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June through November 1958

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NUCLEAR CHEMISTRY

Glenn T. Seaborg and Isadore Perlman in charge

Nuclear ReactionsSPALLATION REACTIONS INDUCED IN URANIUM ISOTOPES
BY HELIUM IONS

Victor E. Viola and T. Darrah Thomas

Experiments are in progress to define in more detail the excitation curves for the (α, xn) reactions of uranium isotopes. Results for the reactions of U^{233} and U^{235} are in agreement with those reported by Vandenbosch et al.,¹ except for the excitation curve for the $U^{233}(\alpha, 3n)Pu^{234}$ reaction. Our measurements show that, at the peak of the excitation curve, the cross section is about 70% higher than was reported by Vandenbosch et al.

REACTIONS OF GOLD WITH CARBON IONS AND OF
PLATINUM WITH NITROGEN IONSH. Marshall Blann, Glen E. Gordon, Robert M. Latimer,
E. Thomas Strom, Victor E. Viola, and T. Darrah Thomas

The excitation functions for the $Au^{197}(C, xn)$ and $Pt(N, xn)$ reactions are being measured. In the gold bombardments products of the evaporation of four, five, and six neutrons have definitely been observed. There is evidence that, at the lowest bombarding energies, the products of two- and three-neutron evaporation have been seen. Alpha activity of 8.97 Mev with a half life of 9 minutes has been observed and assigned to At^{204} on the basis of the excitation curve for its production and the growth of Po^{204} . By milking experiments it has been shown that the alpha-branching ratio of the 9-minute isomer is about 7% (assuming an alpha-branching ratio of 1% for Po^{204}). Similarly, an alpha-branching ratio of between 15 and 25% has been established for At^{205} (assuming an alpha-branching ratio of 0.074% for Po^{205}).

¹Vandenbosch, Thomas, Vandenbosch, Glass, and Seaborg, Phys. Rev. 111, 1358 (1958).

There is an indication that there is an isomer of At^{206} having a half life of ~ 32 minutes and emitting alpha particles of ~ 5.80 Mev, with an alpha-branching ratio of about 6%.

Because of uncertainties in the energy of the carbon ion beam, it is impossible at present to give excitation functions for the reactions observed. Cross sections for the $\text{Au}^{197}(\text{C}, 4n)\text{At}^{205}$ reaction, calculated on the basis of an alpha-branching ratio of 25%, have a maximum of about 70 millibarns, and for the $\text{Pt}^{198}(\text{N}^{14}, 5n)\text{At}^{207}$ reaction have a maximum of 160 millibarns.

Preliminary work on the angular distribution of the fragments from fission induced in gold by heavy ions indicates that the angular distribution is not $1/\sin\theta$, as would be predicted from the most naive considerations. A value of η , the ratio of the velocity of the fission fragments (if formed from a nucleus at rest) to the velocity of the center of mass of the system, equal to 0.22 has been found for the fission fragment Mo^{99} . This value of η corresponds to a fission energy of about 130 Mev, in agreement with what one calculates from mass differences as given by semiempirical mass formula.

Studies of the mass and charge distribution of the fragments from fission induced in gold with carbon ions are being continued.

CROSS SECTIONS FOR THE FORMATION OF A COMPOUND NUCLEUS WITH HEAVY IONS

T. Darrah Thomas

Cross sections for formation of a compound nucleus from various systems of heavy ions and targets have been calculated by using a simple model. The assumptions of this model, which has been taken from Blatt and Weisskopf,¹ are as follows:

1. The potential energy of the system is given by

$$V = \frac{Z_1 Z_2 e^2}{r} + \frac{\hbar^2}{2\mu} \frac{l(l+1)}{r^2}, \quad \text{for } r > R,$$

$$V = -\frac{\hbar^2 K_0^2}{2\mu}, \quad \text{for } r < R,$$

$$K_0 \approx 10^{13} \text{ cm}^{-1},$$

¹ Blatt and Weisskopf, Theoretical Nuclear Physics (John Wiley and Sons, New York, 1952).

where r is the distance between the centers of the two interacting particles, Z_1 and Z_2 are the atomic numbers of target and projectile, μ is the reduced mass of the system, and $\ell \hbar$ is the orbital angular momentum of the incoming particle.

2. R , the radius of interaction, is equal to $r_0(A_1^{1/3} + A_2^{1/3})$, where A_1 and A_2 are the mass numbers of target and projectile.

3. For $r > R$, there is no nuclear interaction, and for $r < R$, there is a strong nuclear reaction, with the result that for $r < R$, the particle is absorbed. Hence, the wave function of the particle in the nucleus can be represented approximately as

$$u \sim e^{-iKr}$$

The formula for the compound-nucleus cross section is

$$\sigma = \pi \chi^2 \sum_{\ell=0}^{\infty} \frac{4 K R S_{\ell}}{\Delta_{\ell}^2 + (K R + S_{\ell})^2},$$

$$S_{\ell} = kR \left(\frac{1}{F^2 + G^2} \right),$$

$$\Delta_{\ell} = kR \left(\frac{FF' + GG'}{F^2 + G^2} \right),$$

$$K = (k^2 + K_0^2)^{1/2},$$

$$\chi = \frac{1}{k},$$

$$F' = \frac{dF}{d(kr)},$$

$$G' = \frac{dG}{d(kr)},$$

where F and G are Coulomb functions and k is the wave number of the incoming particle. The Coulomb functions and their derivatives are evaluated for $r = R$. The methods of Fröberg were used to calculate these functions.²

²C. E. Fröberg, Revs. Modern Phys. 27, 399 (1955).

Comparisons of the calculated cross sections with experimentally determined ones are shown in Figs. 1, 2, and 3. Figure 1 shows cross sections for fission of uranium with different projectiles reported by Flerov.³ It has been assumed that the fission cross section is equal to the cross section for formation of the compound nucleus. Figure 2 shows the sum of fission and-spallation cross sections measured by Flerov and co-workers for nitrogen-ion-induced reactions in gold.⁴ The value of 1.5 fermis for r_0 has been used in these calculations. In Fig. 3, calculations for the system $K^{39} + N$ based on $r_0 = 1.5$ and $r_0 = 1.37$ are compared with experimental values reported by Pinajian.⁵ The lower dashed curve represents the sum of the cross sections for all reactions reported, and the upper curve represents the sum of all cross sections reported plus an estimate for those cross sections not reported.

In Fig. 4 are shown typical angular-momentum distributions, and in Fig. 5 the average value of angular momentum is plotted against energy of incident particle for several systems.

Calculations have been begun to determine the effect of a diffuse nuclear surface on the cross section for formation of a compound nucleus.

³G. N. Flerov, Proceedings of the Conference on Reactions Between Complex Nuclei, Gatlinburg, 1958, ORNL-2606.

⁴Baraboshkin, Karamian, and Flerov, Zhurn. Eksptl. i Teoret. Fiz. 32, 1294 (1957).

⁵J. J. Pinajian, Proceedings of the Conference on Reactions Between Complex Nuclei, Gatlinburg, 1958, ORNL-2606.

MU-16426

Fig. 1. Comparison of experimental and theoretical values of the cross section for compound-nucleus formation. (Data from Reference 3.)

MU-16427

Fig. 2. Comparison of experimental and theoretical values of the cross section for compound-nucleus formation. (Data from Reference 4.)

MU-16429

Fig. 3. Comparison of experimental and theoretical values of the cross section for compound-nucleus formation. Lower dashed curve represents summation of cross sections reported in Reference 5; upper dashed curve is the same plus an estimate of cross sections not reported. (Data from Pinajian⁵.)

WU-16432

Fig. 4. Typical angular-momentum distributions.

Fig. 5. Average angular momentum plotted versus energy of incident particle. Dashed curve is based on a classical calculation.

COULOMB EXCITATION

Frank S. Stephens, Jr., and Richard M. Diamond

Coulomb excitation work has been continued using heavy-ion projectiles from the heavy-ion linear accelerator. This machine will be most advantageous for bombarding the heavier elements, in which the Coulomb barrier is high enough to permit relatively high projectile energies, hence excitation of high-lying (~ 1 Mev) levels. However, preliminary work to date has been on projectile excitations. That is, the energy and yield of the gamma rays from the projectiles themselves have been studied. Since suitable high-energy heavy-ion beams have not previously been available, this is one of the first studies of this type.

Projectiles looked at have been $B^{10,11}$, C^{12} , N^{14} , O^{16} , F^{19} and $Ne^{20,22}$. The Coulomb-excitation gamma rays have been detected with a 1.5×1 -in. or a 3×3 -in. NaI crystal counter.

Of the projectiles studied, no radiations were observed from B^{11} , N^{14} , or O^{16} . This would be expected from previous knowledge of the energy levels of these nuclei. We were just able to observe the 4.4-Mev level of C^{12} . As soon as information is available on the rate of energy loss of C^{12} in gold (dE/dx), we can calculate a half life for the 4.4-Mev level in C^{12} , and compare this with the known half life. The 0.72-Mev level of B^{10} was excited, and the yield of the gamma ray from this level was measured as a function of the B^{10} ion energy. Again, the final results are waiting for information on the dE/dx of B^{10} in gold. (If these dE/dx values are not available soon, we can rather easily make the necessary measurements.) We are currently studying F^{19} , and in addition to the 0.11- and 0.20-Mev levels, excitation is seen in level(s) around 1.5 Mev. More work is required to know exactly which level(s) of F^{19} are being excited in this region.

The most extensive work has been done on Ne^{20} . The yield of the gamma ray from the 1.63-Mev level of Ne^{20} has been rather carefully measured as a function of projectile energy. The results are in general agreement with Coulomb excitation theory, but quantitatively the yield does not increase with projectile energy as rapidly as expected. This is probably due to the inadequacy of the theory at the high probability for excitation encountered here. The angular distribution of the gamma rays with respect to the beam direction was in reasonable agreement with the theoretical expectations. Similar, though not quite so extensive, results were obtained for Ne^{22} .

NUCLEAR REACTION MECHANISMS

Bernard G. Harvey and Jose Gonzalez-Vidal

We have approached the study of nuclear reactions by an examination of the angular distribution of the light products involved. Such a study, together with theoretical treatments, for example, the Butler theory, can give insight into the mechanism of a nuclear reaction.

The determinations were carried out by using the 36-inch scattering chamber at the Crocker Laboratory 60-inch cyclotron. The detector system was composed of two crystals arranged in series so that in passing through the first crystal a given particle lost a small amount of energy and hence gave a light pulse proportional to dE/dx . The particle was then stopped in the second crystal and gave a light pulse proportional to E . The two light pulses were converted into electrical pulses by means of photomultipliers and multiplied together to give a pulse proportional to $E \times dE/dx$, and hence also proportional to mv^2 . Such a procedure permits the identification and separation of different kinds of particles. The multiplied pulse was used as a gate for a pulse analyzer, after appropriate discrimination; this allowed the study of the energy spectrum of any one desired kind of particle.

The system as described was used to study the $Al^{27}(\alpha, t)Si^{28}$ reaction, which had been previously investigated with the use of a range telescope. Only 48-Mev helium ions were used in this case. It was found that the present system is superior to the range telescope system as far as background problems are concerned, since with the range telescope it was possible to observe only tritons corresponding to the ground state and first excited state of Si^{28} . With the present setup it was possible to see tritons corresponding to as many as ten energy levels of Si^{28} . This type of study is an important tool in the determination of high-lying nuclear energy levels.

Another reaction subjected to examination was the (α, pn) reaction, which had been previously investigated radiochemically. Here the main interest was to know how much of the reaction, if any, was really an (α, d) reaction. For this purpose the multiplied pulse itself was fed into a pulse analyzer, with the E pulse used as the trigger and the discriminators on the E pulse set up so that no (α, dxn) would be detected. It was observed that the (α, d) reaction had a cross section comparable to that of the (α, t) reaction. The over-all angular distributions were peaked in the forward direction, thereby indicating the presence of a direct-interaction process. The (α, d) reaction was investigated at several helium-ion energies with the object of obtaining excitation functions. The integrated (α, d) cross sections seem to account for about a third of the radiochemical (α, pn) cross section.

At present a similar system is being tested for use in the study of nuclear reactions produced by heavy ions. A scattering chamber with some novel features is in construction and will be described. For preliminary investigations a rough system has been devised consisting of pipe sections which are provided with a Faraday cup, a target holder, and counting system which allows the investigation of six different angles. Preliminary results have shown that the $E-dE/dx$ system already described will also be useful for studying heavy-ion-induced reactions.

RANGE-ENERGY RELATION FOR HEAVY ATOMS

Bernard G. Harvey, Paul F. Donovan, John R. Morton,
and Ernest W. Valyocsik

Atoms of various known kinetic energies can be produced by bombardment of suitable targets with protons, deuterons, helium ions, and various heavy ions. If a nuclear reaction occurs which is known to involve or can be shown to involve the formation of a compound nucleus, then the momentum of the resulting recoil atom must be equal to the momentum of the incident particle. In this way, recoil atoms with known kinetic energies between a few tenths Mev and several Mev can be generated.

The ranges of such recoil atoms have been studied in various gases (H_2 , D_2 , He, N_2 , CH_4 , Ne, and Ar). The reactions $Ra^{226}(\alpha, n)Th^{227}$ and $Tl^{205}(C^{12}, n)Fr^{212}$ have been used; others will be investigated. The apparatus is shown in Fig. 6. The target material is deposited as a thin layer on a thin foil through which the beam can pass. The recoiling atoms are ejected into a gas-filled space between two parallel aluminum plates. The lower plate is maintained at a potential of -600 volts relative to the upper plate. Recoiling positive ions are stopped by the gas, and then drift down the electric field lines to the lower plate. The plate is finally cut into strips, and the number of recoiling atoms collected on each strip is measured by counting their radiations. In this way, differential range curves such as the curve in Fig. 7 are obtained. This technique was first used in this laboratory by Albert Ghiorso and Torbjorn Sikkeland (unpublished).

Theoretically, the range of recoiling atoms with energies up to a few Mev should be proportional to the energy of the atoms, and this is found to be the case. When ranges are expressed in mass units per unit area, the ranges of recoiling atoms stopping in gases are given by the equation of Lindhard and Scharff,¹

$$R(\mu g/cm^2) = 602 E M_2 \frac{(M_1 + M_2)}{M_1} \frac{\sqrt{Z_1^{2/3} + Z_2^{2/3}}}{Z_1 Z_2}$$

where E is the recoil energy in Mev, M_1 and Z_1 are the mass number and atomic number of the recoil atom and M_2 and Z_2 are the same quantities for the stopping medium. For a heavy-element recoil in a light gas, $M_1 \gg M_2$ and $Z_1 \gg Z_2$, and the equation reduces to

$$R(\mu g/cm^2) \approx 602 E \frac{M_2}{Z_2} \cdot Z_1^{-2/3}$$

For most light gases (e.g., N_2 , Ar, D_2), $M_2 = 2 Z_2$. However, for H_2 , $M_2 = Z_2$, and hence ranges in hydrogen should be only half as great as ranges in the other gases. This is confirmed experimentally for 0.725-Mev Th^{226} recoils.

¹J. Lindhard and M. Scharff, Kgl. Danske Videnskab. Selskab. Mat-fys. Medd. (to be published).

MU-16780

Fig. 6. Apparatus for studying ranges of recoil atoms in various gases.

MU-16872

Fig. 7. Differential range curves for 0.725-Mev Th^{226} recoils
 from $\text{Ra}^{226}(\alpha, 4n) \text{Th}^{226}$.

Gas	Range ($\mu\text{g}/\text{cm}^2$)
H ₂	18
He	30
D ₂	32
N ₂	38
A	40

The width of the differential range curve for monoenergetic recoils should be a function of the mass of the atoms of the stopping medium. Therefore the range curves obtained with argon should be much broader than those obtained with hydrogen. Only a small difference is in fact observed: the hydrogen range curves are much broader than they should be. This is probably due to the method used to generate recoil atoms. The emission of the neutrons broadens the energy spectrum of the recoils, and the observed range curves are indeed consistent with this hypothesis.

ENERGY AND ANGULAR DISTRIBUTION OF RECOIL NUCLEI

Bernard G. Harvey, William H. Wade, and Paul F. Donovan

Studies of the angular distribution of At^{211} recoil nuclei generated by the nuclear reaction $\text{Bi}^{209}(\alpha, 2n)\text{At}^{211}$ were continued. The experimental distributions do not agree well with calculated distributions obtained by assuming isotropic emission of both the neutrons. At incident helium-ion energies greater than about 33 Mev, it seems that the reaction takes place by the emission of a fast neutron in the forward direction followed by isotropic evaporation of the second neutron. Measurements of the kinetic energy of the At^{211} recoil nuclei support this hypothesis. This work is being prepared for publication in The Physical Review.

RANGE-ENERGY RELATIONSHIPS FOR FISSION PRODUCTS

John M. Alexander and M. F. Gallagher

In recent years there has been an increasing interest in the study of the fission reaction by measurements of recoil ranges. A knowledge of the range as a function of the recoil energy is of course essential to these studies. Theoretical guidance on this problem is only qualitative in nature because of the very difficult problem of electron-capture and -loss processes. A number of experimental studies have pointed out the qualitative features of stopping of fission products, but very few investigations have attained a high degree of accuracy.

Of particular interest as stopping materials are the metals aluminum,¹ bismuth,² and uranium.³ A careful radiochemical study of the average ranges of various products in uranium is being carried out by J. B. Niday at UCRL Livermore. However, no energy-loss data are available for this material; the heaviest stopping material for which energy-loss data are available is gold.⁴ Energy-loss measurements have been made for aluminum,⁵ but the total-range measurements from different laboratories^{6, 7} differ by ~10%. It is therefore apparent that accurate range measurements of various fission products in aluminum and gold would be useful. Measurement of these ranges to an accuracy of ~1% is being undertaken by the method described by Douthett and Templeton.⁸

In this method the fragments are stopped in catcher foils of uniform thickness ~1/2 the range to be measured (0.00025-inch aluminum fulfills this requirement and is readily available). The preliminary results obtained to date are shown in Fig. 8. Also shown in Fig. 8 are the measurements from References 6 and 7, both obtained by stopping the fragments in many aluminum foils of ~0.15 mg/cm². Our measurements are seen to be consistently 2% lower than those in Reference 6 and 7% higher than in Reference 7.

