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Structural change of carbonaceous material inferred from Raman spectra in low- to high-temperature metamorphic rocks

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We report the structural change of carbonaceous material (CM) from the samples metamorphosed at temperatures from 165 to 655 ^{o}C based on Raman spectroscopic analysis. 10 samples were selected from contact and regional metamorphic rocks of the Kasuga, Daimonji, and Shirataki areas and 9 samples were collected from accretionary complexes of the Kitagawa, Kure, and Nobeoka areas.

The shapes of CM Raman spectra show a change from broad and complex to sharp and simple ones with increasing metamorphic temperature. The D (”Defect”)-bands dominate in the low temperature range and the G (”Graphite”)-band increases in importance with temperature increase. These changes can potentially be used to construct a CM Raman geothermometer. However, there is no study defining a methodology for decomposing the CM Raman spectrum that is applicable to low to high temperature samples, and it is difficult to discuss the sequence of the development of CM. In particular, there has been little discussion of how peak decomposition of CM Raman spectra in the low temperature range should be carried out. In this study, several band combinations for the Raman spectrum of CM metamorphosed at the low temperature range were tested and parameters for each band were compared.

These studies show that the Raman spectrum of CM can be divided into four groups by the metamorphic temperature range: low-grade CM (150-280 ^{o}C), medium-grade CM (280-400 ^{o}C), high-grade CM (400-650 ^{o}C), and ordered graphite ($>$ 650 ^{o}C).

Low-grade CM: The Raman spectrum of low-grade CM is best considered to consist of four D-bands (D1-, D2-, D3-, and D4-bands). The G-band is not clearly distinguishable in this temperature range. The shape of low-grade CM Raman spectra is qualitatively similar to those for amorphous carbon.

Medium-grade CM: The G-band first appears clearly in samples metamorphosed at temperatures of around $>$ 280 ^{o}C. The D4-band disappears at around 340 ^{o}C and the prominence of the D3-band gradually decreases with increasing temperature. The peak positions of G-, D1-, and D2-bands and intensity ratio with D1- and D2-bands show changes in the temperature range of medium-grade CM. This observation indicates that CM transforms from amorphous carbon to disordered graphite in this temperature range.

High-grade CM and ordered graphite: Most band parameters become almost constant in the Raman spectra of high-grade CM. These changes suggest that the transition of CM from amorphous carbon to disordered graphite is complete at around 400 ^{o}C. A lot of CM grains in the highest temperature sample metamorphosed at 655 ^{o}C show spectra with only a G-band present and no recognizable D1- or D2-bands. This result indicates that CM grain reaches fully-ordered graphite at temperatures around 650 ^{o}C.

The above results are an important first step in establishing a reliable CM Raman geothermometer in low to high temperature range covering the transition of CM.

Keywords: Carbonaceous material, Raman spectroscopy, geothermometer, amorphous carbon, graphite