Minerals and Rocks 20

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Neodymium Isotope Geochemistry

An Introduction

With 104 Figures

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To Lynn,
Tara, and Daniel

Preface

This monograph was begun with two objectives in mind. The first was to provide a review of research involving the application of neodymium isotopic measurements to problems in earth science. In the process of organizing to do this, I realized that the research in this field had produced a need for an updated review of the underlying paradigms. This need had arisen because of the special properties of the samarium-neodymium isotopic system, and because the research had transgressed the traditional boundaries between the subfields of earth science. Without such a review, the significance of the results seemed likely to remain unnecessarily obscure to interested scientists from related disciplines. Consequently, the second objective became the provision of a theoretical framework for the application of neodymium isotopic studies. Much of what this contains is not new, but it is drawn together here for the first time.

At the time the writing was initiated, the literature of the field was still relatively limited. Over the past 5 years it has grown enormously. Considering the rate at which the writing progressed, it became clear that this could not be a fully up-to-date review and still reach completion. The selection of material for the review sections is biased toward earlier studies.

Part I presents most of the background information. Chapter 1 provides some historical context, a qualitative description of the niche filled by Nd isotopic studies, and some basic properties of Sm and Nd. Chapters 2 through 4 contain the "nuts and bolts." Particularly important aspects are the relationship between the notation and models of planetary evolution (Chap. 3) and the relationship between models for igneous processes (Chap. 4) and models for isotopic evolution. Part II contains an overview of the results as they apply to problems on a planetary scale, including applications to sedimentary rocks and oceanography. Part III contains discussion of results related to problems of the origins of igneous rocks.

The manuscript was written while I was at the Department of Earth and Space Sciences at the University of California, Los Angeles. Harriet Arnoff helped with the typing of early versions. Jean Sells did most of the drafting, with contributions from Vickie Doyle-Jones. Terry Gautesen-Borg transferred the text onto personal computer discs, without which it never would have been completed. Lynn Ingram-DePaolo helped with proofreading. G. Lang Farmer, Bruce Nelson, Vickie Bennett, Brian Stewart, Frank Perry, Don Musselwhite, and Steve Semken contributed data and ideas that are integrated into the text. Research support from the U.S. National Science Foundation was instrumental in allowing this monograph to be prepared.

Berkeley, California   DONALD J. DEPAOLO
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Chapter 1 Introduction

1.1 Geologic Perspective

The elements samarium and neodymium have little economic value and are present in only small amounts in most rocks and minerals, yet because of their nuclear and chemical properties, they provide access to information about the origin and evolution of the earth, the moon, and the solar system. They can be considered, like the microscope or the seismograph, to be a tool that can be applied to problems in geology and planetary science. As with other instruments of science, the interpretations of the natural variations of samarium and neodymium isotopic and chemical abundances are governed by principles, models, and assumptions. These and some of the results of the studies to which they have been applied are the subjects of this book.

The geologic utility of these elements stems from the fact that one isotope of samarium (^{147}Sm) is radioactive, and decays by the a-process to one isotope of neodymium (^{143}Nd). The decay proceeds at a very slow rate, the half-life of ^{147}Sm (106 billion years) being some 23 times greater than the age of the earth. Over geologic time, the decay of ^{147}Sm to ^{143}Nd results in changes in the abundance of ^{143}Nd relative to the other Nd isotopes. These small variations can be precisely measured with modern mass spectrometers. The regularity of radioactive decay makes the ^{147}Sm-^{143}Nd pair a timepiece, and one of its most important applications is the determination of the ages of rocks, or more generally, the determination of the timing of major events in the chemical evolution of planets. Sm-Nd studies of igneous rocks can also be used as probes into the earth's interior, capable of sensing the existence of layers or domains of different chemical composition as well as tracing the origin and development of these structures back through time. It is this capability, of determining not only the existence of structure in the earth's interior, but also of determining its age and origin, that makes isotopic studies of ^{147}Sm-^{143}Nd and other similar chronometric nuclide pairs valuable to planetary sciences. Like fossils, these nuclides preserve in the rock record information that allows the ancestry of the modern solid earth to be investigated. The results of these studies relate directly to such diverse fields as igneous, metamorphic and sedimentary petrology and geochemistry, paleo-geography, oceanography, crustal tectonics, and mantle geophysics.

