

Experimental insights into phase relations and liquid lines of descent of low- and high-Si boninitic magmas from the Izu-Bonin-Mariana fore-arc

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Supplementary figures for the main manuscript

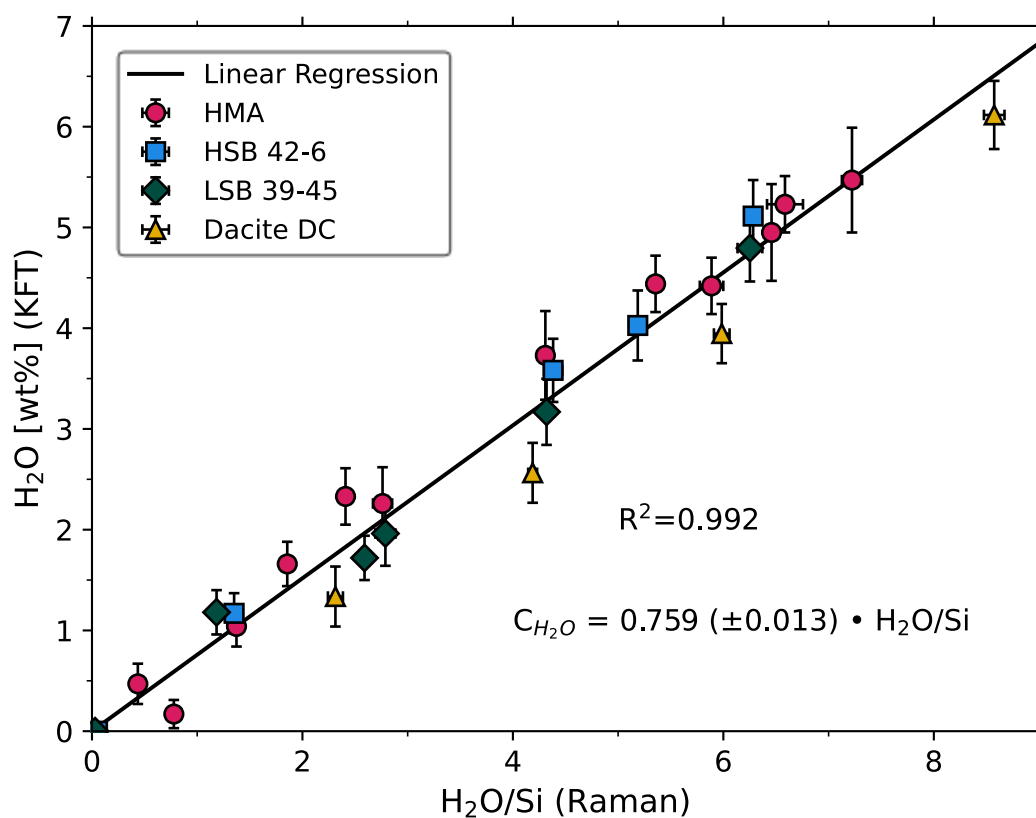


Figure ESM2.1 Linear Raman H₂O calibration for boninitic, high-Mg andesitic and dacitic glasses. Error bars represent the 2 σ standard deviation for H₂O/Si of multiple measurements and the 2 σ uncertainty of the KFT analysis

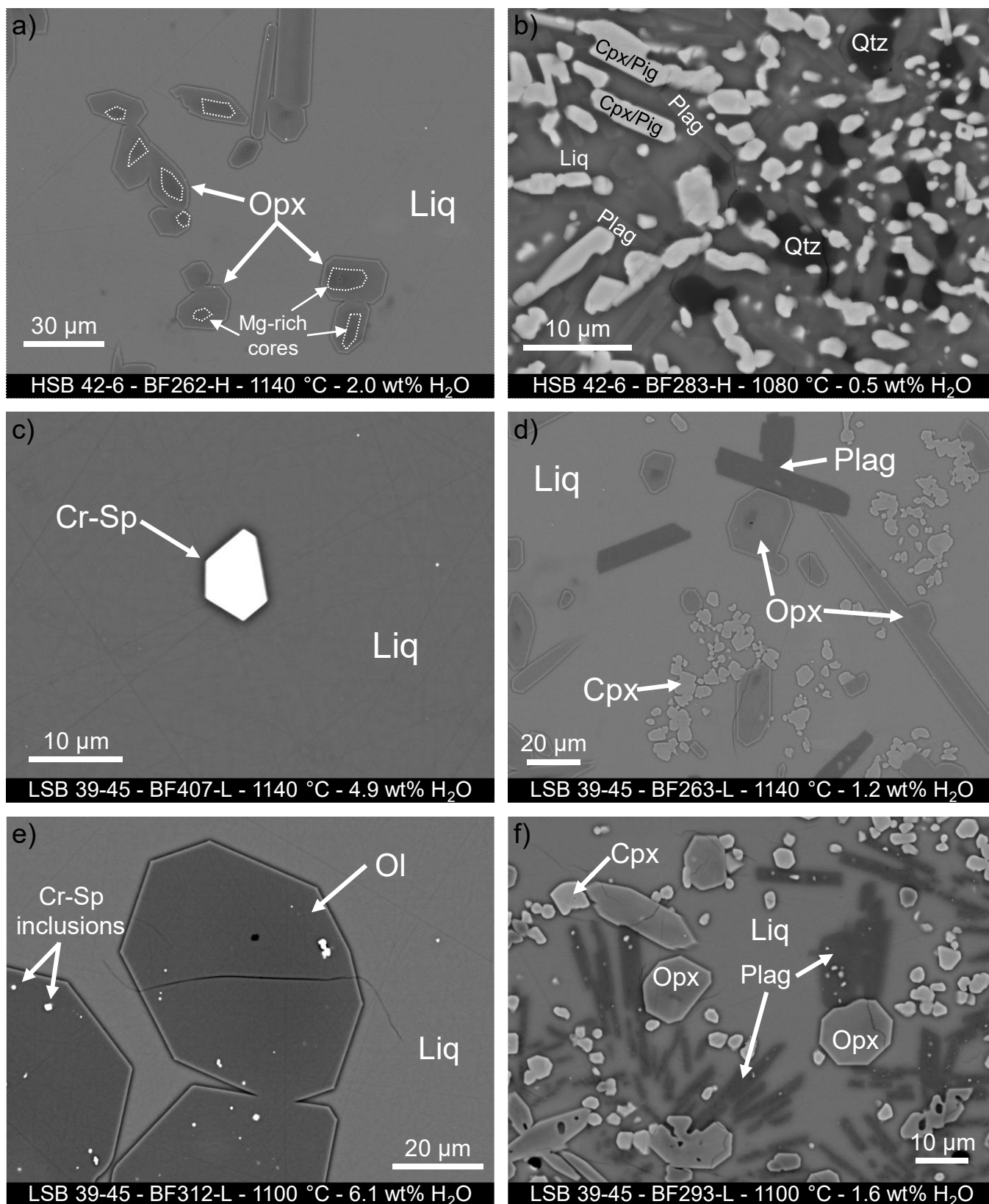


Figure ESM2.2 Supplementary BSE images of experimental run products. Listed H₂O contents correspond to melt H₂O contents. Abbreviations: orthopyroxene (Opx), clinopyroxene (Cpx), olivine (Ol), plagioclase (Plag), Cr-spinel (Cr-Sp), pigeonite (Pig), quartz (Qtz), melt/glass (Liq)

