MonS-124
copy 31A

[redacted]

Contract No. W-35-058-eng-71

***

PROJECT COUNCIL-INFORMATION MEETING

Clinton, March 18-20, 1946

Compiled by E. Rabinowitch

June 6, 1946

DECLASSIFIED
Classification Changed To:
By Authority Of: T 10-1160
By:
ORNL Libraries Division
Y-12 Technical Library
Document Reference Section
Building 9711-1

Date Received: June 6, 1946 Date Issued: June 7, 1946

This document contains information affecting [redacted] of the [redacted] of the Espionage Act, USC 50, 31 and [redacted] transmission or the revelation of [redacted] in any manner to an [redacted] per[redacted].

[redacted]

[redacted]

Contract No. W-7401-eng-37

* *

PROJECT COUNCIL-INFORMATION MEETING

Clinton, March 18-20, 1946

* *

Table of Contents

March 18. Metallurgy

1. Properties of High Temperature Pile Structural Materials
2. The Preparation of Alclad Uranium-Aluminum Alloy
3. Beryllium Reductions
4. Studies of Beryllium Oxide Ceramic Bodies
5. Thermal Conductivity of Beryllium Oxide Bodies
6. Preparation of Beryllium Nitride, Be_3N_2
7. Metallurgy of Be
8. Studies of the Metallurgy of Beryllium

March 19. Symposium on Ion Exchange

9. The Kinetics of Ion-Exchange Adsorption
10. The Use of Ion Exchangers as a Physico-Chemical Tool
11. The Separation of Fission Products
12. Extraction and Decontamination of Plutonium
13. The Status of the Application of Ion-Exchange to Plutonium Separation
14. The Rare Earths and Pu Separation
15. Separation of the Rare Earths
16. Use of IR-1 Columns to Separate Element 95 from Rare Earths
17. Ion-Exchange Work involving Actinium
18. Adsorption Behavior of Protactinium on Amberlite IR-4 and Amberlite IR-1
19. Remote Control Methods in Column Adsorption Operation
20. New Engineering Designs for Adsorption Processes
21. Analytical Application of Ion Exchange to the Separation of Thorium from Uranium
22. Separation of Pu and Other Substances from Biological Materials
23. Uranium Resonance Absorption

[redacted]

Table of Contents (continued)

24. Cross-Section Measurements with Pile Oscillator
25. Temperature Characteristics of the Clinton Pile
26. Studies of Capture Gamma Rays
27. Some Measurements with the 180° Beta-Ray Spectrometer
28. The Double Crystal Neutron Spectrometer
29. Magnetic Moment of the Neutron
30. Preliminary Measurements of C^{14} Radiation with an Ion Chamber and Vibrating Diaphragm Electrometer
31. The Half-Life of C^{14}
32. Cross Sections for Fission Neutrons
33. Mass Assignments and Enrichments
34. Fissionability Studies: Po^{210} and Np^{239}
35. Recent Results of Heavy Isotope Work
36. Cyclotron Bombardments of U^{233}
37. Production of Actinium from Radium; Properties of Actinium and Its Daughters

March 19. Chemistry

38. Contributions to Radio-Chemistry of the 104 Day Y^{88}
39. Summary of Fission Product Investigations at HEW
40. Deuteron and Helium Ion Bombardment of Pa^{231}
41. Color Production in Lithium Fluoride by Radiation
42. Behavior of Pu in the Presence and Absence of Citrate Solutions
43. Potentials of Pu Couples
44. Recent Work on Redox Process
45. Action of Electron Beams and X-Rays on Highly Purified Water
46. A System for the Separation of Uranium from Thorium using Organic Complexing Agents in Solvent Extraction
47. Activity Coefficients of Plutonium
48. Xe Poisoning and Pile Stability
49. Migration Area in Heterogeneous Piles
50. Proposed Experimental Facilities for the Heterogeneous Pile
51. Pile Calculations
52. High Pressure Water as a Heat Transfer Medium in Nuclear Power Plants
53. Resonance Energy Piles
54. Pile Operations at High Temperatures

May 20, 1946

March 18. Metallurgy

1. Properties of High Temperature Pile Structural Materials

C. A. Hutchison, Jr.
(Chicago)

The projected high temperature pile is to contain UO_2 as fuel and beryllia as moderator.

Beryllia has now been produced by hot pressing with a density of 3.00 g/cc, which is close to the theoretical x-ray value. Other physical constants of this material are as follows:

Thermal conductivity at 1000°C - 0.08 cal $^{o}C^{-1}$ x cm^{-1} x sec^{-1}

Young's modulus - 315 x 10^{10} dyne x cm^{-2}

Compressive strength about 1 x 10^{10} dyne x cm^{-2}

Thermal expansion coefficient at 1000°C - 8 x 10^{-6} $^{o}C^{-1}$

Hardness - about that of fused alumina

Vapor pressure at 1500°C about 10^{-8} mm Hg.

Because of the very high thermal conductivity, the temperature gradient in the rods will be only 20°C at 100,000 Kw. The thermal stresses are decreased by the high value of thermal conductivity more than they are increased by the (also very high) value of Young's modulus.

The chemical properties of this material were unknown before. Treated with steam at 1500°C, it lost 10.5% of its weight in 13 hours. Pure cottony beryllia was distilled away, indicating the existence of a volatile hydroxide. The material is gray in color. The origin of the color is unknown; it is not due to carbon impurity. Upon heating, the material becomes white with an increase rather than a decrease in mass.

U_3O_8 is used in high temperature pressing together with beryllia to form the fuel rods. U_3O_8 is converted to UO_2 during this procedure; it is not reoxidized again upon cooling.

The volatilization of uranium from the mixed oxide bodies at 1500°C is illustrated by figure 1.

[redacted]

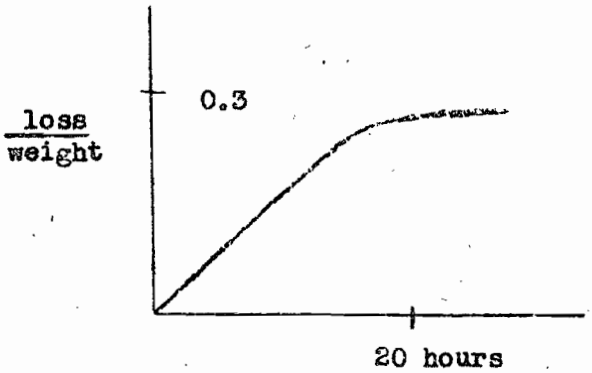


Fig. 1

In vacuo, no loss of uranium occurs after the composition UO_2 has been reached. In air, the uranium continues to evaporate, probably by intermediate formation of volatile UO_3. In hydrogen atmosphere, the uranium loss is less than 4 per cent. We have investigated how much oxygen can be permitted without causing considerable losses of uranium. In an atmosphere containing 0.5 per cent oxygen, the loss was 40 per cent in 30 hours; with 0.14 per cent O_2, the loss was reduced to 10 per cent in 200 hours. In pure oxygen, 80 per cent uranium were evaporated at 1500°C, and condensed in the cooler part of the apparatus as an oxide of the approximate composition U_2O_5. This evaporation could be perhaps used in the reprocessing of material for the separation of uranium from fission products.

* *

2. The Preparation of Alclad Uranium-Aluminum Alloy

by L. H. Grenell (Battelle)

The cladding of Al-U alloys on all sides by hot rolling, to give a density of 15 mg. of U per sq. cm. at 1-mm. thickness, required application of a much thicker cladding layer than is done commercially where the surface layer is from 5 to 10% of the thickness. Regular cladding procedure, for which the cover sheet is wrapped around the leading edge of the core, and a method using this procedure with a "picture frame" surrounding the edges of the core were tried. The first method did not give complete edge closure, but the picture-frame method appears satisfactory.

Tests were run using initial core thicknesses from 0.080" to 0.375", with no great difference in bonding. A number of cleaning methods were tried, but the standard scratch-brushing method seemed most satisfactory. Core and coating thicknesses remain in very nearly the same relation as the starting thicknesses. Alloys with 11 and 15% U were clad successfully. Distribution of the U in the core was more uniform with cross rolling of the core and in the 15% alloy.

[redacted]

3. Beryllium Reductions

T. Magel, M.I.T. (presented by A. R. Kaufman)

(1) Some work has been done at M.I.T. on closed bomb reductions of $BeCl_2$ with Ca metal (and sulfur booster) on an 18 gram (Be) scale. Yields averaged about 75 to 80 per cent coherent, well-consolidated metal. The calcium content of the beryllium varied between 0.5 and 10 per cent. Overall results on $BeCl_2$ reductions indicated no advantage over the reduction of BeF_2. Moreover, since there is no longer a satisfactory source of $BeCl_2$ this work has been discontinued. (2) A few bomb reductions of mixtures of $BeCl_2$ and UCl_4 have been carried out in an attempt to prepare directly the compound UBe_9. Technique similar to that used in $BeCl_2$ reductions was employed with resulting yields of about 75 to 80 per cent, one half of which was in the form of a well consolidated mass and the remainder in the form of a coarse powder. (3) High-purity Be metal has been prepared by high temperature (1700°C), high-vacuum distillation of the Be formed by the reaction of a stoichemetric mixture of BeO and Zr metal (introduced as ZrH_2). Yields of densely deposited Be (on a Ta condenser) of 80 per cent and better have been obtained in about 8 hours. In the present apparatus about 80 grams of Be can be prepared per run.

* *

4. Studies of Beryllium Oxide Ceramic Bodies

H. Z. Schofield
(Battelle Memorial Institute)

Specimens, dust-pressed from a mixture of high-fire grade beryllium oxide and UO_2, disintegrated in air at temperature near "red heat" owing to oxidation of the UO_2 to U_3O_8 and the attendant volume expansion. The use of U_3O_8 in the mixture prevented this disintegration, but a large part of the uranium oxide was lost by volatilization during firing at high temperatures.

Specimens of beryllium oxide, 90%, UO_2, 10%, were fired at 3300°F. without disintegration when an atmosphere of argon was maintained at all temperatures below 2500°F. The fired bodies were porous, however, when high-fire grade beryllium oxide was used. Apparent nonporosity in the fired body was obtained by using fluorescent-grade beryllium oxide.

In the work on nonporous surface films, or "glazes", for the porous $BeO-UO_2$ bodies, some promise was shown when a thin coating, composed of equal parts of BeO and MgO, was applied to the body before firing. Some promise also was shown when a film of silica was deposited on the specimen from the vapor phase in an atmosphere of water vapor and silicon tetrachloride, or by saturating the body with a hot, concentrated solution of beryllium sulfate and drying, before firing.

[redacted]

[redacted]

5. Thermal Conductivity of Beryllium Oxide Bodies

H. R. Nelson (Battelle Memorial Institute)

A knowledge of the thermal conductivity of BeO at elevated temperatures is important in the design of the mixed oxide pile. Previously reported measurements on low-density bodies have shown that BeO has a surprisingly high conductivity which varies with the density of the specimen. New results on high-density material confirm the conclusion that BeO is more metallic than ceramic in its thermal-conductivity properties. Results are summarized below.

Thermal Conductivity in Watt $cm.^{-1}$ $^{o}C.^{-1}$

Temperature °C	BeO Bodies of Two Densities		Pure Iron
	2.16 gm./cm.^{3}	2.87 gm./cm.^{3}	
200	0.57	0.81	0.60
400	0.36	0.54	0.46
600	0.26	0.42	0.32
800	0.20		
1000	0.16		
1200	0.14		

Apparatus has been constructed to permit remote-control measurements on radioactive specimens. This equipment will be used to determine the magnitude of possible changes in the conductivity of a mixed oxide body as a result of neutron radiation.

