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The crystalline lunar spherules: Their formation and implications for the origin of meteoritic chondrules

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Abstract–Crystalline lunar spherules (CLS) from three thin sections of Apollo 14 regolith breccias (14318,6; 14318,48 and 14315,20) have been examined. The objects have been classified and their abundances, size distributions, bulk compositions, and (where possible) plagioclase compositions determined. By number, 64% consist predominantly of very fine-grained equant plagioclase grains but can also contain larger (~50 μm) feldspar crystals (type X), while 22% contain plagioclase lathes in a fine-grained mafic mesostasis (type Y). Plagioclase in both spherule types displays bright yellow cathodoluminescence that is conspicuous among the blue CL of the normal feldspar of the breccias. Type Z spherules (5%) contain feldspar with blue CL and minor amounts of olivine and pyroxene. Type Q spherules (4%) contain feldspar with yellow CL but in a luminescent mesostasis (of quartz or feldspar?). A few spherules are mixtures of type Y and type X textures. Most type X spherules, and a few type Y spherules, have fine-grained opaque rims. Compound objects were also found and consist of two or more CLS that appear to have collided while still plastic or molten. The CLS are thought to be impact spherules that crystallized in free flight, their coarse textures suggesting fairly slow cooling rates (~<1 °C/s). The abundance of the CLS resembles that of chondrules in the CM chondrite Murchison, and their cumulative size-frequency distributions are very similar to those of the chondrules in several meteorite classes. The bulk compositions of the CLS do not resemble regoliths at any of the Apollo sites, including Apollo 14, or any of the common impact glasses, but they do resemble the bulk compositions of several lunar meteorites and the impact glasses they contain. The Apollo 14 site is located on a region containing Imbrium ejecta, and we suggest that the CLS derive from the Imbrium impact. Ballistic calculations indicate that only impact events of this size on the Moon are capable of producing melt spherules with the required free flight times and slow cooling rates. Smaller impacts produce glassy spherules and agglutinates. As has been pointed out many times, the CLS have many properties in common with meteoritic chondrules. While much remains unclear, difficulties with a nebular origin and new developments in chondrule chronology, studies of asteroid surfaces and impact ejecta behavior, and the present observations indicate that meteoritic chondrules could have formed by impact.

INTRODUCTION

The Apollo 14 mission was the first to be sent to a lunar highlands site, the Fra Mauro formation. The site was chosen partly because the formation was thought to contain ejecta from the Imbrium impact (Swann *et al.*, 1977). The returned samples include a variety of complex breccias characterized by a wide range of clast types and textures. The presence of polymict fragmental breccias, regolith breccias, impact-melt breccias, and clast-poor impact melts indicates complex histories of impact melting, fragmentation, shock lithification, and recrystallization. Lithic fragments, glass and mineral fragments, pure glassy spherules, and crystalline spherules are all common components of these samples (*e.g.*, Chao *et al.*, 1972; Warner, 1972). Several Apollo 14 breccias are also notable for an abundance of small spherules that are totally crystalline, although these objects have also been found in samples from other Apollo sites and lunar meteorites (Fredriksson *et al.*, 1970; Roedder, 1971; Kurat *et al.*, 1972, 1974; Nelen *et al.*, 1972; King *et al.*, 1972a,b; Bunch *et al.*, 1972; Keil *et al.*, 1972; Dence and Plant, 1972; Winzer *et al.*, 1977; Roedder and Weiblen, 1977; Bischoff *et al.*, 1987; Koeberl *et al.*, 1991; Warren *et al.*, 1990; Neal *et al.* 1991; Holder and Ryder, 1995). These objects are submillimeter crystalline beads that are spherical or elliptical in shape and contain silicate assemblages that show a variety of morphologies set in a microcrystalline mesostasis. They are normally referred to as "lunar chondrules" or "chondrule-like" objects in the literature. We previously and incorrectly referred to them as "devitrified glass spherules" (Sears *et al.*, 1996a) but now prefer the purely descriptive term "crystalline lunar spherules," or CLS.

The Apollo 14 breccias 14301, 14313 and 14318 contain up to 10 vol% CLS, while the breccias 14305, 14306 and 14311 contain CLS in "moderate" abundances (King *et al.*, 1972b). This is in contrast to the situation at most Apollo sites, where CLS are rare but glassy spherules and agglutinates are common. Many types of glassy spherules have been found in lunar samples, and most are thought to be either impact-produced or volcanic in origin. A set of criteria developed to distinguish the two types have been reviewed by Delano (1986). All of the authors that have discussed the origin of CLS have concluded that they formed by impact melting of lunar surface materials and crystallization during free-flight.

Studies of meteoritic chondrule formation have not yet resulted in a widely accepted theory for their origin (King, 1983; Boss, 1996). Nevertheless, chondrules are an important constituent of some of the oldest solar system materials and must provide important clues to the formation conditions of the major chondrite classes. For several years, the CLS were widely regarded as strong evidence that meteoritic chondrules were also of an impact origin (King *et al.*, 1972a; Fredriksson *et al.*, 1970; Sanders, 1994), but the idea fell into disfavor with many researchers (Taylor *et al.*, 1983; Wood, 1985; Grossman, 1988), although some still argued for other unidentified planetary mechanisms (Hutchison *et al.*, 1988). One key argument against an impact origin for meteoritic chondrules is that "chondrules" are not abundant in lunar samples. Identification of crystalline objects with extensive similarities to meteoritic chondrules found on the heavily impacted lunar surface is therefore extremely important. However, whether or not they are "true" chondrules, the CLS are sufficiently similar to meteoritic chondrules that such comparisons should provide new insights into the origin and history of both. In the present paper, we summarize literature observations of CLS and report new data for the cathodoluminescence (CL), optical and physical properties, and the bulk and phase compositions of CLS in three thin sections of two Apollo 14 breccias 14318 and 14315.

EXPERIMENTAL

The three polished thin sections, 14318,6, 14318,48, and 14315, 20, were examined. Cathodoluminescence images were produced using an MAAS Luminoscope attached to a standard petrographic microscope with a 15 ± 1 keV, 0.7 ± 0.1 mA electron beam focused to a ~1 × 2 cm ellipse. The luminescence was recorded photographically at a magnification of 50× and the film developed using the commercial C-40 process. Photomosaics were assembled then from the prints and each crystalline spherule mapped. The sizes of the CLS were measured in thin section using transmitted light while CL images were used to help locate and classify the objects. Sizes of each object were obtained by averaging the maximum dimension and the dimension perpendicular to this measured using a calibrated reticle in the eyepiece of the microscope.

The major and minor element compositions of the crystalline spherules located in the sections were determined using the Cameca Camebax electron microprobe at the NASA Johnson Space Center. Plagioclase and mesostasis phases were analyzed using a focused (~2 μm diameter) beam in point mode, 15 kV accelerating voltage, and 30 nA absorbed current. Data from 4–6 analyses per spherule were averaged. The bulk compositions of the CLS were determined by rastering a focused beam over a 30 μm square and integrating all counts from the resulting x-rays. Depending on spherule size, between four and ten areas could be analyzed in this way and the data averaged to give an estimate of the spherule's bulk composition. In addition to analyzing a region more representative of the bulk material, rastering the beam also reduces volatile-loss and the current density to the sample (Yang *et al.*, 1992). Mineral standards include kaersutite (Na, K, Si, Ti, Al, Fe, Mg, Ca), 479A (Cr), NVGR (Mn) and apatite (P), and all microprobe data were reduced by the PAP matrix correction program (Pochou and Pichoir, 1991). On-peak count times for phase and bulk analyses were 20 s for Na, K, Si, Ca, P; 25 s for Ti, Al, Cr, Mg; and 40 s for Fe and Mn. Repeated analyses of mineral standards indicate reproducibility for all elements to within 5%.

Correction factors have been applied to the bulk chemical data that attempt to account for the effects of different host-phase densities during rastered beam microprobe analysis (Warren, pers. comm.). As discussed below, the CLS are predominantly plagioclase and, thus, the correction factor for each element is usually comparable to or smaller than the natural variation among the objects.

RESULTS

Classification of the Spherules Based on Observed Properties

We located 303 CLS in the ~10 cm^2 covered by the three thin sections (Fig. 1). Four types of spherules were distinguished on the basis of texture and CL properties, although only two were numerous. Remarkably, few CLS could not be assigned to one of these classes, and assignment was simple to make because (1) the types are highly distinctive and (2) within a type, the CLS were very uniform in their properties. As a simple descriptive device, this classification scheme is very successful and it seems highly likely that this scheme carries genetic implications. This is discussed below. Representative transmitted light photomicrographs of the CLS are shown in Fig. 2 and CL images are shown in Fig. 3. The spherules are circular to elliptical in thin section and all consist of plagioclase crystals in a cryptocrystalline mesostasis of usually mafic silicates. None of them would be considered glassy spherules of the sort discussed by Delano (1986), or the devitrified glasses of Lofgren (1971), who described plagioclase and pyroxene spherulites, generally <1 μm in diameter, which nucleated on the outer edge and grew radially inward, often into a core of glass. In contrast, all of the CLS are completely crystalline, the plagioclase is never spherulitic, the crystallization appears to have occurred randomly, and there is no indication of the plagioclase nucleating perpendicular to the surface. These properties are inconsistent with textures derived from solid-state recrystallization. The CLS textures are clearly igneous.

A significant property of the CLS is the frequent presence of a fine-grained rim. These rims are very similar to rims found on meteoritic chondrules in that they are usually very thin (<~10 μm) and are texturally quite different from the object they surround. The composition of the rim material has not yet been determined, although like many meteorite chondrule rims they appear to contain abundant fine-grained opaque minerals.

Type X CLS display a wide diversity of plagioclase crystal sizes and morphologies (Fig. 2a). These objects are predominantly composed of very fine-grained (typically ~20 μm) equant plagioclase grains imparting an overall granular texture, although they often contain a few larger plagioclase crystals with highly irregular boundaries. The fine-grained feldspar displays a weak yellow-green CL, while the larger feldspar grains are bright yellow (Fig. 3a). The interstitial material does not luminesce. Type X CLS often possess a full or partial thin (5–10 μm) opaque rim of undetermined composition.

Type Y objects consist of euhedral plagioclase lathes embedded in a relatively mafic, cryptocrystalline mesostasis (Fig. 2b). The lathes are usually very thin but have sharp edges. These objects are highly conspicuous on the CL mosaics because the feldspar displays bright yellow CL, the mesostasis shows comparatively little CL, and they are highly uniform in appearance (Fig. 3b). Type Y objects sometimes have thin, opaque rims, but more often they are not rimmed and have irregular edges.

Mixed texture objects are those in which a significant part of the CLS had a typical type X texture while another part had a type Y texture. We found 13 objects of this sort.

Type Z objects are composed of large feldspar grains (10–40 μm) in a feldspathic matrix and often contain small (~5 μm) olivine and/or pyroxene grains and opaques (Fig. 2c). The larger feldspar crystals often have a bright blue CL and the matrix is slightly less intense blue, while the olivine/pyroxene and opaques are nonluminescent (Fig. 3c). These objects are almost never rimmed. Their textures and CL properties somewhat resemble those of the lithic clasts in the breccia.

Type Q spherules consist of laths of plagioclase with the normal yellow CL in a mesostasis that, unlike the mesostases in types X and Y, luminesces, either blue or various shades of yellow or yellow-green. We did not examine these spherules in detail but suspect that the mesostases are either quartz (blue CL) or feldspar (yellow/yellow-green CL). We found seven of them.

