

AEROSOL GENERATION BY SPARK DISCHARGE*

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ABSTRACT

An aerosol generator producing particles of any desired mobility, equivalent radius, and conducting material has been developed. Particles are formed by electrode atomization in a spark discharge. A high yield of charged particles enables generation of high concentration ($> 10^5 \text{ cm}^{-3}$) monodisperse aerosols using electrostatic classification. In order to obtain nonagglomerated particles, the aerosol can be turbulently diluted at the region of particle formation by gas injection through a nozzle. Charged Au particles in the atomic cluster size range (1.3 nm) have been produced in a concentration of $3 \times 10^5 \text{ cm}^{-3}$.

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INTRODUCTION

The generator to be described is based on electrode atomization by spark discharge. It produces ultrafine, highly charged particles of any conducting material. The high yield of charged particles is desirable for production of monodisperse aerosols of high concentration by mobility classification. Universality of the generator with respect to material leads to a wide range of applications. For instance particles of high volatilization temperature are readily generated. The spark generation principle has successfully been applied to Carbon particle production in connection with coagulation studies [1] and simulation of soot particles [2, 3], but a mature generator model and its output characteristics have not been described earlier.

EXPERIMENT

A schematic of the generator is shown in fig. 1. In the model applied for the present study cylindrical electrodes of 3 mm diameter are mounted in jaw chucks of a type used in draftsman's lead pencils. The mounting enables fast exchange of electrodes and precise adjustment of the spacing between them. For the present experiments the spacing was 1.0 mm. A 2.2 nF capacitor is continuously charged by a high voltage power supply (max.12 kV) in constant current mode. Fast discharge occurs when the breakdown voltage is reached. The spark repetition frequency, f , can be adjusted through the charging current I which was variable between 0 and 1 mA. This corresponded to $0 < f < 50$ Hz. N_2 entered through a teflon pipe with a circular aperture of 3 mm diameter ending 4 mm from the spark gap. Windows on either side of and parallel to the spark and flow axes enable viewing of the spark. Conducting material was chosen for the housing (Al) as well as for the output aerosol tube in order to avoid loss of charged particles by electrostatic precipitation to charged insulators.

GENERAL CHARACTERISTICS

A Carbon aerosol was produced in N_2 (impurities $< 10ppm$) using glass-Carbon electrodes (Sigradur G, [Ringsdorff-Werke GmbH, Bonn-Bad Godesberg, W. Germany]). Particles of a certain size interval were selected with a differential mobility analyzer (DMA) of the Hanke type (Hanke GmbH, Gmunden, Austria). A constant DMA input flow of 4 l/min was branched off from the generator output. The DMA resolution $\frac{\Delta D_p}{D_p}$ was 5% (D_p = particle diameter). The particle concentration, n , at the DMA output was measured by means of an aerosol electrometer assuming unit electronic charge per particle. This assumption was verified in a separate experiment by means of repeated mobility analysis after establishing charge equilibrium in an aerosol neutralizer.

Figure 2 shows $n(D_p)$ for $f=25$ Hz for different flows. When decreasing the flow, more material is atomized per unit of gas volume, and the mean particle size becomes larger. Similarly, the mean particle size shifts to larger particles when the repetition frequency or the energy dissipated in each spark is increased. Energy dissipation is increased by raising the capacitance value or the breakdown voltage, which is given by the electrode spacing.

Good stability of aerosol production was reached after letting the generator run at 50 Hz for a day. This results in rounding of electrode edges and the spark then appears with equal probability at all places of the electrode profile. Fig. 3 shows the generator output stability at 25 Hz and 20 l/min.

It must be expected that spark characteristics, and therefore aerosol production, is highly sensitive to the gas composition and that traces of electronegative molecules capturing free electrons may have a large influence. In order to estimate the significance of electronegative components, O_2 and CO_2 have been mixed to the N_2 before the generator. While the shape of the size distribution showed no variation, the total concentration decreased when more than 50 ppm of CO_2 was added. Figure 4 shows the concentration as a function of the CO_2 concentration.

The results for O_2 were similar. Where reproducibility is desired, gases with high enough purity, or identical gases must be applied.

In order to obtain unagglomerated (primary) particles, coagulation is avoided by fast turbulent dilution at the spark gap. For this purpose the Teflon input pipe is replaced by a glass jet with a rectangular aperture of 0.3×3.0 mm, ending 3 mm from the spark gap. The gas pressure required for the maximum flow of 40 l/min was 4 bar.

If every spark produces a certain number N_0 of primary particles, independent of the flow rate Q_x and the spark repetition frequency f , the concentration n is given by

$$n = N_0 \frac{f}{Q}$$

as long as no coagulation takes place. Figure 5 shows $N(\frac{f}{Q})$ measured for $\frac{f}{Q} < 0.036 \text{ cm}^{-3}$. The linearity supports the assumption made. A yield of $N_0 = 3 \times 10^5$ particles per discharge is derived from Fig. 5. The mean diameter of uncoagulated particles showed no sensitivity to f or Q in the range above.

