

Thermoluminescence dating of meteorites

Abstract

Normal TL dating techniques fail with meteorites because of their great age. There is evidence in the literature of a correlation between TL sensitivity—the TL of a sample which has been drained and given a standard test dose—and K-Ar age, which suggests that the former can be used for dating. The effect on TL sensitivity of annealing, and of large doses of γ radiation, has been examined. The results suggest that the correlation is the result of certain meteorites having been annealed in space, since annealing lowers both TL sensitivity and K-Ar age. Except possibly for a region of the glow curve around 400°C, it seems unlikely that TL sensitivity can be used for dating, but it may provide a means of determining the annealing temperature of reheated meteorites.

INTRODUCTION

There are probably three types of event in the history of meteorites which have proved worth dating. Going backwards in time, these are: (i) the date at which the meteorite came to be exposed to cosmic radiation; (ii) the date at which the meteorite parent body broke up; (iii) the date at which the meteorite and its parent body formed (Sears, 1978). The first is determined by measuring the abundance of certain isotopes which are the result of nuclear reactions between cosmic rays and nuclides in the meteorite. These ages are typically 10^7 years. Date (iii) is measured by the whole gamut of geochronological methods, notably those using the Rb/Sr system and the natural decay chains. These usually lead to ages of 4.6×10^9 years. Date (ii) has only recently emerged as one of significance, and so far only the K-Ar method and its new off-spring, the ^{40}Ar - ^{39}Ar technique, are able to put any kind of a date to it. Values are typically 5×10^8 years.

Absolute thermoluminescence dating techniques developed for pottery, and extended to some extent to geological materials, can go back roughly 10^5 years (Aitken, 1974). Subject to the constraints imposed by anomalous fading, thermal decay and saturation, these TL techniques may eventually be applicable to certain exposure age determinations (date (i)). This would seem particularly true of the dozen or so meteorites with exposure ages of less than 10^6 years. For the other dates, these constraints become prohibitive (McKeever and Sears, 1978), and we must resort to empirical techniques based on TL sensitivity—an approach first proposed by Houtermans and Liener (1966) and Liener and Geiss (1968).

“TL sensitivity” is the TL intensity of a sample which has been drained of its natural TL, and given a standard test dose. It is sometimes referred to as “artificial TL”. Liener and Geiss (1968) observed that there was a four-fold

increase in the TL sensitivity in meteorites, which correlated with a ten-fold increase in K-Ar age. They proposed two models to explain the observations (Figure 1). (i) The electron traps in meteorite phosphors (predominantly feldspar) are radiation induced, and build up uniformly with time—TL sensitivity may then be considered as a measure of electron trap numbers. This situation allows TL sensitivity to be used to determine ages, provided some estimate of the rate of sensitivity build up can be made. (ii) TL sensitivity was originally the same in all meteorites, but certain meteorites had their TL sensitivity reduced by some event (e.g. break up of the meteorite parent body) which also lowered the K-Ar age by causing loss of argon. Heymann (1967) has shown that virtually all meteorites with K-Ar ages of 5×10^8 years—unlike those with 4.6×10^9 year ages—have been severely reheated. The cause of reheating appears to have been shock associated with break up of the meteorite parent body. In this instance, TL sensitivity provides a measure of the extent of reheating.