* *

6. Preparation of Beryllium Nitride, Be_3N_2

T. Magel, M.I.T. (presented by A. R. Kaufman)

Be_3N_2 is being prepared at M.I.T. on a 200 gram scale by passing ammonia gas over Clifton flake beryllium metal at 900°C. Only beryllium metal which passes a 28 mesh screen is used; coarser material is difficult to nitride completely under present operating conditions. The metal is placed on a thin molybdenum sheet inside a two inch diameter quartz tube. After evacuating to remove air, a stream of ammonia is passed over the metal while it is heated to 900°C. About 60 hours is necessary for complete conversion to nitride. Shorter heating periods give partial conversion but such material can be completely converted to nitride by additional treatment at 900°C. Analysis of product showed 50.4 per cent N, and 50.0 per cent Be; theoretical is 50.8 per cent N, and 49.2 per cent Be. First batches of M.I.T. nitride were inadvertently contaminated with about 100 to 200 ppm. of titanium and zirconium. The source of contamination has now been removed.

[redacted]

7. Metallurgy of Be

A. R. Kaufman (M.I.T)

Crude Brush beryllium can be refined by melting in an open pot and then casting in air. This process which removes most of the slag in the metal, leads to about a 3 to 5% loss of Be due to oxidation. Vacuum castings up to 25 pounds in weight can be made in a 12" quartz tube furnace using this refined metal.

Extrusions of Be have been carried out using billets up to $6\frac{1}{2}$" in diameter. Rods up to 3" diameter and down to $\frac{1}{2}$" diameter have been made with a four hole die being used in the latter case. A disc or cone of soft iron is placed at the front of the billet to fill up the space at the periphery of the die and thus facilitate the flow of the iron jacket on the billet. Be billets encased in heavy iron have been satisfactorily forged at temperatures as low as 1100°F. Fabrication operations so far carried out indicate that Be crystals in certain orientations are extraordinarily resistant to deformation.

Tensile tests of extruded Be indicate that the optimum combination of strength and ductility, in the direction of extrusion at least, is obtained by extruding at a low temperature (1300°F) and then annealing at about 1380°F.

Distilled Be has been obtained in quantities of several pounds. There is no evidence, as yet, that this material is more ductile than the ordinary metal.

* *

8. Studies of the Metallurgy of Beryllium

H. W. Russell (Battelle Memorial Institute)

It has been demonstrated that large castings of beryllium can be prepared without melting in vacuum. Metal freed from slag was melted in a silicon-carbide (Tercod) crucible without liquid flux. A blanket of argon minimized oxidation. Gassiness, resulting in porous castings, occurred occasionally and erratically, but was controlled in part by bubbling chlorine through the melt. Pure beryllium castings tend to coarse grain and hot cracking; the addition of 2 to 3% of aluminum gives great improvement, and has been the regular practice.

Graphite molds are satisfactory only if all restriction on the normal shrinkage can be avoided. Sand molds of zircon or magnesia give fair results, and are usable where graphite will not permit the necessary shrinkage. Hollow cylinders are readily cast centrifugally in steel molds rotated at a speed to give about 75 times gravity. A melt of 75 pounds of beryllium, containing 3% aluminum, was ladled into a centrifugal mold, forming a casting 52 inches in diameter with a wall thickness of 1-3/4 inches.

There has been contradictory evidence in regard to a possible transformation in beryllium somewhere between 500°C. and 750°C. Measurements of the thermoelectric power of extruded beryllium 0.15 inch in diameter against platinum in the range from 350°C. to 750°C. fail to supply evidence of the existence of a transformation. In this temperature range, the thermoelectric power, in microvolts per degree C., can be represented by

$$de/dt = 1.20 + 2.10t$$

The electrical resistance was found to vary smoothly from 500°C. to 800°C., with no indication of a transformation. The extruded rod used had a resistivity at zero of $\rho_0 = 4.1 \times 10^{-6}$ ohm cm.

Thermal analysis by the method of C. S. Smith (Trans. A.I.M.E., Vol. 137, pp. 236-244, 1940) showed no evidence of a transformation on heating from 500°C. up to 825°C., and back down to 530°C.

Jaeger and Zanstra (Proc. Roy. Soc., Amsterdam, Vol. 36, p. 636, 1933) have reported a new phase, detectable by X-ray diffraction, in beryllium quenched from 630°C. This procedure has been repeated, and, in addition, X-ray diffraction data have been secured in a hot camera at 550°C., 700°C., and 800°C. No lines due to a new beryllium phase could be detected.

* *

March 19. Symposium on Ion Exchange

arranged by J. Schubert (Chicago)

9. The Kinetics of Ion-Exchange Adsorption

L. S. Myers, Jr.

In order to avoid the tedious, time consuming and often wasteful series of experiments necessary to determine the optimum conditions for separating substances such as the rare earths and fission products by column ionic exchange processes, it is desirable that means be found to predict the behavior of any given ion in a column from the results of a few simple batch experiments. In order to accomplish this a knowledge of the mechanism of adsorption and the rate controlling factors must be obtained.

During an adsorption process, a reaction such as is shown by the following equation occurs:

$$nA^+ + mBR = nAR + mB^+ .$$

A^+ and B^+ represent the cation which is adsorbed and the cation initially on the resin, respectively, and BR and AR represent the initial resin and the resin after A^+ is adsorbed, respectively. The equilibrium constant for such a reaction is the ordinary mass action expression in which the activities in the solid phase are represented by the mole fractions.

[redacted]

$$K = \frac{[B^+]^m \quad N_{AR}^n}{[A^+]^n \quad N_{BR}^m} .$$

In such a reaction there are at least three processes, any one of which may be rate controlling. These are: (1) diffusion of A^+ and its anion into the immediate vicinity of BR; (2) the reaction of A^+ with BR to give the products B^+ and AR; (3) the diffusion of B^+ and its anion away from the immediate vicinity of AR and into the solution.

Under conditions of constant concentrations of A^+, B^+ and BR (i.e. tracer concentrations of A^+ and large volumes of solution) very simple rate equations may be derived for each of these processes. If process (1) or (3) is the slowest, the rate of adsorption will be controlled by the laws of diffusion, and the diffusion equation is applicable.

$$\frac{AR}{AR_\infty} = 1 - \frac{6}{\pi^2} \sum_{n=1}^{\infty} \exp. \left(- \frac{D\pi^2 n^2 t}{r^2} \right).$$

AR and AR_∞ are the amounts of A^+ adsorbed at any time and at equilibrium, n is an integer, D the diffusion constant in $cm^2 sec^{-1}$, t the time, and r the radius of the resin particles, which are assumed to be spherical.

If process (2) is the slowest, the rate of adsorption will be controlled by the laws of mass action and the mass action rate equation is applicable.

$$\frac{AR}{AR_\infty} = 1 - \exp. \left(-k_2 B^m t \right).$$

The symbols have the same significance as above, with k_2 being the specific reaction rate constant for the reverse reaction, and B^m the concentration of B ion raised to the appropriate power.

Batch experiments using Amberlite IR-1 resin show that when B^+ is present in low concentrations, the mass action process is the slowest, and hence is rate controlling. As the concentration of B^+ increases, the mass action process increases in velocity until it eventually becomes faster than one of the diffusion processes. Under these conditions the diffusion process becomes rate controlling. The concentration at which this transition occurs varies with the valence of the ions involved. When the ion initially on the resin is monovalent and the adsorbate is monovalent, this transition concentration is of the order of 0.01 M. When the adsorbate is divalent it is of the order of 0.1 M, when trivalent, about 0.3 M.

The values of the equilibrium constant, the mass action rate constant and the diffusion constant show considerable variation with valence. When the resin initially contains a monovalent ion, these constants are of the order of 0.1-3, 10 $M^{-1} sec^{-1}$, and 3×10^{-6} cm^2 sec^{-1} respectively when the adsorbate is monovalent; 3-5, 2 $M^{-2} sec^{-1}$, and 0.4×10^{-6} when it is divalent; and 10-100, 0.2 M^{-3} sec^{-1}, and 0.01×10^{-6} when trivalent. Addition of a

complexing agent to yttrium increases the diffusion constant somewhat. Comparison of several ions of the same valence show that the heavier ions are adsorbed more strongly than lighter ions and that their presence results in smaller diffusion constants. The mass action equilibrium expression is inadequate for trivalent ions.

The mass transfer theory permits prediction of the behavior of an ion during column adsorption if two constants are known, provided that the rate of adsorption is controlled by the mass action process. These constants are known as k and k', and they may be related to batch constants as follows:

$$k = \frac{\text{moles adsorbate per cc of bed at equilibrium}}{\text{moles adsorbate per cc of solution at equilibrium}}$$

$$= \frac{AR_{\infty}\ /g}{A_{\infty}\ /ml} \text{ (Bed density)}$$

$$k' = k_2 B^m.$$

Comparisons of values of k' and k obtained by batch experiment and column studies show that k can be predicted fairly accurately by batch studies, but that k' is apparently a function of the contact time. As the contact time is increased the value of k' obtained from column operation approaches the value obtained from batch studies.

References:

Adamson, A. W., CN-1859, Schubert, J., CN-1873, Adamson, A. W., Mon-C-51 Myers, L. S., Jr., Mon-C-52, Adamson, A. W., PPR, Volume 16-A, Chapters XVII, XVIII.

* *

10. The Use of Ion Exchangers as a Physico-Chemical Tool

Jack Schubert

The procedures by which ion exchangers, utilizing only radiochemical concentrations of elements, can be employed to study fundamental physico-chemical problems is described. Some of the problems which can be profitably studied by the ion exchange method are:

1. Qualitative and quantitative measurements of the dissociation of complex ions.
2. Measurement of the activity coefficients of tracer cations.
3. Detection and behavior of colloids.
4. Qualitative determination of the state of an element in solution.
5. Determination of the valence and relative basicity of cations.

11. The Separation of Fission Products

E. R. Tompkins, J.X. Khym, G. W. Parker, W. E. Cohn and Associates

Reported by W. E. Cohn

The general technique of ion-exchange is shown to be applicable to the problem of separating radioactive fission species from each other in the carrier-free state. This is accomplished by adsorbing all cationic species, after extraction of uranium with ether, on Amberlite IR-1; the anionic species (Ru, Te), are not adsorbed and may be collected in the effluent.

Separation of the adsorbed cations is effected by specific elution as complexed ions with organic acids and their salts. These may be used in various ways, but a practical procedure used on the curie scale has been: (1) elution of Zr and Cb with 1/2% oxalic acid; (2) elution of Pu with 5-7% oxalic acid; (3) elution of Cs with 0.5 N H_2SO_4; (4) elution of rare earths in the order Y, Il, Nd, Pr, Ce by 5% citrate at pH varied from 2.7 to 3.3; (5) elution of Sr and Ba, in that order, by 5% citrate at pH 5-6.

Ion-exchange methods, based on re-adsorption of the active species after decomplexing by acidification, have been developed for purifying the tri- and divalent species of radiochemical and chemical contaminants, for concentration and for metathesis. Since the technique is simple and uniquely adaptable to remote-control handling, the preparation of large amounts of very pure carrier-free fission species is relatively easily achieved. The process is economical since most major fission species may be recovered from a single starting solution; furthermore, the entire process, including concentration, purification, etc., is accomplished by the repetition of a single cycle.

Covered by D-1823 S-2865

* *

12. Extraction and Decontamination of Plutonium

J. A. Ayres

An extraction and decontamination procedure has been developed and tested thoroughly on a pilot plant scale, using one kilogram of uranium metal (one regulation X slug) per run.

The procedure consists of a three step process using adsorption columns. The plutonium is adsorbed on the first column and then eluted with sodium acid sulphate. The uranium passes on into the effluent and is thus separated from the plutonium. Additional carrier is added to the eluate which is then diluted and made alkaline. The supernatant liquid is drained off and the carrier is dissolved in dilute sulfuric acid. The solution is diluted, complexed with 7-iodo-8-hydroxyquinoline-5-sulfonic acid, neutralized to a pH of 5.0-5.8 and then run through a second Amberlite-IR-1 column in the

Ames work Chicago covering

[redacted]

sodium form. The effluent which contains all the plutonium is acidified (pH $\sim$2.5) and run through a third Amberlite IR-1 column. The plutonium is adsorbed on the column. The column is washed with dilute sulfuric acid and dilute oxalic acid, and the plutonium is then eluted with saturated oxalic acid solution.

