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SUMMARY

Radioelements

Uranium-234 Separation and Purification Two aged plutonium samples were processed and 84 milligrams of uranium-234 were recovered. A third sample containing two grams of uranium-234 is currently being processed. Another anion exchange run was made, but analyses are not complete.

Isotope Separation

Helium-3 Thermometry An error was found in the computer program for calculating the thermomolecular pressure gradient of helium-3 and helium-4 from the Weber and Schmidt equation. This error was subsequently corrected, and plots of the thermomolecular pressure gradient as a function of cold temperature were drawn.

The effect of varying the cold and warm temperature in the Weber and Schmidt equation was investigated. The results for saturated helium-3 vapor were only slightly dependent on the room temperature and highly dependent on the cold temperature.

Two experimental runs were made with a modified Dewar system, one with helium-4 and one with helium-3 in the vapor pressure bulb. The resulting thermomolecular pressure differences were measured and compared with calculations based on the Weber-Schmidt equation.

Plastics Research

Four mill lots of a standard asbestos-filled diallyl phthalate formulation were prepared, each containing two parts of a different silane coupling agent. The only significant improvement noted was in the formulation modified with vinyltriethoxy silane, which had a heat distortion value of 214°C as compared to an average of 165°C for standard DAP formulations.

Foams Evaluations Improved epoxy foams were prepared by dissolving diaminodiphenyl sulfone in heated epoxy resin. The samples remained rigid at 330°C. A study of the effect of component ratios on gel time and exotherms was inconclusive. Diaminodiphenyl sulfone and trimethylboroxine do not form a complex when heated together as was previously believed.

Analytical

An automatic sample loading device for a calorimeter was constructed. This device, in conjunction with a constant current supply for the heaters, has made it possible to make unattended assays of high precision and accuracy. Perhaps of greater importance, this device made it possible to assay a given sample indefinitely in a hermetically sealed system. Operating the calorimeter in a constant atmosphere seems to eliminate the slow variations which are now known to be caused by variations in humidity in the case of Calorimeter 51.

Experiments were performed with a plutonium-beryllium neutron source to determine techniques for observing low intensity n- γ interactions in the presence of relatively high level gamma emission from the neutron source itself. It was possible to show the 2.2 Mev gamma that results from the neutron capture by hydrogen in water.

An actinium fluoride sample, approximately eleven years old, is being counted to determine the sodium-22 content. Sodium-22 (half-life = 2.58 years) results from the α -n reaction in fluorine-19.

The 1.28 Mev gamma photon from sodium-22 was tentatively identified in the gamma energy spectrum, but the positron annihilation peak at 0.51 Mev could not be detected in the large background of gamma energies arising from the actinium-227 decay chain.

RADIOELEMENTS

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

URANIUM-234 SEPARATION AND PURIFICATION

Milligram quantities of uranium-234 are being separated from aged plutonium-238 solutions. The uranium is separated from the bulk of the plutonium in nitrate solution by an anion exchange process. Extraction of the uranium into hexone (methyl isobutyl ketone) further reduces the concentration of plutonium and most other impurities. Finally thenoyltrifluoroacetone (TTA) extraction removes the last traces of the plutonium from the uranium. To date, two aged plutonium samples have been processed using this method and 84 milligrams of uranium-234 have been recovered. A sample containing two grams of uranium-234 is being processed through the ion exchange step.

Solutions from the dissolution of a SNAP-3 heat source containing 24.23 grams of plutonium were used as feed solution for the third ion exchange run. Also, raffinates from two previous runs containing 1.8 grams of plutonium were loaded on the column. The ion exchange procedure was the same as earlier runs except that the flow rate was increased to 15-20 ml per minute, and a larger volume of 7.2 N HNO₃ wash was used. Raffinate solutions were collected until the plutonium started to elute, then the first 450 ml of elutriate were collected and held separate. About 4.5 liters of dilute HNO₃ was used to elute the uranium, and this solution was evaporated to less than one liter before analysis. The ion exchange resin showed signs of degradation and decreased capacity and was discarded at the end of the run. Although the solutions have not been completely analyzed, preliminary data indicated that the uranium was being removed, but the plutonium contamination was high. A detailed discussion will be deferred until all the analyses are complete.

A part of the SNAP-3 material had remained as the metal or an alloy in a stainless steel container. This material was dissolved in HCl and converted to the nitrate. The solution was estimated to contain 40 grams of plutonium and is being prepared for future ion exchange runs.

Cold work on the anion exchange behavior of uranium on Dowex 1X4 resin was continued. A feed solution containing 86.3 mg of uranium in 400 ml 7.2 N HNO₃ was loaded on the resin at a flow rate of 6 ml per minute. Samples of the raffinate were collected after each 50 ml passed through the column. The column was washed with one liter of 6.5 N HNO₃ at a flow rate of 10 ml per minute, and samples were collected after every 100 ml. The column was eluted with 500 ml 9.35 N HNO₃ at a flow rate of 9 ml per minute, and samples collected after every 50 ml. Uranium in these samples was determined by the indirect PAN method. The results, shown in Table 1, indicate that 83.1 milligrams or 96.3% of the uranium was recovered with 93.5% in the feed and washed raffinates.

Table 1

URANIUM DISTRIBUTION

Feed Solution		Wash Solution		Elution Solution	
Vol. (ml)	U (mg)	Vol. (ml)	U (mg)	Vol. (ml)	U (mg)
0 to 200	nil	500	8.55	1250	0.93
250	0.04	600	14.25	1300	0.76
300	0.36	700	15.70	1350	0.28
350	0.99	800	13.05	1400	0.24
400	<u>1.94</u>	900	9.55	1450	0.21
	3.33	1000	10.10	1500 to 1700	<u>nil</u>
		1100	4.10		2.42
		1200	<u>2.05</u>		(2.80%)
			77.35		
			(89.6%)		

Dowex 1X4 7.2 N Feed 6.5 N Wash

ISOTOPE SEPARATION

Processes are being developed for separating and purifying the isotopes of a number of elements including the noble gases and carbon. Potential sources of supply of these materials are being evaluated.

HELIUM-3 VAPOR PRESSURE THERMOMETRY

Mound Laboratory is cooperating in an effort to establish the vapor pressure of helium-3 as an international temperature standard. Mound Laboratory's contribution to this effort will be to supply the helium-3 and to determine the thermomolecular pressure drop in a tube possessing a temperature gradient. This pressure drop is being determined as a function of temperature and tube diameter, and applied as a correction to vapor pressure measurements in low temperature thermometry.

