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In situ Viscometry of Primitive Lunar Magmas at High Pressure and High Temperature

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Understanding the dynamics of the magmatic evolution of the interior of the Moon requires accurate knowledge of the viscosity ($\eta$) of lunar magmas at high pressure ($P$) and high temperature ($T$) conditions. Although the viscosities of terrestrial magmas are relatively well-documented, and their relation to magma composition well-studied, the viscosities of lunar titanio-silicate melts are not well-known. Here, we present an experimentally measured viscosity dataset for three end member compositions, characterized by a wide range of titanium contents, at lunar-relevant pressure-temperature range of $\sim$1.1–2.4 GPa and 1830–2090 K. In situ viscometry using the falling sphere technique shows that the viscosity of lunar melts varies between $\sim$0.13 and 0.87 Pa-s depending on temperature, pressure and composition. Viscosity decreases with increasing temperature with activation energies for viscous flow of $E_a = 201$ kJ/mol and $E_a = 106$ kJ/mol for low-titanium (Ti) and high-Ti melts, respectively. Pressure is found to mildly increase the viscosity of these intermediate polymerized melts by a factor of $\sim$1.5 between 1.1 and 2.4 GPa. Viscosities of low-Ti and high-Ti magmas at their respective melting temperatures are very close. However at identical P-T conditions ($\sim$1.3 GPa, $\sim$1840 K) low-Ti magmas are about a factor of three more viscous than high-Ti magmas, reflecting structural effects of Si and Ti on melt viscosity. Measured viscosities differ significantly from empirical models based on measurements of the viscosity of terrestrial basalts, with largest deviations observed for the most Ti-rich and Si-poor composition. Viscosity coefficients for these primitive lunar melts are found to be lower than those of common terrestrial basalts, giving them a high mobility throughout the lunar mantle and onto the surface of the Moon despite their Fe and Ti-rich compositions.

Keywords: moon, viscosity, high-pressure, synchrotron, magmas
INTRODUCTION

A combination of recent analyses of Apollo-era samples (e.g., Borg et al., 2011; Saal et al., 2013; Tartèse and Anand, 2013; Hauri et al., 2015), reassessment of lunar seismic data (Garcia et al., 2011; Weber et al., 2011), orbital measurements from recent lunar missions (e.g., Pieters et al., 2009; Yamamoto et al., 2010; Wu et al., 2012; Wieczorek et al., 2013), and results from advanced experimental and computational studies (e.g., de Vries et al., 2010; Elardo et al., 2011; Jutzi and Asphaug, 2011; Cuk and Stewart, 2012; van Kan Parker et al., 2012; Lin et al., 2017a,b; Charlier et al., 2018; Rapp and Draper, 2018; Zhao et al., 2019) are revolutionizing our view of the formation and general evolution of the surface and interior of the Moon. Detailed models of key aspects of lunar magmatic evolution are now hampered by a lack of quantitative constraints on the physical properties of lunar magma at high pressures ($P$) and temperatures ($T$). This lack of data plagues the accuracy of models of the physical and thermodynamic properties and resulting dynamics of the lunar interior. For example, the plagioclase-rich crust of the Moon exposed in the highland terranes is widely believed to have formed due to flotation of plagioclase in a global, crystallizing lunar magma ocean (Herbert et al., 1977; Elkins-Tanton, 2008; Lin et al., 2017a,b; Charlier et al., 2018; Rapp and Draper, 2018). Dynamic models of this flotation (Warren, 1990; Elkins-Tanton et al., 2011; Suckale et al., 2012) suffer from scarcity of data on the variations of density of both crystals and melts, and on the viscosity of lunar melts (which determine the upward velocity of rising crystals), at lunar high pressure-temperature conditions. Petrogenetic models for the origin of lunar mare basalts and picritic glasses are also strongly dependent on the dynamics of melt transport, and hence on lunar magma viscosities under high P-T conditions, on their route to the surface (e.g., Grove and Krawczynski, 2009).

