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WHY SEPARATE STABLE ISOTOPES

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2

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WHY SEPARATE STABLE ISOTOPES?

INTRODUCTION

One of the most revolutionary discoveries in science was the discovery that all the atoms of a particular element may not necessarily all have the same mass. These atoms, or nuclides, which have practically identical chemical properties but differ in weight have come to be known as isotopes and their discovery has revolutionized the study of the physical sciences. Isotopes are divided into two appropriate classes: stable and radioactive. The advent of the release of atomic energy in 1945 directed the attention of the layman and the scientist to the fact that they were on the threshold of a new field and both felt that it should be explored. The scientists were quick to realize the tremendous possibilities of the equipment and personnel of the existing laboratories which had contributed to the first large scale atomic energy efforts. The existing atomic energy piles at Oak Ridge and Hanford made it possible to produce unheard of quantities of radioactive isotopes. The production type mass spectrographs located at Oak Ridge provided the basic equipment for the production of large quantities of stable isotopes.

Radioactive isotopes can be identified by the radioactive emanations coming from them. The detection of radioactivity is by now a highly developed technique, so radioactive isotopes are extremely convenient tools of research, particularly in the applied fields of science such as medicine, metallurgy, and others.

Before radioactive isotopes could be used many years of research were necessary on the basic principles of radioactivity. From the time of the discovery of natural radioactivity by Becquerel in 1896, stemming mainly from Roentgen's discovery of X-rays in 1895, to the present year 1950 many fundamental experiments were done. Today, at an ever increasing rate, we are all reaping the fruits of such fundamental scientific work. Had it not been for such a background of factual scientific information, the present level of radioisotope application would be much lower. The history of radioactive isotopes is typical in science, a similar history will probably be experienced with stable isotopes.

Sir J. J. Thomson reported in 1913--about 20 years after the discovery of radioactivity among certain elements--the existence of different atomic masses among the atoms of neon. By 1924, through the work of Thomson, Dempster, and Aston, 25 elements had been found to have different stable atomic masses or stable isotopes, the differences being due to the differences in nuclear structure of the atoms of a given element. By 1935 between 50 and 60 elements had been identified as having natural stable isotopes and another 10 or so as having natural radioactive isotopes.

Although heavy hydrogen, that is the hydrogen isotope of mass 2, had been separated in appreciable quantities in the heavy water fraction of electrolysis by Urey, no other isotopes had been separated in appreciable quantities at this time. Then followed the successful isolation of uranium-235 both by the electromagnetic method and by the gaseous diffusion method in Oak Ridge. The atomic bomb application, of course, was responsible

for this accomplishment; the practical application of an isotope naturally makes it of more immediate value than if it is needed for basic studies, the practical value of which cannot be predicted.

BACKGROUND OF THE STABLE ISOTOPE PROGRAM

In the fall months of 1945 a relaxation of wartime pressure made it possible to examine the electromagnetic isotope separation equipment to see how it could be adapted to the separation of isotopes of elements other than uranium. In this way improvements might be made in the calutron, as the electromagnetic isotope separating equipment is called, in its main objective of processing uranium; but, at the same time, enriched isotopes could be collected which might have value in fundamental or applied scientific research. This program is operated now almost entirely for the isotopes produced and the scientific findings which they make possible.

In passing it may be interesting to know what some of the factors are which have to be considered in processing the isotopes of different elements in the calutron. Here are some of those factors: The selection and synthesis of an anhydrous charge material which will supply an abundance of singly charged positive ions and from which sufficient vapor can be obtained at the temperature at which the standard calutron source can be operated, or if such a charge material is not found, to alter the calutron source to meet extreme temperature requirements of the charge compound or element which is available; chemically purifying the charge material; the production in the calutron source of sufficient positive ions of the

isotopes being separated; the development of sources and collectors of materials which will withstand temperature and corrosion conditions experienced with the element being processed; determination of the best geometry of source, accelerating electrodes, ion path, and collector pockets; the maintaining of adequate vacuum and adequate cooling of heated calutron parts; determination of the best accelerating voltage, magnetic field and radius of ion travel; finding the optimum operating control conditions to achieve maximum process efficiency, maximum production, and maximum purity of the desired isotope; chemically purifying the collected isotopes--this requires working out the technique of how to chemically purify small quantities of precious product material; and analyzing chemically and isotopically the product isotopes.

