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NATIONAL USES AND NEEDS FOR SEPARATED STABLE ISOTOPES IN PHYSICS, CHEMISTRY, AND GEOSCIENCE RESEARCH

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Abstract

Present uses of separated stable isotopes in the fields of physics, chemistry, and the geosciences have been surveyed to identify current supply problems and to determine future needs. Demand for separated isotopes remains strong, with 220 different nuclides having been used in the past three years. The largest needs, in terms of both quantity and variety of isotopes, are found in nuclear physics research. Current problems include a lack of availability of many nuclides, unsatisfactory enrichment of rare species, and prohibitively high costs for certain important isotopes. It is expected that demands for separated isotopes will remain roughly at present levels. although there will be a shift toward more requests for highly enriched rare isotopes. Significantly greater use will be made of neutron-rich nuclides below A=100 for producing exotic ion beams at various accelerators. Use of transition metal nuclei for nuclear magnetic resonance spectroscopy will expand. In addition, calibration standards will be required for the newer techniques of radiological dating, such as the Sm/Nd and Lu/Hf methods, but in relatively small quantities. Most members of the research community would be willing to pay considerably more than they do now to maintain adequate supplies of stable isotopes.

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I. INTRODUCTION

In this document we will enumerate the present and future needs for separated stable isotopes, and the uses to which they are being (or will be) put, in physical and chemical research. Other authors will discuss biomedical, clinical, and industrial uses of stable isotopes. This information is especially relevant now, since problems have recently arisen which are beginning to impact the availability of such isotopes. In particular, supplies of many isotopes separated electromagnetically at the Oak Ridge National Laboratory (ORNL) have been exhausted, costs of new supplies have increased enormously, and the ORNL separators (calutrons) are beginning to feel their age.

Since World War II the basic research community, particularly in the field of nuclear physics, has provided the raison d'etre for having a substantial capability to separate isotopes in this country. This is true in the sense that nuclear physicists require a very broad range of separated isotope materials; at one time or another, separated isotopes of nearly all stable elements have been (and continue to be) used in nuclear physics research. Such diverse needs are not surprising, since the particular effects and properties which are of interest to nuclear physicists depend on the specific nucleus being studied, and can change drastically as a result of the addition or removal of even a single nucleon. Thus, investigating nuclear properties without separated isotope materials would be analogous to studying chemical reactions with impure reagents -- it becomes difficult, perhaps impossible, to correlate the observed effects with particular properties of the system under study.

[Of course, certain elements are mono-isotopic, but these constitute only 20 of the 280 stable isotopes. Many of them are utilized in physical and chemical research, but they will not be emphasized here since they require no separation. Furthermore, we will not discuss the needs and uses for gaseous isotopes, such as 3 He, or the relatively common radioactive sources, such as 252 Cf, 148 Gd, 57 Co, 22 Na, etc. These omissions should not be taken to reflect a lack of importance in either case. The use of 3 He for achieving very low temperatures ($<1^{\circ}$ K) is an extremely interesting topic, and even the lowly radioactive sources play a critical role in scientific endeavors ranging from undergraduate teaching at one extreme to calibrating detectors for high energy physics experiments and space shots at the other.]

While it is generally true that research scientists require a great variety of separated isotope materials, the quantities used by an individual researcher are rather modest, usually on the order of 10 to 1000 mg per year for each isotope. This is in contrast to the more "applied" uses of separated isotopes, where the tendency is to utilize a relatively few isotopes in rather large quantities. Maintaining a supply of a few high-demand items is clearly a more economical proposition than maintaining many different isotopically separated materials in moderate amounts. This is the problem which the research community is now facing.

To get an overview of the existing situation with regard to usage of stable isotopes by researchers in the United States, a questionnaire (see Appendix A) was sent to more than 1,000 physics and chemistry departments nationally, including both academic institutions and national laboratories. A list of the institutions contacted is provided in Appendix B; the names of those scientists responding to the survey are listed in Appendix C. Altogether, 231 institutions responded to this request for information, and 553 completed questionnaires were returned. Considering the length of the questionnaire, the number of respondents alone speaks eloquently for the importance the research community attaches to having a steady supply of separated isotopes.

Based on the responses, the research areas relevant to this report with significant requirements for separated stable isotopes are:

- (1) Nuclear Physics/Chemistry
- (2) Medium Energy Physics
- (3) Radjochemistry
- (4) Other Chemistry (Physical, Inorganic, Analytical)
- (5) Other Physics (Atomic, Solid State)
- (6) Geosciences

In Sec. II the separated isotope requirements for each of these areas will be indicated, along with examples of the uses to which the material is put. Of necessity, the selection of examples will not be complete, but should at least give some overview of recent work in the various fields. Problems associated with the present supplies of isotopes will be described in Sec. III, and Sec. IV will discuss trends in future requirements in these research areas. Finally, Sec. V will summarize the current study and present conclusions.

II. PRESENT USES AND REQUIREMENTS FOR STABLE ISOTOPES

A. Nuclear Physics

Nuclear physics involves the study of the structure and properties of nuclei, and is aimed at an understanding of the basic force which holds the nucleus together. The nucleus is perhaps the most versatile quantal many-body system available in nature. It exhibits a wide variety of interesting and complex phenomena, ranging from single particle features (shell structure) to collective features (vibrations, rotations).

In the last decade there have been considerable changes in the study of nuclear physics. For example, there has been a shifting emphasis from "light ion" beams (A \leq 4) to "heavy ion" beams (A \geq 6), and also a trend toward higher beam energies. These trends have opened up new vistas in nuclear physics research which are now being exploited. To give an overview of the needs for separated stable isotopes in this field, Table I lists reported usage in the last three years. Most of the isotopes have been used as target materials, although the use of some isotopes for producing beams has been reported. It is expected, however, that the use of isotopically enriched beam materials will increase significantly as the new generation of accelerators comes on the air. [It should be noted here that present machines often use gaseous isotopes, e.g., $^2_{\rm H}$, $^3_{\rm He}$, $^{18}_{\rm O}$, $^{86}_{\rm Kr}$, and $^{136}_{\rm Xe}$, as beams, but these have been omitted from the present survey because they are not electromagnetically separated.]

In the view of this author, the distinction between nuclear chemistry (as opposed to radiochemistry) and nuclear physics is somewhat arbitrary. Physicists, for example, have been known to study the fusion and deeply inelastic scattering processes, and chemists have been known to do DWBA

calculations. For this reason, the examples described below, which typify the uses of stable isotopes in nuclear physics, include topics from what is sometimes designated as nuclear chemistry. The area of radiochemistry is covered separately in Sec. II-C.

Light Ion Physics

High Resolution Work

Light ion beams, such as protons or deuterons, can be used to measure single-particle properties of nuclei. Because experiments using these beams can be done with very high resolution, a great deal can be learned about the fine details of nuclear level structures. For example, (p,p) resonance experiments have been carried out at the Triangle Universities Nuclear Laboratory with an overall energy resolution of about $400~{\rm eV.}^1$ Such experiments allow observation of various size resonances and determination of proton strength functions. This type of work necessitates the use of very thin and uniform targets. For the work described in Ref. 1, targets of $2~\mu a/cm^2$ (on a carbon backing) were employed.

Polarized Beams

Reactions of light ions can also be studied with polarized beams of protons or deuterons. In this case the measurements give information about various single-particle or collective states, 2 that is, 2 -values, spins and parities, spectroscopic factors, etc. Particularly in the case of spin determinations, the addition of analyzing power measurements can greatly enhance the sensitivity of the experiment. By making comparisons with theoretical nuclear structure calculations across a range of isotopes, it is possible to understand many details of the nuclear structure, such as the influence of deformation or pairing on level schemes, and whether a particular nucleus is better described as vibrational or rotational. For example, a coupled-channels analysis of polarized deuteron inelastic scattering data on the Ge isotopes (Ref. 2) lends support to theoretical predictions of a change in shape from oblate to prolate in going from ^{72}Ge to ^{74}Ge . Targets for this type of work are usually thicker than those required for

ultra-high resolution work. Typical thickness values would be in the 0.1-1.0 $\,\mathrm{mg/cm^2}$ range. Given the losses in the target preparation process, the amount of isotopically separated material required would be about 5-50 $\,\mathrm{mg}$ per target foil.

Ciant Resonances

The study of giant resonance phenomena gives interesting information on the collective structure of nuclei. Besides the familiar giant dipole resonance, evidence has been found in recent years for both giant quadrupole resonances (GQR) and giant monopole resonances (GMR) via inelastic scattering of protons and alpha particles. 4,5 Observation of the GMR allows a determination of the compression modulus of nuclear matter (which depends, in turn, on the nuclear force) if one measures the position of the resonance over a large mass range. In addition, the influence of deformation has been elucidated by comparing data from 144Sm and 154Sm. 5 The GMR apparently splits into two components in the deformed nucleus (154sm), one component being degenerate with the nearby GQR. One puzzling feature of the present data is the apparent absence of the GMR in lighter nuclei. A < 90. This aspect will be pursued in future work. Because the widths of the giant resonance states are large (several MeV), there is no particular advantage to thin targets. Target thickness values of 2-10 mg/cm² are not unusual in these experiments, leading to requirements for enriched materials of as much as 500 mg per target foil.

Exotic Nuclei

An area of light ion induced reactions which depends critically on the availability of separated isotopes is that involving the production of nuclei far from the valley of beta stability. Mass measurements of such exotic nuclei may be used to test predictions of various theoretical models, e.g., that of Garvey-Kelson⁷ or the extensive shell model calculations of Cole et al. As might be expected, most models predict the ground state masses of known nuclei reasonably well, but their predictions tend to diverge as one goes farther from stability. State of Experimentally, one finds that the cross sections for reactions leading to exotic nuclei are small, and that reactions

on isotopic target impurities often lead to considerably higher cross sections (and considerably less negative Q-values), potentially eliminating the ability to observe the more interesting rare nuclei. For example, the "uninteresting" reaction 64 Ni(a, 8 He) 60 Ni has a Q-value of -31.8 MeV and a cross section of about 30 nb/sr, while the corresponding values for the 58 Ni(a, 8 He) 54 Ni reaction, 12 which leads to an essentially unknown nucleus only two neutrons removed from doubly-magic 56 Ni, are -50.2 MeV and 0.5 nb/sr, respectively. Existence of a significant 64 Ni impurity in the target used by Tribble et al. 12 would have made the measurement impossible.

Work of this type 13-15 also allows the observation and mass measurement of complete isospin multiplets, and provides information on the possible existence of high-order charge-dependent forces in nuclei. Although evidence for such forces has been seen in very light systems (where some members of the multiplet are unbound to particle decay), recently completed measurements of the A=36 isospin quintet indicate 15 no evidence for deviations from the simple quadratic form of the Isobaric Multiplet Mass Equation when all members of the multiplet are bound against isospin-allowed particle decay.

Choice of a target thickness for this category of experiments requires making a compromise between the opposing requirements of good energy resolution (favoring thin targets) and adequate counting rates (favoring thick targets). In practice, the tendency is to favor the latter requirement, since poor resolution data are clearly better than no data at all. Typical targets would have a thickness of 0.2-2.0 mg/cm² and would require about 10-100 mg of enriched material for each foil. Even more critical for this work, of course, is the need for highly enriched materials, usually of the low abundance isotopes on the neutron-poor and neutron-rich extremes of the isotope distribution.

Heavy Ion Physics

Deeply Inelastic Scattering

One field of research specific to heavy ions is the study of the deeply inelastic scattering (DIS) process. This process involves the relaxation or

equilibration of a number of degrees of freedom, such as the mass distribution and the kinetic energy. Many researchers worldwide are seeking to determine the mechanism and time scale of the DIS process, and to understand the magnitude and alignment of the angular momentum transferred during the reaction and the nature of the de-alignment that occurs with increasing contact time (increasing kinetic energy loss) between the fragments. Observation of the angular correlation of fragments from the sequential fission of the recoiling target-like nucleus has shown 16 that the angular momentum transfer to the target fragment in DIS of $^{86}{\rm Kr}$ + $^{209}{\rm Bi}$ or $^{238}{\rm U}$ is quite large, J ~ 40 ft, and that the alignment is also quite high, P_{zz} ~ 0.85. Experiments on similar systems which include detection of emitted neutrons have shown $^{17-19}$ that the kinetic energy damping and the neutron-proton ratio equilibrate very rapidly, in about $(5\text{-}10)\times10^{-22}$ sec.

Targets for these experiments are generally about 0.1-0.5 mg/cm^2 and require about 5-25 mg of material for each foil. For experiments where only light particles are of interest, thicker targets (1-2 mg/cm^2) are usable, provided they can survive the heating due to the beam without melting.

Anomalous Large Angle Scattering

One of the more interesting phenomena discovered in recent years is the so-called Anomalous Large Angle Scattering (ALAS) seen in certain heavy ion systems. Although qualitatively similar behavior had been observed in the scattering of very light heavy ion systems, e.g., 16 O + 12 C, 20 in which exchange effects might be expected to play a role, its appearance in heavier systems, such as 16 O + 28 Si (Ref. 21) and 12 C + 28 Si (Ref. 22), was unexpected. The ALAS phenomenon, which also manifests itself in excitation functions, 22 , 23 seems to fall outside the standard optical model description of elastic scattering. 24 Structure effects are clearly important here, since the addition of a single nucleon to the lighter fragment, i.e., 13 C + 28 Si, is enough to completely damp out the oscillations. 22 On the other hand, adding one or two nucleons to the heavier fragment, e.g., 16 O + 29 Si or 16 O + 30 Si, reduces the cross sections by a factor of about 5, but does not eliminate the structure. 25 The most recent results 25 suggest that the structure begins to disappear beyond about 20 Completely, but this cannot

be concluded firmly without higher energy data. Although various attempts have been made to explain the existing data, it cannot as yet be said to be fully understood.

In this type of experiment, much of the "back angle" data are actually measured with reverse kinematics at forward angles, i.e., a beam of $^{28}\mathrm{Si}$ cm a target of $^{A1}2^{O}_3$. Because of the requirement for rather good energy resolution, targets are usually fairly thin, about 0.1 mg/cm². Thus, only 5 mg of enriched material might be needed for each target. On the other hand, a separated isotope beam might require several grams of material.

Fusion

There have been extensive studies of the energy dependence of the fusion process in the lighter heavy ion systems. In certain systems composed of "alpha-particle" nuclei, such as 160 + 12C, unexpected oscillations have been observed 26 in the fusion cross sections in the energy region where they begin to deviate markedly from the reaction cross section. Other nearby systems, such as $^{18}O + ^{12}C$ and $^{19}F + ^{12}C$, however, do not show such behavior 27 and seem to have a very smooth energy dependence to the fusion cross sections. It is now known²⁸ for ^{16}O + ^{12}C that most (>80%) of the fusion cross section goes into just three products, 20Ne. 23Na, and 24Mg, and that the oscillations appear in all channels, but mainly in the ²⁰Ne channel. Also, measurements using gamma-ray detection techniques indicate²⁹ that, in contrast to Ref. 27, the $^{18}\mathrm{O}$ + $^{12}\mathrm{C}$ system does show oscillations, albeit considerably weaker ones than those seen in the $^{16}\mathrm{O}$ + $^{12}\mathrm{C}$ system. Evidence on whether the structures observed in the fusion cross sections correlate with those seen in elastic scattering is presently contradictory. 26,28 It does seem clear, however, that a full understanding of the phenomenon will require careful studies in nearby systems.

In determining fusion cross sections directly by detecting evaporation residues, the energies of the outgoing fragments are not very high, and the cross sections are reasonably large. Moreover, the search for structure in the fusion excitation functions requires that the thickness of the target (in terms of energy loss and straggling) be small compared with that of the structure being investigated. These aspects dictate very thin targets

(<0.1 mg/cm²) for the type of measurements reported in Refs. 26-29, with a corresponding requirement of less than 5 mg of material for each target. In the taboratory energies of the recoiling fragments are reasonably high. Thus, in these cases somewhat thicker targets, say 0.5-1.0 mg/cm², are often acceptable; this means that 25-50 mg of enriched material are needed for each target foil.

Another area of research related to the study of the fusion process involves the question of "complete" versus "incomplete" fusion. At high bombarding energies, Zolnowski et al. 30 have observed a considerable number of forward-angle, high-velocity alpha particles which are in coincidence with fusion-like procestes. Even in very heavy systems there observations 31,32 of considerable emission of forward-peaked protons and alphas which appear in coincidence with fission fragments. The observation in heavy systems of $\mathcal{L}_{\mathrm{crit}}$ values for fusion which substantially exceed the ℓ value for which the liquid drop model predicts the fission barrier to vanish may be related to angular momentum removal by these fast particles. It has been demonstrated in at least one case 33 that the fast alpha particles emitted during incomplete fusion reactions do tend to come selectively from the higher partial waves. In any case, if the $suggestion^{31}$ that these particles are emitted very early in the reaction process turns out to be correct, they will provide an important probe of this interesting stage of the interaction between two heavy ions.

