ENVIRONMENTAL EFFECTS OF NUCLEAR FUEL REPROCESING

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Contents

Summary

During the last decades, the safety, health and environmental consequences of technological and industrial development have included a growing social concern and a incentive for the rapid improvement of risk assessment and management techniques. Thus, the health and environmental impacts of the nuclear fuel cycle, and of fuel reprocessing have been extensively studied. Past experience with various facilities, and a whole spectrum of potential consequences related to normal operation and accidental situations have been analysed in the framework of studies performed at the design stage of installations. Although a vast amount of information based on experience and analysis is available, being important to recognize the fact that some elements of the environmental safety issue remain controversial.

The purpose of this paper is to give overall perspective on the health and environmental consequences of fuel reprocessing taking in account the new realities and the state-of art of the technology. Reprocessing is presently reaching its full industrial maturity in a few countries but some shadows are darkering worldwide nuclear development. In spite of its present stagnation, reprocessing and recycling is nowadays the only route proven for minimizing environmental impacts and that effectively demonstrates the safety of facilities.

1. Introduction

Energy is essential for human life. Urbanization, industrialization and a rising standard of living will lead to growing energy demand. This is a major factor which will give rise to increasing use of energy in the form of electricity.

Despite conservation and increased efficiency in the use of energy, a variety of economically available energy sources will have to be used to meet future energy demands. The selection of appropriate energy sources involves economic, environmental and technical issues, including availability of national resources and balance of payments. The use of any energy sources has some effect on the environment. It is difficult to compare the impacts on environment and public health of all energy sources, but such comparasions are necessary if well informed decisions are to be made.

The purpose of this work is to give a general description of health and environmental aspects of reprocessing, a step of the nuclear fuel cycle.

1.1. Nuclear Energy and the Nuclear Fuel Cycle

The discovery of nuclear fission and the energy that can be released by means of fission are well known today. To describe the meaning of reprocessing some features of nuclear reactor and the relation between fuel reprocessing and other parts of the nuclear fuel cycle will be reviewed.

1.1.1. The Fission Process

Nuclear fission is a highly exothermic process characteristic of certain nuclides; the fissionable materials of importance to reactors are ^{235}U, ^{239}Pu, and ^{233}U. When the process of nuclear fission is carried out under suitably controlled conditions by a self-sustaining chain reaction, the condition is known as *criticality*. Four aspects of fission are of interest: (1) much energy is released; (2) the fissionable atoms are destroyed; (3) neutrons are released; and (4) fission products are formed.

The release of energy is the purpose for which most reactors are built today. The energy may be used for conversion to electricity, for propulsion of ships, or for process heat. The generation of electricity from nuclear energy is already an economic reality in some countries.

The destruction of the fissionable atoms is significant to the fuel cycle because the destroyed atoms must eventually be replaced if the chain reaction is to be maintained. The atomic depletion, or *burn-up*, is measured as a percentage of the fissionable atoms originally present or as the number of megawatt-days of energy liberated per tonne of fissionable and potentially fissionable material originally present. The neutrons that are released and the fission products that are formed have special importance to the fuel-reprocessing plant.

For each atom that fissions, two or three neutrons are emitted. These neutrons have energy of 1 to 10 Mev. They are of importance because of their interations which nuclei of atoms in the reactor. In some cases a neutron rebounds from nucleus which the loss of energy by the neutron and a increase in energy by the interacting nucleus. In other cases the neutron is absorbed in the nucleus, usually with one of the following results: (1) the nucleus fissions, (2) the atom is transmuted to a gamma-emitting isotope of the same element, or (3) the new nucleus is transformed by radioactive decay into some other element.

The energy of neutrons is related to their speed. Thus high-energy neutrons (i.e., neutrons having energies greater than about 0.1 Mev) are called *fast neutrons*, and the loss of energy that occurs during nonabsorption collisions is known as slowing down. Neutrons having speed equal to that which they would have as a result of thermal agitation are called *thermal neutrons*. The energy corresponding to this speed, for ordinary temperatures, is about 0.025 ev, but for practical purposes the thermal-energy range is generally considered to include all energies less than about 1 ev. Since the neutrons produced by fission have na energy of approximately 1 to 10 Mev, they are fast neutrons.

The absorption of neutrons is characterized by the probability of its occurrence. This probability is quantified by a *cross section*. Some atoms have high cross sections for neutrons and are sometimes referred to *as neutron poisons* because they absorb neutrons so readily. The cross section of a nucleus for any given process is a function of the neutron energy.

