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Technology for Nuclear Reprocessing: Present and Future Directions

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Abstract: A variety of factors have served to bring nuclear technology to international attention as a viable option to meet a major fraction of the energy needs of the future. This has also brought the US policy since 1977 of a once-through nuclear cycle into question. Such a process uses only about 5% of the energy content of the nuclear fuel. Although recycling the unburnt nuclear fuel seems reasonable from a technology point of view, proliferation risks led to cessation of the technology, as conventional reprocessing can be used to make weapons grade plutonium. The economic viability of recycling is also questionable, since uranium ore prices make the once-through nuclear fuel cycle cost effective. In addition to the standard reprocessing cycle used in many countries, there are efforts to introduce advanced fuel cycle technologies that deal with some of the issues associated with the older technology. This presentation discusses the concerns with present reprocessing technologies and with the planned repository systems. Advanced separation systems designed to reduce the possibility of diversion of reprocessed material to weapons production are also discussed in the presentation. Other goals of these advanced systems are increased reactor operational safety, the reliable disposal of nuclear wastes from reprocessing for the millennia required to allow radioactive decay.

Keywords: Non-aqueous, AFC, MOX, transmutation

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INTRODUCTION

The availability of adequate energy at a reasonable price is a requirement of modern society. As the world's population increases so does the demand for electricity, transportation, affordable food, water, and other resources. As this demand grows, society also must accept the results of the Greenhouse Effect which will require substantial decreases in energy from use of fossil fuels. Coincident with this is the realization that our present, strong dependence on oil must be decreased as oil supplies decline over the next decades. It is generally recognized by experts that maximum oil production will occur within the next 5 to 25 years. Consequently, the need to have an alternative energy source to replace oil grows ever more important. Presently, approximately 85% of the domestic energy of the United States is produced from fossil fuel, 8% from nuclear and 6% from renewable energy and hydroelectric power (1). This energy use is 22% residential, 52% industrial, and 27% transportation. Efficiencies and conservation are necessary in all these areas to have maximum use of available energy. Nevertheless, it will still be necessary that alternative energy sources become available.

As awareness of these concerns becomes more recognized, much more attention is being given to the use of nuclear energy as a vital option to help solve this problem. New nuclear technologies, including advanced reprocessing cycles, are being developed and further increases in research are planned in these areas. Since 1977, the United States has relied exclusively on a "once-through fuel cycle" for nuclear power generation. As a result, the nuclear utilities extract only about 5% of the energy content of the nuclear fuel as it passes through a reactor (2). This policy had a goal to avoid production and recovery of plutonium which can be used in weapons. Other nations using nuclear power technology reprocess their spent fuel to recycle the unburnt fractions. Such a process can provide up to 96% more energy than the once-through cycle of the United States from the same initial amount of uranium fuel. Such recycling also results in less remaining uranium and plutonium for ultimate disposal in permanent repositories.

For the future, the focus should be on a generation of safe, environmentally viable, and economic nuclear energy systems to reduce the dependence on fossil fuel. This also requires development of safe, reliable nuclear waste repositories, increased research on development of alternate nuclear fuel cycles and on new and improved nuclear plant designs.

CASE FOR NUCLEAR POWER

At present, internationally, nuclear power has reached a good level of scientific and institutional maturity. There are promising plans for significant developments in the nuclear fuel cycle as many nations have made decisions to advance with systems designed to increase the use of nuclear power. An important aspect of such increased nuclear power use is that it will supply electricity; however, it can also be used to provide hydrogen gas for transportation, water desalination, and other systems requiring significant amounts of energy.

Coupled with this replacement of fossil fuel, there will be a global need for a significant increase in energy over the next 50 years as the world population has been predicted to double and, perhaps, even triple over that time. Even if the Greenhouse Effect is ignored, carbon fuel supplies cannot increase equally as oil production will decrease due to exhaustion of resources. The loss of oil will have drastic effects, particularly on transportation, unless hydrogen can be used to replace it. Production of adequate hydrogen for such use requires an amount of energy which can only be provided by nuclear power.

To develop an adequate nuclear energy technology, it will be necessary to extend the availability of nuclear fuel into future centuries by recycling the fuel to recover the unfissioned material for further burning. This use of nuclear energy will have a positive impact on the environment through the displacement of polluting carbon-based energy by electrical generation from the nuclear plants and by the production of hydrogen from these plants. This will require geologic waste repositories to accept the waste for permanent disposal with no danger of release of the spent fuel material for millennia. Ensuring that repository performance is satisfactory for very long time periods (thousands of years) will require a reduction in the lifetime and the level of toxicity of the residual radioactive wastes placed in these repositories. Developments in the nuclear fuel cycle which would remove the long-lived radioactive fission products for other use would avoid their disposal in the repositories.

