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A Study of Heated Sand With Thermoluminescence: An Estimation of Thermal History in Building Materials

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Abstract
When sand is heated slowly, an emission of light will appear and this luminescence phenomenon is called thermoluminescence (TL). TL can be observed by thermally stimulating sands at various temperatures and exposure times. The amount of TL however, is reduced if the sand is exposed to high temperature as in the case of fire. The objectives of this study are to evaluate the natural TL of prepared heated sand of 100°C and 300°C at various exposure times, to compare the natural TL of heated sand treated with 10% and 30% of HCl concentration and to estimate the maximum thermal history reached within heated sand based on self-normalisation method. This is to determine that whether the strength of concrete is structurally sound or not because sand is one of the constituent materials in concrete. The samples of heated sand were prepared same as that of fine-grained dating method and treated with two different HCl concentrations of 10% and 30%. Based on the results, it was found that the natural TL reduced as the exposure times increased. The effect of using higher (30%) HCl concentration yielded 16.8% improvement and a clear TL glow curve shape. The estimation of the maximum thermal history of heated sand using self-normalisation technique is dependent upon the duration of thermal exposure. The result achieved can be used as guidance in estimating maximum thermal history reached within any building materials or structures that contained sand.

Keywords
Thermoluminescence, heated sand, thermal history, building materials

1. Introduction

Research on fire damaged concrete with thermoluminescence has successfully reported by Chew (1989 & 1993) and was first introduced by Placido (1980). Crushed sand extracted from heated concrete has been used to test on its remaining thermoluminescence (TL) signal and maximum temperature history due to fire exposure. The aimed is concerned on the strength of the concrete whether it is structurally sound or not. Chew (1989 & 1993) has proposed the self-normalisation technique to analyse the TL glow curves in

TL test. The basis of TL test appeared to be reliable and non destructive compared with other tests which include color analysis, core testing, ultra pulse velocity test, thermogravimetry etc.

Thermoluminescence (TL) is part of luminescence phenomena. TL is defined as the emission of light during heating of a solid sample (insulator or semiconductor), previously excited by radiation. The emitted light then is measured by light sensitive detector such as photomultiplier and associated electronic equipment. By plotting the measured light output of heated sample against its temperature somehow produces a characteristic glow curve of the irradiated sample. The essential condition for thermoluminescence to occur in a solid is that the material must have been previously exposed to radiation. This radiation (UV, ionising, visible light, etc) is the source of energy that absorbed and stored by TL materials until it get heated. McKeever (1985) stated that three factors contributed for the production of TL. This include; (1) an insulator or semiconductor with a crystalline structure, (2) during the exposure to radiation the material should absorbed some energy at a time and, (3) during heating the material the luminescence emission should be produce.

TL is a luminescence phenomenon of an insulator or semiconductor which can be observed when the solid is thermally stimulated (Bos, 2001 and 2007). The observation of thermoluminescence is under condition of steadily increasing temperature. In normal TL experiments, TL material is irradiated at room temperature and later heated through a temperature range where the luminescence is bright, until a temperature level at which all the charges have been thermally excited out of their metastable levels and the luminescence completely disappears (Furetta, 2004). However, in this study no irradiation sources being applied and TL measurements are solely depends on the natural radiation or original energy stored within the lattice structure. The regenerated curve then being observed with subsequent preheating to ensure of the remaining TL left within the sample.

2. Sand as TL Material

Few research works has been carried out in estimating thermal history of the concrete heated by a fire or other treatments using thermoluminescene (TL) method. Sand is one of the constituent materials in preparing concrete and contained luminescent minerals which capable of emitting light when heated.
Two important constituent of sand are quartz and feldspar which are well known to show thermoluminescence phenomenon. Sand also contains varying concentration of heavy minerals. It can be used as a dosimeter during nuclear emergencies like reactor accidents for monitoring a large number of people. The dosimeter can also be use for personnel radiation dose measurement while handling highly radioactive materials i.e. in kilocurie range (Vaijapurkar *et al.*, 1997).