Weber and Schmidt¹ fitted their data to the following equation for the thermomolecular pressure ratios of helium-4:

$$\log \frac{P_c}{P_w} = \frac{1}{2} \log \frac{T_c}{T_w} + 0.18131 \log \frac{y_c + 0.1878}{y_w + 0.1878} + 0.41284 \log \frac{y_c + 1.8311}{y_w + 1.8311} - 0.15823 \log \frac{y_c + 4.9930}{y_w + 4.9930}$$

where

$$y = \left(\frac{273.15^\circ K}{T} \right)^{1.117} \left(\frac{R_p}{13.42} \right)$$

The subscripts *c* and *w* designate cold and warm quantities, respectively; *R* denotes the tube radius in cm; *p* denotes pressure in microns of mercury; and *T* denotes temperature in °K.

Roberts and Sydoriak² found that for gaseous helium-3 "the thermomolecular ratios between room temperature and liquid helium temperatures measured experimentally do not differ within experimental error from values calculated from the Weber-Schmidt equations, Equation 1, for *R_{pw}* values down to 5 cm".

The Weber-Schmidt equation was programmed for the IBM-1620 and the calculated results for helium-3 and helium-4 were reported previously.^{3,4}

After these data were reported, an error was found in the program for calculating the thermomolecular pressure gradients which accounted for the increasing thermomolecular pressure gradient at increasing temperatures. The program was corrected and the calculated values of the thermomolecular pressure gradient for helium-3 and helium-4 are shown in Figures 1 and 2, respectively. The gradient exhibits a characteristic maximum and then appears to approach zero asymptotically. Figure 3 shows the calculated pressure difference across the warm end of the 1/4-inch and 1/32-inch tubes as a function of cold temperature for helium-3 and helium-4 saturated vapor.

¹S. Weber and G. Schmidt "Determination of the Thermomolecular Pressure Difference near Limiting Condition $P_1/P_2 = T_1/T_2$ and the Comparison with Theory" Leiden Comm., 246c (1936).

²T. R. Roberts and S. G. Sydoriak "Thermomolecular Pressure Ratios for He³ and He⁴" Phys. Rev., 102:2, 304 (1956).

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⁴MLM-1179

Figure 1. *He³ Thermomolecular Pressure Gradient.* **Figure 2.** *He⁴ Thermomolecular Pressure Gradient.*

Figure 3. *Ambient Pressure Difference Between $\frac{1}{4}$ and $\frac{1}{32}$ -Inch Tubes as a Function of Cold End Temperature.*

The variables in the Weber-Schmidt equation are being evaluated to better understand this equation, and to determine if there is sufficiently accurate and precise control and measurement of these variables in the experimental apparatus. The first variable that was selected for evaluation was the warm temperature. Figures 4 and 5 show the dependence of the pressure differential as measured across the $\frac{1}{4}$ -inch and $\frac{1}{32}$ -inch tubes on the ambient temperature. Figure 5 shows that around 300°K, a change of 1°K in the ambient temperature will produce a maximum pressure deviation of about 0.7 microns of mercury. Since the warm temperature will be controlled to $\pm 1^\circ\text{K}$ or better, variations in ambient temperature will cause little or no problem in the thermomolecular pressure measurements.

The data in Figure 4 indicate, however, that as little as 0.1°K error in the measurement of the cold temperature, especially around the maximum of Figures 1 and 2, can cause as much as 50 microns of mercury difference in the thermomolecular pressure gradient. This indicates that particular care must be taken in regulating and measuring the cold temperature.

The following revisions were made on the experimental thermometry system and greatly improved the performance of the system.

1. Aluminum foil was wrapped around the inner dewar to reduce the radiant heat transfer into this dewar. This procedure prevents visual determination of the liquid level in the inner bath, but this level can be calculated by making a material balance on the helium storage tanks. The advantage obtained from this modification can be easily appreciated in view of the Stefan-Boltzman relation for radiant heat transfer in which the temperature appears in the fourth power.
2. Two brass plates were soldered to the thermometer tube jacket, approximately two feet and three feet, respectively, above the flanges of the inner dewar system. The flanges are radiation shields between the inner dewar and the room temperature header assembly. The design and location of these plates will be changed since they cause difficulty in cooling the system to 4.2°K.
3. The nominal $\frac{1}{4}$ -inch and $\frac{1}{32}$ -inch vapor pressure sensing tubes were thermally grounded to the outer liquid helium-4 bath, which reduced the heat leak from the ambient temperature header assembly to the inner bath by approximately 70%. Three holes were drilled in the $\frac{1}{2}$ -inch vacuum jacket surrounding the thermometer tubes and the $\frac{1}{2}$ -inch jacket was plugged with solder through the center hole. The two outer holes were then connected with a piece of $\frac{1}{4}$ -inch copper tubing to evacuate the $\frac{1}{2}$ -inch jacket. This modification calls attention to the unknown effect of the temperature gradient and tube length on the thermomolecular pressure.

After the changes on the experimental system were completed, a five-day checkout run was made using helium-4 in the inner bath and in the thermometer bulb. Midway through the run, a thermostated bath was added to the warm end of the vapor pressure sensing tubes. Above approximately 1.6°K the system could not be properly stabilized. From this temperature down to the minimum temperature ($\sim 1.3^\circ\text{K}$) steady thermomolecular pressures were measured, showing a qualitative similarity to the predictions of the Weber-Schmidt equation as presented in Figure 6. A quantitative disagreement exists, however, which cannot be explained by the scatter of the observations.

Possible reasons for this disagreement as well as for the scatter are being evaluated with the help of computer calculations on the Weber-Schmidt equation. It is believed that a more accurate determination of the inside radius of the small tube must be made as well as a more accurate determination of the absolute temperature and better control of this temperature.

A second run of two days duration was made using helium-3 in the bulb and helium-4 in the inner bath. During the first part of this run, an unsuccessful attempt was made to properly stabilize the system using saturated helium-3 vapor in the same manner as had been done in the previous run with helium-4. The last part of this run was made using unsaturated helium-3 vapor; excellent system stability was obtained. During this portion of the run data could be taken quite rapidly. The agreement with the Weber-Schmidt equation was good at the higher pressures, but an unexplained deviation was observed at the lower pressures, as shown in Figure 6.

Figure 4. Difference Between Thermomolecular Pressure Gradients in a Nominal $\frac{1}{4}$ and $\frac{1}{32}$ Inch OD Tube as a Function of Ambient Temperature.

It is believed that the system could not be stabilized using liquid helium-3 in the bulb because the helium-3 vapor pressure dependence on temperature is about an order of magnitude greater than for helium-4. Thus, more accurate temperature control is necessary when using liquid helium-3 in the bulb than when using liquid helium-4.

