Journal of Aerosol Science 41 (2010) 719–728

Contents lists available at ScienceDirect

Journal of Aerosol Science

journal homepage: www.elsevier.com/locate/jaerosci

Towards traceable particle number concentration standard: Single charged aerosol reference (SCAR)

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ARTICLE INFO

Article history:
Received 7 October 2009
Received in revised form
26 April 2010
Accepted 29 April 2010

Keywords:
Number concentration
Standard
Calibration
Singly charged
Aerosol reference

ABSTRACT

A concept of realizing a standard for aerosol particle number concentration was tested, based on generating singly charged aerosol particles in the size range from 10 up to 500 nm. To this end, a device named single-charged aerosol reference (SCAR) was designed, built, and tested. The device is based on electrical charging of nanoparticles and subsequent growth of the particles. With an accurate measurement of volume flow and electrical current from the singly charged particles, the number concentration can be accurately, and in the end, traceably determined. Laboratory tests have shown that the device can be used to generate a narrow (GSD < 1.3) particle size distribution of singly charged particles. The device can be used for traceable calibration of instruments measuring the number concentration of the particles.

1. Introduction

Aerosol particles play a key role in important scientific and technological issues, such as global atmospheric change (IPCC, 2007), health effects of anthropogenic particulate emissions (CAFE, 2001), and breakthroughs in nanotechnology (e.g. NSF, 2003). In addition to these larger concepts, aerosol particle measurement is widely applied, e.g. in clean room technology and process technology, as well as in studying local and national emissions from traffic, industry, and energy production. Aerosol measurements are also utilized in air quality control, in medical studies and in aerosol technology in general.

Measurement of mass concentration of the particles has long traditions, and international standards exist for this purpose. Along with the development within all the application areas mentioned above, measurement of fine and ultrafine particles has gained importance, exceeding the importance of simple mass concentration. This is evident, since most of the phenomena under interest and of importance deal with particle size, size distribution, number concentration, or surface area concentration. Several continuous flow type aerosol instruments are now capable of measuring these quantities on-line, including optical/laser particle counter (OPC/LPC; e.g. Knollenberg & Luehr, 1976), condensation particle counter (CPC; e.g. Agarwal & Sem, 1980), aerodynamic particle sizer (APS; e.g. Baron, 1984), scanning mobility particle sizer (SMPS; Wang & Flagan, 1990), electrical low pressure impactor (ELPI; Keskinen, Pietarinen, & Lehtimäki 1992) and a number of fast mobility sizers (FMPS, EEPS, DMS; e.g. Biskos, Reavell, & Collings, 2005).

When the number size distribution of fine and ultrafine particles is measured, the particle size can be determined with rather good accuracy. There are several physical properties such as electrical mobility, inertia, and diffusion coefficient to allow this. In addition, spherical custom made latex-particles and even nanosized polymer molecules, with size verification

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doi:10.1016/j.jaerosci.2010.04.012

by SEM and TEM microscopy, are available to perform the calibration in most of the size ranges above 3 nm. However, the measurement of other quantities, particularly particle number concentration has mostly not been addressed by standards. For decades, the clean rooms of production facilities have been practically the only application where particle number concentration and its measurement is covered by standards. Cleanliness is usually monitored by using optical single particle counters (OPC) or laser single particle counters (LPC) (Mirme & Tamm, 2002; ISO 14644-1, 1999). The counting efficiency of these instruments is calibrated by comparison with a reference instrument. However, there is no indication in the standard (ISO 21501-4, 2007) how the absolute counting efficiency of such a reference instrument should be calibrated. Consequently, such reference is not easily available. However, at least the Swiss Federal Office of Metrology METAS offers a particle concentration reference (e.g. Schleicher, 2004).

