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NUCLEAR WASTE MANAGEMENT ORGANIZATION
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NWMO BACKGROUND PAPERS
6. TECHNICAL METHODS

6-4 STATUS OF NUCLEAR FUEL REPROCESSING, PARTITIONING AND TRANSMUTATION

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NWMO Background Papers

NWMO has commissioned a series of background papers which present concepts and contextual information about the state of our knowledge on important topics related to the management of radioactive waste. The intent of these background papers is to provide input to defining possible approaches for the long-term management of used nuclear fuel and to contribute to an informed dialogue with the public and other stakeholders. The papers currently available are posted on NWMO's web site. Additional papers may be commissioned.

The topics of the background papers can be classified under the following broad headings:

1. **Guiding Concepts** – describe key concepts which can help guide an informed dialogue with the public and other stakeholders on the topic of radioactive waste management. They include perspectives on risk, security, the precautionary approach, adaptive management, traditional knowledge and sustainable development.
2. **Social and Ethical Dimensions** - provide perspectives on the social and ethical dimensions of radioactive waste management. They include background papers prepared for roundtable discussions.
3. **Health and Safety** – provide information on the status of relevant research, technologies, standards and procedures to reduce radiation and security risk associated with radioactive waste management.
4. **Science and Environment** – provide information on the current status of relevant research on ecosystem processes and environmental management issues. They include descriptions of the current efforts, as well as the status of research into our understanding of the biosphere and geosphere.
5. **Economic Factors** - provide insight into the economic factors and financial requirements for the long-term management of used nuclear fuel.
6. **Technical Methods** - provide general descriptions of the three methods for the long-term management of used nuclear fuel as defined in the NFWA, as well as other possible methods and related system requirements.
7. **Institutions and Governance** - outline the current relevant legal, administrative and institutional requirements that may be applicable to the long-term management of spent nuclear fuel in Canada, including legislation, regulations, guidelines, protocols, directives, policies and procedures of various jurisdictions.

Abstract

The basic concepts of the reprocessing, partitioning, conditioning and transmutation of nuclear fuel are explained. A Canadian context is established by discussing the characteristics of the fuel from CANDU reactors. The technology of reprocessing is introduced in terms of both the Purex and the dry reprocessing options. The world status of commercial reprocessing is reviewed with detail for the UK, France, Russia, Japan, India, the US and Canada. Current research on the transmutation of fission products and actinides is reported and the fundamental concepts of the fast reactor and Accelerator Driven System approaches presented. The technical aspects of reprocessing relevant to possible decisions on its application in Canada are summarized in issue related sections for easy reference.

Table of Contents

1. Introduction

The purpose of this report is to provide the technical information surrounding a very basic question about nuclear waste management:

- What could be done with used nuclear fuel to reduce the quantity and toxicity of the radioactive materials it contains?

One's first thought is that it would be good idea to do what ever can be done to achieve a smaller volume of less toxic radioactive waste. However, as will be explained below, there are many issues to be addressed concerning the feasibility and desirability of doing so.

1.1 Technical Background

The term nuclear fuel cycle means all the processes involving the fuel for electric power generation in a nuclear reactor. [Those requiring more basic information to follow this paper are referred to books such as references [1] and [2] which explain the essential concepts of nuclear technology in layperson terms.] Most common fuel cycles begin with the mining of uranium. The raw ore goes through a variety of chemical processes to extract, mill, concentrate, and refine the uranium in a suitable form for use in a reactor. Most of the world's nuclear reactors are of the LWR (light water reactor) type. Mined uranium ore contains only 0.7% of the isotope uranium-235 with the remainder uranium-238. For LWRs natural uranium must be enriched in uranium-235 to between three and five percent. However, the fuel for the present generation of Canadian (CANDU) heavy water reactors is natural uranium and enrichment is not necessary. The uranium following purification and conversion to uranium oxide is then incorporated into fuel assemblies manufactured in various sizes and shapes depending on the reactor type.

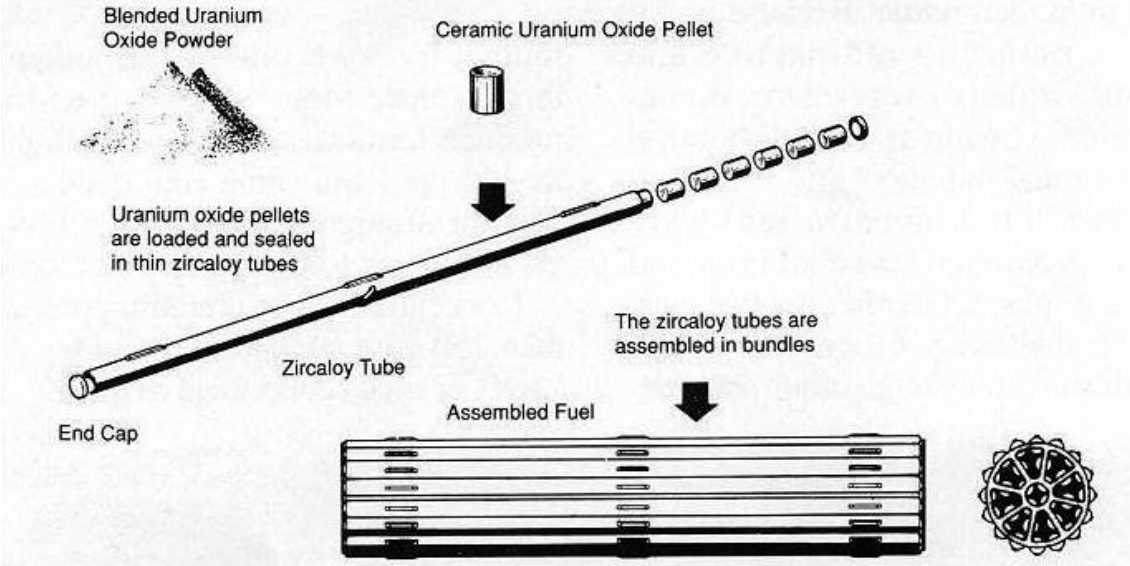

Figure 1 CANDU Fuel Bundle (by permission of AECL)

As shown in Figure 1, CANDU fuel is manufactured by sintering uranium oxide powder into pellets. The pellets are loaded into zircaloy (an alloy of the metal zirconium) tubes which are then welded into a bundle of tubes as shown, i.e. a fuel bundle. The zircaloy parts of the fuel bundle are collectively known as the cladding. The bundles are then inserted into CANDU reactors where nuclear fission reactions in the fuel are used to generate electricity.

There are many possible fission reactions, all producing different fission products. Many of these fission products are radioactive, several of them intensely so. Some fission products absorb neutrons and after a certain point the fuel must be removed because neutron absorption makes the fission chain reaction increasingly difficult to maintain. The net effect is that a substantial percentage of the original uranium-235 remains in the used fuel in addition to a similar amount of plutonium-239, another fission fuel, produced by neutron absorption in uranium-238.

1.2 Reprocessing, Partitioning and Transmutation

Nuclear energy has been in place for some fifty years and has achieved industrial and commercial maturity with about 400 reactors producing electricity worldwide. Most nuclear power reactors are LWRs using enriched uranium fuel. The only other commercially successful reactor type is the Canadian CANDU reactor.

In the early days of nuclear development there was a concern that the natural sources of uranium fuel were limited and therefore, it was essential that reactor concepts should be efficient users of uranium. Both the LWR and CANDU reactor types fulfilled that criterion. Furthermore, substantial plutonium is generated by both reactors and this was considered an asset, particularly for the LWR because plutonium can be recycled to fuel not only reactors with moderated neutrons (thermal reactors) but also fast reactors.

Fast reactors are reactors in which the neutrons from fission are not slowed down or moderated by ordinary (light water) as is the case for the LWR or heavy water as in CANDU. These fast reactors use enriched uranium and plutonium fuel and are cooled by liquid sodium. They generate enough fast neutrons that they can in a “breeder” configuration produce more plutonium than they consume by converting uranium-238 by neutron absorption to plutonium-239.

Fast breeder reactors were considered as the long-term solution to the problem of uranium supply because there is abundant uranium-238 available, 99.3% compared to 0.7% uranium-235 and also because, as will be seen below, used fuel from thermal reactors contains all but a small percentage of its original uranium-238. Moreover, there are large stocks of uranium-238 at enrichment facilities left over from the removal of the uranium-235, known as depleted uranium.

The operating experience with fast reactors has been mixed. Safety problems have been experienced mainly involving sodium leaks. For example, in 1995 Japan’s Monju fast breeder reactor had a serious sodium leak, which was initially concealed by the reactor managers. Monju is slated for restart soon and together with Phénix in France and BN-600 in Russia will be among the few such reactors still in operation. In addition, increasing discoveries of uranium ore bodies, including the very rich ones in northern Saskatchewan, and the global slow down in the deployment of conventional power

reactors has put the original need for fast reactors very much in the future. With proven uranium supplies projected to last for some 50 to 100 years at current rates of consumption, it would require a resurgence of nuclear power to bring this issue to the fore again.

Reprocessing is a general term for applying chemical and physical processes to used (spent) reactor fuel whereas "processing" is taken to mean the preparation of fresh fuel before it goes into the reactor. The recovery and recycling of fissionable isotopes is the main reason for reprocessing reactor fuel in countries operating LWRs. France, Japan, the United Kingdom, and Russia reprocess fuel from light water reactors whereas the United States, with some 100 LWRs in operation, does not.

Reprocessing technology was first developed and exploited in the nuclear weapons programs of the United States, the United Kingdom and Russia and later in the military programs of France, China and India. The aim was to exact plutonium-239 from fuel irradiated in specially designed plutonium production reactors. The other main weapons material, uranium-235, was produced in uranium enrichment plants. Hence, several countries had already made very large military investments in nuclear fuel cycle facilities by the time their civilian nuclear power programs began. The presence of that existing infrastructure clearly influenced the choices made on the adoption of fuel cycles.

In recent years, with no urgency for uranium recycling or for the recovery of plutonium for fast reactors, interest in reprocessing has become more oriented to the role it might have in mitigating the problems presented by the disposition of used nuclear fuel.

Turning to the technology of reprocessing, fuel is taken from the reactors and stored temporarily in water pools in order for it to cool since radioactivity generates substantial heat even after the fuel is removed from the reactor. This is done for all nuclear fuel whether or not it is to be reprocessed. After a few years the radioactivity and consequent heat decrease significantly. The fuel is then taken to a reprocessing plant in large lead and steel casks. Once there the fuel assemblies are opened in facilities designed to prevent the escape of volatile radioactive gases. The fuel pellets are removed from the cladding and dissolved in nitric acid. Chemical separations are performed to remove uranium-235 and plutonium-239. The fission products are also removed and may be further separated into groups of elements. This process of separating and segregating the various components of the fuel is called **partitioning**. Sometimes the term is more narrowly taken to mean the separation of the plutonium in the fuel from the uranium.

The fissionable isotopes extracted from the fuel by reprocessing can be recycled into fresh fuel depending on the particular nuclear fuel cycle being used. However, as we have seen, fission products are not desirable in fresh fuel because among other things they absorb neutrons making the chain reaction difficult to sustain. Therefore, at present the fission products are prepared for long-term geological storage [often called "disposal" but not in this paper –see section 6.3], for example by embedding them in molten glass, and thereafter may be temporarily stored in a nuclear waste management facility. Prior to geological storage, it may also be desirable to change the chemical form of certain isotopes. For example, volatile radioactive iodine gas can be converted to silver iodide a stable solid much preferable for storage. This sort of chemical improvement of nuclear waste prior to storage is known as **conditioning**. It's important to note that even though reprocessing may reduce the volume of the fuel waste by recycling and segregating its highly radioactive components, it does not avoid the need for a geological storage

facility.

Reprocessing and partitioning perform separations and modifications based on chemical and physical properties but do not reduce the quantity or toxicity of the nuclear fuel; they merely rearrange and recycle its components. Nevertheless, it may be possible to transform some of the radioactive components of the fuel into non-radioactive elements using nuclear reactions initiated by neutrons, protons or even photons from lasers. This is called **transmutation** meaning changing one element to another. At the moment transmutation to reduce the radioactivity of nuclear waste is not a practical reality but because of its potential benefits, it is the subject of research programs in many nuclear countries through the world.

