



2nd Oxyfuel Combustion Conference

Carbonation of coal fly ash under oxy-fuel combustion conditions

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1. Introduction

Deposition of coal fly ash in the furnace and convective sections of industrial scale boilers is of great concern because it can result in reduced heat transfer and even serious operational problems. The extent of deposition is largely dependent on ash physicochemical properties as well as the boiler design and operating conditions. Any changes in ash chemistry may have important impacts on its deposition behavior.

Oxy-fuel combustion has much higher flue CO₂ concentrations (>85%) than air combustion (<20%). The elevated CO₂ level is expected to favor the following carbonation reactions:



where M represents Ca, Mg, Na, K, or possibly Fe. The recent work conducted at the University of Stuttgart observed enhanced carbonation of coal ash (primarily Ca-containing species) in oxy-fuel combustion.[1] If this is a general phenomenon, it means that switching from air-firing to oxy-firing can change ash chemistry because of intensified carbonation reactions. As a consequence, the extent of deposition may be affected. If carbonation occurs in the deposits, it will lead to the development of hardness and strength, making ash deposits more difficult to remove.[2]

However, to the authors' knowledge, there is very little data on the carbonation of coal fly ash under oxy-firing conditions, though carbonation of minerals (e.g., Serpentine, Wollastonite) or mineral derivatives (e.g., lime (CaO)) is being actively investigated because the carbonation-calcination cycle is regarded as a promising post-combustion CO₂ capture technology for existing coal power plants.[3-5] In a thermogravimetric analyzer, using simulated flue gas, Wang et al.[6] investigated the carbonation of ash samples collected from the baghouse of a utility circulating fluidized bed (CFB) boiler, which fired a mixture of petroleum coke and coal with limestone added as sulphur

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sorbent. It should be noted that the fly ash studied was mostly derived from the sorbent and thus had a much higher CaO content (55.44 wt%) than common coal fly ash. The results showed that both the temperature and H₂O concentration played important roles in determining the reaction rate and extent of carbonation. Nevertheless, how these results can be extrapolated to oxy-fuel combustion of pulverized coal is not known. Sulfur oxides (SO_x) in practical flue gas can also have impacts on ash carbonation through competitive sulfation reactions,[7] which should be considered as well.

2. Research objectives and work activities

The present work aims to investigate ash carbonation during oxy-fuel combustion of pulverized coal on a drop tube furnace (DTF) under well-controlled conditions. For comparison, air combustion experiments will also be performed as baseline tests. The objectives of this research are to:

- (1) develop viable techniques for speciation and quantification of carbonates in fly ash;
- (2) demonstrate how oxy-firing may differ from air-firing in the extent of ash carbonation;
- (3) identify the dominant factors affecting ash carbonation; and
- (4) illustrate how flue gas component species (e.g., H₂O and SO_x) can affect ash carbonation.

At the date of submittal of this abstract, three coals of different ranks have been purposely selected so that the above objectives can be achieved. They are (a) low sulfur bituminous coal (Bit_LS), (b) high sulfur bituminous coal (Bit_HS), and (c) high calcium subbituminous coal (SubBit). Analysis of coal properties has already been completed. For carbonate characterization, X-ray photoelectron spectroscopy (XPS) and Raman spectroscopy (RS) have been preliminarily evaluated. XPS is a surface chemical analysis technique and can be used to analyze the surface chemistry of the carbonated particles, so that the carbonation mechanisms can be investigated. RS is a spectroscopic technique used to study vibrational, rotational, and other low-frequency modes in a system. The vibrational information is specific to the chemical bonds and symmetry of molecules and thus carbonates in the fly ash can be identified.

Once-through CO₂ will be used to simulate completely cleaned recycled flue gas. Each coal will be burnt under three combustion conditions: (a) air combustion, as baseline tests, (b) 27%O₂/73%CO₂, to match the radiation flux of air combustion, and (c) 32%O₂/68%CO₂, to match the adiabatic flame temperature of air combustion. Both size-segregated ash and bulk ash will be collected. A 13-stage Dekati low pressure impactor (DLPI) will be used to classify ash particles <10 μm into 13 size fractions. Therefore, carbonation of fine ash particles with different sizes can be investigated, for such micrometer-order particles play important roles in deposit initiation.

By the time of the Conference we will have a complete data set of the composition of both size-segregated ash and bulk ash for the three coals studied. The results on coal fly ash carbonation under combustion conditions described above will be presented in the final presentation.

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