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Constraining the timing and sources of volcanism at the Apollo 12 landing site using new Pb isotopic compositions and crystallisation ages

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ABSTRACT

The basaltic suites collected at the Apollo 12 landing site have been interpreted as representing a stratigraphic sequence of volcanic flows emplaced in the Oceanus Procellarum region between approximately 3100-3300 Ma. This study presents Secondary Ion Mass Spectrometry (SIMS) Pb isotopic analyses of samples from each of the basaltic suites, which have been used to constrain precise crystallisation ages and initial Pb isotopic compositions. The new crystallisation ages are consistent with the three main basaltic suites (olivine, pigeonite and ilmenite) being emplaced over a period of approximately 60 million years, and the improved precision of these ages has made it possible to reinterpret the stratigraphic sequence of basalt flows underlying the Apollo 12 landing site. Contrary to previous studies, the three ilmenite basalts are determined as having the oldest ages (with a weighted average of 3187 \pm 6 Ma; 2 σ) and are, therefore, interpreted as representing the lowest unit in the sequence, underlying the olivine and pigeonite basalts (with an age range constrained by the oldest and youngest pigeonite basalts; 3176 \pm 6 Ma and 3129 \pm 10 Ma; 2 σ). The initial Pb isotopic compositions have been compared with recalculated initial Sr and Nd isotopic compositions, and are consistent with the three main basaltic suites originating from magmatic sources that incorporated different proportions of a common primitive mafic cumulate and the residual trapped liquid fraction remaining after a majority of the lunar magma ocean had crystallised. Our data also demonstrate that the feldspathic basalt (12038) is unique, both in terms of its crystallisation age $(3242 \pm 13 \text{ Ma})$ and its derivation from a distinct mantle reservoir.

1. Introduction

The Apollo 12 mission provided 34.3 kg of mainly basaltic samples (43 of the 47 rocks collected) documenting volcanic activity in Oceanus Procellarum between approximately 3100–3200 Ma. All of the basalts returned by the mission are low-Ti mare basalts ($TiO_2 = 1-6$ wt%; following the scheme of Neal and Taylor, 1992). These have been subdivided into three main basaltic suites: olivine, pigeonite and ilmenite basalts (e.g. Neal et al., 1994a). A fourth, feldspathic, suite has been proposed, which is represented by only one rock sample (12038), although several grains from Apollo 12 soil samples have been suggested as additional members (Korotev et al., 2011; Alexander et al., 2016). Crystallisation ages previously determined for the Apollo 12 basalts have not been sufficiently precise to constrain the ages of the basaltic suites relative to each other, given the narrow time interval (20–50 million years) over which the three main suites appear to have been emplaced. Based on the data available at that time, Snyder et al.

(1997) calculated a series of weighted average ages demonstrating the similarity for all but the single anomalous feldspathic basalt (pigeonite = 3180 Ma; olivine = 3190 Ma; ilmenite = 3200 Ma; feldspathic = 3280 Ma).

Modelling of the Apollo 12 basalt sources initially relied on the sample mineralogies and major- and trace-element characteristics (Warner, 1971; Rhodes et al., 1977). This work led to the hypothesis that the olivine and pigeonite basalts were co-magmatic, with the olivine basalts representing more primitive cumulates, while the pigeonite basalts were considered to be formed from more evolved parental magma compositions (Rhodes et al., 1977). Subsequent studies have also attempted to model the sources of the basalts using Sr and Nd isotopic data in addition to incompatible trace-element (ITE) chemistry (Nyquist et al., 1977, 1979, 1981; Neal et al., 1994a, 1994b; Snyder et al., 1997). Neal et al. (1994a) examined the compositions for fine-grained and vitrophyric Apollo 12 basalts thought to be most representative of the parental magmas, focusing on particular ITE ratios

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(e.g. Co/Hf, Sc/Sm, Rb/Sr and Sm/Eu) that would be indicative of fractionation or accumulation, as well as the presence of pyroxene and plagioclase in the mantle sources. The results of this work indicate that the olivine and pigeonite basalt suites are unrelated and were derived from distinct mantle sources.