It goes almost without saying that even the foregoing calutron process problems unearth worthwhile physics, chemistry and engineering data. Of course, the main objective is the research findings obtained with these separated isotopes, most of them stable but occasionally a natural radioactive nuclide appearing at random among stable nuclides, at least at random according to our present knowledge; these are K^{40} , Rb^{87} , Sn^{124} , Sm^{147} , Lu^{176} , and Re^{187} .

ISOTOPES PROCESSED AND SHIPMENTS MADE

What is the story on this inventory of separated isotopes collected during the last 4-5 years?

Up to June 30, 1950, 550 isotope samples had been completely processed--these comprise 173 isotopes of 43 elements. Eight hundred and twenty

individual shipments had been made to 9 Atomic Energy Commission sites and to 34 university and industrial laboratories. Three hundred and fifty isotopic samples are on hand to fill future requests and 80-85 requests are on file which can be satisfied only when additional collections can be done and when some collections previously done can be chemically purified and prepared for shipment.

Table I

Stable Isotope Summary
November 1, 1945 - June 30, 1950

Isotope Samples Completely Processed	550
Individual Elements Involved	43
Individual Isotopes Involved	173
Shipments Completed	820
AEC Laboratories Receiving Shipments	9
University and Industrial Laboratories Receiving Shipments	34
Unfilled Requests for Isotopes on Hand	80-85

USES OF STABLE ISOTOPES

What are the recipients doing with these separated isotopes? Let's look at some of the 140 articles in the technical journals, and papers which have been presented at scientific meetings, in which isotopes were employed; also at some of the proposed experiments which justified the allocation of these isotopes.

First, let me quote from the AEC report of January, 1950 titled, "Atomic Energy and the Physical Sciences" in which neutron cross-sections are discussed.

"A variety of neutron energies is needed in the measurement of neutron cross-sections; that is, the measurement of the probabilities

that neutrons of any specific energy moving into any particular material will pass through unhindered; or be deviated from their course, or be bounced about, or be absorbed--and if the last, which one of the many possible resulting nuclear reactions will ensue.

"To cover this field of investigation completely would require the measurement and tabulation of these various cross-sections for every isotope of every element at every possible neutron velocity from the slowest to the fastest. The investigations actually being carried out concern themselves with the isotopes and the neutron velocities of most immediate practical interest in atomic energy development. The program again further divides itself in accordance with the different facilities available at the different laboratories. Uranium reactors are a tremendous source of slow neutrons, and programs for measuring slow neutron cross-sections are under way at Oak Ridge and at Argonne. Measurements involving fast neutrons, on the other hand, are carried on where there are powerful particle accelerators, as at Berkeley and Columbia University.

"Even when reduced to its essentials, the program for the determination of neutron cross-sections engages a substantial part of the available manpower at every large AEC laboratory and accounts for a sizeable share of the Nation's budget for atomic energy research and development. Typically in the past, the kind of background research involved here--the painstaking collection and tabulation of related measurements that are finally printed in the handbooks of the engineers--was the work of many decades. Today, it is being done systematically and rapidly. It is

speeded by: (1) the development of better experimental techniques, (2) the advancement of nuclear theory, and (3) the promotion of basic research."

One of the most important and widespread applications of stable isotopes is in the determination of neutron cross-sections. The neutron cross-sections of the elements and their individual isotopes are becoming more and more important in those studies and in applications where neutron absorption or neutron transmission is a factor. In economizing on neutrons in a pile, as an illustration, materials are needed which permit the ready passage of neutrons at thermal energies, whereas for shielding personnel from harmful neutron irradiation effects materials are needed which absorb neutrons to a high degree. Every element has a neutron cross-section value which is a weighted average of the individual cross-sections of its isotopes.

Take cadmium as an example. This element is an excellent absorber of thermal neutrons as is evidenced by its being used as control rods in the pile. It has 8 stable isotopes. All are very low absorbers of neutrons except Cd^{113} which has a cross-section of 24,000 barns. Twelve per cent of all cadmium atoms are Cd^{113} , the other 88 per cent are made up of 7 isotopes all of which have neutron cross-sections of 1-2 barns or less--a very fascinating situation. As a result, cadmium as the element has a cross-section of 3000 barns whereas if Cd^{113} alone were used a cross-section of 24,000 barns would be available. Conversely, if all other cadmium isotopes were used--omitting Cd^{113} --the cross-section would be only a few barns. Hence, we can have cadmium of extremely high cross-section or we

can have cadmium of extremely low cross-section. Separations of isotopes in the calutron make this possible. Such cases as this will become more and more valuable in the construction of neutron reactors. We will not only use alloys of elements to gain the proper chemical, physical and structural properties, but we will also use alloys of isotopes to gain in addition the needed nuclear properties, such as high or low cross-sections.

