

**NO_x, Fine Particle and Toxic Metal Emissions from the Combustion of Sewage Sludge/Coal
Mixtures: A Systematic Assessment.**

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ABSTRACT

This research project focuses on pollutants from the combustion of mixtures of dried municipal sewage sludge (MSS) and coal. The objective is to determine the relationship between 1) fraction sludge in the sludge/coal mixture, and 2) combustion conditions on a) NO_x concentrations in the exhaust, b) the size segregated fine and ultra-fine particle composition in the exhaust, and c) the partitioning of toxic metals between vapor and condenses phases, within the process. The proposed study will be conducted in concert with an existing ongoing research on toxic metal partitioning mechanisms for very well characterized pulverized coals alone. Both high NO_x and low NO_x combustion conditions will be investigated (unstaged and staged combustion). Tradeoffs between CO₂ control, NO_x control, and inorganic fine particle and toxic metal emissions will be determined.

Previous research results have demonstrated that the inhalation of coal/MSS ash particles cause an increase in lung permeability than coal ash particles alone. Elemental analysis of the coal/MSS ash particles showed that Zn was more abundant in these ash particles than the ash particles of coal ash alone.

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INTRODUCTION

Municipal sewage sludge (MSS) is formed during wastewater treatment. Sludge is the largest by-product of wastewater treating plants and its processing and disposal represent one of the most environmentally challenging aspects of the wastewater treating process [1]. As discharged, the sludge is contained in a dilute (0.25 to 12 wt%) aqueous suspension and contains hazardous compounds. A description of how sludge is generated can be found in [1].

One of the options currently being considered to improve the treatment of this sludge is a process involving heat treatment (to render the sludge biologically inactive) followed by dewatering, drying, pulverizing, and combustion [2]. The advantages of this combustion process are: a large reduction in disposal volume, thermal destruction of toxic organics, and recovery of the energy value of the carbon sources in the sludge [1]. However, there are a number of potential disadvantages including NO_x formation (due to the high nitrogen content of most MSS), handling of the raw material (due to the toxicity of metals and dioxins in the feedstock), possible boiler slagging and fouling problems (due to the high inorganic content of the feedstock), and the release of toxic trace elements into the environment [3].

A study was conducted to investigate the partitioning characteristics of selected trace elements during the combustion of a dried, treated (Swiss Combi Process), and pulverized MSS at commercially relevant temperatures (i.e. combustion zone and post-combustion zone temperatures similar to those expected in commercial combustors). The objectives of this study were:

- to quantify the partitioning of selected major inorganic (silicon, iron, calcium, and aluminum) and trace elements (zinc, arsenic, selenium, and antimony) during combustion of MSS in a natural gas flame
- to quantify the partitioning of selected major inorganic (silicon, iron, calcium, and aluminum) and trace elements (zinc, arsenic, selenium, and antimony) during co-combustion of MSS with a well-characterized bituminous coal (Ohio 5/6/7 blend)
- to identify mechanisms where possible for trace element vapor-to-solid phase partitioning
- to explore synergistic effects for co-combustion of MSS with coal

In this quarterly report, we explored the combustion of MSS at three different conditions: 1) 100% O₂ enrichment, 2) 50% O₂ and 50% Ar, and 3) assisted by methane and air. Previous research has shown that oxygen enrichment and argon substitution can be used to increase combustion temperatures and still maintain practical velocities through the burner. The importance of the first two experimental configurations was to demonstrate the ability of MSS to combust MSS by itself, without supplemental fuel, and oxygen enrichment was necessary to accomplish that. Furthermore, oxygen enrichment assists in CO₂ removal in the flue gas. Also discussed in this report are 1) experimentation with MSS alone, on the old furnace (with substantial leaks).

METHODS AND MATERIALS

Combustion experiments setup

The equipment set up for the experimental combustion and particle sampling conditions that were carried for this report have been previously described by [1]. Due note, however, that different MSS transport gases were utilized depending on the experimental configuration sought. With the exception of the MSS/methane assisted combustion, in all of the combustion experiments 2.0 Kg/hr of MSS were feed to the combustor. For the MSS/methane assisted combustion experiment, the feed rates of MSS and methane were 1.36 Kg/hr and 1.12 m³/hr, respectively [1]. In all of the combustion experiments, a stoichiometric ratio of 1.2 was implemented.

The physical and chemical composition of the MSS used for this work is demonstrated in Table 1.

Analysis	Value	Units
Heat of combustion	6.647	MJ/Kg
Proximate		
Moisture	5.12	wt %
Volatile Matter	39.20	wt %
Fixed carbon	1.89	wt %
Ultimate		
Carbon	19.70	wt %
Hydrogen	3.08	wt %
Nitrogen	2.22	wt %
Chlorine	0.041	wt %
Sulfur	0.59	wt %
Ash	53.79	wt %
Ash Elemental Analysis		
Al ₂ O ₃	19	wt %
CaO	8.8	wt %
Fe ₂ O ₃	15	wt %
MgO	4.1	wt %
Na ₂ O	1.8	wt %
SiO ₂ *	36	wt %
As	6.7	ppmw
Sb	7.2	ppmw
Se	3.3	ppmw
Zn	664	ppmw

Table 1: Characterization of dried and processed municipal sewage sludge. * Major elements converted to oxides on a sulfur-free basis. Data reported is not normalized.

