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MEASUREMENT OF THE THERMOLUMINESCENCE SENSITIVITY OF METEORITES

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A number of means of measuring the TL sensitivity of meteorites have been examined. The TL sensitivity measured as the slope of the linear portion of the growth curve (a plot of TL versus dose) gives the equivalent results as the TL sensitivity measured from the TL induced in a drained sample by a standard test dose, provided that the test dose is chosen so as to lie on the linear portion of the growth curve.

In UV bleaching, the duration of the exposure to UV must be carefully chosen with reference to the original level of TL to be drained, and the test dose to be subsequently used. We can find no evidence of phototransfer caused by the UV irradiation.

Thermal draining (i.e. heating to 500°C) causes a (48 ± 8) percent decrease in the TL induced by a standard test dose. Furthermore, the slope of the growth curve obtained by plotting TL against the artificial dose imparted to the previously drained sample is (30 ± 14) percent lower than the slope of the plot of TL versus superimposed dose. Because of this decrease, measurements on fresh specimens should not be compared with measurements on specimens which have previously been heated. The decrease in TL sensitivity due to heating occurs only on first heating and is associated with a discoloration of the specimens.

INTRODUCTION

Meteorites have largely remained unaltered since their formation, 4.6 aeon ago. However, some of them (at least one-third of the biggest class, the L chondrites), have suffered a major shock-reheating event, probably 500 my ago (Heymann, 1967; Turner, 1969). This event is generally thought to have been the break-up of the parent body in which the meteorites formed and it resulted in a number of changes to the meteorites' structure. One consequence of the shock event was the loss of ^{40}Ar, so that the K-Ar "clock" was reset. An additional consequence appears to have been a reduction in the TL sensitivity of the meteorites involved. Thus, Komovsky (1961) and Liener and Geiss (1968) have been able to observe a significant correlation between TL sensitivity and K-Ar age in meteorites, such that those specimens with low K-Ar age also have low TL. We wish to make a study of the factors which affect TL sensitivity and to see if the sensitivity can be related, in a quantitative way, to the shock and reheating history of meteorites. It has therefore been necessary to consider the way in which TL sensitivity is best measured. In this paper, we report the results of our investigations.

"TL sensitivity" is the amount of TL produced by a given dose of absorbed radiation. There are several ways by which this quantity may be measured. The most straightforward is to remove the natural TL (by some means) and then to measure the TL induced in the sample by a standard laboratory test dose. The natural TL may be removed either by heating the specimen to 500°C in the TL readout apparatus ("thermal draining") or by exposing it to ultra-violet radiation ("UV bleaching"). In studies on TL phosphors other than meteorites, both methods of draining have caused problems. In some cases, thermal draining has caused physical changes in the phosphor which, in turn, result in changes in the TL sensitivity (e.g. Aitken and Fleming, 1972; Fleming, 1972). Several suggestions can be forwarded to account for the changes in sensitivity, but it is difficult to assess which mechanism is operative in any particular mineral.

The main complication with UV bleaching is that it can cause electrons to transfer from deep to shallower traps ("phototransfer," e.g. Bailiff *et al.*, 1977), so that the TL in certain regions would increase rather than decrease under UV irradiation. A third method for measuring TL sensitivity is to measure the TL in a number of natural samples which have received various doses of radiation. The slope of the TL versus superimposed dose plot is then a measure of the sample's TL sensitivity.

METHODS

Our apparatus for measuring TL consists of a 5 × 1 × 0.0125 cm nichrome strip, heated at a linear rate of either 3 or 5°C s^{-1}. The temperature of the heating strip is monitored with a chromel-alumel thermocouple which feeds an electronic control unit and the X axis of an X–Y plotter. The Y axis of the plotter is used to record the TL signal (photomultiplier tube current). The TL is measured by an EMI-9804 QB photomultiplier tube in front of which are two filters; a heat filter (Chance HA3) and a blue filter (Ilford 621 broad band filter used to suppress black body radiation). Ordinary chondrite TL is blue and is little affected by these filters. The PMT signal is amplified by a Keithley electrometer.

