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NWMO BACKGROUND PAPERS
6. TECHNICAL METHODS

6-14 IMPLICATIONS OF REPROCESSING, PARTITIONING AND TRANSMUTATION ON LONG-TERM MANAGEMENT OF USED NUCLEAR FUEL IN CANADA

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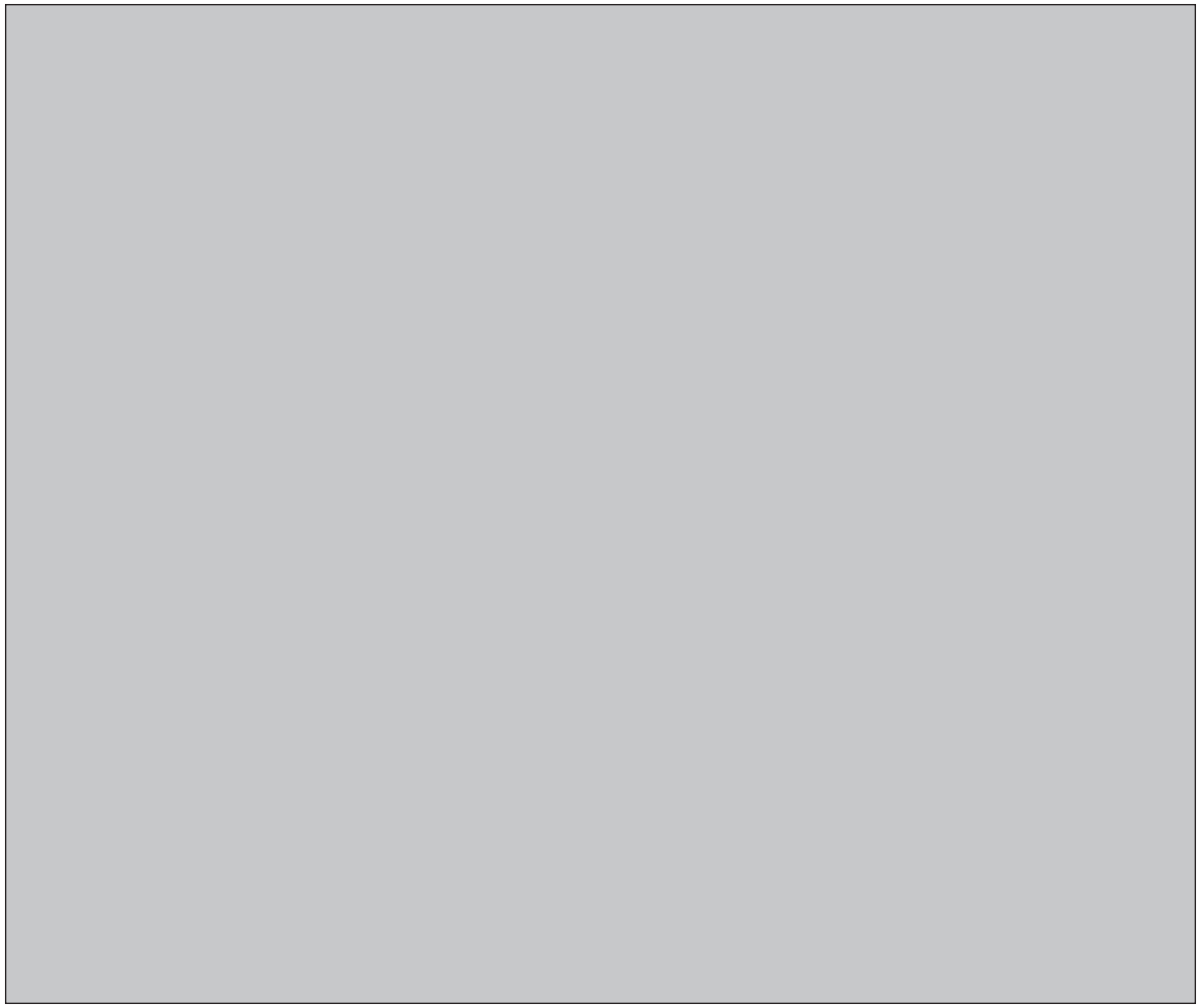

NWMO Background Papers

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 economic and radiological implications of reprocessing, partitioning and transmutation are investigated in the context of the long-term management of used nuclear fuel in Canada. The low burnup of CANDU fuel makes it a substantially less attractive candidate for reprocessing than enriched LWR fuel. On the basis of the best estimates available in the literature, it is concluded that reprocessing used CANDU fuel would add a significant increase to the cost of radioactive waste management in Canada. The occupational and radiation doses to both workers and the public under normal operating conditions would be well within Canadian regulatory limits if the same performance as current commercial reprocessing plants was achieved. Detailed safety and environmental impact assessments would be required. A deep geologic repository would still be needed for the residual high-level radioactive waste if reprocessing of used fuel were to be implemented. The indications are that reprocessing could reduce the size of the deep repository, reduce the radiological hazards of high-level waste, and reduce the length of time that the wastes must be isolated from humans and the natural environment before the waste decays to radiation levels similar to natural uranium ore bodies. Transmutation, now in the research phase, has the potential to completely eliminate some fission products and long-lived minor actinides thereby rendering them harmless. Well-funded research and development programs including experimental accelerator driven transmutation facilities are underway in the Europe, Japan and the US, China, Russia, South Korea and other countries. These programs are being monitored by NWMO to keep abreast of current developments.

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1. Introduction

This report is the sequel to a previous background paper [1] that surveyed the basic technology of the reprocessing, partitioning and transmutation of used nuclear fuel.[1]

The NWMO's position on reprocessing was given in its Discussion Document 2 namely that "...Canada should monitor research on the potential for reprocessing, partitioning and transmutation as possible future options for Canada, although they are not deemed to be practical today…" and "…Canada should maintain a watching brief on research and technology advancements…" [3].

This report further elaborates on some of the potential implications of reprocessing, partitioning and transmutation (RP&T) of used nuclear fuel in Canada and reports some recent progress in the field.

