MASSACHUSETTS INSTITUTE OF TECHNOLOGY

Laboratory for Nuclear Science

PROGRESS REPORT

August 31, 1957
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This is the fiftieth progress report of the Laboratory for Nuclear Science at the Massachusetts Institute of Technology. Progress during the period June 1, 1957 through August 31, 1957 is recorded.

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Reprints of published papers listed herein are available in limited numbers upon request from the Laboratory for Nuclear Science Document Room.
CHEMISTRY OF THE FISSION ELEMENTS GROUP

I. Electrodeposition Behavior of Trace Amounts of Elements

The deposition of nickel onto a mercury pool usually takes place only after applying a relatively large overvoltage and then is virtually irreversible. From stirred acidic solutions of the alkali metal halides, the deposition of nickel has been found to follow first-order kinetics and to be independent of the initial concentration in the range from $10^{-3}$ to $10^{-7}$ M. However, the reduction process is a complex one as indicated by analyses of the relationship between rate of deposition and electrode potential. Though the reduction may proceed through monovalent nickel, as suggested by previous investigators, new evidence appears to favor the existence of two competing paths for the reduction of divalent nickel. For example, an increase in ionic strength caused the first reduction process to decrease markedly without affecting appreciably the second process. A closer examination of depositions from other media, particularly of other halides, would thus appear to be desirable. (C. F. Morrison, Jr.)

II. Rates of Irreversible Polarographic Reactions

Studies involving carbon tetrachloride, dibromoacetic acid and p-dinitrobenzene have clearly demonstrated the effect of a charge in the electrical double layer on the halfwave potential. Each of the above substances has two waves and, by control of pH and similar variables, the electrocapillary maximum can be made to fall between them. Then, by adding potassium thiocyanate, the first wave can be shifted to more cathodic potentials without affecting the second. By adding tetramethylammonium chloride, the second wave can be shifted to less cathodic values without affecting the first. As expected, addition of a mixture of the two salts moves the waves toward one another. Because shifts greater than 100 mv can be produced in this way, small variations in the composition of the supporting electrolytes can lead to serious discrepancies in the values of halfwave potentials reported by different investigators. (L. E. I. Hummelstedt)

Evidence has also been accumulated to show that the transfer coefficient is affected in a systematic way by changes in the double layer. Detailed results of this and the preceding study are being written up for publication. (W. H. Reinmuth)
III. pH Measurements Using a Palladium-Black Electrode

A palladium-palladium black electrode is being compared with a standard hydrogen electrode in a variety of solution. In addition to exhibiting a constant difference in potential from its platinum counterpart, the palladium electrode appears to be more readily polarized, especially in solutions containing substantial percentages of ethyl alcohol. However, unless more serious polarization effects are encountered, the palladium electrode would appear to be a useful substitute that might prove to be more versatile than the standard platinum form because the permeability of palladium to hydrogen makes possible a more convenient design of the electrode. (R. F. Breese)

IV. Luminescence of Chelates

A study of the effect of changes in structure of the chelating agent on the fluorescence spectrum is being undertaken using derivatives of 8-quinolinol. It is hoped that this will facilitate the search for improved reagents for determining traces of elements. (O. Popovych)

V. Thermometric Titrations

Work on thermometric acid-base titrations in the solvent acetonitrile has been continued. Hydrobromic acid as a titrant has proved to be quite satisfactory. A survey of the titration behavior of a wide variety of amines has been concluded. Application of the Taft equation to correlate heat of reaction and ionization constant with structural factors has shown that entropy effects attributable to solvation are much less important in acetonitrile than in water.

Diphenylguanadine in acetonitrile appears to be the most satisfactory titrant for acids, and a survey has been made of the behavior of acids of various structure and strength when titrated thermometrically with this reagent. It is interesting that the strong acids do not all show closely similar heats of reaction with diphenylguanadine nor is the order of conventionally-accepted acid strengths the same as the order in which these acids fall when they are rated according to the heat of reaction.

Efforts are being made to identify the various species in the neutralization solutions by means of infra-red spectrophotometry. (E. J. Forman)

VI. Optical Instrumentation

A working model of an automatic ratio recording spectrophotometer utilizing matched logarithmic amplifiers and multiplier phototubes has been constructed. Tests are under way to
Chemistry of the Fission Elements Group

determine the practicability of this instrument  (R. F. Breese)

VII. Photometric Titration

Efforts are being made to correlate and explain anomalous features of photometric titration curves in glacial acetic acid with the new theory of Kolthoff and Bruckenstein concerning the nature of acid-base reactions in glacial acetic acid. An infra-red spectrophotometric study is being made in an effort to identify some of the intermediate species believed to be formed in neutralization reactions. (L. E. I. Hummelstedt)

VIII. Flame Photometry

Preparations are being made for a systematic study of multiple factor interference effects in flame photometry. Apparatus has been assembled and built consisting of a Beckman DU spectrophotometer as the optical system, a Beckman hydrogen-oxygen burner as the flame source, and a specially designed photomultiplier detector system with automatic recording of luminescence on a potentiometer recorder. Provision is made for automatic wavelength drive so that flame spectra may be scanned and recorded automatically. Preliminary experiments to determine the sensitivity and reliability of the apparatus are under way. The first system to be studied will be the mutual interference effects of the lighter alkali metals. (W. H. Foster)

IX. Fused Salt Systems

A study of molten salt systems as solvents for analytical operations is being initiated. Equipment is being designed, built and gathered for operations in temperatures up to about 900°C. The sodium chloride-potassium chloride eutectic melt has been selected as the first system for investigation and exploratory polarographic investigations are getting under way. (D. L. Maricle)

X. Millicoulometry

The determination of the number of electrons in a polarographic reaction by direct coulometric determination of the amount of the electricity used and the change in concentration as evidenced by the change in diffusion current during a run has been suggested by a number of workers. Although this technique has been successfully applied in a number of cases, serious departure from expected behavior has been observed in others. Some of the largest discrepancies have been observed with particularly well-known and well-behaved polarographic reactions, such as the reduction of cadmium ion to the metal in a chloride solution. An investigation has been initiated to
Chemistry of the Fission Elements Group

determine the basic factors affecting the behavior of millicoulometric processes.

A simplified version of the copper-stripping coulometer of Ehlers and Sease has been developed and its use extended to low-current ranges. Extensive testing in the 5 to 50 microampere range has shown the coulometer to have an efficiency of 98.5 ± 0.5%. Millicoulometric cells consisting of a dropping electrode and a silver chloride anode, accommodating 0.25 ml. of solution have been built and test runs begun. Previous observations that the apparent number of electrons involved in the reduction of the cadmium ion varied from 1 to as high as 3 with potassium chloride concentration have been confirmed. (Barry Miller)

ADDRESSSES


PUBLICATIONS


D. N. Hume
L. B. Rogers
NUCLEAR CHEMISTRY (INORGANIC) GROUP

I. Introduction

Considerable attention has been given to the problem of ionization of mineral acids and hydrogen halometallates in inorganic solvents. The high spots of this are reported below, and full treatment is in draft manuscript or papers submitted for publication. About equal attention has been given to the problem of the anion exchange behavior of metal complexes. A series of seven working seminars was held under the sponsorship of Coryell, Marcus, Poskanzer, Freeman and Leifer, with informal notes available for some of the meetings. The Marcus-Coryell treatment was extended and is now ready for publication.

In mid-June Professor Irvine started sabbatical leave as Scientific Observer for the Office of Naval Research in London. During the summer a large group of people completed their assignments and left for other laboratories. (C. D. Coryell and L. Leifer)

II. Physico-Chemical Studies

A. Freezing-Point Behavior of Dowex 50-X10-HCl-H₂O Systems

Experiments have been continued on the temperature-time dependence during cooling of the cation exchange resin Dowex 50-X10 in the hydrogen form as reported by Leifer and Lundell.¹

Phase transitions have been observed in resin systems which have been equilibrated (and subsequently filtered, flash washed, and air dried) with nominal external HCl concentrations of 9, 6, 3, 1, and 0.1 molar. The total water, milliequivalents of HCl, and resin capacity have been determined on each sample. If one assumes that these observed phase transitions in the resin system are caused by separations of ice or HCl hydrate, the effective composition of the HCl solution within the resin phase may be determined from the freezing point. This work is being extended to lower HCl concentrations and other systems and resins in the hope of determining more precisely the nature of the resin phase in ion exchange systems. (L. Leifer and K. Kreiselmaier)

B. Formation Constants of Metal Complexes Determined by Anion Exchange

The original treatment by Coryell and Marcus² has been examined and revised. The presently accepted form is based on the following assumptions:

1. \( \gamma_\pm = \gamma_\pm = \gamma_{\pm} \) in a 1:1 electrolyte, irrespective of its concentration.

2. \( \gamma_{\text{MA}_{n-1}} \gamma_{\text{MA}_n} \) is a constant in the range of concentrations where \( \text{MA}_{n-1} \) is being converted into \( \text{MA}_n \).
3. $\gamma_- = \gamma_+ = \gamma_\pm$ in the resin phase.

4. $\gamma_B/\gamma_B'$ is constant, where $B$ is a simple monovalent anion in trace amount in a resin RA.

5. $\gamma MA_m + p/\gamma MA_m$ is constant.

Here $\gamma$ is the activity coefficient of the species indicated, $M^{m+}$ is a complex-forming metal cation, $A^-$ a monovalent ligand anion, and $p$ is the number of resin sites occupied by the anion complex.

The letter $r$ as presubscript identifies terms for the resin phase.

Using the following definitions:

$$a = (A)\gamma_+$$
$$k_n = (MA_n)/(MA_{n-1})^a$$
$$\beta_n = k_1 k_2 \ldots k_n$$

$$m = \frac{d \log \sum \beta_n a^n}{d \log a}$$
$$D = r C_M / C_M$$
$$C_M = (M) \sum \beta_n a^n$$

and taking $r C_M = (MA_m + p)_r$, the following equations are derived:

6. $\log r a = \log a + 1/2 \log (r A) - 1/2 \log (r C)$ for a resin RA in equilibrium with the salt of the complex former CA.

7. $rF_a = \log r a - \log r a^0$ is a useful function to correct for invasion. The term $\log r a^0$ is $\log r a$ at $a = 1$.

8. $\log D_B = \log (r B)/(B) = b \log r a - b \log a + \log \gamma_B - \log r \gamma_B$.

9. $d (\log D_B - b r F_a)/d \log a = -b$ is obtained for the slope.

10. $\log D = \log r k_p + p \log r a^0 + \log \beta_m + m \log a - \log \sum \beta_n a^n + p r F_a$, where $r k_p = r (MA_m + p)/r (MA_m) r a^p$ is a constant.

11. $d \log D_0/d \log a = d (\log D - p r F_a) \gamma d \log a = m - \bar{n}$.

In case $A$ is a polyvalent anion, modification in the assumptions (1) to (5) will be necessary, but the final equation obtained is similar to (10) (here, however, $m$ is replaced by the ratio of charges of metal to ligand).

12. $\log D = (\log k_\alpha + (p - m) \log r a^0 + \log \beta_p) + m \log a - \log \sum \beta_n a^n + (p - m) r F_a$, where $k_\alpha = r (MA_p). a^{p-v}/(MA_p). a^{p-m}$ is assumed constant, and then the term in parenthesis in equation (12) is constant.

This holds irrespective of $m$ being integral or non-integral.

If two species are important in the resin, $r C_M = r (MA_m + p) + r (MA_m + q)$, say, then

13. $\log D = \log k_p + \log (1 + k q . r a^{q-p}) + p \log r a^0 + \log \beta_m + m \log a - \log \sum \beta_n a^n + p r F_a$, where $k q = r (MA_v + q)/r (MA_v + p) \ r a^{q-p}$ is assumed constant.
These equations were applied to data for a considerable number of systems of metal tracer HCl with anion exchange resin with results shown in Table 2.1 for the principle complexes in solutions of different HCl concentration. The first column gives the metals and oxidation number, the order being that of atomic number. The second column gives the assumed principle species \( R_m^p MCl_m + p \). The third column gives the identities of the complexes predominating. It will be noted that in very concentrated HCl (near 12 M), some metals show formation of unionized metal complex acids, such as \( HZnCl_4^- \) or \( HGaCl_4^- \).

