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Development of Radioactive Waste Management at Japan Atomic Energy Research Institute

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1. Introduction

Almost twenty thousand MWe of nuclear power plants are now in operation or under construction in Japan. In addition, a spent fuel reprocessing plant of 0.7 t/d at Power Reactor & Nuclear Fuel Corporation (PNC) is under commissioning test and a second one on a larger scale is scheduled to be built in the latter half of 1980's.

To meet the nuclear power development program, the Japan Atomic Energy Commission (JAEC) has decided a waste management policy both for low-level and high level wastes.

For the low-level wastes which are produced mainly in the reactor operation, the sea disposal would be considered. And this sea dumping operation will be put into practice after the safety of the method is confirmed by the results of an experimental sea dumping which is scheduled to start in 1978. For the high-level wastes, the development of solidification and product storage techniques would be performed by demonstration facilities in the next decade, and research on possible procedures of the disposal into geologic formations should be started to decide a final plan within several years.

At JAERI, development of waste management techniques has been carried out to contribute to the future program along the national objectives, and at the same time to meet the practical needs from the routine operation of waste treatments which has been carried out past 20 years.

2. Treatment of Low- and Intermediate-Level Wastes

Three improved techniques of waste treatment will be described.

2.1 Development of Incinerator with Ceramic Filters

Most of the solid radwastes generated from routine operation of nuclear facilities are, in general, combustible wastes of the low-level category. It is considered to be the best method that these wastes are treated with incineration because of high volume reduction and chemically stable state of the product. An incinerator with the wet gas-cleaning system has some problems, for example, of secondary treatment of contaminated washing water and corrosion of construction materials. In order to avoid these difficulties, a test incinerator (capacity:12 kg/h) with two ceramic filter chambers in series was constructed to obtain quantitative data on decontamination characteristics, pressure drop and after-burning effect on the ceramic filter elements, although an operation experience of an incinerator with ceramic filters was known [1].

Decontamination factor of the filters and retention factor in the furnace (radioactivity input to the furnace/radioactivity output from the furnace) measured for four radioisotopes are shown in Table I. The results indicate that the secondary filter has a better dust collection efficiency than the primary, and decontamination factor of the test incinerator including retention factor was found to be 10^5 - 10^6. Results of the pressure drop and after-burning effect were reported elsewhere [2].

In conclusion, it was proved that the ceramic filter has exellent characteristics with respect to decontamination and after-burning effect. Based on this experiment, a new incinerator (capacity:100 kg/h) will be built in the Tokai Research Establishment within few years.

2.2 Treatment of Laundry Waste

In spite of low radioactivity, the laundry liquid waste arisen from nuclear facilities can be hardly treated by a present waste treatment system because of synthetic detergents. Thus, a reverse osmosis treatment apparatus equipped with cellulose acetate semipermeable membrane (spiral wind type, effective area:8.1 m^2) and a thin layer evaporator (capacity: 0.25 m^3/d) have been tested for the treatment of laundry wastes.

Experiments of reverse osmosis treatment were performed until volume reduction of 1/10 was attained by using solution containing 0.1 wt% of detergent and $4.6 \times 10^{-4} \mu Ci\ ^{60}Co/ml$. Both detergent and radioactivity (^{60}Co) were removed effectively, i.e., 94% and 99%, respectively. Concentrated waste from the reverse osmosis was further treated with the thin layer evaporator. Volume reduction was given to 1/9 in the evaporation step. Consequently, combined use of reverse osmosis and evaporation treatments gave ca. 1/100 as overall volume reduction. Concentrated waste from the evaporator was able to be dehydrated by the high frequency heating treatment. Since the reverse osmosis treatment requires low capital and operational cost, a treatment system (capacity:0.4 m^3/h) is in planning at the Oarai Research Establishment, JAERI.

2.3 Incorporation of Spent Ion Exchange Resin in Plastics

Spent ion exchange resins are now only left stored in tanks at power station sites, and their accumulated quantity at the end of 1975 was more than 200 m^3 and is expect to amount ca. 2,000 m^3 after five years (by 1980).

For the treatment of spent resins, it is at present assumed adequate to solidify them in cement or asphalt. It is known, however, that cement matrix tends to crack or break when they are submerged in water. Moreover, cement matrix can contain only 30 wt% of waste and hence offers little advantage in volume. On the other hand, asphalt matrix are poor in thermal stability, ready to deform and liable to catch fire during the production process or storage. In addition, they are likely to swell and disintegrate in water. In order to obtain more stable solidified wastes, experiments were conducted in incorporating spent resins into polyethylene. A twin helical screw type extruder was used for preparing the products.

