Supplementary information (SI)

Mineralogy and thermobarometry of marbles unravel retrogression and hydrothermal imprint of a polymetamorphic terrane (Făgăraș unit, Southern Carpathians, Romania)

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A1) Exsolution lamellae and non-stoichiometric carbonates

The exsolution-like brighter and darker lamellae found in dolomite grains are intriguing, as the microprobe data and WDS mapping showed that both lamellae represent dolomite (with insignificant higher Ba and Fe in the brighter lamellae compared to the darker ones). Very probably the observed exsolution-like lamellae do not represent compositional un-mixing but phase-antiphase domain boundaries, or polysynthetic twinning of dolomite.

Van Tendeloo et al. (1985) studied calcian dolomite (non-stoichiometric dolomite) by electron diffraction and showed indicative evidence for ordered superstructures. The superlattice in non-stoichiometric dolomite appears due to the ordered substitution of Mg by Ca in basal cation layers, and intercalation of additional Ca layers in the rhombohedral stacking, whereas the arrangement of CO_3 groups remains ordered. At higher temperatures, the carbonate mineral tends to be disordered (Ca layers in the rhombohedral stacking interrupting basal Ca-Mg ordered layers) and becomes ordered at lower temperatures. In disordered dolomite, Reeder and Nakajima (1982) found that the twin domain boundaries closely resemble antiphase boundaries (APBs), and therefore it is difficult to determine if the exsolution-like lamellae in dolomite represent phase-antiphase "exsolutions" or twinning. It is therefore easy to imply from Van Tendeloo et al. (1985) and Reeder and Nakajima (1982) that with higher disorder in dolomite (or calcite), the higher are chances to form non-stoichiometric dolomite (or calcite). With decreasing temperature, the non-stoichiometric carbonate becomes ordered, through developing phase-antiphase domains along planes of maximum atom density, resulting in a lamella with the appearance of twinning or exsolution.

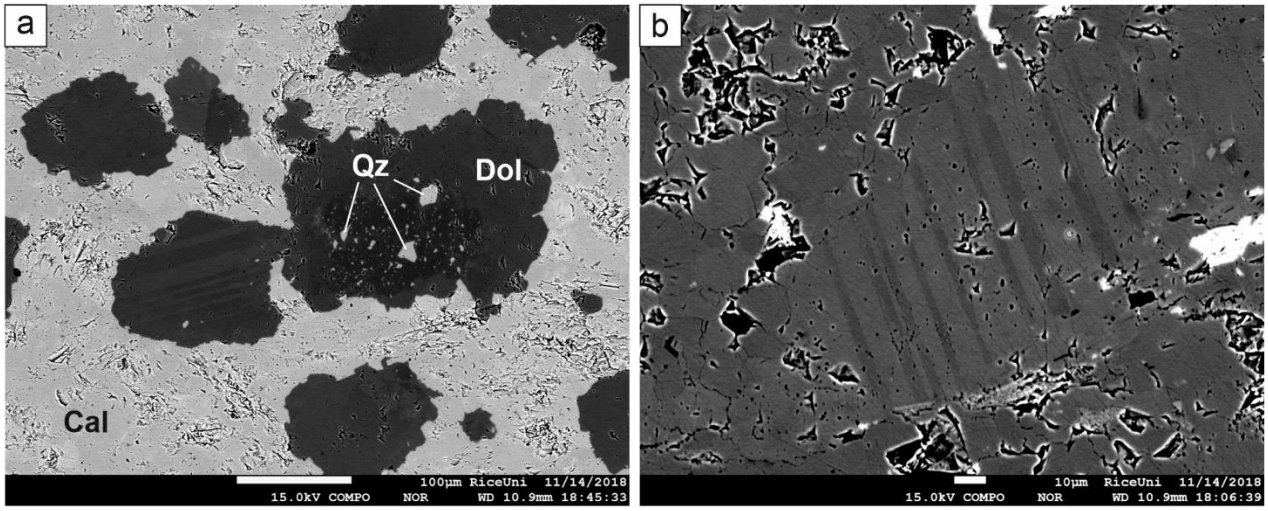

Figure S1. "Exsolution-like" lamellae in dolomite (high contrast BSE images). These lamellae show small differences in Fe and Ba. a) Exsolution lamellae and Qz micro-inclusions in rugged dolomite crystals surrounded by secondary calcite. Sample MPN-1; b) Close-up view of exsolution lamellae in dolomite grain. Sample MPN-1.

