Sulfur compound transformations in hydrous pyrolysis experiments of oil source rocks

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Many studies have documented the pivotal role of early diagenetic conditions with sulfurization of organic matter for source rock potential and kinetics of oil generation [1]. In hydrous pyrolysis experiments of several organic sulfur model compounds decomposition products have been identified and reaction networks inferred [2, 3].

This study is documenting the production – and destruction or incorporation - of H_2S, small alkane thiols, different alkylated thiophenes and dibenzothiophenes in hydrous pyrolysis experiments of initially immature Posidonia shale during the production of oil and gas. To facilitate a differentiation of reactions under kinetic control far from equilibrium in contrast to those reactions approaching equilibrium, a set of isothermal hydrous pyrolysis experiments in gold capsules have been carried out at pressures of 30 MPa for times of 6 to 336 h and at different temperatures (573, 588, 603, 618 K). Time-temperature conditions were selected to cover the maturity range up into the gas window. In addition, some experiments with different time-temperature pairs resulting in identical calculated EASY-R0 maturities have been conducted.

Interestingly, there is a clear distinction of sulfur compounds being formed up to temperature dependent equilibrium concentrations (e.g. H_2S, buffered by the mineral matrix), others which are produced during initial stages of the maturation process and then either destroyed or incorporated in larger molecules (e.g. propane-1-thiol and alkylated thiophenes) and those, whose concentrations are steadily increasing (e.g. alkylated dibenzothiophenes) up to maturities in the gas window. The sulfur isotopic composition of the organically bound sulfur in the kerogen has been significantly altered. These findings have implications for the prediction of quality/sulfur contents of oils and for the interpretation of stable sulfur isotope values of organic sulfur compounds.

[1] Lewan (1998) *Nature* **391**, 164-166. [2] Katritzky et al. (1991) *Energy & Fuels* **5**, 823-834. [3] Nguyen et al. (2013) *J. Anal. Appl. Pyrol.* **103**, 307-319.