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Iron oxides and iron sulfides catalyse organic carbon geopolymerisation under marine sediment conditions

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The transformation of simple organic molecules into complex geopolymers via Maillard-type reactions represents an emerging mechanism for long-term organic carbon preservation in marine sediments [1]. Previous work demonstrates that ferrihydrite abiotically catalyses geopolymer formation under oxic conditions [1]. Yet most continental margin sediments are dominated by suboxic and anoxic zones beneath a thin oxic layer, and it remains unknown whether iron-catalysed geopolymerisation persists with more crystalline iron oxides, mixed-valent iron oxides, and iron sulfides under corresponding redox conditions.

We use laboratory incubation experiments at marine sediment temperatures combined with molecular-scale characterisation to determine geopolymerisation rates and mechanisms for representative iron minerals spanning sedimentary redox conditions: ferrihydrite and goethite (oxic), magnetite (suboxic) and pyrite (anoxic). Geopolymer formation in mineral-free controls is negligible, whereas all iron minerals enhance production by two to three orders of magnitude. Ferrihydrite exhibits the highest production rates, followed by goethite, magnetite and pyrite. Despite lower reactivity under anoxic conditions, magnetite and pyrite sustain substantial catalytic enhancement relative to controls, demonstrating that iron-catalysed geopolymerisation is not restricted to ferrihydrite or to oxic sediments.

Spectroscopic analyses reveal consistent formation of aromatic, nitrogen-substituted, carbonyl- and carboxyl-rich macromolecules across all iron mineral systems, indicating a common transformation pathway independent of mineral type and redox state. Catalytic efficiency scales with mineral surface area, Lewis acidity and redox capacity, consistent with coupled adsorption, surface redox activation and iron(II)-mediated cation bridging. These results extend iron-catalysed geopolymerisation beyond ferrihydrite and beyond oxic surface sediments, suggesting that this mechanism may contribute substantially more to sedimentary organic carbon stabilisation than currently recognised.

[1] Moore O.W., Curti L., Woulds C., Bradley J.A., Babakhani P., Mills B.J.W., Homoky W.B., Xiao K-Q., Bray A.W., Fisher B.J., Kazemian M., Kaulich B., Dale A.W., Peacock C.L. 2023. Long-term organic carbon preservation enhanced by iron and manganese. *Nature* 621, 312-317.