PARTICLE SIZE DISTRIBUTION AND ENRICHMENT OF SELECTED METALS FROM WASTE SLUDGE COMBUSTION AT AIR AND OXY-FUEL COMBUSTION

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Abstract – Comparative tests in air and oxy-fuel combustion with oxygen injection rates ranging from 21% to 40% were conducted in a 30kW$_{th}$ circulating fluidized bed (CFB) pilot plant for waste sludge combustion. General combustion characteristics of the CFB such as pressure profiles, temperatures along the bed, and flue-gas composition, were different under the air and oxy-fuel conditions with various oxygen injection rates. Based on the results, the optimal oxygen injection rate was determined as 25% for the oxy-fuel combustion. In the bottom and fly ash, alkali and heavy metals had different distribution under the air and oxy-fuel combustion conditions. The particle size distribution in fly ash from air combustion was dominated by coarse particles over 2.5 μm in size, whereas with oxy-fuel combustion, most particles were submicron in size approximately 0.1 μm, and a smaller amount of coarse particles over 2.5 μm in size formed than with air combustion. Mass fractions of Al, Ca, and K below 2.5 μm in size found in the ashes from oxy-fuel combustion were higher than those in air combustion were. Submicron particle formation from the Cr, Ni, Cu, and Zn in the fly ash occurred more during oxy-fuel combustion than it did in air combustion.

INTRODUCTION

In recent years, the demand of renewable energy fuels has been increased in worldwide because the capacity of fossil fuel would be not affordable in the future. As one of renewable energy fuels, the production of sewage sludge would be gradually increased by year, and it would be over than 10milion tons in 2015 in Korea. Since ocean dumping was inhibited due to London Convention with being in effective at the end of 2012 in Korea, the combustion of sewage sludge has been emerged as one of alternative technologies of waste to energy. The sewage sludge combustion was oriented from paper and pulp industries. Paper and pulp plants in Europe generated industrial sewage sludge, and the fluidized bed combustion technology was developed to produce superheated steam by utilizing industrial sewage sludge. The fluidized bed combustion (FBC) has been considered as common technology for sewage sludge due to the fuel characteristics of sewage sludge composed lots of moisture and heterogeneous particle size. The capacity of the commercial FBC plant was ranged from 100 to 200 tons as sewage sludge combustion per day. Those FBC plants combusting sewage sludge utilized excess air as combustion air, and generated lots of carbon dioxide as one of green house gases (GHG). However, the issue of global warming has become considerable in worldwide, and the combustion of waste fuels such as sewage sludge, waste biomass, refused derived fuels, and etc. would become one of most attributed emission sources although the fossil fuel combustion has become main emission source. Due to this global warming, the carbon capture & storage (CCS) technology has been developing to reduce GHG such carbon dioxide from stationary emission sources. According to Metz, B. et al.(2005), there are different types of CO_2 capture technologies such as pre-combustion, post-combustion, and oxy-fuel combustion. Oxy-fuel combustion is in the demonstration phase and uses high purity oxygen. Buhre, B. J. P. et al.(2005) wrote that a combination of oxygen typically of greater than 95% purity and recycled flue gas is used for combustion of the fuel during oxy-fuel combustion. By recycling the flue gas, a gas consisting mainly of CO_2 is generated, ready for sequestration without stripping of the CO_2 from the gas stream. Therefore, oxy-fuel technology for new constructed and retrofitted sewage sludge FBC plant would be necessary to generated high purity CO_2 in flue-gas. In the earlier, pulverized coal power plant with oxy-fuel combustion was demonstrated by pilot test performance. Chen, L. et al.(2012) wrote that the earliest study of coal oxy-fuel combustion as pilot scale was conducted for the Argonne National Laboratory (ANL) by the Energy and Environmental Research Corporation (EERC) in their 3MW$_{th}$ pilot facility. This study indicated that oxy-fuel combustion would be capable of applying successfully as a retrofit to a wide range of utility boiler and furnace systems. According to Buhre, B. J. P. et al.(2005), a pilot scale study conducted by the International Flame Research Foundation (IFRF) was evaluated the combustion of pulverized coal in a mixture of O_2 and recycled flue gas with the primary consideration of retrofitting an existing boiler, while increasing CO_2 concentration to above 90% for CO_2 capture. Gotou, T. et al.(2011) wrote that a study by IHI cooperation demonstrated combustion characteristics such as a flame temperature as the function of O_2 concentration, NOx conversion rate, SOx emission, and etc. during oxy-fuel combustion by utilizing $1.2MW_{th}$ test furnace. Also, Buhre, B. J. P. et al.(2005) wrote that a study by Air Liquide and Babcock & Wilcox (B&W) company demonstrated the combustion process based on O_2 enriched flue gas recirculation to provide an easy-to-implement option for multi-pollutant control, including CO_2 capture suitable for retrofitting existing boiler. The test showed that the oxy-fuel combustion generated less NOx than air combustion, and reported that it achieved effective removal of SOx with wet flue gas desulfurization (FGD) equipment and significant reduction of mercury emission. According to Jia, L et al.(2011), the CANMET organization has a long history in experimental results and modeling of the oxy fuel technology. In $0.3MW_{th}$ capacity pilot-scale combustor, the coal combustion behavior in various mixtures of oxygen and CO_2 were studied to demonstrate the effects of several factors on combustion performance including oxygen concentration, recycled ratio, burner performance, and etc. Also, the CANMET organization designed a 0.8MWth pilot plant for the demonstration and evaluation of oxy-fuel CFB process. Feedstock utilized in the tests includes a pet coke, a bituminous coal and a sub-bituminous coal. Along with the pulverized coal power plant, the fluidized bed coal power plant with oxy-fuel combustion has been demonstrated by pilot test performance. Tourunen et al.(2011) wrote that the VTT Technical Research Centre conducted 0.1MWth pilot test under air and oxy-fuel combustion by using seven different fuels including anthracite coal, bituminous coal, lignite coal, and 2 different types of biomass(wood pellet and straw pellet). The selected fuels and fuel blends were tested under air and oxy-fuel combustion. Flue gas emissions were measured and ash samples were collected in different test to evaluate combustion efficiency and emission performance. The main flue gas emission such as CO_2, CO, NO and SO_2 were measured during all tests. Also, there were several on-going demonstration projects for oxy-fuel combustion by several institutes in the worldwide. However, those all pilot and demonstration projects were mostly conducted using coal fossil fuel to demonstrate oxy-fuel combustion of coal-fired power plant. In this study, the concepts of oxy-fuel combustion using CFB combustion technology as a WTE technology and carbon capture have been applied to waste sewage sludge. Combustion characteristics were observed in air and oxy-fuel combustion. In addition, the comparative study of the particle size distributions enriched with metal elements and the behavior of alkali and heavy metals from waste sludge combustion at air and oxy-fuel conditions was conducted using $30kW_{th}$ CFB pilot test bed.

