Goldschmidt 2023 Abstract
https://doi.org/10.7185/gold2023.14810

Origins of deep abiotic hydrocarbons: Insights from fundamental redox reactions within the Fe-C-H-O system

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The deep C-H-O cycle with inputs from the surface and space controls the physical and chemical properties of the Earth system and the evolution of its habitability. Efforts to identify habitable planets must therefore go beyond surface and atmospheres to planetary interiors. However, carbon speciation and notably the diversity of abiotic organic compounds prevailing in the deep Earth is only beginning to be explored. Here, we investigate C-H-O species under deep Earth conditions of temperature (T), pressure (P), and oxygen fugacity (fO_2) by studying fundamental redox reactions within the Fe-C-H-O system. Thermodynamic calculations and *in situ* diamond anvil cell (DAC) experiments up to 10 GPa and 850 °C show that the reducing power of siderite ($FeCO_3$) in water generates aqueous fluids enriched in hydrocarbons such as CH_4, C_2H_6, C_6H_6, as well as solid graphitic carbon, and magnetite. The boundary of the hydrocarbon generation reaction has been precisely delineated in the P-T space. Additionally, fO_2-controlled parallel experiments show that lower fO_2 conditions (i.e., WM-buffered) can significantly reduce the temperature by ~200 °C at which hydrocarbon generation occurs. And hydrocarbon-rich fluids immiscible with water are observed *in situ* at high pressures but low temperatures (< 500 °C), implying that migration and enrichment of hydrocarbons are more effective in a cold subduction zone. Some of the DAC experiments were repeated using large-volume press technology to provide large-size samples for further characterization. GC-MS analysis confirms that C_1-C_4 hydrocarbons are successfully synthesized up to 4.5 GPa and 1000 °C. A Raman band obtained above 1600 cm^{-1} indicates the presence of graphitic carbon containing C-H(-O). These experiments (i) systematically provide the basic chemical principles for synthesizing diverse organics under thermobaric conditions related to subduction zones, (ii) demonstrate the variable-valence-metal- bearing carbonate has the potential to separate H-O in water by auto-redox process and store them in hydrocarbon-bearing fluids and iron oxide minerals respectively; the former may become the basic nutrient for the evolution of life when they migrate to the shallow crust, and the latter will fix O_2 in the deep Earth and contribute to the redox-inhomogeneous mantle.