Kinetics and Equilibrium of Hydrogen Solubility in Water and Brines at High Pressures

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The solubility of hydrogen brine is of importance in processes such as Underground Hydrogen Storage (UHS) in salt caverns and Geological Carbon Storage (GCS) in deep saline aquifers in the presence of hydrogen impurity. In these processes, hydrogen-containing gas is in contact with brine under conditions of high pressure and, possibly, also high temperature. Dissolution of the gas in the aqueous phase, and also the water content of the coexisting gas phase, have an influence on the storage capacity and security of both salt caverns for UHS and deep saline aquifers for GCS. Until recently, no data were available for the solubility of hydrogen in brines under those conditions and, in this work, we have made measurements of pure hydrogen solubility in water and sodium-chloride brine (2.5 mol NaCl per kg of water) at temperatures up to 423 K and pressures up to about 40 MPa. The measurements were made using a static synthetic apparatus with both visual observation and quantitative pressure-volume data used to determine the bubble points. A simple thermodynamic model, based on the Krichevsky-Kasarnovsky equation with inclusion of a Sechenov coefficient, has been used to rationalize the data. The model is also compared with other recently-published data. We discuss the water content of the coexisting gas phase form a modelling perspective and compare with the limited data available in the literature. A key part of the experimental approach was careful monitoring of the approach to equilibrium and we present an analysis of this in terms of interfacial mass transfer, mixing in the liquid phase and the diffusive boundary layer. The analysis shows that the rate at which equilibrium is approached slows down drastically as the bubble point is reached but remains finite.