The plutonium solution is heated with concentrated sulfuric acid to drive off or destroy the oxalic acid. The plutonium may then be dissolved in a few milliliters of acid and concentrated or diluted to any desired volume.

The procedure possesses the following advantages:

1. It is safe since it uses no volatile, explosive, or inflammable solvents.

2. It is simple since it uses only standard procedures which have already been extensively developed on a commercial scale.

3. It is well suited to remote control procedures since it contains no steps requiring close or intricate control and no filtrations or centrifugations.

4. It affords a means of fractionation of the various fission products, providing, if desired, a relatively simple method for obtaining active fission products to a high degree of purity and free of carrier.

5. It is rapid.

6. It gives high yields (>99%) of plutonium and very high decontamination factors.

The only activity other than UX remaining with the Pu is Zr (and Cb daughter). This may be removed very efficiently by any one of several simple specific procedures to give any decontamination factor desired.

* *

13. The Status of the Application of Ion-Exchange to Plutonium Separation

J. A. Swartout

The ion-exchange procedure for separating plutonium from uranyl nitrate dissolver solutions which was recommended for further testing on a Clinton Plant scale in 1944 consisted of the following steps. Upon passage of a 10% UNH dissolver solution through a bed of the cation-exchanger, Amberlite IR-1, adsorption of all of the plutonium, part of the uranium, and most of the fission products occurred. All of the adsorbed uranium was then desorbed along with essentially none of the plutonium by flowing 0.25 M sulfuric

S-1823 Cover

acid through the bed. A wash with 0.04 M oxalic acid effectively removed the fission products, chiefly Zr and Cb, which would otherwise be desorbed by the subsequent plutonium elutriant consisting of 0.4 M oxalic acid. Following the removal of plutonium the adsorbent bed was washed free from residual fission activities by a 1.25 M sodium bisulfate wash. In this procedure a yield of plutonium of greater than 97%, decontamination factors of about 10^2, and a concentration of plutonium by 5 to 10 fold depending upon specific operating conditions were achieved in laboratory experiments with "tracer" amounts of plutonium and the fission elements.

In order to demonstrate the effectiveness of such a process for plant operation it was necessary to show (1) that equally favorable results would be obtained when operating at the levels of plutonium and fission element concentrations which would prevail at Hanford, (2) that reproducible performance would continue over the number of consecutive cycles required for practical plant operation, (3) that mechanical degradation of the adsorbant would not occur during this continued usage, (4) that the adsorptive properties of the adsorbent would be unaffected by the levels of radiation expected at Hanford, and (5) that remote control operation was feasible. Efficient performance at Hanford concentrations was indicated by laboratory experiments (CN-3347). The reproducibility of the process and mechanical stability of the resin were proved in series of semi-works scale experiments (CN-3346). Tests by Burton's group at Chicago demonstrated the stability of the adsorbent under radiation. On the basis of these data equipment to be operated by remote control for testing of the procedure in the Clinton Pilot Plant was designed. Further work on this alternate process was curtailed due to (1) the successful operation of the precipitation process and (2) the failure to achieve sufficient decontamination to permit use of the process as a substitute for the entire precipitation process.

Since the future application of ion-exchange processes for the separation of plutonium depends upon the development of methods giving higher decontamination factors, work in progress or envisioned is aimed at this end. Modifications to effect higher decontamination by an "all-adsorption" process include (1) repetitive cation-exchange cycles, (2) coupling with anion-exchange, (3) use of "tailor-made" adsorbents with specific adsorptive properties for certain ions, (4) the use of complexing agents to preferentially prevent the adsorption of either plutonium of fission elements, and (5) the use of elutriants which are specific for certain ion species. Potential developments to give more efficient and more economical operation include (1) utilization of the entire adsorptive capacity of the adsorbent by employing three or more columns in a tandem system, (2) investigation of new, higher capacity adsorbents, (3) improvements ~~in instruments~~ in instrumentation and (4) the perfection of unique systems such as the counter-current slurry method of Monet.

* *

[redacted]

[redacted]

14. The Rare Earths and Pu Separation

E. M. Gladrow

A set of optimum conditions for the separation of the individual rare earths from each other has been obtained by using an Amberlite IR-1 column and eluting with ammonia buffered citric acid solution. Principle work was done using mixtures of Nd and Pr. The factors investigated were pH, flow rate, composition of starting material, diameter of column, length of column, and the weight of starting sample. The citric acid concentration was kept at 5% by weight and the particle size of the Amberlite used was 40-60 mesh throughout all of the experiments. Best separation was obtained when the pH was 2.55; flow rate 1.5 cm^3/cm^2 cross section/minute, and weight of sample 0.25 - 0.50 grams of rare earth oxide per cm^2 cross section. The diameter of the column had no effect. Increasing the length of column gave increased separation but also increased the length of time required for the experiment. Two passes through a column employing 50% Nd 50% Pr in original mixture yielded 40% of the Nd with a purity > 99.8%. Higher yields than this have been obtained using mixtures of Nd-Sm. The method can also be applied to preparing other rare earths.

MRB

Spedding.

15. Separation of the Rare Earths

Jack E. Powell

The separation of rare earths by means of 5% citric acid solution and Amberlite IR-1 depends chiefly upon the pH of the solution, the bed length, the sample size and the flow rate. The time factor is also important when a separation is to be carried out on a large scale, since lowering the pH, increasing the column length, decreasing the sample size and lowering the flow rate in order to obtain maximum separation all greatly increase the length of time required to process a given quantity of rare earth mixture.

Spedding

The cost of the separation process, using 5% citrate under any set of conditions, is prohibitive. However, by using a citric acid concentration of 0.5% and a pH of about 4, an excellent separation was obtained and the cost of citric acid was reduced by a factor of ten.

Resinous Products Chemical Co. have discontinued the manufacture of IR-1 in favor of the more highly polymerized IR-100. Amberlite IR-100 was compared to IR-1 and was found to be satisfactory for use in the separation process.

For the separation of samarium from neodymium and other impurities and the separation of neodymium from praseodymium on a 50 gram scale, an IR-100 bed 8 feet long and 4 inches in diameter is recommended with 0.5% citrate solution at a pH of 4.0 and a flow rate of 60 cc/min (a linear flow rate of 0.75 cm/min).

[redacted]

Samples rich in samarium were obtained by the sodium amalgam extraction process of Marsh. Such samples contained europium and ytterbium along with traces of cerium and lanthanum. These impurities with the exception of europium were readily removed from the bulk of the samarium by one pass through an IR-100 column (see conditions recommended above).

* *

16. Use of IR-1 Columns to Separate Element 95 from Rare Earths

Chicago Work

S.T. Thompson and G.T. Seaborg

Introduction: Elements 95 and 96, because of their similarity to the rare earths are normally present in rare earth fractions when the usual chemical separations of the elements are made. In general they are more easily complexed than lanthanum and more basic than yttrium. Oxidation or reduction of element 95 from its trivalent state in aqueous solution is so difficult that a method of separation from rare earths based on these properties has not been developed as yet. With element 96 oxidation or reduction does not seem to be possible. Obviously the separation of elements 95 and 96 from rare earths is not easily accomplished.

Summary of Experimental Work: The adsorption of element 96 on columns packed with IR-1 resin followed by its elution with ammonium citrate, according to the method reported by Cohn and his co-workers at Clinton, provides a good separation of 95 from some of the rare earths such as lanthanum and yttrium. However, our work has shown it to be not readily separated from element 61, which is a fission product produced in significant proportions. In this latter case a decontamination factor of the order of two for the separation of element 61 from element 95 in a six foot column was obtained. In our studies we found Y to be much more rapidly eluted and element 61 slightly more rapidly eluted than 95. 61 and 95 were indeed very close in elution characteristics. On the other hand, La and Ce were more slowly eluted than 95. Thus 95 has characteristics placing it in the region of Nd and probably between Nd and 61 with respect to its behavior in this system. For the latter reason we have attempted to find other methods for the separation of element 95 from the rare earth fission products. We have been successful in finding a group separation of the rare earths from elements 95 and 96 (and this separation probably holds for the whole trivalent actinide series starting with actinium). This method involves the use of fluosilicic acid which is a stronger complexing agent for the 95 and 96 than it is for the rare earths. Thus it is possible to precipitate lanthanum or cerium fluorides or fluosilicates rather completely, leaving nearly all of the 95 and 96 in a solution containing a high concentration of fluosilicate. It is interesting that we did not obtain a good separation of 95 and 96 from the rare earth fission products by elution from an IR-1 column with a mixture of HCl and fluosilicic acid. The conditions were such that the elution took place at a suitable rate but both groups came off together. Consideration of these data together with that obtained with citrate might suggest that elements 95 and 96 are more strongly adsorbed on IR-1 than the rare earths

[redacted]

and that the former are also more strongly complexed by citrate than are the rare earths. Since according to Peterson, actinium in the IR-1 citrate system is even more slowly eluted than is La, the positions of 95 leads us to suggest that the trivalent actinide series is analogous to the rare earths in behavior in the IR-1 citrate system, the difference between individuals being of about the same magnitude, and in the same direction with increasing atomic number. Thus element 96 should be more rapidly eluted than element 61 although the separation of these two might not be highly efficient.

Column: 7 ft. length
0.7 to 1 cm diam.
packed with 40-60 mesh IR_1

Conditions used in Separation

Activities adsorbed on column from 0.1 N HCl solution. HCl washed out with water. 5% $Nt/_4$ Citrate of pH 2.75 passed thru column, a large number of fractions collected usually of 5-10 ml vol. Each fraction analyzed for total α and B^- activity. Comparison of 95 in separate experiments with La, Y, and 61 was done in short column of length 12" and diameter 1 cm to determine the relative positions of the elements with respect to each other.

* *

17. Ion-exchange Work involving Actinium

S. Peterson — Chicago?

Almost complete separation of tracer quantities of actinium and lanthanum is obtained by eluting the adsorbed mixture from IR-1 cation exchange resin with a 0.25 M citrate solution of pH near 3. To separate actinium from larger quantities of lanthanum the column must be sufficiently large to adsorb the mixture on a small fraction of the resin bed. A column 1.1 cm. diameter and 55 cm. long is sufficient for 0.20 g. lanthanum.

Tracer experiments have demonstrated that eleven column volumes of 0.25 M mono-ammonium citrate would quantitatively elute Ac from IR-1, but would elute only one part in 10^5 of Ra. This was used to separate the Ac^{227} product from pile-bombarded Ra^{226}. The behavior of the radium series activities showed that Pb follows Ac in the IR-1 process.

* *

18. Adsorption Behavior of Protactinium on Amberlite IR-4 and Amberlite IR-1

Q. Van Winkle (presented by D. Osborne)

Performed at Chicago.

Batch adsorption studies on fluoride solutions of protactinium were carried with Amberlite IR-4 For HF concentrations between 0.1 and 2.7 N and HNO_3 concentrations between 0.2 and 0.9 N, it was observed that [redacted]

$$\frac{X}{1 - X} \quad \frac{(NO_3^-)^2}{(F^-)}$$

was approximately constant, where X is the fraction of protactinium adsorbed. This seems to indicate that an equilibrium between singly charged and doubly charged anions such as -

$$F^- + PaO_2F_2^- \rightleftharpoons PaO_2F_3^{=}$$

controls the degree of adsorption of Pa on Amberlite IR-4.

Column tests showed that the Pa-fluoride complex is readily eluted from Amberlite IR-4 with solutions which are 0.3 to 1.0 N in HNO_3. Separation of Pa from Fe and Ti have been achieved by elution with 0.3 N HNO_3.

