SET-UP AND FIRST OPERATION OF A PLASMA OVEN FOR TREATMENT OF LOW LEVEL RADIOACTIVE WASTES

by

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An experimental device for plasma treatment of low and intermediate level radioactive waste was built and tested in several design variations. The laboratory device is designed with the intention to study the general effects and difficulties in a plasma incineration set-up for the further future development of a larger scale pilot plant. The key part of the device consists of a novel microwave plasma torch driven by 200 W electric power, and operating at atmospheric pressure. It is a specific design characteristic of the torch that a high peak temperature can be reached with a low power input compared to other plasma torches. Experiments have been carried out to analyze the effect of the plasma on materials typical for operational low-level wastes. In some preliminary cold tests the behavior of stable volatile species *e. g.*, caesium was investigated by TXRF measurements of material collected from the oven walls and the filtered off-gas. The results help in improving and scaling up the existing design and in understanding the effects for a pilot plant, especially for the off-gas collection and treatment.

Key words: low level waste, plasma technology, radioactive waste treatment

INTRODUCTION

Radioactive waste from nuclear power plants has to be stored and isolated from the environment in adequate repositories as long as the radioactivity is above acceptable levels. Appropriate long-term storage has to provide a high degree of isolation and retention of the radioactive wastes, ideally without further maintenance. To reach that goal, the waste has to be conditioned, processed and packaged in an appropriate way for easy and reliable handling. The volume of the radioactive wastes and therefore the storage casks, must be as low as possible due to limited space in the repositories. There should be no leaching of radioactive substances from the final packed containers.

Conditioning of the waste is usually done after sorting according to the properties of the waste components. The aim is to minimize the volume of the final product while meeting the regulatory limits on the specific activity per cask.

A comprehensive alternative to conventional low and intermediate level radioactive waste treatment strategies would be plasma treatment, which can be applied to all kinds of materials and results in a volume reduced and stable final product [1].

BACKGROUND

All waste produced during operation of nuclear power plants is usually declared as radioactive and has to be handled specifically. This includes mainly contaminated materials from the working environment, *e. g.*, clothes, gloves, packaging foil *etc*.

When applying plasma to wastes, the waste components are decomposed. Combustible parts are incinerated. The molecular structures of noncombustible components are broken apart. Metallic solids are molten. The peak temperature of the plasma is high enough to decompose any material [2]. The intended effects when treating mixed radioactive operational waste from nuclear power plants are volume reduction and solidification of the residual material. The motivation is to treat all the material in a mixture as produced.

Plasma as thermal treatment technology is applied in various fields for treatment of different hazardous waste materials. Plasma treatment of low level radioactive material is performed in prototype plants, but cannot yet be regarded as an established method [3, 4].

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Some functionalities of plasma treatment are possible to observe in small scale experiments. An experimental device was set-up at the FH Aachen Campus Julich, Germany, for testing of design features and plasma specific properties related to radioactive waste treatment. The main aim is to verify the known advantages of plasma technology on a small scale and in addition, to identify the problems and the properties that can influence the design of a full scale device.

LABORATORY SET-UP

A newly developed microwave plasma torch and a high frequency generator by HHFT was used in our studies [5, 6]. Major advantages of this specific application to common plasma torches, are: the torch provides a high thermal efficiency and is free of maintenance. In addition, a process gas of choice can be applied to allow *e. g.*, combustion in air. The torch is driven by a 2.45 GHz microwave signal. The input power can be regulated up to 200 W. The torch is built up with a hollow copper cannula as an electrode. The length of the copper electrode is $\lambda/2$, with $\lambda = 12.23$ cm being the wavelength of the signal. The plasma gas flows through the cannula and is ignited at the tip where it gets ionized by the high electric field power. The ignition frequency of the torch utilized is 2.40 GHz and the operating frequency is 2.44 GHz as found by calibration. The plasma gas flow has to exceed 0.2 L/min. Welding protective gas (argon) can be used or an argon 98%/hydrogen 2% mixture. In the experiments described in this report the latter was used. A second gas is cooling the cannula and helps focusing the plasma flame. Compressed air was used with a high flow above 2 L/min for this purpose [7].

The thermal power needed for complete treatment of the waste material would then be induced by conventional heating in an induction oven or, in the case of this laboratory device, by a gas burner.

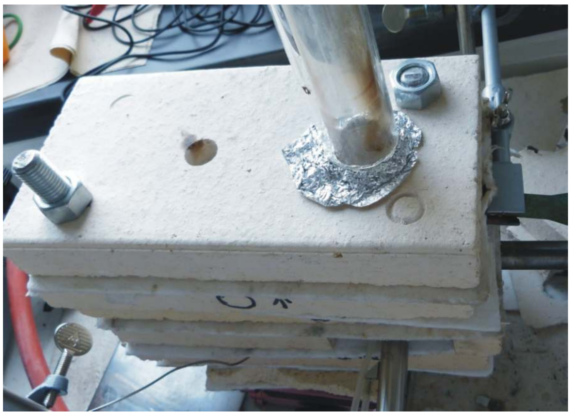

Figure 1. The assembled oven with conventional and plasma burning chambers

The laboratory set-up was planned with the idea of a simplistic and versatile model. The aim was that the setup would be easily amendable to adapt to any new experimental results. The main modifications are an inlet for the plasma torch, one optional inlet for a gas burner, an off gas tube and an inlet for the specimen. Figure 1 depicts the fully assembled oven. The device has been designed as a modular oven with different burning chambers in order to compare the outcome for plasma treatment, conventional burning and combinations of both. For the oven piece a cylindrical piece of a steel tube was used (fig. 2). An aluminum foil bed was added for holding the specimen. The torch power is low enough not to melt down parts of the oven. The effect of the plasma on the material to be treated was of interest. The choice of materials was governed by a detailed study on the composition of operational wastes of nuclear power plants in Europe [8].

