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Thermoluminescence and the origin of the dark matrix of Fayetteville and similar meteorites

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Abstract—The induced thermoluminescence properties and carbon contents of the dark matrix and light clasts of 6 regolith breccias have been measured. The TL sensitivity of the matrix (normalized to that of the clasts) decreases from ~0.8 to ~0.3 with increasing inert gas content, increasing abundances of highly volatile elements and carbon, and decreasing amounts of interstitial, grain-boundary glass. For meteorites with the highest inert-gas contents (Fayetteville and Leighton), the matrix samples have broader TL peaks (by ~10°C) and lower peak temperatures (by ~10°C) than the light clasts, whereas meteorites with low inert-gas contents (*e.g.* Pantar) do not show this difference in the TL properties of the two lithologies. It is argued that the data are consistent with the formation of the dark matrix by comminution of the light clasts with the addition of a component, perhaps CM-like chondrites, with TL properties distinct from those of ordinary chondrites. The present data are inconsistent with the matrix being a new "primitive" material akin to type 3 ordinary chondrites, or with the dark matrix being related to the clasts by purely thermal processes.

INTRODUCTION

BRECCIATION IS EXTREMELY COMMON among the ordinary chondrites, especially among those of the LL class. However, a few chondritic breccias, probably around 30, consist of light clasts in an unusual dark matrix (KEIL, 1982). This unusual matrix material contains solar gases, solar flare tracks, high abundances of carbon and volatile elements and clasts which have been shocked or even melted (SUESS *et al.*, 1964; PELLAS, 1972; REIDER and WÄNKE, 1969). Occasionally, small clasts of a different class to the host are observed (WILKENING, 1976). Clearly these breccias experienced a period of regolith activity. Of several questions posed by the regolith breccias, those which have attracted most attention concern the origin of the dark material and the time and duration of regolith activity.

In their description of the Pantar regolith breccia, FREDRIKSSON and KEIL (1963) suggested that the dark matrix was formed by comminution of clast material. Recent TEM observations of the matrix of Pantar, St. Mesmin and Weston also suggest that the matrix is comminuted host material (ASHWORTH and BARBER, 1976). The inert gas enrichment has subsequently been attributed to the entrapment of solar wind gases (SUESS *et al.*, 1964), while the enrichment in highly volatile elements has been attributed to an influx of volatile-rich material. This influx may be in the form of gases distilled from deeper in the parent body (DREIBUS and WÄNKE, 1980), or of volatile-rich material falling into the regolith from space (MÜLLER and ZÄHRINGER, 1966; MAZOR and ANDERS, 1967). Most of the macroscopic xenoliths found in regolith breccias are CM or CM-like and therefore volatile-rich (WILKENING, 1976). Elemental proportions for the dark matrix are not entirely consistent with any known meteorite class, and may have been disturbed during the regolith processing (CHOU *et al.*, 1981). It has also been argued that the matrix may be an entirely new type of "primitive" material whose unusual properties are "primary" and not produced by the alteration of material from the known classes (BART and LIPSCHUTZ, 1979; LIPSCHUTZ *et al.*, 1983); several authors have likened the dark matrix of regolith breccias to the highly primitive type 3 ordinary chondrites (*e.g.*, BINNS, 1967; WILKENING, 1976).

Discussion of the time of regolith activity has centered on whether it occurred early, more than 4.0 Ga ago (*e.g.* PELLAS, 1972), or whether it immediately preceded exposure to cosmic radiation (*e.g.* SCHULTZ and SIGNER, 1977). Arguments for the former are the great ages, determined by radioisotopes or fission tracks, of the bulk of the breccia clasts, including those which were heavily shocked or melted during regolith working. Arguments for a recent exposure center on the occasional clasts with different cosmic ray exposure history to the bulk of the meteorite and several clasts present in regolith breccias which have formation ages much less than 4.0 Ga. The duration of regolith activity might also be inferred from the spread in cosmic-ray exposure ages and formation ages for the clasts.

