ACCEPTANCE CRITERIA AND THEIR EVALUATION TECHNIQUES FOR SOLIDIFIED WASTE FORMS

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ABSTRACT

General acceptance criteria and their assessmemt technology for the disposal of low- and intermediate-level wastes have been studied. The test and inspection items for solid waste forms were collected, analyzed and then major items with appropriate testing procedures were established. The corresponding Korean Standards were selected for the indigenousness of testing procedures.

Considering the testing results for the solidification of concentrated liquid waste and spent ion exchange resins coming from the nuclear power plants as well as the simulated ones, criteria for the compressive strength and leachability are suggested. These criteria would be utilized for future application to the operation of the Korean repository following confirmation and authorization by the regulatory body, Korea Institute of Nuclear Safety.

INTRODUCTION

In Korea, the Korea Atomic Energy Research Institute (KAERI) has dedicated its resources to the articulation of a plan to begin operation of a land disposal repository in 1996. This has been undertaken following the designation of the institute by an ammendment to the Atomic Energy Act in 1986 to the goal of accomplishing national radioactive waste management.

There are two major radioactive waste producers: nuclear power plants and radio isotope (RI) users. With nine nuclear power plants (NPP) in operation, Korea had accumulated approximately 23,000 drums of solid radioactive wastes by the end of 1989. Out of the nine nuclear power plants, eight plants are pressurized water reactors and one is the CANDU reactor. From 467 RI users, about 310 drums of radioactive wastes were produced in 1989.

In general, waste characteristics and site specific conditions are to be considered in the acceptance criteria for the disposal of radioactive wastes. In 1990 we experienced severe public objection to the geological investigation of candidate diposal sites. Due to this public opposition, the acceptance criteria based on site specific information are not covered here and the final disposal site has not been selected yet.

STATUS OF RADWASTE MANAGEMENT

Classification of Wastes

The methodology of classifing radioactive wastes for the purpose of transportation and packaging has been established in Korea. In the USA, low-level radioactive wastes for disposal are classified into three classes, that is, A, B and C. IAEA suggests five categories for the disposal of wastes. But the specific classification of waste for disposal has not been made yet.

Acceptance Criteria for the Solidified Waste Forms

In view of the necessity of quality control for disposal, general acceptance criteria of waste forms for disposal are required. Among several kinds of wastes to be disposed of, solidified wastes, which occupy about 40% of solid waste from nuclear power plants, are mostly considered in the evaluation of the proposed acceptance criteria. The process acceptance procedure for treatment, packaging and transportation, general acceptance criteria for disposal, and site-specific acceptance criteria are also needed to assure the QA/QC of the disposal of waste.

Quantity of Waste Produced

By the end of 1989 Korea had accumulated about 23,000 drums of solid radioactive wastes at nuclear power plant sites, of which about 40 percent were cemented liquid wastes as shown in Table I. For the burnable wastes which account for another 40 percent, a plan to incinerate them has been made and is under development[1]. Last year KAERI started to accept radioactive wastes produced by RI users. Table II shows the sources and quantity of radioactive wastes from RI users in 1989.

Radioactivity in Cemented Waste Form

The content of radioactive nuclides in the final safety analysis report (FSAR) for NPP Yonggwang 1 & 2 is compared with that by analysis as shown in Table II. The actual activity shows lower values owing to their short operation period of about two years. The main component of liquid concentrates is boric acid.

