PLASMA TREATMENT OF A SIMULATED LOW-LEVEL RADIOACTIVE WASTE

J.A. PULLAO
Comisión Nacional de Energía Atómica / Universidad Nacional de Cuyo / Universidad Nacional de Río Negro
San Carlos de Bariloche, Argentina
Email: juan.pullao@cab.cnea.gov.ar

F.E. BENEDETTO
Comisión Nacional de Energía Atómica / Universidad Nacional de Cuyo
San Carlos de Bariloche, Argentina

I.P. CORADINI
Universidad Nacional del Litoral
Santa Fe de la Vera Cruz, Argentina

L.P. NEIRA
Comisión Nacional de Energía Atómica
San Carlos de Bariloche, Argentina

Abstract

According to the Programa Nacional de Gestión de Residuos Radiactivos (PNGRR), 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 more 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%. This work presents an experiment on gasification by thermal plasma using a simulated low level radioactive waste (SLLW) to analyse its volume reduction and reaction products. The experiments was conducted at Departamento Materiales Nucleares o the Comisión Nacional de Energía Atómica. The SLLW had an initial volume of 9000 cm^3, consisting of nitrile gloves, laboratory paper, and chemical compounds of stable metals Co, Sr, Cs, and Ce to simulate the presence of radioisotopes Co-60, Sr-90, Cs-137, and Ce-144. The volume reduction obtained was 99.6% (34.4 cm^3), and the ashes inside the reactor contained Co, Sr, Cs, Ce, and Cl, along with crystalline phases CuCl, Cu_2O, ZnO, TiO_2, $CuSO_4$, CuO, ZnS, and $TiZn_2O_4$. This significant volume reduction obtained during the experiment would contribute to the more sustainable use on nuclear energy when applied at an industrial scale.

1. INTRODUCTION

Radioactive waste (RW) are materials that contain radionuclides with activity concentrations that exceed the limits established by the Nuclear Regulatory Authority for their dispersion in the environment and, therefore, require options for treatment, conditioning, storage and final disposal [1]. These residues are generated during the operation and maintenance of nuclear power plants, in research centers that work with radionuclides and in medical, agricultural and industrial applications of nuclear technology [2]. In Argentina, RWs are classified as low-level, intermediate-level, and high-level residues. The classification is made according to the half-life of the radionuclides and the concentration of alpha and beta-gamma emitters [3].

A low-level waste is one that has radionuclides with half-lives of less than 30 years, a concentration of beta-gamma emitters of less than 37 GBq/t, and a concentration of alpha emitters of less than 370 MBq/t [3]. As these residues are generated in large quantities, the development of effective, safe and reliable methods for their processing is required. Among the most used techniques are the thermal technologies of incineration, pyrolysis and plasma gasification. These technologies make use of the heat generated by various sources to decompose the waste and obtain final products of lower volume, non-flammable, chemically inert and much more homogeneous than the initial materials, facilitating the storage and final disposal of RW [4].

Thermal plasma is proposed as a desirable technology for the treatment of RW since it is a heat source that reaches temperature values between 5,000-50,000 K [5], allowing the treatment of organic and inorganic solids, liquids, and mixtures of the above, with the consequent reduction in the volume of the original waste close to 100% and the generation of a homogeneous secondary waste that is easy to handle at the outlet [6]. In turn, since this technology does not require fossil fuels, it reduces the generation of greenhouse gases, making it an environmentally friendly process. Although there are some plants in operation for the treatment of RW, such as SIA RADON in Russia, ZWILAG in Switzerland and KOZLODUY in Bulgaria [6], still this technique has a wide variety of possible approaches related to configuration and constructive form of the treatment chambers, the plasma generators and their operating parameters, and the methods and ways of interpreting the characterization of the process and by-products [7].

In the present work, a simulated low-level radioactive waste was prepared and characterized and treated in the thermal plasma gasification system of the Departamento Materiales Nucleares in order to study the volume reduction and the solids and gases generated during the process.

2. MATERIALS AND METHODS

2.1. Experimental set-up

Fig. 1 shows the diagram of the Department's thermal plasma gasification system, which includes the following components: gasification chamber (E-3); plasma torch Hypertherm model Powermax 105 (P-4); compressed air supply (E-1); copper sacrificial electrode (C-1); refrigeration system (buffer tank (E-14); cooling tower (E-15); heat exchanger (E-13) and two pumps (E-15) and (E-18)); gaseous effluent treatment system (washing tower (E-6) and particle filters (E-5 and E-8)); gas sampler and gas detectors.

