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Analysis of results from the operation of a pilot plasma gasification/vitrification unit for optimizing its performance

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Abstract

Plasma gasification/vitrification is an innovative and environmentally friendly method of waste treatment. A demonstration plasma gasification/vitrification unit was developed and installed in Viotia region in order to examine the efficiency of this innovative technology in dealing with hazardous waste. The preliminary results from the trial runs of the plasma unit, as well as the study of the influence of certain parameters in the system performance are presented and analyzed in this paper, contributing to the improvement of the operation performance. Finally, data on the final air emissions and the vitrified ash toxicity characteristic leaching procedure (TCLP) results are provided in order to assess the environmental performance of the system. The produced slag was found to be characterized by extremely low leaching properties and can be utilized as construction material, while the values of the polluting parameters of the air emissions were satisfactory. © 2007 Elsevier B.V. All rights reserved.

Keywords: Plasma technology; Gasification; Vitrification process; Slag

1. Introduction

The operation of a large number of industries located in Viotia region is a great source of waste with high polluting load. More particularly, about 18,000 t of hazardous waste are generated every year, and unfortunately, nonproper management practices are followed in some cases, which can constitute a danger for public health and environment. Moreover, as more and more wastewater treatment plants are applied for the effective management of the generated wastewater, the quantities of the generated sludge are substantially increasing.

Thermal waste treatment is attracting increasing attention as a viable alternative to landfill disposal. It reduces the volume of solid waste significantly but suffers from a bad reputation with the public, representing one of the top industries no one wants "in their backyard". Some of this fear stems from a lack of information about the process, but more justified concerns over emissions remain. Depending on the type and age of the technology applied, potential emissions include dangerous organic molecules, like furans and dioxins, incomplete combustion products, such as carbon monoxide, large amounts of particulates, as well as acid rain precursors like nitrogen oxides and sulfurous compounds. Furthermore, the management of the large amounts of bottom ash and potentially toxic fly ash can cause significant disposal problems in countries where landfill has already been banned [1]. The good news is that the technologies needed to remove the pollutants from the stack gas and clean the ashes are getting better [2,3]. Yet, they remain quite expensive. A promising method that can destroy waste at high temperatures is plasma gasification, which uses electricity to convert waste into a fuel gas and an inert rock.

In this framework, the scientific project entitled 'development of a demonstration plasma gasification/vitrification unit for the treatment of hazardous waste' aimed at studying the potential of plasma gasification and vitrification of waste (PGVW) technology [4]. This paper obviously constitutes continuation of the work described in Ref. [4].

The theoretical basis of plasmas and gasification/vitrification process, as well as different applications [5,6] in various countries were thoroughly investigated and led to the design of a pilot plasma unit, vitrifying the inorganic portion of the hazardous

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Fig. 1. Furnace and its related equipment.

waste into an inert, nonleaching rock, and gasifying the organic portion into a fuel gas, consisting mainly of carbon monoxide and hydrogen. Called plasma gasification and vitrification of waste, this process uses a high temperature furnace equipped with two graphite electrodes to generate plasma arcs. The plasma energy melts the inorganic portion of the waste and provides the initiation energy for the gasification of the organic portion [7].

The pilot PGVW system was designed to treat waste at high temperature in the furnace and treat the off-gas for subsequent release into the atmosphere. The treated waste should have a maximum particle size of 2.5 cm and maximum moisture content of 50%. In case, the particle size is larger, a machine for crumbling the treated waste is first used. If the moisture content is higher than 50%, the waste to be treated is dried in order to reduce the moisture content. The pilot plant has two main sections: (i) the furnace and its related equipment (Fig. 1) and (ii) the off-gas treatment system, including the secondary combustion chamber (SCC), quench and scrubber [4].

The trial tests of the plasma unit started in Canada using waste samples simulating the chemical composition of the waste generated in Viotia region and were continued in Greece after the installation of the facility in Viotia region. The waste samples that were used in Canada had the same origin (industries in the greater area of Montreal). Moreover, measurements and analyses took place in order to ensure that the samples were characterized by similar composition. Some modifications (e.g. by adding metal traces in order to raise the metal content of the waste input) were made when needed. The main advantage of the area selected for the installation and operation of the plasma unit in Greece is the fact that it is located near the industries that generate hazardous waste.

