

1 **Characterization of mesostasis regions in lunar basalts: Understanding late-stage melt evolution**
2 **and its influence on apatite formation**

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13 **ABSTRACT**

14
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**

35 In order to constrain the volatile inventory of the lunar interior many studies have focused on
36 measuring structurally bound OH in apatite [Ca₅(PO₄)₃(F,Cl,OH)] from Apollo and lunar meteorite
37 samples (e.g. Boyce et al. 2010; McCubbin et al. 2010a, 2010b, 2011; Barnes et al. 2013, 2014; Tartèse
38 et al. 2013b, 2014a, 2014b). Yet, translating volatile measurements in apatite to volatile abundances in
39 co-existing melt has proved troublesome, leading many studies to focus on improving understanding
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
42 forms in lunar melts. Apatite begins to crystallize after ~95 % melt solidification in typical mare
43 magmas (McCubbin et al. 2010b), but is likely to start crystallizing earlier (~85 % melt solidification)
44 in KREEP (enriched in incompatible elements such as K, REE and P) basalts (Tartèse et al. 2014a).
45 Apatite, therefore, does not form from bulk basaltic melts but instead forms from differentiated late-
46 stage melts, which are represented by mesostasis regions within lunar samples (Henderson 1970).
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.

55

56

57 **2. Samples**

58 *2.1. 10044*

59 Lunar sample 10044 is a high-Ti ilmenite basalt with a low-K content (Beaty and Albee 1978). It
60 is a subophitic basalt (James and Jackson 1970) that is sometimes described as a microgabbro
61 (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
63 studies carried out on this sample yielded Rb-Sr dates of ~ 3.7 Ga (Papanastassiou et al. 1970), $^{40}\text{Ar}/^{39}\text{Ar}$
64 dates of 3.73 ± 0.04 Ga (Turner 1970; Guggisberg et al. 1979) and a tranquillityite $^{207}\text{Pb}/^{206}\text{Pb}$ date of
65 3.72 ± 0.01 Ga (Tartèse et al. 2013a). Cosmic-ray exposure (CRE) ages of ~ 70 Ma were determined
66 using the abundance of cosmogenic ^{81}Kr (Hohenberg et al. 1970), ^{126}Xe (Srinivasan 1974) and ^{38}Ar
67 (Guggisberg et al. 1979). 10044 contains subhedral to anhedral zoned pyroxene phenocrysts ($\sim 45\%$
68 modal abundance) surrounded by a matrix of subophitic plagioclase ($\sim 35\%$), and ilmenite ($\sim 13\%$)
69 (McGee et al. 1977). Mesostasis areas in this sample are composed of silica ($\sim 7\%$), troilite ($\sim 0.5\%$)
70 associated with metallic iron, K-rich glass, and accessory minerals including apatite ($\sim 0.1\%$) and Zr-
71 rich minerals such as baddeleyite and tranquillityite (McGee et al. 1977). We have examined mesostasis
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 ($\sim 1 \times 2$
74 mm), plagioclase laths ($\sim 1 \times 0.2$ mm) and ilmenite ($\sim 3 \times 0.5$ mm). Multiple mesostasis regions are
75 found throughout the 10044,645 section and are further described in section 4.1.

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 ($\sim 56\%$ modal abundance) intergrown with anhedral plagioclase
80 laths ($\sim 29\%$) (Klein et al. 1971). It is characterized by coarse-grained mesostasis areas ($\sim 9\%$) that
81 contain fayalite, Fe-rich pyroxene, troilite, K-feldspar, K-rich glass, Zr-rich phases, merrillite and
82 apatite (Kushiro and Haramura 1971). Plagioclase compositions range from An_{93} to An_{86} while
83 pyroxene compositions are close to the Fe-rich end-member hedenbergite (Kushiro and Haramura
84 1971). The sample also contains coarse-grained laths of ilmenite (3.5%) associated with ulvöspinel
85 (2%). Small, sporadic grains of pyroxferroite are observed coexisting with clinopyroxene (Klein et al.
86 1971). Rb-Sr (Papanastassiou and Wasserburg 1971) and $^{40}\text{Ar}/^{39}\text{Ar}$ (Horn et al. 1975) dating studies for
87 sample 12064 yielded dates of 3.18 ± 0.09 Ga and 3.18 ± 0.01 Ga, respectively, which have been
88 interpreted as crystallization ages. An exposure age of 255 Ma has been reported by Horn et al. (1975),
89 while exposure ages determined from abundances of cosmogenic ^3He , ^{21}Ne , and ^{38}Ar cluster around

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 ($> 2 \times 2$ mm), intergrown with anhedral
93 plagioclase ($\sim 2 \times 1$ mm) and with minor ilmenite ($\sim 1 \times 0.2$ mm). Multiple mesostasis regions are found
94 across the 12064,130 section and are further described in section 4.1.

