Vitrification Technology for Treating Low-Level Waste from Nuclear Facilities

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The development of technologies for treating nuclear waste generated by nuclear power plants and reprocessing plants during their operation or decommissioning is underway both in Japan and abroad. Of the many types of treatment technologies that have been developed, vitrification technology is attracting attention as being the most promising technology for converting such waste into a stable state. As a brief review of technical developments aimed at reducing nuclear waste and finding a solution to the final disposal issue, this paper describes approaches to completing the development of vitrification technology in Japan, including IHI's activities.

1. Introduction

Many different gaseous, liquid and solid wastes produced from the operation or decommissioning of nuclear power stations, nuclear fuel reprocessing plants (hereinafter referred to as reprocessing plants), and the like are treated or disposed of rationally, depending on the types of their radioactive components or other properties. **Figure 1** illustrates a nuclear fuel cycle and **Fig. 2** illustrates treatment processes for radioactive waste from the operation of reprocessing plants.

High-level radioactive liquid waste (hereinafter, "high-level liquid waste") produced from the reprocessing of spent fuel is characterized by high levels of radioactivity and corrosiveness. For this reason, vitrification technology, which has the ability to stably confine radioactive substances for long periods of time, has been adopted and put to practical use for the treatment of high-level liquid waste inside and outside Japan.

IHI has been engaged in the construction of facilities involved in technology development for the high-level liquid waste vitrification process and for the storage of vitrified waste produced from this process.

On the other hand, low-level radioactive waste (hereinafter referred to as low-level waste) produced at nuclear power stations, reprocessing plants, and the like is characterized by a wide range of radioactivity levels, compositions/ ingredients, among other things, unlike high-level liquid waste. Therefore, various technologies for the treatment of low-level waste are studied and introduced depending on the characteristics of the waste. In Japan, vitrification technology for low-level waste has not yet been introduced, and the following three techniques are commonly used — (1) incineration, (2) compression, and (3) cement

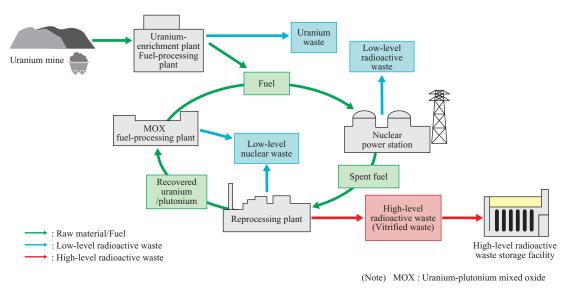
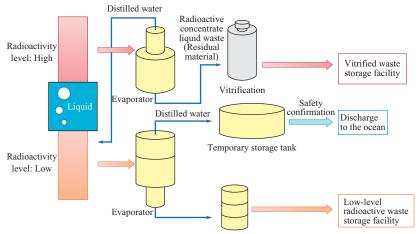


Fig. 1 Nuclear fuel cycle



Volume reduction and solidification

Fig. 2 Treatment process for nuclear waste from reprocessing plants

solidification. In other countries, however, vitrification technology has been introduced for low-level waste, including the following:

(1) AREVA (France)

Low-level liquid waste with relatively high levels of radioactivity from the decommissioning or decontamination of reprocessing plants

(2) U.S. Department of Energy (DOE)

Low-level liquid waste with relatively high levels of radioactivity that is kept in storage at national laboratories

(3) Korea Hydro & Nuclear Power (KHNP) Low-level waste from the operation of nuclear power stations

In light of the aforementioned situations in other countries, IHI believes that vitrification technology is also applicable to low-level waste produced in Japan, and has decided to start developing the technology.

2. Application of vitrification technology to low-level waste produced at nuclear power stations, reprocessing plants, and the like — Technology development activities in Japan —

In the future, Japan will also need treatment technology for low-level waste with relatively high levels of radioactivity (e.g., radioactive liquid waste from decontamination), which will be produced when a nuclear power station, reprocessing plant, or the like is decommissioned. Hence, it is important that we establish a foundation for vitrification technology for low-level waste beforehand in order to reduce radioactive waste (hereinafter referred to as waste), solve the final disposal problem, and more.

