TEST OPERATION OF OXYGEN-ENRICHED INCINERATOR FOR WASTES FROM NUCLEAR FUEL FABRICATION FACILITY

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ABSTARCT

The oxygen-enriched combustion concept, which can minimize off-gas production, has been applied to the incineration of combustible uranium-containing wastes from a nuclear fuel fabrication facility. A simulation for oxygen combustion shows the off-gas production can be reduced by a factor of 6.7 theoretically, compared with conventional air combustion. The laboratory-scale oxygen enriched incineration (OEI) process with a thermal capacity of 350 MJ/h is composed of an oxygen feeding and control system, a combustion chamber, a quencher, a ceramic filter, an induced draft fan, a condenser, a stack, an off-gas recycle path, and a measurement and control system. Test burning with cleaning paper and office paper in this OEI process shows that the thermal capacity is about 320 MJ/h, 90 % of design value and the off-gas reduces by a factor of 3.5, compared with air combustion. The CO concentration for oxygen combustion is lower than that of air combustion, while the O_2 concentration in off-gas is kept above 25 vol % for a simple incineration process without any grate. The NOx concentration in an off-gas stream does not reduce significantly due to air incoming by leakage, and the volume and weight reduction factors are not changed significantly, which suggests a need for an improvement in sealing.

INTRODUCTION

Since IAEA reported that incineration was a convenient means of volume reduction for the management of radioactive wastes produced by radioisotope users in 1965 (1), incineration has been an important technology in radioactive waste treatment. By incineration, waste volume and weight are reduced by factors of 100 and 10, respectively, for combustible solid wastes (2). Incineration also yields residues containing concentrated radionuclides that often are more compatible with subsequent management steps than the original waste form, and has the advantage of versatility to accept and process a wide range of materials including liquid, solid, and semi-solid wastes.

As public concern over incinerator emissions increases, incineration is facing a difficult situation. Incineration has the potential risk to spread contaminants, particulates, radionuclides, and dioxins, and the emission requirement of incineration has been enhanced much more than before. To overcome these problems, alternative incineration technologies, such as thermal (plasma, DC-arc, vitrifier, melter, supercritical water, steam reformer), separation (solvent extraction), chemical oxidation (acid digestion), dehalogenation (solvated electron), and biological methods, have been developed recently (3). All these alternatives have less off-gas than incineration and less potential for hazardous air emissions. None of the alternative technologies alone has, however, the capability of thermal systems to treat the large variety of wastes from nuclear facilities up to now. Alternative non-flame technologies have particular difficulty in treating organically contaminated inorganic matrices such as soils, inorganic sludges and debris (4).

Another method to solve these problems is to decrease emissions and improve off-gas treatment systems, which costs more than other technologies. One good method to minimize the off-gas for the thermal process is the use of oxygen or oxygen-enriched air instead of air as an oxidant. The use of oxygen or oxygen-enriched air leads, in addition, to a higher oxygen partial pressure and a higher combustion temperature, giving a throughput increase, a thermal efficiency improvement, and fuel savings (5-7). Oxygen enrichment means just replacement of air (more exactly nitrogen) with oxygen, resulting in an increase of available oxygen concentration higher than 21 %. This oxygen enrichment technology is now widely used in chemical oxidation and combustion processes in the steel, cement, glass, refining, etc.
Recent applications of oxygen enrichment on incineration have mainly focused on the performance improvement for air-combustion systems by an oxygen injection into the burner air supply (combustion air enrichment) (8) or a direct oxygen supply to a firing grate through a lancing pipe (oxygen lancing) (9). These uses of oxygen enrichment are limited to below 30 % oxygen, not giving a significant reduction in NOx formation, because of a temperature limitation of system components. The use of pure oxygen as an oxidant minimizes the off-gas production, but gives some problems such as higher NOx formation and local overheating due to a higher flame temperature and decrease of gas momentum to give better mixing in the combustion chamber. A higher NOx formation could be excluded by having no air leakage into the incineration chamber from outside. The hot spot formation and loss of gas mixing momentum could be, however, minimized by only the addition of inert gases or diluents.
This paper deals with our trials to minimize the overheating problem by the recycling of a part of off-gas to the combustion chamber; simulation for oxygen combustion with different recycle ratios of off-gas, test operation of laboratory-scale oxygen incineration (OEI) process with a recycle path, and finding the solutions for a stable and effective operation.

