

**AECD-3830**

**MONTHLY DIVISIONAL INFORMATION MEETING  
ISOTOPE RESEARCH AND PRODUCTION  
DIVISION, Y-12**

**July 21, 1950**

**Carbide and Carbon Chemicals Division  
Y-12 Plant  
Oak Ridge, Tennessee**

**Date Declassified: December 7, 1955**

Photostat Price \$ 9.30  
Microfilm Price \$ 3.60

Available from the  
Office of Technical Services  
Department of Commerce  
Washington 25, D. C.

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Paper 1

INTRODUCTION

By C. P. Keim

A very general review will be given in the following papers summarizing the activities of the various sections comprising the Isotope Research and Production Division of Y-12.

This organization has primarily grown out of a program of separating the naturally occurring isotopes, and occasionally some artificial ones, of all the elements. Our research is closely related to isotopes, or involves problems close to the activities of an isotope-separation program. Most of the various departments or sections in this division were operating in the period between 1944 and 1945 and have, for the most part, merely shifted their activity from uranium to many other elements.

Since the fall of 1945 at least forty of the approximately sixty elements below atomic number 83 (bismuth) having naturally occurring isotopes, mostly stable but occasionally radioactive like  $K^{40}$ , and at least two elements containing artificially radioactive isotopes (Be and U) have been successfully processed in the four calutrons available for this separation work. Such an isotope-separation program as this cannot be done without research and development. Research is being carried on in the closely related fields of inorganic chemistry, physics, spectroscopy, metallurgy, and ceramics because their findings are essential not only to the successful performance of such a broad isotope-separation program as this but because they produce results of fundamental value.

Much of the research on enriched isotopes is being done outside this division—particularly in unclassified fields, as it should be—as is evidenced by a large number of isotopes ranging from a few milligrams to several hundred milligrams each being continually sent to at least fifty different research laboratories.

During 1949 at least sixty references were noted in the technical literature, citing the use of enriched isotopes from this separation program. Many of these articles came from Oak Ridge National Laboratory and the rest came from other AEC projects and from thirty university laboratories. In 1950 thus far over fifty such reports have appeared in "The Physical Review." An outstanding example, I think, is the July 1, 1950 issue in which there appears one article, six letters to the editor, and eleven abstracts from the Washington Physical Society meeting, all on research done by means of enriched isotopes supplied from this program. The ultimate value of making these isotopes available, of course, cannot even be estimated.

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Supplying these isotopes for such basic work is in itself one of our main objectives, and the published reports indicate that that objective is a fruitful one. Another objective is the research and development coming within our own laboratories, including not only engineering improvements in the mass separators but experimentation in the basic fields of science as well.

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Paper 2

CALUTRON OPERATIONS AND ISOTOPE-SEPARATION RESEARCH

By C. E. Normand

The objectives of the Isotope Production and Development Section are (1) production of enriched samples of the naturally occurring isotopes and (2) carrying out such research and development as may be required for this production. Of approximately fifty people comprising the group, roughly 60 per cent are engaged directly in production, some 25 per cent in research and development, and the remaining 15 per cent in such activities as building, shop and office services, and administration.

Production is under the supervision of Leon Love, assisted by Bill Bell in calutron operation and Bill Prater in mechanical servicing. This work is carried out on a continuous three-shift schedule.

A group of ten engineers and physicists are engaged in the research and development activities. These men are George Banic, Bill Cobb, Forrest Duncan, Paul Dunn, Harold Green, Bill Leyshon, Bill Ward, George Wells, Grady Whitman, and Paul Wilkinson. We have the services of Roy Goslin from Oglethorpe and Howard Carr from Auburn during the summer months.

The distinction between development and production work is not always clear-cut. Development, if successfully carried out, leads eventually, and sometimes imperceptibly, into production; production, even when it has become well-established and routine in nature, can contribute substantially to development. It has been estimated, however, that during the past year some 25 per cent of the available calutron time was devoted to research and development and the remaining 75 per cent to isotope collection. Such a ratio is naturally not fast.

Tangible results of this effort were the production, during the past fiscal year, of 103 isotope samples of 25 different elements. Of these 25 elements, 5 (La, Ga, Sm, Nd, and Hf) were processed for the first time. Of those remaining some were run to replenish the inventory, others to provide requested quantities of particular isotopes, and still others to meet some specific requirements of the user. In the last class such separations as that of potassium to produce high purity  $K^{40}$  and of carbon to produce 99.99 per cent  $C^{12}$  might be mentioned.

On combining the production in the past year with that of previous years, it is found that 41 elements have been electromagnetically processed to date. Of the stable elements known to have isotopes only 18 remain to be processed, and these fall into three distinct groups as follows: (1) seven are gases ( $H_2$ , He,  $N_2$ , Ne, Ar, Kr, and Xe), (2) six are rare earths

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(Eu, Gd, Dy, E, Yb, and Lu), and (3) five are platinum-like metals (Ru, Pd, Os, Ir, and Pt).

Elements in each of these groups present similar problems of processing. With the gases there are the problems of retention and contamination from neutral particles. In the case of the rare earths actual availability of the starting material is a problem, and operation at rather high temperatures also complicates the situation. In the platinum-like metals high-temperature operation and neager ion production are the principal problems to be met.

Production figures alone provide an incomplete picture of the work of the group in that they do not indicate the production problems encountered or the supporting research and development work required. Typical development problems and wherever possible the production problems that brought these developments about will be sketched briefly.

#### High-temperature Source Units

Development of high-temperature source units was undertaken primarily to meet the demand in processing the rare earth elements. The principal innovation in this development was the substitution of graphite strip heaters for calrods, thereby raising the maximum operating temperature from about 700 to above 1100°C. This change introduced new problems, however. Heavy a-c heater currents resulted in mechanical failures of heater leads extending across the magnetic field; also, the temperature-regulating thermohm proved quite inadequate as a regulator and unreliable as a measure indicator of charge temperature. Both of these difficulties have been overcome, the first by use of heavy mechanically supported leads and the second by redesign of the thermohm and installation of supplementing thermocouples.

#### Receiver-pocket Shielding

Efforts to collect samples of very high isotopic purity have lead to the development and the adoption of shielding to protect the pocket receiving the wanted isotope from material rejected from adjacent parts of the receiver. Higher purity has resulted from this practice.

#### Collector-slot Spacings

Collector-slot spacings, as calculated by the best available methods, do not always conform to the ion-beam separations actually obtained at the receiver. This discrepancy was particularly noticeable in the collection of Be and Li in the beta channels and of K and Te in the Alpha channels. Apparently this is only one phase of the more general problem of ion-beam pattern at the receiver, and this is being studied both theoretically and experimentally.

#### Vacuum-discharge Scrubbers

The use of H<sub>2</sub>S as charge material in the processing of sulfur and the running of beryllium, together with the prospect of running other toxic

materials, osmium, for example, has lead to the installation of effective scrubbers in all of the vacuum-discharge lines.

#### Charge-evaluation Laboratory

This laboratory in which a variety of calutron type experiments can be carried out on a reduced scale is now in operation. Currently the equipment is being used in close-geometry low-voltage-source studies. Maximum evaluations of the operating parameters of this equipment are 5-kv accelerating voltage, 6500-oersted magnetic-field strength, and  $2\frac{1}{2}$  in. beam radius.

#### Magnetic Centrifuge

A theoretical study has been made of the separative action to be obtained from centrifuging an ionized medium in an axially-directed magnetic field. A summarizing report on this work is now being prepared.

QUESTION: Would you say a word about that? I am afraid I don't understand.

ANSWER: Basically the idea was that if, in centrifuging, the material to be centrifuged contains ions, either gaseous ions or ions in solution, and if the centrifuge is about an axis parallel to a magnetic field, then in addition to the centrifugal action there would be the electromagnetic action of the ion particles rotating in the field. You have the possibility of directing the field and the direction of rotation so that these two forces act either together or in opposite directions. By causing the forces to act in opposite directions, there is ideally the possibility that two different isotopes might be caused to migrate in opposite directions radially. The whole problem, of course, involves all the difficulties of analysis of centrifugal problems plus the added complication of the electromagnetic forces.

QUESTION: But nothing has been built?

ANSWER: Nothing has been built. It is purely a theoretical consideration today.

QUESTION: How does it look? Does it look practical?

ANSWER: It does not look very practical. One of the big difficulties is the fact that to get useful separation we need more ions than we see any way of getting in the material to be centrifuged.

QUESTION: Calculations have been made for liquids too?

ANSWER: Both liquids and gases have been considered. I wouldn't say that completely adequate solutions have come out of all these calculations but they have been considered.

#### Abnormal Isotopic Ratios

In the processing of lithium, abnormal isotopic ratios have been observed both in the monitored ion collection and in samples recovered from various parts of the calutron, including the charge bottle and arc chamber. These ratios are abnormal in that they differ from the natural abundance

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ratio in the initial charge material and cannot be explained in terms as simple distillation and diffusion theory. A series of experiments have been carried out to determine any source of mass discrimination that might account for these ratios. These experiments involve mass analyses of numerous samples collected from various parts of the calutron under various conditions of operation. Partial results, which are now available, give supporting evidence of some sort of mass discrimination in the source unit but are not yet sufficient for giving the source of this discrimination.

QUESTION: Could you say a couple of words about that? I take it that what we are talking about is this arc diffusion.

ANSWER: That has been called arc diffusion, possibly a misnomer, but it is the same thing. The initial observation of this arose from the fact that if the received  $\text{Li}^6$  and  $\text{Li}^7$  were monitored at the receiver during calutron separation, the ratio of  $\text{Li}^7$  to  $\text{Li}^6$  turns out to be on the average over a number of runs a value of about 14.3, whereas the normal ratio of  $\text{Li}^6$  to  $\text{Li}^7$  is of the order of 12.3.

QUESTION: And this is caught where?

ANSWER: This is at the receiver — the 180 deg position — and is, on the basis of monitored currents, scanned and collected. As our experimental work has gone on, we have found peculiar ratios in various parts of the samples recovered from various parts of the equipment. Incidentally, this is not a constant thing. It varies during the run and as better operation has been obtained there has been a tendency for this ratio to approach more normal.

QUESTION: Have you set up an experiment in which you simply have an arc and try to see whether or not there is a separation within the arc itself?

ANSWER: The series of experiments involve operating the calutron in the sense of sending lithium out through the arc chamber, both with and without the arc, and collecting samples that can be compared. Obviously our isotopic analyses need comparison on a more or less statistical basis.

QUESTION: Could you indicate which way the  $\text{Li}^7$  goes preferentially to the  $\text{Li}^6$  in the arc?

ANSWER: So far as the analyses of collections from various parts are concerned, they have been taken from so many places I would really hesitate to start giving data because we have reams of it on that. In general, the effect is that with operation there is an increase in the ratio of  $\text{Li}^7$  to  $\text{Li}^6$  within the charge material left in the bottle. That is to be expected, I think. As a matter of fact in the arc chamber we get this analysis depending on whether we used it as a single chamber or whether we split it up into sections and take samples from opposite ends, so the data are rather complex to analyze there.

QUESTION: Is it that there is multiple scattering going on in the arc and therefore we have a diffusion phenomenon within the arc?

ANSWER: Certainly we have that but we can't pin down the extent to which it is effective. The possibility of sort of a selective absorption or a transmission of the lithium through the carbon parts of the arc chamber has also been suggested. A simple diffusion separation takes place. Distillation



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effects probably must take place as the liquids evaporate from the charge material in the bottle, but there also must be secondary evaporations occurring from the various parts of the vapor channel. Just how these secondary evaporations enter in we can not say definitely on the basis of the returns that have come in.

**QUESTION:** How extensive an effort is being made now in trying to find out more about this arc diffusion?

**ANSWER:** We have two men working on that. They are at present modifying their equipment to make use of a small charge evaluation magnet, which I have mentioned, where the experiments can be carried on more conveniently in the small-scale equipment. The work is in a lull at present because of this transition.

#### High-output Production of $\text{Li}^7$

Finally within the list of major development projects is the problem of high-output production of  $\text{Li}^7$ . The possible importance of  $\text{Li}^7$  in power-reactor development has seemed to justify considerable development directed toward a high-output production of this isotope. The principal objectives of this program have been the development of high-output source units, possibly yielding several amperes of ion current, and the development of receivers capable of absorbing the energy and retaining the material from such sources. Choice of charge material and selection of optimum operating parameters such as accelerating voltage, magnetic field, and beam radius might be considered as secondary objectives.

If the operating experience with standard Beta type units is taken as a point of departure, it appears unlikely that these units can give the desired output unless they are drastically modified. (Present limits are  $\sim 350$  ma per arc.) Experiments with grid type slits in these units have given high unit slit-area outputs of as much as 1.5 amp per square inch, but focusing has been poor at these outputs. Close-geometry low-voltage slit systems have not yet given large focus outputs; however, this scheme offers such great advantages in low power consumption and small energy dissipation at the receiver that work along this line is continuing. Different charge materials and operating parameters have been studied with the standard units. Also certain component parts have been experimentally examined with this unit, such as the substitution of an arc chamber, of insulating material largely, in which the activating filament is placed directly within the arc chamber. Results with this arrangement indicate substantially prolonged filament life.

The double Beta source unit represents a direct approach to the high-production problem and consists essentially of two Beta source units having two arcs each, placed side by side in an alpha tank. Designed to operate on a 10-in. radius, source and receiver are mounted on a common face plate. A single filament placed between the ends of abutting arc chambers serves to support two arcs. Total monitored ion currents of almost 1.5 amp have been obtained from this source. Although promising in its operation, this source is still far from a finished production unit.

A number of experiments have been made with a modified Alpha-2 type source. This unit, having four 14-in. arcs, large charge capacity, and replaceable filaments, offers many features desirable in a high-production unit. Source-heater capacity is barely adequate for operating this unit with lithium metal charge material, and heat dissipation and electrical drains promise to be problems. However, the long 14-in. arcs do operate satisfactorily in a weak lithium field, and for high production the unit offers definite possibilities.

Two aspects of the receiver development will be mentioned briefly. One of these is a defocusing receiver in which an insulated graphite rod is placed inside the enlarged receiver pocket directly in the path of the incoming ion beam. The electrical charge accumulated on this insulated rod acts to disperse the incoming ions, spreading them over the surface of the collector, resulting in lower temperatures and an increase in over-all retention from about 50 up to about 80 per cent. Maintaining insulation of this diffusing electrode is a problem that has not been completely and satisfactorily solved.

The other collector approach is that of using a decelerating voltage on the receiver. This has been done at a decelerating voltage about 10 to 15 per cent less than the accelerating voltage. A good reception is realized, and retention of the received material up to as high as 94 per cent is obtained. This arrangement also offers the possibility that by utilizing the principle of power regeneration the power load on the cubicle supply, which is a limiting factor when outputs reach or exceed about 1.5 amp, can be substantially reduced.

