# $^{40}Ar/^{39}Ar$ dating of Apollo 12 impact spherules

Jonathan Levine,[1,2] Timothy A. Becker,[3] Richard A. Muller,[1,4] and Paul R. Renne[3,5]

Received 1 March 2005; revised 8 June 2005; accepted 29 June 2005; published 2 August 2005.

[1] We have used the $^{40}Ar/^{39}Ar$ isochron technique to determine ages of 81 lunar spherules from Apollo 12 soil sample 12023. Most spherules are created in meteoroid impacts, and their ages correspond to the timing of the impacts that formed them. Of the 81 impacts we have dated, most occurred in the last 500 million years. The abundance of spherules from the most recent $\sim$10% of the history of the Moon is consistent with an increase in the meteoroid bombardment of the inner Solar System, but does not require this explanation. The soil sample from which we extracted our spherules was from the ejecta of a recent impact; our spherule age measurements support models of lunar soil mechanics and impact cratering in which ejecta are stratified and inverted relative to the stratigraphy that was present before the impact. **Citation:** Levine, J., T. A. Becker, R. A. Muller, and P. R. Renne (2005), $^{40}Ar/^{39}Ar$ dating of Apollo 12 impact spherules, *Geophys. Res. Lett.*, *32*, L15201, doi:10.1029/2005GL022874.

[2] Glass spherules are quenched droplets of melted or vaporized rock which coalesce while in free flight; on the Moon, these are created in volcanic fire-fountain events [*Reid et al.*, 1973] and, more commonly, in meteorite impacts [*Chao et al.*, 1970; *Glass*, 1971; *Delano and Livi*, 1981]. Radioisotopically determined ages of impact spherules from a well-mixed soil sample are a probe of the meteoroid bombardment history of the Moon [*Muller*, 1993]. The Earth and its satellite have experienced similar histories of bombardment, but evidence of ancient impacts is better preserved on the dry, airless, and tectonically inactive Moon [*Hörz*, 1985]. *Culler et al.* [2000] studied 179 spherules from 1 g of soil collected by the Apollo 14 astronauts, and found evidence for a decline in the meteoroid flux to the Moon from 3000 million years ago (Ma) to 500 Ma, followed by a fourfold increase in the cratering rate over the last 400 Ma. To test the extent to which the abundance of <400 Ma spherules observed by *Culler et al.* [2000] was affected by local geological processes [e.g., *Hörz*, 2000], we have repeated the experiment of *Culler et al.* [2000] using spherules from a soil sample collected from the Apollo 12 landing site.

[3] We received 5 g of Apollo 12 soil sample 12023 from the NASA Lunar Sample Curator. This sample was collected from a trench dug on the flank of Sharp Crater (diameter 14 m), judged on the basis of its fresh morphology to be in the youngest generation of craters visited by the Apollo 12 astronauts [*Shoemaker et al.*, 1970], with a probable age of a few million years (E. M. Jones, *Apollo 12 Lunar Science Journal*, http://www.hq.nasa.gov/office/pao/History/alsj/a12). In the work by *Culler et al.* [2000], the fact that the spherules were taken from the ejecta of a recent, bedrock-penetrating impact was used to argue that their ages would be representative of all impacts through the post-mare history of the Moon (i.e. since $\sim$3200 Ma). We hand-picked 179 spherules from soil fractions with grain size >180 μm, because the precision of $^{40}Ar/^{39}Ar$ dating of lunar spherules is limited by the potassium content of these specimens.

