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A PYROCHEMICAL HEAD-END TREATMENT FOR SPENT NUCLEAR FUELS

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ABSTRACT

A program based upon thermodynamic values and scouting experiments at Argonne National Laboratory is proposed for development of a pyrochemical head-end treatment of spent nuclear fuels to replace the proposed chopping and leaching operation in the Purex process. The treatment consists of separation of the cladding from the oxide fuel by dissolution into liquid zinc; oxide reduction of uranium and plutonium and dissolution into a zinc-magnesium alloy; separation of alkali, alkaline earth, and rare earth fission products into a molten salt; and, finally, separation and recovery of the plutonium and uranium in the alloy. Uranium and plutonium would be separated from the fuel cladding and selected fission products in a form readily dissolvable in nitric acid. The head-end process could be developed eventually into an optimum method for recovering uranium, plutonium, and selected fission products and for minimizing wastes as compact, stable solids. Developmental expenses are not known clearly, but the potential advantages of the process are impressive.

I. INTRODUCTION

We recently reviewed several partitioning proposals for recovery of valuable fission products from spent nuclear fuels.¹ We briefly described the aqueous Purex process, the salt transport process, melt refining, the fluoride volatility process, and a gravimetric separation. All were possible technically, but further research and development is needed to develop practical processes.

We believe that the Purex process will be used to recover uranium and plutonium from light water reactor (LWR) fuels. Under the proposed conditions, however, valuable fission products will be discarded. Massive programs, with developmental costs of at least three million dollars, have been proposed for modifying the Purex process to give effective

partitioning.^{2,3} It is not clear that the developmental task of providing partitioning by the Purex process is easier than that for the alternative methods. In fact, pyrochemical processes would effectively separate selected elements and minimize waste volume.

The pyrochemical head-end treatment proposed here for spent fuels from LWRs and liquid metal-cooled fast breeder reactors (LMFBRs) would be conducted with liquid alloys and molten salts at temperatures varying from 600 to 1400°C. Such temperatures may complicate operations, but studies indicate that uranium, plutonium, and selected actinides can be separated very effectively.

Pyrochemical methods have been applied at the Los Alamos Scientific Laboratory in electrorefining operations to purify plutonium,⁴ in salt extraction recovery of americium at the Rocky Flats Plant,⁵ and

in melt-refining recycling of uranium in the Second Experimental Breeder Reactor (EBR-II).⁶

The liquid alloys and molten salts used in pyrochemical processing are resistant to radiation damage. Therefore, storage times before reprocessing can be minimized and many chemicals can be recycled. Solvent volumes are small relative to those in the Purex process. The remote recovery plant can be operated and maintained simply. Finally, wastes from pyrochemical operations are usually compact solids.

The first phase in replacing the Purex process could be to change the method for separating the cladding from the spent fuel. In the proposed Purex program for the LWR, fuel elements will be chopped into small pieces and the exposed fuel will be leached into nitric acid. However, there is evidence that the spent fuel is not completely dissolved.⁷ In the pyrochemical treatment, the cladding would be dissolved into liquid zinc. The fuel would be reduced and dissolved into a liquid alloy. Alkali, alkaline earth, rare earth elements, and iodine would then be transferred from the alloy into a molten chloride salt. Finally, plutonium, uranium, and other elements that remain in the alloy would be separated and recovered. The final separation and purification could be done in a Purex system. As experience is gained, the pyrochemical methods could develop into an excellent means for safe, efficient reprocessing and for optimum recovery of the actinide elements.

II. THE PYROCHEMICAL HEAD-END TREATMENT

The major purpose of the pyrochemical head-end treatment will be to deliver a uranium-plutonium product to an aqueous (Purex) processing facility. This product would be free of iodine, the alkaline earth, the rare earth, and the gaseous fission products, and it should dissolve completely in nitric acid. The pyrochemical treatment is compact enough that the initial processing could be conducted at the reactor site. Usually, plutonium would be pure enough after this treatment so that it could be recycled directly into fuel fabrication. This would eliminate expensive transportation and much non-productive time, in which plutonium is not in use in

the reactor. The uranium and plutonium, whenever necessary, could be shipped to an aqueous facility as a safe, compact, solid alloy.

