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Experimental study on treatment of simulated radioactive waste by thermal plasma: Temporal evaluation of stable Co and Cs

E.S.P. Prado [a,b,c,*], F.S. Miranda [c], L.G. Araujo [b], G. Petraconi [c], M.R. Baldan [a], A. Essiptchouk [d], A.J. Potiens Jr. [b]

[a] National Institute of Space Research, Av. dos Astronautas, 1758 – Jardim da Granja, São José dos Campos, SP 12227-010, Brazil
[b] Nuclear and Energy Research Institute, Av. Prof. Lineu Prestes, 2242 – Cidade Universitária, São Paulo, SP 05508-900, Brazil
[c] Technological Institute of Aeronautics, Praça Mal, Eduardo Gomes, 50 – Vila das Acácias, São José dos Campos, SP 12228-900, Brazil
[d] São Paulo State University (UNESP), Av. Eng, Francisco José Longo, 777 – Jardim São Dimas, São José dos Campos, SP 12245-000 Brazil

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ABSTRACT

Thermal plasma technology is a process that demonstrates high performance for the processing of different types of waste. This technology can also be applied in the treatment of radioactive wastes, which requires special care. Beyond that, volumetric reduction, inertization, as well as a cheap and efficient process are necessary. In this context, the purpose of this paper is to demonstrate the application of thermal plasma technology for the treatment of solid radioactive waste. For this, stable Co and Cs were used to simulate compactable and non-compactable radioactive waste; about 0.8 g Co and 0.6 g Cs were added in each experimental test. The experimental tests were conducted using plasma of transferred arc electric discharge generated by the graphite electrode inside the process reactor. The behavior and distribution of the radionuclides present in the waste were assessed during the plasma process. The results show that the significant amounts of Co and Cs leave the melt by volatilization and are transferred to the gas phase with a small portion retained in the molten slag. The retention rate of Co in the slag phase is about 0.03% and 0.30% for compactable and non-compactable waste, respectively. On the other hand, Cs is completely transferred to the gas phase when added to the compactable waste. Conversely, when in the non-compactable waste, only 1.4% Cs is retained.

1. Introduction

Technological advancement in the nuclear area has been growing steadily due to the demands of regulatory bodies, usually based on the guidelines of the International Atomic Energy Agency (IAEA). The requirements are strict in terms of the quality of the process in the stage of treatment of radioactive waste; therefore, they require more effective, safe, and reliable methods (IAEA, 2011). For this reason, many nuclear facilities will be shut down and decommissioned shortly. However, a commercial nuclear power plant produces about 40,000 to 50,000 tons of waste during its decommissioning (Min et al., 2014). In this case, it is impossible to implement new technologies without fundamental studies of the radioactive waste treatment process.

In this sense, Thermal Plasma Technology (TPT) applied in the treatment of solid radioactive waste constantly needs new experimental studies due to the high complexity of the plasma system, involving the variation of the experimental configuration, waste composition, and characterization techniques to evaluate the efficiency of and improvements in the process.

Although it is necessary to emphasize that TPT can be applied in the treatment of several types of waste, it has been proven that this technology is especially useful for the treatment of radioactive wastes, due to the high reactivity promoted by the plasma, which results in suitable thermochemical reactions (Sikarwar et al., 2020).

However, thermal plasma processing of radioactive waste still has a wide variety of possible approaches related to the configuration and constructive form of reactors, plasma generators, and their operational parameters, in addition to the methods and forms of interpreting the characterization from the process and of by-products. Many processes are being developed around the world, based on the premise of high plasma reactivity, and they are based on selective oxidation, fluorination, chlorination, or evaporation to isolate radioactive fractions from the waste (Heberlein and Murphy, 2008).

* Corresponding author at: National Institute of Space Research, Av. dos Astronautas, 1758 – Jardim da Granja, São José dos Campos, SP 12227-010, Brazil.
E-mail address: edu.petraconi@gmail.com (E.S.P. Prado).

