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ANALYSIS OF NUCLEAR PROLIFERATION RESISTANCE REPROCESSING AND RECYCLING TECHNOLOGIES

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ABSTRACT

The PUREX process has been progressively and continuously improved during the past three decades, and these improvements account for successful commercialization of reprocessing in a few countries. The renewed interest in nuclear energy and the international growth of nuclear electricity generation do not equate – and should not be equated - with increasing proliferation risks. Indeed, the nuclear renaissance presents a unique opportunity to enhance the culture of non-proliferation. With the recent revival of interest in nuclear technology, technical methods for prevention of nuclear proliferation are being revisited. Robust strategies to develop new advanced separation technologies are emerging worldwide for sustainability and advancement of nuclear energy with a decrease in proliferation risks. On the other hand, at this moment, advanced technologies with reduced proliferation risks are being developed. Until now proliferation resistance as it applies to reprocessing has been focused on not separating a pure stream of weapons-usable plutonium. France, as an example, has proposed a variant of the PUREX process, the COEX^{TM} process, which does not result on a pure plutonium product stream. A further step is to implement a process based on group extraction of actinides and fission products associated with a homogeneous recycling strategy (UNEX process in the U.S., GANEX process in France). Such scheme will most likely not be deployable on an industrial scale before 2030 or so because it requires intensive R&D and robust flow-sheets. Finally, future generation recycling schemes will likely handle the used nuclear fuel in fast neutron reactors. This means that the plutonium throughput of the recycling process may increase. The need is obvious for advanced aqueous recycling technologies that have less proliferation risk than the commercial PUREX process. In this paper, we review the actual PUREX process along with the advanced recycling technologies that will enhance technical barriers that make plutonium diversion more difficult by either not isolating plutonium and/or coexistence of fission products with plutonium.

1. INTRODUCTION

"*Proliferation resistance*" is that characteristic of the nuclear energy system that impedes the diversion or undeclared production of nuclear materials or misuse of technology by the host state in order to develop nuclear weapons or other nuclear explosive devices). (Pasamehmetoglu, 2006). Calculations of the associated stocks and flows of uranium, plutonium and minor actinides indicate that the proliferation risks at mid-century, using current light-water reactor (LWR) technology, may be an issue. There are institutional arrangements that may be able to provide an acceptable level of risk mitigation, but they will be challenging to implement. In developing the next generation of nuclear fuel cycle technologies, it is important to realize that, at this time, there is no such thing as a proliferation proof fuel cycle. However, it is always possible to reduce the risk of proliferation posed by the fuel cycle.

For used fuel recycling, decreasing the proliferation risks of the system has been focused on not separating a pure stream of weapons-usable plutonium. France, as an example, has proposed a variant of the PUREX process, the COEX^{TM} process, which does not result on a pure plutonium product stream. Looking ahead, group actinide extraction processes (UNEX in the U.S., GANEX in France) are under development to support a homogeneous actinide recycle strategy. These processes are currently under investigation at the bench scale, and with intense R&D could be ready for industrialization in the 2030 time frame. As the next generation of fast-spectrum reactors becomes available, the fuel cycle will likely evolve into the management of the plutonium and actinide elements using these fast spectrum systems. For nuclear energy to play an expanded role in the global energy market, innovative approaches will be necessary to address concerns about potential proliferation risks. To meet these challenges at an industrial scale, advanced aqueous recycling technologies that are intrinsically more proliferation resistant than the commercial PUREX process will be needed. In addition, plutonium safeguard strategies may be implemented. In this paper, we review the current PUREX process as a baseline technology, then examine the advanced aqueous recycling technologies designed to reduce the risk of plutonium diversion from the fuel cycle, to inform decision-makers as they evaluate the options for the next generation nuclear fuel cycle.

