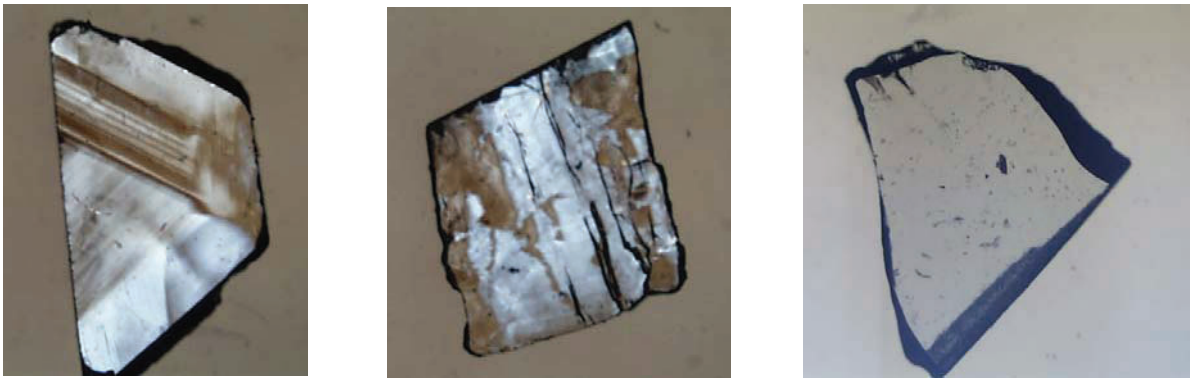


FIGURE 1. (Supplementary information) Photos, made under crossed polars, showing the optical properties of the three natural garnets. a.) Gr-44091, b.) Gr-82-112 and c.) Gr-L/K. The platelets range in size from about 2 to 3 mm in cross section.

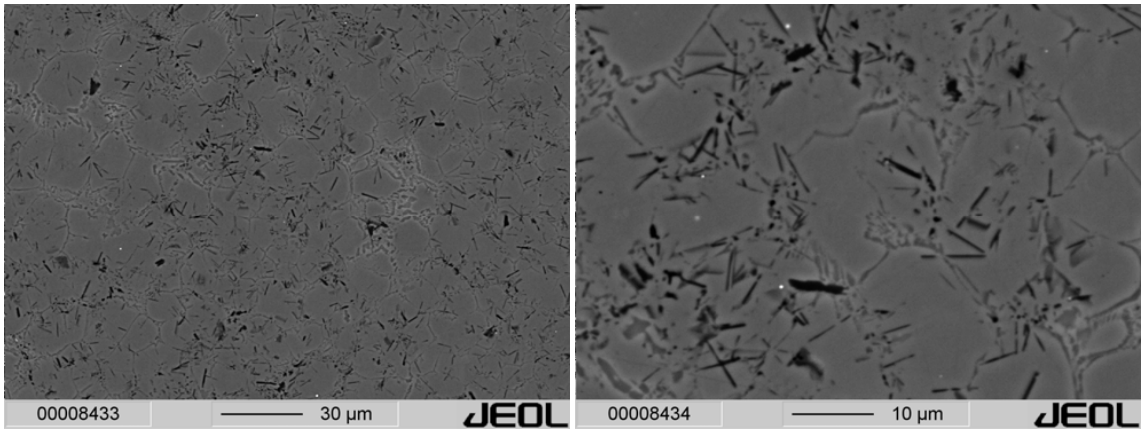
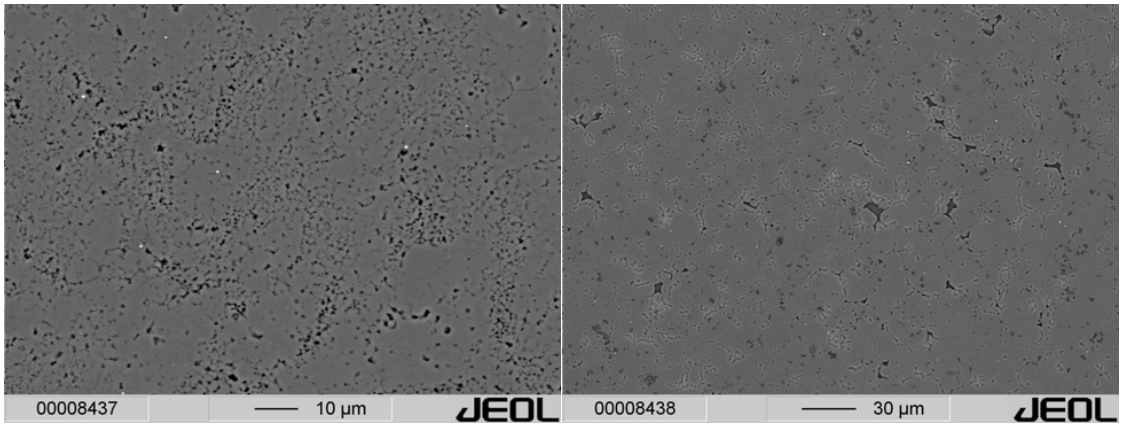


FIGURE 2. (Supplementary information) Backscattered electron images of synthetic grossular. a) and b) grossular of Haselton and Westrum (1980). Note the very fine dark needle-like crystals present within the lighter grossular crystals that form the matrix. The amount of the former varies greatly between different polycrystalline garnet chips and many chips show little to no such crystals, c) Gr-Kiel and d) Gr-92. Here, the dark areas represent spaces between crystals and surface polishing imperfections. The synthetic samples show many 120° triple-point junctions. The crystals are roughly between 5 and 20 µm in size.



