

### Thermal plasma technology for radioactive waste treatment: a review

Eduardo S. P. Prado<sup>1,2</sup> · Felipe S. Miranda<sup>2</sup> · Leandro G. de Araujo<sup>3</sup> · Gilberto Petraconi<sup>2</sup> · Mauricio R. Baldan<sup>1</sup>

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#### Abstract

In this paper, a review of radioactive wastes treatment using thermal plasma technology is presented as a treatment method for radioactive waste management.Virtually all waste streams can be treated by the thermal plasma technologies, resulting in a conditioned product, free from organics and liquids, definitely meeting the acceptance criteria for safe storage and disposal. The application of the thermal plasma system in the nuclear area is still one of the current research topics due to the theoretical and practical complexity of the treatment. This paper discusses the performance of the thermal plasma systems, addressing the advantages and limitations of the method.

Keywords Waste management · Waste treatment · Radioactive waste · Thermal plasma

#### Introduction

The scientific and technological progress in the area of nuclear power observed since the early twentieth century has led to a wide variety of applications in nuclear fission research, medicine, industry, and energy generation [1]. Unfortunately, these practices have the disadvantage of generating radioactive wastes (RW) that requires adequate management and treatment.

According to the RW management glossary published by the International Atomic Energy Agency (IAEA), it defines RW as "any material that contains or is contaminated with radionuclides at concentrations or activities greater than clearance levels as established by the regulatory body" [2].

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Eduardo S. P. Prado edu.petraconi@gmail.com

- <sup>1</sup> Instituto Nacional de Pesquisas Espaciais INPE, Rua dos Astronautas 1758, São José dos Campos, SP 12227-010, Brazil
- <sup>2</sup> Instituto Tecnológico de Aeronáutica ITA, Rua Marechal Eduardo Gomes 50, São José dos Campos, SP 12228-900, Brazil
- <sup>3</sup> Instituto de Pesquisas Energéticas e Nucleares IPEN, Av. Professor Lineu Prestes 2242, São Paulo, SP 05508-000, Brazil

There are interdependencies between all stages of RW management, from its generation to its final disposal [3]. For the treatment of RW, planning must be carried out in advance, so that a balanced method is adopted in the general management program between safety and operability requirements [4].

There are no universal and perfect technologies equally efficient to treat all types of RW; each treatment method has its restrictions [5]. There are increasing demands for further improvements in the efficiency and safety of RW treatment methods. These have stimulated efforts to develop new processes or improved conventional waste processing technologies.

Among the technologies used for waste treatment, the Thermal Plasma Technology (TPT) has attracted significant attention and development as they provide advantages regarding the stabilization of the waste form and high volume reduction [6, 7]. Previously, various review articles on studies of the plasma applications assigned for the processing of different wastes types such as municipal wastes (paper, biomass, plastic, cloth, etc.) and hazardous wastes (from industrial, agriculture, and hospitals) were reported [8–12]. However, to the best of our knowledge, the influence of the most important variables on the treatment of RW by thermal plasma has not been previously discussed, considering the previous studies performed and existing industrial-scale applications of this technology for the treatment of such waste. Also, must be pointed out the advantages and

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limitations of the process and highlight the perspectives of TPT in the nuclear area.

#### Thermal plasma technology (TPT)

#### **Basic description**

Plasma is a partially or fully ionized gas containing electrons, ions, and neutral particles (atoms, molecules, radicals) [13]. These characteristics allow a significant intensification of traditional chemical processes, essentially increasing the efficiency and stimulation of chemical reactions that are difficult to occur in conventional chemical processes [14, 15].

Although plasma is commonly produced by electrical discharges in gases, it can also be obtained in solids and liquids, requiring sufficient energy for their vaporization and ionization [13]. In solids and liquids, plasma can be obtained by means of high concentrations of energies using a laser [14]. In the case of plasma in a gaseous medium, it can be generated and sustained by electromagnetic energy that can be obtained from several sources, such as Direct Current (DC), Alternating Current (AC), Radio Frequency (RF) and microwaves [16].

#### **Plasma: types and classifications**

A typical classification of a variety of plasmas is given in Table 1 [16]. Plasmas can be classified mainly according to the ionization degree (proportion of neutral particles that are ionized into charged particles) and temperature [17]. High temperature plasmas (or hot plasmas) are characterized by presenting similar temperature for electrons and ions and a very high ionization degree ( $\approx$  1), whereas for low temperature plasmas (or cold plasmas), electrons can present higher temperatures than the heavy particles exhibiting, in this case, a partially ionized medium [18]. However, low temperature plasmas can still be divided into thermal and non-thermal plasmas. Where for thermal plasmas, the electrons temperature can be slightly higher or equal to the heavy particles temperature, non-thermal plasmas show a big discrepancy between electron and ion temperature. In addition, thermal plasmas exhibit much higher ionization degree than the non-thermal discharges [16, 18].

The subject of the present review paper is limited to thermal plasma and its application in RW treatment.

