

# Chapter 1

## Atoms and nuclei: their physics and origins

### 1.1 INTRODUCTION

Isotope geochemistry has grown over the last 50 years to become one of the most important fields in the earth sciences as well as in geochemistry. It has two broad subdivisions: *radiogenic isotope geochemistry* and *stable isotope geochemistry*. These subdivisions reflect the two primary reasons why the relative abundances of isotopes of some elements vary in nature: radioactive decay and chemical fractionation.<sup>1</sup> One might recognize a third subdivision: cosmogenic isotope geochemistry, in which both radioactive decay and chemical fractionation are involved, but additional nuclear processes can be involved as well.

The growth in the importance of isotope geochemistry reflects its remarkable success in attacking fundamental problems of earth science, as well as problems in astrophysics, physics, and biology (including medicine). Isotope geochemistry has played an important role in transforming geology from a qualitative, observational science to a modern quantitative one. To appreciate the point, consider the Ice Ages, a phenomenon that has fascinated geologist and layman alike for more than 150 years. The idea that much of the Northern Hemisphere was once covered by glaciers was first advanced by Swiss zoologist Louis Agassiz in 1837. His theory was based on observations of geomorphology and modern glaciers. Over the next 100 years, this theory advanced very little,

other than the discovery that there had been more than one ice advance. No one knew exactly when these advances had occurred, how long they lasted, or why they occurred. Stable and radiogenic isotopic studies in the last 50 years have determined the exact times of these ice ages and the exact extent of temperature change (about 3°C or so in temperate latitudes, more at the poles). Knowing the timing of these glaciations has allowed us to conclude that variations in the Earth's orbital parameters (the Milankovitch parameters) and resulting changes in insolation have been the direct cause of these ice ages. Comparing isotopically determined temperatures with CO<sub>2</sub> concentrations in bubbles in carefully dated ice cores leads to the hypothesis that atmospheric CO<sub>2</sub> plays an important role in amplifying changes in insolation. Careful U-Th dating of corals has also revealed the detailed timing of the melting of the ice sheet and consequent sea level rise. Comparing this with stable isotope geothermometry shows that melting lagged warming (not too surprisingly). Other isotopic studies revealed changes in the ocean circulation system as the last ice age ended. Changes in ocean circulation may also be an important feedback mechanism affecting climate. Twenty-five years ago, all this seemed very interesting, but not very relevant. Today, it provides us with critical insights into how the planet's climate system works. With the current concern over potential global warming and greenhouse gases, this information is extremely "relevant".

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Some isotope geochemistry even seeps into public consciousness through its application to archeology and forensics. For example, a recent *National Geographic* television documentary described how carbon-14 dating of 54 beheaded skeletons in a mass grave in Dorset, England revealed they were tenth century and how strontium and oxygen isotope ratios revealed they were those of Vikings executed by Anglo-Saxons and not visa versa, as originally suspected. Forensic isotopic analysis gets occasional mention in both in shows like *CSI: Crime Scene Investigation* and in newspaper reporting of real crime investigations.

Other examples of the impact of isotope geochemistry would include such diverse topics as ore genesis, mantle dynamics, hydrology, and hydrocarbon migration, monitors of the cosmic ray flux, crustal evolution, volcanology, oceanic circulation, environmental protection and monitoring, and paleontology. Indeed, there are few, if any, areas of geological inquiry where isotopic studies have not had a significant impact.

One of the first applications of isotope geochemistry remains one of the most important: geochronology and cosmochronology: the determination of the timing of events in the history of the Earth and the Solar System. The first “date” was obtained in 1907 by Bertram Boltwood, a Yale University chemist, who determined the age of uranium ore samples by measuring the amount of the radiogenic daughter of U, namely Pb, present. Other early applications include determining the abundance of isotopes in nature to constrain models of the nucleus and of nucleosynthesis (the origin of the elements). Work on the latter problem still proceeds. The origins of stable isotope geochemistry date to the work of Harold Urey and his colleagues in the 1940s. Paleothermometry was one of the first applications of stable isotope geochemistry as it was Urey who recognized the potential of stable isotope geochemistry to solving the riddle of the Ice Ages.

This book will touch on many, though not all, of these applications. We’ll focus first on geochronology and then consider how radiogenic isotopes have been used to understand the origin and evolution of the Earth. Next, we consider the fundamental principles underlying stable isotope geochemistry and then

examine its applications to fields as diverse as paleoclimate, paleontology, archeology, ore genesis, and magmatic evolution. In the final chapters, we’ll see how the horizons of stable isotope geochemistry have broadened from a few light elements such as hydrogen, carbon, and oxygen to much of the periodic table. Finally, we examine the isotope geochemistry of the noble gases, whose isotopic variations are due to both nuclear and chemical processes and provide special insights into the origins and behavior of the Earth.

Before discussing applications, however, we must build a firm basis in the nuclear physics. We’ll do that in the following sections. With that basis, in the final sections of this chapter we’ll learn how the elements have been created over the history of the Universe in a variety of cosmic environments.

### 1.2 PHYSICS OF THE NUCLEUS

#### 1.2.1 Early development of atomic and the nuclear theory

John Dalton, an English schoolteacher, first proposed that all matter consists of atoms in 1806. William Prout found that atomic weights were integral multiples of the mass of hydrogen in 1815, something known as the *Law of Constant Proportions*. This observation was strong support for the atomic theory, though it was subsequently shown to be only approximate, at best. J. J. Thomson of the Cavendish Laboratory in Cambridge developed the first mass spectrograph in 1906 and showed why the *Law of Constant Proportions* did not always hold: those elements not having integer weights had several *isotopes*, each of which had mass that was an integral multiple of the mass of H. In the meantime, Rutherford, also of Cavendish, had made another important observation: that atoms consisted mostly of empty space. This led to Niels Bohr’s model of the atom, proposed in 1910, which stated that the atom consisted of a nucleus, which contained most of the mass, and electrons in orbit about it.

It was nevertheless unclear why some atoms had different masses than other atoms of the same element. The answer was provided by W. Bothe and H. Becker of Germany and James Chadwick of England: the neutron. Bothe and Becker discovered the particle, but mistook it for radiation. Chadwick won the Nobel

Prize for determining the mass of the neutron in 1932. Various other experiments showed the neutron could be emitted and absorbed by nuclei, so it became clear that differing numbers of neutrons caused some atoms to be heavier than other atoms of the same element. This bit of history leads to our first basic observation about the nucleus: it consists of protons and neutrons.

### 1.2.2 Some definitions and units

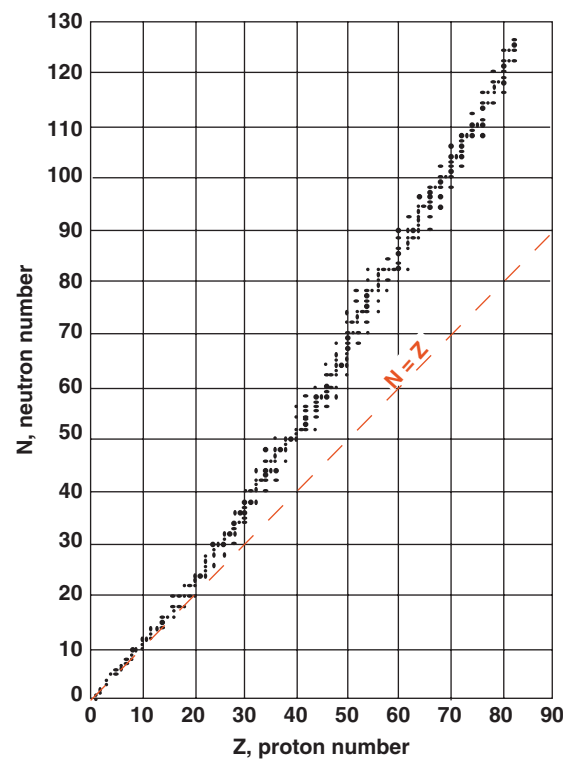
Before we consider the nucleus in more detail, let's set out some definitions:  $N$ : the number of neutrons,  $Z$ : the number of protons (same as atomic number since the number of protons dictates the chemical properties of the atom),  $A$ : Mass number ( $N + Z$ ),  $M$ : Atomic Mass,  $I$ : Neutron excess number ( $I = N - Z$ ). *Isotopes* have the same number of protons but different numbers of neutrons; *isobars* have the same mass number ( $N + Z$ ); *isotones* have the same number of neutrons but different number of protons.

The basic unit of nuclear mass is the unified atomic mass unit (also known as the *dalton* and the atomic mass unit or *amu*), which is based on  $^{12}\text{C} \equiv 12$  unified atomic mass units; that is, the mass of  $^{12}\text{C}$  is 12 unified atomic mass units (abbreviated  $u$ ). The masses of atomic particles are:

$$\begin{aligned} \text{proton: } 1.007276467 \text{ u} &= 1.67262178 \\ &\times 10^{-27} \text{ kg} = 938.2720 \text{ MeV}/c^2 \\ \text{neutron } 1.008664916 \text{ u} \\ \text{electron } 0.0005485799 \text{ u} &= 9.10938291 \\ &\times 10^{-31} \text{ kg} = 0.5109989 \text{ MeV}/c^2 \end{aligned}$$

### 1.2.3 Nucleons, nuclei, and nuclear forces

Figure 1.1 is a plot of  $N$  versus  $Z$  showing which nuclides are stable. A key observation in understanding the nucleus is that not all combinations of  $N$  and  $Z$  result in stable nuclides. In other words, we cannot simply throw protons and neutrons (collectively termed nucleons) together randomly and expect them to form a nucleus. For some combinations of  $N$  and  $Z$ , a nucleus forms but is unstable, with half-lives from  $>10^{15}$  yrs to  $<10^{-12}$  sec. A relative few combinations of  $N$  and  $Z$  result in stable nuclei. Interestingly, these stable nuclei generally have  $N \approx Z$ , as Figure 1.1 shows. Notice also that for small  $A$ ,  $N = Z$ , for large  $A$ ,  $N > Z$ . This is another important

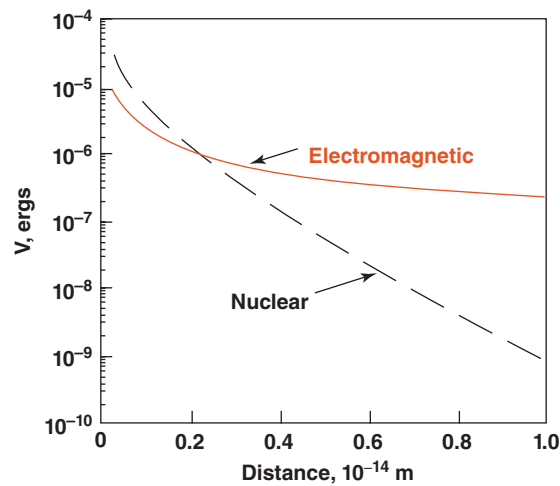


**Figure 1.1** Neutron number versus proton number for stable nuclides. (Source: White (2013). Reproduced with permission of John Wiley & Sons.)

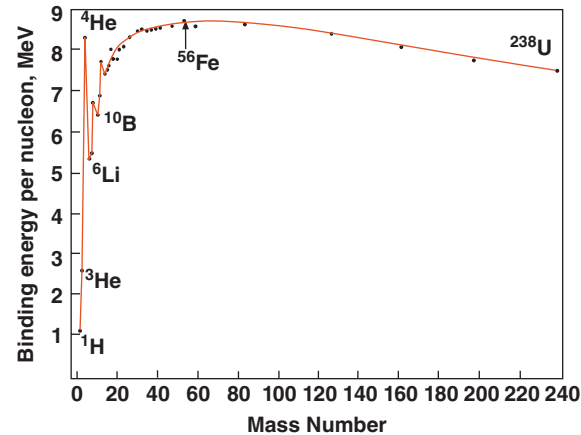
observation that will lead us to the first model of the nucleus.

A significant portion (about half) of the nucleus consists of protons, which obviously tend to repel each other by coulombic (electrostatic) force. From the observation that nuclei exist at all, it is apparent that another force must exist that is stronger than coulomb repulsion at short distances. It must be negligible at larger distances; otherwise all matter would collapse into a single nucleus. This force, called the *nuclear force*, is a manifestation of one of the fundamental forces of nature (or a manifestation of the single force in nature if you prefer unifying theories), called the *strong force*. If this force is assigned a strength of 1, then the strengths of other forces are: electromagnetic  $10^{-2}$ ; weak force (which we'll discuss later)  $10^{-5}$ ; gravity  $10^{-39}$ . Just as electromagnetic forces are mediated by a particle, the photon, the nuclear force is mediated by the *pion*. The photon carries one quantum of electromagnetic force field; the pion carries

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**Figure 1.2** The nuclear and electromagnetic potential of a proton as a function of distance from the proton.



**Figure 1.3** Binding energy per nucleon versus mass number. (Source: White (2013). Reproduced with permission of John Wiley & Sons.)

one quantum of nuclear force field. A comparison of the relative strengths of the nuclear and electromagnetic forces as a function of distance is shown in Figure 1.2.

#### 1.2.4 Atomic masses and binding energies

The carbon-12 atom consists of six neutrons, six protons, and six electrons. But using the masses listed here, we find that the masses of these 18 particles sum to more than 12 u, the mass of  $^{12}\text{C}$  atom. There is no mistake, they do not add up. What has happened to the extra mass? The mass has been converted to the energy binding the nucleons. It is a general physical principle that the lowest energy configuration is the most stable. We would expect that if  $^4\text{He}$  is stable relative to two free neutrons and two free protons,  $^4\text{He}$  must be a lower energy state compared to the free particles. If this is the case, then we can predict from Einstein's mass-energy equivalence:

$$E = mc^2 \quad (1.1)$$

that the mass of the helium nucleus is less than the sum of its constituents. We define the *mass decrement* of an atom as:

$$\delta = W - M \quad (1.2)$$

where  $W$  is the sum of the mass of the constituent particles and  $M$  is the actual mass of

the atom. For example,  $W$  for  $^4\text{He}$  is  $W = 2m_p + 2m_n + 2m_e = 4.03298\text{u}$ . The mass of  $^4\text{He}$  is  $4.002603\text{u}$ , so  $\delta = 0.030377\text{u}$ . Converting this to energy using Eqn. 1.1 yields  $28.28\text{ MeV}$ . This energy is known as the *binding energy*. Dividing by  $A$ , the mass number, or number of nucleons, gives the *binding energy per nucleon*,  $E_b$ :

$$E_b = \left[ \frac{W - M}{A} \right] c^2 \quad (1.3)$$

This is a measure of nuclear stability: those nuclei with the largest binding energy per nucleon are the most stable. Figure 1.3 shows  $E_b$  as a function of mass. Note that the nucleons of intermediate mass tend to be the most stable. This distribution of binding energy is important to the life history of stars, the abundances of the elements, and radioactive decay, as we shall see.