A2) T-X and P-X diagrams

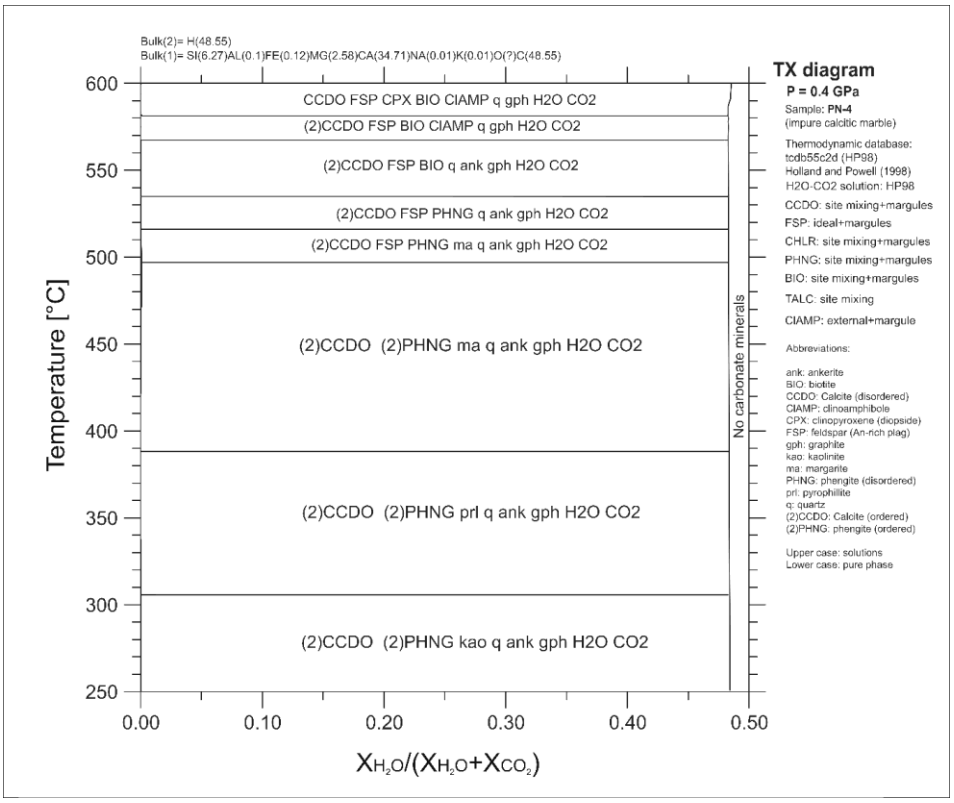

Figure S2. Temperature – $X_{H2O}/(X_{H2O}+X_{CO2})$ diagram for impure calcitic marble PN-4.

