Using TOF-SIMS isotope mapping for studying dissolution and precipitation processes at mineral grains in an experimental CO_2-sequestration setup

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CCS (Carbon dioxide capture and sequestration) is a technique investigated for its possible employment in the reduction of the amount of anthropogenic CO_2 gas emitted into the atmosphere. Deep saline aquifers are one option for storing CO_2 gas streams produced e.g. by the combustion of fossil fuels at power plants. These gas streams contain different impurities depending on their origin, among them O_2, NO_x, SO_x in addition to the CO_2. We have used ToF-SIMS to determine the influence of these impurities on dissolution and precipitation processes at the minerals.

Experiments under in situ pressure and temperature conditions of possible geological storage sites were performed at the German Bundesanstalt fuer Geowissenschaften und Rohstoffe (BGR) in flexible Dickson-type gold-titanium cells and small gold capsules.

Minerals common in the deep saline aquifers (e.g. siderite, calcite) were placed in small reaction cells of thin gold foil in a reactor vessel and exposed to isotopically enriched water ($H_2{}^{18}O$) or carbon dioxide ($^{13}CO_2$) during the experiments.

To facilitate the determination of the amount of dissolved ions incorporated into newly formed precipitates within the reaction chambers, a database of positive and negative ToF-SIMS spectra for a variety of rock-forming minerals were set up to identify minerals and mineral alterations. In addition, preparation techniques were developed for high-resolution measurements of the incorporation of isotope-labeled elements/ions (e.g. ^{18}O) into mineral precipitates. The results show that ToF-SIMS can simultaneously image the elemental, isotopic, and molecular compositions of these minerals with high spatial resolution. Also, elemental and isotopical distributions as a function of depth can be monitored.

Carbonatitic Magmas? A Mineralogical and Isotopic Approach

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Massive deep orange calcites are intensely colored, coarse grained rocks, in contact with silicate rocks that exhibit centimetric crystals of phlogopite, occurs at Serrinha Nucleus (SerN), Brazil. Occeli are a common feature, composed of euhedral fluorapatite, diopsidic pyroxenes and barite. Associated creamy coloured rocks are composed of >90% calcite, 3-8% green-olivine and diopside, 1-2% euhedral sulphides and well developed crystals of purple-spinel and blue-fluorapatites, as well as rounded mafic enclaves composed of amphibole, biotite and pyrite. Coarse calcite veins and/or dykes also cut the 2.1 Ga syenite-lamprophyre association and the meta-gabbros of Itapicuru Greenstone, demonstrating a clear interrelation between carbonates and silicate minerals including Fe-Mg-Mn calcite, diopside, apatite, quartz and Fe-Ni oxides. Rutile, few zircons and pyrite were described from the heavy mineral concentrates.

C and O preliminary results for $\delta^{13}C$ (PDB) between +12 e +13 per mil, $\delta^{18}O$ (PDB) of -10 per mil, and V-SMOW of +20, suggest the involvement of different magmatic or hydrothermal phases in these rocks formation. LA-ICP Pb-Pb analyses of calcites yielded model ages of ~1950 Ma and U-Pb ages from 2 zircons are (i) 2.11 Ga, close to that of SerN syenites and lamprophyres, suggesting that these rocks may be comagmatic and (ii) a clear core-border relationship showing ages of 2.7 and 2.5 Ga, respectively, similar to those reported for zircon xenocrysts in syenites and kimberlites.

Our results show that these rocks are calcite carbonatites, fairly well exposed, and that have not been metamorphosed. The occellar texture suggests liquid immiscibility, indicating silicate magma droplets. Although further work remains to be done, the isotopic data suggest an emplacement age of ~2.0-2.1Ga, coeval with emplacement of the syenites and lamprophyres and to the gold mineralization at Itapicuru Greenstone. This syenite-lamprophyre-carbonatite association is closely related to Au in many Archean cratons worldwide.

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DOI:10.1180/minmag.2013.077.5.18