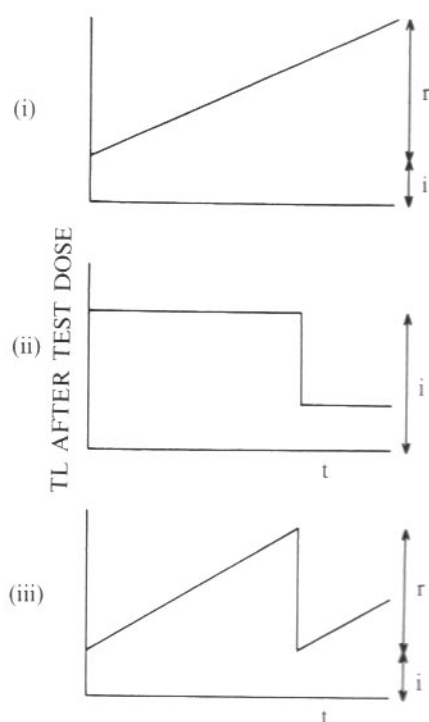


Fig. 1. Schematic representations of possible explanations for the correlation between TL sensitivity and K-Ar age observed by Liener and Geiss (1968). Models (i) and (ii) are essentially those proposed by Liener and Geiss. Model (i) represents a steady build up in sensitivity due to the production of traps by radiation. r represents this "radiogenic" sensitivity, and i is that originally present ("intrinsic" sensitivity). Model (ii) assumes that radiation cannot increase sensitivity, but that some event at time t lowered it and the K-Ar age. The change in sensitivity is a measure of the intensity of the event. Model (iii) is a combination of the previous two. In this instance TL sensitivity is lowered by some event at time t , but builds up again afterwards. This situation is analogous to that for K-Ar dating and allows an estimate of t .

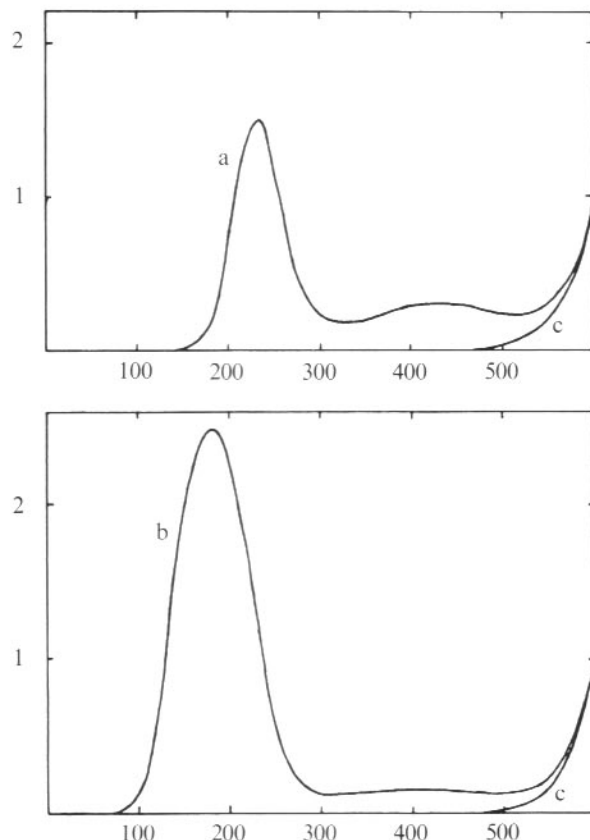


Fig. 2. Glow curves for the Kernouve meteorite: curve (a) for the natural powder, curve (b) for powder which has been drained of its natural TL and given 50 krad γ radiation, and curve (c) is for black body radiation.

A third possibility is that both situations apply. In this case (model iii), sensitivity due to radiation damage was totally destroyed by break up of the meteorite parent body, but afterwards it built up in a time-dependent fashion. This is analogous to the K-Ar situation and it would allow TL sensitivity to be used to measure the date of meteorite parent body break up.

I report here the results of two experimental approaches aimed at identifying the situation which applies. I have subjected meteorite powder to high radiation doses—comparable to those experienced in the lifetimes of the meteorite—in an attempt to increase their TL sensitivity, and I have annealed meteorite samples to see if TL sensitivity can be lowered by reheating. The results of these experiments seem to favour model (ii).

EXPERIMENTAL

The meteorites used in this work are Kernouve and Barwell, which are classified H6 and L5,6, respectively, in the Van Schums and Wood (1967) scheme. The

annealing experiments were made in wire-wound tube furnaces. One of these allowed a continual stream of H_2 at one atmosphere pressure to flow around the specimen during annealing. No difference was observed between a specimen annealed at $500^\circ C$ for 5 minutes and one annealed at the same temperature for 30 minutes, so a single annealing time of 5 minutes was used for all the results discussed here. The TL apparatus and techniques are similar to those described by Mills et al. (1975), except that the heating strip is made of nichrome. For the present work 10 mg of sieved powder, from which the metal and sulphide have been removed with a magnet, was used for the TL measurements. The test dose used to measure sensitivity was 50 or 5 krad from ^{60}Co γ rays, and the dose rate was 100 krad/hour. The heavy irradiations were also made using ^{60}Co γ rays but at a dose rate of 1.359×10^6 rad/hour.

