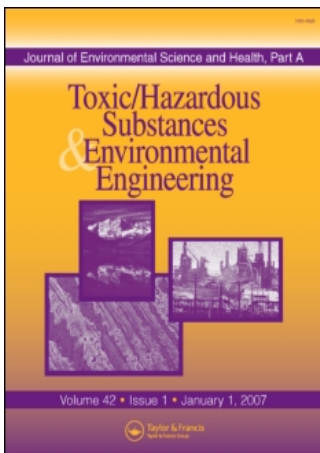


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## NON-COMBUSTIBLE WASTE VITRIFICATION WITH PLASMA TORCH MELTER

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### ABSTRACT

Non-combustible radioactive wastes generated from Nuclear Power Plants (NPPs) are composed of concrete, glass, asbestos, metal, sand, soil, spent filters, etc. The melting tests for concrete, glass, sand, and spent filters were carried out using a 60 kW plasma torch system. The surrogate wastes were prepared for the tests. Non-radioactive Co and Cs were added to the surrogates in order to simulate the radioactive waste. Several kinds of surrogate prepared by their own mixture or by single waste were melted with the plasma torch system to produce glassy waste forms. The characteristics of glassy waste forms were examined for the volume reduction factor (VRF) and the leach rate. The VRFs were estimated through the density measurement of the surrogates and the glassy waste forms, and were turned out to be 1.2–2.4. The EPA (Environmental Protection Agency) Toxicity Characteristic Leaching Procedure (TCLP) was used to determine the leach resistance for As, Ba, Hg, Pb, Cd, Cr, Se, Co, and Cs. The leaching index was calculated using the total content of each element in both the waste forms and the leachant. The TCLP tests resulted in that the leach rates for all elements except Co and Cs were lower than those of the Universal Treatment Standard (UTS) limits. There were no UTS limits for Co and Cs, and their leach rate & index

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from the experiments were resulted in around 10 times higher than those of other elements.

*Key Words:* Vittrification; Non-combustible wastes; Plasma torch; Cs and Co incorporation; High temperature melting; Volume reduction factor.

## INTRODUCTION

The NPPs generate gaseous, liquid, and solid radioactive wastes during their operation and maintenance, which are termed as low- and intermediate-level radioactive waste (LILW). The solid waste is largely classified as combustibles (e.g. protective clothings, shoes, socks, vinyl sheets, spent ion-exchangers, etc.) and non-combustibles (e.g. sand, concrete, glass, metallic waste, spent filters, etc). The combustibles are put in a 208-liter steel drum and then compressed by a super compactor. Some countries such as the U.S.A., Japan, Germany, France, etc. incinerate the combustibles excluding spent ion-exchangers since their radioactivity is much higher than others. The spent ion-exchangers are generally solidified with cement or preserved in a High Integrity Container (HIC) after drying. The non-combustibles are mainly put into a 208-liter steel drum without any treatment, but spent filters are stored in a cement-lined drum (208-liter) because of its high radioactivity (1).

New technologies of LILW treatment are highly demanded on the several problematic respects such as the environment protection, the difficulties in disposal site selection, the disposal cost increase, and the current lack of appropriate means for the handling of LILW waste. Because LILW vitrification, a new technology, can dramatically reduce the volume of the waste and provide more stable waste forms of LILW in reasonable cost, this technology is considered to be the most promising among the new LILW treatment technologies and hence, there has been a world-wide trend to apply it for the treatment of LILW (2,3,4).

Several melters such as plasma torch melter (PTM), cold crucible melter (CCM) heated by direct induction current can be applied to vitrify LILW (1). This paper describes experiments for noncombustible waste melting with a 60 kW plasma torch melter. A plasma torch is a device that converts electrical energy into thermal energy (5,6). The plasma arc creates a "flame" that has temperatures ranging from 4,000 to 7,000°C. Thus plasma torches operate at much higher temperatures, higher enthalpies, and at much greater efficiencies than those of fossil-fuel burners. In addition, plasma torches require only about 5% of the gas needed for fossil fuel burners. Therefore, effluent gases are greatly reduced, and furnace systems can be built that are much more compact than traditional furnaces, at correspondingly reduced capital costs (7).

