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Treatment of radioactive wastes by plasma incineration and vitrification for final disposal

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Abstract

Thermal plasma technology takes the advantages to incinerate the combustible parts of radioactive wastes for volume reduction and to vitrify the noncombustible counter parts simultaneously into glassy slags with very low leaching rate. For developing the proper plasma processes to treat various waste forms, a crucible-type plasma melter and a 10 kg h^{-1} plasma furnace system are built at the Institute of Nuclear Energy Research (INER). Both systems are fired by the home-made 100 kW non-transferred plasma torches. The maximum operation temperatures of the small plasma melter, and the 10 kg h^{-1} plasma furnace are 1700°C and 1650°C, respectively. Glassy or ceramic slags with high quality are obtained from the processing of simulated radioactive wastes by plasma torch. The compressive strengths of these slags are greater than 800 kg cm^{-2}, and the leaching indices of several elements are between 8 and 15, which are all greater than the ROC regulated values for waste forms (i.e. 15 kg cm^{-2} and 6). These promising results encourage INER to further develop plasma system with higher capacity and plasma processes for the treatment of low level radioactive wastes. Future efforts shall be concentrated on the studies of system reliability, secondary waste minimization and volatile radionuclides elimination in off-gas.

Keywords: Plasma torch; Vitrification; Radioactive waste

1. Introduction

Radioactive wastes from nuclear power plants due to normal operation and maintenance works, hospitals and research institutes due to isotope applications are generated daily with a considerable amount. To permanently dispose of these low-level radioactive

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wastes safely and cost effectively, the radioactive wastes should be transformed into the physical and chemical compounds suitable for radionuclides immobilization with maximum volume reduction. Such treatments usually require pretreatment steps to separate the wastes into combustibles, non-combustibles and metals. Combustibles are normally treated by incinerator, with the ash then being mixed with cement and stored in containers (e.g. drums). Non-combustibles are either compacted or directly converted into a solid matrix. Metals are melted and cast into containers. All these existing process technologies have the following disadvantages: (1) different types of waste have to be separated, manipulated and treated with different equipment; (2) volume reduction and vitrification are not achieved within one step [1,2]. But, thermal plasma process bears the capabilities to incinerate the combustibles of radioactive wastes for volume reduction and to vitrify the non-combustibles simultaneously into glassy slags or metal ingots by using the same equipment and in one step [3,4].

Institute of Nuclear Energy Research (INER) has been commissioned by Atomic Energy Council (AEC) of the Republic of China to be responsible for the treatment of all the domestic low-level radioactive wastes except those from nuclear power plants since 1978. The combustible radioactive wastes are treated by a controlled air incinerator with capacity of 40 kg h^{-1}, while the residual ash and non-combustible radioactive wastes are temporarily stored in INER. In order to treat these radioactive wastes effectively and solve the storage problem, INER has commenced developing plasma facilities and plasma processes for various waste forms since 1994. Non-transferred DC plasma torch with 100 kW output power named as INER-100NT DC plasma torch is developed and served as heat source for plasma vitrification. In addition, a crucible-type plasma melter and a 10 kg h^{-1} plasma furnace system are also established for the studies of plasma vitrification of surrogate waste. Preliminary results from vitrification tests of simulated wastes show that thermal plasma processing can produce glasses or ceramic waste forms which are suitable for final disposal.

Thermal plasma process is similar to any other high-temperature process in which high quality slags are produced, in that some particulates and volatile and/or semi-volatile elements such as cesium will escape from the melts [5,6]. An off-gas treatment system is necessary to capture these particulates and volatile/semi-volatile elements. INER has experience of designing and operating off-gas treatment systems for rad-wastes incineration for many years, and a 250 kg h^{-1} pilot-scale plasma furnace plant for low level radioactive waste treatment is to be built in INER. The study on radionuclide inventory of off-gas control which is not included in this paper shall be performed thoroughly in the pilot plant.

2. INER-100NT DC plasma torch

High-power plasma torch is the heart of plasma waste-treatment technology. Cost-effective DC plasma torch is more attractive than RF plasma torch to be used for waste treatment. The INER-100NT plasma torch is a Linde-type arc heater [7,8] which consists of two water-cooled electrodes and a swirl flow generator. The front electrode is a two-end opened copper tube with 25 mm in inside diameter, 45 mm in outside diameter, and 394 mm in length. The rear electrode is a one-end opened copper tube with 25 mm in inside diameter, 35.7 mm in outside diameter, and 194 mm in length. These two electrodes are electrically insulated with 5-mm separation between the open end of rear electrode and one open end of front electrode, the other open end of front electrode is the exit of heated gas. The gas ring of the swirl flow generator which is made of stainless steel has four tangential holes with diameter of 2 mm. The inside diameter and outside diameter of the gas ring are 100.4 mm and 108 mm, respectively. The swirl flow coming out of the gas ring can stabilize the arc inside the electrodes and cool the internal surfaces of electrodes effectively. The total length of INER-100NT plasma torch is 62 cm, while the total weight is about 50 kg.

