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Crystal-bearing lunar spherules: Impact melting of the Moon's crust and implications for the origin of meteoritic chondrules

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Abstract–Crystal-bearing lunar spherules (CLSs) in lunar breccia (14313, 14315, 14318), soil (68001, 24105), and impact-melt rock (62295) samples can be classified into two types: feldspathic and olivine-rich. Feldspathic CLSs contain equant, tabular, or acicular plagioclase grains set in glass or a pyroxene–olivine mesostasis; the less common olivine-rich CLSs contain euhedral or skeletal olivine set in glass, or possess a barred-olivine texture. Bulk-chemical and mineral-chemical data strongly suggest that feldspathic CLSs formed by impact melting of mixtures of ferroan anorthosite and Mg-suite rocks that compose the feldspathic crust of the Moon. It is probable that olivine-rich CLSs also formed by impact melting, but some appear to have been derived from distinctively magnesian lunar materials, atypical of the Moon's crust.

Some CLSs contain reversely-zoned "relic" plagioclase grains that were not entirely melted during CLS formation, thin (≤5 μm thick) rims of troilite or phosphate, and chemical gradients in glassy mesostases attributed to metasomatism in a volatile-rich (Na-K-P-rich) environment. Crystal-bearing lunar spherules were rimmed and metasomatized prior to brecciation. Compound CLS objects are also present; these formed by low-velocity collisions in an environment, probably an ejecta plume, that contained numerous melt droplets.

Factors other than composition were responsible for producing the crystallinity of the CLSs. We agree with previous workers that relatively slow cooling rates and long ballistic travel times were critical features that enabled these impact-melt droplets to partially or completely crystallize in free-flight. Moreover, incomplete melting of precursor materials formed nucleation sites that aided subsequent crystallization.

Clearly, CLSs do not resemble meteoritic chondrules in all ways. The two types of objects had different precursors and did not experience identical rimming processes, and vapor fractionation appears to have played a less important role in establishing the compositions of CLSs than of chondrules. However, the many detailed similarities between CLSs and chondrules indicate that it is more difficult to rule out an origin for some chondrules by impact melting than some have previously argued. Differences between CLSs, chondrules, and their host rocks possibly can be reconciled with an impact-melt origin for some chondrules when different precursors, the higher gravity of the Moon compared to chondrite parent bodies, and the likely presence of nebular gas during chondrule formation are taken into account.

INTRODUCTION

It has long been recognized that some lunar samples contain objects that resemble meteoritic chondrules. These objects, termed crystal-bearing lunar spherules (CLSs), have textures suggestive of crystallization from freely-floating melt droplets and are found mainly in samples of highlands materials (*e.g.*, King *et al.*, 1972a,b; Nelen *et al.*, 1972; Kurat *et al.*, 1972b, 1974; Fredriksson *et al.*, 1973; Roedder and Weiblen, 1977). However, much of the previous research on CLSs was of a preliminary nature and done at a time when far less was known about the Moon and meteorite parent bodies. Recently, interest in CLSs has been revived, as evidenced by the work of Symes *et al.* (1998a).

Crystal-bearing lunar spherules constitute a significant (~8 vol%; Symes *et al.*, 1998a) portion of two lunar breccias, 14315 and 14318. In most lunar samples, CLSs are not especially abundant, but 14315 and 14318 are dominated by glassy and crystal-bearing spherules (*e.g.*, Chou *et al.*, 1972; Symes *et al.*, 1998a). Some researchers have given these samples unique classifications that reflect their unusual characteristics. For example, Chou *et al.* (1972) placed them in a separate category of "spherule-rich microbreccia"; and Kurat *et al.* (1972a, 1974) variously described 14318 as "a chondrite of lunar origin" and as a "polymict breccia with chondritic texture."

All researchers who have studied CLSs agree that impact processes were responsible for their formation, and those who have drawn comparisons to meteoritic chondrules have further concluded that meteoritic chondrules could have formed by impact processes (*e.g.*, King *et al.*, 1972a,b; Kurat *et al.*, 1972b; Fredriksson *et al.*, 1973; Sears *et al.*, 1996a; Symes *et al.*, 1998a). However, as suggested by Boss (1996) and Hewins (1997), most researchers do not currently favor an impact-melt ("planetary") origin for meteoritic chondrules. Instead, most favor an origin in the solar nebula. Taylor *et al.* (1983) marshalled succinct arguments against forming chondrules by impact processes, basing their conclusions in large part on analogies with lunar samples. Their arguments are widely cited by researchers (*e.g.*, Grossman, 1988; Boss, 1996) as the main reasons why an impact-melt origin for chondrules is unlikely. Symes *et al.* (1998a) rebutted all of the arguments made by Taylor *et al.* (1983) and concluded that chondrules could have formed by impact-melting of asteroidal parent bodies.

In this paper, results are presented of a petrographic and electron microprobe study of CLSs in various types of lunar samples, including breccias (14315, 14318, 14313), two soils (68001, 24105), and one igneous-textured, apparently impact-melted rock (62295). Further evidence for the role of impact processes in the origin of the CLSs is provided, and comparisons are made between CLSs and meteoritic chondrules. Preliminary results were described by Ruzicka *et al.* (1998a,b).

Before proceeding, it is prudent to elaborate on terminology. Crystal-bearing lunar spherules (CLSs) are defined as drop-formed or irregularly-shaped lunar objects that are significantly crystalline and that have textures suggestive of crystallization from melts. Included in this definition are objects that probably represent brecciated spherules. In the past, different terminology has been used for CLSs, including "lunar chondrules" (*e.g.*, King *et al.*, 1972a,b; Keil *et al.*, 1972; Kurat *et al.*, 1972b), "chondroids" (Ivanov *et al.*, 1976), and "devitrified glass spherules" (Sears *et al.*, 1996a,b). Although possibly analogous to meteoritic chondrules, the moniker "lunar chondrules" is here avoided as being too suggestive. The term "chondroids" is more neutral but suffers from being nondescriptive. "Devitrified glass spherules" implies a particular origin for the spherules that is probably incorrect (as Symes *et al.*, 1998a, acknowledged); based on textures and mineral-chemical evidence, most of the CLSs appear to have crystallized from melts, not glasses. Here, the term "crystal-bearing lunar spherules" is used instead of "crystalline lunar spherules" (Symes *et al.*, 1998a,b; Symes and Lofgren, 1998) to acknowledge that the objects are not always holocrystalline.

SAMPLES AND ANALYTICAL METHODS

Samples

The polished thin-section collection of selected lunar samples were surveyed at the curatorial facility of the Johnson Spacecraft Center. A review of the literature on CLSs insured that a representative variety of CLS types was analyzed. The samples studied include sections of spheroid-rich breccias 14315 and 14318, breccia 14313, soil samples 68001 and 24105, and igneous-textured rock 62295. Rock 62295 is a spinel troctolite that appears to have formed as a clast-laden melt (*e.g.*, Roedder and Weiblen, 1977; Wilshire *et al.*, 1981) from a source region unlike that typical for Apollo 16 rocks (Taylor *et al.*, 1991). This rock contains barred-olivine

TABLE 1. Petrographic data for crystal-bearing lunar spherules and glass objects in this study.

Object	PTS	Diameter (μm)*	Textural type†	Description†
Luna 24				
24A	24105,15	450	plag-porph (Y)	ellipsoid; skeletal plag set in glass
Apollo 16				
62A	62295,67	350	barred ol	spheroidal barred ol core; granular plag and ol "rim"
62B	62295,69	750 × 390	barred ol	half of originally spherical (?) ol crystal
68A	68001,6046	230	plag-porph (Y)	broken spheroid; tabular and acicular plag set in glass
68B	68001,6046	150	plag-porph (Y)	rimmed ellipsoid; tabular plag set in glass
68C	68001,6028	300	glassy	partly-rimmed, glass-rich spheroid
68D	68001,6028	210	glassy	rimmed, glass-rich spheroid
68E	68001,6028	320	plag-porph (XY)	clast; plag-rich; interstitial ol, pig, glass
68F	68001,6028	315	plag-porph (Y)	clast; tabular plag and mesostasis pig, glass
Apollo 14				
318c1	14318,8	425	plag-porph (Y)	spheroid; tabular plag; mesostasis ol, pig, plag
318c2	14318,8	770 × 460	plag-porph (XY)	consists of glassy and crystal-bearing portions
318c4	14318,8	525	ol-porph	partly rimmed spheroid; skeletal ol; mesostasis glass, plag
318g5	14318,8	350 (core)	mantled glass	glassy ellipsoidal core surrounded by partly crystalline rim
318c5	14318,8	–	plag-porph (XY)	150 μm-thick rim on glassy ellipsoid 318g5
318c6	14318,4	335	plag-porph (XY)	spheroid; tabular/equant plag and mesostasis ol, pig
318c9	14318,4	280	plag-porph (Y)	spheroid; plag laths set in glass
318c10	14318,4	285	plag-porph (Y)	spheroid; plag laths; mesostasis glass, ol, Al-aug
315c1	14315,47	355	plag-porph (Y)	ellipsoid; tabular plag set in meso with glass, pig, ol
315c2	14315,47	210	plag-porph (X)	partly rimmed ellipsoid; anhedral plag, pig, ol
315c3	14315,47	220	plag-porph (X)	clast; equant plag; contains opx, aug, glass, ilm, ZrO_2
315c4	14315,47	230	plag-porph (X)	partly rimmed ellipsoid; plag set in pig + plag mesostasis
315c5	14315,47	350	plag-porph (XY)	comma-shaped object; plag set in pig + plag mesostasis
315c6	14315,47	290	plag-porph (Y)	spheroid; tabular plag and mesostasis pig, glass, ol, sp
315c7	14315,47	180	plag-porph (XY)	clast; tabular/equant plag and mesostasis glass, aug, ol, opx
315c8	14315,47	300	plag-porph (Y)	clast; tabular plag set in glass; contains aug, ol, opx, ilm
315c9	14315,47	175	plag-porph (XY)	brecciated, partly rimmed droplet; equant and tabular plag
31510	14315,47	180,60,60	plag-porph	three-part compound object
315c11	14315,47	300	plag-porph (X)	spheroid; equant plag set in plag, pig, ol mesostasis
315g1	14315,47	300	glass	rimmed spheroid
315g2	14315,47	270 × 140	glass	irregularly-shaped object; five different attached sections
315g3	14315,47	350 × 200	glass	irregularly-shaped object
315d1	14315,47	450 (core)	mantled glass	cryptocrystalline core; ≤150 μm-thick Type X rim
313A	14313,55	925	ol-porph	rimmed spheroid; tabular/skeletal ol set in glass

*Approximate average diameter in thin section.

†Abbreviations: plag-porph = microporphyritic plagioclase; ol-porph = microporphyritic olivine; barred ol = olivine containing parallel bars of plagioclase or feldspathic glass; plag = plagioclase; pig = pigeonite; aug = augite; opx = orthopyroxene; ol = olivine; ilm = ilmenite; X = plagioclase has primarily granular habit; Y = plagioclase has primarily tabular habit.

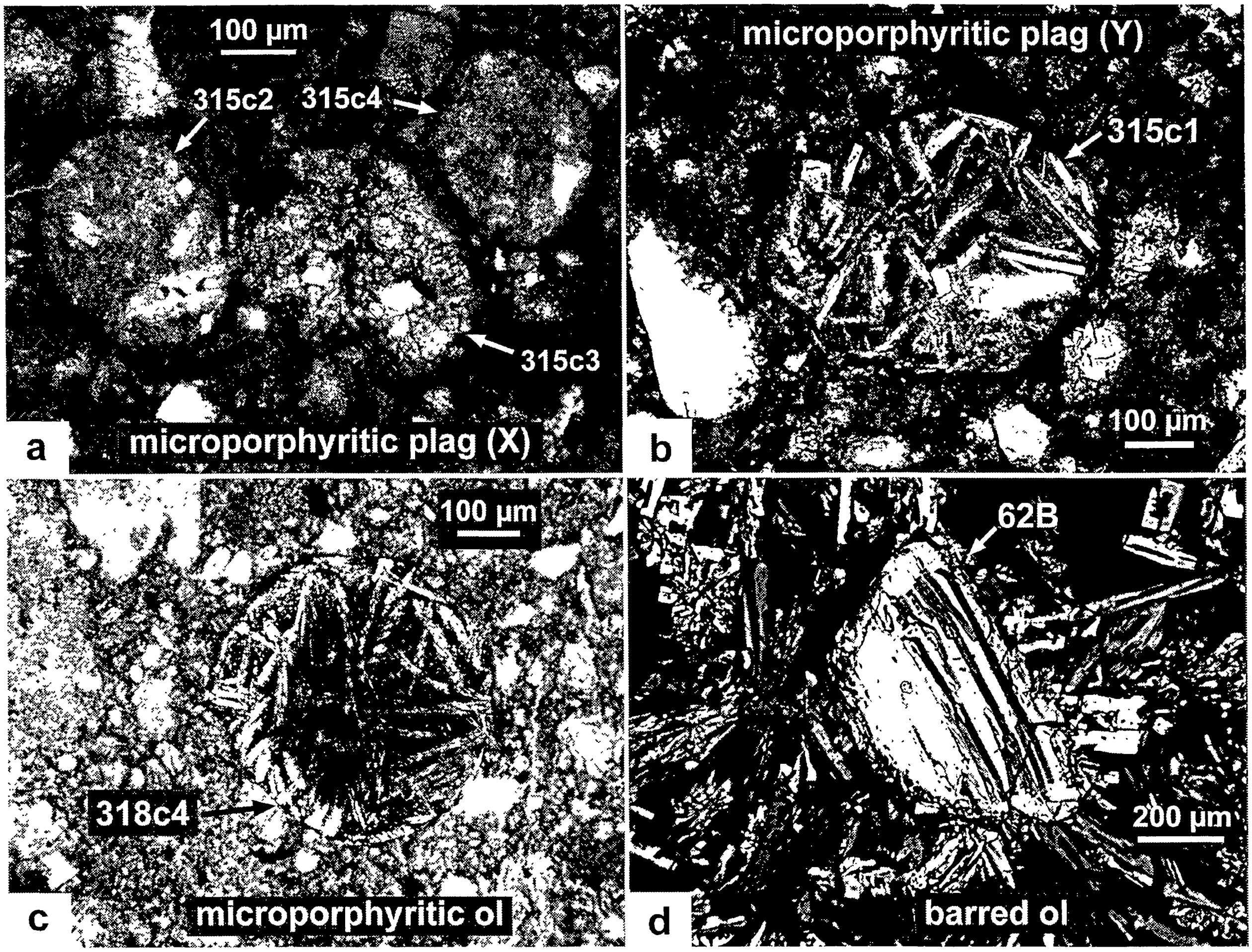

FIG. 1. Micrographs illustrating various CLS types, obtained in plane-polarized transmitted light (a–c) and cross-polarized transmitted light (d). (a) Type X microporphyritic plagioclase CLSs 315c2, 315c3, and 315c4, which contain granular plagioclase grains (white) in a mesostasis of pyroxene and olivine. (b) Type Y microporphyritic plagioclase CLS 315c1, which contains tabular plagioclase grains set in an olivine–pyroxene mesostasis. (c) Olivine-rich CLS 318c4, which contains tabular-to-skeletal olivine microphenocrysts (light) set in a glassy (dark) groundmass. Olivine grains appear to have nucleated along one margin of the CLS (bottom right). (d) Barred-olivine CLS 62B, which consists of a single grain of olivine (white) that encloses parallel bars of plagioclase and glass (dark grey and black) and appears to represent a fragment of a spheroid that was originally twice as large. The host rock has an igneous texture.

objects (Roedder and Weiblen, 1977) that presumably represent solid debris that was incorporated into the melt matrix of the host. For comparison with the CLSs, glassy spheroids and irregularly-shaped glass objects in 14315, 14318, and 68001 were also studied. Data for the CLS and glass objects analyzed are given in Table 1.

