Radioactive Waste Management Associates

Memo

To: David Schlissel

From: Marvin Resnikoff

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Re: GNEP and Radioactive Waste

This memo discusses the Department of Energy's Global Nuclear Energy Partnership (GNEP) as it concerns the repository waste burden and assured fuel supply, two benefits claimed by the DOE. To prepare this memo we have reviewed a large number of documents, including historical technical documents. The idea of partitioning and transmuting waste materials has been discussed for at least 35 years[1], and the claimed benefit since 1975.[2] Yet, little progress has been made to-date.

Waste Burden

One of the ostensible purposes of the GNEP proposal by the Department of Energy (DOE) is to reduce the repository burden. The proposed Yucca Mountain repository has a mandated capacity of 70,000 metric tons, but the potential loadings will likely exceed 200,000 metric tons. However, as we discuss below, in terms of volume, heat or toxicity, it is unlikely that this objective of easing the repository burden can be accomplished with the present state of the art. The GNEP proposal has several stages that must be considered; the standard PUREX reprocessing technology and the advanced fuel cycle that has several refinements. A favorable alternative to the GNEP proposal is continual storage of spent fuel in dry casks at each reactor site or regional sites. This is the present no-action alternative that has several advantages over the GNEP proposal.[3]

PUREX and Pu Recycle

The standard PUREX reprocessing technology, in use since the Manhattan Project, separates out the plutonium (Pu) from uranium (U) from fission products. The radioactive fuel cladding hulls and process wastes remain and are buried in surface landfills. This would be the case under GNEP, though process wastes would increase under the Advanced Fuel Initiative (AFI). At West Valley, the only former commercial reprocessing venture that has operated in the United States, cladding hulls and process wastes were buried in deep holes in an NRC-licensed burial ground. Pu and I-129 contaminated organic wastes have been leaking from this burial ground since the '60's.

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[1] Claiborne, H.C., "Neutron-Induced Transmutation of high-Level Radioactive Waste," Oak Ridge National Laboratory, ORNL-TM-3964, December 1972.

[2] Claiborne, H.C., "Effect of Actinide Removal on the Long-Term Hazard of High-Level Waste," Oak Ridge National Laboratory, ORNL-TM-4724, January 1975.

[3] Lester, R.K., "Energy Conundrums," Issues in Science and Technology, Summer 2006, National Academy of Sciences

Under reprocessing, some long-lived actinides, such as neptunium, and some long-lived fission products, such as technetium-99, remain in part with the uranium[4], but almost all these radionuclides, greater than 95%, are associated with the high-level waste (HLW). Technetium has caused great problems at the gaseous diffusion plants since TcO_6 volatilizes at approximately the same temperature as UO_6 and must be trapped to prevent it from entering the environment or leaving with the enriched U product.[5] Some long-lived fission products, such as iodine-129, remain in the process wastes and also in the fission product high-level waste (HLW).

Under this PUREX stage, the Pu would be recycled into mixed oxide fuel (MOX). To have self-sustaining Pu production, the reactor recipe must be approximately one part MOX fuel (where Pu is mixed with natural U) to 2 parts enriched uranium fuel. That is, under this plan, the recovered uranium, which has less fissionable U-235 than natural U, is not recycled. In addition, recovered U contains U-236, a neutron poison, requiring a higher percent of fissionable U-235 and Pu-239+241. Since 1/3 of the reactor core would use natural (not enriched) U, there is a natural resource saving at this stage, in that less U ore would have to be mined. In order to produce 30 tons of enriched U fuel needed to operate a 1000-megawatt pressurized water reactor for a year, about 272 tons of natural uranium must be fed to the gaseous diffusion plant. Because ore in the U.S. contains a mere 0.15% uranium, approximately 180,000 tons of U ore must be mined to produce the 272 tons U fed to an enrichment plant. Thus, to a small extent, using 1/3 part natural uranium does conserve U resources and also the energy needed to enrich U in the fissionable isotope U-235. It should be noted that storage of U tailings from gaseous diffusion plants is a major environmental problem. Many of the UO6 containers are rusting out.