Some indication of the relative strength of the nuclear binding force can be obtained by comparing the mass decrement associated with it to that associated with binding an electron to a proton in a hydrogen atom. The mass decrement we calculated previously for He is of the order of 1%, one part in  $10^2$ . The mass decrement associated with binding an electron to a nucleus of the order of one part in  $10^8$ . So, bonds between nucleons are about  $10^6$  times stronger than bonds between electrons and nuclei.

### Pions and the nuclear force

As we noted, we can make an *a priori* guess as to two of the properties of the nuclear force: it must be very strong and it must have a very short range. Since neutrons as well as protons are subject to the nuclear force, we may also conclude that it is not electromagnetic in nature. What inferences can we make on the nature of the force and the particle that mediates it? Will this particle have a mass, or be massless like the photon?

All particles, whether they have mass or not, can be described as waves, according to quantum theory. The relationship between the wave properties and the particle properties is given by the *de Broglie Equation*:

$$\lambda = \frac{h}{p} \quad (1.4)$$

where  $h$  is Planck's constant,  $\lambda$  is the wavelength, called the *de Broglie wavelength*, and  $p$  is momentum. Eqn. 1.2 can be rewritten as:

$$\lambda = \frac{h}{mv} \quad (1.5)$$

where  $m$  is mass (relativistic mass, not rest mass) and  $v$  is velocity. From this relation we see that mass and de Broglie wavelength are inversely related: massive particles will have very short wavelengths.

The wavefunction associated with the particle may be written as:

$$\frac{1}{c^2} \frac{\partial^2 \psi}{\partial t^2} - \nabla^2 \psi(x, t) = -\left(\frac{mc}{\hbar}\right)^2 \psi(x, t) \quad (1.6)$$

where  $\nabla^2$  is simply the Laplace operator:

$$\nabla^2 \equiv \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} + \frac{\partial^2}{\partial z^2}$$

The square of the wavefunction,  $\psi^2$ , describes the probability of the particle being found at some point in space  $x$  and some time  $t$ . In the case of the pion, the wave equation also describes the strength of the nuclear force associated with it.

Let us consider the particularly simple case of a time-independent, spherically symmetric solution to Eqn. 1.4 that could describe the pion field outside a nucleon located at the origin. The solution will be a potential function  $V(r)$ , where  $r$  is radial distance from the origin and  $V$  is the strength of the field. The condition of time-independence means that the first term on the left will be 0, so the equation assumes the form:

$$\nabla^2 V(r) = -\left(\frac{mc}{\hbar}\right)^2 V(r) \quad (1.7)$$

$r$  is related to  $x$ ,  $y$ , and  $z$  as:

$$r = \sqrt{x^2 + y^2 + z^2} \quad \text{and} \quad \frac{\partial r}{\partial x} = \frac{x}{r}$$

Using this relationship and a little mathematical manipulation, the Laplace operator in 1.7 becomes:

$$\nabla^2 V(r) = \frac{1}{r^2} \frac{d}{dr} \left( r^2 \frac{dV(r)}{dr} \right) \quad (1.8)$$

and 1.7 becomes:

$$\frac{1}{r^2} \frac{d}{dr} \left( r^2 \frac{dV(r)}{dr} \right) = -\left(\frac{mc}{\hbar}\right)^2 V(r)$$



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Two possible solutions to this equation are:

$$\frac{1}{r} \exp\left(-r \frac{mc}{\hbar}\right) \quad \text{and} \quad \frac{1}{r} \exp\left(+r \frac{mc}{\hbar}\right)$$

The second solution corresponds to a force increasing to infinity at infinite distance from the source, which is physically unreasonable, thus only the first solution is physically meaningful. Our solution, therefore, for the nuclear force is

$$V(r) = \frac{C}{r} \exp\left(-r \frac{mc}{\hbar}\right) \quad (1.9)$$

where  $C$  is a constant related to the strength of the force. The term  $mc/\hbar$  has units of  $\text{length}^{-1}$ . It is a constant that describes the effective range of the force. This effective range is about  $1.4 \times 10^{-13} \text{ cm}$ . This implies a mass of the pion of about 0.15 u. It is interesting to note that for a massless particle, equation 1.7 reduces to

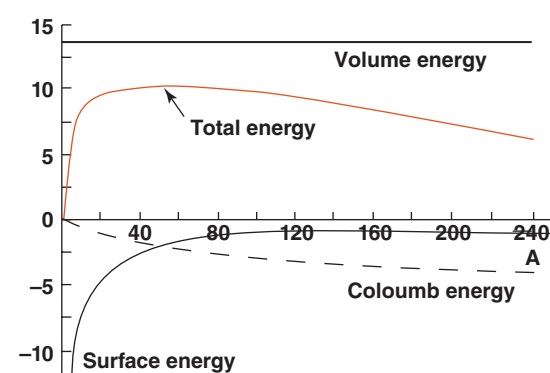
$$V(r) = \frac{C}{r} \quad (1.10)$$

which is just the form of the potential field for the electromagnetic force. Thus, both the nuclear force and the electromagnetic force satisfy the same general Eqn. 1.9. Because pion has mass while the photon does not, the nuclear force has a very much shorter range than the electromagnetic force.

A simple calculation shows how the nuclear potential and the electromagnetic potential will vary with distance. The magnitude for the nuclear potential constant  $C$  is about  $10^{-18} \text{ erg-cm}$ . The constant  $C$  in Eqn. 1.10 for the electromagnetic force is  $e^2$  (where  $e$  is the charge on the electron) and has a value of  $2.3 \times 10^{-19} \text{ erg-cm}$ . Using these values, we can calculate how each potential will vary with distance. This is just how Figure 1.2 was produced.

## 1.2.5 The liquid-drop model

Why are some combinations of  $N$  and  $Z$  more stable than others? The answer has to do with the forces between nucleons and how nucleons are organized within the nucleus. The structure and organization of the nucleus are questions still being actively researched in physics, and full treatment is certainly beyond the scope of this text, but we can gain some valuable insight to nuclear stability by considering two of the simplest models of nuclear structure. The simplest model of the nucleus is the *liquid-drop model*, proposed by Niels Bohr in 1936. This model assumes all nucleons in a nucleus have equivalent states. As its name suggests, the model treats the binding between nucleons as similar to the binding between molecules in a liquid drop. According to the liquid-drop model, the total binding energy of nucleons is influenced by four effects: a volume energy, a surface energy, an excess neutron energy, and a coulomb energy. The variation of three of these forces with mass number and their total effect is shown in Figure 1.4.



**Figure 1.4** Variation of surface, coulomb, and volume energy per nucleon versus mass number. (Source: White (2013). Reproduced with permission of John Wiley & Sons.)

In the liquid-drop model, the binding energy is given as a function of mass number,  $A$ , and neutron excess number,  $I (= N - Z)$ , as:

$$B(A, I) = a_1 A - a_2 A^{2/3} - a_3 I^2 / 4A - a_4 Z^2 / A^{1/3} + \delta \quad (1.11)$$

where:

- $a_1$ : heat of condensation (volume energy  $\propto A$ ) = 14 MeV
- $a_2$ : surface tension energy = 13 MeV
- $a_3$ : excess neutron energy = 18.1 MeV
- $a_4$ : coulomb energy = 0.58 MeV
- $\delta$ : even-odd fudge factor. Binding energy greatest for even-even and smallest for odd-odd.

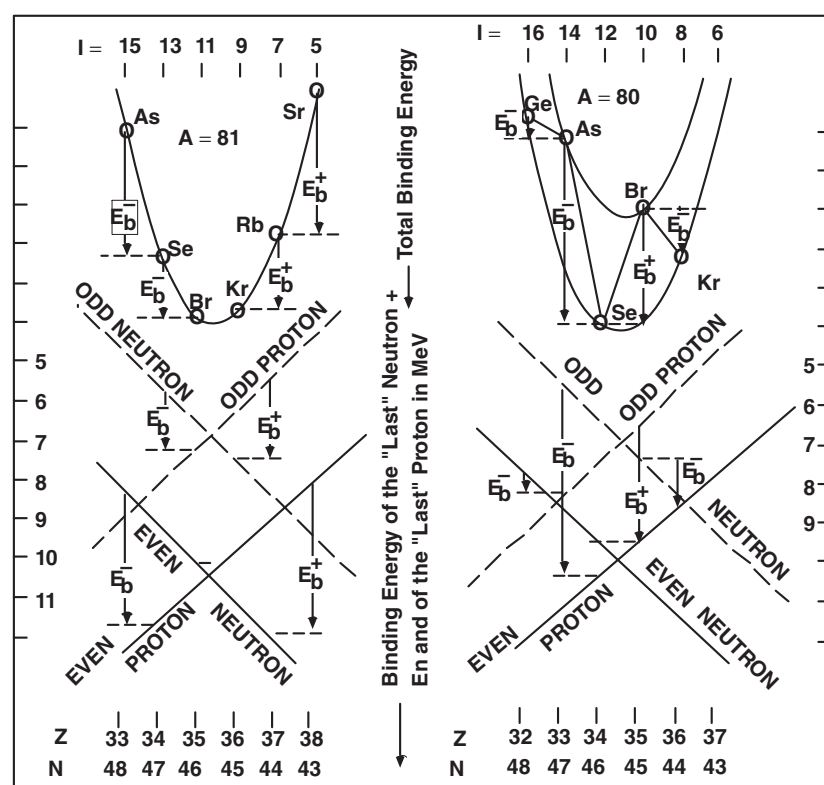
Some of the nuclear stability rules here can be deduced from Eqn. 1.11. Solutions for Eqn. 1.11 at constant  $A$ , that is, for isobars, result in a hyperbolic function of  $I$  as illustrated in Figure 1.5. In other words, for a given number of protons, there are an optimal number of neutrons: either too many or too few results in a higher energy state. For odd  $A$ , one nuclei will lie at or near the bottom of this function (energy well). For even  $A$ , two curves result, one for odd-odd, and one

for even-even. The even-even curve will be the one with the lower (more stable) one. As we shall see, nuclei with either too many or too few neutrons tend to decay to nuclide with the optimal number of neutrons.

### 1.2.6 The shell model of the nucleus

#### 1.2.6.1 Odd-even effects, magic numbers, and shells

Something that we have alluded to and which the liquid-drop model does not explain is the even-odd effect, illustrated in Table 1.1. Clearly, even combinations of nuclides are much more likely to be stable than odd ones. This is the first indication that the liquid-drop model does not provide a complete description of nuclear stability. Another observation not explained by the liquid-drop model are the so-called *Magic Numbers*. The *Magic*



**Figure 1.5** Graphical illustration of total binding energies of the isobars of mass number  $A = 81$  (left) and  $A = 80$  (right). Energy values lie on parabolas, a single parabola for odd  $A$  and two parabolas for even  $A$ . Binding energies of the 'last' proton and 'last' neutrons are approximated by the straight lines in the lower part of the figure. (Source: Suess (1987). Reproduced with permission of John Wiley & Sons.)

Table 1.1 Numbers of stable nuclei for odd and even Z and N.

Z	N	A	Number of stable nuclei	Number of very long-lived nuclei
		(Z + N)		
odd	odd	even	4	5
odd	even	odd	50	3
even	odd	odd	55	3
even	even	even	165	11

Numbers are 2, 8, 20, 28, 50, 82, and 126. Some observations about magic numbers:

1. Isotopes and isotones with magic numbers are unusually common (i.e., there are a lot of different nuclides in cases where N or Z equals a magic number).
2. Magic number nuclides are unusually abundant in nature (high concentration of the nuclides).
3. Delayed neutron emission in occurs in fission product nuclei containing  $N^* + 1$  (where  $N^*$  denotes a magic number) neutrons.
4. Heaviest stable nuclides occur at  $N = 126$  (and  $Z = 83$ ).
5. Binding energy of last neutron or proton drops for  $N^* + 1$ .
6. Neutron-capture cross sections for magic numbers are anomalously low.
7. Nuclear properties (spin, magnetic moment, electrical quadrupole moment, metastable isomeric states) change when a magic number is reached.

The electromagnetic spectra emitted by electrons are the principal means of investigating the electronic structure of the atom. By analogy, we would expect that the electromagnetic spectra of the nucleus should yield clues to its structure, and indeed it does. However, the  $\gamma$  spectra of nuclei are so complex that not much progress has been made interpreting it. Observations of magnetic moment and spin of the nucleus have been more useful (nuclear magnetic moment is also the basis of the nuclear magnetic resonance, or NMR, technique, used to investigate relations between atoms in lattices and the medical diagnostic technique nuclear magnetic imaging).

Nuclei with magic numbers of protons or neutrons are particularly stable or “unreactive.” This is clearly analogous to chemical

properties of atoms: atoms with filled electronic shells (the noble gases) are particularly unreactive. In addition, just as the chemical properties of an atom are largely dictated by the “last” valence electron, properties such as the nucleus’s angular momentum and magnetic moment can often be accounted for primarily by the “last” odd nucleon. These observations suggest the nucleus may have a shell structure similar to the electronic shell structure of atoms, and leads to the shell model of the nucleus.

