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SUMMARY OF PLUTONIUM FUEL TECHNOLOGY SYMPOSIUM HELD AT RICHLAND, WASHINGTON APRIL 22-23, 1958

COMPILED BY

F. W. ALBAUGH

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HANFORD LABORATORIES

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Issued by

F. W. Albaugh
HANFORD LABORATORIES OPERATION

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RICHLAND, WASHINGTON

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V. PLUTONIUM HANDLING PROBLEMS SESSION

FOREWORD

In February of 1958 at a meeting of representatives of the Atomic Energy Commission and several Commission Laboratories it was suggested that a classified symposium be held on the general subject of utilization of plutonium as a power reactor fuel. The Hanford Atomic Products Operation, General Electric Company, was assigned responsibility for organizing the meeting, which was held at Richland, Washington, on April 22-23, 1958.

The purposes of the meeting were (1) to exchange information and review work in progress, (2) to identify deficient areas of research and development in the field of plutonium fuels technology, and (3) to assist the Atomic Energy Commission in determining the feasibility of an unclassified CAP meeting on the state of plutonium fuels technology. In regard to the first purpose, the symposium was apparently successful, bringing together work of different sites as well as work in the various disciplines pertinent to use of plutonium fuel cycles. In regard to the second, no major omissions in the Commission program were obvious; programs specifically directed toward plutonium fuel utilization in fast heterogeneous reactors, fast liquid metal-fueled reactors and thermal heterogeneous reactors are in progress at ANL, LASL, and HAPO, respectively. In addition, much separations processing and weapons development work at these and other sites is pertinent to the problem of plutonium fuel utilization. Finally, it appeared that an early unclassified CAP meeting would be somewhat premature; much of the pertinent information available was classified because of weapons association, and the unclassified programs in the field were too new to have developed a significant fund of conclusive data.

Inasmuch as most of the summaries given in the body of this report were submitted by the authors in the Spring of 1958, data and conclusions given may not in all cases correspond with the current state of knowledge.

F. W. Albaugh

Manager, Reactor and Fuels
Research and Development

FW Albaugh:kb

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SUMMARY OF PLUTONIUM FUEL TECHNOLOGY SYMPOSIUM

HELD AT RICHLAND, WASHINGTON,

APRIL 22-23, 1958

Welcome — H. M. Parker, Manager, Hanford Laboratories

Keynote Remarks — R. C. Dalzell, Atomic Energy Commission

I. PROGRAM REVIEW SESSION

Chairman: F. W. Albaugh, GE-HAPO

1. EBR-II Program - H. O. Monson, ANL

The Argonne Experimental Breeder Reactor II (EBR-II) is under development as one part of the U. S. Atomic Energy Commission's Experimental Power Reactor Program. The EBR-II is an integral nuclear power plant; it includes a complete fuel processing and fabrication facility in addition to the reactor, heat transfer systems, and steam-electric plant. The thermal power rating of the reactor is 62,500 KW. Gross electrical power output is 20,000 KW. Engineering design and component development are presently nearing completion. Construction is scheduled to start in early 1958. Operation is expected to begin in 1960.

Exclusive of fuel recycle aspects, the over-all design objectives of the facility are: to attain high thermal performance (high core power density and high coolant temperature level); to achieve efficient breeding (large breeding ratio); and, to utilize insofar as possible, prototype components (components of such size and design as to permit their use in central station plants with little or no modification).

The facility is comprised of three major systems: (1) the Primary System, consisting of the reactor and the primary sodium cooling system; (2) the Secondary System, or the intermediate sodium heat transfer systems; and (3) the Steam-Electric System.

The complete primary system is contained in a single vessel, or "Primary Tank", 26 feet in diameter and 26 feet in depth. All of the primary system components, including the reactor, sodium pumps, primary piping, heat exchanger, and fuel transfer and storage system, are submerged in the large bulk volume (80,000 gallons) of sodium within this tank. This arrangement provides a number of advantages, among which are: loss of coolant becomes virtually impossible; rapid changes in load demand are prevented from being reflected as temperature changes in the coolant entering the reactor; fuel elements may be removed from the reactor and stored until ready for processing while remaining submerged in the bulk

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sodium, thus obviating the need for an auxiliary cooling system; and, the need for high integrity of the primary sodium piping is eliminated since all leakage is back to the bulk sodium. The two main pumps provided operate in parallel and supply 8200 gpm primary sodium flow. Both of these pumps are of the motor-driven, centrifugal type. The flow path is from the bulk sodium to the pumps, the reactor, the heat exchanger, and back to the bulk sodium, in that order. The temperature of the sodium at inlet to the reactor is 700 F, and at outlet from the reactor, 900 F.

The reactor is of the heterogeneous type, unmoderated and sodium cooled. Control is effected by movement of fuel into and out of the reactor core. A total of twelve peripherally located control rods are employed, of which one is used for regulation. The core is of the shape of an approximate right circular cylinder, with L/D of about 0.8. Maximum core power density is 1400 KW/liter of core volume, or 4300 KW/liter of fuel alloy volume. Breeding blankets of depleted uranium surround the core on top, bottom, and sides. The major fraction of breeding takes place in these blankets; the remainder occurs in the core. For initial reactor loadings, the fissionable material to be employed is U-235, and for subsequent loadings, Pu-239. The approximate conversion (breeding) ratios expected with these loadings are 1.2 and 1.6, respectively.

The secondary system transfers the heat from the heat exchanger in the primary tank to the steam generator. This system is non-radioactive and serves to isolate the steam generator from the radioactive sodium of the primary system. A single A.C. electromagnetic pump provides a sodium flow rate of 6050 gpm. Sodium temperature at inlet to the heat exchanger is 610 F, and at outlet from the exchanger, 880 F.

The steam-electric system receives the heat from the secondary system and converts it to electric power. This system is of essentially conventional design, employing a standard, extracting, condensing, single flow type turbine. An automatic, full capacity, steam bypass system for dumping excess steam directly to the condenser is incorporated to prevent major load changes from effecting changes in primary or secondary system conditions. The condenser circulating water is cooled in a forced convection cooling tower. Turbine throttle flow is 192,200 lb/hr; steam rate is 8.04 lb/KWH. Turbine throttle steam temperature and pressure are 850 F and 1250 PSIG, respectively.

2. LAMPRE Program - D. B. Hall and W. J. Maraman, LASL

The fast reactor program at the Los Alamos Scientific Laboratory is primarily engaged in the investigation of a plutonium-fueled reactor concept known as the Los Alamos Molten Plutonium Reactor Experiment, abbreviated LAMPRE. This concept has evolved from considerations of the well-publicized role which uranium-238 must play in the long range plans for nuclear power and from the several potential advantages which plutonium offers as a reactor fuel. Further stimuli were the considerable experience and facilities at the laboratory in plutonium handling and technology, as well as the non-programmatic research into possible plutonium fuels which has been carried on for many years.

The power reactor concept which seems most attractive calls for a nearly stagnant region of molten plutonium as the fuel, cooled by a liquid metal such as sodium and surrounded by a fertile blanket of uranium. The use of molten fuel avoids many of the problems associated with metallic fuel elements, such as fabrication, irradiation damage and distortion. In principle, the fuel is capable of sustaining unlimited burn-up. However, this concept introduces as many new problems as it solves, and there is no reason for saying that the problems associated with molten plutonium reactors are any easier to solve than those of the more conventional fuel systems. For example, in this reactor design, complete integrity of the container must be assured and simple arguments will also show that the container must last many times longer than is expected for a fuel element cladding.

Plutonium-fueled reactors should operate on a fast neutron cycle to exploit the large breeding gains which are possible. Since fast reactors have relatively large critical masses, the fuel inventory should be kept as low as possible in order to improve the economic condition of the reactors. Fuel processing has unavoidable losses which can be reduced by decreasing the frequency of fuel treatment and this, in turn, is possible with the fast neutron spectrum compared to thermal reactors because the effective poisoning by fission products is very much less.

With these factors in mind the Los Alamos Scientific Laboratory has undertaken a development program which is divided into two phases. The first phase is represented by the reactor experiment LAMPRE I, which is intended to demonstrate the important problems of fuel containment and handling, and is, in reality, little more than a hot critical experiment generating not more than 1 megawatt of heat. The heat will be removed by sodium and dissipated by an air-cooled radiator. The second phase will be a completed reactor package including steam generation and integrated fuel reprocessing. This reactor, designated as LAMPRE II, will probably be scaled at 15 megawatts as a compromise between engineering simplicity and the demonstration of a practical reactor.

LAMPRE I should be regarded as an "in-pile loop". It will, however, provide necessary information on the behavior of this reactor concept under operating conditions. The basic design features are as follows: The fuel selected is 25 kilograms of the 9.5 atomic percent iron-plutonium eutectic. This is to be contained in a tantalum vessel, either a calandria pierced by closely packed tantalum tubes or between two closely spaced elongated sheets of tantalum welded at the perimeter and spiraled about one end. Molten sodium circulated by electromagnetic pumps is the coolant, and the reflector is iron. Control is to be accomplished with annular shims of aluminum or steel. The reactor will be shielded with borated graphite, lead and concrete.

For the first test of the reactor concept, LAMPRE I, it is undesirable to complicate the experiment with a breeding blanket. Indeed, at a power of 1 megawatt, it is difficult to obtain a satisfactory measurement of the breeding rate. Therefore, the reflector chosen for the LAMPRE I is iron, primarily because it is a satisfactory material for use with sodium. The

reflector is to be thin enough so that effective reactivity control can be obtained by reflector movement, but thick enough to reduce the mass of the fuel required for operation.

It is proposed to control the reactor experiment by varying the neutron leakage. There are several ways in which this can be done; the most attractive appears to be the use of a metal annular shim to displace part of the shield. Because of the iron reflector, the reflected fast neutrons are more effective than the moderated neutrons returned from the shield. The choice of shim material is not critical, and steel or aluminum were picked because of their good heat conduction properties and ease of handling. Plans are now being made to incorporate a variable control suitable for making oscillator tests of the behavior of the reactor to transients.

A convenient arrangement for shielding of the reactor experiment is provided by existing concrete and lead. Borated graphite is to be used also.

A mock-up of the reactor core has been constructed using reduced-density plutonium plates clad in nickel to represent the iron eutectic. In addition to tantalum, the mock-up contained aluminum at reduced density in place of the sodium. This assembly (LCX-1) contained about 75 percent by volume of fuel instead of the proposed 50 percent composition, but nevertheless, it has proved extremely useful as a check-point for reactor calculations and the difficult problem of determining control effectiveness. A second critical mock-up(LCS-2) is being prepared to simulate the proposed reactor configurations more closely.

The fuel chosen for the initial experiment is the plutonium-iron eutectic which melts at 410 degrees Centigrade. The phase diagrams of the binary mixtures of plutonium with iron, cobalt and nickel have been investigated in considerable detail. The cobalt eutectic has undesirable phase changes below its melting point of 405 degrees Centigrade, and the nickel eutectic has a melting point approximately 50 degrees higher than the iron eutectic. Although the high plutonium concentration of these fuels makes it difficult to extract heat, they are the best available at present. The fundamental properties of plutonium alloys are being investigated with particular emphasis on those alloys with melting points below 450 degrees Centigrade and with plutonium concentrations in the range of 20 to 90 atomic percent. In fuels for fast reactor applications there are few restrictions which need be placed on the choice of diluent, and therefore, the metallurgical properties appear more important than do the nuclear properties. Hence, a search is being made for diluents which are passive in nuclear and corrosive terms but give promise of improving the otherwise unacceptable characteristics of pure plutonium. The desirable physical properties include high conductivity, thermal stability and freedom from rupturing the container upon freezing and melting.

The search for a container material to hold the plutonium alloy is being directed toward the more refractory elements, niobium, molybdenum, tantalum and tungsten. Of these, tantalum development has been emphasized because of favorable resistance to attack, availability and ease of fabrication. An investigation of tantalum fabrication and joining methods is in progress.

Laboratory experiments are being carried out to determine the compatibility of the several plutonium alloys and tantalum test pieces prepared by various methods from different sources. Both static and dynamic tests are being made at above the temperature of the operating reactor. Some are integral in nature, containing all of the elements of the reactor with the exception of the radiation field. Tests are also under way to determine the additional effect of burn-up on fuel properties; however, since these tests are static, they may not give a complete picture. The problems of designing and assembling a conventional in-pile loop to provide the desired information are just as great as those associated with a small reactor experiment. A laboratory facility is now being prepared to handle and to make metallographic examinations of the highly alpha-active plutonium containing fission products.

Associated with the fuel development is a vigorous attack on the important problems of fuel handling and fuel reprocessing. Since a fast reactor is not sensitive to an accumulation of fission product poisons, it will not be necessary to decontaminate the fuel to very low levels. The objective is to find a reprocessing method which will permit the fuel to be treated at high temperatures, either continuously or batch-wise, without requiring a reactor shut-down.

A survey of methods for fuel reprocessing has been made in the laboratory. Pyrometallurgical experiments have been carried out on about a 100-gram scale using the plutonium-iron eutectic spiked with inert typical fission product elements. The processing conditions are being optimized on a 1-kilogram scale using the same material. Aqueous reprocessing methods have also been investigated and a flow sheet has been developed for the recovery and purification of plutonium from the pyrometallurgical residues.

A hot cell facility which is currently under construction will be used to verify the results obtained from the spiked plutonium on the fuel discharged from LAMPRE I.

For fast reactor applications, liquid metals are the most promising coolants in that they do not present a moderation problem. Of the liquid metals, sodium was first selected because of the favorable attention which it had received at the reactor laboratories. Dynamic loop tests which have been run subsequently with highly purified sodium at Los Alamos have shown satisfactory low rates of attack on tantalum samples. These tests are continuing, and a further test loop is being assembled which contains all of the features of the reactor cooling system except for the actual core structure. It is planned to operate this loop until it has proven satisfactory, and after inspection, its components are to be used in the reactor installation.

Preliminary design and testing of the components for LAMPRE II is proceeding concurrently with those for LAMPRE I. However, final design must await successful completion of LAMPRE I.

Incorporated in this development program is the realization that uncontestable proof of the safety of this reactor concept must be supplied. Calculations are being made to determine the behavior of the LAMPRE concept following reactivity insertions of different rates. The results of these calculations so far have supported the intuitive feeling that this reactor will behave much as a homogeneous reactor with a large negative coefficient of reactivity and be very stable in operation. However, since the precise physical arrangement used in a reactor can never be exactly calculated, it may be necessary to verify the mathematical analysis by observing the behavior of a reactor under the effect of all reasonable malfunctions. The design of a test site in Nevada for such experimentation is being prepared.

The successful demonstration of the operation and safety of LAMPRE II is about as far as the Los Alamos Scientific Laboratory is prepared to go with this reactor concept. Indeed, even with overwhelming successes, this program stretches out into the years past 1960.

3. Operation of the MTR on Plutonium Fuel - D. R. deBoisblanc, Phillips Petroleum Company

The operation of the MTR with plutonium fuel presents a number of problems and hazards not encountered in normal operation. Thus, a number of items must be analyzed to evaluate the safety and feasibility of using this special fuel in the MTR.

A serious control problem arises from the smaller delayed neutron fraction of Pu-239, which is about 0.21% as compared to 0.64% for U-235. This smaller delayed fraction requires modifications in the control system before the MTR can be safely fueled with plutonium. The maximum rate of shim rod withdrawal must be reduced to one-half of that used with U-235. The major effect of this change is the creation of an operating problem by reducing the portion of the cycle in which it is possible to override the xenon in recovering from a scram. The worth of the regulating rod must be reduced so that an excursion of this rod cannot make the reactor prompt critical.

While using plutonium fuel it is necessary to place additional restrictions on the experiments allowed in the reactor. Because of the small value of the delayed neutron fraction, no experiment can be tolerated which is capable of introducing a prompt change in $\Delta k/k$ of greater than 0.1 percent.

The α -emission from plutonium introduces a hazard not normally present in the same magnitude in the MTR. This hazard requires special preparations to be made before using plutonium in order to cope with a possible fission break. A resin bed is made available to process the water in case of α -contamination and a retention basin is kept empty so that the water can be stored until its disposal can be arranged. In addition, special containers are kept on hand to store elements with extensive fission breaks.