**TABLE 2.1**

Chloride Species Found by Anion Exchange

<table>
<thead>
<tr>
<th>Cation</th>
<th>Resin Species</th>
<th>Hydrochloric Acid Concentrations</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>0.1 M</td>
</tr>
<tr>
<td>Fe III</td>
<td>FeCl₃</td>
<td>Fe³⁺</td>
</tr>
<tr>
<td>Co II</td>
<td>R₂CoCl₄</td>
<td>Co²⁺</td>
</tr>
<tr>
<td>Cu II</td>
<td>R₂CuCl₄</td>
<td>Cu²⁺</td>
</tr>
<tr>
<td>Zn II</td>
<td>R₂ZnCl₄</td>
<td>Zn²⁺</td>
</tr>
<tr>
<td>Ga III</td>
<td>R GaCl₄</td>
<td>Ga³⁺</td>
</tr>
<tr>
<td>Ge IV</td>
<td>R₂GeCl₆</td>
<td></td>
</tr>
<tr>
<td>As III</td>
<td>R AsCl₄⁻</td>
<td>AsCl₃⁻</td>
</tr>
<tr>
<td>As V</td>
<td>R AsCl₄⁻</td>
<td>AsCl₃⁻</td>
</tr>
<tr>
<td>MoO_{2+}</td>
<td>R MoO₂Cl₃</td>
<td></td>
</tr>
<tr>
<td>Pd IV</td>
<td>R₂PdCl₄</td>
<td>PdCl₄⁻</td>
</tr>
<tr>
<td>Ag I</td>
<td>R₂AgCl₉</td>
<td>AgCl₂⁻</td>
</tr>
<tr>
<td>Cd II</td>
<td>R₂CdCl₄</td>
<td>CdCl⁺</td>
</tr>
<tr>
<td>In III</td>
<td>R InCl₄</td>
<td>InCl₂⁺</td>
</tr>
<tr>
<td>Sn IV</td>
<td>R₂SnCl₆</td>
<td></td>
</tr>
<tr>
<td>Pt IV</td>
<td>R₂PtCl₆</td>
<td>PtCl₆⁻</td>
</tr>
<tr>
<td>Au III</td>
<td>R AuCl₄</td>
<td>AuCl₄⁻</td>
</tr>
<tr>
<td>Tl I</td>
<td>R₂TlCl₃</td>
<td>TlCl⁺</td>
</tr>
<tr>
<td>Tl III</td>
<td>R₂TlCl₄</td>
<td>TlCl₄⁻</td>
</tr>
<tr>
<td>Pb II</td>
<td>R₂PbCl₄</td>
<td>PbCl⁺</td>
</tr>
<tr>
<td>Bi III</td>
<td>R₂BiCl₅, R₃BiCl₆</td>
<td>BiCl₅⁻</td>
</tr>
<tr>
<td>UO_{2+}</td>
<td>R UO₂Cl₃</td>
<td>UO₂⁺</td>
</tr>
</tbody>
</table>

The results of this treatment will be submitted for publication soon. Reference may also be made to the following papers: Y. Marcus, Bull. Res. Council Israel 4, 326 (1954); ibid. 5A 177 (1956); Acta Chem. Scand. 11, 619 (1957).
C. The Anion Exchange of Metal Complexes: the Cadmium-Chloride System

Distribution data for cadmium between Dowex 1 anion-exchange resin and solutions of HCl and LiCl in the range of 0.01 - 9 M were obtained with the aid of tracer Cd$^{115}$. The batch method was used, and it was observed that the distribution coefficient is independent of Cd concentration in the range 5.10$^{-5}$ to 1.4 · 10$^{-3}$ M, signifying strictly mononuclear complex formation. The distribution curves for HCl and LiCl are identical up to 0.2 M chloride, thereafter the HCl curve is lower, but both curves exhibit a maximum at about 3 M chloride. The invasion of the resin by the HCl and LiCl electrolytes was taken account of to give ideal distribution curves, valid for constant resin-ligand activity. These latter curves were evaluated by the method of Marcus and Coryell (see section II B above) in terms of the stepwise formation of the complexes CdCl$^+$, CdCl$_2^-$, CdCl$_4^{2-}$, and the additional complex HCdCl$_3$ in HCl solutions. The successive formation constants found are: $k_1 = 90$, $k_2 = 3.5$, $k_3 = 0.7$, $k_4 = 0.2$ and the acidity constant of HCdCl$_3$ $k_a = 1.0$ are in good agreement with available published values (e.g. Riley, Leden, King, Vanderzee) (Y. Marcus)

D. The Ionization Constants in Wet Bis(2-chloroethyl) Ether of Mineral Acids

Radioactive nuclides prepared without the use of added carrier offer the opportunity to study the physicochemical behavior of substances down to the concentrations in the range of 10$^{-12}$ molar, but there are many serious technical difficulties in the way of achieving reliable results. Special studies have been made of conditions to obtain reproducible results with bis(2-chloroethyl) ether, and experiments have been carried out with Au(II) and Ga(III) in HCl solutions and with In(III) in HCl and HBr solutions. At fixed acid concentration, the distribution ratio $D$ (organic/aqueous) rises as the metal concentration $M$ is decreased. These effects are ascribed to the fact that the metals under consideration are extracted as HMX$_4^-$, a stronger acid in the organic solvent than the supporting electrolyte HX. A complete theory has been presented for the dependence of $D$ on $M$ at fixed HX, with parameters representing the acid ionization constants $K$ of HMX and HX in the organic solvent. At very low $M$, the main metal species in the organic solvent is MX$_4^-; at increasing M, the predominant form is HMX$_4$, and the transition depends critically on the level of H$^+$ produced by HX. The distribution data provide values for $-\log K = pK$ for HCl and HBr of 6.7 ± 0.2 and 5.6 ± 0.2.

Independent determination of these values is possible from careful measurement of the conductivities of the acids in the wet solvent, bis(2-chloroethyl) ether. Measurements reported earlier have been modified by extensions of the work and reanalysis to give the value for HClO$_4$, $pK = 4.0 ± 0.2$. The other acids were too weak to permit direct determination of the limiting conductance. It was assumed that the acids have the same value as HClO$_4$, so a value of $K$ could be determined for each point, using Debye-Hückel activity coefficients. For HCl the average of 18 points gave $pK = 7.2 ± 0.2$ in good agreement with the extraction value. For HBr there was a trend in K values; the best fit at low concentration corresponds to $pK \sim 7.0$. In addition a rough value of $pK = 4.0$ has been set for HGaCl$_4$. 

-8-
A preliminary report on this constellation of studies has been prepared for the September 1957 UNESCO Conference in Paris on Radioisotopes in Scientific Research, and a detailed report is in preparation. In addition, two papers on the extraction studies have been prepared for the 132nd Meeting of the American Chemical Society in New York. (A. M. Poskanzer, R. J. Dietz, Jr., E. Rudzitis, J. W. Irvine, Jr., and C. D. Coryell)

E. Some Two-Center Overlap Integrals with AO's of Principal Quantum Number Four

Numerical values of thirteen types of two center overlap integrals, involving Slater atomic orbitals of principal quantum number four, have been computed for a wide range of molecular parameters. A table of the integrals $A_k(p)$ where $k$ is non-integral is also given for use in computing these and other overlap integrals.

This work will be submitted for publication in the Journal of Chemical Physics. (L. Leifer, F. A. Cotton, and J. R. Leto)

III. Solvent-Extraction Studies

A. Solubility and Solvent Distribution of As(III) Iodide

Carrier-free As$^{74}$ activity and an As(III) stock solution were prepared. The distribution of As(III) between 2 to 7 M HI and the solvents C$_6$H$_6$, CHCl$_3$, and CCl$_4$ was measured by equilibrating the aqueous and the organic phases for 1 hour at 30.0 ± 0.5°C. Parallel curves, with a maximum at ca. 3 M were obtained by plotting the logarithm of the distribution coefficient versus the HI concentration. The solubility of AsI$_3$, made radioactive by incorporating the tracer As$^{74}$, in HI was measured by precipitating the AsI$_3$ from its solution in water by HI, to give the appropriate concentration of HI. The solubility showed a minimum at 3 M HI.

Results are published more fully in a paper by Brink, Kafalas, Sharp, Weiss and Irvine. (E. L. Weiss)

IV. Radiochemical Separation

Determination of Total Polonium in Bone

Several procedures for the determination of polonium in bone have been proposed. Most of these have been summarized in a recent paper by S. Black. For the purposes of the M.I.T. Radioactivity Center several modifications seemed advisable. The procedure currently being investigated is:

1. To 20 gm of bone in a 250-ml Erlenmeyer flask, add 100 ml of conc. HNO$_3$, and let stand overnight.
Nuclear Chemistry (Inorganic) Group

2. Add anti-foamant and boil down until mixture has the consistency of a thick paste.
3. Add 10 ml of 30% H$_2$O$_2$, and boil gently for 10 minutes.
4. Add formic acid slowly with heating to 90°C until 1 ml can be added without producing brown fumes.
5. Add 20 ml of 6N HCl and boil solution until total volume is 20 ml.
6. Cool, transfer liquid to a 50-ml separatory funnel, and extract fat with 20 ml of petroleum ether.
7. Return the aqueous layer to the Erlenmeyer flask, dilute with distilled water to 100 ml, and heat until sample dissolves.
8. Add 100 mgm of ascorbic acid. Transfer to plating cell and with constant stirring and heating to 90°C plate on silver foil for 2 hours.

The total polonium recovery by this procedure is believed to exceed 90%. (C. E. Gleit)

V. General Nuclear Chemistry

A. Instrumentation

A new P-10 gas line is being installed to provide a more constant flow of gas to the Los Alamos-Sugarman type proportional counters. The main line, which extends along two walls of the counting room, is 1/4" O.D. hard copper tubing. The gas tank is connected to this by a blunt needle bellows valve to allow the system to be pumped out. There are 9-1/8" metering valves tapping the line through tee connections and adapters. From these, 1/8" soft copper tubing connects to the counters. Provision was made for future expansion by capping both ends of the main line with sweat caps. One of the metering valves is designed for attaching a pump or temporary counter to the system.

A 20-channel analyzer, owned by the M.I.T. Physics Department, has been connected to a scintillation counter. As detectors a pair of NaI(Tl) crystals 1" by 1" and 2" by 1/4" purchased from National Radiac Inc. have been employed. This apparatus has been used for energy determinations in several of the following reports. (L. H. Bowen)

B. Search for Ga$^{74}$

It was reported$^8$ that an 8-min $\gamma$-active species was formed by fast neutron bombardment of Ge metal. No chemical separation was made, but the activity was ascribed to Ga$^{74}$. Possible reactions for the formation of Ga$^{74}$ are:

Ge$^{74}$(n, p)Ga$^{74}$ and Ge$^{76}$(d, $\alpha$)Ga$^{74}$.

Since the elements As, Ge, Ga, and Zn will be found in Ge irradiated with deuterons or neutrons, a separation method is necessary. The following procedure gives a good separation:

Ge metal (powdered) after irradiation is dissolved in HF + HNO$_3$, evaporated to dryness, and fumed with conc. HCl to remove the bulk of the Ge as GeCl$_4$. The residue is dissolved in conc.
Nuclear Chemistry (Inorganic) Group

HCl, reduced with a few crystals of NaHSO₃, and extracted three times with benzene, to remove all AsCl₃ and GeCl₄. If Zn is present, the Ga is extracted into bis(2-chloroethyl) ether and re-extracted into 2M HCl. In all cases, the Ga contained in the HCl solution is precipitated by NH₄OH after addition of 5 mg of Fe(III) carrier. The precipitate is filtered, washed, dried, and counted. The time required for the procedure is six to eight minutes, depending on the amount of Ge to be dissolved and fumed away.

The basis of the procedure is the low distribution coefficient D of GaCl₃ between benzene or chloroform and HCl, compared to GeCl₄ and AsCl₃. While for 2 and 4 M HCl, D is 10⁻³ to 10⁻² for Ga, Ge, and As(III), D increases to about 10² or 10¹ for 6-12 M HCl for Ge and As(III)⁶, while D remains about 10⁻² for Ga. Experiments using radioactive tracers of Ge and As showed that these elements do not accompany Ga in the hydroxide precipitate obtained in the procedure outlined above.

The results of cyclotron bombardment experiments on Ge metal with the nuclides identified by their β-decay curves using an end-window, gas-flow proportional counter are presented in Table 2.II.

### TABLE 2.II

<table>
<thead>
<tr>
<th>Projectile</th>
<th>Time of Irradiation</th>
<th>Time before Counting</th>
<th>Elements Separated</th>
<th>Half-lives Found (β)</th>
</tr>
</thead>
<tbody>
<tr>
<td>n (~15 Mev)</td>
<td>10 min</td>
<td>17 min</td>
<td>Ga</td>
<td>Long, 22 ± 0.5 min</td>
</tr>
<tr>
<td>d (14 Mev)</td>
<td>3 min</td>
<td>15 min</td>
<td>Ga</td>
<td>Long ? , 68±2 min, 22±1 min</td>
</tr>
<tr>
<td>d (14 Mev)</td>
<td>5 min</td>
<td>16 min</td>
<td>Ga + As</td>
<td>~28 hr, 80±10 min, 22±1 min</td>
</tr>
<tr>
<td>n (~15 Mev)</td>
<td>15 min</td>
<td>22 min</td>
<td>Ga</td>
<td>14.1±0.3 h, 21±0.5 min</td>
</tr>
<tr>
<td>fast n (~15 Mev)</td>
<td>1.5 min</td>
<td>8.5 min</td>
<td>Zn+Ga+Ge+As</td>
<td>~5 hr, ~80 min, 22±1 min, 1.2 min (?)</td>
</tr>
<tr>
<td>n (~10 Mev)</td>
<td>3 min</td>
<td>1 min</td>
<td>Zn+Ga+Ge+As</td>
<td>~19 hr, 83±3 min, 19±2 min, 1.2±0.5 min</td>
</tr>
</tbody>
</table>

All half-lives found can be ascribed to known nuclides [(viz. ~28 hr: As⁷²(26 hr), As⁷⁶ (26.7 hr); ~19 hr: As⁷², As⁷⁶, Ge⁷⁷(12 h); 14.1 ± 0.3 h: Ga²⁶(14.1 h); ~5 h: Ga⁷³(5 h); 80 ± 10 min: As⁷⁸(90), Ga⁶⁸(68 min); ~80 and 83 ± 3 min also Ge⁷⁷(82 min); 68 ± 2 min: Ga⁶⁸; 22 ± 1, 21 ± 0.5, and 19 ± 2 min: Ga⁷⁰(21 min)] except the short one: 1.2 ± 0.5 min. No evidence for an 8 min β activity could be found. An 8 min γ activity without 8 min β activity would then correspond to an isomeric transition, not necessarily of Ga⁷⁴, although Ga⁷⁴ could have a half-life of 1.2 min.

More experiments concerning the postulated 8 min β activity and eventual confirmation of an 8 min γ activity are planned. Also a faster separation of Ga will be tried, in order to see whether the 1.2 min activity belongs to Ga. (Y. Marcus)
Nuclear Chemistry (Inorganic) Group

C. Study of Short-Lived Isomers of K$_{38}$

A study concerning the existence of a short-lived isomer of K$_{38}$ has been initiated. This is related to work concerning the possible presence of a long-lived K$_{38}$ with a half-life of approximately 10$^9$ years, performed by J. W. Winchester. Recently, Gerling suggested the existence of long-lived K$_{38}$ in natural potassium. On the other hand, Stähelin reported isomers with half-lives of 0.95 sec and 7.7 min produced by K$_{39}$(γ, n)K$_{38}$. Green failed to detect the shorter of these by K(p, pn)K$_{38}$. Others indicated the presence of K$_{37}$, $T_{1/2} = 1.2$ seconds, from the reaction K$_{39}$(γ, 2n)K$_{37}$ and the reaction Ca$_{40}$(p, α)K$_{37}$. Thus at present we have three possible species of K$_{38}$ and two of these are poorly characterized.