1.2 Phylogeny

The application of naturally-occurring radioactive elements and their decay products to geology can be traced back to the discovery of radioactivity in the late 19th century by Becquerel and the Curies. A concise and readable account of this early history can be found in Faure (1977). Isotope geology during the first half of the 20th century was based mainly on the U-Pb method (Rankama 1954, 1963; Russell and Farquhar 1960). Perhaps the biggest single development during this time was the design and construction by Nier (1940, 1947) of mass spectrometers to routinely measure isotopic abundances. This led to a flowering of isotopic research in geology during the 1950s and 1960s (Faure 1977), and in particular, to wider application of the Rb-Sr and K-Ar techniques (e.g. Gast 1960; Faure and Hurley 1963; Hedge 1966; Wasserburg 1966; Aldrich and Nier 1948; Wasserburg and Hayden 1955; Wetherill et al. 1955; Hart 1964; McDougall 1966; Goldich 1968; Doe 1970; Faure and Powell 1972). Modern reviews can be found in Faure (1977) as well as O'Nions et al. (1979a), Hart and Brooks (1980), DePaolo (1983c), Zindler and Hart (1986), and Carlson (1987). The questions of concern in this section are: What determines which isotopes are useful in geology, and why was the Sm-Nd method applied only relatively recently?

   In general, the number of chronometric nuclide pairs available for use in geologic studies has been limited partly by the small number of nuclides with suitable half-lives, and partly by analytical difficulties. In order for measurements of the isotopic composition of an element in rocks and other materials of geologic interest to be feasible, certain requirements must be met. One requirement is to have chemical methods that can be used to separate the element from rocks in sufficiently pure form. The element must also have properties that will facilitate mass spectrometric analysis. With regard to the latter, the basic requirement for analyzability is that the element become sufficiently ionized in the mass spectrometer to produce a measureable ion beam. Standard methods of thermal ionization (Dempster 1918; Inghram and Chupka 1953; Wilson and Daly 1963) have proven most successful for elements that have low ionization potentials and are significantly less refractory than the standard filament materials, tantalum, rhenium, tungsten, and platinum. In some cases special techniques have been developed to ionize nonrefractory elements with large ionization potentials (Crouch 1963; Inghram and Chupka 1953). The most notable example is the use of a silica gel medium to enhance the thermal ionization of Pb (Cameron et al. 1969). However, understanding of the ionization process is limited, and special techniques are usually developed simply by trial and error. Satisfactory methods for the ionization of refractory elements with high ionization potentials are still lacking. Examples are osmium and hafnium, which are of considerable geochemical interest, and have become usable only recently (Allegre et al. 1980; Patchett and Tatsumoto 1980a, b) due to technical limitations. Some of the properties of the elements used in geochronology are summarized in Table 1.1, together

Table 1.1. Chemical properties relevant to mass spectrometry

	Melting point (°C)	First I.P. (eV)	Work function (eV)	Max. ioniza- tion efficien- cy (%)[a]	Smallest analyzable sample (ng)[b]	Crustal abundance (ppm)
Filament materials						
Re	3175	7.88	4.96	–	–	0.001
Pt	1772	9.0	5.65	–	–	0.01
Ta	2996	7.89	4.25	–	–	2
W	3410	7.98	4.55	–	–	1
Daughter elements						
Pb	327.5	7.42	4.25	2 (Si gel)	<1	7
Sr	772	5.69	2.59	0.5	200	400
Nd[c]	1016	5.49	3.3	5 (oxide)	20	20
Hf	2220	7.0	3.9	0.01	1000	3
Os	3015	8.7	4.82	<0.01?	?	0.005
Ca	842	6.11	2.87	0.1	5000	5×10^4
Parent elements						
U[c]	1132	6.08	3.63	0.1	<1	1
Th	1800	6.95	3.4	<0.1	<1	4
Rb	38.9	4.18	2.16	~100	<1	35
Sm[c]	1073	5.63	3.2	1	<1	4
Lu	1663	5.43	3.3	0.1	<1	0.4
K	63.6	4.34	2.30	~100	<1	10^4