The presence of the α -emitting plutonium does not appreciably increase the consequences of a major incident over that from the fission products present near the end of a cycle using U-235.

The fuel requirement of the MTR with plutonium fuel differs from that with U-235 for the following reasons:

(1) The larger number of neutrons per fission in Pu-239;

(2) The larger capture and fission cross sections and the larger capture to fission ratio in Pu-239;

(3) The strong resonance in Pu-239 at 0.3 ev which has no comparable counterpart in U-235;

(4) The strong thermal absorption of Pu-240, which is produced by captures in Pu-239;

(5) The strong resonance in Pu-240 near 1 ev.

The required fuel content has been calculated by a perturbation technique which relates the plutonium concentration to a known U-235 loading. For fuel of 95.5% Pu-239 and 4.5% Pu-241 the calculated loading for approximately 400 megawatt days of operation consists of 23 fuel elements each containing 147 grams of total plutonium and four shim rods each containing 92 grams of total plutonium.

At startup the shim rods are more deeply inserted with plutonium fuel than with U-235. This arises because more plutonium must be consumed to produce a megawatt day and because of the buildup of Pu-240. This deeper insertion of the shim rods produces a flux distortion which in turn causes a heat transfer problem that places a limitation on the power of the reactor in the early portion of the cycle. The higher fast to thermal flux ratio of the plutonium core produces a flux pattern which results in a nine percent increase in the power density at the core reflector interface as compared to a U-235 core operating at the same power.

The calculated void and temperature coefficients for the plutonium core are both negative and have values roughly the same as for the respective coefficients in a U-235 core.

By restricting the experimental loading, making the required modifications in the control system, and proceeding as in the startup of a new reactor, the MTR can be operated on a plutonium core without increasing the probability or consequences of a major incident.

4. The Plutonium Recycle Program - R. M. Fryar, GE-HAPO

An important segment of the over-all development of plutonium utilization was undertaken about mid-1956 by the Hanford Laboratories at the request of the Atomic Energy Commission. The charter of the "Plutonium Recycle

Program" basically is to develop technology for the utilization of plutonium fuels in thermal heterogeneous power reactors. Today I should like to discuss in a broad sense the purpose and organization of the program. In addition, as an aid in orientating some of the later program papers, I should like to describe one of the principal experimental tools of the program, the Plutonium Recycle Test Reactor.

The Plutonium Recycle Program is organized functionally into nine sub-programs dealing with the principal phases of the research and development effort. It is worthwhile to describe very briefly each of these programs. The particular arrangement and order of discussion that I have chosen for the various sub-programs has no particular significance.

Plutonium Fuels Development. Plutonium fuels development is perhaps the most vital area in the development of plutonium recycle technology. It is also a field in which little is known. It is necessary to develop plutonium fuel elements and processes for their fabrication which meet the criteria of (a) in-reactor stability over long periods of exposure, (b) inexpensive fabrication cost, and (c) freedom from personnel hazards in fabrication operations in spite of the toxic nature of plutonium. The accomplishment of these objectives will require conception of novel fuel designs of both metallic and ceramic types and an extensive program of fabrication development, and of ex-reactor and in-reactor testing. Design concepts to be stressed will be those susceptible to a high degree of remotized fabrication, in order to achieve low cost and radiological safety. Several papers will be presented during this symposium covering the problems and accomplishments of this program.

Uranium Fuels Development. The technical scope of this program will include the design, fabrication, and evaluation of ceramic uranium fuel elements that can operate at high specific powers, coolant temperatures, and exposures. The work will include investigation of promising core and cladding materials, development of economical fabrication and assembly processes, and fabrication and irradiation testing of finished elements. In addition, supporting basic studies of the properties of ceramic fuels will be conducted.

The uranium fuels development sub-program is an essential part of PRP for two reasons. First, a principal way in which plutonium fuel can be utilized in a power reactor is in a two-region, "spike" loading, comprising an all-uranium fueled region and an all-plutonium region; the uranium fuels development sub-program will provide the technology required for loading of the all-uranium region. A second way of enriching reactor fuel with plutonium is via uniform enrichment, in which plutonium is uniformly mixed with uranium in all fuel elements. In the development of fuel elements of the uniform enrichment type it is logical and efficient to conduct preliminary development studies with uranium only in order to minimize personnel hazards. I might add at this point that the anticipated conduct for the program will be to first concentrate on the spike approach and follow this with work on the uniform enrichment.

Materials Development. The materials sub-program encompasses the development of zirconium alloy and aluminum alloy fuel cladding materials in support of both the plutonium fuels and uranium fuels programs. It also provides support of PRTR design, through development of zirconium alloy process tubing and nonmetallic materials of reactor construction. Extensive off-site vendor liaison, corrosion studies, mechanical and physical properties studies and radiometallurgical examinations are required in discharging these responsibilities.

Chemical Research and Development. The purpose of Chemical Research and Development sub-program will be to define fuel separations processes for Pu-U mixed or separate fuel cycles, which when considered with reactor operating conditions, fuel element design and performance, and fuel preparation, will yield optimum operability and minimum costs for the fuel cycle.

In addition, program effort will be expended to provide chemical methods, equipment, and materials for the separation, decontamination, and conversion of plutonium or other substances from irradiated fuels of importance to physics, metallurgy, engineering or other programs requiring such services. Further discussion of this program will be presented during the symposium.

Physics Research and Development. The ability to predict accurately the reactivity characteristics and allowable fuel exposure of plutonium-fueled systems as a function of lattice parameters such as plutonium isotope distribution, flux spectrum, etc., is obviously a key aspect of plutonium recycle technology. Attainment of the desired understanding will require theoretical studies and experimental measurements in the PRTR and in other special lattice test facilities such as the Hanford Physical Constants Test Reactor. These measurements will differ from conventional exponential pile or PCTR measurements in that they will be aimed at systems utilizing plutonium as a major portion of the fissionable material and will involve both "green" and irradiated fuel loadings. Further discussion of plutonium recycle physics will be presented to this group later this morning.

Design Development. The purpose of the design development sub-program is two-fold. First, in the early years of the Plutonium Recycle Program, it is providing the design criteria and scope needed as a basis for detailed design and construction of the test reactor. Second, as knowledge of the utilization of plutonium fuel cycles accumulates, this knowledge will provide a basis for reactor design-development studies whose objective is to define in a preliminary way the nature of modifications of the various basic reactor types required to optimize them for the utilization of plutonium fuel. The latter activity represents one of the principal end objectives of the Plutonium Recycle Program as a whole.

Reactor Engineering Development. Inasmuch as the Plutonium Recycle Program is primarily a fuel cycle program rather than a reactor development program, the presently described sub-program is regarded as auxiliary and supporting in nature. It is intended to comprise three principal functions: (1) experimental testing of critical equipment and process features of the PRTR design in order to assure proper performance of the test reactor; (2) continuing provision of specialized functional assistance to other sub-programs

in the areas of heat transfer, fluid flow, out-of-pile mockup testing of components and systems, and shielding technology; and (3) after startup of the reactor, provision of expert engineering assistance as required to assure safe and efficient operation and to make such changes in process or in components as may be desired during the course of the over-all program.

Test Reactor Operation. The purpose of this sub-program is to provide pre-startup planning of the operation of a test reactor required in the Plutonium Recycle Program and, after startup, provide safe efficient operation of the reactor.

A test reactor has been regarded as a necessary experimental tool in the Plutonium Recycle Program to obtain required physics data on the long exposure behavior of plutonium, to provide appropriate quantities of irradiated plutonium-bearing fuel elements for use in chemical reprocessing and fuels development sub-programs, and to provide a means for determining the in-reactor stability of candidate plutonium-bearing fuel elements, a field in which little is known at present.

The test reactor sub-program is concerned with the safety and efficiency of test reactor operations but not with the nature of the tests to be performed or with the scope of the primary research and development for which the reactor is utilized.

Cycle Analysis. The purpose of this sub-program is to provide the over-all studies that are required to properly integrate the Plutonium Recycle Program and plutonium recycle systems as a whole. The need for this program arises from the complex technical and economic interactions that arise in attempting to plan uranium feed supply, reactor design and operation, and fuel reprocessing to achieve optimum design and operation of the over-all system. Specifically, the sub-program includes development of generalized expressions of the effect of reactor parameters on the efficiency of plutonium fuel utilization, economic analyses of various processing methods, planning, in consultation with other groups, of the sequence of PRTR experiments, and analysis and interpretation of the significance to plutonium recycle systems in general of experimental and theoretical data obtained on the Plutonium Recycle Program.

The nine sub-programs I have just described constitute the Hanford Laboratories Program. Several of the programs of direct interest to the members of this symposium will be reported to this group by others. It is difficult to discuss any research and development program without mentioning the facilities required to secure necessary program data. So, at this time I should like to say a few words about one of the major facilities of the program, the Plutonium Recycle Test Reactor. This facility, which is an essential tool of the program, will not be the subject of any of the papers to be presented here, and because of its vital nature to the program, a brief description is quite apropos.

Plutonium Recycle Test Reactor. Very early in the program our studies indicated that while the Hanford Laboratories had the majority of facilities required for conduct of the Plutonium Recycle Program, there was not available at Hanford or elsewhere in the United States an irradiation facility believed necessary if the program goals were to be achieved in a reasonably short time. Planning studies were carried out, and it was concluded that a reactor facility was indeed necessary for the accomplishment of the cycle physics, chemical separations, and plutonium fuel technology, and other programs previously mentioned. The reactor was to provide:

1. for irradiation testing of plutonium-fuel and feed-fuel elements,

2. for direct investigation of reactivity and exposure effects from isotope buildup on the uranium-plutonium fuel cycle,

3. for production of pilot plant quantities of prototypical irradiated fuels for fuels reprocessing and fuel refabrication studies,

4. for investigation of control characteristics, reactor dynamics, and reactor operating problems for plutonium recycle operation, and

5. for providing a convincing demonstration on the pilot plant scale of the economics and practicality of various fuel cycles.

The reactor design that evolved from the series of compromises that were necessary because of the sometimes conflicting demands of the various requirements is characterized by a high degree of flexibility, by low neutron temperature, by independent accessibility of the fuel elements, and by comprehensive instrumentation for test purposes. The Plutonium Recycle Test Reactor has a thermal rating of 70 MW, is moderated by unpressurized heavy water, and is cooled by pressurized heavy water (tube outlet pressure, 1050 psig). The reactor is of the pressure-tube and calandria type. Primary control, including reactor shutdown is obtained by regulating the height of the moderator within the calandria by means of the gas balance method first described for the Canadian NPD-I reactor. Supplementary control, primarily for experimental purposes, is provided in the form of replaceable mechanical shim half-rods. The reactor is housed in an all welded steel cylindrical containment vessel, 80 feet in diameter and 122 feet high designed to withstand an internal pressure of 15 psig. A single story wing of conventional construction, 100 feet long by 80 feet wide is provided to house necessary auxiliary services. Heat generated within the reactor will be removed by the heavy water coolant and dissipated in a heat exchanger by boiling off about 206,000 lb/hr of 425 psia saturated steam. Except for a few minor process uses, the steam will be wasted in a jet condenser located in the Columbia River. Charging and discharging of the fuel elements will be done on an individual basis from the top face of the reactor by means of a self-propelled fueling vehicle while the reactor is shut down. Since the

determination of fuel element designs and the optimum fuel-to-moderator ratios are objectives of the program and are thus not known at present, it is possible that new lattice spacing will be required before conclusion of the program. To this end, the entire core of the reactor is designed to be removable so that any necessary changes may be accomplished at minimum cost.

Design studies were started late in 1956. At the present time the Plutonium Recycle Test Reactor design criteria have been developed, design scope has been completed, and detailed design is well under way and scheduled for completion in October of this year. Funds authorized for the reactor are $15,000,000 including contingencies and the heavy water inventory. The estimated construction schedule is 27 months with startup scheduled for July 1960.

The Plutonium Recycle Test Reactor will be operated to provide data on nuclear physics, fuel element design and performance, and engineering technology essential to the plutonium recycle concept including, but not necessarily restricted to, the self-sustaining cycle. Physics tests will yield information of critical startup conditions, reactor control characteristics, and reactivity effects of various lattice parameters and conditions of operation. Such information as long exposure characteristics, isotope composition, and material balance under both equilibrium and non-equilibrium conditions will be obtained. Testing of a number of fuel element designs will be required in developing feasible and economical high-exposure uranium and plutonium fuel elements. As previously stated, plutonium-bearing fuel elements have received comparatively little study and are of particular importance to the economic feasibility of plutonium recycle. The experimental reactor will be operated to provide the development scale quantities of irradiated fuel elements required for conduct of chemical reprocessing and refabrication studies.

We have great hope and confidence that this and our other facilities will adequately serve the program for the collection of the necessary research and development data and that the objectives of the Plutonium Recycle Program can be attained, and in so doing the recycle concept can be developed into a practical competitive power generating method.

II. PHYSICS AND FUEL CYCLE ANALYSES SESSION

Chairman: P. F. Gast, GE-HAPO

1. ANL Fast Reactor Physics - W. B. Loewenstein, ANL

The nuclear performance of a large central station fast power breeder reactor as currently envisaged remains to be experimentally observed. However, many factors entering into such performance can be obtained in an integral manner by performing a series of pertinent critical experiments. Such experiments are particularly useful toward the determination of parameters which cannot currently be obtained through the measurement of certain microscopic reaction cross sections as a function of neutron energy.

Critical experiments at the Argonne ZPR-III facility were performed to obtain information on assemblies featuring fast neutron spectra which are similar to those expected in the larger more dilute fast reactor systems. These experiments contained much smaller amounts of fissionable material than would be found in the actual systems of primary interest. A conceptual fast power breeder reactor with an 800-liter core volume would require a critical mass of about 470 kg of plutonium or 700 kg of uranium-235. In the experimental program no critical assembly contained more than 245 kg of uranium-235 in the core. This was accomplished primarily by the use of higher density systems with graphite replacing the sodium.

Prior to the performance of such experiments calculations are done to determine the general properties of such assemblies. These include a realistic estimate of the expected critical condition. The optimum cylindrical cores, those with minimum critical mass requirements for a given composition satisfy the relationship $0.8 < \frac{L}{D} < 1$ where L is the core height and D is the core diameter.

Such calculations are often done in spherical geometry using the S_n method. These results may then be interpreted to cylindrical geometry using reasonable shape factors based on both calculation and experiment. The shape factor (S.F.) is defined as the ratio of the critical mass of a spherical core to that of a cylindrical one. Both cores have the same composition and similar reflector parameters. For many such systems a shape factor such that $S.F. = M_{sphere}/M_{cylinder} = 0.95 \pm 0.01$ appears to be reasonable in the L/D range of interest. In addition to investigating the properties and spectra of conceptual systems, a large amount of experimental and analytical effort has gone into determing the nuclear properties of both EBR-I and EBR-II. Both of these systems were mocked up in quite some detail on the ZPR-III. Such detailed engineering mockups have served to confirm the suspicions of the analysts that simplified techniques adequate for predicting the nuclear behavior of idealized uniformly reflected systems are often not adequate for predicting the pertinent nuclear constants of a realistic engineering design. The latter usually feature a non-uniform reflector and various geometric asymmetries.

While the system of most interest for long range planning is that utilizing the plutonium-239 and uranium-238 cycle, most of the experimental work to date has been based on systems utilizing uranium-235 as the primary fissionable material. A number of critical experiments using plutonium are available to the analyst. These are limited to the extremely high density systems with very fast spectra such as are found in the Los Alamos Jezebel and Popsy assemblies. In addition, the worth of plutonium relative to enriched uranium has been measured on nearly all assemblies investigated on ZPR-III. Generally, it has been found that a single atom of plutonium-239 is about 1.6 times as effective as an atom of uranium-235 toward maintaining a chain reaction in a fast spectrum.

