Introduction

This book is a cross between a textbook and a monograph, and it was started as an attempt to link depth with breadth in cosmo- and geochemistry. The need for this becomes obvious when one sees the two opposing trends in this science. On the one hand, much excellent research goes into great depth in a relatively narrow field, unnoticed except by specialists and, on the other hand, wide-ranging textbooks capture the imagination of a broader audience but cannot do justice to the actual data-gathering and interpretation. Thus, if one is interested in cosmochemistry, or the solar system or planetary formation and evolution, one can readily find a number of specific, well-written, textbooks. However, those who want to examine critically how these issues are related, and who would like to see the "big picture" and realize how it came to be, have to dive into the often rather complicated original literature.

As is the case with most branches of science, cosmochemistry and geochemistry have made huge leaps forward in the last 20 years but have become more fragmented. A bewildering amount of isotopic evidence has amassed that links Earth's history to that of the early solar system and, in turn, early solar system history to the evolution of the Galaxy and of the Universe itself. The many papers in which these data have appeared necessarily address specialized issues and although the connection to a grand unifying theme is normally made clear, there is mostly no direct contact with other specialized work that relates to the theme from another niche. This means that possible contradictions, but also cases where different angles of research have strengthened the results, may go unnoticed.

This fragmentation is not necessary, and we have felt that a "history book" describing how matter could have evolved from primordial nucleosynthesis through stellar processes, the formation of a solar nebula and planetary evolution could actually present and discuss large amounts of original data without becoming fragmented and losing sight of the big picture itself.

In pursuing this aim, we have placed much emphasis on isotope data. One reason for this is that relative isotope abundances are fingerprints of the processes in which isotopes were produced or their ratios modified. Isotope compositions of some elements serve as "stellar-thermometers" or "stellar-dosimeters" highlighting intimate features of the birth of the elements. In many cases the relationships between parent and daughter isotopes allow the time of events to be constrained, which is of prime importance if the subject is evolution. On the other hand, in most cases isotope abundance ratios have been much less disturbed than element abundances. They are therefore robust tracers of the early events that set their values. In cases where isotope abundance ratios are fractionated, their behaviour follows simple laws of nature and the resulting variations of isotope compositions help us to understand the underlying chemical and physical processes.

Another reason is that there is simply a very large amount of high-quality isotope data in the literature that combines to tell fascinating and convincing stories but is not sufficiently taken note of in textbooks. The reason for this may be that isotope-ratio interpretation is considered to be difficult and to require involved arguments. This is, however, mostly not the case. Precisely because of their lack of chemical fractionation, isotope data are the easiest geochemical results to interpret. This is why we have chosen a mainly (but not exclusively) isotopic perspective for this book.

This book is aimed at a varied readership: lecturers preparing courses for advanced undergraduate classes; graduate students; young scientists (in any branch of cosmo- or geochemistry) requiring a background in global geochemistry, particularly in its isotopic aspects; and a broader audience interested in examining the basis for our knowledge of the matter from which the Earth was built and how it formed and evolved. The book does not require a specialized knowledge of astrophysics, geology, geochemistry or isotopes: a general science background is probably enough. We have attempted to provide a coherent picture of the history of matter through time, as seen from the perspective of first astrophysics, then solar system origin and early history, including the formation of the Earth and Moon, and finally through geological time on Earth. In this effort at a continuum, we have tried to show at all stages in Earth's evolution how the particular chemical budget, or setup, that we live in, came about. Subjects that are not dealt with, as they are very well covered in many current textbooks, are the question of the origin of life or when this happened, the evolution of life, biogeochemistry and present-day environmental developments.

The book consists of four parts. Broadly, Part I deals with the principles of nucleosynthesis, the evolution of stars and episodes in which they are particularly nucleosynthetically active and the manner in which matter is conserved in interstellar space so that it can be inherited by nascent stars and solar systems. Isotopes play a large part here, first as actors and products in nucleosynthetic processes (so that their abundance ratios act as stellar thermometers and flux indicators), then (in

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the case of short-lived radioactive isotopes) as the illuminators of clouds of supernova ejecta, providing information on their nucleosynthetic processes and finally (in the case of long-lived radioactive isotopes) as clocks for the time scale of nucleosynthesis. Stellar processes also provide an interesting and unusual perspective for isotope geochemists and cosmochemists in that most decay "constants" are not constant in stellar environments. Light-stable-isotope variations in presolar grains are also covered in this chapter, as these data provide an important foundation for improved models of the nucleosynthetic processes that produced them.

