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# KINETIC STUDIES OF QUARTZ THERMOLUMINESCENCE AS APPLIED TO SEDIMENT DATING

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## INTRODUCTION

The calibration of the radiation sensitivity of material with the help of powerful laboratory radiation sources is a fundamental component of the thermoluminescence (TL) dating method. However, the accuracy of using such sources is poorly substantiated, even though it is of special importance since there is a change in dose rate of 6-8 orders of magnitude when transferring from natural to laboratory conditions. Studies of this problem have been one of the principal objectives of our research work. In addition, we have investigated the use of ultra-violet light to bleach the TL stored in minerals since bleaching is the dominant process for zeroing the TL chronometer in sediment dating. To study palaeodosimetric problems more precisely, we focussed our attention on quartz, taking the 300°C TL peak lightsum (integrated emission) as a basic unit of measurement. We also studied the behaviour of the 180°C TL peak which is absent in the natural glow curve, but present with high sensitivity after artificial radiation .

## EXPERIMENTS AND DISCUSSION

Our studies have confirmed that the initial section of the dose response curve is complicated by a form of supralinearity (fig. 1.). Unlike data published elsewhere, which states that this phenomenon is true for a small proportion of samples, we have found supralinearity in all the samples we have studied. In a number of cases it even transforms into a section with negative sensitivity and subsequent normal growth after passing through a minimum value (fig. 1, curve 2). Such a phenomenon is called by us a "dose pit" and is typical for one third of the samples under study.

Good correlation between the level of dose at which the 300°C peak supralinearity finishes and that at which onset of saturation of the 180°C peak occurs can be seen in figure 1. Such behaviour can be explained by shallow traps acting as a competitive mechanism. The dose-pit phenomenon can also be explained within the framework of this theory if a radiation-induced decay component is taken into account as well as a thermal decay component, which is small for deep traps. This process can be described by the following formula:

$$\eta = 1-(1-\eta_0)\exp(-\kappa D) - p[1-\exp(-q\kappa D)]$$

which is presented graphically in figure 2, where

$\eta$ = normalised trap population relative to the saturation level,

$\kappa$ = radiation sensitivity,

D = absorbed dose, and

p and q = parameters characterizing the competing decay processes.

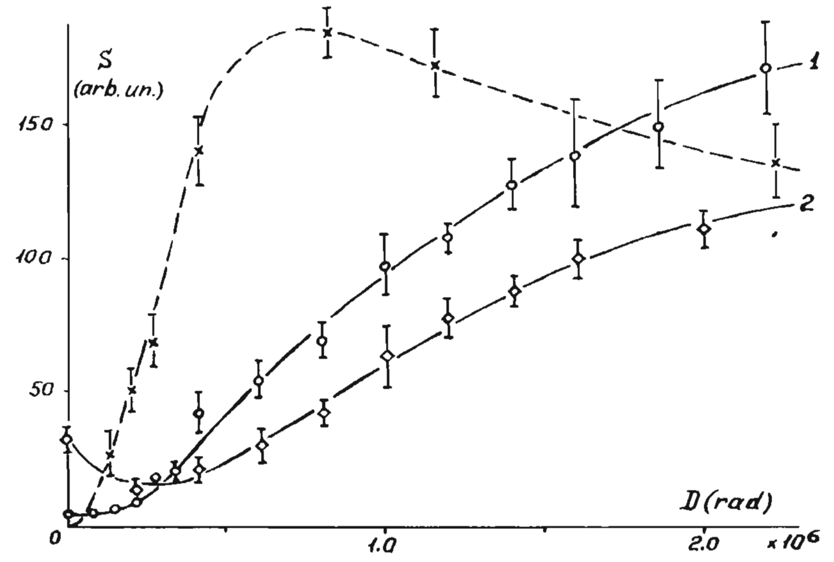



*Figure 1.* *Dose dependencies of a lightsum of the 300°C (solid line) and 180°C (broken line) peaks of the quartz TL showing; 1) simple supralinearity and 2) the "dose-pit".*

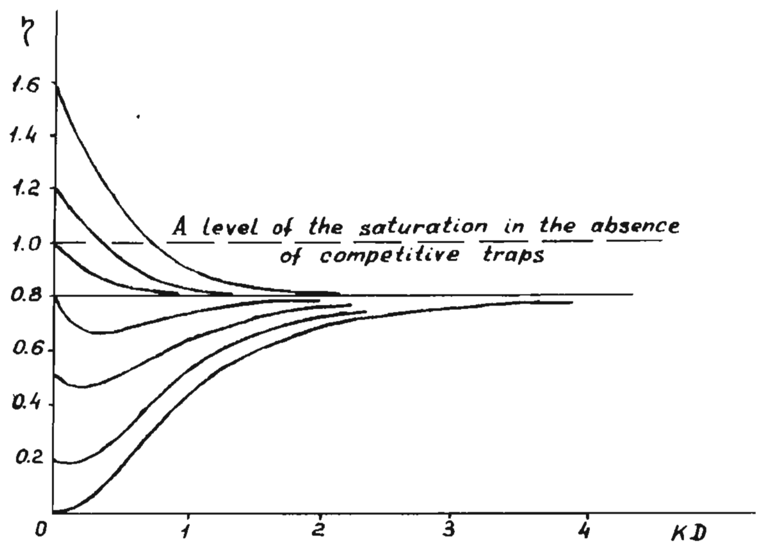



*Figure 2.* *Theoretical dose dependencies in the presence of competing thermal and radiation fading.*

