Oxidation of La Réunion lavas with MORB-like fO_2 by assimilation

R.W. Nicklas^{1*}, R.K.M. Hahn¹, J.M.D. Day¹

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Abstract

Oxygen fugacity (fO_2) is poorly constrained in mantle reservoirs, especially those sampled by ocean island basalts (OIBs). This is partly due to complications from secondary processes acting on fO_2 in OIB parental magmas. To investigate these issues *in situ* trace element data of olivine in lavas from the island of La Réunion are reported. La Réunion lavas are useful for examining post-melting modification due to their limited range in isotopic signatures, indicating a largely homogenous mantle source. Vanadium abundances in olivine were used to constrain the lava fO_2 to between +0.2 and +1.5 ΔFMQ , intermediate between that of mid-ocean ridge basalts (MORBs) and other OIBs. The variable fO_2 of Réunion lavas results from assimilation processes within the volcanic edifice, and not source heterogeneity. The Réunion mantle plume likely has fO_2 similar to that of MORB, despite isotopic evidence for the plume sampling an early-formed isolated mantle reservoir. Assimilation processes can have a pronounced effect on lava fO_2 and should always be accounted for when constraining mantle redox.

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Introduction

Oxygen fugacity (fO_2) of primitive lavas is a proxy for the fO_2 of their mantle sources and is useful for constraining mantle redox variability (Carmichael, 1991; Moussallam *et al.*, 2019; Nicklas *et al.*, 2021). Redox constraints are important for testing models of recycling into the deep Earth, as well as for tracking mantle differentiation and mixing (Carmichael, 1991; Frost *et al.*, 2008; Brounce *et al.*, 2017). While fO_2 in primitive melts can be modified by processes such as fractional crystallisation (Kelley and Cottrell, 2012), crustal contamination (Grocke *et al.*, 2016) and degassing (Moussallam *et al.*, 2019), the fO_2 of erupted basalts is, to a first order, controlled by the fO_2 of their mantle source (Birner *et al.*, 2018). Oxygen fugacity varies strongly with tectonic environment (Carmichael, 1991), as arc basalts ($\Delta FMQ \geq +2$) have elevated fO_2 relative to mid-ocean ridge basalts ($\Delta FMQ \approx 0$), possibly due to metasomatism of the mantle wedge by oxidised recycled materials (Kelley and Cottrell, 2012), or due to distinct differentiation processes (Tang *et al.*, 2018). Global mass balance calculations show, however, that ~90 % of oxidants are not returned in arc volcanism and are instead taken into the deep mantle (Evans, 2012). Ocean island basalts are the result of melting in deep-seated plumes that contain a variety of recycled crustal lithologies (e.g., Hofmann, 1997), and so are expected to contain some of these recycled oxidants, of which Fe^{+3} is the most important (Evans, 2012).

Compared to arc lavas and MORB, OIB fO_2 is not as comprehensively documented. This is partly due to the general absence of glassy materials for X-ray near edge absorption spectroscopy (XANES) measurements meaning that alternative oxybarometry methods are required. It has recently been suggested that OIBs could be equally or even more oxidised than arc lavas

(Moussallam *et al.*, 2019), possibly resulting from sampling oxidised altered oceanic crust (AOC). Sulfur degassing of OIB melts can also have a variable and large (~2 log units) effect on the fO_2 measured in glasses and melt inclusions (Moussallam *et al.*, 2019), making some OIBs appear to be as reduced as MORBs. In contrast with XANES, the V in olivine oxybarometry method records fO_2 as a melt crystallises olivine, which may occur prior to significant sulfur degassing (Nicklas *et al.*, 2021).

In this study, fO_2 is measured in lavas from the La Réunion hotspot. Réunion is an island in the south Indian Ocean composed of two major volcanos including the currently active Piton de la Fournaise (Albarede *et al.*, 1997). Lavas from Réunion are remarkable for their uniform He, Nd, Os and Pb isotope signatures, with elevated $^3He/^4He$ and primitive mantle like Pb isotopes suggesting an ancient deep mantle source (Vlastelic *et al.*, 2006; Füre *et al.*, 2011; Peters *et al.*, 2016). Réunion lavas have elevated $^3He/^4He$ ratios (up to ~14 R_A ; Füre *et al.*, 2011) and are little influenced by recycled AOC, so Réunion is a key locality at which to characterise fO_2 .

