

ENVIRONMENTAL EFFECTS OF NUCLEAR POWER PRODUCTION

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Summary

The introduction encompasses a historical description of the pre-World War II discovery of the fission phenomenon and its military implications, as well as the post-WWII developments that lead to the peaceful uses of atomic energy. All main phases of

the nuclear power production encompassing the front and back ends of the nuclear fuel cycle, as well as the power generation in nuclear reactor plants are covered succinctly, but with in-depth short discussions of each phase. A broad overview of the environmental effects of each phase of the nuclear fuel cycle goes from the high ^{230}Th , ^{226}Ra and ^{210}Po concentrations in local acid uranium tailings, all the way to the global circulation of ^{129}I as a good indication of how much nuclear fuel has been processed thus far in the world. The effects on the environment due to nuclear reactors are discussed under routine and accidental operating conditions. There are different national approaches for high-level waste management. On the one hand, it is observed that policies and costs to deal with high-level nuclear wastes vary considerably whether reprocessing, vitrification and pre-disposal storage are adopted or not. On the other hand, non-high-level waste management is a solved problem in most cases, when one excludes wastes from nuclear military programs. The main difficulty with low-level radioactivity waste management is the risk perception by the public, when one tries to relate risks of the order of one in several thousands with numerical dose limits. The environmental effects of nuclear power production should always be controllable by the operators and local competent authorities to maintain and improve environmental quality locally and globally. The main conclusion is that responsible national authorities and international cooperation on developing new technologies and exchanging relevant information on nuclear safety and other nuclear issues are fundamental for a sustainable energy development.

1. Introduction

On January 16, 1939, Lise Meitner and Otto Robert Frisch wrote a letter to *Nature* telling approximately the following:

At first sight, this result (obtained earlier by Otto Hahn and Fritz Strassman) looks difficult to understand. The formation of elements so much lower than uranium (that means with Z much less than 92) was considered before, but was always rejected by physics arguments, because the chemical evidences were not clear. ... However, based on current ideas about the behavior of heavy nuclei, a picture entirely different and essentially classic of these new processes of disintegration becomes evident. ... It looks possible that the uranium nucleus has only a small shape stability and can, after capturing a neutron, split in two nuclei of similar size.

Such splitting process of a heavy nucleus became known as *fission*. The fission fragments should be unstable and subject to a series of beta emissions, forming *fission decay chains*.

The positive balance of the mass-energy conversion started becoming more seriously considered with the discovery of the phenomenon of fission. Niels Bohr while explaining the mechanism of fission pointed out that ^{235}U was the uranium isotope more likely to undergo fission, but its natural abundance was extremely low. Albert Einstein was soon alerted by Leo Szilard, both of them living in the United States at the time that a fission chain reaction had been proposed simultaneously by H. von Halban, Frédéric Joliot and L. Kowarski, in France, and by Enrico Fermi and Szilard himself in the United States. Taking into the proper account the information gathered by Szilard, Einstein wrote to President Franklin Delano Roosevelt on August 2nd, 1939, urging the

President to start considering the possibility that the Germans were likely to develop a military use of the fission chain.

At the beginning of the year 1939, while some scientists still had some doubts about the feasibility of a nuclear reaction chain, there were others who admitted the possibility of a fission chain, but could not accept that the chain would lead to an explosion.

One must bear in mind that most of the ideas concerning the fission phenomenon discussed at that time were only speculations, without the support of experimental data to validate tentative conclusions. Nuclear Physics was then an inchoative subject of scientific studies. However, by mid-1940 there were already enough experimental data to support two important conclusions:

- a nuclear chain reaction could be established and controlled by using slow neutrons to maintain the fission chain; and
- a fission chain with fast neutrons could occur in a very short time (that means, an explosion), making possible to build an atomic bomb.

Recognizing the military importance that a bomb based on nuclear fission could bear in the future, the United States government laid a shield of secrecy on everything connected with nuclear research. At the beginning the secrecy system was volunteer. A scientist would not stop making research on the subject, but would not publish the results of his (or hers) work.

