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Identifying impact events within the lunar cataclysm from ⁴⁰Ar–³⁹Ar ages and compositions of Apollo 16 impact melt rocks

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Abstract

Lunar impact melt breccias provide a unique record of the timing and frequency of collisional events during the early history of the inner Solar System prior to the development of a significant rock record on Earth. The predominance of ages clustering between 3.8 and 4.0 Ga was a major, unexpected discovery obtained from geochronological studies of lunar impact melts, and is the basis of the concept that a cataclysmic bombardment of large planetesimals struck the Earth and Moon, and possibly the entire inner Solar System, about 3.85 ± 0.10 billion years ago. As a test of the cataclysm hypothesis, we measured high-resolution (20–50 steps) 40 Ar– 39 Ar age spectra on 25 samples of Apollo 16 impact melt breccias using a continuous laser heating system on sub-milligram fragments. Twenty-one of these 25 breccias produced multi-step plateaus that we interpret as crystallization ages, with 20 of these ages falling in the range 3.75-3.96 Ga. We propose that at least four different melt-producing impact events can be distinguished based on the ages, bulk compositions, and petrographic characteristics of Apollo 16 melt breccias. The recognition of multiple impact events within the Apollo 16 melt breccia suite shows that numerous impact events occurred on the lunar surface within a relatively narrow time interval, providing additional evidence of a heavy bombardment of the Moon during the early Archean.

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1. Introduction

The upper crust of the Moon records an integrated history of impact events extending from soon after the main accretionary phase of planet formation to the present day. Lunar impact melt breccias provide a record of the timing and frequency of collisional events that occurred prior to the development of a significant rock record on Earth. Arguably one of the most important and unexpected discoveries obtained from the Apollo expeditions to the Moon is the predominance of impact melt breccia ages between 3.8 and 4.0 Ga (Warren, 2004). This clustering of impact melt breccia ages corresponds to an episode of intense crustal meta-

morphism defined by whole rock U-Pb isotopic compositions of lunar anorthosites (Tera and Wasserburg, 1974: Tera et al., 1974), a coincidence that led Tera et al. (1974) to infer "an event or series of events in a narrow time interval which can be identified with a cataclysmic impacting rate of the Moon at \sim 3.9 Ga." Turner et al. (1973) made a detailed comparison of 40 Ar $^{-39}$ Ar ages of lunar breccias and the photogeologic record of basin formation and concluded that several of the major impact features on the Moon and many smaller ones formed between 3.9 and 4.0 Ga. The idea that a significant spike in the mass flux to the Moon, and by extension the Earth, occurred at \sim 3.9 Ga was developed in greater detail by Turner and Cadogan (1975) and Turner (1979) and subsequently by G. Ryder and colleagues (Ryder, 1990; Stöffler and Ryder, 2001; Ryder, 2002; Ryder et al., 2002), who proposed that 15 of the major nearside lunar basins formed within an interval of ~ 100 to 200 million years (see also Wilhelms, 1987; Spudis, 1993).

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Whether or not the Earth and Moon experienced a cataclysmic bombardment of impacting planetesimals at \sim 3.9 Ga remains an open question with significant implications for understanding the impact history of the inner Solar System. An accurate reading of this impact history is important for several reasons, including a better understanding of the significance of large impact events for crust formation and biologic evolution on Earth (Ryder, 2002), establishing absolute timescales of geological events on other planets (Hartmann and Neukum, 2001; Neukum et al., 2001, 2004), and understanding planetary dynamics in the Solar System (Gomes et al., 2005; Strom et al., 2005).

The cataclysm hypothesis is controversial. An alternate interpretation of the lunar melt breccia age distribution is that a steadily declining impact flux quantitatively erased any record of older events (Hartmann, 2003). In this model, the apparent clustering of impact melt ages at \sim 4 Ga is the time at which the cratering rate dropped to a level sufficient to allow a significant fraction of the upper crust of the Moon to survive intact. Haskin et al. (1998) raised several objections to the notion of a cataclysmic bombardment, and presented cogent arguments that many of the mafic impact melt breccias in the Apollo lunar sample collection were created by the Imbrium event, the largest and youngest of the large nearside lunar basins. Haskin et al. (1998) suggested that the apparent range of ages from 3.8 to 4.0 Ga in these melt breccias reflects complexities within the samples rather than a real spread of ages.