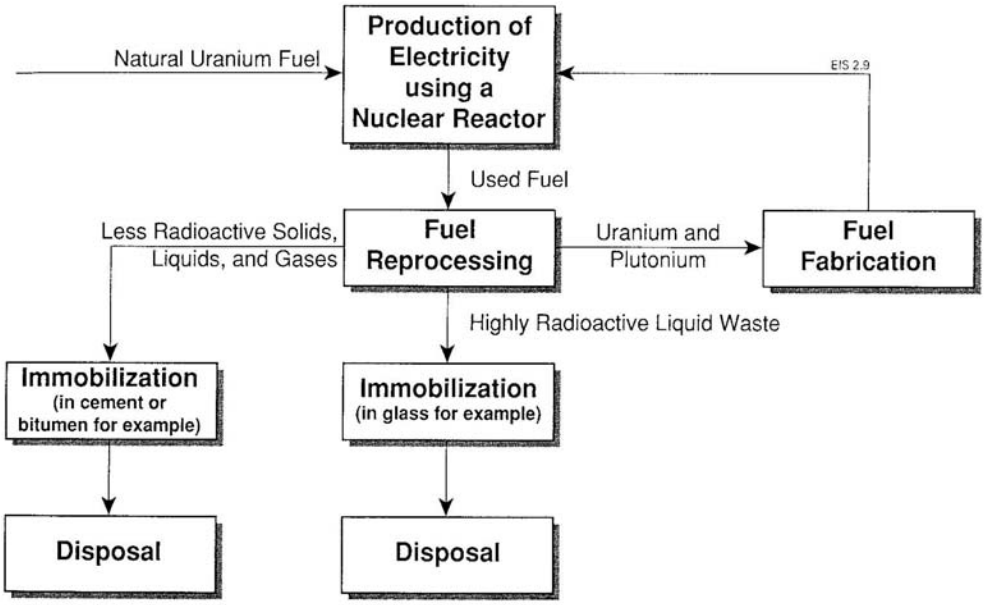

Figure 2 Schematic representation of a nuclear fuel cycle involving reprocessing, partitioning of the fuel into three streams is represented in the Fuel Reprocessing box. Transmutation would be an option just prior to Immobilization steps. From [3] (by permission of AECL)

Figure 2 shows the relationship of the terms defined above for the case of a CANDU system where the input is natural uranium fuel. In the Fuel Reprocessing box, the uranium-235 and plutonium-239 are separated and sent to a Fuel Fabrication step for recycling as new fuel for the Production of Electricity. In partitioning the fuel and removing the fission products, the Fuel Reprocessing step also produces a stream of less radioactive wastes such as gloves, gowns, ion exchange resins, air filters and so forth which have been contaminated in the reprocessing process. Both streams of radioactive by-products undergo an Immobilization step. Here the highly radioactive fission products are embedded in glass blocks and the less radioactive by products are incorporated in cement or bitumen prior to Disposal, meaning long-term geological storage. Transmutation is not shown in the figure because it is not yet a feasible step in reprocessing. When it becomes practical, it would appear before the Immobilization boxes.

2. CANDU Fuel

In Canada there is currently no enrichment of the reactor fuel and no reprocessing of nuclear fuel. The fuel removed from CANDU reactors is stored in water pools for a minimum of six to ten years and is then placed in convection cooled concrete silos or bunkers at the reactor sites for interim dry storage pending a decision on long-term disposition. Nevertheless, it is useful to examine reprocessing and the other issues discussed in this document in a Canadian context to the largest extent possible and thus, the emphasis in this section will be on CANDU fuel.

2.1 Spent CANDU Fuel

CANDU fuel is removed from the reactor when about 67% of the uranium-235 is consumed which takes about one and a half years for an average fuel bundle. Table 1 compares the composition of a standard CANDU fuel bundle (burn up of 685 GJ per kg U) from [3]) after its time in the reactor compared to fresh fuel.

Table 1 Composition of fresh and spent CANDU fuel [3]

Component	Fresh	Spent
Uranium-235	0.72 %	0.23 %
Uranium-238	99.28 %	98.70 %
Plutonium-239	-	0.27 %
Fission Products	-	0.80 %

Several conclusions bearing on reprocessing can be made from Table 1.

- Very little of the material in the fuel is actually changed, only about 1.3% of the original material, even though a large quantity of energy has generated from it, a characteristic of nuclear energy.

- According to numbers provided by NWMO, there are about 1.6 million used CANDU bundles now in storage. In general these bundles are of different types and have had different irradiation histories. To make rough estimates assume each bundle contains on average 19 kg of uranium and that the average content of a spent bundle is as given in table 1. Using these approximations in the about 30,400 tonnes of used fuel there are roughly 82 tonnes of plutonium, 70 tonnes of unused uranium-235, 243 tonnes of fission products and 30,005 tonnes of uranium-238.

- Much of the mass of the fuel remains uranium-238 and therefore, from this point of view alone, substantial reductions in waste volume could be made just by removing this relatively harmless element.

- A significant amount of the isotope plutonium-239 has been produced in the fuel by the absorption of a neutron according to the reaction:

  uranium-238 + neutron → plutonium-239 + ….

This is an example of transmutation whereby a uranium nucleus is converted to a nucleus of plutonium by absorption of a neutron.

- Plutonium-239 is a fissionable fuel. Approximately 0.6% of the original uranium-238 transmutes to plutonium-239. More than half (0.35%) of this plutonium undergoes fission and thus, about 30% of the energy produced from CANDU fuel is from the fissioning of plutonium.

- It is clear from Table 1 that much fissionable material remains in the fuel. The total plutonium-239 and uranium-235 content is 0.5% of the fuel, compared to the starting content of 0.7% uranium-235. Therefore, an incentive for reprocessing the fuel would be to remove the unused uranium-235 and plutonium-239 for recycling in fresh fuel.

The radioactive isotopes in spent nuclear fuel can be conveniently divided for discussion into three classes. The first consists of the fission products. Many products are produced in fission reactions some are stable and others radioactive. The radioactive ones may decay in complex chains to produce even more radioactive isotopes. The radioactive fission products produce the intense radiation and heat of the fuel bundles. Therefore, the bundles must be handled remotely and transferred to a swimming pool type of temporary storage facility where their radiation is shielded and their heat cooled by the water. After a few years of under water storage many of the short lived fission products have decayed but the radioactivity of other isotopes persists for long times - the more important ones are shown in Table 2.

Table 2 The more significant fission products

Fission Product	Half-Life (years)
Krypton-85	11
Strontium-90	29
Technetium-99	210,000
Tin-126	100,000
Iodine-129	16,000,000
Cesium-135	2,300,000
Cesium-137	30

There are observations to be made relevant to reprocessing.

- Some fission products have long half-lives and for this reason alone the fuel bundles will be radioactive for millions of years.

- Kryton-85 and iodine-129 are gases that, unless properly controlled, could escape if the bundles were reprocessed.

- The decay of strontium-90 and cesium-137 is the primary source of the residual heat in the fuel bundle after the cooling period. Therefore, removal of these

isotopes from the fuel would relax the requirements for heat dissipation in a long-term burial depository.

A second category of isotopes present in fuel are the actinides, elements with atomic numbers from 89 (actinium) to 103 (lawrencium). Some times the same elements are referred to as transuranic elements, meaning elements containing more than 92 protons (uranium) in their nuclei. Technically speaking, the difference is that uranium is an actinide but it's not a transuranic. Often the two terms are used interchangeably. Actinides are produced from neutron absorption reactions, which do not result in fission but rather result in transmutations. This type of reaction has been illustrated above for the uranium-238 to plutonium-239 transmutation. Actinides are all radioactive and alpha particle emission is a common mode of decay for them. The main actinides of interest in spent fuel are shown in Table 3.

Table 3 Main actinides in spent fuel

Isotope	Half-life (years)
Uranium-235	710,000,000
Uranium-238	4,500,000,000
Plutonium-239	24,000
Plutonium-240	6,600
Plutonium-242	360,000
Neptunium-237	2,100,000
Americium-241	460
Americium-243	8,000
Curium-244	18

Reprocessing relevant observations about the actinides are as follows.

- As noted previously, the recovery of the fissionable fuels, uranium-235 and plutonium-239, is a major impetus for reprocessing.

- With a few exceptions actinides generally have rather long half-lives and are the primary sources of radiation in a fuel repository after the first few hundred years when much of the decay of the most radioactive fission products has taken place. If the actinides could be destroyed by nuclear reactions (and also a few long lived fission products) then the environmental requirements for long-term burial of the fuel would be much less stringent.

- Based on the variety of actinides present, it can be inferred that attempts to reduce the quantity of actinides by the absorption of the relatively slow (thermal) neutrons found in LWR and CANDU power reactors would just produce even more actinides and little progress would be achieved in destroying them.

The third and final category of radioactive materials in spent fuel comprises the radioactive isotopes resulting from neutron reactions with materials in the fuel cladding as distinct from the fuel itself. They are called activation products since they arise from non-radioactive materials that have been made radioactive (activated) by fission neutrons. Some of them are shown in Table 4.

Table 4 Activation products in Fuel

Isotope	Half-life (years)
Carbon-14	5,700
Clorine-36	300,000
Zirconium-93	1,500,000

Some comments on Table 4 with respect to reprocessing follow.

- For CANDU fuel it turns out that 95% of the radioactivity is in the fuel and only 5% in the cladding.

- Most of the zirconium radioactivity is from zirconium-93 formed by the absorption of a neutron in zirconium-92, which occurs in natural zirconium at 17% abundance. Removal of this isotope from zirconium metal prior to fabricating the fuel assemblies would significantly reduce the radioactivity from activation products.

2.2 Radioactivity and Radiotoxicity of CANDU Fuel

Figure 3 illustrates the decay of the radioactivity over time in a kilogram of irradiated uranium (left hand axis) and in the standard CANDU fuel bundle (right hand axis) as a function of the time in years. The unit Bq, Becquerel, means one decay event per second.

This figure puts the contributions of the various components of CANDU fuel in a comparative perspective. Actinides are included in the group “Uranium and Activation Products in the Fuel Pellets”. It illustrates the following points, some of which have been touched on earlier.

- A period of about a million years must pass before the fuel decays to the same level of radioactivity found in natural uranium ore.

- The radioactivity due to fission products decays to the natural uranium level in about 1,000 years.

- The activation products in the fuel cladding decay to the natural uranium level in less than 100 years.

- After about 1,000 years, the radioactivity is dominated by the actinides i.e. the “Uranium and Activation Products in the Fuel Pellets” category.

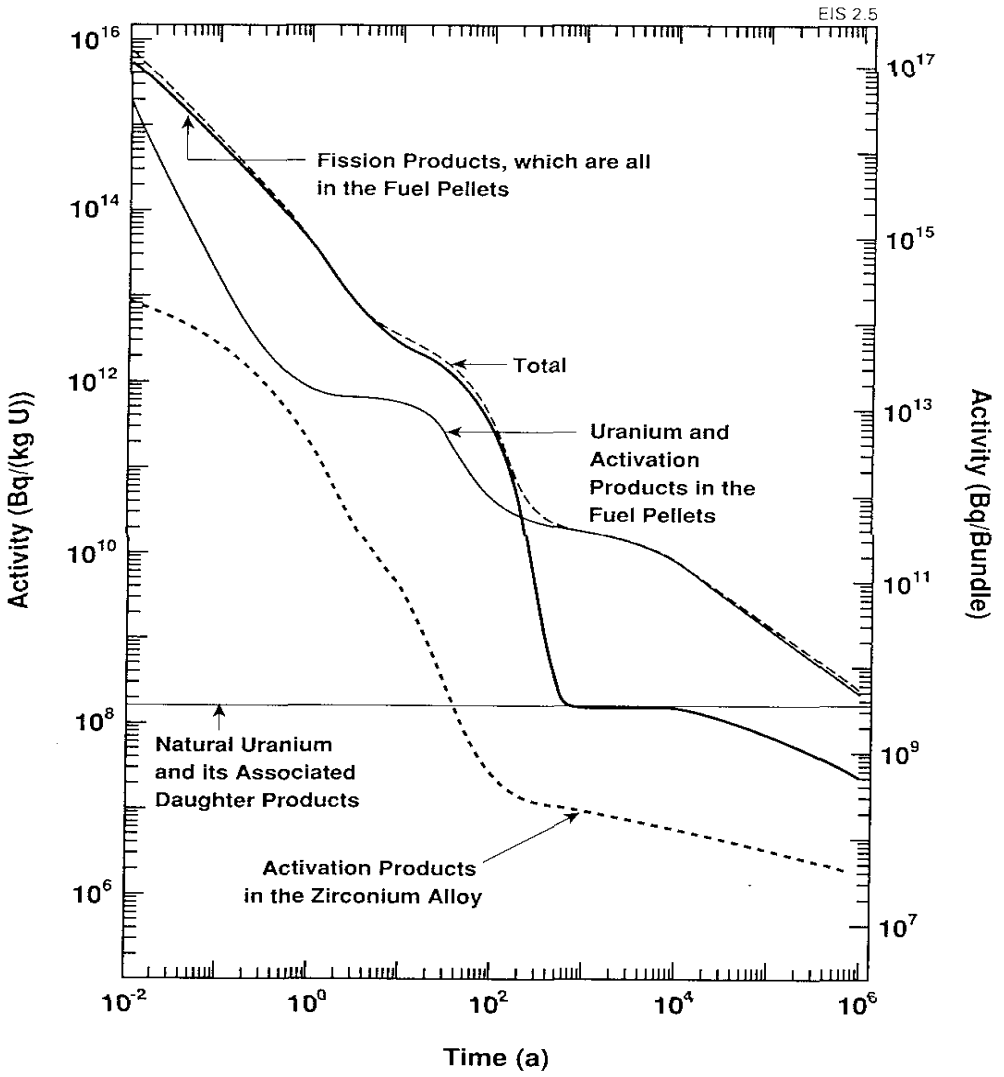

Figure 3 Decay over time in years of the radioactivity from a kilogram of the reference CANDU fuel (left side) and of a standard fuel bundle (right side) – see section 2.1 [3] (by permission of AECL)