#### Description of plasma generating devices

Plasma torches are devices used to stabilize an electrical discharge between two electrodes with gas flow, aiming at the conversion of electrical energy into thermal energy [8]. The high enthalpy plasma results from the interaction of the gas with the electric arc, reaching high conversion efficiency which can be higher than 90% depending on the project of the plasma torch and the working gas [8, 19].

The operation regime of the plasma torch can be classified accordingly the source, which can be: Direct Current (DC), Alternating Current (AC) or Radiofrequency (RF), or by the type of discharge used, which can be non-transferred arc and transferred arc [20]. Between the two methods, the transferred arc is more effective for waste treatment due to its high efficiency in the conversion of electric energy into thermal energy (around 95%) [10, 21].

In a project of waste treatment by thermal plasma, one of the main parameters which need to take into to account is the average value of enthalpy [22, 23]. For estimate the values of average enthalpy, we can use [22]:

$$h_{\rm ave=} \frac{2\pi \int_{0}^{R} \rho v h r dr}{\dot{m}} \tag{1}$$

where  $\rho$ , v and h are, respectively, the density, velocity, and enthalpy of the plasma jet, as functions of the radial position r, R the channel radius and  $\dot{m}$  the total plasma gas mass flow rate.

The average enthalpy is easily determined from an energy balance of the plasma torch [22]:

$$\dot{m}h_{\rm ave} = IV - Q_{loss} \tag{2}$$

where I is the arc current, V the arc voltage and  $Q_{loss}$  the heat lost to the plasma torch cooling water.

High-temperature plasmas (fully ionized plasmas)		Low-temperature plasmas (partially ionized plasmas)		
		Thermal	Non-thermal	
Properties	$T_{\rm e} \approx T_{\rm i} \ge 10^7 {\rm K}$ $n_{\rm e} \ge 10^{20} {\rm m}^{-3}$	$\begin{split} T_{\rm e} &\approx T_{\rm i} \approx T_{\rm g} \\ T_{\rm g} &\leq 10^4  {\rm K} \\ n_{\rm e} &\geq 10^{20}  {\rm m}^{-3} \end{split}$	$T_{\rm e} >> T_{\rm i} \approx T_{\rm g}$ $T_{\rm g} = 300 - 10^3 {\rm K}$ $n_{\rm e} \approx 10^{14} {\rm m}^{-3}$	
Examples	Fusion plasmas, solar core	Atmospheric arc plasma, plasma torch	Corona discharge, dielectric barrier discharge, low pressure glow discharge	

 $T_{\rm e}$  electron temperature,  $T_{\rm i}$  ion temperature,  $T_{\rm g}$  gas temperature,  $n_{\rm e}$  electron density

 Table 1
 Plasmas classification

 [16]
 [16]

The average temperature is defined as the temperature corresponding to the value of the average enthalpy [22]:

$$h_{\rm ave} = h(T_{\rm ave}) \tag{3}$$

The average velocity is defined as [22]:

$$v_{\rm ave} = \frac{\dot{m}}{\rho(T_{\rm ave})A} \tag{4}$$

where, A is the cross-sectional area of the flow channel.

The difficulties of measurements of the real values of these parameters, due to the high complexity of the system making the average measurements attractive. Knowing that only current, voltage, work gas flow rate, and cooling water flow rate and temperature can be precisely measured. These parameters are used together with a table of the thermodynamic properties, and the density and enthalpy as a function of temperature can be estimated [23]. Accordingly to Heberlein et al. [22], the peak values of temperature are typically more than twice the average values, due to this reason, more detailed calculations or measurements are required giving temperature and velocity distributions in the reactor.

The majority of plasma arc generators used in treatment RW use DC rather than AC because there are less flicker generation and noise, a more stable operation, better control, a minimum of two electrodes, lower electrode consumption, slightly lower refractory wear and lower power consumption [8].

There are many companies developing technologies, based on transferred and non-transferred DC torches with water-cooled metal electrodes [10, 24–26], and some based on a transferred DC torch with two graphite electrodes not water-cooled [9, 10]. As reported by Polkanov et al. [6] there are some companies that sell plasma gasification plants based on their own technologies through subsidiaries, thus establishing the market and at the same time disseminating their technological advances. In parallel of these developments of industrial plasma gasification WTE plants, many researchers developed plasma torches [6], or studies using graphite electrodes as an alternative method for generating a transferred arc electric discharge; because it has low complexity in construction, operating and maintenance costs, when compared to plasma torches [27].

