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Activities of Triboluminescence at Sample Failure of Granite

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Abstract

Triboluminescence at a failure of Tsukuba granite was examined. The intensity of triboluminescence was determined by photo-counting at a time resolution of 100 microseconds. The pattern of TL activities of each sample is classified into a sequence of the precursory activity, the burst of triboluminescence at failure, and the following decaying after-glow and is extremely similar to the pattern of the foreshock-main-shock-aftershock sequences for natural eathquakes. The after-glow continues rather long time compared to the process-time of sample failure. The variation of the after-glow intensity as a function of time can be expressed by the same equation for aftershock activities of natural earthquakes;

$$I=\frac{C}{(t+D)^A},$$

where I is the intensity of triboluminescence at time t which is measured from a moment of failure and A, C, and D are constants. The constant A ranges from 0.5 to 0.82. The after-glow was shown to be sustained by consecutive generation of cracks in fragments without being subjected to any external force. Most of the samples showed tangilble precursory activities of triboluminescence which were observed up to twenty milliseconds prior to failure of samples. The initiation of these events closely relates to changes in the stress change rate. The intensity of triboluminescence is affected by humidity contained in a sample.

1. Introduction

Triboluminescence (TL) is light emitted from materials when they are fractured. The occurrence of TL has been known for many years and many triboluminescent substances have been found. A fairly comprehensive list of the triboluminescent substances was given (Wolff et al., 1952), in which only sixteen rock-forming minerals were included. In some textbooks on mineralogy, lists of the triboluminescent minerals are given (for example, Hurlbut, 1971). Comprehensive reviews for a wide variety of characteristics and sources of TL are given by Walton (1977) and Zink (1978).

A strong correlation between the TL activity and the deformation rate of single crystals was observed at a resolution of an order of second (Alzetta et al., 1970). A similar correlation between the rate of stress change and the TL activity was also observed for crystalline sucrose at a rather fast-time resolution (0.1 microsec.) and the beginning of the emission of light and that of a change of stress (meaning the beginning of sample failure) were shown to be simultaneous (Maeda, 1983). A similar correlation between the activity of acoustic emissions (AE) and the deformation rate is very familiar in rock mechanics (for example Mogi, 1962). A relation between TL and the dynamics of formation of cracks was studied on the basis of experiments with a somewhat fast-time resolution of 100 microseconds (Chandra and Zink, 1980). They stress that TL can be used to study fracture.

All these studies already reported were concerned in samples consisting of a single constituent. It is interesting to investigate the TL of multi-constituent materials represented by rocks. There has been no systematic study of the TL of rocks at sample failure with a sufficient time-resolution. In this report, the variation of TL intensity at and around sample failure of granite will be given.

2. Experiment

The experimental assembly is shown in Fig. 1. Thirty five samples of Tsukuba granite having a size of $4 \times 4 \times 8\ mm^3$ were examined. The experiment was performed under room temperature and humidity but several of the samples had been dried at a temperature of 45 or 75 degrees centigrade for at least five hours in order to examine the effect of humidity. The compressional load was measured by an oil pressure gauge. TL and load changes were recorded.

TL was guided through a quartz glass fiber cable and detected by two types of photomultipliers (model R 464 and R 943-02, Hamamatsu Photonics. Response ranges of spectral band are 300-650 nm and 160-930 nm, respectively). The photomultiplier R 464 shows rather low noise (5 counts/sec.) without cooling and has a high sensitivity (3×10^5 A/W: a ratio of the output current measured in ampere to the incident light energy in watt) but the second model, R 943-02, shows higher noise and lower sensitivity (3.4×10^4 A/W) than that of the first model, R 464. The rather high noise of the second model would not be a problem this time because, even if the maximum number of noise pulses reachs

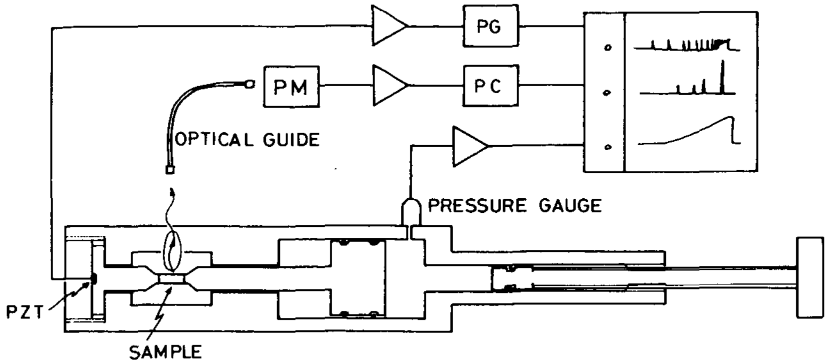


Fig. 1 Block diagram of the experiment. PM: photomultiplier. PC: photo-counter. PG: pulse generator. PZT: piezoelectric transducer. A couple of digital memory units is used as a convertor of the time scale so that the chart recorder can follow the data throughput which was originally too high.

ten thousand per second, the number of noise counted in each counting period of 100 microseconds is unit at most.