Column tests with Amberlite IR-1 showed that the use of 0.1% H_2O_2 in 1 N HCl as an elutant would effect a separation of Ti from Pa. Also 0.5% oxalic acid was found to be a useful eluting agent in separating Pa from Zr and Cb.

* *

19. Remote Control Methods in Column Adsorption Operation

G. W. Parker

(no abstract received)

* *

20. New Engineering Designs for Adsorption Processes

G. P. Monet

The new engineering design is a continuous process where a resin slurry is contacted in a column in parallel or counter-current flow with a feed containing the adsorbate. The resin slurry is separated from the exhausted feed, washed clean with water, and cycled as a slurry to the next column. In a similar fashion, the product is desorbed from the resin, and the resin is regenerated before recycling to the first column as fresh adsorbent.

* *

21. Analytical Application of Ion Exchange to the Separation of Thorium from Uranium

R. W. Bane

Amberlite Resin IR-1 has been employed to separate the strongly adsorbed Th^{+4} ion from the weakly adsorbed UO_2^{++} ion. The addition of hydroxylamine to the influent liquid and the use of dilute hydrochloric

acid to elute adsorbed UO_2 makes it possible to achieve a quantitative separation.

The method was developed specifically for the determination of Thorium in Uranium - Thorium alloys containing 1% to 10% thorium.

A dilute nitric acid (< .1 N) solution of thorium and uranium containing hydroxylamine is passed thru a column of Amberlite IR-1 resin. Adsorbed uranium is eluted by passing thru dilute hydrochloric acid. The thorium is eluted by passing thru a sodium bisulfate solution. The eluate containing the thorium is made ammoniacal and the thorium hydroxide filtered off. The precipitate is dissolved with dilute hydrochloric acid, and the thorium precipitated as the oxalate, ignited and weighed as the oxide.

Recoveries of thorium from control samples is better than 99%.

* *

22. Separation of Pu and Other Substances from Biological Materials

E. R. Russell and J. Schubert

Plutonium is extracted from liter quantities of urine with a resinous cation exchanger. The method involves adsorbing Pu from acidified urine, washing away part of the organic and inorganic impurities adsorbed on the resin with dilute hydrochloric acid. The plutonium is eluted with strong HCl, the eluate evaporated and diluted with distilled water. The adsorption elution step is repeated. This time the Pu is contained in a small volume ($\sim$50 ml) so that the solution is evaporated and mounted directly on a large platinum disc and counted. Yields have averaged 85%.

The adsorption and elution of purines and other urinary constituents have been studied by Dr. S. Schwartz and associates. They find that adenine sulfate and guanine are quantitatively adsorbed by the cation exchange resin IR-100 H from neutral solution. They are desorbed by 6 N HCl on 1 N NaOH. Uric acid is poorly adsorbed by IR-100 H, as is a mixture of pentose nucleatides.

Coproporphyin in solutions weakly acid or alkaline is adsorbed by the anion exchanger, IR-4 and is desorbed by 2 N HCl or a mixture of methyl alcohol, water, and acetic acid. It is also adsorbed by IR-100 but attempts to desorb it have been unsatisfactory.

Kynurenic acid in neutral solution is adsorbed by IR-4 and desorbed by 1 N HCl. It is poorly adsorbed by IR-100 H. This is to be expected because at a pH = 7, kynurenic acid is an anion in true solution and hence does not enter into cation exchange.

[redacted]

March 19. Physics

23. Uranium Resonance Absorption

N. Goldstein

With pile neutrons and depleted U_3O_8 the effective $\int \sigma \frac{dE}{E}$ was measured for uranium metal, various mixtures of graphite and U_3O_8, and graphite using the $\int \sigma \frac{dE}{E}$ for indium as a comparison. Scattering effects of foil materials and adequate thinness of foils were carefully checked. The results were plotted as a function of scattering cross-section per uranium atom. For metal, 8.2 b/U_a scattering cross-section, the $\int \sigma \frac{dE}{E}$ is 9.4 ± 0.5 b, for graphite, the other end point, the $\int \sigma \frac{dE}{E}$ is 237 ± 3b. The error does not include the errors involved in the constants $\int \sigma \frac{dE}{E}$ for In, (thermal cross-sections for In and 238) which were about 10% and would raise or lower all the values proportionally.

* *

24. Cross-Section Measurements with Pile Oscillator

C. D. Moak

Development work on the Clinton Pile Oscillator having been completed, a description of the instrument and its electronic equipment was given. The electronic synchronizing circuits and power supplies were discussed.

The program of measurement thus far is centered around the measurement of cross-sections of small samples of enriched isotopes. Some interest was expressed by the Theoretical Group in the measurement of the isotopes of Fe. It was thought possible that one of the low abundance isotopes of Fe contributes strongly to the total cross-section of Fe. The 2% Fe^{57} has been measured and ruled out, having a cross-section less than 3.0 barns. The separations are being performed at Y-12 and it is hoped that samples of the other Fe isotopes will arrive soon.

Some interest was expressed in the difference between the danger coefficient and activation cross-sections for Cb. Measurements with the pile oscillator indicate a 1.45 barn cross-section which agrees well with other danger coefficient type measurements.

[redacted]

25. Temperature Characteristics of the Clinton Pile

A. J. Ulrich

I. Barometric Effect

Measurements are made when other factors affecting reactivity are constant and a large barometric change takes place. Best value is -0.4 ± 0.05 inhr/mm of Hg.

II. Temperature Effect

When cold pile is brought quickly from zero to a constant power with constant air flow a reactivity transient is established which upon analysis shows two components, the first due to the metal temperatures rising alone with a half-period of about 1.5 minutes followed by the second due to the metal and graphite temperatures rising together with a half period of about 1 hour.

Best values operating	metal temp. coeff.	$= -0.3 \pm 0.05$	inhr/°C
" " "	graphite	$= -0.8 \pm 0.1$	"
" " "	Total	$= -1.1 \pm 0.1$	"

For a uniform temperature distribution of the pile in vacuo

uniform metal temp. coeff.	$= -0.4 \frac{\text{inhr}}{^\circ C}$	$= -1.0 \times 10^5$	K change
" graphite " "	$= -1.9$	$= -4.7$	
" total " "	$= -2.3$	$= -5.7$	

On the basis of these values the following evaluations were made:

	Morrison's values	Our values
Metal coeff. =	$\Delta_r K = -1.3 \times 10^{-5}$	-1.0×10^{-5}
	$\Delta_l K = -2.7$	
	$\Delta_t K = +4.6$	
	$\Delta_\eta K = -9.7$	-6.6
Graphite coeff. =	$\Delta_l K + \Delta_t K + \Delta_\eta K = -7.7$	-4.7
Total coeff. =	$\Delta_r K + \Delta_l K + \Delta_t K + \Delta_\eta K = -9$	-5.7

III. Xenon Effect

72 hours are required to reach 99% saturation. At saturation poisoning is 26 inhr/megawatt. This corresponds to Xe $= 2.9 \times 10^6$ barns.

[redacted]

26. Studies of Capture Gamma Rays

S. Dancoff and H. Kubitschek

A neutron beam from the heavy water pile is allowed to fall on various targets and the gamma radiation resulting from neutron capture is studied. The maximum energy of such radiation is determined for 17 targets. It seems indicated that this method measures the binding energy of the neutron.

The "yield" of gamma rays from neutron capture is defined as the response of a GM tube in a certain specified geometry per neutron captured in the target. This yield, which would be the same for absorbers which gave off identical gamma spectra, is found in fact to exhibit considerable uniformity for over 30 materials investigated. In this experiment, which is preliminary as far as yield measurement is concerned, only thermal neutron beams have been used.

* *

27. Some Measurements with the 180° Beta-Ray Spectrometer

P. W. Levy

The measurements described below were made with two different spectrometers, (a) a 180° beta-ray spectrometer that employs G.M. tubes as the detecting device, (b) the second is photographic spectrometer similar to the one used by Ellis in his classic work on the conversion electrons in natural radioactivities. The coils of the magnet in which these instruments are used have 44,000 turns and utilize currents on the order of 80 milliamperes. Usually magnets of this type have coils of several hundred turns and utilize currents of several amperes. This arrangement was designed so one could use a high voltage rectifier and regulate the field current electronically.

The beta-ray or counter spectrometer was used to determine the spectrum of 2.7 Day Au^{198}. The end point of this spectrum is .91 mev. and the K, L and M conversions of a gamma ray of .41 mev. is superimposed on it. The same activity was investigated with the photographic spectrometer and the value obtained for the gamma ray energy was confirmed. The region of $H\rho$ = 1000 to $H\rho$ = 75,000 was investigated and no other conversion lines were observed. A conversion line of approximately 1% the intensity of the observed Kline should have been detected with either of these instruments.

In the same fashion the beta spectrum of 60 day Iridium was measured with the beta-ray spectrometer and conversion lines that were found superimposed on the beta spectrum were confirmed with the photographic spectrometer. The end point was observed to be .622 mev. and 29 conversion lines attributable to 12 gamma rays were superimposed on the spectrum.*

[redacted]

It is interesting to note that several lines that did not show up above the statistical fluctuations on the curve taken with the counter spectrometer were detected with the photographic spectrometer.

Just recently the beta spectrum of C^{14} was run with the counter spectrometer.

Because of the extremely long half-life of C^{14} so much material was required that the source was far from ideal and even with this source the peak was only 10 times background. However, the end point obtained, .14 Mev., is probably accurate to within 5%.

---

* A detailed analysis of these lines is contained in MON-P-3.

---

* *

28. The Double Crystal Neutron Spectrometer

R. B. Sawyer

A beam of neutrons reflected from the surface planes of a NaCl crystal falls on a second crystal and the transmitted beam, reflected from internal crystal planes is studied. The first crystal is surrounded by a paraffin-cadmium-lead shield, through a hole in which the reflected beam emerges to strike the second crystal. The first crystal is set to reflect neutrons of 0.07 ev energy and the beam has an angular divergence of about 1° 30', corresponding to an energy interval of 0.02 ev. An enriched BF_3 counter is used as a detector. A small electric motor is mounted so as to rotate the second crystal at a constant angular velocity, ω, of 30 minutes of arc per minute of time.

By setting the counter at the proper angle to receive the Bragg reflection from a particular set of planes and rotating the second crystal through the reflection peak, the integrated intensity, E, of the reflection is registered on the scaler. The crystal is then removed and the intensity, Po, of the incident beam is measured. The coefficient of reflection is defined, as it is with X-rays, as

$$R = \frac{E\,\omega}{Po} \quad .$$

Using NaCl as the second crystal, values of R are obtained of the same magnitude as those found with X-rays. Rotation of the crystal about an axis normal to its surface may vary R by a factor of as much as 1.6. This may possibly be due to internal strains introduced in the process of cleaving.

X-ray theory shows that, for a given crystal,

$$R \sin 2\theta \cos \phi = \text{constant} \times F^2,$$

where θ is the glancing angle which the beam makes with the reflecting planes, ϕ is the angle between the beam and the normal to the crystal surface, and F is the crystal structure factor. θ, ϕ, and F are functions of the particular set of planes used for reflection. The following table shows some preliminary values.

Planes Used	R	R sin 2 θ cos ϕ
(200)	5.36×10^{-4}	1.97×10^{-4}
(400)	3.41	2.22
(600)	1.44	1.11 (value questionable)
(220)	4.56	2.07 (two different crystal orientations.
(220)	4.22	2.11
(111)	3.21	0.94
(311)	1.43	0.86

It is seen that the value of F for planes having even indices is quite different from that for planes with odd indices. Further work is in progress.

* *

29. Magnetic Moment of the Neutron

W. H. Zinn

The large neutron flux available from CP-3 should make possible a more accurate determination on the neutron magnetic moment using the method of Alvarez and Bloch (Phys. Rev. 57, 111 (1940).

