Uranium – The Element: Its Occurrence and Uses

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Summary: Uranium metal and its compounds have been of great interest to physicists and chemists due to its use for both civil and military applications, e.g. production of electricity, use in the medical field and for making nuclear weapons. This review paper describes the occurrence, chemistry and metallurgy of the element 'uranium', its conversion to stable compounds such as yellow cake, uranium tetrafluoride and uranium hexafluoride and the enrichment technologies and uses for both civil and military purposes. The paper is meant for ready reference for students and teachers in connection with the recent spate of interest shown in nuclear power generation in Pakistan and abroad.

Keywords: Uranium, Chemistry, Metallurgy, Enrichment, Civil uses, Military uses, Health hazards.

Introduction

Recent interest in nuclear power, both in Pakistan and abroad, has aroused considerable interest in uranium metal and its application to power generation. Moreover, since May 1998, when Pakistan joined the nuclear club using advanced centrifuge uranium enrichment technology, science students and teachers alike are interested in finding out more about uranium and its uses, both civil and military. Hence this tutorial report for ready reference.

Uranium – General Information

As with all subjects and technologies, there are a large number of books and articles available on uranium metal, its mining, refining, radioactivity, uses, etc. Some websites also contain information on this metal, but they cannot always be accepted as authentic. Hence information on uranium metal, its chemistry, metallurgy and uses, both civilian and military, has been collected from reliable books and scientific journals. The references [1-4] contain useful general information on the subject under review.

Occurrence

Uranium was apparently formed in supernovas about 6.6 billion years ago. While it is not common in the solar system, its slow radioactivity (together with the decay of thorium and potassium-40) in the earth's crust is thought to be the main source of heat that keeps the outer core liquid and drives mantle convection, which in turn drives plate tectonics – Fig. 1 [1].

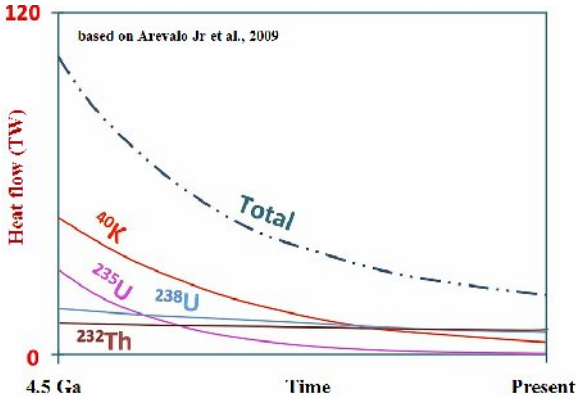

Fig. 1: The evolution of the earth's radiogenic heat flow over time: contribution from 235-U in pink and from 238-U in light blue. Source: Biever, Celeste. First Measurements of the Earth's Core. *New Scientist* July 2005.

Discovery

The discovery of the element is credited to the German chemist, Martin Heinrich Klaproth [5]. While he was working in his experimental laboratory in Berlin in 1789, he was able to precipitate a yellow compound (likely sodium diuranate) by dissolving pitchblende Fig. 2 in nitric acid and neutralizing the solution with sodium hydroxide Fig. 2. He assumed the yellow substance was the oxide of a yet undiscovered element and heated it with charcoal to obtain a black powder, which he thought was the newly discovered metal itself (in fact, that powder was an oxide of uranium). He named the newly discovered element after the planet Uranus, (named

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after the primordial Greek god of the sky), which had been discovered eight years earlier by William Herschel.

Fig. 2: Uraninite, also known as pitchblende, is the most common ore mined to extract uranium [4].

In 1841, Eugène-Melchior Péligot, Professor of Analytical Chemistry at the Conservatoire National des Arts et Métiers (Central School of Arts and Manufactures) in Paris, isolated the first sample of uranium metal by heating uranium tetrachloride with potassium. [6] Uranium was not seen as being particularly dangerous during much of the 19th century, leading to the development of various uses for the element, like the coloring of pottery and glass Fig. 3 [6].

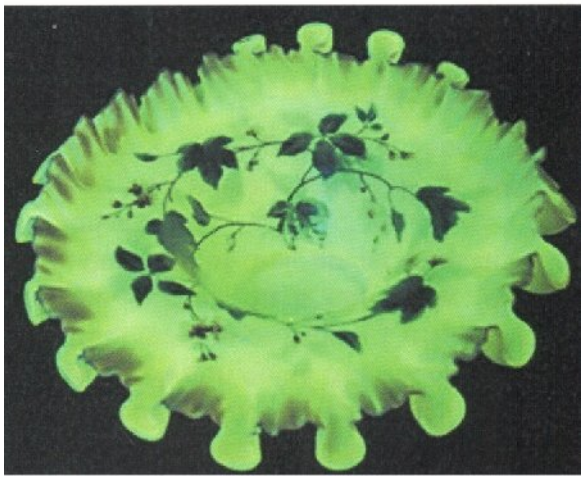


Fig. 3: Uranium glass glowing under UV light. Source: Skelcher, Barrie. Vaseline Glassware. Schiffer Publishing 2007.

Henri Becquerel discovered radioactivity by using uranium in 1896. He made the discovery in Paris by leaving a sample of a uranium salt, $K_2UO_2(SO_4)_2$ (potassium uranyl sulfate), on top of an unexposed photographic plate in a drawer and noting that the plate had become "fogged" Fig. 4. He determined that a form of invisible light or rays emitted by uranium had exposed the plate. [7]

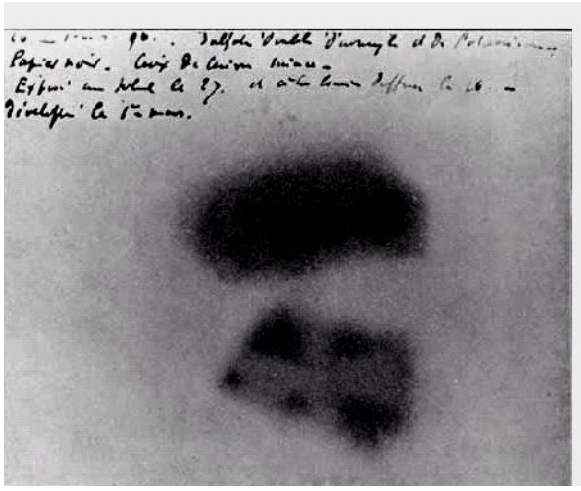


Fig. 4: Antoine Henri Becquerel discovered the phenomenon of radioactivity by exposing a photographic plate to uranium in 1896 [7].

Prehistoric Naturally Occurring Fission: Natural Nuclear Fission Reactor

In 1972 the French physicist, Francis Perrin, discovered fifteen ancient and no longer active natural nuclear fission reactors in three separate ore deposits at the Oklo mine in Gabon, West Africa, collectively known as the Oklo Fossil Reactors. [8] The ore deposit is 1.7 billion years old; then uranium-235 constituted about 3% of the total uranium on earth. This is high enough to permit a sustained nuclear fission chain reaction to occur, provided other supporting conditions exist. The capacity of the surrounding sediment to contain the nuclear waste products has been cited by the U.S. Federal Government as supporting evidence for the feasibility to store spent nuclear fuel at the Uycca Mountain nuclear waste repository.

Pre-Discovery Use

The use of uranium in its natural oxide form dates back to at least the year 79 CE, when it was used to add a yellow color to ceramic glazes. Yellow glass with 1% uranium oxide was found in a Roman villa on Cape Posillipo in the Bay of Naples, Italy by

R.T. Gunther of the University of Oxford in 1912. Starting in the late Middle Ages, pitchblende was extracted from the Habsburg silver mines in Joachimsthal, Bohemia (now Jáchymov in the Czech Republic), and was used as a coloring agent in the local glassmaking industry. In the early 19th century, the world's only known sources of uranium ore were these mines.

Presence in Environment

Uranium is a naturally occurring element that can be found in low levels within all rocks, soil and water. Uranium is the 51st element in order of abundance in the earth's crust. It is also the highest-numbered element to be found naturally in significant quantities on earth and is almost always found combined with other elements [3].

Uranium's average concentration in the earth's crust is (depending on the reference) 2 to 4 parts per million, or about 40 times as abundant as silver. The earth's crust from the surface to 25 km (15 miles) down is calculated to contain 10^{17} kg (2 x 10^{17} lb) of uranium while the oceans may contain 10^{13} kg (2 x 10^{13} lb). The concentration of uranium in soil ranges from 0.7 to 11 parts per million (up to 15 parts per million in farm land soil due to the use of phosphate fertilizers), and its concentration in sea water is 3 parts per billion [3].

Uranium is more plentiful than antimony, tin, cadmium, mercury or silver and it is about as abundant as arsenic or molybdenum. Uranium is found in hundreds of minerals, including uraninite (the most common uranium ore), carnotite, autunite, uranophane, torbernite and coffinite. Significant concentrations of uranium occur in some substances such as phosphate rock deposits and minerals such as lignite and monazite sands in uranium-rich ores (it is recovered commercially from sources with as little as 0.1% uranium) [3].

In nature, uranium(VI) forms highly soluble carbonate complexes at alkaline pH. This leads to an increase in mobility and availability of uranium to groundwater and soil from nuclear wastes, which leads to health hazards. However, it is difficult to precipitate uranium as phosphate in the presence of excess carbonate at alkaline pH. A Sphingomonas sp. Strain, BSAR-1, has been found to express a high activity alkaline phosphatase (PhoK) that has been applied for bioprecipitation of uranium as uranyl phosphate species from alkaline solutions. The precipitation ability was enhanced by overexpressing PhoK protein in E. coli [9].

Production and Mining

Worldwide production of uranium in 2010 amounted to 53,663 tonnes, of which 17,803 tonnes (33.2%) was mined in Kazakhstan. Other important uranium mining countries are Canada (9,783 tonnes), Australia (5,900 tonnes) Namibia (4,496 tonnes), Niger (4,198 tonnes) and Russia (3,562 tonnes) [10].

Uranium ore is mined in several ways: by open pit, underground, in-situ leaching and borehole mining. Mined, low-grade uranium ore mined typically contains 0.01 to 0.25% uranium oxides. Extensive measures must be employed to extract the metal from its ore. High-grade ores found in the Athabasca Basin deposits in Saskatchewan, Canada, can contain up to 23% uranium oxides on average. Uranium ore is crushed and rendered into a fine powder and then leached with either an acid or an alkali. The leachate is subjected to one of several sequences of precipitation, solvent extraction and ion exchange. The resulting mixture, called yellowcake - Fig. 5) [1], contains at least 75% uranium oxides U_3O_8. Yellowcake is then calcined to remove impurities from the milling process before refining and conversion [4].

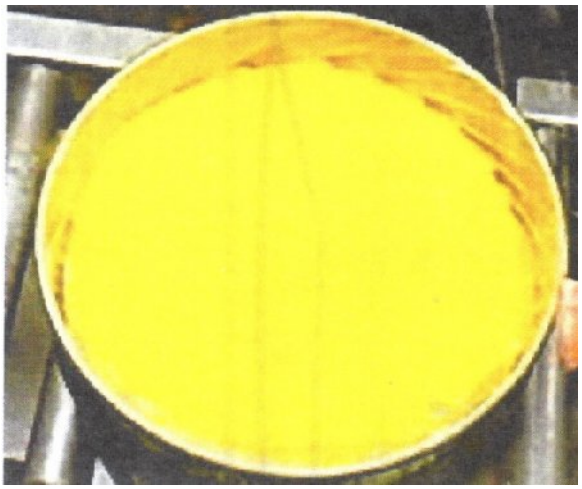


Fig. 5: A drum of yellowcake, a mixture of uranium precipitates. Source: http://encyclopedie-electricite.edf.com.

Commercial-grade uranium can be produced through the reduction of uranium halides with alkali or alkaline earth metals. Uranium metal can also be prepared through electrolysis of KUF_5 or UF_4 dissolved in molten calcium chloride ($CaCl_2$) and sodium chloride (NaCl) solution. Very pure uranium is produced through thermal decomposition of uranium halides on a hot filament [4].

