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Plasma treatment of a simulated low-level radioactive waste

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1. Background and Goal of the present work

According to the Programa Nacional de Gestión de Residuos Radiactivos (2020), 90% of the radioactive waste produced in Argentina is low-level waste. Since these materials occupy a lot of space, resulting in a challenge for the sustainable application of the nuclear energy, treatment techniques have been developed to manage them efficiently. One of these techniques is thermal plasma gasification, which involves heating up waste in a special oven using ionized gas. The process's high temperatures enable the treatment of a wide range of materials, resulting in a volume reduction of nearly 100% (Ojovan, 2011). This work presents an experiment on gasification by thermal plasma using a simulated low level radioactive waste (SLLW) to analyze its volume reduction and reaction products.

2. Materials and methods

2.1 Experimental set-up

The experiments were conducted using a commercial Hypertherm plasma cutting torch, which was connected to its Powermax 105 model source. The Powermax 105 has an output voltage of 160 VDC and a variable current between 30 - 105 amps. For this work, a fixed current of 30 amperes and a power of 4.8 kW was used in all the experiments. Air was used as a plasmatic gas, and the torch was water-cooled to prevent overheating of the internal wires. The compressed air for the torch was supplied using a Kaesser AirCenter SX8 model compressor. The compressor had a working pressure of 8 bar and a flow rate of 8-9 kg/h.

A copper anode with a diameter of 38 mm and a length of 55 mm is utilized to connect the electric arc from the torch. The anode is positioned directly in front of the torch, separated by a distance of 6-9 mm. The refrigeration system is manually controlled using valves and flowmeters and monitored by type K thermocouples that sense the temperature of the inlet and outlet water at each refrigeration point.

Effluent gas monitoring was conducted at the reactor outlet using a Horiba model PG-350 portable gas detector.

2.2 Sample preparation

The experiments were conducted using a simulated low-level radioactive residue, and the preparation of this residue was done using nitrile gloves and laboratory cleaning paper. These materials were chosen because they are commonly found in this type of waste. Table 1 shows the proportions used. The initial volume of these materials was 9000 cm^3.

Table 1. Composition of simulated low-level radioactive waste.

Simulant material	Waste composition	Percentage [%]
Nitrile gloves	200	88.9
Paper	25	11.1

Due to the dimensions of the reactor, the nitrile gloves were ground using a Fristch model Pulverisette 25 tungsten carbide blade grinder and placed in 20 paper wrappers (25 g) of 20 cm x 20 cm each, facilitating the feeding of the residue during the treatment. To these 20 units, 1 g of the stable metals Co, Sr, Cs and Ce were included to simulate the presence of radionuclides Co-60, Sr-90, Cs-137 and Ce-144.These elements were added from the inorganic compounds Co_3O_4, $Sr(NO3)_2$, $CsNO_3$ and CeO_2 in the form of solid powders. The amounts added are indicated in Table 2. The final residue (20 prepared units) had a mass of 231.5 g (200 g gloves + 25 g paper + 6.5 g simulants).

Table 2. Mass of the metal compounds added to the 225 g of simulated waste so that it contains 1 g of each metal.

Simulant compound	Species to simulate	Added mass per gram of metal [g]
Co_3O_4	Co-137	1.36
$Sr(NO_3)_2$	Sr-90	2.42
$CsNO_3$	Cs-137	1.47
CeO_2	Ce-144	1.23

3. Results

Table 3 shows the chemical elements detected in the initial residue together with the percentage by weight of each one.

Table 3. EDS results for simulated low-level radioactive waste materials.

Element	Nitrile gloves [%p/p]			Paper [%p/p]		
	Zone 1	Zone 2	Zone 3	Zone 1	Zone 2	Zone 3
C	72.8	71.8	71.1	50.1	50.4	50.4
O	13.2	13.0	13.3	49.8	49.5	49.5
Na	-	-	-	0.1	0.1	0.1
Al	2.7	-	0.1	-	-	-
Si	-	-	0.1	-	-	-
S	2.1	2.7	2.7	-	-	-
Cl	5.3	7.0	7.0	-	-	-
K	0.4	0.5	0.6	-	-	-
Ca	1.5	2.0	2.1		-	-
Ti	1.0	1.4	1.4	-	-	-
Zn	1.1	1.6	1.6	-	-	-

Table 4 shows the EDS results of the solid powders recovered after treatment (SE1: Sacrificial electrode zone 1; SE2: Sacrificial electrode zone 2; DRM: Deposits on the refractory material lid; SR: Secondary residue). Table 5 presents the results of the XRD analyzes for the same samples.

Table 4. EDS results of the solid powders recovered.

Element	SE1 [%p/p]	SE2 [%p/p]	DRM [%p/p]	SR [%p/p]
Co	0.8	2.1	3.8	2.6
Sr	0.4	0.8	2.2	1.4
Cs	1.6	3.5	-	-
Ce	0.6	0.7	1.6	0.3
Cl	1.6	1.6	-	0.1

Table 5. DRX results of the solid powders recovered.

Compound	Reference code	SE1	SE2	DRM	SR
CuCl	01-082-2114	OK	OK	OK	OK
Cu_2O	03-065-3288	OK	-	-	-
CuO	01-089-5897	OK	-	-	-
TiO_2	01-077-0440	OK	-	-	-
ZnO	01-074-0534	OK	-	-	-
TiO_2	01-075-1537	OK	-	-	-
TiO_2	01-071-0650	-	OK	-	-
$CaSO_4$	01-086-2270	-	OK	-	-
ZnO	01-079-0206	-	OK	-	-
CuO	01-080-0076	-	OK	-	-
ZnS	01-089-7386	-	OK	-	-
$TiZn_2O_4$	01-077-0014	-	-	OK	-
TiO_2	01-082-0514	-	-	OK	-

The final volume of the secondary residue (SR) was 34.4 cm^3 with a mass of 9.53 g, so there was a volume reduction percentage of 99.6% and a mass reduction percentage of 95.9%.

4. Conclusions and Acknowledgements

This study made it possible to reduce the original volume by 99.6%; identify the presence of the elements Co, Sr, Cs, Ce and Cl in the different zones of the system and recognize the formation of the crystalline phases CuCl, Cu_2O, ZnO, TiO_2, $CaSO_4$, CuO, ZnS, $CaSO_4$ and $TiZn_2O_4$ in the remaining powders. The significant volume reduction obtained during the experiment would contribute to the more sustainable use on nuclear energy when applied at an industrial scale.

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