Other Physical Properties

Thermoluminescence of Lunar Samples

Abstract. Appreciable natural thermoluminescence with glow curve peaks at about 350 degrees centigrade for lunar fines and breccias and above 400 degrees centigrade for crystalline rocks has been recognized in lunar samples. Plagioclase has been identified as the principal carrier of thermoluminescence, and the difference in peak temperatures indicates compositional or structural differences between the feldspars of the different rock types. The present thermoluminescence in the lunar samples is probably the result of a dynamic equilibrium between acquisition from radiation and loss in the lunar thermal environment. A progressive change in the glow curves of core samples with depth below the surface suggests the use of thermoluminescence disequilibrium to detect surfaces buried by recent surface activity, and it also indicates that the lunar diurnal temperature variation penetrates to at least 10.5 centimeters.

Thermoluminescence (TL) studies on samples from two medium-grained crystalline rocks (type B), two breccias (type C), and lunar fines (type D, less than 1 mm) that included material from the bulk sample and from core No. 1 were undertaken to determine properties of lunar material and to investigate possible uses of TL for the study of lunar geology (1).

Between 5 and 20 mg of material, in powdered form, was mounted in silicon oil on a small aluminum disk and heated at a rate of 20°C/sec by focused infrared radiation from below. The thermoluminescence was measured by a photomultiplier tube with a Cs₃Sb cathode, and the glow curve and the area under the glow curve were recorded as a function of temperature on an x-y-y' recorder. The instrument was calibrated with a ¹⁴C-activated light source of known luminous flux, which made it possible to read the height of a TL peak in lumens and the amount of TL (the area under the glow curve) in lumen-sec. Samples were irradiated with x-rays of 14-kev effective energy, and the doses were monitored with an integrating dosimeter.

The TL measurements on the fines (sample 10084) from the bulk sample box yielded a glow curve with a single weak peak at about 350°C. In order to identify the TL carrier, a portion of the fines was sieved and separated into fractions with different relative proportions of the major mineral constituents, by means of heavy-liquid and magnetic

30 JANUARY 1970

techniques. The natural TL was measured on portions from nine fractions, including the untreated fines. All of these natural glow curves have a single peak at about 350°C. The same samples were then exposed to 5000 rad of x-radiation, and their "artificial" glow curves were measured 3 minutes after exposure. The artificial glow curves have nearly identical shapes, with prominent peaks at 125° and 185°C and evidence of less prominent peaks at about 260° and 350°C. The ratio N/A was used to compare the relative TL sensitivities of the nine fractions. wher, N is the area per milligram under the natural glow curve from 300° to 350°C and A is the area per milligram under the artificial glow curve from 85°



Fig. 1. Glow curves, showing the decay of artificial TL (5000-rad) as a function of time at 100°C for the 45- to 75- μ m fraction of the lunar fines. *BB*, Blackbody curve.

to 350° C. Although the areas differ by more than an order of magnitude (N from 0.44 to 17.6 nlumen-sec/mg) and A from 6.0 to 283 nlumen-sec/mg), N/A is nearly constant for all fractions, and has a value of 0.063 ± 0.006 (S.D.). The similarity of the natural glow curves, the similarity of the artificial glow curves, and the constancy of N/A show that a single mineral is responsible for the TL. Both N and A increase as the amount of feldspar in the nine fractions increases, which indicates that plagioclase is the primary TL carrier.

In order to observe the change in TL as a function of time and temperature, the following decay experiments were performed on material from the fines: (i) decay of 5000-rad artificial TL in air at room temperature ($\approx 22^{\circ}$ C); (ii) decay of 5000-rad artificial TL in vacuum at 100°C; and (iii) decay of natural TL in vacuum at 100°C. At room temperature, the 125°C peak is almost entirely gone within a few days, but measurable TL in the region above 150°C remains even after a period of 4 weeks. At 100°C the artificial TL of the 125°C- and 185°C-peaks is completely gone by 30 hours (Fig. 1). The 260°C peak is prominent after 1 week at 100°C, but after 4 weeks it has greatly decayed so that the shape of the glow curve closely resembles that of the natural glow curve. There is noticeable decay of the natural TL with storage at 100°C; by 4 weeks the area under the natural glow curve between 300° and 350°C is reduced by approximately one-third.

Although difficult to measure accurately because of mutual interference, the half-lives of the various peaks can be estimated from the decrease in area under the glow curves and the change in peak heights with time. The 125 °C and 185 °C peaks appear to have half-lives of less than 2 weeks at room temperature and will be of little value for lunar geologic studies. At 100 °C ambient temperature, the 260 °C peak decreases with an apparent half-life of about 2 weeks, and the 350 °C peak



Fig. 2. Glow curves on samples from core No. 1 (10005), showing the change in natural TL with depth below the lunar surface. BB, Blackbody curve.

decreases with a half-life of about 1 to 2 months. These half-lives were compared with values calculated from a simple exponential model (2) with

 $t_{\frac{1}{2}} = (\ln 2/s) \exp (E/kT)$ where

$$= (qE/kT_{\rm p}) \exp (E/kT_{\rm p})$$

The activation energy E is approximately 25 $kT_{\rm p}$, q is the heating rate, $T_{\rm p}$ is the temperature of the glow curve peak, kis Boltzmann's constant, and T is the isothermal decay temperature. For isothermal decay at 100°C this approximation yields $t_{1/2}$ values for peaks at 260°C and 350°C of about 1 day and 11 months, respectively; the corresponding values of $t_{1/2}$ for decay at 22°C are 18 years and 60,000 years. The model and the limited experimental data are in reasonably good agreement inasmuch as they predict a rather short lifetime for both peaks at 100°C. In addition, the model predicts that energy storage in the form of TL from ionizing radiation is highly temperature-dependent. A change of only 20°C in the isothermal decay temperature will change the half-life of a peak by more than a factor of 10.