The meteorites used in this study, and their sources, are listed in Table I. Samples were normally prepared as powders (by gentle grinding, removal of the magnetic fraction with a hand magnet and then grinding again to ensure that all the powder passes through a 50 μm sieve). The triboluminescence produced by this process is negligible when compared with the natural TL level. For experiments involving UV bleaching, fine-grain discs were prepared from the powder. 20 mg of sieved powder were shaken in 5 ml of acetone, and those grains which took between two and 20 minutes to settle were deposited on 1 cm aluminium discs. A Camag UV lamp producing radiation of wavelength 366nm and with an intensity of 3.7×10^{-7} W cm^{-2} was used for the UV bleaching.

Artificial irradiations were performed using a ^{60}Co γ–source at a dose rate of about 100 krad h^{-1}.

RESULTS AND DISCUSSION

Measurements of the TL sensitivity of the meteorites listed in Table I were obtained thus: (i) from the slope of the TL versus dose curves; (ii) from a standard test dose after thermal draining; and (iii) from a standard test dose after UV bleaching. The effect upon the glow-curves of the addition of radiation doses on top of the natural dose, N, is demonstrated in Figure 1. When doses of less than 50 krad are superimposed, (Figure 1a), the 200°C peak appears to broaden. In fact, two lower temperature peaks are being filled by the artificial radiation. At doses in excess of about 50 krad (Figure 1b) the superimposed dose is so large that the natural dose has little influence on the shape of the glow-curve. Thus, the overall shape changes little with additional dose, and is the same shape as the glow-curve produced by powder which has become drained of its natural TL and then given various test doses (Figure 1c).

Plots of TL (from the low temperature part of the glow-curve) versus dose, ("growth curves") are shown in Figure 2. Two curves for each meteor-

TABLE I

Meteorites used in this study, their source, class and TL sensitivity as measured in a variety of ways.

Meteorite	Source and catalogue number[a]	Class	Slope of growth curve (TL sensitivity)		Decrease caused by thermal draining (%)	TL induced by test dose (TL sensitivity)		Decrease caused by thermal draining (%)
			N + γ	γ		UV bleaching	thermal draining	
Alta'ameen	AR	LL5	0.55	0.39	29	12.3	6.4	46
Barwell	BM 1966,57	L5	0.24	0.14	42	6.3	3.0	51
Bruderheim	AML H7.45	L6	0.084	0.068	15	2.6	1.6	39
Kernouvé	BM 43400	H6						
Olivenza	BM 1925,430	LL5	0.21	0.14	33	6.8	3.2	51
Wold cottage	BM 1073	L6	0.29	0.20	31	8.0	3.6	55

N.B. Units: Slope of growth curve: TL/krad (TL in arbitrary units).
TL induced by test dose: TL from 50 krad (arbitrary units).

[a] AML—Dr. Glenn Huss, American Meteorite Laboratory, Denver, Colorado.
BM—Dr. R. Hutchison, British Museum (Natural History), London.
AR—Dr. Y. Al-Rawi, College of Science, Baghdad, Iraq.

