

SUPPLEMENTARY MATERIALS

EA1 – ν_1 PEAK DOUBLING

Figure EA1 shows the ν_1 mode of Mg calcites containing 14.5 and 37.8 mol% MgCO_3 . The peak becomes asymmetric to the left as Mg increases, and increasingly difficult to fit with a single peak. This simple fact explains the features observed in Fig. 8d with a marked change of trend above 30 mol% MgCO_3 . The one- and two-peak fits are shown in Fig. EA1. For the two-peak fit, the constraint of FWHM similarity for the two peaks had to be introduced to minimize the number of potential solutions. Figure EA2a shows the peak wavenumbers as a function of Mg content. The position of the major peak is close to that determined with a single peak. The position of the second peak departs significantly from the one of the first peak, with higher data dispersion. However, the slopes of the two datasets are similar. As far as FWHM

is concerned (Fig. EA2b), the data alignment is better than in the one-peak fit and does no longer show divergence at 30 mol% MgCO_3 . As in the case of $2\nu_2$, a maximum seems to be reached at about 50 mol% MgCO_3 . However, this conclusion is difficult to ascertain as no data were collected above 50 mol% MgCO_3 . Note that for the double peak fitting, a single set of FWHM is reported on this figure as the constraint of similarity of FWHM for the two peaks was introduced in the model.

The main purpose of these supplementary materials is to explain the particular behavior of the ν_1 FWHM above ~30 mol% MgCO_3 (Fig. 8d). They indicate that the Raman spectra of the Ca-Mg carbonate solid-solution may show more complicated features than expected. The origin of peak doubling as Mg increases in the solid-solution is not yet fully understood, though a tentative explanation is provided in the main text.