To improve our understanding of the impact history of the Moon and provide an additional test of the cataclysm hypothesis, we measured ⁴⁰Ar-³⁹Ar ages on a suite of impact melt breccias from the central nearside lunar highlands, as sampled by the Apollo 16 mission. Virtually all of the rock samples collected at the Apollo 16 site were impact breccias with a wide range of compositions (Korotev, 1994) and petrographic characteristics (Norman, 2005). Most of the Apollo 16 crystalline melt rocks for which geochronological data exist have ages between 3.80 and 3.92 Ga (Turner and Cadogan, 1975; Dalrymple et al., 2001); a few older ages have been claimed (e.g., Maurer et al., 1978). One group of aluminous melt rocks, including samples 68415 and 68416 chipped from a homogenous boulder, are clearly younger than most other samples (3.75 Ga; Papanastassiou and Wasserburg, 1972). Some glassy melt particles and splash glass coatings are younger still, reflecting small-scale bombardment of the Moon that continues to the present day (Morris et al., 1986; See et al., 1986; Culler et al., 2000; Levine et al., 2005).

Much of the chronological data on Apollo 16 impact melts are derived from ⁴⁰Ar-³⁹Ar determinations made during the first decade of Apollo sample studies. Rb–Sr and Sm– Nd isochron ages are sparse (e.g. Papanastassiou and Wasserburg, 1972), mainly because it is difficult to separate phases from these finely crystalline rocks. Early ⁴⁰Ar-³⁹Ar radiometric ages on lunar impact melt rock are based on step-heating experiments with relatively few temperature release steps and much larger samples than used for the present study. Jessberger et al. (1974) conducted a 'high-resolution' study of whole rock and mineral separates of 65015 in which they used 30–50 heating steps, but more typically ages were based on 7–10 heating steps (e.g., Hueneke et al., 1973; Kirsten et al., 1973; Turner et al., 1973; Stettler et al., 1973; Maurer et al., 1978). The lack of age resolution in these early experiments is worsened by the large samples used by previous studies (typically 10–75 mg), in that undegassed clasts may have been included in the analyzed material (Jessberger et al., 1974). These complications often resulted in relatively large uncertainties (generally ± 0.04 to 0.05 Ga), and did not define any clear age distinctions among the chemical groups of Apollo 16 melt breccias (Korotev, 1994).

Because the Apollo and Luna landing sites are confined to the nearside equatorial region, returned lunar samples may present a biased view of lunar cratering history if they are dominated by ejecta from only a few of the more recent basins such as Imbrium and Serenitatis (Haskin et al., 1998). If this were so, the age distribution of lunar impact melts would not necessarily indicate a late cataclysmic bombardment of the Moon (e.g. Cohen et al., 2000). However, if the melt breccias from a single landing site can be demonstrated to be products of numerous separate and distinct impact events, as asserted for Apollo 15 melt breccias by Dalrymple and Ryder (1993), then this argument would not be valid.

We have improved the chronological database for Apollo 16 melt rocks by obtaining high-resolution (20–50 steps) 40 Ar– 39 Ar age spectra on 25 samples using continuous laser heating of sub-milligram fragments. The small size (≤ 1 mg) of the samples helps avoid undigested clasts, the large number of steps allows better determination of the structure of the gas release, and the large number of samples provides a useful set of internally consistent age data. These features of the current study potentially contribute to improved precision and clearer interpretation of impact melt breccia ages from the central nearside region of the Moon.

2. Methods

The analytical procedures are similar to those reported previously for Apollo 15 and Apollo 17 impact melt breccias (Dalrymple and Ryder, 1993, 1996). We obtained \sim 110 mg splits as interior chips from each rock. Using the neutron activation preparation clean room at Johnson Space Center, we inspected each chip under a binocular microscope to check for homogeneity, lack of obvious large clasts, and freshness of surface. Each chip was then gently crushed, and a few clast-free (as observable) homogenous particles weighing \sim 1 mg were selected for age determinations. The remainder of the sample was ground into a fine powder for other geochemical analyses.

The new radiometric ages were determined by ${}^{40}\text{Ar}{}^{-39}\text{Ar}$ incremental heating experiments at Oregon State University using a Merchantek 10W CO₂ continuous laser system with integrated IR temperature measurement, a Mass Analyzer Products 215/50 mass spectrometer, and methods similar to those in previous studies (Dalrymple and Ryder, 1993,

1996). Samples were irradiated in two batches, for 300 h each in the OSU TRIGA reactor (OSTR) using hornblende Mmhb-1 (513.9 Ma, Lanphere et al., 1990) as the fluence monitor. We used this monitor age for direct comparison with age determinations from Dalrymple and Ryder (1993, 1996), even though more recent intercalibrations place the age of Mmhb-1 at 521–523 Ma (Baksi et al., 1996; Renne et al., 1998). Using the older monitor age would result in a 1.5% increase in the ages reported here. The neutron capture efficiency factor *J* for each lunar sample was calculated from 4 total fusion measurements of 5–6 grains each of the monitor mineral irradiated at the same position as the unknown. The precision of *J* is approximately 0.5%.