Because of this difference in temperature dependence, a run will be made using helium-3 in both the thermometer bulb and inner bath to increase the sensitivity of the bath pressure control device. In any case better pressure controllers than are now being used will be necessary to significantly expand the present narrow temperature range in which stable thermomolecular pressure differences can be measured.

As a result of the last run the experimental difficulties were greatly reduced when using unsaturated helium vapors because the pressure of unsaturated helium vapor is less temperature dependent than saturated vapor.

Figure 5. Difference Between the Thermomolecular Gradients in a Nominal $\frac{1}{4}$ and $\frac{1}{32}$ -Inch OD Tube Per $^{\circ}\text{K}$ Ambient Temperature as a Function of Cold Temperature.

Figure 6. Observations Compared to the Weber-Schmidt Equation ($\frac{1}{4}$ and $\frac{1}{32}$ -Inch Tubes).

CARBON ISOTOPE SEPARATION

The objectives of the carbon-13 program are the preparation of gram quantities of carbon-13 with a purity of 90%, the evaluation of carbon monoxide and methane as feed materials in the thermal diffusion separation system, and the evaluation of various other separation methods, such as chemical exchange and distillation.

Thermal Diffusion A total of 1.4 grams of methane containing over 90% carbon-13 was separated from natural methane in an 11-column thermal diffusion cascade. A batch analysis of approximately half of the product showed a mass-17 enrichment of 96.4% in methane of 94.4% purity. The carbon-13 enrichment of the product was determined to be 93.2% from an analysis of carbon dioxide samples prepared from the methane.

The production rate of the cascade was held at 500 ml per week (see Figure 7). This is equivalent to 0.25 gram of carbon-13 per week. A composition profile was measured on February 28, 1964. The results of the profile measurement (Table 2) show that the cascade inventory of enriched material was partially depleted since the last profile determination (February 5, 1964). Therefore, the production rate of 0.25 gram of carbon-13 per week probably cannot be sustained over a long period of time, and the production capability of the 11-column cascade is probably 0.15 to 0.20 gram of carbon-13 per week. The production rate estimated from cascade calculations was 0.31 gram per week.

On February 29 the 11-column cascade was converted to the separation of partially enriched methane which had been purchased. The carbon-13 content of the first feed of partially enriched methane was 31%. This material is being feed at the rate of 1.6 liters per day to the middle of the fifth stage of the cascade. The product, withdrawn from the top of stage 9 at 300 to 400 ml per day, contains approximately 95% carbon-13. At

Figure 7. Performance of the 11-Column Methane Thermal Diffusion Cascade.

Table 2

COMPOSITION PROFILES FOR THE 11-COLUMN METHANE
THERMAL DIFFUSION CASCADE

Sample Location	Mass 17 Enrichment (%)		
	Dec. 27, 1963	Feb. 5, 1964	Feb. 28, 1964
Feed	1.09	1.09	—
First stage bottom	1.31	1.38	1.32
Second stage bottom	2.45	2.85	2.56
Third stage bottom	3.82	4.26	3.53
Fourth stage bottom	6.93	7.25	5.79
Fifth stage bottom	—	19.10	14.20
Sixth stage bottom	48.90	50.46	40.52
Seventh stage bottom	83.31	93.04	77.17
Eighth stage bottom (product withdrawal)	97.24	95.98	95.94
Ninth stage middle	97.95	97.06	96.87

the present time more carbon-13 is being fed to the cascade than is being recovered from it. In this particular cascade the system inventory of enriched material probably is higher for the separation of partially enriched methane than for the separation of natural methane; hence, additional carbon-13 must be added to the system before steady state conditions can be attained.

Conversion of Methane to Carbon A method is being developed for quantitatively converting methane to elemental carbon, which is one form that carbon-13 users have requested. The technique involved pyrolysis of methane over a platinum filament. Hydrogen was removed from the system by selective oxidation over cupric oxide. Recovery of carbon has been poor. Typically, 50% or less of the carbon put into the system as natural methane has been recovered as elemental carbon. Considerably higher yields must be achieved before attempts are made to convert enriched methane.

NEON ISOTOPE SEPARATION

The one-one overlap thermal diffusion system (see Figure 8) enriched neon-20 to about 99.8% at a rate of 70 ml per hour; neon-22 to about 99.8% at a rate of 4 ml per hour; and separated an intermediate stream of approximately 66% neon-20, 3% neon-21 and 31% neon-22 at a rate of 4 ml per hour. A sample of the feed to the second stage showed that it contained about 27% neon-20, 1% neon-21, and 72% neon-22.

The two column thermal diffusion cascade with the reservoir at the bottom separated about 30 liters of 99.9% neon-20 from the intermediate neon-21 material. After most of the neon-20 has been removed at a high concentration, high purity neon-22 will be removed, leaving enriched neon-21.

Figure 8. One-One Overlap System.

ARGON SEPARATION

The performance of the hot calcium purification system, given in Table 3, showed that at 650°C the impurities were reduced considerably but argon-40 evolved from the calcium metal in the trap, first in large quantity from the surface layers in the calcium, then in decreasing amounts from within the metal.

A new trap filled with calcium metal was carefully prepared by cleaning the metal with trichloroethylene to remove surface oils. After the calcium was placed in the trap, the metal was washed again, and the trap was immediately closed and evacuated. After being evacuated for two days, the trap was heated from 25°C to 650°C in 50°C intervals *in vacuo* to remove argon evolving from the surface layers of the metal.

Table 3

ARGON CLEANUP USING A HOT CALCIUM TRAP IN PRODUCT STREAM

Time (days)	Flow (ml/hr)	Temp (°C)	Composition (%)													
			Argon		Ar-36		Ar-36		Ar-40		H ₂		N ₂		O ₂	
			In	Out	In	Out	In	Out	In	Out	In	Out	In	Out	In	Out
8	0.5	650	65.56	99.66	99.96	83.24	0.03	0.03	0.01	16.73	20.30	0.16	14.14	0.18	0.00	0.00
11	1.7	650	87.16	99.98	99.99	99.06	0.01	0.02	0.00	0.92	7.09	0.00	5.74	0.02	0.00	0.00
13	2.0	650	88.23	99.94	99.99	99.48	0.00	0.02	0.01	0.51	6.61	0.00	5.11	0.05	0.05	0.01
15	2.5	650	90.12	99.91	99.80	99.56	0.01	0.02	0.18	0.42	5.74	0.00	4.12	0.09	0.01	0.00
19	3.0	650	90.83	99.92	99.99	99.40	0.01	0.01	0.00	0.59	5.41	0.00	3.75	0.06	0.01	0.02

This new trap was installed in the top of the final stage with the piping arrangement shown in Figure 9. It consists of a circulating loop through the trap by convection then passing the stream across the top of the final stage. This large circulating stream should mix the argon-40 evolving from the calcium into the convection stream in the column with a continuous purifying action, and maintain the product gas which is drawn off upstream from the trap free from impurities and argon-40.

