

Chapter 1. Fundamentals of uranium enrichment

1. Isotopes

The atoms of all chemical elements consist of a very small nucleus surrounded by a cloud of electrons. The nucleus accounts for all but a tiny fraction of the mass of the atom, but is confined to a volume whose diameter is only one ten-thousandth of the diameter of the atom itself. Since all the reactions between atoms, that is, all of chemistry, take place at the outer fringes of the electron cloud, it is not difficult to understand why the structure of the nucleus, aside from its total electric charge, has only very minor effects on chemical processes.

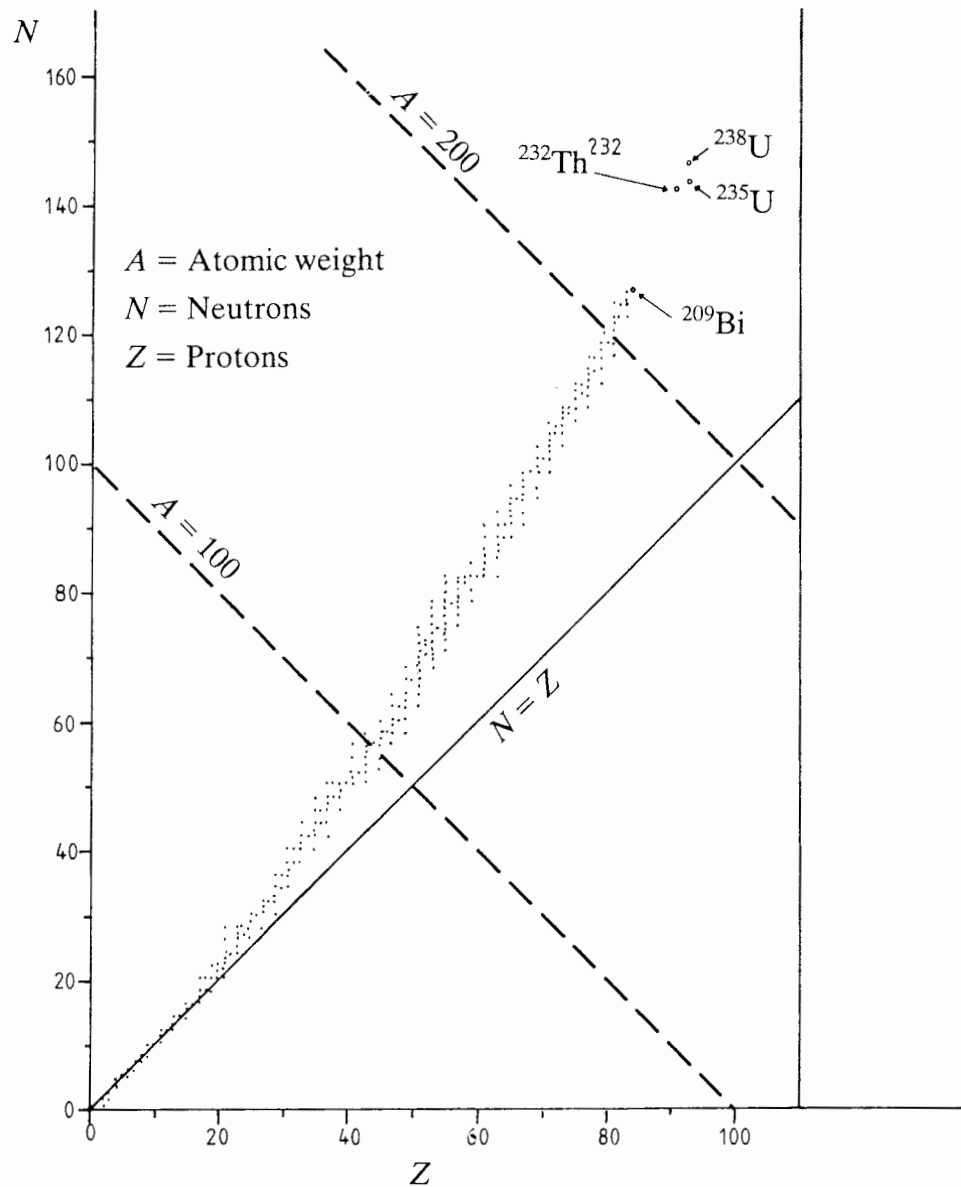
The structure of the nucleus is determined by the numbers of its two constituents, protons and neutrons. Figure 1.1 shows a graph of the stable, naturally occurring elements in which the number of neutrons, N , is plotted against the number of protons, or atomic number, Z [1]. It is the latter value which determines how the element behaves chemically, while the former affects only the mass and detailed structure of the nucleus. A chemical element is determined by its value of Z , and an element can have several isotopes characterized by different values of N .

