Thermoluminescent Response to Low-Energy Protons

Sung-Yen Lin¹, Tieh-Chi Chu¹, Chu-Chung Hsu², Jao-Perng Lin¹,³
¹Department of Nuclear Science, National Tsing Hua University
²Department of Physics, National Tsing Hua University
³Department of Radiation Oncology, Changhua Christian Hospital

INTRODUCTION
With the increasing importance of the application of proton beam in radiation therapy for cancer (1-3), the study of thermoluminescent dosimeter for proton dose measurement has become increasingly important. Hsu et al. have done researches on the response of TLD-300 to protons (4,5). The proton energies ranged in 0.7-3.0 MeV and 15-30 MeV were studied. In their studies, the glow curve of TLD-300 appeared to be relevant to the proton energy. Among the glow peaks, the ratios of the third peak height to the fifth one were related to $\beta$ energy (6). Since no related study on 100 keV proton with highest LET has been carried out for proton therapy (7), this work was to study the thermoluminescent response of TLD-300 to proton at Bragg peak by means of Cockcroft-Walton linear accelerator accelerating protons in the energy range 30-100 keV.

MATERIAL AND METHODS
A. Vacuum Annealing
CaF$_2$:Tm (TLD-300) made by Harshaw Chemical Company, U. S. A. was used in this experiment to study what effect that vapor would cause the glow curve phenomenon when it was diffusing into the TLD surface layer during the annealing process. The selected 20-piece CaF$_2$:Tm with dimension of 3mm$\times$3mm$\times$0.89mm was divided into two 10-piece groups. Each group was then divided into two 5-piece batches. The cooling rate experiment was first proceeded on the first group to study the effect of slower cooling process on thermoluminescence (TL) of CaF$_2$:Tm in the process of vacuum annealing.

After annealed at 400$^\circ$C for 1.5 h, one of the 2 batches in the first group was released from the oven and cooled down to room temperature in a cold room for about 15 minutes. The other batch was still kept in the oven and cooled down to room temperature for about 90 minutes. It was irradiated with 0.662 MeV $\gamma$ radiation before to observe the glow curves.

Similarly, one of the two batches in the second group was released to a cold room and cooled to room temperature. It was then placed in the vacuum chamber. The other batch was placed in aluminum TLD holder as shown in Fig. 1. The TLD holder was wrapped with nickel-chromium alloy wire for heating and attached with a thermocouple to measure the temperature when annealing.

Figure 1. Proton irradiation experiment layout in chamber. Proton is well selected by bending magnet and directed with three slits. After passing through a thin carbon film, proton beam would spread evenly on the TLD surface (as shown the dash line in acrylic TLD holder). Channeltron detector was used closely behind the holder to detector relative amount of the protons. Al TLD holder was placed on another side which wrapped with nickel-chromium alloy wire for heating and attached with a thermocouple to measure the temperature when annealing.

After being heated at 400$^\circ$C for 1.5 h in the TLD holder, the TLD was cooled in vacuum while it was irradiated with 100 keV $H^+$ (equivalent to two 50 keV $H^+$). Then nitrogen was led to the vacuum chamber and the
TLD material was withdrew for measuring. It took less than 20 minutes to complete this process. It was also to prevent that the TLD would have any contact with the humidity in air. After measuring, the variations of the glow curves were studied.

B. Proton Irradiation Experiment

After annealed and cooled in the cold room, CaF$_2$:Tm (TLD-300) were used to be irradiated with 30–100 keV protons in a vacuum chamber. The protons were provided by Cockcroft-Walton linear accelerator which can accelerate particles in the range of 30–100 keV/nucleon. We directed the proton beam with two slits of 3mm in diameter and the other with 1.5mm in diameter. After passing through a 10 µg/cm$^2$ carbon film with Rutherford scattering, the straight proton beam would spread evenly on the TLD surface, which was placed 20cm behind a film at an angle of 20 degrees. The energy loss of the proton beam on the carbon film can be ignored. We then placed the TLD in an acrylic TLD holder. A channeltron was placed behind the holder to detect the relative amount of the protons. After irradiation, we read the TLDs with the procedure of 50-350°C, 5°C/s, 60 sec by Harshaw 3500 reader and recorded its glow curves. The irradiation was carried out again after changing the proton energy.

