

## THE SEARCH FOR INTERSTELLAR COMPONENTS IN INTERPLANETARY DUST PARTICLES

John Bradley

MVA Inc., 5500/200 Oakbrook Parkway, Norcross, GA 30093, USA

Trevor Ireland

Research School of Earth Sciences, Australian National University,  
Canberra, ACT, Australia

### **Abstract.**

Amorphous silicate grains known as GEMS have been found in chondritic porous interplanetary dust particles (IDPs) from comets. GEMS are composed of nanometer-sized FeNi metal and Fe-rich sulfide crystals embedded in silicate glass. The properties of GEMS appear to have been shaped primarily by exposure to ionizing radiation and, since the irradiation occurred *prior to* accretion of the cometary IDPs, GEMS may have formed either in the solar nebula or presolar interstellar environments. The sizes, shapes, structures, and compositions of GEMS correspond to those of interstellar “amorphous silicate” grains. Nanometer-sized superparamagnetic metal inclusions dispersed throughout GEMS provide a logical explanation for alignment of interstellar silicate grains in the galactic magnetic field. Irrespective of their origins, GEMS are an important class of submicrometer-sized chondritic objects. If they were formed in the solar nebula, then they are among the oldest known solar nebula solids. If they are presolar, then they are probably examples of the “amorphous silicate” grains which are ubiquitous throughout the interstellar medium.

### **1. Introduction**

The development of laboratory measurement techniques for distinguishing between cometary and asteroidal IDPs is an important milestone in interplanetary dust research. Distinction between cometary and asteroidal origins for *some* IDPs is now possible as a result of the ability to measure sub-nanogram quantities of He released from IDPs during stepwise heating (Nier and Schlutter, 1993), determination of the masses and densities of IDPs (Love et al., 1993), and development of theoretical models of “most probable entry speed” for cometary and asteroidal IDPs entering the atmosphere (Love and Brownlee, 1994). The temperature range over which an IDP releases its implanted solar wind He (during stepwise heating in the laboratory) is a function of its prior thermal history, specifically frictional heating during atmospheric entry. If the mass, density, and He release temperature are known, the atmospheric entry speed can be estimated. Typical cometary IDPs enter the atmosphere at higher speeds than typical asteroidal IDPs, with the asteroidal contribution becoming minimal at speeds  $>16$  km/sec<sup>-1</sup>. The He release temperatures of 50 IDPs recently measured indicated that ~20% are “almost certainly” of cometary origin (Brownlee, 1996).

Most high-speed “cometary” IDPs belong to the anhydrous chondritic porous (CP) subset (Brownlee et al., 1994), although some CP IDPs which entered the atmosphere at much lower speeds may be of asteroidal origin. CP IDPs differ fundamentally from other classes of IDPs, chondritic meteorites, and



Figure 1. Secondary electron image of a GEM (mounted on a gold substrate).

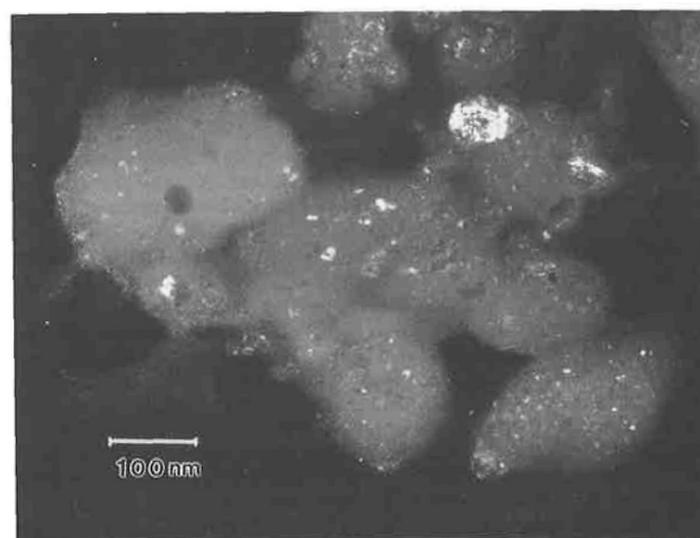


Figure 2. Darkfield electron micrograph of GEMs adhering together in the matrix of IDP U220A19. Adhesion of GEMs may result from irradiation exposure (see Johnson, 1985). Bright specks are metal and sulfide crystals and the uniform grey matrix is silicate glass.

