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Fine Particles Emission from Controlled Combustion of Beech Wood in Laboratory Conditions

Michal Špiláček[a,*], Jan Poláčik[b], Jiří Pospíšil[a], Ladislav Šnajdárek[b], Tomáš Sitek[b]

[a]Sustainable Process Integration Laboratory – SPIL, NETME Centre, Brno Unversity of Technology, Technická 2896/2, 61669 Brno, Czech Republic
[b]Energy Institute, NETME Centre, Brno University of Technology, Technická 2896/2, 61669 Brno, Czech Republic
michal.spilacek@vutbr.cz

This paper deals with the laboratory scale identification of the formation and growth of fine particles during the temperature increase of beech wood samples. The utilized experimental procedure combines the thermogravimetric analysis and identification of particles in flue gas leaving the thermogravimetric analyser (TGA). The aerosol stream leaving TGA enters a Scanning Mobility Particle Sizer (SMPS) where the particle size fractions are separated. The number of particles is identified by the condensation particle counter (CPC). The parametrical study was carried out to assess the composition of the atmosphere on the production and size distribution of fine particles during a heating of beech wood samples. There is clear evidence that the increase of oxygen content in the atmosphere generally results in the lower mass concentration of produced particles.

1. Introduction

Monitoring of air pollution has in the recent years focused on particulate matter (PM) because the negative impact of very and extremely fine particles has been proven by a number of studies. These particles are dangerous to human health, Santibáñez-Andrade et al. (2017), due to their size lower than 1 micrometre (PM1) which is under the minimum size for the lung's self-cleaning ability to work. The smallest particles can even diffuse through the wall of alveolar sacs into the bloodstream.

The extremely fine (or ultrafine) particles are produced during combustion processes. Their creation is connected to an elevated temperature and the occurrence of suitable chemical components. The main producers of ultrafine particles in urban areas are internal combustion engines and small furnaces. Most of the particles produced by the combustion process are PM1 (particles with a diameter of less than 1μm).

The great density of residents in cities causes a greater number of cars and small furnaces in agglomerations. The production of ultrafine particles from those sources is significant and its hazards are given by the conditions of dispersion. Unfortunately, the city buildings are a great obstacle for the air flow at the ground level and create conditions for a long residence time of particulates. Ultrafine particles don't sediment and remain in the environment for days and weeks until they are separated from the air by touching a solid or a liquid surface like walls, roads, vegetation, water bodies or raindrops.

This paper is focused on the study of emissions of very and extremely fine particles from combustion processes occurring in small biomass combusting furnaces. There is a vast variety of those furnaces and in each of them are different combusting conditions. Some parts of the furnaces have intense combustion air flow that causes flying of ash. Other parts are intensively heated by exothermic reactions. There are also parts with a high concentration of volatiles that undergo incomplete combustion caused by a lack of combustion air which subsequently cool down and condensate, leading to nucleation of ultrafine particles.

The measurement of the ultrafine particles contained in the flue gases from actual furnaces is important for identification of effects of operating parameters (power output, a surplus of combustion air, temperature) on the resulting concentration of the particulates. Such measurements are also used for comparison of different fuels in one furnace, Kantova et al. (2018). But these experimental measurements don't provide detailed insight of sub-stages of the process of nucleation and subsequent agglomeration of particles. It is important to experimentally recreate that process in laboratory conditions for combustion of samples of fuels. This

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experimental research is the main focus of this paper. For the identification of the particles emitted from a small specimen is used a tandem of thermogravimetric analysis and identification of particle size distribution of particulate matter. It is very important to understand the inception and creation of these particles in the combustion process to create viable strategies for their elimination.