Resources and Reserves

It is estimated that 5.5. Million tonnes of uranium exists in ore reserves that are economically viable at US $ 59 per lb. of uranium [11], while 35 million tones are classed as mineral resources (reasonable prospects for eventual economic extraction). Prices went from about $ 10/lb. in May 2003 to $ 138/lb. in July 2007. This has caused a big increase in spending on exploration, with US $ 200 million being spent worldwide in 2005, an increase of 54% on the previous year. This trend continued through 2006, when expenditure on exploration rocketed to over $ 774 million, an increase of over 250% compared to 2004. The Organisation for Economic Co-operation and Development (OECD) of the Nuclear Energy Agency said exploration figures for 2007 would likely match those for 2006 [12].

Australia has 31% of the world's known uranium ore reserves and the world's largest single uranium deposit, located at the Olympic Dam Mine in South Australia. There is a significant reserve of uranium in Bakouma, a sub-prefecture in the Prefecture of Mbomou in the Central African Republic.

Some nuclear fuel comes from nuclear weapons being dismantled, such as from the Megatons to Megawatts Program.

An additional 4.6 billion tonnes of uranium are estimated to be in sea water. Japanese scientist showed in the 1980s that extraction of uranium from sea water using ion exchangers was technically feasible [13]. There have been experiments to extract uranium from sea water but the yield has been low due to the presence of carbonate in the water. In 2012, Oak Ridge National Laboratory (ORNL) researchers [14] announced the successful development of a new absorbent material dubbed HiCap, which performs surface retention of solid or gas molecules, atoms or ions and also effectively removes toxic metals from water, according to results verified by researchers at Pacific Northwest National Laboratory [15].

Supplies

In 2005 seventeen countries produced concentrated uranium oxides with Canada (27.9% of world production) and Australia (22.8%) being the largest producers and Kazakhstan (10.5%), Russia (8%), Namibia (7.5%), Niger (7.4%), Uzbekistan (5.5%), the United States (2.5%), Argentina (2.1%), Ukraine (1.9%) and China (1.7%) also producing significant amounts. Kazakhstan continues to increase production and may have become the world's largest producer of uranium soon with an expected production of 12,826 tonnes compared to Canada with 11,100 tonnes and Australia with 9,430 tonnes. In the late 1960s, UN geologists also discovered major uranium deposits and other rare mineral reserves in Somalia. The find was the largest of its kind with industrial experts estimating the deposits at over 25% of the world's then known uranium reserves of 800,000 tonnes [16].

The ultimate available uranium is believed to be sufficient for at least the next 85 years, although some studies indicate under investment in the late twentieth century may produce supply problems in the 21st century Fig. 6 [10]. Uranium deposits seem to be log-normally distributed. There is a 300-fold increase in the amount of uranium recoverable for each tenfold decrease in ore grade. In other words, there is little high grade ore and proportionately much lower grade ore available [17].

Uranium Metal and Its Characteristics

Metal

Uranium is a chemical element with the symbol "U" and an atomic number of 92. It is a silvery white metal in the actinide series of the periodic table. Uranium has the second highest atomic weight, lighter only than plutonium. Its density is about 70%, higher than that of lead but slightly lower than that of gold and tungsten. It occurs naturally in low concentrations of a few parts per million in soil, rock and water and is commercially extracted from uranium-bearing minerals such as uraninite.

In nature, uranium is found as uranium-238 (99.2739 to 99.2752%), uranium-235 (0.7198 to 0.7202%) and a very small amount of uranium-234 (0.0050 to 0.0059%). Uranium decays slowly by emitting an alpha particle. The half-life of uranium-238 is about 4.47 billion years and that of uranium-235 is 704 million years, making them useful in dating the age of the earth.

Uranium-235 was the first isotope that was found to be fissile. Other naturally occurring isotopes are fissionable, but not fissile. Upon bombardment with slow neutrons, the uranium-235 isotope will, most of the time, divide into two smaller nuclei, releasing nuclear binding energy and 2 or 3 more neutrons. If too many of these neutrons are absorbed by other uranium-235 nuclei, a nuclear chain reaction occurs that results in a burst of heat or (under special circumstances) an explosion. [18]

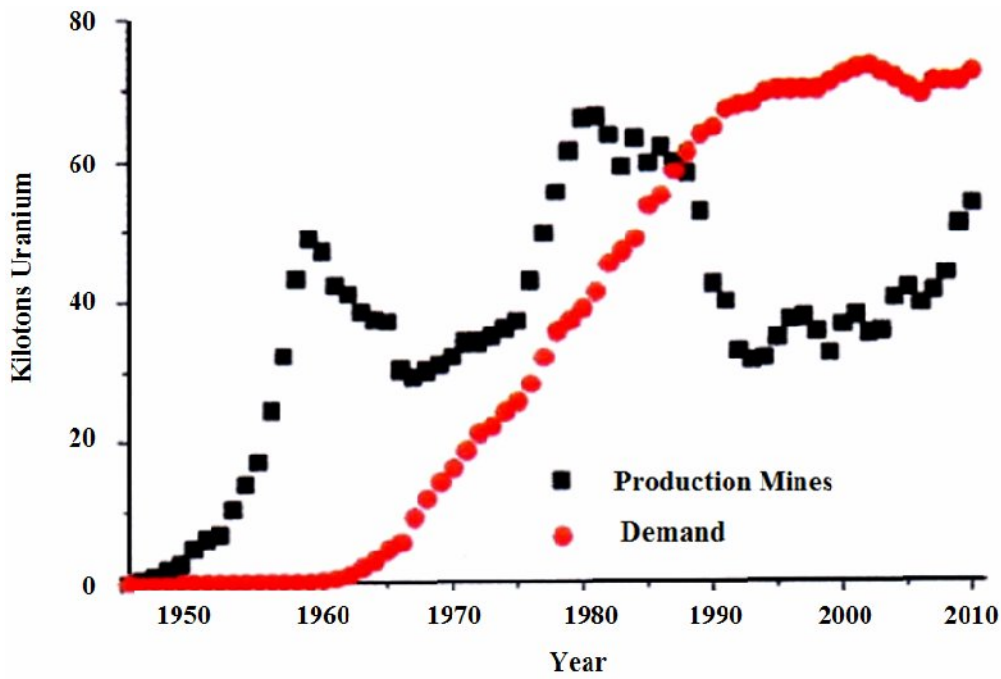


Fig. 6: World uranium production (mines) and demand [10].

Characteristics

When refined, uranium is a silvery white, weakly radioactive metal. It has Mohs hardness of 6, sufficient to scratch glass and approximately equal to that of titanium, rhodium, manganese and niobium. It is malleable, ductile, slightly paramagnetic, strongly electropositive and a poor electrical conductor [3]. Uranium metal has a very high density of 18,800 kg/m^3, denser than lead (11,340 kg/m^3), but slightly less dense than tungsten (19,300 kg/m^3) and gold (19,320 kg/m^3) [4].

Uranium metal reacts with almost all non-metal elements (with the exception of the noble gases) and their compounds, with reactivity increasing with temperature. Hydrochloric and nitric acids dissolve uranium, but non-oxidizing acids other than hydrochloric acid attack the element very slowly. When finely divided, it can react with cold water. In air, uranium metal becomes coated with a dark layer of uranium oxide. Uranium in ores is extracted chemically and converted into uranium dioxide or other chemical forms usable in industry. [3]

Compounds

Oxides and Halides are important compounds which have practical uses in industry. These are briefly summarized below.

Oxides

Calcined uranium yellowcake, as produced in many large mills, contains a distribution of uranium oxidation species in various forms, ranging from most oxidized to least oxidized. Particles with short residence times in a calciner will generally be less oxidized than those with long retention times or particles recovered in the stack scrubber. Uranium content is usually referenced to U_3O_8, which dates to the days of the Manhattan Project when U_3O_8 was used as an analytical chemistry reporting standard.

The most common forms of uranium oxide are triuranium octoxide (U_3O_8) and UO_2 [3]. Both oxide forms are solids that have low solubility in water and are relatively stable over a wide range of environmental conditions. Triuranium octoxide is (depending on conditions) the most stable compound of uranium and is the form most commonly found in nature. Uranium dioxide is the form in which uranium is most commonly used as a nuclear reactor fuel. At ambient temperatures, UO_2 will gradually convert to U_3O_8. Because of their stability, uranium oxides are generally considered the preferred chemical form for storage or disposal [3]. Salts of many oxidation states of uranium are water-soluble and may be studied in aqueous solutions. The most common ionic forms are U_3+ (brown-red), U_4+ (green), UO+2 (unstable) and UO_2+2 (yellow) for U(III), U(IV), U(V) and U(VI) respectively - Fig. 7 [19].

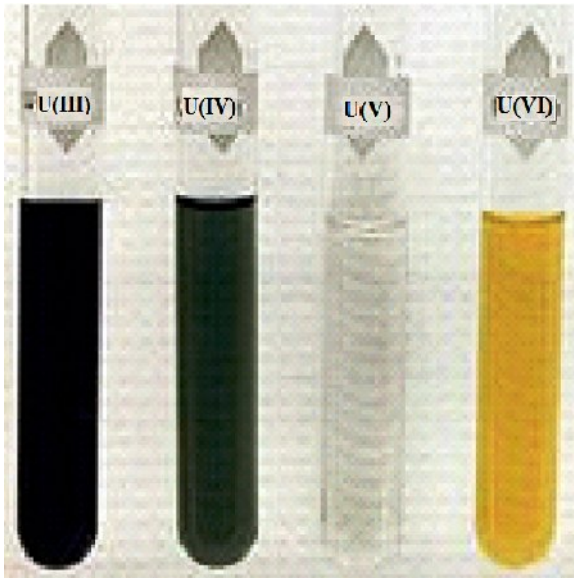


Fig. 7: Uranium in its oxidation states III, IV, V, VI [19].

It does not react with oxygen, nitrogen, carbon dioxide or dry air, but it does react with water or water vapour. For this reason, UF_6 is always handled in leak proof containers and processing equipment. When UF_6 comes into contact with water, such as water vapour in the air, it forms corrosive hydrogen fluoride (HF) and a uranium fluoride compound called uranyl fluoride (UO_2F_6)

Halides

Uranium hexafluoride is the feedstock used to separate uranium-235 from natural uranium. All uranium fluorides are created using uranium tetrafluoride (UF_4). This is itself prepared by hydrofluorination of uranium dioxide. Reduction of UF_4 with hydrogen at 1000°C produces uranium trifluoride (UF_3). Under the right conditions of temperature and pressure, the reaction of solid UF_4 with gaseous uranium hexafluoride (UF_6) can form the intermediate fluorides of U_2F_9, U_4F_{17} and UF_5 [19].

Uranium hexafluoride is a chemical compound consisting of one atom of uranium combined with six atoms of fluorine. This is the chemical form of uranium that is used during the uranium enrichment process. Within a reasonable range of temperature and pressure, it can be a solid, liquid or gas. Solid UF_6 is a white, dense, crystalline material that resembles rock salt [20]. It is used in uranium processing because its unique properties make it very convenient. It can be used as a gas for processing, as a liquid for filling or emptying containers or equipment and as a solid for storage, all at temperatures and pressures commonly used in industrial processes.

At room temperature, UF_6 has a high vapour pressure, making it useful in the gaseous diffusion process to separate the rare uranium-235 from the common uranium-238 isotope. This compound can be prepared from uranium dioxide and uranium hydride by the following process:

$UO_2 + 4\ HF \rightarrow UF_4 + 2H_2O$ (500 ^{o}C, endothermic)

$UF_4 + F_2 \rightarrow UF_6$ (350 ^{o}C, endothermic).

The resulting UF_6, a white solid, is highly reactive (by fluorination), easily sublimes (emitting a vapour that behaves as a nearly ideal gas) and is the most volatile compound of uranium known to exist [21].

One method of preparing uranium tetrachloride (UCl_4) is to directly combine chlorine with either uranium metal or uranium hydride. The reduction of UCl_4 by hydrogen produces uranium trichloride (UCl_3) while the higher chlorides of uranium are prepared by reaction with additional chlorine. All uranium chlorides react with water and air.

Isotopes

Natural Concentrations and Isotopes

Each two or more forms of an element that contain equal numbers of protons but different numbers of neutrons in the nuclei are called isotopes of that element. There are many elements that consist of many isotopes. It is now common knowledge that the nucleus of an atom contains just protons and neutrons and that all the nuclei of a given element contain the same number of protons – the atomic number. For uranium, this is 92. [21]

Natural uranium consists of three major isotopes: uranium-238 (99.28% natural abundance), uranium-235 (0.71%) and uranium-234 (0.0054%) - Fig. 8 [1]. All three are radioactive, emitting alpha particles, with the provision that all three of these isotopes have small probabilities of undergoing spontaneous fission rather than alpha emission. In ordinary uranium, atoms of U-235 occur in the proportion of only one atom in 140, the remaining 139 atoms being U-238.

