



	Experiment title: High energy reflectivity study of CO ₂ hydrate formation and demixing at liquid-liquid interfaces	Experiment number: SI-2146
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On the Spontaneous Formation of Clathrate Hydrates at Water–Guest Interfaces
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Abstract:

The formation of hydrates, cage-like water-gas structures, is of tremendous importance both in industries and research. Although of major significance, the formation process is not completely understood so far. We present a comprehensive study of hydrate formation at liquid–liquid interfaces between water and isobutane, propane, carbon dioxide, and at the liquid–gas interface between water and xenon. We investigated the structure of these interfaces under quiescent conditions in situ by means of X-ray reflectivity measurements both inside and outside the zone of hydrate stability. At the interfaces between water and liquid alkanes, no evidence for a structural change was found. In contrast, the accumulation of guest molecules inside nanothick interfacial layers was observed at the water–xenon and liquid–liquid water–CO₂ interfaces. We show that only those systems initially exhibiting such guest-enriched interfacial layers developed into macroscopic gas hydrates within our observation times (~12 h). Therefore, these layers act as triggers for the spontaneous formation of macroscopic hydrates.