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WASTE TREATMENT AND DISPOSAL PROBLEMS OF THE FUTURE NUCLEAR POWER INDUSTRY

F. R. Bruce

INTRODUCTION

Changing emphasis from the production of fissionable material for weapons purposes to the use of fissionable material for power generation is accompanied by a corresponding change in the nature and magnitude of the waste treatment and disposal problems anticipated. These problems stem, first, from the fact that power reactor fuels are designed for extreme corrosion resistance and must be dissolved in highly corrosive reagents prior to chemical processing. Secondly, the rapid growth of the nuclear power industry will result in very large quantities of waste over the long term. Finally, it is desirable to carry out fuel reprocessing, and, if possible, waste disposal operations near the centers of power consumption, which are expected also to be centers of high population density. The purpose of this paper is to assess some of the elements of waste treatment and disposal that are expected to become important in the development of the nuclear power industry of the future.

GROWTH OF THE NUCLEAR POWER ECONOMY

The magnitude of the waste disposal problem of the future depends on the size of the nuclear power economy. For the purpose of this study the estimate of J. A. Lane,⁽¹⁾ shown in Fig. 1, is used primarily because it falls close to the average of all estimates. According to Lane, about 33,000 megawatts of nuclear heat will be installed by 1970 and nearly 300,000 megawatts by 1990. By the year 2000, about 700,000 megawatts will be installed. These figures establish the quantities of fission products that will be produced and that must be stored.

It is expected that natural or slightly enriched power reactor fuels, on the average, will be irradiated to a level of about 10,000 megawatt-days per ton of uranium, at which time they will be reprocessed for recovery of residual fissionable material. The quantity of fuel that must be reprocessed, also shown in Fig. 1, determines the volume of waste produced. In 1970 fuel will be processed at the rate of 3.3 tons per day, in 1990 at 30 tons per day, and by 2000 at 70 tons per day.

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Fig. 1. Predicted Power Growth and Fuel Reprocessing Capacity.

F. L. Culler has described the aqueous processing techniques in use for current fuels and has presented the available information on the newer techniques, including pyrometallurgical processing and fluoride volatility, which may become important in the future. In this discussion it is assumed that fuel dissolution by aqueous methods and solvent extraction of the solution will be used.

COMPOSITION AND QUANTITIES OF WASTE ANTICIPATED

Expected Reactor Fuel Types

The chemical composition and volume of waste produced by the nuclear power industry will be determined by the processing technique used, which, in turn, will be determined by the fuel composition. In this study it is assumed that the power reactors of the year 1980 will fall into the grouping shown in Table 1. Ten percent of the total power is assumed to be produced by breeders and 90% by converters. Of the 10% produced by breeders, 5% is expected to be derived from fast breeders using stainless steel-clad fuel, and 5% from homogeneous breeders. Of the 90% produced by converters, 40 and 25% are expected to come from reactors using zirconium-clad uranium fuels and stainless steel-clad uranium fuels, respectively, 20% from thorium converters using zirconium-clad fuel, and 5% from thorium converters using carbon-clad fuel.

Table 1. ASSUMED DISTRIBUTION OF REACTOR TYPES IN 1980

Reactor Type	Fuel	Percent of Nuclear Power
Breeders		
Fast	Stainless steel-clad	5
Homogeneous	Aqueous	5
Converters		
Uranium	Zirconium-clad	40
	Stainless steel-clad	25
Thorium	Zirconium-clad	20
	Carbon-clad	5
		<u>100</u>

Zirconium-uranium alloy fuels will be dissolved in ammonium fluoride solution and an extraction feed prepared by the addition of nitric acid. The waste from the extraction step will have a high activity level and will contain nitric acid, zirconium, and ammonium fluoride. Stainless steel-containing fuels in which the uranium and stainless steel are intimately

mixed will be dissolved in dilute nitric-hydrochloric acid, and the chloride will be distilled off before the resulting solution of stainless steel and uranium nitrates is solvent extracted. The waste from this extraction step will contain, per liter, about 50 g of stainless steel nitrates and aluminum nitrates, the latter being introduced as a scrub during extraction.

Stainless steel— or zirconium-clad thorium oxide fuels would be declad in the same way as the analogous uranium dioxide fuels, and the thorium and uranium recovered by the previously described Thorex process.

The wastes expected from power reactor fuels differ from current wastes mainly with respect to their content of dissolved stainless steel, zirconium, and fluoride ion, but they will be less amenable to tank storage than are the simpler wastes resulting from present-day processing of aluminum-clad uranium or uranium-aluminum alloy. Present waste disposal practice usually entails neutralization of waste with caustic prior to storage, but solutions of stainless steel, zirconium, and aluminum form voluminous precipitates on neutralization. The high heat generation rate anticipated in power reactor wastes and the presence of precipitates may require special provisions to remove heat and prevent bumping if the wastes are stored in tanks. The fluoride ion causes serious corrosion problems in long-term storage of the wastes.