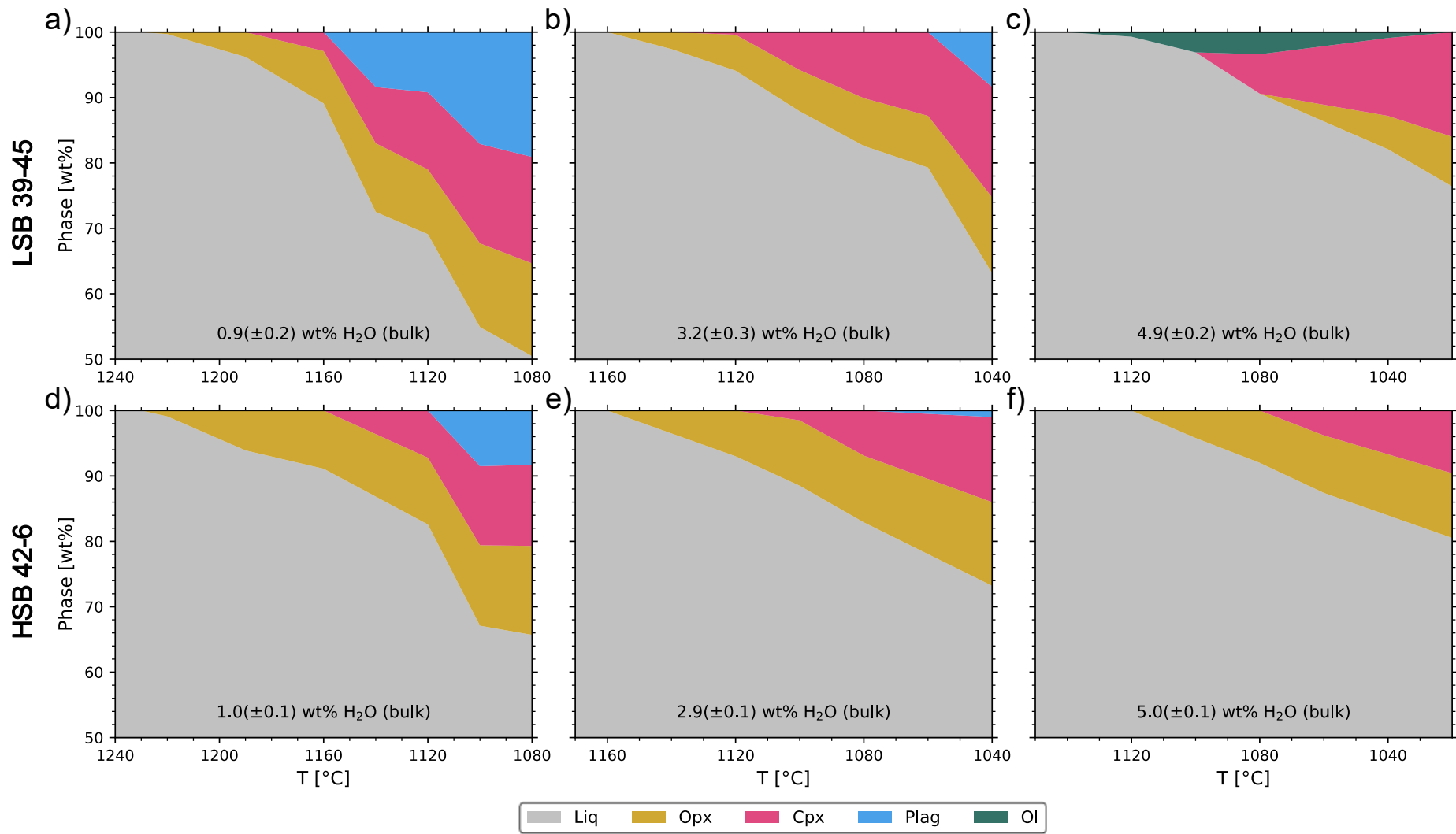


Figure ESM2.3 Evolution of phase proportions (on an anhydrous basis) during EC for different bulk H₂O contents for LSB 39-45 in a)-c) and HSB 42-6 in d)-f) at 200 MPa. Trace amounts of Cr-Sp and an excess fluid phase are not included in the diagrams. The curves for the low H₂O series show slight irregularities (non-smoothness) which can be attributed to temperature deviations during the experimental runs and small differences in bulk H₂O. Abbreviations: melt/glass (Liq), orthopyroxene (Opx), clinopyroxene (Cpx), olivine (Ol), plagioclase (Plag)

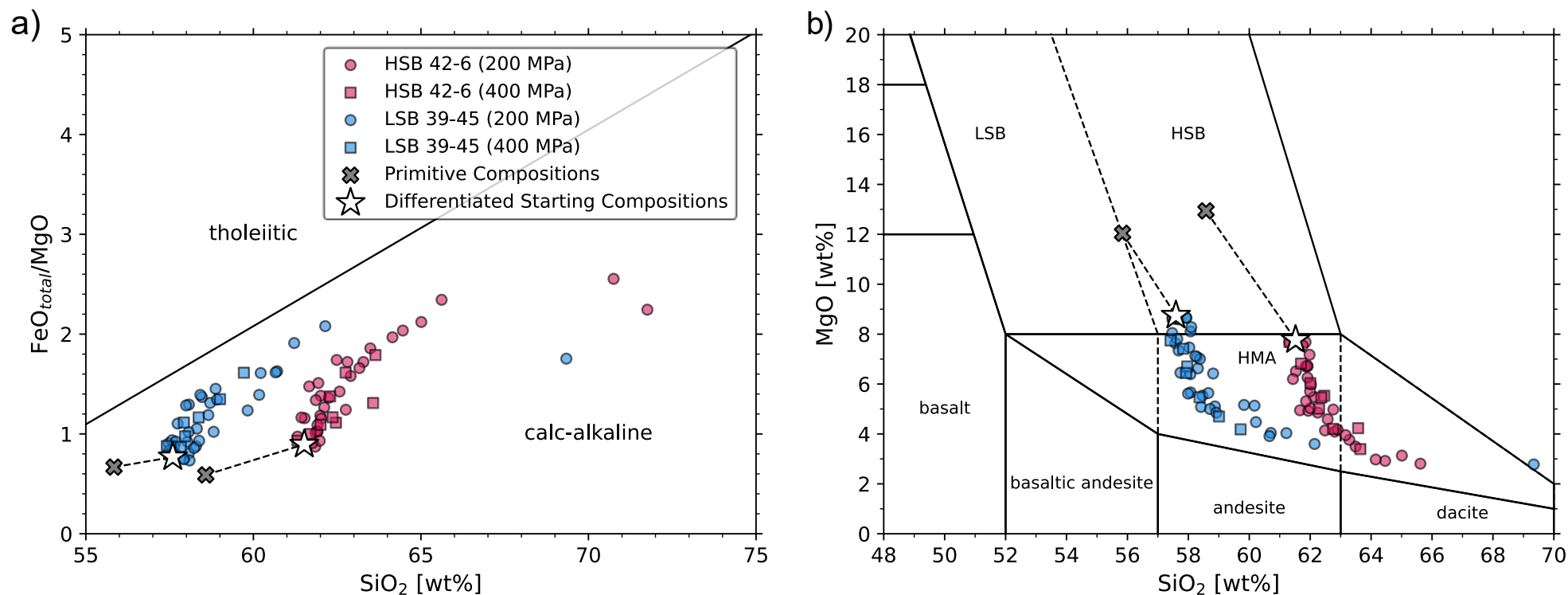


Figure ESM2.4 Experimentally determined differentiation trends of differentiated boninitic magmas (i.e., residual glasses) displayed a) in the $\text{FeO}_{\text{total}}/\text{MgO}$ vs SiO_2 space to discriminate between calc-alkaline and tholeiitic differentiation trends and b) in the MgO vs SiO_2 space. Experimental runs with elevated Fe loss/gain (≥ 10 wt% relative to the starting material) are not shown. Primitive boninitic compositions (i.e., neither fractionation nor crystal accumulation) correspond to the starting compositions used for crystallization modelling by Shervais et al. (2021). Dashed lines connect these primitive compositions with the starting compositions of this study. The solid line separating the calc-alkaline and tholeiitic series in a) is taken from Miyashiro (1974). Boundaries in b) are taken from Pearce and Reagan (2019). Abbreviations: low-Si boninite (LSB), high-Si boninite (HSB), high-Mg andesite (HMA)

