Development of New Waste Forms to Immobilize Iodine-129 Released from a Spent Fuel Reprocessing Plant

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Abstract. I-129 is a very long-lived radionuclide that is released to an off-gas stream when spent fuels are dissolved at a reprocessing plant. An iodine filter can capture I-129 in the form of AgI. However, because AgI is unstable under the reducing conditions of a geological repository and I-129 has a very long half-life, I-129 can migrate to the biosphere. These characteristics make I-129 a key radionuclide for the safety assessment of a geological disposal of radioactive wastes generated from a reprocessing plant (TRU wastes). To improve disposal safety, several new waste forms have been developed to confine I-129 for a very long period in order to reduce the leaching of I-129 from radioactive wastes. These new waste forms have technical objectives of solidifying more than 95% of I-129 into the waste form and achieving a leaching rate of less than 10^{-5}/y. Several iodine immobilization techniques have been examined. This paper presents experimental results concerning the treatment process, leaching behavior, modeling, and related elements of these immobilization techniques.

Introduction

At a reprocessing plant, an iodine filter with silver nitrate ($AgNO_3$) impregnated on an alumina base material (silver quantity of about 10 wt%) is used to adsorb I-129 in the chemical form of silver iodide (AgI) or silver iodate ($AgIO_3$), where the adsorbed quantity of iodine on the iodine filter is set to 6.2 wt% on average. Because AgI and $AgIO_3$ are dissociated into I^- or IO_3^- ions in the groundwater under the reducing conditions of disposal, instantaneous release is assumed to occur. These ions are highly soluble in groundwater, and it cannot be assumed that their radioactivity will be attenuated by the delay imposed by artificial barriers. Therefore, their migration to the biosphere depends mostly on natural barrier conditions such as the groundwater flow rate. A report concerning the concept and the safety evaluation of a geological disposal of radioactive wastes generated from a reprocessing plant including a spent iodine filter and from a mixed oxide (MOX) fuel fabrication plant (the ''TRU report'') has stated that I-129 is the most important of the key

radionuclides [1,2].

Attenuation by confinement in a container is not expected to be a practical way to reduce the radiation dose of the I-129 because of its very long half-life of 15.7 million years. However, reducing the peak radiation dose by solidifying I-129 into a substance that shows good long-term confinement performance of I-129 and would allow I-129 to be released gradually over a long period, may be a more practical solution, in our consideration.

Development objectives

Confinement performance of solidified substances. The radionuclides migration analysis model in the TRU report [2] can be used to calculate the effects of the solidified substance's long-term confinement performance upon radiation dose, for use in evaluation. Table 1 shows the main parameters and codes used in the model's calculations.

Table 1 Main parameters and codes used in the calculation

Confinement Performance (set as a leaching period)	Instantaneous release, 10^4, 10^5, and 10^6 years	
Groundwater transmissivity coefficient	Log-normal distribution mean: -0.99 standard deviation: 1.07	Reference case maximum: $10^{-7} m^2/s$ minimum: $10^{-13} m^2/s$
		Variation case maximum value: $10^{-6} m^2/s$ minimum value: $10^{-12} m^2/s$
Base rock	Granite	
Depth	1,000 m	
Groundwater chemical conditions	High pH, precipitating groundwater	
Groundwater migration distance	100 m	
Biosphere model	River water	
Solubility	Soluble	
Distribution coefficient	Cement mortar: 0.000125 m^3/kg	
	Compressed bentonite: 0 m^3/kg	
	Rock: 0.0001 m^3/kg	
Effective diffusion coefficient	Compressed bentonite: $4 \times 10^{-11} m^2/s$	
	Granite: $3 \times 10^{-12} m^2/s$	
Calculation code	Engineered barrier: FRONT[*1]	
	Natural barrier: CRACK[*2]	