Reprocessing refers to processes whereby the fuel after irradiation in a reactor is chemically treated to separate, or **partition**, its constituent elements for further processing or long-term management. For example, used fuel includes uranium and plutonium isotopes that can be used to make additional nuclear reactor fuel, a process known as **recycling**. There are also fission products (FP) and MA (minor actinides) that contribute most of the radioactivity given off by the used fuel. The FP's are mostly responsible for the intense radioactivity and heat generated by the used fuel in the first few decades after its removal from the reactor whereas the MA's produce less intense, but very long-lived, radioactivity.

Transmutation refers to transforming the fission products and particularly the MA's into non-radioactive isotopes by exposing them to neutrons or possibly other particles. Therefore, transmutation offers the long-term prospect of fundamentally transforming the nature of radioactive waste to render it harmless in the best case. Although, as will be discussed below, there is much research and development (R&D) on Partitioning and Transmutation (P&T) being conducted in many countries throughout the world (but not Canada), it is not yet clear whether the ultimate goal of significantly reducing the toxicity of used nuclear fuel can be achieved.

The first report [1] provided the overall background to reprocessing by considering in a general way the question:

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

The purpose of the present report is to take a closer look at two key implications of reprocessing in the Canadian context:

[1] Readers requiring additional background information on the subject are advised to read the first paper [1] before this one and, if unfamiliar with nuclear technology and its language, to read an introductory explanation in layperson's terms such as [2].

- What would be the cost implications of reprocessing on long-term management of used nuclear fuel?

- Would reprocessing provide a net benefit in terms of the radioactivity to be dealt with?

It's consideration of these issues that in part, will determine long term decisions on reprocessing that may not be made for decades. Even if Canada ultimately decided to embark on a reprocessing program, the time scale for its possible implementation would be many decades into the future, the same sort of time scale as for the implementation of a long-term management approach such as a deep geologic repository. Therefore, the issue in the short term is not whether to implement reprocessing but rather whether provision should be included in Canada's used fuel management strategy to allow for the possibility of reprocessing should it become attractive in the future.

One area where such a provision might be involved is in the question of the retrievability of used fuel. The status of P&T some 50 years or more from now, when a geologic repository might be in operation would have a bearing on deciding whether the used fuel in the facility should be emplaced so that it was easily retrievable. The latter would allow the fuel to undergo beneficial P&T processes at a later time when more advanced technology was available.

There are two main reasons for reprocessing used fuel from commercial power reactors.

- recycling to extract fissionable materials to use again as fuel in order to produce more energy from an initial quantity of uranium; and

- partitioning and transmutation of some radionuclides in used fuel to mitigate or even in some cases eliminate waste management problems.

As discussed in [1], reprocessing was the first method proposed to treat used nuclear fuel. Fuel recycling programs were launched in the 1960's and 1970's when it was assumed that supplies of uranium were limited in view of what was seen at the time as a coming great expansion of nuclear power. However, the growth of nuclear power slowed significantly and large new uranium discoveries with consequent low prices eliminated the need for recycling. Currently much of the R&D in the field of P&T in Europe and North America is directed at the possibilities for the improvement of waste management which is the interest of the NWMO. Very recently recycling has experienced a slight resurgence of interest in North America because of an expectation of new reactor construction on the part of the nuclear industry.

Finally, it should be recognized that some of the issues concerning reprocessing will depend on the future form of the nuclear industry in Canada. Reprocessing done as part of a management approach for a defunct commercial nuclear power industry would be a very different proposition than reprocessing as one stage of the fuel cycle of a fleet of nuclear power reactors continually producing used fuel long into the future.

2. Partitioning CANDU Fuel

Characteristics of Used CANDU Fuel

In order to facilitate the discussions of this paper it is necessary to consider an average or "standard" CANDU fuel bundle. There are several types of CANDU fuel bundles and even within these types each fuel bundle will show slight individual variations. In conformity with [1] the standard CANDU fuel bundle used in the AECL Environmental Impact Statement [4] will be used.

This standard bundle is based on a 37-element Bruce reactor bundle. The total mass of the bundle is about 24 kg of which 19 kg is uranium and 5 kg is the zirconium alloy structure in which the uranium fuel pellets are encased.

It is assumed to have produced 685 GJ per kg U or (7928 MW days per tonne of U) by the time it is removed from the reactor[2]. This energy production (burnup) converts to 63,348 kWh (kilowatt hour) per kg U in the units used by electrical utilities. In terms of energy per bundle the 19 kg would result in 1.204 million kWh per bundle.[3]

The bundle is stored for 10 years after its removal from the reactor and at this point the composition of the bundle is as shown in Table 1 which has been modified slightly from the one given in [1].

Table 1 Composition of fresh and used CANDU fuel as a percentage of the mass of uranium [4]

Component	Fresh	Used
Uranium-235	0.72 %	0.20 %
Uranium-238	99.28 %	98.50 %
Plutonium-239	-	0.28 %
Minor Actinides (MA)	-	0.22 %
Fission Products (FP)	-	0.09 %
Stable Isotopes		0.71 %

Some comments on used or used fuel as made in [1] are worth emphasizing here.

- Uranium-238 from the fresh fuel comprises 98.50% of the used fuel and is the main constituent of natural uranium ore as mined.

- Uranium-235 and plutonium-239 are fissionable and can be recycled in new reactor fuel.

- The Minor Actinides are long-lived radioisotopes (americium, neptunium, curium and others) produced in the fuel by exposure to neutrons in the reactor and comprise the main source of radioactivity in the fuel after the first few hundred years out of the reactor.

[2] Note: This burnup value is slightly less than the reference burnup used in the conceptual design reports prepared for the Joint Waste Owners in Canada.

[3] These are thermal values and the electricity produced is the thermal value times the thermal-electric conversion efficiency, nominally 0.33 for CANDU reactors and the number of full power days.

- The Fission Products, as the name indicates, are radioisotopes resulting from the fission of uranium and plutonium in the reactor; they produce intense radioactivity and heat in the first few hundred years. Other fission products are long lived and contribute to the total radioactivity of used fuel for hundreds of thousands of years.

Reprocessing is a general term for operations on used fuel and partitioning is a more specific term meaning the chemical separation of used fuel into the components shown in Table 1. Partitioning is routinely done in several commercial plants throughout the world and it is estimated that about 80,000 tons of used fuel has been reprocessed to date.

