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Measuring and Modelling Supercritical Gas Adsorption in Clay Minerals and Shales

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Clay minerals are ubiquitous in the subsurface: they are found in CO2 sequestration targets (e.g., sandstones) and in the seals above them, and are major constituents of unconventional shale plays considered for natural gas recovery. A significant fraction of the porosity in clay-rich systems is occupied by micro- and meso-pores that provide a large surface area for physical and chemical interactions with the surrounding fluids. Of particular interest to this study is the adsorption behaviour of CO2 and CH4 that leads to the trapping of these gases in the porous structure at liquid-like densities. From a practical perspective, gas adsorption can lead to (i) an increase of storage capacity in reservoirs having larger clay contents, (ii) an advance in storage safety by limiting gas diffusion through cap rock seals, and (iii) an enhancement of gas production from tight shale formations through an adsorption/desorption (CO2/CH4) process.

Supercritical gas adsorption studies on clay and shale samples that address these aspects are found in the literature, but the picture is still far from being complete. The main reason for this is the intrinsic difficulty in performing these experiments at subsurface conditions (high-pressure and temperature), and in their description, because the interactions between the gases and the rock's constituents (clays, carbonate minerals and organic matter) are quite complex. The lack of a systematic evaluation on the effects of temperature, pore structure and pore chemistry on gas adsorption over the relevant pressure range precludes the development and validation of theoretical models for gas adsorption in rocks that have a sufficient degree of predictive ability. The latter is a necessity if laboratory observations that are inevitably limited in their probed rock volume are to be used to make useful estimates of process-relevant parameters, such as Gas-In-Place and storage capacity.

We report results from a systematic experimental investigation on the adsorption properties of CO2 and CH4 over a wide range of conditions (0-25 MPa and 40-80℃). The systems considered include pure clay minerals (e.g., Na-montmorillonite), shale samples from various (potential) plays (Eagle Ford, Utica and Bowland Shale), as well as reference materials with well-defined surface chemistry and pore structure (micro- and meso-porous zeolites, carbons and silica). Data are interpreted using appropriate quantitative measures, such as the excess adsorption and Henry constants. The measured adsorption isotherms are described using a Lattice Density Functional Theory (LDFT) model that uses as input parameter the pore structure of the material (measured from conventional cryogenic adsorption experiments). As such, the modelling approach is more rigorous, has predictive capability and represents a significant departure from conventional empirical approaches that use Langmuir- or BET-type of models.

References

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