For the present study two series of experiments have been performed to characterize short lived K$_{38}$:

1. The reaction Ca$_{40}$(d, α)K$_{38}$.
   a) 200 mg of CaCO$_3$, prepared from CaCl$_2$ and Na$_2$CO$_3$, was bombarded for 4 min with 15-Mev deuterons. The target was digested with water containing 0.05 m mole KOH, and passed through a double filter. From the first filter the Ca and Sc activities were removed with CaCO$_3$. Approximately 200 mg of colloidal Dowex 50 in the potassium form, sandwiched between the two filters, adsorbed the potassium activity. A half life of 8.1 min was obtained by β counting. However, Na$^{24}$ activity due to Na impurities appreciably masked the K$^{38}$ activity.

b) Calcium metal plate, obtained from Metal Hydrides, Inc. was dissolved in hot water containing a trace of HCl, 0.2 mmole of KOH and a slight excess of Na$_2$CO$_3$ were added, precipitating the Ca. The filtrate was treated with an acid solution of tetraphenyl borate to precipitate the potassium. Sixteen minutes elapsed between the end of bombardment and the commencement of counting. A half-life of 7.74 minutes was observed for more than 10 half lives. Relative yields of the two K activities, $\sigma^{38}/\sigma^{42}$, were determined as 0.55. Absolute yield of K$^{42}$ was slightly greater than 3μc./μah. That is, the yield of K$^{38}$ is the order of 7 nuclide/10$^8$ deuterons from natural calcium.

2. The reaction Cl$_{38}$(α, n)K$_{38}$.

Fused NaCl was bombarded with 30-Mev α particles for 1/4 min at 0.7 μa and counted on an end-window β counter. Half lives of 7.80 minutes (K$^{38}$), 33 minutes (Cl$^{38}$ or Cl$^{34}$?), and approximately 18 hrs (Na$^{28}$) were found. The ratio of activities at the end of bombardment was 970/30/1. The absolute yield of K$^{38}$ was estimated as approximately 1 nuclide/10$^8$ α particles. With this reaction, it is hoped that the 1-sec isomer of K$^{38}$ may be detected. For this purpose a scattering chamber and a tape recorder used for rapid counting would be useful. (M. Honda)

D. Decay Schemes for Neutron-Rich Pd and Ag Isotopes

In the last annual report (Laboratory for Nuclear Science Annual Progress Report, May 1957), the decay properties of six new Pd and Ag species of half-period less than three minutes
were reported, together with more information on the decay properties of other known species in the mass region 113 through 117. The species of interest are listed in Table 2.111 with the new data of this study underlined.

**TABLE 2.111**

<table>
<thead>
<tr>
<th>Mass Number</th>
<th>T/2</th>
<th>Radiations, Energy, and Branching</th>
</tr>
</thead>
<tbody>
<tr>
<td>113</td>
<td>1.4 ± 0.1 min</td>
<td>( \beta ), no prominent ( \gamma )</td>
</tr>
</tbody>
</table>
| \( \text{Ag}^{113m} \) | 1.2 ± 0.15'' | \( \beta < 2.0 \text{ Mev} \) (10\% of equilibrium 5.3-hr \( \beta \))
|              |               | \( \gamma \) 0.14, 0.30, 0.39, 0.56, 0.70 Mev (intensities 40:100:30:10:10:1) |
| 113         | 5.3 ± 0.2 hr | \( \beta \) 2.2 Mev \( \gamma \)(~20\%) 0.30 Mev |
| \( \text{Cd}^{113} \) | 5.3 yr | 0.5 \% of 113 chain\(^b\) comes from 1.2-min \( \text{Ag}^{113m} \) |
| 114         | 2.4 ± 0.1 min | \( \beta \), no \( \gamma \) observed |
| \( \text{Ag}^{114} \) | 5 ± 2 sec | \( \beta \) 4.6 ± 0.4 Mev \( \gamma \) 0.56 |
| \( \text{Ag}^{114} \) (2 min) |               | \( \beta \), \( \gamma \), not observed in fission products\(^c\) |
| 115         | 45 ± 3 sec | \( \beta \), no prominent \( \gamma \) (~100\% decay to 20 sec \( \text{Ag}^{115m} \)) |
| \( \text{Ag}^{115m} \) | 20 ± 10 sec | \( \beta \), no prominent \( \gamma \) (28\%)\(^b\), I.T. (72\%)\(^b\) |
| \( \text{Ag}^{115} \) | 21.1 ± 0.5 min | \( \beta \) 2.9 ± 0.3 Mev, (9\% \( \beta \) decay to 43-day \( \text{Cd}^{115} \))\(^b\) \( \gamma \) 0.14, 0.23 both weak, intensities 1:5. |
| 116         | < 30 sec | \( \beta \) 5.0 ± 0.4 Mev |
| \( \text{Ag}^{116} \) | 2.5 min | \( \gamma \) 0.70, 0.515 (20:80) |
| 117         | < 30 sec | \( \beta \), \( \gamma \) |
| \( \text{Ag}^{117} \) | 1.1 ± 0.1 min | Gives\(^e\) both 3.0 hr \( \text{Cd}^{117m} \) and 50 min \( \text{Cd}^{117} \). |

\(^a\) Two alternative decay schemes A and B are proposed for 1.4-min \( \text{Pd}^{113} \), 1.2-min \( \text{Ag}^{113m} \), 5.3-hr \( \text{Ag}^{113} \); see text.

\(^b\) Branching deduced from fission yields of A. C. Wahl and N. Bonner, Phys. Rev. 85, 570 (1952).

\(^c\) Species reported by R. D. Duffield and J. D. Knight, Phys. Rev. 75, 1613 (1949).

\(^d\) Extractions of \( \text{Ag} \) from rapidly isolated \( \text{Pd} \) formed in deuteron fission give no evidence for \( \text{Pd}^{116} \) or \( \text{Pd}^{117} \) precursors.

\(^e\) Decay relations of \( \text{Cd}^{117m} \), \( \text{Cd}^{117} \), \( \text{In}^{117m} \) and \( \text{In}^{117} \) not yet clarified. Compare C. D. Coryell, H. G. Richter, and P. Lévéque, Phys. Rev. 105, 1763 (1951); C. L. McGinnis, ibid. 94, 371 (1954); C. E. Gleit, unpublished work at M.I.T.
It is attractive to present the rather large body of data in decay schemes, those for the successive mass numbers 113 through 116 being presented as Figures 2.1 through 2.4.

**Figure 2.1**
Alternatives for the decay scheme of Pd$^{113}$ and Ag$^{113}$.

**Figure 2.2**
Decay scheme of Pd$^{114}$ and Ag$^{114}$. 

-14-
Figure 2.3
Decay scheme of $^{46}\text{Pd}^{115}$ and $^{47}\text{Ag}^{115}$.

Figure 2.4
Decay scheme of 2.5-min $^{47}\text{Ag}^{116}$.
 Detailed support of these decay schemes is given in a paper being submitted to Physical Review. Conventional symbols are used. Hypothetical intermediate states or unobserved possible transitions are given as broken lines. Spin values are given at the left of each level, with energies in Mev on the right. Italicized figures are the log ft values for $\beta$ transitions, the $\sim$ sign denoting that estimated $\beta$-decay energies have been used. The $\gamma$-ray transitions are consistent with Weisskopf radiation theory, as modified by Way et al. The decay schemes find general support in the compilations of Goldhaber and Hill and Dzhelepov and Peker.

For mass 113 (Fig. 2.1), choice cannot yet be made between two alternates. Scheme A sets 1.4-min Pd$^{113}$ as 5/2+ and makes all of its decay go to 1.2-min Ag$^{113m}$ (7/2+) which decays 90% by isomeric transition. This is analogous to Pd and Ag level patterns for mass numbers 107 and 109. The alternative Scheme B makes an analogy with the 115 chain (Fig. 2.3) whereby the 1.4-min Pd$^{113}$ is 1/2+ or possibly 3/2+, and the 7/2+ and 1/2-levels of Ag are in the process of exchanging places over the pre-113 pattern, so that isomeric conversion is unimportant. The only difference in the $\beta$-decay patterns of the two Ag$^{113}$ isotopes is in the small branching to the 5.3 yr Cd$^{113m}$ (11/2-), 0.5% in Scheme A and 5% in Scheme B.

Figure 2.2 gives the decay scheme proposed for 2.4-min. Pd$^{114}$ $\rightarrow$ 5 sec Ag$^{114}$ $\rightarrow$ stable Cd$^{114}$. The 2-min Ag$^{114}$ was not observed as a $\beta$-decay product of Ag$^{114}$ presumably because of high spin, probably 5-.

Figure 2.3 is self-explanatory. The case for spin assignments of the Ag isotopes is good, and the high yield of $\sim$20-sec Ag$^{115m}$ (1/2-) sets the spin of 45-sec Pd$^{115}$ as 1/2+ or 3/2+.

The hypothetical decay of < 30 sec Pd$^{116}$ is not entered on Figure 2.4. There is no information on possible direct $\beta$ decay of 2.5-min Ag$^{116}$ to ground-state Cd$^{116}$. The log ft of 6.9 for the $\beta$ decay to the 0.51-Mev 2+ level in Cd$^{116}$ is based on assumption of negligible direct $\beta$ decay to the ground-state.

It is premature to draw a decay scheme for the 117 chain because of lack of information about decay relations among isomers at Cd$^{117}$ and at In$^{117}$. (C. D. Coryell, U. Schindewolf, and J. M. Alexander)

VI. Fission Radiochemistry

A. Investigation of the Isotopes of Cadmium and Indium Formed in Uranium Fission

The nuclides with masses 117 to 120 formed in the fission of uranium play an important part in the theory of fission. Two recent investigations of the 117 chain differ in several respects. A study of these nuclides is in progress.

In these experiments cadmium is extracted from an aqueous basic solution of uranium and mixed fission products containing hydroxylamine, tartrate, and cyanide into a chloroform
solution of dithizone. Cadmium is then back extracted with a dilute mineral acid and precipitated with tetraphenylarsonium chloride. Indium is extracted from cadmium in an 8N aqueous solution of HBr into bis(2-chloroethyl) ether. Indium is back extracted with 12N HCl and precipitated with 8-hydroxyquinoline. Beta, gamma, and beta-gamma coincidence counting has been performed to determine half-lives. Feather analysis and pulse height analysis using a scintillation counter and a 20-channel analyzer have been used to determine energies.

Preliminary results suggest several alterations to the published literature. The 30-min Cd\textsuperscript{118} should be 50 ± 2 min. A new isotope of cadmium, Cd\textsuperscript{119}, has been found. This nuclide has a half-life of approximately 2 minutes, and decays with β rays with maximum energy, of over 2 Mev into 18-min In\textsuperscript{119} and possibly a shorter lived isomer of this In\textsuperscript{119}. The 18-min In\textsuperscript{118} previously produced\textsuperscript{24} by Sn\textsuperscript{119}(γ, p)In\textsuperscript{119}, can now be listed as a fission product. (C. E. Gleit)

B. The Effect of Prompt Neutron Emission on the Fission Products of U\textsuperscript{235}

Kinetic energy data on the products of thermal neutron fission of U\textsuperscript{235} have been employed with a statistical model of the nucleus to predict\textsuperscript{25} the relative emission probabilities of the first five neutrons (prompt) from all nuclei in the range of high fission yield. Charge and mass yield curves are assumed for the initial products formed in fission before prompt neutron emission, and the effect of the neutron emission on these curves examined.

Data on the kinetic energy of the fragments from the fission of U\textsuperscript{235} indicate that there is a probability distribution\textsuperscript{26} of such kinetic energy for a given mass and charge pair formed. From a simple energy balance on the fission act, it is possible to conclude that there is necessarily a probability distribution of excitation energy for any pair of fragments and, considering the kinetic energy data, such a distribution is approximately Gaussian in nature. It is this excitation energy which gives rise to the emission of prompt neutrons observed in fission. It has been the purpose of this work to examine by means of a simple nuclear model the effect of such prompt neutron emission on the mass and charge distributions of the fission products of U\textsuperscript{235}, using the M.I.T. Whirlwind I Digital Computer.

It has been assumed that, at the excitations involved, the nucleus may be considered a statistical assemblage of nucleons. Such an assemblage will emit neutrons with a distribution of kinetic energy.

\[ p(\eta) = \gamma' \eta \exp(-\eta/T') \]

where \( \eta \) is the neutron kinetic energy, \( T' \) is an effective nuclear temperature, a function of the mass of the nucleus, and the excitation energy involved, and \( \gamma' \) is a normalization factor. If the distribution of excitation energy for a given nucleus and the separation energies of its first \( i \) neutrons are known, it is possible to derive equations for the probability of emission of 0, 1, 2, \ldots \( i \) neutrons from the nucleus. Such equations have been derived using the nuclear masses and neutron separation energies computed by Riddell\textsuperscript{27} from Levy's mass systematics.
In order to derive such equations, it is necessary to make some assumption as to how the excitation energy distribution for a nuclear pair (as derived from the kinetic energy data) is split between the two fragments. It has been assumed that such a split occurs in a random manner but that more or less of the energy can be placed on one of the fragments. Two basic computational runs were made with two different energy divisions, the first, Run A, based on the statistical theory of Fong in which 70% of the energy is placed on the heavy fragment, and the second, Run B, suggested by the experimental work of Fraser where 62.5% of the energy is placed on the light fragment.

There are no experimental data on the mass-yield or charge distribution of the products of fission before prompt neutron emission, although there are data concerning mass and charge yields after prompt neutron emission. It was therefore necessary to postulate fission yields before neutron emission, calculate the mass shift of such yields due to neutron emission, and compare the results of the calculations with experimental data.

The results of the calculations are presented in Figures 2.5 and 2.6. Figure 2.1 is a plot

![Figure 2.5](image-url)

Calculated mass distributions, runs A and B, compared to experimental (ref. 30).
of the final yield of all nuclei of mass $A$, $F_A$, versus the mass number $A$. The results of the two runs are given, and a line through the experimental data$^{30}$ presented for comparison. It will be noted that in the heavy mass range, the calculated yields are high directly below the region of 50-proton 82-neutron shell closure ($A = 130-134$). Such high calculated yields are interpreted as being due to poor choice of values of neutron separation energies in this region. In general the fine structure of the two runs is similar to that observed experimentally, and it is concluded that the observed mass yield fine structure is due to prompt neutron emission. The results are not of sufficient accuracy to determine the proper split of excitation energy between the light and heavy fragments.