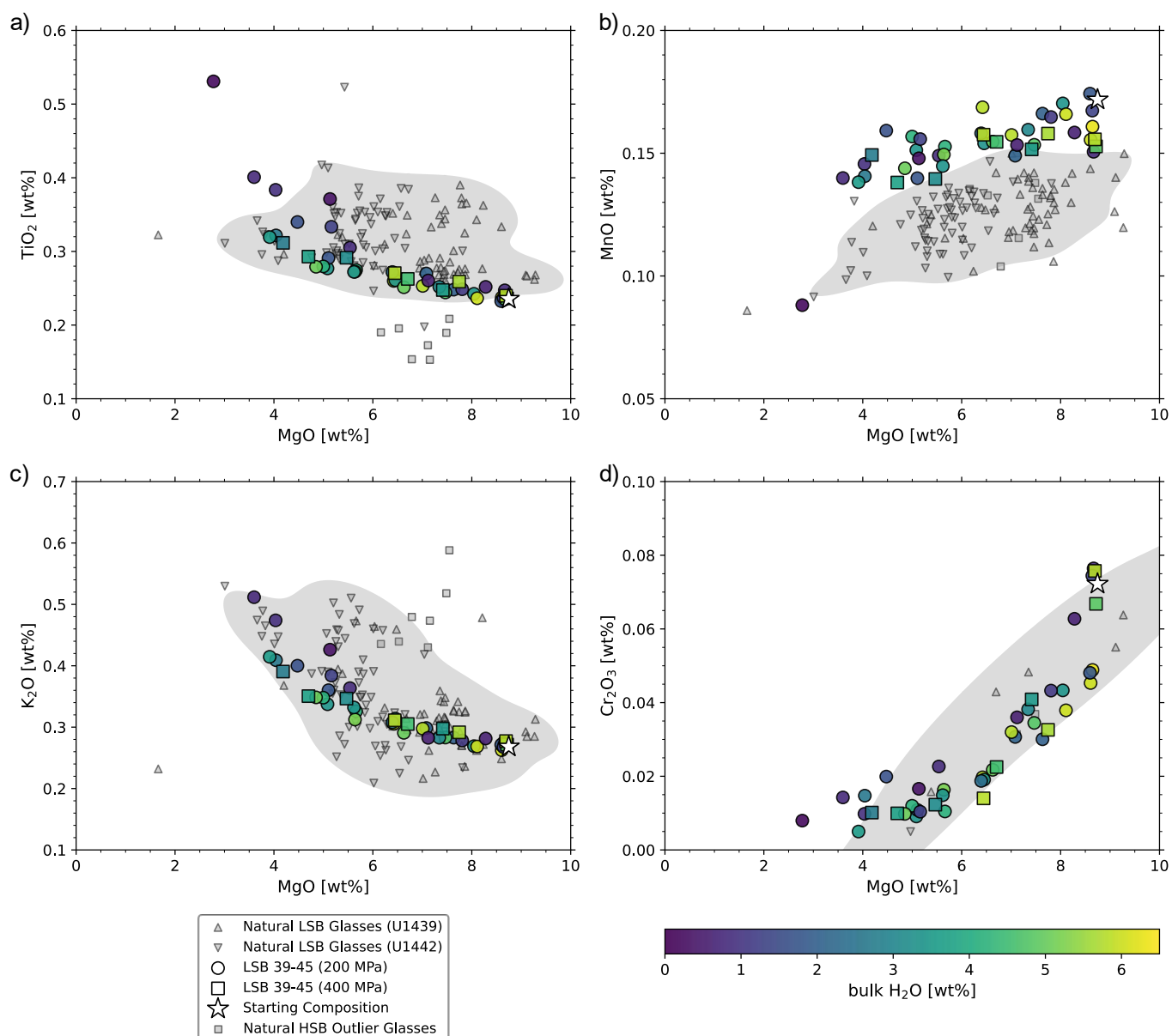


Figure ESM2.5 Additional Harker-type diagrams (major oxides vs. MgO on an anhydrous basis) for residual glasses from the LSB 39-45 crystallization series as well as natural glasses from IBM LSBs (Coulthard et al. 2021a; this study). Experimental runs with elevated Fe loss/gain (≥ 10 wt% relative to the starting material) are not shown. The HSB outlier glasses are also shown for comparison with LSB glasses. The offset for MnO in b) between natural and experimental glasses is explained by too high concentrations of MnO in the starting material for the experiments

