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Optimal Annealing of Natural Beryl for Thermoluminescent Dosimetry

Philip S. Moon

Korea Atomic Energy Research Institute, Seoul, Korea

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Abstract

The annealing of natural beryl powder used for the thermoluminescent dosimetry was investigated in order to eliminate effect of previous exposure. Through the glow curve analysis, the optimal annealing treatment was found to be one hour annealing at 1450°C, but annealing for one hour at 550°C was sufficient for reuse of natural beryl powder. There are two distinguished glow peaks at 65°C and 200°C of heating temperature during readout process. The 65°C glow peak fades away rapidly, but the 200°C glow peak remains very stable. It is, therefore, quite feasible to use the 200°C glow peak for thermoluminescent dosimetry.

요 약

천연 녹주석의 방사선조사에 의한 효과를 없애기 위한 열처리방법을 연구하였다. Glow 곡선분석에서 최적열처리시간과 온도는 1450°C에서 1시간이었고, 녹주석을 다시 사용할 시에는 1450°C에서 1시간동안 열처리를 다시할 필요는 없고 550°C에서 1시간만 열처리를 하면 된다. 1450°C에서 열처리된 녹주석시료를 ^{60}Co 감마선으로 100R 조사한 후 glow 곡선을 그려보니 65°C와 200°C에서 glow peaks를 발견하였다. 65°C glow peak는 곧 없어졌으나 200°C glow peak는 매우 안정하였다. 따라서 열형광방사선측정에는 200°C glow peak를 이용하여 ^{60}Co 감마선을 측정할 수 있다.

1. Introduction

The thermoluminescent dosimetry has been studied by many investigators during the past ten years[1-8]. When ionizing radiation is absorbed in matter, most of the absorbed energy ultimately goes into heat while a small fraction is dissipated to break chemical bonds. In some materials, such as phosphors, a very minute fraction of the energy is stored in metastable energy states. Some of this energy can be recovered later as visible photons if the material is heated. The phenomenon of the visible photons released by thermal means is the thermoluminescence[2,3,4]. Some of the phosphors have very good dosimetric characteristics and now widely used for radiation monitoring and dosimetry. In the thermoluminescent dosimetry, the effects upon the glow curve of thermal annealings are quite important. Therefore, a study has been made of the characteristics of the natural beryl glow curve as a function of annealing time and temperature, both before and after ^{60}Co irradiation. The natural beryl contains about 20 different mineral constituents and it is quite different from other phosphors used in ordinary thermoluminescent dosimetry. There are no reports concerning the annealing and thermoluminescent properties of natural beryl. I have found that the natural beryl has desirable thermoluminescent properties among which are high radiation sensitivity and good stability.

2. Experimental

The material used in this experiment is a natural beryl produced in Chunyang-Gun, Chungchungbuk-Do, Korea, and it was ground and sifted through U.S. Standard sieve in order to make 80-200 mesh powder sample. The color of natural beryl is light green and it has the following composition. (Table 1)

The samples of natural beryl powder were initially annealed for 30 min., 1 hour, 2 hours, 3 hours and 4 hours at 500°C in order to determine the optimal annealing time.

The natural beryl powder samples were put into a porcelain crucible or a silica tube and placed in a furnace for annealing. The samples were annealed for one hour at various degrees of temperature, such as 100°C, 200°C, 300°C, 400°C, 500°C, 600°C, 700°C, 1200°C, 1400°C and 1450°C in order to eliminate the effects of previous exposure.

For the annealing of sample, Thermolyne furnace 1400 was used for heating up to 1200°C and Lindberg silicon combustion furnace was used for heating at 1400°C, 1450°C.

After the annealing, the sample was irradiated to 100R with ^{60}Co gamma-ray. The thermoluminescence was measured 10 minutes after gamma-ray irradiation to allow all the samples to decay for the same length of time.

The thermoluminescent glow curve of each sample was measured and recorded with TLD 7100 and associated dual-channel recorder of Teledyne Isotope Co. The readout system can register the integrated light yield as well as record the glow curve and heating rate curve. The natural beryl powder sample of 50mg was placed on a high-resistivity nichrome heating element, which was connected to the Be-Cu alloy electrodes, in order to get the integrated light yield and glow curve at a heating rate of 10°C/sec upto 280°C. The photomultiplier tube was VMP 11′44K of 20th Century Co., and the sample and PM tube was placed in a light tight compartment for thermoluminescent measurement.

