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Use of plasma reactor to viabilise the volumetric reduction of radioactive wastes

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ABSTRACT

Nuclear reactors, hospitals, industries and research institutes generate considerable amounts of radioactive waste every day. To dispose this waste in a safe and cost-effective manner, it must be treated by immobilising the radionuclides and, for better stocking capacity, it must be volumetrically reduced as much as possible. To this end, plasma technology, among other promising technologies for radioactive waste treatment, exposes radioactive waste to temperatures above 1400 °C, thereby substantially reducing its volume. In the planning and managing of radioactive waste, the challenges related to plasma technology are presented as a motivation factor for the possible implantation of plasma reactors in nuclear plants and research centres, thereby improving radioactive waste management. In this study, a thermal plasma treatment process was established, and a plasma reactor was used for compactable waste processing. After 30 min of thermal plasma treatment, the volume reduction factor reached 1:99. The results demonstrate the viability of using a thermal plasma process for the volumetric reduction of radioactive waste in a safe and cost-effective manner.

1. Introduction

The technological and scientific progress in the area of nuclear power, observed since the beginning of the 20th century, has led to a wide variety of applications in nuclear fission research, medicine, industry and energy generation. Unfortunately, these practices have the disadvantage of generating radioactive waste that requires adequate management and treatment. The management and appropriate treatment of radioactive wastes, following specific procedures and regulations, is necessary to ensure the protection of human life and environment (Tzeng et al., 1998). According to the National Nuclear Energy Commission (CNEN) – the agency responsible for receiving, treating and storing radioactive waste generated in Brazil – 80% of this waste is compactable (i.e. laboratory-, safety- and hygiene-related materials such as gloves, special clothing, glasses, tapes, plastic tubes, and others). Due to its volume, maximum storage capacity in the repositories can be achieved (Prado et al., 2017). In the case of compactable wastes, the method of treatment includes volumetric reduction; this method is employed mainly for economic purposes. The reduction of volume ensures easy subsequent handling, transport, and storage of radioactive wastes. The current method used is mechanical compaction in 200-L drums, achieving a volume reduction factor (VRF) of 1:5 (IAEA, 2003). Presently, buildings designated to the storage and treatment of radioactive waste have their capabilities compromised around the world. As a consequence of the issue of storage, it is reasonable to say that is necessary to apply new methodologies to treat this radioactive waste in order to obtain a larger VRF (Prado et al., 2017). Therefore, the application of thermal plasma technology (TPT) emerged as a viable and promising alternative for volumetric reduction of wastes. The known methods for generation of thermal plasma are by means of direct current (DC) plasma torches which can be non-transferred arc and transferred arc (Gomez et al., 2009). Between the two methods, the transferred arc is suitable and more effective for waste treatment due to its high efficiency in the conversion of electric energy into thermal energy (around 95%) (Zhukov et al., 2007). Another alternative for generating a transferred arc electric discharge is through the use of graphite electrodes; because it has low complexity in construction when compared to plasma torches. In a reactor for the treatment of waste, the discharge is produced between the cathode (graphite electrode) and the anode (bottom of the reactor) (Mosse et al., 2008); the use of graphite electrodes also decreases operating and maintenance costs.

TPT is traditionally applied to process a large amount of waste. Thermal plasma has advantages when compared to other conventional thermal processes (e.g. incineration). The main distinguishing factors between them include the amount of added O_2 and the temperature inside the incineration furnace; in a plasma reactor, incinerators are designed to increase CO_2 and H_2O while thermal plasma treatment systems are designed to maximize CO and H_2 (Fabry et al., 2013). Inside the incineration furnace, there is an oxidizing environment (due to the

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excess of oxygen necessary for this process), causing the generation of NOx and SOx. On the other hand, in the thermal plasma process, there is a reducing environment that inhibits the generation of NOx and SOx (Byun et al., 2012). Another crucial difference between the incineration furnaces and the thermal plasma processes is the temperature. In the furnaces, the temperature reached is around 800 °C, which is below the melting point of ash; this causes inorganic materials contained in the wastes to convert to fly ash. On the other hand, the temperature of the thermal plasma process is over 1400 °C, which is above the melting point of ash (Byun et al., 2012). Additionally, TPT requires small, compact equipment and operational controls achieved through simple practices, enabling shorter startup and shutdown times (Li et al., 2016). However, TPT has certain technical disadvantages and limitations that need to be improved, such as the effective exhaust gas treatment system coupled with the containment of volatile radionuclides (Filius and Whitworth, 1996). For small-scale processing, the process becomes expensive for construction and operation (Deckers, 2011). In this context, taking into account the advantages and disadvantages of the application of thermal plasma technology, the first experimental thermal plasma plant for the treatment of low- and medium-level radioactive waste was constructed by SIA RADON in Moscow, Russia, called "Pluton," which operated with a waste processing rate of 40 kg/h between the years of 1998 and 2001 (IAEA, 2006). In early 2004, in the city of Würenlingen in Switzerland, the first large-scale industrial plant was developed and installed by ZWILAG and is still in operation at present. The maximum capacity of the facility is 200 kg/h of burnt waste and 300 kg/h of fusible waste (Heep, 2017). In 2013, JV IBERDROLA/BELGOPROCESS with MONTAIR PROCESS TECHNOLOGY began construction of another large-scale plasma installation for the treatment of low- and medium-level waste in the town of Kozloduy, Bulgaria. The plasma plant is currently in the testing stage. The facility consists of a plasma reactor equipped with a 500-kW non-transferable arc torch as a heat source with the capacity to process 250 tons per year over 40 operational weeks (Deckers, 2011).

