A Review of Reprocessing, Partitioning, and Transmutation of Spent Nuclear Fuel and the Implications for Canada

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ABSTRACT

The current status of the reprocessing, partitioning, and transmutation of used nuclear fuel are reviewed in the context of assessing the possible application of these technologies to used CANDU fuel. The status of commercial reprocessing is briefly surveyed and recent progress in world R&D programs on the transmutation of FP's and actinides using Accelerator Driven Systems is summarized. The implications of reprocessing for Canada are explored from the point of view of a long strategy for managing used CANDU fuel in terms of the costs of initiating reprocessing domestically at some time in the future including public and occupational radiation doses, and the wastes generated.

I. INTRODUCTION

This paper is a summary based on two background reports [1, 2] commissioned by the Nuclear Waste Management Organization (NWMO). These reports review the basic technology of the reprocessing, partitioning and transmutation of used nuclear fuel [1] and the cost and radiological implications of these technologies if it were decided to apply them in future to the management of used CANDU fuel [2].

Reprocessing refers to a variety of chemical and mechanical processes applied to used nuclear fuel after it is removed from the reactor. Historically the primary reason for reprocessing was to extract plutonium and uranium initially for military use and later for recycling as fuel. At the beginning of civilian 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 relatively efficient users of uranium. Furthermore, substantial plutonium is generated in the fuel by both LWR and CANDU reactors which could be used as a valuable potential fuel. At that time fast reactor programs were established in several LWR-using countries with the aim of extending the energy value of the available uranium resources. Canadian R&D during this era concentrated on the thorium fuel cycle supplemented by an accelerator breeding concept. By the 1970's these plans were largely shelved because the growth of nuclear power had slowed and ample additional uranium resources had been discovered.

Nevertheless, reprocessing has remained on the nuclear agenda in Canada and other nations since some type of reprocessing will be needed for many advanced fuel cycles, except the "once through" cycle. Hence, it has always been an essential component of the long-range plans for the future development of fission power as now exemplified in the Generation IV [3] and INPRO [4] projects. Indeed, reprocessing for advanced fuel cycles will be essential to the long term resource sustainability of fission power. Some reprocessing-based fuel cycles are already in use, for example mixed uranium-plutonium oxide (MOX) fuels in European reactors. China and India are now building fast breeder reactors in anticipation of largely expanded nuclear electricity programs and there even appears to be a modest recent revival of interest in fuel recycling in anticipation of a possible nuclear renaissance in the US.

Notwithstanding the increasing interest in fuel recycling, the primary motivation for the current large world R&D programs on reprocessing of used nuclear fuel is the reduction of its radiotoxicity by the separation of radioactive isotopes for a variety of mitigating treatments and the potential of their eventual destruction by transmutation.

Reprocessing is aimed at separating, or **partitioning**, the constituent isotopes of used fuel for further processing or long-term management. Used fuel contains uranium and plutonium isotopes that can be recycled to make additional nuclear reactor fuel. There are also fission products (FP) and minor actinides (MA) 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. The later statement assumes that the "major actinides", especially the plutonium-239, have been removed before disposal; if not then the long-lived radioactivity from the total actinides is much larger for example in a geological repository containing unreprocessed used fuel.

Partitioning involves segregating the various isotopes in used fuel but does not reduce the total radioactivity in the sense that the total is the same. However, it does isolate the various isotope categories for sequent recycling, chemical treatment to reduce their mobility in the environment (conditioning) or for transmutation.

Transmutation refers to the transformation of FP's and 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 ultimately to make it harmless. There is much R&D on partitioning and transmutation being conducted in many countries throughout the world (but not Canada). It is not yet clear whether the ultimate goal of destroying a significant degree of the toxicity of used nuclear fuel can be achieved by transmutation.

It should be emphasized that a geological repository of some type, albeit it a smaller one with less stringent requirements, will still be needed even if partitioning and transmutation are successfully implemented in the future.

II. REPROCESSING TECHNOLOGY

As documented in [5], several chemical processes were initiated in the 1940's to extract plutonium for military use 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). Other counties followed suit and 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.

Many refinements and variants of the original Purex process have been developed over the years, some of them proprietary, and many specialized side processes to treat specific isotopes are now in use. A simplified summary of a very complex technology is as follows.