*1; 1-dimensional migration calculation code

*2; 2-dimensional migration calculation code

Fig. 1 shows the results of this analysis. The peak radiation dose is larger for the variation case of transmissivity than for the reference case. In both cases, when the leaching period exceeds 10^5 years, the effect of the leaching period becomes dominant. For the confinement performance of the solidified substance, this result indicates that if the leaching period exceeds 10^4 years, then reducing I-129 in the release
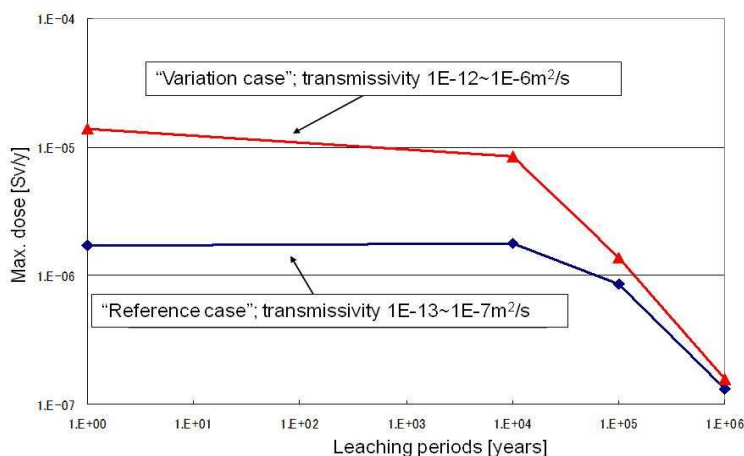

Fig. 1 I-129 max. dose vs. leaching period with different transmissivity

flux from the solidified substance will affects the release flux to the biosphere. This effect is greater in the variation case, which has a larger transmissivity coefficient, than in the reference case. If the leaching period is 10^5 years in the variation case, the peak radiation dose is reduced by about a factor of 10 and becomes lower than the peak radiation dose for the case of instantaneous release in the reference case. Based on this analysis, we set the confinement performance objective of the solidified substance to 10^5 years or longer.

Recovery efficiency at the solidification treatment process. Even if iodine is confined in a solidified substance with superior confinement performance, as described above, some amount of I-129 may not be confined in the solidified substance. Because such unconfined I-129 is considered as instantaneous release, as much of I-129 as possible must be recovered by confinement in a solidified substance. A recovery efficiency of 95% or higher was set as an objective in order not to affect the objective value set for confinement performance.

Development of immobilization techniques
Synthetic rock technique [3–6]**.**
    Outline and principle. The synthetic rock technique is a solidification treatment technique in which I-129 is processed by Hot Isostatic Pressing (HIP) without being desorbed from the spent iodine filter. The alumina adsorbent base material is compounded into a dense solidified substance (synthetic rock), and I-129 is physically confined in the alumina matrix.
    Process flow and treatment conditions. I-129 is treated by HIP processing at a pressure of 175 MPa and a temperature of 1473 K for 3 hours. As a result, iodine can be confirmed to be physically confined in a dense alumina matrix. As a pretreatment for HIP, unused $AgNO_3$ and $AgIO_3$, which can inhibit the matrix densification in the HIP treatment, undergo heat treatment to be transformed into Ag and AgI, respectively. Next, the spent iodine filter is filled into a stainless steel capsule, and the capsule undergoes vacuum degassing and is sealed. Then, HIP treatment is conducted to make the synthetic rock solidified substance. This treatment can lower the probability

of I-129 release during treatment because the treatment is carried out without desorbing I-129 from the spent iodine filter. Fig. 2 shows the treatment flow.
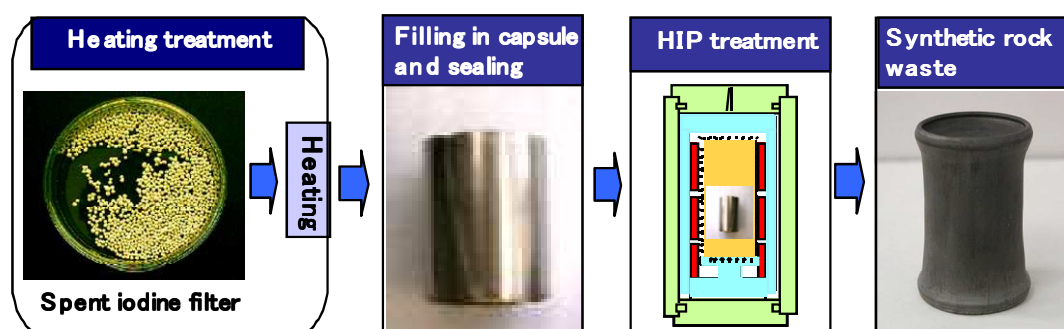

Fig. 2 Flow of synthetic rock treatment by the HIP method

Characteristics of solidified substance. The solidified cross-sectional observation and SEM observation ($\times$ 1,000) of the prepared solidified substances showed that there were no cracks in the solidified substance. Gross porosity obtained from the absolute specific gravity (4.38 g/cm^3) and the bulk density (4.05 g/cm^3) was about 8 % of which open porosity of the cubic piece (10 mm x 10 mm x 10 mm) measured by mercury intrusion porosimetry was 1–3 %. This matrix was a stable corundum (α-Al$_2$O$_3$) with a uniform distribution of iodine, which was immobilized in the chemical form of AgI. The uniaxial compressive strength of the solidified substance was 1.2 GPa.