The study of incomplete fusion described in Ref. 30 is carried out by measuring alpha particles in coincidence with gamma rays. For these measurements the target thickness is generally not a limitation, and thickness values in the range of 0.5-5.0 mg/cm² have been utilized. This corresponds to about 25-250 mg per target.

High Spin States

One of the special features of heavy ion projectiles is their ability to import considerable angular momentum to a nucleus, via either (HI,xn) reactions or Coulomb excitation. In several rare earth nuclei, (HI,xn) reactions have shown that there is an anomalous increase in the moment of

inertia that occurs at about the I=12 level; this phenomenon is called "backbending." It is explained in terms of the crossing of the ground state band by another even-parity band with a higher moment of inertia. This idea has been nicely demonstrated recently for the nucleus $^{164}{\rm Er}$ by Kistner et al. 34 and by Yates et ai. 35 Using both (HI,xn) and Coulomb excitation techniques, Yates et al. were able to follow the ground band sequence to I=22* and also to locate an even-spin "superband" with a large moment of inertia which crosses the ground band at !=12* as well as two other bands (one positive and one negative parity) which also have large moments of inertia. Experiments of this type provide very detailed tests of nuclear structure models such as the rotation alignment model 36 or the Coriolis antipairing model. 37

Another feature of heavy ion reactions important for nuclear structure studier is their ability to impart considerable linear momentum (recoil velocity) to the final system. To take advantage of this aspect, Andrews et al. 38 have studied continuum states in rare earth nuclei by means of Coulomb excitation experiments with $^{86}\mbox{Kr}$. If the products recoil out of the target into a polarized ferromagnetic medium, they experience a very large transient magnetic field (* 4000 T) which allows determination of g-factors of states having sub-picosecond lifetimes. In studying the Coulomb excitation of $^{160}\mbox{Dy}$ and $^{170,174}\mbox{Yb}$, for example, Andrews et al. showed that the g-factors decreased at higher spin values, suggesting a weakening of neutron pairing relative to proton pairing.

All of the experiments discussed in this subsection involved detection of gamma rays. For this reason, the targets can be reasonably thick. A typical target thickness for a Coulomb excitation experiment would be about I mg/cm² (requiring about 50 mg of enriched material), while for the (HI,xn) experiments targets of 5-50 mg/cm² have been employed (requiring 250-2500 mg of material per target).

Exotic Nuclei Revisited

The availability of heavy ion projectiles has considerably enhanced our ability to produce and study nuclei far from stability, and thus provides an opportunity to observe nuclei under very unusual conditions. Because of the

wide variety of products which can be formed in a given reaction, such studies frequently employ on-line isotope separators, such as UNISOR, 39 Tristan, 40 RAMA, 41 etc. A typical target thickness here would be 1-3 mg/cm², corresponding to about 50-150 mg of material. (A recent survey of developments in this field can be found in the proceedings cited in Ref. 39.) Many interesting nuclear properties can be observed in this fashion, e.g., coexistence between spherical and deformed shapes, 42 existence of triaxial shapes, 43 etc. These properties are best elucidated by following the systematic trends of particular levels across a series of isotopes, e.g., $^{187-201}$ Tl. In addition to the normal spectroscopic techniques, it is also possible 45 to use the mass-separated beams to measure hyperfine splittings and isotope shifts, which will yield information on spins and rms charge radii for nuclei heretofore inaccessible. This topic will be discussed in Sec. II-D below.

The deeply inelastic scattering process discussed earlier can also be used as a means to produce exotic nuclei. It has been shown 46 that the yield of projectile-like fragments can be explained in terms of a statistical transfer of particles between the target and projectile while the two are in contact, followed by a statistical evaporation of light particles from the highly excited primary projectile-like nuclei. As a result, several groups have demonstrated 46,47 that choosing a neutron-rich target can lead to production of exotic nuclei, such as $^{41}{\rm Cl}$, $^{53}{\rm Sc}$, and $^{55}{\rm Ti}$, with cross sections of 0.1 to 1.0 mb. A target thickness of about 0.5-1.0 mg/cm² (requiring 25-50) mg of material) is typical for this type of experimental work.

Another worthwhile technique for the study of exotic nuclei employs the fragmentation of relativistic heavy ion projectiles. Westfall et al. 48 produced 14 new nuclides, 22 N, 26 F, 33,34 Mg, 36,37 Al, 38,39 Si, 41,42 P, 43,44 S, and 44,45 Cl, by looking at projectile fragments at 0 arising from interactions of 48 Ca + 9 Be at a calcium beam energy of 212 MeV/A. Given the availability of suitably enriched isotopes, there is no reason to believe this technique would not work equally well with a beam of, sey, 96 Zr or 100 Mo. The thickness of the 9 Be target (900 mg/cm 2) was of secondary importance in this experiment, since the beam was the actual source of the interesting reaction products. The amount of source material required for a

 48 Ca beam is about 0.5 g of enriched material for each electrode (3 are available). Fortunately, it is possible to recover roughly 80% of the unused material from the ion source parts. For the experiment described in Ref. 48, the estimated use rate of 48 Ca was about 10 mg/day prior to recovery, or a net usage of about 2 mg/day.

Neutron Physics

Radiative Capture

The study of nuclear structure with slow neutrons is one of the oldest branches of nuclear physics. Many early experiments were accomplished with neutrons from Ra-Be sources, but modern work takes advantage of the high neutron fluxes available at research reactors, such as the High Flux Beam Reactor at Brookhaven National Laboratory. A major use of reactor neutrons is for radiative capture, i.e., the study of (n,Y) reactions 49,50 using thermal or epithermal neutrons. Compared with most other types of nuclear physics experiments, targets for radiative capture can be quite thick: 5-10 g of enriched material are often required. [Fortunately, however, these samples are used with low beam intensities which do not cause significant activation. Thus, the required isotopes can sometimes be borrowed from the Loan Pool at ORNL.]

One virtue of radiative capture (in contrast to charged-particle) work is its non-selectivity 51 -- essentially all nuclear levels with spins close to that of the capturing state are populated. Thus, radiative capture reactions allow sensitive tests of various nuclear models by making it possible to observe many of the rotational bands in nuclei 52 (and sometimes the transitions between them) as well as such quantities as E1 and M1 , noton strength functions in a wide range of nuclei. $^{49},^{53}$ Experiments can be done with very high precision by detecting the emitted gamma rays with a curved crystal spectrometer rather than a Ge(Li) counter. Also, knowledge of transition multipolarities can be greatly extended by measurements of conversion electrons with a suitable spectrometer. This is necessary to use much smaller samples, i.e., several milligrams rather than several grams, in order to prepare small, intense sources for the

spectrometer. These experiments are usually carried out with an in-pile target in order to maintain acceptable rates.

An example of the structure information which can be obtained via radiative capture is given in Ref. 51. In this experiment, levels of $^{109}\mathrm{pd}$ were in higher to locate the $\mathrm{g}_{7/2}$ and $\mathrm{h}_{11/2}$ neutron strength. Earlier (d,p) experiments 50 had indicated that, contrary to shell model expectations, the $\mathrm{g}_{7/2}$ orbital in $^{109}\mathrm{pd}$ was nearly empty, while the normally higher lying $\mathrm{h}_{11/2}$ orbital was nearly full. Casten et al. 51 have resolved this question by demonstrating that many of the $7/2^+$ assignments in the earlier work were incorrect. After making appropriate modifications to the previously reported spectroscopic factors, the apparent discrepancy was eliminated.

Besides the uses of separated stable isotopes for target materials, several particular isotopes are used for other special purposes. One example is the use of ^6Li and ^{10}B as neutron attenuators; this requires several kilograms of each isotope. Another example is the use of ^{45}Sc and ^{56}Fe as neutron energy filters. Because of destructive interference between the s-wave resonance and potential scattering amplitudes, each of these isotopes has a particular energy region (1.95 \pm 0.5 keV and 24.3 \pm 1 keV for Sc and Fe, respectively) where the total neutron cross section has a sharp minimum. In this region a more or less monoenergetic flux of neutrons is transmitted. About 10 kg of ^{56}Fe were needed to build the neutron filter at BNL. The year the material was obtained from ORNL it put the basic research community back into the big leagues of isotope usage - but only temporarily!

Neutron Scattering and Total Cross Sections

Nuclear deformations can be observed by looking at either the charge distribution or the matter distribution. (In principle, these need not be the same.) Neutrons can serve as an effective probe of the matter deformation of nuclei (as opposed to the charge deformation, which can be studied via electron scattering) by measurements of elastic and inelastic scattering and also total cross sections. Shamu et al. 55 have made a detailed study of deformation effects in various pairs of rare earth nuclides, such as $^{148,154}{\rm Sm}$, over the energy range from about 1-14 MeV. (In each case about 40 g of isotopically enriched material was used as a target.) They found

rather marked effects (as much as 10%), which are quite accurately reproduced with coupled-channels calculations where the influence of the first 2⁺ state is explicitly includeu. The calculations also shed light on the question of whether a rotational or vibrational coupling scheme gives the more appropriate description of the nuclei studied. Thus, it has been nicely demonstrated that neutron total cross section and scattering measurements offer a very precise means of studying deformations of the matter distributions of nuclei.

Electromagnetic Interactions

Electroexcitation

Electron scattering can be utilized to gain a better understanding of various aspects of nuclear sizes and nuclear structure. One example is the search for giant multipole states. We have already discussed the ability of light ion inelastic scattering to locate such states, but inelastic electron scattering can also be used for this purpose. With light targets, electron scattering experiments are quite sensitive to the distribution of quadrupole as well as dipole strength. In 24 Mg, for example, all of the GQR strength was observed in the (e,e') reaction 55 at energies from 100-200 MeV, while only about 50% of the strength was found using inelastic alpha-particle scattering. Determining the spreading of the E2 strength will ultimately allow a sensitive test of nuclear structure calculations in this mass region.

Single-particle transitions are also well identified in (e,e') experiments in light nuclei. Using 70-340 MeV beams from the Bates Linear Accelerator, Hicks et al. 56 studied the 27 Al(e,e') reaction and were able to distinguish the character of the "particle" states (configurations based on excitation of an sd-shell particle into the fp-shell) from the "hole" states (configurations based on a 1p-shell hole in a 28 Si core) via the different form factors for the electroexcitation. Comparisons with both strong- and weak-coupling models indicate that neither is entirely satisfactory in explaining the spectrum of the negative-parity levels in 27 Al. Here too, mcderately thick targets, 10-50 mg/cm², are employed; the corresponding requirement for separated isotopes would thus be about 500-1000 mg per target.

Photonuclear Reactions

Photonuclear reactions allow study of the decay properties of the GDR in heavy nuclei, including photoneutron emission and photofission, 57 because photons are most strongly absorbed into this "state." From the total photoabsorption cross sections, values for the axis ratio of deformed heavy nuclei can be determined or, equivalently, the deformation parameter β_2 or intrinsic quadrupole moment. The values obtained in Ref. 57 by this means agree well with values derived by other techniques. In addition, data on the relative amounts of first- and second-chance fission could be obtained; this separation may aid substantially in our understanding of the details of the fission process. Sample sizes used for these experiments ranged from 20 to nearly 300 q.

In light nuclei, it is possible to observe photoneutron emission from many low-lying states reached via E1, M1, or E2 photoexcitation. Holt et $\varepsilon 1.58$ have studied this process with a $^{13}\mathrm{C}$ target and determined the ground state radiative widths based on a multilevel, multichannel R-matrix analysis. Surprisingly, the results for the strongly interfering E1 excitations at 7.69 and 8.19 MeV agree well with simple weak-coupling wave function predictions but disagree with more sophisticated shell-model calculations. The targets employed in studies such as this can be quite massive; that used in Ref. 58, for example, was about 40 grams.

Photon Scattering

The measurement of elastically and inelastically scattered photons can also provide valuable information on nuclear properties. Compared with scattering of charged particles, photons have the advantage of interacting with nuclei in a well-known fashion, undisturbed by Coulomb effects. Measurements of photon scattering angular distributions allow a determination of the strength distributions for low-multipole giant resonances (E1, M1, E2). In a recent experiment, Bowles et al. 60 used nearly monochromatic photons to measure both elastic and inelastic photon scattering to the GDR region of ^{52}Cr , ^{60}Ni , and $^{92,96}\text{Mo}$, and have compared their results to a model which calculates photon decays of the GDR including coupling to the low-lying quadrupole states. Strengths of the inelastic scattering to the $^{2+}$

levels in these nuclei are at least qualitatively reproduced by the model, but it was found necessary to include an additional isospin splitting of the GDR in the calculations in order to obtain detailed agreement with 'ne data. Here too, targets of several tens of grams can be conveniently employed.

Weak Interactions

Double Beta Decay

Moe and Lowenthal 61 are pursuing ar, experimental hunt for double beta decay in 82 Se. They hope to find direct evidence for this process, either with or without neutrino emission. The choice of 82 Se was based on its very favorable decay energy (3 MeV), its chemical stability in elemental form, and existing geochemical evidence for a half-life for double beta decay (~ 10^{20} years) which is within the range of a direct measurement. Using 38 grams of 97% enriched 82 Se, a target was fabricated which can be viewed by a cloud chamber. Based on the observation of 20 double electron events which had an energy and angular distribution consistent with the process of double beta decay with neutrinos (and which did not appear to be caused by background events), Moe and Lowenthal feel they have obtained suggestive, but not conclusive, evidence for this process. The tentative half-life based on the present observations is about 1×10^{19} years. Improved experiments are under way to obtain more definitive results.

B. Medium Energy Physics

Medium energy physics is an extension of lower energy nuclear physics into a regime where meson production becomes significant. One of the important topics being studied is that of the matter distributions in nuclei. (High energy electron scattering does a good job of providing information on the proton distributions in nuclei but provides relatively little sensitivity to the neutron, or matter, distributions.) A strongly interacting probe seems to be most suitable for investigating the neutron distribution in nuclei. In fact, it appears that pions may be an excellent choice for this task. The reason is that, in the energy region near the (3,3) resonance, the scattering of

negative pions is much more sensitive to neutrons than to protons -- a unique feature among strongly interacting probes. Hopefully, the interaction of pions with nuclei will ultimately provide information not only on nuclear sizes but also on the equation of state of nuclear matter, pion condensation, etc.

At existing medium energy accelerators, e.g., the Los Alamos Meson Physics Facility (LAMPF), the fluxes of π^+ and π^- beams are still rather modest (~ 108/sec and ~ 107/sec, respectively) compared with typical nuclear beams, and the beam sput sizes can be fairly large. For these reasons, the amount of target material required for a medium energy physics experiment is generally considerably larger than that required for most nuclear physics work. Typical target thicknesses for the experiments described in this section are in the range of 100-1000 mg/cm², compared with a value of more like 1 mg/cm² in the majority of the work described in Sec. II-A. The use of separated isotope materials in medium energy physics for the past three years is shown in Table II. Although the variety of isotopic species is presently much less than for nuclear physics (cf. Table I), the amount per sample is usually in the range of 10-100 grams. Note also that for those elements which are presently required, there is generally a need for a wide range of isotopes. This has to do with the fact that, at the present stage of development, it is often prudent to compare data from several isotopes in order to determine the requisite nuclear structure information in the most model-independent fashion. Several examples of this approach will be discussed below.

Proton Scattering

Neutron Density Determination

Elastic scattering of medium energy protons (E $_{\rm p}$ $^{\sim}$ 1000 MeV) is a powerful technique for investigating the matter distribution of nuclei. Such investigations give important information which may be compared with theoretical nuclear structure determinations of ground state matter densities, such as those from Hartree-Fock calculations. A group at LAMPF, for example, has recently completed a study of the p + $^{208}{\rm Pb}$ system at

 $E_p=800~{\rm MeV}.^{62}$ They analyzed their data in terms of a spin-dependent optical potential formalism which uses on-shell proton-nucleon scattering amplitudes as its basic input and treats the target protons and neutrons independently. Hoffmann et al. 62 used a three-parameter Gaussian form for the neutron density in $^{208}{\rm Pb}$ and took the proton density from existing electron scattering data, as described in Ref. 63. A careful error analysis, including both experimental errors and uncertainties related to the choice of nucleon-nucleon scattering amplitudes, indicated that the neutron density is determined very well from about 3.5 to 9.0 fm, and that the rms neutron radius in $^{208}{\rm Pb}$ is 5.593 fm, or 0.14 \pm 0.04 fm larger than the corresponding proton radius.

A similar analysis was carried out in Ref. 64 for the Ca isotopes. In that case, it was found that the <u>absolute values</u> for the neutron radii in the various isotopes depend markedly (* 0.2 fm) on the input scattering amplitudes chosen. On the other hand, the <u>relative differen</u> es in the neutron radii between isotopes were essentially independent of the choice of scattering amplitudes and were therefore greatly superior in terms of a meaningful comparison with theory. A study by Ray et al. 64 indicated that Hartree-Fock calculations do a good job of predicting the relative neutron radius changes with respect to ⁴⁰Ca for all the isotopes except ⁴⁸Ca, whose neutron skin is not as thick as had been predicted. Targets used for these experiments ranged in thickness from 20 mg/cm² at forward angles to 150 mg/cm² at backward angles.