Sorbent injection

2 gram per minute of sorbent Burgess 80 (kaolinite) was injected at the centerline and $\sim\frac{1}{2}$ meter below from the top of the furnace, which was immediately after the post flame. The methodology as to the description of sorbent injection can be found in Fernandez (2002). Flue gas temperatures were taken with a high temperature probe that is equipped with high temperature K-type thermocouple wire.

Theoretical calculations

Equilibrium adiabatic flame temperatures and multi-component chemical equilibrium conditions were completed by utilizing CHEMEQ and CEA software packages [2].

EXPERIMENTAL RESULTS AND DISCUSSION

Combustion of MSS with 33.3% O₂ and 66.6% N₂

The first of the series of experiments was to observe the combustion behavior MSS under 33.3% O₂ and 66.6% N₂ condition. The combustion of MSS under this condition was temporarily substantiated ~20 minutes. After 20 minutes, the MSS was deposited on the tip of the burner, which was quite hot, and swelled inside the burner; which caused the burner to be plugged.

Initial theoretical equilibrium adiabatic flame temperature calculation predicted that the combustion of MSS under this condition can be capable of producing an adiabatic flame temperature of 1805 K. However, the predicted calculations are only limited to ideal conditions and only model limited number of chemical species. Very low heat content.

Combustion of MSS with 100% O₂ – no air

Previously completed theoretical equilibrium adiabatic flame temperature combustion predictions indicated that it would be possible to combust MSS with 100% O₂ at high temperatures ~2599 K. This prediction was important because it would allow the combustion of MSS without the assistance of other fuels –i.e. methane, coal. Furthermore if this was the case, it would be easier to remove highly concentrated CO₂ gas from the flue gas.

The experiment was conducted as indicated. Initially, the MSS burned without any problems. However, one hour into the experiment problems appeared. A complexity associated to this experimental condition was the slow flow rate of MSS/oxygen mixture. The utilized burner contained a stainless steel tip that requires heat removal by higher flow rates of fuel/oxidizer mixtures. Since oxygen is highly reactive at high temperatures, the sustained flame remained very close to the tip of the burner; which caused the tip of the burner to come over heated and crystallized.

Compounding these problems was the composition of MSS. As demonstrated in Table 1, MSS contains large quantities of inorganic materials. As the result of overheated burner tip and slow gas flow rates, the MSS burned inside the burner. The eventual result of this event was the formation of very large glassy granules (slag) which prevented further discharge of MSS into the furnace.

Combustion of MSS with 50% O₂ and 50% Ar – no air

The lessons learned from the previous experiment allowed the implementation of another strategy; in this case, the combustion of MSS with 50% O₂ and 50% Ar. The addition of Ar was important for the transportation of MSS and to increase the gas mixture flow rate. The importance of utilizing Ar and not N₂ is that Ar is a monatomic element whereas N₂ is diatomic molecule. If N₂ was utilized for the transport MSS, then a large amount of energy would be used and wasted in the stretching of the N-N bond. The theoretical equilibrium adiabatic flame

temperature combustion predictions showed that it would be possible to combust MSS at high temperatures -2343 K – but lower than the previously discussed experiment.

This experiment alleviated the flow rate and combustion of MSS inside the burner problems. However, the combustion of MSS under this condition was sustained for about one hour. A possible explanation for this situation is that heat or energy was removed by two possible sources: 1) conduction of heat by the furnace walls and 2) convection of heat by the flue gas.

Combustion of MSS assisted by methane

A previously conducted experiment in this laboratory, [1] looked at the partitioning of trace elements during the combustion of MSS assisted by methane. The same experiment was conducted again but with a different goal. In this experiment, the goal was to test the potential for the removal of vaporized metals found in the flue gas by kaolinite sorbent. Of interest, was the potential removal of vaporized Zn. In controlled doped fuel oil combustion experiments conducted by Fernandez [3], it was determined that Zn was successfully sequestered kaolinite.

Temperature measurements at the point of sorbent injection indicated that the flue gas temperature at that point inside the furnace was greater than 1600 K. Theoretical predictions and previous experimental work indicate that the metal in question needs to be in the gas phase in order to be sequestered. At the measured flue gas temperature, theoretical calculation predict that Zn species (oxides and/or sulfated) are found in the gas phase [3].

Figures 1 and 2 demonstrate the particle size distribution (PSD) of MSS ash particles alone and MSS and kaolinite sorbent ash particles, respectively. Figure 1 demonstrates a trimodal distribution. The trimodal distribution can be broken down into the following three modes: 1) a predominant mode with an aerodynamic cut-off diameter greater than 1 μm , 2) a middle mode with an aerodynamic cut-off diameter in the range of 0.2 to 1 μm , and 3) ultra-fine particle mode with an aerodynamic cut-off diameter in the range of 0.03 to 0.1 μm . This trimodal distribution of combustion generated particles has been reported by several researchers [1, 4]. Furthermore, Seames et al. (2002) have reported the trimodal distribution finding from the combustion of MSS assisted by methane.

