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Zircon formation in impact melts: Complications for deciphering planetary impact histories

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ABSTRACT

We explore the formation conditions and inheritance probability of zircon in impact melts and the implications of using zircon geochronology to investigate planetary impact histories. By modeling the occurrence and crystallization temperature spectrum for zircon in simulated impact melts, we predict the presence of such grains within impactites. We also report U-Pb geochronology of sieve-textured, possibly poikilitic, zircon identified in the pseudotachylyte and granophyre units present within the largest known terrestrial impact crater (Vredefort, South Africa) to explore the accuracy of these grains in dating impact events at an impact structure of known age. Zircons with similar textures have been recently interpreted as growing in an impact melt in lunar meteorite SaU 169 and used to determine the age of the Imbrium impact. Modeling in simulated lunar melt compositions predicts crystallization of zircon in merely ~2% of melting events, in this case via impact. The modeled crystallization temperature spectrum is significantly below Ti-in-zircon crystallization temperatures reported from lunar samples. Zircon formation within an impact melt is dictated by saturation of [Zr] and requires a high abundance for lunar melt compositions. This essentially rules out the possibility of zircon growing in equilibrium with lunar meteorites. Poikilitic textures may be inherited from the lunar crust, presumably due to rapid decompression and/or resorption into an undersaturated magma, as previously recognized in plagioclase. Although either scenario could be due to an impact, endogenic processes cannot be ruled out, and thus lunar poikilitic zircons may not be recording impact melting events. Secondary ion mass spectrometry U-Pb analysis of zircon with similar textures from Vredefort clearly shows that these grains are inherited from the Archean target rocks, with varying degrees of Pb loss, and consequently cannot be used to accurately identify the age of the Vredefort impact structure. Further understanding of the growth and isotopic effects on zircon of shock and heating associated with large impacts could provide another tool that can be used to probe planetary impact histories.

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INTRODUCTION

The intensity of impact bombardment on early Earth has major implications for: (1) the atmosphere (Ahrens, 1993), (2) habitability (Cronin and Pizzarello, 1983; Abramov and Mojzsis, 2009), (3) near-surface environments (Abramov et al., 2013), and (4) volatile budget (Albarède, 2009). Because the early bombardment history of Earth has been all but erased, the lunar surface is widely seen as retaining the most complete impact record in the inner solar system (Fassett and Minton, 2013), and its study is given the highest priority for lunar scientific exploration (Paulikas et al., 2007). However, the few preserved large (~100 km) impact craters formed over the past 2 b.y. offer, via geochemical signatures in rocks and minerals, insights into the physical conditions (i.e., shock pressure, temperatures, etc.) experienced during bombardment and permit modeling of impact melt processes (Wielicki et al., 2012a). Such analysis provides a foundation with which to make interpretations of impact histories from lunar samples and meteorites.

BACKGROUND

The impact history of the solar system remains controversial. Early investigation of lunar samples led to the Late Heavy Bombardment concept (Turner et al., 1973; Tera et al., 1974), which hypothesized a sharp increase in bolide flux centered around ca. 3.95 Ga. Evidence for this cataclysm was first derived from whole-rock U-Pb and Rb-Sr ages (Tera et al., 1974), and recent research has relied on interpretations of ^{40}Ar-^{39}Ar lunar sample data (*Apollo* and lunar meteorites; Dalrymple and Ryder, 1993; Cohen et al., 2000). However, lunar ^{40}Ar-^{39}Ar geochronology can be problematic due to the presence of relic clasts, incomplete Ar outgassing, diffusive modification during shock and heating, and exposure to solar wind and cosmic rays (Harrison and Lovera, 2013). Recent studies have focused on U-Pb geochronology in zircon as a new tool with which to identify ancient large-scale impact events, such as the Late Heavy Bombardment, on the Earth-Moon system. For example, ancient terrestrial zircon grains from Western Australia have been interpreted to show evidence of thermal excursions related to Late Heavy Bombardment–era impacts (Trail et al., 2007; Abbott et al., 2012; Bell and Harrison, 2013). Lunar zircons have also been inferred as being created by large-scale impacts on the Moon and may be an important means with which to address scientific goals of future lunar sample return missions (Crawford et al., 2012).