A second application of enriched stable isotopes has been in establishing the masses of radioactive isotopes artificially produced in the pile, the cyclotron, or other particle accelerating machines by neutrons, protons, or alpha particles. For example, ordinary copper is composed of two stable isotopes, Cu^{63} and Cu^{65} . If natural copper is bombarded with neutrons so as to favor an (n,p) reaction the product will be an isotope of nickel, either Ni^{63} or Ni^{65} . It had been observed that a nickel isotope of 2.5 hour half-life was formed but there was no positive way to determine if it was Ni^{63} from the Cu^{63} isotope or Ni^{65} from the Cu^{65} isotope. From enriched Cu^{65} Swartout and Boyd of ORNL identified Ni^{65} as the 2.5 hour isotope. This was the first application of separated isotopes for positive mass confirmation of artificially produced radioisotopes. Many such uses of enriched stable isotopes have since been reported, particularly by Pool and his students at Ohio State University.

A third contribution of separated natural isotopes has been in studying the properties of naturally radioactive K^{40} and Sm^{147} , which occur with the stable isotopes of those elements. Weaver of the Stable Isotopes Division positively assigned the alpha activity of samarium to Sm^{147} after

the separated isotopes of samarium became available. (At various times Dempster had believed this activity was due to Sm^{147} , Sm^{148} , or Sm^{152}). A number of workers at ORNL, Argonne, Brookhaven, and Columbia used enriched K^{40} to establish more accurately the half-life of K^{40} and to study the beta disintegration spectrum of K^{40} . This was possible because the calutron increased the K^{40} atoms from 1 in 10,000 to over 300 in 10,000. The contribution of K^{40} in this work has been enthusiastically voiced by Wu of Columbia, who is continuing her work on the spectrum of K^{40} , and by Hughes of Argonne and Brookhaven.

Speaking of the beta spectrum of K^{40} --calutron separations of artificial mixtures of isotopes have also been valuable in studying other beta spectra, especially that of Be^{10} . The element beryllium has only one natural atomic species, or nuclide, Be^9 . But if Be^9 is irradiated in the pile some beta radioactive Be^{10} will be formed. Hughes of Argonne had some Be^9 prepared in this way in a Hanford pile but there was only one atom of radioactive Be^{10} in every 500,000 atoms of stable Be^9 ; such a dilute concentration of the radioactive nuclide that he could not get an intense concentration of radiation. Consequently, the calutron physicists enriched the radioactive Be^{10} atoms in Be^9 from 1 in 500,000 to 1 in 1,500, thereby making it possible to obtain accurate fundamental information concerning the energy distribution of the beta rays from Be^{10} .

A practical industrial application of a stable sulfur isotope has been in the vulcanization of rubber. The mechanism of sulfur in this process is only partially understood. By using one of the less abundant

sulfur isotopes in an enriched form and measuring the isotopic ratio of the sulfur isotopes by a mass spectrometer at various stages in the experiment the exchange of sulfur atoms and the mechanism of the reaction can be better understood. Furthermore, the measurement of the infrared absorption spectrum of sulfur containing enriched S^{34} is a valuable procedure for the research chemist in rubber technology.

A staff physician at one of the hospitals has been interested in studying the part potassium plays in body metabolism. (By the way, natural K^{40} has a half-life of about one hundred million years). His research has made use of enriched radioactive K^{40} to be followed as a tracer, and enriched stable K^{39} and K^{41} are also being traced with the mass spectrometer. Furthermore, there is an interest to learn if the long-term effects of potassium are any different between using natural potassium, in which nature has put an occasional radioactive atom of K^{40} , and potassium in which this radioactive isotope has been removed. This reminds me of the biologist who would like to develop a strain of animal in which the nitrogenous material is pure N^{15} rather than natural nitrogen in which the N^{15} is only 0.4 per cent, the other 99.6 per cent being N^{14} . These animals might be physically quite different, no one knows for sure.