The short lifetime and high degree of temperature-dependence of TL indicate that lunar surface temperatures have had a significant influence on the observed TL. Thus the glow curves from material collected near the surface, as the Apollo 11 samples were, will retain only an imperfect record of the radiation history of the samples, and it seems doubtful that TL studies on such material can result in meaningful radiation exposure ages. Finally, the apparent great age of the Apollo 11 samples (1) and the short half-life of TL in the lunar surface environment suggest that the TL observed in a nearsurface sample is probably the result of a dynamic equilibrium between the growth of TL due to radiation and its decay in the lunar thermal environment.

The glow curves of untreated material from core No. 1 show a systematic change with depth below the lunar surface (Fig. 2). Comparison of the 5000-rad artificial glow curves further shows that the differences observed in the natural TL are not due to differences in either mineral content, mineral type, or susceptibility to radiation. The TL of the sample from the top of the core is nearly identical to the TL of untreated fines from the bulk box. With depth, there is an increase in the total amount of TL and the glow curves shift progressively to the left (lower temperatures). This systematic change is probably a result of the rapid decrease in mean temperature with depth below the lunar surface. Near the surface, where temperatures may exceed 100°C during the lunar day, only hightemperature TL remains. The daytime temperatures to which the material is subjected decrease rapidly with depth, resulting in slower decay rates and the preservation of more, and lower-temperature, TL. Because the glow curves in Fig. 2 shift continually to lower temperatures with depth, we conclude that the lunar diurnal temperature variations penetrate to a depth of at least 10.5 cm. It is also interesting to note that from 0 to 7.8 cm there is a steady increase in TL in the region of the glow curve above about 250°C, but that at 10.5 cm there appears to be a reversal in this trend. If real, this reversal may be caused by attenuation of solar radiation with depth.

It seems likely that the TL observed in the core samples is a result of an equilibrium between TL acquisition and TL loss, which suggests that TL disequilibrium may be useful in detecting disturbances in the lunar regolith. For example, if a surface was buried by a few centimeters of material such as might be ejected during the formation of a nearby crater, the normal progression of glow curves with depth would be disturbed. This disturbance would remain until equilibrium conditions were reestablished. The minimum length of time that such an anomalous glow curve pattern would remain detectable can be estimated from the time required to produce glow curves similar to those found in the core samples. Preliminary estimates, with decay neglected, suggest that a radiation dose of at least 10⁴ rad would be required. The radio-



Artificial TL (n lumen-sec/mg)

Fig. 3. Natural TL $(300^{\circ} \text{ to } 350^{\circ}\text{C})$ plotted against artificial TL $(85^{\circ} \text{ to } 350^{\circ}\text{C})$ for samples from breccia 10048: 1 = top; 2 = 4 mm below top; 3 =interior; 4 = bottom. The bars indicate estimates of the standard deviation of precision at the two-thirds confidence level.

active decay of U, Th, and K within the material (1) at present contributes about 0.3 rad/yr. The contribution from solar and cosmic radiation depends on depth and would be about 6 rad/yr at a depth of 10 cm (3). This suggests that a detectable TL anomaly at a depth of 10 cm would remain for more than 10^3 years after burial. Thus TL studies on core tube samples from the lunar regolith may be a means of detecting, and in the future perhaps of dating, recent surface activity that might otherwise go unnoticed.

The natural glow curve and the 5000-rad artificial glow curve were measured on material ground from the surfaces of chips from the top, interior, and bottom of breccia samples 10046 and 10048. The shapes of these curves indicate that the TL carrier in the breccias is identical to the TL carrier in the fines. There was no progressive change in the glow curves with depth, such as was found in the core samples. This is probably because of the lack of significant thermal gradients in the rocks. There is also no significant trend with depth of the ratios N/A except for the sample scraped from the uppermost surface of breccia 10048, which has a natural TL about twice that of the TL from the other three samples, as indicated by the N/Avalues (Fig. 3). This indicates a higher radiation dose, possibly from lowenergy solar radiation, which loses most of its energy within the upper millimeter or less. The TL measurement from the uppermost surface of breccia 10048 was lost because of instrument malfunction, and no significant differ-

SCIENCE, VOL. 167

ences were detected in the interior and bottom samples.