2. RECYCLING OF OXIDE NUCLEAR FUEL

Generation II light water reactors (LWRs), which include pressurized water reactors (PWR) and boiling water reactors (BWR), constitute a majority of all western nuclear power plants. The next generation of LWR designs, commonly categorized as Generation III reactors, are evolutions from these Gen II technologies, designed to improve fuel technology, increase thermal efficiency, include passive safety systems, and standardized design for reduced maintenance and capital costs. Improvements in reactor technology result in a longer operational life (60 years of operation, extendable to 120+ years of operation prior to complete overhaul and reactor pressure vessel replacement) compared with currently used Gen II reactors (designed for 40 years of operation, extendable to 80+ years of operation). All these Gen II and III reactors are designed to use uranium dioxide, UO_2 as the nuclear fuel.

Growth in global nuclear electric generating capacity through this century will result in the production of increasing quantities of UNF that could be dealt with by reprocessing and recycling in order to minimize the stress on uranium resources and mitigate waste disposal issues ands concerns with increasing inventories of plutonium and other fissile materials (IAEA, 2008). Over the last 50 years the principal reason for reprocessing used nuclear fuel (UNF) has been to recover unused uranium and plutonium in the used fuel elements in order to provide fresh fuel for existing and future nuclear power plants, and thereby avoids the wastage of a valuable resource. By closing the fuel cycle, some 10-15% more energy from the original uranium is gained. So far, almost 90,000 t (of 290,000 t discharged) of UNF from commercial power reactors has been reprocessed worldwide. Annual reprocessing capacity is now some 5,700 t per year for normal oxide fuels. Most of the UNF indeed- about 95% is uranium at less than 1% U-235 and up to 1% is plutonium can be recycled as fresh fuel, decreasing the need for the most hazardous operation, mining, and the most resources intensive operation, enrichment, by a combined total of 30% (Nash 2009). It also avoids leaving the plutonium in the used fuel, where in a century or two, the built-in radiological protection will have diminished, possibly allowing it to be recovered for illicit use (though it is unsuitable for weapons due to the non-fissile isotopes present). Recycling UNF may result in a significant reduction in the volume (and radiotoxicity) of highly radioactive and long-lived waste requiring permanent isolation for countries choosing to recycle instead of deploying a once-through fuel cycle. By recycling, the high level waste inventory is restricted primarily to fission products and minor actinides, with relatively low levels of plutonium, from process losses, in the waste stream proliferation even after the radiation barrier begins to fade.

3. THE PUREX PROCESS

The PUREX process, based on liquid-liquid extraction of uranium (U(VI) and plutonium (Pu(IV) by a mixture of tri-n-butylphosphate (TBP) and dodecane is a worldwide known technology and is accessible to all via publications, books etc. The PUREX process has been the basis for commercial recycling worldwide for over 30 years. During this period, the PUREX process has demonstrated high process efficiency and reliability, while providing a high quality product for producing UO_2 and mixed oxide fuels from recycled material. Over the last three decades, industry has worked towards a continuous decrease of 1) solid waste volume, 2) effluents, and 3) environmental impact in terms of radiation doses from facilities using the PUREX process.

Unfortunately, despite all these technological accomplishments, we cannot forget that any country willing to proliferate can do so in small clandestine plants (Pasamehmetoglu 2006). Countries willing to do so, can develop such capabilities based mostly (if not solely) on indigenous capabilities. For a modified open or closed fuel cycle based on the PUREX process, the potential diversion paths are illustrated in Figure 1. To address these diversion paths in the PUREX process, facilities must have a robust and adequate control of the nuclear materials that come in the recycling facility and go out from the recycling facility. With these controls, commercial recycling of UNF utilizing the PUREX process has been successfully practiced on a commercial scale for over 40 years without occurrences of diversion of special nuclear materials. These operations have been both for the purpose of UNF management for the recovery of U and Pu for recycle as UOX and MOX fuel.

Even though, nuclear fuel reprocessing has been in standby for the last 30 years in the United States, one new project, called the MOX project, deserves special attention. In 1999, the National Nuclear Security Administration (NNSA) signed a contract with a consortium, now called Shaw AREVA MOX Services, LLC to design, build, and operate a Mixed Oxide Fuel Fabrication Facility (MFFF). This facility will be a major component in the United States' program to dispose of 34 metric tons of surplus weapon-grade plutonium. If licensed, the MFFF would receive weapons-grade plutonium dioxide, PuO_2, from DOE's Pit Disassembly and Conversion Facility.