Composition of Power Reactor Fuel Wastes

Table 2 shows the important constituents in the anticipated wastes⁽²⁾ from the processing techniques that will be used for the recovery of spent reactor fuel. These processes are under development but are ready to be demonstrated on an engineering scale. The techniques now used for the present aluminum-clad uranium and aluminum-uranium alloy fuels have already been described.

In the new techniques stainless steel cladding of uranium dioxide fuels may be removed by dissolution in 4 molar sulfuric acid. The resulting solution will contain 80 grams of iron, chromium, and nickel per liter as the sulfates. Zirconium cladding from UO_2 fuels will be removed in a solution of 6 molar ammonium fluoride—1 molar ammonium nitrate. The solution will be about 0.5 molar in zirconium, 1.5 molar in ammonium, 3.7 molar in fluoride, and 0.6 molar in nitrate. Both cladding solutions will be sent to waste. The uranium dioxide fuel cores from both types of fuel will be dissolved in 16 molar nitric acid and prepared for solvent extraction.

A question which is very relevant to waste disposal, and which is as yet unresolved, is how much of the fission product activity present in the fuels will follow the decladding solution. Present indications are that about 0.1% of the fission product activity will be in the decladding solution. Although the activity level of this solution will be one-thousand-fold less than that of the main fission product—bearing stream, it is still sufficiently radioactive that it may not be released to the environment and will require handling as a high-activity waste.

Table 2. COMPOSITION OF WASTES ANTICIPATED
FROM VARIOUS TYPES OF REACTORS

Fuel Type	Decladding Solution	High-activity Waste
Natural U		
Aluminum-clad U	NaAlO ₂ , 1.2 M	HNO ₃ , 2 M
Stainless steel-clad UO ₂	(Fe, Ni, Cr)SO ₄ , 80 g/liter	HNO ₃ , 2 M
Zirconium-clad UO ₂	Zr ⁺⁺ , 0.5 M; NH ₄ ⁺ , 1.5 M F ⁻ , 3.7 M; NO ₃ ⁻ , 0.6 M	HNO ₃ , 2 M
Enriched U		
Aluminum-uranium alloy		HNO ₃ , 1.3 M Al(NO ₃) ₃ , 1.6 M
Zirconium-uranium alloy		HNO ₃ , 3.2 M Zr ⁺⁺ , 0.8 M NH ₄ F, 1.8 M
Stainless steel-uranium cermet		Stainless steel, 50 g/liter Al(NO ₃) ₃ , 0.12 M HNO ₃ , 3 M

Quantities of High-Activity Waste Expected

Table 3 shows the volume of waste anticipated from the various power reactor fuel types. The magnitude of the future waste disposal problem depends on the direction of fuel development, with the addition of alloying elements increasing the volumes of waste that must be stored, as well as on the chemical processes used to recover the irradiated fuel. In the case of the natural or slightly enriched fuels the volumes of waste to be stored are generally much lower than for the highly enriched fuels. This stems from the fact that in the former fuels, uranium-238, present as a diluent for the uranium-235, is extracted out of the waste stream in fuel reprocessing. In the case of the latter fuels the diluent, a nonradioactive alloying material which is usually present, remains with the waste stream and contributes to the disposal problem. Present indications are that the uranium oxide and mixed uranium-thorium oxide fuels will be widely used. This type yields roughly tenfold less waste volume per unit of heat extracted than an alloy fuel such as that used in the naval reactors.

Figure 2 shows the waste volumes that will be accumulated. By the year 1980, 36 million gallons of waste will have been accumulated. By way of comparison it has been reported that at the Hanford site about 50 million gallons of waste is stored in underground tanks.⁽³⁾ Thus, it appears that even by 1980 the wastes accumulated from the nuclear power industry will be much less than the volume now stored at Hanford.

Table 3. VOLUME OF WASTE TO BE STORED FROM POWER REACTOR FUELS

Fuel Type	Waste Volume, gallons	
	Per kilogram of uranium	Per megawatt-day
Natural or Slightly Enriched		
Aluminum-clad uranium	0.33*	0.033
Stainless steel--clad UO_2	1.06*	0.106
Zirconium-clad UO_2	1.46*	0.146
Highly Enriched		
Aluminum-uranium alloy	125	0.5
Zirconium-uranium alloy	300	1.2
Stainless steel--uranium cermet	65	0.26

* 0.06 gallon is high-activity waste; the balance is decladding.