Specific processing operations are based upon experiments conducted at Argonne National Laboratory (ANL) in support of the EBR-II melt refining, skull reclamation, and salt transport processes.^{8,9} These experiments indicated that pyrochemical treatments would be practical for recycling spent reactor fuels.

Cladding Removal

After brief storage to allow some intensely radioactive species to decay, fuel assemblies would be lowered into a liquid zinc bath at 800°C. The zinc would be covered with a molten halide salt to reduce losses by vaporization. Most of the gases and iodine would be released from the fuel as the zircalloy or stainless steel cladding dissolved. Gases would be collected in an argon cover gas, and iodine would transfer into the halide salt phase. The oxide fuel would remain as a massive solid.

After the cladding had dissolved, the solution would be siphoned into a mild steel vessel. Zinc would be recovered by vacuum distillation, and the cladding, which is radioactive, would be stored.

The intact fuel could then be dissolved in nitric acid and treated by the Purex process. However, more fission products can be removed efficiently by further pyrochemical processing.⁹ The chop and leach cladding separation would have been replaced by zinc dissolution. Chopping the fuel element is difficult at low radiation levels, and probably will be more so at the higher radiation expected in spent LMFBR fuels. Residues that contain zirconium, rhodium, ruthenium, and palladium remain with the chopped cladding after leaching.¹⁰ Wheelwright found that 0.7 wt% of the total plutonium in a Yankee uranium fuel element (39 000 MW days/ton burnup) remained as a solid after chopping and leaching.¹¹

Zinc decladding is one of several ways to separate the cladding. Cladding will dissolve in a variety of liquid metals or alloys such as bismuth, silver, and copper-antimony. The simplest procedure would be to melt and collect the cladding at about 1600°C.¹² Most of the melt would separate, and the oxide fuel

would be exposed for subsequent treatment. However, the high process temperature would cause reactions between containers and volatile elements in the fuel.

Liquid zinc attacks nickel in stainless steel, and up to 8 wt% of the steel is dissolved.¹³ Zinc slurries containing 15 wt% stainless steel were achieved without external stirring. Zinc-steel solutions were siphoned from a simulated fuel through heated piping.¹⁴ Heat must be supplied to vaporize the zinc from the zinc-steel solution. The distillate must then be cooled to condense.

The gaseous fission products would collect in an inert cover gas during cladding dissolution and be transferred to gas storage tanks. If these gases become valuable, they can be recovered and sold.

Areas of concern in the dissolution process include corrosion of container materials at elevated temperatures, distribution and behavior of fission products during decladding, possible physical trapping of volatile gases, and heat transfer during recycling of the zinc.

Reduction of Oxides and Chloride Slagging

The spent fuel would remain in the dissolution chamber as a massive solid after removal of the zinc and cladding solution. The second step in the pyrochemical treatment will be to convert plutonium and uranium to metals and dissolve both elements in a liquid alloy. This is the oxide reduction process.

Uranium and plutonium can be oxide reduced and dissolved into an alloy by using calcium or magnesium metal as the reducing agent in a liquid magnesium-zinc alloy of low magnesium content.^{8,10,16} The resulting calcium or magnesium oxide will transfer into the cover salt. Any remaining volatile fission product should be released into the cover gas. Experimentally, high-fired uranium dioxide has been reduced by a zinc-magnesium-calcium alloy.¹⁷ Excess calcium was oxidized by magnesium chloride and concentrated in a calcium-potassium chloride salt phase.

