PLUTONIUM ABSTRACTS

PLUTONIUM ABSTRACTS is a "CURRENT AWARENESS SERVICE" TO THE ATOMIC ENERGY COMMISSION AND ITS RESEARCH AND DEVELOPMENT CONTRACTORS FOR NEW DEVELOPMENTS IN ALL AREAS OF PLUTONIUM TECHNOLOGY. MATERIAL IS COMPILED BY THE REFERENCE STAFF OF TECHNICAL INFORMATION, BATTELLE NORTHWEST.

THIS ISSUE COVERS THE FOLLOWING SUBJECT FIELDS:

Biology & Medicine
Chemistry
Health & Safety
Metallurgy
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3319. BIOLOGICAL, MEDICAL, AND ENVIRONMENTAL RESEARCH.

The following sections of this report contain information about plutonium: bone seeking radio-nuclides in animals, p. 154-60; skin studies, p. 163-65; gastrointestinal tract irradiation, p. 165-66; inhalation studies, p. 166-69; and inventory of nuclear debris, p. 191-92.

3320. IN VITRO DISTRIBUTION OF AMERICIUM IN HUMAN BLOOD SERUM PROTEINS.

The distribution of americium in blood serum proteins is compared with that of plutonium. It is concluded that americium forms weaker protein complexes than those of plutonium.

3321. METABOLISM OF TRANSURANIC ELEMENTS.
Jean-M. Debois (Univ., Louvain, Belgium)

In Dutch.


3322. PACIFIC NORTHWEST LABORATORY ANNUAL REPORT FOR 1965 IN PHYSICAL SCIENCES TO THE US AEC DIVISION OF BIOLOGY AND MEDICINE. VOLUME 4: INSTRUMENTATION.

Solid State Gamma Spectrometry Developments. p. 4-10. A spectrometer system using a molecular sieve instead of an ion pump as the pumping agent was assembled. A highly resolved spectrum of $^{239}$Pu was achieved and emissions which had not been observed previously were found.
THE TREATMENT AND EVALUATION OF INTERNAL DEPOSITION FROM A PLUTONIUM WOUND.

W. V. Baumgartner, C. E. Newton, H. V. Larson, G. E. Crook (Battelle Memorial Institute, Pacific Northwest Lab., Richland, Wash.)


Fragments of plutonium with activity exceeding 10 mCi, resulting from an explosive disintegration of plutonium metal, penetrated a hood glove and lodged in the left upper arm of an employee. The health physics and medical aspects of the incident were followed rigorously; urine samples continuously analyzed for plutonium activity; blood samples obtained and analyzed prior to the administration of some 70 intravenous injections of diethylene-triaminepenta acetic acid (DTPA); and the activity in the wound measured frequently. Observations on modifications of prior methods of treatment are presented, i.e., washing open wound with solutions of DTPA and effect of intramuscular injections of DTPA into the wound site.

Discussions are presented that include methods of evaluating the internal deposition. These include a comparative study of past deposition cases that were also complicated with the administration of chelating agents and trends from this study which help to establish estimations of internal depositions from very early bioassay results even when the data are influenced by DTPA treatment. The use of digital and analog computer techniques using the standard models with observed data is analyzed and the value of the analog computer for aiding the understanding of excretion patterns from plutonium deposition cases discussed. (auth.)

WHOLE BODY COUNTING OF PLUTONIUM IN DOGS.

K. L. Swinth (Battelle Memorial Institute, Pacific Northwest Lab., Richland, Wash.)


Thin NaI(Tl) scintillation counters were used for the assessment of body burdens in dogs exposed to Pu$^{239}$ aerosol. We made experiments with both a dog phantom and live dogs. The count rate was affected by changes in dog size, isotopic composition of the plutonium, and by plutonium distribution in the body.
A 50% change in dog weight made a change of a factor of two in counting efficiency in some experiments. The sensitivity for a typical dog when counting the x-rays emitted is 2.1 cpm/nCi and 14.0 cpm/nCi when counting both x-rays and 60 keV gamma rays. This sensitivity is based on body burdens determined by post-mortem alpha-counting analysis of the tissues of several dogs. To date, we have counted 30 exposed dogs, which had body burdens ranging from 0.12 to 2.9 μCi. (auth.)
Chemistry

ANALITICHESKAYA KHIMIYA PLUTONIYA, \(^{242}\)Pu. (ANALYTICAL CHEMISTRY FOR PLUTONIUM, \(^{242}\)Pu).
M. S. Milyukova, N. I. Gusev, I. G. Sentyurin, I. S. Sklyarenko

Chemical Abstracts, Vol. 65, Abs. No. 9736g.
September 26, 1966.

ANALYSIS OF THE CHEMICAL STATE OF PLUTONIUM AND FISSION PRODUCTS IN PROCESS FEED SOLUTIONS.
M. Bonnevie-Svendsen, V. Martini (Institutt for Atomenergi, Kjeller, Norway)

The chemical states of plutonium and fission products in reprocessing feed solutions are discussed and the need for simple routine analytical procedures for the characterization of species with different extractability is stressed. The development of an extraction chromatographic procedure for the determination of Pu(III), Pu(VI), and Pu(IV) is described. The main source of error was a light-induced reduction of Pu, which occurred in the presence of excess uranyl nitrate. Performed under exclusion of light, the method is well suited for the sensitive determination of Pu(III) and Pu(VI) in feed solutions. Less than a microgram of Pu is required, and macroconcentrations of U and technical impurities do not interfere. A fractionation of ruthenium species according to their TBP-extractability can be obtained in the same operation. The possibility for applying a similar procedure to zirconium and niobium is discussed. 24 references. (auth.)

ANALYTICAL CHEMISTRY SECTION PROGRESS REPORT, JANUARY 1-DECEMBER 31, 1965.
J. E. Harrar, comp. and ed. (University of California, Lawrence Radiation Lab., Livermore, Calif.)

The following sections of this report contain information about plutonium: preparation of plutonium samples for oxygen determination, p. 7; and measurement of plutonium emission spectrum wavelengths, p. 9-10.
ANALYTICAL DETERMINATION OF AMERICIUM, PLUTONIUM, AND URANIUM USING THE ANION-EXCHANGE RESIN AMP.
A. M. Vorob'ev, V. I. Formicheva

A method was developed for separating americium, plutonium, and uranium using the resin AMP, based on the difference in the degree of sorption of uranium (VI), plutonium (IV) and (III), and americium (III) ions from hydrochloric acid of various concentrations. This difference is so great that it permits a quantitative separation of these elements.

CALCULATIONS OF THE THERMODYNAMIC PROPERTIES OF GASEOUS URANIUM AND PLUTONIUM AND THEIR IONS UP TO 20,000°K.
L. V. Gurvich, V. S. Yungman (Institut Vysokikh Temperatur An SSSR, Moskva, SSSR)

In Russian.

This paper contains a critical analysis of the results obtained by investigating U(I)-U(V) and Pu(I)-Pu(V) optical spectra. The authors have compiled tables of the energy levels of the corresponding atoms and their ions (with the exception of U(III) and Pu(III)). On the basis of this information calculations have been made of the thermodynamic functions of gaseous uranium and plutonium and their ions in the temperature range from 293.15 to 20,000°K. The calculations were carried out without limitations on the statistical sums since the available data on the energy levels and ionization potentials of the atoms and ions under consideration are extremely incomplete and fragmentary. The authors consider the question of the accuracy of the calculated values for the thermodynamic functions and demonstrate that the lack of reliable data on energy levels leads to substantial errors in these values, particularly at high temperatures. (auth.)
3330. CHARACTERIZATION OF PLUTONIUM COMPOUNDS.
A. G. Miller (Isochem, Inc., Richland, Wash.)

This report describes the establishment of equipment in hoods to make possible the measurement of moisture content, surface area, particle size, oxygen/metal ratio, and pyrohydrolysis of plutonium compounds. 12 references.

3331. COMPOUNDS OF TRIVALENT PLUTONIUM, AMERICIUM, AND CURIUM WITH 8-HYDROXYQUINOLINE AND SOME OF ITS DERIVATIVES.
C. Keller, S. H. Eberle, K. Mosdzelewski
(Kernforschungszentrum, Karlsruhe, W. Germ.)

In German.

Chelates Me (ligand)₃ of trivalent plutonium and americium with 8-hydroxyquinoline are obtained by precipitation from aqueous solutions. As the plutonium compound is very easily oxidized to Pu(IV), it can only be prepared in a completely oxygen-free medium. The properties of the oxinato-complexes of trivalent actinides are much like those of the analogous rare earth compounds.

Solutions of the chelating agent in chloroform extract the compounds Am(oxine)₂⁻, Cm(oxine)₂⁻ and Am(5,7-dichloro-oxine)₃⁺ into the organic phase, Y probably being OH⁻. The overall stability constant of Am(5,7-dichloro-oxine)₃⁺ is logβ₃ = 21.93 at T = 25.0 ± 0.5 °C and μ = 0.1. 18 references. (auth.)

3332. COULOMETRIC DETERMINATION OF CARBON IN PLUTONIUM AND IN URANIUM-MOLYBDENUM ALLOYS.
Bernard Pichotin, Philippe Chasseur (C.E.N., Saclay, France)

In French.

This method is fast and sensitive. The uranium alloy determination takes 10 minutes, the plutonium determination takes 45 minutes, and the limit of sensitivity for both is about 5 ppm in a 1 g. sample.
DEFECT EQUILIBRIA OF PuO$_{2-x}$, 1100 to 1600°C.
L. M. Atlas, G. J. Schlehm (Argonne National Lab., Argonne, Ill.)

The variation of oxygen content and electrical resistivity ($\sigma$) with oxygen pressure ($p(O_2)$) has been measured for PuO$_{2-x}$ between 1100°C and 1600°C. Experimental results are presented as plots of $\Delta G^\theta(0)$ (the relative partial molar free energy of oxygen) vs. log x, $\Delta G^\theta(0)$ vs. temperature, log x vs. log $p(O_2)$ and log $\sigma$ vs. log $p(O_2)$. The log-log plots of x and $\sigma$ against $p(O_2)$ are linear over a wide composition range and at each temperature, the composition line has a steeper slope than the conductivity line. A mass-action analysis of the data suggests that the defects in PuO$_{2-x}$ are predominantly interstitial plutonium ions, and that at 1100, 1200 and 1600°C, they are primarily in a $^+$4 state. Between 1200 and 1300°C dissociation of the interstitials appears to be retrograde and at 1300°C an appreciable concentration of Pu$_{1.3}^+$ occurs. As the temperature rises from 1300 to 1600°C, the concentration of Pu$_{1.3}^+$ diminishes.

A detailed consideration of the enthalpy and entropy changes involved in the formation of plutonium interstitials permits the formulation of a general relation between x, $p(O_2)$ and T. This equation successfully predicts $p(O_2)$ for PuO$_{1.90}$ at 1100, 1300, 1400, 1500 and 1600°C on the basis of data measured at 1200°C. After imposing the condition $\partial (\ln p(O_2))/\partial x=0$, the equation also predicts the transformation of PuO$_{2-x}$ into a mixture of solid phases at a temperature in the region 675-685°C.

THE DETERMINATION OF PLUTONIUM IN URANIUM-PLUTONIUM SOLUTIONS BY DIRECT ALPHA COUNTING.
H. H. Ph. Moeken (Eurochemic, Mol, Belgium)

This method provides a quick determination of plutonium which is sufficiently precise for process control. A pipetted U-Pu solution in tetraethyleneglycol is heated to dryness and counted. The counting result is corrected
Chemistry (Cont'd)

3334. (Cont'd) for the known uranium content. This method
is precise at weight ratios of U/Pu of less
than $10^5$.