To evaluate the I-129 retention capacity of the synthetic rock, leaching tests into solutions that simulated the geological disposal conditions were conducted. Table 2 shows the leaching conditions for the tests and Fig. 3 shows the change in amount of iodine and aluminum leached versus time. The amount of iodine leached tends to decrease gradually with time. This decrease becomes particularly dominant at 200–300 days after the start of leaching, or later.

To evaluate the long-term release behavior of I-129 from the synthetic rock, tests were conducted to confirm the dominant release process among three possible release mechanisms: matrix dissolution, AgI dissolution, and pore diffusion.

Matrix dissolution: the corundum, a component of the solidified substance, is chemically stable; however, under high-alkaline conditions, i.e. the reference disposal environment in the TRU report [1, 2], the solubility becomes larger than under neutral conditions. Accordingly, data concerning the solubility and dissolution rate of the synthetic rock versus pH were acquired, assuming disposal conditions and using NaOH and Ca(OH)$_2$ as pH adjusters. The results, which are shown in Fig. 4 and Fig. 5, can be described by the following equation for the matrix dissolution rate in free water.

$$R(Al) \;=\; k_{Al}(1 - C/C_M) \tag{1}$$

where R(Al): normalized aluminum dissolution rate [mol/cm^2/d],

C: aluminum concentration [mol/L],

C_M: aluminum saturated solubility [mol/L],

k_{Al}: rate constant ($k_{Al}=1.09 \times 10^{-7} \times [H^+]^{-0.309} \times \exp(-27,200/RT)$) [mol/cm^2/d],

        $[H^+]$: hydrogen ion concentration [mol/L],

        R: gas constant [J/K/mol], T: temperature [K]

Table 2 Test conditions for long-term leaching

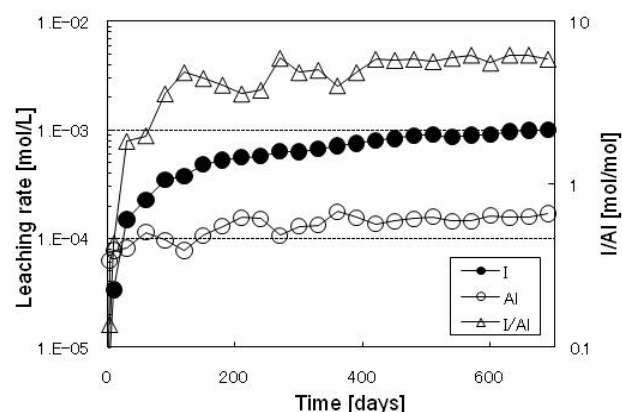| Specimen size	mm	20×20×20
pH of solution		12.5 [by Ca(OH)$_2$]
Temperature	K	308
Reducing agent		Na$_2$S
Concentration	mol/dm^3	3×10^{-3}
Solid liquid ratio (S/V)	cm^{-1}	0.1
O$_2$ concentration in gas phase	ppm	<1

Fig. 3 Results of leaching test
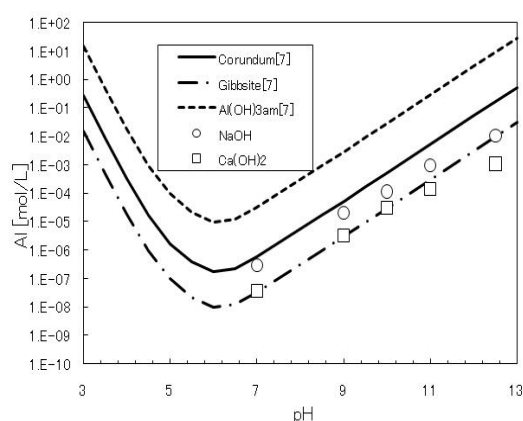(amount of iodine and aluminum leached)

