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General Electric Company, Richland	28-33
Hanford Operations Office	34
Iowa State College	35
Kellex Corporation	36-37
Knolls Atomic Power Laboratory	38-41
Los Alamos	42-44
Massachusetts Institute of Technology (Gaudin)	45
Massachusetts Institute of Technology (Kaufmann)	46
Mound Laboratory	47-49
National Advisory Committee for Aeronautics	50
National Bureau of Standards	51-52
Naval Radiological Defense Laboratory	53
NEPA Project	54
New Brunswick Laboratory	55
New York Operations Office	56-57
North American Aviation, Inc.	58
Oak Ridge National Laboratory	59-70
Patent Branch, Washington	71
Sylvania Electric Products, Inc.	72
Technical Information Branch, ORE	73-87
UCLA Medical Research Laboratory (Warren)	88
University of California Radiation Laboratory	89-92
University of Rochester	93-94
Western Reserve University (Friedell)	95
Westinghouse Electric Corporation	96-97
	97

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SUMMARY OF THE RESEARCH PROGRESS MEETING

April 28, 1949

H. P. Kramer

Thermochemical Properties of Pr and Am Oxides. L. Eyring.

The aqueous oxidation potential of the Am^{+3} - Am^{+4} couple was determined as a part of a program of research on the basic chemistry of the lanthanides and actinides. A knowledge of this potential is useful in the separation of Am from Cm and in the estimation of the oxidation states of the heavy elements of atomic number beyond 96. The only tetravalent compound of Am that is known at present is AmO_2. A knowledge of the oxidation potential will facilitate the prediction of the stability of other tetravalent compounds of americium.

Since the lanthanide rare earths bear a close chemical resemblance to the actinides, it is possible to make inferences about an element of the actinide series by analogy with the corresponding element of the rare earth series. The lanthanide metal that corresponds to Am is Pr. Because of the small quantities available and the high toxicity of Am, Pr was used as a stand-in for Am in the exploratory experiments. Of praseodymium, the following oxides are known: Pr_2O_3, Pr_6O_{11} (of vaguely defined composition that varies with the manner of preparation) and PrO_2.

The oxidation potential of the M^{+3} - M^{+4} couple is determined by means of the two equations:

$$\Delta F = \Delta H - T \cdot \Delta s$$

$$\Delta F = \frac{-E}{nf}$$

where F is the free energy, E is the oxidation potential, n the number of moles involved, f is Faraday's constant, and Δs is the change in entropy in the reaction. In order to calculate the change in heat content ΔH in the reaction $Am^{+3} \longrightarrow Am^{+4}$, the change in heat content was calculated for the equivalent reaction $Am^{+3} + H^{+} \rightarrow Am^{+4} + \frac{1}{2}H_2$. This reaction is equivalent since the oxidation potential of the couple $H^{+} \longrightarrow \frac{1}{2}H_2{}^{\circ}$ is set equal to zero on Latimer's scale of oxidation potentials. The calculation was carried out by the following scheme:

$$\begin{array}{lll}
MO_2 + 3H^{+} \longrightarrow M^{+3} + \frac{1}{4}O_2 + 3/2H_2O & & \Delta H_1 \\
MO_2 + 4H^{+} \longrightarrow M^{+4} + 2H_2O & & \Delta H_2 \\
\frac{1}{2}H_2 + \frac{1}{4}O_2 \longrightarrow \frac{1}{2}H_2O & & \Delta H_3 \\
\hline
Am^{+3} + H^{+} \longrightarrow Am^{+4} + \frac{1}{2}H_2 & & \Delta H
\end{array}$$

so that $\Delta H = \Delta H_2 - \Delta H_1 - \Delta H_3$.

ΔH_1 was measured directly in a micro-calorimeter. The change in temperature was measured by means of the change of resistance in a winding about the calorimeter flask of 50 feet of 40 mil wire. The apparatus is capable of detecting a change in temperature of 10^{-4}° C. ΔH_2 was estimated on the basis of the heats of formation of other actinide tetravalent oxides and tetravalent ions from the metals, and the heat of formation of water ΔH_3 is well known.

It was thought that a very close estimate of the change in entropy in the reaction would be given by the change in entropy of Pu in the analogous oxidation. The values that were arrived at for the oxidation potentials are -2.6 v. for the $Am^{+3} - Am^{+4}$ couple and -3.1 v. for the $Pr^{+2} - Pr^{+4}$ couple.

