Modelling Mixing and Particle Formation in Supercritical Antisolvent (SAS) Processes

J. J. Choi and A. Kronenburg*
Institut für Technische Verbrennung, Universität Stuttgart, Germany
jung-jin.choi@itv.uni-stuttgart.de and kronenburg@itv.uni-stuttgart.de

Abstract

The fluid properties at supercritical conditions such as small thermal diffusivity and close-to-zero surface tension favour complete mixing between solution and antisolvent and thus facilitate the production of ultra-fine nano-sized particles with a narrow size distribution. The supercritical antisolvent (SAS) process exploits these properties by injecting a liquid jet with a solute into a fluid at supercritical conditions, usually CO_2, that acts as antisolvent for the solute. The SAS process is an inherently complex system, that involves the interplay of equilibrium/non-equilibrium thermodynamics/transport properties of supercritical fluids with real gas effects as well as the hydrodynamics for turbulent mixing. In addition, the particle dynamics including nucleation and growth coupled with supercritical thermodynamic properties and mixing at molecular scale need to be carefully examined and modelled. Of all these processes, the mixing is pivotal in the SAS process. It is of primary importance for the formation of supersaturation conditions that lead to particle nucleation and thus controls the dynamics of particle production and evolution. A Large-Eddy Simulation (LES) method is used for the simulation of the flow and mixing fields. Compressibility effects cannot be neglected, and a compressible Navier-stokes formulation employing an $AUSM^{+}$-up scheme needed to be implemented for the description of the compressible but low Mach number flows predominant in SAS processes. The Peng-Robinson equation of state is used to describe real gas effects, and the particle evolution and size distribution are modelled by the population balance equation using the method of moment approach. In LES only the large scale processes flow structures are spatially and temporally resolved. The small, sub-grid processes, in particular the micro-mixing that locally affects supersaturation and particle nucleation, and the interactions between turbulence and particle dynamics need to be modelled. We introduce a conditional moment closure (CMC) method as a sub-grid model to account for the unresolved effects of turbulence on particle nucleation and growth. CMC is a sub-grid model for non-premixed combustion [1] where chemical reaction is strongly dependent on micro-mixing, and the same modelling ideas can be used for the prediction of the nucleation term. Preliminary investigations of particle formation of paracetamol in the mixture of ethanol and CO_2 at T=314K and P=16MPa have been carried out. Computations with varying mole fractions of CO_2 using the equilibrium thermo-hydrodynamics assumption show good agreement with experimental results from [2]. The poster will present preliminary simulation results from a ternary system of paracetamol-ethanol-CO_2, where ethanol and the solute are injected into the mixing chamber by a jet. The LES of the hydrodynamics is fully coupled with the solution of the transport equations of the conditionally averaged moments of the particle dynamics and size distribution. The effects of sub-grid micro-mixing (and essentially turbulence) on particle nucleation and growth can be quantified, and the results can be compared with earlier experiment by Bristow *et. al.* [2].

References

[1] Navarro-Martinez, S., Kronenburg, A. and diMare, F., *Flow, Turbulence and Combustion* 75: 245-274 (2005).
[2] Bristow, S., Shekunov, T., Shekunov, B. and York, P., *Journal of Supercritical Fluids* 21:257-271 (2001).

*Corresponding author: kronenburg@itv.uni-stuttgart.de