Corrections for interfering Ar isotopes produced by undesirable reactions with Ca and K for the OSU TRIGA reactor were those determined previously (Renne et al., 1998; Koppers et al., 2003). The corrections for trapped Ar isotopes are the solar values, while those for cosmogenic Ar isotopes are from Hohenberg et al. (1978); atmospheric Ar is assumed to be negligible. The measured Ar isotopes were corrected for the system background using the results of blank measurements made at the beginning of each day and after every four or five unknown sample temperature steps, and are generally 1–2% of step gas concentrations for m/e = 37, 38, 39 and 40, and <4% for m/e = 36. All of the corrections are known with sufficient precision that they contribute negligible error to the age calculations for the lunar samples we analyzed.

Plateau ages are expressed as the weighted (by $1/\sigma^2$) mean of contiguous, concordant temperature step ages and comprise up to 95% of the total Ar released through the incremental heating experiment. The plateau description is a qualitative assessment of our confidence in interpreting the plateau age as a crystallization age, following Dalrymple and Ryder (1993, 1996). "Excellent" plateaus comprise >50% of the gas released, particularly in the mid- and high-temperature portion of the spectrum, and a majority of the step ages; "Good" plateaus comprise between 30% and 50% of the gas released, and include a significant portion of the many step ages; "Fair" plateaus comprise <30% of the gas released, or show irregular, near-plateaus of <50% of the gas released, over multiple step ages; "Poor" plateaus show only small portions of multiple-step, concordant ages, and complex release patterns. Errors on the plateau ages are reported at the 2σ level. K/ Ca spectra are calculated from the step 39 Ar/ 37 Ar (generated from nuclear reactions with 39 K and 37 Ca). These spectra are used to determine whether similar or dissimilar phases (varying K/Ca compositions) have provided the bulk of Ar released in important portions of the age spectra.

Interpreting plateaus as crystallization ages can be somewhat arbitrary; that is, the omission or inclusion of specific step ages in the weighted mean (see discussion in McDougall and Harrison, 1999). We follow established criteria (Dalrymple and Ryder, 1993, 1996; Dalrymple et al., 2001) in selecting steps and categorizing the quality (i.e., certainty) of the plateau ages. All spectra are disturbed, to greater or lesser degrees, which is why we have adopted a qualitative classification of our plateau ages. The physical basis for the complex argon behavior in some samples may lie in the fact that lunar impact melt breccias are crystalline rocks, not monophase glasses, such that less retentive phases (e.g., glassy mesostasis) might be partially disturbed by subsequent impacts, while more retentive phases (e.g., plagioclase, pyroxene) provide truer representations of the crystallization ages. This interpretation was favored by Jessberger et al. (1974, discussed in McDougall and Harrison, 1999). In addition, we recognize that occasional small unmelted crystals within these breccias may produce irregular step ages at high temperatures because they are not completely degassed from the impact event or they promote recoil effects. Argon loss, apparent in low temperature heating steps, may result from reheating events due to younger impacts.

While a perfect understanding of the complex argon systematics found in lunar breccias may not exist, there are several examples of lunar impact melt rocks that yield ⁴⁰Ar-³⁹Ar plateau ages identical to ages obtained by independent methods such as Rb-Sr and Pb-Pb mineral isochrons. In addition to the example of 65015 discussed in McDougall and Harrison (1999), similar results were obtained for 60315, 62295, and 68415 among others (see Ryder and Norman, 1980). These melt breccias all show lowtemperature disturbance in the Ar release spectra, yet their plateau ages appear reliable. Thus while our results cannot be interpreted in terms of simple diffusion models, we suggest that there is ample corroborating evidence to interpret the Ar plateaus as reasonable (but not necessarily perfect) representations of true crystalline ages, especially within the context of the 'plateau quality' caveats that we apply.

3. Samples

Twenty-five samples were selected for age determinations from all of the major chemical groups and some "unclassified" samples, as defined by Korotev (1994) based on major and trace element compositions. Where possible we analyzed multiple samples from each compositional group. The selected samples are all crystalline melt rocks; their textures range from vitrophyric through intersertal and ophitic–subophitic to poikilitic (Ryder and Norman, 1980).