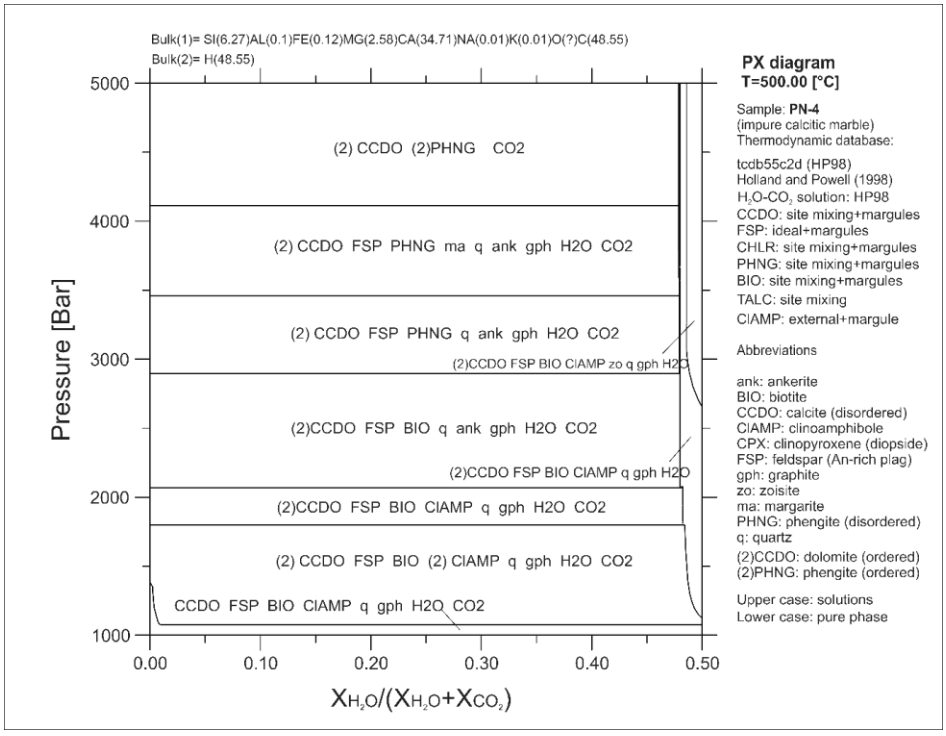

Figure S3. Pressure – $X_{H2O}/(X_{H2O}+X_{CO2})$ diagram for impure calcitic marble PN-4.

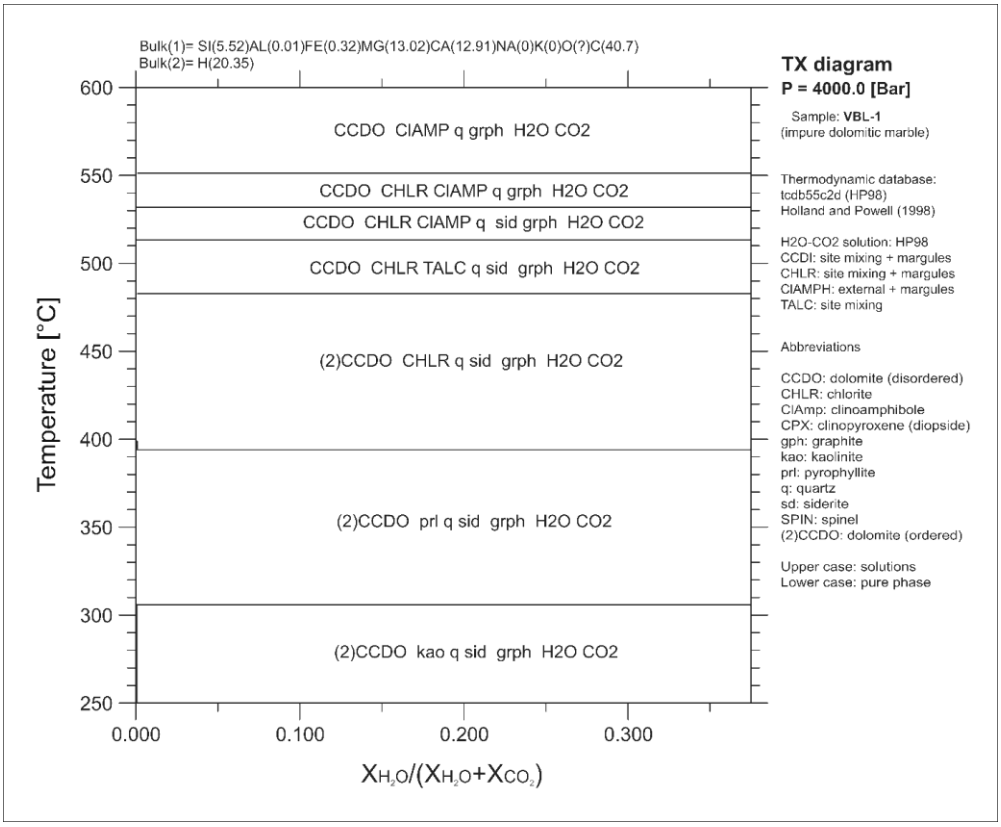

Figure S4. Temperature – $X_{H2O}/(X_{H2O}+X_{CO2})$ diagram for impure dolomitic marble VBL-1.