Analytical Methods

A Cameca SX-52 electron microprobe was utilized for quantitative phase analyses, x-ray mapping, and backscattered electron (BSE) studies. Quantitative wavelength dispersive spectrometer (WDS) analyses were obtained using a 15 keV operating voltage and a beam current of 20 or 30 na. In most cases, the beam was focussed to a ~1 μm diameter spot; but for glassy spheroids, the beam was expanded to approximately 25–50 μm. Modal analyses of CLSs were obtained with a digital x-ray-mapping technique, using a procedure similar to that described by Ruzicka *et al.* (1998c). X-ray images were obtained to cover all of the exposed surfaces of the objects of interest. The typical error in modal abundances for the phases is estimated to be ±5% (relative). The bulk compositions of CLSs were reconstructed by combining modal and phase-compositional data. Areal abundances were converted to weight fractions for the various phases by assuming appropriate densities (as given in Gaines *et al.*, 1997). Errors in the reconstructed compositions were estimated by propagating uncertainties in the modes and phase compositions (the latter assumed to be given by the standard deviations of averaged phase compositions).

RESULTS

Crystal-bearing lunar spherules in the sample suite can be classified into two main types, feldspathic and olivine-rich, which can be further subdivided on the basis of texture. Other rare types of CLSs (including "basaltic" varieties) appear to exist (Kurat *et al.*, 1972b, 1974; Keil *et al.*, 1972; Symes *et al.*, 1998a) but are not present in the sample suite. Data pertaining to the textures, modes, phase compositions, and calculated bulk compositions of CLSs are given in Tables 1–4.

Feldspathic Crystal-bearing Spherules

Feldspathic CLSs contain equant, tabular, or skeletal plagioclase grains set in glass or a largely crystalline mesostasis containing mainly pyroxene and olivine (Fig. 1a–b). Their modes are dominated by plagioclase (22–92 vol%) or feldspathic glass (up to 70%) (Table 2). Sears *et al.* (1996a) and Symes *et al.* (1998a) classified feldspathic CLSs in 14315 and 14318 into two main textural types: Type X and

TABLE 2. Modal composition (vol%) of crystal-bearing lunar spherules.*

	Plag	Ol	Pig	Aug	Opx	Sp	Ilm	Tr	Phos	Glass	Comment
Feldspathic crystal-bearing lunar spherules											
24A	21.9	0.3	0.2	3.7	–	3.1	<0.1	0.4	–	70.4	–
68A	65.4	0.1	0.4	–	–	0.2	–	<0.1	–	33.9	–
68B	76.1	–	–	0.3	–	–	–	1.1	–	22.5	Includes sulfide rim
68E	91.7	5.5	–	–	–	0.1	–	0.2	–	2.5	–
68F	83.2	–	14.5	–	–	0.3	–	0.3	0.1	1.5	–
318c1	70.2	11.2	8.4	–	–	–	<0.1	0.1	0.1	9.0	–
318c5	45.6	7.7	4.9	–	0.9	0.3	–	0.6	–	40.1	Rim of mantled object
318c6	61.0	10.6	19.7	–	–	4.2	<0.1	0.1	0.2	4.3	–
318c9	61.2	3.9	3.1	–	–	0.6	–	0.1	0.1	31.0	–
318c10	60.0	1.6	–	1.8	–	0.8	–	0.1	0.1	35.7	–
315c1	66.5	2.2	6.5	–	–	<0.1	<0.1	0.2	–	24.5	–
315c2	70.3	6.1	23.2	–	–	0.3	–	0.2	–	–	–
315c3	52.0	0.1	–	10.3	30.1	0.1	1.4	0.4	1.4	4.2	Trace baddeleyite
315c4	69.0	0.7	29.6	–	–	0.1	<0.1	0.3	0.3	–	–
315c5	68.6	–	30.3	–	–	0.1	<0.1	0.1	<0.1	0.8	–
315c6	68.5	8.6	12.4	–	–	–	<0.1	0.1	<0.1	9.9	–
315c7	68.3	3.4	–	7.2	2.1	0.4	<0.1	0.4	–	18.1	–
315c8	59.3	2.1	–	7.0	1.6	0.2	2.0	0.6	0.1	27.2	–
315c9	60.4	8.0	–	13.4	1.8	0.7	0.1	0.3	1.0	14.2	–
315c11	82.5	4.9	11.8	–	–	0.5	0.1	0.1	0.1	0.1	–
Olivine-rich crystal-bearing lunar spherules											
62A	27.8	72.3	–	–	–	–	–	–	–	–	Includes core + rim
62B	2.0	96.0	–	–	–	–	–	–	–	2.0	Estimated mode
318c4	23.5	28.7	5.1	–	0.6	–	<0.1	0.4	0.3	41.4	–
313A	minor	53.5	–	<0.1	–	–	–	–	0.7	45.8	Includes phosphate rim

*Abbreviations: Plag = plagioclase; Ol = olivine; Pig = pigeonite (Wo_{5-25}); Aug = augite ($>Wo_{25}$); Opx = orthopyroxene ($<Wo_5$); Sp = spinel; Ilm = ilmenite; Tr = troilite; Phos = phosphate (merrillite).

Type Y. This classification is applicable to other samples as well (Table 1). Type X objects contain equant plagioclase microphenocrysts and are primarily granular-textured (Fig. 1a), whereas Type Y objects contain tabular or skeletal plagioclase microphenocrysts (Fig. 1b). Some CLSs have intermediate textures and are classified as Type XY (Table 1).

Plagioclase grains in the feldspathic CLSs are usually unzoned or slightly normally zoned (calcic cores, sodic rims). Their compositions typically range between An_{90-98} (Fig. 2a). One feldspathic CLS (315c3) contains plagioclase grains that vary widely in An content, An_{74-97} (Fig. 2a); this object is discussed in more detail below. Pyroxene grains are zoned normally (magnesian cores, ferrous and calcic rims) and consist mainly of pigeonite (Wo_{5-25}), although orthopyroxene ($<Wo_5$) and augite ($>Wo_{25}$) are present in some objects (Fig. 2b; Tables 1–3). Olivine grains in feldspathic CLSs often occur near the interface of plagioclase and pyroxene, and have compositions that range between Fo_{54-82} (Fig. 2c, Table 3). Chromium-rich spinel is a common accessory mineral, and ilmenite and phosphate are present in some objects (Table 2). Sample CLS 315c3, the object with the wide range in An contents in plagioclase, has a relatively evolved lithology, which includes ilmenite, trace amounts of baddeleyite, merrillite, and K-Ba-rich glass (Table 2).

Olivine-rich Crystal-bearing Lunar Spherules

Four of the crystal-bearing spherules contain little plagioclase or pyroxene and instead contain large amounts (28–96 vol%) of olivine (Table 2). These olivine-rich CLSs show different textures than feldspathic CLSs and also differ in terms of mineral chemistry. They more closely resemble the mineralogies and textures of many meteoritic chondrules.

Spherules 318c4 (Fig. 1c) and 313A (Fig. 5) are olivine microporphyries that contain euhedral or skeletal olivine set in a glassy groundmass; a few small plagioclase grains occur in the mesostases. The texture and mineralogy of 313A (Fig. 5) resembles that of a large (~0.9 mm in diameter) CLS olivine microporphyry in soil 64811 illustrated by Nelen *et al.* (1972) and Fredriksson *et al.* (1973). Sample CLS 313A also has a texture similar to a small (~0.1 mm in diameter) olivine-rich microporphyry in breccia 65095 illustrated by Symes *et al.* (1998b), except that the latter object has a more crystalline groundmass (Symes, pers. comm.). Previous work on objects 313A (Juan *et al.*, 1972; King *et al.*, 1972b) and 318c4 (King *et al.*, 1972b) led to erroneous descriptions of them being rich in orthopyroxene; microprobe data show that no orthopyroxene is present in them.

Objects 62B (Fig. 1d) and 62A (Fig. 6c) have a barred-olivine texture. The main parts of these objects consist of single grains of olivine that enclose thin (<10 μm-wide) parallel strips of plagioclase and possibly small amounts of glass; the olivine has the thick outer "shells" characteristic of barred-olivine chondrules (*e.g.*, Weisberg, 1987). The shape of 62B (Fig. 1d) suggests that it is a fragment of a spheroid that was originally twice as big. The "barred" olivine grain in 62A is surrounded by a crystalline rim of plagioclase + olivine that appears to be part of the object and contains none of the pyroxene found in the host (see below). These objects appear to be representative of barred-olivine objects in 62295 (Roedder and Weiblen, 1977). They also resemble the barred-olivine CLS in soil 14259 illustrated by Fredriksson *et al.* (1973).

Olivine grains in the olivine-rich CLSs are either normally zoned (313A, 318c4) or vary nonsystematically in fayalite content (62A, 62B). They are notably forsteritic (Fo_{78-98}) compared to olivine in

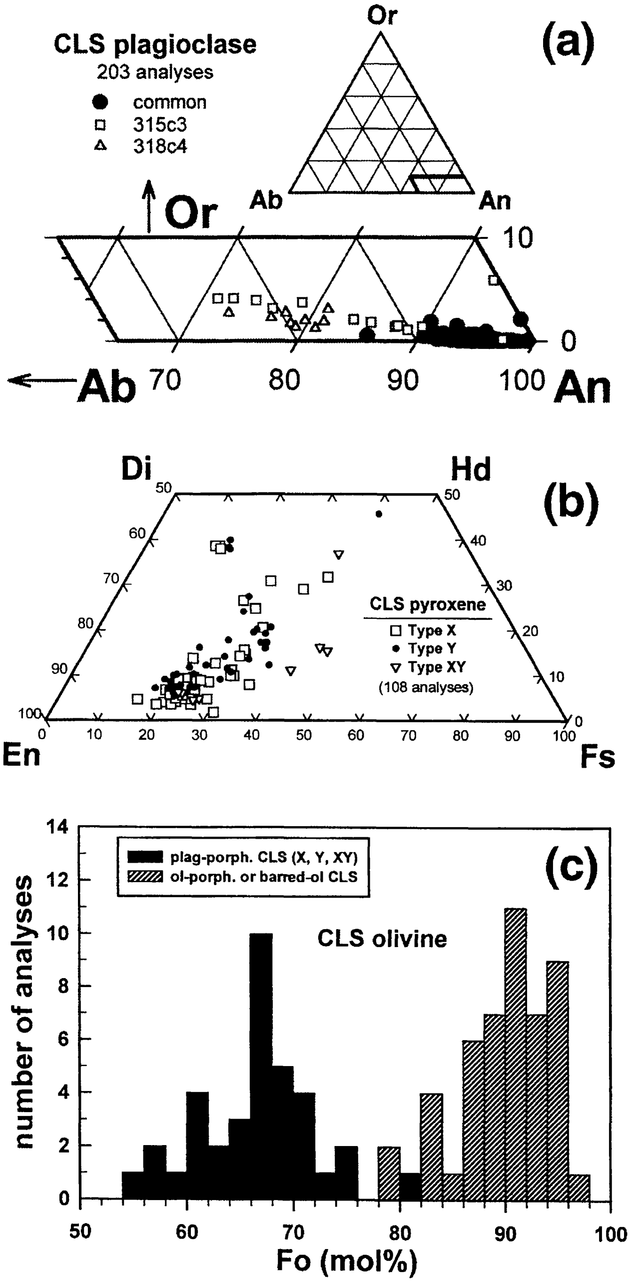

FIG. 2. Diagrams summarizing (a) plagioclase, (b) pyroxene, and (c) olivine compositions in CLSs. Abbreviations as in Table 1.

feldspathic CLSs (Fig. 2c). In contrast to plagioclase in feldspathic CLSs, plagioclase compositions in the olivine-rich CLSs vary widely, from An_{74-81} in 318c4, to An_{90-93} and An_{93-96} in 62A and 62B, respectively, to $An_{97.8-99.9}$ in 313A (Fig. 2a, Table 3).

Relic Grains

Some Type X CLSs (315c3, 315c4, 318c5, 318c6) contain plagioclase grains that have relatively sodic cores and more calcic margins (reverse-zoning patterns). The reversely zoned grains are among the largest grains in their respective objects. Object 315c3 contains the best example of a reversely zoned plagioclase grain (Fig. 3). One grain is clearly more sodic than other plagioclase grains in the object (Fig. 3, Table 3). The zoning pattern in this grain is roughly concentric, with the most sodic composition (An_{71}) found in the core and the most calcic composition (An_{79}) found in the rim. The compositional change is gradual. This reverse-zoning pattern contrasts with the normal-zoning pattern of other large plagioclase grains in the CLS (cores as calcic as An_{97}, rims as sodic as An_{85}) (Fig. 3).

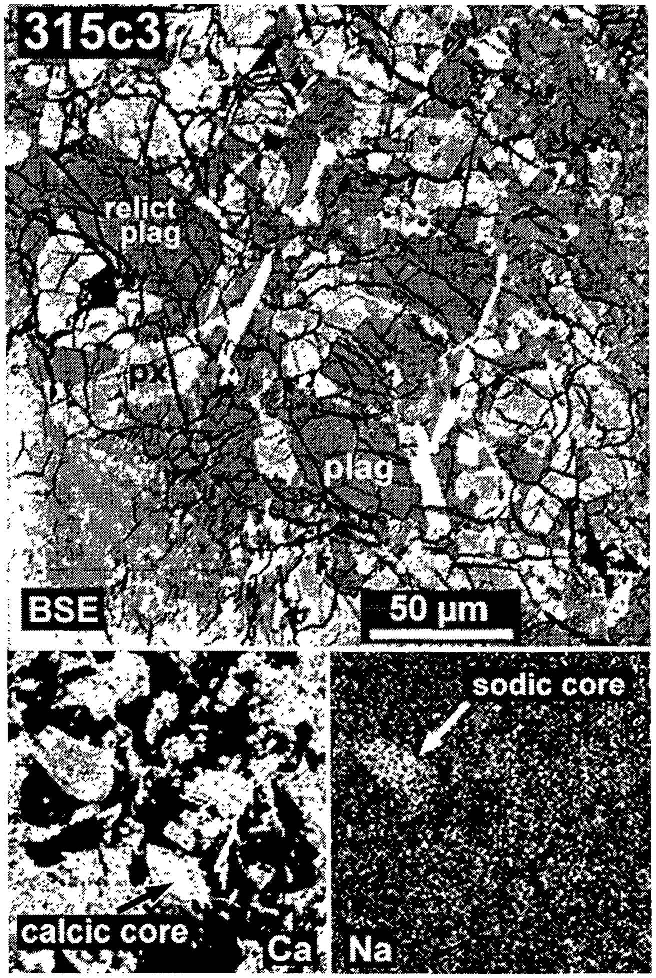

FIG. 3. Images of CLS 315c3 obtained with backscattered electrons (BSE, top), and with $Ca_{K\alpha}$ (lower left) and $Na_{K\alpha}$ (lower right) x-rays. In the BSE image, plagioclase (plag) appears dark grey, pyroxene (px) appears light grey, and ilmenite and phosphate appear white. A relic plagioclase grain in the CLS contains an unusually sodic core and is reversely zoned, unlike other plagioclase grains in the CLS, which have calcic cores.

