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# Laminar jet dispersion and jet atomization in pressurized carbon dioxide

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## Abstract

Laminar jet dispersion and more particularly jet atomization of water, methylene chloride and ethyl alcohol into pressurized carbon dioxide are investigated under experimental conditions of pressure, temperature and liquid flow rate commonly used for supercritical anti solvent (SAS) precipitation processes. The different modes of dispersion are characterized as a function of the jet velocity. Axisymmetrical, asymmetrical and atomized jets are observed at 308 K, under a pressure ranging from 6 to 9 MPa, and for a liquid jet velocity ranging from to 0.14 to 8.02 m $s^{-1}$. A new correlation is given between the jet Reynolds number and the Ohnesorge number, determining the transition to the atomization regime. Experimental values of dynamic interfacial tensions are used to elaborate the correlation.

*Keywords:* Dynamic interfacial tension; Supercritical fluids; Liquid jet dispersion; Jet disintegration; Jet atomization

## 1. Introduction

Supercritical antisolvent precipitation processes have been developed for 15 years and applied to several high added value molecules, and in particular to a lot of pharmaceutical products [1]. Their strong potential for forming ultrafine particles has been widely proved and the current stake is to control characteristics such as, among others, the particle size, the particle size distribution, their polymorphic nature and crystal habit. Basically, these precipitation processes consist in putting in contact a supercritical fluid or a dense gas, which plays the role of the antisolvent, with a liquid solution composed of a solvent and of the solute to be precipitated. The simultaneous mass transfers of the dense gas and of the solvent from one phase to the other induce the supersaturation of the solute, which precipitates. Different antisolvent processes have been developed which differ mainly from one another by the contacting mode of the two phases, the injection device, the batch or semi-continuous processing mode and the transient or steady state conditions.

Nevertheless, the most used operating mode consists in dispersing the liquid solution into a continuous supercritical phase containing the antisolvent, in a semi-continuous mode. This operating mode is involved in the following processes [1]: precipitation with a compressed fluid antisolvent (PCA), supercritical antisolvent (SAS), aerosol solvent extraction system (ASES) and solution enhanced dispersion by supercritical fluids (SEDS).

Except for the last patented process for which a specific injection allows the pre-mixing of the liquid solution with the supercritical fluid at high Reynolds numbers [2], the liquid solution is generally dispersed through a capillary or a nozzle and thus forms a jet or a spray according to the experimental conditions and set-up.

A good control of such processes requires the knowledge of: (a) the mechanisms of jet disintegration, (b) the mass transfer kinetics between the jet (and/or the resulting entities) and the continuous phase, (c) the fluid phase equilibrium of the system, (d) the nucleation and growth mechanisms and kinetics of the crystals.

This study deals with the first point: the mechanisms of jet disintegration. The jet dispersion of a liquid phase into dense carbon dioxide is investigated using water, ethyl

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**Nomenclature**

| | |
|---|---|
| $\rho_\mathrm{l}$ | liquid phase density (kg m$^{-3}$) |
| $\rho_{\mathrm{CO_2}}$ | carbon dioxide density (kg m$^{-3}$) |
| $\mu_\mathrm{l}$ | liquid phase hydrodynamic viscosity (Pa s) |
| $\nu_\mathrm{l}$ | liquid phase kinematic viscosity (m$^2$ s$^{-1}$) |
| $\sigma$ | interfacial tension (N m$^{-1}$) |
| $d_0$ | Capillary internal diameter (m) |
| $d_\mathrm{j}$ | jet diameter (m), $d_\mathrm{j} = d_0/(0.485\,C + 1)$ if $C < 0.62$ and $d_\mathrm{j} = \mathrm{d}_0/(1.51\,C^{0.5} + 0.12)$ if $C > 0.62$, with $C = (g\,\rho_\mathrm{l}\,d_0^2)/\sigma$ |
| $u_\mathrm{l}$ | jet velocity (m s$^{-1}$) |
| $Re_\mathrm{l}$ | jet Reynolds number, ratio of inertial force to viscous force, $Re_\mathrm{l} = \rho_\mathrm{l}\,u_\mathrm{l}\,d_0/\mu_\mathrm{l}$ |
| $We_\mathrm{l}$ | jet Weber number, ratio of inertial force to superficial force, $We_\mathrm{l} = \rho_\mathrm{l}\,u_\mathrm{l}^2\,d_0/\sigma$ |
| $Oh_\mathrm{l}$ | jet Ohnesorge number, $Oh_\mathrm{l} = (We_\mathrm{l})^{0.5}/Re_\mathrm{l} = \mu_\mathrm{l}/(\rho_\mathrm{l}\,\sigma\,d_0)^{0.5}$ |
| $Oh_\mathrm{l}^*$ | modified Ohnesorge number, $Oh_\mathrm{l}^* = (\mu_\mathrm{l}/(\rho_\mathrm{l}\,\sigma d_0)^{0.5})(\rho_\mathrm{g}/\rho_\mathrm{l})^{0.5}$ |
| $Z^*$ | modified dimensionless number, $Z^* = (\mu_\mathrm{l}\,u_\mathrm{l}/\sigma)(\rho_\mathrm{g}/\rho_\mathrm{l})^{0.5}$ |

