

THE NUCLEAR REACTOR CLOSED CYCLE

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Summary

This chapter examines the stages of the closed nuclear reactor cycle from mining of the ore through to spent fuel in preparation for reprocessing and disposal of fission wastes.

There is a discussion of fission, activation and transuranium nuclides and their half-lives and abundances. The various options of dealing with spent fuel - reprocessing or not -

are examined from both a technological viewpoint as well as from the political actions in the U.S.A. which have so far denied the U.S. nuclear industry the Fast Breeder Reactor and the reprocessing option. Some aspects and outcomes of this political decision are examined.

The impact of reprocessing spent fuel or not, on radioactive waste volumes, and the subsequent management and security timeframe, is examined.

The current retirement and destruction of military nuclear warheads from the U.S.A. and former U.S.S.R. arsenals is briefly touched upon in the use of MOX fuel in existing nuclear reactors.

The avoidance of proliferation risks from increasing quantities of non-reprocessed spent fuel in the future because of contained transuranium nuclides, and the assurance of relatively non-polluting energy sources for the future, require that the Fast Breeder Reactor program and reprocessing, be re-implemented in the near future.

In addition, the breeder cycle opens up the immense energy potential contained in surface stockpiles of depleted uranium, and allows it to be used rather than wasted.

Breeding also expands the commercially valuable natural uranium resource by allowing lower grade deposits to be exploited including uranium in seawater, and also opens up the vast resource of energy contained in the much more abundant thorium-232.

1. The Closed Nuclear Cycle

1.1. Introduction

The nuclear reactor cycle, from uranium mining to final waste disposal, comprises several stages. These stages depend upon the reactor design and type, and whether or not spent fuel is re-processed - (the 'closed nuclear fuel cycle'); stored ('once-through'); or the reactor operates with some combination or modification of these cycles.

The closed nuclear fuel cycle is becoming increasingly used in some European countries and Japan.

The use of breeder reactors based upon uranium or thorium, requires a closed nuclear fuel cycle in order to return the bred transuranium nuclides, uranium-233 and unused uranium and thorium back into the cycle.

Some of the stages in the fuel cycle are associated with the production of various classes and volumes of radioactive wastes throughout the world as shown in Table 1.

Reactor Cycle Stage	Radioactive Wastes	Non-wastes for recycling
Front End		
Uranium Mining	1 000 000 000 + tonnes	
Processing	Minor	
Refining	Minor	
Conversion	About 35 000 m ³	

Enrichment	About 16 000 m ³	
Depleted Uranium	'Waste' – only if not recycled	1 500 000 Tonnes
Fuel Fabrication	About 160 000 m ³	
Back End		
Spent Fuel	'Waste' – only if not reprocessed	230 000 Tonnes
Maintenance LILW Wastes	About 6 000 000 m ³	
Spent Fuel Reprocessing		218 000 + Tonnes
Fission Wastes (4 percent)	About 9000 + Tonnes	
Military		
Retired Weapons HEU U-235		500 Tonnes ± (U.S. & Russia)
Retired Weapons plutonium-239		500 Tonnes ± (U.S. & Russia)

Table 1: Summary of Accumulated Radioactive Wastes and Non-wastes in the World to about 2000

The closed-cycle stages from mining to final disposal are shown in Figure 1. The first 8 stages are described in this article. The remaining stages are described in more detail in *Disposal of Nuclear Wastes and Reactor Decommissioning* dealing with nuclear waste disposal and the destruction of military 'wastes' - including retired weapons - in the reactor cycle.

The processes leading up to loading fuel into the reactor are known as the 'front end' of the cycle, and those following discharge of spent fuel from the reactor are known as the 'back end' of the cycle.

The simpler, but more resource-intensive, 'once-through' cycle foregoes - for the moment - the re-processing option and the associated relatively low volumes of wastes. Without reprocessing, the entire discharged spent fuel, containing about 97 percent unused uranium and transuranium elements, may be required to be managed as long-term waste.

With reprocessing, only the low volume, relatively short half-life, fission nuclides would be managed as waste. The 'once-through' cycle requires that world uranium mining production be maintained at a relatively high level to keep up with the demand for new fuel.

Where natural uranium is used in the reactor (e.g., the CANDU), uranium enrichment is not required and reprocessing is not considered at this time, as replacement fuel is cheap at about U.S.\$22 to \$30 kg⁻¹, relative to the costs of enriched fuel, and relative to the costs of reprocessing.