As previously noted, plagioclase, pyroxene, and olivine grains in CLSs tend to be normally zoned. This is consistent with typical igneous (fractional) crystallization, but the reversely-zoned plagioclase grains in 315c3 and other CLSs cannot be explained this way. Instead, these grains probably represent relic grains that were not fully melted during CLS formation. Their zoning patterns suggest that they did not fully equilibrate with the surrounding melts.

Relic grains were not identified in olivine-rich CLSs. However, Symes *et al.* (1998b) illustrated one olivine-rich CLS with a microporphyritic texture in 65095 that appears to contain two unusually large olivine grains with cores that are more forsteritic (up to Fo_{91}) than that of smaller olivine grains (Fo_{78}) in the object (Symes, pers. comm.). These forsteritic olivine grains may be relic (Symes *et al.*, 1998b), which suggests that relic grains in CLSs can

TABLE 3. Representative phase compositions in crystal-bearing lunar spherules.*

Phase Object	Plag† 315c3	Plag‡ 315c3	Plag 313A	Plag 318c4	Opx 315c3	Pig 68F	Aug 318c10	Ol 318c1	Ol† 313A	Glass 313A	Glass§ 313A	Glass 68F
wt%												
SiO_2	43.4	50.6	42.6	49.5	53.6	51.8	51.1	36.6	41.2	55.1	56.8	44.1
TiO_2	–	–	–	–	0.33	0.84	0.47	1.57	0.09	0.97	1.00	1.43
Al_2O_3	36.0	30.1	35.7	28.8	1.85	2.18	1.91	0.07	–	16.4	16.3	4.04
Cr_2O_3	–	–	–	<0.03	0.54	0.32	0.46	0.17	0.24	0.50	0.60	0.38
FeO	0.26	0.54	0.29	1.24	14.2	18.5	9.74	28.9	4.94	7.16	3.66	24.3
MnO	–	–	–	<0.03	0.16	0.25	0.25	0.32	0.03	0.14	0.15	0.25
MgO	<0.03	0.27	0.18	1.52	26.8	20.5	15.7	32.5	53.0	10.5	8.68	14.1
CaO	19.6	14.3	20.0	15.4	1.81	5.14	18.3	0.40	0.12	10.1	8.32	9.14
Na_2O	0.27	2.72	<0.03	2.09	<0.03	0.03	0.16	<0.03	–	0.70	1.14	0.37
K_2O	0.06	0.69	<0.03	0.22	<0.03	–	0.04	–	–	0.18	2.63	0.54
P_2O_5	–	–	–	–	–	–	–	–	–	0.05	0.09	–
Total	99.6	99.2	98.8	99.1	99.8	99.2	99.2	99.1	99.6	98.8	99.4	98.7
mol%												
An	97.2	71.3	99.9	79.2	–	–	–	–	–	–	–	–
Or	0.3	4.1	0.1	1.4	–	–	–	–	–	–	–	–
En	–	–	–	–	74.1	59.0	45.7	–	–	–	–	–
Wo	–	–	–	–	3.6	10.7	38.4	–	–	–	–	–
Fo	–	–	–	–	–	–	–	66.7	95.0	–	–	–

*Phase abbreviations as in Table 2.
†Core of normally-zoned grain.
‡Core of reversely-zoned grain.
§Na-K-P-rich, near rim of CLS.

exist both as plagioclase and as olivine, the principal phenocryst minerals in these objects.

Sulfide and Phosphate Rims

Some CLSs (68B, 318c9, 315c9, 318c4, 313A) contain thin (≤5 μm thick) rims composed primarily either of troilite and lesser FeNi-metal (Fig. 4), or of phosphate (Fig. 5). The phosphate appears to be merrillite (whitlockite).

Under high magnification, the sulfide rims can be resolved into individual, tiny (≤1 μm) sulfide grains that are attached to, and appear to be perched on, the outer surfaces of the CLSs. Sample CLS 315c9 contains a partial sulfide rim, present on the curved margins of the CLS only and absent along margins that appear to have been brecciated. The phosphate rim around 313A is also clearly restricted to the outermost surface of the spheroid. Individual grains are not visible, but x-ray mapping suggests that the phosphate is intergrown with sulfide. Sulfide rims identical to those present around CLSs also partially coat the margins of some glass spheroids (*e.g.*, 68C, 68D, 315gl). The sulfide rim around glass spheroid 68C is present only where the margin is curved and is absent where the margin is irregular and obviously brecciated. These observations suggest that the rims on CLSs and glass spheroids formed in similar ways.

Radial Gradients in Sodium, Potassium, and Phosphorous

Radial gradients in Na, K, and P are observed within the glassy mesostases of some CLSs (68B, 318c9, 318c4, 313A), with the concentrations of these elements increasing towards the margins of the objects (Figs. 4 and 5; Table 3). The gradients occur only in glass and were not detected in microphenocrysts of plagioclase or in other minerals. Similar near-surface enrichments in Na, K, and P are present within some glass spheroids (68C, 68D, 315gl) and have been previously described in glass spheroids from other lunar samples (Kurat and Keil, 1972). As with the CLSs, the gradients in the glass spheroids are diminished or absent along brecciated margins.

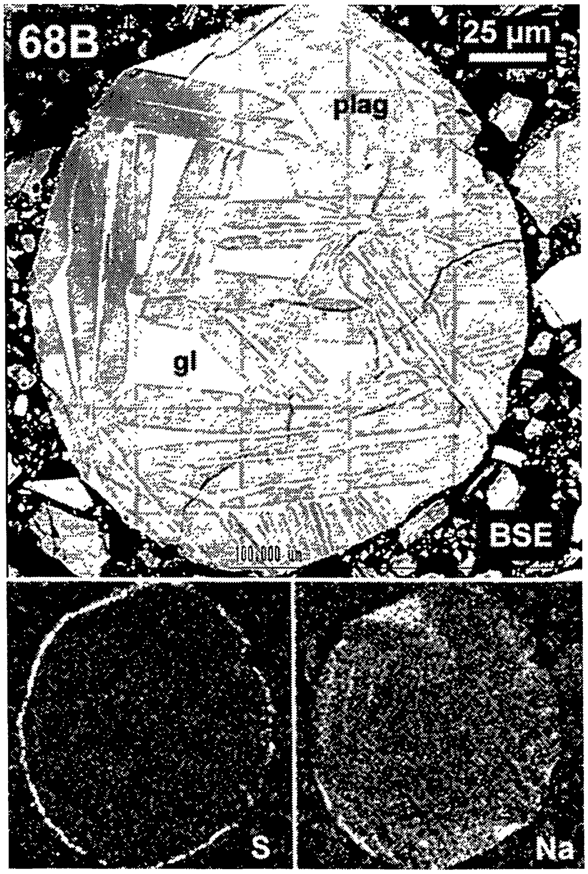

FIG. 4. Images of CLS 68B, obtained with BSE (top), $S_{K\alpha}$ (lower left), and $Na_{K\alpha}$ (lower right) x-rays. In the BSE image, plagioclase (plag) appears dark grey and glass (gl) appears light grey. The CLS is surrounded by a thin rim composed of sulfide and small amounts of FeNi-metal. Glass close to the margins of the CLS is enriched in Na, K, and P and depleted in Ca and Al compared to glass in the center of the object, indicating that 68B was metasomatized.

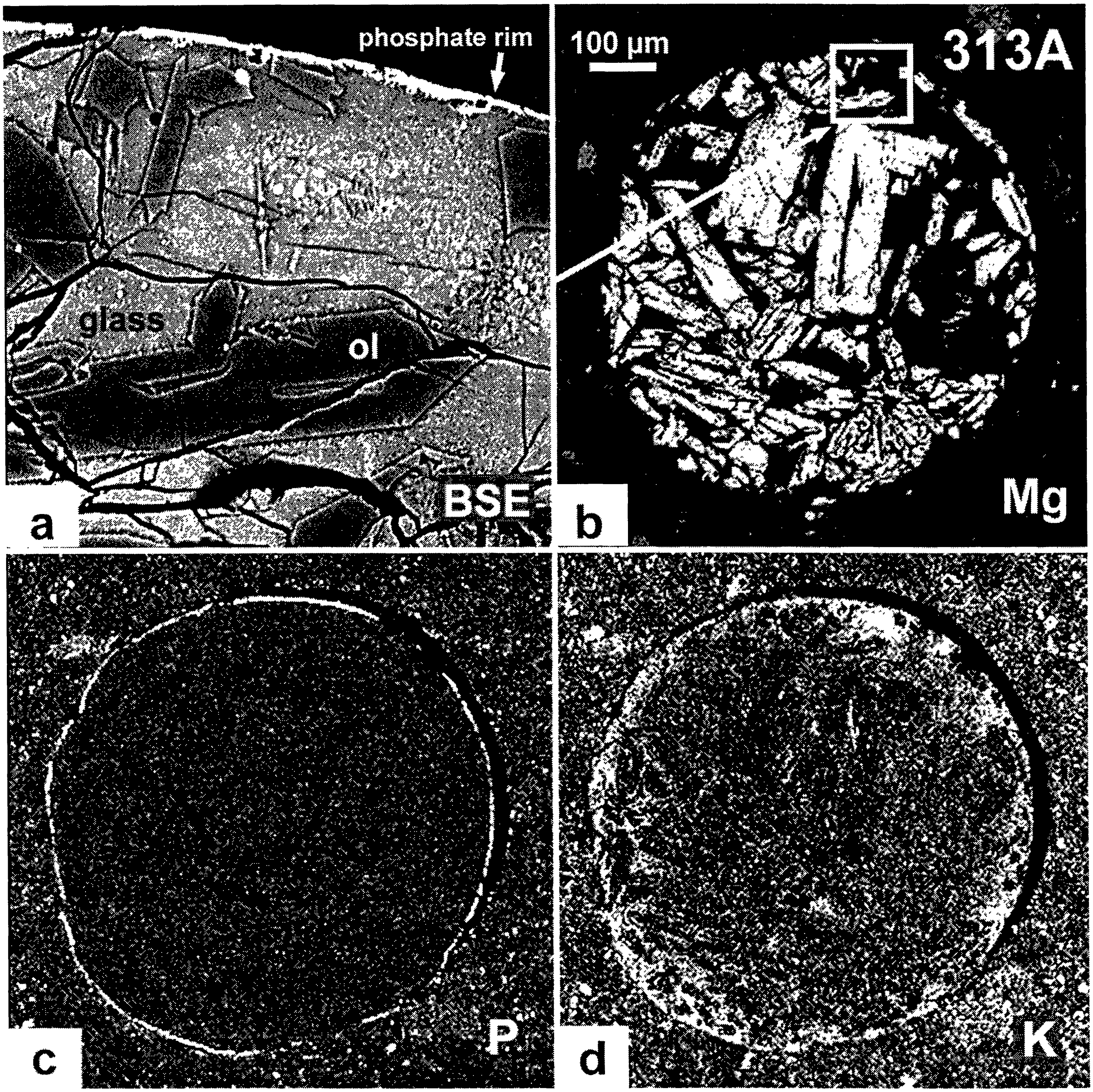

FIG. 5. Images of olivine-rich CLS 313A, obtained with (a) BSE, (b) $Mg_{K\alpha}$ x-rays, (c) $P_{K\alpha}$ x-rays, and (d) $K_{K\alpha}$ x-rays. Parts (b)–(d) have the same field-of-view and show all of 313A, whereas part (a) is a close-up of the area outlined in part (b). Sample 313A consists of olivine grains set in glass. The CLS has a phosphate rim and shows evidence for Na-K metasomatism (enrichments in Na and K in glass close to the margin of the object). In the BSE image (a), olivine (ol) microphenocrysts appear dark grey, and glass (which has devitrified slightly) appears light grey; the phosphate rim and rare FeNi-metal grains inside the CLS appear white.

Compound and Coarsely-Rimmed Objects

Some CLSs are compound objects or have "coarse-grained" rims. Symes *et al.* (1998a) noted that some of the CLSs in 14315 and 14318 are compound objects, and we found additional examples. Figure 6a shows a triple-compound-feldspathic CLS (315c10) in 14315. The textures of the three parts of the object are somewhat different. The largest part is a Type XY spheroid that is mainly granular-textured but which has one prominent plagioclase lath (Fig. 6a). The two smaller parts of 315c10 are less spheroidal; one of these parts is dominated by plagioclase laths and has a Type Y texture; the other part is granular-textured and can be classified as a fine-grained Type X (Fig. 6a).