In the shell model of the nucleus, the same general principles apply as to the shell model of the atom: possible states for particles are given by solutions to the Schrödinger Equation. Solutions to this equation, together with the Pauli Exclusion Principle, which states that no two particles can have exactly the same set of quantum numbers, determine how many nucleons may occur in each shell. In the shell model, there are separate systems of shells for neutrons and protons. As do electrons, protons and neutrons have intrinsic angular momentum, called *spin*, which is equal to  $\frac{1}{2}\hbar$  ( $\hbar = h/2\pi$ , where  $h$  is Planck’s constant and has units of momentum,  $h = 6.626 \times 10^{-34}$  joule-sec). The total nuclear angular momentum, somewhat misleadingly called the nuclear spin, is the sum of (1) the intrinsic angular momentum of protons, (2) the intrinsic angular momentum of neutrons, and (3) the orbital angular momentum of nucleons arising from their motion in the nucleus. Possible values for orbital angular momentum are given by  $\ell$ , the orbital quantum number, which may have integral values. The total angular momentum of a nucleon in the nucleus is thus the sum of its orbital angular momentum plus its intrinsic angular momentum or spin:  $j = \ell \pm \frac{1}{2}$ . The plus or minus results because the spin angular momentum vector can be either in the same



**Table 1.2** Nuclear spin and odd-even nuclides.

Number of nucleons	Nuclear spin
Even-even	0
Even-odd	1/2, 3/2, 5/2, 7/2 ...
Odd-odd	1,3

direction or opposite direction of the orbital angular momentum vector. Thus nuclear spin is related to the constituent nucleons in the manner shown in Table 1.2.

Let's now return to magic numbers and see how they relate to the shell model. The magic numbers belong to two different arithmetic series:

$$n = 2, 8, 20, 40, 70, 112 \dots$$
$$n = 2, 6, 14, 28, 50, 82, 126 \dots$$

The lower magic numbers are part of the first series, the higher ones part of the second. The numbers in each series are related by their third differences (the differences between the differences between the differences). For example, for the first of the previous series:

	2	8	20	40	70	112
Difference	6	12	20	30	42	
Difference		6	8	10	12	
Difference			2	2	2	

This series turns out to be solutions to the Schrödinger equation for a three-dimensional harmonic oscillator (Table 1.3). (This solution

is different from the solution for particles in an isotropic coulomb field, which describes electron shells.)

1.2.6.2 Magnetic moment

A rotating charged particle produces a magnetic field. A magnetic field also arises from the orbital motion of charged particles. Thus, electrons in orbit around the nucleus, and also spinning about an internal axis, produce magnetic fields, much as a bar magnet. The strength of a bar magnet may be measured by its magnetic moment, which is defined as the energy needed to turn the magnet from a position parallel to an external magnetic field to a perpendicular position. For the electron, the spin magnetic moment is equal to 1 Bohr magneton ( $\mu_e$ ) =  $5.8 \times 10^{-9}$  eV/gauss. The spin magnetic moment of the proton is 2.79 nuclear magnetons, which is about three orders of magnitude less than the Bohr magneton (hence nuclear magnetic fields do not contribute significantly to atomic ones). Surprisingly, in 1936 the neutron was also found to have an intrinsic magnetic moment, equal to  $-1.91$  nuclear magnetons. Because magnetism always involves motion of charges, this result suggested there is a non-uniform distribution of charge on the neutron, which was an early hint that neutrons and protons were composite particles rather than elementary ones.

Total angular momentum and magnetic moment of pairs of protons cancel because the vectors of each member of the pair are aligned in opposite directions. The same holds true for neutrons. Hence, even-even nuclei have 0 angular momentum and magnetic

**Table 1.3** Particles in a three-dimensional harmonic oscillator (solution to the Schrödinger Equation).

N	1	2		3			4			
$\ell$	0	1		0	2		1	3		
$j$	1/2	1/2	3/2	1/2	3/2	5/2	1/2	3/2	5/2	7/2
State	s <sup>+</sup>	p <sup>-</sup>	p <sup>+</sup>	s <sup>+</sup>	d <sup>-</sup>	d <sup>+</sup>	p <sup>-</sup>	p <sup>+</sup>	f <sup>-</sup>	f <sup>+</sup>
No.	2	2	4	2	4	6	2	4	6	8
$\Sigma$	2	6		12			20			
Total	(2)	(8)		(20)			(40)			

N is the shell number; No. gives the number of particles in the orbit, which is equal to  $2j + 1$ ;  $\Sigma$  gives the number of particles in the shell or state, and total is the total of particles in all shells filled. Magic numbers fail to follow the progression of the first series because only the f state is available in the fourth shell.

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moment. Angular momentum, or nuclear spin, of odd-even nuclides can have values of  $1/2$ ,  $3/2$ ,  $5/2$ , and non-zero magnetic moment (Table 1.2). Odd-odd nuclei have integer value of angular momentum or “nuclear spin.” From this we can see that the angular momentum and magnetic moment of a nucleus are determined by the last nucleon added to the nucleus. For example,  $^{18}\text{O}$  has eight protons and 10 neutrons, and hence

zero angular momentum and magnetic moment. Adding one proton to this nucleus transforms it to  $^{19}\text{F}$ , which has angular momentum of  $1/2$  and magnetic moment of  $\sim 2.79$ . For this reason, the shell model is also sometimes called the single-particle model, since the structure can be recognized from the quantum-mechanical state of the “last” particle (usually). This is a little surprising since particles are assumed to interact.

**Aside: Nuclear magnetic resonance**

Nuclear magnetic resonance (NMR) has no application in isotope geochemistry (it is, however, used in mineralogy), but it has become such an important and successful medical technique that, as long as we are on the subject of nuclear spin, a brief examination of the basics of the technique seems worthwhile. In brief, some nuclei can be excited into higher nuclei spin energy states by radio frequency (RF) radiation – the absorption of this radiation can be detected by an appropriate RF receiver and the frequency of this absorbed radiation provides information about the environment of that nucleus on the molecular level.

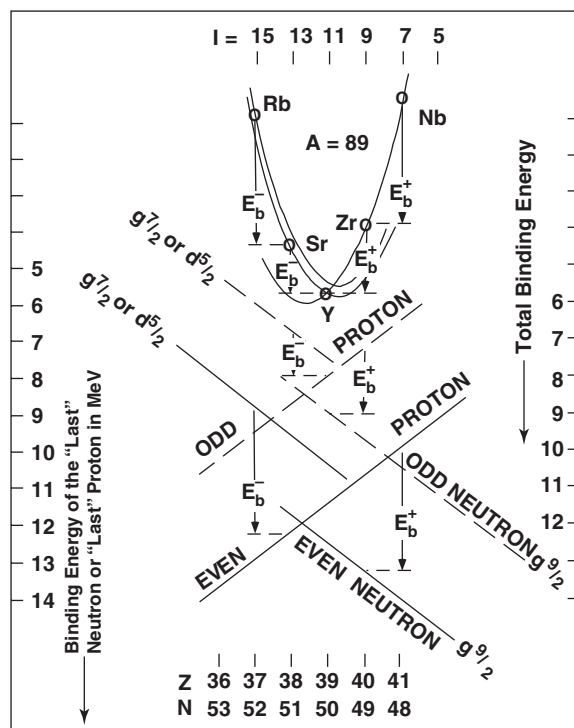
In more detail, it works like this. As we have seen, even-odd and odd-odd nuclei have a nuclear spin. A nucleus of spin  $j$  will have  $2j + 1$  possible orientations. For example,  $^{13}\text{C}$  has a spin  $1/2$  and two possible orientations in space of the spin vector. In the absence of a magnetic field, all orientations have equal energies. In a magnetic field, however, energy levels split and those spin orientations aligned with the magnetic field have lower energy levels (actually, spin vectors precess around the field vector) than others. There will be a thermodynamic (i.e., a Boltzmann) distribution of nuclei among energy states, with more nuclei populating the lower energy levels. The energy difference between these levels is in the range of energies of RF photons (energies are of the order of  $7 \times 10^{-26}$  J, which corresponds to frequencies around 100 MHz). When a nucleus absorbs a photon of this energy, it will change its spin orientation to one having a higher energy level. The precise energy difference between spin states, and hence the precise RF frequency that must be absorbed for the transition to occur, depends on the strength of the applied magnetic field, the nature of the nucleus, and also the atomic environment in which that nucleus is located. The latter is a consequence of magnetic fields of electrons in the vicinity of the nucleus. Although this effect is quite small, it is this slight shift in energy that makes NMR particularly valuable as it allows a non-destructive method of probing the molecular environments of atoms. Non-destructivity is often an advantage for many analytical problems, but, as you can easily imagine, it is particularly important when the sample is a person!

The three-dimensional harmonic oscillator solution explains only the first three magic numbers; magic numbers above that belong to another series. This difference may be explained by assuming there is a strong spin-orbit interaction, resulting from the orbital magnetic field acting upon the spin magnetic moment. This effect is called the Mayer–Jensen coupling. The concept is that the energy state of the nucleon depends strongly on the orientation of the spin of the particle relative to the orbit, and that parallel spin-orbit orientations are energetically favored, that is, states with higher values of  $j$

tend to be the lowest energy states. This leads to filling of the orbits in a somewhat different order; that is, such that high spin values are energetically favored. Spin-orbit interaction also occurs in the electron structure, but it is less important.

**1.2.6.3 Pairing effects**

In the liquid-drop model, it was necessary to add a “fudge factor,” the term  $\delta$ , to account for the even-odd effect. The even-odd effect arises from a “pairing energy” that exists between two nucleons of the same kind. When



**Figure 1.6** Schematic of binding energy as a function of  $I$ , neutron excess number in the vicinity of  $N = 50$ . (Source: Suess (1987). Reproduced with permission of John Wiley & Sons.)

proton-proton and neutron-neutron pairing energies are equal, the binding energy defines a single hyperbola as a function of  $I$  (e.g., Figure 1.5). When they are not, as is often the case in the vicinity of magic numbers, the hyperbola for odd  $A$  splits into two curves, one for even  $Z$ , the other for even  $N$ . An example is shown in Figure 1.6. The empirical rule is: *Whenever the number of one kind of nucleon is somewhat larger than a magic number, the pairing energy of this kind of nucleon will be smaller than the other kind.*

#### 1.2.6.4 Capture cross sections

Information about the structure and stability of nuclei can also be obtained from observations of the probability that a nucleus will capture an additional nucleon. This probability is termed the *capture cross section*, and has units of area. Neutron capture cross sections are generally of greater use than proton capture cross sections, mainly because they are much larger. The reason for this is simply that a proton must overcome the repulsive coulomb forces to be captured, whereas a

neutron, being neutral, is unaffected electrostatic forces. Neutron-capture cross sections are measured in barns, which have units of  $10^{-24} \text{ cm}^2$ , and are denoted by  $\sigma$ . The physical cross section of a typical nucleus (e.g., Ca) is of the order of  $5 \times 10^{-25} \text{ cm}^2$ , and increases somewhat with mass number (more precisely,  $R = r_0 A^{1/3}$ , where  $A$  is mass number and  $r_0$  is the nuclear force radius,  $1.4 \times 10^{-13} \text{ cm}$ ). While many neutron capture cross sections are of the order of 1 barn, they vary from 0 (for  $^4\text{He}$ ) to  $10^5$  for  $^{157}\text{Gd}$ , and are not simple functions of nuclear mass (or size). They depend on nuclear structure, being for example, generally low at magic numbers of  $N$ . Capture cross sections also dependent on the energy of the neutron, the dependence varying from nuclide to nuclide.

#### 1.2.7 Collective model

A slightly more complex model is called the collective model. It is intermediate between the liquid-drop and the shell models. It emphasizes the collective motion of nuclear matter, particularly the vibrations and rotations, both

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quantized in energy, in which large groups of nucleons can participate. Even-even nuclides with  $Z$  or  $N$  close to magic numbers are particularly stable with nearly perfect spherical symmetry. Spherical nuclides cannot rotate because of a dictum of quantum mechanics that a rotation about an axis of symmetry is undetectable, and in a sphere every axis is a symmetry axis. The excitation of such nuclei (that is, when their energy rises to some quantum level above the ground state) may be ascribed to the vibration of the nucleus as a whole. On the other hand, even-even nuclides far from magic numbers depart substantially from spherical symmetry and the excitation energies of their excited states may be ascribed to rotation of the nucleus as a whole.

## 1.3 RADIOACTIVE DECAY

As we have seen, some combinations of protons and neutrons form nuclei that are only “metastable.” These ultimately transform to stable nuclei through the process called radioactive decay. This involves emission of a particle or particles and is usually accompanied by emission of a photon as well. In some cases, the photon emission is delayed and the daughter nuclide is left in an excited state. Just as an atom can exist in any one of a number of excited states, so too can a nucleus have a set of discrete, quantized, excited nuclear states. The behavior of nuclei in transforming to more stable states is somewhat similar to atomic transformation from excited to more stable states, but there are some important differences. First, energy level spacing is much greater; second, the time an unstable nucleus spends in an excited state can range from  $10^{-14}$  sec to  $10^{11}$  years, whereas atomic lifetimes are usually about  $10^{-8}$  sec. Like atomic transitions, nuclear reactions must obey general physical laws, conservation of momentum, mass-energy, spin, and so on, and conservation of nuclear particles.

Nuclear decay takes place at a rate that follows the law of radioactive decay. There are two extremely interesting and important aspects of radioactive decay. First, the decay rate is dependent only on the nature and energy state of the particular nuclide; it is independent of the history of the nucleus, and essentially independent of external influences such as temperature, pressure, and so on. It

is this property that makes radioactive decay so useful as a chronometer. Second, it is completely impossible to predict when a given nucleus will decay. We can, however, predict the probability of its decay in a given time interval. The probability of decay in some infinitesimally small time interval,  $dt$  is  $\lambda dt$ . Therefore, the rate of decay among some number,  $N$ , of nuclides is:

$$\frac{dN}{dt} = -\lambda N \quad (1.12)$$

The minus sign simply indicates  $N$  decreases. Equation 1.12 is a first-order rate law known as the *basic equation of radioactive decay*. Essentially, all the significant equations of radiogenic isotope geochemistry and geochronology can be derived from this simple expression.