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Considering the high demand for the treatment of radioactive waste, studies on different configurations of the plasma system is required. Especially useful is the temporal assessment of the behavior of the elements present in the target radioactive waste during plasma treatment. This is important not only when considering the configuration of the plasma torch but the electrode of choice. Various studies of thermal and electrolytic effects on the behavior and distribution of Cs and Co have been undertaken by researchers and workers (Ghiloufi, 2009; Ghiloufi and Amouroux, 2010; Min et al., 2014; Nachtrodt et al., 2014; Nakashima et al., 2002; Yasui et al., 1997; Yasui and Amakawa, 2003).

For dimensioning and implementing of a thermal plasma plant for processing radioactive waste, all experimental conditions must be previously studied to obtain all possible scenarios of the process. There are currently some plasma plants in operation, which apply TPT as a method of treatment radioactive waste, such as SIA RADON in Russia, ZWILAG in Switzerland, KOZLODUY in Bulgaria, and some smaller installations around the world (Deckers, 2011; Prado et al., 2020a, 2020b). Given this, the motivation for the experimental study and application of thermal plasma as a method of treating compactable and non-compactable solid radioactive waste is the prior investigation of future implementations of small plasma systems in nuclear research institutes.

The use of radioactive isotopes is not common practice in research programs as it creates radioactive contamination and demands special permission for working with radionuclides (Yin et al., 2017). Due to these reasons, stable Co and Cs were used in the present work. The simulated radioactive compactable solid waste (CW) and non-compactable (NCW) solid waste are treated using transferred arc electric discharge plasma generated by the graphite electrode inside of a process reactor, and the mass and volume reduction were investigated. Beyond that, with the experimental tests, the Co and Cs temporal behavior during the thermal plasma treatment process of simulated radioactive solid waste was investigated in detail.

2. Material and methods

2.1. Materials

In this study, simulated non-radioactive solid waste (CW and NCW), provided by the Radioactive Waste Management Facility (RWMF) of the Nuclear and Energy Research Institute (IPEN), were selected to be treated in the thermal plasma reactor of the Laboratory of Plasma and Process (LPP) at the Technological Institute of Aeronautics (ITA). The waste underwent manual sorting first, separated by the materials listed in Table 1, then was homogenized in a five-liter container using a mechanical stirrer, separated into fractions of 250 cm^3, and weighed for processing.

Stable cobalt and cesium were included in all experimental tests as surrogates of ^{58}Co, ^{60}Co, ^{134}Cs and ^{137}Cs, in the compound forms of $CoCl_2{\cdot}6H_2O$ and CsCl (Batdorf et al., 1996). Amounts of about 0.8 g ^{59}Co and 0.6 g ^{133}Cs were added in each sample. These amounts of Co and Cs were selected after exploratory tests, with these amounts being considered sufficient for assessing the behavior of these elements during the thermal plasma treatment.

Table 1
Composition of simulated non-radioactive solid wastes for processing.

	Materials	Waste composition (g)	Ratio (%)
Compactable waste	Cotton, cloth, glove	50.0	62.5
	Plastic	17.5	21.8
	Paper, tapes	12.5	15.7
	Total	80	100
Non-compactable waste	Metal, screw	42.5	35.5
	Lumber	25.0	20.8
	Glassware	20.5	17
	PVC	32.0	26.7
	Total	120 100	

2.2. Thermal plasma system

Fig. 1 shows the schematic diagram of the thermal plasma system. The experiments were carried out according to the schematic drawing in Fig. 2 (Prado et al., 2020a, 2020b) with processing time up to 30 min, which includes the following components: graphite electrode as a discharge cathode, crucible of carbon-based composite material that acts as the anode (grounded) and contains the simulated waste volume, and an off-gas treatment system with sampling filter model FP2000 from HI-Q Whatman. The operating parameters of the discharge were fixed at a current and voltage of 140 A and 70 V, respectively. The compressed air flow rate in the process was 120 L h^{-1}.