Pion Induced Reactions

Pure Proton and Neutron Transitions in Nuclei

In pion scattering from free nucleons near the (3,3) resonance, the ratio of π^- to π^+ cross sections is about 1/9 for protons and 9 for neutrons. In studies of π^- and π^+ scattering leading to collective states in even-even nuclei, however, the observed ratios are close to unity, presumably reflecting the more or less equal contributions of proton and neutron excitations. In contrast to these results, two recent studies of π^+ and π^- inelastic scattering from $^{13}{\rm C}$ at $^{12}{\rm C}$ MeV 65 and $^{18}{\rm C}$ MeV 66 have located a state at 9.5 MeV

with a strong enhancement in π^- scattering and another state at about 16 MeV with strong π^+ enhancement. Compared with free-nucleon values, the enhancement of the 9.5 MeV state is essentially consistent with a pure neutron excitation, while the state at 16 MeV is consistent with a pure proton excitation. Several lower-lying states which are thought to be mainly single-neutron configurations do not show much enhancement, which suggests that they may be significantly admixed with proton particle-hole components. These data are an encouraging sign that pion scattering will become an increasingly important tool in the study of nuclear structure.

This comparison technique was recently extended 67 to pion scattering from a $^{14}\mathrm{C}$ target, with even more striking results. In this case, enhancement factors even larger than the free-nucleon estimate of 9 were found for several states. Preliminary analysis indicates that this can be explained in terms of destructive interference between the proton and neutron components of certain 2-hole and 2-particle-4-hole configurations in the wave functions.

As mentioned earlier, the low beam intensities available for these experiments dictate thick targets. For the work described in Ref. 65 the 99% enriched 13 C target had 72.5 g of material, while 9 g of 14 C was used for the experiment described in Ref. 67.

Pion Charge Exchange

Analog states in nuclei, where all quantum numbers are the same except for the isospin, have been known and studied for many years. ⁶⁸ Recently, Beer et al. ⁶⁹ have investigated the (π^+ , π^0) reaction on a variety of targets, ranging from ^7Li to ^{208}Pb , at a pion energy of 98 MeV. In each case a very strong peak was observed in the π^0 spectrum at precisely the energy expected for the analog state. Calculations are under way to use such states to obtain more information on the isospin-dependence of various higher-order terms of the pion-nucleus interaction responsible for their production. Because of the low π^0 production rate, exceedingly thick targets, ranging from 500-1200 mg/cm², were used for this experiment.

Several other groups 70,71 have been studying the pion double charge exchange (DCX) reaction (π^+ , π^-). This process can probe high isospin

states and can be used to produce exotic nuclei. In a simple picture, it would be expected that this reaction would populate mainly the analog state in the final nucleus. However, the data of Seth et al. 70 for the reaction $^{18}\mathrm{O}(\pi^+,\pi^-)^{18}\mathrm{Ne}$ show that the non-analog $^{16}\mathrm{Ne}(2^+)$ state is populated with the same intensity as the analog transition to the ground state. In addition, they found that they could not fit their angular distribution for the $^{13}\mathrm{Ne}(\mathrm{g.s.})$ with any of the existing reaction theories for the DCX process. Fortunately, the same process, when studied 71 at a higher beam energy of 292 MeV, does appear to exhibit the expected selectivity. Thus, it appears that the DCX reaction will be extremely useful for locating "T+2" analog states in nuclei. For these difficult experiments, targets as thick as 900 mg/cm 2 have been employed.

X-Ray Studies

Nuclear Charge Distributions

It is by now well known that x-ray measurements of muonic atoms can provide very sensitive information on the nuclear charge distribution, including in some cases both static and dynamic quadrupole moments. Powers et al. 72 have studied muonic x-rays (transition energies and hyperfine splittings) in the whole series of isotopes 144,147,148,149,150,152,154 Sm and analyzed their data in terms of a deformed Fermi distribution for the charge density of each isotope. They found that such an analysis must take into account the possibility that the nucleus will be excited during the course of the muon cascade; due to its large deformation, the probability that 154 Sm will be raised to its $^{2+}$ first-excited state is nearly 40%. This excitation shifts the position of the 1s atomic level by more than 1 keV. In terms of equivalent charge radii, Powers et al. 72 found that the charge from 144 Sm to 154 Sm is 0.19 fm, a result which agrees well with other methods of determining this quantity.

Pion-Nucleus Potential

Although the bound atomic pion is not especially useful as a probe of nuclear charge distributions (due to the fact that it interacts via the strong force as well as the well-understood electromagnetic interaction), it offers the possibility of determining some of the properties of the pion-nucleus interaction potential. This can be done by choosing "reference" nuclei for which the charge distribution is already well known from electron scattering and/or muonic atom results, and for which the neutron density is calculable from a good model, such as Hartree-Fock. Powers et al. 73 have used the isotopes of even-even nuclei with 6 < Z < 16 for this purpose. phenomenological potential was obtained from these data by fitting the x-ray results using the Klein-Gordon equation along with a complex optical potential of the type suggested by Ericson and Ericson. 74 Experiments were performed on a series of "test" nuclei, 40,42,43,44,48 Ca and 46,48,50 Ti. to see if the empirical potential parameters derived from the reference nuclei would lead to a correct determination of neutron distributions in the test cases. Since the stopped pion rate was very low, $5x10^{-6}/(g/cm^2)$ -sec, target materials ranged from 2.5 to 14.7 q samples. Data were obtained with a Ge(Li) detector for both transition energies and widths for the 3d to 2p transitions in the Ca and Ti isotopes. At least for these cases, the empirical potential leads to reasonably good agreement for the neutron radius shifts compared with determinations by other techniques (e.g., Ref. 64). If the potential parameters can be fixed in other mass regions, the technique of using pionic atoms for information on the neutron distribution can be extended.

C. Radiochemistry

The question of what topics qualify as "radiochemistry" in the context of stable isotope usage is somewhat fuzzy, since techniques involving radiochemical methods have found wide applicability in many areas of research. Radiochemistry will be used here to mean those areas of nuclear chemistry which utilize chemical techniques for at least a part of their research effort. The topics to be covered here include transactinide

chemistry, fission studies, high energy spallation reactions, and the search for superheavy elements. Nuclear or radiochemical techniques used in the study of other types of physical and chemical problems (e.g., Mössbauer spectroscopy and perturbed angular correlations) will be covered in Secs. II-D and II-E.

Table III shows the use of stable isotopes in radiochemistry over the last three years. Compared with Tables I and II, the distribution of isotopes in Table III tends to be somewhat more concentrated in the high mass half of the periodic table.

Heavy and Superheavy Elements

Transactinide Chemistry

One important area of radiochemical work involves the study of the chemical properties of transactinide elements. Such information can be valuable in making a determination of the atomic number of an unknown radioactive species, as well as for establishing the chemical behavior of yet-to-be-discovered heavy elements (see below), where a chemical separation might be a necessary means of identification. In practice, such studies are becoming more difficult both because of short half-lives for many newly discovered heavy nuclides and because of exceedingly small production cross sections.

The chemistry of element 104, for example, has been studied 75 to see if it behaves (as predicted) as a chemical homolog of Group IV-B elements Zr and Hf. The isotope 261 [104] was produced in the reaction 248 Cm(18 O,5n) using a (degraded) beam energy of 98 MeV. Because the half-life of this nuclide is only 65 seconds, a computer-controlled fast chemistry apparatus was employed; this device allowed a repetitive chemistry experiment to be done every three minutes. To ensure that the behavior of element 104 was similar to that of Hf, a 181 Hf tracer was used in some of the chemistry runs. In spite of detecting only 6 atoms of 261 [104], Hulet et al. 75 were able to demonstrate that its chemical behavior differs markedly from Cm and Fm isotopes, and that it is similar to that of Hf.

Search for Superheavy Elements

In recent years, a considerable amount of effort has none into the attempt to produce and identify so-called superheavy elements (SHE), 76,77 These are elements with 77114 which are expected to have enhanced stability against fission due to the presence of closed shells. At present, it appears that the most promising experimental approach is the study of the ⁴⁸Ca + ²⁴⁸Cm reaction. A previous study ⁷⁶ of this reaction produced no evidence for SHE formation. However, one possible "gap" in the search was the fact that the chemical techniques utilized were not very sensitive to highly volatile products. (Elements 112 and 114 are chemical homologs of Hg and Pb, respectively, and thus are expected to have very low boiling points -- that is, below that of Hq. In addition, relativistic Hartree-Fock calculations suggest that the electronic configurations of these elements will be rather stable, leading to the prediction that they may behave somewhat like noble gases.) Illige et al. 77 repeated the earlier experiment using a 267 MeV 48Ca beam incident on a 24 µg/cm² 248Cm target. experiment, all non-volatile products were eliminated from the detection system, and any volatile products were trapped on a liquid-nitrogen-cooled surface. Unfortunately, no SHE's (in the half-life range from 1 sec to 10^7 sec) were observed in 4 months of counting. Based on updated theoretical predictions 78 recarding the dynamics of the fusion mechanism, however, there is a possibility that the bombarding energy chosen was unnecessarily high. Therefore, new experiments will be conducted at a lower beam energy, and will use physical as well as chemical techniques to span the widest possible range of half-lives.

The amount of the rare isotope ⁴⁸Ca used in these experiments is considerably higher than that used for the experiment described in Ref. 48 (see Sec. II-A). Estimated usage here is about 200 mg/day in the ion source; assuming 80% recovery leads to a net usage of 40 mg/day. Should evidence for SHE production be obtained, a further supply of ⁴⁸Ca would probably be needed. In addition, usage of very heavy target materials would surely increase.

Fission Studies

Fission decay of heavy nuclei is a process which is being extensively studied in a variety of different reactions induced, for example, by thermal neutrons, 79 fast neutrons, 80,81 light ions, 82 heavy ions, 83,84 pions, 85 and even anti-protons. 85 Radiochemical techniques have frequently been employed in such studies, 80,84 since they can provide both Z and A resolution not generally available in experiments where the fission fragments are detected directly. Because the objective of such studies is to determine the fission properties of one particular nuclide, highly enriched targets are very important.

Several research groups have focused on the determination of mass yields of fission products, primarily via gamma-ray and sometimes via beta-particle spectroscopy. Besides providing data aimed toward a theoretical understanding of the fission process, accurately known fission yields can provide information on fuel burn-up in a nuclear reactor. If the may be necessary, however, to have more precise information for this application than can generally be obtained radiochemically. In this instance, mass spectrometric methods may be utilized. Such studies are generally pursued at industrial laboratories and will not be discussed further here.

Weber et al.⁸² have compared data for ²⁵²Cf spentaneous fission to that obtained for fission of ²⁵²Cf at excitation energies near the fission barrier produced in the ²⁵⁰Cf(t,pf) reaction. They found that the total kinetic energy (TKE) released in the induced fission process is about 5 MeV greater than that from the spontaneous fission. Interestingly, however, the shift toward symmetry in the mass spectrum for induced fission was accompanied by an increase in TKE. This is in contrast to results from other cases in which a symmetric mass split occurs with a somewhat reduced TKE release.

Difilippo et al. 81 studied data on neutron-induced fission with very high resolution. An attempt was made to explain the observed strong resonances at 720 and 1210 eV in terms of population of Class I levels (in the inner well of a double-humped fission barrier) or Class II levels (in the outer well), but their data do not allow an unambiguous choice.

Husain and Katcoff⁸⁵ have measured the fission cross sections for Au, Bi, and U targets induced by 1730 MeV anti-protons and 2360 MeV negative pions. They observed that the fission cross section for pions was essentially the same as that for comparable energy protons, but the fission cross section induced by anti-protons was nearly double that for protons. This may be related to the very high excitation energy obtained when the anti-proton annihilates in a nucleus.

High Energy Reactions

Study of the distribution of nuclides produced in high energy nuclear reactions induced by light and heavy ions can provide interesting information on the reaction mechanism. In most cases the range of products is rather broad, and is therefore well-suited to the global nature of radiochemical techniques. (Such studies are also pertinent to other fields of research, such as cosmic ray investigations, cosmochemistry, etc.) One example of this type of work can be found in Ref. 87. in which measurements were reported of the spallation products arising from interactions of 720 MeV alpha particles with targets of 92,96,100 Mo. The data demonstrated that, in spite of previous predictions to the contrary, the yields of spallation products do "remember" the N/Z ratio of the target nucleus. Analysis of the isobaric yields for A=72 showed that the distribution was shifted toward a higher N/Z ratio for the more neutron-rich target, and also that the distribution from this target was skewed toward the high N/Z side. In fact, the most probable N/Z ratio appears to depend linearly on the combined N/Z ratio of the target + projectile system. Several empirical predictions for the isobar vields are able to reproduce the peak shift of the distribution but are presently unable to predict the skewing of the yield curves for different targets.

With the advent of relativistic heavy ion accelerators, experiments can now be performed with a variety of projectiles. Loveland et al. 88 have applied thick catcher foil techniques to the study of products from the reactions of 8000 MeV 20 Ne + 181 Ta at the Bevalac. They found that the condition of "limiting fragmentation" (where the product yields become

independent of the bombarding energy) has apparently not been reached for this system. In addition, they found that products from the Ne + Ta reaction are more forward-peaked than those from proton or $^{12}\mathrm{C}$ bombardments of similar mass targets. Thus, it appears that relativistic heavy ion collisions behave differently than do those of lighter projectiles and that they provide a unique opportunity to gain significant new information on the reaction mechanisms in high energy collisions.

D. Other Chemistry

Aside from the specifically "nuclear" aspects of chemical research, there are several other areas of chemistry which depend critically on the availability of enriched stable isotopes. By far the dominant use is for nuclear magnetic resonance (NMR) spectroscopy. Separated isotopes used in this area include ²H. ¹³C. ¹⁵N. and ¹⁷O: all of these, but especially ²H. are utilized in quantities ranging up to hundreds - or even thousands - of grams per year by individual research groups. Another area of chemistry where separated isotopes are routinely used is molecular spectroscopy; substitution $_{
m of}$ $^{18}{
m O}$ (as well as most of the aforementioned isotopes) into a molecule, for example, will change the rotational or vibrational frequency and thereby make it possible to verify band assignments in such work. 89 An analogous technique is sometimes utilized in mass spectrometric measurements of various molecular reaction products, which can be tagged via isotopic substitution.⁹⁰ Finally, isotope effects on various chemical properties, such as vapor pressure, have been investigated. 91 The technique of Mössbauer spectroscopy, which is important to both physics and chemistry research, will be discussed in Sec. II-E.1 Because most of the uses of stable isotopes in chemistry involve the relatively light elements (H, C, N, O) or gases, they will not be covered here. There are, however, growing numbers of chemistry experiments which do utilize electromagnetically separated isotopes. A few examples of such uses appear below. As can be seen from Table IV. the amounts involved are comparatively small, but probably will grow with time.

NMR Spectroscopy

As just mentioned, the commonly used isotopes for NMR spectroscopy include mainly those of the lighter elements H, C, N, and O. However, there are many heavier nuclei with nuclear spin I=1/2 which are useful for this purpose. Accrete et al. 92 have demonstrated the utility of 183 W for studying the structure of heteropoly- and isopoly-tungstates via NMR. Although 183 W is the only NMR-active tungsten isotope, its NMR sensitivity relative to the proton is 7×10^{-5} ; thus, enriching the 183 W concentration beyond its natural 14.3% abundance will be important. NMR behavior for 25 Mg and 43 Ca is reviewed in Ref. 93, while 45 Sc, 89 Y, 139 La, 47,49 Ti, 51 V, 93 Nb, 181 Ta, 53 Cr, 95,97 Mo, 55 Mn, 99 Tc, 185,187 Re, 57 Fe, 187,189 Os, 59 Co, 103 Rh, 195 Pt, 63,65 Cu, 107,109 Ag, 67 Zn, 111,113 Cd, and last but not least, 199,201 Hg are reviewed in Ref. 94. In general, the amounts of material required are not excessive; a given researcher would probably need quantities on the order of 0.5-1 g per year.

ESR Spectroscopy

Another area where a wide range of isotopes can be utilized to good advantage is in the study of molecules via the technique of Electron Spin Resonance (ESR). Weltner and collaborators $^{95-97}$ have made several studies of the ESR spectra of inorganic molecules trapped in various solids, such as Ar or Kr at 40 K. In each case, an elucidation of the complicated ESR spectrum was accomplished through additional measurements of separated isotope versions of the same compound, e.g., SiN_2 was also observed as $^{29}\mathrm{Si}^{15}\mathrm{N}_2$, etc. From the ESR spectra it was determined that SiN_2 molecules in a pure N_2 matrix were bent, while the same molecules trapped in Ne were linear. The inference is that the molecular bending force is quite low, and that the constraints in the matrix sites are enough to induce bending of the molecule. As part of the studies reported in Refs. 95 and 96, optical spectra were also unrayeled via isotope substitution techniques.