Poikilitic zircon, which appears as branching, interstitial networks of zircon enclosing other phases (Figs. 1G and 1H), within the melt matrix from lunar meteorite SaU 169 and *Apollo 12* samples (Gnos et al., 2004; Liu et al., 2012; Grange et al., 2013) have been interpreted to have grown during equilibrium crystallization of the impact melt. Determining the age of these grains has been suggested as a means to date impact events. Results of such analyses have been offered as constraints on the age of the Imbrium Basin and as such would introduce an important new means with which to probe planetary impact histories (Liu et al., 2012; Grange et al., 2013). Although poikilitic textures have been observed in plagioclase and other phases, they are rare in terrestrial environments (Scoates and Chamberlain, 1995). We examine the likelihood of zircon crystallization within simulated lunar impact events, particularly the formation of poikilitic zircon within lunar meteorite SaU169, and we report secondary ion mass spectrometry (SIMS) U-Pb geochronology of similar-textured zircons found within the melt matrix of the largest terrestrial impact (Vredefort, South Africa) to test the hypothesis that such grains crystallized within an impact melt.

SAMPLE

The Vredefort impact structure in South Africa is the largest known terrestrial impact (Earth Impact Database, 2014 [www.passc.net/EarthImpactDatabase/, accessed August 2014]) and represents the deeply eroded remnant of a ca. 2.0 Ga (Kamo et al., 1996), ~300-km-wide crater (Reimold and Gibson, 1996). Although the impact melt sheet has been eroded, pseudotachylyte breccia and granophyre veins remain. Samples of pseudotachylyte breccia (VD_PB) collected near the quarry at Leeukop, just west of Parys, and granophyre (VD_G) sampled near the Kommandonek Nature Preserve (provided by Roger Gibson, University of Witwatersrand, and W. Uwe Reimold, Humboldt-University Berlin; Gibson and Reimold, 2005) were used for analysis (for sample locations, see Wielicki et al., 2012a). Pseudotachylyte breccia sample VD_PB consists of 1–3 cm clasts of Archean granite to granodioritic gneiss within a fine-grained crystalline matrix (Reimold and Gibson, 2006). Granophyre sample (VD_G) consists of fine-grained crystalline quartz, plagioclase, and alkali feldspar with long laths of hypersthene and small grains of magnetite (Reimold and Gibson, 2006). Zircon grains from both samples average ~100 µm and tend to be fractured. Crystals lack igneous oscillatory zoning in cathodoluminescence (CL) images, and those from the granophyre are intimately intergrown with an Mg-rich pyroxene identified by energy dispersive X-ray analysis (Figs. 1A–1H).

METHODS

Wielicki et al. (2012a) showed that zircon crystallization temperatures from terrestrial impact melts are consistent with the Zr-saturation model for silicate melts (Harrison and Watson, 1983; Watson and Harrison, 1983, 2005; Boehnke et al., 2013) and are a simple function of [Zr], temperature, and rock chemistry (i.e., $M = [2Ca + Na + K]/[Al \cdot Si]$). Thus, knowledge of M, [Zr], T' (i.e., ambient temperature plus the ΔT associated with impacts), and the relationship between M and melting conditions permits any random association of target rock compositions and possible T' to be evaluated for the potential to produce impact zircon (for detailed model description, see Wielicki et al., 2012a). We have further developed this approach to predict both the probability of zircon growth in anhydrous impact melt sheets of

