
Fischer-Tropsch Type (FTT) reactions have been proposed(1-3) as the processes operating in the solar nebula which synthesized the organic matter in carbonaceous meteorites. Presumably these reactions occurred as a result of adsorption of H2 and CO on nebular dust grains, which catalyzed the conversion of these gases to CO2, H2O and a variety of organic species (e.g., \(3\text{CO} + 5\text{H}_2 \rightarrow 2\text{CH}_4 + \text{CO}_2 + \text{H}_2\text{O}\)) and accreted with the products onto parent bodies. The most persuasive evidence supporting such origins consisted of the direction and magnitude (40 to 100 \(^{\circ}/_{oo}\)) of the carbon isotope fractionations reported to occur between the isotopically heavy oxidized carbon (CO2) and isotopically light reduced carbon compounds (hydrocarbons) produced by the FTT synthesis(3). Similar fractionations (relative to the PDB standard) were observed between the carbonate carbon (+30 to +70 \(^{\circ}/_{oo}\)) and organic carbon (-25 to -10 \(^{\circ}/_{oo}\)) in meteorites(4,5). Typically, the organic carbon in meteorites was taken to be the high-molecular weight acid-insoluble fraction. In a few instances, the soluble organic fraction was examined and found to be contaminated with terrestrial organic compounds. Variation in the differences in isotopic compositions between the reduced and oxidized carbon reservoirs was attributed to variations in the temperature of nebular synthesis with larger differences corresponding to lower temperatures(3).

More recent studies of the organic matter of meteorites, particularly the relatively uncontaminated Murchison, have revealed wide ranges in the isotopic composition of C, N, and H which are difficult to account for on the basis of the FTT model(6-11). Nitrogen isotope fractionations in the FTT synthesis were found to be too small(12) to account for the observed variations in meteoritic organic matter(8,11). And a monotonic decrease in \(^{13}\text{C}/^{12}\text{C}\) with increasing carbon number among the low-molecular-weight Murchison hydrocarbons and carboxylic acids has been observed(10). These considerations have led us to re-examine the carbon isotopic fractionation associated with the FTT synthesis.

Static and dynamic Fischer-Tropsch Type (FTT) processes, catalyzed by three different iron-bearing phases, were studied over the 150°-500°C temperature range as models for synthesis of carbon-bearing phases in carbonaceous meteorites. For static experiments catalyst, CO and H2 were heated in sealed tubes for varying lengths of time. Gaseous products were obtained for isotopic analysis by a combined gas chromatography combustion (GCC) technique. In the dynamic mode the gases were passed through a bed of catalyst maintained at elevated temperatures, and gaseous products were analyzed by downstream sampling of the cool gas stream using GCC. Abundances and carbon isotopic compositions were measured for individual
low-molecular weight hydrocarbons, involatile carbonaceous matter, CO and CO$_2$ from these syntheses.

Products were synthesized readily from 500°C down to 250°C, below which temperature CO$_2$ was observed to form but not hydrocarbons. In all experiments in which products were formed under conditions of kinetic control, the carbon isotopic composition of products reflected kinetic isotope effects favoring incorporation of the $^{12}$C over $^{13}$C: relative to $\delta^{13}$C of the initial CO, the resulting saturated hydrocarbons and the involatile carbon deposited on the catalyst were 11-35$\permil$ lighter; and CO$_2$ was 2-13$\permil$ lighter. The unreacted CO was 2-23$\permil$ heavier, an outcome entirely consistent with the preferential transfer of $^{12}$C from reactant into products and the requirement for mass and isotope balance. No systematic carbon isotopic variation with carbon number was observed in the major hydrocarbon (saturated) products. The minor unsaturated hydrocarbons were typically 3-19$\permil$ heavier than their saturated counterpart, although always lighter than the initial CO. Thus, under kinetically controlled conditions, no large isotopic fractionations were observed between the oxidized and reduced carbon components. Only in high temperature or very long duration experiments characterized by >98% conversion of CO to product, did CO$_2$ become isotopically heavier than all other products as well as heavier than the unreacted CO. We attribute this $^{13}$C enrichment to an equilibrium isotope effect the onset of which is favored by higher rather than lower reaction temperatures.

The isotopic systematics of these FTT reactions indicate that if the large isotopic differences between oxidized (heavy) carbon and reduced (light) carbon in carbonaceous meteorites were the product of solar nebula FTT processes, an unusual scenario is called for requiring conversion of virtually all nebular CO to products at temperatures possibly as high as 500°C. Other factors such as orders of magnitude higher H$_2$/CO ratios and lower pressures in the solar nebula than those used in laboratory reactions raise doubts about the efficacy of nebular FTT processes. Even if a nebular synthesis were successful, it is questionable whether the volatile and thermolabile products initially formed survived the nebular cooling to accrete into meteorite parent bodies. As alternatives to putative nebular FTT reactions, heterogeneous gas-solid or liquid-solid syntheses in parent body environments during internal heating, volatiles mobilization and outgassing or in regolith ejecta blankets remain distinct possibilities(13).