URANIUM-CONTAINING WASTES

Uranium-containing solid wastes are generated from two 400-MTU/yr nuclear fuel fabrication facilities at the Korea Nuclear Fuel Company (KNFC), one for a light water reactor and the other for a CANDU reactor. Radioactive materials, concentrated (max. 5 wt %) uranium or natural uranium, exist in wastes as uranium compounds such as UF_6, UO_2F_2, UO_2, U_3O_8, AUC and UO_4. All solid wastes are categorized as metals, concretes, spent filters, lime precipitates, synthetics, and miscellaneous wastes, and packaged in 200-liter steel drum. Synthetics (PE or PVC sheets, rubber or plastic pipes, etc.) and miscellaneous wastes (cleaning fabrics, Tyvek masks, textile clothes, shoes, latex gloves, etc.) are considered as combustibles, and their average activities are 40 and 90 MBq/drum, respectively (10). An average annual generation of combustibles is about 100 drums, and its approximate composition is 35 % cleaning fabrics, 30 % clothes, 20 % Tyvek masks, 10 % PVC sheets, and 5 % shoes. The results of the proximate and ultimate analyses and the calorific values for the above representative samples are shown in Table I. Most combustibles were mainly composed of volatile materials with little ash, and their average calorific value is about 36.5 MJ/kg.

Table I. The proximate and ultimate analyses and the calorific values for waste materials

Materials	Proximate analysis				Ultimate analysis							Calorific value (MJ/kg)
	Moisture	Volatile matter	Fixed carbon	Ash	C	H	O	N	S	Cl	Etc.	
Cleaning fabrics	0.06	0.93	0.00	0.01	0.78	0.13	0.08	0.00	0.00	0.00	0.01	45.1
Clothes	0.10	0.80	0.09	0.01	0.49	0.07	0.40	0.03	0.00	0.00	0.01	18.7
Tyvek mask	0.10	0.89	0.00	0.01	0.81	0.13	0.05	0.00	0.00	0.00	0.01	45.6
PVC sheet	0.04	0.95	0.00	0.01	0.51	0.08	0.06	0.00	0.00	0.34	0.01	45.4
Shoes	0.04	0.76	0.17	0.03	0.71	0.12	0.10	0.00	0.02	0.02	0.03	29.6
Average	0.077	0.877	0.035	0.011	0.07	0.11	0.17	0.01	0.001	0.04	0.01	36.5

SIMULATION FOR OEI WITH U-CONTAINED WASTES

Nitrogen in air is not needed in the combustion process and contributes to the formation of NOx. A significant amount of available heat of combustion is used for heating of the nitrogen, an inert diluent. When a part of the combustion air is replaced by oxygen-enriched air or oxygen, the content of nitrogen would reduce in both volume and partial pressure. Such reduction of nitrogen gives a reduction of the overall volume of off-gas per unit of waste incinerated, and finally affects the reduction of particulate carryover.

Another effect of OEI is a change in the composition of combustion gas. A combustion reaction under oxygen produces the largest portion of water, and then carbon dioxide, while producing in order nitrogen, water, and carbon dioxide, under air combustion. This composition change is caused by an elimination of inert nitrogen, but the total amounts of water or carbon dioxide does not change. If even water were removed from the off-gas stream by an appropriate unit process, the carbon dioxide alone would remain in the off-gas stream.

The maximum flame temperature of methane under air is theoretically about 1900^{o}C, whereas it is about 2800^{o}C under oxygen, because the combustion heat does not consume to increase the temperature of inert nitrogen up to the combustion reaction temperature. Such a high temperature flame sometimes leads to hot spot formation at the point of direct contact with flame, and such a high temperature condition promotes NOx formation.

To overcome these problems coming from the temperature increase due to lack of inert diluent, we added a recycle path to send a part of off-gas composed mainly of carbon dioxide to the combustion chamber when using oxygen as an oxidant and subsequently remove water from the off-gas stream, into a combustion chamber. Such off-gas recycling slightly increases the off-gas volume discharged into the environment, but at the same time a much larger amount of off-gas than nitrogen when using air can be added to the combustion chamber.