In this paper, the melting experiments for concrete, glass, sand, and spent filters were performed with a 60 kW plasma torch system. In order to examine the vitrification possibility for the waste mixture or the individuals, several surrogates were prepared without any flux addition and with addition of non-radioactive Co and Cs to simulate the radioactive waste. The surrogates were put into the plasma torch melter and melted in order to convert into glassy waste forms that were examined to analyze the leaching properties, volume reduction factor, etc. Measurements on specific density and total contents of chemical species, and TCLP tests were performed for the surrogate wastes and the waste forms, respectively.

## EXPERIMENTS

### Materials and Equipment

The plasma torch melting system is comprised of a 60 kW plasma torch melter, DC power supplier, control panel, and off-gas analyzing system as shown in Figure 1. The plasma torch is a solid and straight polarity type. During the experiments, it was mostly operated in transferred mode. In other words, at the startup stage, the startup arcing takes place between a tungsten solid cathode placed at rear of the torch and a copper anode at front (which is called the non-transferred mode). The startup arcing ionizes plasma gas, which provides the conducting passage between the cathode of

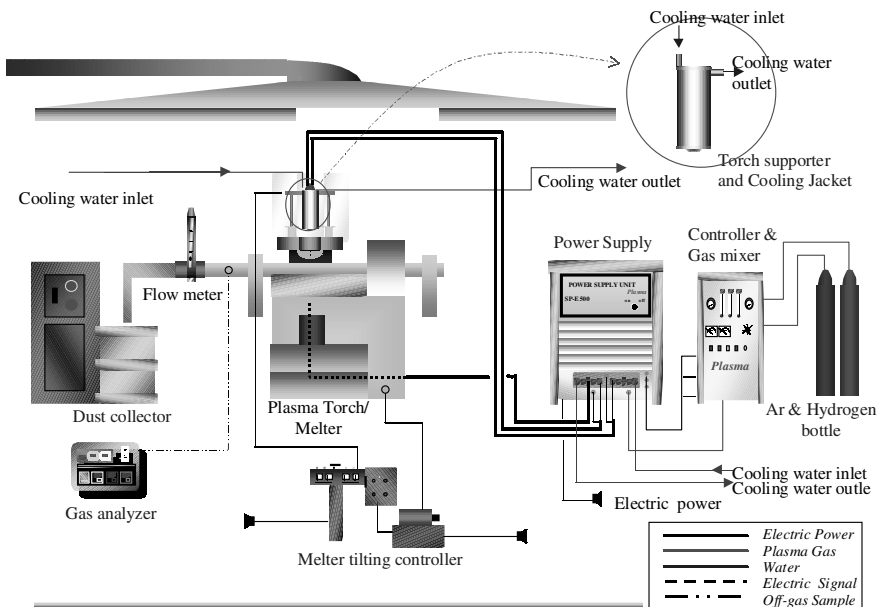


Figure 1. Schematic diagram of a 60 kW plasma torch melting system.

torch and the anode placed on the melter bottom, and then main plasma is generated (which is called the transferred mode). When the main plasma is generating, the startup arcing is automatically disappeared.

The cooling system of torch includes the external cooling jacket whose function is to cool the whole torch body, and the internal cooling loop whose role is to cool down the front electrode (Cu anode) of the torch. The tap water was used as cooling water without any pre-treatment. The melter is made of an alumina refractory lined S.S. vessel that was designed to be suitable for the batch operation. Its dimension is 250 mm both I.D. and effective height. Its internal volume is designed to be able to melt the metallic waste of 5 kg.

Several surrogate wastes were prepared as shown in Table 1, which is showing the chemical composition of the wastes and the glassy waste forms (in lower lows). The surrogates were made using concrete, glass, sand, and filters. It was assumed that the filters were composed of S.S., galvanized steel, glass fiber, and polypropylene with ratio of 2:1:1:1. The surrogates, PT1 to PT5 were simulated by mixing (ratio 3:1) of two materials among concrete, sand, and glass, and PT6 to PT9 using single material. PT10 was prepared with four materials in equal weight of each. The weight of surrogates were 500 g, but PT10 was 600 g. All surrogates include non-radioactive Cs ( $\text{Cs}_2\text{CO}_3$ ) and Co (powder) under the ratio of 5 g per one kg of waste.

