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SUMMARY

Radioelements

Hydrolysis of Plutonyl Ions In preliminary work the hydrolysis of uranyl ions is being determined as a substitute for plutonium. Previous results indicated that uranyl ions hydrolyze to form dimers. The results obtained by increasing the concentration range of the uranyl nitrate solutions indicated that the (2,2) complex was the predominant species, but that additional hydrolytic species are present. The data indicated that hydrolysis was concentration dependent.

Angular Correlation of Gamma Rays in Po^{208} Decay The angular relationships of three gamma rays closely spaced in energy occurring in the electron capture of polonium-208 are being determined. This triplet (534, 565 and 599 kev) is emitted subsequent to emission of a 291 kev gamma ray in the decay. Data were collected for six different angles, and will be reduced by computer operations to angular correlation information.

Po^{208} and Po^{209} from Bi^{209} A bismuth-209 target was bombarded with 21 mev deuterons at Argonne National Laboratory to provide an additional mixture of polonium isotopes. Approximately 0.1 millicurie of polonium-208 and polonium-209 was produced. Results of alpha and gamma ray study of this mixture will be correlated with results of other mixtures to improve energy level assignments of the two isotopes.

Alpha and Neutron Source Development

Neutron Shielding An examination was made of various neutron shielding materials from the viewpoints of optimum thermalization for counting purposes, for construction of a shield for elimination of extraneous neutrons for research purposes, and for health physics shielding. The first of these applications requires the lowest possible capture and the most desirable materials would be paraffin or polyethylene. The second application requires a high degree of neutron absorption in addition to thermalization for which cadmium provides an efficient absorber. For the third application, a boron loaded paraffin or polyethylene appears to be best because it absorbs neutrons with a minimum of resultant gamma radiation.

Alpha Source Development A procedure was developed for the preparation of high intensity plutonium alpha sources. Twenty-two alpha sources from plutonium-238 and plutonium-239 were prepared with an intensity as high as 8.0×10^9 disintegration per minute for the plutonium-238 sources. Approximately 1 mg/cm² was placed in the sources for plutonium-239. The physical sizes varies from 1.0 inch in diameter to 3.14 inches in diameter with an active areas of 1 cm² to 45 cm². The formation of a thin gold protective cover over the plutonium deposited for the high intensity sources is being studied.

Isotope Separation and Purification

CARBON ISOTOPE SEPARATION

Thermal Diffusion The eight-stage methane thermal diffusion system was temporarily converted to process enriched feed. A quantity of 44 stp liters of 54% C¹³H₄ will be separated into a fraction containing 95% C¹³H₄ and a fraction containing 1% C¹³H₄.

Chemical Exchange The new decomposer is not more efficient than the older system. Piperidine in a methanol solution was evaluated; separation was low due to remixing, caused by the low boiling point of the solvent.

STABLE GASEOUS ISOTOPES

Neon Approximately 2.5 liters of 10% neon-21 were accumulated as feed for the new three-column neon tube bundle. Calculations were started on a column profile developed from a ternary mixture.

Argon Studies were made of various sized columns for additional columns to separate argon-38.

Krypton Intermediate krypton is being reprocessed to enrich krypton-86 to 80%. This material will be used as feed for the next pass.

Xenon The calcium trap installed in the thermal diffusion column product xenon-124 line reduced the hydrogen and other impurities to 0.1%.

The heats of adsorption of krypton and xenon on Linde's Molecular Sieve 13X, 50/60 mesh, were calculated to be -1249 and -1953 cal/mole, respectively, based on separations that used nitrogen as the carrier gas.

THERMAL DIFFUSION RESEARCH

Transport Coefficients The source of the discrepancy between theory and experiment for recent thermal diffusion column experiments was found. A stagnant gas barrier appears to be present in the experimental columns. Measured composition profiles are in good agreement with theory except at the top end of the column where the composition is anomalous.

Parasitic remixing coefficients were measured as a function of wire displacement. The data do not agree with theoretical predictions for a plane parallel column.

HELIUM RESEARCH

Vapor Pressure Thermometry The generalized Weber-Schmidt equation derived previously was integrated to give the thermomolecular pressure as a function of pressure at the warm end of the tube. It has been found that round-off errors of the order of one part in 10^6 significantly alter the results. Calculations of higher precision are being made to eliminate these errors.

RADIOELEMENTS

Basic and applied research is being conducted on a number of radioelements to determine their physical properties, to develop analytical techniques, and to study the basic radiochemistry involved. Of particular interest are alpha emitters, their decay chains, their isotopes, and their chemical homologs.

HYDROLYSIS OF PLUTONYL IONS

Previous reports¹ described the hydrolysis of uranyl nitrate at 25°C in preliminary experiments to check the equipment and techniques before checking the hydrolysis of plutonyl ions in aqueous solution. The previous results showed, to a first approximation, that the primary hydrolytic product present at 25°C (ionic strengths of 0.01, 0.04 and 0.1) is the (2,2) cationic complex, i.e., $[(UO_2)_2(OH)_2]^{+2}$. The current report contains the results obtained by increasing the concentration range of the uranyl nitrate solutions, and further refinement of the mathematical techniques used for data processing.

The experimental procedure used was basically the same as described previously¹. A Beckman Model Research pH Meter with a glass-calomel electrode system was used to measure the pH of nine test solutions. The pH meter is capable of measuring pH to a relative accuracy of ± 0.001 pH units. The inert electrolyte added to hold the total ionic strength constant at the predetermined values of 0.1, 0.04 and 0.01 molar was KNO_3 . All the experiments were conducted at $25 \pm 0.05^\circ C$.

