

## Effect of water on the evolution of carbon and hydrogen isotope of lignite during artificial maturation

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Generally, the thermal degradation of sedimentary organic matters occurred in complex inorganic environments. The geochemical evidences, that suggest effects of water on the generation potential of petroleum and natural gas and the isotope fractionation of natural gas, have been extensively reported by field observations and hydrous pyrolysis experiments. To address the effects of water on the thermal evolution of organic matters, a series of non-isothermal hydrous pyrolysis involving lignite ( $R_o=0.4\%$ ) with the presence of distilled water ( $\delta D = -51.1\text{‰}$ ) and sea water ( $\delta D = -6.8\text{‰}$ ) were conducted in this study.

The determination of the yields of products indicated that the presence of two water both enhanced the yields of oils and hydrocarbon gas during lignite maturation, while the hydrocarbon generation time or threshold seemed not affected by the presence of water. The generation of oil mainly occurred at the range of  $R_o=0.5-1.0\%$ . The generation of gas can last to  $R_o=5.0\%$  for lignite, the maximum yields of hydrocarbon gas during hydrous pyrolysis can reach about 300 ml/g.coal. Meanwhile, it can be also observed that the  $^{13}\text{C}$  isotope for methane and ethane was slightly depleted with the presence of two water, which is consistent with the result from our previous hydrous pyrolysis involving hydrocarbons and oils. It is notable that hydrogen isotope fractionation of hydrocarbon gas, oils and residue coal was significantly affected by the presence of water and mainly dominated by the hydrogen isotope of water. The presence of both water resulted in the enrichment of D for hydrocarbon gas. Relatively,  $\delta D$  of methane and ethane derived from hydrous pyrolysis involving sea water ( $\delta D=-6.8\text{‰}$ ) were highest compared with that from anhydrous pyrolysis and hydrous pyrolysis involving distilled water. Moreover, the hydrogen isotope of the oils and residual coal were also enriched by the presence of water. Similarly, the presence of sea water lead to most intensively enrichment of D for oils and coal. Surprisingly, the  $\delta D$  of the residual coal did not continuously increase with the maturity. When  $R_o$  is larger than 2.5%, a sudden decrease of  $\delta D$  of coal appeared in hydrous pyrolysis, which is consistent with the evolution of hydrogen isotope of geological samples.

Base on these results, we can established the isotope fractionation model for natural gas as well as the coal during thermal maturation in different sedimentary environments (terrestrial or marine). These models can be applied to predict the natural gas potential and the isotope compositions of natural gas and residue coal in a particular basin with certain sedimentary environments and thermal history.