Hydrocarbon gas generation and pore evolution of the early Palaeozoic organic rich shales in South China: Insights from closed and semi-closed hydrous pyrolysis

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The investigation of gas generation and pore evolution of organic rich shales is critical to the evaluation of shale gas. This study aims to assess the gas potential of the early Palaeozoic shale intervals in South China. A series of closed and semiclosed hydrous pyrolysis experiments were performed on two sets of shale samples from the Huadian basin and Luquan area, respectively.

Results indicate that the maximum oil expulsion efficiency of the Upper Ordovician Wufeng-Lower Silurian Longmaxi shale is 68.8%. When the value of equal vitrinite reflectance (EqVRo) reaches 3.2%, the total gas generative potential from the retained oil and kerogen is 214.1 mL/g TOC, while that from kerogen only is 69.8 mL/g TOC. Retained oil-formed gas at high mature stages (EqVRo >1.3%) accounts for 60.8% - 68.3% of the total gas, and the kerogen-formed gas occupy 31.7%-39.2%. For the Lower Cambrian Niutitang shale, the gas yields of C1-C5 alkanes obviously decrease with increasing oil expulsion efficiency, and are results of thermal cracking gas caused by a decrease of the retained oil. The change of gas drying indices indicates that EqVRo 1.8% calculated from pyrolysis temperature represents the start of a significant cracking of the heavy gases. Ratios of iC4/nC4 and iC5/nC5 exhibit a gently increasing trend, which may be resulted from the catalysis of clay minerals.

The N2/CO2 adsorption experiment and Frenkel-Halsey-Hill (FHH) model are used to characterize pore structure and fractal dimension of artificially matured shale samples before and after the Soxhlet extraction. The samples mainly develop wedge-shaped or slit-shaped pores. 0.5-0.8 nm and 30-100 nm pores are predominant groups. Retained oil hydrocarbons are able to block certain mesopores and macropores. The evolution of pore structure is more complex than that of pore surface.