

Thermoluminescence of Apollo 12 samples: Implications for lunar temperature and radiation histories

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Abstract—Thermoluminescence (TL), both natural and γ -ray induced, has been studied in contingency fines 12070,112 and in powdered samples from the interior rock chip 12051,15, in the temperature range 20°–600°C. In the natural samples glow peaks are observed at temperatures above $\sim 300^\circ\text{C}$, those below having been thermally drained. Artificial irradiation introduces a prominent new peak at $\sim 170^\circ\text{C}$, and enhances those in the region 300°–550°C. The growth of the high-temperature peaks with γ -ray irradiation is studied in order to deduce both the “natural (or equilibrium) dose” and the “half-dose” (needed to fill half the available “traps”) for the samples. The TL parameters of the traps concerned are determined by the “initial rise” method. These data allow the “effective temperature” of lunar storage ($358 \pm 10^\circ\text{K}$ for the fines and $346 \pm 5^\circ\text{K}$ for the rock chip) and the radiation dose rates received (~ 30 rad/y within a factor of 3, and ~ 60 rad/y within a factor of 2, respectively) to be calculated for the samples. By considering the attenuation of the diurnal heat wave on the moon, the mean subsurface depth of these samples can be estimated (~ 0.5 cm for the fines, and ~ 2.5 cm for the rock chip). Other topics discussed include spectral analysis of TL glow, inhomogeneities in the distribution of TL phosphors, and temperature sensitization of samples.

INTRODUCTION

THE MAIN PURPOSE OF studying the thermoluminescence (TL) of lunar material is to throw light on the thermal and radiation history of the moon. The radiation received by the moon (galactic cosmic rays and solar particles, in addition to internal radioactivity) leads to electrons being trapped in energy levels in the “forbidden” band gap of the solid, while the ambient temperature on the moon causes some of them to be released from these traps. By studying the properties of these traps, such as the growth

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in the number of trapped electrons with radiation, and their release as a result of a systematic heating of the samples, it should, in principle, be possible to draw inferences regarding the two opposing processes: the radiation-filling and the thermal-drainage of traps (Christodoulides *et al.*, 1971). Several groups (see Hoyt *et al.*, 1970, 1971; Dalrymple and Doell, 1970; Doell and Dalrymple, 1971; Geake *et al.*, 1970; Garlick and Robinson, 1971) have studied lunar TL with these and related aims in mind. Two of us have reported work on Apollo 11 and 12 samples (Blair and Edgington, 1970; Blair *et al.*, 1971) with particular reference to a possible relation between TL and transient lunar phenomena. In the present investigation we have laid emphasis on (i) the determination of trap parameters corresponding to the various TL glow peaks observed (both natural and γ -ray induced), (ii) deducing the γ -ray equivalent dose in natural samples and investigating the saturation properties of various glow peaks, and (iii) attempting to interpret the history of lunar radiation and temperature environment from the above information.

EXPERIMENTAL PROCEDURE

The samples investigated by us were (i) unsorted contingency fines 12070,112, and (ii) an interior rock chip 12051,15 (NASA type-code AB, fine to medium grained crystalline igneous rock (olivine basalt); part of a documented sample). The fines were in their natural (as received) state, while the chip had previously been irradiated at AERE, Harwell, with 160 MeV protons to an estimated dose of 200 krad (Blair *et al.* 1971).

Our experiments were carried out on two different sets of TL apparatus. On the first (called hereinafter set A), the amount of sample used was normally ≈ 2 mg, and the linear rate of heating β was 5°C sec^{-1} . On the second set (called B), the amount of sample used was normally ≈ 10 mg, and $\beta = 20^\circ\text{C sec}^{-1}$. The samples used in all cases were in powder form, and generally sieved to obtain various grain-size ranges; the rock samples were first crushed lightly in a mortar. The samples were spread uniformly over a tantalum strip and heated in an oxygen-free nitrogen atmosphere. The rate of heating was kept constant at a predetermined level with the help of a programmed temperature controller operating via a thermocouple welded to the underside of the heating strip directly below the powder. A reflecting cone with an aperture at its apex (positioned over the sample) was used to limit the amount of black-body radiation received by the photomultiplier (PM). The photomultipliers used were specially selected tubes with low dark current. In set A, a quartz photomultiplier (EMI: 6256 SA) was used in a cooled housing (EMI assembly TE 102 TS, to cool the PM to -20°C), thus further reducing the dark current. Set B used a glass PM tube (EMI: 9502 SA). The PM output, measured by a sensitive electrometer, was recorded on an X - t , Y chart recorder. Set A had an extra pen which could record the integrated glow current simultaneously. Between the limiting cone and the PM a heat-shield glass window, and the filter in use, were interposed. For normal work, a broadband filter was always employed, mainly to reduce black-body interference (on set A: Ilford "Bright Spectrum Blue" filter No. 622, transmission band 375 nm to 530 nm; on set B: Ilford "Bright Spectrum Violet" filter No. 621, transmission band 340 nm to 515 nm).

RESULTS

Natural and artificially induced glow curves

Both the natural TL and that following γ -ray irradiation (from a 200 Ci cobalt-60 source) were studied in the unsorted virgin fines. Similar studies were carried out on the powdered rock chip sample (except that it had previously been subjected to a dose of 200 krad of 160 MeV protons). In what follows, TL apparatus A (with a heating rate $\beta = 5^\circ\text{C sec}^{-1}$) was always used, unless otherwise specified. Carefully weighed

samples (usually of a preselected grain-size range but not otherwise separated) of approximately 2 mg were used for each run. Fresh samples were used for successive runs in order to avoid “temperature-sensitization” effects.

Figure 1 shows typical glow curves from the fines samples, both in their natural state and when subjected to artificial γ -ray doses ranging from 35 to 2700 krad (known to an accuracy of $\sim \pm 10\%$). Each of the curves B–E was taken within a fairly short time (~ 15 min to 1 hour) after the end of the corresponding irradiation to minimize effects of thermal drainage at room temperature. It will be noticed that there is virtually no natural TL below 250°C ; above this temperature there is a broad hump extending to $\sim 550^\circ\text{C}$, containing more than one unresolved peak (note that the black-body radiation has been subtracted in all cases, though beyond $\sim 500^\circ\text{C}$ the subtraction becomes uncertain). As increasing amounts of γ -ray dose are added, a prominent new peak (termed peak I) is introduced around 175°C ; it slowly moves towards lower temperatures as the artificial dose is increased (thereby enhancing the relative “weight” of peak I). The TL in the temperature region $300^\circ\text{--}550^\circ\text{C}$ (seen later by the initial rise method to contain peaks II and III) grows gradually with the γ -ray dose imparted, thus indicating that these peaks have not attained saturation in nature.