¹ See pp. 25-26.

² Norbert T. Porile and N. Sugarman, Phys. Rev. 107, 1410 (1957); Sugarman, Compos, and Wielgoz, Phys. Rev. 101, 388 (1956); Norbert T. Porile, Phys. Rev. 108, 1526 (1957).

³ See pp. 27-29.

⁴ Clyde B. Fulmer, Phys. Rev. 108, 1113 (1957).

⁵ H. W. Schmitt and R. B. Leachman, Phys. Rev. 102, 183 (1956).

⁶ P. F. Suzor, Ann. phys. 4, 269 (1949).

⁷ Freedman et al., Paper 44; Finkle et al., Papers 45 and 46, in Radiochemical Studies: The Fission Products, C. D. Coryell and N. Sugarman, Eds., Division IV, Vol. 9, Book 1 (McGraw-Hill Book Company, New York, 1951).

⁸ E. M. Douthett and D. H. Templeton, Phys. Rev. 94, 128 (1954).

MU-16693

Fig. 8. All measurements of range in aluminum of products from the thermal-neutron-induced fission of U^{235} are shown here. The circles are from this work, the squares from Suzor,⁶ and the triangles from Reference 7. The smooth curve was drawn by attributing a systematic error of 7% to the measurements of Reference 7.

These differences can be reasonably attributed to the lack of uniformity of the thin foils which would affect the absolute values but not the relative values of the ranges.

Energy-loss measurements have been made by Schmitt and Leachman for the "median" light and heavy fragments.⁵ The median masses 94.2 and 139.3 are determined by the conservation of momentum between the fragments and the conservation of nucleons. The range-velocity curves shown in Fig. 9 were drawn from the energy-loss data by taking the total range values from the smooth curve of Fig. 8. The low-velocity part of the curves, $v < 0.2$ (Mev/nucleon mass)^{1/2}, was drawn from the theoretical relationship which was verified in other work (Eq. (12), p.41).

In the future, range measurements will be made for several more products and for gold as a stopping material. Range-velocity curves for gold may then be constructed from the energy-loss measurements by Fulmer.⁴ Thus there will be an experimental test of the common assumption that the range of a given product in one material divided by range in another material is independent of the mass and energy of the product.

MU-16690

Fig. 9. The range-velocity curves presented here were drawn from the three sources of information. The average total ranges (filled points) were taken from Fig. 8. Energy measurements corresponding to various degrees of energy degradation were taken from Schmitt and Leachman⁵ (open points). Triangles represent the median light mass of 94.2, and circles the median heavy mass of 139.3. The solid curve for low velocities was taken from Eq. (12) p. 41.

RECOIL STUDIES OF FISSION PRODUCTS

John M. Alexander and M. F. Gallagher

Nuclear fission is one of the predominant reactions occurring when a heavy element is bombarded with neutrons or charged particles. In fact, it has been found that charged-particle bombardment of any element of $Z > 90$ gives rise to fission in at least 90% of the reactions. Thus, an understanding of the nuclear reactions of heavy elements is intimately related to a knowledge of the fission process. One informative experimental technique for the study of fission is the radiochemical method applied to recoil properties of the various products. The recoil properties reflect the energy released in the fission act, the angular distribution of the products, and the momentum of the fissile nucleus at the instant of fission. We are currently engaged in an investigation of these three aspects of the fission reaction for a variety of target elements and a variety of incident bombarding particles.

Several different techniques for recoil measurements have been described in the past,¹⁻⁴ but none of these is alone sufficient to determine simultaneously (a) momentum of struck nucleus, (b) angular distribution of the products, and (c) energy release. We have devised an experimental technique which does simultaneously measure these properties.

In this technique a very thin target ($< 10\%$ of the range of the products) is sandwiched between stacks of 0.00025-inch aluminum foils of carefully measured thickness, and is exposed to a beam of particles. Each foil is dissolved separately, certain products are chemically isolated, and the relative radioactivity and chemical yields are accurately measured.

Let $\Omega(\theta)$ represent the angular distribution relative to the beam direction in the frame of the moving fissile nucleus and $F(t)$ the fraction of the activity penetrating beyond a thickness t of aluminum from a target of thickness W :

$$F(t) = \frac{\int_0^W dx \int_0^{\theta_{\max}} \Omega(\theta) 2\pi \sin \theta d\theta}{\int_0^W dx \int_0^{\pi} \Omega(\theta) 2\pi \sin \theta d\theta} \quad (1)$$

¹E. M. Douthett and D. H. Templeton, Phys. Rev. 94, 128 (1954).

²N. T. Porile and N. Sugarman, Phys. Rev. 107, 1410 (1957).

³Cohen, Jones, McCormick, and Ferrell, Phys. Rev. 94, 625 (1954);
Cohen, Ferrell-Bryon, Coombe, and Hullings, Phys. Rev. 98, 685 (1955).

⁴C. T. Coffin and I. Halpern, Phys. Rev. 112, 536 (1958).

Let x denote the thickness of target material corresponding to the distance of the fissile nucleus from the surface of the target. A quantity η is defined as the velocity v (parallel to the beam) of the fissile nucleus in the laboratory system divided by the speed V of the fission product in the struck nucleus system (see Fig. 16, p. 45). The laboratory angle θ_L is related to the angle in the moving frame θ by

$$\cos \theta_L = \frac{\eta + \cos \theta}{(1 + \eta^2 + 2\eta \cos \theta)^{1/2}}, \quad (2)$$

and the limit of integration $\theta_{\max}$ is determined by the range $R(V, \theta, \eta)$ of the fragments at $\theta_{\max}$ and the thickness (t) of the aluminum in question. The factor C is used to convert thickness of target material to equivalent thickness of aluminum. Thus we have

$$\cos \theta_{L \max} = \frac{Cx + t}{R(V, \theta, \eta)} \quad (3)$$

If the range-velocity relationship is known (see p. 21) Eqs. (2) and (3) can be solved for $\theta_{\max}$ in terms of x , t , V , and η . Previous experimental work³ and theoretical expectation lead us to believe that $\Omega(\theta)$ is symmetric about $\theta = 90^\circ$ and thus Eq. (1) may be integrated for the approximating angular distribution function,

$$\Omega(\theta) = 1 + \sum_{n=1}^N a_{2n} \cos^{2n} \theta.$$

Thus, as an example, if the fission product separated is found in five aluminum foils, then four independent $F(t)$ equations may be used to determine η , V , and two angular-distribution parameters. For cases in which η is very small, experimental uncertainties severely limit the accuracy of the angular-distribution parameters. In these cases it is desirable to measure the angular distribution by an independent technique.

This development is clearly dependent on the following assumptions: (a) the struck nucleus is not slowed down before fission, (b) kinetic energy released in fission is independent of θ , (c) the angular distribution is symmetric about 90° , (d) any velocity component of the struck nucleus perpendicular to the beam is unimportant. Assumption (a) may be considered justified experimentally if the results indicate cases of complete momentum transfer from incident particle to target nucleus. Assumption (c) requires that complementary products indicate identical momentum-transfer and angular-distribution parameters.

Experimental data by this technique have been obtained for uranium targets bombarded with H^2 , He^4 , C^{12} , and Ne^{20} ions, and for Au^{197} and Bi^{209} targets exposed to beams of C^{12} and Ne^{20} . The solution of Eq. (3) requires the use of an electronic computer, and this phase of the analysis is in progress.

THE PRODUCTION OF IODINE FROM URANIUM BY BOMBARDMENT IN THE ENERGY REGION OF BILLIONS OF VOLTS

John M. Alexander and M. F. Gallagher

Cross-section data from various targets reveal striking changes in yield-mass curves which take place as bombarding energies are increased from hundreds of millions of volts to billions of volts. The changes may be summarized very roughly by the statement that the total isobaric yield of each mass, excluding $A < 10$ and $A > (\text{target mass} - 10)$, is approximately constant for Bev bombardments, while at a few hundred Mev, distinct groups of products are observed. In the group called "spallation products" the products have masses not very different from the target. The heavier elements, which are known to undergo nuclear fission, give rise to another group of products with mass about one-half the target mass. This distinction between fission and spallation, which is quite clear from the yield-mass curves at several hundred Mev, loses its clarity in the Bev energy region. We must seek new criteria to distinguish the various features of Bev nuclear reactions. Thus, experimental information other than yield-mass data is required before one can even logically classify the types of reactions present. Measurement of recoil properties have furnished a great deal of useful information about the nature of reactions in the sub-Bev energy region, but few such studies have been made in the Bev region.

A detailed study of iodine products has been undertaken, as the nuclides for this element are in general shielded from beta decay by stable or long-lived parents. Experimental data based on these species therefore reflect the primary formation independent of radioactive decay. Our crude knowledge of the fission process, from lower-energy studies, enables us to draw conclusions about the distribution of the fissile nuclei from the form of the distribution of these primary yields.

Recoil measurements (by the thick-target technique¹) give the average velocity of the struck nuclei and the energy released in the moving frame of reference. The importance of fission with respect to other processes can be estimated from the kinetic energy measurements, and the struck-nucleus velocities are directly related to the initial interactions.

An important aspect in the interpretation of the recoil data is a reaction model which relates the deposition-energy spectrum and residual-nucleus spectrum to the velocity imparted to the struck nucleus. The best model we have for this purpose is the Serber model of nucleon-nucleon interactions. Predictions based on this model are being made by a large group at the Los Alamos Laboratory and elsewhere, but the calculations are not yet available. Interpretation of experimental results is thus postponed until the calculations have been completed.

Preliminary cross-section data are presented in Figs. 10 and 11. The errors indicated are simply the average deviation of the measurements and do not include systematic errors. The most striking feature of the results is the very broad distribution of yields. The apparent separation into two groups of products at 6.2 Bev is outside the random experimental errors and therefore believed to be a real effect.

¹Sugarman, Campos, and Wieljoy, Phys. Rev. 101, 388 (1956).

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Fig. 10. The variation of cross section with mass number is shown for iodine production from U^{238} by 0.72-Bev proton bombardment.

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Fig. 11. The variation of cross section with mass number is shown for iodine production from U^{238} by 6.2-Bev proton bombardment.

ISOTOPES OF MENDELEVIUM

Raymond C. Gatti, Llad Phillips, and M. Luis Muga

A new mendeleevium isotope (Mv^{255}) was discovered by bombarding approximately 1 millimicrogram of E^{253} with helium ions at the 60-inch cyclotron. The results of the experiments are summarized as follows.

Summary of isotopes observed following helium-ion bombardments of E^{253} .

Isotope	Mode of decay	Half life	Cross section (mb)			
			at 24 Mev	at 29 Mev	at 36 Mev	at 42 Mev
Mv^{256}	Electron capture	~1.5 hr	~0.1	~1	~1	~1
Mv^{255}	Electron capture	~1/2 hr	*	~1	~1	~1
	7.34-Mev α	~3/4 hr	*	~0.2	~0.2	~0.2
Fm^{256}	Spontaneous fission	160±10 min	~0.1 ^a	~1 ^a	~1	~1 ^a
Fm^{255}	7.08-Mev α	21.5 hr	~0.4 ^a	~4 ^a	~6 ^a	~6 ^a
Fm^{254}	7.20-Mev α	3.24 hr	*	~0.5 ^a	~10 ^a	~10 ^a

^aThese are over-all cross sections.

* Activity insufficient for calculations of cross section.

A more detailed account of this work has been published in Phys. Rev. Letters 1, 215 (1958).

FISSION STUDIES OF HEAVY ELEMENTS

M. Luis Muga

Investigation of fission phenomena induced by heavy-ion bombardments of the actinide elements continues.

Further experiments on the $U^{238}(Ne^{20}, f)$ and $U^{238}(C^{12}, f)$ reactions to ascertain the possibility of very asymmetric fission do not confirm previous indications that light fission fragments ($A \sim 30$) were being formed.

The study of fission processes by means of nuclear emulsion techniques has resulted in the observation of several interesting events when nuclear emulsions, impregnated with U^{238} , were exposed to a beam of $18A^{40}$ ions. These events, characterized by a large Z^2/A value for the intermediate compound nucleus, are similar in appearance to cosmic-ray stars of 4 to 6 charged recoil particles. It has not been possible at this time to determine whether the argon ion interacts with bromine, silver, or uranium nuclei in the emulsion.

MASS-YIELD DISTRIBUTIONS AND EXCITATION FUNCTIONS
IN HEAVY-ION FISSION

John T. Gilmore

Several bismuth and bismuth oxide targets have been bombarded with carbon ions of varying energies and excitation functions for the formation of certain fission products measured. One goal of these bombardments was to establish the feasibility of radiochemical determination of the mass-yield curve for fission of bismuth with carbon ions of about 50 Mev energy. Bismuth plus carbon ions of this energy gives a compound nucleus in the region of mass and excitation energy where a three-peaked distribution of fission products has been observed. Unfortunately, a bombarding energy of 50 Mev lies about 10 Mev below the Coulomb barrier for compound-nucleus formation, and fission cross sections fall off very rapidly with decreasing energy. It was found that the amount of fission-product activity obtainable at 50 Mev bombarding energy was too low to permit a reliable determination of the mass-yield curve.

A separate program has been initiated to study the competition between fission and neutron emission in heavy-ion bombardments of separated isotopes of iridium and rhenium. Preliminary bombardments of the naturally occurring mixtures of iridium and rhenium isotopes have helped to define experimental techniques, which will be used in bombardments of the separated isotopes.

ANGULAR DISTRIBUTION AND ENERGY SPECTRUM OF PROMPT NEUTRONS FROM THE SPONTANEOUS FISSION OF CALIFORNIUM-252

Harry B. Bowman, Wladyslaw J. Swiatecki, and Stanley G. Thompson

The neutron velocities and fission-fragment velocities were measured in coincidence by use of a cylindrical drum 1 meter in diameter. The drum was evacuated to a pressure of 10^{-6} mm of Hg. A plastic scintillator 2 by 2 inches attached to a 14-stage photomultiplier tube was located in the center of the drum. A Cf^{252} sample of 2×10^6 spontaneous fissions per minute was placed on a flat portion of the plastic scintillator. The detection of the prompt gamma rays in this crystal indicated the occurrence of fission events. A 0.002-inch-thick plastic scintillator was mounted on the face of a 5-inch-diameter photomultiplier located on the curved surface of the drum to detect the arrival of the fission fragments. The arrival of neutrons was detected by a stilbene crystal attached to a 14-stage photomultiplier tube, also located on the curved surface of the drum. The detector outputs were connected to the time-to-pulse-height converters. The pulse heights were in turn converted to digits and punched into IBM cards.

Each fission fragment was characterized as "light" or "heavy" according to its time of flight between the source and the fission detector. The time of flight of neutrons in coincidence with the fragments was measured at angles of 12° , 26° , 40° , and 90° relative to the direction of detected fragments. The measured neutron-velocity spectra were converted to absolute numbers of neutrons per fission per unit volume of velocity space $\rho(v, \theta)$ by applying the corrections due to the counting efficiency of the stilbene crystal (without arbitrary normalization of any kind).¹ Integration over all angles and intervals of velocity space (above a value corresponding to 1.18 Mev energy) gives the absolute value 2.7 as number of neutrons emitted per fission for the energy range considered. This value is close to the number expected for this energy interval on the basis of previous existing measurements.

The final results of the experiments are shown plotted in polar coordinates in Fig. 12. The distance from the origin corresponds to the neutron velocity, and the angle gives the direction of the neutron with respect to the direction of the light fragment. The contour lines connect points of constant ρ at the various measured angles and velocities. The neutrons from the light fragments are shown in the left-hand quadrant. The right-hand quadrant gives the results for neutrons from heavy fragments. The average velocity of light fragments is 1.37×10^9 cm per sec; the average velocity for heavy fragments is 1.04×10^9 cm/sec. Points representing the average fragment velocities are shown on the horizontal

¹ The term "per unit volume of velocity space" means that the neutrons have velocity components v_x , v_y , v_z in a unit cell Δv_x , Δv_y , and Δv_z .

Angular distribution of neutrons per fission per unit volume of velocity space

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Fig. 12. Angular distribution of neutrons per fission per unit volume of velocity space. a- Average velocity of light fragment (1.37×10^9 cm/sec) b- Average velocity of heavy fragment (1.04×10^9 cm/sec) V_L = velocity of neutrons.

axis to the left (light) and right of the origin (heavy). It can be seen that the neutron energy and angular distribution in the laboratory system are strongly influenced by the velocity contribution of the fragments.² It can also be seen that the probability of the emission of neutrons decreases as the velocity increases. Analysis of the results also shows that the gross angular and velocity distributions of the neutrons can be accounted for by isotropic evaporation from the fragments but there is a suggestion of a deviation from this in low-energy neutrons at 90°.

The results show that the method is capable of giving a complete description of the angular and velocity distributions of the absolute numbers of neutrons emitted in the fission process. This experiment is regarded as preliminary, and preparations are being made for a more refined series of measurements which will have the following advantages over the previous work:

- (a) A stronger fission source,
- (b) Measurement of the velocities of both fragments over a longer flight path,
- (c) Measurements of neutron velocities at more angles.
- (d) Measurement of lower-energy neutrons.

² It may be helpful to realize that the distribution represented in one plane in Fig. 12 is better visualized in three dimensions as that of a prolate spheroid. Thus the deviation from a spherical shape is a consequence of the velocity component contributed by the fragments to the neutron velocities.

THE STOPPING OF HEAVY ATOMS

Lester Winsberg and John M. Alexander

The determination of the range of atomic particles in matter is of interest both as regards stopping theory^{1,2} and in its application to other problems, such as the study of nuclear reactions (see pp.52-60). With the advent of heavy-ion accelerators it has become possible to pursue the investigation of heavy-particle stopping with increased vigor.