However, simply because there are no critical experiments involving large amounts of plutonium does not at all imply a lack of confidence in predicting the nuclear behavior of the large conceptual systems. For example, a large conceptual fast power breeder reactor may feature an 800-liter core. About 25 to 30% of the core volume might be occupied by the plutonium-uranium alloy. Such a system would require a critical mass of about 470 kg of plutonium-239. This implies that the ratio of the number of uranium-238 atoms to plutonium-239 atoms per unit volume is around seven. The actual number of plutonium atoms per unit volume is small compared to the fertile, structural and coolant materials. The spectral properties of such a system are determined principally by the latter materials, rather than by the fissile material. Therefore, the more important plutonium constants are concerned with the neutron yield per fission, the fission cross section and the radiative capture cross section, all as a function of neutron energy. The fission cross section is quite well known as is the neutron yield per fission, though the latter is not too well known as a function of neutron energy. Some information is available on the radiative capture cross section on the basis of in-pile measurements from EBR-I as well as some preliminary work as a function of neutron energy done at Los Alamos. The lack of information concerning the inelastic scattering cross section of plutonium does not seriously affect the predicted performance of such a system because the scattering properties of the other core materials are so much more important toward determining the neutron spectrum. It is these other materials which have been quite extensively investigated at ZPR-III and compared with analytical results.

In considering a plutonium loading for EBR_II, where the number of plutonium atoms per unit volume would be much larger than in the system discussed above, the scattering properties of this material are more important and would need to be known with more certainty than they are at present. Some information concerning such properties may be obtained from pertinent material replacement experiments. For instance, if one places a sample of plutonium at the center of a fast system containing no uranium-238, then the observed reactivity effect will be related to the quantity $(\nu-1-\alpha)\sigma_f$ where ν is the neutron yield per fission, σ_f is the fission cross section and $\alpha \equiv \frac{\sigma_c}{\sigma_f}$ where σ_c is the radiative capture

[redacted]

cross section. If the same sample is then placed at the center of a system having a similar neutron spectrum, but containing large amounts of uranium-238 per unit volume, the measured reactivity effect can then again be related to the quantity $(\nu-1-\alpha)\sigma_f$ but additional effects may be attributed to effective inelastic or elastic removal of neutrons from above to below the uranium-238 fission threshold. Actually such experiments can best be interpreted for non-fissionable elements.

Material replacement experiments using a sample of uranium-233 show that this material also shows a great deal of promise in a fast neutron spectrum. Comparing this material with plutonium one finds a lower neutron yield per fission, a higher fission cross section and a lower radiative capture cross section. Atom for atom, one finds that the uranium-233 is generally slightly more effective than plutonium-239 toward maintaining a chain reaction. Another desirable feature of uranium-233 is that $\alpha = \frac{\sigma_c}{\sigma_f}$ appears to increase very slowly with decreasing neutron energy.

It is on the basis of a wide variety of such material replacement experiments as well as the measurement of $\sigma_f^{Pu-239}/\sigma_f^{U-235}$ as well as other spectral indices in typical reactor spectra coupled with the EBR-I measurements on $\alpha = \sigma_c^{Pu-239}/\sigma_f^{Pu-239}$ that the nuclear constants of Pu-239 are much less uncertain today than they were a relatively short time ago. Analysis of the pertinent ZPR-III experiments has served to realistically bracket the uncertainties inherent in the nuclear performance of the conceptual large fast power breeder system.

2. Physics of Plutonium Spikes - B. I. Spinrad, ANL

The problems of reactivity arising in recycled Pu fueled reactors are much simplified if one considered modes of reactor operation whereby fuel is charged and discharged in small increments from any region of the pile. Under these circumstances one can use, for reactivity calculations, the rather reliable concentrations of the various materials obtained from time average of the fuel isotopic concentrations between loading and discharge. The systems become, in effect, systems with constant Pu content for the U-233 as supplied and uranium feed is burned to high depletion as is all the Pu produced from it. In other words, a Pu recycle reactor is a fuel burner, i.e., net conversion rate equal to zero, which uses Pu as a means of being able to use less highly enriched uranium as feed material.

In consequence of this fact, the value of Pu may be considered as equivalent to the value of U-235 incrementally added in the form of enrichment to the feed material. This does not limit the value of U-235 enrichment to above natural but is, in fact, a function of the reactor feed enrichment which can be inferred from relatively simple physics principles. In fact, it is possible to consider Pu recycle even when the feed material is enriched above natural.

When one restricts oneself to the equilibrium situation physics-wise there is no real difference between uniform Pu enrichment, Pu spiking, or Pu seeding. The first two cases represent merely different ways of handling and fabricating the Pu. The third procedure is still another way of handling the fabrication problem but also is a way whereby the processed Pu inventory of the reactor is kept low at the cost of a high throughput rate for the processed Pu.

3. Plutonium Recycle Program Physics - J. R. Triplett, GE-HAPO

The physics portion of the Plutonium Recycle Program may be divided into three main categories:

1. The reactor project: planning, design and startup assistance.
2. Fuel cycle analysis.
3. Experimental physics program.

In the first category the first problem to be solved was the selection of the reactor type which would provide the greatest experimental versatility consistent with demonstration of self-sustaining recycle with natural uranium feed under power reactor conditions of coolant temperature and pressure. The versatility of the pressure-tube concept, as opposed to the pressure-tank, for fuels development led to the rejection of light-water moderation, and of the remaining possibilities heavy-water moderation was chosen as being safer and better adapted to later modifications than graphite. Tube and fuel dimensions, and lattice pitch, were also selected on the basis of versatility for fuels development and physics experiments.

PCTR measurements of lattices resembling those of the PRTR design have been carried out, and calculations of reactor kinetics, power distributions, and heat generations in the core, reflector, and shield have been performed. Extensive zero-power critical experiments at startup are planned.

The second category of effort, fuel cycle analysis, consists of the development and application of computer codes to be used in support of economics analysis of fuel cycles and of the experimental studies of fuel burnup. The customary burnup equations are applied to simplified reactor models, both with and without recycle of plutonium, in order to be able to analyze processing schedules, reactivity transients, etc., for different types of fuels and reactors. General conclusions relating to the plutonium recycle concept have been drawn from these studies. For example, a high moderator temperature is always somewhat unfavorable in recycle systems but is in many cases advantageous in non-recycle systems. Other conclusions of general applicability are that a well-thermalized spectrum is preferable in most recycle reactors to an under-moderated or high conversion-ratio spectrum, and that in fuel cycles using uniformly blended uranium and plutonium under full steady-state conditions, a small processing loss of plutonium has a beneficial effect in view of the resulting reduction of Pu-242 parasitic captures.

These calculations, being based on simplified models, are not sufficiently accurate for the detailed analysis of experiments. An elaborate code known as RBU is being developed, which is in most respects a detailed and accurate mathematical model of an operating reactor, and will provide the needed relationship between the observed fuel burnup and reactor operating characteristics on the one hand, and basic cross-section and similar data on the other. Use of Monte Carlo techniques combined with multigroup-diffusion and burnup equations in this code is expected to result in a very versatile and powerful computational tool.

The experimental physics program is in large part centered on the use of the Physical Constants Test Reactor (PCTR) which is a critical facility designed for the direct measurement of the infinite multiplication constant of a lattice. Thermal utilization, resonance escape probability, and other lattice properties may also be determined in this facility. The PRTR will also be used for several types of direct measurements, such as flux and spectrum distributions and fuel compositions as dependent upon exposure. Finally, the cross-section measurements in the crystal spectrometers will provide invaluable basic data for the recycle program.

4. Plutonium Fuel Cycle Analyses - E. A. Eschbach, GE-HAPO

A major objective of the Hanford plutonium fuel cycle analyses is to determine the value of plutonium as a fuel for thermal reactors. To avoid confusion in our remarks we distinguish the terms "value" and "price". Plutonium value is taken to be a measure of the worth to a prospective purchaser or user. Plutonium price is used to refer to the actual dollars per gram at which a sale is offered or made. This distinction permits the technological and economic factors that establish process value to be considered independently of the many intangibles that may properly belong in the establishment of price.

Before the present HAPO plutonium evaluation code was developed, considerable study was made to determine if the discrete properties of likely combinations of plutonium isotopes could not be used to handily estimate the value of plutonium. Due to the complex interdependence of the pertinent factors no such generic scheme was devised that would not be misleading for some combinations of plutonium isotopes, reactor types, and/or economic environments. This work is summarized in a paper for the 1958 Geneva Conference entitled "The Comparative Economics of Plutonium Fuel Utilization and U-235 Fuel Utilization in Thermal Power Reactors" by Eschbach, Granquist, and Lewis of the Hanford Laboratories Operation, Richland, Washington. It appears that questions pertinent to the use of plutonium as a thermal reactor fuel can best be answered by formulating a model to measure the productivity of plutonium in thermal reactors with all significant interactions and then attempt to separate the effect of different variables by appropriately varying input and output conditions. An HLO computer code characterizes the value of plutonium as a reactor fuel by: making detailed fuel cost estimates for reactors with specific nuclear and operating characteristics optimized with, and without, plutonium enrichment; equating these fuel costs; and solving for an appropriate plutonium value.

It is planned to apply this analysis to whole spectrums of reactors operating in various economic environments as indicated in Table I. Fuel cost is the only consideration of this analysis. Consequently, if it is desired to estimate an over-all plutonium market price by compiling the discrete plutonium values determined for various specific reactor types, care must be exercised to factor in the relative number of each type of reactor actually in operation and the "will" of the discrete operators to participate in a wide market. Such weighting cannot now be done as reactor capital, operating costs, and obsolescence factors are not well enough known to avoid "guessing" rather than estimating the relative number and types of operating power reactors in the future.

TABLE I

VARIABLES AND RANGES UNDER STUDY

Variables	Range of Values
In-Reactor Parameters	
k_{∞}/ε	1.03 to 1.30
Resonance escape probability	Approx. 0.60 to 0.95
Thermal utilization factor	" 0.7 to 0.95
Uranium feed enrichment for U-238 bearing fuel	0.28 to 3.0% U-235
Uranium enrichment for localized or "spike" enrichment	20 to 100% U-235
Average moderator temperature	80 to 600 C
Average specific power	0.1 to 20 MW/ton
Plutonium compositions	Determined from above parameters
Out-of-Reactor Parameters	
Time to reprocess fuel	90 days to 730 days
Reprocessing yields	80 to 99%
Economic Variables	
Use rate for depreciating assets*	1-1/2 to 20%/yr
Use rate for non-depreciating assets*	Zero to 18-1/2%/yr
Chemical reprocessing costs	0 to 20 $/lb U
Uranium-only fuel preparation costs	0 to 40 $/lb U for U-238 & U-235 bearing fuel
Ratio of plutonium-bearing fuel preparation costs to uranium-only costs	1.0 to 4.0
Base price of natural uranium	10 to 80 $/kg U
Separative work unit cost for U-235 from U-238	20 to 320 $/separative unit

*Increments between rates for depreciating and non-depreciating assets of 0, 1-1/2, 3, and 5% are planned.

It is obvious that it is far easier to establish whether or not plutonium has a positive, significant value as a specific thermal reactor fuel than to establish the probable market price of plutonium. Of analyses performed to date the value of plutonium has been significantly different for specific reactors which emphasize the importance of accurate weighting if market price estimates are to be made.

The following preliminary example indicates the range of plutonium values resulting from our studies. The value of plutonium enrichment relative to the value of U-235 enrichment was compared for a "high" neutron economy and a "low" neutron economy reactor as defined in Table II.

TABLE II

Arbitrary Neutron Economy	"Low"	"High"
Average Moderator Temperature	400 C	200 C
K_∞ / ε	1.15	1.10
P	0.75	0.92
f	0.84	0.95
Specific Power MW/ton	10.0	10.0

It appears that the increased neutron absorption cross section of plutonium leads to a greater relative increase in fuel exposure and hence greater value of plutonium for the lower neutron economy reactor. When it costs \$10/#U extra to process plutonium-bearing fuel as compared to uranium-only fuel charges, the value of plutonium for the "high" neutron economy reactor was determined to be \$6.25 per gram of Pu-239 + Pu-241 contained and to be \$10.50 per gram of Pu-239 + Pu-241 for the "low" neutron economy reactor. The following are important to these figures.

1. The "high" neutron economy reactor has the lowest total fuel costs for comparable processing costs per pound of fuel.

2. These reactors do not represent specific designs although it is generally true that high neutron economy reactors cost more to build than low neutron economy reactors, and no corrections are made for such differences as necessary if total cost of electrical power from a reactor were being determined.

3. A somewhat idealized highly thermalized neutron spectrum using appropriate Westcott cross sections is employed to simplify fuel exposure calculations. This is necessary to rapidly survey the value of plutonium in many reactor types, and the limitations of such methods will be determined.

To avoid obsolescence, the plutonium value can be expressed as a function of the cost increment of processing plutonium-bearing fuel over comparable uranium-bearing fuel. This permits determination of the plutonium value for any incremental fuel processing cost within the data range. The

plutonium value for the previous examples can be represented by a simple slope intercept equation as follows, regardless of absolute fuel processing costs up to $60/#U which is the maximum value studied.

Pu value $/gm Pu-239 + Pu-241 = A - (B) (Pu fuel processing cost minus uranium fuel processing cost in $/#U)

for the high neutron economy reactor: A = $11.50 and B = 0.625

for the low neutron economy reactor: A = $15.50 and B = 0.497.

Pertinent to the fuels processer is the loss of plutonium upon reprocessing. An unusual characteristic of plutonium recycle is that at equilibrium recycle, a higher fuel exposure is achieved if some plutonium is discarded during each fuel reprocessing. This is true because the relative quantity of parasite plutonium isotope 242 is reduced in the equilibrium mixture if some plutonium is discarded during each recycle. Computations to date indicate that a maximum attainable exposure is attained with processing losses of 3 to 4% per cycle. At high values of feed enrichment (1.6% U-235) the maxima are very slight indicating little sensitivity to plutonium processing losses. The greatest sensitivity occurs with small leakage reactors with natural or depleted uranium feeds. These results suggest the economy of selling some plutonium during each recycle and, in a high priced plutonium market, minimum fuel costs may occur with a sale of 20 to 30% plutonium per cycle.

III. CHEMICAL REPROCESSING SESSION

Chairman: W. H. Reas, GE-HAPO

1. Aqueous Decontamination of Plutonium From Fission Product Elements - R. S. Winchester and W. J. Maraman, LASL

A survey of aqueous plutonium recovery and purification methods as applied to plutonium-rich reactor fuels has been made. These methods consist of (1) precipitating plutonium fluoride, oxalate and peroxide, (2) sorbing on anion exchange resins, and (3) solvent-extracting with tri-n-butyl phosphate.

The standard fluoride, oxalate and peroxide precipitation methods gave decontamination factors of plutonium from the typical fission product elements zirconium, molybdenum, ruthenium and cerium ranging from 0.002 for cerium to 140 for molybdenum. These results indicate that precipitation methods are unsatisfactory.

Sorption of plutonium on anion exchanger resins from 8 M nitric acid solutions and the subsequent elution resulted in decontamination factors of 16 for ruthenium to 52 for zirconium.

Solvent extraction of plutonium in 2.7 M nitric acid by tri-n-butyl phosphate gave decontamination factors ranging from 1.3 for ruthenium to greater than 100 for molybdenum. Extraction of plutonium from 8 M nitric acid solution by primary, secondary, tertiary and quaternary amines gave greater over-all decontamination.

The secondary amine was selected for recovery processing.

Plutonium is recovered and purified from pyrometallurgical and analytical wastes on a 300-gram scale using secondary amine solvent extraction followed by oxalate precipitation. A flowsheet for this process is discussed.

2. Pyrometallurgical Purification of Plutonium Reactor Fuels - J. A. Leary, R. Benz, D. F. Bowersox, C. W. Bjorklund, K.W.R. Johnson, W. J. Maraman, L. J. Mullins, and J. G. Reavis, LASL

Pyrometallurgical methods for processing spent plutonium fuel have been studied experimentally. Because the principal interest in plutonium fuel is for high burnup, fast power reactors, purification is essentially a problem of fission product bulk removal with no stringent specification on any particular contaminant.

