1 Revision 1

| 3 4 | Experimental confirmation of high temperature silicate liquid immiscibility in multicomponent ferrobasaltic systems | | | | | | | | | | | | |
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24 Abstract

| 25 | Here we report the results of an experimental study aimed at testing the existence of |
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| 26 | stable, super-liquidus immiscibility between silica-rich and Fe-rich multicomponent |
| 27 | melts at temperatures above 1100 °C. Four pairs of the potentially immiscible |
| 28 | compositions were tested in a one-atmosphere gas-mixing furnace $(Ar/H_2-CO_2$ gas |
| 29 | mixture) at 1150 and 1200 °C and at the oxygen fugacity corresponding to that of the |
| 30 | QFM buffer. Pre-synthesized pairs of the silica-rich and Fe-rich starting compositions |
| 31 | were loaded in Pt wire loops, fused separately at 1300 °C, then brought in contact and |
| 32 | kept at constant experimental temperature for more than 24 hours. Three pairs of |
| 33 | compositions out of four used in this study did not mix. Some temperature-dependent |
| 34 | chemical re-equilibration was observed in the Fe-rich liquid phase but, in the cases of |
| 35 | immiscibility, the two liquids remained compositionally distinct and showed sharp |
| 36 | compositional gradients at contacts. One pair of liquids crystallized some tridymite, |
| 37 | whereas the other compositions were clearly above the liquidus. Overall, the results of |
| 38 | the experiments are in good agreement with the earlier centrifuge study and confirm |
| 39 | the existence of stable, super-liquidus immiscibility in some Fe-rich basaltic-andesitic |
| 40 | compositions at temperatures up to 1200 °C. |

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42 Keywords: experimental petrology; silicate liquid immiscibility; ferrobasaltic;
43 multicomponent silicate melts

44 Introduction

45

46 Liquid immiscibility between ferrobasaltic and rhyolitic melts in a broad 47 compositional range of natural magmas has been considered as a potentially important 48 mechanism of magma differentiation (Charlier et al. 2013; Kamenetsky et al. 2013; 49 Namur et al. 2012; Thompson et al. 2007; Veksler et al. 2007; Veksler and Charlier 50 2015). The existence of silicate liquid immiscibility in natural lavas of 51 ferrobasaltic-ferrodacitic composition has long been recognized (e.g., Roedder 1992 52 and Philpotts 1982), and in recent years traces of immiscibility and liquid-liquid 53 fractionation have been found in plutonic rocks in a number of mafic layered 54 intrusions worldwide, such as immiscible silica- and Fe-rich melt inclusions in apatite 55 and late-stage reactive microstructures (Jakobsen et al. 2005, 2011; Holness et al. 56 2011; Namur et al. 2012; Veksler and Charlier 2015). Stable coexistence of Fe-rich 57 and silica-rich immiscible liquids in equilibrium with typical gabbroic mineral 58 assemblages has been reproduced in numerous experiments (Charlier and Grove 2012; 59 Dixon and Rutherford 1979; Longhi 1990; Philpotts and Doyle 1983) but, with only 60 few exceptions (e.g., Roedder and Weiblen 1970; Krasov and Clocciatti 1979), 61 unmixing of ferrobasaltic melts has been documented only at temperatures close to 62 the liquidus of melts that have low liquidus temperatures, usually below 63 1020-1040 °C. The existence of high-temperature, super-liquidus silicate liquid 64 immiscibility at temperatures up to 1200 °C has been proposed for some 65 multicomponent ferrobasaltic-ferroandesitic compositions on the basis of centrifuge 66 experiments (Veksler et al. 2006, 2007), but the evidence and interpretation of 67 experimental results were challenged by Philpotts (2008) who argued that the

68 products of centrifuge experiments were metastable phases formed during quenching. 69 The results of centrifuge experiments were indeed questionable because phase 70 separation in many runs did not develop beyond the formation of sub-micron 71 emulsions, and interpretation of such emulsions and opalescent glasses has been 72 contentious (e.g., Visser and Koster van Groos 1976, 1977; Roedder 1977; Freestone 73 and Hamilton 1977). It has been pointed out (Veksler et al. 2008a,b, 2010) that the 74 protracted stability of the emulsions in the case of stable immiscibility may be due to 75 low interfacial tension between the immiscible liquids, but researchers who have 76 doubted the existence of stable immiscibility in natural ferrobasaltic compositions 77 above 1040 °C seem to remain unconvinced (Kamenetsky et al. 2013)

