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1 2	Characterization of mesostasis regions in lunar basalts: Understanding late-stage melt evolution and its influence on apatite formation
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5 6 7 8 9 10 11 12 13 14	 ¹Planetary and Space Sciences, The Open University, Milton Keynes, MK7 6AA, UK ²Faculty of Earth and Life Sciences, VU University Amsterdam, 1081 HV Amsterdam, NL ³School of Earth, Atmospheric and Environmental Sciences, University of Manchester, UK ⁴Institut de Minéralogie, de Physique des Matériaux et de Cosmochimie, Muséum National d'Histoire Naturelle, Sorbonne Universités, CNRS, UPMC & IRD, 75005 Paris, France ⁵Department of Earth Sciences, The Natural History Museum, London, SW7 5BD, UK *Present address: School of GeoSciences, University of Edinburgh, Edinburgh, EH9 3JW (nicola.potts@ed.ac.uk)
15	Recent studies geared towards understanding the volatile abundances of the lunar interior have focused on the
16	volatile-bearing accessory mineral apatite. Translating measurements of volatile abundances in lunar apatite into
17	the volatile inventory of the silicate melts from which they crystallized, and ultimately of the mantle source regions
18	of lunar magmas, however, has proved more difficult than initially thought. In this contribution, we report a
19	detailed characterization of mesostasis regions in four Apollo mare basalts (10044, 12064, 15058, 70035) in order
20	to ascertain the compositions of the melts from which apatite crystallized. The texture, modal mineralogy, and
21	reconstructed bulk composition of these mesostasis regions vary greatly within and between samples. There is no
22	clear relationship between bulk-rock basaltic composition and that of bulk-mesostasis regions, indicating that
23	bulk-rock composition may have little influence on mesostasis compositions. The development of individual melt
24	pockets, combined with the occurrence of silicate liquid immiscibility, exerts greater control on the composition
25	and texture of mesostasis regions. In general, the reconstructed late-stage lunar melts have roughly andesitic to
26	dacitic compositions with low alkali contents, displaying much higher SiO2 abundances than the bulk
27	compositions of their host magmatic rocks. Relevant partition coefficients for apatite-melt volatile partitioning

- 28 under lunar conditions should, therefore, be derived from experiments conducted using intermediate compositions
- 29 instead of compositions representing mare basalts.

34 1. Introduction

In order to constrain the volatile inventory of the lunar interior many studies have focused on 35 measuring structurally bound OH in apatite [Ca₅(PO₄)₃(F,Cl,OH)] from Apollo and lunar meteorite 36 samples (e.g. Boyce et al. 2010; McCubbin et al. 2010a, 2010b, 2011; Barnes et al. 2013, 2014; Tartèse 37 38 et al. 2013b, 2014a, 2014b). Yet, translating volatile measurements in apatite to volatile abundances in co-existing melt has proved troublesome, leading many studies to focus on improving understanding 39 40 on the partitioning behavior of volatiles between apatite and melt (Boyce et al. 2014; McCubbin et al. 41 2015a). Few studies, however, have focused on constraining the petrographic context in which apatite forms in lunar melts. Apatite begins to crystallize after ~95 % melt solidification in typical mare 42 magmas (McCubbin et al. 2010b), but is likely to start crystallizing earlier (~85 % melt solidification) 43 in KREEP (enriched in incompatible elements such as K, REE and P) basalts (Tartèse et al. 2014a). 44 45 Apatite, therefore, does not form from bulk basaltic melts but instead forms from differentiated latestage melts, which are represented by mesostasis regions within lunar samples (Henderson 1970). 46 47 Although mesostasis regions have been observed within many Apollo basalts and lunar basaltic 48 meteorites, few studies have focused on constraining the silicate melt compositions from which they 49 formed. To provide better understanding on the bulk composition of melts from which lunar apatite 50 crystallized, we have characterized the mineralogy and geochemistry of apatite-bearing mesostasis 51 areas in four Apollo mare basalts (samples 10044, 12064, 15058, and 70035) representative of the high-52 Ti and low-Ti mare basalts. Knowledge of the melt-compositions from which apatite crystallized will 53 inform experimental studies aiding in the determination of appropriate apatite-melt partition 54 coefficients of volatile elements in geochemical models of lunar magma evolution.