Figure 2.5 shows the results for charge distribution from the light mass peak in Run B (the heavy peak and Run A results are similar). The ratio of yield of nuclide $(Z, A)$ to the total yield of all nuclides of mass $A$ is plotted against the distance in charge units from some most probable charge to be formed in fission, $Z_p$, for that mass. Values of $Z_p$ were determined by a technique similar to that of Pappas$^{31}$. Two separate curves result, one of higher yield for nuclei of even neutron number, and a second lower by the factor 1.5 for nuclei of odd neutron number. A curve through the experimental data$^{31}$ (all of which happens to be for odd neutron number nuclei)
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is included for comparison. The divergence in yield between even and odd neutron species is due to the lower separation energy of an unpaired neutron in a nucleus. It is of particular interest to note that no experimental single nuclide yield data are available for even neutron number species, and that there is yet no means of verifying the higher yields predicted here. Furthermore, it has been assumed that even neutron number species are not preferentially formed in fission. If they are, the resulting divergence of the two curves in Figure 2.6 will be greater.

It is concluded that the fine structure observed in the experimental fission product mass yield curve is in general due to prompt neutron emission, but also the fission act itself must give abnormally high yields in the region of the 82-neutron closed shell. Furthermore, it is concluded that there should be two charge distribution curves for fission products after prompt neutron emission, one for nuclei of even neutron number, and another showing lower yield of odd neutron species.

The effect of the relative excitation of the light and heavy mass fragments is examined in the light of experimental data and statistical theory, but the calculations are not of sufficient accuracy to allow choice between possible excitation ratios. Recommendations for further calculational work are included in the thesis.25 (A. E. Herrington)

REFERENCES


12. D. Green and J. R. Richardson, Phys. Rev., 101, 776 (1956); ibid, 96, 858A (1954); ibid,


    H. Walke, ibid. 51, 439 (1937).


    W. J. Henderson et al, ibid. 51, 1107 (1937).
    L. N. Ridenour and W. J. Henderson, ibid. 52, 889 (1937).
    M. M. Ramsay et al, ibid. 72, 639 (1947).

18. V. F. Weisskopf, Phys. Rev. 83, 1073 (1951);
    J. M. Blatt and V. F. Weisskopf, Theoretical Nuclear Physics, J. Wiley and Sons, New York
    1952.

    (1956).


21. B. S. Dzhelepov and L. K. Peker, "Decay Schemes of Radioactive Isotopes", Academy of
    Science, USSR 1957; translated and issued by Atomic Energy of Canada, Ltd., Report
    AEC 457.


30. E. P. Steinberg and L. E. Glendenin, International Conference on Peaceful Uses of Atomic
Nuclear Chemistry (Inorganic) Group


ADDRESS


PUBLICATIONS


THESIS

A. M. Poskanzer, "The Solvent Extraction of Inorganic Complexes", Ph.D. in the Department of Chemistry, August 1957.

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I. Introduction

Work on the mechanism of methanolysis of triphenylmethyl chloride in benzene solution has demonstrated that the rate-determining step is reaction of triphenylcarbonium ion with methanol, a result inconsistent with data and conclusions reported by Ingold and co-workers. Considerable progress has been made on a theory of the origin of isotope effects in light and heavy water, and further experimental data is being collected on alkyl halides and sulfonium salts to test this theory. Tritium is being used in isotope dilution studies of the mechanism of the Cannizzaro reaction. Isotope effects have demonstrated that the oxidation of isopropyl alcohol to acetone by bromine in water solution involves hydride rather than proton removal from carbon, whereas the decarboxylation of \( \beta \)-keto acids involves proton rather than hydride removal from the carboxyl group. Oxygen-18 is being used to find whether the carbonyl and peroxide oxygens of phthaloyl peroxide can be interchanged thermally. Carbon-14 is being used to study stereospecificity in free radical brominations of bibenzyl and \( p, p' \)-dinitrobibenzyl by N-bromosuccinimide.

II. Methanolysis of Triphenylmethyl Chloride

Work on the mechanism of methanolysis of triphenylmethyl chloride in benzene solution using tritium-labeled methanol has been completed. A paper has been written for publication in J. Am. Chem. Soc. Its summary follows.

The methanolysis of 0.1 M triphenylmethyl (trityl) chloride in benzene solution at 25\(^\circ\) has been studied from 5 \( \times \) 10\(^{-2}\) M methanol down to the very low concentration of 5 \( \times \) 10\(^{-7}\) M methanol by a tracer technique specific for the methanolysis reaction. The rate is practically independent of the nature or concentration of tertiary amine used to bind the hydrogen chloride produced. The total kinetic order decreases from approximately third at 5 \( \times \) 10\(^{-2}\) M to second at 10\(^{-3}\) M; below 10\(^{-3}\) M methanol it remains second order over-all (first order in both trityl chloride and methanol). These observations plus Kreevoy's earlier work on exchange with quaternary ammonium salts demonstrate that the mechanism below 10\(^{-3}\) M methanol is a reversible ionization of trityl chloride assisted by only the solvent benzene, followed by a rate-determining reaction of triphenylcarbonium chloride with methanol. The deuterium isotope effect \( k_{\text{CH}_3\text{OH}}/k_{\text{CH}_3\text{OD}} \) is greater than two at 0.05 M but close to unity below 10\(^{-3}\) M methanol. Serious experimental errors in papers by Hughes, Ingold, Mok, Patai and Pocker are noted and it is shown that the mechanisms proposed by these authors are unsound. (E. E. Pegues)
III. Mechanism of the Cannizzaro Reaction

During this period synthesis of a supply of benzaldehyde-p-t for kinetic measurements was completed. The material was purified by distillation in a Holzman column under flowing dry nitrogen, the fraction boiling at 177-178° being collected. The purity of this distillate was checked by measurement of the index of refraction at 25° and the reading obtained agreed very closely with the literature value.

The activity of the benzaldehyde-p-t was measured roughly with the scintillation counter by comparison with a sample of tritiated water of known activity. The activity of the benzaldehyde-p-t measured in this way was 0.38 millicuries per millimole but may be as much as a factor of 10 higher because of self absorption in benzaldehyde.

In conjunction with the isotope dilution method for detecting benzyl benzoate as a possible intermediate in the Cannizzaro reaction, it is necessary to effect complete separation of this ester from possible contaminants such as benzaldehyde, benzyl alcohol and benzoic acid. Accordingly, an investigation was conducted to see if fractional crystallization of benzyl benzoate from a suitable solvent mixture could be employed for this purpose. It was found that benzyl benzoate could be crystallized from 75% ethanol - 25% water solutions at 0° with stirring and addition of a seed crystal. The crystals (m.p. = 18 - 18.5°) were filtered by suction on an ice-cooled fritted glass funnel. It was found that benzyl benzoate could be maintained in the solid state on the funnel as long as the ice packing was present. After two crystallizations followed in each case by drying of the liquid over Drierite, the melting point and the index of refraction of the benzyl benzoate were constant.

Synthesis of an additional supply of benzaldehyde-p-t is under way. A constant-temperature oil bath regulated to control at 100° is being set up for kinetic measurements. It is anticipated that rate determinations will be in progress during the next period. (A. L. Powell)

IV. Oxidation of Alcohols by Bromine

Two alcohols, 2-propanol and 1-fluoro-2-propanol have been oxidized in 0.01 - 0.10 M concentration with 0.0018 - 0.009 M bromine in water solution at 50° to establish the mechanism of oxidation. The stoichiometry is

\[ \text{CH}_3\text{-CHOH-CH}_2\text{-R + Br}_2 \rightarrow \text{CH}_3\text{-CO-CH}_2\text{R + 2HBr}. \]

\( R = \text{H or F} \)

The reactions were followed by measuring the rate of disappearance of bromine. The kinetics indicates that the oxidation of the two alcohols are similar in every respect except that 2-propanol is oxidized 1000 times faster than 1-fluoro-2-propanol. The reactions are first order in alcohol and first order in bromine. The pH was varied by from pH 3 to 1 by adding perchloric acid.
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with no change in rate except with 1-fluoro-2-propanol at pH 1. The ionic strength was varied with sodium perchlorate from 0.01 - 1 M with only small changes in rate, although the rate of oxidation of 2-propanol did increase 4-7% between 0.1 and 1 M. Addition of sodium bromide decreased the rate only to the extent to which tribromide ion was formed.

\[ \text{Br}_2 + \text{Br}^- \rightleftharpoons \text{Br}_3^- \quad K = \sim 10 \text{ at } 50^\circ \]

Tribromide ion was unreactive.

In separate experiments the rates of bromination of acetone and fluoracetone were measured to determine if the products of the oxidation could have an effect on the rate of bromine consumption.

\[ \text{CH}_3\text{-CO-CH}_2\text{-R} + \text{Br}_2 \rightarrow \text{CH}_3\text{-CO-CHBrR} + \text{HBr} \]

The total rate of bromine consumption for both the oxidation of the alcohol and the bromination of the ketone may be expressed by the rate equation

\[ \frac{d[\text{Br}_2]}{dt} = k_1 [\text{CH}_3\text{-CHOH-CH}_2\text{-R}][\text{Br}_2] + k_2 [\text{CH}_3\text{-CO-CH}_2\text{R}][\text{H}^+] \]

To suppress the bromine consumption by the ketone the alcohols were oxidized at pH 3 and at low bromine concentration, 0.004 M, to limit the amount of ketone formed and the amount of acid produced. This was successful in all the runs with 2-propanol and with the runs with 1-fluoro-2-propanol above 0.04 M in alcohol. The runs made below this concentration showed an increase in rate with time.

To learn more about the mechanism, the ratios of the rates of oxidation of the protium compound to the deuterium compound were measured.

\[ \frac{k_{\text{CH}_3\text{-CHOHCH}_3}}{k_{\text{CH}_3\text{CHODCH}_3}} = 1.49 \quad \frac{k_{\text{CH}_3\text{-CHOHCH}_3}}{k_{\text{CH}_3\text{-CDCH}_3}} = 2.96 \]

\[ \frac{k_{\text{FCH}_2\text{-CHOHCH}_3}}{k_{\text{FCH}_2\text{CHODCH}_3}} = 2.10 \quad \frac{k_{\text{FCH}_2\text{-CHOHCH}_3}}{k_{\text{FCH}_2\text{CHODCH}_3}} = 2.83 \]

This clearly indicates that both hydrogen atoms are coming off in the rate-determining step. Only two mechanisms are consistent with this conclusion and the data mentioned earlier. They are

\[ \text{RCH}_2\text{C} = \text{O} \quad + \quad \text{H}_3\text{O}^+ \quad + \quad \text{HBr}_2^- \quad \quad \quad \text{(1)} \]

\[ \text{RCH}_2\text{C} = \text{O} \quad + \quad \text{H}_3\text{O}^+ \quad + \quad \text{HBr}_2^- \quad \quad \quad \text{(2)} \]
The isotope effects may also be used to distinguish between these two mechanisms. In mechanism 1 the O-H bond is being broken with the H being removed as a proton. Because of the inductive effect of the fluorine the effect should be to increase the OH bond breaking in the transition state as compared to 2-propanol and thus to increase the magnitude of the isotope effect.

\[
\frac{k_{\text{FCH}_2\text{CHOHCH}_3}}{k_{\text{FCH}_2\text{CHODCH}_3}} > \frac{k_{\text{CH}_3\text{-CHOHCH}_3}}{k_{\text{CH}_3\text{-CHODCH}_3}}
\]

In mechanism 2 the fluorine should inhibit this bond breaking and reduce this isotope effect.

On the other hand the C-H bond is also being broken in the transition state. Fluorine again because of its inductive effect would be expected to inhibit the removal of H from the carbon via mechanism 1 and thus the isotope effects expected would be,

\[
\frac{k_{\text{FCH}_2\text{CHOHCH}_3}}{k_{\text{FCH}_2\text{CDOHCH}_3}} < \frac{k_{\text{CH}_3\text{CHOHCH}_3}}{k_{\text{CH}_3\text{CDOHCH}_3}}
\]

while mechanism 2 would predict the reverse effect.

The results indicate that mechanism 1 is correct. (R. A. Wiles)

V. Origin of Isotope Effects in Light and Heavy Water

The previous communication reported the development of a theory capable of accounting quantitatively (a) for the thermodynamic differences between light and heavy water, and (b) for the isotope effects observed in the hydration of monatomic ions.

The theory has since been successfully extended to include the solvation of polyatomic ions in light and heavy water. It has been possible to show that the "solvent isotope effect" in such cases has two distinct causes, (a) the effect that the ion has on the librational frequencies of the water molecules (this phenomenon provides the total effect for monatomic ions) and (b) the fact that the ion itself is not capable of free rotation in solution but rather undergoes a librational motion as result of the electrostatic interaction of the ionic field and that of the surrounding water molecules. Two ionic species in particular have been fully discussed, the hydronium and hydroxide ions. The constant governing the more important of the exchange equilibria involving the hydronium and hydroxide ions have been calculated and their significance discussed.

The theory has also been extended to include the isotope effects observed in the use of mixtures of light and heavy water. Agreement between experimental and calculated results has been obtained for both equilibrium and kinetic studies conducted in solutions of the two waters. For example, the calculated values of the dissociation constant for acetic acid as a function of the mole fraction of deuterium present in the solvent show a maximum deviation from the experimentally
determined values of only two per cent. The average deviation is much less.

At the present time the theory is being extended to cover the kinetic isotope effects observed in the use of light and heavy water. (R. F. W. Bader)

VI. Hydrolysis of Alkyl Halides in Light and Heavy Water

Table 3.1 reports first-order rate constants for hydrolysis of methyl halides in light and heavy water.

