

# Thermoluminescence of Meteorites: Relationships with Their K-Ar Age and Their Shock and Reheating History

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The observations of G. F. Komovsky [*Meteoritika* 21 (1961), 64-69] and A. Liener and J. Geiss (in *Thermoluminescence of Geological Materials*, Academic Press, New York, 1968), that the thermoluminescence (TL) sensitivity of meteorites correlates with their K-Ar age, have been confirmed using a suite of 22 ordinary chondrites. In order to interpret this observation, meteorite samples have been exposed to doses of  $\alpha$ ,  $\beta$  and  $\gamma$  radiation comparable with those experienced over the lifetime of the meteorites and given a dose of protons comparable to the total dose received from cosmic rays. There was no increase in TL sensitivity after these treatments, suggesting that, contrary to the ideas of earlier workers, the TL mechanism does not involve radiation damage. The TL sensitivity of meteorites is therefore time independent. On the other hand, samples of meteorite annealed in a furnace at temperatures between 450 and 1250°C for 1 hr suffered up to an order-of-magnitude decrease in TL sensitivity. Similarly, samples of meteorite artificially shocked to pressures of the order of 400 kbar suffered a comparable decrease in TL sensitivity. It is concluded that the correlation between TL sensitivity and K-Ar age is entirely a result of the low K-Ar age meteorites being shocked or reheated. Data on the thermal and mechanical histories of these meteorites, based on <sup>40</sup>Ar-<sup>39</sup>Ar, metallographic, and X-ray diffraction studies, seem to be consistent with this finding.

## 1. INTRODUCTION

Events which lead to shock loading and reheating seem to have played an important part in the history of many meteorites, and are central to models for the origin of at least one major class (Heymann, 1967; Anders, 1978). According to Anders and Heymann, the *L* chondrites were ejected from the Mars-crossing asteroid orbits—in which they were formed—into an Earth-crossing orbit, by an event which caused at least one-third of this class to be severely shocked and reheated. Such meteorites frequently have K-Ar and U, Th-He ages around 500 Ma, which is the postulated age of the event. Better assessments of this age have been made using the <sup>40</sup>Ar-<sup>39</sup>Ar method (Turner, 1969), and this technique also indicates that *H* chondrites have been

involved in similar major degassing events (Bogard *et al.*, 1976).

The shock and reheating effects in stony meteorites have been studied using their metallographic (Buseck *et al.*, 1966; Heymann, 1967; Wood, 1967; Begemann and Wlotzka, 1969; Taylor and Heymann, 1969, 1970, 1971; Smith and Goldstein, 1977), X-ray diffraction (Carter *et al.*, 1968), and macroscopic properties (Fredriksson *et al.*, 1963). Komovsky (1961) and Liener and Geiss (1968) also observed that thermoluminescence sensitivity—the thermoluminescence (TL) induced in a sample by a standard laboratory test dose—correlates with K-Ar age, and Liener and Geiss (1968) proposed two explanations for this observation. One was that the shock and reheating event which lowered the K-Ar age in some meteorites also reduced their TL sensitivity. This being so, TL would also be related in some manner to this important aspect of meteorite history.

Thermoluminescence (TL) is the light

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emitted by a substance as it is heated and before it becomes incandescent. Ionizing radiations which pass through the substance deposit energy and this is absorbed by Valence Band electrons. They are consequently promoted to the Conduction Band and move freely through the lattice until they locate "traps." These are sites in the lattice—usually about 1 eV in energy below the Conduction Band—where the electron is stored until dislodged by heating the sample. Upon heating, they may find their way back to the Valence Band and when this involves a transition which emits visible light, they are said to have found a "luminescence center."

The nature of electron traps and luminescence centers in meteorites is unclear. The TL phosphor in meteorites is predominantly feldspar (Lalou *et al.*, 1970; Sears, 1974), and in all of the terrestrial Ca-bearing minerals examined by Medlin (1968) the TL system was  $Mn^{2+}$  substitution for  $Ca^{2+}$ ; the lattice strain constitutes the trap while the outer orbitals of  $Mn^{2+}$  provide the luminescence center. Similarly, the cathodoluminescence of lunar samples—in which the phosphor is also feldspar—is considerably increased by  $Mn^{2+}$  doping (Geake *et al.*, 1977). However, there is no simple relationship between Ca (and presumably therefore Mn) content and TL in feldspars, and the TL properties of feldspars from different sources vary considerably (Sears, 1974; Wintle, 1973), so these data are probably only suggestive. On the other hand, many phosphors contain electron traps which are due to atom displacements. These may be caused by radiation damage, and by subjecting TL phosphors to radiations of various energies, it is possible to determine threshold energies for the atom displacement in question (Bryant and Hamid, 1970; Levy, 1968). While either of these would seem to be plausible possibilities for meteorite TL, there are very few available data on the subject.

Liener and Geiss (1968) therefore proposed, as a second explanation for their

observed correlation between TL sensitivity and K—Ar age, that TL sensitivity is a time-dependent quantity. Such would be the case if the traps were produced by radiation damage and TL sensitivity was simply a measure of the number of traps. This explanation is not tenable in this simple form because it requires that meteorites with low K—Ar age and TL sensitivity actually formed recently, whereas in fact other chronological data (i.e., Rb/Sr data and I—Xe formation intervals) show that most ordinary chondrites formed within 20 Ma or so of each other, 4.6 Ga ago (Sears, 1978, Sections 5.2.2 and 5.2.5). However, it is perfectly reasonable that this explanation and the earlier proposal—that TL sensitivity is lowered by shock and reheating—were both operative. The resulting, complex situation would then be very similar to that for the K—Ar system.

In the present work, I have confirmed the existence of a correlation between TL sensitivity and K—Ar age, and I have performed several laboratory experiments aimed at providing an interpretation for this observation. Samples of meteorite have been artificially shock loaded, and others have been annealed in furnaces. After each of these treatments a decrease in TL sensitivity was observed. Samples have also been irradiated with doses of radiation comparable with those received by the meteorite over its entire life span, but no resulting increase in TL sensitivity was observed. It therefore seems that the shock and reheating hypothesis alone explains the correlation between TL sensitivity and K—Ar age. A detailed comparison between the TL sensitivity of the present specimens and literature data on their shock and reheating histories seems to be consistent with this interpretation.

## 2. METHODS AND SAMPLES

### 2.1. Thermoluminescence Measurements

The techniques and apparatus for measuring meteorite TL have been described in

great detail by Mills *et al.* (1977) and only a brief outline is necessary here. The specimen is heated in an oxygen-free, nitrogen atmosphere on a nichrome strip whose dimensions are  $5 \times 1 \times 0.0125$  cm. A linear heating rate of  $5^\circ\text{C sec}^{-1}$  is produced by an electronic control unit, and the temperature is monitored by a chromel–alumel thermocouple, which feeds both the control unit and the X axis of an X–Y plotter. The TL is measured by an EMI 9804QB photomultiplier tube in front of which is an Ilford 621 instrument filter and a Chance HA3 heat filter to suppress the blackbody radiation and prevent damage to the PMT. The signal is amplified by a Keithley electrometer and fed to the Y axis of the X–Y plotter. The glow curve, a plot of TL against temperature, is thereby obtained directly (Fig. 1). Most of the TL from meteorites is emitted between 150 and  $250^\circ\text{C}$ , and the sensitivity values referred to here are measurements of the maximum intensity in this region. The laboratory test dose was 50 krad of  $^{60}\text{Co}$   $\gamma$  rays. Each measurement was made several times on at least three samples. A detailed study of different ways of measuring the TL sensitivity of meteorites has

been made by Sears and McKeever (1980), who conclude that this is the most suitable method where many replicate measurements on the same sample are needed. The size of the test dose used is important and has to be carefully chosen.