Viscosity ($\eta$) is a critically important physical property of magma, governing the efficiency, rate and nature of melt transport and affecting the rates of crystallization and differentiation processes. It is a consequence of atomic-scale transport, and is therefore directly related to the structure and thermodynamic properties of the magma. Prediction of variations in viscosity as a function of pressure, temperature, and composition is challenging. Several empirical models have been proposed for predicting the temperature-composition dependence of magma viscosity over a range of anhydrous and hydrous terrestrial magma compositions at atmospheric pressure (Shaw, 1972; Hui and Zhang, 2007; Giordano et al., 2008). While these models are very successful in predicting the viscosities of terrestrial magmas, they have been shown inaccurate for compositions relevant to magmatism on other terrestrial bodies (e.g., Chevrier et al., 2014; Sehlke and Whittington, 2016). One particular feature of lunar magmas is their very wide range of titanium contents (Delano, 1986). The TiO$_2$ content of primitive lunar melts sampled during the Apollo missions varies between 0.26 wt.% for the Apollo 15C green glass to 16.4 wt.% for the Apollo 14 black glass (Delano, 1986). Chemical composition of lunar basalts estimated by remote sensing methods also confirm the wide range of TiO$_2$ contents in lunar basalts ranging from less than ~1 wt% to more than 14 wt% (Giguere et al., 2000; Carter et al., 2009). No systematic viscosity data set is available for the viscosity of titanium-rich magmas that do not occur on Earth but are widespread on the Moon. The empirical nature of current melt viscosity models precludes confident extrapolation to compositions outside of their calibration range. In addition, the effect of pressure on magma viscosity is poorly constrained, precluding extrapolation of the low-pressure model to lunar interior pressures.

This study aims to provide new viscosity data obtained from experimental in situ measurements on a range of primitive lunar melt compositions ranging from the Apollo 15 green glass with low Ti content to the Apollo 14 black glass with very high Ti contents. Using in situ falling sphere viscometry we assess the effects of pressure and temperature on primitive lunar melt viscosity, quantify the difference between measured values and model predictions, and describe the implications of our results for lunar magmatic processes.

MATERIALS AND METHODS

Liquid viscosity at high pressure and high temperature conditions was determined using the in situ falling sphere technique developed over two decades ago (e.g., Kanzaki et al., 1987; Suzuki et al., 2002). In this technique, X-ray radiographic images record the rate of downward descent of an X-ray opaque dense marker sphere as it falls through a molten sample of lower density. Experiments were performed at beamline ID27 of the European Synchrotron Radiation Facility (ESRF) in Grenoble (France) that combined a Vx5-type Paris-Edinburgh (P-E) press with in situ X-ray radiography and diffraction setups (Mezouar et al., 2005; Perrillat, 2008).

Starting material consisted of synthetic equivalents of low-titanium Apollo 15C green glass (0.26 wt% TiO$_2$), high-titanium Apollo 14 black glass (16.4 wt% TiO$_2$), and intermediate Ti-content Apollo 17 (74.220) orange glass (9.1 wt% TiO$_2$). These compositions bracket the observed Ti content variation in Apollo surface samples and enable quantification of the effect (linear or non-linear) of composition on lunar melt viscosity. The nominal bulk compositions of the starting mixtures are given in Table 1. High purity (99.5–100.05%, Alafa Aesar) powdered oxides MgO, MnO, Fe$_2$O$_3$, Al$_2$O$_3$, TiO$_2$, SiO$_2$, Cr$_2$O$_3$ and carbonates Na$_2$CO$_3$, KCO$_3$, and CaCO$_3$ were used to prepare the starting compositions. The oxides MgO, MnO, Al$_2$O$_3$, TiO$_2$, and SiO$_2$ were fired at 1000°C while the other oxides and carbonates were dried at 110°C prior to use. To prepare the starting glasses, required proportions of oxides and carbonates were first intimately mixed under ethanol in an agate mortar, and then dried in air and decarbonated in a Pt crucible in a one-atmosphere furnace by gradually raising the temperature from 650 to 1200°C over a period of 7 h. The Pt crucible had previously been iron-saturated to minimize Fe loss during starting composition preparation. Each individual mixture was finely ground in an agate mortar...
Figure 1

caps, 0.5 mm thick. These caps act as pistons, transferring limited to about one 10th of the diameter of the sample or the sphere settling velocity, the diameter of the sphere was positioned as close as feasible to the center relative to the capsule diameter. In order to avoid wall effects that could affect the position of non-bridging oxygens to the number of network-forming cations, following Mysen (1990), and assuming all Ti cations are tetrahedrally coordinated.

The configuration of the cell assembly used is shown in Figure 1. It consists of a boron epoxy gasket, a cylindrical graphite furnace and a single crystal diamond capsule as sample container. The latter is ideal for these kinds of experiments because of the high melting point, the high X-ray transparency and chemical interaction with its environment. Moreover, the high thermal conductivity of diamond ensures the lack of thermal gradients over the sample. For falling sphere viscometry, we used the high thermal conductivity of diamond to ensure a nominally anhydrous content for the powdered glasses.