An attempt to correlate these results with the thermochemical data for Ce failed because at the acid concentrations that were used, Ce^{+4} exists in the hydrolized form $Ce(OH)^{+3}$ so that the experiments were not reproducible for Ce.

A detailed account of the work is contained in "Thermochemical Studies of Oxides of Praseodymium and Americium and the Estimation of the $Pr^{+3} - Pr^{+4}$, $Am^{+3} - Am^{+4}$ Oxidation Potentials," UCRL 327, by LeRoy Eyring.

Synchrotron Injection. M. Martin

A slit, equal in width to the injection slit, was cut in the back of the injector with the result that the output of the machine jumped from 400-500 mr./hr. to about 1700 mr./hr. Slits of different widths resulted in no improvement of the intensity. The reason for this improvement in operation is at present being investigated theoretically.

Synchrotron Mesons. J. Peterson

Three progressively more discriminating experimental arrangements were tried for obtaining exposures of meson tracks.

Geometry I is shown in Fig. 1a. A stack of plates was placed into the synchrotron x-ray beam at about 26 inches from the target at a point where the half width of the beam is about 1/3 inch. No control over the material in which the mesons are produced is possible with this arrangement. The mesons that were observed may have originated in the glass or the emulsion of the plates. In order to achieve better control over the target material for meson production, geometry II of Fig. 1b was tried. Two stacks of photographic plates were placed two inches apart on opposite sides of the beam in a plane passing through the beam. A sheet of carbon exposed to the flow of x-rays served for a source of mesons. The angular distribution (Fig. 2) of the mesons that were observed showed however that a large number of them must have originated in the plates since it is clearly impossible for mesons that leave the target at angles greater than or equal to 180° and equal to or less than 0° to reach the plates, and yet mesons were observed at those angles. In order to minimize the possibility of meson production anywhere but in the target still further, the beam was first collimated by two lead bricks that allowed a 1 inch aperture for its passage. This geometry is shown in Fig. 1c. Carbon was chosen for a tractable target because of its low atomic number and consequent low cross section for pair production. It was desired to avoid as much as possible the confusion of mesons with electron tracks.

Geometry III was comparatively successful in reducing the darkening of the plates by electrons. This darkening is the main factor that limits the length of exposure of the plates. It was possible to subject the plates in geometry III to 2000 mr., whereas geometry II and I allowed only 500 mr. and 50 mr.

The meson tracks that were observed can be classed into four categories: stars, π -μ decay tracks, clubs, and virgin tracks. The shape of each one of these types is sketched in Fig. 3. Club is the term that is used for a track that ends in a tight little knot that makes it impossible to judge the manner of decay or death. Virgin denotes those tracks that end uneventfully. The other two names are self-explanatory.

It is possible without using a magnetic field for positive identification by drawing on previous experience to estimate the number of positive and negative π mesons and to compute the ratio of $\frac{\text{no. of } \pi^+}{\text{no. of } \pi^-}$. Experience with mesons has shown that all but about 25 percent of π^- mesons make stars in the emulsion. Therefore, the number of π^- mesons is approximately equal to 4/3 of the number of stars. It is believed at present that only π^+ mesons decay into μ mesons, and that each π^+ meson gives rise to a μ meson. Since the juncture of parent with the daughter is often not seen, the probability that a track that may not be assigned to a π^- meson is a π^+ equals the probability that it is a μ. With geometry I, 37 stars, 9 virgins, 6 clubs, and 3 π-μ decays were observed. A calculation carried out according to the above reasoning yields therefore

$$\frac{\pi^-}{\pi^+} = \frac{4/3 \times 37}{\frac{1}{2}\left[55-4/3 \times 37\right]} \cong 17$$

The results of measurements with geometries II and III yield the values 8.1 and 7.4 for this ratio. There are a number of reasons why these numbers should be regarded as no more than indicative of the order of magnitude. The most obvious one is that the statistics are poor. Another one is that it is difficult to distinguish the track of a one prong star and a π-μ decay and that there is considerable uncertainty regarding the identification of π-μ tracks.

Geometry III yielded some crude information on the angular distribution. The graph of Fig. 4 is based only on those tracks that were acceptable. In order to assure that a meson that was counted was actually produced by the interaction of the x-ray beam with the nuclei of the target, it was required of acceptable tracks that their extension intersect with the line of the beam in the target.

A basic energy distribution was obtained by calculating from the geometry the length of the meson path in glass. The preliminary results are sketched in Fig. 5. It must be remembered that the curve cannot represent a true total energy distribution since only those particles are observed that are at angles in the neighborhood of 90°. It is expected that if all angles were seen by the plates, the energy distribution would have more nearly the appearance indicated by the broken line. The threshold for the production of mesons is 180 Mev and the total energy that is available in the x-ray beam is about 300 Mev.