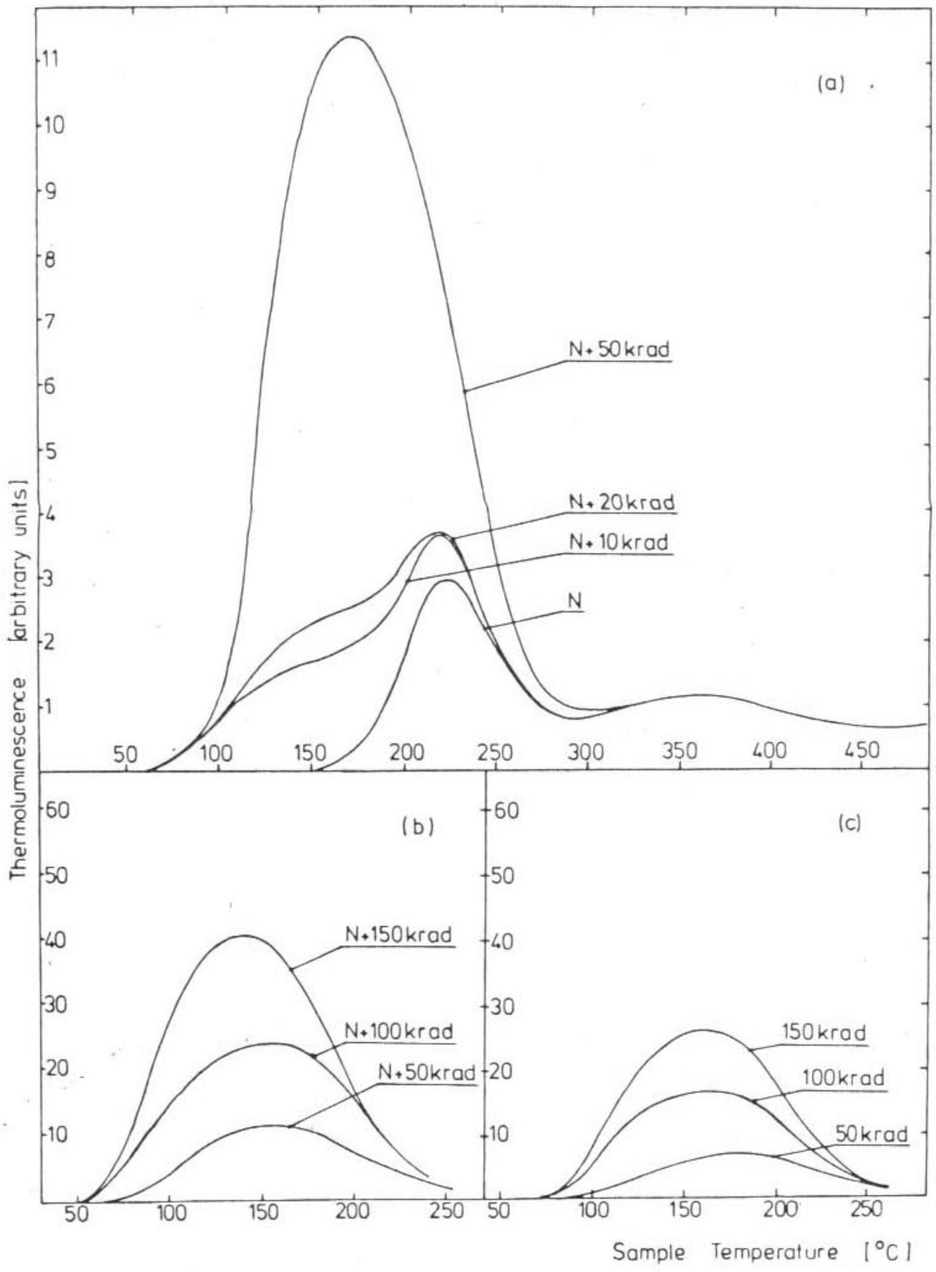

FIGURE 1 (a) TL glow-curves from Wold Cottage showing how the shape of the glow-curve changes when a dose of radiation of 50 krad or less is superimposed on the natural dose. Extra TL centres are being filled to give rise to additional glow around 70–200°C. When doses of greater than 50 krad are added (b) the intensity of the emission increases rapidly (cf. Figure 2) and the natural dose no longer has a significant effect on the shape of the glow-curve. For samples which have been previously drained (c) irradiation produces a glow-curve shape which is similar to those of (b), but less intense because of the effect of heating the sample prior to irradiation. Only the low temperature TL is shown in (b) and (c).

