Distinguishing between basalts produced by endogenic volcanism and impact processes: A non-destructive method using quantitative petrography of lunar basaltic samples

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Abstract

Impact processes play an important role in shaping and reshaping the surfaces of airless planetary bodies. Such processes produce regoliths and generate melts that crystallize and record the homogenization of the geology at the impact site. If the volume of melt is substantial, the resultant crystallized product has an igneous texture and may be free of xenolithic clasts making it difficult to distinguish from melts produced by endogenic magmatic processes. This has been clearly demonstrated during the return of the Apollo samples from the Moon, where Apollo 14 basalt 14310 was initially described as a mare basalt and was only subsequently reclassified as an impact melt following detailed and time consuming crystallization experiments. Another way of distinguishing lunar impact melts from endogenically-derived mare basalts is through the quantification of the highly siderophile elements (HSE: Pd, Rh, Ru, Ir, Pt, Os), which have relatively low abundances in pristine lunar samples but are high in meteorites and, therefore, may be enriched in impact melts. However, these analyses consume relatively large quantities of valuable sample and because of mass constraints cannot be performed on many lunar samples. In this paper we present a quantitative petrographic method that has the potential to distinguish lunar impact melts from endogenically-derived mare basalts through the quantification of the highly siderophile elements (HSE: Pd, Rh, Ru, Ir, Pt, Os), which have relatively low abundances in pristine lunar samples but are high in meteorites and, therefore, may be enriched in impact melts. However, these analyses consume relatively large quantities of valuable sample and because of mass constraints cannot be performed on many lunar samples. In this paper we present a quantitative petrographic method that has the potential to distinguish lunar impact melts from endogenically-derived mare basalts using plagioclase and olivine crystal size distributions (CSDs). The slopes and intercepts of these CSDs are used to show that olivine from impact melts displays a steeper CSD relative to olivine from mare basalts. For plagioclase, generally impacts melts display CSDs with shallower gradients than those from endogenous mare basalts and, as for olivines, plot in a distinct field on a CSD slope vs. CSD intercept plot. Using just a thin section to distinguish impact melts from mare basalts enables the goals of future robotic sample return missions to determine the age of the South Pole-Aitken basin in the Moon, because such missions will potentially only return small (2–4 mm) “rocklets” for analysis, obviating HSE analyses for impact melt identification.

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1. INTRODUCTION

Basaltic samples returned from the Moon can have distinctly different origins through endogenic volcanism (producing pristine mare basalts) or through impact melting processes, and distinguishing between these origins on the basis of texture is difficult. As an illustrative example,
the textures of two Apollo 14 impact melts (14073 and 14276; Fig. 1a and b, respectively) appear texturally similar to a pristine mare basalt (14072) (Fig. 1c). Furthermore, although basaltic sample 14310 was originally classified as a pristine high-Al mare basalt (e.g., Longhi et al., 1972; Ridley et al., 1972; Crawford and Hollister, 1974), subsequent experimental studies of dynamic crystallization demonstrated its impact origin (e.g., Usselman and Lofgren, 1976; Lofgren, 1977). Traditionally, such distinctions required time consuming experiments (e.g., Usselman and Lofgren, 1976; Lofgren, 1977), or destructive analyses using relatively large quantities (0.4 g to >1.0 g) of
material to determine the highly siderophile element (HSE) abundances of the samples (e.g., Anders et al., 1971; Anders, 1978; Wolf et al., 1979). It has been demonstrated that pristine lunar samples contain HSE abundances that are exceedingly low in the pg/g range (e.g., Righter et al., 2000; Walker et al., 2004; Day et al., 2007; Neal et al., 1988ab; Philpotts et al., 1972; Palme et al., 1978; Rose et al., 1972; Ryder and Norman, 1980; Shervais et al., 1985b; Taylor et al., 1972; Vaniman and Papke, 1980; Warren and Wasson, 1978; Willis et al., 1972).

While HSE abundances are a potentially good indicator of a non-pristine origin of impact melt rocks (although not an absolute indicator; e.g., Warren, 1993), not all returned samples are large enough to allow accurate HSE abundances to be determined (e.g., breccia clasts, rake samples, etc.). Given that in the near future any sample return mission to the Moon (or any other planetary body) will be robotic in nature, the sample masses will be small. Furthermore, the National Research Council (2007) placed understanding the bombardment history of the inner Solar System as uniquely revealed by the Moon as its top priority for lunar science. This is reflected in the current NASA Planetary Sciences Division Decadal Survey (National Research Council, 2011) where a sample return from South Pole Aitken basin to determine the age of the impact is a named, high priority New Frontiers class mission. Identification of impact melt samples in the sample return cache is paramount for achieving the goals of the mission. As this robotic sample return mission will collect rake samples, it is unlikely that sufficient sample will be available for HSE determinations and accurate age determinations. Therefore, building on the experimental studies from the 1970s noted above for 14310 (Usselman and Lofgren, 1976; Lofgren, 1977), textures will need to be used to distinguish basaltic samples derived from impacts vs. endogenic volcanism. A textural method that will achieve this is presented here.

Quantitative textural studies of impact-generated rocks have been conducted previously, but have focused on clast-size distributions, rather than crystal-size distributions. For example, Pittarello and Koeberl (2013) used a similar quantitative petrographic approach to study impact lithologies from the El’gygytgyn impact structure in Arctic Russia. As the site contained a variety of volcanic lithologies, the clast size distribution method they developed could not be applied to distinguish impact and unshocked lithologies. Pittarello and Koeberl (2013) focused on the suevite to define lithic clast size distributions by their geometric properties and found the method allowed the classification of unshocked to slightly shocked volcanic clasts included in

![Fig. 2. Siderophile element abundances in the samples being studied in this paper. (a) Ir vs. Au abundances demonstrating that the impact melts used in this study have much higher abundances; (b) Ni vs. Co plot showing that impact melts have higher Ni and lower Co abundances than endogenous mare basalts. Data from: Baedecker et al. (1972), Ebihara et al. (1992), Ehmann et al. (1972), Fagan et al. (2013b), Fischer-Godde and Becker (2012), Hertogen et al. (1977), Hubbard et al. (1972), Karkoski et al. (1973), Laul et al. (1972), Longhi et al. (1972), Morgan et al. (1972), Neal (2001), Neal and Kramer (2006), Neal et al. (1988ab), Philpotts et al. (1972), Palme et al. (1978), Rose et al. (1972), Ryder and Norman (1980), Shervais et al. (1985b), Taylor et al. (1972), Vaniman and Papke (1980), Warren and Wasson (1978), Willis et al. (1972).](image-url)
the suevite. Other examples include the clast size distributions in pseudotachylytes (Shimamoto and Nagahama, 1992; Tsutsumi, 1999) and impactites (e.g., Chanou et al., 2014).