The first step in the Purex 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 FP's 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 to remove any adhering fuel particles. 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 analyzed to ensure that a very high percentage of the materials resident in the uranium pellets have been removed. They are then set aside for separate disposal.

Uranium reacts in the nitric acid solution to form uranyl nitrate $UO_2 (NO_3)_2$. 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 FP's remain in the aqueous nitric acid solution. Thus, the uranium and plutonium are stripped out of the solution with high efficiency.

The separation 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 rationale is that it is a noble gas that has no physiological interaction with human 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.

Major research is also being conducted throughout the world on "dry reprocessing" including pyrochemical and electrolytic technologies in order to achieve the very high purities not attainable in Purex-type processes needed for certain transmutation schemes.

III. GLOBAL STATUS OF REPROCESSING

Reprocessing is a large and apparently profitable international business. Many nations that built large Purex reprocessing facilities for military purposes have adapted the resulting expertise and experience for a successful transition to the reprocessing of civilian fuels [6-8]. Of the approximately 258 ktHM (kilo tonne of Heavy Metal[1]) of used power reactor fuel produced to date, a third, 85 ktHM, has been reprocessed with the rest held in storage either wet or dry.

The two main reprocessing activities at the UK's Sellafield site are reprocessing the fuel from the Magnox and AGR fleet, many of which are being decommissioned and, reprocessing uranium oxide fuel from the commercial nuclear power stations of several nations. The former is done at the B205 plant which alone has handled 40 ktHM to date. 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). THORP, with a capacity of 850 tHM per year, uses a Purex type process to recover uranium and plutonium from used nuclear fuels based on uranium oxide and has reprocessed 3.8 ktHM since its inception. It was reported [6] that in 1999 THORP had an estimated £12 billion ($22.5 billion CDN) worth of reprocessing orders over a 19 year period. About half of these orders were from non-UK sources including 2 tHM of fuel from Canada's decommissioned WR-1 reactor. These profitable foreign orders help to defray the cost of THORP and Sellafield in general. The imminent reorganization of UK nuclear activities has introduced a degree of uncertainty into the future operation of THORP.

The Marcoule UP1 plant in southern France reprocessed 18 ktHM of used fuel during its operating period 1958-1985. Today the major French reprocessing capability resides in

[1] The term "Heavy Metal" is used to include the plutonium and other non-uranium metals present in the used fuel in small amounts. Thus, the mass of uranium is a close approximation to the mass of "Heavy Metal".

two large plants, UP2 and UP3, located at La Hague, Normandy whose combined throughput of 18 ktHM to date is similarly to the UP1 total. 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 with roughly half from France and the other half from Europe and Japan.

Fuel from the Russian LWR-type power reactor, the VVER, is reprocessed at Chelyabinsk at the RT-1 plant operated by the PO Mayak consortium. RT-1 has a capacity of 400 tHM per year and has been operating since 1976. Up to the present a total of 3.5 ktHM of fuel has 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. Russia is the only country thus far that recycles uranium-235 from used fuel. This is done by blending the uranium-235 from VVER 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 RBMK reactors.

Russia has the largest fast reactor program in the world. The BN-600 fast reactor at is the world's most successful fast reactor in terms of sustained reliable operation and at least one new BN-800 fast reactor is planned to be sited there which will make Chelyabinsk the world's most important nuclear fuel cycle complex.

Japan started reprocessing various types of nuclear fuel at its Tokai Reprocessing Plant (TRP) in 1980. TRP has a relatively small capacity, 90 tHM per year, and has reprocessed about 1 ktHM since beginning operation. A new 800 tHM per year plant is nearing completion at Rokkasho, designed 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 FP's returned to Japan. So far some ten electric utilities in Japan have had more than 7 ktHM of fuel reprocessed abroad.

Reprocessing in India began in 1964 with the extraction of plutonium from the fuel of the CIRUS research reactor. India is a large country with limited energy resources and thus, has a strong incentive to recycle nuclear fuel [9] and thus, India operates two reprocessing plants at Tarapur and Kalpakkam. These plants reprocess fuel not only from power reactors but also from research reactors and an experimental fast breeder reactor. While uranium is scarce, thorium is abundant and hence India is particularly interested in the thorium fuel cycle, an attractive possibility for future fuelling of its CANDU-type reactors. Therefore, 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 reprocessing used CANDU fuel. Although there is no reason to doubt that it would be possible, India has demonstrated that CANDU-type 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 for use in CANDU reactors.