Whereas Figure 3 shows the total radioactivity for the groups of elements in the fuel it does not take into account the relative radiotoxicity of those elements. Each one has its own radioactive decay emissions at specific energies. Depending on these factors and including the relative biological impacts of the emitted particles, a radiotoxicity factor can be defined as the number of fatal cancers and serious genetic effects that would be experienced by a person swallowing a gram of the material in question. This is shown in Figure 4.

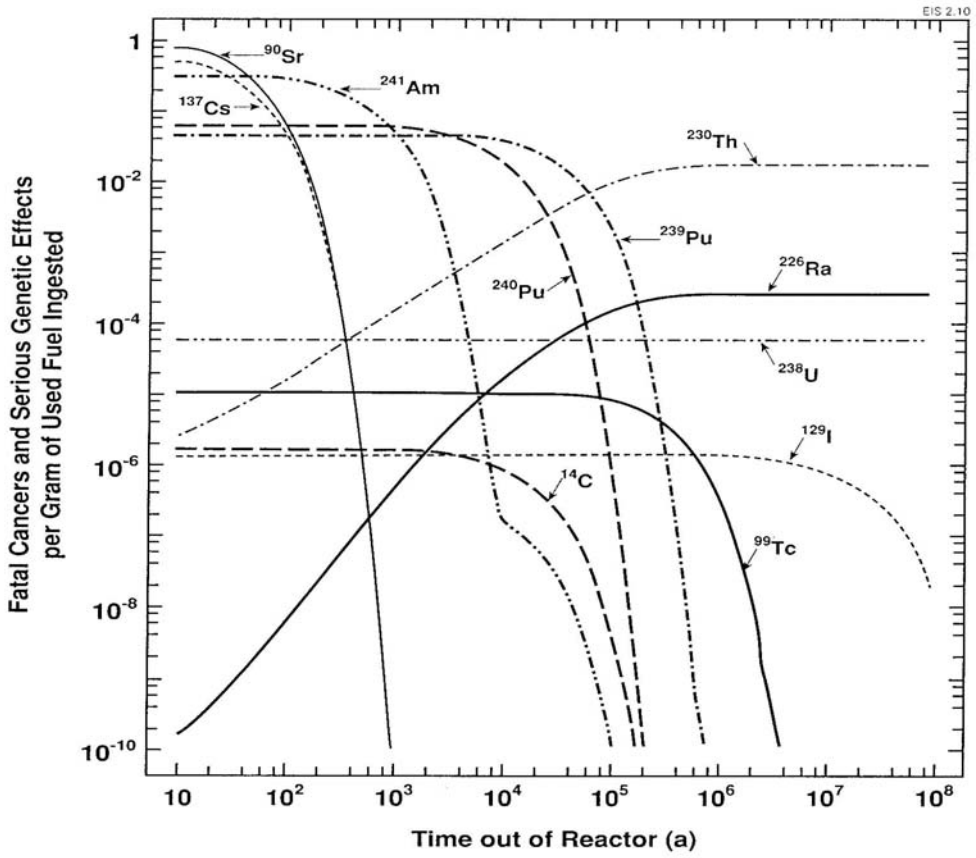

Figure 4 Radiotoxicity of various radioactive isotopes in CANDU fuel [3] (by permission of AECL)

This figure helps to identify the isotopes that would be best removed by reprocessing and/or destroyed by transmutation. (Note that Figure 4 contains estimates based on the 1991 recommendations of the International Commission on Radiation Protection (ICRP) and would need to be recalculated using more recent ICRP values if it were to be used for quantitative purposes. It is included here for qualitative purposes only.)

In terms of designing a depository for long-term geological storage of radioactive materials, either intact fuel or isotopes extracted in reprocessing, another important consideration in addition to relative radiotoxicity has to be folded into the calculations. Namely what radioactive isotopes would have the highest probability of escaping from the facility into the biosphere, for example by ground water transport? Therefore, the optimum means of reducing the total impact on the biosphere of the radioactive material stored can only be understood in terms of the total system. This viewpoint might have strategic implications on where to concentrate efforts on transmutation. Iodine-129 in addition to having significant radiotoxicity for very long times also has relatively high mobility whereas although the radiotoxicity of technetium-99 is larger or comparable, technetium might diffuse much more slowly out of a depository.

2.3 Comparison of CANDU and LWR Fuel

It was mentioned above that several countries operating light water reactor (LWR) plants reprocess their fuel. It is useful therefore to look at the differences between irradiated CANDU fuel and LWR fuel

Table 5 Composition of fresh and spent LWR fuel [3], [4]

Component	Fresh	Spent
Uranium-235	3.3 %	0.81 %
Uranium-236	-	0.51 %
Uranium-238	96.7 %	94.3 %
Plutonium-239	-	0.52 %
Plutonium-240,241,242		0.36
Fission Products	-	3.5 %

Comparing Table 5 with Table 1, the general observation is that there is more of everything present in LWR fuel compared to CANDU fuel. The initial enrichment in uranium-235 means more fission reactions have occurred with more fission products and actinides produced. Significant quantities of uranium-236 and the higher isotopes of plutonium are present because the fuel has been burned to a higher energy level and that increases the time for neutron absorption leading to more actinides. The fissionable components for recycling total 1.34% compared to 0.5% in CANDU fuel and thus, the incentive to recycle is increased two to three fold in terms of deriving the most energy from a given quantity of uranium.

Note that the burn up for this particular LWR example is about 2,800 GJ/kg U compared to 685 GJ/kg U (7,900 MWd/ton U) in typical CANDU fuel. This factor of four difference means that four times as much spent fuel is produced by CANDU reactors compared to LWRs for the same amount of energy. Much of this difference, all uranium-238, is accounted for by the substantial amounts of depleted uranium (uranium-238 containing about 0.2-0.3% uranium-235) left at the enrichment plants. However, it is much easier to deal with depleted uranium than having the “extra” uranium-238 incorporated in spent fuel with fission products and actinides, as is the case for CANDU.

2.4 CANDU Fuel Cycles

To date CANDU fuel waste consists only of irradiated natural uranium fuel bundles. However, many other fuel cycles for CANDU are now under investigation either to decrease the capital costs of building reactors, the objective of the Advanced CANDU Reactor (ACR) now under development by AECL, or to permit the use of other fuels as a response to declining uranium resources. The CANDU reactor is particularly flexible in terms of fuel cycles because of its excellent neutron efficiency, which allows operation with relatively low concentrations of fissile material. Many of the advanced fuel cycles require reprocessing and hence, if adopted domestically, would require a Canadian reprocessing plant or more likely the purchase of reprocessing services at a foreign plant. Figure 5 illustrates some of the more prominent CANDU fuel cycles.

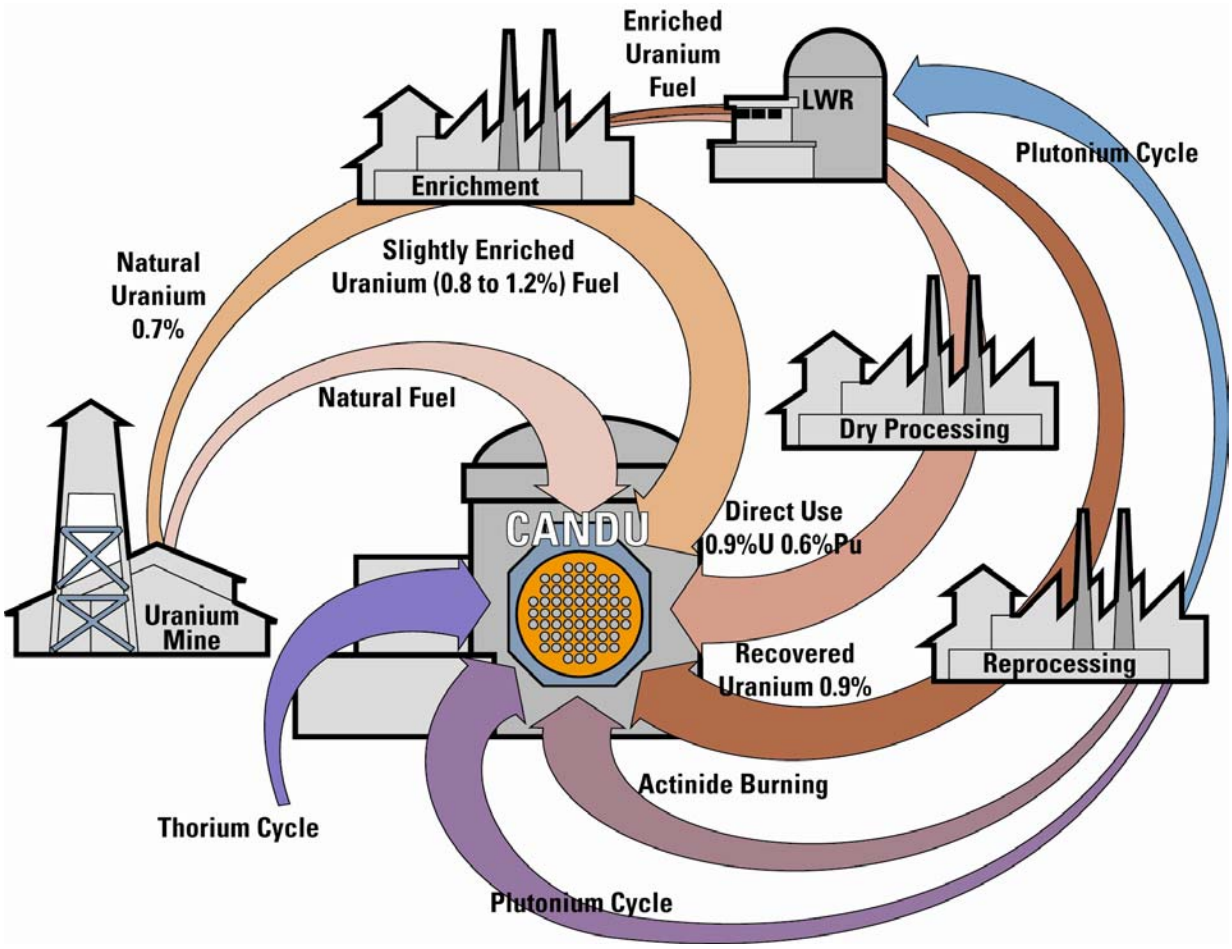

Figure 5 Possible CANDU fuel cycles [5] (by Permission of AECL)

The Natural Uranium Fuel cycle uses uranium not enriched in uranium-235 and is the cycle now used in CANDU reactors. On removal, the fuel is now stored without reprocessing, awaiting final disposition.