Figure 1.1 shows that not all combinations of N and Z are possible, and that the stable isotopes are confined to a relatively narrow band of stability. For low numbers of protons the number of neutrons tends to be close to Z , but as the number of protons increases, the number of neutrons needed to ensure stability increases more rapidly. For example, in a stable isotope of lead there are 82 protons and 126 neutrons, giving a total of 208 particles. This total number is often designated A and is called the atomic weight. Since protons and neutrons have very nearly equal masses, this value of A is a quite accurate measure of the total mass of the nucleus, and therefore of the entire atom.

The physical explanation for the presence of more neutrons than protons in heavy nuclei lies in the role neutrons play in holding the nucleus

Figure 1.1. The stable isotopes

The stable isotopes are indicated by dots whose co-ordinates are the neutron number N and the proton number Z . Note the increasing ratio of neutrons to protons for heavier nuclei. The heaviest stable isotope is ^{209}Bi , and the isotopes of thorium and uranium shown are present in nature only because of their very long lifetimes.



together. Stability in the nucleus is actually the result of a delicate balancing of several forces, the two most important of which are the attractive nuclear force and the repulsive electrical force. Extra neutrons are needed to provide sufficient attractive forces to overcome the mutual repulsion of the protons. By the time the number of protons reaches 80 this balancing becomes quite difficult and, as figure 1.1 shows, there are no stable nuclei beyond $Z = 83$, the single stable isotope of bismuth.

If all elements above bismuth are unstable, then how can the existence of natural uranium (with 92 protons) be accounted for? The explanation is

in the fact that the instability of nuclei can manifest itself over a wide range of lifetimes. There happen to be three particular combinations of protons and neutrons which are so nearly stable that their half-lives (the time in which half of a given sample will decay) are of the order of hundreds of millions or billions of years, comparable to the age of the Earth. These three are: one isotope of thorium, ^{232}Th (with a half-life of 14 billion years); and two isotopes of uranium, ^{235}U (with a half-life of 710 million years) and ^{238}U (with a half-life of 4.5 billion years). These species are also indicated in figure 1.1.

The generally accepted theory of the formation of the Earth is that it condensed from a cloud of material, possibly the remnants of an exploded star. At the time of the Earth's formation the relative amounts of all the elements and their isotopes were determined by whatever nuclear reactions had accompanied the formation of the cloud; but since the formation of the Earth some five billion years ago there has been no significant creation of new heavy elements, and those that solidified with the Earth have been decaying away. After five billion years only the three above-mentioned species remain in any significant quantities. The other heavy elements which are found (e.g., the radium discovered by Marie Curie) are present as a result of the decay of these three.

If the half-lives of the two uranium isotopes¹ are compared with the life of the Earth, an interesting result emerges. First, notice that the age of the Earth is slightly longer than one half-life of ^{238}U . This means that in the early years of the Earth's life there was roughly twice as much ^{238}U present as there is now. But ^{235}U has endured over seven half-lives in the same period, which implies that in the early days there was about 2^7 or 128 times as much as there is now. At present natural uranium consists of 99.3 per cent ^{238}U and only 0.7 per cent ^{235}U , but if these values are extrapolated backwards in time, it can be shown that primordial natural uranium must have contained almost 30 per cent ^{235}U .

From this it is clear that if the Earth had been inhabited soon after its formation, nuclear energy would have been quickly and easily discovered. Actually, long before human life appeared on the planet, a natural nuclear reactor went critical in a rich uranium seam under what is now Gabon in West Africa [2]. This event took place about 1.7 billion years ago when natural uranium was slightly more than 3 per cent enriched in ^{235}U , a value practically identical to that employed today in most light water nuclear reactors (LWRs). This book deals with the elaborate and costly methods which have had to be devised to make up for the arrival of human beings two billion years too late to take advantage of this situation.

