Development of the Volume Reduction Treatment of Solid Waste System by Ultra-High Frequency Induction Furnace - 9356

T. Sakakibara, Y. Aoyama, H. Yamaguchi
Japan Atomic Energy Agency
4-33, Muramatsu, Tokai-Mura, Naka-Gun, Ibaraki pref. 319-1194, Japan

N. Sasaki, T. Nishikawa, M. Murata
Mitsubishi Materials Corporation
1002-14, Naka-City Ibaraki pref. 311-0102, Japan

J. Park, S. Taniguchi
Tohoku University
6-6-02, Aoba-yama, Sendai-City, 980-8579, Japan

M. Fujita
Fuji Electric Thermo Systems Corporation
5520, Minamitamagakicho, Suzuka-City, Mie pref. 513-8633, Japan

T. Fukuda. H, Suzuki
TYK Corporation
3-1, Ohbatacho, Tajimi-City, Gifu pref. 507-8607, Japan

ABSTRACT

The volume reduction treatment of solid waste system by ultra-high frequency induction furnace (UHFIF) was developed from FY2005 to FY2007. Basic data for melting performance were collected by non-radioactive experiments using the bench scale UHFIF with a crucible capacity of 10 liters. Based on the obtained data, engineering specifications were evaluated for a demonstration scale UHFIF with a crucible capacity of 30 liters. A new demonstration scale UHFIF was constructed and melting experiments of surrogate wastes were carried out by this furnace. It was confirmed that the demonstration scale UHFIF can melt ferrous metal, ceramics and aluminum all together and stabilize aluminum by oxidation to alumina. Density, chemical composition, and surface condition of the solidified substances were analyzed, and homogeneity of the solidified substances was confirmed.

Melting behavior in the demonstration scale UHFIF was analyzed by computer simulation and simulation

results agreed well with the experimental ones. From the design study for a full scale UHFIF with a crucible capacity of 100 liters, basic specifications were evaluated for the full scale UHFIF. Based on the obtained specification, melting behavior in the full scale UHFIF was analyzed by computer simulation.

INTRODUCTION

Volume reduction treatment of radioactive waste by melting is effective in simplifying measurement of radioactivity in waste packages because of their homogeneity and reducing waste packages in number for high volume reduction ratio of the melting system. Main constituents of the melting system are an induction melting furnace and a plasma melting furnace, both of which are adopted for nuclear facilities. However, an induction furnace heats solid wastes indirectly with graphite and such things. A plasma melting furnace heats solid wastes on their surface locally. Therefore, it is difficult for the both melting systems to melt solid wastes consist of different kinds of material homogeneously. Accordingly, it is necessary to separate solid wastes exactly into flammable waste, metal waste, ceramics waste and so on, at the pretreatment stage before melting.

Conventional induction furnaces heat metal with electromagnetic induction of frequency of up to 3 kHz. However, molten ceramics can be heated with electromagnetic induction of frequency of more than 100 kHz. The relation between absorbed power and frequency is shown in Fig.1. Based on this principle, ultra-high frequency induction furnace heats both metal and ceramics directly by induction heating with frequency of about 100 kHz.