Simulation to estimate the off-gas volume and concentration was performed under some assumptions as follows:

- Waste: combustible wastes from nuclear fuel fabrication facility
- No air incoming by leak
- Excess O_2 concentration in off-gas stream: above 10 vol %
- Burner is off at steady state operation
- Off-gas recycling to combustion chamber
- Water removal before off-gas discharge: condenser temperature of 50^{o}C
- Oxygen-enriched combustion process: Fig. 1

Simulation results for the oxygen incineration with different recycle ratios, compared with the result of conventional air combustion with an excess air ratio of 2, were shown in Table II, where an excess oxygen (or air) ratio, m is defined as a ratio of actual oxygen (or air) to stoichiometric oxygen (or air), and a recycle ratio, r is defined as a recycled off-gas volume divided by a total combustion off-gas volume. Excess O_2 concentration in off-gas is kept above 10 vol %, because, in Korea, most incineration facilities discharge off-gas with an excess O_2 concentration of above 10 vol % and the concentrations of air pollution materials are normalized and regulated on the basis of standard O_2 concentration, 12 vol %.

By volume, air is made up of about 79 % nitrogen and 21 % oxygen, which means a large amount of nitrogen is supplied in air combustion. Actually, the amount of nitrogen supplied in air combustion with a single chamber and an excess air ratio of 2 is about 11.82 Nm^3/kg-waste, occupying about 75 % of the total off-gas discharged from the stack. The same amount of off-gas as that of nitrogen for air combustion is supplied in oxygen incineration when the recycle ratio, r is 0.85. The off-gas discharged from the stack is, however, reduced by a factor of 6.7, compared with air combustion under the same condition of excess O_2 concentration in off-gas. All concentrations of air pollution materials such as CO_2, SO_2, and HCl increase about 6 times as high as those of air combustion due to the reduction of total off-gas volume.