### Treatment stage in the RW management program

The management of RW must mainly aim at reducing costs and operating doses. It is also necessary to take into account the reduction of secondary RW during the management operation [28, 29]. There are

interdependencies between all stages of RW management, from the generation of the waste to its final disposal. For the treatment of RW, planning must be carried out in advance, so that a balanced method is adopted in the general management program between safety and operational requirements. The treatment of RW may include the following processes [4, 29]:

- Reduction in the volume of wastes (for example, incineration of combustible wastes, compaction of solid wastes and segmentation or disassembly of bulky wastes components or equipment);
- Radionuclide removal (for example, by evaporation or ion exchange for liquid waste streams and filtration of gaseous waste streams);
- Change in the shape or composition of the wastes (for example, through chemical processes, such as precipitation, flocculation, and acid digestion, as well as by chemical or thermal oxidation);
- Change in the shape or properties of the wastes (for example, solidification, sorption or encapsulation; common immobilization matrices include cement, bitumen, and glass).

There is no universal and perfect technology equally efficient to manage all RW streams, each method of treatment or conditioning of wastes has its own restrictions [30]. However, the types of RW to be treated using different heat treatment technologies are shown in Table 2, allowing comparison in relation to the applicability of plasma technology [6].

TPT has advantages when compared to other conventional thermal processes (e.g. incineration) [7]. The main distinguishing factors between them include the amount of added O<sub>2</sub> and the temperature inside the incineration furnace, which are designed to increase  $CO_2$  and  $H_2O_2$ , while thermal plasma treatment systems are designed to maximize CO and H<sub>2</sub> [10]. Inside the incineration furnace, there is an oxidizing environment (due to the excess of oxygen necessary for this process), causing the generation of  $NO_x$  and  $SO_x$  [10]. On the other hand, in the thermal plasma process, there is a reducing environment that inhibits the generation of  $NO_x$  and  $SO_x$  [31]. Another crucial difference between the incineration furnaces and the thermal plasma processes is the temperature [32]. 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 [10, 32]. On the other hand, the temperature of the thermal plasma process is over 1400 °C, which is above the melting point of ash [31]. Additionally, TPT requires small, compact equipment and operational controls achieved

Technology	Waste type							
	Organic liquids	Inorganic liquids	Organic solids	Inorganic solids	Mixed organic- inorganic solids	Mixed organic- inorganic liquids	Spend resins	
Calcination	NA	А	NA	NA	NA	NA	NA	
High-temperature incineration	А	А	А	NA*	A*	А	А	
Incineration	А	А	А	NA*	A*	А	А	
Melting	NA	NA	NA	А	NA	NA	NA	
Molten salt oxidation	А	NA	А	LA	LA	LA	А	
Plasma	Α	Α	Α	Α	Α	Α	Α	
Pyrolysis	А	NA	A**	A**	A**	А	А	
Synroc	NA	NA	А	А	А	NA	NA	
Thermo-chemical treatment	NA	NA	А	А	А	NA	А	
Vitrification	NA	А	A**	A**	A**	NA	А	
Wet combustion	А	NA	А	NA	NA	NA	A***	

 Table 2
 Applicability of thermal technologies to common waste types [7]

A technology is applicable to this type of waste, NA technology is not applicable to this type of waste, LA technology has limited applicability to this type of waste

\*Small pieces of inorganic are acceptable without causing damage or plugging of the system, \*\*applicable only for the granular or powder form of this type of waste, \*\*\*applicable only to organic spent resins

through simple practices, enabling shorter startup and shutdown times [27, 33].

can improve the treatment process even more in a matter of an environmentally friendly more process.

Other researchers reported similar advantages within technical factors, economics, and environment of the process [8, 22, 27].

#### **Treatment of RW by TPT**

#### Advantages of TPT in RW treatment

Numerous literatures have reported several advantages of TPT in RW treatment such as volumetric reduction method, vitrification method, gasification method, radionuclide retention modeling method and thermal processing system method [7, 22, 34-36]. Vanbrabant et al. [34] reported that TPT for RW provides significant advantages over conventional treatment methods considering that the storage cost per unit of packaged waste is high.

According to IAEA [7] reported that of TPT compared to conventional combustion is faster processing the which is a facilitator point for the RW management program. Ghilouf [36] measured that the thermal plasma system takes the advantages to incinerate the combustible parts of RW for volume reduction and to vitrify the noncombustible counterparts simultaneously into glassy slags with very low leaching rate.

Deckers [35] performed studies thermal plasma processing of RW for conditioning of RW and related several advantages commons and specific in relation to incineration, the main advantage is worth mentioning that the combination of a plasma torch and a conventional burner Limitations of TPT in RW treatment

The disadvantages associated with the plasma process, besides the lack of confidence on the part of the competent authorities, although proof of concept and practice are already a reality, lie in the use of electricity, the most expensive form of energy [37]. Consequently, economic considerations provide the strongest barrier for use of plasmas for RW treatment [8, 22]. It is noteworthy that the costs for installation and operation of a plasma treatment facility for RW depend on various factors, like any new technology application for the treatment of RW's, the cost must be taken into account. Nonetheless, electricity costs can be compensated due to the capacity of plasma to process any type of residues, which for the most part cannot be treated with conventional methods [38]. This fact leads to increased storage costs, in addition to the environmental impact. For this reason, the effective cost of operating the plasma treatment can be offset by the added value of the waste when stored or treated by conventional methods [22, 38].