Reprocessing has the potential to significantly reduce the volume of the high-level radioactive waste material for long-term management. Looking at Table 1, the first three rows comprise about 99% of the used fuel mass, i.e. without the cladding used. The uranium-238 could be a candidate for a near-surface waste management facility since it is much less radioactive than the ore from which it was extracted; the challenge is to ensure that the uranium-238 resulting from reprocessing is sufficiently purified of traces of fission products and actinides. The uranium-235 and plutonium-239 could be consumed today as fuel in a reactor.

Transmutation is the process of using neutrons to convert radioactive elements to stable or non-radioactive elements. One promising method of dealing with the MA's would be to transmute them in an Accelerator Driven System (ADS) which consists of a reactor that requires neutrons produced from an accelerator to operate.[4] Another method of disposing of them would be to consume (burn) them as part of the fuel in so-called fast reactors. In both cases the radioisotopes are permanently destroyed.

The approach used at reprocessing plants for most fission products is to store them in glass compounds in order to let them naturally decay since many are intensely radioactive and are essentially gone in relatively short times. For a few long-lived fission products conditioning (changing the chemical form of the material to one better suited to long-term storage) or transmutation schemes may be advantageous.

ADS systems, improving partitioning methods, and developing efficient fuel cycles that consume MA's are among the many topics in this field that are the subject of international R&D programs.

Most of the reactors now producing electricity in the world are light water reactors (LWR) which use uranium enriched in the fissionable component uranium-235 (3 to 4.5%) and therefore, the characteristics of the used fuel are somewhat different from that of CANDU fuel. For example in LWR fuel there is more uranium-235 to be recovered since there was more to start with. The shape of LWR fuel is rods a few metres long whereas CANDU fuel is formed in bundles about half a metre in length. Probably the most important difference is the burnup or energy production. In LWR's the burnup is typically 43,000 to 53,000 MW days per tonne of U compared to the CANDU value of 7,928 MW days per tonne of U, a factor of five or more greater due to the enrichment of the LWR fuel. In other words to produce the same amount of energy that one kilogram of LWR

[4] ADS systems are discussed in the first paper [1] to which the reader is referred for more detail.

fuel needs five or more kilograms of CANDU fuel. The effect of this difference will be felt in costing scenarios based on energy cost per kilogram of reactor fuel. In both reactor types 33% efficiency in converting thermal energy to electricity can reasonably be assumed.

Partitioning CANDU Fuel

A partitioning scheme for CANDU fuel is shown in Table 2.

Table 2 Partitioning of Used CANDU fuel

Component	Potential Disposition	Comments
Uranium-238	• Near-surface waste management facility a possibility if sufficient removal of MA's	•Now stored at reprocessing plants •A long lived isotope of uranium which is less radioactive than mined natural uranium •By far the largest component of used fuel by mass •Can be fissioned in fast reactors
Uranium-235	•Recycling as reactor fuel	•Primary fissionable material in thermal nuclear reactors •Uranium from reprocessing plants now not recycled because fresh uranium is cheaper
Plutonium-239	•Recycling as reactor fuel	•Some fuel cycle concepts leave Pu-239 with the MA's for non-proliferation reasons •MOX (mixed - uranium and plutonium oxide) fuel containing recycled plutonium-239 is burned in about 20 European reactors •Plutonium-239 fuels are also suitable for fast reactors •Large quantities of plutonium now stored at reprocessing facilities have been identified as a concern but build up of americium isotopes from other plutonium isotopes makes it unattractive as fuel or weapons material after 15 to 20 years.
Minor Actinides (MA)	•Transmutation in ADS's •Burning in fast reactors •Geologic disposal	•Long lived MA's are the main reason that geologic repositories designed for 10's of thousands of years still emit radiation •Most significant MA's : -Plutonium-240 (6,600 yr) [5] -Plutonium-242 (360,000 yr) -Neptunium-237 (2,100,000 yr) -Americium-241 (460 yr) -Americium-243 (8,000 yr) -Curium-244 (18 yr)
Fission Products (FP)	•Geologic disposal incorporated in glass (vitrification) •Some FP's may be destroyed by transmutation	•Major Heat Producing FP's : -Strontium-90 (29 yr) -Cesium-137 (30 yr) can be separated from other FP's to simply the thermal engineering of depositories •Transmutation of technicium-99 (213,000 yr) looks to be feasible. •The long lived FP's (LLFP): -Iodine-129 (1,570,000 yr) -Carbon-14 (5730 yr) are the most significant FP's for geologic repositories and their feasibility for transmutation is an important research topic.
Fuel Cladding	•Geologic disposal	•These are the "hulls' consisting of the zirconium alloy structure of the fuel bundles (fuel elements, end plates, caps) after the uranium oxide fuel pellets are removed in reprocessing. •The hulls may be mechanically compacted to reduce their volume and placed in containers and/or concrete. •Most of the radioactivity is from zirconium-93 (1,500,000 yr)

[5] Half life for radioactive decay i.e. the time it takes for the radioactivity of a decaying radioisotope to become half its original value.

Informal discussions with engineers experienced in the field indicate that the reprocessing of CANDU fuel with current technology would be completely feasible. Somewhat different mechanical arrangements for opening the bundles, as compared to opening the fuel rods of LWR's would be needed but otherwise the processes involved would be very similar. Further evidence of technical feasibility is that other countries, for example India, routinely reprocess some hundreds of tonnes of CANDU type-fuel each year.

In terms of used fuel management, at least four times as much CANDU fuel per unit of energy generated compared to LWR fuel would have to be treated in any long-term management scheme. Another way to look at this is that some of the uranium-238 present in natural uranium fuel has in effect been extracted at the facilities used to enrich the LWR fuel in uranium-235. In the CANDU system all of the uranium-238 present in the original natural uranium goes through the reactor and would have to be purified as described above to permit its eventual disposal in a near-surface waste management facility. In contrast, enrichment does not involve radioactive processes and hence, the uranium-238 left at the enrichment plants is immediately suitable for relatively easy disposal. In fact calculations show that the energy generated from a given quantity of natural uranium is somewhat greater for a CANDU reactor than for a LWR, mainly because of the efficiency of enrichment processes. However, as shown, the long-term management of CANDU fuel involves a much greater mass of used fuel to be treated per unit of energy generated.