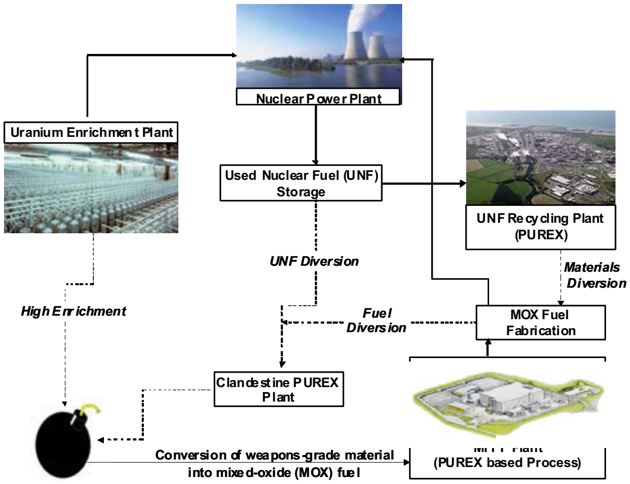

Fig. 1 Potential paths for proliferation and nonproliferation using the actual recycling technology

An additional source of plutonium, also from DOE, known as the alternate feedstock, would be processed separately in the MFFF. The facility would consist of two major areas: the Aqueous Polishing (AP) process based on the commercial PUREX process and the MOX process. In the AP process the weapons-grade plutonium would be dissolved in nitric acid, purified by TBP liquid-liquid extraction, and converted to PuO_2. In the MOX process, the purified PuO_2 obtained from the AP process would be mixed with depleted UO_2 to form the MOX fuel pellets. The pellets would then be used to produce MOX fuel rods, and finally the MOX fuel assemblies for use in nuclear power reactors. These assemblies will be irradiated in two commercial nuclear power reactors in the United States of America (USA). The MOX project is designed to eliminate, destroy, and disperse former nuclear weapons material while producing electricity in commercial nuclear plants. In this particular case, the PUREX process helps in reducing and eventually eliminating existing nuclear weapons stockpiles, clearly demonstrating industry's strong commitment to enhancing the culture of non-proliferation.

4. ADVANCED PUREX OR COEX^{TM} PROCESS

Recycling UNF now and utilizing Pu-239 as a weapons-grade MOX fuel in current reactors provide many short-term benefits, including the reduction of stored inventory (by a factor of 5 to 8), reduction in high level waste production from the fuel cycle, and most importantly reduction in the global stock of Pu-239. The percentage of Pu-239 in a weapons-grade MOX fuel will decrease from 93.5% to 51% (after 40 GWd/ton burnup) while producing energy (ANS 2009). Meanwhile the percentage of Pu-240, an undesirable isotope for weapons applications, will increase from 6.5% to 29 % (after 40 GWd/ton burnup) (ANS 2009).

Table 2: Isotopic Plutonium Concentrations (ANS 2009)

Isotope	Percent in weapons-Grade MOX Fuel before use	Percent in weapons-Grade MOX Fuel after 40 W Gwd/ton Burnup
Pu-239	93.5	51
Pu-240	6.5	29
Pu-241	-	16
Pu-242	-	4

Furthermore, when the commercial nuclear market is ready to adopt fast reactors into the fleet, plutonium will be required upfront to produce the first reactor cores. To reduce the risk of diversion from the recycling process, the COEX^{TM} process was developed. COEX^{TM} process is an evolutionary process, based on the PUREX process, designed to eliminate the production of a pure plutonium product stream. By not separating a pure stream of weapons-usable materials and by being compatible with future scenarios involving homogeneous or heterogeneous recycling of minor actinides, COEX^{TM} process (Fig. 2) aimed at further enhancing proliferation resistance (Senentz 2009).

