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論文要約

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The Earth's life has emerged in the Hadean eon before 4.0 billion years ago. Serpentinite–hosted hydrothermal systems may have played a significant role for the origin and early evolution of life, because abiogenic synthesis of organic compounds can occur through the serpentinization processes. The main purpose of this study is to elucidate the mechanism of abiotic hydrocarbon production based on the field observations of present-day serpentinite-hosted systems; Hakuba Happo hot spring (Japan) and Cabeço de Vide (Portugal). First, systematic hydrogen isotopic study was conducted for the Hakuba Happo hot spring. The observed hydrogen isotopic relationship of CH_4–H_2–H_2O suggests that the methane is directly produced from H_2O, but not from H_2 in the serpentinite–hosted systems (Chapter 2). Second, the compound–specific carbon isotopic analysis was conducted for C_1 to C_5 hydrocarbons in the two on–land serpentinite–hosted systems. The carbon isotopic pattern for the Happo samples suggested that these hydrocarbons were produced by the successive addition of a single carbon compound (C1–compound). Possibly, methane is the C1–compound for the polymerization reaction. A theoretical model of the abiotic polymerization was developed to account for isotopic compositions of Happo hydrocarbons (Chapter 3). The observed isotopic trend is similar to those of the Lost City and Logatchev–II sites, thus the model is also applicable to seafloor serpentinization fields. Furthermore, a new intramolecular ^{13}C analysis of propane, was applied to hydrocarbon samples from serpentinite–hosted systems for the first time (Chapter 4). The difference of $\delta^{13}C$ values between terminal and central carbon positions of propane for serpentinite–hosted Happo sample showed a significantly smaller than thermogenic propane. The observed isotopic distribution within propane molecule is consistent with the theoretical model prediction developed in Chapter 3. Importantly, both compound-specific and intramolecular ^{13}C compositions of hydrocarbons for serpentinite–hosted sample supported the assumption that the production mechanism of these hydrocarbons was kinetically controlled with a constant isotopic fractionation. A series of findings regarding the production mechanisms for hydrocarbons in on–land serpentinite–hosted Hakuba Happo hot spring is summarized

in Chapter 5. The system of Hakuba Happo hot spring seems to be dominated by the present–day serpentinization of ultramafic rocks by meteoric ground waters at low temperature. The dissolved inorganic carbons (DIC) in infiltrated meteoric waters might convert to formate because the formate formation is thermodynamically favorable under the combination of strongly alkaline and highly reducing conditions of Hakuba Happo. Indeed, relatively high concentrations of formate (36–158 μmol/L) was observed in Hakuba Happo hot spring water. This study proposed the hypothesis that methane was abiotically produced from H_2O by potential olivine hydration through formate as an intermediate species. Subsequently, higher hydrocarbons would be formed by the polymerization of the C1–compound of methane (or potentially formate). Assuming the conceptual model of hydrocarbon synthesis for Hakuba Happo is applicable to general serpentinite–hosted systems, the differences in absolute $\delta^{13}C$ value of hydrocarbons in various geological settings can be explained as a variation of carbon reservoirs. The $\delta^{13}C$ values of hydrocarbons for seafloor serpentinite–hosted hydrothermal fields (Lost City and Logatchev–II) were approximately 20‰ higher than those of on–land Hakuba Happo hot spring. In such seafloor serpentinite–hosted systems, a series of reactions for hydrocarbon formation may be considered to start from ^{13}C–enriched carbon reservoirs such as marine DIC and magmatic CO_2. On the other hand, the lower $\delta^{13}C$ values of hydrocarbons at Hakuba Happo might be resulted from isotopically-lighter starting carbon reservoir (i.e. freshwater DIC) that mainly contains biogenic CO_2 from ^{13}C–depleted organic matter in surface. Serpentinite–hosted systems have been likely to occur more commonly in Hadean Earth than in modern Earth. Unlike the present–day serpentinite–hosted systems, komatiite that is petrologically defined as ultramafic extrusive volcanic rocks might be a host rock for serpentinite–hosted systems on Hadean Earth. In the previous experiments of prebiotic organic synthesis (ex., spark discharge of gas mixtures and Fischer–Tropsh type synthesis under the hydrothermal condition), CO_2 has been often difficult to convert to organic compounds. However, such a CO_2–problem might be solved when considering the intermediate of formate in the serpentinite–hosted systems. This study proposed the hypothesis that in the serpentinite–hosted systems, hydrocarbons were synthesized from the inorganic carbon source through the formate. This implied that an important role of serpentinite–hosted systems for birth of life might be to convert inorganic carbon (CO_2) into more reactive formate for the abiotic synthesis of organic compounds at low temperature.