This paper presents a quantitative petrographic method that distinguishes basaltic samples formed through endogenous processes from those formed by impacts using olivine and plagioclase crystal size distributions (CSDs). Comparison of the slopes and y-axis intercepts of these CSDs allows the distinction to be made between the two petrogeneses and builds upon the work of several recent contributions (Oshrin, 2009; Oshrin and Neal, 2009; Hui et al., 2011; Fagan, 2012; O’Sullivan, 2012; Fagan et al., 2013a; and Fagan and Neal, 2012; Donohue, 2013). During these studies, it has been demonstrated that phenocrysts can be distinguished from xenocrysts in impact melts by texture and composition (e.g., Figs. 3–5 in Fagan et al., 2013a,b). This work represents a culmination of 6 years work by current and former Notre Dame graduate students (and coauthors on this paper) in constructing CSDs as part of their respective degree projects.

2. SAMPLES

Plagioclase is ubiquitous in both pristine mare basalts and impact melts; olivine is an early crystallizing phase in some mare basalts and impact melts. Building upon the work reported by Oshrin (2009), Hui et al. (2011), Neal et al. (2010), Fagan and Neal (2012), and Donohue (2013), plagioclase CSDs have been constructed for mare basalts (see Table 1 for specific thin sections) from Apollo 12 (N = 14), 14 (N = 20), 16 (N = 5), and 17 (N = 9), as well as for basaltic impact melts from Apollo 14 (N = 3) and 16 (N = 9). None of these samples exhibit an alignment of crystals indicating they represent a random sampling of crystals that exhibit no flow texture. Complementing the work of Fagan et al. (2013a), olivine CSDs (Table 2) have also been constructed for additional Apollo 12 (N = 5), Apollo 14 (N = 3), Apollo 16 (N = 2) and Apollo 17 (N = 7) basalts as well as for known Apollo 14 impact melts (olivine vitrophyres N = 4; described as impact melts by Allen et al., 1979; Shervais et al., 1988). The mare basalts used in this study represent samples from the Apollo 12 pigeonite and ilmenite basalt suites plus the one feldspathic basalt (12038) (Neal et al., 1994a,b); each geochemical group of the Apollo 14 high-Al basalts (Neal and Kramer, 2006) excluding the very high potassium basalts (“VHK”- Shervais et al., 1985a; Neal et al., 1988a, 1989a,b); three Apollo 16 basalts (Zeigler et al., 2006; Fagan and Neal, 2012); and each geochemical group from the Apollo 17 mare basalts (Rhodes et al., 1977; Neal et al., 1990; Ryder, 1990). Detailed petrographic descriptions for each sample have been previously reported in the literature and the relevant references are contained within Tables 1 and 2.

Not all of the samples analyzed have been analyzed for the highly siderophile elements. All siderophile element data that have been published for the samples studied here
are represented in Fig. 2. For the highly siderophile elements Au and Ir (Fig. 2a), the impact melts contain 1–2 orders of magnitude more than the mare basalts. Substantially more data are available when Ni and Co are considered (Fig. 2b). The impact melts contain substantially different Ni/Co ratios than the mare basalts (8–17 vs. <2) as they contain generally higher Ni and lower Co abundances than mare basalts. Fig. 2 demonstrates that the samples we are classifying as impact melts and endogenous mare basalts have chemical compositions consistent with that classification.

3. METHOD

Crystal size distributions (CSDs) are used to quantify population densities within particular size bins of a given mineral phase and have been developed to define crystallization history for terrestrial samples (e.g., Cashman and Marsh, 1988; Marsh, 1988, 1998; Cashman, 1990). To construct a CSD in this study, individual crystals of a given phase were traced from thin section digital photomicrograph mosaics using Adobe Photoshop® and the outline of the entire section was also traced. Minerals that intersected the edge of the thin section were omitted, as they do not represent complete crystals. The resulting crystal layers were imported into ImageJ (Rasband, 1997; Higgins and Chandrasekhararam, 2007), which measures the major and minor axes, roundness, and area of each crystal. ImageJ is used in preference over the older ImageTool (Higgins, 2000) because ImageJ can accommodate larger image file sizes, thus better retaining the integrity of the smallest crystals. Individual crystals with minor axes <0.03 mm are likely to be a projection of a crystal rather than an accurate measurement due to the approximately equivalent thickness of the thin-section (Higgins, 2000) and a downturn (positive slope) in the CSD is usually seen at these small sizes. Processing of the two-dimensional thin section image to a three-dimensional representation requires the extraction of parameters that describe the intersections of the individual crystals (e.g., length, width, area, perimeter, orientation and centroid location) with the plane of thin section. The major and minor axes of crystals were imported into CSDslice (Morgan and Jerram, 2006) to determine the best-fit short, intermediate, and long axes of the 3-D crystal habit. These data, in conjunction with the major and minor axes, individual crystal area, average crystal roundness, and total sample area, were used in CSDcorrections 1.39 (Higgins, 2000, 2002a; Higgins and Chandrasekhararam, 2007) to determine the 3-D CSD using 5 bins per decade (i.e., per log unit). Higgins (2000) reported that using more than 5 bins per decade can introduce errors because (1) there are fewer crystals in each bin,
and (2) more cycles of correction are needed during stereological conversion. Finally, CSDs are traditionally plotted as the natural log of the population density (number of crystals of a given size per unit volume of rock) against the corrected crystal size length (Cashman and Marsh, 1988; Marsh, 1988). Corrected crystal size length allows the correction of the intersection-probability effect during the examination of a 2D plane (thin section) and how 3D non-spherical crystals intersect it. Higgins (2000) discussed the various methods (direct and indirect) for correcting the stereological affect and noted that Peterson (1996) first applied a correction to CSD data for the intersection-probability effect.