polar micrometeorites in terms of texture, porosity, grain size, mineralogy, and petrography. Although the average grain size in CP IDPs is on a scale of nanometers, an unprecedented level of heterogeneity among known meteoritic materials, progress in microanalytical instrumentation has made it possible to measure the compositions and crystal structures of even the finest grained components (Klöck and Stadermann, 1994; Bradley, 1994a; Thomas et al., 1995). Furthermore, a broad spectrum of *bulk* analytical measurements have now been made on IDPs. They include inorganic (major and trace) elements, organic, noble gas, isotopic compositions, and spectral reflectivities (Zolensky et al, 1994; Schramm et al., 1989; Flynn et al., 1995).

Cometary IDPs are presently the only available known samples of comets. As such they are expected to contain some of the most well-preserved, primitive meteoritic materials (e.g. solar nebula condensates and interstellar grains) (Greenberg, 1982; Brownlee, 1994; Hanner, 1994). This paper describes recent research performed on cometary IDPs using analytical electron microscopy. The focus of the research has been on GEMS (glass with embedded metal and sulfides). GEMS are tiny submicrometer spheroids (0.1-0.5  $\mu\text{m}$  diameter) with bulk compositions that are *approximately* (within a factor of three) chondritic (solar) (Bradley, 1994b). They form the building blocks of anhydrous CP IDPs in general and cometary IDPs in particular. Their compositions, mineralogy and petrography appear to have been shaped by exposure to ionizing radiation. Since the exposure occurred prior to the accretion of cometary IDPs, and therefore comets themselves, GEMS are likely either solar nebula or presolar interstellar grains. The properties of GEMS (size, shape, mineralogy) bear an uncanny resemblance to those of interstellar silicate grains as inferred from astronomical observations.

## 2. Chemistry and Mineralogy of GEMS

GEMS are ubiquitous within the ultrafine-grained matrices of CP IDPs. This matrix is *not* a homogenous mass but rather a loose mixture of discrete aggregates containing nanometer-sized crystals embedded in either glass or carbonaceous material, as well as single mineral grains (e.g. olivine, pyroxene, and Fe-rich sulfides) (Klöck and Stadermann, 1994; Thomas et al., 1995). At least three types of aggregates have been recognized (Bradley, 1994a). GEMS are but one type of aggregate (Figs. 1 & 2). They contain nanometer-sized metal and Fe-rich sulfide crystals embedded in silicate glass. The metal is body centered cubic (bcc) kamacite [Fe,Ni] (Ni < 10 weight %) and the sulfides appear to be pure FeS. Some sulfide crystals may also contain minor Ni. The compositions of GEMS are *approximately* solar (chondritic) for all major elements except C and S (Table 1). C appears to be highly depleted in GEMS although, because IDP thin sections are supported on thin carbon substrates for analysis of GEMS, it is not possible to conclude that they are absolutely carbon-free. Unlike the other major elements whose abundances are highly variable, S is consistently depleted 50-80% below solar abundances (Table 1). GEMS are also non-stoichiometric with respect to oxygen. If Fe, Ni, and S are assigned as metal (FeNi) and sulfides (FeS) and the remaining elements as oxides (MgO, Al<sub>2</sub>O<sub>3</sub>, SiO<sub>2</sub> etc....) there is a stoichiometric "excess" of O (Table 1). Evidence from electron energy loss spectroscopy (EELS) measurements suggest that the excess oxygen is hydroxyl in the form of (-OH) ions (Bradley, 1994b). GEMS exhibit highly unusual chemical gradients. In some GEMS, Mg concentration decreases towards their outer surfaces (Fig. 3) (Brownlee et al., 1996), while in others Mg concentration increases at their

polar micrometeorites in terms of texture, porosity, grain size, mineralogy, and petrography. Although the average grain size in CP IDPs is on a scale of nanometers, an unprecedented level of heterogeneity among known meteoritic materials, progress in microanalytical instrumentation has made it possible to measure the compositions and crystal structures of even the finest grained components (Klöck and Stadermann, 1994, Bradley, 1994a; Thomas et al., 1995). Furthermore, a broad spectrum of *bulk* analytical measurements have now been made on IDPs. They include inorganic (major and trace) elements, organic, noble gas, isotopic compositions, and spectral reflectivities (Zolensky et al. 1994; Schramm et al., 1989; Flynn et al., 1995 ).

