

OXY-FUEL COMBUSTION ASH FORMATION MECHANISMS

Lian Zhang^{*1}, Facun Jiao^{1,2}, Eleanor Binner¹, Luguang Chen¹,
Sankar Bhattacharya¹, Yoshihiko Ninomiya², Chun-Zhu Li³

¹: Department of Chemical Engineering, Monash University, GPO 36,
Clayton, Vic 3800, Australia

²: Department of Applied Chemistry, Chubu University, 1200 Matsumoto-Cho,
Kasugai, Aichi, Japan

³: Curtin Centre for Advanced Energy Science and Engineering, Curtin University
of Technology, 1 Turner Avenue, Technology Park, WA 6102, GPO Box U1987,
Perth, WA 6845, Australia

*E-mail: lian.zhang@eng.monash.edu.au

Oxy-fuel combustion is a process of burning coal in O_2/CO_2 instead of air to generate a CO_2 -rich flue gas which has the potential to be directly sequestered and/or stored with minimal further treatment (Buhre et al., 2005). Due to the different physical properties of CO_2 relative to N_2 , a variety of distinct phenomena relating to coal combustion performance and gaseous emissions in O_2/CO_2 have been observed in both field and laboratory studies (Buhre et al. 2005; Pathnam et al 2009; Molina and Shaddix 2007). Elucidation of the ash formation mechanisms in oxy-fuel combustion is also imperative, as minerals in coal links with corrosion and erosion problems in furnace as well as the release of inhalable, potentially toxic airborne particulates.

A variety of variables could be significantly changed with gas shifting from air to a mixture of O_2 and flue gas. Depending on where the flue gas is re-circulated and if it is cleaned and/or dehydrated, the gas environment in an oxy-fired boiler could be very different. As illustrated in figure 1, in the *case 1* where flue gas is completely cleaned and dehydrated, the flue gas re-circulated back into the boiler can be

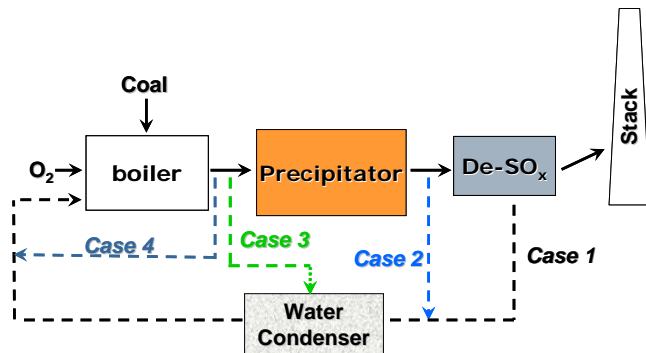


Fig 1 Schematic of flue gas re-circulation during oxy-fuel combustion

regarded as a nearly pure CO_2 . Therefore, oxy-fuel combustion only refers to a substitution of CO_2 for N_2 and a higher O_2 content in the boiler. However, in the other *cases 2-4* where flue gas is less cleaned and/or dehydrated to avoid the loss of the heat in the flue gas, the gas environment in a boiler will be greatly different from air. Plausibly, the gas impurities including steam, SO_2 , HCl and even a portion of fine ash particles can become dominant in an oxy-fuel boiler, which are vital to ash formation. A detailed understanding on these issues is essential for managing the ash-related furnace troubles and environmental impacts. It is also important for managing the CO_2 quality for the downstream carbon capture and storage (CCS).

For the *case 1* with a mixture of pure O_2 and CO_2 for coal combustion, the char temperature, local gas environment on char surface, and the char reaction rate could differ considerably from that in air combustion. In a given O_2 content, coal combustion

possesses lower temperatures for both flame and char in O_2/CO_2 than in O_2/N_2 , due to the larger specific heat capacity of CO_2 . As the diffusivity of O_2 in bulk CO_2 is 20% less than in N_2 with a same content of O_2 (Shaddix and Murphy 2003), replacing N_2 by CO_2 bulk gas could reduce the content of local O_2 while increase the content of CO_2 on char surface. The potential CO_2 -gasification reaction on char surface could also increase the CO/CO_2 ratio. These factors have the potential to affect the vaporization and melting propensities of the mineral grains embedded in char matrix. Figure 2 for the mass loss of individual metals during Victorian brown coal combustion supports this hypothesis. As can be seen, the mass loss of Na is higher in two O_2/CO_2 mixtures than in air, due to the preferential vaporization of Na as NaCl in O_2/CO_2 (Zhang et al, 2010). In addition, the consumption rate of char in O_2/CO_2 would differ from that in O_2/N_2 , it is referable that the collision/agglomeration frequency among the included minerals in char would be distinct in O_2/CO_2 , as evidenced by the results in figure 3 for the particle-size distribution (PSD) of ash formed from combustion of a bituminous coal in air versus two O_2/CO_2 mixtures (21/79 and 27/73) at 1273 K (Zhang et al., 2010).

The gas impurities including steam, SO_2 and HCl in flue gas are influential on ash vaporization, as evidenced in figure 4 for the overall brown coal ash mass loss as a function of the contents of SO_2 or HCl in O_2/CO_2 at 1273 K. This is because of the formation of gaseous chlorides and liquidus sulfates which preferentially condense into ultra-fine particulates that are difficult to capture. Introducing steam into the reactor however inhibits the ash vaporization/loss, due to the preferential formation of solid/molten alumino-silicates which rarely vaporize. Recirculation of fine ash particles into a boiler is also one influential issue which greatly affects the ash formation.

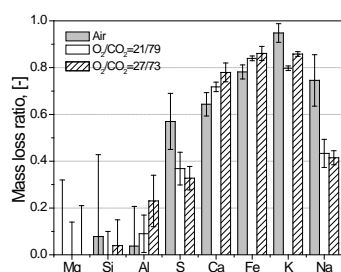


Fig 2 Mass loss of individual metals during Victorian brown coal combustion at 1273 K in Different gases in drop-tube furnace

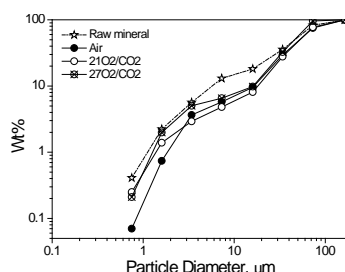


Fig 3 PSD of the ashes of a bituminous coal formed in three gases at 1273 K in DTF

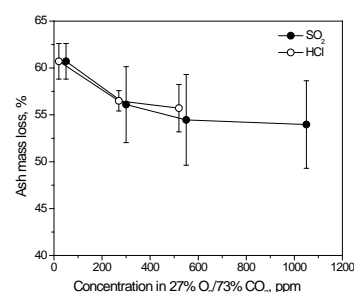


Fig 4 Overall ash mass loss during Victorian brown coal combustion in 27% $O_2/73\%$ CO_2 doped with HCl and SO_2 at 1273 K

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