An important mechanism of radioactive decay in heavy nuclei is nuclear fission. In this process the electrical repulsive forces win out over

¹ A third isotope, ^{234}U , is found in very small quantities in natural uranium. This isotope is too short-lived to have survived since the Earth's formation. It may be a minor product of the decay of ^{238}U .

the nuclear attractive forces, and the nucleus breaks apart into two or more pieces, usually accompanied by the release of a few neutrons. This fission can take place either spontaneously, like other radioactive processes, or it can be induced by the absorption of energy or another particle. There are several nuclei which undergo fission after absorbing a neutron, but generally a rather energetic neutron is required to induce fission.

Of the three naturally occurring, long-lived nuclear species mentioned above, only one, ^{235}U , undergoes nuclear fission when bombarded with slow neutrons. This property is important for the maintenance and control of a sustained, as opposed to an explosive, nuclear chain reaction (i.e., a reaction which is capable of sustaining itself once it has been started). The other two species, ^{232}Th and ^{238}U , can absorb neutrons and be converted (bred) into other forms (^{233}U and ^{239}Pu) which also fission in the presence of slow neutrons, but this would probably never have been more than a scientific curiosity if ^{235}U had not been available in natural form. Only the chain reaction in ^{235}U could have provided at feasible costs the enormous numbers of neutrons needed to breed sufficient quantities of the other isotopes to begin a nuclear fuel cycle or to produce nuclear weapons. Depending on how one assesses the impact of nuclear energy on human history, one can see this remarkably anomalous phenomenon as a stroke of either extremely good or extremely bad luck.

It is possible to maintain a chain reaction in natural uranium. This was in fact what was done by all the countries which contributed to the early development of nuclear energy – Canada, France, the UK, the USA and the USSR. The very small concentration of ^{235}U in natural uranium enforces very stringent requirements for neutron concentration in such a reactor. Neutrons emitted in a fission reaction must be slowed down by many collisions with nuclei in the surrounding medium, called the moderator. If too many are absorbed during the slowing down process, not enough will be left to sustain the chain reaction. Only very pure carbon (graphite) or heavy water (D_2O)² have been found sufficiently resistant to neutron absorption, and producible in sufficient quantities at acceptable cost, to serve as moderators in natural uranium reactors.

It was realized very soon after the discovery of nuclear fission in 1939 that it might be possible to make a powerful nuclear explosive by extracting and concentrating the ^{235}U from natural uranium. It also became clear that if the uranium could be enriched in this isotope it would be possible to substitute ordinary water for the expensive graphite or heavy water in a reactor. These motivations, particularly the first, provided the early impetus for devising means for producing enriched uranium. Later on it was learned that very pure ^{235}U was also extremely useful, if not essential, for the design of thermonuclear explosives. Given these objectives, and the world political situation which emerged from World War II, it is not

² “D” is the symbol for deuterium, an isotope of hydrogen containing one proton and one neutron.

surprising that a great deal of scientific and technological creativity was devoted to the problem of enriching uranium. This book is concerned with some of the results and implications of this creativity.

II. Uses for enriched uranium

As has already been mentioned, even natural uranium can be useful in certain types of nuclear reactors, but, as the proportion of ^{235}U is increased, the ability to use uranium as an energy source becomes more flexible and technically less complicated. Modern light water reactors use uranium enriched to about 3 per cent, although this can vary anywhere between 2.5 and 3.5 per cent depending on the type and operating schedule of the reactor.

As the percentage of ^{235}U increases, the size of reactor for a given power level can decrease, that is, the power density in the core of the reactor increases. Reactors used for ship propulsion use enrichments of at least 10 per cent in order to keep the size of the power plant relatively small. Nuclear submarines, satellites, and many small research reactors use fuel enriched to 90 per cent or more [3a]. Because of the high power densities achievable with such enrichments, research reactors generally require only a few kilograms of fuel to achieve criticality (i.e., a self-sustained nuclear reaction).

Below 10 per cent enrichment metallic uranium cannot be made to explode, since the mass which must be assembled for the explosion, the so-called critical mass, is essentially infinite [4a]. However, the critical mass drops rapidly above 10 per cent enrichment, especially if the uranium is surrounded by a good neutron reflector. This material serves to return neutrons which would otherwise be lost back into the uranium to create more fission reactions. It can also serve to hold the critical mass together for a longer time in order to increase the power of the explosion [5].