2. Facilities and Experimental Method

2.1 Test facility

Figure 1 shows a schematic diagram of test facility of the 30 kWth CFB oxy-fuel pilot test bed. The pilot test was conducted in the CFB combustion system consisting of a riser, a cyclone, a down-comer, and a loop-seal. The CFB combustor has a riser with an inner diameter of 0.15 m and a height of 6.4 m. The CFB combustor was surrounded by heating materials in order to deliver the combustion heat to the waste sewage sludge during the air and oxy-fuel combustion. The optimum temperature for the waste sewage sludge combustion was determined to be 800℃. The feeding rate of the waste sewage sludge was optimized at 13 kg/hr. The experimental tests were carried out in the 30 kWth CFB combustor operating with air and oxy-fuel conditions. The oxygen injection rate in oxy-fuel conditions was 23 %, which was fed into the riser. Once test bed was initiated, test operation was maintained for 3 hours per day with constant sludge feeding rate, oxygen injection rate, and solid circulating rate to verify stable combustion conditions. Series of tests in air and oxy-fuel conditions were conducted in a week. Those tests in air and oxy-fuel conditions with 23 % of oxygen injection rate were repeated in three times for three weeks. Constant operation conditions and stability of CFB system were constantly observed by data acquisition system and continuous analyzing systems. All data were measured during each test period.

2.2 Fuel characteristics

Table 1 shows the basic characteristics of the waste sewage sludge. In the proximate analysis, the volatile fraction in the waste sewage sludge was 45.11%. The fraction of ash in the sludge was 35.04%. The calorific values of the sludge and coal were 3,008 kcal/kg. The sulfur fractions in the sludge and coal were 0.43% and 0.58%, respectively. The theoretical oxygen demand based on the elemental analysis of the sludge was calculated to be 835 L/min when a feeding rate of 13kg/h of waste sewage sludge was used in the 30 kW_{th} CFB oxy-fuel pilot test bed. Based on the theoretical oxygen demand, the input flow rate for the air and oxy-fuel mixtures was set at 900 L/min for this study.