2.2.1. Description of the thermal plasma reactor

The reactor has cylindrical geometry and an internal reaction chamber with a useful volume of 27 L, as shown in Fig. 2. The cover at the top of the reactor is mobile and has a central flange, where it is coupled with the graphite electrode, as well as a viewfinder with a borosilicate glass window. The reactor has a bottom electrode (anode) for operation with the discharge of transferred electric arc. The exhaust of process gases is done through the side flange. A slag leakage system at the bottom of the reactor allows the unloading of melting material if necessary. The reactor is made of stainless steel, has double walls cooled with water, in addition to an internal coating of refractory material, which constitutes the walls of the reaction chamber. Fig. 2 shows the temperature measurement points in the reactor (T1 - external wall of the reactor; T2 - refractory material; T3 – internal wall of the reactor; T4 – exhaust gas) to control the process. For experiments that require 30 min of processing, the maximum temperatures measured were T1 = 25 °C (room temperature), T2 = 620 °C, T3 = 810 °C, and T4 = 200 °C.

All connecting parts, which include the main body, the reactor cover, and the flanges, including the coupling of the graphite electrode, were sealed with a ceramic blanket for high temperature.

2.3. Characterization

To investigate the effects of thermal plasma treatment on the waste, we employed the Thermogravimetric Analysis (TGA) technique; the measurements were taken using a Netzsch STA 449 F3 Jupiter device. The studied samples were exposed to linear heating of 10 °C min^{-1} up to 750 °C under air atmosphere. In addition, for the investigation of the Co and Cs in the samples of slag and gas sampler filter collected, X-ray fluorescence (XRF) was the technique of choice for the bulk chemical composition as well as total Co and Cs contents. The samples were analyzed in a Bruker equipment; model Tiger S8, in the "Quant express–Best Analysis" method, in a helium gas atmosphere with reduced pressure. Loose powders were placed in a sample holder with a 6-μm thick mylar film, and the gas sampling filters were inserted directly into the sample holder.

3. Results and discussion

3.1. Thermal analysis results

Before plasma processing, a TGA was performed to understand the thermal decomposition of the components of the simulated waste without the presence of stable Co and Cs.

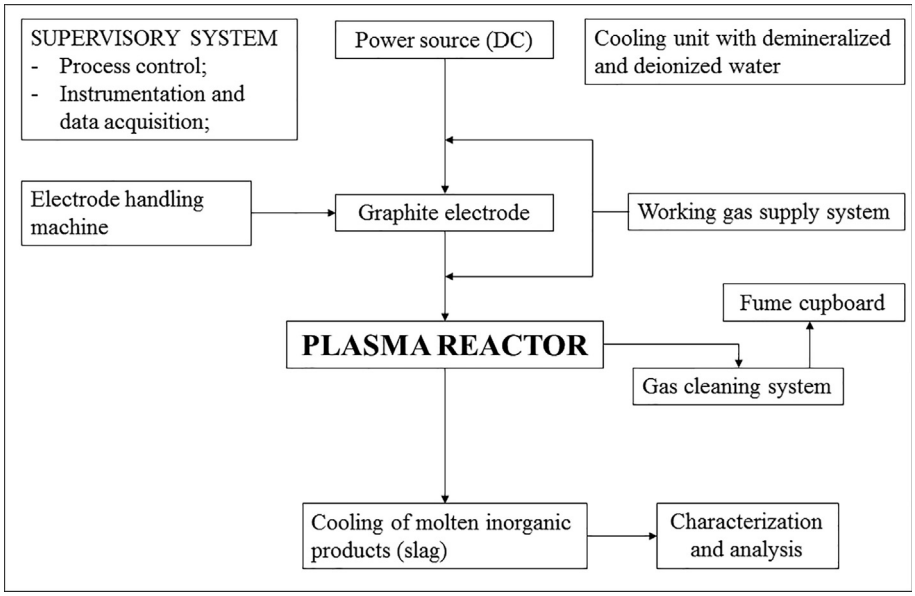

Fig. 1. A schematic diagram of the Thermal Plasma System.

