



Effects of different factors on sulfur trioxide formations in a coal-fired circulating fluidized bed boiler



Baixiang Xiang, Wenfeng Shen, Man Zhang, Hairui Yang, Junfu Lu *

Key Laboratory for Thermal Science and Power Engineering of Ministry of Education, Department of Thermal Engineering, Tsinghua University, Beijing 100084, China

HIGHLIGHTS

- Heterogeneous and homogeneous formations of SO₃ in a coal-fired CFB boiler were measured.
- An improved SO₂/O₂/H₂O/CO₂/CO/NO mechanism was built based on previous mechanisms.
- Homogeneous formation of SO₃ in a coal-fired CFB boiler was numerically calculated.
- Effect of the influencing factors on formations of SO₃ was investigated.

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ABSTRACT

Experimental and numerical studies were conducted to determine the effects of different factors on homogeneous and heterogeneous formations of sulfur trioxide (SO₃) in a coal-fired circulating fluidized bed (CFB) boiler. In the experiments, homogeneous and heterogeneous formations of SO₃ under coal-fired CFB combustion conditions were measured using controlled condensation and S balance, respectively. Meanwhile, homogeneous SO₃ formation under coal-fired CFB combustion conditions was numerically calculated using an improved SO₂/O₂/H₂O/CO₂/CO/NO kinetic mechanism. Measurement results showed that the effect of fly ash on SO₃ concentration was slightly stronger than that of circulating ash. Both experimental and numerical results showed that SO₃ concentration was apparently affected by SO₂, O₂, and H₂O concentrations and temperature but barely affected by CO₂ concentration.

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1. Introduction

Given the presence of sulfur in coal, flue gas typically contains SO₂, some of which further reacts with O₂, H₂O, CO₂, CO, and NO in flue gas to generate SO₃. Flue gas produced by air-fired pulverized-coal combustion contains 0.1–1% SO₃/SO_x (Fleig et al., 2011), which could damage the boiler equipment and the atmospheric environment by causing corrosion, fouling, and blue plumes (Xiang et al., 2016a, 2016b; Moser, 2006; Moser et al., 2006; Fleig, 2012; Chen, 2009; Qi et al., 2011). The effects of various factors on SO₃ in flue gas should be elucidated to predict and control the concentration of this compound in flue gas, which are crucial to meet the increasing demands for energy conservation and emission reduction. To date, previous studies have reported several SO₃ measurement methods, namely, controlled condensation, S balance, salt solution absorption, isopropanol solution, pentol SO₃ monitoring, and use of spiral tubes (Lisle and Sensenbaugh,

1965; Cheney and Homolya, 1979; Fleig et al., 2012; Zhang, 2013; Xiao et al., 2012; Guo et al., 2013; Mueller et al., 2000; Fleig et al., 2011; Cooper and Andersson, 1997; Vainio et al., 2013; Jaworowski and Mack, 1979; Wiedner et al., 1985). Lisle and Sensenbaugh (1965) and Cheney and Homolya (1979) proposed the controlled condensation method based on the principle that sulfuric acid can be measured by cooling down the flue gas to a temperature between its H₂O dew point and acid dew point. The researchers found that this method is more reliable than other measuring methods. However, Fleig et al. (2012), Zhang (2013), Xiao et al. (2012), and Guo et al. (2013) reported that the measurement results obtained from using the controlled condensation method are apparently affected by experimental conditions and operator skills. Mueller et al. (2000) and Fleig et al. (2011) conveyed that SO₃ mole fraction is approximately an order of magnitude larger than any other S-containing species in flue gas, except for SO₂. Thus, SO₃ mole fraction can be reasonably inferred from the measured consumption of SO₂ in flue gas. Compared with the measurement results obtained from the controlled condensation method, those obtained from the S balance method are independent of

* Corresponding author.

E-mail address: lvjf@mail.tsinghua.edu.cn (J. Lu).

Nomenclature

V	the reactor volume	Q_{loss}	the net heat flux directed out of the reactor
\dot{m}^*	the inlet mass flow rate	Q_{source}	the energy deposited into the gas in the reactor
\dot{m}	the outlet mass flow rate	\bar{W}	the mean molecular weight
$N_{\text{inlet}}^{(j)}$	the number of inlets for each reactor j	<i>Greek letters</i>	
N_{PSR}	the total number of reactor modules in the reactor network	ρ	the mass density
R_{rj}	the fraction of the outflow of reactor r that is recycled into reactor j	ω	the molar rate of production by gas-phase chemical reaction per unit volume
A_m	the surface area of the m th material defined within the reactor	<i>Superscripts</i>	
$\dot{s}_{k,m}$	the molar surface production rate of the k th species on the m th material per unit surface area	j	the reactor number
K_g	the total number of gas-phase species	*	the inlet stream quantities
M	the number of materials	<i>Subscripts</i>	
Y_k	the mass fraction of the k th species	PSR	the perfectly stirred reactor
W_k	the molecular weight of the k th species	m	the m th material defined within the reactor
U_{sys}	the total internal energy	k	the k th species
h_k	the enthalpy of the k th species		