Figure 2 shows the TL glow curves from powdered samples of the rock chip 12051,15. Curve A represents TL from a natural sample (containing ~ 200 krad of

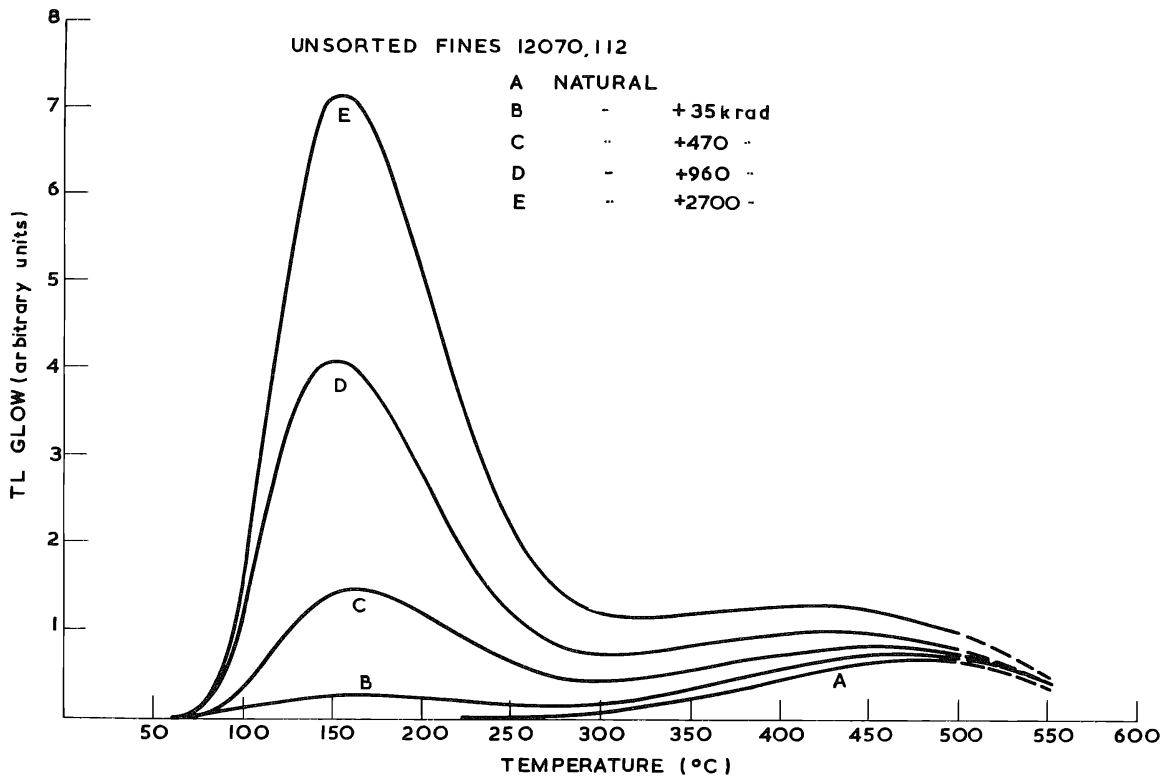


Fig. 1. Typical TL glow curves from the fines sample (grain diameter $d \leq 106 \mu$; rate of heating $\beta = 5^\circ\text{C sec}^{-1}$; a new 2.0 ± 0.1 mg sample used for each irradiation). The black-body contribution has been subtracted (though with increasing uncertainty beyond $\sim 500^\circ\text{C}$); Ilford broad-band filter 622 used.

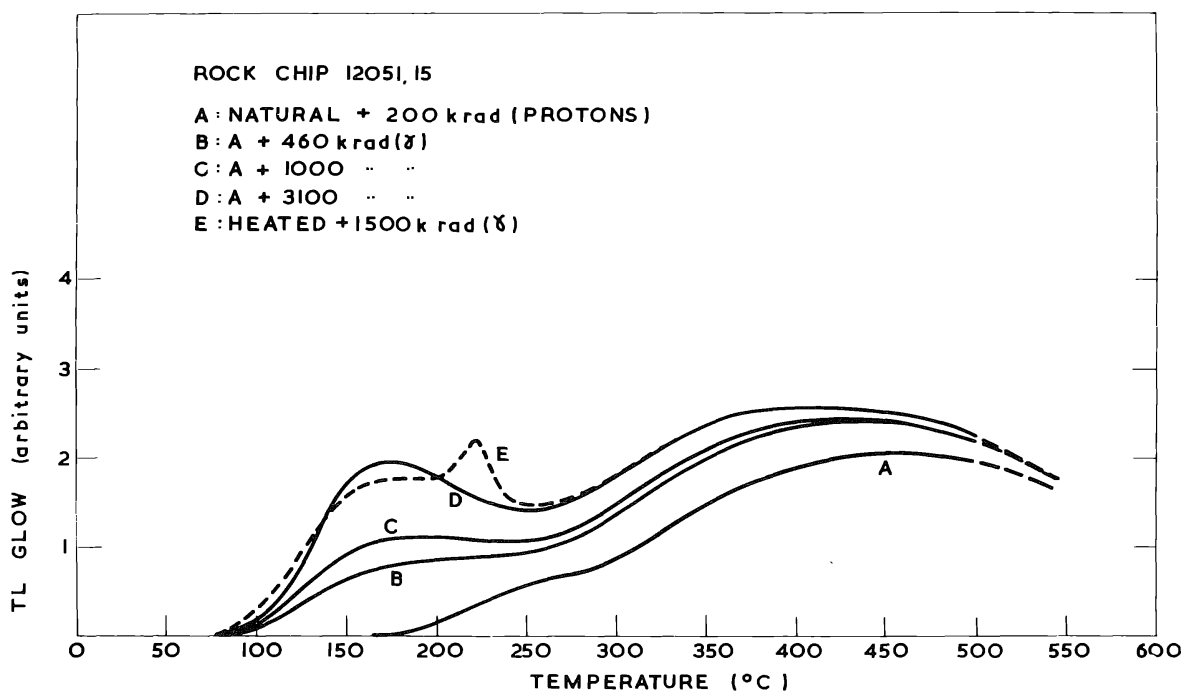


Fig. 2. Typical glow curves from (powdered) rock samples. Experimental conditions, and the scale of the ordinate, are as in Fig. 1 (but $d < 125 \mu$ for curves A–D). The rock sample had, in all cases, an initial dose of ~ 200 krad of 160 MeV protons. The “pip” in curve *E* is discussed in the text. Notice the higher TL response of the heated sample ($d \sim 106$ – 125μ). The near-equality of the areas under curves D and E results from the fact that the sample approaches saturation at ~ 1500 krad (see Fig. 5, 20– 300°C).

