Magma hybridisation and diffusive exchange recorded in heterogeneous glasses from Soufrière Hills Volcano, Montserrat

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ABSTRACT

Arc volcanoes commonly show evidence of mixing between mafic and silicic magma. Melt inclusions and matrix glasses in andesite erupted from Soufrière Hills Volcano include an anomalously K_2O-rich population which shows close compositional overlap with residual glass from mafic inclusions. We suggest that these glasses represent the effects of physical mixing with mafic magma, either during ascent or by diffusive exchange during the formation of mafic inclusions. Many glasses are enriched only in K_2O, suggesting diffusive contamination by high-K mafic inclusion glass; others are also enriched in TiO_2, suggesting physical mixing of remnant glass. Some mafic inclusion glasses have lost K_2O. The preservation of this K-rich melt component in the andesite suggests short timescales between mixing and ascent. Diffusive timescales are consistent with independent petrological estimates of magma ascent time.

INTRODUCTION

Many arc volcanoes are dominated by interaction between mafic and silicic magmas. Recharge by hotter, more mafic material is frequently cited as an eruption trigger, and

transfer of volatiles may be important in both promoting mixing (e.g. *Eichelberger,* 1980) and advecting heat into, and remobilising, the overlying crystal-rich silicic magma (*Bachmann and Bergantz,* 2006). Magma mingling (incomplete mixing) produces quenched magmatic enclaves, crystal clusters and other clear disequilibrium textures (*Anderson,* 1976; *Bacon,* 1986; *Clynne,* 1999). Recognising the products of magma *hybridisation* (complete mixing) is important for understanding the relative proportions and compositions of the end-member magmas, the likely impact of repeated recharge, and the processes causing mass transfer between magmas.

Recent studies have shown that microlites of anomalously An-rich plagioclase (as well as clinopyroxene and Mg-rich orthopyroxene) in intermediate arc magmas originate in mafic magmatic enclaves or 'mafic inclusions' (*Martel et al.,* 2006; *Humphreys et al.,* 2009a). The microlites are transferred into the andesite either during mixing in the chamber (*Martel et al.* 2006) or by physical break-up and disaggregation of mafic inclusions by shearing during ascent (*Humphreys et al.,* 2009a). This physical transfer of crystals will increase magma viscosity in the conduit and therefore has implications for eruption dynamics, as well as the potential to be used as a tracer of the mafic component. With effective mixing, one might also expect to see a mafic melt component. Melt inclusions and matrix glasses are commonly used to track magma evolution paths and assess magma storage conditions (e.g. *Sisson and Layne,* 1993; *Wallace et al*., 1995; *Blundy et al.,* 2006), so it is vital to determine whether melt derived from the mafic magma is entering the andesite, and if so, in what proportions and with what chemical signature. Quantifying the extent and timescale of interaction between andesitic and mafic magma, which is thought to drive the eruption, would also be invaluable for volcano monitoring and hazard assessment. Here we examine evidence that magma mingling *does* involve transfer of mafic-derived melt, preserved as heterogeneity in plagioclase-hosted melt inclusions and matrix glasses.

GEOLOGICAL BACKGROUND AND SAMPLES STUDIED

The Soufrière Hills Volcano on Montserrat lies in the Lesser Antilles subduction zone and has been active for approximately ~300 ka (*Harford et al.,* 2002). The most recent eruption started in 1995, with a series of pulses of dome growth and explosive activity, interrupted by long pauses when no magma was erupted. Major dome

collapses occurred in December 1997, July 1999, July 2001, July 2003 and May 2006. Currently (September 2009) activity is limited to low-level residual activity, although with continuing significant gas emissions (www.mvo.ms).