In Part II the early evolution of the solar system from a disk of gas and dust to planetesimals such as chondrite and achondrite parent bodies, via coagulation, evaporation, recondensation and melting processes, is described using the available data and by modelling. In this part of the book the systematics of stable-isotope fractionation and their relevance to sources of matter and early solar system processes are described. Further, chronological techniques using both the long-lived decay systems (such as U–Pb) yielding absolute ages and the short-lived decay systems (such as Al–Mg), yielding precise relative time spans are dealt with in as much detail as is necessary. The incredibly well-constrained time scale of processes in the first 10 million years of the solar system and some minor contradictions in it are discussed.

Part III of the book concerns planetary accretion. This is first described in general terms and then specifically applied to the Earth–Moon system. The processes associated with planetary accretion, such as core formation, and the apparent paradoxes of the siderophile-element concentrations are considered together with the time scale derived from Hf–W isotope systematics. Also included are the results of new modelling of the core-formation process and the concept of a deep-seated reservoir in the Earth from which primitive noble gases still emanate today. The formation of the Moon by a giant impact is discussed along with the contrast between the ensuing terrestrial mantle-wide magma ocean, which apparently did not fractionate silicates, and the lunar magma ocean, which did. Lunar geochemical and isotope data are tied in with the terrestrial data to provide a consistent picture of the earliest history of our planet. A discussion of the constraints on the earliest atmosphere and its extensive loss is also included. This draws mainly on noble-gas abundance data, including radiogenic and fissiogenic Xe, but also considers the major atmospheric components.

In Part IV, the present-day Earth dynamics and geochemistry are reviewed, as well as the available isotopic and geochemical data base that constitutes "hard data" on the Earth's history. These include, for instance, Hf-isotope data on the oldest terrestrial (detrital) zircons and their interpretation. Present-day data yield important mass-balance considerations relating to mantle dynamics, and the total data set provides constraints for models of the geochemical evolution of the Earth's

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crust and mantle, which are described in some detail. One important question here is whether the mantle convects as a whole entity or in two layers, and another concerns the growth of the amount of continental crust and its partial recycling into the mantle through geological time. In setting up and discussing such models it is a great advantage to have the conclusions of the previous chapters immediately to hand, as these determine the initial geochemical and isotope compositions for the Earth. Further, it is a requirement for successful scenarios to satisfy the principal geochemical and isotope constraints (the Rb–Sr, Sm–Nd, Lu–Hf, U–Th–Pb and K–Ar systematics and the noble-gase abundances); one cannot be eclectic. The interaction of the different reservoirs of planet Earth with one another appears to be essential in all successful models.

Finally a world picture emerges that, in terms of chemistry and isotope compositions, traces its roots back to the very origins of the Universe. In this picture the major processes are mapped out with reasonable confidence but major problems are also highlighted.

We have made frequent use of equations in the text to illustrate points quantitatively. Equations have the advantage of not being vague. However, they usually need explaining and we have padded them in text to cover sharp edges. Systematics such as trace-element partitioning, radiogenic-isotope chronology and geochemistry and stable-isotope fractionation are explained in dedicated sections that are slotted in where they are first needed in the narrative; they are thus distributed over the book but are referred to where necessary and can be readily located using the table of contents.

Further, a comprehensive glossary is included. We have tried to avoid creating new abbreviations; it may be that “SOS” for the solar system is our only invention (which perhaps reflects our concern about what is going on with Nature). Overall we have used those abbreviations that are very frequent in the literature, such as the “H–R diagram” with “RGB and AGB stars” in it and “MORBs and OIBs” for astrophysicists and geologists respectively. Such abbreviations are explained in an appendix. There is also a list of rock and mineral names used in the text as well as a list of meteorite names and a glossary.

