



Multimodal ultrafine particles from pulverized coal combustion in a laboratory scale reactor

Francesco Carbone^{a,*}, Federico Beretta^a, Andrea D'Anna^b

^a Istituto di Ricerche sulla Combustione, Consiglio Nazionale delle Ricerche, piazzale Vincenzo Tecchio 80, 80125 Napoli, Italy

^b Dipartimento di Ingegneria Chimica, Università degli Studi di Napoli Federico II, piazzale Vincenzo Tecchio 80, 80125 Napoli, Italy

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ABSTRACT

Particle size distribution functions have been measured in a ethanol fueled flame reactor fed with a low amount of pulverized coal particles. The reactor is operated in low (5.0 vol.%) and high (76.5 vol.%) oxygen concentrations using two high volatile bituminous Colombian and Indonesian coals. A carbon black powder is also oxidized in the same conditions. Generated particles are sampled using rapid-dilution probes and the size distribution functions are measured on-line by a high resolution Differential Mobility Analyzer. Results clearly show that ultrafine particles, those with sizes lower than 100 nm, have a multimodal size distribution function. These particles have huge number concentrations in both investigated conditions whereas their formation is enhanced in the oxygen enriched condition. Ultrafine particles are almost totally dominated in number by the fraction having sizes below 30 nm. Nanoparticles also account for a significant fraction of total particle mass and slowly coagulate in the reactor. The shape of the size distribution functions is not affected by the coal type, at least for the two investigated coals. Results suggest that ultrafine particles form through the vaporization–nucleation–growth pathway involving inorganic ashes. Moreover the contribution of carbonaceous particles seems particularly important for size smaller than 5 nm.

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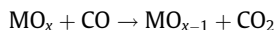
1. Introduction

The increasing demand of energy in the last decades, because of emerging countries such as China and India has been largely satisfied by coal combustion. Coal is also considered to be the major fuel for power generation in the next decades [1]. However, the main drawback of the extensive use of coal for power generation is greenhouse gas, primarily carbon dioxide, and pollutants such as nitrogen and sulfur oxides and particulate matter.

Up to recent times, air blown, atmospheric-pressure pulverized coal furnace has been the principal reactor design for coal combustion. Next generation coal power plants require carbon dioxide capture and sequestration (CCS) to be environmental sustainable [2]. Recently, oxy-combustion technologies are becoming widely used also because of the opportunity to retrofit existing air blown furnaces. Retrofitting of existing plants requires oxygen enriched concentrations (>28 vol.%) in recirculated flue gases to maintain the same heat flux [3].

Oxygen enriched environments in which coal is burned affect the main oxidation process as well as the formation of pollutants. Particulate emission from conventional pulverized coal combustion systems has been extensively studied [4,5] whereas the effects of oxy-

combustion conditions are still under investigation [6]. This is particularly true for ultrafine ashes whose formation, deriving from a vaporization–nucleation–growth pathway, is strongly affected by combustion temperature and oxygen concentration [7]. Indeed metals undergo vaporization and subsequently undergo homogenous nucleation or heterogeneous condensation, usually in the form of refractory oxides. The passage to the gas phase can occur directly, for volatile or organically bonded metals (M), or via reduction to volatile sub-oxides (MO_{x-1}), for refractory minerals (MO_x) [8]:



Vaporization occurs in the high temperature, fuel rich boundary layer surrounding the burning coal particles. It involves a complex chemistry and can be promoted by some elements, among all chlorine, that enhance the formation of volatile metal compounds [9]. Nucleation occurs when burning products move towards lower temperature and oxidizing conditions. This occurs, first, when escaping from the boundary layer surrounding coal particles and, subsequently, at the furnace exhaust where lower boiling point compounds condense. At higher cooling rates, homogenous nucleation is favored over heterogeneous condensation [9].

The process involving ash transformation is, to some extent, coupled with carbonaceous particle inception due to pyrolysis of tar and volatile organic compounds released during coal devolatilization [10].

* Corresponding author. Fax: +39 815936936.

E-mail address: francesco.carbone@irc.cnr.it (F. Carbone).

Coagulation is mainly responsible for particle growth whereas condensation and sintering greatly influence their morphology and surface composition [11].

Ultrafine ashes, those with sizes lower than 100 nm, represent just a small percentage of total fly ash mass at the exhaust of air blown pulverized coal furnaces [12] but it is of concern [13] because recent studies have shown that particle toxicity increases with decreasing particle sizes and strongly depends on particle number more than mass concentration [14]. Moreover, ultrafine particles are not easily removed by aftertreatment devices because filtering efficiency decreases with decreasing particle sizes in the ultrafine range [15]. Recent results obtained by using advanced diagnostics for particle size distribution measurements showed that at high temperature, particle coagulation efficiency dramatically drops with decreasing particle size below 10 nm. This behavior was observed both for carbonaceous [16] and metal oxide [17–19] particles so that nano-ash emission from pulverized coal furnaces might be a problem.

The aim of this paper is to measure the particle size distribution functions of ultrafine and nanosized particles formed in a laboratory scale, pulverized coal combustion reactor. The reactor was firstly operated in conventional conditions, typical of air blown pulverized coal combustion, and, subsequently, in extremely high oxygen concentration. The latter was selected to amplify the effects of oxygen enriched conditions on particle formation. Preliminary information on the nature and behavior of the ultrafine and nanometric fractions were obtained by comparing the results obtained in both conditions and using two types of coals and a carbon black powder.

2. Experimental apparatus

2.1. Flame reactor and operating conditions

A fuel lean flat laminar premixed flame operated at atmospheric pressure is used as reactor. The flame is sustained by droplets of ethanol, monodisperse in size and it is homogeneously doped with pulverized coal particles, not significantly modifying the flame stoichiometry. The reactor attempts to mimic local more than global conditions in furnaces and it allows investigating the kinetic of ash formation from single coal particle combustion in a well controlled environment.

