

## CO<sub>2</sub>-basalt interaction – Numerical simulation and experimental study

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Burning of fossil fuel during the past several decades has lead to increased atmospheric CO<sub>2</sub> content and there are increasing evidences that this change has affects the Earth's climate. A reduction of atmospheric CO<sub>2</sub> is considered an essential step in the control of global warming. One possibility is geological sequestration where CO<sub>2</sub> is injected into permeable rock formations whereas it reacts with the rocks, forming secondary minerals including carbonates mm.

Basaltic rocks are reactive and contain minerals and glasses with substantial Mg, Fe and Ca content needed for carbonate formation. However, the interaction of CO<sub>2</sub> containing water with basalt is complex. Reaction path simulations and closed system batch experiments were carried out in order to evaluate the solution and mineralogical changes during CO<sub>2</sub>-water-basalt interaction. The results indicate that addition of CO<sub>2</sub> (2-30 bar) to aqueous solutions significantly changes the reaction path. Initially, the pH is buffered at 4-6 by the CO<sub>2</sub> ionization, proton consumption upon basalt dissolution and proton consumption/release upon secondary mineral formation. The main secondary minerals formed during this stage are SiO<sub>2</sub> (opal, chalcedony) and Mg-Fe and Mg-Ca carbonates. The system is buffered under these conditions until the concentration of CO<sub>2</sub> has been lowered significantly by carbonate precipitation. Upon further basalt dissolution, the pH rises to >8 and the system becomes buffered by mineral dissolution and precipitation reactions. The associated secondary mineralogy changes with (Ca)-Fe-Mg smectites, SiO<sub>2</sub>, Ca-Na zeolites and some calcite dominating. The formation of Fe and Mg rich smectites limit the mobility and availability of Fe<sup>2+</sup> and Mg<sup>2+</sup> and hence inhibit the formations of Mg and Fe containing carbonates. On the other hand, Ca<sup>2+</sup> availability and alkaline pH causes calcite to become the stable carbonate.

## Generation of plumes at the margins of chemically distinct Large Low Shear Velocity Provinces

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The African and Pacific Large Low Shear Velocity Provinces (LLSVPs) of the Earth's lowermost mantle are the most prominent features of all recent shear wave tomography models. Here we provide further evidence that they are bodies of distinct material: The frequency or occurrence of shear-wave tomography models close to the CMB shows a distinct bimodal distribution, with a larger peak at higher velocities and a smaller peak at lower velocities. The relative size of the peak at higher velocities gets larger higher up in the mantle. Correspondingly, regions of strong lateral gradient in shear wave speed frequently occur (for the smean tomography model) approximately at the -1% shear wave anomaly contour surrounding LLSVPs above the CMB. The two LLSVPs bounded by the -1% contour together occupy a surface area of ~20% close to the CMB, which reduces to ~10% 300 km above the CMB, and thus contain about 2% of mantle material.

Large Igneous Provinces (LIPs) of the past 300 Myr, when reconstructed to their eruption sites, as well as many active hotspots cluster above the margins of these bodies at their base. We therefore infer that plumes are generated at these margins, which appear to have remained rather stable for about 300 Myr or longer. If the LLSVPs are chemically distinct hotter material, these margins are inclined thermal boundary layers, providing a preferred formation environment for thermal instabilities and hence mantle plumes. The bimodal distribution in combination with long-term stability further supports the idea that these bodies are chemically distinct; a purely thermal anomaly would likely not have remained stable for so long. Furthermore, core-mantle boundary topography predicted by geodynamic models is in better agreement with observational constraints, if s-wavespeed anomalies below -1% in the lowermost ~300 km of the mantle have an additional non-thermal density anomaly.

Because of their long-term coherence LLSVPs could form the isolated reservoir invoked by some to explain the distinctive isotopic compositions of terrestrial rocks. Plumes of deep-seated origin appear to get generated only where (i) the outer core, (ii) one of the LLSVPs, and (iii) the seismically faster part of the deep mantle meet. Some of their geochemical characteristics may be be explicable as a consequence of this specific formation environment. For example, the relatively high <sup>3</sup>He/<sup>4</sup>He ratio of hotspots and young LIPs can be attributed to diffusion into the plume generation zones from the hot basal parts of the LLSVPs.