Goldschmidt 2026 Abstract
https://doi.org/10.46427/gold2026.36024

Low-temperature hydrothermal fluids drive micro-scale graphitization of reservoir pyrobitumen in sedimentary basins

HUI TIAN AND LIANQIANG ZHU

Guangzhou Institute of Geochemistry, Chinese Academy of Sciences

The conventional understanding holds that high-degree graphitization of carbonaceous materials requires metamorphic temperatures exceeding 420–500 °C (Buseck and Beyssac, 2014). Challenging this paradigm, we report that ~310 °C hydrothermal fluids convert crude oils to highly graphitized carbon with a graphitization degree of ~79%, based on integrated petrographic, morphological, and structural analyses of Ediacaran samples in the Sichuan Basin, China. Our results show that hydrothermal intrusion triggered in-situ oil cracking in paleo-oil pools, generating mobile bitumen and natural gas. Relatively confined system during oil cracking, as evidenced by the high-pressure methane inclusion hosted in hydrothermal minerals, impeded the loss of light components from bitumen, helping maintain its prolonged mobility. The prolonged mobility of bitumen may be the key factor in reducing graphitization temperature, which facilitates the reorganization and ordered stacking of aromatic layers compared to solid-state transformation. Continued heating by hydrothermal fluids ultimately converted the bitumen into highly graphitized carbon. The resulting highly graphitized carbon exhibits a (002) interplanar spacing of 0.3372 nm, a stacking height (Lc) of 10–20 nm, and an aromatic domain size (La) of 80–450 nm (Figure 1), comparable to graphitized carbon related to metamorphic temperatures of 436–508 °C. The low-temperature graphitization highlights the role of mobility on the formation of highly graphitized carbon from liquid carbonaceous materials and proposes a possible new route for graphite formation related to hydrothermal fluids in addition to direct precipitation (Luque et al., 2009). This new mechanism also defines an efficient pathway for retaining carbon in shallow crust, where hydrothermal fluids convert mobile organic carbon into inert graphitic carbon, thus augmenting the capacity and efficiency of geological carbon sink. Moreover, dehydrogenation via rapid hydrothermal graphitization may also be a potential mechanism for crustal hydrogen, supplementing Fe^{2+} oxidation and radiolysis, as evidenced by hydrogen in reservoirs.

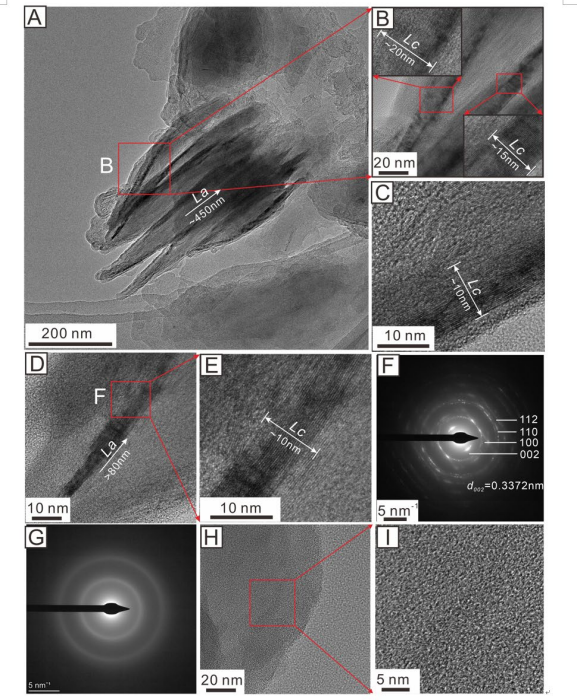

Fig. 1 Structural and chemical characteristics of pyrobitumen (PT1, 5781.4 m). (A, B) HRTEM images revealing flake-shaped lattice fringes with Lc = 15–20 nm and La ≈ 450 nm. (C-E) Well-ordered, stacked aromatic layers with Lc = 10 nm and La > 80 nm. (F) Corresponding SAED pattern showing polycrystalline diffraction rings indexed to the (002), (100), (110), and (112) planes of graphite. (G) Corresponding SAED pattern showing diffraction rings; (H, I) HRTEM images revealing disordered carbons.

Buseck, P.R., Beyssac, O., 2014. From Organic Matter to Graphite: Graphitization. Elements, 10, 421–426.

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This study is supported by NSFC (Grant No. 42330811and 4250020838).