Preliminary experiments were conducted using the 12 atomic percent cobalt - 88 atomic percent plutonium eutectic which has a melting point of 405 C. However, a 9.5 atomic percent iron - 90.5 atomic percent plutonium eutectic, melting at 410 C, has been used in the majority of the experiments. Because shielded facilities for handling high burnup plutonium are not available, all experiments were conducted with the synthetic spent fuel "fissium". This material was prepared by dissolving typical fission product elements in a melt of the eutectic and chill-casting into rods of homogeneous composition. The amount of each fission product element corresponded approximately to a 10 percent fuel burnup.

Pyrometallurgical methods that have been studied are (1) liquation, self-drossing and filtration, (2) slagging by the addition of oxide, carbide or halide, (3) liquid metal extraction, (4) complete conversion to halide, followed by filtration and selective reduction of the plutonium, and (5) electrorefining. Experimental techniques and results are presented for each method.

By holding a melt of fissium at a temperature slightly above the melting point of the fuel, insoluble constituents in the form of elements or compounds have been removed by filtration. Zirconium, lanthanum, and molybdenum are removable to varying degrees by this simple method.

Results obtained by slagging as oxides in magnesia crucibles, as carbides using graphite, and as halides using fluorides or chlorides are presented. In general these results were consistent with predictions based on thermodynamic considerations. In addition, process variables such as temperature and equilibration times are discussed.

The results of solubility studies in liquid-liquid metal extraction systems are presented and discussed from the processing standpoint.

Conversion of the spent fuel to the chloride by hydrochlorination of the hydride has been found to be an effective purification method. After fusing this chloride with molten sodium chloride to form the plutonium chloride - sodium chloride eutectic and to remove volatile fission product chlorides, the material is filtered to remove ruthenium. Removal of rare earth fission products occurs in the reduction step where the plutonium is recovered as the metal. A similar method using a fluoride cycle has also been investigated.

Results on purification by electrorefining in a molten salt electrolyte are also presented.

3. Pyrometallurgical Processing of Plutonium Alloys - M. Ader, ANL

The review presented is limited to recent work of the Chemical Engineering Division applicable to pyroprocessing of plutonium alloys and, in particular, of plutonium present in EBR-II fuel and blanket material.

A proposed loading of the EBR-II core employs "fissium" alloy containing about 70 w/o uranium, 20 w/o plutonium, and 10 w/o fission product elements. The latter elements (Zr, Mo, Ru, Rh, Pd) serve to stabilize the fuel against radiation damage and are allowed to build up to an equilibrium concentration in the reprocessing cycle. The EBR-II blanket will consist of depleted uranium or, possibly, uranium - 5 w/o chromium alloy.

The principal purposes of reprocessing are elimination of radiation damage and reconstitution of the fuel, goals which are realized by melt refining of the core and enrichment with plutonium obtained from magnesium extraction of the blanket.

Melt Refining. Fuel from the reactor, after decladding, is kept molten at 1300 C in an oxide crucible in an inert atmosphere for several hours. A gross separation of fission products is effected by oxide dross formation and volatilization. The metal phase retains uranium, plutonium, and noble fission elements (atomic numbers 40 to 47) and is poured into a mold prior to remote fabrication into fuel pins and replacement in the reactor. The dross, which contains about five percent of the fuel, is treated by a separate procedure (see "dragout" processing below) to recover its uranium-plutonium content and to adjust the noble fission element concentration to a desirable value.

Early melt refining experiments with irradiated uranium containing < 50 ppm plutonium showed that plutonium concentrated in the dross by a factor of about 1.7. [Conc. factor defined as $(Pu/U)_{dross} \div (Pu/U)_{charge}$.] The general behavior of plutonium is not appreciably altered in going from the tracer to the 20 w/o level, a concentration factor of 1.2 in the dross being found with uranium-plutonium fissium alloys melted in zirconia. Similar experiments with uranium - 1 w/o plutonium showed that americium

concentrated in the dross by a factor of 59. Neptunium, at the tracer level, was found to remain with the metal phase.

Mechanism and rate of attack by molten uranium have been investigated for alumina, magnesia, zirconia, thoria and beryllia crucibles. Of these, lime-stabilized zirconia is preferred for use in melt refining because (1) it does not contaminate the melt, as alumina does with aluminum; (2) no gaseous product is evolved to disrupt the dross layer, as in the case of magnesia; (3) it is cheaper than thoria or beryllia; (4) cerium removal rates are faster in zirconia than in magnesia; and (5) it possesses good thermal shock properties. Ingot yields obtained in melt refining two to three kg amounts of uranium "fissium" alloy in zirconia averaged 96.6 percent at 1300 C and 94 percent at 1400 C.

Magnesium Extraction.

a. Liquid-Liquid Extraction from Uranium. Laboratory experiments have indicated a slow approach to equilibrium. In alumina, using plutonium-to-uranium weight ratios of 0.01 and approaching equilibrium from both directions, a mole fraction distribution ratio of 0.19 (+0.01, -0.005) was found for plutonium between magnesium and uranium at 1190 C. This result is in agreement with the value of 0.21 found by Motta at 1150 C at the tracer level.

b. Liquid-Liquid Extraction from Uranium-Chromium. The interest in this alloy stems from the existence of a low melting (859 C) eutectic at 5 w/o chromium. Partition of plutonium, at plutonium-to-uranium weight ratios of 0.01, between uranium - 4.5 w/o chromium and magnesium has been studied in the temperature range 930 to 1195 C, alumina crucibles and approach to equilibrium from opposite directions being employed. The mol fraction distribution ratios may be represented graphically on a plot of log K versus the reciprocal of temperature in degrees Kelvin. A least squares straight line drawn through the points is described by the equation

$$\log K = -2.134 \pm 2010/T.$$

The heat of transfer of plutonium from uranium - 4.5 w/o chromium to magnesium is -9.2 ± 1.6 kcal/g-atom. The presence of 4.5 w/o chromium in uranium has little effect on the distribution coefficient at 1190 C.

Pilot plant studies have been limited to extractions with the uranium - 5 w/o chromium - magnesium system, primarily to circumvent (pressure) operations above the boiling point of magnesium and difficult corrosion problems inherent in uranium-magnesium extractions at 1150 to 1200 C. To date, only simple batch extraction has been investigated although comparison of various extraction schemes indicates that countercurrent extraction is most advantageous from the standpoint of product purity and magnesium economy. Employing four successive batch extractions of 0.68 kg uranium - 5 w/o chromium - 1 w/o plutonium with magnesium at 930 to 950 C in tantalum, 98 percent of the plutonium was extracted using Mg/U-Cr weight ratios of about 0.86. The mol fraction distribution

ratios obtained in each extraction (mg/U-Cr = 0.25 to 0.30) are essentially in agreement with laboratory data obtained in alumina crucibles.

Similar experiments done in graphite crucibles, however, demonstrated considerably lower distribution ratios (0.18 to 0.22) and somewhat poorer plutonium recoveries. This is attributed to the ability of graphite to react with and remove plutonium from magnesium solution, as shown by the following: 506 g of magnesium-plutonium alloy containing 1.27 g plutonium was liquated in a graphite crucible at 900 C under helium and sampled at six-hour intervals. After 18 hours, 98 percent of the plutonium had been scavenged by the graphite. A semi-logarithmic plot of fraction plutonium remaining versus time indicates a first-order reaction. The behavior in tantalum is dissimilar although some loss of plutonium (about 20 to 30 percent) has been noted after 24 hours' liquation. Tests with tantalum as container material are still in progress. Tantalum coated with tantalum monoboride is also being investigated.

While advantageous in lowering operating temperatures, the use of uranium-chromium alloy is disadvantageous in that chromium contaminates the plutonium product. At 950 C chromium extracted by magnesium from a uranium - 5 w/o chromium alloy amounts to 0.156 weight percent of the magnesium phase and about 60 percent of the weight of plutonium extracted, assuming four equal weight magnesium extractions. An eight-fold reduction of the chromium content of the plutonium product may be achieved by cooling the magnesium extract to 675 C, to precipitate 88 percent of the chromium, and separation of chromium prior to distillation of magnesium. A further reduction (factor of 16) in chromium content could be realized if a large percentage of the magnesium were distilled away before precipitating the chromium. Another procedure under investigation for removing chromium from the plutonium-bearing magnesium phase is addition of calcium to form the magnesium - 18 w/o calcium eutectic (m.p. 516 C), the mixture is cooled to 550 C to precipitate 99 percent or more of the chromium previously introduced by magnesium extraction of uranium - 5 w/o chromium at 950 C. After separation of the chromium, magnesium and calcium are distilled off to concentrate a plutonium product.

The build-up of chromium in the reactor core following any of the above-mentioned procedures would be tolerable over many passes of the fuel through the processing cycle.

c. Solid-Liquid Extraction. Solid-liquid extraction experiments indicate that a uranium blanket may be processed for plutonium recovery via magnesium extraction at 800 C without the use of alloying agents, such as chromium. The technique that has been employed involves conversion, in alumina crucibles, of uranium-plutonium alloy to powder by hydriding at 225 to 250 C and dehydriding at 300 C, in the presence of magnesium which is unaffected. Argon is introduced, the temperature is raised to 800 C, and the mixture is agitated by gas bubbling or stirring.

At the tracer level, plutonium was found to extract considerably slower at 700 C than at 800 C. In addition, a reversal of the extraction process appeared to occur after about two hours of mixing at 800 C. Appreciable (48 to 60 percent) extraction of plutonium at 800 C was also obtained by powdering uranium - 1 w/o plutonium alloys and using magnesium-to-uranium weight ratios of 0.2 to 0.4 and one to three hours contact. A double extraction of powder from the same alloy, using magnesium-to-uranium weight ratios of 1.2 and one hour contacts, resulted in over 90 percent of the plutonium being extracted. Graphite crucibles were again found to be unsuitable because of absorption of plutonium (see above).

Although a solid-liquid process for plutonium recovery appears feasible, further experimentation is necessary to establish equilibrium conditions. The mechanism of extraction requires study since it does not appear that true extraction of plutonium from a solid solution in uranium is involved. The hydriding-dehydriding step may effect an initial separation, leaving plutonium preferentially situated in grain boundaries or particle surfaces. Extraction or dissolution of plutonium from these sites may be followed by back diffusion or adsorption. Work on this project has been discontinued.

Pyrozinc Studies. A recent approach to recovery and decontamination of fissile materials is crystallization from liquid metal solvents. In this connection, various applications of molten zinc, grouped under the name "Pyrozinc" processes, are being investigated.

The Pyrozinc processes have in common the following features: (1) alloys or oxides containing the actinide metals are dissolved in zinc or zinc-magnesium at 700 to 800 C; (2) due to the large temperature coefficient of solubility of actinide metals, quantitative precipitation of their zincides is feasible by cooling; (3) separations from contaminants occur during crystallization because of favorable solubility or coprecipitation phenomena; (4) the zincides are separated by decantation, filtration, or centrifugation; (5) the zincides are decomposed by volatilization or leaching to yield product. The product may be coalesced by heating above its melting point.

Some of the advantages, briefly noted, in using Pyrozinc processing are: (1) metal is maintained in the metallic state; (2) operations involved are readily adapted to batch processing; (3) a wide variety of fuels and blankets can be handled; (4) severe inert atmosphere restrictions of operations such as melt refining may be eliminated by the use of oxidation protectants, e.g., dissolved aluminum or salt flux covers; and (5) separations from fission products may be enhanced (e.g., as compared to melt refining) by selective oxidation and reduction prior to recrystallization of zincides.

Solubilities of plutonium and uranium in zinc solution have been determined. A graph of the logarithm of weight percent plutonium and of weight percent uranium in solution versus reciprocal of absolute temperature gives parallel straight lines, the solubility of plutonium in

zinc being almost twice that of uranium in the range 450 to 760 C. The concentration of both plutonium and uranium, however, are reduced when both elements are present in appreciable amounts. At plutonium-to-uranium concentration ratios of 0.01 (below plutonium saturation value) coprecipitation of plutonium by the uranium-zinc intermetallic, U_2Zn_{17}, was found to obey the Doerner-Hoskins logarithmic distribution law, the constant λ being 0.66. A λ value of 0.64 was found at the tracer level where the initial plutonium-to-uranium concentration ratio was 0.0001. Coprecipitation of thorium and americium by U_2Zn_{17} have also been studied at the tracer level, λ values of 1.0 and 0.67, respectively, being found.

Individual solubilities of uranium, zirconium, molybdenum, ruthenium, rhodium, palladium, barium and cerium in zinc have been measured in the temperature range 500 to 800 C. Solubilities of individual and mixed ("fissium") fission product elements in uranium-saturated zinc and uranium-saturated zinc - 6 w/o magnesium solutions have also been determined.

"Dragout" Processing of EBR-II. As stated in the introduction, a small side stream of the fuel material must be processed for removal of noble elements to limit their build-up in the "fissium" alloy. This side stream will probably consist largely of the crucible skull, processing of which is necessary for recovery of occluded fissionable material. Preliminary experiments with skulls resulting from melt refining uranium "fissium" alloys in zirconia crucibles showed that 74 to 89 percent of the uranium was dissolved in situ by zinc - 6 w/o magnesium solution. The extent of UO_2 reduction in these experiments as compared to ordinary dissolution of occluded uranium is not known, although the reduction of uranium dioxide, uranium trioxide, and uranium tetrafluoride to metal by zinc - 6 w/o magnesium has already been demonstrated. Presumably, plutonium could also be recovered from such skulls.

The method proposed for removal of the noble elements from the uranium-plutonium-fission product solution in molten zinc-magnesium involves (a) hot filtration to remove the unreacted oxides and molybdenum (which has low solubility in this solvent), followed by (b) cold filtration to recover the bulk of the actinide metals as their intermettalic compounds. Experimental results indicate that rapid cooling ($\sim$30 to 50 deg C/min) to the filtration temperature minimizes precipitation of a ruthenium containing ternary alloy.

Pyrozinc Processing of EBR-II Blanket. Recovery of plutonium from the EBR-II blanket by a pyrozinc process is presently under investigation. Successful application of this method would eliminate the need for addition of chromium to the uranium blanket (see above) since all operations would be performed at 800 C or lower. A proposed flow sheet involves (a) dissolution of the uranium - 1 w/o plutonium alloy in zinc at 800 C; (b) cooling to $\sim$500 C to precipitate uranium plus plutonium zincides; (c) separation of the zincides by filtration or decantation; (d) addition of sufficient magnesium to form the zinc - 46 w/o magnesium eutectic (m.p. 340 C), thereby dissolving plutonium and precipitating uranium metal: the solubility of uranium in the eutectic at 450 C is -0.05 weight percent; (e) removal

of uranium metal by filtration or decantation; and (f) distilling off zinc and magnesium to yield a plutonium concentrate. Tests of step (d) with U_2Zn_{17} alone have demonstrated that uranium will precipitate as relatively large-sized and filterable aggregates after addition of enough magnesium to form the zinc - 46 w/o magnesium eutectic.

4. Aqueous Processes for Plutonium Recycle Test Reactor Fuels - M. T. Walling, Jr., GE-HAPO

Research and development effort at Hanford in behalf of aqueous processing methods for plutonium recycle reactor fuels has two objectives.

The first is to assure that spent fuel from the Plutonium Recycle Test Reactor (PRTR) can be safely, reliably, and economically processed in existing Hanford separations facilities.

The second objective is to develop separation processes for plutonium recycle reactor fuels which will be sufficiently economic to make the concept of recycle of plutonium economically competitive with other approaches for fueling power reactors.

The major problem encountered in adapting existing Hanford separations plants to the reprocessing of PRTR fuels is devising means of dissolving the Zircaloy-2 clad PRTR fuels in the existing plant dissolvers. These are constructed of 300-series stainless steel and were designed to process aluminum-clad metallic uranium fuels. Present plans call for use of the Zirflex process to selectively dissolve the Zircaloy-2 cladding off these fuels, thus permitting the fuel meats to be dissolved in nitric acid and processed in essentially the same manner as Hanford production fuels. In the Zirflex process Zircaloy-2 fuel cladding is selectively dissolved in either a boiling aqueous solution of ammonium fluoride or in a boiling solution of ammonium fluoride and ammonium nitrate. The stoichiometry of the zirconium dissolution in these media may be adequately represented by the following equations.