Based on the Mg# (atomic Mg/[Fe + Mg]) and the systematics of Li vs. Rare Earth Elements (REE) in similarly fine-grained and vitrophyric Apollo 12 basalts, Snyder et al. (1997) concluded that the three main basaltic suites were derived from partial melts of cumulates formed after 82-94% crystallisation of the Lunar Magma Ocean (LMO). Furthermore, the significantly higher initial ¹⁴³Nd/¹⁴⁴Nd ratios determined for the ilmenite basalts, when compared with those of the olivine and pigeonite basalts, were interpreted as definitive evidence of a distinct source for at least the ilmenite basalts. Combining the Sr and Nd isotopic data for the basalts, Snyder et al. (1997) presented a model in which the parent melts of the three main basaltic suites were formed by varying degrees of partial melting (5-6% for the ilmenite basalts and 7-9% for the olivine and pigeonite basalts) of Lunar Magma Ocean (LMO) cumulates that had incorporated different amounts of ITE-rich LMO residual liquid. In the case of the ilmenite basalts, the relatively high initial ¹⁴³Nd/¹⁴⁴Nd ratios (initial ε^{143} Nd = 10.5–11.3) and low initial ⁸⁷Sr/⁸⁶Sr ratios (0.69920-0.69932) indicate that the ilmenite source was nearly devoid of this residual liquid (< 0.15%), while the olivine and pigeonite sources (initial ϵ^{143} Nd = 4.3–4.7; initial ${}^{87}\text{Sr}/{}^{86}\text{Sr} = 0.69954-0.69960)$ contained more residual liauid (0.3-0.5%). Subsequent work by Hallis et al. (2014) recreated REE abundances measured in olivine, pigeonite and ilmenite basalts starting from a model composition of a low-Ti source with 1% LMO residual liquid and smaller amounts of partial melting (1-2% for the ilmenite basalts and 1-3% for the olivine and pigeonite basalts). The differing conclusions of Snyder et al. (1997) and Hallis et al. (2014) are a reflection of the different modelling approaches and assumptions, but both studies demonstrate the strong effect of very small amounts of LMO residual liquid on the ITE compositions of the basalt sources.

Recent work by Snape et al. (2016) has demonstrated the potential of Secondary Ion Mass Spectrometry (SIMS) Pb isotopic measurements to provide high precision dates for lunar basalts, interpreted to represent igneous crystallisation. The strength of this approach lies in its ability to analyse rapidly a range of phases, providing measurements of both radiogenic Pb isotopic compositions formed through the radioactive decay of ²³⁵U and ²³⁸U in U-rich minerals since crystallisation of a basalt sample and hence indicating the crystallisation age of the sample, as well as compositions closer to the initial Pb component inherited by the basalt from its source. The aim of this study is to use this approach in an attempt to resolve the remaining ambiguity regarding the chronology of volcanic activity at the Apollo 12 landing site and provide new insights to the evolution of the mantle sources from which the Apollo 12 basalts originated.

2. Methods

2.1. Sample descriptions

In addition to the three Apollo 12 basalts investigated previously (feldspathic basalt 12038, pigeonite basalt 12039 and ilmenite basalt 12063; Snape et al., 2016), a further five samples have been analysed in the present study. These samples include two ilmenite basalts (12005 and 12016), two olivine basalts (12002 and 12012) and a pigeonite basalt (12021).

The two olivine basalts analysed in this study (12002,600 and 12012,34; Figs. 1a-b and A.1-A.2) have similar porphyritic-subophitic textures, characterised by olivine (13–23% by modal abundance) and pyroxene (50–56%) phenocrysts (typically 200–600 μ m in size) surrounded by subophitic groundmasses. Both mafic phases are prominently zoned from more Mg-rich cores to more Fe-rich rims, with the cores of the pyroxene crystals also exhibiting more complex zoning