Products from the current eruption are porphyritic andesite, with phenocrysts of hornblende, plagioclase, orthopyroxene and Fe-Ti oxides plus rhyolite glass or groundmass. Disequilibrium crystal textures are common, including rare, resorbed quartz phenocrysts, oscillatory zoning in plagioclase and hornblende, reversely zoned orthopyroxene and sieve-textured plagioclase (*Murphy et al.,* 2000). The groundmass contains microlites of plagioclase, orthopyroxene, clinopyroxene and Fe-Ti oxides as well as rhyolitic glass, and may show extensive crystallisation, incipient devitrification and deposits of cristobalite. Macroscopic mafic inclusions have been described in detail (*Murphy et al.,* 1998; 2000) and contain plagioclase, clinopyroxene, orthopyroxene, Fe-Ti oxides and rhyolitic interstitial glass; larger inclusions also crystallise pargasitic amphibole (*Murphy et al.,* 1998). Many of the microlites in the andesite are derived from mafic inclusions, as are crystal clusters, i.e. mafic-derived fragments that can be recognised by texture and mineral compositions (*Humphreys et al.,* 2009a).

RESULTS

We analysed plagioclase-hosted melt inclusions, matrix glass and residual mafic inclusion glass from 23 samples erupted between July 2001 and July 2008 (see supplementary table 1). Most of the samples represent typical andesite. Sample MVO1532d is a heterogeneous mixture of nearly microlite-free rhyolite glass containing euhedral quartz, plagioclase and hornblende, with fine-grained, crystal-rich patches with very little remaining glass. Sample preparation and analytical methods, together with the procedure used to correct for post-entrapment crystallisation (PEC) of melt inclusions, are described in the auxiliary material.

Melt inclusions

Melt inclusions are rhyolitic, with 71-79 wt% SiO_2 (see supplementary data table 2; figures show PEC-corrected values, normalised to 100% anhydrous). Compositions are similar to those reported by *Edmonds et al.* (2001), *Harford et al.* (2003) and *Buckley et al.* (2006). Two populations can be distinguished on the basis of K_2O

contents (figure 1). Most inclusions have 2–3 wt% K_2O, increasing with SiO_2 and Cl contents, but a minority of inclusions has up to 6 wt% K_2O. For the low-K population, Al_2O_3, CaO and Na_2O show a scattered negative relationship with SiO_2. FeO, MgO and TiO_2 subtly increase with SiO_2, and also correlate with each other (e.g. figure 1d). The high-K inclusion population has lower CaO than the low-K glasses, and slightly lower Cl (figure 1). Two inclusions have high TiO_2 but low K_2O (figure 2). High-K glasses were not present in pumiceous samples.

Matrix glasses

Matrix glass was analysed in samples without significant groundmass crystallisation (supplementary table 1). Matrix glasses are rhyolitic, clustering at the SiO_2-rich end of the melt inclusion trends (75-79 wt% SiO_2, figure 1). There are four groups of matrix glasses: (i) high-K, low-Ti, (ii) low-K, low-Ti, (iii) high-K, high-Ti, and (iii) low-K, high-Ti compositions (figure 2). High-K matrix glasses extend to lower CaO contents than low-K glasses (figure 1b). Some glasses have anomalously low MgO. High-K glasses were not present in the pumice samples.

Mafic inclusion glass

Residual mafic inclusion glasses are also rhyolitic (72-78 wt% SiO_2). In Si, Al, Fe and Na composition they are indistinguishable from the melt inclusions. However, they have distinctive high-Ti, high-K compositions (figure 2) and also show low CaO contents, similar to the other high-K glasses. Many of the residual glasses also have low MgO contents. Cl concentrations are variable but tend to be lower than in the melt inclusions (figure 1).