95

96 2.3. 15058

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

110 Apollo 17 sample 70035 is a medium-grained, vesicular, high-Ti basalt (Ridley and Brett 1973). It
111 is a large crystalline basalt with large ($> 1\text{mm}$) anhedral clinopyroxene crystals ($\sim 46\%$ modal
112 abundance) enclosing armalcolite, ilmenite, and spinel ($\sim 22\%$). Interstitial plagioclase ($\sim 26\%$) is found
113 together with ilmenite and olivine ($\sim 2.5\%$) (Weigand 1973). The mesostasis regions ($\sim 2\%$) include
114 silica, K-feldspar, tranquillityite, ilmenite, ulvospinel, troilite, K-rich glass and apatite (Papike et al.
115 1974; McGee et al. 1977). Metallic iron is found in the cracks and cleavage of opaque oxides and
116 silicates in this sample (El Goresy and Ramdohr 1975). Plagioclase composition in sample 70035 varies
117 from An_{88} to An_{83} (Weigand 1973). The Rb/Sr dates of 3.82 ± 0.06 Ga and 3.73 ± 0.11 Ga determined

118 for 70035 are consistent with each other (Evensen et al. 1973; Nyquist 1977) and with a $^{40}\text{Ar}/^{39}\text{Ar}$ date
119 of 3.75 ± 0.07 Ga (Stettler et al. 1973). Exposure ages determined from abundance of cosmogenic ^{38}Ar
120 (Stettler et al. 1973) and ^{81}Kr (Drozd et al. 1977) are 95 to 100 Ma and 122 ± 3 Ma, respectively. We
121 have studied mesostasis areas in thin section 70035,195. This section is 14×3 mm in size and has a
122 mineralogy consistent with that of the main rock sample, comprising anhedral plagioclase phenocrysts
123 ($> 2 \times 1$ mm), pyroxene ($\sim 1 \times 1$ mm) and ilmenite ($\sim 0.5 \times 0.2$ mm). Small mesostasis pockets are found
124 throughout the studied section and are further described in section 4.1

125

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
130 element maps of the studied samples, mesostasis regions were identified by the presence of small (< 1
131 mm) anhedral and amorphous phases that crystallized between larger surrounding crystals. The
132 presence of symplectite assemblages and/or silicate liquid immiscibility textures was also used to
133 identify mesostasis regions. These regions are rich in fayalite, silica, and glass and host a wide variety
134 of smaller phases (i.e. sulphides, phosphates) compared to the surrounding major minerals. Pernet-
135 Fisher et al. (2014) suggested that coarse-grained silica in mare basalts ($> 100 \mu\text{m}$) is likely a pseudo-
136 eutectic mineral and, therefore, should not be included as a mesostasis phase. For the majority of
137 mesostasis regions investigated here this is not a concern as silica within the regions is fine-grained
138 ($< 100 \mu\text{m}$), and intergrown with other mesostasis phases. For mesostasis regions containing large (> 100
139 μm), elongate silica grains, intergrown with other mesostasis phases, we have considered this as a
140 product of crystallization of the late-stage melt and, therefore, as part of the mesostasis region. In the
141 basalts studied here silica is only found associated with mesostasis regions.