3335. DETERMINATION OF PLUTONIUM THROUGH POTENTIOMETRY AT
CONSTANT INTENSITY. APPLICATION TO THE RAPSODIE
FUEL ELEMENT.
J. Corpel, F. Regnaud (C.E.A., Fontenay-aux-Roses, France)

A new method is proposed for the determination of
plutonium in the UO$_2$-PuO$_2$ pellets for Rapsodie.
The sample is dissolved in a mixture of 11 N nitric
acid and 0.1 N hydrofluoric acid, and the plutonium
is oxidized with silver (II) oxide. Plutonium-(VI)
is reduced in the presence of sulphamic acid with
an excess of iron (II) which is back-titrated with
cerium (IV) solution to a constant-current potentiome-
tric endpoint. Uranium and iron do not interfere
and no separation is required. In routine work,
the method gives a precision of 0.5%. (auth.)

3336. A DIFFERENTIAL SPECTROPHOTOMETRIC DETERMINATION OF PLUTONIUM (III)
USING COLORED GLASS FILTERS.
L. W. Winkle (Isochem, Inc., Richland, Wash.)

A procedure is described for the precise spectro-
photometric determination of plutonium (III) in the
10-15 g/l range. The method is similar to that re-
ported by Phillips, except that the relative absorb-
ance of the sample solution is measured against a
colored glass filter instead of a standard plutonium
solution. The concentration range which gives max-
imum precision was determined for a double-beam
spectrophotometer (Beckman DK-2A) and the influence
of extraneous elements was evaluated. The standard
deviation for 10 samples using a filter as reference
was the same as that reported earlier by Phillips
for aqueous standards ($\pm 0.05\%$). (auth.)
Measurements of solid state diffusion, in an electric field, of various metals in trace concentrations have been made using cerium, uranium and plutonium as solvent metals. An apparatus is described which permits sustained experiments in a controlled atmosphere under constant temperature conditions. Extensive data have been obtained in the case of cerium in the temperature range of 490-650°C at current densities from 250 to 500 A/cm² and over times up to 240 hours. Data are presented for a dozen solute elements. In the case of some transition elements, notably iron, cobalt and nickel, the migration is quite rapid. The use of radioactive tracers, where possible, provided data for quantitative treatment of the results. Spectroscopic analysis provided additional information. Migration rates in uranium measured at 900°C were lower and reduced even more in plutonium at 500°C. However, it was still possible to measure a rate of electrodiffusion of iron. No movement was detected for antimony, magnesium, manganese, silicon or zirconium. With the exception of molybdenum and tin, the metals studied migrated towards the anode. Electrodiffusion presumably results from the net effect of the electric field acting on the ions and from momentum interchange between the ions and conduction electrons. The two effects may thus oppose or reinforce each other. It is felt that the field effect is greater in the cases studied. The tendency of the solute element to form a compound with the solvent metal is one measure of whether migration is to be expected. It is also shown that a relative size effect is important. An interesting aspect of the electrodiffusion of iron in cerium is the very low (≤2 kcal) activation energy. Some comparisons have been made with chemical gradient diffusion. 20 references (auth.)
ELECTROMIGRATION INVESTIGATION OF THE COMPLEXATION OF TRIVALENT PLUTONIUM WITH SOLUTIONS OF ETHYLENEDIAMINETETRAACETIC ACID.

A. V. Stepanov, T. P. Makarova


The complexation of Pu(III) with trilon was investigated using an electromigration method: \( \mu = 0.1 \), \( t^0 = 25^\circ \), \( \text{pH} = 1.1-3.5 \). It was established that under these conditions two forms of complex ions are formed: \([\text{PuH enta}]^0\) and \([\text{Pu enta}]^-\). The latter predominates in solutions with \( \text{pH} > 3 \). The instability constants of complex forms of trivalent plutonium were determined:

\[
\log K_{\text{Pu enta}} = -15.0 \pm 0.3
\]

\[
\log K_{\text{PuH enta}} = -4.6 \pm 0.3
\]

12 references. (auth.)

THE EVAPORATION BEHAVIOR, THERMODYNAMIC PROPERTIES AND SYSTEMATIC TRENDS OF ACTINIDE METAL-OXYGEN SYSTEMS.

R. J. Ackermann, R. J. Thorn (Argonne National Lab., Argonne, Ill.)


Mass effusion measurements and mass spectrometric observations have been combined with existing calorimetric and recent EMF data to yield a consistent thermodynamic description of actinide metal-oxygen systems. Any meaningful description of the evaporation behavior of a given condensed phase must include a careful specification of either the partial pressure of oxygen in the system or the composition of the condensed phase because of all of the solid dioxide phases, for example, exhibit bivariant behavior due to variation in composition. The solid stoichiometric dioxides increase in total vapor pressure in the somewhat inverted order, ThO₂, PuO₂, NpO₂, and UO₂, and in each case the gaseous dioxide is the predominant vapor species. The ease of reduction in vacuum of these phases increases with increasing atomic number giving rise to a MO₂-x phase and/or metal in the case of Th and U, and to lower oxides in the case of Np and Pu. Reduction of a given dioxide results in an increase of the total vapor pressure due to
the rapid increase of the gaseous monoxide which becomes the major vapor species. This behavior is at present most completely resolved in the case of the uranium oxygen system.

There are two significant systematic trends involving the vapor phase: (1) the gaseous dioxides (plus 4 valence states of metal) become less stable ($\Delta G^\circ$ becomes more positive) with increasing atomic number, and (2) the gaseous monoxides (plus 2 valence states of metal) become more stable with increasing atomic number. Other correlations and comparisons of the thermodynamic properties in the solid, vapor and solvated phases of actinide metals and compounds are made with those of lanthanide metals and compounds. The trends in the former case are generally more complex as a result of the more pronounced dual nature of the actinides as transition elements with d-electrons and rare earth elements with f-electrons. 44 references. (auth.)

EXTRACTION OF PLUTONIUM WITH N-BENZOYL-PHENYL-HYDROXYLAMINE.

In this work the extraction of plutonium from aqueous solutions has been studied by means of a cupferron analog, N-benzoyl-phenyl-hydroxylamine (BPH), dissolved in chloroform. Plutonium in the tetravalent state can be extracted from HCl-citric acid solutions in the presence of NaNO$_2$ in the studied pH range, i.e. from 0 to 4.3. For a 0.4 BPH concentration the maximum distribution coefficient is $4 \times 10^3$ at a pH of 1.2. Alpha-emitters which are not tetravalent, U(VI), Np(V) and Am(III), are not extracted under these conditions.

Pu(IV) can also be extracted from HClO$_4$-citric acid solutions with distribution coefficients higher than 250 in the range between 4.5 M acid and pH 4.7, the maximum value being $3.5 \times 10^3$ at pH 0.25.

The influence of other variables on the distribution coefficient has also been studied, such as stabilizing agents (NaNO$_2$, K$_2$Cr$_2$O$_7$), contacting time, and the concentration of BPH in the organic phase.

14 references. (auth.)
A new flat bed gas-solids contactor has been designed and evaluated on a laboratory scale. The unit has the advantages of being critically safe by geometry, easy to scale up, gives excellent gas-solids contact, is operationally insensitive to gas and solid feed rates, and is mechanically simple to operate.

Two reactions studied in the unit were the hydrofluorination of plutonium oxide, and the hydrogen reduction of uranium oxide. Good fluoride conversions were obtained at reasonable throughputs and operating temperatures when oxide produced by decomposition of plutonium oxalate or direct calcined oxide containing an activating agent were used. Hydrogen reduction of uranium oxide was demonstrated although maximum throughputs were not determined.

Based on the results obtained, the feasibility of the contractor design for halogenation and hydrogen reduction of fissile materials has been demonstrated. (auth.)

Fluid-bed volatility processes for recovering uranium and plutonium from spent nuclear reactor fuels appear promising because of the relatively low process waste volumes, excellent probable decontamination, and apparent mitigation of criticality problems. Conceptual plant studies and evaluation of plant-scale component designs are being performed at the Oak Ridge Gaseous Diffusion Plant. Crucial equipment items, including the fluid-bed reactors, will be tested on a semiworks scale using nonirradiated, uranium materials to confirm design assumptions and to evaluate the scale-up problems. Peripheral equipment items, such as gas compressors, piping connectors, valves,
Chemistry (Cont'd)

3342. (Cont'd) filter media, sorbent trapping systems, and solids transfer equipment, are included in the test plans. Preliminary results are given for filter media, compressor, and connector tests; and some materials of construction studies are discussed. The scale-up work and engineering studies at the ORGDP, coupled with the basic experimental programs being carried out at the Argonne, Oak Ridge, and Brookhaven National Laboratories, are aimed at providing private industry with sufficient information to allow design of commercial volatility reprocessing plants. (auth.)

3343. FORCE CONSTANTS OF METAL HEXAFLUORIDES.
S. N. Thakur, D. K. Rai (Banaras Hindu University, Varanasi, India)
The simple and modified UBFF have been applied to thirteen octahedral metal hexafluorides, including PuF₆. The force constants have been adjusted by a least squares method. The force constants show certain regularities. The covalent character of the metal fluoride, metal chloride and metal bromide bonds has been discussed. 12 references. (auth.)

3344. THE HEAT OF SOLUTION OF PLUTONIUM IN HYDROCHLORIC ACID.
V. V. Akhachinskii (Moskovskii Gosudarstvennyi Universitet) M. V. Lomonosova (Moscow, USSR)

In Russian.
The following values for the heat solution of plutonium in 6 M hydrochloric acid have been given in four works published to date:
-141.64 ± 0.2 kcal/mole, -141.02 ± 0.19 kcal/mole, -141.14 ± 0.14 kcal/mole and -138.90 ± 0.9 kcal/mole.
The authors of the last of these works consider their value to be the correct one, but perusal of all the cited works shows that one cannot give a clear preference to their result.

The present paper gives the result of a new determination of the heat of solution of alpha plutonium in 6 M hydrochloric acid. A value of \(-141.5 \pm 0.4\) kcal/mole is found, allowance being made for the impurities in the plutonium. The close agreement between this value and the first three cited above casts doubt on the fourth result and it is therefore essential that the values given in it for the heats of formation of PuCl₃, PuOCl and the Pu³⁺ ion be recalculated.

It should be borne in mind that

\[
\begin{align*}
\Delta H_{298}^0 \text{PuCl}_3 \text{ (crystal)} &= -229.8 \pm 0.5 \ \text{kcal/mole} \\
\Delta H_{298}^0 \text{PuOCl} \text{ (crystal)} &= -222.7 \pm 0.5 \ \text{kcal/mole} \\
\Delta H_{298}^0 \text{Pu}^{3+} \text{Pu}^{3+} \text{ Pu}^{3+} \text{P} \text{O} \text{H}_2\text{O} &= -141.6 \pm 0.5 \ \text{kcal/mole}.
\end{align*}
\]

(auth.)

HIGHER SULFIDES AND SELENIDES OF PLUTONIUM.
Jean-Pierre Marcon, Roger Pascard
In French.

We have prepared the following compounds; PuS₂, PuSe₂, and Pu₂S₃ in three crystalline forms α, β and γ, Pu₂Se₃ in two forms n and γ. All of these compounds are isotopic with their homologs with the lanthanides. In particular, PuS₂ is a polysulfide and PuSe₂ a polyselenide, the structure deriving in both cases from the Fe₂ as type structure. (auth.)

THE INVESTIGATION OF THE RANGE DISTRIBUTION OF PU-FISSION PRODUCTS WITH THE HELP OF THE STRIPPING METHOD.
M. Hollstein, H. Munzel (Kernforschungszentrum, Karlsruhe, W. Germ.)
In German.
The stripping-method used for dissolving thin layers from aluminum absorbers was investigated. It was shown that elements considerably more electropositive than aluminum are only partially stripped. Therefore activity is retained by the catcher in investigations of the range distribution of fission products corresponding to those elements. A correction for this effect is possible. The range distribution in aluminum of cumulatively formed \(^{140}\text{Ba}\) and \(^{127}\text{Sb}\) from the fission of \(^{239}\text{Pu}\) by thermal neutrons was studied by means of the stripping-method. The mean range and relative range-dispersion are, respectively, for \(^{140}\text{Ba}\) 2.95 mg Al/cm\(^2\) and 20.3\%, for \(^{127}\text{Sb}\) 3.6 mg Al/cm\(^2\) and 23.1\%. The low-energy component which occurs in the range distribution curves is discussed. 21 references. (auth.)