During the off stream time while the calcium trap and associated lines were being installed at the top of the last stage, the argon-36 profile increased at the product end of the four stage thermal diffusion system, forcing the argon-38 peak concentration back through the system into Stage 3. Hence, no argon-38 draw-off was made at the bottom of the third stage.

When this trap is placed in the operation satisfactorily, the argon-36 product flow will be regulated to give a steady state system profile such that 99.8% concentration of argon-38 may be removed from the bottom of Stage 3.

Figure 9. Recirculating Hot Calcium Purification System for Argon-36.

KRYPTON ISOTOPE SEPARATION

Experimental Chromatography Chromatography is being used as one method to separate krypton from xenon. The experimental chromatograph is a tubular column 9.5 cm long and 2.54 cm in diameter, packed with 50/60 mesh activated coconut charcoal.

Four runs were made using a column temperature of 193°K. After the krypton-85 peak passed its maximum, the column was heated to 243°K. The data, as shown in Table 4, reveal that nitrogen was significantly better as a carrier gas than helium.

Table 4

EFFECT OF CARRIER GAS ON THE CHROMATOGRAPHIC SEPARATION OF KRYPTON-85 FROM XENON

Run	Carrier Gas	Cleanup, Feed Conc.	Krypton Retention (min)	Time Till Xenon Peak (min)	Recovery in Xenon Trap (%)
$\sigma 1$	He	1120	33.5	41	70
$\sigma 2$	He	a	36	43	74
$\sigma 3$	N ₂	a	3	b	28 ^c
$\sigma 4$	N ₂	a	3	b	79 ^d
τ average (for comparable runs)			34.6 $\pm$ 0.2	48.8 $\pm$ 2.5	71 $\pm$ 0.3

^a Sample analysis not available at this writing.

^b At the predetermined conditions that had to be used in these runs, e.g., a 3.4 liter/min. flow rate, 25 ma was the highest current that could be used in the thermal conductivity cell detector as compared to 165 ma with helium as the carrier gas. The detector is not sensitive enough to detect xenon under these experimental conditions.

^c The charcoal krypton trap had a large nitrogen holdup. The recovery is approximate as some xenon was lost when this nitrogen was pumped off.

^d Since the xenon peak is not observable, the xenon trap was placed in the line when the krypton peak approached the normal base line.

Run $\sigma 1$ and $\sigma 2$ were control runs made under the same conditions as $\tau 2$, 3, 4, and 7, reported previously.⁵ Runs $\sigma 1$ and $\sigma 2$ compare favorable with the τ series. Judging from the cleanup of $\sigma 1$ and recovery of $\sigma 2$, the reduced dead space in the modified chromatograph is reflected by a slightly higher efficiency.

Nitrogen was used for the first time as the carrier gas in Runs $\sigma 3$ and $\sigma 4$. The chromatograms of these runs indicated good krypton-85 removal. The xenon peak was not detectable under the conditions of these runs. The samples for clean-up data are being analyzed.

Since the xenon peak was not detected, the traps in Run $\sigma 3$ were not changed until it became evident that the peak could not be observed. In Run $\sigma 4$ the traps were changed when the krypton peak asymptotically

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approached the base line. Run $\sigma 4$ had a recovery of 79% in the xenon trap (product) compared to about 71% with helium. If the cleanup is comparable to or better than Runs $\sigma 1$ and $\sigma 2$, nitrogen would be superior to helium as a carrier gas. At the same flow rate, the time for a separation using nitrogen is $1/2$ th of that using helium. The charcoal traps held a large amount of nitrogen and interfered with the xenon recovery. The charcoal krypton trap was replaced by a cold trap filled with copper turnings, which worked well.

THERMAL DIFFUSION RESEARCH

Thermal Diffusion Column Theory A rigorous theory for the thermal diffusion column is being developed at Mound Laboratory. One feature of this theory is that the dependence of the transport properties, namely, viscosity, thermal conductivity and density, on the composition of the isotopic mixture is being considered. This composition dependence was not considered in the earlier theory of Jones and Furry. Another feature of the theory is that it allows for a variation in the wire temperature arising from the change in thermal conductivity of the gas.

An equation was derived⁶ for the transport of species¹ up the column. Using the results obtained from this equation⁷, this transport equation can be written in the following form:

$$t_1 = Hr(1-sc_1)c_1c_2 - K_c r(1-5sc_1)\frac{dc_1}{dz} - K_d r(1-2sc_1)\frac{dc_1}{dz} + K_b r(1+sc_1)\frac{d^2c_1}{dz^2} - K_b r s \frac{d}{dz^2} c_1 \frac{dc_1}{dz}$$

where

$$r = m_1/m_2, \quad S = (m_2 - m_1)/2m_2, \text{ and}$$

$$H = 2\pi \int_{T_1}^{T_2(z)} \frac{\alpha G(T)}{T} dT$$

$$K_c = \frac{2\pi}{Q} \int_{T_1}^{T_2(z)} \frac{[G(T)]^2}{\rho D} \lambda dt$$

$$K_d = \frac{2\pi}{Q} \int_{T_1}^{T_2(z)} \rho D r^2 \lambda dT$$

$$K_b = \frac{2\pi}{Q} \int_{T_1}^{T_2(z)} \frac{G(T)\lambda}{\rho D} \int_{T_2(z)}^T \rho D r^2 \lambda dT^2$$

The function $G(T)$ is the solution of the differential equation.

$$Q^3 \frac{d}{dT} \frac{1}{\lambda r^2} \frac{d}{dT} \frac{\eta}{\lambda} \frac{d}{dT} \frac{1}{\lambda r^2 \rho} \frac{dG(T)}{dT} = g \frac{d\rho}{dT}$$

with the boundary conditions

$$G(T_1) = G(T_2) = G'(T_2) = 0$$

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In these equations ρ , λ , and η are the density, thermal conductivity, and viscosity for a pure gas of species 2; O is the coefficient of ordinary diffusion.

