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# Effects of ion implantation on the thermoluminescent properties of natural colourless topaz

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**Abstract**

Natural colourless topaz samples were implanted with different fluences of $Cr^+$, $Al^+$ and $Fe^+$ ions. The thermoluminescence (TL) sensitivity to beta and gamma rays were tested and it was found that for $Cr^+$ implantations in the range of $10^{14}$–$10^{16}$ ions/cm$^2$ and for $Al^+$ in the range $10^{14}$–$10^{15}$ ions/cm$^2$, the overall TL emissions of the samples were enhanced. The TL of the $Fe^+$ implanted samples, on the other hand, was reduced after implantation. © 2002 Published by Elsevier Science B.V.

*Keywords:* Topaz; Ion implantation; Thermoluminescence

## 1. Introduction

The thermoluminescence (TL) properties of natural colourless topaz samples from Santo Antônio do Jacinto, Brazil, have been investigated aiming at the development of a new solid state dosimeter [1,2]. The samples have revealed some very interesting features. The TL signal increases with the radiation dose up to 300 Gy [2], the intensity of the TL peaks of pellets prepared using natural topaz are about five times higher than the intensity of the emission of LiF TLD-100 dosimeters, and the peak temperatures are also higher, using the same heating rate [2]. The main emission is centred at about 410–450 nm, showing that the TL can be easily recorded using regular photomultipliers.

Recently, Marques et al. [3] showed that modifications of the optical properties of topaz occur when the samples are implanted with W and Cr. Although the problems induced by the implantation damage, ion implantation seems to be a promising technique to tailor the properties of topaz.

In the present work we are investigating the modification of the TL properties when the natural samples are implanted with chromium, aluminium and iron aiming at the identification of the mechanisms (charge carrier generation by radiation, charge trapping, release and recombination) involved in the emission of light during the TL process.

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## 2. Experimental

Natural colourless samples of topaz, from Santo Antônio do Jacinto, MG, Brazil, were cut from small rolled pebbles. The ions Cr, Fe and Al were separately implanted in the slices using fluences in the range $10^{14}$–$10^{17}$ ions/cm$^2$ and energies of 140 keV. After implantation the samples were thermally treated at 500 °C for 2 h to allow the diffusion of the ions within the bulk. After the thermal treatments the samples were powdered and irradiated with beta ($^{90}Sr + ^{90}Y$ source) and gamma radiation ($^{137}Cs$ source). The TL measurements were performed from room temperature up to 350 °C, following a linear heating program, in a home made TL reader. RBS were performed with a 1.6 MeV $He^+$ beam after implantation and after different stages of thermal annealing. The backscattered particles were detected at 140° and close to 180° using silicon surface barrier detectors with resolution of 13 and 18 keV, respectively. To minimise the effects of charge accumulation on the surface during the analysis, the beam current on the target was kept below 4 nA.

## 3. Results and discussion

The implanted samples are darker than the natural ones and they are also less resistant to thermal treatment fracturing during the treatments. The samples implanted with higher fluences are normally less resistant than the ones implanted with lower fluences.

Fig. 1 shows the RBS aligned and random spectra obtained before and after implantation of $1 \times 10^{15}$ $Cr^+$/cm$^2$ ions. The high yield for the virgin sample reveals the poor quality of the cleaved surface. The implantation turns the entire implanted region (about 120 nm) fully amorphous (the aligned and random spectra overlap on this region). This result shows that topaz is very sensitive to radiation damage produced by heavy particle irradiation. The damaged region does not recrystallise even after annealing at 900 °C for several hours. The formation of a new surface structure associated with the lost of fluorine observed during annealing above 600 °C can be the reason for this stability. Above 600 °C Cr ions start to diffuse into the bulk in agreement with previous studies [3].

The TL peak of samples implanted with $Cr^+$, $Fe^+$ and $Al^+$ and irradiated with gamma rays are shown in Fig. 2. In samples implanted with $Cr^+$ (Fig. 2(a)), the intensity of the TL peak at 240 °C initially increases for the fluences of $10^{14}$ and $10^{15}$ ions/cm$^2$ while for higher fluences, the peak intensity decreases. The highest increase obtained for the $10^{15}$ ions/cm$^2$ is about 57% of the initial TL peak intensity. The TL peaks at 120 and 280 °C

