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THE VARIATION OF TL PROPERTIES OF SYNTHETIC QUARTZ BY THERMAL ANNEALING

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Abstract — The previously reported supralinear dependence of the 110°C peak in synthetic quartz on the dose of beta excitation has been further studied. The transition from strong supralinearity to linear or weak supralinear behaviour has been investigated in some detail. In the unannealed material, an initial more than quadratic dose dependence has been observed, behaving like D^k where D is the dose and k a number up to 3. Annealing at high temperature increased substantially the initial sensitivity and reduced the degree of supralinearity. The transition occurs between 500 and 1000°C, depending partly on the grain size. In this temperature range the initial sensitivity increases by at least an order of magnitude, and the dose dependence reduces gradually, in some cases down to near linearity. In intermediate temperature annealings within the above mentioned range, a transition between low and high values of the supralinearity factor k has been found in the dose dependence curve, which seems to shed some new light on the understanding of supralinearity in this material.

INTRODUCTION

The change of sensitivity to beta dose of the 110°C peak in fired quartz as a result of an exposure to rather large doses followed by annealing is a well known phenomenon[1,2] in natural quartz which led to the pre-dose method of dating archaeological quartz. The annealings in these cases were usually up to about 500°C.

David *et al*[3,4] showed a substantial increase of the sensitivity by firing to temperatures between 500° and 1000°C (with no prior irradiation). They point out that the originally alpha quartz changes at 573°C to beta quartz and at 870°C to tridymite. In their results, the sensitivity in natural quartz changes by a factor of about 3 near the alpha-beta transition and there is a small peak in the plot of sensitivity against firing-temperature near the second transition near 900°C. Kristianpoller[5] reported an increase in the sensitivity of the 110°C peak by annealing synthetic quartz to about 800°C where the irradiations were by vacuum UV.

The properties of the 110°C peak in synthetic quartz following beta excitation were investigated by Chen *et al*[6]. They found that the dependence on the dose of the unannealed samples was strongly supralinear. Annealing the samples at high enough temperatures increased the sensitivity substantially (much more than the above mentioned results in natural quartz), and changed the dose dependence to be linear or nearly linear. The rather strong supralinearity was attributed to an effect of competition during the heating phase. This had first been suggested by Rodine and Land[7] and later a theoretical formulation was given by Kristianpoller *et al*[8]. According to these authors, the occurrence of a competitor during the heating phase should result in a quadratic or even a more than quadratic dose dependence under certain circumstances. The supralinearity in this case is expected to start from the very low doses. Chen *et al*[6] suggested that the annealing of synthetic quartz removes the competitor which results in an increase in the sensitivity and a decrease in the degree of supralinearity, as expected by the above mentioned theoretical considerations

Another kind of competition, namely competition during excitation, has also been reported[9,10]. In this situation normally expected behaviour would consist of a linear-supralinear-linear sequence, which means that within a certain dose range, the degree of supralinearity increases with the dose.

We report more results of the supralinear dependence on the dose of the synthetic quartz powder. Further results on the dependence of the TL intensity on the annealing temperature are given. These experimental results are discussed in the light of the two models mentioned for explaining supralinearity.

Some preliminary measurements indicate a dependence of the results on the grain size. A dependence of the sensitivity on the grain size in some materials has previously been reported (for a summary see Horowitz[11]). An increase in the sensitivity with grain size was found in some materials whereas a decrease has been seen in others.

EXPERIMENTAL PROCEDURE

The synthetic quartz used in this work was Premium Q material produced by Sawyer Research Products. The quartz samples were crushed and ground. Four to ten milligrams of powder were placed in a stainless steel dish which was placed on a nichrome heating strip. TL was recorded at a constant heating rate of $5°C.s^{-1}$. The emitted light was detected by a 6255B photomultiplier tube. Appropriate optical filters were used in order to reduce the black body radiation. The temperature was monitored by a Chromel-Alumel thermocouple. The oven was evacuated first to about 13.3 Pa, then filled with nitrogen gas. For high temperature firing, the samples were annealed in air in a furnace for an hour and then cooled to RT over two hours.

The irradiation was carried out using a ^{90}Sr β source. Different doses were achieved using different irradiation times ranging from 30 s to 10 min.

RESULTS

Some results of TL dependence on the dose are shown in Figure 1. The strong supralinearity of the unannealed material is seen in Curve A. The slope on the log-log scale is k=3. Curve B depicts the dose dependence results after annealing at 250°C; the results are changed only slightly and the degree of supralinearity reduces to k=2.6. Curve C shows the results seen after annealing at 500°C. The observed curve deviates here from a straight line and is seen to have a break at about 1 Gy. Two slopes can be identified, $k_1=1.2$ and $k_2=2.3$. Firing at 750°C further increases the sensitivity as seen in curve D. The two slopes can be seen here as well, with values of $k_1=1.2$ and $k_2=2.0$; the break occurs at about the same dose as in curve C. Curves E and F represent the dose dependence curves following annealings at 900 and 1000°C respectively. The break in the log-log scale curves does not appear and the slopes are k=1.6 and 1.5 respectively.