Figure 2. Plasma torch in operation inside metallic oven piece

EXPERIMENTS

The aim of these experiments is to prove the correctness of the claim that – in general – all material is treatable and the treatment effects observable especially on this low scale. In addition, the role of the specimen size on the treatment time and outcome was investigated. The assumption should be confirmed, that the required treatment time and energy decreases with higher fragmentation of the specimen. Some experiments involving glass particles were done to find out if any vitrification occurs.

Table 1 lists the experiments of short time plasma treatment of various materials with the specimen mass and treatment time and short remarkable observations.

Activated charcoal was expected to burn away. In addition, some bubble forming on the surface of the charcoal was observable, as shown in fig. 3. Besides evaporation of volatile species formed in the production of charcoal, the peak temperature in the plasma may have been so high, that not only (chemical) burn-

Table 1. Short time plasma treatment of various materials – experiments

Material	Mass [g]	Time [s]	Observations and remarks
Activated charcoal	0.2	120	Completely vanishes, Melting was observable
Paper	0.2	60	No residue
Quartz wool	0.15	60	Volume reduction
Tin plates	0.4	300	No effect
Latex gloves	0.15	60	Glued to oven walls, a lot of smoke production, no residue
Glass particles	0.7	30	Melts together to a single droplet
Brazen chipping	0.3	300	Melts together
PE	0.2	90	Ash, smoke
Textiles	0.2	60	High flame, no residue
Cable	0.3	60	Isolation burned, metal melt where flame touched
Wood	0.3	120	Burns down, orange flame
Mixture	0.5	300	Burns down
Mixture + Glass	0.7	600	Combustibles partially melt together in glass droplet
Brazen chipping + Glass	0.75	460	Melt together to brown, grainy particle
Aluminium chipping	0.33	20	Partially molten
Iron chipping	0.25	50	Sparks, partially molten
Stainless steel chipping	0.67	80	Sparks, partially molten
PVC	0.15	120	Lots of dark smoke, residue on walls
Acrylic glass	0.16	10	Melts away
Stone salt	0.2	30	Breaking of structure, melting
Tungsten wir	–	40	Exp. in air, flame on a spot, wire melts down on that spot
Ceramic tile	–	30	Exp. in air, where the flame touches the ceramic melts

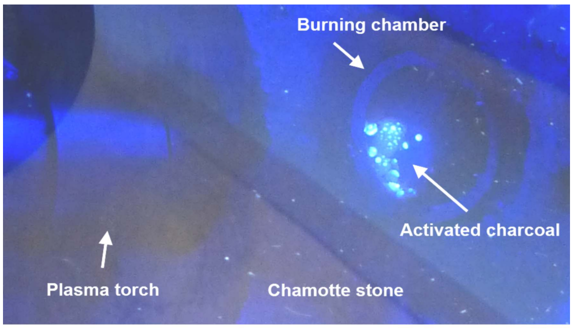

Figure 3. Plasma treatment of activated charcoal observed through a cobalt-glass

ing but physical melting and gasification occurred (melting point: 3550 °C, gasification point: 4027 °C). Some materials that are combustible, but are sorted to the non-combustible waste like PE, PVC, and acrylic glass have been treated with plasma and have burned away. Under unfavorable conditions these materials may form toxic compounds in the flue gas. Hence, detailed investigations and analysis of the flue gas should be performed to establish conditions avoiding their formation.

The various metal chippings showed an effect where the plasma flame touched the material: a spherical melting point was visible at these spots. Some particles were also sintered.

Glass melts down while keeping its volume and mass. The effect of the grain size of glass particles was examined. For this experiment, some glass was crushed and sorted by size. It could be observed that the melting proceeds faster when the particle size is smaller.

The so called "Mixture" in tab. 1 is a typical scaled down composition of combustible material from nuclear power plant operation. In detail the mixture used in this experiment consisted of 0.1 g textiles, 0.17 g PVC and 0.08 g latex glove (synthetics), and 0.12 g charcoal (other). In a subsequent experiment 0.23 g glass (incombustibles) was added. The mixture was easily treatable. In the experiment with addition of glass some dark spots could be found on the molten glass droplet indicating a partial vitrification of the combustibles (see fig. 4).

When treating a mixture (50:50) of brass grains and glass particles, one observed a large grainy, brown droplet had formed besides some residual brass grains.

