Nucleation of H2SO4-H2O in Laminar co-Flow Tube: CFD Simulations and Analytical Modelling.

Trávníčková, Tereza
2018

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Nucleation from vapour mixtures of sulphuric acid, water and other substances like amines, terpenes etc. is one of the key processes that increase the number of aerosol particles in the atmosphere, especially in connection with industrial activity (sulphuric acid) or the occurrence of wooded areas (terpenes). Condensation of the resulting nuclei to form cloud droplets influences both local weather and global climate change due to the increased scattering of the sun rays on the surface of the particles. In addition to atmospheric nucleation, it is possible to encounter nucleation also in industrial applications. In particular, the binary nucleation of sulphuric acid with water is a major problem for the operation of coal-fired boilers, where it reduces the conversion of the reaction and the lifetime of the device.

To describe the binary nucleation of sulphuric acid with water, several mathematical models have been proposed. However, the predictions of these models are often very different. It opens a question of which mechanisms are dominant, because the choice of suitable nucleation mechanisms depends on many factors, such as atmospheric conditions, geographic location, presence of other substances in the atmosphere, etc. Therefore, various experimental devices such as piston expansion chambers, diffusion chambers and laminar flow chambers have been proposed for verifying these models. Recently, the most frequently used are laminar flow chambers with turbulent mixing units, ensuring almost perfect contact between the two components and uniform distribution of the arising clusters. The problem with these devices is the absorption and desorption of sulphuric acid on the device wall and the dispersion of the nucleation zone throughout the whole mixing unit. Thus, the mathematical modelling of these devices is limited only to the calculation of the average values of the nucleation rates estimated from the residence time of the droplets in the chamber and to the correction of the particles and the sulphuric acid losses by experimentally obtained coefficients. In 2018, Trávníčková et al. [1] published pilot measurements on the Laminar Co-Flow Tube (LCFT) which owing to its arrangement minimizes the loss of acid and newly formed particles on the chamber wall. Moreover, due to the laminar nature of the flow, it enables flow dynamics modelling in the system and thus precise definition of the nucleation zone. Unlike turbulent mixing flow chambers, it is also possible to simulate the experimental nucleation rates distribution values in the nucleation zone.

In this work the validity of two mathematical models differing in the complexity of flow dynamics is tested on two new nucleation isotherms measured by LCFT device. The first model is CFD and commercial software Fluent 16.2 is used for hydrodynamics evaluation. The second one is analytical programmed in the Matlab (Coflow). While Fluent used axi-symmetric domain on which commutated transport processes by Navier-Stokes equations solve together with the continuity equation and the scalar equation for description of vapours concentration profiles, the analytical model Coflow considers a homogeneous velocity profile

in mass only transport equations computation. This allows the problem to be modelled as a one-dimensional non-stationary radial diffusion in a coordinate system moving with the gas:

(1)

This equation, has an analytical solution [2]. Both models were used to quantify the semi-empirical nucleation rate [3]:

, (2)

depending on relative acidity and relative humidity , which was then further normalized by the experimentally obtained number of nuclei to obtain the true value of the experimental nucleation velocity.

The value of the experimental nucleation rate depends on the ratio D of the maximum theoretical nucleation velocity and its integral values over the whole nucleation zone. How this ratio varies when using these two models for different experimental conditions can be seen in Figure 1.

Figure 1: Ratio D computed by numerical (circles) and analytical (squares) model for two temperatures (orange 15°C, blue 27°C)

Figure 2: Overestimation of Coflow model in dependence on outer and inner flow ratio

In the Figure 2 it is possible to see how this ratio computed by Coflow program fits the ratio computed by Fluent simulations. It follows from the figure that using Coflow analytical model overestimates the theoretical and hence experimental nucleation rate one to three times. This is an acceptable error compared with uncertainty of experimental measurement, which can reach one order of magnitude.

Acknowledgement: This work was supported by the Czech Science Foundation [grant number 17-19798S]; ACTRIS-CZ-RI [grant number CZ.02.1.01/0.0/0.0/16_013/0001315]; EU project Horizon 2020 ACTRIS2 [grant number 654109].

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