The transport properties of a gas are given in terms of the collision integrals, $\Omega^{(i,j)*}$, which are tabulated functions of temperature. In terms of the quantities $\theta = T/T_1$ and $\Omega^{(i,j)} = \Omega^{(i,j)*}(T)/\Omega^{(i,j)*}(T_1)$,

$$\begin{aligned}\rho &= \rho_1/\theta \\ D &= D_1\theta^{3/2}/\Omega^{(1,1)} \\ \eta &= \eta_1\theta^{1/2}/\Omega^{(2,2)} \\ \lambda &= \lambda_1\theta^{1/2}/\Omega^{(2,2)}\end{aligned}$$

where the subscript 1 indicates that the quantity is to be evaluated at the cold wall temperature. Using these expressions for the transport properties, the differential equation can be written as

$$\frac{d}{d\theta} \frac{\Omega^{(2,2)} \gamma^2}{\theta^{1/2}} \frac{d^2}{d\theta^2} \Omega^{(2,2)} \gamma^2 \theta^{1/2} \frac{d\gamma(\theta)}{d\theta} = \frac{d}{d\theta} \frac{l}{\theta}$$

where $\gamma = r_1/r$ and

$$\gamma(\theta) = \frac{Q^3 \eta_1}{T_1^3 r_1^4 \lambda_1^3 \rho_1^2 g} G(\theta)$$

the annular heat flow Q , and $\gamma(\theta)$ are given by the equations

$$Q = T_1 \lambda_1 f(\theta_0) / \ln \gamma_0$$

$$\gamma = e^{[\ln \gamma_0 f(\theta) / f(\theta_0)]}$$

where $\theta_0 = (T_2/T_1)$, $\gamma_0 = (r_1/r_2)$, and

$$f(\theta) = \int_1^\theta \frac{\theta^{1/2}}{\Omega^{(2,2)}} d\theta$$

The differential equation for $\gamma(\theta)$ can be integrated four times to give

$$\gamma(\theta) = \int_1^\theta \frac{1}{\mu} \int_1^\theta \int_1^\theta \frac{d\theta^3}{\mu} + C_1 \int_1^\theta \frac{1}{\mu} \int_1^\theta \int_1^\theta \frac{\theta d\theta}{\mu} + C_2 \int_1^\theta \frac{(\theta-1)}{\mu} d\theta$$

where $\mu = \gamma^2 \theta^{1/2} \Omega^{(2,2)}$

The constants c_1 and c_2 are determined by the boundary conditions $\gamma(\theta_0) = \gamma'(\theta_0) = 0$; the other two constants which arise in the integration are determined by the boundary conditions $\gamma(1) = \gamma'(1) = 0$, and they are equal to zero. A computer program was written to numerically calculate the integrals for $\gamma(\theta)$ by Simpson's rule.

Using the expressions given above, the integrals K_c and K_d can be written:

$$K_c = 2\pi \frac{\rho_1^3 g^2 r_1^8}{D_1 \eta_1^2} \left[\frac{\ln \gamma_0}{f(\theta_0)} \right]^7 \int_1^\theta \gamma^2 \frac{\Omega^{(1,1)}}{\Omega^{(2,2)}} d\theta$$

$$K_d = 2\pi \rho_1 D_1 r_1^2 \left[\frac{\ln \gamma_0}{f(\theta_0)} \right] \int_1^\theta \frac{\theta d\theta}{\gamma^2 \Omega^{(1,1)} \Omega^{(1,1)}}$$

A Simpson's rule integration to calculate these integrals has also been programmed.

The computer program is currently being checked and tests are being run to determine the number of intervals in Simpson's rule which are necessary to obtain accurate values of the integrals K_c and K_d .

Low Energy Scattering To improve the accuracy of calculated transport properties, the intermolecular potentials must be known more accurately. Evaluation of possible methods for performing this type of measurement on noble gas molecules in the low energy range has shown that a suitable collector for these atoms is unavailable.

An electron multiplier was tested as a neutral particle detector. During this test, the output of the multiplier was connected to an electrometer. To alleviate instability, all tests were run with a 10^8 -ohm input resistor. To test the electron multiplier installation, a crude electron gun was fabricated. When it was directed at the multiplier input, the electrometer could be driven off the 30-volt scale. When the pressure in the bell jar was 10^{-6} torr or less, the electrometer rose sharply to several millivolts when a large burst of helium was admitted through the gas inlet tube. However, this neutral particle sensitivity is much lower than the minimum required for a scattering detector.

Further experiments are planned which shall incorporate an ionizing electron beam directed across the incoming neutral particle beam immediately in front of the electron multiplier inlet.

Center Wire Studies A 24-foot thermal diffusion column with a mercury well used direct current for heating the center wire. The average separation factor was approximately 40,000, neglecting Sample 3, which was removed two hours after the previous sample was taken. The column may not have had time to produce enough enriched gas to allow the column to reach equilibrium.

A spring tension device was attached to the bottom of the wire in a larger well, increasing the volume by approximately 180 cc between the mercury well and the well for the spring tension device. The data are shown in Table 5.

A preliminary run using alternating current instead of direct current in a thermal diffusion column did not cause the wire to vibrate visibly. If the isotope separation of the neon is the same as that obtained when using direct current, alternating current could be used for heating the center wire of a thermal diffusion column when the column is equipped with a damping device.

Magnetic Pump for a Cascade Loop A magnetic pump shown in Figure 10 was designed for circulating gas through an interstage circulating loop and as an intermediate swing pump which may be reversed periodically. A pump of this type has several advantages: (1) there are no external moving parts which could eventually open a leak to the system; (2) there is little holdup, which is essential in a swing system; (3) the pumping rate may be accurately varied; and (4) there is no possible contamination of the process gas by oil or other lubricant.

Table 5

NEON ISOTOPE SEPARATION USING DIRECT CURRENT

Time of Sample (hrs)	0		28		30		35		46	
Sample	1-B	1-T	2-B	2-T	3-B	3-T	4-B	4-T	5-B	5-T
Mercury Well Concentration (mole %)										
Total Ne	99.89	99.88					97.73	90.36	97.44	90.66
Ne-20	50.05	50.02	0.77	99.62	0.82	99.63	0.78	99.63	0.79	99.66
Ne-21	0.20	0.16	0.08	0.07	0.00	0.00	0.04	0.04	0.04	0.04
Ne-22	49.75	49.81	99.16	0.31	99.18	0.37	99.18	0.33	99.18	0.30
Separation Factor	-		41391		32571		38390		41707	
Spring Tension										
Total Ne									99.66	85.91
Ne-20			5.21	99.83	5.18	99.84	5.10	99.85	5.02	99.82
Ne-21			0.16	0.00	0.16	0.02	0.17	0.02	0.16	0.03
Ne-22			94.64	0.17	94.67	0.14	94.73	0.13	94.82	0.14
"Q" Values	-		10667		13035		14267		13468	

Reversible Operation:

To pump in "Y" Direction:
 "A" Open on "R" cycle, "B" closed
 "B" Open on "L" cycle, "A" closed

To pump in "X" Direction:
 "A" Open on "L" cycle "B" closed
 "B" Open on "R" cycle "A" closed

Figure 10. Reversible Magnetic Circulating Pump.