## 1.3.1 Gamma decay

A gamma ray is simply a high-energy photon (i.e., electromagnetic radiation). Just as an atom can be excited into a higher energy state when it absorbs photon, nuclei can be excited into higher energy states by absorption of a much higher energy photon. Both excited atom and nuclei subsequently decay to their ground states by emission of a photon. Photons involved in atomic excitation and decay have energies ranging from the visible to the X-ray part of the electromagnetic spectrum (roughly 1 eV to 100 keV); gamma rays involved in nuclear transitions typically have energies greater than several hundred keV. Although nuclei, like atoms, generally decay promptly from excited states, in some cases nuclei can persist in metastable excited states characterized by higher nuclear spin for considerable lengths of time.

The gamma ray frequency is related to the energy difference by:

$$h\nu = E_u - E_l \quad (1.13)$$

where  $E_u$  and  $E_l$  are simply the energies of the upper (excited) and lower (ground) states and  $h$  is the reduced Planck's constant ( $h/2\pi$ ). The nuclear reaction is written as:



Gamma emission usually, but not invariably, accompanies alpha and beta decay as a consequence of the daughter being left in an excited state. Decay generally occurs within  $10^{-12}$  sec of the decay, but, as noted here, can be delayed if the daughter persists in a metastable state.

### 1.3.2 Alpha decay

An  $\alpha$ -particle is simply a helium nucleus. Since the helium nucleus is particularly stable, it is not surprising that such a group of particles might exist within the parent nucleus before  $\alpha$ -decay. Emission of an alpha particle decreases the mass of the nucleus by the mass of the alpha particle, and also by the kinetic energy of the alpha particle and the remaining nucleus (because of the conservation of momentum, the remaining nucleus recoils from the decay reaction). The  $\alpha$  particle may leave the nucleus with any of several discrete kinetic energy levels, as is illustrated in Figure 1.7.

The escape of the  $\alpha$  particle is a bit of a problem, because it must overcome a very substantial energy barrier, a combination of the strong force and the coulomb repulsion, to get out. For example,  $\alpha$  particles fired at in  $^{238}\text{U}$  with energies below 8 Mev are scattered from the nucleus. However, during  $\alpha$  decay of  $^{238}\text{U}$ , the  $\alpha$  particle emerges with an energy of

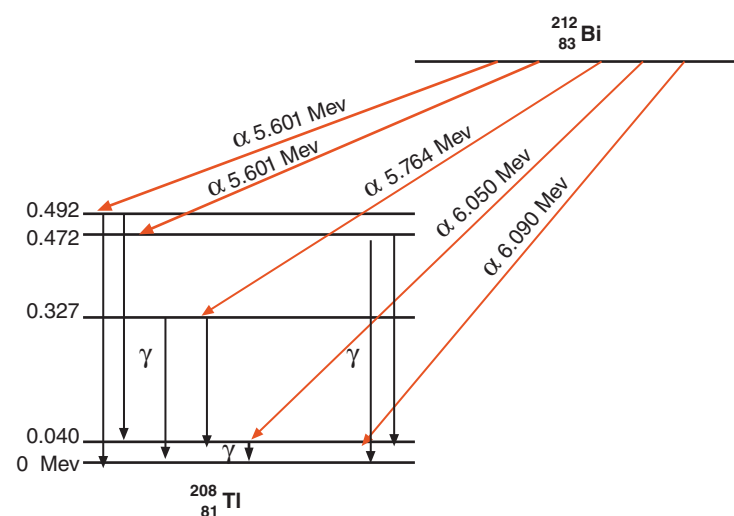
only about 4 Mev. This is an example of an effect called *tunneling* and can be understood as follows. We can never know exactly where the  $\alpha$  particle is (or any other particle, or you or I for that matter), we only know the probability of its being in a particular place. This probability is given by the particle's wavefunction,  $\psi(r)$ . The wave is strongly attenuated through the potential energy barrier, but has a small but finite amplitude outside the nucleus, and hence a small but finite probability of its being located there.

The escape of an alpha particle leaves a daughter nucleus with mass  $<A-4$ ; the missing mass is the kinetic energy of the alpha and remaining nucleus. The daughter may originally be in an excited state, from which it will decay by  $\gamma$ -decay. Figure 1.7 shows an example energy-level diagram for such decay. Note that the sum of the kinetic energy of the  $\alpha$  and the energy of the  $\gamma$  is constant.

Alpha-decay occurs in nuclei with mass above the maximum in the binding energy curve (Figure 1.3), which occurs at  $^{56}\text{Fe}$ . Possibly all such nuclei are unstable relative to alpha-decay, but most of their half-lives are immeasurably long.

### 1.3.3 Beta decay

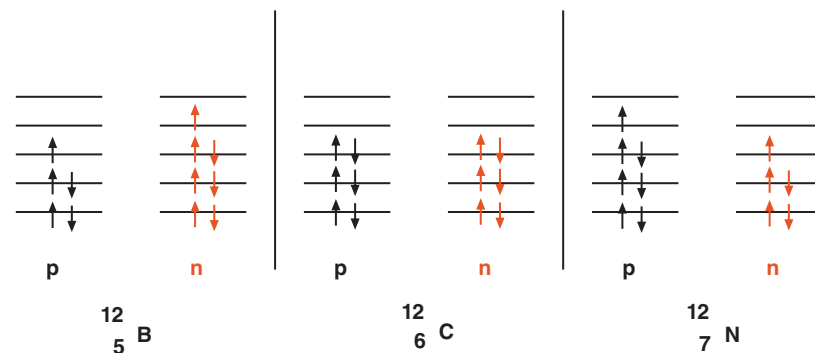
Beta decay is a process in which the charge of a nucleus changes, but the number of nucleons



**Figure 1.7** Nuclear energy-level diagram showing decay of bismuth-212 by alpha emission to the ground and excited states of thallium-208. (Source: White (2013). Reproduced with permission of John Wiley & Sons.)



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**Figure 1.8** Proton and neutron occupation levels of boron 12, carbon 12, and nitrogen 12. (Source: White (2013). Reproduced with permission of John Wiley & Sons.)

remains the same. If we plotted Figure 1.1 with a third dimension, namely energy of the nucleus, we would see the stability region forms an energy valley. Alpha-decay moves a nucleus down the valley axis; beta decay moves a nucleus down the walls toward the valley axis. Beta-decay results in the emission of an electron or positron, depending on which side of the valley the parent lies. Consider the three nuclei in Figure 1.8 (these are isobars, since they all have 12 nucleons). From what we have learned of the structure of nuclei, we can easily predict the  $^{12}\text{C}$  nucleus is the most stable. This is the case.  $^{12}\text{B}$  decays to  $^{12}\text{C}$  by the creation and emission of a  $\beta^-$  particle and the conversion of a neutron to a proton.  $^{12}\text{N}$  decays by emission of a  $\beta^+$  and conversion of a proton to a neutron.

Here, physicists had a problem. Angular momentum must be conserved in the decay of nuclei. The  $^{12}\text{C}$  nucleus has integral spin as do  $^{12}\text{B}$  and  $^{12}\text{N}$ . But the beta particle has  $1/2$  quantum spin units. An additional problem is that rather than having discrete kinetic energies,  $\beta$  particles exhibit a spectrum of kinetic energies, although there is a well-defined maximum energy. Thus beta decay appeared to violate both conservation of momentum and energy. The solution, proposed by Enrico Fermi, was the existence of another, nearly massless particle,<sup>3</sup> called the *neutrino*, having  $1/2$  spin and variable kinetic energy. Thus in beta decay, a neutrino is also released and the sum of the kinetic energy of the beta and neutrino, plus the energy of any gamma, is constant.

Beta decay involves the weak force, or weak interaction. The weak interaction transforms

one flavor of quark into another and thereby a charged particle (e.g., a proton) into a neutral (e.g., a neutron) and vice versa. Both the weak and the electromagnetic forces are thought to be simply a manifestation of one force, called electroweak, that accounts for all interactions involving charge (in the same sense that electric and magnetic forces are manifestations of electromagnetism). In  $\beta^+$  decay, for example, a proton is converted to a neutron, giving up its  $+1$  charge to a neutrino, which is converted to a positron. This process occurs through the intermediacy of the  $W^+$  particle in the same way that electromagnetic processes are mediated by photons. The photon, pion, and  $W$  particles are members of a class of particles called bosons that mediate forces between the basic constituents of matter. However, the  $W$  particles differ from photons in having a very substantial mass (around 80 GeV or almost 2 orders of magnitude greater mass than the proton). Interestingly, *Nature* rejected the paper in which Fermi proposed the theory of beta decay involving the neutrino and the weak force in 1934!

#### 1.3.4 Electron capture

Another type of reaction is electron capture. This is sort of the reverse of beta decay and has the same effect, more or less, as  $\beta^+$  decay. Interestingly, this is a process in which an electron is added to a nucleus to produce a nucleus with less mass than the parent! The missing mass is carried off as energy by an escaping neutrino, and in some cases by a  $\gamma$ . In some cases, a nucleus can decay by either electron capture,  $\beta^-$ , or  $\beta^+$  emission. An

example is the decay of  $^{40}\text{K}$ , which decays to  $^{40}\text{Ca}$  by  $\beta^-$  and  $^{40}\text{Ar}$  to by  $\beta^+$  or electron capture. We should point out that electron capture is an exception to the environmental independence of nuclear decay reactions in that it shows a very slight dependence on pressure.

$\beta$  decay and electron capture often leaves the daughter nucleus in an excited state. In this case, it will decay to its ground state (usually very quickly) by the emission of a  $\gamma$ -ray. Thus,  $\gamma$  rays often accompany  $\beta$  decay (as well as  $\alpha$  decay). A change in charge of the nucleus necessitates a rearrangement of electrons in their orbits. This is particularly true in electron capture, where an inner electron is lost. As electrons jump down to lower orbits to occupy the orbital freed by the captured electron, they give off electromagnetic energy. This produces X-rays from electrons in the inner orbits.

### 1.3.5 Spontaneous fission

This is a process in which a nucleus splits into two or more fairly heavy daughter nuclei. In nature, this is a very rare process, occurring only in the heaviest nuclei,  $^{238}\text{U}$ ,  $^{235}\text{U}$ , and  $^{232}\text{Th}$  (it is, however, most likely in  $^{238}\text{U}$ ). It also occurs in  $^{244}\text{Pu}$ , an extinct radionuclide (we use the term “extinct radionuclide” to refer to nuclides that once existed in the Solar System, but which have subsequently decayed away entirely). The liquid-drop model perhaps better explains this particular phenomenon than the shell model. Recall that in the liquid-drop model, there are four contributions to total binding energy: volume energy, surface tension, excess neutron energy, and coulomb energy. The surface tension tends to minimize the surface area while the repulsive coulomb energy tends to increase it. We can visualize these nuclei as oscillating between various shapes. It may very rarely become so distorted by the repulsive force of 90 or so protons, that the surface tension cannot restore the shape. Surface tension is instead minimized by the splitting the nucleus entirely.

Since there is a tendency for  $N/Z$  to increase with  $A$  for stable nuclei, the parent is much richer in neutrons than the daughters produced by fission (which may range from  $Z = 30$ , zinc, to  $Z = 65$ , terbium). Thus fission generally also produces some free neutrons in addition to two nuclear fragments (the

daughters). The daughters are typically of unequal size, the exact mass of the two daughters being random. The average mass ratio of the high to the low mass fragment is about 1.45. Even though some free neutrons are created, the daughters tend to be too neutron-rich to be stable. As a result, they decay by  $\beta^-$  to stable daughters. It is this decay of the daughters that results in radioactive fallout in bombs and radioactive waste in reactors (a secondary source of radioactivity is production of unstable nuclides by capture of the neutrons released).

Some non-stable heavy nuclei and excited heavy nuclei are particularly unstable with respect to fission. An important example is  $^{236}\text{U}$ . Imagine a material rich in U. When  $^{238}\text{U}$  undergoes fission, one of the released neutrons can be captured by  $^{235}\text{U}$  nuclei, producing  $^{236}\text{U}$  in an excited state. This  $^{236}\text{U}$  then fissions producing more neutrons, and so on. This is the basis of nuclear reactors and bombs (the latter can also be based on Pu). The concentration of U is not usually high enough in nature for this sort of thing to happen. But it apparently did at least once, 1.5 billion years ago in the Oklo U deposit in Africa. This deposit was found to have an anomalously high  $^{238}\text{U}/^{235}\text{U}$  ratio (227 versus 137.82), indicating some of the  $^{235}\text{U}$  had been “burned” in a nuclear chain reaction. Could such a natural nuclear reactor happen again? Probably not, because there is a lot less  $^{235}\text{U}$  around now than there was 1.7 billion years ago. With equations we’ll introduce soon, you should be able to calculate just how much less.

Individual natural fission reactions are less rare. When fission occurs, there is a fair amount of kinetic energy produced (maximum about 200 MeV), the nuclear fragments literally flying apart. These fragments damage the crystal structure through which they pass, producing “tracks”, whose visibility can be enhanced by etching. This is the basis of fission-track dating, which we’ll describe in Chapter 4.

Natural fission also can produce variations in the isotopic abundance of elements among the natural, ultimate product. Xenon is an important product, as we’ll learn in Chapter 12. Indeed, the critical evidence showing that a nuclear chain reaction had indeed occurred in the Oklo deposit was the discovery that fission product elements, such as Nd and Ru, had

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anomalous isotopic compositions. Analysis of the isotopic composition of another fission product, Sm, has led to a controversy over whether the fine scale constant,  $\alpha$ , has changed over time.  $\alpha$  is related to other fundamental constants as:

$$\alpha = \frac{e^2}{\hbar c} \quad (1.15)$$

where  $e$  is the charge of the electron (and  $\hbar$  is the reduced Planck constant and  $c$  is the speed of light). A change in the fine scale constant thus raises the possibility of a change in  $c$ . The change, if it occurred, is quite small, less than 1 part in  $10^7$ , and could be consistent with some observations about quasars and the early universe.

## 1.4 NUCLEOSYNTHESIS

A reasonable starting point for isotope geochemistry is a determination of the abundances of the naturally occurring nuclides. Indeed, this was the first task of isotope geochemists (although those engaged in this work would have referred to themselves simply as physicists). This began with Thomson, who built the first mass spectrometer and discovered that Ne consisted of two isotopes (actually, it consists of three, but one of them,  $^{21}\text{Ne}$ , is very much less abundant than the other two, and Thomson's primitive instrument did not detect it). Having determined the abundances of nuclides, it is natural to ask what accounts for this distribution, and even more fundamentally, what process or processes produced the elements. This process is known as nucleosynthesis.