Cometary IDPs are presently the only available known samples of comets. As such they are expected to contain some of the most well-preserved, primitive meteoritic materials (e.g. solar nebula condensates and interstellar grains) (Greenberg, 1982; Brownlee, 1994; Hanner, 1994). This paper describes recent research performed on cometary IDPs using analytical electron microscopy. The focus of the research has been on GEMS (glass with embedded metal and sulfides). GEMS are tiny submicrometer spheroids (0.1-0.5  $\mu\text{m}$  diameter) with bulk compositions that are *approximately* (within a factor of three) chondritic (solar) (Bradley, 1994b). They form the building blocks of anhydrous CP IDPs in general and cometary IDPs in particular. Their compositions, mineralogy and petrography appear to have been shaped by exposure to ionizing radiation. Since the exposure occurred prior to the accretion of cometary IDPs, and therefore comets themselves, GEMS are likely either solar nebula or presolar interstellar grains. The properties of GEMS (size, shape, mineralogy) bear an uncanny resemblance to those of interstellar silicate grains as inferred from astronomical observations.

## 2. Chemistry and Mineralogy of GEMS

GEMS are ubiquitous within the ultrafine-grained matrices of CP IDPs. This matrix is *not* a homogenous mass but rather a loose mixture of discrete aggregates containing nanometer-sized crystals embedded in either glass or carbonaceous material, as well as single mineral grains (e.g. olivine, pyroxene, and Fe-rich sulfides) (Klöck and Stadermann, 1994, Thomas et al., 1995). At least three types of aggregates have been recognized (Bradley, 1994a). GEMS are but one type of aggregate (Figs. 1 & 2). They contain nanometer-sized metal and Fe-rich sulfide crystals embedded in silicate glass. The metal is body centered cubic (bcc) kamacite [Fe,Ni] (Ni < 10 weight %) and the sulfides appear to be pure FeS. Some sulfide crystals may also contain minor Ni. The compositions of GEMS are *approximately* solar (chondritic) for all major elements except C and S (Table 1). C appears to be highly depleted in GEMS although, because IDP thin sections are supported on thin carbon substrates for analysis of GEMS, it is not possible to conclude that they are absolutely carbon-free. Unlike the other major elements whose abundances are highly variable, S is consistently depleted 50-80% below solar abundances (Table 1). GEMS are also non-stoichiometric with respect to oxygen. If Fe, Ni, and S are assigned as metal (FeNi) and sulfides (FeS) and the remaining elements as oxides ( MgO, Al<sub>2</sub>O<sub>3</sub>, SiO<sub>2</sub> etc....) there is a stoichiometric "excess" of O (Table 1). Evidence from electron energy loss spectroscopy (EELS) measurements suggest that the excess oxygen is hydroxyl in the form of (-OH) ions (Bradley, 1994b). GEMS exhibit highly unusual chemical gradients. In some GEMS, Mg concentration decreases towards their outer surfaces (Fig. 3) (Brownlee et al., 1996), while in others Mg concentration increases at their

surfaces. Some (low-Mg) GEMS exhibit constant Mg and Si depletions towards their outer surfaces (Bradley, 1994). The causes of these compositional gradients are discussed in Section 4.