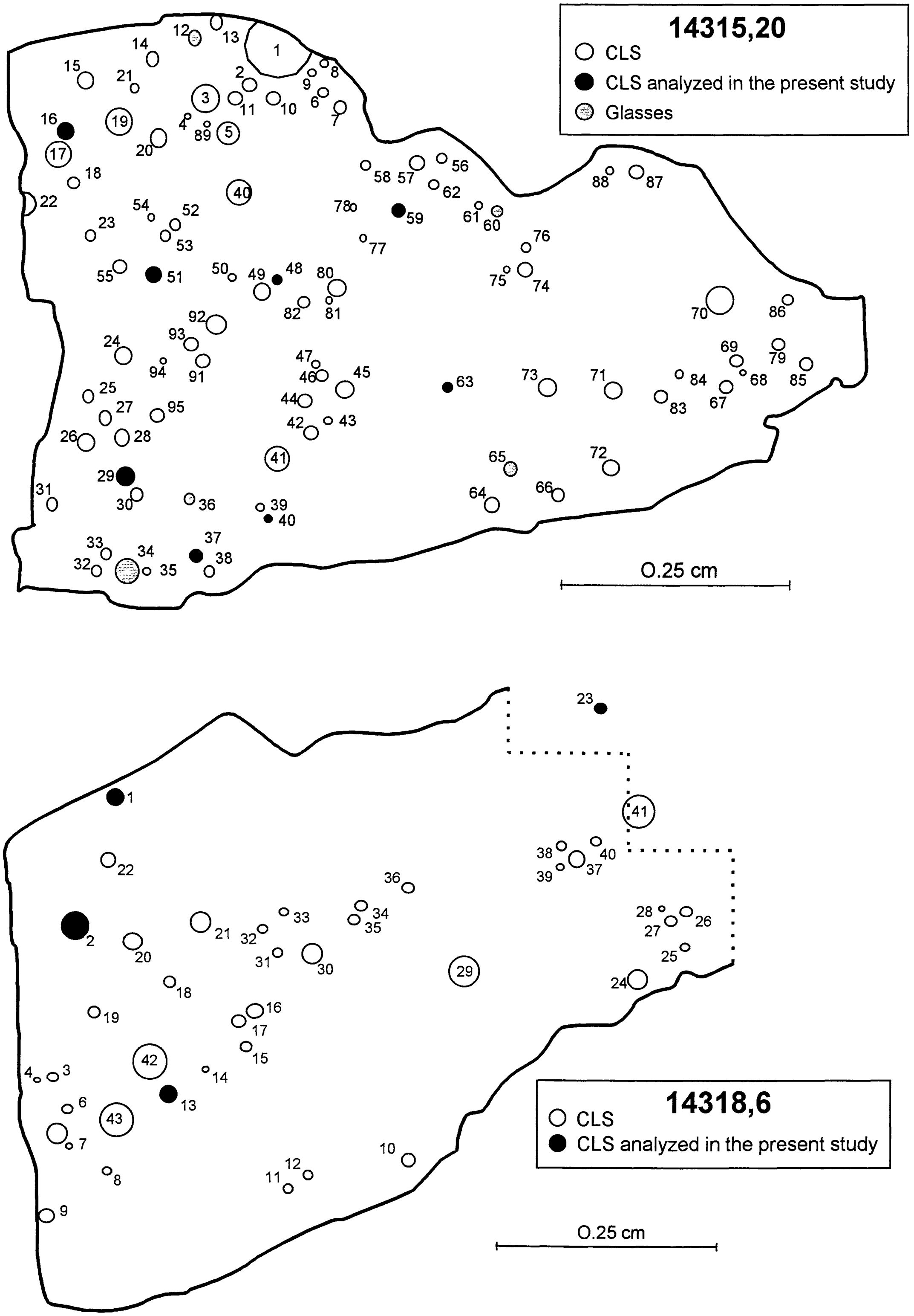

FIG. 1. Maps of two of the three sections used in the present study indicating the location of crystalline lunar spherules (CLS) and glass spherules. The third, much larger section is not shown because of its size. The distribution of CLS is not uniform but rather they are present in comminuted matrix regions of the breccias and not the large coherent clasts.

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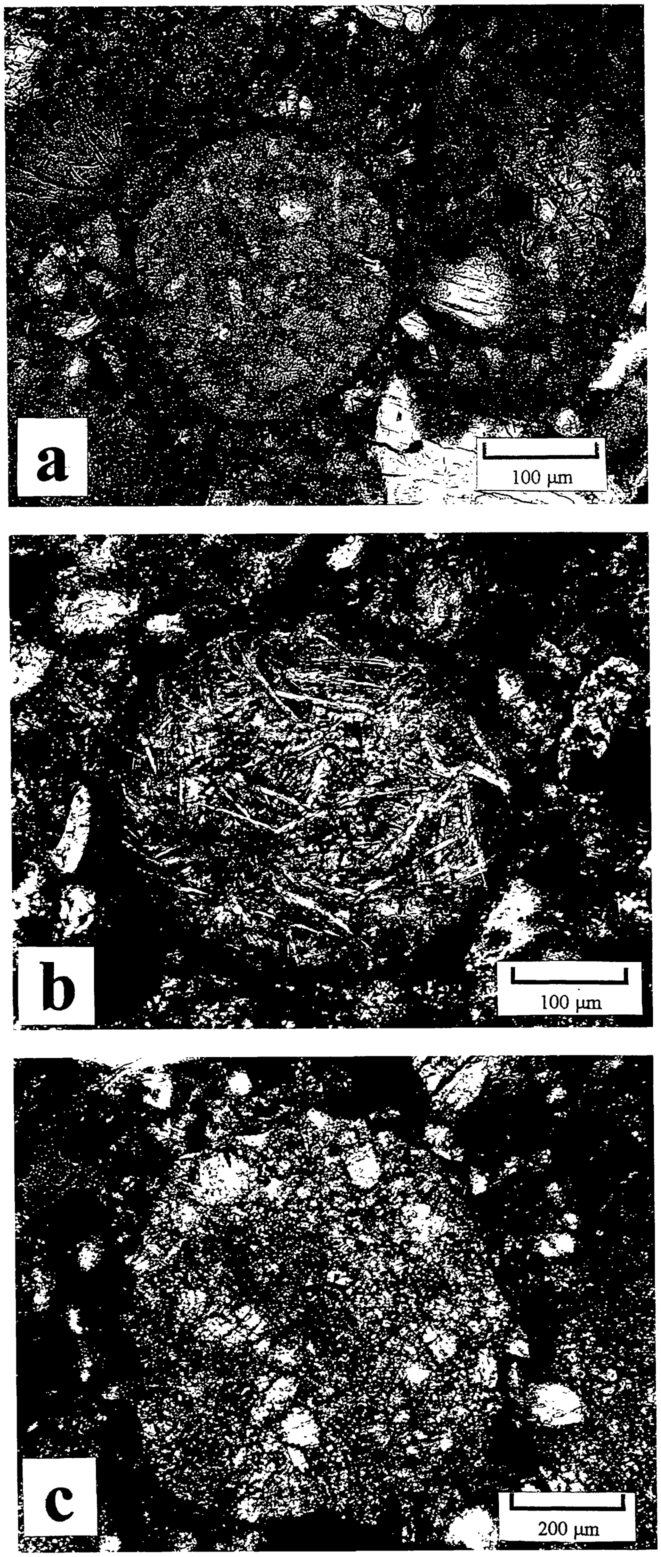

FIG. 2. Transmitted light photomicrographs of representative CLS in Apollo 14 breccias. (a) A 250 μm diameter type X CLS from 14315,20 (#48;); (b) A 325 μm diameter type Y CLS from 14318,6 (#13;); (c) A 670 μm diameter type Z CLS from 14318,48 (#14).

Compound objects are those in which one spherule appears to engulf another or where two or more spherules abut each other. We found several such objects and one is shown in Fig. 3d.

Abundances, Sizes and Shapes

The three sections we examined contain ~8 vol% of CLS, a value similar to the 5–10 vol% estimate reported by Nelen *et al.* (1972) for the same breccias. Our size data are given in Table 1 and the distribution of CLS between the three sections, and their average sizes and aspect ratios (ratios of maximum diameter to the perpendicular diameter), are given in Table 2. Of the 303 CLS we found, 64% (by number) are type X, 22% are type Y, 5% are type Z, 4% are type Q and 4% have mixed textures.

Our size data are plotted in Figs. 4 and 5. The diameters for type X objects ranged from 40 μm to just over 800 μm, with a grand average of 195 ± 93 μm. Type Y objects have a similar range of diameters but their mean is 221 ± 96 μm, so that type Y objects are all ~10% larger than type X objects (but significant only at the 95% level).

The CLS show a marked tendency to sphericity, with an average aspect ratio of 1.28 for type X and 1.35 for type Y, although type X and type Y CLS can show large deviations from circularity with ratios approaching 3.0 sometimes observed (Table 2 and Fig. 6).

Chemical Compositions

The bulk compositions of type X and type Y CLS are shown in Table 3 and Fig. 7. Literature data for basaltic CLS (called type Z in the present study) overlap with impact glasses, while both literature data for CLS and the present type X and type Y do not overlap with pristine (*i.e.*, volcanic) or impact lunar glasses. Table 4 compares CLS bulk compositions with those of "average" Apollo 14 soil where it can be seen that large differences exist among all major elements. The CLS are very feldspathic and there is no evidence for a compositional difference between type Y and type X.

The plagioclase lathes of type Y CLS are An_{95-90}, while the mesostases are mafic (12–20 wt% FeO and 5–21 wt% MgO, Tables 5 and 6). Figure 8 compares plagioclase compositions of type Y CLS with data on similar lunar objects from Kurat *et al.* (1974). Feldspar compositions in type Y objects agree with previous data and suggest derivation from a source region that is rich in anorthositic material. Type X CLS contain phases that are too fine-grained for analysis, but their bulk compositions are indistinguishable from type Y (Table 3) and clearly derive from a similar region.

DISCUSSION

A Brief Survey of Crystalline Lunar Spherules in the Literature

Literature observations of CLS are listed in Table 7. "Lithic," "droplet," "porphyritic" and "non-porphyritic" types have all been described. Some authors describe the presence of rims on the CLS. To a good approximation, type X objects correspond to the "lithic chondrules," and type Y objects correspond to the "droplet chondrules" of King *et al.* (1972a). Type Z objects probably correspond to the "basaltic chondrules" of Kurat *et al.* (1972) but could also be small abraded clasts. Type Q spherules do not appear to have been previously described.

The researchers listed in the references to Table 7 show remarkably universal agreement in likening these objects to meteoritic chondrules, either referring to them simply as "chondrules" or, more conservatively, "chondrule-like." In fact, the main thesis of many of the papers cited in Table 7 is the textural similarity of the CLS to meteoritic chondrules, despite major differences in composition.

A significant property of the CLS described in the literature is their composition. The largest compilation of CLS bulk compositions in the literature is that of Prinz *et al.* (1973) who report data for 18 "anorthositic-noritic-troctolitic (ANT) chondrules," five "alkali high-alumina basalt chondrules," one "high-alumina quartz basalt chondrule" and one "basaltic chondrule," these names reflecting compositional similarities between the CLS and their rock namesakes. The present types X and Y are very similar to each other and closely resemble those of the "ANT chondrules" (loosely termed FAN chondrules) of Prinz *et al.* (1973) and Kurat *et al.* (1974) (Figs. 7 and 8). In short, most CLS have compositions similar to those of highland rocks and remarkably few are basaltic in composition even though most lunar samples were recovered from basaltic regoliths. This is commonly interpreted as indicating that either highland compositions predispose the melts to more easily crystallize or that only the lunar highlands received impacts large enough to produce these objects. We return to this point below.