4. Results

Results of 29 experiments (25 samples, 4 repeats) are summarized in Table 1 and Appendix A. The data are presented in the Electronic Annex. All of the gas-release profiles demonstrate some degree of post-crystallization reheating, probably by later impacts as evidenced by a few to many low-temperature step ages significantly younger than the plateau age. However, 23 of the 29 experiments produced precise, multi-step, mid- to high-temperature plateaus in the incremental heating age spectra that we interpret as crystallization ages. Commonly, the high-temperature portions of the age spectra show the effects of Ar recoil (Turner and

Table 1 Summary of event groups and ${}^{40}Ar - {}^{39}Ar$ incremental heating ages for Apollo 16 impact melt rocks

Sample	Al ₂ O ₃ wt%	Chemical Group (Korotev, 1994)	Plateau			Plateau age $Ma + 2\sigma$
			Description	% of total	Steps	$mu \pm 20$
Mafic Poikilitic						
60315	17.2	1 M	Good	42	9 of 34	3868 ± 31
64816	17.9	1 M	Good	50	11 of 30	3852 ± 12
69945		1 M	Excellent	68	27 of 47	3877 ± 11
62235	18.6	1F	Good	49	26 of 50	3876 ± 32
63596		1F	Excellent	67	19 of 30	3860 ± 13
65015	19.8	1F	Excellent	71	16 of 28	3854 ± 14
64568	22.9	2DB	Excellent	65	18 of 38	3867 ± 9
Mean age (1 SD, $n = 7$)						3865 ± 10
Aluminous subophitic						
63537		3n	Excellent	56	8 of 23	3838 ± 12
63545	22.2	2M	Fair	33	5 of 20	3839 ± 23
63549-1	29.0	3n	None	0	0 of 26	$>$ 3817 \pm 19
63549-2		3n	Excellent	67	15 of 34	3840 ± 11
64817	29.5	Unclassified	Excellent	76	8 of 18	3835 ± 18
65785	23.4	2NR	Poor	16	3 of 23	3826 ± 20
Mean age (1 SD, $n = 5$)						3836 ± 6
Aluminous poikilitic						
61156	22.9	2F	Good	42	7 of 28	3749 ± 36
61569	21.9	Unclassified	Excellent	58	16 of 25	3793 ± 13
Mean age $(n = 2)$						3771 ± 38
Troctolitic vitrophyre						
62295	20.5	2Mo	Good	58	10 of 22	3866 ± 12
64576	22.4	2Mo	Excellent	64	14 of 25	3852 ± 10
60666	18.5	2Mo	None	0	0 of 26	>3820
Mean age $(n = 2)$						3859 ± 16
Dimict						
61015	23.0	2DB	Fair	25	7 of 29	3899 ± 36
64585	22.1	2DB	Good	46	8 of 23	3962 ± 15
61225-1	_	2Md	Fair	36	10 of 32	3885 ± 36
61225-2		2Md	Good	48	10 of 22	3907 ± 15
Mean age (1 SD, $n = 4$)						3913 ± 34
Meta-poikilitic						
64815	18.9	Unclassified	Excellent	84	24 of 41	3886 ± 9
North Ray Crater				(2)	11 6 22	2005 - 20
63525-1		4m	Good	63	11 of 33	3895 ± 36
63525-2	—	4m	Excellent	95	24 of 28	4190 ± 24
63506-1	—	4s	None	0	0 of 23	>4000
63506-2		4s	Poor	8	4 of 32	>4043
Unclassified	24.0		. .	10	5 6 2 5	
66095	24.0	2DB	Fair	19	5 of 25	3676 ± 16
68519	23.5	2F	None	0	0 of 31	>3931

Al₂O₃ contents from the compilations of Korotev (1994), Ryder and Norman (1980), McKinley et al. (1984), Ryder and Seymour (1982).

Cadogan, 1975), which in very fine-grained, multi-phase material results from redistribution of ³⁹Ar from relatively K-rich to K-poor phases, or ³⁷Ar from relatively Ca-rich to Ca-poor phases. In a smaller number of samples we also observe evidence in the very few highest temperature steps of undegassed clasts that produce older than plateau ages. In this section, we present these results organized according to Korotev's (1994) compositional classification of Apollo 16 impact melt rocks. In the Section 5 we consider possible alternative grouping based on petrography, age groups, and bulk compositions.