---

**Table 1**

**Bulk Compositions of GEMS**

atom abundances (normalized to Si)

	O	Mg	Al	S	Ca	Cr	Mn	Fe	Ni	% O excess
1	3.76	0.38	0.12	0.14	0.01	0.02	0.005	0.42	0.03	45
2	3.7	0.24	0.11	0.08	0.02	0.04	-	0.38	0.03	23
3	3.96	1.11	0.08	0.10	0.08	0.03	0.008	0.70	0.05	19
4	3.51	0.65	0.11	0.05	0.03	0.02	0.004	0.12	0.006	22
5	4.99	0.82	0.12	0.12	0.04	0.01	0.01	0.43	0.03	66
6	4.34	1.075	.085	.515	.061	.013	.01	.9	.049	

-----  
 Lines 1 -5 Compositions of 5 GEMS

Line 6 Solar (CI chondrite) abundances (Anders and Ebihara, 1982).

---

### 3. Isotopic Compositions of GEMS

Detection of non-solar isotope abundances in GEMS could potentially provide unambiguous proof of their presolar origins. The first isotopic measurements of individual GEMS were recently attempted in the ion microprobe. Fines from a cluster IDP (L2008 #5) were dispersed onto gold ion-microprobe mounts. (Fines are micrometer and submicrometer-sized fragments of large IDPs (> 35  $\mu\text{m}$  diameter) that break up when they impact the collection surface). Prior to ion microprobe measurements, the exact locations and compositions of several GEMS were determined using scanning electron microscopy (e.g. Fig. 1). Grains like the one shown in Figure 1 are *assumed* to be GEMS from their sizes, spheroidal shapes, and approximately chondritic (solar) abundances of the major elements Mg, Si, S, Fe and Ni. Rigorous identification of GEMS also requires transmission electron microscope (TEM) observations of internal nanometer-sized Fe metal and Fe-sulfide grains embedded in silicate glass matrix. Unfortunately, it is not yet possible to prepare or otherwise subdivide individual GEMS for ion microprobe *and* TEM measurements. Table 2 lists the Mg isotope abundances found in four GEMS. Although the Mg values are not significantly different from solar system values, they at least demonstrate that it is possible to measure the isotopic compositions of individual submicrometer GEMS.

Table 2

## Mg isotopes in GEMS

<u>SAMPLE #</u>	<u><math>\Delta^{25}\text{Mg} \dagger</math></u>	<u><math>\delta^{26}\text{Mg} \ddagger</math></u>
1	$-12 \pm 10$	$9 \pm 16$
15	$-5 \pm 7$	$-9 \pm 14$
18	$-1 \pm 7$	$-5 \pm 12$
21	$-14 \pm 7$	$10 \pm 11$

$\dagger \Delta^{25}\text{Mg}$  is the measured isotopic mass fractionation per amu and comprises instrumental (ion microprobe) and intrinsic fractionation. The values measured are consistent with being solely due to instrumental fractionation.

$\ddagger \delta^{26}\text{Mg}$  is the residual after removal of the measured mass fractionation.

Errors are  $2\sigma$

A variety of presolar grains in meteorites (e.g. diamond, silicon carbide) have been identified as circumstellar grains from their non-solar isotopic compositions (Ott, 1993). Thus, it is tempting to assume that if GEMS are truly interstellar grains, they should also be isotopically anomalous. However, typical interstellar grains (e.g. silicates) may not be significantly different, in a chemical or isotopic sense, than typical solar system grains. The interstellar medium is a dynamic environment where average grains may have been shock melted, sputtered, vaporized, and condensed several times over during their  $10^8$ - $10^9$  year lifetimes (Nuth, 1988). Grains that were originally isotopically anomalous are expected to become more chemically and isotopically "homogenized" (Bradley, et al., 1988). (The average composition of galactic cosmic rays is, with a few exceptions, not significantly different from solar system abundances (Wiedenbeck, 1984)). The bulk "chondritic" (solar) compositions of GEMS are perhaps evidence of this homogenization (Table 1). While non-solar isotope abundances in GEMS might provide unambiguous proof of their pre-solar origins, detection of "normal" isotopic abundances does not rule out a pre-solar origin. Non-solar D and  $^{15}\text{N}$  enrichments consistent with preserved interstellar cloud material have been observed in some IDPs (McKeegan et al., 1985; Stadermann, 1989). Some of the D excesses ( $\sim 25,000 \%$ ) are the largest ever observed in a natural sample in the laboratory (Messenger et al., 1995). The D and  $^{15}\text{N}$  abundances in (GEMS-rich) IDPs certainly support the hypothesis that GEMS are of interstellar origin but, ironically, an (outer) solar nebular origin for these isotope enrichments cannot be absolutely ruled out (McKeegan et al., 1985; Walker, 1994).