One CLS in 14318 may be regarded either as a compound or coarsely-rimmed object (Fig. 6b). This CLS consists of a thick (~150 μm wide) crystal-bearing portion (designated 318c5) surround-

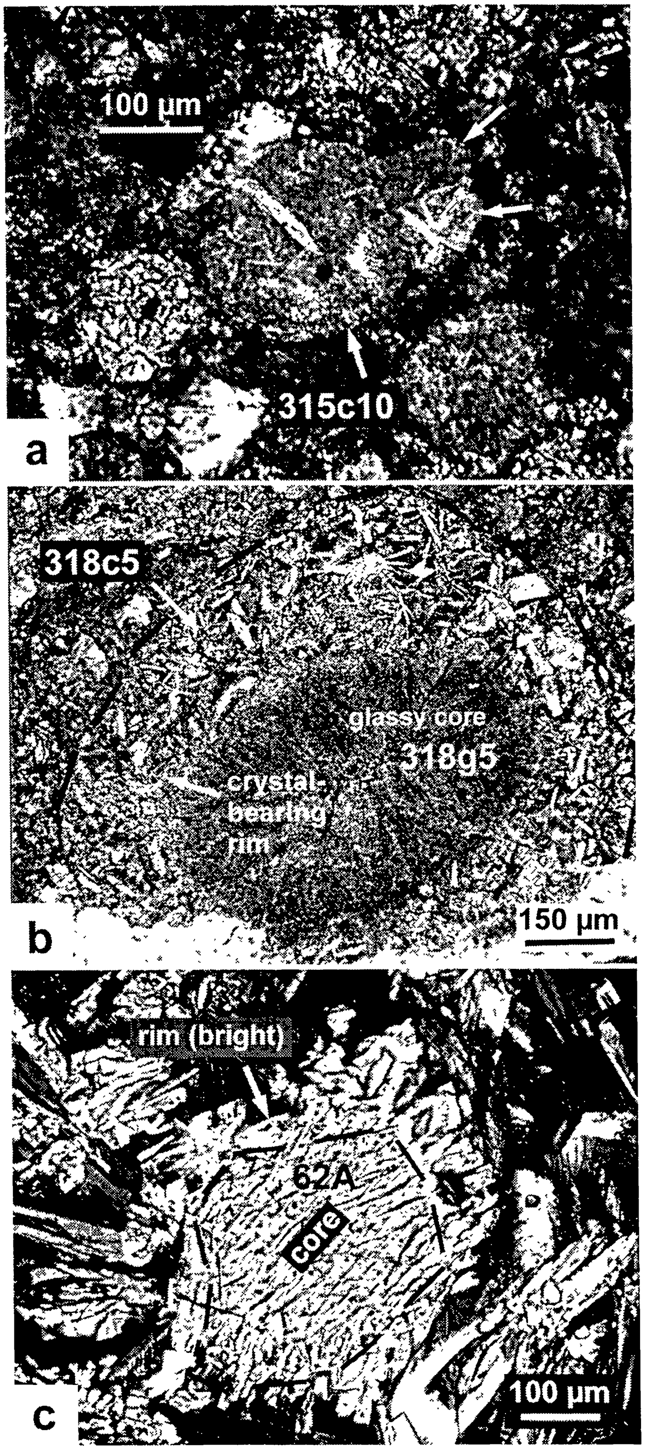

FIG. 6. Micrographs obtained in plane-polarized (a–b) and cross-polarized (c) transmitted light showing compound and coarsely-rimmed CLS objects. (a) Compound feldspathic CLS 315c10 appears to consist of three objects (arrows) stuck together. (b) Compound object 318c5/318g5 consists of a crystal-bearing rim (318c5) surrounding a glassy core (318g5). The rim contains granular plagioclase grains set in a pyroxene-glass mesostasis. The contact between the rim and surrounding host has been outlined to aid in discrimination. The light area at the bottom is the section edge. (c) CLS 62A contains a barred-olivine core and a rim consisting of plagioclase poikilitically enclosing olivine. The host rock has an igneous texture. Plagioclase in the rim appears to have grown away from the barred-olivine core and into the surroundings at top right.

ing a glassy, ellipsoidal core (designated 318g5). The glassy core of the object is fine-grained (<5 μm) and appears to have devitrified, as typical for many glassy objects in 14318 and 14315. The texture and mineralogy of the rim (318c5) is typical of that of an igneous-textured, Type XY feldspathic CLS (Table 1). The largest plagioclase grain is normally zoned (An$_{95-92}$); the second largest grain has weak reverse zoning (An$_{92-93}$).

Another compound object (318c2) in 14318 consists of distinct crystal-bearing and glassy portions. Unlike the object illustrated in Fig. 6b, the crystal-bearing and glassy portions in 318c2 are more randomly distributed.

Sample CLS 62A has a different type of coarse-grained rim (Fig. 6c). As previously noted, the overall texture of 62295 appears to be that of a clast-laden impact-melt, and the barred olivine objects it contains were probably incorporated as xenoliths in the melt-matrix of the host. The core of 62A consists of a single grain of olivine that contains thin (<5 μm), parallel strips of plagioclase or glass, forming a barred-olivine texture. This core is surrounded with a rim composed of plagioclase and olivine, with plagioclase poikilitically enclosing olivine. Most of the plagioclase in the rim has a single crystallographic orientation. Where the rim is thickest (top right, Fig.6c), plagioclase appears to have grown outward into the surrounding host. Other barred-olivine CLSs in 62295 lack similar rims, but at least one other CLS contains plagioclase grains that appear to have nucleated and grown on a barred-olivine core (Roedder and Weiblen, 1977). Most likely, the coarse rim on 62A formed after the core object was incorporated into the melt matrix of the host rock.

DISCUSSION

Lunar Origin for Crystal-bearing Lunar Spherules

The common assumption made by researchers is that CLSs are indigenous to the Moon and do not represent extralunar debris. Evidence for this includes the bulk-chemical and mineral-chemical similarity of some CLSs to other lunar materials (*e.g.*, Kurat *et al.*, 1972a,b, 1974; Keil *et al.*, 1972).

Additional compelling evidence for a lunar origin of CLSs is given by Fig. 7, which compares the bulk FeO and MnO abundances of the CLSs with that observed for lunar samples. All of the CLSs have FeO and MnO abundances that fall within the range observed for lunar materials (Fig. 7). It is well known that the FeO/MnO ratio of planetary materials tends to be relatively constant in the absence of redox variations and that different ratios typify samples from different parent bodies. Lunar samples have a characteristic FeO/MnO weight ratio of 75 ± 5 (Haskin and Warren, 1991). This encompasses the average FeO/MnO value of 73 for CLSs in our study (Fig. 7). In contrast, ordinary and enstatite chondrites have lower FeO/MnO ratios, and carbonaceous chondrites have higher ratios (Jarosewich, 1990), which suggests that the CLSs do not represent chondritic material.

It might be argued that the FeO and MnO abundances and FeO/MnO ratios of CLSs were originally chondritic and were later changed by metasomatism in a lunar environment. However, extensive metasomatism should have obliterated chemical zonation within grains and glasses, contrary to observations (see above). Furthermore, the presence of abundant glass in some objects (Table 2) implies that the objects were not heavily metamorphosed. Instead, the CLSs probably originated by processes indigenous to the Moon.

Incomplete Melting of Precursors

The relic grains observed in some CLSs indicate that these CLSs were not fully melted. One possibility is that the relic grains are

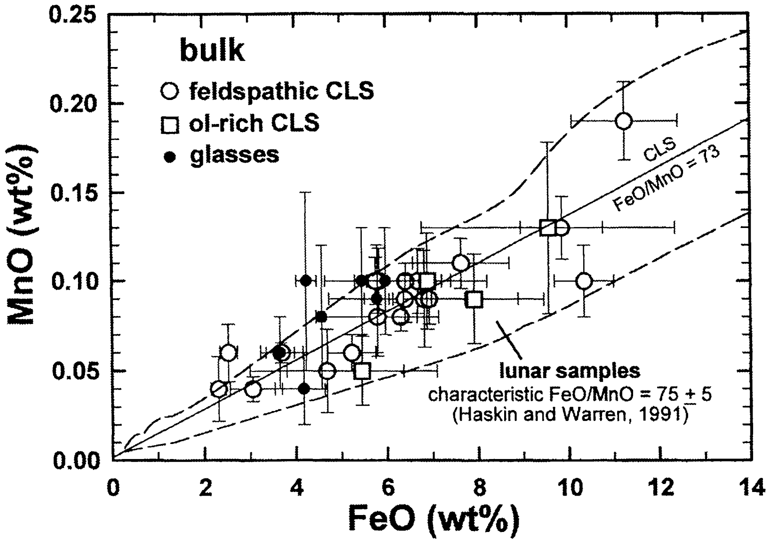

FIG. 7. Bulk FeO and MnO abundances in CLSs fall within the range observed for lunar materials (field outlines based on data given by Haskin and Warren, 1991). The CLSs have an average FeO/MnO weight ratio of 73, indistinguishable from that of lunar materials (75 ± 5; Haskin and Warren, 1991).

xenocrysts that were incorporated into CLS melts during cooling. However, solids that were incorporated into cooling melts would be expected to serve as nucleation sites for other grains in the objects, and no textural evidence for this (*e.g.*, grains that nucleated and grew on preexisting relic grains) is found. More likely, the relic grains represent CLS precursor materials that were incompletely melted during the remelting episode that formed the CLSs. This process appears to be more consistent with the textures of CLSs, as melting would tend to destroy nucleation sites and make them less apparent. Incomplete melting implies either that the precursor materials were heated to subliquidus temperatures, or that heating occurred above the liquidus for brief periods only, allowing the relic grains to persist metastably in the melts.

All four of the CLSs in which relic plagioclase grains were found belong to the Type X (granular) variety, and it is surmised that this variety was incompletely melted. The absence of identifiable relic grains in Type Y (less granular) CLSs could indicate that the nucleation sites in this type of CLS were more effectively destroyed, possibly by more intense heating. These conclusions are consistent with experimental evidence (Symes and Lofgren, 1998) that the textures of Type X CLSs can be simulated by incomplete melting, and that the textures of Type Y CLSs can be reproduced by near-total melting.

Remelting of Highland Lithologies

Bulk-chemical data leave little doubt that materials chemically similar to the lunar highlands were the precursors to most CLSs. Figure 8 shows that the bulk compositions of CLSs more closely resemble monomict breccias from the highlands than mare basalts. This is especially true for feldspathic CLSs, which have FeO and Al_2O_3 abundances and Mg# (Mg/[Mg + Fe^{2+}]) values that fall within the range observed for monomict highland rocks (Fig. 8). Clearly, most feldspathic CLSs were formed by the melting of typical highland lithologies.

Two feldspathic CLSs, 24A and 318c6, lie slightly outside of the monomict highland field (Fig. 8b). In this context, it may be significant that these CLSs contain a higher proportion of Cr-Al spinel than other feldspathic CLSs (Table 2). The bulk compositions of 24A and 318c6 are displaced from the main monomict highland field towards the mare field (Fig. 8), which suggests that they formed by the melting of a mixed highland/mare source, albeit one that was dominated by highland material. In agreement with this possibility, some lunar polymict breccias have compositions (Haskin and Warren, 1991) similar to that of 24A and 318c6.

The olivine-rich CLSs do not chemically resemble typical highland materials as closely as do the feldspathic CLSs (Fig. 8). Although their low FeO abundances are similar to those of highland rocks, the olivine-rich CLSs have alumina abundances (0.5–13 wt% Al_2O_3) that are lower than observed for almost all highland monomict (Fig. 8b) or polymict rocks. Another unusual characteristic of these CLSs is that three (out of four) of them have Mg# values (approximately 0.89–0.94) that are close to the upper limit (0.92) observed in pristine highland samples (Warren, 1993). We will return to the topic of possible lunar precursors and analogues for the olivine-rich CLSs in a later section.

Formation of Crystal-bearing Lunar Spherules by Impact Melting

Constraints on the nature of the melting processes that formed CLSs are obtained by comparing their compositions to that of presumed impact-melt products, such as glasses in samples of the lunar highlands (Table 5, Fig. 9a–c). It is generally accepted that most glass in lunar breccias and soils formed by impact melting of preexisting materials, except for so-called "pristine" volcanic glasses of mare chemical affinity (*e.g.*, Taylor, 1982; Delano, 1986, 1988; Taylor *et al.*, 1991). The two most common (impact-generated) glass compositions found in soil samples are named (somewhat inappropriately) "highland basalt" glass and "Fra Mauro basalt" glass (*e.g.*, Reid *et al.*, 1972; Taylor, 1982).

Most feldspathic CLSs have bulk compositions identical to "highland basalt" glass (Fig. 9, Table 5). This was previously noted for CLSs in 14315 by Dence and Plant (1972) and Vaniman (1990). "Highland basalt" (also known as "anorthositic gabbro") glass is an especially common glass type at the Apollo 16 site (*e.g.*, Naney *et al.*, 1976) and chemically resembles Apollo 16 soils (*e.g.*, McKay *et al.*, 1991). As "highland basalt" glass formed by impact melting (*e.g.*, Taylor, 1982), the close chemical similarity between it and most feldspathic CLSs strongly suggests that the feldspathic CLSs formed by impact melting.

Furthermore, the compositions of feldspathic CLSs and "highland basalt" glass are similar to estimates of average lunar highland crust, as inferred from data based both on Apollo mission samples and lunar meteorites (Table 5) (*e.g.*, Taylor, 1982; Palme *et al.*, 1991). This correspondence reinforces the conclusion, given above, that most feldspathic CLS formed by the remelting of typical lunar highland crustal materials. Indeed, feldspathic CLSs appear to provide a chemically representative sample of the Moon's crust. As the spherules in 14315 and 14318 chemically resemble Apollo 16 soils more closely than they do Apollo 14 soils (*e.g.*, McKay *et al.*, 1991), this implies that they were transported to the Apollo 14 site (*e.g.*, Chou *et al.*, 1972; Symes *et al.*, 1998a) from other, more typical highland terrains.

Although most feldspathic CLSs have compositions corresponding to "highland basalt" glass, two others (315c1 and 315c3) have compositions similar to "Fra Mauro basalt" glass (Fig. 9, Table 5). "Fra Mauro basalt" glass is the most common glass type at the

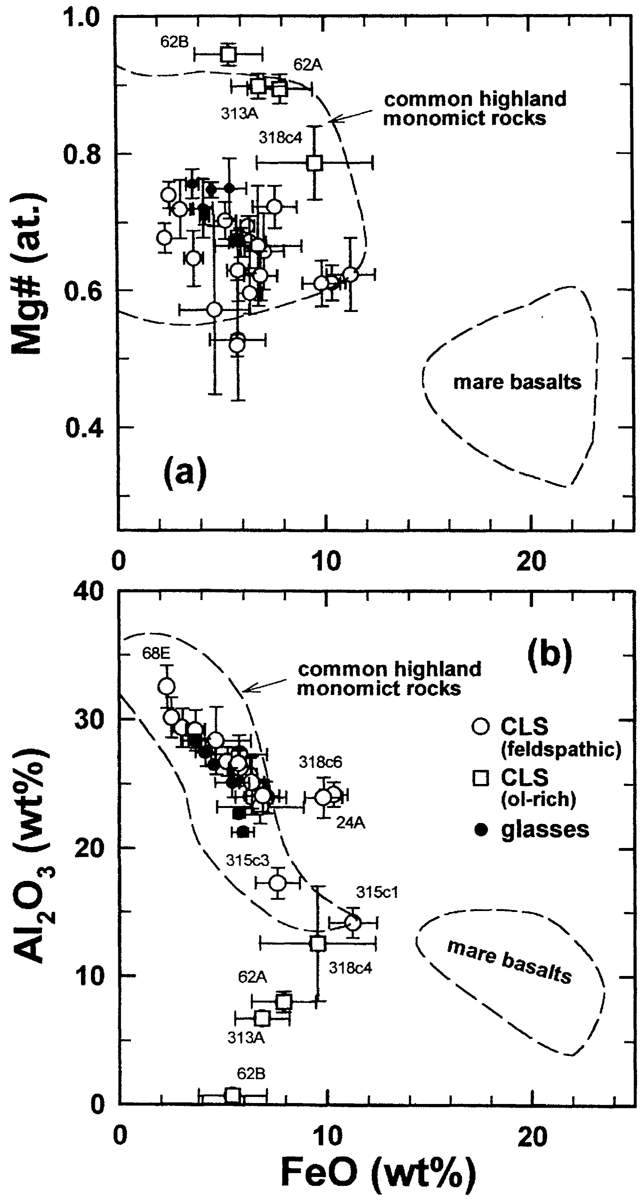

FIG. 8. The bulk abundances of FeO, Al_2O_3, and Mg# (= Mg/[Mg + Fe]) in CLSs more closely resemble that of lunar highland monomict rocks than that of mare basalts (field outlines based on data given by Haskin and Warren, 1991). Feldspathic CLSs have compositions that significantly overlap that of monomict highland rocks.