78 The debate about the extent of silicate liquid immiscibility to higher temperatures 79 in multicomponent compositions has important implications for natural magmatic 80 systems, and it would be good to resolve it by experimental means despite the kinetic 81 obstacles encountered in previous studies (Veksler et al. 2006, 2007, 2008a,b). It is a 82 common practice in experimental petrology to test thermodynamic equilibrium by 83 conducting direct and reverse experiments. A reverse approach to equilibrium, as 84 opposed to the centrifuge experiments, would be to see whether two pre-synthesized 85 immiscible liquid compositions do or do not mix at long exposure times in a 86 conventional static setup. In the original centrifuge study, Veksler et al. (2007) tried 87 the reverse approach but the results were poorly reproducible and inconclusive. To 88 avoid metastable immiscibility, here we report the results of a new study, in which we 89 used two beads of glass (Fe-rich and silica-rich) touching each other in the experiment 90 and achieved good reproducibility of high-temperature immiscibility at exposure 91 times of 24 h or longer. After making minor modifications to the reverse static setup 92 used by Veksler et al. (2007) we have tested immiscibility for a few compositions used in the original centrifuge study and two new ones, and confirmed the existence
of stable immiscibility at temperatures up to 1200 °C in three cases out of four.

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96 Experimental and analytical methods

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98 Preparation of starting mixtures

99 Compositions and sources for the four pairs of the potentially immiscible 100 compositions (MZ, RY, KC and PZH) are listed in Table 1. All the starting materials 101 were synthesized by fusion in platinum crucibles of carefully weighed and mixed 102 reagent-grade chemicals [SiO2, Al2O3, MgO, TiO2, CaCO3, Na2CO3, K2CO3 and 103 $Ca_3(PO_4)_2$]. To avoid strong oxidation and Fe losses to the crucibles, Fe-free glasses 104 were first prepared by repeated fusions in an electric furnace once at 900 °C, and then 105 twice at 1400 °C. The Fe-free glasses were then crushed to fine powders (grain size 106 less than 3 µm), and mixed and ground with reagent-grade FeO in an agate mortar 107 under acetone. The resulting powders of the glass-FeO mixtures were used as starting 108 materials for the experimental charges

109 Experimental technique

110 The experiments were conducted at GeoForschungsZentrum (GFZ) Potsdam, 111 Germany in a 1-atm vertical tube furnace (Vertical Gero Furnace Alsint tube type) in 112 which oxygen fugacity (f_{02}) was controlled by H₂/Ar-CO₂ gas mixtures. Gas flow was 113 regulated by digital Brooks mass flow controllers and a PtRh₆-PtRh₃₀ (type B) 114 thermocouple was used for temperature measurements. The thermocouple had

| 115 | previously been calibrated against the melting point of gold (1064 °C). |
|-----|--|
| 116 | Yttrium-stabilized zirconia electrolyte (SIRO ₂) manufactured by Ceramic Oxide |
| 117 | Fabricators Pty. Ltd (Victoria, Australia) was used for the f_{O2} measurements inside the |
| 118 | furnace. Uncertainties of the cited $\log f_{O_2}$ values and the temperature do not exceed |
| 119 | ± 0.2 and ± 2 °C, respectively. |

120 Fine powders of the Fe-rich and silica-rich reactant mixtures moistened with a 121 drop of water and some water-soluble organic binder were loaded in separate Pt wire 122 loops about 3 mm in diameter, fused at 1300 °C and f_{O_2} corresponding to that of the 123 QFM buffer, and subsequently quenched in air. Preheating and fusion of the 124 potentially immiscible liquid pairs in the separate wire loops was the key modification 125 which we made to the experimental procedure used by Veksler et al. (2007), who had 126 loaded both starting compositions into the same loop. Pre-fused pairs of the Fe-rich 127 and silica-rich glass beads were then suspended in a new position, in contact with 128 each other and kept at constant experimental temperature (1150 °C or 1200 °C) for at 129 least 24 hours. At the end of the run, samples were quenched in air in a few seconds.