The polymerization index NBO/T is calculated from the ratio of non-bridging oxygens to the number of network-forming cations, following Mysen (1990), and assuming all Ti cations are tetrahedrally coordinated.

TABLE 1 | Compositions of synthetic Apollo green, orange, and black glasses based on electron microprobe measurements (from Delano, 1986).

|          | Apollo 15C Green glass | Apollo 17 Orange glass | Apollo 14 Black glass |
|----------|------------------------|------------------------|------------------------|
| SiO₂     | 48                     | 38.9                   | 34                     |
| TiO₂     | 0.26                   | 8.78                   | 16.4                   |
| Al₂O₃    | 7.74                   | 5.81                   | 4.6                    |
| Cr₂O₃    | 0.57                   | 0.67                   | 0.92                   |
| FeO³⁺    | 16.5                   | 22.3                   | 24.5                   |
| MgO      | 18.2                   | 15.7                   | 13.3                   |
| MnO      | 0.19                   | 0.27                   | 0.31                   |
| CaO      | 8.57                   | 7.37                   | 6.9                    |
| Na₂O     | 0                      | 0.26                   | 0.23                   |
| K₂O      | 0                      | 0                      | 0.16                   |
| Total    | 100.03                 | 100.06                 | 101.32                 |
| NBO/T    | 1.57                   | 1.65                   | 1.60                   |

under acetone and subsequently fired at 1500°C. The resulting glasses were ground under ethanol once more in an agate mortar for 1 h, dried and stored at 110°C before use, in order to ensure a nominally anhydrous content for the powdered glasses. The graphite caps were enclosed in an hBN cylinder, acting as P-transmitting media. A poly-ether-ether-ketone (PEEK) ring was added around the gasket to prevent the gasket from flowing out during heating and compression. P-T calibrants were packed in a lateral hand-drilled hole, Ø ~ 0.2 mm, in the middle of the hBN cylinder, touching the outside of the diamond capsule. Calibrants were high-purity Pt powder (99.99%, Alfa Aesar) mixed with hBN powder (99.99 wt%, Goodfellows). This high-pressure sample assembly was fully pressure-calibrated by van Kan Parker et al. (2010), who showed that at T > 1250 K, pressure is fully transmitted by the graphite end caps to the sample.

Following the approach of Rai et al. (2013), after loading the cell assembly in the P-E press, the cells were first cold compressed before increasing the temperature in small steps up to approximately 200 K below the liquidus temperature of the compositions and maintained there for several minutes to allow thermal settling of the assembly. Heating to the final temperature was performed very rapidly at a rate of 100 W/min to ensure full and rapid melting of the sample above the liquidus. At different stages of the experiments, synchrotron X-ray diffraction patterns were systematically collected for both sample and calibrants. All 2D diffraction images were treated and integrated using the Fit2D data analysis software (Hammersley et al., 1996). Sample diffraction patterns were used to assess the physical state of the sample (Figure 2). Note that the diffuse scattering from the liquid that is observed in diffraction patterns (Figure 2 middle) is not readily suitable for structural investigations of the melt structure because (i) incident X-ray energy was optimized for radiography contrast instead of diffraction quality and (ii) measured intensities were not corrected for the response of the Soller’s slit which is non-linear (see Morard et al., 2011). Calibrated diffraction patterns were analyzed using the EXPGUI graphical interface of GSAS which is a comprehensive system for the refinement of structural models to x-ray diffraction data (Toby, 2001). In order to calculate the P-T conditions from the
FIGURE 2 | Typical X-ray diffraction images acquired on sample B2, black glass composition; at (bottom) sub-liquidus conditions, showing diffraction lines of hBN and Pt calibrants; at (middle) superliquidus conditions of 2.4 GPa – 1906 K. Full melting is evidenced by a broad band of diffuse scattering and the absence of diffraction spots. The corresponding integrated intensity spectrum is plotted in Q-range scale. (top) After temperature quench, the sample is fully recrystallized.

diffraction lines of hBN and Pt, third order Birch Murnaghan equations of state (EOS) for Pt (Fei et al., 2004) and hBN (Zhao et al., 1997; Le Godec et al., 2000) were used. Temperature and pressure uncertainties resulting from the cross-calibration of the equations of state of the two calibrants are estimated as ±50 K and ±0.2 GPa, respectively. The absence of peaks from sample X-ray diffraction patterns was used to confirm that superliquidus temperatures had been reached, and the P-T conditions at which this occurred are in agreement with the melting phase relations (Figure 3) determined ex situ for the bulk compositions used (Wagner and Grove, 1997; Elkins-Tanton et al., 2003a; van Kan Parker et al., 2012).