DISCUSSION

In general, the negative correlations of Al_2O_3, CaO and Na_2O with SiO_2 in the melt inclusions indicate decompression crystallisation dominated by plagioclase (e.g. *Buckley et al.,* 2006). The positive correlation between FeO and MgO, and slight increase of both MgO and FeO with SiO_2 suggests minor crystallisation of orthopyroxene or hornblende as observed in the andesite. Ti-Fe variations are consistent with crystallisation of minor Ti-magnetite. The low-K matrix glasses follow mainly the same compositional trends as the low-K melt inclusions but tend towards higher K_2O and higher SiO_2, as K is enriched in the melt during groundmass

crystallisation (*Harford et al.,* 2003). Low-K matrix glasses show decreasing MgO with increasing SiO_2, consistent with groundmass crystallisation of orthopyroxene. The trend of decreasing Cl with increasing K_2O in the matrix glass (figure 1f) indicates degassing of Cl during decompression crystallisation (*Edmonds et al.,* 2001; *Harford et al.,* 2003; *Humphreys et al.,* 2009b).

The low-Ca, low-Mg compositions of the mafic inclusion residual glasses are consistent with significant crystallisation of clinopyroxene in the mafic inclusions. The very high K_2O contents of mafic inclusion glasses are consistent with the lower proportions of amphibole in the mafic inclusions and their lower bulk SiO_2 contents relative to the andesite; their high TiO_2 may be related to the high TiO_2 of the bulk mafic inclusions.

Origin of high-K glass

While the main compositional characteristics of the glass suite are consistent with near-surface processes (see above), the K-rich signature of some glasses is not. The occasional high TiO_2, high-K_2O, low MgO and low CaO contents are also seen in previously reported matrix glasses (*Edmonds et al.*, 2001; 2002; *Harford et al.,* 2003; *Buckley et al.,* 2006; see figure 1). These compositional features are largely shared by the residual mafic inclusion glasses.

The anomalous glass compositions cannot be caused by boundary layer effects during melt inclusion entrapment (*Baker,* 2008) because only slowly diffusing incompatible elements should be enriched in the melt boundary layer, whereas K^+ diffusivities are rapid (*Jambon,* 1983). Similarly, post-entrapment crystallisation of host plagioclase should result in coupled increases of MgO, TiO_2 and K_2O with decreasing CaO, which are not observed, and cannot account for anomalous matrix glass compositions.

K-rich glasses or crystalline products have been ascribed to grain-boundary partial melting of mafic cumulate nodules (*Dungan and Davidson,* 2004; *Heliker,* 1995) or assimilation of biotite-rich cumulates (*Reubi and Blundy,* 2008), with K-rich and host melts mixing during subsequent nodule break-up. However, cumulate nodules are relatively rare in Soufrière Hills andesite and were not observed in any of the samples studied, while the high-K glasses are texturally indistinguishable from 'normal' glasses and their host crystals are not obviously xenocrystic. Finally, K-rich

glasses are also found in mafic inclusions, which are widely agreed to form by rapid quenching against a cooler host (*e.g. Wager and Bailey* 1953*; Yoder* 1973).

Buckley et al. (2006) ascribed the high-K compositions to hornblende breakdown during slow magma ascent and mixing between more- and less-evolved melts. Mass balance between the dissolving hornblende and the observed rims (cpx + opx + plag + oxides) indicated that melts modified by hornblende breakdown should thus be compositionally variable, with high TiO_2 and MgO but low SiO_2 and FeO. The melts should all have high K_2O, Na_2O and Cl (*Buckley et al.,* 2006). Neither of their predicted trends fits with all the observed compositional variations (figure 1). Interstitial melts in hornblende breakdown rims (*Buckley et al.,* 2006) actually show *both* high-K and low-K compositions (figure 1a), not just high-K compositions as expected. We therefore conclude that decompression breakdown of hornblende cannot adequately describe the high-K glasses.