Major Uranium and Plutonium Isotopes

As discussed in [1], the technology for extracting the uranium and plutonium components is applied routinely in existing reprocessing plants throughout the world. Some of the extracted plutonium is burned in MOX (mixed oxide fuel) but now most of it is stored at the plants. Because the price of fresh uranium is so low the uranium-235 is also mostly stored (except in Russia). The uranium-238 component has lower radioactivity than natural uranium ore since about two thirds of the more radioactive isotope, uranium-235, has been burned in the reactor. Therefore, it is a potential candidate for a near-surface waste management facility provided it can be purified to obtain an acceptable level of radioactive impurities.

Removal of these components reduces the mass of the high level waste contained in the fuel pellets by a factor of about 100. Note that none of the radioactivity has disappeared but the radioactive isotopes have merely been redistributed. Some would argue that this volume reduction is of little value because building a smaller geologic repository would not be a large saving. However, concentrating the most radioactive components in smaller volumes may facilitate their treatment with a variety of specialized methods which would not be feasible for large volumes of material.

Fission Products

As noted above, most of the fission products in the used fuel are intensely radioactive and decay to harmless elements a few hundred years after the fuel is removed from the reactor. Because they dissipate naturally and quickly on their own, their disposition is not considered to be a difficulty for geologic repositories nor for reprocessing plants. In reprocessing plants the fission products are mixed with molten glass and the solidified product placed in containers similar to those that might be used in a geologic repository.

Some FP's present special problems, for example the generation of decay heat from some short lived FP's such strontium-90 and cesium-137 affects the design of geologic repositories. If these intensely radioactive but short lived FP's were separated out and used or disposed of elsewhere then significantly more high level waste could be emplaced in the same repository space (see the discussion of Yucca Mountain in section 6 below).

There are some LLFP's (long lived fission products) that require special consideration for their long-term management. These LLFP's include iodine-129, carbon-14[6] and technetium-99. The latter can already be chemically separated from high level waste reprocessing liquids to the level of 80% and it is a good candidate for transmutation. Iodine-129 is a much more difficult problem. At the moment at least 95% of it can be extracted and it is a possibility for transmutation although it has a low neutron capture cross section and appropriate targets would have to be fabricated. Existing reprocessing plants merely release it to the atmosphere or to the oceans in which large dilution with naturally occurring iodine takes place.

Minor Actinides

Some of the minor actinides are significant emitters of radiation for tens of thousands of years and therefore, the main reason for the very long time scales required of geologic repositories. They are called "minor" to distinguish them from the major uranium and plutonium isotopes discussed above; the main MA's are neptunium, americium, curium and some of the less common plutonium isotopes.

From the perspective of waste management, it would be desirable to have a means of destroying actinides. There are two main approaches contemplated at present. The first involves recycling them as fuel in fast (not moderated) reactors. This method clearly requires fast reactors of which there are very few in the world (three operating, two under construction) compared to about 440 commercial nuclear power plants. In the first few decades of nuclear energy a large number of fast reactors were projected which would produce (breed) fuel for LWR's. This didn't materialize but some programs envisage fast reactors for actinide burning. Doing this in today's thermal (moderated) reactors is possible for one cycle of MOX fuel which reduces the actinides by a factor of three but any further recycling would just increase the amount of minor actinides.

Since the construction of fast reactors is a very large and expensive undertaking, the second possibility for MA destruction is a special accelerator-reactor combination dedicated to actinide burning. This system consists of an accelerator beam of protons that is steered into a target (usually liquid lead bismuth) in the centre of the reactor. The impact of the beam on a target made of this particular material yields a large number of neutrons in a process known as spallation. These neutrons are needed to maintain the chain reactor in the reactor which would be unstable if the neutrons were just born internally in its fuel. This is an ADS (Accelerator Driven System) and several ADS experiments are underway in various countries. [1]

[6] Carbon-14 is actually an activation product i.e. it is produced by neutron capture rather than by fission. In this report we lump the two groups together for convenience of exposition.

Cladding

The fuel cladding consists of the metal structure in which the fuel pellets are encased. For the standard bundle containing 19 kg of uranium fuel pellets, the cladding mass is 5 kg of zirconium metal alloy. In reprocessing the cladding is cut mechanically to release the pellets and then set aside for storage. Since it contains the long lived isotope zirconium-93 (1,530,000 yr), it is considered intermediate level waste and therefore, it must be stored in a geologic repository.

3. Economic Implications

Determining the costs of reprocessing is neither easy nor straightforward. Much of the cost information is propriety to the firms who do commercial reprocessing and much of the reprocessing infrastructure has been subsidized by governments. Therefore, a variety of estimating methods must be used quoting a variety of published sources. However, the purpose of this section is not to derive precise cost estimates but rather to determine the economic implications of reprocessing should it be considered as part of a strategy for the long term management of used nuclear fuel.

Cost of Electricity

One measure of reprocessing costs is to compare them to the value of the electricity generated from the fuel to be reprocessed. At this time in Ontario, the total cost of electricity, including various added charges as delivered to the consumer is roughly $0.10 per kWh which is sufficient to make the rough estimates required in this report. Therefore, a kilogram of uranium in the standard CANDU bundle produces $6,289 worth of electricity at 33% thermal to electrical reactor efficiency. This provides one range against which the costs of reprocessing can be compared.

Costs of Current Commercial Plants

A variety of cost estimates for commercial reprocessing have appeared from time to time in the open literature. It is difficult to assess the validity of these estimates and indeed it could be that the same costs are repeated from one report to another paper. The costs found in researching this report are listed in Table 3 with appropriate conversions to 2004 Canadian dollars.

Table 3 Commercial reprocessing costs quoted in the literature

Price ($/kg HM)[7]	Comments
$ 774-1393	•$500-900 (1994$) USD converted to 2004 USD using US Department of Labour CPI multiplier of 1.29 and then to $CDN [5]
$ 420	•Commercial offer from Russia's RT-1 plant as reported in the media [6]
$ 879 -1390	•$370-585 (1982$) USD converted to 2004 USD using US Department of Labour CPI multiplier of 1.98 and then to $CDN [7]
$ 1200-2400	• [8-10]

[7] HM means heavy metal to reflect the small percentage of plutonium included in most cases this makes little practical difference compared to kg U.