Inelastic Scattering

Dagdigian et al. 98 have studied rotational inelastic scattering of 7 LiH molecules in collisions with HCl, DCl, and HCN molecules via a new technique which employs electric quadrupole state selection and laser fluorescence detection. Cross sections leading to various excited rotational states of 7 LiH were observed. It was found that the distribution of states is rather narrow and that the cross sections are large, reflecting the long-range coupling between the very polar molecules, such as 7 LiH, HCN, etc. Theoretical calculations show that the Born Approximation considerably overpredicts the experimental cross sections, but calculations using a Sudden-Approximation agree reasonably well with the data.

Mass Spectrometry

Mass spectrometric techniques utilizing stable isotopes are widespread in chemical research. These techniques are used in the study of molecular reaction mechanisms 99 and also in analytical work to measure concentrations of various species, both stable and radioactive. 100 Many analytical applications are handled with the technique of isotope Dilution Mass Spectrometry (IDMS), which is described in a review article by de Bièvre. 100 [It was through the use of IDMS on samples of 235 U that the discovery was made of a naturally-occurring nuclear reactor which existed at Oklo (Gabon) about 2 billion years ago. This reactor functioned for at least 100,000 years and used up perhaps 1000 kg of 235 U. 100] Because of the accuracy of IDMS techniques, the method can be utilized for half-life determinations if the half-life is in the range of tens of years or if the half-life is very long. 101

Of particular interest to chemistry is the ability of IDMS to provide very accurate atomic weights for the elements. All recent changes in the atomic weights adopted by the International Union of Pure and Applied Chemistry have come from isotope dilution mass spectrometry, and it is expected 100 that this will hold true in the future. The National Bureau of Standards (NBS) is currently pursuing measurements of this type. In order to obtain absolute isotope ratios, however, it is crucial to calibrate the mass

spectrometer over a wide range of isotope ratios (for each element studied) by means of very accurately known standard isotopic mixtures. 100 Creation of these standards by the NBS requires gram amounts of very highly enriched isotopes. These isotopes must also be sufficiently pure chemically that they can be used to prepare standard mixtures by gravimetric techniques. Without pure raw materials, NBS would be unable to create new standards (which are distributed worldwide) and would therefore be unable to improve the accuracy of atomic weight determinations. An example of the usefulness of such a measurement program can be found in Ref. 102, in which a new value for the Avogadro constant, accurate to 1 ppm, was deduced from very accurate measurements of the atomic weight of natural Si samples, along with careful density determinations and a precise measurement of the unit-cell volume in silicon. [For those who are curious, the result was N_{Δ} = 6.0220943x10²³ atoms/mole.] The IDMS technique is also an important component of chronology determinations in the geosciences. This will be discussed in Sec. II-F.

E. Other Physics

Besides nuclear and medium energy physics, there are two other branches of physics which rely on stable isotopes: atomic physics and solid state physics. Table V lists the isotope usage in the last 3 years for these research categories.

One major use of isotopes in atomic physics is for the study of the socalled isotope shift in optical spectra. As we will see below, new and powerful techniques based on tunable dye lasers are making a significant impact on our ability to measure these shifts, and hence nuclear properties, in regions far from the valley of beta stability.

In solid state physics, Mössbauer spectroscopy allows a very sensitive look at the properties of metals, insulators, superconducting compounds, etc. Such studies can also be used to elucidate the properties of chemical bonds and the effects of various ligands. Thus, applications of the Mössbauer technique are ubiquitous in nuclear physics, solid state physics, chemistry, metallurgy, geology, and biology. A closely related experimental technique is

that of perturbed angular correlations (PAC). This technique has much in common with Mössbauer spectroscopy, 103 since it can be used to probe some of the same effects. One difference between the two techniques, however, is that PAC studies require considerably smaller quantities of isotopes.

Atomic Physics

Laser Spectroscopy

In atomic physics the major impact of the "isotopic" nature of atomic nuclei is the so-called isotope shift, which involves a shift in the centroid of the hyperfine levels between isotopes of the same element. This shift arises in part from the mass change of the nucleus about which the electron rotates, 104 and in part from the change in the charge distribution of the nucleus across a series of isotopes. Especially in heavy nuclei, the former effect is rather small and, in any case, the interesting information about the nucleus comes from the latter effect. [In order to avoid problems with this isotope effect, the international unit of length was defined in terms of the wavelength of a particular atomic transition in $^{86}\mbox{Kr}$, rather than natural Kr.] These effects are also the ones being probed with muonic atoms, discussed in Sec. II-B.

In terms of isotope usage, the dominant atomic physics experiments are those involving laser spectroscopy of hyperfine structure. The present status of such work has been reviewed recently by Redi^{105} and by $\mathrm{Schuessler.}^{106}$ As discussed earlier in this paper, the study of isotope shifts can also be carried out with electronic or muonic x-rays. The main benefit of techniques involving optical spectroscopy is that unstable nuclei are also available for study, whereas the x-ray technique is restricted to stable nuclei. Good advantage is being taken of this by coupling laser spectroscopy techniques with existing on-line mass separators. $^{107-110}$ In this manner, isotope shifts have been determined for beta-unstable isotopes of Na, 107 Hq, 108 , 109 and 108 Cs. 110

A very nice experiment by Bemis et al. 111 at ORNL allowed a measurement of the isomer shift in $^{240\mathrm{m}}\mathrm{Am}$, a spontaneous fission isomer with a half-life of only 1 msec. The experiment produced the $^{240\mathrm{m}}\mathrm{Am}$

isomer via the 238 U(7 Li,5n) reaction at 47.5 MeV, thermalized the recoils in helium gas, and then polarized the atoms via the LINUP (Laser Induced NUclear Polarization) technique. Rather than use the light itself to detect the resonance condition, Bemis et al. monitored the rate of fission fragments. At the resonant absorption frequency the beam is optically pumped to the atomic state F=F $_{max}$, M=+F $_{max}$ and the aligned nuclei no longer fission isotropically but fission preferentially along the laser axis. Coincident fission detectors at 90° to this axis were used by Bemis et al. to record the decrease in fission-fragment rate when the laser was tuned to the resonant frequency.

Comparing the experimental shift for $^{240\text{m}}\text{Am}$ relative to ^{241}Am with that for ^{243}Am relative to ^{241}Am , Bemis et al. found that the change in $\langle r^2 \rangle$ for $^{240\text{m}}\text{Am}$, 4.6 fm², was 26.8 times larger than that for ^{243}Am and corresponds to a deformation parameter of β = 0.64, or a quadrupole moment of 30 b, compared with a ground state deformation of β = 0.24. Thus, the identification of the fission isomer as a strongly deformed shape isomer was directly confirmed for the first time.

Solid State Physics

Mössbauer Spectroscopy

Nuclear resonance absorption of gamma rays, referred to as the Mössbauer effect, makes it possible to probe in a very sensitive fashion the environment of an emitting (or absorbing) nucleus. In particular, such measurements give information on the isomer shift, the magnetic hyperfine splitting, and the electric quadrupole splitting. The first of these quantities, the isomer (or chemical) shift, measured in terms of the relative velocity of the source and absorber required to obtain resonance absorption, depends on the size differences between the nuclear ground and excited states and also on factors which modify the electron density within the nucleus. The magnetic hyperfine splitting, which gives rise to multiple resonance peaks due to the removal of the degeneracy between different hyperfine levels (for nuclear spin $I \ge 1/2$), depends on the effective magnetic field acting on the nucleus. This field arises from the motion of the

electrons cutside the nucleus as well as any externally applied fields. The electric quadrupole splitting also gives rise to multiple resonance peaks, due to the removal of the degenerary between different orientations of the nuclear quadrupole moment (I_1) with respect to the electric field gradient present at the nucleus. Given knowledge of the nuclear dipole and quadrupole moments, the observed Mössbauer spectrum can be compared with model calculations of the electronic structure of a Mössbauer nucleus in its medium in order to determine such details as oxidation states, position(s) in a crystal lattice, etc.

A great many nuclei can be used as Mössbauer sources; the main requirements are that the nucleus have a gamma-ray transition of reasonably low energy, say less than about 100 keV, and that the half-life of the Mössbauer gamma ray be on the order of 1 nsec. By far the most com only used source is the 14.4 keV transition in ⁵⁷Fe. fed from decay of ^{57m}Co (270 d). Other commonly used sources include (but are not limited to) 61Ni. 99_{Ru.} 181_{Ta.} 180,182,183,184,186_{W.} 186,188,189,190_{Os.} 191,193_{fr. and} 195_{Pt.} It is almost always preferable to make the precursor of the Mössbauer emitting nucleus from isotopically separated material in order to optimize the specific activity of the source and minimize resonance absorption in the source material. (Most of the production of these sources is done commercially and will not be discussed here.) In addition to the source production, it is often necessary to have an enriched quantity of absorber isotope. This is because the doping of the material to be studied can be difficult when using, e.g., the 2% abundance of ⁵⁷Fe in natural iron. Mass separators are sometimes used to dope the material to be studied. 113 Another method is to fuse the materials of interest together under vacuum. The typical amount of material needed for absorber work is about 0.1 g per researcher per year. Source strengths for, e.g., 57mCo, are in the range of 20-120 mCi (100 mCi is about 12 µg of 57mCo). Thus, elthough the sources are very important, the actual amounts of radioisotopes needed are rather small.

Applications of Mössbauer spectroscopy are quite widespread. A few examples from the recent literature will illustrate some of the uses. Kaizumi and Cathey 114 have used a $^{57m}\mathrm{Co}$ source to study the intermetallic

compound ${\rm Fe}_{0.5}{\rm Co}_{0.5}{\rm Tl}$ and its hydride phases. This compound is interesting because of its ability to absorb a tremendous quantity of hydrogen. It is a good choice for Mössbauer spectroscopy because it already contains the absorber, making it unnecessary to dope the material. By comparing Mössbauer measurements as a function of temperature and hydride content, Koizumi and Cathey were able to demonstrate that there is a phase change in the material as the hydrogen content is increased, and that the two phases coexist. In addition, their analysis suggests that the changes in the Mössbauer spectrum arise from the distortion and expansion of the metal lattice, and that the direct contribution of the H atoms is small.

As another example, Suib et al. 115 have studied the behavior of Eu ions as part of a europium-exchanged zeolite catalyst. Although several studies had already been made on such systems, there still existed several ambiguities, one of which had to to with the possible role of Eu $^{4+}$ in the zeolite. By monitoring the Mössbauer spectrum as a function of time, they were able to show that 95% of the Eu exists as Eu $^{3+}$ and that the addition of water to the zeolite gives only Eu $^{3+}$ and shows no evidence for Eu $^{4+}$.

Of interest to all 57 Fe Mössbauer work, Duff et al. 116 have obtained a new value for the quadrupole moment for 57 Fe which is only about half of the currently accepted value. They have calculated the electric field experienced by the 57 Fe in FeCl $_2$ and FeBr $_2$ via very sophisticated self-consistent-field Hartree-Fock calculations. Their value, coupled with the observed Mössbauer splitting, indicates Q=0.08 b rather than the 0.15 ρ value presently accepted. Should this be confirmed, it would necessitate reevaluation of many current Mössbauer experiments using 57 Fe.

Perturbed Angular Correlations

The study of perturbed angular correlations, like that of Mössbauer spectroscopy, offers opportunities to learn about the properties of matter in a wide variety of circumstances. [For a discussion comparing the two methods, see Ref. 117.] This technique can be used to measure g-factors of recoiling states, ³⁸ to probe the structure of crystals, ¹¹⁸ or to elucidate chemical bonding in molecules. ¹¹⁹ The "perturbation" used for this purpose is the interaction between the quadrupole moment (or magnetic moment) of

the emitting nucleus and the internal or external electric field gradient (or magnetic field) which it encounters.

Requirements for practical PAC emitters are that they have two gamma rays in cascade, and that the transitions be fed by a decay having a half-life of at least 1 hour. Examples of suitable nuclei include $^{111}\mathrm{Cd},^{181}\mathrm{Ta},^{204\mathrm{mpb}},^{181}\mathrm{Re},^{125,127}\mathrm{I},^{154,156}\mathrm{Gd},^{113}\mathrm{Sn},^{\text{etc}}.^{120}$ In general, the specific activity of the material should be high and competing products minimized; these factors argue for using enriched isotope precursors. As mentioned earlier, the amounts of material required for PAC studies are relatively modest compared with the amounts needed for Mössbauer spectroscopy absorbers. A 100 mCi radioactive source of $^{181}\mathrm{Hf},$ for example, is only about 6 $\mu\mathrm{g}$ of material.

One example of the use of the PAC technique was the experiment of Senba et al., 118 who studied the temperature dependence of the hyperfine field H_{hf} of various ions implanted in both the hexagonal close packed (hcp) and face centered cubic (fcc) phases of ferromagnetic Co by means of time differential perturbed angular correlations (TDPAC). The $^{100}\text{Mo}(^{16}\text{O}_{\circ}3\text{n})$ at 56 MeV was used to populate the 740 keV isomeric state of 113 Sn, which recoiled out of the 0.7 mg/cm² enriched 100 Mo target into the Co backing material. The beam was pulsed at a 1 MHz rate, and two NaI counters were used to observe delayed gamma rays at 450 to the beam axis during the beam-off periods. Senba et al. were able observe the exponential decay of the time spectrum modulated by the Larmor precession of the metastable state in the hyperfine field (polarized with a 1.7 kG external field). After correcting for the external field, the value of Hbf was measured and found to actually change sign at about 800° C. Through comparison with existing Mössbauer data, it could be concluded that the behavior of $H_{\mathbf{hf}}$ is completely independent of the method of implantation.

As another example, Ball and Kaplan 119 studied the interaction of the 181 Hf nucleus with its chemical environment via TDPAC. The technique promises to be an informative one, since chemically generated electric quadrupole interactions often reflect specific aspects of the molecular symmetry and electron distributions in chemical bonds. Using high specific activity 181 HfOCl $_2$ ($t_{1/2}$ =42.5 d) in dilute HCl, measurements were carried

out on several different compounds, HfO_2 , $HfO(H_2PO_4)_2\cdot H_2O$, and HfP_2O_7 , via the 133 keV - 482 keV cascade. (Because the recoil energy is very low and the half-life of the first gamma ray is 18 μ sec, it was expected that any "hot atom" effects would have subsided prior to the cascade.) The data were compared with predictions of H_{hf} based on simple bonding calculations, with fairly good agreement. Ball and Kaplan concluded that the bond structures of the two phosphorus compounds were very similar, and that the HfO_2 could not be in an anti-prismatic lattice arrangement because of the observed non-zero effective field gradient. It is apparent that TDPAC data can give information on chemical structure and bonding, and should allow rather detailed understanding of structural and symmetry questions.

F. Geosciences

Investigation of the isotopic abundance of various elements plays a significant role in geophysics and geochemistry. Possibly the best known reason is that isotope ratios provide a sensitive means of identifying the ages of geological objects. Another reason is that many chemical processes exhibit isotope effects 91 which modify physical properties (vapor pressure, melting point, etc.) and chemical properties (reaction rates). consequence, different isotopic ratios can develop in a sample, depending on its history. In addition, there is the interesting question of whether the isotopic abundance of elements on earth is the same elsewhere in the universe. The study of meteorites is particularly informative in this regard, as are the study of moon samples and data from the Viking mission to Mars. Lunar measurements, for example, show fairly large enrichments of the heavy isotopes of various light elements, e.g., 13C, 15N, 18O, 30Si, 34S, and 41K. The present $view^{121}$ is that these enrichments are related to preferential loss of the lighter isotopes caused by alternate vaporization-condensation cycles due to particle bombardment from the solar wind. Similarly, the enrichment of 15N in the Martian atmosphere is nearly twice what it is on earth; this is attributed to selective escape of 14N.

Table VI shows the use of separated isotopes in the geosciences during the last three years. Although a substantial number of different isotopes are

required, the amounts of each needed by a given research group are invariably quite small, 1-50 mg amounts being the norm. Essentially all of the isotopes are utilized for the same purpose, that is, as "spikes" in the IDMS method (see Sec. II-D). This technique, described in Ref. 100, is the standard method of doing accurate quantitative isotope mass spectrometry. In addition to the isotopes listed in Table VI, uses were reported for gaseous separated isotopes not covered here, including 17,18 O, 34 S, 36,38 Ar, Kr, and Xe. Here too only small amounts will probably be needed. A few illustrations of the uses for isotopes in geoscience research appear below.