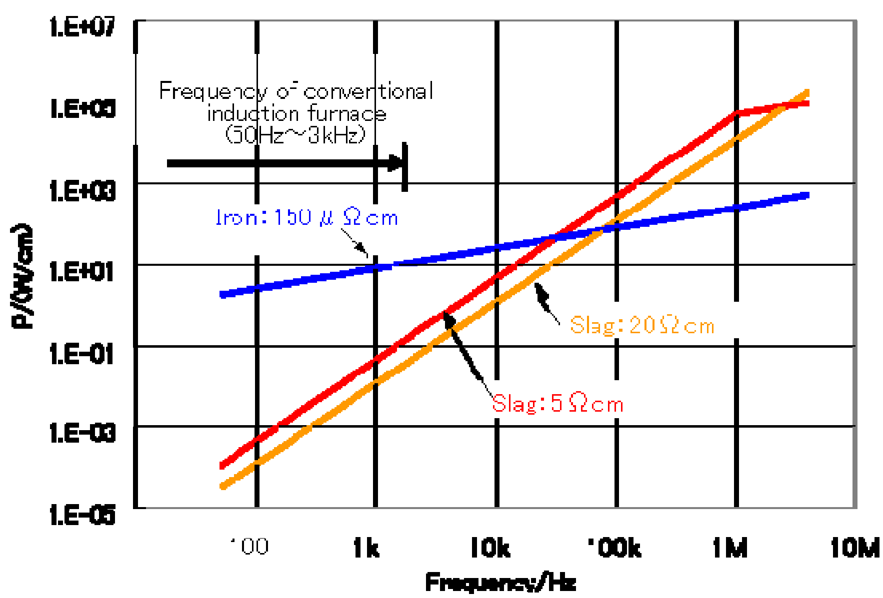

Fig.1. Impressed electric power against frequency

Accordingly, the volume reduction treatment of radioactive solid waste system which can heat both metal and molten ceramics such as grass, concrete, and so on, by UHFIF with very high frequency of more than

100 kHz is developed.

BASIC EXPERIMENT WITH THE BENCH SCALE UHFIF

The specifications of the bench scale UHFIF are shown in Table 1. The coil of bench scale UHFIF was constructed with water cooled copper coil of one turn, because it was operated with very high frequency.

Table 1. The specifications of the bench scale UHFIF

Maximum power	150kW
Maximum frequency	300kHz
Maximum temperature	2000 degree C
Capacity of crucible	10 liter

Experimental conditions of melting test with the bench scale UHFIF are listed in Table 2.

Table 2. Experimental Conditions of melting test with the bench scale UHFIF

<table>
<tr><td rowspan="2">Metal and ceramics simultaneous melting test
Metal : carbon steel
Ceramic : borosilicate glass</td><td>Condition 1</td><td>Volume ratio of metal to ceramics = 1:9</td></tr>
<tr><td>Condition 2</td><td>Volume ratio of metal to ceramics = 5:5</td></tr>
<tr><td>Stainless steel melting test
Metal : stainless steel
Ceramic : borosilicate glass</td><td>Condition 3</td><td>Volume ratio of metal to ceramics = 3:7</td></tr>
<tr><td>Insulator melting test
Metal : stainless steel
Ceramic : borosilicate glass + calcium silicate</td><td>Condition 4</td><td>Volume ratio of metal to ceramics = 3:7
Weight ratio of calcium silicate to glass = 5:5</td></tr>
<tr><td rowspan="2">Aluminum melting test
Metal : carbon steel + Aluminum
Ceramic : borosilicate glass</td><td>Condition 5</td><td>Volume ratio of metal to ceramics = 3:7
Weight ratio of carbon steel to aluminum = 9:1</td></tr>
<tr><td>Condition 6</td><td>Volume ratio of metal to ceramics = 3:7
Weight ratio of carbon steel to aluminum = 8:2</td></tr>
</table>

Metal and ceramics simultaneous melting test

Carbon steel and borosilicate glass were used for melting tests. They were selected as a representative material of ferrous metal and ceramics, respectively. Metal samples and glass samples were mixed to simulate solid wastes, and melted in the bench scale UHFIF. The conditions for mixing ratio of metal and ceramics were

1:9 and 5:5 in volume ratio. In condition 1, a graphite block was added to the sample because the amount of metal, as initial heat source, is little. In the condition 2, only the initially loaded metal was used as a heat source.

In condition 1, all of the initially loaded materials melted completely in 2 hours. And additional materials were loaded several times. After all loaded materials were completely melted, the graphite block was removed, and then molten material was heated up to 1600 degree C. Therefore, even if amount of metal was little, it was confirmed that samples could be molten comparatively easily with graphite in UHFIF.

In condition 2, all of the initially loaded materials melted completely in about 5 hours. However, it was confirmed that samples could be molten without graphite. A cause of time consuming in melting of initial materials was assumed that the power of induction heating of initial metal was weak. This problem is expected to be solved by increasing in an output of a power source.

Stainless steels melting test

In condition 3, stainless steels and borosilicate glass sample were used for initial materials. The volume ratio of metal to glass was 3 to 7. The melting behavior of stainless steels was satisfactory.

Insulator melting test

Insulators exist comparatively a lot in solid wastes as a component of ceramics waste. In condition 4, insulators and carbon steels were used for initial materials. The mixture of 50 wt% of calcium silicate and 50 wt% of borosilicate glass was selected as a representative material of insulator. The volume ratio of metal to insulator is 3 to 7. The melting behavior of Insulators was satisfactory.

