than the heavy element alpha particle emitters. All devices in actual use employ the thermoelectric conversion principle. The SNAP device's versatility is well-demonstrated by its ability to function at the bottom of the ocean, in the frigid climates of the Arctic and Antarctic, and in outer space.

I will mention specifically here only three new radioisotope power supplies which are in the final stages of development and which have exciting futures ahead of them. SNAP 19, a 50-watt supply consisting of twin thermoelectric generators fueled with plutonium-238, will be launched aboard the NIMBUS B satellite later in 1968 on a mission of collecting data on the earth's weather conditions (Figure 10). SNAP 27 is also a thermoelectric power supply fueled with plutonium-238, but of much different design than heretofore used. It is an integral part of the scientific equipment to be left on the moon by the Apollo astronauts (Figure 11). One of the novel features associated with it is that the heat source will be transported to the moon in a container separate from the conversion apparatus and will be inserted in the converter after the landing on the moon. This approach was deemed necessary to provide additional assurance that the integrity of the heat source would be maintained if it became necessary to abort the mission during any part of the sequence from earth launch to lunar landing. J. A. Powers of the Atomic Energy Commission staff will present a paper later in this symposium which goes into detail on the nature of radioisotope heat sources for aerospace power supplies of the kind that I have described here.

In this age of miniaturization, it isn't so surprising that a tiny isotopic power supply is under development also. Its field of application is not so esoteric as that of space exploration, but it may be of far more immediate importance to us. I am thinking of the device now in the testing phase which is designed to power a cardiac pacemaker (Figure 12). As you know, several forms of heart disease

are characterized by failure of the heart to receive a large enough or regular enough electrical stimulus to cause normal pulsation. Stokes-Adams disease is a significant one of these heart malfunctions. Correction is achieved by implanting an artificial electrical stimulator or pacemaker in the thoracic or abdominal cavity and attaching electrodes to the heart surface. Conventional pacemakers use chemical batteries as their source of energy and the batteries limit the life of the devices to from two to three years. The isotopic power supply, which would replace the implanted chemical batteries, is designed to last at least ten years, and hopefully will therefore reduce the frequency of replacement of the power supply for the pacemaker. Of course, the radioisotope heat source would last much longer than ten years, but we're not so sure about the electronics or the patient! The nuclear pacemaker power supply produces about 160 microwatts of electrical power and also will use plutonium-238 as its energy source. Only 300 milligrams of isotope are needed. As many as 10,000 pacemakers per year may be required.

MECHANICAL POWER SUPPLIES

The next category of utility that comes to mind is based on the mechanical power supply. One example will suffice here, and it constitutes one of the most intriguing potential applications for nuclear energy that has been yet devised. Transplantation of human hearts from one individual to another has received world-wide attention recently, and the results obtained have been discussed in every home with access to a newspaper or radio. The public is less well acquainted with a still modest program being carried out by the National Heart Institute of the National Institutes of Health to develop mechanical devices to assist the pumping action of a failing human heart or - in the extreme - to completely replace the human heart.

The ultimate objective of this program is to produce an artificial heart which, after implantation within the patient's body, would restore the person to a condition where he would be a useful member of society again. This means, of course, that the artificial organ would have to have a compact and reliable power supply - no wires plugged into the light socket permitted!

Problems associated with the development of an artificial heart are so complex and severe that I cannot give full assurance that such a device will be available in

any predictable time period, and yet there is reason for optimism because the heart is relatively simple in its function as compared to other organs of the body. One of the very few known possibilities for furnishing power to a completely implantable artificial heart lies in the radioisotope heat source (Figure 13). Since the heart is a mechanical pump, the heat need be converted only to mechanical energy through one of the thermodynamic cycles I mentioned previously.

Preliminary physiological studies with animals indicate that the human body should be capable of dissipating the heat which is unavoidably not converted to pumping action. Only about 7 watts maximum of pumping power are required to sustain the life of a person in normal activity, and this drops to 2 to 3 watts under quiescent conditions, such as in sleeping. If an implanted thermodynamic engine and pump can be made to operate at 20% overall efficiency, then between, say, 15 and 35 watts of thermal power would be required. Should the radioisotope power supply prove to be the best, and if all the other developmental problems can be overcome, demands for radioisotopes to meet the need would amount to requirements for tons per year! At present, plutonium-238, properly purified to reduce its radiation level, appears to be the leading candidate for the power source for the artificial heart.