The maximum width of the output photo-electric pulses is less than 100 nsec. The output pulses were measured by the photo-counting method with a counting period of 100 microseconds (the time resolution is 100 microsec.). The minimum response time of the photo-counter is 100 nsec: If incident light is sufficiently strong (that is, if the number of out-put electric pulses of the photomultiplier exceeds 1000 per 100 microsec. = 10 MHz = 1/100 nsec.), the photo-counter could not correctly count the number of pulses coming because of overlap of consecutive pulses. As will be shown below, this actually happens at the early stage of the TL activities at sample failure. The results of this measurement, therefore, would havd only qualitative meanings. Although the overall sensitivity of the photo-sensing system was not calibrated rigorously, the approximate value was deduced to be the order of 10^{-8} erg. per unit count.

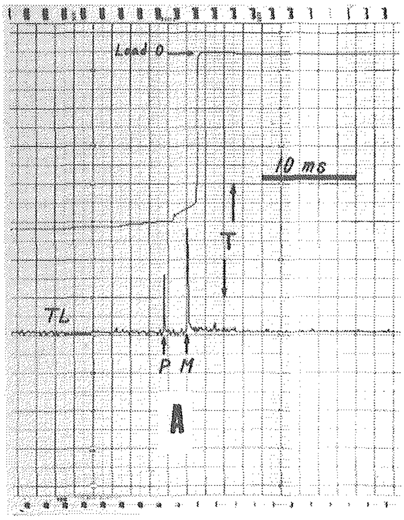
Because the output rate of the photo-counter is too high to be sent directly to the chart recorder, a digital memory unit with an analogue to digital and a digital to analogue converters was used as a temporary storage. Basic ideas for this system and the timing relation between an original signal, the one stored, and the out put signal of the memory have been given by Maeda (1979). The timing for sampling of the memory unit syncronized with that of the counting period. The word length of the memory is 8 bits (it can express only up to +127) but the possible maximum number of photo-electric pulses is 1,000 per 100 microseconds. In order to overcome this difficulty, the output of the photo-counter is once converted to analogue form and then again converted to digital form with suitable voltage adjustment sacrificing the least significant bit or the next; an original number N is stored as N/A, A determined by the voltage adjustment (for example if $N=500$ and $A=4$, 125 will be stored in the memory as a data). The loss of the least significant bit or the next is not important for the present experiment. It is clear from the argument on the number of noise pulses actually counted in one period that the counting period should not be set 10 microseconds in order to circumvent the voltage adjustment and sacrificing of accuracy. The memory is triggered by sudden drop of stress so only data for 0.1 second (corresponding to 1,000 words of the memory) around the moment of failure can be stored.

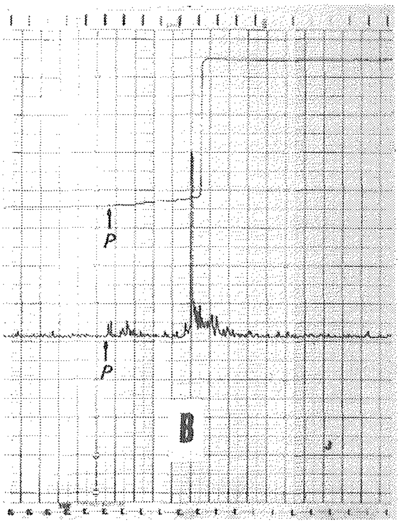
Stress was also measured with this time-resolution. Because the response time of the pressure gauge is less than 10 microseconds and the cut-off frequency of the amplifier is 10 KHz, the measurement of stress with a sampling period of 100 microseconds is meaningful. The function of the memory unit used for the stress data is the same as that for TL and the timing of sampling syncronizes with that of the photo-counter. The loading rate was controlled manually but the stress change around the moments of sample failure was completely out of control, in the period in which data are stored. It means that observed changes in load reflect the actual fracture process of samples. After completion of failure the stored data of TL and stress are read out at a sufficiently low rate so that the recorder can follow.