Table 3 indicates that the prices from the several sources are in reasonable agreement with the exception of that from RT-1 which may be a "lost leader" intended to encourage commercial contracts. It would seem that $1,000 to $2,000/kg U would be a reasonable range for the spot price of commercial reprocessing.

An average reprocessing cost of $1,500/kg U is about 24% of $6,335, the average value of the electricity generated per kg U. Therefore, reprocessing the estimated 3.7 million used CANDU fuel bundles produced to the end of life from the current fleet of commercial nuclear power reactors in Canada would cost about $107 billion or about 24% of the approximately $445 billion value of the nuclear electricity generated. On the other hand, if the assumption is continued operation of Canada's current reactors at the pre-1997 rate of consumption of 113,000 bundles per year [4], then the annual value of the electricity generated would be $13.6 billion and the estimated annual reprocessing costs at 24% would amount to $3.3 billion. However, these simple estimates don't take into account such factors as the value of the fuel materials derived from the reprocessing or the savings in geologic disposal costs due to volume reduction.

Harvard Study

Bunn and his colleagues in the Project on Managing the Atom at Harvard University have done a particularly detailed study of the economics of LWR fuel reprocessing compared to direct disposal of the fuel [11]. They make as detailed estimates as possible with well justified assumptions while acknowledging the difficulty of obtaining real reprocessing price data from commercial sources. Their basic costing method is to determine what the market price of fresh uranium must be to balance the equation:

[cost of interim storage & disposal] =

[cost of reprocessing & disposal of wastes] – [value of recovered plutonium & uranium]

Another way of putting this is: at what uranium market price point is it worthwhile to reprocess? This is clearer if the last term of the above equation is restated as:

[value of recovered plutonium & uranium] =

[enriched natural uranium cost] – [recovered uranium & plutonium cost]

Note that in this methodology economic value is assigned to the recovered fuel unlike the commercial prices quoted in Table 3. In fact the calculations were made from the point of view of a US utility contemplating whether reprocessing would be advantageous. A variety of other costs such as conversion (from uranium to uranium hexafluoride for enrichment purposes), enrichment, fuel fabrication, discount rates for capital and so on are incorporated in their formulas to make them as realistic as possible. However, the parameter ranges for LWR fuel are not easily applicable to CANDU fuel mainly because the typical burnup LWR values used (43,000 and 53,000 MW day per tonne) are much greater than the standard bundle value (7,928 MW day per tonne U).

To get a sense of the results of this approach, at a uranium price of $40 US/kg and with a reprocessing price of $1,000 US/kg, reprocessing would increase the cost of nuclear electricity by 1.3 mills US per kWh or $ 0.00156 CDN per kWh. Reprocessing would

increase the US retail electricity price by up to 3%. A very crude estimate is that CANDU fuel with its lower burnup but with no front-end conversion and enrichment costs might cause a variation of a factor of 2 to 3, i.e. a 6 to 9% increase in electricity prices in Canada compared to the 24% derived earlier with no credits for reprocessing.[8] This is not a surprising result because fuel prices are generally a small percentage of the electricity costs of nuclear reactors.

Comparison with Disposal Costs

The Harvard group [11] quote a direct disposal cost of 1.8 mills CDN per kWh which would amount to $2,150 CDN per CANDU bundle for interim storage and geologic disposal. NWMO estimates a cost of $16.2 billion (2002$) for a deep geologic repository including interim storage, retrieval and transportation. The estimated number of fuel bundles to be stored is 3.7 million which works out to $4,378 per bundle or 3.6 mills CDN per kWh which is about 4% of the value of the electricity generated. The Japan Atomic Energy Commission did a recent study [12] that concluded reprocessing at their Rokkasho-Mura plant was a factor of 1.5 to 1.6 times more expensive that geologic disposal. Applied to the Canadian situation this would mean approximately 6% of the cost of electricity. All of these estimates are reasonably consistent.

Capital Costs of Reprocessing Plants

For purposes of estimation, NWMO assumes that a total of 3.7 million used fuel bundles will need to be cared for in a long-term management facility. Therefore, a total of 88,800 tonnes of used CANDU fuel would need to be reprocessed of which 70,300 tonnes would be heavy metal in uranium fuel pellets. Making the assumption that reprocessing would take place over a period of 100 years, then 703 tonnes of used fuel per year would have to be reprocessed. Table 4 shows the capacities and capital costs of some fairly recent reprocessing plant [13]. THORP and the French plants have been operation for some years whereas Japan's Rokkasho-Mura is now accepting fuel for reprocessing and will begin full operation in July 2006.

The capacity of 703 tons per year clear the backlog of used fuel over 100 years is well within the capacity of current plants.

Table 4 Capacity and Capital Costs of Recent Reprocessing Facilities

Country	tonne U per yr	Cost $CDN
France - UP2,UP3	1600	$19.2 billion
UK, Sellafield (THORP)	900	$ 7.1 billion
Japan (Rokkasho-Mura)	800	$21.6 billion

The steady state used fuel bundle production of Canadian reactors would be 113,000 bundles per year or 2,147 tonnes of uranium metal per year for a fully operational nuclear electricity system of a size similar to today's [4]. This would require two or three reprocessing plants of current size [14].

[8] Because fuel burnup in CANDU is so low, a detailed study of the type conducted by Bunn et al [11] would be required to verify these crude guesses for CANDU reactors.

Decommissioning Costs of Reprocessing Plants

Decommissioning and refurbishment costs for THORP and UP3 are estimated [11] to be in the order of $120 per kg U or the basis of a fully capitalized commercial facility. In the same calculation, the reprocessing price comes out to $2,112 per kg U or about 6%. Again this would seem reasonable in the context of other nuclear installations.

The low burnup of CANDU fuel makes it a substantially less attractive candidate for reprocessing than LWR fuel. However, on the basis of the best estimates available in the literature, it is concluded that reprocessing would add a significant but tolerable percentage increase to the cost of electricity to Canadian consumers. Hence, reprocessing for waste management purposes cannot be ruled out on purely economic grounds.