2.3 Sampling and analysis

The heating value of sludge and biomass fuel was analyzed by AC-350 calorimeter from LECO Corp. USA. The proximate analysis was conducted by TGA-601 from LECO Corp. USA. The elemental analysis was conducted by 2400 series Ⅱ CHNS/O analyzer from Perkin Elmer Inc. The gaseous analysis in flue-gas was conducted by the portable gas analyzer.

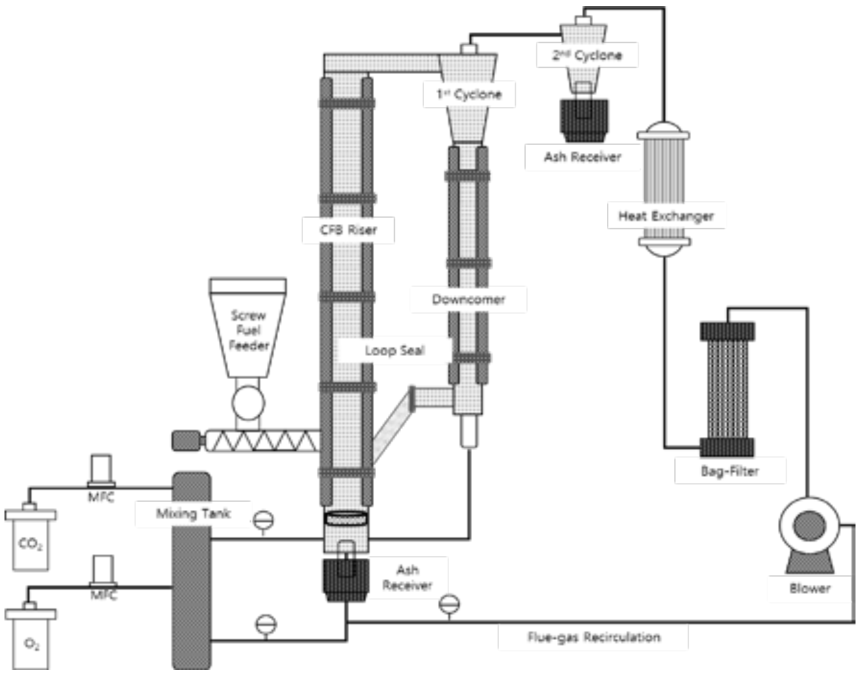


Fig. 1 Schematic diagram of the 30 kWth CFB oxy-fuel pilot test bed

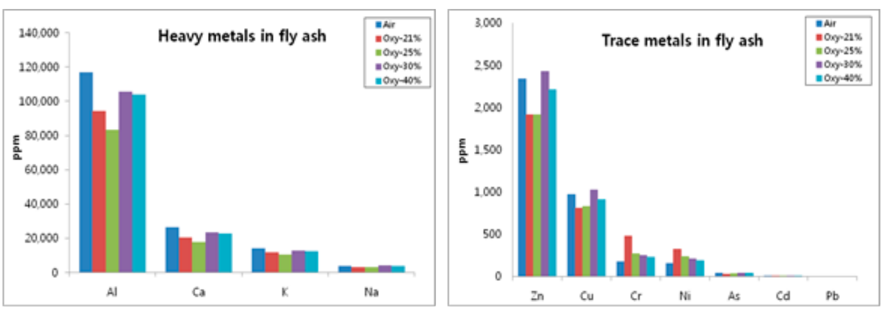
Table 1. Results of the basic characteristic analysis of waste sewage sludge

Proximate analysis (Wt, %)		Element analysis (Wt, %)	
Moisture	7.32	Carbon	28.14
Volatile	45.11	Hydrogen	4.74
Fixed carbon	12.25	Nitrogen	4.43
Ash	35.04	Oxygen	23.90
-	-	Sulfur	0.43
Calorific value (kcal/kg)	3,008	Chloride	0.053
Selected metals analysis			
Heavy metals analysis (ppm)		Trace metals analysis (ppm)	
Al	21,700.0	Zn	635.4
Ca	11,204.7	Cu	305.2
K	8,249.0	Pb	44.1
Na	5,874.9	Cr	42.5
-	-	Ni	30.8
-	-	As	6.1
-	-	Cd	1.6