Since the 180°C peak is absent from the natural glow curve, we have every reason to assume absence of supralinearity under natural irradiation conditions (i.e. during antiquity). Under laboratory conditions, in addition to the 180°C peak, there are other peaks present at lower temperatures which may be capable of yielding an analogous effect.

When observing changes in the lightsum at 300°C using doses of up to 5 krad, we have also observed complex fine structure which is difficult to characterize. We presume this arises from competition processes within the trap spectrum which introduce additional distortions into a laboratory dose curve.
At higher doses (above100 krad) we have also observed an "abnormal" signal in the region of the 300°C peak, which is capable of being several times greater than the "normal" component. The main distinguishing feature of this abnormal TL signal is its insensitivity to UV radiation and its generation only under artificial ionizing radiation .
On the basis of these studies, we came to the conclusion that there were fundamental differences in the TL response of samples under natural and laboratory irradiation and that this brings into doubt the validity of established methods of using quartz as a palaeodosimeter in which the natural TL is compared with the response to laboratory irradiation.

**The Effect of UV Radiation**
In addition to de-activation of the 300°C peak, we have found that UV irradiation can also cause activation. The latter has been observed in the form of a regenerated 300°C peak after complete thermal annealing of the sample (450°C,10 min.). Lower temperature peaks, similar to those produced by gamma irradiation, are also produced. The growth of the 300°C peak lightsum with UV dose is not of a simple exponential form and the equilibrium level is the result of competing UV-stimulated activating and deactivating processes. Under conditions of solar exposure at middle latitudes, the UV equilibrium lightsum is achieved after one month of exposure. This value, which may serve as a "theoretical zero" of the TL chronometer for sediments, is easily reproduced under laboratory conditions using unfiltered light from a 120W mercury lamp (@ 25cm for 1h).

## RESULTS

Our research work has resulted in the development of a new approach to TL dating, the main feature of which is the exclusion of individual radiation calibration of samples. In the absence of competition by shallow traps and "abnormal" TL, the dose dependence of the 300°C peak lightsum follows a simple law of saturation. At the present time, we do not possess sufficient data to establish the order of the kinetics. However, use of two extreme possibilities, with either first or second order kinetics, yields no more than a 20% difference (fig. 3) in the age calculation. For first order kinetics the following exponential growth curve is obtained:

$$S = S_\infty - (S_\infty - S_o) \exp(-\kappa D)$$

where: S = a lightsum, and "o" &"∞" subscripts indicate the initial state and level of saturation respectively.
When determining D, the absorbed dose, for a given value of $S_o$ it is necessary to know the two parameters $\kappa$ and $S_\infty$. $\kappa$ is a physical constant universal for a given crystalline lattice with a particular trapping centre, and may be determined in advance and used for all the samples. Individual sample properties determined by particular trap concentrations are reflected in the $S_\infty$ parameter.

