

Characterization of nanoparticle flow produced by gas aggregation source



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ABSTRACT

A simple and compact gas aggregation cluster and nanoparticle source based on a planar magnetron (Haberland type) without mass separation was characterized. Such source produces a beam of neutral, positively and negatively charged particles that are dragged by a buffer gas expanding from the aggregation chamber. Sizes, speeds and charges of nanoparticles have been determined using a combination of TEM micrographs, electrostatic deflection setup and numeric modeling. The strong dependence of the velocity of the nanoparticles on their size was confirmed both experimentally and by modeling.

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1. Introduction

In the last decades, gas phase production of clusters and nanoparticles and their deposition has attracted special interest because of a wide range of possible applications of such materials [1,2,5]. As a consequence of this, various kinds of gas aggregation nanocluster sources were designed (e.g. reviews [4,6]). The concept of gas aggregation originally presented by Sattler et al. [3] was further developed by Haberland et al. [7], who used a planar magnetron as a source of the material for nanocluster growth. Such sources are able to produce a large number of charged clusters and nanoparticles that are dragged by the flow of the carrier gas and extracted from the source through an orifice into the deposition chamber with a substantially lower pressure. The original two-stage UHV design offers mass filtering and acceleration of negatively charged clusters, albeit on the expense of the intensity of the cluster beam. It was recently demonstrated that this system may be simplified using only a single-stage expansion of the gas without mass filtration and may use all clusters (neutral as well as positively and negatively charged). This leads to a dramatic increase of the

deposition rate of the nanoparticles (e.g. Ag, Ti, Pt [8–10,18]), albeit with certain mass distribution and slow landing on the substrate.

Important parameters of such nanocluster sources are size and mass distribution [11], charge and velocity of produced nanoclusters or nanoparticles. These parameters determine the transport of the nanoparticles from the source to the substrate. When they are known, additional manipulation of the process of deposition of nanoparticles is possible. In this study we focused on the characterization of their velocity and charge [12,13]. The experimental approach was supported by simulations. The simulation of aerodynamics of the nanoparticles is following an approach similar to these reported by Smirnov et al. in [18] and Skorovodko et al. [14]. Silver was chosen as a model material for the nanoparticles.

2. Theory and modeling

2.1. Gas dynamics and acceleration of nanoparticles

In the Haberland gas aggregation nanocluster source [15] the material is sputtered from the target and forms nanoparticles that are dragged by the flow of the carrier gas. These nanoparticles are then extracted from the source through an orifice into the deposition chamber with a substantially lower pressure. Since the pressure inside the source is usually from several Pa to several hundred Pa and the velocity of the carrier gas flow inside the source is some tens of cm/s, the carrier gas can be treated as viscous

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laminar flow in the cylindrical part of the source. Under such conditions the nanoparticles have much bigger cross-section than the atoms of the carrier gas and their velocity is equal to the drift velocity of the gas.

However, such continuum approximation is not appropriate for the area near the orifice, where a pressure drop of several orders of magnitude occurs over the length of several millimeters. This is accompanied by a strong acceleration of the carrier gas flow due to expansion. Since the mass vs. cross-section ratio of the nanoparticle increases with its diameter and the carrier gas density decreases during expansion, the heavier particles do not undergo enough collisions with light gas atoms to equalize their velocity and direction with the gas. This can also cause differences in the flight path between lighter and heavier nanoparticles that can lead for example to so called aerodynamic lensing [16,17]. Even when the effect on the shape of the flight path of particles of various size is negligible, the differences in their velocity still remain.

The acceleration of the nanoparticles near the orifice with purely conical walls has been nicely treated by Smirnov et al. in [18]. The analytical theory leads to the dependency of the final velocity v of the nanoparticle on its mass m in the form $w \approx m^{-2/9}$ for a particular pressure and temperature in the source. For the source with the pressure 75 Pa at room temperature and orifice of the diameter 1.5 mm, i.e. conditions used for experiments reported in this work, this calculation predicts the speed of 30 m/s for silver nanoparticles of 5 nm diameter and 7 m/s for nanoparticles of 40 nm diameter.

In order to inhibit the effects of aerodynamic focusing, the orifice of our source is not purely conical, but it has a cylindrical shape in the last 2 mm. The expansion from such orifice is different from the expansion from a purely conical orifice. To have a better understanding of the conditions in the orifice we have made a direct 2D Monte Carlo simulation of the carrier gas flow near the orifice using the DS2V code [20]. We have simulated a domain from 13 mm before the orifice plane to 50 mm after it (2D maps of gas flow properties are shown in Fig. 2). From this domain we have taken the data of velocity, density and temperature near the axis of the orifice from 10 mm before to 10 mm after the orifice. As shown below, this region is safely several times longer than the region where almost all of the acceleration and expansion of the gas occurs.

