Contribution ID: **29** Type: **Poster**

Ice freezing and gas hydrate formation in water-in-oil emulsions

Tuesday 9 January 2018 12:35 (1h 30m)

Experimental investigations carried out in this work allowed us to elucidate a number of new aspects in the ice freezing and gas hydrate formation processes in water-in-hydrocarbon emulsions. Various types of oils, including one with different biodegradation levels, and n-decane were used as disperse media. As it has been shown there are several hydrate and ice formation ways in the emulsions. (1) Primary nucleation of a solid phase on any of water droplet leads to a rapid secondary nucleation in a region of the emulsion adjacent to the droplet. The thermal effect is characterized by a rapid rise in temperature at the leading edge and a slow decrease at the trailing edge. Since a significant thermal effect can be observed by thermal methods, it makes possible to use them to study primary nucleation. (2) Primary nucleation of a solid phase on any of water droplet leads to a rapid secondary nucleation in a small number of neighboring droplets. Then the secondary nucleation decays. In this case, there is one or more small exothermal effects on the thermal curve. (3) Primary nucleation on any of the water droplet leads to an occurrence of rapid secondary nucleation only in a certain fraction of cases (if nucleation has occurred in a droplet agglomerate for instance). In this case, the thermal effect can be indistinguishable from the case (1). However, the statistics of the thermal effects appearance reflects not only the dynamics of the primary nucleation, but also the structural features of this emulsion. (4) Primary nucleation of the solid phase on any of the water droplet leads to a slow secondary nucleation in some region of the emulsion adjacent to this drop. This type of nucleation process relates to hindered germination of the crystallites through the medium. Release of heat in this process seems to will increase due to the involvement of free water droplets into it until some moment. In this case, an extended exothermal effect will appear on the thermal curve, the shape of which may be close to a symmetric. (5) Primary nucleation of the solid phase on any water droplet never leads to the secondary nucleation process. In this case, latent crystallization of the solid phase occurs during the emulsion cooling. A noticeable exothermal effect is manifested only about a point of homogeneous ice nucleation. It should be noted that the presence of secondary nucleation can have a significant effect on the agglomeration of hydrate and ice particles in oil suspensions. The germination of a crystal from one water particle to another indeed creates a mechanical connection between them. Thus, the particles involved in the secondary nucleation process will be agglomerated to a great extent. It was also shown that the appearance of biodegradation products in oil leads to an easier process of secondary nucleation of gas hydrates and ice in the emulsion and slightly increases the probability of hydrate nucleation.

Acknowledgments (This work was supported by Russian Science Foundation grant No 17-17-01085)

Significance statement

The results of this work extend comprehension of the mechanisms of ice freezing and gas hydrates formation in water-in-oil emulsions.

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Session Classification: Poster & Lunch