APPENDIX

Reprints of a few Published Papers
Thermoluminescence of Quartz: Part 1—Glow Curve & Spectral characteristics

Virgin (natural) quartz samples exhibit glow peaks at about 250, 315 and 350°C. On irradiation with γ-rays, the samples showed extra peaks at about 71, 110 and 169°C. The observed glow emission consists of two bands with maxima for most of the samples at 350 and 470 nm.

Thermoluminescence (TL) of quartz has gained enhanced importance within the last two decades because of its application in diverse fields like, archaeology, geology and dosimetry of nuclear radiations. It is, therefore, important to have a knowledge of TL characteristics of as many varieties of quartz as possible and to examine their similarities and dissimilarities. Published literature (by different workers) on different specimens cannot be compared with each other, since dissimilarities of glow curve shapes and peak temperatures can arise because of the difference in experimental conditions (of individual workers), such as heating rate during TL recording, pre-irradiation treatment, etc. Further, studies of the spectral nature of TL emission are inadequate and scanty. Keeping the above points in view, samples of different types are collected from various places and their TL glow curves and TL emission spectra are recorded and the results are discussed in this note.

Natural specimens were crushed to powder, and the fine powder sieved through a mesh of 100 'Tyler' size was used in all our experiments. The apparatus to record TL glow curves and TL emission spectra has been described in detail elsewhere. Care was taken to avoid the tribo-thermoluminescence by reading the sample with nitrogen flow through the sample reader chamber.

The TL glow curves of virgin specimens show peaks (Fig. 1), at temperatures approx. 250, 315 and 350°C. On gamma irradiation (1.25 x 10⁵R) after heating at 400°C to remove natural TL, glow peaks are observed in most of the samples at average temperatures of 71, 110, 169, 235, 310 and 350°C as shown in Fig. 2. Some samples have glow peaks at 425 and 500°C also. The absence of glow peaks at temperatures lower than 250°C in virgin samples is due to fading at ambient earth surface temperature.

As seen from Fig. 1, the intensities of individual glow peaks as well as the overall TL intensities are widely different from sample to sample. These variations can be attributed to the following factors: (1) Natural thermoluminescence (NTL) is induced by natural radioactivity present in the specimen and its environment which may vary from sample to sample and place to place. (It is evident from Fig. 1 that specimens from uranium or thorium bearing rocks and sands, have, in general, one order higher TL intensity than others). (2) The intensity of NTL peak temperatures can differ from one specimen to other due to changes in impurity contents and lattice defects.

In contrast to earlier works, the glow peak, which appears most consistently and prominently in all the
irradiated specimens without exception, is the one that would depend upon the geological age of the specimen, since the longer the age, the larger will be the time integrated radiation dose to the sample, which is responsible for inducing TL. (3) The TL characteristics like radiation sensitivity as well as glow occurs at an average temperature of 71°C (Fig. 2). The intensity of a given glow peak for a given radiation exposure varies from sample to sample by as much as three orders of magnitude. The relative intensities of different glow peaks of the same sample also change with exposure (received by the sample). The growth pattern as well as the saturation levels are different for different peaks (Fig. 3). The intensity of the 66°C glow peak of pink quartz, for example, rises linearly with exposure till it reaches a maximum at $3 \times 10^2$ R. The peaks of 175 and 310°C grow supralinearly and continue to rise up to an exposure of $1 \times 10^5$ R and $3 \times 10^9$ R respectively. The peak at 175°C starts rising sharply after the peak at 66°C attains saturation. Similarly, the peak at 310°C continues to rise even after the peak at 175°C has saturated. The highest intensities attained by peaks at 175°C and 310°C are respectively 7 and 17 times larger than that of the peak at 66°C even though at smaller doses the peak at 66°C is the strongest one.

The spectra of the glow peaks (Fig 4) show two major regions of emission at about 350 and 470 nm. Some samples have emissions at 425 instead of at 470 nm. The band at 470 nm is stronger in intensity than that at 350 nm except in two of the samples (Fuchsite and Pink variety). The 470 nm band in smoky quartz has been attributed to aluminium substitutional impurity by some workers, while McMorres has attributed it to Ge-alkali centres. The emission band at 350 nm has been attributed by Medlin to unidentified lattice defects. According to him, the impurities play a secondary role in TL of this material. Lattice defects and impurities undoubtedly play the most important role in TL, though precise knowledge of these entities and their function in respect of individual glow peaks is not available. A particular impurity may not be responsible to determine the temperature of appearance of glow peak but it might determine the spectrum of the emitted glow. Large differences in sensitivity to irradiation in specimens having glow peaks at identical temperatures, could be attributed to the change in the concentration of some impurity ions.