Meanwhile, to observe the damage done on the TLD surface, CaF$_2$:Tm were used to be irradiated with 100 keV proton of the same dose, since 100 keV proton has the highest LET. The thermoluminescent output was recorded after reading.

RESULT AND DISCUSSION

A. Vacuum Annealing

![Figure 2. Glow curves of TLD-300s for room (x ) and oven cooling (□)after 0.662 MeV γ-ray irradiation (two curves are almost the same).](image)

![Figure 3. Glow curve deconvolution analysis of vacuum annealed (△) and air annealed (□) TLD-300s after 100 keV H$_2$+ irradiation. Two curves are approximately equaled in P2 (125°C), P3 (150°C) and P5 (260°C)](image)

The glow curves of TLD-300 in both the annealing oven and the cold room are shown in Fig. 2. We found that the cooling rate has no effect on TLD-300 in thermoluminescent response.

The glow curves of TLD-300 in a vacuum chamber and in air after irradiated with 100 $H_2^+$ are reported in Fig. 3, where both annealing methods did not cause any effect on the second, the third peaks and the fifth peak. Therefore, both annealing methods would not cause any effect on the fifth peak. As for the disturbing thermoluminescence, the reason could be that the temperature was not well controlled in vacuum.
B. Proton Irradiation Experiment

The relationship between the thermoluminescent output and the number of irradiation times for TLD-300 after irradiated repeatedly with 100 keV protons are shown in Figure. 4. We found the thermoluminescent output of TLD-300 decreased as the number of irradiation times increased. We then analyzed the Figure 5 illustrate the relationship between the number of irradiation times and the second (125°C), third (150°C) and fifth peak (260°C) heights of TLD-300. The P2 and P3 of TLD-300 increased as the number of the irradiation times increased. The fifth peak (P5) height decreased as the number of the irradiation times increased. The gain of P2 by 48% and P3 by 81%, and the loss of P5 by 26% indicated that P5 appeared to be saturated while P2 and P3 appeared to be sensitized. For the trap at P5 and beyond have higher electron-capture ability, thus the P5 has higher electron-capture ratio than that at P2 and P3. After irradiation and annealing, the deep trap is mostly filled with electrons. Therefore, there are shallow traps to capture electrons. Thus, the shallow traps would have higher probability to capture electrons. The thermoluminescence was then increased. Such saturation of deep traps and sensitization of shallow traps corresponded to the deep trap competition model (8).

![Figure 4](image1.png)

**Figure 4.** The relationship of TL output of TLD-300 after equal dose of 100 keV H\(^+\) irradiation with repeated irradiation times. It shows that TL output increases with increased repeated irradiation times.

Figures 6 showed the relationship between the TL output and protons with different energy ranges for TLD-300. We found that TL output increased as the proton energy decreased. This corresponded to the hypothesis of track structure theory that direct recombination probability of electrons and centers increases with high ionization density caused by high LET particles. As a result, the decrease of the capture probability led to lower TL output (9).

![Figure 5](image2.png)

**Figure 5.** The relationship of peak height of P2 ( ), P3 ( ) and P5 ( ) of TLD-300 with repeated irradiation times. It shows that the gain of P2 by 48% and P3 by 81%, and the loss of P5 by 26% after nine repeated irradiation times than that of TLD-300.
Figure 6. The relationship of TLD-300 TL output per unit dose with different proton energy (30-100 keV). TL output decreased with increased proton energy between 30-100 keV.

CONCLUSION

According to the experiment, the TL output of TLD-300 increased as the LET of proton decreased corresponded to track structure theory. The analysis of the glow curves of TLD-300 proved the correspondence to the hypothesis of deep trap model and the dependence of P2, P3 and P5 on proton energy.

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REFERENCE