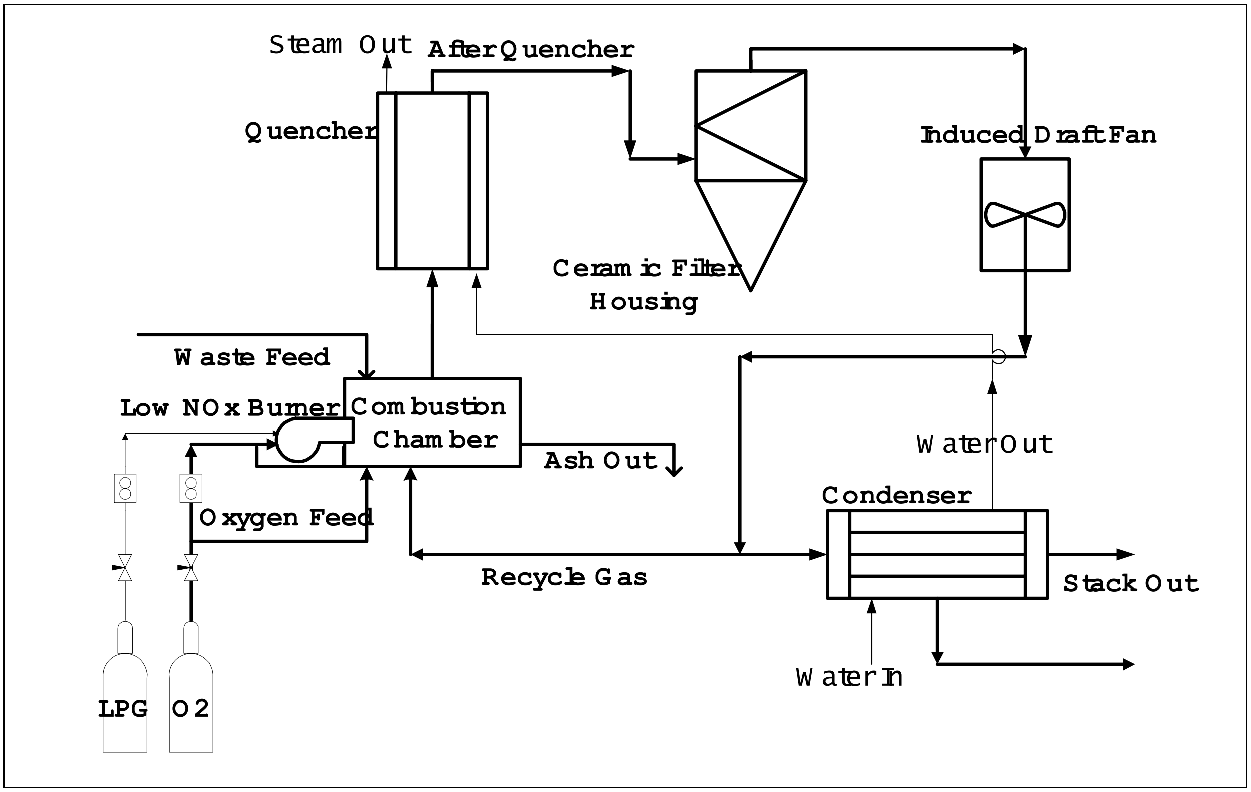

Fig. 1. Oxygen-enriched combustion process for simulation

Table II. Simulation results for oxygen combustion with different recycle ratios

	Oxygen combustion, m=1.2[a]				Air combustion (single chamber), m=2.0
	r = 0.5[b]	r = 0.7	r = 0.8	r = 0.9	
O_2 conc., vol.%	17.0	15.0	13.0	11.9	9.9
Off-gas discharged, Nm^3/kg-waste	1.87	2.09	2.24	2.48	15.86
Ratio	0.12	0.13	0.14	0.16	1

[a] m: excess oxygen (or air) ratio = actual oxygen (or air) / stoichiometric oxygen (or air)
[b] r: cycle ratio = recycled off-gas volume / total combustion off-gas volume

LABORATORY-SCALE OEI PROCESS

Process Description

The laboratory-scale OEI process consists of an oxygen feeding and control system, a combustion chamber, a quencher, a ceramic filter, an induced draft fan, a condenser, a stack, an off-gas recycle path, and a measurement and control system. This process has a capacity of 10 kg/h (thermal capacity of 350 MJ/h) on the basis of combustible wastes from nuclear fuel fabrication facilities. All unit processes except oxygen feeding and control system were made of stainless steel. After liquid oxygen is vaporized, oxygen gas is sent to the oxygen-feeding tank, and finally fed to the chamber through an aperture within refractory. The combustion chamber, a cylindrical type positioned horizontally, has a double gate for waste feeding, a low NOx oxygen burner, an ash removal gate, and three oxygen-feeding tanks on the surface of the chamber. The combustion gas output path comes in contact with the quencher, a water-evaporating heat exchanger. The 3x3 units filter cartridges are equipped within the filter housing. The induced draft fan, operating in connection with pressure of combustion chamber, is used for draft of off-gas and recycling a part of off-gas to combustion chamber. The condenser, a water-cooled heat exchanger, eliminates water in off-gas discharged at stack. This process is actually only for the development and testing of the OEI process; therefore, other off-gas treatment systems, such as an HEPA filter, wet scrubber, etc., were not used in this step.

Experimental procedure

Test operation of the OEI process was performed with two paper wastes: cleaning paper and office paper composed of spent Xerox paper or books. Thermal properties of waste materials were analyzed and listed in Table III.

Paper wastes were packaged in about 1 kg/package and put into paper bag to be fed easily into the incineration chamber through the double gate. The test operation procedure is as follows: first, oxygen and liquefied petroleum gas (LPG) are fed to the oxygen burner, and then the burner is started. When the combustion chamber is preheated to 450 ^{O}C using the oxygen burner alone, the oxygen and the paper waste packages are fed into the chamber, and then the oxygen combustion is started. Waste packages are fed in continuously until the combustion temperature reaches a desired temperature, namely, the "combustion temperature". Once the combustion temperature reaches a desired temperature, the waste is fed in at every fixed temperature to maintain a constant combustion temperature. Air combustion is also performed according to the same procedure as oxygen combustion, except for the feeding of compressed air instead of oxygen. Detailed operation conditions are shown in Table IV.