Magma hybridisation and diffusive contamination

We propose that the K-rich melts are derived from, or affected by mixing with intruding mafic magma. The K-rich compositions are similar to those of mafic inclusion residual glass, and incorporation of K-rich melt into the host matrix is consistent with transfer of microlites into the andesite groundmass by disaggregation of mafic inclusions (*Humphreys et al.,* 2009a). However, K_2O-TiO_2 concentrations demonstrate the presence of four distinct glass compositions (see earlier; figure 2): (i) low-K, low-Ti; (ii) low-K, high-Ti; (iii) high-K, high-Ti; and (iv) high-K, low-Ti. This indicates that the mafic inclusion glasses are, for the most part, not being transferred unmodified into the host andesite, and suggests diffusive modification. Breaking open partially crystalline mafic inclusions would allow interaction between host (rhyolite) melt from the andesite and residual rhyolite from the interior of the mafic inclusions. Similarly, complete disaggregation of mafic inclusions would result in physical transfer of K-rich, Ti-rich residual rhyolite, which can be modified by diffusive re-equilibration with the host melt. Diffusion of TiO_2 is much slower than that of K_2O (see later), so residual mafic glass that has lost K_2O by diffusion still retains its high-Ti signature, whereas the high-K host rhyolite cannot gain TiO_2 by diffusion (figure 2). The result is anomalously K-rich (but Ti-poor) host rhyolite melt, and K-poor (but Ti-rich) residual mafic inclusion glasses. This process explains the lack of ubiquitous Ti-enrichment of high-K melt inclusions and matrix glasses

compared with mafic inclusion glass. Once a pocket of K-rich melt is present in the matrix of the andesite, it can be incorporated into melt inclusions by sealing of proto-inclusions during ascent-driven crystallisation (see *Humphreys et al.,* 2008, figure 11).

Timescales of between mixing and eruption

The glass compositions and distribution can give further insight into the physical processes involved in transfer of material. For example, the lack of K-rich glasses in pumiceous samples (see earlier) implies that K-enrichment occurs during slow ascent. Many of the K-rich compositions are matrix glass, with relatively few high-K melt inclusions. This also suggests that transfer occurs primarily during low-pressure ascent and crystallisation, and could be explained by lower shear stresses in the conduit during rapid ascent of less viscous, less crystalline magma (e.g. *Melnik and Sparks* 2005) compared with the highly viscous, strongly crystalline magma that erupts slowly during dome growth.

Elemental diffusivities can be used to assess the timescales of this process (*Sparks et al.* 1977; *Baker*, 1991). Alkali and alkaline earth diffusivities (D) in anhydrous rhyolite are $D_{Na} \sim 1.6 \times 10^{-6}$ cm^2/s, $D_K \sim 8.9 \times 10^{-8}$ cm^2/s, and $D_{Ca} \sim 4.0 \times 10^{-10}$ cm^2/s at 900 °C (*Jambon* 1983). A first-order approximation of timescales (t) can be made, ignoring the effects of possible differences in melt H_2O content, from $x \sim 2\sqrt{(Dt)}$, where x is the diffusion lengthscale, here taken to be 1 cm. Diffusive timescales are 43 hours (Na^+), 32 days (K^+) and 20 years (Ca^{2+}). In other words, alkaline earth diffusion is slow relative to alkalis; diffusion of highly charged ions (e.g. Ti^{4+}) should be even slower than the alkaline earths (*Henderson et al*. 1985). Diffusive contamination of alkalis should therefore be rapid, but modification of other elements would be prohibitively slow (*Baker,* 1991). The K-enrichment of the rhyolite matrix of the host andesite must therefore have occurred ~1 month or less prior to eruption of the magma at the surface, in order to preserve the high-K signature. This is consistent with timescales estimated by preservation of Fe-Ti oxide zoning (*Devine et al.* 2003) and decompression breakdown rims on hornblende (*Rutherford and Devine* 2003). The few high-Ti, low-K glasses, and the wide range of K_2O contents of residual mafic inclusion glasses (figure 2) may reflect a spread to longer timescales, allowing complete or partial re-equilibration of K_2O with the host rhyolite. We also note that the K_2O contents of mafic inclusion and high-K host melts

237 are similar, whereas a diffusion couple should give slightly lower K_2O contents in the
238 host melt. There are two possible explanations for this: (i) the mafic inclusion glasses
239 are already diffused and their observed compositions are not primary, or (ii) the high
240 K_2O in the host rhyolite represents an uphill diffusion 'spike' similar to that observed
241 in experimental diffusion couples (*Bindeman and Davis* 1999; *van der Laan et al.*
242 1994). In either case, this observation reinforces the short timescales between mixing
243 and eruption.