<table>
<thead>
<tr>
<th>Compound</th>
<th>Temp., °C</th>
<th>$k_{H_2O} \times 10^5$, sec.$^{-1}$</th>
<th>$k_{D_2O} \times 10^5$, sec.$^{-1}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Methyl chloride</td>
<td>79.9</td>
<td>1.790 ± 0.003</td>
<td>1.366 ± 0.008</td>
</tr>
<tr>
<td>Methyl chloride</td>
<td>70.0</td>
<td>0.686 ± .004</td>
<td>0.519 ± .002</td>
</tr>
<tr>
<td>Methyl iodide</td>
<td>79.9</td>
<td>6.061 ± .008</td>
<td>5.047 ± .008</td>
</tr>
<tr>
<td>Methyl iodide</td>
<td>70.0</td>
<td>2.68</td>
<td>2.08</td>
</tr>
</tbody>
</table>

It seems likely that at least one of the four rate constants obtained for the hydrolysis of methyl iodide is in error. The two isotope effects ($k_{H_2O}/k_{D_2O}$) concerned are 1.20 at 79.9° and 1.29 at 70.0°. It would be surprising if there were this much change in isotope effect at two temperatures ten degrees apart and in this region. These reactions are therefore being repeated to determine whether the same results are obtained. (R. N. Griffin)

VII. Decarboxylation of $\beta$-Keto Acids

A sample of the p-toluylacetic acid-d used in the study reported previously was analyzed by the falling drop method and found to contain 1.055 deuterium atoms per molecule.

Enolization of the salts of $\beta$-keto acids in aqueous base was studied by consumption of bromine. Since the results of this work indicated that at least some of the benzoylacetic acids must have contained high percentages of deuterium in the methylene group, benzoylacetic acid-d and benzoylacetic-d$_2$ acid-d, containing 1.05 and 2.77 deuterium atoms per molecule respectively were synthesized and decarboxylated. The rates of the two decarboxylations were the same within experimental error.

It was also shown by bromination studies that no enolization takes place in benzene solution at 50° except that incidental to the decarboxylation. (R. N. Griffin)
VIII. The Solvolysis of t-Butyldimethylsulfonium Chloride and Perchlorate in 90% Acetone and Acetic Acid

Previous experiments by Kaiser\(^1\) and Knee\(^2\) with t-butyldimethylsulfonium chloride and perchlorate can be interpreted by assuming that the t-butyldimethylsulfonium cation decomposes without assistance from chloride ion (equation 1). Subsequent reaction, (equation 2 or 3), produces the solvolysis product. If this is true, the rates of the two salts in the same solvent should be the same. Kaiser found that there was a difference in rate of 20% between t-butyldimethylsulfonium chloride and perchlorate solvolysis in 90% acetone. The object of this investigation was to remeasure the rates in 90% acetone to see if they were the same and to measure the rates of both salts in 100% acetic acid.

Remeasurement of the rates of solvolysis of t-butyldimethylsulfonium chloride and perchlorate in 90% acetone continue to show that the chloride reacts faster (here 18%) than the perchlorate. Rate constants could be duplicated within 7% with the chloride and within 1% with the perchlorate. Realizing that the chloride ion may be reacting with the sulfonium ion introducing a second-order term, a rate of solvolysis of t-butyldimethylsulfonium chloride was measured with a ten-fold excess of chloride ion present. This was compared with the rate of a similar run using the same concentration of sodium perchlorate. The rate was faster with added sodium chloride. However, in 90% acetone, a 0.1 M solution of sodium chloride is almost saturated. If the water was tied up solvating the sodium and chloride ions, it would make the remaining solution less aqueous and less polar. Since there is probably spreading of the charge in the transition state, the lowered polarity may increase the rate. Consequently, two comparative runs are now being made with approximately 10\(^{-2}\) M t-butyldimethylsulfonium perchlorate plus equivalent sodium chloride and a ten-fold larger quantity of sodium perchlorate added in the first and no sodium chloride but added sodium perchlorate to keep the total salt concentration the same in the second. (D. A. Kuhn)

IX. Decomposition of Sulfonium Salts in Light and Heavy Water

Until recently, there has been much confusion in the literature concerning the decomposition of sulfonium salts. Gleave, Hughes, and Ingold\(^3\) proposed that the mechanism for the decomposition of trimethylsulfonium compounds is

\[
R_3S^+ X^- \xrightarrow{\text{slow}} R^+ + R_3S + X^- \quad \xrightarrow{\text{fast}} R_3S^+ X^- \quad (1)
\]
The first, slow, rate-controlling step according to this mechanism would be the fission of the sulfonium cation to form a carbonium ion which then reacts with the anion to form a halide. Kinetically this process should be first-order and the rate should be largely independent of the nature of the anion. Kaiser\(^1\) has shown this mechanism to be incorrect for trimethylsulfonium salts and tribenzylsulfonium salts. She found the decomposition of these salts to be a second-order reaction if the ionic strength of the reaction medium was kept high enough, by means of an inert salt, to eliminate the large salt effects which the earlier investigators had failed to note. The mechanism of the decomposition of trimethylsulfonium salts and tribenzylsulfonium salts in 90\% acetone - 10\% water is then a bimolecular attack of the anion on the sulfonium cation.

\[
R^+ + X^- \rightarrow RX
\]

The fact that trimethylsulfonium perchlorate and tribenzylsulfonium perchlorate failed to react is further proof that the rate is dependent upon the nature of the anion, since the perchlorate anion is much less nucleophilic than any of the other anions studied.

Dimethyl-t-butylsulfonium salts, however, do decompose by mechanism 1,1,4 Swain, Cardinaud and Ketley\(^5\) have studied the hydrolysis of these salts in light and heavy water to determine whether there was an "isotope effect" and, thereby, learn more of the role of the medium in such reactions. They reported no difference in rates and explained this lack of an isotope effect on the basis that the hydrolysis involves splitting off of an uncharged molecule, dimethyl sulfide, which does not need electrophilic solvation.

The present investigation is also designed to study the role of the medium in the decomposition of sulfonium salts. The decomposition of tribenzylsulfonium salts will be studied in light and heavy water to determine whether there is an "isotope effect" caused by the difference of light water - anion bonds of solvation and heavy water - anion bonds of solvation. The size and nucleophilicity of the anion will be changed (azide, halides, phenolate, etc.) to determine kinetic dependence on the nature of the anion and also to study the "isotope effects" caused by the medium due to the changes in nature of the anion.

Methods are being investigated for studying the kinetics of these reactions. The halides can be conveniently studied by use of the Volhard method, but anions such as azide and acetate present a problem. Since the common product is dibenzylsulfide in all of these reactions, a study is being made of the feasibility of using a tracer technique involving isotope dilution.

Tribenzylsulfonium perchlorate has been prepared in good yield by the method of dePascual.\(^6\) An attempt to prepare tribenzylsulfonium chloride from this compound by ion exchange was unsuccessful. Other attempts have been made to prepare tribenzylsulfonium chloride with special emphasis on the method of Lee and Dougherty.\(^7\) The most recent reaction yielded a solid which had a melting point of 76\(^\circ\). They report a m.p. of 82-84\(^\circ\) for tribenzylsulfonium chloride. Upon recrystallizing from methanol, the compound decomposed to dibenzylsulfide. (C. R. Morgan)
X. Proton Exchange Between Ammonium Ions and Methanol

In further work on the mechanism of exchange between tritium-labelled ammonium bromide and methanol in dimethylformamide solution at 25°, it was found that the reaction is first order in ammonium bromide below 0.10 M ammonium bromide. The ionic strength was kept constant at 0.20 M with sodium bromide. It was also found that exchange between ammonium ion and acid (conjugate acid of dimethylformamide) is extremely slow. No significant proton exchange occurred in 96 hours at 25°. These results are in accord with the data and mechanism previously published.8 (J. W. Ladbury)

XI. Investigation of the Nature of the Stability of Phthaloyl Peroxide

The marked stability of phthaloyl peroxide in carbon tetrachloride solution (50% decomposition of a solution, 0.05 M, requires 15 days at 80°) may be attributed to the stability of this diacyl peroxide ring system per se or to facile ring closure following thermal fission of the oxygen-oxygen bond. Experiments in which carbonyl oxygen could be distinguished from oxygen of the oxygen-oxygen bridge might provide evidence on these possibilities. The use of oxygen-18 provides a physical distinction between these positions and reaction of phthaloyl peroxide with olefins provides a chemical distinction between the two types of oxygen.

\[
\begin{align*}
\text{O}^{18} & \quad \text{O}^{18} \\
\text{O} & \quad \text{O} \\
\text{C}_6\text{H}_5\text{C}-\text{C}_6\text{H}_5 & \text{CCl}_4,80^\circ \\
\text{NaOH, EtOH} & \\
\text{H} \quad \text{OH} & \\
\text{C}_6\text{H}_5 - \text{C} - \text{C}_6\text{H}_5 & \text{OH} \quad \text{H}
\end{align*}
\]
Phthaloyl chloride was treated with water containing 1.4 atom % excess oxygen-18. The phthalic acid -018 was recovered and converted to phthaloyl chloride-carbonyl 018 by use of phosphorus pentachloride. The phthaloyl chloride was converted to phthaloyl peroxide-carbonyl 018.

The isotopically-labeled phthaloyl peroxide was treated in two ways. One portion was added directly to trans-stilbene in carbon tetrachloride. After 16 hours the cyclic phthalate I was isolated and purified. A second sample of phthaloyl peroxide was heated in carbon tetrachloride for 94 hours at 80°. One equivalent of trans-stilbene was added and after 16 hours the cyclic phthalate I was isolated. Both samples of cyclic phthalate were hydrolyzed to dl-stilbenediol. The two cyclic phthalate samples and the derived diol samples have been submitted to Professor Bender for oxygen-18 analysis. Further discussion awaits the results of these analyses. (F. D. Greene)

XII. Stereospecificity of Free Radical Reactions

Previous work in these laboratories has shown a qualitatively high degree of stereospecificity in the peroxide-catalyzed reaction of bibenzyl with N-bromosuccinimide. A quantitative analysis of this problem is under investigation by means of isotopic dilution techniques using C14.

Benzoic acid-carboxyl-C14 has been prepared from bromobenzene and reduced to the correspondingly labeled benzyl alcohol by lithium aluminum hydride. Conversion of the alcohol to benzyl bromide by hydrogen bromide and coupling of the bromide by magnesium afforded bibenzyl-benzyl-C14, of specific activity 0.5 µc/mmole. Reaction of the bibenzyl with N-bromosuccinimide afforded the meso-dibromide in 92% yield. To the residual material was added a fixed amount of unlabeled dl-dibromide. By the re-isolation of dl-stilbene dibromide and analysis of its specific activity, it was established that this isomer is formed in the NBS reaction of bibenzyl in 2% yield. The stereospecificity in meso- vs. dl-product formation thus appears to be 45 to 1. The more rigorous experimental test, involving addition of all dl-dibromide prior to the start of the reaction, is in progress.

The possibility of decomposition of the chain-carrying carbon radical, C6H5CHBrCHC6H5, to a bromine atom and stilbene followed by re-addition has been rendered unlikely by the inertness of added stilbene under the reaction conditions. (G. Maciel)

Reaction of p, p'-dinitrobibenzyl with N-bromosuccinimide in carbon tetrachloride under peroxide catalysis affords a high-melting dibromide in 85% yield. The reported formation of a dibromide of the same m.p. from the photochemical bromination of trans-p, p'-dinitrostilbene makes it very probable that the material is the meso-isomer. N-bromosuccinimide bromination of the dinitrobibenzyl on a small scale has afforded 85% yield of the high-melting dibromide, isolation of a small amount (~ 1%) of material containing the succinimide residue but no isomeric dibromide fraction. Large scale experiments in progress may reveal the presence of such an isomer. Proof of structure of the high-melting dibromide is also being sought by degradation experiments and by independent synthesis of the unknown dl-dibromide. (P. Zanet)
REFERENCES


THESES

Elizabeth E. Pegues, "Mechanism of Methanolysis of Triphenylmethyl Chloride in Benzene Solution", Ph.D. in the Department of Chemistry, August 1957.

A. MacLachlan, "Effect of Solvent on the Rate of Solvolysis of Alkyl Halides", Ph.D. in the Department of Chemistry, August 1957.

C. G. Swain
F. D. Greene II
I. Multiplate Cloud Chamber Analysis

The rescanning and analysis of the pictures taken with the large multiplate cloud chamber at the Brookhaven National Laboratory are being continued. The scanning and measuring projectors and the allied analysis equipment have been moved to the Compton Laboratory.

One portion of the program is concerned with determining the cross sections for strange particle production by 1.8-Bev $\pi^-$ mesons and 2.6-Bev protons on iron. The proton work is essentially completed, except for some systematic corrections which will be made on the basis of the $\pi^-$ work, for which many more events are available. Preliminary results show that the $\pi^-$ mesons are considerably more effective in producing strange particles than are the protons. The hyperon production cross sections in iron are $28 \pm 11$ mb (of which $22$ mb is $\Lambda^0$ or $\Sigma^0$) for pions and $2.0^{+1.3}_{-0.6}$ mb for protons. Despite this large difference, the cross section for producing $K^+$'s seems to be about the same for pions and protons, since the $\pi^-$'s produce predominantly $\Lambda^0$'s, $\Sigma^0$'s, and $\theta^0$'s. Additional evidence for the pion-proton total cross section difference is provided by the observation that secondaries (mainly pions) from interactions of the proton beam tracks in about one mean free path of iron in the chamber plates produce about one-half as many strange particles as are "directly" produced by the protons. This gives a measure of how much of this "direct" production is actually due to intermediary pions within a given iron nucleus.

Our data indicates also that the K-pair production process may be of more importance relative to hyperon-K production for protons than for pions. First, both associated events we have observed with protons are K pairs, and secondly, the total K cross section (greater than $2.0 \pm 1.0$ mb for $K^+$ plus $1.4^{+1.6}_{-0.5}$ mb for $K^0$) for protons exceeds the total hyperon cross section.