130 Electron microprobe analyses

Run products were mounted in epoxy, ground and polished, and studied by electron microprobe at the GeoForschungsZentrum Potsdam using a JEOL Hyperprobe JXA-8500F in wavelength-dispersive spectrometry (WDS) mode at 15 kV accelerating voltage and 15 nA beam current with spot sizes ranging from 10 to 15 This is a preprint, the final version is subject to change, of the American Mineralogist (MSA) Cite as Authors (Year) Title. American Mineralogist, in press. (DOI will not work until issue is live.) DOI: http://dx.doi.org/10.2138/am-2015-5285

| 135 | μm depending on the properties and compositions of the analyzed materials. Counting |
|-----|--|
| 136 | time for all the elements was set to 20 s on peak and 10 s on background. The |
| 137 | following synthetic and natural standards were used for the calibration: orthoclase (Al |
| 138 | and K), rutile (Ti), wollastonite (Si and Ca), albite and jadeite (Na), apatite (P), |
| 139 | hematite (Fe), diopside and periclase (Mg). The precision for oxide concentrations is |
| 140 | better than 1%. |
| 141 | Compositional mapping of a sample (PZH1200) for calcium was conducted using |
| 142 | a highly sensitive four-segment backscattered electron (BSE) detector. We used the |
| 143 | electron microprobe combined with X-ray, backscattered electron and |
| 144 | cathodoluminescence (CL) maps. By scanning over a specified target area, CL |
| 145 | wavelength and X-ray element distribution maps are acquired in parallel, enabling |
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148 **Results**

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As explained in the introduction, the idea of the reverse experiments was to see if the pre-synthesized pairs of the Fe-rich and silica-rich liquid droplets would mix when held for more than 24 hours in contact with each other at constant temperature and fO_2 close to the QFM buffer. Mixing was supposed to be by chemical diffusion. However, some minor mechanical stirring of the liquids due to wetting and gravity forces was unavoidable, especially at the very beginning of the run. Surface tension merged two droplets into one and the buoyant silica-rich liquid usually floated up and 157 positioned itself on top of the denser and less viscous Fe-rich liquid.

158 In summary, one pair of compositions (labeled MZ in Table 1) completely mixed 159 in both runs at 1150 °C and 1200 °C, which we take to indicate that the run durations 160 were long enough to expect complete mixing by diffusion for all compositions if the 161 two liquids were actually miscible. The other three pairs of the starting compositions 162 did not. In these latter cases, the two liquids remained compositionally distinct and 163 showed sharp chemical gradients at the contacts (Fig. 1). One pair of liquids (KC) 164 crystallized some tridymite (Fig. 1b), whereas the other compositions were clearly 165 above the liquidus. Tridymite crystallization in the RY compositions was also 166 observed in the centrifuge experiments by Veksler et al. (2007) and the results of the 167 direct and reverse experiments appear to be in a good agreement in this respect.

168 Electron microprobe analyses of all the run products are presented in the 169 supplementary data set. The analyses of those samples that did not mix completely 170 showed that liquid compositions, and especially the compositions of the Fe-rich 171 phases, significantly changed in comparison with the starting compositions. Partial 172 dissolution of the silica-rich liquid *Lsi* in the Fe-rich liquid *Lfe* was observed in all the 173 runs in which the liquids did not mix completely. After the runs, unmixed Lfe 174 compositions contained 53-56 wt. % SiO₂ (Table 2) instead of 38-50 wt. % SiO₂ in 175 the starting *Lfe* mixtures (Table 1) and compositional contrast decreased for all the 176 other major components. Mutual solubility of the melts with starting compositions RY 177 and KC appears to increase with temperature, whereas the conjugate Lfe and Lsi melts 178 with the starting mixture PZH were practically unchanged at 1150 °C and 1200 °C 179 (Table 2). Losses of Fe into the Pt wire loops and losses of more volatile components 180 by evaporation appear to be insignificant, except for phosphorus in the P_2O_5 -rich 181 mixtures RY, where P_2O_5 contents decreased by more than 50 % relative to the 182 concentrations in the starting reactant mixtures.

