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Thermochemical oxidation of hydrocarbons in deep-buried clastic strata: Features and mechanism constrained by differential distribution of oil, gas and water

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Oxidation of hydrocarbon plays an important role in fluid-rock interactions, hydrocarbon generation and evolution in the Earth's crust. To unravel its geological features and mechanism, we conducted a comparative study in the Lower Triassic red beds in the Mahu Sag (Junggar Basin, NW China). The strata contain a variable oil, gas and water content as well as abundant high-valence Fe/Mn oxide. In the west slope area containing oil and gas, but no free pore-water, three generations of authigenic calcite occur: (i) non-luminescent, early diagenetic calcite (MnO < 0.3%, $\delta^{13}C_{VPDB}$: –5.6 to –4.1‰, $T\Delta_{47}$=40°C); (ii) bright-orange luminescent late-stage I calcite (0.75–5.23% MnO, $\delta^{13}C$: –51.4 to –25.8‰, $T\Delta_{47}$=81–107°C), and (iii) dull-orange late-stage II calcite (4.10–12.93% MnO, $\delta^{13}C$: –91.4 to –30.9‰, $T\Delta_{47}$=107–132°C). The $\delta^{13}C$ values as low as –91‰ indicate that late-stage II calcite is mainly derived from the thermochemical oxidation of methane ($\delta^{13}C$: –46.8 to –39.3‰) induced by high-valence Mn/Fe oxide, whereas late-stage I calcite is the final product of both oxidation of methane and C_{2+} hydrocarbon. For late-stage I calcite, hydrocarbon oxidation was most likely promoted by high temperature. The higher precipitation temperature of late-stage II calcite demonstrates that the oxidation of methane requires higher activation energies than C_{2+} hydrocarbons.

In the east slope area, the strata contain free pore-water and oil/gas. Hydrocarbon oxidation also differentially occurred in oil layer, oil-water transition zone (OWTZ), and water layer. In the oil layer, the oxidation of hydrocarbons including methane generates ^{13}C-depleted CO_2, causing the dissolution of alkaline minerals like K-feldspar, accompanied by the generation of solid bitumen, ferrochlorite, and extremely ^{13}C-depleted calcite (4–16% MnO, $\delta^{13}C$<–50‰). In OWTZ, the presence of water promotes hydrocarbon oxidation, generates amounts of organic acids and CO_2, significantly promoting the extensive dissolution of K-feldspar, forming abundant secondary porosity, ferrichlorite, and ^{13}C-depleted calcite (2–3.5% MnO, $\delta^{13}C$: –50 to –35‰). In the water layer, the organic acids and CO_2 produced in OWTZ diffuse downward, the dissolution of alkaline minerals decreases, and precipitating less calcite (<1.5% MnO, $\delta^{13}C$:–35 to –20‰).