Figure 6 shows $n(D_p)$ of primary carbon particles for $f=25$ Hz and $Q=40$ l/min. The solid line represents a log-normal fit. Such distributions showing a "tail" on the large particle side are typical for coalescence growth [4].

For gold, this distribution for the same conditions is shown in Fig. 7. Here, the primary particles are as small as 1.3 nm. This is the size of atomic clusters containing about 50 atoms. Since the mobility of these particles ($0.8 \text{ cm}^2/\text{Vs}$) is in the range of typical ionic mobilities (ca. $2 \text{ cm}^2/\text{Vs}$), tests were carried out to make sure that the measured species is actually gold, and not clusters of gasion molecules formed in the spark with impurities. Nitrogen ions can be ruled out, since the negative polarity is chosen, and negative nitrogen ions do not form. The first test was a coagulation experiment: The aerosol was passed through a vessel (residence time 8 min) before the DMA, and normal coagulation or coalescence

growth was observed by the size distribution change. This points to a species with low vapor pressure. Secondly, the particles were sampled and high resolution transmission electron micrographs were taken. They show patches consistent with 1.3 nm particles at the resolution limit. Only high atomic weight elements such as Au show detectable electron scattering for these sizes. The solid line in Fig. 7 represents a fit to a normal distribution. Such size distributions have been found for growth processes involving absorption of single atoms [5].

CONCLUSION

An aerosol generator with a high yield of charged ultrafine particles has been developed. Even 1.3 nm Au particles are obtained charged. This particle size has not been observed before in aerosol measurements. The reason is that such particles are not accessible by optical methods, and charging methods are ineffective.

The high yield of charged particles enables monodisperse generation of ultrafine aerosol concentrations above 10^5 cm^{-3} . For applications requiring particles with defined diffusional or aerodynamic behaviour of arbitrary shape, monodisperse aerosols of any selected particle size and chemically stable conducting material are producible. Air can be used as a carrier gas with inert materials like Au.

Applications include inhalation diagnosis with radioactive monodisperse aerosols. Recent studies have shown that the use of monodisperse ultrafine aerosols for such purposes has great advantages concerning interpretability [6] and radioactive dose [7] compared to conventionally applied polydisperse fine aerosols.

Noncoagulated particles were obtained in limited size ranges. The influence of material, spark characteristics (energy, duration), carrier gas, etc. on the primary particle size distribution remains to be investigated.

Compared to conventional evaporation aerosol generators, the generator shown here has the advantage of being quite universal with respect to material, and extremely simple. The material to be atomized is heated very locally, and no other

material is evaporated or desorbed. This enables production of very clean particles, and makes the generator principle applicable to studies of adsorption or catalysis on small particles or clusters. Furthermore, no heating-up time is required. Only very small amounts of material are needed, if electrodes are coated with the material to be dispersed. By coating, extension to chemically stable nonconductors should be possible, because any material conducts in electrical breakdown.

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REFERENCES

1. H. Burtscher, A. Schmidt-Ott, *Phys. Rev. Lett.*, 48, 1734, (1984)
2. H. Burtscher, A. Schmidt-Ott, *J. Aerosol Sci.*, 17, 699, (1986)
3. H. Burtscher, A. Schmidt-Ott, in *Aerosols, Formation and Reactivity*, Pergamon New York, p. 833 (1986)
4. C. G. Granqvist, R. A. Buhrman, *J. Appl. Phys.*, 47, 2200, (1976)
5. G-H. Comsa, *J. Physique*, 38, C2-185 (1977)
6. C. Schuemichen, *Proc. of Erstes Grafschafter Kolloquium, Schmollenberger Grafschaft* (1987)
7. J. Heyder, J. Gebhart, G. Rudolf, C. F. Schiller, W. Stahlhofen, *J. Aerosol Sci.*, 17, 81, (1986)

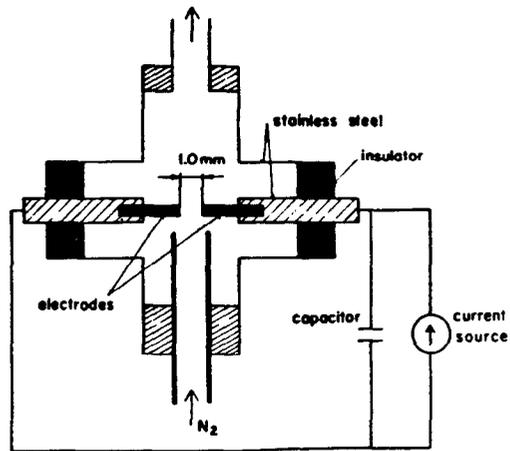


Fig. 1. Schematic of the generator.