It is important to note that a thermal plasma plant does not require pre-treatment of waste, decreasing related costs [39]. In addition, it does not need screening or other methods for secondary treatment, directly going to the final disposal [40]. In scenarios where the whole waste stream is produced or stored at the plasma plant site, transportation costs are eliminated as well [40, 41].

However, the thermal plasma treatment has certain technical disadvantages and limitations that need to weaken or solved, such as [35, 42, 43]:

- An effective exhaust gas treatment system coupled with the containment of volatile radionuclides;
- For small-scale processing, the process becomes expensive for construction and operation.

## Other factors influencing the operation of thermal plasma system in the RW treatment

Plasma treatment is a thermal process in which waste is exposed to extreme thermal conditions (approximately 2000–14,000 °C) of temperature [44]. Several subsystems and factors are involved in the plasma treatment of RW providing different necessary functions, as follows:

#### Gas handling system

- *Precursor gas supply* The source materials or precursors are in most cases gases in high-pressure cylinders or liquids with sufficiently high vapor pressures.
- *Mass flow controllers* These are used to measure and control the flow of the different gases fed to the reactor.
- *Exhaust system comprising motors of the fans and pressure controller* The plasma reactors for RW processing operate at negative pressures. However, lower background pressure is often required to ensure the cleanliness of the process. The entire gas stream is propelled through the off-gas treatment system by redundant flue gas extraction fans. They have a dual function of enabling transportation of the flue gases and ensuring that the required continuous negative pressure inside the entire system is maintained.

#### **Plasma reactor**

In general, the study carried out by Li et al. [33] reported that plasma torch can be connected to the reactor through two modes in which the most conventional is a single-stage reactor that couples the plasma torch to the reactor in just one body where the plasma jets are located at the bottom or the top of the reactor, and waste is heated directly by plasma jets. Another mode is a two-stage reactor, in which a conventional reactor is followed by a plasma converter, wherein previously the crude syngas and the solid residue are reformed, and then processed in the plasma reactor [10, 45]. Fourcault et al. [46] compiled that in the second case, the plasma heat is used to provide the heat to 'polish' the crude syngas, and/or to vitrify the solid residues derived from the conventional thermal process.

Agon et al. [47] deduced that a single-stage plasma system shows more favorable characteristics for syngas production, while the two-stage plasma system favor the process of vitrification of solid waste. For each facility, the plasma reactors will be described in detail in later sections of this review paper.

#### **Power supply**

There are different types of power supply, which the most common are: Direct Current (DC), Alternating Current (AC) and Radio Frequency (RF) [12]. The role of the power supply is very important for the project of the treatment system since from the power supply will be determined the processing capacity and the adequate project of the plasma torch (or plasma generator) [48]. Beyond that, the power supply has to sustain the plasma in the reactor and provide an operational control system for the process [49].

#### Safety devices

The thermal plasma process is similar to any other hightemperature process in which high-quality slags are produced, but in such case, some particulates and volatile and/or semi-volatile elements can escape of the slag [50]. Continuous inspection of off-gas is usually performed to ensure that the gas does not include undesirable matters. Activity detectors, emissions detections, and other devices are routinely utilized to detect these undesirable components [35].

#### Feeding systems

The choice of feed systems has a direct effect on processing parameters; basically, there are two main types of feed systems, namely batch and continuous [35]. In a batch feed system, the drum (generally 200-L) containing the RW (with metals, concrete debris, and organic material) is placed whole inside of the process chamber of the plasma reactor [35]. The feed process generally is made by a robotic arm, in order to avoid contact with the operator [50]. In case the waste contains a considerable amount of organic material, the off-gas system should be sized for the large instantaneous flue gas flow caused by its vaporization [35].

In the continuous method, a shredder is used to provide a continuous/uniform feeding, smoothes and reduces peak off-gas flow rates [51]. Shredders allow a 200-L drum to be fed into a primary chamber, usually insufficient in terms of size to handle the entire drum, without treatment [35].

#### **End product**

The main advantage of TPT in the treatment process of RW is the characteristics of the end product [51]. Once that the process changes the characteristics of the waste and results in an end product adequated to safe disposal and storage. Knowing that the end product has the homogeneous form and consists mainly of amorphous material with a portion around of 10-20% crystalline phase, have very high mechanical and chemical durability exceeding the equivalent properties of the glass matrices [36, 50].

Deckers [35] emphasized that processing of RW containing both organic and inorganic materials may result in a heterogeneous slag in which the molten debris, metal integrate different forms in the final product. The author further emphasized that technically this has no direct impact on the acceptability of the final product form for disposal but may have a specific activity higher than the original as-generated waste, depending on the waste composition and volume reduction factor. This may impact the waste classification and acceptance criteria for the disposed of the package [7]. A study of thermal decomposition of the waste constituents is necessary to understand the series of phenomena occurring in slag during the plasma treatment [50]. Though literature pertaining to the treatment of RW's is available such as [3, 6, 52], there are no many studies investigating the effect of different atmospheres/ambients and the physicochemical reactions taking place in the slag are available. For instance, the determination of reaction kinetics during the plasma process similar to the study by Suneel et al. [53] in the vitrification process.

