



# The role of steam in silica vaporization and ultrafine particulate matter formation during wet oxy-coal combustion



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## HIGHLIGHTS

- H<sub>2</sub>O enhances the vaporization of silica in wet oxy-coal combustion.
- Improved model for SiO<sub>2</sub> vaporization was developed to analysis the effect of H<sub>2</sub>O.
- Changes of char combustion characteristic caused by H<sub>2</sub>O are the primary factors.

## ARTICLE INFO

### Article history:

Received 23 March 2014

Received in revised form 1 June 2014

Accepted 17 July 2014

### Keywords:

Silica  
Vaporization  
Ultrafine particulate matter  
Oxy-fuel combustion  
Steam

## ABSTRACT

Oxy-coal combustion produces a combustion environment that contains large amounts of CO<sub>2</sub>, H<sub>2</sub>O compared to conventional air combustion. Silica (SiO<sub>2</sub>) is a major component of the mineral impurities in coal and the vaporization of silicon containing species controls the initial phase of formation of ultrafine particulate matter (ultrafine PM) via vaporization–condensation mechanism. To investigate the vaporization behavior of silica in wet recycle oxy-coal combustion conditions, synthetic chars with SiO<sub>2</sub> inclusions were burned at 1873 K in the O<sub>2</sub>/CO<sub>2</sub>/H<sub>2</sub>O environments with steam of up to 10 vol.%. Experiments were carried out with a high temperature drop tube furnace (HDTF) system and ultrafine PM was collected with a low pressure impactor (LPI). Meanwhile, modeling work was carried out for further investigation of the effect of H<sub>2</sub>O on the vaporization of silica. The improved model was developed based on a model built in conventional air combustion conditions and verified by the experiment. The results indicate that, under the wet recycle oxy-coal combustion conditions, H<sub>2</sub>O in combustion atmosphere significantly enhanced the vaporization of SiO<sub>2</sub> and considerably increased the yield of the ultrafine PM. The variation of char combustion characteristics such as burning temperature and gas properties surrounding the mineral inclusions caused by H<sub>2</sub>O seemed to be the primary cause. Especially, effect of H<sub>2</sub> inside the char particle cannot be ignored.

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## 1. Introduction

Ultrafine particulate matter (ultrafine PM, particles with aerodynamic diameters of <0.3 μm) is one of the most critical air pollutants in China [1]. A considerable part of the particles is believed to be derived from coal combustion. Ultrafine PM is generally rich in hazardous materials such as heavy metals and polycyclic aromatic hydrocarbons (PAHs), which are seriously harmful to public health. Unfortunately, there is currently no effective technology to control the emissions of ultrafine PM from combustion systems [1,2]. Understanding the formation of ultrafine PM is essential for developing technology for PM emission control.

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Si is well known to be one of the major refractory elements in the ultrafine PM emitted from the combustion of coals [1–5]. Moreover, silica (SiO<sub>2</sub>) plays an important role in the formation of ultrafine PM, as the condensation of SiO<sub>2</sub> vapor provides the initial nuclei [4,6]. Generally, the vaporization of SiO<sub>2</sub> dominates the initial phase of formation of ultrafine PM via the vaporization–condensation mechanism [4,6]. Therefore, it is of particular importance and has attracted extensive investigation [2,4,7–9]. During the combustion of coal particles, SiO<sub>2</sub> inclusions undergo reduction reactions and vaporize as volatile SiO in the local reducing atmosphere [4,8]. In the early 1980s, Quann [4] systematically investigated the vaporization behavior of refractory minerals by burning various coals in air-combustion conditions on a drop tube furnace (DTF) and proposed a model to describe the reduction reaction of SiO<sub>2</sub> and the transportation of the generated SiO in char pores [10]. They pointed out that combustion

**Nomenclature**

$\alpha$	stefan flow factor	$T$	temperature
$\varepsilon$	porosity	$V$	vaporization rate of each mineral inclusion
$\theta$	volume fraction of mineral inclusions in coal particle		
$\rho$	density	<i>Subscript</i>	
$\tau$	tortuosity	$a$	apparent
$\Phi$	thiele modulus	$e$	effective
$a$	reaction activity factor	$i$	initial
$c$	concentration	$k$	Knudsen
$d$	pore diameter	$o$	oxygen
$f$	vaporization fraction	$p$	coal particle
$r$	radius	$m$	mineral inclusions
$t$	burnout time		
$x$	molar fraction	<i>Superscript</i>	
$A$	ash content	$b$	bulk gas
$D$	diffusivity	$e$	equilibrium
$Ke$	equilibrium constant of reaction	$s$	surface
$M$	relative molecular mass		
$P$	partial pressure		

conditions, especially the combustion atmosphere, have a great impact on the vaporization of SiO<sub>2</sub>, which is consistent with the studies by Lee et al. [11] and Sui et al. in our group [12]. Buhre et al. [7] investigated the factors governing the vaporization of silica based on experiments with five bituminous coals, and concluded that the occurrence of silica in coal as well as the char burning temperature significantly affects the vaporization behavior. Because the vaporization processes of SiO<sub>2</sub> and the factors governing vaporization are complicated, the evaporation and related ultrafine PM formation are not fully understood. More fundamental research is still required particularly for those in coal combustion processes involving emerging technologies.