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Part I

The elements

In this part of the book the processes of nucleosynthesis and the environments in which they are occurring, and have occurred are sketched out.

To understand the principles of nucleosynthesis, it is important to appreciate the factors that determine the relative stability of different nuclides, and this subject is treated in Chapter 1. The grand scene is introduced in Chapter 2, without too much detail. Chapter 3 deals with data and observations concerning the chemical and isotopic composition of stars, galaxies and the solar system. This follows a broad chronological order, starting with the D/H and He/H ratios that lend support to the hypothesis of Big Bang nucleosynthesis, following through with the most primitive stellar matter and heterogeneities in presolar grains and then focussing on the composition of the solar system. Models and explanations of these data are contained in Chapters 4 to 8, which relate the data to results derived from astrophysical modelling. This helps us to understand first how the chemical elements were and are produced and second how they were scattered in space, to be incorporated in stars and solar systems that formed later.

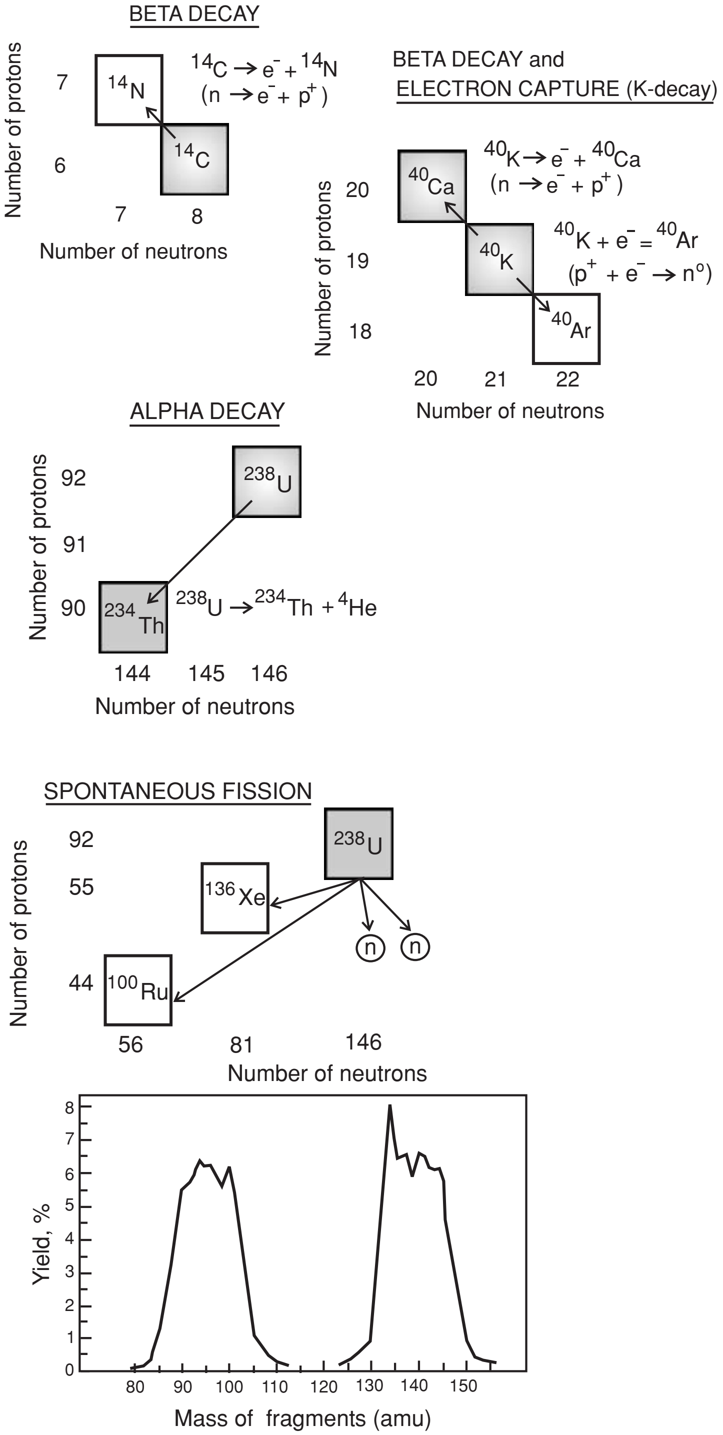
1

Isotopes: weights and abundances

1.1 Introduction: nuclei and their behaviour

Atoms are the smallest units of matter that characterize a chemical element. An atom consists of a positively charged core or nucleus and negatively charged electrons orbiting around the core. In nuclear physics, a host of different particles is known to make up atomic cores, but for the purpose of cosmochemistry and geochemistry the simplified model suffices, in which we consider just two kinds of nuclear particles (nucleons): positively charged protons, p, and neutral neutrons, n. For a neutral atom the number of protons in the core, Z (the atomic number), is equal to the number of electrons around it. As Z determines the electron configuration and therefore the chemical behaviour, a family of atoms of equal Z constitutes a chemical element. Such a family generally includes nuclei with a varying number of neutrons, N. The atomic mass number $A = Z + N$, the total number of nucleons, then varies accordingly. Atoms of an element that have different values of N (and therefore A) are called isotopes, a term with Greek roots indicating that these different nuclides occupy the same position in the periodic table. The lightest element, hydrogen, includes three isotopes, ^{1}H, ^{2}H (D) and ^{3}H, having 0, 1 and 2 neutrons in the core, respectively. Most elements consist of a larger number of isotopes; therefore the approximately 100 currently known elements include approximately 1000 isotopes.