One movement reflecting the aforementioned situation is the "basic research programs for the next generation vitrification technology" (hereinafter referred to as the basic research programs), which have been implemented since FY2014 as commissioned projects of the Agency for Natural Resources and Energy of the Ministry of Economy, Trade and Industry.

In the basic research programs, the following activities are being carried out: ① the development of the foundations of vitrification technology for transforming low-level waste produced at nuclear power stations, reprocessing plants, and the like into stable solidified waste with a substantially reduced volume, a technology that has not yet been developed; and ② a study on upgrading the vitrification technology for high-level liquid waste that is in practical use in Japan (achieving higher waste loading and enhancing operation control technology) by applying the findings from the development of the foundations.

Participants in the basic research programs are four entities constituting the joint venture entrusted with the program — IHI Corporation, Japan Nuclear Fuel Limited, the Japan Atomic Energy Agency, and the Central Research Institute of Electric Power Industry, which are well-established in the field of vitrified radioactive waste and research, development and testing of it, as well as those entities to which the program is recommissioned from the joint venture — research institutes and companies in Japan specialized in glass, steel, nuclear and other relevant industries, as well as research institutes inside and outside Japan specialized in vitrification. In other words, the programs contain the collective wisdom of experts inside and outside Japan.

Section 2.1 and later describe the outlines of approaches pertaining to low-level waste in the basic research programs.

2.1 Development objective

The objective is to implement the following development for low-level waste produced at nuclear power stations, reprocessing plants, and the like.

(1) Glass composition development

Techniques for selecting a glass composition will be established for waste that is difficult to treat using the treatment technologies currently being considered and waste for which vitrification technology would be able to outperform the treatment technologies currently being considered in terms of reducing the volume of waste.

(2) Development of operation control technology

Various glass compositions will be selected for various wastes. Then, melting methods will be considered from the perspective of suitability for such glass compositions.

2.2 Concept of applying vitrification technology to low-level waste

IHI is seeking to apply technology called "fused glass solidification technology" to low-level waste. As defined below, this technology applies the conventional vitrification technology. The conventional vitrification technology, that is, the vitrification technology used for the treatment of high-level liquid waste, vitrifies waste by adding a glass raw material, such as borosilicate glass. On the other hand, the fused glass solidification technology vitrifies waste by using a component of the waste (e.g., SiO_2) as a glass-formers, thereby minimizing the amount of additive. The basic research programs utilize this technology that IHI developed in its independent research. **Table 1** lists the features of the fused glass solidification technology.

The fused glass solidification technology makes it possible to adjust volume reduction, operability, and the stability of solidified waste in accordance with the business operator's needs or disposal method as well as to combine one waste with a different waste to transform them into a stable solidified waste. Wastes to which the fused glass solidification technology could potentially be applied effectively are selected based on the following objectives.

- (1) Objective I
 - To stabilize (improve the water resistance of) waste to which the currently considered treatment technology is difficult to apply

Example: High dose ion exchange resin, which must be mineralized

- (2) Objective II
 - To outperform the currently considered treatment technology in terms of reducing the volume of waste
 - Example: Ash containing SiO_2 , CaO, or the like as the chief component

For example, results obtained from a trial calculation of the volume of solidified waste produced from the treatment of 0.3 t of ash indicate that the fused glass solidification technology is able to reduce the volume of solidified waste to approximately 1/4 the volume of solidified waste from cement solidification. **Figure 3** shows an example assessment of the volume reduction effects of vitrification.

