PARTICULATE GENERATION FROM THE VITRIFICATION PROCESS OF SIMULATED DRY ACTIVE WASTE IN A PILOT FACILITY IN KOREA

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ABSTRACT

Pilot scale vitrification tests on low- and intermediate-level radioactive wastes are being performed for the commercialization of the process in Korea. In order to evaluate and finally optimize the vitrifying condition for combustible dry active waste (DAW) from nuclear power plants, dust generation characteristics were investigated for various waste types (polyethylene, cellulose, mixed waste, etc.), melter configuration, excess oxygen amounts and waste feeding rates. Particulate concentration and size distribution in the off-gas were analyzed using an isokinetic dust sampler and a cascade impactor, respectively. Amount of dust collected at the bottom of a High Temperature Filter was measured to evaluate the total dust generation ratio and to analyze the elemental compositions. Results showed that operating parameters, such as waste types, melter configuration and duration of waste feeding time, etc., affected the dust concentration. Carbon content in the dust, which implies combustion efficiency, was proportional to the dust generation ratio and greatly reduced by adjusting the melter's configuration and increasing the oxygen amount. Size distribution of particulate generated during a treatment of a mixed DAW showed that the ratio of the large particles increased by decreasing the oxygen amount fed into the melter. Evaluation of the dust characteristics makes it possible to optimize the process parameters and to determine the effective dust collection equipment.

INTRODUCTION

NETEC-KHNP (Korea Hydro & Nuclear Power Co. Ltd.) is developing a vitrification technology to treat low- and intermediate-level radioactive wastes generated from the pressurized water reactors. A pilot plant consisted of a cold crucible melter (CCM) heated by an induced current, a plasma torch melter (PTM), and an off-gas treatment system (OGTS) was constructed by KHNP, Hyundai MOBIS, and SGN in Korea. By vitrifying solid and liquid radioactive wastes generated from the nuclear power plants, it is possible to immobilize hazardous isotopes and components in a more chemically durable glass matrix with remarkable volume reduction. KHNP has successfully completed its 1st phase pilot plant performance tests for various waste types.

The purpose of the performance tests was to confirm a feasibility of the vitrification technology as a whole process including the off gas treatment system, to optimize the operation parameters, and to evaluate the performance of each equipment. The final objective will be the acquirement of detailed design data for the commercialization of vitrification technology in Korea.

In Korea, no clear guidelines for vitrification facility are formulated yet. However, it is thought that most requirements applied to the industrial incinerator and the radwaste treatment system are also applied to the commercial vitrification facility. In this study, following requirements are considered.

- Complete combustion of organic materials
- Minimization of the volatilization of hazardous components
- Minimization of the radioactive by-product generation

The first requirement, complete combustion, is the essential one to be satisfied to reduce the generation of CO, PICs (products of incomplete combustion) and particulate matters in the industrial incineration system. It is reported that most dioxins are formed from the carbon in fly ash in the cooling process of off-gases and metals such as Cu and Fe included in dust serve as catalysts for the formation of dioxin (1). On the other hand, chlorine content in waste streams is not significantly related with the dioxin formation (2). It means that the dust entrained in the off-gas is directly related to the dioxin formation. Therefore, the complete combustion must be one of the basic requirements for vitrification process.

The second one is an important issue in the radwaste treatment process since it can affect the system safety and operation/maintenance cost as well as the waste glass characteristics. The main purpose of the vitrification is to separate radionuclides and hazardous components from waste feed and to incorporate them into the glass matrix. In this respect, volatilization of such species can be one of the most severe problems in high temperature vitrification process. Therefore, volatilization characteristics of hazardous chemicals should be investigated in the pilot plant tests.

Ash or residues not incorporated into the glass matrix or not properly thermal-treated (e.g. small residence time) can be entrained into OGTS by the extraction fans, and cause an increse in the dust level significantly. In particular, radioactive dust entrainment may require an additional treatment process for secondary waste and special consideration for the protection of workers. Therefore, by-product generation should be minimized by regulating operation and/or design parameters.

These three requirements are interleated each other. If wastes are completely oxidized with sufficient retention time and oxidative environment, entrained dust amount that may include radionuclides can be reduced. It also results in the less amount of radioactive by-product during the vitrification. Therefore, optimizing the parameters to meet these three requirements will be one of main purposes of the pilot plant tests.