RADIATION APPLICATIONS

Turning now from the consideration of isotopes as heat sources, let us give our attention to those applications of the actinides which are based on their radiation properties. Each of the actinides has isotopes which are alpha particle emitters, which decay by spontaneous fission and hence are sources of neutrons, or which produce characteristic gamma rays.

Starting with the lower end of the atomic number range, americium-241 is finding a variety of uses. It is in plentiful supply and even larger quantities will be available in future years, as will be pointed out. Its long half-life of 433 years minimizes the necessity for frequent decay correction, but does make its specific activity too low to be considered as an alpha particle heat source at present.

One property of particular interest in the decay of americium-241 is the 60 KeV gamma ray emitted in about 35 per cent of the alpha particle decays. This predominant photon emission is excellent for use in various

types of measuring equipment such as thickness gauges and level gauges because its attenuation coefficient in materials of moderate to low density is neither too high nor too low.

Another use for the gamma ray from americium-241 is its ability to excite elements in the middle of the periodic table in order to detect their presence and determine their relative amounts by observing their characteristic x-rays produced by the process of fluorescence. An unusual application of this technique that is now being studied by Dr. Paul Hoffer at the Argonne Cancer Research Hospital involves moving a collimated 300 millicurie americium-241 gamma source and a solid state radiation detector around the area adjacent to a patient's thyroid gland (Figure 14). The stable iodine-127 present in the thyroid emits x-rays at an intensity proportional to the amount and distribution of iodine, and an indication is thereby obtained of the functional adequacy of the thyroid. This method could conceivably replace or supplement the now widely used radioiodine uptake and scanning technique because there is no residual radioactivity left in the patient, and the radiation dose to the patient is markedly reduced.

For periods of time up to a few years, the correction for decay of americium-241 is often less than the statistical counting error and can be ignored. Popular applications for americium-241 include its use in static eliminators, smoke detectors, radiography units, and oil well logging devices. In oil well logging, it functions as an americium-beryllium neutron source.

Several actinide elements have alpha particle emitting isotopes which are available in large enough quantity to be of commercial or scientific interest. Plutonium-239, plutonium-238, americium-241, curium-242, and - surprisingly - einsteinium-254 have been used as alpha particle sources for one or more of the following purposes:

1. To produce neutrons by the alpha-n reaction with elements of low atomic number such as beryllium;

2. To provide external beams of alpha particles as the excitation medium in new methods of chemical analysis, such as alpha-excited x-ray analysis and alpha scattering analysis; and

3. In methods of analysis of physical properties such as density or thickness based on absorption or backscattering of alpha particles.

Whereas 15 or 20 years ago I could note with wonder the emerging industrial uses of isotopes, I must now turn to more exotic applications to avoid reciting the commonplace. It is with some personal pride that I note that the first use of radioisotopes on an extraterrestrial body - the moon - involved the isotopes of two actinide elements, curium-242 and einsteinium-254. The 162-day curium-242 was the source of alpha particles in the equipment on board Surveyor V which performed the first direct chemical analysis of the lunar surface by alpha particle scattering and alpha-proton reaction techniques. The source strength was about 100 millicuries. The einsteinium-254 was placed near the alpha particle detectors to act as an energy marker at 6.44 MeV. The maximum energy of the curium-242 alpha particles is 6.11 MeV.

APPLICATIONS OF CALIFORNIUM-252

As far as future applications of the actinides as radiation sources are concerned, I look forward with the greatest interest to the fruition of developments now taking place with californium-252. Since this isotope will be available in at least gram quantities within a few years, it is not at all out of place to speculate on its large-scale uses, all of which at present are based on the fact that this isotope, with its nominal 2.6 year half-life, decays by spontaneous fission in about 3% of the decay events. Since spontaneous fission is accompanied by neutron emission this is equivalent to a neutron yield of about 4.4×10^9 neutrons per second per curie of californium-252 - or about 300 times as many neutrons per alpha particle disintegration as can be obtained with the best alpha-n neutron source, such as a radium-beryllium source. One curie of californium-252 weighs about 2 milligrams, so it is just about as intense a neutron source as a power reactor! The general advantage of californium-252 over machine or reactor sources of neutrons is its inherent compactness and portability. Among the applications for it being studied are:

Radiotherapy - Neutron irradiation of cancerous tissue is a promising approach in cancer therapy, and is important because neutrons overcome the radiation resistance of anoxic cells in tumors. Several applicators in the form of needles, each containing a few micrograms of californium, have been supplied to medical researchers already. The convenience and economy of the needles are most evident here, since patients would not have to be taken to reactor or accelerator sites for treatment. This is a good example of the mountain coming to Mohammed!

