

1 HIGHLIGHTS AND BREAKTHROUGHS

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3 **Stable and metastable silicate liquid immiscibility in**
4 **ferrobasalts**

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11 **Abstract:** The onset of immiscibility in ferrobasaltic systems has been the subject of much
12 research in the last years. The compositional space of the two-liquid field and the maximum
13 temperature of the binodal surface have been investigated experimentally but results from static
14 and centrifuge experiments are controversial. In the article by Hou and Veksler (this issue)
15 entitled “Experimental confirmation of high temperature silicate liquid immiscibility in
16 multicomponent ferrobasaltic systems”, the authors present experimental evidence for
17 immiscibility between silica- and iron-rich melts at 1150-1200 °C, thus significantly higher to
18 previous studies (ca. 1000-1025 °C). These results have important implications for potential
19 large-scale differentiation of magmas by liquid unmixing and for the formation of both Fe-Ti-P-
20 rich melts and rhyolites.

21 **Keywords:** Experimental petrology, binodal, basalt, rhyolite

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23 The formation of immiscible silica-rich and iron-rich melts during cooling of ferrobasalts
24 has been recognized experimentally and in plutonic and volcanic rocks (Philpotts 1982; Charlier
25 and Grove 2012; Veksler and Charlier 2015). However, unmixing of immiscible melts under
26 equilibrium conditions (binodal above the liquidus) or as a metastable process (binodal below the
27 liquidus) has long been discussed. The distinction is essential regarding the ability of liquid
28 immiscibility to produce large-scale differentiation of magmas and contrasting liquid
29 compositions. A metastable process would stay highly localized during eruption of supercooled
30 lavas or late-stage solidification of a crystal mush.

31 The paper by Hou and Veksler (2015) complements the experimental work of Veksler et
32 al. (2007) that has been highly debated. This last study aimed at supporting potential separation
33 of immiscible melts in ferrobasaltic systems at temperature above 1100 °C using high-
34 temperature centrifugation. In these experiments, sub-micrometer globules were produced but
35 clear pools of equilibrium melts separated by a meniscus were nowhere observed. Based on its
36 own experience from static experiments, Philpotts (2008) commented Veksler's results and
37 interpreted the emulsion of immiscible melts as a metastable process during cooling in the
38 centrifuge of the homogeneous melt that reached a sub-liquidus binodal surface. Static
39 crystallization experiments in 1-atm vertical furnace and rapid quench have usually been used to
40 constrain the binodal surface of ferrobasalt to a maximum temperature of 1025 °C (Dixon and
41 Rutherford 1979; Philpotts 1979; Philpotts and Doyle 1983; Charlier and Grove 2012). In these
42 experimental studies, sharp two-liquid interfaces are usually observed. However, Charlier and
43 Grove (2012) and Longhi (1990) report some static experiments with diffuse contacts between
44 the two liquids, illustrating the difficulty of the equilibrium immiscible melts to separate from
45 each other. This is a consequence of very low interfacial tension between contrasting iron- and

46 silica-rich melts with easy nucleation of immiscible liquid droplets and very slow coarsening
47 (Veksler et al. 2010).

48 In a detailed reply, Veksler et al. (2008) further explained and reaffirmed the evidence for
49 high-temperature liquid immiscibility. The paper by Hou and Veksler (2015) is a new effort to
50 prove that silicate liquid immiscibility can occur at higher temperature. The approach is based on
51 mixing rather than unmixing experiments. Pairs of potentially immiscible compositions were first
52 fused separately in 1-atmosphere vertical tube furnace at QFM buffer. Fused beads were then
53 suspended in contact with each other and run at 1150 or 1200 °C. Compositional reequilibration
54 of the paired melts is observed but a compositional gap exists between an iron-rich basaltic
55 andesites (53-56 wt% SiO₂ and 14.7-17.7 wt% FeO_{tot}) and rhyolitic melt (69-71 wt% SiO₂ and
56 4.0-7.9 wt% FeO_{tot}). Interestingly, this compositional range do not include classical ferrobaltic
57 composition at maximum iron-enrichment (ca. 45-50 wt% SiO₂ and 14-19 wt% FeO_{tot}; Charlier
58 et al. (2013)).

59 Because the two-liquid field broadens with decreasing equilibration temperature, it is
60 expected that immiscible melts will have less-contrasting compositions at high-temperature. It is
61 thus interesting to observe that Hou and Veksler (2015) obtained iron-rich immiscible melts with
62 53-56 wt% SiO₂ and 14.7-17.7 wt% FeO_{tot} above 1150 °C, while they range from 30-50 wt%
63 SiO₂ and 18-32 wt% FeO_{tot} below 1020 °C (Charlier and Grove 2012). This means that with
64 increasing temperature, the binodal surface moves from ferrobalt-rhyolite compositions to
65 basaltic andesite-rhyolite end-members. Consequently, although the experiments of Hou and
66 Veksler (2015) convincingly support the existence of a stable super-liquidus two-liquid field in
67 ferrobaltic systems above 1150 °C, it would be important to identify whether silicate melts
68 produced along tholeiitic liquid lines of descent can reach SiO₂ content above 52 wt% at such
69 high temperatures. Indeed, silica-enrichment above ca. 50 wt% during tholeiitic evolution is

70 produced by crystallization of Fe-Ti oxides that appear on the liquidus below 1100 °C (Juster et
71 al. 1989; Snyder et al. 1993; Toplis and Carroll 1995). Possibly, the results of Hou and Veksler
72 (2015) have implications for the evolution of tholeiitic andesite, in which immiscible globules
73 have also been reported (Philpotts 1982). The Upper Zone of the Bushveld complex is an
74 example of plutonic evolution of magma with andesitic composition (VanTongeren et al. 2010)
75 for which the development of immiscibility and its role for magma differentiation are highly
76 controversial (VanTongeren and Mathez 2012; Cawthorn 2013).

77 The study by Hou and Veksler (2015) is an important advance in the understanding of
78 silicate liquid immiscibility. Further experimental studies must pursue in testing the existence of
79 a stable or metastable two-liquid field by running unmixing and mixing experiments for different
80 magma compositions in a range of magmatic temperature. Further progress should also come
81 from in situ experimental methods and the development of micro-analytical facilities.

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