The Slightly Enriched Uranium (SEU) fuel cycle involves fuel with a slight enrichment in uranium-235 between 0.8 and 1.2% compared to the natural content of 0.7%. It could be used in existing CANDU reactors without changes to the reactor design and would increase the energy produced per kilogram of fuel by a factor of 2 to 3 while reducing fuel costs by about 30%. [5] The quantity of spent fuel produced per unit of electricity produced would also be reduced. However, Canada would have to buy enriched uranium on the world market.

A Low Enriched Uranium (LEU) fuel, between 2 and 4% uranium-235, is projected for AECL's Advanced CANDU Reactor (ACR) now under development. Using this fuel would reduce the size of an ACR by up to 30% compared to a current CANDU of the same power. Reducing the size would also reduce the comparative capital cost of the

reactor for a given power. Again the quantity of spent fuel per unit energy would decrease.

DUPIC stands for “Direct use of PWR fuel in CANDU” fuel cycle. A PWR is the most common configuration of LWR. Table 5 shows that fuel from a LWR contains about 1.3% fissile materials (uranium-235 and plutonium-239), which, although too low a percentage for re-use in a LWR, could be used in a CANDU. The cladding of the fuel would need to be mechanically removed in a special facility and the fuel treated by a dry process to remove fission products. The resulting powder would be pressed into pellets and then made into fuel bundles. This dry reprocessing would have advantages over traditional aqueous methods. The resulting fresh fuel would yield a factor of two more energy than the original fuel. This concept is currently being studied by Korean utilities, who would like to optimize their electrical production by first burning fuel in LWR stations and then recycling it in the Korean CANDU’s.

In the 1960’s the Thorium fuel cycle was put forward by the originators of the CANDU concept as the best means of extending the stocks of nuclear fuel [6]. By neutron capture, the naturally occurring isotope thorium-232 transmutes to uranium-233, which can fissioned. It takes some uranium or plutonium to get the thorium cycle started but the production of uranium-233 is much more efficient than that of plutonium-239 in other fuel cycles. This high efficiency means that only a relatively small amount of fresh fissionable fuel needs to be added on each cycle for the system to be self-sustaining. This make up fuel could be produced by other means such as an accelerator. An additional feature of the thorium cycle is that fewer long lived actinides are produced.

The Recovered Uranium fuel cycle uses uranium recovered from the reprocessing of LWR fuel in CANDU fuel. For example reprocessing plants in Europe extract uranium with 0.9% uranium-235. This enrichment is suitable for CANDU fuel but not LWR fuel and thus, would recycle LWR uranium. This recycling not only extracts more energy from the originally mined uranium, but also significantly reduces the amount of waste per unit energy to be disposed of.

The Tandem fuel cycle uses mixed oxide (MOX) fuel, formed by mixing plutonium-239, as plutonium dioxide, with natural uranium dioxide to form fuel (in the form of ceramic pellets). Plutonium from European reprocessing plants is currently recycled with enriched uranium and now used in more than thirty European reactors. In a similar manner, mixed oxide fuel can be made from plutonium obtained by reprocessing US and Russian military plutonium from dismantled nuclear weapons. The CANDU is particularly well suited for plutonium recycling with MOX. Experiments have recently been performed in Canada using MOX containing plutonium derived from both US and Russian weapons.

Although the examples discussed in this section are for the CANDU reactor, some important points can be made on fuel cycles in general.

- All reactor types have several potential fuel cycle options, some of which can be very complex involving the use of more than one reactor type.

- Nuclear fuel cycles can be optimized to extend the supply of nuclear fuels, to reduce the radiotoxicity of the resulting fuel or for other reasons such as the maximum production weapons materials.

- Reprocessing is an integral component of many fuel cycles.

- As yet, fuel cycles requiring uranium enrichment have not been used in Canadian power reactors but this may happen in the future and hence, may require either the construction of domestic enrichment facilities or more likely the purchase of enriched uranium abroad.

- Similarly, it is not clear whether the future use of advanced fuel cycles in Canada would create a domestic need for reprocessing or the purchase of offshore reprocessing services.

- Using fuel cycles other than the current once through natural uranium cycle in Canada would result in spent fuel with somewhat different characteristics. Depending on the degree of difference, changes in the strategy for nuclear fuel waste management might be required, perhaps even to the extent of requiring reprocessing.

3. Reprocessing Technology

Historically the primary objective of reprocessing reactor fuel was to extract plutonium and uranium for recycling but more recently the reduction of radiotoxicity and the separation of radioactive isotopes for conditioning and eventual transmutation has become increasingly important.

Partitioning the fuel during reprocessing separates it into the following groups.

- plutonium
- uranium-235 and uranium-238
- minor actinides (MA)
- fission products

The minor actinides (MA) group consists of isotopes of neptunium, curium and americium (see Table 3). The plutonium group is mainly plutonium-239 but plutonium-240, 241 and 242 are present in smaller quantities. Similarly the uranium-235 group will contain some uranium-232, -233, -234 and uranium-236. The fission product group is extensive; some of the more important ones are shown in Table 2. Reprocessing opens up a wide variety of options for the individual treatment of these groups. Recycling of plutonium and uranium and eventual transmutation of MA and fission products are possibilities.

The major technology for reprocessing is the Purex process, which is used in every large plant throughout the world. This is a wet process involving the dissolving of the fuel in nitric acid. Dry processes usually grouped under the heading “pyrochemical” are more sophisticated methods that need to be used to separate isotopes that are hot, both radioactively and thermally, to a very high level of purity.

3.1 The Purex Process

As documented in [7], several chemical processes had been used in the 1940's to extract plutonium for weapons from irradiated reactor fuel. It was found that the best was the Purex process developed in the early 1950's at General Electric in the US. A Purex pilot plant started in 1952 at Oak Ridge, Tennessee. Large Purex plants were constructed at Savannah River, South Carolina (1954) and at Hanford, Washington (1956). Subsequently, most of the plutonium used in the world's nuclear arsenals was extracted using the Purex method. It also became the major reprocessing method used for civilian nuclear fuel throughout the world.

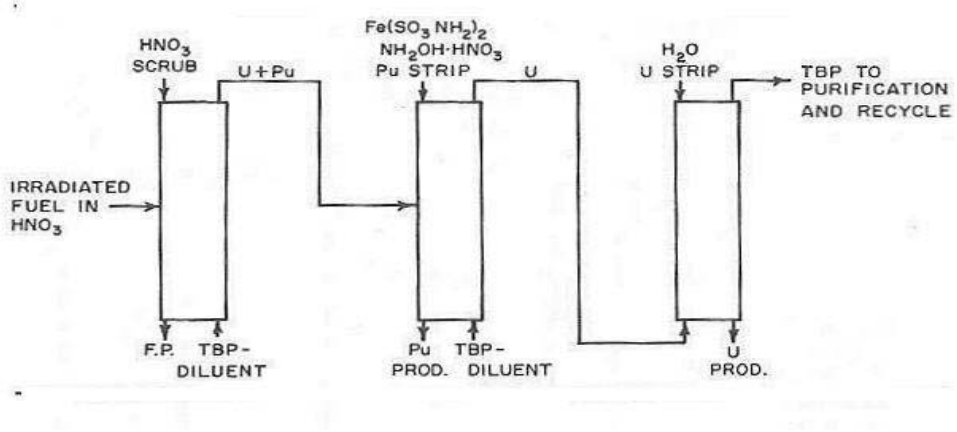

Figure 6 Schematic of the Purex Process from [6] (by permission of AECL)

Following Figure 6, the first step of the process is to open the fuel assembly by sawing, cutting or shearing so that the fuel pellets can easily be extracted from the cladding. Since finely divided zirconium can burn, this must be done in an inert atmosphere or under water. When the fuel is opened gaseous fission products such as krypton-85, xenon-135, carbon-14 and tritium are released and must be taken off in separate streams for specialized treatment.

The fuel and cladding are then immersed in hot nitric acid. The cladding is unaffected by the nitric acid but any fuel particles adhering to it are removed. Oxides of nitrogen given off during the dissolution step are absorbed to reconstitute the nitric acid. At this point the remaining fission product gases trapped within the fuel are released and piped to the off-gas treatment streams. The fuel assemblies are then washed in water and checked to ensure that a high percentage of the fission products and fuel have been removed. They are then appropriately prepared for geological storage.

The uranium reacts to form uranyl nitrate $UO_2(NO_3)_2$ resident in the nitric acid solution. The pH of the nitric acid solution is adjusted and the plutonium is converted to its tetravalent form by addition of N_2O_4 resulting in plutonium appearing as a nitrate,

$Pu(NO_3)_4$. The next step is the key solvent extraction process. A 30% solution of tributyl phosphate (TBP) in a solvent with high boiling point (usually n-dodecane or other paraffin-like compound) is flowed through a pulsed counter-current column to contact the nitric acid solution. Both the uranium and plutonium nitrates preferentially concentrate in the TBP whereas most of the fission products remain in the aqueous nitric acid solution. Thus, the uranium and plutonium are stripped out of the solution with high efficiency.

The separation, or partition, of the plutonium is done by reducing it to its trivalent state which is insoluble in TBP by adding, for example, hydroxylamine which is not strong enough to reduce the uranium. The plutonium nitrate is then purified by additional solvent extractions in separate loops. After the partitioning of plutonium, the uranyl nitrate in solution is washed out of the TBP with water and then purified in another system of columns.

The volatile gases are a particular concern in reprocessing. There is no practical way of storing large quantities of krypton-85 and this gas is vented to the atmosphere. The argument is that it is a noble gas that has no physiological interaction with humans or animal life and therefore, does not damage the biosphere. With a half-life of 11 years, there is some validity in this line of reasoning. Iodine-129, half-life of 16 million years with an affinity for the human thyroid, is released to the sea for dilution at some reprocessing plants, a somewhat more controversial procedure.

The above is a highly simplified summary of a very complex process, elaborated in great detail in [7]. Many variants and extensions of Purex have been developed over the years, some of them proprietary. Similarly many specialized side processes to treat specific elements are now in use.

It should be emphasized that reprocessing plants are expensive both to build and to operate since they must fulfil several stringent requirements.

- Reprocessing procedures must be carried out by remote manipulation because of the high radioactivity of the fuel.

- Elaborate and effective containment measures must be used to prevent the release of fission products to the environment, particularly the volatile gaseous ones.

- Rigid control of radiation exposure to plant workers must be exercised in a highly radioactive environment in order to ensure occupational safety.

- Since plutonium-239 and uranium-235 are nuclear weapons materials, tight physical security of the plant is essential and strict accountability of these materials during every stage is needed in order both to avoid unauthorized diversion and also to satisfy international monitoring and inspection.

- Economic operation requires that nitric acid, TBP, solvents and other high value chemicals be recycled with losses minimized.

- Hazards to safe operation such as spontaneous criticality, chemical fire and explosion and venting, leaking or other breaches of containment must be avoided.

3.2 Dry Reprocessing Methods

Pyrochemical and pyrometallurgical processes for nuclear fuel have advantages compared to liquid methods such as potentially more effective separations for certain isotopes and less secondary waste streams. These methods could lead to better separation of actinides and of long-lived fission products like cesium-135. They have already been used with good success for experimental reprocessing of fast reactor fuels in the US (in the 1960's) and later in Russia and are now the subject of considerable research and development worldwide.

Several options for pyrochemical processing methods are now used and researched. [8]

Under specific conditions passing an electric current between two electrodes embedded in molten salt results in the "plating" out of a pure material usually at the cathode (or negative electrode). In this class of electrochemical processes the fuel including opened cladding forms the other electrode (anode) and the radioactive isotopes in the fuel are transported in the salt from the anode to the cathode. By varying factors like the concentration of the salt, the salt temperature and other parameters, the process can be adjusted to be highly selective for certain isotopes. The molten salt has to be at high temperature (500°C) and the decay heat of the fuel can be used to advantage to achieve this. Depending on the details of the particular process used, the molten salt can be a mixture of chlorides, for example lithium chloride and potassium chloride. This is called electro-refining and a variation with fuel isotopes present as components of the molten salt is called electro-winning. Multiple cathodes designed to capture individual isotopes are possible as are liquid metal (cadmium) cathodes to differentially plate out actinides.