The general equation for the hydrolysis of a metal ion, in this case, the uranyl ion, is given by

It was shown previously¹ that to a first approximation, i.e., when it is assumed that only one hydrolytic species is present to any appreciable extent and that for that species $q = p$, the following equation holds:

$$\log [H^+] = \frac{q}{1+q} \log \left[\frac{M - [H^+]}{1 + Q_n(NO_3^-)} \right] + \frac{1}{1+q} \log (qQ_{qq}) \quad (2)$$

where
$$Q_n = \frac{(UO_2NO_3^+)}{(UO_2^{+2})(NO_3^-)}$$

and Q_{qq} is the formation constant q of the specie $[(UO_2)_q(OH)_q]^q$

this equation shows that if the above stated conditions are met, a plot of $\log [H^+]$ versus $\log \left[\frac{M - (H^+)}{1 + Q_n(NO_3^-)} \right]$ will yield a straight line having a slope of $\left[\frac{q}{1+q} \right]$. Previous results¹ showed a straight line relationship with $q = 2.0$ to 2.1 . These results indicated that the (2,2) species was the predominant species. Since the q value was greater than 2, one or more species in which $q > 2$ for $q = p$, and/or $q > p$ when $q \neq p$ must also be present. The inclusion of such a species into the general equation, from which Equation 2 was derived, would result in a higher value for the slope. Therefore, by neglecting the presence of this species, the calculated slopes would be less.

¹MLM-1287

During the past month, a greater number of solutions was studied. The data obtained were plotted again according to Equation 2 (see Figure 1). A straight line was obtained, but the slope changed from 0.67 to 0.58. The q value calculated from this slope was ≈ 1.4 . This result indicated that the (2,2) complex was still the predominant species, but for the other species in solution q was less than p for $q \neq p$ and/or $q = p = 1$, since $q < 2$. It was unlikely that $q = 1$ and $p \geq 2$, since this would have resulted in neutral and/or anionic species, which probably would not exist at the concentrations used in this study.

If it is assumed that, in addition to the (2,2) complex, the (1,1) complex was also present, the following derivation can be made:

$$[UO_2^{+2}] = M - \sum_{p=1}^P \sum_{q=1}^Q q [(UO_2)_q (OH)_p]^{(2q-p)} \quad (3)$$

where M = stoichiometric molarity of uranyl ion

$$[H^+] = \sum_{l=1}^P \sum_{l=1}^Q p [(UO_2)_q (OH)_p]^{2q-p} \quad (4)$$

$$Q_{qp} = \frac{[(UO_2)_q (OH)_p]^{(2q-p)} [H^{+2}]^p}{[UO_2^{+2}]^q} \quad (5)$$

If $q = p = 1$ and $q = p = 2$, the combination of Equation 3, 4 and 5 yields

$$\left[\frac{M}{[UO_2^{+2}]} - 1 - Q_n(NO_3^-) \right] H^+ = 2Q_{22} \frac{[UO_2^{+2}]}{[H^+]} + Q_{11} \quad (6)$$

Figure 1. $-\log [H^+] \text{ vs } -\log \left[\frac{M - H^+}{1 + Q_n(NO_3^-)} \right]$

where

$$[UO_2^{+2}] = \frac{M - (H^+)}{1 + Q_n(NO_3^-)} \quad (7)$$

Equation 6 predicts that if the (1,1) and (2,2) species were present, a straight line should be obtained with a slope equal to $2Q_{22}$ and an intercept equal to Q_{11} . This plot as shown in Figure 2 shows that a straight line was obtained in the concentration range of $\approx 95 \times 10^{-4}M$ to $\approx 20 \times 10^{-4}M$. The data deviated from linearity, which can be explained by the presence of an additional hydrolytic species. They also show that the hydrolysis is concentration dependent. The concentration of the third species is small from 20 to $95 \times 10^{-4}M$, and then its concentration increases as the concentration of uranyl nitrate is lowered.

The reason for this can be quantitatively seen from Equation 5. For the (1,1), (2,2) system, the $[UO_2^{+2}]$ and the $[H^+]$ concentration vary with the same exponential dependence, so that the ratio of their concentrations will not radically change. Therefore, the $[(UO_2)_q(OH)_p]^{(2q-p)}$ will remain relatively constant. As the solutions were diluted further, the $[UO_2^{+2}]$ increased and the $[H^+]$ decreased. Since q must be less than p because q was shown to be less than 2 from our first approximation, $[H^+]^p$ was decreasing faster than $[UO_2^{+2}]^q$ increased. In order for Q_{qp} to remain constant, the $[(UO_2)_q(OH)_p]^{2q-p}$ must increase to a greater extent in dilute solutions.

In order to determine the additional complex, Equations 3 and 4 were substituted in Equation 5 to yield:

$$Q_{qp} = \frac{\frac{[H^+]}{p} [H^+]^p}{\left(\frac{M - q[H^+]}{p} \right)^q} \quad (8)$$

Figure 2. Plot of Equation Resulting from Assumption of (1,1);(2,2) Hydrolytic Species.

Taking logarithms, Equation 9 results:

$$pH = \frac{-\log(Q_{qp} p)}{p+1-q} - \frac{q}{p+1-q} \log \frac{M}{(H^+)} - \frac{q}{p} \quad (9)$$

By taking various q/p ratios, a graph of the dependence of pH on $\log M/(H^+) - q/p$ may be plotted, and from the tangent of the slope the number of hydrogen and uranyl ions participating in the hydrolysis is determined. This graph² is shown in Figure 3 for $q/p = 1$

Figure 3. pH vs $\log \frac{M}{[H^+]} - 1$

As seen in Figure 3, there is a straight line dependence as predicted by Equation 9. However, it is very interesting to note that there is a change in slope as the solutions become more dilute. The change in slope occurs at $\sim 20 \times 10^{-4} M$, which is in agreement with Figure 2. The slope of this curve is only dependent on the values of q and p , and substantiates the argument that the concentration of the third hydrolytic species increased as the solution became more dilute.

The slope of the curve in the concentration range of 95×10^{-4} to $20 \times 10^{-4} M$ is approximately 2, which corresponds to the (2,2) complex. The (1,1) species be related to the (2,2) species by the following equilibrium:

$$Q_D = \frac{[(UO_2)_2(OH)_2^{+2}]}{[UO_2OH^+]^2} \quad (10)$$

In the more dilute portion of the curve, the slope is approximately 1.4, which again agrees with the previous conclusion that for the third hydrolytic species $q < p$. The complex which seems to fit best would be the (3,5). The presence of this species, in any appreciable concentration, would tend to lower the value of the slope. This conclusion is also substantiated by previous workers.³

In an attempt to quantitatively show that the (3,5) is present, curve fitting techniques are being employed. Preliminary values given in Table I were calculated by hand for the hydrolysis constants, Q_{22} , Q_{11} , Q_{35} .