Volumetric reduction tests using several methods have been performed previously (Garamszeghy, 2011; Mosse et al., 2008; Polkanov et al., 2011), and the results are still inefficient considering the volume reduction factors achieved; this demonstrates that a more effective method to optimize wastes storage in repositories should be investigated. Therefore, in this paper, the viability of the use of thermal plasma as a method of volumetric reduction using a process reactor will be presented. The results demonstrated that for compactable solid radioactive waste, a VRF of 1:99 can be achieved when processed in the plasma reactor using a graphite electrode.

2. Materials and methods

2.1. Sample preparation

In the experiments, samples of compactable non-radioactive solid wastes were used, similar to the wastes that are stored for treatment in the Nuclear and Energy Research Institute (IPEN). The use of simulated waste aims to establish an adequate and safe methodology for the future processing of radioactive wastes. The wastes were ground and homogenized by a mechanical agitator composed of a stainless-steel propeller and rod set and separated into fractions with a volume of 250 cm^3. Furthermore, all samples had their masses determined for processing. The volume of solid treatment product (slag) in powder form after processing was calculated with a graded beaker.

2.2. Experimental setup

The process reactor presented in Fig. 1 was used for the volumetric reduction of simulated waste in the plasma process. The process consisted of a cooling system that was used to maintain the low temperature of the reactor wall, power supply, and the electrode. A control system maintained the proper running of all the parts during operation. This system operated with an average power of 8.62 kW and used air such as working gas (flow of 120 L/h). The experiments were performed duplicated divided into time steps from 5 to 30 min.

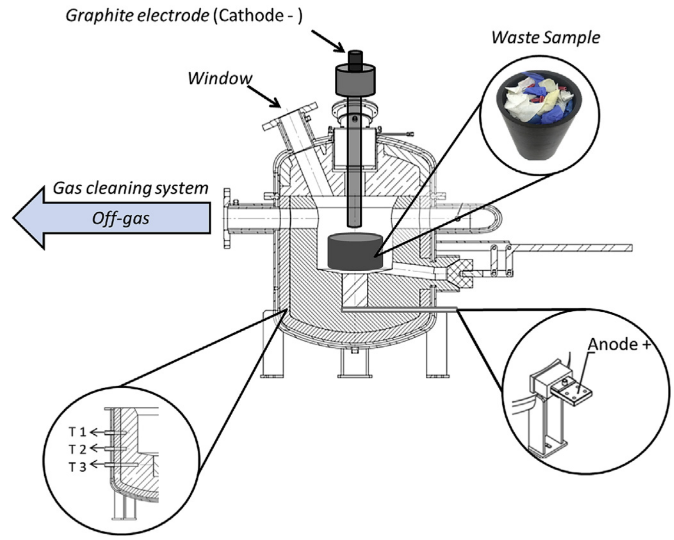

Fig. 1. Schematic drawing of the volumetric reduction reactor highlighting the main components of the experimental setup.

The temperature measurements were performed by thermocouples (T1, T2, T3 represented in Fig. 1). T1 refers to the reactor outer wall, T2 refers to the refractory material and T3 is relative to the closest temperature of the process chamber (waste sample, Fig. 1). For the experiments that require 30 min of processing, the maximum temperatures measured were T1 - 25 °C (room temperature), T2 – 620 °C and T3 – 810 °C.

The graphite electrode was fixed in the mechanic arm (elevator) as a discharge cathode; the arc closed directly over a crucible of carbon-based composite material that acted as the anode (grounded) and contained the simulated waste volume. Due to the characteristics of the waste, a crucible of carbon-based composite material was designed to couple the sample to the centre of the reactor process chamber, avoiding the molten bath at the bottom of the reactor that interfered with the collection of the slag. The mass, density and volume of the waste were measured before and after the plasma process with a density analysis by gas pycnometer, model Ultrapyc 1200e.

2.3. Off-gas treatment system

The reactor was connected to a gas scrubber where the process gases counter-flow to the scrubbing water sprayed by an internal shower at the top of the scrubber. The wash water flowed with the help of gravity through a filler layer made up of several tubular ceramic parts, while the gas flowed through forced convection promoted by a centrifugal fan installed in the exhaust of the gas washer. The several cavities formed in the filler layer increased the gas residence time and particulate retention efficiency. Finally, the gases passed through a HEPA filter to clear out unwanted particles and were then exhausted to the outside of the laboratory through a chimney.