(1) Dissolution in NH_4F

$$Zr(s) + 6NH_4F \longrightarrow 2NH_4^+ + ZrF_6^= + 4NH_2(g) + 2H_2(g)$$

(2) Dissolution in NH_4F-NH_4NO_3

$$Zr(s) + 6NH_4F + 0.5\ NH_4\ NO_3 \longrightarrow 2NH_4^+ + ZrF_6^= + 5.0\ NH_3(g) + 1.5\ H_2O$$

The stoichiometry indicated is inferred from analysis of the gaseous products and from the fact that the dissolution process approaches a negligible rate as the ratio of fluoride to zirconium in solution approaches six.

A mixture of ammonium fluoride and ammonium nitrate is preferred as the dissolving medium since the concentration of hydrogen in the off-gas is thereby reduced to a near-negligible amount and it is possible with this medium to dissolve the tin in Zircaloy-2.

Uranium or uranium dioxide is slightly attacked by the decladding medium. Fortunately, however, the product is sparingly soluble uranium(IV) fluoride (or double fluorides of Uranium(IV) and ammonium ion) so that little loss of uranium (or plutonium) results in the decladding operation.

Anion exchange has been demonstrated to be a powerful reprocessing method for feeds from which only plutonium need be recovered and is accordingly worthy of consideration for plutonium recycle reactor fuels.

Since plutonium is one of the very few metal ions which form stable anionic nitrate complexes in solutions of moderate nitrate ion concentration, it is possible to obtain excellent separation from virtually all contaminants. Separation from uranium by a factor of greater than 10^6 has been demonstrated in a single cycle of anion exchange. Niobium-95 limits fission product decontamination. However, gamma decontamination factors in excess of 10^4 have been obtained in a single cycle and of the order of 4×10^7 in two successive cycles.

With commercial anion exchange resins of sufficient stability, kinetics limits the concentration of plutonium which can be established on a resin column. In many cases, however, raising the temperature to 50 to 60 C yields acceptable column loadings (60 - 120 g Pu per liter of resin, depending on the resin used) and enables the plutonium to be eluted off the resin (in dilute nitric acid) at concentrations of 50 to 60 g Pu/l. Although plutonium loading and elution behavior is quite acceptable with Dowex-1, X-4 (50-100 mesh), Permutit SK (20-50 mesh) is preferred for this application because of its greater stability toward radiation. This resin remains usable after exposure to gamma radiation doses (cobalt-60 source) of up to 4×10^8 R.

5. Continuous Plutonium(IV) Oxalate Precipitation, Filtration, Calcination, and Hydrofluorination - H. W. Crocker, GE-HAPO

Plutonium(IV) oxalate has been precipitated, filtered, calcined, and hydrofluorinated to plutonium tetrafluoride in a batch process at HAPO for several years. The batch process time cycle is approximately eight to ten hours for the complete conversion from plutonium(IV) nitrate solution to plutonium tetrafluoride powder. Processing capacity has been restricted due to a fixed number of processing stations of limited capacity, and associated nuclear safety considerations. Plutonium losses to the filtrate have resulted in high recycle to the recovery operation. Batch process product is not of uniform quality. High maintenance costs have been incurred due to the corrosive action of chemicals on furnaces, filter boats, and other processing equipment.

The 234-5 Development Operation has completed the development of an efficient continuous processing system for the conversion of plutonium nitrate solution to plutonium tetrafluoride. A large number of experimental runs, including periods of sustained operation, have successfully demonstrated the dependability of the process and experimental equipment. Operation and maintenance of the equipment is simple. The plutonium tetrafluoride product can be prepared in high yields and is suitable for reduction to plutonium metal.

Plutonium nitrate feed solution approximately 100 g/l Pu and 3.5 - 5.5 M HNO_3 is continuously metered into an agitated reactor simultaneously with 1 M $H_2C_2O_4$. A side outlet is provided on the reactor to permit overflow of the plutonium oxalate slurry into the filtration pan. Slurry residence time in the reactor is fifteen minutes. Precipitation takes place at room temperature. Filtration, cake washing, and air drying are done on a rotary vacuum drum filter which rests in the filtration pan. Filtrate is continuously removed by vacuum to a filtrate storage vessel where excess oxalate is decomposed by contact with potassium permanganate, followed by hydrogen perioxide in a batch or continuous process. The plutonium oxalate cake is removed from the revolving drum filter by a scraper and falls into the continuous screw calciner. Calcination is accomplished at 300 C with a powder residence time of 15 minutes in the calciner. Oxide powder is then transferred into the hydrofluorination reactor where it is contacted with a countercurrent stream of gaseous anhydrous hydrogen fluoride at 500 C. Off-gases are filtered to retain any plutonium solids, and then transferred to the waste sump. The plutonium tetrafluoride powder is collected in a receiver vessel and is periodically removed for shipment. The reactor is controlled at a negative gauge pressure of four inches of water during the processing operation.

A glass reactor five inches in diameter and twelve inches high served as the precipitation vessel. The polyethylene rotary drum filter is eighteen inches in diameter, three inches wide, and has eight filter sections. The filter cloth is Dynel SD-9. Total filtration area is 0.8 square feet. The filter nests into a 304-L stainless steel filter pan. The pan and filter are critically safe. The screw calciner is thirty-six inches long, two inches in diameter, with a two-inch screw mounted on a one-inch shaft enclosed in a 304-L stainless steel shell. A rotary grinder is located one foot from the entry end to pulverize lumpy cakes. The calciner, as designed, is critically safe. The hydrofluorination tube is of inconel, eight feet long and two inches in diameter, and is lined with an 0.01-inch thick Baker Gold Alloy 413 liner. The tube is sized to prevent nuclear criticality when completely filled with a powder of 2.47 g/cc plutonium density. Electric vibrators provide the powder bed movement within the reactor. The vibratory method of powder movement was selected because of mechanical simplicity and low powder entrainment.

In the experimental program, plutonium nitrate feed solutions were prereduced with hydrogen peroxide to attain a 100 percent plutonium(IV) valence state. Reduction was done on a batch-basis or as part of the continuous precipitation by using an oxalic acid-hydrogen peroxide precipitant mixture. A five-inch mercury vacuum was required to obtain a

dry cake. Oxalic acid excess must be maintained within a few hundredths of 0.1 M to prevent pasty cake formation which tends to blind the filter cloth. Plutonium losses to the filtrate were normally two percent. Plutonium nitrate solutions of 10 g/l to > 100 g/l have been processed in the equipment. Spectrographic analyses of plutonium oxide and plutonium tetrafluoride samples have confirmed the absence of corrosion product contamination from the experimental equipment. Conversion of the plutonium oxide to plutonium tetrafluoride was greater than 98 percent. The plutonium tetrafluoride may be reduced to metal with yields normally in the 98 percent range. Normal processing capacity of the experimental equipment is 500 grams of plutonium per hour.

The experimental equipment was used as a basis for a plant scale plutonium processing facility. A number of mechanical difficulties were experienced during the startup of the plant unit. The inadequacies were corrected and normal plant production is now processed through the new facility.

6. The Recovery of Plutonium from Various Sources at the ORNL Metal Recovery Plant - W. H. Lewis and R. E. Brooksbank, ORNL

Aside from the recovery of fissionable material from reactor fuel elements, the ORNL Metal Recovery Pilot Plant has engaged in several programs for plutonium recovery from unusual materials. Plutonium has been recovered from (1) plutonium-aluminum alloyed rods from the Canadian NRX reactor; (2) plutonium oxide powder, dust, billets and chips from Chalk River; (3) gallium-alloyed nickel coated plutonium slugs encased in mild steel cans from the Clementine reactor; and (4) bomb test debris and sand from the Nevada proving ground. Processing of these materials consisted of dissolution or leaching of the feed material, separation and decontamination by solvent extraction and product concentration by ion exchange or evaporation methods.

Two separation programs have been undertaken in the facility in which plutonium-aluminum alloy rods were processed. Feed for the process was prepared by dissolving 11-12 slugs, batchwise, in boiling 5.4 to 6.0 M nitric acid containing 0.05 M mercuric nitrate. The first dissolution of this type required 62 hours of digestion at 108 C to obtain 80 percent dissolution. Two additional 44-hour digestions were required for complete dissolution of the slugs. The second dissolution was made using 6.0 M HNO_3 containing 0.05 M $Hg(NO_3)_2$ and a total of 36 hours was required for complete dissolution; however, the solution was refluxed for 58 hours to further insure complete dissolution. The average dissolution rate was approximately 3 $mg/min/cm^2$. The solutions obtained from this operation was blended with 2 M uranyl nitrate solution and adjusted to the Purex process feed specifications. Dilution of the feed was necessary because of limited plant shielding. After two cycles of solvent extraction the plutonium product was sorbed on cation resin for concentration.

Approximately 1.5 kg of plutonium oxide in powdered form and 377 grams of miscellaneous solid scrap was received from Chalk River for plutonium recovery. Complete dissolution of the oxide was obtained in 52 hours

(~ 350 grams/dissolving) using boiling 13.4 M HNO_3 and 0.02 M HF as a dissolvent. Acid concentrations of 10 - 13.4 M nitric acid and 0.01 to 0.03 M HF were tried before the optimum reagent concentration was selected. The scrap, mostly reduction crucible slag, billets and chips required 70 hours for complete dissolution. One cycle of solvent extraction which consisted of an extraction column and a stripping column, using a Purex second plutonium cycle flowsheet was required for purification of the plutonium. Concentration was by ion-exchange.

Elements from the Clementine reactor were nickel-coated, gallium-alloyed plutonium rods encased in mild steel cans. The plutonium slug along with a 30-gram normal uranium wafer was dissolved in a boiling solution 13 M HNO_3 and 0.05 M HF. Complete dissolution of one (455 g) fuel assembly could be accomplished in 12 hours by refluxing the dissolvent. The average dissolution rate for these slugs was 17 mg/min/cm^2. One cycle of solvent extraction was required for separation and decontamination. The plutonium product was concentrated by ion-exchange.

A feasibility study of recovering uranium and plutonium from bomb test debris was conducted in the Metal Recovery plant. The debris which consisted of sand, rocks, wood splinters, and metal fragments was packaged in 30-gal drums for shipment to ORNL. The drums and contents were dissolved and leached with nitric acid. Each batch dissolving used 1620 pounds of the raw material. Dissolution of the iron drums, necessary to minimize the health hazard, was accomplished using 3.0 M HNO_3 heated to 70-80 C. After the initial reaction had subsided, concentrated nitric acid was added in small (50-gal) increments and the mixture agitated and refluxed for four hours. After the digestion period, the solution was transferred to the feed tank and the sand contacted with fresh nitric acid. After leaching and dissolving the sand was slurried with water and removed to waste. The plutonium obtained from this operation has ranged from 5.5 to 25 grams per ton of sand and averaged 10 g/ton. One cycle of solvent extraction consisting of an extraction and stripping column was required for recovery. Concentration of the product was by evaporation. The residual plutonium content of the sand after leaching was 0.5 g/ton, indicating a recovery of 95 percent.

7. Possible Uses of Plutonium Hexafluoride in Reactor Fuel Reprocessing - R. K. Steunenberg, ANL

The spent fuel elements from a nuclear reactor may contain plutonium produced by the neutron capture of uranium-238, or plutonium might have been placed in the fuel prior to its insertion into the reactor. In either case, it is desirable to recover decontaminated purified uranium and plutonium separately from the spent reactor fuel.

Fluoride volatility processes for the recovery of uranium from reactor fuels are based on the volatility of uranium hexafluoride (triple point 64 C). In one such process, metallic uranium fuels are dissolved in liquid bromine trifluoride. The dissolution reaction converts uranium to the hexafluoride, plutonium to the tetrafluoride, and the fission products to their respective fluorides, most of which are non-volatile. The uranium

hexafluoride and the bromine fluorides are distilled from the dissolver leaving the plutonium and non-volatile fission product fluorides behind as a residue. The uranium is then separated from the bromine fluorides and volatile fission product fluorides by fractional distillation. Irradiated fuel was subjected to this process in the pilot plant and good quality uranium hexafluoride was produced with a fission product decontamination factor exceeding 10^8.

Two methods have been proposed for the recovery of plutonium from the residue. The original plan was to wash the dissolver with an aqueous solution and to recover the plutonium by a small-scale Purex-type solvent extraction procedure. It has been shown, however, that the plutonium can be volatilized as the hexafluoride by treating the residue with fluorine at a moderate temperature (300-400 C). It is believed that the final purification and decontamination of the plutonium hexafluoride might be accomplished by fractional distillation or by fluorination-decomposition cycles.

Many reactor fuels are clad or alloyed with stainless steel, zirconium or aluminum, which cannot be dissolved in bromine trifluoride. These fuels might be handled by an aqueous dissolution followed by calcination of the dissolver solution to the oxides. These oxides could then be fluorinated, perhaps in fluidized beds, volatilizing the uranium and plutonium as the hexafluorides. Laboratory work on this type of process is in the initial stages at the present time.

Plutonium hexafluoride is similar to uranium hexafluoride in its physical properties, but its chemical stability is very limited. It is decomposed by heating, by its own alpha emission, and by reduction on most materials of construction. Thus, rather special techniques are required to handle it as a process material.

Recent laboratory data, however, lend encouragement to this scheme. It was shown that plutonium tetrafluoride from various sources, including residues from the dissolution of uranium-plutonium alloys in bromine trifluoride, is converted to the hexafluoride at a reasonable rate for a chemical process. The kinetics of the reaction were investigated under varying conditions of time, temperature, fluorine pressure and type of material. The fluorination rate of plutonium tetrafluoride from any one source at one atm fluorine pressure may be expressed by the equation:

$$\text{mg } PuF_4 \text{ reacted} = k\, S\, t\, e^{-\frac{\Delta E}{RT}}$$

where k is a constant, S the surface area, t the time, ΔE the activation energy, and T the absolute temperature. The activation energies were 10 to 12 kcal/mole for the various types of plutonium tetrafluoride. The effect of the partial pressure of fluorine on the fluorination rate is not clearly understood. As the partial pressure was increased from 0.25 to 1.0 atm, the fluorination rate passed through a maximum at about 0.75 atm fluorine pressure.

[redacted]

Experiments have also been conducted on the transfer of plutonium hexafluoride in the vapor phase. It appears to be transferred with relative ease in a stream of fluorine. In a helium stream, however, it was necessary to prefluorinate the nickel equipment very thoroughly before quantitative transfers could be made.

The conclusions pertaining to this work may be summarized as follows:

1. Plutonium hexafluoride can be produced at a satisfactory rate under reasonable process conditions by fluorination of the tetrafluoride.

2. Plutonium hexafluoride can be transferred satisfactorily in the gas phase in a stream of fluorine; in a helium stream, good transfers are possible only under conditions of careful prefluorination.

3. Plutonium hexafluoride can be deposited in a heated packed section, probably as a result of thermal decomposition to a non-volatile form.

4. In consideration of these facts it seems possible that fluoride volatility processes might be applied profitably to the recovery of uranium and plutonium from spent reactor fuels.

8. The Los Alamos Plant for Remotely Controlled Production of Plutonium Metal (Film) - A. N. Morgan, Jr., R. D. Baker, W. C. Hazen, A. V. Henrickson, W. D. McNeese, and R. L. Thomas, LASL

Plutonium metal has been produced in kilogram quantities in remotely controlled equipment at the Los Alamos Scientific Laboratory since 1951. The chemistry consists of peroxide precipitation of plutonium from a nitrate feed solution, conversion of the peroxide to plutonium tetrafluoride, and reduction of the tetrafluoride to plutonium metal. The aqueous chemistry is performed in a glass reaction vessel.

Solid plutonium peroxide is separated from the supernatant liquid by filtering through a sintered platinum disc. Direct conversion to plutonium tetrafluoride is accomplished by passing anhydrous hydrogen fluoride through the peroxide cake. The tetrafluoride powder is then mixed with calcium, poured into a ceramic crucible and reduced to plutonium metal. Chemical operations are carried out on a batch basis.