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57 2. Samples

58 *2.1. 10044*

Lunar sample 10044 is a high-Ti ilmenite basalt with a low-K content (Beaty and Albee 1978). It
is a subophitic basalt (James and Jackson 1970) that is sometimes described as a microgabbro
(Anderson et al. 1970). A similarity between 10044, 10047, and 10058 has been noted (Beaty and Albee

62 1978) with a suggestion that these samples could be fragments of a larger block. Geochronological studies carried out on this sample yielded Rb-Sr dates of ~3.7 Ga (Papanastassiou et al. 1970), ⁴⁰Ar/³⁹Ar 63 dates of 3.73 ± 0.04 Ga (Turner 1970; Guggisberg et al. 1979) and a tranquillityite ²⁰⁷Pb/²⁰⁶Pb date of 64 3.72 ± 0.01 Ga (Tartèse et al. 2013a). Cosmic-ray exposure (CRE) ages of ~70 Ma were determined 65 using the abundance of cosmogenic ⁸¹Kr (Hohenberg et al. 1970), ¹²⁶Xe (Srinivasan 1974) and ³⁸Ar 66 (Guggisberg et al. 1979). 10044 contains subhedral to anhedral zoned pyroxene phenocrysts (~45% 67 modal abundance) surrounded by a matrix of subophitic plagioclase (\sim 35%), and ilmenite (\sim 13%) 68 (McGee et al. 1977). Mesostasis areas in this sample are composed of silica (\sim 7%), troilite (\sim 0.5%) 69 associated with metallic iron, K-rich glass, and accessory minerals including apatite (~0.1%) and Zr-70 rich minerals such as baddeleyite and tranquillityite (McGee et al. 1977). We have examined mesostasis 71 72 areas in thin section 10044,645, which is 6×10 mm in size. The mineralogy of this section is consistent 73 with that of the main rock sample described above, containing anhedral pyroxene phenocrysts ($\sim 1 \times 2$ mm), plagioclase laths ($\sim 1 \times 0.2$ mm) and ilmenite ($\sim 3 \times 0.5$ mm). Multiple mesostasis regions are 74 found throughout the 10044,645 section and are further described in section 4.1. 75

76

77 2.2. 12064

78 Sample 12064 is a coarse-grained low-Ti ilmenite basalt that displays a subophitic texture 79 containing anhedral pyroxene crystals (~56% modal abundance) intergrown with anhedral plagioclase 80 laths (~29%) (Klein et al. 1971). It is characterized by coarse-grained mesostasis areas (~9%) that 81 contain fayalite, Fe-rich pyroxene, troilite, K-feldspar, K-rich glass, Zr-rich phases, merrillite and apatite (Kushiro and Haramura 1971). Plagioclase compositions range from An₉₃ to An₈₆ while 82 pyroxene compositions are close to the Fe-rich end-member hedenbergite (Kushiro and Haramura 83 1971). The sample also contains coarse-grained laths of ilmenite (3.5%) associated with ulvöspinel 84 (2%). Small, sporadic grains of pyroxferroite are observed coexisting with clinopyroxene (Klein et al. 85 1971). Rb-Sr (Papanastassiou and Wasserburg 1971) and ⁴⁰Ar/³⁹Ar (Horn et al. 1975) dating studies for 86 sample 12064 yielded dates of 3.18 ± 0.09 Ga and 3.18 ± 0.01 Ga, respectively, which have been 87 interpreted as crystallization ages. An exposure age of 255 Ma has been reported by Horn et al. (1975), 88 while exposure ages determined from abundances of cosmogenic ³He, ²¹Ne, and ³⁸Ar cluster around 89

90 200 Ma (Hintenberger et al. 1970). We have examined mesostasis areas in thin section 12064,130, 91 which is 8×4 mm in size. The mineralogy of the studied sample is consistent with modal abundances 92 reported for the bulk rock, containing anhedral pyroxenes (> 2 × 2 mm), intergrown with anhedral 93 plagioclase (~2 × 1 mm) and with minor ilmenite (~1 × 0.2 mm). Multiple mesostasis regions are found 94 across the 12064,130 section and are further described in section 4.1.

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96 2.3. 15058

Apollo 15 sample 15058 is one of the largest basalts returned from the Moon. It is a coarse-grained 97 low-Ti pigeonite basalt (Ryder 1985). Olivine ($\sim 2\%$ modal abundance) in this sample is found only in 98 the cores of large pigeonite grains (\sim 71%). This sample also contains radiating plagioclase (\sim 24%) 99 clusters (Butler 1971). Dates determined for sample 15058 range from 3.36 ± 0.03 Ga (40 Ar/ 39 Ar; 100 101 Husain 1974) to 3.46 ± 0.04 Ga (Rb/Sr; Birck et al. 1975). 15058 is characterized by a multistage exposure history with increased ⁸¹Kr near the surface of the sample but constant at depth (Eugster et al. 102 1984). We have studied mesostasis areas in thin section 15058,20, which is 6×8 mm in size. The 103 104 mineralogy of this section, consistent with that of the main rock sample, comprises anhedral pyroxene 105 phenocrysts (> 2 × 2 mm), plagioclase (\sim 1 × 0.5 mm) and minor ilmenite (< 0.5 × 0.2 mm). Mesostasis 106 regions in 15058,20, which are further described in section 4.1, are observed mostly in a few clusters 107 in the center of this thin section.