Radiological Dating

Radiological dating is the technique used to determine the age of various geological objects. In order to obtain an absolute chronology, time measurements should be based on a process which has been occurring at the same rate throughout the earth's history. Radioactivity is the only known process which meets this requirement. The dating of very old objects requires a radioactive decay with a comparably long half-life. Fortunately, there are several isotopes which have half-lives of 10^9 - 10^{11} years (the age of the earth being about 6×10^9 years 122) and are therefore suitable for this purpose. [Assuming that such long half-life nuclides as 238 U(4.5×10 9 y), 232 Th(1.4×10 10 y), 147 Sm(1.1×10 11 y), 87 Rb(4.8×10 10 y), and 40 K(1.3×10 9 y) are not currently being produced in nature, this time scale provides a rough upper limit for the age of the universe.]

Dating methods based on most of these long-lived isotopes are being used at present. The methods based on $^{232}{\rm Th}$ and $^{238}{\rm U}$ decay are not always completely reliable because of the poorly defined isotopic ratios for Pb, which have to do with the fact that both radiogenic and non-radiogenic Pb exist in nature. In the recent literature, the two most generally used dating schemes involve $^{40}{\rm K}/^{40}{\rm Ar}$ and $^{87}{\rm Rb}/^{87}{\rm Sr}$. Both of these techniques obtain a date for an object by measuring, with a mass spectrometer, the ratio of atoms of the two species in it, and then connecting this to a time via the radioactive decay laws. 122 The $^{40}{\rm K}/^{40}{\rm Ar}$ technique requires $^{38}{\rm Ar}$ of very high purity to use as a "spike" to obtain the absolute $^{40}{\rm Ar}$ content via IDMS. (This material is apparently obtained entirely from foreign suppliers

at the present time.) The $^{67}\text{Rb}/^{87}\text{Sr}$ method of dating various mineral samples is illustrated in Refs. 123 and 124. In Ref. 124 the results for certain samples were cross-checked against the $^{40}\text{K}/^{40}\text{Ar}$ technique. In general the agreement was satisfactory, but it was suspected 124 that the ages obtained by both techniques (up to 500 million years) were too low in some cases, because the Sr had been leached from the samples and some of the Ar had escaped. Nonetheless, the $^{87}\text{Rb}/^{87}\text{Sr}$ technique appears in most cases to yield reliable chronological information.

Isotopic Anomalies

As mentioned earlier, one question of interest to geoscientists is whether the isotopic ratios found on earth are consistent with those found in extraterrestrial materials. Wasserburg and coworkers at the California Institute of Technology have studied this question for various isotopes of uranium, 125 titanium, 126 and silver 127 in meteorite samples. No anomalies were found in the $^{235/238}$ U ratio, although some had been reported earlier. The conclusion was that the earlier samples were contaminated with 235U from a spike solution. (Chen and Wasserburg 125 avoided this problem by making up a 233 U/ 236 U double spike instead.) In the case of the titanium isotopes, however, significant anomalies were found, especially in the very neutron excess isotope 50 Ti (also 48 Ca). Based on all of their data, the researchers at Cal Tech concluded that the isotopic ratios were probably explainable only by assuming that several different nucleosynthesis processes were involved in the production of these nuclides. Finally, analysis of the Santa Clara and Piñon meteorites has shown an anomalous 107Aq/109Aq ratio, ranging from 1.7 to 2.8, compared with a normal value of 1.09. The data are consistent with the "extra" 107Ag arising from decay of 107Pd, but the possibility of an intense local cosmic ray irradiation production mechanism cannot be excluded.

Table 1

Usage of Electromagnetically Separated Stable Isotopes and Derived Radioisotopes in Nuclear Physics/Chemistry. The amounts shown represent usage (in mg) for the 3-year period, 1979-1981.

Н		(a)	Ca		Ge	
He ∟i	,	(a)	40	74,000	70	1000
L	,	7 400 000	42	1300	72	800
	6 7	7,400,000	43	300	74	700
_		297,000	44	73, 000	76	100
Вe		4.)	48	22, 000	As	
_	9	(b)	Sc		75	(b)
В	10	F 200 000	45	(b)	Se	
	10	5,300,000	Ti		76	5500
_	11	176,000	46	1000	78	450
С		1/00	48	300	80	450
	12	1600	50	800	82	41,000
	13	189,000	V		Br	
7		(a)	51	200	Kr	(a)
O F		(a)	Cr		Rь	
۲		4.3	50	700	Sr	
Ne	19	(b)	52	60,000	84	100
		(a)	53	60,000	86	3100
Na	0.7	<i>(</i> -1	Mn		87	110,000
	23	(b)	55	(b)	88	51,000
Mg		7400	Fe		Υ	4.3
	24	3600	54	228,000	_ 89	(b)
	25	160	56	1 7, 00 0, 000	Zr	750
	26	41,000	57	1100	90	750
ΑI		6.3	58	600	91	350
٠.	27	(b)	Co		92 94	700
Si		7000	59	(b)		700
	28	3000	Ŋi		96	700
	29	155,000	58	330,000	Nb	4.5
_	30	56 , 000	60	182,000	93	(b)
Р		4.5	62	41,000	Mo	
_	31	(b)	64	43,000	92	2906
5	70	400	Cu _		94	600
	32	600	63	175,200	95	200
	34	1100	65	175,000	96	450
-	36	100	Zπ		98	1200
Cl		222	64	1300	_ 100	1700
	35	200	66	500	Tc	(c)
	37	100	67	200		
Ar K		(a)	68	800		
	40	700	70	110		
	40	300	Ga			
	41	2				

Table 1, cont.

Ru		Cs		Ho	
96	550	133	(ь)	165	(b)
98	100	Ba		Er	
99	100	134	10,000	162	250
100 101	350 100	135	200	164	200
101	100	136	200	166 167	40 60 0 0
102	300	138	150	168	52,000
Rh	200	La Ce		170	250
103	(ь)	136	10,000	Tm	270
Pď		140	400,000	169	(b)
102	500	142	100,000	Υb	
104	75	Pr	•	171	190,000
105	20	141	(b)	173	300
106	75	Nd		174	1000
108	600	142	120,000	176	500
110	350	143	50,000	Lu	
Ag	350	144	600	176	300
107 109	300	145	51,000	⊢f	
Cq	200	146 148	80 110	176 380	100 650
106	2000	150	74,000	80 Ta	620
110	10,000	Pm 170	(c)	180	160
111	10,000	Sm	(2)	w	100
114	200	144	2000	180	550
In		147	800	182	51,000
5n		148	1200	Re	•
112	1400	149	700	Os	
116	31,000	150	450	184	10
118	32 , 000	152	2100	186	3300
119	200	_ 154	3800	187	3000
120	210,000	Eu ,,,	650	188	50,000
122 124	32,000 33,000	151 153	650 150	189	50,000
126	250	Gd	170	190 192	52,000
Sb	270	152	8500	Ir	52,000
Te		153	35,000	 191	2300
120	100	155	21,000	193	2300
122	350	156	212,000	Pt	-5
124	300	157	130,000	192	15,000
125	6000	158	1200	194	2000
126	30¢	160	2000	195	2000
128	300	ть		196	500
130	6000	159	(P)	198	2800
I 127	(b)	Dy	50.000	Au	
Xe 127	(a)	162 163	59,000 50,000	197	(ь)
Ve.	(4)	164.	51 , 000		
		104.	719000		

Table 1, cont.

Hg		Nρ		At	(c)
196	150	237	100,000	Rn	(c)
200	2000	Pu	100,000	Fr	(c)
204	650	239	140,000	Ra	(e)
205	1000	240	51,000	Ac	(c)
Tl		244	3500	Th	(-,
Pb		Am	7300	232	360,000
204	8500	Cm		Pa	,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,
206	41,000	248	1	່ເງື	
207	100	Bk	1	233	100,000
208	550,000	249	1	234	16,000
Bi		Cf	•	235	940,000
209	(b)	249	1	236	30,000
Po	(c)	447	1		
P0	(C)			238	710,000

Notes: (a) Stable isotopes used in research, but not separated electromagnetically.
 (b) Mono-isotopic in nature.
 (c) No stable isotopes exist.

Table II

Usage of Electromagnetically Separated Stable Isotopes and Derived Radioisotopes in Medium Energy Physics. (The amounts shown represent usage (in mg) for the 3-year period, 1979-1981.

Li			Fe		Sn	
	6	810,000	54	7300	112	7000
	7	810,000	56	1300	116	7000
В		ŕ	Ni		118	70,000
	10	60,000	58	210,000	120	37,000
	11	60,000	60	7500	122	6000
С		•	62	6000	124	12,000
	12	10,000	64	6000	Te	12,000
	13	320,000	Cu		125	20,000
	14	50,000	63	500	Sm	-0,000
Mo	1	·	Zn		144	500
_	24	40,000	64	2	148	500
	25	40,000	Ge	-	150	20,000
	26	1.42,000	72	6000	152	500
Si		· _,	76	500	154	6000
-	28	20	Se	200	Gd	0000
	29	1000	76	500	155	21,090
	30	22,000	Sr	700	Dy	21,000
5		,	87	2000	161	18,000
•	32	500	88	2000	Er	10,000
	34	6000	Z r	2000	166	6000
Ca			90	84,000	Yb	8000
	40	20,000	Mo	0-,000	176	6000
	42	180,000	92	40,000	Hf 170	0000
	44	26,000	Ru ´²	40,000	177	25,000
	48	29,000	100	5000	Ta Ta	27,000
Τi	40	2,,000	104	5000	181	10
	48	200,000	Pd	7000	PP 101	10
	50	50,000	110	10,000	204	3000
ν	-0	20,000	Cd	10,000	204	3000
•	51	10	110	50,000	207	170,000
Сг	71	10	111	50,000	208	115,000
CI	50	300	112	50,000	U 208	117,000
	52	41,000	112	000,000	238	2
	54	300			230	2
	/-	200			•	

Table III

Usage of Electromagnetically Separated Stable Isotopes and Derived Radioisotopes in Radiochemistry. The amounts shown represent usage (in mg) for the 3-year period, 1979-1981.

Li		Sn		⊢lf	
6	223,000	116	2000	172	2
7	1,000,000	118	50	176	1.0
8		120	2000	177	10
10	5000	124	200	179	20
Ca		Te		180	10
48	4000	130	500	W	
Ti		Ce		180	2
48	2000	140	300	Ir	
Fe		Nd		191	5
57	500	146	200	192	5 5
58	50	148	200	Pt	
Cu		150	1	198	15
63	376	Sm		Рb	
6 5	1920	144	700	208	5900
Zn		148	200	Th	
68	20	149	200	230	10
Mo		150	250	IJ	
92	500	154	400	233	200
9 6	500	Eu		235	75
100	700	151	200	238	50
Ru		153	200	Pu	
96	10	Yb		239	5
Ag		170	100	242	1 5
109	50	176	300	244	5
Cd		Lu			
106	10	176	300		
108	10,000				

Table IV

Usage of Electromagnetically Separated Stable Isotopes and Derived Radioisotopes in Other Chemistry. The amounts shown represent usage (in mg) for the 3-year period, 1979-1981.

Li 6 7	10 , 000 10 , 000	Sr 87 Mo	500
B 10 11	5500 500	95 Nd 142	300 50
C 13 Si	61,000	144 146 148	50 50 50
29 Ti 47	5100	Dy 164	100
C r 50	400 1000	Er 167 170	100 200
53 Fe 56	1200 8100	ҮЬ 171 W	200
57 Co 57	1600 1	183 Hg	4200
Cu 63 65	200 1500	200	100

Table V

Usage of Electromagnetically Separated Stable Isotopes and Derived Radioisotopes in Other Physics. The amounts shown represent usage (in mg) for the 3-year period, 1979-1981

L i		Sb	
6	180,000	123	100
7	380 , 000	125	100
В		Te	
11	10,000	126	200
C		Sm	
13	2200	154	200
Mg		Eu	
24	100	151	10
Fe		Gd	
56	1000	160	400
57	2800	Dy	
Co		160	200
60	700	Er	
Ni		168	200
58	100	Yb	
- 60	50	172	1500
Zn		W	
67	10	180	10
Se	***	Hg	100
77	500	198 199	100
Mo	70		10
92 94	70 70	200 201	200
94 95	70 70	201	100
	70 70	202	100
96 97	70 70	71 Tl	6
98	70 70	203	500
100	70 70	Pb	300
Te	70	207	300
99	3000	U	200
Ag	0000	235	1000
Ay 107	50	Am	1000
109	1000	241	5000
Cq	1000	243	2000
114	500	Cm	2000
Sn	700	244	500
118	260	277	200
119	1000		
11,	1000		

Table V:

Usage of Electromagnetically Separated Stable Isotopes and Derived Radioisotopes in Geosciences. The amounts shown represent usage (in mg) for the 3-year period, 1979-1981.

Li		Eu		Sr	
ϵ		151	41		84 750
7	' 30	153	1		86 260
Ŗ		Gd		Zr	
	.0 30	155	2		91 30
1	.1 30	157	10		96 30
Mg		158	30	Pd .	
	4 30	160	30		102 30
2	5 31	Ðy			108 30
	6 1	160	30		110 30
Si		161	2	Ag	
2	9 750	Er			107 30
3	0 750	164	30		109 30
K		167	2	-Dd	
4	0 210	ΥЬ			106 30
4	1 800	170	30		108 1
Ca		171	2		111 1
4	2 500	176	30	:	113 1
4	3 100	Lu		Sn.	_
4		176	41		118 1
4	8 100	Hf			120 1
Ti		179	40	Ba	
5	0 30	TI TI			134 30
V		203	31		35 1
. 5	0 30	205	1		36 400
Cr		Pb	-		37 30
51	0 30	204	31	La	
5		205	130		.38 41
5		206	250	Ce	.,,
Fe	, ,,	207	30		.38 30
5.	4 30	208	450		.42 42
5.		Th 200	420	Nd	76 72
51		230	31		45 43
Ni	, ,	U	71		48 30
62	2 30	233	30		49 1
64		234	30		50 70
Zn	, ,	235	5	Sm	20 /5
64	4 1				44 30
66		236	30		47 60
67					49 15
70					50 30
Rb /	, ,1			1	/0 /0
170					

III. STABLE ISOTOPE SUPPLY PROBLEMS

As part of the survey (see Appendix A), respondents were asked to indicate whether they had experienced any problems or significant delays in obtaining isotopes. Some 26% of the respondents in all categories answered in the affirmative. These problems should not be taken as a reflection on the ORNL personnel working in the Stable Isotope Sales area, who were specifically praised for their cooperation. The breakdown of the responses by research category is shown in Table VII; there is some tendency for the major isotope users to have a larger percentage of the difficulties.

Reported problems can be grouped into four main areas: unavailability of isotopes, marginal enrichment or chemical purity, delays in obtaining isotopes, and high cost. Disturbingly, nearly half of the respondents reporting a problem (12% of all respondents) found certain isotopes unavailable. Particular isotopes that were specified as being unavailable or having too low enrichment included $^{26}\mathrm{Mg}$, $^{29,30}\mathrm{Si}$, $^{50}\mathrm{Ti}$, $^{67}\mathrm{Zn}$, $^{77}\mathrm{Se}$, $^{84}\mathrm{Sr}$, $^{96}\mathrm{Ru}$, $^{110}\mathrm{pd}$, $^{113}\mathrm{Cd}$, $^{112,118,124}\mathrm{Sn}$, $^{134}\mathrm{Ba}$, $^{138}\mathrm{La}$, $^{150}\mathrm{Nd}$, $^{154,160}\mathrm{Gd}$, $^{176}\mathrm{Lu}$, $^{180}\mathrm{W}$, $^{189,190}\mathrm{Os}$, $^{191}\mathrm{Ir}$, $^{198}\mathrm{pt}$, $^{201}\mathrm{Hg}$, and $^{233}\mathrm{U}$.

The lack of sufficiently enriched materials is becoming a particularly serious problem. Although many isotopes are technically "in stock," they are of marginal utility to the research community because of low enrichment or unacceptable chemical contaminants. A notable example of insufficient enrichment is ⁸⁴Sr, which is heavily used in geochronometry. Until recently, the National Bureau of Standards provided this isotope as a high-purity Standard Reference Material (SRM-987 to its friends), but it is no longer available. In practice, replacing material of >95% isotopic purity with material from ORNL having only 82% purity introduces significant uncertainties into the IDMS technique discussed in Secs. II-D and II-F. For

most, if not all, of the fields considered in this paper, there is a distinct tendency to require the highest isotopic purity of the most rare isotopes -both neutron-poor and neutron-rich -- since these frequently lead to the most unusual products in a nuclear reaction or provide the least potential interference in mass spectrometry measurements. As a consequence of the lack of highly enriched isotopes, several groups have had to resort to the tactic of purchasing a low-enrichment isotope and having it further enriched on a colleague's isotope separator, or, in the case of $^{38}\mathrm{Ar}$, purchasing all of their supplies of >99% enriched material from a foreign (Swiss) supplier. This is both inefficient and time consuming, and is clearly impractical as a long-term solution to the basic difficulty.

A number of respondents reported a complete lack of availability of certain separated isotopes. Nearly 5% of the researchers indicated that they had abandoned at least one planned experiment in the past three years due to inability to procure the requisite material. Although this number is not (presently) overwhelming, one cannot help but make an analogy to the failure of a string of resistors: each time one fails, the load on the rest increases until the next weakest one goes, and so on.