The amount of trapped gas, volatile elements, and solar flare charged-particle tracks in the dark matrix can vary by orders of magnitude from one regolith breccia to another. PELLAS (1972) pointed out that the relatively gas-poor regolith breccias were better consolidated than the others (Fayetteville excepted), and suggested that the lithification process responsible for consolidation caused gas-loss, so that gas-content was *inversely dependent* on the extent of regolith working. Consistent with this, BISCHOFF *et al.* (1983) found that the abundance of interstitial grain-boundary glass, which was assumed to be associated with lithification, varied inversely with gas-content. However, it is also possible that trapped

* Research performed as part of a consortium to study the Fayetteville meteorite.

gas content, volatile elements and track density reflect different durations of regolith activity.

The regolith breccias therefore pose a number of questions which existing data only partially resolve. In the present paper, we report induced thermoluminescence data for six gas-rich regolith breccias. The TL sensitivity of all but a few ordinary chondrites depends on the amount of feldspar, while the temperature and width of the dominant TL peak depends on its thermal history (GUIMON *et al.*, 1985, 1986; HARTMETZ and SEARS, 1987). Feldspar is acutely sensitive to the types of process (metamorphism, shock, partial fusion) discussed in connection with the regolith breccias. We also report new data for the carbon content of the same six breccias which, with the TL data, provide new insights into the history of regolith breccias.

EXPERIMENTAL

Our samples and their sources are listed in Table 1. The Fayetteville samples are from the University of Arkansas specimen used for the Fayetteville consortium study; the sample nomenclature and locations are described by SCHWARZ and SEARS (1988). The Pantar samples were broken from a 2 mm thick slice from the collection at Arizona State University and are described in detail in HAQ (1987). The remaining samples were provided by M. E. Lipschutz (Purdue University) and are unused fragments from the studies of BART and LIPSCHUTZ (1979) and LIPSCHUTZ *et al.* (1983).

Fragments whose weight is given in Table 1 were lightly ground, the metal removed with a hand magnet, then re-ground to pass through a 100 mesh sieve. Aliquants weighing 5 ± 1 mg were placed in a Cu pan (5 mm diameter, 2 mm depth, 0.01 mm base thickness) for TL measurement. After heating to 500°C in the TL apparatus, to remove natural TL, the sample was exposed to a 250 mCi Sr-90 beta source until the absorbed dose was ~2 krad. The induced TL of each sample was measured 3 times, and samples of Dhajala were run at the beginning and end of each series of measurements to act as normalization standard and monitor stability of the apparatus. As in previous work, TL sensitivity, peak temperature and width data were measured from the glow curves, peak width being defined as the full width at half maximum peak height (SEARS and WEEKS, 1983). Uncertainties are standard deviations calculated from replicate measurements, compounded with the uncertainty for Dhajala in the case of TL sensitivity.

Total carbon was determined using methods outlined and reviewed by MOORE *et al.* (1971, 1972).

RESULTS

The data are presented in Table 1 and plotted in Figs. 1 and 2. For all meteorites, the dark matrix has lower mean TL sensitivity than the light clasts, but the extent of the difference depends on the meteorite (Fig. 1). For Pantar the difference is very small, the dark matrix having a TL sensitivity typically 80% of that of the light clasts. SEARS' (1981) data for Plainview shows a similarly small difference in TL sensitivity between clasts and matrix. St. Mesmin and Cangas de Onis show a somewhat greater difference between clast and matrix, typically the dark matrix has a TL sensitivity 50–70% of the light clasts. The largest differences are shown by Weston, Leighton and Fayetteville, where the matrix/clast TL sensitivity ratios are 0.2–0.4. The "diffuse clast" is a region of dark matrix which contained several small fragments of light clast, but was otherwise like much of the dark matrix of Fayetteville (E.R.D. SCOTT, pers. commun.; see also SCHWARZ and SEARS, 1988). Its TL sensitivity is the same as the other matrix samples in the present study, and possibly it is a clast of "matrix-within-matrix". Split number 26 (clast d) is a ~1 cm-wide, dark region of matrix which was found to be coarsely crystalline in thin section (R. T. DODD, pers. commun.). The low TL sensitivity of this material is consistent with melting of the feldspar.