[4] We used geochemical criteria to test which, if any, of our spherules might have been formed in volcanic eruptions, rather than in impacts. Impact spherules have been distinguished on the basis of several characteristics, including low Mg/Al weight ratios, inter- and intra-spherule chemical heterogeneity, exotic inclusions, surface-correlated volatiles, and high ferromagnetic resonance intensity [e.g., *Delano and Livi*, 1981; *Stone et al.*, 1982]. Our diagnostic tests for spherule origin were performed without sectioning, polishing, or heating the spherules, so as not to compromise our ability to date each by the $^{40}Ar/^{39}Ar$ isochron technique. We examined each spherule in a field-emission scanning electron microscope at the National Center for Electron Microscopy, measuring major-element abundances by energy dispersive x-ray spectroscopy on multiple $\sim$10 μm spots. We used these spot analyses to estimate bulk Mg/Al weight ratios and to probe for heterogeneous concentrations of major elements (Al, Mg, Si, Ca, and Fe). Over 90% of the spherules were inconsistent with a volcanic origin on the basis of these two criteria (Supplement 1[1]).

[5] In preparation for $^{40}Ar/^{39}Ar$ dating, we sent the spherules, along with mineral standards Hb3gr [*Turner et al.*, 1971; *Renne*, 2000] and FCs [*Renne et al.*, 1998], for 207 hours of neutron irradiation at the McMaster Nuclear Reactor in Hamilton, Ontario (Supplement 2). Interfering reactions were minimized by encasing the spherules in cadmium foil to attenuate the thermal neutron flux; appropriate correction factors for these reactions were measured and applied.

[6] The irradiated spherules were degassed of their argon by laser step-heating to an extraction line connected to a MAP 215 noble gas mass spectrometer at the Berkeley Geochronology Center. We employed a $CO_2$ laser with power output 0.25–10 W. Spherules <250 μm in diameter were analyzed in 6–9 steps, and larger spherules were analyzed in 12–20 steps. Instrumental mass discrimination was monitored by regular analyses of air aliquots.

[1]Department of Physics, University of California, Berkeley, California, USA.

[2]Now at Laboratorio TANDAR, Departamento de Física, Comisión Nacional de Energía Atómica, Buenos Aires, Argentina.

[3]Berkeley Geochronology Center, Berkeley, California, USA.

[4]Lawrence Berkeley National Laboratory, Berkeley, California, USA.

[5]Department of Earth and Planetary Science, University of California, Berkeley, California, USA.


0094-8276/05/2005GL022874$05.00

[1]Auxiliary material is available at ftp://ftp.agu.org/apend/gl/2005GL022874.

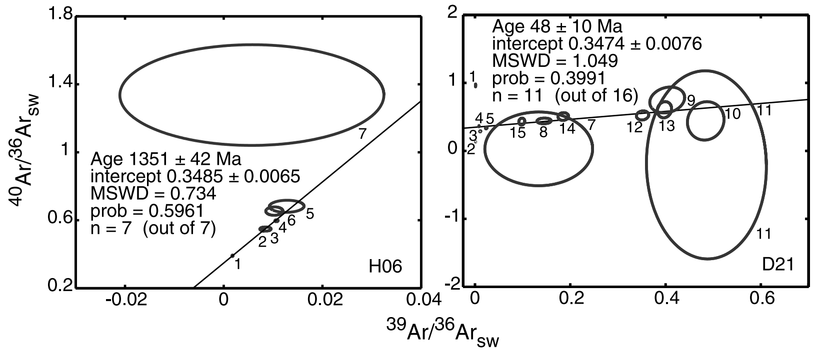


**Figure 1.** $^{40}$Ar/$^{39}$Ar isochrons, correlating ratios of $^{40}$Ar and $^{39}$Ar to solar-wind $^{36}$Ar, for 2 spherules. Ellipses contour the 1σ confidence level for each measurement. Ages are determined from the slope of the best fitting lines; intercepts correspond to the $^{40}$Ar/$^{36}$Ar ratio of surface-implanted (trapped) components. Quoted uncertainties represent 1σ. MSWD = mean square weighted deviance of the measurements from the line. Prob = probability that scatter from a straight line can be explained by experimental uncertainties alone. For spherule D21 (right), the first heating step has possible contamination from terrestrial atmosphere; the second and third show that the spherule may have lost argon on the Moon, and the 6th and 16th steps (not shown) released argon consistent with instrumental blank levels. All these steps are excluded from the isochron fit.