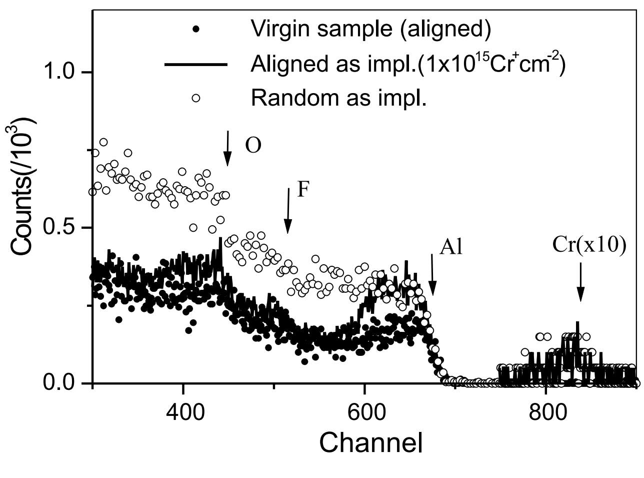


Fig. 1. RBS aligned and random yield obtained before and after implantation of $1 \times 10^{15}$ $Cr^+$/cm$^2$ ions.

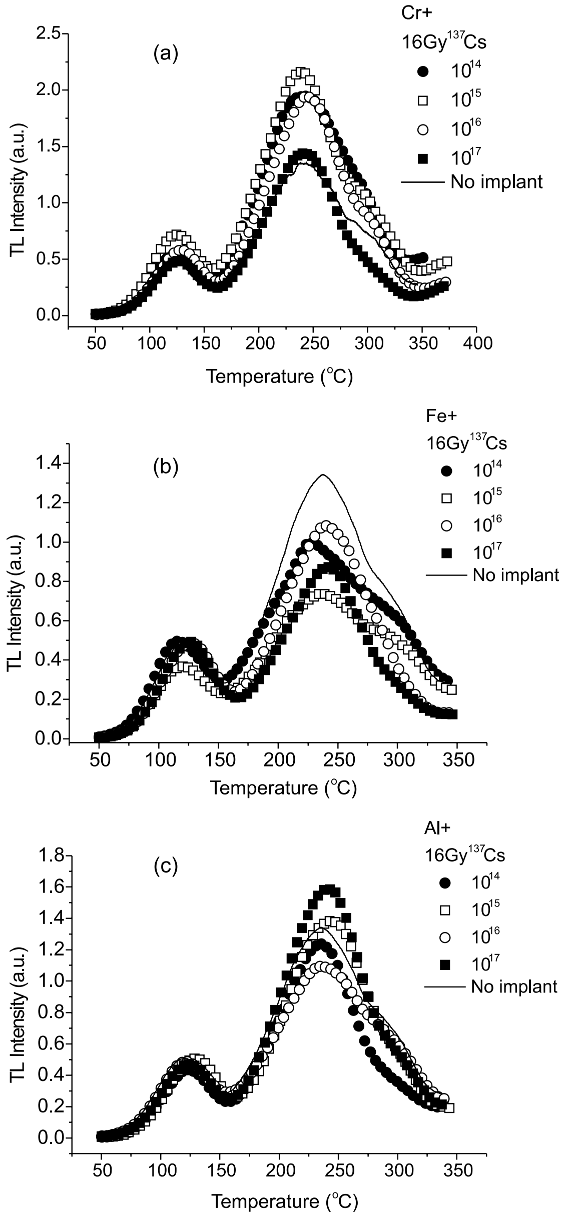


Fig. 2. TL glow curve of implanted topaz samples irradiated with gamma rays ($^{137}$Cs source), as a function of the fluence of $Cr^+$ (a), $Fe^+$ (b) and $Al^+$ (c) implantation.

follow essentially the same behaviour of the 240 °C peak, but while for the latter the intensities of the implanted samples are always higher than the virgin sample, for the formers the TL intensities of the $10^{17}$ ions/cm$^2$ implanted sample are lower than the peak intensities of the non-implanted one. In the samples implanted with $Fe^+$ (Fig. 2(b)), the TL intensity of all three peaks are lower than the non-implanted one for all fluences used.

For the samples implanted with $Al^+$ (Fig. 2(c)) and irradiated with $\gamma$ rays, the intensity of the first TL peak at 120 °C show no appreciable change in the fluence range used during the implantation process. The intensity of the TL peak at 280 °C shows a similar trend but for the lowest fluence ($10^{14}$ ions/cm$^2$) the peak intensity is lower than the non-implanted sample. The intensity of the TL peak at 240 °C, on the other hand, firstly slightly decreases for the fluence of $10^{14}$ ions/cm$^2$, followed by an increase for the fluence of $10^{15}$ ions/cm$^2$. For the fluence of $10^{16}$ ions/cm$^2$ the intensity decreases again, followed by an increase reaching the highest intensity for the fluence of $10^{17}$ ions/cm$^2$. All these changes in the 240 °C peak intensity, however, are within 14% of the intensity of the non-implanted samples.