## Description of facilities for the treatment of RW by thermal plasma: global scenario

There are limited numbers of plants in operation, which apply thermal plasma technology as an RW treatment method. Only in 2004, the first plasma plant became operational, since that, other facilities are under construction or in development and some even in advanced stages of investigation/trials [7]. Therefore, the main plants in partial or complete operation can be listed below:

#### Russia

The first experimental thermal plasma plant for the treatment of RW of low and medium levels of radiation was commissioned by SIA RADON, in Moscow, Russia called

"Pluton" (operating period: 1998–2001). SIA RADON then built an industrial-scale plant for the treatment of mixed RW in a plasma shaft furnace with melted slag pour, which has a capacity of up to 200–250 kg  $h^{-1}$  [7].

#### Description of the Pluton plasma plant

As described by Dmitriev et al. [54] the process chamber of Pluton was made of refractory-coated steel (as used in processing ovens). With a height of 6.4 m and a diameter of 0.8 m it had the capacity to be filled with 3.5 m3 of waste. In the process chamber two plasma torches were used, which operated at a maximum power of 150 kW. Such power made the system reach high temperatures around 1500 to 1800 ° C during the process, so the treated material was collected from the bottom of the process chamber [6, 7]. The plant shown in Fig. 1 allowed drying, pyrolysis, gasification, oxidation and pouring of the materials/waste to be treated.

The feeding is made through falling by gravity into the first chamber, called of loading unit; this chamber is sealed in order to prevent air from entering into the process chamber. With the absence of free oxygen, waste passes through stages of drying and pyrolysis, led by intensive gas formation [7]. The resulting gases are processed by an afterburner, where the components are burned at a temperature range from 1200 to 1300 °C. Subsequently, off-gases are cooled to 300 °C in a contact heat exchanger, and remaining aerosol particles are trapped in a sleeve filter [54]. The gaseous components (HCl, NO<sub>x</sub>, SO<sub>2</sub>, etc.) are neutralized in the absorber and washed with an alkaline solution. The off-gases are then cooled, filtered and released into the atmosphere [6, 54].

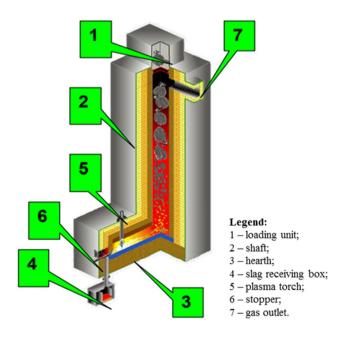


Fig. 1 The general view of the shaft furnace [7, 54]

#### Switzerland

In early 2004, in the city of Würenlingen in Switzerland, the first large-scale industrial plant was developed and installed by ZWILAG (Fig. 2) [7], and it is still in operation. The maximum capacity of the facility is 200 kg  $h^{-1}$  for burnable waste and 300 kg  $h^{-1}$  for fusible waste. This plant operates twice a year and each process remains for about 10 weeks, the restriction or limited time of use is not due to technological limitation but for logistical and organizational purposes [7, 35].

#### Description of the ZWILAG plasma plant

The actual plasma reactor consists of a cylinder with approximately three meters in diameter and four meters in height. The reactor, including the interior refractory lining, weighs a total of about 28 tonnes [55]. It comprises two, twin-walled, water-cooled steel parts: the cover and a lower section. The cover contains all the necessary openings for feeding [55]. The feeding is made by drums and the residues are processed by a thermal plasma torch. As described by Heep [55], the processing of waste by batches takes place by charging the plasma furnace with 200-L drums of untreated waste, which are introduced automated and controlled remotely to the process chamber where is located the plasma stream. From the horizontal drum feeder, the waste falls onto the molten slag. Inorganic material is melted and becomes slag. Organic material is vaporized, and the remaining volatile gases are fed to the afterburner chamber [7, 55]. A rotating crucible (centrifuge) in the primary processing chamber moves the molten slag, which directed the slag from the pour hole during processing [55]. The slag moves towards the center and pours through the throat into a mold located directly below the throat in the slag collection chamber, pouring is achieved by opening the outlet of the throat and slowing down the centrifuge [56].