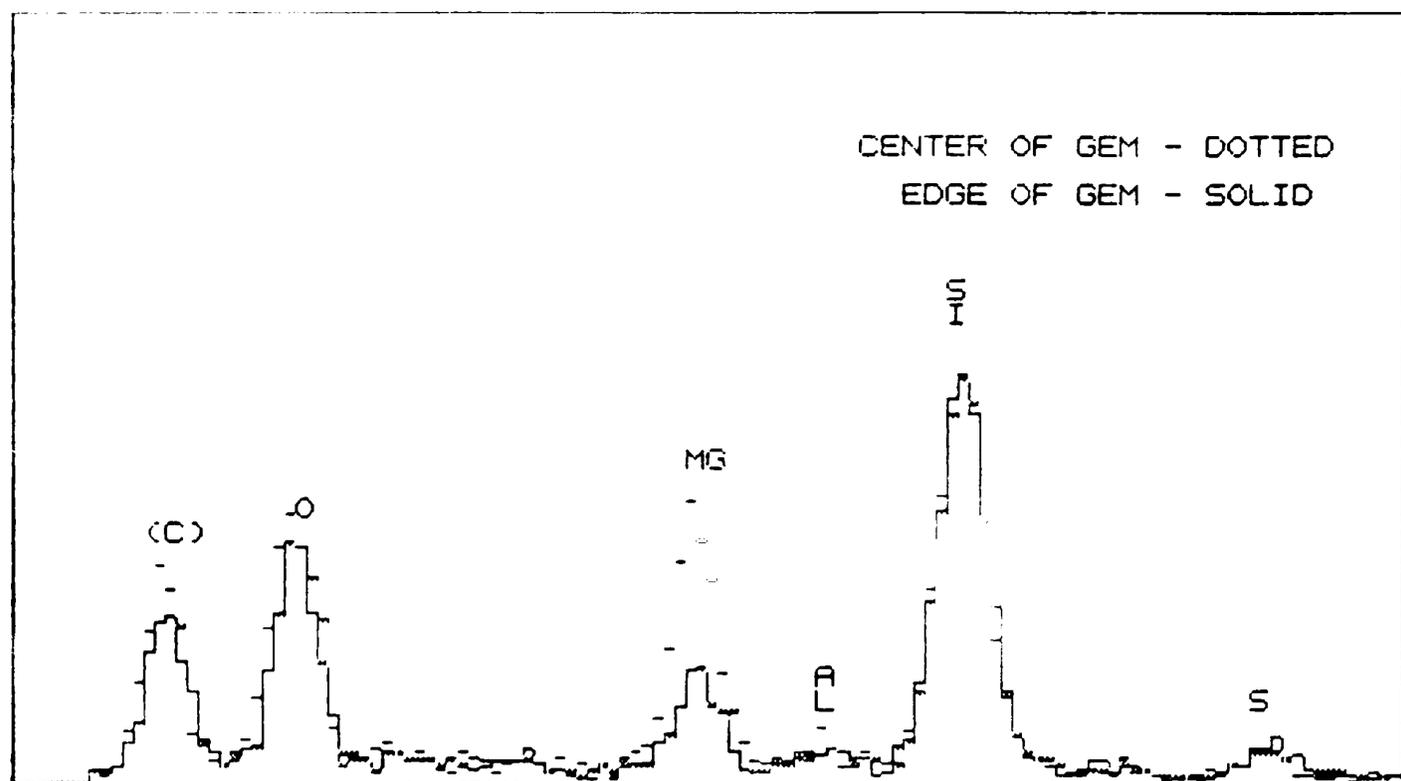


Figure 3. Energy-dispersive x-ray spectra collected at the center (solid trace) and outer edge (dotted trace) of a thin section of a GEM in IDP W7027F11. Mg/Si falls steadily from the center to the edge of the GEM (see Bradley, 1994b & Brownlee et al., 1996). Spectra were collected from ultramicrotomed thin-sections (<80 nm thick) of a GEM using a 200 KeV, ~20 nm diameter electron probe.