Reacting natural uranium with fluorine to produce uranium hexafluoride (UF_6) is the first step in uranium enrichment. In fact about 80% of Canada's uranium exports are shipped in this gaseous form. Several other actinide fluorides are possible, PuF_6 among them, and a variety of isotope separation schemes can be envisaged using this approach, called fluorine volatilization. Other variations under consideration involve the volatility of chlorides such as $ZrCl_4$. Research in this promising area concerns the control of these processes to yield high efficiency separations and the difficulties of controlling very corrosive gases.

Combinations of both of the above general classes of methods in addition to selective reduction techniques appear to have promise to achieve the degree of isotope separation required for the transmutation schemes to be discussed below. Thus, to a large extent the eventual practicality of transmutation will depend on the progress made in bringing these methods to practical fruition.

A disadvantage of dry processes is that they are essentially batch processes, meaning that only a limited amount of material can be treated at any given time, in contrast to wet (aqueous) methods such as Purex in which continuous streams of materials are pumped through the system. This limitation negatively impacts the economics of current pyrochemical processes and may also incur additional costs, for example, the need to

replace and safely dispose of the large crucibles used for pyrochemical processes. Another difficulty is in developing materials able to withstand the effects of corrosive chemicals at high temperatures. Much research needs to be done before dry processes can go into large-scale commercial production.

4. Global Status of Reprocessing

4.1 Overview

Reprocessing is a large and potentially profitable international business. Many nations built large reprocessing facilities for military purposes and with the resulting expertise and experience were easily able to transition to the reprocessing of civilian fuels. [9-11]

Table 6 World Commercial Nuclear Fuel Reprocessing Capacity

Country	LWR Fuels (tonnes/year)	Other (tonnes/year)
France, La Hague	1600	
France, Marcoule		400
UK, Sellafield		1500
UK, Sellafield (THORP)	850	
Russia, Chelyabinsk (Mayak)	400	
Japan	90	
India		200
Total	**2940**	**2100**

Table 6 shows the main players in civilian reprocessing. The UK and France handle the largest volumes with both of them reprocessing the fuel from several countries. Countries such as China [12] and South Korea are also getting into the field. The table is arranged in terms of LWR fuel and “Other”. Generally speaking the latter covers mostly metal fuels from Magnox, research and fast breeder reactors. For India this category includes the reprocessing of the fuel for their CANDU type reactors. LWR fuel is initially in the form of oxides and generally oxide and metal fuels are reprocessed in different facilities. To date about 75,000 tonnes of civilian fuel have been processed by all countries. [9]

4.2 United Kingdom

Sellafield (Figure 7) is a large multipurpose nuclear complex established in 1946 to produce nuclear materials, notably plutonium, for the British nuclear weapons program. It was also the site of the world’s first nuclear commercial power station, the four-unit Calder Hall, which began electricity production in 1956. Reprocessing of the nuclear fuel from Calder Hall and the other UK power reactors that followed was done to extract the plutonium for use in weapons. Weapons materials are no longer produced but a large

stockpile (50 to 100 tonnes) of weapons and civilian plutonium is located at Sellafield. [10], [13].

Figure 7 Sellafield nuclear complex in the UK. The THORP facility is in the large complex with the white roof in the centre of the photograph (by permission British Nuclear Fuels Limited)

The first reactors at Sellafield used natural uranium metal fuel with a graphite moderator and air coolant, which was the best way at that time to produce the plutonium. The graphite in one of the dedicated plutonium production reactors, Windscale 1, caught fire in 1957 and burned for about a day. This tendency of graphite to burn under accident conditions was also a factor in the Chernobyl accident where the release of fission products was about 1,000 times greater than the Windscale accident. Both plutonium production reactors were shut down soon after the accident.

This same reactor configuration, called Magnox, was subsequently used for the four Calder Hall reactors on the Sellafield site. Eventually 26 Magnox reactors were built for electric power generation. Since Magnox fuel is made from uranium metal, it is not suitable for long-term storage or burial because of its chemical reactivity, unlike fuels based on uranium oxide which is a ceramic. Therefore, it is essential to reprocess Magnox fuel and for this reason alone, the UK required a reprocessing plant.

At present the two main activities at Sellafield are reprocessing the fuel from the UK Magnox fleet, many of which are being decommissioned and, reprocessing uranium oxide fuel from the commercial nuclear power stations of several nations. In 1994, the Thermal Oxide Reprocessing Plant (THORP) began operation. Owned and operated by British Nuclear Fuels Ltd. (BNFL), it is a capital-intensive facility, built at a cost of £1.85

billion ($4.2 billion CDN) with the required ancillary clean up plants costing an additional £1 billion ($2.25 billion CDN) [10]. THORP, with a capacity of 850 tonnes per year (Table 6), uses a Purex type process to recover uranium and plutonium from used nuclear fuels based on uranium oxide. It was reported [10] that in 1999 THORP had an estimated £12 billion ($22.5 billion CDN) worth of reprocessing orders up to 2010 and acceptance of orders beyond that are subject to government approval. About half of these orders were from non-UK sources.

The policy on reprocessing for foreign countries is to return all the products extracted from the fuel, including the fission products, to the country where the fuel originated. These profitable foreign orders help to defray the cost of THORP and Sellafield in general.

Other activities at Sellafield include a MOX fuel manufacturing capability and some processing of nuclear wastes from other countries, for example, the extraction of slightly enriched uranium from liquid nitric acid wastes from Purex reprocessing in the US weapons program.

4.3 France

France is the other main player in global commercial reprocessing after the UK. [14]

There is little domestic uranium but a strong desire for energy independence in France. Therefore, the government selected the reprocessing option early in the civilian nuclear program. A law was passed in 1991 that research and development in the following areas should be performed with respect to nuclear fuel until 2006.

- separation and transmutation of long-lived radioactive nuclides,
- retrievable or non-retrievable underground repositories
- nuclear waste conditioning and long-term storage.

The French nuclear strategy includes a MOX fuel program and operation of a fast reactor, Phénix. The main players are Electricité de France the utility which operates about 60 LWR power reactors producing 75% of France's electricity, COGEMA the nuclear fuel company that does the reprocessing (and has uranium holdings in Canada), ANDRA the national agency responsible for waste repositories and CEA the nuclear research organization.

The primary use of the recycled plutonium is for MOX fuel, which is now used in many of Electricité de France's reactors, particularly the larger ones (up to 1450 MWe). Generally these reactors use mostly standard enriched uranium fuel with a minority component (30%) of MOX fuel. MOX fuelling strategies are under study concerning the enrichment level of the uranium and the plutonium content to maximize the overall energy from the fuel cycle.

Small MOX fuel plants are located in Belgium and southern France with the main one (MELOX) operated by COGEMA at Marcoule in Normandy. In addition to production for European utilities, Japanese MOX fuel will be made at MELOX.

The major French reprocessing capability resides in the two large plants at La Hague, UP2 and UP3, in Normandy (Figure 8). These plants reprocess not only French nuclear fuel but also fuel from Germany, Switzerland, the Netherlands, Belgium and Japan. Both

plants are operated by COGEMA with UP2 dedicated to French fuel and UP3 to foreign fuel. By 1998, the total fuel recycled at La Hague had exceeded 15,000 tonnes, roughly half from France and the other half from Europe and Japan.

Figure 8 Reprocessing plant at La Hague France (COGEMA Archives/Phillipe Lesage)

Future fuel recycling in France envisages a double strata (two reactors) approach employing both thermal and fast reactors with reprocessing step in between. Research and development on separation and transmutation in carried out under the SPIN program.

4.4 Russia

Russia in its past as a nuclear superpower built very large reprocessing facilities for military purposes. Therefore, a substantial technological base was available for civilian reprocessing. [15]

Fuel from the Russian LWR reactor type, the WWER, is reprocessed at the RT-1 plant in Chelyabinsk run by the PO Mayak consortium. RT-1 has a capacity of 400 tonnes per year and has been operating since 1976. By 1999 a total of 3000 tonnes of fuel had been reprocessed. Reprocessing has been performed not only for the countries of the former Soviet Union but also for countries of the former Warsaw Pact: Bulgaria, Czechoslovakia, and Hungary. RT-1 uses the same Purex process used elsewhere.

It is interesting that Russia is the only country that recycles uranium-235 from fuel. This is done by blending the uranium-235 from LWR fuel with that derived from fuel from ship propulsion and breeder reactors. This is fabricated into in an LEU (2-2.4% enrichment) fuel for Russia's RMBK reactors.

Substantial amounts of plutonium are stored at the Chelyabinsk site including 30 tonnes of civilian plutonium and 50 tonnes of military plutonium from dissembled weapons. Like other countries removing plutonium from fuel, Russia cannot consume it fast enough even though it does have the largest fast reactor program in the world. The BN-600 fast reactor is the world's most successful fast reactor in terms of sustained reliable operation and three new BN-800 fast reactors are planned. The new reactors, like BN-600, will be at Chelyabinsk where a MOX fuel plant is also under construction (Complex 300). This will make that site the world's most important nuclear fuel cycle complex.

Russia increasingly participates in collaborative research programs on nuclear fuel topics such as ADS and recently entered into cooperation on the Energy Amplifier concept (see below) with Italy and Spain. Russia has also considered going into competition with the UK and France for commercial reprocessing business, perhaps even offering facilities for long-term storage of fission products and MA (minor actinides). However, this business has not yet been approved by the Russian government [16].

4.5 Japan

Japan started reprocessing various types of nuclear fuel at its Tokai Reprocessing Plant (TRP) in 1980. TRP has a relatively small capacity (90 tonnes per year). A new 800 tonnes per year plant is under construction at Rokkasho, which will be able to serve all of Japan's reprocessing needs. In the meantime fuel is being sent to Sellafield and La Hague for reprocessing and the plutonium and fission products returned to Japan. So far some ten electric utilities in Japan have had more than 7,000 tonnes of fuel reprocessed abroad. [17]

The fuel is shipped to Europe via the Panama Canal. To date about 160 fuel shipments have been made and one shipment of plutonium has been returned to Japan. MOX fuel is being made for Japan at Sellafield and from now on plutonium will be sent back in MOX form. Fission products are mixed with molten glass in stainless steel cylinders 1.3 m high weighing 400 kg. When they arrive back in Japan they are stored at Rokkasho pending final disposition.

The fuel and the fission product cylinders are sealed into large shipping casks built to withstand any conceivable hazard. Shipping of these materials is done in the specially built ships of Pacific Nuclear Transport Limited owned by BNFL, COGEMA and the Japanese utilities. These ships incorporate specialized safety features such as double hulls and enhanced buoyancy to ensure their survival in extreme weather or collision. There are also extensive but classified security systems and armament. Over 4,000 casks have been shipped in a 20 year period with no security incident or radioactive release.

Research in Japan is concentrated in the OMEGA program for partitioning and transmutation. It was reviewed by government agencies with a report [18] in 2000. This report concluded that the research had made significant progress but that before proceeding further, decisions will be needed on the future plans for Japan's fast breeder reactor program.

4.6 India

Reprocessing in India began in 1964 with the extraction of plutonium from the fuel of the CIRUS research reactor. CIRUS was built by Canada with the heavy water moderator supplied by the US. The reprocessing operation was done at the CIRUS site with the objective of serving as a test bed for the development of a domestic reprocessing capability. Later an early model power reactor of the CANDU type, called RAPP, was also built by Canada in India and this became a prototype for a succession of small domestic CANDU-like reactors. Plutonium derived from CIRUS fuel was used in India's first nuclear weapon detonated in 1974 and since then nuclear cooperation between Canada and India has been limited [19].

From the perspective of its civilian nuclear program, India is a large country with limited energy resources and thus, has a strong incentive to recycle nuclear fuel [20]. India operates two reprocessing plants at Tarapur (Prefre-1) and Kalpakkam (Prefre-2), which reprocess fuel not only from the CANDU-type power reactors but also from research reactors and an experimental fast breeder reactor. These plants are based on the Purex process with variations depending on the fuel type.

While uranium is scarce, thorium is abundant and hence, India is particularly interested in the thorium fuel cycle (Section 2.4), which is an attractive possibility for future fuelling of its CANDU-type reactors. Development of thorium fuels is a major component of India's nuclear power strategy.

The Indian experience is particularly relevant to the technical aspects of the issue of reprocessing in Canada. Although there is no reason to doubt that it would be possible, India has demonstrated that CANDU fuel can be successfully reprocessed on a large scale. Furthermore, if there were to be a scarcity in uranium supply in the long-term, it would seem that India has gone an appreciable distance in developing the technology of the thorium cycle that can readily be used in its CANDU-type reactors.