This summarizes briefly the work of the Production and Development Group.

### Paper 3

## ISOTOPE CHEMISTRY SECTION ACTIVITIES

By B. S. Weaver

During the past year alone the Isotope Chemistry Section has performed a variety of functions essential to the separation of the isotopes of some 28 elements and has conducted research and production activities involving synthesis and study of the chemical properties of compounds of most of these and several other elements. This paper is limited to a discussion of recent work with a few specific elements of special interest.

### LITHIUM

The current intensive research by the Division on the production of  $\text{Li}^7$  has made it especially important that charge material of the highest possible quality be supplied to the calutrons. At present it appears that pure lithium metal is preferable to any of its compounds. Since the presence of a small amount of sodium in the best commercial lithium metal causes loss of operating time while it is being pumped off in the tanks, we have undertaken and successfully accomplished the removal of all the sodium. This is done by vacuum distillation from special equipment which accommodates two charge bottles simultaneously. Precise conditions for effective separation have been determined, and the metal produced has greatly increased the calutron operating efficiency. This process, known among us as "denatification," was developed by the charge-preparation group, consisting of Stan Fairbourne, Cliff Sheridan, and Charlie Harrison. This group is called on to supply a pure anhydrous volatile charge material for each element.

The next chemical step in isotope production is removal of all impurities and conversion of the isotope to a useful, stable compound. The impurities comprising most of the common metals, especially Cu, Fe, Cr, Ni, Ag, Si, Ti, Pb, Sn, Mn, Mo, Al, B, and Ca, must be removed by a series of chemical operations, including precipitations, electrolyses, extractions, filtrations, and ignitions. Most of the recent refinement work on lithium has been done by Ken Allen. The success of his efforts is attested by analysis of a recently produced large sample of  $\text{Li}^7$ . This material, weighing 296 g as  $\text{Li}_2\text{SO}_4$ , was found spectrographically to contain: Fe, 0.02 per cent; Mg, 0.01 per cent; Na, <0.15 per cent; and Si, <0.08 per cent.

There have also been numerous small experimental samples which required only partial purification and conversion to iodide for mass analysis. Ken Allen and Frank Scheitlin have set up procedures for the rapid handling of such samples on the milligram scale from various sources.

Since the eventual aim of the lithium-isotope - separation project is to supply pure  $\text{Li}^7$  in the metallic form, the chemists have had to consider carrying the purification much farther to meet as yet unknown specifications, finally reducing a lithium compound to pure lithium metal without loss of significant amounts of the element. The advice of experts from the industrial producers of lithium will be sought on this problem. However, it is hoped that Dr. Normand's group will be able to collect the element in such a way that the major part of it can be obtained directly from the collectors as pure metal.

#### GALLIUM

The rare element gallium has occupied a considerable part of our efforts during recent months. Gram quantities of  $\text{Ga}^{71}$  of high purity, both isotopically and chemically, have been distributed to users, and  $\text{Ga}^{69}$  is now in process of refinement. Gallium iodide, made directly from the elements or by way of the oxide, was found to be a very satisfactory charge material. A very helpful observation made here was the fact that hydriodic acid is a superior solvent for gallium oxide.

Since gallium is a relatively expensive metal, the several hundred grams of it deposited on the inner walls of the calutron tank liner was recovered and separated from most of the large quantities of stainless steel constituents and other elements washed out with it. After further purification in small batches some of this material has been used in other research on gallium.

Gallium isotopes were collected on carbon, which has such remarkable absorbing powers for this metal that the surfaces of the carbon pockets did not show any signs of erosion by the ionic beams nor any visible deposits of gallium. There had previously been fears that the low-melting metal would run out of the pockets and be lost. Ignition of the carbon gave the first evidence that the gallium was still present.

Bill Davis refined the first isotope with considerable effort, partly because of the difficulty of removing sodium introduced to make the oxide soluble. Refinement of the second isotope is proceeding more rapidly as a result of his experience. The final spectrographic analysis of the  $\text{Ga}^{71}$  oxide (10.46 g  $\text{Ga}_2\text{O}_3$ ) was as follows: Al, 0.04 per cent; Mg, 0.02 per cent; Ag <0.04 per cent; Si, <0.08 per cent; and Pt <0.08 per cent.

Milligram quantities of material for mass analysis were conveniently made by dissolving oxide in hydriodic acid, drying in vacuo, and finally sublimating the iodide. Iodides are finding increasing applications in mass analysis as well as in calutron charge material. Frank Scheitlin has become expert in the optimum procedures and conditions for synthesis of milligram quantities of iodides of several elements.

Our interest in gallium chemistry has recently been greatly extended by the receipt of a contract for the synthesis of gallium compounds for use in the cancer-research program of the Medical Division of the Oak Ridge Institute of Nuclear Studies. The aim is to synthesize some compound of gallium which will carry this element through the blood stream and then allow it to be deposited in the bones. In such a form radiogallium would bring a high level of activity in contact with malignant bone growth.

Radioactive gallium has already been used as the citrate in the treatment of bone cancer in human beings. However, the citrate ion is unacceptable to the body because of its power of complexing the calcium in the blood. Its introduction must be compensated for by the simultaneous injection of calcium gluconate. The few other gallium salts of organic acids reported in the literature may be no better.

E. V. Jones, who is here this summer from the University of Alabama, has been attempting to synthesize gallium succinate and ascorbate. Bill Davis is investigating the possibility of double salts containing one of the alkali metals. There is still a long list of unexplored organic acids. While the most direct route possible will be taken toward the very urgent main goal, this research should result in the synthesis of several new and interesting compounds of gallium.

#### ZIRCONIUM

The first zirconium isotopes separated were all found to be contaminated slightly with hafnium. Even this theoretically excellent physical method was incapable of separating this pair of chemically similar elements. Today, as a result of the work of the Chemical Development and Chemical Production and Engineering Divisions and many individuals at Y-12, they are no longer regarded as chemically synonymous in all respects and large quantities of zirconium oxide essentially free of hafnium have been produced. Some of this was available for our most recent separation of zirconium isotopes.

High-quality zirconium tetrachloride was synthesized for charge material by the action of chlorine and carbon tetrachloride on hot zirconium oxide, followed by vacuum distillation of the volatile contaminants.

Bill Davis and Fred Thomas separated the enriched isotopes from their copper collectors and numerous impurities by a combination of several reactions, the most important of which was the precipitation of zirconium tetramandate. Mandelic acid has become a standard reagent in the laboratory, and much experience in purifying both zirconium and hafnium has made us well acquainted with both the effectiveness and limitations of this reagent.

Among other interesting observations is the fact that zinc is partially and titanium completely coprecipitated with zirconium, though neither of them comes down alone under the conditions used for zirconium precipitation. Complete separation from the impurities introduced by calutron operations requires a combination of several purification steps, with the mandelate precipitation the last in the series. Typical analyses of isotopes refined in this manner are: Zr<sup>90</sup> (16.25 g ZrO<sub>2</sub>), Pt, 0.04 per cent; Hf, 0.005 per cent; and Zr<sup>94</sup> (7.27 g ZrO<sub>2</sub>), Cu, 0.02 per cent, Fe, 0.02 per cent, Pt, 0.08 per cent, Mg, 0.005 per cent, Si, 0.08 per cent.

You will notice that there is still a trace of hafnium. This is due to the fact that Hf<sup>180</sup> doubly charged equals a mass of 90 in the calutron. A quantity of normal zirconium oxide, prepared especially for spectrographic standards, had no detectable impurities except 1 ppm boron, but, if the final step had been done in platinum, that might have been absent.

Our most extensive zirconium research project has been in connection with the production of zirconium tetrafluoride for use by the Materials Laboratory

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as raw material for a new method of producing the metal. The fluoride had to be free of oxide and any considerable amounts of most other elements. Jo Williams, with the help of other members of the section and of chemical operators, has succeeded in supplying the needs for very pure  $ZrF_4$  and at the same time has conducted experiments which have greatly extended our knowledge of the reaction between  $ZrO_2$  and HF, which does not go to completion under any known conditions.

By the aid of the x-ray diffraction facilities in the Physics Section, two intermediate compounds have been detected. One of these is believed to be  $ZrOF_2$  and the other has a F-Zr ratio of 3. Is such a compound as  $ZrOZrF_6$  possible? (This would be somewhat parallel to  $U_3O_8$  with F replacing O.) The latter compound has been observed only once and has not been studied further.

The compound  $ZrOF_2$  is produced by a reaction between  $ZrO_2$  and  $ZrF_4$  at temperatures above  $650^\circ C$  and reacts with HF only at lower temperatures. While most of our pure  $ZrF_4$  was produced by incomplete hydrofluorination of  $ZrO_2$ , followed by vacuum sublimation in separate equipment, the observed chemical properties of  $ZrOF_2$  will probably make possible the production of  $ZrF_4$  in a single reactor by successive reactions at different controlled temperatures. There is much room for additional research on this problem. A preliminary research report, Y-619, has been written on our past work.

#### HAFNIUM

The separation of hafnium isotopes was made possible by the production of high-quality hafnium as a by-product of the zirconium-purification project. Several kilograms of  $HfO_2$  containing only small amounts of  $ZrO_2$  was produced at our request by reprocessing the hafnium-rich effluents from the zirconium plant. Part of this oxide was separated from other impurities, especially titanium, and converted to  $HfCl_4$  by the charge-preparation group. Chemical refinement of the enriched isotopes is now in process. Procedures used in the refinement and mass analysis of zirconium are applicable to hafnium.

Hafnium tetrafluoride has also been produced in our laboratory and used by the Materials Laboratory in the production of unusually large amounts of hafnium metal. Hafnium oxide apparently reacts with HF more readily and completely than does  $ZrO_2$ .

The availability of the first large quantities of pure compounds of both zirconium and hafnium should make possible more accurate determinations of some of their physical properties. Work on some of the more easily determined properties is now under way in the Chemistry Section. Bob Bailey has made preliminary preparations for the redetermination of the densities of both  $ZrO_2$  and  $HfO_2$  and for a comparison of the solubilities of the tetramandelates of the two elements in water. This work has been crowded into his regular schedule of refining the isotopes of such elements as lead, zinc, and titanium.

Some time has already been spent in testing a new method of determining the relative amounts of zirconium and hafnium in mixtures. This method consists of weighing the material first as the tetramandelate and then as the

oxide. The ratio of molecular weights of pure zirconium mandelate and zirconium oxide is 5.6463, that for the corresponding hafnium compounds is 3.7184. Mixtures of the two have intermediate ratios. This considerable difference in weight ratios and the high purity attainable by precipitation of the mandelates permits more precise analysis of such mixtures than does any other known chemical method. Analyses of some test samples by Bill Davis have given excellent results, and others have pointed out the care necessary in washing, drying, and igniting the mandelate. We expect to publish the results of the research soon.

It is interesting to observe that the presence of titanium gives high results for the hafnium content. This indicates that while titanium is apparently coprecipitated quantitatively, it comes down as some compound which has a much lower molecular weight than does titanium mandelate. At present we do not know what that compound is.

#### CARBON

Preparation of charge material for the separation of the isotopes of carbon was no problem since the requirement of a volatile compound was easily satisfied by carbon tetrachloride. Chemical refinement of the collected elemental carbon, leaving it in the elemental state, is a much more difficult and still unsolved problem. The impurities are laid down among the atoms in the carbon flakes and cannot be removed by aqueous reagents. Preliminary experiments with one sample achieved removal of most of the copper, the chief contaminant, by chlorination at temperatures up to 1100°C. Extreme purity may be attainable by chlorination at much higher temperatures, followed by high-temperature hydrofluorination, but equipment necessary to perform these operations without loss of carbon has not been devised yet.

Louis Gilpatrick has already prepared samples for mass analysis of numerous collections of carbon. The carbon was submitted for analysis as CO<sub>2</sub> after taking great precautions to prevent the introduction of any natural carbon.

Gilpatrick has also served the Physics Section by preparing a long series of samples of cyanogen chloride from potassium cyanide containing known amounts of C<sup>13</sup>. These samples were used in the standardization of microwave equipment for use in the mass analysis of carbon. At present he is attempting to synthesize methyl mercury chloride for similar use in the study of mercury isotopes. Frank Scheitlin is synthesizing tungsten carbonyl for use in Dr. Baldock's studies of ions. Carbonyls of various elements are of interest for use as possible charge materials in the calutron and other electromagnetic equipment. The synthesis of these compounds will be considered as a part of the growing function of preparing unusual compounds for research by all parts of the Division.

#### RARE EARTHS

This Division has long been interested in the separation of the isotopes of the rare earths and expects to progress as fast as suitable materials become available. Separations have already been made of the isotopes of cerium,

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lanthanum, neodymium, and samarium. Oxides of all these elements of sufficient purity were available on the market, and anhydrous chlorides were prepared in the charge laboratory.

Sufficient europium has recently been offered for sale at an unprecedented low but still prohibitive price, \$20,000 per 500 g. Concentrates high in gadolinium have been obtained on contract with the Lindsay Light & Chemical Co. Bill Burns and Fred Kappelmann are now busily engaged in attempting to separate the middle fraction of gadolinium and samarium by fractional crystallization of rare earth magnesium nitrates. These two elements will then be separated by reduction of the samarium. If this is not too difficult there may be a chance of going on to obtain dysprosium from the same or similar concentrates. The hopes of some for separated isotopes of lutecium must remain, at present, a dream of the distant future.



Paper 4a

ISOTOPE PHYSICS AND SPECTROSCOPY RESEARCH: INTRODUCTION

By L. E. Burkhart

In the Isotope Physics Section there are fifteen technical people: three have Ph. D. degrees and the others have Master's and Bachelor's degrees.

It might be said that the objective of the Isotope Physics Section is to study the properties of molecules, atoms, and nuclei. It is granted that this is a broad objective, but we approach that through our techniques as applied to the electromagnetic-spectrum projects which are developed within the laboratory. There is the responsibility for a Y-12-plant-wide service in spectrochemical analysis and x-ray diffraction analysis.

The Isotope Physics Section, in the calendar year 1949, turned out 24 reports, 16 of which were unclassified and 8 of which were classified. Seven of the unclassified reports were published in the open literature.

Figure 1 shows our program as it is related to the electromagnetic spectrum.

At the top is seen the spectrum, frequency being in cycles per second. The wave lengths are given below. The solid portion on top is the spectrum, as a whole divided into regions applying to our program. The solid section below it is the scope of our program at present.

In the case of nuclear induction, shown here in megacycles and meters, the first figure shows what our active range is, the second, what our capacity is with our equipment. The same is true with microwave. The top number given shows what our present capacity is and the second number shows what our ultimate capacity might be.