The liquid radwaste containing boric acid generated from the PWRs is generally concentrated by evaporation and then solidified. In Korea the waste is concentrated up to 12 wt% boric acid and the concentrate is then solidified with portland cement. As the boric acid strongly inhibits the hydration of cement, slaked lime is added to eliminate this effect. The formulation ratio of concentrated waste :

TABLE I

Solid Waste Stored at NPP Sites

	Type	1989	unit: 200 L drum up to 1989 (%)	
Non-combustible	Concentrate	1,397	9,167	(39.8)
	Spent resin	467	2,076	(9.0)
	Spent filter	89	539	(2.4)
	Others	449	1,893	(8.2)
	Sum	2,402	13,675	(59.4)
Combustible	Paper,PE etc.	2,116	9,346	(40.6)
	Total	4,518	23,021	(100)

TABLE II

Radwaste Arisen from RI Users in 1989

(Unit: 100 L Drum)

Organization(No)		Usage	Drum(%)
Hospital	(89)	Diagnosis Treatment	557(91)
University, Institute	(54)	Tracer	32(5)
Industry, Others	(319)	Luminance Survey	26(4)
Total(419)			615(100)

cement : lime is 39 : 55 : 6 by weight. The density is 1.78 g/cm^3. The compressive strength is about 20 MPa as shown in Fig. 1[2]. Table III shows the content of radionuclides in a 200 L drum[3].

Indigenization of Testing Methods

There are a variety of testing procedures and properties to be considered for the characterization of solidified waste forms. Testing all the test items is not necessary to confirm the waste forms. To ascertain properties of solidified forms, each requirement of handling, storage, transportation and disposal should be considered and the testing items should be selected according to the importance of the testing items[4].

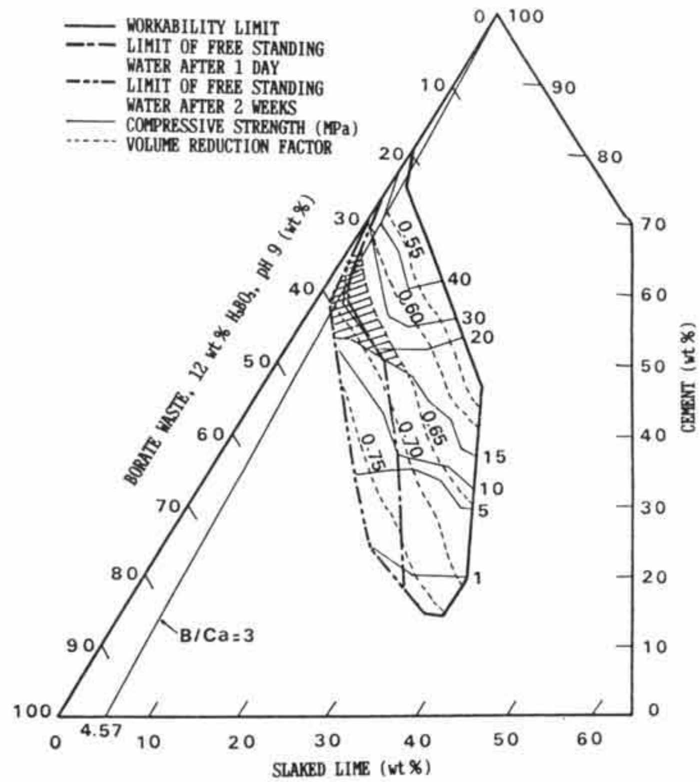

Fig. 1. Three-phase diagram of cement, slaked lime and borate waste (12 wt%).

Testing methods which can confirm the satisfaction of acceptance criteria easily should be selected. Properties such as physical, mechanical, thermal, temperature, chemical, water resistance, radiation resistance, biodegradation and others are considered. The testing methods should be standardized to compare with each other. Indigenization of the testing method is also important. After reviewing foreign standards, Korean Standards are selected. Destructive and non-destructive methods for analyzing radionuclides in the waste forms are excluded.

Solidified Waste Form Test Facility

The Solid Waste Form Test Facility is a *facility to test* and evaluate characteristics of waste forms, such as physicochemical properties, mechanical properties, thermal properties, water resistance and leachability. Such characteristics of solid waste forms are strongly required to meet conditions for long term storage or for final disposal of wastes[5].

At this time, efforts to discover the most effective management methodology for the radioactive wastes generated from NPPs and RI users are being conducted in this facility.