Histories: Textures and Cooling Rates

Most observers have concluded that the CLS are melt spherules that crystallized during free flight. The present data are also consistent with this conclusion. Figure 9a is a plot of the partition coefficients

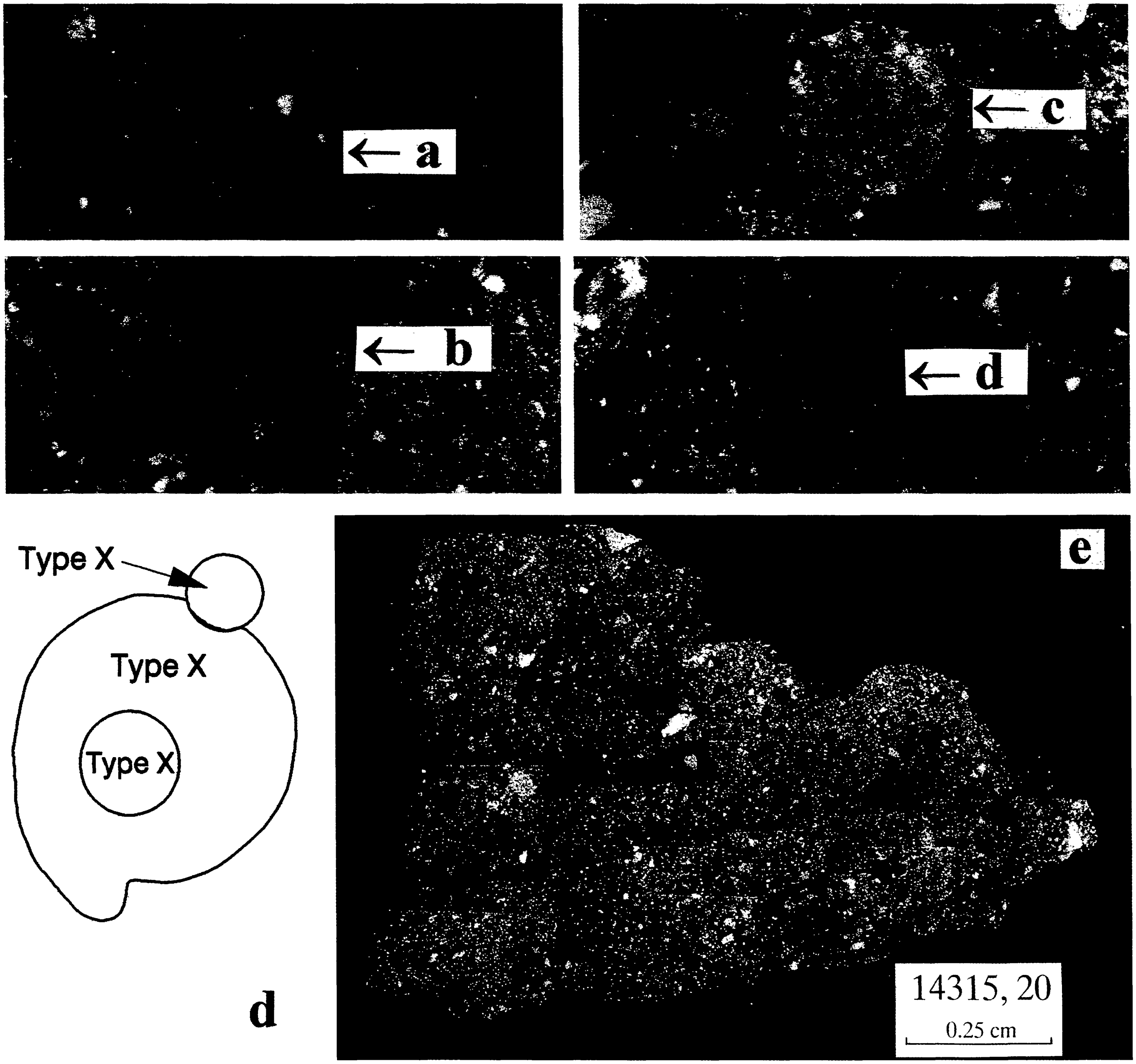

FIG. 3. Cathodoluminescence images of the objects shown in Fig. 2. (a) A type X CLS from 14315,20 (#48); (b) A type Y CLS from 14318,6 (#13); (c) A type Z CLS from 14318,48 (#14); (d) a compound object in which a type X CLS encloses a type X object; (e) a cathodoluminescence mosaic of thin section 14318,6. The brecciated texture of the sample is clearly evident from the cathodoluminescence image. Virtually all yellow objects are crystalline lunar spherules.

for the plagioclase grains and the coexisting mesostasis. While the plagioclase elements (Ca, Al and Na) are obviously enriched in the grains, and Si is equally distributed between phases, most of the remaining elements are enriched in the mesostasis with plagioclase/mesostasis partition coefficients of ~0.05 (they range from ~0.02 to 0.2). This partitioning behavior suggests that the plagioclase and the mesostasis crystallized from the same melt. Figure 9b shows Weill and McKay's (1975) experimentally derived partition coefficients for melts of lunar highland composition. Values very similar to those obtained for the present CLS are observed, which suggests equilibrium crystallization close to the experimental range of 1320–1200 °C.

Assuming that the CLS are impact spherules that crystallized in free flight after ejection by impact, then following Arndt *et al.* (1984) we might expect them to have cooled in a vacuum at a rate that can be calculated from the Stefan-Boltzmann equation:

$$\frac{dT}{dt} = \frac{6\epsilon\sigma}{D\rho C_p}(T^4 - T_s^4)$$

where T, D, ρ, and C_p are the temperature, diameter, density and specific heat of the droplet, T_s is the ambient temperature (taken as 200 K, but values up to 1400 K do not significantly alter the result), ϵ is total emissivity, and σ is the Stefan-Boltzmann constant (5.67×10^{-12} W cm^{-2} K^{-4}). For a 0.02 cm droplet with T = 1800 K, ϵ = 0.7 ("average" value used by Arndt *et al.*, 1984), ρ = 2.74 g cm^{-3} (corresponding to An_{90}), and C_p = 0.3267 cal g^{-1} K^{-1} (the specific heat

TABLE 1. Diameters (μm) of crystalline lunar spherules from three thin sections of two Apollo 14 breccias*.

No.	Dia.	No.	Dia.	No.	Dia.	No.	Dia.	No.	Dia.	No.	Dia.	No.	Dia.	No.	Dia.
Type X		69	250	6	140	81	140	153	380	26	220	173	140	**Compound**	
14315,20		71	110	7	215	82	300	156	275	27	220	174	110	**14318,6**	
1	815	74	240	8	185	85	165	157	125	29	450	178	120	21	265
2	200	75	170	10	300	90	255	159	220			181	435		
6	200	76	95	11	140	91	110	160	230	**Type Y**		184	340	**Type Q**	
7	140	77	140	12	125	92	115	164	180	**14318,48**		186	250	**14318,48**	
8	130	78	40	15	95	93	75	165	190	9	230	***Avg Y***	***221***	25	275
9	150	79	75	17	175	96	135	167	280	24	215	***Std Dev***	***96***	35	310
11	130	80	400	18	120	97	230	168	250	45	235			36	305
12	170	82	235	19	135	99	200	170	305	50	440	**Type Z**		66	785
13	170	84	90	20	225	100	370	176	170	51	400	**14315,20**		68	435
14	300	85	140	21	145	102	180	179	125	52	350	19	400	117	400
15	185	86	180	22	190	104	105	180	250	56	395	49	300	119	390
16	200	87	190	23	160	106	205	182	220	57	105	81	50	***Avg Q***	***337***
17	530	88	170	26	255	109	305	183	155	65	155	92	255	***Std Dev***	***164***
18	210	90	380	28	225	110	230	185	310	67	115	94	50		
20	250	91	165	29	190	111	180	***Avg X***	***195***	69	235			**Mixed**	
21	130	93	155	31	70	112	135	***Std Dev***	***93***	77	255	**Type Z**		**14318,48**	
23	150	95	140	32	215	114	175			78	220	**14318,6**		58	210
25	155			33	80	118	150	**Type Y**		83	100	16	230	71	450
26	380	**Type X**		34	230	122	150	**143515,20**		86	110	17	215	73	85
29	335	**14318,6**		37	215	123	310	79	115	89	105	19	220	84	225
31	145	4	105	39	175	125	105	5	375	94	130	31	145	105	390
32	110	5	525	40	200	126	130	10	230	95	140	39	95	124	350
33	180	6	205	41	235	127	195	22	200	98	95			135	155
35	100	8	215	42	210	128	325	30	195	101	180	**Type Z**		141	160
39	150	11	155	44	175	129	205	37	205	103	255	**14318,48**		145	225
40	120	12	180	46	195	130	245	43	170	107	300	14	670	161	325
41	380	14	110	49	135	131	170	53	120	108	175	16	160	163	145
42	225	15	90	53	325	132	115	55	190	113	120	38	270	177	235
44	220	18	195	54	205	133	165	63	160	115	165	88	160	***Avg***	***246***
46	185	20	185	55	175	134	130	70	415	120	250	116	125	***Std Dev***	***110***
48	250	22	180	59	195	136	200	72	210	121	160	175	415		
51	200	37	195	60	155	137	270	83	255	140	155	***Avg Z***	***235***		
52	140	40	120	61	60	138	195	89	155	148	200	***Std Dev***	***158***		
54	90	41	485	62	105	139	100			149	235				
56	205			63	100	142	190	**Type Y**		154	175	**Type Q**			
57	170	**Type X**		64	140	143	150	**14318,6**		155	225	**14315,20**			
58	130	**14318,48**		70	315	144	340	1	235	158	145	28	380		
59	255	1	230	72	310	146	250	2	320	162	230	38	150		
61	150	2	260	74	255	147	250	3	200	166	95	45	265		
62	145	3	115	75	70	150	245	13	325	169	225	50	150		
64	165	4	245	76	140	151	125	23	195	171	85	66	185		
67	190	5	250	80	120	152	275	24	360	172	205	73	350		

*As described in the text, type X refers to crystalline lunar spherules (CLS) containing equant plagioclase grains, type Y have lathes of plagioclase, type Z are essentially basaltic in composition, and type Q have plagioclase grains in a mesostasis of undetermined compositions but known to be unlike the others. Mixed refers to single objects with multiple textures.

of anorthite at 1800 K; Robie *et al.*, 1979), the estimated cooling rate is ~3300 °C/s. This is essentially an instant quench and pure glass beads should have resulted. Clearly, the textures of both type X and type Y CLS, especially type Y, suggest much slower cooling rates than this calculation would suggest. Experiments on synthetic melts with compositions similar to Apollo 15 green glass indicate that cooling rates of <1 °C/s are required for crystallization (Arndt *et al.*, 1984), while synthetic glasses of Apollo 17 orange and black glass composition required <100 °C/s for crystallization (Arndt and Engelhardt, 1987). Similar experiments with melts of chondritic composition and cooling rates <1 °C/s also produce textures similar to those observed in the CLS (Lofgren and Russell, 1986; Hewins, 1988; Lofgren, 1989), as do melts of feldspathic composition (Lofgren, 1977). While composition, maximum temperature, amount of supercooling, and the presence of nucleation sites all affect crystallization, it seems safe to conclude that the CLS cooled at rates of <100 °C/s, probably <1 °C/s, which is much slower than cooling in a vacuum. Like the volcanic lunar spherules (Arndt and Engelhardt, 1987; Arndt *et al.*, 1984; Kring and McKay, 1984) and meteoritic chondrules (Grossman, 1988; Hewins, 1988, 1989), the CLS must have been surrounded by hot gases and dust during their crystallization. Also, the CLS appear to have solidified before reimpacting the lunar surface as evidenced by their well-preserved spherical shapes. If CLS cooled at rates of 1 °C/s, this would imply solidification times of ~5 min to cool from, say 1300 to 1000 °C. This, in turn, requires long flight times and distant source regions.