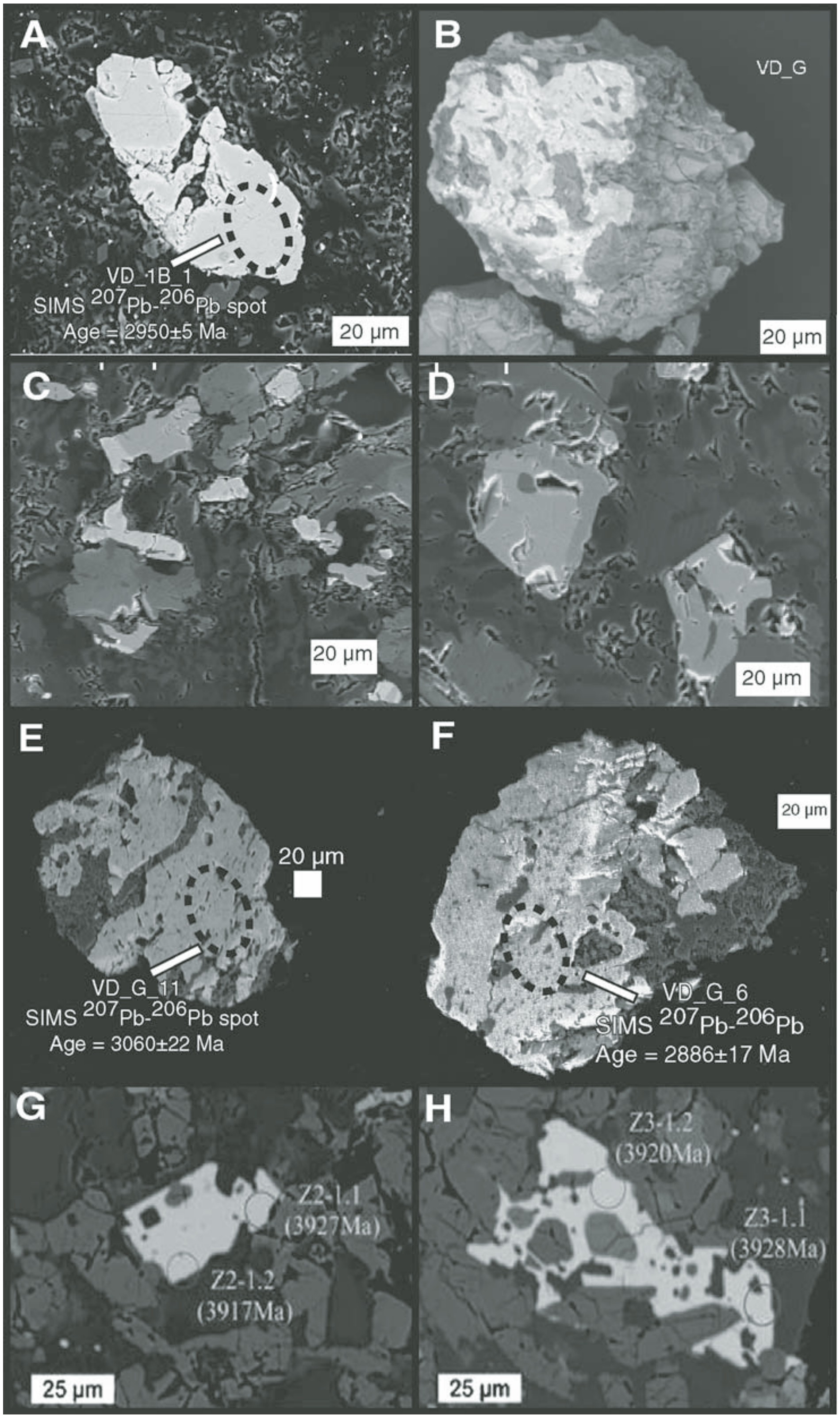

Figure 1. Backscattered-electron images of (A) in situ VD_PB zircon and (B–F) VD_G zircon compared to (G–H) images of lunar meteorite SaU 169 (Liu et al., 2012), highlighting similar poikilitic textures. See text for further description.

