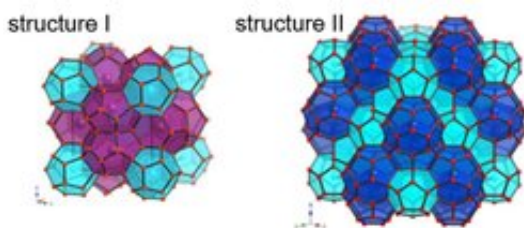


# Neutrons reveal fast methane translational diffusion at the interface of two clathrate structures

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Unit cells of clathrate structure I and clathrate structure II. Different types of polyhedral cages are represented in different colours. Credit: Ranieri et al., *Nature Comm.*

Gas clathrate hydrates are ice-like solids, in which gas molecules or atoms are trapped inside crystalline frameworks formed by water molecules. They have attracted considerable attention over the last decade for their potential as a geo-organic fuel resource, as they naturally form in large quantities within marine sediments and below continental permafrost. Exchanging the guest gases in existing methane hydrate deposits with CO<sub>2</sub> has also been recently indicated as a promising two-in-one approach of energy recovery and concomitant carbon dioxide mitigation.

On the molecular scale, gas clathrate hydrates are characterised by polyhedral water cages of different shapes and sizes. Those cages can be

combined in different ways to form a crystalline [structure](#). Four different structures are known so far and the two most common among them are generally referred to as clathrate structures I and II. The [gas molecules](#) or atoms occupy the centre of the cages; they are efficiently trapped and can barely escape.

The gas molecules or atoms are able to diffuse during non-equilibrium processes however. Such as formation or decomposition of a clathrate structure, or conversion between different structures. Their diffusion even plays a paramount role during such processes and some stages of these key phenomena are gas diffusion-limited. Understanding the mechanism of gas diffusion is thus crucial to prove the viability of all energy applications involving gas clathrate hydrates.

For clathrate hydrates of [methane](#), structure I is the thermodynamically preferred structure, and structure II is a metastable form that is kinetically favoured and transiently detected in the initial stages of the [hydrate](#) formation process. Remarkably, structure II can persistently coexist with structure I in methane clathrate hydrates under high pressure.

A recent study conducted by an international collaboration of research institutes (Institut Laue-Langevin (ILL), École Polytechnique Fédérale de Lausanne (EPFL), University of Göttingen, and University Pierre and Marie Curie) has exploited the exceptional persistence of structure II methane clathrate hydrates under high pressure, in order to measure the translational diffusion of methane molecules at the interface of clathrate structures I and II. Quasielastic neutron scattering experiments under a high pressure of 0.8 GPa were conducted at ILL to make these observations.

This was achieved by using a dedicated high-pressure apparatus: a Paris-Edinburgh press. The IN6 spectrometer at the ILL allowed the team to

measure the diffusion of [methane molecules](#) at the interface of the two clathrate structures on the picosecond time and Å length scales.

These findings reveal that methane translational diffusion is remarkably fast at the interface of clathrate structures I and II. The obtained diffusion coefficients are one order of magnitude higher than that of methane dissolved in water at low pressure, and a factor of two to three higher than that expected for pure bulk supercritical methane at comparable pressure and temperature.

Umbertoluca Ranieri, PhD student at ILL and EPFL, and lead author of this study says: "These results are important in improving our understanding of many fundamental non-equilibrium phenomena involving methane clathrate hydrates; for example, the replacement kinetics during gas exchange in case of conversion between the clathrate structures I and II. This knowledge will also help us to tackle important energy and environmental issues such as methane recovery from marine hydrates sediments and carbon dioxide capture in the future."

In addition to these advances, the high-pressure properties of methane clathrate hydrates are also crucial to planetary science. Clathrate hydrates of methane are believed to be the main methane-bearing phases in the interiors of some icy bodies of the Universe. Therefore, the results of this study could be used in future models of methane clathrate hydrates layers embedded in the cryosphere of such bodies, where [high pressure](#) is experienced.

**More information:** Umbertoluca Ranieri et al. Fast methane diffusion at the interface of two clathrate structures, *Nature Communications* (2017). [DOI: 10.1038/s41467-017-01167-2](https://doi.org/10.1038/s41467-017-01167-2)

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