When the implanted samples are irradiated with beta rays, the behaviour is slightly different (Fig. 3(a)). For all Cr implanted samples, the TL emission is markedly enhanced, as compared to the non-implanted one. The highest increase, of more than 160%, was observed in the 240 °C TL peak for the $10^{16}$ $Cr^+$/cm$^2$ implanted sample. The intensity of the TL peaks at 120 and 280 °C also increase and the highest efficiency is obtained for $10^{16}$ $Cr^+$/cm$^2$ and $10^{14}$ $Cr^+$/cm$^2$, respectively.

The TL response of the Fe implanted sample and irradiated with $\beta$ rays (Fig. 3(b)) is again lower than the non-implanted sample. The peaks at 240 °C and at 280 °C decrease after implantation and the first TL peak at 120 °C firstly decreases for the fluence of $10^{14}$ ions/cm$^2$ following to a slightly increase as the fluence increases.

For the Al implanted samples irradiated with $\beta$ rays (Fig. 3(c)), the highest TL response was observed for the samples implanted with $10^{14}$ ions/cm$^2$, where an enhancement of about 30% is observed for the TL peaks at 120 and 240 °C. The 280 °C TL peak remains unchanged for all implanted samples.

Comparing all the results one can see that the most efficient implantation concerning the increase in the TL signal is obtained when Cr was used,

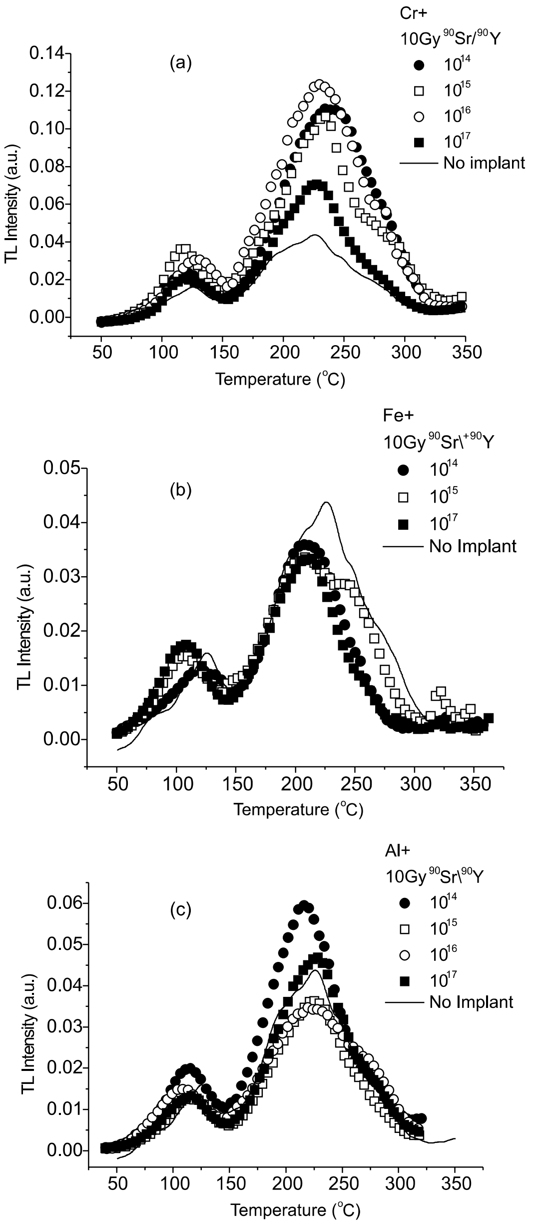


Fig. 3. TL glow curve of implanted topaz samples irradiated with beta rays ($^{90}Sr/^{90}Y$ source), as a function of the fluence of $Cr^{+}$ (a), $Fe^{+}$ (b) and $Al^{+}$ (c) implantation.

where enhancements of about 160% could be attained. Al implantation is less efficient with the highest improvement in the TL response of about 30%. On the other hand, Fe acts mainly as a TL suppressor and, except for the first TL peak at 120 °C, where some slightly enhancement could be obtained, for most of the implanted samples and irradiations used, the overall TL signal decreased, as compared to the non-implanted samples.

The above results can be understood considering that the Cr could act as a dopant that induces colour centres in topaz and thus induces extra defects that can trap charges generated during the irradiation. Marques et al. [3] observed a markedly change in the AO spectra for samples implanted with Cr revealing that extra defects are indeed induced by the Cr implantation in topaz.

The effect of the implantation of Al in the TL response can be explained taking into account two main features: (i) Al is a normal constituent of the crystalline matrix and (ii) the spectra of the emitted light during the TL process is similar to the well-known $[AlO_4]^{\circ}$ defect in quartz and seems also to be also responsible for the TL emission of topaz [4]. This centre act mainly as a hole trapping centre and during the TL measurements, as the main recombination centre in colourless topaz. When the samples were implanted with Al, part of the incident ions can be only displacing the Al already sit at the Si sites thus generating very few extra charge traps. Moreover, considering the fluences used in the present work, the extra amount of Al implanted in the samples represents only a molar fraction of $10^{-9}$–$10^{-6}$ of the already existing Al ions per $cm^2$. This indicates that even if all the implanted ions were generating extra centres, very few defects would be generated during this process. The increases in the TL response observed in some of the implanted samples are probably due mainly to other defects generated along the ion tracks, rather than to the Al ions implanted in the samples.