PRESENT SYSTEMS

While the United States today uses the once-through fuel cycle, other countries generally use a fuel cycle that recycles the spent fuel. Recycling requires separating and purifying the uranium and plutonium from the spent fuel and using it in new fuel rods for production of more power. Recycling of commercial reactor spent fuel should be done by a system which does not produce separated plutonium for potential use in weapons.

Spent nuclear fuel reprocessing has both benefits and risks. In countries where reprocessing is done, such as France, the reuse of the separated uranium in fresh nuclear fuel is important; however, the reprocessing is expensive and creates significant amounts of weapons-grade plutonium and fission products. There are technologies which can be used to reduce the risks from these products. One such technology is known as transmutation (3). In this system, some of the fission product materials produced in the reactor operation are transmuted into less radioactive nuclides, thereby reducing the heat load and the long term radioactivity of the spent nuclear fuel considerably. It is estimated that with transmutation, repository utilization could improve by as much as a factor of 100 (4). At present, however, reprocessing has been of somewhat less interest internationally as it increases the cost of nuclear electricity relative to the once-through cycle by approximately 10%. Reprocessing would not eliminate the need for a repository; but would delay filling a repository. Also, to proceed with reprocessing internationally, it will be necessary to reduce the probability of proliferation of weapons from the plutonium. This requires development of a proliferation-resistant, economic reprocessing technology.

New technologies have been proposed for reprocessing that do not involve separating pure plutonium as a fuel material, but rather would produce a plutonium/actinide mixture (5). This would increase the difficulty of diverting plutonium and protect it from theft and handling. The actinides could also make the plutonium not usable as weapon material without sophisticated chemical separations technologies which few countries possess.

What is the situation for nuclear power in the United States? At present, license renewal requests are being prepared for at least 50% of the existing nuclear plants while five new plants are on order. In addition, the existing plants have had a 10% annual increase in electrical output beyond the initial output (6). By 2020, it is anticipated that 25% of US electricity will come from nuclear plants with 50 new plants built. The waste treatment is expected to be modified to provide a significant reduction in the level of toxicity and volume of radioactive waste to be placed in repositories. By 2050, 50% of US electricity is predicted to be produced by nuclear plants while nuclear power will be used also to produce hydrogen to provide 25% of the energy needed for transportation (7). To achieve this, it is necessary that the public recognize that the best solution to the replacement of fossil fuel is nuclear power as wind and solar energy are too limited to provide the amounts of energy needed by modern society.

A major environmental problem of nuclear power is uranium mining. However, by using the excess weapons uranium and plutonium, the United States can avoid further uranium mining for at least 100 and, possibly, 300 years. In addition, it is important to understand that new reactors will be passive (i.e., they will eliminate the possibility of operator mistakes which caused the Three Mile Island and Chernobyl accidents). Research in reprocessing methods to minimize the volume of nuclear wastes indicates a strong probability that very successful new methods will be developed to meet this goal.

ALTERNATE PROCESSES TO AQUEOUS TREATMENT

There is a growing interest in application of non-aqueous processes to the treatment of radioactive waste (8). The advantages of such processes include:

- higher radiation resistance
- more compact equipment

- production of smaller amounts of secondary waste volume
- more proliferation resistant

However, there are disadvantages such as:

- greater difficulty in conducting the separations
- smaller decontamination factors

Nevertheless, it seems probable that the advantages outweigh the disadvantages and as we move to the future generations of nuclear reactors and nuclear reprocessing systems, it is very likely that there will be a substantially diminishing use of aqueous processing with development of gaseous and non-aqueous solvent separation systems.

The systems that have been studied in the United States for reprocessing spent fuel for non-aqueous methods include molten salt systems ($NaF + LiF + ZrF_4$). Such molten salt systems provide higher radiation resistance with simpler and smaller equipment. In such systems, it would be possible to use volatility to distill UF_6 and PuF_6 from the fission products. An alternate method would be to use solvent extraction with a heat resistant solvent such as 100% tributyl phosphate. It has also been proposed to use systems of molten solutions of electropositive metal oxides. The fission products which are evolved in this process have to be removed, but the actinides could remain in the molten bath for recycling.

The most developed of these non-aqueous systems at present uses electrorefining (9). Argonne National Laboratory is using an ElectroRefining System to process uranium in spent nuclear fuel (SNF) that has been stored from the weapons program (Fig. 1).

This technology is also one of the methods proposed for use in the Partitioning & Transmutation Program (P&T) as an alternative to direct disposal. The P&T program includes both treatments by transmutation in nuclear reactors as well as by accelerator driven systems. Laboratories in France, the Czech Republic, and the Republic of Korea are developing non-aqueous systems, most of which are based on ElectroRefining. Japan is also engaged in significant research on this and other non-aqueous processes.