In this paper, dust characteristics generated from the melter were analyzed to determine the optimum conditions for the vitrification of the dry combustible wastes. Dust generation amount was used as a parameter for the quantative evaluation of the vitrification condition. It is because the amount of particulate has a close relationship with all three requirements mentioned above and particulate removal can play an important role in controlling the emissions (particulates themselves, toxic metals, radionuclides, and hydrocarbon) from a thermal treatment process (3).

Waste Types

The combustible dry active wastes (DAWs) are being generated at about 41 % of overall low- and intermediate-level radioactive wastes from Korean nuclear power plants (4). It consists of clothes, papers, plastics, woods, etc., and compacted in drums for temporary storage. Investigation has been undertaken to examine the material composition of the DAW and its generation ratio (see Table I). Results showed that the wastes mainly consist of six materials and the cellulose is a dominant one (47.4 %).

For pilot plant tests, four materials (cellulose, PVC, PE, and rubber) were selected as representatives of the combustible DAWs because their proportion exceeds 90 %.

Table I. Material composition of DAW

Wastes	Cellulose	PVC	PE	Rubber	Polyester	PP	SUM
	56.0			9.6	34.4		100
Papers	93.8		6.2				100
Vinyl		61.3	38.7				100
Rubber/Plastics	Clothes		7.1	92.9			100
Lancing Filter						100	100
Woods	100						100
AVERAGE	47.4	18.6	14.5	10.8	8.1	0.6	100

Vitrification Process

In our pilot system, the DAWs are directly fed into the melter after being shredded into 5 x 5 mm of nominal size. Direct vitrification may entail combustion, oxidative pyrolysis, pyrolysis, or combination of these processes depending largely on the concentration of oxidants. Inorganic and metallic components in the wastes will be incorporated into molten glass. Off-gases produced from the direct vitrification process are drawn to OGTS by the extraction fan. Particulates and hazardous gases such as CO, SOx, NOx, HCl are removed by proper treatment methods.

A waste feeder supplies the waste materials continuously into the upper part of the melter at a constant rate. Off-gases from the melter are cooled when passing a Pipe Cooler, then particulates are filtered out inside a High Temperature Filter (HTF). The main function of the HTF is to remove particulates from the off-gas stream and, thereby, to protect the following system from the radioactive contamination. It is equipped with 64 ceramic filters and maintained inside temperature at 150 ~ 200 ℃ to avoid moisture condensation in the filters. Clogged dusts on the outside surface of the filters are removed by the pulse-jet declogging method and fallen down to the drum of the filter bottom.

The gases from the HTF are destroyed and cleaned passing through Post Combustion Chamber (PCC), Wet Scrubber, HEPA filter, and NOx removal system (SCR; Selective

Catalytic Reduction). Finally, gases are released to stack and monitored by the on-line Continuous Emissions Monitoring System.

PILOT PLANT TESTS FOR VITRIFICATION

Operation Parameters

In order to optimize the DAW vitrification process, four operation parameters were selected as follows: waste type, configuration of the melter, wastes feeding rate, and excessive oxygen amount. Pilot plant tests were performed at various conditions combining the four operation parameters and dust generation characteristics were analyzed. Operation time for each condition was about 1 to 3 hours.

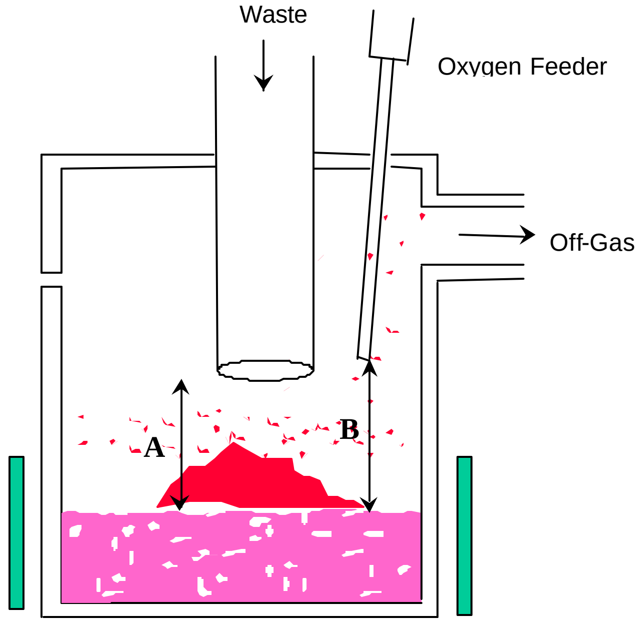
Waste type : Three types of the wastes were tested; cellulose, PE, and mixed waste (50% of cellulose, 20% of PVC, 20% of PE, and 10% of rubber). The wastes were fed into the melter at a constant rate after shredding. Table II shows the elemental composition of each waste type calculated based on the material compositions.

