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Ejector-based sampling from low-pressure aerosol reactors

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ABSTRACT

Online measurements of nanoparticles are necessary when rapid information about the particle size and mass distribution is needed. Currently, the application of online measurement techniques with commonly used instruments such as SMPS, CPMA and ELPI+ is not possible at low-pressure conditions. In this work, a commercial vacuum ejector is used as a simple tool to transfer nanoparticles from a low-pressure region to atmospheric pressure. The vacuum ejector is investigated for different process pressures between 120 and 170 mbar to measure size-selected aerosols in the range from 10 to 100 nm. It was found that the sampling with the vacuum ejector does not change the particle size. The gas and particle dilution factors as well as the particle losses are determined, so that quantitative measurements of the aerosol size distribution can be obtained. Additionally, the applicability of the vacuum ejector is tested during particle synthesis in a low-pressure microwave plasma reactor with a combination of online instrumentation. The direct transfer of the aerosol to atmospheric pressure allows real-time measurements. The primary particle size, mass mobility exponent and effective density are calculated exemplary based on parallel online ELPI+, SMPS and CPMA measurements and are compared to offline TEM analysis.

1. Introduction

Aerosol instrumentation plays an important role in the study of aerosol reactors and process equipment where aerosols have relevance, such as semiconductor processing equipment. The instrumentation can be divided in online and in-situ measurements. In-situ measurements allow to obtain information of the evolution of the particle size inside the actual particle formation zone, usually by non-intrusive optical methods such as laser-induced incandescence (Eom et al., 2004), and laser-based diagnostics (Dreier & Schulz, 2016) which requires however relatively large particle number concentrations, light-absorbing particles and a substantial amount of information about the particle properties. Although, for online measurements the aerosol has to be sampled from a specific reactor zone and conditioned before size analysis can be performed, the advantage is the availability of a range of different measurement methods, which in combination can also be used to extract information about the particle morphology, such as agglomerate density, primary particle size and mass fractal dimension (Eggersdorfer, Grohen et al., 2012; Eggersdorfer, Kadau et al., 2012). Common necessary conditioning steps are rapid cooling and dilution before the aerosol can be analyzed. When the reactor volume is large enough, the finite size of the sampling probe (usually 5–15 mm in diameter) does not disturb the process substantially.

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A fundamental difficulty for the online measurement techniques is that the process pressure is often substantially below atmospheric pressure. In inert-gas evaporation of nonreactive metals, the process pressure is along with the type of carrier gas the main process parameter determining the particle size (Granqvist & Buhrman, 1976). In aerosol reactors, both low-pressure flame synthesis (Zhao, Liu, & Tse, 2009) as well as chemical vapor synthesis apply non-atmospheric process pressures, usually in the range 30–500 mbar. From a scientific viewpoint, low-pressure flames owe their popularity to the fact that the computational modeling is simplified due to a simpler axisymmetric velocity flow field, in which temperature and species profiles are one-dimensional (Janzen, Kleinwechter, Knipping, Wiggers, & Roth, 2002; Janzen, Knipping, Rellinghaus, & Roth, 2003).

In chemical vapor synthesis of nanoparticles, the process pressure is one of the main factors determining particle size, size distribution, and production rate (Schilling & Winterer, 2014). Commercially available aerosol instrumentation is however not designed to operate at pressures below 500 mbar. Some studies have been performed in order to investigate the performance of specific aerosol instruments at lower pressures (Seifert et al., 2004) or have developed differential mobility analyzers adapted for lower pressures (Nanda & Kruis, 2014; Seto et al., 1997). The drawbacks of these procedures is the requirement that all of the components have to be tested over the full pressure range of the process. Mobility analysis as being applied in commercial scanning mobility particle sizers (SMPS) is heavily dependent on the availability of experimentally evaluated charging probability. However, the precision of the charging probability measurements is at lower pressures not sufficient, as a result of the obligatory use of an electrometer as particle counter in lack of a low-pressure condensation particle counter. Other instruments such as the aerosol particle mass analyzer (APM) (Ehara, Hagwood, Coakley, 1996) are – due to their intricate construction as a result of the very high rotational speeds – unsuitable for a low-pressure adaption. Therefore, a more convenient route would be to bring the aerosol from the low-pressure reactor environment to ambient pressure to enable conventional aerosol measurements such as SMPS, ELPI and APM.