Stars in Oxygen and Helium. J. Tracy.

A study has been carried out on a number of stars produced in the cloud chamber by the exposure of oxygen, H_2O vapor and helium at a pressure of $\frac{1}{2}$ atmosphere to 90 Mev neutrons. The aim of the analysis was the identification of the atoms whose disintegration initiated the stars and of the emitted particles whose tracks are seen as the prongs of the star. 82 stars were examined. However, complete identification of only a few was possible.

Events that take place in the cloud chamber are recorded photographically. The length of exposure of the film is controlled by the duration of a spark that is synchronized with the beam. The photographs are examined in a stereographic viewer that permits under optimum experimental conditions the measurement of the radius of curvature of the track, the change of the radius of curvature, and the spatial orientation of the track. When the prong of a star ends in the illuminated region of the cloud chamber, it is also possible to determine its length and thus gain information about the momentum of the particle. The density of ionization specified as light, medium, or heavy and the qualitative appearance of the track were also drawn on for help in the identification.

The principles of conservation of charge, energy and momentum were used in the analysis. Of all particles emitted in a disintegration only those carrying a charge are capable of ionizing the gas of the cloud chamber, producing tracks, and therefore, being seen on the photographs. A charge balance was therefore always possible. A consideration of charge leads to the immediate conclusion that stars with more than two prongs can only be due to the disintegration of an oxygen atom. The principle of conservation of energy could not be used for identification except in the elimination of alternatives that required the '90 Mev' neutrons to come in with an energy in excess of the maximum. A momentum balance also could be used only for the elimination of alternatives since the possible emission of invisible neutrons had to be considered.

It was possible however in the case of 7 He stars to establish an energy balance since the range of all the prongs could be measured and therefore their energy could be determined by means of the range energy curves for various charged particles.

The details of this analysis are set forth in "Star Fragments in Oxygen and Helium under Bombardment by 90 Mev Neutrons," J. Tracy and Wm. Powell, UCRL 346.