Methods and Results

The sample set includes seven Réunion olivine-rich lavas, which have been characterised for their bulk rock geochemistry previously (Peters *et al.*, 2016). Laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS) analyses of cores of olivine were performed for 24–36 grains in each sample (Supplementary Information; average olivine trace element concentrations are reported in Table S-1). The V content of the calculated parental melt for each sample was coupled with the V content in olivine to calculate $D_V^{olivine/melt}$ of each sample.

1. Scripps Institution of Oceanography, University of California San Diego, La Jolla, CA, 92093, USA

* Corresponding author (email: rnicklas@ucsd.edu)

Calculated $D_V^{\text{olivine/melt}}$ and temperature (Nisbet *et al.*, 1993) were then used with the empirical equation of Wang *et al.* (2019) to compute the fO_2 for each lava. Parental magma major element and V concentrations and oxygen fugacities are listed in Table 1; calculation details are in the Supplementary Information. Reported uncertainties of fO_2 values are based on the uncertainties of V concentrations in olivine and parental lavas, as temperature and parental magma major element composition have only a second order effect on the reported fO_2 . Vanadium concentrations in olivine vary between 6.0 ± 1.3 and 8.6 ± 2.1 ppm. Also analysed as a proxy for depleted ocean lithosphere olivine was a depleted harzburgite from the Tonga Trench (Day and Brown, 2021), with olivine containing 0.7 ± 0.2 ppm V.

Réunion olivine shows a broader range in V content than other OIB olivines (Fig. 1). The fO_2 of the lavas show wide variation, between $+0.20^{+0.36}_{-0.29}$ and $+1.45^{+0.38}_{-0.31}$ ΔFMQ , and do not correlate with parental magma composition. The fO_2 values range between MORB at $+0.60 \pm 0.15$ (Nicklas *et al.*, 2019) and the global OIB average at $+1.7 \pm 0.8$ ΔFMQ , both measured using the same oxybarometry method (Nicklas *et al.*, 2021). The Réunion lavas also overlap with MORB fO_2 estimates obtained by other oxybarometry methods (Li and Lee, 2004; Berry *et al.*, 2018; Zhang *et al.*, 2018; Wang *et al.*, 2019; Novella *et al.*, 2020). The V in olivine method assumes that the measured olivine is either phenocrystic or antecrystic, and that olivine has not been diffusively reset since crystallisation (see discussion in Supplementary Information). In short, although the studied olivine grains cannot conclusively be shown to be ubiquitously phenocrysts or antecrysts, it is unlikely that samples such as RU0705 (~49 % modal olivine; Peters *et al.*, 2016) would have assimilated such high quantities of unrelated olivine.

Figure 1 Average V concentrations in olivine cores plotted against forsterite number (from Peters *et al.*, 2016), along with olivine from a Tonga Trench forearc peridotite, MORB olivine (Nicklas *et al.*, 2019), and olivine from other OIBs (Nicklas *et al.*, 2021).

The pervasively high $^3\text{He}/^4\text{He}$ measured in olivine separates from these lavas (Füri *et al.*, 2011) strongly suggests that the olivine grains ultimately originate from recent Réunion magmas reinforcing the concept that the calculated fO_2 variations relate to assimilation processes, rather than reflecting mantle source variations.

Table 1 Parental magma compositions; oxides are in wt. %, V is in ppm (~10 % uncertainty). Average ppm V in olivine is also listed. NBO/T – non-bonding oxygens divided by tetrahedrally bonded oxygens in each parental melt. ΔFMQ – oxygen fugacity relative to FMQ buffer. Up – positive uncertainty, down – negative uncertainty. $^{187}\text{Os}/^{188}\text{Os}$ are bulk rock values (Peters *et al.*, 2016). Ol add/subtract – the amount of olivine added or subtracted from the bulk rock composition to calculate the parental magma composition. Positive percentages are added olivine, negative percentages are subtracted olivine. Temperature is calculated using the formulation of Nisbet *et al.* (1993).

