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pH controlled modification of metal oxide surfaces in mine tailings measured by low field ^{1}H NMR relaxometry

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Abstract

Porous media are prevalent in both natural formations and engineered systems, playing a key role in processes such as mineral carbonation and groundwater flow. However, understanding the physiochemical interactions within their complex pore networks remains challenging. Nuclear Magnetic Resonance (NMR) techniques, particularly, transverse relaxation time (T_2) measurements offer a powerful, non-invasive means of inferring pore-scale behaviour [1]. These measurements are highly sensitive to factors such as surface chemistry, mineral deposition and magnetic susceptibility. In systems containing iron oxides, these effects are even more pronounced due to the paramagnetic nature of Fe^{3+}. Additionally, surface charge, and thus surface relaxivity is influenced by pH, electrolyte concentration, and the isoelectric point (IEP) of the mineral surface. This study investigates how NMR T_2 relaxation of hematite-containing porous media responds to changes in pH and electrolyte concentrations, using a low-field rock core analyser. A stepwise increase in NMR T_2 relaxation time was observed when mine tailings were mixed with $MgCl_2$, attributed to acidification and the presence of sufficient divalent cations. The results imply that selective adsorption of divalent cations can shift the IEP of mineral surfaces to higher pH values [2], leading to significant shifts in NMR T_2 relaxation. To isolate these effects, a model system of borosilicate glass beads and hematite was used, where a threefold increase in T_2 was observed regardless of whether the porous media was exposed to acidic or basic solutions. These findings suggest that under acidic conditions, the formation of amorphous hydroxyl groups reduces surface relaxivity, while under basic conditions, FeOOH formation increases surface relaxivity [3]. This study highlights the importance of identifying IEP in systems with sufficient magnetic susceptibility and emphasizes the need for continuous pH monitoring for accurate interpretation of NMR results.

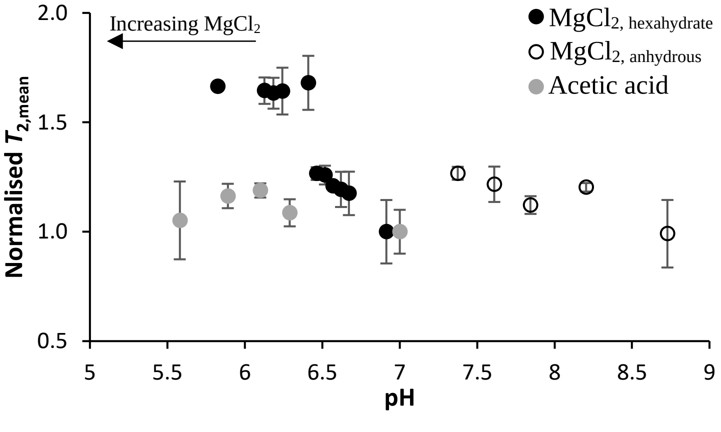

Fig. 1: Normalised $T_{2,\mathrm{mean}}$ as a function of pH for tailings mixed with $MgCl_{2,\mathrm{hexahydate}}$, $MgCl_{2,\mathrm{anhydrous}}$ and acetic acid solutions. Concentration of $MgCl_2$ ranges from 0.5M to 2.5M.

References: [1] Robinson et al., Phys. Rev. Lett. (2023). [2] Kosmulski, Adv. Colloid Interface Sci. (2016). [3] Parks, Chem. Rev. (1965).