

Evaluation of transient kinetics during the supercritical CO₂ drying

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Production of aerogels starting from almost all precursors requires a supercritical drying step to preserve the mesoporous structure of the gel. Such systems include most of the organic gels derived from synthetic polymers and almost all biopolymers. To understand, optimize and scale-up the supercritical CO₂ drying process, several mathematical models have in the past been developed and fitted to experimental data. Since there is a lack of correlations describing the convective-dispersive transport of the solvent during the supercritical drying process, the existing models assume an ideal behavior of the bulk domain. Thus, they are likely to be overfitted and may not represent the real drying dynamics but rather biased and thus of limited practical applicability.

Nowadays, most of the measurement systems track the concentration of solvent at the outlet of the autoclave. The obtained signal is a convolution of the mass transport occurring in the gel and the convection and dispersion of the solvent from the surface of the gel to the detector. The convective-dispersive transport of the solvent through the system dominates the signal, hiding the true nature of the drying dynamics. Decoupling the main transport processes has been proven to be a difficult task due to the transient kinetic nature of the process. In order to decouple the drying kinetics and to generate data that is equipment independent, new correlations describing the system dynamics for the deconvolution of the experimental signal need to be developed.

In this project we are measuring the residence time distribution of the process at different compositions, pressures, temperatures and flow rates. The dispersion coefficient and mean residence time which describe the system dynamics are correlated using the Aris-Taylor equation. The Aris-Taylor equation has been proven to describe the system at the measured conditions. It was found out that the mean residence time at process conditions relevant to the industry is in the range between 500 s and 2000 s together with dispersion coefficients 0 to 40 times larger than the molecular diffusion coefficient of the system.

Also, during the process of supercritical drying two different dynamic regimes take place. At the beginning of the process the large flux out of the gel together with the large density difference between pore fluid and bulk fluid form a stratified system. At later stages of the process a more dispersive behavior is observed. Due to the non-ideal behavior of the bulk domain and large gravity influence, the bulk dynamics play a key role on the kinetics of the supercritical CO₂ drying.