The main characteristics of COEX^{TM} are: 1) no plutonium separation at any point of the process; and 2) use of a co-conversion process to produce a mixed solid solution of uranium and plutonium (U, Pu)O_2.

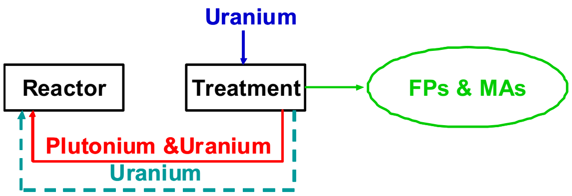

Fig. 2 Schematic of the COEX^{TM} Process (Warin, 2008)

Numerous variations are possible for COEX^{TM}. The implementation presented by Drain (Drain, 2008) is aimed at minimizing plutonium as a solid, even while mixed with uranium as well as integrating reprocessing and recycling facilities on a single site. Co-conversion can also be done with MAs (Np or Am) included in the fuel mixture. For example, a simple modification of the Pu stripping flow-sheet of the industrial PUREX process, in the first extraction cycle, allows Np to follow U-Pu stream, resulting in a U-Pu-Np mixed oxide fuel product. In the U-Pu purification cycle, Np will follow U-Pu stream towards conversion, providing some minor flow-sheet modification.

Co-conversion processes play an important role by closing the actinides partitioning and producing, at the same time, mixed actinides solid compounds as starting materials for the fabrication of new fuel elements or target/blankets for transmutation (Grandjean 2009). For advanced fuel cycles in which it is desired to manage americium and curium to further reduce radiotoxicity and heat load of High Level Waste (HLW), it could be envisaged to recycle those MAs by implementing new advanced separation systems such as the DIAMEX/SANEX/GANEX processes being developed in France or the TALPSEAK process in the U.S.

5. MINOR ACTINIDES SEPARATION

One of the intermediate term goals of the United States Nuclear Power (NP) 2010 program for the advanced light water reactor (Gen III) is to reduce used nuclear fuel storage volume while reducing the proliferation risks. One proposed solution includes increasing burn-up of the discharged used fuel and mixing MAs (Np-237 and Am-241) in the high burn-up fuel. Thus the UNF volume can be reduced, and the isotopic ratio of ^{238}Pu/Pu will be increase. For future advanced nuclear systems, MAs are viewed more as a resource to be recycled and transmuted than to be disposed off directly into a nuclear repository. As an example, on-going investigations are adding MAs such as Am-241 to a UO_2 fuel to decrease proliferation risks and improve fuel cycle performance for the intermediate term-goal of future nuclear energy systems.

Several countries are investigating the separation of MAs from the raffinate issued from a PUREX based process using new extractant molecules with two possible options for actinides separations (IAEA 2008):

1) Selective separation of MAs for interim storage, pending a decision regarding their transmutation in heterogeneous recycling mode either in Fast Reactor (blankets) or in Accelerator Driven System (ADS) (TALSPEAK process in the US, DIAMEX/SANEX Processes in France, TODGA process in Japan), and

2) Group actinide separation using an integrated fuel cycle (online fuel reprocessing and re-fabrication) with the prospect of their homogeneous recycling in Fast Reactors (GANEX process in France, NEXT process in Japan).

In the United States, the TALSPEAK process concept (Trivalent Actinide Lanthanide Separation by Phosphorus reagent Extraction from Aqueous Komplexes) is being investigated. This process was developed in the late 1960s at Oak Ridge National Laboratory as a potential replacement of the TRAMEX process (Nash 2009). Conventional TALSPEAK process combines Di-(2-ethylhexyl) phosphoric acid (HDEHP) with an aqueous medium composed of diethylenetriamene pentaacetate (DTPA) and lactic acid at a suitable pH to separate trivalent actinides from trivalent lanthanides, trivalent actinides having a stronger affinity with DTPA. Though not deployed as such for accomplishing lanthanide-trivalent separations at process scale, this process gives good promises for the future.