The scattering cross section for neutrons oriented with their magnetic moments parallel to the field in a block of iron is different from the cross section for anti-parallel neutrons. The increased transmission for one component results in a transmitted beam that is partly polarized and in an increased total transmission. If a neutron beam passes thru two separated blocks of magnetized iron, such as those between the pole pieces of magnets I and III as shown in figure 3, in CP-3490, the intensity of the transmitted beam will reflect any depolarization that occurs between the two blocks.

In order to find the neutron magnetic moment a coil, whose axis is colinear with the neutron beam, is placed between the pole faces of magnet II and generates an alternating field of several megacycles. The neutrons will precess in the field H of magnet II with a frequency f given by

$$f \cong 2uH/h$$

where u is the magnetic moment of the neutron in nuclear Bohr magnetons ($eh/4\pi Mc$) and h is Planck's constant. If the frequency of the oscillating

field is made equal to this Larmor precession frequency, the polarized beam from magnet I will be depolarized and the transmission through the iron in magnet III will decrease.

The change in transmission with increasing magnetic saturation of the iron is shown in figure 4 in report CP-3490. The upper curve shows the ratio of magnetized to unmagnetized intensities with the blocks of iron in magnets I and III magnetized in the same direction. The lower curve is obtained when I and III are magnetized anti-parallel, and gives an indication of the amount of depolarization which occurs between the magnets due to stray fields. If no depolarization occurred the ratio for the lower curve would be 1.0. If magnets I and III were magnetized in the same direction, and complete depolarization was achieved in magnet II, one would expect a 'dip', or decrease in the intensity of the transmitted beam, of one half the difference between these two curves -- about 4½% at 250 ma. These curves also show that saturation of the 3 cm armco iron blocks is not complete and that if higher fields were obtained the dip would be 2 or 3 times as large.

Figure 5b in report CP-3490 shows the dip in intensity which occurred when, with a 10 megacycle alternating field in the r.f. coil, the field of magnet II was increased through resonance. The current in I and III was 250 ma (about 6100 oersteds), the polarizer and analyzer were armco iron blocks 2.54 x 2.86 cm in area and 3 cm thick in the direction of the beam. The dip was 4% of the total intensity of 12,000 cpm on the BF_3 counter. The beam was collimated to 2.5 cm high and 0.9 cm wide. The field of magnet II was measured to be 3370 ± 70 oersteds. This gives a value of the magnetic moment of

$$u = \frac{f\,h}{2\,H} = \frac{10^7 \times 6.62 \times 10^{-27}}{2 \times 3390} = 0.978 \times 10^{-23}$$

or 1.94 ± .04 nuclear Bohr magnetons.

Bloch and Alvarez obtained 1.93 ± .02 for the moment, but since the probable errors overlap so far the difference is not significant. The frequency of the oscillator was set to zero beat with the 10 mc signal of WWV of the National Bureau of Standards so practically all the uncertainty in our result is in the measurement of H. Fig. 5a in report CP-3490 is a dip obtained at 5.3 mc and 1800 oersteds.

At present an attempt is being made to measure the proton moment in the same field that is used for the neutron. This would give the ratio of the two moments in terms of a ratio between two frequencies which could be measured with high accuracy. A permanent magnet has been constructed with a field of 1880 oersteds, uniform to 0.5% over an area of 3" x 5", to replace magnet II, and extra coils have been added to magnets I and III which increases their fields by 36% for the same 250 ma to which heating limits them.

* *

30. Preliminary Measurements of C^{14} Radiation with an Ion Chamber and Vibrating Diaphragm Electrometer

R. K. Swank and William P. Jesse

Preliminary measurements have been made of the ionization produced by thin solid samples of C^{14} in the form of sodium carbonate and sodium acetate in a specially designed ion chamber coupled to a vibrating-diaphragm electrometer. This chamber is fitted with a drawer which pulls out to receive the sample, usually mounted on aluminum foil. The measurement is made by timing the travel of the needle of the output meter over its scale. The electrical capacity of the system is 20 m.m.f. and full scale voltage sensitivity is 100 millivolts. The samples were kindly furnished us by Dr. Finkel, who gave us a rough estimate of their activity in disintegrations/sec based on measurements made by him with a mica-window counter. Our measurements with the ion chamber system on thin samples have so far proved quite as reproducible and reliable as those made with a mica-window counter. A set of ten different measurements, each of 20 minutes duration, gave a standard deviation from the mean of 3%. No single measurement differed by more than 5% from the mean. This sample would count about 30 c/min above background on a mica-window counter.

* *

31. The Half-Life of C^{14}

by L.D. Norris

Several samples of $BaCO_3$ containing C^{14} were analyzed by Dr. A. J. Dempster's group in Chicago. Small weighed portions of these samples were then counted in a low absorption counter to determine the absolute disintegration rate. Then, very simply,

$$T_{\frac{1}{2}} = \frac{.69\ N_o}{N_\tau}$$

The low absorption counter was of the usual type, i.e., a counter tube (with a weightless zapon window), mounted in a large, glass desiccator fitted with a rotating tray of aluminum absorbers. This counter was standardized with UX, which had been evaporated upon films of weightless zapon in order to eliminate backscattering.

Portions (approximately 0.15 milligrams) of the $BaCO_3$ samples were weighed on a micro balance, transferred to holders of one mil aluminum, slurried in alcohol by means of a micro stirring rod, dried, and counted in the low absorption counter. Upon making a previously determined back-scattering correction, the total number of disintegrations in the sample were obtained, and $T_{\frac{1}{2}}$ calculated. Two weighed portions of one $BaCO_3$ sample

[redacted]

[redacted]

gave values for $T_{\frac{1}{2}}$ of 20,000 years, and 18,000 years. Since this counting work is still in a preliminary stage, we shall not attempt to evaluate the significance of the above values for the half life.

* *

32. Cross Sections for Fission Neutrons

by D. J. Hughes

The work previously reported (CF-3403) on radiative capture cross-sections for fission neutrons showed extremely low values for light elements. We have now extended the measurements to (n - p) and (n α) reactions for the light elements, where in some cases the cross sections exceed the (n γ) cross-sections. The values obtained for the average cross-section for fission neutrons range from 0.01 millibarns for the O^{16} (np) to 8 mb for the Be(n α). The average cross-sections decrease with increasing threshold energies roughly parallel to the decrease of the number of neutrons in the fission energy spectrum, hence give information on the extrapolation of the spectrum above measured values.

* *

33. Mass Assignments and Enrichments

by A. J. Dempster

A sample of carbon 14, sent to us by Dr. Snell, has been analyzed by Mr. Inghram with a Nier type mass spectrometer. The sample was in the form of barium carbonate and carbon dioxide was obtained, on the advice of Dr. Willard, by simply heating to about 1000° in a quartz tube. The small amount CO_2 given off at low temperatures was not as rich in carbon 14 as the main amount, suggesting that the absorbed carbon dioxide from the air will exchange in time with the carbon in the barium carbonate. The two samples had 3.35 and 3.23 per cent of carbon 14. A recent mass assignment is the 13 hour palladium isotope to mass 109 and not to 107 as was once supposed. The analysis of the neodymium samples prepared by Dr. Gladrow at Ames by adsorption processes are still uncertain. There appears to be an enrichment of the light isotopes in the tail fraction. However, there are alterations in the heaviest isotope at mass 150 that cannot yet be interpreted.

* *

34. Fissionability Studies: Po^{210} and Np^{239}

by J. J. Floyd

Samples of Np^{239} and Po^{210} have been studied for thermal fissionability. The method used was photographic involving special treated commercial materials. The sample for study is placed in contact with the photographic

[redacted]

plate and exposed to thermal neutron flux of 10^{11} neutrons per square centimeter. The exposed photographic plate is studied for fission tracks with a high power microscope. Limiting cross sections may be determined upon the basis of the number of events observed. The cross section of Po^{210} is certainly less than 10^{-25} centimeters^2 and the cross-section of Np^{239} is probably less than 3×10^{-24} centimeters^2.

* *

35. Recent Results of Heavy Isotope Work

by Glenn T. Seaborg

A. Properties of Element 95

I. Oxidation states.

The most stable oxidation state in aqueous solution is almost certainly III. This conclusion is based on (a) the pronounced similarity in chemical properties of aqueous solutions of element 95 and of the tripositive rare earths, (b) the adsorption-elution behavior of 95, relative to the rare earths, on amberlite columns (the behavior of 95 is that expected for a tripositive ion having the radius predicted for the sixth member of the actinide series) and (c) the similarity of the aqueous solution absorption spectrum of element 95 to that of tripositive europium, its rare earth analog.

A higher oxidation state may be obtained by oxidation with bromate in ~15 M HNO_3. The formal oxidation potential of the 95 (III-III + x) couple in 4 M HNO_3 is almost certainly more negative than ca. -2 v (since there is no oxidation by Ag^{++} in 4 M HNO_3).

In alkaline solution the flesh pink color ofthe solid hydroxide, $95(OH)_3 \cdot XH_2O$ is changed to reddish brown by the addition of H_2O_2 or ClO^-. This may be evidence for oxidation in alkaline solution.

The IV state is known in the black oxide, $95O_2$ (identified by W. H. Zachariasen). This oxide is obtained by igniting the nitrate or the hydroxide of the III state in air at 500-600°C. There is some preliminary evidence that thermal decomposition of the dioxide (presumably to the sesquioxide) may be brought about at higher temperatures.

Polarographic evidence of a very preliminary and tentative nature indicates that 95(III) is reduced at the dropping mercury electrode at a potential of ca. +0.8 v (latimer convention).

II. Compounds.

In addition to the compounds already mentioned, a trifluoride has been prepared by precipitation from aqueous solution, and a trichloride by the action of CCl_4 vapor on the oxide at 700°C.

III. Aqueous ions.

The color of aqueous solutions of the tripositive ion in 1-2 M HNO_3 or HCl is pink. The absorption spectrum of the aqueous ion in 1 M HNO_3 has been determined in the region 350-1200 millimicrons, using the Beckman quartz spectrophotometer. The characteristics of the principal absorption bands are as follows:

Location of Maximum ($m\mu$)	Apparent E	Half-width ($m\mu$)
503 ± 1*	300*	5*
812 ± 5	60	30
987 ± 2	20	15

*Not completely resolved by the spectrophotometer.

In 15 M HNO_3 a maximum occurs at 506 ± 1 $m\mu$, and there is a pronounced decrease in the value of E.

IV. Emission spectrum.

Fifty-one lines in the emission spectrum (copper spark) of element 95 have been determined by Mark Fred and Frank Tompkins. The wave lengths of ten of the most prominent lines in the region 2700-4000A° are as follows:

3926.24	(Probably unresolved triplet)
3696.43	
3161.88	
3038.39	
2972.57	
2969.29	
2966.71	
2920.61	(Strongest line)
2756.55	
2755.78	

About 0.4 microgram of the element may be detected spectrographically.

V. Specific activity and half-life

The specific activity has been determined as 3.6×10^6 counts per minute per microgram in a standard "50%" counter. This figure corresponds to a half-life of ~490 years. The error in these figures is almost certainly less than ± 30%.

[redacted]

B. Fissionability of Heavy Isotopes

			Barns σ_f	Chemists Responsible for Purification
88	Ra	223 (acX)	<100*	Peterson
		226	$\leq 1.1 \times 10^4$	Ames
		228 (MsThI)	<2	Peterson
89	Ac	227	< 20	Peterson
90	Th	227 (RaAc)	540 ± 200	Peterson
		229	40 ± 10	Studier
		230	$\leq$ 0.0012 ≠	Van Winkle
		232	$\doteq 2 \times 10^{-5}$ ≠	Van Winkle
91	Pa	231	0.010 ± .005	Van Winkle
		233	< 0.1	Studier, Hageman
92	U	232	70 ± 10	Van Winkle
93	Np	237	0.018 ± .005	Osborne, Hindman, Magnusson
94	Pu	238	$\leq$ 18	Osborne
		240	$\leq$ 180	Westrum, James, Florin
95	Am	241	$\leq$ 2.8	Cunningham
96	Cm	242	(~600)*	Thompson, S. G.