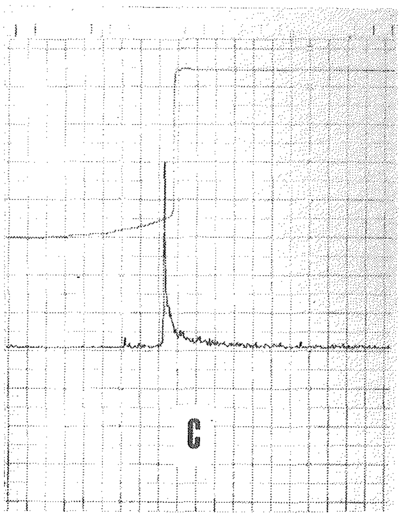
3. Results

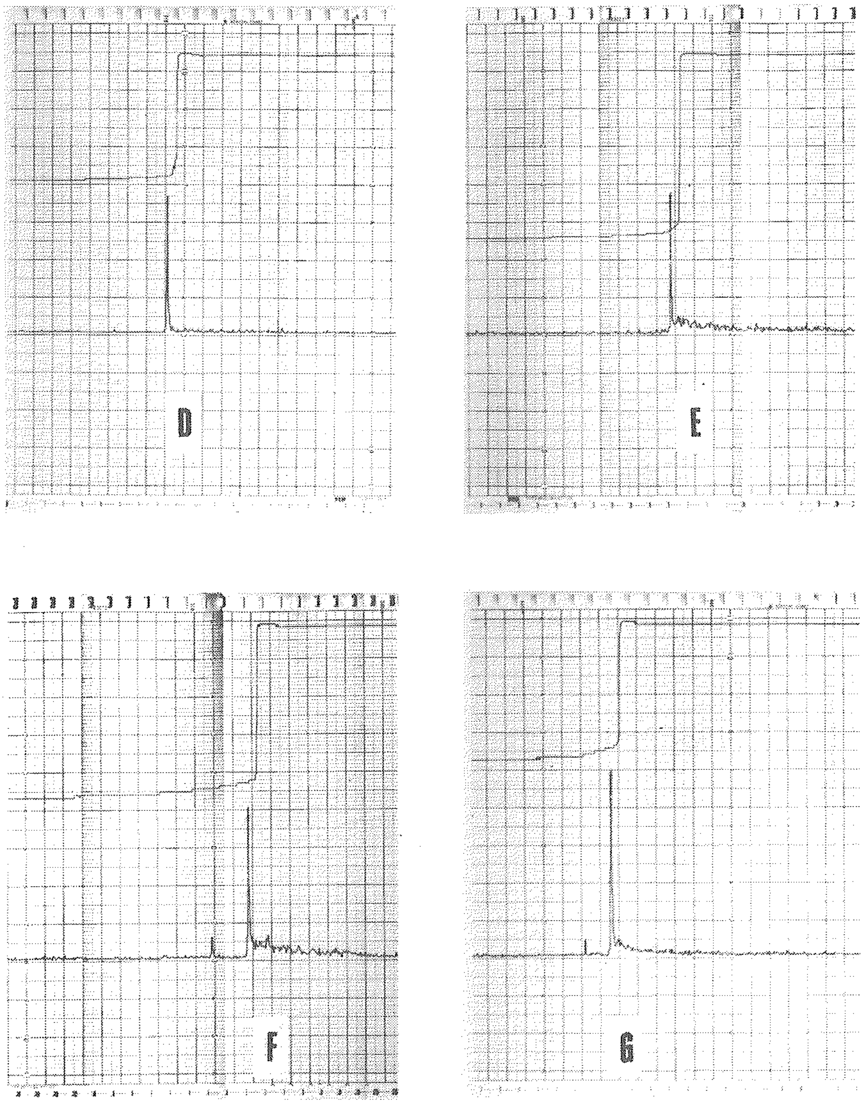
Records are shown in Fig. 2. We will explain Fig. 2-A as an example and the same scale is applied to the remaining ones. The upper trace is a load variation (the upper most line is the zero load as indicated) and lower one is that of TL intensity. The relative pen positions meaning the same mement are indicated by up- and down- arrows with letter T. Then one can see the TL activity indicated by M (Main event) corresponds to the sudden drop of load to zero. TL activities prior to the moment should be called precursory activities the beginning of which is indicated by P (see also Fig. 2-B). A time scale of 10 miliseconds is indicated by a horizontal bold line. The maximum load of this case corresponds approximately to a stress of 2.4 Kb on the sample. Small steps in the load trace are due to the digital sampling without sufficient word length. Records 2-A to D were obtained by the photomultipier R 464 and the remainings were by R 943-02.

Fig. 2 Examples of records. All have the same time scale which is indicated in A. The vertical scale for TL is as follows ; the counted number of photoelectric pulses of 250 has the same length as that of the time scale bar of 10 miliseconds except records M and N. In each record the upper trace is load variation and the lower one is TL activity around moments of failure. The upward and downward arrows marked by T indicate relative pen positions. Arrows marked by P mean precursory activities of TL. The arrow marked by M shows the moment of the final failure of a sample and the onset of TL burst corresponding to it.





