CHEMICAL ENGINEERING TRANSACTIONS Volume 17, 2009
Editor Sauro Pierucci
Copyright © 2009, AIDIC Servizi S.r.l., ISBN 978-88-95608-01-3 ISSN 1974-9791
DOI: 10.3303/CET0917122

Effect of Process Parameters on Ultrafine and Fine Particle Emissions

Erkki Lamminen, Henna Isherwood
Dekati Ltd.
Osuusmyllynkatu 13 33700 Tampere, Finland

Industrial processes such as combustion, high temperature manufacturing and application of nanotechnology are potential sources of ultrafine and fine particles. Particle generation and subsequent particle transformation during these processes can be strongly affected by changes in the process parameters. Due to the growing number of health studies showing a correlation with exposure to particles and adverse health effects, an understanding of particle generation and transformation is important.

In this study, we take a look at small scale biomass combustion, welding and generation of nanoparticles with a focus on the effect of process parameters on the produced particles in terms of particle size and concentration.

The studied particle generation processes display the unstable nature of particle formation and the difficulty to predict the emission of particles. As a general guideline it can be said that an efficient and stable process causes less particle emissions compared to a transient, inefficient process. Characteristics of the emitted particles are observed to change over time. Therefore, when the potential health effects from these processes are considered, the transformation of the particulate emission before exposure should be taken into account.

1. Introduction

Ultrafine and fine particles are formed in a wide range of industrial processes. High energy processes such as combustion and high temperature manufacturing are typical sources of ultrafine and fine particle emissions. In addition to these processes where the particles are usually an unwanted by-product, recent development of nanotechnology depends on the generation of engineered ultrafine particles.

Ultrafine particles typically form through nucleation from supersaturated vapor. This process is extremely sensitive to temperature, vapor concentration and depends on the background particle concentration. After nucleation the ultrafine particles grow into accumulation mode (fine) particles through condensation and agglomeration. Due to the unstable nature of the particle formation, changes in an industrial process can have a major effect on the particle emissions. A common example from combustion is if the

Please cite this article as: Lamminen E. and Isherwood H., (2009), Effect of process parameters on ultrafine and fine particle emissions, Chemical Engineering Transactions, 17, 729-734 DOI: 10.3303/CET0917122

quantity or quality of fuel or combustion temperature changes, it will most probably result in a shift in particle size and concentration. This example can be extrapolated to different processes so that if the amount or properties of the vaporized material or the process temperature change, it will likely cause the particle emissions to change.

In this paper we show how particle emissions from small scale biomass combustion, welding and generation of nanoparticles shift due to changes in process parameters. The particle emissions are measured in real-time and analyzed for particle size and particle concentration. Measurements are carried out with the Electrical Low Pressure Impactor (ELPI™, Dekati Ltd.) and a two stage dilution system is applied in all measurements to condition the particle sample.

2. Experimental Methods

A similar measurement setup was used in all measurement locations. A raw sample was extracted with a probe and led into a two-stage dilution system. The diluted sample was then led from the dilution system directly into the ELPI™. All known changes in the measured processes were carefully documented for data analysis purposes.

2.1 Electrical Low Pressure Impactor (ELPI™)

The ELPI™ operating principle can be divided into three major parts; particle charging in a unipolar corona charger, size classification in a cascade impactor and electrical detection with sensitive electrometers. The particles are first charged into a known charge level in the charger. After charging the particles enter a cascade low pressure impactor with electrically insulated collection stages. The particles are collected in the different impactor stages according to their aerodynamic diameter, and the electric charge carried by particles into each impactor stage is measured in real time by sensitive multichannel electrometers. This measured current signal is directly proportional to particle number concentration and size Keskinen et al. (1992), Marjamäki et al. (2000), Virtanen et al. (2001).

The particle collection into each impactor stage is dependent on the aerodynamic size of the particles. Measured current signals are converted to (aerodynamic) size distribution using particle size dependent relations describing the properties of the charger and the impactor stages. The result is particle number concentration and size distribution in real-time. The operation principle is presented in Figure 1.

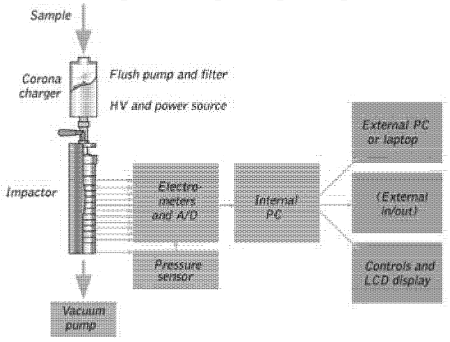


Figure 1. Electrical Low Pressure Impactor operation principle

2.2 Two Stage Dilution System

A two stage dilution system was used to condition the sample during all measurements. The first dilution stage was always heated to the raw sample temperature, thus lowering the vapor pressure of volatile compounds and water vapor. This enables the use of room temperature dilution in the second stage to reduce the temperature of the sample while ensuring that the volatile vapors and water do not nucleate into ultrafine particles. The residence time within the dilution system is approximately 1 second and therefore agglomeration within the diluter can be considered to be negligible. Particle losses in the diluters have been determined to be below 5% for a single dilution stage, and are not taken into account in this study Samaras et al. (2005).

