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Effects of feed gas composition and sediments on the formation process and the composition of the resulting mixed gas hydrate in Qilian Mountain permafrost, China

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Abstract

The occurrence of mixed gas hydrates has been proved in Qilian Mountain permafrost (QMP), China. This deposit is characterized by a shallow depth below thin permafrost, and a more complex gas composition compared to other hydrate reservoirs. Gas hydrate samples recovered from QMP contain a high percentage of hydrocarbons besides CH_4, suggesting that the hydrate belongs to structure II hydrate. The gas source is mainly composed of deep or crude oil-concomitant gases, occasionally mixed with microbial source in shallower depths. The reservoir contains a system of faults over the deep potential hydrocarbon reservoir. In this case, gas mixtures derived from deep hydrocarbon reservoir are being transported from the place of origin at greater depths through fractures to the shallower hydrate stability zone for the formation of gas hydrates.

It is demonstrated that the formation of a more-or-less homogeneous hydrate phase was difficult with the presence of a complicated gas mixture. In this work, two gas mixtures were used for the synthesis of mixed gas hydrates at 274 K, 3.2 MPa: one of them composed of 63 mol% CH_4, 15 mol% C_2H_6, 15 mol% C_3H_8, 7 mol% CO_2 and the other contained 71 mol% CH_4, 4 mol% C_2H_6, 8 mol% C_3H_8, 1 mol% iso-C_4H_{10}, and 1 mol% n-C_4H_{10} and 15 mol% CO_2. In additional experiments, a natural core sample was grinded into homogeneous powders with a grain size of less than 63 µm and added into the pressure cell for the formation of gas hydrates as well. The formation of gas hydrates was monitored applying *in situ* Raman spectroscopy combined with microscopic observations throughout the process. Through continuous characterization of the gas and hydrate phase over several days, important information about the gas enclathration on a µm-scale were revealed, indicating a competitive enclathration of guest molecules into the hydrate crystal lattice depending on their guest-to-cavity ratio and solubility. It is worth mentioning that no coexisting hydrate phases were observed during the formation of mixed gas hydrates without the presence of sediments. The results only demonstrated the heterogeneous character of the resulting hydrate phase with minor composition differences within the measuring points in one single crystal. Statistical calculations revealed that gas hydrate formation was promoted with the presence of sediments. Interestingly, a coexisting solid phase in the system with QMP sediments was also observed, which depicted totally different Raman spectra showing only CH_4 signals as compared to those from typical sII hydrate crystals. The existing hydrophobic minerals such as plagioclase or organic compounds in QMP sediments might be a possible reason as they could perturb the arrangement of water molecules in specific areas. In these areas, the nucleation and growth of hydrates might be retarded or even prevented. These findings are of importance to understand the mixed gas hydrate system in QMP and may suggest the potential effects of local sediments

Keywords: Mixed gas hydrates, Raman spectroscopy, Qilian Mountain permafrost, Gas composition, Sediments