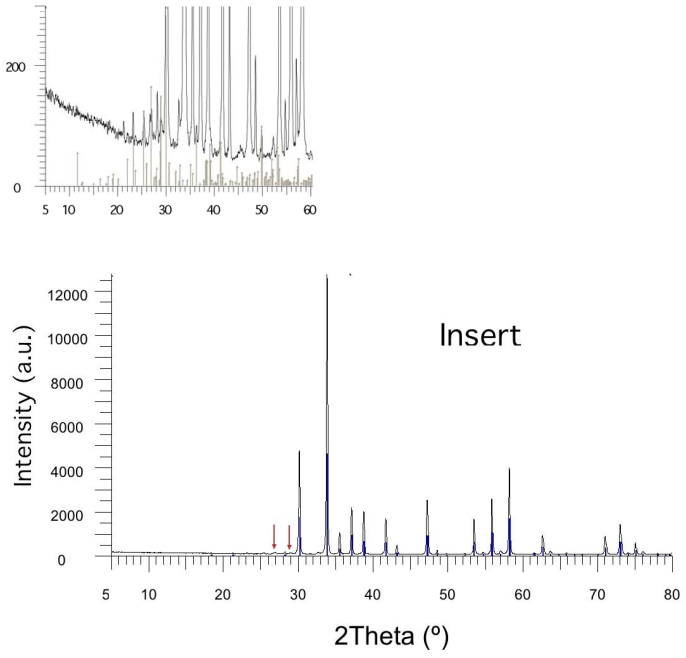


FIGURE 3. (Supplementary information) X-ray powder diffractogram for the sample Gr-Has (The other synthetic samples show similar patterns). Most of the very weak non-grossular reflections can be assigned to wollastonite or a wollastonite-like phase as shown, for example, by the arrows. The inset shows a portion of the diagram between 5 and 60° 2Θ θ. Wollastonite peaks are marked by the vertical lines.

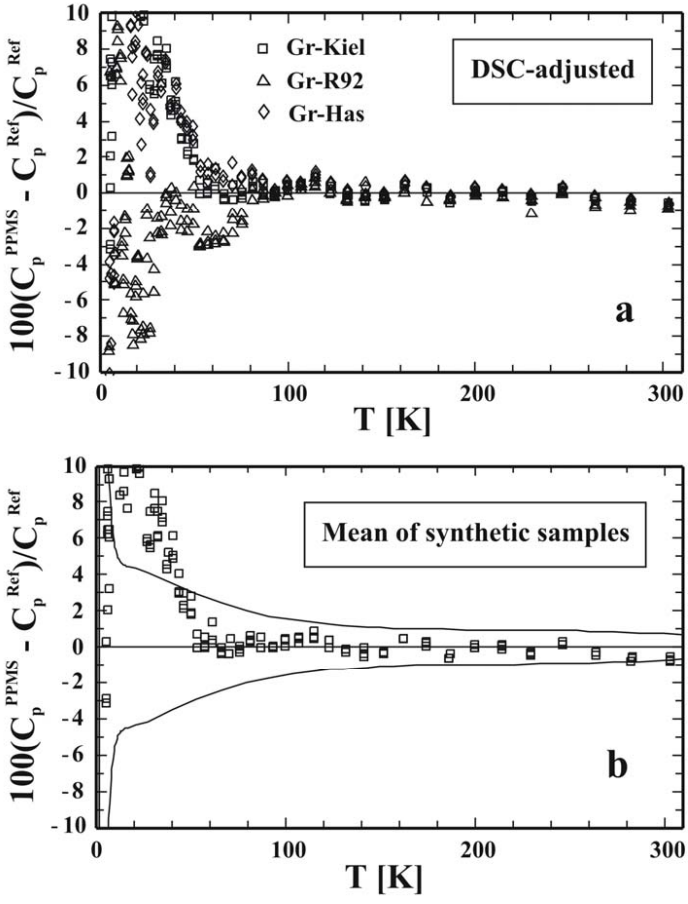


FIGURE 9. (Supplementary information) Percent difference plot between DSC-adjusted low-temperature heat capacity values (C_p^{PPMS}) for synthetic grossular and published values (C_p^{Ref}) obtained from adiabatic calorimetry (Haselton and Westrum 1980). (**a**) The three synthetic grossular samples Gr-Has, Gr-R92 and Gr-Kiel, (**b**) Their mean C_p values (the solid line is a $\pm 1\sigma$ uncertainty envelope).