108

109 *2.4.* 70035

Apollo 17 sample 70035 is a medium-grained, vesicular, high-Ti basalt (Ridley and Brett 1973). It 110 is a large crystalline basalt with large (>1mm) anhedral clinopyroxene crystals (~46% modal 111 abundance) enclosing armalcolite, ilmenite, and spinel (~22%). Interstitial plagioclase (~26%) is found 112 together with ilmenite and olivine (~2.5%) (Weigand 1973). The mesostasis regions (~2%) include 113 silica, K-feldspar, tranquillityite, ilmenite, ulvospinel, troilite, K-rich glass and apatite (Papike et al. 114 1974; McGee et al. 1977). Metallic iron is found in the cracks and cleavage of opaque oxides and 115 silicates in this sample (El Goresy and Ramdohr 1975). Plagioclase composition in sample 70035 varies 116 117 from An₈₈ to An₈₃ (Weigand 1973). The Rb/Sr dates of 3.82 ± 0.06 Ga and 3.73 ± 0.11 Ga determined

for 70035 are consistent with each other (Evensen et al. 1973; Nyquist 1977) and with a 40 Ar/ 39 Ar date of 3.75 ± 0.07 Ga (Stettler et al. 1973). Exposure ages determined from abundance of cosmogenic 38 Ar (Stettler et al. 1973) and 81 Kr (Drozd et al. 1977) are 95 to 100 Ma and 122 ± 3 Ma, respectively. We have studied mesostasis areas in thin section 70035,195. This section is 14 × 3 mm in size and has a mineralogy consistent with that of the main rock sample, comprising anhedral plagioclase phenocrysts (> 2 × 1 mm), pyroxene (~1 × 1 mm) and ilmenite (~0.5 × 0.2 mm). Small mesostasis pockets are found throughout the studied section and are further described in section 4.1

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126 **3.** Methods

127 *3.1. Identifying mesostasis regions*

128 The aim of this study was to characterize the regions in which apatite is found in lunar basalts, 129 therefore, mesostasis regions containing apatite were primarily investigated. Using whole section element maps of the studied samples, mesostasis regions were identified by the presence of small (< 1130 mm) anhedral and amorphous phases that crystallized between larger surrounding crystals. The 131 132 presence of symplectite assemblages and/or silicate liquid immiscibility textures was also used to identify mesostasis regions. These regions are rich in fayalite, silica, and glass and host a wide variety 133 134 of smaller phases (i.e. sulphides, phosphates) compared to the surrounding major minerals. Pernet-Fisher et al. (2014) suggested that coarse-grained silica in mare basalts (> 100 µm) is likely a pseudo-135 eutectic mineral and, therefore, should not be included as a mesostasis phase. For the majority of 136 mesostasis regions investigated here this is not a concern as silica within the regions is fine-grained 137 $(<100 \,\mu\text{m})$, and intergrown with other mesostasis phases. For mesostasis regions containing large $(>100 \,\mu\text{m})$ 138 µm), elongate silica grains, intergrown with other mesostasis phases, we have considered this as a 139 product of crystallization of the late-stage melt and, therefore, as part of the mesostasis region. In the 140 141 basalts studied here silica is only found associated with mesostasis regions.

A large variety of textures and mineral phases were observed in the different mesostasis regions.
We determined the two-dimensional extents of the different regions in multiple ways. For silica-rich
mesostasis regions the boundary was drawn around the area including silica. Pernet-Fisher et al. (2014)

145 proposed that up to 30 vol.% of mesostasis melt could have been equilibrated with larger surrounding phases. Re-equilibration of pre-existing plagioclase and pyroxene grains with mesostasis melt modified 146 their chemistry and created chemical zonations. In some mesostasis regions clear modification of pre-147 existing pyroxene associated with overgrowth of mesostasis phases was observed on back-scattered 148 149 electron (BSE) and false-color X-ray images, and later confirmed with electron probe microanalysis (EPMA). In regions with no clear modification of pre-existing phases, mesostasis boundaries were 150 drawn around mesostasis phases only. Compositional differences between phases within and outside 151 mesostasis regions are detailed in section 5. For modal abundance estimates, where possible, mesostasis 152 regions were limited to areas that included late-stage phases with limited incorporation of surrounding 153 pre-existing phases. It is worth noting that thin sections only provide 2D sections through 3D mesostasis 154 regions, which likely implies a sampling bias. To try and mitigate the possible effects associated with 155 this sampling bias, we analyzed multiple mesostasis regions $> 50 \times 50$ µm. 156

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158 *3.2. Electron Microscopy*

The BSE maps were collected using the FEI Quanta 3D dual beam scanning electron microscope (SEM) at The Open University. The instrument is fitted with an Oxford Instruments 80 mm X-MAX energy dispersive X-ray detector, which allowed X-ray maps of each sample to be obtained by using the SEM in Energy Dispersive Spectroscopy (EDS) mode. Elemental mapping was carried out using a 20 kV accelerating voltage and a 0.60 nA beam current. X-ray maps were acquired at resolutions of 512 × 448 and 1024 × 896 pixels with a magnification of 200 to 300.