In terms of volume, heat and toxicity, this stage does little to reduce the repository burden. Since the fission product high-level waste contains americium (Am), curium (Cm) and neptunium (Np), this waste will remain toxic essentially forever. Over time, Am-241 decays to Np-237 (half-life, 2 million years), and I-129 (half-life, 16 million years) implying HLW will remain toxic, essentially forever. This can be seen in Fig. 1. It is important to note that the Fig. 1 does not take into account I-129 and also the buildup of actinides when Pu is recycled several times. Nevertheless as seen, the HLW hazard is greater for MOX fuel (shown in the figure as PWR-Pu fuel) than standard U fuel. The continual recycling of Pu in MOX fuel leads to the buildup of greater amounts of Am, Cm and higher isotopes of Pu. Comparing U to MOX fuel where the Pu has been recycled 5 times (Type E fuel), 8 times as much Pu-242 and Am-241 are created, 7 times as much Cm-242 and 15 times as much Cm-244. This is shown in Table 1, taken from the NRC's GESMO EIS[6]. These alpha-emitting radionuclides increase the heat production and toxicity of high-level waste. The heat production of HLW is important because the high-level waste (HLW) repository will have a limit on the heat production or power per acre (kw/acre) implying that MOX HLW will have to be spaced further apart. In short, under the standard reprocessing scenario, neither the hazard, nor the heat output of HLW is reduced. In terms of radiotoxicity, because of the buildup of actinides, spent MOX fuel is 8 times more toxic than spent U oxide fuel.[7]

Done exactly, an evaluation of the hazard of HLW is not a simple calculation. One must develop a scenario for how each radioactive material escapes its container and the geologic repository, and reaches humans. Two simplified approaches to calculate this hazard have been used in the literature. The IAEA estimates the amount of alpha emitting radionuclides in the waste over time and compares

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[4] IAEA (2003)

[5] dela Merced, M, B Hintermann and M Resnikoff, "Groundwater Movement at the Portsmouth Gaseous Diffusion Plant," Radioactive Waste Management Associates, report prepared for PRESS and the Uranium Enrichment Project, February 2002.

[6] Nuclear Regulatory Commission, "Final Generic Environmental Statement on the Use of Recycle Plutonium in Mixed Oxide Fuel in Light Water Cooled Reactors," NUREG-0002, August 1976.

[7] International Atomic Energy Agency, "Partitioning and Transmutation: Radioactive Waste Management Option, " Workshop on Technology and Applications of Accelerator Driven Systems (ADS), ICTP, Trieste, Italy, 13-17 October 2003.

this quantity to the transuranic limit, 100 nanocuries per gram, and to that in uranium ore.[8] An alternative approach is to estimate the volume of water required to dilute the radionuclides in HLW to "safe" levels, using the maximum permissible concentrations found in Nuclear Regulatory Commission regulations, 10 CFR Part 20, and to compare this water volume to that needed by U ore.[9]

While the volume of HLW in the repository is a function of the power output, the industry also considers the volume of the waste itself and claims there is a waste volume reduction in the PUREX process. The waste form being considered for HLW is borosilicate glass. In terms of waste volume, the borosilicate glass waste volume is many times greater than the original volume of the spent fuel sent to the reprocessing plant. To chemically separate Pu, U and fission products, chemicals must be added to the mix – nitric acid, kerosene, tributyl-n-phosphate, among others. The liquid HLW must then be solidified with glass frit to produce solid borosilicate glass. At the former West Valley reprocessing plant, the volume of solidified HLW compared to spent fuel was approximately 25 to 1. This increased volume primarily affects transportation impacts to a repository, but not the volume occupied within a repository; that volume is primarily based on the heat output.

In addition to repository considerations, it is also important to evaluate the amount of other waste produced at the reprocessing stage. Cladding hulls would generally be disposed at a surface landfill, along with other so-called "low-level waste." Process wastes, generally spent chemicals in reprocessing, would also go to a radioactive landfill. At West Valley, 8,000 gallons of process wastes were disposed in the NRC-licensed burial ground. During reprocessing, an organic chemical, such as kerosene and tributyl-n-phosphate, becomes contaminated with Pu and I-129 over time and must be disposed of. In addition, when a reprocessing plant is decommissioned, a portion of those wastes would also go to a radioactive landfill.

Advanced Fuel Cycle

In order to ease the repository burden, or as DOE describes it, to obtain "the full benefit envisioned for the separations process," several modifications must be made at the reprocessing stage. Essentially, the reprocessing stage requires several additional chemical steps, so that long-lived minor actinides (MA), such as Am, Cm and Np, and long-lived fission products, such as Tc-99 and I-129, are removed from HLW. Once these actinides and fission products are removed, they must be managed in some manner. These materials obviously are created in a light water reactor (LWR) faster than they are consumed. Thus, the GNEP plan is to "burn" these materials in a reactor that has a greater neutron flux, probably a sodium-cooled breeder reactor. By "burn" we mean, convert these long-lived radionuclides to shorter-lived radionuclides. We discuss the individual radionuclides below.

