Report about the experiment HS2330 performed on ID30 beamline

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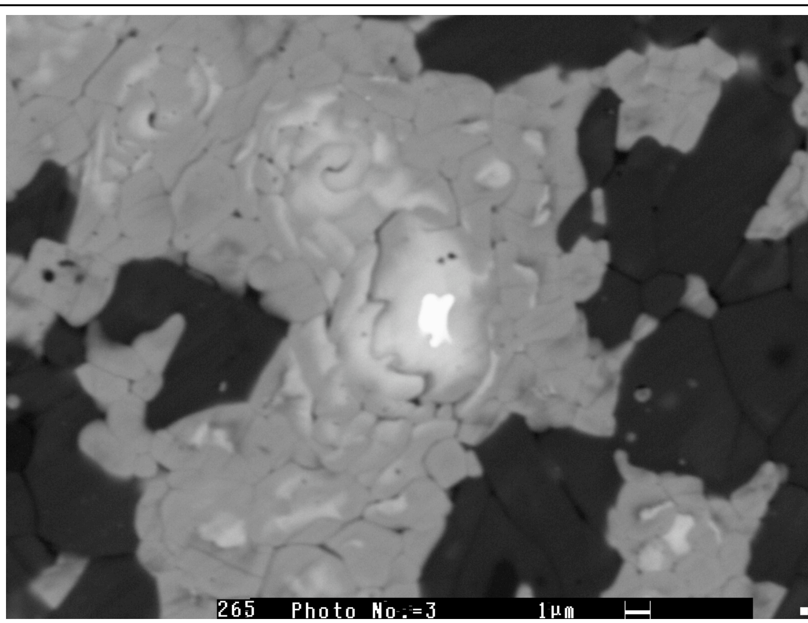
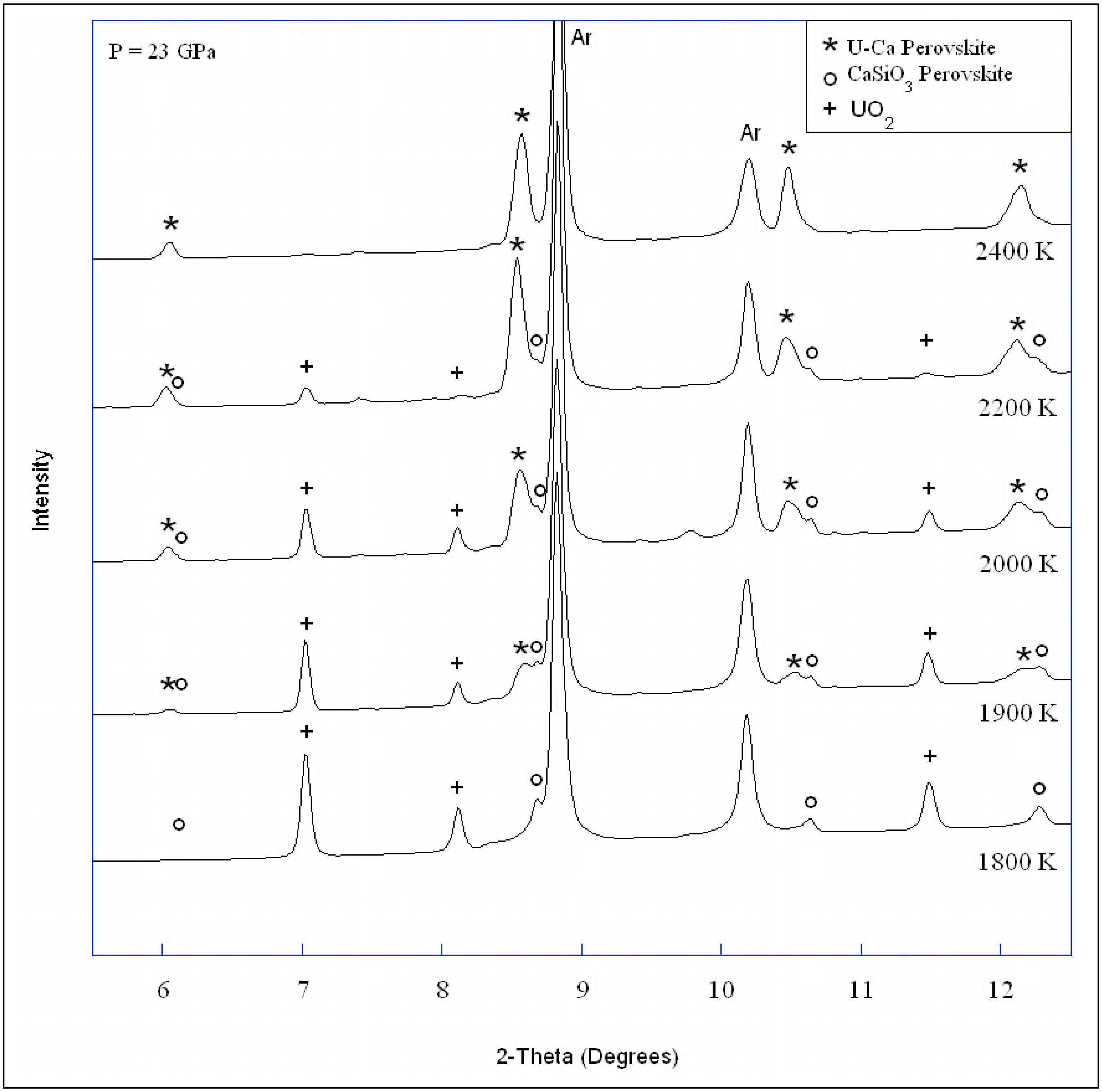
It is generally agreed that about half of the total heat flux (44 TW) at the Earth surface comes from the mantle, through the radioactive decay of uranium, thorium and potassium[1], with about 9 TW accounting for U alone. It is essential to determine the location of these heat sources for a better understanding of the geodynamics and thermal behaviour of the Earth. In order to determine the eventual host minerals for uranium in the deep mantle, we have performed solid-solid reactions between synthetic $Ca_3Al_2Si_3O_{12}$ grossular garnet and natural uraninite UO_2 at high pressures and high temperatures using laser-heated diamond anvil cell at ESRF ID30 beamline.
Preliminary multi-anvil experiments revealed that a phase of stoichiometry CaSiO3 was able to incorporate large amounts of uranium. The following figure displays a backscattered electron image of a so-called cell of diffusion of uranium : we can see clearly that the diffusion is occuring from UO_2 (white) to the silicate phase (light and dark grey).



