Binary nucleation of H_2SO_4-H_2O in laminar co-flow tube

[1]T. Trávníčková, [1]L. Škrabalová, [1,2]J. Havlica, [3]J. Hrubý, [1]V. Ždímal
[1]Institute of Chemical Process Fundamentals of the CAS, v. v. i., Rozvojová 2/135, 165 02 Prague 6, Czech Republic; phone: +420 220 390 262, fax: +420 220 920 661
e-mail: travnickovat@icpf.cas.cz; [2]University of Jan Evangelista Purkinje, České mládeže 8, Ústí nad Labem, 400 96, Czech Republic; [3]Institute of Thermomechanics of the CAS, v. v. i., Dolejškova 1402/5, 182 00 Prague 8, Czech Republic.

Atmospheric aerosols significantly affect many areas of human being, whether it be on the global climate, local formation of clouds, visibility and signal transmission, or human health. One of the main mechanisms of fine aerosols formation is a binary nucleation of sulphuric acid with water. Because accurate description of all processes occurring in the atmosphere and conditions under which these phenomena occur is very difficult, the mechanism of nucleation of sulphuric acid with water remains still insufficiently described. In order to overcome this problem several types of experimental devices have been proposed. For this purpose flow chambers with turbulent mixing are most commonly used. However, these chambers encountered problems, how to determine the position of the nucleation zone and the concentration of sulphuric acid in this zone.

We measure binary nucleation of sulphuric acid with water using Laminar Co-Flow Tube, which overcomes both of these problems. In this paper we present the first results of measurements on this device. It appears that with this type of chamber equivalent results as measured on the flow chambers with turbulent mixing can be obtained. Moreover, the use of super-clean gases in our experiment helps to eliminate the impact of amines and other compounds to stabilize the clusters.

Laminar Co Flow Tube is actually a tube within tube, where the inner tube is fed with a mixture of saturated vapour of sulphuric acid in a nitrogen stream, and the packing stream is at approximately equal gradual rate fed with a mixture of saturated water vapour in nitrogen. The central tube ends after several tens of cm and on its edge, it comes to diffusion contact of these two mixtures, which then further flow throw the chamber together. Laminar parallel flow can be modelled and gain by this way a position of nucleation zone in which stable nuclei of a new phase are formed. These nuclei in the supersaturated mixture continue to grow and are carried out of the chamber. Thanks to the placement of acid into axial flow, and because the diffusion coefficient of acid in nitrogen is much lower than the diffusion coefficient of water in the nitrogen, losses of H2SO4 on device walls are effectively suppressed. At the outlet of the chamber, the particles are detected by a condensation particle counter with enlarger of clusters (PSM AirModus).

Mass and momentum balances were solved using commercial software tool FLUENT 14.5. Composition of the inlet stream was obtained by calculation from the saturated vapour pressure of water (Katz 1967) and sulphuric acid (Kulmala 1990) at the saturation temperature. Based on knowledge of temperature, these pressures and flow rates composition of the mixture were calculated at every point of the apparatus and the profiles of relative humidity *RH* and the relative acidity *Ra* were obtained. The theoretical nucleation rate was calculated according to Wyslouzil al. (1991) and experimental nucleation rate J_{exp} as a function of *T* and *Ra* was determined by Anisimov and Wagner (1993) parametrization from the particle flow detected at the outlet from the chamber using the PSM (see Fig. 1).

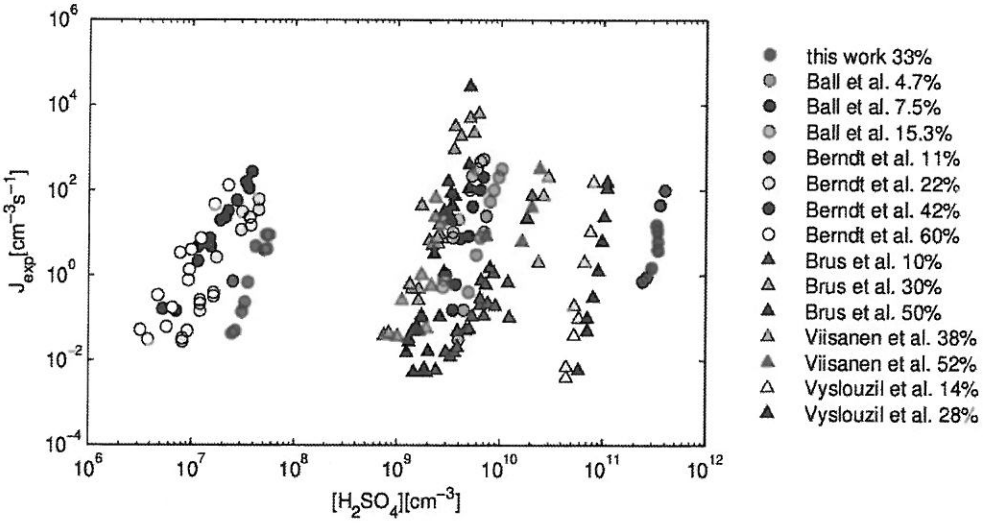

Fig. 1: Comparison of our data with previous studies - Dependence exp. nucleation rate on the concentration of H2SO4 at $T = 25$ ° C.

The obtained nucleation rates are roughly two orders of magnitude lower than those obtained by other authors on flow chambers with turbulent mixing. A shift of nucleation isotherm towards higher concentrations of H2SO4 may be caused by the high purity of the used substances, which are free of amines and other ingredients stabilizing critical clusters. We have shown that LCFT is a relevant tool for the measurement of nucleation rate of binary mixtures.

Acknowledgements:

The authors wish to thank for the support the work of the grant P105 / 12/0664 of Grant Agency of the Czech Republic and the EU project 7. RP EU Marie-Curie ITN HEXACOMM č. 315760.

Literature:

Reiss, H., Margolese, D.I., Schelling, F.J., Experimental study of nucleation in vapor mixtures of sulfuric acid and water. *J. Colloid Interface Sci.*, 56, 511–526, (1976).

Katz, J.L., Ostermier, B.J., Diffusion cloud-chamber investigation of homogeneous nucleation, *J. Chem. Phys.*, 47, 478, (1967).

Kulmala, M. and Laaksonen, A.: Binary nucleation of water-sulfuric acid system: Comparison of classical theories with different H2S04 saturation vapor pressures, *J. Chem. Phys.*, 93, 696–701, (1990).

Wyslouzil, B.E., Seinfeld, J.H., Flagan, R.C., Okuyama, K., Binary nucleation in acid water-systems.2. sulfuric-acid water and a comparison with methanesulfonic-acid water, *J. Chem. Phys.* 94, 6827 (1991).

Wagner, P.E., Anisimov, M.P., Evaluation of nucleation rates from gas flow diffusion chamber experiments, *J. Aerosol Sci.* 24, S103-S104 (1993).