Neptunium: After fuel dissolution, U and Pu is associated with the organic stream (kerosene and TBP), and the fission products are associated with the aqueous stream. Np-237 is partly associated with the U/Pu organic stream, but primarily is associated with the aqueous stream. Of all the radionuclides, Np-237 is probably the easiest to recover. For years, the DOE had been recovering Np-237 from spent fuel at the Savannah River Plant, then irradiating the Np-237 rods to create Pu-238, used for heat generators in satellites. Np-237 is therefore one radionuclide that can be successfully transmuted, but at a penalty in an LWR, that is, an LWR must have a greater amount of fissile U-235 and/or Pu-239+241 to compensate for the neutrons absorbed by Np-237. Under fast neutron bombardment, Np can fission; its critical mass is 55 kg. Thus, Np-237 must be safeguarded.

Americium/Curium: However, it is not sufficient to simply remove Np-237 from HLW, in order to reduce the repository burden since Am-241 decays to Np-237. Thus, the GNEP goal is also to separate Am and Cm from liquid HLW at the reprocessing stage. The separation efficiency must be on the order of 10,000, to sufficiently reduce the repository hazard.[10] This separation efficiency is not presently possible. Once separated, these minor actinides cannot simply be placed into an LWR,

[8] *Ibid.*
[9] Claiborne (1975).
[10] IAEA (2003)

without simultaneously increasing the fissionable materials (U-235 and Pu-239 and Pu-241). The GNEP goal is therefore to place these actinides into a fast flux reactor, and transmute them to shorter-lived materials. This part of the process becomes quite murky. The only presently viable method is to store and dispose of specially prepared Am/Cm. That is, it does not appear that this part of the repository burden problem has been resolved, even in DOE's most optimistic view.

Technetium: Tc-99 has a long half-life and needs to be removed from HLW. A great deal of Tc-99 is created in an LWR, approximately 26 to 27 kg/GWe-yr, or 1.2 kg/MT[11]. Soluble TcO_4^- can be extracted from liquid HLW, but the insoluble Tc component cannot be easily separated. This radionuclide can be transmuted to shorter lived radionuclides, but its small cross-section requires high neutron fields. Transmutation requires a breeder reactor.

Iodine: I-129 has a radiotoxicity comparable to the actinides. It occurs in liquid and gaseous forms in a reprocessing plant. About 7.1 kg/GWe-yr is produced in an LWR. At a reprocessing plant, I-129 is generally captured as a gas or particulate on silver zeolite beds, or contaminates the organic solution and is disposed of in a landfill. I-129 cannot be easily transmuted to shorter-lived materials because of its small cross-section. The GNEP plan for I-129 is similarly murky. IAEA says that it can be disposed in the ocean.[12] It also can be disposed of in a HLW repository, but this defeats the purpose of separating I-129 from HLW.

Assured Fuel Supply

Part of the purpose of the GNEP plan is to create a system where the United States would be the world-wide supplier of fuel for civilian nuclear reactors, providing foreign countries foreswore reprocessing spent fuel and separating Pu. That is, DOE claims a non-proliferation purpose for the GNEP proposal. Foreign policy is far removed from my expertise, but it is inconceivable to me that any foreign government would wish to partner with the United States under the present administration. Essentially a foreign country could be subject to blackmail. Take Iran as one example. This administration has several grievances against Iran, any one of which could lead to a cessation of fuel supply.

GNEP does little for the U.S. fuel supply since it is highly unlikely that recovered U would be re-used because of its low fissile content and the presence of the neutron poison, U-236. The nuclear industry in the United States must rely on foreign suppliers for our nuclear fuel. That is, under the GNEP plan we will continue to need U from outside the U.S.

Effective GNEP Plan

The rationale for the GNEP proposal - proliferation, energy independence, etc. are reassuring code words. But, most likely, the GNEP proposal, because of its cost and its lack of feasibility, will be arrested at an early stage. Here is an alternative scenario. Under GNEP, the Allied General reprocessing plant (AGNS) in Barnwell, SC will be started up. Fuel will be reprocessed; the recovered Pu would be made into MOX at the Savannah River Plant next door. And that will be it. This would allow the utilities to ship their spent fuel off their reactor sites and will allow DOE to resolve their HLW contractual problems with utilities. The HLW will not be shipped to Nevada, but will remain indefinitely in glass logs. This will not solve the waste problem and other rosy promises, but this abridged GNEP will solve the immediate DOE and utility problems. Or, spent fuel will be shipped to AGNS and be stored there indefinitely.