Quantitative mineral compositions were determined using a CAMECA SX-100 Electron Probe Micro Analyzer (EPMA) at The Open University. An accelerating voltage of 20 kV and a beam current of 20 nA were used except for plagioclase and glass analyses where the beam current was reduced to 10 nA. The beam diameter varied from 1 μ m to 10 μ m, depending on the dimensions of the analyzed phase. Standard count rates of 20 to 40 s per element were used, with a background counting time of half the peak counting time before and after peak analysis. A selection of natural standards were used for calibration, including feldspar (Si, Al, K), jadeite (Na), forsterite (Mg), hematite (Fe), rutile (Ti), 172 and apatite (P). The apatite compositions used here for bulk mesostasis composition calculations are 173 those reported by Tartèse et al. (2013b) for apatite grains located in the same thin sections but not necessarily in the same mesostasis regions. For 70035, the average apatite composition of those 174 analyzed in the other samples by Tartèse et al. (2013b) was used as these authors did not analyze apatite 175 176 in this sample. The modal mineralogy and the quench glass abundances, for each mesostasis area, were calculated using pixel histograms from BSE-images in the ImageJ[®] software. For each mesostasis 177 region a bulk composition was calculated using average EPMA compositions for each phase and the 178 modal mineralogy within individual regions. 179

180

181 4. Results

182

183 The BSE images and false-color compositional maps of representative mesostasis regions for

4.1. Mesostasis textural descriptions

the four samples are shown in Figures 1 to 8, and the modal mineral abundances in Table 1.

185 In sample 10044,645, eight mesostasis regions were analyzed, and representative BSE images 186 of some of these regions are shown in Figure 1, while Figure 2 displays false-color X-ray elemental maps combining Si, Ca and Fe. These regions are $\sim 200 \times 200 \,\mu\text{m}$ and are located between pre-existing 187 plagioclase laths (Figs. 1B & 1C) and pyroxene phenocrysts (Figs. 1A, 1C, & 1D). All of the regions 188 189 contain apatite, pyroxene and ilmenite, while silica, plagioclase, K-glass, and K-feldspar were found in 190 the majority of areas. Two mesostasis regions contained favalite grains large enough (i.e. $>10 \mu$ m) for multiple microprobe spot analyses (Figs. 1A & 1D). Symplectite texture (labelled as 'pxf' in 191 192 accompanying figures), formed from the breakdown of pyroxferroite into fayalite, silica, and 193 hedenbergite, was observed in two regions within 10044,645 (Figs. 1D/2C & 2F). Two regions within 194 this sample contained coarse-grained silica (Figs. 1B & 2D). Spheroidal textures comprising of Si-K-Ba-rich glass (hereafter referred to as K-glass) enclosed by fayalite, characteristic of silicate-liquid 195 196 immiscibility (SLI) (Pernet-Fisher et al. 2014), were observed in five of the regions and shown in Figure 197 1D. Such textures were described by Pernet-Fisher et al. (2014) as 'sieve' textures and classified as mature (large glass droplets) or immature (small glass droplets). The mesostasis region in Figure 1D 198 displays immature 'sieve' texture, with fine-grained droplets of K-glass trapped within host fayalite. 199

Seven mesostasis regions within basalt 12064,130 were analyzed. These regions are $\sim 100 \times$ 100 µm and found mostly between pyroxene phenocrysts (Figs. 3A, 3C, & 3D) and plagioclase laths (Fig. 3B). Mesostasis regions contain varying amounts of apatite, silica, pyroxene, K-feldspar, fayalite, and ilmenite (Figs. 3 & 4). Pyroxferroite breakdown products are present in some regions (Figs. 3B, 4B, 4C, 4D & 4F). Both mature (Fig. 3A) and immature (Figs. 3B & 3D) 'sieve' textures, indicative of SLI, were observed in all but two mesostasis region of 12064 (Figs. 3C & 4D). One region within 12064 contained both mature and immature SLI textures (Fig. 3A & 4A).