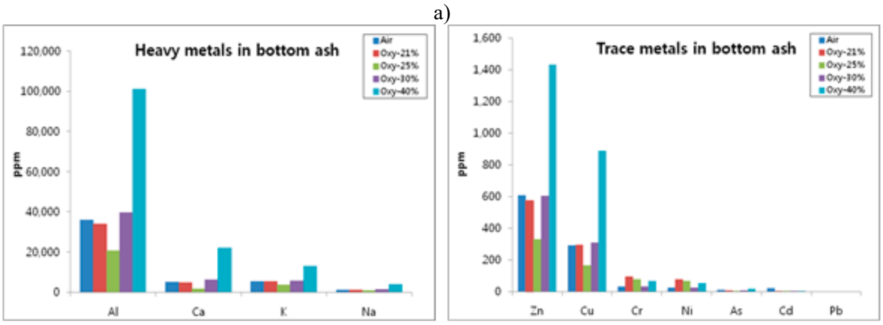
3. Results and discussion

3.1 Analysis of flue-gas and selected metals

The temperature profile of the air combustion was relatively higher than it was for the 21% of oxygen injection rate in oxy-fuel combustion. The heat capacity of carbon dioxide was much higher than that of nitrogen. This was likely because the flame temperature of the 21% oxygen injection rate in waste sewage sludge combustion was lower than it was for the air combustion due to the waste sewage sludge containing many volatiles. This leads to delayed devolatilization and total ignition time of waste sewage sludge at this oxygen rate due to a larger capacity of carbon dioxide substituted for nitrogen during the waste sewage sludge combustion. However, the temperature trend of oxy-fuel combustion of 23% of oxygen injection rate was higher than that of the air combustion. This indicates that oxy-fuel combustion with 23% of oxygen injection rate occurred at a high flame temperature compared to air combustion of waste sewage sludge, and devolatilization and total ignition time rapidly decreased with increasing oxygen injection rate. Figure 2 shows comparative concentration of selected heavy and trace metals in the fly ash from the air and oxy-fuel waste sewage sludge combustion with oxygen injection rate ranged from 21 % to 40 %. As shown figure, the species of Aluminum (Al), Potassium (K) and Calcium (Ca) were dominant in the fly ash from waste sewage sludge combustion in the air and oxy combustion. It was considered that those species are concerned with particle formation mechanism such as vaporization, condensation, and coagulation, which caused the growth of fly ash from waste sludge combustion. Figure 2b) shows comparative concentration of selected heavy and trace metals in the bottom ash from the air and oxy-fuel waste sewage sludge combustion with oxygen injection rate ranged from 21 % to 40 %. As shown figure, the concentrations of Al, K and Ca compounds in bottom ash were more decreased in the range from 21 % to 25 % of oxy-fuel condition than air and oxy-fuel condition above 30 %. It was indicated that oxy-fuel combustion with the range from 21 % to 25 % would mitigate agglomeration, fouling and corrosion problems from waste sewage sludge combustion, and economically beneficial in terms of long time operation of waste sewage sludge combustion facility. As shown figure 2 a) and b), Zn and Cu compounds in bottom ash and fly ash from waste sewage sludge combustion showed as similar trends with heavy metal compounds in both ashes, which were more decreased in the range from 21 % to 25 % of oxy-fuel combustion than air combustion. Those results are related the difference of combustion characteristics in air and oxy-fuel combustion.



a)



b)

Fig. 2 Concentrations of heavy and trace metals in ash from air and oxy-fuel waste sewage sludge combustion: a) fly ash, b) bottom ash