2. Thermal Decomposition of Wood

The solid phase of woods affected by heat flux can be divided into three layers. These are the char layer, pyrolysis layer and virgin wood. Char front is defined as the transition between the char layer and the pyrolysis layer. It is the zone where thermal degradation of wood and char formation occurs. This transition is usually considered to take place at the 300 °C isotherm, called the char-line, Cachim and Franssen (2009).
When external heat flux affects the wood surface, part of the heat is reflected from the surface. Convective heat transfer between surrounding gases and wood surface also occurs.
In the pyrolysis layer, water evaporation occurs together with pyrolysis reactions and production of gas volatiles. Vapour and gas volatiles penetrate through pores and leave the wood.

3. Particulate formation

Released volatile components have different values of partial pressure. After reaching the saturation point, the formation of a new phase begins. We call it the nucleation process. The molecules are clustered into ultrafine particles up to 0.1 μm in size. The resulting particles can be further enhanced by coagulation mechanisms (aggregation of colloidal and macromolecular organic particles into larger aggregates), by agglomeration (bonding based on the adhesion of the surfaces), and by oxidation reactions or condensation on the particle surface, Obaidullah et al. (2012). Particles that are formed in the combustion chamber are called primary particles. Secondary particles are formed in the flue gas duck and in the atmosphere. According to volatility, the resulting organic compounds are divided into volatile organic compounds (VOC) and semi-volatile organic compounds (SVOC), Sippula (2010).

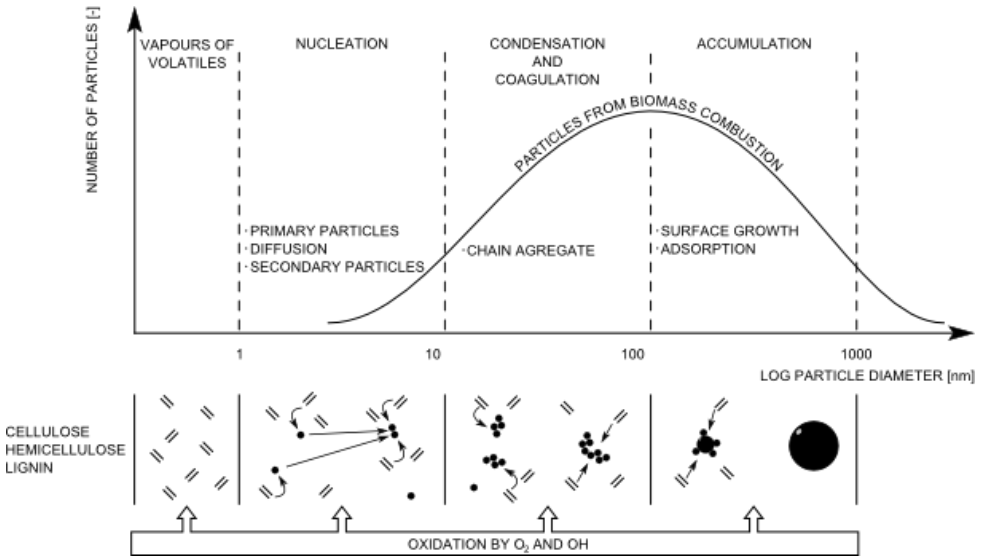

Figure 1: Nucleation and growth of combustion particles

3.1 Soot

Soot is a spherical particle of impure carbon coated in PAHs, Battin-Leclerc et al. (2013). These pollutants are usually the result of a fuel-rich combustion in high temperature (over 900 °C) area. Combustion in low temperature (under 700 °C) area results in CO, VOC and smoke species and soot coated in O-PAH, Williams et al. (2012). Production of soot can also occur in fuel lean conditions if some parts of the fuel cannot be accessed by the combustion air both in the fuel bed and in freeboard flue gas.
Modelling of soot formation is based on experimental work on laminar flames of simple and clean fuels like ethylene and methylene and to date, no model is describing the formation precisely. But there is agreement on the basic mechanism of the soot formation, and a mechanism known as HACA, that also describes the evolution of PAHs, manages to well describe the nucleation of soot, Desgroux et al. (2017). For the description of this