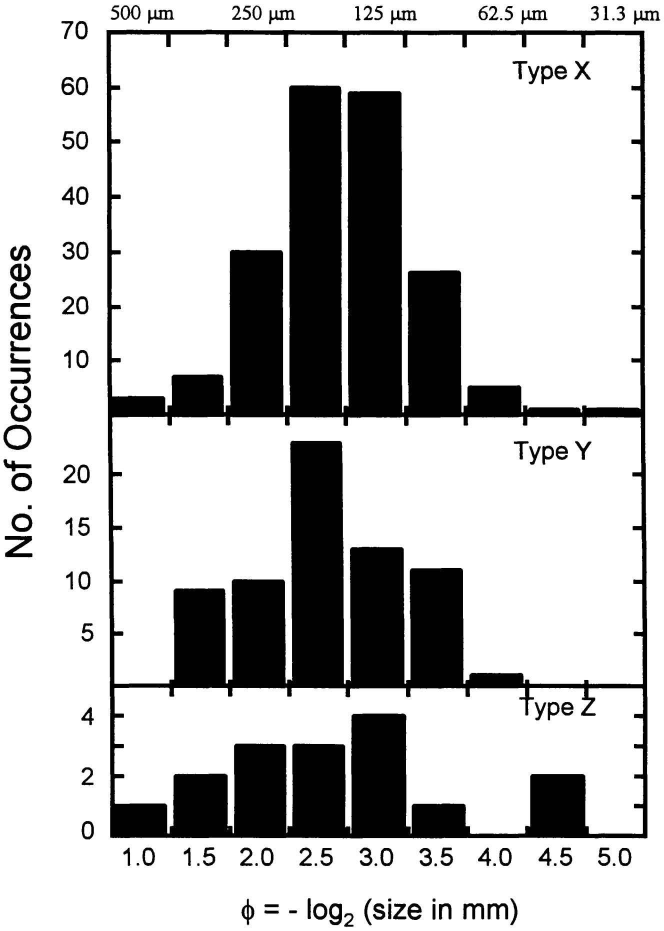

FIG. 4. Histograms of maximum diameters for crystalline lunar spherules in Apollo 14 breccias 14315 and 14318. The type Y CLS size distribution appears to be skewed to slightly larger sizes by ~10%. Type Z (basaltic) CLS appear to have similar dimensions to the other types.

TABLE 2. Distribution, size, and shape data for crystalline lunar spherules from three Apollo 14 breccia thin sections.

CLS type	Number of objects 14315,20	14318,6	14318,48	Total	Mean size (μm)†	Aspect ratio†‡
X	61	22	111	194	195 ± 93	1.28 ± 0.36
Y	14	9	44	67	221 ± 96	1.35 ± 0.27
Z	5	5	6	16	235 ± 158	1.28 ± 0.28
Q	6	0	7	13	337 ± 164	1.30 ± 0.24
Mixed*	0	0	13	13	246 ± 110	1.75 ± 0.61

*Mixed = single object with multiple textures.
†Uncertainties are 1σ.
‡Ratio of largest to smallest diameter.

Compositions and Source Regions

Delano (1986) and others have argued that most impact spherules have bulk compositions similar to the regolith from which they formed but that ballistic transport can sometimes result in the presence of exotic material. Of the 360 impact glasses in soil 64001, for instance, 23% were thought to be exotic to the site and had been transported at least 300 km (Delano, 1991). Delano (1991) also reported that ~40% of the impact spherules in MAC 88105 and <5% in ALH 81005 were exotic and not the product of impact melting of local

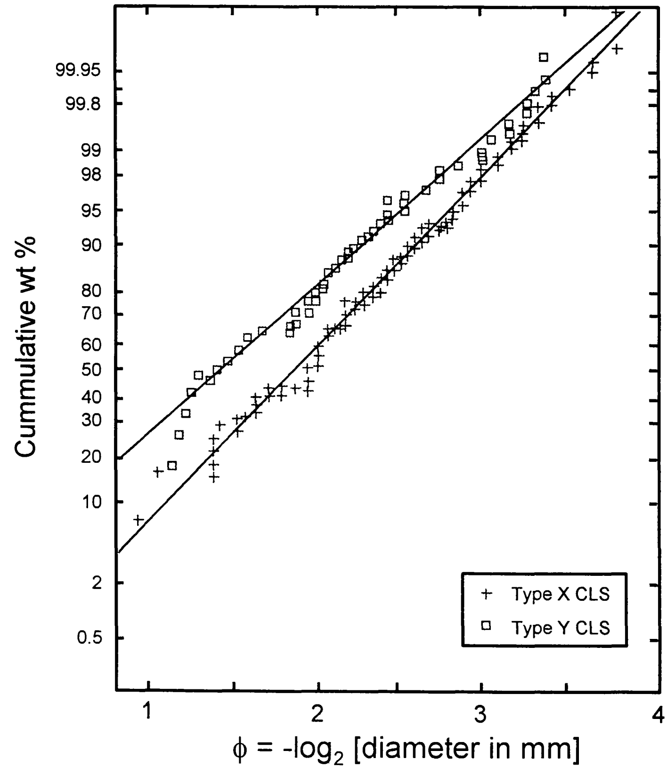

FIG. 5. Cumulative size-frequency distributions for crystalline lunar spherules. $\phi = -\log_2$ (diameter in mm) is a commonly used parameter for describing grain-size distributions in sediments. Consistent with the histograms, the cumulative size-frequency distributions indicate that type Y are larger than type X.

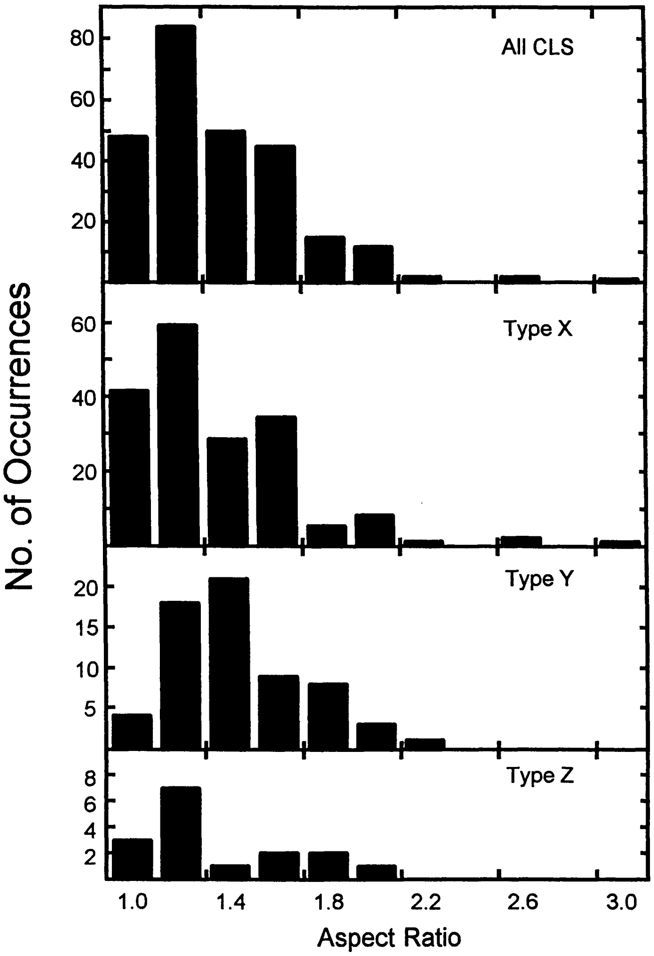

FIG. 6. The aspect ratios (ratios of the largest to the smallest diameters) for crystalline lunar spherules from Apollo 14 breccias 14315 and 14318. Most of the objects are spherical, but some highly elliptical objects are also observed.

regolith. Figure 10 is a plot proposed by Delano that uses diagnostic combinations of nonvolatile elements to identify source materials for impact spherules. Most strikingly, the CLS from the Apollo 14 regolith breccias do not plot with the Apollo 14 highland regolith, or the highland regolith of any of the Apollo sites, but instead have bulk compositions similar to the bulk compositions of lunar highland meteorites (Y-791197, ALH 81005, Y-82192, Y-86032, MAC 88105) and the impact spherules they contain.

A possibility to account for the compositions of Apollo 16 high alumina-silica poor (HASP) glasses is the process of differential vaporization during an impact melting event (Naney *et al.*, 1976). Compared to the inferred parent composition, the HASP glasses were depleted in Si and Na but enriched in Ti, Al, and Ca. However, relative to "average" Apollo 14 regolith, the CLS have near-normal Si and Na, are depleted in Ti, Fe, Mg, and P, and have high Ca and Al (Table 4) so that CLS compositions could not be produced by melting Apollo 14 soil. Furthermore, the only phenocrysts present in the CLS are plagioclase, which would be expected from a high-Al parent melt. These trends suggest that the CLS derive from a source region more feldspathic than Apollo 14 regolith. Differential vaporization probably did not play a role in the production of the observed CLS compositions.

We conclude that the CLS are exotic to the Apollo 14 site. One possibility is that they were transported to the site by the Imbrium impact since much of the material in the Fra Mauro region is thought to contain Imbrium ejecta. The question then becomes whether the Imbrium impact would have resulted in flight times long enough to permit the very slow cooling rates required by the CLS.

Origin of the Crystalline Lunar Spherules

According to Melosh (1989), the ballistic range, R_b, of ejecta is given by:

$$R_b = 2R_m \tan^{-1}\left(\frac{(v_e^2/gR_m)\sin\theta\cos\theta}{1-(v_e^2/gR_m)\cos^2\theta}\right)$$

where v_e is the initial ejection velocity, R_m is the radius of the Moon, g is the acceleration due to gravity, and θ is the angle of ejection (arbitrarily assumed to be 45°). The ejection velocity, v_e, depends on r/R, where r is the distance of the ejecta from the center of a crater of radius of R (Melosh, 1989, Fig. 6.4 caption):

$$v_e = 0.28\ (r/R)^{-1.8}\ (g D_t)^{0.5}$$

where D_t is the transient crater diameter. We restrict our use of this relationship to r/R values between ~0.35–0.9 because the equation is only valid for ejecta deriving from a distance from the crater center that is larger than a few times the projectile radius (which we take to be $r/R = 0.3$) and because crater formation moves from the strength regime to the gravity regime at $r/R \cong 1$ when a different ejecta velocity equation would be required (Melosh, 1989; Housen *et al.*, 1983).

The transient crater diameter is used in the ejecta velocity expression since the Imbrium Basin is a complex, multi-ringed structure that experienced significant slumping and crater modification long after the initial kinetic energy of the impactor had been transferred to the early leaving, high-speed ejecta. Melosh (1989) suggests that for complex craters, the transient crater diameter is 0.5 to 0.65× the final crater diameter, and since the rim-to-rim diameter of the Imbrium Basin is 1200 km (Wilhelms, 1984), we can take the transient crater as being 600 km in diameter.