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5 Medlin W L, J. chem Phys., 38 (1968), 1132.
Fig. 4—Emission spectra of different varieties of natural quartz samples. (Q-numbers in the figure are the identification numbers of the specimens)


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Received 19 June 1976, revised received 27 Sept. 1976
Thermoluminescence of Quartz: Part II—Sensitization by Thermal Treatment

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Received 19 June 1976

Thermoluminescence (TL) sensitivity of quartz is found to be enhanced by a factor of 20 when it is heated to temperatures exceeding 600°C and then quenched to 0°C. Slow cooling, on the other hand, reduces the TL sensitivity below that of the sample prior to heat treatment. The maxima in the curve of TL sensitivity as a function of temperature of heating, coincide approximately with the temperatures of phase change in quartz from alpha to beta and from beta to tridymite form. The results are explained on the basis of generation of vacancies in the crystal lattice by thermal treatment at elevated temperatures.

Studies by the authors on thermoluminescence (TL) glow curves and their spectra for a number of natural quartz specimens have been presented earlier. Most of the naturally available quartz is in the alpha form. On heating it changes to beta (at 573°C) and to tridymite (at 870°C). Ichikawa has found that TL sensitivity of quartz increases on heating. The present communication gives details of a systematic study (by us) of the effect of heating on TL and its relation with phase change.

A sample of natural pink quartz was selected for these studies since it is easily available and has sufficient TL sensitivity. The same experimental procedure as described in an earlier paper was followed for recording TL glow curves. The specimen was crushed for TL reading, and the powder between 100 and 200 Tylor mesh was used. Test exposures were given in ice bath to avoid decay of 70°C glow peak which has a half-life of 36 min at room temperature.

Three quartz capsules of 3 cm length, 0.5 cm internal diameter and 1 mm thickness were packed with the specimen powder. These were then kept in a muffle furnace at 600°C for 90 min. The first capsule was dropped from the furnace into an iced water bath, the second was taken out in a silica crucible and allowed to cool in air and the third capsule was left in the furnace and allowed to cool in it by switching off the furnace. A test exposure of $4 \times 10^4$ R was given to each in ice bath and TL was read from room temperature to 500°C. Fig. 1 presents the glow curves along with the glow curve of virgin sample exposed to the same test dose. The sample quenched in ice water bath produced maximum TL (Fig. 1). A number of 50 mg batches of powdered specimens were heated in silica crucibles for 90 min at different temperatures varying from 400°C to 900°C in a muffle furnace at intervals of 50°C. These were quenched to room temperature by withdrawing the crucibles from the furnace into open air. A test exposure of $10^5$ R was given and the TL was read. The results are shown in Fig. 2. For comparison the TL of untreated sample is also included in Fig. 2. In Fig. 3 is shown the effect of time of heating. The intensity values plotted in Figs. 2 and 3 are integrated intensities of full glow curves. Fig. 4 shows the integrated TL intensity as a function of exposure for the four types of samples namely the virgin untreated, 600°C heated slowly cooled, 600°C heated air quenched and 600°C heated ice quenched.

The TL spectra of glow peaks were recorded using Jarrell-Ash 0.25 m Ebert monochromator with scanning speed of 100 nm/min while the samples were maintained at fixed temperatures of about 30°C lower than those of respective glow peaks. Fig. 5 shows the spectra taken at 40° and 140°C which correspond to the emission of 70°C and 175°C glow peaks respectively.

![Fig. 1—Curves showing the effect of different thermal treatments on TL sensitivity. 1. Natural sample; 2. Sample heated at 600°C for 90 min. and quenched to 0°C; 3. Same as in 2 but quenched to room temperature; and 4. Slowly cooled in the furnace to room temperature.](image-url)
Fig. 5—Emission spectra of a pink quartz sample heated at 600°C for 90 min. and cooled in different ways [Spectra taken at: - - - - 40°C; ---- 140°C. Test exposure: 2.55 X 10^4 R(y) in ice bath]

temperature ranges before and after about 600°C shows that the rate of generation of the vacancies (with respect to rising temperature) leading to the formation of trapping sites, is different in the different phases of quartz crystal. These phase changes in quartz occur sharply at the given temperatures, whereas the vacancies and the related defects are formed by the reversible thermodynamic process varying comparatively slowly with the temperature. This makes the discontinuities (Fig. 2) at 600 and 850°C clearly noticeable.