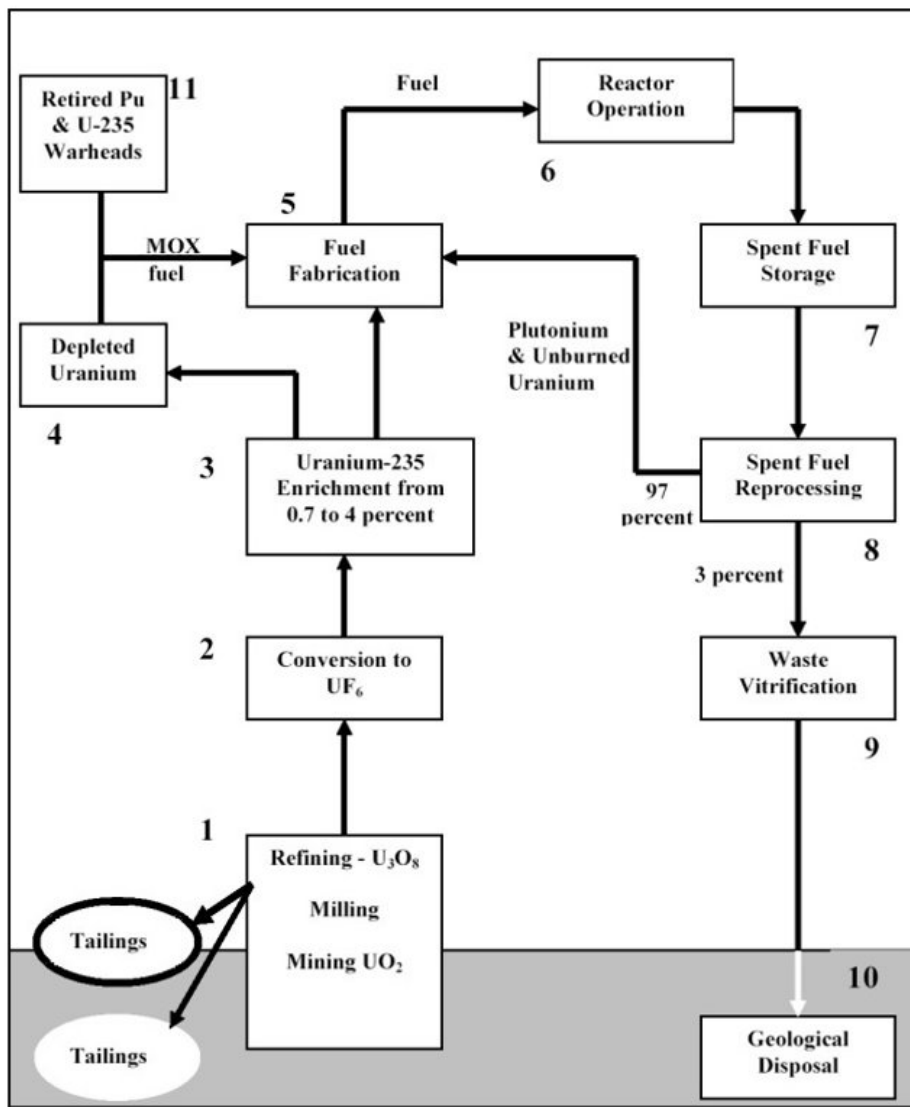


Figure 1: The Closed Nuclear Fuel Cycle for a Typical PWR or BWR Reactor

2. Uranium Mining, Processing, Refining

The first mining efforts to deliberately recover uranium occurred in central Europe to extract uranium for use in coloring glass and glazes. When radium - one of the radioactive progeny of uranium - was discovered in 1897 and immediately became of value in medical radiation treatments, a mining boom of known uranium-bearing deposits took place, with the uranium itself being treated mostly as a by-product or waste.

Mining methods of economically viable deposits may be by open pit (about 38 percent at the end of 2000), underground mining (about 33 percent), in situ leaching (ISL) (about 17 percent), or as a by-product of other mining or industrial process (about 12 percent).

By-product uranium is recovered from activities such as phosphate mining and

processing for fertilizer production; formerly, from the processing of some alum shale deposits in Sweden; formerly, from some low-grade coal deposits in the U.S.A.; and from some gold and copper mines. Increasingly, more uranium deposits at the present time are exploited by *in situ* leaching of the deep ore body to extract uranium, which is then pumped in solution to the surface for extraction.

This method produces neither rock waste nor tailings. Where the ore is mined, rather than chemically leached, it is crushed at the mine site, reduced to sand-sized particles, leached with a solvent solution, and then is further processed to extract and purify the uranium.

The residual wastes from mining the common low-grade deposits (ranging mostly from about 0.1 percent to 1 percent uranium) amount to large quantities of rock and process tailings containing traces of residual uranium too difficult to extract, and radium (of no significant value today) along with most of its progeny.

Such wastes today amount to more than about 200 million tonnes in surface waste piles in the U.S.A. alone, and possibly ten times more at existing and former uranium mining operations throughout the world. Most are now gradually being addressed to ensure that they are adequately covered and protected to minimize radon gas leakage from them; to limit moisture penetration and acidic drainage; and to protect them from weather erosion.

