



Full Length Article

A novel Ti-based sorbent for reducing ultrafine particulate matter formation during coal combustion



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ABSTRACT

Ultrafine particulate matter (PM) is an important part of $PM_{2.5}$, which is enriched with hazardous components and more harmful; meanwhile it cannot be effectively removed by the common-used dust collectors. In-furnace sorbent injection is an emerging technology to reduce the emission of PM in the coal combustion and the sorbent is a key factor determining its feasibility. In this study, to seek new PM sorbents, eight minerals were first separately added into a pulverized coal and burned in a drop-tube furnace (DTF) at 1773 K. The derived PM was collected via a Dekati Low pressure impactor (DLPI) sampling system and impacts of each mineral on the PM emission were evaluated. Then, the tested minerals were burned with pure sodium acetate (NaAc) under the same conditions to determine their Na fixation abilities. Finally, the PM reduction mechanism of the sorbent was discussed based on the particle size distribution, mass yield, composition and micromorphology of PM and the chemical and physical properties of the sorbent. A novel Ti-based PM reduction sorbent was screened out, which exhibited an ultrafine PM ($PM_{0.2}$) reduction efficiency of $\sim 39\%$ under the experimental conditions at an addition ratio of 5% (wt. %, coal basis). The Ti-based sorbent reacted with Na-contained vapour and formed sodium titanates. Fixation of the Na-contained vapour by the sorbent was considered to be the primary PM capture mechanism. What's more, the high sintering temperature of the Ti-based sorbent facilitates its PM reduction performance. Under the high temperature combustion conditions, the Ti-based sorbent exhibited a good performance in capturing ultrafine PM and Na-contained vapour, indicating its potential of being a high temperature sorbent.

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1. Introduction

Particulate matter (PM) emitted from the coal-fired power plants is causing severe ambient PM pollution in China [1,2]. Ultrafine PM (PM with the aerodynamic diameter of $<0.2 \mu\text{m}$) is of particular concerns as it is enriched with the hazardous components (e.g., trace elements, PAHs, etc.) and is much more harmful to the human health [1,3,4]. Coal combustion in the power plants is one primary PM emission source, especially the ultrafine PM [1,5]. At present, dust collectors (e.g., ESP, fabric filter) are widely installed downstream of the furnace to remove PM in the flue gas [5]. Although these dust collectors generally have total PM collection efficiency of over 99%, they cannot effectively remove the ultrafine PM [5–8]. Adding more chambers and/or electrostatic fields in the dust collectors, or improve their operation parameters may help improve the removal of fine and ultrafine PM meanwhile it is not economical or not feasible due to lacking space on the existed

power plant units. To effectively and economically improve the removal of ultrafine PM, the PM sorbent technology has been proposed, in which certain sorbents/additives are injected into the furnace and reduce the emission of PM and trace elements simultaneously during the combustion process [5,9–14]. In this way, the yield of ultrafine PM is reduced in the furnace, thereby easing the burden of the traditional PM removal devices (e.g., electrostatic precipitator, baghouse, etc.) downstream of the furnace and finally reinforcing the control of ultrafine PM.

Sorbent is a key factor affecting the feasibility of this technique and developing high efficient PM sorbents has been a focus in this field. Briefly, three types of PM sorbents have been reported in public, including the Si-/Al-based, the Ca-based and the Mg-based sorbents. These sorbent materials are different in the composition and exhibit different PM removal performance and mechanism [12]. Kaolin, alumina and silica are the most widely studied Si-/Al-based sorbents [10,12,15,16]. As reported, the Si-/Al-based sorbent particles injected into the furnace would react with the mineral vapour (e.g., alkali metal, heavy metal, etc.) released from the burned coal and inhibit their migration into

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the ultrafine PM. In this way, the formation of ultrafine PM is reduced by the so-called vapour-capture mechanism [9,17–19]. Laboratory studies by Linak et al. [9] showed that kaolin addition into the residual oil combustion reduced the ultrafine nuclei (less than 0.1 μm) by $\sim 35\%$. In another laboratory study on coal combustion, kaolin was reported to reduce the yield of PM_1 (PM less than 1 μm) by 20–60% with an addition ratio of 5 g-kaolin/100 g-coal. Moreover, the tested kaolin exhibited the best performance at 1373 K when the combustion temperature increased from 1173 to 1573 K [10]. Our previous study further showed that kaolin (3 g-sorbent/100 g-coal) can reduce the emission of $\text{PM}_{0.2-0.5}$ by 9–48% at the combustion temperature of 1773 K and the PM reduction performance of kaolin depended closely on the properties of minerals in coal and combustion atmosphere [12]. Besides, some hazardous heavy metals such as Pb, Cd and Cr are simultaneously captured by the Si-/Al-based sorbents [17–19]. The Ca-based PM sorbents mainly include limestone, hydrated lime and calcined lime [10,11]. Studies on a lab scale drop tube furnace (DTF) showed that the addition of limestone (5 g-sorbent/100 g-coal) reduced the emission of PM_1 by 20–70% during the combustion of a high S-content coal at 1173–1573 K. Different from the Si-/Al-based sorbents, the Ca-based sorbents added into the furnace primarily capture the acidic species such as SO_x released out of the burning coal particles. And the performance of such Ca-based sorbents is observed to be related to the S content in coal [10]. The Mg-based PM sorbents are developed by Wei and Ninomiya et al. [13,14], which mainly are $\text{Mg}(\text{OH})_2$ and $(\text{CH}_3\text{COO})_2\text{Mg}$. It was reported that Mg-based sorbents exhibited removal efficiencies of 40–50% and 40–60% for PM_1 and $\text{PM}_{2.5}$ respectively with the coal-based addition ratio of 5%. Different from the aforementioned Si-/Al-based and Ca-based sorbents, Mg-based sorbents would enhance the melting of minerals and thereby capture the small PM through an additional liquidus-capture mechanism besides the vapour-capture mechanism above. In conclusion, PM sorbent could effectively reduce the emission of fine PM from the combustion of coal. However, up to now, only few PM sorbents are available in public and they might hardly meet the various combustion conditions (e.g., combustion temperature, atmosphere, coal property, etc.). Therefore, more efforts are needed to seek new effective PM sorbents in developing the in-furnace PM control technique.