Bringing the aerosol back to ambient pressure can be done in a discontinuous way, e.g. by using a flexible evacuated bag which sucks in the low-pressure aerosol and letting the bag inflate to atmospheric pressure (Ober, Mayer, Büttner, & Ebert, 2002). This is a time-consuming and labor-intensive as well as slow sampling procedure. A continuous sampling procedure from low-pressure into ambient pressure would be of great advantage for online methods to achieve real-time measurements. A first report on the technical feasibility of such a sampling was given by Wang et al. (2005), who extracted an aerosol from a low-pressure environment using an ejector. Although they collected particles from the low-pressure reactor, they did not study the transfer behavior of the ejector. Ejectors are commonly used as a tool to generate low-pressure regions on the basis of the Venturi principle. A high-velocity gas provides the driving force to entrain a side gas, usually the gas to be sampled, by the use of a converging nozzle where the pressure is locally decreased. When this local pressure is lower than the pressure of the gas to be sampled, the gas to be sampled will be sucked in. Then the gas mixture is slowed down in a diverging diffuser section and the final pressure of the mixed gas is in between that of the driving gas and the sampled gas, and usually at atmospheric pressure.

In this work, a vacuum ejector is investigated as a suitable transfer system for sampling nanoparticles from a low-pressure process. This study is divided in experiments with and without particle load to find optimal working conditions of the vacuum ejector as well as to determine the pressure dependency of the gas dilution factor (GDF) of defined sample flow rates. Changes in the incoming particle size distribution (PSD) are measured as function of pressure and size. Additionally, the vacuum ejector is assessed with a combination of online measurement instrumentation in a case study of a low-pressure microwave plasma reactor with high production rates on the lab scale.

2. Experimental

2.1. Setup for measuring the gas dilution factor

The ejector used in this work is a commercial ejector designed to sample a gas from a low-pressure region, specified for up to approx. 100 mbar (VIP-4, Landenfeld, Kassel, Germany) and a maximum flow rate of the driving gas Q_{in} of 63 slm (standard liter per minute). It contains three consecutive chambers, each with a Venturi nozzle which are separated from each other by elastic flaps. The Venturi nozzles are designed to deal with smaller to larger pressure regions in flow direction. The final pressure is reached quickly by closing the flaps successively when the final pressure of each Venturi nozzle is reached. In this work only the smallest Venturi nozzle is effectively in use. The ejector is driven by purified nitrogen gas having several bars of overpressure, thereby entraining the gas to be sampled which can be at a pressure lower than atmospheric, denoted by p_{vac}.

The gas dilution factor (GDF) is defined as the ratio of the diluted gas flow rate after the ejector Q_{out} and the gas sampled from a low-pressure system Q_{vac}. Knowledge about the gas dilution factor is required when the particle losses are to be determined on the basis of the measured particle dilution factor (PDF). Therefore, a setup was built that allows to investigate the driving gas Q_{in}, the sampling flow rate Q_{vac} and the outflow rate Q_{out} in dependency of the driving gas pressure p_{in} and the process pressure p_{vac} (Fig. 1). The system provides a gas at defined sub-atmospheric pressures by varying the needle valve opening. The flowrates Q_{out} and Q_{vac} are measured with flowmeters (Model 4040 and 4143, TSI, Minneapolis, US) as function of the system pressure p_{vac} with a vacuum gauge (TTR 101, Leybold, Cologne, Germany).