142 A large variety of textures and mineral phases were observed in the different mesostasis regions.
143 We determined the two-dimensional extents of the different regions in multiple ways. For silica-rich
144 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
146 phases. Re-equilibration of pre-existing plagioclase and pyroxene grains with mesostasis melt modified
147 their chemistry and created chemical zonations. In some mesostasis regions clear modification of pre-
148 existing pyroxene associated with overgrowth of mesostasis phases was observed on back-scattered
149 electron (BSE) and false-color X-ray images, and later confirmed with electron probe microanalysis
150 (EPMA). In regions with no clear modification of pre-existing phases, mesostasis boundaries were
151 drawn around mesostasis phases only. Compositional differences between phases within and outside
152 mesostasis regions are detailed in section 5. For modal abundance estimates, where possible, mesostasis
153 regions were limited to areas that included late-stage phases with limited incorporation of surrounding
154 pre-existing phases. It is worth noting that thin sections only provide 2D sections through 3D mesostasis
155 regions, which likely implies a sampling bias. To try and mitigate the possible effects associated with
156 this sampling bias, we analyzed multiple mesostasis regions $> 50 \times 50 \mu\text{m}$.

157

158 *3.2. Electron Microscopy*

159 The BSE maps were collected using the FEI Quanta 3D dual beam scanning electron microscope
160 (SEM) at The Open University. The instrument is fitted with an Oxford Instruments 80 mm X-MAX
161 energy dispersive X-ray detector, which allowed X-ray maps of each sample to be obtained by using
162 the SEM in Energy Dispersive Spectroscopy (EDS) mode. Elemental mapping was carried out using a
163 20 kV accelerating voltage and a 0.60 nA beam current. X-ray maps were acquired at resolutions of 512
164 \times 448 and 1024 \times 896 pixels with a magnification of 200 to 300.

165 Quantitative mineral compositions were determined using a CAMECA SX-100 Electron Probe
166 Micro Analyzer (EPMA) at The Open University. An accelerating voltage of 20 kV and a beam current
167 of 20 nA were used except for plagioclase and glass analyses where the beam current was reduced to
168 10 nA. The beam diameter varied from 1 μm to 10 μm , depending on the dimensions of the analyzed
169 phase. Standard count rates of 20 to 40 s per element were used, with a background counting time of
170 half the peak counting time before and after peak analysis. A selection of natural standards were used
171 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
174 necessarily in the same mesostasis regions. For 70035, the average apatite composition of those
175 analyzed in the other samples by Tartèse et al. (2013b) was used as these authors did not analyze apatite
176 in this sample. The modal mineralogy and the quench glass abundances, for each mesostasis area, were
177 calculated using pixel histograms from BSE-images in the ImageJ[®] software. For each mesostasis
178 region a bulk composition was calculated using average EPMA compositions for each phase and the
179 modal mineralogy within individual regions.

180

181 **4. Results**

182 *4.1. Mesostasis textural descriptions*

183 The BSE images and false-color compositional maps of representative mesostasis regions for
184 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
187 maps combining Si, Ca and Fe. These regions are $\sim 200 \times 200 \mu\text{m}$ and are located between pre-existing
188 plagioclase laths (Figs. 1B & 1C) and pyroxene phenocrysts (Figs. 1A, 1C, & 1D). All of the regions
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 fayalite grains large enough (i.e. $>10 \mu\text{m}$) for
191 multiple microprobe spot analyses (Figs. 1A & 1D). Symplectite texture (labelled as 'pxf' in
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-
195 Ba-rich glass (hereafter referred to as K-glass) enclosed by fayalite, characteristic of silicate-liquid
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
198 mature (large glass droplets) or immature (small glass droplets). The mesostasis region in Figure 1D
199 displays immature 'sieve' texture, with fine-grained droplets of K-glass trapped within host fayalite.

200 Seven mesostasis regions within basalt 12064,130 were analyzed. These regions are $\sim 100 \times$
201 $100 \mu\text{m}$ and found mostly between pyroxene phenocrysts (Figs. 3A, 3C, & 3D) and plagioclase laths
202 (Fig. 3B). Mesostasis regions contain varying amounts of apatite, silica, pyroxene, K-feldspar, fayalite,
203 and ilmenite (Figs. 3 & 4). Pyroxferroite breakdown products are present in some regions (Figs. 3B,
204 4B, 4C, 4D & 4F). Both mature (Fig. 3A) and immature (Figs. 3B & 3D) ‘sieve’ textures, indicative of
205 SLI, were observed in all but two mesostasis region of 12064 (Figs. 3C & 4D). One region within 12064
206 contained both mature and immature SLI textures (Fig. 3A & 4A).