Table 1.1. Critical masses of uranium for varying degrees of enrichment^a

Enrichment (per cent)	Critical mass of uranium-235 (kg)
10	100
20	50
50	25
100	15

^a The critical mass values, which assume a good neutron reflector, are taken from reference [4b].

By the time enrichment reaches 20 per cent, uranium can be considered a highly sensitive material. This enrichment has in fact been chosen by the International Atomic Energy Agency (IAEA) as the line above which enriched uranium is treated in the same category as plutonium, the other nuclear explosive material [6a]. Table 1.1 shows that the critical mass required for a nuclear weapon drops rapidly above enrichments of 20 per cent, and that even enrichments as low as 50 per cent must be viewed as potentially dangerous. However, as the rest of this chapter and the next will show, the production of even relatively small amounts of highly enriched uranium is no simple task.

III. Isotope separation

Uranium enrichment is a special case of the much more general problem of separation of isotopes. There are many applications, for example, in biological research and medicine, for which it is necessary or desirable to obtain purified samples of a particular isotope; but, generally the amounts required are extremely small: a gram of purified isotope is considered a large amount in a biological laboratory. Only for uranium and hydrogen have people found it necessary to perform isotope separation on quantities of material measured in millions of kilograms.

Since the number of neutrons in a nucleus has only minute effects on the chemistry of an atom, the separation of isotopes cannot be accomplished by the use of chemical techniques of the kind normally used to purify substances. The separation must somehow take advantage of those properties of the nucleus which are affected by the number of neutrons: its mass, size and shape, magnetic moment or angular momentum. By far the most important of these is the difference in mass, which is utilized in all of the proven enrichment processes and all but one of the advanced methods currently under development.

There are two broad classes of effects which are sensitive to the difference in mass of uranium isotopes. The first relies on the differences in the average speed of atoms or molecules with different masses which are mixed together in thermal equilibrium. The second utilizes the different inertias, that is, resistance to acceleration, of different masses when they are subjected to the same force. Lighter atoms or molecules move faster on the average than heavier ones in mixtures at a given temperature, and lighter molecules respond more readily (i.e., accelerate faster) than heavier ones when a force is applied to them.

In the first class can be placed all of the currently demonstrated separation techniques: gaseous diffusion, centrifugation and aerodynamic techniques such as the jet nozzle process and the vortex tube process. All of these processes operate on uranium in the form of uranium hexafluoride gas, UF_6 .

At any temperature the molecules of a gas will be in constant motion, colliding with each other millions of times per second and exchanging energy each time they collide. All of these collisions ensure that the energy of motion of the molecules (their kinetic energy) is equally shared among them on the average. This equal sharing of average kinetic energy is one of the fundamental principles of the kinetic theory of gases, or more generally, the branch of physics called statistical mechanics which deals with systems containing very large numbers of particles.

If two particles have the same kinetic energy, the one with the smaller mass will have the larger velocity. In the gaseous diffusion process this difference is exploited by allowing the gas to diffuse through a solid barrier permeated by many small holes or pores. The faster-moving molecules pass through the holes more frequently, and the mixture which emerges on the other side of the barrier is therefore somewhat richer in the light species than the original sample.

The centrifuge and aerodynamic methods also use diffusion, but add strong accelerations to magnify the effect. In all of these methods the gas is accelerated into rapid rotation creating a centrifugal force field in the gas which accelerates particles towards the periphery of the circle.

This force has a property similar to gravity in that it accelerates particles at a rate which is independent of their masses. For example, when a car turns a sharp corner, everything and everybody in the car is thrown to the outside of the turn at the same rate. It follows that centrifugal force by itself cannot be used to separate isotopes. All it can do is create a pressure variation in the gas, against which particles of different masses will diffuse at different rates. It is this differential diffusion against a pressure gradient which underlies the centrifuge, nozzle and vortex tube techniques.

There are forces which do accelerate particles of different masses at different rates. In this class are electromagnetic forces, interatomic binding forces and the forces between colliding atoms or molecules. Actually all of these are electromagnetic in origin, but it is more convenient here to treat them separately.