A number of research laboratories are investigating the superconducting properties of various isotopes, like those of mercury and of tin. Certain unexplained inconsistencies have been found. According to laws of physical chemistry relating to superconductivity--a material being

considered superconducting at the low temperature where its resistance drops to zero--one would expect the transition temperature to superconducting to be lower the higher the atomic weight of the mercury or the tin. For tin isotopes this has been found to be the situation but with mercury isotopes the reverse has been observed. This seeming anomaly is under further study and may be important.

The 7 natural stable isotopes of mercury are interesting and apparently worth investigating in various ways. They were difficult to process in the calutron, mainly because of the relatively high vapor pressure of mercury at ordinary temperatures, so that natural mercury would not only keep the pressure in the vacuum chamber of the calutron too high for successful processing, but normal mercury would diffuse into the individual isotope collectors and undo the isotope separations accomplished in the magnetic field between source and collectors. Also, if the collectors were not cooled to a low temperature the isotopes could not be effectively retained in the collectors. Amalgamation or chemical combination in the collector aided retention but low temperatures were necessary to really be effective.

As you all know mercury arc lamps emit the characteristic spectrum of the element mercury. If the odd isotopes of mercury, Hg^{199} and Hg^{201} , which comprise 30 per cent of all the isotopes could be removed the spectrum would be greatly simplified. (The hyperfine structure of the odd mass mercury isotopes complicate the spectral picture). Furthermore, if one of the even isotopes were obtained almost 100 per cent pure, then

a single spectral line of a definite wavelength could be observed in the spectroscope.

Dr. Meggers of the Bureau of Standards has proposed that Hg^{198} be used as a standard wavelength since it can be prepared better than 98 per cent pure from Au^{198} in the pile. He has made mercury lamps using this Hg^{198} , and he proposes that the single spectral line be the standard primary wavelength from which all measurement be standardized instead of from the standard platinum-iridium meter bar--certainly a spectral line would be a much more permanent and scientific primary standard than the meter bar.

Physicists and calutron technicians have enriched natural Hg^{202} to over 98 per cent purity and have observed the pure spectrum from it. This pure spectral line of Hg^{202} could serve as a standard of measurement as well as Hg^{198} --the main justification for the choice between Hg^{198} and Hg^{202} resting on the availability of either isotope.

One can readily see the value of having such a primary standard of measurement in our laboratories, whether industrial or academic. From these clean spectra of Hg^{198} and Hg^{202} the wavelength of their lines can be measured to an accuracy of at least one part in a million million centimeters.

Enriched lithium isotopes have various possibilities. A useful unclassified application has been in photographic films used for detecting neutrons. The incorporation of enriched Li^6 in such emulsions is regularly done and these films are widely employed by nuclear physicists.

Enriched isotopes have many possibilities as starting materials in the artificial production of radioisotopes in the pile by neutrons or in the cyclotron by protons or alpha particles. A few milligrams of enriched stable isotope, an amount readily enriched in the calutron, will often produce a large number of millicuries of a radioisotope. As an example, enriched Fe^{58} irradiated in the pile produces 46-day Fe^{59} , a valuable radioisotope in medical research, whereas normal iron containing appreciable Fe^{54} produced intolerable amounts of 4-year Fe^{55} . Forty-six-day Fe^{59} can be safely used in human experiments but the 4-year Fe^{55} cannot.

Much more could be said about the use of enriched stable isotopes in fundamental physics research. In summary, however, these isotopes are used in many investigations of optical spectroscopy, particularly in hyperfine structure and in the determination of isotope shifts, investigations of nuclear energy levels, nuclear spins and of magnetic moments, and other properties involving the nucleus. Associated with such stable isotope research programs will result developments in instruments of research in electronics, microwave spectroscopy, optical spectrographs, methods of mass analysis, the design and use of magnets, nuclear research tools, and similar equipment.

Furthermore, much basic work is being done in chemistry associated with calutron charge choice and preparation, and in the chemical refinement of the precious isotope product. A large amount of research is being done and has contributed to the fundamental chemistry of many elements, notably gallium, hafnium, zirconium, rhenium, cerium, and other rare earth elements.

Metallurgy and ceramics have contributed measurably to the success of enriching isotopes and have in return received isotopic materials for research and development. Because of the objective of separating hafnium isotopes, for example, highly purified hafnium oxide was produced by the joint efforts of several groups of personnel, and as a result not only have zirconium and hafnium isotopes been obtained in very pure form but the metallurgists in the Materials Laboratory at Y-12 have produced appreciable amounts of pure zirconium and hafnium metal for research studies.