The unit consists primarily of an enclosed horizontal conveyor, which transports the plutonium between chemical processing stations, and pneumatic cylinders, for vertical transmission into each processing station. Incorporated in the design are features which exclude all accessory equipment from the contaminated enclosure. In addition, each processing station is constructed as an integral part so that replacements can be made simply without spread of contamination. Manual servicing of the unit requires loading of the plutonium nitrate container, crucible and gasket, extraction of the plutonium pellet and residues, and maintaining reagent supplies.

Contamination control is successful. High-purity metal is produced consistently with good yield.

9. Preparation and Reprocessing of Plutonium-Aluminum Alloy by the Cryolite Process - W. L. Lyon, GE-HAPO

The studies described in this paper have been carried out in support of the Plutonium Recycle Program wherein plutonium-aluminum alloy is being considered as a fuel material for the PRTR. Ways are being sought to prepare and reprocess this alloy simply and inexpensively.

The Preparation of Plutonium-Aluminum Alloys. The usual metallurgical practice for the preparation of alloys is to introduce the alloying constituents as metals to the solvent melt. In the case of aluminum alloys it is often possible to use compounds such as oxides or halides which are reduced to the metallic state by aluminum. This technique has been used by Saller[1] in the preparation of aluminum-uranium alloys and by Runnalls[2] in the preparation of aluminum-plutonium alloys.

Plutonium oxide is reduced by aluminum according to

$$3\ PuO_2 + 4\ Al \longrightarrow 3\ Pu + 2\ Al_2O_3.$$

For this reaction a driving force may be calculated from thermodynamic data:

$$\Delta F_{1000\ K} = -42\ \text{kcal}.$$

In practice, use is made of cryolite, Na_3AlF_6, as a fluxing agent to slag off the aluminum oxide. At the same time, a large excess of aluminum is present to dissolve the plutonium metal formed. These factors, combined with the favorable free energy change for the reaction, result in the reaction proceeding essentially to completion.

An acceptable procedure for preparation of plutonium-aluminum alloys consists in combining the desired weights of reactants, aluminum metal, plutonium oxide and cryolite, in a graphite crucible, and heating to 1050 to 1100 C for a period of 1/2 hour. The quantity of cryolite should be sufficient so that the Al_2O_3 formed does not exceed 20 weight percent in the salt phase when the reaction is complete. (Otherwise, the salt layer becomes viscous and a low yield may result.) Alloys containing up to 50 weight percent plutonium have been prepared in this manner. Plutonium yields in the range of 99.5 to 99.9 percent and averaging 99.8 percent were obtained during preparation of 20 to 200-gram batches of 1.5 to 15 weight percent alloys.

The process may be carried out on a semi-continuous basis by keeping the crucible continuously at a temperature above 1000 C, and pouring the metal and slag into separate molds at intervals, followed by recharging with reactants. Using a one-liter capacity tilting induction furnace for top pouring of the melt, a production rate of one kilogram per hour of 8.25 weight percent master alloy was demonstrated.

A cryolite furnace having a working capacity for 6250 grams of 15 weight percent plutonium-aluminum alloy has been designed for use in a plutonium fabrication pilot plant. This facility will be used in the preparation and fabrication of plutonium fuel elements for the Plutonium Recycle Test Reactor.

Reconstitution of Irradiated Plutonium-Aluminum Alloy. It is probable that the principal objective of reprocessing of a plutonium fuel will be the replacement of fissionable plutonium which has been burned out of the fuel material. In the case of plutonium-aluminum alloy fuel, this may conceivably be accomplished, either by addition of new plutonium, or by the removal of aluminum, so that the concentration of fissionable plutonium is again at the desired level.

Perhaps the most simple and direct method for enriching plutonium-aluminum alloy which has undergone depletion in a fuel cycle would be to add new plutonium in the form of plutonium oxide using the cryolite process. Conceivably, either the irradiated alloy could be processed in the cryolite step, or a master alloy could be produced for enriching the fuel in a remelting operation. There may be some merit in contacting the irradiated alloy with cryolite since a degree of fission product removal is thereby accomplished.

Several processes have been studied for the selective removal of aluminum from recycled plutonium-aluminum alloy fuel material. Such processes may be especially applicable to plutonium fuel utilization schemes in which it is desired to segregate plutonium on the basis of isotopic composition.

One process for the selective removal of aluminum from plutonium-aluminum alloy is to take advantage of the volatility of aluminum subfluoride. When metallic aluminum and its trifluoride are heated together in vacuo above 660 C, a white sublimate forms whose composition corresponds to that of AlF, but which has been identified as only Al and AlF_3. It has been concluded by Klemm and Voss[3] that aluminum subfluoride probably forms and exists in the vapor phase according to

$$2\ Al + AlF_3 \rightleftharpoons 3\ AlF$$

within a certain temperature range.

In order to explore the utility of this reaction for concentrating an alloy, several experiments were performed in which uranium-aluminum, or plutonium-aluminum alloys were heated in vacuo with cryolite, aluminum trifluoride, and admixtures of these fluoride salts. Removal of aluminum has been evidenced by losses in weight and by analysis of the metal charge. When the reaction occurred between two molten phases, it was found to be very rapid, often expelling the contents from the crucible. On the other hand, the reaction between molten aluminum alloy and solid AlF_3 at 800 to 1000 C proceeded smoothly and could be controlled.

The removal of aluminum as volatile aluminum trichloride offers another possible way of recovering plutonium from the aluminum alloy. The use of chlorine to volatize $AlCl_3$ from aluminum-uranium alloy at 340 C has been reported.(4)

The reaction between molten zinc chloride and plutonium-aluminum alloy has been demonstrated to proceed smoothly at 450 to 500 C with the formation of $AlCl_3$ sublimate.

Another process explored consists in the oxidation of some of the aluminum present in plutonium-aluminum alloy. If the alloy is partially oxidized by exposure to air at 900 C, and then further heated with molten cryolite, the aluminum oxide produced is dissolved away. Any plutonium oxide formed is immediately reduced by the remaining aluminum. Thus, the alloy becomes impoverished only in aluminum.

In order to achieve the selective oxidation of aluminum without this extravagant use of cryolite, the process may be accomplished electrolytically much in the manner of aluminum refining. The alloy to be enriched is made the anode of a cell, using molten cryolite as the electrolyte, and aluminum as the cathode. Experiments indicate that aluminum is removed from the anode and deposited at the cathode with apparently no significant transfer of the heavy metal.

Aluminum is a powerful reducing agent for oxides of all but the most active metals owing to the marked thermodynamic stability of aluminum oxide. Thus, when uranium or plutonium oxide containing also oxides of the fission elements is introduced into the aluminum-cryolite system, it may be expected that the noble and less active fission elements will be found in the metal phase. Similarly, if irradiated plutonium-aluminum alloy fuel material is contacted with cryolite in the presence of air, most of the fission element activities present should remain in the reduced state - except, however, that activities due to certain alkaline earths should tend to be oxidized and distribute into the salt phase. Also, volatile activities should be released during the process.

Experiments were performed wherein irradiated UO_2 was heated with aluminum and cryolite to 1050-1100 C, and the system equilibrated for 30 minutes in this range. The resulting alloy was sampled and the fission element activities were chemically separated and counted. As expected, the rare earth activities were largely present in the alloy, while alkali and alkaline earth element activities were not. Cs-Ba-137 and Sr-90 were each removed by a factor of at least 10^3. The fact that cesium is removed is somewhat surprising since Cs_2O should be readily reduced by Al. It may be postulated, however, that CsF becomes the stable form by reaction with the fluoride salt. Decontamination factors on the order of ten were observed for Ru-Rh-106 and Zr-Nb-95 activities, due possibly to the volatilization of the oxide or fluoride species.

References

(1) Saller, H.A., Preparation and Properties of Aluminum-Uranium Alloys, Journal of Metallurgy and Ceramics, 1:9 (1948).

References (cont)

(2) Runnalls, O.J.C., The Preparation of Plutonium-Aluminum Alloys, Reactor Science and Technology, Vol. 3, No. 3 (1953).

(3) Klemm, W. and Voss, E., Z. Anorg. Chem., 251, 233 (1943).

(4) Blanco, R.E., Processing of Power Reactor Fuels, Nuclear Science and Engineering 1, 412 (1956).

10. Plutonium Trichloride Process Development - H. H. Hopkins, Jr., GE-HAPO

The 234-5 Development Operation has undertaken to develop a plutonium trichloride process for the purpose of using plutonium trichloride as the reducible compound in the metal preparation process. Use of plutonium trichloride instead of the tetrafluoride will greatly reduce neutron exposure problems encountered in the present operation. This neutron source is overwhelmingly due to the alpha-n reaction on fluoride, which does not occur with chloride.

Early work on the preparation of plutonium trichloride was conducted at Chicago and at Los Alamos. It was shown that plutonium oxalate could be chlorinated by a mixture of hydrogen and hydrogen chloride at 700 C. Likewise, the oxalate or the dioxide could be chlorinated by carbon tetrachloride at a temperature of 800 C. HAPO workers several years ago showed that carbon tetrachloride and phosgene could be used to chlorinate plutonium dioxide at a much lower temperature, in the range 350 - 475 C and with reasonable time cycles, two to four hours.

Early work on the reduction of plutonium trichloride to plutonium metal indicated that the reaction would go to completion. Kennedy used a 25 percent excess of calcium and heated to 975 C. Tolley performed successful bomb reductions using an iodine booster.

Current work at HAPO on the preparation of plutonium trichloride is emphasizing the use of carbon tetrachloride and plutonium dioxide. At 450 C, good conversions are obtained in 2-1/2 hours for a powder depth of 0.7 cm. Further work will study the use of phosgene and the use of a vibrating tube reactor for carrying out this step.

The hygroscopic properties of plutonium trichloride present special handling problems. Thus, the powder may not be permitted to pick up sufficient water to cause pressurization in the mixture or bomb. The weight gain of powders exposed to air of various dew points has been studied. As an example, powder of one-inch depth can be exposed to air of dew point -13 C for 10 hours before a weight gain of 0.1 percent is observed. Similar data indicate that a well-dehumidified atmosphere will be required and that the powder must probably be kept in covered containers.

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Recent work on the reduction of plutonium trichloride to metal has been carried out on a 30-gram scale. Reduction yields are favorable and only a moderate sticking problem has been found to date. Again, an iodine booster is being employed. Product quality appears satisfactory.

The recovery of plutonium from slag, crucible, and other residues will require a preliminary chloride removal step. At present, a nitric acid boil-down treatment is visualized similar to that used in the Darex process.

Materials of construction are being investigated since chloride corrosion occurs readily in stainless steel equipment. A number of materials are promising. For the chlorinator, glass-lined steel, or platinum are possibilities. For the off-gas system, Hastelloy F, Teflon, or PVC are possibilities. For the gas supply, Monel will be used. For the chloride removal system, glass, titanium, or tantalum are planned.

The chemistry of a chloride-based process is now firmly established. However, process variables and operating conditions are not yet completely worked out. Technology must insure process safety and corrosion resistance of existing hood facilities. The reduction in neutron exposure will easily be worth the development effort.

IV. FUEL PREPARATION AND IRRADIATION SESSION

Chairman: O. J. Wick, GE-HAPO

1. Plutonium Metallurgy - F. W. Schonfield, LASL

The purpose of this talk is to define the metallurgical characteristics of plutonium and plutonium alloys, in a general manner, for the information of those interested in the use of plutonium as a reactor fuel. It is emphasized that the state of knowledge of plutonium physical metallurgy is such that much of the information presented must be considered to be tentative and subject to change.

The suitability of any fuel for a given reactor application is determined by its ability to meet the economic, nuclear, chemical, and metallurgical requirements of the application. Considering the metallurgical requirements, and only those requirements that are usually present no matter what the reactor system may be, one can say that a metallic fuel should exhibit:

(1) a positive coefficient of thermal expansion;

(2) no discontinuous volume changes between the minimum and maximum operating temperatures;

(3) sufficiently high thermal conductivity to dissipate the generated heat without having undesirably high temperature gradients;

(4) a sufficient degree of compatibility with the container material to permit fuel maintenance separate from the rest of the system (e.g., mechanical and chemical interaction should be of such small magnitude that the identities of fuel and container can be maintained, and the fuel must not migrate from its proper place as by mechanisms such as diffusion or effusion);

(5) a reasonable degree of resistance to radiation damage; and

(6) resistance to permanent deformation during thermal cycling.

These requirements alone seem to remove unalloyed plutonium from the classification of desirable fuels. Therefore, one must investigate alloy systems.

Data concerning the interactions of plutonium with other metals are sufficient to delineate a few of the general characteristics of its alloying behavior. Plutonium seems to be completely immiscible, in both liquid and solid states, with the alkali metals (data are available only for lithium, sodium, and potassium); and to show liquid miscibility gaps of different widths with the alkaline-earth metals (although not shown in the Pu-Be diagram, the existence of a small miscibility gap is suspected; the Pu-Mg system has a large gap, and calcium, stontium and barium are believed to be completely immiscible with plutonium in both liquid and solid states).

The behaviors of scandium and yttrium toward plutonium are not known. Lanthanum forms no compounds with plutonium and the system has a wide liquid miscibility gap, but cerium, praseodymium and neodymium are completely miscible with plutonium in the liquid state and show significant solid solubilities, but compounds are not formed. The actinide elements thorium and uranium show complete liquid miscibilities, partial solid miscibilities, and do form intermediate phases with plutonium.

The characteristics of the binary systems involving titanium, zirconium, and hafnium are not well established. It is known that all show complete miscibilities in the liquid states and appreciable solid solubilities; and at least one compound is known to be formed with zirconium.

The refractory metals vanadium, chromium, niobium, molybdenum, tantalum, and tungsten have complete liquid miscibilities but essentially zero solid miscibilities.

Manganese and the Group VIII elements (in the cases where reactions are known) possess the capacity to lower markedly the melting point of plutonium. Only one compound is formed in the Pu-Mn system, whereas the Group VIII metals form greater (and variable) numbers of compounds.

The binary systems with copper, silver, and gold are not well established. It is known that a liquid-miscibility gap exists in the Pu-Ag system and that all three elements form several compounds with plutonium. Other B-subgroup metals tend to form, with plutonium, intermediate phases having comparatively high melting points. All of these systems show complete

liquid miscibilities and a number of them are characterized by solid solubilities sufficient to retain the delta-phase of plutonium to room temperature.

In general, while the undesirable thermal expansion characteristics of solid "pure" plutonium, and its discontinuous volume changes, can be easily eliminated by small alloy additions, increases of conductivity and decreases of reactivity are probably most easily attained in solid solutions containing relatively small amounts of plutonium; and resistance to radiation and thermal cycling damage may be obtained by selecting alloys of high crystallographic symmetry.

Insofar as solid fuels for high-temperature operation are concerned, the alloys of plutonium with zirconium, thorium, and uranium are of interest, as are many intermediate phases. The oxides, carbides, and nitrides possess many potential advantages and should also be considered for use. If the operating temperatures can be decreased to below about 500 C, then the field of possible solid fuels is extended to include the stabilized delta-phase alloys.

It appears at present that the binary alloys of plutonium with the Group VIII metals will form the bases for liquid fuels. Considerable variation may be introduced through use of three, four, or more components, but such alloys will probably be based on the low melting eutectics typified by the iron, nickel, and cobalt systems.

2. Physical Metallurgy of Plutonium Fuel Alloys - R. J. Dunworth and L. R. Kelman, ANL

At ANL the physical metallurgy of plutonium fuel alloys is carried out in four programs: (1) the U-Pu-Fs alloy program, (2) the high purity Pu program, (3) the U-Pu alloy program, and (4) the phase diagrams of Pu and Fs elements.