The plasma torch is powered by a constant current DC power supply with a high-frequency high-voltage starter. The open circuit voltage of the power supply is 875 V, while the maximum output current is 200 A. The front electrode is connected to positive voltage, and the rear electrode through a 10-turn magnetic coil wound around itself is connected to negative output end of the power supply. Axial magnetic field, generated by the magnetic coil when arc current is conducting, makes the arc root attached on the internal surface of rear electrode spin rapidly, thus, the thermal loading of cathode surface and the erosion rate of cathode are greatly reduced. The plasma torch is ignited under current setting at 50 A and argon atmosphere with flow rate in between 60 and 80 *sl* $\min^{-1}$, then the working gas is gradually changed into air. The output power of plasma torch can be increased by increasing the arc current and gas flow rate. Fig. 1 shows the picture of INER-100NT plasma torch during operation under the following conditions. The arc current is 200 A, the voltage drop between cathode and anode is 600 V, and the air flow rate is 700 *sl* $\min^{-1}$. The length of plasma flame under these operation conditions is about 30 cm. Energy conversion efficiency defined as the ratio of output thermal power and input DC electric power is an important parameter to

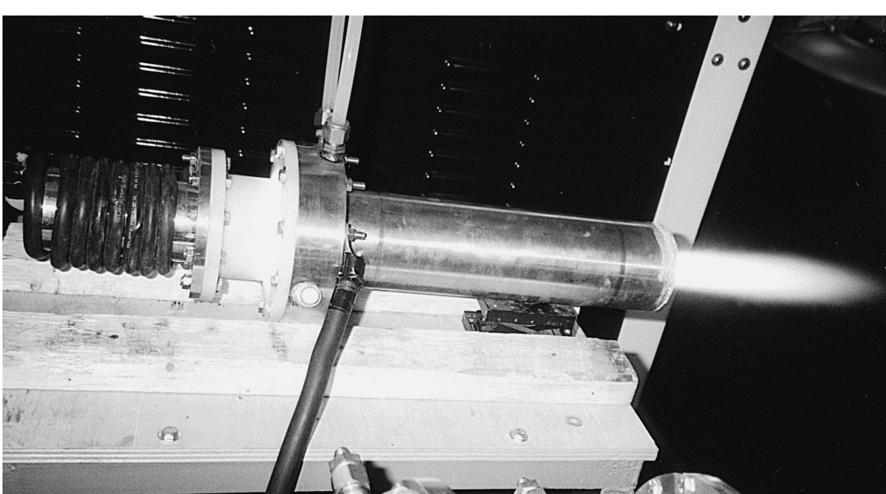

Fig. 1. Photo of INER-100NT DC plasma torch during operation.

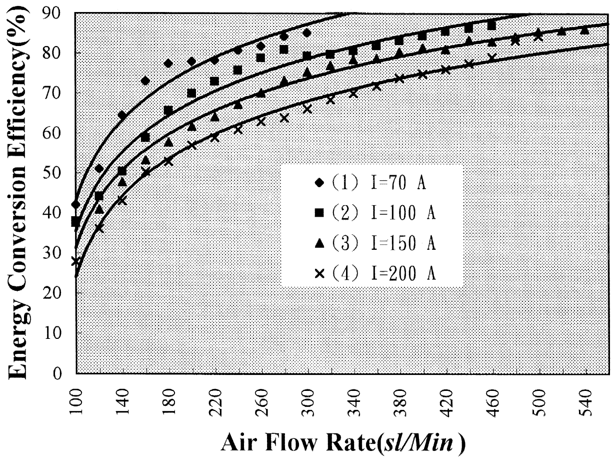

Fig. 2. Energy conversion efficiency of INER-100NT DC plasma torch vs. air flow rate and current.

evaluate a plasma torch system. Fig. 2 shows the energy conversion efficiency curves of INER-100NT DC plasma torch at four arc currents (i.e. $I = 70$, 100, 150, 200 A) vs. air flow rate. The energy conversion efficiency increases as the air flow rate increases, and reaches to a maximum value within the operation range. The characteristics of INER-100NT plasma torch is listed as follows:

- Electric power range = 20–120 kW
- Maximum arc current = 200 A
- Maximum working voltage = 600 V
- Air flow rate = 100–700 *sl* $\min^{-1}$
- Maximum energy conversion efficiency = 87%
- Average temperature of heated gas = 5000–6000°C
- Enthalpy of heated gas = 6–10 MJ kg^{-1}
- Cathode/Anode lifetime > 150/500 h