This chart is not meant to convey the impression that we expect to cover the whole electromagnetic spectrum. On the contrary, we expect to cover only that part of it which applies to our own capabilities, equipment, and program.

QUESTION: You have equipment for the whole range?

ANSWER: We have equipment for the range that is indicated by the second number in the lower portion of the figure. There are a lot of blanks in the spectrum, however.

These upper sections should not convey the amount of emphases because in the optical spectroscopy region, which is very small, there is considerable emphasis in our program and a lot of interest.

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I would like to acknowledge the cooperation of the X-10 Chemical Division, which has been very helpful to us, especially Fred Fitch, who has helped us with the handling of plutonium. We are in the process of getting facilities to handle material like plutonium and some of the transuranic elements, and we expect to call on our good neighbors some more to help us.

From this paper you will be able to see where we can attack problems and observe properties of these elements in our program.

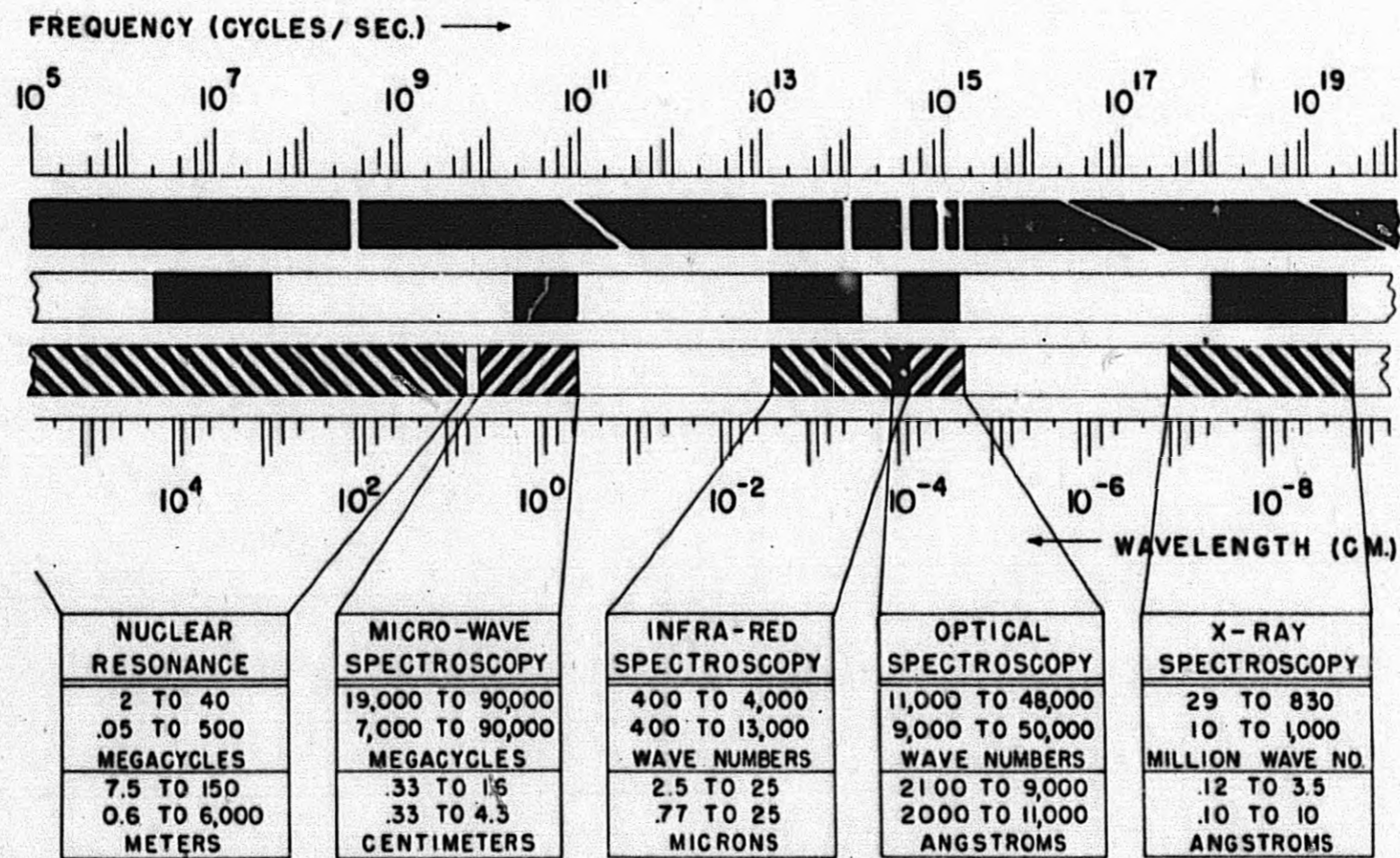


Fig. 1—Regions of electromagnetic spectrum covered by the Isotope Physics and Spectroscopy Group.

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Paper 4b

ISOTOPE PHYSICS AND SPECTROSCOPY RESEARCH: ATOMIC SPECTROSCOPY

By J. R. McNally, Jr.

This paper will discuss two of the small sections shown in Fig. 1 of Paper 4a, namely, optical spectroscopy and one slightly larger but not covered by us as much, x-ray spectroscopy.

The breakdown of our work, as has been mentioned, covers services and research in the field of spectrochemical analysis. We handle approximately 650 samples a month for about 4000 element determinations. Some of the important samples have been mentioned: hafnium, zirconium, beryllium, lithium, all the raw materials of the calutron group, and all their finished products for chemical analysis.

Messrs. Spitzer and Smith are in charge of the people working in the spectrochemical laboratory and all the credit really devolves on them.

In the field of x-ray spectroscopy, besides a certain amount of service work such as identification of crystal structures by means of x-ray diffraction, there has been some emphasis on the use of the L-3 absorption edge of uranium as a possible means of analyzing solutions for uranium content.

Past work in the field of x-rays has covered the obtaining of the characteristic K and L spectra of such elements as 43 and 61, which were prepared in the X-10 laboratory, and element 84, which was prepared in one of the other AEC laboratories. The x-ray work is being carried on by Messrs. Peed and Dunn, and again all the credit devolves on them rather than on the supervisors. Dr. Bennett from the University of Louisville has been working with the x-ray group this summer.

In the field of optical spectroscopy, we cover perhaps our largest multitude of sins. We are doing work in the following fields: high-resolution spectroscopy, wave-length measurement on the heavy elements, and isotope shift displacement. Perhaps the best way to discuss our work in these fields would be to refer to the figures.

Figure 1 shows some of the standards which we use in the analysis of zirconium material for hafnium in purity. Actually, you cannot follow the hafnium line all the way down, but it illustrates the scope of coverage in terms of parts per million, etc.

Figure 2 shows some of the past work which has been reported at least orally. It illustrates the hyperfine structure of one of the many lines of the  $U^{235}$  isotope. The predicted pattern for a nuclear spin of  $I$  equals  $5/2$ . When a quadrupole correction is introduced it agrees quite well with the actual observed pattern, whereas a spin of  $7/2$  or larger, or  $3/2$  or smaller, definitely cannot fit the pattern.

Figure 3 illustrates the purity of the Hg-202 sample which had been separated by the calutron group. On the left is the Fabry-Pérot interference pattern of natural mercury, showing both isotopic structure and hyperfine structure of the odd-mass component. On the right is the Hg-202 sample which was better than 98 per cent pure. This has also been reported in at least one or two scientific meetings.

Figure 4 illustrates some of the emphasis being placed on uranium. The relative positions of certain isotopic lines,  $U^{238}$  down through  $U^{233}$ , are shown. Two of those,  $U^{233}$  and  $U^{236}$ , are not naturally occurring, and the other three are naturally occurring. This study has been important in terms of the possibility of assaying uranium for constituent isotopes by means of a rather cheapened spectroscopic method. It has done a fairly good job, and we hope, possibly with an electronic technique, to improve accuracy and make it even more useful.

QUESTION: Is there considerable isotope effect in these lines?

ANSWER: There is considerable isotope effect but not too much hyperfine structure in this particular line.

QUESTION: I am talking about the isotopes.

ANSWER: This is not the actual pattern you would see on a plate; this is schematic showing relative positions only.

QUESTION: But the positions are different from what you would get just from the reduced mass?

ANSWER: Yes.

QUESTION: How much is the discrepancy here?

ANSWER: A factor of 0.0010 over what you would expect for reduced mass plus nuclear motion correction. It is an entirely different type of effect in this case, having to do with the volume of the nucleus rather than the motion of the nucleus. It is found that beginning about the center of the periodic table you have a very small nuclear-motion effect and you have the beginning of a nuclear-volume effect, which tends to increase with increase in mass number, so that by the time you get to uranium, plutonium, or thorium, the separation effect is fairly reasonable.

QUESTION: Could you tell us of the status of comparison between theories and experiments on the isotope effect now?

ANSWER: Most of it has been empirical with the exception of hydrogen-like atoms where you can get fairly good measurement of what the wave function of the particular electrons may be at the nucleus. In this case, where you have electronic structure of the type of  $f^3 ds$  or  $f^3 s^2$ , the corrections which come in are apparently such that you cannot make a careful theoretical calculation. In the case of the light elements, the effect of nuclear-motion calculations have been made not only for the simple case of hydrogen but for two- and three-electron spectra and I believe, also, that some calculations were made on magnesium some 8 or 10 years ago.

Figure 5 shows the isotope-shift effect in the lithium resonance doublets. This investigation was carried out on separated isotopes  $Li^6$  and  $Li^7$ , and you will note that the fine structure or doublet splitting in either the upper or lower section is plainly identical (and actually turns out to be identical, within a few per cent) with the isotope shift.

I hope to carry out more careful measurements of these shifts because there has been an indication that the fine-structure splitting is slightly different in the two isotopes. This has been published previously in the literature, and with enhanced isotopes it might be possible to answer that question successfully.

Figure 6 shows the beautiful isotope shift that is obtained when use is made of end masses, that is, the highest and lowest masses in enhanced state, and all those in between are eliminated. It also points out the complexity of line spectra. When you visualize that a spectrum line of a mixture of samarium isotopes will be of the order of this breadth, you have quite a problem in measurement and classification of spectra, which is one of our aims - to separate isotopes and obtain beautifully sharp spectrum lines.

Figure 7 shows isotope shifts between  $\text{Th}^{230}$  and  $\text{Th}^{232}$  spectrum lines. Here the sample contained about 13 per cent  $\text{Th}^{230}$  (this was sent to us by Dr. Manning of Argonne) and, I believe, the thorium was separated from naturally occurring uranium material. This indicates a possible origin in  $\text{Th}^{232}$  as a decay from uranium elements and possibly indicates that  $\text{U}^{236}$  did exist in some prehistoric time.

The work of the optical spectroscopy group is being done by quite a few people: Messrs. Griffin, Strukenbroeker, Yonts, Werner and Smith. I hope that I have not omitted anyone.

Our biggest project, if you wish to look at it that way, is to extend the classification of spectra of the heavy elements. This is important if it is desired to evaluate such things as nuclear magnetic moment, even on a semiquantitative basis. To this end we have under construction a better electromagnet with which we hope to obtain a magnetic field of about 100,000 gauss; although it will be designed to possibly give 200,000 gauss. We hope to work at 100,000 gauss and obtain Zeeman effects of spectra of the heavy elements.

We have obtained very good spectra for plutonium, thorium, and uranium with samples of the order of 100  $\mu\text{g}$ . We should be able to obtain fairly reasonable Zeeman effects of the more intense lines and hence identify characteristic quantum numbers of these transuranic elements.

In addition to carrying out measurements on these heavy elements which have multitudinous line spectra, it is important, especially in the Zeeman effects where that number is multiplied by anything from 10 to 30 (Zeeman components), that there be an accurate and rapid method of measuring these data. We cannot wait until infinity to get all these answers; we want to get some today or next week, and to that end we are designing and considering the construction of an automatic comparator which should carry out most of that routine work of hand and eye use of comparators.

Mr. Werner is doing most of the work on that, and it will tie in the use of facilities of the International Business Machines Corp. and carry out such things as calculation of wave numbers from wave lengths in air and numerous other features, such that we hope the operator can put a plate on the automatic comparator, smoke a cigarette, and come back half an hour later and have an answer for the whole plate of perhaps 2000 or 3000 spectrum lines.

The measurement of the spin of  $\text{Xe}^{135}$  is pretty stagnant now and has been for some time. Although we have equipment which can be set up and used, we

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have been limited by the available samples. At present we have no  $\text{Xe}^{135}$  or xenon samples available for study.

We have done some study on  $\text{Ca}^{43}$ , which was separated at Y-12. We have done some studies on rhodium, which is 100 per cent isotopic pure and has no spin listed in the literature.

At present we have gone to very high orders of interference, and, although there is an indication of structure there, the indications are that the nuclear magnetic moment is very small. We have no idea as to the spin. The same might be said of  $\text{Ca}^{43}$ , which is one of the lighter isotopes for which the spin has not been determined.

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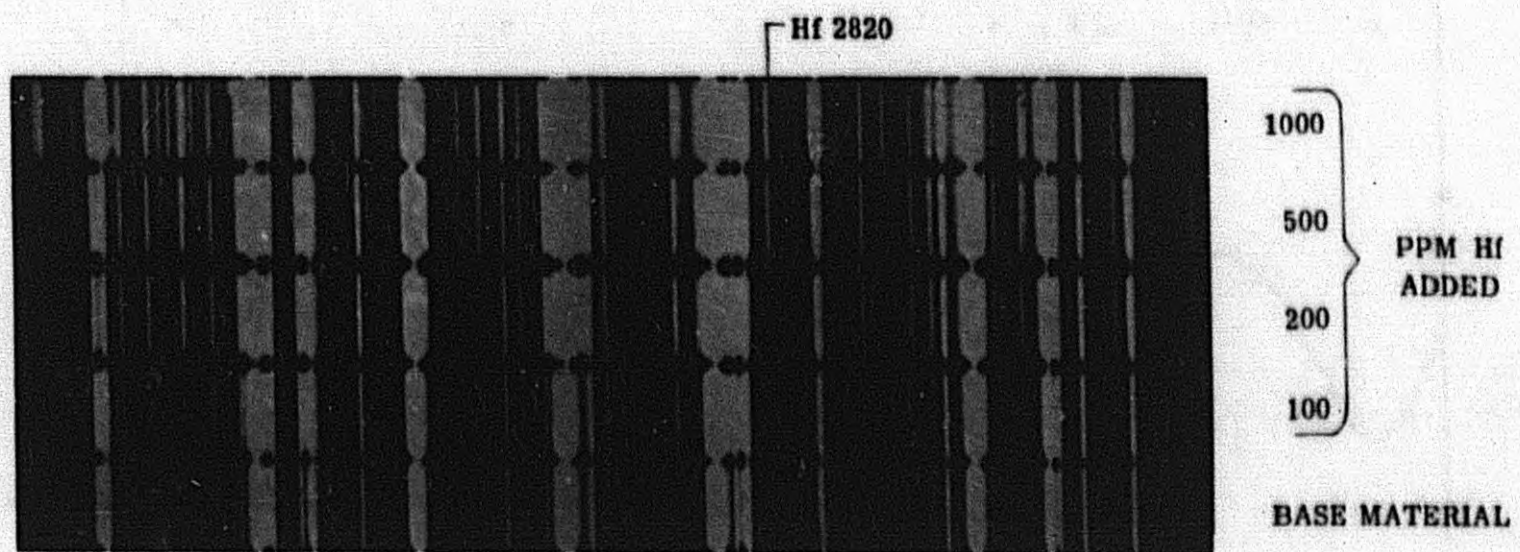


Fig. 1--Hafnium in  $ZrO_2$  standards (Y-12 material).