The polyethylene composites thus obtained have following properties. The volume of the product containing 50 wt% of resin is about 0.6 times larger than that of original wet resin, i.e., the volume reduction is about 1.7. The uniaxial compressive strength is nearly 300 kg/cm^2 even when it contains 50 - 60 wt% of resin. This strength declines from 300 to 200 kg/cm^2 upon irradiation by γ-rays with a dose of 10^8 rad, beyond which this strength remains constant up to 10^9 rad. Leaching test was also carried out using ^{137}Cs in accordance with the method proposed by IAEA. The leaching ratio for the specimens of polyethylene composite is as small as 0.1 % after a period of one year, which is about 1/1000 of that for the cement matrix.

Thus, the proposed plastic solidification can be considered to be one of the most desirable method so far for immobilizing spent resins.

3. Safety Evaluation of Solidified Wastes for Sea Disposal

In compliance with JAEC's decision, several governmental and public organizations, under the leadership of the Science and Technology Agency, take their share of relevant studies including oceanographic investigations on promising areas for sea dumping in the northwestern Pacific. JAEC showed, at the same time, the Japanese provisional guidelines of monolithic type low-level cement solidified packages for sea dumping. In JAERI, evaporator concentrate — cement composites solidified in 200 l drums have been subjected to high hydrostatic pressure tests and leaching tests under high pressure [2]. Multi-stage type packages have also been examined for their integrity by high hydrostatic pressure tests and for their leachability under ordinary pressure.

3.1 Monolithic Type Low-Level Cement Solidified Packages

Evaporator concentrate – cement composites solidified in 200 l drums were subjected to high hydrostatic pressure up to 500 kg/cm^2. This pressure was attained within about 30 min., corresponding to falling velocity of the package in sea water. In case of the solidified in a sealed drum, no damage was observed both for the solidified and drum, except that the drum was deformed when void existed between solidified and drum. On the other hand, in case of the solidified with some exposed bare surface, the exposed parts of the solidified were cracked. The degree of damage decreased as decreasing the increasing rate of pressure.

Leaching tests of the solidified containing ^{137}Cs or ^{60}Co have been performed under the condition of hydrostatic pressure of 500 kg/cm^2 and temperature of 2°C, which simulated the condition of the proposed deep sea bottom. Results are shown in Fig. 1

The solidified seemed to be intact even in case when its upper part was exposed, i.e., without lid of the drum, due to very low pressure increasing rate. It is estimated from curves in Fig. 1 that the leaching ratio of ^{137}Cs for the first one year is lower than 3×10^{-3} and that of ^{60}Co is 2×10^{-5}. In the case of the solidified in a sealed drum, i.e., a drum with lid, the leaching rate seemed to be suppressed only at the beginning.

In conclusion, the solidified in a sealed drum would remain intact even when it is dumped into the deep sea of 5000 m, and that the rate of releasing radioactivity on the seabed appeares to be very low.

3.2 Multi-Stage Type Waste Packages

Radioactive wastes are packaged in multi-stage concrete containers at the Oarai Research Establishment, JAERI. Low-level wastes solidified with cement, concrete or asphalt, and compacted rags are packed into 200 l steel drums lined with reinforced concrete. The container is sealed with reinforced concrete pouring onto each top. Packages are likely to be consistent with the guidelines prepared by OECD/NEA in 1974.

Three packages in 200 l steel drum with 50 mm concrete liner contained a compacted lump of contaminated wet rags and vinyl films, etc. were subjected to leaching test under ordinary pressure. Description of these packages and observed saturated leaching ratios are tabulated in Table II. The leaching were found to saturate after about 200 day dipping into tap water at ambient temperature.

Radioactive analysis of the leachant was tried with Ge(Li) detector, but could not identified any nuclide due to a very small quantity of activities leached.

4. Management Technique for High-Level Wastes

At present, studies on management technique for the high-level wastes at JAERI have two main objectives. One is safety evaluation of solidified products in storage and disposal conditions, and another is partitioning of the waste to some fractions of fission products and actinoids.