The TL peak temperature and peak width data are shown in Fig. 2. For Leighton, Fayetteville and Weston (Fig. 2a), there is a significant difference between the dark matrix and light clasts in the clustering of these data. The matrix samples are generally ~10°C lower in peak temperature than the

Table 1. Induced thermoluminescence properties of matrix and clast samples from six breccias.*

Meteorite†	Split Number+	Mass (mg)	TL Sens. (Dhajala=1)	Peak Temp. (°C)	FWHM‡ (°C)	Description
Fayetteville						
	28(91)	55	1.6±0.2	183±3	153±2	Matrix
	17(108)	58	1.5±0.2	193±2	144±2	Matrix b
	18(126)	66	1.5±0.2	182±3	154±2	Matrix
	26(113)	31	0.3±0.2	173±2	153±2	Dark clast d
	39(175)	62	2.3±0.2	183±2	152±2	Matrix
	33(80)	81	1.5±0.2	182±2	150±2	Matrix
	5(76)	59	1.4±0.3	178±2	148±3	Matrix
	31(134)	107	2.2±0.1	183±2	154±2	Matrix
	22(67)	54	2.2±0.1	181±2	152±2	Matrix
	7(162)	112	1.8±0.1	186±2	152±2	Matrix
	37(154)	35	1.6±0.2	188±3	152±1	Diffuse clast k
	6(189)	18	6.4±0.2	188±5	150±2	Clast ak
	19(19)	50	7.8±0.1	183±5	144±2	Clast z
	8(98)	59	7.5±0.2	188±2	152±3	Clast i
	20(186)	54	5.8±0.2	183±2	142±2	Clast h
	14(179)	104	5.8±0.1	198±3	145±2	Clast g
	32(115)	56	2.8±0.2	185±2	145±2	Clast bc/j
	50(177)	57	3.8±0.2	187±2	147±2	Clast n
Cangas de Onis		86	2.46±0.08	184±2	144±2	Matrix
		100	5.06±0.05	182±2	142±2	Clast
Leighton						
	B-D1	~100	0.5±0.2	187±3	154±5	Matrix
	F-D0	~100	0.2±0.1	188±9	161±6	Matrix
	F-D1	~100	1.0±0.2	191±3	149±2	Matrix
	F-D2	~100	0.35±0.07	190±5	156±3	Matrix
	F-D3	~100	0.2±0.1	180±6	162±2	Matrix
	F-L0	~100	0.71±0.07	196±3	149±2	Clast
	F-L1	~100	2.24±0.08	195±5	150±2	Clast
	Light(1t)	~100	1.8±0.1	195±2	146±2	Clast
Pantar						
	D-1	~100	2.3±0.1	187±4	145±2	Matrix
	D-2	~100	2.7±0.2	190±7	143±2	Matrix
	D-3	~100	2.41±0.04	184±2	145±2	Matrix
	D-4	~100	2.0±0.2	188±7	143±2	Matrix
	D-5	~100	2.6±0.3	185±2	146±2	Matrix
	D-6	~100	2.2±0.3	190±14	144±2	Matrix
	L-1	~100	2.6±0.3	185±2	148±2	Clast
	L-2	~100	2.9±0.3	189±2	146±2	Clast
	L-3	~100	3.0±0.3	189±2	147±2	Clast
	L-4	~100	3.0±0.3	189±6	145±2	Clast
	L-5	~100	2.9±0.3	188±2	146±5	Clast
	L-6	~100	2.9±0.4	192±9	143±2	Clast
St. Mesmin						
	D-4	2000	4.11±0.02	191±6	154±2	Matrix
	D-5A	1630	5.4±0.1	188±3	148±2	Matrix
	D-7A	1200	3.3±0.3	188±5	154±2	Matrix
	368		6.78±0.06	188±2	156±2	Clast
Weston						
		420	1.1±0.2	189±3	150±2	Matrix
		1090	3.2±0.4	184±10	143±2	Clast

*Errors quoted are ±1 sigma based on replicate measurements of the same powder.
‡Full width of the peak at half its maximum intensity.
+In the case of Fayetteville, the "sub-split" number is given in parenthesis.
†See the following for details: Fayetteville, Schwarz and Sears (1987); Pantar, Haq (1987); Leighton, Bart and Lipschutz (1979); Cangas de Onis, Weston, St. Mesmin, Lipschutz et al. (1983).

