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Are time and temperature the only constraints to the simulation of organic matter maturation?

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Abstract—Confined pyrolysis, hydrous pyrolysis and high pressure hydrous pyrolysis were performed with various samples (Toarcian shale and kerogen, Woodford shale and kerogen, Mahakam coal) and under different experimental conditions, in order to test the effects of pyrolysis variables on the simulation of maturation of organic matter. Results show that: (i) experimental time and temperature are not exchangeable parameters as suggested by first order kinetic law models; (ii) increasing confining pressure (300–1300 bars) in gold cell experiments performed on Woodford samples has a limited influence on the reduction rate of the oil potential, the TSOM (Total Soluble Organic Matter) yield and the structure of the polars; (iii) the presence of excess liquid water (0–100 weight%) in confined pyrolysis experiments does not significantly influence organic matter maturation. However, comparisons with classic hydrous pyrolysis (200 weight% water) could indicate major differences in timing and composition of the TSOM (bitumen + expelled fraction) as well as of the solid residue. It appears that excess water is not necessary to simulate organic matter maturation when the system is sufficiently confined. Hydrous pyrolysis conditions (low partial pressure of products) delay Woodford kerogen conversion. In addition, results concerning the Mahakam coal suggest that the structure of the organic matter plays an important role concerning the water effect; (iv) increasing water pressure in hydrous conditions (220–1300 bars) drastically lowers and delays Woodford kerogen maturation. Quantitative analysis of the water produced during confined pyrolysis and comparison of the extent of aromatization in hydrous and confined pyrolysis suggests that two competing hydrogen transfer mechanisms occur during organic matter pyrolysis. This study shows that additional parameters must be defined when comparing the results of the experimental simulation of organic matter maturation.

Key words—hydrous pyrolysis, confined pyrolysis, Toarcian shales, Woodford shale, Mahakam Delta coal, hydrogen transfer mechanisms

INTRODUCTION

The respective roles of time and temperature are recognized as critical in the processes of organic maturation and hydrocarbon generation. Pioneering experimental work performed in the beginning of the century suggested that time and temperature were two essential parameters of coalification (Bergius, 1913). More recently, laboratory experiments allowed the different coalification and hydrocarbon generation stages to be evidenced on time–temperature diagrams (Bostick, 1973). Such observations, experimentally or empirically (Connan, 1974; Wright, 1980), relate time and temperature by thermal maturity scales (Lopatin, 1971) or by mathematical models based on a first order Arrhenius law (Tissot and Espitalié, 1975). Recent developments in kinetic modelling of hydrocarbon generation (Ungerer and Pelet, 1987; Burnham and Braun, 1990) have assumed that the transformation of organic matter during thermal treatment can be depicted by a series of parallel and successive reactions involving primary and secondary cracking of kerogen and bitumen (Carlsen *et al.*, 1993). Subsequent calculations of distributed activation energies allowed the kinetic evolution of organic matter to be described. However, the use of these kinetic parameters is still questionable because there is no proof that kerogen and oil cracking follow a first order reaction. Such theoretical and experimental results are currently used in order to predict the maturity of a source rock and to reconstruct the thermal history of sedimentary basins.

Parallel to these heating experiments—generally conducted in open systems—pyrolysis devices dedicated to reproduce natural maturation of organic matter have been developed. It has been shown that the simple heating of source-rocks, coals or kerogens did not lead to satisfactory evolution of the geochemical parameters (especially concerning the release of liquid hydrocarbons). Different experimental studies demonstrated that the simulation of natural maturation may be achieved when excess water pressure or high confinement are applied to organic matter (Monthioux *et al.*, 1985; Lewan, 1979, 1993a; Horsfield *et al.*, 1989; Landais *et al.*, 1989a; Vandenbroucke *et al.*, 1993). Artificial maturation experiments provided adequate data sets for describing the different maturation stages and quantifying the oil and gas yields (Monin *et al.*, 1990). Such data cannot always be derived from a natural series of samples. Comparison of artificial and natural series allowed