Radiography - Neutron radiography is a technique complementary to the more familiar gamma or x-radiography. It is expected to be of value in the medical or biological fields because it offers improved delineation of soft tissue morphology and elimination of excessive bone contrast. It may also be useful in non-destructive inspection methods where large thicknesses of heavy materials must be penetrated by radiation. High atomic number materials have relatively low mass absorption coefficients for neutrons. Californium-252 neutron sources for this purpose would be much more adaptable to field or laboratory use than reactors or accelerators. A problem still to be resolved, however, is the development of a good imaging technique for neutrons of fission energy. Most of the development work on imaging with neutrons has been done in the thermal region.

Activation Analysis - Since neutron activation analysis is now a widely accepted technique, the application of californium-252 as a portable neutron source is quite apparent. It would be particularly advantageous for mineral exploration and in remote regions, such as the ocean floor or the surfaces of other planetary bodies.

Radiation Effects - A number of experiments come to mind out of the past in which unshielded reactors have been operated above ground surface for the purpose of generating sufficient neutrons to determine their effect on the surrounding ecology or to measure the attenuation afforded by various types of shielding. This kind of research is important for the future, primarily for defense research purposes. It has been proposed that a californium-252 source be used instead of a reactor or accelerator neutron source. A 40-gram californium source would produce 10^{14} neutrons per second, and would require very little in the way of supporting facilities other than means to remove the 1.6 kilowatts of thermal power that would be generated.

Hydrology, Petroleum Exploration, and Other Industrial Applications - Almost all oil wells and an increasing number of water wells are logged for geophysical purposes by one or more techniques, including neutron scattering. Alpha-n neutron sources are in common use for this purpose, and in fact this is one of the largest uses for americium-241. Californium-252 would offer the advantages of greater neutron intensity and much more compact size relative to other isotopic neutron sources or to small accelerators which produce neutrons by the D-T reaction. Other possibilities

for its use include the routine measurement by neutron scattering of the water content of soils, wood, concrete, coal and other bulk materials, and on-line neutron activation analysis of flowing process streams.

REACTOR FUELS BEYOND PLUTONIUM-239

Although I am not discussing here the well-known applications of plutonium-239 as a nuclear fuel, worth mentioning briefly are some possible special applications of heavier actinide isotopes to this purpose.

The potential importance of the fission properties of higher mass number isotopes of plutonium and of the heavier actinide elements is not often considered. For example, the availability of increasing amounts of americium and curium isotopes may make feasible the construction of thermal neutron reactors fueled with such isotopes as the 152-year americium-242, with a thermal neutron fission cross section of about 6,000 barns, or the 9,300-year curium-245, with a cross section of about 2,000 barns; and, perhaps further in the future, we may consider the use of such isotopes as the 360-year californium-249 and the 800-year californium-251, which also have been reported to have very high thermal neutron fission cross sections.

With such isotopes as these, it may become feasible to construct a very small reactor with a minimum amount of fuel which someday might find importance.

Perhaps closer at hand is the possibility of using plutonium-239 and heavier plutonium isotopes in an application which has been studied in the past at such installations as Pacific Northwest Laboratory - the "Phoenix fuel." Here advantage is taken of appropriate combinations of these isotopes, so that one of the isotopes serves as a non-fissionable burnable poison whose neutron capture product is fissionable. Mixtures of plutonium-239, nonfissionable plutonium-240, and fissionable plutonium-241, the Phoenix fuel, make possible the design of a reactor system which, from the nuclear standpoint, is able to sustain almost constant reactivity over very long periods of time. It is conceivable that in future reactor applications this may prove to have some very important advantages.

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ACTINIDES FOR THE CHEMIST

The actinide elements constitute a fascinating transition series of great intrinsic scientific interest to the chemist. In addition, as these elements become a more familiar and important part of our technical and economic life, it is natural to expect that chemists will want to study their chemical and metallurgical properties in greater depth than is now being done. Such exploration of the actinides will be more convenient and meaningful as some of their especially long-lived isotopes become available, as the result of intensive neutron irradiations followed by electromagnetic separation, in macroscopic quantities which are nearly isotopically pure. Problems arising from radiolysis of solutions, degradation of crystal structures, the handling of highly radioactive materials, and transmutation by decay will then be minimized.