TABLE 1. ION MICROPROBE U-Pb ZIRCON ISOTOPIC DATA FROM VREDEFORT GRANOPHYRE AND PSEUDOTACHYLYTE BRECCIA

Sample: Vredefort	$^{207}Pb/^{235}U$ value	1 s.e.	$^{206}Pb/^{238}U$ value	1 s.e.	Correlation of concordia ellipses	$^{206}Pb/^{238}U$ age (Ma)	1 s.e. (Ma)	$^{207}Pb/^{235}U$ age (Ma)	1 s.e. (Ma)	$^{207}Pb/^{206}Pb$ age (Ma)	1 s.e. (Ma)
VD_1A_1	15.75	0.7077	0.4974	0.0221	0.9988	2600	95	2860	42.9	3050	3.6
VD_1A_2	12.19	0.6299	0.4338	0.0222	0.9942	2320	99.8	2620	48.5	2860	9.1
VD_1A_3	9.209	0.4496	0.3528	0.0173	0.9955	1950	82.5	2360	44.7	2740	7.7
VD_1A_4	12.86	0.8148	0.4668	0.028	0.99	2470	123	2670	59.7	2820	15.3
VD_1A_5	16.04	0.9469	0.5368	0.0308	0.9937	2770	129	2880	56.4	2960	10.8
VD_1B_1	15.8	1.077	0.5314	0.0361	0.9992	2750	152	2870	65.1	2950	4.3
VD_1B_3	11.72	0.416	0.4373	0.0161	0.9848	2340	72	2580	33.2	2780	10.5
VD_1B_4	10.01	0.4831	0.3557	0.0166	0.9869	1960	78.9	2440	44.5	2860	12.8
VD_G_1	13.19	1.052	0.4812	0.0362	0.9867	2694	75.3	2532	157.6	2817	21.9
VD_G_2	14.06	1.323	0.5256	0.0435	0.9321	2754	89.2	2723	183.9	2777	56.4
VD_G_3	11.91	0.6746	0.4931	0.025	0.9439	2597	53.1	2584	108	2607	31.5
VD_G_5	11.76	0.6246	0.474	0.0229	0.9441	2586	49.7	2501	100.3	2652	29.2
VD_G_6	7.386	0.3061	0.3269	0.0109	0.8812	2159	37.1	1823	53.1	2496	33.4
VD_G_7	15.97	0.8252	0.5435	0.0294	0.9908	2875	49.4	2798	122.9	2930	12.3
VD_G_8	14.7	0.6422	0.514	0.0201	0.975	2796	41.5	2674	85.4	2886	16.8
VD_G_9	9.948	0.6674	0.4372	0.0226	0.8646	2430	61.9	2338	101.4	2508	57.7
VD_G_10	22.68	1.707	0.6698	0.0459	0.9932	3213	73.2	3305	177.4	3156	17
VD_G_11	18.7	1.475	0.5867	0.043	0.986	3026	76.1	2976	174.7	3060	22.3
VD_G_12	18.65	1.027	0.6002	0.0302	0.9893	3024	53.1	3031	121.5	3019	14.6
VD_G_13	14.32	0.7285	0.5473	0.0274	0.9789	2771	48.3	2814	114.1	2740	17.1

lunar composition and their crystallization temperature spectra. Because lunar rocks have distinctive compositions, we utilized the solubility model of Dickinson and Hess (1982) derived specifically for lunar compositions. To simulate lunar target chemistry, we used a wide range of *Apollo* and lunar meteorite compositions. We used the selenotherm of Dyal et al. (1973) together with the Late Heavy Bombardment–like impact thermal perturbation spectrum of Abramov and Mojzsis (2009) to assign target rock temperatures. Although the Abramov and Mojzsis (2009) model was developed for Earth, the relative thermal effects on the terrestrial and lunar lithospheres scale linearly. Thus, each rock is associated with an M and [Zr], a calculated solidus (T_{sol}) and liquidus (T_{liq}) temperature (i.e., via the M vs. T relationships), and a value of T'. Note that M will be much lower than the bulk rock value for low melt fractions; this will be particularly pronounced for mafic compositions, thus requiring parameterization of M with melt fraction (based on synthetic experiments using MELTS; Ghiorso and Sack, 1995). The following logical statements were then applied to each of the collective data sets in the sequence:

(1) Did the rock melt due to impact (i.e., is $T' > T_{sol}$)? If not, the event does not produce magmatic zircon. If true, T' is recorded, and the calculation continues.

(2) Is $T^{sat}_{zir} < T_{sol}$? If true, the whole-rock composition will not saturate zircon, and the calculation terminates, unless $T' > T_{liq}$, in which case, we assume the melt differentiates and thus produces zircon with $T^{xtln}_{zir} = T^{sat}_{zir} + 50$ °C if $> T_{sol}$ (i.e., we account for the high-temperature onset of zircon saturation in crystallizing intermediate to mafic melts by adding the conservative lower limit of the observed 50–100 °C difference between T^{xtln}_{zir} and T^{sat}_{zir}; see Harrison et al., 2007). If $T^{sat}_{zir} > T_{sol}$ and $T_{sol} \leq T' \leq T^{sat}_{zir}$, then T' is taken to be T^{xtln}_{zir}.

(3) In cases where $T_{sol} < T' < T_{liq}$, we scaled M according to the relationships $M' = 0.4M$ if $T_{sol} < T' < (T_{liq} - T_{sol}) \times 0.5 + T_{sol}$, or $M' = 0.8M$ if $T_{sol} < T' > (T_{liq} - T_{sol}) \times 0.5 + T_{sol}$. (These relationships approximate the compositional response of the Wyllie [1977] rock analyses to melting using the MELTS algorithm.)

Only ~2% within each run batch resulted in conditions permitting zircon formation. Thus, in order to attain a robust result, we executed the algorithm 1000 times.

For U-Pb SIMS analysis, thin sections were first examined using a LEO 1430 VP scanning electron microscope (SEM) to assess zircon size and abundance. Mineral separates were obtained from bulk rock samples by standard heavy liquid separation procedures. Separated zircon crystals were also imaged (Fig. 1) and then handpicked and mounted in 2.54-cm-diameter epoxy mounts together with AS3 zircon standard (1099 ± 1 Ma; Paces and Miller, 1993). Ion microprobe analyses were conducted with a CAMECA ims1270, at University of California–Los Angeles, using an ~8–12 nA mass-filtered $^{16}O^-$ beam focused to spots between ~20 and 35 μm (Compston and Williams, 1984). In situ ion microprobe analysis on thin sections permits data collection from grains present within the impact melt and ensures avoidance of inherited clasts that could contribute to the separated samples. All analytical results are reported in Table 1.

RESULTS

Modeled crystallization temperature spectra for zircon growth in lunar impact melts indicate that zircon crystallizes in only ~2% of the 1000 simulations, reflecting the high [Zr] necessary to nucleate zircon in the predominantly mafic compositions (Boehnke et al., 2013) present on the lunar surface. Interestingly, model temperatures, which range from ~850 °C to 1050 °C, are typically ~100–200 °C higher than those seen by Wielicki et al. (2012a) for the terrestrial case, presumably reflecting the anhydrous nature of lunar melts, although they are significantly lower than Ti-in-zircon crystallization temperatures reported for lunar grains (Taylor et al., 2009; Fig. 2).

In situ U-Pb geochronology of zircon grains (8 of 14 grains were large enough to analyze; Fig. 1A) within the melt matrix of the pseudotachylyte breccia (VD_PB) yielded ages of ca. 3 Ga. As this is nearly ~1 b.y. older than the known age for this impact event, these grains are presumably inherited from the Archean target. The grains record no evidence for a thermal event associated with the Vredefort impact (Fig. 3A). Grains show Pb loss broadly correlated with postimpact thermal events associated with the Kibaran orogeny at ca. 1.1 Ga (Reimold et al., 2000; Erickson et al., 2013).