Although these materials were only out of stock temporarily, rather than forever, there is often not much difference as far as basic research is concerned. In some fields of research, e.g., high energy physics, a particular experiment may take 5 years or more from conception to completion, but for most of the fields considered in this document the time scale is much shorter. Thus, the unavailability of a particular enriched isotope for a period of several years very likely means that the experiment is, to all intents and purposes, dead. [At many major nuclear physics facilities a "scheduling cycle" for a series of approved experiments is 4-6 months: experiments not completed within one year from the date of the original proposal are considered sufficiently out of date that they are automatically "removed from the books."] Furthermore, in nuclear physics and nuclear chemistry, which dominate the research usage of enriched isotopes, there is a tendency to exhibit "resonance-like" purchasing patterns, A newly discovered phenomenon such as backbending caused a resonance in rare earth purchases, while the ALAS phenomenon in ¹⁶O + ²⁸Si scattering generated a resonance

in Mg and Si purchases (for both of which the heavier isotopes are now out of stock). It would seem best to deal with such fluctuations by having enough material on hand to average them out over a period of several years. As of now this is obviously not happening, since in July 1981 there were 65 isotopes reported to be out of stock.

Fortunately, much of the research with large requirements for enriched isotopes --neutron, photonuclear, pion, electron, and double beta decay experiments-- can do nicely with target materials loaned from the Research Isotopes Pool at ORNL. The results of the survey indicate that this part of the system is functioning reasonably well. Occasional delays occur when an isotope is signed out, but it seems that the scientists concerned are generally able to negotiate directly to arrive at a mutually satisfactory solution. Since the number of respondents worried about the availability of pool isotopes is roughly balanced by those who already have isotopes and are worried about how long they can keep them, the system seems to be in equilibrium. Of course, these isotopic targets are fairly rugged and can presumably be used many times without incident. [Targets for the majority of nuclear physics experiments, however, tend to be thinner and more fragile, and are frequently destroyed by interactions before, during, or after the experiment, e.g., interactions with fingers, screwdrivers, or occasionally malevolent vacuum systems. Insofar as these items are a routine part of most experimental setups, this problem will remain with us.]

With respect to costs, the one issue raised repeatedly was the astronomical cost of $^{48}\mathrm{Ca}$. Because of its large N/Z ratio, this isotope is prized in many experiments, ranging from the search for superheavy elements to studies of nuclear matter radii with pions. For many projects a comparison of the behavior of $^{40}\mathrm{Ca}$ and $^{48}\mathrm{Ca}$ is the most straightforward way to elucidate nuclear structure effects, but the high cost of $^{48}\mathrm{Ca}$ has begun to price the scientific community out of the market. Other statements regarding isotope costs must be "normalized" to some extent. At one extreme there were comments about the expense of having to order a "special calutron run" to produce a needed isotope, while at the other extreme one user commented that his "entire research budget would not cover the interest on the loan" to acquire a parameter isotope. Nonetheless,

the problem of excessive and rapidly rising costs for some isotopes is real and must be dealt with somehow. If special calutron runs were to become the rule, rather than the exception, the entire community of isotope users would clearly suffer.

As mentioned above, some researchers have been forced to abandon certain experiments due to the lack of suitable materials. Others have been able to solve their problems, at least temporarily, by borrowing targets or enriched materials from colleagues at other institutions. Although one researcher tried "ordering well in advance," within the limited statistics of the present survey we find that this approach is not in wide use. A majority of respondents appears to have adopted the "begging and waiting" approach. One isotope user claimed to have solved his problem by "developing patience," but it seems unlikely that this will ever become a general solution for the research community.

<u>Table VII</u>

Reported Problems with Electromagnetically Separated Isotope Supplies

Category	Unavailable ^{a)} (%)	Enrichment a) (%)	Delays ^{a)} (%)	Cost ^{a)} (%)	Total ^{a,b)} (%)
Nuclear Physics	13	6	20	4	43 (14)
Medium Energy Physics	23	4	-	-	27 (2)
Radiochemistry	16	11	5	5	37 (2)
Other Chemistry	4	2	4	4	14 (3)
Other Physics	4	6	1	1	12 (2)
Geosciences	16	4	-	9	29 <u>(3)</u>
Totals	(12)	(4)	(6)	(4)	(26)

a) Ench number refers to the percentage of the total responses in that category only - values in different categories are not directly comparable.

b) Numbers in parentheses are the percentage of total responses from all six categories.

IV. TRENDS IN FUTURE SEPARATED ISOTOPE REQUIREMENTS

In this section, we will look at how the various uses of stable isotopes are expected to change in the next 5 years, and at how such changes will affect the demand for separated stable isotopes. In a similar survey 128 on stable isotopes which was carried out by a National Research Council panel in 1968, it was stated that "since nuclear structure, spectroscopy, and reaction studies are concerned with the systematics of all the isotopes of an element, separated samples of nearly all isotopes have been and continue to be required." The present investigation (see Tables I-III) shows that this is still the case. Of the elements between hydrogen and bismuth which have more than one isotope, all but eight were utilized for nuclear science experiments in the past three years. (If one includes the other research areas covered here, for which isotope utilization is shown in Tables IV-VI. only separated isotopes of Ga, Br, In, and Re were not used during this period.) The overall amounts used, along with their approximate costs, are summarized in Table VIII. (Some attempt was made to estimate and exclude the non-destructive uses in determining these costs.)

Because of the manner in which basic research is carried out, it is at best difficult to make predictions of future needs in a quantitative fashion. For this reason, no attempt will be made here to provide detailed numerical estimates for each isotope. Rather, we will look at the trends in scientific programs and comment on areas where significant changes in isotope usage are likely. One of the topics covered in the survey was expected future needs, especially where they might differ from present needs. The survey showed a surprisingly uniform attitude in all six research areas. There was a clear consensus that research needs for stable isotopes would be approximately constant during the next 5-year period. (Even those who felt

that changes would occur indicated their such changes would be "slight" or "moderate.") This does not mean, of course, that the research itself will be the same, since it is clear that there will be changes in the mix of such isotopes and the experiments for which they will be used.

First, let us consider the field of nuclear physics/chemistry, which has historically been (and is likely to continue to be) the largest user of stable isotopes for basic research purposes. In the next 5 years, it seems probable that nuclear physics will continue an evolution that, to some extent, parallels that of high energy physics. ifistorically, nuclear physics has been carried out at a large number of facilities, mainly situated on university campuses, and has involved a correspondingly large number of independent research groups, each with its own professor, perhaps a few postdoctoral scientists. and several areduate students. More recently, however, the trend has been toward fewer but larger facilities (in terms of beam energy and availability of experimental equipment). Thus, nuclear physics has entered the "user" phase of its evolution. Much can be said about whether this is good or bad. but it is nonetheless a reality. This has several effects on the way nuclear physics is done. One of the most obvious is the shrinkage in the amount of beam time allotted to each research group. Because many of the smaller facilities are being closed, researchers now find it increasingly necessary to formally submit proposals for beam time, have them approved, and then schedule time on an accelerator which may be hundreds, or even thousands, of miles away from their home laboratory. In practice, this tends to mean that a group will perform fewer experiments per year, although an individual experiment may be longer than was the case previously. Thus, a research group uses fewer targets per year, but performs more sophisticated measurements on each. It is also true that most major facilities (e.q., the Holifield Heavy Ion Research Facility at ORNL, the Brookhaven Tandem Van de Graaff Laboratory, the LBL SuperHILAC) are considerably oversubscribed,

Several patterns are evident in experimental nuclear physics programs. One is a general trend toward heavier projectiles and higher beam energies. This will have an effect on allowable target thicknesses such that more material per target will become the rule. [Of course, this statement refers mainly to the relatively thin targets used in heavy ion experiments; for

neutron, electron, or pion work, a restriction on target thickness would not be coupled to the beam energy.] In addition, there has been considerably more emphasis on investigating rare processes which, because of their low cross sections, require thicker targets to achieve acceptable counting rates. With regard to choice of targets (which have always made up the bulk of the separated isotope usage for nuclear physics), the clear consensus in the research community is that it is impossible to predict exactly which isotopes will be needed in the future. Nonetheless, some general points did emerge from the survey.

The rare earth isotopes will remain in high demand, since they have regularly provided us with the most exciting and surprising information on nuclear structure. Other materials which seem likely to keep their high appeal are those with a wide range of available isotopes, such as Ca. Ni. Zr. Mo. Sn. Sm. and Pb. These isotopes allow the exploration of changes in nuclear structure and reaction mechanisms due to changes in N/Z ratio. distance from closed shells, and deformation. There is also likely to be a considerable demand for isotopes in the Mq-Si region. This mass region has shown evidence for interesting but as yet poorly understood structures which will undoubtedly be investigated in more detail. We have already noted, however, that many of these isotopes are now out of stock and may not be available for several years - a clearly unfortunate situation! In general terms, it is probably safe to say that targets of isotopes on the extremes of the mass distribution for any element (i.e., both neutron-rich and neutronpoor species) will be in demand, since these offer the best possibilities for producing nuclei far from beta stability and studying their properties. At UNISOR, for example, there is a need for the lightest isotopes of all refractory elements for producing neutron-deficient nuclei. Finally, it can be predicted that the demands for radioactive targets will continue to expand. In general, the amounts needed for the above targets will be similar to current needs, i.e., about 500-1000 mg per isotope for thick targets or about 50-100 mg per isotope for very thin targets.

As in the past, a crucial aspect of the targets will be their enrichment. In most cases the addition or removal of even a single neutron can have a significant effect on a nuclear reaction. Moreover, the problem of

competing reactions with an isotopically mixed target leads to background processes which almost invariably obscure the reaction of interest (or at least make its analysis complicated and ambiguous). Unfortunately, this tends to be especially true for the isotopes on the extremes of the mass distribution, e.g., ^{48}Ca or ^{112}Sn , whose natural abundance is low. Thus, the isotopes of the lowest natural abundance are just the ones for which very high isotopic purity (>95%, and preferably >99%) is most crucial. The lack of high purity rare isotopes is a weakness in the present supply system that must be corrected.

Another clear trend in heavy ion experimental programs is that new accelerators just coming on-line will all have the capability of accelerating relatively heavy beams to energies well above the Coulomb barrier on most targets, (The SuperHILAC, and Michigan State University Phase II when completed, will do so with any mass ion, while HHIRF and Atlas will do so for ion masses up to about 160 and 130, respectively.) This will stimulate the use of stable isotopes as exotic projectiles, a use that has only recently begun to be exploited. It is clear that the most interesting choice for a projectile is ⁴⁸Ca. followed by several other neutron-rich ions, such as ²⁶Ma. 30_{Si.} 36_{S.} 50_{Ti.} 58_{Fe. and} 64_{Ni.} These ions allow studies of nuclei far from the valley of beta stability, in relatively unknown territory as far as nuclear structure goes. The use of stable isotopes for beams implies that the quantities required will increase significantly. It is likely that 1-5 g amounts of many of the isotopes just listed will be needed at various accelerators. Based on the experience at the SuperHILAC, which for many years has been accelerating 48Ca beams as well as enriched beams of 86Kr and 136Xe, it is possible to recover a substantial portion of the rare isotope if proper care is taken in the design of the ion source and its attendant vacuum system. (Approximately 80% of the ⁴⁸Ca isotope in the source is recoverable at the SuperHILAC.) In addition, it should be recognized that the requirement for isotopic purity is much less severe when the material is used to provide a beam than when it is used as a target. This is because most accelerators have reasonably good mass separation properties, thereby allowing only the isotope of interest to be accelerated. In fact, once the enrichment gets much beyond 50% the law of diminishing returns comes into play, in the

sense that a further increase in enrichment to 99% improves the beam intensity by only a factor of two. The 70-80% enrichments provided by ORNL for some rare isotopes are ideal from this point of view.

Separated isotopes, particularly ^{6,7}Li, ¹⁰B, and ⁵⁶Fe, are also expected to be required in large quantities for the fabrication of special shielding, collimators, and filters for neutron experiments. In these applications, kilogram amounts are often utilized. Also emphasized in neutron work will be studies of structural materials, such as Cr, Fe, and Ni (for which 100 g samples will be employed), and studies of stable fission products and actinide nuclei, such as ²³⁹⁻²⁴⁴Pu (which will utilize about 1 mole of each nuclide in metallic form). An expected increase in resonance-averaged neutron capture studies (relative to thermal neutron work) will correspond to greater needs for 10-50 g samples. Here too, there will be a tendency to shift the experimental emphasis to species of lower abundance.

In medium energy physics, the demand for rare isotope targets will continue to grow. Studies of exotic reactions on isotopes at the extremes of stability, for example with (π^+,π^-) , (π^-,π^+) , and (π^-,p) reactions, will require 100 g quantities of material. For electron scattering and pion/muon work, radioactive targets such as ^{41}Ca and ^{205}Pb will be of interest. It should be mentioned here that facilities such as the Bates Linear Accelerator and LAMPF generally have a substantial library of targets on hand. Because of the large mass and "structural stability" of targets used at these accelerators, they can be shared by many groups. Stockpiles also exist in the nuclear physics community, most notably at some of the national laboratories and larger university facilities. The difference in this case, however, is that the amounts of material available tend to be rather small, say 50-100 mg quantities of materials scattered throughout the periodic table, and do not form much of a hedge against supply shortages at ORNL.

Radiochemistry research will require substantial amounts of ⁴⁸Ca (at least 10 g) in the future for use as a projectile in the search for superheavy elements. Also needed for this purpose are actinide and transactinide target materials, but due to their highly radioactive nature the amounts required (<1 mg) will not be large in an absolute sense. Clearly, however, the discovery of positive evidence for the existence of SHE's could be expected

to increase demand for the above nuclides. Another area where stable isotopes will continue to be in demand is in the production of various radioactivity standards for the NBS; particular isotopes mentioned in this regard are $^{104}\mathrm{Ru}$ and $^{107}\mathrm{Ag}.$ NBS provides Standard Reference Materials of more than 60 radionuclides at present, and the list will no doubt continue to expand.

Utilization of separated isotopes in chemistry includes experiments with NMR, ESR, and mass spectrometric techniques. It is likely that the need for electromagnetically separated isotopes in NMR work will increase in the next several years, particularly for ²⁹Si, ⁴³Ca, ⁵⁷Fe, ⁶¹Ni, ⁶³Cu, ⁶⁷Zn, ⁷⁷Se, 95 Mo. 99 Ru, 113 Cd, 123 Te, 183 W, and 187 Os. These isotopes will be used in amounts of about 0.1-1.0 g at many NMR facilities, which are routinely involved in studies of newly synthesized compounds, heterogeneous catalysts, organo-iron compounds, etc. Although the sensitivity of NMR devices for natural abundance materials has increased substantially, the range of experimental activities has more than kept pace. Thus, many important experiments will continue to require the enhanced signals from isotopically enriched samples. For ESR measurements, 63Cu will be in demand for analysis of protein structures; in addition, various complexes will be investigated with isotopes of ⁴⁷Ti, ⁵³Cr, ⁹⁵Mo, and ¹⁸³W, in amounts on the order of 100 mg each. (Much of this work is aimed at biomedical questions and will be covered in detail elsewhere.)

Mass spectrometric experiments will continue to require very high purity (>99%) "spike" solutions for quantitative isotopic analyses. For example, analysis of nuclear fuel elements will require $^{236}{\rm Np}$ for measurements of $^{237}{\rm Np},~^{233}{\rm U}$ for measurements of $^{235}{\rm U}$, and $^{97}{\rm Tc}$ (produced from enriched $^{96}{\rm Ru}$) for determination of $^{99}{\rm Tc}$. A wide variety of isotopes will also be needed to monitor fission products. In general, the needs for mass spectrometric analysis in geological and environmental work will also continue to grow, and accompanying this growth will be new requirements for isotopically pure materials. The quantities involved in such work are small, however; only microgram quantities are needed for any particular analysis, and 50 mg of material can last for several years.

Isotope usage in solid state and atomic physics does not appear to be

expanding substantially at present. In Mtssbauer spectroscopy the primary source will continue to be $^{57}\mathrm{Co}$ (produced from $^{57}\mathrm{Fe}$), although $^{119}\mathrm{mSn}$ (produced from enriched $^{180}\mathrm{sn}$), $^{181}\mathrm{Ta}$ (produced from enriched $^{180}\mathrm{w}$), and several other sources will undoubtedly be used. The magnitude of present needs is indicated in Table V. One area where some expansion may occur is in neutron scattering studies of condensed matter. Appropriate substitution of isotopes can change the scattering lengths and make it possible to separate the contributions to the neutron scattering from different components of the material. Isotopes which may be needed here include $^{10}\mathrm{B}$, along with various Ni and Se isotopes in quantities of 10-100 grams.