Apart of using heated concrete to test with TL, heated sand has been used for this study as fundamental of concrete exposed to fire so that one can estimate the maximum thermal history attained within it. Sand exhibited good luminescent mineral and capable to absorbed and stored the energy which previously irradiated by ionising radiation such as U, Th, K, cosmic rays, etc. (Michael *et al.*, 1997). When sand has been exposed to heat for a particular time, it TL signal will be reduced depending on how deep the electron trapped within the lattice. The remaining TL left in heated sample then measured in TL reader based on self-normalisation method. The result somehow gives an indication of maximum thermal history when TL emission data plotted as a function of temperature.

The idea of this study is to perform an experimental study of heated sand with thermoluminescence using TL dosimeter reader with higher constant heating rate of $10°Cs^{-1}$ and subject to the following objectives:

a) To evaluate the natural thermoluminescence of heated sand with determined temperature of 100°C and 300°C at five exposure times of 5 minutes, 10 minutes, 30 minutes, 1 hour and 2 hours,
b) To assess and compare the natural thermoluminescence of heated sand treated with 10% and 30% of HCl concentration , and

c) To investigates the estimated maximum thermal history reached within heated sand based on self-normalisation method

3. Experiment Set-Up

The main objective of this research is to study and perform the maximum thermal history of heated sand based on a range of temperature with determined exposure times. Therefore the following experimental programmes were undertaken.

a) Sample preparation by using fine-grain technique,
b) Measurements of natural TL signal of heated samples, and
c) Measurement of remaining TL left in the heated sand based on the self-normalisation technique

3.1 Sample Collection

The raw sand particles are range in diameter from 0.0625 (or 1⁄16 mm) to 2 millimetres that have been exposed to natural sources of radiation (U, Th, K, cosmic rays, etc.). The technique used for sample preparation was fine-grained method (Zimmerman, 1971). This is because sand consists of polymineral grain and this method is one of the options for avoiding inhomogeneities caused by non-uniform radiation field. Before sand taken for thermal exposure, it should be sieve and dry in order to obtain the smallest sand particles. The smallest sand grain then crushed with an agate pestle and mortar to further fine the grains. The crushed sand is now ready for thermal exposure.

3.2 Determination of Sand Thermal Exposure

To demonstrate a ‘so-called’ fire scenario, an isothermal furnace has been employed as a basis of thermal exposure for sand samples. The processed was reliable in maintaining constant temperature at desired exposure times thus makes reproducible sample for this TL measurements study.

a) Furnace

Carbolite furnace with temperature controller was used for heating of the samples. Powerful free radiating coiled wire elements on both sides and ceiling of the chamber ensure good thermal uniformity. It uses rigidized low thermal mass insulation to ensure rapid heating and hard ceramic hearth provides robust base.

Figure 1: Sample attached with thermocouple.

Table 1: Samples subjected to various thermal exposures.

Temperature	Exposure Time
100°C & 300°C	5 minutes, 10 minutes, 30 minutes, 1 hour & 2 hours

b) Procedure

A sample of crushed sand (about 500 mg) then placed in a small aluminium container into the preheated oven at a pre-selected isothermal temperature. This thin layer of sand was inserted with thermocouples type K (chromel-alumel) at the centre of the container as shown in **Figure 1**. In order to observed constant heating during thermal exposure, thermocouple wire is connected to a data logger Yokogawa DAQ station type DX230. The sample size required for thermal exposure depends on the number of TL disc and distribution of the amount of sand particles for each TL disc. Table 3.1 shows various sand samples subject to determined thermal exposure. These heated samples are now ready for chemical cleaning process.

c) Chemical Cleaning Process

The heated sand samples contained of carbonates and other soluble compound which later affect on the TL intensity during the read-out. To remove this unwanted compound, heated sample has been carefully treated with 30% concentration of HCl. The quantity of HCl used is double the amount of sample. Then another set of same heated samples will be treated with 10% of HCl concentration. Later, HCl has been rinsed off from the samples using water. The remaining water is then removed using methanol followed by acetone. The fine grains particles ranging from 1 to 8µm are fully gained and suspended in acetone. The samples are now ready to be deposited on TL disc.