Table I

U(VI) Hydrolysis at 25°C ($\mu = 0.04$)

Reaction	Equilibrium Quotient	Literature Equilibrium Quotient
(1) $UO_2^{+2} + H_2O = UO_2OH^+ + H^+$	$Q_{11} = 1.8 \times 10^{-6}$	$(2 \pm 1) \times 10^{-6}$ ref 2
(2) $2UO_2^{+2} + 2H_2O = (UO_2)_2(OH)_2^{+2} + 2H^+$	$Q_{22} = 1.3 \times 10^{-6}$	$(1.2 \pm 0.1) \times 10^{-6}$ ref 3
(3) $3UO_2^{+2} + 5H_2O = (UO_2)_3(OH)_5^+ + 5H^+$	$Q_{35} = 1.8 \times 10^{-16}$	$\approx 10^{-16}$ ref 2

²It has been shown (a) that the form of the curve is constant regardless of whether $q/p = 1, 2, 3$, etc

³Baes C. F. and Meyer J. J. *Inorg Chem*, 1, 780-86 (1962)

ANGULAR CORRELATION OF GAMMA RAYS IN Po^{208} DECAY

The decay scheme of polonium-208 is represented by the diagram shown in Figure 4. It will be noted that three of the gamma rays, rather closely spaced in energy (534 kev, 565 kev, 599 kev), are emitted subsequent to the emission of the 291 kev gamma ray, and thus, should be in time coincidence with it. In principle, this permits the measurement of the angular correlation of each of these three gamma rays relative to the 291 kev gamma ray. Such measurements could then be interpreted theoretically to yield, for example, the mixing ratios of the gamma rays involved.

Figure 4. Alpha and Electron Capture Decay Schemes for Po^{208} .

According to the theory of angular correlation, which has been thoroughly developed, if none of the gamma rays involved have a spin greater than 2, and if the relevant nuclear states all have spins no less than 2 (both of these conditions hold for the electron capture decay of polonium-208) then the angular correlation of each of the three gamma rays (mentioned above) relative to the 291 kev gamma ray will be described by an equation of the form

$$W(\theta) = 1 + A_2 P_2(\cos\theta) + A_4 P_4(\cos\theta)$$

where θ is the angle between the emission directions of the two gamma rays, A_2 and A_4 are constants which are related to the spins and parities of the nuclear states involved in the transitions and to the multiplicities and/or mixing ratios of the gamma rays involved, and P_2 and P_4 are Legendre polynomials. Since the numerical values of A_2 and A_4 can be measured experimentally, it is clear that one can obtain nuclear information providing most of the theoretical quantities appearing in A_2 and A_4 are known. The experimental values then can be used to obtain further nuclear information

In practice, of course, the situation is much more complicated than suggested above. Apart from experimental problems to be described below, one can ask whether "unique nuclear answers" are possible in an angular correlation experiment. A literature search has revealed that this is, and has been, the case only when all of the quantities appearing in A_2 and A_4 are known from other experiments with the exception of one. For example, if all spins and parities are known, and all but one of the multiplicities are known, then the remaining multiplicity may be determined through an angular correlation experiment. In the present problem none of the multiplicities and/or mixing ratios are known.

To obtain reasonable statistics for the coincidence counting data, four days of counting (including background) would be necessary for each angle. Six angles were measured using the multichannel analyzer, and these data are now ready for analysis

Analysis of Experimental Data As mentioned above, the three gamma rays of interest in this experiment are closely spaced in energy. According to conversion electron studies made earlier at Mound, they are separated by 30-35 kev in energy. The resolution of the best scintillation counter available was about 50 kev, so it is clear that the gamma rays were not resolved. The significant part of the "singles" gamma ray spectrum is shown in Figure 5. It will be noted that the part of the spectrum labeled "composite peak"

Figure 5. Singles Gamma Ray Spectrum of Po^{208} .

covers the energy range spanned by the gamma rays of interest. The same spectrum taken in coincidence with the 291 kev gamma ray is shown in Figure 6. Although the peak is far more evident in the coincidence spectrum than in the singles spectrum, careful study reveals that both the shape and width of the photo-peak are inconsistent with that produced by a single gamma ray. This supports the conclusion that more than one gamma ray is responsible for the peak.

The first part of the problem, to decompose this composite experimental spectrum into its three components, is far from trivial. One must know the energies of three gamma rays quite accurately, and one must know in detail the shape of the spectrum which would be produced by a single gamma ray of each energy. Some studies were made to see how sensitive the resulting composite peak was to both the energies and the intensities of the component gamma rays. The procedure is illustrated in Figure 7 where three peaks are indicated along with the resulting sum peak. In fact, the resulting composite peak is sensitive both to the energy and to the spectral shape. What is needed in this experiment is to obtain the intensities of the three component peaks. Unfortunately, there does not exist even one radioactive material having a gamma ray of the correct energy which could be employed to determine the correct spectral shape. Thus, the sources available had to be used, and the spectral shapes will be obtained by interpolation.

Standard spectra have been taken for beryllium-7, chromium-51 and strontium-85. The gamma rays present in these sources nicely straddle the region in question (in the neighborhood of 565 kev). The spectra will be plotted together, and a spectrum for each of the three energies (534 kev, 565 kev, 599 kev) obtained by interpolation.

The procedure outlined above must be done for each of the six angles for which coincidence data were obtained. Using the standard spectra to obtain the intensities at each angle, one is then in a position to study the angular dependence for each of the three gamma rays separately.

Figure 6. Coincidence Spectrum.

IBM 1620 Computer Application The best method for determining the relative intensities of the three component gamma rays which contribute to the composite peak from a knowledge of the (interpolated) spectral shapes of each gamma ray and a knowledge of the experimental spectrum seemed to be to employ the method of least squares. Thus, one postulates intensities for the three gamma rays, calculates a sum spectrum, subtracts this calculated spectrum from the experimental spectrum, and then minimizes the squares of the residuals. Clearly, such a job is too long and tedious to do by hand, so a computer program was written and tested which will accomplish the task quickly and accurately.

