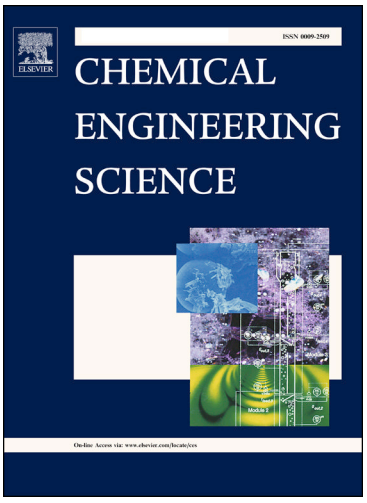

A droplet-based millifluidic method for studying ice and gas hydrate nucleation

Dyhia Atig, Abdelhafid Touil, Manuel Ildefonso, Laurent Marlin, Patrick Bouriat, Daniel Broseta

PII: S0009-2509(18)30569-4
DOI: https://doi.org/10.1016/j.ces.2018.08.003
Reference: CES 14418

To appear in: *Chemical Engineering Science*

Received Date: 1 May 2018
Revised Date: 20 July 2018
Accepted Date: 3 August 2018

Please cite this article as: D. Atig, A. Touil, M. Ildefonso, L. Marlin, P. Bouriat, D. Broseta, A droplet-based millifluidic method for studying ice and gas hydrate nucleation, *Chemical Engineering Science* (2018), doi: https://doi.org/10.1016/j.ces.2018.08.003

A droplet-based millifluidic method for studying ice and gas hydrate nucleation

Dyhia Atig, Abdelhafid Touil, Manuel Ildefonso, Laurent Marlin, Patrick Bouriat, Daniel Broseta

CNRS /TOTAL/UNIV PAU & PAYS ADOUR/ E2S UPPA, LABORATOIRE DES FLUIDES COMPLEXES ET LEURS RESERVOIRS-IPRA, UMR5150, 64000, PAU, France

Abstract

We design and implement a simple and versatile droplet-based millifluidic method for investigating nucleation and growth processes in crystal-forming aqueous systems. It consists in generating and storing in a transparent capillary a train of identical and regularly-spaced droplets of an aqueous phase in a carrier oil phase, and then in video-monitoring crystal nucleation and subsequent growth and melting events as temperature and/or pressure are varied. Compared to previous investigations, the novelty is the possibility of working with aqueous solutions containing dissolved gas under controlled pressure, thus opening the way to gas hydrate studies. In the absence of dissolved gas, *i.e.*, at ambient pressure, we observe ice nucleation to be weakly promoted by titanium oxide and montmorillonite particles, and strongly promoted by silver iodide, in agreement with literature results. Ice nucleation is also promoted when the carrier oil is more wetting towards the capillary, which is the case for fluorinated oil as compared to n-hexane. With cyclopentane, a hydrate-former, as the carrier oil, and dissolved CO_2, also a hydrate-former and a "help gas" for cyclopentane hydrate formation, we find evidence for hydrate nucleation along with that of ice, and monitor the different solid phases as temperature varies.

Keywords: millifluidics, ice, gas hydrates, nucleation, crystallization kinetics