2.3 Survey of waste produced from nuclear facilities

In order to carry out the development of vitrification technology for low-level waste, the following procedure is adopted. First, business operators are interviewed about low-

Item	Vitrification	Malt solidification technology		
	Vitrification (conventional)	Fused glass solidification	Melt solidification technology	
Brief description	Method in which vitrification is performed by adding a predetermined glass raw material (e.g., borosilicate glass) to waste	Method in which vitrification is performed by using an component of the waste (e.g., SiO_2) or the like as a glass-formers, thereby minimizing the amount of additive	Method in which waste is melted and solidified as slag	
	Waste + Glass raw material Welting Vitrified waste	Waste + Additive Melting Vitrified waste	Waste Melting solidification	
Volume reduction		0	0	
	Volume reduction is low because a constant waste loading factor is maintained by adding a glass raw material.	Volume reduction is relatively high because the amount of additive is minimized to the extent that stability can still be maintained. However, volume reduction varies depending on the waste composition.	Volume reduction is high because no additive i used.	
Operability	\bigcirc	0		
	Heating and pouring conditions are fixed (do not vary among melting operations) because the properties of fused glass at high temperature can be controlled to within a fixed range. In cases of vitrification for high-level liquid waste, however, operation is affected by the control of noble metals.	Operation without substantial fluctuations in heating and pouring conditions is ensured because the properties of fused glass at high temperature can be controlled to within a predetermined range.	Heating and pouring conditions must be specified per melting operation because the properties at high temperature vary depending on the waste composition.	
Stability of solidified waste	0	0		
	Excellent stability is exhibited because the post-vitrification composition is constantly controlled to within a fixed range.	Relatively high stability is exhibited because the amount of additive is minimized to the extent that stability can still be maintained. However, stability varies depending on the waste composition.	Stability is variable because the slag composition depends on the waste composition.	

Table 1 Comparison of vitrification processes

(Notes) 1. Characteristic of low-level wastes

- Many low-level wastes contain a glass-formers such as SiO₂ or Al₂O₂.
- 2. Technology evaluation results
 - \bigcirc : Excellent, \bigcirc : Good, \triangle : Acceptable

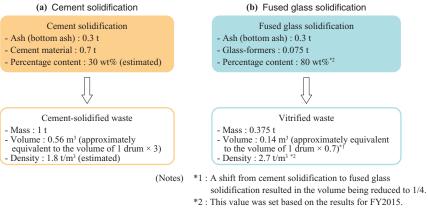


Fig. 3 Comparison between vitrification and cement solidification

level waste produced from nuclear power stations and reprocessing plants. Next, wastes to which the fused glass solidification technology has high applicability are explored and then target wastes are selected for the basic research programs. **Table 2** shows the features of waste from nuclear facilities.

2.4 Glass composition development

In order to define glass compositions (glass matrices) for

various wastes, studies are conducted through the following approaches towards the development of fused glass solidification technology providing excellent volume reduction, operability, and solidified waste stability.

- (1) Approach based on actual results in other countries
- (2) Approach leveraging the know-how of the relevant industries

As mentioned in the previous section describing survey

		Purpose			Solves problem	
No.	Target waste		II	Characteristics	Glass composition development	Operation control development
1	Ion exchange resin	0		 High dose Contains moisture and organic matter. 	O	0
2	Low-level radioactive concentrate liquid waste, radioactive liquid waste from decontamination (Liquid waste with a high sodium nitrate concentration)	0	0	- High sodium nitrate concentration	O	O
3	Low-level radioactive concentrate liquid waste (Phosphate liquid waste)		0	- Contains phosphate ions and a small amount of nitric acid.	O	0
4	Boric acid liquid waste		0	- Contains B, Na, and moisture.	0	_
5	Ion exchange resin eluent		0	- High dose - High S concentration	O	0
6	Ash (including fly ash)		0	 High dose Substantial composition variation 	O	—
7	HEPA filter, Metal (Al) plate, etc.	0		- Contains metal (Al).	—	0
8	Sludge, etc.		0	- High Fe concentration	O	—
9	Asbestos	0		- Chemically stable. Hazardous form.	—	0
10	Toxic metal waste	0		- Contains heavy metals, such as lead and mercury.	0	0
11	Abrasive paper		0	- Made of polypropylene.	—	0
12	Liquid filter	0		- Contains organic matter.	_	0
13	Abrasive for blasting		0	- Al ₂ O ₃ (main component), Si, Fe, etc.	O	_
14	Activated coal		0	- Main component: C		0