3. Plasma incineration and vitrification systems

3.1. 10 kg h^{-1} plasma furnace system

3.1.1. Process flow diagram

The process flow diagram of INER's 10 kg h^{-1} plasma furnace system is shown in Fig. 3. The primary reaction chamber (PRC) and secondary combustion chamber (SCC) are all fired by one INER-100NT plasma torch, and can be heated up to their maximum operation temperatures, i.e. 1650°C and 1350°C, respectively. As PRC and SCC reach to their own feeding temperatures, i.e. 1500°C and 1200°C, respectively, surrogate wastes are fed into PRC through the feeder at the top of furnace. Combustibles are pyrolysed into small organic molecules which are then drawn into SCC to be oxidized completely by adding proper amount of air into SCC. The retention time of small organic molecules

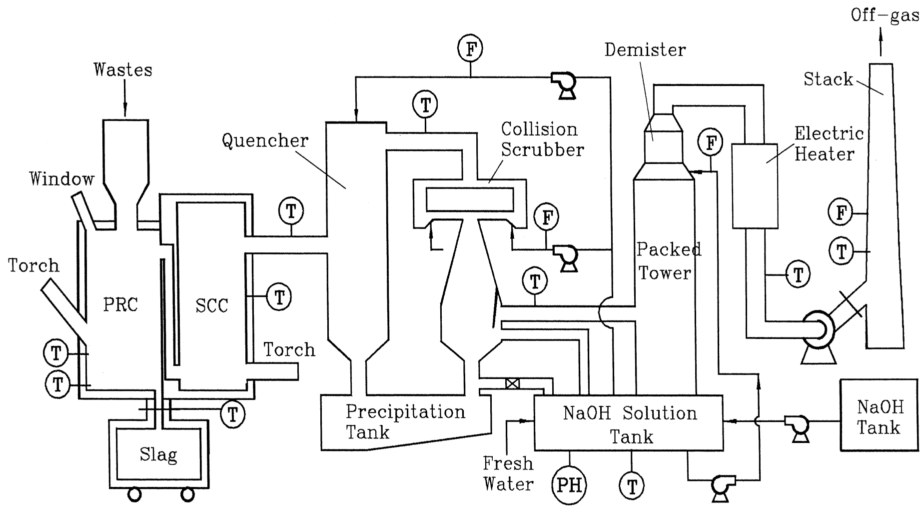

Fig. 3. The process flow diagram of 10 kg h^{-1} plasma furnace system.

in SCC is more than 1 s. Noncombustibles such as Fe_2O_3, Al_2O_3, SiO_2, CaO, etc. and metals are melted in PRC, and then the molten slags or metals flow into the collection chamber at the bottom of furnace. To keep the molten slags sufficiently fluid during discharge, the flow passage is heated by another torch and maintained at a temperature higher than 1600°C. High temperature off-gas coming from secondary combustion chamber is rapidly quenched by spaying NaOH solution into the quencher. The pH value of NaOH solution is maintained at 8.5 ± 0.5 by a PID pH controller and a metering pump. The collision scrubber removes most of dusts and part of acid gases. Then, the off-gas goes through the packed tower, and acid gases are eliminated. After passing the demister to remove most of water drops greater than 3 μm, the scrubbed off-gas is heated to a temperature above its dew point ($\sim$ 81°C) by the electric heater. Finally, the clean off-gas is drawn by induced draft fan to the stack, and discharged into atmosphere. The plasma furnace system is maintained at a pressure ranging between 1 in. to 5 in. water column negative by the blower. The blower is equipped with a frequency converter to help maintain constant pressure during processing to prevent the materials inside the plasma furnace from escaping into working environment. To capture volatile or semi-volatile radionuclides in off-gas stream when hot tests are to be performed, HEPA filters should be inserted between the electric heater and the blower of this process flow diagram.

3.1.2. The structure of furnace

Two vertical cross-section views of the 10 kg h^{-1} plasma furnace are shown in Fig. 4. The outer shell of the furnace is made of 8-mm stainless steel, and all the surfaces except the bottom surface are cooled by circulating water. The hearth is constructed by using the proper combination of 1800°C and 1650°C standard high alumina firebrick, 1400°C insulation brick, and 1260°C glass–fiber plate with 2.5 cm in thickness. The