If an atom or molecule can be ionized, that is, given an electric charge, it can then be accelerated by either electric or magnetic fields or both. The earliest successful uranium isotope separating device utilized strong magnetic fields to deflect beams of ^{235}U and ^{238}U ions by slightly different amounts. This device, called the calutron, produced the highly enriched ^{235}U which destroyed Hiroshima in 1945. Another technique which was suggested in the 1940s, but which has only recently begun to show some promise of success, is the use of oscillating electromagnetic fields to accelerate ionized atoms in circles, using a phenomenon called ion cyclotron resonance.

Interatomic forces hold molecules together, and a useful model of a chemical bond is a small spring connecting two masses. This spring can be stretched and compressed, so that the masses can oscillate with certain characteristic frequencies. These frequencies tend to lie in the infra-red

part of the electromagnetic spectrum, and the vibrations can be excited either by collisions with other molecules or by absorption of infra-red radiation at well-defined frequencies. The vibration frequency depends on the stiffness of the spring (i.e., the nature of the chemical bond) and the values of the masses; but since the chemical bonds formed by two isotopes are virtually identical, the masses alone determine the vibration frequency shifts between molecules containing different isotopes.

The principle of molecular laser isotope separation (MLIS) is to tune an infra-red laser to a precise vibration frequency of $^{235}\text{UF}_6$. Then, only these molecules are excited into vibration, and once they are, they can be separated in any of a number of ways by ionization, dissociation, or chemical reactions. Recent proposals also suggest that this method of selective infra-red excitation can be used as a way of giving the molecules more thermal energy and utilizing this to enhance various diffusion effects. The difference in vibration frequencies also leads in a rather subtle way to a tendency for light isotopes to concentrate in certain chemical compounds in preference to others. To exploit this effect the two chemicals must be brought into contact with each other and encouraged, usually with catalysts, to exchange uranium atoms. When this exchange reaches

Table 1.2 Uranium enrichment techniques, according to physical principles and mechanisms used

Property of isotope	Physical principle	Physical mechanism	Separation process
Nuclear mass	Newton's law of acceleration	Acceleration by electro-magnetic forces	Calutron Plasma centrifuge Ion cyclotron resonance
		Shift in chemical equilibrium	Chemical exchange
	Equipartition of energy	Selective absorption of infra-red light	Molecular laser isotope separation (MLIS)
		Flow through porous barrier	Gaseous diffusion
		Diffusion against pressure gradient	Centrifuge Jet nozzle Vortex tube
Nuclear size, shape, spin, magnetism	Quantum theory of atomic structure	Selective absorption of visible or ultraviolet light	Atomic vapour laser isotope separation (AVLIS)

equilibrium a slight, but significant, concentration of isotopes is sometimes achievable. This is the basis of a number of chemical-exchange techniques which are currently under development.

There is one isotope separation process which does not depend very much on the nuclear mass difference. This is atomic vapour laser isotope separation (AVLIS) which utilizes very small shifts in the frequencies at which uranium atoms absorb light. These shifts are caused only partially by the different masses of the two nuclei; more important are the small changes the extra neutrons make in the magnetic properties of the nucleus as well as in its size and shape. These changes are very small, and it was only the advent of the laser which made possible the selective use of light to distinguish such small frequency shifts. The process uses laser light to excite and ionize only ^{235}U atoms in a chamber containing metallic uranium vapour. Then the ^{235}U ions are collected by electromagnetic fields.

Table 1.2 summarizes this brief introductory discussion by listing the enrichment techniques mentioned above along with the basic principles and mechanisms on which they are based. From this list it is clear that there is no shortage of clever ideas for the separation of uranium isotopes. The basic physical principles underlying most of the above-mentioned methods are really quite simple, and all have been well understood for many years. But the transition from an elegant physical principle to an economically viable industrial technique is usually a long and arduous one. The reasons for this are made clear in Part Two where the practical engineering aspects of these ideas are explored.

IV. Basic principles of enrichment

The basic component of any enrichment facility, no matter which process is used, is the enrichment element (see figure 5.1, p. 95). An element is a device which separates an incoming feed stream into two outgoing streams: a product stream in which the process material is enriched to some degree in the desired isotope and a tails stream (sometimes inappropriately called waste) which is somewhat depleted in this isotope.

