Characterisation Of Aerosols From The Co-combustion Of Forest Biomass And Sewage Sludge In A Bubbling Fluidised Bed Reactor

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Abstract. In the present study, particulate emissions from the co-combustion of forest biomass residues with sewage sludge in a pilot-scale bubbling fluidised bed combustor were characterised. The combustion flue gas was exhausted to the atmosphere after passing through a cyclone separator. Physical-chemical characteristics of the particles were studied: *i*) morphology and aerosol size, surface and volume distributions before the cyclone and *ii*) chemical composition (carbonates, water soluble-inorganic ions, organic and elemental carbon) before and after the cyclone. Chemical composition data were used to calculate aerosol density and refractive index. Aerosols showed a unimodal size distribution with a geometric mean diameter of 2.25±0.02 μm and a geometric standard deviation of 1.27±0.01. The surface and volume mean diameters were 2.64±0.02 μm and 2.91±0.05 μm, respectively. Water-soluble inorganic ions were predominant in the fine particle fraction ($PM_{2.5}$). The filters were loaded of crystallised mineral particles. The analysis revealed a dominance of calcium carbonate/oxide and halide (NaCl or KCl), sulphate and aluminosilicate nanocrystals forming larger mixed aggregates.

Keywords: Aerosol, biomass, co-combustion, fluidized bed, sewage sludge.

INTRODUCTION

Valorisation of the industrial biomass wastes constitutes a sustainable alternative to improve safety in industrial waste management. Co-combustion, i.e. the simultaneous combustion of two or more fuels in the same plant for energy production, constitutes a promising option for valorisation of some industrial wastes. Among the technologies available for co-combustion, fluidised bed combustion (FBC) is one of the most advantageous due to its fuel flexibility and operating conditions [1]. FBC has become attractive in the pulp and paper industries as an alternative for energetic valorisation of some of their solid wastes [2]. Thus, forest residues and sewage sludge are generated in different processes in this industry. The co-combustion of these residues in fluidised furnaces offers a feasible alternative to manage these residues and at the same time, to generate income through energy recovery. However, attention must be paid to operating and environmental problems associated with this process. Focusing on air quality, particle and gas emissions acquire a particular relevance when sewage sludge is used as additional fuel [3]. The emission of fine particles constitutes an important challenge since the current air pollution control devices cannot effectively remove

them from the flue gas [4]. As numerous studies have pointed out, besides other impacts, these particles are particularly dangerous for human health because they are able to enter the respiratory system.

In this study, particulate emissions from the co-combustion of forest biomass residues and sewage sludge from a pulp and paper industry were characterised. The combustion experiments were conducted in a pilot-scale bubbling fluidised bed combustor.

MATERIAL AND METHODS

Fuel Characteristics

Forest biomass residues from eucalyptus felling and sewage sludge from the pulp and paper industry were used as fuel. Forest biomass residues were air dried, chopped and sieved in order to obtain particles in the size range of 1-5 mm. Sewage sludge was air dried and also sieved in the particle size range of 1-5 mm. The final fuel was composed of 40%wt of secondary sewage sludge and 60%wt of forest biomass residues.

Pilot-Scale Bubbling Fluidised Bed Combustor Characteristics

The combustion experiments were conducted in a pilot-scale bubbling fluidised bed combustor (BFBC) [5]. The reactor is a thermally insulated AISI 310 SS tube with an internal diameter of 0.25 m and 3 m height. The bed was operated with silica sand particles in the 0.25-1.00 mm size range. The combustion air was staged, with primary air through the distributor plate and secondary air through a vertical tube located inside the freeboard. The solid fuels were fed together with the secondary air, and discharged continuously at bed surface. Pressure, temperature and composition of the combustion flue gas were monitored by means of nine water-cooled sampling probes located at several heights along the reactor, two are immersed in the bed and the others are located along the freeboard. O_2 and CO_2 were also monitored. The flue gas was exhausted to the atmosphere after passing through a cyclone separator. Particle sampling was carried out at the exhaust duct, before and after the cyclone separator, with the reactor operating at steady state conditions.

Combustion Experiments And Operation Conditions

The FBC was operated with a bed temperature of 800±5 ºC, and with an excess air of 50%. The fuel was continuously fed to the bed at a feed rate of 4.2 kg h^{-1}. The combustion air was staged, with the primary air accounting to 80% and the secondary combustion air accounting to 20% of the total combustion air. A set of eight water-cooled probes located at the bed level allowed the control of the bed temperature at the desired value. The freeboard temperature was controlled by a heat exchanger operated with liquid water at a constant flow rate of 1 L min^{-1}. The temperature of the flue gases at the cyclone was around 150 ºC.

