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Thermal alteration of the R1 parameter of carbonaceous matters

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Burnham and Sweeney (1989) concluded that the vitrinite (VTN) maturation during heating could be treated most simply by considering the following four independent parallel reactions: VTN -> VTN + H_2O, VTN -> VTN + CO_2, VTN -> VTN + CH_n and VTN -> VTN + CH_4. Huang (1996) proposed the empirical power law rate model for the evolutions of VTN reflectance controlled more by temperature than time based on his heating experiments of lignite.

The thermal maturity of carbonaceous matters (CMs) may also be represented by the R1 ratio (measured commonly as D/G peak intensity ratio) of the Raman spectra. Muirhead et al. (2012) estimated the kinetic rate of the thermal maturity of CMs indicated by the R1 ratio as $R1 = B + 0.441557 * \exp(-402/T) * t\hat{}(6.04E{-}5 * T + 0.011304)$, where t is the duration of heating in seconds, T is the temperature in Kelvin and B the initial ratio of R1 (=0.23 in his study), from his flash pyrolysis experiments of CMs for 5 to 80 seconds heating. However, the CMs taken in his experiments were collected from Murchison meteorite and show quite different Raman spectra from those of CMs in non-metamorphosed pelitic rocks. Also, it should be noted that his R1 ratios may be D/G peak area ratios.

Therefore, we have made heating experiments of CMs in pelitic rocks collected from the Kanoashi Group of Jurassic complex, SW Japan and measured the R1 (D/G peak intensity) ratios before and after the experiments. Reflectances of the CMs indicate their diagenetic temperatures as around 284C. The initial R1 ratios before heating are about 0.5.

Polished rock chips were kept in an Ar purged capsule, and then taken into an electric oven. Unfortunately, the oven is not available for quick heating, it took, for example, 14 minutes to achieve 800C. The experiments have been done under 290, 500 and 800C with different durations. Note that the both 500 and 800C experiments, the same rock chips were used, that is, repeatedly heated, to follow the evolution of the R1 ratios of the individual CMs.

Small peaks between the D and G bands of the Raman spectra appeared in some CMs after short heating (< 10 minutes at 500C and (1) of 800C). However, they disappeared after longer and higher heating experiments. Other two types of the thermal evolution of the R1 ratios could be observed during 500C experiments. The one is that the R1 ratios systematically increased with increasing heating durations (i.e., repeating times), while the other is that the ratios increased after a short time heating and then decreased to the initial ratios and again increased by further heating. On the other hand, the R1 ratios after 800C heating experiments show two types of their evolutions. The one is that the ratios did not exceed 1.5 after heating, while the other did. The latter showed various R1 ratios irrespective of heating durations. Those characteristics of the thermal evolution of the R1 ratio may depend on the difference of the precursors of CMs.

Therefore, it can be concluded that the large grained CMs of which R1 ratios do not exceed 1.5 should be taken to estimate a kinetic rate for the thermal maturation of CMs. The power term on time of a preliminarily estimated kinetic rate of the thermal evolution of the R1 ratios estimated by the present experiments is one order of magnitude higher than that of Muirhead et al. (2012), implying that the rate depends not only on temperature and time but also on the initial R1 ratio. Also, it should be investigated in a future study that there may be maximum value(s) of the R1 ratios as shown by Beyssac et al. (2002), which in turn may also affect the kinetic rate.

Keywords: carbonaceous matters, R1 ratio, Raman