In addition, overall technical standards for test and inspection of solid waste forms such as the standardized

TABLE III

Comparison of Radionuclides Concentration in the Evaporator Concentrates

		(NPP Yonggwang 1 & 2)	
Radionuclides	From PSAR kBq/cc	Analyzed by MCA kBq/cc	After Cementation MBq/drum
Cs-134	5.2	0.104	12.8
Cs-137	35	0.112	13.8
Mn-54	0.11	0.015	1.85
Co-58	4.1	0.026	3.20
Co-60	0.56	0.0074	0.91
Nb-95	0.13	0.00825	1.01
Ag-110M	0.31	0.00844	1.04

Formulation ratio: concentrate/cement/additives = 39/55/6 (wt%) Density after cementation : 1.78 g/cm^3 Density of concentrate : 1.122 g/cm^3 Package efficiency : 95% Volume of concentrate in a drum: 210 L x 1.78 x 0.95 x 0.39/1.122 = 123 L

equipment and processes in the facility will be estabilished in the beginning of 1990s.

GENERAL ACCEPTANCE CRITERIA FOR DISPOSAL

Testing items should be selected to get early results and also to be effective and economical. To obtain results rapidly and precisely, discussion on the selection of assessment and testing procedures should be carried out. Based on the properties to be evaluated, test items are selected. Based on these test items, test methods are discussed and as a result criteria are set up and suggested to be employed as a general acceptance criteria for the disposal of radioactive wastes.

Major Categories of the Solidification Media

The acceptance criteria and test items vary in accordance with solidification media, weather, and the general disposal policy applicable to each country. The solidification media is affected significantly according to the properties of waste form. Test items should be introduced following a consideration of the characteristics of waste forms.

There are many stabilization and solidification techniques to treat radioactive wastes prior to disposal. According to the solidification media they can be divided into four groups as follows:

- Cementation,
- Bituminization,
- Plastic solidification,
- Vitrification and ceramic solidification.

The importance of waste form characteristics should be considered for each of the four groups as regards solidification, interim storage, transportation and final disposal. Main assessment items for each group are discussed in the literature [6].

In Korea the cemented waste forms are produced at nuclear power plants and KAERI uses asphalt to treat low level liquid waste. KAERI has done some research work on plastic solidification [1,7].

Selection of Test Items

Table IV shows test items chosen for the assessment of the general acceptance criteria. The chosen test items are explained with discussion for each of the characteristic property of solidification method.

The selection of criteria are based on the properties that should be assessed and the point of time when the test is done for the received wastes.

Physical Properties

Physical properties relevant to disposal are homogeneity, porosity, water content, dimensional deformation and free liquid. Of these properties, the change of homogeneity or deformation affects the mechanical properties of the waste forms. Density is useful for the confirmation of homogeneity and packing efficiency in land disposal. On the other hand, in ocean disposal the density should be over 1200 kg/m^3 in order to assure sinking to the bottom.

Waste forms containing free-standing liquid, which is drainable liquid, are not acceptable for disposal. Free liquid usually accelerates corrosion of steel drums and consequent radionuclide leakage.

ANS 55.1[8] specifies method of testing free liquid. For stable wastes in a package the quantity of acceptable free-standing liquid is less than one volume percent. Although the quantity is also applicable to the high integrity container, no free standing water is preferred for the solidified waste forms. In practical terms that is less than 0.5 volume percent and the acceptable pH of the liquid is between 4 to 11, which should be suitable for cement waste forms. In the French criteria no water is squeezed out under the pressure of 0.35 MPa, which is suitable for asphalt or plastic waste forms. Visual inspection followed by checking free standing liquid limit of 0.5 volume percent is suggested as test criteria.