[7] We found statistically acceptable isochrons (i.e. partial releases along binary mixing lines in $^{39}$Ar/$^{36}$Ar$_{\text{solar-wind}}$ versus $^{40}$Ar/$^{36}$Ar$_{\text{solar-wind}}$ space) for 81 spherules (Figure 1, Supplement 3, Supplement 4). The fraction of spherules with acceptable isochrons is much lower than in the work by *Culler et al.* [2000], but this is partly due to a slight overestimate by *Culler et al.* [2000] of their analytical uncertainties. Re-analyzing the raw data of *Culler et al.* [2000], and we find acceptable isochrons for 61% of their Apollo 14 spherules (Supplement 5), compared with 45% of Apollo 12 spherules in this study [*Levine*, 2004]. An isochron results from the mixing of two isotopic components of argon: a radiogenic component, whose $^{40}$Ar/$^{39}$Ar ratio is a function of the age of the spherule, and a surface-implanted component containing solar-wind $^{36}$Ar and so-called parent-less $^{40}$Ar [*Wieler and Heber*, 2003]. (We assume that no cosmogenic $^{40}$Ar is present.) Spherules without statistically acceptable isochrons represent complex thermal and/or argon histories on the Moon, were incompletely degassed during formation, or contain argon from additional (and unresolvable) isotopic sources.

[8] The distribution of Apollo 12 spherule ages that we measure (Figure 2) is dominated by spherules younger than Ma, but is very different from the distribution of Apollo 14 spherules ages determined by *Culler et al.* [2000]. Our re-analysis of the data gathered by *Culler et al.* [2000] yields a spherule age distribution broadly similar to that reported initially, but in which the abundance of <400 Ma spherules is of less statistical significance [*Levine*, 2004] (Supplement 5). The overabundance of young spherules in the Apollo 14 sample could be explained by spherules produced in the Cone Crater impact at 25 Ma, by a veneer of post-Cone spherules deposited on top of a mixed layer of Cone ejecta, or by an increase in the meteoroid bombardment rate since 400 Ma. Our observation of abundant spherules younger than 500 Ma in Apollo 12 soil is consistent with a recent increase in the impactor flux in the inner Solar System [*Culler et al.*, 2000; *Grieve and Shoemaker*, 1994; *McEwen et al.*, 1997; *Schmitz et al.*, 1997], but does not require it. We consider alternative explanations of the spherule age distribution (Figure 2) below. The observation of many <500 Ma