[a] Maximum ionization efficiency is here defined as the ratio of the total number of "measurable ions" reaching the mass spectrometer ion collector to the number of atoms initially present on the source filament. "Measurable ions" refers to that portion of the run when the ion beam is of suffi- cient intensity and stability to yield precise isotopic ratio data.

[b] Measurements can be made on smaller samples of the parent element because only the concen- tration need be determined. Precise isotropic ratio measurements on the daughter element general- ly require larger samples.

[c] Elements that can be measured as oxide species (e.g. NdO^+, UO_2^+). The ionization energy can be more favorable for the oxide, although the actual values are not available.

with an indication of the ease with which they can be analyzed by thermal ionization mass spectrometry.

Geochemical considerations are closely tied to the technical problems. The variability of the parent/daughter ratio in nature, which is a function of the chemical contrast between the two elements, and the half-life, determines the magnitude of isotopic variations that can occur. The variations must be substantially larger than the precision of a measurement in order to be useful. The attainable precision is dictated by the properties of the element as discuss- ed above, combined with limitations imposed by the design and construction of the mass spectrometer. Finally, the natural abundances of the elements are important, since elements present in extremely low concentrations in rocks can present problems in chemical separation procedures.

The relatively recent application of the Sm-Nd method to problems in geology was made possible by a series of technical developments which took

place over 3 decades. As is true of many techniques in the earth sciences, the lead-up developments originated in other fields, chemistry and physics in this case, and the implementation proceeded in small steps, over a substantial time period, as those developments were made available to geochemists who could identify the potential applications.

The first step toward the eventual use of Sm-Nd studies in geology can be traced to the development of ion exchange methods for the efficient separation of rare earth elements (REE). These were first developed in the 1940s to separate uranium fission products produced in nuclear reactors (Spedding et al. 1947; see reviews by Peppard 1961 and Powell 1961). In the 1960s these procedures were employed to partially separate REE for the purpose of determining their abundances in rocks by neutron activation (Haskin et al. 1966). The methods for high purity separation of individual rare earths for mass spectrometric measurements were first applied by Eugster et al. (1970) who studied neutron-induced isotopic effects in Gd and Sm from meteorites and lunar samples.

The second step involved the improvement of the precision of isotopic abundance ratio measurements. The variation of the abundance of ^{143}Nd in nature could be estimated to be small, of the order of a few tenths of a percent, due to the long half-life of ^{147}Sm and the limited variation of Sm/Nd. The usefulness of the Sm-Nd system depended upon the ability to make precise measurements of Nd isotopic abundances. The typical precision for isotopic ratio measurements that was attainable with the thermal ionization mass spectrometers used until the 1960s was $\pm 0.1\%$ (e.g. Hedge 1966); about the same magnitude as the variations in ^{143}Nd. This particular limitation was imposed by the stability of the ion beam, the detector characteristics, and the data acquisition procedures rather than by the properties of the elements. Measurements were made by manually reading peak heights from a chart recorder while the mass spectrum was scanned in analog mode by varying the magnetic field. The development of a computer-interfaced mass spectrometer with rapid magnetic field switching and digital data acquisition by Wasserburg et al. (1969) improved the precision by a factor of 30 and brought the use of the Sm-Nd system into the range of feasibility.

In pioneering studies of the meteorite Juvinas and a lunar basalt, Lugmair (1974) and Lugmair et al. (1975 a) combined the chemical methods of Eugster et al. (1970) with modern mass spectrometric techniques to become the first to exploit the Sm-Nd system in geochemistry. Their studies demonstrated that the system could be used to obtain precise ages of old rocks, and also yielded important evidence in support of the early chemical differentiation of the moon.

