

Reactions between CaCO₃ and bridgmanite at mantle conditions and the role of carbonates as possible trace element carriers into the Earth's deep interior

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Understanding the processes within the Earth's lower mantle and transition zone and their physical properties as well as compositional variations is one of the grand challenges in modern geoscience. One of the large unknowns concerning the Earth's lower mantle is the role of carbonates and their impact on the chemistry and dynamics of our planet. Recent studies have shown that carbonates are stable along the geotherm down to lower mantle conditions [1-3] where they may react with silicates [4, 5] to form new phases. These reactions will redistribute elements, which are carried by the carbonates, mainly Ca, Mg, Fe and CO₂ but also trace elements, such as Sr or REE. However, the stability of carbonates in presence of mantle silicates at relevant temperatures is far from being well understood. Related to this, very little is known about distribution processes of trace elements between carbonates and silicates. To shed light on these processes, we studied reactions between Sr and REE-containing CaCO₃ and bridgmanite at high pressure and temperature using synchrotron radiation based μ -X-ray diffraction and μ -X-ray fluorescence in a laser-heated Diamond Anvil Cell at beamline P02.2 at PETRA III and beamline ID27 at ESRF. X-ray diffraction allows monitoring structural changes whereas XRF shows chemical changes. Experimental conditions were chosen on basis of the stability field of bridgmanite and reached 40 GPa and about 2500 K. XRD and XRF maps covering the laser-heated spot were collected before and after the heating process. Single point XRD patterns recorded during heating of the assemblage allowed to observe the reaction between CaCO₃ and bridgmanite:



A comparison of the XRF intensity maps before and after heating indicates a change in the elemental distribution. By superimposition of the XRF-maps to the spatial distribution of the phases as determined from

XRD-maps, an increase in Sr-concentration can be found around the newly formed CaSiO₃-perovskite. Our recent findings on the elemental partitioning of La and Eu in carbonate-silicate reactions show no incorporation into silicate phases. Instead, La and Eu tends to form isolated oxide phases upon breakdown of CaCO₃. Further analyses were performed using TEM on FIB-foils cut from the recovered sample. This technique allowed us to gain more information on the elemental partitioning between carbonates and silicates on a nanoscale. The incorporation of Ca into MgSiO₃ silicate phases at high pressure and high temperature was observed and was used to follow kinetic and textural effects from heating.

Based on these experiments, we can conclude that certain elements (e.g. Sr, Ca) are redistributed among the newly formed phases. This effect is most pronounced upon breakdown of the carbonates. If the carbonate is not in contact with the silicate and remains stable, redistribution is more sluggish and could not be unequivocally constrained. In any case, our first findings imply that silicates may take up the trace elements that are provided by the carbonates and indicate that carbonates may have a major effect on the trace element contents of mantle phases.

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