The CSDcorrections program also reports errors (summarized below from Higgins, 2000) on the population density for each size bin on the basis of the:

1. **Counting Error**: the square root of the number of crystals within an interval (usually only significant for larger crystal sizes where the number of crystals is smaller).
2. **Shape Probability Parameters**: applied when converting the 2D thin section to a 3D representation. The **Probability Parameters** are precisely defined for fixed convex shapes, but most crystals are irregular or have variable shapes so it is difficult to estimate the contributions from this to the total error. However, it is relatively easy to estimate the counting errors of other intersections with the plane of the thin section and assess their impact on the total correction of a given interval. This source of error is most important for small crystal sizes.
3. **Conversion to Crystal Lengths**: occurs in the conversion of intermediate (for the length of crystals that intersect the plane of the thin section) or short crystal dimensions (for intersection width measurements) to crystal lengths. This will produce systematic errors in population density and the size.

The slope of a CSD is proportional to \(-1/G\) (growth rate) multiplied by \(\tau\) (residence time) (Fig. 3a). Crystal nucleation rate generally increases with time as an initially wholly molten package of magma cools (Cashman, 1990) and will produce a linear CSD if the cooling rate remains constant (Higgins, 2000). The intersection of the CSD with the y-axis represents the nucleation density (Fig. 3a). Non-linear CSDs indicate that dynamic and/or kinetic processes affected crystallization and produced a change in cooling rates and nucleation density (Cashman, 1990; Marsh, 1998). Crystals formed at depth (“slow” cooling) carried in the initial magma may vary in size and can easily be recognized as phenocrysts crystallized from the present host magma (Marsh, 1996, 2002), whereas the groundmass phase is much smaller and formed at a “fast” cooling rate at the surface. This produces an overall “kinked” CSD for the rock (e.g., Fig. 3b), with the larger crystals (“mega-crysts” in Fig. 3b) forming the shallower sloped portion of the CSD and the smaller crystals forming the steeper sloped limb of the CSD. Kinked CSD profiles can also be achieved through magma mixing if, prior to mixing, both magmas were crystallizing the same phase at different cooling rates (Fig. 3c). Curved CSDs (Fig. 3d) may reflect the different nucleation and growth conditions of distinct crystal populations in the rock (e.g., Higgins, 1996; Marsh, 1998; Higgins and Roberge, 2003). For example, if a magma has experienced crystal fractionation, the removal of larger crystal sizes yields a concave-down CSD profile (Fig. 3d), whereas crystal accumulation would produce a concave-up CSD profile (Fig. 3d) (Higgins, 2000).

For the method presented here, samples containing a variety of crystal sizes need to be compared with each other. Therefore, only a portion of each CSD is used and that is the portion that is present in all samples. For olivine, only the portion of the CSD that is \(\leq 0.4\) mm and has a negative slope is used, omitting any change in gradient that can occur in small crystal sizes. The calculated intercept of this slope is equivalent to the nucleation density (Fig. 3a;
Table 1

Samples where plagioclase crystal size distributions have been determined for this study.

<table>
<thead>
<tr>
<th>Sample #</th>
<th>Sample Type</th>
<th>Al₂O₃ (wt.%)</th>
<th>MgO (wt.%)</th>
<th>Petrographic Reference</th>
<th>Whole Rock Reference</th>
<th>CSD Slope</th>
<th>CSD Intercept</th>
<th>Residence Time (Yrs.)</th>
<th># of Crystals</th>
</tr>
</thead>
<tbody>
<tr>
<td>12038,254</td>
<td>Fe-Diaphyic</td>
<td>12.45</td>
<td>19.2</td>
<td>[1]</td>
<td>[23]</td>
<td>-1.27</td>
<td>6.59</td>
<td>71.2</td>
<td>2198</td>
</tr>
<tr>
<td>12016,41</td>
<td>Ilmenite</td>
<td>12.65</td>
<td>8.3</td>
<td>[5]</td>
<td>[26]</td>
<td>-2.67</td>
<td>3.30</td>
<td>85.9</td>
<td>92</td>
</tr>
<tr>
<td>12051,58</td>
<td>Ilmenite</td>
<td>7.95</td>
<td>3.3</td>
<td>[2]</td>
<td>[24]</td>
<td>-2.34</td>
<td>3.96</td>
<td>10.4</td>
<td>1940</td>
</tr>
<tr>
<td>12054,126</td>
<td>Ilmenite</td>
<td>10.47</td>
<td>6.76</td>
<td>[2]</td>
<td>[25]</td>
<td>-1.98</td>
<td>2.61</td>
<td>12.3</td>
<td>563</td>
</tr>
<tr>
<td>12065,21</td>
<td>Ilmenite</td>
<td>10.3</td>
<td>8.1</td>
<td>[3]</td>
<td>[26]</td>
<td>-3.65</td>
<td>2.62</td>
<td>4.9</td>
<td>148</td>
</tr>
<tr>
<td>12063,24</td>
<td>Ilmenite</td>
<td>9.27</td>
<td>5.64</td>
<td>[2]</td>
<td>[27,28]</td>
<td>-3.92</td>
<td>5.50</td>
<td>6.2</td>
<td>658</td>
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<tr>
<td>12007,17</td>
<td>Pigeonite</td>
<td>11.28</td>
<td>5.86</td>
<td>[4]</td>
<td>[25]</td>
<td>-0.83</td>
<td>0.56</td>
<td>29.2</td>
<td>272</td>
</tr>
<tr>
<td>12017,25</td>
<td>Pigeonite</td>
<td>10.02</td>
<td>7.63</td>
<td>[4]</td>
<td>[26]</td>
<td>-5.37</td>
<td>6.27</td>
<td>4.5</td>
<td>1421</td>
</tr>
<tr>
<td>12019,5</td>
<td>Pigeonite</td>
<td>11.01</td>
<td>7.94</td>
<td>[4]</td>
<td>[26]</td>
<td>-6.19</td>
<td>7.46</td>
<td>3.9</td>
<td>872</td>
</tr>
<tr>
<td>12039,4</td>
<td>Pigeonite</td>
<td>10.52</td>
<td>5.75</td>
<td>[4]</td>
<td>[25]</td>
<td>-2.44</td>
<td>3.08</td>
<td>10.0</td>
<td>369</td>
</tr>
<tr>
<td>12045,35</td>
<td>Pigeonite</td>
<td>10.1</td>
<td>7.68</td>
<td>[4]</td>
<td>[29]</td>
<td>-2.45</td>
<td>4.78</td>
<td>10.0</td>
<td>389</td>
</tr>
</tbody>
</table>


