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Physicochemical and hydrodynamical Controls on Methane Bubble Dissolution within the Hydrate Stability Field

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We recently reported on extended lifetimes of methane bubbles within the hydrate stability field as a consequence of the formation of a hydrate skin (1), based on *in situ* measurements of methane and argon bubble dissolution in the depth range from 400–800 m. These observations were extended to depths between 900 m and 1800 m. Single bubbles were injected from ROV Ventana into an attached, back-illuminated, flow-through imaging box. The ascent of individual bubbles within the imaging box was then recorded with *Ventana'*s HDTV camera system by piloting the 3-ton vehicle upward, at the exact rise rate of the bubble, for up to 400 m of vertical transit.

The observed rise rates were of the order of 30cm/sec for all depths. Post-dive analysis of the HDTV tapes allowed detailed measurements of the bubble shrinking rates. The results of the earlier and new experiments lead to the following conclusions: (A) Methane bubbles released below the hydrate stability field showed markedly enhanced lifetimes, attributed to hydrate skin formation. The change in radius with time, dr/dt, varied from -7.5 micrometer/s above the hydrate stability field to about -1 micrometer/s at the deepest release depth. (B) Bubble lifetime within the hydrate stability field increased with distance in P-T space from the hydrate phase boundary. (C) Before hy-

drate skin nucleation, dr/dt for CH4 bubbles was comparable to dr/dt above the hydrate stability field. Although variable, the nucleation onset time generally decreased with distance from the hydrate phase boundary (super-pressurization and super-cooling with respect to hydrate formation). (D) Even before the onset of nucleation, a slight decrease of dr/dt with increasing depth can be observed.

We relate these findings to formal calculations of the solubility and density of methane as a function of pressure, and reviewing the effect of super-pressurization on hydrate nucleation. Deviation from ideal gas law and Henry's law were implemented into the bubble dissolution model of Leifer and Patro (2) and the effect of surface immobility after hydrate formation was accounted for. The simulations suggest that the modifications above improve the ability of the model to predict methane bubble behavior within the gas hydrate stability field, but also points to an overestimation of the dependence of the gas transfer rate on bubble size for 1000 micrometer $<$ r $<$ 3500 micrometer in most bubble dissolution models used today.

References: (1) Rehder et al. Geophys. Res. Lett., 29, 1731, (2002); (2) Leifer and Patro, Cont. Shelf Res., 22, 2409 (2002).