Waste feeding rate: Pilot plant tests were performed at 5 to 20 kg/h of waste feeding rates to determine the maximum feed capacity of the CCM.

Table II. Theoretical elemental composition of DAW (Unit : %)

Element / Waste	C	H	O	Cl	SUM
Cellulose	44.4	6.2	49.4	-	100.0
PVC	85.7	14.3	-	-	100.0
PE	38.4	4.8	-	56.8	100.0
Rubber	88.2	11.8	-	-	100.0

Configuration of melter: As shown in Fig. 1, the waste feeding pipe is connected to the upper chamber of the melter, and six oxygen injectors are installed around the central feeder. It was found from the previous resin tests that heights of the feeder and oxygen injectors are important factors affecting the combustion condition. Three conditions are selected to optimize the melter's configuration as shown in Fig. 1. In each condition, the heights of the feeder and oxygen injectors are adjusted by 5 ~ 10 cm.



Configuration / Height	Conf. I	Conf. II	Conf. III
A	High	Low	Medium
B	Low	Low	High

Fig. 1. Configurations of the melter during the DAW vitrification

Excessive oxygen amount: Oxygen is introduced into the melter through the oxygen injectors and the waste feeder for a complete oxidation of waste meterial. Stoichiometric oxygen concentration was calculated for each waste type using the elemental composition data shown in Table II. Effect of excessive oxygen amount was investigated at 50%, 70%, and 100% of excessive oxygen conditions.

Analysis of Dust Generation Characteristics

In order to evaluate the dust amount, dust concentration was measured at an outlet of the CCM according to the Korean Standard Sampling and Analysis Methods. This method is to collect particulates at a filter in the sampling equipment by the isokinetic off-gas sampling. The dust collected at the bottom drum of the HTF was also used for the analysis of the dust generation characteristics. As the air pulse clear away clogged dust in the HTF at a constant frequency (3 min), mass of the collected dust can give information about the dust generation amount. Some of the dusts drawn to the OGTS can be deposited onto the internal surface of the Pipe Cooler and some can be escaped from the HTF. However, the acculumated amount in the Pipe Cooler was not so significant compared with the HTF dust amount. In addition, it is very difficult to evaluate the accumulated amount in the Pipe Cooler during the operation. Dust passing through the HTF was ignored because the HTF has 99.99 % of removal efficiency for 1-micrometer particle.

The weight of the dust collected in the HTF drum was measured with a time interval of about one-hour. And it was normalized for each condition in order to exclude the effects of waste feeding rate and feeding time. That is, total dust amount was divided by waste feeding time and feeding rate. It was called here as "dust generation ratio" of which unit is 'g/kg'. It implies the ratio of the generated dust weight to the supplied waste weight.

Dust size distributions were measured with a Cascade Impactor equipped with ten impactor stages. Sampling method was identical to the dust comcentration measuring method.

RESULTS AND DISCUSSION

Dust Generation Characteristics of PE waste

Pilot plant vitrification tests for PE waste were carried out for six operating conditions by varying the three parameters; CCM configuration, waste feeding rate, and excessive oxygen amount (Table III). Glass temperature was maintained at approximately 1200 ~ 1220 ℃ while feeding the waste.

Table III. Operation parameters for PE vitrification

Parameter \ Condition	Cond. 1	Cond. 2	Cond. 3	Cond. 4	Cond. 5	Cond. 6
Waste feeding rate (kg/h)	5	10			15	20
Excessive oxygen (%)	70		100			
CCM configuration	Conf. I			Conf. III		

Fig. 2 shows the dust generation characteristics for six operating conditions. A remarkable difference was found in the dust generation ratios between two groups;

condition 1/2/3 and condition 4/5/6. The first group, which was operated at melter's configuration type I, produced about 10 times more dust than the second group operated at configuration III. It implies that melter 's configuration is the most dominant parameter to influence the vitrification condition inside the melter. Sampling results of the dust concentration in off-gas showed a big deviation even at one condition. However their trend was similar to that of dust generatio ratios.