However, as the idea to build an atomic bomb became a mature plan of action, whatever was related to the plan was put under State Secrecy. The number of persons with access to relevant nuclear data was reduced to a minimum. It was common scientific knowledge, however, that ^{235}U was a fissile isotope, which could be fissioned by slow and fast neutrons, but its natural abundance was only 0.72% (i.e., corresponded to only one part in 139 of the more common ^{238}U).

The first letter that Einstein sent to Roosevelt did not have the immediate effect that most people think. A second letter was sent to Roosevelt on March 7th, 1940. Einstein in this second letter expressed to the American President his worries about the German nuclear research on uranium. The nuclear military developments in the United States are registered in a large number of books, not necessarily included in the bibliographical list at the end of this article. For some time now, the history of the Manhattan Project, the mammoth effort made by the United States during the Second World War (WWII) to develop the first atomic bombs, is fairly well known.

All sufficiently industrialized nations that had in 1940 a critical mass of scientists in conditions to understand what had happened in the last year of the previous decade started military nuclear projects, with greater or lesser enthusiasm of scientists and governments. Accordingly, England, France, Germany, Japan, the Soviet Union, as well as the United States all had some kind of nuclear military project at the beginning of, or during the WWII.

The history of the German nuclear project is fairly well known through a series of articles, but the 1967 David Irving's book is by far the best source of information.

Accounts of the nuclear military projects developed in England, Japan and the Soviet Union can be found in the open literature, but it is worth mentioning the books written by Gowing in 1964, Wilcox in 1985, and Holloway in 1994 concerning the military nuclear activities of England, Japan and the Soviet Union, respectively.

After the WWII, nuclear research and development programs, aimed not only to military applications but also to civilian uses of the then new technology, started in most industrialized nations. The First International Conference of the United Nations on the Peaceful Uses of Atomic Energy met in Geneva in 1955. This First Conference dealt most entirely with the fission of the atom.

The United Nations General Assembly, in its resolution 912 (X) on the peaceful uses of atomic energy, adopted unanimously on December 1955, recommended a second international conference to be held in two or three years' time. Preliminary meetings to organize the second conference occurred in September 1956, May 1957 and May 1958.

The Second United Nations International Conference on the Peaceful Uses of Atomic Energy was held in Geneva, 1 September – 13 September 1958. The complete Proceedings of this Second Conference comprise 33 volumes. A vast amount of scientific information on nuclear energy and its applications was published then for the first time.

The prospect of using nuclear power for generating electricity at that time was discussed in the Second Conference under the chairmanship of Sir John Cockcroft. Substantial progress had then been made in the programs to advance the technology of nuclear power plants since the First Conference three years earlier.

Today, forty years have passed since the Second Conference, and the nuclear industry for generating electricity achieved its maturity many years ago. The environmental effects of this mode of producing electricity became focus of intense debates throughout the last few decades.

Environmental effects are present in each phase of the nuclear fuel cycle, though the ways these effects appear and are controlled vary from phase to phase. Thus, they need to be addressed separately in order to be better understood and minimized to the extent possible.

The following sub-sections will summarize the main environmental implications concerning the five main phases of the nuclear fuel cycle. FIGURE 1 illustrates a simplified nuclear fuel cycle to examine the environmental effects of each phase. Future power generation systems and military uses of nuclear energy will not be examined here.