The number concentration calibration of continuous flow type instruments usually reduces to accurate calibration of two properties, the sample flow rate and the count efficiency (instrument response to known number concentration). The flow rate can in the end be compared with traceable standards of the international and national institutions, such as NIST in the US (Wright, Mattingly, Nakao, Yokoi, & Takamoto, 1998) and MIKES in Finland (Sillanpää, Niederhauser, & Heinonen, 2006). The counting efficiency, on the other hand, forms a much more difficult problem. In practice, the counting efficiency can be calibrated by challenging the instrument to a known particle number concentration. For large particles, the number concentration can be calibrated against reference measurement of mass concentration. As an example, Armendariz and Leith (2002) calibrated aerodynamic particle sizer (APS) against gravimetric mass concentration measured by a cascade impactor. Volckens and Peters (2005) used fluorometry to detect particle mass. However, these methods become impractical for finer particles due to prolonged measurement times. Quite recently Koch, Pohlmann, and Schwarz (2008) introduced a number concentration reference where an aerosol with a well defined starting time, coagulates in a chamber and has a number concentration which can be unambiguously derived from the life time of the aerosol. This 'ab initio'-method applies best for submicron particles, but as a disadvantage the particle size is changing with time.

The differential mobility analyzer (DMA) is a convenient source of (nearly) monodispersed submicron particles. Combined with a calibrated aerosol electrometer, the DMA can be used as a number concentration standard to calibrate particle counting instruments (Liu, Pui, Hogan, & Rich, 1975). This has become the most popular method to calibrate continuous flow CPC's (e.g. Banse, Esfeld, Hermann, Sierau, & Wiedensohler, 2001; Hermann et al., 2007; Liu & Kim, 1977; Sem, 2002). In addition to CPC calibration, differential mobility analyzer method can be used in particle size calibration (Mulholland et al., 2006). With proper calibration of the flows and voltages in the DMA, it can be used to select particles with the known electrical mobility. The main drawback of the DMA method for both the number and size calibration is the fact that particles with different charge and size combinations can have the same electrical mobility. Only the smallest particles can be trusted to be singly charged. In number concentration calibration with the electrometer, multiply charged particles give higher signal because of their higher charge. In size calibration, multiply charged particles have larger size than the singly charged particles. It is possible to reduce these errors by careful selection of particle sizes from a narrow distribution (e.g. Hillamo & Kauppinen, 1991; Keskinen, Marjamäki, Virtanen, Mäkelä, & Hillamo, 1998; Wang & John, 1988) or by applying a calculated correction for doubly charged and triply charged particles (Hoppel, 1978; Kauppinen & Hillamo, 1989). Both methods reduce the uncertainties in the measurements but are not ideal solutions for calibration purposes. As a recent attempt to solve the practical problems in CPC calibration, Fletcher, Mulholland, Winchester, King, and Klinedinst (2009) describe a method of comparing aerosol electrometer and CPC. They achieve a total accuracy in the order of 5–27% in total number concentration, depending on the concentration level.

Okuyama, Shimada, Choi, and Han (2005) describe a method for generating nearly singly charged particles over a wide size range by using an X-ray source to charge the particles and a DMA to classify the particles according to their electrical mobility. They describe how to generate singly charged aerosol particles such that the relative fraction of doubly charged particles is equal to or less than 5%, by adjusting operating current and voltage of the X-ray source.

In a conference abstract in 2002, Mirme and Tamm discussed a novel idea of first charging nanoparticles and then growing them into larger size. This way the charge distribution of the grown particles would follow the nanoparticle charge distribution, with very few multiply charged particles. Recently, they reported tests on a particle generator applying the idea (Uin, Tamm, & Mirme, 2009). They concentrated on validating their basic idea and on characterizing the quality of the output aerosol, obtaining a multiply charged fraction well below 5%. In this paper, we concentrate on applying the same basic idea for the purpose of realizing a particle number concentration standard for a wide size range. In addition to the particle generator, our system includes an accurate measurement of volume flow and electrical current from the charged particles. In this paper, we take the method into a real calibration unit. We describe the concept, the practical apparatus and its operation, and discuss its performance. We also present examples of performing number concentration measurement calibration of various particle counting devices. Our development work has been included in the Joint Research Project called: Traceable Characterization of Nanoparticles in the European Metrology Research Programme since June 2008 (EURAMET, 2009).

2. Concept and device

In the single charged aerosol reference-device (SCAR), realized in this work, the following units are used in the numbered order to generate a narrow, singly charged output aerosol size distribution: 1. Generation of primary