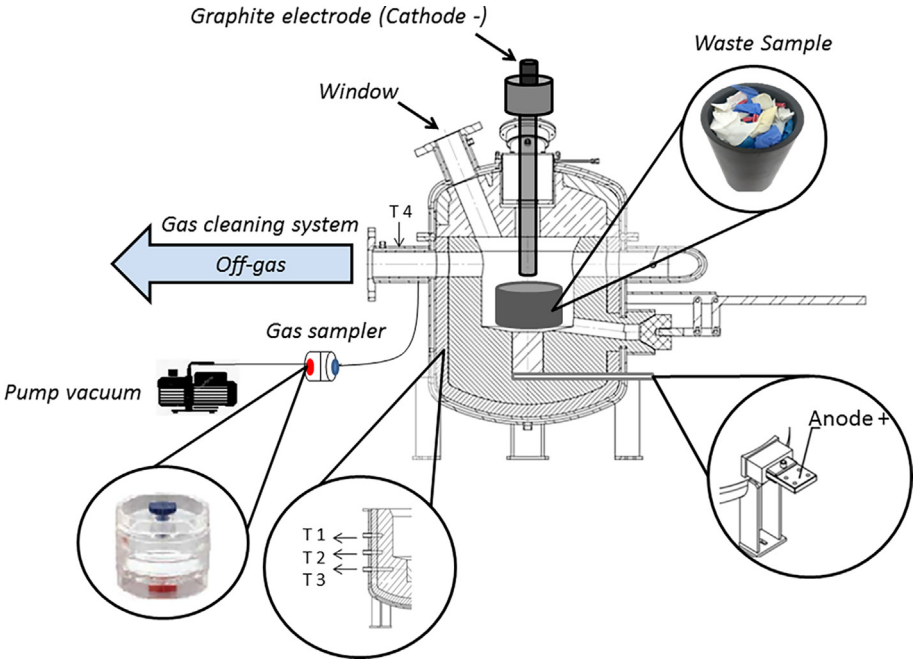

Fig. 2. Schematic drawing of the thermal plasma reactor (Prado et al., 2020a, 2020bs).

Fig. 3 presents the thermogravimetric (TG) data for simulated non-radioactive solid waste samples (CW and NCW) in oxidizing (synthetic air: 20% oxygen and 80% nitrogen) atmosphere. The TG curves of the waste show combustion profiles between 30 °C and 750 °C. The maximum mass loss rate during combustion took place between 250 °C and 500 °C, as a result of the decomposition of all organic matter, loss of volatiles, and waste gasification. The differences from the profiles are due to the variation in the composition of the wastes, being that the loss of mass for CW was approximately 78% and 30% for NCW, which can be explained as a difference in the concentration of organic materials, acting to increase the output of volatiles.

After this process, the final stage begins at 500 °C with 11% mass loss for CW and 8% for NCW. The final decomposition is of inorganic matter.

3.2. Thermal plasma treatment

3.2.1. Mass and volume reduction

Fig. 4 presents the results of mass and volume loss rate after the plasma treatment process. These results show that the mass and volume loss rates were higher for the CW compared to NCW for all processing times.