experimental conditions and operator skills. Cooper and Andersson (1997) found that SO_3 in flue gas can be measured using alkali solutions, such as NaCl, NaOH, KCl, CaCl_2 , and K_2CO_3 . However, Vainio et al. (2013) determined that SO_3 concentration measured with the salt solution absorption method is apparently affected by experimental conditions, operator skills, and salt solution. Fleig et al. (2012), Xiao et al. (2012), and Guo et al. (2013) found that the measurement results obtained from the salt solution absorption method are less reliable than those obtained from the controlled condensation method under the same conditions. The same researchers reported that the results obtained from the isopropanol solution method are apparently affected by experimental conditions and operator skills, and are less reliable than those obtained from the controlled condensation method under the same conditions. Therefore, among the SO_3 measurement methods reported in previous studies (Lisle and Sensenbaugh, 1965; Cheney and Homolya, 1979; Fleig et al., 2012; Zhang, 2013; Xiao et al., 2012; Guo et al., 2013; Mueller et al., 2000; Fleig et al., 2011; Cooper and Andersson, 1997; Vainio et al., 2013; Jaworowski and Mack, 1979; Wieder et al., 1985), the controlled condensation method is the most reliable regardless of the influence of operator skills, and the S balance method is the least affected by experimental conditions and operator skills.

With the SO_3 measurement methods mentioned above as a reference, many studies investigated the effects of various factors on homogeneous and heterogeneous formations of SO_3 in flue gas (Fleig et al., 2011; Ahn et al., 2011; Fleig et al., 2013; Reidick and Reifenhauer, 1980; Belo et al., 2014; Spörl et al., 2014; Jørgensen et al., 2007; Wang et al., 2012; Thibault et al., 1982; Chen et al., 2011). Fleig et al. (2011) focused on the gas-phase chemistry and examined the impact of different combustion parameters and atmospheres on SO_3 formation in oxyfuel and air-fuel flames; they found that SO_3 formation is influenced by the direct and indirect effects of SO_2 , O_2 , NO, and CO contents on flue gas. Ahn et al. (2011) investigated the effects of increased oxygen and CO_2 concentrations on SO_3 formation in two types of pilot-scale furnaces – a pulverized-coal and a circulating-fluidized-bed fired system – and found that higher SO_3 emissions are generated under air-fired conditions than under oxy-fired conditions in pulverized-coal testing; meanwhile, similar amounts of SO_3 emissions are produced during air-firing and oxy-firing in circulating fluidized bed (CFB) testing. Fleig et al. (2013) studied the effects of different combustion parameters on SO_3 formation in a flow reactor under post-flame conditions and found that reactive gases

(e.g., NO, CO and CH_4) promote this process. Meanwhile, Belo et al. (2014) investigated the catalytic effects of different fly ash samples derived from Australian sub-bituminous coals on heterogeneous SO_3 formation in flue gas, with air and oxy-fuel firing at 673–1273 K. Their results showed that the catalytic effect accounts for roughly 95% of the total conversion of SO_2 – SO_3 at 973 K, whereas the catalytic effects of fly ash samples derived from air and oxy-fuel firing under different flue gas environments are similar. Thibault et al. (1982) studied the absorption of SO_3 by CaO and MgO particles and found that the uptake rate is mainly dependent on intragranular diffusion and essentially independent of gross particle size in the range 0.06–0.33 mm. In consideration of the significant differences in grain size and porosity between CaO and MgO particles, the uptake rate in MgO is considerably faster than that in CaO. Moreover, Chen et al. (2011) measured the SO_3 removal capacities of CaO, $\text{Ca}(\text{OH})_2$, and CaCO_3 by using a fixed-bed reactor with different flow rates at temperatures ranging from 573 K to 673 K. They found that the SO_3 removal capacity of $\text{Ca}(\text{OH})_2$ is greater than those of the two other compounds; in addition, the removal efficiency decreases with increasing flow rate increases, and temperature only slightly affects the removal efficiency. With the rapid development of coal-fired CFB combustion and in-furnace sorbent injection-based desulfurization technologies, understanding the effects of different factors on homogeneous and heterogeneous formations of SO_3 in flue gas under coal-fired CFB combustion conditions has become highly necessary. However, insufficient information is available about the effects of different factors on homogeneous and heterogeneous formations of SO_3 in a coal-fired CFB boiler; thus, the increasing demands for energy conservation and emission reduction have yet to be met (Fleig et al., 2011; Ahn et al., 2011; Fleig et al., 2013; Reidick and Reifenhauer, 1980; Belo et al., 2014; Spörl et al., 2014; Jørgensen et al., 2007; Wang et al., 2012; Thibault et al., 1982; Chen et al., 2011).

Aside from the experimental studies on the different factors influencing homogeneous and heterogeneous formations of SO_3 in flue gas, several kinetic modeling works have been also conducted (Mueller et al., 2000; Alzueta et al., 2001; Giménez-López et al., 2011; Rasmussen et al., 2007; Yilmaz et al., 2006; Hindiarti et al., 2007; Sendt et al., 2002; Allen et al., 1997; Spencer et al., 1012). Mueller et al. (2000) developed and validated a detailed chemical kinetic mechanism based on the reaction profile measurements of CO, CO_2 , O_2 , NO, NO_2 , and SO_2 in a flow reactor. Spencer et al. (1012) proposed a detailed chemical kinetics

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