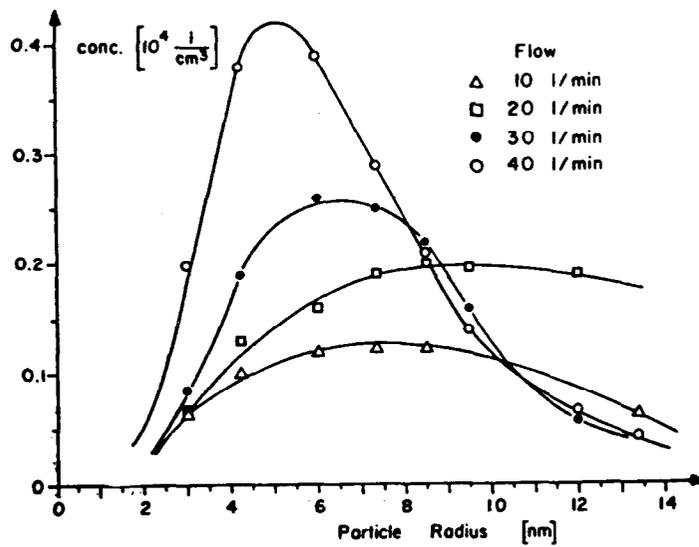


Fig.2. Concentration vs. radius of carbon particles for different flows

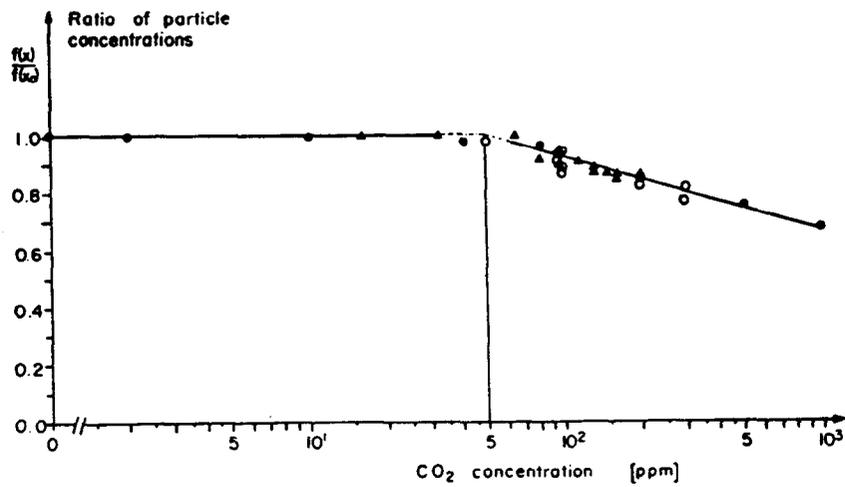


Fig. 3. Ratio of particle concentration with and without CO₂ added.

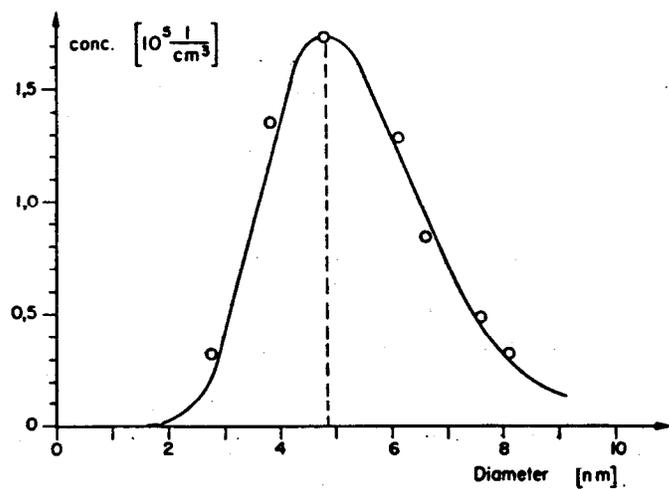


Fig. 4. Size distribution of primary carbon particles.

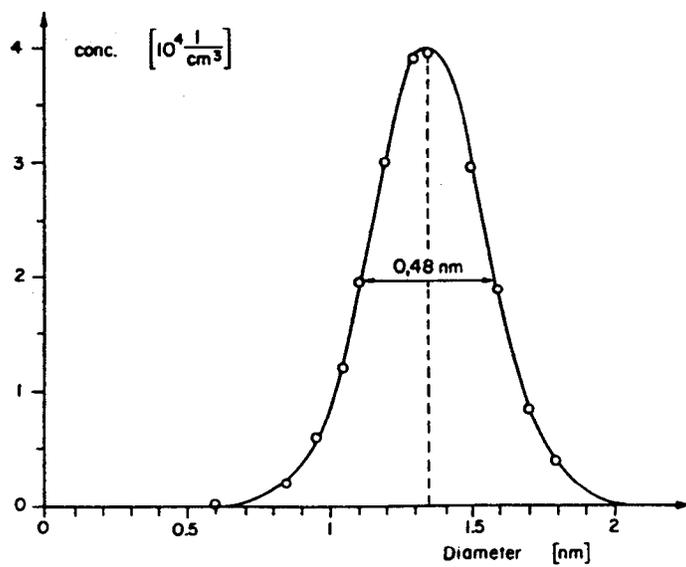


Fig. 5. Size distribution of primary gold particles.