Crucial to the formation of the CLS is the time of flight. The time of flight (T_a) for ejecta from a crater being formed on a body where crater radius is a significant fraction of the parent body radius, so that the curvature is important, was given by Ahrens and O'Keefe (1978):

$$T_a = \frac{a^{3/2}}{\sqrt{gR_m^2}}\left\{\pi - \left[2\tan^{-1}\left(\sqrt{\frac{1-e}{1+e}}\tan\left(\frac{\pi}{2}-\frac{R_b}{4R_m}\right)\right) - \frac{e\sqrt{(1-e^2)}\sin(\pi - R_b/2R_m)}{(1+e\cos(\pi - R_b/2R_m))}\right]\right\}$$

when $v_e^2/gR_m > 1$, or,

$$T_a = \frac{2a^{3/2}}{\sqrt{gR_m^2}}\left[2\tan^{-1}\left(\sqrt{\frac{1-e}{1+e}}\tan\left(\frac{R_b}{4R_m}\right)\right) - \left(\frac{e\sqrt{(1-e^2)}\sin(R_b/2R_m)}{(1+e\cos(R_b/2R_m))}\right)\right]$$

when $v_e^2/gR_m \le 1$. The ellipticity of the trajectory, e, is given by:

$$e^2 = \left(\frac{v_e^2}{gR_m} - 1\right)^2 \cos^2\theta + \sin^2\theta$$

TABLE 3. Raw data for the bulk compositions (wt% oxide) determined by rastered-beam electron microprobe analysis of randomly selected type X and type Y crystalline lunar spherules*.

PTS/Object	SiO_2	TiO_2	Al_2O_3	Cr_2O_3	FeO	MnO	MgO	CaO	Na_2O	K_2O	P_2O_5	Total
Type X												
14315,20/48	45.71	0.23	26.17	0.13	6.14	0.09	4.78	14.24	0.49	0.20	0.10	98.30
14315,20/40	44.50	0.27	25.80	0.24	6.45	0.09	5.07	14.04	0.55	0.15	0.08	97.22
14315,20/51	45.59	0.29	24.40	0.14	6.74	0.10	5.53	13.53	0.53	0.30	0.07	97.22
14315,20/59	43.86	0.24	27.27	0.08	5.03	0.08	4.44	14.78	0.54	0.16	0.04	96.51
14315,20/98	45.30	0.29	24.05	0.22	7.01	0.11	5.45	13.45	0.63	0.38	0.13	97.03
14315,20/16	44.38	0.19	24.42	0.11	8.08	0.11	6.62	13.12	0.54	0.32	0.12	98.00
14315,20/29	44.59	0.23	26.91	0.28	5.33	0.09	5.04	14.54	0.53	0.12	0.09	97.76
Mean	*44.85*	*0.25*	*25.58*	*0.17*	*6.40*	*0.09*	*5.28*	*13.96*	*0.54*	*0.23*	*0.09*	*97.43*
Std. dev.	*0.69*	*0.04*	*1.30*	*0.08*	*1.03*	*0.01*	*0.70*	*0.61*	*0.04*	*0.10*	*0.03*	*0.62*
Rel. dev.(%)	*2*	*16*	*5*	*47*	*16*	*11*	*13*	*4*	*7*	*43*	*33*	
Type Y												
14318,6/23	46.41	0.20	28.55	0.07	2.71	0.05	2.18	15.40	1.02	0.45	0.27	97.31
14318,6/1	45.26	0.24	25.10	0.08	6.46	0.09	4.93	14.04	0.57	0.30	0.07	97.14
14318,6/13	44.55	0.17	26.97	0.12	5.13	0.07	4.82	14.75	0.52	0.10	0.06	97.25
14318,6/2	45.76	0.17	23.56	0.21	6.51	0.10	7.07	13.10	0.49	0.08	0.06	97.10
14315,20/63	45.33	0.28	24.03	0.14	7.29	0.10	6.39	12.86	0.72	0.25	0.12	97.51
14315,20/37	45.70	0.34	24.05	0.21	6.96	0.10	6.04	13.24	0.59	0.22	0.13	97.58
14315,20/99	45.50	0.15	23.68	0.07	7.21	0.09	6.99	12.26	0.98	0.16	0.10	97.19
Mean	*45.50*	*0.22*	*25.13*	*0.13*	*6.04*	*0.08*	*5.49*	*13.66*	*0.70*	*0.22*	*0.11*	*97.30*
Std. dev.	*0.57*	*0.07*	*1.92*	*0.06*	*1.64*	*0.02*	*1.71*	*1.11*	*0.22*	*0.13*	*0.07*	*0.19*
Rel. dev.(%)	*1*	*32*	*8*	*46*	*27*	*25*	*31*	*8*	*31*	*59*	*64*	

*Each value is an average of typically five analyses. See Symes (1996) for a complete data listing. Std. dev. refers to standard deviation for all the objects and Rel. dev. is the percent relative deviation among the objects of each type.

and a is the semimajor axis of the trajectory:

$$a = \frac{R_m[1 - e\cos(R_b / 2R_m)]}{(1-e)(1+e)}$$

Figure 11 shows the distance from the basin center that ejecta lands (R_b + r) as a function of original position of ejected material (r/R) for the Imbrium impact. Alongside each point is the calculated time of flight. Ejecta coming from the outer parts of the crater (r/R > 0.5) experience only a few minutes of flight, whereas a small fraction of material from the central regions of the crater (r/R < 0.5) can travel half way around the Moon and take hours to settle. The Fra Mauro region is ~1200 km from the center of the Imbrium basin and lies near the edges of the expected zone of continuous ejecta, which extends ~1100 km from the center of the basin (Oberbeck *et al.,* 1974). Material from r/R = 0.45 has an appropriate ballistic range for landing in this region. It takes ~30 min to travel that distance.

The often perfectly circular shapes of the CLS indicate that they were completely solidified in-flight so that deformations caused by being plastic or molten at the time of impact with the surface occurred occasionally but were not common. Estimates of the liquidus and solidus temperatures can then constrain first-order cooling rates because of the limited time-of-flight. Due to the similarity in composition between the CLS and synthetic charges used by

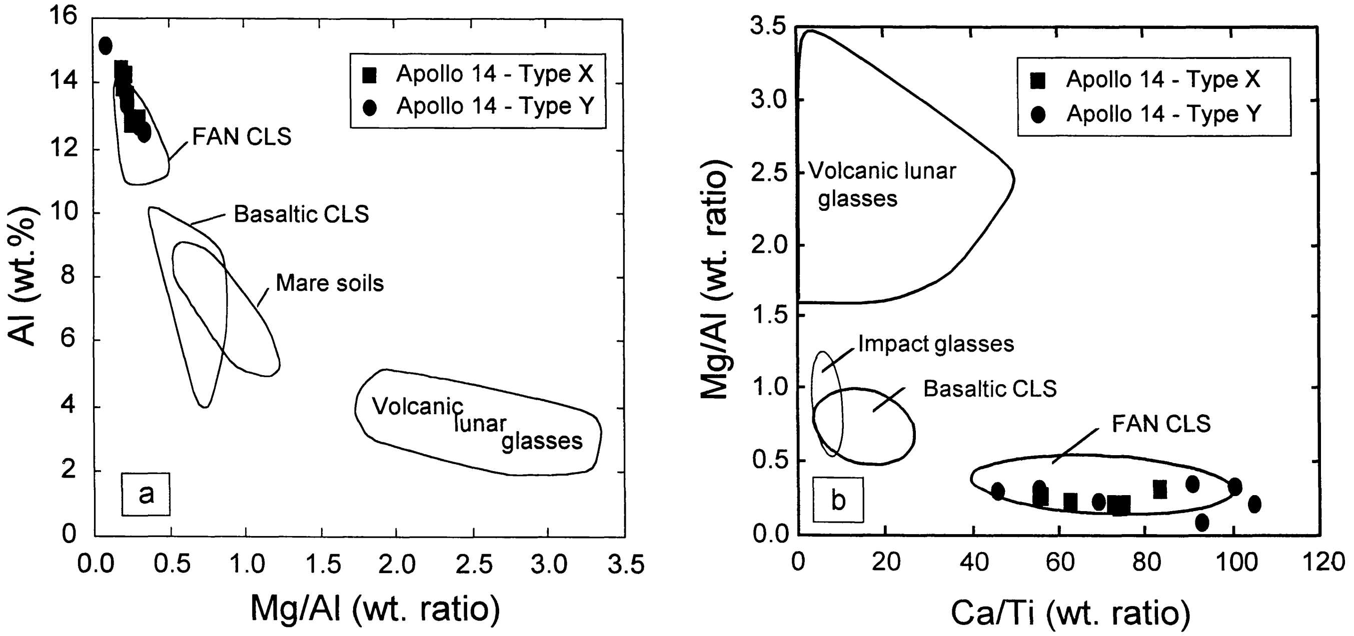

FIG. 7. Comparisons of CLS bulk compositions (raw data) obtained in the present study and literature data for CLS with the compositions of volcanic lunar glasses, impact glasses, and mare soils. Literature CLS data from Prinz *et al.* (1973) (for "ANT" chondrules which for our purposes are equivalent to "FAN") and the remaining literature data from Delano (1986). (a) Al (wt%) *vs.* Mg/Al. (b) Mg/Al *vs.* Ca/Ti (element ratios are all weight ratios). Literature data for basaltic CLS (called type Z in the present study) overlap with impact glasses, while both literature and the present type X and type Y CLS data do not overlap with pristine or impact lunar glasses. They have lunar highland compositions and there is no evidence for a compositional difference between type Y and type X CLS.

Lofgren (1977), a reasonable estimate for the liquidus temperature of the CLS would be ~1300 °C. If complete solidification is assumed to occur at ~1000 °C, then estimated cooling rates are <1 °C/s, which is entirely consistent with the observed textures. Of course, the spherules may have solidified well before landing. A cooling rate of 1 °C/s would imply solidification after only 15–20% of the ballistic path length.

It is possible that the crystallization of the CLS resulted when spherules with quenched, hard rims (but mostly molten interiors) landed in the regolith and were then insulated by warm ejecta to allow slow cooling. It seems unlikely, however, that 250 μm objects could have a temperature gradient so large as to have a molten interior with a solidified rim. Certainly no such gradient would exist so long as to still be present at the end of the ballistically required 30 min flight.

If the type Y spherules are equivalent to droplet chondrules and type X spherules were equivalent to lithic chondrules, as suggested by King *et al.* (1972a), then very different modes of origin are implied. However, we doubt this is the case because we observed a few CLS that were type X for one half of the texture and type Y for the other, yet they were clearly a single entity that was at one time entirely melted. We suspect that the different textures are the result of different nucleation processes, but we stress the distinctiveness of the two textures and the lack of transitionary textures. Above, we alluded to the superficial similarity between type Z CLS and the larger lithic clasts and suggested that they might be small abraded clasts, in which case the type Z CLS are more equivalent to lithic chondrules.

It takes major impacts on the Moon to produce CLS. Smaller impacts would be expected to produce melt-spherules that cool too

TABLE 4. Average bulk compositions (wt%) for Type X and Type Y objects compared to Apollo 14 average soils.

Oxide	Uncorrected Type X	Uncorrected Type Y	Corrected Type X	Corrected Type Y	Bulk soil 14163,778*	"Average" A14 Soil†
SiO_2	44.85	45.50	45.74	46.41	47.3	48.1
TiO_2	0.25	0.22	0.32	0.29	1.6	1.7
Al_2O_3	25.58	25.13	24.04	23.63	17.8	17.4
Cr_2O_3	0.17	0.13	0.20	0.15	0.200	0.23
FeO	6.40	6.04	8.44	7.97	10.5	10.4
MnO	0.09	0.08	0.11	0.10	0.135	0.14
MgO	5.28	5.49	6.17	6.42	9.6	9.4
CaO	13.96	13.66	13.96	13.66	11.4	10.7
Na_2O	0.54	0.70	0.51	0.66	0.70	0.70
K_2O	0.23	0.22	0.21	0.20	0.55	0.55
P_2O_5	0.09	0.11	0.10	0.12	n.a.	0.51
Total	97.43	97.30	99.82	99.62	99.8	99.8

*From soil "reference suite" Papike *et al.* (1982); n.a. = not analyzed.
†Average composition of soils 14003, 14148, 14149, 14156 (McKay *et al.*, 1991).

