



## Full Length Article

## Effect of the interaction between sodium and soot on fine particle formation in the early stage of coal combustion



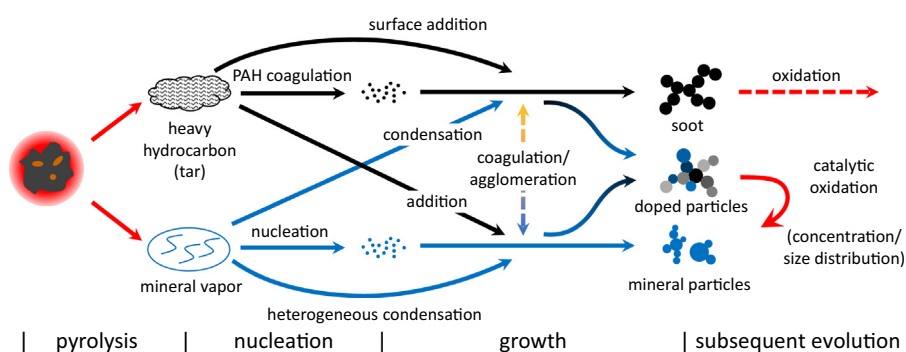
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## HIGHLIGHTS

- Na-soot interaction in the early stage of coal combustion was investigated.
- Na doping formed through flame synthesis reduced the oxidation temperature of soot.
- Na-soot interaction reduced the fine particle yield of a bituminous with Na addition.
- A schematic of the interactions between evaporative minerals and soot is proposed.

## GRAPHICAL ABSTRACT



## ARTICLE INFO

## Article history:

Received 26 January 2017

Received in revised form 18 April 2017

Accepted 5 June 2017

## Keywords:

Sodium doping

Soot oxidation

Fine particulate matter

Coal combustion

## ABSTRACT

Mineral transformation and soot formation are the two governing mechanisms for fine particle formation in the early stage of coal combustion. The interaction between the evaporative alkali species and soot, which can have significant effect on fine particle formation, however, is not fully understood. To investigate the interaction between Na and soot in the early stage of coal combustion, we characterized the particles derived from four coals samples, including acid-washed and Na-loaded samples, under both the pyrolytic and oxidizing atmospheres. It was found that Na can reduce the soot yield and particle size under the pyrolysis condition. The Na-doped soot was formed by flame synthesis of vaporized minerals and soot precursors, and the Na content had a strong effect on soot oxidation activity. The Na-soot interaction was found to reduce the fine particle yield in the early combustion stage, as the high Na concentration and strong sooting tendency of a sodium doped bituminous (YK-Na) resulted in a lower fine particle yield. A schematic of the Na-soot interaction pathways is proposed based on the experimental results. Catalyzing soot oxidation and eliminating ultrafine mineral particles were found to be the two principal ways by which Na-soot interaction affects the formation and evolution of fine particles.

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## 1. Introduction

Coal combustion is one of the key contributors to particulate matter pollution. Due to increasing concerns about the environ-

mental and the health effects of particulates, significant research has been conducted on elucidating particulate formation mechanisms and developing emission reduction technologies [1,2]. Recently, the early stage of coal combustion has attracted special attention, since it has been proved that this short period plays an important role in ultrafine particle formation [3,4]. The early combustion stage is characterized by volatile release and combustion, in a typical time of only tens of milliseconds [5]. But the release

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and subsequent reaction of volatile and vaporized mineral species under elevated temperature contributes most ultrafine particulates [6,7]. As a result, a substantial amount of ultrafine particulate matter can be observed in the flame region, with a residence time as short as 100 ms [8,9]. Thus, to further understand ultrafine particle formation mechanisms, investigation on the early combustion stage is of importance.

Previous studies found that the formation and evolution of fine particles in the early combustion stage is governed by mineral transformation and sooting process [1,3]. Evaporative minerals, especially the alkali and alkali-earth minerals (AAEM) existing in water-soluble form or organic form, exhibit extensive gas-phase release at high temperatures, forming ultrafine particles through gas-to-particle conversion [10–14]. Experiments carried out in laboratory-scale furnaces have revealed the particle size distribution (PSD) evolution with residence time, as well as the enrichment behavior of evaporative mineral species in ultrafine particles [8,9,15,13]. More recently, with the combination of flat-flame burners and different measurement methods, including dilution sampling and laser diagnostics, a better insight has been achieved into the control mechanism and dynamic behavior of ultrafine particle formation during the early combustion stage [3,4,16]. Besides the mineral transformation, heavy hydrocarbons, or tar, undergo secondary reactions and generate soot [17], which is a potential source of particulate matter in industrial and residential coal combustion systems [18,19]. Dilution sampling carried out on a quasi-one dimensional furnace showed that soot formation and oxidation have a strong impact on the composition and size distribution of particles in the flame region [9]. Measurements of soot loading evolution were also carried out under the pyrolysis conditions [20,21] and combustion conditions [22], as well as a laser diagnostic for the 2D planar distribution of soot concentration [22]. Separation methods for char and soot particles were also developed and the two types of carbonaceous particles were found to possess distinct properties [23]. As an insight into the two governing mechanisms, however, the PSD in the early combustion stage is less well characterized, especially for coals with different AAEM contents and sooting tendencies.