In Fig. 4, a sharp rise is observed in TL intensity with exposure after about 4 X 10^4 R. This effect is due to further sensitization by high exposure. The high-irradiation caused sensitization superimposes over the thermal sensitization effect in this exposure range. Ultimately a limit is reached and the TL output saturates. The radiation sensitization effect which is by far overwhelming in slowly cooled sample in comparison to quenched or even virgin natural samples, brings the saturation level (intensity) equal to the limiting level in all the samples. Enhancement in TL sensitivity due to high radiation exposure is discussed in a separate paper.

References
Thermoluminescence of Quartz—Part III: Sensitization by Pre-Gamma Exposure

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Received 24 June 1977; revised received 31 October 1977

Thermoluminescence (TL) response of quartz to gamma radiations is enhanced by two to three orders of magnitude if the sample has received a pre-gamma exposure of about $10^7$ R. A strong glow peak develops at about 250°C when the pre-exposure exceeds $10^4$ R. The spectrum of this newly generated peak is different from that of the glow peaks produced without pre-irradiation. Prolonged heating of the pre-irradiated sample at 700-900°C reduces the sensitivity of this peak and causes an enhancement in the other three glow peaks appearing below 200°C. It is tentatively proposed that high irradiation causes the formation of a defect near an impurity and that the centre thus formed gives rise to a glow peak with the spectrum of the impurity. Subsequent heating at 700-900°C transforms these centres into traps of lower temperature peaks.

1. Introduction

Results of a study on thermoluminescence (TL) glow curves and emission spectra of a large variety of quartz specimens have been reported in an earlier paper.1 The TL sensitivity of quartz has been observed to be enhanced by large factors if the sample is subjected to thermal quenching.2 The present paper deals with the sensitization of TL response of quartz by pre-exposure to gamma radiation. Sensitization produced by pre-gamma doses in LiF (Ref. 3), CaF$_2$ (Ref. 4), CaO$_2$ (Ref. 5) and Al$_2$O$_3$ (Ref. 6) has been studied by different workers. Fleming2 observed the sensitization of 110°C glow peak of quartz by pre-irradiation followed by thermal treatment at 500°C or more. Ichikawa8 also observed the enhancement of TL response after high irradiation of quartz. In the present paper it is found that the sensitization caused by pre-gamma exposure is in fact due to the development of a strong glow peak at about 250°C.

2. Experimental Results

The results presented in this paper pertain to a sample of pink quartz, although similar results are observed in other quartz samples of different colours and origin. Experimental set-up for recording the TL glow curves and TL emission spectra are described elsewhere.4,6

Samples were irradiated at room temperature to different gamma exposures ranging from $10^5$ to $10^7$ R. These were then heated at 400°C for 90 min, to remove the induced TL. A test gamma exposure of $4.5 \times 10^4$ R was then given and the glow curves recorded. These are shown in Fig. 1.

An experiment was carried out to find whether the pre-gamma induced sensitization could be removed by thermal treatments. Sensitized sample (pre-gamma exposure: $10^7$R) was annealed at 400°C for 90 min and was divided into six parts. Two of these were heated at 700°C, the other two at 800°C and the last two at 900°C. One from each set was quenched from the respective temperatures to room temperature while the other was allowed to cool to room temperature very slowly by switching off the furnace. After a standard heat treatment at 400°C, all the six samples were irradiated to a test gamma exposure of $3.75 \times 10^4$R and glow curves were recorded (Fig. 2).

Emission spectra of different glow peaks were recorded by maintaining the samples at fixed temperatures. The selected temperatures were about 30°C lower than the respective glow peak temperatures. The spectra are shown in Figs. 3(a) and 3(b) for the virgin and the sensitized samples respectively after a test exposure of $10^4$R.

These observations can be summarized as follows:

- There is a rapid increase in the gamma ray sensitivity of the TL glow peaks appearing above 200°C with increasing pre-exposure received by the sample. A new glow peak appears with strong intensity at about 250°C (Fig. 1). A plot of the height of this peak (produced by a fixed test exposure as a function of the pre-exposure values) shows that when pre-exposure increases beyond $10^4$ R, there is a steep rise in the TL output (Fig. 4). On subsequent thermal treatment at 700-900°C, this glow peak loses its sensitivity while the other three peaks appearing below 200°C are enhanced (Figs. 2 and 5).
heating of pre-irradiated quartz at 700-900°C it is found that TL sensitivity increases in the glow peaks appearing below 200°C and decreases in those appearing above 200°C (Figs. 2 and 5). This behaviour is different from thermal sensitization of quartz without pre-exposure, which is nearly uniform in all the glow peaks and is observed only on quenching. In the present case, same effect is seen whether the sample is quenched or slowly cooled.