The U-Pu-Fs Alloy Program. The early U-Pu alloys were brittle, pyrophoric, and susceptible to irradiation damage. Alloys of U-Pu-Mo and U-Pu-Fs, however, have excellent resistance to irradiation damage and air corrosion, less than 0.5% thermal cycling growth, a thermal expansion coefficient of $18(10^{-6})/^{o}C$ between 200 C and 500 C and about 50% of the thermal conductivity of unalloyed uranium. These alloys have been top poured, injection cast, and extruded successfully. Both Mo and Fs tend to stabilize B.C.C. gamma U and make an isotropic U-Pu alloy. The reference alloy is 70 w/o U - 20 w/o Pu - 10 w/o Fs. The Fs contains 0.5 w/o Zr, 2.6 w/o Mo, 4 w/o Ru, 0.6 w/o Rh and 2.3 w/o Pd.

Several problems with these alloys have arisen: (1) recent pins were severely damaged by irradiation tests; (2) the oxygen content of the alloy is quite high - between 65 and 150 ppm O_2; (3) the thermal expansion curves showed two contractions (at 500 C and 700 C) and two breaks (at 520 C and 600 C); (4) a metallographic examination of the injection cast pins showed dendrites, segregation and porosity. The reactions are mostly sluggish. The present work involves metallographic and x-ray studies of alloys after various heat treatments. The properties of certain alloy structures will be determined.

The High Purity Plutonium Program. High purity plutonium will be made by electrolytic deposition from a fused salt bath of LiCl, KCl, and PuF_4. A metallographic study of different grades of unalloyed Pu has been started. The common impurities in high purity reduction buttons from Los Alamos total about 200 ppm and in regular plutonium between 1000 and 2000 ppm.

The U-Pu Alloy Program. The addition of $12^{w}/o$ Pu to U expands the lattice. The a_o and c_o parameters increase 0.14% but the b_o parameter increases 1.2%. A preferred orientation study of U and $1^{w}/o$, $3^{w}/o$, $5^{w}/o$ and $10^{w}/o$ Pu has been started. A 20% reduction in area has been produced in the rods by rolling at 450 C.

Phase Diagrams of Pu and Fs Elements. The binary phase diagrams of plutonium and each of the major fissium elements Zr, Mo, Ru, Rh, and Pd will be roughed in. The work on Pu-Zr has just started and some thermal arrests have been obtained.

3. Radiation Damage to U-Pu Alloys - J. H. Kittel, ANL

The first irradiation experiments on uranium-plutonium alloys were on small nickel-plated pins with the compositions U - 10 w/o Pu and U - 15 w/o Pu. The pins showed little damage after burnups ranging up to 0.35 a/o, although the amount of restraint contributed by the nickel cladding was not known.

The next irradiations were made on large specimens which were not clad before irradiation. The larger specimens included castings of uranium containing 3.7, 6.6, and 13.0 w/o plutonium, and extruded specimens containing 9.5, 14.1, and 18.7 w/o plutonium. It was found that the cast specimens were badly damaged from severe surface roughening by burnups of only a few tenths a/o, apparently because of excessively large grain sizes. The extruded specimens maintained smooth surfaces under irradiation but developed large elongations, which were dependent upon plutonium content. For example, in samples with 0.4 a/o burnup, a 14.1 w/o plutonium alloy specimen elongated 96%, whereas an 18.7 w/o plutonium alloy specimen elongated only 8.4%.

Irradiations were also made on cast specimens of the alloy uranium - 20 w/o plutonium - 5 w/o molybdenum. Part of the specimens were given a heat treatment after casting which consisted of holding them at 560 C for 25 minutes followed by furnace cooling to room temperature. The uranium-plutonium-molybdenum specimens were found to show excellent dimensional and surface stabilities under irradiation. Information on performance at elevated temperatures was not obtained.

More recently, attention has been directed to uranium-plutonium-"fissium" alloys. Fissium is the term applied to a group of alloying elements which will be present in the fuel alloy used in EBR-II because of the pyrometallurgical reprocessing scheme which will be used. It is anticipated that the composition of the fuel alloy used in the second loading of EBR-II will have approximately the composition uranium - 20 w/o plutonium - 10 w/o fissium. The fissium alloys are characterized by sluggish transformations, so

that if sufficient fissium is present, the gamma phase is present even with slow cooling.

Uranium - 20 w/o plutonium alloys containing additions of 5.4 and 10.8 w/o fissium have been found to be very stable under irradiation provided irradiation temperatures do not exceed the swelling temperature. Sufficient experimental data are not yet available to define this temperature, but it appears to be in the range of 500 to 600 C. Specimens irradiated above the swelling temperature have been observed to show volume increases ranging up to 70%. Swelled uranium - 20 w/o plutonium - 10 w/o fissium specimens are highly pyrophoric in air and have been observed to spontaneously explode or ignite and burn to oxide.

4. Plutonium Fuel Fabrication and Technology at Argonne National Laboratory - A. B. Shuck, ANL

Much of the emphasis of research and development on fuels will be shifted from uranium to plutonium at Argonne National Laboratory. Three facilities are under construction or being planned to facilitate the work. These are the Fuel Fabrication Facility for development of plutonium fuel manufacturing methods; the Fuels Technology Center for general metallurgical and engineering research on reactor materials including plutonium fuels and uranium-233; and the EBR-II Fuel Process Plant, to reprocess and refabricate irradiated EBR-II fuel.

The Fuel Fabrication Facility building is completed and installation of hoods, process equipment and services is under way. This facility was designed for a maximum of versatility in the development of reactor fuel elements. The process equipment is completely hooded with provision for inert gas atmosphere operation of the equipment lines.

The Fuel Technology Center is under design and with construction to begin this fiscal year. It will replace temporary metallurgy facilities at Argonne National Laboratory but will be designed so that major emphasis may be shifted to research on plutonium, uranium-233, and irradiated reactor fuels. Provision will be made for the addition of future additions for general engineering metallurgy.

The EBR-II Fuel Process Plant to be built at Arco, Idaho, is designed specifically to demonstrate the possibility of pyrometallurgical processing, refabrication, and reassembly of the highly radioactive irradiated fuels from the EBR-II. It will be an all remote control, shielded, sealed and inert atmosphere ventilated plant equipped for the remote manufacture of the EBR-II fuel elements. A measure of flexibility has been achieved by design of componentized equipment, the components of which are replaceable by remote control. This plant is scheduled for operation in 1960.

5. Fabrication of Plutonium Discs (Film) - W. D. McNeese, J. W. Anderson, J. D. Carter, B. N. Robbins, J. W. Romero and T. K. Seaman, LASL

Twenty-five plutonium discs of a fixed density and close dimensional tolerances were required for criticality measurements for the Los Alamos Molten Plutonium Reactor Experiment (LAMPRE). These discs were to 0.125 inch thick, 5.9370 inches in diameter, and have a density of 16.47 g/cc. They were to be handled and tested in equipment and laboratories used for other than plutonium work; therefore, it was necessary that they be in tight alpha-containing cans.

Two methods were used for making these discs: casting and direct machining, and blanking from sheet stock made by extrusion.

In the machining method, an oversize disc was cast in an alumina-coated graphite crucible in a vacuum furnace. This casting was machined to dimensions in a lathe.

In the blanking method, a cast feed ring was rough machined to weight and then extruded into a tube of the proper wall thickness in a specially designed direct extrusion die assembly. This tube was split and flattened by hand. Discs were cut from the roughly flattened sheet with a blanking die in a slow movement hydraulic press. The discs were then flattened and pressed to exact size in a coining die.

The discs were canned in 0.005 inch thick nickel cans which had been formed by blanking and drawing. The discs were carefully assembled in the cans and the edges sealed with a lead-tin solder.

6. Fabrication and Irradiation of Plutonium Fuels - M. D. Freshley, GE-HAPO

One of the key objectives of the Plutonium Recycle Program at Hanford is the development of desirable plutonium-bearing fuel elements that can be fabricated safely and at low cost. Elements containing plutonium for spike enrichment use and plutonium-uranium elements for uniformly enriched reactor loadings are both of interest.

Fuel Element Designs. At present, more fuel fabrication and irradiation experience has been obtained with Al-Pu alloys than with any other Pu-containing fuel material and alloys of Al with Pu will be the first to be evaluated as spike enrichment for the PRTR. Al-Pu alloys minimize the Pu contamination problem, are relatively easy to fabricate, and the fuel material is an ideal matrix, or dispersion, type which has demonstrated in-reactor stability.

In the PRTR irradiations of Pu-Al, a 19-rod cluster geometry will be employed. Rods will be one-half inch in diameter and about seven feet long and clad in Zircaloy-2. The fuel composition for the initial PRTR loading will be Al - 1.8 w/o Pu and thereafter plutonium higher in Pu-240, 241,

and 242 will be used, requiring higher total plutonium contents in the alloy to maintain equivalent reactivity.

A number of other fuel designs and compositions will be investigated for possible spike enrichment use. Oxide mixtures of interest are mixtures of PuO_2 or PuO_2-UO_2 mixed crystal with diluents such as Al_2O_3, MgO, SiO_2, BeO, or ThO. Impregnation of porous inert carrier materials with PuO_2 may be a simple way of obtaining Pu-bearing fuel cores for spike enrichment. Several types of carrier materials are available, one of which is graphite. A somewhat different spike element concept is the "self-shielded" fuel element. In this design plutonium is utilized in the concentrated metal form taking advantage of the high thermal neutron absorption cross-section of Pu to limit the fissioning of the Pu to the external surface of the fuel core. This type of element permits longer residence time in the reactor for a given change in reactivity and reduces the rate of fuel element throughput. The disadvantages are the high heat transfer rates involved and the high Pu inventory.

After about the first two years of PRTR operation emphasis will change from spike systems to uniformly enriched loadings. UO_2-PuO_2 mixtures, in concentric tubular and rod cluster geometries, fabricated in various ways will then be the subject of intensive study. Metal alloy systems containing uranium and plutonium will also receive attention.

Fabrication Development. Whether or not a particular type of fuel element is economical depends to a large extent upon the method of fabrication. Because of the toxicity of Pu and the restrictions that are thus imposed upon its handling, the incentive and the need is great to develop cheap, simple, and reliable fuel element fabrication techniques.

Al-Pu alloys have been fabricated, starting with plutonium metal, by melting, casting and machining, but this is an expensive procedure. Several simplified fabrication and reprocessing schemes are under study in an attempt to minimize costs. Hanford studies have confirmed that the cost of preparing Al-Pu alloy material by directly reducing PuO_2 with aluminum under a cryolite flux is substantially less than by conversion of oxide to fluoride, bomb reduction and alloy melting. Further cost reduction appears possible by casting Al-Pu alloy core material directly into a jacket using air or mechanical pressure, a potentially inexpensive technique easily adapted to remote operation. Satisfactory Al castings up to eight feet long with densities up to 96 percent of theoretical have been obtained in 1/2-inch diameter jacketing tubes. Also, extrusion of Al-Pu fuel elements has certain advantages in respect to the amount of handling required and, thus, fuel element fabrication costs.

With ceramic core fuel elements emphasis has also been on the development of simplified fabrication techniques. Because of the similarity between PuO_2 and UO_2 and because the UO_2 will be diluted with PuO_2 in such small amounts, UO_2 has been used wherever possible to develop the process under study.

UO_2 has been formed into fuel element shapes by pressing, extrusion or slip casting of a green compact followed by sintering in hydrogen to yield final densities as high as 98% of theoretical and then grinding to final size.

Several fabrication methods yielding unsintered cores are under study. In one of these, powder is vibrated or mechanically tamped into the cladding tube to intermediate densities. In another, powder is packed to an intermediate density, the element is sealed off, and pressure up to ca. 50,000 psi is applied in an isostatic press to yield core densities over 70 percent of theoretical. Still another method under study is the swaging of a metal tube containing the ceramic fuel material. Densities greater than 90 percent of theoretical have been obtained.

Irradiation Testing. The irradiation program at Hanford is just commencing. To date, aluminum alloys containing about 1.7 w/o Pu have been irradiated to as much as 50 percent of burnup of the Pu atoms. These 1/2-inch diameter samples were clad in Zircaloy-2 and operated with a central core temperature of 400 C. A volume increase of about 1.4 percent was observed. Many more irradiation tests of Al-Pu and other Pu-containing fuel materials are being planned.

Irradiation of UO_2 core materials fabricated by the methods which were described have been performed under a variety of test conditions. Fuel elements containing unsintered UO_2 powder enriched to three percent U-235 were irradiation tested at power levels sufficiently high to cause extensive sintering, relocation, and fusion of the core. It appears that central core temperatures higher than previously believed may be tolerated. An aluminum clad tubular fuel element containing enriched UO_2 powder compacted by isostatic pressing has been successfully irradiation tested.

Capsules containing UO_2 compacted by swaging to about 90 percent of theoretical in Zircaloy tubing have been irradiated to relatively low exposures without incident.

7. Fabrication of MTR Plutonium Fuel Loading - T. C. Nelson, GE-HAPO

Late in 1957 Hanford was contacted in regard to producing a plutonium fuel loading for the MTR reactor located at Arco, Idaho. At the time of these earlier discussions it was contemplated that Hanford would cast and form the fuel into suitable cores, assemble and weld into a convenient sandwich. Subsequent discussions enlarged the scope of the original job to also include the rolling and sizing operation. Early in 1958 the job was further enlarged to include the complete fabrication of fuel assemblies ready for insertion into the reactor.

Briefly, the fabrication process is as follows:

Plutonium of suitable purity is alloyed with aluminum in a resistance type melting furnace in a glove box enclosure. The molten metal is cast into graphite molds. These castings are subsequently cut into short sections or billets. These billets are press forged to the desired shape in a set of hot dies. All of this equipment is located within glove box enclosures. The forged billets are then examined by radiography for segregation or internal flaws. The acceptable pieces are cleaned utilizing ultrasonic techniques until they are relatively contamination free.

All of the aluminum components such as the core frame, picture frame, cover plates and evacuation tubes are cut from 2S aluminum sheet, given a blister test, cleaned, degreased, and sent to the assembly operation. The sandwiches are then assembled with the plutonium-aluminum core placed in the center. These components are held together with clamps until the edges are welded. Three edges of the sandwich are welded in a consumable electrode automatic welding machine. The time cycle for each weld is about nine seconds. The remaining edge through which the evacuation tube is introduced is, by necessity, hand welded. The assembly then undergoes a helium leak check on the mass spectrometer. The next operation is a hot out-gas to remove all air and undesirable gases from the assembly. This is accomplished by placing twenty-four of the welded components in the furnace operating at 520 C at a vacuum less than 0.1 micron Hg for a minimum of ten to twelve hours. The tubes are then sealed by pinching with a modified bolt cutter and welding the end.

These out-gassed assemblies are then placed in a furnace operating at 590 C for a minimum of three hours prior to rolling. They are hot rolled from the 3/4" original thickness down to approximately 0.070" in six passes. This amounts to about 30% reduction in thickness per pass. The rolled out plates are then rough sheared to a convenient length for a flux annealing operation. This operation consists of painting the surfaces of the plate with a water mixture of eutectic 190 flux. A split temperature and time cycle is used to reduce the time at high temperatures. The current practice is 1/2 hour at 530 C followed by 1/2 hour at 600 C.

After the flux annealing operation the plates are cleaned and inspected for any surface flaws such as blisters, pits, or obnoxious surface blemishes. The plates that have not been rejected are then reduced to the final thickness of 0.060" by cold rolling. They are then examined for surface blemishes such as pits, scratches or any undesirable flaws.

The plates are then examined by radiography to determine the core location and segregation in the core area. The plates are then indexed and milled to size. A second x-ray is then taken to determine the edge dimension, end clearances, and core thickening or "dogboning".

The next operation is the blister test where the plates are subjected to 500 C for one-half hour followed by 600 C for an additional one-half hour. The plates are then given a very close examination for blisters or any physical defects and are again checked for the proper dimensions.

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The plates are then curved in a set of curving dies, cleaned, and prepared for assembly in the brazing operation.

The location of any plate in the assembly is determined by its plutonium content. It is desirable that plates with higher than normal concentration are placed in the center of the assembly where the flow of cooling water is maximum.

The fuel plates and side plates are coated with a water diluted brazing flux (currently ALCOA 33). They are assembled in a steel jig and transferred to a ceramic jig for insertion into the furnace. Current practice is to place the unbrazed assembly into a furnace which is operating at 604 C for one and one-half to two hours. After the brazing operation, the assembly is cleaned, inspected for proper plate spacing, curvature of plates, width, length and all other necessary dimensions. The brazed assemblies are also checked for surface contamination by sensitive alpha counters.