France has largely invested for more than 20 years in the selective separation of trivalent actinides. Liquid-liquid extraction process is under development to recover the trivalent actinides mainly Am and Cm in only one step. The one step DIAMEX SANEX process (Fig. 3) is based on the selectivity of a synergetic mixture of malonamide and HDEHP to load all the minor actinides and lanthanides in an organic phase. Transuranic elements (TRU) are then successively stripped from the solvent by complexation with DTPA. DIAMEX-SANEX processes have been tested on genuine raffinate from the PUREX process. (Paviet-Hartmann 2009)

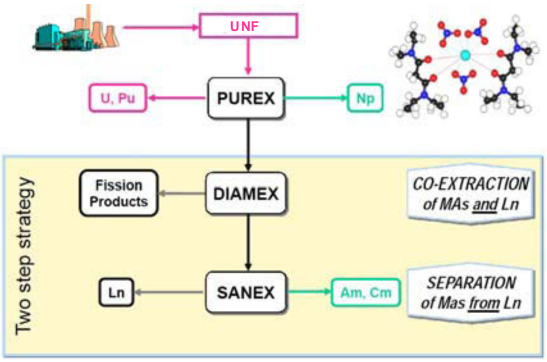

Fig. 3: French Minor Actinides (MAs) Partitioning Strategy (Courtois 2004)

The subsequent step of either the TALSPEAK process or the DIAMEX-SANEX process is the separation of Am(III) from Cm(III). In principal, both elements could be transmuted together in a fast reactor or ADS system. However because of the high heat decay and neutron emission of curium, any dry or wet fabrication process will require remote handling and continuous cooling in hot cells behind thick concrete shielding. One option involved the interim storage of curium for about 100 years, after which the relatively short lived Cm isotopes (Cm-242, Cm-243 and Cm-244) decay to plutonium isotopes which could be separated from americium. Therefore an effective method for separating americium from curium prior to re-fabrication is a major prerequisite for the discussion of further fuel cycle scenarios. The separation of Am(III) from Cm(III) is extremely delicate due to the very similar properties of both actinides. One process being investigated in Germany, the LUCA process (Modolo, 2009), uses aromatic dithiophosphinic acid to separate Am(III) from Cm(III). The promising results obtained with a surrogate solution should allow a hot demonstration to be performed in the near future.

6. TRISO FUEL FOR HIGH TEMPERATURE REACTOR

In the context of the Gen IV initiative, one particular technology being investigated is the High Temperature Reactor (HTR), with two options: 1) the High-Temperature Gas-Cooled- Reactor (HTGR), and 2) the Advanced High Temperature Reactor. Use of HTGRs for reactor-based transmutation to destroy fissionable plutonium is a logical extension of developed technology (DelCul 2002). HTGRs are able to accommodate a wide variety of mixtures of fissile and fertile materials without any significant modification of the core design. Based on helium-cooled, graphite-moderated core, this technology aims to run fuels to very high burn-ups (up to 15% FIMA (Fission per Initial Metal Atom) thanks to the high strength of the coated particles (TRISO fuel) (Fig. 4). Different kinds of particles with various kernel diameters containing either fertile or fissile isotopes may be accommodated in the same fuel

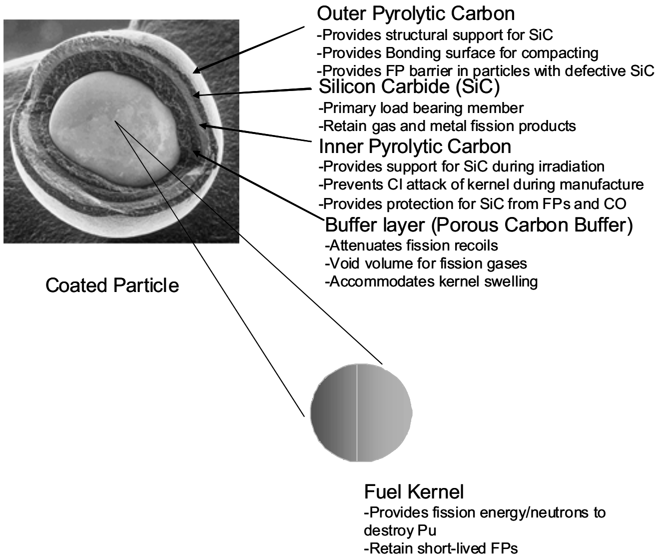

Fig. 4 Configuration of HTR TRISO Fuel (DelCul 2002, Forsberg 2010)