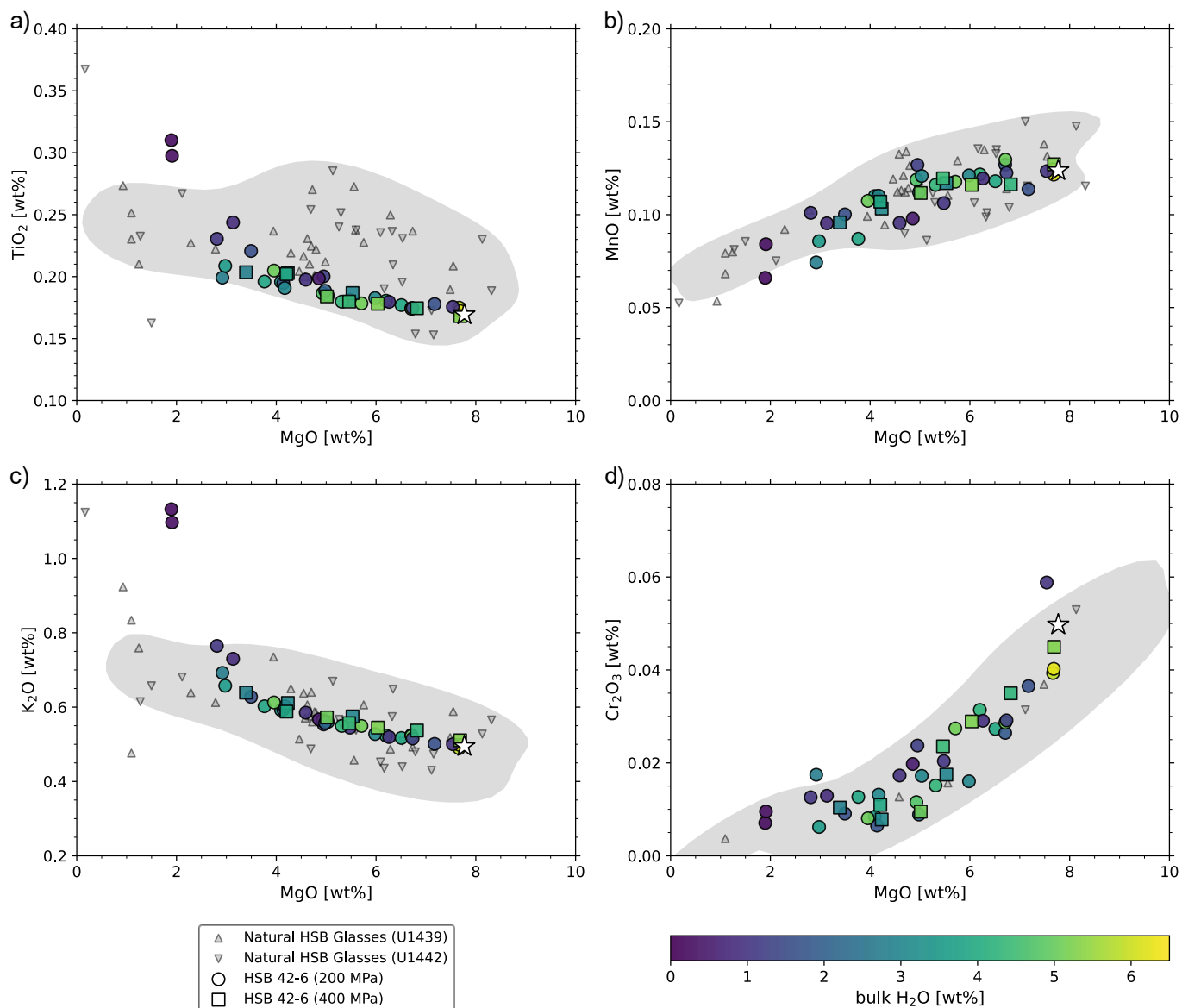


Figure ESM2.6 Additional Harker-type diagrams (major oxides vs. MgO on an anhydrous basis) for residual glasses from the HSB 42-6 crystallization series as well as natural glasses from IBM HSBs (Coulthard et al. 2021a; this study). Experimental runs with elevated Fe loss/gain (≥ 10 wt% relative to the starting material) are not shown

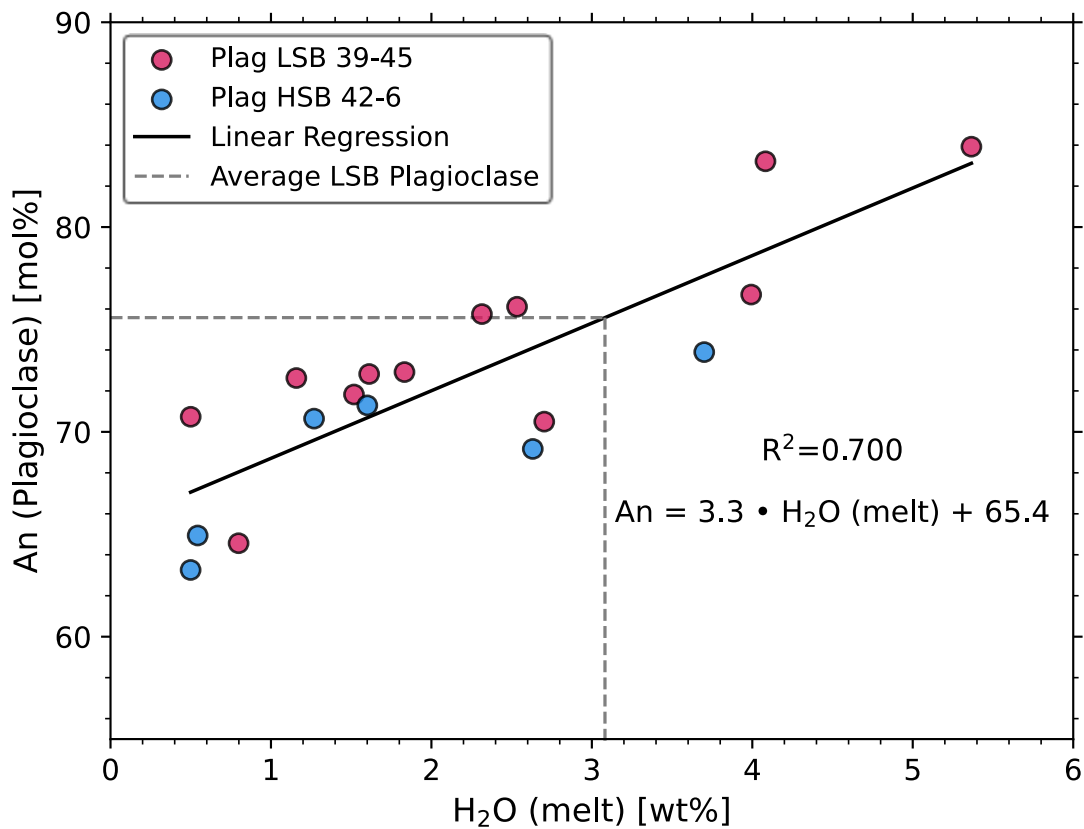


Figure ESM2.7 Anorthite (An) content of experimental Plag as a function of melt H_2O content. The linear regression includes LSB 39-45 and HSB 42-6 experiments. Applying the linear equation to the average An content of natural LSB Plag (An_{76}) results in a melt H_2O content of 3.1 wt%