The way in which the samples are prepared for these measurements has also been considered carefully. Liener and Geiss (1968) used 10 mg of  $50\text{-}\mu\text{m}$ -sieved powder, from which the magnetic material had been removed with a hand magnet. Five specimens are common to the present study and that of Liener and Geiss (1968) and our results are compared in Table 1. The precision on both sets of data is thought to be  $\pm 5\%$ , but clearly our accuracy is lower; the maximum deviation about the (normalized) mean is 70% (Tieschitz), while the mean deviation is 25%. This is probably a consequence of small differences in the extent of rusting in the fragments examined. Small amounts of rust, which are present even in observed falls, are sufficient to reduce the TL by an order of magnitude in particularly bad cases. Samples have therefore also been prepared as fine-grained disks. These consist of about 1 mg of  $\leq 10\text{-}\mu\text{m}$  grains deposited onto a 1-cm-diameter aluminum disk from acetone suspension (Sears, 1978b). The grain size is controlled by collecting only those grains which fall after 2 min. For reasons which are discussed below, the presence of small amounts of rust does not affect measurements of disks, but as a double precaution the powders were shaken for a few minutes in 10 M HCl and then washed twice in distilled water and twice in acetone. Disks have the advantage of being less susceptible to differences in albedo and friability of the meteorites. A priori, it is unclear whether the smaller-grain-sized material on these disks is more, or less, representative of the bulk powder than, say, a 10-mg aliquot measured in the normal way. The excellent agreement between low-albedo samples measured as powder and those measured as disks (see

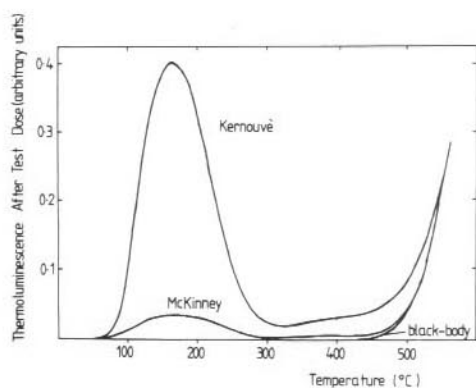


FIG. 1. Plots of TL against temperature (glow curves) for two meteorites with different K–Ar ages. Kernouvé has a K–Ar age of 4.47 Ga and McKinney is a black chondrite with a K–Ar age of 2.15 Ga. The TL is that induced by a standard laboratory test dose of 50 krad in samples which had previously been heated to  $500^\circ\text{C}$  to remove their natural TL. It is usually referred to as “TL sensitivity.”

TABLE I

COMPARISON BETWEEN THE PRESENT DATA FOR MEASUREMENT ON POWDERS AND THOSE OF LIENER AND GEISS (1968).<sup>a</sup>

	Liener and Geiss (1968) ( $\mu\text{m}$ )	Present work (arbitrary units)	Normalized to Appley Bridge	
			Liener and Geiss (1968)	Present work
Appley Bridge	620	4.30	1.00	1.00
Olivenza	266	2.80	0.43	0.65
Bruderheim	190	1.10	0.31	0.26
Tieschitz	48	0.08	0.08	0.02
Farmington	24	0.17	0.04	0.04

<sup>a</sup> Both sets of data are TL sensitivities using a 50-krad test dose and 10 mg of 50- $\mu\text{m}$ -sieved powders.

below) suggests that they are probably equally representative. However, the reproducibility of measurements on disks is much poorer than that for measurements on powder and the greatest care is necessary in their preparation. Table II lists measurements on fine-grained disks prepared from three samples of the Kernouvé (H6) chondrite. The standard deviation is 13% and, unless otherwise stated, this is the error assigned to the measurements.

## 2.2. Samples

The samples used in this work are listed with their sources in Table III. They were chosen because  $^{40}\text{Ar}$ - $^{39}\text{Ar}$  data existed for them, or because there were data in the literature to suggest they had suffered severe reheating or shock. I have also included Allan Hills A77214, because a fragment at hand resembles a black chondrite. Potassium-argon and U, Th-He ages for

these specimens have been calculated using the compilation of inert gas contents in meteorites by Schultz and Kruse (1978). Decay constants and the assumptions necessary for the calculations are given in footnotes to the table. Potassium-argon and U, Th-He ages for Barratta, Farmington, Lubbock, McKinney, and Wickenberg (stone) were also calculated by Heymann (1967), and the agreement between his estimates and those given here is very good.

Thirteen of the present specimens have been examined by the  $^{40}\text{Ar}$ - $^{39}\text{Ar}$  technique (Turner, 1969, 1975; Turner *et al.*, 1978; Bogard *et al.*, 1976). Six of them have good 4.6-aeon plateaus, indicating little or no disturbance since the onset of Ar retention. Mangwendi and Appley Bridge suffered early degassing and give  $^{40}\text{Ar}$ - $^{39}\text{Ar}$  ages around 4.0 aeons, while Ambapur Nagla, and Allegan have poorly determined ages

TABLE II

REPEATABILITY IN TL SENSITIVITY MEASUREMENTS ON THE KERNOUVÉ H5 CHONDRITE FOR SAMPLES PREPARED AS FINE-GRAINED DISKS.\*

Disk	J18	J19	J20	K18	K19	K20	L18	L19	L20
Measurement 1 <sup>a</sup>	0.42	0.44	0.32	0.41	0.47	0.43	0.42	0.72	0.43
Measurement 2	0.32	0.42	0.33	0.48	0.47	—	0.41	—	—

\* Mean, 0.41; SD, 0.05 (13%).

<sup>a</sup> Measurements made 4 months apart. Missing values from measurement 2 occur where the samples were being used in other long-term experiments.