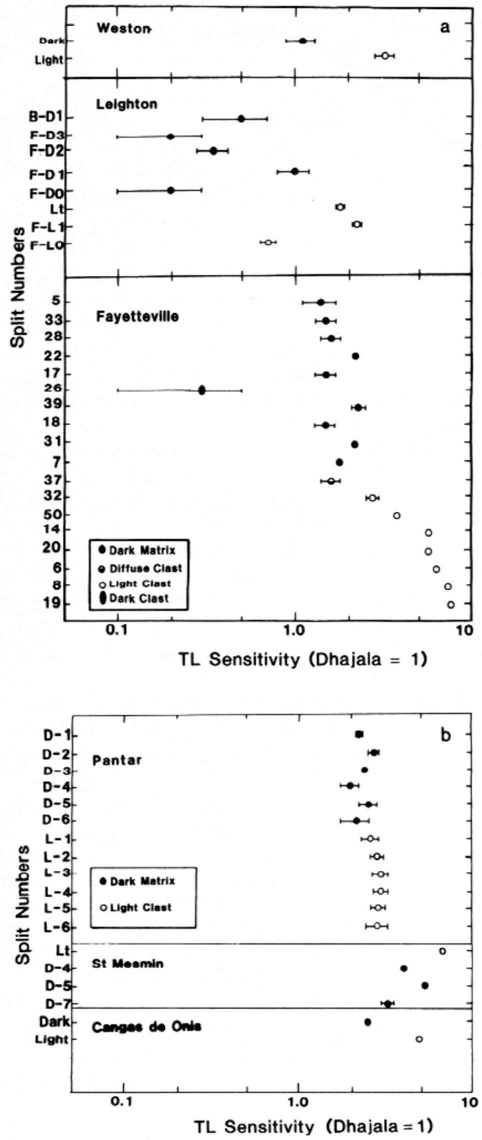

FIG. 1. Thermoluminescence (TL) sensitivity of the dark matrix and clasts from 6 regolith breccias, (a) three meteorites whose dark matrix has high inert gas contents (^{4}He $<10^5 \times 10^{-8}$ cc STP g^{-1}) and (b) three meteorites whose dark matrix has low inert gas contents. Error bars refer to ±1 sigma calculated from triplicate measurements, lack of error bar indicates uncertainties comparable with size of symbol. In all cases, the clasts have higher TL sensitivity than the matrix, this is especially true of the more gas-rich meteorites.

clast samples and have peaks which are usually ~10°C broader. The two Weston samples plot in the matrix and clast fields consistent with the other data, but the relative positions of the two points differs from that of the two clusters; however, in view of the scatter in the data and the size of the error bars, this is probably not significant. In contrast to the data in Fig. 2a, the clast samples from Pantar (Fig. 2b) do not appear to be significantly different in their peak temperatures and peak widths from matrix samples. The same is probably true of Cangas de Onis, although the data are meagre. The St. Mesmin samples have comparable peak temperatures to those of Cangas de Onis and Pantar, but 3 out of 4 of the samples have larger peak widths. The relative distribution of the St. Mesmin clasts and matrix samples on the peak temperature *vs.* peak width plot is unlike that of Fayetteville, Leighton and Weston. St. Mesmin is the only LL chondrite in the present suite, the others being H chondrites, but previous studies have not shown any systematic TL differences between L and H chondrites and in the present study St. Mesmin normally behaves in a way consistent with the others.

The new carbon data are shown in Table 2. Pantar, St. Mesmin and Plainview show little, if any, enhancement of