The present plans for the next reactors, the Generation-4 nuclear systems, propose Advanced Fuel Cycle technologies. The Advanced Fuel Cycle (AFC) has as its primary objective the management of LWR (Light Water (i.e., H_2O) Reactor) spent fuel with the development of advanced nuclear systems (Generation IV), Fig. 2. Such an AFC would include the following operations:

- quantitative recycling of U and Pu into LWR–Mixed Oxide (MOX) fuel
- reprocessing of the spent LWR–MOX fuel
- separation of minor actinides (e.g., Np, Am, Cm) from the HLLW
- fabrication of Fast Reactor (FR) Fuel (MOX, metallic on nitride) with limited content, burnup in SR

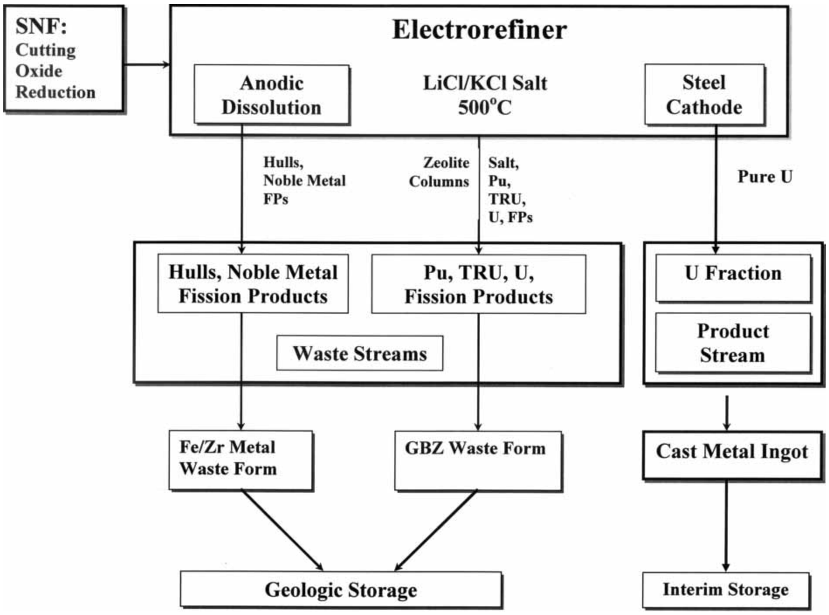

Figure 1. ANL electrometallurgical process for treatment of SNF.

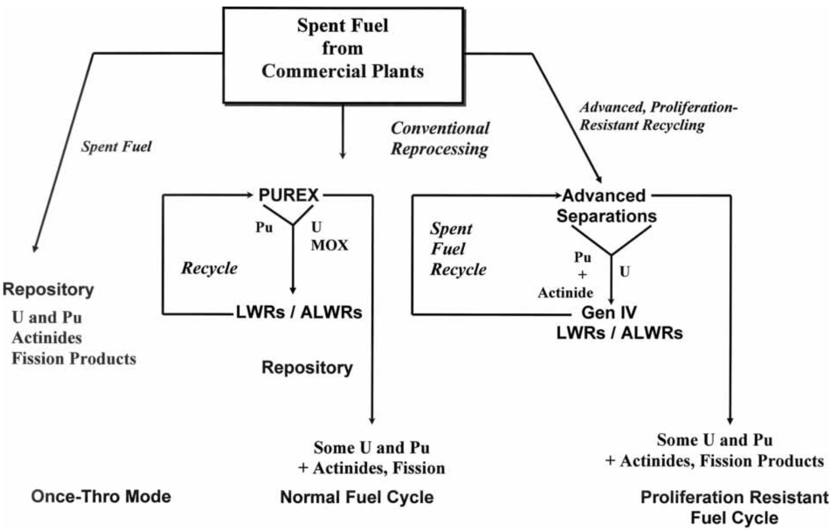

Figure 2. Advanced fuel cycle technologies.

- reprocessing of spent nuclear fuel (SNF)
- quantitative separation of all transuranium (TRU) elements from the fission products (FR)
- separation of certain fission products for disposal
- separation of other fission products for use in industry, medicine, etc.

In Fig. 2, the "Once-Through Mode" is the system used in the United States; the PUREX processing system labeled the "Normal Fuel Cycle," is used internationally. The advanced separation system, titled "Proliferation Resistant Fuel Cycle," would treat the spent fuel to provide recycling with separation of plutonium plus actinides and of uranium by advanced methods. The result would make the fuel cycle proliferation resistant with reduced amounts of actinide and fission products.