≠ Since Th^{230} contained Th^{232} value for Th^{232} was used to get net effect due to Th^{230}.

* If 95^{241} σ_f = 2.8·b and no Pu^{239} contamination.

C. Suggested Names for Elements 95 and 96

Elements 95 and 96 should of course have names and we propose the following in which these actinide elements are given names by analogy with the corresponding members of the lanthanide earths. We suggest for element 95, with its six 5f electrons, the name "americium", symbol Am; thus this element would be named after the Americas, or New World, by analogy with europium, with its six 4f electrons, which was named after Europe. For element 96, containing seven 5f electrons, we suggest "curium", symbol Cm, after Pierre and Marie Curie, historical leading investigators in the field of radioactivity; this is by analogy with gadolinium, containing seven 4f electrons, which recalls Gadolin, the great investigator of the rare earths.

[redacted]

[redacted]

36. Cyclotron Bombardments of U^{233}

by Martin H. Studier

Low atomic weight neptunium and plutonium isotopes were prepared by bombardment of U^{233} with 22 Mev deuterons and 44 Mev helium ions. A 4.5 day orbital-electron capture activity (accompanied by a 1.8 Mev gamma ray) is assigned to Np^{234}. The limit of alpha branching is set at $< 0.01\%$. An isotope with an apparent half life for alpha decay of $\sim$ 10 hours and an energy of ca 6 Mev is assigned to Pu^{234}. The absence of any evidence for Pu^{235} indicates its half life for decay by orbital-electron capture to be $<$ 12 hours or $>$ 5 years. A 40 day orbital-electron capturing isotope previously assigned by James, Florin, and Hopkins to Pu^{235} or Pu^{237} may now be definitely assigned to Pu^{237}. Evidence for the reaction U^{233} (d, α n) Pa^{230} was found.

* *

37. Production of Actinium from Radium; Properties of Actinium and Its Daughters

by S. Peterson

(No abstract received)

* *

March 19. Chemistry

38. Contributions to Radio-Chemistry of the 104 Day Y^{88}

by R. Overman

The gamma-emitting, long-lived yttrium isotope from deuteron bombardment of strontium is used in various fields of applied radiochemistry. Data are given for a highly purified preparation of the isotope. Radiation absorption curves are presented with a discussion of secondary effects observed in standard counting procedures; under conditions of low scattering and magnetic deflection of Compton electrons, a component was discovered which corresponds to the K X-rays of strontium. The half-life was found to be 104 $\pm$ 2 days. Mass spectrographic analysis of the sample by Dr. R. L. Hayden of the Metallurgical Laboratory shows the radiations arising only from mass 88.

Bombardment of yttrium with high energy neutrons gives the two isomers of Y^{88}, of 2.0 hour activity and the 104 day activity, the 60 hour activity of Y^{90}, the 55 day activity of Sr^{89}. Absorption measurements of the positron energy associated with the 2.0 hour activity give a value of 1.65 Mev rather than the 1.2 Mev value reported in the literature. This new value is in better accord with the theory of the existence of independent isomers.

* *

[redacted]

39. Summary of Fission Product Investigations at HEW

by W. H. Sullivan

Experimental work at HEW was directed to establish the existence or prove the absence of certain long-lived activities which have been reported in previous project and published literature or which theoretically could be present, according to the Bohr-Wheeler calculations for unstable nuclei. Most of the activities which were sought occur in elements whose isotopes lay on the extremities of the fission yield vs. mass number curve and which could be studied most easily in solutions of high specific activity.

A summary of these investigations is included below.

1. Activities in daughter extractions of active Ce.
The trans-Ce activities derived from Ce parents which had been reported in previous project literature do not exist in pile fission material. According to experimental evidence the "55d Pr" was probably due to 57d Y^{91} contamination; the 11d "trans-Ce" was due to the 13½d Pr^{143} contamination; and the "1y trans-Ce" activity, to the 275d Ce^{144} activity.

2. Rh
No long-lived Rh activity is produced in pile fission. Calculation of the upper limit of the fission yield leads to the value of $< 6 \times 10^{-7}$% for a Rh isotope of $\sim$25d half-life.

3. Mo
No long-lived Mo activity is produced in pile fission. Calculation of the upper limit of the fission yield gives a value of $< 4 \times 10^{-9}$% for a Mo isotope of $\sim$60d half-life.

4. Element 43.
A long-lived activity having decay and radiation characteristics unlike any fission product thus far identified was found in 25 gt material. Estimates of the half-life, making assumptions concerning fission yield and chemical recovery, lead to a value of $> 5 \times 10^3$ yr. and probably less than 10^6 yr.

5. Pd
No long-lived Pd activity was found in the plant dissolver solution. If Pd^{107} is long-lived, its half-life must be of the order 10^8 years.

6. Sn
A long-lived Sn activity with a half-life of 130d was identified in the plant dissolver solution. It was found to emit only beta particles whose maximum energy according to Feather analysis was 1.5-1.6 Mev. The fission yield was observed to be 0.0012%. The activity is probably an isomer of either the 10d $Sn^{(123)}$ or the 62h $Sn^{(121)}$ activity, both of which decay to stable antimony isotopes.

7. Sb
The controversial long-lived (250-400 day) Sb activity was found to exist. Its half-life as determined by measurements over several months was $\sim$2.7 years. Associated with this activity are $\sim$0.5 Mev electrons, 0.5_6 Mev gamma rays, and $\sim$0.026 Mev x-rays. From considerations concerning the fission yields, possible isotope assignments and decay mechanisms, it is considered possible that the activity is a nuclear isomer of stable isotope Sb^{123}.

[redacted]

8. I

An I sample, which was collected from the stack gases in a scrubber column and which at the end of bombardment contained more than 40 curies of 8d I^{131} as based on an extrapolation of the experimental decay curves, was found to contain only ~30 counts/minute of a long-lived activity after 371d of decay. Calculations of the lower limit of the half-life for isotope I^{129} lead to a value of $> 3.8 \times 10^9$ yr., if observable β radiations are present.

9. As

No long-lived As activity with a half-life of 40-400d was found to be formed in fission with a yield $> 5 \times 10^{-6}$%.

* *

40. Deuteron and Helium Ion Bombardment of Pa^{231}

by D. W. Osborne, R. C. Thompson and Q. Van Winkle

(Presented by D.W.Osborne)

Bombardments of Pa^{231} by 22 Mev deuterons and by 44 Mev helium ions have produced two new uranium activities, in addition to U^{230} and U^{232}: a 4.1-day x-ray activity believed to be due to K-capture by U^{231}, and a 1.5-day alpha-activity tentatively assigned to U^{229}. Repeated milkings of thorium from the uranium fraction isolated after the deuteron bombardment have shown that the half-life of U^{231} for alpha emission is over 100 years, corresponding to an alpha branching of less than 0.01%. The only activities found in the protactinium fractions were the previously known β^- emitting Pa^{232} and Pa^{230}, although there was some evidence for a daughter of Pa^{229}, (known from previous work to be an alpha emitter with 33-hr. half-life). Pa^{230} emits a gamma-ray with 0.8 Mev energy, and the Feather range for the β^- is about 450 mg/cm^2 in aluminum (1.1 Mev). The half-lives for Pa^{232} and Pa^{230}, as determined by decay of the radiation through 5 g/cm^2 of lead, are 1.4 days and 18.2 days, respectively. The neptunium fraction from the helium ion bombardment had a 4.3-day x-ray and gamma activity, probably Np^{234}, which had also been found in deuteron bombardments of U^{233} and U^{235}, and an unidentified softer Geiger activity with 1.5-day half-life.

41. Color Production in Lithium Fluoride by Radiation

by Arild J. Miller

Crystals of synthetic lithium fluoride are colored by electron, x-ray, and pile radiation (neutron). The principal effect of pile radiation is caused by the $Li^6(n, \alpha)H^3$ reaction, the α and H^3 particles having a total kinetic energy of ~4.6 Mev. Qualitatively, the color depends upon the amount of irradiation, and progresses from a light green to a deep red on sufficient exposure. The absorption spectra of these crystals, colored by the various radiations, show discrete bands with peaks at 250, 310, 375, 445, and 550 mu which are identical in position for the different irradiations, and are identified as F, R, R, M, and F°-bands, respectively. Heating at 300°C restores the original transmission.

[redacted]

42. Behavior of Pu in the Presence and Absence of Citrate Solutions

by Jack Schubert

Fig. 2 shows how the ultrafiltrability of plutonium declines with increasing pH

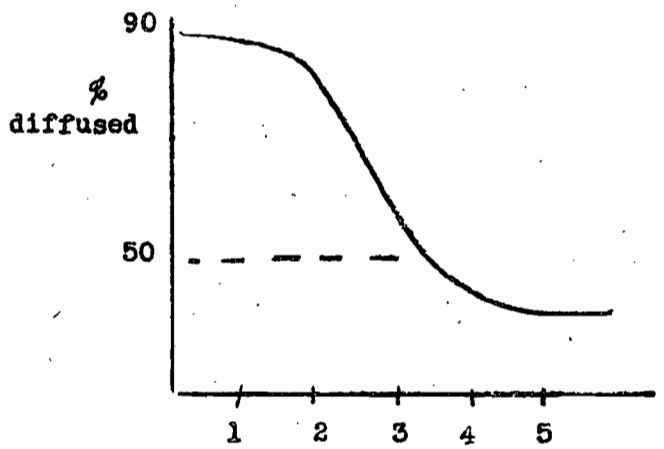


Fig. 2

The drop at pH 2.5 is due to polymerization. At the same pH, material begins to adhere to the membrane. Addition of 10^{-5} m. citrate increases diffusion markedly; at 10^{-3} m. citrate, diffusion becomes complete, and no sticking to the membrane is observed. This is the result of the formation of a plutonium-citrate complex. Fig. 3 shows how this complexing affects the adsorption of Pu on IR-130 resin.

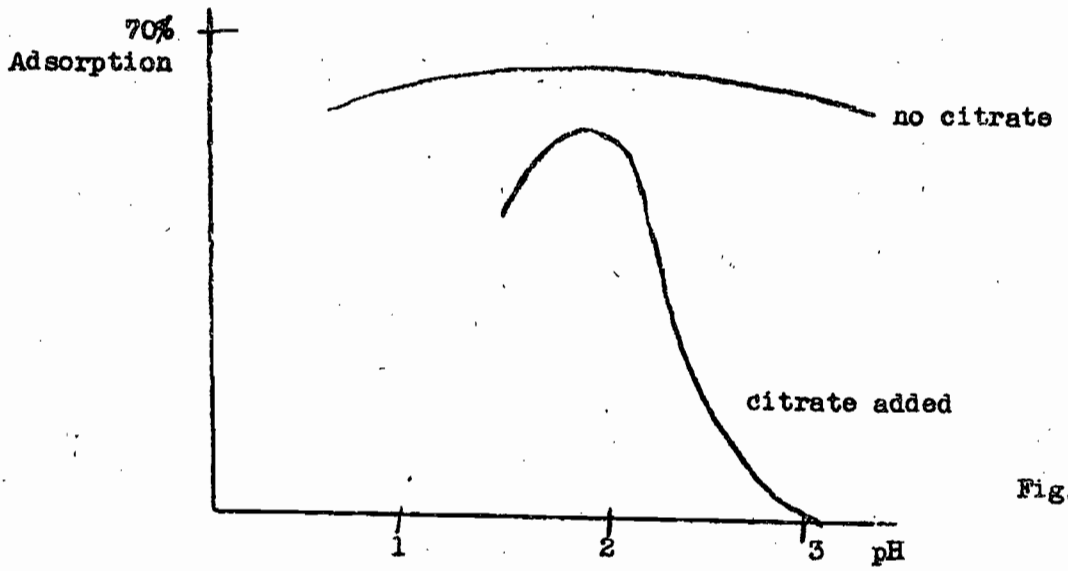
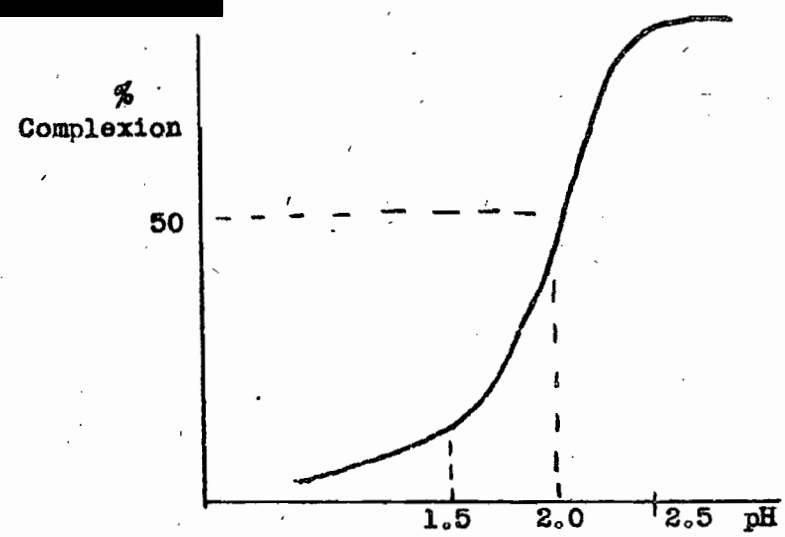