### Experimental Procedure

Because the surrogates were non-conductive materials, a molten metal should be formed in order to operate the plasma torch in transferred mode. Therefore, carbon steel (4,000 g) was melted for molten metal formation prior to the waste feeding. The plasma torch cooling system and the off-gas treatment system have to be started before the torch ignition. The mixture of Ar (18 lpm) and  $\text{H}_2$  (2 lpm) is injected to the plasma torch as plasma generation gas, and then the start-up button for arc generation is pushed from control panel. Non-transferred mode operation is immediately started, and then the torch operation is automatically switched to the transferred mode. After switching to the transferred mode, electrical current rate was adjusted to 500 A. This rate was kept whole through the control of distance between the torch bottom and the metal pool.

The waste is fed onto the molten metal after sufficient melting of the carbon steel in the melter bottom. If the waste is thoroughly melted, the molten waste and metal are separated from each other due to the density difference. As soon as the plasma torch operation stopped, the molten waste and metal are drained into a mold by tilting of the melter. It takes about 3 minutes for melting of carbon steel, 7–8 minutes for both waste feeding and melting, and 2 minutes for the drainage of molten material.

**Table 1.** Chemical Compositions of Wastes and Glassy Waste Forms

Oxides	Compositions of Wastes and Glassy Waste Forms (wt.%)									
	PT1	PT2	PT3	PT4	PT5	PT6	PT7	PT8	PT9	PT10
SiO <sub>2</sub>	50.50	67.27	45.38	51.94	60.33	42.11	75.66	55.22	88.59	59.60
Al <sub>2</sub> O <sub>3</sub>	49.26	57.74	56.65	45.55	61.26	52.68	69.44	62.62	55.56	49.11
MnO	7.96	2.69	13.75	20.06	17.43	10.59	0.05	23.22	0.09	10.59
CaO	22.06	12.24	11.92	22.02	14.41	20.47	2.39	5.35	21.21	21.17
MgO	0.58	0.41	0.67	0.69	0.61	0.66	0.32	0.70	0.06	0.53
Na <sub>2</sub> O	0.58	0.41	0.41	0.49	0.56	0.92	0.36	0.59	0.56	0.78
K <sub>2</sub> O	1.96	2.23	3.30	6.25	6.38	1.83	2.36	7.72	0.01	3.72
P <sub>2</sub> O <sub>5</sub>	1.45	1.08	1.09	5.51	1.20	1.11	3.09	12.45	1.82	1.80
Fe <sub>2</sub> O <sub>3</sub>	2.47	2.63	2.11	1.54	1.62	2.39	2.71	1.26	0.02	1.99
TiO <sub>2</sub>	2.35	2.36	2.33	1.82	0.18	2.39	0.23	1.92	2.38	2.38
Σ(others)	4.12	5.08	2.97	1.64	2.12	3.64	5.56	0.97	0.53	3.21
CaO/SiO <sub>2</sub>	3.26	3.45	3.48	3.93	3.56	3.52	3.22	3.33	3.59	3.99
SiO <sub>2</sub> +	0.51	1.08	0.84	2.07	2.36	0.22	1.37	2.69	0.58	1.37
Al <sub>2</sub> O <sub>3</sub>	1.56	1.73	1.35	2.00	1.95	2.42	2.09	2.08	2.02	2.06
Σ(RO+R <sub>2</sub> O)	0.02	0.02	0.02	0.03	0.03	0.02	0.02	0.03	0.00	0.02
Waste	0.07	0.08	0.08	0.00	0.00	0.05	0.01	0.06	0.03	0.02
Ingredients*	20.26	14.11	18.81	9.77	7.00	23.33	11.03	5.25	2.35	12.53
	18.59	21.26	22.35	19.36	18.28	18.83	20.02	14.11	15.98	18.99
	0.32	0.21	0.35	0.30	0.25	0.37	0.15	0.28	0.04	0.25
	0.33	0.36	0.36	0.35	0.21	0.19	0.24	0.38	0.28	0.27
	31.90	18.61	30.98	15.81	8.73	38.56	11.97	8.22	10.12	18.99
	19.48	20.99	22.77	18.68	16.88	16.49	19.18	11.66	12.86	18.71
	0.039	0.033	0.073	0.120	0.106	0.043	0.031	0.140	10 <sup>-4</sup>	0.062
	0.029	0.019	0.019	0.121	0.020	0.021	0.004	0.199	0.033	0.037
	58.46	69.96	59.13	72.00	77.76	52.70	75.71	78.44	88.68	70.19
	71.32	69.98	68.57	67.57	75.67	73.15	71.83	67.97	76.77	70.28
	9.64	11.43	9.89	12.19	13.51	8.74	12.32	13.34	1.20	10.82
	9.20	9.03	8.66	13.75	7.45	10.36	8.99	20.37	10.37	11.01
										C:25
										G:25
										S:25
										F2:25