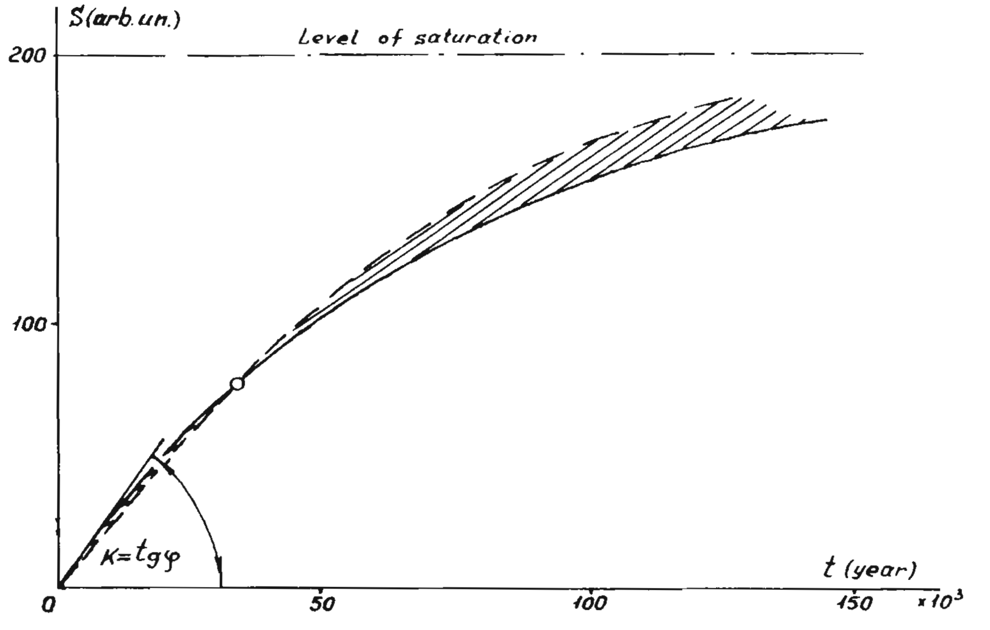
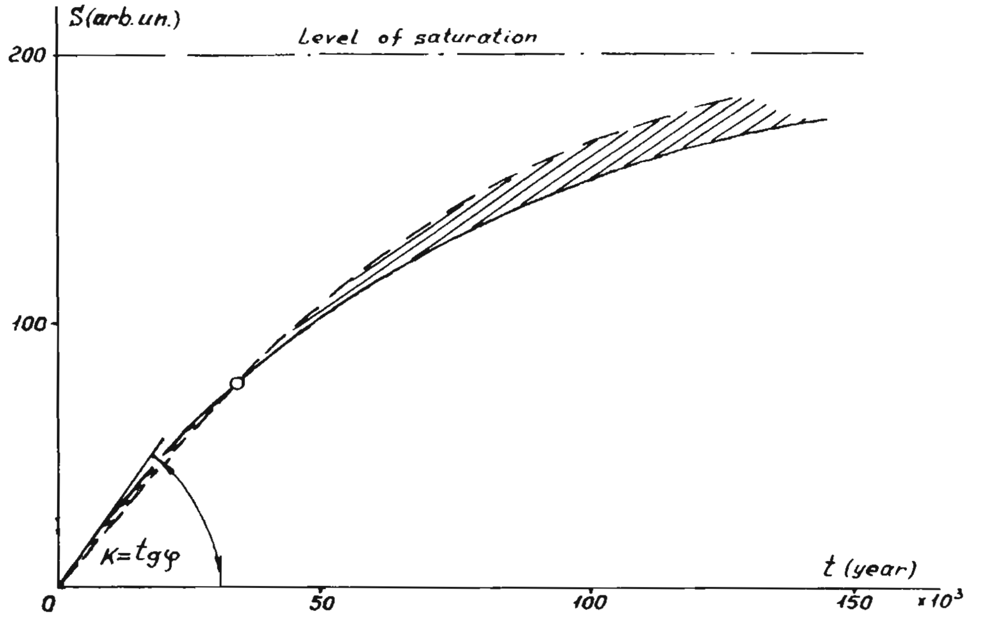



*Figure 3.* *Dependence of the lightsum arising from natural irradiation;*

| | |
|---|---|
| ___ | *1st order* |
| - - - | *2nd order* |
| ///// | *limits of uncertainty* |
| o | *calibration point* |

The analysis of samples from a collection covering a vast area of the European part of the USSR has revealed that for samples with ages ranging from the beginning of the Middle Pleistocene to the Upper Cretaceous, the natural glow curves are identical within experimental error limits. We thus infer that they are all in saturation, and can be used to obtain $S_\infty$ for this area. In this way, one of the unknown parameters has been provided by nature itself. A contradiction between a measured saturation of 300-400 ka and a theoretical prediction of 2-3 Ma, based on the thermal stability of the trapped electrons, should be noted. We can explain this contradiction by the exclusion of a radiation-induced decay component in the theoretical model; however, its existence has been recorded by us experimentally. Hence early saturation testifies to the predominance of a radiation-induced decay component over a thermal one.

Having established $S_\infty$, the radiation sensitivity value $\kappa$ can now be obtained for a sample of known age (dated by $^{14}C$). We estimate $\kappa$ to be $4.10^{-4}$ rad $^{-1}$ when the age, t, and the in-situ dose-rate, E, are inserted in the equation above with D = t.E .

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The age of an unknown sample may now be calculated with the following formula,

$$t = (\kappa E)^{-1} \ln \{ (S_\infty - S_0) / (S_\infty - S) \}$$

where, E , the in-situ dose-rate, was determined using an industrial scintillating (NaI) prospecting radiometer. The $S_0$ value (a theoretical zero) was taken to be the lightsum remaining after a three hour exposure performed with the mercury lamp. We assume that the error arising from incomplete zeroing under natural conditions is small compared with the age being measured.

The method was tested using material from Middle-Late Pleistocene deposits from the Central Russian Plain and the lower Volga. With a few exceptions, the results obtained show good internal consistency and compare well with geological estimates of the age.

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We wish to thank Dr. M.V. Fock for regular consultations and practical recommendations on problems of thermoluminescence kinetics.

**Invited Paper.** This paper was first presented by the authors at a conference "Methodology of TL Dating" held at Tallinn 20-24 April 1986.