TABLE III  
SAMPLES USED IN THE PRESENT STUDY AND SOME LITERATURE DATA

Meteorite <sup>a</sup>	Class	Source and Catalogue No. <sup>b</sup>	<sup>3</sup> He <sup>c</sup> (10 <sup>-8</sup> cm <sup>3</sup> STP/g)	<sup>4</sup> He <sup>c</sup> (10 <sup>-8</sup> cm <sup>3</sup> STP/g)	<sup>40</sup> Ar <sup>c</sup>	Ca (wt%)	K (wt%)	Ref. <sup>d</sup> (Ga) <sup>e</sup>	K-Ar age (Ga) <sup>f</sup>	U, Th-He age (Ga) <sup>g</sup>	<sup>40</sup> Ar- <sup>39</sup> Ar age		Ref. to data on shock and reheating <sup>h,i</sup>
											(Ga)	Ref. <sup>d</sup>	
Allan Hills A77214	L or LL	MWG (NSF)											
Allegan	H5	BM1920,281	9.0	1625	5700	{ 1.180 1.201	0.075 0.077	(o) (m)	4.53 4.48	5.00	~4.6	(b)	
Ambapur Nagla	H5	BM81117	6.85	138	1850				2.64	0.44	>4.39	(c)	
Apple Bridge	LL6		180	887	4310	1.07	0.10	(n)	3.58	3.10	3.19-4.1	(b)	
Barratta	L4	BM1929,1284	14.24	188	340	1.10	0.09	(n)	0.76	0.48	0.47 ± .05	(e)	(f, g)
Barwell	L5	BM1966,57	5.65	1068	5140	1.29	0.10	(p)	3.87	3.70	4.45 ± .03		
						1.29	0.17	(r)	1.28				
Bruderheim	L6		48.9	530	1251	1.24	0.11	(s)	1.73	1.10			(f, h)
						1.27	0.10	(q)	1.84				
Butsura	H6	BM34795	58.5	1975	4390				3.98	5.20	4.48 ± .03	(a)	
Farmington	L5	BM67016	0.06	145	341	{ 1.36 1.315	0.11 0.083	(n) (m)	0.65 0.82	0.60			(f, g, i, j)
Kernouvé	H6	BM43400	4.88	1070	4430(a)				4.47(a)	3.70	4.46 ± .05	(a)	
Kingfisher	L	HES	24.85	216	184				0.50	0.36			(f, g, k)
Lubbock	L	BM1959,887	19.0	192	690				1.43	0.38			(j, k, l)
Mangwendi	LL5		26.0	900	5050				4.21	2.80	4.0-4.1	(b)	
McKinney	L4	HES	5.53	120	1285	{ 1.27 1.258	(0.18) 0.079	(n) (m)	1.25 2.16	0.39			(f, g)
Monroe	H4	BM25462	18.50	440	1290				2.15	1.30	1.1 ± 0.2	(d)	
Olivenza	LL5		12.25	1168	5620				4.39	3.80	4.49 ± .03	(a)	(g)
Saratov	L4	BM1956,169	33.0	1593	3930				3.80	4.60	4.44 ± .03	(a)	
Tenham	L6	BM1935,792											(n, m)
Tieschitz	H3	BM1975,M11	46.8	1623	2280				2.94	4.50			
Uccia	H	JEV	10.05	256	271				0.80	0.80			
Wickenburg (stone)	L5	BM1959,1023	3.05	56	900				1.72	0.17	4.45 ± .05	(a)	(f, g, j, o)
Zomba	L6	BM84357	26.7	462	820				1.61	1.30	0.44 ± .05	(e)	

<sup>a</sup> Finds underlined

<sup>b</sup> MWG (NSF)—Meteorite Working Group of the U.S. National Science Foundation, BM—Dr. R. Hutchison, British Museum, Natural History, London. HES—Professor H. E. Suess and Dr. M. Herndon, University of California, San Diego. JEV—Dr. J. E. Vaz, Instituto Venezolano de Investigaciones Científicas, Caracas, Venezuela.

<sup>c</sup> Means for bulk meteorite from the compilation by Schultz and Kruse (1978).

<sup>d</sup> References: (a) Turner *et al.*, 1968. (b) Turner, 1975. (c) Knight, personal communication. (d) Bogard *et al.*, 1976. (e) Turner, 1969. (f) Heymann, 1967. (g) Hey, 1966. (h) Carter *et al.*, 1968. (i) Wood, 1967. (j) Smith and Goldstein, 1977. (k) Taylor and Heymann, 1970. (l) Waldschmidt, 1940. (m) Von Michaelis *et al.*, 1969. (n) Wilk, 1969. (o) Jarosewich and Mason, 1969. (p) Jobbins *et al.*, 1966. (q) Duke *et al.*, 1961. (r) König, 1964. (s) Baadsgaard *et al.*, 1961.

<sup>e</sup> Calculated from the <sup>40</sup>Ar and K values listed or assuming a K content of 800 ppm, a <sup>40</sup>K abundance of 0.012% of total K,  $t_{1/2}$  (<sup>40</sup>Ar) equal to  $5.30 \times 10^{-10}$  year<sup>-1</sup>, a branching ratio of 0.110, and all <sup>40</sup>Ar radiogenic.

<sup>f</sup> Calculated from listed <sup>3</sup>He and <sup>4</sup>He data and assuming a U content of 11 ppb,  $t_{1/2}$  (<sup>238</sup>U) =  $1.525 \times 10^{-10}$  year<sup>-1</sup>,  $t_{1/2}$  (<sup>235</sup>U) =  $9.848 \times 10^{-10}$  year<sup>-1</sup>,  $t_{1/2}$  (<sup>232</sup>Th) =  $4.984 \times 10^{-11}$  year<sup>-1</sup>, Th/U = 3.7,  $^{235}\text{U}/^{238}\text{U}$  = 140 and cosmogenic <sup>4</sup>He/<sup>3</sup>He = 5.2.

<sup>g</sup> See Appendix 1 for details.



between 4.0 and 4.6 aeons. Barratta and Zomba have ages near 0.45 aeons, while Monroe has a 1.1-aeon age. The 10 specimens which were chosen because there were data on their shock and reheating history are described in Appendix 1.

### 2.3. Radiation Sources and Procedures

A  $^{60}\text{Co}$   $\gamma$ -ray source in the University of Birmingham Radiation Centre was used for administering a standard 50-krad test dose (dose rate  $7.5 \times 10^4$  rad/hr) and the high-dose  $\gamma$  irradiations (dose rate  $1.4 \times 10^6$  rad/hr). A  $^{90}\text{Sr}$  source delivering about  $6 \times 10^4$  rad/hr was used for the  $\beta$  irradiations. The Nuffield cyclotron in the Physics Department was used to perform 40-MeV/nucleon  $\alpha$ -particle and 10-MeV proton irradiations.

The procedures for determining the effect of high-radiation doses on TL sensitivity were much the same for all radiations, except that powders were used for  $\gamma$  irradiations. This was found appropriate in view of source geometries and particle ranges. For each type of radiation, nine samples of the Kernouvé meteorite were drained of their natural TL by heating to 500°C, and the TL induced by the test dose was measured ( $S_1$ ). They were then given a range of selected radiation doses and then heated to 500°C again to remove the induced TL. The same test dose was administered a second time and the induced TL measured ( $S_2$ ). The ratio of TL sensitivity after and before irradiation was expressed as the normalized TL ratio ( $S_2/S_1$ ).

During the proton irradiation experiments, some samples were not thermally drained, but instead were exposed to 365-nm uv radiation for 6 weeks. After this exposure, the TL induced by the proton irradiation was reduced to a few percent of the natural level, and the samples could be considered drained. The samples were then given the same test dose as before to enable a comparison to be made of uv bleaching and thermal draining after the heavy irradiation.

In this way it was shown that the thermal draining, after the heavy irradiation, was not removing any possible radiation damage.