183 Discussion

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185 In our view, the results of this study strongly support the existence of stable liquid 186 immiscibility in some multicomponent, geologically relevant aluminosilicate 187 compositions at temperatures up to 1200 °C. Except the complete mixing between the 188 starting compositions MZ, the results are generally in good agreement with the earlier 189 centrifugation study by Veksler et al. (2007). The coexisting immiscible liquid 190 compositions in this study and in the centrifuge experiments are not exactly the same, 191 but some compositional differences should be expected in view of the very different 192 redox conditions in the direct centrifuge and the reverse static experiments (below the 193 IW and at the FMQ buffers, respectively). Contacts between the immiscible liquids 194 are not razor-sharp menisci but they are somewhat blurred. Diffuse contacts between 195 the immiscible liquids have been also observed in some direct static experiments (e.g., 196 Longhi 1990; Charlier and Grove 2012) and such textures have been interpreted as a 197 result of ineffective phase separation and sluggish chemical diffusion. In the case of 198 reverse "mixing" experiments, which is discussed here, diffuse contacts probably 199 formed at the start of the static runs when the liquid droplets were moved and 200 deformed by the forces of surface tension and gravity. Stirring by convection in the 201 low-viscosity Fe-rich liquid may be another reason for the formation of diffuse 202 contacts. If the liquids were close to chemical equilibrium, gradients of chemical 203 potentials of major components were probably very low across the diffuse contacts. 204 Therefore, sharpening by chemical diffusion of the contacts initially blurred by 205 mechanical stirring may require much a longer time than the exposure times of our 206 experiments. Nevertheless, the same result for the MZ sample, i.e. mixing, at two different temperatures was observed, whereas all the other samples remained unmixed for two temperatures. Therefore, we believe that MZ was not just more thoroughly mechanically mixed (by chance) than the other three samples, but that the observations indicate differences in miscibility for these different compositions.

211 Implications

212 Overall, the results of the experiments are in good agreement with the earlier 213 centrifuge study (Veksler et al. 2007) and strongly support the existence of stable 214 liquid immiscibility in some multicomponent, geologically relevant aluminosilicate, 215 i.e. Fe-rich basaltic-andesitic compositions at temperatures up to 1200 °C. Take the 216 example of the Panzhihua layered gabbros (Wang et al. 2013): based on the chemical 217 composition of the daughter minerals, including plagioclase and clinopyroxene and 218 the melt inclusion, we calculated the liquidus temperature of the immiscible melts in 219 this system. Our results showed that the liquidus temperature is about 1000 °C. Thus, 220 with temperature decreases, the compositions of the conjugate silicate liquids moved 221 further apart along the solvus, consistent with the previous studies (Charlier and 222 Grove 2012; Kamanetsky et al. 2013), but the upper limit (critical point) of the solvus 223 of the tholeiitic system is moderately higher than previously thought. Thus, silicate 224 liquid immiscibility in a broad compositional range of natural magmas could be 225 considered as a potentially important mechanism of magma differentiation.

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237 SIGNATURES

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- 320
- 321
- 322 Figure caption

Figure 1. A series of back-scattered electron images of run products and electron microprobe compositional profiles across the contacts between silica-rich (*Lsi*) and Fe-rich (*Lfe*) glasses (a, RY1150; b, KC1150; c, PZH1200).

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Figure 2. Ca distribution map in the sample PZH1200 showing a steep
compositional gradient at the contact between two immiscible liquids (*Lfe* and *Lsi*).

330

| 331 | Table captions |
|-----|--|
| 332 | Table 1. Compositions of synthetic starting mixtures normalized to 100 wt.%. |
| 333 | |

- **Table 2.** Representative compositions of experimental products based on electron
- 335 microprobe analyses of quenched glasses.