The alloy containing uranium and plutonium could be solidified and shipped to a central Purex processing facility for further purification and recovery. However, more fission products can be

separated and the plutonium can probably be recovered for recycling by contacting the alloy with a molten chloride salt. This operation, chloride slagging, will transfer the alkali, alkaline earth, and rare earth elements from the alloy into the salt phase. The alloy would be siphoned into another container, and the salt phase would remain in the dissolution chamber.

The slag can be stored in this form, or the fission products can be removed and the slag can be recycled.¹⁸ Fission products can be recovered by a selective Purex process or by transfer into another alloy such as aluminum-magnesium.

After the reducing and slagging, the composition of the alloy containing the uranium and plutonium can be adjusted to precipitate the uranium. The remaining plutonium would be concentrated by distilling away the alloy. Both the alloy and plutonium could then be recycled. The uranium would be shipped to a Purex facility for reprocessing. Eventually, impurities in the plutonium would reach poisoning levels, and both elements would be shipped as a solid alloy to the Purex facility.

Shipment of a solid alloy should be safer than similar transfer of spent oxide fuels. If spills, leaks, or accidents occur during shipment, the alloy will effectively prevent widespread contamination and minimize cleanup. Any chance of nuclear criticality would be eliminated. Finally, both uranium and plutonium in the alloy form should dissolve readily in nitric acid for Purex processing.

Product Recovery

The salt phase, which would contain many of the fission products, would be siphoned off, cooled, and stored. Valuable elements, such as americium and the rare earths, could be recovered by a Purex treatment or by contact with alloys and salts selected for purification of given elements. The salt itself could be purified for recycling by transferring the fission products into a second alloy for storage as a compact, stable waste.

The uranium, plutonium, and more noble elements would remain in the zinc-magnesium alloy. This solution could be dissolved immediately in nitric acid, or the uranium-plutonium product could be concentrated either by vacuum distillation or

precipitation before further processing. The zinc-magnesium alloy could be recycled without further modification.

A container resistant to liquid zinc, uranium, and plutonium is required for the distillation. A number of alloys or oxides may be satisfactory, but, so far, only tungsten has been used.¹⁹ Equipment must be carefully designed so that a critical mass cannot form during the distillation. Heat transfer problems will be encountered during zinc distillation and recycling.

Rather than using distillation, one can add oxygen, nitrogen, or a selected solid to the alloy to precipitate plutonium and uranium. For example, both plutonium and uranium nitride will precipitate if calcium nitride is added.¹⁹ Oxide precipitation will cause magnesia to form as a major impurity. If a precipitation method were adopted, a less volatile alloy such as copper-magnesium could be used in place of the zinc and magnesium for the oxide reduction and slagging.

High-purity fission products can also be recovered from the salts and alloys. Problems in developing a practical commercial process will be formidable, but the advantages would be large. Waste volumes would be reduced, plutonium storage time would be minimized, fission products would be separated in commercially useful forms, wastes would be stable solids, and reagents could be recycled.

III. THE SALT TRANSPORT PROCESS

If the alloy from oxide reduction of the spent fuel is treated with a series of liquid metals and molten salts to isolate specific fission products, the head-end treatment becomes the salt transport process.^{9,20} The actinides would be separated in a series of salt transfer cells, probably in specially designed mixer-settler compartments, according to chemical reactivity. For example, the element to be purified would transfer from the initial donor alloy into the liquid salt carrier in exchange for a second element common to the salt and the acceptor alloy. The element to be purified will transfer through the salt and then exchange with the common element into the acceptor alloy. Use of multiple series of selected alloys and transfer salts would provide very effective separations. Alloys and salts, which are radiation resistant, could be recycled. Wastes would be compact solids.

The problems in material selection, equipment design, and operation procedures in the salt transport process should be similar to those in the pyrochemical head-end treatment. Development of the salt transport process would aid in planning a total process for treating spent LMFBR fuels. Such fuels will contain more fission products, be at higher temperatures, and radiate more intensely than the present spent LWR fuels. These changes should encourage development of a practical salt transport process.