Sensitization of 110°C TL glow peak of quartz by heat treatment subsequent to pre-irradiation has been shown earlier by Fleming7 and Fleming and Thomson12 Fleming7 explained the sensitization by assuming that the luminescence centres get charged by heating the pre-exposed sample at 500°C. The heat treatment at 500°C is presumed to-transfer charges (holes) from certain deep lying 'killer' centres (which are filled up during the pre-exposure) to the luminescence centres. Although Fleming’s results deal with small exposures (< 500 R) when development of the glow peak at 250°C cannot be seen, the present results are similar in the sense that the glow peaks at temperatures below 200°C get sensitized after thermal treatment subsequent to pre-exposure. The increase in intensity of the glow peaks appearing at temperatures below 200°C on one hand and the decrease above 200°C on the other suggests that the TL centres might be getting transformed from the latter type to the former during thermal treatment.

5. Conclusion
Pre-irradiation induced changes in quartz are very dissimilar to those in other TL phosphors. High pre-exposure in the range of 10^6-10^9 R, produces a strong glow peak at a temperature of about 250°C. Prolonged heating at 700-900°C subsequent to pre-exposure produces increase in TL sensitivity of glow peaks.
References

GEOCHRONOLOGY AND PROSPECTING OF RADIOACTIVE ORES BY THEIR THERMOLUMINESCENCE

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ABSTRACT
Preliminary studies have been conducted to investigate the possibility of applying thermoluminescence (TL) techniques in radioactive mineral prospecting and geological dating. TL measurements were done on six uranium ore samples collected from various localities all over India and seven thorium bearing monazite samples from the length of the Indian west coast. The TL age estimates obtained have been very low and found to bear an inverse proportionality with the respective alpha activities of the samples; this is presumably due to a continuous production of alpha-irradiation-induced TL traps through the geological times thus rendering the present day TL calibrations incorrect. On the other hand, encouraging results have been obtained regarding prospecting of monazite deposits from their natural TL emissions and this may serve as a convenient supplementary method to the conventional radiometric survey for the radioactive mineral prospecting.

INTRODUCTION
Thermoluminescence (TL) has been customarily thought to be a promising tool in geological applications such as age estimation, mineral prospecting, stratigraphy, paleothermometry etc.; however only limited success has so far been reported in isolated cases and efforts are still continuing in search of new areas where the technique could be fruitfully employed. This paper presents the preliminary results obtained in a similar attempt on some uranium and thorium bearing ore deposits in India.

SAMPLES AND TECHNIQUES
The investigations have been carried out on six uranium ore samples collected from deposits scattered all over India and seven thorium bearing sand samples from monazite deposits along its west coast (Table 1). TL glow curves have been recorded from virgin samples (−80 and +200 tyler) as well as after artificial 60Co gamma irradiation. These will be respectively referred as natural and artificial TL-NTL & ATL-glow curves. Alpha irradiation of these samples (a plated 241Am source was used on samples powdered to −325 tyler) produced less than 1% TL outputs as compared to gamma irradiation on equal absorbed dose basis; hence alpha induced TL is neglected in the ultimate analysis of results. TL efficiency for beta irradiations is the same as for gammas.

Where possible, quartz grains were separated from the ore samples and these 'quartz extracts' were then examined...
TABLE 1
LIST OF U AND Th ORE SAMPLES STUDIED FOR THEIR TL