We have chosen two reactions that are convenient in this type of work: the production of astatine isotopes from gold, and of terbium-149 from praseodymium by carbon-12 bombardment. These reactions probably proceed via a compound-nucleus mechanism. It is therefore assumed that the entire momentum of the C^{12} ion is transferred to the compound nucleus. The energy of the recoiling nucleus can therefore be readily calculated. Some uncertainty is present because several different charge states of carbon are often accelerated. The recoil energies were calculated from the average energy of the beam.

In the experiments performed at the Berkeley machine, sets of foils of various thicknesses, such as are shown in Fig. 13, were stacked together and exposed to the C^{12} beam. The target foils for experiments No. 1 and No. 2 were prepared by evaporating gold on aluminum, and for experiment No. 3 by electroplating praseodymium hydroxide on nickel. The alpha activities of the foils were directly measured by alpha-particle detectors and corrected for absorption. In experiment No. 1 the gold was separated from its backing and each foil was counted separately. In succeeding experiments the targets and backing foils were counted together.

The results from each set of foils in the three experiments were analyzed in order to determine the mean range of the recoiling alpha-particle emitters. In addition the straggling parameter in the distribution of ranges about the mean range was determined in Experiments 1 and 3. The analysis of Experiment 3 was straightforward, since the target and the catcher foils were thin and the recoil alpha-particle emitters were expected to be spread through the aluminum foils in a Gaussian distribution about the mean range.

In the first two experiments the target foils were thicker than the mean range. It was therefore necessary to develop a mathematical treatment of the results from these experiments.

If the range straggling is neglected the mean range in the target material is simply the product of the target thickness, T , and F_c , the alpha activity found in the forward catcher foil divided by the total alpha activity that had been formed,

$$R_0 = T \cdot F_c \quad (1)$$

¹N. Bohr, Kgl. Danske Videnskab Selskab. Mat.-fys. Medd. 18, No. 8 (1948).

²J. Lindhard and M. Scharff (as quoted by Leachman and Atterling⁴).

Fig. 13. The arrangement of foils in experiments at the Berkeley heavy-ion linear accelerator.

The characteristics of the foils were as follows:

Experiment No.	Foils	Element	Thickness of foil used (mg/cm ²)
1	1, 4	Al	2.4
	2, 3 ^a	Au	0.5-0.6
2	1	Al	1.8
	2 ^a	Au	0.6-1.5
	3	Al	1.9
3	1	Ni	2.2-2.4
	2 ^a	Pr	0.04-.06
	3-10	Al	0.09-.17

^aThese foils consisted of the target attached directly to the backing foil (see text).

If straggling is not neglected the distribution of activities from a thin foil is given as a Gaussian function,^{1,2}

$$P(R) = \frac{1}{K\sqrt{\pi}} \exp \left[- \left(\frac{R - R_0}{K} \right)^2 \right], \quad (2)$$

where $K = \sqrt{2} R_0 \rho$, ρ being the straggling parameter.

Let us consider the target foil to consist of two parts; the first, into which the beam enters, has a thickness t , and the second, a thickness $T-t$. This is the case for foils No. 2 and No. 3 in Experiment 1 (Fig. 13).

The fraction of the total activity left in the first foil is

$$F_t = \frac{1}{KT\sqrt{\pi}} \int_0^T \int_0^t \exp \left[- \left(\frac{r-s-R_0}{K} \right)^2 \right] dr ds \quad (3)$$

or

$$F_t = \frac{K}{2T} \sum_{L_1=L_1}^{L_4} (-1)^{i+1} \int_0^{L_i} I(x) dx, \quad (4)$$

where s is the point at which the recoiling atom originates and $r-s$ is the distance it travels in the forward direction.

The probability integral is $I(x) = \frac{2}{\sqrt{\pi}} \int_0^x \exp(-\mu^2) d\mu$, and

$$L_1 = \frac{R_0 - t}{K}, \quad (5)$$

$$L_2 = \frac{T + R_0 - t}{K}, \quad (6)$$

$$L_3 = \frac{T + R_0}{K}, \quad (7)$$

$$L_4 = \frac{R_0}{K}. \quad (8)$$

Equation (4) reduces to

$$F_t = \frac{K}{2T} \sum_{L_i=L_1}^{L_4} (-1)^{i+1} \left[\frac{\exp(-L_i^2)}{\sqrt{\pi}} + L_i I(L_i) \right] \quad (9)$$

The fraction of the total activity left in both target foils, F_t , can be obtained if t is replaced by T in Eqs. (5) and (6).

Since L_2 and L_3 are large, Eq. (9) can be simplified to

$$F_t = \frac{K}{T} \left[F\left(\frac{R_0 - t}{K}\right) - F\left(\frac{R_0}{K}\right) \right], \quad (10)$$

where

$$F(y) = \frac{1}{2\sqrt{\pi}} e^{-y^2} - y/2 \left[1 - I(y) \right]. \quad (11)$$

Here $F\left(\frac{R_0}{K}\right)$ is small and may be neglected. The values of $I(y)$ can be found in tables.³

The values of R_0 and K in the first experiment were obtained by successive approximations until the experimental and calculated values of F_t and F_T agreed. The three values of K/R_0 were plotted as a function of recoil energy. The values for K for the second experiment were obtained from this curve. The values of R_0 were then calculated from the experimentally determined values of F_T by use of Eq. (10). The results of both experiments are given in Table I, and the values of R_0 are plotted as a function of recoil energy in Fig. 14. Also shown in Fig. 14 is the range-energy curve for astatine atoms stopping in aluminum.⁴

The recoil properties of Tb^{149} from C^{12} bombardment of praseodymium (Experiment 3 in Fig. 13) were obtained by fitting the distribution of Tb^{149} alpha activity in the aluminum catcher foils to Eq. (2). This was done by the procedure described by Leachman and Atterling.⁴ The results are listed in Table I. The values of R_0 and K are only approximate values, since the thicknesses of the aluminum foils used to obtain the 3.79-Mev result are yet to be determined, and, for the other two energies, were not measured directly. In the latter cases aluminum leaf, which proved to be nonuniform, was used. It was only after the foils had been discarded that the weights, which had been determined on a different part of the leaf, were found to be unreliable. However, the values of ρ are probably reliable. The ranges will be determined more carefully later. The range values at 4.46 and 4.80 Mev are shown in Fig. 18 (p. 54).

³B. O. Peirce and R. M. Foster, A Short Table of Integrals (Ginn and Co., 1956).

⁴R. B. Leachman and H. Atterling, Arkiv Fysik 13, No. 9 (1957).

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Fig. 14. Range versus energy for astatine atoms in gold and aluminum.

Table I

Ranges and straggling parameters for the stopping of heavy atoms.

Experiment No.	Target; → Recoil	E_{recoil} (Mev)	R_0 (mg/cm ²)	R_0/E (mg/cm ² Mev)	K (mg/cm ²)	ρ
1	Au ¹⁹⁷	3.68	0.459	0.125	0.279	0.430
	→ At ²⁰²⁻²⁰⁵	5.23	0.640	0.122	0.399	0.441
		6.67	0.824	0.123	0.537	0.461
2	Au ¹⁹⁷	2.90	0.382	0.132		
	→ At ²⁰²⁻²⁰⁷	3.93	0.503	0.128		
		4.73	0.592	0.125		
		5.41	0.692	0.128		
		6.01	0.760	0.126		
		6.58	0.834	0.127		
	Average			0.126		0.444
	Theoretical			0.213		0.408
3	Pr ¹⁴¹	3.79	0.39 ?		0.16 ?	0.288
	→ Tb ¹⁴⁹	4.46	0.37±.04	0.082	0.13±.01	0.256
		4.80	0.48±.05	0.101	0.21±.02	0.300
	Average			0.091		0.281
	Theoretical			0.105		0.294

For the recoil energies studied here, the range is predicted to be proportional to the energy.^{1,2} This is found to be the case for the astatine isotopes in Fig. 14 and is not inconsistent with the Tb^{149} results.

The predicted ratio² of range to energy is

$$\frac{R_0}{E} = 0.600 \frac{m(m+M)}{M} \frac{\sqrt{z^{2/3} Z^{2/3}}}{z Z} \frac{mg}{\text{cm}^2 \text{Mev}}, \quad (12)$$

where m and M , z and Z are respectively the atomic masses and atomic numbers of the stopping material and the recoiling atoms. The predicted value² of the straggling parameter is

$$\rho = \left[\frac{2 m M}{3(m+M)^2} \right]^{1/2} \quad (13)$$

The predictions of the theory for ρ are in rough agreement with the experimentally determined values in Table I. Leachman and Atterling find a similar agreement for At^{203} and At^{205} stopping in aluminum.

The theoretical value of R_0/E is in rather good agreement with the experimental value for Tb^{149} stopping in aluminum, but not for astatine atoms stopping in gold. This result is not unexpected, since the theoretical R_0 is the total path length of the recoiling atom, whereas the experimental value is its projection along the beam direction. The path of heavy atoms is expected to be more nearly straight in a light material (aluminum) than in a heavy material (gold).

We are indebted to Mr. Daniel O'Connell and Mr. Arthur Johns for preparing the special foils used in our experiments.

RECOIL PROPERTIES OF SPALLATION PRODUCTS

John M. Alexander and Lester Winsberg

The compound-nucleus model of nuclear reactions has been shown to give predictions that agree with experiment in very many cases. The model is expected to predict correctly most of the nuclear reactions that occur between nucleons and nuclei at low energy. As the kinetic energy of the incident nucleon is increased the utility of the model is expected to decrease. Experiments have shown this to be the case. In reactions between nuclei and complex incident particles the limits of applicability of the compound-nucleus picture are not clear. Several specific reactions (such as the d, p stripping reaction) have been shown to proceed primarily by non-compound-nucleus interactions. However, it is difficult to predict the relative importance of these reactions compared with compound-nucleus processes. The purpose of this experimental program is to obtain information on the contributions of non-compound-nucleus processes to the formation of spallation products as a function of the energy and mass of the incident particle. Similar studies of the fission products are also in progress. These studies were pursued at the Berkeley heavy-ion linear accelerator, which delivers particles of masses up to 40 with energies of 10.5 Mev per nucleon.

The technique used was the measurement of recoil properties as described elsewhere (see pp. 35-41). At this time we can report the determination of the average recoil energies of spallation products produced by the bombardment of Bi^{209} with Ne^{20} .

This system is interesting because it is known that the fission reaction takes place with high probability, and mainly by the compound-nucleus mechanism (see pp. 25-26). A study of the spallation products is thus rather sensitive to the presence of non-compound-nucleus mechanisms.

Targets consisted of bismuth metal evaporated onto 0.00025-inch aluminum foils arranged as shown in Fig. 15.

Fig. 15. Arrangement of target foils.

The stack was exposed to the neon beam at very low intensity (average beam 0.05 μ amp) in order to avoid thermal diffusion of the products. After the irradiation the alpha activity of each foil was observed over a period of several days. The decay curves revealed many half periods from ~ 20 minutes to several days from a complex mixture of polonium, astatine, and emanation species. Even though no precise statement can be made about the particular species observed, the following interesting observations were made.

1. Less than 0.5% of the activity produced recoiled in the direction opposite to the beam (as determined from the ratio of Foil 1 to Foil 2).
2. The ratio of the alpha activity of each target to the adjacent recoil catcher was constant to $\sim 2\%$ as a function of time.
3. The ratio of catcher activity to target activity increased as the energy of the beam was degraded.

The first observation simply verifies the expectation that the recoil momentum gained by the decay of some intermediate excited nucleus is less than that imparted by the incident ion.

The second observation can be interpreted in two ways: (a) the spectrum of momentum transfers is rather narrow, or (b) the complicated decay chains of electron capture and alpha decay that lead from the primary products to the observed products give the result that each species observed is the product of the same average initial interaction.

The third point indicates that we have indeed observed non-compound-nucleus processes and that the probability of these processes increases with increasing bombarding energy.

In Table II the results of the experiment are recorded. The average ranges (column 3) were calculated from the fraction of the total alpha activity found in the forward catcher, according to Eq. (1), p. 35. The average recoil energies were determined by use of the range-energy data of the previous report corrected to that for bismuth by Eq. (12), p. 41.

Table II

Recoil range measurements of spallation products from the reaction $\text{Bi}^{209} + \text{Ne}^{20}$					
1 Target foil number	2 Neon energy (Mev)	3 Average range of alpha-emitting products (mg/cm ²)	4 Average energy of alpha-emitting products (Mev)	5 Kinetic energy of center of mass (Mev)	6 Ratio of Col. 4 to Col. 5
3	177	$0.44 \pm .01$	3.4	15.45	0.22
6	154	$0.49 \pm .00$	3.8	13.45	0.28
8	132	$0.62 \pm .01$	4.8	10.5	0.46
10	111	$0.75 \pm .01$	5.8	9.7	0.60

THE MATHEMATICAL TREATMENT OF DATA FROM RECOIL EXPERIMENTS

Lester Winsberg

The study of recoil phenomena has provided useful information concerning the mechanism of nuclear reactions.¹⁻³ The mathematical treatment of these experiments has been given for special cases. It is the purpose of this article to present a general treatment of the subject in order to facilitate this type of investigation. The treatment follows that given by Porile and Sugarman.¹

For the purpose of the analysis, it is assumed that the nuclear reaction proceeds in two stages. In the first stage, the incoming particle imparts a velocity v in the laboratory system along the direction of the incoming particle to some intermediate nucleus. The final results are only slightly affected by a perpendicular component, $v_{\perp}$.¹ In the second stage, the intermediate nucleus breaks up to form the final nuclear fragment with a velocity V in the system of the moving intermediate nucleus. The vector V may be at any angle θ with respect to v . It is convenient to introduce $\eta = v/V$ and θ_L , the angle in the laboratory system at which the final fragment is moving with respect to the direction of the incoming particle. The relation between these quantities is shown in Fig. 16, on the assumption that $v + V$ is nonrelativistic. (This is the case for most recoil studies.)

The distance R_L that an atom of velocity $\vec{v} + \vec{V}$ travels in matter depends on the relation between its range and its velocity. For most cases studied until now by the recoil technique, the range is given by

$$R = k V^N, \quad (1)$$

where $N \approx 1$ for fission fragments and $N = 2$ for heavy atoms moving with smaller velocities.¹⁻⁴

Therefore we have

$$R_L = R (1 + \eta^2 + 2 \eta \cos \theta)^{N/2}, \quad (2)$$

where R is the range in the stopping material of an atom of velocity V .

¹ N. T. Porile and N. Sugarman, Phys. Rev. 107, 1410 (1957); with corrections in Phys. Rev. 111, 1746 (1958). Other pertinent articles are referred to here.

² John M. Alexander in Chemistry Division Semiannual Report December 1957 through May 1958, UCRL-8369, July 14, 1958, p. 13.

³ Lester Winsberg, The Production of Terbium-149 etc., this report, p. 52.

⁴ Lester Winsberg and John M. Alexander, The Stopping of Heavy Atoms, this report, p. 35.

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Fig. 16. This diagram shows the relationship between the various angles and vectors for a nuclear reaction. If all lengths are multiplied by V , the more familiar velocity-vector diagram results.

The projection of R_L in the direction of the beam, t , is given by

$$t = R(\eta + \cos \theta)(1 + \eta^2 + 2\eta \cos \theta)^{(N-1)/2} \quad (3)$$

The maximum thickness of material the atom can traverse in the forward (+) or backward (-) direction is $R(1 \pm \eta)^N$. Thus Eq. (3) relates the variables involved in a recoil experiment in which a target foil of thickness t is oriented perpendicular to the incoming beam. We denote by F and B the fraction of the atoms, produced in a nuclear reaction, that recoil out of the target in the forward and backward directions respectively.

The projection of R_L on a given direction perpendicular to that of the beam is given by

$$t = R \sin \theta (1 + \eta^2 + 2\eta \cos \theta)^{(N-1)/2} \cos \phi, \quad (4)$$

where ϕ is the angle between the given direction and that corresponding to $\sin \theta$ in Fig. 16. The maximum thickness of material the atom can traverse in the perpendicular direction is

$$R \left\{ 1 + \frac{N(N-1)}{2} \eta^2 \right\}.$$

Terms in η to the fourth and higher powers have not been included. For $N = 1$, the maximum thickness is exactly equal to R . Equation (4) relates the variables if the target foil is placed parallel to the beam. The fraction of atoms that recoil out of one face of the target is denoted by P .

We first consider cases in which the scattering is isotropic in the system of the moving intermediate nucleus for various thicknesses W of the target foil. In every case B can be obtained by changing the sign of η in the expression for F . The cases for $N = 1$ have been worked out before,¹ but are included for completeness. For all cases considered here $\eta \leq 1$.

Case 1. $W = 0$:

$$F = 1/2 \int_0^{\arccos(-\eta)} \sin \theta d\theta = \frac{1+\eta}{2}, \quad (5)$$

$$B = \frac{1-\eta}{2}, \quad (6)$$

$$\eta = F - B. \quad (7)$$

Of course, $P = 1/2$ for each of the two perpendicular directions.