\* C: concrete, G: glass, S: sand, F1(simulated filters): S.S.(200 g) + galvanized steel(100 g) + glass fiber(100 g) + polypropylene(100 g), F2(simulated filters): S.S.(60 g) + galvanized steel (30 g) + glass fiber(30 g) + polypropylene(30 g).

## RESULTS AND DISCUSSION

### Volume Reduction Factor

In order to estimate the volume reduction factor (VRF), densities were measured for the wastes and the glassy waste forms. The measurement for wastes was performed for weight and volume of each waste. Two measurement methods were applied for the waste forms. One is to measure the weight

and volume of the waste forms in powder form, whereas the other one is to measure the waste form in itself. The powders are prepared by grinding the waste forms to the size of 0.25–1.0 mm in diameter for removal of cavities within them. We call the densities measured by the former method *the intrinsic density* and by the latter *the superficial density*. Table 2 describes the intrinsic and the superficial densities, and the volume reduction factor. It shows that the VRF is the largest in PT9 because the surrogate waste of PT9 contains polypropylene. The VRFs of PT2 and PT7 show no difference between the intrinsic VRFs and the superficial VRFs, which means that there are no bulky cavities in glassy waste forms. It is desirable to exclude the bulky cavities from a viewpoint of the volume reduction and the waste form quality. In order to remove the bulky cavities, various techniques such as the flux addition and the optimization of operating parameters (melting temperature, redox potential, etc.) might be adopted.

### Composition Analysis for Wastes and Glassy Waste Forms

The compositions of surrogates were calculated using the oxides compositions of concrete, sand, and glass as shown in Table 1. Those of waste forms were measured with an XRF (x-ray Fluorescence) manufactured by Shimadzu in Japan (model 1700). Figure 2 describes the compositions of network formers ( $\text{SiO}_2 + \text{Al}_2\text{O}_3$ ), network modifiers ( $\Sigma(\text{RO} + \text{R}_2\text{O})$ ), and other oxides. The network formers are elements that can replace silicon. And the network modifiers are elements that do not enter the network, but form ionic bonds with nonbridging oxygen atoms in the structure of silica glass network (8). This figure also shows that despite the compositions of wastes largely fluctuate, the fluctuation for those of glassy waste forms