4.1 Solidification

Selection of solidification matrix is important in relation to both the safe operation during solidification and the product durability. A study on materials for vitrification was carried out using natural zeolite, which is found in Japan. This method would be expected to have advantages of (1) suppressing volatization of radioactive components due to adsorption during the heating, (2) increasing the product durability owing to the effective ingredients such as Al_2O_3, TiO_2, and (3) a good use of natural resources. The zeolite used contains clinoptilolite and mordenite over 50 wt%, and its cation exchange capacity is 100-150 meq/100 g. The laboratory scale tests showed that the matrix made of zeolite and other additives such as Na_2CO_3, H_3BO_3, could contain up to 25 wt% waste oxides in melting at a temperature of 1200°C.

Various solidified products were prepared for the characteristics evaluation by use of an bench-scale apparatus, which are composed of rotary kiln calciner (feed rate: 10 l/h), a vitrifying melter with an induction heating (1.5 l glass/batch) and a hot press (0.2 l ceramics/batch).

The results of leaching tests on the products with powder boiling method have proved that higher operating temperature and higher content of B_2O_3 decrease leachability of cesium and the tendency becomes prominent over 1250°C and 6 wt%, respectively. It has also been found in heating tests that cesium volatilizes from vitrified products at 950°C two to four times as much as at 800°C. The effects of devitrification and of irradiation (10^{10}rad with electron beams) on the products are now being examined.

4.2 Partitioning of High-level Liquid Wastes

Partitioning of the wastes into some fractions of fission products and actinoids, according to their half-lives or radiological toxcities, will greatly assist the rationalization of management schemes for highly radioactive liquid wastes. Furthermore, it will open the gates to study the new possibility such as a nuclear burning of the hazardous nuclides or to set up some utilization scheme of ^{90}Sr, ^{137}Cs, ^{147}Pm and so on. However, the partitioning scheme should be the one which does not increase the varieties and amounts of waste.

In a developed scheme, most of platinum element, zirconium etc. was first removed as a deposit during the acidity adjusting treatment from 1 - 2N to ∿ 0.1N. Rare earth elements (RE) accompanied with transuranum elements (TRU) were extracted from the 0.1N nitric acid solution with normal paraffin solvent containing di(2-ethylhexyl) phosphoric acid (DEHPA). Strontium-90 was successively extracted with the same solvent after adjusting the aqueous phase to pH5. Cesium-137 was then collected on zeolite column remaining the rest of components in aqueous phase.

During the procedure, strict attention was paid to avoid the generation of new kinds of wastes or increase in the amount of wastes; for example, each fraction was separated with the same kind of reagents only by changing the separation condition, the adjustment of acidities was proceeded with formic acid instead of ordinary neutralization method, and so on.

The separated fractions were further devided into several parts if necessary. In case of (RE + TRU) fraction, further partitioning was achieved with di-ethylentriaminepentaacetic acid (DTPA) by using pressurized cation exchange column system. The exchanger was a newly developed porous type having a rapid exchanging speed, and was resistible against irradiation up to 2.5×10^8 rad. Promethium-147 could be isolated from the rest of RE and TRU in a satisfactory purity.

A bench scale experimental unit has been installed in a shielded containment, consisting of a denitration and evaporation vessel, a mixer-settler bank and ion-exchange columns, and the cold test by suing simulated waste has defined that this system may work well in about 1 kCi level hot run.

REFERENCES

[1] Bähr, W. et al., Die Verbrennungsanlage für radioaktive Abfälle des Kernforschungszentrums Karlsruhe, KFK 2300 (1976).

[2] Machida, C. et al., Developments and Studies for Sea Disposal of Radioactive Wastes, International Symposium on the Management of Radioactive Wastes from Nuclear Fuel Cycle, IAEA, Vienna (1976).

Table I. Decontamination Factor of Incinerator

Nuclide	Retention Factor	D.F. of Ceramic Filter			Total
	(a)	Primary (b)	Secondary (c)	(b)x(c)	(a)x(b)x(c)
^{137}Cs	4.1	1.7×10^{2}	2.5×10^{2}	3.9×10^{4}	1.6×10^{5}
^{85}Sr	9.3x10	5.5x10	1.0×10^{3}	5.5×10^{4}	5.1×10^{6}
^{58}Co	4.8x10	4.5x10	2.1×10^{2}	9.5×10^{3}	4.5×10^{5}
^{32}P	1.9x10	3.7x10	1.1×10^{3}	7.6×10^{4}	7.6×10^{5}

