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Understanding of physicochemical properties and formation mechanisms of fine particular matter generated from Canadian coal combustion

Chong Tian [a,*], Qingye Lu [b], Yuxi Liu [b], Hongbo Zeng [b], Yongchun Zhao [a,*], Junying Zhang [a], Rajender Gupta [b]

[a] *State Key Laboratory of Coal Combustion, Huazhong University of Science and Technology, Wuhan 430074, PR China*
[b] *Department of Chemical and Material Engineering, University of Alberta, Edmonton, AB T6G 2V4, Canada*

HIGHLIGHTS

- Systematic investigations on fine particles emissions from Canadian coals combustion were presented.
- The tri-modal mass size distributions was confirmed by the size-segregated elemental distributions.
- High volatile content in coal gives priority for the fine particle formation.
- Included minerals contribute greater for fine particles formation than the excluded minerals.

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ABSTRACT

A systematic study on the fine particular matter emissions from coal combustion of four kinds of coals of different ranks (lignite, sub-bituminous, bituminous, coking coal), including three typical Canadian coals and a Chinese coal have been presented. The results show that a tri-modal mass size distribution (coarse, fine and ultrafine mode) was commonly identified, and it was confirmed by size-segregated elemental distributions. The tri-modal distribution is of temperature and coal ranks independence. Coarse particles ($Da \sim 5$ μm) presented as round-shaped sphere with smooth surface. Both round-shaped particles and irregular-shaped particles were observed in fine particles ($Da \sim 0.5$ μm). Clusters of nano-particles ($Da \sim 0.05$ μm) were identified in the ultrafine mode. The concentration of fine particles ($Da < 1$ μm) generated from the TECK coal is the highest (about 150 mg/Nm^3), while fine particles ($Da < 10$ μm) generated from Boundary coal (about 580 mg/Nm^3) is highest among the four coals. Higher volatile content in coals gives priority for enhancing the formation of fine particles. Excluded minerals would mostly fragmented to form ash residual with irregular shapes, while part of them would experience coalescence individually to form round-shaped particles with size larger than 10 μm. Included minerals adhered firmly on the surface of char particles contribute greater in the irregular-shaped particles in fine mode, while the shedding included minerals from char particles should take the major responsibility for the round shaped particles in fine mode.

1. Introduction

Pollutant emissions from coal utilization have been causing irreversible environment disruptions in the past decades, and restrictions on emissions of the pollutants have been imposed by many countries. For instance, the particulate matter (PM) generated from coal combustion is of a great concern due to the serious risks to human health. The inhalable portions of the fine particulate matter have the capability to transport and get deposited in

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Abbreviations: PM, particular matter; PM$_1$, sub-micro particular matter ($Da < 1$ μm); PM$_{10}$, particular matter ($Da < 10$ μm); DTF, drop tube furnace; DLPI, Dekati Low Pressure Impactor; GC, Genesee coal; BD, Boundary coal; TK, TECK coal; YT, Yantang coal; XRF, X-ray fluorescence; BSEM, Back Scattered Electron Microscope; SEM-EDX, scanning electron microscope with energy-dispersive X-ray spectroscopy.

* Corresponding authors. Tel.: +86 27 8754 4779; fax: +86 27 8754 5526.
E-mail addresses: ctian@hust.edu.cn (C. Tian), yczhao@hust.edu.cn (Y. Zhao).

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the lung, and the toxic heavy metals enriched in the PM might be the causative agent for many diseases [1]. A comprehensive understanding of the properties and formation mechanisms of fine particulate matter is of both fundamental and practical importance in achieving a better solution for reducing the pollutant emissions [2].

Extensive research reports related to PM generated from coal combustion issues have been published. Physicochemical properties of PM have been completely presented, and mature theories regarding to the PM formation mechanisms have been built up. Three primary ash aerosol size modes would be commonly identified in the pulverized coal combustion [3,4]. While formation of PM in the three modes are led by a combination of various mechanisms. Classical theories regarding to the formation mechanisms of the PM generated from coal combustion include inorganic species vaporization, condensation, and aggregation [5,6], surface ash shedding during char combustion [6–8], bursting of cenosphere [9], agglomeration of the melting minerals [10,11], included minerals coalescence [12–15], char fragmentation [16–18], and excluded minerals fragmentation [12,19,20]. Most of the mechanisms can be attributed to the coal mineral properties [21,22] and the fragmentation behavior of coal/char at high temperature [23,24]. To sum up, these two aspects can be attributed to the coal ranks. Some previous studies indicated that the correlations between coal ranks and the yields of PM are ambiguous [25,26], and it is rather complicated to come to a unified conclusion. This is because that coal mineral matter is regularly deemed as a whole in many previous researches without distinguishing included and excluded minerals in coal. Actually, excluded minerals and included minerals would undergo distinct pathways prior to forming ash residuals [21]. Transformation behaviors of excluded minerals are majorly dominated by coal decomposition and oxidation reactions [22]. In contrast, included minerals would undergo a relatively complex process, e.g higher temperature [23,24], reducing atmosphere [27], etc. prior to exposing on the surface of char particles, which is with resultant from its close associations with the carbonaceous matter in coal. The devolatilisation and burnout behaviors of char particles would intimately impact on the shedding, aggregation, and coalescence of the included minerals. Herein, a clarification of contributions of included mineral species for PM formation is value of importance for better elucidating the formation mechanisms of PM.