The mass and volume reduction ratios varied depending on the composition of the waste and the operation conditions of the thermal plasma. The ignition of the electric arc drives to a fast increase in temperature at the beginning (0–5 min), leading to a decrease in mass and volume of both wastes. The decrease of mass was 65% for CW and 43% for NCW at only 5 min. Longer times provided a higher reduction of mass and volume for CW and NCW, which were almost complete for CW, reaching values superior to 90% after

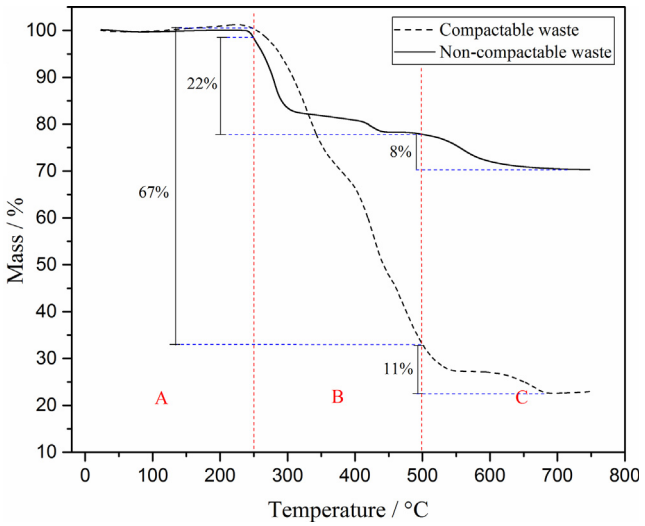

Fig. 3. TGA curves for CW (dash line) and NCW (solid line), heating rate 10 °C/min.

30 min of treatment. For NCW, the thermal plasma process was lesser efficient, reaching loss rates of about 60% mass and 70% volume.

The volumetric reduction rates (Fig. 4B) highlighted that the waste composition plays an important role under thermal treatment, considering similar conditions of operation. CW, which is composed of cotton, cloth, glove, plastic, paper, and tapes, had a greater volumetric reduction, about 99%, as previously reported in (Prado et al., 2020a, 2020b). Conversely, for NCW, which is composed of metal, screw, lumber, glassware, and polyvinyl chloride (PVC), after 30 min of the process, the volume reduction was significantly lower, approximately 76%.

Computer model studies confirm that due to the exposure time of the radioactive waste under plasma, there is a significant increase in the surface temperatures of the bath influencing the volatilization and loss of mass (Ghiloufi, 2009; Ghiloufi and Amouroux, 2010).

3.2.2. Concentrations of Co and Cs in the products after the plasma process

The behavior of Co and Cs concentrations in the slags after the thermal plasma processing is shown in Fig. 5. The Co concentration value after 30 min of processing was 5×10^{-3} mol·kg^{-1} for CW and 1×10^{-1} mol·kg^{-1} for NCW. The Cs concentration for CW was below the limit value of detection for X-ray fluorescence spectrometry, but for the NCW, it was 1×10^{-4} mol·kg^{-1}; therefore, according to the amount of Cs retained in the slag, it is possible to conclude that the surface temperature of the bath is above 2000 K (Ghiloufi and Amouroux, 2010). Ghiloufi (Ghiloufi, 2009) and Min (Min et al., 2014) investigated the behavior of the radionuclides Co and Cs in thermal plasma processing. Both studies showed that the addition of metals and semiconductors (mainly silicon) in oxidizing plasma environment catalyzes reactions with Co and Cs to form oxides of these elements in the condensed phase, retaining the Co and Cs in slag after vitrification. These results indicated that due to the composition of the CW, the loss of the Co and Cs in the slag for the exhaust line was more significant than for NCW.