4. Radiological Implications

The issue addressed in this section is the impact of reprocessing on the total radiological hazard to the public and to workers in reprocessing plants. The specific question is how much could reprocessing reduce biological hazard or radiotoxicity from used fuel.

Reprocessing Wastes

The high and intermediate level wastes expected from reprocessing are of two broad types:

- The zirconium alloy structure of the fuel bundles must be removed to extract the uranium fuel pellets contained inside. The standard CANDU fuel bundle is comprised of about 19 kg of uranium pellets and 5 kg of zirconium alloy which constitute ‘hulls’.

- The liquid waste solutions containing fission products and minor actinides together with any remaining fuel material, stable isotopes and chemical by products of the process.

In addition there will be quantities of the usual low and intermediate level waste typical of nuclear process (for example cloths, wipes, gloves, filters and so on).

The zirconium hulls are intermediate level waste (ILW) and in today’s commercial plants are stored in cement or in containers either compacted or in original form.

The standard approach used in other countries for the high level waste (HLW) component present in the reprocessing liquids is to concentrate them to solids which are then incorporated in a glass matrix, namely vitrification. This technology has been licensed for commercial use for 20 years and is widely used in reprocessing plants worldwide. The glass blocks will eventually be stored in a geologic repository – at present, they are in interim storage.

Note that reprocessing will not eliminate the need for a geologic repository although in general it may reduce the size of the repository, reduce the radiological hazard and

reduce the length of time that the wastes must be isolated from humans and the natural environment. For example, a recent Nuclear Energy Agency report [15] has estimated that the time for used fuel to decay to levels similar to a natural uranium ore body is about 500,000 to 1,000,000 years, whereas the time for vitrified HLW to decay to the levels in uranium ore bodies is about 10,000 years.

Occupational and Public Radiation Exposure from Reprocessing Plants

In [1], occupational exposure at the French UP2 and UP3 reprocessing plants was discussed and a chart reproduced from [15] that showed that in 20 years from 1977 to 1997 the average radiation dose experienced by workers, expressed in sievert (Sv) per person per year, was reduced from about 7 mSv to 0.56 mSv. In [13] the average reduction for workers in all reprocessing plants over two decades is quoted to be from 10 to 1.5 mSv annually, similar to the UP plants. The regulations of the Canadian Nuclear Safety Commission for nuclear workers in Canada are that the annual dose to a worker should not exceed 20 mSv averaged over 5 years. Therefore radiation doses to workers at present day reprocessing plants are well within Canadian regulatory limits.

Public radiation exposure from reprocessing plants has similarly declined. In the United Kingdom the public radiation dose from reprocessing emissions is now below 0.1 mSv per person per year compared to an individual's annual exposure from natural background radiation of 2.2 mSv in the UK [13]. The CNSC regulatory limit for annual radiation exposure to a member of the Canadian public is 1 mSv per year.

Therefore, experience in operating reprocessing plants indicates that a hypothetical plant in Canada would not constitute a significant radiological hazard to its workers or to the public in normal operation. Abnormal occurrences, for example criticality and other types of accidents, would form part of a detailed safety assessment prior to plant operation – again the excellent safety record of the UP plants and THORP is encouraging.

Wastes Arising from Reprocessing

Ko and his colleagues [17] calculated the wastes arising from four scenarios including the CANDU-OT (once through - used fuel bundles to a geologic repository with no reprocessing) fuel cycle and the DUPIC fuel cycle which involves the dry reprocessing of used LWR fuel to make fuel for CANDU's. Two other fuel cycles were also included. DUPIC was shown to have advantages in terms of waste production. The CANDU-OT has the highest levels of LLW (low level waste) and ILW (intermediate level waste), primarily because of the larger size of CANDU reactors that results in larger quantities of decommissioning wastes. In conjunction with the discussion of LWR-MOX reprocessing fuel cycle, the high level waste (HLW) plus used fuel is 9 -11 cubic metre per GW year of electricity in contrast to the CANDU-OT used fuel which amounts to 200 of the same units. This is plausible since in reprocessing the volume of HLW mainly comes from the vitrified FP's and MA's which have a much smaller volume than the original used fuel, all of which must be considered HLW.

Ideally for the purposes of the present report, the key comparison would be between reprocessed CANDU fuel and CANDU OT which wasn't done. However, in general the wastes arising from recycling are not very different from those from direct through fuel

cycles [18-19]. The removal of even the major isotopes significantly reduces the volume of the HLW that must be handled but the entire original radioactivity is still present.

Influence of P&T on Post Closure Assessment of a Deep Geologic Repository

The AECL Environmental Impact Statement [4] discusses in some detail the expected impact of the isotopes in the used fuel in the years after the deep geologic repository is closed. Over a long period of time isotopes can migrate through a variety of physical barriers, for example a failed used fuel container, the backfill material and the rock around the repository and appear in the biosphere. Generally the barrier materials affect different isotopes in different ways and even after tens of thousands of years only very small quantities reach the surface environment. This safety analysis was calculated using elaborate and complex computer models based on parameters derived from years of experiments. It turns out that the post closure assessment shows that the isotopes iodine-129 and carbon-14 are the key radionuclides that may enter the biosphere after tens of thousands of years although the predicted quantities are very small.

Removing and treating the iodine-129 and the carbon-14 with special methods such as changing their chemical form (conditioning) or enclosing them in specially designed containment could mitigate the already very small risk they pose. However, since about 90% of the iodine-129 in used fuel is already tightly bound to the uranium fuel matrix and the reference design lifetime of the used fuel container in a deep geologic repository is 100,000 years, the benefits of removing and treating these isotopes is likely to be very small.

5. Status of Partitioning and Transmutation R&D

The series of information exchange meetings on P&T provide an excellent current picture of progress in the field [20-21].

United States

The Administration's energy policy as announced in 2001 [22-23] includes policy recommendations to:

- support expansion of nuclear energy in the US;
- develop advanced nuclear fuel cycles and next generation technologies; and
- develop advanced reprocessing and fuel treatment technologies.

New initiatives in the US in P&T arise from the Advanced Fuel Cycle Initiative (AFCI) with objectives:

- recovery of the energy value from used nuclear fuel;
- reduction of the civilian inventory of plutonium;
- reduction of the toxicity and heat of nuclear waste; and

- more effective use of the Yucca Mt. Repository.