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[11] IAEA (2003)
[12] IAEA (2003)

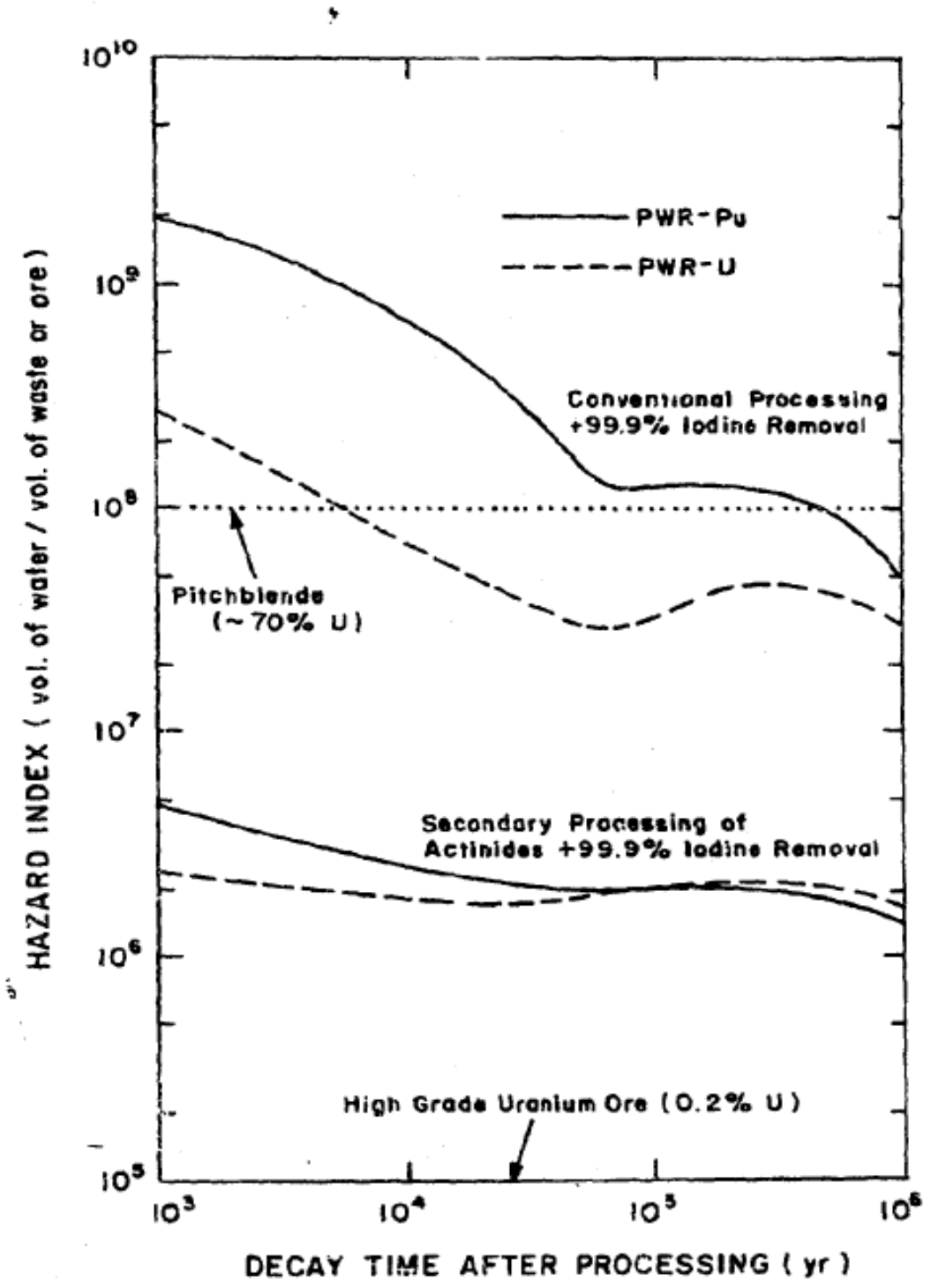

Fig. 1. Hazard of PWR waste as a function of decay time.

Table 1. PWR Spent Type E Fuel

Radionuclides	U only		MOX only	
	kg	%U	kg	%U
U-234	2.99	0.0017	0.36	0.0044
U-235	148.39	0.89	22.61	0.275
U-236	126.03	0.72	2.97	0.036
U-237	0.2	0	0.1	0
U-238	17346	98.4	8204	99.7
Np-237	14.6		0.6	
Np-239	1.47		0.61	
		% Pu		% Pu
Pu-238	5.46	3.3	10.84	3.43
Pu-239	96.22	57.5	110.78	35.1
Pu-240	40.11	24	84.89	26.9
Pu-241	19.11	11.4	60.29	19.1
Pu-242	6.53	3.9	48.97	15.5
Am-241	0.46		3.65	
Am-242	0.02		0.24	
Am-243	1.73		28.7	
Cm-242	0.19		1.31	
Cm-244	0.56		16.1	
Cm-245	0.03		1.78	
Cm-246	0.007		0.17	

From GESMO, NUREG-0002.