In the United States, unlike other countries with LWRs, no post-reactor used fuel reprocessing is performed and permanent geological burial of the used fuel in an underground facility is planned. Even though the US had large military reprocessing facilities for plutonium production, no civilian processing has taken place there since 1977. At that time the President made it a tenet of US non-proliferation 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. A few reprocessing plants had been built by then but they were soon closed down. Since then uranium prices have fallen as new uranium reserves were discovered and the expansion of nuclear power in the US has ceased. Therefore, up until recently there was no economic or other reason for reprocessing in the US. The US administration has recently somewhat relaxed the prohibition on reprocessing and has a policy of encouraging the development of additional nuclear power in the US.

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. Considerable research was also done on thorium fuel cycles.

By the 1970's in Canada the emphasis had shifted from uranium conservation to economics that is to optimizing fuel cycles for minimum cost. Research with several fuel cycles showed that the SEU (slightly enriched uranium) fuel cycle gave the best value followed by the natural uranium once through cycle. At present SEU fuel is being fabricated for use in domestic CANDU's.

Some time after the US policy declaration of 1977 Canada's policy on used fuel reprocessing changed from reprocessing to geological disposal to accord with the US policy. There is no pressure now from domestic nuclear utilities to recycle because the cost of fuel remains only a small percentage of the cost of electricity generated by a CANDU reactor. There is also no interest in reprocessing for uranium conservation in Canada, the world's largest exporter of uranium, but there may well be future interest for long-term used fuel management.

IV. 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 the standard CANDU fuel bundle (burnup of 685 GJ per kg U) used in the AECL Environmental Impact Statement [10] ten years after its removal from the reactor to fresh fuel.

Table 1: Composition of fresh and used CANDU fuel [10]

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

As the table shows very little of the material in the fuel is actually changed, only about 1.3% of the original material. 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. 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.

Using the nuclear utilities estimate that 3.7 million bundles of used CANDU fuel will ultimately have to be dealt with, assuming that the percentage plutonium-239 is as given in the table and the standard bundle contains 19 kg of uranium, yields a total plutonium content of 190 tonne. Similarly the total uranium-235 content is 162 tonne. One could also argue that the uranium-238 might eventually be used in fast reactors and thus, may have an energy value in the future. However, there is a great deal of this material already available in the form of DU (depleted uranium) and there is no incentive to extract it at present other than to reduce the total volume of the used fuel waste.

Together the plutonium-239 and uranium-235 content is 0.5% of the used fuel, compared to the starting content of 0.7% uranium-235. Therefore, a clear incentive for reprocessing the fuel would be to remove the unused uranium-235 and plutonium-239 for recycling in fresh fuel. A further incentive for recycling would be to remove these two fuel isotopes, or "major actinides", for waste management purposes since otherwise they will make large contributions to the long term activity of used fuel in a geological repository.

It is important to note that the burnup of CANDU fuel is a factor of at least five or less than that of LWR fuel. The later has typical burnup in the range of 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 at least five greater due to the enrichment of the LWR fuel. In other words to produce the same amount of energy that one kilogram of LWR 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.

A partitioning scheme for CANDU fuel is shown in Table 2 with comments on the various partitions.

Table 2: Partitioning of Used CANDU fuel

Component	Disposition	Comments
Uranium-238	• Near-surface waste management facility a possibility if sufficient removal of MA's	•Now stored at reprocessing plants •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 fuel containing recycled plutonium-239 is burned in 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 military material after 15 to 20 years.
Minor Actinides (MA)	•Transmutation in ADS's •Burning in fast reactors •Geological disposal	•Long lived MA's are the main reason that geological repositories designed for 10's of thousands of years still emit radiation •Most significant MA's : -Plutonium-240 (6,600 yr) -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)	•Geological 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) •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 geological repositories and their feasibility for transmutation is an important research topic.
Fuel	•Geological	•These are the "hulls' consisting of the zirconium

Cladding	disposal	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)

V. REPROCESSING ISSUES

Reprocessing CANDU fuel is technically feasible and any decision to reprocess would depend on many factors. It can be anticipated that the time scale for the initiation of reprocessing in Canada would be several decades into the future. Even if there were to be a nuclear renaissance involving growth in the domestic nuclear reactor fleet large enough to strain uranium reserves with consequent high uranium prices, the economic incentive to recycle would not materialize for some time into the future since the price of uranium fuel is a very small percentage of the cost of nuclear electricity. The time needed to design, site and construct new reactors followed by a long period to exhaust today's large uranium reserves would encompass decades.