* Represents plagioclase CSD avoiding the quench zone around xenoliths/xenocrysts.

ND = No Data; Hi-Ti = High Titanium; VLT = Very Low Titanium.

Highlighted samples are those where <250 crystals are used to construct the CSD. Samples in italics are not used because of the criteria discussed in the text and in Fig. 7.

Marsh, 1988). For plagioclase, similar criteria are followed, with the exception that the corrected crystal size range is $\geq 0.3$ mm. For both olivine and plagioclase, only consecutive data points with errors $\leq 15\%$ are used to determine the slope and intercept of the CSD in these ranges. Plagioclase is volumetrically more abundant than olivine in mare basalts and impact melts, which means fewer statistically relevant olivine CSDs can be constructed relative to plagioclase.

### 4. RESULTS

Plagioclase and olivine CSDs for the individual samples noted in Tables 1 and 2 are presented in Figs. 4 and 5, respectively. Previous studies have qualitatively shown that the plagioclase CSDs of endogenous basalts and impact melts from Apollo 14 exhibit a decreasing CSD slope from Group A to Group B to Group C and to the impact melts (Fig 4b; Oshrin, 2009; Neal et al., 2010; Hui et al., 2011). For olivine, the impact melts appear to have steeper CSDs than those from mare basalts (see Fig. 5a where the impact melts are represented by the A-14 Olivine Vitrophyres; Fagan et al., 2013a). However, it is difficult to differentiate all sample types when CSDs are presented in this way. This paper quantifies these qualitative observations to better compare the diverse CSDs shown in Figs. 4 and 5.

It has been recommended that “as a minimum, 75 crystals are required to robustly define crystal habit for CSD measurements if crystals are tabular in shape, with more acicular shapes requiring a minimum of 250 sections, suggesting a sample size of $>250$ sections to be used for shape determination in natural examples where the true 3D shape is unknown” (Morgan and Jerram, 2006). With plagioclase often exhibiting an acicular habit, there are 10 samples in our database that have plagioclase CSDs defined using $<250$ crystals (Table 1; Fig. 6). We have evaluated these CSDs in qualitative terms using the smoothness of the profile, and in quantitative terms using the errors in population density – if they are $\leq 10\%$ for at least three consecutive

### Table 2

Samples where olivine crystal size distributions have been determined for this study.

<table>
<thead>
<tr>
<th>Sample #</th>
<th>Sample type</th>
<th>$\text{Al}_2\text{O}_3$ (wt.%</th>
<th>MgO (wt.%</th>
<th>Petrography reference</th>
<th>Whole rock reference</th>
<th>CSD slope</th>
<th>CSD intercept</th>
<th>Residence time (yrs.)</th>
<th># Of crystals</th>
</tr>
</thead>
<tbody>
<tr>
<td>12004,137</td>
<td>Olivine</td>
<td>8.51</td>
<td>12.53</td>
<td>[1]</td>
<td>[14]</td>
<td>$-6.33$</td>
<td>$4.80$</td>
<td>1.59</td>
<td>143</td>
</tr>
<tr>
<td>12016,41</td>
<td>Ilmenite</td>
<td>7.23</td>
<td>12.65</td>
<td>[2]</td>
<td>[17]</td>
<td>$-8.36$</td>
<td>$6.85$</td>
<td>1.20</td>
<td>215</td>
</tr>
<tr>
<td>14321,1260</td>
<td>Group A</td>
<td>12.4</td>
<td>10.7</td>
<td>[4]</td>
<td>[4]</td>
<td>$-12.30$</td>
<td>$9.14$</td>
<td>0.82</td>
<td>133</td>
</tr>
<tr>
<td>14321,1473</td>
<td>Group A</td>
<td>11.3</td>
<td>11.0</td>
<td>[5]</td>
<td>[5]</td>
<td>$-11.91$</td>
<td>$9.09$</td>
<td>0.84</td>
<td>233</td>
</tr>
<tr>
<td>71957,12,13</td>
<td>&gt;0.4 mm</td>
<td>7.9</td>
<td>15.8</td>
<td>[6]</td>
<td>[18]</td>
<td>$-3.30$</td>
<td>$1.60$</td>
<td>3.04</td>
<td>165</td>
</tr>
<tr>
<td>71957,12,13</td>
<td>&lt;0.4 mm</td>
<td>7.9</td>
<td>15.8</td>
<td>[6]</td>
<td>[18]</td>
<td>$-13.41$</td>
<td>$5.12$</td>
<td>0.75</td>
<td>165</td>
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<tr>
<td>71048,3,6</td>
<td>Type B2</td>
<td>8.4</td>
<td>8.0</td>
<td>[7]</td>
<td>[7]</td>
<td>$-9.10$</td>
<td>$6.43$</td>
<td>1.10</td>
<td>114</td>
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<td>74235,60</td>
<td>Type A</td>
<td>8.61</td>
<td>8.35</td>
<td>[8]</td>
<td>[19]</td>
<td>$-4.61$</td>
<td>$4.81$</td>
<td>2.18</td>
<td>198</td>
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<tr>
<td>74275,312</td>
<td>Type C</td>
<td>8.72</td>
<td>10.36</td>
<td>[8]</td>
<td>[19]</td>
<td>$-12.81$</td>
<td>$7.49$</td>
<td>0.79</td>
<td>617</td>
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<td>75115,4</td>
<td>Type B2</td>
<td>8.9</td>
<td>9.3</td>
<td>[9]</td>
<td>[20]</td>
<td>$-2.32$</td>
<td>$4.81$</td>
<td>4.34</td>
<td>83</td>
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<tr>
<td>77516,30</td>
<td>Type B2</td>
<td>7.8</td>
<td>9.4</td>
<td>[9]</td>
<td>[21]</td>
<td>$-2.12$</td>
<td>$3.74$</td>
<td>4.75</td>
<td>81</td>
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<tr>
<td>79516,9</td>
<td>Type B2</td>
<td>8.4</td>
<td>8.0</td>
<td>[10]</td>
<td>[10]</td>
<td>$-5.04$</td>
<td>$4.83$</td>
<td>2.00</td>
<td>91</td>
</tr>
<tr>
<td>60003,10-16</td>
<td>High-Ti</td>
<td>5.5</td>
<td>13.0</td>
<td>[11]</td>
<td>[11]</td>
<td>$-56.68$</td>
<td>$13.80$</td>
<td>0.18</td>
<td>695</td>
</tr>
<tr>
<td>65703,9-13</td>
<td>VLT</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>$-24.91$</td>
<td>$9.97$</td>
<td>0.40</td>
<td>168</td>
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<td>12.84</td>
<td>19.2</td>
<td>[12]</td>
<td>[12]</td>
<td>$-31.80$</td>
<td>$11.13$</td>
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<td>14321,1471</td>
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<td>13.9</td>
<td>19.7</td>
<td>[13]</td>
<td>[22]</td>
<td>$-28.32$</td>
<td>$11.28$</td>
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<tr>
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<td>–</td>
<td>–</td>
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<td>$-25.07$</td>
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</table>