-700 -600 -500 -400 -300 -200 -100 0 100 200 300 400 500 600 μm -800 -700 -600 -500 -400 -300 -200 -100 0 100 200 300 400 500 600 -800 -700 -600 -500 -400 -300 -200 -100 0 100 200 300 400 500 600



| Sample | N | νZ | R | RΥ ⁵ | I | <c th="" ّ<=""><th>P2</th><th>ZH"</th></c> | P2 | ZH" | | | | |
|------------------|-------|-------|-------|-----------------|-------|--|-------|-------|--|--|--|--|
| Phase | Lfe | Lsi | Lfe | Lsi | Lfe | Lsi | Lfe | Lsi | | | | |
| SiO ₂ | 45.66 | 64.56 | 38.40 | 72.50 | 49.86 | 71.82 | 43.38 | 71.31 | | | | |
| TiO ₂ | 5.41 | 1.80 | 3.50 | 0.80 | 4.45 | 1.63 | 1.46 | 0.25 | | | | |
| AI_2O_3 | 9.78 | 13.25 | 8.20 | 15.20 | 2.93 | 8.50 | 9.03 | 14.84 | | | | |
| MgO | 4.15 | 2.32 | 6.60 | 1.00 | 6.79 | 2.02 | 5.59 | 0.65 | | | | |
| FeO | 21.00 | 7.39 | 20.00 | 4.50 | 22.07 | 7.62 | 20.97 | 2.82 | | | | |
| CaO | 10.65 | 6.34 | 14.00 | 1.39 | 9.96 | 2.98 | 16.68 | 5.26 | | | | |
| Na_2O | 2.44 | 2.81 | 1.00 | 2.90 | 1.95 | 2.45 | 1.14 | 2.89 | | | | |
| K ₂ O | 0.48 | 1.37 | 0.30 | 1.70 | 0.96 | 2.98 | 0.31 | 2.31 | | | | |
| P_2O_5 | 0.43 | 0.15 | 8.00 | 0.00 | 1.03 | 0.00 | 1.44 | 0.26 | | | | |
| Total | 100.0 | 100.0 | 100.0 | 100.0 | 100.0 | 100.0 | 100.0 | 100.0 | | | | |

 Table 1. Compositions of synthetic starting mixtures normalized to 100 wt.%.

Notes:Lfe, Fe-rich glass; *Lsi*, silicic glass.

^{a.} Immiscible pairs produced by the centrifuge experiments of Veksler et al. (2007), where the starting composition is based on estimates of the liquid composition at the top of the Lower Zone of the Skaergaard layered intrusion.

^{b.} Natural immiscible liquids in melt inclusions (quenched glasses) in native iron (c.f. Veksler et al. 2007).

^{c.} Natural immiscible liquids (quenched glasses) in mesostasis of basaltic lavas (c.f. Veksler et al. 2007).

^{d.} Natural immiscible liquids in melt inclusions hosted in apatite from the Middle Zone of the Panzhihua layered intrusion,

China (Wang et al. 2013).

