

## The oldest remnant of life in 3.8 Ga old early Archaean rocks

OHTOMO, Yoko<sup>1\*</sup> ; KAKEGAWA, Takeshi<sup>2</sup>

<sup>1</sup>Kochi Institute for Core Sample Research, JAMSTEC, <sup>2</sup>Graduate School of Science, Tohoku University

The suggestion that graphite in early Archaean rocks represents materials of biogenic origin has met with a degree of scepticism. Isotopic compositions of graphite in >3.7-billion-year-old rocks from the Isua Supracrustal Belt (ISB), western Greenland, which are believed to be of sedimentary origin, suggest that vast microbial ecosystems were present in early Archaean oceans. However, results of more recent studies suggest that most of graphite-bearing rocks were formed through interactions between crustal fluids and surrounding igneous rocks during later metasomatic events, thereby casting doubt on the existence of an extensive sedimentary sequence in the ISB and on the biogenic origin of constituents. In contrast, <sup>13</sup>C-depleted graphite globules, which are considered to form from biogenic precursors, have been reported from the metamorphosed clastic sedimentary rocks in the ISB. However, these were found at a single locality. It therefore remains unclear whether traces of life at other localities in the ISB were lost during metamorphism or were originally absent. The presence of additional clastic sedimentary rocks containing graphite may provide evidence for the preservation of organic constituents in early Archaean rocks, thus supporting the notion that microbes were active in early Archaean oceans.

We conducted a geological survey along the northwestern area of the ISB. Banded iron formations contain interbedded black to grey schist layers, typically 40-80 cm thick. Rare earth element patterns in samples lie close to that in Post Archaean Australian Shale, suggesting that the protoliths of the schist was clastic marine sediments. The black-grey schist samples contain abundant reduced carbon (0.1-8.8 wt%), identified as graphite by X-ray diffraction analysis. The range of  $\delta^{13}\text{C}$  values was -23.8 to -12.5 per mil (average, -17.9 per mil), which is within the range of values reported in previous studies. Scanning transmission electron microscope and high-resolution electron microscope observations present different nanoscale morphologies between the graphite of metasediment and secondary vein samples. Examined metasediment included graphitic polygonal grains and nanotubes. Sheeted flakes were a dominant morphology of secondary graphite, whereas polygonal grains and nanotubes were absent from them, suggesting a different origin from the secondarily derived graphite.

We modelled the theoretical  $\delta^{13}\text{C}$  values of fluid-precipitated graphites. The lowest  $\delta^{13}\text{C}$  values exceed -16.4 per mil when Rayleigh-type isotope fractionation operates in the fluids. Therefore, <sup>13</sup>C-depleted biogenic organic matter in Isua clastic sediments is postulated as an initial carbon source to explain the lightest carbon isotope compositions (e.g., -23.8 per mil) in the present study. Distorted structures are common in pyrolysed and pressurized organic compounds. Such precursors commonly contain non-planar carbon ring compounds associated with abundant pores. Biogenic organic matter, which contains various molecules and functional groups, is suggested as the precursors of the graphite observed in metasediment.

In summary, the graphite in metasediment from the northwest ISB is distinct from the graphite in secondary vein samples. The combined information on geological occurrences, graphite morphologies, nanoscale structures, and isotopic compositions of the graphite in the metasediment suggests a biogenic origin of the graphite. High concentrations of <sup>13</sup>C-depleted graphite in these rocks would require widespread biological activity to support the high rate of production and sedimentary delivery of organic matter to the >3.7-billion-year-old ocean floor.

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