Zircons isolated from the granophyre unit (VD_G) of the Vredefort impact show an intimate relationship with Mg-rich pyroxene (i.e., poikilitic texture) similar to that discovered in

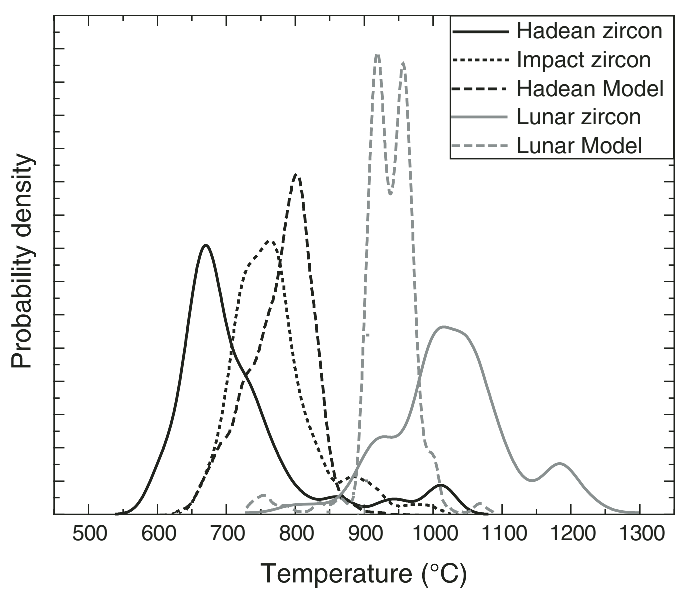

Figure 2. Modeled crystallization temperature spectrum for zircons within simulated impact events on the lunar surface. Predicted zircon crystallization temperatures are significantly higher than reported Ti-in-zircon crystallization temperatures within lunar impact melts, suggesting that these grains did not form within lunar impact melts. Also shown are modeled results for impacts on an ancient terrestrial crust as compared to Hadean zircon from the Jack Hills, Western Australia, and zircon grown in large terrestrial impact melts (Wielicki, 2011).

