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Molecular dynamics study of mineral impact on mass transport and hydrate formation

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Extended molecular dynamics studies were performed in several layered systems containing carbon dioxide hydrate. The hydrate slab was composed of two 4x4 layers of perfect hydrate units cut out from a larger hydrate crystal and allowed rotational motion only. This setup created two outer layers of partially destroyed and originally empty cages. Liquid layers directly facing the hydrate included a thin aqueous layer and a larger water phase containing either a freely-floating calcite or hematite fragment or a fixed calcite slab. Liquid carbon dioxide layer of 20 Å was inserted in between the aqueous layers. Maestro/Jaguar quantum chemistry package version 7.6 utilizing B3LYP with PS LACVP basis set was used to estimate the partial charges in vacuum and with continuum solvent model for a 210-atom calcite slab cleaved along the dominant 1-0-1 over bar-4 plane. Except for the crystal edges, vacuum charges proved to be quite uniform and were mapped onto a 1620-atom calcite slab. Calcite fragment used charges found in literature; hematite charges were obtained using an eight-atom fragment retaining hematite crystal symmetry.
All systems showed evidence of vigorous carbon dioxide transport heavily affected by the presence of minerals. Carbon dioxide molecules crossed both aqueous layers to fill a large number of partial hydrate cavities. This caused the neighboring free water molecules to form hydrate-like hydrogen bond structures. It was found that the presence of minerals contributed to increased admixture of carbon dioxide, with hematite fragment encouraging carbon dioxide absorption in water well in excess of its solubility limit. Combined with the highly volatile carbon dioxide – water interface displayed by both calcite and hematite systems, this appears to indicate an increased potential of hydrate formation in water-carbon-dioxide-mineral systems found in reservoirs and during pipeline transport of natural gases.