@ 165 represents the total number of olivine crystals used to construct the entire CSD for 71597.

**Abbreviations:** Vit. = Vitrophyre; VLT = Very-low-Ti; Ol. Vit. = Olivine Vitrophyre. Whole-rock composition for 14321,1305 taken to be the same as 14321,1158 due to textural and modal similarities (Shervais et al., 1988).


* Unclassified Apollo 17 basalt that has experienced accumulation of olivine (Warner et al., 1977). CSDs from two thin sections were combined for these samples in order to have a sufficient number of olivine crystals to make the CSD statistically relevant. The full CSD for 71597 Olivine is shown in Fig. 4b.
data points, the profile can be used. In order for plagioclase CSD profiles to be used in our classification scheme samples must pass both criteria. Of these 10 samples (highlighted in Table 1), 6 are not used any further in this study – 12016,41; 14321,1475; 14321,9057; and 14321,9090. Six samples (12056,21; 60635,2; 60639,52; 70315,27; and 71557,1) were not used.

Fig. 6. Plagioclase CSDs from 10 samples where <250 crystals were used in their construction (cf. Morgan and Jerram, 2006). Each has been evaluated in a qualitative sense (e.g., smoothness of profile) and in quantitative terms (errors calculated for population density, which need to be ≤10% for at least three consecutive data points for the profile to be used). (a) 12016,41; (b) 12056,21; (c) 14321,1475; (d) 14321,9090; (e) 14321,1608; (f) 14321,9057; (g) 60635,2; (h) 60639,52; (i) 70315,27; and (j) 71557,1. Of these 10 samples (highlighted in Table 1) only four pass both criteria and are used in this study – 12056,21; 14321,1475; 14321,9057; and 14321,9090. Six samples (12016,41; 60635,2; 60639,52; 14321,1608; 71557,1; 70315,27; 60635,2) were not used. All samples used to construct olivine CSDs (Table 2) contained >70 crystals that could be used. The smallest population used to define a plagioclase CSD is 92 (12016,41; Table 1), and for olivine it is 81 for 77516,30 (Table 2). All CSDs reported here have uncertainties that are <15%, indicating
a representative statistical population of crystals for each thin section was sampled.

Marsh (1998) concluded that igneous systems display a significant range of relations between CSD slope, maximum crystal size, and intercept. The CSD data presented here are depicted by plotting the slope of the CSD in the size ranges noted in the previous section against the intercept, and plotting the reciprocal of the residence time against the ratio of the slope to the intercept (Figs. 7 and 8). The residence time is related to the slope of the CSD (Fig. 3a) and a negative relationship should be evident from Figs. 7b and 8b. These plots show how nucleation density and residence time can be used to distinguish between impact melts and pristine mare basalts.

5. DISCUSSION

5.1. Plagioclase

The plagioclase CSD data (Fig. 4) exhibit a number of features when presented on a plot of CSD Slope vs. CSD Intercept (Fig. 7a). The Apollo 14 basalts define a distinct
field from the overlapping data from Apollo 12 and Apollo 17 basalts. Residence times, calculated using the average plagioclase growth rate in basaltic magmas at low pressure given in Burkhard (2005) of $1.304 \times 10^{-9} \text{ cm s}^{-1}$, show that the Apollo 14 high-Al basalts give increasing values (although overlapping) from Group A, through Groups B and C, to the impact melts (Table 1), supporting the qualitative observations of Oshrin (2009), Neal et al., 2010), and Hui et al. (2011). In general, the impact melts form a cluster of data that for a given CSD intercept have a lower CSD slope relative to mare basalts, which means that they have a higher nucleation density. They also have some of the highest residence times (Table 1; Fig. 7b). These relationships indicate that relative to the mare basalts, the impact melts have plagioclase on the liquidus for longer periods and have a higher nucleation density for this mineral relative to mare basalts.