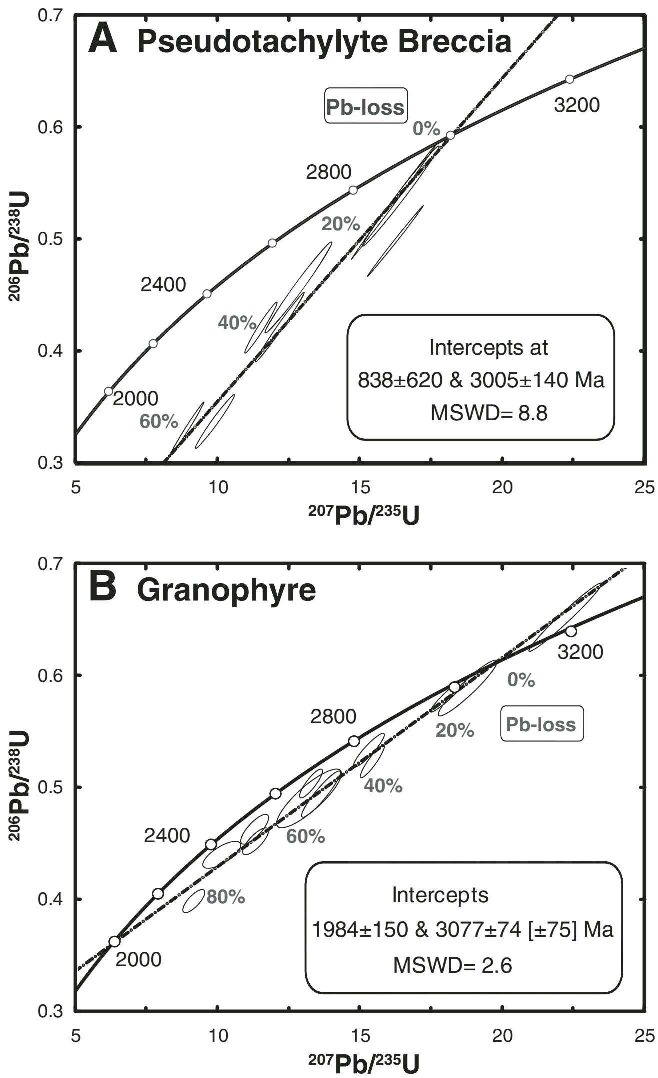

Figure 3. Concordia diagrams of zircon from the ca. 2 Ga Vredefort impact calibrated against AS3 zircon standard (Paces and Miller, 1993). (A) Grains analyzed in situ within the melt matrix of VD_PB show these grains are inherited from the Archean basement and possibly record thermal effects associated with emplacement of the ca. 1 Ga Anna's Rust Sheet. (B) Grains isolated from the granophyre unit, which exhibit poikilitic textures, also show inheritance from the Archean target, with varying amounts of Pb loss and a lower intercept presumably associated with the impact. MSWD—mean square of weighted deviations.

lunar meteorite SaU 169 (Liu et al., 2012). Zircon grains were imaged via backscattered-electron detector as unpolished, single loose grains (Fig. 1B), in polished thin sections (Figs. 1C and 1D), and in polished 2.54 cm epoxy mounts (Figs. 1E and 1F), and these were compared to images from polished thin sections of lunar meteorite SaU 169 (Figs. 1G and 1H; Liu et al., 2012). The presence of zircon with such a texture within the largest terrestrial impact crater deserved further investigation. Twelve of 20 isolated grains were analyzed; the other eight were so intergrown that no continuous surface was available to place the ion beam without possibility of common Pb contamination. One grain was rejected due to high common Pb, presumably contributed from contaminated areas or from the intergrown pyroxene. Results indicate that all the grains were inherited from the Archean target, as noted in the previous reports (Fig. 3; Wielicki et al., 2012a), with a crystallization age of 3077 ± 74 Ma, similar to that of VD_PB, and a lower intercept of 1984 ± 150 Ma (mean square of weighted deviates [MSWD] = 2.6). No grains appear to have grown or been "reset" within the impact melt. Interestingly, grains from a small hand sample (and thus in close proximity) showed dramatically varying amounts of Pb loss (0%–80%; Fig. 3).

DISCUSSION

Modeled crystallization temperatures appear to argue against most lunar zircons as forming in response to impact melting (Wielicki et al., 2012b; Grange et al., 2013). One possible complication of Ti crystallization temperatures for lunar zircons is the different behavior of elements in melts of variable redox states, i.e., presumably much more reduced in lunar as compared to terrestrial melts. However, partitioning coefficients of Ti between zircon and melt were found to be independent of fO_2 (Burnham and Berry, 2012), suggesting little presence of Ti^{3+} in even the most reduced melts and similar behavior in lunar and terrestrial melts. Ti-in-zircon crystallization temperatures of lunar poikilitic grains might provide another discrimination criterion with which to identify whether or not grains are melt neoformed or neoblastic. To our knowledge, Ti concentrations have yet to be reported for any poikilitic lunar grains.