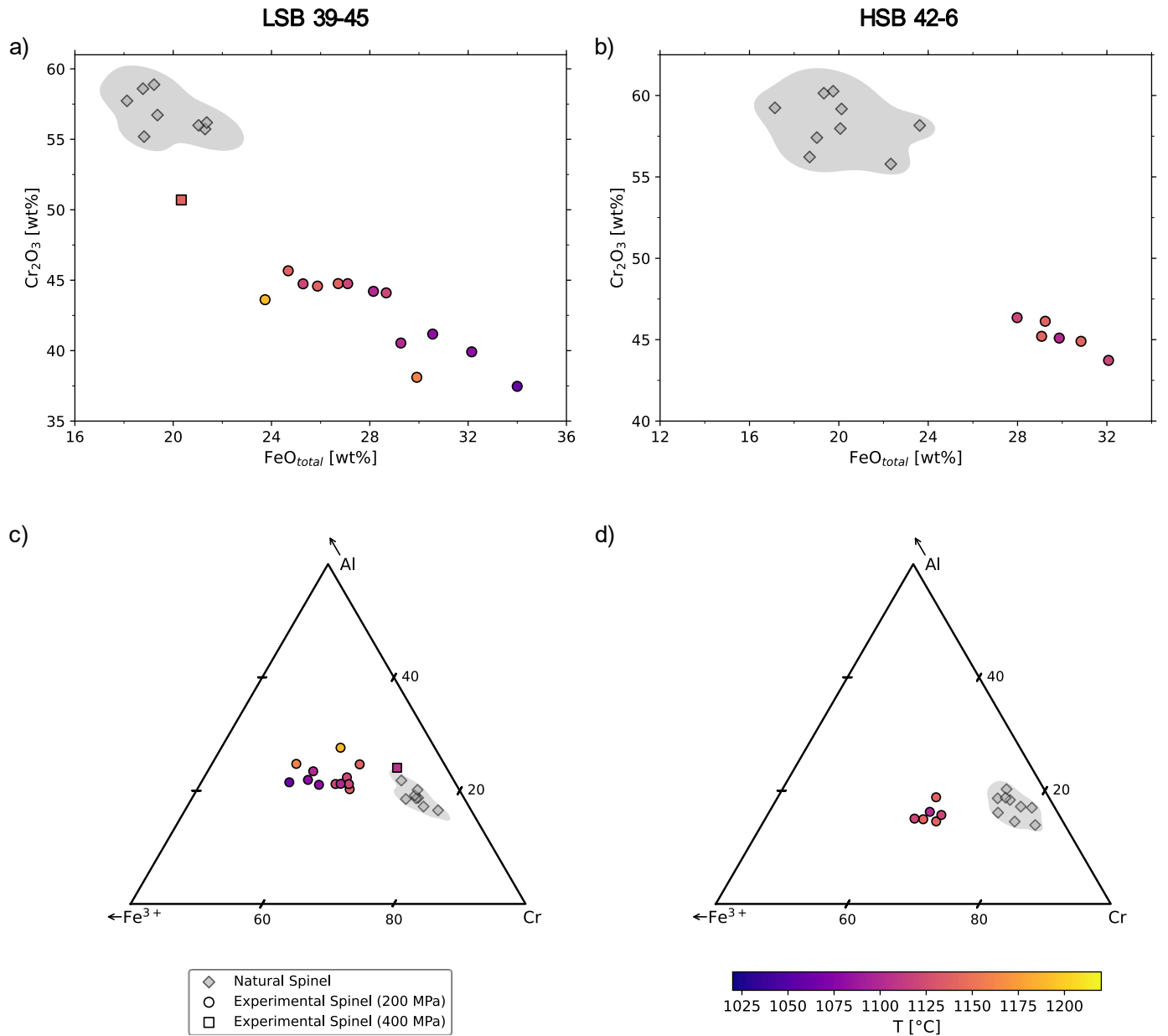


Figure ESM2.8 Comparison between experimental Cr-Sp and Cr-Sp from natural boninites from the IBM fore-arc sampled during IODP Expedition 352. Diagrams in the left column are related to LSBs while diagrams in the right column are related to HSBs. Natural Cr-Sp compositions comprise analyses of Whattam et al. (2020), Scholpp et al. (2022) and analyses within the frame of this study. Compositions shown in a) and b) are not normalized to 100 wt%. Panels in c) and d) display the Cr-rich (≥ 50 mol% Cr) corner of the trivalent cation diagram for spinel (Fe^{3+} calculated based on stoichiometry and charge balance). As discussed in the main manuscript, Fe^{3+} estimates and Cr-Sp compositions in general should be taken with caution

Supplementary text for the main manuscript

Details on Raman spectra processing

As described in the methods section in the main manuscript, the H₂O contents of the experimental and natural boninitic glasses were determined with Raman spectroscopy. The processing and evaluation of the spectra was conducted using the recently published software SilicH₂O (van Gerve and Namur 2023). Additional information and details on the correction of nanolite signals and on the general background correction method are given in the sections below.

Correction of nanolite contamination

Raw spectra of natural glasses were free of peaks of crystalline phases (i.e., no sharp additional peaks) and showed the typical silicate glass bands as well as H₂O bands only. In contrast, some spectra of experimental glasses contained weak but sharp additional peaks at ~1010, ~675 and ~340 cm⁻¹ indicating the presence of nanolites (e.g., Di Genova et al. 2017; Bondar et al. 2025) that potentially formed during the rapid quenching of the samples. In such cases, a pure nanolite spectrum had to be subtracted from affected spectra in order to avoid a disturbance of the Raman bands of the aluminosilicate network (which were later used during H₂O quantification). At first, a pure nanolite spectrum had to be obtained. For that, a nanolite-unaffected spectrum was subtracted from a strongly nanolite-affected spectrum from the same sample. Subsequently, the obtained difference spectrum (i.e., the pure nanolite spectrum; see the black spectrum in Figure ESM2.9) could be used for correction of all other nanolite-affected spectra from this study because the sharp nanolite bands always occurred at the same wavenumber. Finally, this correction step was conducted by subtracting the pure nanolite spectrum from all nanolite-affected spectra using the interference subtraction and interpolation feature of the Raman processing software SilicH₂O (van Gerve and Namur 2023). An example for the nanolite correction is shown in Figure ESM2.9: Subtraction of the nanolite spectrum (black) from the affected spectrum (red) results in the corrected spectrum (green) which is almost identical to a nanolite-free spectrum of the same sample (blue).

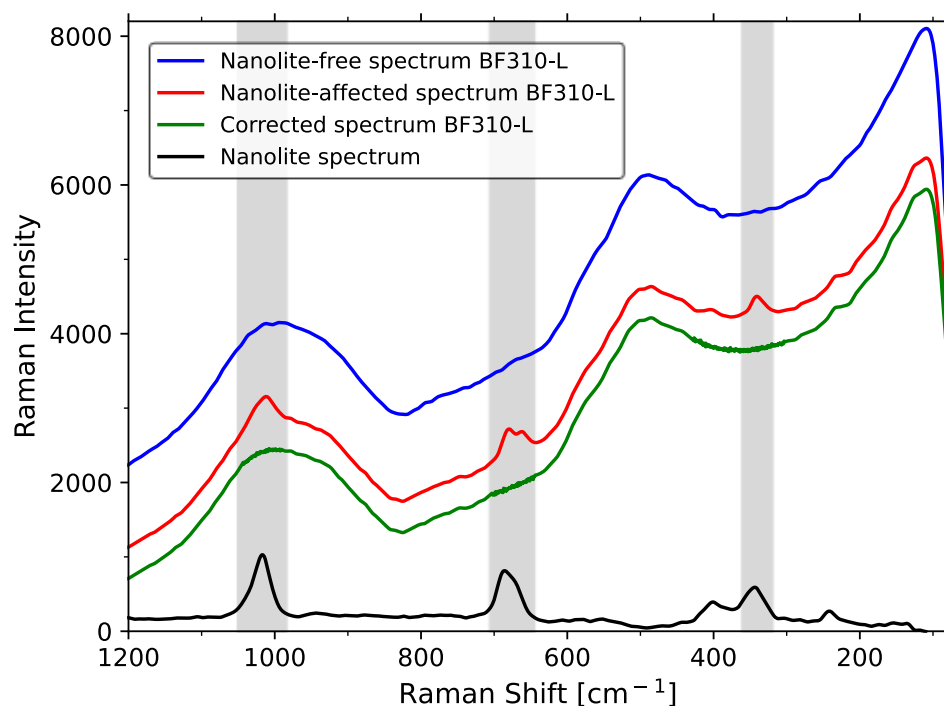


Figure ESM2.9 Exemplary processing of a nanolite-bearing Raman spectrum (before Long-correction) for the hydrous glass of experiment BF310-L (LSB 39-45, 200 MPa, 1100 °C). The spectral range shows the Si region only. Spectra are offset for clarity

Processing and evaluation of Raman spectra

During initial attempts of setting up a general Raman H₂O calibration (i.e., known H₂O vs. H₂O intensity normalized against the intensity of the aluminosilicate network) for glasses with varying chemical composition, we found that applying the Long-correction (Long 1977) to Raman spectra improved the calibration by minimizing the compositional effect. In principle, the Long-correction adjusts Raman intensities for the frequency-dependent Raman scattering and it has been previously applied when using Raman spectra for quantification (e.g., Behrens et al. 2006; Le Losq et al. 2012). Hence, all Raman spectra of our study were processed by applying the Long-correction (Long 1977). Subsequent background correction of Raman spectra was performed using SilicH₂O (van Gerve and Namur 2023) by fitting cubic spline baselines individually for every obtained spectrum by defining five baseline interpolation regions (BIRs). As a result, the peak area of the fundamental OH stretching vibration at ~3550 cm⁻¹ (H₂O) and the peak area of the aluminosilicate network bands between 200 and 1300 cm⁻¹ (Si) were determined. An example for the background correction procedure is shown in Figure ESM2.10. Finally, the H₂O signal was normalized against the Si signal (H₂O/Si) to correct absolute intensities for the analyzed glass volume.