A four-column ion exchange purification-concentration process for americium.
R. S. Kingsley (General Electric Co., Hanford Atomic Products Operation, Richland, Wash.)

A four-column ion exchange purification process is described for americium from a synthetic Americium Recovery Facility product containing gross amounts of the transition, alkaline earth, and rare earth metals as impurities. A decontamination factor of \(>50\) was obtained for plutonium. The column used for plutonium and iron purification is described in some detail. 22 references.

This article includes a history of the production of plutonium \(^{237}\text{Np}\) used as a source of \(^{238}\text{Pu}\), the use of \(^{248}\text{Pu}\) as an energy source in satellites, and a comparison of the chemistry of neptunium and plutonium. 13 references.
PLUTONIUM PURIFICATION WITH ION EXCHANGE RESINS.
SPECIAL REPORT NO. 1.
(Centre d'Etude de l'Energie Nucleaire and Societe Belge pour l'Industrie Nucleaire, Brussels, Belgium)

The first part of the present report describes the loading conditions of plutonium from nitric-hydrofluoric solutions, free from metal impurities (about 5 to 110 g Pu/l; about 7.5 and 11 M HNO₃) on Permutite SK (20-50 and 40-70 mesh), Dowex 1°XL, X₄, X10, Amberlite IRA 400 and Dowex 50 resin columns, thermostatized at 60°C. Then the washing operation and elution conditions of the adsorbed plutonium are dealt with. In order to explain experimental data, a theoretical interpretation of the exchange kinetics during loading and elution is given. The second part discusses the loading, washing and elution conditions of plutonium by means of an identical resin column, but fed with nitric solutions (7 and 11 M) containing metal impurities such as zirconium, iron, uranium, molybdenum, etc., in variable ratios. The behavior of these elements was particularly studied during the washing operation. The decontamination factors obtained after one or two purification cycles are also reported.

Appendices I and II relate the plutonium absorption and desorption techniques under well-defined working conditions. Appendix III describes the purification equipment required for the needs of the program, as well as the procedure and the material balance. Finally, in Appendix IV a comparison is made between the reprocessing procedure by means of ion exchangers, on the one hand, and by amines on the other. 68 references. (auth.)
Several important high temperature reactions for processing plutonium involve equilibria between the metal and a fused salt system. In almost all these processes, the kinetics are rapid, and equilibria are of overwhelming importance. Therefore, the application of appropriate thermodynamic data and estimates are of major importance in selecting the proper reaction system.

Examples of high temperature plutonium processes where thermodynamic considerations are of principal importance are bomb reduction of plutonium halides by an active metal such as calcium, electrorefining of liquid plutonium in fused salt media, and extraction of impurities such as radionuclides from molten plutonium by fused salt media or solid oxide media. The thermodynamic principles involved in these processes, including selection of container materials, are reviewed and compared with experimental results. 42 references. (auth.)

In Spanish.

Chemistry (Cont'd)


Dicesium plutonium hexachloride has been proposed as a primary analytical standard for plutonium, but the cesium interferes in some assays. This paper shows that cesium can be removed from this compound and the plutonium subsequently can be quantitatively recovered, thus increasing the versatility of dicesium plutonium hexachloride. 12 references.


The kinetics of the sorption of plutonium (IV) on Dowex 1 x 4 from nitrate solutions are presented. Sorption rates from various nitric acid and mixed aluminum-nitrate/nitric-acid solvents were studied. The particle-diffusion controlled reaction proceeds with an apparent average diffusion coefficient which decreases exponentially with increasing plutonium sorption. The results agree reasonably well with a computer solution of Fick's law with the local intraparticle diffusion coefficient proportional to the local intraparticle fraction of unreacted exchange sites. The trace-sorption diffusion coefficient is strongly dependent upon the composition of the aqueous phase, and is empirically shown to be roughly inversely proportional to the product of the trace-sorption distribution coefficient and the ratio of the hydronium ion-to-water concentrations. These kinetic results and previously reported equilibrium parameters may be coupled with the fundamental partial-differential equation which describes the operation of a fixed-bed anion-exchange column to allow computer optimization of plant-scale processing equipment. The effects of the variation of such parameters as volume and mass flow rate, temperature, plutonium feed concentration, and macro concentrations of aluminum nitrate in the feed are qualitatively discussed for one equipment design. 14 references. (auth.)
PROCESSING OF PLUTONIUM BY ION EXCHANGE. VIII.
SELF DIFFUSION STUDIES IN ANION-EXCHANGE RESIN.
D. B. James, W. A. Beyer (Los Alamos Scientific Lab., Los Alamos, N. Mex.)

The isotopic migration rate of plutonium (IV) in Dowex 1 anion-exchange resin was studied with varying temperature, nominal percent cross-linking, and fractional equilibrium loading with plutonium. The diffusion coefficient decreases very rapidly with increasing loading. A diffusional model that is suggested by these results is qualitatively discussed. (auth.)

PRODUCTION OF URANIUM, THORIUM, AND PLUTONIUM AND THEIR COMPOUNDS.
J. J. Barghusen, P. A. Nelson
Summer 1966.

This review article contains information on the reduction of plutonium by U(IV), the precipitation of plutonium trifluoride, conversion of plutonium dioxide to plutonium trichloride, the production and properties of uranium and plutonium carbides, and the production and properties of uranium and plutonium mixed-anion fuels. 37 references.

QUANTITATIVE ELECTRODEPOSITION OF ACTINIDES FROM ISOPROPYL ALCOHOL.
M. Yates Donnan, E. Kenneth Dukes (E.I. du Pont de Nemours & Co., Savannah River Lab., Aiken, S. Car.)

An electrodeposition method was developed for the preparation of thin, uniform alpha sources for precise counting and accurate pulse height analysis. The method employs an isopropanol electroplating medium and is applicable to deposition of actinides from both organic and aqueous media. Moderate amounts of aluminum and iron do not decrease deposition yields. (auth.)
By radiochemical range determinations on some fission fragments from the fission of $^{239}$Pu, the change of the energy minimum was investigated as a function of the excitation energy of the fissioning nucleus. These studies were carried out at an excitation energy range between 5 and 40 MeV. At 5 MeV the fission yields from symmetric fission were approximately 3 orders of magnitude below those of asymmetric fission whereas they were equal at 40 MeV. A method was developed by which it was possible to prepare gold foils of uniform thickness. The range of some fission products ($^{91}$Sr, $^{92}$Sr, $^{97}$Zr, $^{111}$Pd, $^{138}$Ba) from the fission of $^{239}$Pu by thermal neutron and deuterons at 12 and 20 MeV were determined by the thin target-thin collector method. The ranges of some fission products ($^{91}$Sr, $^{92}$Sr, $^{112}$Ag, $^{118}$Ag, $^{118}$Cd, and $^{139}$Ba) from the fission of $^{239}$Pu by thermal neutrons and deuterons at 20 and 50 MeV were determined by the thick target-thick collector method. An evaluation was made of the excitation energy and the frequency of the intermediate nuclei and fission nuclei in the fission by neutrons of varying energies. The most probable charge of the fission fragments, and the number of evaporated neutrons could then be calculated. From the ranges, the values of the total kinetic energies were calculated and the results were compared with previous results. The comparison showed that the value of the total kinetic energy in the area of mass ratios from 0.8 to 1.3 (asymmetric fission) are independent of the excitation energy of the fissioning nucleus. The value of the total kinetic energy in the area of symmetric fission increases by about 8 MeV from 5 to 20 MeV excitation energy and remains constant thereafter. It could be shown that these results are in good agreement with the hypothesis of the 2 independent fission types for symmetric and asymmetric fission. The usual assumption for the distribution of the excitation energy of the fissioning nucleus on the primary fission fragments in the ratio of their neutron numbers appears incorrect. This could be evaluated from the number of evaporated neutrons in the area of symmetric fission. 50 references.
Chemistry (Cont'd)


The following ions were separated: Tb(III), Hf(IV), Cd(II), and Zn(II) from U(VI), Am(III) from Pu(VI), Te(IV) from Sb(V), and Ni(II) from Co(II) and Cu(II). 12 references.


This report includes a review of the nuclear and chemical features of plutonium of interest to the radiochemist, a discussion of sample dissolution and counting techniques, and a collection of radiochemical procedures for plutonium. The literature search is complete through September 1964. 455 references.


In German.

A method was developed by which it is possible to measure the ranges of fission products in aluminum in a "thick target-thick catcher" arrangement. The target consists of a very dilute alloy of $^{239}$Pu in aluminum; the catcher of pure aluminum. Therefore a correction for the different scattering of fission products in target and catcher can be neglected. By means of this method the fragment ranges of $^{139}$Ba and $^{139}$Cs as well as the cumulative ranges of $^{139}$Ba, $^{139}$Cs and $^{139}$Xe were determined. According to our results the fragment ranges $R_f$ within an isobaric chain are different. Their relative values normalized to the isobaric range $R_{is}$ are linearly dependent upon the difference $Z-Z_p$: $R_f/R_{is} = 1.0-0.037 (Z-Z_p)$. Using this equation and assuming a constant range disper-
Chemistry (Cont'd)

3361. (Cont'd)  

It follows that the dispersion in the kinetic energy of primary fission fragments is about a factor of 0.5 smaller than previous values. 13 references. (auth.)

3362.  


Research centered around the use of chlorine trifluoride, at low temperature, as a fluorinating agent for the production of uranium hexafluoride. In this case the plutonium remains in the residue and is transformed into a hexafluoride by fluorine at around 550°C.

Normal progress is reported in the other fields of research, namely plutonium volatilization, decladding, chemical attack on uranium carbide, conversion of plutonium fluoride into an oxide in a molten salts medium, detection of traces of fluorinating agents, and the chemical resistance of the ventilation filters.

Construction of the hot semi-pilot continued with the installation of the main alpha cell. (auth.)

3363.  

RESEARCH AND DEVELOPMENT ON NONAQUEOUS PROCESSING.  
J. J. Barghusen, W. J. Mecham, L. Burris, G. A. Bennett  

This review article contains information about fluid-bed fluorination of UO₂-PuO₂ pellets, the separation of uranium from plutonium, the preparation and properties of plutonium fluoride complexes, salt-transport processes, conversion of fluorides to oxides in fused-salt media, and purification of plutonium by electrorefining. 64 references.
Chemistry (Cont'd)

3364. SOME KEY THERMOCHEMICAL DATA FOR PLUTONIUM COMPOUNDS
M. H. Rand (Atomic Energy Research Establishment, Harwell, U.K.)

The key data for thermochemical calculations involving plutonium compounds are discussed. The problems involved in determining reliable standard entropies for plutonium compounds are emphasized, and it is suggested that much more use will have to be made of accurate measurements of equilibria involving the gaseous plutonium species whose standard entropies are known (namely) Pu(g) and PuF$_6$(g) to obtain entropies of condensed phases. A number of equilibria which could be of use for this purpose are discussed. The data for the plutonium carbides have been critically assessed. 28 references. (auth.)

3365. THERMODYNAMIC DATA FOR PLUTONIUM OXIDES.

Thermodynamic data have been obtained for plutonium oxides with O/Pu ratios between 1.53 and 2.00 at temperatures between 700 and 1140°C, using a high temperature galvanic cell, containing solid electrolytes reversible to oxygen ions only and employing two reference electrodes (Ni, NiO and Fe, wustite). A large change in $\Delta H$($O_2$) occurs between PuO$_2$ and PuO$_{1.9}$ and the thermodynamic data is sufficiently precise to give reliable values of $\Delta H$($O_2$) and $\Delta S$($O_2$) for all compositions between PuO$_{1.52}$ and PuO$_{1.98}$ for 700 < $T$ < 1140°C.