Melter configuration I was equipped with the high waste feeder and the low oxygen injectors than those of configuration III. The reason why the configuration I condition generates more dust is judged that shredded PE with a low bulk-density is more easily blown off by the flow caused by oxygen injection flow and extraction fan force. That is, the effects of off-gas flow and oxygen flow on waste dispersion may be more vehement in configuration I because the distance between the injector outlet and accumulated waste surface is shorter than that of configuration III. Therefore, it may be difficult for wastes to be stably accumulated on the molten glass surface and to have a sufficient retention time for complete oxidation in the configuration I condition. Another evidence to support this explanation can be found from the results of Cond. 2 and 3 in Fig. 2. These two conditions have the same melter's configuration and the waste feeding rate (10 kg/h) except the amount of oxygen supply. As the excess oxygen flow increased from 70 % (Cond. 2) to 100 % (Cond. 3), more unburned particles were collected at the HTF. It may suggest that oxygen flow induce some disturbance in the combustion process inside the CCM in the configuration I.

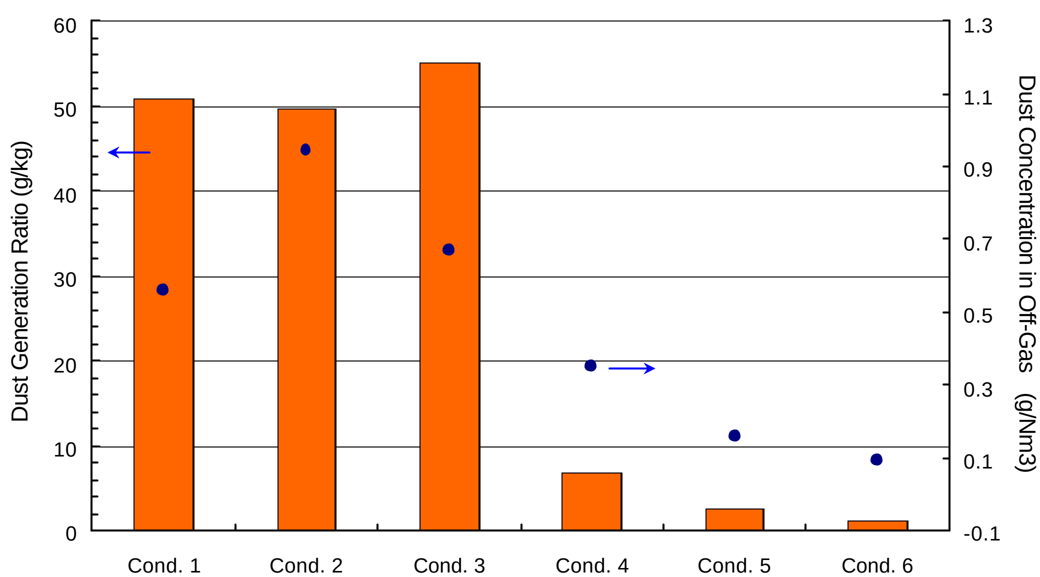


Fig. 2. Dust generation ratios (bar) and average dust concentrations (●) in off-gas on the operating conditions for PE waste vitrification

In the same CCM configuration I, significant differences in the dust generation ratios were not observed by changing waste feeding rate (Cond. 1, 2 in Fig. 2). On the other hand, as shown in Cond. 4/5/6 in the Fig. 2, the dust generation ratios decreased with the increase of waste feeding rate in the configuration III.

Various reactions may occur inside the melter. The surface of the accumulated waste pile will be exposed to the more oxidative environment and the interface between the waste and molten glass will form an oxygen-deficient condition. Hence, the main reaction is supposed to be the oxidation reaction on the surface of waste pile and pyrolysis or oxidative pyrolysis on the glass surface (3). Most products from pyrolysis and oxidative pyrolysis are products of incomplete combustion (PICs). Therefore, most particulates are supposed to be produced from the interface of waste and glass if fed waste is settled down on the glass surface and not dispersed by any flow. In the melter configuration III, the depth of the waste pile will increase as the waste feeding rate increases. This phenomenon may reduce the amount of PICs escaped to the OGTS through the upper cold layer of the accumulated wastes.

Consequently, it is thought that the fed waste is stably accumulated on the glass surface as the waste feed rate increases, and then, the thick layer of accumulated waste blocks escape of the particulates to the OGTS penerating through the layer. Mason in VECTRA Technologies, Inc. reported that deep waste bed on top of the glass functioned as a cold-cap to efficiently remove particulate and condense volatile radionuclides and heavy metals by reincorporating them into the glass (5).

Results in Fig. 2 imply that large amount of waste can be treated with less dust generation by optimizing the melter configuration. Similar trends were observed when treating the other waste types.