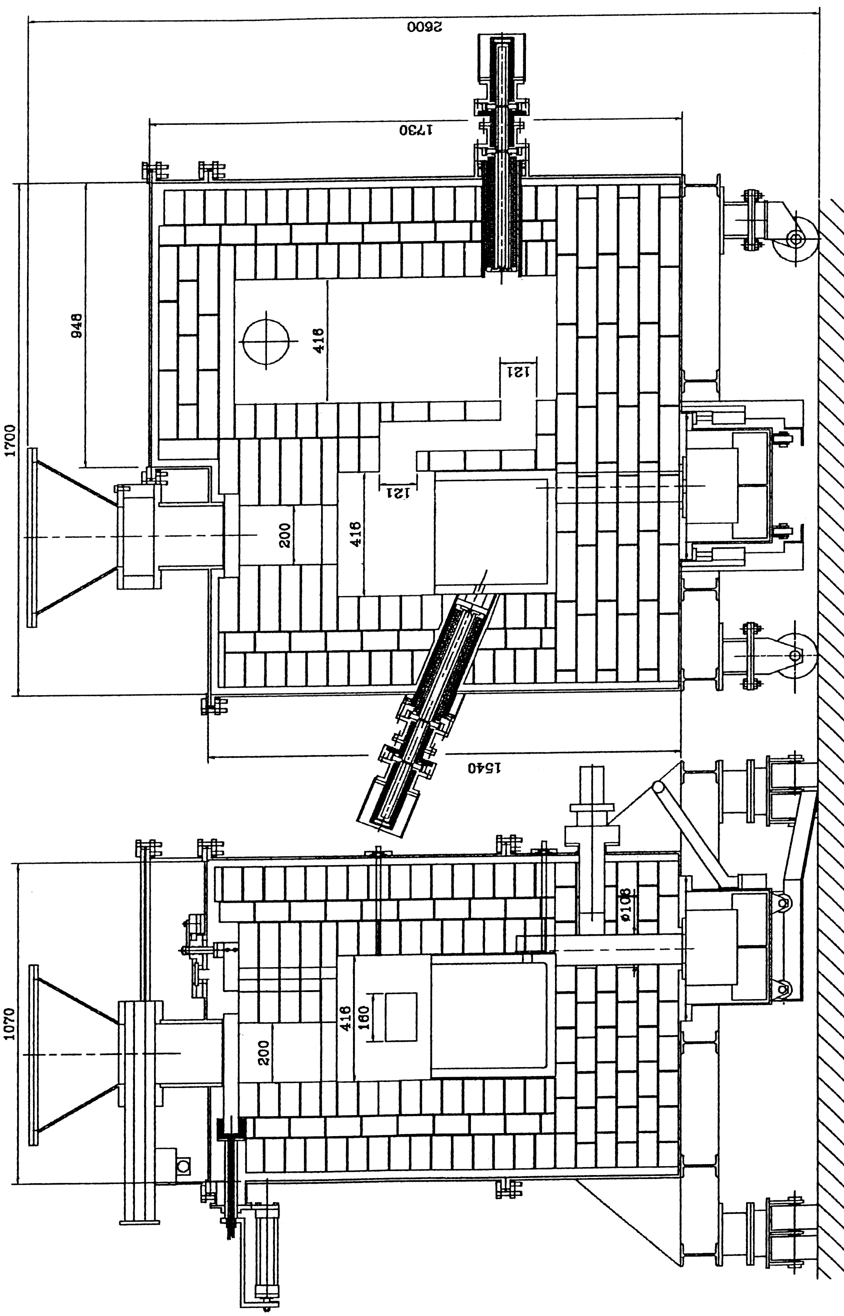

Fig. 4. The vertical cross-section views of the 10 kg h^{-1} plasma furnace.

internal volume of PRC and SCC are $41 \times 41 \times 80$ cm^3 and $41 \times 41 \times 110$ cm^3, respectively. One square crucible is placed inside PRC for the purpose of vitrification. The crucible is made of castable mix with high alumina contents, and is sintered above 1500°C before being put inside PRC. The internal dimension of the crucible is 35 cm × 35 cm × 37 cm, and its thickness is 3 cm. There is an opening ($\phi = 6$ cm) through crucible wall, which is located 20 cm above the bottom of the crucible and inclined at an angle of 22° with horizontal line. This opening allows the heated gases or plasmas to reach the wastes inside the crucible, and process them directly. Another opening ($\phi = 3$ cm) located 5 cm above the internal bottom surface of the crucible allows the melt to flow into slag collection chamber. The feeding door is controlled by two gates to feed the wastes into PRC without sucking excess air. One window at the top of PRC can be used to monitor the dynamics inside the furnace. A tunnel having 16 cm × 12 cm cross-section inside the adjacent wall of PRC and SCC allows fuel gases from PRC to be sent into SCC for combustion. All the temperatures of PRC, outer surface of crucible, SCC, and slag passage are monitored by using B-type thermocouples (100–1800°C).