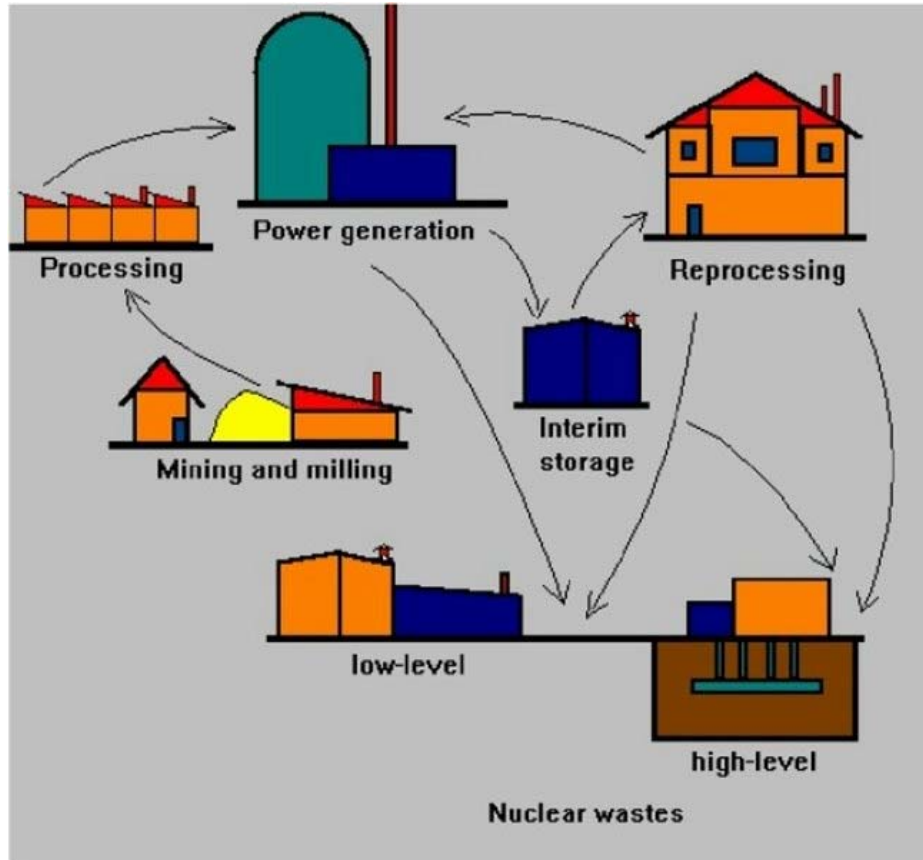


Figure 1. Simplified scheme of the nuclear fuel.

The *front end* of the nuclear fuel cycle consists in those phases from prospecting for uranium ores to the supply of fabricated fuel assemblies to the nuclear power plant, all of them occurring before the nuclear power generation. The *back-end* of the nuclear fuel cycle consists of the phases starting with the removal of spent fuel racks from cooling pools at the reactor site and includes all others all the way up to the disposal of the spent fuel, or of the wastes separated in reprocessing plants. The subsequent sub-sections will summarize the main environmental and regulatory aspects of the different phases of the nuclear fuel cycle.

2. Mining and Milling Nuclear Fuel

After being discovered by prospecting, uranium can be mined in underground or open pit mines. There were in the past deposits with uranium concentrations over 10%, however, most of the economically feasible ores contain around 0.2 % uranium.

In the past, underground mining had important occupational implications, mostly because of radon (^{222}Rn) gas accumulation in galleries. Later, forced ventilation at rates as high as $100 \text{ m}^3 \cdot \text{s}^{-1}$ decreased the radon concentrations in underground mines. Air discharged from underground mines reached radon concentrations as high as $26 \text{ kBq} \cdot \text{m}^{-3}$ ($1 \text{ Bq} = 1 \text{ disintegration per second}$) which was of the order of the natural radon flux from about 1 km^2 of the earth's surface. Underground mining can also be undertaken by *in situ* leaching, mostly when uranium production is resumed in former

mines. The environmental impacts of open pit mines to the environment are usually related to the ^{226}Ra in the overburden and used water, which may be left in the mining site. The average ^{226}Ra concentration in uranium mine overburden was estimated to be about 1 kBq.kg^{-1} in the Western states of the United States.