mechanism was created a basic chemical and physical frame. In the chemical frame, the main precursor of soot formation is considered acetylene C_2H_2 that is reacting to create higher hydrocarbons that are the basis of PAH, Saini and De (2017). And once PAH is formed, the physical frame can be used to describe the rest of the formation that consists of 4 steps: formation (nucleation), coagulation, condensation and surface growth, Saini and De (2017). Coagulation and condensation are shown in Figure 1. These two steps are creating agglomerates that after further reactions turn into impure carbon nuclei. Oxidation by O_2 and OH appears at all steps, Desgroux et al. (2017). The main source of these pollutants is incomplete combustion and is controllable to some degree, because in grate biomass combustion pyrolysis always takes place, by the quality of combustion process.

3.2 Fly ash

Biomass generally contains trace amounts of metals and the most important of them is potassium, Williams et al. (2012). These elements are released during combustion and create compounds responsible for fouling the equipment after condensing on its walls. But some of these compounds leave the combustion equipment in the form of aerosols that are dangerous for human health by themselves or create particles of fly ash in the similar fashion as the soot particles are formed and these two mechanisms are connected.
The inception and growth of ultrafine particles are very complex processes in which it is complicated to quantify the intensity of individual incremental changes. A certain help in the quantification is numerical modelling, but despite the great advances in development of models and greater computational resources it is not yet possible to account for every process and create an exact model of the of the particulates inception and growth. And models of biomass combustion mostly do not account for particulates, Abricka et al. (2016).

4. Experimental procedure and measurement device

From the previously stated reasons, the focus of this paper is on laboratory measurements of the size distribution of ultrafine particles emitted from combustion of beech wood. The laboratory procedure which was developed combines the advantages of thermogravimetric analysis with the detailed monitoring of the size distribution of produced fine particles. Thermogravimetric analysis (TGA) allows monitoring the exact temperature influence of a small fuel sample according to the desired schedule. TGA also influences the composition of the atmosphere flowing around the sample. During the test, the TGA monitors the weight of the heat-affected sample and identifies its loss. From the analysis of the development of temperature rise, it further identifies the presence of endothermic and exothermic reactions.
The measurement was carried out with utilizing the TGA device NETZCH – Jupiter F3. The base component of the STA-449 Jupiter analyser is a very precise digital weighing system with the vertical design. The weighting system is connected to a shielded ceramic module (TG-module) into which the analysed samples are placed. During the measurement, the entire module is inserted into a gas-tight laboratory furnace with a controlled heating rate. The result of the measurement is a TGA curve showing the weight change in dependence on the temperature of the sample.
The gaseous components released from the fuel sample during the TGA are scattered in the test atmosphere and removed from the device. Subsequent flow through the connecting pipeline cools the stream to the ambient temperature. The cool aerosol stream enters a Scanning Mobility Particle Sizer (SMPS) where the particle size fractions are separated.
Nearly every particle has some level of electric charge. Electrostatic classifier requires aerosol to achieve a steady state of charge distribution. This is achieved by Aerosol Neutralizer device, which provides discharging and charge neutralization process. After this process, particles pass through a bipolar charger and reach equilibrium charge level on the particles. Then, aerosol flows into the electrostatic classifier (DMA) where sizing occurs (Figure 2).
In a particle sizing system, the Electrostatic Classifier separates particles by size for high-resolution measurements of particle size distribution. Scanning Mobility Particle Sizer (SMPS) allows classifying particles in the range from 10 to 1000 nanometres in diameter. Differential mobility analyser (DMA) selects flowing particles of a specific size from a polydisperse aerosol. Particles are separated according to their electrical mobility. As a result, there is a highly monodisperse aerosol containing particles of a certain size. Monodisperse particles leaving the electrostatic classifier continue to a Condensation Particle Counter (CPC). This device measures particle number concentration. A number of particles in the particle-size fractions are identified by the condensation particle counter (CPC).
In the CPC, particles pass through a heated saturator in which Butanol is vaporized. The aerosol together with evaporated Butanol flows into a cooled condenser. Particles, as condensation nuclei, get enwrapped by the supersaturated Butanol vapour. They become larger droplets so they are easily counted passing through an optical detector, Kantova et al. (2018).