IV. THE EXPERIMENTAL PROGRAM

Problems of corrosion, solubility, distribution, and recycling should be examined by a series of research projects. Corrosion-resistant materials must be used throughout the pyrochemical process. They must resist attack by liquid alloys, zinc, plutonium, the actinides, and molten halide salts. A variety of materials have been used successfully at high temperatures. Molybdenum-30 wt% tungsten alloy, which can be machined, was used to pipe molten zinc at 800°C.¹⁴ In electrorefining, molten plutonium is collected in spun tungsten crucibles.²¹ Americium is purified by molten salt extraction in tantalum crucibles thinly coated with tantalum oxide.²¹ Mullins used magnesia crucibles in electrorefining and chloride slagging.^{4,22} These crucibles were in contact with plutonium, selected actinides, liquid alloys, and molten salts. Spent fuel from the EBR-II was processed in lime-stabilized zirconia crucibles.⁶

The search for suitable materials should include review of materials for LMFBR and LWR piping and consideration of newer alloys and ceramics. Initial high costs of suitable container materials can be offset by reduction of maintenance and replacement costs. Consultations with experts on corrosion should also be valuable. Laboratory work should include static and dynamic corrosion studies on many materials. The most promising would be used in process-related experiments in a continuing evaluation.

Quantitative iodine removal into the molten cover salts should be verified. Simulated fuels can be used for these tests with liquid zinc and molten halide salts. Further studies of volatile fission product release during decladding and oxide reduction will be necessary to demonstrate complete recovery.

Selective purification of valuable gases from the cover gas should also be demonstrated.

Experiments are needed to prove that spent oxide fuels are, indeed, reduced quantitatively during oxide reduction and are dissolved into the alloy. The concentration of the products that remain in the alloy after slagging should be demonstrated with simulated and spent fuels, both by distillation of the alloy and by precipitation of the fission products.

The distribution of fission products between the slagging salt and the alloy should be determined. It can be estimated for a chloride salt in contact with an alloy that contains magnesium as an active element from the stabilities of the fission product chlorides relative to magnesium chloride. Those elements that form more stable chlorides than magnesium chloride concentrate in the salt phase, whereas those that form less stable chlorides concentrate in the alloy phase. If activity coefficients are known or can be estimated, the distribution can be estimated from the standard energies of formation of the chlorides by calculating a thermodynamic equilibrium constant for the reaction

where M is the fission product. Standard free energies and equilibrium constants, assuming unit activity, are listed in Table I.²²

Lanthanum and cesium should be converted to chlorides and transferred to the salt phase. In fact, all alkali, alkaline earth, and rare earth elements should concentrate in the salt phase. The refractory and noble metals such as chromium, nickel, and rhodium would remain in the alloy. Plutonium would be expected in both the salt and alloy; in experimental studies, however, it concentrated in the alloy phase.²⁴

Johnson used experimentally determined values for uranium and plutonium solubilities in the individual solvent metals to estimate these elements' activities, and thus their solubilities, in selected alloys.²⁵ His estimates of uranium solubility in zinc-magnesium alloys and of uranium distribution coefficients between molten magnesium chloride and zinc and magnesium alloys agreed with available experimental data. In distribution studies, neptunium, plutonium, uranium, and curium were concentrated in the alloy, whereas californium and einsteinium