In both Cr and Al implanted samples, one could see that there are optimum fluences that produces maximum TL output. The optimum fluence is radiation dependent for the Al implanted samples ($10^{17}$ $Al^{+}/cm^2$, for $\gamma$ radiation and $10^{14}$ $Al^{+}/cm^2$, for $\beta$ radiation) and the change induced by the implantation is much lower than the ones observed for the Cr implanted samples. Also, the total light output of the Al implanted samples displays an oscillating behaviour as a function of the fluence. Moreover, comparing Figs. 2(c) and

3(c), one can see that the TL intensity for the $\gamma$ irradiated samples is about 20 to 30 times higher than the TL induced by irradiation for all implanted samples. The $\beta$ radiation only induces charge generation and trapping and, as a consequence, TL light output arising from the surface of the material while $\gamma$ radiation can penetrate deeply in the sample and the corresponding TL output is a bulk effect. This indicates that lower fluences of Al during implantation is more efficient to generate charge trapping defects throughout the sample, while higher fluences of Al seem to induce mainly modifications in the surfaces of the samples.

For the Cr implanted samples, on the other hand, the optimum fluence that produces the maximum TL is approximately radiation independent (in the range $10^{15}$–$10^{16}$ $Cr^+/cm^2$ for both $\gamma$ and $\beta$ radiation). The larger modification on the TL intensity, as compared to the non-implanted sample, was obtained during $\beta$ irradiation, although the TL response to $\beta$ irradiation is much lower than the $\gamma$ ones. These results can be explained considering that Cr ions are heavier than Al and were deposited mainly at the surface of the sample, according to Fig. 1. This indicates that the enhancement of the TL light output when $\gamma$ or $\beta$ rays are used arise mainly from the defects induced in the surface and the layers close to the surface of the material due to the Cr implantation and, as a consequence, no appreciable difference should be noticed when these two radiation types were employed.

This behaviour is consistent with the results of the RBS (Fig. 1) and also the previous observation that after annealing, $F^-$ leaves the damaged region [3]. Considering that the beta irradiation only fills the traps of the surface of the sample, and considering that the TL traps seem to be mainly due to $OH^-$ groups [4], it is expected that the TL response to $\beta$ irradiation of the Cr-implanted samples would markedly increase.

Another important feature observed in the present work, concerning the dosimetric applications, is the dose response of the implanted samples with $10^{15}$ $Cr^+/cm^2$ (Fig. 4). The dose dependence was fitted to the following equation:

$$I = I_0 D^a, \qquad (1)$$

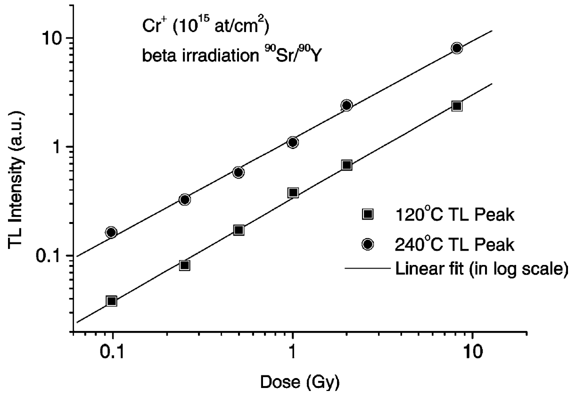


Fig. 4. Dose dependence of the TL intensity of the samples implanted with $10^{15}$ $Cr^+/cm^2$, irradiated with beta rays from a $^{90}Sr/^{90}Y$ source.

where $I$ and $I_0$ indicates the TL peak intensity at a given dose and the initial TL intensity, respectively and $D$ is the dose. The obtained values for the a parameter were $0.92 \pm 0.03$ and $0.96 \pm 0.02$ for the TL peaks at 120 and 240 °C, respectively, indicating that the dose dependence is close to linear.

## 4. Conclusions

We have shown that the TL sensitivity of natural colourless topaz samples to radiation doses can be enhanced with implantation of Cr and Al ions. The best results are obtained when Cr is implanted with a fluence in the range of $10^{14}$–$10^{16}$ ions/cm$^2$. Fe, on the other hand, act as TL suppressor when implanted in topaz. Linear dose dependency was observed for Cr implanted topaz when irradiated with $\beta$ rays.

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