### 2.4. Annealing Experiments

Fifty-micrometer sieved powders of the Kernouvé meteorite were annealed in wire-round tube furnaces for 60 min at a range of temperatures between 400 and 1250°C. Some samples were annealed in a hydrogen atmosphere (at 1 bar pressure) in a specially constructed hydrogen furnace in the Industrial Metallurgy Department, University of Birmingham. Other samples were sealed in quartz vials at a pressure of less than  $10^{-3}$  mbar. For TL measurement, the powders were usually prepared as fine-grained disks, although in a few cases 10 mg of powder was used for the measurements for comparison.

### 2.5. Shock Experiments

Six disks, 2 cm in diameter and 0.5 cm thick, of the Kernouvé meteorite were vacuum sealed in cylindrical blocks of aluminum alloy (HE 30) measuring 7.5 cm in diameter by 7.5 cm in length. The sealant was an araldite resin and the sealing pressure was just above that required to boil the araldite at room temperature, probably about 0.1 mbar. The six sample blocks, together with two "dummies" containing concrete disks, were sent to K. D. Burrows and R. L. Gyton of the Atomic Weapons Research Establishment, Foulness, for shock loading. A plane-wave-geometry, flying-plate technique, similar to that described by Stöffler (1972) but without a momentum trap, was used to shock load the specimens. The shock work will be described more fully elsewhere.

## 3. RESULTS AND DISCUSSION

### 3.1. Observations

Figure 2 presents the data of Liener and Geiss (1968) showing the relationship between TL sensitivity and K-Ar age. These

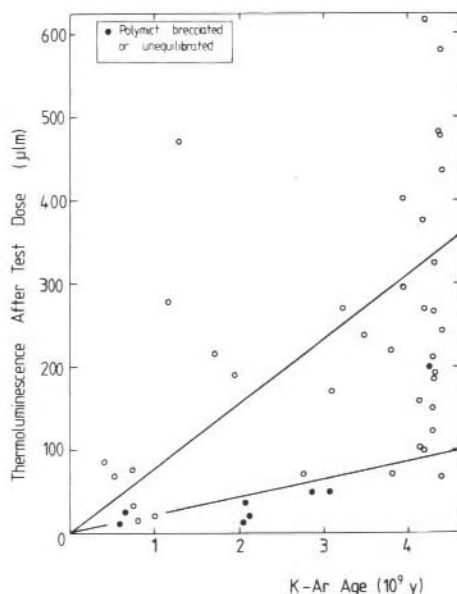


FIG. 2. Plot of TL after a 50-krad  $\beta$ -particle test dose against K-Ar age. Both sets of data are from Liener and Geiss (1968). These authors proposed that there were two relationships between TL and K-Ar age, one involving polymict brecciated and unequilibrated chondrites (the lower diagonal line), and another involving "ordinary" chondrites (the upper diagonal).

authors interpreted the data in terms of two relationships which are indicated by the two diagonal lines. The lower is a regression line for polymict brecciated and unequilibrated chondrites, and the upper line refers to the other chondrites. In Fig. 3, I have replotted the data by putting TL sensitivity on a logarithmic scale and updating the K-Ar ages, using the assumptions and new data sources listed in Table III (see Appendix 2). It seems that while polymict brecciated meteorites—these are also listed in Appendix 2—lie on the lower limit of the observed range of TL sensitivity values for a given K-Ar age, they do not produce a significantly different population. There are only two unequilibrated chondrites in the Liener and Geiss (1968) suite of specimens but, as discussed below, there is good evidence that unequilibrated chondrites have low TL sensitivities. More important for our present purposes, there is a statistically significant correlation between the TL sen-

sitivity and K-Ar age. The data in Fig. 3 yield a regression line with a correlation coefficient of 0.45 (significant at the 99% level), while the largest ordinary chondrite group (the *L* chondrites) have an even more significant regression line with a correlation coefficient of 0.70.

Table IV lists the measured TL sensitivities of the 22 meteorites in the present study. To facilitate comparison with other data, the meteorites are listed in order of decreasing TL sensitivity. I report both the TL sensitivities obtained when the samples are prepared as fine-grained disks, and those obtained when the samples are 10 mg of 50- $\mu$ m-sieved powder. It is enlightening to compare the two sets of measurements (Fig. 4). Meteorites with low TL sensitivity ( $\leq 0.5$ ) measured as disks differ markedly from those measured as powders. Measurements on powders are subject to large errors owing to the presence of rust, probably because rust lowers the albedo of a sample (Sears, 1978b). Essentially, meteorites with high albedos produce an excess of light when measured as powders, because TL from subsurface grains can find its way to the surface by multiple reflection from other grains (Fig. 5). Samples with low albedos—most of the meteorites with TL sensitivities  $\leq 0.5$  are dark; some are well known as "black chondrites"—only give detectable TL from surface grains when measured as powders and, for a given aperture size, the TL intensity will be the same for powders and disks. TL sensitivities measured on fine-grained disks are generally the more meaningful. Because of the strong albedo effect caused by rust, I omitted finds when plotting Figs. 2 and 3 since the measurements were made on powders.

The TL sensitivities measured in the present study, and using the fine-grained disk data, are compared with K-Ar ages in Fig. 6. These data also display a statistically significant correlation ( $r = 0.75$ ,  $n = 17$ ), confirming Komovsky's (1961) and Liener and Geiss' (1968) observations. According to Heymann (1967), meteorites