Overall, the total Co and Cs concentrations for CW throughout the study in the thermal plasma process decreased over the processing time. However, the initial times for CW had a slight increase in Co concentration, going from 1.66×10^{-1} mol·kg^{-1} to 1.8×10^{-1} mol·kg^{-1}; for NCW, a small increase was maintained for Co concentration up to the final of process. As regards Cs concentration, there was a significant increase at 5 min from 3.5×10^{-2} mol·kg^{-1} to 6.5×10^{-2} mol·kg^{-1} and then, it declined up to 1×10^{-4} mol·kg^{-1}. This may be due to differences in waste compositions, especially the inorganic fraction, which is in greater quantity in NCW than in CW, resulting in greater retention of Co and Cs in the slag (Nakashima et al., 2002; Yasui and Amakawa, 2003). According to Ghiloufi (Ghiloufi, 2009; Ghiloufi and Amouroux, 2010), the increase in the processing time intensifies the vaporization speed and the volatilized quantities of the elements Co and Cs. In this study, due to batch processing, the effect of time is directly related to the increase in the process temperature. It should be noted that the thermal equilibrium of the reactor is associated with the energy supplied by the plasma generators; in this case, for a processing time of 30 min the process reactor is still in the heating phase (Mosse et al., 2008).

The previous analysis makes it possible to consider that the Co and Cs are present in the process gases. In such a case, for the exact determination of the total amount of Co and Cs concentration present in the gases, it is necessary to improve the system, primarily the specific equipment for measurement of the total gas flow in the exhaust line. However, it was assumed that the chemical composition of the gases is similar to the chemical elements present in the sampling filter after 30 min of processing, as shown in Table 2. On the other hand, it has not yet been possible to conclude on the

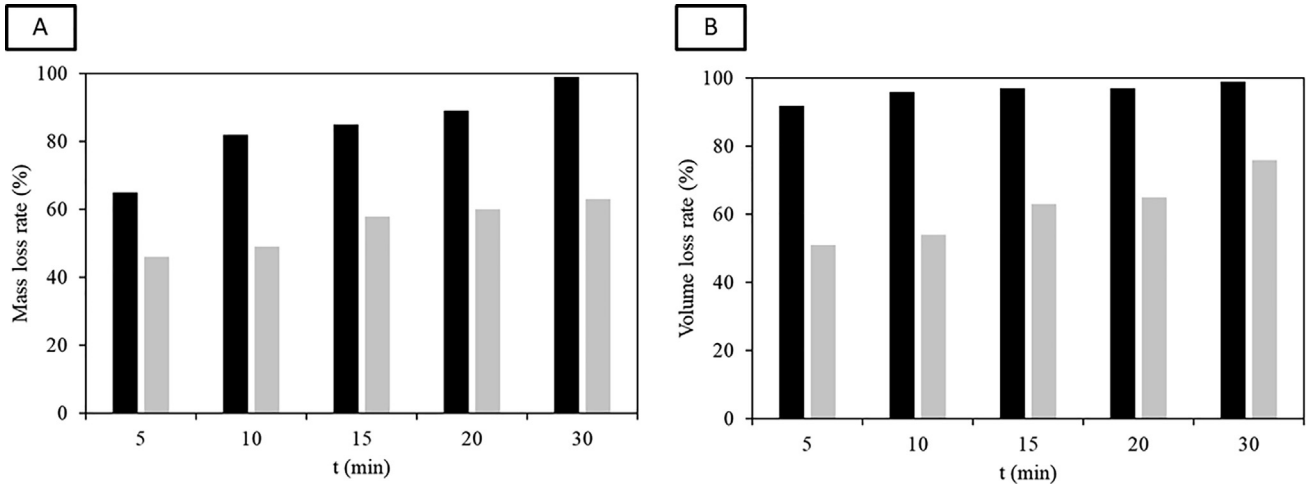

Fig. 4. Loss rate for (A) mass (%) and (B) volume (%). (In black) CW; (In grey) NCW.