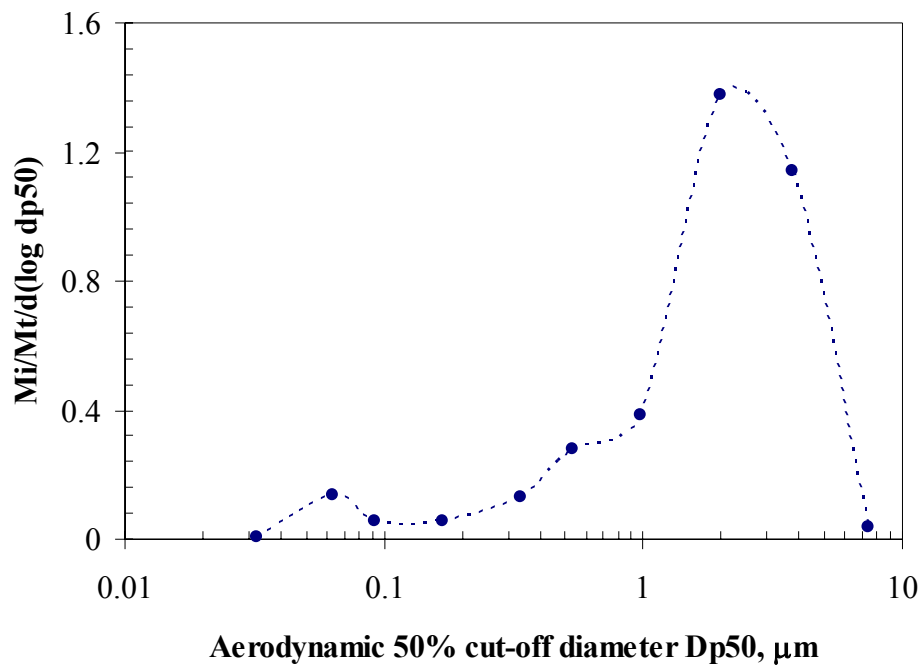


Figure 1: Particle size distribution from MSS ash particles.

Figure 2 demonstrates the PSD from MSS assisted by methane and after the injection of kaolinite sorbent. As demonstrated in this figure, the particle distribution appears to have a more predominant bimodal distribution. The predominant appears to more dominant than in the MSS ash particle distribution alone –Figure 1. Furthermore, Figure 2 demonstrates that there is also a more predominant appearance of a middle mode and an almost a disappearance of the ultra-fine mode. The near disappearance of the ultra-fine mode suggests that some of the vaporized metals found the flue gas were possible sequestered by the kaolinite particles.

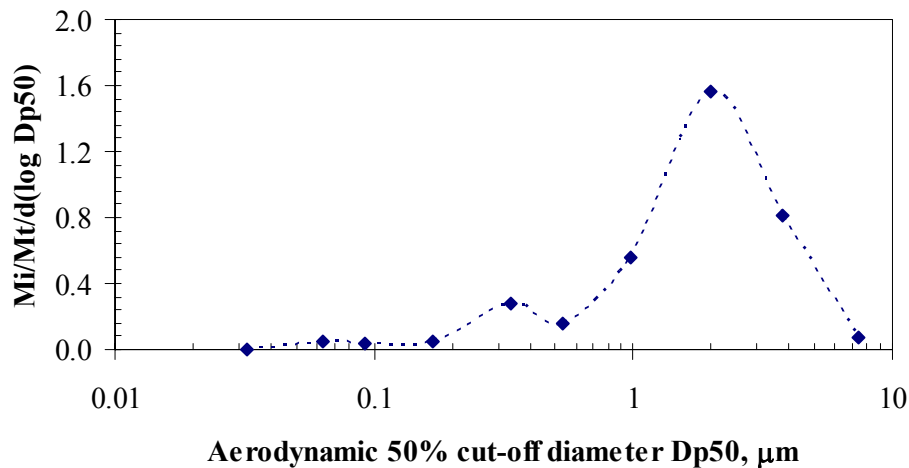


Figure 2: Particle size distribution from MSS ash particles and kaolinite sorbent.

CONCLUSIONS

Several MSS combustion experimental configurations were accomplished. The experiments ranged from the 100 % to 20% oxygen enrichment. Theoretical work indicated that it would be possible and ideal to combust MSS in 100% presence of oxygen but mixture flow rate proved otherwise. Other oxygen enhancements with air and Ar were accomplished but the result proved to be negative. The best combustion condition occurred when MSS was pre-mixed with methane.

The combustion of MSS assisted by methane generated a trimodal PSD. When kaolinite sorbent was added to the same aforementioned experiment after MSS/methane combustion, it appears that the sorbent sequestered vaporized metals that contributed the trimodal distribution and more precisely the ultra-fine mode.

FUTURE WORK

Complete the elemental analysis of the collected ash particles to determine the effectiveness of sorbent for the removal of vaporized metals in the flue gas. In addition, future work will focus on trade offs between NO_x and fine particle emissions under staged combustion conditions.

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