The end hardware is then joined to the brazed assembly, held in position in a set of jigs and welded. The welded assembly is machined to the final dimensions. The final rod is then subjected to a series of dimensional inspections and if acceptable is prepared for shipment.

During the development of the current process, several operational obstacles had to be overcome. One of the major defects which became evident early in the program was the problem of plutonium diffusing through the aluminum clad. After a short investigation it was found that the time at or near 600 C seriously affected the diffusion rate. The time at high temperature or the operating temperature was lowered from the original specified temperatures in many of the operations. Preliminary investigations were made to determine what the diffusion of plutonium in aluminum might be.

During the fabrication of these earlier elements it was not recognized that the problem of core-end thickening was serious. When the temperatures of various processes were lowered, however, this thickening or "dogboning" became extremely apparent. This was the reason for a long investigation into ways to eliminate dogboning. It was felt that removal of core material near the corners of the core would result in less thickening at the ends of the resultant rolled plates. This, however, proved to not always be the case. Other people, experienced in this type of work, were consulted and their suggested techniques were tried. Several hundred cores were made with various angles, edge sizes and core lengths. It was found that whenever there was a sharp angle or change in direction, which formed a sharp edge in the core, it resulted in dogboning. It should be pointed out also that it was desirable to limit the maximum allowable dogboning to less than 5% of the core or 0.001". The core configuration which looks most acceptable has a rather short taper with all sharp corners removed by adequate blending or tapering. This shape is not much unlike a parabola. New dies to produce this shape are being fabricated and installed in the press. A short series of plates will be run through this operation, rolled, and evaluated prior to starting the full fledged production program.

V. PLUTONIUM HANDLING PROBLEMS SESSION

Chairman: R. D. Baker, LASL

1. Radiation and Contamination Control in a Hanford Plutonium Production Area - G. E. Backman, GE-HAPO

The process equipment used in this facility is housed in stainless steel hoods with lucite windows having glove ports for arm length rubber gloves, sphincter ports and airlocks for entry of equipment into the hood and plastic ports of varying sizes for entry for removal of equipment. Hoods are connected together when feasible and materials transported with some remotely controlled conveyors of various types. Contamination control during routine operation is achieved by having reasonably tight hoods and a constant vacuum of approximately one inch of water on each hood.

Potential exists for more serious contamination spreads during removal and replacement of process equipment. The use of plastic has become exceedingly important in the use of contamination control. Nearly all equipment is removed through plastic bag ports. It is placed in plastic bag and then a seal made on the bag between the equipment and the hood. The greenhouse technique involves building a small room with plastic around equipment to be moved. This method is employed when a serious potential exists of the contamination spread. When the potential becomes even more extreme a "plastic man" is used inside the greenhouse. The "man" has a plastic tunnel attached and through it personnel enter or leave the suit. By keeping positive pressure on the suit very little contamination is encountered when tears or cuts occur.

The radiation exposure problem can be divided into hand dose and whole body penetrating dose. Surface dose rates on plutonium contribute primarily to hand dose and have been rather difficult to determine. At one time it was considered to be 200 mr/hr, later this was revised to 800 mr/hr, and then again revised to 1600 mr/hr after a reduction step and 1200 mr/hr after it would be remelted. These are the dose rates that are being used presently; however, they could be somewhat inaccurate because exposure does increase with time after decontamination and depends on the amount of the different plutonium isotopes encountered.

The breakdown of the effective energies of the surface dose rate shows that 75% is approximately 17.5 kev, 12% is approximately 60 kev, and 13% is equal to or greater than 200 kev. These energies are mentioned to show that a small amount of shielding can reduce the dose rate significantly.

Body exposure has been a serious problem also. At present personnel receive 40 to 50 mr/kilogram of plutonium processed from the nitrate to the metal. A breakdown of the energies of the radiation dose rate received shows that approximately 15% is due to 17.5 kev, 10% due to 60 kev, and 75% due to 200 kev or greater.

Shielding has been employed in attempts to reduce exposure. Hood gloves with a 10-mil overlay of zinc impregnated rubber have been shown to reduce hand exposure by 50%, and recently hood gloves containing 10 mils of lead impregnated rubber have been employed and appear to reduce exposure by 60 or 65%.

Body exposure comes from neutrons that are primarily the result of an alpha, n reaction occurring in one of the plutonium compounds and gamma radiation from plutonium isotopes, daughter products, and fission products. The neutrons have an effective energy of approximately 1.4 mev and shielding is rather difficult since it takes about six inches of a high hydrogen containing material to reduce the neutron exposure by a factor of 10. Water walls, masonite and lucite are materials that are most frequently employed. Safety glass has been used to remove low energy x-rays; however, it appears that it will be necessary to resort to lead glass to minimize the exposure problem properly. It will also be necessary to use lead or thicker steel in non-transparent portions of the hoods.

The original design of equipment is most important in having both contamination and radiation control. A few items that I suggest be considered when any new plutonium handling facility is designed are:

1. Design hoods so that they are easily decontaminated, both inside and out.

2. Design hoods so that plutonium is kept near the center whenever practical, particularly when it is in storage. (The reason for this is to prevent the neutron exposure from spontaneous fission of the Pu-240 isotope from being a serious shielding problem.)

3. Investigate the possibility of remote operation for each section of the operation.

4. If possible, design equipment so that it can be removed from areas of high radiation for repair.

5. All wiring, piping, etc., should enter hood in as few locations as possible and should be enclosed in easily decontaminated conduit (as much equipment as possible on one large conduit).

6. All motors, pumps, etc., located near process hoods should be compartmentalized in containers that are easily decontaminated.

7. Shielding should be considered for all hoods primarily for gamma radiation.

8. Shielding of the plutonium-containing process equipment should be investigated.

One point to remember is that regardless of how the process is constructed a most important item in maintaining control is to expect all employees to continually be aware of their responsibility in this direction. Without this type of emphasis on the problem, a control program will not be successful.

2. Dosage from High Exposure Plutonium - W. C. Roesch, GE-HAPO

When plutonium isotopes decay, a small part of their energy is given off as X and gamma rays. These together with gamma rays and neutrons from spontaneous fission produce a significant dose rate at the surface of plutonium metal or compounds. This rather than the alpha emission is responsible for the external radiation hazard from plutonium. The dose rate depends on the relative amounts of the different plutonium isotopes that are present. This report describes estimates of the dose rate to be expected from the plutonium isotopes in the metal and compounds, an example of high exposure plutonium, and related problems.

The dose rate due to an arbitrary energy gamma emitter in plutonium metal was calculated by allowing for exponential absorption in the metal, neglecting scattering in the metal, making an empirical correction for backscattering from tissue in contact with the metal, and allowing for the production of fluorescence radiation. Then the decay schemes of the plutonium isotopes and their daughters were examined for gamma rays, x-rays following internal conversion, and gamma rays from spontaneous fission. The dose rate at the surface of the individual pure isotopes when in radioactive equilibrium was calculated; the results are in Table I. The surface dose rate of any composition of plutonium can be calculated by weighting the entries in Table I by the fractions by weight of the different isotopes and allowing for radioactive buildup.

As an example of the use of the figures in Table I, the dose rate at the surface of plutonium made by long exposure of uranium in a reactor was calculated.

Table I. Equilibrium Plutonium Surface Dose Rates

Isotope	Component	rad/hr
238	X-rays	2
	Hard	980
239	X-rays	0.61
	Hard	0.056
240	X-rays	14
	Hard	0.37
	Fission	0.25
241 (Am-241)	X-rays	237
	Hard	221
(U-237)	Hard	23

Triplett's (HW-33912) calculations of product composition for a natural uranium reactor with initial resonance escape probability 0.8 and temperature 0.05 ev were used.

The total X and gamma-ray dose rate of freshly purified metal rises from about 1 rad/hr for low reactor exposure to a little over 4 rad/hr at 10,000 MWD/T. The dose rate changes with time after purification due to buildup of daughter products. The change in the total dose rate is relatively small at low exposures but at 10,000 MWD/T the dose rate increases to 6 rad/hr in five days.

The total photon dose rate is made up of a large proportion of x-rays of about 17 kev energy that result from internal conversion of higher energy rays. These x-rays are readily absorbed in thin layers of high atomic number materials and in many circumstances can be eliminated. This leaves a hard component that is about 0.1 rad/hr for freshly purified metal of low exposure and increases slowly to about 0.2 rad/hr for 10,000 MWD/T. The hard component contains most of the dose rate from the daughter products Am-241 and U-237, resulting from the branching decay of Pu-241. These daughters cause significant changes in the hard component with time after purification. Even at low reactor exposure the hard component about doubles in a few weeks. At 10,000 MWD/T it increases to almost 2 rad/hr in five days, an increase by an order of magnitude. There is advantage in planning to process the plutonium as rapidly as possible and to return scrap to a point in the process at which uranium will be eliminated.

Neutrons from spontaneous fission add to the total dose rate at the surface of plutonium metal. For small pieces of metal, the dose rate was calculated by neglecting the absorption in the metal. The RBE dose rate at the surface of a sphere of one kilogram rises from zero at low exposure to about 0.9 rem/hr at 10,000 MWD/T. It will not change with time after purification.

Some plutonium compounds emit neutrons and/or gamma rays as a result of alpha particle bombardment of the light elements in the compound. Fluorides are particularly important in this respect. Neutrons result from an (alpha, n) reaction in the fluorine; gamma rays, from inelastic alpha scattering. The alpha rays of the different isotopes are sufficiently alike in energy that the number of these reactions is about proportional to the curies/gm of alpha activity in the plutonium. The neutron surface dose rate for low exposure is a few rem/hr; for 10,000 MWD/T it will be about double this. The total gamma-ray dose rate is about 1.5 rad/hr; about 0.5 rad/hr of this comes from the inelastic scattering and will double at high exposure.

When the dose rate at a distance from the surface is desired, it can be estimated from

$$\frac{\text{dose rate}}{\text{surface dose rate}} = \frac{\text{solid angle subtended by source}}{2\pi}$$

This is expected to be accurate for the gamma rays and approximately true for neutrons. For example, at 10,000 MWD/T, the gamma-ray dose rates of 4 (total) and 0.2 (hard) rad/hr become 0.11 and 0.006 rad/hr at 10 cm from a one-kilogram sphere and follow inverse square at greater distance.

The surface dose rate considered so far is that on a piece of plutonium infinitely thick. Practically, "infinite" here means a few mean-free paths. For the x-rays, a mean-free path is 0.004 inch; for the harder components, about 0.05 to 0.1 inch. When plutonium is distributed as contamination on hood surfaces, the x-ray dose close to the hood is usually nearly equal to that on a solid piece because even a thin layer of contamination is effectively infinitely thick. Due to the solid angle rule, above, the x-ray dose rate in or near the hood is the same (or may be almost twice as much) as on a solid piece. For hood contamination, the harder components are usually much reduced because there is not enough plutonium to give a "thick" source.

When plutonium is dispersed among lighter elements to form a compound, the actual mass dilution of the plutonium is not very great. The absorption of the x or gamma rays is decreased in nearly the same proportion because the light elements are relatively inefficient absorbers. The result is very little difference in dose rate due to rays emitted by the plutonium between plutonium and its compounds. The problem of greater dilutions such as in alloys or solutions has not been studied.

3. Design and Operation of Hanford's Plutonium Metallurgy Facilities - O. J. Wick and I. D. Thomas

A brief history of Hanford's metallurgical research and development facilities which first began operation in 1951 is presented. Since startup the laboratory has expanded considerably to keep pace with an enlarged fabrication development program. An active plutonium fuel development program at Hanford was started in 1957, and a developmental pilot plant was designed for the fabrication of a wide variety of experimental plutonium fuels. This new facility is now under construction and will be in limited operation by 1959.

The handling of plutonium in large quantities is discussed from the standpoint of additional precautions required over those needed for a research operation. Large scale fabrication development work is of such a nature that an occasional glove rupture or other incident must be expected. Design of facilities at Hanford to minimize the spread of uncontrolled contamination and to facilitate decontamination procedures include: (1) compartmentation of the laboratory area, (2) flow of laboratory air from areas of low contamination potential to those of high potential, (3) down draft air flow to floor exhausts below glove boxes and hood; (4) provisions for static inert gas or dry air atmospheres in glove boxes with adequate exhaust capacity to provide a minimum flow of 120 lineal feet per minute into hoods through openings such as ruptured gloves, air locks, etc.

The operational philosophy is given for the prevention of the accumulation of a critical mass which cannot be completely guaranteed by the design of a versatile developmental facility. Thorough training of engineers and technicians in nuclear safety coupled with operational rules must be relied upon to maintain safe handling conditions.

Radiation exposure problems in plutonium handling are discussed which are becoming more serious as more highly irradiated plutonium must be fabricated. Personnel must be shielded from the beta, gamma and x-ray dosage which is related to some of the isotopes of plutonium and their decay products. With the present material of moderate exposure, adequate protection to allow almost unlimited working time is afforded by lead glass equivalent to one eighth inch of lead. Lead impregnated neoprene gloves to maintain reasonable working times with safe hand exposures are now available. Plutonium of much higher isotopic content (20-30,000 MWD/T) will probably require remote handling techniques or automatic fabrication processes which result in little or no personnel exposure.

Design concepts for a new fuel fabrication facility are presented. The Plutonium Fuels Pilot Plant has been designed to provide full scale metal and ceramic fabricating equipment for development of plutonium-containing reactor fuels. This plant will provide experimental reactor loadings for evaluation in the Plutonium Recycle Test Reactor at Hanford. Fabrication equipment for each unit process is housed in individual cells for contamination control and the building structure is designed as a secondary containment shell to control free contamination. This facility will incorporate maximum flexibility for the development of plutonium fuels coupled with improved continuity of operation and personnel safety.

4. Plutonium Metal Fabrication Facility at Los Alamos Scientific Laboratory - P. J. Peterson, R. L. Thomas, and J. W. Anderson

The Plutonium Metal Fabrication Line at the Los Alamos Scientific Laboratory is described in this paper. Its purpose is to furnish facilities for research and development on plutonium and plutonium-alloy fabrication techniques, as well as for routine fabrication. It was designed to provide a versatile, enclosed facility for casting, machining, drawing, pressing, assembling and nickel-coating plutonium and its alloys. Provision was made for the addition of equipment which may be needed for other fabrication methods.

5. Hot Cells for Plutonium Reactor Fuel Research - P. J. Peterson, R. L. Thomas, and J. L. Green, LASL

A hot cell facility, now under construction, is to be used in support of plutonium-fueled fast reactors. Experiments will be conducted on a kilocurie scale to develop pyrometallurgical and aqueous reprocessing techniques for spent fuel.

[redacted]

The structure consists of four 6' by 6' by 10' high cells in a line backed by a common corridor 11' by 30' long. Shielding is accomplished with a combination of magnetite and ordinary concrete, the magnetite concrete being used where space considerations dictate.

One pair of Argonne Model No. 8 manipulators are used in each cell and a General Mills Mechanical Arm, in the corridor. The former will be used for actual experimentation and the latter to transfer material into and out of the cells.

Because of the types and quantity of plutonium to be handled, the material will be processed within a portable enclosure located in a cell. The enclosure is designed for versatility.

The Model No. 8 Manipulators will have access to the interior of the enclosure through contamination-tight plastic booting developed at the Los Alamos Scientific Laboratory and can be remotely introduced and withdrawn. Their function is to perform the lighter tasks required and to service the mechanical aids used for heavier work.

Services to the enclosure can be remotely disconnected and the enclosure can be remotely removed. Special services are ventilation, negative-pressure circulating water system and induction heating facility.

Plutonium is transferred to and from the enclosure in an alpha container by the Mechanical Arm. The container is attachable to the enclosure and the plutonium removed by remote operation without spread of contamination.

6. Plutonium Handling and Plant Design Problems - I. D. Venable, Dow Chemical Company

Mr. Venable described methods and equipment found best in Rocky Flats Plant experience for plutonium processing and fabrication work.

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