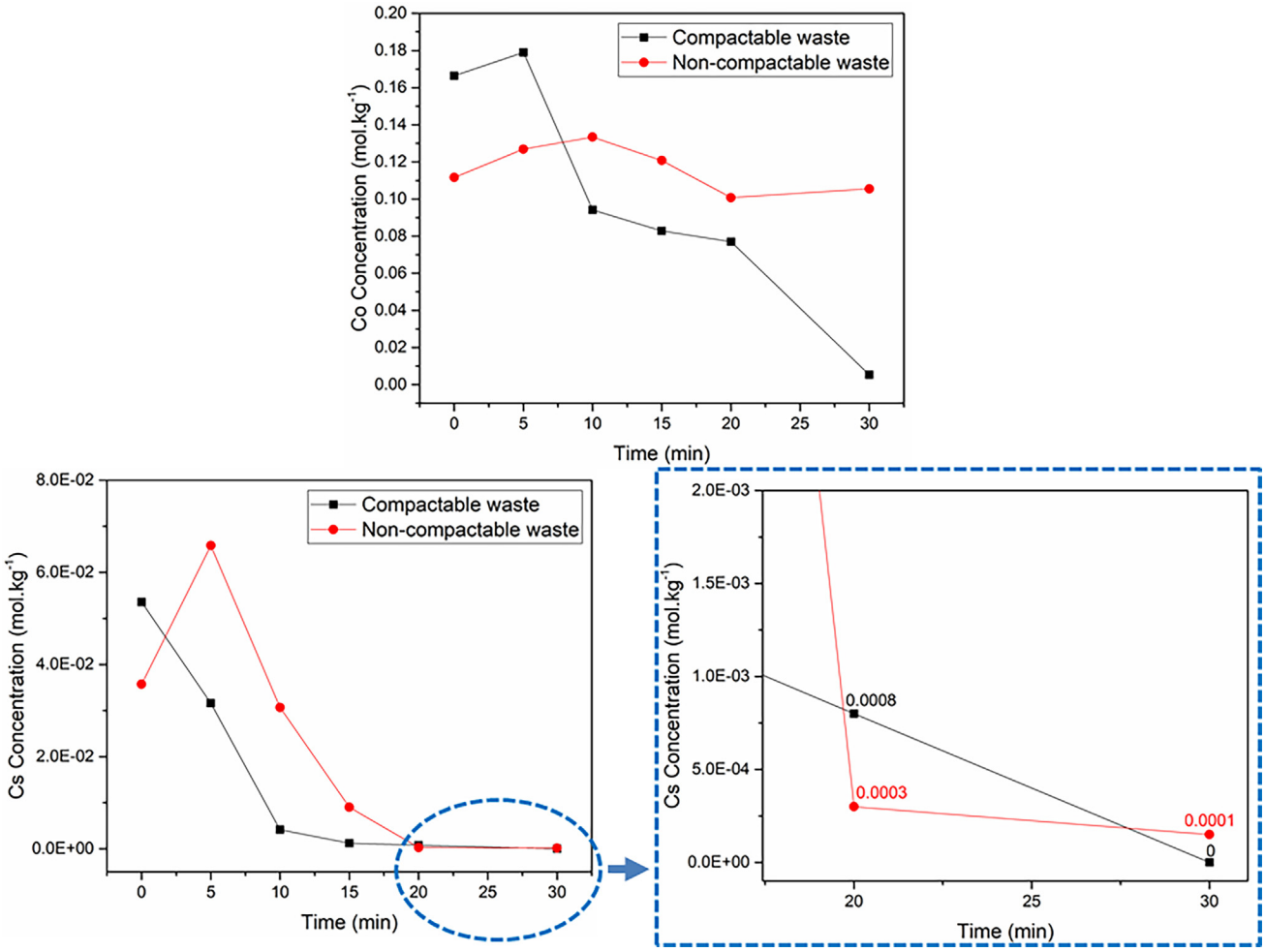

Fig. 5. Investigation of Co and Cs concentrations in the slags after the thermal plasma processing.

Table 2
Elementary composition in the gas sampling filter after 30 min of processing.

