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Clathrate Hydrates on Titan: Investigating Partial Dissociation and Substitution processes

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Titan, Saturn's largest moon, exhibits a unique environment characterized by a dense atmosphere, active organic chemistry, and a frigid surface shaped by hydrocarbon flows. Clathrate hydrates have long been inferred to constitute a major crustal component of its icy shell, and are believed to play a critical role in replenishing atmospheric methane through dissociation or substitution processes. Clathrates of methane and tetrahydrofuran (THF), its ambient pressure analog, undergo partial dissociation at temperatures as low as ~200 K in the presence of ammonia, a strong antifreeze agent likely present in Titan's outer ice shell. Interactions with NH_3-bearing liquids in Titan's subsurface could thus potentially trigger clathrate dissociation. In addition, the ternary H_2O-THF-NH_3 system has been shown to exhibit a great chemical complexity with the reported formation of several crystallization stages upon cooling, including a binary THF-NH_3 clathrate and an unknown THF-NH_3-rich phase.

In order to establish the effects of NH_3 on clathrates and better assess their contribution to methane outgassing and exchange processes on Titan, we conducted laboratory and modeling studies. In particular, we investigated the ternary H_2O-THF-NH_3 system and the CH_4-C_2H_6 replacement kinetics. Our X-ray diffraction data confirmed the partial dissociation of THF clathrates in aqueous solutions of ammonia between 200 and 270 K, and enabled the characterization of the unknown THF-NH_3-rich phase, which was previously observed below 220 K. Our analysis suggests a trigonal crystal system for this novel compound, with a unit cell volume of 1617 $Å^3$ at 90 K.

Further Raman spectroscopy investigations showed that ethane can replace methane in the large sI clathrate cavity, providing a mechanism for methane release and ethane trapping in Titan's subsurface. These findings support the hypothesis that dynamic processes involving clathrates contribute to the exchange of volatiles between the surface and atmosphere on Titan.