RESULTS

Annealing experiments

Figure 2 shows glow curves for the Kernouve meteorite in its natural state, and after the drained powder had been irradiated to 50 krad with γ rays. There are two "bands" of TL, which I refer to as LT (100-250°C) and HT (300-450°C). Each of these probably consists of a number of peaks. In the natural state LT is about 5 times as intense as HT while the TL sensitivity of LT is some 17 times that of HT. This reflects the extensive thermal draining of LT, both in space and on earth. HT is also much higher in the natural specimen, showing that its equivalent dose is, as expected, very much greater than 50 krad—one might predict that it could be as much as 10^9 rad (i.e. one rad per year for 10^9 years).

In Figure 3 I show the results of the annealing experiments. The sensitivity of LT behaves very similarly when the annealing was performed in air, and when the atmosphere during annealing was hydrogen. Until $500^\circ C$ there is no change. Above this temperature annealing causes a lowering of TL sensitivity. Initially the drop seems fairly steady, despite a discontinuity at $750^\circ C$ which is most probably caused by a mineralogical change in the powders. At this temperature it has dropped to about one half its unannealed value. By $1100^\circ C$ the TL sensitivity has fallen to one quarter its unannealed value. After this there is a very sharp drop, and material annealed to $1200^\circ C$ has a TL sensitivity only 1/50 that of the unannealed material. There is no obvious change in the shape of the glow curve with annealing, suggesting that the traps corresponding to the various peaks throughout LT are annealing at the same rate.

In initial experiments, some material was annealed in air without prior removal of metal. This caused a new peak to appear at $260^\circ C$, reminiscent of a peak at this temperature in powder from meteorites which showed signs of rusting (Sears and Mills, 1974). I am therefore inclined to believe that it is associated with the oxidation of iron.

The effect of annealing on the TL sensitivity in the HT region depends on the annealing atmosphere. Figure 4 shows the glow curves of two specimens which