It was also observed during the operation that the dust generation ratios were decreased with the increasing of the waste feeding time even at the same condition. Off-gas sampling data showed the similar phenomenon. It indicates that vitrification conditions were not stabilized within 2~3 hours after changing the operating parameters. The results shown in Fig. 2 may involve the effect of the operation time. In order to examine the effects of the operating variables, off-gas sampling data on the combustion process should be obtained in the stabilized conditions.

Carbon contents in the HTF ashes generated during PE and cellulose treatments were analyzed. The results showed that carbon content in the dust is proportional to the dust generation ratio. It means that the dust generation ratio can be used as a parameter indicating the combustion efficiency. If carbon contained in the waste is not converted completely into gases due to incomplete combustion conditions, ashes containing carbon element will increase.

Flyash typically contains 5 ~ 15 % of carbon by soot and condensible hydrocarbon contents (6). Although this small amount of carbon will not normally support a flame, high carbon contents (over 70 %) analyzed in the HTF ash will be capable of smoldering when ignited by a hot ember. If this happens, the resulting high temperature could damage the OGTS. Therefore, carbon content should be controlled for safe operation of the OGTS.

Dust Generation Characteristics on Waste Types

Fig. 3 shows the dust generation ratios with waste feeding rates for PE, cellulose, and mixed waste. All tests were carried out with 100 % of excessive oxygen. The melter was operated with the configuration III for PE and mixed waste and with the configuration II for cellulose waste.

A similar trend was observed on the relationship between dust generation ratio and waste feeding rate irrespective of waste types: dust generation ratio decreased as the waste feeding rate increased

However, there was a significant difference depending on the waste type. The dust amount was much larger when treating the mixed waste than in the case of PE and cellulose. Mixed waste contains 20 % of PVC and 10 % of rubber. It was experimentally measured that ash contents were 6.8 % for PVC and 2.4 % for rubber, while those of PE and cellulose were 0.3 ~ 0.5 % (4, 7). And the major components of ash were determined to be specific additives, such as plasticizer, filler, and pigment. They are added during the manufacturing process for easy handling of raw material or providing specific properties of the final product. Elements composing the HTF ash were analyzed to be B, Na, C, and metal/inorganic components of waste ash. Therefore, high ash contents of PVC and rubber are assumed to be one of reasons causing high dust generation ratio.

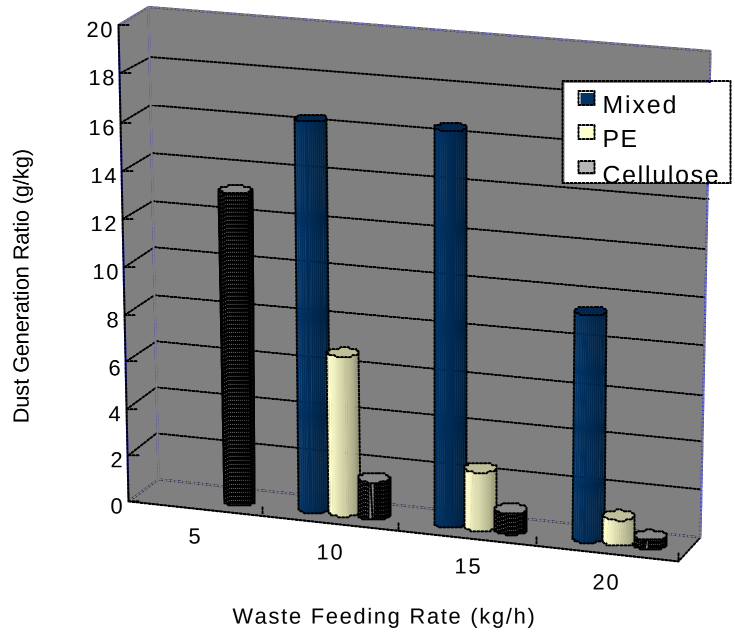


Fig. 3. Dust generation ratios for three waste types on the waste feeding rates

In addition, glass temperature variation is suspected to contribute to the high dust generation ratio. It was observed in the previous tests that a sharp change in the glass temperature resulted in an instant increment of CO concentration and off-gas flowrate. During the cellulose tests, glass temperature was maintained almost constantly (about 1200 ℃), while the temperature variations were 60 ~ 80℃ and 90 ~ 180℃ for PE and mixed waste, respectively. The dust generation ratio of each waste type tends to be increased with the temperature change of molten glass.