TABLE 5. Compositions of plagioclase lathes in type Y CLS (wt% oxide).*

	14318,6				14315,20			
Object	1	2	13	23	37	63	*Mean*	*Std. dev.*
SiO_2	44.37	46.47	45.19	46.39	44.2	45.47	*45.36*	*0.99*
TiO_2	0.03	0.02	0.03	0.07	0.04	0.09	*0.05*	*0.03*
Al_2O_3	35.52	33.72	34.47	33.39	34.25	33.82	*34.19*	*0.75*
Cr_2O_3	0.02	0.03	0.02	0.01	0.01	0.01	*0.02*	*0.01*
FeO	0.76	0.59	0.78	0.79	0.69	0.57	*0.70*	*0.10*
MnO	0.02	0.01	0.01	0.01	0.02	0.01	*0.01*	*0.01*
MgO	0.48	0.58	0.63	0.38	0.28	0.26	*0.44*	*0.16*
CaO	18.57	18.18	18.45	17.66	18.36	17.83	*18.17*	*0.36*
Na_2O	0.46	0.72	0.58	0.78	0.56	0.71	*0.64*	*0.12*
K_2O	0.05	0.04	0.04	0.38	0.05	0.09	*0.11*	*0.14*
P_2O_5	0.03	0.02	0.02	0.04	0.04	0.05	*0.03*	*0.01*
Total	*100.31*	*100.38*	*100.22*	*99.9*	*98.5*	*98.91*	*99.71*	*0.81*
No. of ions on the basis of 8 oxygens								
Si	2.047	2.134	2.085	2.145	2.064	2.118		
Al	1.931	1.825	1.874	1.816	1.914	1.857		
Σ	*3.98*	*3.96*	*3.96*	*3.96*	*3.98*	*3.97*		
Ca	0.918	0.894	0.912	0.873	0.920	0.890		
Na	0.041	0.064	0.052	0.070	0.051	0.064		
K	0.003	0.002	0.002	0.022	0.003	0.005		
Σ	*0.96*	*0.96*	*0.96*	*0.97*	*0.97*	*0.96*		
Norm (mol%)								
An	95.39	93.12	94.4	90.39	94.38	92.79		
Ab	4.33	6.64	5.37	7.28	5.27	6.67		
Or	0.28	0.24	0.23	2.33	0.35	0.54		

*Each value for the wt% oxide is an average based typically on five analyses. See Symes (1996) for a complete listing of data. Mean and Std. dev. refer to the grand mean and standard deviation for all analyzed lathes.

TABLE 6. Compositions of mesostasis regions in type Y CLS (wt% oxide)*.

PTS/Object	SiO_2	TiO_2	Al_2O_3	Cr_2O_3	FeO	MnO	MgO	CaO	Na_2O	K_2O	P_2O_5	Total
14318,6/1	51.02	0.66	17.17	0.24	12.85	0.19	5.18	12.69	0.39	0.54	0.14	101.1
14318,6/2	49.22	0.34	11.30	0.40	12.12	0.19	13.95	11.82	0.22	0.07	0.06	99.7
14318,6/13	47.29	0.38	15.21	0.31	12.45	0.18	12.29	9.90	0.28	0.12	0.12	98.5
14318,6/23	44.74	0.65	2.48	0.27	22.34	0.31	21.29	6.16	0.08	0.14	0.10	98.5
14315,20/37	50.78	0.65	6.68	0.47	14.86	0.24	17.85	6.05	0.26	0.26	0.14	98.3
14315,20/63	48.67	0.41	9.34	0.22	15.83	0.26	15.25	7.88	0.42	0.22	0.15	98.6
14315,20/99	48.79	0.40	8.42	0.12	19.98	0.28	14.97	6.66	0.39	0.07	0.22	100.3
Mean	*48.64*	*0.50*	*10.09*	*0.29*	*15.77*	*0.24*	*14.40*	*8.74*	*0.29*	*0.20*	*0.13*	*99.3*
Std. dev.	*2.15*	*0.15*	*5.01*	*0.12*	*3.97*	*0.05*	*5.00*	*2.75*	*0.12*	*0.17*	*0.05*	*1.1*

Norm (mol%)	Or	Ab	An	Di	Hy	Ol	Il	Qt
14318,6/1	2	3	33	14	25	0	2	21
14318,6/2	0	2	25	24	40	8	1	0
14318,6/13	1	2	35	7	44	10	1	0
14318,6/23	1	1	4	17	28	48	2	0
14315, 20/37	1	2	14	11	69	1	2	0
14315, 20/63	1	3	19	13	49	14	0	0
14315,20/99	0	3	17	9	54	15	1	0

*Each value is an average based typically on five analyses. See Symes (1996) for a complete listing of data. Std. dev. refers to standard deviation for all the objects.

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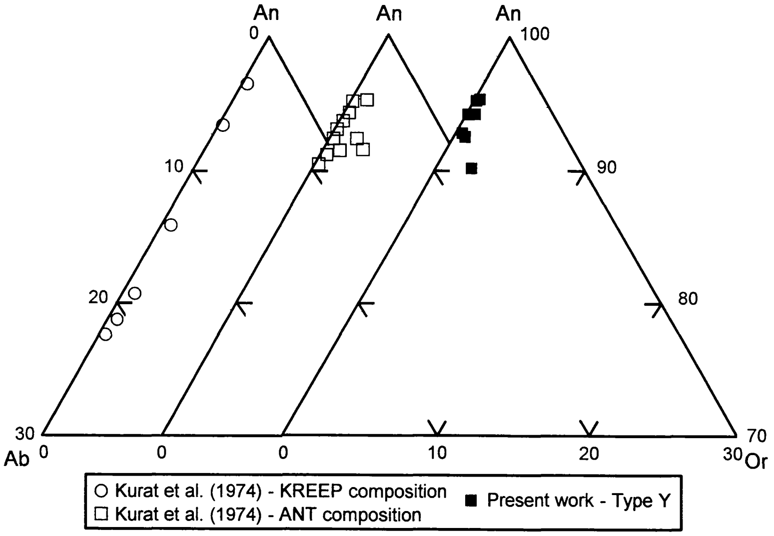

FIG. 8. Composition of feldspar in type Y CLS from Apollo 14 breccia thin sections 14318,6 and 14315,20 compared with literature data. The present data agree with literature data and suggest derivation from the anorthosite-rich highland crust.

quickly for appreciable crystallization because they do not produce a mass of hot gas and dust to insulate the spherules in free flight. Still smaller impacts will result in very short flight times and produce agglutinates because the melt globules impact the surface before solidification. Figure 12 shows calculations similar to those described above (Fig. 11) for four relatively small (1, 2, 5, and 10 km) craters on the Moon. These small (numerous) cratering events produce ejecta with orders of magnitude shorter flight times, and any melt droplets formed would probably cool radiatively since no transient atmosphere would be produced to slow down cooling. These factors would increase the proportion of glassy spherules and agglutinates in the ejecta of these small craters. Kring and McKay (1984) suggest that agglutinates were essentially quenched at <600 °C/min, so that compositional gradients in olivine were either vestiges of the original igenous gradients or were the result of subsequent slow cooling period at ~10 °C/min.

Implications of Crystalline Lunar Spherules for the Formation of Chondrules on Asteroids

As discussed above, the textural similarities between some CLS and meteoritic chondrules are well documented (Table 7). The current data also show similarities in size distributions, shapes, and the presence of fine-grained rims and compound objects. Nebular models for chondrule formation have run into a number of difficulties: (1) Chondrule cooling rates are orders of magnitude slower than apply to reasonable nebular conditions; (2) oxygen and sodium fugacities are many orders of magnitude higher than cosmic; (3) chondrule number densities appear to have been very high; (4) chondrules and matrix have complementary compositions requiring that they be cogenetic and not readily separable; and (5) cosmogenic tracks are absent, implying that the chondrules did not exist independently and unshielded in interplanetary space. It makes sense to investigate more closely the possibility that meteoritic chondrules are, like the CLS, of impact origin (Sears *et al.*, 1995). Arguments against a formation of chondrules by impact were summarized by Taylor *et al.* (1983), and a similar approach to the discussion will be followed here.

Chondrule Abundances and Compound Chondrules–Since some type 3 chondrites consist of up to 75 vol% chondrules, it has been argued that the mechanism that produced them must have been very efficient. The efficiency of formation by impact would be governed primarily by the gravitational field. On a parent body like the Moon, giant impacts are necessary to provide a medium for slow cooling and flight times long enough to allow crystallization. Such impacts were rare and the relative abundances of CLS, glass spherules, and agglutinates are qualitatively consistent with crater size-frequencies (see Hörz *et al.*, 1991), so that CLS are globally rare. On small parent bodies, the reduced gravity facilitates long flight times and the formation of an insulating cloud of gas and dust, especially if the body has a porous surface and was volatile rich.

It is possible that chondrules are actually rare on asteroids because our meteorite collections are heavily biased towards meteorites that can be ejected from the asteroid belt and survive atmospheric passage. Certainly, the frequency of occurrence of the various asteroid types is quite different from the frequency with which their look-alikes fall to Earth, even allowing generously for space weathering or other processes that camouflage asteroid surfaces (Sears, 1997). The chondrites falling to Earth may be coming from a few tough asteroids conveniently located near orbital resonances, the asteroid "escape hatches." Gaffey (1996) argues that all of the H chondrites derive from a single asteroid, 6 Hebe, for instance.

The apparent lack of compound chondrules on the Moon is no longer a point of difference between meteoritic chondrules and lunar CLS. We found several compound CLS in just the three thin sections we examined, amounting to 8% by number of our spherules.

The Relative Number of Agglutinates and Glass Spheres–Since agglutinates are known to be an abundant component of the lunar regolith, it has been argued that an impact origin for meteoritic chondrules would have resulted in more agglutinates being present in the meteorites than chondrules. Agglutinates have been observed in two gas-rich regolith breccia meteorites, the Fayetteville chondrite (Taylor *et al.*, 1983) and the Jodzie howardite (Bunch and Rajan, 1988), so the differences in the processes occuring on the surfaces of the Moon and the meteorite parent bodies are of degree not kind. Regolith formation, evolution, and dynamics on small bodies and the Moon are expected to be very different (*e.g.*, McKay *et al.*, 1989a), and agglutinates should not be expected in rocks formed on small bodies for the following reasons.

(1) Lunar agglutinate formation in large amounts requires that the material to be turned into agglutinates remains in the upper few centimeters of the regolith for extended periods of time (*e.g.*, McKay and Basu, 1983), but burial rates on asteroids exceed excavation rates due, in part, to a lower ratio of small to large impacts than is found at 1 A.U. (Housen *et al.*, 1979).

(2) Since the large gravity field of the Moon allows even the smallest impactors to produce melt, lunar agglutinates are largely formed by local small-scale micrometeorite impact. Only the largest impacts on asteroids are expected to produce significant melting.

(3) Asteroidal impacts that do have sufficient kinetic energy to produce melts result in crystalline spherules rather than agglutinates because of systematically longer flight times (recall time of flight is proportional to $1/\sqrt{g}$) and solidification before landing in the lower gravity field.