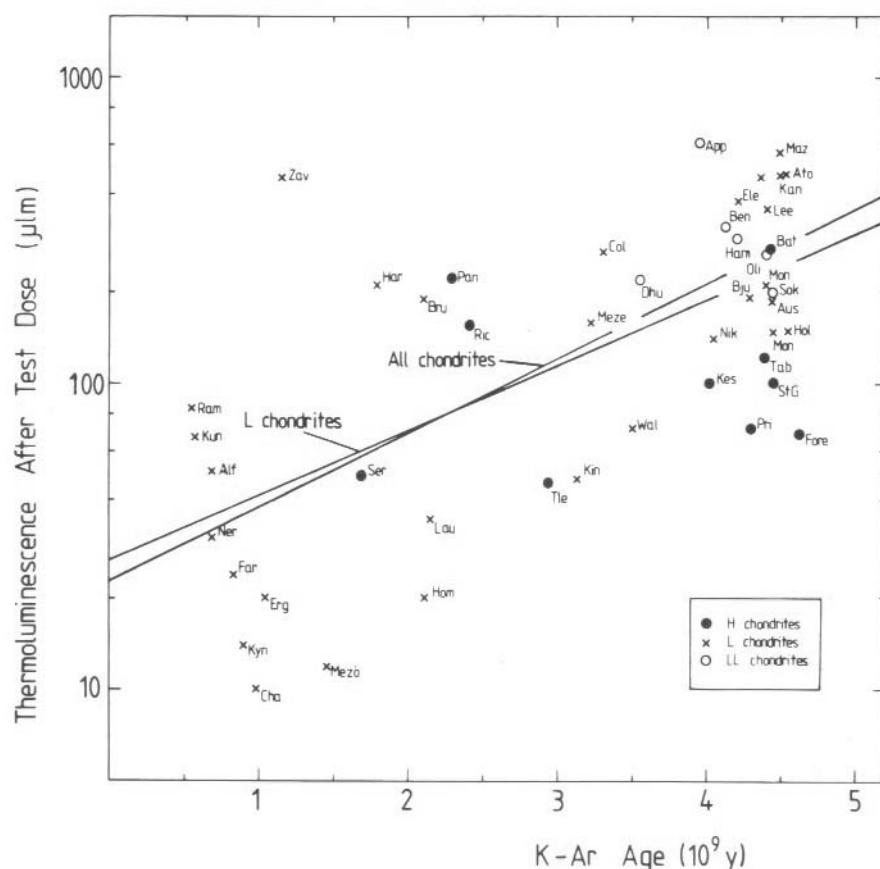


FIG. 3. Plot of TL after a 50-krad  $\beta$ -particle test dose against K-Ar age using a logarithmic TL scale and updated K-Ar ages. The points are labeled with the first three or four letters of the meteorite name, which may be identified from the data tabulation in Appendix 2. There now appears to be a single relationship between the two parameters. The correlation has a high statistical significance for all the chondrites plotted ( $r = 0.45$ ,  $n = 46$ ) and for the L chondrites considered as a subset ( $r = 0.70$ ,  $n = 29$ ).

with low K-Ar age tend to be heavily shocked, so these data have been examined in the light of evidence for shock and reheating. To help in this, in Table IV I have assigned the meteorites a ranking order; details are given in Appendix 1. Meteorites with good 4.6-Ga  $^{40}\text{Ar}/^{39}\text{Ar}$  plateaus are believed to have suffered no major shock or reheating, as this would have led to degassing. Such meteorites, assigned a 1 in the ranking order, have a remarkably uniform TL sensitivity; their mean disk value is 0.53 with a standard deviation of 0.05 (9.4%), while their median value is 0.4. Allegan and Olivenza have TL sensitivities a factor of 2

or so higher than the mean. There seems to be no reasonable explanation for this other than sample inhomogeneity. Saratov and Tieschitz, however, have particularly low TL sensitivity for meteorites of this kind.

Tieschitz does not produce a good  $^{40}\text{Ar}/^{39}\text{Ar}$  plateau, although its  $^{40}\text{Ar}/^{39}\text{Ar}$  age is  $4.45 \pm 0.05$  yr and it was assigned a 1 in Table IV. The  $^{40}\text{Ar}/^{39}\text{Ar}$  release pattern in Tieschitz, and those in most petrologic type 3 chondrites, is unusual and is believed by Turner *et al.* (1978) to be associated with their fine-grained nature rather than with any unusual history. The TL sensitivity of Tieschitz is similar to those of



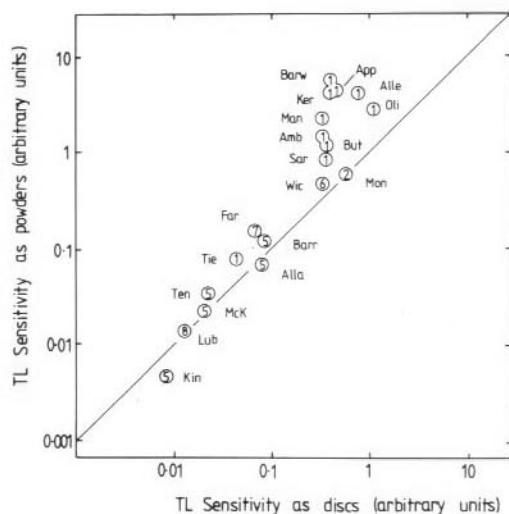


FIG. 4. A comparison of the measurements made on 10 mg of sieved powders with those made on fine-grain disks. The diagonal line has a slope of unity. Low-sensitivity meteorites give the same results, regardless of sample preparation, whereas high-sensitivity meteorites are brighter when measured as powders. See Fig. 5 for the probable explanation. The numbers indicate the degree of shock and reheating suffered by the meteorites, which are identified by the first three or four letters of their names (see Table II).

heavily shocked meteorites like Farmington and Barratta and one is tempted to ascribe its low TL sensitivity to a shock event within the first few million years of formation. Hutchison *et al.* (1979) have found numerous examples of deformed chondrules in Tieschitz, but they did not believe they were of shock origin because of the lack of other deformation textures. However, there are alternative explanations for its low TL sensitivity. Sears *et al.* (1980) have observed that type 3 chondrites have TL sensitivities 10 to 1000 times lower than those of type 6 meteorites and that their TL correlates with the extent of metamorphic alteration. Following Liener and Geiss (1968), we might suggest that low TL is sometimes a consequence of low petrologic type. This seems reasonable since in type 3 meteorites the feldspathic mineral exists as a glass rather than as crystals, and glasses generally have poorer luminescence than crystalline substances. In the case of

Tieschitz, another factor also seems to be involved. Despite a 4.45-Ga  $^{40}\text{Ar}/^{39}\text{Ar}$  age and a 4.50-Ga U, Th/He age, Tieschitz has a K-Ar age of only 2.94 Ga; presumably, its K content is lower than the value of 800 ppm assumed for the age calculation. This might be reflected in a lower TL phosphor content, which would also contribute to the low TL sensitivity of Tieschitz.

Saratov's fine-grained disk value in Table IV is comparable with the other 4.6-Ga  $^{40}\text{Ar}/^{39}\text{Ar}$  age specimens, but an earlier determination on different material gave a low value, consistent with the powder value here. The K-Ar age for this meteorite is low (3.8 Ga) while its U, Th-He age is normal (4.60 Ga), so a low K (and presumably feldspar) content is probably responsible for its slightly low TL sensitivity.

The most striking feature of the shock/reheating ranking order and TL sensitivity in Table IV is that, with the two exceptions already discussed, the order 1 meteorites have the highest TL sensitivity. There is also some relationship between the decrease in TL sensitivity and the shock/reheating ranking order. With the exception of Wickenburg, the specimens ranked 5 to 8 have a TL sensitivity an order of magnitude or so below that of the specimens ranked 1 (Fig. 4). Wickenburg has a TL sensitivity more comparable with that

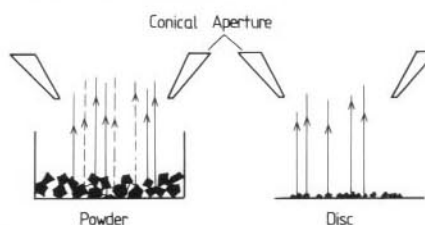


FIG. 5. Diagrams illustrating how sample albedo will influence TL measurements on powders, but not fine-grain disks. Left, 10 mg of 50- $\mu\text{m}$ -sieved powders will lie two, three, or even more grains deep and, in addition to the TL received directly from surface grains, a significant proportion will come from subsurface grains via multiple reflection. However, this "excess TL" will be negligible if the sample has a low albedo and absorbs rather than reflects the light. Right, fine-grained disks enable light to go directly to the detector without scattering.