In sample 15058,20, six mesostasis regions were analyzed. These regions are $\sim 300 \times 300 \,\mu\text{m}$ and found between pre-existing plagioclase (Fig. 5B) and pyroxene (Figs. 5A & 5C). Most of these regions contain a high abundance of silica (Figs. 5A, 5B, 6A, 6C, 6E & 6F). Five of the regions contained apatite, pyroxene, plagioclase, and ilmenite, while one area did not contain apatite. K-glass was found in some of these regions. Textures indicative of SLI or pyroxferroite breakdown products were not observed in this sample.

Seven mesostasis regions within sample 70035,195 were analyzed. The mesostasis regions in this sample are $\sim 100 \times 100 \mu$ m, and found between pre-existing pyroxene and plagioclase grains, and they generally contain silica, pyroxene, plagioclase, ilmenite, and K-glass (Figs. 7 & 8). These regions contain small (< 2 to 10 µm) apatite crystals co-existing with merrillite grains (e.g., Fig. 8A). Out of the nineteen areas initially investigated we found that most of the regions contained merrillite. Apatite was not widespread but instead found concentrated within a few of the mesostasis regions.

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4.2. Composition of mesostasis phases

For all four samples plagioclase compositions vary from An_{93} to An_{68} (Fig. 9A). The K-rich plagioclase feldspars are those firmly within mesostasis regions (Fig. 9A). Most of the pyroxene analyses in samples 10044 and 12064 plot close to the Hd – Fs binary (Fig. 9B). In contrast, all pyroxene analyses for 70035 are more Mg-rich, plotting towards the En and Di regions (Fig. 9B). The few analyses of pyroxene compositions in 15058 range between those in 70035 and in samples 10044/12064(Fig. 9B). 227 Glass compositions from mesostasis regions are displayed in a Total-Alkali-Silica (TAS) diagram in Figure 10, together with compositions of the bulk-rock mare basalts studied and of lunar granites 228 (Kushiro and Haramura 1971; Rhodes and Hubbard 1973; Rhodes et al. 1976; Beaty and Albee 1978; 229 Seddio et al. 2013). In general, the glass compositions plot in a similar region of the diagram to the 230 231 lunar granites (~4 - 10 wt.% alkali and ~68 wt.% to 78 wt.% SiO₂).

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4.3 Reconstructed bulk mesostasis compositions

Average modal abundances for mesostasis regions in the four studied samples are given in Table 1, 234 while average compositions of mesostasis mineral and glass phases are given in Tables 2 - 5. These 235 236 data were combined to calculate the bulk mesostasis compositions, and average compositions for the 237 latter are given in Table 6. Compositional variations between different mesostasis regions within 238 individual samples are generally larger than variations between different samples. Bulk SiO₂ contents within 10044 mesostasis regions, for example, varied from 57 wt.% to 83 wt.%, while SiO₂ contents 239 varied by about 12 wt.% across the different mesostasis regions in 70035. Large variations between 240 241 bulk compositions of mesostasis regions were also observed for TiO_2 in sample 70035 for example, which varied by 14 wt.%. Significant variations in CaO (6.2 wt.% difference) contents in sample 12064 242 243 were also observed between different mesostasis regions. Very little variation within regions was 244 observed for MnO, Na₂O, K₂O, and P₂O₅ contents, which is related to the lower abundance of these oxides in the calculated meostasis compositions. 245

The reconstructed bulk mesostasis compositions display large variations between each sample. The 246 largest variation is observed for SiO₂, which varies from 70 wt.% (10044) to 56.2 wt.% (70035). There 247 is no correlation between the reconstructed SiO₂ content of the mesostasis regions and the SiO₂ content 248 of the bulk sample. Sample 70035 does have the lowest bulk-rock SiO₂ content (37.8 wt.%; Rhodes et 249 al., 1976) out of the four samples studied here, but 10044 has a lower bulk-rock SiO₂ content (42.2 250 wt.%; Beaty and Albee 1978) than both 12064 (46.0 wt.%; Scoon, 1971) and 15058 (47.8 wt.%; Rhodes 251 and Hubbard, 1973) and, yet, the highest bulk-mesostasis SiO₂ content. The reconstructed mesostasis 252 compositions for the four samples also display large variations in TiO₂ content. Sample 70035, a high-253 254 Ti basalt, has the highest mesostasis TiO₂ content (7.0 wt.%), however, high-Ti basalt 10044 has a 255 relatively low mesostasis TiO₂ content (1.5 wt.%). TiO₂ contents of mesostasis regions in low-Ti basalts 12064 and 15058 are 0.9 wt.% and 3.6 wt.%, respectively. FeO contents of mesostasis regions in 256 samples 10044 (10.8 wt.%), 15058 (11.1 wt.%) and 70035 (11.2 wt.%) are fairly similar, while it is 257 much higher in 12064 (23.0 wt.%). The CaO contents of bulk mesostasis are similar in samples 10044 258 259 and 15058 (6.4 wt.% CaO), while bulk mesostasis in 12064 and 70035 have varying CaO contents (5.7 - 8.8 wt.% CaO). The Al₂O₃ contents of mesostasis regions in 10044, 15058, and 70035 are similar 260 (9.3, 9.2, and 10.1 wt.%), while 12064 has a lower Al₂O₃ content of 4.1 wt.%. The MgO contents of the 261 mesostasis regions are highly variable between the four samples, ranging between 4.9 wt.% (70035) 262 and 1.7 wt.% (15058), down to 0.8 wt.% (12064) and 0.3 wt.% (10044). The Na₂O contents of the 263 mesostasis regions display little variation, ranging from 0.1 wt.% to 0.7 wt.%. The K₂O contents, 264 265 however, show relatively large variation between 0.1 wt.% (70035) and 0.4 wt.% (12064), and up to 266 0.9 wt.% for sample 15058 and 0.5 wt.% for 10044.