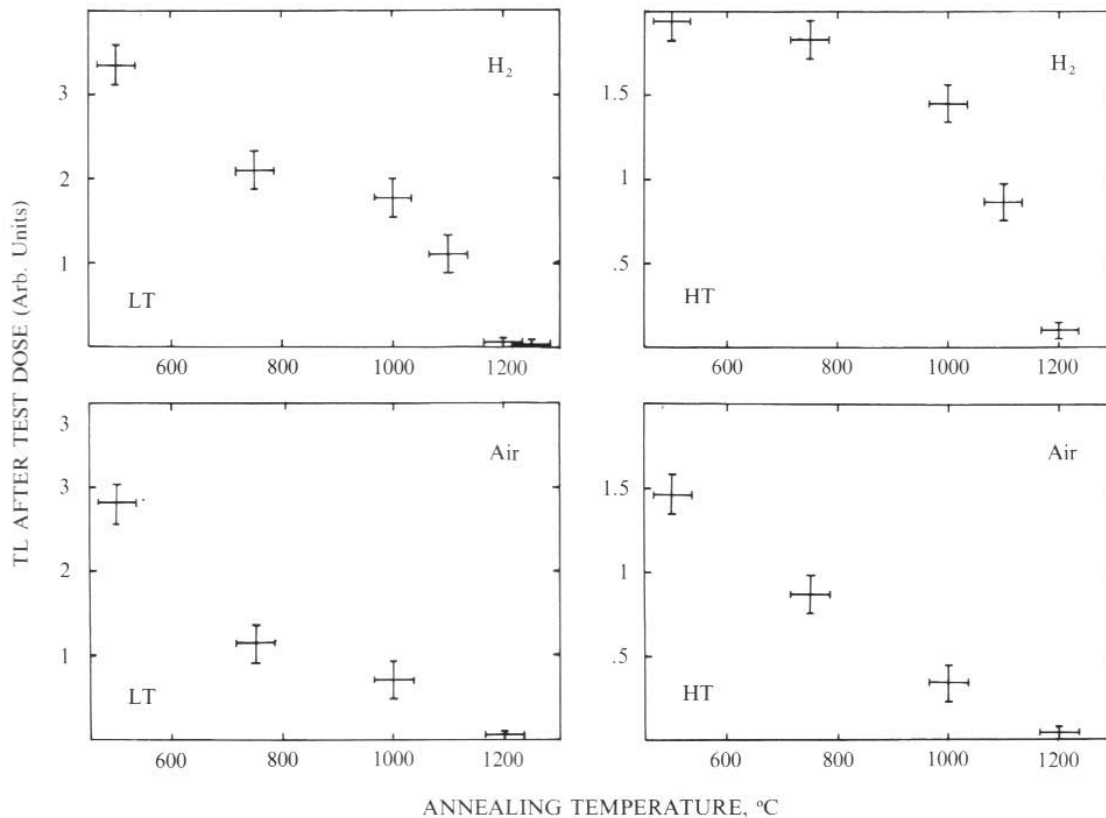


Fig. 3. Results of the annealing experiments. The TL intensity was measured after a standard test dose of 50 krad γ radiation. The height of the low temperature peak is shown in the left two plots, and the TL intensity at 400°C is shown in the two plots on the right. The atmosphere in which the annealing was done is shown in the top right hand corner of each plot.

have been annealed at 1000°C. That which was annealed in air seems to lose sensitivity in this region faster than that which was annealed in hydrogen. For the latter specimen, the drop in TL sensitivity with annealing temperature exceeds 1000°C (Figure 3). Oxygen displacement would seem to be a likely mechanism for radiation induced traps in silicates (G. F. J. Garlick, personal communication), and annealing traps produced by this process might well be affected by the atmosphere in which the annealing is done.

Irradiation experiments

Both annealed and unannealed meteorite powders were subjected to large doses of γ radiation in an attempt to increase their TL sensitivity. A sample which had been annealed to 1200°C for 5 minutes in a hydrogen atmosphere was exposed

to ^{60}Co γ rays until a dose of 2×10^8 rad had been reached. The TL was then drained, and the sample, together with a control specimen which had not been irradiated, given a 50 krad test dose. The resulting glow curves are shown in Figure 5. Except possibly for part of the HT region, there seems to have been no increase in the TL sensitivity as a result of the irradiation—certainly not by the factor of fifty or so needed to restore LT to its initial (pre-annealing) level. Around 400°C the TL sensitivity was increased by a factor of five or so by the heavy irradiation.

There is a high probability that the annealing produces a permanent change in the meteorite's TL properties, other than annealing out traps—for example, I suspect there are substantial mineralogical changes in the phosphor. For this reason, aliquots of meteorite powder in its natural state were also given large doses of radiation artificially to see if there could be any radiation-induced increase in TL sensitivity. The results are shown in Figure 6. From these data, it again seems that there is little increase in the TL sensitivity of the LT region as a consequence of the high doses experienced. A close examination was also made of the TL in the 400°C region to see if there was any sensitization similar to that observed in the annealed material above. However, there was none.