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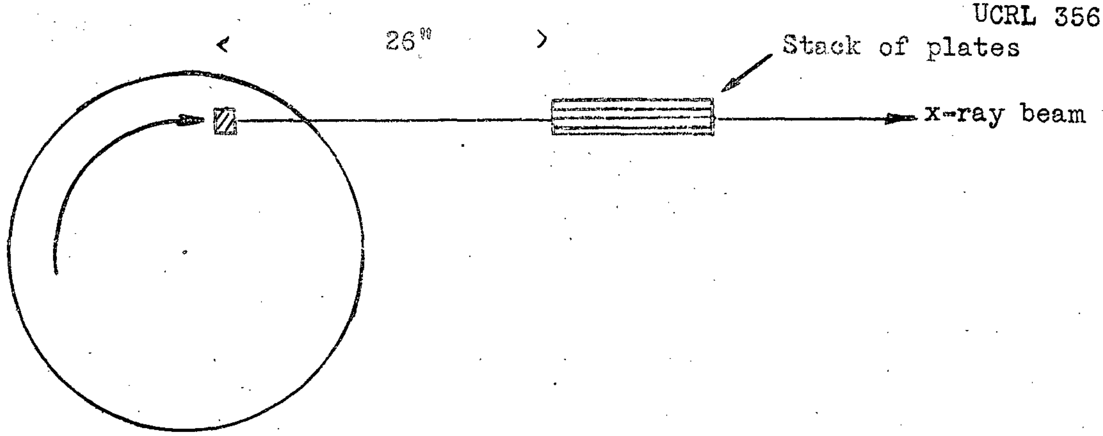
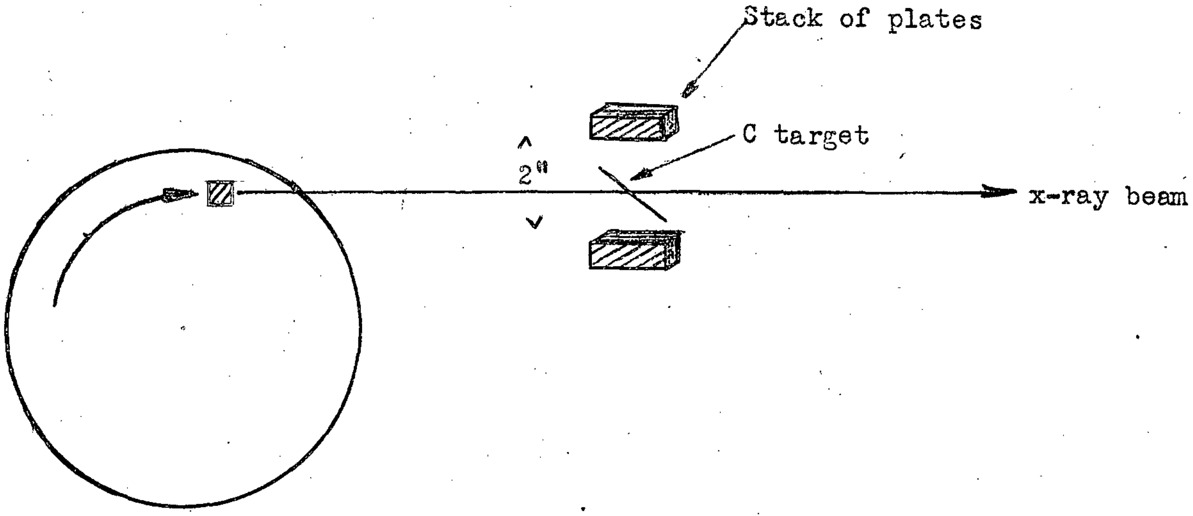
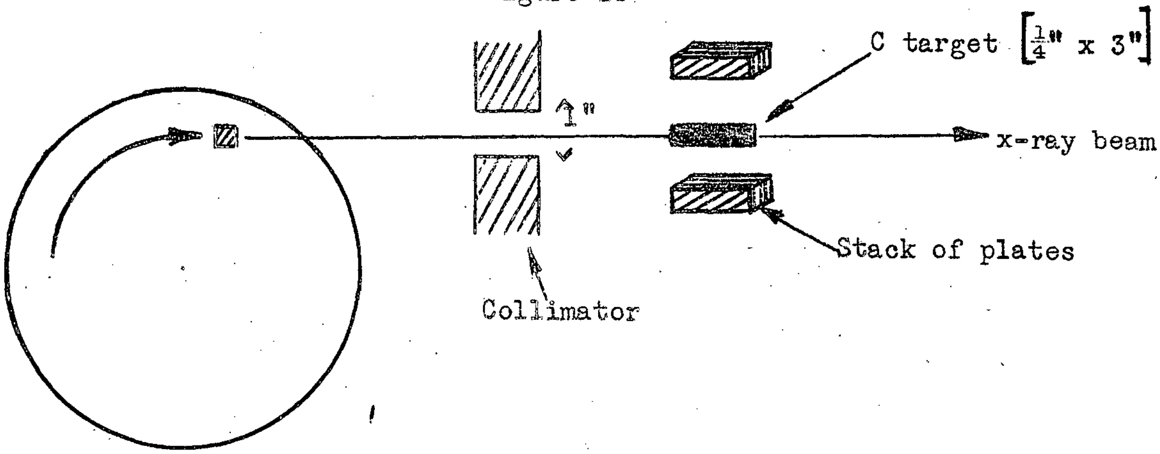