Fig. 2. Accumulated Volume of High and Intermediate Level Waste Stored.

Prospects for markedly decreasing the volumes of waste resulting from power reactor fuels lie in two areas. First, mechanical processing methods are under development for separating cladding agents from the fuel core. Since chemical dissolution of cladding contributes the largest fraction of the volume to be stored, the mechanical techniques, if perfected, will result in a marked reduction in waste volume. Secondly, methods for converting power reactor wastes to a solid form prior to storage are being studied. In the case of a zirconium-clad UO_2 fuel, which produces 1460 gallons of waste to be stored per ton of fuel, the stored volume could be reduced to 25 gallons per ton by conversion of the waste to a solid. Not only would this process reduce the volume of waste to be stored, but it would also eliminate the bumping and corrosion problems associated with storage of the waste in liquid form. At the same time it introduces new problems of heat removal and evolution of radioactive gases.

The combination of the high toxicity of the fission products and their expected production in very large quantities in the nuclear power industry makes it essential that methods for their containment be absolutely safe. Figure 3 shows the accumulated radioactivity resulting from the predicted nuclear power industry.⁽⁴⁾ A feeling for the magnitude of the hazard potential associated with the waste may be obtained by considering the following. By the year 1980, 10 billion curies of activity will be stored. Eight hundred million curies of strontium-90 will be contained in 36 million gallons of solution.

Gaseous Wastes

Fission product gases appear in the off-gas system from fuel reprocessing plants. The only important one from the standpoint of long-term waste disposal is krypton-85, which has a 10-year half-life. Iodine-131 is by far the most significant when short-decayed fuel is processed. Although processes are currently in use to remove iodine from chemical plant off-gas systems, the present techniques probably do not achieve complete enough removal to permit locating chemical plants in congested areas. Further process development is required in this regard.

Although the hazard associated with krypton is slight, its continued buildup necessitates developing methods for removing it from power reactor fuel reprocessing off-gases. By the year 2000, 700 megacuries of krypton-85 will have been released to the atmosphere.⁽⁵⁾ This quantity mixed with the air surrounding the earth to a height of 10 miles would increase the background by 4.2% assuming that the air background is now 0.015 milliroentgen per hour. While it appears that the release of rare gases to the atmosphere may be permissible during the early stages of the nuclear power industry, it will eventually be necessary to remove the rare gases from chemical plant waste and retain them for decay.

Fig. 3. Accumulated Radioactivity Resulting from Predicted Nuclear Power Industry.

Transuranic Elements (6,7)

The recycle of power reactor fuel will result in the production of transuranic elements which are only less important as hazards in long-decayed waste than are strontium-90 and cesium-137. The most important of these heavy elements are americium-241, plutonium-238, plutonium 239 and 240, and curium-242. The quantities of these elements expected to be accumulated in the waste system by 1990 are

americium-241	4.8×10^5 curies	plutonium-239 and 240	1.2×10^5 curies
plutonium-238	3.3×10^5 curies	curium-242	1.4×10^6 curies

They become important in waste from power reactor fuel because the fissionable material is recycled, with resulting buildup of the higher isotopes. Plutonium-238 is formed by successive neutron capture by uranium-235 and 236, and neptunium-237. Plutonium-239 is used as a fuel itself, and captures a neutron to produce plutonium-240. Americium-241 results from plutonium-240 capturing a neutron and decaying to americium-241. Curium-242 arises from americium-241 capturing a neutron and decaying to curium-242. The quantities of plutonium isotopes shown above assume a 0.1% plutonium loss in fuel reprocessing. The chemical behavior of americium and curium is similar to that of the rare earth fission products, and they appear in the waste stream. Any program aimed at treating wastes for hazards removal prior to discharge to the environment must take cognizance of the transuranic elements as well as the fission products. Since the process chemistry of americium and curium in waste solutions has been relatively little studied, the problem may be a formidable one.

Low-activity Wastes

Fuel reprocessing operations produce large quantities of low-activity wastes. These wastes are now dispersed to the environment in remote areas, but such dispersal will probably not be feasible in heavily populated sections where central fuel reprocessing plants are likely to be located. If these wastes are stored, their large volume will adversely affect the economics of nuclear power. It is hoped that it will be possible to recycle essentially all this low-activity waste within the chemical plant, thereby minimizing the volume to be disposed of.

Distribution of Radioactivity in the Power Reactor Complex

A study has been made of the distribution of accumulated radioactivity over the power reactor complex, which consists of the reactor, decay storage prior to chemical processing, and, finally, waste storage. (5,7) The results show that the waste disposal system will contain the bulk of the long-lived, hazardous radioactivity produced by the power industry. In the case of cesium-137 and strontium-90 expected to accumulate by the year 2000, about 4% will be in the reactors, 7% in fuel decay storage, 1% in the chemical reprocessing plant, and 88% in the waste disposal system.

