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An Optical and SEM-EDS Study of Solid Bitumens in Two Chinese Basins: Relevance to Gas Generation at Shallow and Deep Sedimentary Strata

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Solid bitumens within geological environments can be primary and secondary in origin. Primary bitumens are presumably formed when constituents of sedimentary organic matter including kerogen within a hydrocarbon source rock transform into a variably soluble, slightly semi-solid to solid bitumen during thermal maturation, some of which within the oil window are extractable with organic solvents. Secondary bitumens in contrast are derived from microbial, oxidative or thermal alteration of a pre-existing bitumen or liquid hydrocarbon. Thermally derived secondary bitumens at advanced stages of thermal maturation (pyrobitumens) are essentially formed through disproportionation reactions forming new liquids and/or gas, plus a residual pyrobitumen. Many studies have evaluated the optical properties and petrography of primary and secondary solid bitumens within hydrocarbon source rocks and carrier beds, in order to evaluate anomalous thermal events associated with ore mineralization or the thermal maturity level of the host rock with respect to oil and gas generation. In most of these cases, a marked difference in optical properties of bitumens within a given geological sample is attributed to different thermal events and/or temporal pulses of hydrocarbon migration through the host sedimentary strata. Bitumens possessing optical coke textures are generally attributed to anomalous heating events. This presentation will use two Chinese case studies to illustrate that other factors, such as bacterial (BSR) and thermochemical sulfate reductions (TSR), can also control the resultant optical properties and coke microtextures of primary and secondary bitumens. This has significant implications for biogenic and thermogenic gas generation within sedimentary strata, and thus can be used to determine the effective source rocks for biogenic gases and constrain the TSR processes associated with crude oil cracking.

In the first case study, the organic petrology of primary bitumens and other organic constituents from shallow Quaternary clastic gas reservoirs from Sanhu region in eastern Caidam

Basin has been investigated, where geochemical data of gases clearly demonstrate that the gases are of biogenic origin and were formed by breakdown of organic matter by anaerobic bacteria. The gases are dominated by methane, with minor C_{2+} hydrocarbons, N_2 and CO_2 (all below 1%). The $\delta^{13}C$ values of methane in these gases range from -68.51‰ to-65.00‰, typical of biogenic gas. The δD values of methane in these gases, ranging from-227.55‰ to -221.94‰, are much higher than those generated from acetate fermentation, indicating a probable origin from carbon dioxide reduction. Quaternary strata in the Sanhu area are characterized by inland evaporitic lacustrine sediments, with generally low organic content (on average only 0.3% TOC) and locally organic-rich carbonaceous layers (up to 40% TOC). The organic input is predominately from terrigenic herbaceous and low shrubbery plants, relatively rich in cellulose, hemicellulose, and carbohydrates such as starch, fat and pectin. As the average geothermal gradient in this area is only about 25^{o}C/km, the Quaternary sediments have been exposed to less than 70^{o}C, with the measured vitrinite reflectance values for shallow sediments with burial depth less than 2000 m being far below 0.5 %Ro. Against conventional thinking, many previous studies on the area suggest that the Quaternary deposit with a low TOC content be the effective source rocks for the bioigenic gases. The organic-rich carbonaceous layers were deliberately excluded, based on earlier mass balance calculation in the literature claiming that a 10% conversion of sediments containing greater than 0.2% TOC can potentially generate a free gas phase. Our organic petrographic results indicate that sulfate occurs widely in the Quaternary sediments in the Sanhu region. Organic matter in almost all of the organic-lean Quaternary samples (with less than 1% TOC) are either inertinite or highly mineralized bitumenite, concurrent with sulfate and ferrous oxide minerals. The miniscule amounts of organic extracts in these samples also reveal ample molecular evidence for oxidative degradation of sedimentary organic matter, such as anomalously high sterane transformation ratios, which are incompatible with the thermal maturity levels of the host sediments. This suggests that these organic-lean sediments were deposited and are still in aerobic environments in spite of significant sedimentary burial! This is in contrast with the organic-rich carbonaceous layers, where large amounts of well-preserved plant material co-occur with pyrite, a reliable indicator for anaerobic environment and sulfate-reducing bacteria. The latter are common in anaerobic environments where they aid in the degradation of organic materials. In these anaerobic environments, fermenting bacteria extract energy from large organic molecules; the resulting smaller compounds such as organic acids and alcohols are further oxidized by acetogens and methanogens and the competing sulfate-reducing bacteria. The results indicate that only these organic-rich layers have been capable of providing methanogens with sufficiently available substrates in subsequent diagenesis! When the sediments are buried to some depth and reach the optimal temperatures for methanogens to thrive, the dilution of water salinity by groundwater infiltration would greatly facilitate the flourish of the methanogens in shallow sediments and hence the generation of large quantities of biogenic methane.

In the second case study, the organic petrology of pyrobitumens from Lower Triassic Feixianguan Formation and Upper Permian Changxing Formation carbonate gas reservoirs from Puguang gas field in northeastern Sichuan Basin were investigated using incident light microscopy and Scanning Electron Microscope - Energy Dispersive Spectrometry (SEM-EDS). The optical properties (reflectance in oil), coke microtextures and relative EDS sulfur to carbon ratios of reservoir pyrobitumens from the geological environment are evaluated in terms of and compared with Upper Devonian geological analogs in Western Canada Sedimentary Basin. The degree of optical anisotropy and the type of microtextures formed within reservoir pyrobitumens is related to the chemical composition of the parent material, the reactivity of its components and the physical conditions of dolomitic carbonate rock matrix. Reflectance properties (%Romax and %Romin), coke microtextures and relative S/C ratios are used to classify the pyrobitumens. In reservoir units with low porosity where NSO-enriched, low aromaticity crudes may have been dominant, oil to gas transformations produced mainly low reflecting isotropic to fine-grained mosaic coke microtextures within the reservoir pyrobitumens. In deep reservoirs with high porosity where normal black oils were most likely dominant in the reservoir units prior to thermal cracking, pyrobitumens with high bireflectance and very anisotropic coarse flow to domain cokes dominate. Oil to gas cracking within some of the reservoir units resulted in overpressuring and is represented by highly deformed pyrobitumens with fibrous coke textures and high bireflectance. Relative concentrations of different types of pyrobitumens vary vertically and temporally, and are attributed to crude oil oxidation and sulphur enrichment during thermochemical sulfate reduction (TSR), which is contemporaneously with crude oil cracking. The interaction between crude oil and sulphur-enriched, highly alkaline fluids is suggested by the presence of high S/C, very fine-grained, late stage calcined needle cokes within brecciated pyrobitumens, as well as highly variable non-hydrocarbon contents in the gas fluids produced from these reservoirs (5.1-58.3% H_2S and 7.9-18.0% CO_2). TSR in the study area appears to have been limited by sulfate concentrations in the reservoirs, as clearly indicated by the sporadic presence of high S/C dots within a largely low S/C pyrobitumen background in most reservoir cores under investigation. Both gas geochemistry and pyrobitumen chemistry/petrography suggest that TSR reactions in the study area involve dominantly with liquid hydrocarbons due to insufficient sulfate supply, and methane is a main product, not the dominant organic reactant for TSR as suggested by previous studies.