As shown in Table 1.1, the chemical properties of Nd are favorable for precise isotopic measurements, especially the low ionization potential and relatively low melting point. In fact, using standard techniques, Nd samples can be analyzed with such ease that measurements can be made to the precision limits of present-day mass spectrometers on only 2×10^{-8} g of separated Nd.

For a typical rock sample that has a Nd concentration of 20 parts per million (ppm) by weight, this amount of Nd is supplied by only 1 mg of rock; a piece the size of the ball on a ball-point pen.

1.3 Characteristics of the Sm-Nd System and Comparison with Rb-Sr and U-Th-Pb

The application of Sm-Nd isotopes draws on an extensive groundwork laid by the previous studies of ^{87}Rb-^{86}Sr-, ^{235}U-^{207}Pb, ^{238}U-^{206}Pb, and ^{232}Th-^{208}Pb. The value of ^{147}Sm-^{143}Nd relative to these other pairs can be likened to the value of an electron microscope relative to an optical microscope or an X-ray diffractometer. Just as each instrument is sensitive to a certain aspect of mineral structure which cannot be detected with the other instruments, ^{147}Sm-^{143}Nd is sensitive to certain geologic processes and insensitive to others, in a manner that is complementary to the other nuclide pairs. The principles are the same, the differences lie only in the chemical properties of Sm and Nd, which enable them to magnify the effects of some processes which would be unresolvable using the other nuclide pairs, while being oblivious to other processes that are better investigated with the other nuclides.

    The study of Nd isotopic variations is unique among isotopic investigations in that the first measurements were preceded by more than a decade of intensive study of the distribution of Sm and Nd and the other rare-earth elements in rocks and minerals (e.g. Haskin et al. 1966; Schilling and Winchester 1967; Frey et al. 1971; Taylor 1964; Gast 1968; Kay and Gast 1973; see Haskin and Paster 1979; Hanson 1980; and Frey 1982 for reviews). REE studies have contributed immensely to knowledge of the petrogenesis of a wide variety of rock types, and had developed into an important geochemical field well before the first Sm-Nd isotopic measurements were made. The Sm-Nd studies came at a time when the geochemistry of both parent and daughter were already well understood, providing the opportunity to add time constraints to an already valuable geochemical technique.

1.3.1 General Geochemistry and Cosmochemistry

For Sm-Nd, both the parent and daughter elements are refractory in the sense that they are thought to occur early in the sequence of elements that would have condensed from the cooling solar nebula 4.5 billion years ago (Grossman and Larimer 1974). Consequently, parent/daughter fractionation during condensation of the solar nebula should have been negligible because both elements were quantitatively transferred into the early-formed solid bodies that later accreted to form the planets. Boynton (1975) has calculated that only under conditions of extreme disequilibrium will Sm and Nd be fractionated

during condensation. Therefore, most of the variation of Sm/Nd in the earth today is a result of the earth's internal differentiation rather than a result of condensation processes (Chap. 3). In contrast, Pb is much more volatile than U and Th, and Rb more volatile than Sr under the conditions thought to have prevailed in the solar nebula, so large parent/daughter fractionation may have occurred during condensation.

Available data support these inferences. The ratio of U/Pb in the earth is 25 times higher than the solar value, and Rb/Sr is about 8 times lower. The moon is a more extreme example. The ratio U/Pb in the moon is perhaps 100 times higher, and the ratio Rb/Sr 30 times lower than the solar ratio. By comparison, isotopic data on terrestrial basalts (e.g. Hofmann and Hart 1978) suggest that the variability of Rb/Sr in the earth's mantle today is of the order of a factor of two, and the variability of U/Pb is even less. Since these variations are dwarfed by the possible variations produced during condensation, it is more difficult to distinguish variations due to planet formation processes from those caused by the planet's internal differentiation. On the other hand, the large parent/daughter fractionations in the Rb-Sr and U-Th-Pb systems provide precise information on the time of planet formation, whereas the Sm-Nd clock is insensitive to that event (Chap. 3).