It is more likely that in Canada the motivation for future reprocessing would be as a strategy for the long term management of used fuel than for recycling of nuclear fuel. A primary consideration would be the success of transmutation systems whereby substantial reductions could be made in the radiotoxicity of the used fuel resulting in much increased confidence in the efficacy of the long term geological burial of the remaining used fuel FP's and MA's. Since the operation of a geological repository is some 50 or more years into the future, there would ample time to make a decision on this issue. At that juncture, the future form of the Canadian nuclear power industry would also be determined. Reprocessing done as part of a used fuel 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. Of course, by that time nuclear fusion might well have supplanted fission as the primary nuclear power system.

V.A *Costs*

It is very difficult to derive realistic costs for reprocessing since cost information is propriety to commercial reprocessing. However, estimates can be made from a variety of published sources in order to determine roughly the level of reprocessing costs.

One metric for 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 price of electricity, including various added charges as delivered to the consumer is about $0.10 per kWh which is certain to increase in the near future. Using this price, a kilogram of uranium in the standard CANDU bundle produces $6,289 CDN worth of electricity at 33% thermal to electrical efficiency.

In [2], a range of reprocessing costs from $1,000 to $2,000 CDN per kg of U (or per kg HM) was derived from the open literature as typical of commercial installations. Using $1,500 as an average then this would represent 24% of the value of the electricity produced by a kilogram of uranium.

Using the same average cost for the 3.7 million bundles estimated by the utilities to be the total output of the current Canadian power reactor fleet to the end of its operating

lifetime, gives a total cost of about $107 billion. This is about 24% of the approximately $445 billion value of the nuclear electricity generated. While the cost of reprocessing is great, the value of the electricity generated is a factor of four greater. Looking at it in another way, if continued operation of Canada's current reactors at the pre-1997 output of 113,000 bundles per year [10] is assumed 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-minded estimates don't take into account such factors as the value of the fuel materials derived from the reprocessing or the savings in geological repository costs due to volume reduction.

A group at Harvard University have done a detailed study with well justified assumptions of the economics of LWR fuel reprocessing compared to direct disposal of the fuel [11]. Unlike the commercial reprocessing costs considered above, in this study value is assigned to the recovered fuel. In fact the calculations were made from the point of view of a US utility contemplating whether reprocessing would be advantageous. Other costs such as conversion, enrichment, fuel fabrication, discount rates for capital are taken into account to make their estimates realistic as possible. However, the parameter ranges they use 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 CANDU bundle value (7,928 MW day per tonne U).

Their results show that in a typical case 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. This is not a surprising result because fuel prices are generally a small percentage of the electricity costs of nuclear reactors.

Comparing with the costs of direct once through disposal, the Harvard group [11] quote a direct disposal cost of 1.8 mills CDN per kWh which is $2,150 CDN per CANDU bundle for interim storage and geological disposal whereas NWMO estimates a cost of $16.2 billion (2002$ CDN) for a deep geological 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 concluded reprocessing at their Rokkasho-Mura plant was a factor of 1.5 to 1.6 times more expensive that geological disposal. Applied to the Canadian situation this would mean approximately 6% of the cost of electricity. All of these estimates are reasonably consistent.

The capital costs for the construction of reprocessing plants are debatable because of substantial government subsidization. Published costs range for current plants range from about $8,000 (UK) to $27,000 (Japan) per kg U capacity. Using $17,500 CDN per kg U capacity, a domestic plant to process 3.7 million used fuel bundles over a period of 100 years would cost about $12 billion. To handle 113,000 bundles per year in a steady state reactor economy would require up to three such plants at a total cost of about $36 billion.