Fig. 4 Solubility data of synthetic rock
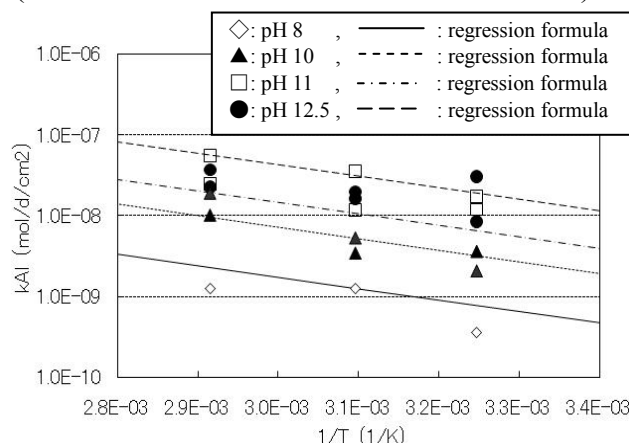substance matrix

Fig. 5 Results of solubility test of
synthetic rock substance matrix

According to Eq. 1, the matrix dissolution rate at pH 12.5 was estimated to be 9.7×10^{-7} g-Al/cm^2/d and was converted into the iodine dissolution rate to be 1.5×10^{-7} g-I/cm^2/d using the assumption of the congruent dissolution of the matrix.

AgI dissolution: AgI can elute I$^-$ by a reducing reaction (AgI + e$^-$ → Ag^0 + I$^-$) using HS$^-$, which is supposed to be a reducing species in the groundwater at the disposal environment, and also by a chemical reaction (2AgI + HS$^-$ = Ag$_2$S + 2I$^-$ + H$^+$). Accordingly, a dissolution test of AgI in Na$_2$S solution was conducted. The results showed that, under the presence of HS$^-$, AgI dissolution progressed rapidly as a result of a chemical reaction using Ag$_2$S precipitation and that an Ag$_2$S dissolution-precipitation equilibrium was almost reached within a reaction time of several days or shorter. At this point, the AgI dissolution rate at pH 12.5 was estimated to be 4.9×10^{-4} g-I/cm^2/d,

which is faster than the matrix dissolution rate stated above.

Pore diffusion: to obtain estimates of the diffusion coefficient, permeation diffusion tests were conducted on a solidified substance test piece of 1 mm thickness using the Cs^+ ion. The result was that no Cs^+ permeation was observed during the test period of 270 days. This lack of permeation is considered to result from the low open porosity of only 1–3 % and the low continuity of the pores. The low continuity can be inferred from the fact that permeation range was limited to a shallow surface area in the test of permeation from the solidified substance surface using the internal diffusion method.

According to these test results, the release of iodine from the synthetic rock is expected to occur in a process like this: the matrix within the solidified substance dissolves in a solution, then the AgI that comes into contact with the solution dissolves quickly, and iodine is released. It is considered possible that the pores on the surface of the solidified substance may affect the permeation of the solution into the matrix and the release of iodine. However, because the open porosity was low and no predominant pores were found in cross-sectional observation, this effect is considered to be small. Even though iodine and aluminum were dissolved congruently, Fig. 3 shows the I/Al release ratio to be several tens times the stoichiometrical ratio of approximately 0.15, as calculated from the composition of the sampled solidified substance. However, because the aluminum concentration measured in the leaching test was lower than the concentration in the acquired solubility data with the powdered solidified substance, this difference is assumed to be caused by the precipitation of aluminum as a secondary mineral. On this assumption, the release ratio was corrected to be approximately 0.9 which is still higher than the stoichiometrical ratio. If the solubility calculated using the PHREEQC with thermodynamic data of corundum [7] shown in Fig. 4 is used, the I/Al release ratio is corrected to be approximately 0.01 which is much lower than the stoichiometrical ratio.

The long-term performance of the synthetic rock solidified substance was evaluated by calculating the life of the solidified substance, assuming that the matrix instantaneously reaches solubility equilibrium with the groundwater in a cement environment of pH 12.5. The solubility data for $Ca(OH)_2$ in Fig. 4 show that this environment is a severe one for the synthetic rock. The aluminum concentration of the groundwater was set to 0 for the calculation. For hydraulic conditions, the hydraulic gradient was set to 1% and the permeability coefficient set to 10^{-8} m/s, based on the values in the TRU Report [1]. The result of the calculation using the acquired solubility data is a life of 10^5 years or longer for the solidified substance.

