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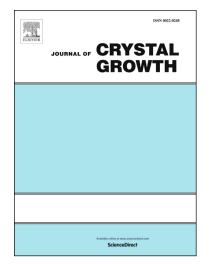
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## **ACCEPTED MANUSCRIPT**

## Visual Observation of Gas Hydrates Nucleation and Growth at a Water – Organic Liquid Interface

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#### **Abstract**

Visual observation of nucleation sites of methane and methane-ethane-propane hydrates and their further growth in water – organic liquid – gas systems with / without surfactants was carried out. Sapphire Rocking Cell RCS6 with transparent sapphire cells was used. The experiments were conducted at the supercooling  $\Delta \Gamma_{\text{sub}} = 20.2^{\circ}\text{C}$ . Decane, toluene and crude oils were used as organics. Gas hydrate nucleation occurred on water – metal – gas and water – sapphire – organic liquid three-phase contact lines. At the initial stage of growth hydrate crystals rapidly covered the water – gas or water – organics interfaces (depending on the nucleation site). Further hydrate phase accrete on cell walls (sapphire surface) and into the organics volume. At this stage, growth was accompanied by water «drawing out» from under initial hydrate film formed at water – organic interface. Apparently, it takes place due to water capillary inflow in the reaction zone. It was shown that the hydrate crystal morphology depends on the organic phase composition. In the case of water-in-decane emulsion relay hydrate crystallization was observed in the whole sample, originating most likely due to the hydrate crystal intergrowth through decane. Contacts of such crystals with adjacent water droplets result in rapid hydrate crystallization on this droplet.

Keywords. B1. gas hydrate; B1. ice; A1. morphology; B1. decane; B1. crude oil; A1. nucleation

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