Given the relative intensities of the three gamma rays at each angle (as calculated on the computer) together with the total composite intensity, it is then desired to obtain the best experimental fit of the angular distribution for each gamma ray by an expansion of Legendre polynomials so that the result may be compared with the theory. Again this task, which also involves least square analysis, was deemed to be too difficult to do by hand, so a computer program has been written and tested which will accomplish the task.

Figure 7. *Simulated three Photon Peak (numbers on curves refer to peak channels).*

Po^{208} and Po^{209} from Bi^{209}

A stacked bismuth target was bombarded with 21 mev deuterons for 10 hours at Argonne National Laboratory in August. The front foil consisted of a 0.003-in. thick deposit of bismuth metal sputtered on a 0.005-inch thick copper disc. The back-up target was a 0.050-inch thick disc of bismuth metal. It was estimated that about 0.1 millicurie of polonium-208 and -209 would be produced in the front foil and perhaps a tenth of this in the back-up foil. The polonium-208 to polonium-209 ratio in the front foil was projected to be many fold greater than any material studied thus far.

After the material was received, the polonium was purified chemically. Substantial quantities of nuclides, such as copper-64 and zinc-65, were produced in the copper backing, and even trace amounts of these impurities in the polonium fraction would be detrimental.

Spectral determinations of alpha particles, conversion electrons and gamma rays were made with the thin front foil. Although these data have not been analyzed completely at this time, several typical spectra are noteworthy.

A gamma spectrum obtained on a 3 x 3-inch NaI(Tl) detector, in Figure 8, was made using a 30 minute measurement inside the two-inch lead shield. The familiar peaks at 76, 260, 580 and 900 kev are evident. In addition, the 803 kev gamma-ray from polonium-210 and the significant broadening from the 295 kev photons in polonium-208 are observed. Broadening on the high energy side of the 260 and 585 kev peaks are believed to be caused by x-ray gamma-ray summation from the high geometry.

The peaks at 160 and 210 kev were caused by scatter inside the lead shield. This portion of the spectrum was observed in detail with a 1/8-inch thick, thin window NaI(Tl) crystal. Because of its size, the direct to scatter ratio of the crystal should be significantly less than that of the 3 x 3-inch detector. The absence of these two peaks in spectra with the smaller detector confirmed that they were caused by scatter.

Figure 8. Gamma Spectra of Po^{208} and Po^{209} from August 1965 Deuteron Bombardment of 3-mil Bi^{209} Target (30 minute measurements).

Figure 9 is a conversion-electron spectrum obtained on a 110 mm² lithium drifted silicon detector at room temperature over a 5000 minute measurement interval. All the main peaks in this spectrum were observed in earlier work, and further confirm the assignments made in those studies. Some possible structure exists between the triad of peaks at 443, 474, 508 kev and the 810 kev electron line. Unfortunately, the counting rate was too low and the resolution was too poor to more positively identify possible lines in this energy range. Resolution was estimated as 25 kev during this run.

Figure 9. Conversion Electron Spectrum of Po^{208} - Po^{209} from August 1965 Deuteron Bombardment of 3 mil Bi^{209} Target Using 110 mm² Lithium Drifted Silicon Detector (5000 minute measurement).

ALPHA AND NEUTRON SOURCE DEVELOPMENT

Special alpha and neutron sources are fabricated from the radioisotopes polonium-210 and plutonium-239. The techniques developed in the fabrication of these sources are supplied to American Industry. In addition, basic nuclear studies are being made on these sources.

NEUTRON SHIELDING

The neutron shielding ability of several different materials was examined using a plutonium-239 beryllium fast neutron source (total emission of 1.24×10^6 neutrons per second). A Texas Nuclear helium-3 proportional chamber filled to a pressure of approximately one atmosphere was used as the detector. This type of chamber, essentially a thermal neutron detector, is insensitive to gamma rays. The neutron source was located a few centimeters from the detector. Calculations were made of the neutron flux from the source at the helium-3 detector. Then, various shielding materials were located between the source and the detector, and the neutron attenuation was measured.

The problem of neutron shielding depends upon the viewpoint of the individual making the analysis. In the construction of a "wide range" neutron detector, a thermal neutron detector surrounded by an adequate amount of shielding material to produce a maximum count rate is used. In such a case, the curves presented in Figure 10 and the data tabulated in Table 2 would be of interest. From an examination of Figure 10 and Table 2, the use of polyethylene would require the least amount of material. On the other hand, the maximum neutron count rate is obtained with paraffin. Even so, polyethylene is the easier to fabricate into a hollow cylinder into which the detector can be placed.

Figure 10. Relative Neutron Count Rates for Various Shielding Materials.

Table 2

Relative Neutron Count Rates for Various Materials

Material	Approximate Thickness to Produce Maximum Count	Relative Neutron Count Rate
Paraffin	7.5 centimeters	1.00
Benelex 70	10.5	0.95
Polyethylene	7.0	0.90
Plexiglas	8.8	0.89

Another aspect would be the construction of a shield in which a neutron counting experiment is to be conducted. Here, the reduction of neutrons external to the experiment would be important and the research would be interested in such information as that contained in Figure 11 and Table 3. Assuming the use of a "gamma ray insensitive" neutron detector, a cadmium liner can be employed between the counting chamber and the shielding material. From a consideration of the thickness of material required to produce a minimum count rate (Table 3), either polyethylene or paraffin would be adequate.

Figure 11. Relative Neutron Count Rate for Various Shielding Materials.

Still another viewpoint would be that of the health physicist. In this case, the concern would be that of the dose rate received by an individual working behind a shielding material. Computations of the relative neutron dose rates through several different materials included a consideration of their ability to thermalize and capture the fast neutron emission from the source. The results of this phase of the study are presented in Figure 12. The minimum possible shielding thickness and its respective relative neutron dose

Table 3

Relative Neutron Shielding Ability

Material	Minimum Possible Thickness (centimeters)	Relative Neutron Count Rate
Borated Polyethylene	<5	1.00
Plexiglas*	5.5	0.26
Benelex 70*	6.5	0.27
Boral	~1	0.70
Paraffin*	~4	0.26
Polyethylene*	4.0	0.22

*0.07 cm of cadmium used as a "liner".