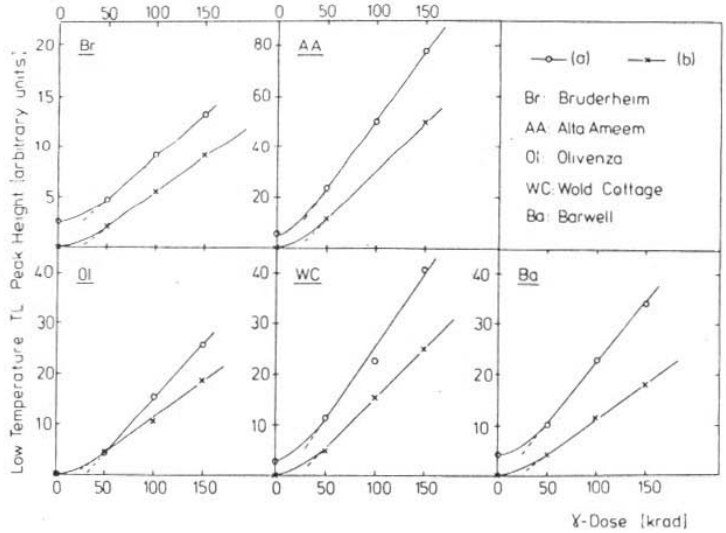

FIGURE 2 "Growth curves" for five different meteorites. Two curves are shown for each sample indicated; (a) the growth of TL as artificial doses of radiation are superimposed on the natural TL (N + γ); and (b) the growth of TL as the artificial dose is increased for specimens from which the natural TL has been thermally drained (γ). The slope of the growth curves for each of the γ specimens is less than that for the N + γ specimens.

ite are presented; one in which the artificial dose has been added to the natural dose (these are termed the "N + γ powders") and one in which the artificial dose was given to powder which had previously been heated to 500°C in the TL apparatus to remove its natural TL (the "γ-powders"). For most part the curves are linear, but at low doses (<50 krad) they are both supralinear. Supralinearity is a well-known phenomenon, but in many phosphors it is poorly understood. In this case, it is probably associated with the existence of several discrete TL peaks within the glow-curve. From the slope of the linear portions of the growth curves, we can derive a measure of the TL sensitivity (Table I). For each meteorite, the sensitivity of the N + γ powders is 14–40 percent higher than that of the γ powders. This cannot be attributed to the effect of the natural dose present in the N + γ powder, because it only affects the intercept of the growth curve, not its slope. We believe it is associated with a slight discolouration suffered by each meteorite examined when they are first heated. Fresh powders are recognisable paler in colour than heated powder.

We now consider the determination of TL sensitivity from a single measurement of the TL induced by a standard laboratory test dose; this is a frequently used procedure and it removes the need to produce a full growth curve. We need to consider (i) a means of removing the natural TL, and (ii) the size of the test dose used. The test dose must be chosen so that the TL lies on the linear portion of the growth curve. From our discussion of supralinearity, a dose of 50 krad or larger is required. However, the TL level ceases to increase linearly with increasing does at doses >300 krad—i.e. the TL approaches saturation (Figure 3). In our measurements, we have used a test dose of 50 krad.

Removal of the TL by exposure to UV is illustrated in Figure 4. The samples were thermally drained and given a 50 krad test dose prior to UV exposure. Under our conditions of UV bleaching, the TL has dropped to ten percent of its initial value after 6.3h and to two percent after 80h. In order to sufficiently drain the natural TL of a meteorite sample, prior to measuring the TL induced by a 50 krad test dose, 80h of exposure to UV would seem to be required. In many experiments involving high radiation doses, the TL is saturated. Using an extrapolation of the solid line in Figure 4 (inset), we find that it requires a 40 day exposure to UV to cause the TL at its saturation level to fall to two percent of its 50 krad test dose level.

Phototransfer does not seem to be a problem in

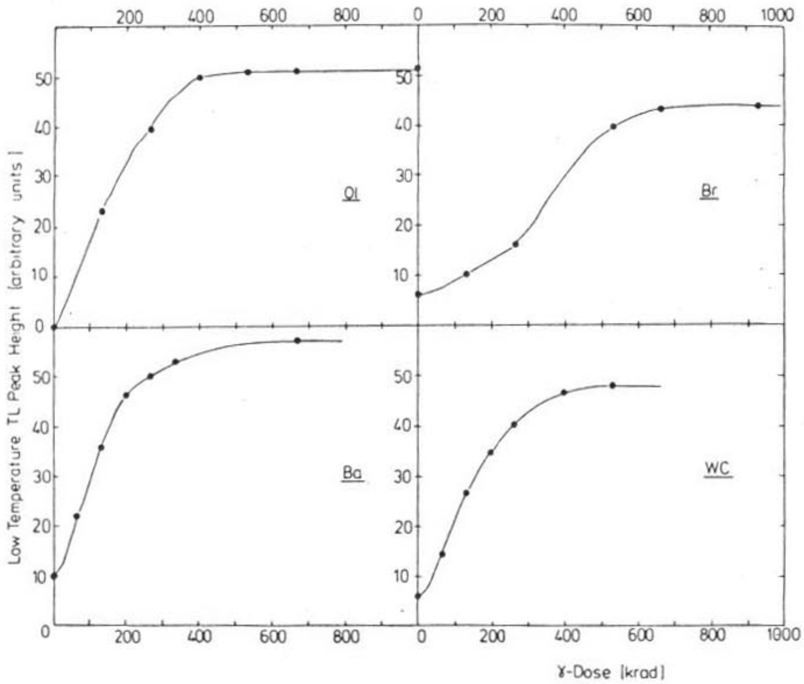

FIGURE 3 The growth curves for four meteorites using N + γ specimens and large artificial doses. Saturation begins to be reached at doses of >300 krad for each meteorite.