Impact melt 60635 is an outlier of the impact melt field (Fig. 7a), having the shallowest slope and lowest intercept; this sample contains the largest plagioclase crystals of any impact melt in this study, suggesting it came from a slow cooling interior of a thick impact melt sheet. This is supported when the residence time is considered, as it yields a value of 140.5 years (Table 1). However, the CSD of 60635.2 does not meet the criteria outlined above for using the CSD in this study, and it is omitted from Fig. 7b, as are 12016.41, 14321,1608, 60639.52, 70315.27, and 71557.7, while they are marked with an “X” in Fig. 7a and b.

The Apollo 16 basalt fragment 65703.9-13 reported by Zeigler et al. (2006) has a relatively short residence time of 2.1 years and plots slightly above the Apollo 14 Group A basalts in Fig. 7a. It is unclear whether 65703.9-13 has a similar composition to the Apollo 14 Group A basalts, or is a VLT basalt similar to those returned by Luna 24, as proposed by Zeigler et al. (2006); the only major elements reported were TiO$_2$ (0.5 wt.%), FeO (21.4 wt.%), CaO (12.1 wt.%), and Na$_2$O (0.218 wt.%). This basalt plots close to the Apollo 14 Group A high-Al mare basalts (Fig. 7a and b) and in conjunction with olivine CSD data (see below) this sample is not designated as an impact melt.

Apollo 14 Group C basalt 14053 falls close to (but not within error of – see below) the impact melt field (Fig. 7a), thus extending the trend defined by Apollo 14 high-Al basalts by plotting at the lower CSD slope end of the trend and apart from other Group C basalts. Although geochemically classified as a Group C high-Al basalt, it has been demonstrated that 14053 is unique in that the exterior surfaces are highly reduced (Mayne and Taylor, 2003; Patchen and Taylor, 2004; Taylor et al., 2004). This was caused by exposure at the lunar surface with implantation of solar wind H, followed by metamorphism via heating from being covered with an impact ejecta blanket, before re-exposure and sampling by the Apollo 14 mission. The thin section used in this study (14053.18) was one of 12 thin sections taken from potted butt 14053.2. The original chip of 14053.2 had an exterior surface on one side and fresh interior on the other. This appears to be the “recon” chip made by the Apollo 14 Preliminary Examination Team to make the initial characterization. Subsample14053.18 was one of the last thin sections made from 2, and it is likely that it is from the exterior portion of the sample. The CSD results are consistent with this section being taken from an area that experienced thermal metamorphism via a hot impact ejecta blanket, thus promoting some textural coarsening (i.e., growth of larger crystals at the expense of smaller ones; also known as Ostwald ripening; e.g.,
Marqusee and Ross, 1984; Voorhes, 1985, 1992) that reduces the overall CSD slope and intercept and moving closer to the impact melts in Fig. 7a.

The plagioclase CSD data are also presented in terms of the reciprocal of the residence time (Table 1) plotted against the ratio of the slope to the intercept (Fig. 7b). The impact melts plot as an extension of the Apollo 14 high-Al basalt trend, but have a higher slope to intercept ratio and a longer residence time (lower reciprocal of residence time). They are distinguished from Apollo 12 and 17 basalts, which have similar residence times but have generally lower slope/intercept ratios, although some overlap does occur (Fig. 7b). The Apollo 12 and 17 basalts exhibit less well-defined trends relative to the Apollo 14 high-Al basalts and the impact melts, and generally plot below them.

Mare basalt 12007 plots as an outlier from the rest of the Apollo 12 data in Fig. 7b. The plagioclase CSD for this sample is concave-up, but has a much lower y-axis intercept and extends to much larger crystal sizes than the other Apollo 12 basalts studied here, with the possible exception of feldspathic basalt 12038 (Fig. 4a). It is hypothesized that textural coarsening has affected this sample, reducing the slope of the CSD as well as the y-axis intercept. This can occur if the temperature is kept relatively stable near the mineral liquidus (Voorhes, 1992; Higgins, 1998, 1999, 2002b; Higgins and Roberge, 2003), which in turn implies that 12007 represents the interior of a relatively thick lava flow or sill. This is in agreement with its petrographic description as a relatively coarse grained microgabbro (e.g., Baldridge et al., 1979).

5.2. Olivine

The olivine CSDs for impact melts and endogenous mare basalts behave in the opposite sense to plagioclase in that those from impact melts are steeper than for mare basalts (Fig. 5a). The number of samples in this part of the study is lower than for plagioclase because mare basalts and impact melts contain fewer olivines; this means fewer statistically relevant olivine CSDs can be constructed. In a plot of CSD Slope vs. CSD Intercept (Fig. 8a), the impact melts and mare basalts plot in distinct fields. Here, two Apollo 16 basalts (65703,9-13 and 60603,10-16) reported by Zeigler et al. (2006) are shown. Sample 65703,9-13 plots with the Apollo 14 Group A high-Al mare basalts (as with plagioclase in Fig. 7a), whereas 60603,10–16 extends the impact melt field to steeper negative slopes and higher y-axis intercepts (Fig. 8a).

Olivine residence times have been calculated in a similar way to those for plagioclase but using the olivine growth rate defined by Borell and Kilinc (2012) of $3.1623 \times 10^{-9}$ cm s$^{-1}$ (Table 2). The mare basal olivines have the longest residence times (0.59–4.75 years) and the impact melts plus the Apollo 16 basalts 60603,10–16 have the shortest (0.2–0.4 years) (Fig. 8b). As shown in Figs. 5a and 8a, the impact melts have higher intercept (i.e., nucleation density) values and steeper CSD slopes than the mare basalts. The two Apollo 16 basalts (65703,9-13 and 60603,10-16) plot with the mare basalts and impact melts, respectively, in Fig. 8a and 8b, suggesting similar crystallization conditions. In Fig. 8b, the mare basalts and impact melts form a linear trend with the only exception being the olivine cumulate 71597 that falls below this trend. Conversely to the similar plot for plagioclase (Fig. 7b) the impact melts have lower olivine CSD slope/intercept ratios and shorter residence times relative to the mare basalts, although they have higher nucleation densities (Fig. 8a).