Various kinds of mineral matter present in coal, and their modes of occurrence in coal are rather complex. Consequently, more or less discripencies of the PM emissions would always exist on various coal combustions in real cases. Hence, a comprehensive evaluation of PM emissions generated from coal combustion prior to utilization of the coal widely is essential, since it is helpful to predict the pollution level of the coal used in the local district. Goodarzi [28] has investigated the morphologies and chemistry of fine particles generated from a sub-bituminous coal combustion in a Canadian power plant, which provides us with valuable information on PM emissions from power plant in Canada. However, with increasing consumption of coal in Canada, the understanding of the PM emission generated from Canadian coal combustion is important and may provide guidance for the emission and pollution controls of coal combustion in Canada.

This paper presents a systematic study on the PM emissions generated from three kinds of Canadian coals, including Genesee coal (GC), sub-bituminous; Boundary coal (BD), lignite; TECK (TK), coking coal. A typical Chinese coal (Yantang coal (YT), bituminous) is also studied as a comparison. Influences of the external parameters, including combustion temperature and coal ranks, on the PM formation were detailed investigated, and the minor discrepancies caused by the parameters were also analyzed. Further, for understanding play in the PM formation, a detail process of the included minerals exposing, shedding and coalescence were presented and analyzed for better understanding the role of the included minerals. The overall mechanisms for PM formation in different modes were also analyzed and proposed.

2. Experimental

2.1. Experimental approach

The coal combustion experiments were carried out by using a laboratory scale of drop tube furnace (DTF) [29] as shown in Fig. 1, and three Canadian coals as well as a Chinese coal were chosen in the study. Basically, a screw feeder with pulsating walls and stirring rod in combination with a primary flow of N_2 was used to entrain the feeding coal into the reactor, the coal feeding rate was 0.3 g/min. N_2 and air as the secondary gas were firstly preheated to 400 °C before injecting into the reactor, and it quickly reached to the reaction temperature at a very short time. The ratio of primary gas to secondary gas was controlled at 1:9. The total flow rate of gas is 10 L/min, which was precisely controlled by mass flow controllers. The pressure inside the reactor was always negative during the experiment. The reaction temperatures were ranging from 1200 °C to 1400 °C with intervals of 100 °C, and the residence time was about 2 s. Coal ash samples were collected by a water-cooled collection probe with N_2 as the quench gas. Following the collection probe, the cooled stream was passed through a cyclone where char and ash samples, with diameter larger than 10 μm, were separated and collected. Afterwards, the fine particles with size less than 10 μm (PM_{10}) would pass through a Dekati Low Pressure Impactor (DLPI), in which aluminum foils greased with Apiezon-L grease and/or polycarbonate foils were installed on the stages to trap and segregate the fine particles by inertia force. The fine particles could be segregated into 13 stages with aerodynamic diameter ranging from 0.03 μm to 10 μm. In the system, a bypass went directly through a bag filter and a condenser to trap the particles and take out the remaining water vapor. After that the gas compositions were analyzed online using an Agilent micro-GC (CP-4900).

2.2. Samples characterizations

Three typical Canadian coals of different ranks (GC, sub-bituminous; BD, lignite; TK, coking coal) and a Chinese coal from southwestern China (YT, bituminous) were chosen for the study. All the coal samples were grinded and sieved into specific size fractions (45–75 μm), and vacuum dried at 105 °C prior to the combustion experiments. Fine particulate matters collected on the aluminum foil were used to evaluate the mass size distributions of the PM_{10} emissions. Particles collected on the polycarbonate foil were used to investigate the elements distributions. Element distributions of the PM_{10} of different sizes were determined by using the X-ray fluorescence (XRF). The micro-morphologies of the PM_{10} and char/ash were observed by scanning electron microscope with energy-dispersive X-ray spectroscopy (SEM-EDX), which were a JAMP-9500F (Jeol, Japan). All the experiments and measurements were conducted in the department of Chemical and Material Engineering in University of Alberta, Canada.

3. Results and discussions

3.1. Coal property

The coal property (proximate and ultimate analysis) and chemical compositions of the high temperature ash from the four coals were characterized and listed in Tables 1 and 2, respectively.