Case 2A. $W > R(1+\eta)^N$:

$$F = 1/2 \cdot W \int_0^{\arccos(-\eta)} \sin \theta d\theta \int_0^{R(\eta + \cos \theta)(1 + \eta^2 + 2\eta \cos \theta)^{(N-1)/2}} dt \quad (8)$$

For $N = 1$,

$$F = \frac{R}{4W} (1+\eta)^2, \quad (9)$$

$$R = \frac{W (F-B)}{\eta}. \quad (10)$$

For $N = 2$,

$$F = \frac{4R(1+\eta)^2}{15W} \left\{ 1 + \frac{\eta}{2} - \frac{(1-\eta)^2 (1 - \sqrt{1-\eta^2})}{8\eta^2} \right\} \quad (11)$$

$$R = \frac{15 W (F-B)}{4 \eta (5+\eta^2)} \quad (12)$$

Case 2B. $W > R \left\{ 1 + \frac{N(N-1)}{2} \eta^2 \right\}$:

$$P = \frac{1}{2\pi W} \int_0^\pi \sin \theta d\theta \int_0^{\frac{R \sin \theta (1+\eta^2 + 2\eta \cos \theta)^{(N-1)/2}}{dt}} dt \int_0^\phi d\phi, \quad (13)$$

where ϕ is given by Eq. (4).

$$\text{For } N = 1, \quad P = R/4 W. \quad (14)$$

$$\text{For } N = 2 \quad P = \frac{R}{4W} (1 + 3/8 \eta^2). \quad (15^*)$$

Case 3A. $0 < W < R (1 + \eta)^N$:

$$F = \frac{1}{2} W \int_0^W dt \int_0^\theta \sin \theta d\theta, \quad (16)$$

where θ is given by Eq. (3).

* Not exact; terms containing the fourth and higher powers of η have been omitted.

For $N = 1$,

$$F = \frac{1+\eta}{2} - \frac{W}{4R} \quad (17)$$

For $N = 2$,

$$F = \frac{1+\eta}{2} - \frac{W}{4R\sqrt{1-\eta^2}} + \frac{1}{6} \frac{\eta (W/R)^2}{(1-\eta^2)^2} + \dots \quad (18^\dagger)$$

Case 3B. $0 < W < R$ $\{1 + \frac{N(N-1)}{2} \eta^2\}$:

$$P = \frac{1}{2\pi W} \int_0^W dt \int_{\theta_2}^{\pi-\theta_2} \sin \theta \, d\theta \int_0^\phi d\phi, \quad (19)$$

where θ_2 is given by

$$t = R \sin \theta_2 (1 + \eta^2 + 2\eta \cos \theta_2)^{(N-1)/2} \quad (20)$$

For $N = 1$

$$P = 1/2 \left(1 - \frac{W}{2R}\right). \quad (21)$$

For $N = 2$

$$P = 1/2 \left\{1 - 1/2 W/R - 1/8 W/R \eta^2 + 5/16 \frac{W^2 \eta^2}{R^2} \dots\right\} \quad (22^\dagger)$$

If the scattering in the system of the intermediate nucleus is not isotropic but has the form $a + b \cos^2 \theta$, we get for a thin target,

[†]The terms indicated are usually sufficient for the accuracy of most recoil experiments. The exact solution is not included because of lack of space.

Case 4. $W = 0$:

$$F = 1/2 \left\{ 1 + \frac{1 + b \eta^2/3a}{1 + b/3a} \eta \right\}. \quad (23)$$

Porile and Sugarman consider the anisotropic case for $N = 1$ for a thick target.¹

A special case of current interest in the study of fission is for $R = k V - \Delta$ with the angular distribution of the form $a + b \cos^2 \theta$.

Case 5A. $W > R (1 + \eta) + \Delta \eta$; $R = k V - \Delta$:

$$F = \frac{R(1+\eta)^2}{4W} \left\{ 1 + f_1(\eta) \frac{\Delta}{R} + (1-\eta)^2 \beta + f_2(\eta) \beta \frac{\Delta}{R} \right\}. \quad (24)$$

$$f_1(\eta) = \frac{2\eta}{3(1+\eta)^2} \left[1 + \eta \left\{ 1 + \frac{(1-\eta^2)(1-\eta^2/2 - \sqrt{1-\eta^2})}{\eta^4} \right\} \right], \quad (25)$$

$$f_2(\eta) \approx \frac{8\eta}{3(1+\eta)^2} (1 + 6/5 \eta + 9/35 \eta^2), \quad (26^*)$$

$$\beta = \frac{b/6a}{1 + b/3a}. \quad (27)$$

Case 5B. $W > R + \Delta \left\{ 1 - \frac{1}{\sqrt{1+\eta^2}} \right\}$; $R = k V - \Delta$:

$$P = \frac{R}{4W} \left\{ 1 - f_3(\eta) \frac{\Delta}{R} - \frac{1}{2} \beta - f_4(\eta) \beta \frac{\Delta}{R} \right\}, \quad (28)$$

$$f_3(\eta) = 1/4 \eta^2 \left\{ 1 - \eta^2/3 - \frac{8(1+\eta^2)(1+\eta^2/2 - \eta^4/8 - \sqrt{1+\eta^2})}{3\eta^4} \right\}; \quad (29)$$

$f_4(\eta)$ has not been determined since it involves an expression which cannot be integrated analytically. However, $f_4 \beta (\Delta/R)$ is usually small because the largest term in f_4 is of the order η^2 , and $\beta (\Delta/R) \ll 1$.

Lack of space prevents the presentation of other cases.

In general, a specific value of R and of η need not be associated with a given species of recoiling atoms. The values of F , B , and P given by the above equations will then be average values resulting from processes involving various values of R and η .

Thus for a thin-target experiment, i.e., Case 1, Eq. (7) and Case 3A, Eq. (17), we would actually get

$$\langle \eta \rangle = F - B, \quad (30)$$

and if we add F and B in Eq. (17) we get (for $N = 1$)

$$F + B = 1 - W/2 \langle 1/R \rangle, \quad (31)$$

or instead of a unique value of R , the quantity

$$\langle 1/R \rangle^{-1} = W/2 (1 - F - B). \quad (32)$$

For a thick-target experiment, Case 2A, with $N = 1$ we get, from Eq. (9),

$$\langle R \eta \rangle = W (F - B) \quad (33)$$

and

$$\langle R (1 + \eta^2) \rangle = 2 W (F + B). \quad (34)$$

Thus, instead of determining a unique value for η and R from Eqs. (9) and (10), we actually get

$$\frac{\langle R \eta \rangle}{\langle R \rangle} \quad \text{and} \quad \frac{\langle R (1 + \eta^2) \rangle}{\langle 1 + \eta^2 \rangle}$$

respectively. If η^2 is small relative to unity, which is the case for most recoil experiments that have been performed, then we have

$$\frac{\langle R (1 + \eta^2) \rangle}{\langle 1 + \eta^2 \rangle} \approx \langle R \rangle. \quad (35)$$

Similar results are obtained for range-velocity relations other than $R = k V$.

Equations (30) to (35) are summarized in Table III.

If there is a unique value for R , the values obtained from both types of experiments will be the same. The application of Table III is illustrated on pp. 52.-60.

Table III

Quantities measured in recoil experiments performed with thin and with thick targets			
Target thickness		Quantities measured	
$W \rightarrow 0$		$\langle 1/R \rangle$	$\langle \eta \rangle$
$W > W_{\max}$		$\langle R \rangle$	$\langle R \eta \rangle$

The thickness, $W_{\max}$, depends on the range-velocity relation and is given in the first part of this article.

As can be seen from Eqs. (5) and (30), the value of $\langle \eta \rangle$ does not depend on the range-velocity relation if the target is sufficiently thin. This is not true if the target is thick. Similarly, the value of $\langle R \rangle$ obtained from a thick-target experiment depends only slightly on the range-velocity relation, provided η^2 is small relative to unity.

In order to determine the spectrum of R and η values, an experiment can be performed with a thin target ($W \rightarrow 0$) sandwiched between stacked foils of some suitable material. It is, of course, necessary that the product being investigated cannot be formed directly in the catcher foils.

The fraction $F(t)$ of all the atoms produced that go beyond a thickness t in the forward direction is given by

$$F(t) = 1/2 \int_0^\theta \sin \theta \, d\theta = \frac{1 - \cos \theta}{2}, \quad (36)$$

where $\cos \theta$ is given by Eq. (3):

For $N = 1$,

$$F(t) = (1 + \eta - t/R)/2. \quad (37)$$

For other values of N , Eq. (37) will be the first term of a series expansion. A similar expression is obtained for $B(t)$ but with the sign of η changed. Equation (37) is for unique values of R and η . For a spectrum of R and η values we get

$$F(t) = 1/2 \sum_i p_i \left(1 + \eta_i - \frac{t}{R_i} \right), \quad (38)$$

where p_i is fraction of the atoms having the values η_i and R_i . The summation is restricted to values of i for which $t/R_i = \eta_i < 1$.

For $N = 1$ the analysis of this type of experiment is straightforward because a plot of $F(t)$ or $B(t)$ versus t is a sum of straight lines, each one corresponding to a given value of η and R . The analysis of other cases is much more complicated.

THE PRODUCTION OF TERBIUM-149 FROM THE BOMBARDMENT OF TANTALUM, GOLD, AND BISMUTH BY HIGH-ENERGY PROTONS

Lester Winsberg

The production of Tb^{149} in tantalum, gold, and bismuth by protons with energies greater than 0.4 Bev was measured by direct counting of the alpha activity in the target foils and in adjacent aluminum foils. The latter were employed to catch any products that recoiled out of the target. The beam intensity was monitored from the amount of Na^{24} produced in an aluminum foil included for this purpose. Previously, the excitation functions of the alpha activity of Tb^{149} had been determined to 3.0 Bev.¹ These were extended to 6.2 Bev. In order to determine the total cross section for the formation of Tb^{149} , the alpha-branching ratio was measured and found to be 0.10 ± 0.02 (see page 67). These cross sections, determined at the Bevatron and the 184-inch synchrocyclotron, are shown in Fig. 17. Extension of the curve below 1 Bev for tantalum was made with the aid of the Brookhaven data, but it will be remeasured directly here at a future date. The cross section to produce the monitor Na^{24} was taken to be 10.5 mb in the entire energy region studied. There is some evidence that there is actually a dip in the excitation function of Na^{24} at 3 Bev.² This indicates that the secondary hump in the Tb^{149} excitation functions at this energy may not be real.

The fraction of Tb^{149} recoiling out of the target both in the forward and backward directions was also determined to provide further evidence on the mechanism of the reaction. The analysis follows the general procedure given elsewhere (pp. 44-52). In order to interpret the results it is necessary to know how the range of Tb^{149} in the target materials varies as a function of energy. This relationship has been experimentally determined for $\text{At}^{203-207}$ with kinetic energies between 4 and 7 Mev stopping in gold (pp. 35-41). In agreement with theory, it is found that the range is proportional to energy. This is also the case for astatine nuclides stopping in aluminum. By means of the theory these results were used (pp. 35-41) to calculate the range-energy relation for Tb^{149} stopping in aluminum, tantalum, gold, and bismuth (Fig. 18). A direct experiment on Tb^{149} (Fig. 18) confirmed the aluminum curve (Fig. 14) and is reassurance that the other curves are probably correct.

The mathematical treatment of the recoil results has been developed for this range-energy relation; see pp. 44-52. The definitions of quantities discussed below are given in the same article.

¹Duffield, Friedlander, and Miller (Brookhaven National Laboratory) (unpublished).

²D. W. Barr, Nuclear Reactions of Copper Induced by 5.7-Bev Protons, UCRL-3793, May 1957, Figure 3.

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Fig. 17. The cross section for the formation of Tb^{149} as a function of the proton energy. The secondary hump at approximately 3 Bev is probably not real. See text.

Fig. 18. Range versus energy for Tb^{149} in various stopping materials. The two circled points on the aluminum curve were determined directly (see pp.35-41).

In Fig. 19 are given values of R as a function of energy for thick and for thin targets. Experiments in which the proton beam enters perpendicular to the stack of foils give values of R that are in agreement within experimental error with those obtained in experiments in which the beam entered parallel to the foil stack. This indicates either (a) that η_1 and any anisotropy in the scattering of the Tb^{149} in the moving frame of reference are both too small to be detected by this type of measurement, or (b) that the effects due to η_1 and the anisotropy cancel each other. The equations for the isotropic case therefore have been used in the treatment of the experimental results.

From Fig. 19 we see that the experimentally determined value of R depends on the thickness of the target. It is therefore apparent that R does not have a unique value. A similar result is obtained for η (Fig. 20). In these cases, the quantities that are actually measured are approximately as given in Table III. Thus we get $\sim \langle R \rangle$ from a thick-target experiment and $\sim \langle 1/R \rangle^{-1}$ from a thin-target experiment.

In order to determine the spread in values of R , thin targets (0.18 mg/cm^2) of gold sandwiched between stacks of thin aluminum foils were exposed to beams of 1.7- and 4.5-Bev protons. The results of these experiments are presented in Fig. 21 as the fraction of Tb^{149} stopping in 0.1 mg Al/cm^2 for a given thickness of aluminum penetrated.

An approximate analysis of the data at 1.7 Bev was made to obtain the spectrum of R values. In Fig. 22 is given the probability $P(R)$ of getting a given value R , divided by R , as a function of R . From this we obtain the approximate relation

$$P(R)dR = \frac{R \exp(-R/t)}{t^2} dR \quad (1)$$

$$\text{where } t = \frac{0.155}{0.693} = 0.224 \frac{\text{mg Al}}{\text{cm}^2}.$$

Similar results are obtained from the analysis of the 4.5-Bev data.

It is now possible by means of Eq. (1) to calculate the values of R expected from thick- and thin-target experiments. For the thick-target experiments we get

$$\langle R \rangle = \int_0^\infty R P(R) dR = 2t \quad (2)$$

For the thin-target experiments we get

$$\langle \frac{1}{R} \rangle^{-1} = \left\{ \int_0^\infty \frac{1}{R} P(R) dR \right\}^{-1} = t, \quad (3)$$

$$\therefore \frac{\text{value for thick target}}{\text{value for thin target}} \approx \frac{\langle R \rangle}{\langle \frac{1}{R} \rangle^{-1}} = 2. \quad (4)$$

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Fig. 19. Experimentally measured values of R (pp.44-52) as a function of the proton energy. Points marked R_p were determined from a perpendicular-type experiment. The tantalum foils were either 8.4 or 10.3 mg/cm^2 thick. The unmarked and bismuth points were determined with foils 1.5 to 2.5 mg/cm^2 thick. The thicknesses of the other foils were (a) 3.2 and 3.7 $\text{mg Au}/\text{cm}^2$, (b) 4.2 $\text{mg Bi}/\text{cm}^2$, (c) 0.18 $\text{mg Au}/\text{cm}^2$, (d) 0.085 $\text{mg Bi}/\text{cm}^2$.

Fig. 20. Experimentally measured values of η (pp. 44-52) as a function of the proton energy. The unmarked points are for the thickest targets. When slightly thinner targets were used, the values were corrected to infinitely-thick-target values by an adjustment of a few percent. The thicknesses of the other foils were (a) 0.18 mg Au/cm², (b) 0.085 mg Bi/cm².

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Fig. 21. The fraction of Tb^{149} stopping in 0.1 mg Al/cm^2 as a function of the thickness of aluminum penetrated. The overall results of these experiments were:

E_p	1.7 Bev (●)	4.5 Bev (◻)
Thickness of Al (mg/cm^2)	0.182	0.183
Fraction in back catchers	0.1748	0.2484
Fraction in target	0.1301	0.1451
Fraction in front catchers	0.6951	0.6065

Fig. 22. The probability $P(R)$ for a given value of R , divided by R , as a function of R .

The ratio is in agreement with the values for bismuth at 1.7 and 4.5 Bev (Fig. 19). The thickness of the thin target was 0.085 mg Bi/cm^2 ; of the thick targets, 2 to 4 mg Bi/cm^2 . Apparently, the gold targets were not thin enough to satisfy this test.

At 1.7 Bev, $2T = 0.447 \text{ mg Al/cm}^2$. This corresponds to an energy of 5.0 Mev. From the data directly (Fig. 22) we get 4.6 Mev for the average recoil energy. From thick-target experiments at 1.0, 1.7, and 2.0 Bev we get a value of 0.94 mg Au/cm^2 for the range. This corresponds to an energy of 5.0 Mev. This agreement between the two types of experiment is well within the experimental errors and the uncertainty of the range-energy curves.

Since range is proportional to energy, we also obtain the relation

$$P(E)dE = \frac{E}{T^2} \exp(-E/T) dE, \quad (5)$$

where $T \approx 2.5 \text{ Mev}$ and 2.6 Mev for gold and bismuth targets respectively. The values of thick-target ranges in Fig. 19 indicate that T decreases but little in value from 1 to 6 Bev.

Since only thick-target experiments were performed in the case of tantalum we have no direct evidence that Eq. (5) is valid for tantalum. If it is, the T value for tantalum is $\approx 1.7 \text{ Mev}$, from the range values in Fig. 19.

At present I do not wish to go into any detailed consideration of these results in terms of the mechanisms that lead to the production of Tb^{149} from bombardment by high-energy particles. More experiments are contemplated with protons and other projectiles. Speculation will be on a more reliable footing when these experiments are completed and when the spectrum of η values has been calculated. However, it is possible to conclude that Tb^{149} is not produced, in the main, by a fission mechanism. Were it a fission product, it would have a unique recoil range corresponding to a kinetic energy of the order of 15 Mev. This is clearly not the case.

NUCLEAR REACTIONS INDUCED BY He^3 IONS

Samuel S. Markowitz and Jane M. Hall

Beams of He^3 ions have recently been accelerated at the Crocker 60-inch cyclotron to a maximum energy of 17.7 Mev and at the Hilac to a maximum energy of 31.2 Mev. Using dilute He^3 in He^4 gas mixtures (about 4% He^3), we obtained an internal beam intensity of 1 microampere of doubly charged He^3 at the cyclotron and about 0.01 microamperes of singly charged He^3 at the Hilac. The shape of the magnetic field as a function of radius in the cyclotron (as it now operates without special "shim" devices) prevents acceleration of the He^3 ions to an energy greater than 17.7 Mev. Because of the higher energy and the convenient beam collimation into the center of a target stack (rather than into the leading edge as in the cyclotron) the Hilac was used as a source of 31.2-Mev He^3 ions; lower energies were obtained by degrading the beam in absorbers. In some of the Hilac experiments, 15% He^3 in He^4 was used for a short time; the beam intensity increased in rough proportion to the concentration of He^3 , allowance being made for the variations in operating conditions of the accelerator from day to day.