(4) A lower abundance of agglutinates in meteorites can also result from the fragility of agglutinates. Chondrites have porosities <20% (Yomogida and Matsui, 1982), implying that agglutinates may have been destroyed by lithification. Experimentally, lunar agglutinates are easily destroyed (Simon *et al.,* 1986), and most Apollo 15 and 16 breccias with <25% porosity contain <1 vol% agglutinates despite I_s/FeO and solar gas contents that are indicative of high maturity (McKay *et al.*, 1986, 1989b). Similarly, many lunar meteorites, that are necessarily compact since they survived ejection and passage through Earth's atmosphere, are regolith breccias essentially free of agglutinates (*e.g.*, Palme *et al.*, 1991).

Rates of Impact Comminution Versus Melting–Since comminution is so efficient on the Moon, and assuming impact-melting is inefficient, it has been argued that if chondrules were formed by impact, chondrites ought to consist mostly of unmelted, clastic debris. Of course, a great many complex events occur during impact and only 15% of the energy of a terrestrial impact goes into comminution (Gault and Heitowit, 1963). The high burial rates on asteroids and the lack of large numbers of energetic micrometeorites, relative to the Moon, mean that comminution on asteroid surfaces is expected to be less efficient than on the Moon. The gas-rich regolith breccia meteorites, in which the "dark matrix" is comminuted clast material, often contain well-preserved chondrules. Ngawi is a particularly good example (Sears *et al.*, 1991).

Taylor *et al.* (1983) and Keil *et al.* (1997) have argued that impact melting on a planetary surface is an inefficient process. If

TABLE 7. Crystalline lunar spherules reported in the literature.

Source rock*	Authors' description and details	Reference†
11085	Skeletal olivine (Fa_{24-26}) in Mg-Fe-poor, Si-Al-rich glass, like some Chainpur chondrules; 400 μm across	Fredriksson *et al.* (1970); Fig. 2
14076,7	"round micropoikilitic chondrule, 0.6 mm across."	Warren *et al.* (1990); Fig. 13
14168	Basaltic fragment with rim; 660 μm across.	Nelen *et al.* (1972); Fig. 13
14259	Barred olivine Fa_3; 90 μm across.	Nelen *et al.* (1972), Fig. 10
14313	Lithic chondrule; 600 μm across.	King *et al.* (1972a); Fig. 2
14313	Lithic chondrule; 200 μm across.	King *et al.* (1972a); Fig. 2
14313	Lithic chondrule; 400 μm across.	King *et al.* (1972a); Fig. 2
14313	Lunar chondrule similar to primitive meteorites; neither droplet nor lithic; 400 μm across; thick rim.	King *et al.* (1972a); Fig. 3
14313	Euhedral pyroxene and olivine in brown transparent glass; 750 μm across.	King *et al.* (1972b), Fig. 1
14313/14318	Abraded clasts, 400 μm, across.	King *et al.* (1972b), Fig. 5
14313/14318	Abraded clasts, 400 μm across.	King *et al.* (1972b), Fig. 5
14315,11	Figure of two (maybe more) "chondrule-like" clasts. Bulk analyses for 10 objects	Dence and Plant (1972)
14315	Chondrule cluster; plagioclase needles; glass with spherules and fragments; 5–10% or rock.	Nelen *et al.* (1972), Fig. 7
14318,4	Plagioclase laths in fine grained fibrous matrix; plastic deformation; 280 μm across; ANT composition.	Kurat *et al.* (1972), Fig. 1
14318,4	Plagioclase laths in fine grained fibrous matrix; vague outlines for spherules; 240 μm across; ANT composition.	Kurat *et al.* (1972), Fig. 2
14318,4	Plagioclase and olivine in fine grained opaque matrix; 140 μm across; KREEP composition.	Kurat *et al.* (1972), Fig. 3
14318,4	Plagioclase microphenocrysts and laths in fine grained matrix; 400 μm across; attached to sphene; plastic deformation; ANT composition.	Kurat *et al.* (1972), Fig. 4
14318,4	Fine grained crystals in dark red opaque matrix; basaltic composition; bulk Fe# 0.596; 25 μm across.	Kurat *et al.* (1972)
14318,4	Fine grained crystals in dark red opaque matrix; basaltic composition; bulk Fe# 0.355; 40 μm across	Kurat *et al.* (1972)
14318	Fluid drop; pyroxene and plagioclase crystals nucleate at edges in brown glass; 500 μm across.	King *et al.* (1972a), Fig. 1
14318	Plagioclase (An_{87-90}); 260 μm across.	Nelen *et al.* (1972), Fig. 12
15365	43.3 SiO_2, 15.8 MgO, 21.3 FeO, 9.0 CaO, 8.3 Al_2O_3, 0.10 Na_2O	Bunch *et al.* (1972)
61175	"... a droplet chondrule ... filled with plagioclase"	Winzer *et al.* (1977), Fig. 2a
62275	Barred olivine "chondrules"	Roedder and Weiblen (1977)
68001,6039 (ddt)	"chondrule-like object"; olivine within clinopyroxene and yellow glass, 2 mm across	Holder and Ryder (1995), figure
68001,6044 (ddt)	"chondrule-like object," 1 mm across	Holder and Ryder (1995)
68001,6051 (ddt)	"chondrule-like object," 500 μm	Holder and Ryder (1995)
Luna 16 core tube	Igneous; large plagioclase crystals in fine grained anisotropic matrix; "textures not distinguished from meteorites," "porphyritic to excentro-radial"; compositions given.	Keil *et al.* (1972), Fig. 12
MAC 88105,87	"chondrule-like" objects	Koeberl *et al.* (1991)
Y 82192	crystallized spherule	Bischoff *et al.* (1987), Fig. 6a
Y 82193	crystallized spherule	Bischoff *et al.* (1987), Fig. 6b

*ddt, double-drive tube.
†With figure number in original reference, if applicable.

this were true, it would be difficult to understand why 30% of highland Apollo samples are of impact melt origin (Simonds *et al.*, 1976) or why impact melt lithologies have been recognized at over 60% of the known terrestrial impact structures (Grieve *et al.*, 1977). Although impacts tend to break substantial portions of rock, and most lunar rocks and meteorites are breccias, more important is the amount of melt that is ejected from the crater. Dence *et al.* (1977) calculate that ~75% of the generated melt volume was ejected from the simple Brent crater (4 km diameter) while ~50% was ejected from the complex Manicouagan structure (75 km). In the case of Manicouagan, the melt volume was ~35% of the transient cavity volume (Dence *et al.*, 1977). The reduced gravity of smaller bodies should result in smaller amounts of melt relative to impact on a larger body, but the highly porous regoliths present on the Moon and asteroids will substantially enhance melt production (Schaal *et al.*, 1979). The lower gravity on asteroids will also ensure that a greater fraction of the melt leaves the crater.

Hypervelocity Impact Pits–Microcraters from high-velocity micrometeorite impacts have been observed on the surfaces of many lunar regolith particles, yet none have been found on chondritic chondrules. This also reflects differences in regolith dynamics between large and small bodies. Acquisition of hypervelocity impact craters requires a long surface exposure and a high flux of high-velocity micrometeorites, which are two conditions not expected on asteroidal

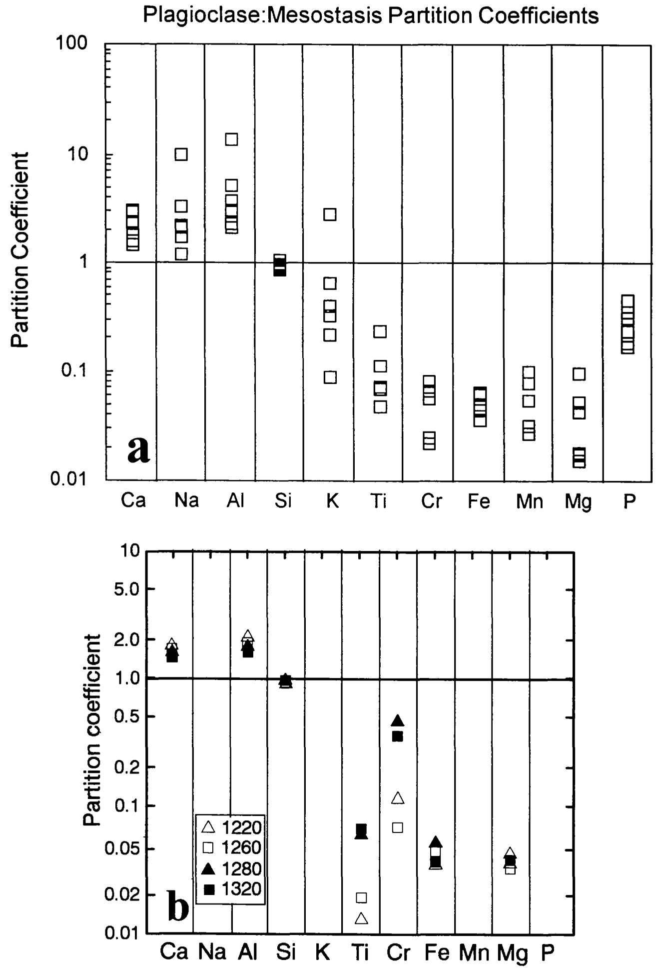

FIG. 9 (a) Apparent partition coefficients for plagioclase and mesostasis in the present CLS and (b) partition coefficients derived from experiments with melts of lunar highland composition (Weill and McKay, 1975). The values 1220–1320 refer to temperatures in degrees celsius at which the experimental charges crystallized.

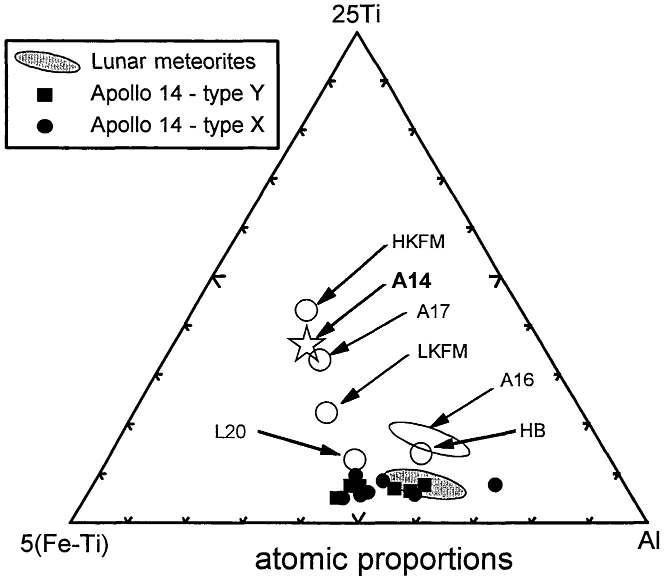

FIG. 10. A ternary plot, after Delano (1991), of element concentrations thought to be diagnostic of lunar impact glass spherule sources. A14, A16, A17 and L20 refer to Apollo 14, 16, 17 and Luna 20 highland regolith compositions. Also plotted are compositional fields representing three groups of widespread highland glass (HKFM, LKFM, and HB refer to high-K Fra Mauro basalt, low-K Fra Mauro basalt and highland basalt). The lunar meteorite field represents bulk compositions of Y-791197, ALH 81005, Y-82192, Y-86032, and MAC 88105 and the impact spherules they contain. See Delano (1991) for data sources for all fields other than the current data. The Apollo 14 CLS bulk compositions resemble the highland meteorites and their impact spherules but are unlike the common impact glasses or any Apollo highland regoliths, including that of Apollo 14. The CLS raw data are plotted since the "density-corrected" data are not significantly different.