Many isotopes exist indefinitely, at least in normal conditions, and these are known as stable isotopes, S. The nuclei of the great majority of isotopes are, however, not stable and can spontaneously decay, i.e. turn into other nuclei, by emitting or absorbing a particle as summarized in Fig. 1.1. These decaying isotopes are termed radioactive or parent isotopes, ^{r}R, and the decay products are radiogenic or daughter isotopes, ^{r}D. Generally after decay an excited daughter nucleus "cools down", emitting γ-rays (high-frequency electromagnetic radiation). Each radioactive isotope species has its own specific rate of decay, λ, known as the


BETA DECAY
Number of protons
7
6
14N
14C
$^{14}C \rightarrow e^- + {}^{14}N$
$(n \rightarrow e^- + p^+)$
7
8
Number of neutrons
BETA DECAY and
ELECTRON CAPTURE (K-decay)
$^{40}K \rightarrow e^- + {}^{40}Ca$
$(n \rightarrow e^- + p^+)$
$^{40}K + e^- = {}^{40}Ar$
$(p^+ + e^- \rightarrow n^o)$
Number of protons
20
19
18
40Ca
40K
40Ar
20
21
22
Number of neutrons
ALPHA DECAY
Number of protons
92
91
90
238U
234Th
$^{238}U \rightarrow {}^{234}Th + {}^4He$
144
145
146
Number of neutrons
SPONTANEOUS FISSION
Number of protons
92
55
44
238U
136Xe
100Ru
n
n
56
81
146
Number of neutrons
Yield, %
0
1
2
3
4
5
6
7
8
80
90
100
110
120
130
140
150
Mass of fragments (amu)

decay constant; if R is the number of radioactive atoms then the decay is described by

$$\mathrm{d}R/\mathrm{d}t = -\lambda R. \tag{1.1}$$

The solution of Eqn (1.1) gives

$$R = R_0 \mathrm{e}^{-\lambda t} \equiv R_0 \exp(-\lambda t), \tag{1.2}$$

where t is the time elapsed since some time t_0 in the past and $R_0 \equiv R(t_0)$. Commonly, the decay is also characterized by the time interval τ during which the number of atoms R decreases by a factor 2; this is the half-life of the isotope. As $R(\tau) = R_0/2$, the relation between the decay constant and the half-life τ of a radioactive nuclide is $\tau \equiv \ln 2/\lambda$. The mean life of a radioactive isotope is $1/\lambda = \tau \ln 2$. Some radioactive isotopes decay by more than one mechanism, producing different daughter nuclides; for example ^{40}K can decay into ^{40}Ca (with corresponding λ_{Ca}) or into ^{40}Ar (λ_{Ar}), so that the total decay rate is $\lambda_{40} \equiv \lambda_{\mathrm{Ca}} + \lambda_{\mathrm{Ar}}$ and the number of $^{40}\mathrm{Ar}^*$ atoms generated by ^{40}K decay during time t equals $(\lambda_{\mathrm{Ar}}/\lambda_{40})\ ^{40}\mathrm{K} \exp(-\lambda_{40} t)$. In some cases decay competes with nuclear reactions (Section 5.4). The general term for such situations is branching.