<table>
<thead>
<tr>
<th>Sample code</th>
<th>Locality</th>
<th>Rock type</th>
<th>Radioactivity level (gross alpha counts per hour)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A. Uranium Ore Samples:</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>MI-1</td>
<td>Morni, Ambala Dt., Haryana</td>
<td>Indurated sandstone</td>
<td>24,827</td>
</tr>
<tr>
<td>MH-20(a)</td>
<td>Umra, Udaipur Dt., Rajasthan</td>
<td>Schistose shales</td>
<td>17.825</td>
</tr>
<tr>
<td>MH-27</td>
<td>Turamdih, Singhbhum, Dt., Bihar</td>
<td>Schist</td>
<td>1,284</td>
</tr>
<tr>
<td>MH-31</td>
<td>Sevattur, Dharmapuri, Dt., Tamilnadu</td>
<td>Carbonatite</td>
<td>247</td>
</tr>
<tr>
<td>MH-32Q</td>
<td>Kalaspura, Chikamangalur, Dt., Karnataka (Quartz extract)</td>
<td>Conglomerate</td>
<td>2,305</td>
</tr>
<tr>
<td>MH-34</td>
<td>Nerwpahar, Singhbhum, Dt., Bihar</td>
<td>Uraniferous ore</td>
<td>5,212</td>
</tr>
<tr>
<td>B. Thorium Ore Samples</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>00</td>
<td>Purakkadu, Kerala</td>
<td>Beach sand</td>
<td>98</td>
</tr>
<tr>
<td>01</td>
<td>Trikkunnapuzha, Kerala</td>
<td>Beach sand</td>
<td>87</td>
</tr>
<tr>
<td>02</td>
<td>Arrattupuzha, Kerala</td>
<td>Beach sand</td>
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<td>Beach sand</td>
<td>2480</td>
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<td>Beach sand</td>
<td>2840</td>
</tr>
<tr>
<td>05</td>
<td>Chavara, Kerala</td>
<td>Beach sand</td>
<td>6840</td>
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<tr>
<td>06</td>
<td>Thskkumbhagum, Kerala</td>
<td>Beach sand</td>
<td>8264</td>
</tr>
</tbody>
</table>

is one of the more common TL sensitive minerals generally encountered in geological studies. However in the present collections, only the monazite sand samples yielded appreciable quantities of quartz and the prospecting aspect of our investigations was thus restricted to monazite deposits only.

The build up of the TL peak corresponding to the NTL of each sample was followed by studying the TL outputs for incremental artificial gamma irradiations of the virgin samples and the NTL output is calibrated in terms of an equivalent gamma dose. Details of the TL instrument and experimental procedures used are the same as described elsewhere (Nambi et al. 1978).

The gross alpha activity of each sample (as given in Table 1) was determined by counting an ‘infinitely thick’ sample in contact with a ZnS scintillator screen and a photomultiplier (Turner et al. 1956). The sample area in our case was 9.83 cm² and the counting efficiency, 87%. The alpha, beta and gamma annual dose rates due to the U and Th contents are estimated from the gross alpha activity using the conversion factors suggested by Bell (1977). These dose rate values are used assuming each sample to be surrounded by its own bulk to an equilibrium thickness of about 30 cm on all sides in their respective deposits.

The NTL exposure could be assumed to arise mainly from the beta and...
gamma emanations of the uranium and thorium series neglecting the alpha component which produces negligible TL. (Alphas do produce defects and not necessarily filled traps and this aspect is discussed later). The cosmic ray contributions to the NTL dose have also been omitted for lack of data on background radiation levels at the respective sampling locations and this is expected to result in an over estimation of the TL age within a factor. The ratio of the NTL dose in equivalent gamma dose to the beta, gamma annual dose rate within each sample could thus yield an upper limit to the TL age of the sample through which it has been subjected to this self irradiation.

RESULTS

Geochronology

a) TL age of uranium ore samples

Figure 1 presents the NTL and ATL glow curves obtained for the six uranium ore samples. Additional lower temperature TL peaks always appear in the ATL glow curves which have evidently decayed and not seen in the NTL. Significant quantity of quartz grains could be extracted only in the case of sample MH32 and the improvements obtained both in resolution and intensity of the NTL are shown in Fig. 2. The NTL outputs and TL ages obtained are given in Table 2. The upper limits of the TL age estimates are found to be quite low.
b) TL age of Thorium ore samples: The NTL obtained from a 'whole' sample as well as its 'quartz extracts' are shown in Fig 2. The pattern of glow curves obtained for all the seven regions is the same with differences only in the relative intensities. The NTL equivalent gamma doses and the TL ages obtained are given in Table 3. The TL age estimates have been extremely low, the oldest value being only $4.76 \times 10^4$ yrs. It is well known that these monazite sands are Detrital deposits which can be quite young. Views have been expressed to the effect that the region around Chavara is probably of Tertiary age (Kaul 1968).