Certain materials (^{238}U and ^{232}Th) are not themselves fissionable under most conditions, but they can be transmuted into fissionable materials by the absorption of neutrons. Such materials are called *fertile materials*. The absorption of neutrons to produce fission or to convert fertile materials into fissionable materials are two desirable process, but other absorptions are undesirable and are frequently referred to as *parasitic absorption* or *parasitic capture*.

The part of a reactor that contains the fissionable material is called the *core*. In addition to fissionable material, the core contains coolant for carrying away the heat liberated, structural material to support the fissionable material and to protect it from corrosion, and sometimes a moderator for slowing down the neutrons to the thermal-energy range. All these materials have some cross section for parasitic absorption, but this cross section is kept as small as is feasible.

The fissionable material in most reactors is in the form of a solid (a metal or an oxide) and is nearly always contained in a cladding material to prevent fission-product escape and corrosion of the fissionable material by the coolant. The combination of the fissionable material contained in cladding and supported in a structural assembly is called a *fuel element*.

1.1.2. Reactor Types

Reactors are classified in several ways. The kind of fissionable material used, the energy of the neutrons responsible for most of the fissions, and/or the extent of utilization of fertile materials determines the basic reactor type. Reactors that use ^{239}Pu or ^{233}U can be characterized by the fissionable isotope alone, but reactors that depend upon the fission of ^{235}U are characterized by the concentration of ^{235}U atoms in the total uranium present, i. e., the *enrichment*. Uranium in nature contains 0.72 at. % of ^{235}U; this concentration is adequate to support a chain reaction under suitable conditions. Many rectors, however, use slightly or highly enriched uranium.

According to neutron speed (energy), reactors are classified as *thermal reactors* or *fast reactors*. Intermediate reactors, which use neutrons intermediate in energy, i.e., between fast and thermal, have been built, but they appear to offer no compelling advantages. Thermal reactors are easily controlled and can use natural uranium as a fuel. Fast reactors are generally small in size and in some cases have an advantage in the conversion of fertile materials.

According to their utilization of fertile materials, reactors can be classified as (1) burners, (2) converters, or (3) breeders. Converters and breeders can be further classified according to whether the fertile material is in the reactor core (one-region reactor) or outside of the core in a region called the *blanket* (two-region reactor). A *burner* is a reactor that consumes fissionable material without converting fertile material to any appreciable extent.

A *converter* reactor produces a significant amount of fissionable material from fertile material but does not produce as much fissionable material as it consumes. A *breeder* reactor produces as much fissionable material as it consumes or more than it consumes. The distinction between converters and breeders is thus rather artificial and depends on the breeding ratio, i.e., the ratio of atoms of fissionable material bred to atoms of fissionable material consumed. A reactor having a breeding ratio near unity (about 0.95 to 1.05) is called near breeder.

Three kinds of reactors are of current or potential importance: thermal converters, fast breeders, and thermal breeders. Nearly all the power reactors built in the early 1960's are thermal converters and use natural or slightly enriched uranium as the fuel. They have a breeding ratio of 0.6 to 0.7. Fast breeder reactors use ^{239}Pu or ^{235}U as the fissionable material.

The breeding ratio for ^{239}Pu is 1.6, and for ^{235}U, about 1.2. Although ^{233}U can breed in both a thermal reactor and a fast reactor, the fast ^{233}U breeder is usually considered less attractive than the other types of breeders. The breeding gain that can actually be

obtained from a reactor is less than that indicated by the neutron emission per absorption in fissionable atoms and by fission-capture cross sections for fertile materials. This is due to parasitic losses of neutrons to fission products, structural materials, control rods, reflector, and moderator (if used) and to reprocessing and re-fabrication losses.

1.1.3. Nuclear Fuel Cycle Description

The nuclear fuel cycle deals with the preparation of the fuel for the safety utilization in the nuclear reactor. The main activities in the nuclear fuel cycle, comprise the following: mining, milling, refining and conversion, enrichment, fuel fabrication, spent fuel reprocessing and disposal. It is not included nuclear plants were the nuclear chain reaction occurs. There are two possible configurations of the fuel cycle (see Figure 1):

- The *once through* fuel cycle, in which the ore is made into fuel passed through the reactor once and is then stored in waiting for final disposal.
- The *reprocessing cycle*, in which the fuel is passes through the reactor, reprocessed an passed through the reactor again.