Shredded mixed waste was not homogeneously mixed but conglomerated. When it is fed into the melter, the heat generated by the combustion reactions will not be maintained constant because the combustion enthalpy for each material is different (cellulose: 16.1 kJ/kg, PVC: 17.5 kJ/kg, PE: 46.3 kJ/kg). Then, such a change in the combustion heat may also induce an instant change in the glass temperature. This may cause a larger temperature variation in the treatment of mixed waste than the homogeneous waste.

Dust Generation Characteristics of Mixed Waste

Dust generation ratio during the mixed waste treatment was investigated at the feed rate of 20 kg/h by varying the excessive oxygen amount. Dust generation ratio was inversly propertional to the excessive oxygen amount (Fig. 4).

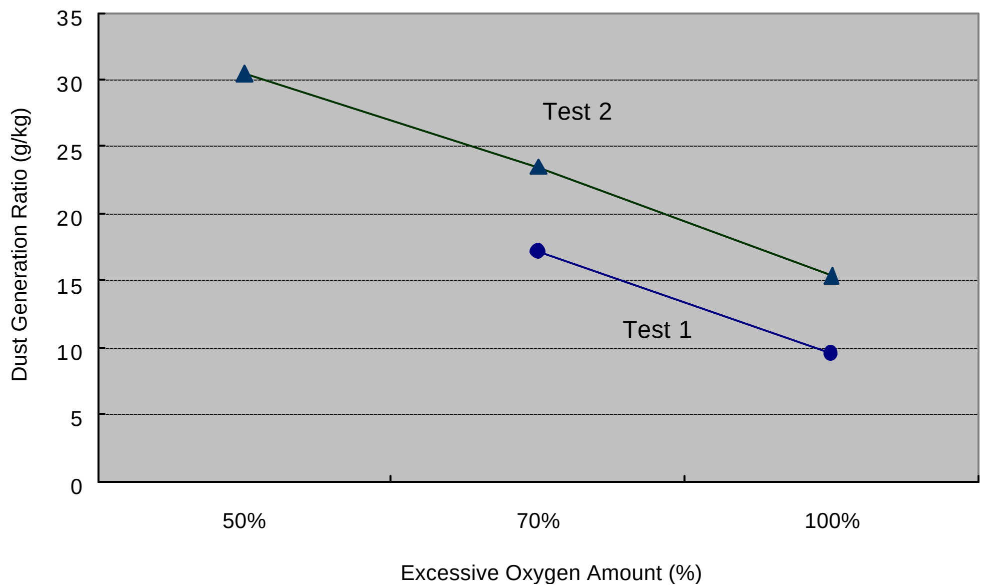


Fig. 4. Dust generation ratios on the excessive oxygen amounts for the mixed DAW vitrification tests

The results from two tests showed almost similar decrease tendency according to the excessive oxygen amount. However, absolute values showed some difference. In the Test 1, glass bath temperature was maintained stable from 1150℃ to 1200℃ compared to the range of 1090 ~ 1250℃ in the Test 2. In general, proper oxygen amount and temperature controls are essential for suppressing the generation of PICs during thermal treatment. Ultimately, it is expected that temperature variation influenced on the dust generation ratios.

Dust size distributions were measured during the Test 2 with the excessive oxygen amounts (50, 70, 100 %). The results showed that the amount of large particles (greater than 5.6 microns) increased with the decrease in the excess oxygen amount. It is thought that partially oxidized or pyrolyzed products were abundantly generated in the oxygen deficient condition.

CONCLUSION

The dust characteristics generated from the melter were analyzed to derive the optimum conditions for the vitrification of the DAWs by varying the three operating parameters; melter configuration (configuration I, II, III), exc essive oxygen amount (50%, 70%, 100%), and waste feeding rates (5, 10, 15, 20 kg/h).

The test results showed that the dust generation characteristics have a close relationship with the operation variables. Among the tested parameters, the melter configuration was found as the dominant one to affect the DAW vitrification process. Dust generation ratio was minimized in the condition of melter configuration II or III and 20 kg/h of waste feeding rate for all waste types. It implies that large amount of waste can be treated with less dust generation. There was a significant difference in the dust generation ratio depending on the waste type. The dust amounts were increased when treating the mixed wastes compared to the PE and the cellulose. The dust generation ratio was inversly proportional to the excessive oxygen amount.

Since other parameters such as temperature control and operation time have also affected the waste vitrification process, tests to optimize these parameters and to maintain the steady combustion condition should be performed in the future.

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