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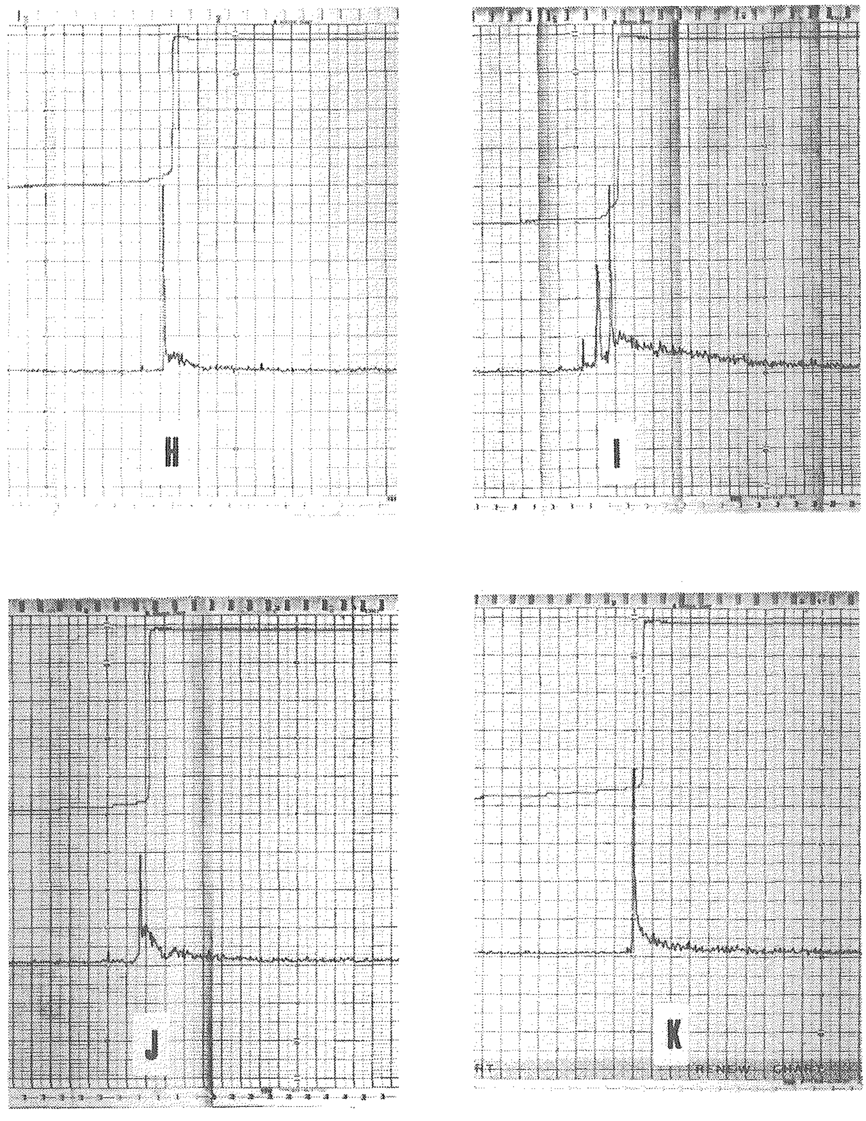
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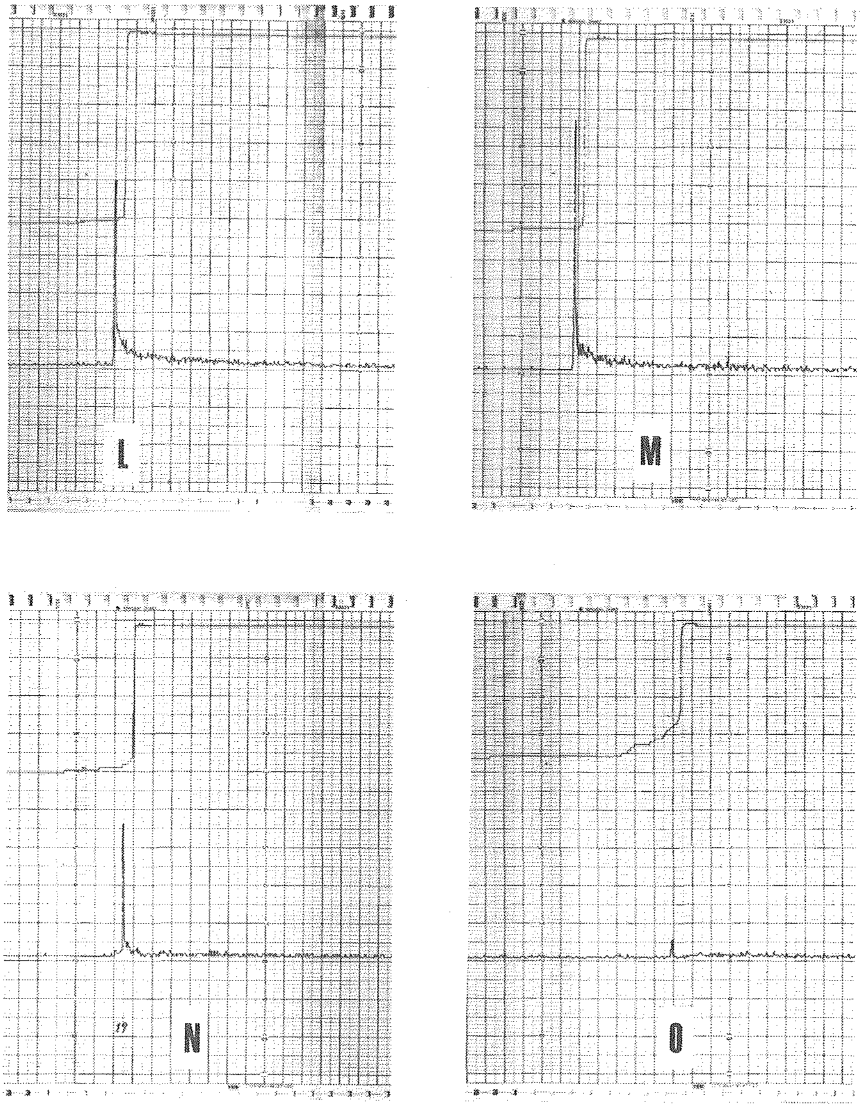