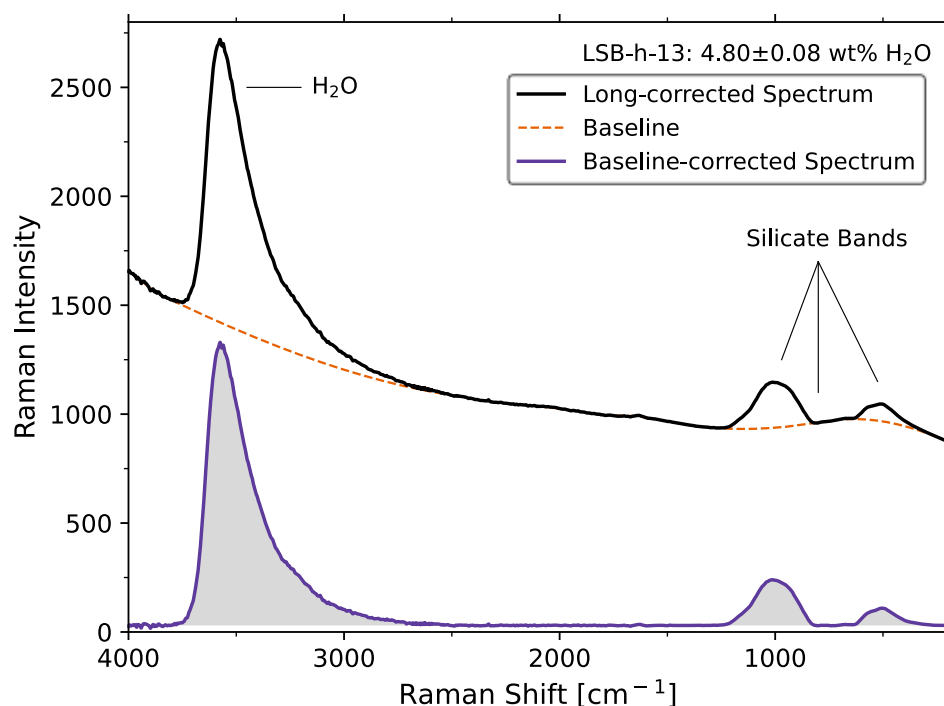


Figure ESM2.10 Exemplary processing of a long-corrected Raman spectrum of the internal H₂O standard glass LSB-h-13. The baseline (dashed) is fitted to the Long-corrected spectrum using the software Silich₂O (van Gerve and Namur 2023) by defining five baseline interpolation regions (BIRs). As a result, the peak areas for the silicate and H₂O bands of the baseline-corrected spectrum can be determined (grey areas). Spectra are offset for clarity

Petrography, compositions and H₂O contents of natural boninites

Petrographic features of the natural boninites are summarized in Table ESM1.7 (Online Resource 1). The boninites all have a vitrophyric and (in most cases) vesicular texture. Phenocryst assemblages of LSBs are typically Ol+Cr-Sp or Opx+Cpx±Ol±Cr-Sp±Plag. HSBs typically have Opx+Cr-Sp±Ol±Cpx. Some samples show tiny acicular and skeletal pyroxenes (or plagioclase) within the matrix glass indicating rapid crystallization due to quenching during eruptions. Olivine, orthopyroxene and clinopyroxene often show normal and oscillatory crystal zonation. Furthermore, olivine typically shows resorption features. These observations all agree with descriptions of samples from IODP Expedition 352 of previous studies (Reagan et al. 2015a; Scholpp 2020; Whattam et al. 2020; Brounce et al. 2021; Coulthard et al. 2021a; Shervais et al. 2021; Scholpp et al. 2022). Exemplary BSE images of natural samples are shown in Figure ESM2.11.

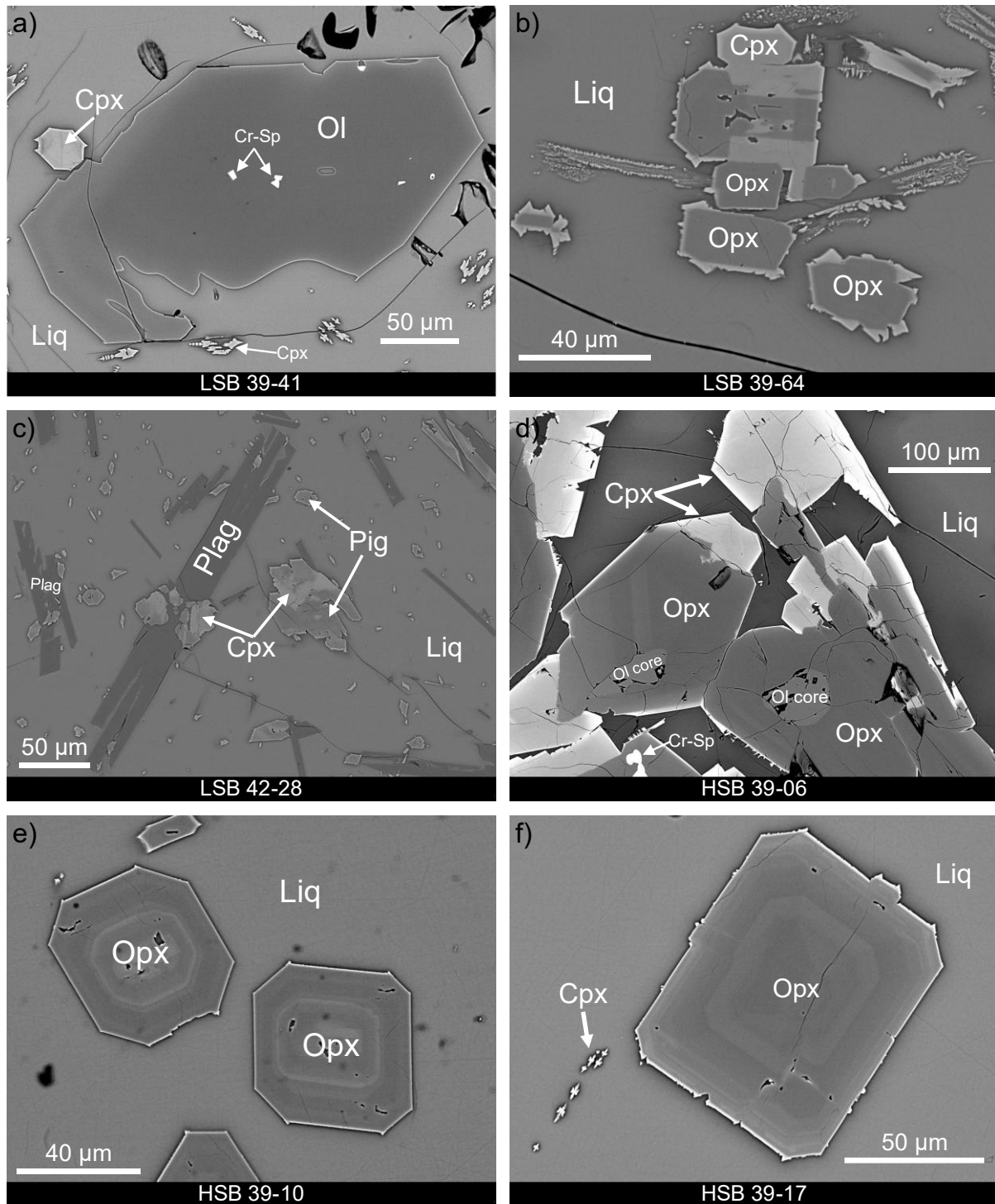


Figure ESM2.11 Exemplary BSE images of natural boninites. Abbreviations: orthopyroxene (Opx), clinopyroxene (Cpx), olivine (Ol), plagioclase (Plag), Cr-spinel (Cr-Sp), pigeonite (Pig), melt/glass (Liq)

Chemical compositions of glasses and minerals as well as glass H₂O contents of the natural boninites are reported in Table ESM1.8 (Online Resource 1) and shown in Figure ESM2.12 (MgO vs. SiO₂). The glasses of the samples classify as differentiated boninites and high-Mg andesites (MgO between 4.6 and 9.3 wt%; SiO₂ between 57.1 and 62.3 wt%) in accordance with boninite glasses from IODP Expedition 352 measured by Coulthard et al. (2021a). Sample 39-06 (HSB) represents an exception because of its anomalous low MgO concentration (~1.1 wt%) and can thus be classified as dacitic. Similarly, Scholpp (2020) and Coulthard et al. (2021a) also reported low MgO concentrations (i.e., <3 wt%) in some glasses and melt inclusions

from IODP Expedition 352 samples. In case of sample 39-06, such low MgO contents could have been the result of extensive Opx and Cpx crystallization whilst suppressing Plag formation (requiring high H₂O melt contents). Alternatively, secondary alteration could have led to MgO depletion but since the glasses appear fresh and show no signs of devitrification, alteration effects can be excluded. H₂O contents of glasses range between 2.1 and 2.9 wt% and generally agree with measurements by Brounce et al. (2021) on the boninites from IODP Expedition 352. Again, sample 39-06 (HSB) represents an exception with an elevated glass H₂O content of 3.6 (±0.2) wt%.