2.3 Measurement Locations

Measurements for this study were carried out at a biomass power plant, welding station and at a nanoparticle flame spray oven.

The biomass power plant was a 7 MW wood firing plant that produced energy for district heating. The plant was equipped with gasification and flue gas recirculation to reduce both particulate matter and nitrogen oxide emissions. Effects of addition of fuel, grate movement and adjustment of flue gas recirculation are presented in this study. Recirculation of the flue gas improves the gasification of the fuel by increasing the air flow in the gasification chamber which stabilizes the gasification temperature.

Particle emissions from welding were studied at a MIG welding station. Sampling point for the emission measurement system was situated at the welder breathing area. Effect of welding process on the particulates and the effect of particle transport from the point of welding to the breathing area are presented in this study.

Nanoparticle flame spray was studied for the effect of different spray liquids and for the effect of operating related equipment such as spray oven access ports and substrate transport sleds inside the oven.

3. Results and discussion

3.1 Biomass Power Plant

Typical particle emissions with and without flue gas recirculation from the biomass power plant are presented below in Figure 2.

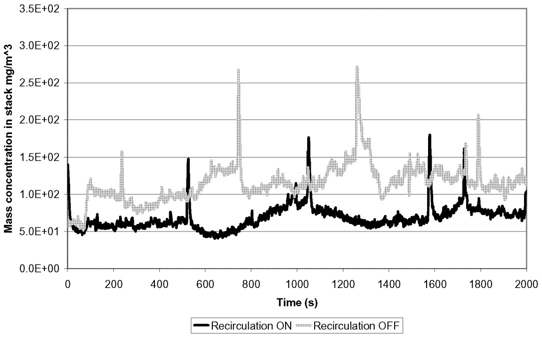
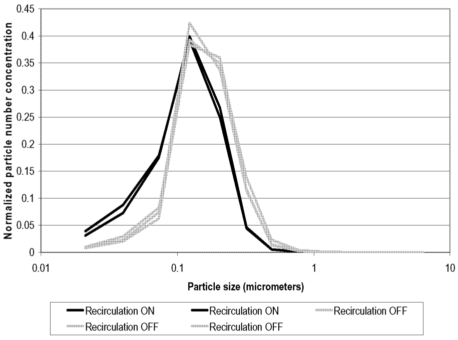


Figure 2. Mass concentration in flue gas with and without re-circulation

Typical particle mass concentration in the flue gas is in the range of 120 mg/m^3 with recirculation off and approximately 30-40% lower with recirculation on. The recirculation of flue gas stabilizes the gasification temperature and eliminates cold spots in the gasification chamber that produce particles and particle precursors.

Normalized particle number distributions with recirculation on / off can be seen in Figure 3 and distributions from stable and unstable operating conditions can be seen in Figure 4. The unstable operation conditions are caused by addition of wood into the gasification chamber and by grate movement, which removes ashes from the gasification chamber.



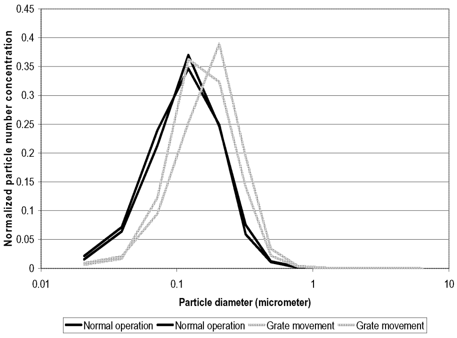


Figure 3 (on the left). Normalized particle number distributions with recirculation on and off

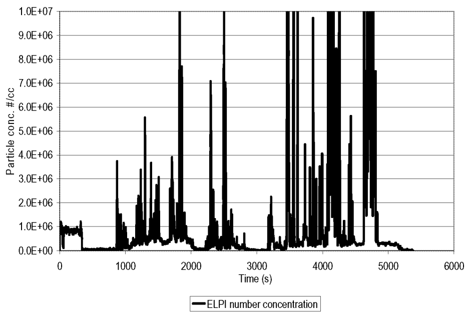
Figure 4. (on the right) Normalized particle size distributions during normal operation and grate movement

As can be seen in Figure 3, turning recirculation of flue gas off and on causes a repeatable change in the particle emissions, while grate movement and addition of wood cause an unstable distribution of generally larger particles when compared to the original distribution.

3.2 Welding

Particle number emissions measured from the worker breathing area during MIG welding can be seen in Figure 5. Particle number emission fluctuates radically with the start and stop of MIG welding. Also during continuous welding the number concentration is highly unstable due to changes in wire-metal distance which causes large temperature gradients.

