

Effect of HCO_3^- , CO_3^{2-} and SO_4^{2-} ions on the zeta potential of intact carbonate rock sample

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Zeta potential is an important interfacial property that controls electrostatic interactions between mineral, water, and non-aqueous phase fluids. These interactions play an important role in defining the wetting state of reservoir rocks and transport of ionic species through porous media. The zeta potential is shown to be an efficient means for a broad range of applications including monitoring of single- and multi-phase flows in subsurface settings, characterization of fracture networks, efficiency of CO_2 sequestration, hydrogen underground storage and enhanced oil recovery. It is widely agreed that the zeta potential in carbonate rocks is controlled by the concentration of potential determining ions (PDI), but the understanding of the underlying mechanisms is still limited as there are little experimental data on quantitative characterization of the dependence of the zeta potential on concentration of negative potential determining ions (PDI) such as SO_4^{2-} , CO_3^{2-} , HCO_3^- , especially when their concentration is high and exceeds that of the positive PDIs.

In this study, the streaming potential method was used to measure the zeta potential of natural carbonate rock samples in contact with natural aqueous solutions of three constant ionic strengths while varying concentration of sulphate (SO_4^{2-}) and carbon (C_4) related (HCO_3^- , CO_3^{2-}) ions

We employed the surface complexation model (SCM) of calcite to explain the results and demonstrated that although previously published predictive SCM was successful in reproducing dependence of the zeta potential on varying concentration of HCO_3^- and CO_3^{2-} , it significantly overestimated the impact of SO_4^{2-} . Our results demonstrate the variation of the negative zeta potential with \log_{10} of SO_4^{2-} concentration is non-linear suggesting varying mechanisms of this PDI's specific adsorption. Moreover, the results demonstrate that the zeta potential strongly depends on the total ionic strength, interpreted from slopes of the linear regressions for each negative PDI in different background solution. This observation suggests that equilibrium constants of negative PDI specific adsorption may be affected by the total ionic strength. Our findings improve the current understanding of the complex physicochemical processes that take place at calcite-water interface and provide important insights into electro-geochemical controls of wettability essential for underground storage of CO_2/H_2 in carbonate rocks.