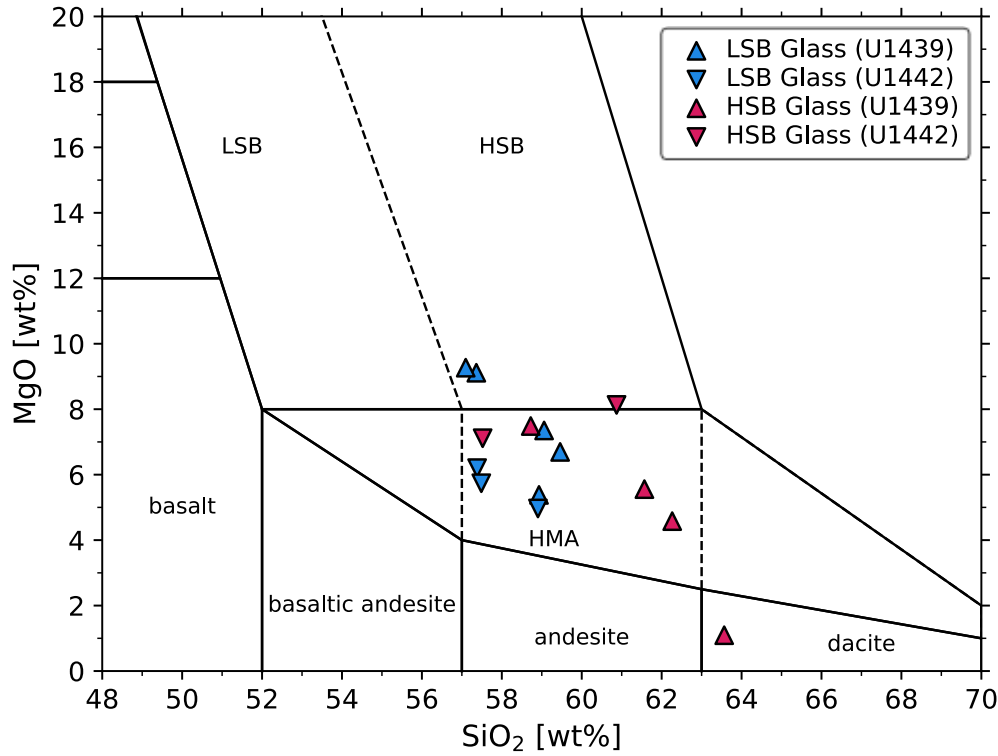


Figure ESM2.12 Compositions of natural boninitic glasses of this study in the MgO vs. SiO₂ space. Boundaries are taken from Pearce and Reagan (2019). Abbreviations: low-Si boninite (LSB), high-Si boninite (HSB), high-Mg andesite (HMA)

As described in the methods section, only mineral rims were analyzed because they are interpreted to be in equilibrium with the interstitial glasses. Olivine varies between Fo₈₅ and Fo₉₀. Low-Ca pyroxenes were classified as orthopyroxenes (N=10) or pigeonite (N=1). Their enstatite content varies between En₇₆ and En₈₆. Clinopyroxenes were classified as augite (Wo₃₇-Wo₄₀) with enstatite contents between En₄₉ and En₅₄. Plagioclase is anorthite-rich (An₇₃-An₇₉) and orthoclase-poor (Or_{0.3}-Or_{0.4}). Cr-Spinel was only analyzed for 3 natural samples. Cr-Spinel is high in Cr# (76-84) and Fe³⁺/Fe_{total} (molar, calculated via stoichiometry) ranges between 0.26 and 0.31. The mineral compositions agree with those reported by Whattam et al. (2020) and Scholpp et al. (2022). K_D values ($K_D = (Fe_{total}/Mg)^{Mineral} / (Fe_{total}/Mg)^{Liq}$) for olivine range between 0.21 and 0.28. K_D values for low-Ca pyroxenes and clinopyroxenes range between 0.24 and 0.28 and between 0.27 and 0.33 respectively. Anomalous sample 39-06 (HSB) represents an exception with low K_D values of 0.06 (Ol), 0.05 (Opx), and 0.09 (Cpx) due to the low MgO content of the residual glass which indicates that these minerals were not in equilibrium with the evolved melt (i.e., the crystals likely represent antecrysts). K_D values ($K_D = (Ca/Na)^{Plag} / (Ca/Na)^{Liq}$) for plagioclase show a narrow range of 1.43-1.53.

Details on the Opx±Liq thermobarometry calculations

For thermobarometric calculations, the “Orthopyroxene P-T” Excel-spreadsheet of K. Putirka (available online at K. Putirka’s website at Fresno State; Putirka 2025) was used. It includes the established Opx(±Liq) thermometers and barometers (Beattie 1993; Putirka 2008) as well as two new additional simplified barometers (“Global” and “Felsic”; Putirka 2008 with modifications, personal communication).

Experimental samples

The initial experimental dataset comprised all low-Ca pyroxene-bearing samples of this study (LSB 39-45 and HSB 42-6; N=80), orthopyroxene-bearing samples from HSB crystallization experiments (Coulthard et al. 2021b; N=7), orthopyroxene-bearing samples from HMA crystallization experiments (Jiang et al. 2025; glass starting material only; N=19) and all orthopyroxene-bearing samples from near-liquidus experiments on a high-Mg andesite (Koch et al. 2025; N=54). Because of this compositional range, the experiments are very suitable to investigate the performance of the thermobarometers within boninitic systems. Another advantage is that melt H₂O concentrations were determined via spectroscopic methods for the majority of the experimental run products.

The experimental data was filtered for Opx-Liq pairs (i.e., average Opx and corresponding average glass compositions) that passed the equilibrium test defined by Putirka (2008): $0.23 \leq K_D = (\text{Fe}_{\text{total}}/\text{Mg})^{\text{Mineral}} / (\text{Fe}_{\text{total}}/\text{Mg})^{\text{Liq}} \leq 0.35$. The final filtered dataset comprised 105 samples. Instead of solving the equations for P and T iteratively, the experimental pressure was used as a known input variable for temperature calculations and vice versa. This helps to reduce the error sources and uncertainties (Wieser et al. 2025) and allows to assess thermometers and barometers independently. However, this procedure represents an ideal situation (with one known and one unknown parameter instead of two unknown parameters) and, thus, will only rarely be the case for natural samples. For some Opx-Liq pairs, the application of barometer equations 29a, 29b and 29c resulted in negative pressures (21 cases). In such cases, the calculated pressure was treated to be 0 MPa. Furthermore, numerical calculation errors occurred in some cases for barometers 29a and 29b (27 cases).