Radiographic images were acquired every 20 ms during the final temperature increase using a high-speed CCD camera (Perrillat et al., 2010). Images recorded the fall of the dense Re sphere as soon as the melting temperature was reached. During and after radiographic observations, P-T conditions were held constant and diffraction patterns of the liquid and P-T calibrants were collected. The sample was then quenched in temperature by cutting power to the furnace.

RESULTS

We performed a total of seven successful runs covering the temperature range between 1830 and 2090 K and pressure range from 1.1 to 2.4 GPa (Figure 3 and Table 2). Time-lapsed radiographic images of a typical successful run are shown in Figure 4, with a falling rhenium sphere measured at 2.4 GPa and 1906 K in a molten black glass sample (sample B2, also shown in Figure 2). The corresponding time versus falling distance profile (Figure 4) is constructed from the location of the top and bottom points of the sphere, tracked on each individual frame using the Metavue® image software. The time-falling distance
plot shows a sigmoid shape with non-linear regions at both ends, reflecting acceleration as the sphere starts to fall before achieving the terminal velocity, and deceleration of the sphere as it approaches the bottom of the capsule. The time versus falling distance profiles in all runs reported in this study do not exhibit variations or discontinuities that could be related to convection or thermal gradients in the cell. The velocity curve in the inset of Figure 4 is derived by fitting the time-distance profile with a cubic spline function and numerical differentiation. It shows a plateau of constant velocity consistent with the achievement of steady state velocity. The terminal velocity is determined by linear regression of this flat-top interval. Comparison of rates derived from the motion of the top and bottom of the spheres

| Sample | P (GPa) ±0.2 | T (K) ± 50 K | r_s (µm) ±2 µm | V (µm/s) | Experimentally measured η (Pa·s) | Model predicted Giordano et al. (2008) η (Pa·s) | Model predicted Sehlke and Whittington (2016) η (Pa·s) |
|--------|-------------|-------------|----------------|---------|--------------------------------|--------------------------------|--------------------------------|
| G1     | 1.1         | 2090        | 58             | 106 ± 5 | 0.17 ± 0.02                     | 0.05                          | 0.15                          |
| G2     | 1.2         | 1830        | 53             | 25 ± 2  | 0.87 ± 0.08                     | 0.26                          | 0.87                          |
| G3     | 2.1         | 1980        | 66             | 84 ± 4  | 0.43 ± 0.03                     | 0.15                          | 0.28                          |
| G1     | 1.3         | 1840        | 66             | 61 ± 2  | 0.58 ± 0.04                     | 0.48                          | 0.81                          |
| B1A    | 1.3         | 1860        | 58             | 93 ± 14 | 0.27 ± 0.07                     | 1.15                          | 0.61                          |
| B1B    | 1.1         | 2080        | 58             | 181 ± 18| 0.13 ± 0.02                     | 0.2                           | 0.14                          |
| B2     | 2.4         | 1906        | 76             | 111 ± 6 | 0.37 ± 0.03                     | 0.75                          | 0.43                          |

Model predicted viscosities at 1 atm. pressure from Giordano et al. (2008) and Sehlke and Whittington (2016) are listed for comparison.
in the same run and errors calculated from the fitting procedures yield an absolute uncertainty of ±2 to ±18 μm/s in the resulting terminal velocities.

For a solid sphere of density $\rho_s$ and radius $r_s$ falling in a liquid of density $\rho_l$, the viscosity $\eta$ is given by Stokes’ relation:

$$\eta = \frac{2gr_s^2(\rho_s - \rho_l)}{9V} \cdot \frac{F}{K}$$

(1)

where $g$ is the acceleration due to gravity, and $V$ the settling velocity. The parameters $F$ and $K$ correspond to the wall effect (Faxen, 1925) and finite length (Kingery, 1959) corrections respectively, where:

$$F = 1 - 2.104 \left( \frac{r_s}{r_c} \right) + 2.09 \left( \frac{r_s}{r_c} \right)^3 - 0.95 \left( \frac{r_s}{r_c} \right)^5$$

(2)

$$K = 1 + 3.3 \left( \frac{r_s}{h} \right)$$

(3)

In these equations, $r_c$ is the internal diameter of the capsule ($r_c = 0.25$ mm), and $h$ the sample length, taken to be the entire fall distance. The densities of the melts were calculated from the equation of state parameters of the Apollo green, orange and black glasses reported in van Kan Parker et al. (2012). The density of rhenium at high pressure and temperature was calculated from its equation of state (e.g., Ahrens and Johnson, 1995).