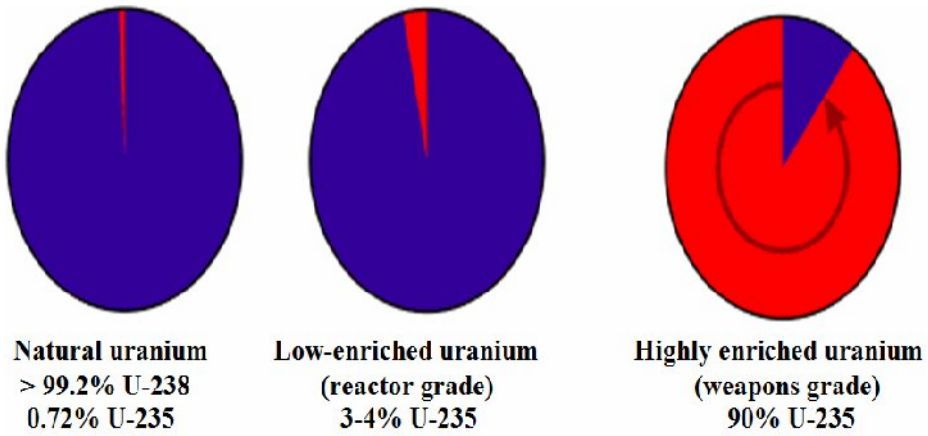


Fig. 8: Proportions of uranium-238 (blue) found naturally and enriched uranium-235 (red) Source: http://commons.wikimedia.org.

Uranium-238 is the most stable isotope of uranium with a half-life of about 4.468 X 10^9 years, roughly the age of the earth. Uranium-235 has a half-life of about 7.13 X 10^6 years and uranium-234 has a half-life of about 2.48 X 10^5 years. For natural uranium, about 49% of its alpha rays (= two protons plus two neutrons, the most energetic form of radiation but moving relatively slowly. It is found in uranium and plutonium-239) are emitted by each of ^{236}U atom and 49% by ^{234}U (since the latter is formed from the former) and about 2.0% by the ^{235}U. When the earth was young, probably about one-fifth of its uranium was uranium-235, but the percentage of ^{234}U was probably much lower than this [3].

Uranium-238 is usually an α emitter (occasionally it undergoes spontaneous fission), decaying through the "Uranium Series" of nuclear decay, which has 18 members, all of which eventually decay into lead-206 by a variety of different decay paths.

The decay series of ^{235}U, which is called the actinium series, has 15 members, all of which eventually decay into lead-207. The constant rates of decay in these decay series make the comparison of the ratios of parent to daughter elements useful in radiometric dating.

Uranium-234 is a member of the "Uranium Series" and it decays to lead-206 through a series of relatively short-lived isotopes [21].

Uranium-233 is made from thorium-232 by neutron bombardment, usually in a nuclear reactor and ^{233}U is also fissile. Its decay series ends with thallium.

Uranium-235 is important for both nuclear reactors and nuclear weapons because it is the only uranium isotope existing on earth in nature, in any significant amount, that is fissile. This means that it can be split into two or three fragments (fission products) by thermal neutrons - Fig. 9 [22].

Uranium-238 is not fissile but is a fertile isotope because, after neutron activation, it can produce plutonium-239, another fissile isotope. Indeed, the ^{238}U nucleus can absorb one neutron to produce the radioactive isotope uranium-239. ^{239}U decays by beta emission to neptunium-239, also a beta-emitter which, in turn, decays into plutonium-239 within a few days and can be used for both civil and military purposes.

Enrichment Technologies

The need to enrich uranium, *i.e.* to increase the concentration of uranium-235, arose from the fact that this isotope was found to be easily fissile and thus could be used for both civilian (enriched to 3% as fuel for power reactors) and military (enriched to 95% as core for nuclear weapons) purposes. Since this characteristic of U-235 was discovered when the Second World War was raging, all efforts were aimed at enriching the material for weapon use.

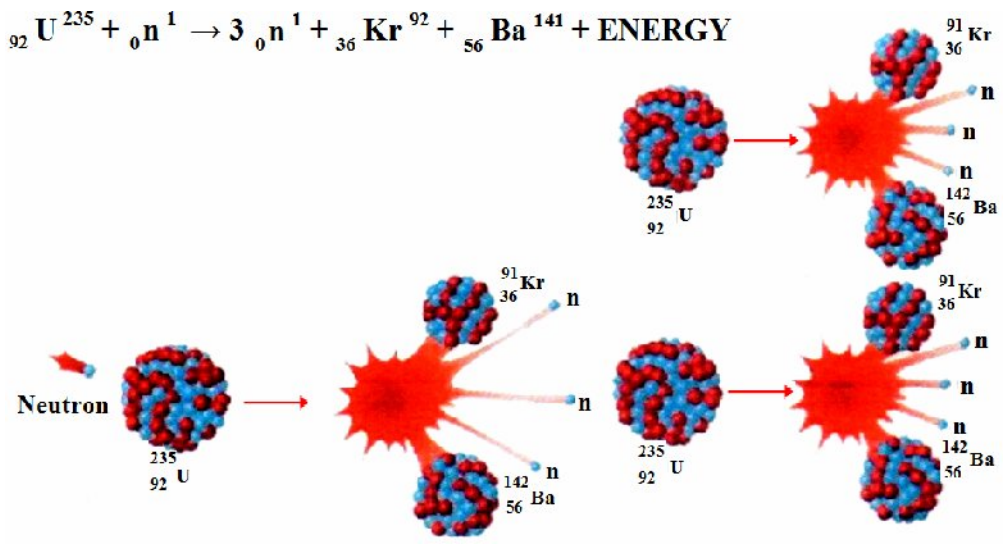


Fig. 9: Fission chain reaction of uranium, which splits into krypton, barium and three separate neutrons. Source: www.thestargarden.co.uk/Nuclear.

In nature, uranium is found as uranium-238 (99.2742%), uranium-235 (0.7204%) and uranium-234 (0.0054). Isotope separation concentrates (enriches) the fissionable uranium-235 for nuclear weapons and most nuclear power plants, except for gas-cooled reactors and pressuirized heavy water reactors Fig.10a and 10b [4]. In order to be considered "enriched", the uranium-235 fraction should be between 3% and 5%. This process, producing uranium that is depleted of uranium-235 and, with a correspondingly increased fraction of uranium-238, is called depleted uranium or DU.

Fig. 10: (a) A billet of highly enriched uranium metal. Source: http://commons.wikimedia.org.

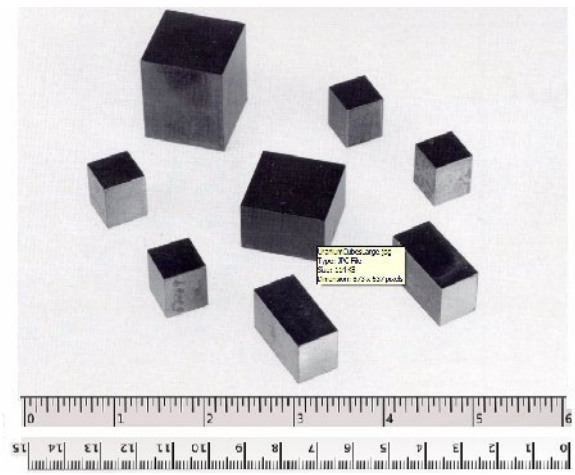


Fig. 10: (b): Cubes and cuboids of uranium produced during the Manhattan Project. Source: http://commons.wikimedia.org

There exist literally dozens of ideas for separating uranium and other isotopes. New ideas are still being proposed every now and then. So far, only two have been found suitable for industrial application. These are diffusion and ultracentrifuge methods. However, for these two processes to work, it was first necessary to choose a uranium compound which could be readily turned into a gas. Uranium hexafluoride UF_6, which is solid at room temperature but is easily vaporized, was the best choice Fig. 11a [4]. The molecule of uranium hexafluoride contains a uranium atom in the centre and the six fluorine ones arranged symmetrically on three perpendicular axes - Fig. 11b [4]. A number of enrichment technologies

tried out over the years, but not used on an industrial scale, are summarized below:

1. Aerodynamic Separation Process.
2. Nozzle Process.
3. Helikon Process.
4. Chemical Exchange Process.
5. French Chemical Process.
6. Japanese Ion Exchange Process.
7. Laser Isotope Separation Process.
8. Atomic Vapour Laser Isotope Separation Process.
9. Molecular Laser Isotope Separation Process.
10. Electro-magnetic Isotope Separation Process.

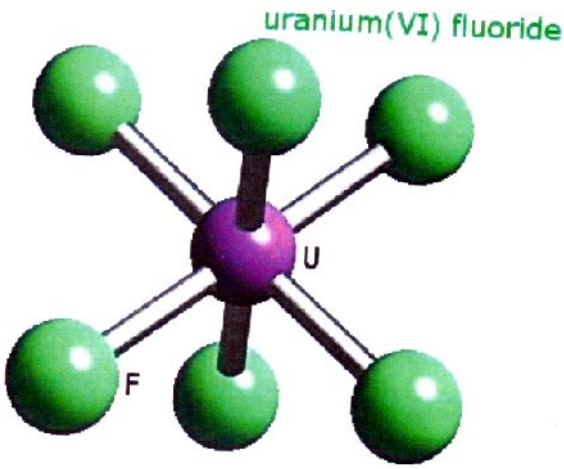


Fig. 11: (a) A uranium hexafluoride (UF6) molecule. Source: www.webelements.com.

Fig. 11: (b) UF6 in a glass ampoule. Source: web.ead.anl.gov/uranium/guide/UF6

Here we will be discussing the two most popular and industrially successfully utilized technologies – the Diffusion and Centrifuge Isotope Separation Processes.

Diffusion Isotope Separation Process

This was the only process used to enrich large quantities of uranium by the five big nuclear powers for almost 30 years. The gaseous diffusion process uses the differences in the rates at which a gas of different molecular weight diffuse through a porous membrane, or barrier, to separate the isotopic forms of elements, e.g. uranium hexafluoride gas [21]. The process was based on the discovery made by F.A. Lindemann and F.W. Aston in 1919 in England that the lighter of two neon isotopes, Ne-20, had a higher average velocity than the heavier isotopes [23]. The average kinetic energy of isotopic molecules in a gas must be the same, *i.e.*

$$\frac{1}{2} m v_1 \times v_1^2 = \frac{1}{2} m_h \times v_h^2$$

where m_h, m_1, v_h and v_1 are the masses and velocities of the molecules containing the heavy and light isotopes, respectively. The maximum theoretical separation factor, α in gaseous diffusion, is given by

$$\alpha = v_1/v_2 = (m_h/m_1)^{1/2}$$

For the isotopic molecules $^{238}UF_6$ and $^{235}UF_6$, the theoretical value of α = 1.0043. Thus the ideal separative power, $\xi_{d,\ is}$

$$\xi_d = \alpha - 1 = 0.0043 - 1 = 4.3 \times 10^{-3}$$

However, the experimental value of ξ_d, in this case, is further reduced to 0.002, which is about 100 times less than the separative power of the gaseous centrifuge method, which will be discussed later.

Gaseous diffusion plants require a very large capital investment due to the very low enrichment factor. This involves extensive repetitions of process equipment and physically large hardware and other facilities Furthermore; it requires a power loading of 6060 megawatts for an annual separative capacity of a plant of 17,230,000 separative work units.

The basic unit of the process is called a stage. A stage consists of a diffuser, or a converter, which contains a permeable membrane (barrier), a compressor, a control valve, a gas cooler and a motor. Since the degree of enrichment is very small in every single stage, a number of stages are connected in series, or cascades, to obtain the desired uranium-235 concentration. About one half of the gas diffuses through the barrier and is fed to the next, higher stage. The waste, or remaining undiffused portion, is recycled to the next lower stage Fig. 12a and Fig. 12b.

