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Fe-bearing carbonates in the lower mantle: an experimental approach

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Carbon, a constituent element of life, is continuously exchanged between the surface and the deep Earth's interior. During subduction, this element is brought inside the Earth mainly by means of carbonate, and Fe-bearing carbonates are potential carbon carriers down to the deep lower mantle. Indeed, the presence of iron influences the stability of these phases at high pressures and high temperatures (HPHT), partly due to the spin-pairing of Fe-d electrons. Our study is focused on identifying and characterizing the dominant carbon phases in the deep mantle at the relevant conditions of fO_2, P and T, and investigating their thermoelastic properties. To achieve this goal, we performed experiments in a laser-heated diamond anvil cell (DAC) to generate the HPHT conditions prevailing in the Earth's lower mantle. We used a variety of in situ analysis methods, including X-ray diffraction, Mössbauer, Raman and XANES spectroscopies and nuclear inelastic scattering (NIS). All samples were enriched in 57Fe to ensure a strong signal for Mössbauer spectroscopy and NIS. We determined the spin crossover pressure of Fe in single-crystal $FeCO_3$ and $Mg0.74Fe0.26CO_3$ at room temperature using Mössbauer and Raman spectroscopies to be 41(1)-45(1) and 45(1)-48(1) GPa, respectively, showing a slight effect of composition on the transition pressure. We observed that spin crossover of both compositions shifts to higher pressures at $T > 650$ K based on HPHT Raman experiments. We discovered that new C-rich phases formed from breakdown of these compositions in laser-heated DAC experiments using in situ XANES and synchrotron Mössbauer source (SMS) measurements, which was confirmed by ex situ XRD patterns after quenching. The new C-rich phases contain Fe3+ as observed using SMS spectroscopy, both in situ and after quenching, which implies that redox reactions took place. For comparison, our experiments have also included room temperature measurements of $NiCO_3$ using Raman and XANES spectroscopies, which show no transitions up to 67 GPa. The presentation will focus on our most recent results and discuss their implications for the carbon cycle in the deep Earth's interior.