Laser spectroscopic investigations of isotope shifts benefit most from the study of a range of isotopes. Thus, it is likely that such candidates as Zr and Mo isotopes will be subjects for this type of study. Other candidates for future studies include 107,109 Ag, 113 In, 198 Hg, 203 Tl, and 204,207 Pb, in amounts on the order of tens of milligrams each.

The primary use for electromagnetically separated isotopes in the geosciences will be for the IDMS technique discussed earlier. (Other needs for isotopes will arise in the study of various fractionation processes, but such studies tend to utilize mainly the lighter elements such as oxygen, nitrogen, and sulfur, which do not come from ORNL.) Specific future needs will probably include standards of $^{149}\mathrm{Sm}$ and $^{145}\mathrm{Nd}$ for the Sm/Nd dating method, $^{205}\mathrm{Pb}$ for the U/Pb method, and $^{175}\mathrm{Lu}$ and $^{174,177}\mathrm{Hf}$ for the Lu/Hf method. Once again, the technique of mass spectrometry does not require large amounts of material, but isotopic purity can be dominant in determining the absolute error of a measurement.

Although it is not a primary concern of this report, the future costs of separated isotopes will obviously be relevant to how extensively they are used. The price of ⁴⁸Ca was discussed in Sec. III as an example of how a very useful isotope is becoming virtually unusable because of its expense. In spite of tightening research budgets, however, respondents to the survey clearly indicated that, given a choice between paying much higher prices for separated isotopes or not being able to get them at all, they would prefer to pay more. This is not surprising, since virtually <u>all</u> respondents indicated that their research would come to a complete halt without access to stable

isotopes.

One other issue relevant to the future need for stable isotopes is the question of who will be using them. What is somewhat worrisome, at least as regards the nuclear physics community, is the downward trend in the number of both senior scientists and graduate students. This trend, which was commented on by a number of respondents, has been confirmed by the Nuclear Science Advisory Committee (NSAC) Subcommittee on Manpower, which has just completed its 1980 census of basic nuclear scientists in the United States. The subcommittee report 129 offers the following conclusions:

- There has been an overall decline of approximately 10% in the number of basic nuclear scientists in the two years between 1978 and 1980;
- The postdoctoral population has stayed essentially constant during this period; and
- 3) The graduate student population has decreased by at least 16% during this period, with the ratio of students to faculty members becoming about 1:1.

This trend is already having a negative impact on the usage of isotopes, and, if left unchecked, will have an even bigger impact in the future.

<u>Table VIII</u> Summary of Isotope Usage (1979-1981)

Table	No. of Species	Gram-Atoms	Gram-Atoms ^{a)}	Approx. Cost ^{b)}
1	187	2213 (1824) ^{b)}	408 (18) ^{b)}	\$ 730K
11	69	323 (251) ^{b)}	67 (1) ^{b)}	50K
111	53	181	0.4	400K
IV	26	8.8	5.2	35K
٧	47	86	1.3	270K
VI	_87	0.15	0.1	90K
	220	2812 (2351) ^{b)} [937 (784) ^{b)}] ^{c)}	482 (26) ^{b)} [161 (9) ^{b)}] ^{c)}	\$1575K [\$ 525K] ^{c)}
		()) () () ()	[101 ()/]	[サーンケンパー]

a) Excluding 6,7Li, and 10B.

b) Including only "destructive" uses (which cannot be satisfied by loans).

c) Annual usage.

V. SUMMARY AND CONCLUSIONS

In this document we have presented information on the research uses for electromagnetically separated stable isotopes, and radioisotopes derived therefrom, in the broad areas of physics, chemistry, and the geosciences. The information contained in this report is based on a nationwide survey of more than 1,000 physics, chemistry, and geology departments. As has been true in past studies, we found that research use of stable isotopes is greatest in the field of nuclear physics/chemistry. However, many other research areas were also found to have significant needs for separated isotopes, including medium energy, atomic, and solid state physics; physical, inorganic, and analytical chemistry; geochronometry and isotope geology. Demand for separated isotopes in the United States has remained very substantial; of the elements between hydrogen and bismuth which have more than a single isotope, only four were not used during the three-year period covered by this report. Altogether, 220 different isotopes were utilized in the research areas of physics, chemistry, and geology, corresponding to a total amount of material of nearly 3000 gram-atoms, at a cost of almost \$1.6M. Approximately 85% of the material was consumed in these experiments, while 15% was put to "non-destructive" uses.

Several problems were reported, however, in the supply of stable isotopes. The most critical of these was the complete lack of a number of important isotopes. Nearly 5% of the respondents to the survey indicated that they had recently been forced to abandon plans for at least one experiment due to an inability to obtain a required isotope. At present, the amount of time needed to generate new supplies of an out-of-stock isotope can be as long as 2-3 years, which is incompatible with the shorter time frame in which most research is carried out. On the positive side, the

Research Isotopes Pool arrangement appears to be operating reasonably smoothly, and the research community has taken great advantage of it.

Another problem is the lack of sufficient enrichment for many isotopes of low natural abundance. There is a definite trend toward increased use of nuclides at the extremes of an isotope distribution. At present these are frequently not enriched sufficiently to be useful either as targets in nuclear physics experiments or for the production of high-purity radioisotopes. It is expected, however, that these isotopes, at least up to about A=100, will also see substantial service in the production of exotic ion beams. For this use, an enrichment of 50-80% should be acceptable.

Skyrocketing isotope costs are also a matter of concern. Such important isotopes as 48 Ca are rapidly becoming unusable due to their high cost. In spite of this, there was a clear consensus among the respondents that the value of separated isotopes to their research is so great that, within reason, they would be willing to pay significantly higher costs in order to ensure a steady supply.

There appears to be a downward trend, at least in nuclear physics, in available research personnel. If this trend continues, it will eventually begin to have a substantial negative impact on isotope utilization. It presently appears, however, that the rate of isotope usage in the next 5 years will remain essentially constant. Of course, the particular choices will change during this period. It is expected that future emphasis will be on rare earth nuclei, most of the lighter neutron-rich isotopes, and on those elements having a large number of stable isotopes, such as Ca, Mo, Sn, and Sm. In addition, fabrication of isotopically pure shielding and filters for neutron work will take kilogram amounts of such isotopes as ^6Li , ^{10}B , and ^{56}Fe .

The availability of separated isotopes is absolutely essential to the continued health of much of the physics, chemistry and geoscience research in this country. We are at a crossroads in terms of our ability to provide adequate supplies of suitably enriched isotopes for the future. It is critical, therefore, that the availability of stable isotopes be improved to the point where adequate supplies exist to handle the fluctuating demands of an active research community. Not to do so will eventually mean that a large portion of the physical science research in this country cannot be performed.

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Appendix A SURVEY OF ISOTOPE USAGE

Lawrence Berkeley Laboratory

University of California • Berkeley, California 94720

September 18, 1981

Dear Colleague:

In response to a request from the Office of Basic Energy Sciences (OBES) of the U.S. Department of Energy, the National Research Council is organizing a Workshop on the Applications of Stable Isotopes and Derived Radioisotopes, under the aegis of its Subcommittee on Nuclear and Radiochemistry. Basically, the OBES would like documentation from the user community on present and projected applications of, and requirements for, electromagnetically separated stable isotopes in physics/chemistry research, biomedical research, clinical medicine, and industry. One goal of the Workshop, therefore, is to generate a report which summarizes the needs for these isotopes and also makes quantitative estimates of the amounts required in the future.

As part of this effort, I have been asked to gather information on the uses of stable isotopes in the broad areas of physical and chemical research, including nuclear physics/chemistry, geophysics/geochemistry, mass spectroscopy, etc. Because of the somewhat amorphous nature of the area for which I am responsible, it is difficult to arrive at an inclusive mailing list. For this reason, it is essential that you distribute copies of this note to any colleagues at your institution who have (or expect to nave) need for either separated stable isotopes or the radioisotopes derived therefrom.

My suggestion is that you appoint a "contact person" at your institution (perhaps a secretary) who will ensure that the enclosed questionnaire is copied, distributed, collected and returned to me. It would be helpful to have the name and phone number of this person as soon as possible; a simple form for this purpose is attached. Should there be no members of your institution who use stable isotopes in their research, please indicate this on the return form and I will delete your name from our mailing list.

Please have all your questionnaires filled out and returned to me by October 16, 1981. The information you provide will help the OBES ensure a continuing supply of stable isotopes in the future.

Thank you very much for your cooperation in this effort.

Telephone: (415) 486-4000 FTS 451-4000

Please return your completed questionnaire by Oct. 16, 1981 to:
Michael S. Zisman
Building 71
Lawrence Berkeley Laboratory
Berkeley, CA 94720

Questionnaire

	Grescionnaire
	Applications of Stable Isotopes and Derived Radioisotopes
	(Please type or print - use black ink.)
1.	Name; Title
2.	Institution; Mailing Address; Phone Number
3. a)	Identify and describe your field(s) of research.
ь)	$\underline{\textbf{List}}$ a few of your recent publications in the above fields.

4. a) Describe (briefly) how stable isotopes, or radioisotopes derived

therefrom, are used in your research, e.g., as target materiais, to produce ion beams, as tracers, etc.

	6)	amounts of such isotopes used in the past 3 years, e.g., ⁵⁶ Fe (500 mg).
5.	a)	Where were your isotopes obtained?
	ь)	Have you had problems with availability of required isotopes or significant delays in obtaining them? If so, how did you solve these problems?
	c)	Do you precently have a stockpile of separated isotopes? If so, what materials (and quantities) are included?
6.	a)	If you utilize radioisotopes derived from separated stable isotopes, what reactions were used for the conversion?
	b)	Where and by whom were the derived radioisotopes produced?
	c)	Indicate why <u>separated</u> stable isotopes must ce used in the radioisotope production process, e.g., unwanted competing reaction products, etc. Be specific.

So much for the easy part. Now for the future . . . (Please note that it is important to be realistic here; the questions below do not involve research budgets!)

- a) Indicate the isotopes you envision using in the next 3-4 years and their amounts (as in question 4a).
 - b) Describe how these isotopes will be used, emphasizing differences compared with your answer to question 4. (For example, do you anticipate significantly different uses for the isotopes or radioisotopes, or significant changes [up or down] in the amounts which will be required? If so, why?)

c) How do you anticipate your needs for stable isotopes changing in th; long term (beyond the next 5 years)? (Do you see your needs remaining nearly constant or increasing/decreasing significantly? Please explain.)

8. How important are separated stable isotopes or derived radioisotopes to your research? Would your research be significantly impaired if certain isotopes were temporarily or permanently unavailable, or if the prices of such isotopes were substantially higher than at present?

Congratulations! You have completed the hardest part. Now all that's left is for you to mail the questionnaire back to me at the address given on the first page. Thank you for your help.

Appendix B INSTITUTIONS CONTACTED

Alabama

University of Alabama
University of Alabama in Birmingham
University of Alabama in Huntsville
Auburn University
Tuskegee Institute

Alaska

University of Alaska at Fairbanks
University of Alaska at Juneau

Ar.zona

Arizona State University University of Arizona Northern Arizona University

Arkansas

Arkansas State University University of Arkansas at Fayetteville University of Arkansas at Little Rock University of Central Arkansas Ouachita Baptist University

Cali fornia

Aerospace Corp. Space Science Laboratory California Institute of Technology California Institute of Technology - Jet Propulsion Laboratory California Polytechnic State University at San Luis Obispo California State Polytechnic University at Pomona California State University at Fresno California State University at Fullerton California State University at Hayward California State University at Humbolt California State University at Long Beach California State University at Los Angeles California State University at Northridge California State University at Sacramento University of California at Berneley University of California - Lawrence Berkeley Laboratory University of California - Lawrence Livermore National Laboratory University of California at Davis University of California at Irvine University of California - Institute of Geophysics and Planetary Physics University of California at Riverside
University of California at San Diego
University of California at Santa Barbara
University of California at Santa Cruz
Naval Postgraduate School, Monterey
Pacific Union College
University of the Pacific
San Diego State University
San Francisco State University
University of San r-rancisco
San Jose State University
University of Southern California
Stanford University - High Energy Physics Laboratory
Stanford University - Accelerator Center

Colorado

Atomic Physics Chemistry Laboratory, Boulder Colorado School of Mines Colorado State University University of Colorado University of Colorado at Colorado Springs University of Colorado at Denver University of Denver, Colorado Seminary National Center for Atmospheric Research University of Northern Colorado Space Environment Laboratory, NOAA Western State College of Colorado

Connecticut

University of Bridgeport
Central Connecticut State College
University of Connecticut
University of Connectic : - Institute of Material Science
Guinnipiac College
University of Hartford
Saint Joseph College
Trinity College
Wesleyan University
Yale University

Delaware

University of Delaware

District of Columbia

The American University
Carnegie Institute of Washington
Carnegie Institute - Geophysics Laboratory
The Catholic University of America
George Washington University
Georgetown University
Howard University
National Bureau of Standards

Florida

University of Central Florida Florida Atlantic University Florida Institute of Techi-logy Florida International University Florida State University at Tallahassee University of Florida University of Miami University of South Florida

Georgia

Albany State College Atlanta University Columbus College Emory University Georgia Institute of Technology Georgia State University University of Georgia

Hawaii

University of Hawaii

Idaho

Idaho National Engineering Laboratory Idaho State University University of Idaho

Illinois

Argonne National Laboratory Bradley University University of Chicago University of Chicago Enrico Fermi Institute DePaul University Eastern Illinois University Fermi National Accelerator Laboratory Illinois Institute of Technology Illinois State University University of Illinois at Chicago Circa University of Illinois at Urbana-Champaign Loyola University of Chicago Northeastern Illinois University Northern Illinois University Northwestern University Roosevelt University Southern Illinois University - Carbondale Southern Illinois University - Edwardsville Western Illinois University

Indiana

Ball State University Butler University Indiana State University Indiana University Indiana University - Purdue University at Indianapolis University of Notre Dame Purdue University Rose-Hulman Institute

Iowa

Drake University
Jowa State University of Science and Technology
Jowa State University
Jowa State University - Ames Laboratory
University of Jowa
University of Northern Jowa

Kansas

Emporia State University
Fort Hays State University
Kansas State University
Kansas State University - MacDonald Atomic & Nuclear Physics
Laboratory
University of Kansas
Pittsburgh State University
Wichita State University

Kentucky

Eastern Kentucky University University of Kentucky University of Louisville Murray State University Western Kentucky University

1 ouisiana

Louisiana State University
Louisiana Tech University
McNeese State University
University of New Orleans
Northeast Louisiana University
Northwestern State University of Louisiana
Southern University
University of Southwestern Louisiana
Tulane University
Xavier University of Louisiana

Maine

University of Maine at Orono Woods Hole Oceanographic Institute

Maryland

Frostburg State College
Goddard Space Flight Center
The Johns Hopkins University
The Johns Hopkins University - Applied Physics Laboratory
Loyola College
University of Maryland
University of Maryland, Baltimore County
University of Maryland, Eastern Shore

Massachusetts

Amherst College Boston College Boston University Brandeis University

Bridgewater State College

Clark University

Harvard University

Harvard University - Cyclotron Laboratory

College of the Huly Cross

University of Lowell

Massachusetts College of Pharmacy

Massachusetts Institute of Technology

Massachusetts Institute of Technology - Bates Linear Accelerator

Laboratory

Massachusetts Institute of Technology - Center for Space Research
Massachusetts Institute of Technology - Francis Bitter National Magnet
Laboratory

Massachusetts Institute of Technology - Nuclear Science Laboratory

University of Massachusetts at Amherst

University of Massachusetts at Boston (Harbor Campus)

Mount Holvoke College

Northeastern University

Smith College

Smithsonian Astrophysics Laboratory Southeastern Massachusetts University

Tufts University

Worcester Polytechnic Institute

Michigan

Andrews University
Central Michigan University
University of Detroit
Eastern Michigan University
Michigan State University
Michigan Technological University
University of Michigan
Northern Michigan University
Saginaw Valley State College
Wayne State University
Western Michigan University

Minnesota

Mankato State University
University of Minnesota
University of Minnesota at Minneapolis
University of Minnesota at Duluth
St. Cloud State University
Southhampton College

Mississippi

Alcorn State University
Delta State University
Jackson State University
Mississippi State University
University of Mississippi
University of Southern Mississippi

Missouri

Fontbonne College
University of Missouri - Columbia
University of Missouri - Kansas City
University of Missouri - Rolla
University of Missouri - St. Louis
Northeast Missouri State University
St. Louis University
Southeast Missouri State University
Southwest Missouri State University
Washington University

Montana

Montana College of Mineral Science & Technology Montana State University University of Montana

Nebraska

Creighton University Kearney State College University of Nebraska - Lincoln

Nevada

University of Nevada at Las Vegas University of Nevada at Reno

New Hampshire

Dartmouth College University of New Hampshire

New Jersey

Fairleigh Dickinson University, Teaneck
Fairleigh Dickinson University, Florham-Madison Campus
Montclair State College
Monmouth College
New Jersey Institute of Technology
Princeton University
Princeton University - Center for Environmental Studies
Princeton University - Geophysics Fluids Dynamics Laboratory
Princeton University - Plasma Physics Laboratory
Rutgers University - The State University of New Jersey
Seton Hall University
Stevens Institute of Technology
Trenton State College

New Mexico

Eastern New Mexico University

Los Alamos Scientific Laboratory

New Mexico Highlands University

New Mexico Institute of Mining and Technology

New Mexico State University

University of New Mexico

New York

Adelphi University

Brookhaven National Laboratory

PhD Program in Chemistry of the C.U.N.Y.