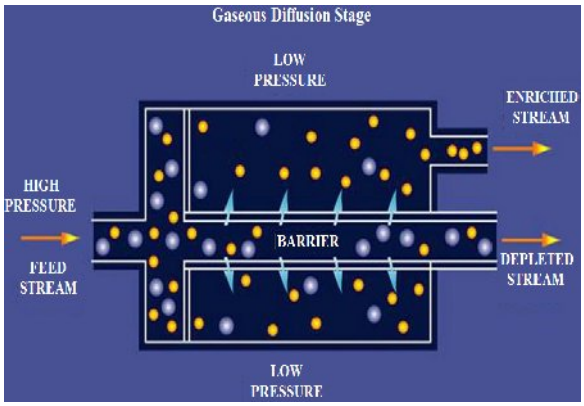


Fig. 12: (a) A stage in gaseous diffusion. Source: www.atomicheritage.org/history/isotope-separation-methods.

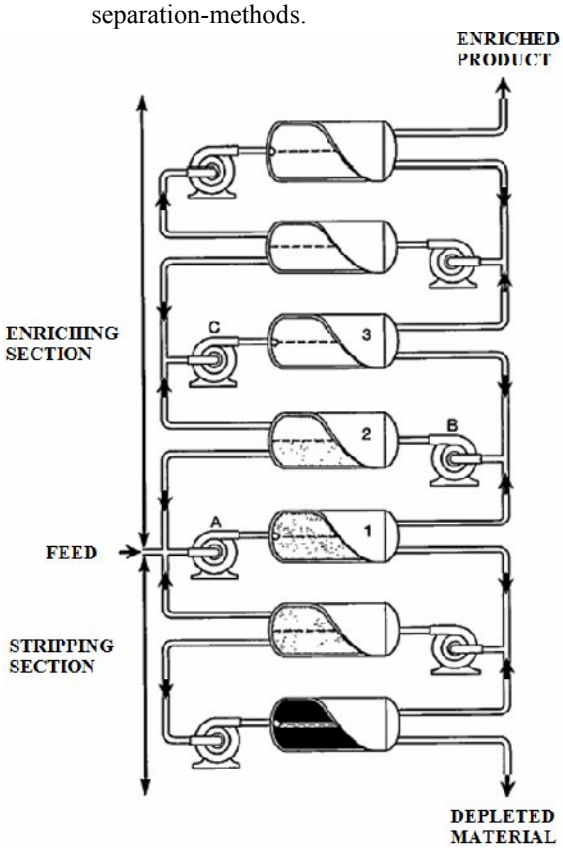


Fig. 12: (b) Schematic diagram of stages of gas flow in gaseous diffusion. Source: www.atomicheritage.org/history/isotope-separation-methods.

In the gaseous diffusion process, gaseous uranium hexafluoride (UF_6) is compressed in axial compressors and fed to the converters, which contain a cooler to remove compressed heat, and a large number of porous tubes. In the space around the porous tubes, a vacuum, maintained by the compressor of the separation stage, causes some of the UF_6 to diffuse through the porous tube wall. In this way the lighter molecules become enriched in the diffusate because of their higher average gas velocity. This occurs due to collision of molecules with the wall when migrating through the pores of the membrane. Collision among the particles does not occur, as otherwise it would cause the heavier and lighter molecules to push each other in the pores, rendering separation impossible. While the gas enriched U-235 is fed to the converter of the next stage after compression, the depleted residue flows to the compressor of the preceding stage. Since the residual fraction is at higher pressure than the fraction that penetrated through the porous wall, the compressors are provided with 2 suction converters maintained at different pressure levels.

Some 1400 diffusion stages are required in a series to produce the 3.5% enriched material used as fuel in light-water reactors [3]. More than 4500 stages are required to produce the 90% enriched material needed for nuclear weapons. Only the USA, USSR, UK, France and China still have old diffusion plants which are now obsolete and these are now mostly being replaced by centrifuge plants. The diffusion process is highly complicated and expensive, requiring huge plants, a lot of electric power and billions of dollars.

Centrifuge Isotope Separation Process

Lindeman and Aston were the first to show the possibility of separating isotopes in 1919 [23]. In 1938, Dr. J.W. Beams of the University of Virginia, USA, first demonstrated an operating gas centrifuge and separated the isotopes of chlorine [23]. Gas centrifuges were used in the USA and in Germany by Profs. Martin, Beyerle and P. Hartek [24-26] respectively during World War II for the separation of uranium isotopes on a laboratory scale. The idea was considered during the early days of the Manhattan Project, but was abandoned because of the difficulty in achieving the high rotational speed (70,000 rpm) necessary for separation. The gas diffusion process was adopted instead.

Though gas diffusion had been the all-dominating process for U-235 enrichment for many years, its small separation factor, together with the large energy consumption, encouraged interest over the next 50 years in developing the gas centrifuge process for industrial scale purposes. At the moment 65% of the world demand for enriched uranium for civilian use is met by the centrifuge method [27, 28].

In a gas centrifuge the light molecules (U-235) are accumulated at the centre and the heavy molecules (U-238) at the periphery of the centrifuge. There is a short shaft attached to the centre of the

maraging steel end cap fitted to the bottom of the maraging steel rotor tube, which acts as a stator. The rotor rests on a motor, which is driven through Eddy currents by a high frequency inverter at 1000 – 1200 Hz. The uranium hexafluoride gas is fed at about the mid-point of the stationary post on the axis. In addition to the feed tube, this post also contains conduits for removing the enriched product and the depleted waste streams from the machine. The rotor is centered by the bearing assembly at the top, where contact between moving and stationary parts is avoided by ring magnetic bearings. The space between the spinning rotor and the casing is evaluated by a diffusion pump. The removal lines for the enriched and the depleted fractions are designed as stationary tubes. The upper system of the feed and collection tubes is used at the same time to generate a convention flow in the axial direction, which multiplies the elementary separation effect - Fig. 13 [27]. Internal gas circulation is, in the largest part, responsible for the separative properties of the centrifuge. The approximate ratio of the radial concentration, X(1), to the axis concentration, X(2), for isotopic molecules under equilibrium conditions and at low pressure (neglecting mixing due to Brownian movement) for n centrifuges (stages) connected in series is:

$X(1)/X(2) \approx \exp(Sn)$

where $S = (MH-ML)\ 10^{-3}v^2.r^2/2RT$ and

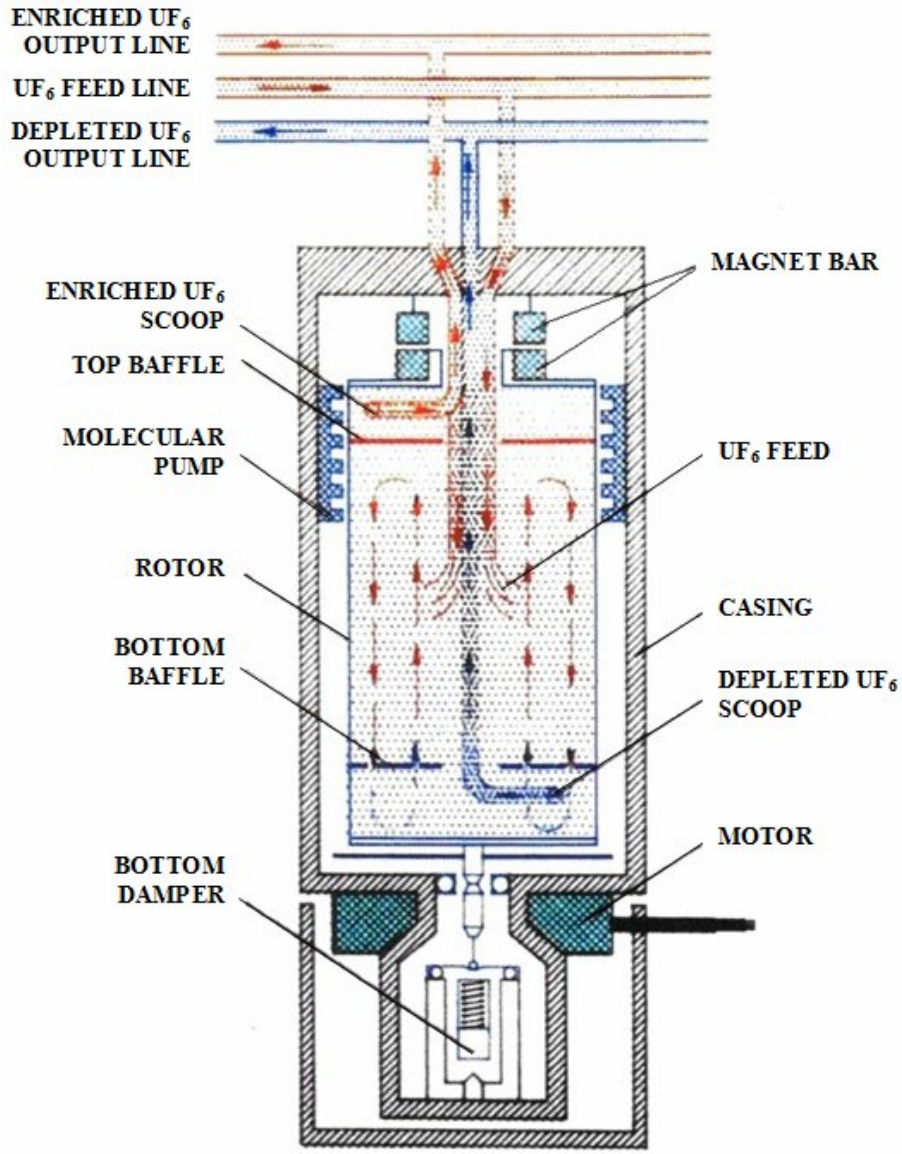


Fig. 13: Schematic diagram of an ultracentrifuge. Source: Kamran, Dr. Mujahid. Dr. A.Q. Khan on Science, Technology and Education. Sang-e-Meel Publications (2003).

MH and ML are the masses of the heavy and light components respectively; r is the centrifuge radius and v the speed of rotation.

Separation factor n = 1 + S

Present centrifuges operating on uranium hexafluoride vary from 3 to 5 meters in length and a 20 cm diameter with rotational speeds of 50,000 to 80,000 rpm. The Americans claim to have 3 meter tall machines with 40 to 50 cm diameters. The enrichment obtained in one stage is limited by the material strength of the centrifuge rotor, which is the product of v and r and amount to 1100 m/sec. Typical separation factors are 1.2 – 1.3 per stage. Thus 12 – 14 stages are required to enrich U-235 from 0.7% to 3% with a 0.2% tail. In comparison, a diffusion plant requires about 1400 stages to achieve this. In the centrifuge method, 50 to 55 stages would be required to obtain the 90% enriched uranium necessary for nuclear weapons. A large number of centrifuges are connected in series forming a stage and these stages are connected together in parallel to form a cascade - Fig. 14. There can be anything from a few hundred to thousands of machines in a cascade. Usually, 9 stages are on the enriching side and 4 stages on the stripping or the depleted side to produce 3% enriched reactor-grade material and about 0.25% to 0.3% depleted uranium [27].

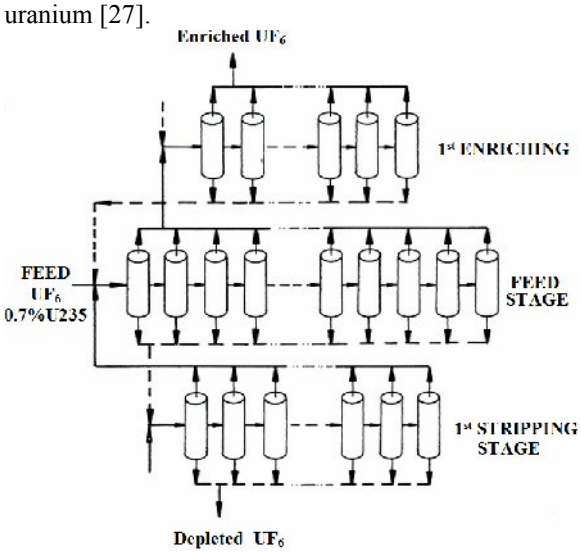


Fig. 14: Schematic diagram of a cascade. Source: Dr. Mujahid Kamran. Dr. A.Q. Khan on Science, Technology and Education. Sang-e-Meel Publications (2003).