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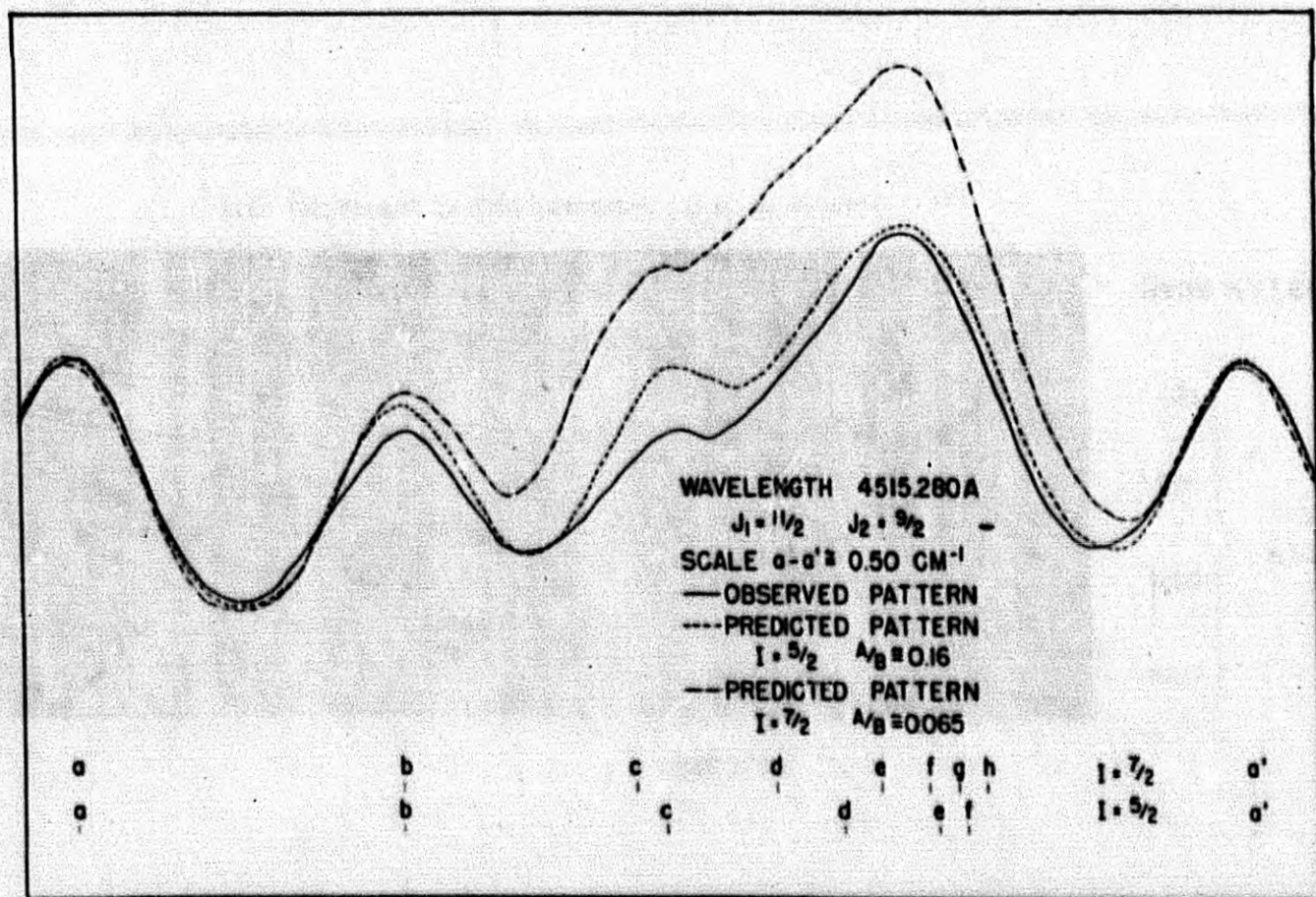


Fig. 2—Hyperfine structure in  $U^{235}$ .

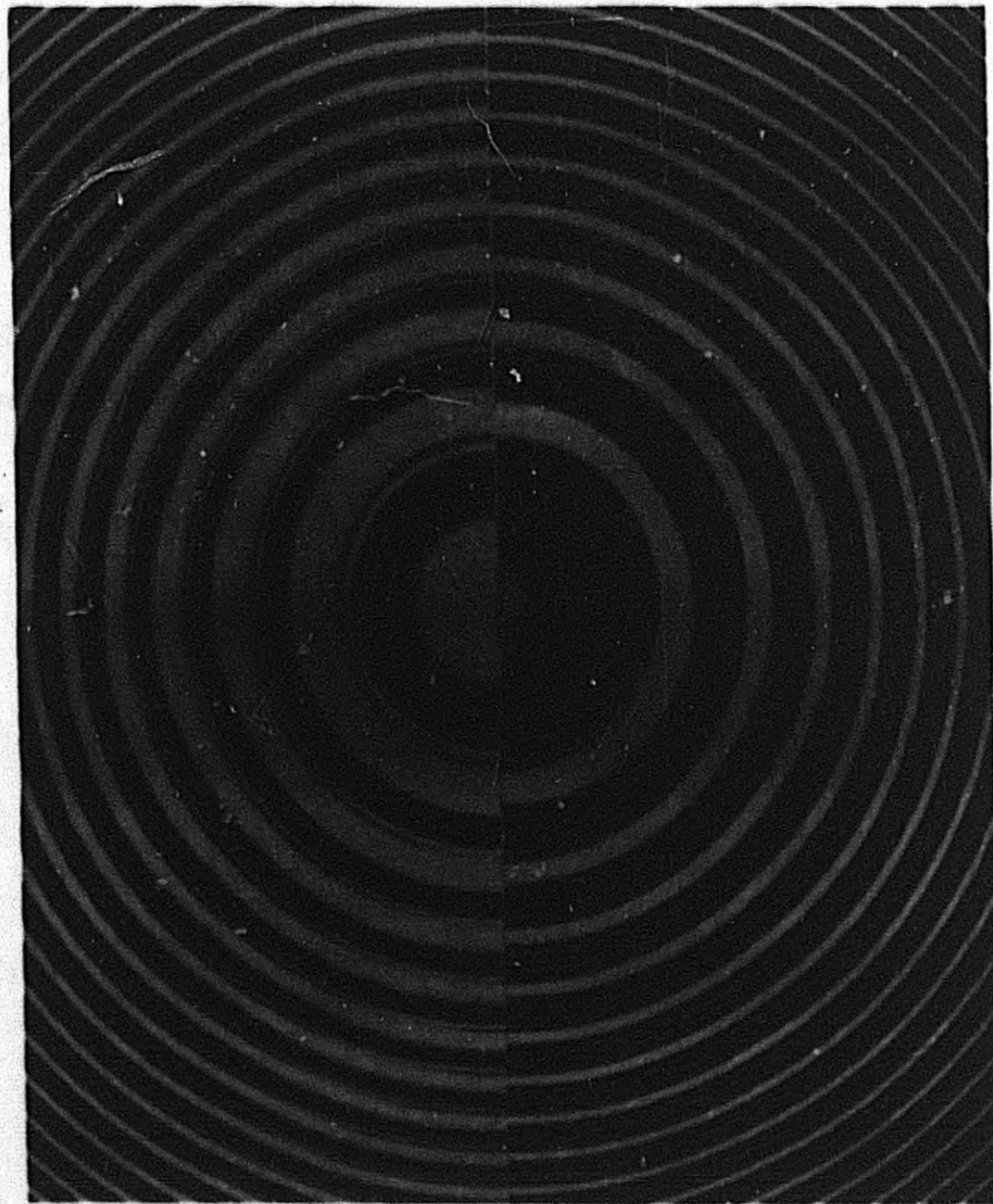


Fig. 3--Natural and artificial mercury (5461A).

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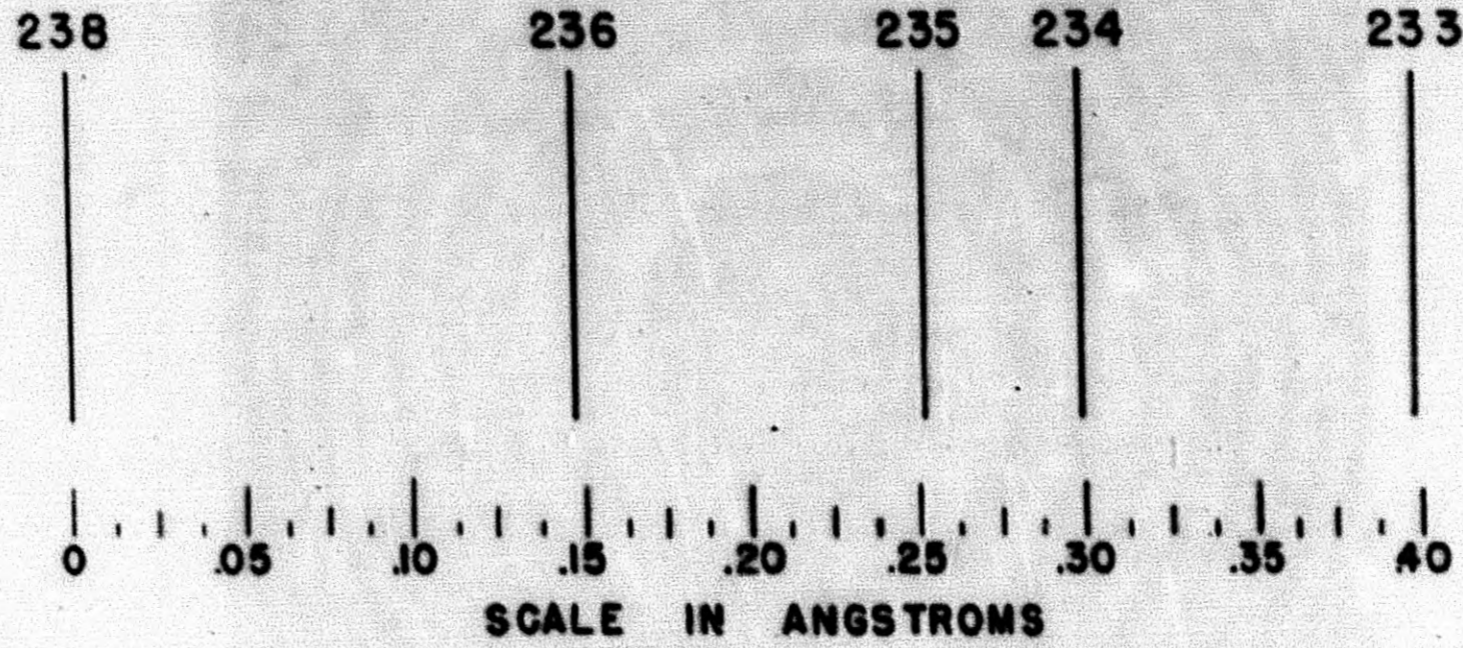


Fig. 4--Isotope structure of uranium line 4244A.

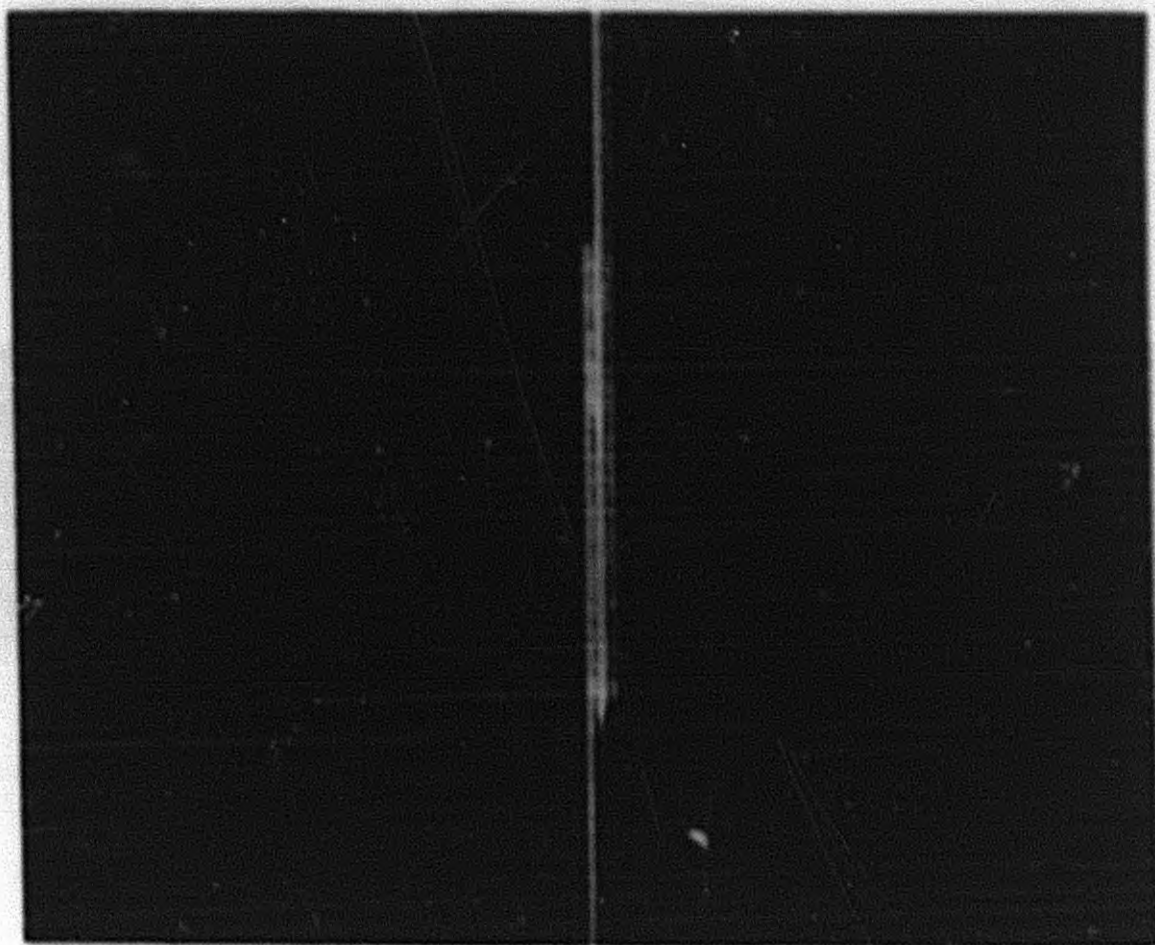


Fig. 5--Isotope shift in lithium resonance  
doublet at  $\lambda$  6707A.