He^3 is a very interesting nucleus with respect to its participation in nuclear reactions as an incident projectile. The low binding energy of 7.7 Mev (compared with 28.2 Mev for a He^4 nucleus) causes many simple nuclear reactions to be exoergic; compound nuclei if formed with He^3 can be relatively highly excited with a low-energy incident particle. The ability of the He^3 to "pick up" a neutron from a target nucleus to form He^4 and its probability for "stripping" in various ways, should lead to a better understanding of "direct interactions" and the binding of nucleons at the surfaces of target nuclei. The purpose of our research in this relatively new and unexplored field is then apparent.

Important applications of He^3 -induced nuclear reactions are, in addition to the above,

(a) production of neutron-deficient isotopes in good yield with low-energy particles e. g., via $(\text{He}^3, 3n)$ and $(\text{He}^3, 4n)$ reactions; and

(b) as an aid in determination of the mass number of a new isotope by cross-bombardment technique, especially when yields are compared with those for He^4 bombardment.

Excitation functions have been measured by stacked-foil experiments for the following reactions (calculated "Q" values from mass data of Wapstra are shown¹):

¹A. H. Wapstra, *Physica* 21, 367 (1955).

Reaction	Q (in Mev)
$\text{Al}^{27}(\text{He}^3, 2\text{He}^4)\text{Na}^{22}$	- 1.9
$\text{Al}^{27}(\text{He}^3, 3\text{He}^4)\text{F}^{18}$	-10.4
$\text{Al}^{27}(\text{He}^3, \text{He}^4_2\text{p})\text{Na}^{24}$	-10.9
$\text{C}^{12}(\text{He}^3, \text{He}^4)\text{C}^{11}$	+ 1.9
$\text{C}^{12}(\text{He}^3, \text{d} \& \text{np})\text{N}^{13}$	- 3.6 and -5.9
$\text{Be}^9(\text{He}^3, \text{n})\text{C}^{11}$	+ 7.7
a $\left\{ \begin{array}{l} \text{O}^{16}(\text{He}^3, \text{p})\text{F}^{18} \\ \text{O}^{16}(\text{He}^3, \text{n})\text{Ne}^{18} \xrightarrow[1.6 \text{ sec}]{\beta^+} \text{F}^{18} \end{array} \right.$	+ 2.0 - 2.9

^a Cumulative yield from (He^3, p) and (He^3, n) reactions measured.

The excitation functions are shown in Figs. 23, 24, and 25. The energy scale near 0 Mev, computed from the range-vs-energy curves of Rich and Madey (UCRL-2301) is in error. This is probably due to inaccuracies in the range-energy relations at low energies coupled with the increase of the energy spread of the beam as it proceeds through the stacked foils. We shall attempt to check the beam energy and uniformity by direct measurement. The shapes of the excitation functions are not greatly changed by this except to lower the points plotted at the low He^3 energies and make the rise in cross section more steep than shown in Figs. 23-25. Excellent agreement with the data of Cochran and Knight² taken at Los Alamos indicates that the absolute cross-section measurements (determined by standard radiochemical proportional-counter techniques) for the $\text{C}^{12}(\text{He}^3, \text{He}^4)\text{C}^{11}$ and $\text{C}^{12}(\text{He}^3, \text{d} \& \text{np})\text{N}^{13}$ reactions above 13-Mev He^3 energy are accurate (Fig. 24).

We have not drawn conclusions based solely upon the shapes of the excitation functions shown in this semiannual report; we shall attempt to supplement these data with cross-section experiments with other target nuclides. More detailed experiments, concerning the angular distributions of the outgoing particles, will help to determine what part of the total reaction cross section for a given process proceeds via compound-nucleus or pickup and stripping mechanisms. With respect to compound-nucleus processes, statistical theories--at least in the region of light nuclei studied here--can not be adequately applied with confidence.

An interesting secondary effect observed in the production of F^{18} from Al^{27} via the $(\text{He}^3, 3\text{He}^4)$ reaction was the high probability for formation of F^{18} deep in the stack of aluminum foil (see Fig. 23b). This was attributed

² D. R. F. Cochran and J. D. Knight, Bull. Am. Phys. Soc. Ser. II 3, 323 (1958).

MUB-237

Fig. 23. Excitation functions for He^3 -induced reactions in aluminum.

MUB-235

Fig. 24. Excitation functions for He^3 -induced reactions in carbon.

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Fig. 25. Excitation functions for He^3 -induced reactions in beryllium and oxygen.

to the reaction of He^3 with oxygen on the surface of the aluminum. A separate experiment, in which Mylar (a plastic which contains oxygen) was bombarded, showed a 290-mb cross section at about 7 Mev for the production of F^{18} from O^{16} (Fig. 25b). This suggests the possibility of a convenient He^3 activation analysis for surface oxygen and dissolved oxygen in various metals and other substances. We shall pursue further the details of this process with respect to advantages, disadvantages, ultimate sensitivity, cost, range of applicability, and other factors.

"Nuclear Reactions in Light Nuclei Induced by He^3 Ions" was presented (by SSM) at the New York Meeting of the American Physical Society, January 28-31, 1959 (Bull. Am. Phys. Soc. II, 4, 8 (1959)).

Radioactive Decay

THE ALPHA-BRANCHING RATIO OF TERBIUM-149

Lester Winsberg

Samples of neutron-deficient terbium isotopes were prepared by C^{12} bombardment of praseodymium oxide at the heavy-ion linear accelerator. After chemical purification¹ the samples were transferred to 1/4-mil aluminum foil and dried. The alpha activity was determined by counting the samples in an alpha-particle detector. The total decay via other modes was determined by counting the samples in a well-type scintillation counter. The crystal was a 3-in. -diameter 3-in. -long sodium iodide crystal with a hole 1/4 in. in diameter and 1.5 in. deep.

The sample was counted in the well-type counter for a time sufficiently long to resolve the decay curve into the Tb^{149} component. The discrimination was set as low as possible in order to get the maximum detection efficiency without too large a background. The efficiency for the detection of the 60-keV gamma radiation from Am^{241} was measured as approximately 90%.

The decay scheme of Tb^{149} is complex and involves the emission of many gamma rays² in addition to x-rays and positrons. Most of the gamma rays were found to be in coincidence with the remainder of the gamma spectrum, with the x-rays, and with the positrons.³ Less than 2% of the alpha particles was found to be in coincidence with the rest of the spectrum. Thus there is a high probability that every decay event via electron capture or positron emission was detected with the well counter.

The alpha-branching ratio was found to be 0.103, with a statistical uncertainty of about 10%. This value is rounded off to 0.10 ± 0.02 , because some direct transitions to lower states of Gd^{149} may not have been detected.

The positron activity was measured on a proportional counter and also determined from the gamma spectrum. The branching ratio for positron emission was found to be approximately 0.20.

¹ Kenneth S. Toth, Nuclear Studies in the Rare Earth Region (Thesis), UCRL-8192, May 1958.

² Kenneth S. Toth and John O. Rasmussen, Gamma Rays Following the Beta Decay of Rare Earth Alpha Emitters, J. Inorg. Nuclear Chem. (to be published).

³ This was done in collaboration with Frank Asaro.

ALPHA DECAY OF NEUTRON-DEFICIENT ASTATINE ISOTOPES

Richard W. Hoff, Frank Asaro, and Isadore Perlman

The alpha-decay properties of the neutron-deficient isotopes of astatine as produced in heavy-ion bombardments have been investigated. Gold foil targets were irradiated with C^{12} , N^{14} , and O^{16} ions accelerated in the heavy-ion linear accelerator to produce light isotopes of astatine and adjacent elements. The astatine and polonium were removed from the gold by volatilization onto a water-cooled plate, usually platinum or aluminum. No further chemical purification was made.

The energies, half-lives, and relative intensities of the alpha groups from the decay of the astatine isotopes were measured by using a double-focusing magnetic alpha-particle spectrograph. The mass assignment of all observed alpha groups has been made on the basis of recoil-collection experiments and relative formation cross sections as a function of bombarding-ion energy. These data for astatine isotopes with masses 201 to 207 are summarized as follows.

Mass Number	Half life (min)	Alpha-particle energy (Mev)
201	1.5 ± 0.1	6.348
202	2.9 ± 0.2	6.231 (36%)
	3.0 ± 0.2	6.133 (64%)
203	7.4 ± 0.3	6.086
204	9.3 ± 0.3	5.950
205	26.2 ± 0.5	5.899
206	21.7 ± 2.6	5.699
207	1.6 hr	5.750

The alpha-particle energies were measured relative to Cm^{242} (6.110 Mev) and Cm^{244} (5.800 Mev) as standards.

It should be noted that the data for the odd-mass isotopes, 201, 203, 205, and 207, agree well with those reported by Barton et al.¹ However, we have found no evidence for the alpha decay of the 25-minute At^{204} and the 2.9-hour At^{206} electron-capturing isotopes reported by Barton et al.¹ and Stoner² (for At^{206}). It is thought that in both cases we may be observing the predominant alpha-particle emitter of a pair of isomers, similar to the situation at mass 208.

¹Barton, Ghiorso, and Perlman, Phys. Rev. 82, 13 (1951).

²Allan W. Stoner, Nuclear Properties of Some Neutron-Deficient Isotopes of Emanation, Polonium, and Astatine (Thesis), UCRL-3471, June 1956.

Recoil-collection experiments were performed in which the astatine was first volatilized from the gold target onto a platinum plate. Recoil atoms were collected from this source at regular intervals on a negatively charged (-300 volts) platinum or aluminum plate. Although the experiments were originally designed with the intention of collecting bismuth isotopes from the recoil of the alpha-emitting astatines, the predominant activities on the recoil plates proved to be polonium isotopes. Subsequent recoil collections were made in which it was shown that Po^{201} , Po^{202} , and Po^{204} were being collected at such rates that 1.5-min, 3.0-min, and 9.3-min activities must be the respective parent nuclides. These facts represent a portion of the evidence upon which the mass assignments of At^{201} , At^{202} , and At^{204} are based. The mechanism of the recoil reactions apparently involves recoil from emission of either energetic positrons or conversion electrons by the parent astatine isotopes. The efficiency of such collection is approximately 3% for Po^{204} . The process was carefully checked to insure that the atoms collected were indeed positively charged, so as to eliminate possible volatilization from the original plate. The absence of large quantities of astatine activity on the recoil plate helps support this premise.

The two alpha groups of At^{202} suggest that two levels of Bi^{198} , separated by 98 kev, are being populated and that photons of this energy might be expected to be in coincidence with the 6.13-Mev alpha particles. On the basis of the alpha-gamma and alpha-L x-ray coincidence experiments that we have performed, we can set a limit of $< 5\%$ on the number of transitions of 100 kev or lower energy that are in coincidence with the 6.13-Mev alpha group. A possible explanation of this fact is that the 98-kev transition deexciting the level in Bi^{198} is an M2, E3, or higher multipole transition, in which case the conversion coefficient would be large and the lifetime would become long enough that, because of the resolving time ($\sim 0.5 \mu\text{sec}$) of the coincidence apparatus, it would be difficult to detect.

Similar coincidence experiments performed on other astatine isotopes have shown the following upper limits for photons between 20 and 100 kev in coincidence with alpha particles: $\text{At}^{201} \leq 0.15\%$, At^{202} (6.23 Mev) $\leq 0.6\%$, At^{202} (6.13 Mev) $\leq 0.45\%$, $\text{At}^{203} \leq 0.18\%$, and $\text{At}^{204} \leq 1.2\%$.

For At^{206} , L x-rays and a photon of approximately 68 kev were found in coincidence with the 22-minute 5.699-Mev alpha particles. Since no fine structure has been observed in the alpha spectrum for At^{206} , it is assumed that the alpha decay proceeds predominantly to a 68-kev excited level in Bi^{198} which decays to the ground state. The photon is approximately 24% abundant with respect to the alpha particles, indicating a conversion coefficient of 3, which would be consistent with an M1 transition.

A NEW ISOTOPE OF TERBIUM-152

Kenneth S. Toth, Kenneth T. Faler, and John O. Rasmussen

A paper has been submitted to Physical Review Letters.

ALPHA-DECAY THEORETICAL WORK

Richard R. Chasman and John O. Rasmussen

Work has continued on the effects of the nuclear quadrupole interaction on alpha decay of deformed nuclei; one paper has recently appeared,¹ and a second paper is soon to be submitted.

Further computations have been made on barrier penetrabilities with the diffuse optical-model potential derived by Igo² from data on alpha-particle elastic scattering. One paper is being published,³ and a second with calculations on all known alpha groups of odd-mass alpha emitters is nearly completed.

DECAY OF 11-HOUR TANTALUM-175

Kenneth T. Faler

Tantalum-175 has been produced both on the heavy-ion linear accelerator and on the 60-inch cyclotron by alpha-particle bombardment of lutetium. The gamma spectrum has been obtained from heavy-ion-produced samples, and conversion-electron spectra have been obtained from the alpha-particle bombardment samples. Conversion electrons have been studied both on the permanent-magnet spectrographs and on the double-focusing electron spectrometer. Intensity data together with multipolarity assignments are given in Table IV for the most intense transitions.

In addition to these transitions, weaker ones have also been identified at 77.4, 157.1, 186.0, 190.8, 193.3, 235.6, 349.0, 394.1, and 437.3 keV.

The half life of the isotope is 11 hours. No 511-keV radiation has been observed; that is, there is no detectable positron emission.

Several of the observed transitions are incorporated into a suggested decay scheme (Fig. 26) based on energy sums and on the assumption, on the basis of its large intensity, that the 81.6-keV transition goes to ground. Spins of the first three members of the ground-state rotational band are assigned in analogy to Lu^{173} decay. The ground-state band is believed to have the Nilsson state assignment $5/2^-$ (512).

¹Chasman and Rasmussen, Alpha Decay of Deformed Even-Even Nuclei, Phys. Rev. 112, 512 (1958).

²George Igo, Phys. Rev. Letters 1, 72 (1958).

³John O. Rasmussen, Alpha-Decay Barrier Penetrabilities with an Exponential Nuclear Potential. I. Even-Even Nuclei, Phys. Rev. (in press).

Table IV

Some transitions in decay of Ta¹⁷⁵

Transition energy (kev)	Relative electron intensity	Normalized gamma intensity	Multi- polarity	α		K/L		$L_I/L_{II}/L_{III}$	
				Theoret.	Obs.	Theoret.	Obs.	Theoret.	Observed
50.5	31		60% E2 40% M1	4.9				1/.95/1.0	1/.90/1.1
70.5	15(L)		35% E2 65% M1	9.1		1.0		1/2.0/1.9	1/2.5/1.5
81.6	18(L)	< 9	7% E2 93% M1	6.2		3.9		1/.30/.22	1/.31/.30
104.3	53		7% E2 93% M1	3.5		5.2	3.8	1/.22/.14	1/.24/.11
126.2	40	29.5	94% E2 6% M1	1.35	1.35	0.84	1.0	1/4.9/3.9	1/4.7/3.7
162.5	12	7	M1	1.14	1.7	7.2	8		
179.4	2.5		E2	0.36		1.4	2.8		
207.9	5.5	25	M1	0.53	2.2	6.9	5.8		
267.2	17	20	M1	0.26	.85	7.3	6

Fig. 26. Suggested decay scheme for Ta^{175} .

GAMMA SPECTRA OF TANTALUM-176

John O. Rasmussen and David A. Shirley

Beta spectroscopic studies of Ta¹⁷⁶ (unpublished) at the University of Washington in August of 1958 by Hugh Nutley and John Rasmussen showed a weak and complex positron spectrum and numerous conversion lines of energies up to ~1.6 Mev.

We have restudied this isotope in Berkeley, taking gamma spectra with a 3×3-inch NaI(Tl) crystal with 100-channel pulse-height analysis. The gamma spectrum is highly complex and only partially resolvable. Those peaks, mostly broad and complex, which are resolvable, and rough relative intensity measurements, corrected for counting efficiency, are as follows.

<u>Energy (Mev)</u>	<u>Relative intensity</u>	<u>Remarks</u>
0.5	1	Annihilation radiation
~ 0.70	1	-----
1.19	22	Broad, complex
1.67	10	Broad, complex
~ 1.9	6.3	Broad shoulder ahead of 1.67-Mev peak
~ 2.5	2.0	Broad
~ 3.0	--	Broad

Work is continuing on the complicated decay scheme of Ta¹⁷⁶.

Instrumentation

AN OSCILLOSCOPE COINCIDENCE UNIT

John A. Stone and Donald Strominger

Gamma-gamma coincidence counting is an important tool for the study of disintegration schemes. The usual method is to fix the pulse height from one scintillation counter and determine the coincident spectrum with another scintillation counter. This procedure is then repeated for several if not all of the resolvable gamma rays. To facilitate these experiments it would be convenient to record all the coincidence data simultaneously. Also, if all the coincidence data could be recorded simultaneously, then gamma-gamma coincidence experiments could be performed on some short-lived isotopes for which the usual method is impractical.

Grodzins¹ has described a technique for recording coincidences with the pulse heights of both counters simultaneously on photographic film. We have essentially duplicated 'Grodzins' system except that we have adapted standard UCRL units for several of the components. A block diagram of our system is shown in Fig. 27. We have used this system to obtain coincidence spectra of Na²² and Bi²⁰⁷ samples.

We are now in the process of building a permanent oscilloscope coincidence unit to be used in conjunction with the Penco analyzers.

AN ACCELERATING ELECTRON SPECTROGRAPH

Royal G. Albridge, Jr.

A permanent-magnet spectrograph capable of pre- or postacceleration of conversion electrons has been designed and built. A regulated power supply with a continuous range from ~7 to 40 kv is employed. The voltage regulation is constant to within ~0.01%. The spectrograph employs a 50-gauss permanent magnet and photographic recording.