Additional measurements of the isotopic compositions of GEMS are a high priority. Some GEMS may contain subtle remnant isotopic signatures of circumstellar precursors or even isotopically distinct spallation products. A systematic examination of a larger population of grains dispersed on gold ion microprobe substrates is in progress. Previous *bulk* isotope measurements of Mg, Si, and O in CP IDPs have yielded negative results for isotope anomalies (Esat et al., 1979). While these results may indicate that anhydrous CP IDPs are isotopically normal with respect to silicate mineralogy, they may also indicate that *bulk* measurements of IDPs (containing tens or even hundreds of GEMS) provide a highly averaged isotope measurement. Clearly, development of ion microprobes capable of high sensitivity and nanometer-scale isotopic measurements could have a major impact on our understanding of GEMS.

#### 4. Irradiation Histories of GEMS

Amorphous silicates, nanocrystals of metallic Fe, non-stoichiometry, and surficial Mg depletions in GEMS are all consistent with a “sputtered target” that has been chemically altered by bombarding ions. The same effects were recently observed on the surfaces of (solar wind) irradiated lunar pyroxene and feldspar crystals, as well as on silicate crystals that had been exposed to high doses of 1 keV H<sup>+</sup>, 20 keV H<sup>+</sup>, and 4 keV He<sup>+</sup> in laboratory simulations (Bradley, 1994b; Bradley et al., 1996). GEMS with surficial Mg enrichments may contain “sputtered residues” on their outer surfaces.

Where and when were GEMS irradiated? Since GEMS are distributed throughout IDPs and were irradiated *prior to* accretion of the IDPs, they must predate IDPs. In the case of cometary IDPs, the GEMS must have formed *prior to* the accretion of comets. This suggests two possible formation environments, the solar nebula during an early phase of the Sun when irradiation episodes may

have been severe, or the presolar interstellar environment (where grain lifetimes are  $10^8$ - $10^9$  years and irradiation exposures are prolonged) (Mathis, 1993). A solar nebula origin seems problematical for the following reason. GEMS are admixed with what appear to be non-irradiated mineral grains in the matrices of CP IDPs; i.e. grains were either heavily irradiated or not irradiated at all, with no continuum in between. It is difficult to envisage how GEMS could have been heavily irradiated in the vicinity of comet accretion while other grains were not. A more logical explanation may be that cometary IDPs contain material from two fundamentally different reservoirs, "ancient" (preaccretionally irradiated) interstellar grains (GEMS) and "recent" (unirradiated) solar nebular grains (Ney, 1977; Greenberg, 1982; Hanner, 1994). It is *inevitable* that interstellar grains surviving in comets were preaccretionally irradiated.

## 5. Summary

A population of "amorphous silicate" grains known as "GEMS" have been found in interplanetary dust particles which are most likely from comets. The properties of GEMS correspond to the properties of interstellar amorphous silicate grains (Bradley, 1984b; Flynn, 1994; Martin, 1995; Goodman and Whittet, 1995). Astronomical observations indicate that interstellar silicates are  $\sim 0.25$   $\mu\text{m}$  radius, predominantly "amorphous", and may contain nanometer sized metal inclusions which would explain their ability to align in the galactic magnetic field (Mathis, 1993). GEMS are  $\sim 0.25$   $\mu\text{m}$  radius, they contain amorphous silicate glass, and they contain nanometer-sized metal (and sulfide) inclusions. Even the depletions of S measured in GEMS (Table 1) are consistent with the expected compositions of interstellar silicates (Martin, 1995). GEMS truly are a "raisin pudding of ferromagnetic clusters in non-magnetic matrix.....", first proposed by Jones and Spitzer (1967) to explain interstellar grain alignment. However, a solar nebula origin for GEMS cannot yet be ruled out. Clarification of their origin(s) is being sought through isotope measurements on *individual* GEMS, experiments designed to determine the magnitude and duration of irradiation exposure, and high-resolution analytical electron microscopy of the mineralogy and chemistry of GEMS and other fine-grained components in IDPs.