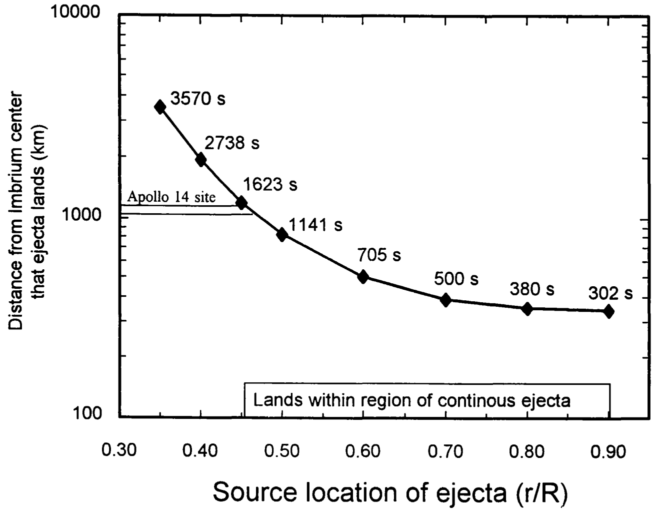

FIG. 11. Plot of total distance from crater center that ejecta lands for material ejected during the formation of the Imbrium basin against the original location of the material within the crater (in terms of fractions of the crater radius). The Fra Mauro region lies near the limit of continuous ejecta. The time above each datum is the free-flight duration. (Method of Ahrens and O'Keefe, 1978, and 45° ejection angle were used).

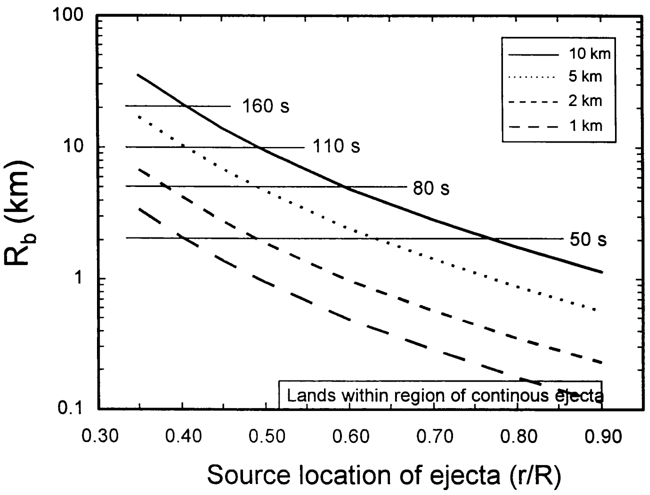

FIG. 12. Plot of the ballistic range (R_b) of material ejected during formation of relatively small (1, 2, 5, and 10 km) craters on the Moon. The times indicated are the free-flight durations of ejecta with a given ballistic range. Same methods as in Fig. 11, although ballistic range is shown instead of total distance from center since the craters are small and the two quantities are nearly equal.

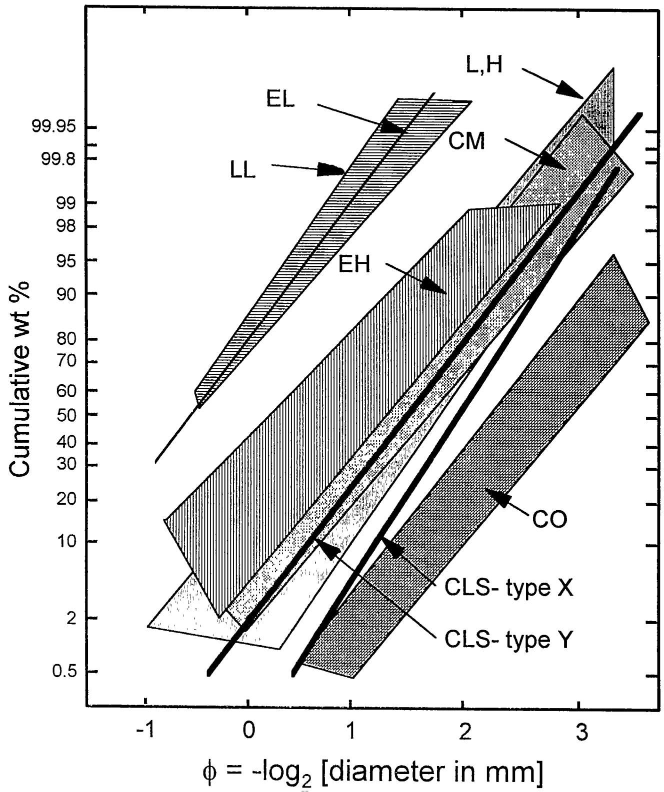

FIG. 13. Cumulative size-frequency distributions for CLS compared with chondrules from meteorites. Crystalline lunar spherules have very similar cumulative size-frequency distributions to meteoritic chondrules from several classes of meteorite. Data sources for meteoritic chondrules are as follows: Murchison CM chondrite, Sears *et al.* (1993); L, H chondrites, King and King (1979); EH chondrites, Rubin and Grossman (1987); EL chondrites, Taunton *et al.* (1996); the Semarkona LL chondrite, Huang *et al.* (1996); CO chondrites, Rubin (1989). In the case of the LL, CM, CO and EH fields, chondrules of different classes define the long edges of the fields. In the case of the L, H field, L chondrites plot to the left of the field and H chondrites define the right of the field since chondrules in H chondrites tend to be smaller than those in L chondrites.

bodies due to the high burial rates and low flux of micrometeorites mentioned above. Thus, impact pits should not be expected to be found on chondritic chondrules.

Ages–If chondrules formed by impact, then some should be younger than 4.4 Ga, since the target must be younger than its impact products. Also, if the impacts were associated with the 3.9 Ga event that affected the Moon and the basaltic meteorite parent body, then chondrules should have 3.9 Ga ages. There is now strong evidence that chondrules formed after their parent bodies, but there is no evidence that the chondrite parent bodies experienced the 3.9 Ga terminal cataclysm.

The refractory calcium-aluminum inclusions (CAI) in CV and other chondrites are probably the first solar system solids to form, followed by several classes of igneous meteorite. They have the oldest Pb-Pb ages and high initial $^{26}Al/^{27}Al$, and low initial $^{87}Sr/^{86}Sr$ (Podosek and Cassen, 1994; MacPherson *et al.*, 1995). Chondrules formed up to several million years after CAI (Hutcheon *et al.*, 1994; Hutcheon and Jones, 1995). The absence of ^{26}Al suggests formation 1.6 to 6.3 Ma after CAI formation, while Mn-Cr systematics suggest a 6 Ma interval between chondrule formation and the formation of several diverse classes of meteorite. In fact for Chainpur, chondrule formation may have lasted as long as 50 Ma (Swindle *et al.*, 1996).

Impact Velocities–The discovery that chondrules are several million years younger than the first formed solids in the early solar system resolves another difficulty with chondrule formation by impact which is that impact velocities in the asteroid belt were assumed to be insufficient for melt production. However, within a few hundred thousand years of the age of the solar system, Jupiter or proto-Jupiter will have formed (Cameron, 1995; Wetherill and Stewart, 1987; Zuckerman *et al.*, 1995) and by gravitational interactions with material in the asteroid belt have caused relative velocities to reach 5 km/s, which is sufficient for significant melt production by impact (Davis *et al.*, 1979).

Absence of Prechondrule Target Rock in our Meteorite Collections?–An impact scenario requires the existence of precursor target rock. Assuming parent bodies accreted from fine-grained nebular dust, and that this material could sometimes survive chondrule formation, we might expect to see it in our meteorite collections. Huang *et al.* (1996) proposed that the target rock for the ordinary chondrites was "CI-chondrite-like"; maybe it was the CI chondrites, in which case we have several in our collections. However, the meteorites falling to Earth are not representative of the asteroid belt, partly because few friable dust or mud balls (such as "fine-grained nebula dust") would survive passage through the Earth's atmosphere (Boss, 1996; Sears, 1997).

Chondrule Rims–Many chondrules in unequilibrated ordinary chondrites are rimmed and any theory for chondrule origin must account for these rims. Taylor *et al.* (1983) thought that "...the only possibility offered by impact models is rolling around in regolith dust" and since individual mineral grains, rock fragments, and glass spheres from the Moon do not have rims, this process was thought unrealistic. In fact, CLS do have rims, albeit much thinner than meteorite chondrule rims. Hewins (1989) and Huang *et al.* (1996) have recently suggested that meteorite chondrule rims were produced when volatiles, perhaps lost from the chondrules during heating, condensed onto their surfaces as they cooled during free flight. In this case, much thinner rims on lunar CLS would be consistent with the Moon's volatile-poor surface materials. Impact vapor deposition may be an important process for many lunar surface materials (Keller and McKay, 1993, 1995).

Chondrule Compositions–In order for an impact scenario to account for the wide compositional variety displayed by chondrules, the chondrite parent bodies would have to be compositionally heterogeneous so that numerous impacts could melt a wide variety of targets. It is just as likely as not for the parent body to be heterogeneous on the milligram scale, but more importantly, it is possible (we would actually argue very likely) that chondrule diversity was created during chondrule formation as some chondrules experienced FeO reduction and evaporative loss of major and minor elements while others did not (Alexander, 1996; Huang *et al.*, 1996; Sears *et al.*, 1996b).

SUMMARY AND CONCLUSIONS

Crystalline lunar spherules have shapes and igneous textures that are consistent with crystallization from molten droplets at cooling rates well below that expected by cooling in a vacuum. Some crystalline lunar spherules have thin, fine-grained, opaque rims. Compound CLS exist. Crystalline lunar spherules are as abundant in the present Apollo 14 breccias as chondrules are in certain chondrite classes such as the CM chondrites that are ~5 vol% chondrules. They also have very similar cumulative size-frequency distributions to many meteorite chondrule populations, which might suggest a similar mode of formation (see King and King, 1979; Rubin and Grossman, 1987; Rubin, 1989).

Based on the requirement that CLS are produced only in major impacts, we would expect them to be much less abundant on the lunar surface than glass spherules or agglutinates. They should be even more rare on Earth due to the much larger size of the target and the necessary increase in the size of the impact required to provide long enough flight times to allow crystallization. It is clear from terrestrial crater studies that melt spherules are commonly produced as a result of impact. Thirty centimeter thick beds of impact melt spherules have been found in the ejecta component of the K/T impact event (Bohor and Glass, 1995). Glass spherules somewhat analogous to the lunar glass spherules, with minimum crystallization, have been found in breccia from the Ries crater (Graup, 1981; Sears *et al.,* 1996c), but nothing approaching the CLS have been found. Clearly, whether melt spherules created in an impact event crystallize or remain glassy critically depends on flight time, which in turn is governed by the gravitational field of the target.

Several crystalline spherules have been found in howardites and were called "chondrules" by Olsen *et al.* (1989). They have textures and other properties similar to meteoritic chondrules, but they have bulk compositions (in particular Fe/Mn) similar to the host meteorites in which they are found. They appear to be spherules produced by impact on a basaltic surface. It also seems, after artificial satellite flybys of Phobos and the asteroids, and contrary to the views of two decades ago, that thick regoliths are possible on asteroids and that relatively small amounts of ejecta actually escape entirely (Housen *et al.*, 1979; Langevin and Maurette, 1980; Asphaug and Nolan, 1992; Housen, 1992; Asphaug and Melosh, 1993). Chondrules arguably formed well after their postulated parent bodies and after Jupiter or proto-Jupiter had increased relative velocities in the asteroid belt to 5 km/s, which is sufficient for melt production. Based on our investigations of CLS, we suggest that meteoritic chondrules could have had an origin resulting from impact onto a planetary surface.

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