Sample Name	RU0702	RU0703	RU0705	RU0707	RU0709	RU0715	RU0719
SiO ₂	46.3	48.6	46.3	49.1	47.2	48.3	46.4
TiO ₂	2.35	2.46	2.42	2.35	2.48	2.13	2.40
Al ₂ O ₃	12.4	12.9	12.3	12.8	12.5	12.4	12.4
FeO	13.1	11.4	13.1	11.2	11.9	12.0	12.8
MnO	0.19	0.17	0.20	0.17	0.18	0.19	0.18
MgO	12.7	11.1	13.2	11.0	11.6	11.7	12.5
CaO	10.0	10.0	9.7	10.1	10.8	10.0	10.2
Na ₂ O	2.03	2.39	2.10	2.37	2.23	2.37	2.15
K ₂ O	0.59	0.66	0.52	0.60	0.72	0.68	0.65
P ₂ O ₅	0.27	0.29	0.28	0.28	0.29	0.29	0.28
V (ppm)	163	139	294	263	315	283	313
NBO/T	1.14	0.97	1.16	0.96	1.07	1.04	1.13
Olivine V (ppm)	6.49	5.96	6.94	6.83	8.55	6.66	6.00
2 s.d.	1.06	1.32	1.00	1.07	2.06	0.79	1.05
ΔFMQ	0.20	0.33	1.03	1.19	0.98	1.24	1.45
Up	0.36	0.47	0.32	0.35	0.51	0.28	0.38
Down	0.29	0.37	0.27	0.29	0.39	0.24	0.31
Temperature (°C)	1255	1221	1264	1220	1233	1233	1250
$^{187}\text{Os}/^{188}\text{Os}$	0.13205	0.13110	0.13194	0.13179	0.13328		0.13273
2 s.e.	0.00007	0.00024	0.00006	0.00028	0.00013		0.00013
Ol add/subtract	-37.7 %	+10.7 %	-50.4 %	+11.3 %	+1.4 %	+2.0 %	+28.1 %

Oxygen Fugacity of Réunion Lavas

The calculated fO_2 of Réunion lavas is highly variable compared with other OIBs (Fig. 2). The more reduced Réunion lavas overlap with the MORB range, unlike other OIBs. The variability of the data is unanticipated given the limited range of isotopic compositions suggesting a homogenous mantle source (Albarede *et al.*, 1997; Vlastelic *et al.*, 2006; Peters *et al.*, 2016). A possibility that can be precluded to explain the variations in fO_2 is variable amounts of recycled AOC in the plume, due to the uniform and unradiogenic Pb and Os isotope signatures of lavas (Vlastelic *et al.*, 2006; Peters *et al.*, 2016). Alternatively, the most oxidised Réunion lava (RU0719 at +1.5 ΔFMQ) may be representative of the plume with the other lavas being reduced by degassing of SO_2 . Sulfur degassing can lead to reduction of a melt (Moussallam *et al.*, 2019), with loss of ~4000 ppm S from Canary and Cape Verde OIBs leading to a ~2.5 log unit fO_2 reduction. Réunion lavas have been estimated to have degassed at most 1510 ppm S, but likely only 100 ppm (Bureau *et al.*, 1998; Collins *et al.*, 2012). If S degassing has a similar redox effect for Réunion lavas as it does in other OIBs, even maximum loss of 1510 ppm S would cause only a 1.05 log unit reduction in fO_2 , smaller than the range of the data even in this most extreme case. For this reason, S degassing can only explain part of the fO_2 variations, assuming that such degassing took place during olivine crystallisation.

Assimilation of material from within the volcanic edifice is a well known phenomenon in OIB. For Réunion, if the lavas were to assimilate altered and oxidised basaltic material, then radiogenic isotope modification from such assimilation would be minimal due to the young age of Piton de la Fournaise. The whole rock dataset of Peters *et al.* (2016) includes concentrations of both ferrous and ferric iron, with $Fe^{+3}/\Sigma Fe$ ratios varying between 0.03 and 0.90, ranging significantly higher than mantle derived basalts at <0.3 (Moussallam *et al.*, 2019), indicating that many of the basalts have suffered oxidative alteration. Starting from the lowest fO_2 Réunion sample (+0.2 ΔFMQ) and using $Fe^{+3}/\Sigma Fe$ ratios of 0.14, 0.21, and 0.90, mixing calculations were performed to determine the amount of assimilation needed

Figure 2 Oxygen fugacity plotted against bulk rock $^{187}Os/^{188}Os$. Literature OIB data from Nicklas *et al.* (2019, 2021). The MORB estimate using V in olivine oxybarometry (Nicklas *et al.*, 2019) is shown as an inverted triangle. Also plotted is the range of MORB oxygen fugacity estimates using other oxybarometry methods as “Literature MORB Range” (Li and Lee 2004; Berry *et al.*, 2018; Zhang *et al.*, 2018; Novella *et al.*, 2020).

