

Appendix 1

Extended methods

A1.1. Preparation of bulk rock samples

Bulk tephra samples were first rinsed with tap water to remove ultra-fine dust and organic materials. Samples were then dried in an oven overnight at ~80 °C and sieved into <250 µm, 250-500 µm, 500-1000 µm, 1000-2000 µm, and >2000 µm size fractions. Aliquots of lapilli clasts were collected from the >2000 µm size fraction, taking care to obtain clasts with the least alteration. Samples were then crushed to pea-sized grains in plastic bags using a 3 lb sledge hammer. The crushed sample was then rinsed with deionized water at least three times, dried in an ~80 °C oven overnight, and, again, clasts having the least alteration were handpicked for powdering.

Samples were powdered using an alumina–ceramic ball mill at Lamont-Doherty Earth Observatory (LDEO). Prior to running the ball mill, and between samples from different volcanoes, the vessel was cleaned with quartz powder, deionized water, and methanol. The vessel was also pre-contaminated by adding a small amount of the least altered sample to the vessel, running the machine, and wiping the vessel clean with a Kimwipe. Each sample was in the ball mill for ~5 minutes, or until a homogeneous powder was achieved. In between samples from the same volcano, the vessel was rinsed with deionized water, dried with a Kimwipe, rinsed with methanol, and dried with a Kimwipe.

Samples were digested using the protocol in Kelley et al. (2003), which we briefly describe here. Redundancy cleaning of 23 mL Teflon beakers was performed with pipetting 0.75 mL of 8N HNO₃ and 0.25 mL of HF into each beaker, sealing the lids, placing on a hotplate at 150 °C for a few hours, and discarding the acid mixture along with 3x rinses of Milli-Q. Powders were weighed

(~0.05 g) and added to the beakers along with a drop of Milli-Q. Then 3 mL of 8N HNO₃ and 1 mL of HF were added, lids were sealed, and beakers were heated overnight on a hot plate at 150 °C. Lids were then removed and samples were dried. Finally, samples were picked-up with 6 mL of 8N HNO₃, sonicated for 20 minutes, returned to the hotplate at 150 C for a few hours, transferred to acid-cleaned 125 mL HDPE bottles, and diluted to 1:2000 powder to solution by mass (~100 g final mass of solution).

A1.2. Major and trace element analyses of bulk rocks

To analyze the major and trace element contents of bulk rock solutions, we followed the method outlined by Wade et al. (2005) that we will briefly discuss. Trace elements were analyzed using a PQ ExCell ICP-MS at LDEO. BHVO-1, W2, BIR-1, and MAS1722 were used as calibration standards. Analyses were corrected by the subtraction of a blank, which was prepared in the same batch of solutions as the unknown and standard samples. Samples were corrected for drift and calibrated using the standard samples. Major elements were analyzed using an Agilent 700-series ICP-ES. Measurements of SiO₂ are not possible by this method because it is volatilized during HF digestion. Therefore, SiO₂ was calculated by difference after accounting for total volatile content measured by loss on ignition. SiO₂ calculated using this method has been shown to give totals that are accurate to within 0.5 wt.% (Wade et al., 2005).

A1.3. Preparation of melt inclusions

The ash size fractions (0.5-2 mm) of cleaned and sieved pyroclastic samples were examined for olivine. Samples containing olivine were put through heavy liquid separation (using LST, ~2.8 g/mL), washed, and dried in an oven at 80 °C for a few hours. Heavy liquid separates were then handpicked for olivine, which was then submerged in mineral oil and examined for melt inclusions. Olivine grains containing fully-enclosed, glassy melt inclusions were mounted to 1”

glass round slides using CrystalBond, ground until melt inclusions were exposed, and polished to 1 μm using diamond suspensions on two sides.

References

- Kelley, K.A., Plank, T., Ludden, J., and Staudigel, H. (2003) Composition of altered oceanic crust at ODP Sites 801 and 1149. *Geochemistry, Geophysics, Geosystems*, 4(6).
- Wade, J.A., Plank, T., Stern, R.J., Tollstrup, D.L., Gill, J.B., O'Leary, J.C., Eiler, J.M., Moore, R.B., Woodhead, J.D., and Trusdell, F. (2005) The May 2003 eruption of Anatahan volcano, Mariana Islands: Geochemical evolution of a silicic island-arc volcano. *Journal of Volcanology and Geothermal Research*, 146(1-3), 139-170.