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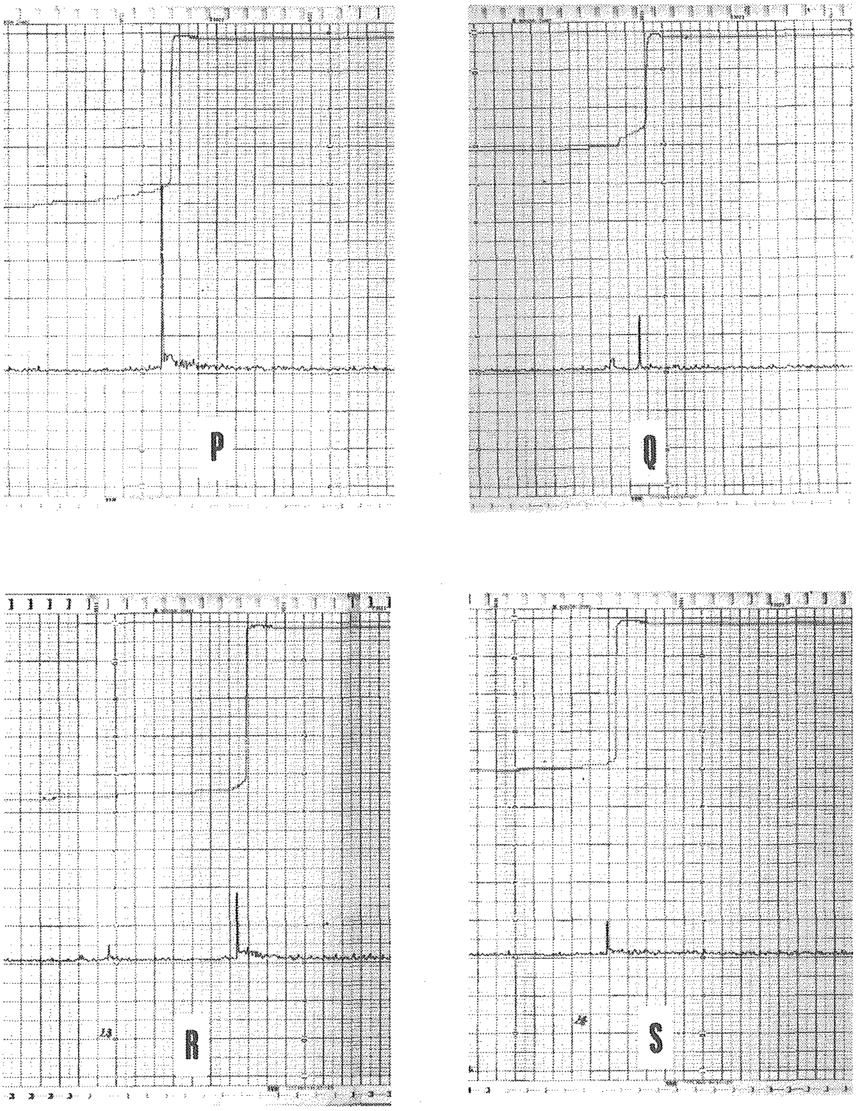
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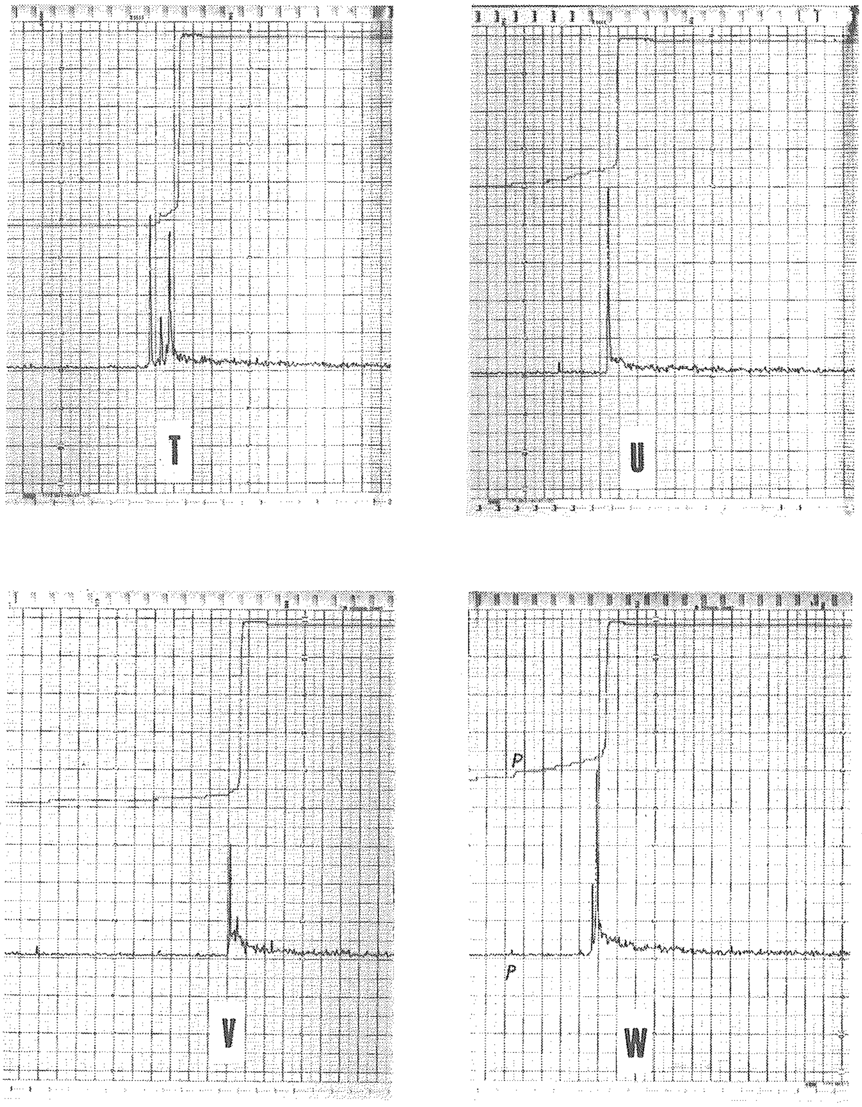
T
U
V
P
P
W