Waste	Elementary concentration (wt.%)																				
	C	Pb	Cl	Zn	Si	Fe	Cu	K	Ca	Ba	Cs*	P	Co*	Al	Mg	S	Cr	Sr	Mn	Ni	Rb
CW	48.28	15.9	12.4	8.14	3.14	2.22	2.17	1.07	0.989	0.789	0.611	0.381	0.157	0.121	–	0.091	0.0294	0.0218	0.0202	0.0174	0.00
NCW	66.43	1.43	9.39	2.81	6.9	2.29	1.1	1.34	2.48	3.12	0.518	0.54	0.0854	0.413	0.0831	0.441	0.0873	0.0475	0.0382	0.0766	–

* Stable elements.

aspects of mass balance due to the difficulty in assessing the total concentration of all elements in the off-gas.

Although the determination of chemical composition in the sampling filter does not present enough data for correlation of the values of the total concentration at the outlet gas, it is possible to claim that, in both cases, the presence of Co and Cs in filters is the reason for future studies of reprocessing filters with the addition of metal and semiconductors materials. The objective is to better retain the Co and Cs, and decrease the generation of secondary waste or in addition, apply the gas treatment methodology reported in the studies of (Deckers, 2011; Min et al., 2015, 2014; Prado et al., 2020a, 2020b; Tzeng et al., 1998).

4. Conclusions

In the present, experiments were performed using thermal plasma for the treatment of simulated radioactive waste: CW (higher organic concentration) and NCW (higher inorganic concentration), unlike what is commonly performed in thermal plasma plants, which treats mixtures of CW and NCW. The reason as to why the mixture of CW and NCW is treated is because the slag (from NCW) has a higher retention rate of radionuclides. Therefore, the use of stable Co and Cs in the processing of solid radioactive waste by thermal plasma was investigated. Mass and volume reduction were studied for CW and NCW with stable Co and Cs for better processing conditions.

During treatment, the interval that most influenced the mass loss rate was 250 °C–500 °C, as a result of the decomposition of organic matter, loss of volatiles, and waste gasification. Different mass loss were observed for the two materials investigated — 78% for CW and 30% for NCW — probably because of the higher organic content in the former.

In the thermogravimetric analysis, we can conclude that longer times of treatment provided higher degradation of mass and volume for both varieties of radioactive waste. Regarding mass loss, 30 min was necessary to degrade most of the CW, but for NCW,

no significant loss was observed after 15 min of treatment. The results obtained in the volumetric reduction highlighted that, similar to the mass loss rate, variety of solid radioactive waste to be treated is decisive when the thermal treatment technique is utilized under similar conditions of operation. In other words, composition plays a major role in this process.

The concentration of cobalt found after 30 min of processing was 5×10^{-3} mol•kg^{-1} for CW and 1×10^{-1} mol•kg^{-1} for NCW. After plasma treatment, the concentration of Cs for CW was below the limit of detection and was very low for NCW, with only 1×10^{-4} mol•kg^{-1}. Overall, the total Co and Cs concentrations for CW throughout the study in the thermal plasma process decreased over the processing time. Meanwhile, differences were observed, probably as a result of the variance in waste composition, especially the inorganic fraction, which increased the retention of Co and Cs in the slag.

However, it is worth mentioning that if an efficient gas treatment system for radionuclide retention is obtained, the process is highly efficient for optimizing the storage of radioactive waste, given that the repositories have their capacities compromised.

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CRediT authorship contribution statement

E.S.P. Prado: Conceptualization, Methodology, Validation, Formal analysis, Investigation, Data curation, Writing - original draft, Visualization. **F.S. Miranda:** Conceptualization, Methodology, Validation, Investigation, Formal analysis, Data curation, Writing - original draft. **L.G. Araujo:** Investigation, Data curation, Writing - original draft, Visualization. **G. Petraconi:** Conceptualization, Methodology, Supervision. **M.R. Baldan:** Resources, Resources, Supervision. **A. Essiptchouk:** Conceptualization, Methodology, Resources, Supervision, Funding acquisition. **A.J. Potiens:** Conceptualization, Methodology, Resources, Supervision, Funding acquisition.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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