Aluminum melting test

Aluminiferous metal exists as a component of metal wastes. In Japan, aluminum metal must not be disposed of in an underground disposal plant, because aluminum metal generates hydrogen gas in alkaline condition with mortar. Therefore, it is necessary for disposal of aluminum wastes to be oxidized or made to alloy with iron. Accordingly, the melting behavior of aluminum added materials were examined.

In conditions 5 and 6, the weight ratio of aluminum to carbon steels was 1 to 9 and 2 to 8, respectively. In both conditions, the volume ratio of metal to glass was 3 to 7. The melting behavior of aluminum added materials was satisfactory. The chemical composition of solidified molten materials was analyzed. Aluminum was not detected in metal layer of solidified material, and almost all aluminum were detected in slag layer. Therefore, almost all of loaded aluminum were oxidized and distributed in slag layer.

Selection of material of crucible

The requirements for crucible are as follows.

High electrical resistance.

High heat resistance at least 1700 degree C.

Good compatibility with basic slag and acid slag

High thermal shock resistance

High mechanical strength

Candidate oxide materials for crucible are evaluated in Table 3. The property of those materials was evaluated in five grades; the larger number, the better property. No oxide material satisfied those properties singly, but those properties were expected to be improved by mixing with each others. The most important property of crucible for in-can melting was breaking resistance, and thermal shock resistance was taken priority. Therefore, silica was selected as main composition of crucible. Alumina was mixed with silica in order to improve corrosion resistance of crucible. In addition, silicon carbide which has effect to improve mechanical strength was added as a modifier. Finally, the crucibles were made with both of two material systems, silica – alumina and silica – alumina – silicon carbide.

Table 3. The results of evaluation of oxide materials for crucible

	Melting point (degree C)	Thermal shock resistance	Corrosion resistance	
			To acid slag	To basic slag
Silica	1723	5	4	1
Alumina	2050	3	5	3
Mullite	1810	4	3	2
Magnesia	2800	1	1	5
Spinel	2100	2	2	4
Lime	2620	1	1	5

Analysis of electromagnetic and thermal behavior in the bench scale UHFIF by computer simulation

Electromagnetic and thermal behavior in UHFIF was calculated in order to obtain data for design of the demonstration scale UHFIF. The calculation condition is shown in Table 4.

Table 4. The condition of computer simulation of electromagnetic and thermal behavior in UHFIF

Number of coil turn	1
Frequency	175kHz
Current	1350A

Employed physical properties of metal layer and slag layer for computer simulation is shown in Table 5.

Table 5. The employed physical properties of metal and slab layer for computer simulation

	Conductivity (S/m)	Density (kg/m^3)	Viscosity (Pa・s)
Slag(CaO-50wt%SiO_2)	32	2600	0.207
Metal(carbon steel)	7.22×10^5	7000	5.28×10^{-3}

The calculated electromagnetic field in UHFIF is shown in Fig.2. When the metal layer and the slag layer coexisted in UHFIF, the magnetic field in the bottom of slag layer was remarkably decreased by skin effect of metal later.