160 MeV protons, i.e., a total dose N' , say); while further doses of γ -rays (ranging from 460 krad to 3100 krad) have been superimposed upon N' in the case of curves B to D. The glow structure and its growth are broadly similar to those of fines (the hump in the region of $\sim 200^\circ$ – 300°C is compatible with being the room-temperature remnant of the proton irradiation, which had been carried out some three months earlier).

Two points in Fig. 2 are worth noting. First, the rate of growth of peak I is much lower relative to the same peak in Fig. 1 (both drawn to the same scale in the ordinate): thus the relative heights of peak I at ~ 1 Mrad imparted γ dose are $\sim 1:4$ for rock to fines samples. Secondly, a sharp hump (at $\sim 225^\circ\text{C}$) is observed from rock samples if powdered samples from a previous TL read-out are re-irradiated and then used for TL measurements. This was a persistent phenomenon and a number of experiments were carried out to investigate possible causes. The feature is further discussed below.

Spectral response

A series of Ilford filters (nos. 600, 607, and 621–626; typical band width at half-height ~ 50 nm) were used on apparatus B to determine the glow spectrum from various traps in the rock sample 12051,15 after irradiation with 270 krad of γ -rays. The photomultiplier current generated at any given temperature of read-out is a function of the emitted TL intensity $I(\lambda)$, the transmission characteristics $F(\lambda)$ of the filter in use, and the quantum efficiency $Q(\lambda)$ of the photomultiplier, λ being the

wavelength. It is possible, by assuming as a first approximation that the intensity I at a given temperature is a slowly varying function of λ over the waveband of the filter, to deduce the (relative) value of $I(\lambda)$ from the observed current and from the known values of $Q(\lambda)$ and of $F(\lambda)$ for the various filters employed. This is done by carrying out the numerical integration $\int F(\lambda) Q(\lambda) \lambda d\lambda$ over the transmission band of each filter in turn (and by deriving a self-consistent solution by reiteration if necessary, though in our case it was not found to make a significant difference).

Figure 3 shows the emitted TL spectrum at three different temperatures (450° , 350° , and 200°C). The main conclusions derived from these and other similar curves (not shown) are that (i) very little TL intensity is observed at wavelengths ≥ 550 nm; and (ii) although at all temperatures the short wave length component (~ 380 – 490 nm, violet to blue) predominates, yet the long wavelength part (~ 500 – 570 nm, green to yellow) constitutes a more significant fraction of the total output at lower temperatures of emission ($\lesssim 300^\circ\text{C}$).

Dose response and saturation effects

In Figs. 4 and 5 are shown the growth of the TL glow (integrated over the temperature intervals 400° – 500°C , 300° – 400°C , and 20° – 300°C) as increasing amounts of γ -ray dose are imparted to the fines and the rock chip samples, respectively. Both types of sample show saturation in their 300° – 500°C TL output at natural + ~ 2.5 Mrad γ -ray dose. By extrapolating backward the initial trend of the TL growth curve (see equation (6) below) it is possible to estimate the natural TL in each case. For

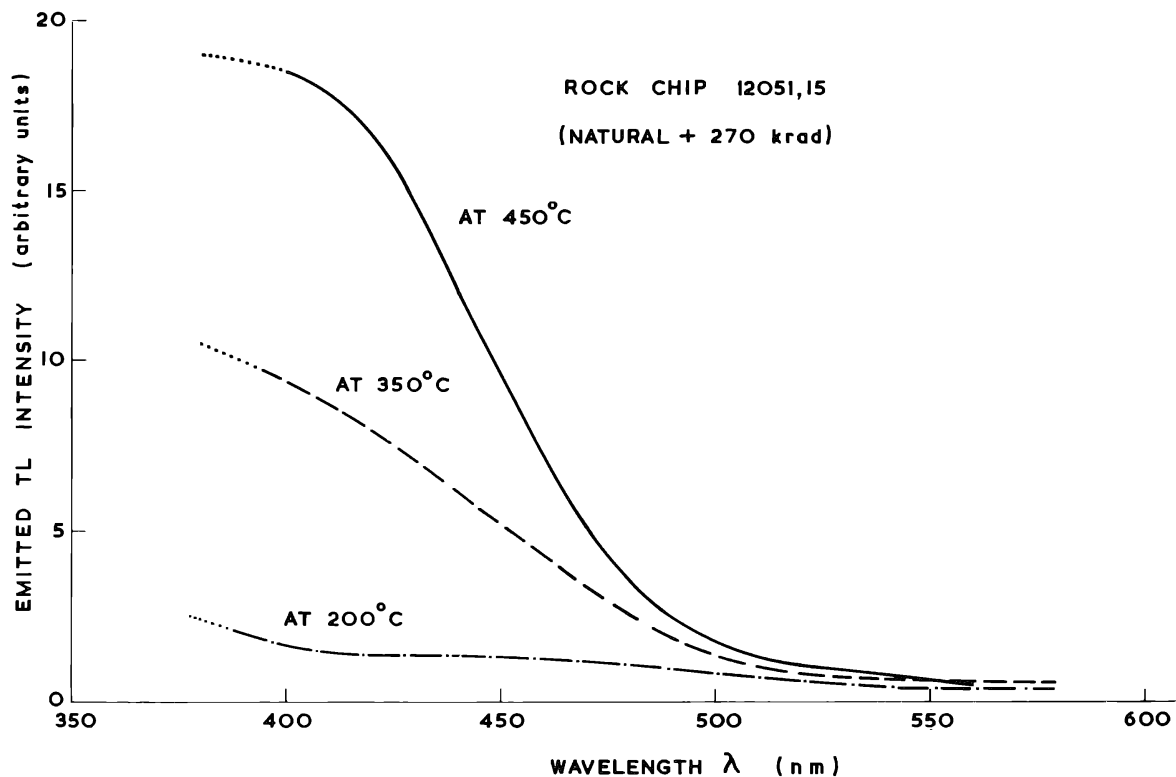


Fig. 3. Spectrum of TL emitted at three different temperatures by the irradiated rock sample (grain size, $125 \mu < d \leq 355 \mu$). See text for method of analysis.

