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Formation and thermal evolution of insoluble reservoir bitumen in Angolan carbonate reservoirs

Andrew Mort¹, Fatima Laggoun-Défarge¹, Isabelle Kowalewski², Alain-Yves Huc², Jean-Noël Rouzaud³, Fabrice Muller⁴

Pyrobitumen resulting from the thermal cracking of crude oil is a frequent occurrence in petroleum reservoirs. Despite the detrimental implications of pyrobitumen for the poroperm qualities of the reservoir; little is known about the evolution of pyrobitumen properties as a function of increasing thermal stress.

A suite of pyrobitumen-bearing reservoir cores from the carbonate Jurassic Pinda formation in offshore Angola has been studied using geochemical and petrographic techniques (including elemental analysis, Rock Eval pyrolysis, GC, FTIR, XRD, SEM, TEM) in order to characterise the physical, chemical, and optical properties of the pyrobitumen as fully as possible. The 11 core samples contain pyrobitumen at reflectance values ranging from 0.55-2.24%R_o and display varying degrees of solubility in dichloromethane from 98% insoluble bitumen to 45% insoluble, indicating a substantial spread of maturity. However, these indicators of maturity, and further classical maturity indicators such as the H/C and T_{max} show surprisingly poor mutual agreement.

The reliability of maturity parameters such as vitrinite reflectance, T_{max} , H/C, has been proven by their widespread and long-standing application to coals and kerogens. However, the

discordance shown by these parameters (e.g. bitumen reflectance and bitumen solubility in DCM) indicates that thermal evolution of solid bitumens is not a straightforward process based on cracking and condensation reactions alone. Given that values of bitumen reflectance for solid reservoir bitumens



Figure 1. Reflectance and solubility data for a suite of solid reservoir bitumens from offshore Angola

with equivalent degrees of insolubility display substantial variation, it appears that additional reaction pathways are responsible for the thermally driven insolubilisation of crude oil in petroleum reservoirs.

Despite reflectance values in excess of $2.2\%R_o$, and low H/C ratios between 0,6 and 0,8, condensed polyaromatic structures remain poorly ordered, as indicated by XRD analysis of the molecular structure (maximum stack height of basic structural units (BSUs) indicates only 7-8 aromatic layers). Although the FTIR spectra show development of these condensed polyaromatic structures, the retention of large amounts of the aliphatic fraction even at high values of reflectance and low H/C is one of the most striking features of the bitumen samples. Bitumen structure was studied at multiple scales to elucidate the relative order of condensed polycyclic aromatics. TEM *a posteriori* analysis of heat treated bitumens reveals that the maximum degree of order of BSUs to form molecular orientation domains (MODs) is attained when the semi-coke precursor is oxygen-depleted and hydrogen rich. On a microscopic scale, observation in polarised light reveals varying degrees of anisotropy (fine mosaic optical microtextures) in all but the 3 most immature samples.

Development of highly reflective, optically anisotropic microtextures has previously been attributed to bitumens originating from "normal" oils rather than NSO-rich heavy oils but the geochemical evidence points to a significant influence of the NSO component in precocious insoluble bitumen formation in this series. Photonic microscopy of the bitumen *in situ* also provides corroborative evidence for a heavy precursor, and suggests collateral influences on bitumen formation such as oil-mineral interactions and the depletion of light hydrocarbons by TSR in this sample series.

The origin of insoluble bitumen is unlikely to be identifiable by a single means of analysis (e.g. petrography) but must be elucidated from a wide body of evidence. FTIR could provide the means for rapid identification of the thermal maturity of natural reservoir bitumens and the calibration of bitumen reflectance as a proxy for vitrinite reflectance but further study of larger numbers of samples is necessary to confirm the usefulness of this technique.

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