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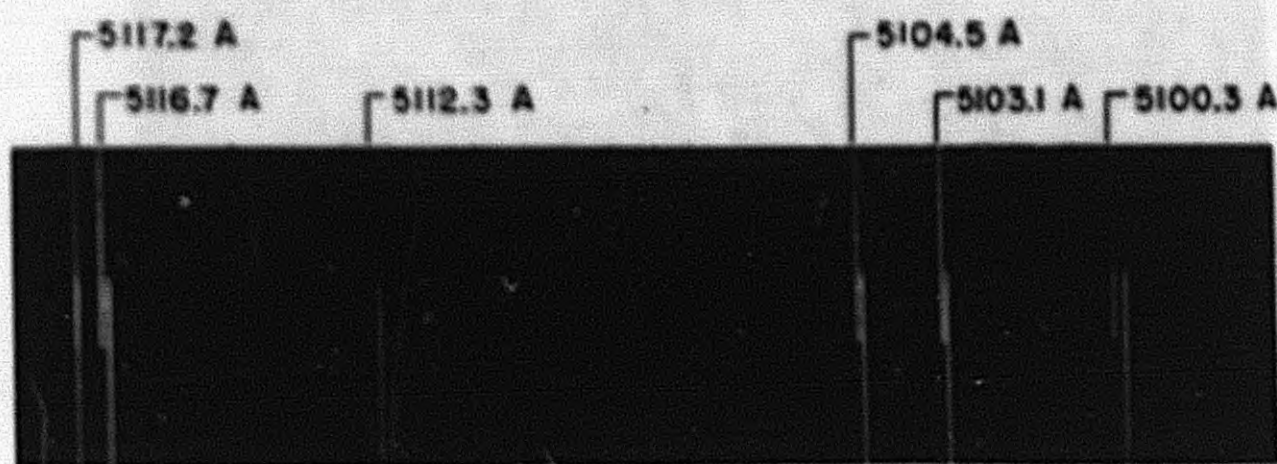


Fig. 5—Spectrum shifts in enhanced samarium samples. Top spectrum, Sm<sup>144</sup>. Bottom spectrum, Sm<sup>154</sup>.

4085.030

4086.520

4094.751

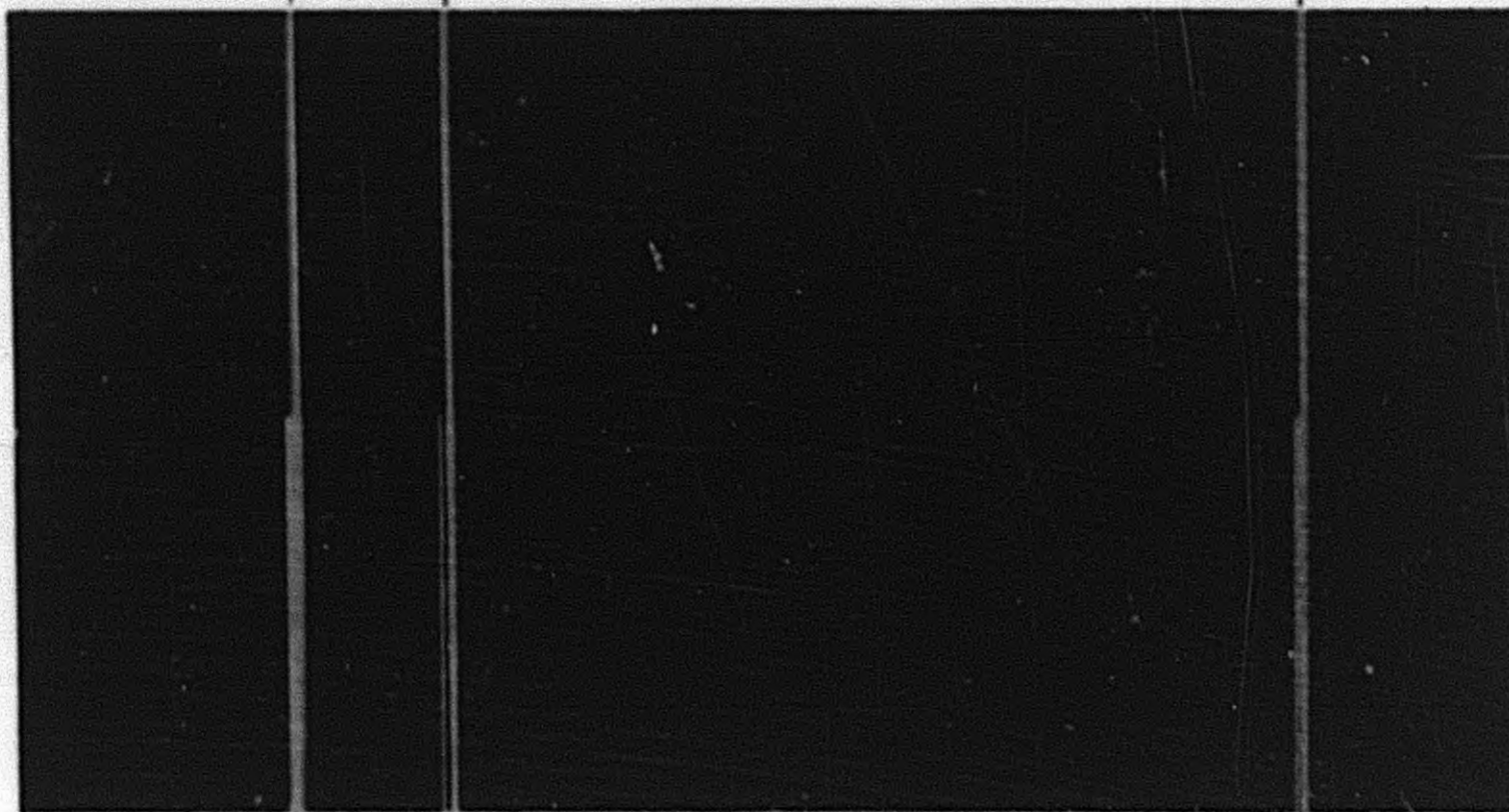


Fig. 7—Isotope shifts between  $\text{Th}^{230}$  and  $\text{Th}^{232}$  spectrum lines.

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Paper 4c

ISOTOPE PHYSICS AND SPECTROSCOPY RESEARCH:  
NUCLEAR INDUCTION AND MICROWAVE SPECTROSCOPY

By G. W. Keilholtz

Harold Walchli and Roy Zimmerman are working on the nuclear-induction program. The program has been one of equipment and technique development and will be applied to the analysis of isotopes, particularly uranium. In addition, studies are to be made of the heavy elements and the rare earth elements.

A sample sealed in its container can be examined by nuclear induction methods and returned in the original form without loss of material. Nuclear-induction methods are applied to isotopes with odd mass, except in cases of odd-odd structures such as  $H^2$ ,  $Li^6$ ,  $B^{10}$ ,  $N^{14}$ ,  $Na^{22}$ ,  $F^{19}$ , and  $Lu^{176}$ . Samples can be solid, liquid, or gas. The liquid samples are preferred for optimum conditions of concentration and signal response.

The present equipment includes a locally constructed instrument with a 16,000-gauss magnet and an instrument constructed by Varian Associates, Inc. having a 12,000-gauss magnet. Operating frequencies are available from 3 to 9 megacycles.

The nuclear-induction spectrograph, with associated magnetic and control equipment as received from Varian Associates, has undergone some modifications for research with  $U^{235}$ . A fixed frequency of 2.5 megacycles and magnetic scanning are now being used for the investigation.

The work on  $U^{235}$  should give further information on fundamental properties of the nucleus, i.e., spin and magnetic moment. The application of nuclear-induction developments to the gaseous-diffusion process or reactor control is a possibility. Stable isotopes will also be studied in the present program.

A. L. Southern and H. W. Morgan are working on the microwave program, which is active in molecular spectroscopy and isotopic analysis.

Molecular spectroscopy in microwaves includes the determination of both molecular and nuclear properties. With the availability of the high resolution the effects of isotopes are very pronounced in microwave spectra. Molecular-structure studies include frequency determination, moments of inertia, isotope effects, nuclear spins, quadrupole effects, and Stark effects for linear, symmetric-top, and asymmetric-top molecules.

Microwave techniques require materials in a gaseous state, and the molecule must have a permanent dipole moment. Quantitative measurements involve absorption of energy at fixed frequencies similar to the techniques used in infra-red. The resolution is high enough to give lines with intensities dependent on isotopic concentrations.

The microwave spectrograph has been applied to the isotopic analysis of  $N^{15}$  in ammonia and  $C^{13}$  in cyanogen chloride.

Table 1--Nuclear-induction Signals

| Sample                        | Isotope           | Magnetic field,<br>gauss | Frequency,<br>megacycles | Signal to noise<br>ratio |
|-------------------------------|-------------------|--------------------------|--------------------------|--------------------------|
| H <sub>2</sub> O              | H <sup>1</sup>    | 1,550                    | 6.58                     | 150:1                    |
| D <sub>2</sub> O              | H <sup>2</sup>    | 11,000                   | 7.16                     | 200:1                    |
| P-10 wax                      | P <sup>19</sup>   | 1,600                    | 6.40                     | 60:1                     |
| KI                            | I <sup>127</sup>  | 7,500                    | 6.36                     | 25:1                     |
| AlCl <sub>3</sub>             | Al <sup>27</sup>  | 6,200                    | 6.88                     | 15:1                     |
| NaAsO <sub>2</sub>            | Na <sup>23</sup>  | 5,700                    | 6.41                     | 20:1                     |
| V <sub>2</sub> O <sub>5</sub> | V <sup>51</sup>   | 5,800                    | 6.48                     | 12:1                     |
| LiNO <sub>3</sub>             | Li <sup>7</sup>   | 3,490                    | 6.52                     | 60:1                     |
| LiCl                          | Li <sup>7</sup>   | 3,490                    | 6.52                     | 90:1                     |
| Cu                            | Cu <sup>63</sup>  | 4,400                    | 5.00                     | 44:1                     |
|                               | Cu <sup>65</sup>  | 4,200                    | 5.00                     | 26:1                     |
| NaBr                          | Br <sup>79</sup>  | 4,200                    | 5.02                     | 35:1                     |
|                               | Br <sup>81</sup>  | 5,600                    | 5.02                     | 50:1                     |
| RbCl                          | Rb <sup>85</sup>  | 12,000                   | 5.00                     | 35:1                     |
|                               | Rb <sup>87</sup>  | 5,400                    | 7.80                     | 30:1                     |
| Tl(Ac) <sup>+</sup>           | Tl <sup>203</sup> | 3,000                    | 7.82                     | 15:1                     |
|                               | Tl <sup>205</sup> | 3,000                    | 7.91                     | 35:1                     |

Table 1 shows the number of compounds which have been tested on the present nuclear-induction equipment. The equipment will operate between 3 and 9 megacycles. The P-10 wax, the V<sub>2</sub>O<sub>5</sub>, and the copper represent solid materials; the remainder represent liquids or solutions. The solutions run from 1N up to saturation. These are not new compounds but represent a trial run on our nuclear-induction equipment.

QUESTION: How long does it take to get a signal when you place a sample in the equipment?

ANSWER: It is possible to get them in just a few minutes if you know where to tune the equipment.

QUESTION: This is with magnetic catalysts?

ANSWER: Yes.

QUESTION: The reason I ask is because I understand some of the X-10 people went over to Y-12 about the general question of whether these methods are at all practical as a straightforward chemical analytical method.

ANSWER: I believe you will find that they are, providing you have your series of frequencies prepared ahead of time and just tune to the particular frequency for the particular isotope.

QUESTION: Was this the general consensus, that the thing might be made into a practical spectroscopic analytical method?

ANSWER: We have not gone at it from that angle. We are going at it by studying ratios, spin, and magnetic moment and with the possibilities of using



the intensity of the signals as a quantitative measurement. But it could be used as a qualitative instrument.

QUESTION: I was thinking of it as a quantitative analysis but I was wondering how, considering the difficulties in getting the signals, it would compare with the optical-spectroscopy method as a straightforward method?

ANSWER: I believe it would just be a matter of tuning the dial to a particular frequency.

QUESTION: So you do think it might be a practical scheme if it worked out?

ANSWER: I think it will take considerable work, but I think it would go in that direction.

QUESTION: You do feel though that the difficulties in finding the resonance and tuning in, etc., are not prohibitive?

ANSWER: No, they are not, but there are so many little variables that creep in that it is going to take some time to iron it out of the electronic equipment.

QUESTION: Do I take it then that you have not found a signal from any of the uranium isotopes?

ANSWER: We have not found a signal. Searches have been made on solid uranium fluoride and on solutions of uranium sulfate, but we have not seen a signal.

QUESTION: Have you looked for  $U^{233}$ ?

ANSWER: No.

Figure 1 shows a recorder trace of the  $N^{15}$  in ammonia. Notice the comparisons: there are three separate concentrations, going from 0.38 to 1.3 and 2.5, in which the  $N^{14}$  is compared to the  $N^{15}$ .

Figure 2 is similar to Fig. 1 for  $C^{13}$  and chloride in which the measurements are again made over with over-all peak.

Figure 3, taken from the experimental data, shows the straight-line graph running again from 0.38 up to 4.3 per cent. Experimentally this has been a straight line and is being used as a calibration curve in the analysis of  $N^{15}$ .

Figure 4 is an experimental standardization curve for  $C^{13}$ . From normal abundance to 1.1 up to 5 per cent, there is a straight-line relation. We have experimental results up to 10 per cent. There is a slight curve due to nonlinearities in the electronic system.

Isotopic analysis by microwave techniques in the present stage of development does not meet the high precision of a Nier type spectrograph. However, routine results for carbon or nitrogen by microwave methods would be  $1.00 \pm 0.03$  and the  $1.000 \pm 0.003$  for the Nier mass spectrograph.