The plutonium-oxygen phase diagram has recently been re-investigated for O/Pu ratios between 1.50 and 2.00 at temperatures between 25 and 900°C, using a high temperature X-ray powder diffraction
A new phase diagram is suggested which attempts to summarize the work of the several investigators.

The thermodynamic data, obtained from EMF cell measurements, agrees with the suggested phase diagram, of which it is entirely independent, extremely well, and an extrapolation of the thermodynamic data from 700°C to room temperature has been made that is consistent with the suggested phase diagram. For the reaction \( 2 \text{Pu}_2\text{O}_3 + \text{O}_2 \rightarrow 4 \text{PuO}_2 \), an estimated value of \( \Delta C_p \) has been taken from the value for the corresponding cerium oxide reaction.

The partial molar heats and entropies have been calculated for the several phase regions and \( \Delta S(O_2) \) and \( \Delta H(O_2) \) for the entire region from \( \text{Pu}_2\text{O}_3 \) hex to \( \text{PuO}_2 \), has been integrated and the heats of formation and standard entropies calculated for \( \text{Pu}_2\text{O}_3 \) hex and \( \text{PuO}_{1.61} \). 

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THE VAPOR PRESSURE OF PLUTONIUM.
R. N. R. Mulford (Los Alamos Scientific Lab., Los Alamos, N. Mex.)


The vapor pressure of liquid plutonium has been determined over the temperature range 1100 to 1800°C by the Knudsen effusion method. The least-squares equation which fits the data is \( \log_{10} p(\text{atm}) = -17.420/T(\text{K}) + 4.913 \). The standard deviation corresponds to about \( +10\% \) in the pressures calculated from this equation. The heat of vaporization computed from the temperature dependence of the experimental data is \( \Delta H_{298}^0 = 82.3 \text{ kcal/g-at} \). The heat computed by combining independent entropy and heat capacity data with the present measurements is \( \Delta H_{298}^0 = 82.1 \text{ kcal/g-at} \). Effects of oxygen upon the volatility of liquid plutonium were sought by comparing the vapor pressures observed with the liquid in contact with tantalum, tantalum carbide, magnesia, and plutonium sesquioxide. No differences were found. In addition, the vapor pressure was measured with different degrees of vacuum in the system. No effect was found here either, except that in very poor vacuums a surface film of oxide apparently formed and reduced the volatility by about a factor of 2. (auth.)
CHARACTERISTICS OF THE AEROSOL PRODUCED FROM BURNING PLUTONIUM.
H. J. Ettinger, W. D. Moss, H. Busey (Los Alamos Scientific Lab., Los Alamos, N. Mex.)

Safety analysis of sodium-cooled, plutonium-fueled, fast reactor plants must be concerned with the possibility of fires involving these materials. Design of an air cleaning system for such a facility requires basic data defining the aerosol characteristics of sodium and plutonium released during a fire.

Size characteristics of the aerosol produced during plutonium fires were determined for different atmospheres ranging from 20.8% oxygen, 79.2% nitrogen to 0.5% oxygen, 99.5% nitrogen. Plutonium-cobalt-cerium alloy, and alpha- and delta-phase plutonium metal were burned. Data were obtained to estimate the fraction of plutonium alloy airborne during a plutonium fire. Fires simulating a reactor accident involving both fuel and coolant defined the relative airborne concentration of plutonium and sodium. 10 references. (auth.)

CONSEQUENCES OF ACTIVITY RELEASE.
K. E. Cowser

The radiation-safety measures needed for medical work with sealed and unsealed sources of radioactive materials in small laboratories, clinics, and hospitals are reviewed. Pu-238, Pu-239, Pu-240, Pu-241, and Pu-242 are among the isotopes considered. 52 references.

DESIGN CRITERIA, PLUTONIUM BUY-BACK FACILITY.
M. E. Borgeson, D. C. Nelson (General Electric Co., Hanford Atomic Products Operation, Richland, Wash.)

Plutonium is produced as a valuable by-product in the operation of uranium-fueled nuclear reactors for research or for power. The Atomic Energy Commission has offered to buy back from private industry plutonium produced by power reactors operating on fuel supplied by the AEC. It is expected that the plutonium can be sold at some
Health & Safety (Cont'd)

3369. (Cont'd) later date for use in research programs or for fueling of plutonium recycle reactors.

The Richland Operations office of the Atomic Energy Commission has been assigned responsibility for receiving, storing and repackaging-for-shipment this recovered plutonium. The plutonium will be separated from fuels and purified at Nuclear Fuel Service (NFS) facilities in New York state, and shipped to HAPO as a plutonium nitrate. The design criteria presented herein provide the bases for the facilities required at Z Plant to receive and repackage for storage or shipment the plutonium nitrate to be received from NFS. In addition, bases are provided for facilities to enable packaging of HAPO produced plutonium liquids for off-site shipment. (auth.)

3370. DOSE CONSTANTS OF GAMMA-RAYS.  
W. Marth (Gesellschaft für Kernforschung mbH, Karlsruhe, Germany)  
In German.

Partial and total dose constants are given for many nuclides generated by reactor activation, including Pu-240. 9 references.

3371. INDEXED BIBLIOGRAPHY OF CURRENT NUCLEAR SAFETY LITERATURE-6.  
(Oak Ridge National Lab., Oak Ridge, Tenn.)  

This quarterly bibliography includes 20 entries on plutonium.

3372. INCIDENTS RELATED TO NUCLEAR SAFETY IN AEC OPERATIONS.  

A summary of these accidents during 1965 includes 6 plutonium incidents.
Recent publications which have discussed the production of particulate material by oxidation of plutonium and the effects of particle size and composition on retention in the human respiratory system are summarized. 26 references.

Recommendations are given for design, manufacture, and maintenance of packings for Pu-239 and Pu-241 and other fissile materials.
AUTORADIOGRAPHY AS A METHOD FOR DIFFERENTIATING PuO₂ FROM UO₂ IN IRRADIATED MECHANICALLY MIXED OXIDE FUELS.
W. J. Gruber, J. L. Hascall, J. O. Blakeley, D. J. Deschane (Battelle Memorial Institute, Pacific Northwest Lab., Richland, Wash.)

Autoradiographs made with cellulose nitrate have been used to locate particles of PuO₂ in a matrix of UO₂, and determine migration and diffusion of PuO₂ in irradiated mechanically mixed oxide fuels. Procedures for making and photographing autoradiographs are described. (auth.)

BASIC PHYSICAL RESEARCH PROGRAM.

The following sections of this report contain information about plutonium: transuranium elements in nature; and self-irradiation damage of plutonium.

BEHAVIOR OF LIQUID Pu-Co-Ce ALLOYS IN CONTACT WITH SODIUM.
I. OUT-OF-PILE FUEL PIN VENTING EXPERIMENTS.
J. C. Clifford (Los Alamos Scientific Lab., Los Alamos, N. Mex.)

To help determine the feasibility of venting fission gases from fuel capsules in a sodium-liquid plutonium alloy fast reactor system, open tantalum capsules containing Pu-Co-Ce fuel have been exposed to flowing sodium at 650°C. These phenomenological experiments have shown that fuel climbs up the walls of annealed tantalum capsules, leading to measurable fuel losses within 1000 to 3000 hours. Cobalt depletion in the climbing fuel (because of Ta-Co intermetallic compound formation at the capsule walls) leaves a plutonium-cerium residue which is more corrosive than the original fuel. The use of a carbide layer on capsule inside surfaces ameliorates these conditions to some, as yet undefined, extent. (auth.)
COMPATIBILITY OF U-Pu-Fz FUEL ALLOYS WITH POTENTIAL CLADDING MATERIALS.

C. M. Walter, J. A. Lahti (Argonne National Lab., Argonne, Ill.)

The cladding materials tested against uranium-plutonium-fizzium alloy can be arranged in order of decreasing compatibility at 650°C as follows: 1) V-20wt% Ti; 2) V-10wt% Ti; 3) molybdenum; 4) 304 stainless steel; 5) Hastelloy-X; and 6) niobium and Nb-1wt%Zr. Increasing plutonium content of the fuel decreases its compatibility with all the cladding materials investigated. The results of this study indicate that the compatibility of a given metal fuel with various claddings may be predictable, on a relative basis, by comparison of the binary phase diagrams of the major elements in the fuel and in the cladding.

10 references. (auth.)

CONTRIBUTION TO THE STUDY OF THE DELTA+GAMMA TRANSFORMATION OF PLUTONIUM.


DENSITIES OF SOME LOW MELTING PLUTONIUM ALLOYS.


The change in fuel density with temperature is an important parameter in nuclear reactor design. For molten fuels, such as are used in LAMPRE-type reactors, it is also necessary to know the volume change on melting. A volumeter employing NaK as a working fluid was used to obtain these data for various plutonium and cerium base alloys over the range 25-800°C. Cerium and several low-melting...
binary cerium alloys were studied with this equipment. Cerium, Ce-Co, Ce-Ni, and Ce-Cu alloys all exhibit an increase in density on melting, while a Ce-Mn alloy expands on melting. The melting temperatures of several of these alloys differ from those reported in the literature, and the compositions of several eutectics in these systems are also reported incorrectly. The densities of unstabilized and gallium-stabilized plutonium and Pu-10 at.%Fe were measured and compared over this temperature range. All these materials expand on freezing. At 675°C, molten unstabilized plutonium is approximately 2% more dense than Pu-1 wt.%Ga alloy. Molten Pu-Fe alloy containing 0.2 wt.%Ga at 435°C is 0.8% less dense than unstabilized alloy. This indicates that there is short-range ordering of plutonium atoms by gallium in the liquid state. The materials containing gallium melted over a 20°C temperature range, while the unstabilized materials melted sharply. Pu-Co-Ce alloys containing 3, 5, 6.2 and 8 g Pu/cm³ were investigated. They all melt in the range 425-442°C and expand on freezing. This expansion increases with increasing plutonium content from 1.3% for the 3 g Pu/cm³ alloy to 3% for the 8 g Pu/cm³ material. Manganese additions to this fuel system are being studied in an attempt to reduce this expansion on freezing. (auth.)

Fabrication and testing techniques for forming about three tons of plutonium alloys at various concentrations are described. Alloys as clad rods were used for various critical experiments and oscillation experiments in test reactors. Two types of experiment were performed. In the first, UPu elements were substituted in the lattices of a uranium-heavy water reactor, a uranium-graphite (cold) reactor, and a hot reactor. Results were precise but require large quantities of fuel - about 700-1000 kg. Oscillation experiments
3381. (Cont'd)

were also made with UPu elements in the same lattices. Only a small amount of fuel (about 7.5 kg) is required.

3382.

A GAMMA-PHASE IN THE PLUTONIUM-MERCURY SYSTEM
A. F. Berndt (Argonne National Lab., Argonne, Ill.)
September 1966.

An investigation of the Pu-Hg system was undertaken to gain a better understanding of the structure of PuHg4. The preparation and powder patterns of PuHg4 are described. A table compares the observed data for the Pu-Hg compound with published intensity data for the γ-phases of compounds having A5B21 stoichiometry.

9 references.

3383.

GROWTH OF SINGLE CRYSTALS OF PLUTONIUM. PROGRESS REPORT JANUARY 1, 1966 to MARCH 30, 1966 (Nuclear Materials and Equipment Corp., Apollo, Pa.)

Studies were confined to delta plutonium this quarter. Zone melting was evaluated as a means of growing single crystals of delta plutonium, and it was concluded that solid state techniques involving recrystallization and grain growth are more favorable.

3384.

GROWTH OF SINGLE CRYSTALS OF PLUTONIUM. PROGRESS REPORT APRIL 1, 1966 TO JUNE 30, 1966
(Nuclear Materials and Equipment Corp., Apollo, Pa.)

Large grains of alpha 2.0 mm x 2.4 mm were grown by subjecting plutonium with an average grain size of .030 mm to a pressure of 25 kilobars at 350°C for 8 hours followed by cooling to room temperature under pressure. A constant temperature oil bath to be used for growing large grains of alpha by an isothermal transformation technique was designed, constructed, tested and installed in a glove box.