**Table 2.** Densities and Volume Reduction Factors

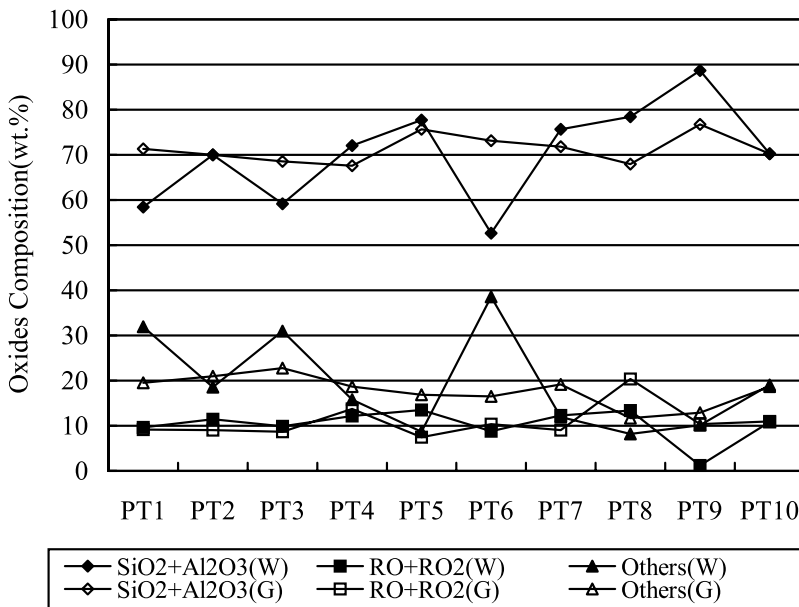
I.D. #	Densities ( $\text{g}/\text{cm}^3$ )				Volume Reduction Factors	
	A	B	C	B-C	B/A	C/A
PT1	1.316	3.450	–	–	2.622	–
PT2	1.250	2.990	3.000	–0.010	2.392	2.400
PT3	1.351	2.740	1.680	1.060	2.028	1.243
PT4	1.563	3.330	2.130	1.200	2.131	1.363
PT5	1.563	2.360	2.000	0.360	1.510	1.280
PT6	1.250	3.070	1.720	1.350	2.456	1.376
PT7	1.282	2.180	2.110	0.070	1.700	1.646
PT8	1.429	2.550	1.700	0.850	1.785	1.190
PT9	0.439	3.230	–	–	7.364	–
PT10	0.938	2.440	1.830	0.610	2.603	1.952

Note; A: waste, B: intrinsic density, C: superficial density.

becomes smooth. The composition ranges for network formers, network modifiers, and the others in waste forms are 68–77 wt.%, 9–20 wt.%, and 12–21 wt.%, respectively. The ranges are similar to those of waste forms produced by ISV (In-Situ Vitrification) of INEEL soils, which are corresponding to 84.7 wt.%, 9.8 wt.%, and 5.5 wt.% (8).

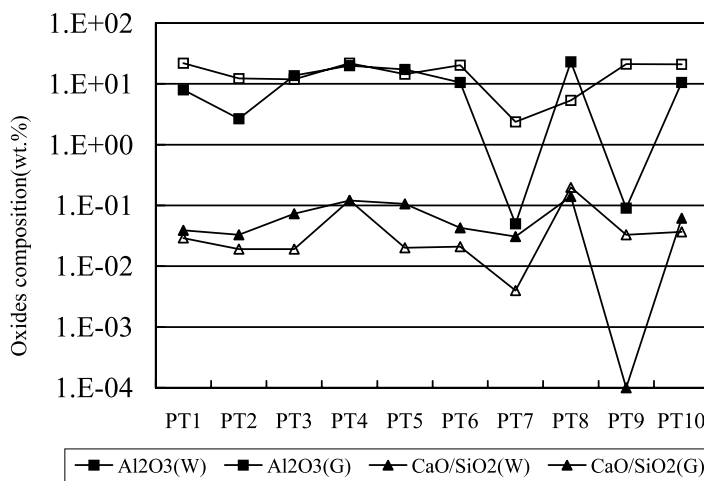
As shown in Table 1 and Figure 3, alumina contents in waste forms such as PT1, PT2, PT7, and PT9 are higher than those in corresponding wastes, but PT8 shows the reverse phenomenon. Alumina dissolution from wastes or waste forms may result in the smooth variation of oxide composition in waste forms. The degree of alumina enrichment and reduction in waste forms might be changed in accordance with the basicity ( $\text{CaO}/\text{SiO}_2$ ) of wastes. The basicity is classified as the acid waste (0.2–0.7), the neutral (0.7–1.2), and the basic waste (1.2–4.0) according to the value of  $\text{CaO}/\text{SiO}_2$  (9). It is thought that the enrichment source of alumina is the alumina refractory of plasma torch melter.