244 The estimated diffusion timescales also suggest that Na_2O contents would
245 quickly be homogenised by diffusion between host andesite and mafic inclusions,
246 while original CaO contents should be preserved. We would also anticipate rapid
247 diffusion of volatiles (e.g. CO_2 and H_2O), particularly in more H_2O-rich melt (*Baker*
248 *et al.* 2005). It is difficult to assess the effects of mixing and diffusion on Ca and Na
249 as these elements are compatible in the crystallising assemblage and therefore
250 strongly affected by fractionation of plagioclase, whereas K is strongly incompatible.
251 However, the K-enriched glasses are slightly depleted in CaO relative to the normal
252 glasses, as is the mafic inclusion residual glass. The different diffusivities of CaO and
253 K_2O are not consistent with diffusive contamination of both elements: timescales long
254 enough for significant Ca diffusion would also eliminate any K_2O signature. The
255 lower CaO contents cannot be produced by crystallisation of quartz, or diffusion
256 gradients around growing plagioclase or pyroxene grains, as discussed earlier. We
257 suggest that the lower CaO might be related to continued crystallisation of
258 plagioclase.
259

260 **CONCLUSIONS**

261 Glass compositional heterogeneity from Soufrière Hills Volcano, Montserrat, is
262 interpreted as the result of mingling between hotter, mafic magma and the host
263 andesite. High-K_2O melt inclusions and matrix glasses in the andesite overlap with
264 the compositions of residual glass from mafic inclusions. However, K_2O and TiO_2
265 contents are decoupled: many high-K melt inclusions do not show high Ti as seen in
266 residual mafic inclusion glasses. This can be explained by diffusive exchange between
267 disaggregated mafic inclusion melt and host matrix melt. The host rhyolite gains K_2O
268 from mafic inclusions, but the original low TiO_2 contents are unchanged. Conversely,
269 high-Ti glasses with normal K_2O contents probably represent residual mafic inclusion
270 glass that has lost K_2O by diffusion. The preservation of such heterogeneity can be

used to estimate the timescales between mingling and magma ascent to the surface. The timescales necessary to preserve K heterogeneity are on the order of a month, which is consistent with magma ascent times estimated from hornblende breakdown and Fe-Ti oxides.