A prototype model was constructed using two modified Skinner 10-watt solenoid valves. Operation of the pump has been good until it becomes hot. When hot, the pumping action slows and becomes irregular. Further piston modifications may eliminate this difficulty.

Center Wire Embrittlement A study is being made to determine why center wires in thermal diffusion columns burn out so rapidly. It is suspected that the crystal lattice in the metal fractures from the formation of water vapor within the lattice structure. Oxygen is contained within the metal lattice along the grain boundaries, and the metal has a certain solubility of hydrogen which is a function of the oxygen content, temperature and pressure. Upon entering the crystal lattice of the metal, the hydrogen atoms combine with the contained oxygen to form water. Since the hydrogen diffuses through the crystal lattice more rapidly than the formed water vapor can escape, the pressure increases and ruptures the lattice structure. This rupture occurs at the grain boundaries because of: (1) the inherent weaknesses due to lesser lattice binding forces; (2) the more abundant oxide formation there; and (3) the hydrogen enters the lattice more readily at these points to combine with the oxygen⁸. The internal lattice pressure causes expansion to occur as shown by lesser hydride density.

Rhines and Anderson⁹ conclude that hydrogen is the only gas that will produce this type of fractural condition for embrittlement. N. P. Allen¹⁰ has shown that trapped gases exert enormous internal pressures in metals, which cause numerous, minute, "blow holes" to congregate at the crystal grain boundaries in the lattice. Allen and A. C. Street¹¹ observed that this type of porosity occurs when the oxygen content exceeds a certain critical value which is a function of the solid solubility of the oxygen in the metal.

This mechanism may contribute to embrittlement in Nichrome V wires used in thermal diffusion columns. When the wire is heated, hydrogen combines with oxygen in the wire to form water vapor, causing fracturing at the grain boundaries. The embrittlement later causes breakage after repeated shutdowns.

A burned out wire was examined at 60X magnification. From its appearance at the breaking point, the wire evidently had melted from within. The outer surface was covered with cracks and fractures in a pattern which resembled possible grain boundary lines.

Oxidation of the wire surface was observed in well defined portions of the wire. The break occurred in a bright portion of the surface near the boundary of an oxide layer.

⁸ F. N. Rhines, Trans. A.I.M.E., 156, 33 (1944).

⁹ F. N. Rhines and W. A. Anderson, Trans. A.I.M.E., 143, 312 (1941).

¹⁰ N. P. Allen, J. Inst. Metals, 49, 317 (1932).

N. P. Allen, J. Inst. Metals, 43, 81 (1930).

¹¹ N. P. Allen and A. C. Street, J. Inst. Metals, 51, 233 (1933).

PLASTICS RESEARCH

Mound Laboratory is conducting research on the molding properties of diallyl phthalate in an effort to obtain superior plastic parts for the detonator program.

ASBESTOS-FILLED DIALLYL PHTHALATE

A silane coupling agent was added to the standard asbestos-filled diallyl phthalate plastic formulation to determine its effect on the physical properties. The formulation contained:

- 100 parts resin
- 100 parts acetone
- 98 parts Johns Manville asbestos floats
- 2.0 parts calcium stearate
- 2.0 parts titanium dioxide
- 2.0 parts tert-butyl perbenzoate
- 2.5 parts pigment (Harshaw 25AD Scarlet)
- 0.03 parts hydroquinone
- 2.0 parts silane coupling agent

Four silane coupling agents were used: A151, A172, A4086 and A4087 (Union Carbide Corporation). The silanes were added to the formulation just prior to the milling operation.

The A151 coupling agent increased the heat distortion temperature to 214°C as compared to 165°C for the standard DAP formulations. No other improvements in the physical properties were noted.

FOAMS EVALUATION

Properties of epoxy foam systems prepared previously¹² which contained epoxy resin (Epon 828), 4,4-diaminodiphenylsulfone (DDS), and trimethoxyboroxine (TMB), (Formulation EF-1) were unsatisfactory. A study of the reaction of DDS with TMB, component ratios, and a new preparative technique were initiated to improve the properties of EF-1 foams.

Earlier workers postulated that the thermal stability of foams prepared with TMB and DDS was due to an initial polymerization of TMB and DDS. The highly cross-linked DDS-TMB polymer then cured the epoxy resin. However, various attempts to obtain a polymeric substance from a TMB-DDS mixture were unsuccessful. No reaction occurs when TMB and DDS are mixed together at room temperature. When the two materials are heated to approximately 150°C, a clear amber liquid forms with the evolution of a gas. A solid material results when the liquid is subsequently cooled.

The material was identified by placing a mixture of 24 grams of TMB and 16 grams of DDS into a flask fitted with a thermometer and a water-cooled condenser. The flask was heated in an oil bath at 150°C and the liquid which distilled off was collected. The vapor temperature during distillation ranged between 50 and 70°C. The contents of the flask were cooled and dissolved in ethanol. Recrystallization from ethanol gave a brown material melting at 173°C, which proved to be DDS. Apparently, the TMB and DDS did not react. The liquid which distills is probably trimethyl borate formed by the decomposition of the TMB.

¹²MLM-1185

The gel times and exotherms generated during gelation with various ratios of TMB and DDS were determined to optimize the reactant ratio. Gel times were determined using a Tecam gel meter. Exotherms were determined using an iron-constantan thermocouple connected to a recorder. The results are shown in Table 6. Heat resistance of the samples was determined by placing weighed samples in a circulating air oven at 230°C for one hour and determining the weight loss.