We first choose records B, I, K, and W as typical examples. The record K shows almost sudden failure of the sample without discernible precursory reduction of stress and a burst of TL activity accompanying the failure with almost indiscernible precursory activity. The TL activity is decaying but continues for long time. In the record W, precursory reduction of stress the beginning of which is indicated by a letter P is clearly seen (We call this length of time span a long term in the extent of the present time scale) and slight TL activity accompanied by it can also be seen though much more short term precursory of TL activity is conspicuous. The record B shows more conspicuous long term precursors in stress and TL activity the beginning of which is again indicated by a letter P. The record I shows rather high TL activity before and after the main failure. This unusual high activity is thought to be caused by a large feldspar grain accidentally contained in the sample.

Most of the remaining records are intermediate between the records B and K and a few of the remaining are similar to the record I. The earliest precursory TL activities could be traced back up to 20 miliseconds prior to the main event. Some of them show low intensity TL activities, reasons of which will be discussed below. The patterns of the activity of TL resemble incredibly those of earthquakes associated with an large earthquake, the classification of which was given by Utsu (1970). According to Utsu's classification, the activities K, W, B, I of TL can be assigned to the main shock-aftershock (MA for short) type A1, the foreshock-main-shock-aftershock (FMA for short) type $B_0 2$, the FMA type $B_2 1$, and C1, respectively. This strong resemblance would not be ostensible because both TL and earthquakes are excited by generating cracks. This observation will be supported by somewhat more quantitative analysis of the decay rate of TL activity.

In Fig. 3, the log intensities of TL of the records K and W in Fig. 2 are plotted against log time. The plotted points after some time elapse are the average values among data around each moment of time plotted. Many of the first data were off scale. A strong linear trend can be recognized and will be expressed in the following form:

$$\log(I) = -A.\log(t) + B$$

or

$$I = \frac{C}{t^A}, \qquad (1)$$

where I is the intensity of TL at time t and A, B, and C are coefficients. The coefficient A ranges from 0.5 to 0.82 and B or C varies from sample to sample.