alcohol and methylene chloride at 308 K under immiscible as well as miscible conditions. The experimental conditions used are typically those of SAS processes, the only difference being the absence of solid solute in the liquid phase.

The theory of liquid dispersion briefly exposed below is widely used in this work to describe and interpret our experimental results. Before presenting our approach, we will give an overview of liquid jet dispersion into dense fluids.

### *1.1. Theory*

Three principal regimes of liquid phase dispersion are observed for liquid–liquid and liquid–gas systems, depending on the liquid flow rate [3]: (i) the dripping regime, in which the droplets are formed at the outlet of the capillary; (ii) the laminar regime, in which the jet has a smooth and continuous aspect before a break-up zone where there is quasi-periodic emission of droplets identical in size; (iii) the turbulent regime, in which the jet surface presents irregularities and the resulting droplets have different sizes.

The experimental conditions and set-up generally used in SAS processes (liquid velocities, nature of the organic solvent) lead to the formation of laminar jets and there are several modes of dispersion for laminar jets, essentially depending on the liquid phase velocity. The appearance of the unstable modes of laminar jets is under the control of the competing effects acting on the jet. The capillary, inertial, aerodynamic and viscous forces are involved whereas gravity effects are neglected.

For the lower Reynolds numbers, the first mode observed for a laminar jet is characterized by axially symmetrical disturbances, named varicoses, producing the jet break-up. The first theory on the mechanism of drop formation resulting from an axisymmetrical jet has been proposed by Rayleigh [4]. The Rayleigh analysis assumes that surface tension is the chief force controlling the break-up of an axisymmetrical jet. Weber extended Rayleigh's theory to the break-up of a viscous jet, when both viscous and inertial forces offer significant resistance [5].

For higher Reynolds numbers, the inertial forces compete with the capillary forces. The jet break-up zone follows a lateral motion with increasing amplitude, which leads to the formation of an asymmetrical jet, which can be either sinuous or helicoidal.

Then, when the flow rate goes beyond a certain value, the aerodynamic effects become quite strong and the jet is atomised. The jet presents a smooth region at the outlet of the orifice, usually with a length of about 15 times the jet diameter (see nomenclature), and after a break-up zone there is a cone of atomized solution. The break-up is highly chaotic. There is a sharp transition from the helicoidal jet dispersion mode to the atomization jet dispersion mode. Regardless of the type of flow, disintegration or atomization are favoured by air friction and the break-up distance decreases as the pressure increases.

A laminar jet can be characterized by: (i) *the jetting velocity* (velocity at the transition from the dripping to the laminar jet regime), (ii) *the jet length* (length of coherent portion of liquid jet) or break-up length, (iii) *the critical jet velocity* (velocity at the atomization transition).

The effects of the liquid properties and jet velocity on the mechanism of jet disintegration can be investigated with an approach based on a dimensional analysis. When a liquid jet is dispersed in a non dense fluid, the break-up mechanism is dependent on the jet diameter, jet velocity, liquid density, interfacial tension and viscosity. The break-up mechanism can be predicted using dimensionless numbers: the Reynolds number and the Ohnesorge number ($Oh_\mathrm{l} = \mu_\mathrm{l}/(\sigma\,\rho_\mathrm{l}\,d_0)^{0.5}$). At atmospheric pressure, empirical correlations can be used to classify the jet disintegrations. The Ohnesorge chart describes the dispersion modes as a function of the Reynolds number and the liquid properties [6]. Three different zones are distinguished: the Rayleigh break-up zone, a second zone where the jet has a sinuous or a twisted shape and finally the atomization zone. Qualitatively, when the density of the continuous phase increases, this chart is no longer valid and the boundaries between the different zones shift to lower Reynolds numbers. As a matter of fact, an increase in the continuous phase density leads to an increase in the aerodynamic effects acting on the jet.