21 references. (auth.)
Metallurgy (Cont'd)


A high-temperature ionization chamber for the investigation of the penetration of containers by molten plutonium has been developed. At 1000°C it is capable of detecting 0.1 ug of Pu alloy which has diffused through the grain boundaries to the surface of a tantalum container. (auth.)


Eight clad specimens of (U_{0.8} Pu_{0.2})C_{0.95} were irradiated to two burnup levels (~5 x 10^{20} and 11 x 10^{20} fiss/cc average) at average power levels of 330 to 530 w/cm. A description of the experiments and their results are given. 13 references.


Plutonium may contain a large volume of microcracks which form during the β→α transformation. The volume of microcracks increases with increasing transformation temperature, metal purity, specimen size and applied uniaxial tensile stress. Transformation cycling causes additional microcracking, with concomitant swelling in all directions and extensive surface rumpling. For a given β→α transformation temperature, more damage occurs if the parent beta was formed from alpha rather than gamma. Alpha plutonium containing no microcracks can be produced by quenching the beta phase to -75°C during casting. No
Metallurgy (Cont'd)

3387. (Cont'd)

physical damage occurs during $\gamma \rightarrow \beta$, $\delta \rightarrow \gamma$, or $\delta \rightarrow \beta$ transformation cycling if the metal initially contains no microcracks. 24 references.

3388.

THE PLUTONIUM-AMERICIUM SYSTEM.

The plutonium-americium phase diagram has been determined by micrographic and X-ray diffraction methods. $\delta$-Plutonium and $\beta$-americium form a continuous series of solid solutions, which is stable at room temperature in the composition range from about 6 to 80 at % Americium. No intermediate phases were found. (auth.)

3389.

RADIOGRAPHY OF VERY HIGH ACTIVITY IRRADIATED FUEL ELEMENTS.
Norbert Chassende-Baroz, Lucien Hayet (C.E.N., Saclay, France)

In French.

Using a Rapsodie UO$_2$-PuO$_2$ fuel, characteristics of radiographs made at LÉCI (Saclay) with improved Kodak films are given. 10 references.

3390.

SELF-DIFFUSION STUDIES OF GAMMA-PLUTONIUM.
R. E. Tate, G. R. Edwards (Los Alamos Scientific Lab., Los Alamos, N. Mex.)

The self-diffusion of orthorhombic gamma phase plutonium has been investigated in the temperature range 215-310°C. Roll-bonded couples of plutonium, mainly $^{239}$Pu with one side of the couple enriched with 0.5% $^{238}$Pu as a tracer, were used. $^{238}$Pu penetration-concentration curves for the annealed couples were obtained by sectioning the couples on a lathe and by pulse
height analysis of the alpha activity from representative sections. The preliminary data now available suggest the following equation for the self-diffusion coefficient:

$$D(\text{cm}^2/\text{s}) = 2.1 \times 10^{-5} \exp \left[-\frac{16700}{RT}\right]$$

Data from couples now being processed are being integrated with the preliminary data, and a refined expression for the diffusion coefficient will be determined with a computer least-squares technique. (auth.)

SINTERING OF (U, Pu)O₂ PELLETS.
G. Dean, C. Pelou, D. Beugnies (C.E.N., Fontenay-aux-Roses, France)
CEA-R-2737. May 1965. 32 p.

In addition to the conventional process using UO₂ and PuO₂ as nuclear materials, and hence dependent on the fabrication of sinterable UO₂, a "chamotte" process has been perfected which only requires the use of UO₃ or U₃O₈ and PuO₂ oxides. It is still too early to decide whether this new process will produce a decisive improvement so far as the original target is concerned, i.e. to improve the dimensional reproducibility of sintered pellets. It is however considered that it may constitute a promising process by reason of the following advantages:

- As there is no prior preparation of "sinterable" UO₂, the powders handled are stable in regard to atmospheric oxidation and the operations can be entirely effected in air.

- It resolves in a simple way the problem of producing pre-diffused powders, the basic material necessary for other processes such as compaction-vibration or the obtaining of carbides by carbothermy.

- The process is self-recycling, i.e. the waste from one fabrication can be directly used for the following fabrication.

- The pellets are pressed without bond or lubricant.
Finally, if the question of dimensional reproducibility is not too exacting, a simplified version makes it possible to obtain dense pellets with one single heat treatment. Moreover, the said pellets can be ground.

The products obtained by the two processes have identical characteristics:
- density: between 96 and 99% of the theoretical density,
- micrography: a single phase product with grains approximately 10 to 20 μ, 
- X-rays: one single phase, without variation of composition,
- stoichiometry: the formula of the solid solution varies from $(U, Pu)O_{0.95}$ to $(U, Pu)O_2$ according to the moisture content of the hydrogen, pure or diluted, in the sintering atmosphere. (auth.)

THE SOLID-LIQUID PHASE DIAGRAM FOR THE UO₂-PuO₂ SYSTEM.
W. L. Lyon, W. E. Baily (General Electric Co., Vallecitos Atomic Lab., Pleasanton, Calif.)

The liquidus and solidus were experimentally determined for the UO₂–PuO₂ system by a thermal arrest method. The phase diagram is typical of a binary pair with complete solid solution and without maximum or minimum. A melting point of $2840±20^\circ$C was observed for UO₂, and a melting point of $2390±20^\circ$C was observed for PuO₂. Calculated theoretical liquidus and solidus curves exhibit remarkably good agreement with the experimental values found. 11 references. (auth.)
THERMODYNAMICS OF DILUTE SOLUTIONS OF PLUTONIUM IN LIQUID MAGNESIUM.
Irving Johnson, James B. Knighton, Robert K. Steunenberg (Argonne National Lab., Argonne, Ill.)

The activity coefficient of plutonium in liquid magnesium, over the temperature range 650° to
800°C, was obtained from measurements of the distribution of plutonium between a 50 mole pct
MgCl₂-30 mole pct NaCl-20 mole pct KCl molten-salt mixture and liquid Zn-Mg alloys. For dilute
solutions (0.08 at. pct Pu) the activity coefficient of plutonium was found to vary from 10.1 at 650°C
to 12.2 at 800°C. The activity coefficients of plutonium in dilute liquid solutions of plutonium
in uranium, silver, lanthanum, cerium, and calcium were estimated to be 𝛾Pu(U)=2.8 (1190°C),
𝛾Pu(Ag)=0.18 (1225°C), 𝛾Pu(La)=6.7 (1230°C), 𝛾Pu(Ce)
=5.3 (1225°C), and 𝛾Pu(Ca)=2100 (1150°C). The dis-
tribution data indicate a value of about 0.1 at
800°C for the activity coefficient of PuCl₃ dis-
solved in the above ternary salt mixture.
16 references. (auth.)
COMPARATIVE COST STUDY OF THE PROCESSING OF OXIDE, CARBIDE, AND METAL FAST-BREEDER-REACTOR FUELS BY AQUEOUS, VOLATILITY, AND PYROCHEMICAL METHODS.
M. Levenson, V. G. Trice, Jr., W. J. Mecham (Argonne National Lab., Argonne, Ill.)

An estimate was made of cost differentials among alternative cases in the processing of commercial fast-reactor fuels in facilities to be operated in about 1980. For each of the three alternative fuel types, flowsheets and plant designs were specified, and capital and operating costs were estimated for the three types of processes. A major conclusion drawn from the study is that on-site, non-aqueous, reprocessing plants requiring relatively small amounts of risk capital will probably be capable of processing spent fast-reactor fuel at unit costs very little, if any, higher than those achievable in larger central plants, no matter what process is used in the central plant. 144 references.

TECHNICAL STATUS AND ECONOMIC POSSIBILITIES OF USING PLUTONIUM.
Harvey A. Wagner (The Detroit Edison Co., Detroit, Mich.)

Uses for plutonium in the period between the end of the AEC guaranteed buy-back of plutonium in 1970 and the time when fast reactors will need significant quantities of plutonium in the late 1970's are discussed. The author feels increased use of plutonium must be made in thermal reactors during this period.
Physica

3396.

AN ANALYSIS OF THE FISSION CROSS SECTIONS OF $^{232}$Th, $^{233}$U, $^{234}$U, $^{235}$U, $^{236}$U, $^{237}$Np, $^{238}$U, $^{239}$Pu, $^{240}$Pu, $^{241}$Pu, and $^{242}$Pu FROM 1 keV TO 10 MeV.

William G. Davey (Argonne National Lab., Idaho Falls, Ida.)


The published cross sections of $^{232}$Th, $^{233}$U, $^{234}$U, $^{235}$U, $^{236}$U, $^{237}$Np, $^{238}$U, $^{239}$Pu, $^{240}$Pu, $^{241}$Pu, and $^{242}$Pu from 1 keV to 10 MeV have been carefully studied to select best cross sections for fast reactor analysis. Frequently, the measurement is not of the absolute cross section but of the ratio between the unknown and some reference cross section; thus in comparing measurements, it is important to determine if the same reference data were used in all cases. In this study, emphasis has been placed on determining the reference data used and, where necessary, the published data have been revised to accord with more accurate, currently accepted cross sections. Thus, it is believed that a consistent set of cross-section data has been derived.

Some cross checks have been made by comparing calculations based on the selected cross sections with integral measurements in broad fast-neutron spectra.

The study shows the great importance of the $^{235}$U fission cross section in deriving other cross sections and emphasizes the necessity of re-evaluating nearly all fission cross sections, if it proves necessary to revise the $^{235}$U data.

68 references. (auth.)

3397.

BARRIER PENETRABILITIES AND THE ALPHA-NUCLEUS POTENTIAL.

Gy. Bencze, A. Sandulescu (University of Helsinki, Helsinki, Finland)


Arguments are given that the discrepancy in the absolute value of the theoretical alpha-decay rates can be explained by using the correct alpha-nucleus potential and an accurate method for calculating the barrier penetrabilities. A table is included of the penetrabilities for the $^{238}$Pu-$^{235}$U$_{92}$ alpha transition. 10 references.
3398. \textbf{CALCULATION OF ROTATION GAMMA-VIBRATION INTERACTION IN TRANSURANIC NUCLEI.}
Vojislav Radojevic (University of Belgrade and Boris Kidrič Institute of Nuclear Sciences, Belgrade, Yugoslavia)

Calculations were performed for Th-228, Th-230, U-232 and Pu-238 under the assumption of one γ-vibrational degree of freedom. 12 references.

3399. \textbf{CAPTURE AND FISSION CROSS SECTIONS OF $^{240}$Pu.}
D. H. Byers, B. C. Diven, M. G. Silbert (Los Alamos Scientific Lab., Los Alamos, N. Mex.)

The capture and fission cross sections of $^{240}$Pu are presented in the energy interval 20 eV to 2 MeV. The results were obtained using the technique of neutron cross section measurement by the time-of-flight method employing a nuclear detonation as the pulsed neutron source. (auth.)

3400. \textbf{CHANNEL ANALYSIS OF $^{238}$Pu NEUTRON FISSION.}
P. E. Vorontnikov, S. M. Dubrovina, G. A. Otroshchenko, V. A. Shigin

Results are presented of cross-section and angular-distribution measurements of fragments from fission of $^{238}$Pu by 50-1400 keV neutrons. The data obtained were analyzed for the purpose of separating individual fission channels with different values of K, the projection of the angular momentum of the fissioning nucleus on its symmetry axis. It is shown that the sequence of occurrence of channels with different K is as follows: 1/2$, 3/2$, 1/2+. The values of the thresholds, fission widths, and fission-barrier curvatures were determined for each channel. The results obtained, together with data on photofission of $^{239}$Pu, show that the shape of the fission barrier is nearly parabolic and permit us to obtain the cross section for dipole photoabsorption for the nucleus $^{239}$Pu. 22 references. (auth.)
CHARACTERISTICS OF FISSION OF Th$^{232}$ BY DEUTERONS AND THE DEPENDENCE OF THE FRAGMENT KINETIC ENERGY ON THE EXCITATION ENERGY OF THE FISSIONING NUCLEI.
Yu. A. Selitskii, S. M. Solov'ev, V. P. Eismont

Various fission properties—the mass distribution of the fragments and the mean values and dispersions of the kinetic energy distributions of fragments with equal masses—have been determined by measuring the kinetic energies of equal fragments at two deuteron energies, and are compared with each other. The results of this investigation and of the analysis of other papers on the dependence of the kinetic energies of fragments with given masses on the excitation of the fissioning nucleus are discussed in terms of nuclear shells in the fragments for two independent types of fission. This is in agreement with experimental data in the medium-energy region. Data are included for Pu$^{237}$ and Pu$^{242}$. 24 references. (auth.)