Apollo 17 high-Ti basalt 71597 is unique among the Apollo 17 mare basalt suite. It has experienced significant olivine accumulation in a high-Ti mare basalt flow.
Whole rock major and trace element analyses and major element mineral compositions indicate that 71597 experienced up to 24–27% olivine and possible minor ilmenite accumulation (Warner et al., 1977). There is a bimodal distribution of olivine Fo-content between large (~2 mm to >5 mm) subequant olivine crystals (Fo73–79) and smaller (0.1–0.5 mm) “matrix” olivine crystals (Fo63–69). The olivine CSD demonstrates this multi-modal distribution (Fig. 5b) and possibly indicates there are three groups of olivine crystals: <0.5 mm, 0.5 to 2.2 mm, and >2.2 mm; however, errors on the data points defining the larger group are beyond the 15% limit and the largest size bins could represent a continuation of the 0.5–2.2 mm trend (Fig. 5b). Therefore, 71597 is represented as two connected points on olivine CSD plots (Fig. 8a and b) solely order to demonstrate these differences. When CSD slope and intercept are considered, both 71597 crystal populations plot within the mare basalt field (Fig. 8a).

5.3. Errors and limitations

The errors on each data point in Figs. 7a and 8a have been calculated by CSDCorrections (Higgins, 2000, 2002a,b; see Section 3) and error bars are presented for all data points in these diagrams. Data apparently free of error bars mean that the errors are within the size of the symbols. Inclusion of errors demonstrates that the CSD data for impact melts are distinct from those of pristine mare basalts both for olivine and plagioclase besides the stated exceptions. In addition, the CSDs for the impact melts and each mare basalt suite were constructed by six different people (coauthors on this paper), demonstrating that there is relative consistency in the way CSDs have been constructed. To illustrate this, two separate CSDs from 12038 were constructed by different people. Slopes and intercepts were calculated over approximately the same size interval (0.3–1.9 mm). Calculated slopes were -2.028 ± 0.003 and 2.180 ± 0.002, with intercepts of 4.26 ± 0.1 and 4.16 ± 0.06. On this basis, we submit that CSD data carefully derived from different analysts can be used to distinguish between lunar basalts derived from endogenic igneous processes and impact melt without the introduction of unacceptable errors.

Another source of error could be induced because a thin section represents a 2D slice through a 3D rock sample and one thin section may not represent the true CSD (see description in the Section 3). Therefore, plagioclase CSDs were constructed from two different thin sections of one sample, impact melt 67559 (.1 and .28). The results indicate that the method used here yields data that are within error of each other when two sections of the same sample are examined (Fig. 9; Table 1). This allows the combination of data from two thin sections to achieve a statistically representative crystal population, which was done for 71597, a high-Ti mare basalt, by examining two thin sections (.12 and .13; Fig. 5b).

Impact melts can be heterogeneous in nature with cooling rates highly variable at the centimeter scale if xenoliths and xenocrysts are present and have not been fully melted. Cooling rate can have a significant effect on the slope of a CSD. In order to investigate this potential complication with the quantitative petrography method described in this paper, sample 60235 was examined in detail through analysis of thin section 60235,17 (Fig. 10).

Fig. 9. Duplicate plagioclase CSDs from impact melt 67559 taken from thin sections .1 and .28. Both profiles are within error of each other giving confidence that the profiles taken from a single thin section of a given sample represent the crystal size distribution of the sample.

Fig. 10. Photomicrograph (in plane-polarized light) of thin section 60235,17. Note the presence of a microbreccia xenolith and two plagioclase xenoliths. There is a slight reduction in grain size of the impact melt immediately adjacent to the xenolith.
(1980) described 60235 as a plagioclase-rich impact melt containing clastic material consisting of plagioclases and plagioclase-rich breccias. This description is supported by thin section 60235,17 (Fig. 10). Crystal size distributions were constructed for all plagioclases in the melt portion of the thin section, represented by 60235,17 on Fig. 7 a and b. This CSD plots within the Apollo 14 high-Al basalt field. A second plagioclase CSD was constructed omitting the plagioclases in a 0.5 mm zone around the xenolith as the grain size diminishes slightly presumably due to the cooling effect of the xenolith. This CSD is also plotted on Fig. 7 a and b as 60235,17 (No Xeno) and falls within the impact melt field. This demonstrates that impact melt cooling rates (and the slope of the plagioclase CSD profile) is influenced by the presence of xenoliths. Using the plagioclase CSD method to distinguish impact melts that contain xenoliths should be used with caution.

5.4. Influence of bulk composition

The samples analyzed in this study show distinct CSD slopes, intercepts, and residence times between impact melts and pristine mare basalts (Figs. 7 and 8). This suggests distinct crystallization regimes, which could be influenced by bulk composition. Fig. 11 is a plot of whole rock MgO (wt.%) vs. Al₂O₃ (wt.%) for all samples studied here, as they are dominated by olivine and plagioclase, respectively. The whole rock data used have the following caveats:

- Olivine vitrophyre (impact melt) 14321,1486 has no whole-rock data reported in the literature. Fagan et al. (2013a) described this as an impact melt on the basis of petrography and mineral chemistry.
- Olivine vitrophyre (impact melt) 14321,1305 (equivalent whole rock aliquot = 14321,1180) was reported by Shervais et al. (1988), but only whole-rock FeO, CaO, Na₂O, and Cr₂O₃ were reported for 1180. However, the values for these elements are similar to those reported for olivine vitrophyre 14321,1158 (Shervais et al., 1988) whose Al₂O₃ and MgO values are adopted here to represent 1305.
- No Al₂O₃ or MgO data were reported for 65703,9-13 (Zeigler et al., 2006).
- No MgO values have been recorded for impact melt 14073, but its Al₂O₃ content (20.8 wt.%; Laul et al., 1972) is similar to that in 14310 (20.74 wt.%; e.g., Philpotts et al., 1972), thus the same MgO value is used for 14073 (8 wt.%).