After complete oxidation in a secondary combustion chamber, the flue gases emitted from the centrifuge chamber are routed to an off-gas treatment system consisting of a wet physical process and a wet chemical process [7, 55, 57]. Part of the thermal energy from the flue gases, still laden with contaminants at this stage, is drawn off in the recovery boiler and used for reheating, the remaining gases pass by a

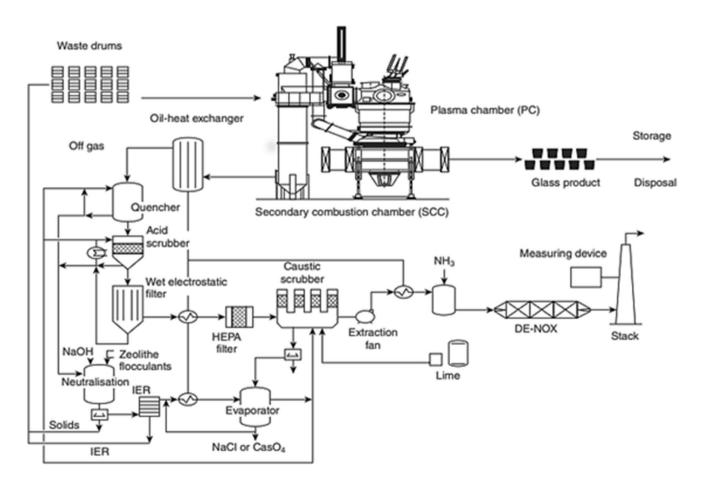


Fig. 2 Flow diagram of ZWILAG plasma facility [7]

series of filters to retain all possible contaminants and then can be released to the atmosphere [35, 55].

#### Bulgaria

In 2015, Bulgaria started the construction of the large-scale plasma plant, in the city of Kozloduy, for the treatment of low and medium radiation waste from the Kozloduy Nuclear Power Plant. The plasma facility was taken in nuclear operation in 2018 [27, 55].

#### Description of the KOZLODUY plasma plant

The facility consists of a tilting plasma furnace equipped with a nontransferable torch of 500 kW as a heat source and will treat 250 tons per year, spread over 40 operational weeks. The tilting furnace developed was designed to pour the slag in a controlled way into a slag mold [57, 58].

The RW is shredded to feed the furnace; the feeder has a rotating connection so that the feeder is mounted to the tilting furnace. The contaminated process gases, with temperature about 1300 °C, are directed to the treatment chamber. The system processes mixtures of organic waste with inorganic waste, and depending on the incoming waste composition, a glassy-like slag or a metal-like slag is obtained. When the process reaches 200 L of slag, the slag is poured into the mold, about 50 L of slag remains in the furnace, and it is used as thermal protection for the refractory [55, 57]. An overview of the plasma facility is depicted in Fig. 3, the components of the plasma facility were thoroughly described by [57, 58]:

#### Main TPT facilities around the world

The development of thermal plasma processing for waste disposal began in the USA, Europe, and Japan in the 1980s [8]. Now, more than 150 industrial plasma plants are used mainly in the processing of municipal waste [59]. Table 3 shows some plasma plants for the processing of different wastes in several locations from all over the world.

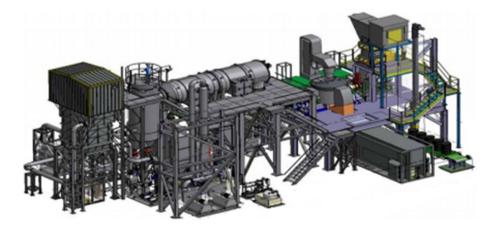
# Recent studies related to the RW processing system by thermal plasma in the nuclear area

The importance of the systematic study of chemical and physical properties of solidified products and the distribution of trace elements serve as a tool to optimize the process of treatment of RW by thermal plasma. The major problem of RW vitrification by thermal plasma is the volatility of radionuclides which end up leaving the reactor and reaching the gas cleaning system [61, 62].

There are few papers on volatility of radionuclides during thermal plasma processing. Several types of radionuclides have been used to study the products of the process, especially cesium and cobalt, including europium and ruthenium [36, 62–64].

Ghiloufi [36] studied the distribution of radionuclides <sup>137</sup>Cs, <sup>60</sup>Co and <sup>106</sup>Ru and other species possibly present in radioactive waste by a computer model. Some of the results were compared with those obtained previously by the same author Ghiloufi and Amouroux [65]. In [36], the author modeled the behavior of these three elements in the vitrification of waste, considering a two-phase system (liquid and gas). The model derived from a dynamic equilibrium with reaction sequences and the mass transfer equation, electrolysis effects and diffusive transport in a closed system. According to Ghiloufi [36], the model can handle 20 elements, 99

**Fig. 3** Overview of the plasma facility KOZLODUY [57]



**Table 3** The main plasma plants for processing of different waste currently in operation in the world and plant projects for the coming years [10, 33, 60]