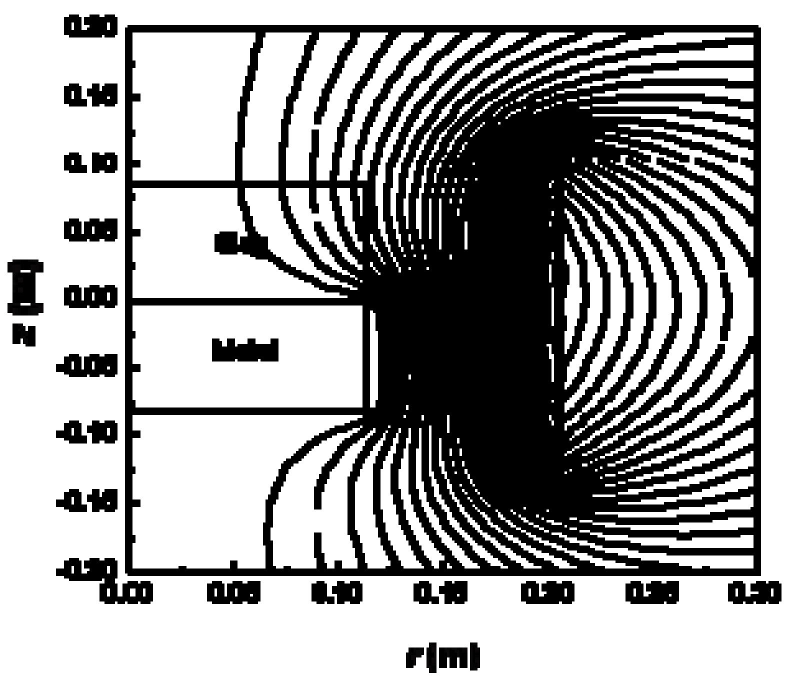

Fig.2. Calculated electromagnetic field in UHFIF

The heat transfer and the flow of molten metal and slag were calculated based on the analysis of the distribution of electromagnetic field.

Design and construction of the demonstration scale UHFIF

The demonstration scale UHFIF was designed and constructed based on the results of examination with the bench scale UHFIF and the analysis by computer simulation. The capacity of crucible was determined to be 30 liters in order to insure feasibility.

Study of frequency, electric power, and the number of coil turn

Frequency, electric power, and the number of coil turn of the demonstration scale UHFIF were studied. Molten metal and slag could be heated to 1500 degree C with primary electric power of 80 kW within 2 hours by assuming the frequency of 170 kHz, the efficiency of power supply of 80 %, and the efficiency of power input of 75%. However, the frequency of 170 kHz was so high that starting of UHFIF became difficult. Therefore, the frequency was lowered to the practical specification.

Necessary electric power, electric current, and voltage for heating molten metal and slag to 1500 degree C within 2 hours were calculated by assuming 3 conditions which was the frequency of 30 kHz, 50 kHz, and 100 kHz. The results of calculation are shown in Table 6. Efficiency of input power increases with frequency, and the electric power could be saved.

Table 6. Necessary specification for each frequency

Frequency (kHz)	30	50	100
Efficiency of input power (%)	65	70	75
Electric power (kW)	90	85	80
Electric current (A)	4811	3910	2826
Voltage (V)	2184	2956	4262

Though the calculated necessary electric power was 80 kW, the electric power was determined to 220 kW in order to leave a margin because of experience with the bench scale UHFIF examination.

Furthermore, the number of coil turn was determined to be 3 for satisfaction of both the upper limit of current density enough to cool coils and the upper limit of voltage enough to prevent sparking. Fig.3. shows the demonstration scale UHFIF.

Fig.3. Exterior view of the demonstration scale UHFIF

EXAMINATION WITH THE DEMONSTRATE SCALE UHFIF

Ferrous metal, Aluminum, and ceramics simultaneous melting test

The composition of melting materials is shown in Table 7.

Table 7. Composition of melting materials

		Condition 7		Condition 8	
		Weight ratio of metal	Weight (kg)	Weight ratio of metal	Weight (kg)
Metal	Carbon steel	9	54	8	36
	Aluminum	1	6	2	9
Ceramics	Borosilicate glass	-	51	-	37

Based on the results of the bench scale UHFIF examination, it was known that aluminum was oxidized to alumina which had high melting point, and distributed to slag layer. Therefore, cryolite was added to samples as a melting point depressor, because alumina made slag layer difficult to melt.

The chemical composition of solidified molten metal and slag were analyzed. Aluminum concentration in metal layer was less than 1 wt %. Before melting, alumina concentration in ceramics was 13 wt %. However, in conditions 7 and 8, alumina concentration in solidified molten slag is 15.7 wt % and 27.7 wt %, respectively. Therefore, it was confirmed that aluminum was oxidized and stabilized by melting with ferrous metal and ceramics with UHFIF.

Elements distribution test

The distribution behavior of elements between molten metal and slag was examined by adding non-radioactive elements to samples as simulated nuclide in TRU wastes. The set of simulated nuclides and added elements are listed in Table 8.

Table 8. Simulated nuclides and added elements

Nuclides in TRU wastes	Simulated non-radioactive elements	
	element	composition
Co-60	Co	Co
Sr-90	Sr	$Sr(NO_3)_2$
Tc-99	Mo	MoO_2
Ru-103/106	Ru	RuO_2
Sn-126	Sn	SnO_2
Cs-134/137	Cs	$CsNO_3$
U isotopes	Ce	CeO_2
Np-237		
Pu isotopes		
Am isotopes		
Cm-244		

The composition of melting materials is shown in Table 9.