High-performance cement technique [8, 9]**.**

Outline and principle. The high-performance cement technique is a solidification method that can immobilize IO_3^- with a certain designed special cement material. The cement is minerally enriched with the calcium-aluminate ferrites, i.e. ettringite (AFt) and monosulfate (AFm), and has an oxyanion substitution ability. AFt has a needle crystalline structure with a hexagonal cross-section and is described by the chemical formula $[Ca_6(Al(OH)_6)_2 \cdot 24H_2O](SO_4)_3 \cdot 2H_2O$, and

AFm is a hexagonal crystal described by the chemical formula $[Ca_2(Al(OH)_6)_2](SO_4)\cdot 6H_2O$. The chemical formulas show a positive-charged column structure for AFt and a laminated plate structure for AFm. SO_4^{2-} and hydrated water are located in channels between the columns and plates. It is known that SO_4^{2-} in the structure can be replaced by a different anion, such as OH^- or CO_3^{2-}. After desorption from a spent iodine filter by a reductive reaction, liquid-phase I^- (I-129) is formed as the oxyanion IO_3^- by an oxidative reaction. The oxyanion easily substitute for SO_4^{2-} of AFt and AFm. Aft and AFm are known as stable minerals in natural conditions.

Process flow and conditions. The process flow of high-performance cement solidification is shown in Fig. 6. Iodine-129 in the chemical form of AgI impregnated in the spent iodine filter is dissolved into the chemical form of I^- by a reducing reaction using Na_2S. Then all of the I^- is oxidized to IO_3^- by ozone gas scrubbing. Finally, the IO_3^- solution is mixed with the high-performance cement material, which contains large amounts of AFt and AFm.

Characteristics of solidified substance. The physical properties and workability of the high-performance cement material are shown in Table 3. These properties and workability are comparable to those of a

Fig. 6 High-performance cement solidification process for I-129

conventional cement. To scale up the cement solidification process, 20 L of the solidified substance was manufactured using the established ratios, then the homogeneity and quality of outward appearance were confirmed. Table 4 shows the mineral composition of the cement solid and the corresponding iodine fractions. The table shows that iodine is present in AFt and AFm, which account for approximately 80% and 6% of the total iodine. In addition, the presence of iodine in the minerals containing calcium, aluminum and sulphur was confirmed by applying SEM-EDS techniques to the solid form.

In order to evaluate the iodine immobilization capability of the cement solid, accelerated dissolution tests were conducted by a repeated replacement with an ion-exchanged water as the immersing fluid. Fig. 7 shows the concentrations of cementitious elements as a function of the overall liquid-to-solid ratio (L/S). The results show that the iodine concentration after the alteration of the solid is in an acceptably low range. In addition, AFt remained in the altered cement solid after replacement with ion-exchanged water.

Based upon these findings, we believe that IO_3^- is immobilized in the cement solid by transfer through ion-exchange substitution to the anion site of cementitious minerals, especially AFt, and that iodine may be released from the cement solid if AFt that has undergone IO_3^- substitution is dissolved. Therefore, the iodine release from this cement solid may be evaluated by the solubility equilibrium model, which is based on the concept that the solubility of cementitious minerals is
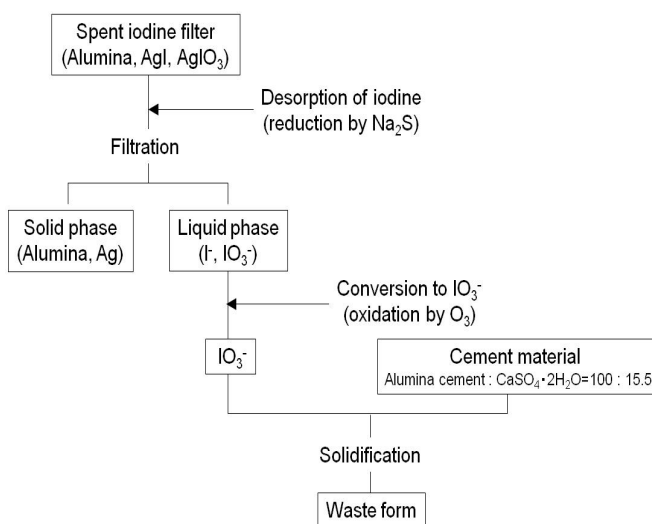
affected by the chemical properties of groundwater. The alteration of minerals in the cement and the release of iodine due to immersion were evaluated in thermo-equilibrium conditions by using the PHREEQC Interactive v2.155 [7]. The initial composition of the minerals is listed in Table 4 and the chemical properties of groundwater were estimated based on a fresh groundwater.