Extraction of uranium by *in situ* leaching avoids most of these problems. Modern mining is much more stringently regulated and controlled than previously, with ongoing environmental protection and remediation requirements and activities.

	Tonnes	Percent
Australia	890 000	26
Kazakhstan	560 000	17
Canada	510 000	15
South Africa	350 000	10
Namibia	260 000	8
Brazil	230 000	7
Russia	150 000	4
United States	125 000	4
Uzbekistan	120 000	4
Niger	70 000	2
Ukraine	45 000	<1
Others (28 countries)	>50 000	1
Total*	3 360 000	
* At 41 000 tonnes a ⁻¹ production, this estimated resource will last for less than 100 years at this price, without reprocessing, and without the adoption of the fast breeder cycle. Data are from various sources.		

Table 2: Estimated Recoverable World Uranium Resource at US\$80 kg⁻¹ of Uranium

The richest uranium deposits at the present time are found in Canada and Australia which dominate world production (see Table 2). The richest ores are the primary oxides of uranium (uraninite and pitchblende) - usually in mineralized veins with other metals, such as silver, copper, bismuth, cobalt, molybdenum and lead as sulfides, selenides, tellurides and arsenides along with various oxides and silicates. Many of the richest known deposits have generally been mined out, with a few notable exceptions in Canada and Australia. There are also many secondary and very complex uranium-vanadium minerals - often brightly colored green and yellow - which tend to be more widely dispersed through the sedimentary strata in which they are found, as in Colorado, and in many other low grade uranium deposits throughout the world.

The extraction of uranium from seawater (even at about 3 parts per billion, or 3 milligrams per tonne of seawater) is a possible future massive source of uranium. In Japan, the Takasaki Radiation Chemistry Research Establishment conducted extraction experiments several years ago and determined that uranium and other metals could be readily extracted from seawater, but not yet economically at the prevailing price for uranium.

The Uranium contained in the Black Current flowing off the coast of Japan carries about 5 million tonnes of uranium (comparable to the estimated remaining uranium resource on land) each year along the coastline. Japan uses about 6000 tonnes of uranium annually, of which only about 3 percent is actually consumed in the reactors during each fuel cycle, so extraction of even a small fraction of that carried in this current, could meet their needs as well as those of the rest of the world. With reprocessing and the use of a fast breeder cycle the resource is essentially unlimited.

2.1. Ore processing, Concentration and Refining

This converts the extracted and purified uranium to U_3O_8 , also known as yellowcake. This is a pure, but very low-level radioactive material. It is traded internationally and safely shipped around the world in 100 L steel drums to uranium enrichment facilities, or may be fabricated into natural uranium fuel for use in those reactors (CANDU and GCR) fueled by natural uranium.

The world production of uranium in the year 2000, controlled by 8 major mining companies operating in about 16 countries, was about 41 000 tonnes of U_3O_8 . With an average grade of about 1 percent UO_2 in the feed ore, this implies that more than 4 million tonnes of radioactive mine wastes are produced annually from these deposits.

Comparable radioactive wastes are produced from many other base metal mining operations, to the extent of about 1 billion tonnes each year. Most of these other mine wastes are controlled primarily to minimize acid mine drainage effects and erosion, and attract little attention because of their contained radioactivity, which is usually unmeasured, uncontrolled, and ignored.

The largest producers of uranium for sale on the international market are Canada and Australia (annually about 11 000 and 8000 tonnes respectively in 2000) producing more than 50 percent of the world supply. Kazakhstan appears to be making a major effort -

announced in 2002 - to eventually become the dominant producer in the world.

3. Conversion to UF₆

Conversion is the process of changing U₃O₈ (yellow-cake), to uranium hexafluoride UF₆ for enrichment in the uranium-235 isotope.

There are five commercial conversion plants in the world: in the U.S.A., Canada, France, the United Kingdom, and Russia. Two other countries, Brazil and China also operate relatively small conversion facilities but not, at present, commercially. Total available capacity in the seven facilities to the end of 2000, is about 66 000 tonnes/a, but annual world requirements for conversion are approximately 57 000 tonnes.

There are only minor low-level uranium wastes associated with such conversion. The cumulative total of such wastes throughout the world up to the year 2000, amounts to about 35 000 m³.

4. Enrichment

Natural uranium contains 99.3 percent U-238 and 0.7 percent U-235. Nuclear fission reactors based upon uranium, cannot operate without the uranium-235 isotope, and in the case of light water moderated reactors, require the concentration to be greater than about 3 percent.

Enrichment is the process of augmenting the percentage of uranium-235 in uranium hexa-fluoride (UF₆) and rejecting a stream of uranium-238 (also known as depleted uranium), before the U-235-enriched uranium is processed into the oxide fuel for use in the reactor. Some uraniumiferous wastes are produced during this process, with world cumulative totals up to the year 2000 amounting to about 16 000 m³.