Regarding longitudinal temperature and pressure profiles along the reactor height, the temperature registers a minimum value at the reactor base (around 390 ºC) as it is measured in the fixed bed region. Then, it increases in the fluidised bed section and achieves a maximum value of around 970 ºC near the secondary air and fuel feeding location [5]. After that, the temperature decreases along the freeboard due to heat transfer to the reactor walls, water-cooled sampling probes, and heat exchanger located in the freeboard. The pressure ranges between 101 kPa and 103 kPa along the pilot-scale BFBC during the co-combustion experiment. The CO_2 concentration in the exit flue gas is around 14%v (dry gases) and that of O_2 around 6.5%v (dry gases).

Particle Sampling And Measurement Techniques

The FBC was operated under pre-set steady-state conditions, and the particle samples were collected under isokinetic conditions. Two different systems were used for particle sampling: a) a low volume sampler unit to collect $PM_{2.5}$ onto quartz filters directly from the inlet and outlet cyclone ducts, and b) a Venturi system that allows the sampling of the combustion flue gas at 10.0±0.3 L min^{-1} (at atmospheric pressure and temperature) directly from the inlet duct of the cyclone by using dry filtered compressed air (100±3 L min^{-1}, at atmospheric pressure and temperature). Two different instruments used this flue gas flow for particle sampling: i) an optical particle counter (OPC), for the continuous monitoring of particle size distributions and ii) a Gent stacked filter unit PM_{10} sampler using polycarbonate filters aiming to determine the morphological characteristics of the particles by means of Scanning Electron Microscopy (SEM). As a consequence of practical limitations related to relatively high under-pressure operation downstream the cyclone, it was not possible to operate the Venturi sampling system at this location. Consequently, it was not possible to get information from the OPC and Gent stacked filter unit downstream the cyclone.

Analytical Methodologies

The particulate matter mass deposited on the filters was quantified by gravimetry. Filters were analysed by different techniques in order to determine the concentrations of carbonaceous content and soluble inorganic ions. Organic and elemental carbon (OC and EC) were analysed using a thermal-optical transmission technique [6], after a previous sample acidification to remove carbonates. The carbonate fraction was determined by sample acidification with phosphoric acid. The major soluble inorganic ions (Cl^-, NO_3^-, PO_4^{3-} y SO_4^{2-}, Na^+, NH_4^+, K^+, Mg^{2+} y Ca^{2+}) were determined by ion chromatography in liquid phase. A set of blank filters was analysed to correct the obtained values.

RESULTS AND CONCLUSIONS

The $PM_{2.5}$ samples presented a mean concentration of 255±16 mg Nm^{-3} and 220± 30 mg Nm^{-3} before and after the cyclone. Percentage Total carbon (TC)/$PM_{2.5}$ mass ratios of 6.9%wt and 1.9%wt, before and after the cyclone, were obtained. The TC is

mainly composed of OC, representing mean percentages of 97%wt and 69%wt before and after the particle separator.

Water-soluble inorganic ions were the dominant $PM_{2.5}$ mass fraction (>50%wt), with potassium, sulphate and chlorine being the most abundant. OC, EC and carbonate registered mean concentration values of 17±3, 0.5±0.2 y 21±4 mg Nm^{-3}, respectively, before the cyclone (Fig. 1).

Aerosol size distribution and morphology of aerosols were studied before the cyclone. The bulk composition of the particulate matter samples was used to calculate aerosol density and refractive index [7]. A unimodal particle size distribution was observed, with a mean geometric diameter of 2.25±0.02 µm and a geometric standard deviation of 1.27±0.01. The number concentration of particles with size lower than 1 µm was relatively low. The surface and volume mean diameters of the particle distribution were 2.64±0.02 µm and 2.91±0.05 µm respectively.

Emissions factors of $3.7 \cdot 10^{11}$ particles ($PM_{0.1\text{-}12}$) kg^{-1} fuel (as burned) and 4.0±0.2 g$PM_{2.5}$ kg^{-1} fuel (as burned) before the cyclone and 3.7±0.5 g $PM_{2.5}$ kg^{-1} fuel (as burned) after the cyclone were estimated.

The morphology of the aerosols present in the flue gas from FBC of biomass and sludge has been analysed by SEM. Particles are mainly composed of mineral crystals of mean diameter of 150 nm forming larger aggregates. The chemical composition of those nanometric crystals is complex, revealing a dominance of calcium compounds, mainly carbonates and oxides (that contain less than 5%wt Mg), and sodium/potassium chlorides, calcium sulphates and silicates in a lesser extent. The presence of needle-fibre elongated crystals was remarkable. Considering that non-biogenic micro-rods are rarery found in atmospheric samples, and those structures might have direct implications on lung disease, this finding is of special interest and requires further study.