After being mined, uranium ore is shipped to mills to be transformed into an uranium concentrate, U_3O_8 , known as yellow-cake. In the mills the ore is ground to small size grains. Uranium is then separated by either acid or alkaline leaching. Over 90% of the uranium is removed from the processed ore in the mills, but its decay products such as ^{230}Th and ^{226}Ra remain in the tailings. Dusts containing finely divided grains with ^{230}Th and/or ^{226}Ra can lodge in the lungs. The inert gas radon released from the rocks by the mining operations can deposit its solid daughters, like the alpha emitting ^{218}Po , ^{214}Po and ^{210}Po , in aerosols that after inhalation can lead to significant radiation doses to the lung tissue.

Ranges of ^{230}Th , ^{226}Ra and ^{210}Po concentrations in acid uranium tailings impoundments vary from place to place. Table 1 presents the ranges of ^{230}Th , ^{226}Ra , ^{210}Po and natural uranium concentrations in selected uranium tailings in the United States and Canada.

NUCLIDE	CONCENTRATION RANGES (Bq.g^{-1})	
	Lowest	Highest
^{230}Th	0.7	8400
^{226}Ra	4.0	490
^{210}Pb	7.4	620
Natural uranium (99.8% ^{238}U)	5.1	360

Table 1. Ranges of ^{230}Th , ^{226}Ra , ^{210}Po and natural uranium concentrations in selected uranium tailings impounds.

Most of the ^{226}Ra in tailings effluents is in the solid fraction, however, up to 5% may be found in solution. The potential contamination of surface waters by ^{226}Ra that come in contact with uranium mill tailings is a subject that needs further study, because ^{226}Ra in its soluble form is available to be taken up by plants and animals, becoming part of food pathways. There were mining and milling operations in several parts of the world that resulted in significant ^{226}Ra contamination of surface waters.

After an uranium mill ceases to operate either due to depletion of the ore body or changes in the uranium concentrate procurement policy, the mill is dismantled or use for other type of ores. Protection against environmental releases of radionuclides is then established through a series of procedures. Such procedures may vary from place to place, depending on the limits adopted by national authorities.

Exception made of the highest concentration for natural uranium, all other highest values listed in Table 1 are well above the concentration limits usually recommended by international agencies and adopted by national authorities. As a consequence, there is a

need to mitigate the environmental effects of uranium mines and mills after they cease to operate.

Uranium in yellow-cake is further purified by dissolution in nitric acid and chemical processing. The subsequent steps of the nuclear fuel cycle will be briefly discussed in the next sub-sections.

3. Nuclear Fuel Processing

Pure uranyl nitrate, obtained after chemical processing, is further processed to yield one of the following:

- uranium hexafluoride, a gaseous compound, to be used in enrichment processes;
- metallic uranium for fuelling Magnox reactors; or
- uranium dioxide for fuelling CANDU reactors.

Natural uranium consists approximately of 99.3% ^{238}U and 0.7% ^{235}U . Thus, enrichment processes are used to produce a material containing 3 to 4 % ^{235}U required for light water reactors (LWR), or higher enrichment for more advanced and research reactors. All enrichment processes are based on techniques of isotopic separation. The most common enrichment processes are the following:

3.1. Magnetic Separation

Magnetic separation, which is based on the same principles as mass spectrometry, was used at the beginning of the Manhattan Project. Uranium isotopes are separated by deflecting through magnetic fields electrically accelerated ion beams. Such uranium beams are made of ionized molecules of uranium hexafluoride or vaporized uranium. The lighter uranium isotopes describe circular paths with radii smaller than heavy ones.

3.2. Diffusion of Uranium Gas

Diffusion of uranium gas compounds through porous walls or membranes was the first enrichment process to be used successfully. This process of uranium enrichment was used first in the Manhattan Project. Later on, diffusion plants were also developed in United Kingdom, France and the former Soviet Union. A cascade of membranes is used to enrich uranium hexafluoride vapor from its natural abundance to a higher percentage of ^{235}U isotope. The quality of the membranes has great influence in the efficiency of the process, and a large number of stages are necessary to achieve a significant enrichment. The depleted product is mixed with the feed to the preceding stage for passing again through a membrane. The diffusion process requires a large number of stages, because the separation achieved in each stage is quite small. A typical diffusion plant may have over one thousand stages for enrichment and stripping of ^{235}U .