Sm and Nd are lithophile in their geochemical character. In contrast, U and Th are lithophile, but Pb is both lithophile and chalcophile. If planetary core formation involved the incorporation of sulfides into the core (Murthy and Hall 1970), large U-Th-Pb fractionations might be expected, but Sm-Nd fractionation would not be expected. These inferences, of course, are based on the near-surface properties of the elements, which might not apply to their behavior during core formation if high pressures and temperatures were involved (e.g. Lewis 1971).

In summary, Sm/Nd fractionation during planetary evolution probably occurs only as a result of magmatism in the silicate portion of the planets, and not during condensation or core formation. This is different from Rb/Sr,

Table 1.2. Concentrations (ppm) in some typical terrestrial rooks and in the silicate portion of the earth (mantle and crust)

Element	Silicate earth	MORB	Oceanic alk. basalt	Granite (G-1)	Av. Shale
Sm	0.29	3.5	8.5	8.3	5.8
Nd	0.90	10	44	55	24
Rb	0.5	1.1	30	220	140
Sr	17	130	900	250	300
U	0.018	0.10	1	13.4	3.7
Th	0.070	0.20	4	50	12
Pb	0.13	0.5	6	48	20
Lu	0.048	0.45	0.2	0.19	0.6
Hf	0.23	2.3	5	5.2	5

**Fig. 1.1.** Enrichment factors of elements important in geochronology, relative to the concentrations in average chondritic meteorites. The normalizing chondritic values used are those listed for the silicate earth (Table 1.2) divided by 1.5 except for Rb (2.5 ppm) and Pb (3.8 ppm)
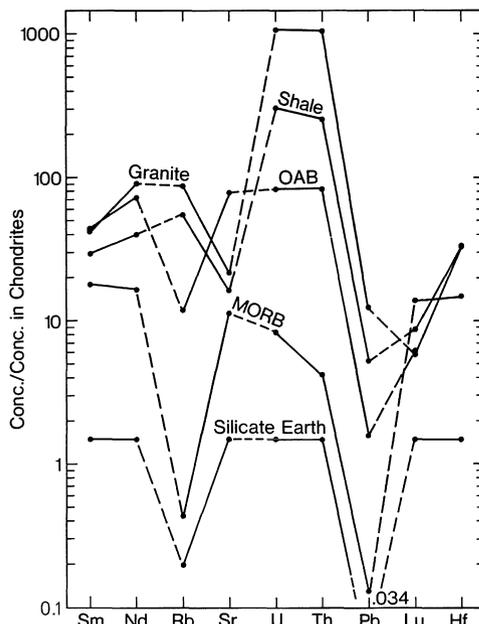
U/Pb, and Th/Pb, each of which can be affected by condensation and/or core formation. The abundances of Sm, Nd, Rb, Sr, U, Th, and Pb are given in Table 1.2 for chondrites, the estimated "bulk silicate earth" and some common rock types. The depletion of the silicate part of the earth in Rb and Pb is noteworthy. However, the Pb abundance, estimated from abundances in surface rocks and mantle samples, could be too low for the whole earth if the earth's core contains a large amount of Pb. Figure 1.1 shows the enrichment factors relative to the average chondritic abundances. Crustal rocks are highly enriched in all of these elements relative to the silicate earth, and with the exception of Pb, relative to chondrites.