The prospects for this approach appear to be very good. Neptunium-237 already qualifies in this regard. Plutonium-244, with its 76 million year half-life, will be a relative pleasure to work with in the laboratory; its preparation in macroscopic quantity will require electromagnetic separation from other plutonium isotopes. Americium-243 has a half-life almost 20 times as long as americium-241, is more convenient for chemical research and will be available in large quantities. By comparison with curium-242 and curium-244, the isotopes curium-245 through curium-248, singly or in mixtures, would almost be considered to be stable by the radiochemist! Curium-247, with a half-life of 16 million years, is the best single isotope of curium. The 470,000-year curium-248 is also good and is easier to prepare in high isotopic purity; a special way of preparing it in high isotopic purity is as the alpha particle decay product of californium-252. These heavier curium isotopes will also require magnetic separation for their preparation in high isotopic purity.

Berkelium-247 has a half-life of about 1,400 years, but no easy way to produce it in macroscopic amounts has appeared yet.

The 360-year californium-249 can, fortunately, be prepared in isotopically pure form by chemically isolating its beta particle-emitting parent, berkelium-249, and letting it decay to the daughter californium-249. The even longer-lived californium-251 (half-life about 800

years) would require very complete electromagnetic separation from "hot" californium-252 in order for it to be useful for chemical studies. Beyond this point the state of affairs promises to remain "hot" or scarce.

LARGE-SCALE PRODUCTION OF ACTINIDES

What are the present and anticipated capabilities for producing the actinide elements in large quantity in the United States? To answer this question, I will essentially exclude consideration of accelerators and charged particle reactions for this purpose because the quantities produced are usually too small and too expensive for practical applications. I therefore also exclude production of elements above fermium, since they have not yet been observed as the product of neutron-induced nuclear reactions.

Neptunium-237 is an important isotope because it is the precursor of plutonium-238. Fortunately, it is produced incidentally in nuclear reactors by the (n, 2n) reaction on uranium-238 and by successive (n,γ) reactions on uranium-235 and uranium-236 to form uranium-237, which undergoes beta decay to neptunium-237. This long-lived alpha particle emitter (half-life of two million years) is now being recovered from irradiated AEC production reactor fuels. At least two private companies are considering recovering it from spent power reactor fuels, and if our predictions of the rate of growth of the nuclear power industry materialize, it can be expected that 200 kilograms per year from this source could be available by 1975 and about 500 kilograms per year by 1980.

Plutonium-238 is produced by the thermal neutron irradiation of neptunium-237 in AEC production reactors (Figure 15). It has been limited in its rate of production by the amount of neptunium-237 available. However, essentially all requirements for it are being met. The AEC has had a commitment to produce 900 kilograms of plutonium-238 for use by NASA in the 1964-1980 time period, contingent upon materialization of NASA long-range plans for a variety of space missions. Sufficient production reactor space and neptunium target material should be available for this purpose. In fact, this isotope could be available in ton quantities to meet other large requirements that might develop, including the artificial heart and pacemaker programs. As I indicated previously,

it would be desirable to produce a higher quality isotope; that is, with reduced plutonium-236 content, for biological applications. This could be accomplished by storing the plutonium-238 while the relatively short-lived (2.85 years) plutonium-236 decayed preferentially. A ten-fold reduction in plutonium-236 content occurs in about ten years. Such a procedure would be relatively expensive, and would require another chemical separation of the decay products at the end of the storage period. A more attractive approach would be to minimize the formation of plutonium-236 (which is produced by the (n, 2n) reaction on neptunium-237) by increasing the proportion of thermal neutrons in the reactor used to convert neptunium-237 to plutonium-238.

Americium-241 is valuable for its own uses and as a starting material for isotopes of higher mass number. It also is incidentally produced as the beta particle decay product of the plutonium-241 which is present in all plutonium formed in production and power reactors. The AEC has been the only domestic source of this isotope in years past. It will be available in much larger quantities from increased power reactor operation in the future, particularly because power reactors subject the plutonium formed to higher neutron exposures than in production reactors, thereby increasing the relative concentration of plutonium-241. However, this isotope would be mixed with americium-243 at the time of discharge of the spent fuel. To recover pure americium-241, it would be necessary to separate the mixture of plutonium isotopes in the conventional manner, and allow the americium-241 to grow in as the result of decay of plutonium-241. If plutonium is being well utilized in plutonium recycle reactors or fast breeder reactors, it might not be economically attractive to store the plutonium, allow the decay to occur, and then process the plutonium again to recover the americium.

It is with some satisfaction that I note the current and future plans for the recovery of large quantities of neptunium-237 and americium-241 as byproducts of nuclear reactor operation, a course of action that I have advocated with various degrees of insistence for more than 20 years.