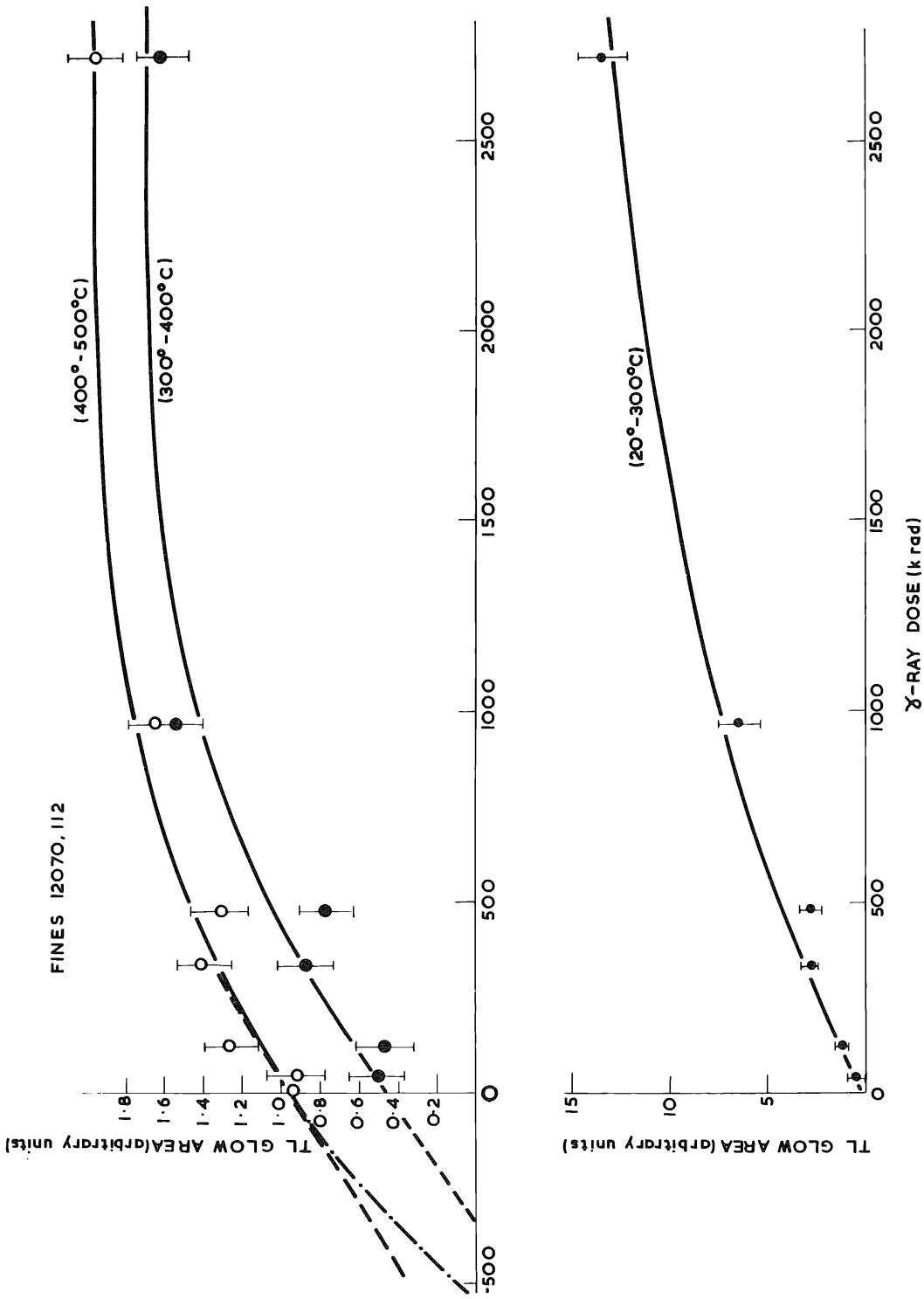


Fig. 4. Dose response of the fines (grain size $d < 106 \mu$). By extrapolation of the initial trend of the TL growth curve (shown as a series of dots and dashes (-.-.-) for the top curve (400-500°C)), the natural (or equilibrium) dose in each sample may be estimated (see equation (9)). The dashed line (---) is merely the tangent to the curve at the origin. The error bars result from repeated measurements, intrinsic inhomogeneities of samples, weighing uncertainties, etc.

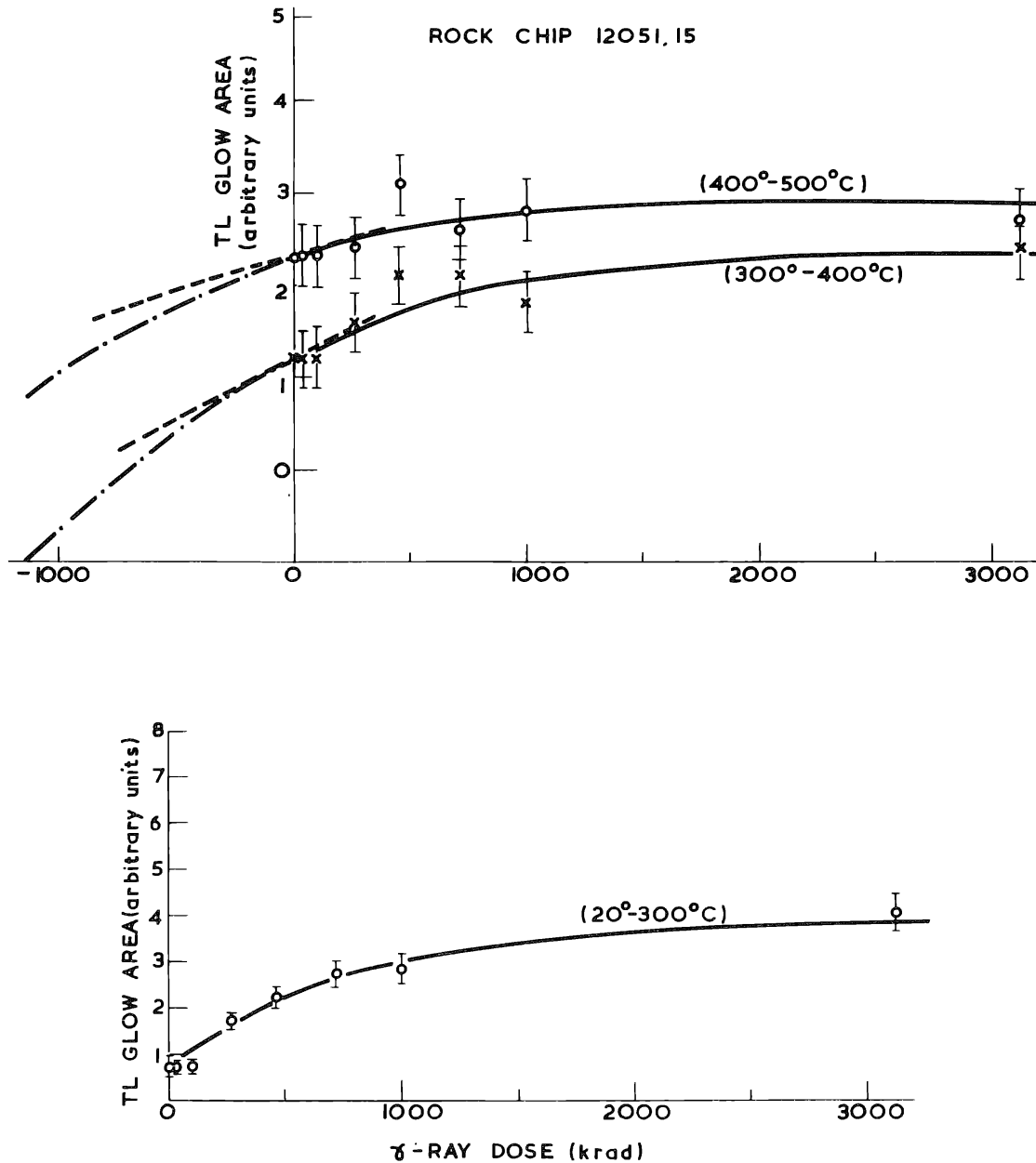


Fig. 5. Dose response of the rock sample ($d < 125 \mu$) drawn to the same scale in the ordinate as Fig. 4. By extrapolation of the initial trend of the TL growth curve, the natural (or equilibrium) dose in each sample may be estimated (see discussion in text).