Because of the large contrast between the density of Re and the density of silicate melts, the viscosity estimates are not significantly affected by uncertainties in the densities of the sphere and the melt. For example, an error of 500 g cm$^{-3}$ on the parameter $(\rho_s - \rho_l)$ in Eq. 1 results in <3% error in the calculated melt viscosity. Overall uncertainty mainly stems from the dispersion in falling velocities leading to absolute errors of ±0.02 to ±0.08 Pa·s on melt viscosities. Experimental conditions, sphere diameters, terminal velocities and calculated viscosities are summarized in Table 2. The viscosity coefficients obtained in this study range from 0.13 Pa·s for the black glass at 1.1 GPa and 2080 K to 0.87 Pa·s at 1.2 GPa and 1830 K for the green glass composition.

**DISCUSSION**

**Effects of Temperature, Pressure, and Composition**

Measurements were made over the temperature range 1830–2090 K, which for all three compositions includes the region above the liquidus. A graphical illustration of the logarithm of viscosity as a function of reciprocal temperature for each glass composition is given in Figure 5. Within the investigated temperature range, the viscosities can be reasonably assumed to follow a simple Arrhenius relationship:

$$\log \eta = \log \eta_{0,T} + \frac{E_a}{RT}$$

(4)

where $\eta_{0,T}$ is a pre-exponential factor, $E_a$ the activation energy for viscous flow, and $R$ the gas constant. The values of $\log \eta_{0,T}$ and $E_a$ in Eq. 4 were calculated from simple linear regression of $\log \eta$ on $1/T$ from nearly isobaric experiments at 1.1–1.2 GPa for the green glass, and 1.1–1.3 GPa for the black glass composition. The best fit parameters are $E_a = 201 ± 26$ kJ/mol − log $\eta_{0,T} = -5.8 ± 0.7$ Pa·s and $E_a = 106 ± 46$ kJ/mol − log $\eta_{0,T} = -4.1 ± 1.5$ Pa·s for the green and black glass, respectively. For the orange glass, we consider the activation energy $E_a$ to range between the two above values considering its intermediate composition.

These activation energies suffer from significant uncertainties because of the limited dataset; however, they are in agreement with activation energies for viscous flow determined experimentally at high-temperature (T > 1600 K) and ambient pressure for a variety of silicate melts (Mysen and Richet, 2005). For example, Liebske et al. (2005) measured an activation energy $E_a = 197$ kJ/mol for a peridotite liquid. For Ti-bearing ferrobasaltic melts, Chevrel et al. (2014) reported $E_a = 179$ kJ/mol for a 1.1 wt% TiO$_2$ and 20.36 wt% FeO composition, and Dygert et al. (2017) derived $E_a = 153$ kJ/mol for a 4.1 wt% TiO$_2$ and 29.9% FeO liquid. For lunar mare basalt compositions, the high-temperature data of Uhlmann et al. (1974) on a synthetic Apollo15 green glass showed $E_a = 167$ kJ/mol, and the more recent experimental measurements by Sehlke and Whittington (2016) on a high-Ti (9.54 wt% TiO$_2$) sample produced $E_a = 165$ kJ/mol. The present activation energy for the green glass $E_a = 201 ± 26$ kJ/mol is also consistent with the value of $E_a = 180$ kJ/mol recently estimated by Dufilis et al. (2018) using molecular dynamics simulation on a similar green glass (A15G) composition.

As for temperature, the pressure dependence of melt viscosity can be described by an Arrhenian behavior:

$$\log \eta = \log \eta_{0,P} + P.V^*/RT$$

(5)
where $\eta_{0,P}$ is the zero pressure viscosity, and $V^*$ the activation volume of viscous flow. Experimentally determined viscosities as a function of pressures are reported in Figure 6. To evaluate the variation of melt viscosity with pressure at constant temperature, we have interpolated viscosity data using the above Arrhenius parameters $E_a$ and $\log \eta_{0,T}$ to common isotherms $T = 1980$ K and $T = 1906$ K, for the green and black glass respectively. Both isotherms show a linear variation of $\log \eta$ with pressure, with best fitted parameters of linear regression $V^* = 2.2 \pm 2$ cm$^3$/mol – $\log \eta_{0,P} = -0.5 \pm 0.1$ Pa·s for the green glass, and $V^* = 5.9 \pm 2$ cm$^3$/mol – $\log \eta_{0,P} = -0.8 \pm 0.1$ Pa·s for the black glass (Figure 6). Previous studies found the pressure-viscosity relationship to depend on the degree of polymerization of the melt (for a review see, Wang et al., 2014). While viscosities of depolymerized melts (NBO/T > 2) increase monotonically with pressure, those of polymerized melts (NBO/T < 1) initially decrease with pressure, defining a viscosity minimum. For example, Liebske et al. (2005) found a positive activation volume $V^* = 4.9$ cm$^3$/mol for peridotite liquid (NBO/T $\approx 2.5$) in the 0–7 GPa range. In contrast, for liquid basalt (NBO/T $\approx 0.8$) Sakamaki et al. (2013) observed a decrease up to 4 GPa following an activation volume of $V^* = -5.1$ cm$^3$/mol. For the high-Ti black glass composition of our study with NBO/T $\approx 1.6$, the positive values of $V^*$ imply an increase of viscosity by a factor of $\sim 1.5$ between 1.1 and 2.4 GPa. This minor increase in pressure is consistent with the free-volume theory, and with predictions from MD simulations (Dufils et al., 2018) on green (A15G), orange (A17O), and black (A14B) glass compositions.

Equations 4 and 5 can be used to predict viscosity values at common $P$-$T$ conditions, enabling isolation of melt compositional effects. In Figure 7 the viscosities of green, orange, and black glass melts at their respective melting temperatures ($T_m$) at $\sim 1.2$–1.3 GPa are plotted against their TiO$_2$/(TiO$_2$ + SiO$_2$) contents. Also reported are the viscosity data of samples G2, O1, and B1A corrected to a common pressure – temperature point of 1.3 GPa and 1840 K, close to the experimentally investigated conditions. While the viscosities of low-Ti and high-Ti magmas at their respective liquidi are very close, under similar temperature conditions, the viscosity of the green glass melt is found to be the highest, the orange glass melt having an intermediate value and black glass melt with the highest Ti content being least viscous. Quantitatively, our data suggest a viscosity decrease by a factor of $\sim 3$ from low-Ti to high-Ti lunar magma at identical $P$-$T$ conditions.

At first sight, this compositional dependence may seem unexpected since titanium is generally considered to be a network former, in which case increasing amounts of TiO$_2$ in silicate melts would make it more polymerized and hence more viscous (Mysen et al., 1980). However, the structural role of titanium in silicate melts has previously been found to be complex, with more than one structural position of Ti$^{4+}$ and
viscosity to decrease with increasing TiO$_2$ (Mysen et al., 1980); and could lead to the observed melt O-Si bonds, because the Ti cation is larger than that of Si of Ti in models. 

reflects the difficulty to capture the complex structural effect not reproduced in MD simulations (Dufils et al., 2018) which low-Ti counterparts. This compositional dependence is however of quench growth textures in Ti-rich melts relative to their similar P-T conditions, the viscosity is found to significantly decrease with the Ti content.

Several hypotheses have been proposed to explain this dependence. The substitution of Si by Ti in tetrahedral sites probably makes Ti-O-Ti bonds weaker than Si-O-Al and Si-O-Si bonds, because the Ti cation is larger than that of Si (Mysen et al., 1980); and could lead to the observed melt viscosity to decrease with increasing TiO$_2$ content. Moreover, increasing the TiO$_2$ content of the melt, a significant proportion of Ti$^{4+}$ cations can be coordinated with more than four oxygens, forming Ti[5] or Ti[6] polyhedra (Dickinson and Hess, 1985; Henderson and Fleet, 1995). Such an increase in coordination number would reduce the degree of polymerization and hence viscosity. Dygert et al. (2013) proposed that the addition of TiO$_2$ in these lunar silicate melts leads to the formation of regions enriched in Fe-O-Ti complexes, effectively depolymerizing the melt. Interestingly, the X-ray intensity scattered by the present molten glasses (Supplementary Figure S1) displays a change in the first sharp diffraction peak (FSDP) position as a function of composition. The position of the FSDP is shifted to higher Q with increasing Ti content (where Q = 4πsinθ/λ, is the momentum transfer, θ the half scattering angle, and λ the wavelength). The FSDP is generally accepted to be related to intermediate-range order (IRO) of the network (Salmon, 1994; Inamura et al., 2004). Hence, the observed FSDP shift argues for a more closely packed IRO of the black glass relative to the green one, which would agree with the more depolymerized and less viscous character of these Ti-rich lunar magmas. A definitive answer on the origin of the observed Ti viscosity dependence would require thorough additional experimental investigations and/or numerical modeling of the structural effects of Ti incorporation in silicate liquids.