FAST TRANSIENTS IN LIQUID PLUTONIUM.
B. M. Carmichael (Los Alamos Scientific Lab., Los Alamos, N. Mex.)
LA-DC-7748. 1966. 9 p.

Fast transients in clean fuel were studied using a code, TRN, which is being developed specifically for applications to molten plutonium capsules. The code calculates distributed values for the power, temperature, pressure, and motion of the fuel in capsules for given reactivity input functions. Results for a $100/\text{sec}$ ramp in a LAMPRE-type core containing clean fuel, and for the Molten Plutonium Burn-up Experiment at ramps of $500, \$100, \$10, \text{and} \$1/\text{sec}$ are given.
FISSION CROSS SECTION OF $^{239}$Pu, 20 eV to 20 MeV.
Edward R. Shunk, W. K. Brown, R. LaBauve
(Los Alamos Scientific Lab., Los Alamos, N. Mex.)

The fission cross section of $^{239}$Pu has been measured by time-of-flight using neutrons from an underground nuclear explosion. The Petrel event in June, 1965 at the Nevada Test Site provided an extremely high neutron flux from 20 eV to 2 MeV. The neutron flux was determined from simultaneous measurement of $^6$Li(n, α) for $E < 100$ keV, and $^{235}$U(n,f) for $E > 10$ keV. Backgrounds were measured by recording signals from blank backing foils.

FISSION-fragment ENERGY-CORRELATION MEASUREMENTS FOR THE THERMAL-NEUTRON FISSION OF $^{239}$Pu AND $^{241}$Pu.
J. N. Neiler, F. J. Walter, H. W. Schmitt (Oak Ridge National Lab., Oak Ridge, Tenn.)
September 23, 1966.

Fission-fragment mass and energy distributions and mass-versus-energy correlations have been obtained for $^{239}$Pu and $^{241}$Pu thermal-neutron-induced fission. Silicon surface-barrier detectors were used in energy-correlation measurements; absolute fragment energies were obtained by means of a recently developed mass-dependent energy calibration. Average total fragment kinetic energies before neutron emission are found to be 177.7 ± 1.8 MeV for $^{239}$Pu and 179.6 ± 1.8 MeV for $^{241}$Pu. Detailed experimental results are given and compared with those of other experiments. Observed fine structure in the fragment mass distribution and in the average total fragment kinetic energy as a function of mass is correlated with the energetically preferred even-even nucleon configurations in the fragments. New determinations of the root-mean-square width of the total-kinetic-energy distribution as a function of fragment mass show structure which also appears to be correlated with the energetically preferred even-fragment configurations. Fission neutron and gamma-ray data of other experiments are used with the new fragment kinetic energies presented here to examine the total energy balance for fission for the two cases studied. A comparison of the two mass distributions shows the heavy-fragment groups almost superimposed;
Physics (Cont'd)

3404. (Cont'd) the light-fragment groups are separated almost uniformly by 2 amu. 24 references. (auth.)


The total kinetic energies of symmetric fragments from fission of Pu^{239} by thermal neutrons and by neutrons transmitted through a samarium filter have been measured with the help of two surface-barrier semiconductor counters. The kinetic energies proved to be equal within the experimental errors (+2 MeV). The yield of fragments in the symmetric region was about 30% lower for resonance-neutron fission. The dependence of the total kinetic energy of the fragments and of the dispersion of this quantity on the mass ratio was determined for thermal-neutron fission of Pu^{239}. (auth.)


Measurements of the energy spectra of isomeric gamma rays from the neutron fission of U^{235} and Pu^{239} at a number of time intervals between 50 and 600 usec showed six prominent gamma rays for both cases of fission. The intensities and half-lives for these gamma rays indicate that there are three fission-fragment isomers, each giving rise to a pair of gamma rays in cascade. The energies of the cascade gamma rays and the half-life for each
isomer are: 1260, 450 keV (80 μsec), 850, 250 keV (54 μsec), and 990, 710 keV (32 μsec). Absolute gamma-ray intensities were obtained by measuring the integral number of gamma rays above a fixed bias energy as a function of time and normalizing these data at 10 msec to the absolute intensities derived from the theoretical work by Griffin and the data of Fisher and Engle. The intensities of delayed gamma rays from photofission of U^{235}, Pu^{239}, and U^{238} were also measured at early times, starting at 2 μsec after fission. An intense short-lived component with an apparent half-life of a few μsec was observed for all three cases of photofission. (auth.)

ISOTHERMAL DROP CALORIMETER FOR ALPHA ACTIVE, PYROPHORIC MATERIALS.
Howard Savage (Argonne National Lab., Argonne, Ill.)

A copper block drop calorimeter to measure the heat content of intensely alpha active materials has been constructed. The furnace, calorimeter, and aluminum isothermal jacket are contained within an inert atmosphere glovebox. This permits the use of unencapsulated materials without exposing personnel to an alpha contamination hazard. The inert atmosphere also permits measurements to be made on materials which are pyrophoric in air. The apparatus is equipped with a suppressed range recorder to monitor continuously the resistance thermometer signal with a resolution of 0.4 cal/μV. Electrical calibration of the calorimeter yielded a precision of 0.08% in the thermometer and recording system. The heat content of a sample of Calorimetry Conference sapphire obtained from the National Bureau of Standards was measured, and the results deviated from the Bureau's reported data a maximum of 1.0% at 1008°K. The useful range of the apparatus extends from 306 to about 1450°K. 12 references. (auth.)
M, N, AND O SUBSHELL CONVERSION COEFFICIENTS IN $^{228}\text{Th}$ AND $^{240}\text{Pu}$.

A. V. Ramayya, B. van Nooijen, S. R. Amtey, J. H. Hamilton
(Vanderbilt University, Nashville, Tenn.)

The M, N, and O subshell conversion coefficients of the 57.9-keV, $2^+\rightarrow 0^+$ transition in $^{228}\text{Th}$ and the M and N subshell conversion coefficients of the 42.88-keV, $2^+\rightarrow 0^+$ transition in $^{240}\text{Pu}$ have been measured with an iron-free double-focusing spectrometer. In the case of $^{228}\text{Th}$, conversion coefficients obtained for the $M_1$, $M_2$, $M_3$, $M_4+M_5$, $N_2$, $N_3$, $O_2$, and $O_3$ shells are 0.89±0.22, 19.1±1.5, 17.8±1.3, 0.55±0.34, 5.16±0.46, 4.97±0.45, 1.33±0.22, and 1.23±0.20, respectively. For the transition in $^{240}\text{Pu}$, the conversion coefficients of the $M_1$, $M_2$, $M_3$, $N_1$, $N_2$, $N_3$ and $N_4+N_5$ are 5.6±2.1, 107.2±7.5, 97.8±7.0, 2.9±0.8, 20.3±2.9, 28.7±2.9, and 2.3±1.0, respectively. Results from the M subshells are (15±8)% to (85±45)% higher than Rose's theoretical values after correcting for screening according to the semiempirical method of Chu and Perlman. The M-shell conversion-coefficient ratios for these E2 transitions agree with Rose's unscreened values within our experimental errors. This agreement suggests that the corrections due to screening for the different members in a particular shell are approximately the same. (auth.)

NEUTRON CROSS SECTIONS FOR $^{244}\text{Cm}$ AND $^{238}\text{Pu}$.

A. Prince (General Electric Co., Nuclear Materials and Propulsion Operation, Cincinnati, Ohio)

The neutron cross sections of $^{244}\text{Cm}$ and $^{238}\text{Pu}$ have been calculated from thermal energies to 1 MeV. The calculations have been cross checked in most cases by as many as three independent theoretical approaches, giving a high degree of confidence in the results. It appears desirable, however, to verify these calculations by performing cross section experiments at an early date. An estimation of the minimum critical mass based on these cross sections, has been made in this study to determine the extent of the criticality problem in the use of $^{244}\text{Cm}$ and $^{238}\text{Pu}$ as a heat source. 52 references. (auth.)
NEUTRON MULTIPLICATION DETERMINATION OF PLUTONIUM-238 OXIDE.
R. A. Wolfe, J. B. Kahle (Monsanto Research Corp., Mound Lab., Miamisburg, Ohio)

A neutron multiplication experiment was performed with 21 storage capsules containing high isotopic analysis plutonium dioxide. Each storage capsule contained approximately 80 grams of the plutonium-238 isotope. The capsules were assembled in a heterogeneous array spaced four inches center-to-center and measurements were made independently in a water and in an air medium. The results indicated that no neutron multiplication was detectable in the fully assembled array in a water medium. However, a slight neutron multiplication of 1.29 existed in the fully assembled array in an air medium. (auth.)

NEUTRON STUDY OF PLUTONIUM IN THERMAL NEUTRON REACTORS.
(Commissariat a l'Energie Atomique, Paris, France)

The main purpose of this contract was to develop the methods required for studying uranium-plutonium lattices moderated with graphite and heavy water. The accurate measurement of the Pu/U ratio of a metal fuel, especially in the case of alloys with small Pu content (less than 1000 ppm), proved to be a major difficulty. A double dilution determination using a tracer with U233 and Pu242 base was chosen as most accurate at a satisfactory operating speed (at least 100 analyses per year). The spontaneous fissions of Pu240 have proved remarkably efficient for checking the preparation and finished rods. Special apparatus was designed for oscillation tests. The theoretical study of the neutron thermalization made it possible to design calculation methods which are exceptionally simple and well adapted to the systematic study of U/Pu lattices (spectrum indices, fine structures, evolution problems) which are now used on a large scale. It is anticipated that by the end of 1967 a complete stock of information on the use of plutonium in heavy-water lattices and in graphite lattices up to 450°C and for exposures up to 5000 MWD/t will be available.
NEW MEASUREMENTS IN PLUTONIUM L EMISSION SPECTRUM USING AN ELECTRON PROBE MICRO-ANALYZER.

Jean-Louis Bobin, Jean Despres (C.E.N., Saclay, France)

CEA-R 2921, April 1966. 32 p.

Further studies by means of an electron-probe micro-analyser, allowed the authors to set up a larger plutonium X ray spectrum table. Measurements of plutonium L_I and L_{III} levels excitation potentials were achieved. Apparatus performance data (such as spectrograph sensibility, resolving power and accuracy) is found in the appendix. 14 references.

NON-ELASTIC INTERACTION CROSS SECTIONS OF NEUTRONS WITH 7Li, 12C, 14N, 27Al, 56Fe, Cu, Pb, 235U, 238U, and 239Pu.

Yu. G. Degtyarev


The neutron transmission coefficients were measured in spherical samples of these materials in inverse geometry. A detector which is insensitive to inelastically scattered neutrons was used to measure the flux of neutrons from an external monokinetic source, first with the source inside the spherical sample, and then with the sample removed. Since elastically scattered neutrons are automatically compensated for in this method, the transmission coefficient of the sphere is a measure of the inelastic interaction cross section only.

OLBUSTAP: A BURNUP PROGRAM FOR HEAVY WATER MODERATED UNIFORM LATTICES.