The abundances of naturally occurring nuclides are now reasonably well known – at least in our Solar System. We also have what appears to be a reasonably successful theory of nucleosynthesis. Physicists, like all scientists, are attracted to simple theories. Not surprisingly then, the first ideas about nucleosynthesis attempted to explain the origin of the elements by single processes. Generally, these were thought to occur at the time of the Big Bang. None of these theories was successful. It was really the astronomers, accustomed to dealing with more complex phenomena than physicists, who successfully produced a theory of nucleosynthesis that involved a number of processes. Today, isotope geochemists continue to be involved in refining

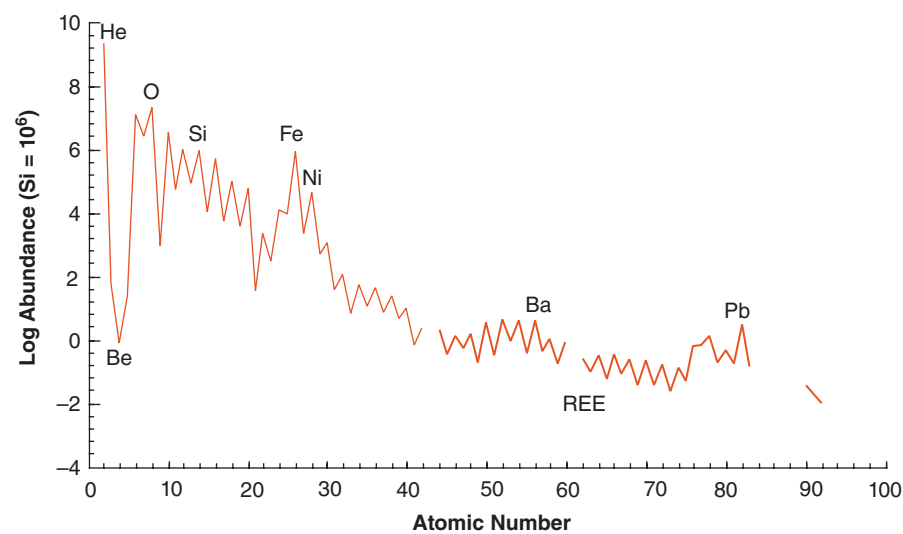
these ideas by examining and attempting to explain isotopic variations occurring in some meteorites; two recent examples are papers by Jadhav et al. (2013) and Haenecour et al. (2013).

The origin of the elements is an astronomical question, perhaps even more a cosmological one. To understand how the elements formed we need to understand a few astronomical observations and concepts. The Universe began about 13.8 Ga<sup>4</sup> ago with the Big Bang. Since then the Universe has been expanding, cooling, and evolving. This hypothesis follows from two observations: the relationship between red-shift and distance and the cosmic background radiation, particularly the former. This cosmology provides two possibilities for formation of the elements: (1) they were formed in the Big Bang itself, or (2) they were subsequently produced. As we shall see, the answer is both.

Our present understanding of nucleosynthesis comes from three sorts of observations: (1) the abundance of isotopes and elements in the Earth, Solar System, and cosmos (spectral observations of stars), (2) experiments on nuclear reactions that determine what reactions are possible (or probable) under given conditions, and (3) inferences about possible sites of nucleosynthesis and about the conditions that would prevail in those sites. The abundances of the elements in the Solar System are shown in Figure 1.9.

Various hints came from all three of these observations. For example, it was noted that the most abundant nuclide of a given set of stable isobars tended to be the most neutron-rich one. We now understand this to be a result of shielding from  $\beta$ -decay (see the discussion of the r-process next).

Another key piece of evidence regarding formation of the elements comes from looking back into the history of the cosmos. Astronomy is a bit like geology in that just as we learn about the evolution of the Earth by examining old rocks, we can learn about the evolution of the cosmos by looking at old stars. It turns out that old stars (such old stars are most abundant in the globular clusters outside the main disk of the Milky Way) are considerably poorer in heavy elements than are young stars. This suggests much of the heavy element inventory of the galaxy has been produced since these stars formed (some



**Figure 1.9** Solar System abundance of the elements relative to silicon as a function of atomic number. (Source: White (2013). Reproduced with permission of John Wiley & Sons.)

10 Ga ago). On the other hand, they seem to have about the same He/H ratio as young stars. Indeed  $^4\text{He}$  seems to have an abundance of 24–28% in all stars. Another key observation was the identification of technetium emissions in the spectra of some stars. Since the most stable isotope of this element has a half-life of about 100,000 years and for all intents and purposes it does not exist in the Earth, it must have been synthesized in those stars. Thus the observational evidence suggests (1) H and He are everywhere uniform implying their creation and fixing of the He/H ratio in the Big Bang and (2) subsequent creation of heavier elements (heavier than Li, as we shall see) by subsequent processes.

As we mentioned, early attempts ( $\sim 1930 - 1950$ ) to understand nucleosynthesis focused on single mechanisms. Failure to find a single mechanism that could explain the observed abundance of nuclides, even under varying conditions, led to the present view that relies on a number of mechanisms operating in different environments and at different times for creation of the elements in their observed abundances. This view, often called the polygenetic hypothesis, is based mainly on the work of Burbidge, Burbidge, Fowler and Hoyle. Their classic paper summarizing the theory, “Synthesis of the elements in stars” was published in *Reviews of Modern Physics* in 1957. Interestingly, the abundance of trace

elements and their isotopic compositions were perhaps the most critical observations in development of the theory. An objection to this polygenetic scenario was the apparent uniformity of the isotopic composition of the elements. But variations in the isotopic composition have now been demonstrated for many elements in some meteorites. Furthermore, there are quite significant compositional variations in heavier elements among stars. These observations provide strong support for this theory.

To briefly summarize it, the polygenetic hypothesis proposes four phases of nucleosynthesis. *Cosmological nucleosynthesis* occurred shortly after the Universe began and is responsible for the cosmic inventory of H and He, and some of the Li. Helium is the main product of nucleosynthesis in the interiors of normal, or “main sequence” stars. The lighter elements, up to and including Si, but excluding Li and Be, and a fraction of the heavier elements may be synthesized in the interiors of larger stars during the final stages of their evolution (*stellar nucleosynthesis*). The synthesis of the remaining elements occurs as large stars exhaust the nuclear fuel in their interiors and explode in nature’s grandest spectacle, the supernova (*explosive nucleosynthesis*). Finally, Li and Be are continually produced in interstellar space by interaction of cosmic rays with matter (*galactic*



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*nucleosynthesis*). In the following sections, we examine these nucleosynthetic processes as presently understood.

## 1.4.1 Cosmological nucleosynthesis

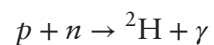
Immediately after the Big Bang, the Universe was too hot for any matter to exist – there was only energy. Some  $10^{-11}$  seconds later, the Universe had expanded and cooled to the point where quarks and anti-quarks could condense from the energy. The quarks and anti-quarks, however, would also collide and annihilate each other. So a sort of thermal equilibrium existed between matter and energy. As things continued to cool, this equilibrium progressively favored matter over energy. Initially, there was an equal abundance of quarks and anti-quarks, but as time passed, the symmetry was broken and quarks came to dominate. The current theory is that the hyper-weak force was responsible for an imbalance favoring matter over anti-matter. After  $10^{-4}$  seconds, things were cool enough for quarks to associate with one another and form nucleons: protons and neutrons. After  $10^{-2}$  seconds, the Universe has cooled to  $10^{11}$  K. Electrons and positrons were in equilibrium with photons, neutrinos, and antineutrinos were in equilibrium with photons, and antineutrinos combined with protons to form positrons and neutrons, and neutrinos combined with neutrons to form electrons and protons:



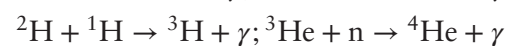
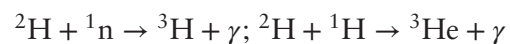
This equilibrium produced about an equal number of protons and neutrons. However, the neutron is unstable outside the nucleus and decays to a proton with a half-life of about 15 min. So as time continued passed, protons became more abundant than neutrons.

After a second or so, the Universe had cooled to  $10^{10}$  K, which shut down the reactions above. Consequently, neutrons were no longer being created, but they were being destroyed as they decayed to protons. At this point, protons were about three times as abundant as neutrons.

It took another 3 min to for the Universe to cool to  $10^9$  K, which is cool enough for  $^2\text{H}$ , created by



to be stable. At about the same time, the following reactions could also occur:



and  $^3\text{He} + ^4\text{He} \rightarrow ^7\text{Be} + \gamma; \quad ^7\text{Be} \rightarrow ^7\text{Li} + e^- + \gamma$

One significant aspect of this event is that it began to lock up neutrons in nuclei where they could no longer decay to protons. The timing of this event fixes the ratio of protons to neutrons at about 7:1. Because of this dominance of protons, hydrogen is the dominant element in the Universe. About 24% of the mass of the Universe was converted to  $^4\text{He}$  in this way; less than 0.01% was converted to  $^2\text{H}$ ,  $^3\text{He}$ , and  $^7\text{Li}$  (and there is good agreement between theory and observation). Formation of elements heavier than Li was inhibited by the instability of nuclei of masses 5 and 8. Shortly thereafter, the universe cooled below  $10^9$  K and nuclear reactions were no longer possible.

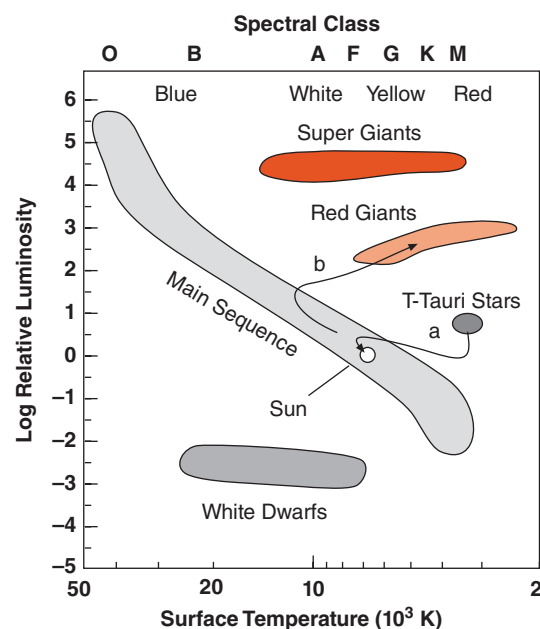
Thus, the Big Bang created H, He and a bit of Li ( $^7\text{Li}/\text{H} < 10^{-9}$ ). Some 300,000 years or so later, the Universe had cooled to about 3000 K, cool enough for electrons to be bound to nuclei, forming atoms. It was at this time, called the “recombination era” that the Universe first became transparent to radiation. Prior to that, photons were scattered by the free electrons, making the Universe opaque. It is the radiation emitted during this recombination that makes up the cosmic microwave background radiation that we can still detect today. Discovery of this cosmic microwave background radiation, which has the exact spectra predicted by the Big Bang model, represents a major triumph for the model and is not easily explained in any other way.

## 1.4.2 Stellar nucleosynthesis

## 1.4.2.1 Astronomical background

Before discussing nucleosynthesis in stars, it is useful to review a few basics of astronomy. Stars shine because of exothermic nuclear reactions occurring in their cores. The energy released by these processes results in thermal expansion that, in general, exactly balances gravitational collapse. Surface temperatures are very much cooler than temperatures in stellar cores. For example, the Sun, which is





**Figure 1.10** The Hertzsprung–Russell diagram of the relationship between luminosity and surface temperature. Arrows show evolutionary path for a star the size of the Sun in pre- (a) and post- (b) main sequence phases. (Source: White (2013). Reproduced with permission of John Wiley & Sons.)

in many respects an average star, has a surface temperature of 5700 K and a core temperature thought to be 14,000,000 K.

Stars are classified based on their color (and spectral absorption lines), which in turn is related to their temperature. From hot to cold, the classification is: O, B, F, G, K, M, with subclasses designated by numbers, for example, F5. (The mnemonic is “*O Be a Fine Girl, Kiss Me!*”). The Sun is class G. Stars are also divided into Populations. Population I stars are second or later generation stars and have greater heavy element contents than Population II stars. Population I stars are generally located in the main disk of the galaxy, whereas the old first generation stars of Population II occur mainly in globular clusters that circle the main disk.

On a plot of luminosity versus wavelength of their principal emissions (i.e., color), called a Hertzsprung–Russell diagram (Figure 1.10), most stars (about 90%) fall along an array defining an inverse correlation between these two properties. Since wavelength is inversely related to temperature, this correlation means simply that hot stars are more luminous and give off more energy than cooler stars. Mass and radius are also simply related to

temperature and luminosity for these so-called “main sequence” stars;<sup>5</sup> hot stars are big, small stars are cooler. Thus O and B stars are large, luminous, and hot; K and M stars are small, cool, and (comparatively speaking) dark. Stars on the main sequence produce energy by “hydrogen burning,” fusion of hydrogen to produce helium. Since the rate at which these reactions occur depends on temperature and density, hot, massive stars release more energy than smaller ones. As a result, they exhaust the hydrogen in their cores much more rapidly. Thus there is an inverse relationship between the lifetime of a star and its mass. The most massive stars, up to 100 solar masses, have life expectancies of only about  $10^6$  years or so, whereas small stars, as small as 0.01 solar masses, remain on the main sequence more than  $10^{10}$  years.