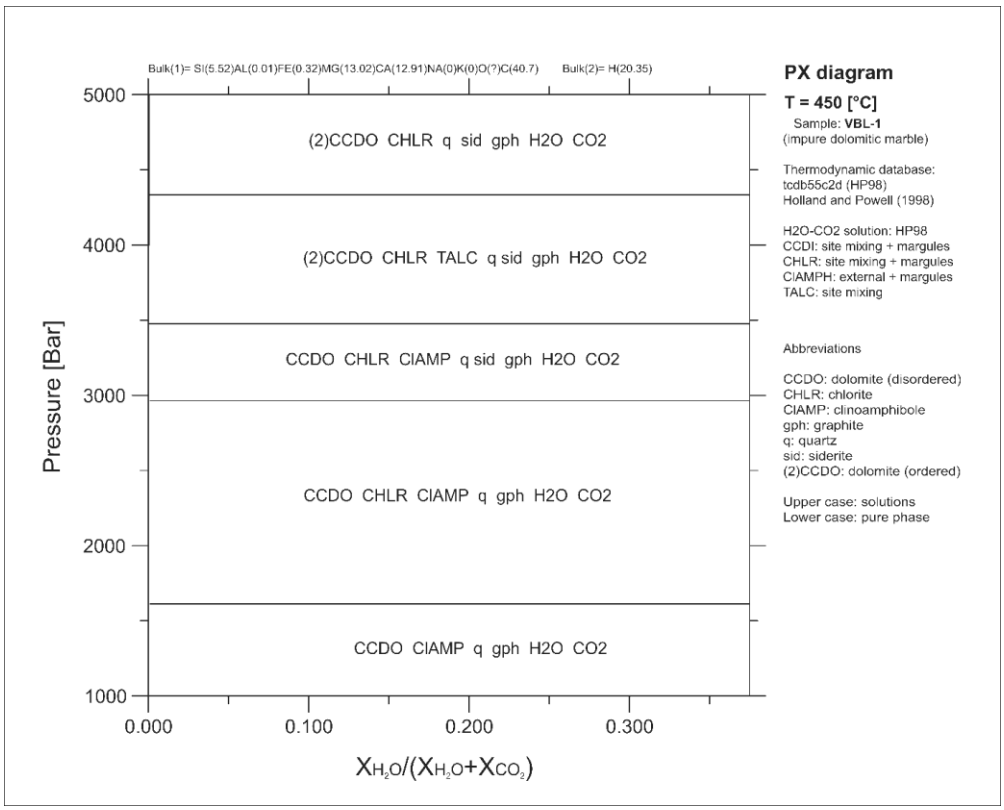

Figure S5. Pressure – $X_{H2O}/(X_{H2O}+X_{CO2})$ diagram for impure dolomitic marble VBL-1.

References

Reeder RJ, Nakajima Y (1982) The nature of ordering and ordering defects in dolomite. Phys Chem Miner 8(1):29–35. https://doi.org/10.1007/BF00311160

Van Tendeloo G, Wenk HR, Gronsky R (1985) Modulated structures in calcian dolomite: A study by electron microscopy. Phys Chem Miner 12(6):333–341. https://doi.org/10.1007/BF00654343

Supplement tables (Data repository)

SupplTable S1. EPMA analysis of carbonate minerals from Făgăraș Mountains, Southern Carpathians.
SupplTable S2. EPMA analysis of silicates and other minerals contained in the marbles of Făgăraș Mountains, Southern Carpathians.