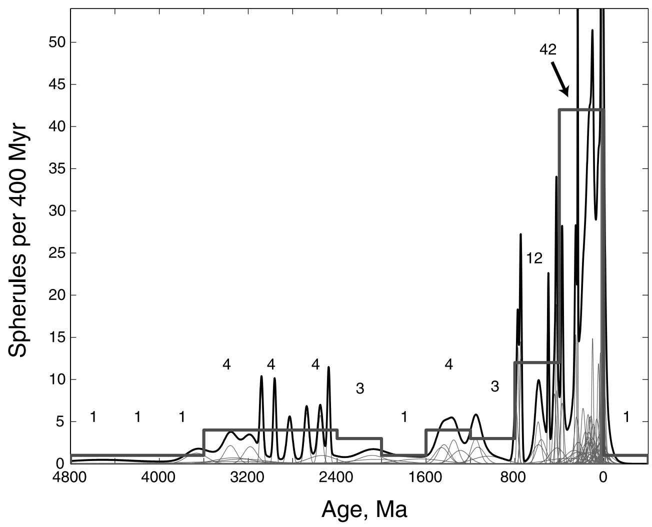


**Figure 2.** Age distribution of 81 Apollo 12 spherules. Each measured age is represented by a Gaussian of equal area (light curves), whose width corresponds to the uncertainty in the measurement, i.e. tall, narrow Gaussians represent precisely determined ages, and short, wide Gaussians denote imprecisely measured ages. An ideogram (dark curve) is created by summing the Gaussians; it is our best statistical estimate of the density of spherule ages in the Apollo 12 soil sample. The vertical scale for the Gaussians and ideogram is arbitrary. To blur the peaks caused by individual well-dated spherules, and to illustrate statistical significance more intuitively, we represent the same age distribution with a histogram, shown here with 400 million year bins. Numbers above each bin represent the number of spherule ages in each interval. Both the ideogram and the histogram show an abundance of spherules younger than 400 Ma, and a relative paucity of spherules older than 1000 Ma, which we attribute to the soil sample having come from the bottom of an ejecta deposit of recently inverted stratigraphy.

spherules cannot be explained by the spherules' being volcanic; there is no known volcanic activity on the Moon in this interval.

[9] First, the fact that we gathered spherules from Sharp Crater ejecta might have ensured that we dated many spherules created in the impact which excavated Sharp Crater. However, not more than $\sim$10 of our dated spherules have ages consistent with the likely age of Sharp Crater ($\leq$10 Ma). Thus the Sharp impact is not sufficient to explain the overabundance of young spherules that we observe.

[10] Second, perhaps spherules are destroyed or degassed of their argon with a mean lifetime on the Moon of $\sim$300 Ma. If a mechanism could be found that sets such a lifetime, we would expect spherules with older $^{40}$Ar/$^{39}$Ar isochron ages to be increasingly rare, approximately as we observe. However, the abundance of >3000 Ma spherules in the Apollo 14 sample of *Culler et al.* [2000] argues against the routine destruction of old spherules.

[11] Third, spherules older than a few hundred million years might have been removed from the Apollo 12 landing site by a large impact, with the subsequently developed regolith containing few older spherules. This model can be falsified on geological grounds: ejected rays from the Copernicus Crater, $\sim$400 km to the north, are continuous across the Apollo 12 landing site and date to $\sim$800 Ma [*Eberhardt et al.*, 1973], so no subsequent impact has ejected large amounts of regolith from this location. An event older than $\sim$800 Ma is insufficient to explain why there are so many more <400 Ma spherules than spherules from 400–800 Ma.

[12] Fourth, the Sharp Crater ejecta may include material only from the upper fraction of the pre-Sharp regolith. Sharp Crater is 3 m deep, and *Shoemaker et al.* [1970] consider it bedrock-penetrating or nearly so. If the full column of regolith from the crater were ejected, and if the ejecta were thoroughly mixed, the distribution of spherule ages would be representative of all lunar impacts since the emplacement of the local bedrock $\sim$3200 Ma. However, *Melosh* [1989] cautions that in computer simulations and laboratory experiments, only material from the top one-third of a crater is ejected, the remainder being displaced downward or laterally. If the pre-Sharp regolith were stratified, with younger spherules toward the top, then the distribution of ejected spherules should show a scarcity of older ages, consistent with our observations. However, for the top one-third of the regolith to be populated by spherules not more than 15% of the age of the local bedrock, one would also need to postulate a dramatic increase in the rate of soil accumulation at the Apollo 12 landing site since 400 Ma, as by an increased meteoroid flux.

[13] Observations of Barringer Meteor Crater, Arizona [*Shoemaker*, 1962] and laboratory impact experiments [*Melosh*, 1989] find overturned strata in the rim crest deposits of meteoroid impact craters, though the extent to which strata are mixed must vary with proximity to the crater and from impact to impact. *Melosh* [1989, p. 79] illustrates that even unlithified sand, in the laboratory, can be overturned by an impact as though it consisted of coherent layers. On the basis of a core sample collected on the Sharp Crater rim deposit, *Morris and Lauer* [1982] argue for overturned strata in the Sharp Crater ejecta, though this is disputed by *Nagle* [1980]. The overturning of strata may explain the distribution of spherule ages that we measure: trench sample 12023 was collected 20 cm deep into an ejecta deposit that is 23–28 cm thick [*Nagle*, 1980; *Morris and Lauer*, 1982], and therefore contains material that had been near the top of the regolith before the Sharp impact. In this model, our observation of many <400 Ma spherules implies the presence of abundant young spherules, and the near absence of old ones, in the uppermost layers of the pre-Sharp soil. Regolith could be stratified in this way if soils accumulate by slow deposition of ejecta from distant impacts, so that some spherules deposited in each unit have sufficient time at high temperature to be completely degassed. Most of the rest of the spherules may be only incompletely degassed, perhaps explaining why the majority of our spherules did not yield statistically acceptable isochrons. The few spherules we observe with ages >1000 Ma must have been heated very little when they were transported to the pre-Sharp regolith, so that their argon isotope systematics are undisturbed.