267

268 5. Discussion

269 5.1. Relationships between bulk-rock chemistry and mesostasis composition

270 Analysis of mesostasis regions in samples from this study and those studied by Pernet-Fisher et al. 271 (2014) show that these regions have distinct mineralogy and textures, which vary greatly across a single 272 sample, and display both mature (Figs. 3A & Fig. 4A) and immature (Figs. 4B, 4C, & 4E) SLI-related 'sieve' textures. Mesostasis compositions appear to be more dependent on the composition of 273 274 surrounding mineral phases than on bulk-rock composition, which is consistent with the observations 275 of Pernet-Fisher et al. (2014). The similarity of Al₂O₃, CaO, and K₂O concentrations of the mesostasis 276 regions of 10044 and 15058 on one hand, and of 12064 and 70035 on the other hand, is a reflection of surrounding pre-existing grains. The majority of mesostasis regions within 10044 and 15058 are mostly 277 surrounded by pre-existing pyroxenes and smaller plagioclase grains, while mesostasis regions in 12064 278 are found between pre-existing plagioclase and pyroxene grains, and those in 70035 are commonly 279 observed between pre-existing plagioclase grains. Phase boundaries in contact with mesostasis melt are 280 thought to re-equilibrate with this melt. However, precisely quantifying the extent of diffusion into pre-281 282 existing phenocrysts is not possible from our dataset. It is worth noting, however, that mesostasis areas

surrounded by pre-existing plagioclase that was not included in melt reconstructions may lead to modeled melts having lowerAl₂O₃, contents for example, assuming some of the melt Al₂O₃ is lost as diffusion into pre-existing grains.

Mineral compositional variations between different mesostasis regions within a sample also support fractional crystallization as an important process in controlling mesostasis bulk compositions and explaining differences for different mesostasis regions. For example, Figure 11 highlights how feldspar and pyroxene compositions within different mesostasis regions in sample 10044 vary. It is important to note that in addition, sampling bias, notably due to the 2D nature of mesostasis regions observed in thin sections while they are in fact 3D objects, can also be responsible for part of the variations observed between different mesostasis regions in individual samples.

293 The bulk-rock samples from this study are all relatively enriched in Fe (Mg# = 37 for 12064, 38 for 294 10044 and 44 for 15058), the less differentiated being sample 70035 (Mg# = 48). Sample 12064 has the 295 lowest Mg# and the highest mesostasis FeO content. Sample 10044 has a similar Mg# but a lower bulk 296 mesostasis FeO content compared to 12064. Pernet-Fisher et al. (2014) have suggested that lunar basalts 297 with low Mg# (<50) are more likely to undergo SLI than those with higher Mg# (>50). Even though all 298 the studied samples have low-Mg#, the two samples that do not display evidence of SLI, 15058 and 299 70035, have the highest Mg#, which supports the observation of Pernet-Fisher et al. (2014). Also, 300 Pernet-Fisher et al. (2014) found that their low Mg# basalts displayed the largest range in pyroxene 301 compositions. In the studied samples, pyroxene have more restricted compositions. Pyroxene in samples 302 70035 and 15058 displays the largest range in compositions (Fig. 9b), which is opposite to the 303 observation of Pernet-Fisher et al. (2014) since these two samples have the highest Mg#.

The SiO₂ contents of K-rich glass for all samples cluster together at around 74-78 wt.%, while the Na₂O+K₂O contents of glass in 70035 (\sim 7.0-7.5 wt.%) are slightly lower than in basalts 10044, 12064 and 15058 (> 8 wt.%; Fig. 10). This may suggest that mesostasis regions in 70035 generally crystallized earlier than in other mare basalts, having slightly less evolved chemical compositions (Table 6), which is also reflected in the more Mg-rich nature of pyroxene in 70035 mesostasis regions. In general there is no relationship between SiO₂ in the bulk-rock, bulk-mesostasis, and mesostasis

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glasses, supporting the hypothesis that fractional crystallization of major silicate minerals may not bethe main process controlling the compositional characteristics of mesostasis regions.