Table 6

**GEL TIME, EXOTHERM, AND HEAT RESISTANCE OF
VARIOUS EPOXY FOAM FORMULATIONS**

Sample ^a	Formulation (parts)		Gel Time (min)	Exotherm (F)	Heat Resistance (% wt loss)
	DDS	TMB			
1-1	10	10	62	85	0.76
2-1	20	10	34.5	103	1.19
3-1	30	10	18.0	125	1.98
4-1	40	10	15.0	119	2.53
5-1	50	10	10.0	125	2.70
1-2	10	20	17.5	151	1.99
2-2	20	20	12.5	185	2.54
3-2	30	20	8.5	152	3.14
4-2	40	20	6.5	148	3.05
5-2	50	20	5.5	178	3.22
1-3	10	30	11.5	188	5.49
2-3	20	30	10.5	208	5.60
3-3	30	30	8.5	219	6.82
4-3	40	30	5.5	160	7.52
5-3	50	30	4.5	168	6.75
1-4	10	40	9.5	211	10.74
2-4	20	40	7.5	211	9.72
3-4	30	40	5.5	188	9.44
4-4	40	40	4.5	187	8.10
5-4	50	40	3.5	165	8.55
1-5	10	50	8.5	198	12.60
2-5	20	50	6.5	189	11.26
3-5	30	50	4.5	168	11.0
5-5	50	50	3.5	158	9.83
1-6	10	60	8.5	195	15.09
2-6	20	60	6.5	187	12.65
3-6	30	60	5.5	188	11.48
4-6	40	60	4.5	177	12.19
5-6	50	60	3.5	174	10.92
0-2	0	20	23.0	- - -	1.37
0-3	0	30	15.0	185	2.56
0-4	0	40	12.5	185	6.37
0-5	0	50	12.0	228	10.40
0-6	0	60	10.5	185	13.30

^aSample Designation: First Number x 10 ~ PHR DDS; second number x 10 ~ PHR RMB. All samples were prepared with 20 grams Epon 828.

The fastest gel times were obtained with 40-60 parts TMB and 50 parts DDS. However, since the foams obtained in this study were not rigid at 230°C, the optimum reactant ratio could not be determined. The weight loss increased as the DDS and/or TMB concentration increased. This indicated that a reaction of components at these concentrations is incomplete. After exposure to the test temperature the samples contained small glossy particles dispersed throughout the material.

It was reported (Technical bulletin SP-24B, Shell Chemical Co.) that DDS dissolves in heated epoxy resin. Twenty grams of Epon 828 were heated to 100°C with known amounts of DDS. The solution was stirred until it was clear; the hot solution was then transferred to a small paper cup and allowed to cool for 25 minutes. TMB was then added to the mixture. The solution of DDS in Epon 828 was quite viscous at room temperature; however the viscosity was lowered when TMB was added. Samples prepared using this technique were superior to those prepared previously. They had excellent foam rise, cell structure, and heat resistance.

Formulation EF-1-32 had the best thermal stability. Variations from this formulation result in higher viscosity, greater weight loss and poorer cell structure. This formulation does not need an auxiliary blowing or a nucleating agent.

The effect of silicone surfactants was determined by using several silicone cell control additives in formulation EF-1-32 samples. The surfactant was used in one percent quantities (based on total weight of foam components); the silicone fluid was added after the DDS was solvated. The incorporation of the surfactants increased the foam rise to such an extent that the samples collapsed upon cooling. Formulations and results of heat resistance tests are shown in Table 7.

Table 7

FORMULATIONS AND HEAT RESISTANCE OF EPOXY FOAMS

Sample ^a	Formulation (Parts)			Silicone ^b Surfactant	Heat Resistance ^c		
	828	DDS	TMB		270°C	300°C	330°C
EF1-23	100	20	30	-----	9.33	16.40	20.02
EF1-22	100	20	20	-----	7.56	14.84	19.44
EF1-33	100	30	30	-----	9.16	14.40	18.43
EF1-32	100	30	20	-----	6.33	12.06	17.61
EFL-32A	100	30	20	1% DC-113	6.30	11.63	18.93
EF1-32B	100	30	20	1% XF-1066	6.20	13.00	18.62
EF1-32C	100	30	20	1% DC-201	6.99	12.78	19.42

^aSample designation: Last two numbers are indicative of parts of DDS and TMB respectively.

^bBased on total weight of 828, DDS and TMB.

^c% wt. loss.

Epoxy foams prepared using a solution of DDS in Epon 828 improved the thermal stability. All samples retained their rigidity at 330°C, but the samples were extremely brittle. Preliminary density determinations gave values in the 5-7 lb/ft³ range. The use of one percent silicone surfactant had little effect on heat resistance, but caused the foam to collapse.

ANALYTICAL

Methods of analyzing elements and compounds are being developed to support other programs at Mound Laboratory. These methods include instrumental techniques and classical wet methods.

CALORIMETRY

Automatic Sample Loading of a Calorimeter Automatic sample loading devices are being used to increase the output of calorimetric data and to improve the precision and accuracy of the data. The devices are used primarily for half-life determinations with calorimeters 90, 91, and 58. A detailed description has not been given previously, although one has been used¹³ since October, 1962.

The loading devices made it possible to hermetically seal the calorimeter from the room. A Lucite cover was first used¹⁴ with calorimeter 90. For a single measurement this has little advantage, but for repeated measurements on a sample over a period of several weeks, the slow variations disappear and the power measurements approach a steady value in less than ten days. Over a period of a year, half-life measurements on a plutonium-238 sample with calorimeter 90 have a probable error per observation of only 0.001%, which is almost five times smaller than any previous work.

The mechanical arrangement of the automatic loading device is shown in Figure 11 lifting a sample can from Calorimeter 90. The device consists of a sample-holder suspended on a line which passes through an arm and pulleys to a motor shaft. Two limit switches are used to sense the position of the sample holder.

Two lifting mechanisms are used; one for the sample and one for the dummy. The dummy sample holder is used during the heater run to produce similar conditions to the sample run. This is to prevent errors that might arise such as differences in end losses, time constants, stray light, and weight.

The operational sequence is as follows:

1. The sample is lowered into the calorimeter.
2. At the end of the timing period, the program timer energizes the lifting motor on the sample side.
3. When the sample holder is completely removed, the "up" limit switch is tripped which stops the motor and energizes the dummy side time delay relay. After two seconds, the dummy side motor lowers the dummy. When it reaches the bottom, the "down" limit switch opens. The motor continues to run for approximately five seconds until the time delay relay opens. This additional running time permits the line to become completely slack.
4. At the end of the timing period the program timer energizes the lifting motor on the dummy side. The sample side goes through the same step as described in 3 above.
5. The cycle will be repeated until stopped manually.

Figure 11. Automatic Loading Device for Calorimeter 90.

¹³MLM-1144

¹⁴MLM-1152

The program timer, through a dry reed relay, also switches the heater current into the calorimeter during the heater run. During the sample run the relay switches the current into the heater leads and a dummy heater resistor. The dummy heater resistor keeps the load within the range of the constant current controller. Heater lead current during the sample run balances the lead losses during the heater run.