Current results on the program to determine the branching ratio $R = \theta_1^0 - 2\pi^0/\theta_0^0$ indicate that this ratio is considerably smaller than that predicted by the $\Delta T = 1/2$ selection rule ($R = 1/3$), and is even too small to be compatible with an admixture of $\Delta T = 3/2$ ($R > 1/4$), and hence the experimental result presents an interesting theoretical problem. If all of our events showing $\gamma$-ray showers near identified $\Lambda^0$'s are assumed to be due to the decay $\theta_0^0 \rightarrow 2\pi^0 \rightarrow 4\gamma$, then $R = 0.13$. However, if one makes an appropriate background subtraction, $R \approx 0.03$. The latter value is also consistent with our having observed just one case of a multiple shower. This one multiple-shower case provides, however, excellent evidence that the $\theta_1^0 \rightarrow 2\pi^0 \rightarrow 4\gamma$ decay does indeed exist, although it is compatible with some other neutral decays, such as $\theta_2^0 \rightarrow 2\gamma$. Fortunately, the two $\gamma$ rays which made the showers apparently carried nearly all the energy of the $\pi^0$'s, since the two showers are within $5^0$ of being coplanar with the origin, and since their energies, $300 \pm 100$ and $490 \pm 110$ Mev, are almost those which the $\pi^0$'s would have to have had
(295 ± 10 and 600 ± 35 Mev, respectively), if they came from a 90° decaying at the observed angle. (E. Boldt, H. Bradt, H. S. Bridge, D. O. Caldwell, Y. Pal, and B. Rossi)

II. The Velocity Spectrum of Cosmic Ray μ-Mesons at Sea Level

The absolute differential range spectrum in air of vertical cosmic-ray mu mesons was determined from the direct measurement of the time-of-flight spectrum of particles between two scintillation counters separated by about 9.5 m of air. The statistical dispersion in the time-of-flight measurements is 1.7 millimicroseconds, and a statistical analysis was done to determine the effect of this dispersion on the data. The analysis procedure involved the selection of events according to a criterion on each measured variable. For μ mesons the variables were the time of flight, the energy of the stopped μ meson, the energy loss of its decay electron, and the time separation of the μ meson and its decay electron. We evaluated the effects of the selection criteria and of errors which were inherent in the experimental method, and we applied appropriate corrections to the data. The errors inherent in the experimental method were caused by multiple coulomb scattering in all parts of the apparatus, the radioactive decay of μ mesons during their flight through the apparatus. The final results for the average intensities of vertical cosmic-ray μ mesons at sea level and 50° north geomagnetic latitude in the range intervals 5.3 to 30.8 g cm^{-2} and 56.2 to 82.2 g cm^{-2} of air are $4.61 ± 0.54 \times 10^{-6}$ sec^{-1} g^{-1} sterad^{-1} and $4.92 ± 0.77 \times 10^{-6}$ sec^{-1} g^{-1} sterad^{-1} respectively. In addition, we have evaluated the intensity of vertical cosmic-ray protons. An estimate of the average intensities of vertical cosmic-ray protons at sea level and 50° north geomagnetic latitude in the range intervals 6.1 to 24.0 g cm^{-2} and 57.2 to 78.2 g cm^{-2} in air is $1.27 ± 0.32 \times 10^{-6}$ sec^{-1} g^{-1} sterad^{-1} and $1.85 ± 0.41 \times 10^{-6}$ sec^{-1} g^{-1} sterad^{-1} respectively. (G. W. Clark and J. S. Strickland)

III. Air Shower Monitors

Air shower experiments being conducted at Chacaltaya, Bolivia, are being continued. Experiments conducted with the monitor in Colorado have been discontinued. (H. Bradt, J. Hersil, R. W. Williams, in collaboration with I. Escobar, Laboratorio de Fisica Cosmica, La Paz, Bolivia)

IV. Large Meson Telescope

The testing of the second pair of scintillators was satisfactorily completed, and the output of this telescope section was added to the output of the first section by means of a discriminator-mixer designed and constructed for this purpose. The counting rate integrator was then connected
Cosmic Ray Group

to this combined output, and the integration period was reduced to 30 seconds, so that the same number of total counts would be recorded in half the time. This results in similar calibration and statistical fluctuations for each point, but in finer time resolution and better statistics for any given period than before.

Local barometric pressure and upper atmospheric data were collected and evaluated for 10 a.m. and 10 p.m. (the standard times for radiosonde measurements) daily during April and May, 1957. The best method of calculating the effects of the atmosphere on our counting rates was worked out. Some periods were discarded because of experimentally doubtful upper atmosphere or counting rate data. A double regression calculation was made of the correlation between local barometric pressure and the height of the 100 millibar pressure level at Boston with our cosmic-ray meson counting rate, for April and May separately. A further breakdown of the period of calculation, comparison of the measured counting rate with the calculated counting rate and with magnetic disturbances, and a more careful correction for biasing effects is now being undertaken to resolve some discrepancies with previous results.

A detailed check was made of the counting rates recorded during several of the larger flares occurring since April first. No significant fluctuations were noticed. No further magnetic storm decreases have been definitely established, although several short decreases are being compared with other cosmic-ray data.

With a 0.7% statistical accuracy on a 30-second time base, no significant short-time fluctuations in cosmic ray intensity have been observed. (R. D'Arcy, R. Palmeira, R. W. Williams)

V. Large Air Shower Experiment

Results obtained through May 1957, from the large air shower experiment were reported at the Varenna Conference on Cosmic Rays sponsored by the International Union of Pure and Applied Physics and will appear with the conference proceedings in a Supplement to the Nuovo Cimento.

During the summer a thorough analysis of the accuracy of our determinations of shower size and core location was carried out with the aid of the IBM 704 computer in the M.I.T. Computation center. In addition, work proceeded on the preparation of a new data reduction program for use on this computer. (B. Rossi, G. W. Clark, J. Earl, W. L. Kraushaar, J. Linsley, F. Scherb, and T. Cline)

VI. Mu-Meson Component of Large Air Showers

The experiment on the \( \mu \)-meson component has been completed and has been written up as a thesis by James Earl. (J. A. Earl and W. L. Kraushaar)
VII. Arrival Directions of Air Showers

Results on the arrival directions of 7000 showers recorded at Kodaikanal, India, were reported at the Varenna Conference on Cosmic Rays. No indication of anisotropy in the distribution of arrival directions of the primaries of these showers was found.

A total of about 42,000 events has been recorded and the data are being processed.

(E. Chitnis and G. W. Clark)

VIII. The Polarization of Cosmic-Ray $\mu$ Mesons

The experiment previously described has been completed with positive results. We find a difference of $(5.2 \pm 1.6)\%$ between the downward fluxes of decay electrons from positive $\mu$ mesons stopped in brass with and without a depolarizing magnetic field. The minimum polarization of low-energy $\mu$ mesons at sea level which this result indicates is $0.19 \pm 0.06$, and this quantity is consistent with the value expected according to the theory of S. Hayakawa.

The experimental work we have carried out and the earlier theoretical work of S. Hayakawa have been submitted as two articles for publication in the Physical Review.

(G. W. Clark and J. Hersil)

IX. Search for Long-Lived Particles Using the Cosmotron

Because of the prolonged shut-down of the Brookhaven cosmotron, no new experimental information has been obtained on the energetic long-lived decay process described in the last Laboratory for Nuclear Science Annual Progress Report (May 1957). However, in anticipation of a forthcoming run using the external beam of the cosmotron, new instrumentation has been developed. Additional counters and their associated coincidence-anticoincidence circuitry have been built and tested, so as to increase both the number of types of measurements which can be made, and also the number which can be made simultaneously.

(E. Boldt, H. S. Bridge, B. Rossi, and D. O. Caldwell)

X. Energy Flow in Air Showers

An experiment to measure the energy in the electronic component of extensive air showers is being undertaken at the Agassiz site; this will provide information correlating the energy of the particles in the shower front with the known particle density as a function of the distance from the core of each shower of known size. The latter data are provided by the air shower experiment proper.
The experimental arrangement is as follows: four scintillators, each a square meter in area, are stacked such that the pulses from the advancing shower particles can be recorded with the other air shower data by photographic oscilloscopes. In this way, by placing an appropriate amount of material sandwiched between the scintillators, the energy loss of the advancing shower sample can be determined.

The individual scintillators and circuits will be periodically calibrated by photographing pulses from single vertical \( \mu \) mesons, triggered by a Geiger tray telescope, and by photographing artificial pulses the sizes of which can be compared with the continuously recorded cosmic ray level. The displayed pulse heights triggered by the air showers, will be assumed to be roughly linear with respect to the original energy losses.

The device has been placed immediately beside the center detector in the air shower array, so that an independent measurement of the local particle density will be known. Continuous operation is expected for the next eight to ten weeks. (T. Cline)

XI. Giant Air-Shower Experiment

Through the kind assistance of Professor Victor Regener of the University of New Mexico, we have located four square miles of suitable land on Volcano Ranch, 15 miles northwest of Albuquerque. The elevation is 6000 feet. Preliminary negotiations with the owner indicate that use of the land can be arranged on acceptable terms.

The facility for manufacture of scintillating plastic has been overhauled with the object of reducing the manpower requirement. Production of plastics will begin within two or three weeks.

A prototype 3.5 m\(^2\) quatrefoil scintillation counter has been operating satisfactorily for two months. About one fourth of the electronic equipment has been built and tested. (J. Linsley)

ADDRESSES


Cosmic Ray Group

B. Rossi, "Raggi Cosmici e Brillamenti Solari", Bologna, Italy, June 1, 1957.


 I. B. Rossi, "General Description and Preliminary Results Concerning the Size Spectrum and the Zenith Angle Distribution".

 II. W. L. Kraushaar, "The Lateral Distribution of Electrons and $\mu$ Mesons".

 III. G. W. Clark, "The Arrival Directions of Air Showers".

 IV. J. Linsley, "Instrumentation of M.I.T. Air Shower Experiment".

App. I. G. W. Clark, "Methods of Analysis".

App. II. W. L. Kraushaar, "Fluctuations in Shower Development".


PUBLICATIONS


THESES


B. Rossi
A. SYNCHROTRON OPERATION AND MAINTENANCE

In the past quarter, 75% of the time has been spent making various changes to increase the reliability and intensity of the synchrotron. The rest of the time has been spent on experimental work.

I. Magnetic Field Tracking

While operating with a 15 millimicrosecond injector pulse it was found that the acceptance time for this pulse was about .1 μsec. or less. If the timing is mis-set the beam will reappear should the Van de Graaff voltage be changed without resetting the contractor pulse timing. The voltage change is 20 kv per microsecond. For this reason we equipped the Van de Graaff with an aluminum shell surrounding the head such that if pulsed, it would capacitively change the Van de Graaff voltage at the desired rate. The liner was tested as an energy analyzer and it operated as desired. However, when the liner was used there was no appreciable increase in beam intensity. We have some evidence that the expected increase does occur for very small beam currents. The reason for this non-linearity is not known at present; calculated space charge effects do not appear large enough to account for the trouble.

II. Vacuum System

The rubber boot system for vacuum sealing sections of the doughnut was not reliable. In order to get a more positive sealing a mechanical clamp was used. The clamps were strapped to the sections with stainless steel bands; flat gaskets were placed at the edges of the sections. This system is more reliable. (P. D. Luckey, W. Lobar, L. S. Osborne, C. J. Strumski, and A. Wattenberg)

B. SYNCHROTRON RESEARCH

I. Scattering of γ Rays by Protons, Deuterons, and Other Nuclei

The revised detecting equipment described previously has been calibrated, and now works well except that the pulse stretcher did not seem to remove the sensitivity of the pulse height analyzer channel widths to bias setting, and was therefore removed. The liquid hydrogen target has been simplified also. This experiment is now set up in the synchrotron beam. (R. Ely, D. H. Frisch, L. G. Hyman, and M. A. Wahlig)
II. One-Quantum Annihilation of Positrons

Testing of the double focusing magnetic spectrometer has proceeded. We find a dispersion of about 30''/Mev and a resolution of about 3%. The collection efficiency at present is about 4% for a monochromatic source of electrons. The magnet has been slightly reshaped in an attempt to improve the focusing and collection efficiency, and focusing studies of the new magnet shape must be made. (R. M. Weinstein and F. Y. Yap)

C. NUCLEAR EMULSIONS RESEARCH

I. K⁺ Forward Scattering

Work has been completed on the elastic scattering of K⁺ mesons at forward angles to determine whether the scattering is constructive or destructive with Coulomb scattering. To avoid uncertainties inherent in previous work the following precautions were observed.

1. Low-energy K mesons (~ 50 Mev average energy) were observed. This was to insure first that a track, even if scattered, would end in the stack and be identified unequivocally by its decay. Secondly, that the scattering be predominantly S-wave and that the sign of the potential therefore be relatable to a definite scattering state.

2. Care was taken in scanning to extend the measurements down to all angles with projections greater than 50, thus insuring that the region of interference with pure Coulomb scattering be completely covered. This interference region should occur for collisive momenta transfers of \( \sim 2.5 \cdot 10^{12} \text{ cm}^{-1} \cdot \text{h} \). The measurements were extended to collisive momenta transfers down \( \sim 1.0 \cdot 10^{12} \text{ cm}^{-1} \cdot \text{h} \). This precaution was taken to insure that the interference region was not missed.

3. Considerable care was taken with the angular measurements. In order to measure dip angles reproducibly to \( \sim 10^\circ \) a "tilting microscope stage" was constructed and used.

Figure 5.1 shows the experimental results, and for comparison are shown the calculated results for pure Coulomb scattering only. The experimental points lie systematically over the whole range above the pure Coulomb curves indicating no destructive interference. The agreement with the data of other groups is good. (G. Costa, D. M. Ritson, and R. A. Schluter)

II. Hyperon Study Program

The experiment described in the annual report (L.N.S. Annual Progress Report, June 1, 1956 to May 31, 1957) was scheduled at Brookhaven sometime during the summer of 1957; however, the Brookhaven Cosmotron has not been operating. The experiment has therefore been delayed until the fall. In the meantime some improvements have been made. The most important of these is the use of ignitron switching, which has proved convenient and reliable even at peak currents of 80,000 amperes. (R. A. Schluter, G. G. Fazio, and B. Ursin)
III. Polarization of Recoil Protons from Pion Scattering

Very soon after the resumption of operation of the Brookhaven Cosmotron, a run was obtained in which the recoil protons from the scattering in liquid hydrogen of 950-Mev negative pions were recorded in nuclear emulsions. The plates from this run are now being processed and the scanning for proton scatterings will be started as soon as the plates become available.