Fig. 3

From these observations, the complexion curve can be derived:

[redacted]



Pu^{IV} has a stronger tendency for complexing with citrate than La^{III} or Ba^{II}. the pH's corresponding to 50% complexing are 2, 3 and 4 for Pu^{IV}, La^{III} and Ba^{II} respectively.

There is some evidence that Pu complexes with proteins in blood. This is to be tested by ultrafiltration.

* *

43. Potentials of Pu Couples

by K. A. Kraus (Clinton)

Trivalent plutonium in HCl is probably not complex up to pH 7. From there on, complexion occurs,

$$Pu^{3+} \xrightarrow{pK\ 7.2} Pu(OH)^{2+} \longrightarrow Pu(OH)_3$$

Tetravalent plutonium probably forms free Pu^{4+} - ions in strong acid, but begins to hydrolyze at pH $>$ 1.

$$Pu^{4+} \xrightarrow{pK\ 1.3\text{-}1.4} Pu(OH)^{3+} \xrightarrow{\text{Rapid polymerization to colloid}}$$

$$\left\{ Pu(OH)^{+0.2}_{3.8} \right\}_n \longrightarrow \text{precipitation}$$

Pentavalent plutonium presumably is PuO_2^+ up to 0.5 N acid. It does not hydrolyze up to about pH 9 (pK $\cong$ 9.7).

Hexavalent plutonium is supposed (but not actually proved) to be PuO_2^{++} up to about pH 5. It then hydrolyzes

[redacted]

$$PuO_2^{++} \xrightarrow{pK\ 5.5} PuO_2(OH)_2 \xrightarrow{pK\ 9.8} PuO_2(OH)_3^-$$

We can accordingly predict the pH-dependence of E_o° for several Pu-couples. For example, the Pu^{III}/Pu^{IV} potential should be independent of pH below pH 7. At pH 1.5, when Pu^{IV} rapidly polymerizes, E_o° changes with $[H^+]^4$.

Pu^{III} must be capable of reducing water from pH 6 on.

The potential Pu^{IV}/Pu^{V} can be calculated from the known potentials of Pu^{IV}/Pu^{VI} and Pu^{V}/Pu^{IV}. No complete agreement exists between all measurements. The E_o° is pH-independent (at -0.86 volt) above pH 5, since both reductant and oxidant concentrations are proportional to $[H^+]^3$.

The Pu^{V}/Pu^{VI} couple is practically reversible.

The measured E_o° - value is -0.92 volt at pH 3.5.

* *

44. Recent Work on Redox Process

by I. Scaffner (Chicago)

The redox method has been under development at Chicago since August. A pilot plant, operating on batches, has been built and found to work satisfactorily. It has been inspected by duPont, and a larger pilot plant (processing 0.1 ton U/day) will be erected at Hanford.

Five columns give 99.2% yield and a decontamination factor of $10^{7.3}$ (in β's as well as γ's). Plutonium produced contains only 0.1% uranium; the residual activity is due mainly to Ce and Ru.

An alternate salting-out agent is now being studied.

In column I B, the use of ferrous sulfate with hydrazine and 8N NH_4NO_3 lead to a 0.5 - 0.8% loss of Pu.

By reducing HNO_3 and replacing it by acetic acid, the loss can be cut to 0.2%. Another attempted substitution was that of $Al(NO_3)_3$ for NH_4NO_3.

[redacted]

45. Action of Electron Beams and X-Rays on Highly Purified Water*

By John A. Ghormley

Pure water in pyrex cells was decomposed by electron beams and x-rays to yield hydrogen, hydrogen peroxide, and oxygen. The initial products were shown to be hydrogen and hydrogen peroxide. By suitable adjustment of the operating variables, net decomposition of water or net formation of water from the decomposition products could be made to occur. The concentration of products at the steady state wherein no net change occurred, increased with increasing radiation intensity and with decreasing temperature. The observed initial yield (G_{H_2}) varied from 0.1 to 0.5 molecules H_2 per 100 ev. All results are explained on the basis of free radical mechanisms, H and OH being formed as a result of ionization of water molecules by fast electrons. The steady state concentration was found to increase when water was allowed to stand in the pyrex cell for three weeks, presumably because of sensitization of the reaction by dissolved glass. Mechanical agitation of the cell during electron bombardment produced a large effect on the steady-state concentration; this effect indicates presence of a relatively long-lived free radical, presumably HO_2.

* *

46. A System for the Separation of Uranium from Thorium using Organic Complexing Agents in Solvent Extraction

by O. K. Neville

A method has been developed for the recovery of small amounts of uranium from a concentrated thorium solution by the use of organic complexing agents and solvent extraction techniques.

Certain dithiocarbamates ($R_1R_2NCSSNa$) have been found to be excellent complexing agents for uranium in the presence of thorium and to promote the solvent extraction of uranium from thorium in very high yields. Of the dithiocarbamates that have been prepared, those derived from secondary amines are better agents for the purpose than those derived from primary amines. Those derived from phenyl amines are superior both in complexing ability and stability. The best agents found are the sodium salts of: ethylphenyl-, butylphenyl-, and benzylphenyldithiocarbamates.

The experimental procedure is as follows: A uranium-thorium solution, 1 molar in thorium and 3×10^{-5} molar in uranium, and which has been previously adjusted with ammonia to a pH of 3, is shaken 5-15 minutes with an equal volume of ethyl acetate to which has been added the organic reagent to the extent of 10^{-3} molar. When the layers are separated, over 99% of the uranium has been extracted into the organic phase. Less than 0.5% thorium is extracted.

Stripping to the extent of 95% may be accomplished with an equal volume of 1 M HNO_3. If 0.5 M HNO_3 is used, uranium recovery is 90% per batch.

If a one-half volume of ethyl acetate is used in the first extraction, the uranium is extracted to the extent of about 95% on a small scale. This

[redacted]

may be improved on a larger scale. One-half volumes of 1 M HNO_3 for stripping cut the uranium recovery to 85-90 per cent.

The work on the process has been taken over by the Chemistry Section of the Technical Division at Clinton.

* *

47. Activity Coefficients of Plutonium

by R. E. Connick (Berkeley)

We studied the extraction of Pu into benzene with trifluro acetyl acetonate as chelating agent

$$Pu^{4+} + 4\ HK \text{ in } (C_6H_6) \rightleftharpoons Pu\ K_4 \text{ in } (C_6H_6) + 4H^+$$

The extraction is proportional to $[K]^4$:

$$\text{Const} = \frac{(PuK_4)\ (H^+)^4}{(Pu^{4+})\ (HK)^4}$$

where parentheses refer to activities in the benzene phase. Assuming (PuK_4) to be proportional to concentration (since we deal with tracer quantities), and the same to be true of HK (because the concentration is low), we obtain

$$\text{Const} = \frac{\gamma_{H^+}[H^+]\,[PuK_4]}{\gamma_{Pu^{4+}}[Pu^{4+}]\,[HK]^4}$$

where square brackets indicate concentrations.

What is measured, is $[PuK_4]$.

We studied the effect of mixed acid, e.g. that of addition of HCl to $HClO_4$. The results indicate the extent of complexing of Pu with Cl^-.

From $\gamma_{Pu^{4+}}$ and the potentials, one can calculate γ-values for Pu^V and Pu^{VI}.

[redacted]

48. Xe Poisoning and Pile Stability

by G. Nordheim and H. Soodak
(Clinton)

In all actual piles, the reactivity does not only depend on the position of the control rods, but also on quantities like the temperature and fission product poisoning which in turn depend on the power level. Therefore, piles have a certain equilibrium power. The question of pile stability consists in determining whether deviations from the equilibrium state decay, in which case the pile is stable, or build up, in which case the pile is unstable.

Since an increase of power decreases the amount of Xe in the pile (by neutron capture) and therefore increases the reactivity, the Xe is an unstabilizing factor. The temperature, on the other hand, is a stabilizing factor for piles with a negative temperature coefficient. Calculations were made of the periods with which deviations from equilibrium build up or decay, not only for the X and W piles but also for the proposed light-water cooled, heavy-water moderated heterogeneous pile.

The following table gives the unstable periods due to the Xe for the three piles with and without taking the temperature effect into account.

Unstable Period

	X - Pile	W-Pile	New Pile
Xe alone	98 min	11 min.	.9 min
Xe + Temp.	No pos. period	14 min.	~8 min

* *

49. Migration Area in Heterogeneous Piles

by R. Scalettar and A. Weinberg
(Clinton)

In the investigation of the characteristics of enriched lattice piles the simplest procedure is to determine the pile constants in such a way that, at once, the formalism of the homogeneous pile equation is applicable and that, of course, the correct results will be obtained for critical sizes, motion of the pile, etc.

Since most of the slowing down is accomplished by the moderator the age is practically unaffected by the lattice structure (in enriched piles of low concentration). The diffusion length is the only quantity which needs to be adjusted for the heterogeneity of the pile. The problem then is to determine the diffusion length, L, in such a way that the homogeneous thermal

theory equation

$$L^2\Delta + k_\infty - 1 = 0$$

gives the correct Laplacian, Δ, and consequently the correct pile size; here k_∞ is the reproduction constant for an infinite medium and is given by $\eta \epsilon f_\infty$ (neglecting resonance absorption which is small in the enriched pile). η is the number of neutrons produced per absorption by a fissionable atom, ϵ the fast effect contribution to reproduction, and f_∞ is the thermal utilization for an infinite pile. If, for the finite lattice pile, we define f_{eff} = number of neutrons absorbed in U in cell/number of neutrons produced in the cell, the critical condition for the lattice is

$$k_{eff} = \eta \epsilon f_{eff} = 1$$

Comparison of these two equations which must express the same critical condition we find for the L^2 which gives the correct Laplacian

$$L^2 = \frac{1}{1\Delta 1} \frac{f_\infty - f_{eff}}{f_{eff}}$$

The evaluation of f_∞ and f_{eff} was carried out for a bare rectangular lattice pile infinite in the X-direction and finite in the Y and Z directions, the half thickness per cell of uranium being X_o and of moderator X_1. It was assumed that the number of neutrons becoming thermal in the moderator was independent of X and that the uranium did no slowing down. Under these assumptions one obtains (where L_o is the diffusion length in the uranium, L_1 in the moderator).

$$L^2 = L_1^2 (1 - f_\infty) - L_o^2 f_\infty + \text{correction terms}$$

The significance of the first two terms is that if one imagined our lattice to be smeared into a homogeneous structure these terms would be closely the correct diffusion length for the mixture. The correction terms are

$$\text{Correction terms} = f_\infty \left[\frac{\sigma_{a_1} X_1}{\sigma_{a_o} X_o} \frac{X_o^2}{3} \left\{ 1 - \frac{(2+1\Delta 1 L_o^2)}{15} \frac{X_o^2}{L_o^2} + 1\Delta 1 L_1^2 \left(1 - \frac{\overline{2+1\Delta 1 L_o^2}}{15} \frac{X_o^2}{L_o^2} + \cdots \right) \right\} + \frac{1}{3} \frac{X_1^2}{L_1^2} \left\{ \frac{(1+1\Delta 1 L_1^2) X_1^2}{15} + L_o^2 \left(1 - \frac{1}{15} \frac{X_1^2}{L_1^2} + 1\Delta 1 L_1^2 \left(1 - \frac{2}{15} \frac{X_1^2}{L_1^2} + \cdots \right) \right) \right\} \right]$$

[redacted]

[redacted]

Here σ_{a_0} and σ_{a_1} are respectively the absorption cross sections in uranium and moderator. The correction terms for all usual geometries are small and for the volume ratios of the enriched pile amount to only about 4% of the first two terms. Thus the critical size may be calculated correctly using the homogeneous expression for the diffusion length.