An investigation of volatile metallic compounds is under way for such compounds as  $VOCl_3$ , methyl mercuric chloride, and lithium ethyl.

In order to work with lighter molecules such as lithium methyl and carbon monoxide, it is necessary to extend the frequency range considerably. Work is being directed to 40,000 megacycles; plans are then to proceed to 60,000 megacycles with klystrons. The range of 60,000 to 90,000 megacycles will require the use of second and third harmonics of lower-frequency tubes.

The nuclear-induction and microwave programs are planned for both practical applications and fundamental research, and we shall have plenty of each this coming year.

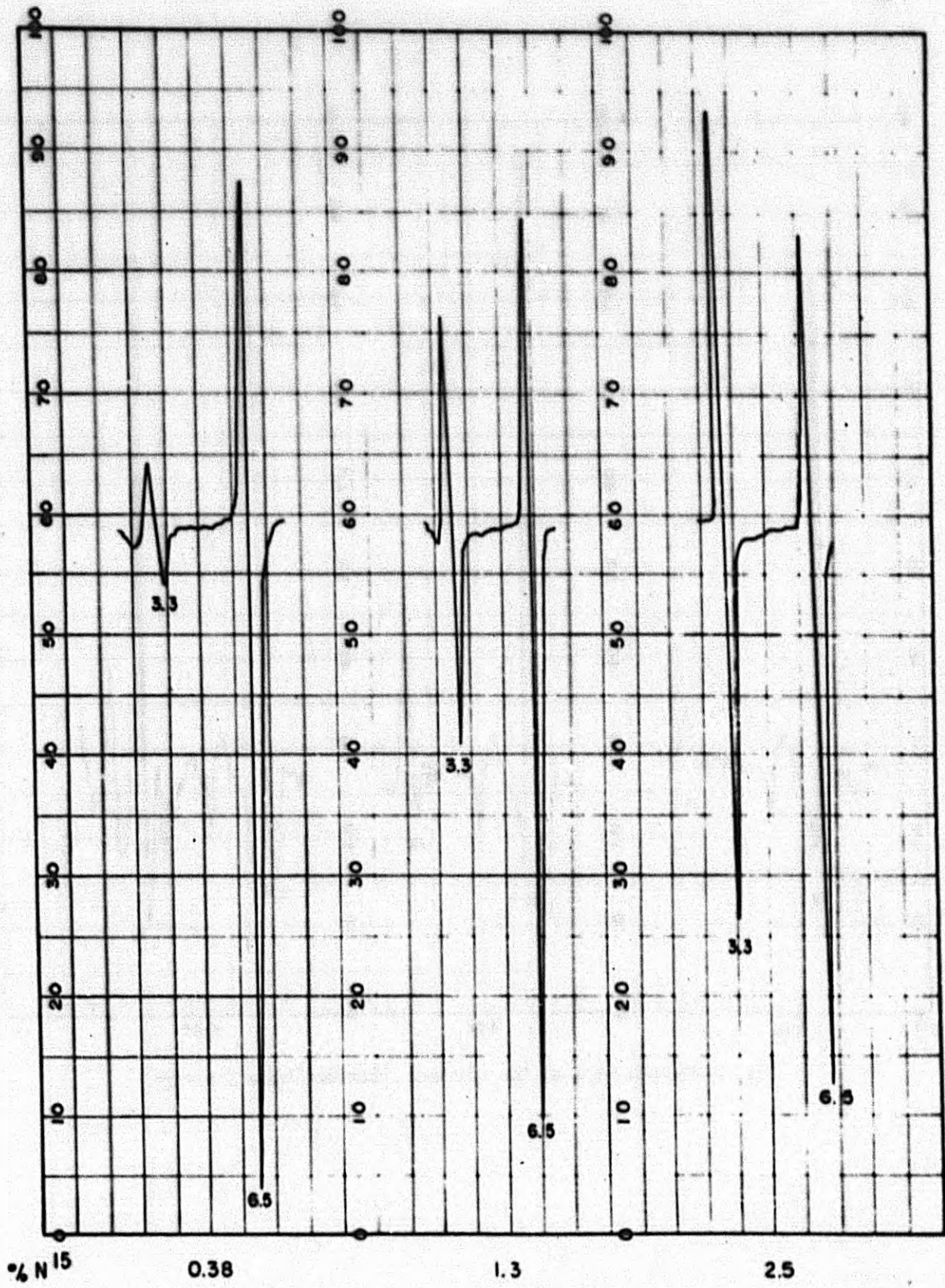


Fig. 1—Recorder traces for ammonia (125 μ pressure).

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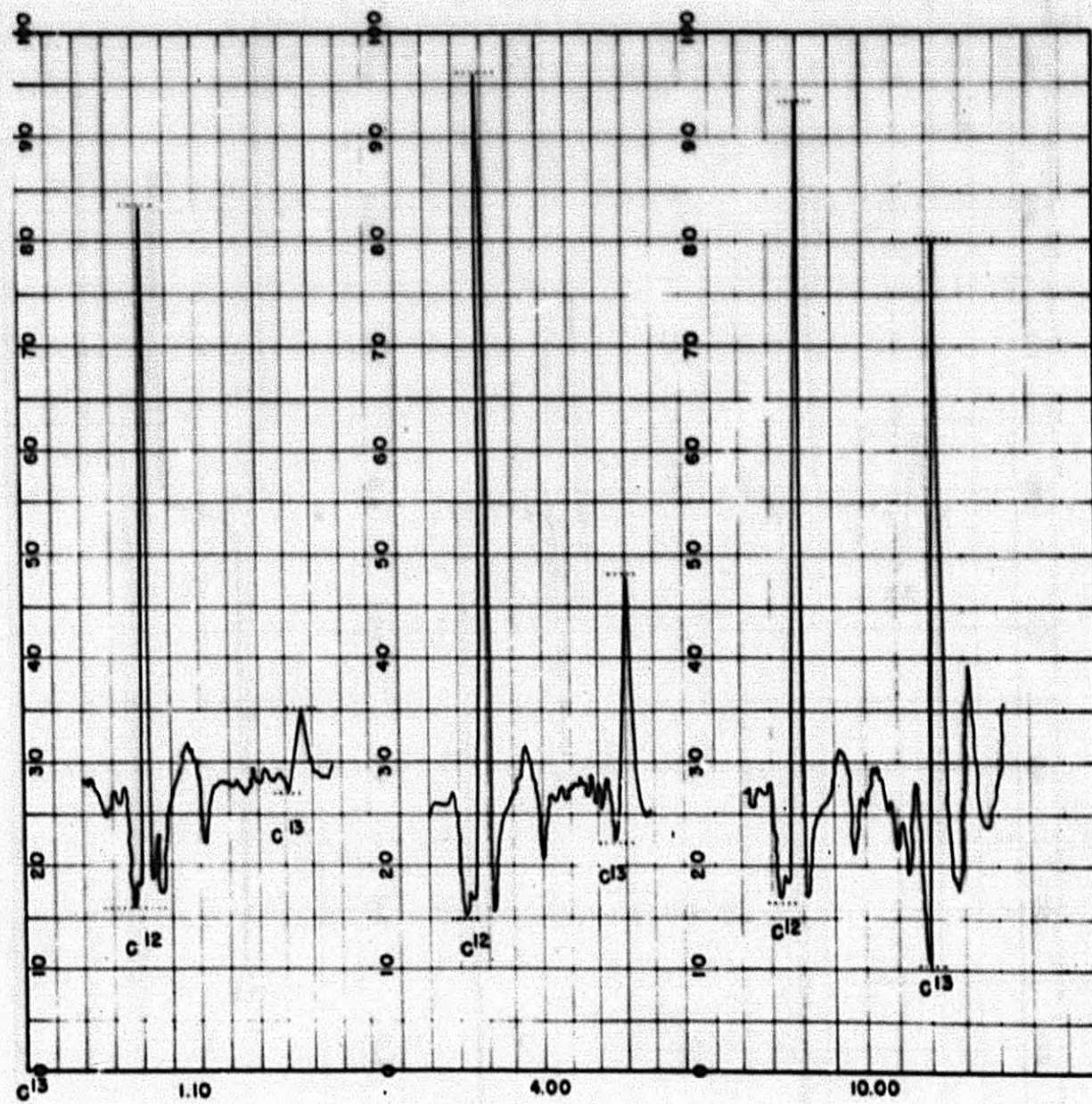


Fig. 2—Recorder traces for cyanogen chloride (100 μ pressure).

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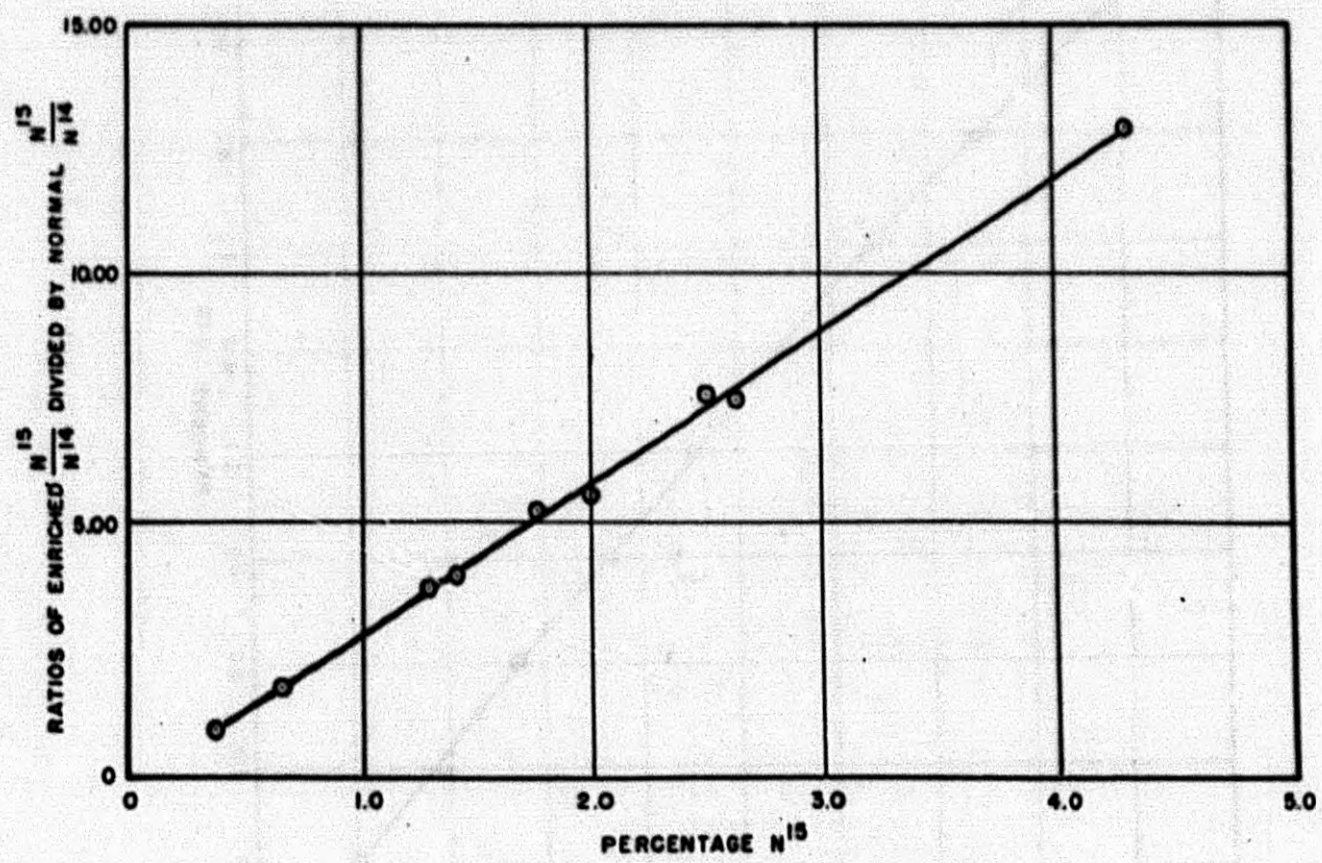


Fig. 3—Standardization curve for N<sup>15</sup> analysis.

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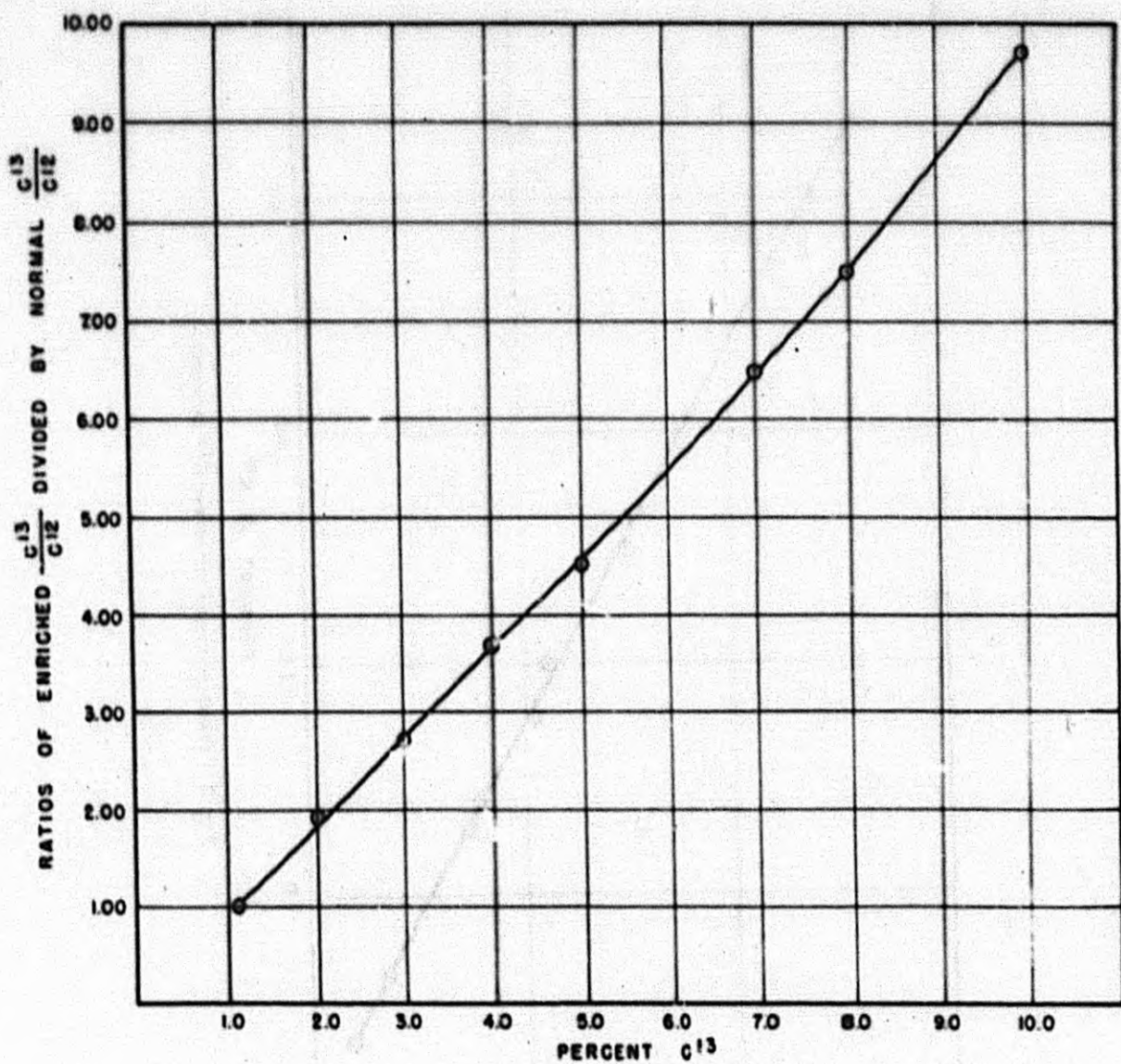


Fig. 4--Standardization curve for  $C^{13}$  analysis.