[14] Our explanation of stratified soil being overturned by impact offers two possible interpretations for the observations of *Culler et al.* [2000] at the Apollo 14 landing site. If the sample studied by *Culler et al.* [2000] was taken from the top layers of inverted Cone Crater ejecta, then it included the oldest pre-Cone material, along with young spherules created in the Cone impact or deposited subsequently. In this interpretation, the data do not require an increase in the meteoroid bombardment flux in the last 400 Ma. Alternatively, if the spherules dated by *Culler et al.* [2000] came from a distal part of the Cone ejecta blanket in which higher-velocity ejecta are more thoroughly mixed [*Melosh*, 1989], then the Apollo 14 spherule ages are representative of the post-mare bombardment history of the Moon, and the abundance of <400 Ma spherules implies an increased cratering rate since that time.

[15] Dating glass spherules from different depths in a cored regolith sample can probe the meteoroid bombardment history of the Moon if the studied soils accumulated through a large fraction of lunar history. We suggest dating spherules from either a drive tube core though much of the regolith or through the condensed stratigraphic section offered by the rim crest deposit of a recent impact. Such an impact must have penetrated at least 3 times the thickness of the regolith for the ejecta to include soils from all depths [*Melosh*, 1989]. Differences in the spherule age distribution as a function of depth would be useful constraints for models of regolith accumulation and impact mechanics. Material for this experiment could come from the Apollo archives or possibly from a future sample-return mission to the Moon.

[16] **Acknowledgments.** We are grateful to the NASA Lunar Sample Curator for providing the lunar samples, and to the staff of the National Center for Electron Microscopy. We thank W. Alvarez, F. Asaro, J. Feinberg, D. Karner, D. O'Connor, R. Rohde, D. Shimabukuro, and P. Weir for invaluable discussions and assistance in this research, and we acknowledge constructive reviews of our manuscript by B. Weiss and B. Cohen. Supported by the National Science Foundation, through a Graduate Research Fellowship (JL), the Ann and Gordon Getty Foundation, and the Folger Foundation.

## References

Chao, E. C. T., J. A. Boreman, J. A. Minkin, O. B. James, and G. A. Desborough (1970), Lunar glasses of impact origin: Physical and chemical characteristics and geologic implications, *J. Geophys. Res.*, *75*(35), 7445–7479.

Culler, T. S., T. A. Becker, R. A. Muller, and P. R. Renne (2000), Lunar impact history from $^{40}$Ar/$^{39}$Ar dating of glass spherules, *Science*, *287*, 1785–1788.

Delano, J. W., and K. Livi (1981), Lunar volcanic glasses and their constraints on mare petrogenesis, *Geochim. Cosmochim. Acta*, *45*, 2137–2149.

Eberhardt, P., J. Geiss, N. Grögler, and A. Stettler (1973), How old is the crater Copernicus?, *Moon*, *8*, 104–114.

Glass, B. P. (1971), Investigation of glass recovered from Apollo 12 sample 12057, *J. Geophys. Res.*, *76*(23), 5649–5657.

Grieve, R. A. F., and E. M. Shoemaker (1994), The record of past impacts on Earth, in *Hazards Due to Comets and Asteroids*, edited by T. Gehrels, pp. 417–462, Univ. of Ariz. Press, Tucson.