It should be noted that the term “decay constant” does not apply to stellar environments, where β-decay rates can vary by orders of magnitude owing to the extreme temperatures and pressures. These variations, when known, shed light on nucleosynthetic processes (see for example Section 5.4). For planetary conditions λ values are constant, with some rare exceptions; for instance, the λ_3 value for ^{3}H β-decay is measurably dependent on the chemical state of hydrogen (Akulov and Mamyrin, 2004) and the value for ^{7}Be increases with pressure, by about 1% at 40 GPa (Liu and Huh, 2000).

←

Fig. 1.1 Radioactive decay and fission.
Top left, β-decay: a neutron n in the nuclei of carbon-14 decays to a proton p^+ and electron e^-, which is then emitted leaving behind nitrogen-14.
Top right, e-capture: a proton in the nucleus of ^{40}K captures an electron from the innermost orbit to produce a neutron and the nucleus of ^{40}Ar. Potassium-40 nuclei also decay via β-emission. Middle, α-decay: a nucleus of the heavy radioactive element ^{238}U emits an α-particle consisting of two protons and two neutrons; the resulting isotope is ^{234}Th. Bottom, nuclear fission: the nucleus of ^{238}U disintegrates into two fragments (generally the mass ratio is $\sim 1/2$) and emits two to three neutrons. As the fragments initially have too many neutrons relative to protons (for the given mass range), β-decay occurs until the “stability valley” (Fig. 1.3) is reached. When short-lived heavy isotopes ($A \sim 260$) exhibit fission, the fragment mass ratio approaches 1.

Nuclei can be modified not only by spontaneous decay but also by nucleus–particle (or nucleus–γ) interactions known as nuclear reactions. These can be destructive (breaking nuclei up) or constructive (building heavier nuclei). The interaction of nuclei with other nuclei or with protons is impossible at low temperatures, as both are positively charged. However, at $T \sim 10^7$ K or higher temperatures, this "Coulomb barrier" can be overcome: nuclei can collide and fuse, which is the basis for the existence of all nuclides other than the proton, ^{1}H.

Neutrons can easily penetrate nuclei even at low temperatures. For instance,[1] neutron capture by ^{56}Fe(n, γ)^{57}Fe and ^{57}Fe(n, γ)^{58}Fe gives rise to heavier iron isotopes. Further n-capture, ^{58}Fe(n, γ)^{59}Fe, followed by β-decay yields the next element, cobalt: $^{59}\mathrm{Fe} \rightarrow \beta^- \rightarrow {}^{59}\mathrm{Co}$. Such n-capture and associated β-decay has produced all the elements beyond Fe. These are therefore called n-capture elements. An example of a destructive nuclear reaction is the nuclear fission of ^{235}U: after neutron capture, ^{235}U disintegrates into two heavy fragments with different masses and a few neutrons (Fig. 1.1). Its heaviest brother, ^{238}U, exhibits spontaneous fission in addition to α-decay, but with a much lower probability. Another important example is ^{6}Li(n, α)^{3}H: this reaction produces radioactive ^{3}H (tritium), which β-decays into daughter ^{3}He.

Investigations of the heaviest nuclei have shown that the heavier a nucleus is, the higher the probability that it will disintegrate via fission. Extrapolation of the relationships between Z, A and the fission rate suggests a limit of $Z \sim 120$, $A \sim 310$ for possible nuclei. Thus, the full range of the elements extends from hydrogen (1 amu) to an, as yet unknown, superheavy element (~ 300 amu).