TABLE I

STANDARD FREE ENERGIES OF
FORMATION OF CHLORIDES
AND THERMODYNAMIC
EQUILIBRIUM CONSTANTS
FOR REACTION WITH MgCl_2

MCl_n	Free Energy (1000 K)	Equilibrium Constant
BaCl ₂	83.4	1.7×10^{11}
KCl	81.4	1.5×10^6
RbCl ₂	81.2	1.4×10^5
SnCl ₂	81.0	1.5×10^{10}
CsCl	80.0	7.4×10^4
SmCl ₂	80.0	5.6×10^9
LiCl	78.8	4.1×10^4
CaCl ₂	77.9	6.8×10^5
NaCl	75.7	8.6×10^3
LaCl ₃	67.0	8.6×10^3
PrCl ₃	66.3	4.4×10^5
CeCl ₃	66.3	4.4×10^5
ThCl ₃	65.3	9.6×10^4
NdCl ₃	64.2	1.8×10^4
YCl ₃	61.2	2.0×10^2
PuCl ₃	58.9	6.1
MgCl ₂	57.7	1.00
UCl ₃	54.0	3.8×10^{-3}
ZrCl ₂	49.2	1.9×10^{-4}
MnCl ₂	42.3	1.9×10^{-7}
ZnCl ₂	35.0	1.2×10^{-10}
CrCl ₂	31.9	5.3×10^{-12}
CdCl ₂	30.4	1.2×10^{-12}
FeCl ₂	26.6	2.5×10^{-14}
NbCl ₅	24.6	6.8×10^{-37}
CuCl	22.0	1.6×10^{-8}
NiCl	20.0	3.3×10^{-17}
MoCl ₂	8.0	1.9×10^{-23}
TcCl ₃	7.0	5.7×10^{-34}
RhCl	5.8	4.5×10^{-12}
PdCl ₂	3.8	2.8×10^{-24}
RuCl ₃	1.4	1.2×10^{-37}

concentrated in the salt phase.²² Americium was present in significant amounts in both phases. In other slagging studies, rare earth impurities were transferred into lithium-sodium-potassium slags by

magnesium, but the plutonium in a simulated spent fuel remained in the alloy.²³

Estimates of fission product distribution, solubilities, and possibilities of physically trapping simulated fuel must be checked experimentally. The slagging should be optimized for greatest separation of the rare earth and alkaline earth elements.

Heat transfer during vacuum distillations in zinc recycling and product concentrating must be examined. Consultation with industrial engineers familiar with commercial pyrochemistry application would be useful in designing the equipment for these operations. Recycling of zinc, its alloys, and the chloride salts is important to the success of the pyrochemical process, and it should be demonstrated on a pilot plant scale.

The pyrochemical treatment can be improved further by developing special equipment such as chambers for decladding, mixer-settler vessels, and vacuum distillation retorts. This equipment should be designed and tested as the process is developed.

Finally, individual fission product recovery should be demonstrated. Recovery of these elements could become extremely profitable as commercial applications develop. It is entirely possible that sale of these elements will more than pay for the complete recovery cycle.

V. SUMMARY

The pyrochemical head-end treatment for spent nuclear fuels consists of a series of remote processing operations. The cladding would be separated from the spent fuel elements by dissolution in liquid zinc (the decladding step). The spent fuel elements would then be reduced and dissolved in a liquid alloy (the oxide reduction step). Selected fission products would be extracted into molten chloride salts (the slagging step), and plutonium, uranium, and other fission products would then be recovered from the alloy.

The pyrochemical products can be transferred to aqueous (probably Purex) processing after the cladding is removed, after oxide reduction of the spent fuel, and after the contact with chloride salts. Completing all three operations will remove the volatile elements, alkali, alkaline earth and rare earth fission products, and iodine before aqueous

processing. Adding a series of liquid alloy and molten salt extractions after the oxide reduction would permit individual fission products to be separated and purified.

The pyrochemical head-end treatment will separate cladding, fission products, and volatile gases at a compact, remotely operated facility. Reagents will be radiation resistant and should be recyclable. The inventory of plutonium that is not in reactor fuel would be minimized. Wastes would be generated as compact solids, and both wastes and products would be in shippable forms.

Initially, pyrochemical treatment would replace the chopping and leaching process. Later, research could be expanded to develop an excellent pyrochemical process for recovering the valuable fission products that will be present in spent LMFBF fuels.

The proposed pyrochemical head-end process is technically feasible, and its technological basis has been established. More research and developmental programs will be needed, however, to make it commercially practical.

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