4.7 United States

In the United States enrichment is necessary for its LWR fuel but, unlike other countries with LWRs, no post-reactor reprocessing is performed and permanent burial of the used fuel in an underground facility is planned [9].

Even though the US had large military reprocessing facilities for plutonium production, no civilian processing has taken place there since 1977. At that time President Carter made it a tenet of US nonproliferation policy that there would be no reprocessing of commercial reactor fuel. He hoped, as it turned out wrongly, this would set an example to the rest of the world. Since then uranium prices have fallen as new uranium reserves were discovered and the expansion of nuclear power in the US has ceased. Therefore, there is no economic or other reason for reprocessing in the US.

Prior to the 1977 ban, the US had two 300 tonne/year plants at West Valley, NY and Morris Ill. The West Valley plant was closed in 1972 because it couldn't meet regulatory requirements. The Morris plant never worked very well and was slated for closure. A 1500 tonne/year facility under construction at Barnwell, SC was never completed. The

1977 decision and the subsequent cancellation of the fast breeder reactor program have resulted in a US strategy for spent fuel mitigation very different from Europe, Japan and other countries. This program will be further discussed below.

4.8 Canada

In the early years of nuclear development Canada, like all other countries with civilian nuclear programs, was concerned about a future shortage of uranium fuel. Experimental reprocessing was done on research reactor fuel to extract plutonium and some recycling was done [21]. A considerable research program on thorium fuel cycles was performed because CANDU reactors are particularly favourable for thorium and thorium is 7,000 times more abundant in the earth's crust than uranium [6].

By the 1970's the emphasis had shifted from uranium conservation to economics that is to optimizing fuel cycles for minimum cost. Research with several of the fuel cycles mentioned in Section 2.4 showed that the SEU (slightly enriched uranium) fuel cycle gave the best value followed by the natural uranium once through cycle. All the other fuel cycles were more expensive. Fuel cycle development continued into the 1980's mainly to demonstrate CANDU's fuelling versatility to prospective buyers not blessed with ample uranium deposits. At present research is concentrated on LEU (low enriched uranium) fuel for the ACR (Advanced CANDU reactor).

Another unique feature of the Canadian fuel cycle program was the concept called electronuclear breeding. An accelerator would propel an intense beam of protons to impinge on a lead-bismuth liquid metal target producing large fluxes of fast neutrons in a process called spallation. These fast neutrons would be used to breed fissionable materials in thorium targets, which would be used to make the thorium fuel cycle fully sustainable. Much research funding was invested in this overly ambitious and, as it turned out, completely unnecessary program, the Intense Neutron Generator, until its cancellation in 1968 [22].

Canada's policy on reprocessing at some point changed to accord with the US policy declared by President Carter in 1977 although it appears no similar high level announcement was made by the Canadian government [23-24]. Canada has never done reprocessing on an industrial scale and there is little or no home grown reprocessing expertise. There is also no pressure from the nuclear utilities to reprocess because the cost of fuel remains only a small percentage of the cost of electricity generated by a CANDU reactor. Furthermore, there is unlikely to be interest in reprocessing for uranium conservation in the world's largest exporter of uranium.

5. Transmutation

Reprocessing, partitioning and conditioning provide means to reduce the volume of nuclear fuel, to segregate its components for individual treatment and to alter its chemical composition for better storage. Transmutation is aimed at destroying the radioactive isotopes by nuclear reactions in order to convert them to stable isotopes.

Reprocessing is now widely used in several countries as reviewed in Part 4 above. In contrast, transmutation of nuclear fuel isotopes is not yet a practical reality but is the subject of research and development programs in many laboratories worldwide.

There are different possibilities for delivering the neutrons necessary for transmutation. Many power reactors with thermal neutrons and a few fast reactors are available at the present time. Other possibilities such as Accelerator Driven Systems (ADS) and the Energy Amplifier may be available in the future and are now being researched. Further in the future could be neutrons from nuclear fusion (see for example [25]) and high energy photons (see for example [26]), which is mentioned here only for completeness.

5.1 Physics of Transmutation

Transmutation encompasses two basic means of destroying radioactive isotopes. The first is conversion of a radioactive isotope to a stable isotope by neutron absorption. For example:

technetium-99 + neutron → technetium-100

technetium-100 → ruthenium-100 + $beta^-$ (17 seconds)

Ruthenium-100 is stable and thus, the long lived radioactive fission product technetium-99 has been transformed to ruthenium and thus, destroyed by neutron absorption.

In order for such a reaction to be practical for transmutation two conditions must apply.

- The probability of the reaction (called the cross section) must be sufficient that the transmutation occurs rapidly enough to be practical
- The isotope in question must be sufficiently separated from other isotopes so that more radioactive isotopes aren't produced under neutron bombardment.

As will be seen below these conditions are not always easy to fulfill.

The other means of destroying isotopes is by inducing them to fission. Attempting to transmute actinides in a thermal reactor would simply yield more radioactive actinides. However, in a fast reactor the probability of actinide fission is similar to that of producing another actinide and thus, the actinides would be consumed. Figure 9 illustrates this situation for neptunium.

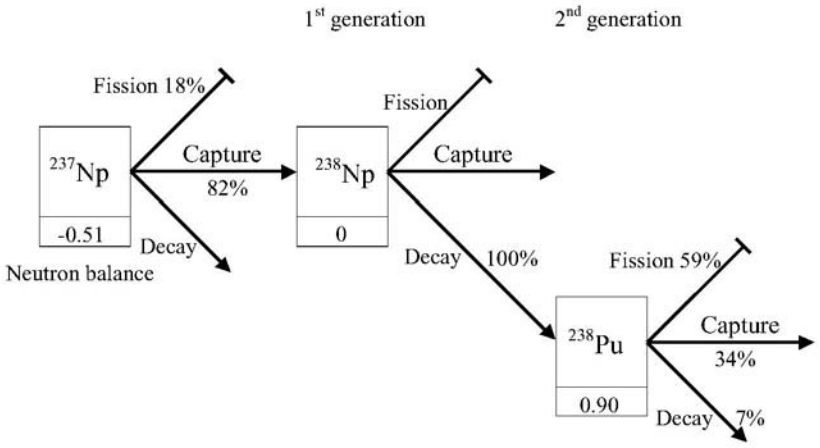

Figure 9 Transmutation Scheme for Neptunium-237 [8] (by permission of the NEA)

In the figure a neptunium-237 nucleus absorbs a fast neutron and 18% of the nuclei fission while 82% transmute to neptunium-238. Essentially all of the neptunium-238 decays to plutonium-238 which with another neutron (2nd generation) fissions in 59% of cases, 34% transmute to plutonium-239 and 7% decay. Thus, in the first two generations 66% of the original neptunium-237 has been destroyed. The neutron balance is a measure of the neutrons used in the process of destroying the neptunium. In fast reactors, the overall neutron balance is positive meaning additional neutrons are available because neptunium fission can be achieved. This would not be the case in thermal reactors since the fissioning of actinides (except uranium and plutonium) is improbable. Note that fission of actinides results in the production of fission products that are essentially the same that result from the fission of uranium and plutonium. The important point is that the long-lived actinides have been replaced by short-lived fission products.

5.2 Transmutation of Fission and Activation Products

Neutron absorption is the only really feasible nuclear reaction for transmuting fission products. The neutrons for example could be delivered by exposing specially designed target assemblies to thermal or fast neutron fluxes. Unfortunately, for some fission products the transmutation reaction rates are small for both thermal and fast neutrons.

Considering the fission products shown in Table 2, strontium-90 and cesium-137 have half-lives of 29 and 30 years respectively and this makes them the major sources of the radioactivity and heat in spent fuel. It turns out that it is impractical to transmute these isotopes because of their relatively low probability for neutron absorption. No source currently available could deliver a sufficiently large neutron flux to make transmutation proceed faster than radioactive decay [27]. However, the radioactivity from both these isotopes will decay to negligible levels after 300 years. Thus, they could be segregated from other fission products and allow to decay in dedicated storage facilities in order to reduce the heat load requirements for long-term storage and burial facilities.

Figure 4 shows that in terms of long-term risk, the isotopes it might be most desirable to destroy by transmutation would be in order of importance: iodine-129, technetium-99, cesium-135, and tin-126. The activation products (Table 4) zirconium-93, carbon-14 and chlorine-36 are also significant hazards. Transmutation is not necessarily the best approach for each of these fission products and, as suggested above for strontium-90 and cesium-137, conditioning, partitioning and segregated confinement may be more efficient and economical.

Iodine-129 is highly mobile, chemically reactive and radiotoxic due to its specificity for the thyroid. Between 95-98 % of the iodine can be removed in Purex type-reprocessing but it would be desirable to have that percentage much higher. As mentioned above, changing iodine's chemical form to AgI or $PbIO_4$ reduces its mobility but would not necessarily prevent its eventual escape to the biosphere because of its long half-life. Transmutation is difficult because the iodine-129 would have to be completely separated from the other isotopes of iodine so neutron absorption would not merely result in more iodine-129. It may also be difficult to make stable targets for irradiation in appropriate neutron fluxes. At this time iodine-129 at most reprocessing plants is just released into the sea on the grounds it has a negligible effect given the very large dilution factor of the natural iodine-127 in sea water.

Technetium-99 is a promising candidate for transmutation. It has a sufficiently large neutron absorption cross section to make transmutation feasible and stable targets could be fabricated from the metallic form. Chemical problems in partitioning technetium mean that not more than 80% can be removed in standard Purex processes. Therefore, the emphasis for this isotope is in developing better pyrochemical separation technologies.

Cesium-135 has a half-life of 2.3 million years compared to the much more radioactive cesium-137 with a half-life of 30 years. It is possible to transmute cesium-135 by neutron absorption to the stable isotope barium-136. However, unless all the stable isotope cesium-133 was removed to very high level of purity more cesium-135 would be produced than destroyed by neutron bombardment.

Zirconium-93 can be transmuted to the stable zirconium-94 but not very efficiently because the relevant reaction rate is small, some five times less than the comparable rate for cesium-135. A highly effective means of separating zirconium isotopes would be needed and if this were to become available then, as suggested in section 2.1, it might be better to manufacture cladding from zirconium containing as little zirconium-92 as possible on the principle that non radioactive materials are easier to work with.

Tin-126 has not only a very low neutron absorption reaction rate but also the same sort of difficulties as technetium in extracting it from fission product mixtures. Therefore, it doesn't appear to be a good prospect for transmutation.

The conclusion of this section is best summarized by a quote from [27]*: "The primary risk of geologic repositories is related to the release of long-lived fission products. With the exception of technetium-99, however, the transmutation of long-lived fission products appears to be difficult because of low neutron reaction cross-sections (rates) and the necessity of isotopic separations. This means that, for most fission products, special conditioning and confinement is the only practical method to reduce the radiological impact."*

5.3 Transmutation of Actinides

Figures 3 and 4 show the long-term radiotoxicity of spent fuel is primarily caused by the presence of actinides with very long half-lives. By far the largest amounts of actinides are the uranium and plutonium isotopes and reprocessing can extract them. In the MA (minor actinides) category neptunium, americium, curium and others make significant contributions to radiotoxicity. Therefore, an important thrust of transmutation research is aimed at destroying the MA, which could reduce by hundreds of thousands of years the time requirements for spent fuel depositories.

Assuming that the uranium isotopes in the fuel have removed by reprocessing, the next question concerns what to do with the plutonium-239. One possibility is to remove it by Purex reprocessing which can be done with 99.9% efficiency. Much of the plutonium so extracted is stored to await the advent of a fast reactor nuclear economy; hundreds of tonnes are now stockpiled at reprocessing plants throughout the world. Alternately it can be used in a MOX fuel cycle by blending with added uranium, a fuel now used in many reactors in Europe and Japan. It turns out that with one MOX recycle, the total radiotoxicity of all the fuel isotopes is reduced by a factor of about three. However, if the plutonium from the irradiated MOX fuel is further recycled then the radiotoxicity summed over the total fuel cycle exceeds that of the original fuel because of the additional actinides and fission products generated.