The coals and the carbon black powder are first milled in a Planetary Mono Mill (Pulverisette 6, Fritsch) using tungsten carbide bowl and balls, and subsequently suspended in ethanol. The suspension, containing about 1.5 wt.% of coal/carbon black, is intensively sonicated to fragment agglomerated coal particles. More than 95% of the suspended particles have size smaller than 30 μm , as verified by dispersion granulometric analyses (Hydro 2000S, Malvern Instrument).

A Berglund–Liu-type Vibrating Orifice Aerosol Generator (VOAG model 3450, TSI), generating 80 μm monodisperse suspension droplets, is used to feed the flame. It supplies a constant flow of 0.30 cm^3/min through a 40 μm orifice oscillating at a frequency of 20 KHz. The VOAG syringe pump is immersed into an ultrasonic thermostatic bath at 40 $^\circ\text{C}$, to prevent particle sedimentation or agglomeration and the consequent orifice clogging. The resulting coal/carbon black flow rate, added into the ethanol flame, is of about 4.5 mg/min.

The suspension droplets, once generated, are suddenly dispersed and diluted in an oxidizing gas mixture with a flow rate of 2.3 l/min (STP), preventing their coalescence. The resulting gas/droplet mixture is fed to the burner.

The burner consists of two coaxial stainless steel tubes and it is similar to that used by Arabi-Katbi et al. [20]. The inner tube (18 mm ID) is used to carry the reacting mixture and a 40 mm long

Mullite Zirconia honeycomb (400CPSI, CTI s.a.) is placed on its top. Inner tube wall temperature was kept constant at 90 $^\circ\text{C}$ to prevent ethanol condensation. The ring (24 mm ID and 34 mm OD), supplied by the outer tube, has a flow of 9.0 l/min (STP) of sheath argon to minimize surrounding air perturbation and reduce flame flicker. A flat plate is also placed at 90 mm from the burner mouth.

Ethanol droplets completely evaporate through the honeycomb and the vapor burns stabilizing a flat premixed flame on the honeycomb mouth (cold gas velocity is 16.5 cm/s). Coal/carbon black aggregates, less than 20 μm in sizes, are homogeneously carried into the ethanol flame where they devolatilize, ignite and oxidize. The particle combustion environment can be adjusted by controlling the ethanol flame stoichiometry and dilution allowing coal/carbon black particles to react in different conditions in term of gas composition and temperature. Further details on the flame reactor are reported elsewhere [19,21,22].

Measurements have been performed using two high volatile bituminous Colombian and Indonesian coals whose properties are reported in Table 1. Ash composition and minor elements concentrations, measured by Agilent 7500 ICP-MS are also listed in Table 1. A Carbon Black powder (N762, Phillips Petroleum Co.) with primary particle size between 76 nm and 100 nm, has been also oxidized in the same conditions to evaluate the unburnt carbon and to analyze the role of carbonaceous particles in the build-up to the particle size distribution.

Table 1
Properties of the used Colombian and Indonesian coals.

| | Colombian coal | | Indonesian coal | |
|--|----------------|-------|-----------------|-------|
| | As received | Dry | As received | Dry |
| <i>Proximate analysis</i> | | | | |
| Moisture (%) | 4.90 | | 3.11 | |
| Volatile (%) | 38.31 | 40.28 | 40.20 | 41.49 |
| Ashes (750 $^\circ\text{C}$) (%) | 6.11 | 6.42 | 7.40 | 7.64 |
| Fixed carbon (%) | 50.68 | 53.3 | 49.29 | 50.87 |
| HHV (kcal/kg) | 6862 | 7216 | 7245 | 7478 |
| LHV (kcal/kg) | 6560 | 6957 | 6938 | 7198 |
| <i>Ultimate analysis, wt.%</i> | | | | |
| C | 70.95 | 74.60 | 71.68 | 73.98 |
| H | 5.32 | 5.03 | 5.62 | 5.44 |
| N | 1.32 | 1.39 | 1.38 | 1.42 |
| S | 0.40 | | 0.53 | |
| <i>Ash analysis, wt.% on ash basis</i> | | | | |
| SiO ₂ | 47.05 | | 44.47 | |
| Al ₂ O ₃ | 23.94 | | 22.28 | |
| Fe ₂ O ₃ | 5.99 | | 9.67 | |
| MgO | 1.43 | | 1.41 | |
| CaO | 3.29 | | 1.98 | |
| Na ₂ O | 2.03 | | 1.14 | |
| K ₂ O | 1.40 | | 2.31 | |
| MnO | 0.04 | | 0.04 | |
| <i>Minor elements in coal as received, ppm</i> | | | | |
| Ti | 176 | | 114 | |
| V | 21 | | 16 | |
| Cr | 7 | | 8 | |
| Co | 56.1 | | 24.5 | |
| Ni | 10 | | 8.7 | |
| Cu | 5.7 | | 9.7 | |
| Zn | 7.8 | | 13.7 | |
| As | 2.5 | | 1.9 | |
| Se | 4 | | 0.4 | |
| Mo | 2.1 | | 0.7 | |
| Sr | 71.8 | | 96 | |
| Zr | 9.6 | | 4.4 | |
| Cd | 0.1 | | 0 | |
| Sn | 0.2 | | 0.3 | |
| Ba | 241 | | 62.7 | |
| Hg | 2.7 | | 6.8 | |
| Pb | 2.3 | | 3.5 | |
| Cl | 101 | | 164 | |
| F | 16 | | 45 | |

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