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Hydrothermal Carbonation and Potential for Molecular Hydrogen Generation in High-Titanium Basalts from the Serra Geral Group, Paraná Large Igneous Province, Brazil

TIAGO DE ABREU SIQUEIRA, PHD[1], VICTOR KOVASKI CESCANI[1], THIERRY MENDES DOS SANTOS[1], MAURÍCIO GOMES[1], RENAN GUILHERME MARIN[1], MORGANA BAZZAN DESSUY[1], ANDREA JELINEK[1], ANA MARIA MIZISAKI[1], MARCO THOALDO ROMEIRO[2] AND ROMMULO VIEIRA CONCEIÇÃO[1]

[1]Universidade Federal do Rio Grande do Sul
[2]PETROBRAS

The interaction between CO_2-rich fluids and basaltic rocks plays a crucial role in geological carbon sequestration, promoting carbonate mineral formation while potentially generating molecular hydrogen (H_2) through water-rock reactions. This study investigates the thermodynamic and geochemical conditions favoring carbonate precipitation and H_2 production simultaneously using high-titanium basalts from the Paraná Large Igneous Province, Brazil, under controlled hydrothermal batch experiments.

Experiments were conducted at 150°C and pressures of 70-80 bar using 10 g of solid basalt in 50 g of high-ionic-strength saline solution simulating Brazilian seawater, with agitation at 100 rpm. Reaction times of 5, 10, and 20 days were completed, with additional experiments ongoing for longer durations. Numerical modeling using PHREEQC was applied to compare experimental results with theoretical predictions, evaluating pH evolution, alkalinity, and carbonate saturation states.

Preliminary results indicate a progressive increase in pH and alkalinity, suggesting silicate mineral dissolution and subsequent carbonate supersaturation. Dissolved Ca, Mg, and K concentrations increased over time, supporting the breakdown of primary basaltic minerals. The concentration of dissolved $CaCO_3$ arose from an initial 2,255 mg/L to 3,964 mg/L after 10 days, indicating an increasing potential for carbonate precipitation. Concurrently, mineral dissolution reactions are expected to promote redox conditions conducive to H_2 generation via Fe^{2+} oxidation.

The data suggest that optimal conditions for simultaneous CO_2 mineralization and H_2 production occur at temperatures around 150°C and pressures near 120 bar, where silicate mineral dissolution releases divalent cations, leading to carbonate supersaturation. Simultaneously, H_2 production is driven by Fe(II)-bearing mineral oxidation and water reduction reactions, particularly under more reducing conditions. At lower temperatures (<100°C), carbonation kinetics are slower, while at higher temperatures (>150°C), transient undersaturation conditions may delay carbonate stability.

Further investigations at longer reaction times will clarify the kinetics of carbonate precipitation and H_2 evolution, offering insights into the viability of Paraná basalts for combined CCUS and hydrogen generation strategies.