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5.2. Fractional crystallization modeling and the influence of bulk-rock composition on late-stage
melts.

To provide further understanding on the evolution of these lunar basalts, crystallization modeling 315 316 was undertaken using whole-rock major element compositions for each of the four samples. Liquid lines of descent were calculated using the 'Simulating Planetary Igneous Crystallization Environments' 317 package (SPICEs; Davenport et al. 2014), which incorporates the FXMOTR, MAGFOX, and 318 MAGPOX programs (Longhi 1991). The use of MELTS for modeling basaltic systems is well 319 320 demonstrated in the literature (Ghiorso and Sack 1995; Ghiorso et al. 2002; Gualda et al. 2012) and has 321 been used for extraterrestrial applications (e.g. Anand et al. 2003; Day et al. 2006; Liu et al. 2009; Fagan 322 et al. 2014). However, the SPICEs program has the unique advantage of being calibrated for extra-323 terrestrial systems. Here, the MAGFOX program was used to model fractional crystallization of the 324 four Apollo samples under lunar relevant conditions. The MAGFOX program has been widely utilized 325 for modeling Apollo basalt evolution (Neal et al. 1994a, 1994b) and more recently to investigate SLI in 326 late-stage melts (Pernet-Fisher et al. 2014). Results from the MAGFOX algorithm are plotted on the 327 AFM 2D pseudo-ternary ($Na_2O + K_2O - FeO - MgO$; Fig. 12).

328 For each sample, all available bulk compositions were input into the MAGFOX algorithm. The 329 liquid line of descent (LLD) for each sample varies with composition indicating the sensitivity of the 330 model to relatively minor changes in bulk composition (Fig. 12). To investigate the possible effect of pressure, we used the bulk composition from Engel and Engel (1970) for sample 10044 and varied the 331 pressure between 1 kb, 5 kb, and 10 kb, which had very little effect on the until the final stages of 332 crystallization (Fig. 12A). When MgO becomes exhausted in the system, the 1 kb LLD moves towards 333 lower FeO contents while the 10 kb moves in the opposite direction on the AFM (the run at 5 kb did 334 not crystalize to the point of MgO exhaustion (Fig. 12A). Based on modeling for sample 10044, we 335 considered the effect of pressure much less significant than the effect of the bulk rock composition, and 336 337 runs were only performed at a pressure of 1 kb for the remaining samples.

338 MAGFOX was unable to model crystallization up to 100% fractionation for any of the samples. The volume of crystals fractionated for 10044 was 64 % using the bulk composition from Wänke et al. 339 (1970), 98 % using that of Engel and Engel (1970) and Beaty and Albee (1978), and 99 % using those 340 from Agrell et al. (1970), Wakita et al. (1970) and Dymek et al. (1975) (Fig. 12B). For sample 12064, 341 342 the volume of crystals fractionated ranged from 88 % using the composition from LSPET (1970) to 99 % with those from Kushiro and Haramura (1971) and Scoon (1971) (Fig. 12C). Finally, the volume of 343 crystals fractionated for sample 15058 varied from 97 % using the composition of Rhodes and Hubbard 344 (1973) to 99 % with that of Willis et al. (1972) (Fig. 12D) while the crystallization of 70035 reached 345 96 % (Rhodes et al. 1976). 346

347 For all the samples the LLD's plot within the mare basaltic field on the AFM (Fagan et al. 2014). 348 The LLD's for 10044 are in good agreement with some of the compositions of late-stage melts 349 calculated in this study (Fig. 12B). The bulk-rock composition from Beaty and Albee (1978) has a low initial bulk alkali content (Na₂O + $K_2O = 0.02$ wt.%), resulting in a LLD which is too depleted in Na₂O 350 351 and K₂O to correspond with any of the late-stage melt calculated compositions. The LLD calculated 352 using the bulk-rock composition of Wänke et al. (1970), which has an initial alkali content much higher 353 than the other compositions, also has alkali contents which are higher than the majority of the mesostasis 354 region calculated compositions. There is good agreement for the majority of calculated mesostasis regions and the compositions of Dymek et al (1975), Engel and Engel (1970), and Wakita et al. (1970). 355 356 The LLD's for 12064 plot closely together and show fairly good agreement with the calculated late-357 stage melt compositions (Fig. 12C). For samples 15058 and 70035, the LLD's do not correspond with 358 calculated mesostasis region compositions (Fig. 12D