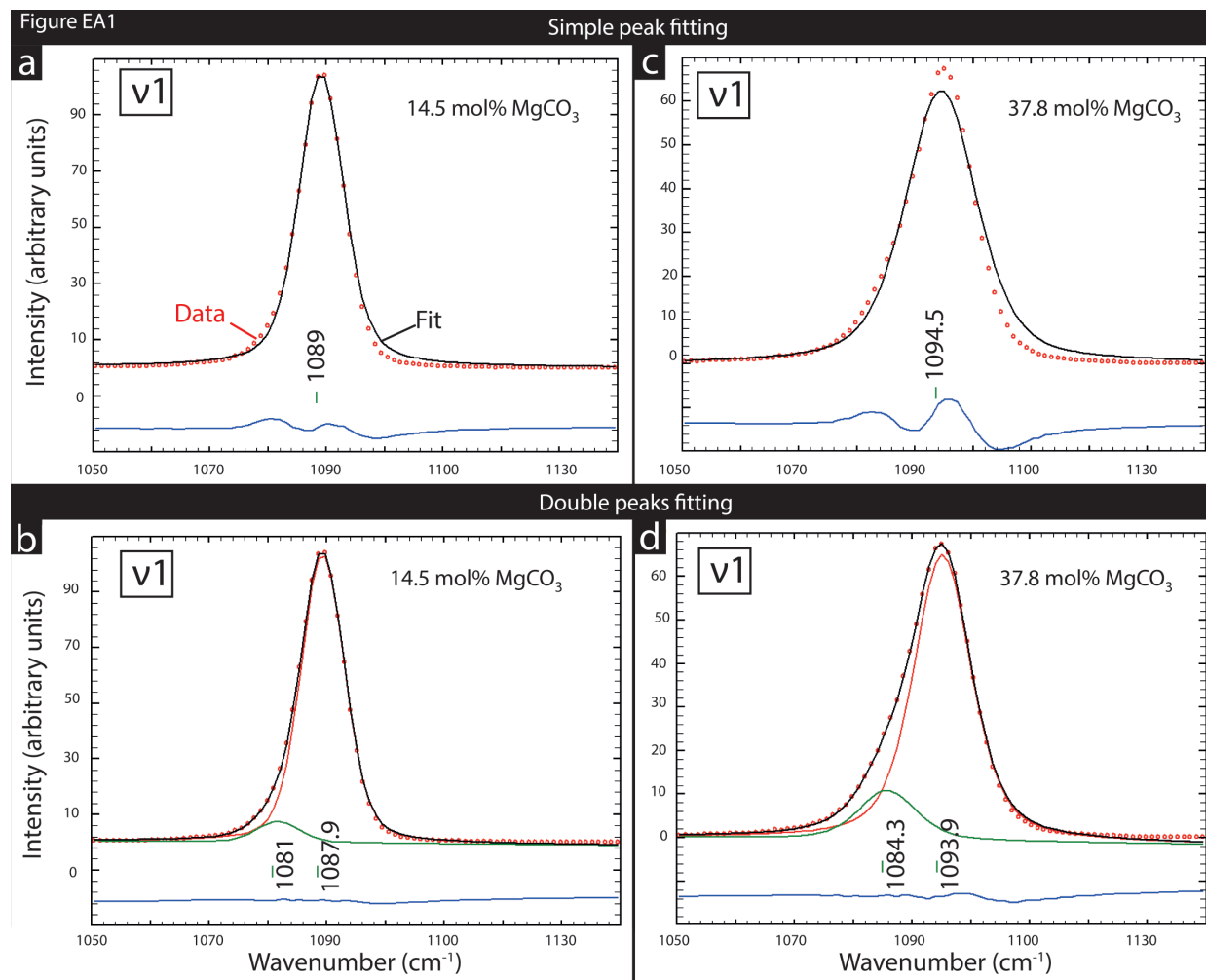


FIGURE EA-1: Simple (a, c) and double (b, d) peak fitting for Mg calcite syntheses with 14.5 and 37.8 mol% MgCO_3 .

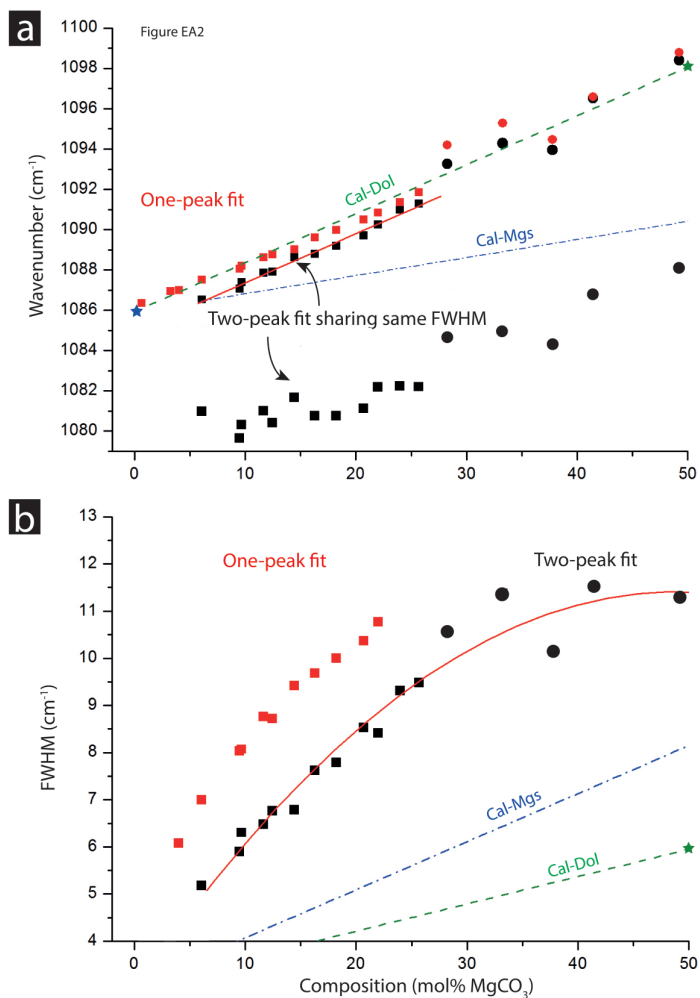


FIGURE EA-2: (a) Wavenumber of the ν_1 Raman mode of synthetic Mg calcites as a function of composition in the range 0–50 mol% MgCO_3 . Results of simple and double peak fittings are shown in red and black, respectively. Cal-Dol and Cal-Mgs lines are shown for comparison. (b) FWHM of the ν_1 Raman mode of synthetic Mg calcites as a function of composition in the range 0–30 and 0–50 mol% MgCO_3 for the simple (red) and double (black) peak fitting, respectively.