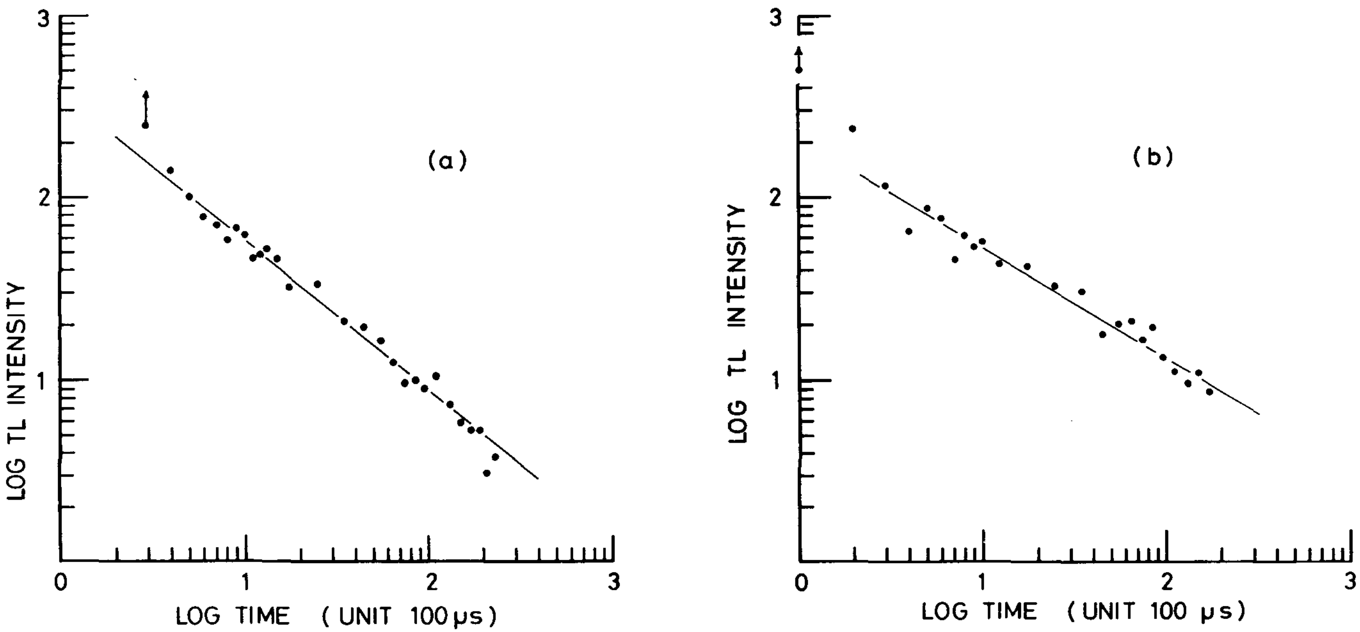

Fig. 3 Plots of log TL intensities against log time. Data attached to an arrow and data obtained before that were off scale. Plots (a) and (b) correspond to records K and W in Fig. 2, respectively. The unit of TL intensity is the number of photoelectric pulses counted for each period of 100 microsecond.

It seems that the intensity of TL activity (indicated by B or C if A is constant) was affected by humidity of air contained in the samples (see below). Such a tendency, however, could not be discerned for the coefficient A. In the case of earthquakes, the corresponding quantity of the intensity of TL might be the energy of earthquakes occurring in a unit of time. Utsu (1961) studied the variation of energy of aftershocks and showed a relation similar to Eq. (1):

$$E=\frac{C}{(t+D)^{A}}, \tag{2}$$

where E is the energy and D is an additional coefficient which does not affect the overall linearity but means that there is a deviation from the linearity at the opening activity of aftershocks. This activity deviates from a line indicating the linearity to lower-side of it if we see the variation reversely in time axis (Utsu, 1961). As can be seen in Fig. 3, the activity of TL also devates from the linearity at the beginning though in opposite direction. This means that both TL and earthquake activies can be expressed by the same equation (2) but tendencies of the deviation, that is, upward or downward, are different between them. For earthquakes, the coefficient A is about two (Utsu, 1961). The activity of aftershocks can also be expressed as a rate of aftershock occurrence and the rate is also expressed in the form of Eq. (2) with N (number of earthquakes occurred in a unit of time) in place of E though A is about unit (Utsu, 1961).

Scholz (1968) reported that the activity of AE after the failure of samples can be expressed in the first form of Eq. (1) with N in place of I though the parameter A varied sample to sample. Generally, AE is detected by transducers such as the piezoelectric transducer attached on a sample or end pieses and therefore detection of AE after breakdowns of samples by uniaxial stress is not assured because of loosing the contact (or the elastic impedance matching). Scholz used a sufficiently stiff machine to test samples in order that the samples were not scattered. Even so, states of the contact would vary experiment to experiment and also would vary during measurement for a sample. This suggests that the apparent decay rate of AE activity would be different from experiment to experiment. It can be considered that such a fluctuation in the resulting parameter A is a drawback of the measurements made by using contacting transducers when compared to measurements made by using non-contacting sensors detecting TL as an information source for fracture, in which rather stable values of A were obtained.

Meyer et al. (1970) gave TL decay curves of ZnS doped with several metal elements with different concentration and found that ZnS : Mn shows exponential decay. Unfortunately, because their experimental way (compaction of thin powdery sample by plates) is completely different from the present one we cannot compare their results with present ones directly.

A correlation between initiations of precursory TL activities and those of load changes is clear. For example, record A shows two spike-like TL activities which syncronized with initial moments of two stages in load change. The reason why the precursory TL activity did not continue to the main even though the load kept decreasing is explained as follow : In average, the rupture velocity in rocks is an order of the Rayleigh wave velocity, in the present samples, 3 km/sec. This means a rupture can run through a sample of the present size in less than 10 microseconds (process time). After the rupture expansion has been completed, then frictional sliding which continues for a much longer time than the process time would take place. If TL is emitted most effectively during the rupture expansion, the spike-like TL activites must syncronize with a moment of sudden change in load change rate. Many records with precursory spike-like TL activities (A, B, C, H, Q, T, and U) support this assumption. We made a subsidiary Brazilian test using single crystals to know the process time associated with a single crack expansion and found that it is sufficiently short compared with the present time resolution. These observations and arguments suggest that TL does not come from heating by the frictional sliding. Inversely speaking, the number of TL spikes means the number of large scale (relative to the sample size) ruptures occurring in a sample.