Figure 3 indicates the relationship between the alumina enrichment amount and the basicity of waste. We can find out that all wastes are nearly acid waste and the wastes having the basicity of about 0.06 and less result in the alumina enrichment in the waste forms. The enrichment degree increases as the basicity decreases. For the basicity more than 0.07, the alumina contents in wastes and waste forms are nearly the same as each other or the contents in waste forms are lower than those in waste.



**Figure 2.** Oxides composition of the waste and the glassy waste forms (closed: wastes, opened: waste forms).





**Figure 3.** Variation trends for alumina composition in waste forms with respect to basicity of waste (closed: wastes, opened: waste forms).

### Leaching Characteristics

The leaching test was carried out in accordance with TCLP established by U.S.A. EPA (10). The Cs content in leachant was analyzed with ICP-MS (Inductively Coupled Plasma – Mass spectrometer (model: VG plasma Quad PQ2 Turbo made by Shimadzu)), and contents for the other elements with ICP-AES (Inductively Coupled Plasma – Atomic Emission Spectrometer (model: ICP S-1000 IV, Shimadzu in Japan)).

As described in Figure 4, all waste forms satisfy UTS limits of EPA for all elements excluding Cs and Co. We should know that there are no UTS limits for Cs and Co so that we can not interpret the results. The leached amount of Co and Cs was about 10 times more than those of the other elements. In addition, the leached Co (2.4–67.19 mg/l) were around 2 to 5 times more than the leached Cs (0.5–28.84 mg/l). These results were supposedly caused by higher contents of Co and Cs in waste (5,000 ppm) than the contents of other elements, and by the solubility difference in silica glass (3–5 wt.% for Co and 15–25 wt.% for Cs). The leaching property for mercury (Hg) was also examined, but not detected due to the low boiling point (356.9°C) and the low solubility in silica glass (<0.1 wt.%) (8).

The leached amount variation of elements may be depending on the total content in waste forms. Therefore, the leached fractions (LF) were investigated for the absolute comparison. The leached fraction is defined as follows:

$$LF_i = \frac{C_{il}}{C_{ig}} \times 20, \quad (1)$$

where  $C_{il}$  (mg/l) and  $C_{ig}$  (mg/kg) are respectively the concentration in leachant and total concentration in a glassy waste form for element  $i$ . The value, 20 is the weight (g) of a sample per 1 liter of digestive solution which should be utilized to dissolve metal elements thoroughly in a waste form in the case of Microwave Digestion (MD) analysis.

As shown in Figure 5, LF has a wide variation according to the type of metal. We can see that LF of Cr is the smallest (<0.1 wt.%), but that

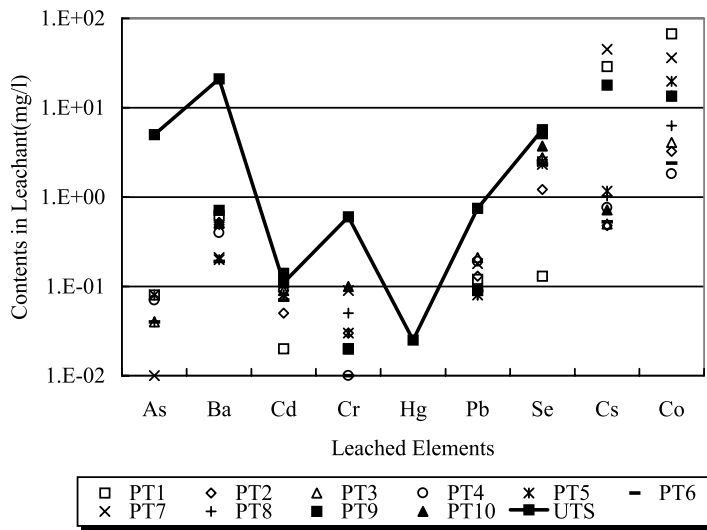


Figure 4. Elements concentration in leachant with respect to the waste forms.

