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Transparent carbon films containing graphene in 3.2 Ga sedimentary rocks in the Moodies group, Barberton Greenstone Belt, South Africa

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Carbon materials (e.g., fullerene, carbon nanotube, graphite, diamond) have been found in rocks throughout geologic ages [1,2]. These solid carbons have been actively investigated as products formed in specific conditions as well as important materials for carbon neutral technologies [3]. Understanding the formation conditions in ancient rocks would give insights on carbon cycle at the era and guide us to innovations for new carbon materials. Here, we report peculiar transparent carbon films containing graphene in 3.2-billion-year-old metasedimentary rocks in the Moodies Group of Barberton Greenstone Belt, South Africa.

A total of 24 iron-rich siliciclastic rocks predominantly composed of detrital materials, but also contain chemically precipitated iron and silica as well as ^{13}C-depleted organic carbon. Organic matters in the rocks are transparent unlike general Archaean sedimentary organic matters. They occur as several tens of microns in diameter films or occasionally a hundred microns of filaments with acid-resistance, containing trace amounts of sulfur and nitrogen. The transparent carbon films exhibit submicron scale-like layered or folding microstructures, in which submicron Fe, Ti and Si-euhedral minerals are attached or included. Chemical structural analysis combined with microscopic observation demonstrates that the carbon films primarily comprising aggregates of kerogeneous nanoparticles enveloped by ultrathin membranes, both of which contain graphene-like structure. Considering with possible conditions for graphene formation in tectonic history of the Moodies Group, the microscopic internal and molecular structures of the carbon films, and chemistry of the associated minerals suggest that the transparent carbon films were formed in specific degradation-recondensation processes involving organic solvents derived from microbial biomolecules during solidification of the siliciclastic sediments. Our discovery provides variety of natural solid carbon material formations associated with low temperature graphene formation at least <300°C in Archaean sedimentary rocks.

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