Apollo 14 site (*e.g.*, Reid *et al.*, 1972; Dence and Plant, 1972; Vaniman, 1990), and it chemically resembles average Apollo 14 soils and most Apollo 14 regolith breccias (*e.g.*, Vaniman, 1990; McKay *et al.*, 1991). This implies that 315c1 and 315c3 were derived by impact melting of the chemically distinctive highland materials that typify the Fra Mauro Formation, on which the Apollo 14 mission landed.

Lack of Vapor Fractionation

The overall chemical similarity of feldspathic CLSs to common glass and soil compositions suggests that the genesis of these CLSs did not involve major vapor fractionation. Although phosphate and sulfide rims around some CLSs imply that some Fe, S, and P were in vapor (see below), less volatile elements such as Si do not appear to have been appreciably vaporized. Evidence that some (mainly small) lunar impact-glasses experienced loss of Si relative to Al to produce high-alumina, low-silica (HASP) glass was presented by Naney *et al.* (1976), Vaniman (1990), and Papike *et al.* (1997). The HASP glasses form distinctive chemical trends in Al–Si plots, with the abundances of Al and Si varying inversely (Fig. 9a–b). The CaO/Al_2O_3 ratios of HASP glasses are relatively unaffected by silica loss (Fig. 9c), as would be expected based on the similar, refractory nature of Ca and Al.

Vaniman (1990) suggested that Apollo 14 HASP, Apollo 16 HASP, and high-Al HASP chemical trends were produced by vapor-fractionation effects operating on typical Apollo 14 soil, typical Apollo 16 soil, and pure anorthosite starting materials, respectively. In Fig. 9, these trends are shown as A, B, and C, respectively. Trace-element data appear to corroborate the vapor-fractionation hypothesis (Papike *et al.*, 1997).

However, the data for CLSs show no convincing evidence for vapor fractionation. Feldspathic CLSs have Al and Si abundances that cluster around "highland basalt" or "Fra Mauro basalt" glass compositions (Fig. 9a–b), and the CLSs that deviate the most from these compositions (24A and 68E) do not follow HASP trends (Fig. 9b). Moreover, even the comparatively volatile element Na has abundances in feldspathic CLSs that are similar to, or higher than, those in their common glass counterparts (Table 5). Similarly, although the four olivine-rich CLSs show chemical variability, they do not show the inverse correlation between Al and Si abundances that would be expected for Si loss (Fig. 9a), or the inverse correlation between Al and Fe abundances that would be expected for Fe loss (Fig. 8b). (The feldspathic CLSs do show an inverse correlation between Al and Fe, but this co-variation is similar to the trend for monomict highland rocks (Fig. 8b) and probably reflects variations in precursor compositions.) Evidently, CLS formation did not result in significant loss of Si, Fe, or Na. This is consistent with the relatively large sizes of the CLSs compared to HASP glasses, as the effects of vapor loss on composition would be expected to increase as melt particles became smaller and their surface-area-to-mass ratios increased (P. Warren, pers. comm.).

Crystallization

The close chemical correspondence between feldspathic CLSs and common lunar glass types (Table 5) has additional significance beyond suggesting that the CLSs formed by impact melting. Namely, the data suggest that compositional effects are not primarily responsible for producing the crystallinity of CLSs. Instead, thermal or physical histories must have been important in determining whether a particular melt spherule would crystallize. This conclusion disagrees with that of Kurat *et al.* (1972b), who did not find glass compositions similar to that of feldspathic CLSs in 14318, and, on this basis, concluded that feldspathic CLSs had compositions that were especially conducive to crystallization. However, devitrified glasses with essentially the same composition as most feldspathic CLSs are present in both 14315 and 14318 (Table 4).

All previous workers who have studied CLSs have concluded that their drop-formed shapes and overall textures are best explained by these objects having crystallized and solidified from melt droplets while in free-flight. This implies long transit times before landing on the surface. Consequently, impacts sufficiently large to loft material high above the surface are needed (*e.g.*, Symes *et al.*, 1998a). Large impacts are also implied by the prevalence of CLSs in highland samples, as the highlands are older and contain a greater number of large craters compared to the maria.

TABLE 4. Bulk compositions (wt%) of crystal-bearing lunar spherules and lunar glasses.

	SiO_2	TiO_2	Al_2O_3	Cr_2O_3	FeO	MnO	MgO	NiO	CaO	Na_2O	K_2O	P_2O_5	Fe	S
Feldspathic crystal-bearing lunar spherules														
24A	41.3	0.11	24.2	0.54	10.3	0.10	9.11	0.04	13.2	0.48	0.02	–	0.43	0.25
68A	46.3	0.14	28.4	0.09	4.68	0.05	3.50	–	16.0	0.65	0.19	0.04	0.02	0.01
68B	43.8	0.23	29.4	0.06	3.06	0.04	4.39	–	16.4	0.73	0.10	0.02	1.14	0.66
68E	43.4	0.09	32.5	0.03	2.31	0.04	2.72	–	18.2	0.35	0.01	–	0.20	0.12
68F	45.3	0.26	29.2	0.09	3.69	0.06	3.79	–	16.6	0.58	0.05	0.03	0.27	0.16
318c1	44.8	0.07	25.7	0.12	6.30	0.08	8.02	0.02	14.2	0.47	0.02	0.03	0.14	0.08
318c5*	44.4	0.08	26.7	0.31	5.22	0.06	6.91	–	14.9	0.41	0.04	–	0.61	0.35
318c6	42.8	0.14	24.0	1.22	9.85	0.13	8.64	–	12.6	0.36	0.04	0.10	0.13	0.07
318c9	45.8	0.25	27.3	0.11	5.79	0.08	3.62	–	16.2	0.50	0.07	0.04	0.13	0.07
318c10	45.5	0.24	26.2	0.43	5.82	0.10	5.53	–	15.4	0.54	0.06	0.05	0.08	0.04
315c1	50.3	0.63	14.2	0.51	11.3	0.19	10.4	–	10.9	0.49	0.52	0.25	0.20	0.12
315c2	45.8	0.11	24.0	0.21	6.67	0.10	8.39	–	13.9	0.42	0.02	–	0.21	0.12
315c3	48.3	1.69	17.0	0.23	7.56	0.11	11.0	–	11.4	0.77	0.66	0.64	0.36	0.21
315c4	46.3	0.18	24.0	0.25	6.40	0.10	7.32	–	14.5	0.33	0.04	0.12	0.31	0.18
315c5	47.6	0.14	23.2	0.16	6.81	0.09	7.59	–	13.6	0.48	0.14	–	0.14	0.08
315c6	47.2	0.25	24.1	0.09	6.92	0.09	6.37	–	14.0	0.49	0.37	0.01	0.08	0.05
315c7	45.7	0.28	26.6	0.22	5.75	0.10	3.48	–	16.6	0.46	0.13	–	0.42	0.24
315c8	44.4	2.09	25.1	0.15	6.39	0.09	5.27	–	15.1	0.43	0.04	0.03	0.58	0.33
315c11	45.6	0.10	30.1	0.13	2.53	0.06	4.03	–	16.7	0.46	0.03	0.04	0.14	0.08
Olivine-rich crystal-bearing lunar spherules														
62A	41.7	0.01	7.99	0.04	7.91	0.09	37.5	0.01	4.52	0.20	0.02	–	–	–
62B	41.9	0.01	0.69	0.12	5.45	0.05	51.2	0.04	0.48	0.02	0.06	–	–	–
318c4	45.2	1.39	12.6	0.34	9.56	0.13	19.7	–	9.03	0.90	0.29	0.13	0.44	0.25
313A	46.4	0.38	6.71	0.36	6.88	0.10	33.8	0.01	4.30	0.40	0.32	0.33	–	–
Crystal-bearing lunar spherules														
Unc.§	1.4–2.0	0.01–0.05	1.0–1.5	0.01–0.07	0.5–1.5	0.01–0.02	0.4–1.0	0.01	0.5–1.0	0.04–0.15	0.01–0.03	0.01	0.01–0.02	0.01
Glasses and devitrified glasses														
68C	45.3	0.30	26.5	0.13	4.57	0.08	7.58	–	15.3	0.25	0.03	–	–	–
68D	44.2	0.30	28.3	0.12	3.66	0.06	6.36	–	16.7	0.24	0.01	–	–	–
318g5‡	46.1	0.11	27.4	0.28	4.17	0.04	6.02	–	15.4	0.38	0.05	–	–	–
315g1	51.1	0.27	21.3	0.26	5.96	0.10	6.79	–	13.2	0.83	0.22	–	–	–
315g2	49.5	0.34	22.7	0.25	5.78	0.09	6.80	–	14.4	0.15	0.03	–	–	–
315g3	45.9	0.22	27.5	0.10	4.23	0.10	5.78	–	16.0	0.16	0.01	–	–	–
315d1†	45.0	0.19	25.1	0.16	5.45	0.10	9.10	–	14.5	0.27	0.12	–	–	–
Glass														
Unc.§	0.2–0.3	0.01–0.03	0.1–1.0	0.02–0.09	0.1–0.8	0.02–0.05	0.1–0.8	–	0.1–0.8	0.03–0.11	0.01–0.15	–	–	–

*Granular rim, surrounding glassy core 318g5.
‡Glassy core, mantled by rim 318c5.
§Typical 1σ uncertainty (wt%) based on phase inhomogeneity and (for crystal-bearing spherules) uncertainty in modes.
†Core region of object.

One often-cited possibility is that CLSs originated in the Imbrium impact (*e.g.*, King *et al.*, 1972a,b; Kurat *et al.*, 1974; Symes *et al.*, 1998a). Assuming that CLSs followed a ballistic trajectory in Imbrium ejecta, Symes *et al.* (1998a) calculated a transit time of ~30 min to reach the Apollo 14 site, which was deemed sufficient to permit crystallization. (Symes *et al.* (1998a) noted that turbulence in the ejecta could lengthen this travel time.) However, glass spheres of similar composition probably also solidified in free-flight, yet they are not appreciably crystalline. Evidently, long transit times are not a sufficient condition for explaining why CLSs are crystalline.

The presence of relic grains in some CLSs suggests incomplete melting of precursor materials. Consequently, CLSs may owe their crystallinity, in part, to the preservation of nuclei in the melts out of which they formed. Nuclei would have been preserved if heating temperatures did not exceed the liquidus for long periods of time. The lack of evidence for vapor fractionation in the CLSs (see above) is consistent with this inference.

Crystallization would also have been aided if cooling rates were relatively slow. Based on comparison with experimental analogues, cooling rates for CLSs have been estimated as approximately 50–2000 °C/h (Symes and Lofgren, 1998). These relatively slow cooling rates imply that the CLSs were warmed by their surroundings, either by a high-temperature gas or by numerous other nearby, radiating objects (*e.g.*, Symes *et al.*, 1998a).

Chemical and Physical Interactions of Crystal-bearing Lunar Spherules with Their Environment

Crystal-bearing lunar spherules experienced physical and chemical interactions with their surroundings prior to their final assembly in the host samples. These interactions included the formation of rimmed and compound objects, and the process that produced radial chemical gradients in some objects.

The radial gradients of Na, K, and P observed in some CLS and glass spheroids are best explained by the diffusion of Na, K, and P into glass or melt in the objects from an external alkali- and P-rich

TABLE 5. Composition (wt%) of feldspathic crystal-bearing spherules compared to common lunar glasses and two estimates for average lunar highlands.

	Average feldspathic CLS* (this study)	Range "highlands basalt" glass†	Lunar highlands‡	Lunar highlands§	Range 315c1 and 315c3 (this study)	Range "Fra Mauro basalt" glass†
SiO_2	45.5 ± 2.0	44.6–46.6	45	–	48.9–50.3	47.5–51.1
TiO_2	0.37 ± 0.54	0.16–0.33	0.56	0.27	0.63–1.71	1.20–2.38
Al_2O_3	25.4 ± 4.2	23.0–29.3	24.6	26.1	14.2–17.2	12.1–17.4
Cr_2O_3	0.26 ± 0.27	0.06–0.14	0.10	0.12	0.24–0.51	0.11–0.27
FeO	6.36 ± 2.60	3.6–6.4	6.6	5.24	7.62–11.3	8.7–15.1
MnO	0.09 ± 0.03	–	–	0.07	0.11–0.19	–
MgO	6.32 ± 2.46	3.4–8.8	6.8	6.14	10.4–11.1	7.4–11.7
CaO	14.8 ± 1.9	14.3–16.9	15.8	15.4	10.9–11.0	9.7–11.6
Na_2O	0.49 ± 0.11	0.0–0.46	0.45	0.33	0.49–0.78	0.48–0.95
K_2O	0.13 ± 0.18	0.01–0.05	0.08	0.02	0.52–0.67	0.43–0.69
P_2O_5	0.11 ± 0.17	–	–	0.02	0.25–0.64	0.2–0.5

*Average (and standard deviation) of 19 feldspathic CLS from Table 4.
†Observed in 1–2 mm fines of sample 14258,34 (Dence and Plant, 1972).
‡Estimated from Apollo orbital x-ray data and from sample-based element-element correlations (Taylor, 1982).
§Average of five lunar highland meteorites (Palme *et al.*, 1991).