ECONOMIC IMPLICATIONS OF WASTE DISPOSAL (8-10)

Tank Storage

In order to assess the significance of waste disposal cost in the production of economic nuclear power, let us assume that power is to be produced for 8 mills per kilowatt-hour of electricity and that 2% of this cost, or 0.16 mill per kilowatt-hour, may be allocated to waste disposal. Figure 4 shows the allowable waste disposal cost as a function of fuel burnup and waste volume per ton of fuel. It now seems likely that the expected power reactor fuels will achieve burnups of about 10,000 megawatt-days per ton and that the volume of high-level waste produced will be 1200 to 1300 gallons per ton. Under this set of assumptions, we can afford to spend about \$8 per gallon for waste disposal.

In current practice, tank storage of high-level, neutralized waste from aluminum-clad uranium fuel varies from \$0.40 to \$1.00 per gallon, depending on the tank design.⁽³⁾ Therefore, power cost allocated to high-level waste storage may be about 0.02 mill per kilowatt-hour. It seems apparent that the difference between the current cost of high-level waste storage and the amount which may be allocated to waste disposal, namely 0.014 mill per kilowatt-hour, is more than sufficient to treat and dispose of the low-level waste.

It is obvious that the initial cost of waste storage in tanks does not contribute greatly to the cost of nuclear power. However, since steel tanks have a finite life and will have to be replaced many times, it is not now possible to assign a realistic cost to tank storage as a method of ultimate waste disposal.

Impact of Radioactive Wastes on Fuel Reprocessing Plant Location

The cost of shipping liquid waste from a central fuel reprocessing plant to an ultimate storage site is not prohibitive. However, this practice requires the transit of such large quantities of waste that it raises questions of safety. In order to achieve freedom in locating fuel reprocessing plants in the optimum location, attention must be given to possibilities of converting wastes to a safe form for storage and to safeguarding against atmospheric contamination.

Power reactor fuel reprocessing studies indicate that attaining reprocessing charges which are consistent with economical nuclear power requires a minimum plant capacity of 6 tons of fuel per day.^(11,12) Thus, a central reprocessing plant will service many reactors. In order to minimize fuel shipping costs it is desirable to locate the plant centrally with respect to the reactors. Two sites appear attractive for early location of a fuel reprocessing plant: one on the Ohio River and the other on the Great Lakes, perhaps between Cleveland and Buffalo. Both these areas are heavily populated and the hazard associated with storage of large quantities of fission products has stimulated consideration of waste shipment to permanent disposal in salt beds.

Fig. 4. Allowable Waste Disposal Cost as a Function of Burnup and Process Waste Volume.

The magnitude of the waste shipment problem anticipated in the year 1980 is shown by the following values:

	1980	2000
Number of carriers in transit	505	3,270
Probable carrier weights, tons	6	6
Volume of waste in transit, gallons	227,000	1,470,000
Amount of fission products in transit, curies	1.5×10^8	9.8×10^8

In this study⁽¹³⁾ it was assumed that wastes were shipped a distance of 500 miles after 2000 days' decay, the decay time which minimized the combined decay storage costs and shipping costs. Under these conditions, the total cost for decay storage and shipping 1000 miles was about \$1.80 per gallon, or 0.04 mill per kilowatt-hour of electricity.

CONCLUSIONS

The prospective nuclear power industry will produce large quantities of spent fuel, containing fission products. It is economically desirable to recover the fissionable and fertile material from this fuel in central reprocessing plants which should be located near the centers of power consumption and population. This will necessitate developing improved methods for removing iodine from off-gases, and over the long-term future, krypton-85 also.

Tank storage of power reactor fuel wastes will be complicated by their content of dissolved zirconium, stainless steel, and fluoride ion. The initial cost, 0.02 mill per kilowatt-hour, of storing power reactor wastes in tanks is not economically significant, but this storage method does not represent a permanent solution to the problem. In addition, storage of large quantities of liquid waste near centers of population must be examined from a safety standpoint. The alternative of shipping liquid wastes from the fuel reprocessing site to another location for disposal is not prohibitive costwise, but involves movement of such large quantities of radioactivity that questions of safety are raised.

Research and development on waste disposal methods for the power reactor industry should be concentrated on devising improved methods for decontaminating off-gases and on conversion of wastes to a more suitable form than the liquid for storage. The indications are that, with adequate development, these objectives can be attained at a cost commensurate with economic nuclear power. The disposal procedures resulting are expected to be sufficiently safe to make waste disposal non-restrictive in the location of fuel reprocessing plants.

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