For the reuse of natural beryl sample, the optimal pre-annealing schedule was investi-

Table 1. Chemical Analysis of Natural Beryl*

(weight in %)

SiO_2	Fe_2O_3	Al_2O_3	BeO	Bi	Mg	Sn	C	S	P	
82.5	2.19	2.48	9.46	0.10	0.18	0.001	0.065	0.078	0.003	
Pb	Mn	Mo	Cu	Na	Zn	Ag	Ti	Ca	Cr	Ig. loss
0.003	0.033	0.031	0.020	0.14	0.033	<0.0001	0.08	0.40	0.009	0.97

* Analysis was done by KIST

gated with the sample irradiated with 100R ^{60}Co gamma-ray.

Instead of weighing each sample, the powder sample dispenser was used together with the automatic vibrator. When the dispenser is clean and the powder sample dry and running free, the standard deviation of the weight of a series of aliquots are less than 0.5%. This dispenser is designed to be used with 80-200 mesh powder sample. The much finer powder does not dispense readily.

The glow curve of each sample was taken for analysis of thermoluminescent response. Also the fading of glow peak was examined in order to select the stable glow peak on a series of glow curves.

The natural beryl sample was cooled rapidly after annealing and stored at room temperature in the dark.

3. Result and Discussion

Figure 1 shows the thermoluminescent response of natural beryl sample versus heating time at 500°C. The optimal pre-annealing time for natural beryl is one hour within a 5% confidence limit.

The control and 100°C-300°C annealed samples were not easy to measure the thermoluminescent response, because there were too much light emission left after annealing and the samples were still not completely annealed out. No thermoluminescent response was found in the samples annealed for one hour at 400°C, 500°C, 600°C, 700°C, 1200°C, 1400°C and 1450°C. Each of these samples was irradiated by ^{60}Co gamma-ray to 100R prior to the glow curve recording.

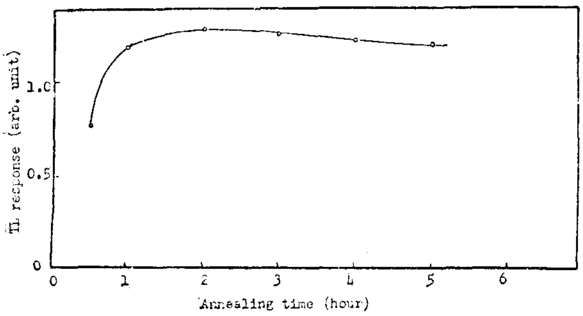

Fig. 1. The TL response of natural beryl vs. annealing time at 500°C

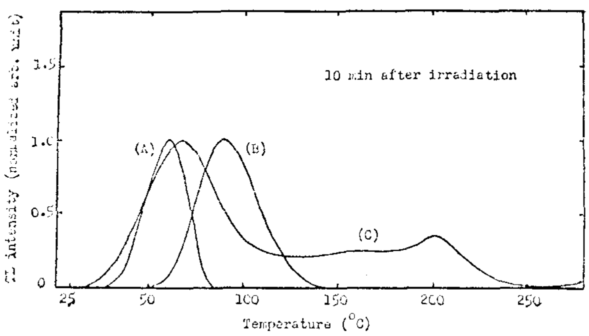

Fig. 2. Glow curves of natural beryl annealed at various temperatures. normalized to the same maximum emission of luminescence. (A) Annealed at 400-700°C for 1hr. (B) Annealed at 1,200°C for 1hr. (C) Annealed at 1,450°C for 1hr. Heating rate: 10°C/sec Dose: 100R

Figure 2 shows the glow curves of natural beryl annealed at various temperatures for one hour and irradiated to 100R by ^{60}Co gamma-ray, and the glow peak was normalized to the same maximum emission of thermoluminescence.