The overall broad agreement between petrological and modeling estimates of mesostasis melt compositions in sample 12064, and to a lesser extent 10044, suggests that SPICEs may be an efficient tool for calculating the composition of late-stage melts (Fig. 12). However, SPICE modeling was unable to reconcile compositions in samples 15058 and 70035. Additionally, AFM diagrams only considers Na₂O + K₂O, FeO, and MgO contents modeled using SPICEs, and other chemical parameters may differ in their reconciliation. Petrological estimates of mesostasis regions compositions for 15058 appear to slightly overestimate MgO and Na₂O + K₂O contents (Fig. 12D). This is similar for 70035, which shows 366 a large discrepancy between the reconstructed bulk mesostasis compositions and those modeled by SPICEs, as the reconstructed bulk mesostasis compositions are Mg-rich and would plot near the 367 beginning of the LLD, when SPICEs modeling suggests that mesostasis compositions should be similar 368 to those in sample 10044. The high MgO content calculated for the bulk mesostasis regions in 70035 369 370 (Table 6) presumably results from the high proportion of pyroxene relative to plagioclase surrounding mesostasis regions in this sample. The pyroxene grains in 70035 have also higher Mg contents than 371 those of the other samples (Fig. 9B). As highlighted by the other samples, however, the bulk-rock 372 composition inputted in SPICE can have a significant influence on the calculated LLDs and with only 373 one available composition for 70035 it is difficult to make strong conclusions. Overall the outcome of 374 MAGFOX modeling indicates that fractional crystallization can produce the compositions seen for 375 376 some late-stage melts but not all. This suggests, therefore, that other processes such as equilibrium crystallization and/or alteration of early-crystallized pyroxene and/or plagioclase phenocrysts are also 377 378 affecting late-stage melt compositions. The variability of late-stage melts also highlights that apatite 379 within lunar mare basalts does not crystalize from a fixed melt composition.

380

381 *5.3. Silicate liquid immiscibility*

382 We have argued that the composition of phases between which mesostasis regions become trapped 383 and crystallize have a large influence on these late-stage melt compositions. After these late-stage melts 384 become trapped, SLI is thought to exert additional control on the compositions of these melts (Pernet-385 Fisher et al., 2014). It is not known, however, if SLI commonly operates in all late-stage melts. There 386 are many regions, in the samples studied here, where SLI has obviously occurred. Where present, 387 however, the extent of SLI appears limited and has no influence on the bulk composition of these regions as both fractions remain. It should be noted, however, that the absence of SLI-related textures, 388 such as sieve textures, does not indicate that SLI did not occur, only that it was not followed by 389 immediate quenching if it occurs. In the sections studied here, apatite was not found in direct contact 390 with textures directly related to SLI within mesostasis regions, preventing a thorough investigation of 391 whether apatite crystallized before, during or after SLI. Volatiles, such as F, Cl and OH, can be 392

fractionated during SLI (Lester et al. 2013), which, therefore, could add further uncertainty into reconstructing volatile abundances in silicate melts based on analysis of volatiles in apatite.

395

396 6. Summary

397 The lack of relationship between bulk-rock chemistry and reconstructed mesostasis melt composition argues for distinct chemical evolution of the different mesostasis regions within a single 398 crystallizing basalt. Mesostasis regions within a single sample can vary in composition and textural 399 maturity, which suggests that volatile contents could also vary within these regions. Our study also 400 shows that mesostasis regions have bulk compositions that differ significantly from those of the initial 401 402 basaltic melts that formed the bulk sample. As such, partition coefficients derived from experimental 403 studies of apatite-melt partitioning designed for basaltic melts may not be fully applicable. Future 404 experiments on apatite-melt volatile partitioning should, therefore, be conducted with more silicic 405 starting compositions given the strong dependence that melt composition has on partitioning in this 406 system (Mathez and Webster 2005).

407 The mineralogy and petrology of mare basalts are well characterized in the literature, yet detailed 408 analyses of mesostasis regions in Apollo samples have been largely overlooked. This study has 409 highlighted the variability of mesostasis regions in lunar basalts. Once a mesostasis melt is trapped 410 between pre-existing grains, its evolution becomes distinct and separated from the evolution of melts trapped in other mesostasis regions. It is expected that volatile evolution in these mesostasis regions is 411 equally distinct, and as such, detailed petrographic information of apatite surroundings should be 412 413 collected when conducting apatite analysis. In general, the mesostasis melts are elevated in SiO₂, slightly enriched in alkali elements, and contain varying amounts of TiO₂ and FeO. The bulk-mesostasis 414 compositions calculated here may also be used to guide future experimental work on apatite-melt 415 volatile partitioning under lunar conditions. 416

417

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- 426

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