For other information see caption to Fig. 4.

the fines, the natural dose in the temperature interval 400° – 500°C is estimated to be ~ 550 krad, and in the rock chip ~ 1700 krad; while the estimates for the temperature interval 300° – 400°C are: ~ 350 krad (fines), and ~ 1100 krad (rock chip).

The fact that the natural dose corresponding to the higher temperature region is larger than that for the lower region is reasonable, as the traps in the former case are deeper and are therefore better able to retain their dose. It must, however, be pointed

out that there is a considerable scatter in the TL output from individual 2 mg samples. This effect, which is attributable to inhomogeneities in the distribution of the TL phosphors, is discussed below.

TL parameters

In discussing the thermal and radiation history of a thermoluminescent material, it is essential to know the values of the basic TL parameters E (trap depth) and s

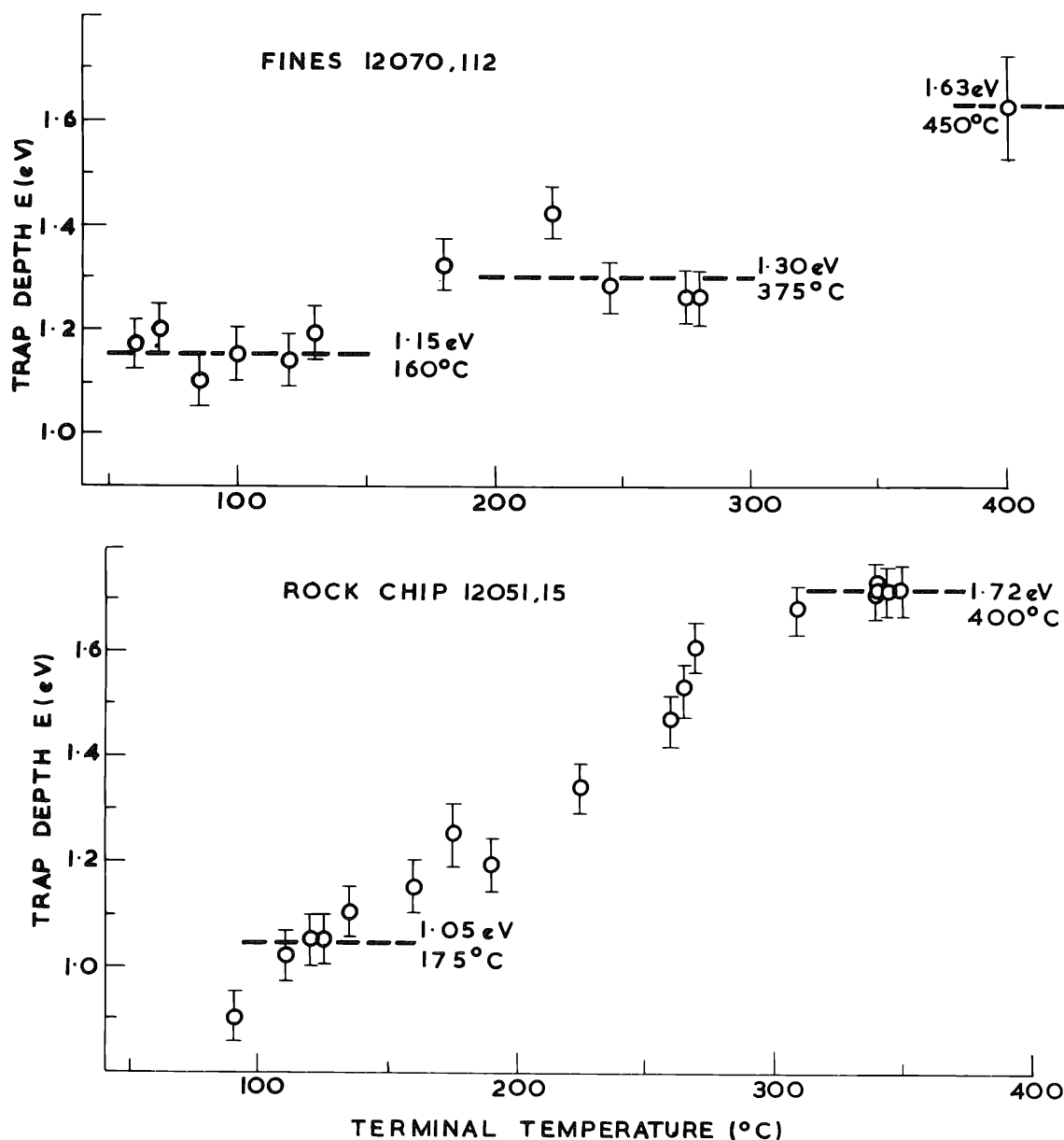


Fig. 6. Determination of the trap depth E for the various peaks in the fines and the rock samples by the initial rise method. The samples are heated to successively increasing terminal temperatures (followed by cooling). The horizontal clusters represent the peaks whose E and T^* values are marked. The values of T^* were obtained by a separate series of experiments involving the "thermal cleaning" of peaks.

(frequency factor) for it. In the first-order kinetics, these parameters are related as follows (Randall and Wilkins, 1945)

$$n(t, T) = n_0 \exp(-s \cdot t e^{-E/kT}) \quad (1)$$

where n is the number of filled electron traps at time t in a body kept at an absolute temperature T , n_0 is the number at $t = 0$, and k is Boltzmann's constant. This leads to a mean life τ ($= \tau_{\frac{1}{2}}/\ln 2$) for the trapped electrons at temperature T , given by

$$\tau(T) = \tau_0 \cdot e^{E/kT} \quad (2)$$

where $\tau_0 = s^{-1}$ (and is characteristic of the trap).