2. System fundamentals

Plasma is an ionized gas that conducts electricity [8,9]. In order for the air to conduct electricity, it must be subjected to a large differential in electrical potential. This is done between two electrodes, which are separated by air. When this potential is large enough, electrons can be pulled from the normally neutral molecules in the air. These electrons then move with the electric field and impact other molecules, releasing more free electrons at an exponential rate. This phenomenon is called electron cascade, and when adequate electrons are moving with the electric field, an arc is created between the electrodes. All of this occurs within a fraction of a second [1].

As an electrical circuit, the air gap where the arc is created can be viewed as resistance. While going through the resistance, the electrical current releases a large amount of heat. A large number of technologies have been developed to use this heat source that can reach temperatures exceeding $5000 \,^{\circ}\text{C}$ [1,10]. The main dissociation mechanism of hazardous waste is that chemical bond of hazardous material is degraded by the hightemperature plasma energy under the reducing or the oxidizing condition of plasma gas [11]. The tests described in this paper have been performed using air, but in fact, several gases are especially suitable for the technical realization of the processes and the operation of the unit [12,13]. Plasma gases can be divided into three classes depending on the availability of oxygen. These are:

- with free oxygen suitable for combustion (air, oxygen, and air enriched with oxygen);
- with partially free oxygen suitable for gasification (carbon dioxide and steam);
- without oxygen or with oxygen that is so tightly bound, that it is not available for oxidation processes (noble gases, nitrogen, hydrogen, carbon monoxide, and their mixtures). When no oxygen is present at all, the process of breaking down large molecules into smaller ones by heat, not by flame, is called pyrolysis [14].

In order to achieve the optimum results, depending on target pollutants and concentration of pollutants, different types of plasma processes and integrated pollution control systems can be used [15]. Nevertheless, the developed system has been designed to treat a large number of waste streams. The large size waste should be pretreated with a shredder. The shredded waste is loaded in the solid feeder when the system is cold, and the feed rate is calibrated. The waste size should be no more than approximately 25 mm diameter.

The feeding rate of the feeder should be calibrated for each waste feeding material. The feed rate varies depending on the bulk density of the test material. The system can be started in hot mode with residual slag from a previous run. In this case, the furnace is not opened, the electrodes are lowered to the surface of the slag and graphite pieces and/or metal shavings are placed between the two electrodes to initiate the arc. Graphite electrodes with male-female threads are used. The graphite-arc plasma system [1,16] uses the energy available in an electrical arc by transferring the energy directly to the material that will be destroyed. A high current (ranging from a few hundred amperes to several thousand amperes) is directed through long cylindrical graphite electrodes into the furnace, lined with refractory material. The electrical energy typically jumps from an electrode to another (nontransferred arc mode) or from an electrode to the waste (transferred arc mode), crossing air gaps in the process. The arcing generates tremendous heat, which serves to heat the waste. The temperature in the furnace can attain $1500 \,^{\circ}$ C, where the inorganic part of the waste under treatment is vitrified [17,18]. Vitrification is the process in which inorganic components (e.g. silicates) melt into a viscous liquid, which traps heavy metals into a solid matrix once solidified. It is a favored process for immobilization and destruction of dangerous waste, producing amorphous inorganic materials with high chemical durability [19–22]. The molten pool of hot slag at the bottom of the furnace serves as hot mass of energy, and in this way, waste can be processed at quite high feed rates.

The furnace is preheated using a portable propane burner. The purpose of preheating is to remove residual moisture from the furnace refractory and to ease arc start-up.

The off-gas system is started before the arc is started. During portable propane burner preheating, it is not necessary to start the off-gas system, as long as the bypass chimney is installed on the furnace. The system is controlled by the control program.

A procedure is established to control arc and power output. To start, low values of current and voltage are normally used, for example: 200–300 A DC and 150–250 V DC (approximately 75–100 V for each electrode). A common practice is to let the electrodes heat up in the position for a few minutes (3–10 min) to start up the arc and then slowly pull out the electrodes to increase arc voltage until a stable arc is obtained.