The production of fine particles during the process of burning the wood sample was measured by TSI-SMPS device (Model 3080-Series Electrostatic Classifiers including CPC 3775, TSI Inc.).

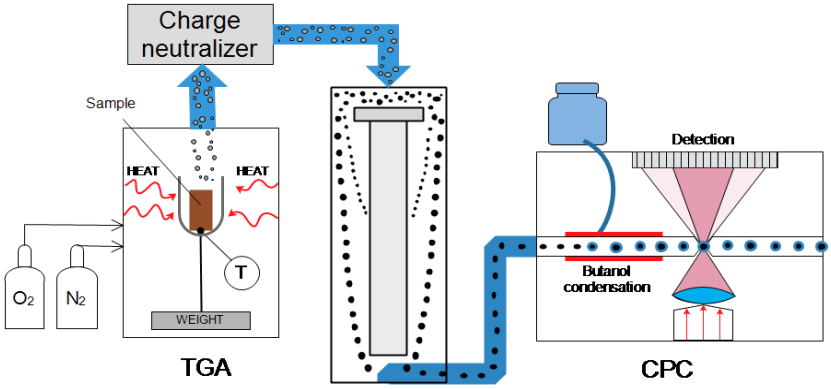

Figure 2: Scheme of the measuring equipment and the particle flows

5. Experimental measurement and discussion

During the combustion process, many of its parameters change with temperature and combustion air surplus being the most changing depending on location in the combustion chamber. For the research of inception and growth of the very and extremely fine particles in the flue gas was used a measuring apparatus described in the previous section. For the specimen were used parts of beech wood made into blocks each weighing 80 mg which were during the experimental procedure heated up in the thermogravimetric analyser from 20 °C to 620 °C over the course of 120 min by a constant +5 °C/min temperature change. To simulate the wide spectrum of possible conditions of the combustion process the samples were heated up in atmospheres with different oxygen concentrations, namely with the concentrations of 0 %, 5 %, 15 % and 20 % of oxygen. Each atmosphere was prepared by mixing pure oxygen and pure nitrogen from pressurised vessels connected to the TGA device.

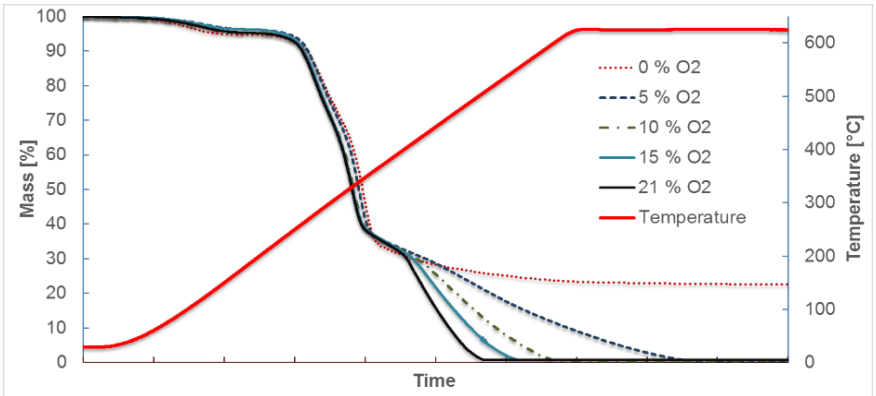

Figure 3: Record from the thermogravimetric analysis of beech samples in different atmospheres

A mass loss is occurring during the controlled heating up the samples as shown in Figure 3. This figure presents the temperature pattern of wood samples (increasing curve) and the associated weight loss of the samples (decreasing curves). During temperature increase up to 120 °C, water evaporates from the tested samples. In the interval 120 °C to 250 °C, light volatile compounds are released. The interval 250 °C to 300 °C is associated with the rapid loss of samples weight induced by the intensive release of other volatile compounds. Exothermic reactions are commenced within these temperatures. The associated heat generation promotes the release of other volatile compounds. The interval 300 °C to 500 °C is associated with the gradual charcoal decomposition of carbon. The relations presented at the Figure 3 show the different intensity of incineration of samples in different atmospheres.