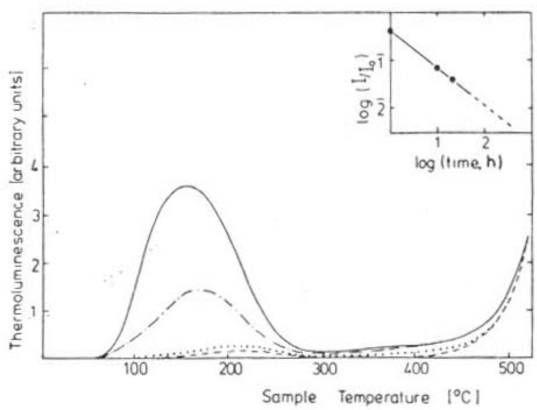

FIGURE 4 The decay of TL from the Kernouvé meteorite due to exposure to 366 nm UV radiation. Exposures of one hour (– · – · – · –), ten hours (· · · · · ·) and 22 hours (– – – – – –) were used. The full line represents the TL from a sample irradiated with 50 krad γ without any exposure to UV. From the decay of TL with time of exposure (inset) it can be seen that 6.3 hours of exposure are required to reduce the TL to ten percent of its original level, whilst 80 hours of UV exposure are required to reduce the TL to two percent of its original level. In the inset diagram, I/I_0 is the fraction of TL remaining after a given number of hours of UV bleaching.

the present work. The glow curves in Figure 4 show no signs of that phenomenon. As an additional check, a sample of thermally drained meteorite powder was exposed to UV for 100h and its TL measured. If phototransfer occurred, it would have transferred electrons from the traps which were too deep to be emptied by the thermal draining, into traps which would produce measureable TL. In fact, there was no detectable TL signal.

Also listed in Table I are the TL sensitivity estimates based on the TL induced in two samples of each meteorite by a 50 krad test dose. One sample was drained of its natural TL by an 80h exposure to UV, whilst the other was drained by heating to 500°C. As with the previous sensitivity determinations, the samples which have been thermally drained have significantly lower TL sensitivity than those which have not. In fact, the decrease is slightly greater (48 ± 8 percent) than the decrease in the slope of the growth curve (30 ± 14 percent), although it is more uniform. (The errors are simply chosen so as to cover the observed ranges.)

It is frequently necessary to repeatedly measure the TL of a given specimen. Figure 5a shows a plot

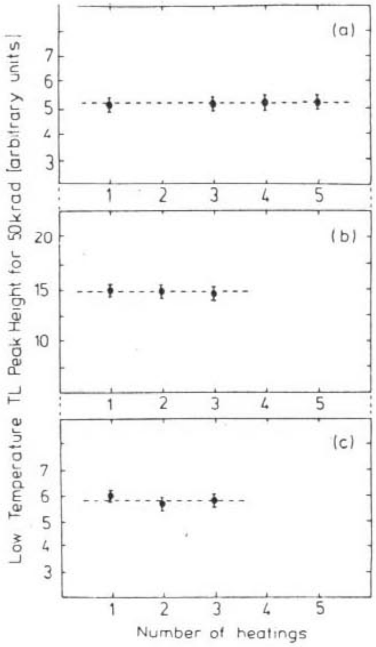

FIGURE 5 (a) The TL from a standard test dose (50 krad) given repeatedly to a previously drained sample of the Wold Cottage meteorite and readout after each irradiation, plotted against the number of times the sample had been previously heated. After the first heating, no additional decrease in TL output is observed. (b) and (c) show similar plots for Alta'ameen and Barwell, but here the samples were heated several times *before* irradiation and then subjected to a standard test dose. Again, no additional decrease in TL is observed following the first heating.