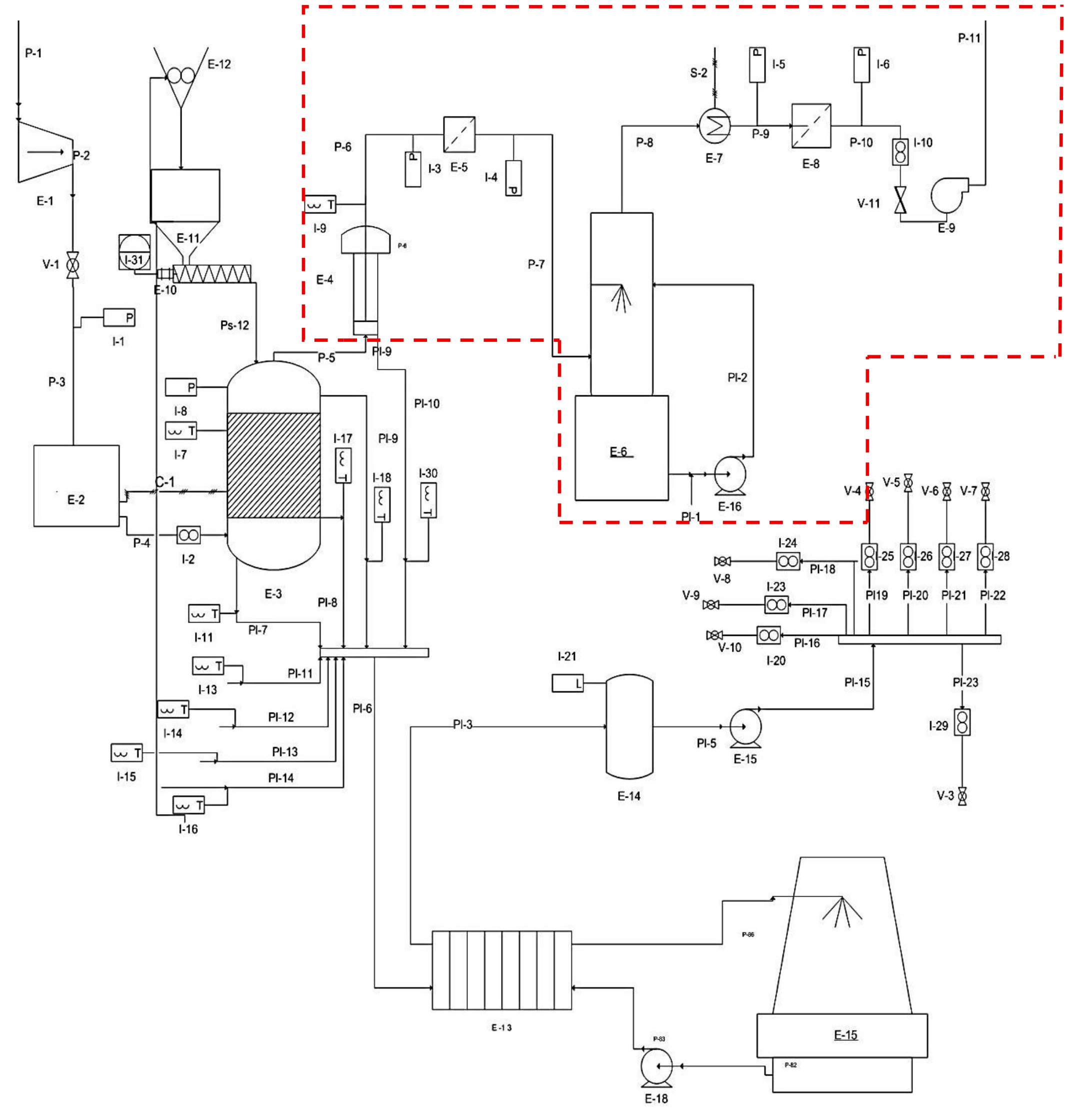

FIG. 1. Diagram of the plasma gasification process developed in the Department's laboratories. The red box --- represents the gaseous effluent treatment system. This stage is currently 90% installed.

The gasification chamber has a cylindrical geometry with a useful volume of 2 L. Its walls are built with refractory cement Surecast 60 brand S.A.E.M.S.A (60.0-62.0% Al_2O_3 and 31.0-35.0% SiO_2). Suitable to work with a maximum temperature of 1650 °C. In addition, on the side walls of the chamber there are two perforations through which the plasma torch and the sacrificial electrode enter for the operation with transferred electric arc discharge. The upper lid has an entrance through which the waste to be treated enters and a gas outlet. The entire chamber is placed inside a metal box and is surrounded by a layer of ThermalCeramics brand Kaowool 1260 insulating mineral blanket (53.0% SiO_2 and 47.0% Al_2O_3).

