

Clay sediments, a multiscale playground for hydrate formation.

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Gas hydrates are crystalline structures consisting of cages resulting from the hydrogen bonding of water molecules (host) enclosing relatively small gas molecules (guest), such as hydrogen, methane and other small hydrocarbons [1]. Gas hydrates occur naturally on continental margins and in the permafrost region where the pressure and temperature conditions are favorable for hydrate formation and stability [2,3]. Methane hydrates represent the largest natural reservoir of methane on Earth (with estimates ranging between 600-10000 GT). However, large uncertainties remain about the amount of methane trapped and the mechanisms governing methane fate, limiting conclusions about climate implications [4,5]. The Romanian sector of Black Sea is known to host a large amount of hydrates, and samples have been collected at around 700m water depth (70bars), where the seafloor temperature is close to 282K. Furthermore, the scientific cruises Ghass (2015) and Ghass2 (2021) conducted by Ifremer, revealed that the hydrates are formed in a clay-rich sediment. In the context of the study of the Black Sea hydrate deposit, the influence of the complex clay/sand matrix on the methane hydrate formation is studied using Raman spectroscopy, Inelastic Neutron Scattering and Neutron diffraction. Methane hydrate structural properties, spatial distribution in sediments and formation kinetics are investigated in presence of both natural and synthetic sedimentary environment and NaCl. These physical-chemistry results provide new insights on the formation mechanism, including original findings on the impact of clay structure and salinity on hydrate kinetics with investigations ranging from minutes to several months.

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