The centrifuge process requires less than 7% of the power consumed by a diffusion plant or less than 200 MW for 10Mkg SWU/yr. This makes the environmental impact of gas centrifuge plants minimal compared to gas diffusion plants, which require substantial electric power installation and cooling towers with large water vapour effluent. Smaller centrifuge plants are already economical and their output can be readily multiplied by the installation of parallel processing stages. A very large number of centrifuges are required due to their small size (each having a capacity of less than 15 kg SWU/yr), but this does not lead to excessive construction costs due to mass production [27].

The centrifuge process has been deployed on a commercial level in Russia. Their plants at Seversk, Zelenogorsk, Angarsk and Novouralsk account for about 40% of world capacity. In 2012, Russia commissioned 8th generation centrifuges with a service life of upto 30 years. The last 6th and 7th generation ones were installed in 2005 and 8th generation equipment has been supplied since 2004 to replace 5th generation models with a service life of only 15 years. With surplus capacity, Russian plants operate at low tails assays (underfeeding) to produce low-enriched uranium for sale.

In Europe, Urenco, an industrial group formed by the British, German and Dutch governments, operates enrichment plants in the UK, the Netherlands and Germany and it is building one in the USA Fig. 15.

In Japan, JNC and JNFL operate small centrifuge plants. JNFL's plant at Rokkasho was planned to be 1.5 million SWU/yr.

China has two small centrifuge plants imported from Russia, one at Langzhou of 0.5 million SWU/yr and the other at Hanzhun, operating at 1 million SWU/yr, which has been operational since 2011. It's capacity is expanding considerably, in line with domestic requirements.

- Brazil has a small plant which is being developed to 0.2 million SWU/yr.
- Pakistan has also developed centrifuge enrichment technology at Kahuta (Rawalpindi district).
- Iran has sophisticated centrifuge technology which is operational with an estimated 9000 SWU/yr.
- In both France and the USA, plants with 6th generation Urenco centrifuge technology are now being built to replace ageing diffusion plants, not least because they are more economical to operate. As noted, a centrifuge plant requires as little as 50 kWh/SWU power (Urenco at Capenhurst, UK, input 62.3 kWh/SWU for the whole plant in 2001-02, including infrastructure and capital works.)

Fig. 15: Inside view of the Dutch Centrifuge Plant at Almelo [29].

Areva's new EUR 3 billion French plant – Georges Besse II – started commercial operation in April 2011 and will ramp up to full capacity of 7.5 million SWU/yr in 2016. In October 2013 it had 4.6 million SWU/yr online.

Urenco's new $ 1.5 billion National Enrichment Facility in New Mexico, USA, commenced production in June 2010 with a capacity of supplying 10% of US electricity needs.

Areva is planning to build a $ 2 billion, 3.3 million SWU/yr plant at Idaho Falls, USA named Eagle Rock, which will commence construction in 2015 with first production in 2018. In 2009 it applied for doubling its capacity toi 6.6 million SWU/yr.

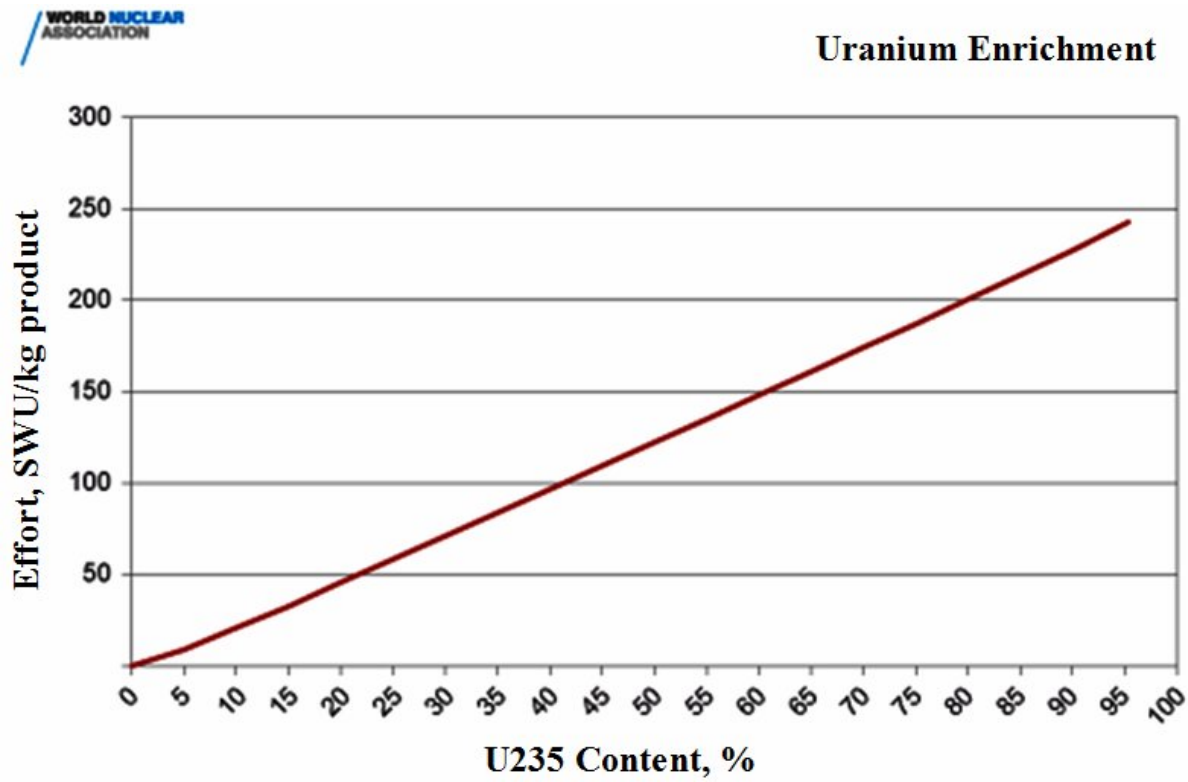
USEC (United States Enrichment Corporation) has been building a plant in Piketon, Ohio on the same Portsmouth site where DOE's (Department of Energy) experimental plant operated in the 1980s as the culmination of a very major R & D program. Operation from 2012 on was envisaged at a cost of $ 3,5 billion. It is designed to have an initial annual capacity of 3.8 million SWU/yr, though its license application is for 7 million SWU/yr to allow for expansion. Authorization for enrichment up to 10% was sought – most enrichment plants operate up to 5% U-235 product, which is becoming a serious constraint as reactor fuel burnup increases. A demonstration cascade started up in September 2007 with about 20 prototype machines and a lead cascade of commercial centrifuges started operation in March 2010. These are reported to be very large machines, 13 meters tall, each with about 350 SWU/yr capacity. However, the whole project was largely halted in July 2009 pending further financing. A total of $ 1.95 billion had been spent from May 2007 to December 2010 and a further $ 2,8 billion cost was then projected. In March 2010, DOE made $ 45 million available to USEC for continued development. Table-1 below lists world enrichment capacities.

Table-1: World Enrichment Capacity – Operational and Planned (Thousand Swu/Yr).

Country	Company and Plant	2013	2015	2020
France	Areva, Georges Besse I & II	5500	7000	8200
Germany Netherlands UK	Urenco: Gronau, Germany Alemelo, Netherlands Capenhurst, UK	14200	14200	15700
Japan	JNFL, Rokkaasho	75	1050	1500
USA	USEC, Paducah & Piketon	0*	0	3800
	Urenco, New Mexico	3500	5700	5700
	Areva, Idaho Falls	0	0	3300
	Global Laser Enrichment	0	0	3000
Russia	Tenex: Angarsk, Novouralsk Zelenogorsk, Seversk	26000	30000	37000
China	CNNC, Hanzhun & Lanzhou	2200	3000	8000
Other	Various	75	500	1000
	TOTAL SWU/yr approx	51550	61450	87200
	Requirements (WNA referencescenario)	49154	51425	59939

*Diffusion, closed mid 2013. US centrifuge proposed

'Other' includes Resende in Brazil, Rattehallib in India and Natanz in Iran. At end of 2012, Iran had about 9000 SWU/yr capacity operating, according to ISIS and other estimates. (ISIS = Centre for Research, Rutherford Appleton Laboratories, University of Oxford, Oxford, UK) Source: WNA Global Nuclear Fuel Market_Report 2013, Areva 2014 Reference Document for most 2013 figures.



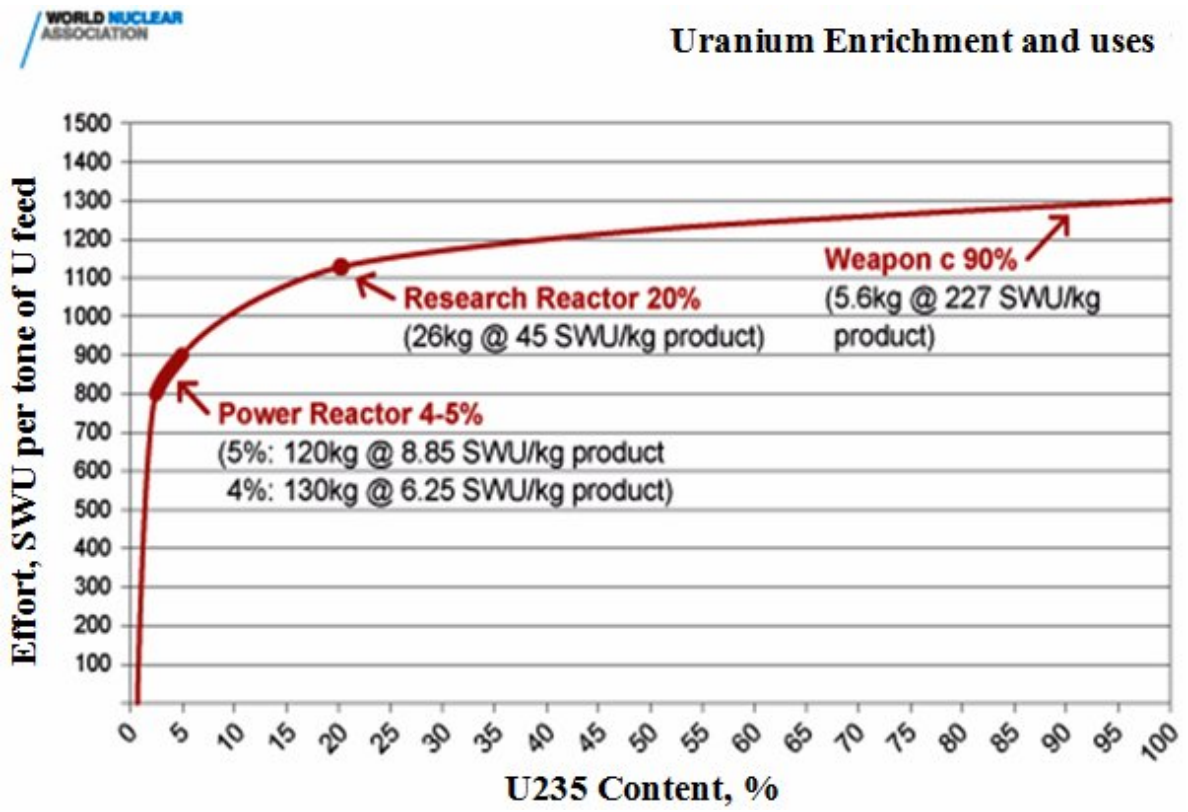


Fig. 16: Uranium Enrichment and Uses [29].

It is interesting to note that, to produce one kilogram of uranium enriched to 5% U-235, it requires 7.9 SWU if the plant is operated at a tails assay of 0.25% or 8.9 SWU if the tails assay is 0.20% (thereby requiring only 9.4 kg instead of 10.4 kg of natural feed). There is always a trade-off between the cost of enrichment SWU and the cost of uranium – Fig. 16 [10].

In the graphs of Fig. 16, the curve flattens out so much because the mass of material being enriched progressively diminishes to these amounts from the original one tonne, so requires relatively less effort to that which has already been applied in order to progress a lot further in percentage enrichment.

The relatively small increment of effort needed to achieve the increase from normal levels is the reason why enrichment plants are considered a sensitive technology in relation to preventing weapons proliferation and are very tightly supervised under international agreements. Where this safeguard supervision is compromised or obstructed, concerns arise. About 140,000 SWU is required to enrich the annual fuel loading for a typical 1000 MWe light water reactor at today's higher enrichment levels. Enrichment costs are substantially related to electrical energy used. The gaseous diffusion process consumes about 2,500 kWh (9,000 MJ) per SWU, while modern gas centrifuge plants require only about 50 kWh (180MJ) per SWU.