**Comparison With Previous Work and Model Predictions**

The first viscosity measurements on lunar lavas were performed soon after the Apollo missions in the early seventies (Murase and Mc Birney, 1970; Weil et al., 1971; Uhlmann et al., 1974). These early works already outlined the lower viscosity of Ti-bearing lavas relative to terrestrial basalts (Hawaii, Columbia River Basalts). Uhlmann et al. (1974) reported a 1 atm viscosity for molten Apollo 15 green glass of $\eta$ = 1.07 Pa-s at 1785 K, and Murase and Mc Birney (1970) obtained $\eta$ = 0.6 Pa-s for molten Apollo 11 orange glass at 1773 K – 1 atm. For comparison with our high-temperature-pressure data, the 1 atm measurement of Uhlmann et al. (1974) was extrapolated to $T = 1980$ K using the $E_g$ for green glass derived in this study, and then compared with the one extrapolated at ambient P along the 1980 K isotherm in Figure 6. The corrected Uhlmann et al. (1974) viscosity $\eta$ = 0.54 Pa-s is higher than the one derived from our data $\eta$ = 0.33 Pa-s; however the difference is not unacceptable considering experimental uncertainties. Recently, Sehlke and Whittington (2016) measured the 1 atm viscosity of a synthetic high-Ti lunar basalt (LM sample, 9.52 wt% TiO$_2$), and obtained $\eta$ = 0.19 Pa-s at 1850 K, which extrapolates to $\eta$ = 0.14 Pa-s at 1906 K. This value is again close to our present estimate for black glass composition $\eta$ = 0.15 Pa-s at 1 atm – 1906 K. Dygert et al. (2017) reported a viscosity of $\eta$ = 0.22–1.45 Pa-s at experimental conditions, 1573–1873 K and 0.1–4.4 GPa, for a late Lunar Magma Ocean (LMO) analog. Although a direct comparison cannot be made because the composition of the late LMO (Fe- and Ti-rich ferrobasalt) is quite different from the present primitive lunar melts, both studies point to very low viscosities for these lunar melts.

Owing to experimental difficulties, published data on the viscosity of primitive lunar melts at high P-T conditions are scarce. The only literature data are a single viscosity point for the green glass composition at 3.1 GPa and 1915 K by Maeda et al. (2004), and six viscosity points for the black glass composition measured by Suzuki et al. (2009) in the 1733–1803 K and 1.0–3.5 GPa range. These data are plotted in
The viscosities of a wide range of hydrous and anhydrous experimental observations.

Reflects the prediction of the models of increasing viscosity is higher for the black glass than the orange glass, and model values are significantly higher than the experimentally below our extrapolated value at 1 atm., while the Sehlke and Whittington (2016) based on composition predicted by the empirical model of Giordano et al. (2008) and Sehlke and Whittington (2016). These models are based on composition and temperature (Bottinga and Weill, 1972; Giordano and Dingwell, 2003; Hui and Zhang, 2007; Giordano et al., 2008; Sehlke and Whittington, 2016). These models are based on experimental measurements done on natural terrestrial and some planetary compositions and their synthetic analogs at atmospheric pressure (Hui and Zhang, 2007; Giordano et al., 2008). Figure 6 and Table 2 show a comparison of the experimentally measured viscosities for the green, orange and black glass compositions to the viscosity values predicted by the empirical model of Giordano et al. (2008) and Sehlke and Whittington (2016) based on composition and temperature at ambient pressure. For comparison we also extrapolated our high-pressure data to 1 atm using the Arrhenius relationship (Figure 6), although these low pressure estimates are actually very dependent on the uncertainty on the activation volume $V^*$. The green glass model predicted viscosity by Giordano et al. (2008) at 1980 K falls significantly below our extrapolated value at 1 atm., while the Sehlke and Whittington (2016) model better reproduce the experimental data. However, for the orange and black glasses we find that both model values are significantly higher than the experimentally measured viscosity values. This overestimation of viscosities is higher for the black glass than the orange glass, and reflects the prediction of the models of increasing viscosity with increasing TiO$_2$ content of the melt, contrary to our experimental observations.

This shows that empirical models that can successfully predict the viscosities of a wide range of hydrous and anhydrous terrestrial compositions cannot be extrapolated to predict viscosities of Ti rich lunar melts. This could be due to reasons that include (1) the TiO$_2$ content of the black and orange glasses are out of the range of compositions on which this model is based upon and (2) our viscosity measurements were done at high pressure conditions whereas models are based on 1 atm data. Considering the recent identification of significant amount of water in the source of several basaltic lunar magmas (Hauri et al., 2011, 2015; Hui et al., 2013; Saal et al., 2013), there is a clear need for more experimental data on melt viscosity determinations on lunar compositions including the effect of H$_2$O and other volatile species.