d) Chemical Cleaning Process

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d) Sample and Disc Preparation

In order to achieve good heating contact between sample and heater plate, very thin samples and TL disc are made. This is to make sure no thermal gradient across the thickness of aluminium disc which relatively higher temperature occurred at the bottom surface that directly contacted the hotplate. Magnetic stirrer has been used to agitate the mixture of the particles and acetone so that it equally distributed into the disc. Calibrated pipette with fixed volume aliquot has been used to take out the grains from the test tube and mounted on 0.5 mm thick and 10 mm diameter of aluminium disc. Drying of samples was performed at room temperature to minimize thermal bleaching effects and under low light illumination. All dried samples are kept in light tight containers and prepared for TL measurements.

3.3 Measurement of TL

3.3.1 Self-Normalisation Technique

A self normalisation method that first developed by Chew (1988, 1993) has been applied for interpretation of heated concrete TL results. This is to make sure that the difficulties in interpretation of TL result from the same sample can be resolved.

The TL reader used was Harshaw Bicron model 3500 manual TL dosimeter reader (Figure 2a). As this is a manual reader, the measurement is carried out one disc at a time. A series of heated samples from the same thermal exposure are now ready for read out process. Five samples disc will be measured for 'first' TL signal at 10°C/s^{-1} of heating rate for reproducibility of TL glow curve. During the read out, TL signal is increased as temperature increased in a linear manner. The best average curve will be taken as a standard curve and noted as 'natural' TL which can be explained as the TL emission produced by the unirradiated portions. It is known as the natural TL, since it results from the radiation dose accumulated by the sample in the natural environment over geological time. The remaining samples are then subjected to various preheated exposures before their TL is measured and the same process applied (five samples for each preheated exposure).

For example, a series of 100°C of 30 minutes exposure will be tested for 'Natural' TL and others are preheated to 100°C, 150°C,200°C, etc before their TL is measured. This preheated TL results are then compared with 'Natural' TL curve. The lowest preheating curve that is able to reduce the TL signal or the one that fits best with 'Natural' curve is presented as peak thermal history for this particular heated sample (100°C-30mins). The disc has to be placed exactly above the position where the thermocouple is fixed on the planchet so that good temperature transfers to the sample.

(a) **(b)**

Figure 2: (a) Harshaw 3500 TLD reader and (b) planchet heating with read-out pan

Table 2: Samples with preheated temperatures.

Thermal exposures of samples treated with 10% & 30% HCl	Preheated Temperature
100°C-5 MINS	100°C, 150°C & 200°C
100°C-10 MINS	100°C, 150°C, 200°C & 250°C
100°C-30 MINS	100°C, 150°C, 200°C, 250°C, 300°C & 350°C
100°C-1 HR	100°C, 150°C, 200°C, 250°C, 300°C, 350°C & 400°C
100°C-2 HRS	100°C, 150°C, 200°C, 250°C, 300°C, 350°C & 400°C
300°C-5 MINS	300°C, 350°C & 400°C
300°C-10 MINS	300°C, 350°C & 400°C
300°C-30 MINS	350°C & 400°C
300°C-1 HR	350°C & 400°C
300°C-2 HRS	400°C

4. Results and Discussion

The TL intensity based on the light emission of the heated Thermoluminescence sample of 100°C and 300°C with determined exposure times of 5, 10 and 30 minutes, 1 hour and 2 hours was measured using a Harshaw-Bicron model 3500 TLD reader. The reader was calibrated to ensure on the reproducibility of the glow curves as shown below. The glow curves were obtained using a constant heating rate of 10°Cs^{-1} with an operating temperature of 0°C up to a maximum temperature of 400°C. The samples tested for TL are of the same size, weight, storage and handling.

4.1 Reproducibility of TL Glow Curve

The measurements of TL intensity are plotted as a function of temperature (or time) and the resulting graph is called glow-curve. The glow-curves have one or more maxima called glow-peaks and are function of various energy level traps. The reproducibility of TL glow curve has been checked by running of five TL measurements from the same heated sample. The significant is that by repeating a measurement several times would yield the same curve shape during the measurement.