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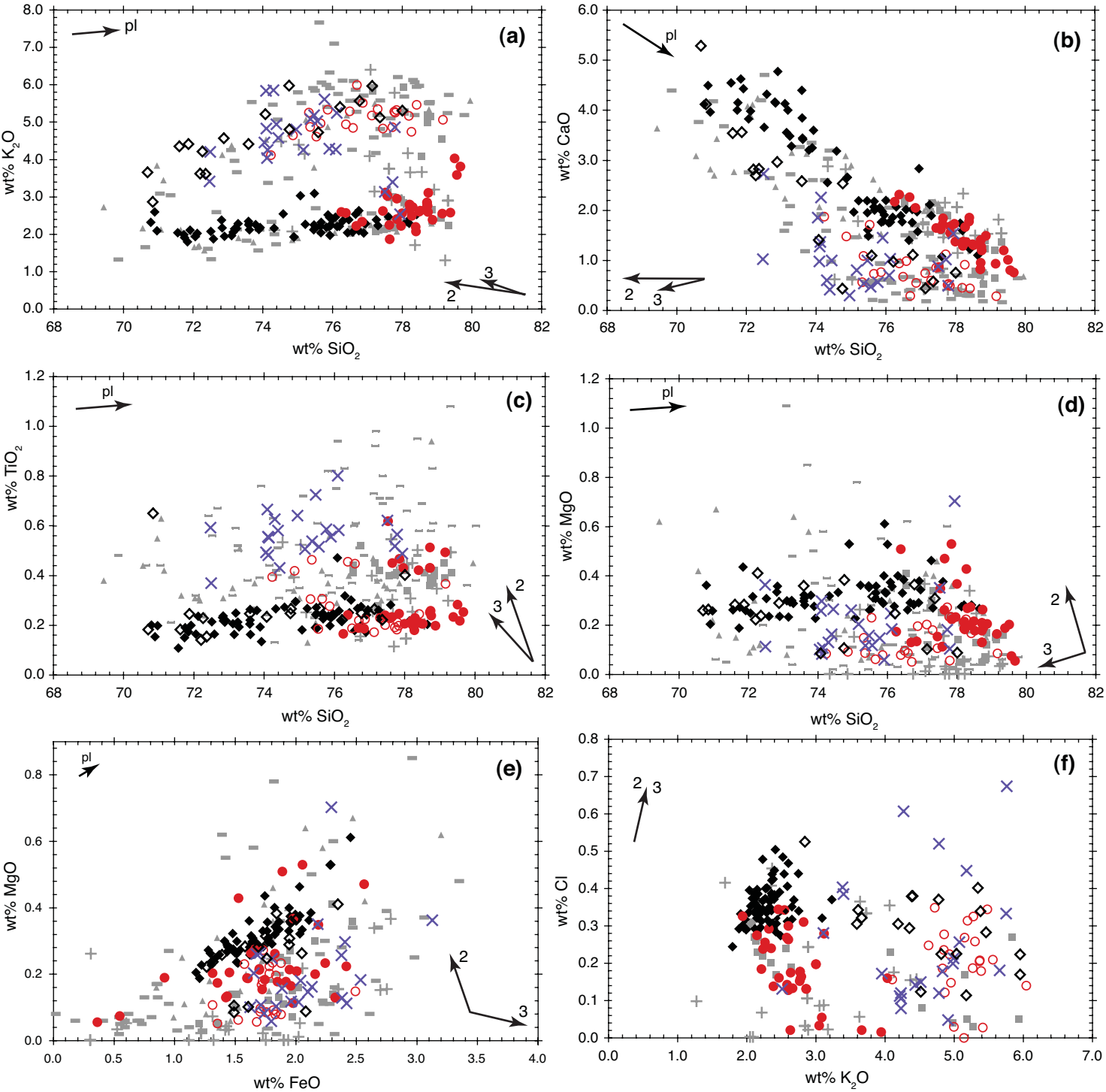
Figures

Figure 1

Compositions of melt inclusions, matrix glasses and mafic inclusion residual glasses. Melt inclusions (diamonds) and matrix glasses (circles) are divided into high-K (open symbols) and low-K (filled symbols) compositions. Crosses represent mafic inclusion glasses. Grey symbols represent previously published glasses from Soufrière Hills (pluses, *Edmonds et al.*, 2001, 2002; squares, *Harford et al.*, 2003; triangles, *Buckley et al.*, 2006). Grey dashes: glasses in hornblende breakdown rims (*Buckley et al.,* 2006). Large arrows indicate the schematic effects of hornblende breakdown reactions (reactions 2 and 3, *Buckley et al.,* 2006), or the effect of 5% post-entrapment crystallisation of plagioclase (pl).

Figure 2

Decoupled compositional variations in K_2O and TiO_2 for all glasses. Thick grey arrows indicate how diffusive contamination of K_2O affects melt compositions. TiO_2 is unaffected because of its very low diffusivity. Symbols as for figure 1.