Figure 12. Relative Neutron Dose Rates for Various Shielding Materials as Compared with the Neutron Dose Rate from an Unshielded Pu^{239}Be Source (total emission of 1.24×10^6 neutrons/sec. The neutron dose rate from the Pu^{239}Be source was taken as unity).

rate is tabulated in Table 4 for each material. In employing the results provided in Table 4 and Figure 12 the health physicist must include a consideration of this particular shielding problem. For general shielding of this fast neutron source, less than 5 cm of borated polyethylene appears to be the best of those examined, since the most energetic Al capture gamma which originates in the Boral is 7.724 mev, and would increase the total gamma dose. On the other hand, the most energetic boron capture gamma is only 0.478 mev.

Table 4
Relative Neutron Shielding

<i>Shielding Material</i>	<i>Minimum Possible Thickness (centimeters)</i>	<i>Relative Neutron Dose Rate</i>
None	—	1.00
Boral	~1	1.5×10^{-4}
Borated Polyethylene	<5	2.1×10^{-4}
Polyethylene	7.0	9.7×10^{-4}
Plexiglas	8.8	9.7×10^{-4}
Paraffin	7.5	1.1×10^{-3}
Benelex 70	10.5	1.0×10^{-3}

The use of 0.07 cm of cadmium behind such materials as polyethylene, Plexiglas, paraffin and Benelex 70 does decrease their respective relative neutron dose rates to values of less than those computed for Boral and borated polyethylene. On the other hand, the relative gamma ray dose rates are increased as a result of the introduction of the "cadmium capture gamma".

ALPHA SOURCE DEVELOPMENT

Previous attempts to prepare high intensity alpha sources by electroprecipitation from an acid solution have not been very successful. The few literature references available on this subject are contradictory in that individual results have not been reproducible.

One of the big problems is the proper solution which is necessary to prepare depositions of one milligram (or more) per cm² and maintain a high deposition yield. The only literature reference found concerning large depositions was that of Miller and Brouns⁴. In this report, plutonium(VI) in 1.5-2 N KOH was reduced to plutonium(IV) by electrolysis and the plutonium hydrous hydroxide was deposited on the cathode. By this technique, Miller and Brouns claimed to have deposited up to 1-2 mg/cm² of the plutonium compound on platinum disks.

The use of a basic solution from which the plutonium could be electroprecipitated appeared to offer two distinct advantages over past investigations in an acid media.

- (1) When using an acid media, the mechanism of electroprecipitation involves the production of hydroxide ions at the cathode. The presence of a high hydroxide ion concentration at the cathode results in the precipitation of the plutonium hydrous hydroxide at the electrode surface. However, as the electrolysis continues, the pH of the bulk solution is also raised, resulting in the precipitation of the plutonium from the bulk solution and limiting the amount of plutonium precipitated at the electrode surface.
- (2) The plutonyl ion must be stabilized in a basic solution probably as a hydroxy complex ion (no literature reference has been found to support this fact) such that the deposition is no longer dependent on the formation of the hydroxide at the electrode surface. The mechanism most probably taking place in this medium is the reduction of the plutonium(VI) to plutonium(IV) at the electrode surface and because of the excess hydroxide present there is an immediate precipitation of plutonium(IV) hydrous hydroxide.

Because of these factors, it was decided to attempt to prepare high intensity alpha sources from a basic solution containing plutonium(VI). The following is a brief outline of the procedure used in this investigation:

Approximately 1-10 mg of either plutonium-239 or plutonium-238 (in an HCl medium) was placed in a 100-ml beaker along with 10-12 drops HNO_3 . The resulting solution was heated on a hot plate until the final solution volume was 1-2 ml. To the nitrate solution, 0.5 ml of 1 M NaBrO_3 solution was added and the mixture was again heated until the color of the solution turned from a blue to a light brown. Approximately 10 ml distilled water was added and the solution allowed to cool. The acidity of the plutonium solution was adjusted by adding 4 N KOH slowly until a slight amount of precipitate was observed, and then one drop of conc HNO_3 was added. The resulting solution was transferred to the electrolysis cell which contained the required volume of 2 N KOH. The solution was electrolyzed for varying periods of time and current density. The cathode was removed from the cell, washed twice with distilled water and pressed dry with Kimwipes. The sources were removed from the hood, the edges and back were wiped with acetone and then counted. After the sources were counted, they were ignited at 500-550°C for 24 hours, cleaned and re-counted.

A total of 21 sources were made, varying in depositions from 4 μg to 1 mg over areas varying from 1 to 42 cm^2 (the intensity of the sources varied from 10^{11} to 10^7 d/m). The larger sources, after ignition, had a tendency to wipe off, i.e., only 10-15% remained, whereas the smaller sources retained essentially all of their precipitated activity. The yield of the electroprecipitation was quite low for the larger sources, (approximately 5%) and the yield increased as the concentration of the original electrolysis solution was decreased, up to 50% for the smaller sources.

Different cathode materials were used such as type 304 stainless steel, nickel, and platinum. Little can be said concerning the use of nickel cathodes since too few experiments were run using this substrate. The efficiency of electroprecipitation on stainless steel appears to be much greater than that of platinum; however, the experiments were not primarily designed for testing substrate efficiency.

A few of the larger sources were gold plated in an attempt to form a protective cover; the results were not favorable, but again the number of trials were too few for conclusive results.

ISOTOPE SEPARATION AND PURIFICATION

Under the sponsorship of the Division of Research, Mound Laboratory is responsible for furnishing material to be used in the physical sciences to further the progress of science and technology in the public interest. After recovery, purification and concentration, research quantities of isotopes are furnished to installations for research programs in the physical sciences. Purified helium-3, the isotopes of other noble gases, and carbon-13 are available for distribution in research quantities. Methods of separation and purification are being studied.