R. Gopalan, T. A. Subramanian (Atomic Energy Establishment, Trombay, India)


This report describes the program OLBUSTAP (Open Lattice Burnup Studies of Thorium and Plutonium) developed for heavy water moderated, uniform lattices. It is in Fortran language for the computer CDC-3600. A system of burnup equations is solved numerically and the resulting isotopic compositions are used for calculating various lattice parameters by a conventional recipe. (auth.)
3416. PARAMETERS OF Pu-239 LEVELS.
Yu. V. Ryabov, Yung-chang Wang, E. Dermendzhiev, Pei-shu Chang
(Joint Institute for Nuclear Research, Dubna, USSR)
JINR-P-2713, 1966. 18 p.

In Russian.

3417. THE PROBABILITY OF ISOMERISM IN THE DECAY OF HEAVY
EXCITED NUCLEI.
V. P. Zommer, A. I. Prokof'ev
Soviet Journal of Nuclear Physics, Vol.3, No.3:289-92,
September 1966.

A method of calculating the isomerism ratio is
described for an arbitrary nuclear reaction on
heavy nuclei, under the condition that the com-
petitive processes are fission and emission of
neutrons and γ quanta. Theoretical results are
obtained for the reaction Pu$^{242}(d,2n)$Am$^{242}$ and are
in good agreement with the experimental data.
It follows from the comparison with experiment
that the energy of the isomer state in the Am$^{242}$
nucleus, decaying through spontaneous fission
with period 14 msec, is 2.8 MeV. (auth.)

3418. REACTOR NOISE EXPERIMENTS IN SMALL PLUTONIUM SYSTEMS.
G. M. Hess (Battelle Memorial Institute, Pacific North-
west Lab., Richland, Wash.)

The noise method was investigated as a measure
of neutron lifetimes, on the assumption that a
comparison between experimental and calculated
lifetimes is a more sensitive test of a cross
section set than is a comparison of calculated
and experimental critical masses. The noise
method excels near criticality because of the
large multiplication. It is concluded that
an on-line system can be developed which will
measure to within one per cent the break
frequency of an epithermal plutonium system from
delayed critical to approximately seven dollars
subcritical. 27 references.
Physics (Cont'd)


An extension of the Iridium technique for the measurement of $^{240}$Pu resonance capture described by Jakeman and Maunders is suggested: in the original method the resonances above 2 eV introduced considerable unwanted contributions to the total reaction rate, which limited the accuracy of the method. This paper suggests a modification of the technique which minimizes these contributions by using the sandwich method with carefully optimised foil thickness and a filter of Cd-Ag alloy to eliminate contributions from the 5.36 eV resonance. The results of measurements made using this technique could be analysed without the aid of a digital computer. (auth.)


Experiments have been made to measure the shielding effect of U-238 resonances on epicadmium fissions in U-235 and Pu-239 and on epicadmium activations of Mn-55. The experimental method is based on the comparison of detector reaction rates in cadmium-covered rods of depleted uranium and lead/U-235 placed in the center channel of a subcritical assembly. The measured shielding factors have been compared with the predictions of a simple theoretical model and reasonably good agreement is obtained. (auth.)
AN ASSESSMENT OF HIGH TEMPERATURE POWER REACTORS.

C. A. Rennie

The status of the high temperature reactor is reviewed by first discussing development costs, engineering design, fuel technology, full cycle costs, capital costs and generating costs. The present position seems very favorable but it is clear that no reactor system can survive in the long term unless it is truly competitive both now and in the future. The utilization of natural uranium resources, the effect of increases in the price of uranium, the actual resources available and the nuclear power potential and requirements are discussed. The arguments put forward show that the high temperature reactor is well suited to meet future requirements and, after a brief discussion of uranium enrichment capacity and helium supplies, the place of the high temperature reactor in nuclear power programs is discussed, and the conclusion drawn that the high temperature reactor should have an important role to play both in the short term and long term future of nuclear power. Data on plutonium are included. 20 references. (auth.)

COMMERCIAL ASPECTS OF FUEL PROCESSING.

W. J. Mecham, A. A. Jonke

The Commercial Plutonium Fuel Conference held last March is summarized. Evaluations have been published of methods for measuring the chemical concentration and isotopic composition of U and Pu in various forms and materials. The comparative costs of processing fast reactor fuels in facilities to be in operation in about 1980 are tabulated. Recent papers on the long range economics of nuclear fuel costs, especially with regard to plutonium, are summarized. 36 references.
The production of pellets for the EBR-II irradiation rods is continuing. The greater proportion of the coprecipitated and master blended fuel pellets have been produced. The production of the mechanically mixed fuel is underway and the necessary sintering trials continue. The apparent lack of correlation in sintering results between trial batches and production lots is most likely due to the difference in mixing techniques. Control of density is a persistent problem at the levels and limits considered necessary at this stage. Yields and material losses encountered to date are tabulated.

Fuel rod welding parameters have been established. The characteristics of all fuel rods to be irradiated are described in detail. The capsule sodium filling setup has been completed and the design setup is given.

The metallography of the preliminary UO₂-clad powder specimens heated to 1250°C for 24 hours indicates no reaction zone.

The thermal expansion work by x-ray diffractometer is currently being performed. Results of the initial runs are shown.

The difference in PuO₂-UO₂ melting points under argon and N₂-6 v/o H₂ appears to be quite significant and details are presented. The work with the setup for melting tungsten encapsulated specimens is continuing.

The heavy duty mixer has been received and has been modified to minimize criticality hazards. The necessary box design changes to permit incorporation of the mixer in a glove box setup have been made and work commenced. For correlation purposes the significant extrusion work which used the 25 ton press will now be rerun with the new mixer and 75 ton press combinations. (auth.)
EVALUATION OF EXISTING REACTORS FOR THE IRRADIATION OF LIQUID PLUTONIUM FAST REACTOR FUELS.
G. L. Ragan (Los Alamos Scientific Lab., Los Alamos, N. Mex.)

The potential of the 5-megawatt Omega West Reactor for providing useful irradiation data on liquid plutonium fuels is explored.

FAST REACTOR DEVELOPMENT IN THE UNITED STATES: LOS ALAMOS MOLTEN PLUTONIUM PROGRAM.
William H. Hannum (Los Alamos Scientific Lab., Los Alamos, N. Mex.)
LA-DC-7550. 1965. 9 p.

A core fueled with molten plutonium (LAMPRE I) has been operated successfully for over two years. A versatile ternary fuel Pu-Co-Ce, has been selected which appears adequate for use in a well-designed central-station fast breeder core. The reliable containment of this fuel appears to be feasible, and the MPBE will demonstrate this capability. In the present concepts, a tantalum container is used. The associated parasitic neutron losses are tolerable in a hard spectrum, if the plutonium content of the fuel is properly selected as a compromise between heat transfer and breeding potential.

The major potential advantages for molten plutonium in a near-term application (e.g., second generation, central-station fast breeders) lie in safety and fuel element lifetime and performance. The main incentive for the program, however, lies less in its near-term capabilities, which are significant, than in the fact that such a near term use would constitute a base for realistic and potentially spectacular further advances. (auth.)

FUEL AND FERTILE MATERIALS.

This article reviews recent work on solubility of Nb in liquid Pu, attempts to grow single crystals of high-purity αPu, the physical properties of several Pu-base intermetallic com-
Reactor Technology (Cont'd)

3427. (Cont'd) pounds, the development of U-Pu-Ti, U-Pu-Zr, and U-Pu-Fe alloys, the preparation of UO₂-PuO₂ pellets, the preparation of PuC and (U,Pu)₃C, the preparation of (U,Pu)(OCN), and the preparation of (U,Pu)S solid solutions and UN-PuN. 77 references.

3428. FUEL CYCLES FOR POWER REACTORS.
E. Schröder, J. H. Blomstrand, H. Bruneder

In addition to the high thermal efficiency and the excellent neutron economy of high temperature reactors, the freedom to utilize plutonium or U²³⁵ as fissile material and U²³⁸ or thorium as fertile material in all possible combinations, is an additional advantage of this reactor type. The paper summarizes the results of survey calculations on fuel cycle performance and cost for these combinations and various fuel management schemes, ranging from batch loadings over part load system to continuous charge/discharge operations, the latter on-load. Under the present economic conditions the reactor would be best utilized with a feed breed cycle with continuous charge/discharge of the feed. Assuming equal charges for fissile materials, this cycle gives fuel costs of 0.095 d/kWh with U²³⁵ feed and thorium breed and 0.112 d/kWh with plutonium feed. When reprocessing facilities are available fuel cycle costs can be further reduced for reprocessing cost below £200/kg heavy metal. 11 references. (auth.)

3429. FUEL ELEMENT FABRICATION FOR THE DRAGON REACTOR EXPERIMENT.
M. S. T. Price, J. R. G. Gough, G. W. Horsley

This discussion includes data comparing the costs of PuC₂ and UC₂ coated particle fuels. 36 references.
HAS ATOMIC ENERGY GOTTEN INTO ITS STRIDE?
H. de Laboulaye (C.E.A., France)
In French.
A general discussion of the use of plutonium and enriched uranium fuels in the United States, the United Kingdom, and in France is included.

THE IRRADIATION OF LIQUID PLUTONIUM FUELS IN A THERMAL REACTOR.
Richard L. Cubitt, George L. Ragan, Donald C. Kirkpatrick (Los Alamos Scientific Lab., Los Alamos, N. Mex.)
In-pile test loops for irradiating liquid plutonium fuel alloys have been designed and operated in the Omega West Reactor. These loops provide a sodium environment for test fuel capsules at temperatures corresponding to those expected for IMBFR. Seven fuel capsules have been irradiated with up to 2% burnup of plutonium. Irradiation of two fuel specimens is currently in progress. The report describes the design, construction and operation of the irradiation loops and handling of the irradiated test fuel capsules. (auth.)

IRRADIATION TESTING OF PuO₂-UO₂ FUELS. QUARTERLY PROGRESS REPORT, JANUARY 1-MARCH 31, 1966.
(Nuclear Materials and Equipment Corp., Apollo, Pa.)
Photomicrographs of irradiated PuO₂-UO₂ fuels, prepared by five different procedures, are presented and discussed. A plot of fission gas release as a function of the fuel volume showing the effects of grain growth indicates that homogeneous and heterogeneous substoichiometric fuels behave similarly. There appears to be little difference in the fission gas release from co-precipitated and substoichiometric mechanically-blended mixtures operating at the same temperature. Mechanically-blended stoichiometric oxides showed significant grain growth in comparison to the other specimens, but very little difference in the amount of fission gas released. (N.S.A., Vol.20, Abs:No.29805)
The present paper represents the concept of new type mercury-boiling fused salt fast reactor "WARS". The fuel consists of: $^{239}$PuCl$_3$ - fissionable material, $^{238}$UCl$_3$-fertile material and NaCl and/or KCl-diluent. Mercury metallic was chosen as primary cooling agent. Mercury liquid is pumped into the fused salt giving dispersed system. Then it is heated to the boiling temperature. The boiling heat of mercury is a decisive element of heat balance in this type of reactor. Outlet mercury vapor is moderately superheated.

The simplified characteristics of such a reactor are the following:
- Reactor power: 1000 MW/t
- Power density in fused salt, average: 400 KW/liter of salt
- Reactor core volume: 10,500 liters
- Outlet temperature of mercury vapor: 740°C
- Inlet temperature of mercury liquid: 356°C
- Pressure in reactor core: 40 ata
- Fissionable material inventory $^{239}$Pu: 1000 kg
- Fertile material inventory $^{238}$U: 4000 kg.

10 references. (auth.)
Reactor Technology (Cont'd)

3434. A NEW-FUEL CONFIGURATION TO INCREASE PLUTONIUM VALUE.
D. E. Deonigi, J. H. Nail ( Battelle Memorial Institute,
Pacific Northwest Lab., Richland, Wash.)

The present paper considers the burnup characteristics and the value effects of concentrating plutonium in the center of each fuel rod of a water moderated reactor. Such a concentrated fuel configuration makes possible (1) an adjustable cross section of plutonium, (2) an improvement in the alpha of Pu$^{239}$, and (3) some use of Pu$^{242}$ as a fissile fuel. (auth.)