As there is no standard exists in KS for free-standing liquid testing, ANS 55.1 is suggested for use. Prior to a

TABLE IV

Proposed Criteria with Related Standards

Items	Related Standard	Testing Methods	Criteria
Free standing water	ANS 55.1		No water (<0.5 vol%)
Compressive strength	KS L 5105/ KS M 3816		Compressive strength->5MPa
Water resist. -Immersion test		Immersion into water depth of 10 cm	After 100 days -No swelling, crumbling -Variation of compressive strength <20%
-Leaching test	IAEA standard	Leach rate to 100 days	$\beta, \gamma < 1x10^{-10}$m/s $\alpha < 2x10^{-12}$m/s
Thermal cycling	KS F 2456/ KS L 5105/ KS M 3816	24h for 1 cycle +10°C : 2 h +40°C : 10 h +10°C : 2 h -20°C : 10 h	After 30 cycles -Variation of compressive strength <20%
Irradiation test	KS L 5105/ KS M 3816	γ-source, Co-60 rate:10^3-10^4Gy/h total:10^6 Gy	Variation of compressive strength <20%
Biodegradation	KS A 0702/ KS L 5105/ KS M 3816		-No growth -Variation of compressive strength <20%

boring test to find drainable liquid, confirmation of no free liquid while testing compressive strength is preferable.

Mechanical Properties

Compressive strength, tensile strength, impact resistance, shock resistance, stackability and durability are the important mechanical properties in disposal of solidified waste forms. The compressive strength measures the structural stability to maintain its original form and dimension under the disposal conditions. The compressive strength is important to the cemented waste form and it is easy to measure. On the other hand, quantitative measurement of stackability by measuring the degree of deformation is not easy.

Each country has its own standard for testing compressive strength in relation to civil engineering. Korea has its Korean Standards for compressive strength, KS-L-5105, KS-M-3816, and KS-F-2351 for cement, plastics and asphalt, respectively. A cylindrical specimen, with a ratio of diameter to length at 1.5, is suggested.

The acceptance criterion for the compressive strength is 60 psi(0.41 MPa) in the United States, which considers stackability at a shallow land disposal site. In Sweden and Italy it is 50 kg/cm^2(4.9 MPa). The limit of compressive strength for the German disposal site, Konrad, is 10 MPa. Considering ocean disposal in Japan and Taiwan it is 150 kg/cm^2(14.7 MPa), while it is 15 kg/cm^2(1.47 MPa) for shallow land disposal. For the French disposal site La Manche it is 30 MPa and 8 MPa for cement and plastic waste forms, respectively.

In Korea, research on the process control program of the cementation process to solidify the evaporator concentrates from PWR resulted in a limit being set at 15 MPa as shown in Fig. 1. Considering the different results between batch and continuous mixer and the failure in immersion test in the case of the cemented form of ion exchange resin, whose compressive strength is less than 5 MPa [7], the limit is suggested to be 5 MPa. For asphalt products the compressive strength test is excluded.

Thermal Properties

Thermal criteria for waste forms should indicate fire safety during handling and storage or under the weather conditions of disposal site. Stability to weathering could be shown by consistency of mechanical properties, usually compressive strength, and no deformation after simulated accidental conditions.

Generally the test conditions for fire accident during storage and handling are suggested as 30 minutes at 800°C in IAEA transportation accident test procedure. The probability of the accident is very low in comparison with that during transportation. Stability on the variation of temperature and weather in the repository area is more important. Flash point, softening point, burning point, thermal conductivity, thermal expansion, specific heat and gas releases are the items which can be measured during research for integrated waste forms. For receiving waste, a thermal cycle test, which measures the effect of temperature and weather by simulated conditions, is more important.

Korean Standard KS-F-2456 "Test method for freeze/thaw resistance of concrete" was chosen for use. Considering that the period of time to raise the temperature at the center of real wastes to the ambient temperature takes more than 6 hours, a resident period of 10 hours at high and low temperatures and for raising or decreasing temperature set to medium temperature for 2 hours each are set for one cycle, and a 30-cycle experiment is chosen. Considering the temperature in Korea, high, medium and low temperatures are chosen to be 40, 10 and -20°C, respectively. The variation of compressive strength less than 20% after 30 cycles is suggested as a criterion.