There are several ways of determining E and s experimentally in favorable circumstances. The one we found satisfactory for the Apollo 12 samples is the so-called initial rise method (Garlick and Gibson, 1948). This method is based on the fact that, at temperatures sufficiently below the peak temperature T^* for a given trap, the TL intensity $I(T)$ rises with temperature as $e^{-E/kT}$, irrespective of the order of kinetics and the rate of heating β involved. On plotting $\ln I$ against $1/T$ the initial part of the glow curve thus yields a straight line with slope $-E/k$. A number of heatings (interrupted at successively increasing temperatures, and followed by cooling) yield a series of values of E , until deviations from linearity become significant when the temperature of a glow peak T^* is approached. Figure 6 shows the resulting E values, plotted against the temperature of interruption, for both types of sample. The horizontal clusters correspond to the trap depths for the respective peaks. The s factor is then calculated from the (first-order kinetics) relation

$$\beta \cdot E/kT^{*2} = s \cdot e^{-E/kT^*} \quad (3)$$

Table 1 records the trap parameters for the peaks found in the fines and the rock chip samples by the initial rise method. Also shown in the table are the half-lives $\tau_{\frac{1}{2}}$ of trapped electrons corresponding to these peaks, computed from equation (2), at -30°C (approximately the mean temperature of the moon unaffected by the diurnal heat wave), 20°C (room temperature), and 120°C (approximately the maximum lunar surface temperature).

Table 1. Trap parameters* and half-lives of peaks at different storage temperatures

Sample	Peak			Frequency factor s (sec^{-1})	Half-life $\tau_{\frac{1}{2}}$ at		
	No.	Temp. ($^\circ\text{C}$)	Trap depth E (eV)		-30°C	$+20^\circ\text{C}$	$+120^\circ\text{C}$
12070,112 (Fines)	I	160	1.15 ± 0.05	$\sim 7 \times 10^{12}$	2×10^3 yr	73 days	55 sec
	II	375	1.30 ± 0.05	$\sim 2 \times 10^9$	1×10^{10} yr	2.5×10^5 yr	182 days
	III	450	1.63 ± 0.10	$\sim 4 \times 10^{10}$	4×10^{15} yr	7×10^9 yr	5×10^2 yr
12051,15 (Rock chip)	I	175	1.05 ± 0.05	$\sim 2 \times 10^{11}$	6.5×10^2 yr	47 days	100 sec
	II	400	1.72 ± 0.05	$\sim 1 \times 10^{12}$	1×10^{16} yr	7×10^9 yr	3×10^2 yr

* Determined by the initial rise method (see text); dose given: 3.9 mrad of γ -rays; rate of heating: 5°C sec^{-1} .

DISCUSSION

Determination of lunar temperature and radiation dose rate

A good deal of information on the thermal and radiation history of samples may be gleaned from studying their natural and artificially induced TL curves. Thus it is seen (Fig 1) that peak I in the natural samples of lunar fines has been completely drained. This is readily understood since, from Table 1, its half-life at room temperature is only 73 days, whereas the experiments were performed some 18 months after retrieval of the samples from the moon. This underlines the importance of placing lunar samples, destined for TL study, in deep freeze (say at liquid nitrogen temperature, though even -20°C storage would help), as well as of their prompt distribution for such work. Vital information, particularly from core tube samples which had previously remained at low ambient temperatures on the moon, may otherwise be irretrievably lost.

Glow peaks which have not reached saturation in nature (e.g., the peaks above $\sim 350^{\circ}\text{C}$ in Figs. 1 and 2) but have attained a dynamic equilibrium in the prevailing radio-thermal environment (as a result of competition between the thermal emptying and radiative filling of the corresponding traps) can be used to estimate the environmental temperature if a certain dose rate is assumed, and vice versa. For details of the general method, reference may be made to Christodoulides *et al.* (1971) or Hoyt *et al.* (1971).

Briefly, consider a sample with N traps of depth E and frequency factor s irradiated with a dose rate r while being held at a temperature T . Then, if p is the probability per unit dose of filling one such trap, the number n of traps filled at time t is given by

$$n = \frac{N}{1 + \frac{s}{rp} \cdot e^{-E/kT}} \cdot \left\{ 1 - \exp \left[-rp \left(1 + \frac{s}{rp} \cdot e^{-E/kT} \right) t \right] \right\} \quad (4)$$

The number n_{eq} of traps filled at equilibrium, for large t , is thus

$$n_{\text{eq}} = \frac{N}{1 + \frac{s}{rp} \cdot e^{-E/kT}} = \frac{N}{1 + 1/r\tau p} \quad (5)$$

where, in the second equality in equation (5), we have used $\tau = \frac{e^{E/kT}}{s}$ given by equation (2).

If the sample is irradiated artificially at such high dose rates (or at such low temperatures) that decay (or drainage) of filled traps may be ignored, then equation (4) reduces to

$$n = N(1 - e^{-rt \cdot p}) \quad (6)$$

The probability p may then be expressed in terms of observable quantities by considering the total artificial dose rt for which the emitted TL under the glow peak is just half of the saturation TL (i.e., $N/2$ traps are filled). Such a dose may be termed $R_{\frac{1}{2}}$, or "half-dose" on the analogy of half-life; then

$$p = 0.693/R_{\frac{1}{2}} \quad (7)$$

Substituting p in equation (5), we obtain

$$n_{\text{eq}} = \frac{N}{1 + \{(R_{\frac{1}{2}}/r) \cdot se^{-E/kT}/0.693\}} = \frac{N}{1 + (R_{\frac{1}{2}}/r\tau_{\frac{1}{2}})} \quad (8)$$

where $\tau_{\frac{1}{2}} = 0.693\tau$.

If we wish to determine the artificial dose R_{eq} that would produce the same amount of TL in a sample (starting with all traps initially empty) as it had when at equilibrium in nature (usually called natural dose), we solve equation (6) (with $rt = R_{\text{eq}}$ and $n = n_{\text{eq}}$) simultaneously with the second equality in equation (8) to obtain the useful relation

$$R_{\text{eq}} = \frac{R_{\frac{1}{2}}}{0.693} \cdot \ln \left(1 + \frac{r\tau_{\frac{1}{2}}}{R_{\frac{1}{2}}} \right) \quad (9)$$

This formula is applicable to curves such as those in Figs. 4 and 5.

The ratio of the natural TL under the glow peak emitted by a sample at equilibrium to the TL emitted by an artificially saturated sample gives us the value of n_{eq}/N . Then, if the dose rate r in nature is known, the first of the equalities in equation (8) allows us to calculate T . Thus

$$T = \frac{E/k}{\ln \left[\frac{sR_{\frac{1}{2}}}{0.693r \left(\frac{N}{n_{\text{eq}}} - 1 \right)} \right]} \quad (10)$$

The value of T is only weakly dependent on the values of r and N/n_{eq} used, since they occur in the logarithmic term—indeed, a factor of 2 in the product $r \cdot [(N/n_{\text{eq}}) - 1]$ changes the calculated value of T , in the cases of interest to us, by only $\sim 1\%$ in equation (10).