Particle number distributions from MIG welding can be seen in Figure 6. The large differences are caused by different transport times from the welding spot to the measurement point and by different initial concentration of primary particles thus causing different agglomeration speeds.



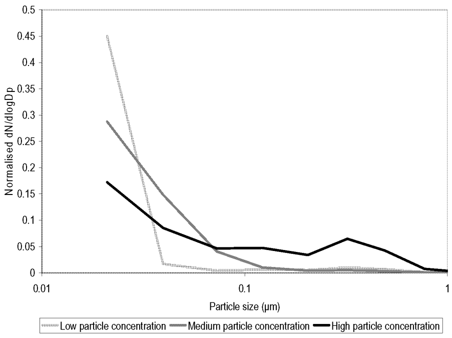
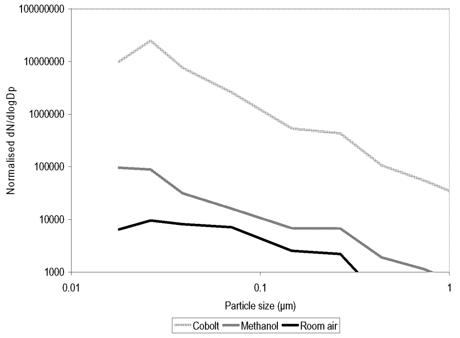


Figure 5. (on the left) Particle number concentration from welding,
Figure 6. (on the right) Particle size distributions with different initial particle concentrations

3.3 Flame spray of nanoparticles

Particle distributions from flame spraying of methanol and cobalt can be seen in Figure 7. In addition, indoor air particle distribution outside the oven is also plotted into the same Figure for comparison purposes. Particle measurements from inside, from the tail and from beside the flame can be seen in Figure 8. Methanol produces smaller particles than cobalt and in much lower concentrations. The particles generated from methanol are caused by impurities and possibly incomplete combustion, while the particles from cobalt are the purposefully generated cobalt nanoparticles. In the particle measurements measured from in and around the flame it can be clearly seen how the aerosol ages very quickly. Especially in the measurement from beside the flame the distribution is bimodal showing a mode of freshly generated particles and a mode of aged particles grown inside the oven through agglomeration. There was no observed change in the oven background particle concentration when the flame spray related equipment was operated without the flame.



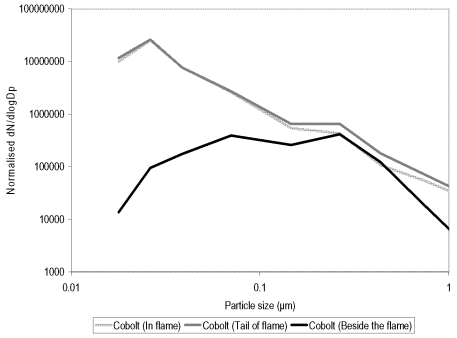


Figure 7. (On the left) Particle size distributions from cobalt, methanol and room air
Figure 8. (On the right) Particle size distributions from different spatial locations around the flame

4. Conclusions

Emissions of ultrafine and fine particles from process sources were found to be highly sensitive to process conditions. In biomass combustion, stabilizing the gasification process with recirculation of flue gas reduces particle emissions, while grate movement and addition of wood cause instability and higher emissions of particles. During welding, the instable process causes high emissions of particles. The particles also change in size depending on the initial concentration and on the transport of particles to the worker breathing zone, thus making the evaluation of potential health effects complex. The evolution of a particle size distribution can also be clearly seen during the flame spray of nanoparticles, with the particles growing and decreasing in concentration when the measurement point is moved from within the flame to different spatial locations inside the oven. Particles generated from different liquids were also found to have very different concentrations and size.

Generation of ultra-fine and fine particles in industrial processes is unstable and the characteristics of the particulates are extremely difficult to predict without direct measurements. Incomplete combustion, instability of temperature and other process conditions, high temperature and presence of impurities in the process generally escalate particle generation. For process development purposes it is most useful to measure the particle emissions directly at the source. However, due to the transformation of the particulates in the atmosphere, these results cannot be directly used for health effect studies.

References

Keskinen, J., Pietarinen, K. and Lehtimäki, M. (1992) Electrical Low Pressure Impactor, J. Aerosol Sci. 23, 353-360.

Marjamäki, M., Keskinen, J., Chen, D-R. and Pui, D. Y. H. (2000) Performance Evaluation of the Electrical Low-Pressure Impactor (ELPI), Journal of Aerosol Science 31:2, pp. 249-261.

Samaras, Z., Ntziachristos, L., Thomson, N., Hall, D., Westerholm, R., Boulter, P., Characterisation of Exhaust Particulate Emissions from Road Vehicles (PARTICULATES)-Final Deliverable No. 16.

Virtanen, A., Marjamäki, M., Ristimäki, J., Keskinen, J. (2001). Fine particle losses in electrical low-pressure impactor, Journal of Aerosol Science, vol 32, pp 389-401.