Table 9. Composition of melting materials

		Condition 9			Condition 10		
		Initial loaded	Adding loaded	total	Initial loaded	Adding loaded	total
Metal	Carbon steel (kg)	65	68	133	24	30	54
	Aluminum (kg)	-	-	-	23	28	51
Ceramics	Glass (kg)	10	18	28	2.6	3.4	6.0

The results of elements distribution test is shown in Table 10. It was confirmed that Co, Mo, Ru, and Sn were distributed to metal layer, and Sr, Cs, and Ce were distributed to slag layer. Those results were agreed with tendency of standard free energy of formation of oxides, and molten metal and slag were in equilibrium thermodynamically in UHFIF. The standard free energy of formation of oxides is shown in Fig.4. Therefore, behavior of other elements is assumed from its standard free energy of formation of oxides.

Table 10. Results of distribution ratio (%)

		Co	Sr	Mo	Ru	Sn	Cs	Ce
Condition 9	Slag layer	0.2	100.0	1.8	0.5	0.8	99.2	98.9
	Metal layer	99.8	0.0	98.2	99.5	99.2	0.8	1.1
Condition 10	Slag layer	0.4	100.0	1.5	0.9	2.0	99.7	99.5
	Metal layer	99.6	0.0	98.5	99.1	98.0	0.3	0.5

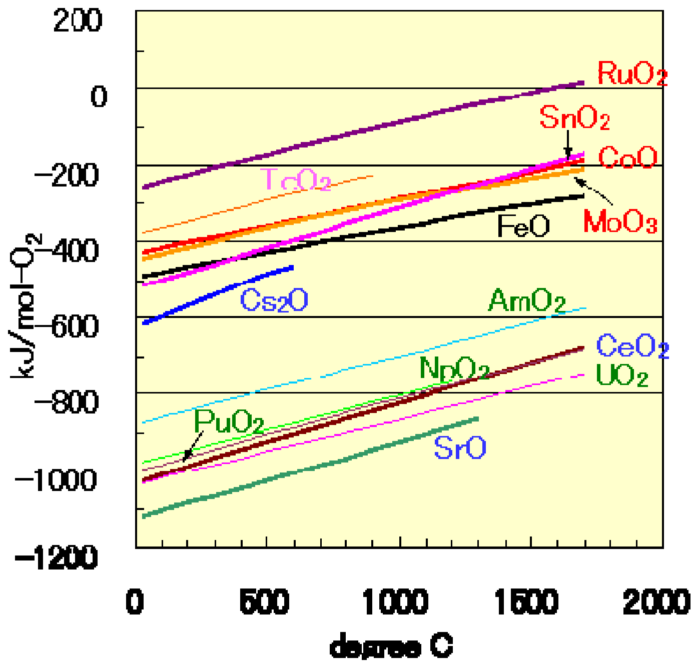

Fig.4. Standard free energy of formation of oxides against temperature calculated by MALT II computer code.

Analysis of electromagnetic and thermal behavior in the demonstration scale UHFIF by computer simulation

The changing in distribution of temperature and velocity vector of molten metal and slag with time for 1000 seconds was calculated. The condition of calculation is shown in Table 11. The flows of molten metal and slag were unsteady from the results of calculation. It was estimated that, in slag layer, natural convection was generated by mixing of downward flow formed at surface and upward flow formed by induction heating at peripheral area and contact heating at interface with metal layer. On the other hand, in metal layer, it was estimated that agitation by electromagnetic power was more dominant than natural convection. Those calculated results were agreed with the observed behavior of molten materials at melting tests.

Table 11. Condition of calculation

Frequency		100 kHz
Current		1425 A
Number of coil turn		3
Volume ratio of metal to ceramics		1 : 1
Calorific value	Metal layer	13.5 kW
	Slag layer	15.3 kW
	Total	28.8 kW

Calculation of homogeneity of molten metal and slag

The necessary time that SiC powder spread homogeneously in molten metal and slag was calculated by assuming that SiC powder put into a point in molten metal and slag.