3.2 Particle size distribution and mass fraction of metals

Based on the analysis of flue-gas and behavior of selected metals, oxygen injection rated in oxy-fuel combustion was decided as 23%. Figure 3 shows Particle size distribution in fly ash from air and oxy-fuel combustion of sewage sludge. The alkali metals, Al, Ca, K, were mainly distributed in the super-micron particle size ranged from 1µm to 2.5 µm under both the air and oxy-fuel combustion conditions. Generally, super-micron particles are formed directly from inherent metals in fuel during combustion. Those metals were inherent refractory metals in the sewage sludge, and the particles were formed by solid to particle mechanisms of inherent refractory metals. On the other hand, sub-micron particles are formed by particle formation mechanisms such as vaporization, nucleation, and condensation. Among those refractory metals, Al, Ca, and K showed significant accumulation in the size mode that ranged from 1 µm to 2.5 µm from sewage sludge in air and oxy-fuel combustion. Originally, K was one of the highly volatilized metals, formed into sub-micron particles below 1 µm in size by particle formation mechanisms such as vaporization and condensation. However, K also formed super-micron particles over than 1 µm in size. It was explained that K metal vapor would be diffused from gas to the surface of residual ash by condensation and chemical reactions, and bonded to inherent refractory metals, such as Al and Ca, by surface growth mechanism. Figure 4 shows mass fractions of alkali and heavy metals from air and oxy-fuel combustion. In oxy-fuel combustion, the mass fractions of Al, Ca, and K were more accumulated in the particle mode below 1 µm in size than they were for air combustion. In addition, the mass fractions of Al, Ca, and K from oxy-fuel combustion that ranged from 1 µm to 2.5 µm in size were slightly higher than they were for air combustion, whereas the mass fractions of those metals from air combustion for particles over than 2.5 µm in size were higher than they were from oxy-fuel combustion. It was considered that the ignition time delay from oxy-fuel conditions was shorter than it was from air combustion, and sub-micron particle formation from those refractory metals was enhanced by a series of particle formation mechanisms such as volatilization and condensation.

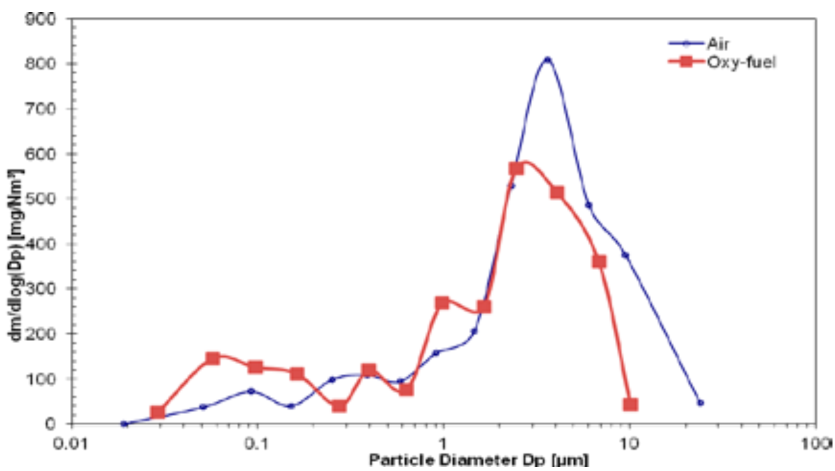
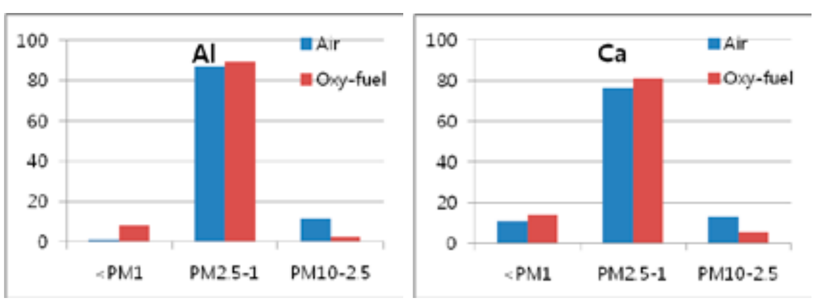


Fig. 3 Particle size distribution in fly ash from air and oxy-fuel combustion of sewage sludge



