Can CO_2 be sourced by mineral reactions in deep sediments?

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CO_2, a frequent constituent in natural gas fields, can come from various sources, either organic or inorganic. A better knowledge of geochemical systems and conditions able to produce large amounts of CO_2 would help to address several issues in the context of petroleum exploration.

Among the potential CO_2 sources, the present work deals with mineral reactions likely to occur in conditions of low-grade metamorphism, reached in the deep part of some sedimentary basins. The role of overmature kerogens was also investigated. The adopted methodology combined thermodynamic modelling and experimental work.

Numerical calculations were achieved using Arxim-GEM, a code able to solve thermodynamic equilibrium through free energy minimization. The associated database is consistent with Berman's one [1] enriched with properties of numerous pure phases and solid solutions –including clay minerals. The fluid phases are described by the CPA-Electrolyte equation of state parameterized for H_2O-CO_2-CH_4-NaCl systems [2]. The thermodynamic properties of kerogens were calculated using a group contribution approach [3]. CO_2 formation in Si-Al-Na-K-Ca-Mg-C-H-O systems was systematically explored in a range of temperature and pressure encompassing 100-500 °C and 300-2,500 bar. A set of CO_2-prone mineral reactions was identified, and shows the respective role played by carbonates, alumino-silicates and residual kerogen.

In order to validate the thermodynamic calculations, gold tubes experiments were conducted between 350 and 475 °C at 2,500 bar with a mixture of dolomite, kaolinite, quartz and water during 100 days. Despite some discrepancies between modelling and experimental results, attributed to the formation of hydrated smectites, an overall agreement was obtained and key-reactions were observed: kaolinite disappearance, devolatilization of dolomite to form CO_2 and calcite, and precipitation of micas and chlorites.

[1] Berman (1988) *J. Petrol.* **29**, 445-522. [2] Courtial *et al.* (2014) *Geochim. Cosmochim. Acta* **142**, 1-14. [3] Richard & Helgeson (1998) *Geochim. Cosmochim. Acta* **62**, 3591-3636.