The DC power supply for the electrodes has a maximum power output of 200 KVA (plasma arc power supply, input: 600 VAC-30–60 Hz, $3 \text{ A} \times 200 \text{ A}$ fuses). Secondary current and voltage varies with tap selection.

Once the furnace has run for a few hours and the furnace temperature has reached sufficient level or if slag is present in the furnace and it is well melted, the feed can be started with a precalibrated feeding rate. Controlled values can vary widely from one run to the other and one type of waste to the other. Most of the data is recorded automatically by the computer and can be printed at the end of the trial. Additional data is recorded by the operators.

3. Testing the pilot gasification/vitrification system - results

Regarding the chemical composition of the waste generated in Viotia region, it should be noted that the NTUA working team has made a detailed inventory of the generated waste in the Viotia region. A large amount of visits took place in 281 units located in this region, waste samples were collected from different waste sources of the industrial units and were transferred to the lab of the unit of environmental science and technology for determination of the polluting parameters of the collected waste samples (measurements and analyses). In addition, the production processes of all industries, where visits were made, were recorded and the existing waste management systems were assessed. This paper focuses on results from the treatment of ash from foundries. The majority of the tests that are described below took place using ash derived from foundries. Data on the composition of the ash used in provided in Table 1.

Table 1				

Characterization of the	he waste input	(ash from foundries)
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Polluting parameter	Range (mg/kg dry weight)		
As	2.8–3.2		
Ba	160-240		
Cd	5-240		
Cr	195-8180		
Cu	125–19390		
Ni	24–495		
Pb	310-23500		
Zn	6100-115800		
SO_4^{2-}	12450-20220		
Cl ⁻	3450–5250		

3.1. Testing of the performance of the feeding system using different types and mixtures of waste

In the initial design, waste was introduced through the front side of the furnace and fell in a limited area, in a circle of approximately 30.5 cm in diameter. This method of feeding was found inefficient, as some localized areas of metal oxide reduction occurred, and waste only melted in a much-localized area.

Two actions were taken to improve the process:

- 1. The waste fed from the top instead of the side, and a refractory cone was used to spread the waste evenly over the surface of the molten bath. Hence, the surface of the whole bath was used to melt the waste, instead of a localized area.
- It was also found that keeping a layer of untreated waste on top on the molten pool improved the operation in two ways:

 (i) the arc was more stable (less frequent extinguishing of the arc) and (ii) the energy efficiency of the process was higher, since waste provided an insulation for the molten slag.

Fig. 2 shows the range of feed rates obtained with the pilot unit before and after the change in the feeding pattern. With side feed, feed rates higher than approximately 25 kg/h were difficult to obtain, because waste would not melt fast enough to allow for higher feed rates. With even feed from the top, much higher feed rates were obtained, up to 92 kg/h, almost double the original design feed rate of 50 kg/h. The influences to the feed rates were similar for all kinds of waste fed to the system.



Fig. 2. Range of feed rates with side feed and even top feed.



Fig. 3. Electrode consumption under different environment.

3.2. Performance of the graphite arc system using mixture of sludge

Graphite electrodes with male–female threads are used. The electrode dimensions were 7.6 cm in diameter and 106.7 cm in length.

Electrodes are installed with the female end down in order to avoid dust accumulation in the threads. Two electrodes were screwed together on each side (anode and cathode).

The graphite electrode consumption was measured under nitrogen and air atmosphere.

Electrode consumption was measured in grams per hour. This gave the best basis for comparison. The electrode consumption was four times higher in the air atmosphere compared to the nitrogen atmosphere (Fig. 3).

The electrode consumption, due to erosion, was estimated by measuring the electrode length change and multiplying it by the linear density of graphite (72 g/cm). The linear density was measured on the received electrode: 106.7 cm long by 7.6 cm in diameter with a weight of 8.5 kg. Using the average electrode density (1.66–1.73 g/cc) and a diameter of 7.6 cm, the linear density was very similar: 77 g/cm.