3. Results and discussions

Fig. 2 illustrates the crucible with waste sample to be treated (Fig. 2 (A)) and the solid product after 30 min of the plasma processing (Fig. 2 (B)). Due to the high temperature that is attained in the process and the low volatilization conditions of the used material, it melts at a lower temperature when compared to the material of the crucible, facilitating the occurrence of the analysed effects, such as the vaporization of the

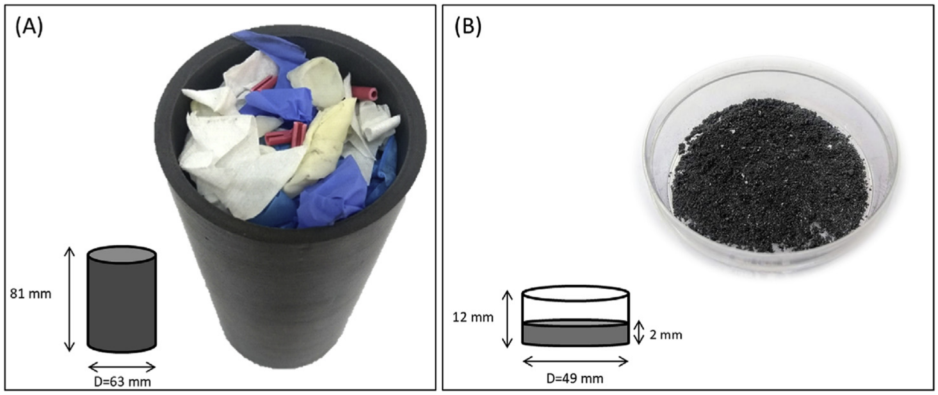

Fig. 2. (A) The crucible with the sample of compactable waste and the dimensions of the crucible. (B) Waste in a container after thermal plasma treatment and the dimensions of the container with final volume representation.

organic fraction of the waste. A small part of the crucible was also degraded due the oxidation process, occurred due the contact with oxygen during the experiments.

The processed wastes from each treatment were subsequently removed from the crucible after cooling and agglomerated in a Petri dish for mass and density determination. Higher percentage reduction was obtained in Experiments 9 and 10 with 30 min of processing as shown in Fig. 3.

The results obtained in the present work reached factors of volume reduction between 1:90 and 1:100. The thermal plasma technology get a difference considered when compared to conventional treatment of the compactable radioactive waste (in-drum compactation) that usually has a moderate volume reduction factor between 1:2 to 1:10 (Garamszeghy, 2011). The processing model of the (Mosse et al., 2008) mentions volume reduction factor the range of 1: 100 using two process chambers for burning of radioactive wastes, this process has more operational difficulties compared to the one developed in the present work, due to the use of two plasma torches.

4. Conclusion

The preliminary results indicate a high potential of the application of the compactable solid radioactive waste treatment with plasma technology in the process of volumetric reduction using graphite electrode to generate the plasma arc transferred. The initial experiments were performed using simulated radioactive waste exposed directly to the plasma arc in order to reduce the volume. For the application of radioactive waste treatment it is necessary to implement the gas cleaning system considering the presence of radionuclides. The acquired results during the 30 min of processing reached 1:99 of VRF over the sample before processing. It is concluded that TPT is a promising and effective method for the treatment of compactable solid radioactive waste.

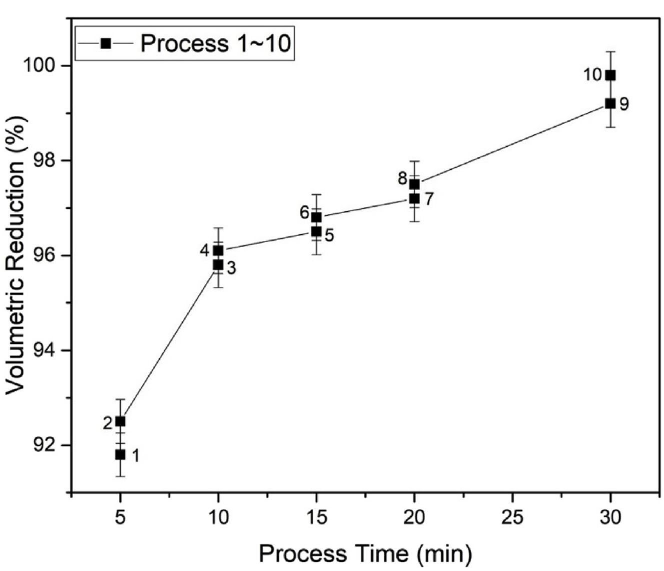

Fig. 3. Percentage of volumetric reduction of the experiments as a function of the processing time.

Authorship statement

All persons who meet authorship criteria are listed as authors, and all authors certify that they have participated sufficiently in the work to take public responsibility for the content, including participation in the concept, design, analysis, writing, or revision of the manuscript. Furthermore, each author certifies that this material or similar material has not been and will not be submitted to or published in any other publication before its appearance in the Radiation Physics and Chemistry.

Authorship contributions

All authors (Prado, E.S.P., Miranda, F.S., Petraconi, G. and Potiens Jr, A.J.) contributed to the development of the experiments, data acquisition, discussion of results, and manuscript writing.

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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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.radphyschem.2019.108625.

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