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Gas selectivity in mixed gas hydrates formed from pressurized ice.

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In addition to their various applications in the environment, energy and technology fields, gas hydrates are supposed to be involved in the formation of planetesimals, comets and other planets, such as Titan or Enceladus, both being Saturn's moons. For instance, CO gas hydrate was one of the hydrates especially highlighted in the first work outlining this hypothetical hydrate formation in astrophysical conditions. Moreover, recent theoretical works suggest that the nitrogen depletion observed on the Jupiter family comet 67P/Churyumov-Gerasimenko might be due to N2 encapsulation within hydrates and so assume that 67P agglomerated from these crystalline compounds. The preferential trapping of carbon monoxide with respect to nitrogen inside mixed gas hydrate allow to determine the temperature range of the cometary grains formation in the protosolar nebula. The present work is dedicated to the experimental investigation of such a preferential encapsulation by means of Raman scattering in various mixed gas (CO, CO2, N2) hydrates. Beyond the astrophysical interest, the molecular selectivity in mixed gas hydrate plays an important role in many areas such as process engineering or geoscience.
The investigation is based on the study of the gas selectivity in gas hydrates formed with high driving force, i.e. by ice exposure to different gaseous mixtures (in various pressure and temperature conditions). These studies have been carried out on the pure gas hydrates (CO, N2 and CO2) as well as on the corresponding mixed gas hydrate. Such a series of gaseous molecules offer the opportunity to explore the impact of various physical-chemistry parameters onto the gas selectivity by forming hydrates. Many factors, such as steric hindrance or thermodynamic conditions of formation, govern this selectivity. CO2 possesses a strong water affinity while CO and N2 do not. CO and N2 molecules have similar steric hindrance and close hydrate formation P-T conditions. CO is the only considered guest molecules possessing a dipolar moment. Raman spectroscopy results have put forward a clear preferred encapsulation of carbon monoxide molecules compared to nitrogen molecules during the mixed gas hydrate formation. Similar preferential encapsulation is observed in the case of CO2 molecules with respect to CO and N2 molecules. Moreover, neutron powder diffraction data have shown phase mixtures involving various structures (so-called type I and type II) and structural metastability. This whole set of results will be discussed in conjunction with DFT calculations, kinetic formation conditions and structural metastability properties evidenced in these gas hydrates.

Significance statement

This work (Claire Petuya's PhD) provides the first experimental evidences of preferred gas trapping of CO in CO/N2 mixed gas hydrate as well as of vibrational signatures of guest partitioning for the considered mixed gas hydrates, formed from pressurized ice.

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