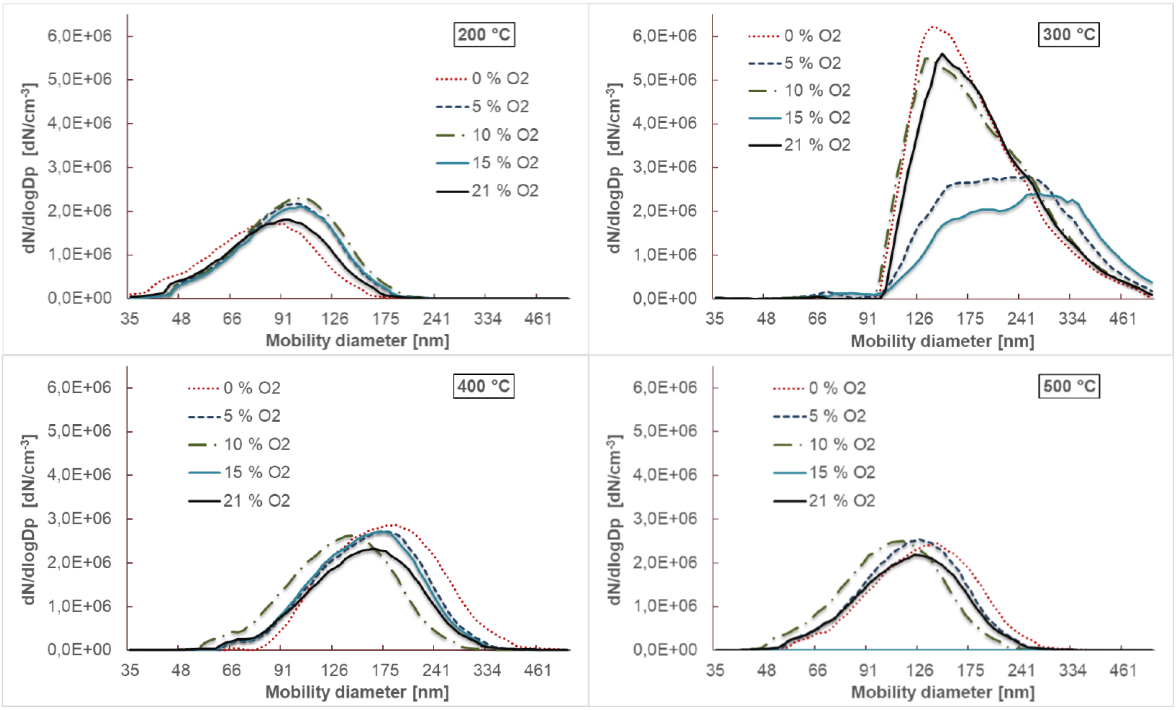

Figure 4: The distribution curves of fine particles emitted at the different temperature levels

The thermal decomposition of the beech wood samples is influenced by the intensity of the oxidation reactions. The result of the thermal decomposition is a production of ultrafine particles scattered in the gaseous products leaving the TGA apparatus. The Figure 4 presents the obtained distribution of generated particle concentration at the temperatures 200 °C, 300 °C, 400 °C and 500 °C for all tested concentrations of oxygen in the atmosphere. At the temperature of 200 °C very significant concentrations of ultrafine particles in the range of the particle mobility diameter of 45 nm – 180 nm. The maximum concentration of particles was identified for the particle size close to 100 nm.

