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# Earth science: Role of $fO_2$ on fluid saturation in oceanic basalt

[Brief Communications Arising]

Scaillet, Bruno; Pichavant, Michel

Assessing the conditions under which magmas become fluid-saturated has important bearings on the geochemical modelling of magmas because volatile exsolution may profoundly alter the behaviour of certain trace elements that are strongly partitioned in the coexisting fluid 1. Saal et al.2 report primitive melt inclusions from dredged oceanic basalts of the Siqueiros transform fault, from which they derive volatile abundances of the depleted mantle, based on the demonstration that magmas are not fluid-saturated at their eruption depth and so preserve the mantle signature in terms of their volatile contents. However, in their analysis, Saal et al.2 consider only fluid-melt equilibria, and do not take into account the homogeneous equilibria between fluid species, which, as we show here, may lead to a significant underestimation of the pressure depth of fluid saturation.

For any basalt melt that is at fixed temperature and pressure in fluid-saturated conditions with known  $H_2O$  and  $CO_2$  concentrations, the corresponding volatile fugacities,  $fH_2O$  and  $fCO_2$ , can be calculated 3. The phase rule states that this in turn fixes the fugacities of all other C-O-H fluid species, including  $fO_2$  (ref. 4). Figure 1a shows the covariation of the mole fraction of  $H_2O$  and  $CO_2$  ( $XH_2O$  and  $XCO_2$ ) in a C-O-H fluid calculated for various  $fO_2$  at 1,200 °C and 400 bar ( $fO_2$  expressed in log units relative to the solid buffer Ni-NiO, referred to here as NNO). It can be seen that at a very low mole fraction of  $H_2O$  ( $XH_2O < 0.05$ ), reduced fluids are poorer in  $CO_2$  than oxidized ones: for instance, at  $\Delta NNO = -2$  the mole fraction of  $CO_2$  is 0.8, whereas at  $\Delta NNO = -0.8$ , it is 0.95. This is due to the progressive reduction of  $CO_2$  into CO, which becomes significant at  $fO_2$  below  $\Delta NNO = -1$  (ref. 4).

Figure 1b shows the  $H_2O$  and  $CO_2$  concentrations of basalt melts that coexist with fluids shown in Fig. 1a. Under oxidizing conditions ( $fO_2 > \Delta NNO = -1$ ), the overall shape of the curve resembles the pattern of the curve when it is calculated by considering only fluid-melt equilibria 2. By contrast, for  $fO_2 < \Delta NNO = -1$ , the isobaric curve displays an asymmetric bell-shaped pattern characterized by a strong lowering of the melt  $CO_2$  content at low  $H_2O$ . As already stated, this is the result of the reduction of  $CO_2$  to CO at low  $fO_2$ , CO being an insoluble species in silicate melts at low pressures 5. The two curves merge at melt  $H_2O$  contents higher than 1 wt%, which shows that, for basalt melts with a higher meltwater content, the calculation of pressure for fluid saturation in the C-O-H system does not require an accurate knowledge of their redox state - unlike  $H_2O$ -poor basalts, such as oceanic basalts 2.

The  $fO_2$  of primitive melt inclusions at Siqueiros is at present not well constrained but is estimated to be around  $\Delta NNO = -2$  (ref. 2), which would fall at the upper end of the range of  $fO_2$  estimated for mid-ocean-ridge basalt 6 (MORB). However, given the general inverse correlation between  $fO_2$  and MgO of MORB documented worldwide 6, the Siqueiros magmas would be expected near the lower end of the range (that is,  $\Delta NNO = -3.5$ ; ref. 6). The  $CO_2$  contents of Siqueiros melt inclusions average at  $132 \pm 34$  p.p.m. but range from 43 p.p.m. up to 243 p.p.m. (ref. 2).

Figure 1c shows the evolution of the pressure of fluid saturation with  $fO_2$  of basalt melts having 40, 132 and 240 p.p.m.  $CO_2$  and 0.1 wt%  $H_2O$ . It can be seen that, except for the lowest  $CO_2$  contents, most melts would be fluid-saturated at their collection pressure for an

$fO_2 < \Delta NNO = -2.5$ . Considering the uncertainties associated with the determination of dissolved  $CO_2$  in MORB glasses ( $\pm 15$  p.p.m.) and with the redox state of Siqueiros magmas, we contend that the condition of fluid saturation before eruption cannot be disregarded for at least the most  $CO_2$ -rich Siqueiros melt inclusions.

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We note that this condition is in agreement with earlier findings showing that the redox state of oceanic basalts is compatible with mantle melting under fluid-present or graphite-saturated conditions 7,8. Therefore, although the variable  $CO_2$  content of quenched oceanic basaltic glasses results from syneruptive degassing 3, part of this variability may also reflect regional-to-local variations in  $fO_2$ . In general, a quantitative modelling of volatiles' behaviour in MORB magmas will require explicit consideration of the role of  $fO_2$  (ref. 9).