The Monte Carlo code calculates 2D data of the flow, but since our source produces a relatively narrow beam of nanoparticles (half-angle of about 6°), we treated in this work the acceleration region as basically a 1D problem. The problem of acceleration of the particle can be treated as a movement of a single particle in an undisturbed gas, since the volume and mass fraction of the nanoparticles in the buffer gas is less than 10^{-9} and 10^{-3} , respectively,

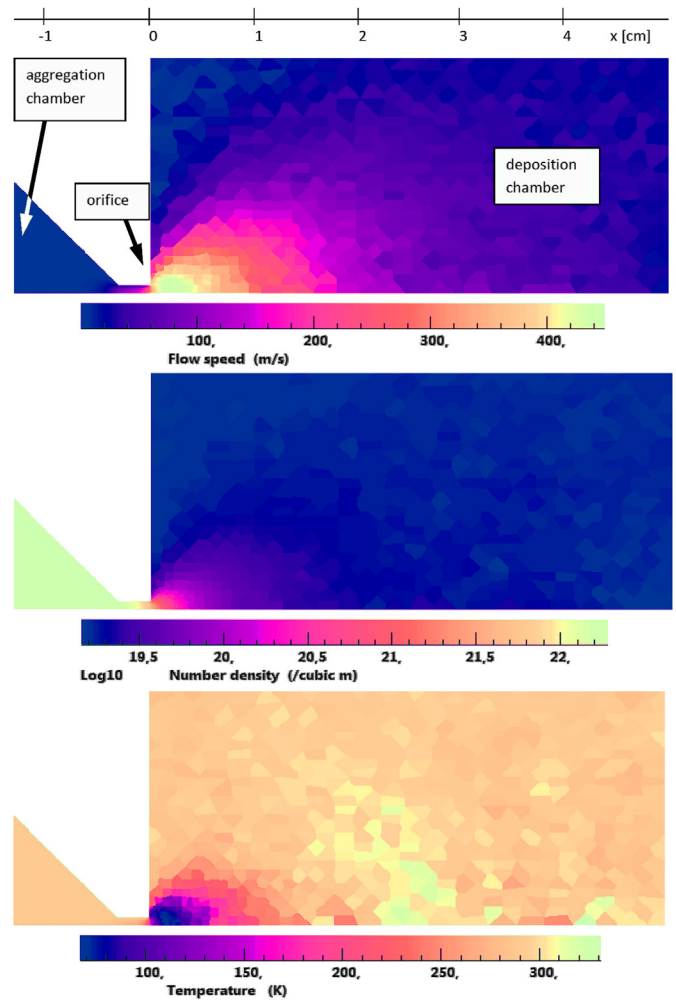


Fig. 2. 2D map of the gas velocity, density and temperature near the orifice of the nanoparticle source obtained using DSMC modeling. Bottom line of each picture is the axis of symmetry, the orientation of the x-axis is shown on top. Position $x = 0$ corresponds to the orifice.

according to the estimates based on the deposition rates of the nanoparticles.

According to [18], the acceleration of a spherical particle in the gas flow can then be described by the equation:

$$dw/dt = v^*(u - w) \quad (1)$$

where w is the velocity of the particle, u is the drift velocity of the gas and v is proportional to the collisional frequency. It is equal to:

$$v = (m/M) * N * v^* \sigma \quad (2)$$

where m is the mass of the atom of the gas and M is the mass of the particle, N is the numeric density of the gas, v^* is the average thermal velocity of the gas atoms and σ is the particle cross-section.

The original equation does not account for the dependence of the collision frequency of the particle with the gas on the drift velocity of the particle. When we add the velocity vector of the gas molecule to the vector of difference between drift velocities of gas and the particle, we can obtain an integral that describes the relative velocity v_{eff} of the gas molecules and the particle:

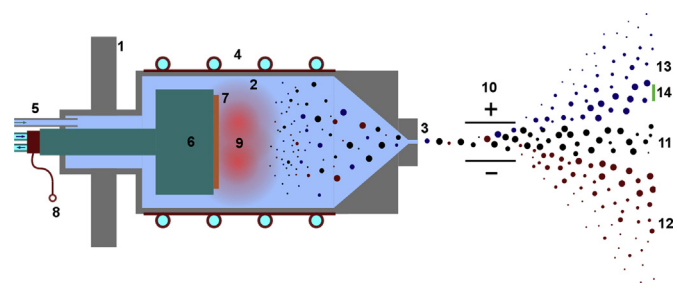


Fig. 1. Scheme of the source of nanoparticles and of the general layout of the experiment. 1 – main chamber; 2 – aggregation chamber; 3 – orifice; 4 – water cooling; 5 – carrier gas inlet; 6 – water-cooled magnetron; 7 – target; 8 – power source; 9 – plasma; 10 – deflection plates; 11 – neutral nanoparticles; 12 – negatively charged nanoparticles; 13 – positively charged nanoparticles; 14 – substrate.

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