CARBON ISOTOPE SEPARATION

Thermal Diffusion The eight-stage methane thermal diffusion system was temporarily converted from natural abundance feed to partially enriched feed. A quantity of 44 stp liters of 54% $C^{13}H_4$ will be separated into two fractions, one which contains 95% $C^{13}H_4$ and one which contains less than 1% $C^{13}H_4$.

The cascade configuration to be used for the separation is shown in Figure 13. Three of the four first stage columns of the natural abundance cascade have been dropped as superfluous and the feed point was moved from stage 1 to the middle of stage 4. These changes were guided by cascade performance calculations reported earlier⁴. The feed point chosen for operation, however, was $\frac{1}{2}$ stage below the calculated optimum. This choice was made primarily for mechanical convenience. The calculations show that only a slight reduction in product flow will result.

Operation with 54% feed requires a much higher system holdup of $C^{13}H_4$ than does operation with natural abundance feed. The additional holdup was estimated⁴ at 4 stp liters. This additional holdup was accumulated in the following way.

Product flow was stopped when the cascade was switched to enriched feed. Waste or depleted methane was continuously removed from the top of the cascade at an average rate of 520 stp ml/day. Enriched methane was fed at the same rate at the feed point indicated in Figure 13. Thus, over a period of 14 days 7.3 stp liters of 54% $C^{13}H_4$ entered the cascade while 7.3 stp liters of 0.4% $C^{13}H_4$ were removed in the waste stream. The net addition to the cascade was 3.9 stp liters of $C^{13}H_4$.

At the conclusion of the above operation the depleted methane flow was reduced to 300 stp ml/day and product flow was started at approximately the same rate. These flow rates will be adjusted as operating experience is acquired.

Chemical Exchange Two series of two reaction runs were completed on the carbon dioxide-carbamate chemical exchange system. The first series consisted of two runs using monoethanolamine in methanol, the conditions were identical to some of the reactions in the past. The purpose of these runs was to compare the efficiency of the new decomposer system with the older system.

Figure 13. Methane Cascade Configuration for Separation with Enriched Feed

⁴MLM-1274

Figure 14. Comparison of two Similar Reaction Runs with Different Decomposer.

The results of the comparison are shown in Figure 14. Within experimental and analytical error, the results of the two curves show the decomposers to be of about the same efficiency. Further examination of the decomposed carbamates for carbon dioxide content will better reveal the difference in efficiency between the two decomposers.

In the second series of reaction runs, piperidine was evaluated. Piperidine is a stronger base than the monoethanolamine, and was more difficult to decompose under similar operating conditions. There was no improvement in separation using the piperidine in the methanol solvent, but the remixing caused by incomplete dissociation of the carbamate was greater, as expected. A preliminary carbon dioxide analysis of the decomposed carbamate from one of the piperidine runs showed that over 1% carbon dioxide was retained by the decomposed carbamate. Stronger heating conditions are necessary to dissociate this carbamate more completely. This will necessitate running the reaction in a higher boiling solvent.

STABLE GASEOUS ISOTOPES

Neon After 109 successive days of operation, the four-stage neon cascade was shut down due to a column leak caused by an electrical short at the top of a column which damaged the Nylon plug. To date, 2.5 liters of neon-21 enriched to 10.9% have been accumulated for feed into the new 36-foot tube bundle.

In preparation for the start-up of the new tube bundle, the tubular heaters were heated to 750°C before the hot wall temperature static tests were run. During the test, the system will be run at static conditions until the column comes to equilibrium at various pressures. The middle column will then be sampled at the top and bottom to obtain data to plot the logarithm of the separation factor as a function of pressure. These data will be compared to theoretical predictions.

The column profile of the new system using ternary thermal diffusion theory is being calculated. In order for the computer program to calculate accurately the profile of the three isotopes in the column, all the parameters of the system, especially the dead volume, must be known. The column measurements and dead volume have been accurately calculated, and are given in Figure 15. Knowledge of the entire volume of the

Figure 15. Dead Volume in the 36-foot Neon Cascade. (The cold-wall diameter is 0.995 inch and the tubular heater diameter is 0.314 inch. The water volume of the system is 6026 ml. The dead volume of the system is 21%).

system permits the calculation of the exact amount of gas put into the system, and provides data for calculating an accurate column profile. Of the 13 feet of tubular heater length, only 11 feet 11 inches of the heater are heated to 750°C. Therefore, only 11.9 feet of the column are active during thermal diffusion separation. This volume is called the hot volume. All other volumes, including that at the cold area along the heater at the top and bottom of the column and all circulating lines, are called the cold volume. The dead volume, which is actually the ratio of the inactive to the active volume, has been defined as follows in this work,

$$\text{dead volume (\%)} = \frac{\text{cold volume}}{\text{hot volume}} \times 100.$$

Argon To enrich argon-38 more than that obtained from the four-stage cascade, studies are being made for the addition of a tube bundle. Since argon-38 is a middle isotope of low abundance, the two criteria of a new system are; (1) that it uses a reasonable amount of column hold-up gas, and (2) that separation comparable to the present system is obtained. Several columns with various cold wall diameters have been studied.

The logarithm of the separation factor per centimeter of column length as a function of pressure is shown in Figure 16. The results indicate that either ¾-inch extra heavy or standard copper pipe can serve as a suitable cold wall and still give a reasonable column operating pressure. The ¾-inch extra heavy copper pipe is easier to fabricate than the standard copper pipe.

Calculations were made to determine how much the separation may be increased by putting tubular heaters into the 24-foot, 11-column cascade. Figure 17 shows the logarithm of the separation factor per centimeter of column length as a function of pressure for the present column and for the column with a tubular heater. The curves show that replacing the 1/16-inch wire with a 0.314-inch tubular heater would increase the separation factor by roughly 70% but moves the operating pressure from the present 400 torr to 1000 torr. This increases the column volume by a factor of 2.5.

Krypton A two-column, two-stage thermal diffusion cascade is being used for enriching krypton-84 and -86. The present step of reprocessing the waste stream to obtain intermediate feed of 80 per cent krypton-86 was interrupted due to mechanical difficulties. When intermediate krypton stock has been depleted in krypton-86 content, the 80 percent krypton-86 will be used as feed for the final pass.