Multi-anvil experiment

Cell of diffusion of Uranium (white) in a matrix of silicate (grey).
In dark accessory materialsI

In situ angle-dispersive x-ray diffraction performed at ID30 beamline allows us to observe the same reaction of incorporation of uranium in a phase $CaSiO_3$ with a perovskite-type structure. This study shows a good correlation between Multi-Anvil Press (MAP) and Laser-Heated Diamond Anvil Cell (LHDAC) experiments. The figure below displays the successive diffraction spectra at 23 GPa, while temperature increases. We see peaks of UO2 disappearing while those of a new phase (slightly distoreted compared to the pure CaSIO3) appear. This is an experimental evidence of the incorporation of uranium in a new high pressure phase.



It is believed that about 20 ppb of U is present in the bulk silicate earth[3] (that is, mantle + crust) and about 50 % of total U in the Earth is stored in the lower mantle. This U-bearing $CaSiO_3$ high pressure phase could be part of a compositionally distinct and dense layer containing heat-producing elements, which has been proposed at depth from around 1600 km to the Core-Mantle Boundary[2]. Since calcium is the largest of the major elements present in the Earth's mantle, it is then a good candidate for a substitution by a voluminous element like uranium : thus Ca-rich high pressure phases are possible hosts for large cations like uranium.

Note that the behaviour of natural uraninite UO_2 at high pressure has been determined : UO_2 adopts an orthorhombic structure at pressures above 35 GPa, and the expected cotunnite-like structure was not observed at pressures up to 80 GPa.

References

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