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Paper 5a

MATERIALS LABORATORY ACTIVITIES: INTRODUCTION

By P. J. Hagelston

The Materials Laboratory of the Isotope Research and Production Division is engaged in research, development, investigational and developmental service, and consultant work.

Principal fields covered are ceramics, plastics, metals, and certain combinations thereof. The projects cover: (1) independent research and development, such as the work being done on zirconium, hafnium, and other metal reductions, and studies of unconventional ceramics; (2) developmental problems and studies in cooperation with and/or as a service to other divisions (in this category there have been such developmental jobs as corrosion studies, welding and plating of metals, hydride preparations, graphite purification, and fabrication of special ceramics and metal parts); and (3) the provision of services and consultation as a result of the availability of the particular skills and equipment available in the laboratory.

There are light people in the laboratory, including myself. These people are divided into two general groups. In a department of this size though, as is usual, there is very close coordination between the two groups and most of the time the lines are very broad.

In one of the groups which we call the "Ceramics Group," dealing with ceramics, graphite purification, etc., we have Jesse Griffin and Fred Boody. Ralph Hutchison heads this group.

In addition, during the summer, we have the services of Dr. McVay from the University of Alabama on the Research Participant Program.

The other group, which mainly concerns itself with metals, is composed of Roy Roseberry, Stan Fulkerson, and Frank Knox. This group is headed by Frank Lambert.

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Paper 5b

MATERIALS LABORATORY ACTIVITIES: CERAMICS AND GRAPHITE

By R. O. Hutchison

Our ceramics program differs from the regular research program in that we have two primary functions instead of one. The first function is the development of ceramics for use within our own division, in other divisions in ORNL, Y-12, and X-10, and at K-25. The second function, which is both research and development, embraces longer ranged problems in which the AEC is particularly interested.

The general types of work carried on consist of development work on electrical insulators, cast ceramic heaters, ceramic reactors for high-temperature chemistry, ceramic crucibles for melting such materials as uranium, zirconium, hafnium, and lithium, and ceramic crucibles for holding some of these special queer AEC-sponsored alloys, such as Bi-Zr, Li-U, etc.

The laboratory is also used to furnish information on properties of ceramics which might be helpful in developing techniques of fabricating ceramics and to advise on source of supply when possible. A certain amount of experimental electroplating is carried out. Some examples of this are a smooth copper plate on graphite, rhenium plating of tantalum, and, at present, an investigation of the silver plating of uranium for the MTA program. The latter includes shortening the present process and experimenting with an entirely different process.

A carbon tube furnace is maintained in the laboratory for the purpose of purifying carbon and graphite parts. This purifying process consists of chlorinating the graphite and carbon parts, crucibles, etc. in an atmosphere of approximately 85 per cent chlorine and 15 per cent carbon monoxide at a temperature of about 1800°C. The average impurity of new extremely pure graphite ranges from 1000 to 2000 ppm; after processing these impurities range from 25 ppm downward. This furnace will handle pieces up to 5.5 in. in diameter and 27 in. long. Used calutron parts, encrusted with all types of contaminants, are purified to the same degree. Since the furnace was constructed, several thousand pieces have been purified in this manner, and because of the constant use of the furnace we are adding a new one that will handle pieces approximately 16 in. cube.

I would like to give a few specific examples of the type of problems we have handled, starting with ceramic heaters.

As the calutron begins to process higher and higher temperature compounds, particularly the rare earths, the calrod heaters generally used had to be abandoned for high-temperature heaters (see Paper 2). A research



program was carried out in close cooperation with the production personnel, and it has resulted in producing a heater that will operate satisfactorily for long periods of time above 1000°C.

The first approach to this problem consisted in embedding molybdenum elements in various ceramic materials. The body giving the best results was aluminum oxide with a few per cent glaze binder. However, the heater proved unsatisfactory owing to frequent burn-outs, and this led to the development of a lavite heater, meaning silicate, with graphite elements. This heater is in use today.

In working with a number of these unconventional ceramics, it is necessary to first synthesize some of these compounds. This entails high-temperature-chemistry techniques usually involving the use of purified graphite reactors in vacuum or controlled atmospheres. Zirconium nitride, zirconium carbide, thorium sulfide, boron nitride, and tantalum carbide are a few of the materials that have been synthesized in this manner. Ceramets, or ceramels, is a mixture of conducting and insulating materials, as the mixture of a metal in an oxide, the functions of the metal powder being twofold. In some cases metal acts as a producer of free electrons and in others it acts as a reducing agent for the oxide.

Semiconductors have been fabricated from magnesium oxide, carbon and zirconium oxide, zirconium oxide, iron oxide, and copper powder; and mixtures of thoria and molybdenum powder have been prepared by cold-pressing with a binder, usually fired about 1600°C in a vacuum. These pieces, which were in the shape of washers, were used in experiments on low-temperature-ignition filaments (see Paper 6).

The fabrication of pure oxide parts, zirconia, magnesia, and aluminum oxide, is usually done in the laboratory in one of three ways. The first method is by slip casting, which consists in preparing a fluid mixture of the material, about the consistency of cream, pouring it into plaster molds, and adding material until a sufficient wall thickness is reached. The material is then taken out, carefully dried, and fired to the desired temperatures. The second method is cold-pressing, which would also be called "tamping," and consists in pressing materials at room temperature with an organic binder. I might add here that a good part of our plastic work is used as binders for these ceramics. The material is then fired in the usual manner. A great deal of this type of work has been done on magnesium oxide.

Another example of cold-pressing and firing to various temperatures is our recent work on calcium fluoride as a refractory. This material was developed as a reactor lining for the zirconium-metals production and as a reactor lining for the fluorination of zirconium oxide. It was found that by sintering extremely pure calcium fluoride at approximately 1000°C, pre-grinding to a specified mesh, and re-firing to about 1200°C, a good piece of ware was obtained.

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The third type of fabrication is hot-pressing. This consists in packing the material in a graphite die and surrounding the die with thermatomic carbon, which in turn is surrounded by a quartz container. Outside of that is placed the induction coil for induction heating. The material is then pressed in graphite, approximately 1000 to 1200 psi, and raised to 1800 to 2000°C. While at this temperature the pressure is maintained for a few minutes after which the piece is pressed out into a container of graphite or carbon powder and allowed to cool.

Using these various methods of research and development, the search goes on for material to hold uranium, zirconium, hafnium, with varying degrees of success.

Several divisions at Y-12 have taken advantage of these services, and it is our hope that we can make the services available to every division in ORNL that is interested.

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Paper 5c

MATERIALS LABORATORY ACTIVITIES: METALS

By F. J. Lambert

The metallurgy program we are working on can be applied to four phases: zirconium-metal reduction, hafnium-metal reduction, lithium and uranium alloying, and investigations and development work.

ZIRCONIUM-METAL REDUCTION

The work in the reduction of zirconium fluoride has been continuing for seven months, during which time techniques were developed for producing a ductile zirconium. The present method being used to produce ductile zirconium can best be illustrated by a sketch of the reactor vessel, Fig. 1.

In the reactor vessel we have a steel shell, which has a tube welded on each side, in which we tamp a calcium fluoride liner. A mixture of zirconium fluoride, iodine and calcium metal, is placed in the center, and a cover of calcium fluoride is placed on top. Through the tube we push a tantalum rod which has high resistance in the center so that when we pass a current through this rod it will get hot and set the charge off. This rod has some D-29 gaskets placed on it and a Kerotest fitting tightened on to make it vacuum tight.

This bomb is then closed with an iron lid, and a copper gasket is seated between the lid and the bomb itself in order to produce a good joint. We evacuate this bomb three times and fill it with argon three times, finally ending up with an atmosphere of argon. We are then ready for firing.

Concentration of effort is currently being placed on the elimination of impurities which have been definitely established as detrimental to the corrosion resistance of zirconium metal. The ones that are now above the tolerable limits are aluminum and magnesium. Nitrogen, which is also bad, has previously been running about 500 ppm, but it is now down to about 70 ppm. The chief source of the nitrogen had been the calcium metal used. Fresh batches of calcium metal low in nitrogen are now being used.

The aluminum appears to be coming from the calcium fluoride liner, and tests are now in progress to lower the aluminum content in the calcium fluoride. The calcium metal appears to be the greatest carrier of magnesium and only calcium metal of a low magnesium content will be used in future runs. When the aluminum and magnesium impurities are lowered below this tolerable limit, we should have a corrosion-resistant zirconium.

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QUESTION: Does your argon have any nitrogen in it?

ANSWER: Yes, it has nitrogen in it, but, as I said, the tolerable limit of nitrogen in the metal is 100 ppm, and we are running about 70 ppm.

QUESTION: Is that the tolerance for corrosion or cross section?

ANSWER: Corrosion.

#### HAFNIUM-METAL REDUCTION

The reduction of hafnium fluoride is being carried out in very much the same manner as that of zirconium. We use the hafnium fluoride instead of the zirconium fluoride. The apparent higher melting point of hafnium makes it difficult for the metal to consolidate into a button. Out of several reductions we have produced one button; the other reductions have produced pieces of metal. We are continuing to experiment by varying the quantities of the charge material. When hafnium metal of sufficient quantity is produced, a research program designed to study the physical properties of hafnium will be carried out in cooperation with Dr. Andersen and Mr. Manly of the X-10 Metallurgy Division.

#### LITHIUM AND URANIUM ALLOYS

An attempt to make a 10 per cent Li-U alloy by firing a mixture of lithium fluoride-uranium fluoride, iodine, and calcium in a magnesium-lined bomb appeared to be very promising, except for the fact that the lithium soaked through the magnesium oxide liner and it was impossible to separate. A molybdenum cup was used in place of the magnesium oxide liner in an attempt to overcome the problem of soaking. Instead of a 10 per cent lithium alloy, a 3 per cent lithium alloy was attempted. By reducing the lithium it was hoped that a button would result which would not oxidize as rapidly as the 10 per cent lithium alloy. The button formed contained only 60 ppm; the remaining lithium was evidently tied up with the slag. A third attempt to make a 10 per cent U-Li alloy by reducing uranium tetrafluoride in molten lithium also failed because of segregation of the uranium (Fig. 2).

#### INVESTIGATION AND DEVELOPMENT

Under this heading fall those requests that require examinations of metal-equipment failures and investigations and development of new methods for doing specific jobs.

Recently cold-welding of aluminum and copper was developed industrially, and Mr. Case of Y-12 foresaw possible application of this process in the fabrication of aluminum liners. The advantage gained would be the reduction of warpage which occurs in arc-welding these liners. Mr. Case had the opportunity to witness the cold-welding operation and submitted samples for investigation.

The welding operation consists in stamping two pieces of aluminum sheet between dies of identical shape. The sheets are left with an indentation which extends about halfway through. The samples I had, after cold-welding, looked something like that shown in Fig. 3.

The sheets were about 0.068 thick. This was pressed down about half-way through, which was about 0.034. The weld zone is about half the length of what you might call the "welding surface" of the die. All these welds were 1 in. long.

For those who might have use for this type of weld, the Y-12 shops will be in a position to do fabrication by this method in the very near future.

At the request of Mr. Graham of the Reactor Development Group of X-10, a study was made to determine the feasibility of making pressure welds on pipe in the laboratory. This method of welding was being considered in replacing the pump in the homogenous reactor when it failed. Of course this would have to be done by remote control. The conditions needed to produce welds of good quality were found to be too exacting, and consequently it was decided to shelve this method in lieu of another which will probably be inert arc-welding.

Work is being carried on in cooperation with Dr. Livingston of Y-12 and C. D. Smith of X-10 on the development of the fabrication method to be used for the linear accelerator.

Plating procedures for uranium are being studied at the Materials Laboratory and at Y-12 plating shops. Samples are being furnished Mr. Smith for bonding tests to other metals. Heater plates for use in hot pressure bonding are being fabricated for this project. Castings used for bonding experiments will also be made.