As a check on the method of detecting proton polarization by scattering in nuclear emulsions, an exposure was made to a 70% polarized proton beam of 150 Mev from the Harvard Cyclotron. Approximately 500 scatterings have been observed and large right and left asymmetries of the appropriate magnitude and sign have been measured in the angular range 10° - 25°. Below 10° the polarization appears to vary rapidly with angle, even changing sign at small angles, and to depend strongly on the proton energy. Such effects are to be expected as a result of the interference between Coulomb scattering and the nuclear scattering responsible for the polarization. A crude calculation using the model of Fermi\(^1\) indicates that such effects should be expected in the angular and energy range in which they have been observed. A more accurate computation by the method of Sternheimer\(^2\) is now being programmed for the M.I.T. computing machine. (B. T. Feld and B. Maglich)
D. MAGNETS AND INSTRUMENTATION

Glass-Chip Counter

Cosmic rays traversing the glass-chip-in-alpha-chloro-methyl-naphthalene counter do not give the expected pulse height and uniformity. This is possibly because the phosphate glass does not appear to be "water white" as previous (smaller) samples would have indicated. At the moment we do not have enough people to continue this development. (A. J. Morency and M. A. Wahlig)

E. BROOKHAVEN COUNTER GROUP

K-Meson Scattering

The differential Cerenkov counter designed by L. S. Osborne and D. O. Caldwell has been given some tests with electrons from the M.I.T. Synchrotron and \( \pi \) mesons from the Brookhaven Cosmotron, with results which are promising but inconclusive as yet. Further tests are being made at Brookhaven, where Dr. D. Hill of our research staff is now in residence.

The working Cerenkov fluid, the fluorochemical FC-75, behaves well on isotherms of 250°C and above. One must be sufficiently above the critical temperature so that the sensitivity of index of refraction to temperature and pressure near the critical point is avoided. Also, this liquid appears possibly to be a mixture of several fractions, judging by the separation out of layers near the critical point.

Our major difficulties have been with the weakness of quartz windows and the carbonization of silicone rubber gaskets. We now use highly polished quartz and are testing metal gaskets. (C. Burrowes, D. H. Frisch, S. G. Hamilton, D. A. Hill, and D. M. Ritson)

REFERENCES

During this period, the group acquired a 40-cubic-feet-per-minute air compressor which greatly simplified the operation of our bubble chambers. The 9-inch aluminum chamber was taken to Cornell University and exposed there to the 1.05-Bev gamma-ray beam. A well-collimated beam (1/8" in diameter) was passed through the center of the chamber. Approximately 100,000 exposures were made. A pulse height analyzer was used so that pictures were taken only when the gamma-ray beam was of the right intensity. The purpose of the experiment is to study the angular dependence and energy dependence of π-pair production by gamma rays. It is hoped that some strange particle production events will also be found.

The 9-inch stainless steel chamber to be used with a methyl iodide propane mixture was completed and tested satisfactorily.

Plans were completed for a 15-inch diameter by 14-inch deep stainless steel chamber. The 14-inch diameter by 6-inch thick glass has arrived from Germany. (P. D. Luckey, A. E. Brenner, I. A. Pless, A. Tuchman, R. W. Williams, and R. Yamamoto)
Photoneutron Time-of-Flight Measurements at the Linear Accelerator

The time-of-flight measurements of photoneutron spectra from heavy elements have been continued in essentially the same form described previously. The only major improvement has been the use of the RCL 256 Channel Analyzer to record data.

Measurements of neutron spectra from Bi, Au, and Ta induced by 16 Mev Bremsstrahlung have been repeated and a first attempt has been made to infer the neutron energy distributions from monochromatic X-rays. Figure 7.1 represents the difference between the neutron yield induced by 16 plus Mev Bremsstrahlung and that induced by 14 plus Mev Bremsstrahlung as a function of neutron energy. The normalization is per atom and, in the photon difference, corresponds to equal numbers of photons greater than about 5 Mev; the insert shows the photon difference spectrum. The best guess is the solid curve, but the photon spectrum could be anywhere within the cross-hatched area. The difference is a crude approach to monoenergetic X-rays, but is much better than the raw Bremsstrahlung spectrum. The neutron statistics are good (the difference spectrum for each element represents about 10,000 counts), but to date the reproducibility is somewhat worse than the statistics. The limitation is in the magnetic analyzing system which defines the electron spectrum incident on the Bremsstrahlung target. This system must be improved before more refined difference experiments are attempted.

Figure 7.1
Photoneutron energy distributions.
The quantity plotted in Figure 7.1 is $1/E$ times the neutron energy spectrum, so that a Maxwellian distribution becomes a straight line. The most striking feature is the peak (removing the $1/E$ factor) in the gold spectrum at about 4.5 Mev. In the absence of any suspicion of anomalous energy dependence of level densities in Au$^{198}$. It is natural to consider this peak as evidence for a direct photoeffect as described by Wilkinson. Because of the single particle nature of the dipole operator, the excited states of the target nucleus immediately after the photon absorption are closely related to a group of states of the residual nucleus. They are these states with a single excited nucleon$^{2,3}$. If the nucleon escapes fast enough, the residual nucleus will be left in one of these states. How fast is fast enough depends on $W$, the cloudy part of the nuclear potential. In the present case, for $W = 2$ Mev, one would expect about a 4% "direct effect", and a 7% or 8% direct photoneutron emission for $W = 1$ Mev. With this interpretation, making a subtraction of the lower energy "statistical" neutrons, the direct neutrons from gold are 7-11% of the total yield. Although for Bi there is no high-energy peak in the neutron spectra, a reasonable subtraction of evaporation neutrons would yield such a peak for a direct process. For Ta, if a peak exists at all it is much flatter.

It is difficult to precisely answer the question "what is the temperature?" for the approximately Maxwellian low-energy part of the spectra. The primary uncertainty is whether or not the detection of second neutrons from $(\gamma, 2n)$ processes is contributing appreciably to the region between 1 and 2 Mev. $(\gamma, 2n)$ thresholds from mass formulae are about 14 Mev, but are uncertain. A threshold as low as 13 Mev might appreciably affect a spectrum shape at 1 Mev neutron energy. Uncertainties in the energy variation of counter efficiency are also important at low neutron energies. At about 5 Mev excitation in the residual nucleus, temperatures for Au and Ta are not more than about 0.5 Mev however. This is lower than one would expect if the energy dependence is $(E/a)^{1/2}$ as predicted by the Fermi gas model with $a = \text{about 10 Mev}^{-1}$. (W. Bertozzi, P. T. Demos, F. R. Paolini, and C. P. Sargent).

REFERENCES


P. T. Demos
The work of the Rockefeller Generator Group during the quarter from June through September 1957 was concerned with the following items:

A. Continuation of work on adaptation of the generator to pulsed operation.

B. Experiments on parity non-conservation using positron emitters generated by \((d, n)\) reactions.

A. Pulsed Generator

1. Terminal Head. The construction of the pulsed terminal head described in the Laboratory for Nuclear Science Annual Progress Report (May 1957) was completed. Bench testing of the head is now in progress.

2. Main Pumping System. The main pumping system for the present generator has been redesigned for faster speeds. The present diffusion pump capable of speeds of 100 litres per sec is being replaced by a 500 litre per sec pump. All the component parts for the system have now been bought or manufactured, and the changes on the system are scheduled for the near future. With this greater pumping speed it should be possible to operate the generator reliably at a Mev above the 4-Mev limit for which it was originally designed.

3. Pure Sodium Iodide Scintillation Counter. The detecting equipment designed for use with the pulsed generator has been described in the Annual Progress Report for 1957. The detectors employed were photomultipliers with plastic scintillators.

However, in connection with proposed experiments on low-lying isomeric states of nuclei, plastic scintillators would not be suitable. In these experiments, it is necessary to detect the de-excitation gamma rays (100 kv - 200 kv), and in such phosphors the resultant pulse heights would be comparable to background noise. For gamma rays, these energies NaI (Tl) would normally be suitable as a detecting phosphor; in this instance, however, the decay time \((250 \times 10^{-9} \text{ sec})\) is too long for the resolving times required (a few times \(10^{-9} \text{ sec}\)).

To meet this need, following the investigations of Van Sciver\(^1\), a scintillation detector has been built with a pure NaI crystal which can be cooled to liquid air temperatures. It has been found that an eight-fold increase in light output can be obtained by cooling the crystal from room temperature to that of liquid nitrogen. For radiation of 120 kev, this corresponds to an average number of electrons emitted from the photocathode of 40, for this prototype unit. Such a light output is less than would be expected from NaI (Tl) at room temperature, but only by a factor of two or three.

The decay time of the phosphor at the low temperature has been measured as approximately \(15 \times 10^{-9} \text{ sec}\). This unit was substituted for one of the plastic scintillator units in the
time-to-pulse-height conversion systems previously described (Annual Progress Report, May 1957).

As before, coincidences were debated between the two annihilation quanta for Na\(^{22}\) source. The resultant time spectrum corresponded to a distribution with a full width at half height of 2.7 \(\times 10^{-9}\) sec.

Further investigation on the suitability of this counter in relation to fast (10\(^{-9}\) sec) detection of low-energy gamma quanta is scheduled. (L. E. Beghian, G. Kegel, M. Salomaa, and R. P. Scharenberg)

B. Positron Longitudinal Polarization Experiments

In conjunction with the Radioactivity Group, an experiment is in progress to determine the degree of polarization of positrons emitted from Al\(^{26}\). The Al\(^{26}\) is generated from the reaction Mg\(^{25}\)(d, n) Al\(^{26}\) obtained by bombarding separated Mg\(^{25}\) isotopes with 3-Mev deuterons.

A fuller description of the experiment is to be found in the current report of the Radioactivity Group (August 31, 1957). (M. Deutsch, B. Gittleman, and R. P. Scharenberg)

REFERENCES


PUBLICATIONS

G. Goldring and R. P. Scharenberg, "Gyromagnetic Ratio of Rotational 2\(^+\) States", submitted for publication to The Physical Review.

W. W. Buechner
M. Deutsch
ONR GENERATOR GROUP

I. Introduction

A considerable portion of the period covered by this report was spent in making various repairs and alterations in the accelerator. The r.f. ion-source bottle, which has been used since May 1956, was replaced because of decreased ion output. During the replacement, signs of tracking were noticed on the glass insulators between the source focus electrode and the first tube electrode. A shield was installed to prevent further damage to the insulators. As a result of admitting air to the vacuum tubes, a troublesome leak developed, and this prompted us to carry out an experiment which had long been planned. The accelerating tube and the differential pumping tube were disconnected and evacuated separately. No difficulty was experienced in operating the accelerator without the differential pumping action of the auxiliary tube. Electron loading in this tube was still apparent, however, and as a subsequent experiment the differential pumping tube was filled with nitrogen to a pressure equal to that in the generator tank. This has resulted in improved generator operation, and it is apparent that, with the present ion-source outlet size, differential pumping is not required. When the generator tank is next opened, we plan to remove the auxiliary tube completely.

After the repairs to the generator were completed, a number of new investigations were undertaken and are still in progress. These include studies of a number of \((d, \alpha)\) reactions in the mass region between 40 and 65, the primary objective being to determine the ground-state Q-values for a number of important mass-linking reactions. The analysis of the data from various experiments carried out before the generator shutdown has been continued, and some of the results obtained thus far are reported in the following sections.

II. Inelastic Proton Scattering From Nickel

The study of the excited states of nickel has been continued by measuring the inelastically scattered protons from \(\text{Ni}^{62}\). A target enriched in \(\text{Ni}^{62}\) was bombarded with 7.0-Mev protons, and the inelastically scattered groups were observed at angles of 50, 90, and 130 degrees. Our preliminary results indicate levels in \(\text{Ni}^{62}\) at 1.172, 2.048, 2.301, 2.335, 2.887, 3.053, 3.155, 3.172, 3.254, 3.366, 3.470, and 3.515 Mev. A target enriched in \(\text{Ni}^{94}\) has also been bombarded, but the results have still to be analyzed. (C. H. Paris)
III. (d, p) Reactions from the Iron Isotopes

The analysis of the proton groups obtained from the deuteron bombardment of targets made of natural iron and of enriched isotopes of Fe$^{54}$, Fe$^{57}$, and Fe$^{58}$ is continuing. The energies and intensities of several hundred groups have been measured from the three bombardments made from each isotope with 6.54- and 7.01-Mev deuterons. Identification of the individual peaks is being made, chiefly on the basis of intensity considerations. Thus far, thirty-six groups have been assigned to Fe$^{55}$, twenty groups to Fe$^{57}$, forty-one groups to Fe$^{58}$, and ten groups to Fe$^{59}$. For each of these isotopes, the regions of excitation thus far analyzed are 4.273, 2.703, 4.998, and 1.648 Mev, respectively. It is expected that the analysis of the remaining groups will be completed shortly, and the final results for each isotope will appear in the next Progress Report.

(A. Sperduto)

IV. (p, α) Reactions from the Iron Isotopes

An examination of the nuclear-track plates exposed in the inelastic proton scattering studies of the four stable iron isotopes has revealed a number of alpha-particle groups that have been attributed to the (p, α) reactions from iron. A total of eight groups were observed at 130 degrees from a natural iron target and from targets enriched in Fe$^{57}$ and Fe$^{58}$. No groups were observed from the corresponding exposure with the enriched Fe$^{54}$ target. All groups were relatively weak, and only one measurement was made of each group. The results from these studies are summarized in Table 9.1.

<table>
<thead>
<tr>
<th>Reaction</th>
<th>Q-Value (Mev)</th>
<th>Ex (Mev)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fe$^{58}$(p, α)Mn$^{53}$</td>
<td>-0.220</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td>-0.720</td>
<td>0.500</td>
</tr>
<tr>
<td>Fe$^{57}$(p, α)Mn$^{54}$</td>
<td>0.240</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td>0.190</td>
<td>0.050</td>
</tr>
<tr>
<td></td>
<td>0.080</td>
<td>0.160</td>
</tr>
<tr>
<td></td>
<td>-0.170</td>
<td>0.410</td>
</tr>
<tr>
<td>Fe$^{58}$(p, α)Mn$^{55}$</td>
<td>0.400</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td>0.270</td>
<td>0.130</td>
</tr>
</tbody>
</table>
The ground-state Q-value obtained in each case is in agreement with the value calculated from the known masses involved. The group corresponding to a level in Mn$^{55}$ at 0.130 Mev is in agreement with the value of 0.127 Mev obtained from the Mn$^{55}$(p, p')Mn$^{55}$ reaction. None of the alpha groups has been observed previously, and no other data on the levels in Mn$^{53}$ and Mn$^{54}$ appear in the literature. (A. Sperduto)

V. Multi-gap Spectrograph

The machine drawings for the new type spectrograph have been essentially completed. As yet, no acceptable bid has been received on the ten-ton iron core, but as a result of some recent contacts, we hope that the order for the iron can be placed during the coming quarter. The design of the coils has been changed. We now plan to use 0.8-inch square copper bars with a 1/2-inch hole for water cooling. There will be a coil of ten turns between each air gap. All coils will be electrically series connected, but there will be four parallel water circuits. A high-speed mercury diffusion pump, previously used on the air-insulated accelerator, has been reconditioned for use with the spectrograph. An auxiliary vacuum-test chamber has been built for use with this pump, so that the vacuum properties of the various insulating materials which will be used within the spectrograph tank can be studied. (H. A. Enge, M. Mazari, and J. A. Taylor)

REFERENCES


Addresses

Invited Papers

H. A. Enge, "Results of (d, p) stripping measurements on some light-to-intermediate nuclei, Radiation Laboratory, University of California Colloquium, July 18, 1957.