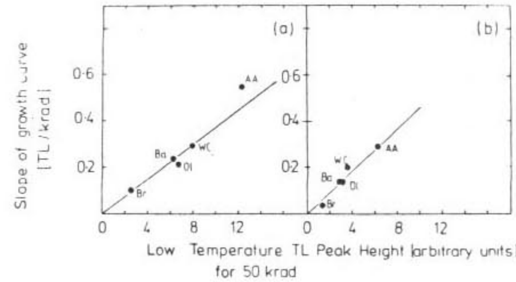

FIGURE 6 Comparison of the different ways in which TL sensitivity is measured for (a) samples which have not been heated, and (b) samples which have been heated. The TL/krad from the slope of the growth curve is not the same as that from a test dose of 50 krad because the supralinearity in the growth curve (cf. Figure 2) means that the linear portion of the curve will not pass through zero when extrapolated back. Otherwise, TL sensitivity as measured by a standard test dose is equivalent to taking the slope of the growth curve provided that the test dose which is used is on the linear portion of the curve. The result should be expressed in terms of the test dose used—e.g. "TL from 50 krad." Furthermore, a sensitivity measurement on a sample which has not been heated should not be compared with that of a sample which has been heated.

of the TL produced by repeatedly giving the same test dose (50 krad) to a sample of Wold Cottage and measuring its TL each time. The TL is plotted against the number of times the sample had been previously heated. Clearly, there is no change in the TL signal due to repeated measurement. In case this result is due to a combined effect of pre-dose irradiation and thermal cycling, samples of Alta'ameen and Barwell were heated several times *before* they were given a test dose. The plots in Figure 5 (b and c) show the TL signal produced from the test dose plotted against the number of times the samples had been previously heated. Again, it can be seen that, following the initial decrease in sensitivity upon first heating, the TL does not decrease further with additional heatings.

In Figure 6 we compare the TL sensitivity estimates obtained in different ways. Figure 6a compares the slope of the TL versus superimposed-dose curve with the TL from a standard test dose (50 krad) following UV bleaching of the natural TL—i.e. determinations which do not involve thermal draining. Figure 6b compares the slope of the TL versus artificial-dose curve with the TL from a standard test dose (50 krad) following *thermal* drainage of the natural TL. In each case, the agreement between the two types of measurement is good; we therefore believe that measuring the TL induced by a 50 krad test dose is equivalent to determining the slope of the growth curve. It is, however, ill-advised to compare sensitivity determinations which involve thermal draining with those which do not. Furthermore, it must be noted that the TL/krad derived from the slope is not equal to that derived from a standard test dose because the supralinearity seen in Figure 2 causes an extrapolation of the linear portion of the curve not to pass through zero. Because of this, when TL sensitivity is determined by a standard test dose, it is advisable to express the sensitivity in terms of the actual dose used—e.g. "TL per 50 krad" (say) and not TL/krad (i.e. a "normalised" value).

CONCLUSIONS

The assessment of TL sensitivity from the slope of the growth curve and from the TL induced in a drained sample by a laboratory test dose are essentially equivalent operations, provided that the test dose is suitable and is quoted when giving the result. A suitable test dose is one which lies on the linear portion of the growth curve; for meteorites, 50 krad seems to be a satisfactory value. Because of the decrease in TL sensitivity caused by the first heating, measurements of the TL sensitivity of samples in their natural state should avoid thermal draining; the slope of the $N + \gamma$ curve or UV bleaching followed by a 50 krad test dose would be appropriate methods. Phototransfer does not appear to pose any problems for UV bleaching, but the UV exposure time has to be chosen to suit the level of TL being drained and the size of the test dose to be used. In the present work, typical natural TL levels required 80h, and saturation TL levels required 40d of UV exposure for draining prior to the measurement of TL sensitivity with a 50 krad test dose.

In certain cases, thermal draining is implicit in the measurements—for example, when measuring the TL sensitivity of a given sample before and after some treatment. It is ill-advised to compare sensitivity determinations which involve thermal draining with those which do not, so it is necessary to heat the sample once before the first sensitivity determination is carried out. This is acceptable because subsequent to the first heating, further heatings cause no additional change in the TL sensitivity.

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