MOX recycle is an example of a fuel cycle that becomes increasingly negative from a nuclear waste perspective while tending to maximize the total energy extracted from a given quantity of uranium. This illustrates the important point that in order to choose the best nuclear fuel cycle all its inputs and outputs need to be carefully accounted for in a life cycle approach. It is also clear that what is best in terms of the efficient use of nuclear fuels may not necessarily be best in terms of nuclear waste reduction.

Another possibility is to leave the plutonium and MA together as fuel for a fast reactor. This will effectively consume the plutonium and fission many of the minor actinides. However, there is a difficulty in controlling the criticality of a fast reactor fueled by plutonium alone due to the very small reactivity margin provided by the small number of delayed neutrons from decay. The solution to the problem is to add uranium-238 but this just results in the production of more fission products because the rate of uranium-238 fission for fast neutrons is much higher than in a power reactor with thermal neutrons. More plutonium and MA would also come from neutron absorptions in the uranium-238. It turns out that a significant amount of plutonium and MA can be usefully consumed in fast reactors with added uranium-238 in the fuel but there comes a point when no further improvement is possible if the objective is to mitigate the radiotoxicity of fuel components. Therefore, fast reactors are only part of the solution [28].

5.4 Accelerator Driven Systems for Transmutation

Obviously fast reactor transmutation schemes depend on having a fast reactor program as in France, Japan and Russia. However, the fast reactor program of the US was closed down in the 1980's and it is the country that has accumulated the most used nuclear fuel, estimated to be a total of 87,000 tonnes at the end of the lifetimes of the present LWRs. The policy of the US is now the long-term burial of the fuel from the

power reactors at Yucca Mountain, Nevada with no prior reprocessing, partitioning or conditioning.

Researchers in the US national laboratory system, especially those concerned with accelerator development, have promoted an accelerator-based concept for plutonium and MA burning called ATW for Accelerator Transmutation of Waste [29]. This is one of several similar ADS (Accelerator Driven System) ideas now being researched worldwide. In most cases the purpose of the ADS is to perform the final destruction of plutonium and MA after the fast reactor step, in the US it is proposed to put all the plutonium and MA into an ADS.

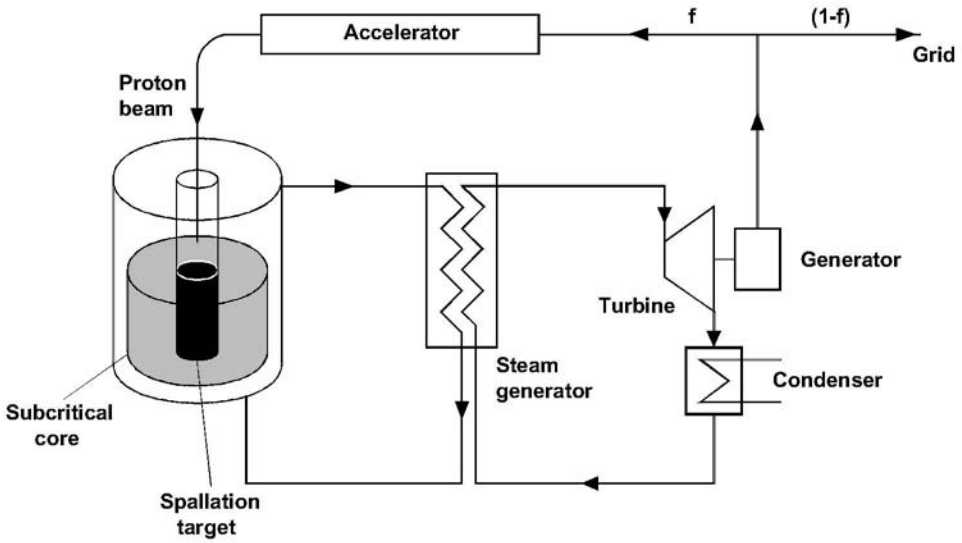

Figure10 Accelerator Driven System (ADS) for transmutation [8] (by permission NEA)

The basic ADS idea is shown in Figure 10. An accelerator is used to produce an intense beam of high energy protons. The beam is directed to a fast reactor core consisting of plutonium and MA, which is subcritical, that is incapable of sustaining a chain reactor on its own. The protons strike a lead-bismuth target which yields copious fast neutrons by spallation. These neutrons bring the core to criticality and the desired fissioning of the plutonium and MA occurs. Heat is taken from the reactor to generate electricity and a fraction of it, f, is used to supply the accelerator with the remainder sent to the grid.

A related concept, invented by Nobel Laureate Carlo Rubbia, is called the Energy Amplifier [30]. It uses a thorium fuel cycle to both generate electricity and to burn plutonium and MA. It has advantages in terms of accelerator requirements, for example, but goes into the not very well-known territory of thorium fuels.

ADS is the subject of several cooperative research programs internationally but such technology is not expected to be available for practical deployment for several more decades. The US ATW program is estimated to require 27 years of research and demonstration followed by 90 years of reprocessing and transmutation to deal with the expected US production of 87,000 tonnes of LWR fuel. The estimated cost would be $280 billion (US 1999$) [29]. An energy bill passed by the US Senate in June 2003

allocates $865 million US to research in areas related to nuclear waste mitigation including ATW [31].

6. Reprocessing Issues

The purpose of this section is to summarize in a conveniently accessible form the technical aspects of the main issues surrounding reprocessing. In presenting these issues care has been taken not to intrude opinions on social, political, and ethical factors on which the ultimate decisions will most likely be based. Nevertheless, it must be acknowledged that scientists and engineers can also have opinions on technical issues and striking a balance is attempted when this is encountered.

6.1 Recycling of Uranium and Plutonium

The once through nuclear fuel cycle, from mine to reactor to burial without reprocessing, is wasteful of nuclear fuel resources. From Table 1, it is clear that more energy could be extracted from the remaining fissionable uranium-235 and plutonium-239 if they were recycled. If the fuel was buried in a long-term geological storage facility then this potential for additional energy would be wasted. This consideration would also be a factor in deciding whether a depository should be designed for fuel retrievability or not. For that reason advocates of recycling decline to use the term “waste” for used or spent reactor fuel and would rather consider it a resource.

At the present time there is no scarcity of low cost uranium due in large measure to the vast and rich uranium ore bodies of northern Saskatchewan. Therefore, there is not sufficient economic incentive even to use the uranium-235 from reprocessing plants since the recycled uranium-235 contains a small amount of other uranium isotopes that make it somewhat less reactive. It’s cheaper just to buy fresh fuel.

Some reprocessed plutonium-239 is now consumed as MOX fuel. Nevertheless, much more is extracted in reprocessing and at Sellafield, for example, between 50-100 tonnes of plutonium are stockpiled. In addition, the current trend to nuclear disarmament is freeing up for civilian use significant quantities of plutonium and uranium-235 from dismantled nuclear weapons. The fast reactor stage of nuclear development, expected to consume the plutonium, has not come to pass. Hence, there is little or no demand for plutonium.

A uranium shortage was much feared in the 1950’s and 1960’s when the first nuclear power plants were being deployed. While the global total of nuclear power plants has grown to well over 400 worldwide, growth has been slow in the last two decades. Nuclear fission technology has not succeeded in dominating electricity production to the extent expected by its original proponents. Moreover, large uranium discoveries have been made in recent years. There is now enough uranium in current known reserves to last for 50 to 100 years at the present rate of consumption [9].

A future resurgence of nuclear fission power, perhaps to combat climate change or for hydrogen production could change this picture but at this time uranium conservation is not a compelling reason to reprocess nuclear fuel. According to their various advocates, renewable energy or fusion power may come on stream before all the uranium is

exhausted at which point fission fuel requirements would no longer be relevant.

6.2 Reduction of Volume and Radiotoxicity

There is no doubt that reprocessing spent fuel can greatly reduce the volume of the radioactive material ultimately to be buried in a repository. For CANDU fuel removal of the uranium-238 and the cladding and separating out the uranium-238 and plutonium-239 would leave only a small percentage (~0.8 %) of fission products including MA. Almost the entire radioactivity in the fuel is concentrated in this component.

Another advantage of reprocessing is that it provides opportunities to condition certain isotopes in the fuel. Their chemistry can be changed to lower mobility, and hence the net potential for radiotoxicity in geological storage. Nevertheless, whatever reprocessing is done there will still be a need for long-term geological storage of various radioactive isotopes. In considerations of volume reduction, the volume of low and intermediate wastes generated in reprocessing (including those from decommissioning of the reprocessing plant) must also be taken into account. If the fission products are to embedded in glass the volume of the final waste form that is the blocks instead of the fission products themselves needs to be considered. As a final thought, if a long term geological storage facility were to be built, how significant would volume reduction be in reducing the overall cost of the facility?

As explained in sections 2.1 and 2.3, CANDU reactors using natural uranium produce at least (depending on LWR fuel enrichment) four times more spent fuel per unit energy produced as LWRs. Therefore, reprocessing to reduce spent fuel volume would be particularly appropriate for the CANDU fuel cycle. However, at the 1993 (maximum) Canadian level of nuclear electricity production quoted in section 2.1, over 2,000 tonnes of uranium in spent CANDU fuel result each year. Note that in Table 6 the world capacity for LWR (uranium oxide) fuel reprocessing is only about 3,000 tonnes. Thus, reprocessing of domestic CANDU spent fuel would require new and large facilities.

6.3 Destruction of Radioactive Isotopes

Reprocessing including the partitioning and conditioning it involves, does not get rid of any radioactive material. Many separations into various components for recycling and storage in addition to chemical changes can be usefully made. However, except for the decreases due to radioactive decay which occur independently of reprocessing, the same quantity of radioactive materials is present. No radioactive isotopes are destroyed in reprocessing itself. Hence, those with concerns about the ultimate disposition of used nuclear fuel prefer to use the terms “long-term burial” and “geological storage” instead of “disposal”. This is technically correct in the sense that the radioactive isotopes are simply being stored, albeit for very long times in geological formations, until their radioactivity decays away.

Transmutation does offer the prospect of truly disposing of radioactive isotopes. It requires the development of much more efficient partitioning systems otherwise neutron bombardment of impurities may produce additional unwanted radioactive isotopes.

Another problem is delivering enough neutrons of the appropriate energy to perform the needed transmutations. Neutrons from thermal power reactors would simply lead to more MA because they are not capable of fissioning them. Fast reactors, of which there are only a few still operating, can perform the necessary fissions but only up to a point where reactor criticality and stability control become problems. Accelerator Driven Systems are necessary to complete the desired transmutations. These systems are still in the conceptual stage and may not be available until well into the present century and thus, comprise another energy eventuality, a category already mentioned above.

6.4 Economics

If needed, the technology and experience for reprocessing CANDU fuel could be acquired from India. Firms experienced in building and operating reprocessing facilities, BNFL and COGEMA would probably be eager to construct a reprocessing facility in Canada provided it didn't compete with their international business. Therefore, if it were considered desirable or necessary, a domestic reprocessing plant would be feasible. It would be a rather expensive undertaking. Duplicating in Canada a reprocessing facility similar to THORP at Sellafield or U3 at La Hague would likely cost in the order of at least $5 billion and perhaps, much more. An alternative approach would be to send used CANDU fuel to the UK, France or another country for commercial reprocessing at their facilities which no doubt would also be expensive and probably impractical due to the large annual tonnage of CANDU spent fuel.

It seems at present the only grounds for considering reprocessing in the Canadian context would be to improve the overall situation for long-term storage of nuclear fuel. Calculating the costs and benefits would require an in depth analysis of factors such as the cost of reprocessing versus the benefits of a smaller depository. The likely high costs of transmutation, if and when it becomes commercially feasible, would have to be weighed against the presumably lower cost of burying the fuel for the long-term in an appropriate geological depository. Engineering cost estimates would be a factor in decisions on which option to pursue but social, political and economic considerations would be likely be decisive.

6.5 Weapons Proliferation

Both the major isotopes involved in reprocessing, plutonium-239 and uranium-235, are fissionable isotopes and therefore, are potential materials for nuclear weapons. In fact, the world's first nuclear weapons used by the United States against Japan consisted of a uranium-235 bomb ("Thin Man") and a plutonium-239 bomb ("Fat Boy"). The uranium-235 was separated from uranium-238 at the world's first enrichment plant. The plutonium was produced by reprocessing natural uranium fuel that was irradiated in heavy water reactors built solely for that purpose.

