Artificial maturation experiments on Qingshankou Formation shale: Porosity changes and implications for hydrocarbon expulsion behavior

Yan, Yu1,2,3; Misch, David3; Wang, Min1,2; Shi, Xiangyun2; Skerbisch, Lukas3; Sachsenhofer, Reinhard3

1 School of Geosciences, China University of Petroleum, 66 West Changjiang Road, Huangdao, Qingdao 266580, P.R. China; 2 Key Laboratory of Deep Oil and Gas, China University of Petroleum, Qingdao, China; 3 Department of Applied Geosciences and Geophysics, Montanuniversität Leoben, A-8700 Leoben, Austria.

The Upper Cretaceous lacustrine Qingshankou Formation (Songliao Basin, NE China) is a promising shale oil target due to its remarkable source potential. The formation reaches total organic carbon (TOC) contents > 10 wt.-% and hydrogen index (HI) values of > 700 mg HC/g TOC in its most prolific parts, resulting in a substantial shale oil potential. Previous studies targeted the expulsion behavior of the Qingshankou Formation and reported a maximum of extractable organic matter, pore volume occluded by bitumen, as well as Rock-Eval S1 hydrocarbons and high molecular weight bitumen (solvent-extractable part of the Rock-Eval S2 peak) in place at a vitrinite reflectance (VR) of ~ 0.8 %Ro. Furthermore, a reportedly sharp decrease in these parameters at > 0.9 %Ro indicates a good expulsion efficiency of the Qingshankou Formation in comparison to other oil-prone source rocks. However, work on natural source rock maturation profiles cannot exclude lithological variations as well as possible uncertainties in maturity determination. Hence, laboratory-based maturation experiments such as hydrous pyrolysis offer a great opportunity to expand the knowledge on the organic matter transformation-related pore space evolution within a potential shale oil reservoir. This contribution presents microstructural data gained from a series of artificially matured Qingshankou Formation shale samples. Hydrous pyrolysis experiments have been conducted on an initially immature sample at 0.55 %Ro with an initial TOC of 11.0 wt.-% and an initial HI of 744 mg HC/g TOC. Six sub-specimens originating from the same sample have been pyrolyzed up to corresponding VR values of 0.8, 1.0, 1.1, 1.2, 1.3, and 1.5 %Ro and investigated via broad ion beam – scanning electron microscopy (BIB-SEM) porosimetry (detectable pore size range from 30 nm to ~2 μm) to capture porosity changes related to i) hydrocarbon generation and resulting pore occlusion, as well as ii) hydrocarbon expulsion and secondary cracking. The change in image-based porosity shows a clear trend of pore occlusion, which results in a drastic decrease of porosity from 8.9 vol.-% at 0.8 %Ro to 0.7, 1.4, and 2.5 vol.-% at 1.0, 1.1, and 1.2 %Ro, respectively. A sharp increase in porosity to 13.6 vol.-% at 1.3 %Ro suggests that the main phase of hydrocarbon expulsion occurs between 1.2 and 1.3 %Ro at the given experimental maturation settings, resulting in a drastically changed microstructure of the rock. This is remarkably late compared to the suggested main expulsion phase around 0.9 %Ro reported for the natural maturity profile. It is yet unclear if this difference is due to a limited comparability of fast artificial laboratory-based and slow natural thermal maturation (despite matching VR), or caused by lithological heterogeneities (e.g., changes in grain size or bulk mineralogy). At the onset of the dry gas window (1.5 %Ro) the image-based porosity of the pyrolyzed sample decreases to 7.2 vol.-%. This may be due to the inherent variability of the hydrous pyrolysis experiment, but may also indicate further occlusion of pores due to ongoing hydrocarbon cracking, or possibly stronger pore pressure-relaxation effects at the main stage of expulsion at 1.3 %Ro. In summary, the results of this study emphasize the dramatic pore space changes during maturation of a highly oil-prone source rock. This has important implications for shale oil extraction behavior, but also for the matrix permeability evolution of organic-rich seal rocks.