| Sample | MZ1150 1150 | MZ1200 | D0 RY1150 0 1150 | | RY1200 | | KC1150 | | KC1200 | | PZH1150 | | PZH1200 | |
|--------------------------------|----------------|--------|--|----------|----------|----------|----------|----------|----------|----------|----------|----------|----------|----------|
| T (°C) | | 1200 | | | 12 | 1200 | | 1150 | | 1200 | | 1150 | | 1200 |
| Phase | L(N=3) | L(N=3) | Lfe(N=3) | Lsi(N=3) | Lfe(N=3) | Lsi(N=3) | Lfe(N=3) | Lsi(N=3) | Lfe(N=3) | Lsi(N=3) | Lfe(N=3) | Lsi(N=2) | Lfe(N=3) | Lsi(N=3) |
| | 54.86 | 55.16 | 52.89 | 71.27 | 54.23 | 68.87 | 53.31 | 70.81 | 56.00 | 71.05 | 54.50 | 69.32 | 54.34 | 71.17 |
| SIO ₂ | (1.29) | (1.31) | (1.17) | (1.28) | (1.25) | (1.13) | (1.20) | (1.25) | (1.36) | (1.26) | (1.27) | (1.16) | (1.26) | (1.27) |
| TO | 3.93 | 4.02 | 2.05 | 0.79 | 2.01 | 1.09 | 4.39 | 1.90 | 4.22 | 1.72 | 1.01 | 0.27 | 1.09 | 0.28 |
| 1102 | (0.20) | (0.20) | (0.10) | (0.12) | (0.10) | (0.05) | (0.12) | (0.10) | (0.21) | (0.09) | (0.05) | (0.01) | (0.05) | (0.01) |
| | 13.76 | 12.92 | 11.37 | 12.81 | 11.92 | 14.69 | 3.52 | 8.21 | 4.45 | 8.48 | 11.37 | 14.34 | 11.26 | 14.72 |
| AI ₂ O ₃ | (1.24) | (1.16) | (1.02) | (1.15) | (1.07) | (1.32) | (0.32) | (0.74) | (0.22) | (0.42) | (0.57) | (0.72) | (0.56) | (0.74) |
| | 8.39 | 8.38 | 16.25 | 5.65 | 14.72 | 4.00 | 17.76 | 7.92 | 16.21 | 7.29 | 12.88 | 3.35 | 13.29 | 1.46 |
| FeO | (0.17) | (0.17) | (0.33) | (0.56) | (0.29) | (0.31) | (0.36) | (0.40) | (1.46) | (0.66) | (1.16) | (0.30) | (1.20) | (0.13) |
| M-0 | 4.18 | 4.22 | 3.72 | 1.01 | 3.57 | 1.76 | 7.45 | 2.22 | 6.13 | 2.21 | 3.50 | 0.73 | 3.71 | 0.64 |
| IVIgU | (0.21) | (0.22) | (0.19) | (0.15) | (0.18) | (0.09) | (0.37) | (0.11) | (0.31) | (0.11) | (0.18) | (0.04) | (0.19) | (0.03) |
| 6-0 | 9.27 | 9.19 | 8.87 | 3.42 | 8.27 | 3.99 | 10.81 | 3.81 | 9.44 | 3.85 | 12.45 | 5.09 | 12.80 | 5.25 |
| CaU | (0.46) | (0.45) | (0.44) | (0.47) | (0.41) | (0.20) | (0.20) | (0.19) | (0.47) | (0.19) | (0.62) | (0.25) | (0.64) | (0.26) |
| K 0 | 0.79 | 0.77 | 0.64 | 1.94 | 0.73 | 1.64 | 0.91 | 3.03 | 1.15 | 3.10 | 1.02 | 3.44 | 0.99 | 3.31 |
| K ₂ O | (0.04) | (0.04) | (0.06) | (0.04) | (0.05) | (0.03) | (0.05) | (0.15) | (0.06) | (0.08) | (0.05) | (0.07) | (0.05) | (0.07) |
| | 3.65 | 4.02 | 1.68 | 2.81 | 1.89 | 3.03 | 1.36 | 2.45 | 1.56 | 2.46 | 1.54 | 2.81 | 1.63 | 3.01 |
| Na₂O | (0.18) | (0.18) | (0.08) | (0.07) | (0.08) | (0.15) | (0.07) | (0.05) | (0.08) | (0.05) | (0.08) | (0.06) | (0.08) | (0.06) |
| | 0.22 | 0.21 | 3.04 | 0.16 | 2.79 | 0.63 | 0.94 | 0.10 | 0.86 | 0.05 | 0.97 | 0.28 | 1.13 | 0.31 |
| P_2O_5 | (0.02) | (0.02) | (0.06) | (0.02) | (0.04) | (0.05) | (0.08) | (0.02) | (0.10) | (0.01) | (0.05) | (0.05) | (0.05) | (0.02) |

Table 2. Representative compositions of experimental products based on electron microprobe analyses of quenched glasses.

| Total | 99.09 | 98.92 | 100.5 | 99.87 | 100.1 | 99.69 | 100.44 | 100.46 | 100.02 | 100.19 | 99.22 | 99.62 | 100.23 | 100.14 |
|-------|-------|-------|-------|-------|-------|-------|--------|--------|--------|--------|-------|-------|--------|--------|
| | | | | | | | | | | | | | | |

Note:

N represents the number of the analyses points of the immiscible phases that are away from the contact zone, and the composition is an average of these analyses. Lfe,

Fe-rich glass; Lsi, silicic glass. Standard deviations are in parentheses