H. A. Enge, "Results of (d, p) stripping measurements on some light-to-intermediate nuclei, Oak Ridge National Laboratory Colloquium, August 9, 1957.

Publications


ONR Generator Group


Mazari, Sperduto, and Buechner, "Excited States in Co$^{59}$ and Fe$^{56}$, Phys. Rev. 107, 365 (1957).

W. W. Buechner
RADIOACTIVITY GROUP

Polarization of Positrons

An attempt is being made to extend the measurements of the longitudinal polarization of high-energy positrons previously reported by us for Ga$^{66}$ and Cl$^{34}$ to the case of Al$^{26}$. This decay is presumably a pure Fermi Transition and could provide a valuable confirmation of our earlier results. The very short half-life of Al$^{26}$ makes it imperative that the measurements be carried out very close to the accelerator which produces the activity. Despite the larger activity which can presumably be obtained from the cyclotron, considerations of background and ease of access lead us to attempt production by bombarding separated Mg$^{25}$ with 3-Mev deuterons from the Rockefeller generator.

As in the earlier experiments the polarization will be detected by the absorption of the annihilation radiation in magnetized iron. Since Al$^{26}$ decays without emission of gamma rays, it may be possible to avoid the use of a magnetic spectrometer to separate the positrons from the other radiations but provision has been made for the use of a short-lens spectrometer if it should prove to be necessary. A compressed-air operated mechanism transports the target between bombarding and counting positions. This mechanism as well as the control circuits for the bombarding and counting cycle have been installed and tested. Preliminary measurements indicate that the available intensity will be marginal unless the stable operating energy of the accelerator can be raised somewhat.

The method used by us for the determination of the positron helicity, through the helicity of the annihilation radiation is well suited for high positron energies. It seemed of interest to investigate a method which might more easily yield results at lower energies. Such a method might be provided by the dependence of the annihilation cross-section for polarized positrons on the relative spin direction of the target electrons. Since the annihilation in flight competes with energy loss by ionization, we may expect the intensity of high-energy annihilation radiation produced by longitudinally polarized positrons in magnetized iron to depend on the direction - parallel or antiparallel to the positron momentum - of the magnetization. In particular, calculation by Page$^1$ shows that the emission of the highest-energy quanta in the forward direction is due exclusively to electrons and positrons with anti-parallel spin. The effect diminishes rapidly, however, for larger angles of emission, and correspondingly lower gamma-ray energies, especially with high positron momenta. For sufficiently large angles the effect changes sign, so that for positron energies above about 4 Mev the integral annihilation cross section is greater for parallel than for antiparallel spin. It is therefore important to maintain sufficient angular and energy resolution in this experiment.
Figure 10.1

Experimental arrangement for measuring longitudinal polarization of positrons by method of annihilation in flight.
We have carried out preliminary experiments with a source of Cu$^{62}$, in equilibrium with its Zn$^{62}$ parent. The source nuclide was chosen because of the convenient half-life and energy and because of the absence of nuclear gamma radiation which reduces background problems. Collimation and energy selection of the positrons was performed by means of the magnetic spectrometer lens much in the manner of our previous experiments. The annihilation-converter was a hollow cone of iron, illustrated in Figure 10.1, designed to minimize the possibility of annihilation in unmagnetized material. The wall thickness of the cone varied in such a manner that saturation of the iron was maintained over the whole surface.

Table 10.1 summarizes the results obtained with a source of about 6 mC strength at three positron energies, after correction for background counts. The last column shows the magnitude of the effect expected for positrons completely polarized with positive helicity, considering the angular and energy spreads involved. The agreement is seen to be good but the experimental errors of these measurements are still rather large.

**TABLE 10.1**

<table>
<thead>
<tr>
<th>$E_\gamma - E_{\beta}^+$ (Mev)</th>
<th>0.3</th>
<th>0.5</th>
<th>0.7</th>
<th>Theoretical 0.7</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.15</td>
<td>2.7 ± 1</td>
<td>2.1 ± 1</td>
<td>6.2 ± 1.7</td>
<td>10</td>
</tr>
<tr>
<td>1.5</td>
<td>1.7 ± .8</td>
<td>1.8 ± 1.1</td>
<td>6.8 ± 2.8</td>
<td>9.0</td>
</tr>
<tr>
<td>2.0</td>
<td>2.5 ± 0.7</td>
<td>4.2 ± 1.1</td>
<td>2.4 ± 2.2</td>
<td>7.5</td>
</tr>
</tbody>
</table>

(B. Gittelman, R. Scharenberg and M. Deutsch)

REFERENCES

During the interval covered by this report, the effort of the cyclotron group was directed towards the installation and alignment of the strong-focussing quadrupole-pair magnetic lens system.

When the old focussing system, consisting of some stacked iron plates in the cyclotron fringe field and a uniform field turning and focussing magnet*, was removed, it was found that the fringe field of the cyclotron caused the beam to turn to such a large extent (~200°) that an in-line system would have brought the beam to an undesirable part of the scattering chamber room. We therefore had to bring the beam out of the cyclotron in a magnetic channel consisting of two concentric iron cylinders about 15 inches long and each one about 3/4 inch wall thickness. This shield served to make the beam go in practically the same direction as the old focussing system.

Another source of trouble in alignment was the interaction of the quadrupole fields and the cyclotron. This was remedied by moving the magnets as far away from the cyclotron as possible. This distance turns out to be about five times the cyclotron pole piece gap.

After the installation of the magnets in this position, but before final alignment, the cyclotron field was re-shimmed, by trial and error, to compensate for all the changes made.

The performance of this focussing system is quite satisfactory in that we have had an effective increase in the target current of a factor of at least three, comparing peak beam currents and a factor of 5-10 comparing routinely available proton beams.

The peak beam current we have seen to date is 2μa of protons with an average beam current available from day to day of about 1μa. Previously our readily available proton beam current was about 0.2μa.

We furthermore now have a much better idea of our beam optics and believe it would be possible to improve our target beam even further by the addition of one additional magnet to more completely compensate for the horizontal defocussing in the cyclotron fringe field. We have no plans at present to attempt to obtain any greater beam current because of the limitations on our shielding and scattering room vault.

The magnets which have been described in earlier Laboratory for Nuclear Science Progress Reports derive their excitation from separate motor-generator sets which are controlled by a D. C. amplifier feedback arrangement which has a stability of better than 1 percent and is easily adjusted for the point of quiescent operation. The circuit for one of these regulators is given in Figure 11.1. (C. W. Darden, K. S. Lee, and N. S. Wall)


M. Deutsch
Figure 11.1
Magnet control circuit.
THEORETICAL GROUP

I. Nuclear Saturation

We solved the Bethe-Goldstone equation for s-waves, forgetting the coupling between s and d states due to the center-of-mass motion and assuming the hard core potential to be such that

\[ V_u = A \delta(x - c) \]

The energy shift was then calculated making use of the relation

\[ E = (u_o V_u) = A \frac{\sin K}{K} \]

where \( A \) was fixed by the asymptotic behavior of the solutions. The total energy of a hard core Fermi gas was then obtained as a function of density and applied to the saturation phenomena. The attractive energy was calculated neglecting the tensor force (using the singlet force only). We are now in the process of understanding the effects of the tensor force on nuclear matter. (L. C. Gomes, D. Walecka, and V. F. Weisskopf)

II. Optical Model of the Nucleus

A dispersion relation for the real and imaginary parts of the optical potential is determined for the case of an infinite nucleus. For finite nuclei the method used gives rise naturally to potentials which vary sharply with energy. These can be shown to give rise to fine structure resonances in the cross section and thus afford another insight into the theory of resonance reactions. The cloudy crystal ball model may now be obtained by finding the equation for a wave packet of suitable width in energy. (H. Feshbach)

III. Fluctuations in Nuclear Reactions

Using the Porter-Thomas distribution functions for nuclear reactions with either 1, 2, or a large number of exit channels has been determined. These distributions are rather wide so that the distribution of nuclear reactions in terms of averages and fluctuations is not sufficient. (H. Feshbach)

IV. High-Energy Nucleon-Nucleus Scattering

The high-energy approximation for nucleon-nucleus scattering has been put into a useful form for the analysis of experimental data. The potential well which consists of a flat portion plus
a tail is used. Spin-orbit interaction has been included. The analysis of the available experimental data is in progress. (J. Ricci)

V. Investigation of Bremsstrahlung

The problem of bremsstrahlung (continuous x-radiation) in collisions of elementary particles has been investigated, and the following theorem proved: If the differential bremsstrahlung cross section is expanded in powers of the energy loss, then the first two terms in the expansion may be expressed in terms of the charges, magnetic moments and elastic scattering amplitudes of the participating particles with no reference to the mechanism of interaction between them.

The usefulness of this theorem is two-fold: (1) it provides an independent method of measuring scattering amplitudes and (2) it shows that one cannot use the low-energy end of the bremsstrahlung spectrum to test a theory which has correctly predicted the scattering amplitude. (F. E. Low)

VI. Coupling Constant Correction to Pion-Nucleon Interaction

Assuming that the breakdown of charge independence in the pion-nucleon (π-N) system is of electromagnetic origin, then, besides the π\(^0\) - π\(^\pm\) mass difference, there should appear differences between the various coupling constants (π\(^\pm\) - N, π\(^0\) - p, π\(^0\) - n). An attempt to calculate these differences is being made, using \(\gamma_5\) coupling for the π-N system and introducing a Pauli moment term into the Hamiltonian. The following diagrams contribute to lowest order in the electromagnetic interaction:

![Diagram](https://via.placeholder.com/150)

Order of magnitude results can be obtained by choosing separate cutoffs for the Pauli moment and intermediate photon to agree with those which produce the mass difference, as calculated by Feynman and Speisman.\(^1\) (D. M. Greenberger)

VII. Electromagnetic Self-Energy

The self-energy of a Dirac particle coupled through its charge with the electromagnetic field is investigated without perturbation theory (at the cost of assumption 3). The assumptions made are:
1. Relativistic invariance and absence of negative energy states. The propagation function of the electron and its inverse are then

\[ S(p) = - \int_{-\infty}^{+\infty} \rho(a) \frac{i \gamma p - a}{p^2 + a^2 - i\epsilon} \, da \]

\[ S^{-1}(p) = \frac{1}{R} \int_{-\infty}^{+\infty} \rho(a) \frac{i \gamma p + a}{p^2 + a^2 - i\epsilon} \, da \]

where \( R = \left[ \int \frac{a \rho(a) \, da}{p^2 + a^2 - i\epsilon} \right]^2 + \rho^2 \left[ \int \frac{\rho(a) \, da}{p^2 + a^2 - i\epsilon} \right]^2 \)

with \( \rho(a) > 0 \).

2. Dyson's integral equation:

\[ S(p) = S_0(p) + S_0(p) \sum^*(p) S(p) \]

\[ \sum^*(p) = \int \gamma \mu S(p-k) \Gamma^\mu (p-k) D(k) d^4k \]

3. No vacuum polarization and no overlapping graphs:

\( D \longrightarrow \frac{1}{k^2}, \quad \Gamma_\mu \longrightarrow \gamma_\mu \),

\( \sum^*(p) \longrightarrow \int \rho(a) \left\{ A(a) + S_0^{-1}(a) \left[ B(a) + \sum^2(a, p) \right] + S_0^{-2} \sum^{-1}(a)p \right\} \, da \)

in the notation of Jauch and Röhrlich.

The mass shell conditions (physical and bare) in conjunction with 1, 2, 3 give divergence-free integral equations for \( \rho \):

\[
\begin{align*}
\int a \rho(a) \frac{p^2 + m^2}{p^2 + a^2} \, da &= m - \int \int \rho(a) \rho(a') \bar{V} (a, a') \, da \, da' \\
\int \rho(a) \frac{p^2 + m^2}{p^2 + a^2} \, da &= 1 + \int \int \rho(a) \rho(a') \bar{U} (a, a') \, da \, da'
\end{align*}
\]
where $\bar{U}$ and $\bar{V}$ are the symmetric parts of $U$, $V$:

$$(p^2 + a'^2) \bar{U}(aa') = -A(a)(a' + m) - (B(a) + \sum^2_a(a))(aa' + p^2 + m(a - a'))$$

$$- \sum'(a)(a'(a^2 - p^2 - 2am) + 2ap^2 + m(a^2 - p^2))$$

$$(p^2 + a'^2) \bar{V}(aa') = A(a)(a'm - p^2) + (B(a) + \sum^2_a(a)(p^2(a' - a) + m(aa' + p^2))$$

$$+ \sum'(a)[a'(p^2a + m(a^2 - p^2)) - p^2(a^2 - p^2 - 2am)] .$$

$\rho(a)$ should contain a $\delta$-like singularity at the physical mass: $\rho(a) = z \delta(a - m) + \sigma(a)$. It has been shown that

$$\rho(a) = 0 \quad a < m_0$$

i.e. the virtual mass spectrum of this theory is of the type shown in Figure 12.1.

---

**Figure 12.1**

**Spectrum of Virtual Electron Masses**

It is being investigated whether (i) the integral equations lead to such a singularity, (ii) $\delta m = m - m_0$ is finite. (If $\delta m < \infty$ as $m_0 \to 0$, the entire mass of the electron can be attributed to the electromagnetic field.) (iii) (*) remains true when the restrictions (3) are removed.

(G. Sandri)

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**REFERENCES**


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