

# Impacts of Caprock Wettability and Rock-Fluid Interfacial Tension on Hydrogen and CO<sub>2</sub> Geo-Storage

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## Abstract

Underground hydrogen storage (UHS) and carbon dioxide geo-storage (CGS) are two essential methodologies employed to address the worldwide energy demand and facilitate the transition towards a low-carbon global economy. The distribution of H<sub>2</sub> and CO<sub>2</sub> at the pore scale, the storage capacity of these gases at the reservoir scale, and the security of their confinement are all greatly affected by interfacial properties. These variables include the equilibrium contact angle ( $\theta_E$ ), as well as the interfacial tensions between the solid and liquid phases ( $\gamma_{SL}$ ) and between the solid and gas phases ( $\gamma_{SG}$ ). Nevertheless, due to the technological limitations associated with empirically obtaining these parameters, they are frequently computed using advancing and receding contact angle measurements. There is a limited availability of data about  $\theta_E$ ,  $\gamma_{SL}$ , and  $\gamma_{SG}$ , especially about caprock's hydrogen structural sealing potential, which has not been documented in existing literature. In this study, the authors integrated Young's equation and Neumann's equation of state to theoretically calculate three parameters ( $\theta_E$ ,  $\gamma_{SL}$ , and  $\gamma_{SG}$ ) under reservoir conditions (5 – 20 MPa, 308 – 343 K, and 10 wt.% NaCl salinity) for the possible geo-storage of H<sub>2</sub> and CO<sub>2</sub>. The study aimed to examine the wetting behavior of pure mica, organic- aged mica (four organic acids are taken into this study, hexanoic, lauric, stearic, and lignoceric acid at various concentrations of 10<sup>-2</sup> – 10<sup>-9</sup> mol/L), and alumina nano-aged mica substrates (four nanofluid concentrations are taken into this study, 0.05, 0.1, 0.25, and 0.75 wt.%), with a specific focus on understanding the factors that contribute to the phenomenon of rock wetting and the possibility of these substrates acting as caprock sealants. The study's findings indicate an increase in  $\theta_E$ , while there is a drop in  $\gamma_{SG}$  as pressure, organic acid concentration, and alkyl chain length increase. However, there is a decrease in both  $\theta_E$  and  $\gamma_{SG}$  as temperature increases. The values of  $\theta_E$  and  $\gamma_{SL}$  exhibit a drop, whereas  $\gamma_{SG}$  demonstrates an increase when the concentration of alumina nanofluid increases from 0.05 wt.% to 0.25 wt.%.

In contrast, the values of  $\theta_E$  and  $\gamma_{SL}$  exhibit an upward trend, whereas  $\gamma_{SG}$  demonstrates a downward trend when the concentration of alumina nanofluid increases from 0.25 wt.% to 0.75 wt.%. The wettability of mica, which serves as a proxy for caprock, exhibited a lower degree of hydrogen wettability than carbon dioxide wettability under similar thermo-physical conditions. The interfacial data presented in this work play a critical role in predicting changes in caprock wettability and the subsequent impact on the structural sealing ability of UHS and CGS systems.