Interfacial tension between immiscible liquids in the system K$_2$O-FeO-Fe$_2$O$_3$-Al$_2$O$_3$-SiO$_2$ and implications for the kinetics of silicate melt unmixing

**ILYA V. VEKSLER,**1,2,* JOHANNES KÄHN,3 GERHARD FRANZ,2 AND DONALD B. DINGWELL3

1GFZ German Research Centre for Geosciences, Section 3.3, Telegrafenberg D-14473 Potsdam, Germany
2Technical University Berlin, Department of Mineralogy and Petrology, Ackerstrasse, 71-76, D-13355 Berlin, Germany
3Earth and Environmental Sciences, University of Munich, Theresienstrasse 41/III, 80333 Munich, Germany

**ABSTRACT**

Interfacial tension between immiscible liquids is an important thermodynamic parameter of silicate melt unmixing and a property that determines the kinetics of phase separation. In this study, we present experimental measurements of interfacial tension between immiscible Fe-rich and silica-rich melts in the system K$_2$O-FeO-Fe$_2$O$_3$-Al$_2$O$_3$-SiO$_2$. We have also measured densities and surface tensions of the individual immiscible liquid phases. The measurements were carried out in air at 1500–1550 °C by the maximum detachment force method employing vertical cylinder geometry and using a gravimetric balance system. We have chosen the most oxidized and contrasting liquid compositions containing 73 and 17 wt% SiO$_2$ and 14 and 80 wt% FeO, respectively, that have been shown to coexist in air at and above 1465 °C. Interfacial tension between the synthetic immiscible liquids decreases with increasing temperature from 16.4 ± 3.1 mN/m at 1500 °C to 7.8 ± 1.1 mN/m at 1550 °C. Interfacial tension between natural, less compositionally contrasting ferrobasaltic and rhyolitic melts should be even lower by a factor of 2 or 3. Very low interfacial tension implies easy nucleation of immiscible liquid droplets and very slow coarsening of resulting silicate emulsions.

**Keywords:** Immiscibility, interface, tensiometry, nucleation

**INTRODUCTION**

Immiscibility in silicate and other glass-forming liquids has been extensively studied in igneous petrology (Roedder 1978; Philpotts 1982) as one factor to explain the diversity of magmatic rocks, and because of important applications to glass technology, e.g., in manufacturing of glass ceramics (Shelby 2005). Decades of experimental and theoretical research resulted in comprehensive thermodynamic models of silicate liquid immiscibility (see review by Hudon and Baker 2002), and good understanding of physical and chemical driving forces behind unmixing and liquid-liquid element partitioning (Schmidt et al. 2006; Veksler et al. 2006). On the other hand, the kinetics of liquid immiscibility has not received enough attention. To the best of our knowledge, only one study specifically dealt with the kinetics of immiscibility in natural basaltic magma (Martin and Kushiro 1991). Our recent work (Veksler et al. 2008) revealed sluggish nucleation and phase separation kinetics during unmixing of synthetic Fe-bearing aluminosilicate melts. Interfacial energy is a key parameter determining the kinetics of nucleation and growth of immiscible liquid droplets. Theoretical considerations (Hammel 1967) and the morphology of quenched immiscible glasses (Veksler et al. 2008) have implied a very low interfacial tension between immiscible silicate liquids but, as far as we know, interfacial tension has never been measured directly by experimental methods for any pair of immiscible silicate melts.

Here we report results of experimental measurements of interfacial tension between Fe-rich and silica-rich immiscible melts in the system K$_2$O-FeO-Fe$_2$O$_3$-Al$_2$O$_3$-SiO$_2$. We measured interfacial tension in air between the most contrasting, oxidized liquid compositions. Therefore, the values are likely to represent the upper limit of interfacial tension between immiscible Fe-rich and silica-rich aluminosilicate melts. In the discussion, we examine relationships between interfacial tension and surface tensions of the coexisting liquids and touch upon some implications for immiscibility in natural basaltic magmas.

**EXPERIMENTAL METHODS**

Surface and interfacial tension was measured by the method of maximum detachment force of a vertical cylinder wetted to the interface. The force was measured using a gravimetric balance system. This version of the detachment technique is a classical method that has been broadly used in liquid-gas and liquid-liquid tensiometry (Walker and Mullins 1981; Rusanov and Prokhorov 1996), and its practical and theoretical aspects have been investigated and described in great detail (e.g., Padday et al. 1975). The method and the equipment that we used were successfully tested in our recent study of interfacial tension between immiscible melts in alkaline-earth-boron oxide binaries MgO-B$_2$O$_3$, CaO-B$_2$O$_3$, and BaO-B$_2$O$_3$ (Veksler et al. 2010).

**Starting materials**

The detachment method requires relatively large volumes of liquids, at least a few cubic centimeters in our case. A sufficiently large area of the interface is needed to minimize effects from crucible walls (Rusanov and Prokhorov 1996). To facilitate the formation of homogeneous, perfectly separated immiscible liquid layers, pre-synthesized conjugate liquid compositions are loaded separately into a vessel used for the measurements, one composition on top of the other in the order of decreasing density.

The compositions of the two synthetic starting liquids used in this work (Table 1) are based on electron microprobe analyses of the conjugate Fe-rich ($L_{Fe}$) and