Tungsten wire was examined to see if the effective temperature is high enough to melt even high temperature resistant material (melting point: 3422 °C). When treating a wire fixated in front of the plasma

Figure 4. Plasma treatment of a waste mixture + glass particles melt together

torch, it was observed to break after ~40 s of treatment. A very bright glare was seen through a cobalt glass used to protect the eyes.

In treatment of ceramic tiles only a local effect around the plasma focus was observed: the surface melts with an appearance similar to molten glass.

We found materials to behave differently in plasma incineration, plasma aided incineration and conventional combustion. The most remarkable effect could be seen when treating quartz wool (see fig. 5). Whereas with conventional burning no visible effect could be observed, the plasma flame burned the quartz wool down. The material shrank significantly where touched by the plasma flame and its volume was highly reduced.

The power required for complete treatment is mass-dependent and necessitates an up-scaling of the plasma torch and its electric power supply. At the current power level the plasma torch is not suitable for treatment of massive materials. For future applications an upgrade of the torch for power inputs of several kW is foreseen.

Caesium-137 is a radionuclide with a half-life of 30.1 years volatilizing at 671 °C. ^{137}Cs is a leading nuclide for all radioactive waste material. Due to its low gasification point, it is a special threat in heat treatment. The aim of this experiment is to find out the effect of the plasma on caesium and analyze the distribution of the caesium in different parts of the oven after treatment. Filter papers were impregnated with low concentrations of Cs salts and plasma treated. Caesium was washed from the oven piece and the off-gas tube by demineralized water. The resulting solutions were analyzed for Cs by TXRF system.

It was found that >97% of the Cs was recovered from cold surface areas of the assembly as expected. Only <3% could not be recovered. That may be lost in the off-gas or through leakages or by the dissolution process. The well-known behavior of Cs is therefore confirmed and reproduced for high temperature plasma combustion. In a set-up with a high temperature in a larger volume, such as an industrial oven, one should foresee a zone to collect the volatile Cs, *e. g*., at the first cold surface in the off-gas stream or by chemical absorbers.

Figure 5. Comparison of plasma treated (left) and conventionally thermal treated (right) quartz wool

About two thirds of the total Cs was found to deposit locally in the burning chamber and 1/3 to travel with the off-gas stream to the region of the off-gas tube. This distribution, however, seems more to be a characteristic of this specific oven setup and not for plasma incineration in general. The deposition of caesium in different oven designs will be investigated in further experiments.

CONCLUSIONS

Several conclusions can be drawn for up-scaling of the effects and development of a full scale plasma treatment device for radioactive wastes from the experiments described.

It is indeed possible to treat all kinds of materials by applying plasma. In principle, the temperature is high enough to incinerate and melt even a material of high temperature resistance, such as iron, tungsten and ceramics.

The thermal power output of the torch is an important factor to consider. In our experiments it was not possible to melt down larger specimens due to the low power of the plasma torch (200 W) applied. In addition to increasing the power, it is helpful to also decrease the size of the material to be treated, by controlled crushing or milling.

Plasma, as a thermal treatment technology, has some clear advantages compared to conventional combustion. Not only can all materials be treated due to the very high plasma temperature, but also the combustion process is faster and more effective.

Caesium volatilizes under the influence of plasma. In our experiments it was found locally close to the treatment zone deposited on cold surfaces. Only a small fraction of the total caesium amount could not be recovered. When regarding a full scale plasma treatment plant, one has to account for a caesium collection device in the flue gas stream to prevent contamination by radioactive caesium. It is hoped that accumulation in the oven resulting in dose build-up can be avoided by improved design of the combustion chamber.

AUTHOR CONTRIBUTIONS

Theoretical analysis was carried out by F. Nachtrodt and experiments were carried out by F. Nachtrodt and U. W. Scherer. All authors analysed and discussed the results and prepared the manuscript.

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ПОСТАВЉАЊЕ И ПРВО ПУШТАЊЕ У РАД ПЛАЗМА ПЕЋНИЦЕ ЗА РУКОВАЊЕ РАДИОАКТИВНИМ ОТПАДОМ НИСКОГ НИВОА

Експериментални уређај за третирање плазмом радиоактивног отпада ниског и средњег нивоа направљен је и тестиран у неколико варијанти. Лабораторијски уређај дизајниран је са намером да се изуче општи ефекти и потешкоће код спаљивања плазмом у циљу будућег развоја већег пилот постројења. Кључни део уређаја је нови микроталасни горионик плазме којег покреће електрична снага од 200 W и који ради при атмосферском притиску. Специфична карактеристика дизајна горионика омогућује постизање високе температуре са ниском снагом у поређењу са другим горионицима плазме. Експерименти су спроведени на материјалима који се типично појављују као отпад ниског нивоа. У прелиминарним хладним тестовима испитивано је понашање сталних испарљивих врста, на пример, цезијума, користећи TCRF мерења материјала сакупљених са зидова пећнице и филтрираног гаса. Резултати дају допринос побољшању и скалирању постојећег дизајна, као и у разумевању ефеката код пилот постројења, нарочито за третирање и сакупљање гаса.

Кључне речи: отпад ниског нивоа, плазма технологија, руковање радиоактивним отпадом