Considering that the consumption under a partially oxidizing atmosphere corresponds to 7 kg/t, electrode consumption could theoretically be reduced to approximately 4 kg/t if oxygen levels in the furnace were kept at 1-2% by volume. Assuming that all consumption due to oxidation could be eliminated, a consumption of 3 kg/t could be achieved.

3.3. Graphite arcs control system evaluation

The graphite arcs were controlled manually by setting the position of the electrodes. The relation between the voltage and the relative position of the cathode and anode was studied (Fig. 4). The zero point is at the molten slag surface. It was found that electrode voltage could be very well controlled by position. Therefore, future systems could include automatic voltage control.

The pivots of the graphite electrodes were cooled using water-cooling to reduce graphite oxidation. This led to improved



Fig. 4. Cathode and anode voltage.

electrode consumption. The cooling was also necessary to limit the heating effects outside the furnace.

3.4. Slag tapping mechanism evaluation

Regarding tapping, as feeding continues, the volume of molten slag increases and the effective space of the furnace reduces. Slag must be tapped out of the furnace once the level is close to the maximum of the crucible.

The tapping hole was plugged with a carbon-based paste during the operation of the furnace. Two methods of tapping were investigated [4].

Tapping of slag produced two types of products:

- solid ingot by tapping into molds (Fig. 5);
- slag granules by tapping into water.

Good physical and mechanical properties can be achieved and the results could be comparable with the commercial products [23].

Fig. 6 provides an example of the produced granules' size distribution.



Fig. 5. Slag ingot in pouring mold.



Fig. 6. Slag granule size distribution.

3.5. Level control of molten pool

During all the pilot studies, the slag tapping was only conducted periodically. The level control of the molten pool was not part of this study. Continuous tapping of the molten slag is commonly used in the industry, and the level control of the molten pool should not represent any technical difficulties.

3.6. Accuracy of data acquisition

Special attention has been given to the type of temperature sensors used in and around the furnace and the secondary combustion chamber due to the high temperatures encountered at various points in the system.

Type K or J thermocouples have been used for all cold temperature duties (cooling water, quench, scrubber, etc.). Type B was successfully used in all high temperature (over 800 °C) locations, the furnace, and SCC. Ceramic sleeves provided a good shield for extremely high temperature locations inside the furnace and the SCC.

The data acquisition system provided a fairly accurate representation of system operation. The information was further processed using an excel spreadsheet to recover various trends (like temperature, voltages, power, etc.) during the experimental operation.

3.7. Off-gas combustion system

The performance of the off-gas combustion system was assessed during the experimental trials. Automatic flame detec-

Table 2			
Effect of air and	steam	injection rate	

tors ensured that the propane was cut off if the flame was lost on any one of the burners. The combustion of the synthesis gas from the PVGW (plasma vitrification and gasification of waste) furnace maintained a temperature around 1000–1100 °C in the SCC. This temperature ensured full combustion of the synthesis gas (carbon monoxide and hydrogen) to carbon dioxide and water. The main system blower maintained a negative pressure in the SCC ensured effective evacuation of all combustion gases to the system stack. No operational problems were encountered with the SCC.

3.8. Furnace operation

A large number of trials were performed on the furnace using various combinations of waste. Waste from other sources was also tested in the system so as to determine if any difference in the mode of operations could be identified.

Maintaining the furnace under constant negative pressure was challenging at times. If a cold top was not maintained, the gasification of the organic portion of the waste led to occasional system pressurization, causing gas leaks around the furnace. For best furnace operation, close control over the level of the cold top in the furnace was essential. The presence of a cold top and the distribution of the batch in the melter strongly influence the system performance [24–28].

3.9. Location of air and steam injection into the furnace

The location of the air injection into the furnace was found to be very important for achieving an effective gasification process. The best location was found to be at the center of the furnace, inside the cold top, not too far from the slag/cold top interface.

3.10. Waste to air and steam feed ratio

This following series of tests (Table 2) were performed to evaluate the impact of gasification air and steam flow on synthesis gas composition. Ash from foundries located in Viotia region was used in order to make conclusions. It was found that adding air only had a dilution effect on the gas. The heating value of the gas produced with the highest airflow rate was found to be the lowest.

ng value (MJ/Nm ³)

The addition of steam marginally improves the heating value of the synthesis gas under no air addition conditions. Too much steam can dilute the synthesis gas, and as such, reduce its heating value.