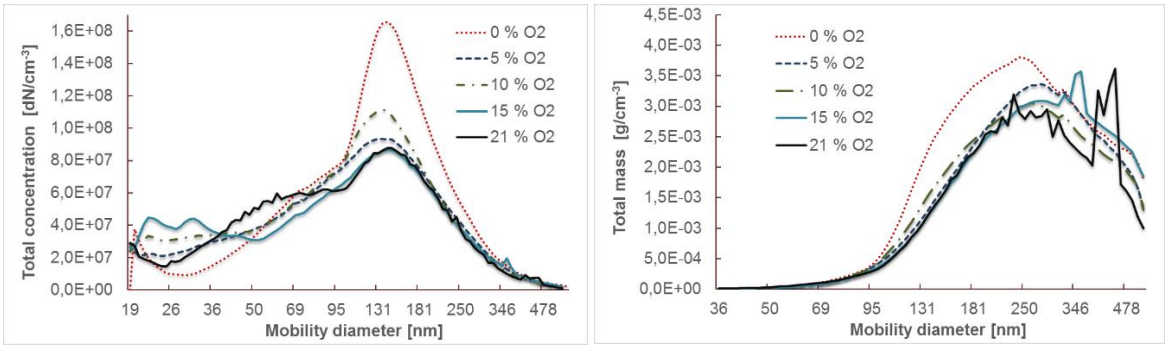

Figure 5: The relation between the total particles concentration and the mobility diameter of particles

At the temperature of 300 °C were measured the highest concentrations of particles. This temperature is close to the ignition temperature of the samples. For this temperature is characteristic an intense production of volatile compounds that subsequently lead to nucleation of a great number of particles. This high concentration of nuclei causes an intense coagulation of particles that leads to the growth of bigger particles.
The concentrations at the temperatures of 400 °C and 500 °C are close to the concentrations measured at 200 °C. Significant concentrations of the ultrafine particles occurred within the particle mobility diameter of 50 nm – 340 nm and the maximum concentration corresponds to particle size around 130 nm. With the increasing temperature, the production of particles diminishes due to a lower concentration of volatile compounds in the atmosphere.

The Figure 5 (right) shows the total concentration of particles for the entire temperature range of the samples from 20 °C to 620 °C. It shows the general maximum mass concentration of particles for particle mobility diameter of 135 nm. The Figure 5 (left) presents the total mass concentration as a function of the mobility diameter. The maximum Total mass concentration was identified for particle size close to 250 nm. With an increase of oxygen concentration in the atmosphere the total mass concentration of particles decreases.

6. Conclusions

The very and extremely fine particles produced by combustion processes represent a significant health risk. The entire process of inception and growth of these particles is very complex and still not sufficiently described. The study of this issue at actual combustion devices is very difficult due to the instability and diversity of the combustion process for diverse types of devices and fuels. In this paper is presented a research focused on the emission of ultrafine particles from a controlled heating up of small size samples of beech wood. A connection of a thermogravimetric analysis and a mobility diameter measurement of particulates was realised for this purpose. The carried out the study was a parametric sweep tracking the influence of different concentrations of oxygen in the atmosphere on the production of particulates during a controlled heat up.

From the carried out experimental measurement can be concluded that increasing temperature leads to a higher emission of particles produced by the combustion process. This applies until the ignition temperature is reached. This is most likely due to the increasing concentration of volatile vapours. Just before the ignition temperature the highest amount of volatile vapours is evaporating from the sample which leads to the most intensive nucleation of particles resulting in their high concentrations, coagulation and growth. At the moment the ignition temperature of the volatile vapours is reached, which can only happen is oxygen is present, the concentration of these vapours diminishes and so does the production of the particulates. This diminishing process occurs until the maximum experimental temperature of 620 °C.

The utilisation of the thermogravimetric analysis seems to be suitable for experimental measurements of particulates production from combustion and a suitable tool for detailed insight into the sub-processes of the particulates inception and growth during the combustion process.

Acknowledgments

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