## Acknowledgment

This research is sponsored by NASA contract NASW-5835 and NASA Grant NAGW 3636.

## References

- Anders, E. and Ebihara, M. (1982) *GCA* 46, 2363.
- Bradley, J. P. (1994a) *GCA* 58, 2123.
- Bradley, J. P. (1994b) *Science* 265, 925.
- Bradley, J. P. *et al.* (1988) in *Meteorites and the Early Solar System* J. F. Kerridge and M. S. Matthews, Eds. (Univ. Arizona Press) p 861.
- Bradley, J. P. *et al.* (1996) *LPSC 27*, *in press*.
- Brownlee, D. E. (1994) in *Analysis of Interplanetary Dust*. M. E. Zolensky, T. L. Wilson, F. J. M. Rietmeijer, and G. J. Flynn, Eds. (AIP Conf. Proc. 310) p5.
- Brownlee, D. E. (1996) personal communication.
- Brownlee, D. E. *et al.* (1994) *LPSC 25*, 185.
- Brownlee, D. E. *et al.*, (1996) *LPSC 27*, *in press*.
- Esat, T. M. *et al.* (1979) *Science* 206, 190.

- Flynn, G. J. (1994) *Nature* 371, 287.
- Flynn, G. J. *et al.* (1995) *Meteoritics* 30, 505.
- Goodman, A. A. and Whittet, D. C. B. (1995) *Ap. J.* 455, L181.
- Greenberg, J. M. (1982) in *Comets*, L. L. Wilkening, Ed. (Univ. Arizona Press, Tucson 1982) p 131.
- Hanner, M. S. (1994) in *Analysis of Interplanetary Dust*. M. E. Zolensky, T. L. Wilson, F. J. M. Rietmeijer, and G. J. Flynn, Eds. (AIP Conf. Proc. 310) p23.
- Johnson, R. E. (1985) in *Ices in the Solar System*. J. Klinger et al. Eds. (Riedel Publishing Company). p. 337
- Jones, R. V. and Spitzer, L. (1967) *Ap. J.* 147, 943.
- Klöck, W. and Stadermann, F. J. (1994) in *Analysis of Interplanetary Dust*. M. E. Zolensky, T. L. Wilson, F. J. M. Rietmeijer, and G. J. Flynn, Eds. (AIP Conf. Proc. 310) p 51.
- Love, S. J. *et al.* (1993) *LPSC* 24, 901.
- Love, S. J. and Brownlee, D. E. (1994) *Meteoritics* 29, 69.
- Martin, P. G. (1995) *Ap. J.* 445, L63.
- Mathis, J. S. (1993) *Rep. Prog. Phys.* 56, 605.
- McKeegan, K. D. *et al.* (1985) *Geochim. Cosmochim. Acta* 49, 1971
- Messenger, S. *et al.* (1995) *Meteoritics*, 30, 546.
- Ney, E. P. (1977) *Science* 195, 541.
- Nier, A. O. and Schlutter, D. J. (1993) *Meteoritics* 28, 675.
- Nuñh, J. A. (1988) in *Meteorites and the Early Solar System*. J. Kerridge and M. S. Matthews, Eds. (Univ. Arizona Press, Tucson) pp 457-497
- Ott, U. (1993) *Nature* 364, 25
- Schramm, L. S. *et al.* (1989) *Meteoritics* 24, 99.
- Stadermann, F. J. *et al.* (1989) *Meteoritics* 24, 327
- Thomas, K. L. *et al.* (1995) *GCA* 59, 2797
- Walker, R. M. (1994) in *Analysis of Interplanetary Dust*. M. E. Zolensky, T. L. Wilson, F. J. M. Rietmeijer, and G. J. Flynn, Eds. (AIP Conf. Proc. 310) p 203.
- Wiedenbeck, M. E. (1984) *Adv. Space Res.* 4, 15.
- Zolensky, M. E. *et al.* (1994), "Analysis of Interplanetary Dust" (American Institute of Physics Conference Proceedings, New York, 1994) pp357