As reviewed above, PM sorbents reduce the emission of ultrafine PM mainly via capturing the PM precursor (e.g., vapourized mineral matter such as alkali metal vapour, SO_x , etc.) during the coal combustion. In this regard, sorbents/additives that are developed to capture gaseous mineral species at high temperature may also be used as PM sorbents. Such additives include clay minerals (mainly containing SiO_2 and Al_2O_3) and functional materials (e.g., volcanic ash, coal fly ash, etc.) used in solving the ash deposition, corrosion and slagging problems involved in the combustion/gasification of coal or biomass [20–24]. For example, Shadman and Punjak et al. [25,26], Lee et al. [27] reported that emathlite, silica gel, alumina, diatomaceous earth, activated bauxite, attapulgus clay and dolomite could react with the vapourized mineral matter (e.g., NaCl, Pb, Cd, etc.) and fix them in the solid particles as aluminosilicates. And their performance appeared to be depended on the Si/Al content and their structures. Other materials such as bentonite, hollloysite, bauxite and coal fly ash were also used to capture Na and K in biomass combustion and IGCC (Integrated Gasification Combined Cycle) [28–30]. Generally, above sorbent materials have distinctive physical properties (e.g., melting temperature, specific area, etc.) and/or chemical properties (e.g., reactivity, capacity, etc.), which facilitate their reaction with the vapourized mineral matter and are believed to bring them the ability to inhibit the partitioning of mineral matter into ultrafine PM. However, as far as we know, there are still no studies quantita-

tively evaluate the effects of above materials on the formation of ultrafine PM during coal combustion and their potential of being PM sorbent.

In this paper, the effects of five clay minerals (montmorillonite, attapulgite, rectorite, vermiculite and wollastonite), one volcanic rock (pumice), one metal oxide (anatase) and the well-studied sorbent-kaolinite on the emission of the ultrafine PM are studied. First, yields of ultrafine PM from the combustion of coal with each sorbent were compared with that of the blank run without sorbent addition to evaluate the performance of sorbents. And then, treatment of each sorbent with alkali metal vapour were carried out to ascertain the possible reactions and determine their Na capture capacities under the experimental conditions, which together with the particle size distribution, composition and morphology of PM help explore the reduction mechanism of the sorbents on ultrafine PM. Finally, properties led to the better performance of selected sorbents than the others were investigated.

2. Experimental

2.1. Materials

A Chinese lignite coal (denoted as HLHC) with the particle size between 45 and 90 μm was used in the experiments. The ash content of coal HLHC is 21%, and other properties of coal are shown in Table 1. As can be seen, coal HLHC has a relatively high Na content.

In total, 8 mineral materials were involved in the present study. They were 6 clay minerals (i.e., kaolin, montmorillonite, attapulgite, rectorite, vermiculite and wollastonite), a volcanic rock (pumice) and a metal oxide (anatase or titanium dioxide). The formula, Sauter mean diameter $[D(3, 2)]$ and specific surface area (SSA) of each sorbent were listed in Table 2 and their elemental composition and minerals were listed in Table S1 in the supplementary material. Briefly, kaolin (KAO), montmorillonite (MON), vermiculite (VER) and rectorite (REC) were layer structure clay minerals with Si and Al as the primary components; meanwhile, they were of different $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratios. Anatase (ANA) was also of a layer structure while its primary component was titanium (Ti). Pumice (PUM) was a typical porous volcanic rock, which was mainly composed of Si and widely used in the water purification. Sorbent candidates used in the study were obtained from different mines, and few impurities were in them except for some SiO_2 and FeS_2 (see Fig. S1). Prior to the experiment, sorbents were dried (298 K), pulverized and screened, and the portion of a narrow size distribution was used. As shown in Table 2, Sauter mean diameters of the used sorbents were mostly located in the size range of 3.7–10.7 μm . The specific surface areas of the materials differed significantly from each other (1–140.99 m^2/g). In the second part experiments, sodium acetate (NaAc, chemical pure) powders were used.

2.2. Combustion of coal/sorbent blends and PM sampling

Two part experiments were carried out successively. In the first part, pulverized coal/sorbent blends as well as the coal or sorbent alone were burned in a high temperature drop tube furnace (DTF) system to evaluate the effects of the each sorbent on the PM emission. As sketched in Fig. 1, DTF system consisted of a SANKI piezo bowl vibratory feeder, a corundum tube reactor, a gas distributor and a furnace controller, and more detailed information has been described in our previous reports [31,32]. Before the experiment, each sorbent was blended with the pulverized coal mechanically at a mass ratio of 5 g-sorbent/100 g-coal. During the experiment, coal/sorbent blends were fed into the furnace at a constant rate

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