Figure 16. Separation/cm of Column Length as a Function of Pressure for Various Cold Wall Radii Using a 0.40 cm Radius Tubular Heater. (Comparison is made to the 3/4-in column used in the present argon separation system)

Figure 17. Separation as a Function of Pressure for the Argon Columns. (The two curves are a theoretical comparison using two different heating elements).

Xenon

Thermal Diffusion The calcium trap installation, shown in Figure 18, effectively removed contaminants from the xenon-124 product in the 24 tube bundle. Results are given in Table 5.

Table 5
Performance of Calcium Trap

Component	Analysis, Mol %	
	Prior to trap installation	5 days after trap installation
Xenon	0.25	99.90
Hydrogen	99.53	0.04
Water	0.14	0.01
Nitrogen	0.07	0.05
Argon	0.01	—

Figure 18. Xenon Calcium Trap in a Tube Bundle Cascade.

A leak interrupted enrichment of xenon-124 beyond the level reported last month. In order to reduce pressure surging in the first stage and to prevent an upset from disruption of compressed air supply, a pneumatic control valve was replaced by an in-line regulator.

The six stage swing unit is being operated to accumulate approximately five liters of xenon-136 enriched to 75-80% for use as feed for a second pass to enrich this isotope to 90%. Approximately 700 ml of xenon-136 enriched to about 75% are being drawn off each week. To date, approximately two liters have been accumulated.

Experimental hot wire temperature data reported previously⁵ was compared with theoretical data from the program which computed the temperature gradient between the hot and cold wall from thermal conductivity, radii and other input data. Figure 19 shows that the experimentally determined hot wire temperatures were approximately 20-30°C higher than the theoretical temperatures.

Figure 19. Hot Wire Temperature for 24-ft. Xenon Thermal Diffusion Columns.

Chromatography Xenon obtained as a reactor off-gas contains the radioisotope krypton-85 as an impurity. This impurity is removed chromatographically. Continuing research in this area is directed at further reduction of the krypton-85 level while at the same time increasing the xenon yield.

A series of separations on Molecular Sieve 13X, 50/60 mesh, at 166°K, 210°K, and 243°K was completed. Nitrogen was used as the carrier gas. The calculated heats are -1249 cal/mole and -1953 cal/mole for krypton and xenon, respectively, which is very close to those found for Molecular Sieve 5A, 50/60 mesh, in an identical series of separations, -1208 cal/mole and -1901 cal/mole, for krypton and xenon respectively. Molecular Sieve 13X is superior to Molecular Sieve 5A, however, since the 13X exhibited considerably less peak tailing than the 5A. It follows that 13X would produce cleaner xenon than 5A.

⁵MLM-1254

THERMAL DIFFUSION RESEARCH

Transport Coefficients

Convective and Diffusive Remixing Terms Two sets of measurements are required to determine the coefficients H' , K'_c , and K'_D in the differential equation for isotope transport in a thermal diffusion column,

$$\tau = H' p^2 c(1-c) - (K'_c p^4 + K'_D) \frac{dc}{dZ} \quad (11)$$

where p is pressure, c is fraction of one of the isotopes and Z is distance along the column. The required experiments are (1) measurement of static separation as a function of pressure and (2) measurement of separation as a function of flow rate.

Over a period of time various sets of static separation data for $\text{Ne}^{20}\text{-Ne}^{22}$ in the four 12-ft (3.65 m) experimental columns⁶ have been in excellent agreement with theoretical predictions based on Lennard Jones shape factors. The results of flow experiments, however, have been persistently in poor agreement with theory. Last month⁷ it was postulated that a stagnant diffusion barrier was present at the top of each column, and calculations were made to show that the experimental discrepancies could result from the presence of such barriers.

This postulate was verified in a recent series of experiments. Gas flows were set up as in all previous experiments, i.e., gas was circulated at a high rate across the top of the experimental column while a stream of gas was withdrawn at a measured slow rate from a port near the bottom of the column. In the recent series of experiments, however, gas samples were taken at intermediate points along the column in addition to the usual samples of the gas in the circulating loop and of the gas withdrawn from the bottom.

The experimental setup and a typical set of results are depicted in Figure 20. The anomalous concentration resulting from diffusion across a stagnant layer at the top of the column is immediately obvious. (The solid curve in Figure 20 is a profile calculated from the measured product composition with theoretical values of the column transport coefficients.) It is quite clear that the poor agreement with theory observed in earlier experiments resulted from use of the anomalous composition to obtain H' from the experimental data.

In all future flow experiments compositions will be measured at intermediate points and the evaluation of H' will be based on these compositions rather than the top composition.

Parasitic Remixing Parasitic remixing effects are known to result from asymmetric thermal diffusion column geometry and temperature distribution. The observed separation is very sensitive to small devi-

Figure 20. Flow Experiment with Column E1 (1359 STP ml/hr).

⁶Dimensions and operating conditions for these columns are given in MLM-1245.

⁷MLM-1297

ations. Dickel⁸ has shown experimentally that the deviations have an unimportant effect on H' but a large effect on the convective remixing part of Equation (11), i.e., the coefficient of p^4 .

The transport for an asymmetric column is thus

$$\tau = H' p^2 c(1-c) - [(K'_c + K'_p)p^4 + K_d] \frac{dc}{dZ} \quad (12)$$

where K'_p is the parasitic remixing coefficient. Very little experimental work has been reported on the problem of relating K'_p quantitatively to deviations from symmetry.

A series of experiments, the object of which is to relate K'_p to known deviations from symmetry, is now in progress. The six-foot (1.82 m) experimental column is being used. Static separations as a function of pressure measurements are being measured for Ne²⁰-Ne²² separation at wire displacements of 0, 0.16, 0.32, and 0.48 cm from the axis of the column. In these experiments, special blocks were made to hold the center wire at a known displacement throughout the entire active length. Data available to date from this series are plotted in Figure 21. The data obtained at a 0.48 cm displacement are not complete and will be reported later. The values obtained thus far appear in Table 6. The ratio $(K'_p + K'_c)/K'_c$ is plotted against displacement divided by column radius in Figure 22. A result obtained earlier in an experiment with one of the 12 foot columns⁹, is also plotted in Figure 22.

