

Structural Evolutions of Small Aromatic Mixtures Under Extreme Temperature Conditions: Insights from ReaxFF Molecular Dynamics Investigations

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ABSTRACT

Conversion mechanisms of carbonaceous materials during controlled pyrolysis under high temperature currently rely on empirical understanding, rather than a detailed atomistic description of the underlying chemical evolution. In this work, a series of molecular dynamics simulations, based on reactive force field, were performed to probe the structural evolutions of small aromatic (number of aromatic rings ≤ 4) mixtures under extreme conditions (temperature 3000-4000 K). For any temperature studied and independent of the presence of alkane radicals, three major gas-phase byproducts, H_2 , C_2H_2 , and C_2H_4 , were found in bulk treated by rapid heating. Temperatures as well as alkane addition can affect the production/consumption rates and fractionation of these gas species. Compared to alkane radicals, temperatures also play important roles in modulating the initial growth pathways of solid carbon clusters. At 3000 K, aromatic carbon cluster formation is mainly initiated through the direct combination of aryl radicals. Conversely, at 4000 K, carbon cluster formation is initiated by the assembly of chained radicals, since substantial polyacetylenic chains were produced. Through cross comparison on different simulation models/methods, as well as with experiments and literature works, the effect of processing conditions on carbonaceous materials conversion was revealed, and underlying mechanisms responsible for the observed phenomena are proposed.