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

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

219

220 *4.2. Composition of mesostasis phases*

221 For all four samples plagioclase compositions vary from An_{93} to An_{68} (Fig. 9A). The K-rich
222 plagioclase feldspars are those firmly within mesostasis regions (Fig. 9A). Most of the pyroxene
223 analyses in samples 10044 and 12064 plot close to the Hd – Fs binary (Fig. 9B). In contrast, all pyroxene
224 analyses for 70035 are more Mg-rich, plotting towards the En and Di regions (Fig. 9B). The few
225 analyses of pyroxene compositions in 15058 range between those in 70035 and in samples
226 10044/12064(Fig. 9B).

227 Glass compositions from mesostasis regions are displayed in a Total-Alkali-Silica (TAS) diagram
228 in Figure 10, together with compositions of the bulk-rock mare basalts studied and of lunar granites
229 (Kushiro and Haramura 1971; Rhodes and Hubbard 1973; Rhodes et al. 1976; Beatty and Albee 1978;
230 Seddio et al. 2013). In general, the glass compositions plot in a similar region of the diagram to the
231 lunar granites (~4 - 10 wt.% alkali and ~68 wt.% to 78 wt.% SiO₂).

232

233 *4.3 Reconstructed bulk mesostasis compositions*

234 Average modal abundances for mesostasis regions in the four studied samples are given in Table 1,
235 while average compositions of mesostasis mineral and glass phases are given in Tables 2 - 5. These
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
239 within 10044 mesostasis regions, for example, varied from 57 wt.% to 83 wt.%, while SiO₂ contents
240 varied by about 12 wt.% across the different mesostasis regions in 70035. Large variations between
241 bulk compositions of mesostasis regions were also observed for TiO₂ in sample 70035 for example,
242 which varied by 14 wt.%. Significant variations in CaO (6.2 wt.% difference) contents in sample 12064
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
245 oxides in the calculated mesostasis compositions.

246 The reconstructed bulk mesostasis compositions display large variations between each sample. The
247 largest variation is observed for SiO₂, which varies from 70 wt.% (10044) to 56.2 wt.% (70035). There
248 is no correlation between the reconstructed SiO₂ content of the mesostasis regions and the SiO₂ content
249 of the bulk sample. Sample 70035 does have the lowest bulk-rock SiO₂ content (37.8 wt.%; Rhodes et
250 al., 1976) out of the four samples studied here, but 10044 has a lower bulk-rock SiO₂ content (42.2
251 wt.%; Beatty and Albee 1978) than both 12064 (46.0 wt.%; Scoon, 1971) and 15058 (47.8 wt.%; Rhodes
252 and Hubbard, 1973) and, yet, the highest bulk-mesostasis SiO₂ content. The reconstructed mesostasis
253 compositions for the four samples also display large variations in TiO₂ content. Sample 70035, a high-
254 Ti basalt, has the highest mesostasis TiO₂ content (7.0 wt.%), however, high-Ti basalt 10044 has a

255 relatively low mesostasis TiO_2 content (1.5 wt.%). TiO_2 contents of mesostasis regions in low-Ti basalts
256 12064 and 15058 are 0.9 wt.% and 3.6 wt.%, respectively. FeO contents of mesostasis regions in
257 samples 10044 (10.8 wt.%), 15058 (11.1 wt.%) and 70035 (11.2 wt.%) are fairly similar, while it is
258 much higher in 12064 (23.0 wt.%). The CaO contents of bulk mesostasis are similar in samples 10044
259 and 15058 (6.4 wt.% CaO), while bulk mesostasis in 12064 and 70035 have varying CaO contents (5.7
260 – 8.8 wt.% CaO). The Al_2O_3 contents of mesostasis regions in 10044, 15058, and 70035 are similar
261 (9.3, 9.2, and 10.1 wt.%), while 12064 has a lower Al_2O_3 content of 4.1 wt.%. The MgO contents of the
262 mesostasis regions are highly variable between the four samples, ranging between 4.9 wt.% (70035)
263 and 1.7 wt.% (15058), down to 0.8 wt.% (12064) and 0.3 wt.% (10044). The Na_2O contents of the
264 mesostasis regions display little variation, ranging from 0.1 wt.% to 0.7 wt.%. The K_2O contents,
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
273 ‘sieve’ textures. Mesostasis compositions appear to be more dependent on the composition of
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 Al_2O_3 , CaO, and K_2O 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
277 surrounding pre-existing grains. The majority of mesostasis regions within 10044 and 15058 are mostly
278 surrounded by pre-existing pyroxenes and smaller plagioclase grains, while mesostasis regions in 12064
279 are found between pre-existing plagioclase and pyroxene grains, and those in 70035 are commonly
280 observed between pre-existing plagioclase grains. Phase boundaries in contact with mesostasis melt are
281 thought to re-equilibrate with this melt. However, precisely quantifying the extent of diffusion into pre-
282 existing phenocrysts is not possible from our dataset. It is worth noting, however, that mesostasis areas