C) Prospecting of monazite deposits: The seven monazite deposits listed in Table 1 were surveyed extensively using a sensitive scintillation survey meter as well as calcium fluoride thermoluminescent integrating dosimeters. (Sunta et al 1971). The TLDs were kept at site for a period of 2 months and the integrated gamma exposure was evaluated in the laboratory. Plots of NTL outputs of quartz extracts versus the gross alpha activities of the respective 'whole' samples are shown in Fig. 3. The radiometric survey results have also been included in the same figure.
DISCUSSIONS AND CONCLUSIONS

Geochronology

The very low TL age estimated obtained can be explained from two different considerations:

1) Low life-times of NTL traps: As a thumb rule, unless the NTL peak occurs at a temperature well above 350°C and has a thermal activation energy much higher than 2 ev, the draining of TL signal at ambient temperature will be significant enough to hinder the correct age evaluation of samples of even 50 m.yr. age (Bonfiglioli 1968). Most of our uranium ore samples possess NTL peaks at temperatures less than 350°C and estimates by 'initial rise method' have yielded activation energy values in the range of 1.14 to 1.98 ev only. In all probability these NTL traps have much shorter mean lives (at temperatures actually experienced throughout their geological existence) compared to the geological ages involved; the NTL may actually represent a state of dynamic equilibrium between the daily build up and drainage rates. In such a case, the TL ages will represent only the order of magnitude of the respective trap meanlives and not the geological ages. Such a consideration does not however apply to the
GEOCHRONOLOGY AND PROSPECTING OF RADIOACTIVE ORES

Figure 4: TL age index v/s alpha activity content of various monazite (solid circle) and uranium (centred circle) ore samples

extremely low values obtained from the 375°C NTL peaks in monazite samples.

ii) Effect of alpha activity on TL age: Zeller et al (1963) have noted a common tendency for the relatively highly radioactive samples to appear too young and explained it in terms of a linear trap production process by the alpha irradiation from the radioactive impurities; hence calibration of NTL by artificial gamma irradiation at present times leads to a lower estimate. Such a trend seems to be operative in our samples too as shown in Fig. 4, where the TL age index is plotted against the alpha activity. This observation at first sight rules out any possibility of using TL of highly radioactive samples for dating. The straight lines obtained in Fig 4 can aid in estimating the geological ages provided a calibration line could be constructed from similar samples of same known age but different radioactivities (Hutchison 1968). However, it may be better to work with TL sensitive minerals collected from locations well separated from the radioactive zones of the deposits, such an effort has already been reported to yield meaningful TL age estimates (Kaul 1965 & 1968).

Prospecting

The monazite prospecting results presented in Fig 3 may cause a surprise in context of the knowledge that the TL sensitive mineral quartz in monazite may have been contributed by rocks of different types and ages in a continuous washing process, hence the various quartz grains may have recorded different NTL doses. Since monazites are mainly contributed by pegmatites (Tipper 1914), the linear relationship obtained in Fig 3 may be taken to indicate that the NTL output has been mainly contributed by pegmatict quartz. The slopes of the two straight lines in the same figure point out that the ‘NTL method’ of prospecting is less sensitive than the radiometric survey method either by using a survey meter or an integrating TL dosimeter. Hence it can only be stated that the ‘NTL method’ can provide at best a convenient supplementary measurement to the conventional radiometric survey measurement. In contrast to the recent suggestion (Vaz et al 1977) that TLDs can be implanted in thorium deposits over long periods and the accumulated TL can be calibrated against Th concentration,
the present method does away with the requirement to make two field trips to implant and then to retrieve TLDs. It is only necessary to collect samples during the conventional radiometric survey trip, separate out TL sensitive minerals like quartz and read the NTL in the laboratory which will provide with supplementary data on radioactive levels.

ACKNOWLEDGEMENTS

Thanks are due to Dr K M V Jayaram of Atomic Minerals Division, Hyderabad for the supply of the uranium ore samples.

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Manuscript Received September 15, 1977
Revised Manuscript Received January 30, 1978
Thermoluminescence of Quartz: Part IV—Effect of Stress on Sensitivity

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Received 20 October 1978

Radiation-induced thermoluminescence (TL) is studied as a function of stress in natural quartz. Stress and strain caused by progressive and impact pressure loading produce change in the TL sensitivity. It is found that sensitivity increases as the amount of stress increases and reaches a maximum at about 1400 kg/cm²; thereafter it decreases. The sensitization obtained is attributed to dislocation produced by stress.