The international collaboration to develop advanced reactor designs to follow after the next generation of reactors about to be constructed in the next decade, for example the Advanced CANDU Reactor is of the third generation, is called the Generation IV (Gen IV) program. Ten nations, among them the US and Canada, participate in this program which aims at developing safer, more reliable, and more economic nuclear power plants with high proliferation resistance and much improved waste management, in sum sustainable nuclear energy. Because advanced fuel cycles will be essential, P&T is part of the Gen IV program.

Until 2003 the emphasis in the US P&T program was on the Accelerator Transmutation of Waste (ATW) program. The ATW document [24] envisaged a program involving 27 years of R&D followed by 90 years of P&T for 87,000 tonnes of LWR fuel at an estimated cost of $280 billion (1999$ USD).

The ATW was abandoned in 2003 because it would have been prohibitively expensive to implement. The fundamental reason is that the US is the only country in the world that requires as a matter of nonproliferation policy that the plutonium and the MA's be keep together in all recycling scenarios including transmutation. Other programs in addition to transmutation in ADS include plans to burn the plutonium as MOX fuel in thermal reactors (already done in Europe reactor) and/or together with the actinides to be burned in fast reactors. Requiring that plutonium be transmuted makes the amount of material for transmutation prohibitively large. For example, some 64 ADS's at a cost of $100 billion or more would be needed. That's simply not economically practical and hence, the US ATW program was scrapped.

In place of the ATW program the US is pursuing programs looking at what could be done in thermal reactors to recycle plutonium and MA's. A dominant theme is to reduce the volume and heat production of nuclear waste in order to avoid what is called the "second repository" problem which means either increasing the planned capacity of Yucca Mt. in the face of equity-based protests from Nevadans or going through the very arduous and lengthy process of getting approval for another repository elsewhere, both unattractive options for the US. One method of extending the storage capacity of Yucca Mt would be to reduce the heat given off by the used fuel which would allow more fuel to be stored in a given area. Radioactive decay of fission products in the fuel, notably strontium-90 and caesium-137, is the main source of heat and thus, their removal to a relatively small specially engineered high-heat storage area would substantially increase the storage capacity of the other areas. Thus, there is a substantial effort in the AFCI program to partition these two fission products. In the program's phased approach the plutonium and the MA's would initially be disposed of as high level waste, perhaps in subsequent phases consumed in thermal reactors and ultimately destroyed by actinide burning in a Gen IV reactor.

With the demise of ATW, there remains only a science investigation of accelerator driven transmutation based in US universities (Idaho, Texas A&M, UNLV); the RACE (Reactor-Accelerator Coupling Experiment) program [25], is funded at the $2M to $3M level.

Total funding for the AFCI was $68M US in 2004 and the budget request is $46.3M in 2005. Since there is no operating fast reactor in the US and not enough hot cells for a

full P&T program, the US program heavily emphasizes international collaboration particularly with the more widely based programs in Japan and Europe.

Europe

Europe has a vigorous program in P&T. Much of the impetus for the EURATOM program on P&T originated in France. The French law of December 30, 1991 directed that R&D in used fuel waste management be done in the following areas [26]:

- minimization of waste volume and toxicity by partitioning and transmutation;
- waste packaging and conditioning for long-term containment and the feasibility of long term surface storage; and
- study of the feasibility of deep geologic disposal, with or without retrievability.

In 2005, the results of the R&D programs performed under this law will be reviewed to determine the strategic direction in which France should proceed.

The EUROTRANS transmutation program, the partitioning program EUROPART, and a program on the impact of P&T on waste management (RED-IMPACT) together have total funding of €90M ($135M CDN) from the Euratom 6th Framework Program financed by the European Commission. About a third the budget is for P&T and the remainder is for geologic repository R&D. The EURATOM contribution is at least matched by contributions from the member countries in whose laboratories most of the R&D is performed.

According to [27] the rationale for P&T in the 6th framework program is based on:

- mitigating the negative “public perception that the safety of repositories can not be guaranteed for tens of thousands of years”;
- extracting the fuel components for recycling thus, reducing the toxicity and volume of the used fuel and increasing the storage capacity of planned repositories; and
- reducing the high waste temperatures “in the first 200 years that otherwise would restrict the packing capacity of the repository”.

The value of P&T for mitigating the requirements for a geologic repository and in contributing to the public acceptability is clearly recognized in the European program.

The recent increase in the number of European Union countries to 25 has made joint R&D under the Framework programs very difficult to organize. There are now 45 European partners from 14 countries in the EUROTRANS program [28] including 18 national laboratories, 10 industries and 17 universities. The purpose of the program is to determine the feasibility of doing transmutation on an industrial scale. Initially an experimental ADS system (XT-ADS) with power less than 100MW (thermal) will be designed in parallel with tests to enhance knowledge of ADS systems. The long-term goal is an ETD (European Transmutation Demonstration) by about 2020.

A key component of EUROTRANS is the TRADE experiment which consists of a proton accelerator, neutron spallation target, and TRIGA reactor to investigate reactor-accelerator coupling which is the critical issue for an ADS. There are many supporting

technology programs such as fuel development, nuclear data, liquid metal targets and neutronics calculations. Interestingly, there is also a training program for up to 15 graduate students to address the general low level of interest by European university students in nuclear careers.

Japan

Japan has made the decision to reprocess used nuclear fuel and its new reprocessing plant at Rokkasho-Mura is nearing completion. The stated aim of Japan's P&T program [29] is "to reduce the burden of waste disposal and to utilize waste as resources".

The Japanese P&T program, called OMEGA (Options for Making Extra Gains from Actinides), is currently based in three organizations:

- Japan Atomic Energy Research Institute (JAERI);
- Japan Nuclear Cycle Development Institute (JNC); and
- Central Research Institute of the Electric Power Industry (CRIEPI).

JAERI's program involves an ADS system for transmutation of the MA's following partitioning. JNC and CRIEPI are researching fuel cycles using fast reactors with JNC responsible for MOX fuel and aqueous reprocessing and CRIEPI responsible for metallic fuel and dry reprocessing.