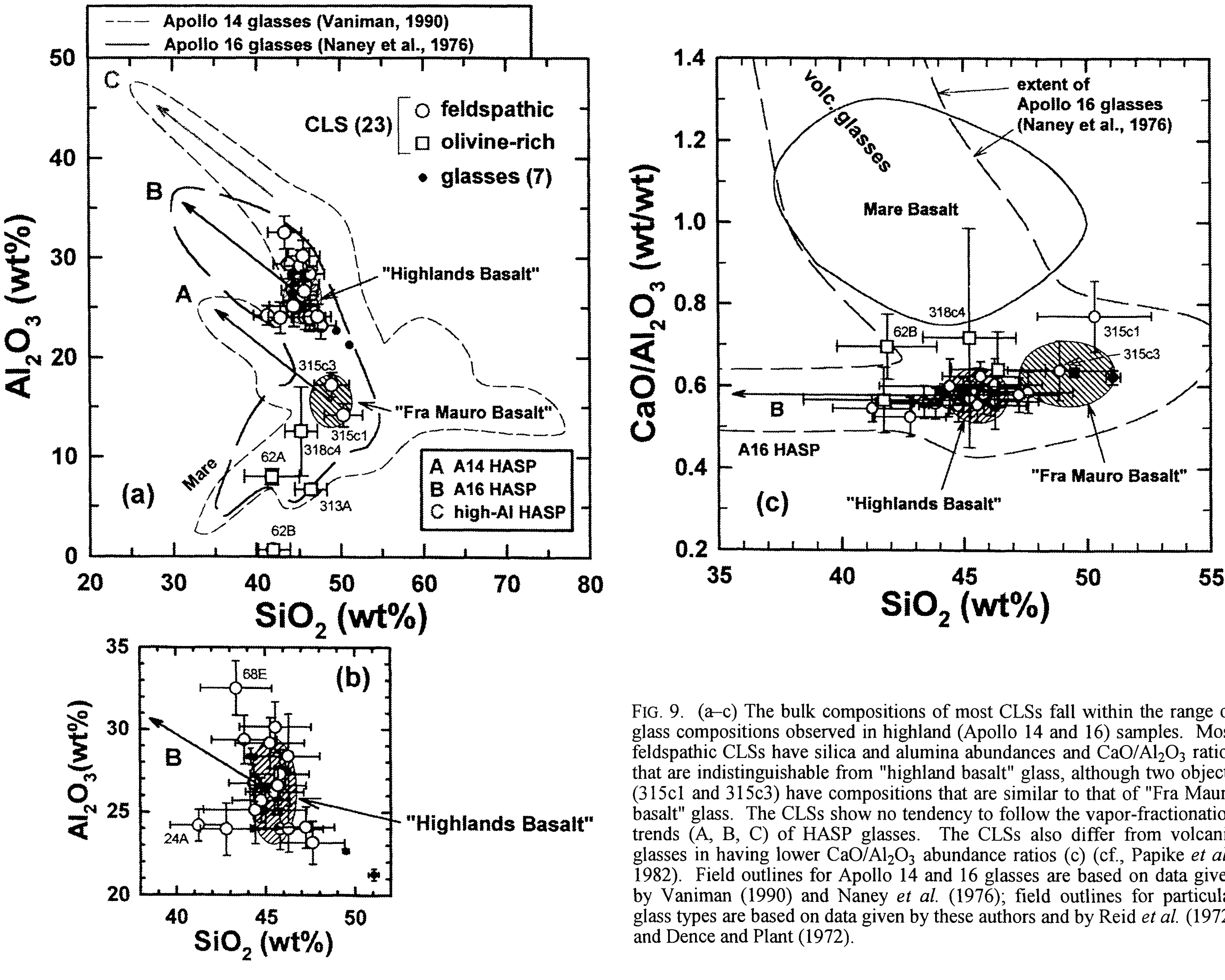

FIG. 9. (a–c) The bulk compositions of most CLSs fall within the range of glass compositions observed in highland (Apollo 14 and 16) samples. Most feldspathic CLSs have silica and alumina abundances and CaO/Al_2O_3 ratios that are indistinguishable from "highland basalt" glass, although two objects (315c1 and 315c3) have compositions that are similar to that of "Fra Mauro basalt" glass. The CLSs show no tendency to follow the vapor-fractionation trends (A, B, C) of HASP glasses. The CLSs also differ from volcanic glasses in having lower CaO/Al_2O_3 abundance ratios (c) (cf., Papike *et al.*, 1982). Field outlines for Apollo 14 and 16 glasses are based on data given by Vaniman (1990) and Naney *et al.* (1976); field outlines for particular glass types are based on data given by these authors and by Reid *et al.* (1972) and Dence and Plant (1972).

source. This constitutes evidence that the objects were metasomatized in a volatile-rich medium. Metasomatism clearly occurred prior to brecciation, as broken margins of the spheroids lack volatile-element enrichments. Metasomatism may have occurred while the objects were suspended in a gas phase within an impact-ejecta plume. Alternatively, chemical exchange could have occurred in a regolith environment, after these objects had solidified, but while they were sufficiently warm to allow significant diffusional exchange.

The sulfide and phosphate rims on various CLSs and glass spheroids can be explained by the accretion or vapor-phase condensation of tiny grains on the surfaces of preexisting melt droplets. An alternative explanation, that the rims formed by the migration of sulfide and phosphate from within the objects to their surfaces, would require complete separation of the phases from within the objects, as sulfide and phosphate are rarely found within the interiors of the objects. Such complete separation seems unlikely. In any case, as with metasomatism, the absence of rims along irregular margins of the objects implies that rimming occurred before brecciation. Moreover, as for metasomatism, rimming involved relatively volatile elements (S, Fe^{2+}, and P). These similarities suggest that rimming and metasomatism were coeval and involved interaction of the CLSs and glass spheroids with an external, volatile-bearing environment, which could have been an impact-generated gas plume (Housley, 1979).

Compound objects are best explained as having formed by the attachment of droplets that were free-floating. In the case of 315c10 (Fig. 6a), three independently-formed melt droplets became attached. At the time of attachment, it appears that the main portion of 315c10 had largely solidified, as evidenced by its near-perfect droplet morphology (Fig. 6a). In contrast, the two smaller parts of the object may have been plastic and easily deformable, as suggested by their less regular shapes (Fig. 6a). They may have been partly molten, or at least warm and still deformable. In any case, for three droplets to have become attached implies that they formed in an environment that had numerous, closely-spaced melt droplets.

Compound object 318g5/c5 (Fig. 6b), with its glassy core and crystal-bearing rim, provides perhaps the most striking evidence of interactions between objects that had solidified to differing degrees. The glassy core appears to have been largely solid at the time the rim formed, based on its ellipticity, the fairly sharp boundary with the rim, and the absence of crystals that could have penetrated from the rim into the core had the latter been molten. The rim could have been molten or semi-molten, as suggested by its apparently igneous (Type XY) texture. In this case, the object may have formed by the accretion of a crystal-laden melt (the rim) onto a preexisting glass spheroid (the core). The core (318g5) and rim (318c5) have similar bulk compositions (Table 4), implying that they were derived from similar sources. Conceivably, both the core and rim formed in the same melting event, and the rim (crystal-laden melt) overtook and accreted onto the core (a largely solidified melt) as a result of turbulence in an impact-plume. Alternatively, 318g5/c5 may have formed after a glassy object (represented by the core) was coated with coarser, solid debris (the rim) in a regolith, but it is unclear whether the texture of the rim (which appears "igneous") is compatible with this origin.

Unlike CLS 318c5/g5, compound object 318c2 has a more random distribution of crystal-bearing and crystal-poor portions, and a less-regular shape. It may have formed by the coalescence and incomplete assimilation of crystal-bearing and crystal-poor melts.

It is evident that the collisions between melt droplets that produced compound objects occurred under a variety of circumstances, resulting in different textural associations. Some collisions occurred between largely soldified objects (315c10); others involved largely molten objects (318c2); and still others (318c5/g5) may have involved the accretion of melt onto a largely solidified core.

Compound objects probably formed in an ejecta plume. The alternative, that they formed after landing in a regolith or base-surge deposit, seems unlikely, as this process would be expected to produce either agglutinatic (*e.g.*, vesiculated) or brecciated material. No agglutinatic material attached to CLSs was found, and brecciation would tend to destroy compound objects, not produce them.

Regardless of the precise environment in which compound objects formed, this environment appears to have been turbulent, allowing melt droplets in various states of solidification to collide. Moreover, for the compound objects to have become attached, low-velocity collisions and some "stickiness" (perhaps caused by annealing between warm objects) are required.

Altogether, the data suggest that CLSs experienced various physical and chemical interactions in a turbulent impact-ejecta plume, in some cases involving low-velocity collisions between numerous, closely-spaced melt droplets, and in other cases becoming rimmed or metasomatized by interaction with the surrounding vapor phase. A finite amount of time was needed to accomplish these various physical and chemical interactions; they could not all have occurred instantaneously. This is consistent with the need for relatively long ejecta transit times to enable crystallization to occur in the CLSs (see above).

Analogues and Precursors to Olivine-rich Crystal-bearing Lunar Spherules

In contrast to feldspathic CLSs, the compositions of some olivine-rich CLSs do not correspond to common rock types or impact-produced glasses. It was noted above that these objects are FeO-poor, high in Mg#, and more closely resemble the compositions of common highland monomict rocks than mare basalts, but that the match to highland materials was not especially good. Especially noteworthy is the high Mg# values (approximately 0.89–0.94) for olivine-rich CLSs 62B, 62A, and 313A (Table 4, Fig. 8a). Highland rocks with Mg# values as high are rare (Haskin and Warren, 1991). For example, considering "pristine" highland rocks, only 11 samples (mainly troctolites) among 260 are listed in Warren's (1993) compilation as having Mg# ≥ 0.89, and only 2 of these samples have Mg# > 0.90. None have Mg# as high as the most magnesian CLS (0.94 ± 0.01).

Possible explanations for the distinctive compositions of the olivine-rich CLSs include (1) nonrepresentative sampling, (2) reduction of FeO to produce high-Mg# objects, (3) volatility-related loss of FeO to form high-Mg# objects, and (4) derivation from distinctive or otherwise unsampled lunar sources. It might be argued that the bulk compositions of the olivine-rich CLSs reflect nonrepresentative sampling of olivine-bearing highland precursor rocks, with the CLSs preferentially sampling olivine. This is a distinct possibility. However, high Mg# values are difficult to explain by non-representative sampling, as both Mg and Fe should largely be contained in olivine and pyroxene in reasonable precursor materials, and as Mg and Fe do not fractionate strongly between these phases. Alternatively, high Mg# could be caused by the reduction of FeO to Fe-metal, but this process would tend to produce anomalously low bulk FeO/MnO ratios in these objects, for which there is no evidence (Fig. 7). Another possibility is that the high-Mg# values for the olivine-rich CLSs were produced by volatility-related loss of FeO. However, this process would be expected to result in an inverse correlation between FeO and Al_2O_3 abundances, opposite to the observed trend for the olivine-rich CLSs (Fig. 8). Thus, it appears that the high values of Mg# and low FeO abundances for some olivine-rich CLS reflect their derivation from relatively magnesian precursors.

Although some magnesian lunar glasses with appropriately high Mg# exist, these too appear to be unsuitable precursors for the high-Mg# olivine-rich CLSs. Picritic volcanic glasses of mare affinity have values of CaO/Al_2O_3 (>1 wt/wt) that are higher than observed for the olivine-rich CLSs (<1) (Fig. 9c). Forming the olivine-rich CLSs from sources similar to that which produced the picritic volcanic glasses would require loss of clinopyroxene relative to olivine and plagioclase components, which seems *ad hoc* and unlikely. Similarly, the rare high-Mg# (>0.90) "komatiitic" glasses of highlands affinity found in Apollo 16 soils (Wentworth and McKay, 1988; Shearer *et al.*, 1990) have alumina abundances (approximately 12–23 wt% Al_2O_3; Shearer *et al.*, 1990) that are higher than in three of the four olivine-rich CLSs (<8 wt%) (Fig. 8b). If the olivine-rich CLSs were derived from such a "highland komatiitic glass" source, preferential loss of plagioclase relative to an olivine component would be required. This is unlikely for CLS 313A, which is a relatively large (925 μm) object compared to the "komatiitic" glasses (<120 μm; Shearer *et al.*, 1990). Thus, an appropriate lunar source region for the three high-Mg# olivine-rich CLSs (62A, 62B, 313A) remains elusive.

Compared to other olivine-rich CLSs, the fourth object (318c4) has lower Mg# and higher alumina contents, which fall within the range of monomict and polymict highland rocks (Fig. 8; Haskin and Warren, 1991). A possible analogue to CLS 318c4 is olivine vitrophyre in lunar breccia 14321, described by Allen *et al.* (1979) and Shervais *et al.* (1988) (Table 6). The olivine vitrophyre is present as large (up to 6.5 × 4.5 cm) clasts and is believed to have formed by impact melting of an Mg-rich precursor (Allen *et al.*, 1979; Shervais *et al.*, 1988). The compositions of olivine vitrophyre in 14321 and CLS 318c4 are similar (Table 6), implying that they could have formed by impact melting of similar precursors.

TABLE 6. Composition (wt%) of CLS 318c4 compared to average olivine vitrophyre (AOV) clasts in lunar breccia 14321.

	CLS 318c4[*]	AOV[†]	AOV[‡]
SiO_2	45.2 ± 1.9	45.7	46.5
TiO_2	1.39 ± 0.9	1.03	1.30
Al_2O_3	12.6 ± 4.5	10.3	12.4
Cr_2O_3	0.34 ± 0.21	0.17	0.23
FeO	9.56 ± 2.79	10.8	9.86
MnO	0.13 ± 0.05	0.12	0.13
MgO	19.7 ± 2.5	24.3	19.2
CaO	9.03 ± 0.99	6.27	7.9
Na_2O	0.90 ± 0.40	0.78	0.80
K_2O	0.29 ± 0.09	0.46	0.51
P_2O_5	0.13 ± 0.01	–	–

[*] This study, Table 4.
[†] Average of four Mg-rich olivine vitrophyre clasts (Allen *et al.*, 1979).
[‡] Preferred average olivine vitrophyre composition of Shervais *et al.* (1988).

Precursors: Crustal Sources and Lithologic Mixing

Chemical evidence suggests that the CLSs sampled at least three source regions. These include: (1) common feldspathic CLSs, derived from the same source as "highlands basalt" glass; (2) feldspathic CLSs 315c1 and 315c3, derived from the same source as "Fra Mauro basalt" glass; and (3) olivine-rich CLSs, derived from one or more Mg-rich source regions atypical of the lunar crust. Source region (1) resembles the average regolith at the Apollo 16 site, and source region (2) resembles the average regolith of the Apollo 14 site. This chemical similarity to regoliths suggests that the feldspathic CLSs were derived from lithologically-mixed sources. Moreover, as previously noted, the bulk compositions of two feldspathic CLSs (24A, 318c6) can be explained if they incorporated an admixture of a mare component to a predominantly highland lithology.

Mineral-chemical data provide additional evidence for the nature of the CLS source regions. The current paradigm (*e.g.*, Taylor *et al.*, 1991; Warren, 1993; Papike *et al.*, 1998) is that the Moon's crust originally contained plutonic rocks consisting mainly of anorthosites, norites, troctolites, and dunites, identifiable by their coarse textures (signifying a "plutonic" setting) and by their low abundances of siderophile trace elements (signifying minimal meteoritic contamination). Based on mineral-chemical criteria, these "pristine" rocks can be classified into two types, including (1) ferroan anorthosites (FANs), and (2) norites, troctolites, and dunites (rocks of the Mg-suite). An incompatible-element-enriched (KREEP) component, identified mainly by trace-element signatures, is a third important component of the Moon's crust (Taylor *et al.*, 1991).