3.1.3. Pattern monitoring and control system

The pattern monitoring and control system as shown in Fig. 5 is consisted of one personal computer Pentium 100 with 16M RAM, and a series of analog and digital I/O modules. The I/O modules used are ADAM-4000 series remote data acquisition and control modules developed by Advantech. The softwares used include: (1) operation platform: DOS 6.22 + Windows 3.1 (Chinese Version), (2) pattern control software: GENIE-data acquisition and control software developed by Advantech, and (3) application programs: user programs developed by INER based on GENIE software. Two application programs have been developed by INER for monitoring the parameters of

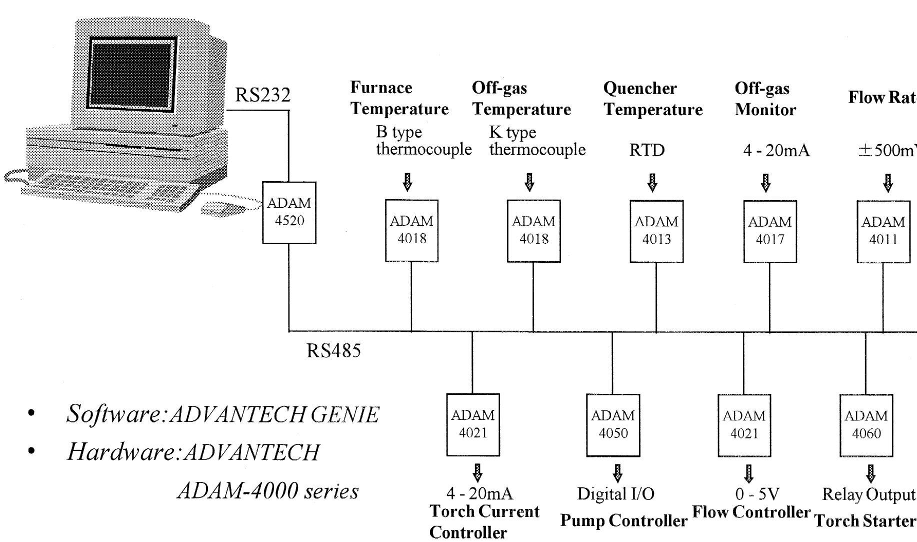

Fig. 5. Structural diagram of pattern monitoring and control system.

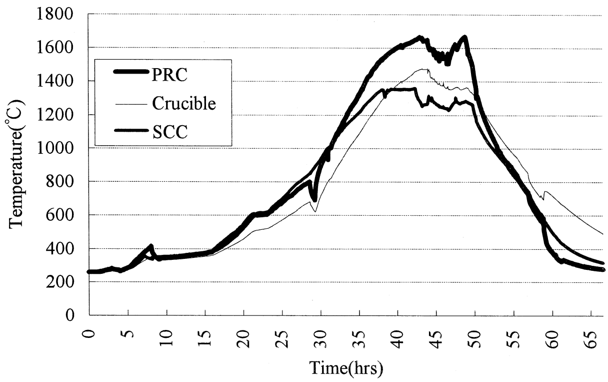

Fig. 6. Typical heating curves of 10 kg h^{-1} plasma furnace system.

plasma furnace system and controlling the plasma torches. PLASMA9.GNI system monitoring program has four display frames (DISP 1-4). DISP 1 shows the schematic flow diagram of 10 kg h^{-1} plasma furnace system, and displays system parameters including temperatures of process units, flow rates of NaOH solution and off-gas, and off-gas concentrations (O_2, CO, NO, NO_2, SO_2, HCl, THC, dust, opacity, etc.). DISP 2 shows the trend curves of 10 important process temperatures and displays the digital data. The data are updated and recorded every 1 s, and the time span of trend curves is 150 min. DISP 3 shows three historical tracking curves of the temperatures of PRC, crucible outer surface and SCC. All those data within 15 days can be reviewed at any time. DISP 4 shows the off-gas concentration curves generated by those data received from off-gas analyzers. Flowc4.GNI torch control program can be used to control the ignition and stopping of plasma torches, to change the working gases of torches, to control the flow rates of the working gases, and to display torch voltages and currents. All the torch parameters can be recorded immediately and displayed on trend curves.

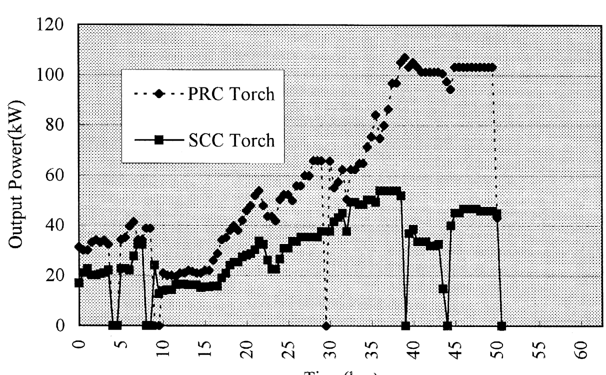

Fig. 7. Torch power corresponding to Fig. 6.