between pigeonite and augite compositions. The subophitic groundmass surrounding the mafic phenocrysts comprises pyroxene intergrown with plagioclase crystals (typically 50-400 µm; 23-27% by modal abundance; Table B.1), which are generally elongated, occasionally with hollow intrafasciculate morphologies, and in the case of 12002 are sometimes clustered to form acicular bundles. Ilmenite laths (up to approximately 150 µm long; 2% by modal abundance) and spinel crystals (50-100 µm; 1%) are common throughout the groundmass of the two samples, with the spinels commonly exhibiting well defined chromite cores and ulvöspinel rims. Also present are grains of a silica polymorph (most likely cristobalite; 10-50 µm; 1%; Table B.1), which tend to occur within or adjacent to the plagioclase crystals. Mesostasis areas, approximately 50 um in size, are commonly found at the boundaries between plagioclase and the most Fe-rich pyroxene. These typically contain K-rich glass, sulfide grains and Ca-phosphate, often as fine-grained (1-5 µm) intergrowths.

The pigeonite basalt (12021,619; Figs. 1c and A.3) contains several very large (1–4 mm) pyroxene phenocrysts in a variolitic groundmass primarily composed of pyroxene (total pyroxene modal abundance of 64%), plagioclase (28%), ilmenite (4%) and grains of silica (4%; Table B.1). The silica grains form either fine laths or less elongated subhedral-anhedral crystals (previous studies indicate that the former is likely tridymite while the latter is cristobalite; Weill et al., 1971). Less common phases in the groundmass include occasional grains of both chromite and ulvöspinel (< 1%; Table B.1). Relatively large (up to approximately 200 μ m) symplectite assemblages and smaller (50–100 μ m) mesostasis areas typically occur at the boundaries between the Fe-rich pyroxene grains and the other groundmass phases. The mesostasis includes K-rich glass, K-feldspar and Ca-phosphate crystals.

The ilmenite basalt 12005 has been previously identified as being a unique lunar basalt, due to its high Mg# (62) and olivine content (30% by modal abundance), as well as its "cumulate texture" (Rhodes et al., 1977). As is typical of the sample, the section analysed in this study (12005,78; Figs. 1d and A.4.) contains rounded olivine grains (typically 50-300 µm in size; 37% by modal abundance; Table B.1) poikilitically enclosed by pyroxene (typically 100-500 µm; 40%), plagioclase (typically 50-200 µm; 19%) and (less commonly) ilmenite (typically 100-200 µm; 3%), while euhedral to subhedral grains of ulvöspinel and chromite (typically 10-50 µm) are common throughout the sample (1%), occasionally forming glomerophyric clusters, with the ulvöspinel often exhibiting ilmenite exsolution. The EDS element map of the sample (Fig. A.4.) indicates that the major phases in the section are generally equilibrated and homogenous in major element composition. Grains of FeNi-metal and troilite (between 1 and $20 \,\mu m$; < 1% by modal abundance; Table B.1) are also present within the pyroxene and plagioclase grains, while several mesostasis areas (approximately 50 µm) of additional accessory phases are present interstitially between the major phases. The accessory phases in the mesostasis (typically between 1 and 10 µm) include Ca-phosphates, K-rich glass, K-feldspar, sulfides and ilmenite grains.

Ilmenite basalt 12016 was described by Dungan and Brown (1977) as an equigranular, medium-grained microgabbro. The section analysed in this study (12016,39; Figs. 1e and A.5.) contains pyroxene (54% by modal abundance; Table B.1), plagioclase (25%), olivine (15%) and ilmenite (4%). As has been previously reported (Dungan and Brown, 1977), the pyroxene grains (up to approximately 600 μ m in size) are characterised by complex intergrowth of augite and pigeonite. The olivine grains (typically 50–500 μ m) exhibit some zonation from more Mg-rich cores to Fe-rich rims, and typically have embayed grain boundaries with the pyroxene. Anhedral plagioclase grains (typically 50–400 μ m) occur interstitially to the olivine and pyroxene and occasionally subpoikilitically enclosing the mafic phases. After ilmenite, spinel (both chromite and ulvöspinel) is the next most abundant phase (~1%; Table B.1). Mesostasis areas (between approximately 20–100 μ m in size) occasionally occur in interstitial sites between the Fe-rich rims

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