4.2 Comparison Of Natural Tl Curve - 30% And 10% Of HCl.

Figure 4a and 4b shows a comparison of heated sample of 100°C-5 minutes and 100°C-10 minutes treated with two different HCl concentrations. A slight difference of TL is observed for sample treated with 30% of HCl compared with that of 10% of HCl concentration. It can be deduced that sample treated with 30% HCl have less impurities, thus have an effect on the reproducibility of TL curves. Further comparisons of 300°C samples with different acid treatment are not required since the result deduced not much different as in the case of 100°C samples mentioned above.

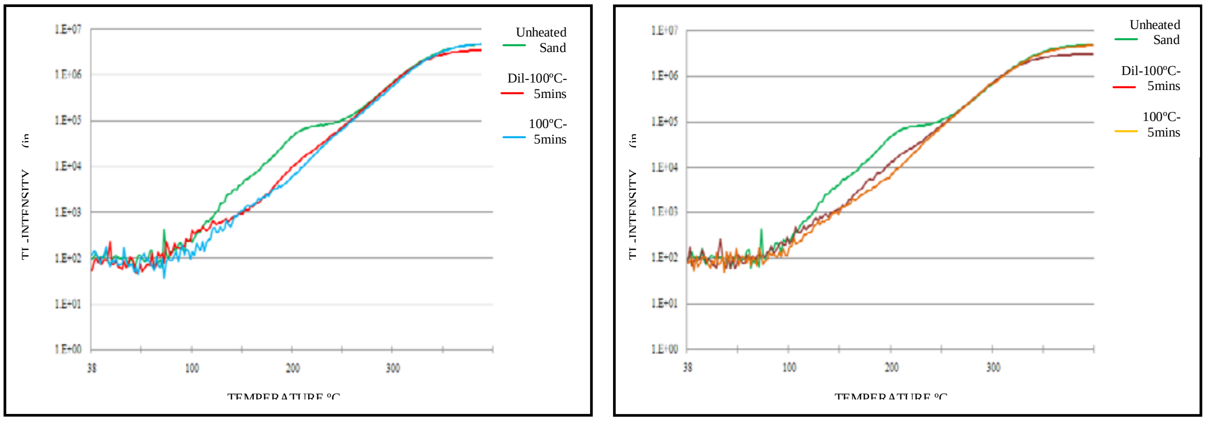
4.3 Self-Normalisation Results

4.3.1 Natural and Preheated Glow Curve for Sample Heated to 100°C and 300°C

In the case presented here, the natural TL intensities for the heated sample of 100°C and 300°C at various exposure times are of the same plot shape either treated with 30% or 10% HCL concentration. Although there are notable reductions of TL with regard to the increase of duration of exposure when treated with 10% of HCl, the treatment with 30% HCL concentration shows a better result. This is attributed to the elimination of impurities during the chemical treatment process. By application of two set chemical treatment mentioned above, samples which had been treated with 30% HCl concentration are taken as a standard curve for the analysis of the self-normalisation technique (Figure 4a and 4b).