Table II. Leaching Test of Multi-Stage Packages

No.	Main Nuclide	Estimated Radioactivity (mCi)	Surface Dose Rate (mR/h)	Leaching Ratio	
				β(γ) Measurement	γ Measurement
1	^{60}Co, ^{54}Mn, ^{137}Cs	28	79	3.3×10^{-6}	2.0×10^{-5}
2	ditto	24	58	7.0×10^{-7}	3.3×10^{-5}
3	ditto	29	66	1.9×10^{-6}	5.8×10^{-5}

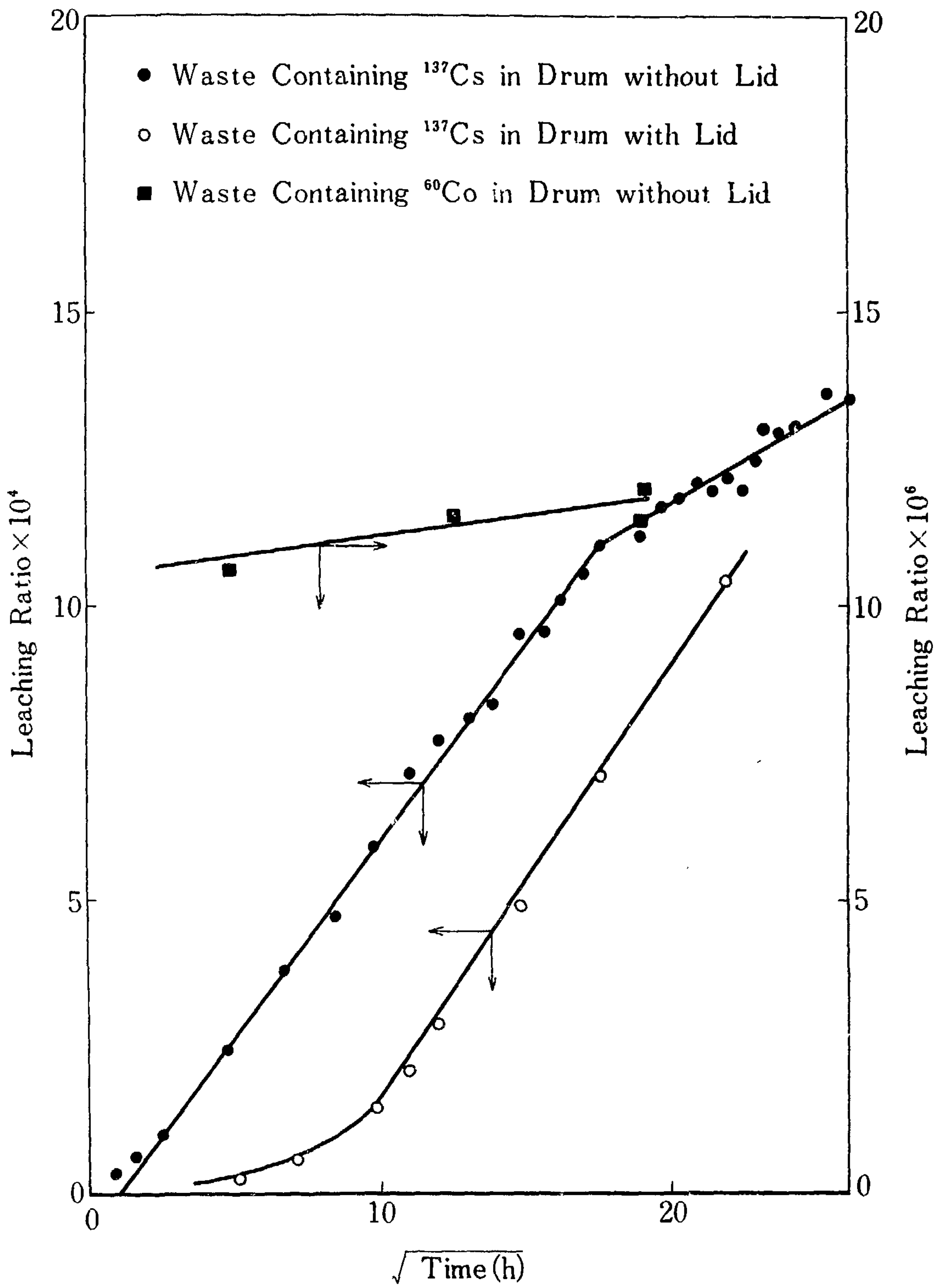

Fig. 1. Leaching under High Pressure from Low-Level Waste—Cement Composite Solidified in 200 l Drum