The two most important exceptions to the main sequence stars, the red giants and the white dwarfs, represent stars that have burned all the H fuel in their cores and have moved on in the evolutionary sequence. When the H in the core is converted to He, it generally cannot be replenished because the density difference prevents convection between the core and out layers, which are still H-rich. The interior

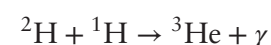
part of the core collapses under gravity. With enough collapse, the layer immediately above the He core will begin to “burn” H again, which again stabilizes the star. The core, however, continues to collapse until T and P are great enough for He burning to begin. At the same time, energy from the interior is transferred to the outer layers, causing the exterior to expand; it cools as it expands, resulting in a *red giant*; a star that is over-luminous relative to main sequence stars of the same color. When the Sun reaches this phase, in perhaps another 5 Ga, it will expand to the Earth’s orbit. A star will remain in the red giant phase for of the order of  $10^6$ – $10^8$  years. During this time, radiation pressure results in a greatly enhanced solar wind, of the order of  $10^{-6}$  to  $10^{-7}$ , or even  $10^{-4}$ , solar masses per year. For comparison, the present solar wind is  $10^{-14}$  solar masses/year; thus, in its entire main-sequence lifetime, the Sun will blow off 1/10,000 of its mass through solar wind.

The fate of stars after the red giant phase (when the He in the core is exhausted) depends on their mass. Nuclear reactions in small stars cease and they simply contract, their exteriors heating up as they do so, to become *white dwarfs*. The energy released is that produced by previous nuclear reactions and released gravitational potential energy. This is the likely fate of the Sun. White Dwarfs are underluminous relative to stars of similar color on the main sequence. They can be thought of as little more than glowing ashes. Unless they blow off sufficient mass during the red giant phase, stars larger than 8 solar masses die explosively, in supernovae (specifically, Type II supernovae). (Novae are entirely different events that occur in binary systems when mass from a main sequence star is pulled by gravity onto a white dwarf companion.) Supernovae are incredibly energetic events. The energy released by a supernova can exceed that released by an entire galaxy (which, it will be recalled, consists of on the order of  $10^9$  stars) for a period of days or weeks!

#### 1.4.2.2 Hydrogen, helium, and carbon burning in main sequence and red giant stars

For quite some time after the Big Bang, the Universe was a more or less homogeneous, hot gas. More or less turns out to be critical wording. Inevitably (according to fluid dynamics),

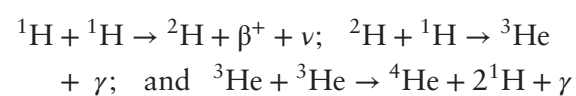
inhomogeneities in the gas developed. These inhomogeneities enlarged in a sort of runaway process of gravitational attraction and collapse. Thus were formed protogalaxies, thought to date to about 0.5–1.0 Ga after the Big Bang. Instabilities within the protogalaxies collapsed into stars. Once this collapse proceeds to the point where temperatures reach 1 million K, deuterium burning can begin:



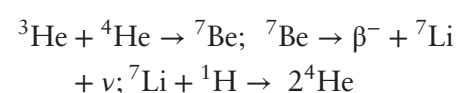
This occurs while pre-main stars are still accreting mass and growing and temporarily stabilizes the star against further collapse. This may continue for several million years in smaller stars such as the Sun. (In low mass objects that will never reach temperatures and pressures for hydrogen burning to initiate, known as brown dwarfs, deuterium burning can occur and continue for hundreds of millions of years before the deuterium is exhausted. This requires a mass at least 13 times that of Jupiter to occur.)

When deuterium is exhausted and the stellar core reaches a density 6 g/cm and temperature 10–20 million K, *hydrogen burning*, or the *pp process* begins and continues through the main sequence life of the star. There are three variants,

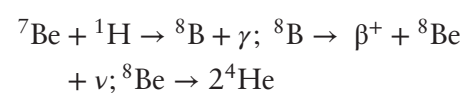
PP I:



PP II:



and PP III:



Which of these reactions dominates depends on temperature, but the net result of all is the production of  $^4\text{He}$  and the consumption of H (and Li). All main sequence stars produce He, yet over the history of the cosmos, this has had little impact on the H/He ratio of the Universe. This in part reflects the observation that for small mass stars, the He produced remains hidden in their interiors or their white dwarf remnants and for large mass stars, later

**Figure 1.11** Illustration of the CNO cycle, which operates in larger second and later generation stars.

$^{14}\text{N}(\text{p}, \gamma)^{15}\text{O}$ . As a result, there is a production of  $^{14}\text{N}$  in the cycle and net consumption of C and O. The CNO cycle will also tend to leave remaining carbon in the ratio of  $^{13}\text{C}/^{12}\text{C}$  of 0.25. This is quite different than the Solar System (and terrestrial) abundance ratio of about 0.01. Because of these rate imbalances, the CNO cycle may be the principle source of nitrogen in the Universe.

The CNO cycle and the PP chains are competing fusion reactions in main sequence stars. Which dominates depends on temperature. In the Sun, the PP reactions account for about 98–99% of the energy production, with the CNO cycle producing the remainder. But if the Sun were only 12–30% more massive (and consequently a few million K hotter), the CNO cycle would dominate energy production.

Once the H is exhausted in the stellar core, fusion ceases, and the balance between gravitational collapse and thermal expansion is broken. The interior of the star thus collapses, raising the star's temperature. The increase in temperature results in expansion of the exterior and ignition of fusion in the shells surrounding the core that now consists of He. This is the *red giant* phase. Red giants may have diameters of hundreds of millions of kilometers (greater than the diameter of Earth's orbit). If the star is massive enough for temperatures to reach  $10^8\text{K}$  and density to reach  $10^4\text{g/cc}$  in the He core, *He burning*, (also called the triple alpha process) can occur:

$$\begin{aligned} & {}^4\text{He} + {}^4\text{He} \rightarrow {}^8\text{Be} + \gamma \\ \text{and } & {}^8\text{Be} + {}^4\text{He} \rightarrow {}^{12}\text{C} + \gamma \end{aligned}$$

The catch in these reactions is that the half-life of  $^8\text{Be}$  is only  $7 \times 10^{-16}$  sec, so three

$^{12}\text{C}(\text{p}, \gamma)^{13}\text{N}$

is equivalent to:

$$^{12}\text{C} + \text{p} \rightarrow ^{13}\text{N} + \gamma$$

It was subsequently realized that this reaction cycle is just part of a larger reaction cycle, which is illustrated in Figure 1.11. Since the process is cyclic, the net effect is consumption of four protons and two positrons to produce a neutrino, some energy, and a  ${}^4\text{He}$  nucleus. Thus, to a first approximation, carbon acts as a kind of nuclear catalyst in this cycle: it is neither produced nor consumed. When we consider these reactions in more detail, not all of them operate at the same rate, resulting in some production and some consumption of these heavier nuclides. The net production of a nuclide can be expressed as:

$$\frac{dN}{dt} = (\text{creation rate} - \text{destruction rate}) \quad (1.16)$$

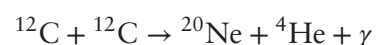
Reaction rates are such that some nuclides in this cycle are created more rapidly than they are consumed, while for others the opposite is true. The slowest of the reactions in Cycle I is

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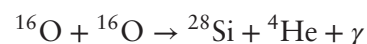
He nuclei must collide nearly simultaneously (this reaction is sometimes called the triple alpha process for this reason), hence densities must be very high. Depending on the mass of the star, the red giant phase can last for as much as hundred million years or as little as a few hundred thousand years, as a new equilibrium between gravitational collapse and thermal expansion sets in. Helium burning also produces  $^{16}\text{O}$  through fusion of  $^4\text{He}$  with  $^{12}\text{C}$ . Upon the addition of further He nuclei,  $^{20}\text{Ne}$  and  $^{24}\text{Mg}$  can be produced.  $^{14}\text{N}$  created by the CNO cycle in second generation stars can be converted to  $^{22}\text{Ne}$ ; however, production rates of nuclei heavier than  $^{16}\text{O}$  is probably quite low at this point. Also note that Li, Be, and B have been skipped: they are not synthesized in these phases of stellar evolution. Indeed, they are actually consumed in stars, in reactions such as PP II and PP III.

Evolution for low-mass stars, such as the Sun, ends after the Red Giant phase and helium burning. Densities and temperatures necessary to initiate further nuclear reactions cannot be achieved because the gravitational force is not sufficient to overcome coulomb repulsion of electrons. Thus nuclear reactions cease and radiation is produced only by a slow cooling and gravitational collapse. Massive stars, those greater than about four solar masses, however, undergo further collapse and further evolution. Evolution now

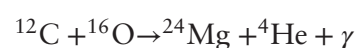
proceeds at an exponentially increasing pace (Figure 1.12), and these phases are poorly understood. But if temperatures reach 600 million K and densities  $5 \times 10^5 \text{ g/cc}$ , carbon burning becomes possible:



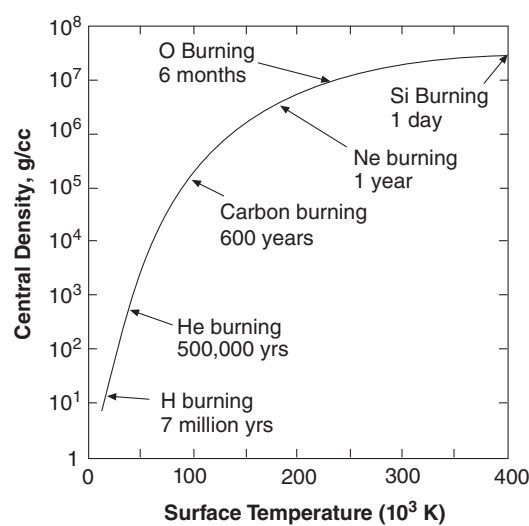
The carbon-burning phase marks a critical juncture in stellar evolution. As we mentioned, low mass stars never reach this point. Intermediate mass stars, those with 4–8 solar masses can be catastrophically disrupted by the ignition of carbon burning. The outer envelope of the star is ejected, leaving an O-Ne-Mg white dwarf. But in large stars, those with more than 8 solar masses, the sequence of production of heavier and heavier nuclei continues. After carbon burning, there is an episode called Ne burning, in which  $^{20}\text{Ne}$  “photodisintegrates” by a  $(\gamma, \alpha)$  reaction. The  $\alpha$ s produced are consumed by those nuclei present, including  $^{20}\text{Ne}$ , creating heavier elements, notably  $^{24}\text{Mg}$ . The next phase is oxygen burning, which involves reactions such as:



and



A number of other less abundant nuclei, including Na, Al, P, S, and K are also synthesized at this time.

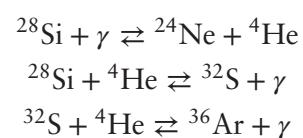


**Figure 1.12** Evolutionary path of the core of star of 25 solar masses (after Bethe and Brown, 1985). Note that the period spent in each phase depends on the mass of the star: massive stars evolve more rapidly. (Source: ©Ian Worpole. Reproduced with permission.)

During the final stages of evolution of massive stars, a significant fraction of the energy released is carried off by neutrinos created by electron-positron annihilations in the core of the star. If the star is sufficiently oxygen-poor that its outer shells are reasonably transparent, the outer shell of the red giant may collapse during last few  $10^4$  years of evolution to form a *blue supergiant*.

#### 1.4.2.3 The *e*-process

Eventually, a new core consisting mainly of  $^{28}\text{Si}$  is produced. At temperatures above  $10^9\text{K}$  and densities above  $10^7\text{g/cc}$  a process known as *silicon burning*, or the *e* process, (for equilibrium) begins, and lasts for only day or so, again depending on the mass of the star. These are reactions of the type:



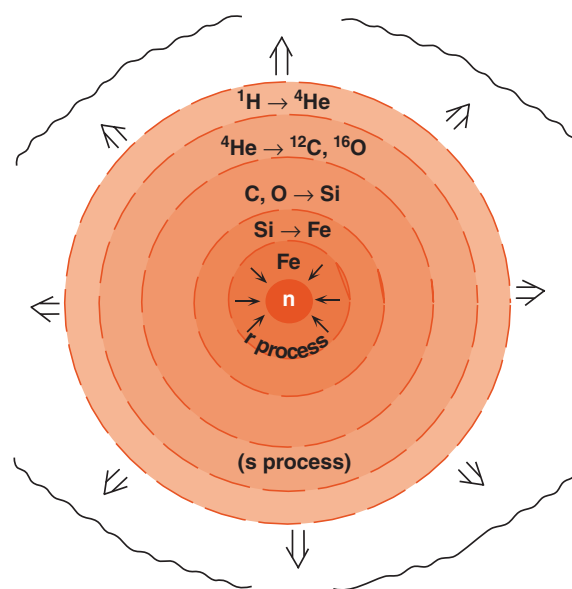
While these reactions can go either direction, there is some tendency for the build up of heavier nuclei with masses 32, 36, 40, 44, 48, 52, and 56. Partly as a result of the *e*-process, these nuclei are unusually abundant

in nature. In addition, because of a variety of nuclei produced during C and Si burning phases, other reactions are possible, synthesizing a number of minor nuclei. The star is now a cosmic onion of sorts (Figure 1.13), consisting of a series of shells of successively heavier nuclei and a core of Fe. Though temperature increases toward the interior of the star, the structure is stabilized somewhat with respect to convection and mixing because the each shell is denser than the one overlying it.

Fe-group elements may also be synthesized by the *e*-process in Type I supernovae. Type I supernovae occur when white dwarfs of intermediate mass (3–10 solar masses) stars in binary systems accrete material from their companion. When their cores reach the Chandrasekhar limit, C burning is initiated and the star explodes. This theoretical scenario has been confirmed in recent years by space based optical, gamma-ray, and X-ray observations of supernovae, such as the Chandra X-ray observatory image in Figure 1.14.

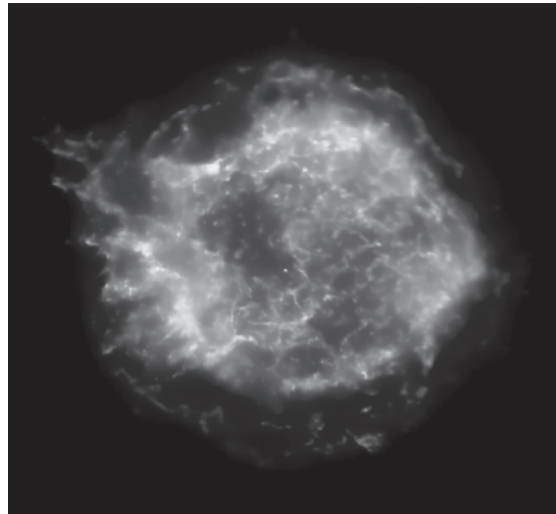
#### 1.4.2.4 The *s*-process

In second and later generation stars containing heavy elements, yet another nucleosynthetic process can operate. This is the slow



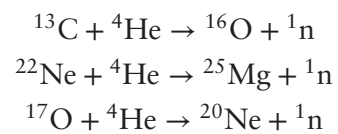
**Figure 1.13** Schematic diagram of stellar structure at the onset of the supernova stage. Nuclear burning processes are illustrated for each stage. (Source: White (2013). Reproduced with permission of John Wiley & Sons.)





