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Mineralogical and chemical changes induced by experiments of interaction between supercritical CO_2 and plutonic mafic rocks. A case study in Portugal.

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The focus of this research is a qualitative study of mineralogical and chemical changes in plutonic mafic rock samples after exposure to a CO_2-rich brine, under supercritical conditions (SC), to clarify the behavior of brine and rock in the initial stages of mineral carbonation. The studied rock consists of a gabbro-anorthosite from the Odivelas massif, in southern Portugal. The sample was exposed to a SC CO_2-rich brine (P≈8 MPa, T≈40□C) for runs of 0, 30 and 90 days. Experiments were conducted in batch mode, ie. with no CO_2 flow, and with a proportion of CO_2 to brine of 0.226 for 30 days and 0.033 for 90 days. In addition, numerical modeling was applied to complement the experimental observations, reproducing the experimental observations and simulate the chemical behavior for longer times. The chemical analysis of the brine, before and after, the experiment, shows: (i) increase of magnesium (Mg^{2+}), calcium (Ca^{2+}) and silica (SiO_2) for the 30 and 90 days runs and (ii) decrease of pH (8.1 to 6.1 and 8.1 to 6.3, respectively). Experimental and numerical results indicate that the rock sample suffered a slight dissolution process with mineralogical/textural readjustments on the external area of the specimens studied. This is thought to mimic the initial dissolution process under early-stage mineral carbonation. After 90 days, apart from halite, there are no significant new mineral phases. However, the elemental association in the EDS maps of carbon and magnesium dissociated from silicon suggests the residual crystallization of magnesite.