In Fig. 2, curve (A) shows the glow curve of sample annealed for one hour at 400°C-700°C and irradiated with 100R of ^{60}Co gamma-ray. Curve (B) shows the glow curve of sample annealed for one hour at 1200°C and irradiated 100R of ^{60}Co gamma-ray. Curve (C) shows the glow curve of sample annealed for one hour at 1450°C and irradiated with 100R of ^{60}Co gamma-ray. As one may see in Fig. 2, the glow curve (B) is shifted to higher temperature region than glow curve (A): the glow peak was at 60°C for (A) and 90°C for (B). Glow curve (C) shows two distinguished glow peaks at 65°C and 200°C.

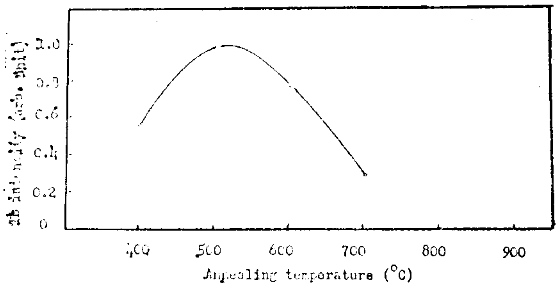

Fig. 3. The TL light sum *vs.* annealing temperature. Exposure dose: 100R

Figure 3 shows the thermoluminescent light yield of natural beryl versus annealing temperature after 100R ^{60}Co gamma-ray irradiation.

The maximum thermoluminescent light sum was found at the temperature of around 550°C, and consequently 550°C was chosen as a re-annealing temperature for repeated use in this experiment. Accordingly the samples annealed at 1200°C, 1450°C and stored in room temperature were annealed again at 550°C in order to eliminate the effect of exposure other than ^{60}Co gamma-ray irradiation.

Figure 4 shows the fading pattern of natural beryl thermoluminescent light sum on storage at room temperature after annealing and 100R ^{60}Co gamma-ray irradiation. In this figure, curve (A) shows the fading of thermoluminescent light sum of sample annealed for one hour at 400°C and it faded away rapidly with half-life of 45 min., which shows that it is not suitable for thermoluminescent dosimetry. Curve (B) shows the fading of thermoluminescent lighht sum of sample annealed for one hour at 600°C. It also faded away with half-life of 120 min., and it is also not suitable for thermoluminescent dosimetry. Curve (D) shows the fading of thermoluminescent light sum of sample annealed for one hour at 1450°C and heated for one hour at 550°C. It faded very

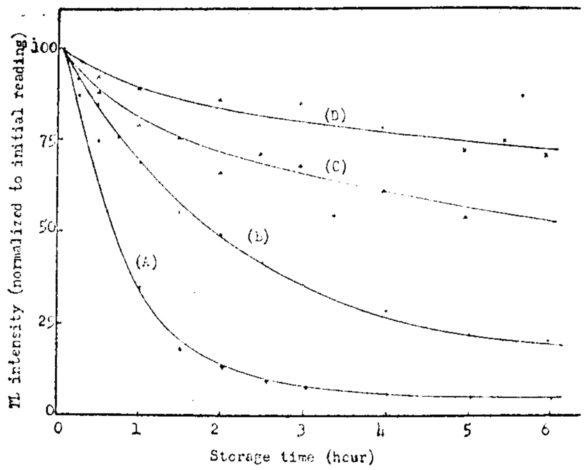

Fig. 4. Fading of natural beryl TL light sum on storage at room temperature. (A) Annealed at 400°C for 1 hr. (A) Annealed at 600°C for 1hr. (C) Annealed at. 1,200°C for 1 hr. (D) Annealed at 1,450°C for 1 hr. Exposure dose: 100R

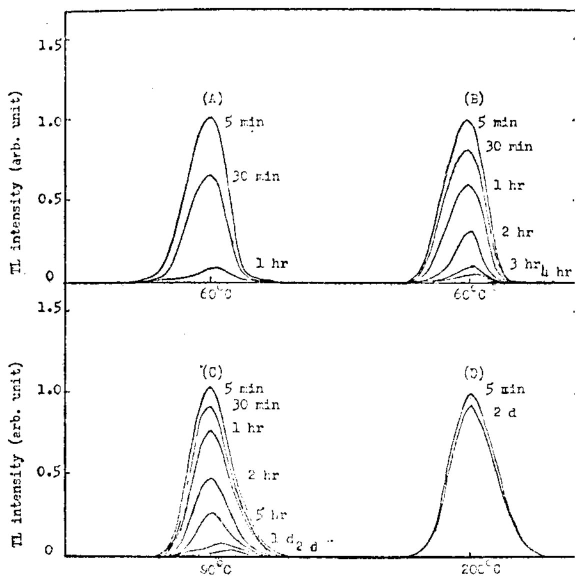

Fig. 5. Fading of natural beryl glow curves stored at room temperature in the dark room. Exposure dose: 100R