The assumption in the above calculation concerning the constancy of thermal neutron production in the X-direction is not strictly correct but it is not believed that this will produce more than a second order correction since the result depends on total thermal neutron production rather than its distribution. However, a calculation on the basis of the two group theory to check this point is in progress. The question of the correct expression to be used for L^2 during the motion of the pile (non-stability) is also under investigation.

* *

50. Proposed Experimental Facilities for the Heterogeneous Pile

John R. Huffman

The present conception of the P-9 moderated light-water cooled 25 heterogeneous pile at Clinton envisions 30 active sub-assemblies spaced in a lattice of diameter 93 cm and length 100 cm, surrounded by 20 cm of P-9 reflector top, bottom, and sides. A ring of 26 thorium rods is situated on a diameter of 137 cm which is backed up by 20 cm of P-9 reflector. An aluminum reactor tank 175 cm in diameter and 140 cm high results. This is surrounded on the sides by 75 cm of graphite containing 69 C^{14} or cooling holes, 6" of iron thermal shield, and 8' of concrete biological shield replaced by 42" of W-type shield at three points. The reactor tank is indented at five points to permit experimental facilities to approach the lattice from the sides.

The majority of experimental facilities will permit utilization of neutron densities from 10^{13} to 2×10^{14}. As many facilities as possible have been provided, even to the extent of using upward and downward inclined as well as horizontal experimental holes. About ten holes 4" to 12" in diameter piercing the pile structure and the tank permit experimentation at at 10^{14} nv. An additional five holes reach to a field of 3×10^{13} nv (tank wall). Twelve 2" ID pneumatic tubes are provided, four of which pass through the top reflector region of the tank at 10^{14} nv. Two "donut" tubes are provided in the graphite at $\sim 10^{13}$ nv.

A side thermal column 6' x 6' x 8' is provided with a water cut-off tank at the inner end. A twelve inch square stringer is provided on the center line of the column. In addition, five 4" holes approach the sides and top of the column and one 4" hole passes through the column transversely.

A biological hole 4' x 4' x 3" is provided below the side thermal column by bringing the neutrons down at an angle in a graphite path.

[redacted]

In the center of the reactor tank a 4" ID central thimble with access at both ends is provided. This is the only large vertical hole penetrating the tank. Any of the 47 available uranium holes, the 41 available thorium holes, and the 69 C^{14} or cooling holes can be used for other experiments.

Eight instrument holes are planned in four banks of two each, with six tangent to the outside of the iron thermal shield and two penetrating into the graphite region.

All holes are filled with shielding plugs normally. Experiments in general will have to be built in special plugs with cooling facilities and rather elaborate charging and discharging coffins. All holes can be served by pneumatic devices. The accessibility of vertical holes in the lattice structure will be extremely limited.

* *

51. Pile Calculations

by Gale Young

In a general way, breeding gain increases and specific power rate decreases as one raises the energy at which neutrons are absorbed in a pile. The rate at which the supply of fissionable material can be enlarged depends on the product of these two quantities; calculations to date suggest that this product may have its greatest value in intermediate or resonance energy piles (cf. talk by Goldberger).

All cooled piles to date have operated on thermal energy neutrons. The unit being designed by Zinn will run on fast neutrons and, it is hoped, will provide the first demonstration of breeding gain. Liquid metal cooling is being developed for fast piles, and may be of use in resonance and thermal piles as well. The approximate composition of the Zinn pile is 1 atom U, 0.6 atom Fe, and 0.2 atom of coolant (Na_2K). The capture cross-section of Fe is still somewhat uncertain.

The above mentioned fast piles can give both breeding and power, and operate on either the 49-28 or the 23-02 cycle. Another type which is of interest is the thermal power pile operating on the 23-02 cycle; such systems are not expected to show true breeding gain, but it is hoped that they may be able to maintain the cycle and thus effectively burn thorium to yield power. Be metal, BeO, and graphite are possible moderators for units of this type; with helium cooling it may be possible to run gas turbines. Work on BeO high temperature piles has been initiated by Daniels. The temperature coefficient of reactivity in such units has been the object of study by Way, Wilkins, and Goldberger; other reactivity changes (e.g., that due to Xe) have been considered by Way. In addition to the more standard rod controls (cf. talk by Sachs) Goldberger and Wilkins have considered methods of control by gaps which create less disturbance in the pile and cooling stream temperature pattern.

[redacted]

[redacted]

52. High Pressure Water as a Heat Transfer Medium in Nuclear Power Plants

by F. Murray and A.M. Weinberg

Useful work can be extracted efficiently from a nuclear chain reaction only if the coolant operates at high temperatures. This fact leads directly to the conclusion that water at ordinary pressures is unsuitable as a power plant coolant. To avoid this difficulty inherent in water cooling, alternative heat transfer fluids such as gases or liquid metals (either of which can be raised to very high temperatures) have been proposed.

There is another coolant*, neither liquid metal nor gas, which is capable of withstanding rather high temperatures. This is water under pressure. The critical temperature of water is 374°C, its critical pressure is about 215 atmospheres. Decomposition of water under pile radiation is a phenomenon which has been studied rather extensively; the decomposition products, H_2, O_2, and H_2O_2 can be coped with satisfactorily. Corrosion by hot water is not excessive in stainless steel systems and possibly even in aluminum systems. These facts suggest that a high pressure water power plant may be built with less development work than either the gas or liquid metal plants, and that such a plant might be very reliable.

The main objection to hot water as coolant is that nowhere in such a system is there a really high temperature; the use of superheat is therefore almost precluded. This will reduce the thermal efficiency of such a plant as compared to one in which superheat is available. But the magnitude of this loss is small since during the superheating part of a steam engine cycle the heat absorption is very far from being isothermal. This means that much less is added to the efficiency of a steam cycle by superheat than would be calculated from the simple Carnot formula.

The main reason for investigating the high pressure water system is not that such a system will necessarily compete thermodynamically with either the gas or metal system. Rather it is that hot water (either light or heavy) will probably have to be used in thermal breeders or converters. Any large scale program of breeding or converting makes little sense unless the by-product power is used. Thus, even if the hot water system is less favorable as a power plant than other systems, a point will very probably arise in the development of large scale nucleonics where power from hot water will have to be extracted as a by-product of 23 production.

---

*The use of fluorocarbons or diphenyl or other low vapor pressure liquids is not considered here because the decomposition of these materials under pile conditions probably makes them very awkward to handle.

[redacted]

[redacted]

53. Resonance Energy Piles

M. L. Goldberger

Resonance energy piles are piles in which most of the neutrons are captured in the energy region from 10 to 1000 ev. Resonance piles are of interest because it is possible to obtain comparatively high breeding gains and power outputs without the extremely small size concomitant with the fast breeder (CF-3107).

We have made calculations of critical sizes of piles using Pu as the fissionable isotope and Be as the moderator. It is imagined that the pile would be cooled with liquid Na or Na-K alloy provided these coolants do not attack the Be-Pu mixture. No real design calculations have been carried out.

The greatest uncertainty in the calculation is the number of neutrons produced per capture in Pu (η_{49}); we have used the values given by Weinberg (CF-2848) multiplied by $\frac{2.95}{2.90}$ since it is now believed that the maximum η_{49} is nearer 2.95 than 2.90.

The negative laplacian for a resonance pile is given by the formula

$$\overline{\varkappa}^2 = \overline{\varkappa}_o^{\,2} \left(1 + \beta \ln \overline{k}\right)$$

$$\overline{\varkappa}_o^{\,2} = \frac{\ln \overline{k}}{\overline{\tau}}, \quad \beta = \frac{1}{2}\left(\frac{\overline{\tau^2}}{\overline{\tau}^2} - 1\right)$$

where τ is the usual Fermi age, $\overline{k}$ is the reproduction factor for an infinite pile, averaged over reproduction, $\overline{\tau}$ is the average age from fission to capture, and $\overline{\tau^2}$ is the mean square age from fission to capture. (For details, see forthcoming report by Friedman and Goldberger.)

Critical sizes have been computed for bare spherical reactors cooled with sodium which occupies the same volume as the Be + Pu. We assume a flow rate of 10 m/sec, sp. ht. = 0.3 cal/cm^3 and a temperature rise of 400°C. The flux in these piles is of the order of 5×10^{15}. The results of these calculations for three concentrations are given in the table below.

[redacted]

[redacted]

RESULTS

Pile Quantity ↓	No. atoms Be / No. atoms Pu → 50	100	150
$\bar{k}$	2.887	2.642	2.521
$\bar{\tau}$ (cm^2)	55.51	62.56	67.20
$\overline{\mathcal{H}}_o^2$ (cm^{-2})	1.924×10^{-2}	1.554×10^{-2}	1.376×10^{-2}
$\overline{\mathcal{H}}^2$ (cm^{-2})	2.065×10^{-2}	1.617×10^{-2} (est.)	1.404×10^{-2} (est.)
R_c (cm)	21.9	24.7	26.5
V_c (liters)	43.8	63.2	78.1
Total Weight (kg)	118	145.	168
Weight of Pu (kg)	41.1	30.3	25.4
Weight of Pu $\frac{1}{2}$ Na ducts (kg)	137	101	84.6
V_c ($\frac{1}{2}$ Na ducts) (liters)	292	421	521
Total Output (KW)	4.43×10^5	5.63×10^5	6.50×10^5
KW per kg of Pu	3240	5580	7580
Mean life in days	264	155	112
Loss per destruction of Pu (est.)	0.15	0.20	0.25
No. neutrons produced per destruction	2.90	2.70	2.60
Breeding Gain	0.65	0.50	0.35
Doubling Time (days)	405	310	320
"Goodness" $\frac{KW}{kg}$ x Gain	0.75	1	0.95

[redacted]

54. Pile Operations at High Temperatures

R. G. Sachs

Estimates have been made of the amount of control required for the BeO pile operating on a temperature range of 1000°C. It is found that six control rods of 3 in. diameter placed in a hexagonal pattern nine inches on an edge with an additional rod at the center can compensate a change in B of 50%. Since the total change in B from hot pile with Xe to cold pile without is expected to be about 25%, this control system should provide a considerable margin of safety.

The temperature effect on the reflector has also been considered. Increasing the temperature of the reflector increases the reactivity of the pile, thereby partly compensating the temperature effect on the pile itself. It is estimated that the effect will amount to at most a 5% change in B for the BeO pile.

JUN 1946
Clinton Laboratories
Central File
Room

[redacted]

57446