

Formation of reduced carbon compounds using natural catalysts in hydrothermal experiments

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The alteration of peridotites (the so called serpentinization reactions), leads to the production of H₂ and thus propagates low oxygen fugacity conditions in the fluids which may in turn favor the reduction of CO₂. This reduction reaction is invoked for the production of methane and other reduced carbon molecules (e.g. carboxylic acids, formic acid, methanol) observed in hydrothermal fluids circulating through the oceanic lithosphere. These reactions are usually interpreted in terms of Fischer-Tropsch type (FTT) processes and have been experimentally studied for natural systems leading to contradictory results. In this study, we carried on an experimental work to study the catalytic properties of some spinel-structure phases (magnetite and chromite) and sulfides (Fe, Cu, Zn-sulfides) which are suspected to play some role in these FTT processes. Hydrothermal batch experiments were performed in gold capsules on nano-fayalite or nano-Fe-rich forsterite powder mixed with a potential catalyst at 200 bar, 200°C during 3 weeks. The source of carbon was a NaHCO₃ solution, labelled in ¹³C in order to detect any contamination. Blank experiment without any additional source of C was also performed. After the experiment, alkanes (C1-C4) and H₂ were measured in the gas phase for all capsules by gas chromatography; this shows however a very small rate of conversion of inorganic carbon to alkanes (<1%) for any catalyst involved. Thermodynamic modelling revealed that all experiments lie on a CO₂-disordered-C phase equilibrium, suggesting that some reduced carbon could have been formed as a solid product. We indeed evidenced such C-rich solid phases by electron microscopy. Their characterization by scanning transmission X-ray microscopy (STXM) showed the presence of aromatic groups, together with aliphatic and carboxylic groups, indicating a possible role of this carbonaceous material as intermediate in the synthesis of organic molecules in the CO₂-H₂ system. Organic contamination will be further tested using nanoSims on these condensed carbon phases in ¹³C-labelled experiments.