Table 2Features of waste from nuclear facilities

(Notes) Objective I : To stabilize waste that is difficult to treat using the currently

considered treatment technology Objective II: To outperform the currently considered treatment technology in

terms of reducing the volume of waste

: Waste to be tested in the basic research programs

: Waste to be tested in the basic resea
 : Target waste suited to the objective
 : Technology development is require

: Technology development is required to solve problems.

eropinent is required to some problems.

results, low-level wastes vary from each other in composition and properties. It is therefore necessary to define a glass composition according to each waste. For this reason, glass compositions are predicted (candidate glass compositions are predefined) in accordance with waste compositions by utilizing the existing international glass database system (INTERGLAD Ver. 7, New Glass Forum) and the like, and vitrification tests are conducted in order to improve the development efficiency.

Next, data concerning the above candidate glass compositions, including their characteristics and properties, is obtained, and such compositions are assessed in terms of stability after disposal and applicability to glass melter operation.

During the development, fused glass solidification was performed on ion exchange resin, ash, and simulated liquid waste with a high sodium nitrate concentration, and results were obtained that demonstrated that vitrification would be possible at a melting temperature of 1 100°C. **Table 3** shows the details of the assessment regarding the applicability of vitrification to low-level waste. In addition, the glass compositions that had been considered were assessed from other perspectives, including viscosity and water resistance. Since no domestic standards are available for such an assessment, fused glass solidification conditions that satisfy reference values established with reference to U.S. standards, for example, are being studied in order to demonstrate the superiority of vitrified waste over other types of solidified waste.

3. Application of vitrification technology to waste produced from the Fukushima Daiichi Nuclear Power Station accident — Technology development activities based on IHI's independent research —

The following sections discuss the applicability of vitrification technology being studied by IHI in its independent research to waste from the accident at the Fukushima Daiichi Nuclear Power Station (hereinafter referred to as the Fukushima Daiichi NPS accident), as well as describe the situation of glass melter development.

3.1 Approach to selecting a melting furnace

Wastes from the Fukushima Daiichi NPS accident include a lot of wastes that have never been produced at nuclear facilities before, and such unprecedented wastes include a lot of wastes suitable for fused glass solidification.

For the fused glass solidification of wastes, it is necessary to conduct a study of melting methods taking into consideration the characteristics of the wastes. We conducted such a study in consideration of the results of overseas case studies and the like. As a result, it is suggested that a Cold Crucible Induction Melter (CCIM) is the most suitable. The CCIM (**Fig. 4**) has the following features.

- (1) This glass melter is heated by high-frequency heating, and the furnace wall (the part that comes in contact with molten glass) is made of metal.
- (2) The furnace wall is water-cooled to cause the formation of a skull layer, which has the advantage of reducing the corrosiveness of fused glass to the furnace wall material. This structural feature makes the glass melter usable with highly corrosive glass as well and extends the service life of the glass melter.
- (3) It takes up to four hours to start up the glass melter and likewise to shut it down. Waste to be treated can easily be changed. For example, the waste to be treated could be changed from day to day or week to week.
- (4) The glass melter is applicable to many different kinds of wastes, including sludge, zeolite, spent resin, liquid wastes, ash, and combustible wastes.

3.2 CCIM development

Shortly after the Great East Japan Earthquake of 2011, IHI started research on the treatment of waste from the Fukushima Daiichi NPS accident and a study of the application of fused glass solidification to such waste. In addition, IHI entered into an agreement with KHNP (Korea) in FY2012, which has a proven track record in the treatment of low-level waste with a CCIM, and since then, IHI has been studying the applicability of the CCIM to waste from the Fukushima Daiichi NPS accident. From 2013 to 2015, demonstration testing was conducted using full-scale CCIM facilities. Currently, a conceptual examination of the full-scale facilities is being carried out on the assumption that the