TABLE IV  
MEASUREMENTS OF THE TL SENSITIVITY OF 22 ORDINARY CHONDRITES<sup>a</sup>

	TL sensitivity (arbitrary units)		K-Ar age (Ga) from Table III	Shock/reheating ranking order
	Measured as 10-mg powders	Measured as fine-grained disks		
Barwell	5.60 ± 0.28	0.42 ± 0.06	4.24	1
Appley Bridge	4.30 ± 0.21	0.48 ± 0.07	3.95	1
Allegan	4.20 ± 0.21	0.82 ± 0.12	4.42	1
Kernouvé	4.00 ± 0.20	0.41 ± 0.06	4.47	1
Olivenza	2.80 ± 0.14	1.16 ± 0.17	4.39	1
Mangwendi	2.20 ± 0.11	0.40 ± 0.06	4.21	1
Ambapur Nagla	1.41 ± 0.07	0.35 ± 0.05	2.64	1
Butsura	1.26 ± 0.06	0.39 ± 0.06	3.98	1
Ucera	1.16 ± 0.06	—	0.80	3
Bruderheim	1.10 ± 0.06	—	1.85	2
Saratov	0.80 ± 0.04	0.38 ± 0.06	3.80	1
Monroe	0.60 ± 0.03	0.60 ± 0.09	2.15	2
Zomba	0.60 ± 0.03	—	1.61	4
Wickenburg (stone)	0.55 ± 0.10	0.35 ± 0.05	1.72	6
Farmington	0.17 ± 0.01	0.070 ± 0.010	0.84	7
Barratta	0.100 ± 0.005	0.090 ± 0.013	0.84	5
Tieschitz	0.080 ± 0.004	0.050 ± 0.007	2.94	1
Allan Hills A77214	0.062 ± 0.003	0.087 ± 0.013	—	5
Kingfisher	0.041 ± 0.001	0.009 ± 0.001	0.50	5
Tenham	0.033 ± 0.008	0.025 ± 0.004	—	5
McKinney	0.022 ± 0.001	0.022 ± 0.003	2.15	5
Lubbock	0.015 ± 0.001	0.014 ± 0.002	1.43	8

<sup>a</sup> Listed in order of decreasing TL sensitivity.

of specimens ranked 2 to 4; that is, of the low end of the range observed in specimens ranked 1.

To summarize Section 3.1, the 22 meteorites studied here display a significant correlation between TL sensitivity and K-Ar age, and seem to display a relationship between TL sensitivity and the extent of shock and reheating.

### 3.2. Interpretations

In the first section of this paper two interpretations were offered for these observations. First, TL sensitivity may be the result of a buildup with time due to radiation damage, combined with a partial or complete resetting to zero by shock or reheating. This situation is analogous to that for the K-Ar system. Second, the TL sensi-

tivity in meteorites may have originally been uniformly high but in certain cases it was reduced by shock and reheating. The observed correlation between TL sensitivity and K-Ar age is equally consistent with either interpretation, but the relationship between shock/reheating and TL sensitivity might be more consistent with the second interpretation.

It is possible to perform laboratory experiments to test these hypotheses. If radiation damage does cause TL sensitivity to increase then we should be able to observe a significant increase in the TL sensitivity of samples which have been exposed to high radiation doses in the laboratory. Figure 7 shows the results of such laboratory experiments. To account for the correlation, one would expect a 10-fold increase in TL sen-

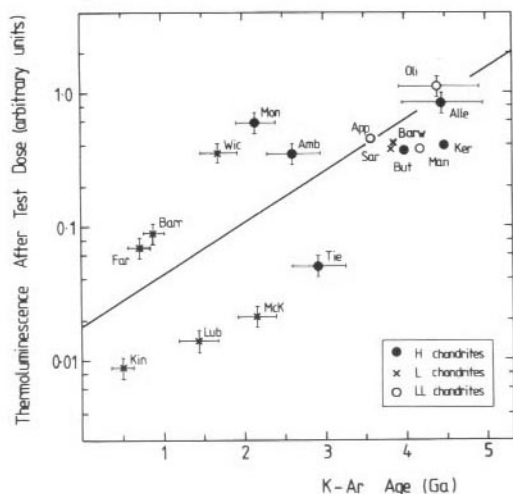


FIG. 6. Thermoluminescence after test dose against K-Ar age for 17 of the specimens studied here. The TL data are those obtained using fine-grained disks. An error of  $\pm 15\%$  has arbitrarily been assumed for the K-Ar ages; it may well be an underestimate. The regression line has a correlation coefficient of 0.74 ( $n = 17$ ) and is statistically significant at the 99% level. The points are labeled with the first three or four letters of the names of the meteorites to which they refer (see Table III).

sitivity for a comparable, or slightly smaller, increase in dose absorbed. Conversely, by giving the powder a dose of radiation comparable to that experienced

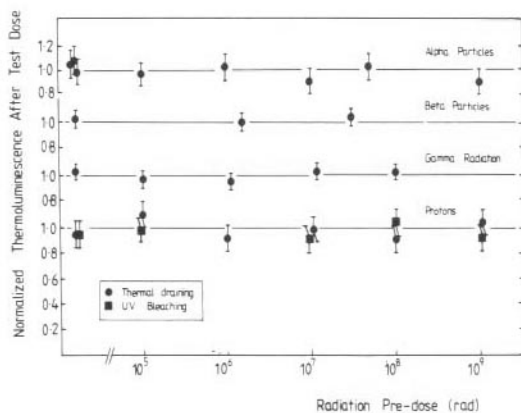


FIG. 7. Preirradiation with very high doses of protons and  $\alpha$ ,  $\beta$ , and  $\gamma$  radiation does not seem to cause any measurable change in the TL induced by a standard test dose. Before the test dose was administered, samples were drained of existing TL either by heating to  $500^\circ\text{C}$  or by exposing to uv radiation for 6 weeks.

over its total life span, one might expect a doubling of the natural TL sensitivity. Assuming chondritic abundances, I calculate that the total dose received by a meteorite from its contents of U, Th, and K, is about  $1.4 \times 10^8$  rad;  $4.7 \times 10^7$  rad from  $\alpha$  radiation,  $6.5 \times 10^7$  rad from  $\beta$  radiation, and  $2.6 \times 10^7$  rad from  $\gamma$  radiation (where 1 rad is 100 ergs of absorbed energy per gram of meteorite). Meteorites have also received a dose of about  $10^8$  rad from cosmic rays. This is not relevant to our attempts to interpret the TL sensitivity vs K-Ar correlation, but it might explain some of the scatter in Figs. 2, 3, and 6. The same experiment was therefore performed using 10-MeV protons. This energy is well below the 3 GeV normally assumed typical of cosmic rays, but two orders of magnitude or more larger than that required to produce atom displacements (Bryant and Hamid, 1970; Saidoh and Townsend, 1975).

None of the radiations used produced any measurable increase in the TL sensitivity of the Kernouvé meteorite material, despite having exceeded the natural dose by an order of magnitude in the case of  $\alpha$  particles and protons. The possibility that cosmic rays will cause sensitivity changes in meteorites has been discussed previously and it now seems unlikely that any such effect is significant (Sears, 1975; Vaz and Sears, 1977). It would seem that, rather than being due to atom displacements produced by radiation damage—as envisaged by Houtermans and Liener (1966) and Liener and Geiss (1968)—electron traps in meteorites are probably due to impurity centers. The present data concern only the LT band of meteorite TL ( $150$ – $250^\circ\text{C}$  in the glow curve) and I have not made a detailed study of a possible effect in the HT region (Sears, 1979).