In molten metal, SiC powder was dispersed homogeneously within 40 seconds, because of effect of agitation by electromagnetic power for mass transfer. In molten slag, 220 seconds were needed to become homogeneous. Therefore, the necessary time of homogeneity of molten metal and slag was 5 minutes. Practically, it was assumed to be enough to retain temperature of molten metal and slag for three times the calculated period, and the homogeneous solidified molten material was able to be made with retention time of 15 minutes.

Analysis of solidified molten materials

The density, composition and surface condition of the solidified molten metal and slag made by the demonstration scale UHFIF was analyzed, the homogeneity of those substances were estimated. The chemical stability of those substances was also estimated by leaching test conforming to shortened MCC-1 method.

The analyzed density and composition are shown in Table 12. and Table 13., respectively. It was confirmed that the solidified molten substances made by UHFIF were homogeneous.

The samples for leaching test were collected from the solidified material made by elements distribution test, and examined by the method conforming to shortened MCC-1 method. The examination condition is shown in Table 14. The ratio of normalized elemental mass loss is shown in Table 15.

In metal layer, the ratio of normalized elemental mass loss of Co, Mo, Ru, and Sn was increase with Fe which was principal component of metal layer. In slag layer, Cs, and Ce was leached with Na, Mg, and Si which was principal component of slag layer. Therefore, the elements simulating nuclides in TRU wastes were retained stably in metal and slag layer.

Table 12. Analysis result of density of a solidified material

Sampling point			Density g/cm^3
	Horizontal	Vertical	
Solidified metal layer	Center	Upper part	7.84
		Middle part	7.84
		Lower part	7.84
	Periphery	Upper part	7.84
		Middle part	7.84
		Lower part	7.84
Solidified slag layer	Center	Upper part	2.62
		Middle part	2.62
		Lower part	2.62
	Periphery	Upper part	2.61
		Middle part	2.62
		Lower part	2.62

Table 13. Analysis result of composition of a solidified material

	Sampling point		Concentration wt%				
Solidified metal layer	Horizontal	Vertical	Cr	Ni	Mn	Al	-
	Center	Upper part	0.1	<0.1	1.1	<0.1	-
		Middle part	<0.1	<0.1	0.9	<0.1	-
		Lower part	<0.1	<0.1	0.8	<0.1	-
	Periphery	Upper part	<0.1	<0.1	0.8	<0.1	-
		Middle part	0.1	<0.1	1.0	<0.1	-
		Lower part	<0.1	<0.1	0.9	<0.1	-
	Coefficient of variance %		-	-	11.6	-	-
Solidified slag layer	Horizontal	Vertical	Al_2O_3	Na_2O	MgO	CaO	FeO
	Center	Upper part	3.5	11.4	3.3	7.0	5.5
		Middle part	3.6	11.7	3.3	7.1	5.5
		Lower part	3.6	11.3	3.3	7.2	5.4
	Periphery	Upper part	3.6	11.5	3.3	7.2	5.5
		Middle part	3.6	11.6	3.3	7.2	5.6
		Lower part	3.6	11.5	3.3	7.3	5.5
	Coefficient of variance %		1.0	1.1	0.0	1.3	1.0

Table 14. Leaching test condition of a solidified material

Sample	Solidified metal layer	Solidified slag layer
Weight (g)	31.0	6.8
Dimension (mm)	40 x 40 x 2.5 t	-
Particle diameter (mm)	-	$75x10^{-6}$-$150x10^{-6}$
Specific surface area (cm^2/g)	36.0	440
SA/V (cm^{-1})	0.65	74.8
Volume of deionized water (cm^3)	55	40
Temperature (degree C)	90	
Period of test (day)	3, 7, 14, 32, 56, 91	

Table 15. Normalized elemental mass loss of a solidified material (mg/cm^2)

Leaching period (day)		3	7	14	32	56	91
Solidified metal layer	Mn	0.025	0.068	0.133	0.370	0.586	0.882
	Fe	0.2630	0.583	1.401	3.601	5.821	8.911
	Co	0.228	0.544	1.275	3.115	4.649	7.445
	Mo	0.075	0.248	0.472	1.257	1.280	2.272
	Ru	0.038	0.244	0.260	<0.03	<0.03	0.422
	Sn	0.162	0.409	0.655	3.096	4.319	6.059
Solidified slag layer	Na	0.012	0.016	0.020	0.027	0.033	0.042
	Mg	0.005	0.007	0.007	0.008	0.010	0.010
	Al	0.005	0.007	0.007	0.010	0.012	0.013
	Si	0.003	0.003	0.004	0.004	0.005	0.006
	Ca	0.006	0.007	0.008	0.010	0.013	0.014
	Fe	0.008	0.008	0.011	0.012	0.014	0.015
	Sr	0.008	0.008	0.009	0.012	0.014	0.015
	Cs	0.008	0.007	0.008	0.010	0.013	0.013