Based on $PM_{2.5}$ concentrations determined by gravimetric analysis (255±16 mg Nm^{-3} and 215±30 mg Nm^{-3} before and after the cyclone, respectively) it seems that the cyclone doesn't play an important role in removing particles with an aerodynamic diameter less than 2.5 µm; in fact, it is recognised the relatively low removal efficiency of cyclones for particulate matter on this size range. In this context, a similar aerosol size distribution and morphology of $PM_{2.5}$ could be expected after the cyclone, when compared to that observed upstream the cyclone.

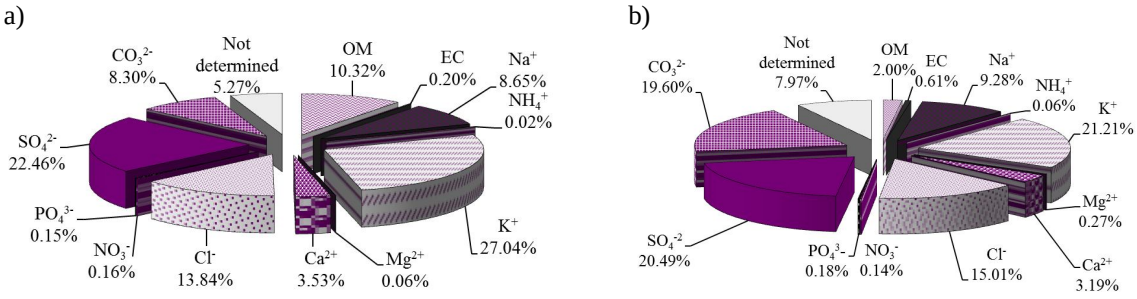

FIGURE 1. Aerosol composition (%wt) registered a) before and b) after the cyclone. Organic matter (OM) has been estimated multiplying the OC concentration by a factor of 1.55.

PERSPECTIVES

Future studies will focus on: a) study the impacts of aerosols from co-combustion on climate, health, ecosystems, etc; b) investigate the origin and formation conditions of calcium needles; c) study the hygroscopicity and activation ability of calcium oxides and calcium carbonate; d) quantify the nanocrystals formation; e) application of climate change models based on non-spherical particles; f) test other air pollution control devices such as catalytic filters or electrostatic precipitators; g) determine the speciation of the organic fraction of aerosols; h) test different percentages of forest biomass and sewage sludge as fuel; i) evaluate the effect on particulate matter emissions of recycling a fraction of bottom bed after treatment (water leaching).

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REFERENCES

1. Rink, K.K., Kozinski, J.A., Lighty, J.S. *Comb.Flame* **100**, 121–130 (1995).
2. Oral, J., Sikula, J., Puchyr, R., Hajny, Z., Stehlik, P., Bebar, L., *J. Clean. Prod.* **13**, 509–515 (2005).
3. Shao, J., Yan, R., Chen, H., Yang, H., Lee, D.H., Liang, D.T., *Energy Fuels* **22**, 2278–2283 (2008).
4. Yao, Q., Li, S.Q., Xu, H.W., Zhuo, J.K., Song, Q., *Energy* **35**, 4480–4493 (2010).
5. Tarelho, L.A.C., Neves, D.S.F., Matos, M.A.A. *Biomass Bioenerg.* **35**, 1511–1523 (2011).
6. Alves, C., Vicente, A., Nunes, T., Gonçalves, C., Fernandes, A.P., Mirante, F., Tarelho, L., Sánchez de la Campa, A.M., Querol, X., Caseiro, A., Monteiro, C., Evtyugina, M., Pio, C., *Atmos. Environ.* **45**, 641–649 (2011).
7. Levin, E.J.T., McMeeking, G.R., Carrico, C., Mack, L., Kreidenweis, S.M., Wold, C.E., Moosmüller, H., Arnott, W.P., Hao, W.M., Collett, J.L., Malm, W.C. *J. Geophys. Res.* **115** (2010).

PUBLICATIONS

A part of this work was presented at the European Aerosol Conference held in Granada (Spain), in September 2012 (Particulate emissions from the co-combustion of forest biomass waste and sewage sludge in a bubbling fluidised bed) and has been published in the international journal *Fuel Processing Technology* (Particulate emissions from the co-combustion of forest biomass and sewage sludge in a bubbling fluidised bed reactor, 114, 58-68 (2013)).