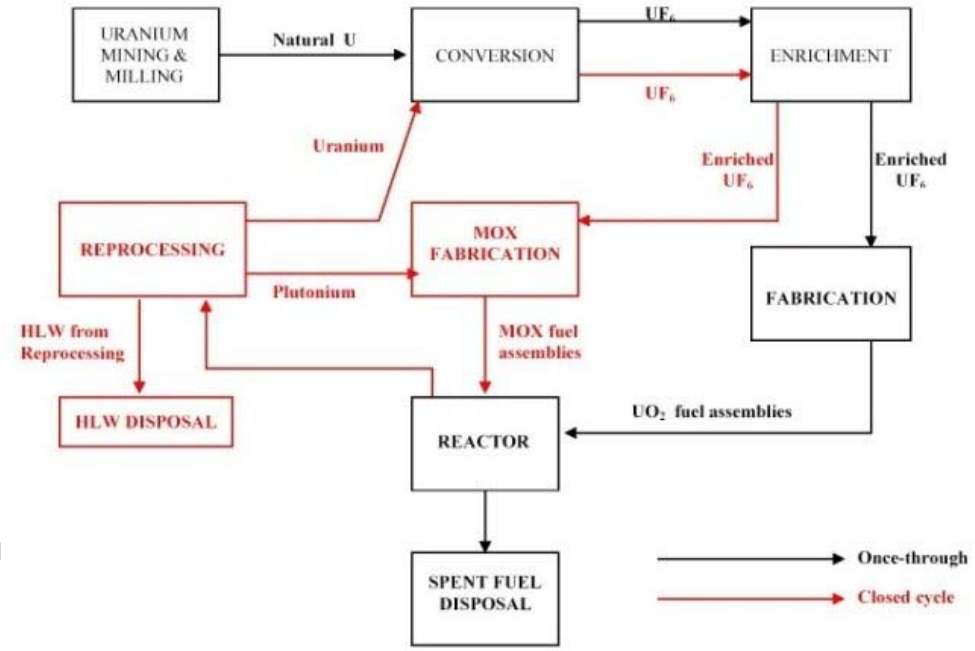

Figure 1. Schematic representation of once-through and closed fuel cycles.

The difference between the once through cycle and the reprocessing cycle is that the reprocessing cycle makes more efficient use of the fuel through extraction of Pu and recycling of ^{235}U.

2. Reprocessing

Energy production using nuclear fission generates fission products and actinides. Reprocessing is an important step of the nuclear fuel cycle and deals with the chemical

separation of the fission products from the actinides remaining in the spent fuel and/or produced during the reactor operation from the irradiation of ^{238}U.

2.1. Decay

Solid fuel elements in a nuclear reactor must sooner or later be removed and replaced for one or more of the following reasons: (1) replenishment of fissionable material; (2) excessive corrosion and radiation damage; (3) recovery of converted (bred) fissionable material; and (4) removal of neutron-absorbing fission products from the recovered material.

When the closed cycle is choose, prior to reprocessing, the fuel is stored for a period of time to allow a portion of the radioisotopes to decay (see Figure 2).

The purpose of the decay is to simplify the shipment (if necessary) and the reprocessing by:

- the essentially complete removal of certain troublesome fission product isotopes having short *half-life*;
- a reduction in the amount of high-energy gamma-emitting isotopes, as well as in the overall intensity of the radiation emitted, and thus a reduction in the amount of shielding necessary and in the damage done to process chemicals and equipment;
- the formation, sometimes, of a desired isotope by the decay of a precursor specie (the obtention of ^{233}U from the decay of ^{233}Pa, in the ^{232}Th cycle).

The optimum length of time for the decay is the time that permits decay of the most troublesome fission-product activities consistent with the fuel-inventory charge. The longer the decay period, the less expensive is the reprocessing owing to lower shielding requirements and disapperance of some troublesome radioactive species, but the advantage diminishes rapidly with time.

On the other hand, the inventory charge for fissionable material increases with increasing length of the decay period. The minimun over-all cost must balance these two factors. If long irradiation times are employed in the reactor, the length of time for decay is a less important fraction of the total inventory charge.

The decay of radioactivity is very rapid at first but becomes increasingly slow with the passing of time. For many purposes the minimum decay period is considered to be the time necessary for the practical disappearance of ^{131}I. The decay period can also be influenced in an overriding manner by the time necessary for the formation or disappearance of a transmutation product.