In contrast to exposure to high radiation levels, both shock and reheating seem to have an effect on the TL sensitivity of meteorites. Figure 8 reports the results of the annealing experiments. Similar results were found for each of the variations in the

annealing conditions. There is a steady decrease in TL sensitivity as higher annealing temperatures are reached. A sample annealed at 1250°C for 1 hr suffered a 10-fold decrease in TL sensitivity. Of course, we have no idea of the duration of the annealing naturally experienced by reheated chondrites, but qualitatively we observe that TL sensitivity decreases of the right order are observed by annealing to temperatures which are thought probable in the light of metallographic data (see Appendix 1). It is beyond the scope of this paper to consider the causes of the drop in TL sensitivity with annealing. Numerous mechanisms are viable, such as the vitrification of the phosphor or the provision of alternative pathways to the ground state for the excited electrons.

The results of the shock-loading experiments are presented in Fig. 9 and Table V. The effect of shock on both the level of natural TL and on the TL sensitivity are presented in this plot, although it is the latter we are concerned with here. A full discussion of the shock results will appear elsewhere. Shock pressures less than 200 kbar seem to have had little or no effect on

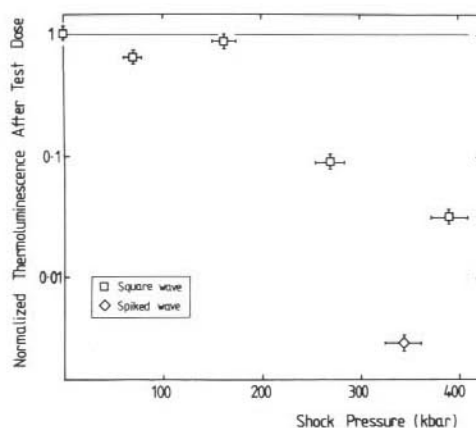


FIG. 9. The effect of dynamic shock loading on the TL sensitivity of the Kernouvé meteorite.

the TL sensitivity, but a considerable decrease is observed when the pressures exceed this value. The sample shocked to 270 kbar had a TL sensitivity 0.09 times that of the unshocked sample, while the TL sensitivity of the specimen shocked to 391 kbar was only 0.03 that of the unshocked specimen. The 335-kbar specimen was shocked in a manner slightly different from that in which the others were shocked. Instead of a square shock pulse being passed through

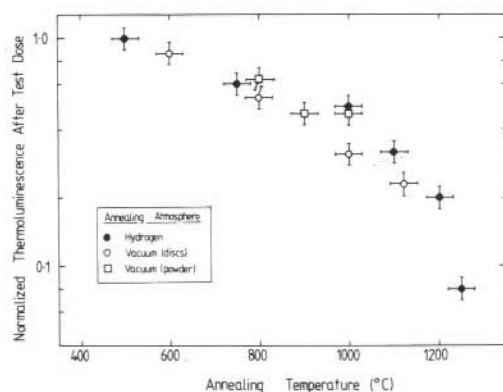


FIG. 8. The effect of annealing on the TL after test dose for the Kernouvé meteorite. The TL has been normalized to that induced by the same test dose before annealing. Some samples were annealed in a hydrogen atmosphere at atmospheric pressure while others were annealed in evacuated (better than  $10^{-3}$  mm Hg) quartz vials.

TABLE V

RESULTS OF SHOCK LOADING EXPERIMENTS

Shock level ( $\pm 5\%$ ) (kbar)	Duration of shock ( $\mu\text{sec}$ )	Normalized TL sensitivity ( $\pm 10\%$ )
0	0	1.00
66	$\sim 3$	$0.66 \pm 0.07$
163	$\sim 5.5$	$0.99 \pm 0.10$
269	$\sim 1.5$	$0.091 \pm 0.009$
335 <sup>a</sup>	$\sim 1$	$0.0029 \pm 0.0003$
390	$\sim 1$	$0.033 \pm 0.003$
150–300 <sup>b</sup>	—	0.93
300–500 <sup>b</sup>	—	0.093

<sup>a</sup> Spike pulse shock. Time given is that for pulse to decay to half its value. A square pulse of equivalent energy would give a shock value of  $\sim 500$  kbar.

<sup>b</sup> From Liener, 1966.

the meteorite, a spiked pulse was used. This has the advantage of making the material easier to recover after the shock; only about 50 mg of the 391-kbar material was recovered. The 335-kbar shock-pressure value may be an underestimate; a square pulse of the same energy would have delivered a shock pressure of  $\sim 500$  kbar. The TL sensitivity had fallen to 0.003 of its preshock value and the test sample had been completely blackened, whereas the 391-kbar specimen was not blackened by the shock.

The only existing data with which to compare these results are those of Liener (1966). He reported that a sample of Stålldalen shocked to 150–300 kbar had virtually unchanged TL sensitivity, while a sample shocked to 500–800 kbar had a TL sensitivity only 0.09 of its original value. These findings are in general agreement with the present results. A brief, but valuable, review of the effect of shock on the TL of nonmeteoritic material was compiled by Stöffler (1974).

The observed decreases in TL sensitivity are comparable with the range in TL sensitivity observed in the 22 meteorites studied here. However, the maximum pressure available in our experiments is probably lower than those which have been suggested for several of the black chondrites. Heymann (1967) suggested that certain black chondrites had been shocked to between "a few hundred kilobars up to perhaps 1 or 1.5 Mbar." Carter *et al.* (1968) found a similar range, 0.15 to  $\sim 1$  Mbar, while Taylor and Heymann (1971) reported a range of 0.5 to  $>0.8$  Mbar for seven black chondrites including Lubbock and Wickenburg.

It appears therefore that both shock and reheating are separately capable of causing appreciable decreases in the TL sensitivity of meteorites. Meteorites are known to have suffered both effects and it is impossible from the present study to say which has been the dominant one. We cannot even rule out the possibility that it is the

heating, which is associated with the shock, which is responsible for the TL sensitivity decrease in our shock experiment. An elaborate series of laboratory experiments, coupled with a knowledge of the appropriate shock-wave physics, would probably enable a reasonable assessment to be made, leading to the application of TL sensitivity to studying meteorite shock and reheating histories. For our present purposes, these data suffice to show that the correlation between TL sensitivity and K–Ar age is almost entirely a result of the shock and reheating experienced by certain meteorites. This correlation is apparently a result of shocked and reheated meteorites suffering  $^{40}\text{Ar}$  loss following the high temperature experienced.

#### 4. SUMMARY AND CONCLUSIONS

The finding of Komovsky (1961) and Liener and Geiss (1968), that there is a positive correlation between the TL sensitivity and K–Ar age of meteorites, has been confirmed. It is possible to lower TL sensitivity by artificially shock loading meteorite samples, or by annealing them at temperatures in the order of  $1000^\circ\text{C}$ . However, no observable change in their sensitivity resulted from exposure to high doses of radiation. It is therefore concluded that the correlation between TL sensitivity and K–Ar age is the result of the shocking and/or reheating of low K–Ar age meteorites. A detailed comparison of literature data on shock and reheating for the present meteorites, and their TL sensitivity seems to be consistent with this conclusion.