Production of curium isotopes begins to cause special problems. Curium-242, produced by thermal neutron irradiation of americium-241, has a relatively short half-life and is formed in high yield only if the neutron flux is high and the irradiation time short on a comparative basis. Although curium-242 has been considered as a heat source candidate in applications where the mission life is relatively short - perhaps up to six months - and where use could be made of

its intense thermal emission rate - 120 watts per gram - no such application has reached fruition. However, hundreds of grams per year of curium-242 could be produced on relatively short notice if the need arose.

Curium-244, the alpha particle emitting isotope with an 18-year half-life, is proposed as an alternative to plutonium-238 in heat sources. It has the advantage of exhibiting a higher thermal emission rate than the latter isotope, and the potential of earlier production in larger quantities, but suffers the disadvantage of having a higher radiation level due to neutrons from spontaneous fission.

Six kilograms of curium-244 are now in the final stages of production at Savannah River; the process involves irradiating plutonium-239 for several years, since five successive neutron captures are involved. To even accomplish the irradiation within this time scale it has been necessary to increase the thermal neutron flux of a reactor substantially above levels found in other reactors. The highest continuous peak neutron flux ever reported, 6.1×10^{15} neutron/cm^2-sec., has been achieved as part of the heavy element program at Savannah River. Studies indicate that this can be increased still further to 2.5×10^{16}, which would correspond to a time average flux at reactor center of 0.7×10^{16}. In the future, recovery of americium-curium fractions from spent power reactor fuels could provide excellent starting material for production of curium-244 in subsequent high-flux irradiations. Costs would be reduced substantially by this approach, which would make feasible the production of tons of this isotope.

THE HIGH FLUX ISOTOPE REACTOR

As we go to higher mass number curium isotopes or beyond the curium isotopes, conventional power or production reactors no longer serve to provide the intensive neutron fluxes required to increase the mass number and atomic number. At least three different means appear to be available for this purpose.

First, we have the high performance research reactor epitomized by the High Flux Isotope Reactor, or HFIR, at Oak Ridge National Laboratory. In operation since 1966, this 100 Mw reactor can produce a flux as high as 5×10^{15}, which is perturbed to about half this value when a plutonium-242 target of 250 grams is inserted in the flux-trap region of the core. This reactor has now produced about seven

milligrams of californium-252, which have been isolated in the transuranium processing facility (TRU) adjacent to the reactor. The plutonium-242 target material comes from Savannah River, where it is prepared by high-flux neutron irradiation of plutonium-239. By the mid-1970's, the HFIR will be producing californium-252 at the rate of 1 gram per year, along with 0.1 gram of berkelium-249 (and therefore potentially of its useful daughter, californium-249), 10 milligrams of einsteinium-253, 0.03 milligram of einsteinium-254, and lesser amounts of fermium isotopes.

The Savannah River production reactors, operating in the thermal high-flux mode, also produce these higher isotopes, of course, as indicated in Figure 16. Twenty-five milligrams of californium-252 have been produced there as part of the curium-244 campaign described previously. If a Savannah River reactor were to operate at a high thermal flux for two years, an additional 3 to 4 grams of californium-252 could be available by about 1973.

Additional high-flux irradiation capability will come on the line with the completion of the Argonne Advanced Research Reactor (A^2R^2). This reactor, using a core identical to that of HFIR, will operate at about the same flux. Yields of actinide isotopes that can be anticipated from reactors like HFIR and A^2R^2 are shown in Figures 17a, b, c.

THE RESONANCE REACTOR CONCEPT

Another possibility for increasing the production rate of heavy elements involves changing the mode of operation of a Savannah River reactor to produce a high neutron flux in the resonance energy region, rather than in the thermal region. Computations indicate that fluxes above thermal energy in the range of 10^{14} to 10^{15} might be achieved. Sufficient target material to make effective use of such a large reactor would not be available for some years, when large quantities of americium-243 and curium-244 will be separated from spent power reactor fuels. One estimate indicates that hundreds of grams per year of californium-252 could be produced by a reactor operating in the resonance mode, although neutron absorption resonance integrals are not sufficiently well known at this time to make this an accurate prediction.

UNDERGROUND NUCLEAR EXPLOSIONS FOR PRODUCTION OF ISOTOPES

You will hear during this symposium a report by J. E. Evans and co-workers of the Lawrence Radiation Laboratory, Livermore, about techniques for recovering heavy elements