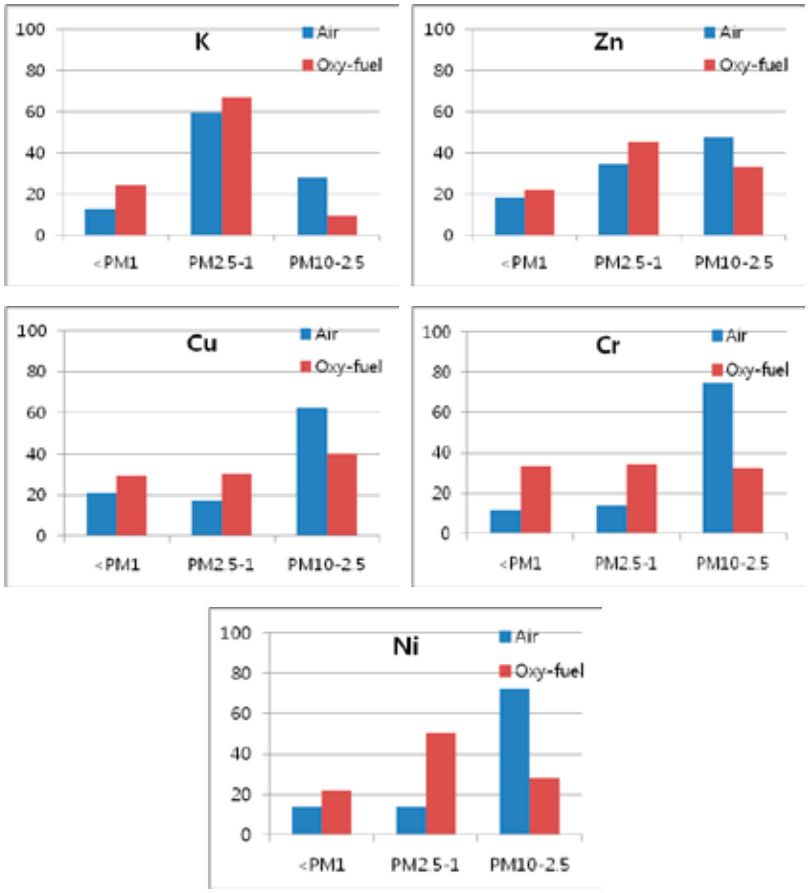


Fig. 4 Mass fractions of alkali and heavy metals from air and oxy-fuel combustion

In the case of the heavy metals, such as Cr, Ni, Cu, and Zn, sub-micron particle formation was occurred more actively in oxy-fuel combustion than it did in air combustion. It was explained that oxy-fuel combustion has different combustion surroundings by O_2/CO_2 mixture compared with conventional air combustion. The sub-micron particles could be formed by particle formation mechanism by chemical reaction of the refractory metal oxides. Generally, the refractory metal oxides could be reduced to sub-oxides by reacting with carbon monoxide in the combustion surroundings. The sub-oxide metals were de-volatilized easily due to their low melting point, and those sub-oxide metals vapor could be more actively re-oxidized to the gas phase to form sub-micron particles in oxy-fuel combustion than it was in conventional air combustion due to an enriched carbon dioxide injection under the oxy-fuel conditions. Sub-micron particle formation from Zn and Cu in heavy metals would be occurred by both particle formation mechanisms, such as volatilization and condensation, because the melting point of those metals was lower than the adiabatic flame temperature from air and oxy-fuel combustion, whereas the melting point of Cr and Ni in heavy metals was greatly higher than the adiabatic flame temperature. On the other hand, those heavy metals, such as Cr and Ni, were mainly distributed in coarse particles greater than 2.5 μm in size during air combustion. This particle formation could be occurred by coalescence of the included mineral, which was not volatilized as sub-oxide metals, and by a solid to particle mechanism of inherent refractory metals that have high melting points.

4. Conclusion

The comparative study on particle size distribution and the behavior of alkali and heavy metals from waste sludge combustion at air and oxy-fuel conditions was conducted using $30kW_{th}$ CFB pilot test bed. The particle size distribution in fly ash from air combustion was dominated by coarse particles greater than 2.5 μm in size, whereas that from oxy-fuel combustion showed each accumulation mode as sub-micron particle below 1 μm and super-micron particle ranged from 1 μm to 2.5 μm. Those results occurred by particle formation mechanisms of metal elements by different surroundings during air and oxy-fuel combustion. The concentrations of Al, Ca, and K from oxy-fuel combustion below 2.5 μm in size were higher than they were from air combustion. It was considered that the ignition time delay from oxy-fuel combustion was shorter than it was from air combustion, and sub-micron particle formation from those refractory metals was enhanced by a series of particle formation mechanisms by volatilization and condensation. Sub-micron particle formation from Cr, Ni, Cu, and Zn was more actively occurred in oxy-fuel combustion than it did in air combustion. It was considered that those sub-oxide metals vapor could be more actively re-oxidized to the gas phase to form sub-micron particles during oxy-fuel combustion than it was during conventional air combustion due to enriched carbon dioxide injection under oxy-fuel conditions.

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