Japan has made considerable progress on a variety of advanced chemical separation methods in many cases using real used fuel solutions. For example, JNC has done lab scale experiments on the extraction of noble metals (ruthenium, rhodium, palladium, and others) for use as catalysts in electrolytic hydrogen production. They have also extracted strontium-90 and cesium-137 for use as industrial radioactive sources and also for heat production.

JAERI and JNC will merge next year and it is anticipated that this may help in getting Japan's fast reactor, MONJU, operating again. A major sodium coolant leak and a subsequent attempt by reactor management to conceal the seriousness of the accident lead to its shutdown in 1995.

JAERI is contributing to the construction of a linear accelerator (LINAC) in the Japan Proton Accelerator Research Complex research (J-PARC) park [30]. The 600 MeV proton beam from the LINAC will be guided into a sub-critical assembly TEF-P (Transmutation Experimental Facility - Physics) which will start construction in 2007. There will also be a TEP-T for liquid metal targets. Feasibility studies for an 800 MW (thermal) ADS capable of transmuting 250 kg of MA's annually are also underway.

Japan's P&T program is comparable in size to the European program with many collaborating institutions participating.

South Korea

South Korea [31], like Canada, currently does no uranium enrichment and reprocessing of its own but unlike Canada it does R&D in P&T. The DUPIC fuel cycle is of particular interest to Korea since that country has both LWR and CANDU reactors. DUPIC

involves repackaging used LWR fuel in CANDU form using dry processing. In fact dry reprocessing, including pyroprocessing, are major subjects of R&D interest. The design of a conceptual ADS system called HYPER is also underway.

China

By 2050 China plans to expand its nuclear electricity generation capacity by a factor of 40 to 240GW (thermal), the equivalent of about 100 Darlington reactors, in order to meet its projected energy needs [32]. Using that many LWRs would require more than 2.5 million tons of uranium. This not feasible and a fast breeder reactor system will be needed. An experimental breeder reactor, CEFR, will be in operation by 2007-08 and China is also pursuing R&D in an ADS program.

Russia

The policy of Russia is to have a closed nuclear fuel cycle with a final product equivalent in radiation to mined uranium ore [33]. There is about 16,000 tons of used fuel in storage, increasing at a rate of 850 tons per year. At the present time about 20% of the used fuel is reprocessed. An ADS experiment, the Sub-critical Accelerator Driven (SAD) assembly program at Dubna is expected to start in 2007-08. The cost is projected to be $30M USD, mostly provided by the European Union under the ISTC program with equivalent in-kind contributions by the participating Russian institutes. There is also an extensive nuclear data effort to determine high energy neutron reaction cross sections for actinides.

Additional Technical Developments

Developments in other fields of science and technology are underway which may affect the long term management of used nuclear fuel of which three examples follow.

The current burgeoning field of nanotechnology including the discovery of carbon nanotubes and fullerenes offers novel and intriguing possibilities for the treatment of radioisotopes. For example, a recent measurement [34] has demonstrated a 3% increase in the decay rate of a beryllium-7 nucleus enclosed in a fullerene structure. Fullerenes, also called “bucky balls”, are geodesic spheres comprised of 60 carbon atoms, named after the American inventor Buckminster Fuller who was the first person to construct geodesic domes. The more rapid radioactive decay rate is attributed to the electron distribution within the sphere.

Theoretically, radioactive decay may also be accelerated by frequent observation which is a consequence of uncertainly considerations as applied to proving the inapplicability of the quantum Zeno effect to decay [35]. While now somewhat abstruse, the authors say that “Another prediction is AZE [Accelerated Zeno Effect] for β-decay…may be achievable by perturbing the decaying nuclear state with a broadband fluctuating γ-ray source. Thus the present findings may serve as clues to our ability to manipulate diverse decay processes.” This appears to be an interesting line of research as is the recent work on actinide fission by laser [36] earlier mentioned in [1].

Research on the application of fusion devices to the destruction of actinides remains a lively topic. The reason is that a fusion reactor would produce abundant high energy neutrons from fusion reactions independently of the composition of the used fuel

components to be transmuted. In contrast, fast reactors and ADS systems require that MA's, plutonium and other materials be fabricated into fuels obeying stringent reactivity and safety guidelines. Thus, for these systems the materials for transmutation are intrinsic to the generation of the neutrons in the system which would not be the case in fusion, potentially a great advantage. Stacey and his colleagues propose [37] using a tokamak fusion reactor as a neutron source in a sub-critical reactor in place of the accelerator used in ADS systems.

In addition to the large and well-funded programs in Europe, Japan and other countries directed at improved partitioning and transmutation, a variety of technological developments in other fields of science may well influence P&T.

6. Conclusion

Returning to the questions originally asked in Section 1:

- What would be the cost implications of reprocessing on the long-term management of used nuclear fuel?

The conclusion of this study is that reprocessing for partitioning and conditioning of low burnup CANDU fuel would significantly increase the cost of the nuclear fuel cycle. However, this increased cost could be partially offset by the value of the nuclear fuels recovered for recycling in an on-going nuclear reactor program. The costs of transmutation are much more difficult to assess since this technology is still in the laboratory. However, given the relatively small masses of Minor Actinides that would have to be dealt with after partitioning, it can be inferred that these costs may not be prohibitive. Therefore, it is concluded that reprocessing used fuel is potentially economically feasible in the case of a continuing nuclear fission reactor program in Canada.

- Would reprocessing provide a net benefit in terms of the radioactivity to be dealt with?

At this point in time, the answer to this question must be a guarded yes. The radioactivity involved in reprocessing itself appears reasonably tolerable in terms of the radiation doses to the public, to workers and to the environment. Furthermore, partitioning allows certain isotopes to be singled out for special treatments. However, the real advances in reduction of radioactivity will come with advanced fuel cycles involving actinide burning in fast reactors and when transmutation systems are developed that can work on an industrial scale.

Large R&D programs on partitioning and transmutation are underway in many countries throughout the world with ample opportunities for Canada to be involved in scientific collaborations in these areas. Furthermore, developments in other technical fields may well have a significant impact on not only used fuel management *per se* but also on the issue of the retrievability of used fuel to be emplaced in a deep geologic repository. Systematic monitoring of this evolving scientific environment will continue to be an important function of the NWMO.

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