V.B *Radiological Consequences*

The issue addressed in this section is the impact of reprocessing on the total radiological hazard to the public and workers. 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 perhaps with unpredictable consequences and hence, it would be best to leave them sealed and bury them intact. The question is whether one is better off by reprocessing in terms of used fuel waste management.

The zirconium alloy structural frameworks of the bundles would be treated as intermediate level waste (ILW) and stored in cement or in containers, either compacted or in original form as at today's commercial plants. The bulk of the high level waste (HLW) is in the form of solutions containing FP's and MA's with residual fuel material, stable isotopes and chemical by- products of the process. There will also be quantities of the usual low and intermediate level waste typical of nuclear process.

The standard approach used for the HLW 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 contaminated glass in appropriate containers will eventually be stored in a geological repository – at present, they are in interim storage.

Reprocessing will not eliminate the need for a geological 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 [12] 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.

As noted in [8] occupational exposures at the French UP2 and UP3 reprocessing plants is now less than 1 mSv per year and the average for all reprocessing plants is in the order of 1.5 mSv per year. Similarly, public exposure in the UK due to reprocessing is below 0.1 mSv. 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 and the exposures to the workers and the public would be well within CNSC regulations.

V.C *Safety*

As in all aspects of the nuclear enterprise safety must be a priority and reprocessing saw many accidents in the early plants [5] since the process involves nitrates and organic solvents that must be carefully handled. In 1955 the only AECL work-related fatality at Chalk River occurred as a result of a nitrate explosion at a plutonium extraction facility [13]. Fires can occur even in operations such as encasing low level radioactive wastes in

bitumen as happened in 1995 at TRP [14]. As the technology matured it was found 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 can be mitigated by the design of modern reprocessing plants, which retain and contain any radioactive materials released within thick-walled vault structures.

A particularly concerning aspect of reprocessing is the potential for criticality accidents i.e. creating an unintentional chain reaction in a mixture of nuclear materials. There have been more than 30 such accidents since the beginning of nuclear fuel processing including a particularly severe one at Tokai, Japan in 1999. A company 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 root causes of this easily avoided accident were incompetence and stupidity [15].

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. This a particularly timely example at present when Canada is for the first time is moving into fuel blending to produce SEU fuel. Any procedure involving enriched uranium opens up the possibility of criticality accidents which up to now have not been a danger in Canadian facilities dealing only with natural uranium. 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 domestic reprocessing plants if proper containment was built into them.

VI. TRANSMUTATION

Transmutation encompasses two basic means of destroying radioactive isotopes by neutron bombardment. The first is conversion of a radioactive isotope to a stable isotope by neutron capture. To be of practical use the reaction cross section must be sufficient that the transmutation occurs rapidly enough compared to decay and the isotope in question must be sufficiently isolated from other isotopes so that more radioactive isotopes aren't produced by neutron capture. This in turn requires that a highly efficient partitioning method is available for each isotope of interest.

The other means of destroying isotopes is by fission. Fast reactors are necessary since fissioning 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. Note that fission of actinides results in the production of FP's that are essentially the same as those resulting from the fission of uranium and plutonium. Thus, generally the long-lived actinides have been replaced by short-lived FP's.

The transmutation of FP's is problematic. For example, 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 used fuel. However, it is impractical to transmute these isotopes because of their relatively low neutron capture cross sections. No source currently available could deliver a sufficiently large neutron flux to make transmutation proceed faster than radioactive decay [16]. However, the radioactivity from both these isotopes will decay to negligible levels after 300 years. Thus, they could be segregated from other FP's and allow to decay in dedicated storage facilities in order to reduce the heat load requirements for long-term storage and burial facilities. On the other hand technetium-99 is one of the few FP's which is a promising candidate for transmutation. It has a sufficiently large neutron capture cross section to make transmutation feasible and stable targets could be fabricated from the metallic form. The emphasis for this isotope is in developing better pyrochemical separation technologies.

The primary attraction of transmutation lies in its application to the long-lived actinides because they are the main contributors to the long-term radiotoxicity of used fuel. The largest quantities of actinides are the uranium and plutonium isotopes and reprocessing can extract them. The MA's consist of neptunium, americium, curium and others that make significant contributions to radiotoxicity. Therefore, transmutation R&D is aimed at destroying the MA's, which could reduce by hundreds of thousands of years the time requirements for used fuel repositories.