(A) Annealed at 400°C, heating rate: 10°C/sec

(B) Annealed at 600°C, heating rate: 10°C/sec

(C) Annealed at 1,200°C, heating rate: 10°C/sec

(D) Annealed at 1,450°C, heating rate: most rapid

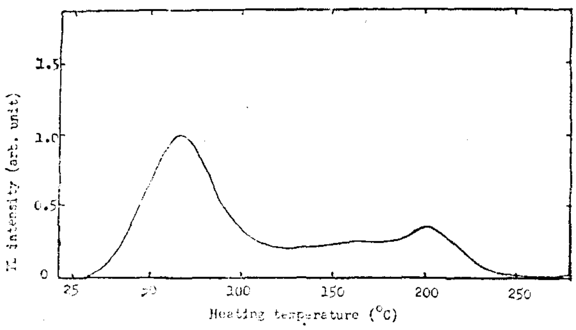

Fig. 6. The TL glow curve of natural beryl 10 min. after gamma-ray irradiation. Exposure dose: 100R Heating rate: 10°C/sec

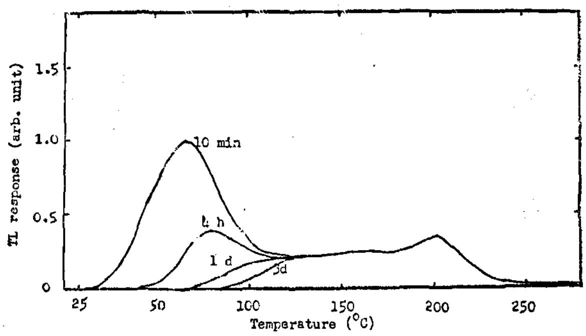

Fig. 7. Fading of natural beryl glow curves stored at room temperature in the dark. Dose: 100R Heating rate: 10°C/sec

slowly.

Figure 5 shows the fading of glow curves of natural beryl stored at room temperature in the dark after annealing and 100R ^{60}Co gamma-ray irradiation. Glow curve (A) of sample annealed at 400°C faded quickly. Glow curve (B) of sample annealed at 600°C faded rather quickly. Glow curve (C) of sample annealed at 1200°C and heated for one hour at 550°C faded slowly, but in two days the glow curve disappeared. Glow curve (D) of sample annealed at 1450°C and heated for one hour at 550°C remained very stable.

As one may see in Fig. 5, it is apparent that the natural beryl annealed at 1450°C shows some thermoluminescent dosimetric characteristics.

Figure 6 shows a typical glow curve of natural beryl an nealed for one hour at 1450°C and then heated for one hour at 550°C and irradiated by ^{60}Co gamma-ray with 100R. The glow curve was taken after 10 min. of ^{60}Co gamma-ray irradiation and the heating rate of readout system was 10°C/sec. As one may see, there are two distinguished glow peaks at 65°C and 200°C. The sample annealed at 1400°C for one hour did not show the stable glow peak at 200°C

Figure 7 shows the fading of glow curve of natural beryl annealed for one hour at 1450°C and then heated for one hour at 550°C and irradiated by ^{60}Co gamm-ray with 100R. The 65°C glow peak faded away completely after three days, but 200°C glow peak remained stable and this 200°C glow peak seemed most suitable for thermoluminescent dosimetry.

4. Conclusion

The optimal annealing temperature for natural beryl is 1450°C and one hour of annealing time is necessary to erase all the effects induced by previous exposure. It is not necessary to anneal the natural beryl sample for one hour at 1450°C, if it has previously annealed at that temperature. For reuse of natural beryl sample, one hour annealing at 550°C is sufficient to eliminate the previous gamma-ray exposure. The glow peak at 200°C of natural beryl annealed at 1450°C is quite suitable to use for thermoluminescent dosimetry of gamma-ray.

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