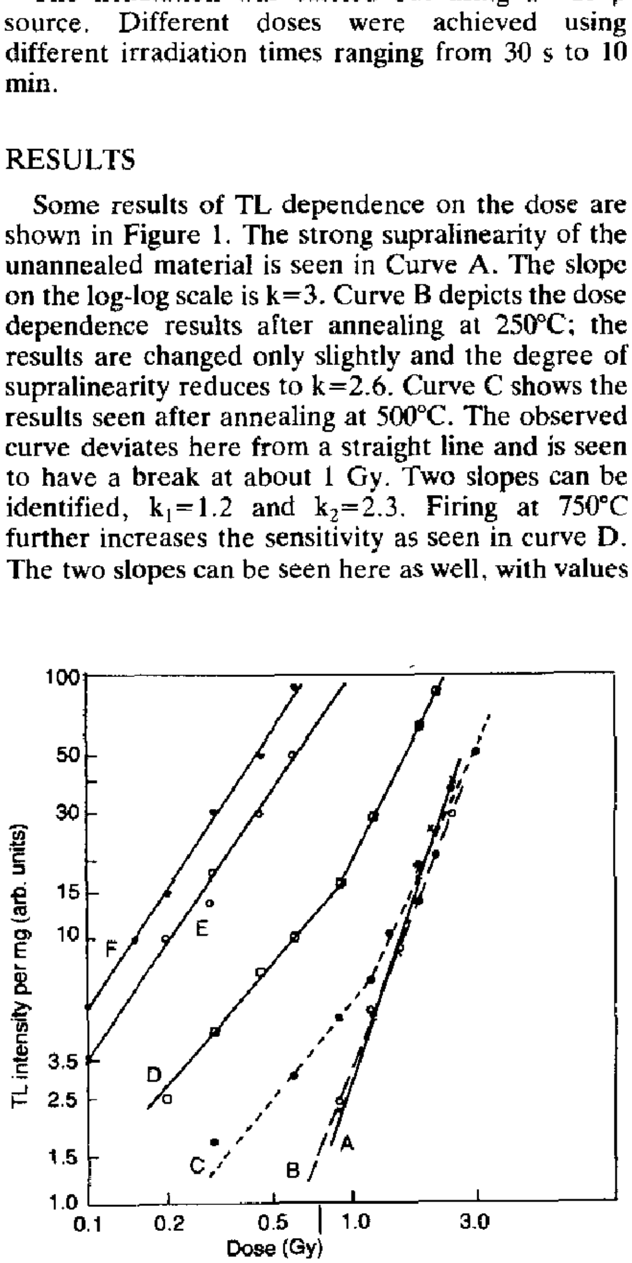

Figure 1. Dose dependence of synthetic quartz powder following different annealing temperatures. (A) Unannealed sample. (B) Annealed at 250°C. (C) Annealed at 500°C. (D) Annealed at 750°C. (E) Annealed at 900°C. (F) Annealed at 1000°C.

Some preliminary measurements have been performed on the dependence of the TL results on the grain size of the synthetic quartz samples. It appears that the sensitivity of the samples reduces with smaller grain size and that the dose dependence curves vary with the grain size. More measurements are needed which may help to unfold the processes involved.

DISCUSSION

In the paper by Chen *et al*[6] which first described the supralinear dependence of the 110°C peak in synthetic quartz, the two kinds of competition leading to supralinearity have been considered. As mentioned in the introduction, these are the competitions during the excitation and heating process. The former is usually characterised by a dependence which is linear at low doses, followed by supralinearity before a second linearity and saturation are approached. Since results at the very low doses may not be seen, this situation may be characterised by an increase in the degree of supralinearity with dose in a certain dose range. This feature appears in curves C and D above. The removal of the competitor in this case is expected to increase the sensitivity and reduce the slope to unity, which indeed occurred in some synthetic quartz samples[6]. In the present work, the slope remained about 1.5 even after an annealing at 1000°C.

The competition during heating is expected to be quadratic or slightly more than quadratic at the very low doses with a decreasing slope at higher doses. In the previous work[6], the latter possibility was favoured since, in most cases, the behaviour was supralinear right from the very beginning. The present results shed some doubt on this conclusion. The fact that after firing to 500°C and 750°C, the initial slope is only slightly above unity (k=1.2) and at higher doses it increases to 2.3 and 2.0 respectively, seems to indicate that competition during excitation plays some role. It appears that until further experimental results are available, the correct model to be used for explaining the supralinearity in synthetic quartz cannot be conclusively determined. One can speculate that since features of both kinds of competition are present, the complex results seen here are due to the existence of both kinds of competitors.

As explained before[6], the occurrence of supralinearity due to the existence of a competitor during the heating phase, is related to the fact that under these circumstances, one has $S \propto m_0 n_0$ where S is the total TL emission and m_0 and n_0 are respectively the initial occupancies of centres and traps. If one of these, say n_0, is more than linearly dependent on the dose due to competition during excitation, the final dose dependence may be more than quadratic. Some support for this idea may be found in curves E and F of Figure 1 where, following annealings to 900 and 1000°C which may very efficiently empty the heating phase competitor, the dose dependence is still supralinear with k being about 1.5. The temperature of annealing that influences the TL properties seems to be higher than previously reported. The main variations of the sensitivity and the dose dependence occur between 600°C and 900°C. It would be interesting to find whether the transitions from alpha to beta quartz (at 573°C) and then to tridymite (at 879°C) are involved in the TL properties described here.

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