In fast reactors leaving the plutonium and MA together would effectively consume the plutonium and fission many of the MA's. 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. Adding uranium-238 can provide the necessary stability but this results in more FP's because the fission rate of uranium-238 for fast neutrons is much higher than in a thermal reactor. More plutonium and MA's would also come from neutron capture in 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 [17]. Moreover, today there are only three operating fast reactors in the world with two more under construction. An additional means of transmutation is needed.

The Accelerator Driven System (ADS) concept is an alternate approach to transmutation being actively investigated by many of the world's nuclear programs. Figure 1 is a schematic of an ADS. An accelerator is used to produce an intense beam of high energy protons which is directed into a sub-critical fast reactor core consisting of plutonium and MA fuel. The protons impinge on a lead-bismuth target yielding copious fast neutrons by spallation. These neutrons bring the core to criticality and the desired fissioning of plutonium and MA's occurs. As indicated in the figure, in its fully developed version an ADS could be used to generate electricity although current systems are experimental.

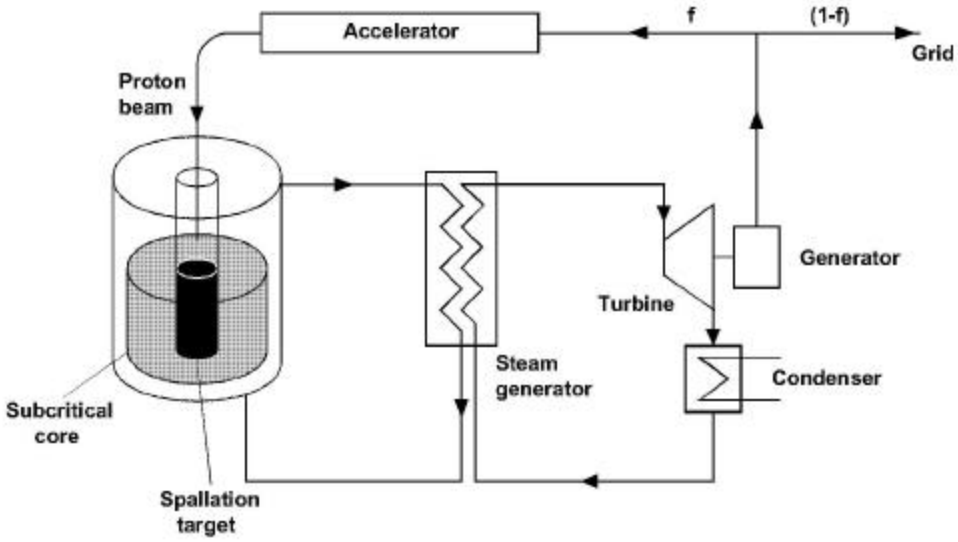

Figure 1: Accelerator Driven System (ADS) for transmutation [17]

In most of the nuclear programs planning to use the ADS concept, its purpose will be the final destruction of the MA's. However, initially in the US it was proposed to burn all the plutonium and MA recovered from used fuel in ADS facilities. The US ATW (Accelerator Transmutation of Waste) program was 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. Some 64 ADS systems were projected. The estimated cost would have been be $280 billion (US 1999$) [18]. The ATW plan was abandoned because of its very large costs which arise from the US non-proliferation policy to put all of the plutonium recovered from used LWR fuel through the ADS systems. In contrast other countries plan to recycle most of the plutonium both as MOX thermal reactor fuel and also as fast reactor fuel with only a small proportion of the plutonium used as fuel for the ADS whose primary purpose will be to consume the MA's.

VII. STATUS OF PARTITIONING AND TRANSMUTATION R&D

The international series of information exchange meetings on partitioning and transmutation R&D sponsored by the OECD Nuclear Energy Agency (NEA) provide an excellent current picture of progress in the field [19] which is summarized in this section.

There is a very high level of international cooperation and collaboration in this area. In addition to the activities of the NEA, the Generation IV (Gen IV) [3] program is an international collaboration lead by the US to develop advanced reactor designs to follow after the next generation of reactors about to be constructed in the next decade. Ten nations, among them 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, partitioning and transmutation is part of the Gen

IV program. A related program, INPRO, an International Atomic Energy Agency initiative [4] is aimed at providing the user requirements for innovative fission power systems and fuel cycles. In addition, there is a European Union program, MICANET, to develop innovative nuclear technologies. Therefore, there are ample opportunities for Canada to collaborate with other countries on partitioning and transmutation R&D if a domestic program in the area were to be undertaken.