WHY SEPARATE STABLE ISOTOPES?

In one hand I hold a vial of almost pure isotope, Hg^{202} , and in the other hand probably the purest hafnium metal ever available to mankind. What the value of these products, and others like them, may be to our future national security, industry, or personal well-being no one will know until more research is done with such materials, only available because a stable isotope separation program is maintained by our government through the Atomic Energy Commission. These, then, to me are the most important reasons for separating stable isotopes: To make available for the extension of knowledge in the basic and applied sciences new materials never before available to mankind.

APPENDIX

The following is a summary of some unclassified uses of stable isotopes; this compilation was prepared by Boyd Weaver of the Isotope Chemistry Laboratory.

SOME UNCLASSIFIED USES OF ELECTROMAGNETICALLY CONCENTRATED ISOTOPES

<u>Uses as Supplied</u>	<u>Users</u>	<u>Isotopes Supplied</u>
Study of natural activities	University of Chicago Princeton University Massachusetts Institute of Technology University of Michigan University of Southern California	K 40 In 113, 115 Sn 116, 124 Zr 90, 96
Spectroscopy Isotope shift, hyperfine structure, spin, magnetic moment and discharge tube phenomena	University of Wisconsin University of Buffalo Harvard University Massachusetts Institute of Technology National Bureau of Standards University of Southern California University of Pittsburgh Westinghouse Research Laboratory Yale University	Li 6, Pb 204, 207 Ba 134, 135, 137 Cr 53, W 180, 183 Te 120, 122, 123, 124 Te 125, 126, 128, 130 Mo 95, 97, Mg 25 Ca 44, Ti 47, 49, Fe 57 Ge 73, Se 77, Zr 91 Si 29, Ni 61, Zn 67 Cd 111, 114, Hg 199, 201, 202 Cu 63
Infrared absorption spectrum	B. F. Goodrich Research Center	S 34
Microwave	Harvard University State University of Iowa	S 33
Mass Spectroscopy	University of Chicago University of Minnesota	Cl 37 K 40
Superconductivity	National Bureau of Standards Rutgers University Yale University	Hg(all), Sn(all), Tl 203, 205
Medical research	Presbyterian Hospital, Philadelphia	K 40

Uses as SuppliedUsers

18

Isotopes SuppliedUses Involving IrradiationStudy of pile-produced
activitiesUniversity of Illinois
Massachusetts Institute of Technology
University of Michigan
Northwestern University
Reed College
Vanderbilt UniversityCe 136,138
Sn (all)
Te 122
Li 6
Fe 54, Mo 96
Se 74
Cd 108,114Study of cyclotron phenomena
and activities producedCarnegie Institute of Technology
Johns Hopkins University
Indiana University
Massachusetts Institute of Technology
University of Michigan
Ohio State University
Princeton University
Purdue University
University of Rochester
Washington State College
University of Illinois
University of Notre Dame
Yale UniversityLi 6, Cu 63, Ag 107,109
K 41, Fe(all), Se 74,76,77
Pb 123, Tl 203,205
Pb 206,207,208, Sn(all)
Cr 50,52,54, Zn(all)
Hg(all), Mo(all)
Ni 54, Zr(all), Ti(all)
W 182,193,196, Re 185,187
Ba(all), Ce(all)
Cd(all), Ge 70,76Study of betatron phenomena
and activities producedUniversity of Chicago
University of Illinois
University of PennsylvaniaSr 84, Ca 40,42,48, Sb 121
W 184,186, Hg 196,201,202
204, Si 28, Te 130, Ti 46
Fe 54,58, Ni 58,64, Zn 54,
Zn 70, Ge 70,76, Se 74,80,82
Zr 90, Mo 92,95,100, Cd 106,
Cd 114,116, Li 7, Sn 119
Sn 120,122,124, Cr(all),
S 32,34, Pb(all), Mg(all),
B 11, K 39,41Study of Van de Graeff phenomena
and activities producedBartol Research Foundation
California Institute of Technology
University of Minnesota
Massachusetts Institute of Technology
Rice InstituteSi 28,29,30
Li 6,7
Mg 24,25,26

Medical Research

Harvard University

Cr 50

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