1.2 Atomic nuclei and binding energy, with some predictions on isotope abundances

Mass, energy and binding energy

The atomic nuclei are quite small: the radius r_A of a nucleus with atomic mass number A is about $1.4 \times 10^{-13} A^{1/3}$ cm. Thus, for the heaviest possible nuclides, $r_A \sim 10^{-12}$ cm. The shape of atomic nuclei varies between spheroidal and ellipsoidal. The whole atom, i.e. the nucleus plus the electronic cloud, is a factor $\sim 10^5$ larger. For example, the radius of the first electronic orbit of the hydrogen atom is 0.53×10^{-8} cm. However, the nucleus makes up almost all the mass of an atom. Generally this mass is measured in so-called atomic mass units, defined as 1/12 of the mass of the neutral isotope ^{12}C; i.e. 1 amu $\equiv 1.660\,53 \times 10^{-24}$ g. Thus the mass of an atom in amu is numerically $\approx A$, the atomic mass number. The precise masses of the proton, $M_\mathrm{p} = 1.007\,282\,6$ amu, and neutron, $M_\mathrm{n} = 1.008\,671\,3$ amu, are larger

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[1] The following notation abbreviates $^{56}\mathrm{Fe} + \mathrm{n} \rightarrow {}^{57}\mathrm{Fe} + \gamma$ etc.

by a factor of about 2×10^3 than the mass of the electron, $m_e = 0.000\,548\,58$ amu. The nuclear masses and radii (e.g. $238 \times 1.66 \times 10^{-24}$ g corresponds to $\sim 10^{-12}$ cm) allow the density of nuclear matter to be estimated at $\sim 10^{14}$ g cm^{-3}.

High-resolution mass spectrometry allows the isotope masses $M(A, Z)$ to be obtained precisely. These masses are without exception smaller than the sum of the masses of the constituent particles, protons + neutrons + electrons:

$$[Zm_p + (A - Z)M_n + ZM_e] - M(A, Z) = \Delta M > 0. \tag{1.3}$$

Note that the measured $M(A, Z)$ also includes Zm_e, so that ΔM is the difference in nuclear mass. From this, the binding energy of nuclei can be calculated. According to the Einstein relationship,

$$E = Mc^2 \tag{1.4}$$

where E is the energy in ergs; $c = 3 \times 10^{10}$ cm s^{-1} is the light velocity in vacuum and M is the relativistic mass in g: $M = M_0/\sqrt{}(1 - (v/c)^2)$, where M_0 is the rest mass and v is the velocity of the body. One atomic mass unit is thus equivalent by (1.4) to the energy 1.49×10^{-3} erg or 0.932×10^9 eV = 932 MeV (1 MeV $\equiv 1.60 \times 10^{-6}$ erg). Substituting ΔM from Eqn (1.3) into Eqn (1.4) gives the total binding energy ΔW of a nucleus,

$$\Delta W = \Delta Mc^2, \tag{1.5}$$

and the binding energy per nucleon for that nucleus,

$$\varepsilon = \Delta W/A. \tag{1.6}$$

A comparison of the mass of deuterium, 2.014 74 amu, with the total mass of its constituent proton and neutron, 2.017 12 amu, gives $\Delta M = 0.0024$ amu, $\Delta W =$ 2.2 MeV and $\varepsilon = 1.1$ MeV nucleon^{-1}. This is the energy yield from deuterium nucleosynthesis. Conversely, a neutron is heavier than a proton by about 1 MeV and readily decays, when in isolation, producing a proton, electron and neutrino. A similar estimate for the ^{4}He nucleus gives $\Delta W = 28$ MeV and $\varepsilon = 7$ MeV nucleon^{-1}.

It is instructive to compare nuclear energy values with those for chemical interactions, say, that required to separate an electron from a hydrogen atom. The total energy of an electron having a charge $e = -1.6 \times 10^{-19}$ C and orbiting the nucleus at a distance $r = 0.53 \times 10^{-8}$ cm is the sum of its kinetic and potential energies:

$$E = \frac{mv^2}{2} - \frac{e^2}{4\pi\epsilon r} = \frac{-e^2}{8\pi\epsilon r},$$