For preacceleration a negative potential is applied to the source, a electrodeposited sample on a 0.010-inch-diameter platinum wire. The acceleration occurs between the source and grounded slit. A special film holder has been designed to maintain the photographic film and the tungsten wire grid immediately before the film at a positive potential for postacceleration.

Preliminary experiments with accelerating voltages in the range of 8 to 14 kv have shown little or no line broadening as a result of the acceleration. For low-energy electrons the film darkening for a given exposure time is doubled by use of the accelerating voltage.

¹L. Grodzins, Rev. Sci. Instr. 26, 1208 (1955).

MU-16778

Fig. 27. Block diagram of oscilloscope counting unit. The data are recorded photographically with a Dumont 'scope camera.

ISOTOPE SEPARATOR FOR C^{14} -ION BOMBARDMENT

Fred L. Reynolds

This report describes primarily the construction details of a small isotope separator to be used in low-energy ion bombardment of organic compounds. The main use of the machine is to label complex organic molecules by substitution of the C^{12} with radioactive C^{14} by bombardment techniques. Other studies are planned to ascertain the mechanism involved, using this method of atom exchange.

The entire separator is a compact unit, mounted on an angle iron stand 40 by 30 inches. The sector 90° magnet is C-shaped, with the coil positioned as in Fig. 28. This position of the coil does not provide a highly efficient magnet, but if the coil or coils were on the pole tips it (they) would greatly interfere with the ion source and the required collector assembly. The sector has a normal radius of curvature of 15 cm, and a 2.5-cm gap width, with the focus points located symmetrically 17.5 cm from the real pole faces. The fringe field moves the focus out about 2.5 cm from the theoretical 15-cm focus point.

A considerable amount of effort has gone into the design, testing, and redesign of the low-voltage arc source. The low-voltage capillary arc was chosen because it gives beam intensities in the microampere region, and secondly, the energy spread of the ions would be around 1 volt. Since we desired to operate the arc with an accelerating maximum voltage of 6 kilovolts, and as the machine is directional-focusing only, the capillary arc source seemed to be a proper choice. A low extraction voltage was also desired for an additional reason, since it seemed necessary to provide ion deceleration at the target end of the instrument. This arrangement will allow studies on the ion energy as related to the labeling process. The construction of the ion source is shown in Fig. 29, a cross-sectional view showing the arc chamber, extraction electrode, focusing electrode, and slit electrode. Both the extraction and slit electrodes are movable along their axis by rack-and-pinion motion through Wilson vacuum seals. The arc chamber is made of stainless steel, water-cooled with low-conductivity water flowing through a small glass heat exchanger. The capillary is 1/8 in. in diameter, 3/8 in. long, tapering to tubes of 5/8-in. diameter which hold the tantalum anode and cathode assembly. Both anode and cathode can be removed readily for cleaning or replacement. Gas flows into the arc from a side tube in the anode section, and ions are extracted through a 0.025-in. diameter hole to the first electrode, which is kept at ground potential.

The focusing electrode, mounted inside the extraction electrode on insulator standoffs, focuses the divergent beam onto the slit with a beam diameter of about 1/8 in. The operating potential on this electrode is only slightly less than the potential on the arc box and is of the same polarity. The final slit is operated at ground potential. Typical operating conditions using argon as the arc gas are as follows:

MU-16779

Fig. 28. Magnet yoke and coil.

MU-16781

Fig. 29. Schematic of capillary arc source and extracting and focusing electrodes.

Arc current (cathode to anode), 1 ampere
Gas pressure in arc chamber, $\approx 10^{-3}$ mm Hg
Beam intensity at focus electrode, 100 microamperes
Arc voltage (cathode to anode), 50 volts
Arc chamber, extraction voltage, 6 kv (+)
Focusing-electrode voltage, 5.8 kv (+)

The cathode filament has been constructed of 0.020-in. -diameter tungsten or tantalum wire. Considerable difficulty has been experienced in maintaining a reasonable life expectancy for these filaments under the above arc conditions. With argon the arc may operate for as long as 10 hours before the filament fails, but when gases containing oxygen are used, considerable chemical erosion takes place in addition to the normal positive-ion erosion. Sometimes the filament fails after 1 hour of running under oxidizing conditions. From 1 to 100 microamperes, the extracted beam is visible through a lucite window mounted on top of the ion-chamber assembly.

Construction of the vacuum pumping system had two major design features. The first was the necessity for high pumping speed and the second one of recovery of active CO_2^{14} gas, since this feed gas is expensive. Figure 30 is a diagram of the system; the main trajectory chamber is pumped by a 700-liter/sec oil pump, and the arc chamber is pumped by a separate 300-liter/sec oil diffusion pump. On either side of the liquid nitrogen traps it was necessary to place suitable valves so that the active CO_2^{14} could be pumped and recovered from the traps after a bombardment.

The receiver end is mounted on a pyrex glass insulator so that the entire assembly can be floated electrically to act as retarding field to the ion beam. Before the beam enters the target region it passes by two split electrodes which can deflect the beam along a horizontal plane. Placing a saw-toothed voltage on these plates makes the beam oscillate over a larger target area. The target area is essentially a cold-thimble-type cold trap, built square-shaped at the bottom to accommodate four targets. By rotation of the trap flange these targets can be positioned. If gas or volatile samples are to be studied, they may be frozen on the trap surface by bleeding the sample through a valve and through an inlet tube looking at the target area. A schematic drawing of the target assembly appears in Fig. 31.

MU-16783

Fig. 30. Block diagram of vacuum system.

MU-16782

Fig. 31. Schematic diagram of target section.

Radiation Chemistry

CHLOROETHYLENES

Amos S. Newton, Michael P. Sweeney, Jean Futrell,
and Aldo F. Sciamanna

Recent work on dichloroethylene oxide formation and the radiation chemistry of cis- and trans-dichloroethylene have been published as UCRL reports.^{1,2} This work has also been submitted to the J. Am. Chem. Soc. for publication.

Trichloroethylene

The radiation chemistry of trichloroethylene has been found to be more complex than previously thought, and may prove too complex to interpret completely with our present methods of analysis of the products. The symmetrical dichloroethylenes and trichloroethylene yield, in addition to a series of low-boiling products, two groups of high-boiling products. One is a series of four-carbon polyhalides resulting from dimerization of the monomer, and the other consists of higher-boiling products; trimers, tetramers, etc. of the monomer. Henne and Ruh³ have studied the free-radical-induced dimerization of trichloroethylene, and the products found were hexachlorobutenes. We have compared the products from the free-radical-induced dimerization of trichloroethylene formed by the decomposition of benzoyl peroxide in boiling trichloroethane with the high-boiling products found in the radiolysis.

Gas chromatograms of the chemical products show it to contain two hexachlorobutenes in the ratio of about 1:30. The high-yield product may or may not be a single isomer. No compounds of higher molecular weight than hexachlorobutene were found. The mechanism may be written as follows.

Initiation

¹J. H. Futrell and A. S. Newton, The Radiation Chemistry of the Symmetrical Dichloroethylenes, J. Am. Chem. Soc. (to be published).

²A. S. Newton and J. H. Futrell, The Synthesis of Cis- and Trans-1,2-Dichloroethylene Oxides, J. Am. Chem. Soc. (to be published)

³A. L. Henne and R. P. Ruh, J. Am. Chem. Soc. 69, 279 (1947).

Propagation Termination

The chain length for propagation observed was about 10, based on the yield of dimer produced and the amount of benzoyl peroxide initially added.

The exact structures of the heptachlorobutyl radical and the hexachlorobutene formed are not known. The mass spectrum of the products can be interpreted as either the 1,1,3,4,4,4-hexachlorobutene-1, as written, or as 1,2,3,4,4,4-hexachlorobutene-1, as stated by Henne and Ruh.³ The different products are formed according to whether the addition is head to head (as written in the equation) or head to tail, as stated by Henne and Ruh.

Among the products, we observed both pentachloroethane and a product the mass spectrum of which is best interpreted as that of a dichlorostyrene, so that the mechanism written appears quite straightforward.

The radiolysis products, however, contain no single dominant product but a mixture of at least 10 products, including at least three hexachlorobutenes. The higher-boiling materials cannot be formed by the same propagation chain as forms the chemical product. In particular, the higher-molecular-weight polymer cannot be formed by a mechanism corresponding to the one postulated for higher polymer in the dichloroethylene paper,¹ namely the addition of another monomer unit to the dimer radical. The chemical results show that heptachlorobutyl radicals do not add to another trichloroethylene molecule. Therefore another mechanism is operative in the radiation chemistry process which is not operative in the chemical process.

The investigation of this mechanism may be limited by the difficulty of obtaining the pure forms of products. Some of these are labile compounds and their decomposition products contaminate the stable products in the gas-chromatographic separations.

BENZYL CHLORIDE AS A RADICAL SCAVENGER

Amos S. Newton, Michael P. Sweeney, Jean H. Futrell,
and Aldo F. Sciamanna

In some exploratory work on methyl phenyl ether it was observed that a trace of benzyl chloride impurity yielded substantial yields of methyl chloride and chloromethyl ether. Some investigations of the use of benzyl chloride as a radical scavenger have therefore been started. As methanol has been investigated previously, this was used as a test compound. The results of a series of experiments in which the concentration of benzyl chloride was varied from 0.25% to 2.5% by volume indicate that benzyl chloride does not act as a general radical scavenger. For most gaseous products, H_2 , CH_4 , C_2H_6 , and CO , the only effect was a gradual decrease in yield with increasing concentration of benzyl chloride. This may be attributed to a small protective effect of an aromatic group. There is, however, a rapid growth of yield of methyl chloride and dimethyl ether.

In benzyl chloride solutions all formaldehyde is converted to methylal (HCl catalysis). This product does not vary appreciably in yield.

Any chloromethyl ether that might have been formed was not seen by the methods of analysis used. The products indicate some reactions of benzyl chloride with radicals, but not as efficiently as has been observed by using iodine as a scavenger according to the work of Meshitsuka and Burton.⁴ In particular, the yields of hydrogen and methane are not reduced by the presence of benzyl alcohol in the same manner nor to the same degree as occurred with iodine. Therefore benzyl chloride cannot be considered a scavenger for primary radicals. The formation of methyl chloride and excess dimethyl ether suggests that there is some scavenging action for secondary radicals.

Table V

Yields of methyl chloride and dimethyl ether in the radiolysis of benzyl chloride solution in methanol				
G Product	Conc. benzyl chloride (ml/100 ml CH_3OH)			
	0	0.25	0.75	2.5
CH_3Cl	0	0.012	0.13	0.73
CH_3OCH_3	0.13	0.18	0.33	0.68

⁴G. Meshitsuka and M. Burton, Rad. Research 8, 285 (1958).

Chemical Properties

HIGH-PRESSURE WORK WITH ACTINIDE METALS

Denis B. McWhan, James C. Wallmann, Burris B. Cunningham,
George Jura, and Juozas R. Vaisnys

We are doing some work on the high-pressure transformations in various actinide metals. Our method consists of measuring the resistance as a function of pressure from 30,000 atmospheres up to about 150,000 atmospheres. We are interested in studying americium, as the atomic spectra show that the 5f and 6d levels are quite close in energy, and an electronic transition similar to that seen in cerium may take place under pressure.

Our sample consists of a piece of metal which fills a 13.5-mil-diameter hole in a disk of silver chloride which is 8 mils thick. Around the disk of AgCl is placed a pipestone ring which is very strong with respect to radial pressure. This assembly is loaded between a set of anvils.

We encountered an interesting chemical problem in loading the samples: when a piece of reactive metal, such as one of the lanthanides or actinides, was pushed into the hole in the AgCl disk a reaction started to take place, as indicated by bubbling around the edges of the hole. This reaction could be stopped by blowing nitrogen on the ring while loading it. The surprising thing was the speed with which the reaction would start again if the stream of nitrogen were taken away (approximately 3 to 4 seconds). It is believed that the reaction is the dissolution of the metal by water, slightly acid, which is present on the AgCl surface; perhaps it is condensed by capillary action at the interface between the metal and the AgCl.

At present we have run neptunium, plutonium, and americium metal, and preliminary results show a transition only in neptunium (at about 95,000 atmos). The possibility exists that some transformations occur below the 30,000-atmos pressure at which our studies began. An apparatus to study phase transformations in this low-pressure region is being constructed.

CALORIMETRY

John D. Axe, Jr. and Hans Ihle

Measurements have been carried out on the heat of reaction of Pu(VI) with Fe(II) in 1 M HClO₄. Assuming that perchlorate complexing is negligibly small, the net reaction of millimolar Pu(VI) with a twofold stoichiometric excess of Fe(II) can be written

Under these conditions the reaction is rapid (essentially complete in less than 1 minute) and well suited to calorimetric study. In our experiments a freshly oxidized Pu(VI) solution (ozone was chosen for this oxidation) was allowed to react with a Fe(ClO₄)₂ solution in a microcalorimeter (previously described^{1,2}). The reaction was initiated by shattering a glass ampule containing a Fe(ClO₄)₂ solution, also in 1 M HClO₄. Separate experiments were made to determine the thermal effect of this operation. The amount of plutonium undergoing reaction was determined both by a radiometric and a gravimetric method. The gravimetric results were about 3% higher than the radiometric ones and were shown to be unreliable because of impurities (Ca, Mg, Si) in the Pu stock, present to the extent of several percent. The Pu²³⁹ aliquots were α-counted on a precision low-geometry counter and corrections were made for small amounts of Pu²³⁸, Pu²⁴⁰, and Am²⁴¹ isotopes present. We believe this analytical procedure to be accurate to about 1%.

Data from three determinations yield a value $\Delta\bar{H}_{298} = -51.6 \pm .8$ kcal/mole for Reaction (1). If, from the work of Connick and McVey³ and Fontana,⁴ we adopt a mean value for the reaction

with no correction for the change to 1 M HClO₄, we calculate

The previously accepted value for this reaction $\Delta\bar{H}_{298} = -77.8$ kcal/mole, given by Connick and McVey,³ derived from work by Evans⁵ has since been questioned by these authors.⁶ Rabideau and Cowan⁷ find a value of -78.4 kcal/mole for the above reaction in 1 M HCl. The difference may be due to the effect of chloride complexing in this medium.

¹Stuart R. Gunn, Thermodynamics of the Aqueous Ions of Americium (Thesis), UCRL-2541, April 1954.

²John D. Axe, Jr. and Hans Ihle, in UCRL-8369, p. 69.

³R. E. Connick and W. H. McVey, J. Am. Chem. Soc. 73, 1798 (1951).

⁴Quill, ed., The Chemistry and Metallurgy of Miscellaneous Materials, Natl. Nuclear Energy Series, Div. IV, Vol. 19B (McGraw-Hill, New York, 1950) p. 321.

⁵Seaborg, Katz and Manning, The Transuranium Elements, Natl. Nuclear Energy Series, Div. IV, Vol. 14B (McGraw-Hill, New York, 1949) p. 282.

⁶R. E. Connick and W. H. McVey, J. Am. Chem. Soc. 75, 474 (1953).

⁷S. W. Rabideau and H. D. Cowan, J. Am. Chem. Soc. 77, 6145 (1955).

THERMODYNAMICS: AMERICIUM

Harry L. Conley, Jr.

Thermodynamic data are essential to understanding the chemistry of any element. In the field of transuranium elements thermodynamic data are somewhat sparse and relatively inaccurate. To help remedy this situation it seemed necessary to obtain an accurate measurement of the entropy change associated with the reaction

With this entropy change the absolute entropy of AmO_2^+ can be determined if the entropy of AmO_2^{++} is estimated to be equal to the entropy of PuO_2^{++} , namely - 18 eu. Gunn and Cunningham¹ gave $\Delta H = +36.9 \pm 1.0$ kcal for

and using Penneman and Asprey's value² of $E_f = -1.60$ v for this couple they calculate $S_{\text{AmO}_2^{++}} - S_{\text{AmO}_2^+} = -16 \pm 4$ eu. To afford a check on these data the E and ΔH for the reaction given above will be measured. The ΔH will be measured calorimetrically and the E will be measured in the manner to be described below.

A Leeds and Northrup type K-2 potentiometer will be used for the potential measurement. The potentiometer can be read to 0.00002 v. A high-sensitivity Leeds and Northrup galvanometer with the following properties will be used;

Sensitivity: 0.00036 μ amp/mm,
Critical external damping resistance: 10,000 ohms,
Period: 6.8 sec,
Resistance: 553 ohms.

A lamp and scale-reading device will be used in conjunction with the galvanometer. An Eppley standard cell will be used in calibrating the potentiometer. The cell may be represented as follows:

An AmO_2^{++} solution will be prepared electrolytically and this solution will be allowed to undergo autoreduction to the AmO_2^+ ion as the potential is measured. The total americium concentration will be measured and the AmO_2^{++} concentration will be measured spectrophotometrically; thus the ratio $(\text{AmO}_2^{++})/(\text{AmO}_2^+)$ will be known. The junction between the two half cells will be made by means of an ungreased ground-glass joint, so that the resistance of the cell will be high and will have to be compensated for if the best galvanometer operation is to be had.

¹ S. R. Gunn and B. B. Cunningham, J. Am. Chem. Soc. 79, 1563 (1957).

² R. A. Penneman and L. B. Asprey, The Formal Potential of the Am(V)-Am(VI) Couple, Los Alamos Sci. Lab. Report, AECU-936, Sept. 1950.