Natural samples

The Opx±Liq thermobarometers have the advantage that they are directly applicable to the majority of the natural boninitic samples (containing Opx as the dominant phenocryst) and that the results can be critically assessed based on the general performance of the various calibrations on experimental boninitic samples. Furthermore, a direct comparison with the results of Opx±Liq thermobarometry on IODP Expedition 352 boninites of Whattam et al. (2020) is possible.

The thermometers and barometers of Putirka (2008) were solved iteratively since no independent constraints on P and T were available. For T calculation, only thermometer 28a (Putirka 2008) was applied because of its good performance on the experimental run products. Here, pressure obtained by the “Global” barometer (Putirka 2008 with modifications, personal communication) was used as an input variable because the more complex barometers (Putirka 2008) are prone to numerical calculation errors (Wieser et al. 2025). Temperature calculations were performed assuming melt H₂O concentrations of 4 and 2 wt%. For pressure calculation, all five barometers were applied using temperatures from Eq. 28a assuming an intermediate melt H₂O content of 3 wt%.

In total, 11 out of 14 investigated natural boninites are orthopyroxene-bearing. Barometer and thermometer calculations were conducted for every individual measuring point on orthopyroxene instead of using the average orthopyroxene composition for one sample directly. By this, individual Opx-melt pairs where numerical calculation occurred or where the Opx-Liq equilibrium conditions were not met could be

excluded. Furthermore, barometer calculations that resulted in negative pressures were set to $P=0$ MPa (but not discarded) similar to the experimental samples. Finally, average pressures and temperatures were calculated based on the filtered and enhanced dataset for every orthopyroxene-bearing sample except for sample 39-06 (HSB) because no Opx-Liq pair passed the test for equilibrium (Putirka 2008).

Volatile saturation pressures

Fresh interstitial glasses of natural boninite rocks from the IBM are highly hydrous with H_2O contents of up to 3.6 wt% (Scholpp 2020; Brounce et al. 2021; this study). In accordance, the application of a Ca-in-olivine hygrometer (Gavrilenko et al. 2016) by Whattam et al. (2020) showed that boninitic magmas had H_2O contents between 2 and 4 wt%. Furthermore, IBM boninites often show degassing textures (i.e., bubbles within interstitial glasses) indicating magmatic water degassed during eruption (i.e., Brounce et al. 2021; Shervais et al. 2021) inferring that measured glass H_2O contents likely represent minimum values.

Reagan et al. (2023) interpreted H_2O contents of boninitic glasses to represent equilibrium H_2O solubilities during subaquatic eruptions of the boninitic magmas at the IBM fore-arc. Therefore, based on the volatile contents of the glasses, they estimated ocean water depths in the range of 2-5 km at the fore-arc of the nascent subduction zone during boninite emplacement (i.e., a water column of 2-5 km, corresponding to a hydrostatic pressure of 20-50 MPa, was required to prevent complete degassing and to maintain the observed high H_2O contents). To further constrain the hydrostatic pressure during eruptions of boninitic magmas and to simultaneously provide minimum pressures for magma storage, volatile saturation pressures were calculated for 72 hydrous glasses reported by Coulthard et al. (2021a) and Brounce et al. (2021) and for the 14 hydrous glasses that were measured within the frame of this study. The saturation pressures were calculated by applying the Magmasat model (Ghiorso and Gualda 2015) via the VESlcal web-app (Iacovino et al. 2021). For simplicity, a magmatic temperature of 1150 °C was assumed and Fe^{3+}/Fe_{total} was set to 0.20 for all boninitic glasses.

LSB and HSB glasses both have an average volatile saturation pressure of 48 MPa (LSBs range between 39 and 74 MPa and HSBs range between 24 and 97 MPa; Figure ESM2.13). Generally, HSB glasses show a wider range in calculated pressures. No significant differences are evident between the two boninite drilling sites U1439 and U1442. In conclusion, calculated pressures imply water depths mainly between 4 and 5.5 km (assuming a hydrostatic pressure of 10 MPa per 1 km) which is in general accordance with estimates by Reagan et al. (2023) but points to slightly greater depths. Similarly, Newman and van der Laan (1992) estimated water depths (i.e., depths of eruption) between 3 and 6 km for boninitic magmas at the IBM fore-arc based on their volatile contents (ODP Leg 125). The calculated saturation pressures can also be seen as minimum pressures for magma storage because these pressures were at least required to dissolve the observed volatile contents in the first place. Hence, the results already indicate deeper storage than estimated by Whattam et al. (2020) via Opx±Liq barometry (20-40 MPa) supporting the overall results on magma storage depths at pressures of around 100-300 MPa established in this study.

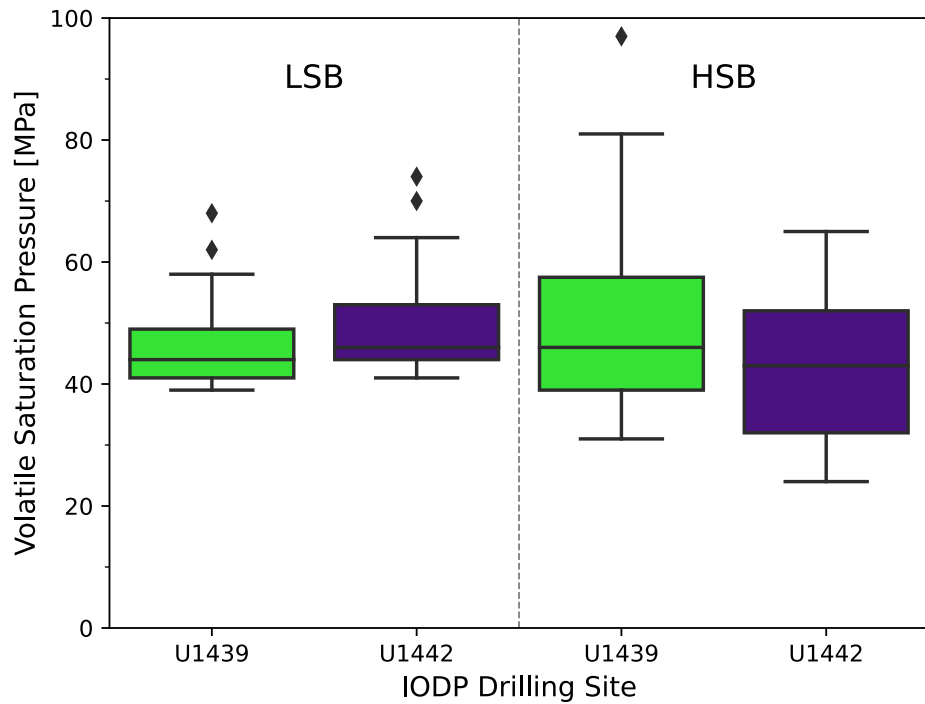


Figure ESM2.13 Boxplot showing the calculated volatile saturation pressures for LSB and HSB magmas from the two IBM drilling sites U1439 (Reagan et al. 2015b) and U1442 (Reagan et al. 2015c)

In general, high H₂O concentrations in boninitic glasses (e.g., >3 wt%) could also be the result of secondary alteration and hydration (Newman and van der Laan 1992) and, as a consequence, calculated volatile saturation pressures could represent overestimations. In general, analyzed boninitic glasses from IODP Expedition 352 were described as fresh and care was taken to avoid altered samples for analyses (Brounce et al. 2021; Coulthard et al. 2021a; Shervais et al. 2021). For instance, the 14 glasses analyzed within this study also appeared fresh and their Raman spectra showed no signs of secondary modification. However, since alteration and secondary gain of H₂O cannot be ultimately excluded, the highest H₂O contents and the highest volatile saturation pressures obtained should be taken with caution.

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