Fig. 8. Photo of BF-B slag ($28 \times 54 \times 49$ mm^3).

3.2. The crucible-type plasma melter

The crucible-type plasma melter is used for the vitrification testing of small amount surrogate wastes in relatively short time. The outer dimension of this small melter is $43 \times 40 \times 45$ cm^3. The melter is constructed by 3-mm SUS304 stainless steel, 1260°C glass–fiber insulation plate, and 1800°C high-alumina firebrick to form an internal volume of $23 \times 23 \times 28$ cm^3. INER-100NT DC plasma torch placed on the top of the melter is served as arc heater. The off-gas is drawn from the opening at the bottom of the melter and sent to the off-gas treatment system of the 10 kg h^{-1} plasma furnace system. Surrogate wastes in crucibles can be placed inside the melter from the front door, and gradually heated up by controlling the output power of the plasma torch. The temperature of the melter is measured by a B-type thermocouple. The maximum operation temperature of this small melter is restricted to 1700°C [9].

4. Experimental results

4.1. Plasma system testing

The typical heating curves of the 10 kg h^{-1} plasma furnace is shown in Fig. 6. PRC reaches to 1670°C by spending 44 h. SCC is heated to 1350°C in 37 h. The average

Table 1
Chemical composition of slags (wt.%)

Test no.	Oxide							
	SiO_2	Na_2O	CaO	K_2O	SO_3	MgO	Fe_2O_3	Al_2O_3
E-22	57.37	8.86	23.43	0.88	0.33	3.11	1.14	4.88
E-23	51.19	7.05	31.29	0.85	1.70	2.84	1.73	3.35
E-24	46.43	6.02	31.87	0.70	1.75	2.67	1.99	8.57
E-26	18.56	5.06	34.52	0.61	2.51	1.84	2.08	34.82
BF-B	60.98	9.31	0.39	4.12	–	0	3.33	21.87

heating rate is about 0.62°C min^{-1}. There is a minimum in the PRC curve at the center of the highest temperature region, which is caused by the feeding of surrogate wastes due to heat absorption of the wastes. The corresponding output power of plasma torch is shown in Fig. 7. To maintain the maximum temperatures of PRC and SCC at 1670°C and 1350°C, the responsible plasma torches must deliver 105 kW and 50 kW electric power respectively. The small plasma melter can be heated up at a higher heating rate in between 6 and 9°C min^{-1}, and 40 kW plasma power is enough to maintain the highest temperature at 1700°C.

4.2. Vitrification of surrogate wastes

4.2.1. Feldspar powder mixed surrogate waste

Feldspar powder mixed surrogate waste (FPMSW, 37.13 kg), which is consisted of feld spar powder (33.84 kg, 91.2 wt.%), polyethylene bottles (1.18 kg, 3.1 wt.%), and iron cans (2.11 kg, 5.7 wt.%), and packed in 11 PE bottles and 18 iron cans, is served as simulated waste for vitrification testing in the 10 kg h^{-1} plasma furnace system. As PRC and SCC reach 1650°C and 1350°C, respectively (see Fig. 6), FPM surrogate wastes are fed into furnace at an average rate of 10 kg h^{-1}. High quality slag in black color shown in Fig. 8 and denoted by BF-B is obtained after vitrification. The compressive strength of BF-B slag is about 1225 kg cm^{-2}, and its composition is listed in Table 1. Fig. 9 shows the X-ray diffraction pattern of BF-B slag, and reveals that the slag is in amorphous structure.