TRISO particles are dispersed in a graphite fuel matrix forming the HTR fuel element. These elements can provide significant barriers to the diversion of material. Because the fuel is very diluted by the fuel element graphite (low fuel volume fraction) and because of the technical difficulty to retrieve materials from within the refractory fuel coatings, unirradiated TRISO fresh fuel can provide a significant technical barrier to diversion. The TRISO UNF has these same characteristics plus self-protecting high radiation fields. Furthermore, the low volume fraction and isotopic dilution of plutonium due to the high burn-up of the TRISO UNF make it particularly unsuitable for use in weapons.

The development of recycling technology for TRISO fuels can build to some extent on experience gained during the 1970s when a process involving burning the outer layers of graphite was developed (IAEA 2008). The planned fuel is usually urania (UO_2) but can also be a mixture of urania and uranium carbide as well. Two attractive and recycling options for used TRISO fuels have been proposed by DelCul (2002). One option produces a nitric acid solution that can be introduced into an aqueous-based separation scheme typified by existing industrial scale PUREX processing plants. The other option produces a chloride salt that can be fed into pyroprocessing separation schemes, which are presently under development. This non-aqueous process includes fluoride and chloride volatility processes, carbon-chlorination processes and direct electrochemical dissolution (IAEA 2008).

DelCul et al. (2002) took into account simple processing steps, waste minimization, economy or reagents and the utilization of exiting industrial-scale processing options. Conceptually the process involved removing the fuel compacts from the graphite fuel elements, grinding the compacts to expose the fuel kernel, separating the lighter carbon particles from the heavier fuel particles, and leaching or dissolving the fuel components form the remaining carbon fines in a nitric acid solution.

The primary process that requires development and careful attention to detail include (1) crushing and milling, (2) carbon-kernel separation, (3) multi-stage dissolution and leaching of oxide particles from a matrix of carbon and silicon carbide particles, (4) solid-liquid separation, and (5) carbon waste consolidation (Fig 5).

The path of generating the least amount of waste and the one having the fewest number of process steps includes coring to remove fuel compacts, grinding and milling the fuel compacts, solid-solid separation to remove a fraction of the carbon from the fuel oxides, and dissolution to produce actinide and fission product nitrate solution. Solid-liquid separation may be combined with the dissolution process. Waste consolidation to produce a compact graphite-carbon high level waste from is included as part of the overall process.

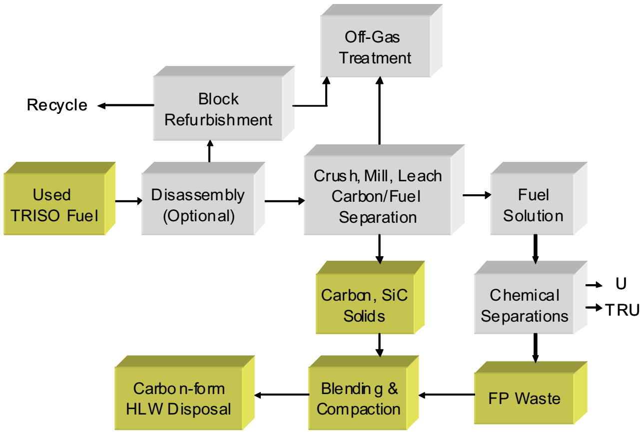

Fig. 5: Used TRISO-coated Fuel Recycle (Forsberg 2010, Collins 2010)