Low-level was	stes		Vitrification		III1	
Name	Main component	Reagent added	Waste loading factor (wt%)	Melting temperature (°C)	High temperature viscosity	Leaching rate ^{*1}
Ion exchange resin	Fe	$SiO_2 - Na_2O$	$40 \rightarrow 35$	1 100	0	
ion exchange resin	ге	P ₂ O ₅	40	1 100	—	0
Ash (Bottom ash)	Si/Ca/Al	$B_2O_3 - Li_2O$	75 or more	1 100	0	0
Low-level radioactive		SiO ₂	40	1 100	—	_
concentrate liquid	Na	$SiO_2 - B_2O_3$	40	1 100	×	×
waste (Sodium nitrate liquid waste)		$\mathrm{SiO}_2-\mathrm{B}_2\mathrm{O}_3-\mathrm{Al}_2\mathrm{O}_3-\mathrm{CaO}$	30	1 100	0	

 Table 3 Evaluation of the applicability of vitrification technology in treating low-level waste

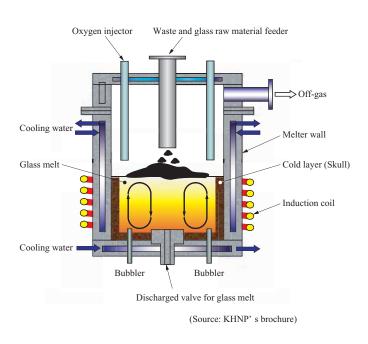
(Notes) *1 : PCT (Product Consistency Test) results

 \bigcirc : No problem, \triangle : Slight deviation from the standard value,

× : Considerable deviation from the standard value, —: No data (Untested)

(a) CCIM overview







(c) Pouring into CCIM



Fig. 4 Appearance of a cold crucible induction melter (CCIM)

facilities will be used for the treatment of waste from the Fukushima Daiichi NPS accident.

While laboratory-scale testing was conducted using a crucible and such to assess the applicability of vitrification to waste from the Fukushima Daiichi NPS accident, the following demonstration tests were conducted as CCIM demonstration tests.

- (1) FY2013: Demonstration test for zeolite and ash Based on the results of the zeolite test in FY2013, the high applicability of the CCIM was confirmed. For this reason, the melter was tested with other wastes in FY2014 and FY2015, and these tests also resulted in the high applicability of the melter being confirmed.
- (2) FY2014: Demonstration test for AREVA sludge (glass matrix used: iron-phosphate glass)
- (3) FY2015: Demonstration test for ALPS* slurry (Carbonate slurry)
 - * ALPS: Advanced Liquid Processing System (intended for radioactive substance removal)

4. Conclusion

In the basic research programs for the next generation vitrification technology of the Agency for Natural Resources and Energy of the Ministry of Economy, Trade and Industry, vitrification technology for low-level waste, which has yet to be proven in Japan, is being developed in order to transform low-level waste with relatively high levels of radioactivity into solidified waste with a substantially reduced volume that can be disposed more safely and in a more stable state.

The applicability of fused glass solidification to low-level waste was assessed based on the outcomes of IHI's independent research and the basic research programs. As a result, the following findings were obtained.

- (1) Ion exchange resin: It seems that waste existing as residue (inorganic matter) can be contained in glass at a ratio of approx. 40 wt%. In fused glass solidification, mineralization and stabilization is performed on the treated waste stored in slurry form within one process. It can therefore be expected that this technique will provide a higher volume reduction effect than other techniques in which mineralization and stabilization are performed separately.
- (2) Ash: By making effective use of a glass-formers present in waste (e.g., SiO_2), it was possible to increase the waste content to 75 wt% or more.
- (3) It was confirmed that fused glass solidification was applicable to other kinds of waste as well, and it is necessary to continue our research.
- (4) As the results of laboratory-scale tests make up a large fraction of the outcomes of a series of research activities, it is necessary to carry out future tests and studies taking actual treatment into consideration in order to solve issues involved in waste treatment.
- (5) IHI is assessing the applicability of fused glass solidification to waste from the Fukushima Daiichi NPS accident, and it has selected a CCIM as the glass melter

and confirmed that the melter is capable of treating ALPS slurry and other substances on a realistic scale.

From now on, IHI will continue to develop a glass melter suitable for low-level waste through its independent research and the basic research programs, endeavoring to expand the scope of application of IHI's vitrification technology as a whole and improve the reliability of such technology.

- Acknowledgements -

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