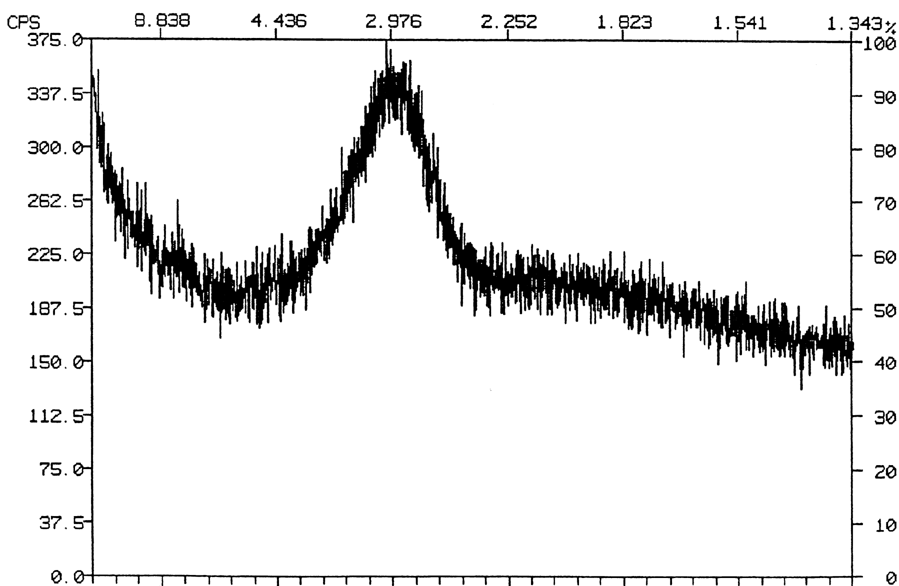

Fig. 9. The X-ray diffraction pattern of BF-B slag.

Table 2
Plasma vitrification conditions and results of cement-solidified sodium sulfate simulated wastes

Test no.	Waste / glass weight ratio	Process temperature (°C)	Residence time (min)	Weight reduction ratio (WR)	Volume reduction ratio (VR)	Slag density (g cc^{-1})	Compressive strength (kg cm^{-2})
E-22	1:1	1430	40	0.69	1.10	2.75	1218
E-23	2:1	1430	30	0.85	1.44	2.92	1356
E-24	10:3	1500	30	0.98	1.68	3.10	845
E-26	1:0	1580	30	1.68	2.77	2.78	1891

4.2.2. Sodium sulfate cement-solidified surrogate wastes

Sodium sulfate concentrate generated from BWR nuclear power plant is usually solidified by Portland type II cement in Taiwan, and transported to temporary disposal site in an isolated island after more than 28 days of curing. Recently, one reveals the fact that some of these cement solidified low-level waste drums are fractured into powders or small pieces, or in bad shape after about 20 years storage. If the fractured cement-solidified wastes are repacked by cement solidification or repacked directly in special containers, the final volume of these repacking wastes increases more than 50%, and incurs the storage problems due to limited space of the temporary disposal site. Since thermal plasma can melt any material, INER proposes to treat these fractured cement-solidified wastes by using plasma technology for volume reduction and vitrification. Plasma vitrification of cement solidified wastes which has never been performed before to authors' knowledge are worth further studying, and the test results could be useful for the community of rad-waste treatment.

Sodium sulfate cement-solidified surrogate wastes (SSCSSW) are prepared by mixing 11 wt.% Na_2SO_4, 51 wt.% Portland type II cement, and 38 wt.% H_2O, then the mix is poured into PE containers ($\phi = 50$ mm, height = 100 mm) and cured for at least 28 days. The density of SSCS surrogate waste is about 1.8 g cm^{-3}. To study the effect of glass additive on slag quality and processing temperature, four conditions with weight ratio of SSCS surrogate waste and glass additive by 1:1 (E-22), 2:1 (E-23), 10:3 (E-24), and 1:0 (E-26, no additive) are tested. These surrogate waste mixtures are placed in small crucibles ($\phi = 63$ mm $\times$ 105 mm, 10 wt.% Cr_2O_3 and 90 wt.% Al_2O_3), and vitrified in the small plasma melter. The vitrification conditions and results are listed in Table 2. The melting point, weight reduction ratio, and volume reduction ratio decrease

Table 3
Average leaching indices of slags generated from plasma vitrification of cement-solidified sodium sulfate surrogates

Test no.	Element							
	Si	Na	Ca	K	S	Mg	Fe	Al
E-22	11.7	10.9	10.3	11.6	8.69	9.78	14.7	12.0
E-23	11.3	10.9	10.1	11.6	9.26	10.3	15.3	12.5
E-24	11.0	10.6	9.92	11.7	9.08	10.2	15.2	12.4
E-26	11.6	9.53	10.1	11.2	8.15	10.3	15.2	11.8

Table 4
Average leaching rate of slags generated from plasma vitrification of cement-solidified sodium sulfate surrogates (g cm^{-2} day^{-1})