3.3. Gas Centrifuge

Gas centrifuge separates lighter and heavy uranium isotopes by rapidly rotating a cylinder filled with uranium hexafluoride vapor around its vertical axis. The heavier

molecules go towards the outer wall of the cylinder, while the lighter ones stay closer to the central axis. Gas centrifuge is quite efficient to separate ^{235}U from ^{238}U , in which the number of stages required to achieve the same degree of enrichment is less than that of a diffusion plant by a factor of about 2/100. In addition, the power supply required to run a gas centrifuge plant is about 4% of that needed to run a diffusion plant of the same size. There are gas centrifuge plants in operation, or under construction, in the United Kingdom, United States, Germany, Japan, and Brazil.

3.4. Jet nozzle

Jet nozzle is a uranium isotopic separation process based on forces acting in a jet of uranium hexafluoride mixed with an inert carrier gas, usually hydrogen, which describes a curvilinear trajectory. Typical feed gas has 4% uranium hexafluoride and 96% hydrogen. This process has been developed at the Nuclear Research Centre, Karlsruhe, Germany. In 1975 a cooperative effort started between the German nuclear center and the Brazilian company NUCLEBRAS. There are (or there were) research projects on the jet nozzle process in Germany, South Africa, and Brazil. The efficiency of the jet nozzle project is not as good as that of the gas centrifuge.

3.5. Laser Separation

Laser separation is based in exposing high temperature uranium vapor to a laser specially tuned to ionize the isotope ^{235}U . Once the isotope ^{235}U is ionized it can be electro-magnetically separated from the non-ionized isotope ^{238}U . There is ongoing research on laser separation in a number of countries.

3.6. Chemical and Other Methods

Chemical and other methods can also be used to separate light isotopes. Such methods can be potentially used for uranium enrichment. In the case of chemical exchange methods the separation factors for heavy elements like uranium are rather small. Other methods, such as fractional distillation, thermal diffusion, chromatography, electrolysis, physical adsorption, as well as rotating plasmas have been reported to be potentially applicable to separate isotopes.

The most important environmental effects expected to occur in uranium enrichment facilities are those due to leaking of fluoride compounds. An accidental leak of uranyl fluoride in the Kerr Mcgee plant, in the United States, on January 05, 1986 is the best known event of this kind. One person died, and various workers had to be evacuated from the immediate vicinity of the site of the accident.

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Biographical Sketch

Anselmo Salles Paschoa was born in Rio de Janeiro, Brazil, on December 15, 1937. Mr Paschoa holds a PhD degree from New York University. He is a Full Professor at Pontifícia Universidade Católica do Rio de Janeiro – PUC RJ. He was Visiting Associate Professor at the University of Utah, Guest Scientist at Brookhaven National Laboratory, and Visiting Scientist at Memorial Sloan-Kettering Cancer Center. Dr Paschoa was Guest Lecturer in courses offered by the Pan-American Health Organization, the Universidad de Sevilla, the International Atomic Energy Agency (IAEA), and the International Center for Theoretical Physics. He is member of several scientific and professional societies and associations. He was member of the Board of Directors of the International Union of Radioecologists, and Vice-President of the International Radiation Physics Society for Central and South Americas. Dr Paschoa has published

more than 100 scientific articles and papers. He is member of several international scientific committees in the fields of Radiation Physics and Environmental Sciences. Dr Paschoa was Director for Radioprotection, Nuclear Safety and Safeguards in the Brazilian Nuclear Energy Commission (CNEN) from May 1990 to August 1992. In September 1991, he was head of the Brazilian Delegation to the General Conference of IAEA and Alternative Governors of Brazil to the Board of Governors of the IAEA from 1990 to 1992. Recently, Prof. Paschoa was member of the United States National Academy of Sciences Committee on Evaluation of EPA Guidelines for Exposure to Naturally Occurring Radioactive Materials, National Research Council.

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