**Implications for Lunar Evolution**

Primitive lunar melts occur as basaltic flows filling impact basins and as picritic glass beads erupted in lava fountains. These volcanic rocks have crystallization ages typically between 3900 and 3200 Ma, and have been sourced from deep silicate reservoirs in the lunar mantle generated during the LMO crystallization (e.g., Grove and Krawczynski, 2009). Petrogenetic models account for their elevated Ti abundances by either invoking a hybrid source produced by mixing late-stage ilmenite-pyroxene cumulates of the LMO with low-Ti rocks during a mantle overturn (Ringwood and Kesson, 1976; Hess and Parmentier, 1995), or by assimilation of Fe-Ti oxides in picritic glass melts as they migrate to the surface (Hubbard and Minear, 1975; Wagner and Grove, 1997). The combination of inferred deep formation (i.e., 200–500 km) and observed high titanium and iron contents begs the question of mobility of these primitive melts relative to their sources rocks. Gravity-driven transportation of magmatic liquids within a planetary interior is proportional to hydrostatic melt mobility $\Delta \rho/\eta$, where $\Delta \rho$ is the density contrast between the magmatic liquid and the surrounding solid silicate and $\eta$ the viscosity of the melt (Kono et al., 2014). Since current viscosity prediction models are not applicable to lunar melt compositions at non-ambient pressures, it becomes crucial that only relevant input values determined through experimental measurements are used in lunar interior evolution models. By looking at different bulk compositions our data provide insight into the first-order effects of titanium on melt physical properties. Calculating values for melt densities from EoS and the lunar mantle density profile from van Kan Parker et al. (2012), and using the viscosity values derived from this work, we find that under identical conditions of $P$-$T$ (1.3 GPa, 1840 K), the black glass melt would exhibit maximum mobility (1.64 g cm$^{-3}$ Pa$^{-1}$s$^{-1}$), the orange glass melt would show intermediate mobility (0.9 g cm$^{-3}$ Pa$^{-1}$s$^{-1}$) and the green glass melt would show lowest mobility (0.62 g cm$^{-3}$ Pa$^{-1}$s$^{-1}$). This suggests that the low buoyancy of high-Ti melts relative to the Moon's interior density profile is counterbalanced by their very low viscosity, enabling their rise through the lunar mantle. Additionally, these very high melt mobilities, much higher compared to terrestrial basaltic lavas (Sato, 2005; Villeneuve et al., 2008; Sakamaki et al., 2013), would lead to fast migration rates of these picritic glasses. Rapid magma ascent would affect the extent of chemical reaction (i.e., assimilation, fractionation) of magmas with cumulate piles during their
route to the surface, and also aid final eruption through the anorthosite crust.

CONCLUSION

We have presented the first comprehensive experimentally measured viscosity of pristine lunar melts that span a range of TiO$_2$ contents from 0.26 to 16.4 wt%, showing that viscosity is a complex function of chemical composition under high pressure and high temperature conditions. We find an Arrhenian dependence of lunar melt viscosities on temperature, and an increase in viscosity with pressure for these intermediate polymerized melts. We also show that there can be a substantial change in lunar melt viscosities with changing TiO$_2$ content of the melt.

The possibility of linking viscosity variations to local melt structure variations paves the way to comprehensive structural models of magma viscosity. We find that experimentally derived viscosity coefficients for the lunar glasses are lower than those reported for terrestrial basalts. Such low viscosities would be consistent with thin and extensive lunar lavas flows as observed (Head, 1976). We also find significant differences between the experimentally derived values and model predicted values, which underlines the need for a systematic experimental determination of viscosities of lunar melts under relevant high $P$-$T$ conditions. In spite of recent development of empirical viscosity models for terrestrial compositions, there is still a need for a more extensive model that would also be applicable to terrestrial silicate melts for a range of temperature and pressure conditions.

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AUTHOR CONTRIBUTIONS

NR, J-PP, and WvW designed the project, analyzed the results, and wrote the manuscript with input from all authors. All authors performed the experiments.

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SUPPLEMENTARY MATERIAL

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FIGURE S1 | X-ray intensity scattered by the green, orange, and black molten glasses under similar conditions of 1.2 ± 0.2 GPa and 1850 ± 50 K. The first sharp diffraction peak (FSDP) position is shifted to higher Q with increasing Ti content.
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