Supercritical Assisted Atomization: a Performance Comparison between Laboratory and Pilot Scale

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Summary
The SAA (Supercritical Assisted Atomization) process has been developed up to pilot scale. Some experiments have been performed on laboratory and pilot apparatuses to compare the performance of the process on different scales. Zyrconil nitrate and rifampicin have been used as model compounds. Different concentrations of zyrconil nitrate in water and rifampicin in methanol have been tested. Particles of zyrconil nitrate with mean diameters ranging between 0.3 and 0.6 μm and of rifampicin ranging between 0.5 and 2.5 μm have been obtained on both apparatuses.

Introduction
Supercritical assisted atomization has been recently proposed for the generation of particles of fine chemical compounds in the micronic range and with controlled particle size distributions. It is based on the solubilization of controlled quantities of supercritical CO_2 in liquid solutions containing a solid solute and on the subsequent atomization of the ternary solution through a nozzle. SAA process efficiency is based on a two-step atomisation mechanism: primary droplets are formed at the exit of the nozzle. Then, CO_2 is released from the primary droplets and secondary, smaller droplets are produced. Several compounds have been succesfully micronized on the laboratory scale: superconductor and catalyst precursors, ceramics, and pharmaceutical compounds, using some different liquid solvents such as water, methanol, and acetone[1]. Particles ranging between about 0.5 and 4 μm have been as a rule obtained; amorphous or crystalline particles have been produced depending on the chemical nature of the solid product and on the temperature of the precipitation vessel. In this work, a pilot scale plant has been developed to assess if SAA large scale processing is feasible. The design of the new plant has been finalized to obtain a ten times increase of the production rate.

Experimental
The apparatus used for SAA experiments mainly consists of three feed lines used to deliver supercritical CO_2, the liquid solution and an inert gas. CO_2 and the liquid solution are sent to a high-pressure vessel (saturator), loaded with stainless steel perforated saddles using two high-pressure pumps. An adequate residence time produces the dissolution of the gaseous stream in the liquid. Then, a spray is produced delivering the ternary mixture to an 80 μm diameter stainless steel nozzle in the case of the laboratory scale plant and a 200 μm diameter nozzle in case of the pilot plant. Droplets are formed in the precipitator (I.V. 3 dm^3 blench scale, 12 dm^3 pilot scale) that operates at atmospheric pressure. A flow of heated N_2 is also delivered to the precipitator with the scope of favouring the evaporation of the liquid solvent. Samples of the processed powder were observed by Scanning Electron Microscopy (SEM). Particle Size Distributions (PSDs) were measured considering about 1000 particles in each calculation. More details on the SAA apparatus and experimental procedures were previously published[2].

Results and discussion
Systematic experiments were performed to micronize zyrconil nitrate, a catalyst precursor, in H_2O at different concentrations of the solute using the bench plant and repeating the same

experiments on the pilot plant at the same operating conditions. The main difference between the two apparatuses is the heating system of the precipitation vessel. In the case of the bench plant we used electric heaters bended around the vessel, in the case of the pilot plant we used a water jacket. This arrangement assures a better heating efficiency and a smoother temperature distribution in the precipitator. Both apparatuses yield amorphous spherical particles with a relatively narrow particle size distribution (Figure 1).

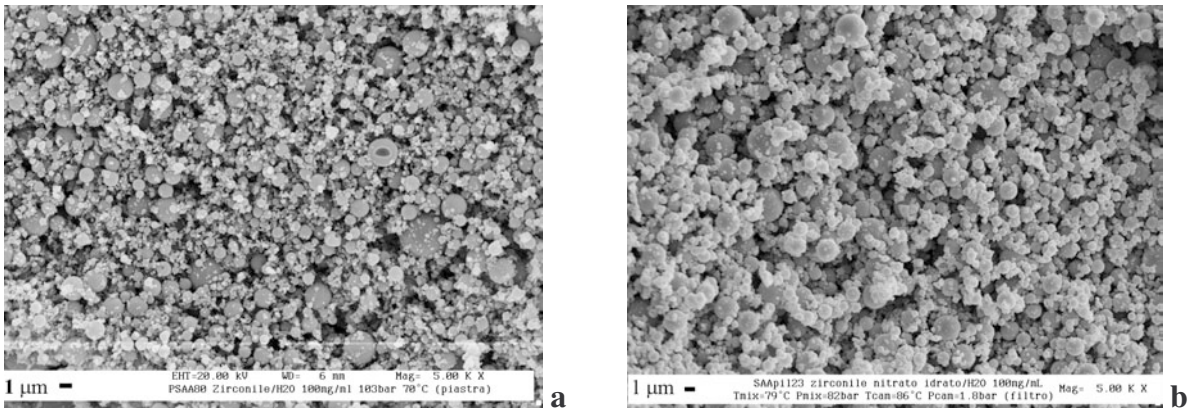

Figure 1 : Comparison of SEM image of zyrconil nitrate particles precipitated from H_2O at 100mg/mL (**a**) laboratory scale plant, (**b**) pilot scale plant

We observed only a slight broadening of the particle size distribution in the case of the pilot plant (see Figure 2). This behaviour has been observed in all the experiments. Varying the solution concentration from 50 to 100 mg/mL we observed an increase of the particle size from 0.63 to 0.78 µm in the case of the bench scale plant and from 0.63 to 0.87 µm in the case of the pilot plant.

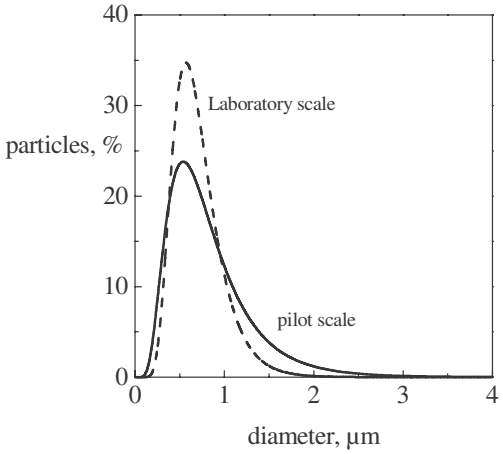

Figure 2 : PSDs in terms of particle number percentages of zyrconil nitrate from H_2O at 100mg/mL

The good results obtained for the system zyrconil nitrate-H_2O were confirmed micronizing a compound of different chemical nature, rifampicin, a pharmaceutical principle, using methanol as liquid solvent. For what concerns the pilot plant productivity, the best performance has been 200g/h of precipitated material.

References

1. E. Reverchon, Ind. Eng. Chem. Res., 41, 2405 (2002)
2. E. Reverchon, G. Della Porta, J. Supercrit. Fluids, 26, 243 (2003)