Figure 10 shows that feldspathic CLSs have plagioclase An contents and olivine and pyroxene Mg# values that cluster between the compositions observed in FANs and Mg-suite rocks. Some

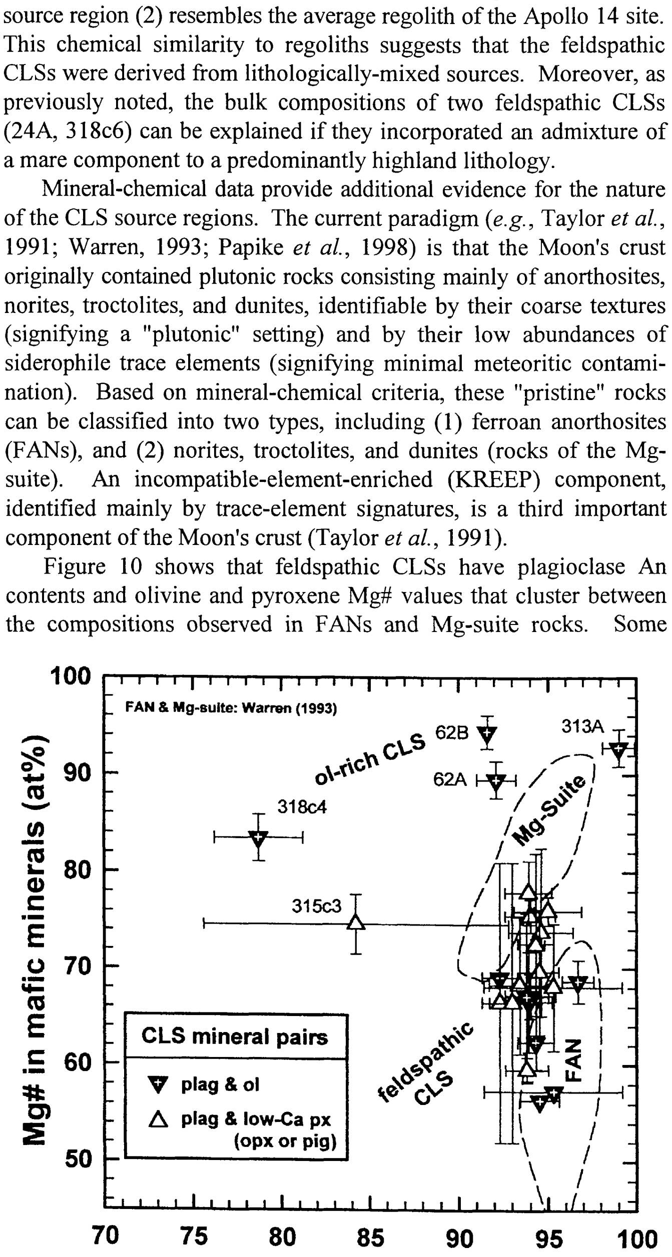

FIG. 10. Most feldspathic CLSs have mineral compositions that lie between that of ferroan anorthosites (FANs) and Mg-suite rocks, implying that they could have formed from precursors that were mixtures of these rock types. In contrast, olivine-rich CLSs appear to have been derived from more magnesian precursors. Crystal-bearing-lunar spherule compositions represent averages and associated standard deviations for each object. Field outlines for FANs and Mg-suite rocks are from Warren (1993). Abbreviations: An = Ca/(Ca + Na + K) in plagioclase; Mg# = Mg/(Mg + Fe) in olivine or low-Ca pyroxene (orthopyroxene or pigeonite).

overlap is present, but the mean compositions for most CLSs clearly lie between the two fields (Fig. 10). This implies that the source materials of most feldspathic CLSs were mixtures of the original, principal rock types in the Moon's crust, namely FANs and Mg-suite rocks. The feldspathic CLSs do not have mineral compositions strongly biased towards either the FAN or Mg-suite fields (Fig. 10). This could be explained if the feldspathic CLS precursors consisted, on average, of roughly equal amounts of FAN and Mg-suite rocks.

As the major element compositions of common feldspathic CLSs are similar to estimates of the Moon's crust (Table 5), the implication is that the lunar highlands are composed of approximately similar amounts of FAN and Mg-suite rocks. This is consistent with the observation that roughly the same number of samples of pristine FANs and Mg-suite rocks have been found (Taylor *et al.*, 1991; Warren and Kallemeyn, 1993; Warren, 1993), and with estimates (based on Al mass balance) that the lunar highlands contain about 70% FAN and 30% non-FAN rocks (Warren and Kallemeyn, 1993). However, a higher proportion of FAN has been advocated by Tompkins and Peters (1999), based on Clementine spectral data for fresh craters on the Moon.

Other CLS objects sampled different materials. Feldspathic CLS 315c3 contains both relic sodic and normal calcic plagioclase, accounting for the large dispersion in its plagioclase composition (Fig. 10). Based on the presence in 315c3 of such phases as K-Ba-rich glass, ilmenite, phosphate, and baddeleyite, the precursor to this object appears to have been more chemically evolved than for other CLSs. It probably incorporated a significant KREEP component.

Unlike the common feldspathic CLSs, the olivine-rich CLSs (313A, 62B, 62B, 318c4) have mineral compositions that are not intermediate to Mg-suite and FAN-type rocks (Fig. 10). Instead, the olivine-rich CLSs appear to form an array displaced towards relatively high Mg# (Fig. 10). This supports the conclusion, based on bulk-chemical data, that the olivine-rich CLSs were derived from distinctive, magnesian sources (the lunar mantle?), atypical of the lunar crust.

Rarity of Crystal-bearing Lunar Spherules

It is clear that CLSs are relatively rare in lunar samples, compared to other impact-melt products, such as clast-rich and -poor melt rocks, glassy spheroids, and agglutinates (*e.g.*, Taylor *et al.*, 1991; McKay *et al.*, 1991; Papike *et al.*, 1998). This paucity could be explained either by inefficient production or preferential destruction of CLSs compared to other impact-melt products on the Moon.

Although CLSs could have been subjected to a long period of destruction by regolith processes on the Moon, it seems unlikely that this can explain the relative dearth of these objects. As previously discussed, CLS formation was probably restricted to basin-scale impacts (Symes *et al.*, 1998a) prior to 3.8 Ga ago (*e.g.*, Wilhelms, 1987). This is in contrast to impact-melt rocks, impact-glass spheroids, and agglutinates, which could have continued to form in smaller-scale impacts throughout lunar history. However, many impact-melt rocks with old ages (>3.8 Ga) have been analyzed (*e.g.*, Wilhelms, 1987; Taylor *et al.*, 1991; Papike *et al.*, 1998), and these, like the CLSs, would have been susceptible to destruction by later impacts. The existence of numerous, old impact-melt rocks implies that early-formed impact melts were not completely destroyed on the Moon. Similarly, many impact-glass spherules were probably produced early, yet they outnumber CLSs in lunar samples. Indeed, in breccias 14315 and 14318, CLSs and glassy spherules may have formed in the same impact event and could have similar brecciation histories, yet the glassy spherules are more common.

Altogether, the data imply that CLSs were not significantly destroyed by regolith processes compared to impact-melt rocks and impact-glass spherules. Instead, it appears that CLS formation was a relatively inefficient process and that unusual conditions on the Moon were needed to produce these objects.

As noted above, thermal or physical histories must have been important in determining whether a particular melt droplet in an ejecta plume would crystallize to form a CLS or solidify to form a glass spherule. Two conditions that would promote crystallization of the droplets include (1) incomplete destruction of nuclei in the melt, to provide nucleation sites for crystals; and (2) subsequent slow cooling, to allow crystals to grow larger. Incomplete destruction of nuclei could be explained if the time spent above the liquidus was relatively short, as would be expected for melt produced by hypervelocity impact processes. In contrast, relatively slow cooling of melt-droplet ejecta in free-flight may have been uncommon on the Moon. Melt ejecta would tend to cool rapidly, unless it was buried in regolith and no longer in free-flight, or unless it was thermally insulated by hot vapor, or by nearby warm solids or melted debris in the ejecta curtain. The CLSs may have formed in localized regions of ejecta plumes that had an unusually high vapor or melt content. Slow cooling may have required large impact events to produce a sufficient quantity of hot vapor or melt in the ejecta curtain. Coupled with the need to keep the melt ejecta suspended above the surface for unusually long durations, this could explain the dearth of CLSs compared to other impact-melt products on the Moon.

IMPLICATIONS FOR METEORITIC CHONDRULES

It has long been noted that CLSs have important implications for the formation of meteoritic chondrules. The argument has been that if CLSs formed by impact melting, as seems certain, chondrules may have as well. There would be little argument with this proposition if chondrules, CLSs, and their host rocks were identical in every way, but they are obviously not, and the question of whether chondrules could have formed by impact-melting requires careful evaluation. To this end, Table 7 summarizes CLS and chondrule characteristics. Discussion is first made of the properties listed in Table 7 and then of the broader implications of the data.

Properties

Textures and Zoning Patterns–Both chondrules and CLSs most commonly have microporphyritic or granular textures; barred-olivine textures are also present in both. Zoning patterns within phenocrysts (in weakly metamorphosed samples) are consistent with igneous (fractional) crystallization. These similarities testify to the igneous origin of the objects.

Sizes–The overall sizes of chondrules and CLSs are similar. Furthermore, the size-frequency distribution of CLSs in 14315 and 14318 are similar to the size-frequency distribution of chondrules in some chondrite types (Symes *et al.*, 1998a). This implies either that chondrule and CLS formation resulted in melt objects of similar size, or that similar size-sorting processes affected the objects after they had formed.

Shapes and Mineralogies–Some CLSs and chondrules have drop-formed shapes, suggesting that they crystallized from melts while they were free-floating objects. Other objects have more irregular shapes, possibly indicative of collisional (*i.e.*, brecciation) processes. The mineralogies of CLSs are dominated by Ca-rich plagioclase, whereas those of most chondrules are dominated by either olivine or low-Ca pyroxene. The difference in mineralogy

TABLE 7. Comparison of characteristics for crystal-bearing lunar spherules and chondrules.

	Characteristic	Crystal-bearing spherules	Chondrules
1.	Textures	mainly microporphyritic/granular; also barred olivine	mainly microporphyritic/granular; also barred olivine, radial pyroxene, cryptocrystalline [1–3]
2.	Diameters	approximately 0.1–0.9 mm	approximately 0.2–1 mm [2]
3.	Shapes	drop-formed and irregular	drop-formed and irregular [4]
4.	Mineralogy	plagioclase- or olivine-rich	olivine- or pyroxene-rich [1, 2]
5.	Relic grains	plagioclase; also olivine [5]	olivine [2, 6, 7, 9]; also low-Ca pyroxene [8, 9]
6.	Heating	short; subliquidus to superliquidus [10]; minimal vaporization; no FeO reduction	short [4, 26]; subliquidus to superliquidus [3, 4, 26]; partial vaporization of Na, Fe [11–13]; FeO reduction [6–8, 12, 13]; repeated melting [14, 17, 18]
7.	Cooling	approximately 50–2000 °C/h [10]	approximately 10–2000 °C/h [3, 25, 26]
8.	Compound objects	present	present [14]
9.	Rims	thin (≤ 5 μm); mainly fine-grained sulfide; no layering; coarse-grained rims absent or rare	thick (*e.g.*, 5–40 μm in UOC), fine-grained sulfide and silicate, sometimes layered [15, 16]; coarse-grained rims present [14, 17, 18]
10.	Metasomatism	minor Na, K, P influx	Na influx [19–22], possibly major [19]
11.	Precursors	at least three differentiated sources on the Moon	olivine- and pyroxene-rich [1, 2, 23]; probably vapor fractionated with diverse Al/Mg/Si [2, 11, 23]
12.	Abundance	low but found in many highland breccias; up to ~8% in 14315/18 [24]	0 to ~75% in chondrites [2]
13.	Ages	unknown	close to 4.55 Ga [27]

References: [1] Dodd (1981); [2] Grossman *et al.* (1988); [3] Hewins (1988); [4] Hewins (1998); [5] Symes *et al.* (1998b); [6] Nagahara (1981); [7] Rambaldi (1981); [8] Rambaldi *et al.* (1983); [9] Jones (1996); [10] Symes and Lofgren (1998); [11] Hewins *et al.* (1997); [12] Huang *et al.* (1996); [13] Sears *et al.* (1996c); [14] Wasson *et al.* (1995); [15] Allen *et al.* (1980); [16] Alexander *et al.* (1989); [17] Rubin (1984); [18] Krot and Wasson (1995); [19] Grossman (1996); [20] Ikeda and Kimura (1985); [21] DeHart *et al.* (1988); [22] Matsunami *et al.* (1993); [23] Grossman (1988); [24] Symes *et al.* (1998a); [25] Weinbruch *et al.* (1998); [26] Lofgren (1996); [27] Brearley and Jones (1998).

almost certainly reflects a more felsic and less mafic or ultramafic composition for the precursors of CLSs.

Relic Grains–Some chondrules and CLSs contain relic grains. These grains do not appear to have crystallized from the melts that solidified to form the remainder of the objects. Most likely, this indicates that some chondrules and CLSs were incompletely molten and were formed by the melting of precursors that were not exclusively fine-grained. Relic grains consist mainly of olivine in chondrules and of plagioclase in CLSs, probably owing to differences in precursor composition (see above).

Heating Conditions–Experimental work suggests that the textures and zoning patterns of chondrules and CLSs were produced by heating mainly to near-liquidus or subliquidus temperatures (*e.g.*, Hewins, 1988; Lofgren, 1996; Symes and Lofgren, 1998). The heating duration was probably "short" for chondrules (Hewins, 1989, 1997) and for CLSs. Crystal-bearing lunar spherules show no evidence for FeO reduction during heating, contrary to the situation for some chondrules (*e.g.*, Huang *et al.*, 1996; Sears *et al.*, 1996c; Rambaldi, 1981; Rambaldi *et al.*, 1983). This may indicate that chondrules were exposed to one or more reducing agents (H_2 gas? graphite?) during heating and that these agents was less prevalent or absent during CLS formation. Vapor fractionation appears to have been important in affecting the compositions of chondrules (*e.g.*, Grossman *et al.*, 1988; Grossman, 1988; Sears *et al.*, 1996c; Huang *et al.*, 1996; Alexander, 1996; Hewins *et al.*, 1997), but the same is not true for CLSs. However, regarding chondrules, it is not entirely clear to what extent the precursors to chondrules were vapor fractionated, and to what extent chondrule formation involved vapor fractionation.

Cooling Conditions–The inferred cooling rates for chondrules (about 10–2000 °C/h) and CLSs (about 50–2000 °C/h) are essentially identical and relatively low, implying that the objects were warmed by their surroundings as they crystallized (*e.g.*, Hewins, 1988; Weinbruch *et al.*, 1998; Symes and Lofgren, 1998).

Compound Objects–Some chondrules (*e.g.*, Wasson *et al.*, 1995) and CLSs are compound objects. These include objects that appear to have coalesced as a result of low-velocity, "sticky" collisions, and melts that appear to have enveloped preexisting objects. High densities of melt objects in the source regions for CLSs and chondrules are implied.