The off-gas stream passes through the quencher, where the temperature of the off-gas decreases to below 170 ^{O}C, and then goes into a ceramic filter housing. After the off-gas stream removed particulates passes through the induced draft fan, the off-gas stream is divided into two: one for recycling to the combustion chamber and the other for discharge through a condenser and then the stack.

The temperatures at the combustion chamber, chamber outlet, and quencher outlet are recorded every two minutes during the test operation. A hot wire anemometer (V-02-AD500, Denshi Co., Ltd) is used for the flow rate of off-gas flowing inside the unit process, and a stack gas analyzer (TESTO-350, Testo GmbH & Co.) is used for analysis of the off-gas discharged from the stack.

Table III. The proximate and ultimate analyses and the calorific values for paper waste materials

Materials	Proximate analysis				Ultimate analysis							Calorific value (MJ/kg)
	Moisture	Volatile matter	Fixed carbon	Ash	C	H	O	N	S	Cl	Etc.	
Office paper	0.083	0.809	0.060	0.007	0.410	0.059	0.522	0.000	0.000	0.001	0.008	15.6
Cleaning paper	0.061	0.733	0.101	0.146	0.324	0.051	0.468	0.001	0.000	0.001	0.155	13.5

Table IV. Operation conditions for test run

	O4032	O4033	O4034	O4101	O4102	O4262	O4263	O4264	O4265	A4133	A4134
Oxidant	Oxygen									air	
Oxidant flow rate, m^3/h	25	15	10	20	20	20	15	12	10	170	170
Waste material	cleaning paper			Office paper							
Set point of feeding, ^{O}C	800			850				900		850	
Burner operation	OFF			ON	OFF						
Chamber press., mmH_2O	-18			-16		-7	-8	-12	-12	-8	
Recycle ratio	0.5		0.7	0.5	0.7	0.5	0.90	0.90	0.90	0	
Operation time, min	30	15	20	40	60	35	5	15	15	35	30

Results and Discussion

The typical temperature profiles for oxygen combustion of cleaning paper (2a) and office paper (2b) are shown in Fig. 2, and the experimental results are summarized in Table V.

Table V. Experimental results from test operation

		O4032	O4033	O4034	O4101	O4102	O4262	O4263	O4264	O4265	A4133	A4134
Av. temp., ^{O}C	Chamber	920			1,035		996		1017		968	
	Ch. Outlet	825			882		890		909		877	
	Qu. Outlet	136			151		157		149		224	
Inc. capacity	kg/h	24	20	15	18	24	26	24	26	26	26	33
	MJ/h	380	310	230	240	320	350	320	350	350	350	440
Av. vol. reduction factor		50			27		NM[a]				20	
Av. wt. reduction factor		11			8		NM				8	
Flow rate, Nm^3/h	Recycle	32	30	31	29	31	24	33	32	32	0	0
	Discharge	NM	NM	NM	33	48	37	13	8	6	146	230
Off-gas concentration	O_2, %	37.3	27.5	20.5	26.7	25.7	22.8	21.0	18.2	18.3	10.9	14.2
	CO, ppm	NM	40	40	120	70	8,400	5,000	2,600	6,000	2,700	170
	SO_2, ppm	NM	6	4	180	40	90	120	80	100	130	80
	NOx, ppm	80	70	50	170	130	180	140	100	120	70	40
Discharged off-gas per unit waste, Nm^3/kg-waste		NM	NM	NM	1.7	2.0	1.4	0.5	0.3	0.2	5.6	7.0

[a] NM : Not measured

Incineration capability

This oxygen combustion process was originally designed for incineration of combustible wastes, whose calorific value is about 2.5 times as high as that of paper wastes. Our first concern is how much paper

waste is burnt out in this process. When office paper was burnt out at a set point of combustion temperature of 850 ^{O}C, the incineration throughput was about 25 kg/h (thermal capacity of about 340 MJ/h), showing about 2.5 times as large a design value, but almost the same value in thermal capacity.
Incineration capacity increased linearly with oxygen feed rate when cleaning paper was burnt out at a combustion temperature of 800 ^{O}C. However, the oxygen feed rate seemed not to affect the combustion of office paper during our short operation of about 30 minutes at a combustion temperature of 900 ^{O}C. The waste feeding method, feeding at every moment that the temperature of the chamber outlet reached the set point and keeping a constant combustion temperature, might have some problems like an oversupply of paper waste with a low calorific value, to reach and keep at such a high temperature condition. Such unstable operation by the oversupply of waste is realized by very high CO concentrations in off-gas.

