Microdroplets initiate organicinorganic interactions and mass transfer in thermal hydrous geosystems

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Organic-inorganic interactions regulate the dynamics of hydrocarbons, water, minerals, CO2, and H2 in thermal rocks, yet their initiation remains debated. To address this, we conducted isotope-tagged and in-situ visual thermal experiments. Isotopetagged studies revealed extensive H/O transfers in hydrous n-C₂₀H₄₂-H₂O-feldspar systems. Visual experiments observed water microdroplets forming at 150-165 °C in oil phases near the water-oil interface without surfactants, persisting until complete miscibility above 350 °C. Electron paramagnetic resonance (EPR) detected hydroxyl free radicals concurrent with microdroplet formation. Here we propose a two-fold mechanism: water-derived and n-C₂₀H₄₂-derived free radicals drive interactions with organic species, while water-derived and mineral-derived ions trigger mineral interactions. These processes, facilitated by microdroplets and bulk water, blur boundaries between organic and inorganic species, enabling extensive interactions and mass transfer. Our findings redefine microscopic interplays between organic and inorganic components, offering insights into diagenetic and hydrousmetamorphic processes, and mass transfer cycles in deep basins and subduction zones.