3.11. Waste residence time

The unprocessed waste forms a layer (cold top) on top of the molten slag in the furnace. This arrangement has provided the optimum condition for controlled gasification of the organic portion of the waste. Since the system was only tapped periodically, it is difficult to provide a definite residence time estimate. At the end of an experiment, sufficient time was provided to melt the cold top before tapping. An average residence can be estimated from the residual heel of molten slag in the furnace of 6-8 cm. This provides an estimated waste residence time of 1.5-2 h.

3.12. Effect of gasification air on synthesis gas composition

DC steam plasma can be used to treat carbonized wastes, such as the carbide of the hazardous waste, in order to reduce the weight of graphite and to produce the combustible gas from graphite [29]. The effect of air feeding to the system was tested. Evaluation of the effect of gasification airflow rate on synthesis gas composition was part a critical parameter for studying. By reducing the amount of air, carbon monoxide, and hydrogen content of the syngas increased, improving its heating value. It is observed from the test results that the CO and H₂ content varies as a function of air flow rate when other operating conditions and the type of waste are fixed. Thus, it has been demonstrated that, by adjusting the amount of air fed to the furnace, the composition of synthesis gas can be controlled.

3.13. Effect of water content

Plasma technology is characterized as flexible and the change in the composition of the waste treated has no noticeable effect on the operation of the demonstration plasma unit. This was the outcome of experiments with a variety of waste types. As mentioned before, the waste can have maximum moisture content of 50%. In order to investigate the effect of water content, several trial runs were carried out, which showed that the water content quantity does not cause unusual disturbances in the operation of the plasma unit and the system demonstrated great flexibility in treating higher water content waste.

3.14. Environmental performance

Without doubt, the environmental performance of a thermal treatment system is of paramount importance for assessing its effectiveness, reliability, and application potential and ensuring public acceptance. The evaluation of the environmental performance of the developed system was based on two main figures:

- the quality of the produced off-gases (air emissions), being the main concern for all thermal methods;
- the toxicity characteristic leaching procedure (TCLP) tests performed for the vitrified slag.

The most crucial point of the operation of a thermal treatment unit is the nature and the amount of the final air emissions. At first, it should be noted that the amount of the gases generated is quite low, since the amount of the available oxygen is less at the gasification/vitrification process. The demands of the European legislation concerning air emissions from the thermal waste treatment have become strict nowadays because of the awareness and the pressure of the public opinion towards a safer and cleaner environment. These demands are contained in the directive 2000/76/EC of the European parliament and of the council of 4 December 2000.

Next, the results from the analysis of the final air emissions from the ash treatment are presented (Table 3).

The above tables include the average measured values. It should be noted that the measured values of the polluting parameters of the air emissions did not present great variations in function of time, excluding the start of the operation of the pilot plasma unit. Apart from the average values, the relevant limits of directive 2000/76/EC were not exceeded in any case, too.

The low measured values of acids (SO_2, HCl, HF) show that the operation of the packed bed scrubber is effective and the acid components of gas are neutralized by the caustic soda solution. In addition, the low measured values of carbon monoxide (CO)

Table 3

Measured daily average values of final air emissions and daily average limit values (directive 2000/76/EC of the European parliament and of the council of 4 December 2000 on the incineration of waste)

Polluting parameter	Measured daily average value (mg/m ³)	Daily average limit values according to directive 2000/76/EC (mg/m ³)
Total dust	5.5	10
Organic substances as gases or steam, measured as total organic carbon (TOC)	2.0	10
Hydrogen chloride (HCl)	2.9	10
Hydrogen fluoride (HF)	0.4	1
Sulfur dioxide (SO ₂)	14.2	50
Nitrogen monoxide (NO) and nitrogen dioxide (NO ₂), calculated as NOx for existing units with capacity of over three tonnes of waste per hour or new units	102.6	200
Carbon monoxide (CO)	2.1	50