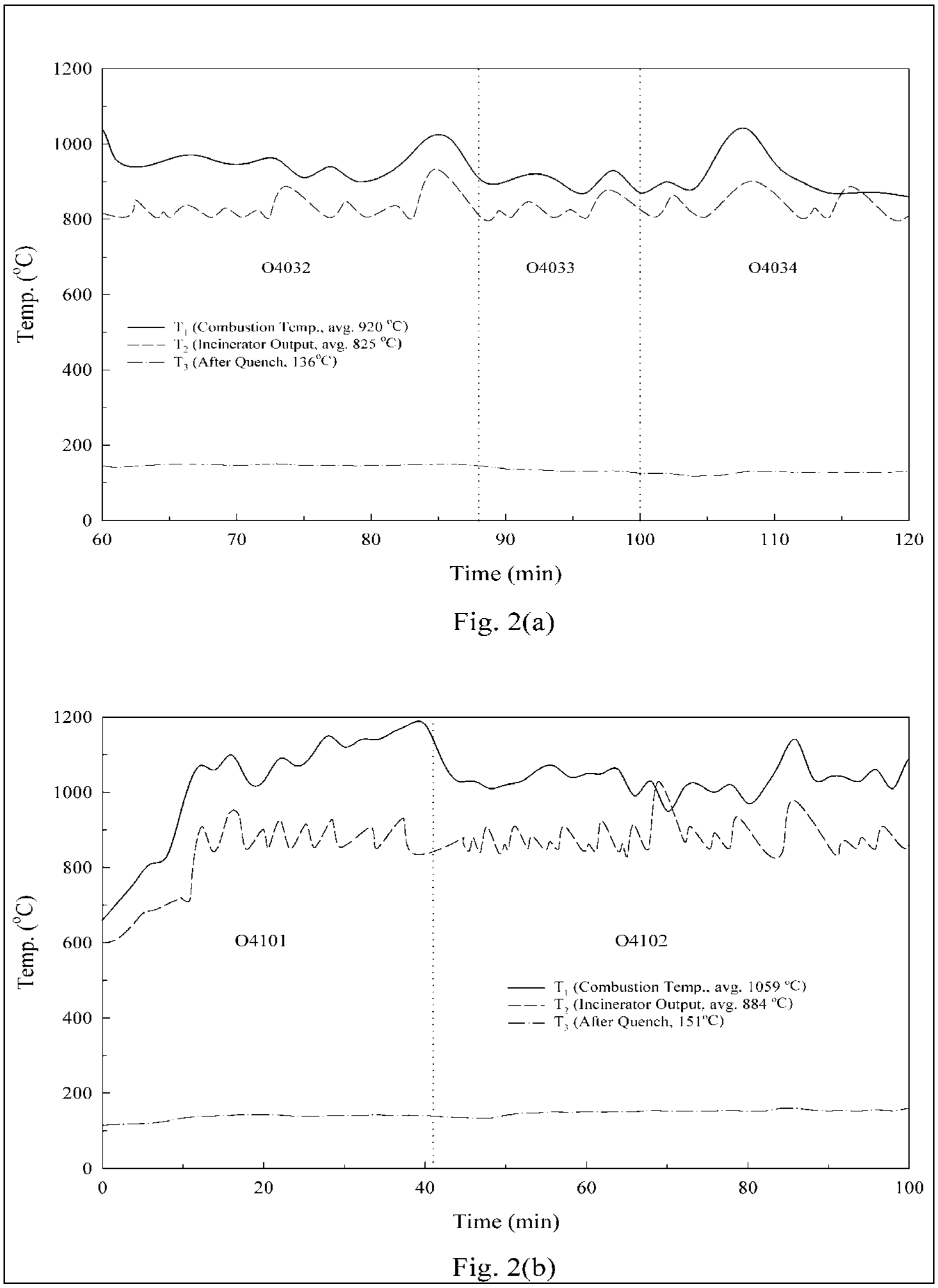

Fig. 2. Temperature profiles for oxygen incineration of cleaning paper (a) and office paper (b)