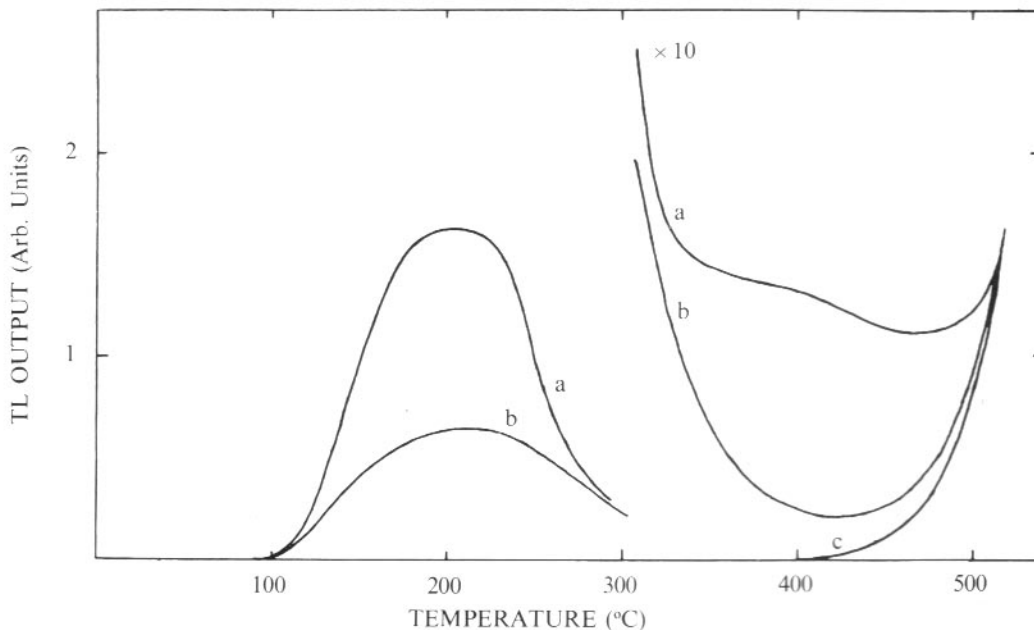


Fig. 4. Glow curves after a 50 krad test dose for two specimens annealed at 1000°C. Curve (a) is for the specimen annealed in hydrogen, and curve (b) is for the specimen annealed in air. Curve (c) is the black body radiation. Notice particularly the difference in shape of the curves around 400°C.

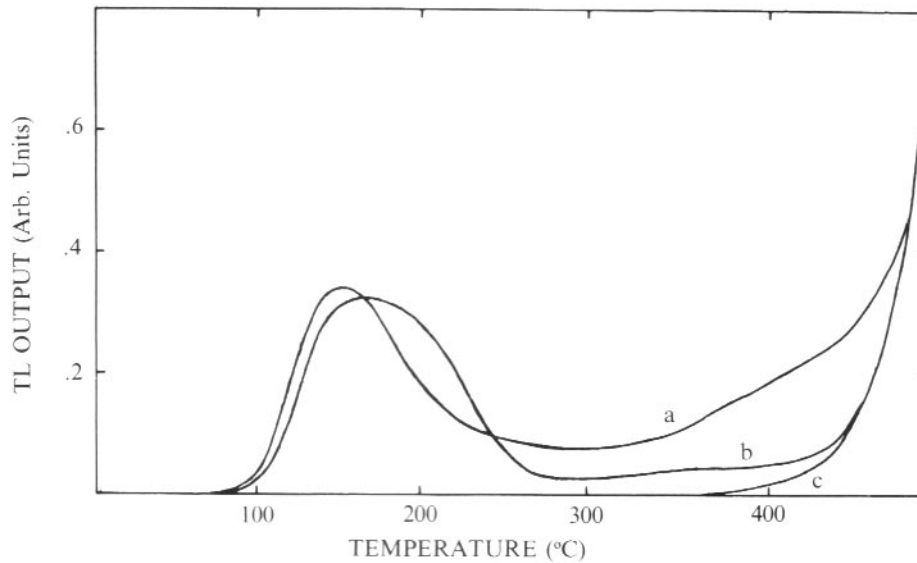


Fig. 5. Two glow curves for material annealed at 1200°C and given a 50 krad test dose. Curve (a) was irradiated to 2×10^8 rad with γ radiation and drained prior to giving the test dose. Curve (c) is black body radiation.