Enrichment accounts for almost half of the cost of nuclear fuel and about 5% of the total cost of the electricity generated. In the past is has also accounted for the main greenhouse gas impact from the nuclear fuel cycle where the electricity used for enrichment is generated from coal. However, it still only amounts to 0.1% of the carbon dioxide from equivalent coal-fired electricity generation if modern gas centrifuge plants are used [29].

The gas centrifuge process already has significant cost advantages over the gas diffusion method and this margin will further increase as improved centrifuges are deployed. Thus the discussion on centrifuges can be summarized by stating that this technique has reached maturity and has sidelined gaseous diffusion. As pointed out earlier, 65% of the world's enriched uranium demand is now being met through centrifuge technology. A centrifuge plant has many advantages over a diffusion plant and needs only about 13 – 14 stages to prepare reactor-grade material, as compared to about 1400 stages in a diffusion plant, and about 50 stages for weapons-grade material. Moreover, a centrifuge plant can be built step-wise and expanded as and when needed. These advantages make the centrifuge process so attractive [27]. However, the design and production of centrifuges and the putting up of a functional industrial plant is a difficult task. Pakistan mastered this very sophisticated technology thanks to the engineers and scientists at Kahuta.

With reduced demand for enriched uranium following the Fukushima accident enrichment plants have continued running since it is costly to shut down and re-start centrifuges. The surplus SWU output can be sold or the plants can be underfed so that the enricher ends up with excess uranium for sale, or with enriched product for its own inventory and later sale. The inertia of the enrichment process thus exacerbates over-supply in the uranium market.

Obsolete diffusion plants have been retired, the last being some belated activity at Paducah in 2013. Table 2 shows the present status of the three enrichment technologies and projections for 2017 [29].

Table-2: Enrichment Technologies.

Supply Source	2000	2010	2017 (Projected)
Diffusion	50%	25%	0%
Centrifuge	40%	65%	93%
Laser	0%	0%	3%
HEU Ex Weapons	10%	10%	4%

Report World Uranium Mining: World Nuclear Association

Separative Work Unit

The efficiency of any enrichment method is judged by what is called the "Separative Work Unit" (SWU). This is the amount of separation done by an enrichment process and is a function of the concentrations of the feedstock, the enriched output and the depleted tailings. It is expressed in units which are so calculated as to be proportional to the total input (energy/machine operation time) and the mass processed. Separative work in NOT energy. A kilogram of separative work is known as a SWU. The same amount of separative work will require different amounts of energy depending on the efficiency of the separation technology. Separative work is measured in SWU, KgSW or KgUTA (from the German "Urantrenarbeit" – literally meaning uranium separative work).

- 1 SWU = 1 kg SW = 1 kg UTA
- 1 kSWU = 1 tSW = 1 tUTA
- 1 MSWU = 1 ktSW = 1 ktUTA

There are six variables related by two material balance equations.

On U: $F = P + W$

On U-235: $Fx_f = Px_p + Wx_w$

where F, P and W are the mass of U in the feed, product and tails respectively, and x_f, x_p and x_w are the weight fractions of U-235 in the three streams. In these equations, P and x_p are determined by the in-core fuel management scheme; x_f is given by the U-235 content of natural uranium; and x_w is set to optimize enrichment plant operations (see below). Thus we have two equations in two unknowns. Solving for F, we get that

$$F=P \frac{\chi_p-\chi_w}{(\chi_f-\chi_w)}$$

for $x_p = 0.03$; $x_f = 0.00711$; $x_w = 0.02$, F/P = 5.5.

In many enrichment technologies it is useful to express the magnitude of the separation task in terms of the separative work, S, where S is given by:

$$S = P\,(2x_p - 1)\ln\frac{\chi_p}{1-\chi_p} + W\,(2x_w - 1)\ln\frac{\chi_w}{1-\chi_w}$$

$$- F(2x_f - 1)\ln\frac{\chi_f}{1-\chi_f}$$

(It can be shown that S is proportional to the total volume of material flowing within an ideal cascade producing this amount of separation and this, in turn, is proportional to the inputs of both energy and capital necessary to perform the separation task. Thus the total cost of the separation task is directly proportional to S.)

The total cost of an enrichment task = C_s x S, where C_s = cost per kilogram of separative work [30]

Neutron

The discovery of neutrons was the turning point for the advent of the use of uranium in nuclear technology development. We would, therefore, like to throw some light on the discovery of this important sub-atomic particle. The story of the discovery of the neutron and its properties is central to the extraordinary developments in atomic physics that occurred in the first half of the 20th century, leading to the development of atomic power for civilian and military purposes.

In 1908, Ernest Rutherford and Thomas Royds [31] proved that alpha radiation (2 protons and 2 neutrons tied together) is helium ions. Rutherford's model for the atom of 1911 showed that atoms have their mass and positive charge concentrated in a very small nucleus [32]. The essential nature of the atomic nucleus was established in the discovery of the existence of the neutron in 1932 [33]. By mid century, these discoveries and subsequent developments had ushered in the atomic age.

The 1911 Rutherford model was that the atom was made up of a massive central positive charge of small spatial extent surrounded by a larger cloud of negatively charged electrons. This model had been developed from the extraordinary finding that alpha rays, on occasion, scattered to high angle when passing through gold foil, indicating that alpha particles were occasionally reflecting from a small, but dense, component of atoms. Rutherford and others noted the disparity between the atomic number of an atom, or number of positive charges, and its mass computed in atomic mass units. The atomic number of an atom is usually about half its atomic mass. In 1920 Rutherford suggested that the disparity could be explained by the existence of a neutrally charged particle with the atomic nucleus. [18] Since, at the time, no such particle was known to exist, yet the mass of such a particle had to be about equal to that of the proton. Rutherford considered the required particle to be a neutral double consisting of an electron closely orbiting a proton. [18] The mass of protons is about 1800 times greater than that of electrons [33].

There were other motivations for the proton-electron model. As noted by Rutherford at the time, "We have strong reason for believing that the nuclei of atoms contain electrons as well as positively charged bodies." [33], *i.e.* it was already known that electrons were emitted from the nucleus.

Rutherford called these (neutral double) uncharged particles *neurons*, apparently from the Latin root for *neutral* and the Greek ending *–on* (by imitation of *electron* and *proton*).

In 1931, Walther Bothe and Herbert Becker in Giessen, Germany, found that, if the very energetic alpha particles emitted from polonium fell on certain light elements, specifically beryllium, boron or lithium, an unusually penetrating radiation was produced [34]. Since this radiation was not influenced by an electric field (neutrons have no charge), it was thought to be gamma rays (= electromagnet radiation of very short wave length which cannot be deflected by electric or magnetic fields). The radiation was more penetrating than any gamma rays known and details of the experimental results were difficult to interpret. The following year Irène Joliot-Curie and Frédéric Joliot in Paris showed

that if this unknown radiation fell on paraffin, or any other hydrogen-containing compound, it ejected protons of very high energy [35]. This observation was not in itself inconsistent with the assumed gamma ray nature of the new radiation, but detailed quantitative analysis of the data became increasingly difficult to reconcile with such a hypothesis.

On hearing of the Paris results in 1932, neither Rutherford nor James Chadwick at the University of Cambridge was convinced by the gamma ray interpretation. Chadwick had searched for Rutherford's neutron by several experiments throughout the 1920s without success. Chadwick quickly performed a series of experiments showing that the gamma ray hypothesis was untenable. He repeated the creation of the radiation using beryllium, used better approaches to detection and aimed the radiation at paraffin following the Paris experiment. Paraffin is high in hydrogen content, hence offers a target dense with protons. Since neutrons and protons have almost equal mass, protons scatter energetically from neutrons. Chadwick measured the range of these protons and also measured how the new radiation impacted the atoms of various gases. He found that the new radiation consisted, not of gamma rays, but uncharged particles with about the same mass as the proton; these particles were neutrons [36]. Chadwick won the Nobel Prize in Physics for this discovery in 1935.

In 1935 Chadwick and his doctoral student, Maurice Goldhaber, reported the first accurate measurement of the mass of the neutron, finding it to be slightly greater than the mass of the proton [37, 38]. They therefore predicted that an unbound neutron is unstable and would undergo beta decay. (Beta rays are fast electrons having less energy than alpha particles, emitted by nuclei which have too many neutrons to be stable, *i.e*. lower energy state.

The discovery of the neutron immediately gave scientists a new tool for probing the properties of atomic nuclei. In Rome, Enrico Fermi bombarded heavier elements with neutrons and found them to be radioactive. By 1934 Fermi had used neutron sources to induce radioactivity in 22 different elements, many of these elements of high atomic number. Noticing that other experiments with neutrons at his laboratory seemed to work better on a wooden table than a marble one, Fermi suspected that the protons of the wood were slowing the neutrons and so increasing the chance for the neutron to interact with nuclei. Fermi, therefore, passed neutrons through paraffin wax to slow them down and found that the radioactivity of bombarded elements increased by a hundredfold. The cross section for interaction with nuclei is much larger for slow neutrons than for fast neutrons. In 1938, Fermi received the Nobel Prize in Physics for his "demonstrations of the existence of new radioactive elements produced by neutron irradiation and for his related discovery of nuclear reactions brought about by slow neutrons [39].

Jointly with Lise Meitner and his pupil and assistant, Fritz Strassmann, Otto Hahn furthered the research begun by Fermi and his team when he bombarded uranium with neutrons at his laboratory in Berlin. Between 1934 and 1938, Hahn, Meitner and Strassmann found a great number of radioactive transmutation products from these experiments, all of which they regarded as transuranic. The decisive experiment on 16 – 17 December, 1938 (the celebrated "radium-barium-mesothorium-fractionation") produced puzzling results: the three isotopes consistently behaved not as radium, but as barium. By January 1939, Hahn had concluded [40] that he was seeing light platinoids, barium, lanthanum and cerium. Hahn and his collaborators had observed nuclear fission, or the fractionation of uranium nuclei into light elements induced by neutron bombardment. In their second publication on nuclear fission, Hahn and Strassmann predicted the existence and liberation of additional neutrons during the fission process [41]. Halban, Joliot and Kowarski proved this phenomenon to be a chain reaction in March 1939. [42] In 1945 Hahn received the 1944 Nobel Prize in Chemistry "for his discovery of the fission of heavy atomic nuclei".

The discovery of nuclear fission at the end of 1938 marked a shift in the centers of nuclear research from Europe to the United States. Large numbers of scientists migrated to the United States to escape the troubles in Europe and the looming war. The new centers of nuclear research were universities in the United States, particularly Columbia University in New York, the University of Chicago where Enrico Fermi had relocated, and a new research facility at Los Alamos, New Mexico, beginning in 1942, the new home of the Manhattan Project.

Thus the discovery of the neutron by James Chadwick in 1932, its application to bombard and split uranium by Enrico Fermi in 1934, the subsequent discovery of the release of 2 – 3 neutrons after each fission by Hans von Halban and his colleagues in 1939 [41] and the resultant discovery of the chain reaction by John Dunning and his associates in 1940 [43], led to the rapid development of nuclear weapons.

It is pertinent to realize that, when James Chadwick found the neutron, very few physicists were aware of its importance. The Dutch/Austrian/German physician, Fritz Houtermans, and the British physicist, Robert E. Atkinson were the first to state, shortly after the discovery, that the particle, neutron, might be capable of releasing the powerful forces (energy) dormant in the atom [44]. Even such giants as Niels Bohr, Albert Einstein and Ernest Rutherford dismissed this suggestion. Similarly, a German scientist (chemist), Ida Noddack, when she heard of Fermi's experiments of neutron bombardment of uranium and his subsequent statement that trans-uranic elements heavier than uranium were produced, published a paper [45] suggesting that a nucleus of uranium, after absorbing a neutron, might break up into two fragments. In other words, Fermi had produced nuclear fission. On 10th January, 1939 Lise Meitner and Otto Frisch wrote: "It seems possible that the uranium nucleus has only small stability of form and it may, after neutron capture, divide itself into two nuclei of roughly equal size". They also predicted that a relatively large energy is liberated during fission [46].

But for their ignoring the most important statements made by Fritz Houtermans and Ida Noddak, the Germans would definitely have succeeded in making nuclear weapons before the Americans did and the history of the world would have been totally different.