3435. ORGDP FUEL REPROCESSING STUDIES SUMMARY PROGRESS REPORT JULY THROUGH DECEMBER, 1965.
S. H. Smiley, D. C. Brater, J. H. Pashley ( Union
Carbide Corp., Nuclear Div., Oak Ridge Gaseous
Diffusion Plant, Oak Ridge, Tenn.)

The Oak Ridge Gaseous Diffusion Plant Technical Division is participating with Argonne and Oak Ridge National Laboratories in studies of a group of processes aimed at purifying and recovering valuable uranium and plutonium from spent nuclear reactor fuels by volatilization as the respective hexafluorides. The ORGDP portion of the program includes two main phases: 1. preparation of conceptual plant studies with concomitant definition of problem areas associated with the process technology; and 2. component development, including scale-up and testing of crucial process equipment and auxiliaries. The current report is the second in a series of progress reports to be issued semiannually. Topics covered include status of the conceptual study of the multipurpose fluid-bed plant for high enrichment fuels; status of low enrichment fuel flow sheet studies; composition of irradiated fuels; theoretical aspects of uranium hexafluoride-plutonium hexafluoride-fission product separation by distillation; design of a reactor for producing bromine pentafluoride from bromine trifluoride; sizing of condenser systems for collection of bromine pentafluoride-uranium hexafluoride mixtures; sodium fluoride fluid-bed sorber criticality calculations; corrosion testing and related studies; process and reactor studies, including cold semiworks plant
design; outlet gas filter testing; product purification sorption-desorption system using batch sodium fluoride pellet traps; connector testing; status of valve selection; peripheral and diaphragm compressor evaluation; and reactor fabrication procedures.

(auth.)

3436. POWER REACTOR CORE STUDIES. 
P. Marien 

General thermal design formulas—and related computer programs—which can be used to optimize the core design of various reactors, using different fuels and coolants are presented. These formulas are applied to the study of high temperature gas cooled power reactors and the feasibility is demonstrated to design such reactors with acceptable core temperatures, for different fuel cycles and fuel management schemes, under the operating conditions selected for the Dragon power reactor assessment studies. The incorporation of the thermally optimized fuel cell geometries in fuel elements, with due consideration of corrosion, fission product migration, core stability, fuel handling and manufacturing problems is discussed. Data on plutonium are included. (auth.)

3437. QUARTERLY STATUS REPORT ON ADVANCED REACTOR TECHNOLOGY (ART) FOR PERIOD ENDING APRIL 30, 1966. 

The molten plutonium program is being modified since the AEC decided not to continue with the Fast Reactor Core Test Facility. This report contains information about "mixed core" fuel which would be a molten Pu driver core used for irradiation of possible Liquid Metal Fast Breeder Reactor fuels. New data is also presented on the MPBE core originally planned as the first experiment in the FRCTF. Supporting research has been done on plutonium oxides, uranium-plutonium carbides, preparation of plutonium monocarbide by arc melting, aluminum additions for reprocessing Pu-Ce-Co fuel alloys, phase transition properties, and solubilities of selected elements in liquid plutonium.
Reactor Technology (Cont'd)


The Molten Plutonium Program was terminated by the AEC and the phasing out of the program is described. The results of studies of irradiated fuels and containers in the OWAREX experiment on liquid Pu fuels are reported. The results of thermocycling tests of Pu-Co-Ce fuel alloys are given. The compressibility of liquid Pu and thermal conductivity of Pu metal are discussed. 18 references.


In French.

We review the power densities necessary for the production of electrical energy by thermoelectric or thermionic methods from the radioactive decay of (isotopic) sources. Taking into account the thickness of the radioactive capsule, only $^{210}\text{Po}$ and $^{242}\text{Cm}$ are useful for direct thermionic conversion. For thermoelectric conversion, the alpha emitters $^{238}\text{Pu}$ and $^{244}\text{Cm}$, the beta emitters $^{170}\text{Tm}$, $^{90}\text{Sn}$, $^{144}\text{Ce}$, and $^{137}\text{Cs}$ are useful. Some prototype developments are suggested.


In a study of the best radioisotope for use as the heat source material in such a system it is pointed out that while Po-210 is advantageous from a cost and availability standpoint and has a more acceptable weight penalty, Pu-238 provides growth potential for longer duration flights.
Reactor Technology (Cont'd)

3441. REACTOR DEVELOPMENT AND TECHNOLOGY.
AEC, 1965.

The following sections of this report contain information about plutonium: plutonium lattice experiments, p. 17-19; measurements of fast-neutron cross-sections, p. 23-25; fluidized-bed volatility process, p. 50-52; properties of plutonium hexafluoride, p. 53; liquid metal-salt extraction systems, p. 53-54; fissiochemical processes, p. 54-55; Doppler effect measurements, p. 58-59; metallic fuels, p. 68-69; U-Pu-Pz fuel-clad compatibility, p. 69-71; ceramic fuels, p. 71-74; fast ceramic reactor program, p. 74-75; molten fuels, p. 75.

3442. SHORT TERM DYNAMIC EFFECTS UPON THE UTILIZATION OF PLUTONIUM AS A REACTOR FUEL.
James R. Sheff (Battelle Memorial Institute, Pacific Northwest Lab., Richland, Wash.)

With the availability in the near future of reactor fuels in sizable quantities other than U-235, it seemed necessary to estimate quantitatively the effects produced upon several of the characteristic parameters of the reactor associated with its control and inherent safety. In particular, calculations have been made of the average delayed neutron fractions, the mean neutron lifetime, the prompt neutron decay constant, and a parameter representative of the control strength required for fuel burnup. These calculations were done for two specific reactor types, BWR and PWR, for two basic levels of plutonium enrichment in uranium, and for two reactor fueling modes. The results were obtained as a function of goal fuel exposure. (auth.)

3443. SODIUM-COOLED REACTORS, FAST CERAMIC REACTOR DEVELOPMENT PROGRAM. QUARTERLY REPORT NO. 16, AUGUST-OCTOBER 1965.
C. E. Breizy, comp. (General Electric Co., Advanced Products Operation, San Jose, Calif.)

The following sections of this report contain information about plutonium: sodium-fuel compatibility,
Reactor Technology (Cont'd)

3443. (Cont'd) p. 3.4-3.7; axially restrained fuel experiments, p. 4.4-4.12; center temperature limits, p. 5.1-5.7; plutonium migration, p. 5.8-5.10; fuel properties - melting point studies, p. 5.10-5.11, and fast flux irradiation of fuel, p. 6.1-6.7.

3444. SOME FUTURE REQUIREMENTS FOR LARGE-SCALE ISOTOPE IRRADIATION IN REACTORS.

Some of the isotopes suited for isotopic power generation have to be obtained through neutron irradiation, and some aspects of the possible future irradiation needs are dealt with. The irradiation charges for the different isotopes are compared and it is shown that there are large differences in the contribution of the irradiation cost to the total cost per initial thermal watt. Consequently, a large scale isotope irradiation program could lead to a better overall neutron economy depending upon the isotope chosen for power generation. Data are included for Pu-238, Pu-239, Pu-240, Pu-241, and Pu-242. (auth.)

3445. STUDY OF THE FEASIBILITY OF USING THORIUM IN FAST POWER REACTORS.

The results of studying the possibilities of using $^{233}$U and thorium in a reactor system with breeding are described. It is shown that the most promising from the point of view of fuel doubling time is the combined use of $^{233}$U-Th and $^{239}$Pu-$^{238}$U in a mixed fast-reactor fuel cycle on condition that the thorium is placed in the shields and $^{233}$U, $^{239}$Pu and $^{238}$U in the cores.

The burning and regeneration of $^{233}$U and $^{239}$Pu in a mixed fuel cycle is effected in such a
way that the ratio of the quantity of each
remains constant. The doubling time of a
system of fast reactors operating on a mixed
fuel cycle mode is substantially less than
the doubling time of reactors using only \(^{233}\text{U}\)
and thorium. The main raw material in the
mixed fuel cycle is thorium.

A method of obtaining isotopically pure
\(^{233}\text{U}\) with a \(^{232}\text{U}\) content \(\sim 10^{-4}\) per cent is
given. (auth.)

USE OF PLUTONIUM AS A FUEL IN NUCLEAR REACTORS.
QUARTERLY REPORT NO. 7.
(Societe Belge pour l'Industrie Nucleaire,
Brussels, Belgium)
EURAE C-1552, December 1965. 79 p.

A. Development of ceramic fuels

Various methods of preparing the mixed
powders required for the program were
considered during this quarter. The
sintering of powders, the sintering of
nodules and melting may be mentioned.
The last method is based on the in-
duction of high frequency current in
the hot oxide.

The heterogeneous plutonium distribution,
arising from the enrichment of fine grade
alone in the powder fuel, appears to be
due chiefly to resonance effects during
the vibrational tamping. The shape and
roughness of the coarse grade grains also
play a very important part; from the re-
sults available to date, the melted powders
seem the most satisfactory.

Positioning of the fuel inside the slug
was attempted using brazed elements.
However, brazing is complicated by the
gettering of the powders.

Fabrication of the Vulcain and Venus/Vulcain
elements was completed.

The influence of transversal PuO\(_2\) segre-
gation on the overall count rate of a section
and the count rate of two opposed counters
was examined in detail for the Vulcain fuels. The most unfavorable case was found for a rod with a local count rate excess of 17% and a ratio of 1.28 between the count rates of two diametrically opposed detectors. According to the most pessimistic hypotheses, this bar would have 62% surplus heat flux along a peripheral width of the order of 3 mm.

B. Irradiation

B.1. Hydraulic conveyor

The ceramographic and contact radiographic examinations made of the fuel samples irradiated during the first series of irradiations did not show any structural change during irradiation.

The second series on the hydraulic conveyor was started on 2nd September 1965. One set of 3 samples was irradiated at low power, and a second set at higher power. On the basis of data currently to hand, the maximum power values were probably of the order of 450 to 700 W/cm respectively.

B.2. Boiling water capsule

Three samples of UO₂-PuO₂ fuel have been undergoing irradiation in the BR-2 reactor since 10 August 1965. The maximum power during the first cycle is of the order of 100 W/cm. The mean burn-up ratios reached during this cycle are of the order of 600 MWd/t.

B.3. BR-3/Vulcain

The selection of the samples for the BR-3/VN reactor is representative of the present stage of development of the fabrication method. An analysis of the influence of heterogeneities in the plutonium distribution for the most pessimistic hypotheses showed that this factor had an unfavorable effect on fuel performance, in the present state of uncertainty surrounding the effect of hot points on the burn-out margin. For this reason the vibrated rods could not, in the cases referred to here, be placed at the few most affected points of the fuel subassembly.
C. Neutron physics of the UO$_2$-PuO$_2$-H$_2$O lattices

The study of the 15.4 and 22 mm pitch lattices carried out on the Thetis lattice was completed. A critical approximation with cylindrical and square geometries, and variable poisoning measurement were made for these two lattices. The measuring methods used were: source ejection and pulsed measurement. The results of these two measurements are in agreement. The slope and linearity for the function $a = f$ (number of rods) make it quite easy to estimate the prompt critical mass. 11 references. (auth.)

USE OF PLUTONIUM AS A FUEL IN NUCLEAR REACTORS.
QUARTERLY REPORT NO. 8, OCTOBER 1-DECEMBER 31, 1965.
(Societe Belge Pour l'Industrie Nucleaire, Brussels, Belgium)
EURAEC-1593. February 1966. 120 p.

Progress on the development of ceramic fuels is reported. A study was made of the transversal distribution of the heat flux in rods containing pellets which were assumed to be eccentric, in order to compare the effect of transversal PuO$_2$ segregation in an element prepared by the vibration method, with the plutonium being introduced in the fine grade alone, and by the pelletization method. The progress of the irradiation of UO$_2$-PuO$_2$ fuel samples is reported. Work done on neutron study of UO$_2$-PuO$_2$-H$_2$O lattices is described. 42 references.