



## **Anomalous zeta potential trends in natural clay-rich sandstone**

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It is widely accepted that zeta potential in natural sandstones is negative and increases in magnitude with increasing ionic strength. However, the impact of clay minerals remains poorly understood. Here we report measurements of zeta potential, obtained using the streaming potential method, in intact, clay-rich Berea sandstone saturated with NaCl and CaCl<sub>2</sub> electrolytes of varying ionic strength.

We found that diluting NaCl electrolyte the (negative) zeta potential increased in magnitude (became more negative). The observed relationship between zeta potential and ionic strength is consistent with numerous previous experimental and modelling studies. However, with further dilution, the zeta potential also decreased in magnitude (became less negative). The initial increase in magnitude of the zeta potential with dilution is consistent with expansion of the electrical double layer, but the decrease in magnitude with further dilution has not been observed previously and is not consistent with double layer expansion. Diluting CaCl<sub>2</sub> electrolyte yielded no change in zeta potential irrespective of the ionic strength. The constant value of zeta potential has not been observed previously and is not consistent with double layer expansion.

Our results show that the presence of clay minerals can significantly modify relationships between zeta potential and ionic strength in simple monovalent and divalent electrolytes. The zeta potential behaviour is more complex than has been observed and modelled in clay-poor and clay-free sandstones.