1.3.2 Petrogenetic Significance of the Parent/Daughter Ratio

Because the REE differ chemically only with respect to ionic radius, which decreases regularly with increasing atomic number, the fractionation that occurs within the REE group is systematic. Some general features of rare-earth abundance patterns are illustrated in Fig. 1.2. The abundances measured in the rocks are normalized to the average abundances of chondritic meteorites and are plotted against atomic number from La (57) to Lu (71). The abundances of the light REE in rocks vary over several orders of magnitude, whereas the heavy REE show much less variability. In general, the curves are
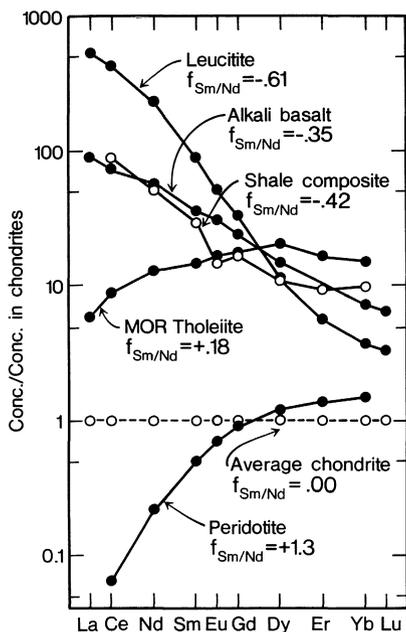
Fig. 1.2. Chondrite-normalized rare-earth element patterns of some terrestrial rock samples. The parameter $f_{Sm/Nd}$ gives the difference of the Sm/Nd ratio relative to chondrites (see Chap. 3). The rare-earth elements are listed by increasing atomic number from *left to right*. Data are from Kay and Gast (1973), Frey et al. (1971), O'Nions et al. (1979a), Haskin and Paster (1979)

smooth functions of atomic number. The pattern for a composite of North American shales, with light REE enriched relative to heavy REE, is typical of the vast majority of continental crustal rocks and is considered to be representative of continental crust (Haskin et al. 1966). The lower curve, a sample of peridotite derived from the upper mantle, has a distinctly different abundance pattern, with light REE relatively depleted, and low abundances of all rare earths. The other three patterns shown in Fig. 1.2 are from basaltic rocks. These examples demonstrate that the abundance patterns are highly variable even among rocks that are classed as "basalt". The REE patterns vary systematically with other aspects of the bulk chemistry.

Both Sm and Nd are light rare earths, and their position within the group is such that the overall abundance pattern is usually accurately reflected by the Sm/Nd ratio. Low Sm/Nd is indicative of a light REE-enriched pattern, and high Sm/Nd is characteristic of a light REE-depleted pattern (relative to heavy REE). This is an important property of Sm and Nd, because it means that information gained from Nd isotopic studies provides information about the overall REE chemistry of the parent reservoirs from which the rocks were derived (Chap. 3). This information can then be integrated with the broad base of information about the behavior of REE in petrogenetic processes to help understand both the petrogenesis of the rocks and the evolution of the parent reservoir (Chap. 4). The link between REE fractionation and igneous petrogenesis is provided by extensive studies of REE partitioning between

minerals and magma, and theoretical models of trace element fractionation during magma genesis (Schilling and Winchester 1967; Schnetzler and Philpotts 1970; Gast 1968; Kay and Gast 1973; Weill and McKay 1975; Drake and Weill 1975; Hanson 1980; Allegre and Minster 1978; DePaolo 1981d; Richter 1986).

The Rb-Sr system is similar to Sm-Nd in that the properties of Rb, an alkali element, differ from the properties of Sr, an alkaline earth, in ways that allow Rb/Sr fractionation to be interpreted in terms of petrogenetic processes that tend to separate the Group I and II elements. However, the correlation of K/Ca, Cs/Ba, and Na/Mg fractionation with Rb/Sr fractionation is only approximate at best. The most important consideration is that the behavior of Rb and Sr during magma genesis is now quite well understood as a result of experimental studies of partitioning between minerals and magma (see Irving 1978 for review and Chap. 9). The U/Pb-Th/Pb system is quite different in this regard. The geochemistry of U, Th, and Pb has been studied extensively but there are not close chemical homologues of these elements that can be used to infer the broader petrogenetic character of the fractionation processes. Furthermore, accurate determinations of partition coefficients for minerals and magma have only recently been undertaken (e.g. Benjamin et al. 1980). On the other hand, the U-Pb system has the unique advantage of a coupled pair of uranium isotopes, both of which decay to Pb isotopes. The systematics can often be used to unravel complicated geologic histories (e.g. Doe 1970; Tera and Wasserburg 1974).