283 surrounded by pre-existing plagioclase that was not included in melt reconstructions may lead to
284 modeled melts having lower Al_2O_3 contents for example, assuming some of the melt Al_2O_3 is lost as
285 diffusion into pre-existing grains.

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

293 The bulk-rock samples from this study are all relatively enriched in Fe ($\text{Mg}\# = 37$ for 12064, 38 for
294 10044 and 44 for 15058), the less differentiated being sample 70035 ($\text{Mg}\# = 48$). Sample 12064 has the
295 lowest $\text{Mg}\#$ and the highest mesostasis FeO content. Sample 10044 has a similar $\text{Mg}\#$ but a lower bulk
296 mesostasis FeO content compared to 12064. Pernet-Fisher et al. (2014) have suggested that lunar basalts
297 with low $\text{Mg}\#$ (<50) are more likely to undergo SLI than those with higher $\text{Mg}\#$ (>50). Even though all
298 the studied samples have low- $\text{Mg}\#$, the two samples that do not display evidence of SLI, 15058 and
299 70035, have the highest $\text{Mg}\#$, which supports the observation of Pernet-Fisher et al. (2014). Also,
300 Pernet-Fisher et al. (2014) found that their low $\text{Mg}\#$ 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 $\text{Mg}\#$.

304 The SiO_2 contents of K-rich glass for all samples cluster together at around 74-78 wt.%, while
305 the $\text{Na}_2\text{O}+\text{K}_2\text{O}$ contents of glass in 70035 (~ 7.0 -7.5 wt.%) are slightly lower than in basalts 10044,
306 12064 and 15058 (> 8 wt.%; Fig. 10). This may suggest that mesostasis regions in 70035 generally
307 crystallized earlier than in other mare basalts, having slightly less evolved chemical compositions
308 (Table 6), which is also reflected in the more Mg-rich nature of pyroxene in 70035 mesostasis regions.
309 In general there is no relationship between SiO_2 in the bulk-rock, bulk-mesostasis, and mesostasis

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

312

313 *5.2. Fractional crystallization modeling and the influence of bulk-rock composition on late-stage*
314 *melts.*

315 To provide further understanding on the evolution of these lunar basalts, crystallization modeling
316 was undertaken using whole-rock major element compositions for each of the four samples. Liquid
317 lines of descent were calculated using the ‘Simulating Planetary Igneous Crystallization Environments’
318 package (SPICES; Davenport et al. 2014), which incorporates the FXMOTR, MAGFOX, and
319 MAGPOX programs (Longhi 1991). The use of MELTS for modeling basaltic systems is well
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 ($\text{Na}_2\text{O} + \text{K}_2\text{O} - \text{FeO} - \text{MgO}$; 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
331 pressure, we used the bulk composition from Engel and Engel (1970) for sample 10044 and varied the
332 pressure between 1 kb, 5 kb, and 10 kb, which had very little effect on the until the final stages of
333 crystallization (Fig. 12A). When MgO becomes exhausted in the system, the 1 kb LLD moves towards
334 lower FeO contents while the 10 kb moves in the opposite direction on the AFM (the run at 5 kb did
335 not crystallize to the point of MgO exhaustion (Fig. 12A). Based on modeling for sample 10044, we
336 considered the effect of pressure much less significant than the effect of the bulk rock composition, and
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.
339 The volume of crystals fractionated for 10044 was 64 % using the bulk composition from Wänke et al.
340 (1970), 98 % using that of Engel and Engel (1970) and Beatty and Albee (1978), and 99 % using those
341 from Agrell et al. (1970), Wakita et al. (1970) and Dymek et al. (1975) (Fig. 12B). For sample 12064,
342 the volume of crystals fractionated ranged from 88 % using the composition from LSPET (1970) to 99
343 % with those from Kushiro and Haramura (1971) and Scoon (1971) (Fig. 12C). Finally, the volume of
344 crystals fractionated for sample 15058 varied from 97 % using the composition of Rhodes and Hubbard
345 (1973) to 99 % with that of Willis et al. (1972) (Fig. 12D) while the crystallization of 70035 reached
346 96 % (Rhodes et al. 1976).