Brooklyn College of the C.U.N.Y.

City College of the C.U.N.Y.

Hunter College of the C.U.N.Y.

John Jay College of Criminal Justice of the C.U.N.Y.

Queens College of the C.U.N.Y.

Clarkson College of Technology

Columbia University

Cooper Union for the Advancement of Science and Art

Cornell University

Cornell University - Atomic and Solid State Physics Laboratory

Fordham University

Long Island University, Southampton College

Manhattan College

New York University

Polytechnic Institute of New York

Polytechnic Institute - Radiation Physics/Environmental Measurements

Laboratory

Rensselaer Polytechnic Institute

Rochester Institute of Technology

University of Rochester Rochester University

Saint Bonaventure University

St. John's University

S.U.N.Y. at Albany

S.U.N.Y. at Binghamton

S.U.N.Y. at Buffalo

S.U.N.Y., College of Environmental Science and Forestry

S.U.N.Y. at Fredonia

S.U.N.Y. at Geneseo

S.U.N.Y. at New Paltz

S.U.N.Y. at Oneonta

S.U.N.Y. at C. rego

5.U.N.Y. at Plattsburgh

S.U.N.Y. at Stony Brook

College of Staten Island

Syracuse University

Union College

Vassar College

North Carolina

Appalachian State University Duke University

East Carolina University

North Carolina Agricultural and Technical State University

North Carolina Central University

North Carolina State University

University of North Carolina at Chapel Hill

University of North Carolina at Charlotte

University of North Carolina at Greensboro

Wake Forest University

Western Carolina University

North Dakota

North Dakota State University University of North Dakota

Ohio

Air Force Institute of Technology (Wright-Patterson AFB)

The University of Akron

Bowling Green State University

Case Wastern Reserve University

University of Cincinnati

University of Cincinnati - Laboratory of Basic & Applied Nuclear

Research

Cleveland State University University of Dayton

John Carroll University

Kent State University

Kent State University - Lewis Research Center

Miami University

The Ohio State University

Ohio University (Athens)

University of Toledo

Wright State University

Xavier University

Youngstown State University

Oklahoma

Central State University

Oklahoma State University University of Oklahoma

The University of Tulsa

Oregon

Oregon Graduate Center 'Beaverton)

Oregon State University

University of Oregon

Portland State University

Pennsylvania

Bryn Mawr College

Bucknell University

Carnegie-Mellon University

Drexel University Duquesne University East Stroudsburg State College Indiana University of Pennsylvania Lehigh University The Pennsylvania State University The Pennsylvania State University - Applied Research Laboratory The Pennsylvania State University - Materials Research Laboratory University of Pennsylvania University of Pittsburgh Saint Joseph's University University of Scranton Shippensburg State College Swarthmore College Temple University Villanova University West Chester State College Wilkes College Rhode Island Brown University

University of Rhode Island

Providence College Rhode Island College

South Carolina
Clemson University
Furman University
University of South Carolina

South Dakota

South Dakota School of Mines and Technology South Dakota State University University of South Dakota

Brown University - Materials Research Laboratory

Tennessee

East Tennessee State University
Fisk University
Memphis State University
Middle Tennessee State University
Oak Ridge Associated Universities (UNISOR)
Oak Ridge National Laboratory
Tennessee State University
Tennessee Technological University
The University of Tennessee
Vanderbilt University

Texas

Baylor University
East Texas State University
University of Houston, Central Campus
University of Houston, Clear Lake City
Lamar University

North Texas State University Rice University Sam Houston State University Southern Methodist University Southwest Texas State University Stephen F. Austin State University Tarleton State University Texas A & I University Texas A & M University Texas A & M University - Cyclotron Institute Texas Christian University Texas Tech University The University of Texas at Arlington The Univer ty of Texas at Austin The University of Texas at Dallas The University of Texas at El Paso The University of Texas at San Antonio Texas Woman's University Trinity University West Texas State University

Utah

Brigham Young University Utah State University University of Utah

Vermont

Middlebury College University of Vermont

Virginia

Lynchburg College
Old Dominion University
Radford University
University of Richmond
Virginia Commonwealth University
Virginia Institute of Marine Science
Virginia Polytechnic Institute and State University
Virginia State University
University of Virginia
College of William and Mary

Washington

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West Virginia

Marshall University
West Virginia Institute of Technology
West Virginia University

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McMaster University University of British Columbia

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C. P. Slichter	(Physics)	Laboratory	-titles
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Robert L. Switzer	(Chemistry)	John C. Hill Robert S. Houk	(Physics) (Chemistry)
Dale J. Van Harlingen	(Physics)	Géorge Kraus	(Chemistry)
Albert Wattenberg Hartmut Zabel	(Physics)	,	,,
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Northern Illinois Universit	•	William C. Stwalley	(Physics)
Carla Montgomery	(Geoscience)	Kansas	
Northwestern University		Fort Hays State University	ty
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K. K. Seth	(Physics)	Richard Kjonaas	(Chemistry)
William W. Traessley	(Chemistry)	Maurice H. Witten	(Physics)
<u>Indiana</u>		Kansas State University	
Ball State University		John S. Eck	(Physics)
Malcom Hults	(Physics)	University of Kansas	
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Massachusetts Institute of Technology

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Xavier University of Louis		∷enneth L. Williamson	(Chemistry)
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Paul J. Dagdigian John W. Kohl	(Chemistry) (Physics)	Edward L. Fireman	(Chemistry)
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University of Maryland		George J. Thomas, Jr.	(Chemistry)
Glen E. Gordon	(Chemistry)	Tufts University	
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Missouri		Los Alamos Scientific La	eboratory
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University of Missouri - C		Helmut Baer	(Physics)
University of Missouri - C James J. Carni Gary J. Ehrhardt	Columbia (Chemistry) (Chemistry)	•	. *
James J. Carni	(Chemistry)	Helmut Baer Richard L. Boudrie Merle E. Bunker M. Leon Joel Moss	(Physics) (Physics) (Physics) (Physics) (Physics) (Physics)
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Cornell University	(55, ,	Joseph Jach W. Le Noble	(Chemistry) (Chemistry)
Barbara A. Baird	(Chemistry)	Charles S. Springer, Jr.	(Chemistry)
Barry K. Carpenter	(Chemistry)	Syracuse University	•
David D. Clark	(Physics) (Chemistry)	George C. Levy	(Chemistry)
David B. Collum Michael E. Fisher	(Chemistry)	Union College	,
Jack H. Freed	(Chemistry)	Alfred T. Goble	(Physics)
Bruce Ganem Melvin J. Goldstein	(Chemistry) (Chemistry)	North Carolin	•
Paul Houston	(Chemistry)		
Fred W. McLafferty	(Chemistry)	Duke University	(C)
L. Que James R. Rasmussen	(Chemistry) (Chemistry)	F. O. Purser R. L. Walter	(Physics) (Physics)
H. A. Scheraga	(Chemistry)	Chris Westerfeldt	(Physics)
David A. Usher Peter T. Wolczanski	(Chemistry) (Chemistry)	University of North Carol Chapel Hill	ina at
Long Island University, B	rooklyn Center	Paul D. Fullagar	(Geoscience)
Raymond DiMartini	(Chemistry)	Edward J. Ludwig	(Physics)
New York University		Wake Forest University	
Burton Budick	(Physics)	Ysbrand Haven	(Pnysics)
Randall B. Murphy Olav Redi	(Chemistry) (Physics)	Western Carolina Universi	ty
Rensselaer Polytechnic In	stitute	Gene F. Morris	(Chemistry)
Herbert M. Clark	(Chemistry)	North Dakota	!
	,,	University of North Dakot	a
		Glenn I. Lykken	(Chemistry)

<u>Ohio</u>		John A. Gardner David M. Nelson	(Physics) (Geoscience)
Air Force Institute of Technology (Wright-Patterson AFB)		University of Oregon	(raedacterice)
George John	(Chemistry)	J. C. Overley	(Physics)
Bowling Green State Un	niversity	Portland State Universit	sy .
Edgar B. Singleton	(Physics)	Rudi H. Nussbaum	(Chemistry)
Case Western Reserve U	Jniversity	Pennsylvan	
John P. Facler	(Chemistry)	Carnegie-Mellon Univers	ity
Willam M. Ritchey	(Chemistry)	John L. Anderson	(Chemistry)
University of Cincinnati		Peter D. Barnes	(Physics)
S. Jha	(Physics)	A. A. Caretto Morton Kaplan	(Chemistry) (Chemistry)
John Carroll University		Paul J. Karol	(Chemistry)
Joseph Trivisonno	(Physics)	Drexel University	
Kent State University		Jack G. Kay	(Chemistry)
Bryon Anderson	(Physics)	Allan L. Smith	(Chemistry)
Richard Madey David L. Uhrich	(Physics) (Physics)	Lehigh University	
John Watson	(Physics)	L. H. Sperling	(Chemistry)
University of Miami		The Pennsylvania State I	University
Norman K. Grant	(Geoscience)	R. P. Danner	(Cr.emistry)
Michael R. Van De Ma	rk (Chemistry)	Peter Deines Edgar Earl Graham	(Geoscience) (Chemistry)
The Ohio State University	ty	E. R. Vance	(Physics)
Gunter Faure	(Geoscience)	University of Pittsburgh	
Ming-Daw Tsai	(Chemistry)	D. P. Balamuth	(Physics)
Ohio University at Ather	ns	Manfred A. Biondi	(Physics) (Physics)
Roger W. Finlay	(Physics)	T. Chapuran Wilfried W. Daehnick	(Physics)
Xavier University		H. T. Fortune	(Physics)
Theodore C. Thepe	(Chemistry)	Irving J. Lowe J. V. Maher	(Physics) (Physics)
Oaklahoma	•	R. Middleton	(Physics)
Oklahoma State Universit	•	J. X. Saladin	(Physics)
	•	L. E. Seiberling	(Physics)
E. J. Eisenbraun	(Chemistry)	Darel K. Straub R. W. Zurmuhle	(Chemistry) (Physics)
<u>Oregan</u>			(1.193103)
Oregon Graduate Center	(Beaverton)	Temple University	(5)
J. K. Hurst	(Chemistry)	David R. Dalton	(Chemistry)
Thomas M. Loehr Lois E. Rasmussen	(Chemistry) (Chemistry)	Rhode Island	<u>d</u>
Oregon State University	(Siletifietty)	Brown University	
-	(0	David E, Cane	(Chemistry)
T. M. Reasley	(Geoscience)		

John O. Edwards	(Chemistry)	Texas	
Ronald G. Lawler	(Chemistry)	University of Houston, C	lear Lake City
Philip H. Rieger Michael Squillacote	(Chemistry) (Chemistry)	Ruthann A. Masaracchia	(Chemistry)
J. William Suggs	(Chemistry)	North Texas State Univer	sity
University of Rhode Islan	nd	R. W. Gracy	(Chemistry)
Stanley J. Pickart	(Physics)	Ben G. Harris	(Chemistry)
South Caroli	•	Andras G. Lacko Ruthann A. Masaracchia	(Chemistry) (Chemistry)
University of South Card		S. J. Norton	(Chemistry)
•		Robert M. Pirtle	(Chemistry)
F. T. Avignone Gary S. Blanpied	(Physics) (Physics)	Richard E. Thompson	(Chem:stry)
Barry M. Preedom	(Physics)	Rice University	
B. G. Ritchie	(Physics)	Robert F. Curl	(Chemistry)
Tennessee		Edward S. Lewis	(Chemistry)
East Tennessee State Un	iversity	Ronald J. Parry	(Chemistry)
David M. Close	(Chemistry)	Texas A & M University	
		Wiley P. kirk	(Physics)
Oak Ridge Associated Un	nivers itie s	J. B. Natowitz	(Chemistry)
(UNISOR)		Jeevak M. Parpia	(Physics)
E. H. Spejewski	(Physics)	Hans A. Schuessler	(Physics)
Oak Ridge National Labo	ratory	Texas A & M University Institute	- Cyclotron
R. Auble	(Physics)		<i>i</i>
J. R. Beene	(Physics)	David R. Haenni	(Chemistry)
C. E. Bemis, Jr.	(Physics) (Physics)	R. P. Schmitt Robert Tribble	(Chemistry) (Physics)
F. E. Bertrand J. Kirk Dickens	(Physics)	David Youngblood	(Physics)
K. Erb	(Physics)	-	(···) 0.00)
R. L. Ferguson	(Chemistry)	Texas Tech University	
J. Ford	(Physics)	John N. Marx	(Chemistry)
C. Fulmer	(Physics)	Robert W. Shaw	(Chemistry)
J. Gomez del Campo	(Physics)	Henry J. Shine	(Chemistry)
M. L. Halbert	(Physics) (Physics)	Pill Soon Song	(Chemistry)
D. C. Hensley D. Horen	(Physics)	The University of Texas a	t Austin
Noah Johnson	(Chemistry)	Lean E. Long	(Geoscience)
R. Novotny	(Physics)	Fred W. McDowell	(Geoscience)
F. E. Obenshain F. Plasil	(Physics) (Physics)	The University of Texas a	t Dallas
R. L. Robinson	(Physics)	William I. Manton	(Geoscience)
D. Shapira	(Physics)	Texas Woman's University	•
T. Sjoreen	(Physics)	•	_
G. R. Young	(Physics)	Norman G. Foster James E. Johnson	(Chemistry) (Chemistry)
Vanderbilt University		depide L. dellison	(Chemistry)
Leo T. Burka Joel Tellinghuisen	(Chemistry) (Chemistry)		

<u>Utah</u>		J. Michael Schurr	(Chemistry)
Brigham Young University		D. W. Storm Robert Vandenbosch	(Physics) (Chemistry)
Max Hill	(Physics)	West Virgin	ia
Kenneth A. Solen James Thorne	(Chemistry) (Chemistry)	West Virginia University	-
Utah State University		N. S. Dalal	(Chemistry)
Jack T. Spence	(Chemistry)	Wisconsin	
University of Utah		Marquette University	
Stanley H. Evans, Jr.	(Geoscience)	Kazuo Nakamoto	(Chemistry)
Robert W. Parry C. Dale Poulter	(Chemistry) (Chemistry)	University of Wisconsin a	
Vermont		James M. Cosk Russell F. Howe	(Chemistry) (Chemistry)
University of Vermont		Calvin O. Huber	(Chemistry)
K. Spartalian	(Physics)	James T. McFarland James Otvos	(Chemistry) (Chemistry)
Virginia		Frank Shaw, III	(Chemistry)
Virginia Polytechnic Instit State University	tute and	Kenneth L. Watters University of Wisconsin a	(Chemistry) at Oshkosh
Marvin Blecher	(Physics)	Roy R. Knispel	(Physics)
Kazuo Gotow David G. I. Kingston	(Physics) (Chemistry)	Wyoming	
Washington	(Chemistry)	University of Wyoming	
Washington State Universi	itu	Anthony L. Hines	(Chemistry)
Steven Brown	(Chemistry)	Ray Kunselman Emanuel Pedram	(Physics) (Chemistry)
H. H. Cheng	(Chemistry)		
K. W. Hipps	(Chemistry)	British Columb	<u>Dia</u>
J. P. Hunt J. A. Magnuson	(Chemistry) (Chemistry)	McMaster University	6 1
K. D. McMichael	(Chemistry)	P. G. Ashbaugh D. G. Burke	(Physics) (Physics)
M. Pall D. M. Roundhill	(Chemistry)	J. R. D. Copley	(Physics)
M. J. Smerdon	(Chemistry) (Chemistry)	R. H. McNutt	(Geoscience)
B. Srinivasan	(Chemistry)	A. A. Pilt W. V. Prestwich	(Physics) (Physics)
University of Washington	-	Henry P. Schwarcz	(Geoscience)
Bruce E. Eichinger	(Chemistry)	W. W. Smeltzer	(Chemistry)
Norval Fortson	(Physics)	Ian D. Spenser	(Chemistry)
Geruas M. Hinn	(Physics)	R. G. Summers-Gill J. C. Waddington	(Physics) (Physics)
Albert I. Lazzarini John Maklin	(Physics) (Chemistry)	Colin E. Webber	(Physics)
Donald R. McAlister	(Chemistry)	University of British Colu	mbia
Eric B. Norman	(Physics)	Richard Lee Armstrong	(Geoscience)
Y. Pocker B. S. Rabinovitch	(Chemistry) (Chemistry)	K. L. Pinder	(Chemistry)
Brian R. Reid	(Chemistry)		