Calculations using the PHREEQC model revealed that iodine concentration released from the cement solid on the order of 10^{-5} mol/L for lower L/S values and less than or equal to 10^{-3} mol/L at higher L/S values. In addition, the model suggests that AFt that has undergone IO_3^- substitution will remain in the cement solid for conditions up to L/S=500. According to a simulation of disposal conditions described in the TRU report [2], the L/S=500 condition may be equivalent to an elapsed time of 10^5 years or more after disposal. Therefore, it is believed that the cement solid retains its iodine confinement ability for as long as 10^5 years.

Table 3 Physical properties of cement solidified substance

Measurement Items		Values
Mixing properties	Flow value [cm]	29.2, 27.8
	P funnel value [sec.]	12.2, 12.7
	Coefficient of viscosity [dPa·s]	16, 12
Hardening properties	Hardening time [min]	Initial setting: 45, final setting: 120
	Bleed [%]	After 3 hrs: 0 Over 20 hrs: 0
	Coefficient of expansion [%]	0.35 %
Characteristics of cement solid*	Axial compression strength [MPa]	After 30 days: 22
	Density [g/cm^3]	1.74
	Porosity [%]	30
	Iodine content [%]	1.7
	External appearance	Good (no cracks)

* Test pieces: diameter = 5 cm, height = 10 cm

Table 4 Mineral composition of cement solid and iodine fraction

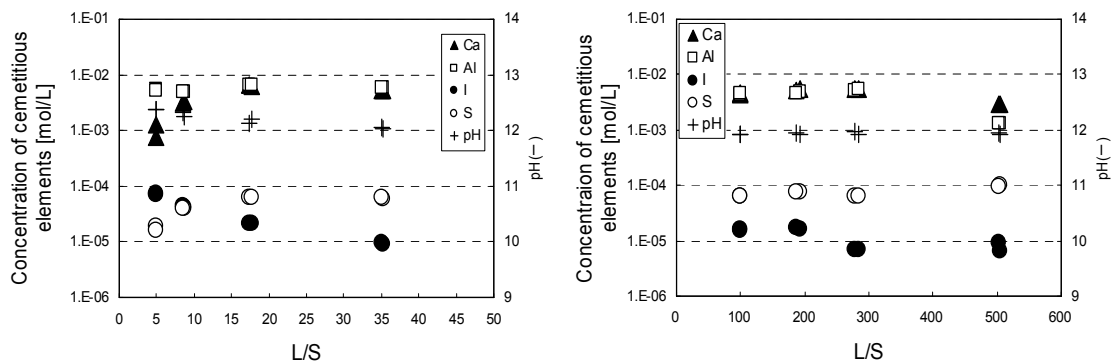| Minerals	Ettringite (AFt)	Monosulfate (AFm)	Hydrogarnet	Calcite	Monocalcium aluminate	Gibbsite
Composition [%]	12	23	19	0	2	45
Iodine fraction [%]	79	6	16	0	0	0

Fig. 7 Concentrations of cementitious elements as a function of overall liquid-to-solid ratio (L/S)

BPI vitrification technique [10–13]**.**

   Outline and principle.   The BPI vitrification technique consists of an iodine recovery from a spent iodine filter using H_2 and a low-temperature vitrification to form a homogeneous dispersion of iodine in a PbO-B_2O_3-ZnO glass system.

   Process flow and conditions. Initially, iodine is desorbed as HI from a spent iodine filter using H_2. Then the iodide ion is fixed within $BiPbO_2I$ (BPI) by reaction with an inorganic anion exchanger, $BiPbO_2NO_3$ (BPN). The ion exchange reaction can be described as follows:

$$BiPbO_2NO_3 + I^- \rightarrow BiPbO_2I + NO_3^- \tag{2}$$

The vitrification process uses a low melting temperature of 813K to avoid an iodine volatilization during the vitrification with PbO-B_2O_3-ZnO glass frit (Fig. 8).
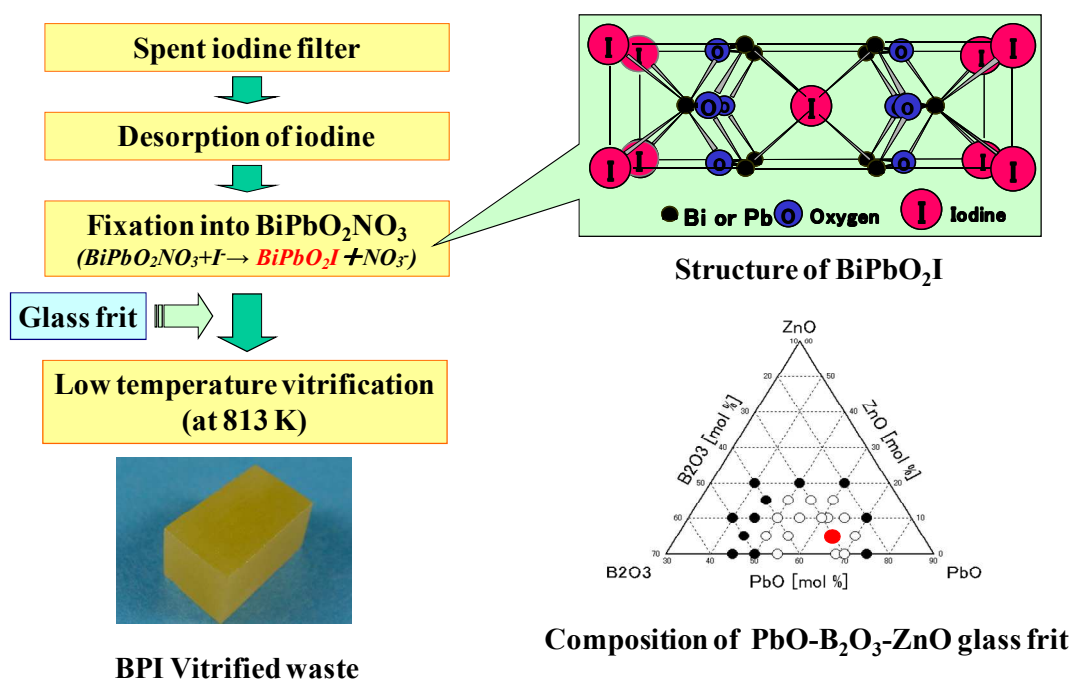

Fig. 8 Outline of BPI vitrification process

   Characteristics of solidified substance. To advance the understanding of the BPI glass, its microstructure was analyzed by ^{11}B MAS-NMR, XRD, XPS and XAFS. Based on ^{11}B NMR spectrums of the PbO-B_2O_3-based glasses with iodine contents for 0.2, 1.0, and 2.0 wt%, the ratio of tri-coordinated boron (B3) and tetra-coordinated boron (B4) was approximately 7:3. The RDF curves obtained by XRD analysis of the above glasses using SPring-8 (Super Photon ring-8GeV) are similar, which suggests that these glasses have nearly the same bulk structure. In addition, peaks can be found at 0.24 nm and 0.39 nm, which represent the Pb-O bond and the Pb-Pb pair, respectively. XPS and XAFS analyses show that iodine is present in the BPI glass in the iodide ion

state and selectively forms Pb-I bonds, and that the microstructures surrounding the iodine in the glass matrix are nearly the same.

To demonstrate the feasibility of the BPI vitrification process, a prototype melting furnace was developed to produce the BPI vitrified waste in quantities of 0.5 L. The samples were divided into 90 pieces and underwent XRD analysis, which found the samples to be amorphous. The dispersion of iodine contents and the specific gravity in the samples was small enough to be considered homogeneous. The uniaxial compressive strength of the BPI glass was over 40.8 MPa. The volatilization rates of iodine during the BPI vitrification were found to be in the range of 5–6 %.

Leaching tests were conducted to evaluate the iodine leaching rate from the BPI vitrified waste in various solutions that simulated geological disposal conditions. Table 5 and Fig. 9 show the experimental conditions for the leaching tests and normalized leaching rates of boron (NL_B) and iodine (NL_I) that resulted.

