Geophysical Research Abstracts
Vol. 15, EGU2013-8982, 2013
EGU General Assembly 2013

Investigations on the fate of subducted carbonates

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Carbon storage in the deep Earth is currently the subject of ongoing debate. There is much evidence for the presence of carbon cycling through the Earth's interior, such as the occurrence of diamonds from the upper and lower parts of the mantle, carbonate inclusions in diamonds and mantle xenoliths, the existence of carbonatite magmas, the presence of CO_2 in volcanic eruptions, etc. Now that carbonate-bearing subducting slabs are believed to pass through the transition zone and enter the lower mantle, interest has been focused on which carbon-bearing phases are the most stable at pressures and temperatures down to the core-mantle boundary. The solubility of carbon in the dominant mantle phases is low; hence carbon is likely stored in accessory phases, e.g., carbonates, diamonds/graphite, methane, carbides. Its distribution is a function of pressure, temperature, bulk composition and oxygen fugacity. At highly reducing conditions, the crystalline form of carbon is graphite or diamond, depending on P and T. At more oxidizing conditions carbonates are favored, due to the reaction between elemental carbon and oxygen to form (CO_3)2- groups, which bond to other cations such as Ca2+, Mg2+, Fe2+ and Na+ depending on the original bulk assemblage. Recent discoveries favor the presence of Fe-bearing carbonates at ultra high pressures and temperatures, suggesting the presence of carbonate solid solutions in the deep Earth. Several questions remain unsolved, however: 1) Which phase compositions are stable at pressures and temperatures of the lower mantle? 2) What is the effect of Ca on the redox stability of iron-bearing carbonates at high pressures and high temperatures? 3) Do spin transitions occur at lower mantle conditions in iron-containing $(Mg,Ca)CO_3$ phases? Our interest is currently focused on the study of carbonates at lower mantle conditions. We achieve high pressure and temperature conditions using externally heated or laser heated diamond anvil cells, which enable us to reach pressures and temperatures down to the bottom of the lower mantle. We are currently studying the spin transition of iron at lower mantle conditions in the pure end-member siderite ($FeCO_3$) and Fe-bearing solid solutions using Mössbauer spectroscopy. Siderite and carbonate solid solutions are being enriched in 57Fe to ensure a strong signal for Mössbauer spectroscopy. Our ultimate goal is to identify which phases may be the dominant carriers of carbon into the deep mantle at the relevant conditions of oxygen fugacity. Our experiments involve in situ analyses on carbonate end-members and solid solutions at lower mantle conditions using X-ray diffraction, Mössbauer, Raman and eventually IR spectroscopies. Samples analyzed after quenching to room pressure and temperature will be investigated using SEM, electron microprobe, TEM, Mössbauer spectroscopy and X-ray diffraction. The presentation will focus on our most recent results.