Heterogeneous nucleation of sulfur vapor on tungsten oxide and NaCl nanoparticles: Determination of the radius and the contact angle of critical nucleus.

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The heterogeneous nucleation plays an important role in the atmospheric processes, in the condensation particle counters and can be used to generate composite nanomaterials. Heterogeneous nucleation depends on the surface property of the seed particles. If surface of the seed particle is not perfectly wettable, critical nucleus with radius r and contact angle Θ is formed on the surface of particle.

In this work we describe the method of determination of the radius and the contact angle of the critical nucleus that formed on the surface of the seed nanoparticles during heterogeneous nucleation in the flow chamber. The experimental setup is shown in Fig. 1. It consists of three main parts [1]: the particle generator, the flow nucleation chamber and an Automatic diffusion battery (ADB) coupled with a condensation nucleus counter to measure aerosol size distribution and particle number concentration. Two different types of seed particles (tungsten oxide and NaCl) were used in the experiments. The particles were obtained by evaporation of sample material in the crucible (located in the high-temperature zone) followed by vapor condensation in the flow. Particle size distributions are well described by log-normal law. For tungsten oxide particles: geometric mean diameter d_g = 7.8, geometric standard deviation σ_g = 1.45, for NaCl: d_g = 7.3 nm, σ_g = 1.45.

The flow of argon with seed particles enter in the nucleation chamber where the heterogeneous nucleation takes place. Fig. 2. shows the size distributions of the incoming seed particles (solid line) and the particles at the outlet of chamber (after the heterogeneous nucleation). One can see that size distribution at the output is bimodal. One of these modes (left) is a part of the spectrum of original particles which is smaller than the size required for the start of heterogeneous nucleation. The second mode (right) is formed by particles on which critical nucleus were generated followed by condensation growth. During the experiments we varied the vapor pressure of sulfur at the unchanged size distribution of seed nanoparticles. The axial and radial temperature profiles were measured by a thermocouple. The radial profiles of sulfur vapor concentration and supersaturation were determined by solving the mass transfer equation.

Next, we used the Fletcher`s formula for the rate of heterogeneous nucleation with pre-exponential factor, offered by us. Free parameter in this formula is the contact angle of the critical nucleus on the surface of seed particle. For each experiment, we have found a contact angle that give good agreement with experimental data (part of enlarged particles) As a result, we obtained the dependence between the contact angle of lenticular critical nucleus and the radius of maternal seed particle (Fig. 3, triangles - tungsten oxide particle, rings - NaCl particle). Thus, we have a monotonically increasing dependence between the contact angle of critical nucleus and the size of the maternal seed particle. Qualitatively, this is consistent with the dependence results [2, 3], where the heterogeneous nucleation of n-nonane and n-propanol on the surface of tungsten oxide and silver seed nanoparticles was studied. By using the process of condensation growth in the flow of sulfur vapor, we have prepared samples of submicron droplets of sulfur at the surface of formvar substrate (Fig. 4). This specimen was shaded through deposition of silver in a vacuum evaporator at the angle of 15^0 to the specimen surface. This makes it possible to determine the height (thickness) of sulfur lenticular particles. As a result, it was calculated that the contact angle for a range of droplet sizes 100 – 300 nm amounts to 63 ± 6^0. In addition, in [4] provided data on the contact angles of the sub-millimeter droplets of sulfur on a silicon substrate (58 ± 2^0) and teflon (82 ± 2^0). Based on above data, it can be assumed, that the contact angles for macro-droplets of sulfur on tungsten oxide and sodium chloride are higher than for nano-sized range. In this case, applying the approach, developed in [2, 5], we can conclude that the line tension of studied sulfur critical nuclei is negative. Our nearest plan is to obtain the missing experimental data, which will give a more reliable description of the studied process.

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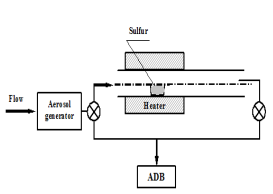

Fig 1.

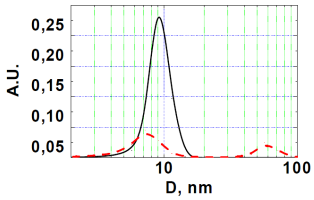

Fig 2.

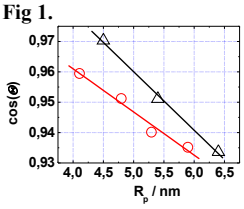

Fig 3.

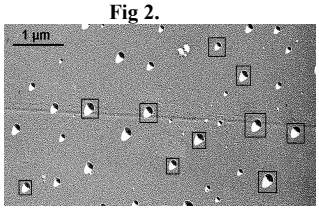

Fig 4.