Table 5 Experimental conditions for leaching tests

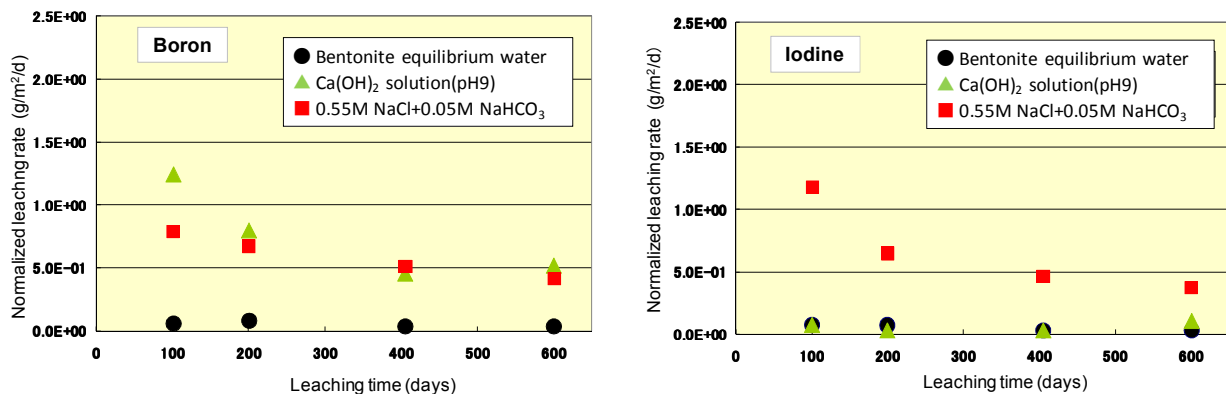| Item		Contents
Ratio of liquid/solid		10 mL/g
Solid	Glass	BPI glass,　10 mm×20 mm×5 mm (7 g)
Liquid	Leachant	1) Ca(OH)$_2$ solution (pH 9) 2) 0.55M NaCl + 0.05M NaHCO$_3$ solution 3) Bentonite-equilibrium water
	Temperature	Room temperature
	Reductant	Electrolytic iron powder (1 wt %)
Atmosphere		Low oxygen (less than 1 ppm)

Fig. 9 Normalized leaching rate of boron and iodine

Fig. 10 shows the conceptual model of the iodine dissolution from the BPI glass. The dissolution behavior can be categorized into two types depending on the properties of the solution used. One type of behavior is that iodine dissolves congruently with boron, which indicates the dissolution of the glass matrix. This behavior can be observed in a bentonite equilibrated water, a NaCl solution, and similar media. The other type of behavior is that iodine dissolves quite slowly

compared to boron. This behavior can be observed in a $Ca(OH)_2$ solution.

When iodine dissolved congruently with boron, an altered layer was observed at the glass surface. Also, both boron and iodine were depleted in the altered layer. These observations suggest that the glass dissolution rate is related the formation rate of the altered layer. Moreover, the dissolution rate of boron and iodine was proportional to time $t^{0.5}$, which suggests that diffusion occurs.

In the other case, in which iodine does not dissolve congruently with boron, a precipitation layer was observed in addition to the altered layer. An EPMA analysis showed significant concentrations of iodine and bismuth in the precipitation layer, and a XANES analysis suggested that a compound similar to a BPI crystal was precipitated. The iodine dissolution rate appears to be controlled by the extent of immobilization in the precipitation layer.

Fig. 10 Conceptual model of iodine dissolution behavior from BPI vitrified waste

Summary

This report presented developments and results of I-129 immobilization techniques. Several alternative techniques were proposed and, after the preliminary study and evaluation, three candidate techniques are now under study. Although each technique demonstrates a potential to achieve the long-term I-129 confinement objective by a solidified substance, each technique shows its own specific features as well. Synthetic rock may demonstrate the superior confinement performance in a neutral pH, fresh groundwater environment, where alumina matrix solubility is low. High-performance cement can be used for all types of groundwater with the exception of a saline groundwater where accelerated matrix dissolution makes it difficult to achieve the confinement objective. BPI glass has a simple modified layer in a bentonite-equilibrium water

environment or a saline groundwater environment that dissolves congruently with boron. However, BPI glass will form a complex modified layer on its surface when immersed in a groundwater environment that is influenced by the leaching of calcium from cement materials.

Future research will focus on long-term immersion tests to develop the evaluation model of long-term performance of each solidified substance under a variety of groundwater conditions. The research to evaluate treatment feasibility by obtaining the basic process data for achieving an I-129 recovery rate of 95 % or greater will be also pursued.

Acknowledgements

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