

Natural gas geochemical responses to thermochemical oxidation of hydrocarbon in deep siliciclastic reservoirs of Junggar Basin, China

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The occurrence of high-valence Mn/Fe oxides in deeply buried siliciclastic reservoir rocks promotes thermochemical oxidation of hydrocarbons (TOH). This study aimed to better understand this process and its effects on natural gas geochemistry via sampling gases from the Lower Triassic red beds with depths of >3.5 km in the Junggar Basin, NW China. The gas compositions, carbon and hydrogen isotopes of C₁-C₄ alkanes, and carbon isotopes of carbon dioxide were analyzed. The gases are almost mature thermogenic gases generated by sapropelic kerogen from the deeper Lower Permian source rock. The carbon isotopic compositions ($\delta^{13}\text{C}$) of C₁-C₄ alkanes mostly follow the normal trend of $\delta^{13}\text{C}_{\text{CH}_4} < \delta^{13}\text{C}_{\text{C}_2\text{H}_6} < \delta^{13}\text{C}_{\text{C}_3\text{H}_8} < \delta^{13}\text{C}_{\text{C}_4\text{H}_{10}}$, whereas the hydrocarbon gases display a unique reversed trend of $\delta^2\text{H}_{\text{CH}_4} > \delta^2\text{H}_{\text{C}_2\text{H}_6} < \delta^2\text{H}_{\text{C}_3\text{H}_8} > \delta^2\text{H}_{\text{n-C}_4\text{H}_{10}}$ (Fig. 1). The average $\delta^{13}\text{C}$ values of C₁-C₄ increased by 10.0‰, 6.0‰, 4.4‰, and 5.1‰, respectively, compared with the initial gases from source rocks, and exhibit a gradually declining trend. In contrast, the $\delta^2\text{H}$ values of C₁-C₄ increased more from those of normal thermogenic gas, and the size of the ²H-enrichments varies, eventually resulting in the unique reversals. The $\delta^{13}\text{C}$ and $\delta^2\text{H}$ increments of methane are much higher than those of C₂-C₄. The $\delta^2\text{H}$ reversals cannot be explained by current models for hydrocarbon gas generation, such as mixing of gases from different sources and thermal maturities. Instead, the results are consistent with a model where TOH caused a systematic ¹³C- and ²H-enrichment of C₁-C₄ after hydrocarbons charged the reservoir beds enriched in high-valence Mn/Fe oxides. It is possible that the heavy isotopic enrichment of methane was increased by the Rayleigh fractionation effects of methane through TOH and the decomposition of C₂-C₄ to methane via TOH, whereas the isotopic enrichment of C₂-C₄ was mainly caused by the Rayleigh fractionation as TOH proceeded. Involvement of formation water in the reaction intermediates of TOH is likely another factor causing the abnormally heavier hydrogen isotopic enrichment and the reversals.

Fig. 1 Carbon and hydrogen isotopic variations between the gases produced from the P₁f source rocks and the T₁b reservoirs.