It should be pointed out that, long before the actual experimental results confirmed it, Niels Bohr had, in 1939, predicted that a neutron could cause fission in U-238 only if its velocity exceeded a certain critical value. Too few of the neutrons available for sustaining the fission process have this critical velocity and a chain reaction is not possible using only U-238. Bohr also predicted that a nucleus of U-235 would undergo fission when any neutron, even one moving very slowly, collided with it and that a chain reaction was possible using U-235. In order to obtain fast neutrons for the fission of U-238, a mixture of U-235 and U-238 is used and the fast neutrons generated in the U-235 cause fission in U-238 [18].

The release of enormous energy in fission is explained in terms of the famous Einstein equation. In 1905, Einstein had advanced the then revolutionary concept that matter and energy are two aspects of a primal cosmic substance rather than two entirely separate phenomena. Moreover, he claimed that matter could be converted into energy and expressed the relationship between them by the equation

$$E = mc^2$$

where *E* represents the energy equivalence of a quantity of matter (or mass) *m* and *c* is the velocity of light. It invariably happens that the total sum of the masses of the fission products and the fission neutrons is less than the mass of the U-236 nucleus. The energy accompanying fission is equal to this mass difference multiplied by the square of the velocity of light. Although the mass difference involved is very small, the velocity of light squared is an enormous number and, therefore, the amount of energy liberated is very large. In fact, the complete fissioning of one gram of U-235 would release about 23,000 kilowatt-hours of heat. Put in another way, one ton of uranium has roughly the same potential fuel value as 3 million tons of coal or 12 million barrels of oil [18].

Production of Plutonium from Uranium

Substances like U-235, which will undergo fission after capturing either a slow or a fast neutron, are called fissile materials and these are of fundamental value for the utilization of nuclear energy. In experiments performed in the Berkeley Radiation Laboratory during 1941 it was found that plutonium-239 was a fissile material, a discovery which immediately aroused great interest in it [18].

Pu-239 is produced when U-238 nuclei absorb slow neutrons. The U-239 nuclei thus formed do not undergo fission but are ultimately transformed into Pu-239. U-239 is an example of a radioactive isotope, or radionuclide, and as such it spontaneously disintegrates or decays [18].

A U-239 nucleus decays by emitting an electron which is, so to speak, shot out of the nucleus like a bullet. But when this process was discovered, it was not at first realized that the particle emitted was an electron, and so it was called a *beta particle*, a name which is still used today. The emission of a beta particle occurs when one of the neutrons in the radioactive nucleus spontaneously changes into a proton. A new element is formed by this fascinating event since the new nucleus contains an additional proton. Thus, when U-239 decays, an isotope with an atomic number of 93 is produced, namely neptunium-239 (Np-239). Np-239 is also radioactive, undergoing beta decay to produce Pu-239 (atomic number 94). [18]

Military and Civilian Uses Of Uranium

Before discussing the military and civilian uses of uranium, it should be pointed out that uranium was supposed to be sold only to those countries that are signatories to the Nuclear Non-Proliferation Treaty (NPT) and which allow international inspection by the International Atomic Energy Agency (IAEA) to verify that it is used exclusively for peaceful purposes. These were the guidelines given by the Nuclear Suppliers Group (NSG), but for political expedience, the US, Australia and Canada have agreed to supply nuclear technology and uranium to India, a non-NPT signatory.

Military Uses

Once scientists had discovered that uranium consisted of 3 isotopes (U-238 of 99.28% natural abundancy, U-235 of 0.71% abundancy and U-234 of 0.0054% abundancy), that U-238 is split by very fast neutrons and U-235 is split by slow and fast neutrons respectively, (U-235 being fissile and U-238 being fertile) and that huge amounts of energy are released during the chain reaction, the first option for its use was to produce nuclear weapons due to the prevailing Second World War. The US was successful in enriching uranium to more than 95% U-235 and also in producing plutonium-239 in Production Reactors (reactors which are used to convert U-238 into Pu-239) [18].

During the later stages of World War II, during the entire extent of the Cold War and, to a lesser extent later on, U-235 has been used as the fissile explosive material to produce nuclear weapons Fig. 17. Initially, two major types of fissile bombs were produced – a relatively simple device using U-235 and a more complicated one using Pu-239 derived from U-238. Later a much more complicated and far more powerful type of fission/fusion bomb (thermonuclear weapon) was produced that used a Pu-based device to cause a mixture of tritium ($^{3}_{1}H$) and deuterium ($^{2}_{1}H$) to undergo nuclear fusion. Such bombs are jacketed in a non-fissile (depleted) uranium casing and they derive more than half of their power from the fission of this material by fast neutrons from the nuclear fusion process [47].

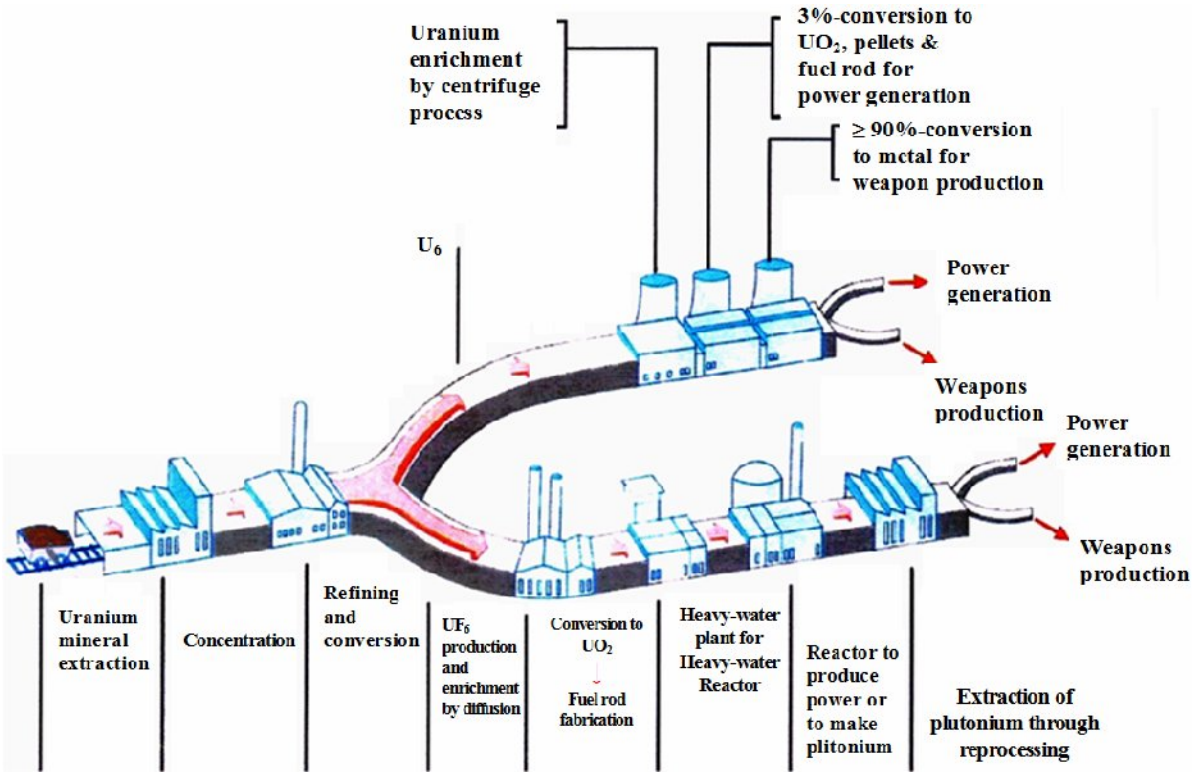


Fig. 17: Two processes for productrion of electric power and nuclear weapons. (1) centrifuge; (2) conventional. The first route is cheaper, more efficient and less investment incentive. Source: Kamran, Dr. Mujahid. Dr. A.Q. Khan on Science, Technology and Education. Sang-e-Meel Publications (2003).

The other major application of uranium in the military sector is in high density penetrators, mainly for anti-tank weapons, the Armour Piercing Disposable Sabo (APDS) - Fig.18. The penetrator, about 30 cm long with a diameter of 4 to 5 cm, not only manages to pierce the armoured plate, but then breaks into pieces, which instantly light up, putting the tank on fire. Tank armour and other removable vehicle armour are also hardened with depleted uranium plates. The use of depleted uranium became politically and environmentally contentious after the use of depleted uranium munitions by the US, UK and other countries during wars in the Persian Gulf and the Balkans raised moral and health questions of uranium compounds left in the soil [48, 49].

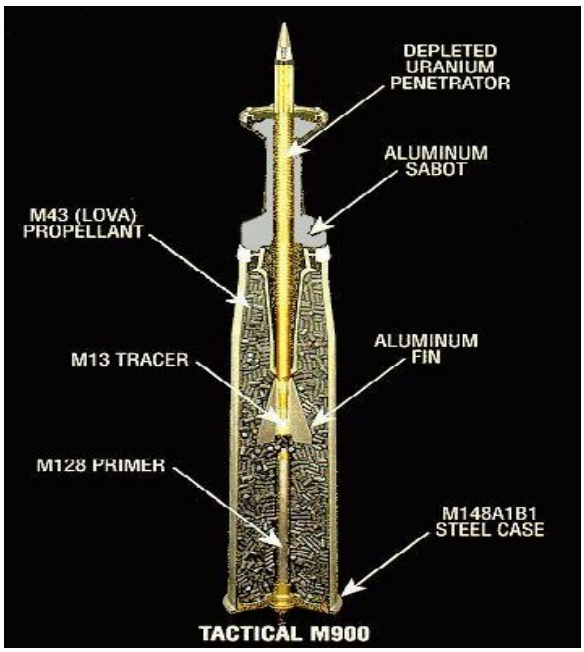


Fig. 18: High density penetrator. Source: http://commons.wikimedia.org.

Depleted uranium is also used as a shielding material in some containers used to store and transport radioactive materials. While the metal itself is radioactive, its high density makes it more effective than lead in halting radiation from strong sources such as radium. Other uses of depleted uranium include counterweights for aircraft control surfaces, as ballast for missile re-entry vehicles as shielding material and counterweights in ships. Due to its high density, this material is found in inertial guidance systems and in gyroscopic compasses. Depleted uranium is preferred over similarly dense metals due to its ability to be easily machined and cast, as well as its relatively low cost. The main risk of exposure to depleted uranium is chemical poisoning by uranium oxide rather than radioactivity (uranium being only a weak alpha emitter).

Civilian Uses

Many people, when talking about nuclear energy, have only nuclear reactors (or perhaps nuclear weapons) in mind. Few people realize the extent to which the use of radioisotopes has changed our lives over the last few decades.

The main use of uranium in the civilian sector is to fuel nuclear plants. One kilogram of uranium-235 can theoretically produce about 20 terajoules of energy (2 X 10^{13} joules), assuming complete fission: as much energy as 1500 tonnes of coal [18, 21, 50].

Commercial nuclear power plants use fuel that is typically enriched to around 3% uranium-235. This 3% enriched UF_6 gas is converted to uranium oxide (UO_2) pellets, which are then placed in zirconium metal tubes known as fuel rods. These rods are assembled in bundles to become fuel elements for the core of the reactors. As per 2014 IAEA reports, about 12% of the world's electricity is generated from uranium in nuclear reactors. This amounts to over 2500 billion kWh each year, as much as from all sources of electricity worldwide in 1960. This electricity comes from some 430 nuclear reactors with a total output capacity of about 380,000 megawatts (MWe) operating in 31 countries. More than 70 additional reactors are under construction and another 170 are planned. Belgium, Bulgaria, Czech Republic, Finland, France, Hungary, Japan, South Korea, Slovakia, Slovenia, Sweden, Switzerland and Ukraine all get 30% or more of their electricity from nuclear reactors. The USA has 100 reactors operating, supplying 20% of its electricity. France gets three quarters of its electricity from uranium reactors. Table 3 below indicates nuclear electricity generation [52].

The CANDU and Magnox designs are the only commercial reactors capable of using unenriched uranium fuel. Fuel used for United States Navy reactors is typically highly enriched in uranium-235 (the exact values are classified). In a breeder reactor, uranium-238 can also be converted into plutonium through the following reaction

$$\frac{238}{92}U + n \rightarrow \frac{239}{92}U + y \xrightarrow{\beta-} \frac{239}{93}Np \xrightarrow{\beta-} \frac{239}{94}\text{Pu}$$

This plutonium can also be used for power generation.