1.3.3 Magnitude of Isotopic Variations

The relative chemical similarity of Sm and Nd results in rather modest separation in nature, which in turn yields only small isotopic effects. The typical range of the weight ratio of Sm/Nd in rocks is from about 0.1 to 0.5 (cf. Richard et al. 1976). Minerals generally show about the same variation, although some rare minerals, such as xenotime, may have Sm/Nd somewhat greater than one. By contrast, in the U-Th-Pb system, the strong chemical differences between the parent and daughter elements result in the very large fractionation during geologic processes. The large fractionation causes enormous variations in the isotopic composition of Pb, which permit a clear determination of the times of fractionation (e.g. Krogh 1982a,b). Rb and Sr are similar to U-Th-Pb in that they also have extremely different chemical properties and can be greatly fractionated during magmatic differentiation, metamorphism, and weathering. Rb/Sr in rocks and minerals can vary from nearly zero to 10^4 or more. Consequently, isotopic variations in Sr can be thousands of times larger than the largest possible isotopic variations in Nd.

1.3.4 Behavior in the Near-Surface Environment

An advantage of the Sm-Nd system relative to Rb-Sr and U-Th-Pb is that the rare earths are less mobile under near-surface conditions. Therefore, the systematics are less likely to be disturbed. The "mobility" of an element is a loosely defined property that depends on several factors. A particular problem with the U-Th-Pb system is that the U and the radiogenic Pb are typically concentrated in minor minerals. In contrast, the bulk of the Sm and Nd is more often contained in the major minerals of rocks. Smaller grains are generally more susceptible to leaching or diffusive loss than are the larger mineral grains of the major phases. Furthermore, the large amount of energy released by U decay tends to cause substantial damage to the crystal structure and therefore enhances diffusive loss of U and Pb. The energy released by U decay in the lifetime of a 2.7-billion-year-old rock is about 1.8 $\times 10^6$ kcal g^{-1} U or about 2000 times more than is released by ^{147}Sm decay in the same period. Since the U-bearing minor minerals can have U concentrations that are much higher than the Sm concentrations in the major minerals, the factor per unit volume can be much higher. Severe Pb loss from zircons associated with metamictization (radiation damage) has been amply demonstrated (e.g. Silver and Deutsch 1963). It is the coupled pair of radioactive U isotopes and the apparently systematic way in which Pb is lost from zircons that make them useful in geochronology.

An additional factor that contributes to mobility is the chemical incongruence of parent and daughter. Radiogenic Pb atoms must occupy structural sites in minerals that may have been chemically favorable for U atoms, but are unsuitable for the Pb atoms. These Pb atoms are probably not bound tightly in the mineral structure and therefore may be especially mobile. Furthermore, the decay of U atoms to Pb involves a large number of intermediate daughter products, some of which are gases that are particularly mobile. Similar problems exist for Rb and Sr. Although Rb decay releases relatively little energy, the extremely different chemical properties result in radiogenic ^{87}Sr atoms being located in unfavorable crystal sites. In the case of phyllosilicates, which are important in Rb-Sr dating, the Rb atoms and hence also the radiogenic ^{87}Sr atoms tend to occupy crystal sites that are easily leachable, and in some cases even exchangeable. The subtle chemical differences between Sm and Nd are probably insignificant in comparison, so a Nd daughter atom can occupy the same site as its Sm parent with little energy change. However, the radiogenic ^{143}Nd nucleus does experience a substantial recoil from the 2.23 MeV α-decay, whereas ^{87}Sr would experience essentially no recoil from the β-decay.

Mobility of atoms may also be a function of their solubility in groundwater and hydrothermal fluids that circulate through rocks. Only a small amount of data exist on this problem for REE, and there is presently only a sketchy understanding of the mechanisms of trace element redistribution during metamorphism and weathering (e.g. Fletcher and Hofmann 1974; Coller-