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 Beatty and Albee (1978) has a low
350 initial bulk alkali content ($\text{Na}_2\text{O} + \text{K}_2\text{O} = 0.02 \text{ wt.}\%$), resulting in a LLD which is too depleted in Na_2O
351 and K_2O 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
355 regions and the compositions of Dymek et al (1975), Engel and Engel (1970), and Wakita et al. (1970).
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)

359 The overall broad agreement between petrological and modeling estimates of mesostasis melt
360 compositions in sample 12064, and to a lesser extent 10044, suggests that SPICEs may be an efficient
361 tool for calculating the composition of late-stage melts (Fig. 12). However, SPICE modeling was unable
362 to reconcile compositions in samples 15058 and 70035. Additionally, AFM diagrams only considers
363 $\text{Na}_2\text{O} + \text{K}_2\text{O}$, FeO, and MgO contents modeled using SPICEs, and other chemical parameters may differ
364 in their reconciliation. Petrological estimates of mesostasis regions compositions for 15058 appear to
365 slightly overestimate MgO and $\text{Na}_2\text{O} + \text{K}_2\text{O}$ contents (Fig. 12D). This is similar for 70035, which shows

366 a large discrepancy between the reconstructed bulk mesostasis compositions and those modeled by
367 SPICEs, as the reconstructed bulk mesostasis compositions are Mg-rich and would plot near the
368 beginning of the LLD, when SPICEs modeling suggests that mesostasis compositions should be similar
369 to those in sample 10044. The high MgO content calculated for the bulk mesostasis regions in 70035
370 (Table 6) presumably results from the high proportion of pyroxene relative to plagioclase surrounding
371 mesostasis regions in this sample. The pyroxene grains in 70035 have also higher Mg contents than
372 those of the other samples (Fig. 9B). As highlighted by the other samples, however, the bulk-rock
373 composition inputted in SPICE can have a significant influence on the calculated LLDs and with only
374 one available composition for 70035 it is difficult to make strong conclusions. Overall the outcome of
375 MAGFOX modeling indicates that fractional crystallization can produce the compositions seen for
376 some late-stage melts but not all. This suggests, therefore, that other processes such as equilibrium
377 crystallization and/or alteration of early-crystallized pyroxene and/or plagioclase phenocrysts are also
378 affecting late-stage melt compositions. The variability of late-stage melts also highlights that apatite
379 within lunar mare basalts does not crystallize 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
388 regions as both fractions remain. It should be noted, however, that the absence of SLI-related textures,
389 such as sieve textures, does not indicate that SLI did not occur, only that it was not followed by
390 immediate quenching if it occurs. In the sections studied here, apatite was not found in direct contact
391 with textures directly related to SLI within mesostasis regions, preventing a thorough investigation of
392 whether apatite crystallized before, during or after SLI. Volatiles, such as F, Cl and OH, can be

393 fractionated during SLI (Lester et al. 2013), which, therefore, could add further uncertainty into
394 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
398 composition argues for distinct chemical evolution of the different mesostasis regions within a single
399 crystallizing basalt. Mesostasis regions within a single sample can vary in composition and textural
400 maturity, which suggests that volatile contents could also vary within these regions. Our study also
401 shows that mesostasis regions have bulk compositions that differ significantly from those of the initial
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
411 trapped in other mesostasis regions. It is expected that volatile evolution in these mesostasis regions is
412 equally distinct, and as such, detailed petrographic information of apatite surroundings should be
413 collected when conducting apatite analysis. In general, the mesostasis melts are elevated in SiO₂,
414 slightly enriched in alkali elements, and contain varying amounts of TiO₂ and FeO. The bulk-mesostasis
415 compositions calculated here may also be used to guide future experimental work on apatite-melt
416 volatile partitioning under lunar conditions.

417

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426

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