The degree of separation which can be achieved in a given element is measured by a parameter called the separation factor. This varies widely from one technique to another, being smallest for the chemical-exchange processes and gaseous diffusion and largest for the modern resonance techniques based on lasers or plasmas. When the separation factor is very small only a small enrichment can be achieved in a single element, and the process material must be passed through many elements in order to achieve useful enrichments. For example, several thousand separate

enrichment elements must be employed in a chemical-exchange facility to enrich uranium from its natural composition of 0.71 per cent ^{235}U to 3 per cent ^{235}U . On the other hand it is possible that an atomic vapour laser enrichment element will be able to obtain the same separation in a single element.

Another important property of any enrichment element is the rate at which it can process feed material. Some elements, such as large gaseous diffusion units, can process tons of material per minute, while others, such as those which use atomic vapour might process only a few grams per minute or even less. Both the separation factor and the flow rate are needed to specify the separative power of an element, which is usually rated in separative work units per year (SWU/yr). For example, typical ratings for modern, high-speed gas centrifuges are from 5 to 100 SWU/yr depending on their design and operating conditions.

An enrichment facility designed to produce fuel for several nuclear reactors must have a total capacity of hundreds of thousands of SWU per year. Such a plant must therefore consist of many elements arranged in a cascade. Such an arrangement is illustrated in figure 5.4, p. 103 which shows a series of stages each consisting of a number of elements. By adding elements to a given stage the flow capacity of the stage can be increased, and by cascading many stages in series any desired enrichment can be achieved in the final product.

In general, it can be said that the smaller the separation factor of a stage the larger the flow rate necessary to achieve a given separative capacity. This can be achieved either by inserting many individual elements in a stage (as is done with centrifuges) or by building very large elements which constitute a stage in themselves (as in gaseous diffusion). The more elements that are required, or the larger their sizes, the larger the size of the facility. Also, the larger and more numerous the elements become, the larger is the process inventory in the plant. Since the inventory must consist of uranium it is an important economic factor in the design of enrichment plants. Large plant sizes and process inventories also make it difficult to hide such facilities or to operate them in ways for which they were not designed.

For most enrichment techniques there is a time delay between when a plant is completed and when useful product can be extracted. The cascade must first be filled with unenriched feed material and then run for a while to allow the concentration of desired isotope to build up at the product end. A standard measure of the time required for this build-up is the equilibrium time of the cascade. This time depends on the final enrichment desired, on the inventory, and on the time required for a sample of material to pass through a typical element. It can range from virtually instantaneous for the laser and plasma processes, to weeks or months for plants with large inventories and small separation factors. An extreme example is the 30–40 year equilibrium time required for obtaining 90 per cent enriched product in one type of chemical-exchange facility.

An enrichment cascade is normally designed to produce product with some desired degree of enrichment from feed with some specified initial isotopic composition. This is usually natural uranium, but in principle a cascade can be fed with material of higher enrichment, and the product of such a cascade will be correspondingly more highly enriched. This leads to the possibility of batch recycling to produce highly enriched material. Batch recycling is generally a wasteful way of achieving high enrichments, but may be an option if obtaining relatively small amounts of highly enriched material is of high priority. The suitability of any technique for batch recycling is obviously related to its equilibrium time and inventory.

There are two more features of a cascade which should be introduced here in preparation for the comparative analysis of the next chapter. The first is reflux and the second is criticality.

Reflux is the process by which part or all of the output of an element is recycled back to the input to achieve higher separation factors. In effect this amounts to running the material through a given element more than once, and the ease with which this can be done depends on the nature of the process material and the enrichment mechanism. If, as in chemical-exchange techniques, the chemical nature of the process material is changed by the enrichment process, then the reflux must consist of a chemical reconversion. This adds considerably to the cost and technical difficulty of chemical enrichment techniques.

Criticality problems arise in any process where enough fissionable material might collect to produce a self-sustaining nuclear reaction (i.e., a critical mass). This is not a serious problem where the process inventory is in gaseous form, but when liquids (especially aqueous or organic solutions) are used, special precautions must be taken to make sure that no critical mass can ever accidentally be assembled.

This completes a brief introduction to the basic principles and terminology of uranium enrichment. With this introduction the non-technical reader should be able to follow the arguments of the next chapter in which the proliferation dangers of the various processes are compared. Readers desiring a more detailed and quantitative discussion of these concepts are referred to Part Two.