Figure 3 Results of mixing calculations between a parental magma composition of RU0702 and an $Fe^{+3}/\Sigma Fe$ of 0.071 and an altered basalt with the same composition but $Fe^{+3}/\Sigma Fe$ values of 0.14, 0.21 and 0.90. Oxygen fugacity was calculated assuming bulk mixing, the calibration of Kress and Carmichael (1991), and a temperature of 1200 °C. Horizontal dashed line represents the maximum measured Réunion lava fO_2 of +1.45 ΔFMQ .

to oxidise RU0702 to +1.5 ΔFMQ (Fig. 3). This modelling shows that 35 % to 5 % edifice assimilation would be required to explain the range in data if assimilated materials had $Fe^{+3}/\Sigma Fe = 0.21$ and 0.90, respectively. Although 5 % assimilation is reasonable for a high MgO magma, 35 % assimilation will spur extensive fractional crystallisation from cooling and could not plausibly produce primitive magma compositions (~12.5 wt. % MgO), suggesting that assimilation of highly oxidised materials is necessary. The presence of such materials in the volcanic edifice indicates that assimilation is a plausible mechanism for explaining the range in measured fO_2 . This model requires magmas to either cross the olivine liquidus in edifice level magma chambers or for altered material to be present at depth. Better constraints on the depth of olivine crystallisation beneath Réunion are needed to fully examine the role of edifice assimilation.

Another process potentially operative in Réunion is inheritance of olivine by lavas. Accumulation of olivine is necessary to explain ‘oceanite’ lavas containing up to 49 % modal olivine, which are not realistic parental melt compositions. Whether this olivine is antecrystic (related to the melt) or xenocrystic (unrelated to parental melts) affects the estimated fO_2 . For example, accumulation of low V olivine xenocrysts would have a profound effect on V in olivine oxybarometry if not properly accounted for. Xenocrystic olivine with low V would artificially drive the calculated fO_2 to higher values if included in the sampled olivine data. For a lava at +0.2 ΔFMQ (RU0702), ~62 % xenocrystic olivine with a V content of ~1 ppm, similar to the Tonga Trench olivine, must be added to sufficiently lower the average olivine content so that the calculated fO_2 is equal to that of RU0719. None of the ~30 vanadium measurements for olivine cores in RU0702 approach 1 ppm, with a minimum of 4.9 ppm (Supplementary Information). Even if xenocrystic olivine were to re-equilibrate with the carrier magma, 62 % is a high proportion of olivine to be accidentally included within any OIB.

Implications for V in Olivine Oxybarometry and fO_2 in OIBs

Vanadium in olivine oxybarometry measures the fO_2 of a melt early in its evolution and is assumed to record fO_2 prior to

secondary processes (Nicklas *et al.*, 2019). Réunion data presented here suggest that this may not always be true. In this setting, assimilation of AOC (~1.3 log unit oxidation with ~5 % assimilation) as well as reduction from SO₂ degassing likely affected the estimated fO_2 . A possible complicating factor with the oxybarometry method is xenocrystic olivine; however, such olivine would likely be from within the Réunion edifice. Such xenocrysts may be common in some OIBs (Wieser *et al.*, 2019) and care is therefore needed in oxybarometry studies to account for them. Olivine populations should be examined for both high Ca concentrations (indicating non-mantle origin) and for unimodal trace element systematics. None of the sampled lavas contain mantle xenocrysts, as mantle olivine has much lower V contents (0.7 ± 0.2 ppm, Fig. 1) than any olivine analysed.

Other OIBs have been measured using V in olivine oxybarometry (Fig. 2) but these data are unlikely to reflect the processes shown here for olivine-rich Réunion oceanites as all those samples show consistent fO_2 regardless of bulk composition, isotopic signatures or location. Given that secondary processes affected the lavas, the uncontaminated fO_2 of Réunion plume is likely to be within the range of contemporary MORB ($\Delta FMQ \approx 0$). This is dissimilar to other OIBs (Fig. 2) and may indicate that the Réunion plume entrains significant MORB source mantle. Isotopic studies, however, have shown a minimal contribution from the MORB source mantle and that Réunion lavas instead come from a primitive source with elevated ³He/⁴He, low ¹⁸²W/¹⁸⁴W and low, highly siderophile element abundances (Vlastelic *et al.*, 2006; Furi *et al.*, 2011; Peters *et al.*, 2016; Rizo *et al.*, 2019). This would suggest that upwelling deep mantle material has fO_2 similar to that of the depleted upper mantle source of MORB despite being isolated from the MORB source mantle for much of Earth's history.

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Additional Information

Supplementary Information accompanies this letter at <https://www.geochemicalperspectivesletters.org/article2205>.

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