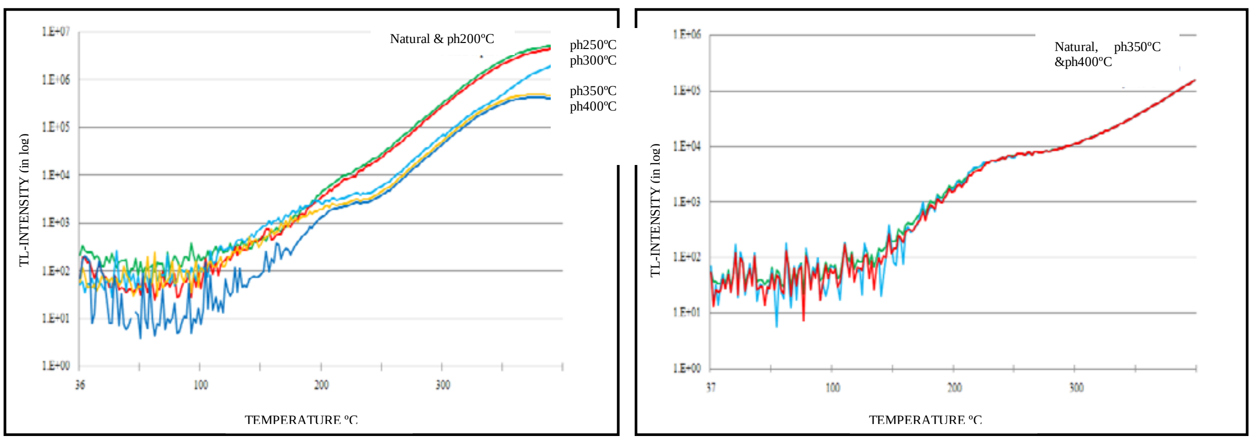
It should be mentioned that in the case presented here, the unstable curves or spikes are always occurred at the lower temperature during the TL read out of the heated samples or at subsequent preheating TL measurement. Generally, the spikes may be caused by electrical interference producing very high readings (Piniella, 2001). However, in custom dosimetry, it is possible to prove the existence of abnormal curves occurring due to spurious effects. Such spurious effects do occur even though extra precautions have been taken during preparation, handling and storage of the heated sample. Some of the possible phenomena that may account for this effect are intrinsic defect such as ion vacancies, crystal defect, chemical treatment process, etc.

The TL response of samples that had undergone the same treatment may not necessarily be the same. That is why self-normalisation method (Chew, 1988) has been applied for determination of the maximum thermal history of heated sand. Table 3 shows the summary results and apart of using sample drilled from the heated concrete, heated sand also exhibited similar TL signal during TL measurement.



(a) **(b)**

Figure 3: Effect of the (a) 100°C-5 minutes and (b) 100°C-10 minutes sample upon different chemical treatment.



(a) **(b)**

Figure 4: Effect of subsequent preheating glow curves upon (a) 100°C-2 hours and (b) 300°C-2 hours sample.

Table 3: Summary of the self-normalising results

Sample	Max. Temperature by self-normalising (°C)
100°C-5 MINS	100°C - 150°C
100°C-10 MINS	150°C - 200°C
100°C-30 MINS	200°C - 250°C
100°C-1 HR	200°C - 250°C
100°C-2 HRS	200°C - 250°C
300°C-5 MINS	300°C - 350°C
300°C-10 MINS	350°C - 400°C
300°C-30 MINS	350°C - 400°C
300°C-1 HR	350°C - 400°C
300°C-2 HRS	> 400°C

5. Conclusion

This study presents thermoluminescence as a technique applied to evaluate the TL signal of the heated sand. The self-normalisation method was employed to estimate the maximum thermal history of the heated samples. It was found that this indirect method has effectively overcome the difficulties encountered during the interpretation of the TL result especially when dealing with the samples of same exposure time, batch or homogeneity, storage, handling, etc. The critical findings derived from the experimental results are summarised as follows:

1. For the natural TL intensity of heated sand of 100°C and 300°C with various exposure times, it was found that the TL decreased as the exposure times increased. The shape of the natural TL curves yielded two distinct peaks with first peak occurred between the temperatures of 200°C and 250°C and the second peak at temperature greater than 300°C. This could be explained by the presence of electron traps in the heated sand which is attributed to the defects in the lattice structure.
2. The natural TL curve yielded 16.8% improvement when treated with 30% of HCl compared to 10% of HCl. This improvement can be seen at the lower temperature of the curve as less spike curve or abnormal curve occurred during the initial TL read-out. Furthermore, higher HCl concentration removed more impurities contained within the samples thus spurious TL was reduced during TL measurement. By this, the natural TL results for samples treated with 30% of HCl were taken to estimate the maximum thermal history of heated sand.
3. For the estimation of the maximum thermal history of heated sand using self-normalisation technique, it was found that the results are dependent upon the duration of thermal exposure. This is shown by the differences in the estimated maximum temperature history from the samples that were subjected to different exposure times.

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