DISCUSSION AND CONCLUSIONS

On the strength of the present data, it seems unlikely that radiation can induce any TL sensitivity increase in meteorites—at least not in the region where TL sensitivity is at a maximum (100–250°C). This would eliminate models (i) and (iii) described earlier and shown schematically in Figure 1. The remaining alternative requires that TL sensitivity can be lowered by reheating, and this I have shown is the case. The drop in sensitivity produced by annealing to temperatures around 1000°C is of the same order as that observed by Liener and Geiss (1968)—roughly a factor of four. Furthermore, Heymann (1967) has observed that meteorites with low K-Ar age have been reheated. Recently, Smith and Goldstein (1977) have estimated that several chondritic meteorites they examined have been reheated to temperatures around 1000°C. These observations fit together rather well in support of model (ii)—which suggests that TL sensitivity was originally fairly uniform but has been lowered in certain meteorites by annealing to temperatures around 1000°C. The annealing also resulted in the loss of argon which caused the K-Ar clock to be reset.

There are two provisos which should be added. Firstly, if the traps were the result of radiation damage one would also expect them to be removed by annealing, so the annealing results do not, as of themselves, argue against radiogenic traps. Secondly, only γ irradiation has been used in the attempts to increase TL sensitivity and other radiations may produce different results. We now know rather well the relative efficiencies of α , β and γ radiation in promoting electrons to traps (Aitken, 1974), but nothing is known about their relative abilities

to cause sensitivity increases—even though this phenomenon is the basis of the pre-dose dating technique using quartz (Fleming, 1973). There is some evidence that cosmic rays (mainly protons) produce sensitivity increases in small meteorite samples (Vaz and Sears, 1977), although this was not the case with the large Estacado specimen (Sears, 1975). In any event, there is clearly a need to extend the heavy irradiation experiments to other types of radiation.

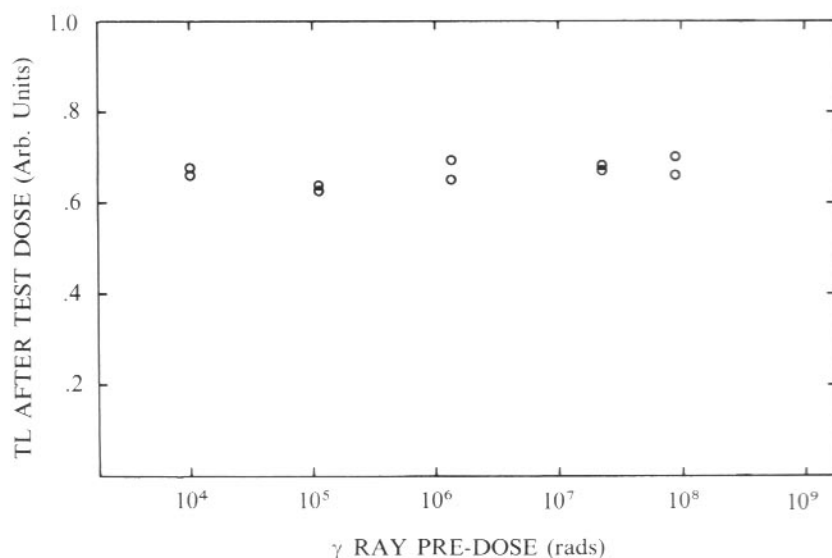


Fig. 6. Height of the low temperature peak after a test dose of 5 krad of γ radiation. The samples had previously been subjected to high doses of γ radiation, in an attempt to increase sensitivity, and then drained.

Some interesting effects seem to be apparent in the 400°C region of the glow curve. Unlike the low temperature region just discussed, the drop in sensitivity depended on the annealing atmosphere. In fact the drop was more abrupt, and required higher temperatures, when the annealing atmosphere was hydrogen than when it was air. The implication of these results on ideas of the nature of meteorite TL will be pursued elsewhere. Also in this region, an increase in TL sensitivity was observed after heavy γ irradiation of annealed material. However, this was not the case with unannealed material. It was therefore either associated with new phosphors produced by the annealing, or is so low in TL sensitivity that it could only be seen in samples whose general TL sensitivity is low. Further experiments are required to resolve this, but it would seem that any attempt to use TL sensitivity for TL dating should concentrate on this region of the glow curve.

In another paper presented at this seminar, P. Levy proposed the use of TL sensitivity—in this case in a terrestrial feldspar—to measure reheating temperatures.

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