The two uranium isotopes cannot be separated chemically but have slightly different masses (about 1.3 percent difference), so are physically separable though with considerable difficulty.

There are two common multi-stage enrichment processes - gaseous diffusion, used for the Manhattan Project in the 1940s, and gaseous ultra-centrifuging in Calutrons - with others (laser ionization coupled with magnetic separation) being researched. The process, taking into account the market price of uranium and the high electrical energy cost of enrichment (described in Separative Work Units – SWUs – the amount of electrical energy needed to produce 1 kilogram of enriched uranium), still leaves about 0.25 - 0.3 percent U-235 in the rejected uranium-238.

The total U.S. nuclear electrical capacity of about 100 GW(e) from more than 100 large reactors requires some 12 million SWUs per year to enrich the fuel. Each SWU - using the gas diffusion process - requires about 2500 kWh of electricity or the equivalent of about 4.2 percent of the total nuclear electrical output (late 1990s). Competing offshore interests, seeking to break into the lucrative U.S. enrichment market, suggest that they can achieve this separation at a much lower cost. Future advances in isotope separation in the U.S.A., and lower separation costs, may make it economically advantageous to

re-process some of the stockpiled depleted uranium to strip out more of the residual U-235 if the adoption of a breeder reactor cycle and spent fuel reprocessing continues to be politically rejected or remains economically unattractive at the present price of uranium.

To produce about 4 percent uranium-235 enrichment from 0.7 percent feed material requires an almost 8 fold concentration. For every tonne of Low Enriched U-235 produced for the Light Water Reactor (4 percent U-235), about 7 tonnes of depleted uranium (about 99.7 percent U-238) is rejected from the process. For every tonne of High Enriched Uranium (say 20 percent U-235 - the minimum enrichment used in nuclear submarine and ship reactors) about 39 tonnes of depleted uranium is rejected.

In general, the more enriched the uranium, the greater the reactivity margin to over-ride the effects of fission poison build-up, the smaller the required fuel load to maintain a large power output, and the more compact the reactor, as in nuclear submarines and ships.

Uranium enrichment - an expensive and technologically demanding process - was initially a virtual monopoly of the U.S.A. The early reactor programs of most other countries were based upon the U.S. PWR or BWR reactor designs and U.S. enriched fuel. Other countries either accepted this as the price to be paid for nuclear co-operation and development, or began to develop their own independent enrichment programs, or sought to build reactors that were fueled by natural uranium (as in the U.K. and Canada).

Commercial 'enrichment' (as opposed to 'conversion') is carried out in the U.S.A., France, the United Kingdom, Germany, the Netherlands and Russia. These countries effectively control the enriched uranium fuel supply to many other countries which operate Light Water reactors. All of these countries must be signatories of the Nuclear Non-Proliferation Treaty (NNPT) and allow International inspection of nuclear facilities, materials and operations to ensure that there is no clandestine diversion of restricted materials. Other countries with enrichment facilities for their own programs include China, Japan, and Pakistan. There are still a few states like India, Pakistan, Israel and North Korea, that possess nuclear facilities and nuclear weapons, but resist signing the NNPT or threaten to withdraw from it (N. Korea).

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Biographical Sketch

Dr. John K. Sutherland obtained a First Class Honors degree and PhD in Geology at Manchester University in England. He accepted a position in a research laboratory in Canada in which he was responsible for chemical and instrumental analyses of rocks, ores and minerals, including the development and use of X-ray diffraction, X-ray Fluorescence and Electron Microprobe analyses. In 1980, he joined the Health Physics Department of the local utility. He was responsible for the Environmental Radiation Monitoring Program, and for external beta-gamma dosimetry for the 600 nuclear facility employees over almost 20 years. He developed analytical techniques for the analysis of strontium-90, and for rapid analysis of vanadium in fly ash, and wrote the complete chemical analytical procedure for analyzing coal and fluidized bed combustion products in a coal burning facility. He was responsible for revising the Derived Emission Limits for the nuclear facility and for contributing to Shift Supervisor training and Radiation Protection training for plant employees, and contributed to Emergency Response Co-ordination and training. He conducted a radiation monitoring program of the nuclear waste and storage facilities over many years and was also engaged in monitoring spent fuel transfers to the storage facilities. During plant maintenance and other outages, he participated in Radiation Protection activities during maintenance work, including Fuel Channel replacement. He became an adjunct professor at the University of New Brunswick, where he taught Nuclear Safety and Reliability to graduate engineers. Since retiring, he has engaged in consulting work, including developing nuclear and radiation training for plant employees, emergency responders, and at other facilities where radioactive materials are used. He teaches and writes extensively on all radiation related issues.