Table 4 Vitrified ash TCLP results

Metal	Sample (mg/L)					
	1	2	3	4		
As	<0.002 (N.D.)	<0.002 (N.D.)	<0.002 (N.D.)	0.03		
Ва	<0.4 (N.D.)	<0.4 (N.D.)	<0.4 (N.D.)	0.96		
В	<0.3 (N.D.)	<0.3 (N.D.)	<0.3 (N.D.)	1.4		
Cd	0.010	<0.006 (N.D.)	0.006	<0.01 (N.D.)		
Cr	<0.03 (N.D.)	<0.03 (N.D.)	<0.03 (N.D.)	<0.01 (N.D.)		
Cu	0.08	0.19	<0.03 (N.D.)	-		
Ni	<0.04 (N.D.)	<0.04 (N.D.)	<0.04 (N.D.)	-		
Pb	0.55	0.18	<0.06 (N.D.)	<0.01 (N.D.)		
Se	0.004	<0.004 (N.D.)	0.009	<0.01 (N.D.)		
Zn	0.46	0.13	<0.02 (N.D.)	-		
Hg	<0.0002 (N.D.)	<0.0002 (N.D.)	<0.0002 (N.D.)	<0.0004 (N.D.)		

N.D., non detectable.

1, Bowater granules; 2, Bowater hard block; 3, HSPP glassy rock; 4, HSPP fly ash rock.

prove the effectiveness of the secondary combustion chamber. Its role to convert the synthesis gas (CO, H_2), produced in the gasification/vitrification furnace, to CO₂ and H_2O is achieved. Also, the operation of the hot cyclone resulted in low concentrations of particulates in the final air emissions.

The measured values of the polluting parameters of the air emissions show that the plasma technology is environmentally friendly thermal method for ash treatment. In fact, this constitutes the major advantage of the proposed innovative technique, which easily meets the strict demands of the recently set legislative framework regarding the air emissions.

Toxicity characteristic leaching procedure tests were performed on a number of slag samples. TCLP gives a measure of the stability of a solid waste. The measured TCLP numbers were compared to the EPA regulation (Table 4). For all the samples tested, the TCLP measured was well below the regulated limits, often orders of magnitudes below. For example, the arsenic level is regulated at 5 mg/L. The measured level was less than 0.002 for three samples and 0.03 for another sample, less than one hundredth to a thousandth the regulated limit.

4. Discussion and conclusions

The plasma process is capable of treating all types of mixed waste due to the extreme high temperature and drastic conditions achieved. Plasma units are able to treat separately or mixed municipal solid waste, industrial solid or wastewater, oily waste, medical, tires, sewage sludge, etc. due to the fact that the heat necessary for the gasification derives from the plasma and not from the waste oxidation. The flexibility regarding the waste that can be treated with plasma process is an advantage of vital importance. Of course, the expansion of the use of plasma technology for the management of several waste streams would require the modification of several operating parameters and further investigation. A fundamental advantage of the plasma gas. This also means the free choice of the reaction atmosphere.

The vitrification process converts the inorganic components into a solid rock with extremely low leaching properties that can

Furthermore, the limited application of thermal methods in Greece for the effective waste management supports the use of plasma technology for the treatment of other waste streams in future. Necessary precondition for the feasibility of such a plan is the energy recovery from the produced synthesis gas. Therefore, the economic feasibility of applying plasma technology is greater in the case of waste having major organic part, because more organic part means more gasified product and substantially more energy available for recovery, since a clean syngas can be converted to energy quite efficiently using either a turbine or an internal combustion engine. Furthermore, the syngas can be turned into other gases or chemicals, such as methane, hydrogen, or methanol. There are several advantages to producing a syngas rather than a fully combusted gas. Gasification leads to the production of a smaller gas volume, because no fossil fuels are burnt to generate heat and there is no need for excess oxygen as in the majority of the other thermal treatment processes. In addition, smaller gas volume means lower expenses for being cleaned.

It is true that due to their high cost, plasma systems have been used primarily for the vitrification of high toxicity waste and mainly at pilot scale. As these systems become more accepted and their design becomes simpler, the use will be more extensive. Finally, a long testing period is also of vital importance for achieving optimum results.

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