Location	Type waste	Capacity	Date
North America			
Anniston, USA	Catalytic converters aluminum	24 t/day	1985
Libby, USA	MSW	45 t/day	1987
Jonquiere, Canada	Aluminum slag	50 t/day	1991
Honolulu, USA	HSW	1 t/day	2001
Bristol, USA	MSW	4.5 t/day	2001
Montreal, Canada	MSW	2.5 t/day	2001
Richland, USA	HW	4 t/day	2002
Alpaca, USA	MW	10 t/day	2003
US Navy	Wastes on board	7 t/day	2004
Monterrey, Mexico	MSW	90 t/day	2005
Hawthorne, USA	MW	10 t/day	2006
Madison, USA	CW	18 t/day	2009
Los Angeles, USA	Biomass	18 t/day	2009
Hurlburt Field, USA	HSW/ISW/HW	10.5 t/day	2011
Quebec, Canada	ILW	1.2 t/day	2013
Ottawa, Canada	MSW	85 t/day	Project
Port Hope, Canada	MSW	400 t/day	Project
Tallahassee, USA	MSW	910 t/day	Project
Europe		-	0
Landskrona, Sweden	Fly ash	200 t/day	1983
Bordeaux, France	MSW ash	10 t/day	1998
Moscow, Russia	LLRW	1 t/day	1998
Morcenx, France	Asbestos	22 t/day	2001
Kędzierzyn, Poland	ISW	10 t/day	2001
Bergen, Norway	Tannery waste	15 t/day	2001
Würenlingen, Switzerland	LLRW	7 t/day	2004
Swindon, England	MSW	0.25 t/day	2008
Morcenx, France	ISW/Biomass	137 t/day	2012
Hirwaun, Wales	MSW/ISW	750 t/day	2015
Kozloduy, Bulgaria	LLRW	1.5 t/day	2018
Sunderland, England	ISW/Biomass	107 t/day	Project
Hull, England	ISW/Biomass	107 t/day	Project
Barrow, England	ISW/Biomass	107 t/day	Project
Barry, Wales	ISW/Biomass	107 t/day	Project
Belgium	MSW/HSW/HW	246 t/day	Project
Swindon, England	MSW	-	Project
Asia			0
Kinura, Japan	MSW ash	50 t/day	1995
Yongin, Korea	MSW ash	14 t/day	1997
Yoshi, Japan	MSW	151 t/day	1999
Mihama-Mikata, Japan	MSW/sludge	28 t/day	2002
Utashinai, Japan	MSW/scraps	300 t/day	2002
	r	•	
	MSW ash	41 t/dav	2002
Shimonoseki, Japan Imizu, Japan	MSW ash MSW ash	41 t/day 12 t/day	2002 2002

339

Location	Type waste	Capacity	Date
Maizuru, Japan	MSW ash	6 t/day	2003
Lizuka, Japan	ISW	10 t/day	2004
Tainan, China	ISW/HW	5 t/day	2005
Taipei, China	HSW/HW	4 t/ day	2005
Zigong, China	HW	3 t/ day	2006
Osaka, Japan	PCBs	4 t/day	2006
Hiemji, Japan	MSW ash /PCBs	5 t/day	2006
Kaohsiung, China	ISW/HW	0.5 t/day	2007
Cheongsong, Korea	MSW	10 t/day	2008
Liquan, China	SSW/HSW	5 t/day	2008
Pune, India	HW	68 t/day	2009
Nagpur, India	HW	68 t/day	2010
Taichung, China	ISW/HW	1.5 t/day	2011
Shanghai, China	HSW	1.5 t/day	2013
Shanghai, China	HSW	30 t/day	2014
Dongguan, China	MSW	30 t/day	2016
Shenzhen, China	MSW/HW	25 t/day	2019
Bijie, China	MSW	600 t/day	Project
Beijing,China	MSW	200 t/day	Project

Table 3 (continued)

*MSW* municipal solid waste, *HW* hazardous waste, *ISW* industrial solid waste, *HSW* hospital solid waste, *SSW* sewage sludge waste, *CW* construction waste, *LLRW* low-level radioactive waste, *PCB* poly chlorinated biphenyl

species and 10 different phases for a given temperature and total pressure.

The volatility of the three radionuclides was evaluated by the proposed model under different process conditions. These conditions were temperature, furnace atmosphere, current, and matrix composition [36].

Regarding temperature, Ghiloufi [36] emphasizes two different behaviors in relation to the volatilities of <sup>60</sup>Co and <sup>106</sup>Ru. The former is not volatile at temperatures below 2000 K, while above 2000 K, cobalt vaporization occurs with different vaporization rates. The latter has contrasting volatility behaviors, depending on the temperature or temperature range. Generally, <sup>106</sup>Ru's volatility increases as the temperature increases. However, in the range of 1700–2000 K, volatility decreases as the temperature increases. In a previous study, Ghiloufi and Amouroux [65] studied the volatility of <sup>137</sup>Cs at different temperatures and found that its behavior is similar to that of <sup>60</sup>Co.

For the furnace atmosphere, Ghiloufi [36] used the argon/oxygen mixture as the carrier gas,and temperature, total pressure and plasma current were kept constant. The volatility of compounds  $^{60}$ Co and  $^{106}$ Ru was studied under four partial pressures of oxygen (PO<sub>2</sub>) in the carrier gas. For the former, an oxidizing environment with increasing amounts of oxygen resulted in less vaporization and less volatile element. On the other hand,  $^{106}$ Ru showed the

opposite behavior. This difference is due to the redox character of several species present in the biphasic system, mainly in the condensed phase and in the gas in equilibrium [36].