Dissolution and growth of zircon in an impact melt are a function of: (1) the solubility of Zr, which itself is a function of composition (i.e., cation ratio M = [Na + K + 2Ca]/[Al·Si]; (2) the diffusivity of Zr; and (3) the temperature and rate of cooling (Harrison and Watson, 1983; Watson and Harrison, 1983; Watson, 1996). For lunar zircons incorporated within predominantly mafic lunar melts (SaU 169 M = 3.32; Liu et al., 2012), a high degree of resorption is likely given the propensity of high-M magmas (i.e., M = ~3.0) to dissolve zircon (Boehnke et al., 2013), potentially accounting for the poikilitic texture. For the composition of SaU 169, growth of zircon would require 20 times higher [Zr] (50,000 ppm; Boehnke et al., 2013) than that reported by Liu et al. (2012) of 2260 ppm at 1200 °C and over 10,000 ppm at an estimated solidus temperature of 1000 °C, essentially ruling out the possibility of this grain growing in equilibrium with the impact melt. Neoformed zircons of a size suitable for microanalysis are present in the largest differentiated impact melt sheets (e.g., Sudbury, Manicouagan, and Morokweng; $M \approx 1.5$), but they are rare in terrestrial impact melt–bearing breccias and undifferentiated impactites (Wielicki et al., 2012a).

Our observation of Vredefort zircon with inherited U-Pb systematics and apparent poikilitic textures further supports our conclusion that poikilitic grains from *Apollo 12* and lunar meteorite SaU 169 (Liu et al., 2012; Nemchin et al., 2008) are also likely inherited from target rocks and are not primary zircons grown in impact melts (Grange et al., 2013). Thus, such grains should be used with caution when investigating impact scenarios in terrestrial and extraterrestrial samples. Possible sources for inherited lunar zircons include granitoid and, Mg- or alkali suite–anorthositic clasts, such as those found within lunar meteorite SaU 169 (Liu et al., 2012). We note that the only occurrence of such textures in terrestrial zircon is from the Laramie anorthosite complex, Wyoming (Scoates and Chamberlain, 1995), which is compositionally similar to an important constituent of the lunar crust (Ryder, 1982), supporting the likelihood of inheritance. All grains analyzed in this study from the 2 Ga Vredefort impact were inherited from the Archean basement. Interestingly, the lower concordia intercept of 1984 ± 150 Ma obtained from poikilitic-textured grains from VD_G overlaps with the impact age; however, precise determination of the age of the impact event is not possible in this case. As few regional thermal events are seen within this time, the lower intercept may reflect thermal effects associated with the Vredefort impact as opposed to overprints from endogenic thermal pulses.

Poikilitic texture within zircon may be similar to those observed in the Laramie anorthosite, Wyoming (Scoates and Chamberlain, 1995), or may be related to sieve textures in plagioclase due to rapid decompression (Nelson and Montana, 1992), possibly from excavation of an impact crater, and/or from resorption (Stewart and Pearce, 2004). Interestingly, no such grains have been reported in the ancient detrital record, a time when large-scale impacts are predicted to have been common (see review in Harrison, 2009). The lack of such grains, as well as shocked zircon (Erickson et al., 2013), within the ancient detrital zircon record (Hopkins et al., 2010; Cavosie et al., 2010) strongly suggests that such grains do not survive sedimentary recycling.

CONCLUSION

Given fewer inherent complications than commonly used ^{40}Ar-^{39}Ar dating of lunar materials, U-Pb dating of zircon may become an important new tool with which to probe planetary impact history. However, a better understanding of zircon crystallization in response to impact shock and heating is required. Specifically, being able to discriminate inherited zircons grown in endogenic environments as opposed to impact melts is important for their use in establishing impact histories. Only in the case of small parent bodies without the thermal capacity for prolonged endogenic magmatism can zircon age and geochemistry currently be used to conclusively identify an impact. Our results suggest that zircon crystallization in response to impact melting is rare on the Moon, and zircons are unlikely to have crystallized from lunar impact melts but instead are inherited from the lunar crust. Thus, their use to constrain impact histories is limited. Poikilitic zircons from lunar meteorite SaU 169 are either xenocrysts inherited from the zircon-bearing anorthositic crust (Mg and/or alkali suite), or they record rapid decompression and/or resorption of the grain into a Zr-undersaturated magma, possibly associated with an impact event, but not requiring one.

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