Recently, with the rapid development of oxy-coal combustion (O<sub>2</sub>/CO<sub>2</sub> combustion) technology, more and more attention is focused on the formation of ash particles in the O<sub>2</sub>/CO<sub>2</sub> atmospheres [3,9,13]. The variation of the vaporization of SiO<sub>2</sub> appears to be a significant cause of the change in the ultrafine PM generated from the O<sub>2</sub>/CO<sub>2</sub> combustion compared with that from the conventional air combustion [3,9]. Kazanc et al. [3] comprehensively investigated the vaporization of SiO<sub>2</sub> in the O<sub>2</sub>/CO<sub>2</sub> combustion condition, based on experiments and a model modified from Quann et al.'s model. They concluded that the vaporization of SiO<sub>2</sub> decreases as the result of the lower char burning temperatures and the change of the gas properties, and their results confirm the reports of Suriyawong et al. [9]. While previous studies on the vaporization of SiO<sub>2</sub> were limited in dry (O<sub>2</sub>/N<sub>2</sub> or O<sub>2</sub>/CO<sub>2</sub>) combustion conditions; there have been no studies on the vaporization behavior of SiO<sub>2</sub> in wet oxy-coal (O<sub>2</sub>/CO<sub>2</sub>/H<sub>2</sub>O) combustion conditions. In the wet recycle mode there is a high content of H<sub>2</sub>O in addition to CO<sub>2</sub> [13–17], and the effect of H<sub>2</sub>O on coal combustion and direct sulfation of limestone cannot be ignored as reported by Hecht et al. [18], Duan et al. [19] and Hu et al. [20]. Therefore, it is essential to clarify the effect of H<sub>2</sub>O on the vaporization of SiO<sub>2</sub> and the formation of ultrafine PM in oxy-coal combustion conditions, especially for wet oxy-coal combustion conditions.

In this study, the vaporization behavior of SiO<sub>2</sub> and the formation of ultrafine PM in O<sub>2</sub>/CO<sub>2</sub>/H<sub>2</sub>O environments were studied based on combustion experiments using synthetic chars with SiO<sub>2</sub>. Moreover, an improved model that considers the effect of H<sub>2</sub>O in the atmosphere to describe the vaporization of SiO<sub>2</sub> was developed based on Quann et al.'s model. The effect of CO<sub>2</sub> and especially H<sub>2</sub>O on the vaporization behavior of SiO<sub>2</sub> and the formation of ultrafine PM during char combustion are discussed.

**2. Experimental****2.1. Material**

Synthetic char containing SiO<sub>2</sub> inclusions was used for combustion experiments. This model material was prepared with furfural alcohol (chemically pure) and SiO<sub>2</sub> powder (5 μm) with carbon black (20 nm) as a pore former following the procedure of Senior [21]. The proximate and ultimate analyses and heating values are shown in Table 1 and are similar to that of bituminous coal. As reported by Senior et al. [21,22], this model material has combustion characteristics similar to coal, especially in the char burning phase. The ash composition of the char was analyzed with X-ray fluorescence (XRF, EDAX EAGLE III), which is also shown in Table 1. The pore structure was analyzed by N<sub>2</sub> absorption (BET method) (ASAP2020) and mercury penetration (AutoPore IV 9500). The results are listed in Table 2.

Synthetic char is a type of well-defined model material of coals, which has been successfully used in various studies, such as those of char combustion, gaseous pollutant formation and mineral transformation [21,23]. Pulverized synthetic char particles of 45–75 μm were burned in experiments. It can be seen in Table 1 that the ash content of the synthetic char was 6.7%, and 90.9% of which was SiO<sub>2</sub>. The unique composition allows this study to focus on the vaporization behavior of SiO<sub>2</sub> while minimizing interferences from impurities (Na, Al, Ca, etc.). Moreover, the mineral inclusions added to the synthetic char were well controlled, which provided accurate parameters for the modeling work, especially the mineral particle size.

**2.2. Combustion conditions**

Combustion experiments of the synthetic char were carried out on a high temperature drop tube furnace (HDTF). The HDTF consists of a material feeding system, gas delivery system, high temperature furnace and sampling system, as reported elsewhere in detail [24]. Synthetic char was fed into the furnace at a rate of 0.1 g/min by a SANKI piezo bowl vibratory feeder, and then burned in a simulated atmosphere at a furnace wall temperature of 1873 K. O<sub>2</sub> concentrations in the simulated air combustion (ON21), dry recycle oxy-coal combustion (OC31) and wet recycle oxy-coal combustion (OC31 + 10H<sub>2</sub>O) conditions were 21%, 31% and 31% by volume, respectively. In the OC31 + 10H<sub>2</sub>O conditions,

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