Studies of the crystalline rocks (10003 and 10044) are still in progress, but it is evident that the TL carrier (plagioclase) in these rocks is different from the TL carrier in the breccias and the fines. There is more TL, by several orders of magnitude, than there is in the fines; there is a natural peak above 400°C, with no evidence of a peak at 350°C; and the artificial glow curves are unlike those of the fines. There also appear to be differences between the glow curves of the two crystalline rocks, although these differences are not as pronounced as those between the crystalline rocks and the fines and breccias. These differences in TL characteristics are probably caused by dissimilarities either in composition or in the degree of shock metamorphism.

Finally, it is of interest to consider the amount of energy represented by TL in the lunar material. This has been estimated for the fines and breccias by taking the entire area under the glow curves (in lumen-sec), converting it to units of energy through use of the conversion factor for monochromatic green light at 5550 Å, and assuming that the light is emitted from the sample equally in all directions. The values are 8×10^{-9} cal/g for the fines and 2×10^{-9} cal/g for the breccias. Although only rough estimates, these data indicate that TL energy stored in the Apollo 11 samples is exceedingly small.

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References and Notes

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Luminescence and Thermoluminescence Induced by Bombardment with Protons of 159 Million Electron Volts

Abstract. We have observed a red luminescence, qualitatively similar to that of enstatite achondrites, in unsorted fines, their separated mineral phases, and rock chips. The energy efficiency of the plagioclase fraction is approximately 1 percent. At -196° C the effect is enhanced by a factor between 1 and 2. All fractions except ilmenite exhibit blue thermoluminescence with a glow peak near -135° C and an energy efficiency $\approx 4 \times 10^{-6}$. Unlike the thermoluminescence of terrestrial and meteoritic material, it is nonrepeatable even after annealing at 200° C. Similar thermoluminescence is seen in rock chips, but in unsorted fines it is masked by the opaque fractions.

There are two reasons for studying the luminescence of lunar rocks. First, the results may be used, alone or in combination with results from terrestrial and meteoritic rocks, as indicators of specific crystalline defects, hence giving information on the thermal and chemical history of the lunar surface.

Table 1. Luminescent and thermoluminescent parameters of lunar rocks.

| Material | Energy efficiency of direct luminescence | | Thermoluminescence | |
|----------------|--|-----------------------|----------------------|---|
| | 196°C | +100°C | Glow peak (°C) | Efficiency |
| Unsorted fines | 1.97×10^{-3} | 1.13×10^{-3} | | |
| Plagioclase | $1.07 	imes 10^{-2}$ | 6.66×10^{-3} | -134 | 4.32×10^{-6} |
| Pale pyroxene | $7.16 	imes 10^{-3}$ | 4.12×10^{-3} | -135 | 0.72×10^{-6} |
| Dark pyroxene | 3.97×10^{-3} | 2.80×10^{-3} | -135 | 4.09×10^{-6} |
| Ilmenite | $2.54 	imes 10^{-3}$ | 1.83×10^{-3} | | ,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,, |
| Surface chip | | | -132 | |
| Interior chip | Optical depth not known | | -145 | Optical depth not known |

30 JANUARY 1970

Second, the existence of a class of transient lunar phenomena has often been ascribed to luminescent effects, and studies of the energy efficiency of such processes, in conditions approximating those found on the moon, are clearly relevant to this interpretation.

Using techniques similar to those we have employed previously (1), we have measured the luminescence of samples from Apollo 11 when irradiated in the extracted proton beam of the synchrocyclotron at the Atomic Energy Research Establishment, Harwell. Our beam flux of about 4 \times 10⁸ protons cm⁻² sec⁻¹ gave rise to a local energy density of ionization of 2 \times 10¹⁵ ev g⁻¹ sec-1, which is insufficient to cause radiation damage during the measurements. This energy density is comparable with that resulting from the solar wind on the lunar surface and is a factor of 107 less than the value obtained with low-energy, electrostatically accelerated protons (2). Spectra were observed with a set of seven wide-band interference filters and an EMI 9558 photomultiplier tube. The specimens could be cooled to liquid nitrogen temperature and heated to 200°C. The phototube current and thermocouple voltage were recorded on a two-pen recorder.

The specimens studied were two unsorted aliquots of the < 1-mm fines, sample 84; four separated mineral phases from these fines; and two rock chips, a surface and an interior piece, from sample 17. The mineral separation was performed by washing and sieving, followed by magnetic separation of the fraction in the size range 40 to 150 μ m. Modal analysis of the four phases vielded the following compositions: (i) plagioclase 88 percent (with 10 percent colorless glass and 2 percent pyroxene composites); (ii) pale pyroxene 68 percent (with 17 percent plagioclase, 13 percent glass, and 2 percent ilmenite); (iii) dark pyroxene 66 percent (with 26 percent ilmenite composites, 4 percent plagioclase and 4 percent glass); (iv) ilmenite 80 percent (with 16 percent pyroxene composites, 2 percent plagioclase, and 2 percent glass). Fractions B and C each contain about 3 percent of unidentified olivine. Note that this size range contains only about 5 percent of glassy fragments and spherules.

The six powder specimens were mounted on aluminum disks using nonluminescent silicone grease. The amount of plagioclase and pale pyroxene was insufficient to completely cover the