Figure 21. Separation Fraction per Unit Length as a Function of Pressure for On-and-Off-Center Wires in the Six-Foot Experimental Column.

⁸Dickel, G. and Burkholz, A. *Z. Naturforsch.* 16a, 760 (1961)

⁹MLM-1238

These preliminary results can be compared to a theoretical treatment by Dickel¹⁰ which states that

$$\frac{K'_c + K'_p}{K'_c} = 1 + \frac{47}{6} \left(\frac{\epsilon}{\Delta x} \right)^2 \quad (13)$$

where ϵ is the displacement and Δx is the nominal distance from the hot wall to the cold wall. This relationship is also plotted in Figure 22. It appears that this theory does not account for the experimental observations; however, derivation of the theory was based on plane-parallel geometry, thus an arbitrary extension of the theory to the extreme cylindrical geometry of these experiments is not justified.

Table 6

Coefficient Ratios for Asymmetric Thermal Diffusion Column Geometry

Wire displacement (cm)	$\frac{K_d}{H'}$	$\frac{K'_c + K'_p}{H'}$	$\frac{K'_p + K'_c}{K'_c}$	Theoretical	
				$\frac{K_d}{H'}$	$\frac{K'_c}{H'}$
0	38.2	29.3	—	37.9	32.9
0.16	30.5	68.4	2.33	—	—
0.32	36.1	126	4.30	—	—

Figure 22. Parasitic Remixing as a Function of Wire Displacement.

¹⁰Dickel, G., Z. Naturforsch. 16a, 755 (1961).
¹¹MLM-1287

HELIUM RESEARCH

Vapor Pressure Thermometry The generalized Weber-Schmidt equation derived in last month's report¹¹ was integrated to give the thermomolecular pressure as a function of the pressure at the warm end of the tube. The integration, following Weber and Schmidt, was accomplished by transforming

$$\frac{dP}{P} = \frac{1}{2} \frac{dT}{T} \left(\frac{\pi}{24K_1} (y^2 + 4K_2 y) + \frac{1 + gy}{1 + hy} \right)^{-1} \quad (14)$$

where

$$y = \frac{r}{\lambda} = rP \left(\frac{T}{T_0} \right)^{1+en} \text{ ex constant} \quad (15)$$

to

$$-\frac{dP}{P} = dy \left(\frac{A}{y} + \frac{B}{y + m_1} + \frac{C}{y + m_2} + \frac{D}{y + m_3} \right) \quad (16)$$

by the method of partial fractions. The constants in Equation 16 are related uniquely to those in Equation 14. Several useful relations appear in reducing Equation (14) to (16). Two of them that will bear on the present discussion are

$$-A = B + C + D, \quad A = \frac{I}{2n + 1} \quad (17)$$

and

$$Bm_1 + Cm_2 + Dm_3 = 0 \quad (18)$$

Equation (17) allows A to be eliminated from Equation (16). Equation (16) is easily integrated to give

$$\ln \frac{P_w}{P_c} = \frac{1}{2} \ln \frac{T_w}{T_c} - \frac{2n + 1}{2(n + 1)} \left[B \ln \frac{y_w + m_1}{y_c + m_1} + C \ln \frac{y_w + m_2}{y_c + m_2} + D \ln \frac{y_w + m_3}{y_c + m_3} \right] \quad (19)$$

which is the general form of the Weber-Schmidt equation.

Equations (19) and (15) are solved to give the thermomolecular pressure ΔP as shown in Figure 23 where the four curves represent four different values of the constant in Equation (18). The disturbing features of Figure 23 is that none of the curves go to zero as P_w gets large and that the effect changes signs. The reasons for this behavior in the high pressure limit can be seen from Equation (19), which can be rewritten as,

$$\frac{P_w}{P_c} = \left(\frac{T_w}{T_c} \right)^{1/2} \left[\frac{y_w + m_1}{y_c + m_1} \right]^B \left[\frac{y_w + m_2}{y_c + m_2} \right]^C \left[\frac{y_w + m_3}{y_c + m_3} \right]^D \quad (20)$$

where $N = (2n + 1)/2(n + 1)$. In the limit of P_w large, $y_c \gg y_w \gg 1$, and since $m_1 \approx m_2 \approx m_3 \approx 1$,

$$\left(\frac{P_c}{P_w} \right)^{1-N(B+C+D)} = \left(\frac{T_c}{T_w} \right)^{1/2 + (2n+1)/2 (B+C+D)} \left[1 + \frac{Bm_1 + Cm_2 + Dm_3}{y_w} \right]^{N(B+C+D)} \quad (21)$$

¹¹MLM-1287

Figure 23. Round Off Errors in Weber-Schmidt Calculation.

Now according to Equation (17), $(2n+1)/2 (B+C+D) = -1/2$ and from Equation (18), $Bm_1 + Cm_2 + Dm_3 = 0$. Therefore, P_w/P_c should equal 1. In considering the reasons for $P_w/P_c \neq 1$, first assume that the temperature exponent is exactly 0 and look at the effect of Equation (18) being not exactly zero. Then, since $y_w \gg 1$ and $y_w = \alpha P_w$,

$$\frac{P_c}{P_w} = 1 + \frac{N(B+C+D)}{1-N(B+C+D)} \left(\frac{Bm_1 + Cm_2 + Dm_3}{\alpha P_w} \right) \quad (22)$$

or

$$P_w - P_c = - \frac{N(B+C+D)}{1-N(B+C+D)} \frac{(Bm_1 + Cm_2 + Dm_3)}{\alpha} = \text{Const.} \quad (23)$$

Thus $Bm_1 + Cm_2 + Dm_3 \neq 0$ causes the constant non zero values of ΔP at large P in Figure 23. Secondly, consider that Equation (18) holds exactly and $(2n+1)/2(B+C+D) \neq -1/2$. The divergence of Curves II and III for $P_w > 10^5 \mu$ is caused by $(2n+1)/2 (B+C+D)$ differing from $-1/2$ by ± 0.00001 . These errors are being eliminated by doing double precision computer calculations of ΔP .