No attempt has been made in the present work to derive quantitative information on the shock/reheating histories of meteorites from their TL properties, but it is clearly a possibility for the future. Detailed time and temperature relationships between annealing and TL sensitivity would be required, as well as relationships between shock and annealing temperature.



## APPENDIX 1

DISCUSSION OF THE SHOCK  
AND REHEATING RANKING ORDER IN  
TABLE IV

As is discussed in the text, meteorites with  $^{40}\text{Ar}$ - $^{39}\text{Ar}$  ages of  $\sim 4.5$  Ga are assumed to have suffered minimal gas loss and to have remained relatively unreheated. Mangwendi and Appley Bridge have poorly defined plateaus at 4.0 Ga, suggesting an intense reheating early in their history. No shock or reheating symptoms were observed by Sears and Axon (1975), who examined the metal from these meteorites, and I nervously clump them together with the meteorites having  $^{40}\text{Ar}$ - $^{39}\text{Ar}$  ages of 4.5 Ga. Assigning shock and reheating ranking orders to the other meteorites is difficult because of the diffuse and qualitative nature of the data. Smith and Goldstein (1977) give the following reheating temperatures for three meteorites studied here: Lubbock, 1100–1150°C; Farmington, 1050–1100°C; Wickenburg, 950–1000°C. These are in agreement with Taylor and Heymann's (1971) estimates for Lubbock and Wickenburg (900–1200°C and 670–1000°C, respectively). Taylor and Heymann (1970) also made a detailed study of Kingfisher and concluded that it had been heated to at least 600°C. In order of increasing reheating temperature therefore,

*Kingfisher*  $\approx$  *Wickenburg*  
 $<$  *Farmington*  $<$  *Lubbock*.

Heymann's (1967) original study of shocked chondrites included seven of the present specimens. He placed Farmington, Lubbock, and Wickenburg together in his "very heavily shocked" class, and McKinney, Barratta, and Kingfisher in his "heavily shocked" class, on the basis of metallography and X-ray diffraction patterns. Assuming, after Taylor and Heymann (1971), some relationship between shock and reheating, we can now write

*McKinney, Kingfisher,*  
*Barratta*  $<$  *Wickenburg*  
 $<$  *Farmington*  $<$  *Lubbock*.

These meteorites are all listed by Hey (1966) as being black chondrites, which the present shock data, and those of Fredriksson *et al.* (1963), show is the result of shock loading to pressures in excess of 300 kbar. According to the equation of state of McQueen *et al.* (1970), this probably corresponds to a reheating temperature of  $\sim 450^\circ\text{C}$ . Bruderheim, which belongs to Heymann's "moderately shocked" class, Monroe, and Ucera are not black, although Ucera's lower K-Ar age might suggest outgassing more extensive than that of the other two. Consequently, we arrive at

*Monroe, Bruderheim*  $<$  *Ucera*  
 $<$  *Barratta, McKinney, Kingfisher*  
 $<$  *Wickenburg*  $<$  *Farmington*  
 $<$  *Lubbock*

It remains only to insert Zomba, Tenham, and Allan Hills A77214 into the ranking order. The only data on Zomba are its low  $^{40}\text{Ar}/^{39}\text{Ar}$  plateau age of 0.44 Ga and its low K, Ar and U, Th/He ages (1.61 and 1.30 Ga, respectively). In these it seems comparable with Kingfisher, McKinney, and Barratta; however, it is not black and presumably belongs between Barratta and Ucera in the ranking order. With existing data, the placement of Allan Hills A77214 and Tenham is difficult. Allan Hills A77214 was included because of its black appearance and Tenham because of the presence of veins containing high-pressure phases (Binns *et al.*, 1969; Putnis and Price, 1979). Whether these phases are present in all veined chondrites, or whether they demonstrate an intense shock history for Tenham, is unclear. For the moment, I very tentatively place them in the ranking order with Kingfisher, McKinney, and Barratta. The complete sequence is then

*Meteorites with  $^{40}\text{Ar}/^{39}\text{Ar}$* *ages of greater than 4.0 Ga (1)**< Bruderheim, Monroe (2)**< Ucera (3) < Zomba (4)**< Kingfisher, McKinney, Barratta,**Tenham, Allan Hills A77214 (5)**< Wickenburg (6) < Farmington (7)**< Lubbock (8),*

where the number in parentheses is the assigned order listed in Table IV.

## APPENDIX 2

## TABULATION OF DATA USED TO PRODUCE FIGS. 2 AND 3

Meteorite <sup>a</sup>	Class	Thermoluminescence sensitivity ( $\mu\text{lm}$ )	K-Ar age ( $10^4$ yr)	Meteorite <sup>a</sup>	Class	Thermoluminescence sensitivity ( $\mu\text{lm}$ )	K-Ar age ( $10^4$ yr)
Bath	H4	278	4.43	*Homestead	L5	20	2.11
Forest Vale	H4	68	4.62	Kandahar (Afg.)		482	4.50
Kesen	H4	100	4.02	*Knyahinya	L5	50	3.14
Pantar	H5	220	2.30	Kunashak	L6	68	0.584
Pribram	H5	72	4.31	Kyushu	L6	14	0.889
Richardton	H5	158	2.42	*Lanzenkirchen	L	36	2.15
Seres	H3, 4	50	1.69	Leedey	L6	376	4.40
St. Germain	H	102	4.44	Maziba	L	580	4.48
Tabor	H5	124	4.39	Mezel	L	160	3.23
*Tieschitz	H3	48	2.94	*Mező Madaras	L3	12	1.48
				Mocs	L6	150	4.45
Alfianello	L6	52	0.675	Monte de Fortes	L	212	4.39
Atoka	L	436	4.53	Nerft	L6	32	0.695
Ausson	L5	186	4.45	Nikolskoe	L4	164	4.04
Bjurböle	L4	192	4.29	Ramsdorf	L	84	0.554
Bruderheim	L6	190	2.11	Walters	L6	72	3.51
*Chantonay	L6	10	0.993	Zavid	L6	470	1.16
Colby (Wisc.)	L6	270	3.30				
Elenovka	L5, 6	402	4.21	Appley Bridge	LL6	620	3.95
Ergheo	L5	20	1.05	Benton	LL6	324	4.13
*Farmington	L5	24	0.841	Dhurmsala	LL6	218	3.57
Forksville	L6	480	4.38	Hamlet	LL3, 4	296	4.20
Harleton	L6	216	1.80	Olivenza	LL5	266	4.41
Holbrook	L6	150	4.65	*Soko Banja	LL4	200	4.45

Note. TL sensitivities are taken from Liener and Geiss (1968). Finds were excluded because of weathering effects. The K-Ar ages have been recalculated using assumptions and data given in the footnote to Table III.

<sup>a</sup> Asterisks denote meteorites believed by Liener and Geiss (1968), on the basis of literature data, to be unequilibrated or polymict brecciated.

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