4.1. Group 1

Six samples from Group 1 (incompatible element-rich poikilitic melt breccias) were analyzed. These include three examples of each subgroup 1M (mafic; 60315, 64816, and 69945) and 1F (felsic; 65015, 62235, and 63596). All samples yielded plateau ages that we considered either 'good' or 'excellent'. Both subgroups yielded similar ages (3852–3877 Ma for 1M, 3854–3876 Ma for 1F); 2σ measurement uncertainties on four of the six samples were ±11–14 Ma, and ±31–32 Ma on the remaining two samples. Represen-



Fig. 1. Ar release spectra for Apollo 16 impact melt breccias 69945 and 65015 representing the mafic poikilitic group. Korotev (1994) classified these samples as Group 1M and 1F, respectively, based on their chemical compositions. Their similar ages and petrographic characteristics suggest that all of these breccias were derived from the same impact melt deposit.

tative age and K/Ca spectra are shown in Fig. 1. These samples produced easily interpreted, broad plateaus with small amounts of Ar recoil at the highest temperatures of gas release. The K/Ca compositions decrease smoothly over a restricted range, consistent with degassing from a fine-grained mineral assemblage. The unweighted mean ages for Subgroups 1M and 1F are 3866 ± 13 and 3863 ± 11 Ma, respectively, with the uncertainty on the mean calculated as 1 SD of the individual determinations. The unweighted mean age for all Group 1 samples is 3865 ± 11 Ma (1 SD, n = 6).

4.2. Group 2

Group 2 melt breccias have higher Al_2O_3 and lower incompatible lithophile element abundances compared to Group 1. Korotev (1994) recognized several subgroups based on their petrographic and lithologic associations, and subtle differences in trace element compositions. Argon release spectra of Group 2 samples yield age plateaus ranging in quality from 'excellent' to 'fair.' The four samples classified as Group 2DB based on their chemical compositions (61015, 64568, 64585, and 66095) yielded ${}^{40}Ar - {}^{39}Ar$ ages that define the entire range observed in this study, from 3676 ± 16 Ma for 66095 to 3962 ± 15 Ma for 64585 (Table 1). For only one of these samples (64568), however, was the Ar release plateau considered to be 'excellent' (Table 1) with complex spectra obtained from the other three samples (Fig. 2). Samples 62295 and 64576, both assigned to the 2Mo troctolitic group, produced clear plateaus at 3866 ± 12 and 3852 ± 10 Ma, respectively, with Ar recoil at higher temperature steps (Fig. 3). Two Group 2 samples (60666, 68519) produced strong Ar loss patterns from which only minimum ages could be estimated. In the Section 5 we present a possible alternative classification of Group 2 samples based on their Ar ages and petrographic characteristics.

4.3. Group 3

Samples classified as Group 3 by Korotev (1994) have lower absolute abundances of incompatible lithophile trace elements, lower Sm/Sc ratios and higher Al₂O₃ than Groups 1 and 2. Korotev (1994) recognized two subgroups (3n, 3s) based on siderophile element ratios (Ir/Au, Ir/Ni) and REE contents. We analyzed two samples from Group 3n (63537, 63549). Splits of both samples yielded excellent plateau ages within uncertainty of each other (3838–3840 \pm 11–12 Ma), although a replicate split of 63549 did not give a plateau age (Supplementary table). The age and K/Ca spectrum for sample 63549-2 are shown in Fig. 4. In this example, a clear plateau is formed (3840 \pm 11 Ma) followed by a small recoil-affected portion of the highest temperature steps. The K/Ca decreases smoothly for the plateau steps as seen in other well-behaved samples.



Fig. 2. Ar release spectra for Apollo 16 impact melt breccias 64568 and 64585 representing the compositional Group 2DB of Korotev (1994). The large age differences measured for the four Group 2DB samples suggest that multiple impact melt deposits are represented in this compositional group.



Fig. 3. Ar release spectra for Apollo 16 impact melt breccias 62295 and 64576 representing the compositional group 2Mo of Korotev (1994). The similar ages and petrographic characteristics suggest Group 2Mo samples were derived from the same impact melt deposit.

4.4. Group 4

Group 4 melt breccias are predominantly from the North Ray Crater area of the Apollo 16 landing site. They are highly aluminous with very low abundances of incompatible lithophile trace elements (Korotev, 1994), and they tend to be finer-grained and richer in clasts than the other Apollo 16 melt rocks (Ryder and Norman, 1980). We analyzed two samples representing Group 4; both yielded equivocal results. Replicate splits of 63506 yielded no reliable age plateaus and showed evidence for both older (≥ 4



Fig. 4. Ar release spectra for the Apollo 16 impact melt breccia 63549-2 representing the aluminous subophitic group of breccias recognized by this study. The similar ages and petrographic characteristics suggest this group of melt breccias samples were derived from the same impact melt deposit.

Ga) and younger (≤ 3 Ga) events in the age spectra (Fig. 5; Appendix A). In contrast, replicate splits 63525 yielded plateau ages regarded as "good" and "excellent," respectively (Fig. 5; Appendix A), but these ages differed considerably from one another (3895 vs. 4190 Ma; Table 1).