Rims–Both types of objects are sometimes surrounded by fine-grained rims, with troilite being an important constituent in most such rims. However, the fine-grained troilite and phosphate rims around CLSs are thinner (≤ 5 μm) and less complex than those around chondrules. For example, rims around chondrules in ordinary chondrites are typically 5–40 μm thick and consist of a mixture of troilite and silicates (mainly ferrous olivine) that are often complexly layered (*e.g.*, Allen *et al.*, 1980; Matsunami, 1984; Alexander *et al.*, 1989). "Coarse-grained rims" of the type found around some chondrules (Rubin, 1984; Krot and Wasson, 1995) are generally absent around CLSs, although lunar object 318c5/g5 (Fig. 6b) may be an exception. The "coarse-grained rims" around chondrules were interpreted to be fine-grained rims that were flash-remelted (Rubin, 1984; Krot and Wasson, 1995; Wasson *et al.*, 1995), which suggests that chondrules experienced multiple, brief melting events (see above). Possible reasons for differences in rims between CLSs and chondrules are discussed below.

Metasomatism–Radial gradients in Na are sometimes observed in chondrule mesostasis glasses (*e.g.*, Ikeda and Kimura, 1985; DeHart *et al.*, 1988; Matsunami *et al.*,1993; Grossman, 1996) and similar gradients in Na, K, and P are found in some CLS mesostasis glasses. Some mega-chondrules experienced Na-K-P metasomatism (Ruzicka *et al.*, 1998c). Evidently, both chondrules and CLSs interacted with volatile-rich environments. Chondrules and CLSs may differ in the extent to which they were metasomatized. Most CLSs have Na-poor compositions and show no evidence that their overall compositions were significantly affected by Na-metasomatism. However, this may not be true for normal-sized chondrules (Grossman, 1996; Ruzicka *et al.*, 1998c).

Precursors–The precursors to chondrules and CLSs were clearly different. The CLSs were derived from at least three already differentiated source regions, two of which were felsic. In contrast, chondrule precursors were more mafic and consisted largely of olivine and low-Ca pyroxene, and trace element data do not support the idea that the precursors to chondrules were differentiated (*e.g.*, Dodd, 1981; Grossman *et al.*, 1988; Grossman, 1988). Chondrule precursors may have formed by vapor-fractionation processes that resulted in diverse Al-Mg-Si ratios (*e.g.*, Grossman *et al.*, 1988; Grossman, 1988; Hewins *et al.*, 1997).

Abundances–Chondrules are the principal constituent of some chondrite types, but CLSs tend to be rare in lunar samples. The high abundances of chondrules in many chondrites imply that the chondrules were efficiently produced, concentrated, and preserved in chondrites. In contrast, the rarity of CLSs in most lunar samples is best explained if CLS production on the Moon was a relatively inefficient process.

Ages–The ages of CLSs are uncertain but are probably old (≥3.8 Ga), corresponding to the period when basin-scale impacts were occurring on the Moon (*e.g.*,Wilhelms, 1987), as only large impacts can keep material suspended above the lunar surface for sufficiently long durations (Symes *et al.*, 1998a). If CLSs formed in the Imbrium impact (*e.g.*, King *et al.*, 1972a,b; Kurat *et al.*, 1974; Symes *et al.*, 1998a), this would indicate they formed ~3.85 Ga ago (Wilhelms, 1987). The ages of individual chondrules are poorly known, but whole-rock and internal-isochron ages for chondrites using a variety of chronometers are typically in the range of 4.3–4.55 Ga, with the older ages interpreted as formation ages and the younger interpreted as being caused by later disturbances (*e.g.*, Brearley and Jones, 1998, and references therein). Most likely, chondrules formed close to 4.55 Ga ago, and CLSs formed significantly later.

Implications

It is obvious that there are many detailed similarities between chondrules and CLSs. Of the 13 characteristics for chondrules and CLSs listed in Table 7, only 3 or 4 (Mineralogies, Precursors, Abundances, and possibly Ages) indicate fundamental differences between the two types of objects. The question is, do these differences require significantly different origins for the two types of objects? The answer to this question is "probably not." The case can be made that differences in precursor compositions, local gravitational fields, and the likely presence of ambient gas during chondrule formation ultimately may be responsible for all of the differences between chondrules and CLSs and their host rocks. We suggest that many chondrules, but not necessarily all, may have formed by impact melting.

As noted above, differences in precursor compositions can explain the differences in overall mineralogies and types of relic grains found in the objects. Less obvious is the possible influence of precursor composition on evidence for vapor-fractionation trends. A distinction must be made between vapor fractionation that affected chondrule precursors (*e.g.*, Grossman *et al.*, 1988; Grossman, 1988; Hewins *et al.*, 1997) and that which occurred during the chondrule-forming process itself (*e.g.*, Sears *et al.*, 1996c; Huang *et al.*, 1996; Hewins *et al.*, 1997). Obviously, if the compositions of chondrules and CLSs were inherited from their precursors, and the precursors for chondrules were already vapor fractionated, whereas those for CLSs were not, only the chondrules would show evidence for vapor fractionation. Moreover, if the source regions for the CLSs (highland crust of the Moon) were deficient in volatiles such as Na and FeO compared to chondrule precursors, evidence for vapor loss of Na and FeO would be more obvious in the case of chondrules simply because there would be more volatiles to lose during heating.

A similar argument can also partly explain why rims are thicker around chondrules, as previously noted by Symes *et al.* (1998a). Rims rich in sulfide and FeNi-metal around chondrules may have formed by recondensation of S and Fe on chondrule surfaces following the partial vaporization of these elements in the precursor materials (*e.g.*, Zanda *et al.*, 1995; Huang *et al.*, 1996). A similar process, involving some combination of grain accretion and condensation, appears to have occurred to form CLS rims. Assuming that the precursors to chondrules were richer in volatiles such as S and FeO compared to CLS precursors, and that these species were partly vaporized during chondrule and CLS formation, more S- and Fe-rich gas would be generated during chondrule formation. Consequently, thicker rims could form on chondrules compared to CLSs, even for heating events of similar intensity and duration.

Besides differences in precursor composition, another important difference between CLSs and chondrules is the presence of stronger gravity on the Moon compared to the asteroidal-sized parent bodies of chondrites. The complexity of some chondrule rims, and especially the tendency for chondrule rims to be layered, suggests that chondrules acquired their rims over a longer time period than for CLSs, possibly in an episodic fashion. This can be explained if chondrules were suspended for a comparatively long period of time and had more opportunity of interacting with different environments than the CLSs. Longer suspension times and an enhanced opportunity for traversing different environments (*e.g.*, containing different proportions of sulfide and silicate dust) would be expected for chondrules orbiting or drifting between irregularly-shaped, low-gravity, asteroidal parent objects (Symes *et al.*, 1998a).

The rarity of CLSs compared to chondrules and the likely older ages of the latter are the most obvious differences between these objects. These differences probably indicate that the efficiency of producing chondrules in the earliest moments of solar system history was higher than that of producing CLSs at a later date. As noted above, CLS formation on the Moon was probably a relatively inefficient process, both because the high gravity of the Moon limited the amount of melt ejecta that could crystallize in free-flight, and because cooling rates for the melt ejecta were usually too rapid. We suggest that chondrules were more efficiently produced than CLSs because chondrules were produced in lower-gravity conditions at a time when nebular gas was still present.

On asteroidal-sized parent bodies, low gravity would allow more impact-ejecta to become free-floating, for longer periods of time, than is the case for the Moon. Indeed, the apparent need to form

CLSs in large impacts on the Moon does not apply to asteroidal-sized objects, because even smaller impact events on asteroids could potentially send melted material into orbit or into escape trajectories. However, relatively large impacts may still be needed to produce large amounts of impact melt. Impacts sufficiently large to produce CLSs on the Moon could lead to the disruption of asteroidal-sized parent bodies. In this case, 100% of any impact melt produced in the collision could become dispersed and free-floating, potentially resulting in many chondrules.

Chondrules probably formed in the solar nebula (*e.g.*, Grossman, 1988; Boss, 1996; Scott *et al.*, 1996; Hewins, 1997) when nebular gas was present, and we speculate that the presence of this gas may have been important, either to provide the necessary slow cooling rates for chondrules, or to enable chondrules to agglomerate after impact events. If nebular gas was needed for either reason, it would explain why chondrites were produced only in the early solar system close to 4.55 Ga ago.

Thus, provided that sufficient material could be melted by impact events, and provided that such material could crystallize and ultimately reaccrete on asteroidal objects, chondrule-rich rocks (*i.e.*, chondrites) could be formed. It has been argued that an insufficient quantity of melt can be produced by impacts and that ejected melt could not ultimately reaccumulate onto asteroids (*e.g.*, Taylor *et al.*, 1983; Keil *et al.*, 1997). We dispute the first point but acknowledge that further work is needed to determine the fate of ejecta produced on asteroidal parent bodies, especially in the presence of an ambient (albeit low-pressure) gas.

The amount of impact melt that could have been generated in the asteroid belt is considerable, if the Moon can be used as a guide. Hörz *et al.* (1991) estimated that impact melt constitutes 30–50% of all hand-specimen-sized rocks from the lunar highlands, and some 50% of all soil materials from the maria and highlands. A single terrestrial impact seems to produce ~10% impact melt in its ejecta, which implies that multiple impacts were involved in producing the high fraction of impact melt near the surface of the Moon (Hörz *et al.*, 1991). Higher porosity and smaller grain sizes in the impact target increase the fraction of impact melt that can be generated (*e.g.*, Hörz *et al.*, 1991). Assuming that early-formed chondrite parent bodies were small and consisted of agglomerations of dust grains, it seems likely that they would have been both more porous and finer-grained than the differentiated crust of the Moon, and these factors would promote the formation of more impact melt. One would expect impact melting to have been an important process on asteroids unless collisional velocities in the asteroid belt were much lower than at the Moon. The well-known effect of Jupiter to cause perturbations of asteroid orbits implies that appreciable collisional velocities were probably attained in the asteroid belt at some stage. This could have been true even if nebular gas had not fully dissipated.

Thus, large amounts of impact melt could have been generated in the asteroid belt. The question is not so much whether impact melting could have been an important process, but whether any of this impact melt became incorporated in meteorite parent bodies and would be identifiable as such. On the Moon, one would expect most impact melt to have originally resided in lenses, pools, or sheets within craters, to produce clast-laden or clast-poor impact-melt breccias; or in ejecta that landed close to parent craters, where it could form either impact-melt breccias, agglutinates, or glassy spherules. On asteroids, impact-melt breccias could still be produced within craters, but lower gravitational fields on asteroids might significantly change the nature of the cratering process, resulting in melt rocks that did not necessarily resemble their lunar counterparts in all ways. Similarly, most ejecta on asteroids could become free-floating for at least short periods of time, preventing (or at least inhibiting) the formation of agglutinates or impact-glass-coated materials. Consequently, a detailed similarity of impact-produced rocks on the Moon and asteroids would not necessarily be expected (*e.g.*, McKay *et al.*, 1989).

We speculate that following the agglomeration of chondrules (some of which were produced by impact melting) and unmelted material, subsequent impact events either remelted some chondrules or processed additional unmelted material through a melt stage. This would tend to increase the apparent efficiency of chondrule production, just as the fraction of impact melt on the Moon was apparently increased by multiple impact events. Multiple episodes of impact melting would result in "recycling" of some chondrules through more than one episode of melting. Such recycling, either with or without accompanying volatile-loss, has been proposed for chondrules (*e.g.*, Alexander, 1996; Jones, 1996; Wasson *et al.*, 1995). Remelting of fine-grained rims on chondrules to produce igneous-textured rims (Rubin, 1984; Krot and Wasson, 1995; Rubin and Krot, 1996), which presumably requires short but intense heating, could conceivably occur by the partial melting or "sintering" of preexisting, rimmed chondrules during a later impact event. Alternatively, the igneous-textured rims on chondrules may have formed by the accretion of melted material around a preexisting chondrule, perhaps in a manner analogous to that which formed lunar object 318c5/g5.

Different chondrite types have different physical characteristics (*e.g.*, chondrule size-distributions, chondrule abundances, chondrule rim thicknesses), and there is no obvious reason why impact processes could not have been responsible for producing such differences. Differences in parent body sizes or shapes, ambient gas densities, impactor velocities or sizes, and overall bombardment history would presumably affect the number and sizes of impact-melt droplets that were produced, the number and thickness of rims produced during the free-floating stage, and any "sorting" processes during the reaccretion or agglomeration of chondritic material.

To summarize, we see no "fatal flaw" in forming chondrules and CLSs in similar ways, by impact melting. The differences between the two types of objects can be reconciled with an impact-melt origin by chemical differences in their precursors, by differences in free-flight times owing to differences in gravity fields of different parent bodies, by the likely presence of an ambient low-pressure (nebular) gas at the time of chondrule formation, and by differences in cooling rates experienced by melt ejecta. Cooling rates may have been lower for melt ejecta produced in the asteroid belt if nebular gas or substantial dispersed material was present and able to act as a thermal insulator.

However, not all chondrules necessarily formed by impact melting. Evidence provided by large inclusions ("mega-chondrules" and "large, igneous-textured clasts") in ordinary chondrites suggests that the inclusions represent two chemical populations of objects, with Na-rich inclusions having formed by impact melting, and Na-poor inclusions having formed by an unknown mechanism that involved vapor-fractionation processes (Ruzicka *et al.*, 1998c, 2000). If chondrules formed in the same way as the larger inclusions, as evidence suggests (Ruzicka *et al.*, 1998c), there may have been more than one way to produce chondrules.

CONCLUSIONS

Crystal-bearing lunar spherules can be subdivided into two major types, feldspathic and olivine-rich. Feldspathic CLSs were produced by impact melting of feldspathic crust on the Moon, involving target materials that consisted of a mixture of roughly equal portions of anorthositic and Mg-suite rocks. Olivine-rich CLSs were produced by impact melting of magnesian lunar materials atypical of the upper crust. Some CLSs were rimmed and metasomatized in a volatile-rich environment before they were brecciated. Crystal-bearing lunar spherules were sometimes incompletely melted, resulting in relic grains. Incomplete melting, slow cooling rates, and long ballistic travel times in impact ejecta curtains were probably the key features that allowed CLSs to partly crystallize. These conditions do not appear to have been simultaneously attained often on the Moon, resulting in the relative rarity of CLSs. The many similarities between CLS and chondrules suggest that an impact-melt origin for chondrules is more difficult to rule out than some have previously argued. Differences between CLSs and chondrules can be reconciled with an impact-melt origin for chondrules when differences in precursor compositions, parent body gravity fields, and the likely presence of ambient nebular gas during chondrule formation are taken into account. More work on collisional processes affecting asteroidal-sized objects in the early solar system is needed, especially to evaluate the dynamics of impact-ejection and reaccretion processes in a gaseous medium.

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