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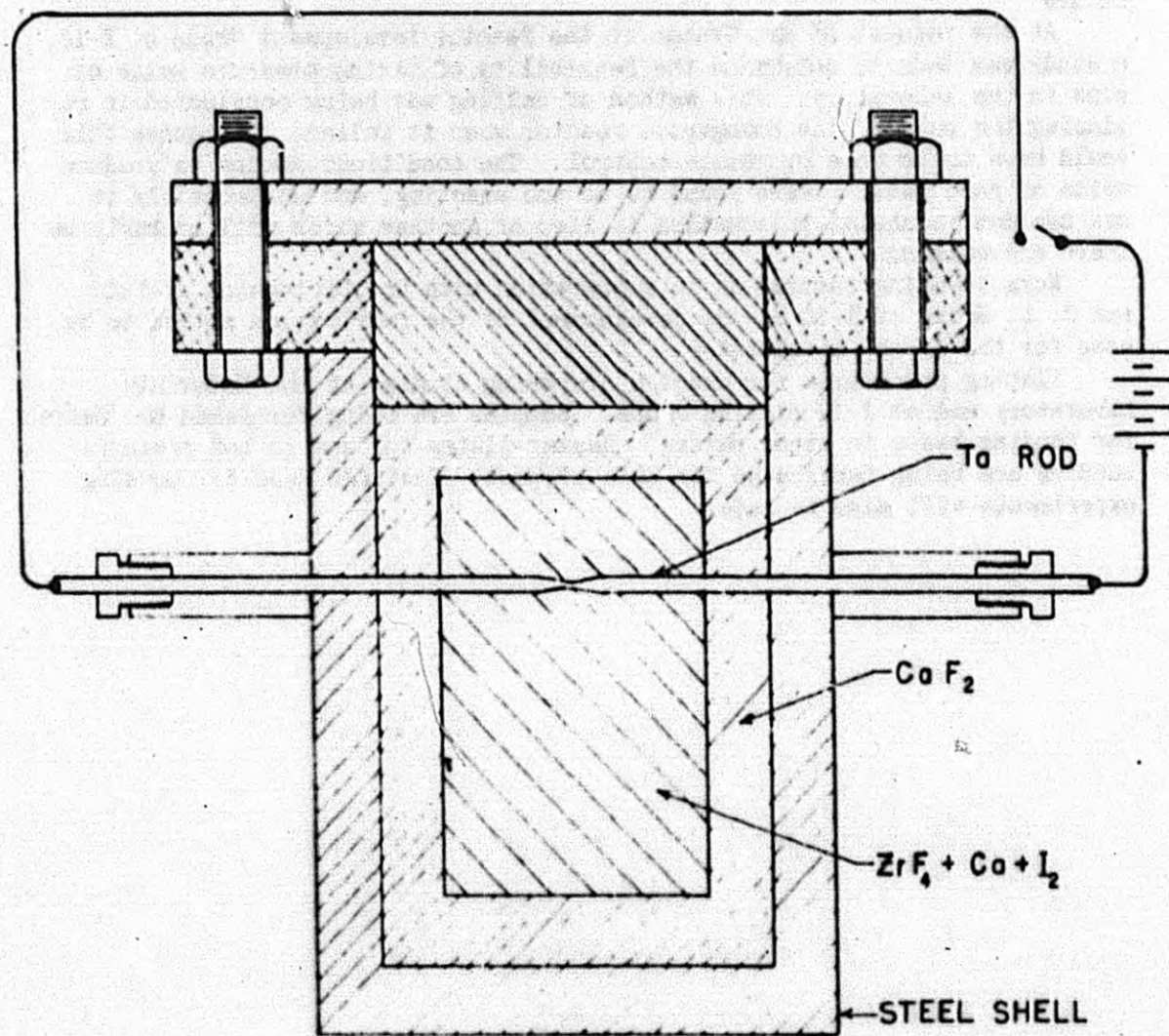


Fig. 1—Reactor vessel.

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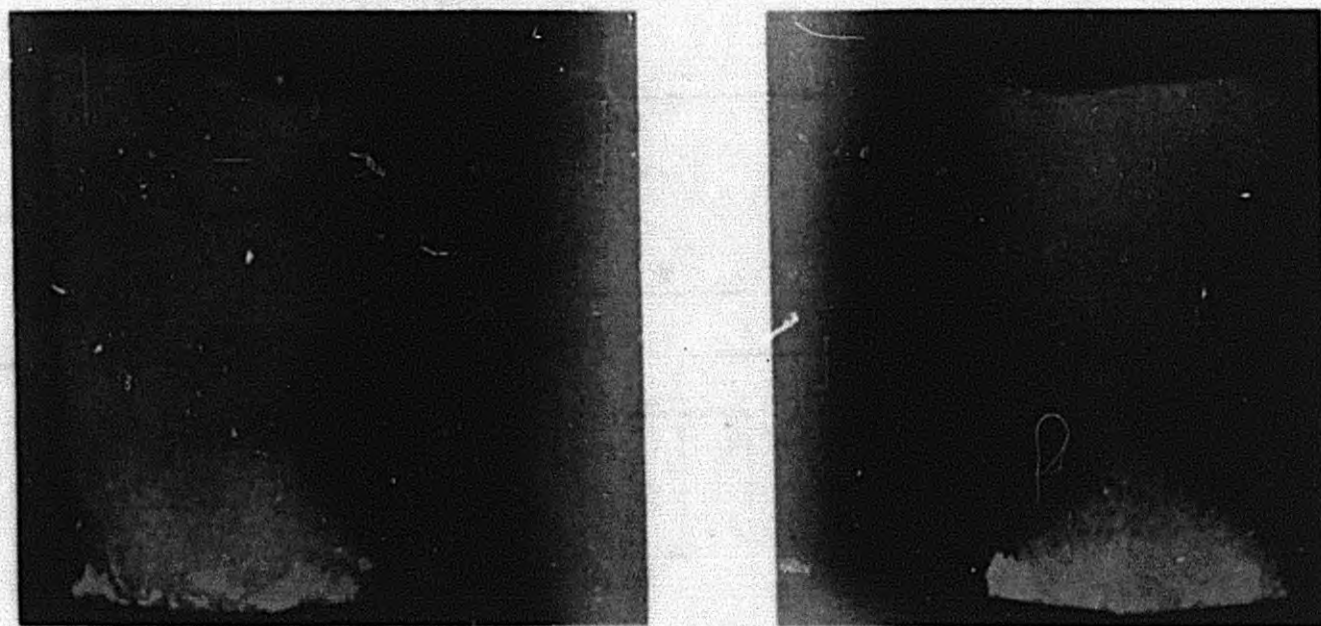


Fig. 2—X-ray picture showing segregation of uranium in the ingot cast of U-Li alloy.

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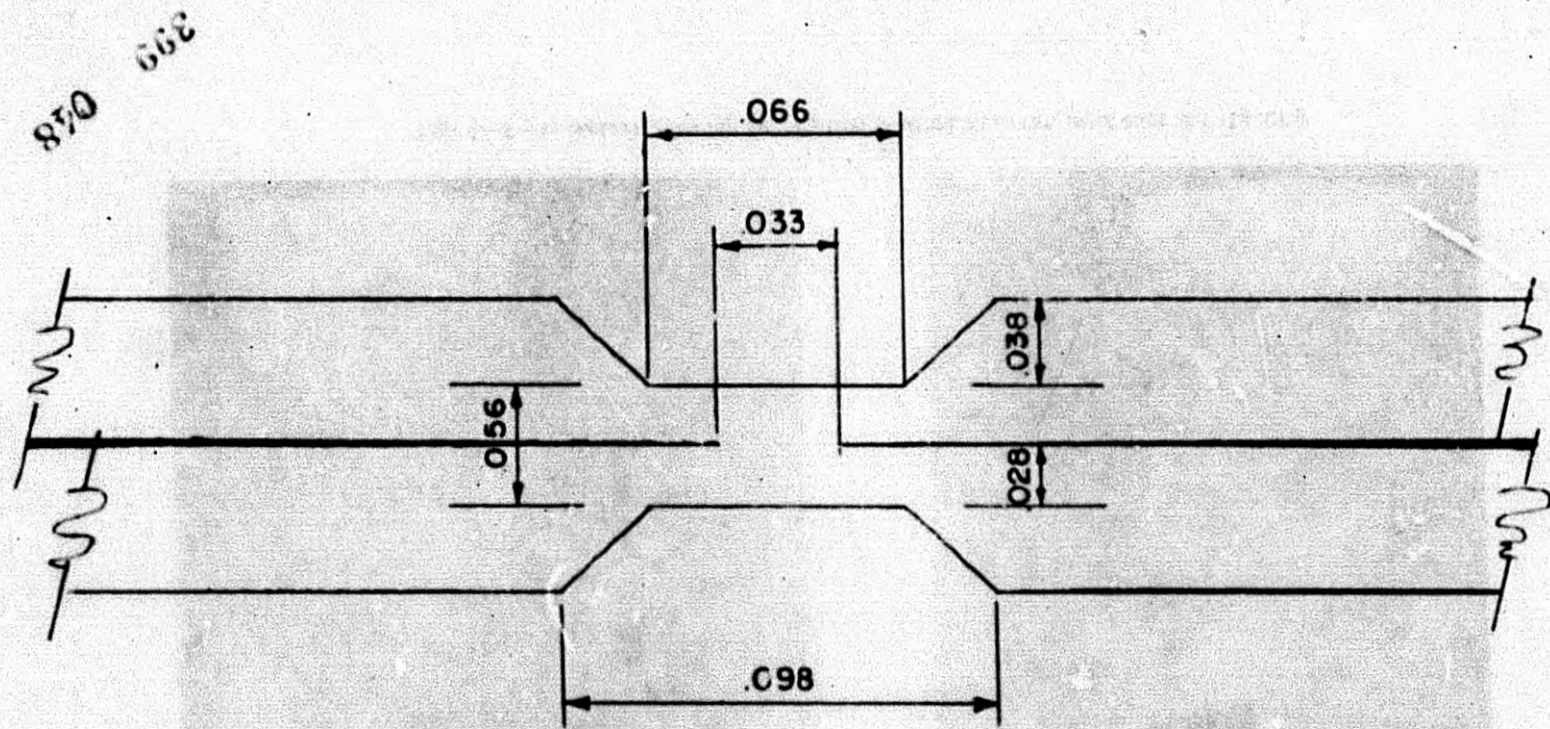


Fig. 3—Cold-welded sample.





Fig. 4—Microphotograph of cold-welded aluminum.

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Paper 6

SOURCE, FILAMENT, AND VACUUM RESEARCH

By C. R. Baldock

The principal objective of the source and filament study is to carry out a program of research in the collision-process phenomenon related to, or encountered in, calutron production of the isotopes. Our chief piece of equipment for the ion-source studies is a 60 deg neotype specially constructed mass spectrometer. There are several features of this equipment which I think would be of interest.

It has an 8 in. radius of curvature for the ions. The pole gap is 1 in., and we are able to easily get up to 11,000 gauss in the gap. Perhaps one of the most novel features of the magnet is the fact that it is mounted from above the vacuum system, which allows a great deal of accessibility and ease of servicing. Another novel feature is that we are driving the magnetic field directly from an amplidyne generator rather than from the usual type of electronics.

This instrument, we find, is most suitable for our particular type of problems because it enables us to subject the ion source to a wide variety of conditions, particularly those varying the magnetic field without disturbing the magnetic field used for analyzing and sorting out the fragments. Mr. John Sites is in charge of this particular laboratory.

During the past few months we have been investigating a number of charge compound cracking fractions. The reason we have chosen to look into these cracking patterns is that we have believed that by so doing we could predict the operating characteristics of these compounds when used as charge materials in the calutron. Our work in the past three months seems to bear out this prediction very well. I will cite only three examples.

In the case of tungsten hexachloride, in spite of our long operating experience, we have never been able to get very satisfactory ion yield. On studying the cracking patterns under single-collision-process conditions, we have found, in addition to a very high percentage of all of the possible chlorides, oxychlorides, and oxides as well, which came as no great surprise to the charge-preparation chemists, that the electrons which produced the single plus metal ion, in the case of tungsten, must have a velocity corresponding to a minimum of 20 volts. This is rather significant when you realize that in the calutron arc the average velocity of the electrons probably does not exceed 2.5 to 4.5 volts and that you only have available, perhaps, the high-energy tail of some possible secondaries to do the ionizing when you are getting the single plus metal ions.

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Just to check something that was known to be an extremely fine positive-ion producer, thallic chloride was checked, and it was found, as expected, that about 98 per cent of all ions produced were the single plus metal ions and that electrons with velocities even less than 10 volts could easily produce the desired ion.

We have recently checked rhenium tetrachloride, which was proposed as a possible material for obtaining rhenium isotopes. The mass spectrometer analysis showed that all one was doing when he heated the compound was to drive off the rhenium and that none of the metal ions appeared at all. On that basis it was predicted that it was not possible to serve as a calutron charge material.

Subsequently, a test was made running an arc in a magnetic field, and spectroscopic means were used to check it. It was proven conclusively that all one did was to drive off the chlorine, and we were very glad to be able to return all the rhenium for further work.

There are a number of fields that we want to move into, such as improving ion-source deficiency, but I will not discuss our ideas about that. We would also like to eventually breach the gap that exists between the high precision of the mass spectrometer and the high ion currents that one obtains in the calutron.

In the filament laboratory we have as our prime objective, of course, the extension of the life of filaments which are used to produce the primary electrons needed to run the arcs. We found there a rather complex mixture of filament evaporation, chemical attacks, and erosion on the ion bombardment.

We have a well-equipped laboratory operated by Mr. Lees. In addition to the vacuum system and magnet we have a-c and d-c supplies and all the necessary equipment for high-precision current measurement, vacuum or pressure measurement, and measurement of flow rates, optical pyrometers and radiation instruments for temperature measurements, etc.

Our work has proceeded in four fields and I will say only in passing that attempts at producing activation of filaments under positive-ion bombardment have yielded some success, but it is extremely difficult in that you always have to proceed under the assumption that you have a mechanism for constantly replacing the material since any preactivated surfaces are immediately cleaned up under positive-ion bombardment and the electron production from such surfaces is extremely erratic.

Our most hopeful program to obtain a filament to withstand the high currents in ion bombardments will probably come out of our studies of some of the semiconductor materials. We are grateful to the Materials Laboratory for the development work they have done in fabricating some test pieces, and we hope they will soon have the hydraulic presses needed to give us the exact types needed for detailed testing.

Basic studies of a large number of properties are carried on continuously because we can predict one thing with certainty, that is, every time you go to a new element you have an entirely different type of filament problem usually of a nature you never would have predicted.

In regard to our vacuum research, I would like to say that we have been extremely fortunate in having Dr. Normand directing this work since the early part of the project. Perhaps you do not know this but it was he and

Dr. Robert Thornton who did all the early research and development work which made it possible to go from the laboratory type of a few liters per second pump up to the many thousand liters per second pump which has operated so successfully ever since the beginning of the electromagnetic project. His work comprises the development through experimental determination and theoretical analyses, techniques and information regarding equipment, materials, and design principles and techniques, which are applicable to the construction and operation of high-vacuum equipment and associated processes.

I will mention only three recent phases of his work. Incidentally, he has a well-equipped laboratory with a large number of pumps, parts, and measuring devices. Mr. William Rathkamp is in charge of this laboratory. Tests have recently been made on a large series of vacuum greases for high-vacuum work, and it has been found that their relative merits can be shown both by weight and pump-down curves; he is in the process of completing a report on that study.

He has also investigated the effects of various gasket materials, and he finds that the initial very marked outgassing or evaporation is a transient rather than a permanent phenomenon and once the materials are removed from the vacuum system they quickly return to their previous bad behavior. Finally, I would say that he has done extensive work with regard to gauge calibration, and it is his finding that for general purpose use it is hardly worth doing much precise calibrating in the ion gauges because the variations with conditions of use are large and most unpredictable.

Our group is very happy to have with us this summer Dr. Carr from Auburn. His vast experience with chlorine and chlorine-containing compounds has been invaluable to us in evaluating some of these materials.

**END**

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