4.5. Unclassified

Three samples that Korotev (1994) referred to as 'unclassified' (64815, 64817, and 61569) yielded excellent age plateaus of 3886 ± 9 , 3835 ± 18 , and 3793 ± 13 Ma, respectively. The age and fine-grained subophitic texture of 64817 are both similar to the two Group 3n samples (63537 and 63549), whereas the composition and age of 61569 resembles that of sample 61156 more closely than the Group 1 poikilitic breccias.

5. Discussion

Twenty-one of the 25 samples of Apollo 16 impact melt breccias produced multi-step plateaus that we interpret as crystallization ages. Of these, 20 fall in the range 3.75– 3.96 Ga. This confirms the strong concentration of lunar impact rock ages within a relatively narrow interval of ~200 million years. Central to evaluating the significance of this apparent spike in lunar impact melt rock ages is



Fig. 5. Ar release spectra for the Apollo 16 impact melt breccia 63506-2 and 63525-2 representing compositional Group 4 of Korotev (1994). The ages of Group 4 breccias are not well determined due to the complex release spectra of these samples, perhaps suggesting incomplete degassing and/or later disturbance of the argon systematics in these breccias.

the clear identification of distinct impact events within the overall group of lunar impact breccias. We propose that at least four separate impact events can be recognized within the Apollo 16 impact melt rocks based on their ages and textures (Figs. 6 and 7). The event groups proposed here correspond in a general way to the compositional groups recognized by Korotev (1994) with some modifications based on the new ages and petrographic characteristics of the melt breccias. To avoid complicating the lunar melt breccia classification scheme unnecessarily, here we refer to the event groups by general terms reflecting key petrographic and chemical characteristics of the melt breccias that we assign to these groups, e.g., mafic poikilitic, aluminous subophitic, aluminous poikilitic, and troctolitic vitrophyre (Figs. 6 and 7).

5.1. Mafic poikilitic group (3865 Ma)

Based on their similar ages (Table 1), petrographic characteristics (Bence et al., 1973; Simonds et al., 1973), and restricted range of major and trace element compositions (Korotev, 1994), we propose that all six samples of the Group 1 melt rocks formed in the same impact event, to which we assign an age of 3865 ± 10 Ma (Table 1). Considering its poikilitic texture, mineralogy, and well-defined age



Fig. 7. Summary diagram showing ${}^{40}\text{Ar}{}^{-39}\text{Ar}$ ages of Apollo 16 impact melts determined by this study grouped according to petrologic type. The stippled boxes indicate the 1σ standard deviation around the mean ages for the groups (Table 1). We propose that each of these petrologic-age groups represent a discrete impact event. The Group 4 (North Ray Crater) and unclassified samples (Table 1) are not plotted.

 $(3867 \pm 9 \text{ Ma})$, we have also assigned sample 64568 to the mafic poikilitic event group even though it was classified as Group 2DB by Korotev (1994).



Fig. 6. Photomicrographs illustrating the textures of four groups of Apollo 16 impact melt breccias proposed to represent discrete impact events based on their ages (Table 1), textures, and compositions. (a) 60315, mafic poikilitic group (mean age 3865 ± 10 Ma). Field of view 1×2 mm. (b) 63549, aluminous subophitic group (mean age 3836 ± 6 Ma). Field of view 1×2 mm. (c) 61569, aluminous poikilitic group (mean age 3771 ± 38 Ma). Field of view 2×4 mm. (d) 62295, troctolitic vitrophyre group (mean age 3859 ± 16 Ma). Field of view 1×2 mm.

5.2. Aluminous subophitic group (3836 Ma)

Samples 63537, 63545, 63549, and 64817 have clast-poor. fine-grained subophitic textures (Fig. 6) and well-defined ages (3835–3840 Ma) that are younger than the Group 1 samples (Fig. 7). Considering the subophitic texture, bulk composition, and young (albeit poorly defined) age of 65785, we tentatively assign this sample to the aluminous subophitic event group as well. The bulk compositions of these samples are somewhat variable (23–29% Al₂O₃), which is reflected in Korotev's (1994) classification of 63545 as Group 2m, 63537 and 63549 as Group 3n, 65785 as 2NR, and 64817 as unclassified. Based on the tight clustering of ages and similar petrographic characteristics of these breccias, however, we propose they are all samples of the same impact melt sheet, to which we assign an unweighted mean age of 3836 ± 6 Ma (1 SD, n = 5) (Table 1). Excluding the poorly defined result for 65785 produces a mean age of 3838 ± 2 Ma.

