Iterative, randomized fitting of Raman spectra of carbonaceous material based on genetic algorithms

Lünsdorf, N.K.[1,*], Lünsdorf, J.O.[2]

[1]Sedimentology & Environmental Geology, Georg-August-University Goettingen, Germany
[2]R&D Division Energy, OFFIS Oldenburg, Germany
**e-mail: keno.luensdorf@geo.uni-goettingen.de*

Raman spectroscopy of carbonaceous material (RSCM) is a frequently used method in geosciences (e.g. Beyssac et al., 2007; Wiederkehr et al., 2011). It is fast, nondestructive and gives the maximum metamorphic temperature of the examined sample (Aoya et al., 2010; Beyssac et al., 2002a; Beyssac et al., 2002b; Lahfid et al., 2010).

The method is based on the progressive transformation of carbonaceous material into graphite during burial or regional and contact metamorphism (Aoya et al., 2010; Beyssac et al., 2002b; Lahfid et al., 2010; Wopenka & Pasteris, 1991; Yui et al., 1996). During the transformation the shape of the Raman spectrum changes also, and this change is quantified by spectral deconvolution. Common techniques are given in Beyssac et al. (2002b), Sadezky et al. (2005), and Lahfid et al. (2010). A set of functions (Voigt, Lorentzian, Gaussian, etc.) is fitted to the spectrum after baseline correction and from the parameters of the functions (i.e. center, height, area, full width at half maximum [fwhm]) different ratios are calculated to quantify the spectrum. However manual intervention is often required, especially during baseline correction. This introduces a certain degree of subjectivity and increases the time necessary to apply the method. Furthermore the data quantities are increasing drastically when spectral maps are generated or when the samples require solid statistics. Thus a need for automated evaluation of CM spectra is given.

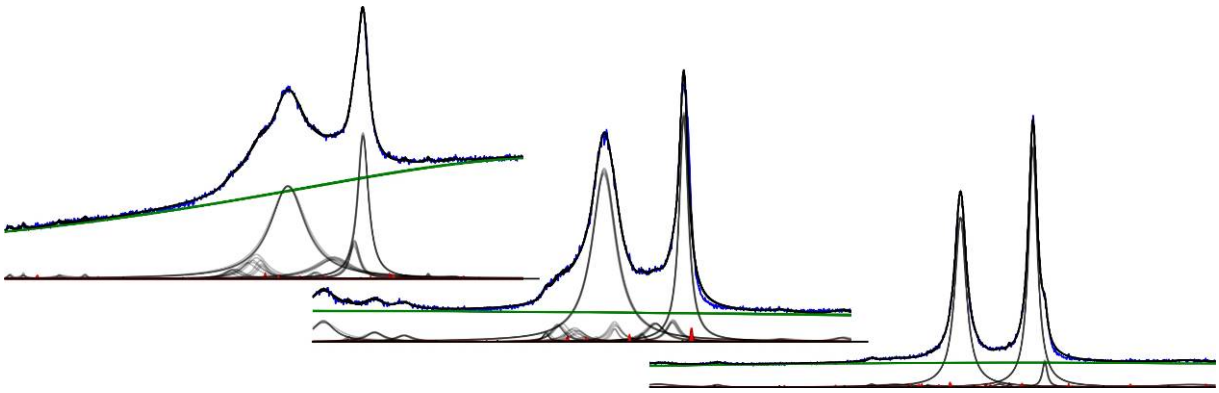

Fig. 1: *Automatic fits of CM. The structural ordering increases from upper left to lower right. Each spectrum has been fitted five times. The variability within the fits decreases with increasing structural order. Green line = Baselines, Black lines = Models, Gray lines = Model functions, Red lines = Noise funtions.*

Here we present a software package that is specifically designed to automatically evaluate CM Raman spectra. The software iteratively fits a model, which is the sum of a series of Lorentzian functions, to a given spectrum by random variation of function parameters. Because no preprocessing is applied to the spectrum, the model will consist of noise-, baseline- and signal-functions (Fig. 1). Based on the function parameters noise, baseline, and signal can be separated, and various ratios can be derived from the parameters of the isolated signal-functions. Especially in CM from sub-greenschist metamorphic conditions the Raman bands

overlap which leads to ambiguous results. Because of automation each spectrum can be evaluated several times which allows to estimate the fitting variability (Fig. 1). Additionally the subjectivity of the evaluation process is overcome by the new software. The Method and results will be illustrated by real data from geological examples.

The presented software enables the automatic and objective evaluation of CM Raman spectra which will increase the comparability of studies using the RSCM method and it will help in studies which involve evaluation of numerous Raman spectra.

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