We have used the characteristics of peaks III and II in lunar fines and the rock chip, respectively, to calculate from equation (10) the effective lunar storage temperature in each case (see Table 2). The dose rate from galactic cosmic rays (in “ 2π geometry”) in the vicinity of the earth is known (Haffner, 1967) to be ~ 9 rad/y, with a comparable contribution from solar flares. The efficiency of these radiations, relative to 1 MeV β and γ , in producing TL in materials is unknown; but preliminary experiments carried out by two of us (S.A.D. and W.P.) on meteorites bombarded with 7 GeV protons suggests the relative efficiency to be close to 1.

If the lunar temperature is assumed, for simplicity, to vary sinusoidally about a mean temperature T_0 with an amplitude A such that

$$T(\phi) = T_0 + A \sin \phi \quad (11)$$

where ϕ is the phase angle, the temperature T_{eff} (which will have the same effect on TL as the actual cycle) may be defined by the relation.

$$e^{-E/kT_{\text{eff}}} = \frac{1}{2\pi} \int_0^{2\pi} e^{-E/kT(\phi)} d\phi \quad (12)$$

The temperature T_0 of a body in space at 1 AU from the sun may be taken as 240°K, and the value of A at the surface of the moon, say A_s , as 150°K (Kopal, 1969). The amplitude of temperature modulation is sharply attenuated with depth d

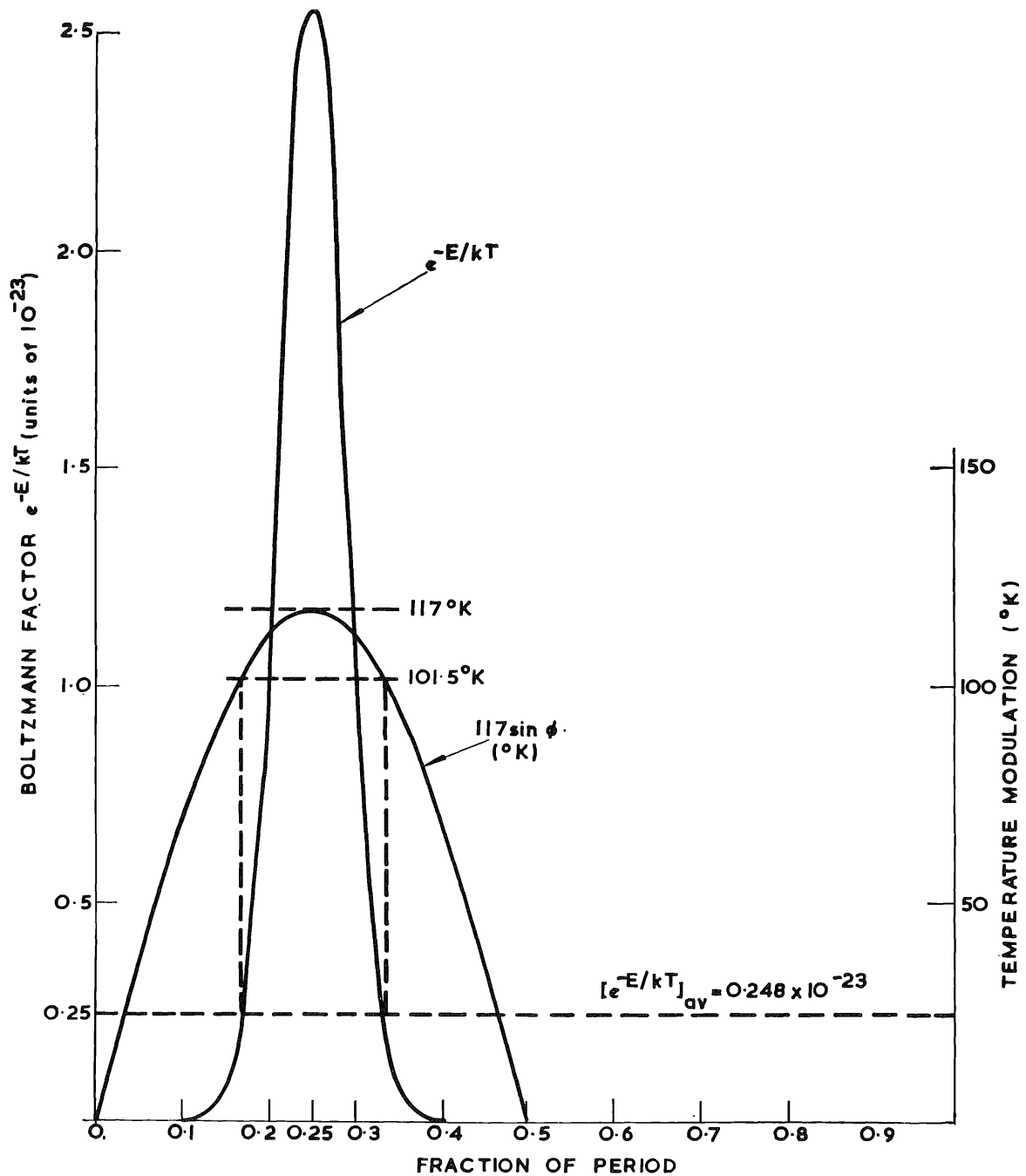


Fig. 7. Determination of the "effective temperature" T_{eff} that has the same effect on draining the TL as a sinusoidally varying temperature cycle on the moon. The average value $[e^{-E/kT}]_{\text{av}}$ is such that, if maintained over a lunation, it will have the same area under it as $\exp(-E/kT(\phi))$. By projecting the points of intersection of the average line and the Boltzmann factor onto the modulation curve, T_{eff} can be found graphically. (Alternatively, T_{eff} may be found by numerical integration.)

below the lunar surface. Following Hoyt *et al.* (1971), we assume a value of 24 cm for λ , the thermal wavelength for Apollo 12 fines, in the relation

$$A = A_s e^{-2\pi d/\lambda} \quad (13)$$

Figure 7 shows how the integral in equation (12) may be determined graphically. With $E = 1.6$ eV and $A = 117^\circ\text{K}$ (this being the amplitude at $d \approx 1$ cm), the value of T_{eff} is found to be 341.5°K , which may be compared with $T_{\text{max}} = 240^\circ\text{K} + 117^\circ\text{K} = 357^\circ\text{K}$ (at 1 cm depth).

It is possible to use the effective storage temperature found for the fines sample (Table 2) to estimate the mean depth below the lunar surface from which it came. Thus $T_{\text{eff}} = 358^\circ\text{K}$ is found to correspond to $A = 136^\circ\text{K}$ (i.e., $T_{\text{max}} = 376^\circ\text{K}$), which yields a value of mean depth $d \approx 0.4$ cm.