The potential of the Ce^{+3} - Ce^{+4} couple has a value of -1.70 in 1.00 N HClO_4 , as reported by Smith and Goetz.³ A measurement of this couple will be made in order to check the experimental procedure and equipment. In preliminary experiments, the potential of the following cell was measured,

The cell reaction was

A small U-shaped glass tube filled with agar saturated with KNO_3 was the salt bridge. The ratio $(\text{Ce}^{+4})/(\text{Ce}^{+3})$ was changed by adding $\text{FeSO}_4(\text{NH}_4)_2\text{SO}_4 \cdot 6\text{H}_2\text{O}$. Neglecting any junction potential and using concentrations instead of activities, one could calculate the potential of the cell by

$$E_{\text{calc}} = E_{\text{Ce}^{+3}-\text{Ce}^{+4}}^0 - 0.0591 \log \frac{(\text{Ce}^{+4})}{(\text{Ce}^{+3})} + E_{\text{KCl}(\text{sat.}); \text{Hg}_2\text{Cl}_2, \text{Hg}}$$

$E_{\text{Ce}^{+3}-\text{Ce}^{+4}}^0 = -1.44 \text{ v}$ as reported by Kunz,⁴ and

$$E_{\text{KCl}(\text{sat.}); \text{Hg}_2\text{Cl}_2, \text{Hg}} = 0.2415 \text{ v}.$$

The ratio $(\text{Ce}^{+4})/(\text{Ce}^{+3})$ was determined by starting with a known Ce^{+4} solution and adding weighed amounts of $\text{FeSO}_4(\text{NH}_4)_2\text{SO}_4 \cdot 6\text{H}_2\text{O}$.

$E_{\text{exptl.}}$	E_{calc}	$(\text{Ce}^{+4})/(\text{Ce}^{+3})$
-1.47	-1.49	8.827
-1.45	-1.47	4.009
-1.43	-1.46	2.323
-1.41 ^a	-1.44	1.000

^aThis value is extrapolated from a plot of E_{exptl} vs. $\log (\text{Ce}^{+4})/(\text{Ce}^{+3})$.

³G. F. Smith and A. Goetz, Ind/ Eng. Chem., Anal. Ed. 10, 191 (1938).

⁴A. H. Kunz, J. Am. Chem. Soc. 53, 98 (1931).

ATTEMPTED PREPARATION OF
THORIUM TRICHLORIDE FOR PARAMAGNETIC RESONANCE STUDY
OF THE ELECTRONIC STRUCTURE OF THORIUM

Ru-Tao Kyi

The preparation consists of a reduction of ThCl_4 in LaCl_3 by some reducing agent, and subsequent growth of a single crystal of LaCl_3 containing small amounts of thorium (III) ($\sim 0.1\%$). LaCl_3 is used as a diluent to reduce the spin-spin interaction--i.e., the mutual effect of one paramagnetic ion on another--and also as a stabilizer to the product ThCl_3 , since they both have the same crystalline structure.

According to the free-energy change for the reduction and from previous experiments, ammonia was chosen as the reducing agent. ThCl_4 and LaCl_3 of proper ratio were sublimed in a tube under vacuum and then ammonia was admitted to a pressure of 0.2 atmosphere. The tube was sealed and the crystal grown in an oven at 900°C for 5 days.

The residual gas was taken out for mass analysis and the results show that N_2 and H_2 were present as the product of decomposition of NH_3 . Paramagnetic-resonance measurement of the crystal showed the existence of an impurity, gadolinium, but no other lines.

LaCl_3 was then purified from gadolinium as follows:

1. La_2O_3 was dissolved in 6 N HCl .
2. Lanthanum was incompletely precipitated by NH_3 gas and the supernatant solution was decanted.
3. The precipitate was dissolved and similarly reprecipitated five times.
4. The final precipitate was dissolved in 6 N HCl and dried in an evacuated desiccator, which resulted in the formation of a powder.
5. Purification was again accomplished by sublimation.

No gadolinium resonance line was shown by paramagnetic resonance in the crystal of purified LaCl_3 .

A second reduction of ThCl_4 in purified LaCl_3 was attempted, using thallium metal as a reductant.

The paramagnetic-resonance spectrum of this sample has not yet been determined.

Optical Spectroscopy

ABSORPTION SPECTRUM OF URANIUM TETRAFLUORIDE

John G. Conway

The absorption spectrum of UF_4 has been measured through the wave-length range of 2000 Å to 8 μ . Most of the work was at liquid nitrogen temperatures. Experiments at liquid helium temperatures did not sharpen up the lines. The observed wave-length lines due to U^{+4} as the fluoride in a matrix of CaF_2 are listed below. Three crystals of differing concentrations were used.

Wave lengths of U^{+4} as fluoride in CaF_2

		(continued)
2349	angstroms	8955
3920		8965
4193		8981
4418		8995
4745		9157
4803		9179
4813		9199
4823		9298
4831		10797
4837		11080
4847		11128
5256		
5391		
6269		1.17 ^a microns
6488		1.569
6507		1.592
6545		1.603
6562		1.674
6569		1.684
6578		1.697
6592		1.704
6600		1.716
6659		2.42 ^b
7715		
8788		
8935		

^aBroad^bVery broad

Intermediate field calculations have been made by using the parameters given by Elliott, Judd, and Runciman¹ for 5f electrons.

Work is now directed to testing the fit of experimental data to the calculated diagrams and if possible to find ways of improving the fit.

¹Elliott, Judd, and Runciman, Proc. Roy. Soc. A 240, 509 (1957).

ATTEMPTS TO OBSERVE THE EMISSION SPECTRUM
OF ASTATINE

Ralph D. McLaughlin

Rough calculations indicate that it should be possible to produce as much as a few tenths of a microgram of astatine by bombarding bismuth metal with alpha particles from the 60-inch cyclotron. Experience with the use of the electrodeless discharge tube as a spectroscopic source indicates that this amount of material is sufficient to produce observable spectra.

Two methods of transfer of the astatine from the bismuth target to the electrodeless discharge tube have been attempted. In one method the discharge tube was one of the arms of a U-tube. The U-tube was placed in a cold bath, the astatine distilled into the U-tube, and the arm containing the astatine sealed off. In the other method the astatine was sublimed from the target to a Pt foil which was placed in the discharge tube, and the spectra were observed.

So far, three bombardments have been completed in which close to the maximum amount of astatine has been produced. Two of these three bombardments have yielded spectral lines which cannot be easily explained from a knowledge of the emission lines tabulated in the Massachusetts Institute of Technology wave-length tables. The wave lengths of these lines are 2162.3, 2244.0, 2564.5 and 2566.2 Å.

SPECTROCHEMICAL DETERMINATION OF STRONTIUM-TO-CALCIUM
RATIOS IN MILLIGRAM-SIZE SAMPLES OF ASHES

George V. Shalimoff

A spectrochemical analytical method has been developed and is being used to determine the ratio of strontium to calcium in milligram samples of ashes of babies' diapers from a control experiment conducted by Crocker Laboratory. It has been shown earlier that strontium-to-calcium ratios can be obtained with good spectrographic precision in small samples of food and products of cows.¹ The method in this study differs only in the manner of sample preparation, namely, that one part by weight of sample is mixed with 10 parts of finely powdered germanium dioxide. In a dc arc the vaporization of germanium dioxide creates a smooth burning condition which improves the reproducibility of excitation of the samples. Ratios of strontium to calcium in the range of 10^{-2} to 10^{-4} have been obtained for various ash samples.

¹Shalimoff, Conway, and Pitzer, Spectrochemical Determination of Strontium-to-Calcium Ratios in Food, Milk, Cream, Blood, Feces, and Urine of Cows, in "Applied Spectroscopy," Vol. 12, No. 4 (1958).

OPTICAL SPECTROSCOPY OF RARE EARTH
AND ACTINIDE +3 IONS

John B. Gruber

Investigations are being undertaken to identify the electronic ground states and the low-lying excited states of the rare earth ions Eu^{+3} , Gd^{+3} , and Tm^{+3} and the actinides Am^{+3} , Cm^{+3} , Bk^{+3} , and Cf^{+3} in LaCl_3 as a matrix. Single crystals of the +3 ions in concentrations of 3 to 10% in LaCl_3 were grown by passing the sublimed material, sealed in quartz tubes, through a temperature-controlled tube furnace at 900°C . The single crystals thus grown contain the +3 ions in C_{3h} point symmetry at the ion sites in the lattice. LaCl_3 is isomorphous with the +3 ions investigated. The optical spectra observed for each one of the rare earth ions showed no impurity lines, indicating that the impurity in the rare earths used was less than 0.1%.

The spectrum of Gd^{+3} (ground state: $^8\text{S}_{7/2}$) was studied in the u. v. Our results agree with those reported in the literature. A remarkable similarity between the Gd^{+3} and the Cm^{+3} spectra was observed. The spectrum of $\text{GdCl}_3 \cdot 6\text{H}_2\text{O}$ was also studied.

The optical spectra of Eu^{+3} in the range from 2000 to 8000°A (ground state: $^7\text{F}_0$) was next considered. Single crystals of $\text{EuCl}_3 \cdot 6\text{H}_2\text{O}$ were grown from solutions of 6 M HCl in a vacuum desiccator. There is some question as to the crystal structure of the hydrate $\text{EuCl}_3 \cdot 6\text{H}_2\text{O}$. Hellwege and Kahle¹ claim it is monoclinic, but their claims are based on unpublished data and from an analysis of their spectral data. Low-lying states $^5\text{D}_0$, $^5\text{D}_1$, $^5\text{D}_2$ were observed in the hydrate spectrum. These were presumably transitions from the $^7\text{F}_0$ ground state.

The optical spectrum of the anhydrous EuCl_3 was also observed and the spectral lines were measured. Single crystals containing 3% EuCl_3 in isomorphous LaCl_3 were grown from the melt. One-half atmosphere of Cl_2 was placed over the material at all times so that no large amounts of Eu^{+2} would be formed as a result of thermal decomposition of Eu^{+3} . The observed spectral lines agreed with those of Sayre and Freed and other workers.² One sees no lines in the yellow region of the spectrum (5800°A), two lines in the green (5200°A), and one line in the blue (4650°A). The spectrum taken below 4000°A showed the setting in of a continuum due to the strong f-to-d electron transition from the very small amounts of Eu^{+2} present. We intend to compare the Eu^{+3} spectrum with that of Am^{+3} in the near future.

The optical spectrum of Tm^{+3} (ground state: $^3\text{H}_6$) is being studied in order to find lines that have not yet been reported in the literature, but which according to theory ought to be present. The excited $^3\text{P}_{0,1,2}$ states are expected by theory to be observed in the region from 2500°A to 3000°A . Difficulty in observing any transitions in this region is due to the strongly allowed electronic transition from 4f to 5d shells, which results in a broad continuous band.

¹ Hellwege and Kahle, Z. Physik. 129, 62 (1951).

² Sayre and Freed J. Chem. Phys. 23, 2066 (1955).

X-Ray CrystallographyTHE CRYSTAL STRUCTURE OF THE 1-1 ADDITION COMPOUND
CARBON TETRABROMIDE · p-XYLENE

Frederick J. Strieter and David H. Templeton

It is known that p-xylene forms addition compounds with several halogenated methanes at reduced temperatures. The compound with carbon tetrabromide is solid at room temperature, melting at 53°C. Its crystal structure has been investigated to reveal the type of bonding configuration.

X-ray-diffraction data on a single crystal showed the symmetry to be orthorhombic, and a satisfactory structure was found in space group Cmc₂m. The unit cell, containing 4 molecules, is

$$\begin{aligned} a &= 8.43 \text{ \AA}, \\ b &= 8.96 \text{ \AA}, \\ c &= 17.45 \text{ \AA}. \end{aligned}$$

The atomic coordinates found by least-squares refinement are

	<u>x</u>	<u>y</u>	<u>z</u>
Br(1)	0	0.204	0.339
Br(2)	0.184	0.450	1/4
C (3)	0	0.332	1/4
C (4)	0.361	0.431	0.019
C (5)	1/2	0.372	0.050
C (6)	1/2	0.231	0.097

These coordinates correspond to C-Br bonds in CBr₄ of 1.88 and 1.94 Å; C-methyl bonds of 1.52 Å; and C-C aromatic bonds of 1.42 and 1.39 Å. The accuracy is no better than 0.07 Å because of large thermal vibrations. The interesting feature of the structure is that each benzene ring has a bromine atom on its axis on each side. This configuration suggests secondary bonding of bromine to the π electrons of the ring. Of each CBr₄, two bromine atoms are involved in this bonding.

CRYSTAL STRUCTURE OF PROPIONIC ACID

Frederick J. Strieter and David H. Templeton

Low-temperature x-ray studies have permitted determination of the structure of solid propionic acid. The crystals are monoclinic, space group $P2_1/c$, with 4 molecules in a unit cell:

$$\begin{aligned}a &= 3.99 \text{ \AA}, \\b &= 9.066 \text{ \AA}, \\c &= 10.98 \text{ \AA}, \\\beta &= 91.3^\circ.\end{aligned}$$

It is known that formic and acetic acids crystallize in long hydrogen-bonded chains, while heavy fatty acids are hydrogen-bonded dimers. The structure found for propionic acid is a dimer, with bond distances as indicated:

There is a center of symmetry at the center of the dimer. These distances are calculated for a structure which gives an "unreliability index" of 11.6%, achieved by least-squares refinement.

THE CRYSTAL STRUCTURE OF AMMONIA MONOHYDRATE

Ivar Olovsson and David H. Templeton

Ammonia monohydrate or ammonium hydroxide, $\text{NH}_3 \cdot \text{H}_2\text{O}$, freezes at -79°C . Its crystal structure has been determined from x-ray data taken at -160°C from a single crystal. The structure was refined by least-squares fitting of three-dimensional data.

The crystal is orthorhombic, space group $\text{P}2_12_12_1$, with four molecules in a unit cell of these dimensions (at -160°C):

$$\begin{aligned}a &= 4.51 \pm 0.01 \text{ \AA}, \\b &= 5.587 \pm 0.003 \text{ \AA}, \\c &= 9.700 \pm 0.005 \text{ \AA}.\end{aligned}$$

The density is calculated as 0.95. The coordinates for oxygen and nitrogen (in fourfold general positions) are:

	<u>x</u>	<u>y</u>	<u>z</u>
O	0.241	0.110	0.017
N	0.264	0.272	0.334

The structure consists of a hydrogen-bonded network. Each oxygen is hydrogen bonded to two others at a distance of 2.76 Å in a zigzag chain. Each oxygen is also bonded to four nitrogen atoms at distances 2.77, 3.21, 3.26, and 3.29 Å. Each nitrogen atom has four oxygen neighbors at these same distances. The configuration about nitrogen is approximately tetrahedral, while that around oxygen is somewhat like the sixfold coordination of nitrogen in solid ammonia. The short bonds are attributed to the hydrogens of water, while the long bonds are attributed to the hydrogens of ammonia, three of which compete for a pair of electrons on oxygen.

This assignment of hydrogen atoms to the bonds accounts for all of them, except that in the oxygen-oxygen bonds there are two possible choices of configuration for each chain. The possibility of randomness in a chain of this sort would not lead to observable entropy, however, because all bonds of a chain must be consistent with one another.

OTHER CRYSTAL STUDIES

George L. Hardgrove, Jr., Quintin C. Johnson, Ivar Olovsson,
Helena W. Ruben, and David H. Templeton

Structural work is in progress on several substances. The refinement of the structures of $\text{Na}_2\text{SO}_4 \cdot 10 \text{H}_2\text{O}$ (Glauber's salt) and semicarbazide hydrochloride is well advanced. A structure for $\text{Na}_2\text{C}_2\text{O}_4 \cdot 4 \text{H}_2\text{O}$ gives reasonably good agreement with the data, but it is not yet certain if it is correct. A structure has been found for cis-1,2-dichlorobenzocyclobutene at room temperature, but the accuracy is low because of large thermal vibrations. The work is being repeated at a lower temperature to improve the result. A different crystal structure has been found for cis-1,2-dibromobenzocyclobutene and the isomorphous iodine compound. In this case the symmetry of the space group, $\text{Pna}2_1$, does not define the origin of the c axis. There results a strong correlation of z coordinates which has severe effects, as it interferes with convergence in the least-squares refinement as ordinarily carried out. A full-matrix (40×40) refinement is being computed for the bromine compound to shed light on the operation of the least-squares method and incidentally to improve the accuracy of the structure of this interesting fused-ring system.

GENERAL CHEMISTRY

Leo Brewer, Robert E. Connick, and Kenneth S. Pitzer in charge

Metals and High-Temperature Thermodynamics

SPECTROSCOPIC STUDIES OF C_2 AND C_3

John Engelke, Sandor Trajmar, and Leo Brewer

Additional temperature-coefficient measurements are in progress to fix the heat of sublimation of C_3 more accurately. The C_3 spectrum is being examined on the 21-foot grating for evidence of structure.

NEW LIGHT SOURCES

Earl Worden and Leo Brewer

A steady bright CN arc source has been developed and its brightness temperature has been measured. The brightness temperatures of a number of other light sources have been measured for comparison. This problem has been completed and has been reported separately.¹

ABSOLUTE-LIFETIME APPARATUS

Richard Brewer, Fred Stafford, and Leo Brewer

The lifetime apparatus has been completed and the lifetime of the upper electronic state of the visible bands of I_2 has been measured and has been found to be less than 3×10^{-7} sec. The sensitivity of the apparatus is being improved and measurements are in progress on sodium vapor. It is also planned to make additional measurements on iodine before measurements on high-temperature molecules such as CN, TiO , and C_3 are attempted. These results have been reported elsewhere.²

TEMPERATURE SCALE AT HIGH TEMPERATURES

John Engelke, Earl Worden, R. Brewer, Fred Stafford,
Robert Berg, and Leo Brewer

Leo Brewer, Earl Worden, and Oscar Krikorian attended the Pyrometry Conference held at the National Bureau of Standards in October. The results of temperature calibrations at Berkeley and Livermore were

¹ Earl F. Worden, Jr., High-Intensity Light Sources (Thesis), UCRL-8509, Oct. 1958.

² Richard G. Brewer, I. A Method for Determining Radiative Lifetimes of High-Temperature Molecules. II. The Probability of Spontaneous Nuclear Reaction in Molecular Hydrogen (Thesis), UCRL-8387, July 1958.

presented. The most serious discrepancies of the temperature-scale calibrations have now been eliminated. Elimination of deviations of the order of 10 to 20° between the thermodynamic scale and the working-temperature scales depends upon work in the various national standardizing laboratories which is aimed at fixing the gold point on the thermodynamic scale. Additional refinements depend upon more widespread use of photoelectric pyrometers.