Test no.	Element							
	Si	Na	Ca	K	S	Mg	Fe	Al
E-22	4.19×10^{-6}	9.80×10^{-6}	2.08×10^{-5}	4.62×10^{-6}	2.35×10^{-5}	6.72×10^{-6}	2.0×10^{-8}	5.22×10^{-7}
E-23	8.71×10^{-6}	1.06×10^{-5}	2.72×10^{-5}	3.95×10^{-6}	2.52×10^{-5}	7.61×10^{-6}	2.0×10^{-8}	6.57×10^{-7}
E-24	1.00×10^{-5}	1.05×10^{-5}	2.39×10^{-5}	3.15×10^{-6}	2.64×10^{-5}	6.94×10^{-6}	2.0×10^{-8}	6.56×10^{-7}
E-26	3.47×10^{-6}	5.29×10^{-5}	2.56×10^{-5}	6.30×10^{-6}	8.71×10^{-5}	6.68×10^{-6}	2.0×10^{-8}	1.23×10^{-6}

as the glass additive percentage increases. The compressive strengths of these four slags are 1218, 1356, 845, and 1891 kg cm^{-2}, respectively. The volume reduction ratios (VR) of these tests range from 1.1 to 2.77, while the weight reduction ratios (WR) are between 0.69 and 1.68. The densities of the slags are between 2.75 g cm^{-3} and 3.1 g cm^{-3}. Average leaching indices and average leaching rates are measured by using ANS 16.1 standard testing method [10] through 10 times sampling within 90 days. Table 3 shows the average leaching indices of these slags for eight elements, which are in the range of 8 and 15. Table 4 shows the average leaching rates of the slags which range from 8.7×10^{-5} to 2.0×10^{-8} g cm^{-2} day^{-1}. Chemical compositions of the slags analyzed by ICP method are listed in Table 1. X-ray diffraction patterns show that slags from E-22 and E-23 tests are in amorphous structures, while slags from E-24 and E-26 tests are in microcrystalline structures. Fig. 10 shows the X-ray diffraction pattern of the slag from E-26 vitrification test without glass additive. Though the slags are in different structures, but the average leaching indices are no significant difference.

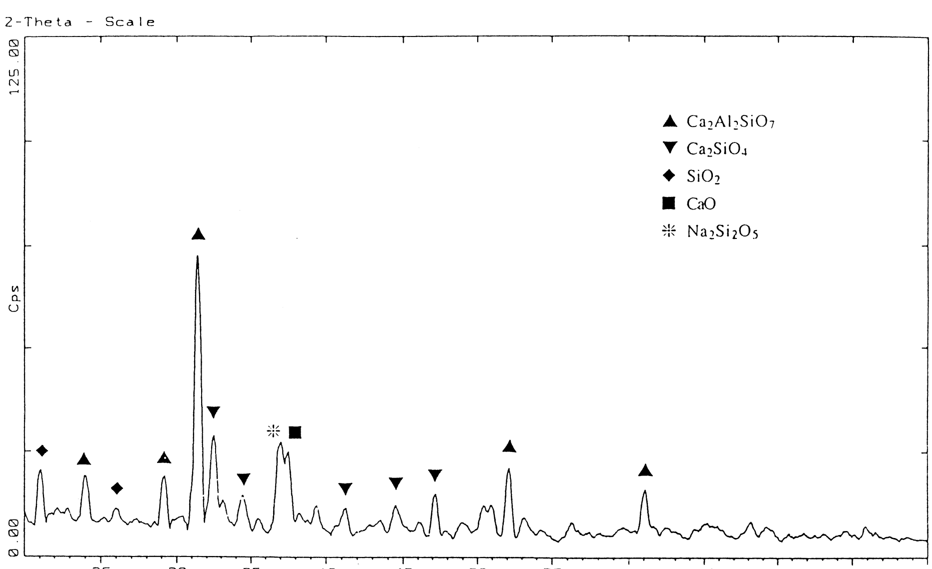

Fig. 10. The X-ray diffraction pattern of E-26 slag.

5. Discussion

Institute of Nuclear Energy Research has established some facilities including INER-100NT DC plasma torches (100 kW), a small plasma melter (1700°C), and a 10 kg h^{-1} plasma furnace system (1650°C). Preliminary vitrification testing of surrogate wastes shows that thermal plasma processing is a robust innovative technology for the treatment of low level wastes. Sodium sulfate cement-solidified surrogate wastes can be vitrified easily by plasma torch, and be converted into slags with high quality. The compressive strengths of the vitrified slags are all greater than 800 kg cm^{-2}, and their leaching indices for eight elements are between 8 and 15, which are all greater than the ROC regulated values for radioactive waste forms (i.e. 15 kg cm^{-2} and 6). These promising results encourage INER to develop high capacity plasma system and plasma processes for the treatment of low level radioactive wastes. A 250 kg h^{-1} pilot-scale plasma furnace plant for low level radioactive waste treatment is under construction in INER [11]. A wet off-gas system modified from the existing incineration systems is expected to be used for the plasma vitrification treatment. To prove that the INER's plasma processing system is suitable for long-term and safe operation under the radioactive environment, much efforts shall be placed on the studies of extending the electrode lifetime of plasma torch measuring radionuclides distributions of plasma system and recycling the secondary wastes into plasma furnace, etc.

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