Figure 12 is a simplified schematic diagram of the timer circuit. The lifting parts are wired to four pin connectors for interchangeability and manual operation. L.S. 1 and L.S. 3 are the limit switches which stop the motors on the up cycle and permit the opposite side to lower. L.S. 2 and L.S. 4 are the limit switches which stop (through the time delay relays) the motors on the down cycle. T.D. R1 and R.D. R2 are amperite two-second N.O. thermal time delay relays. While these do not have the same delay on opening as closing, they were chosen so that they would not produce heat during the run. The opening time may be adjusted over a small range by inserting a resistor in series with the heating elements.

The program timer is a modified Eagle Signal "Multipulse" repeat cycle timer with a 12-hour base time. The timer was modified to operate two switches together to prevent an interaction with the heater relay.

The heater relay is an Allen Bradley dry-reed relay. The calorimeter heater circuit was omitted from Figure 12. The lifting motors are single phase reversible motors with 93-ounce-inch of torque at 9.3 revolutions per minute. Smaller motors have been used, but were unsatisfactory for larger samples.

It is probable that the automatic loading devices, as they have been described here, will be used for many years since they have a proved performance and it is not desirable to make major changes during a half-life measurement.

Figure 12. Simplified Schematic Diagram of Timer Circuit.

Slow Variations in Calorimeter Outputs The equilibrium stability of all calorimeters is much better than the reproducibility from one run to the next. An extensive investigation was initiated to discover the cause of these semi-random slow variations¹⁵ which have periods ranging from a week to a year. The primary cause of the variations in Calorimeter 51 was caused by variations in the humidity of the room atmosphere. The output was observed for 20 months while a sample container was raised and lowered automatically four times a day. The cause of the slow variations which occurred during this time was obscure until the past winter, when the humidity control was not operating. Since the air gaps of Calorimeter 51 were sealed with black wax at the time of construction in 1949, it was difficult to see how changes in humidity could cause the zero bridge potential to change unless it was an interior heating effect. It was proved that the resistance of the bridge changed, but not because of self-heating and a change in the sensitivity. The most logical explanation of the slow variations is as follows:

1. When the sample container is inserted, fresh air with a different humidity content is introduced into the calorimeter.
2. The sample container, which has been heated (or cooled) by the room, will cause a temperature transient in the thermal, and consequently, in the air gap.
3. A pressure transient will result which causes some of the fresh air to be pulled into the air gap through a leak that has developed.
4. The varnish on the bridge wire, which are exposed to the air gap, absorbs (or desorbs) water vapor from the fresh air and causes a swelling of the insulation which stresses the wire and changes its resistance. Fortunately, no varnish has been used on a bridge since 1952.

The secondary cause of the slow variations in Calorimeter 51 is caused by barometric pressure changes in conjunction with removing and inserting the sample container, and can be explained by the same sequence listed above.

Probably most of the slow variations that occur to a lesser extent in other calorimeters also are caused by variations in humidity, although the more recently constructed calorimeters are made of materials that do not change weight (by sorption-desorption processes) with temperature and have enameled-covered bridge wires. This insulation is the best available for protection against humidity. However, since the hermetically sealed loading devices (described earlier) tend to remove the slow variations, it suggests that some constituent of the air causes trouble.

NUCLEAR COUNTING

2.2 Mev. Gamma Ray from Neutron Capture by Hydrogen The capture of a thermal neutron by a hydrogen nucleus is accompanied by the emission of 2.2 Mev gamma photon. Because of an appreciable thermal neutron cross section of hydrogen (0.33 barn) this effect can be observed with a neutron source and a relatively simple counting assembly.

The experimental arrangement was as follows: A 2 x 2-inch NaI(Tl) scintillation crystal detector was mounted vertically inside an open-end cylindrical lead shield. A metal beaker containing approximately two liters of water was placed over the open end of the shield, and a plutonium-beryllium neutron source with an emission rate of approximately 10^6 neutrons per second was positioned next to the water container in such a way as to be shielded as well as possible from the sodium iodide detector. The detector was connected into a 400 channel pulse height analyser.

The source was gamma counted for 30 minutes live time with water in the container, then the source was counted in the subtract mode for 30 minutes live time with no water in the container. Background counts

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¹⁵MLM-1138.

were removed from the gamma energy spectrum in a similar manner. A detectable peak in the residual gamma energy spectrum was obtained by this procedure; this peak was compared to the gamma spectrum of cobalt-60, and was found to be 2.19 Mev. A section of the residual spectrum containing this peak is given in Figure 13.

Figure 13. Section of Residual Gamma Spectrum of Moderate Pu-Be Neutron Source showing 2.2 Mev. Gamma from Neutron Capture by Hydrogen Nuclei in Water.

The integrated intensity of this peak showed a net total of 4641 counts for the 30 minute period, or a net counting rate of approximately 2.6 counts per second.

The water container was moved around to various locations without disturbing the neutron source and the intensity of the 2.19 Mev peak varied accordingly. This shows that the origin of the gamma ray was in the water and not a room background effect. No peak was observed at this energy when the neutron source was removed.

Sodium-22 in Aged Actinium Fluoride An aged sample of actinium fluoride (AcF_3) which was prepared 11 years ago at Mound Laboratory to study the growth rate and intensity of neutron emission was analyzed to determine the sodium-22 content.¹⁶ Because of alpha-neutron reactions involving fluorine-19, sodium-22 (half-life = 2.58 years) could now be detected in the sample.

The sample had originally contained 44 millicuries of actinium and had achieved an equilibrium neutron emission rate of approximately 55,000 neutrons per second. The main mode of decay of sodium-22 is positron

¹⁶ K. W. Foster and J. G. Stites, J. Phys. Chem., **60**, 1017 (1956).

emission followed by a 1.28 Mev gamma from the daughter nucleus, neon-22. The quantities of sodium-22 to be expected in this sample after 11 years is of the order of 10^{-10} gram, so that the sample would be expected to show a sodium-22 decay rate of approximately 4×10^4 disintegrations per second.

The actinium-fluoride sample was counted by a 2 x 2-inch NaI(Tl) crystal detector connected to a 400-channel pulse height analyzer. The sample was contained in a lead during counting to reduce the intense radiation from the lower energy gammas in the actinium decay chain. A faint peak was tentatively identified at approximately 1.3 Mev. However, it was not possible to distinguish the 0.51 Mev positron annihilation peak because of the predominance of Compton edges and general broadening of the actinium spectrum by the lead container. Coincidence techniques will be used to obtain more reliable data on the sodium-22 content of the sample.