Design of the full scale UHFIF

The full scale UHFIF was designed based on the results of examination with the demonstrate scale UHFIF. An object of the full scale UHFIF was to produce TRU waste packages of 200 liter drum. For this reason, the crucible capacity for the full scale UHFIF was determined to be 100 liter.

Operating condition of the full scale UHFIF

It is assumed that UHFIF was operated only on the day shift, and operating time of UHFIF was determined to be as follows. The temperature rising time was 90 minutes, and the time for charging of additional loading materials is 120 minutes.

Basic specification of the full scale UHFIF

Impressed electric power in metal layer and slag layer were calculated based on operating conditions and parameters shown in Table 16.

Table 16. Parameters for calculation of impressed power in UHFIF

Frequency	30 kHz, 50 kHz, 75kHz
Number of coil turn	3, 4, 5
Volume ratio of metal to ceramics	1:9, 9:1

The results of calculation are shown in Table 17. The impressed electric powers were almost equal with each others. However, it was difficult to make a power source because electric power was too high assuming by 3 coil turns, and voltage was too high assuming by 5 coil turns. Therefore, the specification of the full scale UHFIF was determined practically to be 50 kHz of frequency and 4 coil turns.

Table 17. Results of calculation of specification of the full scale UHFIF

Crucible capacity	100 liter								
Volume ratio of metal to slag	9:1								
Operating electric power	270 kW								
Number of coil turn	3			4			5		
Frequency(kHz)	30	50	75	30	50	75	30	50	75
Current (A)	6573	5721	5101	4899	4260	3807	3900	3399	3032
Voltage(V)	3220	5928	6247	4246	6555	8249	5290	7684	10281

Analysis of electromagnetic and thermal behavior in the full scale UHFIF by computer simulation

The electromagnetic and thermal behavior in the full scale UHFIF was analyzed by calculation. The flows of molten metal and slag were unsteady similarly to the demonstrate scale UHFIF.

The necessary time of homogeneity of metal and slag layer was calculated. Molten slag and metal need 63 and 56 seconds to become homogeneous, respectively. Therefore, the homogeneity of solidified molten materials is expected to be ensured, even if the crucible capacity is scaled up to be 100 liter. From the above, the full scale UHFIF can be designed and constructed practically.

CONCLUSION

The conclusions are as follows.

Experimental melting data were collected with the bench scale and demonstrate scale UHFIF.

It was demonstrated that metal and ceramics were able to be melt and stabilized simultaneously by induction heating with UHFIF.

The solidified molten materials were confirmed to be reliable, because of their homogeneity and retentiveness of nuclides.

It was demonstrated that melting behavior was able to be predicted with computer simulation, and the design of progressed UHFIF was practicable, because of agreement between experiment results and calculated results.

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REFERENCES

1. N.SASAKI et al., Proceedings of 2006 Fall Meeting of the Atomic Energy Society of Japan, A11, (2006) (in Japanese).
2. Y.MIYAMOTO et al., Proceedings of 2006 Fall Meeting of the Atomic Energy Society of Japan, A12, (2006) (in Japanese).
3. N.SASAKI et al., Proceedings of 2007 Fall Meeting of the Atomic Energy Society of Japan, N33, (2007) (in Japanese).
4. T.NISHIKAWA et al., Proceedings of 2007 Fall Meeting of the Atomic Energy Society of Japan, N34, (2007) (in Japanese).
5. Y.AOYAMA et al., Proceedings of 2007 Fall Meeting of the Atomic Energy Society of Japan, N35, (2007) (in Japanese).
6. Y.AOYAMA et al., Proceedings of 2008 Annual Meeting of the Atomic Energy Society of Japan, H37, (2008) (in Japanese).
7. N.SASAKI et al., Proceedings of 2008 Annual Meeting of the Atomic Energy Society of Japan, H38, (2008) (in Japanese).