Off-gas production

During the stable oxygen combustion of office paper, considered a low CO concentration in off-gas, off-gas of about 1.7 Nm^3/kg-waste was produced when an oxygen burner was run and about 2.0 Nm^3/kg-waste when the oxygen burner was not run. On the other hand, in air combustion with a similar condition, the amount of off-gas was 7.0 Nm^3/kg-waste. Because off-gas of only 0.9 Nm^3/kg-waste was expected to be produced from the simulation for oxygen combustion of office paper, almost the same rate of air leakage as the oxygen feed rate seemed to infiltrate into the combustion chamber.

Oxygen concentration in off-gas

The oxygen for the combustion reaction was supplied on the basis of the simulation result above 10 vol % of O_2 concentration in off-gas. The actual oxygen concentrations in off-gas for oxygen combustion were about 10 vol % higher than those from other simulations for oxygen combustion with the same capacity of paper waste, which meant that the incineration capacity, calculated from total number of waste package fed only above the set point of the combustion temperature, was somewhat over estimated.

Carbon monoxide (CO) concentration in off-gas

So long as the O_2 concentration in off-gas was kept above 25 vol %, the CO concentration was much lower than that of air combustion. However, the CO concentration increased very much when the O_2 concentration in the off-gas was lower than 22 vol %, which suggests that such an oxygen combustion process without any grate to help the oxygen diffuse well needs to keep the O_2 concentration above 25 vol % to give good combustion.

Nitrogen oxide (NOx) concentration in off-gas

An advantage expected by using oxygen as an oxidant is a significant decrease in NOx formation because thermal NOx doses not occur during the combustion reaction due to the absence of nitrogen. During the test operation, the NOx concentration in the off-gas was actually 3 or 4 times higher than that of air combustion. The total generation considering off-gas volume was, however, a minimum of 20 % lower than that of air combustion, which meant that our OEI process was not effective in decreasing NOx formation. Such a result seems to mainly come from a failure in sealing for leakage, which suggests that an improvement should focus on sealing for all identified sources of leakage.

Volume and weight reduction ratios

Volume or weight reduction ratio, defined as the volume or weight of total waste fed into the combustion chamber divided by the volume or weight of ash, respectively, after combustion, were obtained only by the averaged values because ash removal from the chamber was performed by batch. Little change in the volume and weight reduction ratios was shown, compared with air combustion. This result was another proof of the need for the bottom grate to help the diffusion of oxygen and the oxygen supply. A simple replacement of air by oxygen does not give a good diffusion of oxygen; it only gives a reduction of off-gas volume.

CONCLUSION

A test operation of the oxygen combustion process with paper wastes has been performed in a laboratory-scale incinerator designed for application of uranium-containing wastes from nuclear fuel fabrication facilities. The oxygen combustion can reduce the off-gas production by a factor of 6.7 theoretically, compared with conventional air combustion. A laboratory-scale oxygen enriched incineration process with a thermal capacity of 350 MJ/h is composed of an oxygen feeding system, a combustion chamber with a low NOx oxygen burner and a double gate, quencher, ceramic filter, induced fan, condenser, stack, and off-gas recycle path to prevent over heating and to give sufficient gas momentum. Test operation with cleaning paper and office paper in this OEI process shows that the thermal capacity is about 320 MJ/h, 90 % of the design value and the off-gas reduces by a factor of 3.5, compared with air combustion. The CO concentration for oxygen combustion is lower than that of air combustion, so long as the O_2 concentration in the off-gas is kept above 25 vol % for a simple incineration process without any grate. The total amount of NOx does not reduce significantly due to air incoming by leakage, and the volume and weight reduction factors are not changed significantly, which suggests a need for an improvement in sealing.

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