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Influence of iron redox state on SO_2 scavenging by rhyolitic glass

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We investigate high-temperature SO_2 scavenging by natural calc-alkaline rhyolite particles. We simulate this process experimentally, by reacting powdered rhyolite glass particles of a range of grainsize distributions with SO_2 atmospheres at high temperature for various exposure times. The gas phase is either hydrous (SO_2-H_2O) or anhydrous (SO_2). Our results show that the principal factors affecting reaction rates are temperature and particle size such that SO_2 uptake is amplified by using small particles at high temperatures. Electron probe analysis measurements of polished thick sections of the pre- and post-treated samples, show depletion of calcium near the edges of the particles, and a diffusion gradient toward the centre of the particles, which remain at the initial value. This implies that it is diffusion of calcium toward the surface that accommodates the reaction. Inspection of the surfaces shows prodigious quantities of sulphur-bearing calcium salts. Leachates of the treated glass powder confirmed that calcium and sulphur are combined in soluble surface compounds. We propose that this reaction involves the reaction of SO_2 with CaO surface sites, initially forming $CaSO_3$, further oxidizing to a more stable from, i.e. CaSO4. In a second step, we show that the outward motion of calcium is charge balanced by auto-oxidation of iron such that the bulk Fe3+/Fetotal ratio, and varied from an original (i.e. untreated glass powder) value of 0.15, up to 0.51. The increase of the Fe3+/Fetotal ratios correlates with calcium depletion. To assess the influence of iron content on calcium diffusion, thus on sulfur uptake, we produced a wide range of rhyolitic-like glasses (haplogranitic composition; K2O-Na2O-Al2O_3-SiO_2) with various amounts of CaO; 1 and 2 wt%, and Fetotal; 0, 0.1, 1, 1.5, 2, and 2.5 wt%. Such experiments will give us a closer insight into the role of Fetotal in glasses on sulfur uptake at high temperatures and the cause-effect-like processes linked to calcium diffusion and Fe2+ oxidation. Ultimately, our work demonstrates the conditions and compositional-dependence of sulphur sequestration process, and has wide implications for in-plume and in-conduit scavenging during rhyolite eruptions.