We now make some explanations on the postponed problem of low intensity TL. The case of O may be attributed to the rather gradual load drop to zero. Such a gradual advance of fracture may choose weeker paths avoiding hard mineral grains. If it were true, TL intensity would be low because a possibility of cracking qualtz which is hard and an effective TL mineral in granite is reduced. Cases of Q, R, and S are due to humidity. Although humidities were not measured, taking account of an appearance of source of water vapor, we could concluded that the humitity had increased. By heating samples up to 45 and 75 degree centigrade, all samples showed the normal intensity (records T to W) even in the same damp atmosphere. This proves the cause of lowering TL intensity and suggests that humidity inside of samples is important.

Relating to the problem of TL intensity, we have to point out the fact that there is no clear intensity difference between data obtained by different photomultipliers. It has been thought to be advantageous to use a model having a wider response range reaching to infrared if the main TL energy is emitted in the form of black body radiation. The above observation shows that the lower sensitivity of R 943-02 is compensated by its wider response range. This meets with what expected from the chracteristics of the black body radiation, because the wavelength of the peak spectral energy of the radiation is in infrared region for the source temperatures obtained by Weichert and Schronert (1978). Reduction in TL intensity by water vapor consists with the theory by Weichert and Scronert because, at a crack tip, atomic bonds are weakend by water molecules (Lawn and Wilshaw, 1975) and weakened bonds cannot sustain high stress which causes high energy concentration resulting in black body radiation.

We now describe in what state samples or their fragments keep emitting light. After a complete breakdown of each sample, the sample or its fragments fly in the air for two or three miliseconds then bump against the walls which confine the fragments from flying out of the field of the sensing device. This was confirmed by removing the walls (corresponding records are Fig. 's 2-A to H). If there is no wall, the observed intensity of TL shows a large irregular

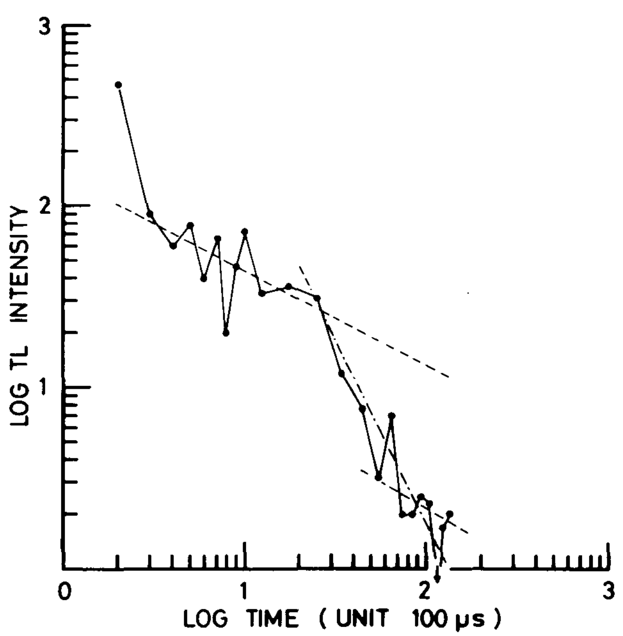

Fig. 4 Plot of log TL intensity against log time similar to Fig. 3, the original record of which is B in Fig. 2. It can be clearly seen that the deflection from the earlier trend of decay occurred at around 3 milliseconds after failure. This deflection was due to the fact that fragments could fly out of the field of the sensing device in this case.

variation and suddenly drops a few miliseconds after failure. Fig. 4 (original record is Fig. 2-B) shows an example of this. In order to see this easily, data points are connected by lines in time order. A rough estimate shows the velocity of scattered particles to be the order of several meters per second. This estimation agrees with observations that scattered fragments were distributed within a 1 m radius when there was no barrier. These observations show that, during the whole span of time after failure, rocks emit TL without being subjected to any external force. Together with the above argument on the spike-like precursory activities, small scale fluctuations in the after-glow curves indicate that the after-glow is caused by consecutive generation of cracks and not by frictional heating at sample failure.

4. Conclusions

TL from granite at sample failure has been studied with a time resolution of 100 microseconds. Overall TL intensity variation (or TL activity) at and around sample failure is quite similar to the pattern of earthquake activity accompanying a large earthquake. All sample show TL after their complete failure. TL intensity during the after-glow decays with time linearly in log-log space and the after glow is not caused by frictional heating at the ultimate break down of a sample but sustained by consecutive generation of cracks without being subjected any external load. Many samples show precursory activities of TL several to 20 miliseconds prior to the main or ultimate failure of specimens. The precursory activities are spike-like and each spike is simultaneous with a moment of change in stress change rate. TL intensity is affected by the humidity contained in a sample. All suggest that TL is the black body radiation caused by the energy concentration at crack tips and not by frictional heating in a usual sense.

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