Noble Gas Insights on Stable carbon isotope reversals in Natural Gases

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Increasing demands for cleaner burning sources of energy have dramatically increased the exploration and production of natural gas from unconventional resources, such as shales, in recent decades. Geochemical analyses of these different shale gases revealed that many gases produced from high thermal maturity shales experienced partial or full isotopic reversals, where the stable isotopes of carbon in methane display less negative values compared to ethane (partially reversed) and propane (fully reversed). In conventional petroleum systems, oil and natural gas is derived from the temperature-dependent cracking of kerogen, which fractionates the stable carbon isotopic values of the lightest hydrocarbon molecules in predictable manners ($\delta^{13}C\text{-}CH_4 < \delta^{13}C\text{-}C_2H_6 < \delta^{13}C\text{-}C_3H_8$).

The often high production experienced in shale formations with isotopic reversals has led to great interest in understanding the processes that cause these distinct geochemical characteristics. All of the present models to explain isotopic reversals, which can include mixing of gases with varying thermal maturity, microbial oxidation of hydrocarbons, redox controlled rayleigh fractionation, diffusive fractionation, and secondary cracking of heavier hydrocarbons, may effect the $\delta^{13}C$ values in similar or ambiguous manners. Therefore, additional geochemical techniques may be required to better understand these hydrocarbon systems. Noble gases represent inert tracers that are unaffected by microbial or redox processes. Their well understood abundances and production in the hydrosphere and crust make them suitable for understanding hydrocarbon evolution. The temperature-controlled release of radiogenic noble gases (^{4}He, $^{21}Ne^*$, and $^{40}Ar^*$) from mineral grains into pore fluids can constrain hydrocarbon maturity, the $^{4}He/CH_4$ ratio can be used to identify oxidation, and the atmospherically derived components (^{20}Ne, ^{36}Ar, ^{84}Kr) can be used to understand gas to water volumes and migration processes.

We present hydrocarbon molecular (C_1/C_2+) and stable isotopic compositions ($\delta^{13}C\text{-}CH_4$, $\delta^{13}C\text{-}C_2H_6$), and noble gas isotopic data from shale gases in the Appalachian Basin and the Fort Worth Basin that display isotopic reversals. Our data suggest these gases were produced from closed systems and retain greater amounts of the ASW components compared to conventionally produced gases.