5.3. Troctolitic vitrophyre group (3859 Ma)

Samples 62295, 64576, and 60666 have vitrophyric textures (Fig. 6), troctolitic (olivine + plagioclase) mineralogies, and highly magnesian bulk compositions that are unusual for Apollo 16 impact melts. Korotev (1994) recognized their distinctive compositions and classified all of these samples as 2Mo (olivine-rich). The 40 Ar $^{-39}$ Ar release patterns of 62295 and 64576 are well-behaved and yield similar ages of 3866 ± 12 and 3852 ± 10 Ma (Fig. 3), respectively. Sample 60666 yielded only a minimum age of >3820 Ma. The ages of 62295 and 64576 are within the range obtained for the Group 1 mafic poikilitic breccias (Fig. 7), but due to their unique bulk compositions and textures, we assign the troctolitic vitrophyre (Group 2Mo) breccias to a separate event group with an unweighted mean age of 3859 ± 16 Ma (Table 1).

5.4. Aluminous poikilitic group (3771 Ma)

Samples 61156 and 61569 have poikilitic textures (Fig. 6) but they are chemically and texturally distinct from the Group 1 mafic poikilitic breccias (Simonds et al., 1973). Both samples have less pyroxene and more olivine than the Group 1 poikilitic rocks (Albee et al., 1973; Simonds et al., 1973), and they have more aluminous bulk compositions and lower abundances of incompatible trace elements than the mafic poikilitic breccias (Simonds et al., 1973; Korotev, 1994). Sample 61156 was classified as Group 2F by Korotev (1994), whereas 61569 was unclassified due to compositional heterogeneity among the analyzed splits. We tentatively assign these two samples to a single event group with a mean age of 3771 ± 38 Ma, which is younger than the inferred age of 3865 ± 10 Ma for the Group 1 mafic poikilitic breccias (Fig. 7). The small number of samples and less distinctive petrographic and chemical characteristics of the aluminous poikilitic breccias makes this event assignment less robust than the other three groups discussed above.

5.5. Dimict breccias (3913 Ma)

The four melt breccis classified as Group 2DB by Korotev (1994) that we analyzed (61015, 64568, 64585, and 66095) have diverse ages (3676–3962 Ma) and textures. Although Korotev's Group 2DB nominally represents the distinctive Apollo 16 'black and white' or dimict (melt breccia + anorthosite) breccias, in fact only one of our Group 2DB samples (61015) was taken from a rock with the characteristic physical intermingling of melt breccia and anorthosite lithologies that characterizes the typical 'dimict breccias' (Ryder and Norman, 1980).

We suspect that the four Group 2DB samples analyzed for this study are not related to a single event group, and additional studies of dimict breccias selected based on macroscopic lithologic characteristics are needed. However, the fine-grained, mesostasis-rich, subophitic texture of 64585 appears similar to that of the melt portion of other dimict breccias, including 61015, and a number of samples classified as dimict breccias (Ryder and Norman, 1980) collected at Apollo 16 Station 4 along with 64585. The ⁴⁰Ar-³⁹Ar ages of 61015 (3899 \pm 36 Ma) and 64585 (3962 \pm 15 Ma) are among the oldest observed in Apollo 16 melt breccias, and we tentatively propose that dimict breccias represent one of the oldest melt breccia groups sampled at the Apollo 16 site (Fig. 7). The age of replicate splits of 61225 (3907 \pm 15 and 3885 ± 36 Ma, respectively), previously classified as group 2Md by Korotev (1994), and the thick coating of adhering anorthositic debris present on exterior surfaces of this sample (Ryder and Norman, 1980) may point toward affinities with the dimict breccias, although a detailed petrographic description of this sample is not available. We tentatively assign an unweighted mean age of 3913 ± 34 Ma (1 SD, n = 4) to the dimict breccia group (Table 1).

Samples 64568 and 66095 were classified as Group 2DB by Korotev (1994) but they appear to have petrographic and age characteristics unlike to those of the typical dimict breccias. As discussed above, sample 64568 has more affinities with the mafic poikilitic breccias than the other Group 2DB samples. The Ar release pattern of 66095 ('Rusty Rock') indicates substantial disturbance and the young age obtained from a relatively small plateau (five steps representing 19% of the total ³⁹Ar released) may not be a reliable crystallization age.