It is evident that the impact melts have distinct compositions, being either higher in Al₂O₃ or MgO (Apollo 14 olivine vitrophyres) than the pristine mare basalts (Fig. 11). Higher Al₂O₃ would enable plagioclase to be on the liquidus for a longer period of time, which is consistent with the impact melts having shallower CSD slopes and longer residence times than the mare basalts (Fig. 7). However, the shallower CSD slope may also reflect a slower...
cooling rate for the impact melts relative to the mare basalts. The field of impact melts in Fig. 9 appears to form a negative correlation and suggests that bulk composition is controlling the texture. However, for olivine the impact-generated Apollo 14 olivine vitrophyres have the highest MgO, which suggests that olivine is on the liquidus for longer. This should result in shallower olivine CSD slopes and longer residence times than the mare basalts (as seen with plagioclase), but the opposite is true. In addition, Apollo 16 basaltic sample 60603,10-16 (Zeigler et al., 2006) has an olivine CSD similar to the impact melts (Fig. 8), yet its whole-rock composition is similar to the mare basalt field (Fig. 11), suggesting that whole rock composition is not controlling the olivine CSD profile.

Therefore, it is concluded that the mode of origin and cooling rate are the main factors in olivine textural development. Conversely, bulk composition may have some control over the plagioclase CSD, but our dataset suggests that mode of origin and cooling rate are also very important.

6. SUMMARY

This quantitative petrography study of lunar basaltic rocks has shown that the suite of impact melts and endogenous mare basalts analyzed as part of this study can be distinguished on the basis of plagioclase and olivine textures by using CSDs. This distinction is made using the slope and intercept of specific crystal size ranges of the CSD: $\geq 0.3$ mm for plagioclase; $\leq 0.4$ mm for olivine. This contradicts the findings of Higgins (2002a,b), who stated that correlation of CSD slopes and intercepts was not significant, if the volumetric phase portion is constant. The two origins are distinguishable using this method.

Impact melts have plagioclase and olivine CSDs that appear to give conflicting information; plagioclase impact melt CSDs generally have shallower slopes and relatively large crystal sizes relative to the mare basalts, while olivine CSDs have steeper slopes for impact melts and relatively small crystal sizes (Figs. 4 and 5). Olivine and plagioclase CSDs have been constructed for four mare basalt samples (12016,4i; 14321,1473; 14321,1482; 65703,9-13) and confirm the dichotomous behavior (compare Figs. 7a and 8a). This could not be confirmed with the impact melt samples studied here as no impact melt sample permitted construction of CSDs for both plagioclase or olivine.

The dichotomous behavior between olivine and plagioclase CSDs for the mare basalts and impact melts does not appear to be controlled primarily by bulk composition. For example, mare basalt 14053 plots close to the impact melts in Fig. 7a and b, but bulk compositions appear to be distinct (Fig. 11). Apollo 16 basalt 60603,20-16 plots with the impact melts on the basis of olivine CSD data, and 65703,9-13 plots with the mare basalts on the basis of both olivine and plagioclase CSD data. This is consistent with the findings of Donohue et al. (2013, 2014) who used mineral chemistry to define the origins of these samples.

On the basis of the quantitative petrography presented in this paper, a classification scheme that defines lunar impact melts from mare basalts is shown in Fig. 7a for plagioclase and Fig. 8a olivine CSD data. The textural classification is consistent with siderophile element abundances for the samples analyzed here (Fig. 2a and b). While further data will update the limits of these fields, the current dataset supports the initial classification of lunar basaltic samples on the basis of quantitative petrography.

6.1. Importance of this study

It is unlikely that, in the foreseeable future, hand-specimen sized extraterrestrial samples will be returned from the Moon or any other extraterrestrial planetary body. Therefore, planetary scientists must learn to work with smaller and smaller sample sizes. Destructive analyses (e.g., HSE) are not ideal methods to test if a lunar sample is of impact origin or is a product of partial melting of the lunar interior. For example, South Pole-Aitken basin sample return, with the goal of collecting SPA impact melt to age-date the basin event, is an identified mission in the latest decadal survey for NASA’s Planetary Science Division (National Research Council, 2011). The current plan for this sample return mission is to return $\sim 2$ kg of 2–4 mm fragments high-graded from lunar regolith. Thus, it is critical to the success of such a mission that we be able to reliably distinguish between impact melts and pristine mare basalt samples. The small sample sizes would negate HSE determinations, so if a distinction can be made on the basis of petrography, more sample would be available for other studies, including geochronology. The quantitative petrography method for plagioclase and olivine described here shows promise in being able to distinguish between these two types of igneous rock and allow such a mission to achieve its science goals.

The quantitative petrography method will also open up a wealth of new research possibilities in the existing lunar sample collection. Re-examination of the 2–4 mm soil fragments returned by the Apollo and Luna missions using this method could help refine mare basalt volcanism as well as impact events. Examination of small clasts from returned breccia samples will also be possible in order to determine their impact vs. pristine origin. In essence, the quantitative petrography method can determine the origin of a clast while maximizing the amount of remaining sample for other studies.

7. CONCLUSIONS

The quantitative petrography method presented in this paper shows promise in distinguishing impact melts from endogenous melts of the lunar interior (i.e., mare basalts) using just a thin section. It first requires the construction of crystal size distributions for olivine and/or plagioclase. Analysis of known lunar impact melts and pristine mare basalts demonstrates that, at least for the samples studied here, the two origins are distinguishable using this method.
(one sample, 14053 shows ambiguities in its texture that are related to post-eruption thermal metamorphism via a hot impact ejecta blanket). Specifically, olivine from impact melts displays a steeper CSD relative to olivine from mare basalts. For plagioclase, generally impacts melts display CSDs with shallower gradients than those from endogenous mare basalts and, as for olivines, plot in a distinct field on a slope vs. intercept plot. Using just a thin section to distinguish impact melts from endogenous mare basalts enables the goals of future robotic sample return missions that will potentially return small (2–4 mm) “rocklets” for analysis.

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