**Figure 1.14** Chandra X-ray image of the supernova remnant Cassiopeia A (Cas A). The original Chandra X-ray image of the supernova remnant Cassiopeia A has red, green, and blue regions that show where the intensity of low, medium, and high energy X-rays, respectively, is greatest. The red material on the left outer edge is enriched in iron, whereas the bright greenish white region on the lower left is enriched in silicon and sulfur. In the blue region on the right edge, low and medium energy X-rays have been filtered out by a cloud of dust and gas in the remnant. (Source: Photo: NASA.)

neutron capture or *s-process*. It is so-called because the rate of capture of neutrons is slow, compared to the *r-process*, which we will discuss next. It operates mainly in the red giant phase (as evidenced by the existence of  $^{99}\text{Tc}$  and enhanced abundances of several s-process elements) where neutrons are produced by reactions such as:



(but even H burning produces neutrons; one consequence of this is that fusion reactors will not be completely free of radiation hazards). These neutrons are captured by nuclei to produce successively heavier elements. The principal difference between the r- and s-process (discussed in the following) is the rate of capture relative to the decay of unstable isotopes. In the s-process, a nucleus may only capture a neutron every thousand years or so. If the newly produced nucleus is not stable, it will decay before another neutron is captured. As a result, instabilities cannot be bridged as they can in the r-process discussed next. In the s-process, the rate of formation of stable

species is given by

$$\frac{d[A]}{dt} = f[A-1]\sigma_{A-1} \quad (1.17)$$

where  $[A]$  is the abundance of a nuclide with mass number  $A$ ,  $f$  is a function of neutron flux and neutron energies, and  $\sigma$  is the neutron capture cross section. Note that a nuclide with one less proton might contribute to this build up of nuclide  $A$ , provided that the isobar of  $A$  with one more neutron is not stable. The rate of consumption by neutron capture is:

$$\frac{d[A]}{dt} = -f[A]\sigma_A \quad (1.18)$$

From these relations we can deduce that the creation ratio of two nuclides with mass numbers  $A$  and  $A-1$  will be proportional to the ratio of their capture cross sections:

$$\frac{[A]}{[A-1]} = \frac{\sigma_{A-1}}{\sigma_A} \quad (1.19)$$

Here, we can see that the s-process will lead to the observed odd-even differences in abundance since nuclides with odd mass numbers tend to have larger capture cross sections than even mass number nuclides. The s-process also

explains why magic number nuclides are particularly abundant. This is because they tend to have small capture cross sections and hence are less likely to be consumed in the s-process. The r-process, which we discuss next, leads to a general enrichment in nuclides with  $N$  up to 6–8 greater than a magic number, but not to a build up of nuclides with magic numbers. That the s-process occurs in red giants is confirmed by the overabundance of elements with mainly s-process nuclides, such as those with magic  $N$ , in the spectra of such stars. On the other hand, such stars appear to have normal concentrations of elements with  $25 < A < 75$ , and show normal abundances of r-only elements. Some, however, have very different abundances of the lighter elements, such as C and N, than the Sun.

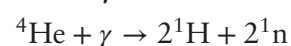
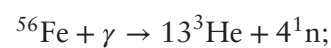
#### 1.4.3 Explosive nucleosynthesis

##### 1.4.3.1 The r-process

The e-process stops at mass 56. In an earlier section, we noted that  $^{56}\text{Fe}$  had the highest binding energy per nucleon, that is, it is the most stable nucleus. Fusion can release energy only up to mass 56; beyond this the reactions become endothermic, that is, they absorb energy. Thus, once the stellar core has been largely converted to Fe, a critical phase is reached: the balance between thermal expansion and gravitation collapse is broken. The stage is now set for the most spectacular of all natural phenomena: a supernova explosion, the ultimate fate of large stars. The energy released in the supernova is astounding: supernovae can emit more energy than an entire galaxy (the recent supernova SN2011fe in the Pinwheel Galaxy, M101, provides an example; at its peak brightness, the supernova was visible with a small telescope, even though the galaxy was not).

When the mass of the iron core reaches 1.4 solar masses (the Chandrasekhar mass), further gravitational collapse cannot be resisted, even by coulomb repulsion. The supernova begins with the collapse of this stellar core, which would have a radius similar to that of the Earth's before collapse, to a radius of 100 km or so. This occurs in a few tenths of a second, with the inner iron core collapsing at 25% of the speed of light. As matter in the center 40% of the core is compressed beyond the density of nuclear matter

( $3 \times 10^{14}$  g/cc), it rebounds, colliding with the outer part of the core, which is still collapsing, sending a massive shock wave back out less than a second after the collapse begins. As the shock wave travels outward through the core, the temperature increase resulting from the compression produces a break down of nuclei by photodisintegration, for example:



This results in the production of a large number of free neutrons (and protons). The neutrons are captured by those nuclei that manage to survive this hell. In the core itself, the reactions are endothermic, and thermal energy cannot overcome the gravitational energy, so it continues to collapse. If the mass of the stellar core is less than 3–4 solar masses, the result is a neutron star, in which all matter is compressed into neutrons. Supernova remnants of masses greater than 3 solar masses can collapse to produce a singularity, where density is infinite. A supernova remnant having the mass of the Sun would form a neutron star of only 15 km radius. A singularity of similar mass would be surrounded by a black hole; a region whose gravity field is so intense even light cannot escape, with a radius of 3 km.

Another important effect is the creation of huge numbers of neutrinos by positron-electron annihilations, which in turn had “condensed” as pairs from gamma rays. The energy carried away by neutrinos leaving the supernova exceeds the kinetic energy of the explosion by a factor of several hundred, and exceeds the visible radiation by a factor of some 30,000. The neutrinos leave the core at nearly the speed of light (and may contribute to the explosive rebound of the star). Although neutrinos interact with matter very weakly, the density of the core is such that their departure is delayed slightly. Nevertheless, they travel faster than the shock wave and are delayed less than electromagnetic radiation. Thus, neutrinos from the 1987A supernova arrived at Earth (some 160,000 years after the event) a few hours before the supernova became visible.

The shock wave eventually reaches the surface of the core, and the outer part of the star is blown apart in an explosion of unimaginable

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violence. Amidst the destruction new nucleosynthetic processes are occurring.

This first of these is the *r process* (rapid neutron capture), and is the principle mechanism for building up the heavier nuclei. In the *r*-process, the rate at which nuclei with mass number  $A + 1$  are created by capture of a neutron by nuclei with mass number  $A$  can be expressed simply as:

$$\frac{dN_{A+1}}{dt} = fN_A\sigma_A \quad (1.20)$$

where  $N_A$  is the number of nuclei with mass number  $A$ ,  $\sigma$  is the neutron capture cross section and  $f$  is the neutron flux. If the product nuclide is unstable, it will decay at a rate given by  $\lambda N_{A+1}$ . It will also capture neutrons itself, so the total destruction rate is given by

$$\frac{dN_{A+1}}{dt} = -fN_{A+1}\sigma_{A+1} - \lambda N_{A+1} \quad (1.21)$$

An equilibrium distribution occurs when nuclei are created at the same rate as they are destroyed, that is:

$$N_A\sigma_A f = \lambda N_{A+1} + N_{A+1}\sigma_{A+1}f \quad (1.22)$$

Thus, the equilibrium ratio of two nuclides  $A$  and  $A + 1$  is:

$$N_A/N_{A+1} = (\lambda + \sigma_{A+1}f)/\sigma_A f \quad (1.23)$$

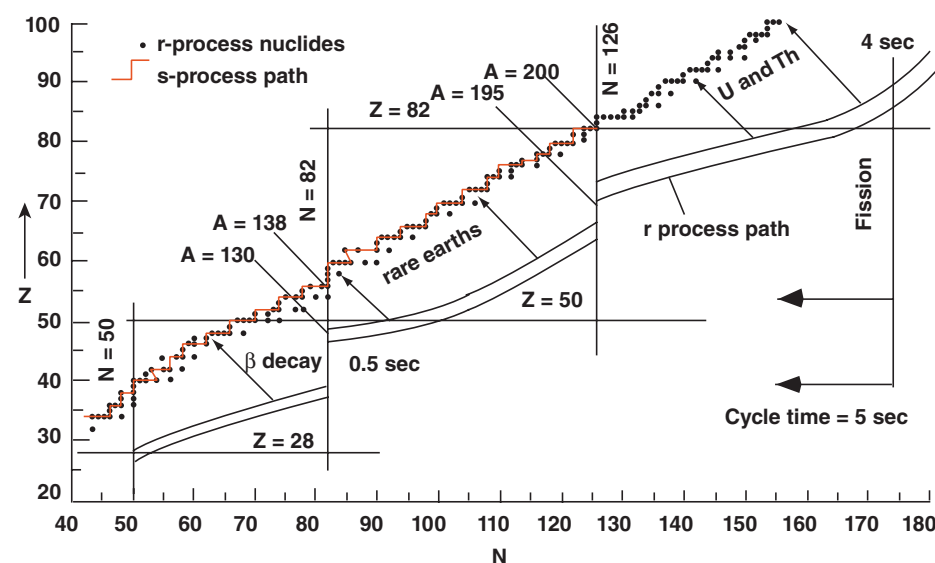
Eventually, some nuclei capture enough neutrons that they are not stable even for short periods (in terms of Equation 1.23,  $\lambda$  becomes large, hence  $N_A/N_{A+1}$  becomes large). They  $\beta^-$  decay to new elements, which are more stable and capable of capturing more neutrons. This process reaches a limit when nuclei beyond  $Z = 90$  are reached. These nuclei fission into several lighter fragments. The *r*-process is thought to have a duration of 100 sec during the peak of the supernova explosion. Figure 1.15 illustrates this process.

During the *r*-process, the neutron density is so great that all nuclei will likely capture a number of neutrons. And in the extreme temperatures, all nuclei are in excited states, and relatively little systematic difference is expected in the capture cross sections of odd and even nuclei. Thus, there is no reason why the *r*-process should lead to different abundances of stable odd and even nuclides.

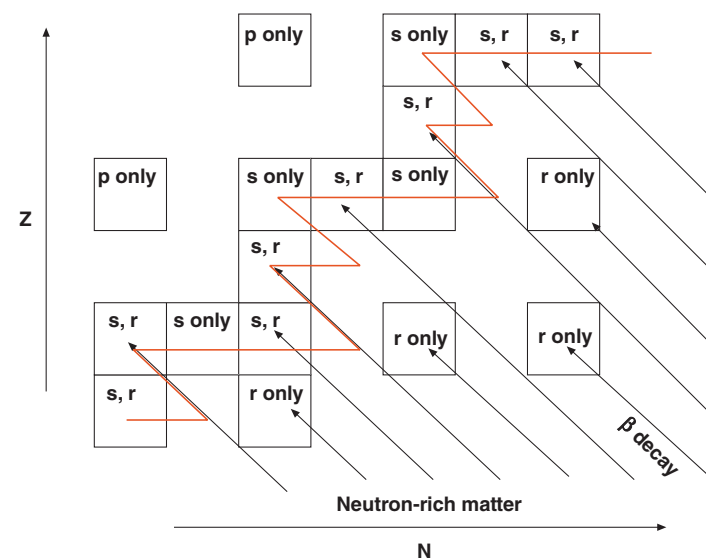
The fact that the *r*-process occurs in supernovae is confirmed by the observation of  $\gamma$ -rays from short-lived radionuclides.

#### 1.4.3.2 The *p*-process

The *r*-process tends to form the heavier isotopes of a given element. The *p*-process



**Figure 1.15** Diagram of the *r* process path on a  $Z$  versus  $N$  diagram. Dashed region is *r*-process path; solid line through stable isotopes shows the *s*-process path. (Source: White (2013). Reproduced with permission of John Wiley & Sons.)



**Figure 1.16** Z versus N diagram showing production of isotopes by the r- s- and p-processes. Squares are stable nuclei; black lines are beta-decay path of neutron-rich isotopes produced by the r-process; solid red line through stable isotopes shows the s-process path. (Source: White (2013). Reproduced with permission of John Wiley & Sons.)

(proton capture) also operates in supernovae and is responsible for the lightest isotopes of a given element. The probability of proton capture is much less likely than neutron capture and hence it contributes negligibly to the production of most nuclides. The reason should be obvious: to be captured the proton must have sufficient energy to overcome the coulomb repulsion and approach to within  $10^{-13}$  cm of the nucleus where the strong nuclear force dominates over the electromagnetic one. Since the neutron is uncharged, there is no coulomb repulsion and even low energy neutrons can be captured. Some nuclides, however, particularly the lightest isotopes of elements, can only be produced by the p-process. These p-process-only isotopes tend to be much less abundant than those created by the s- or r-process.

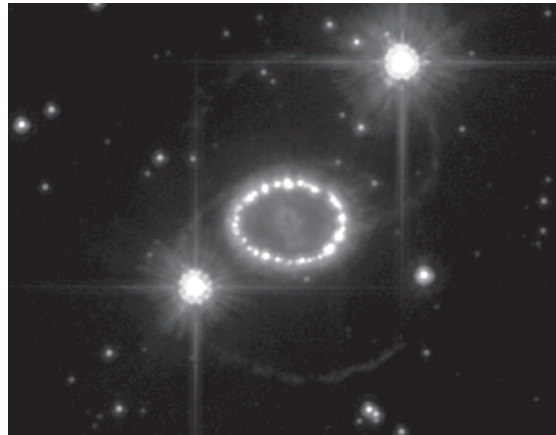
Figure 1.16 illustrates how the s- r- and p-processes create different nuclei. Note also the shielding effect. If nuclide X has an isobar (nuclide with same mass number) with a greater number of neutrons, that isobar will “shield” X from production by the r-process. The most abundant isotopes will be those created by all processes; the least abundant will be those created by only one, particularly by only the p-process.