(a)
pl
2
3
wt% K_2O
wt% SiO_2
(b)
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2
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wt% CaO
wt% SiO_2
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3
wt% TiO_2
wt% SiO_2
(d)
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wt% MgO
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2
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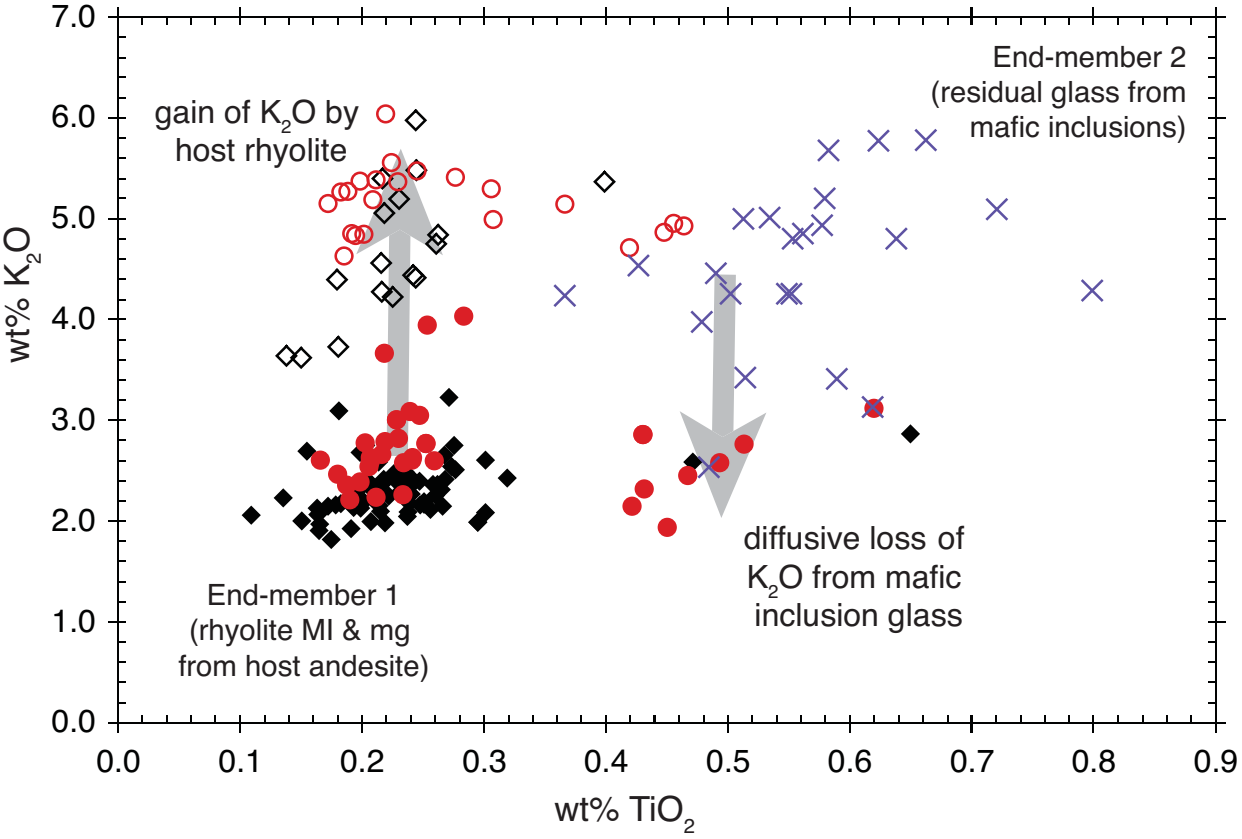
wt% Cl
wt% K_2O

Figure 1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6.0
5.0
4.0
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1.0
0.0
wt% K_2O
0.0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
wt% TiO_2
gain of K_2O by host rhyolite
End-member 2 (residual glass from mafic inclusions)
diffusive loss of K_2O from mafic inclusion glass
End-member 1 (rhyolite MI & mg from host andesite)

Figure 2