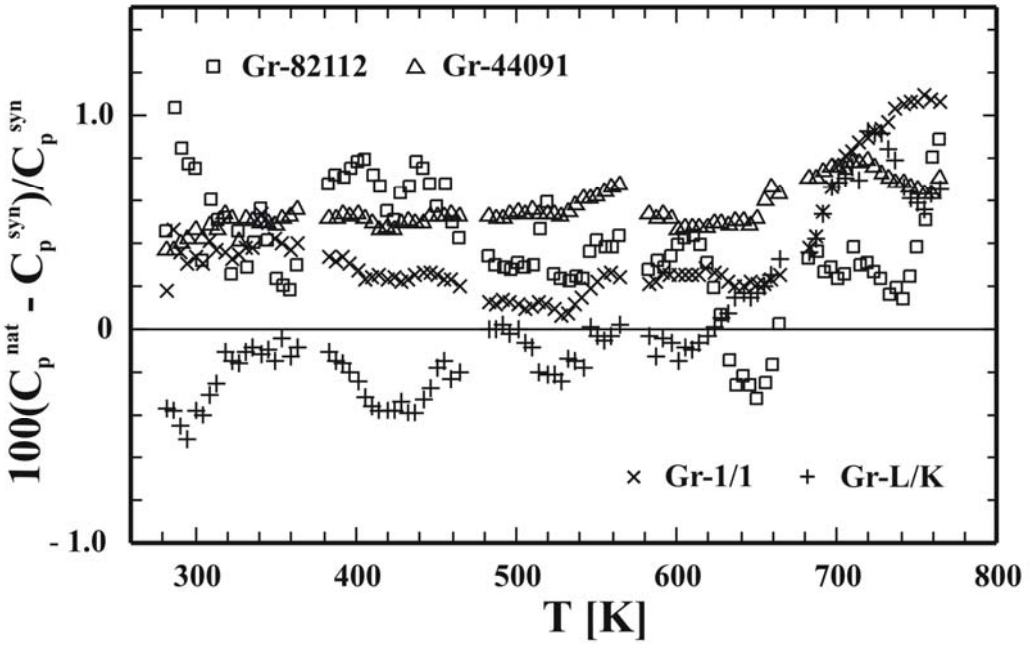


FIGURE 15. (Supplementary information) Percent difference plot between the high-temperature heat capacity values of natural grossular (C_p^{nat}) and synthetic grossular (C_p^{syn}) from data in this study. Natural garnets are Gr-82112 (squares), Gr-44091 (triangles), Gr-1/1 (X symbols) and Gr-L/K (crosses) (see Tab. 4). C_p^{syn} has been calculated using Eq. (4).

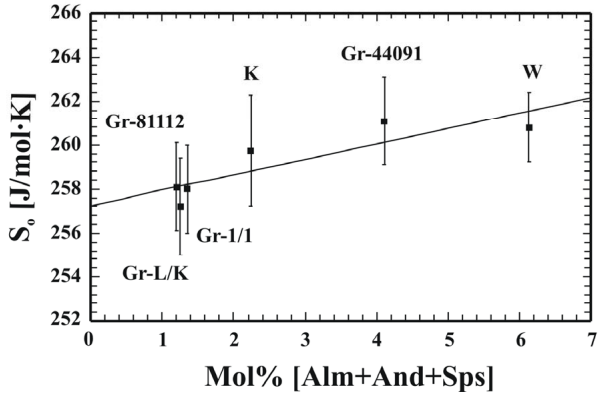


FIGURE 16. (Supplementary information) Standard entropy, S^o, values for the natural grossulars from this study as function of their mole percent almandine + andradite + spessartine components (Table 3). Two S^o values from the low-TAC study of Westrum et al. (1979) and Kolesnik et al. (1979) are also shown (labeled W and K, respectively). A straight line fit to the data intersects the ordinate at S^o = 257.6 J/mol×K.

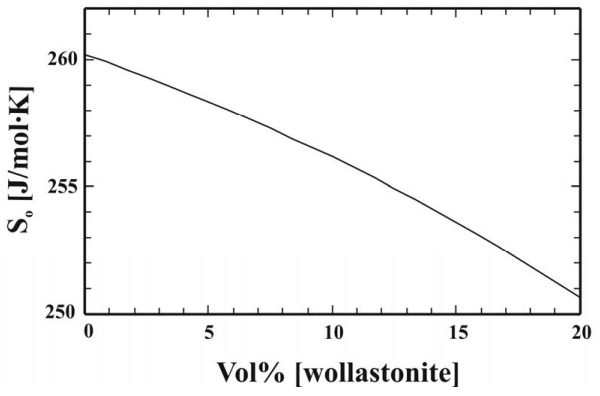


FIGURE 17. (Supplementary information) Plot showing the S^o value for grossular as a function of the volume % wollastonite impurity (see text for further details).