Hörz, F. (1985), Mass extinctions and cosmic collisions: A lunar test, in *Lunar Bases and Space Activities of the 21st Century*, edited by W. W. Mendell, pp. 349–358, Lunar and Planet. Inst., Houston, Tex.

Hörz, F. (2000), Time-variable cratering rates?, *Science*, *288*, 2095.

Levine, J. (2004), Lunar glass spherules as probes of the meteoroid impact history of the Moon, Ph.D. thesis, Univ. of Calif., Berkeley.

McEwen, A. S., J. M. Moore, and E. M. Shoemaker (1997), The Phanerozoic impact cratering rate: Evidence from the farside of the Moon, *J. Geophys. Res.*, *102*(E4), 9231–9242.

Melosh, H. J. (1989), *Impact Cratering: A Geologic Process*, Oxford Univ. Press, New York.

Morris, R. V., and H. V. Lauer (1982), Stratigraphy of core 12027 and section 64002 of core 64001/2: FeO and $I_s$/FeO depth profiles, *Lunar Planet. Sci.*, *13*, 544–545.

Muller, R. A. (1993), Cratering rates from lunar xenospherules, *Tech. Rep. LBL-34168*, Lawrence Berkeley Lab., Berkeley, Calif.

Nagle, J. S. (1980), Preliminary description and interpretation of drive tube 12027, *Lunar Planet. Sci.*, *11*, 786–788.

Reid, A. M., G. E. Lofgren, G. H. Heiken, R. W. Brown, and G. Moreland (1973), Apollo 17 orange glass, Apollo 15 green glass and Hawaiian lava fountain glass, *Eos Trans. AGU*, *54*(6), 606–607.

Renne, P. R., C. C. Swisher, A. L. Deino, D. B. Karner, T. L. Owens, and D. J. DePaolo (1998), Intercalibration of standards, absolute ages and uncertainties in $^{40}$Ar/$^{39}$Ar dating, *Chem. Geol.*, *145*, 117–152.

Renne, P. R. (2000), $^{40}$Ar/$^{39}$Ar age of plagioclase from Acapulco meteorite and the problem of systematic errors in cosmochronology, *Earth Planet. Sci. Lett.*, *175*, 13–26.

Schmitz, B., B. Peucker-Ehrenbrink, M. Lindström, and M. Tassinari (1997), Accretion rates of meteorites and cosmic dust in the early Ordovician, *Science*, *278*, 88–90.

Shoemaker, E. M. (1962), Interpretation of lunar craters, in *Physics and Astronomy of the Moon*, edited by Z. Kopal, pp. 283–259, Elsevier, New York.

Shoemaker, E. M., et al. (1970), Preliminary geologic investigation of the Apollo 12 landing site, part A, in *Apollo 12 Preliminary Science Report*, *NASA SP-235*, 113–156.

Stone, C. D., L. A. Taylor, D. S. McKay, and R. V. Morris (1982), Ferromagnetic resonance intensity: A rapid method for determining lunar glass bead origin, *Proc. Lunar Planet. Sci. Conf. 13th*, Part 1, *J. Geophys. Res.*, *87*, suppl., A182–A196.

Turner, G., J. C. Huneke, F. A. Podosek, and G. J. Wasserburg (1971), $^{40}$Ar-$^{39}$Ar ages and cosmic ray exposure ages of Apollo 14 samples, *Earth Planet. Sci. Lett.*, *12*, 19–35.

Wieler, R., and V. S. Heber (2003), Noble gas isotopes on the Moon, *Space Sci. Rev.*, *106*, 197–210.

T. A. Becker and P. R. Renne, Berkeley Geochronology Center, 2455 Ridge Road, Berkeley, CA 94709, USA. (prenne@bgc.org)

J. Levine, Laboratorio TANDAR, Departamento de Física, Comisión Nacional de Energía Atómica, Avenida del Libertador 8250, 1429 Buenos Aires, Argentina.

R. A. Muller, Department of Physics, University of California, Berkeley, CA 94720, USA.