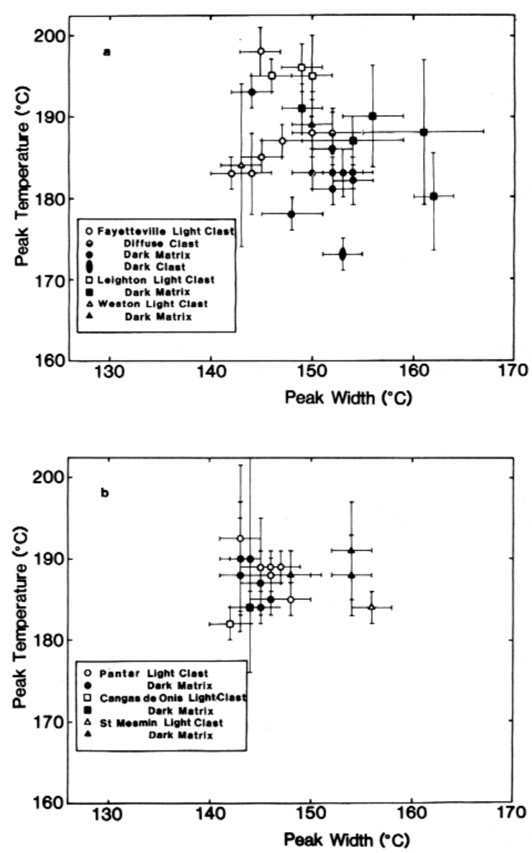

FIG. 2. TL peak temperature against peak width for 6 regolith breccias. (a) Three meteorites whose dark matrix contains high gas contents (^{4}He $> 10^5 \times 10^{-8}$ cc STP g^{-1}). Matrix samples tend to plot in a field displaced about 10°C towards lower peak temperatures and broader widths from the data for the clasts. (b) Three meteorites whose dark matrix contains relatively low levels of gases. Clast and matrix data plot in a single field, which is the same as the clast data in Fig. 2a. Error bars refer to ±1 sigma calculated from triplicate measurements.

Table 2. Total carbon content of gas-rich chondrites

Meteorite	Catalog/ Split Number	Mass (mg)	C* (wt.%)	Description
Fayetteville⁺	69	52.9	0.239	Matrix a
		60.0	0.232	
	81	51.6	0.173	Matrix
		53.7	0.180	
	99	51.4	0.284	Matrix
		46.8	0.277	
	181	52.7	0.073	Clast g
		51.6	0.072	
St. Mesmin	USNM 262	99.8	0.103	Matrix
		92.6	0.133	Clast
Plantar	ASU 503.3	108.4	0.084	Matrix
		112.5	0.082	Clast
Plainview	ASU 92.342	104.5	0.088	Matrix
		105.2	0.125	Clast
	ASU 92.342	103.8	0.125	Matrix
		103.3	0.175	Clast
Leighton	ASU 1192	102.6	0.820	Matrix
		31.8	0.240	Clast
Weston	ASU 2385b	103.0	0.250	Matrix
		101.8	0.124	Clast

⁺See Schwarz and Sears (1987) for sampling details.
*One sigma uncertainties are 0.005 wt % C.

C in the matrix compared to the clasts, while Fayetteville, Weston and especially, Leighton, show considerable C enhancement in the matrix.

DISCUSSION

TL sensitivity variations of the dark matrix

To a good approximation, the TL sensitivity of ordinary chondrites depends on the amount of feldspar in the sample. Minor differences can also be caused by albedo, darker substances have lower TL sensitivity, and the type of feldspar—high-temperature feldspar has a TL sensitivity 60% of the same feldspar in the low-form (GUIMON *et al.*, 1985). However, these effects are usually minor compared with differences in TL sensitivity caused by shock, which melts or converts the feldspar to maskelynite (SEARS, 1980), or metamorphism, which produces feldspar from primary igneous glass (SEARS *et al.*, 1980; GUIMON *et al.*, 1986).

The lower TL sensitivity of the dark matrix compared to the clasts more reasonably reflects the presence of a low TL sensitivity component, where the amount of this component varies from ~20% in the case of Pantar to ~75% in the case of Leighton. Their are several candidates for this low TL component, some being petrographically observed; they are material of low petrological type (BINNS, 1967; BART and LIPSCHUTZ, 1979), heavily shocked material (DODD, 1974; PELLAS, 1972) or grain-boundary, interstitial glass (BISCHOFF *et al.*, 1983; ASHWORTH and BARBER, 1976).

BISCHOFF *et al.* (1983) argued that the lithification process involved the formation of feldspathic, interstitial grain-boundary glass. The conversion of feldspar to glass certainly lowers its TL sensitivity, however this is not the major factor causing the range of TL sensitivities observed for the present matrix samples. Breccias of BISCHOFF *et al.*'s (1983) class A (minimal grain-boundary glass, *e.g.* Fayetteville and Weston) have the lowest TL sensitivities while those of class B (intermediate grain-boundary glass, *e.g.* St. Mesmin and Pantar) and class C (maximal grain-boundary glass, *e.g.* Cangas de Onis and Plainview) have relatively high TL sensitivity.