These same two routes, an enrichment plant or reprocessing the fuel from a heavy water (or graphite) moderated reactor fuelled by natural uranium, are still the main means that can be used to acquire nuclear weapons materials today. Therefore, any state could in principle with time and sufficient funding produce its own weapons materials. The main factor that prevents this is a system of international treaties with the probability of sanctions should states not fulfil their obligations. The International Atomic Energy

Agency (IAEA), a United Nations agency, operates an inspection service, which monitors states who are parties to the Non-Proliferation Treaty (NPT) to ensure they are following the treaty's intention of not diverting nuclear materials to weapons. It should be stressed that these measures are voluntary: *"Cooperation between the IAEA and a State is necessary for the successful implementation of safe guards in any context."* [32]

There are those who argue that the best way to safeguard the plutonium in nuclear fuel is to leave it there rather than to extract it because the intense radioactivity of the fuel would discourage any tampering. Others argue that the plutonium is best consumed in MOX fuel or burned in fast reactors and thus, permanently removing it from potential diversion. In either case the security of the nuclear materials must be very high. Given the current status of nuclear development in the world, it would seem improbable that additional reprocessing for the remediation of used nuclear fuel would add to the risks of nuclear proliferation.

6.6 Environmental Issues

The environmental issues of reprocessing are closely related to the economic issues. It would seem that minimizing the environmental impact over the entire nuclear fuel cycle would be a more plausible optimisation criterion instead of minimizing its total cost. The complicated fuel cycles aimed at consuming most of the radioactive isotopes arising from spent fuel, of the type being studied in particularly Europe, involve more than one reactor and multiple reprocessing steps. They need to be examined very carefully, particularly in terms of the resulting reprocessing wastes. Comparisons need to be made with the environmental impacts of long-term geological storage to ensure that the radiotoxicity and net radioactive burden to the biosphere were really decreased by reprocessing.

There are those who argue that opening a fuel bundle for reprocessing is equivalent to opening the mythical Pandora's Box because the radioactive isotopes are no longer confined in the fuel but released to do mischief. In this view it would be best to leave them sealed and bury them intact. The issue is whether reprocessing can be done without severe environmental impacts.

During the early stages of the Cold War in the 1950's and 1960's, an intense nuclear arms race gave rise to very large scale processing of nuclear fuel to extract plutonium. This fuel was irradiated in purpose-built reactors like Windscale-1, section 4.2. Virtually no attention was paid to the environment in what many on both sides perceived to be a contest for survival. The outcome was very large quantities of poorly stored and badly characterized reprocessing wastes needing costly remediation in locations like Hanford in the US and Chelyabinsk in Russia.

Therefore, the issue is whether reprocessing is fundamentally an environmentally risky business or whether it was just done badly in the Cold War period but now is sustainable in a civilian context. The Sellafield site has seen both military and civilian reprocessing in addition to a reactor accident and electricity production from Calder Hall. Sellafield is continually criticized in the UK press for environmental problems although it is difficult to judge the validity of these accusations. La Hague pursues a rather more open approach and regularly publishes the occupational exposure of its staff and analyses of seafood, local produce and other biosphere radiation measurements. Figure 11 shows a plot over

time of staff radiation exposure in UP2 and UP3. It shows a rapid decline even as the tonnage of fuel reprocessed increases [14]. La Hague's record of minimal environmental impact, at least in the short term, indicates that civilian nuclear fuel reprocessing can be done in an environmentally responsible manner.

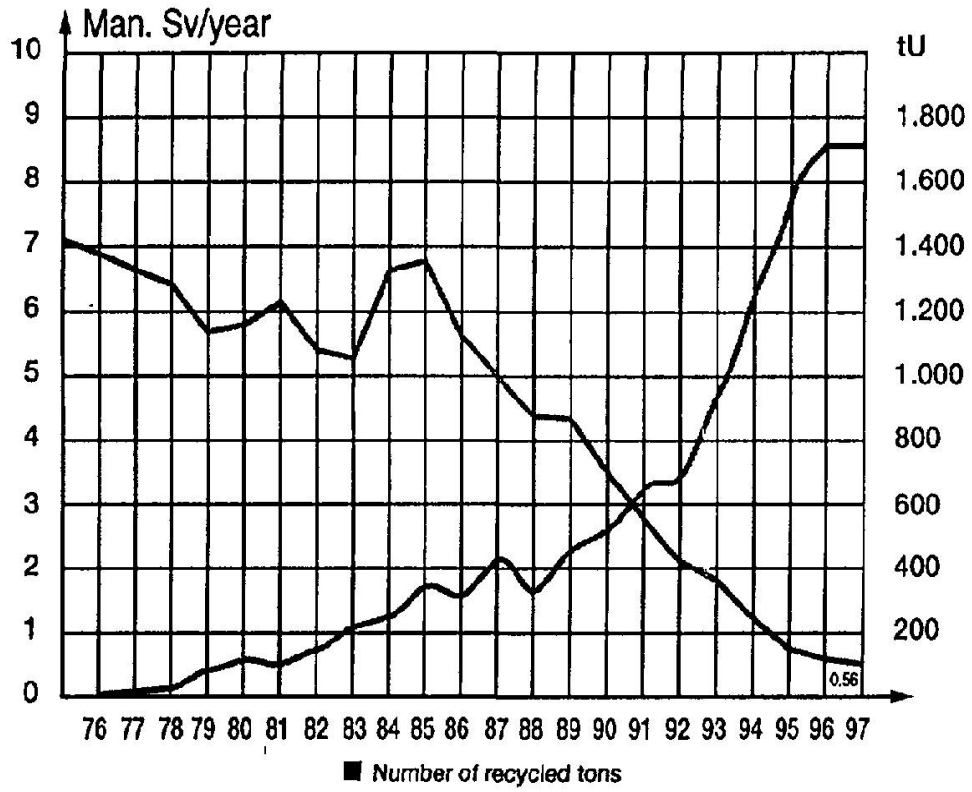

Figure 11 Total occupational exposure at La Hague plants UP2 and UP3 compared to tonnage reprocessed [14] (by permission IAEA).

6.7 Transportation

Reprocessing requires the transportation of nuclear fuel from the reactor sites to the reprocessing plants and the subsequent transportation of the products (plutonium, uranium and fission products) in the case of commercial plants, back to their countries of origin. This is now done routinely throughout the world using massive containers to shield persons from the high levels of radiation from their contents. These containers are specially engineered to withstand the worst rail and road accidents that can be hypothesized. Severe tests such as long exposure to high temperature fires and dropping from heights onto an unyielding surface, and onto hardened steel pins, must be passed before the containers are approved for use. Spectacular tests such as train crashes have also been carried out [33-34].

Security in the transportation of materials such as plutonium is also stringent. Certain government agencies specialize in securing such shipments and Japan, for example, has built armed vessels especially for the transport of materials that might be used in

weapons. The record on transportation has been excellent in terms of both radiation safety and security. Evidently transportation to and from reprocessing facilities can be done safely and the primary issue is likely to be the cost of specialized transportation facilities and personnel rather than technical feasibility.

6.8 Reprocessing Safety and Criticality Accidents

The early days of reprocessing saw many accidents within the plants [7]. The organic solvents used must be carefully handled and fires can occur even in operations such as encasing low level radioactive wastes in bitumen as happened in 1995 at TRP [17]. It was found that as the technology matured that the key to preventing these incidents is a continuous emphasis on safety culture among the employees combined with a high degree of quality control. The consequences of such accidents are mitigated by the design of modern reprocessing plants, which retain and contain any radioactive materials released within thick-walled vault structures.

A particularly damaging reprocessing accident occurred in 1957 at Kyshtym some 80 km from Chelyabinsk. Kyshtym was a waste area associated with the military plutonium production operation at Chelyabinsk. A powerful chemical explosion occurred in a concrete waste storage tank containing 80 tonnes of fission products and MA. Failure of the tank's cooling system caused a mostly solidified mixture of sodium acetate and sodium nitrate to ignite. About 20 million curies of radioactivity were released contaminating an area of 15,000 square kilometres. For comparison, the Chernobyl reactor accident released about 50 millions curies. This Soviet era accident was not officially reported to the IAEA for some 30 years. [35]

There is another type of accident unique to nuclear facilities known as a criticality accident. This occurs when a mixture of nuclear materials and usually water is put together in a confined space accidentally creating the conditions for a chain reaction. There have been more than 30 such accidents since the beginning of nuclear processing and a particularly severe one took place at Tokai, Japan in 1999. A company known as JCO was preparing enriched uranium fuel and instead of using the carefully designed system of pumps and vessels put in place to avoid criticality, the workers routinely added uranium solutions to a vat with buckets. On the occasion in question this practice resulted in a chain reaction, which went on for some 20 hours. Two workers died from the radiation sickness and another one was badly injured. Since this particular building did not have a containment system residents of the surrounding area were exposed to levels of radiation that were later assessed not to be harmful. The causes of this easily avoided accident were incompetence and stupidity [36].

The Tokai experience illustrates the reality that even in an advanced industrial country no amount of safety culture or regulatory supervision can completely eliminate the possibility of accidents. Hence, nuclear facilities such as reprocessing plants must be built to contain and limit any accidental releases of radioactivity within the plant itself. The technology to do this is now widely available and, if deployed, safety although it must always be an important concern would probably not form a technical obstacle to building additional reprocessing plants.

6.9 Recovery of Valuable Isotopes and Elements

Canada currently supplies a large percentage of the world's radioisotopes for medical and industrial purposes. Some, notably technetium-99m (the "m" denotes a metastable isotope), are fission products. At Chalk River highly enriched uranium targets are irradiated in a reactor and the technetium extracted [1]. Uranium quantities of few tens of grams are typical of this process compared to the 100 tonne scale of commercial reprocessing.

It is technically possible to extract such useful radioisotopes from spent fuel but it appears that the market for them is limited to relatively small quantities already supplied by other means [4]. Similarly, precious metals and rare earth elements could also be extracted but before they could be marketed for industrial or commercial use, any radioactive isotopes present would have to be eliminated to a very high degree. A careful study would be required to determine whether the costs of this radioactive purification would price these materials out of the market.

While there may be opportunities to derive useful and valuable by-products from reprocessing, the revenue they would bring in would be unlikely to have any significant impact on the costs of, or justification for, a reprocessing plant.

7. Conclusion

Many countries have long-term nuclear power strategies based on the future need for the conservation of uranium resources and the deployment of fast reactors. Recycling of fuel is an integral part of these strategies and in many cases the domestic reprocessing capability is based on the prior development of related expertise for weapons purposes. Reprocessing of civilian nuclear fuel is done commercially on a large scale. Much the same technology, the Purex process, is used in all of the world's reprocessing plants.

Reprocessing enables the partitioning, or separation, of the spent fuel into its components: uranium, plutonium, minor actinides (MA) and fission products. The plutonium can be recycled for consumption as mixed oxide (MOX) fuel in conventional power or fast reactor fuel. However, the usage of plutonium for these purposes is still small and today most of it is stockpiled at the reprocessing plants. Fissionable uranium taken from the fuel can also be recycled as reactor fuel but, because of the low cost of uranium, is only done in Russia.

The fission products and the MA are now embedded in molten glass for long-term burial in a geological facility and some are stored in highly active liquid waste tanks and thus, reprocessing must include treatment of the resulting low and intermediate wastes. The chemical form of some isotopes can be altered to decrease their mobility and radiotoxicity in conditioning processes. Hence, although reprocessing can reduce the volume and improve the form of these wastes prior to geological storage, a repository is still needed.

Transmutation is aimed at destroying the radioactive isotopes, primarily the long-lived fission products and the MA, by neutron bombardment. It requires research on improved partitioning and the development of subcritical accelerator driven systems. Even if today's research programs are successful, it will be many decades before these

technologies can be deployed for practical purposes. Thus, transmutation offers the potential for the ultimate mitigation of fuel waste but only in the long-term.

Up to now, Canadian reactors have exclusively used a once through fuel cycle in which the fuel is removed from the reactor and stored at the reactor sites pending decisions on a long-term option for its disposition. Many more advanced fuel cycles are available for CANDU reactors that are primarily aimed at the optimum use of uranium. Several of these fuel cycles have a requirement for reprocessing. If in the future there was a decision to reprocess CANDU fuel either for reasons of uranium conservation, or more likely to reduce the volume and radiotoxicity of the fuel, a brief survey of the current status indicates there would be no purely technical obstacle to domestic reprocessing.

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