Water Resistance

Chemical reaction, decrease of stability, gas generation and corrosion by contact with ground water and soil should be considered for long term storage at disposal site, which takes into consideration the chemical stability along with environmental conditions of disposal site.

Ground water affects the rate of nuclides leaching from waste forms. Water resistance can be divided into leach characteristics and water immersion stability. Leaching is diffusional mass transfer of radionuclides from solidified forms by contacting with ground water. Immersion stability is the dimensional consistency of waste forms against water absorption.

Immersion stability is especially important for solidification of ion exchange resins. Immersion in water at a depth greater than 10 cm for more than 4 weeks is preferable for the test. Considering the leaching test, immersion for 100 days is suggested. The criteria are no swelling, no crumbling and less than 20 % of compressive strength variation in comparison to the compressive strength before immersion.

Standard method of leach test was raised by Hespe for the intercomparison of waste forms in 1971 as IAEA recommendation [9]. Other test methods are compared by Han et al. [3]. Besides IAEA recommended method, ISO 6961 [10] of France and ANS 16.1 [11] of the United States are well known. IAEA standard method is recommended. Limits of leach rate less then 1×10^{-10} m/s and 2×10^{-12} m/s for beta and gamma nuclides, and alpha nuclides are recommended, respectively.

Effect of Irradiation

Waste forms should maintain their stability for 300 years, which is a period of management. For the cumulative exposure (in the United States 10^6 Gy, in France 10^5 Gy) the waste forms should not show swelling, gas generation, evaporation of volatile component and deterioration of properties. For the organic media this test has some effects.

We have no experience in radiation exposure test. The test method is chosen to prevent the specimen from melting and not to cause increase of the specimen temperature over 80°C [12]. In France the exposure rate is 2×10^2 Gy/h[12] in the acceptance criteria and that is 5×10^2 - 5×10^3 Gy/h in the standard test method. Cumulative exposure of 1×10^6 Gy by Co-60 with exposure rate of 1×10^3 - 1×10^4 Gy/h is recommended [13]. During the test no gas generation and less than 20% of compressive strength variation after the radiation exposure is the criterion.

Biodegradation

Even though synthetic polymers are often not biodegradable, as they cannot be easily absorbed into microbial cells, biodegradability of organic resins has to be considered for long-term storage and disposal. Physical destruction, mobility increment of radionuclides and gas generation by biological decomposition can affect the stability at the disposal site. Thus safety tests due to microbe, fungi or bacteria should be assessed.

KS-A-0702 "Fungi resistance testing method" can be used for testing the effect of microbe, fungi and bacteria. In the case of positive growth ASTM G21[14], G22[15] can be used. When symptom of growth is found, by using method of Bartha-Pramer[16] testing should be done for more than six months to measure the biological growth rate. The decrease of carbon quantity by biological growth should be less than 10% for 300 years by extrapolation to the real waste form[17].

After testing by KS-A-0702 to check negative biological growth, measurement of the compressive strength to see the variation less than 20% is recommended.

Others

Up to this point, the assessment methods of waste forms have been described for the case where wastes are received at a disposal site. The radioactivity content should be measured and controlled by a destructive sampling method or nondestructive direct assay method [3]. For the operation of repository the period of testing should be shortened, for example, less than one week. A preliminary acceptance [18], which checks free liquid, compressive strength and the results from the derived short period water immersion and leach test, is done regularly and to receive a report on freeze/thaw test, long period leach test, radioactivity resistance and biodegradation is useful. In addition, safety should be assured not only on the long term storage and disposal but also on the radwaste treatment facility, interim storage, and transportation [19]. A confirmation of the process acceptance procedure for final disposal should be made.

CONCLUSION

Based on the literature survey and research results the general acceptance criteria for solidified waste forms, which should be checked at a disposal site, are suggested as shown in Table IV. After the assessment and thereby supplement of the criteria by the Korea Institute of Nuclear Safety, a regulatory body, the criteria will be used at Korean disposal sites with a supplement of site specific criteria.

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