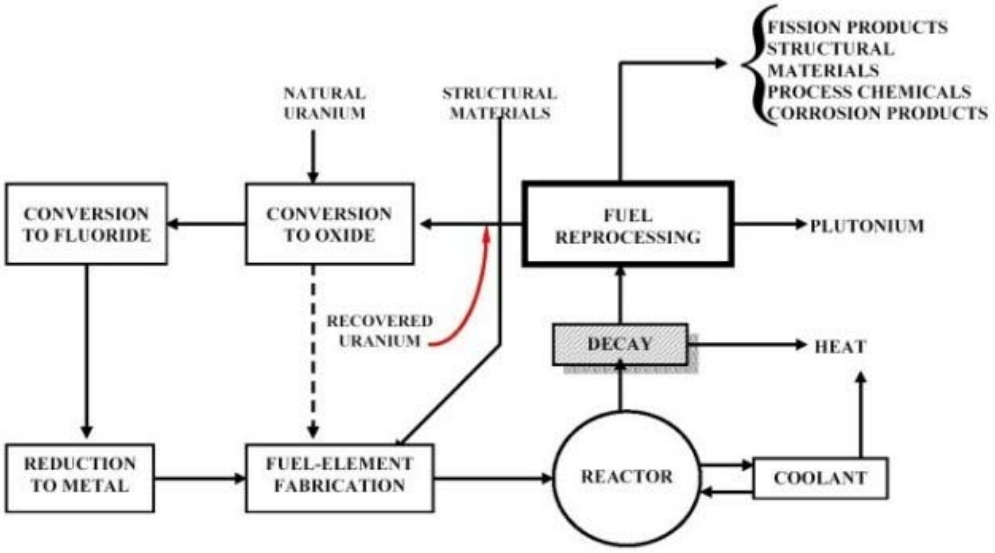

Figure 2. Fuel cycle for a natural-uranium reactor with aqueous fuel reprocessing.

2.3. Build-up of Radioactive Nuclides

The reprocessing of spent fuel requires an understanding of the laws governing the formation of radioactive nuclides in the neutron flux of a reactor and the subsequent decay of these nuclides both in the reactor and after their removal from it. This knowledge is necessary to an understanding of the separation that must be accomplished, the precautions needed to avoid criticality, the amount and kind of biological shielding that must be provided, the amount of heat that must be removed from irradiated material, and the composition of the radioactive wastes that must be stored or otherwise disposed of.

The laws describing the physical processes are amenable to rigorous mathematical formulation, and most of the necessary physical constants (cross sections, fission yields, half lives, and branching ratios) are known, though not always with high precision. Hence, in principle, the properties of spent fuel can be calculated for any condition of irradiation and decay. To do so here is impractical. However, because of the almost infinite variety of possible irradiation and decay conditions and because of the strong influence of these conditions on the properties of spent fuel.

Radioactive nuclei in spent nuclear-reactor fuel emit alpha, beta, and gamma radiations; delayed neutrons are also emitted from fuel immediately after removal from the neutron flux of a reactor. These emissions reduce the energy level of the excited nuclei to a stable state, and no further radioactivity is then released. The number and nature of the desintegrations required before the stable state is reached is a characteristic of the particular nuclei. Most radioactive nuclei go through several transformations.

Radioactive-decay emissions were named for the first three letters of the Greek alphabet in the progressive order of their penetrating powers. Alpha radiation, a particle that is identical to the nucleus of the helium atom, is the least penetration; it can be stopped by a sheet of ordinary paper. Although alpha particles cannot penetrate the skin, they can

cause serious local damage to tissue if atoms that emit then are deposited internally by ingestion or inhalation or in an open wound. For example, plutonium is an alpha emitter that tends to locate in the bone marrow, where an accumulation of minute quantities can be fatal.

The second radioactive-emission is the beta particle, an energetic electron. With the emission of a beta particle the nucleus having atomic number Z is transmuted to a new element having atomic number Z + 1. Practically all fission products are beta emitters because the fission process leaves the nucleus with an overabundance of neutrons and a paucity of protons. Hence the nucleus tends to eject an electron (which in effect transforms a neutron into a proton) to attain a more stable ratio of neutrons to protons.

The third means by which an excited nucleus loses energy in reaching the ground state is by the emission of a gamma photon. The gamma photon is electromagnetic radiation similar to visible light but having relatively high energy. The wave length is a factor of about 10^5 shorter than that of light, and the penetration power of gamma radiation is appreciable in all matter. Unlike alpha and beta radiations, the emission of a gamma photon does not transmute the atom but only reduces the atom to a lower energy state.

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Biographical Sketch

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