As for the solid rock sample, if the thermal conductivity of lunar regolith in the top 1 m, reported from the Apollo 15 heat flow measurements (Langseth *et al.*, 1972) to be ~ 10 times that for Apollo 12 fines, is used, this results in a $\lambda \sim 70$ cm. The temperature of the rock chip ($T_{\text{eff}} = 346^\circ\text{K}$, i.e., $T_{\text{max}} = 362^\circ\text{K}$ or $A = 122^\circ\text{K}$) then yields a value of mean subsurface depth $d \approx 2.5$ cm. This depth, however, must only be treated as approximate in view of all the uncertainties involved.

Finally, if one takes the storage temperature as known, it is possible to estimate the dose rate experienced by the sample by using equation (9)—though not very accurately because of the logarithmic term involved. The value of $T_{\text{eff}} = 358 \pm 10^\circ\text{K}$ for the fines leads to a half-life $\tau_{\frac{1}{2}} \sim 2 \times 10^4$ yr (varying by a factor of ~ 3 either way owing to the $\pm 10^\circ\text{K}$ uncertainty in temperature). Hence the natural dose $R_{\text{eq}} \sim 550$ krad (corresponding to the temperature interval $400^\circ\text{--}500^\circ\text{C}$ for peak III in the fines (Table 2) yields a value of ~ 30 rad/y for the dose rate, accurate to a factor of ~ 3 either way. The value estimated for the rock chip using peak II (Table 2) is found, in a similar way, to be ~ 60 rad/y within a factor of ~ 2 either way.

It is clear from the foregoing that useful quantitative inferences can be drawn regarding the thermal and radiation histories of lunar samples by studying their TL. This will be increasingly true in future when data from the temperature sensing probes to be installed during the forthcoming Apollo missions (Langseth *et al.*, 1972) become available so that some of the uncertainties regarding the temperature gradients in core tube samples can be eliminated (no such samples were available for use in the present investigation).

Table 2. Effective lunar storage temperature T_{eff} calculated from equation (10), assuming a dose rate of 10 rad/year*

Sample	Peak		Trap depth E (eV)	Frequency factor s (sec^{-1})†	Half-dose $R_{\frac{1}{2}}$ (krad)	N/n_{eq}	T_{eff} ($^\circ\text{K}$)
	No.	Temp. ($^\circ\text{K}$)					
12070,112 (Fines)	III	723	1.63 ± 0.10	3.8×10^{10}	500	2.1	358 ± 10
12051,15 (Rock chip)	II	673	1.72 ± 0.05	1.4×10^{12}	500	1.3	346 ± 5

* For explanation of symbols, see text. Values of $R_{\frac{1}{2}}$ and N/n_{eq} are for the temperature interval $400\text{--}500^\circ\text{C}$. A change in the dose rate r by a factor of 2 changes the value of T_{eff} by $\sim 1.3\%$.

† The value of s changes inversely by a factor of ~ 2 for each 0.05 eV change in E .

One interesting use of the above calculations of the lunar temperatures is to speculate on the temperature history of the earth's surface over the corresponding period of $\sim 10^4$ – 10^5 yr during which the equilibrium doses have been accumulated by the lunar samples. If the constancy of lunar temperatures over this period is accepted as a guide, then the glaciation cycles of the earth during the last $\sim 100,000$ yr cannot be attributed to sustained increases in solar heat output leading to enhanced cloud formation and precipitation of snow near the poles. Other causes for the glaciation pattern will then have to be invoked. Of course the existence of periods of low solar heat output in the past cannot be excluded on the above evidence.

Inhomogeneities and temperature effects in samples

In interpreting thermoluminescence data, a constant source of uncertainty is the variation in the TL output per unit mass from sample to sample. This is attributable to inhomogeneities in the distribution of phosphors responsible for TL, as first pointed out by Fremlin and Srirath (1964) in the case of pottery sherds, and also noted by Durrani and Christodoulides (1969) in meteorites. These nonuniformities are liable to be accentuated when small amounts of (independent) samples have to be used, as in the present investigation. The effect was particularly noticeable in the case of lunar fines, as can be seen from the scatter of experimental points on the dose response curve (Fig. 4). The ultimate solution of the problem may well be to work with individual grains of separated minerals as attempted by Hoyt *et al.* (1972).

It may be argued that better uniformity in TL response might be achieved by using physically the same sample over and over again after giving it varying artificial doses. But here one comes up against the problems of temperature sensitization and predose effects (Zimmerman *et al.*, 1965); for instance some tektite and meteorite samples have been shown (Durrani *et al.*, 1970; Christodoulides *et al.*, 1970) to yield six to eight times as much TL per unit dose after being annealed as prior to it. In the case of Apollo 12 samples, it was established that, for a test dose of 920 krad for fines and 1500 krad for the rock chip, the sensitivity of a given sample preheated to 500°C was enhanced by ~ 30 – 40% for fines and by ~ 10 – 20% for the rock.

Another effect of temperature which we noticed in the present investigation is the sharp hump (at $\sim 225^\circ\text{C}$) visible in heated (and then irradiated) rock samples (Fig. 2). A number of subsidiary experiments were carried out to exclude possible causes (chemiluminescence; vacuum grease used in the previous proton-irradiation experiments; TaO_2 from the heating strip; oxidation of samples). It was established that the hump was observed only if the rock powder had been previously heated to at least 500°C; it became much more marked as a result of pre-heating up to 600°C. The area under the hump grew with dose and roughly in proportion with the body of the main curve. The only explanation that can be offered at this stage (in the absence of a definitive crystallographic analysis) is that a phase change brought about by heating the sample to above 500°C is apparently responsible for the phenomenon. No such "pip effect" was observed in the case of fines, or of any Apollo 14 samples subsequently examined.

In an effort to minimize the intrinsic inhomogeneities in the samples used, grain-

size control was implemented in each series of related TL observations, as mentioned earlier. The advantage of such a procedure was established by a subsidiary investigation in which grains from unirradiated fines, subdivided into four groups by size (with diameters d ranging from $20 \mu < d \leq 53 \mu$ up to $d > 355 \mu$), were examined for TL output. It was found that the specific TL (output per unit mass) went down monotonically as the grain size increased (and the surface to volume ratio fell). The 2 mg samples were always spread out uniformly and thinly enough on the heating strip that loss of glow received by the photomultiplier through layering and intergrain scattering of light was minimal. Since each 2 mg sample consisted of several thousand grains, it was hoped that the phosphors chiefly responsible for TL output would be reasonably randomly distributed between samples (short of actual mineralogical separation) and that at least specific TL would be kept within narrow limits by means of grain-size control.

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