1. Introduction

Effects of treatments like heating and irradiation on the TL of geological quartz specimens have been reported earlier. Stress is an important factor which can affect the minerals in their geological surroundings. McDougall has described various modifying effects of stress on the radiation-induced TL of mineral quartz. Zeller et al. have shown an apparent relationship between geological age and the pressure-induced changes in the TL of carbonate sediments. Haldin et al. have studied the effects of pressure on TL and colouration of certain rocks and minerals. Angino has shown that pressure-induced TL in limestone increases with increasing time of application of pressure. Younger rocks have shown larger increase in TL by application of pressure in comparison with older rocks. Pressure effects on TL of a number of minerals include shift in glow peak temperatures. Effects of pressure on TL of quartz have not been studied systematically so far. The present paper forms a part of our investigation on the TL of geological quartz.

2. Experimental Procedure

Thin plates of 0.5 cm thickness were cut from natural transparent single crystals of quartz along the direction perpendicular to c-axis. These plates were then polished to obtain flat surfaces to ensure uniformity of stress in the samples.

An Instron tensile testing machine was used to apply various amounts of stresses to the samples. Two kinds of stresses were applied to the crystal plates along the c-axis: (1) stress by progressive loading, and (2) stress by impact loading. Quartz plates were then powdered using agate mortar and sieved to a mesh size of less than 100 Tyler for TL studies. The powder samples (5 mg each) were irradiated using a Co-60 gamma source at a rate of $6.9 \times 10^3$ R/min. TL glow curves were read using a laboratory-made apparatus. The readings were taken by heating the sample at 25°C/min.

The desired stresses by progressive loading were developed on the sample by loading at a rate of 500 kg/min. The sample was kept in the state of peak stress for 3 min and then the stress was released at the same rate as in the case of loading. The stresses given to different plates were 308, 617, 1436, 2875 kg/cm². At the stress of 2875 kg/cm², extensive cracks were seen in the crystal.

Stress by impact loading was given by dropping a load of 200 kg from a height of 30 cm on a quartz plate (10 × 8 × 2 mm). The sample got crushed.

3. Results

3.1 Natural Thermoluminescence (NTL) of Stressed Samples

NTL (the signal naturally present in the sample prior to any artificial irradiation) glow curves were recorded for all the stressed samples along with an unstressed natural sample for comparison (Fig. 1). Two glow peaks were observed, one at 250°C and another at 350°C. Significant variation in TL output...
as well as in relative intensities of peaks was observed. Areas under NTL glow curves were also plotted (Fig. 2).

3.2 Radiation-induced Thermoluminescence of Stressed Samples

3.2.1 Progressive loading—The natural samples after different amounts of stresses were subjected to artificial (test) irradiation at \(2.4 \times 10^6\) R from a Co-60 gamma source and the glow curves recorded (Fig. 3). An overall change in the TL output was observed in most of the stressed samples. No selective enhancement or reduction was observed in the individual glow peaks. Areas under the glow curves were plotted against the stresses (Fig. 4).

The stressed samples and an unstressed sample were subjected to a heat treatment at 400°C for 90 min so as to remove NTL completely. These samples were then irradiated at \(2.4 \times 10^5\) R and the glow curves drawn (Figs. 5 and 6).

In another experiment, a stress of 2000 kg/cm\(^2\) was given to a quartz plate. The sample was kept in the state of peak stress for a duration of 42 hr and then released at the same rate as used in loading (500 kg/min). The TL response of the sample vis-a-vis that of an unstressed sample is given in Fig. 7.
then irradiated to a test exposure of $8 \times 10^4$ R and the glow curve was recorded. It is found that the TL sensitivity is reduced uniformly in all the glow peaks by a factor of 5.

4. Discussion

There is in general no effect of stress on the temperature of the glow peaks except that the glow peaks of 250 and 350°C apparently get closer to each other when TL intensities are large either due to pressure or dose. This apparent change may be due to superimposition of the peaks. Also there is no change in the relative intensities of the individual glow peaks (Fig. 3) except when the sample is heated in between the stress and the test radiation exposure (Figs. 5 and 6). Absence of change in glow peak temperature means that the defect centres responsible for TL emission are the same in the stressed and the unstressed samples. Enhancement in TL intensity by stress should mean an increase in the number of these defect centres or in their radiative recombination or both (Figs. 1, 2, 4-6). High pressure beyond a certain limit, however, causes damage to TL intensity.