#### 1.4.3.3 SN 1987A

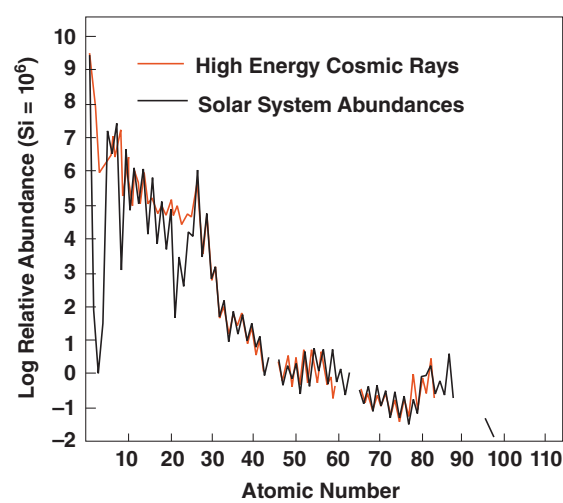
The discussion previously demonstrates the importance of supernovae in understanding the origin of the elements that make up the known Universe. On February 23, 1987, the closest supernova (Figure 1.17) since the time of Johannes Kepler appeared in the Large Magellanic Cloud, a small satellite galaxy of the Milky Way visible in the Southern Hemisphere. This provided the first real test of models of supernovae as the spectrum could be analyzed in detail. Overall, the model presented earlier was reassuringly confirmed. The very strong radiation from  $^{56}\text{Co}$ , the daughter of  $^{56}\text{Ni}$  and parent of  $^{56}\text{Fe}$  was particularly strong confirmation of the supernova model. There were of course, some minor differences between prediction and observation (such as an overabundance of Ba), which provided the basis for refinement of the model.

#### 1.4.4 Nucleosynthesis in interstellar space

Except for production of  $^7\text{Li}$  in the Big Bang, Li, Be, and B are not produced in any of the earlier situations. One clue to the creation of these elements is their abundance in galactic cosmic rays: they are over abundant by a factor of  $10^6$ , as is illustrated in Figure 1.18. They



**Figure 1.17** Rings of glowing gas surrounding the site of the supernova explosion named Supernova 1987A photographed by the Hubble Space Telescope in 2006. The shock wave produced by the supernova explosion is slamming into the ring, about a light-year across, that was probably shed by the star about 20,000 years before it exploded. (Source: NASA photo.)



**Figure 1.18** Comparison of relative abundances in cosmic rays and the solar system. (Source: White (2013). Reproduced with permission of John Wiley & Sons.)

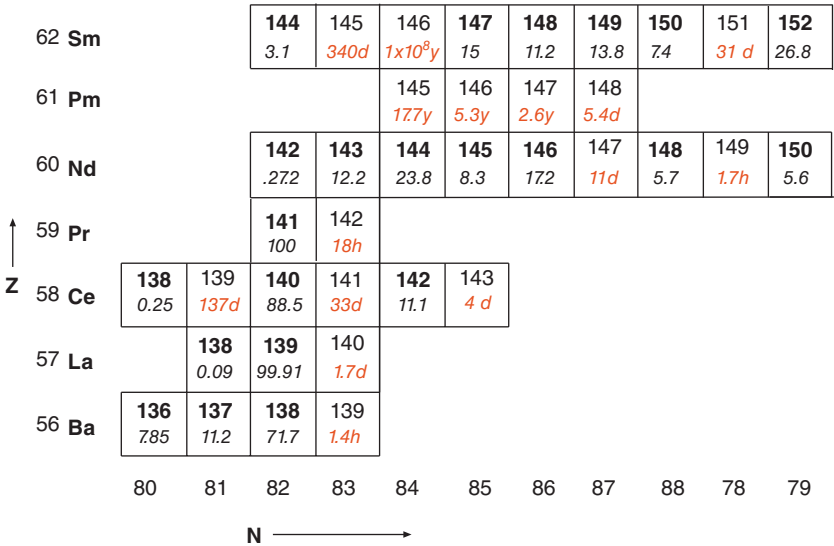
are believed to be formed by interactions of cosmic rays with interstellar gas and dust, primarily reactions of  $^1\text{H}$  and  $^4\text{He}$  with carbon, nitrogen and oxygen nuclei. These reactions occur at high energies (higher than the Big Bang and stellar interiors), but at low temperatures where the Li, B, and Be can survive.

#### 1.4.5 Summary

Figure 1.19 is a part of the  $Z$  versus  $N$  plot showing the abundance of the isotopes of elements 56 through 62. It is a useful region

of the chart of the nuclides for illustrating how the various nucleosynthetic processes have combined to produce the observed abundances. First we notice that even number elements tend to have more stable nuclei than odd numbered ones – a result of the greater stability of nuclides with even  $Z$ , and, as we have noted, a signature of the  $s$ -process. We also notice that nuclides “shielded” from  $\beta^-$  decay of neutron-rich nuclides during the  $r$ -process by an isobar of lower  $Z$  are underabundant. For example,  $^{147}\text{Sm}$  and  $^{49}\text{Sm}$  are more abundant than  $^{148}\text{Sm}$ , even though the





**Figure 1.19** View of part of chart of the nuclides. Mass numbers of stable nuclides are shown in bold, their isotopic abundance is shown in italics as percentages. Mass numbers of short-lived nuclides are shown in plain text with their half-lives also given.

former have odd mass numbers and the latter an even mass number.  $^{138}\text{La}$  and  $^{144}\text{Sm}$  are rare because they “p-process only” nuclides: they are shielded from the r-process and also not produced by the s-process.  $^{148}\text{Nd}$  and  $^{150}\text{Nd}$  are less abundant than  $^{146}\text{Nd}$  because the former are r-process only nuclides while the latter is by both s- and p-processes. During the s-process, the flux of neutrons is sufficiently low that any  $^{147}\text{Nd}$  produced decays to  $^{147}\text{Sm}$  before it can capture a neutron and become a stable  $^{148}\text{Nd}$ .

NOTES

1. We define “fractionation” as any process in which two substances, in this case two isotopes of the same element, behave differently. Thus, fractionation is a process that causes the relative abundances of these substances to change.
2.  $u$  has been the internationally accepted symbol for this unit since 1961. However, the older abbreviation,  $amu$  is still often used (even though its defined value is slightly different).

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*Dalton* is a newer, but still not fully accepted, name for this unit now commonly used by biochemists and likely to be officially adopted in the future.

3. The mass of the neutrino remains a subject of active research in physics. Results of the most recent experiments indicate that it is non-zero, but extremely small; no more than a few electron volts. By comparison, the mass of the electron is about 0.5 MeV.
4. Ga is an abbreviation for giga-annum or  $10^9$  years. Other such abbreviations we will use in this book are a, years, Ma,  $10^6$  years, and ka,  $10^3$  years.
5. It was originally believed that stars evolved from hot and bright to cold and dark across the diagram, hence the term “main sequence.” This proved not to be the case. Stars do, however, evolve somewhat to hotter and brighter during the main sequence part of these lives. The Sun is now about 30% brighter than it was when it first reached the main sequence; this, however, is small compared to the orders of magnitude range in luminosity.

## 30 ISOTOPE GEOCHEMISTRY

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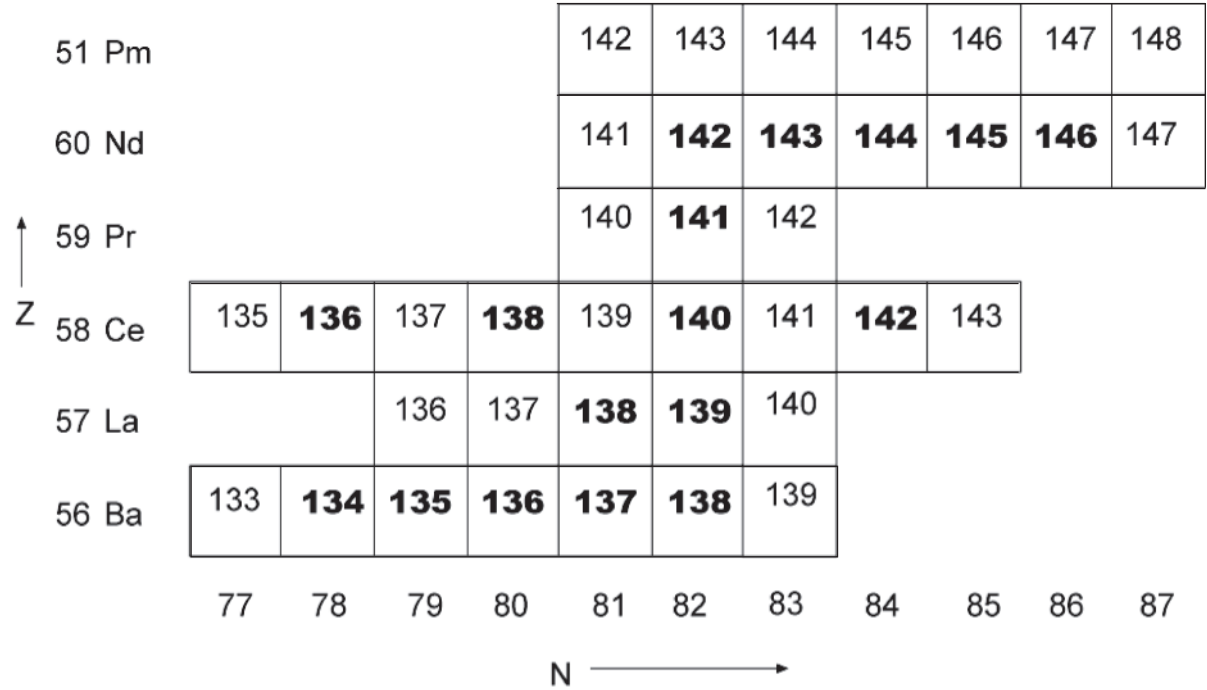
## PROBLEMS

Useful facts for these problems:

Avagadro's number is  $6.02252 \times 10^{23} \text{ (mole)}^{-1}$   
 Speed of light:  $c = 2.997925 \times 10^8 \text{ m/sec}$   
 $1 \text{ eV} = 1.60218 \times 10^{-19} \text{ J}$ ;  $1 \text{ J} = 1 \text{ kg m}^2/\text{sec}^2$ ;  $1 \text{ J} = 1 \text{ VC}$   
 electron charge:  $q = 1.6021 \times 10^{-19} \text{ C (coloumb)}$   
 $1 \text{ u} = 1.660541 \times 10^{-27} \text{ kg}$   
 $1 \text{ Gauss} = 2.997925 \times 10^4 \text{ J/mC}$

- (a) How many moles of Nd (AW = 144.24 u; u is unified atomic mass units) are there in 50 g of Nd<sub>2</sub>O<sub>3</sub> (AW of oxygen is 15.999 u)?  
 (b) How many atoms of Nd in this?
- Given an electron and positron of equal energy, how much more energy is the positron capable of depositing in a detector?
- What are the binding energies per nucleon of <sup>87</sup>Sr (mass = 86.908879 u) and <sup>143</sup>Nd (mass = 86.90918053 u)?
- What is the total energy released when <sup>87</sup>Rb (mass = 86.909183) decays to <sup>87</sup>Sr (mass = 86.908879 u)?
- Using the equation and values for the liquid-drop model, predict the binding energy per nucleon for <sup>4</sup>He, <sup>56</sup>Fe, and <sup>238</sup>U (ignore even-odd effects).
- How many stable nuclides are there with N = 82? List them. How many with N = 83? List them too. Why the difference?
- Calculate the maximum β<sup>−</sup> energy in the decay of <sup>187</sup>Re to <sup>187</sup>Os. The mass of <sup>187</sup>Re is 186.9557508 u; the mass of <sup>187</sup>Os is 186.9557479 u.
- 28% of <sup>228</sup>Th atoms decay to <sup>224</sup>Ra by emitting an alpha of 5.338 MeV. What is the recoil (kinetic) energy of the <sup>224</sup>Re atom? Is the <sup>224</sup>Ra in its ground state? If not, what is nuclear energy in excess of the ground state? (Mass of <sup>228</sup>Th 228.0288 u, mass of <sup>224</sup>Ra is 224.0202 u, mass of alpha is 4.002603 u).

9. A section of the chart of the nuclides is shown next. Mass numbers of stable isotopes are shown in bold; unstable nuclides, shown in plain typeface, can be assumed to be short-lived. The chart shows all nuclides relevant to the questions next.
- (a) Show the s-process path beginning with  $^{134}\text{Ba}$ .
  - (b) Identify all nuclides created, in part or in whole, by the r-process.
  - (c) Identify all nuclides created *only* by the p-process.
  - (d) Which of the stable nuclides shown should be least abundant and why?
  - (e) Which of the cerium (Ce) isotopes shown would you expect to be most abundant and why? (Your answer *may* include more than one nuclide in d and e.)



Detailed information on the nuclides can be found on a web version of the Chart of the Nuclides maintained by Brookhaven National Laboratory Site ([www.nndc.bnl.gov/chart/](http://www.nndc.bnl.gov/chart/)).

10. Both  $^{122}\text{Te}$  and  $^{123}\text{Te}$  are created only in the s-process.  $^{122}\text{Te}$  constitutes 2.55% of Te and  $^{123}\text{Te}$  constitutes 0.89% of Te. What was the ratio of capture cross sections of these nuclides during the s-process? (Your answer might differ from capture cross sections listed in tables as the probability of neutron capture varies with neutron energy.)
11. What is the recoil energy of  $^{208}\text{Tl}$  (mass = 207.9820187 u) in the 5.601 MeV alpha decay illustrated in Figure 1.7?
12. A certain radionuclide emits radiation at the rate of 15.0  $\mu\text{W}$  at one instant of time and at 1.0  $\mu\text{W}$  (microwatts) 1 h later. What is its half-life?