To treat the simulated residue, the system torch was used with a current of 30 A, a voltage of 160 VDC and in roughing mode. Compressed air was used to produce plasma gas at a flow rate of 9 kg/h at 7 bar pressure. The sacrificial electrode cooling flow was 10 L/min. The feeding was carried out by adding the packets progressively inside the gasification chamber (batch type feeding). The entire experience was carried out under a hood with gas extraction.

During the experiment, the gaseous effluents at the outlet of the gasification chamber were monitored with a Horiba model PG-350E portable detector and a Testo model 350 detector. Also, a control was made of the gaseous species present in the work area using an RKI Instruments Eagle brand 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, $Sr(NO_3)_2$, $CsNO_3$ 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. EXPERIMENTAL RESULTS

3.1. Characterization of simulated low-level radioactive waste.

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 [Wt%]			Paper [Wt%]		
	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	-	-	-

3.2. Characterization of the solid powders recovered after treatment.

Table 4 shows the EDS results of the solid powders recovered after treatment while Table 5 shows the results of the XRD analyses for the same samples.

TABLE 4. EDS results for simulated low-level radioactive waste materials (SE1: Sacrificial electrode zone 1; SE2: Sacrificial electrode zone 2; DRM: Deposits on the refractory material lid; SR: Secondary residue)

Element	SE1 [Wt%]	SE2 [Wt%]	DRM [Wt%]	SR [Wt%]
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

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; 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 and identify the presence of gases NO, NO_2, NO_X, SO_2, CO, CO_2, H_2, H_2S, HC and O_2 during the treatment. The significant volume reduction of low level radioactive wastes obtained in the conducted experiments using a thermal plasma gasification technique justify further investigations aiming to obtain related industrial processes for radioactive waste management contributing to the more sustainable use of nuclear energy,

5. FURTHER INFORMATION

5.1. Author affiliation

J.A. PULLAO, National Atomic Energy Commission, National University of Cuyo and National University of Río Negro, juan.pullao@cab.cnea.gov.ar
F.E. BENEDETTO, National Atomic Energy Commission and National University of Cuyo, benedetto@cab.cnea.gov.ar
I.P. CORADINI, National University of the Litoral, irinapaucarc@gmail.com
L.P. NEIRA, National Atomic Energy Commission, leonardo.neira@cab.cnea.gov.ar

ACKNOWLEDGEMENTS

The authors express their sincere thanks to Dr. Carlos Bertoli and Tec. Bernardo Pentke for carrying out the SEM-EDS analyzes of the recovered samples, Dr. Gastón Galo Fouga for performing the XRD analyses of the remaining solids, Lic. Gastón Goldmann for analyzing the simulated waste materials by neutron activation, Ing. Agustín Nasjleti for the FTIR analysis of the recovered gas.

REFERENCES

[1] Programa Nacional de Gestión de Residuos Radiactivos (PNGRR)., Joint Convention on the Safety of Spent Fuel Management and on the Safety of Radioactive Waste Management Seventh Report. National, Comisión Nacional de Energía Atómica, Argentinian republic, 2020.

[2] International Atomic Energy Agency (IAEA)., Final disposal of radioactive waste. Retrieved on February 9, 2023 from IAEA: https://www.iaea.org/es/temas/disposicion-final (2012).

[3] RIVERO, R, E., Treatment of radioactive waste through thermal processes. Nuclear Energy Technological Applications Area Management. Nuclear Materials Department. Division: Fuels and Waste., Comisión Nacional de Energía Atómica, Argentinian republic, 2017.

[4] Binetti Basterrechea, G. F., Pullao, J. A., & Benedetto, F. E. Reduction of the volume of radioactive waste simulated by means of thermal plasma, summer scholarships from the Instituto Balseiro, Comisión Nacional de Energía Atómica (2022).

[5] Hrabovsky, M., "Generation of Thermal Plasmas". Plasma Gasification and Pyrolysis. New York: CRC Press Taylor & FRancis Group (2023)

[6] Ojovan. M., Incineration and plasma processes and technology for treatment and conditioning of radioactive wastes. Handbook of advanced radioactive waste conditioning technologies. Cambridge: Woodhead Publishing Limited (2011).

[7] Prado, E., Miranda, F., Araujo, L., Petraconi, G., Baldan, M., Essiptchouk, A., & Potiens, A., Experimental study on treatment of simulated radioactive waste by thermal plasma: Temporal evaluation of stable Co and Cs. 160(108433) (2021).