Except the stress of 308 kg/cm², all other stresses used in the present study are higher than the tensile strength of quartz (7000 psi) (Ref. 10). Such stresses would cause extensive dislocations in the lattice. It appears that the TL centres (traps and recombination centres) produced in this process are of the same type as present in the virgin sample. There is no change in the glow peak temperatures. If the stress-induced sensitization is related to the dislocations, it would be initiated at the elastic limit and would reach a maximum during the early stage of permanent deformation. At still higher pressures, the TL centres might be getting annihilated due to the extensive damage to the lattice, like conversion to glassy state. The reduction in TL sensitivity due to impact loading may be attributed to this type of annihilation of TL centres due to heavy impact.

The changes in TL intensity at 250 and 350°C glow peaks are quite conspicuous (Figs. 1, 2, 5, 6). These glow peaks are of geological significance, since these are the only major peaks which survive in the geological environmental conditions. These results show that in geological applications of TL, such as TL dating etc., this factor (stress to which the specimen was subjected in the host rock during its geological life) will have to be taken into consideration.

References

Thermoluminescence of Quartz: Part V—Effect of Polarization on Sensitivity

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Received 20 October 1978

Effect of polarization on thermoluminescence (TL) glow peaks of a natural quartz sample is studied. The glow curve consists of five distinct glow peaks at 70, 120, 180, 250 and 350 °C of which only the last two are present in the virgin natural sample. Polariation enhances the intensity of these two glow peaks in the natural sample. Gamma irradiation after polarization also enhances only these two peaks, the other three glow peaks are unaffected. Polarization during irradiation suppresses the intensity of all the five glow peaks but produces a new glow peak at 300 °C. Effect of polarization is removed on heating. These results are discussed and attributed to different configurations of Al³⁺ impurity centre in the SiO₂ lattice.

1. Introduction

Radiation-induced thermoluminescence (TL) of natural quartz is known to be affected by different treatments like heat, pre-irradiation and stress. This paper shows that the TL sensitivity is found to be changing also with the electric field applied across the crystal before and during irradiation.

2. Experimental Details

A simple device is made to give electric potential to the crystal. This consists of two bakelite plates of 6 x 6 x 1 cm size with electrodes attached in the middle (Fig. 1). The sample is kept in between the electrodes and the assembly is fixed with four screws at the corners of the plates. The set-up is small and can be easily placed in the gamma irradiation chamber. The irradiation facility consists of Co-60 pencils arranged in annular fashion inside a lead chamber. Samples can be sent to the chamber through a drawer, for irradiation. The exposure rate is about 1000 R/min.

The quartz sample used in this study is a clear transparent natural crystal grown in nature under hydrothermal conditions. The sample was cut perpendicular to c-axis to form plates of 4 mm thickness and polished well to achieve parallel surfaces for a good contact of the crystal with the electrodes. The sample was sandwiched between the electrodes and the electric potential was given across the crystal from a high voltage dc unit. The instrumentation to read the TL signals is fabricated in the laboratory and described elsewhere.

After polarization, these crystals were powdered to a size of less than 100 Tyler mesh, and these samples were used for the TL measurements. In each reading, 5 mg of sample was heated at a rate of 25 °C/min and the TL glow curves are recorded simultaneously.

3. Results and Discussion

Without removing the natural thermoluminescence (NTL), one sample was kept under a dc voltage of 1300 V for 16 hr. Readings were taken using this sample along with an unpolarized natural sample for comparison. Glow curves recorded are shown in Fig. 2 (curves 1 and 2). This polarized sample was given an exposure of 6 x 10⁴ R in the Co-60 gamma chamber and the glow curves were recorded. These glow curves together with readings of an unpolarized sample are also given in Fig. 2 (curves 3 and 4).
observations on optical absorption and electron paramagnetic resonance (EPR). The colouring has been attributed to substitutional ion in the SiO₂ lattice. In the unirradiated state, the Al³⁺ ion is associated with an adjacent interstitial monovalent positive ion such as Na⁺ or H⁺. According to the model, irradiation removes an electron from this centre leaving a hole behind, which is mostly associated with oxygen adjacent to the aluminium. In this process, the positive compensating ion moves away from the vicinity of the Al³⁺ ion, since its presence is no more essential for charge compensation. Annealing at 350°C causes the recombination of the electrons and holes and thus bleaching of the colour centres.

A similar colouration is observed by Krefft in his experiment on electrolysis of quartz at high temperature and in high vacuum. In contrast to natural and radiation-induced colouration, this type of colouration (by electrolysis) is found to be stable even at a temperature of 1000°C. This result has been attributed to the removal of the compensating ion as well as the electron permanently from the