7. CONCLUSIONS

The selection of strategy for UNF management is a complex decision with many factors to be taken into account including technical issues associated with the composition of the domestic reactor fleet, national legal and regulatory framework, political and public acceptance, economics and environmental protection, proliferation risks. Conventional recycling technology is expected to play an important role in the medium term (IAEA 2008). The PUREX process has been progressively and continuously improved during the past three decades, and these improvements account for successful commercialization of reprocessing in a few countries such as France, Japan and UK. We should not forget though that one of the greatest proliferation concern could be associated with any enrichment process. It is clear that some techniques may be more susceptible to various kinds of abuse or misuse than others. But this should not stop the renewed interest in nuclear energy and the international growth of nuclear electricity generation. Indeed, the nuclear renaissance presents a unique opportunity to enhance the culture of nonproliferation. The nuclear industry will play a major role in strengthening this culture. While a few countries have taken irresponsible actions in the nuclear field that threaten the international and regional peace and security, the international nonproliferation system has, on the whole, been highly successful in limiting the spread of nuclear weapons. One hundred and eighty seven states now adhere to the Treaty on the Non-Proliferation Of Nuclear Weapons (NPT). Only three states, Pakistan, Iran and North Korea have elected not to join NPT, and some states such as South Africa and Libya have abandoned or dismantled their nuclear weapon programs altogether. (Lauvergeon, 2009)

Notwithstanding the recent achievements made by the second generation of reprocessing plants, there are still a number of issues to be addressed in preparation for the forthcoming new generations (Gen III and IV) of facilities aiming at replacing the operating plants at the end of their life (IAEA, 2008). The near- and medium-term challenges for recycling includes economic competitiveness, possibly through a combination of 1) higher economic value for the recycled products as a result of increasing uranium ores/conversion/enrichments costs, and 2) credits for enabling a significant increase in the ultimate disposal capacity of geologic repositories by reducing the long-term heat load and radiotoxicity of the wastes. An additional challenge is the adaptation of current technologies and plants to meet more stringent national and international regulations while at the same time accommodating increasing demands for higher in-reactor fuel performance such as higher discharge fuel burn-ups. The experience acquired by existing facilities and their operators in criticality control, higher throughputs, lower emissions and working with high level radiation allow them to be confident about the adaptability of their plants to future market and regulatory charges.

Several national and international initiatives have been launched in preparation for technical innovation in the next generation of power plants and fuel cycle facilities looking beyond the current horizon of technical innovation. However, some of these novel technical concepts are still in the development phase, and several decades may be needed to have these new advanced technologies commercially available. The full potential for industrialization of most of these novel processes will require a full understanding of their technical feasibility throughout the course of their development. (IAEA, 2008)

A critical issue for safeguarding reprocessing plants is the risk of diversion of separated fissile materials and their possible misuse for non-peaceful ends. In fact one of the stated main reasons for the move towards a direct disposal policy for UNF management in the U.S. that began in the 1970s, was concern over proliferation. The controversy as it related to PUREX-based reprocessing culminated in the latter half of the 1970s with extensive debates during the International Nuclear Fuel Cycle Evaluation (INFCE), which examined various fuel cycle concepts that might be able to mitigate the proliferation concern (IAEA 2008). Although the discussions from INFCE concluded there was no “technical fix” to the proliferation problem, it did provide an opportunity for international recognition of some technical features that could be complementary to institutional factors.

With the recent revival of interest in nuclear technology, technical methods for deterrence to nuclear proliferation are being revisited: Japan for instance (in the Rokkasho-Mura plant) mixes some uranium with plutonium. At the end of the process no pure plutonium exits in the final reprocessing stage. France has proposed a variant of its process, the COEX^{TM} process, which does not result on a pure plutonium product stream.

A further step is to implement a process based on group extraction of actinides associated with a homogeneous recycling strategy (GANEX process in France). Such scheme will most likely not be deployable on an industrial scale before 2030 or so (IAEA, 2008).

Finally, future generation recycling schemes may handle the UNF in fast neutron reactors. This means that the plutonium throughput of the recycling process may increase. These new technologies will enhance technical barriers making plutonium diversion more difficult by not isolating plutonium or/and coexistence of fission products with plutonium (low decontamination) and by degrading plutonium to have lower plutonium-fissile enrichment (lower attractiveness) (Inoue, 2009).

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