Table-3: Nuclear Share of Electricity Generation in 2014.

Country	Number of Operated Reactors	Total Net Electricity Capacity (MW)	Nuclear Electricity Supplied (GW.h)	Nuclear Share %
Argentina	3	1627	5258.17	4.0
Armenia	1	375	2265.64	30.7
Belgium	7	5927	32093.74	47.5
Brazil	2	1884	14463.39	2.9
Bulgaria	2	1926	15013.99	31.8
Canada	19	13500	98588.12	16.8
China	23	19007	123807.69	2.4
Czech Republic	6	3904	28636.78	35.8
Finland	4	2752	22645.96	34.6
France	58	63130	418001.40	76.9
Germany	9	12074	91783.70	15.8
Hungary	4	1889	14777.73	53.6
India	21	5308	33231.87	3.5
Iran, Republic of	1	915	3723.60	1.5
Japan	48	42388	0.00	0.0
Korea, Republic of	23	20717	149199.22	30.4
Mexico	2	1330	9311.60	5.6
Netherlands	1	482	3873.51	4.0
Pakistan	3	690	4577.53	4.3
Romania	2	1300	10753.68	18.5
Russia	34	24654	169064.84	18.6
Slovakia	4	1814	14420.34	56.8
Slovenia	1	688	6060.82	37.2
South Africa	2	1860	14762.70	6.2
Spain	7	7121	54860.38	20.4
Sweden	10	9470	62270.05	41.5
Switzerland	5	3333	26467.90	37.9
Ukraine	15	13107	83122.79	49.4
United Kingdom	16	9373	57918.48	17.2
United States of America	100	99244	798616.26	19.5
TOTAL	439	376821	2410372.94	NA
	The following information is included in the totals			
Taiwan, China	6	5032	40801.06	18.9

Source: [51] Power Reactor Information System (PRIS) developed and maintained by the IAEA. Last updated 2015-05-28

The discovery of the radioactivity of uranium ushered in scientific and practical uses of the element. The long half-life of the isotope uranium-238 (4.51 X 10^9 years) makes it well-suited for use in estimating the age of the earliest igneous rocks and for other types of radiometric dating, including uranium-thorium dating, uranium-lead dating and uranium-uranium dating. Uranium metal is used for X-ray targets in the making of high-energy X-rays [53].

Using relatively small, special-purpose nuclear reactors using uranium as fuel, it is possible to make a wide range of radioactive materials (radioisotopes) at low cost. For this reason the use of artificially produced radioisotopes has become widespread since the early 1950s and there are now over 200 "research" reactors in 56 countries producing them. These are essentially neutron factories rather than sources of heat. The neutrons thus produced are used to bombard elements to produce isotopes. These isotopes are used in food preservation, in medical applications, in dating of ancient rocks and artifacts, etc. etc.

There are many other uses. For instance, a radioisotope derived from the plutonium (made from uranium) formed in nuclear reactors is used in most household smoke detectors. Radioisotopes are used to detect and analyse pollutants in the environment and to study the movement of surface water in streams and groundwater [53].

There are also other uses for nuclear reactors run on uranium-based fuels. About 200 small nuclear reactors power some 150 ships, mostly submarines, but ranging from icebreakers to aircraft carriers. These can stay at sea for long periods without having to make refueling stops. In the Russian Arctic, where operating conditions are beyond the capability of conventional icebreakers, very powerful nuclear-powered vessels operate year-round where previously only two months allowed northern access each year [53].

The heat produced by nuclear reactors can also be used directly rather than for generating electricity. In Sweden and Russia, for example, surplus heat is used to heat buildings. Nuclear heat may also be used for a variety of industrial processes

such as water desalination. Nuclear desalination is likely to be a major growth area in the next decade. High-temperature heat from nuclear reactors is likely to be employed in some industrial processes in the future, especially for making hydrogen." [53]

Enrichment of Reprocessed Uranium

In some countries used fuel is reprocessed to recover its uranium and plutonium and to reduce the final volume of high-level wastes. The plutonium is normally recycled promptly into mixed-oxide (MOX) fuel by mixing it with depleted uranium.

Where uranium recovered from reprocessing used nuclear fuel (RepU) is to be re-used, it needs to be converted and re-enriched. This is complicated by the presence of impurities and two new isotopes in particular: U-232 and U-236, which are formed by, or following, neutron capture in the reactor and increase with higher burn-up levels. U-232 is largely a decay product of Pu-236 and increases with storage time in used fuel, peaking at about ten years. Both decay much more rapidly than U-235 and U-238 and one of the daughter products of U-232 emits very strong gamma radiation, which means that shielding is necessary in any plant handling material with more than very small traces of it. U-236 is a neutron absorber which impedes the chain reaction and means that a higher level of U-235 enrichment is required in the product to compensate. For the Dutch Borssele reactor, which normally uses 4.4% enriched fuel, compensated enriched reprocessed uranium (c-ERU) is 4.6% enriched to compensate for U-236. Being lighter, both isotopes tend to concentrate in the enriched rather than the depleted output, so reprocessed uranium which is re-enriched for fuel must be segregated from fresh enriched uranium. The presence of U-236 in particular means that most reprocessed uranium can be recycled only once – the main exception being in the UK with AGR fuel made from recycled Magnox uranium being reprocessed. U-234 is also present in RepU, but as an alpha emitter it does not pose extra problems. Traces of some fission products such as Tc-99 may also carry over.

All these considerations mean that only RepU from low-enriched, low-burnup used fuel is normally recycled directly through an enrichment plant. For instance, some 16,000 tonnes of RepU from Magnox reactors (not using enriched uranium and usually known as Magnox depleted uranium) in the UK has been used to make about 1650 tonnes of enriched AGR fuel via two enrichment plants. Much smaller quantities have been used in France and Japan. Some re-enrichment fuel, eg. In Switzerland, Germany and Russia, is actually done by blending RepU and HEU [29].

It can thus be said that uranium is a renewable, sustainable fuel whereas fossil fuel, gas, etc, once used, cannot be recycled or reversed.

International Control- Multilateral Approaches to Control Proliferation.

Since the appearance of fanatic terrorist organizations, the world has become extremely conscious of the threat of nuclear proliferation and very effective and stringent measures to control nuclear materials and technology have been taken.

Following proposals from the International Atomic Energy Agency (IAEA) and Russia, and in connection with the US-led Global Nuclear Energy Partnership (GNEP), there are moves to establish international uranium enrichment centres to control enriched uranium. These are one kind of multilateral nuclear approaches (MNA) called for by IAEA. Part of the motivation for international centres is to bring all new enrichment capacity, and perhaps eventually all enrichment, under international control as a non-proliferation measure. Precisely what "control" means remains to be defined and will not be uniform in all situations. But having ownership and operation shared among a number of countries at least means that there is a level of international scrutiny which is unlikely in a strictly government-owned and – operated national facility.

The first of these international centres is the International Uranium Enrichment Centre (IUEC) at Angarsk in Siberia, with Kazakh, Armenian and Ukrainian equity so far. The centre is to provide assured supplies of low-enriched uranium for power reactors to new nuclear power states and those with small nuclear programs, giving them equity in the project, but without allowing them access to the enrichment technology. Russia will maintain majority ownership and in February 2007, the IUEC was entered into the list of Russian nuclear facilities eligible for implementation of IAEA safeguards. The USA has expressed support for the IUEC at Angarsk. IUEC will sell both enrichment services (SWU) and enriched uranium product.

In some respects this is very similar to what pertains now with the Eurodif set-up where a single large enrichment plant in France with five owners (France – 60%, Italy, Spain, Belgium and Iran) is

operated under IAEA safeguards by the host country without giving participants any access to the technology – simply some entitlement to share of the product and even that is constrained in the case of Iran. The French Atomic Energy Commission proposed that the new Georges Bess II Plant, which replaces Eurodif, should be open to international partnerships on a similar basis and minor shares in the Areva subsidiary operating company, Societ d'Enrichissement du Tricastin (SET), have so far been sold to GDF Suez, a Japanese partnership and Korea Hydro and Nuclear Power (KHNP) – total 10%.

The three-nation Urenco set-up is also similar, though with more plants in different countries – UK, Netherlands and Germany – and here the technology is not available to host countries or accessible to other equity holders. Like Russia with IUEC, Urenco (owned by the UK and Netherlands host governments plus E.On and RWE in Germany) has made it plain that, if its technology is used in international centres, it will not be accessible. Its new plant is in the USA where the host government has regulatory but not management control.

A planned new Areva plant in the USA has no ownership diversity beyond that of Areva itself, so will be essentially a French plant on US territory. The only other major enrichment plant in the Western world is USEC's very old one in the USA [29].

Health Hazards

Human Exposure

A person can be exposed to uranium (or its radioactive daughters, such as radon) by inhaling dust in air or by ingesting contaminated water and food. The amount of uranium in the air is usually very small; however, people who work in factories that process phosphate fertilizers, live near government facilities that made or tested nuclear weapons, live or work near a modern battlefield where depleted uranium weapons have been used, or live or work near coal-fired power plants, facilities that mine or process uranium ore or enrich uranium for reactor fuel, may have increased exposure to uranium. Houses or structures that are over uranium deposits (either natural or man-made slag deposits) may have an increased incidence of exposure to radon gas.

Most ingested uranium is excreted during digestion. Only 0.5% is absorbed when insoluble forms of uranium, such as its oxide, are ingested, whereas absorption of the more soluble uranyl ion can be up to 5%. However, soluble uranium compounds tend to quickly pass through the body whereas insoluble uranium compounds, especially when inhaled by way of dust into the lungs, pose a more serious exposure hazard. After entering the bloodstream, the absorbed uranium tends to bio-accumulate and stay for many years in bone tissue because of uranium's affinity for phosphates. Uranium is not absorbed through the skin and the alpha particles released by uranium cannot penetrate the skin.

Incorporated uranium become uranyl ions, which accumulate in bone, liver, kidneys and reproductive tissues. Uranium can be decontaminated from steel surfaces and aquifers [48, 50, 53].

Effects and Precautions

Normal functioning of the kidneys, brain, liver, heart and other systems can be affected by uranium exposure because, besides being weakly radioactive, uranium is a toxic metal. Uranium is also a reproductive toxicant. Radiological effects are generally local because alpha radiation, the primary form of U-238 decay, has a very short range and will not penetrate skin. Uranyl (UO_2 + 2) ions, such as from uranium trioxide or uranyl nitrate and other hexavalent uranium compounds, have been shown to cause birth defects and immune system damage in laboratory animals. While the Center for Disease Control and Prevention (CDC) has published one study that no human cancer has been seen as a result of exposure to natural or depleted uranium, exposure to uranium and its decay products, especially radon, are widely know significant health threats. Exposure to strontium-90, iodine-131 and other fission products is unrelated to uranium exposure, but may result from medical procedures or exposure to spent reactor fuel or fallout from nuclear weapons. Although accidental inhalation exposure to a high concentration of uranium hexafluoride has resulted in human fatalities, those deaths were associated with the generation of highly toxic hydrofluoric acid and uranyl fluoride rather than the uranium itself. Finely divided uranium metal presents a fire hazard because uranium is pyrophoric; small grains will ignite spontaneously in air at room temperature.

Uranium metal is commonly handled with gloves as a sufficient precaution. Uranium concentrate is handled and contained so as to ensure that people do not inhale or ingest it [48, 50, 53].

Conclusion

During the last few years many countries have, to a certain extent, switched to nuclear power.

France, Belgium, Sweden, USA, UK, Armenia, Bulgaria, Czech Republic, Finland, Hungary, Switzerland, etc. now derive a substantial percentage of their power requirements from nuclear power plants. Pakistan is, at the moment, deriving only about 4.3% of its requirements from nuclear power, but some nuclear power plants have been planned, which will substantially increase this percentage. In the light of this development, in which a central role is played by uranium, this article has reviewed the element uranium, its occurrence, metallurgical and chemical properties, enrichment for reactor grade (or weapon grade) material, uses for power generation and other civil and military purposes and subsequent health hazards. Except for its unfortunate use by the USA to drop nuclear bombs on Japan in 1945, the element uranium and its associated technology has made invaluable contributions to maintaining world peace, albeit through the mutually assured destruction (MAD). There have been local wars and skirmishes, but luckily no world wars [54].

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