Examining the role of aqueous organo-silica complexes on silicification during carbonate diagenesis

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Silicification is a diagenetic process that commonly occurs in carbonate rocks. During silicification, silica-rich solutions deposit silica minerals (e.g., quartz, chalcedony) that replace carbonate minerals (e.g., calcite) in the rock [1]. The presence of silica in carbonate rocks negatively impacts hydrocarbon exploitation because silica blocks up porosity and permeability, which hinders the migration of hydrocarbons into and reservoir development within them. However, the fundamental geochemical mechanisms that drive silicification of carbonate rocks during diagenesis remain poorly understood. An existing model has proposed that carbonate silicification proceeds through an interface-coupled dissolution-precipitation reaction, wherein the force created by silica growth in confined pore spaces leads to dissolution of carbonate at the silica-carbonate interface [2]. However, it lacks a mechanism that enables pore fluids to become significantly supersaturated with respect to silica to allow nucleation and growth. Field and experimental observations have demonstrated that dissolved organic carbon (DOC) can significantly enhance silica solubility, such that quartz rapidly dissolves into DOC-rich solutions far above even the thermodynamic solubility of amorphous silica [3]. We hypothesize that enhanced silica solubility caused by geologically ephemeral organo-silica complexes creates silica-supersaturated pore fluids, which, in turn, drives carbonate silicification as the organo-silica complexes become unstable as environmental conditions evolve. To examine this hypothesis, we present a suite of batch experiments dedicated to: (i) examining the nature and stability of aqueous organo-silica complexes at variable conditions using FT-ICR-MS and ICP-OES analysis; and (ii) evaluate the physical and chemical characteristics of carbonate replacement by silica during interaction with Si-DOC rich solutions using SEM-EDS, ICP-OES, and FT-ICR-MS analysis. The results of this study will directly impact our understanding of carbonate silicification reactions.