Investigation of the Supersaturation of the Water-Rich Phase after Gas Hydrate Dissociation by Raman Spectroscopy

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Gas hydrates are crystalline solids composed of a three-dimensional network of hydrogen bonded water molecules stabilized by the inclusion of gas molecules. Their formation can be a boon or bane: The undesired formation of gas hydrates can cause pipeline blockage or even destruction, whereas considerable amounts of carbon dioxide could be captured in the form of gas hydrates. After the complete melting of hydrate, the generated water-rich phase often exhibits a so-called 'memory' of the hydrate structure, which facilitates the re-formation of gas hydrate. There are two possible explanations in literature for this phenomenon. One is that residual hydrate structures remain in the water-rich phase, the other one is that the water-rich phase contains excess hydrate guest molecules (here CO_2). In our experimental Raman studies, we observed an increased amount of carbon dioxide in the water-rich liquid phase after complete melting of the hydrate. A shift in the Raman signal indicates that the carbon dioxide does not exist in a dissolved form, but in a liquid state, potentially in the form of liquid microbubbles. In this work we investigate the stability of this micro emulsion, by varying different parameters during the heating phase, e.g. superheating, time, temperature gradient applied and the stirrer speed. With these measurements, it is possible to determine the conditions at which the memory effect still exists or can be avoided depending on whether gas hydrate formation is desired or should be prevented.

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