Peak temperature and width

Although the details of the mechanism are known in only a fragmentary way, it is clear the TL peak temperature and width of chondritic meteorites are governed by thermal history. For example, type 3.5–3.9 ordinary chondrites have broad peaks at high temperatures, while the type 3.2–3.4 ordinary chondrites have narrow peaks at low temperatures. Furthermore, peak temperatures and widths can be affected by annealing and formation temperature (GUIMON *et al.*, 1985, 1986, 1987; HARTMETZ and SEARS, 1987). The change in peak shape is indirectly associated with disordering the Al and Si atoms in the feldspar lattice, and mixtures of high and low feldspars are responsible for the variety of peak shapes observed (KECK *et al.*, 1986).

It is unusual for TL peak temperature and widths to cluster in the manner shown in Fig. 2a. Severely annealed chondrite samples may on occasion show a tendency for peaks to narrow and move to higher temperatures; in this case it is thought to reflect the formation of especially pure high form of the plagioclase (KECK *et al.*, 1986; GUIMON *et al.*, 1986; HAQ *et al.*, 1988). However, this cannot be the correct interpretation for the present data because the changes are in the opposite direction to those required. The only reasonable interpretation of the separation of clast and matrix data in Fig. 2a, based on existing data, is that there is a significant abundance of a component in the dark matrix which has a TL phosphor with a broad peak at low temperatures. There have been no TL studies of CM chondrites, which most closely resemble the dominant form of "contamination" of the dark matrix (*e.g.* WILKENING, 1976), but the Allende meteorite contains a number of phosphors that have the required TL properties, most notably gehlenite (GUIMON and SEARS, 1986), which is present in CM chondrites. This being so, it seems that while the dark matrix of Leighton and Fayetteville contain amounts of this component detectable by its TL properties, there is no TL evidence for its presence in Pantar, for which a comparable data base exists. The unequilibrated olivines occasionally observed in the dark matrix (*e.g.* Fa_5-Fa_{40} in Leighton according to (GRAHAM *et al.*, 1985, page 211) may also represent contamination with such unequilibrated CM-like material.

Origin and history of the dark matrix

By examining TL sensitivity as a function of regolith maturity it should be possible to distinguish between the idea that the dark matrix was formed by comminution of the clasts with "contamination" by volatile material from the idea that the matrix was originally primitive material. Low petrologic type chondrites have TL sensitivities $<10^{-3}$ times those of equilibrated, unshocked chondrites, while heavily shocked chondrites have TL sensitivities 0.1–0.01 times equilibrated, unshocked chondrites. Thus, if low TL sensitivity reflects a large component of shocked material, then one might expect the TL sensitivity of the dark matrix to decrease with regolith maturity as shocked clast material is added to the matrix. On the other hand, if the low TL sensitivity component is little-metamorphosed (*i.e.* type 3) ma-

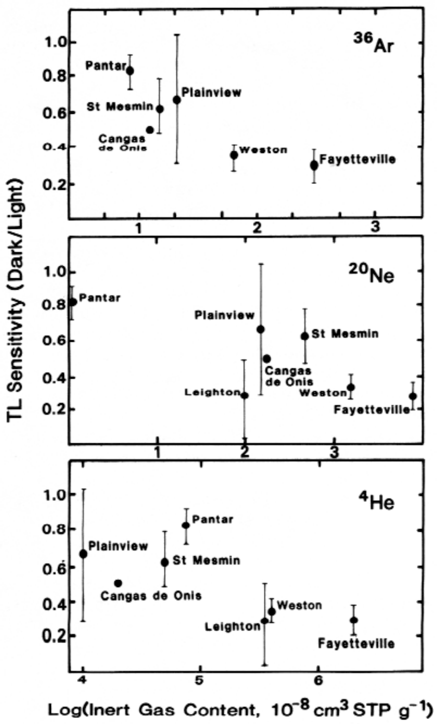

FIG. 3. The ratio of the TL sensitivity of the dark matrix to that of the light clasts plotted against three solar wind implanted isotopes; ^{4}He, ^{20}Ne, ^{36}Ar. In each case, the TL ratio decreases with increasing inert gas content. (Plainview TL data from SEARS, 1981, inert gas data from compilation by KRUSE and SCHULTZ, 1983).

terial, then its TL sensitivity will even increase with regolith maturity as shocked clast material is added to the matrix.