To summarize, reasonable policy for future nuclear energy reprocessing would include:

- reprocessing spent nuclear fuel by non-aqueous systems
- removal of unburnt uranium/plutonium for recycle and further burning
- remove long-lived nuclides (e.g., Tc-99, I-129, Np-237) for transmutation
- remove nuclides for use in industry, medicine, space, etc.
- permanently dispose of remaining waste in deep disposal repository

DISPOSAL OF EXCESS WEAPONS Pu

A concern among the nuclear weapons nations in the disposal of such weapons is that the treatment of the Pu and enriched uranium in the weapons be such that the products could not be used in new weapons in the future. For uranium, this would most likely be achieved by dilution of the isotopic purity of the highly-enriched uranium to the level required for nuclear fuel use which is much below the level needed for weapons use. However, for plutonium, this is not possible, and, consequently, the best way to protect against the future use of plutonium in weapons is by disposal of the Pu as MOX (mixed oxide) reactor fuel (10). This involves mixing the plutonium with uranium in the oxide fuel burnt in many reactors. The AIRES Process shown in Fig. 3 is used for the treatment of plutonium from weapons by reduction to PuH_3, followed by oxidation to PuO_2. The PuO_2 could be disposed of directly into a repository. However, such a disposition system does not eliminate the danger that in future centuries (even millennia), the plutonium would be retrieved and made into weapons as the half-life of Pu^{239} is 24,900 years. An alternative treatment is to destroy the plutonium in MOX fuel by burning it in reactors. This has the advantage of not only destroying the plutonium but also of providing significant additional energy for public use (11).

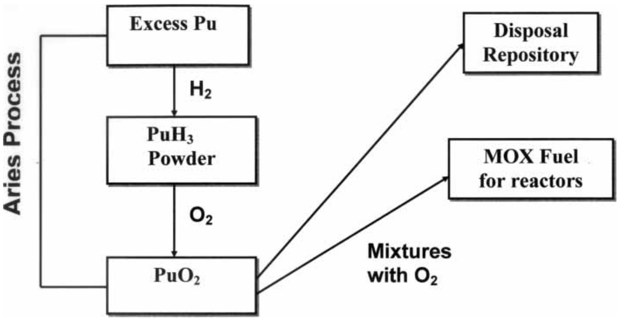

Figure 3. Disposal of excess weapons Pu as PuO_2 or by burning as MOX.

ENERGY FUTURE

The future increase in nuclear power requires the development of passive reactors with reliable accountability and protection from terrorist and other attacks. There is much talk at present of building smaller reactors in groups with reprocessing done on site. This reduces the concern of transportation of spent fuel with the possibility of terrorist attacks during such transportation.

There is also interest in the Pebble Bed Reactors (12) in which the fuel is in small "pebbles" and a fraction of these pebbles is continuously removed and reprocessed. Such processing would occur on-site at the reactor location with recycling of the U and Pu for further burnup in reactors at the same site. Again, this would avoid the transportation of material which is the basis of the highest level of terrorist concern. It is very likely that aqueous reprocessing systems would be replaced by more efficient non-aqueous alternatives. Such reactors would provided electricity and, probably, be used at the same time, as discussed earlier, for hydrogen generation as nations move into the nuclear plus hydrogen energy era.

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REFERENCES

1. Energy Information Administration, Annual Energy Review 2003, U.S. Department of Energy: Washington, D.C.
2. Choppin, G.R. (1999) Overview of chemical separation methods and technologies. In *Chemical Separation Technologies and Related Methods of Nuclear Waste*

Management: Application, Problems and Research Needs; Choppin, G.R. and Khankhasayev, M.K. (eds.), Kluwer Academic Publishers, 1–15.

3. A Roadmap for Developing Accelerator Transmutation of Waste (ATW) Technology, U.S. Department of Energy, DOE/RW–0519.
4. Nuclear Wastes: Technologies for Separation and Transmutation, Report of the N.R.C./N.A.S. National Academies Press: Washington, D.C., 1996.
5. Advanced Fuel Cycle Initiative (AFCI). Report U.S. Department of Energy, May 2005.
6. Nuclear Energy Institute, Washington, D.C., 20006-3708, www.nei.org
7. World Nuclear Association, Issue and Information, 2005, www.world-nuclear.org.
8. Choppin, G.R. (2002) Non-aqueous methods. In *Chemical Separations in Nuclear Waste Management, The State of the Art and a Look to the Future*. Choppin, G.R., Khankhasayev, M.K. and Plendl, H.S. (eds.), DOE/EM-0591.
9. U.S. National Research Council Reports on Electrometallurgical Technologies for USDOE Spent Fuel Treatment, National Acad. Sci. Press: Washington, D.C., 1995 1996a,b, 1996, 1998.
10. Choppin, G.R. (2004) Actinide chemistry: From weapons to remediation to stewardship. *Radiochim. Acta*, 92: 579.
11. Schwarty, K.L. (1998) Process Modeling of Pu Conversion and MOX Fabrication for Pu Disposition. Amarillo National Resource Centre for Plutonium. ANRCP–1998-17.
12. Clery, D. (2005) Nuclear industry dares to dream of a new dawn. *Science*, 309: 1172.