Moreover, in most studies on particle formation during coal combustion, mineral transformation and soot formation were considered as two separate pathways, while the researches on soot derived from hydrocarbon flame [24–26], diesel engine [27–29] and biomass soot [30] all suggested the existence of strong interactions between soot and mineral species in combustion. Several interaction mechanisms have been proposed, including the ionic effect, homogeneous reaction and catalytic oxidation [25,26], as well as the cation-molecule binding which was recently studied by several researchers using *ab initio* computation [31,32]. In particular, alkali metals were among the most active metallic elements that affect soot formation and oxidation, due to their low ionization potential [25,26] and high catalytic activity [33,34]. Experiments on sodium-doped soot showed that as low as 1% of sodium content could result in a 140 K decrease in the soot oxidation temperature [33,34]. As for early stage of coal combustion, interaction between soot and AAEM species might have strong impact on fine particulates formation, especially for the low-rank coals with high sodium content [12,14,35]. Although doping of Na and Fe has been observed in the coal-derived soot [9,36], the effect of this interaction on fine particulates formation has not been investigated in detail.

Thus, this work focuses on the effect of Na-soot interaction on particulate formation. Four coal samples with different sodium concentrations and sooting tendencies were tested in a flat-flame reactor, and the fine particulates derived under pyrolysis and combustion conditions were sampled and characterized. The interac-

tion mechanisms are further discussed based on the experimental results.

## 2. Experimental section

### 2.1. Experimental apparatus and operating conditions

Experiments were conducted in a downward flat-flame reactor, as shown in Fig. 1. A 42 mm diameter Hencken burner made of a honeycomb and hundreds of capillary tubes was employed to provide a uniform high-temperature post-flame environment. The combustion gas was isolated from the outer atmosphere by a quartz chimney. A de-agglomeration particle feeder was applied to generate a well dispersed coal particle stream. The particle stream was extracted from the reactor by a sampling probe, in which the combustion aerosol was quenched and diluted to avoid further reaction and agglomeration. A detailed description of the burner and sampling probe can be found in our previous work [3].

A mixture of 20% methane and 80% carbon monoxide was used for fuel gas. The flowrates of fuel gas, oxygen, and nitrogen were adjusted by mass flow controllers to obtain specific conditions, including temperatures, oxygen concentrations and gas velocities. During the experiments, the flame temperature was set to 1500 K, while the aerosol was extracted at a 50 mm axial distance from the burner surface, where the residence time was 30 ms. The conditions of pyrolysis and 20% O<sub>2</sub> were tested to investigate the particle formation with and without soot oxidation, respectively. A stoichiometric ratio of 1.1 was chosen for the pyrolysis condition to eliminate oxygen in the post-flame environment. The coal entraining gas had the same oxygen concentration as the post-flame environment, and the flow rate was set to 200 mL/min. To obtain the mass of coal fed, a precision balance was used to weigh the particle feeder before and after each experiment. The coal feeding rate was 0.10–0.20 g/min for all coal samples.

### 2.2. Aerosol sampling and analysis

Two sampling systems were employed in this study. One system, consisting of a 10 μm cyclone and an electrical low pressure impactor (ELPI+, Dekati Ltd.), was used for 14-stage size-selective sampling. Greased aluminum foil was used for the particle size distribution measurement. Nylon membrane was used for scanning electron microscope (SEM) and energy dispersive spectroscopy (EDS) analysis (MERLIN, Carl Zeiss AG). The particle deposition time for SEM-EDS samples was limited to 30 s to reduce particle packing and overlapping. Carbon coating was carried out on the samples with an ion beam coater (Model 681, Gatan Inc) after particle deposition.

The other sampling system, including a virtual impactor, two cyclones and a filter, was utilized to collect the coal derived particles under the pyrolysis condition. The cut-off size of the trap impactor was designed to be approximately 15 μm, which was based on a Stokes number of  $Stk_{50} = 0.6$ , as reported by Kim et al. [37]. The two cyclones were designed and calibrated to possess cut-off sizes of 6 μm and 1 μm, respectively, based on the work of Kenny et al. [38,39]. The trap impactor collected large char particles, and the remaining coarse particles, such as fragmented char particles, were removed by the two cyclones. Finally, submicron particles, which mainly consisted of soot and vaporized mineral aerosol, were collected by quartz filters (Pallflex Tissuquartz, Pall Corp.). Prior to use, the quartz filters were calcined at 650 °C for 12 h to eliminate the problem of subsequent mass loss at the high experimental temperature. To investigate the oxidation activity of soot, thermogravimetric analysis (TGA, STA449 F3, Netzsch GmbH) was performed on the loaded quartz filters with 4% oxygen in

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