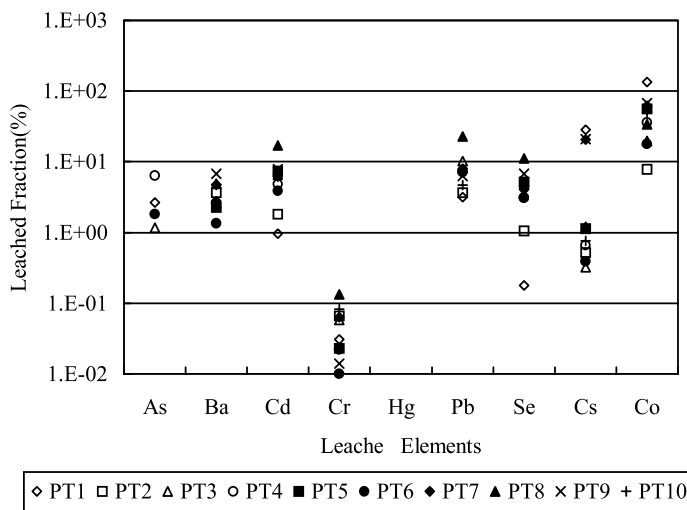


Figure 5. Leached fraction for 9 elements in waste forms with respect to waste type.

of Co is very large up to 100 wt.% for PT1 waste form. The incorporation ratio for Cs and Co was also investigated and the ratio was turned out to be 30–80 wt.% and 2–25 wt.%, respectively. The result is unexpected because the volatility of Cs is higher than that of Co. This phenomenon is supposed to be caused by that the chemical forms of the added Cs and Co are different, which are from  $\text{Cs}_2\text{CO}_3$  and Co powder, respectively. It was known that the  $\text{Cs}_2\text{CO}_3$  can slightly improve retention of Cs-137 but the mechanism was not determined (11).

## CONCLUSION

In order to examine the feasibility of vitrification for non-combustible wastes from nuclear power plants, the melting experiments with a 60 kW plasma torch system were performed for the wastes such as concrete, sand, glass, and pseudo filters, and some combined wastes of them. Ten surrogate wastes were prepared and substitutional non-radioactive Cs as  $\text{Cs}_2\text{CO}_3$  and Co powder were added to simulate the radioactive waste. The surrogates were melted with the plasma torch and converted into the glassy waste forms. These waste forms were analyzed to find out the best conditions for glass formation.

All waste forms show satisfactory results for TCLP test on As, Ba, Cd, Cr, Hg, Ce, and Pb compared with UTS limits. However, considering volume reduction factor and leaching property for Co and Cs, the best waste form was the one made from the waste of concrete (25 wt.%) and glass (75 wt.%). Its composition was the glass network formers ( $\text{SiO}_2 + \text{Al}_2\text{O}_3$ ) of 69.98 wt.%, the network modifiers ( $\Sigma(\text{RO} + \text{R}_2\text{O})$ ) of 9.03 wt.%, and the rest of 20.99 wt.%. The VRF of waste form from the waste of concrete (75 wt.%) and glass (25 wt.%) was turned out to be the poorest. The VRF of other waste forms from the combination of wastes were larger in the following order as PT3 (concrete 75 wt.% + sand 25 wt.%), PT4 (concrete 25 wt.% + sand 75 wt.%), and PT5 (glass 25 wt.% + sand 75 wt.%). The waste formers from the combination of wastes had the range of superficial volume reduction factor in 1.28–2.40. The range of oxides composition of the waste forms was the glass network formers ( $\text{SiO}_2 + \text{Al}_2\text{O}_3$ ) in 67.57–75.67 wt.%, the network modifier ( $\Sigma(\text{RO} + \text{R}_2\text{O})$ ) in 7.45–13.75 wt.%, and the rest in 16.88–22.77 wt.%. In addition, it might be concluded that the basicity ( $\text{CaO}/\text{SiO}_2$ ) of wastes is required to be 0.07 or more in order to prevent the corrosion of alumina refractory. Among the waste forms PT6 to PT9, which were made of concrete, glass, sand, and pseudo filters, respectively, PT6 shows the best result from a viewpoint of incorporation of Co and Cs. PT7 was the best for superficial volume reduction factor.

Further work should be carried out in terms of the following categories: (1) The melting test and examination for waste forms made from variable mixing of wastes with addition of another chemical form such as CsCl and CoCl<sub>2</sub>; (2) The study is necessary to increase the incorporation rate of Cs and Co with much low concentration.

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