Geometry I

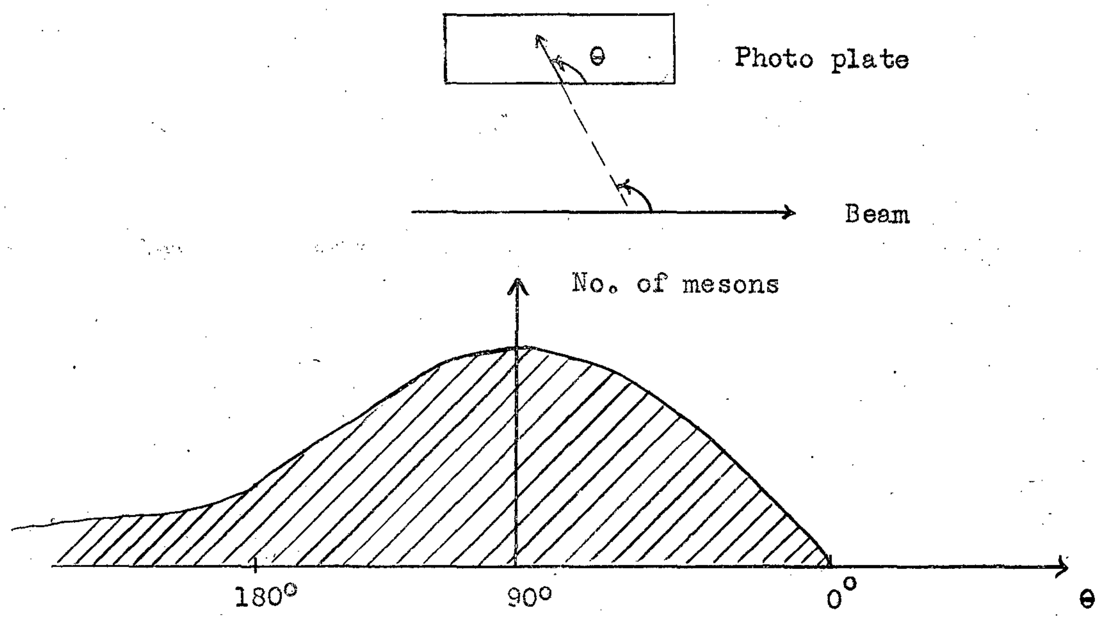
Figure 1a



Geometry II
Figure 1b



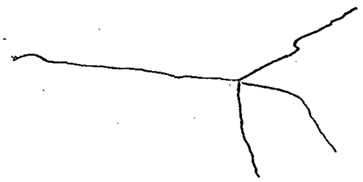
Geometry III
Figure 1c



Angular Distribution for Geometry II

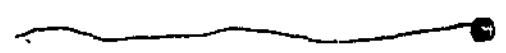
Figure 2

1. Star



2. $\pi - \mu$

3. Club



4. Virgin

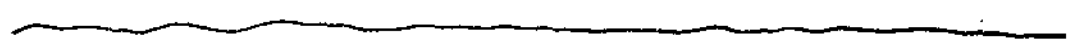
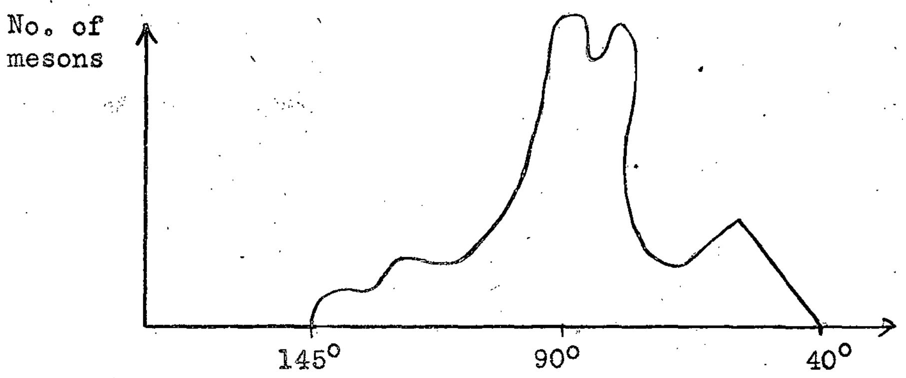
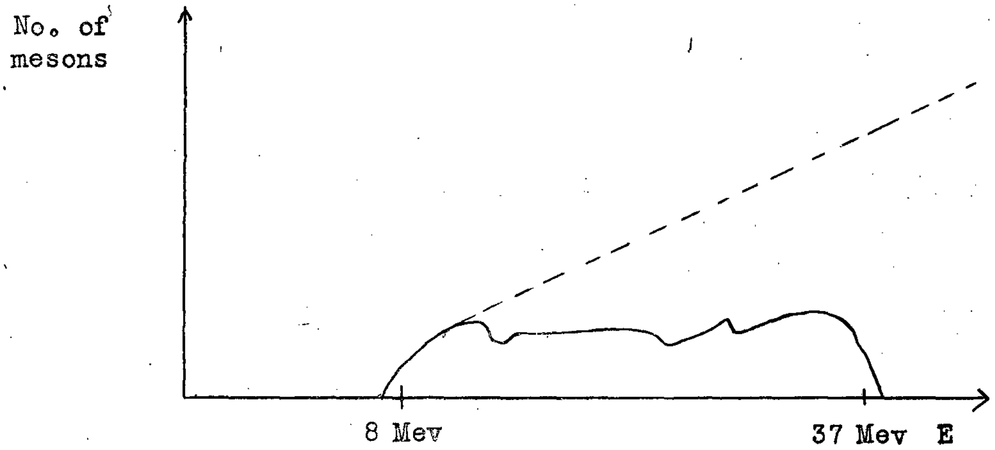


Figure 3



Angular Distribution for Geometry III

Figure 4



Basic Energy Distribution for Geometry III

Figure 5

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