Ghiloufi and Amouroux [65] observed that  $^{137}$ Cs under different PO<sub>2</sub> behave similarly to  $^{60}$ Co, concerning the influence of the current on the volatility of radionuclides, the temperature and PO<sub>2</sub> were kept constant. The plasma current ranged from 0 to 600 A. The three radionuclides behaved in a similar manner, with an increase in the rate of vaporization and the amount of vaporized elements with the increase in the plasma current [65].

Ghiloufi [36] evaluated three matrices, which were silicon with basalt in different proportions or just basalt. Only <sup>60</sup>Co and <sup>137</sup>Cs were considered in this study because, in the liquid phase, the <sup>106</sup>Ru volatility would not change with any modification in the containment matrix. The increase in the percentage of silicon in the matrices improved the incorporation of <sup>60</sup>Co and <sup>137</sup>Cs through the formation of silicon oxides.

Yasui and Amakawa [63] studied the rate of vaporization of Cs from the slag by the plasma fusion process with an eye toward the definition of the optimum conditions of the process. Two parameters were considered: first, the chemical composition of each slag, which varied in terms of amounts of the oxides  $Al_2O_3$ ,  $SiO_2$ , CaO FeO,  $Fe_2O_3$ , MgO and Cs<sub>2</sub>O and second, the crucible's cross-sections were 0.254, 0.314, and 0.415 m<sup>2</sup>. The vaporization rate of Cs was evaluated in electric resistance and plasma melting furnaces, and the results indicated that the vaporization rate occurs mainly in a specific location, this location is the high-temperature region, where the plasma is connected [63].

Yasui and Amakawa [63] placed the plasma on the surface of the slag, and they observed that the how larger is the surface area of the crucible, less the influence of this high-temperature region. This result is probably due to the spread of material in the crucible, reducing the influence of this region. In addition, the constant values of the apparent vaporization rate reach a maximum value that does not increase with the increase in surface areas. However, they are dependent on the thermodynamic properties of the molten slag [66].

Nakashima et al. [64] investigated the treatment of various simulated wastes with the addition of radioactive tracers (<sup>60</sup>Co, <sup>137</sup>Cs, <sup>152</sup>Eu) using a non-transferred plasma torch, and the chemical compositions were evaluated by simulated ash based on a real Low-Level Waste. The mechanical resistance of the slag was also considered and the information was provided by various indexes of its physical properties. These indices were specific gravity, Vicker's hardness number and compressive strength of solid products. Through the results of chemical composition and mechanical resistance, the authors investigated the

homogeneity of the products and the behavior of selected radioactive tracers embedded in the waste. No more than 15% of the radionuclide concentration in the solidified product was identified, showing the approximately uniform distribution of these elements. On the other hand, the nonuniformity of these elements was also verified in some cases as a result of the higher coefficients of variation. According to the authors, this inconsistency is due to the contrasting melting conditions that the concrete fragments can undergo [64]. The complex interaction between the melted residues with the concrete, such as the sorption of radionuclides, can significantly interfere with the homogeneity of the treated wastes [67].

The reported differences from the aforementioned studies [36, 63–65] on the distribution and retention of nuclides may be related to the difference in plasma condition. These conditions include the type of plasma torch (transferred or non-transferred), chemical composition of slag, longer heating time, etc. However, the homogeneity of the slag is the crucial element for determining the inventory of radionuclides present in the process products [64].

#### **Concluding remarks**

Radioactive waste is generated at the stages of manufacturing and using nuclear fuel, the use of radioisotopes in industries, medical clinics, hospitals, and research centers, and materials removed from decommissioned radioactive facilities. However, these wastes require suitable treatments to ensure the protection of human health and the environment.

Studies for the treatment of radioactive waste using a variety of thermal methods are reported in the literature. But the treatment of radioactive waste by thermal plasma is still a subject of study with a wide variety of possible approaches regarding the configuration and constructive form of reactors and their operational parameters.

Most literatures have reported the technical feasibility of TPT in the nuclear and environmental field. However, it is quite confusing, that the applicability of thermal plasma for the treatment of radioactive waste is in accordance with regulatory, economic and socio-political factors. It is also observed that experimental conditions are different for each type of RW processed. The reported volumetric reduction values and application of the different study radionuclides of interest from various literature sources showed a wide variability of results. This huge variability might be resulted from different process conditions such as experimental methods, plasma torch, plasma reactor, process time etc. This variation makes it more difficult to study the ideal model for radionuclides transport and subsequently the plasma system performance. In addition, the investigation and characterization of the process products must be improved, as well as the standardization of the processing parameters must be tackled because this is still an impediment to the broad use of TPT on an industrial scale, affecting not only the applicability of technology but also the study guidelines of the process. All these improvements will result in the enhanced commercial viability of the process.

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