A brief review of the activities of some of main countries in the field follows.

The US administration's energy policy of 2001 includes policy recommendations that recognize the need to develop advanced nuclear fuel cycles, next generation technologies and advanced reprocessing and fuel treatment technologies. These objectives are pursued through international collaboration and the domestic Advanced Fuel Cycle Initiative (AFCI) program.

Since the end of the ATW program in 2003, the US has been 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 FP's 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 FP's. 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 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 partitioning and transmutation program, the US heavily emphasizes international collaboration particularly with the more widely based programs in Japan and Europe.

Europe has a large and well funded R&D program on transmutation with much of the impetus for the program originating from France. The EUROTRANS transmutation program, the partitioning program EUROPART, and a program on the impact of transmutation on waste management (RED-IMPACT) together have total funding of €90M ($135M CDN) from the European Commission's 6th R&D Framework Program About a third the budget is for transmutation and the remainder is for geological

repository R&D. The Commission's contribution is at least matched by contributions from the member countries in whose laboratories most of the R&D is performed.

The value of partitioning and transmutation for mitigating the requirements for a geological repository and in contributing to the public acceptability of nuclear power is clearly recognized in the European program. There are now 45 European partners from 14 countries in the EUROTRANS program 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 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 partitioning and transmutation program is "to reduce the burden of waste disposal and to utilize waste as resources".

Japan has made considerable progress on a variety of advanced chemical separation methods in many cases using real used fuel solutions. For example, Japan Nuclear Company 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 is contributing to the construction of a linear accelerator (LINAC) in the Japan Proton Accelerator Research Complex research (J-PARC) park. 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 partitioning and transmutation program is comparable in size to the European program with many collaborating institutions participating.

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. 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's policy is to have a closed nuclear fuel cycle with a final product equivalent in radiation to mined uranium ore. There are about 16,000 tonne of used fuel in storage, increasing at a rate of 850 tonne per year. At 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.

South Korea [31], like Canada, currently does no uranium enrichment and reprocessing of its own but unlike Canada it does R&D in partitioning and transmutation. 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.

VIII. CONCLUSION

The future need for the conservation of uranium resources and the deployment of fast reactors makes recycling of used fuel an integral component of any sustainable fission power system. This includes many of the advanced fuel cycles envisaged for CANDU reactors.

Reprocessing of civilian used nuclear fuel is technically feasible and is done commercially on a large scale. Reprocessing enables the partitioning of used fuel into its components: uranium, plutonium, MA's and FP's.

The plutonium can be recycled for consumption as mixed oxide (MOX) fuel in conventional power or fast reactor fuel. Fissionable uranium taken from the fuel could also be recycled as reactor when the cost of uranium makes this practical.

Removal of the recyclable fuel components, particularly the plutonium, from used fuel is advantageous in terms of long term waste management. The FP's and the MA's can be embedded in molten glass for long-term burial in a geological facility. The chemical form of some isotopes can be altered to decrease their mobility and radiotoxicity in conditioning processes. However, although reprocessing substantially reduces the volume and radiotoxicity of the used fuel through recycling and conditioning prior to long term geological storage, a repository is still needed.

Transmutation is aimed at destroying the radioactive isotopes, primarily the long-lived FP's and the MA's, by neutron bombardment. It requires R&D on improved partitioning and the development of ADS driven systems. The time scale for the success of today's transmutation research programs is similar to the time for construction of a geological repository i.e. 50 or more years from now. Thus, the design of such a repository should

allow the used fuel to be easily retrievable to permit future partitioning and transmutation in the future.

Reprocessing as applied to low burnup CANDU fuel would significantly increase the cost of the nuclear fuel cycle. 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 MA's 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 but would not need to be initiated on an industrial scale for some decades into the future.

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. Therefore, systematic monitoring of this evolving technology should continue to be an important function of the NWMO. In terms of effectively participating in world developments in both advanced fuel cycles and long term waste management Canada should launch a modest university-based domestic R&D program of its own on partitioning and transmutation.

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