Figure 3 compares the ratio of the TL sensitivity for the matrix to that of the clasts with the contents of three inert gas isotopes abundant in the solar wind. For each isotope, there is a relationship between the TL sensitivity of the dark matrix and gas content; while the TL sensitivity decreases by a factor of $\sim$4, the content of solar wind gas increases by more than two orders of magnitude. A similar relationship probably exists between TL sensitivity and solar flare track density, since grains from the dark matrix of Fayetteville show a surface solar flare track density of $\sim 10^{11}$ cm^{-2}, compared with $\sim 10^{9}$ cm^{-2} more typical of gas-rich regolith breccias (BARBER *et al.*, 1971; PELLAS, 1972). The highly volatile elements, In, Tl and Bi, and non-volatile carbon, also show a relationship with TL sensitivity of the dark matrix; the $\sim$4-fold decrease in TL sensitivity is accompanied by $>$100-fold increases in In, Tl, and Bi and a 3-fold increase in carbon content (Fig. 4).

If solar wind gas content, solar flare track density, volatile element content, and carbon content are measures of regolith maturity, then the inverse relationship between these quantities and the TL sensitivity ratio for the matrix and clasts suggests that the matrix is predominantly comminuted clast, rather than primitive material.

If, on the other hand, solar wind gas content reflects the extent of gas-loss during regolith activity, as suggested by PELLAS (1972) and BISCHOFF *et al.* (1983), then presumably the variations in track density, carbon and volatile element content, and TL sensitivity also reflect element-loss or annealing during regolith activity. But this is not consistent with laboratory data on the loss of volatile elements during heating or annealing of charged particle tracks; temperatures and times sufficient to cause loss of In, Tl and Bi would have totally removed charged-particle tracks (IKRAMUDDIN *et al.*, 1977; PELLAS and STORZER, 1976). Similarly, it is difficult to imagine elemental carbon, which is refractory, being lost during regolith processes, and we would presume its removal would involve the formation of CH_4. However, the removal rates would be different for the noble gases, C, Tl and In.

It is possible that, while inert gas content reflects gas-loss duration lithification, tracks, carbon and volatile elements predominantly reflect regolith maturity. Other minor processes may have occurred on a high localised scale; for example, Weston shows traces of terrestrial or extraterrestrial weathering (ASHWORTH and HUTCHISON, 1975). In view of the correlations between inert gases and the other data, we think it most plausible that although element redistribution and track annealing may have occurred during regolith activity on a highly localised (*i.e.* individual grain or small clast) basis, these and other secondary processes did not dominate in determining composition, track density and TL sensitivity of the matrix as a whole. The amount and texture of interstitial, grain-boundary glass will depend on several factors besides regolith processing, including degree of compaction prior to lithification, burial depth and petrologic type of the precursor dust. Post-compaction recrystallization could also have been important (ASHWORTH and BARBER, 1976). In fact, there is probably competition between the formation of large contiguous networks of interstitial glass (which is associated with the degree of consolidation) and the disruption and dispersion of such glass during regolith processing. The tendency for the most gas-rich regolith breccias to have least interstitial glass, and be least well consolidated, could well

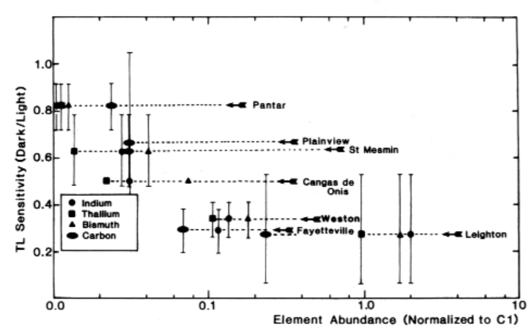

FIG. 4. Ratio of the TL sensitivity of the dark matrix to that of the light clasts against CI-normalized abundances of three highly volatile elements and carbon. The elemental abundances increase with decreasing matrix/clast TL sensitivity ratio in a manner similar to that of solar wind gases (Fig. 3). (Data from BART and LIPSCHUTZ, 1979; LIPSCHUTZ *et al.*, 1983; RIEDER and WÄNKE, 1969; CHOU *et al.*, 1981; ANDERS and EBIHARA, 1982; and the present work). Pantar is apparently a mixture of breccia types and this is reflected in the trace element data. The data plotted are for ASU 503.3, the fragment used in the present study.

reflect an *extended* period of regolith activity. We conclude that the dark matrix formed by comminution of the clasts with the addition of a component approximately resembling CM chondrites. The range of properties observed reflect, predominantly, the duration of regolith processing.

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