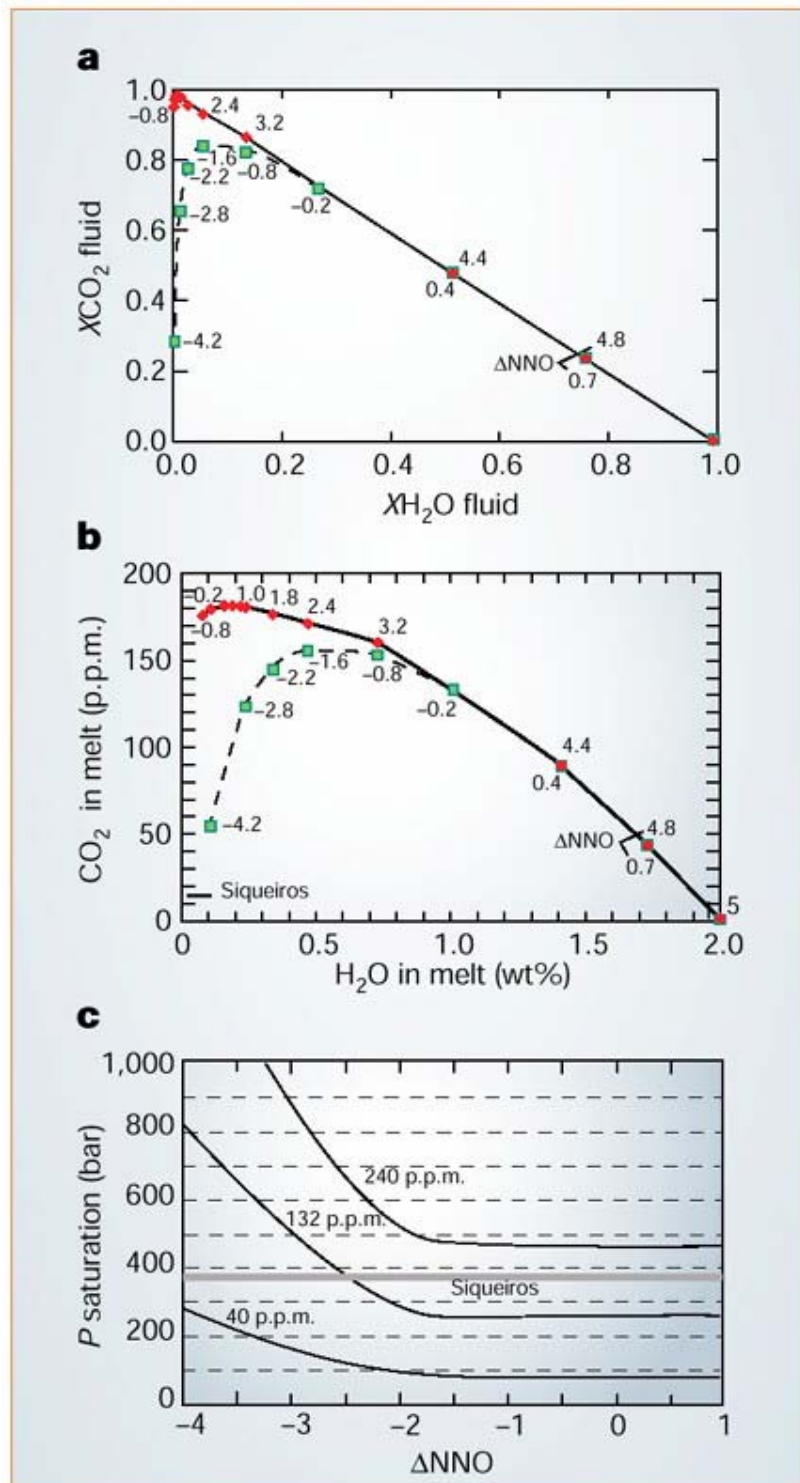


Figure 1 Effect of  $f_{O_2}$  on fluid speciation and fluid saturation in basalts. a, Covariation of  $X_{H_2O}$  and  $X_{CO_2}$  (where  $X_i$  is the mole fraction of species  $i$ ) in a C-O-H fluid calculated for various values of  $f_{O_2}$  (numbers along each curve). The calculations were done by fixing  $f_{H_2}$  (either 0.01 bar or 1 bar, corresponding to red and green symbols, respectively) and  $f_{H_2O}$ , which allows us to calculate  $f_{CO_2}$  in the C-O-H system 4. Once  $f_{H_2}$  and  $f_{H_2O}$  are fixed,  $f_{O_2}$  can be calculated through the equilibrium  $H_2 + 0.5 O_2 = H_2O$ . T, 1,200 °C; P, 400 bar. b, H<sub>2</sub>O-CO<sub>2</sub> solubility diagram for a basalt at 1,200 °C and 400 bar and equilibrated with fluid compositions shown in a. For any given  $f_{H_2O}$  and  $f_{CO_2}$  set of values, the corresponding H<sub>2</sub>O and CO<sub>2</sub> contents of the melt are computed according to ref. 3. The  $f_{O_2}$  is shown along each line in log units calculated relative to the solid buffer Ni-NiO. The Siqueiros bar shows the range of H<sub>2</sub>O content of Siqueiros melt determined by Saal et al. 2. c, Evolution of the pressure of fluid saturation with  $f_{O_2}$  of a basalt melt having 40, 132 and 240 p.p.m. CO<sub>2</sub> and 0.1 wt% H<sub>2</sub>O, which are minimum, average and maximum CO<sub>2</sub> contents, respectively, of the Siqueiros melt inclusions 2. At an  $f_{O_2}$  below  $\Delta NNO = -1$ , the pressure of saturation in fluid rises because of the continuous increase in CO of the coexisting gas phase. Grey horizontal line corresponds to the average collection pressure of Siqueiros basalts.