5.6. Old ages at North Ray Crater?

Apparent ages >4.0 Ga in the highly aluminous, clastrich Group 4 breccias are enigmatic due to the lack of useful plateaus in sample 63506, and the poor reproducibility among the replicate splits of 63525. Similarly old ages have also been reported for melt breccia clasts associated with North Ray Crater fragmental breccias (Maurer et al., 1978; Stöffler et al., 1985; Duncan and Norman, 2005), but there is no consensus on the interpretation of these ages. They may represent an older impact event, or (perhaps more likely?) incomplete degassing of clastic debris incorporated in these melts, and/or slow diffusion in these highly aluminous melt rocks.

5.7. Sample 64815

Breccia 64815 appears to be a unique sample. It has a poikilitic texture but experienced a greater degree of postcrystallization annealing than the Group 1 mafic poikilitic breccias (Simonds et al., 1973; Ryder and Norman, 1980). Korotev (1994) recognized the compositional uniqueness of 68415 among Apollo 16 impact melt breccias, primarily due to its high Ti, Sc, Cr, and HREE abundances. It has a well-defined age that is somewhat older (3886 \pm 9 Ma) than both the mafic (3865 Ma) and the aluminous (3771 Ma) poikilitic groups defined here (Fig. 7). The significance of this sample is not apparent; it may represent still another discrete impact event.

5.8. Compositional variations in lunar impact melt breccias

Impact melt sheets at terrestrial craters are remarkably homogeneous in major and trace element compositions. Major element variations within terrestrial impact melt sheets typically are similar in magnitude to analytical precision, and lithophile trace element dispersions are $\sim 10\times$ less than that of the country rocks (Reimold et al., 1990 and references therein). It is often assumed that co-genetic lunar impact melt breccias should display a similar degree of compositional homogeneity (e.g., Spudis and Ryder, 1981) although the actual variability of major and trace element compositions within lunar impact melt deposits is unknown.

One implication of our proposed grouping of lunar impact melt breccias based on 40 Ar– 39 Ar ages and petrographic characteristics is that lunar melt breccia deposits can show a considerably greater internal major and trace element variability than would be predicted based on terrestrial crater analogues. For example, our conclusion that all of the Group 1M and IF (mafic poikilitic) samples are related to a single event implies a compositional range of ~17–22 wt% Al₂O₃ in these melt breccia deposits, whereas the aluminous subophitic melt breccias have Al₂O₃ contents ranging from 23 to 29 wt%. This corresponds to major element variability of ~20 to 30% relative compared to 2–5% variation in terrestrial melt sheets (Floran et al., 1978; Reimold et al., 1990).

Some of this compositional variation in lunar melt breccias might reflect non-representative sampling of incompletely digested clasts (Korotev, 1994), although the possibility that lunar impact melt deposits are inherently more heterogeneous than the coherent melt sheets within terrestrial craters must be considered a realistic possibility (Spudis, 1993, pp. 172–174). In particular, the extent of compositional variability between a melt sheet preserved within a crater and discontinuous melt ejecta deposits emplaced outside of a crater is difficult to evaluate, in part because erosion has removed the melt ejecta deposits from most large terrestrial impact craters, and because of the limited field data available for individual lunar craters. Additional studies of clast compositions and abundances in lunar melt breccias may help clarify these relationships.

6. Conclusions

High-resolution 40 Ar ${}^{-39}$ Ar age spectra have been measured on 25 samples of Apollo 16 impact melt breccias using a continuous laser heating system on sub-milligram fragments. Twenty-one of these 25 breccias produced multi-step plateaus that we interpret as crystallization ages, with 20 of these ages falling in the range 3.75–3.96 Ga. This confirms the strong concentration of lunar impact rock ages within a relatively narrow interval of ~200 million years.

At least four different melt-producing impact events are identified based on groupings defined by similarities in age and petrographic characteristics. Members of these proposed event groups also share compositional characteristics, although the range in compositions within each group tends to be somewhat greater than classification schemes based on major and trace element geochemistry alone (Korotev, 1994). The range of major and trace element compositions observed in melt breccia groups defined by petrography and age may provide a better measure of compositional variations in lunar impact melts than assumed analogies with terrestrial melt sheets, which tend to be compositionally homogeneous.

The recognition of multiple impact events within the suite of Apollo 16 melt breccia supports the conclusion that numerous impact events occurred on the lunar surface within a relatively narrow time interval, providing additional evidence of a heavy bombardment of the Moon (cataclysm), and presumably the Earth, during the period 3.75–3.96 Ga.

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Appendix A

40Ar/39Ar radiometric dating plateau and K/Ca diagrams for 25 Apollo 16 lunar impact melt rocks. Includes 4 pairs of repeat experiment results for samples 61225, 63549, 63506 and 63525.

















Appendix B. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.gca.2006. 05.021.

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