THERMODYNAMIC COMPILATION OF DATA ON GASEOUS OXIDES

Mallasetty S. Chandrasekharaiah and Leo Brewer

Free-energy functions have been calculated as a function of temperatures for the MO and MO₂ gaseous oxides of the periodic table. Analysis of published equilibrium data is in progress to fix the best values of the heats of formation.

Theoretical Studies

POLYMER CARBON MOLECULES

Enrico Clementi and Kenneth S. Pitzer

Work is in progress as referred to in the preceding Semiannual Report (UCRL-8369, p. 92).

ELECTRONIC STATES OF C_2 MOLECULE

Enrico Clementi and Kenneth S. Pitzer

Energy-state calculations for the $^1\Pi_u$, $^3\Pi_u$, $^1\Sigma_g^+$, $^3\Sigma_u^-$, $^1\Delta_u$, $^1\Sigma_u^+$ states (corresponding to the lowest configurations

$\sigma_g^2 \sigma_u^2 \pi_u^3 \sigma_g$, $\sigma_g^2 \sigma_u^2 \pi_u^4$, $\sigma_g^2 \sigma_u^2 \pi_u^2 \sigma_g^2$) are in advanced status. A

modification of conventional A. S. M. O. approximation is used: without inclusion of C. I., exact dissociation values are obtained. One-center repulsion integrals are calculated from atomic data. One-center (α) and two-center (β) core integrals for π as well as for σ electrons are in calculation by an iteration technique (α and β are obtained from spectroscopic data as a function of interatomic distance). The 701 electronic computer was used for the solution of secular equations. A program for the above iterative computation is being studied.

ELECTRONIC STATES OF C_3 MOLECULE

Enrico Clementi and Kenneth S. Pitzer

The lowest states $^1\Sigma_u^+$, $^3\Sigma_u^+$, $^1\Sigma_u^-$, $^3\Sigma_u^-$, $^1\Delta_u$, $^3\Delta_u$ and $^1\Sigma_g^+$ (arising from π -electron excitation) and the $^1\Pi_u$, $^3\Pi_u$ (σ -to- π excitation) are considered. The A. S. M. O. type of approximation with inclusion of nonneighborhood terms is used. One-center repulsion integrals and core integrals are obtained from C and C_2 data.

The indication so far obtained is that the $^1\Sigma_g^+$ state is substantially below the first Π excited state. This implies that the excited states of C_3 are not crowded near the ground state as they are in the C_2 molecule.

INTERACTION OF TWO MOLECULES WITH A SURFACE

Oktaý Sinanoglu

The mutual Van der Waals repulsion of two molecules and a surface, as has been observed in physical adsorption,¹ was derived by using quantum mechanical third-order perturbation theory for a general plane surface.

¹Mark P. Freeman, in Chemistry Division Semiannual Report, UCRL-8369, July 1958, p. 93.

EQUATION OF STATE OF ALKALI VAPORS

Oktay Sinanoglu and Kenneth S. Pitzer

For the calculation of equation of state and thermodynamic and transport properties of alkali vapors (and Hg, etc.), the usual "method of significant structures" (i. e., considering monomer and dimer species) is not satisfactory, even when anharmonicity corrections are included, because of the relatively weak interaction between these atoms. It is necessary to have the virial coefficients for a realistic potential-energy function and thus calculate the properties of such vapors as those of an imperfect monatomic gas. Suitable diatomic potentials are being considered, and tables of reduced second virial coefficients for the reduced temperature range $0.1 < T^* < 3.0$ and for a range of reduced potential parameters will be computed along with the first and second temperature derivatives and the first quantum correction to $B(T)$. The work is in progress.

Inorganic Chemistry

RUTHENIUM STUDIES

Dwight Fine and Robert E. Connick

Investigation of chloride complexes of ruthenium (III) is being continued. Two cationic species have been identified as RuCl_2^+ by ion-exchange and analytical methods. These species are presumed to be cis- and trans-isomers, although specific assignment of configuration is not possible at this time. It is hoped that application of the Kerr effect will prove enlightening as to configuration. Effort is currently being directed toward separation and identification of anionic species.

CHEMISTRY OF +1 IODINE IN ALKALINE SOLUTION

Yuan T. Chia and Robert E. Connick

This work has been completed and is reported separately.¹

SPECIES IN REFLUXED CHROMIC NITRATE SOLUTIONS

James Finholt and Robert E. Connick

Attempts to prepare a known +4 model compound were abandoned because of unexpected synthetic difficulties.

At the April 1958 meeting of the American Chemical Society, Dr. Plane stated that one of the species produced by refluxing Cr(III) would be obtained more conveniently by oxidizing Cr(II) with O_2 . The material prepared in this manner has the same charge per Cr atom (+2) and the same spectra as the blue species formed in refluxing.

All attempts to crystallize a Br^- or ClO_4^- salt of this material failed.

Charge-per-species experiments at constant ionic strength were run on this species. A result of $+4 \pm 1$ was obtained. The large uncertainty was due primarily to analytical uncertainty. This difficulty will shortly be corrected.

BIARSINE

Leon Burright and William L. Jolly

A modified dew-point method is being used to determine the vapor pressure of a volatile arsenic hydride, probably biarsine. Molecular weight determinations are being made to decide whether the gas is biarsine or not.

¹Yuan-Tsan Chia, Chemistry of +1 Iodine in Alkaline Solution (Thesis), UCRL-8311, July 1958.

STIBINE

Ladislav H. Berka and William J. Jolly

The study of a new method for the preparation of stibine, that of reacting aqueous solutions of potassium or sodium borohydride with antimony trichloride solutions, showed the percentage yield based on borohydride to be greater at higher pH, lower antimonous concentrations, and lower reaction temperatures.

Attempts to determine the nature of the reaction(s) between stibine and sodium in liquid ammonia were inconclusive owing to the catalytic effect of ammonia upon the decomposition of stibine into its elements.

Preliminary measurements of the vapor pressure of stibine at various temperatures (-95°C to -22°C) have indicated that similar data reported in the literature may be incorrect by as much as 10%. Present work is being done to obtain more accurate and reliable data over this temperature range.

Spin Resonance and Infrared Spectroscopy

INTERNAL ROTATION IN MOLECULES

Tad Beckman and Kenneth S. Pitzer

Infrared study in the CsI region is being employed to determine the barrier to internal rotation in ethyl chloride. The study has the twofold purpose of resolving the scatter in data already reported and to complete the series of barrier studies on chlorinated ethanes.

The general problem of internal rotation is also receiving some consideration. A review of past theories indicates that the cause of the rotation barrier is a complex phenomenon involving bonding and electron distributions. New calculations have been initiated with the hope that barrier trends may be obtainable in spite of the fact that barrier magnitudes are not.

INFRARED SPECTRA OF METAL-AMMONIA SOLUTIONS

Enrico Clementi and Kenneth S. Pitzer

Work is in progress as referred to in the preceding report (UCRL-8369, p. 92). The reflection cell is completed.

MAGNETIC-RESONANCE STUDIES OF METALS IN METAL HALIDES

Judith Brown and Kenneth S. Pitzer

An investigation of solutions of metals in metal halides in the liquid state is being undertaken, utilizing the techniques of electron-spin resonance and nuclear magnetic resonance(NMR). These studies should yield new information on the hypothesis relating solubility to subhalide formation. It is hoped that the development of high-temperature techniques will make possible an investigation of the state of the conduction electron introduced in solutions of the alkali metals in the alkali halides and a comparison of this type of system with that of sodium in liquid ammonia.

NMR SPECTRA OF SOME MOLECULAR COMPLEXES

Juana Acrivos

The proton NMR spectra of the molecular complexes of I_2 with benzene and mesitylene are being studied. Small negative shifts are found besides a broadening of the absorption lines. Fe^{+3} in benzene is also being studied in order to determine shifts caused by indirect hyperfine interactions between the electron spin of the $^6S_{5/2}$ Fe^{+3} and the benzene protons in the 1:1 molecular complex.

STUDY OF HYDROGEN BONDING IN ALIPHATIC ALCOHOLS BY NMR SPECTROSCOPY

Jefferson C. Davis, Jr., and C. N. Ramachandra Rao

Attempts are being made to study substituent effects on the hydrogen bonding in aliphatic alcohols by nuclear magnetic-resonance spectroscopy.

PARAMAGNETIC RESONANCE

Rollie J. Myers, Norman Edelstein, Larry R. Doyle,
and Robert G. Hayes

The instrument construction which we hoped would be completed during the summer has extended into the fall. However, we have started some work on impurity centers in calcite and on the paramagnetic resonance of the OH radical.

NMR STUDIES OF HYDROGEN BONDING

Jefferson C. Davis, Jr., and Kenneth S. Pitzer

The nuclear magnetic resonance spectra of several carboxylic acids have been studied in various solvents in order to determine the equilibria between the monomeric species of the acids and the hydrogen-bonded dimers. The electronic shielding of the carboxylic protons in these two species has also been of interest. Although it has not been possible to determine the equilibrium constants directly, even at higher temperatures at which the amount of monomer becomes appreciable, information about the electronic shielding has been obtained. It appears that the monomeric protons are highly shielded, even more so than the corresponding protons in alcohols. Solutions of alcohols in which the equilibria are more easily studied are also being investigated, with the object of obtaining more exact information about systems of this kind. Quantum-mechanical calculations are being made to elucidate some of the results obtained.

Solid-State and Low-Temperature Thermodynamics

HEAT CAPACITY OF SOLID HYDROGEN UNDER PRESSURE

William Orttung and Kenneth S. Pitzer

A project for the measurement of the specific heat of solid hydrogen under high pressure has been started. At 1 atmosphere there is an anomaly in the specific heat in the temperature range between 1°K and 10°K caused by a splitting of the rotational levels of the ortho molecules. At low pressures the zero-point energy of the lattice keeps the molecules rather far apart, and the splitting is the result of electric quadrupole interactions between the adjacent ortho molecules. By applying pressures between 5000 and 10,000 atmospheres we hope to move the molecules close enough together to be able to observe the effect of exchange repulsion on the splitting. At high enough pressures the rotation should become an oscillation. It is hoped that high enough pressures will be obtained to approach the rotation to vibration transition. It is also hoped to carry out the experiments on D₂ and HD.

ANOMALOUS HEAT CAPACITY OF SILVER OXIDE BETWEEN 20° AND 45°K

Lawrence V. Gregor, C. N. Ramachandra Rao, and Kenneth S. Pitzer

Attempts are being made to investigate possible causes of the anomalous heat capacity of silver oxide between 20° and 45°K. The solid-state chemistry of silver oxide is being studied by chemical, x-ray, and differential thermal analyses.

LOW-TEMPERATURE HEAT CAPACITIES OF METALS

Norman E. Phillips

Modifications in the apparatus have permitted an extension of the measurements to 0.1° K. Measurements on aluminum in the normal and superconducting states, in the temperature interval 0.1 to 4.0° K, have been completed and submitted for publication. Measurements on zinc have been completed between 0.1 and 1.2° K and the results have been published.¹ Measurements on cadmium have been undertaken. The results for zinc and aluminum at very low reduced temperatures are not in agreement with predictions based on the energy-gap model of current theories of superconductivity.

HELIUM-3 CALORIMETER

William H. Lien and Norman E. Phillips

Work on the construction of a calorimeter for temperatures between 0.3 and 30° K is continuing. The calorimeter will employ He³ to attain temperatures below 1° K.

¹ Norman E. Phillips, Phys. Rev. Letters 1, 363 (1958).

LOW-TEMPERATURE HEAT CAPACITY OF Ag_2O

Roger Gerkin and Kenneth S. Pitzer

This work has been completed and a report is being written.

CHEMICAL ENGINEERING (PROCESS CHEMISTRY)

ZONE MELTING

William R. Wilcox and C. R. Wilke

Although separations of mixed crystalline materials have been attained by pulling the test rod up vertically through a stationary heater, large bubbles form on the freezing interface. This situation defies theoretical treatment. Therefore the apparatus is being turned upside down, i. e., the rod will now be pulled down.

The first known rigorous treatment of pure diffusional mass transfer in zone melting has just been completed. The differential equation and boundary conditions were derived and solved numerically for constant distribution coefficients and for eutectic-forming systems. The results will now be compared with the experimental results.

The refrigeration system for the new cooled-roller apparatus for continuous fractional crystallization has been completed. The roller apparatus itself is nearing completion.

MULTICOMPONENT DISTILLATION STUDIES USING AN IBM-701 TWO-FRAME DIGITAL COMPUTER

John H. Duffin, Donald N. Hanson, and C. R. Wilke

The problems mentioned in the preceding report have all been solved, and in addition energy balances have been successfully applied to the multistage processes. Current work involves solution of various types of problems that arise in multistage process work, in order to illustrate the methods involved. For the most part the methods are steady-state solutions.

STABILITY OF PERFORATED-PLATE TRAYS

Robert S. Brown, Donald N. Hanson, and C. R. Wilke

This work has been completed and is the subject of a separate report.¹

HEAT TRANSFER BETWEEN IMMISCIBLE FLUIDS

Sidney B. Lang, Victor L. Ledesma, and C. R. Wilke

Basic equipment was constructed for the studies of direct-contact heat transfer from an organic liquid to water, resulting in the vaporization

¹Robert S. Brown, Bubbling from Perforated Plates (thesis), UCRL-8558, Dec. 1958.

of a portion of the water and of the separation of the organic phase from the residual water. Monsanto Arclor 1248, a chlorinated diphenyl, was selected as the organic phase because of its low volatility, high specific gravity, and relatively low cost. The heat-transfer coefficients obtained with the equipment were very high. Future work will include a study of direct-contact heat transfer between condensing steam and an organic phase, and a more detailed study of separation of liquid phases by settling.

CONDUCTIVITIES OF MULTIPHASE MIXTURES

Robert E. Meredith and Charles W. Tobias

Conductivity studies of two-phase mixtures indicate that the effects of shape, size distribution, and surface resistance of the dispersed particle cannot be neglected in accurate conductivity measurements. In a study of dilute emulsions where the electrical conductivity of the dispersed phase was larger than the conductivity of the continuous phase, a "surface resistance" has been observed. The effect of this phenomenon has been overcome by making conductivity measurements at frequencies ranging from 10 to 100 kc instead of the customary 1-kc value.

MASS TRANSFER AT HORIZONTAL SURFACES IN FREE CONVECTION

Eugene J. Fenech and Charles W. Tobias

In all previous experimental work (already reported¹), the rectangular electrodes were limited on two sides by the wall of the electrolysis vessel. At present, experiments are performed with surfaces "free" on all sides. Preliminary results with circular and rectangular geometry indicate that the dependence of mass-transfer coefficients on process variables has been correctly represented by the correlation presented in the above report.

Differential equations for a simple model of the physical processes involved have been set up. Since exact solutions are impossible, numerical solutions will be attempted to see if an agreement with experiment can be found.

ELECTROCHEMICAL STUDIES IN CYCLIC CARBONATE ESTERS

William S. Harris and Charles W. Tobias

A report has been issued on this subject: William S. Harris, Electrochemical Studies in Cyclic Esters (thesis), UCRL-8381, July 1958.

¹Eugene J. Fenech, Correlation of Limiting Currents of Horizontal Electrodes Under Free-Convection Conditions (M. S. Thesis), UCRL-3586, Jan. 1956.

FORCED CONVECTION BOILING IN TUBES

Roger M. Wright, Robert Sani, and Leroy A. Bromley

A series of runs has been completed on the initial boiling-test section. This section consisted of a copper-finned tube (3/4 inch i.d., 0.110-inch wall thickness) in a steam jacket. Difficulty in accurately determining heat flux as a function of position along the tube has led to conversion of the boiling-test section to electrical heating.

The electrically heated test sections are 6-foot-long 3/4-inch (o.d.) 304 stainless steel tubes with two wall thicknesses: 0.016 and 0.020 inch. The electrical power is supplied by a General Electric 25-kva motor-driven induction voltage regulator connected to an air-cooled step-down transformer capable of giving 825 amperes at 40 volts. The range of relatively small vapor fractions is being studied to determine heat-transfer coefficients and pressure drops with greater accuracy than possible with the copper-finned tube section. Heat flux and inside wall temperatures are evaluated from thermocouple readings at the well-insulated outside wall. Uniform heat generation is assumed with the application of appropriate corrections.

Now in design is a steam-jacketed thick-wall copper test section (3/4-inch wall, 3/4-inch i.d.) to be constructed in 6-inch sections. Thermocouple installation will give inside and outside wall temperatures and heat flux throughout the full 6-foot length of the section.

LIQUID-LIQUID EXTRACTION AND AGITATION:
PERFORMANCE OF PACKED EXTRACTION COLUMNS

John E. Cotter, Norman Li, and Theodore Vermeulen

Axial-mixing measurements for two-phase countercurrent flow have been made with water-kerosene and water-mineral oil systems, in columns packed randomly with 1/2-inch Raschig rings and with 3/8-inch stoneware spheres. Results for the continuous phase confirm earlier data by G. Jacques, and indicate that the dimensional term,

$$\frac{(\text{particle diameter})(\text{dispersed-phase superficial velocity})}{(\text{sphericity})(\text{continuous-phase superficial velocity})}$$

can be used as a correlating factor. For the dispersed phase, different results are obtained, depending upon whether or not this phase wets the packing; but the Peclet number (ratio of particle diameter to mixing length) is always substantially less than in single-phase flow.

Apparatus modifications have been designed which will allow steady-state extraction runs to be made for checking the theoretical and experimental work that has been done so far. A sampling probe has been developed for selective removal of organic phase or aqueous phase from within the packing, and a manifold has been constructed for applying vacuum to an entire set of sampling lines. Also, phase-equilibrium measurements have been made for the purpose of selecting suitable solvent-solute systems for the extraction runs.

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