Anomalous behaviour of thermoluminescence from quartz: A case of glow peaks from a Nigerian quartz

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Abstract

Anomalous behaviour displayed by a thermoluminescence (TL) glow peak as radiation dose increases in a Nigerian quartz is presented. The glow curves of the phosphor have four clear glow peaks. The peak temperatures of the first three glow peaks were found to be constant with change in radiation dose at 82, 148 and 200ºC for the sample readout at a heating rate of 1 ºCs^{-1}. The peak temperature of the fourth peak, which is at around 320ºC for a sample irradiated to a dose of 63 Gy and heated at 1 ºCs^{-1}, displays anomalous behaviour with increase in dose relative to the first three peaks. The temperature at which this peak occurs increases with dose to about 335ºC for 177 Gy and then decreases thereafter as dose is further increased. The change is explained on the assumption that the peak may be complex consisting of several overlapping first-order glow peaks each with different TL behaviour.

1. Introduction

Quartz is abundant in nature and can be found in archaeological artefacts as well as geological materials (Sawakuchi and Okuno, 2004 and Yazici and Topaksu, 2003). The interest in quartz for dating and retrospective dosimetry applications (Chen and Mckeever, 1997, Pagonis et al., 2002 and Woda et al., 2002) has led to many studies on its thermoluminescence (TL) characteristics (Adamiec, 2005, Spooner and Franklin, 2002 and Franklin et al., 2000), effects of thermal treatments on these characteristics (Adamiec, 2005, de Lima et al., 2002 and Nakagawa and Hashimoto, 2003) and on kinetic analysis (Yazici and Topaksu, 2003 and Spooner and Questiaux, 2000). These studies revealed that quartz exhibits a number of TL peaks when heated from room temperature to 500ºC after irradiation. For instance, glow peaks around 100,180,220,260,305,350 and 450ºC have been reported for an Australian sedimentary quartz at a heating rate of 5 ºCs^{-1} (Franklin et al., 1995). Another example is that of an Arkansas quartz where peaks were seen at 97, 147 and 197ºC for readout at 4 ºCs^{-1} (Kitis et al., 2003). Various other TL glow-peak positions have been reported for quartz of different origin including Brazilian, Norwegian, Sea sand, as well as synthetic quartz (Pagonis et al., 2002 and de Lima et al., 2002).

Studies on the influence of excitation dose and thermal treatment on the TL properties of glow peaks in quartz have been given considerable attention. de Lima et al. (2002) and Yang and Mckeever (1990) have shown that pre-irradiation annealing produces profound changes in TL sensitivities of the peaks. Various other studies on quartz, including that of Yazici and Topaksu (2003) on a synthetic quartz and Kitis et al. (2003) on an Arkansas quartz have also shown that positions of the glow peaks in quartz either remain constant or shift to lower temperature as excitation dose given to the sample is increased.

In this work, an anomalous shift of a TL glow peak in a Nigerian quartz relative to other peaks from the same sample, first increasing and later decreasing as irradiation dose increases, is reported. The dose-dependent TL characteristics of this glow peak and the other glow peaks in the quartz are reported.

2. Materials and methods

2.1. Sample preparation

Samples used in this work consisted of natural quartz from Akure, Ondo state, Nigeria ground to grains of size less than 90µm. Before use, the samples were annealed at 900ºC for 10 min (Gallenkamp Muffle Furnace) and cooled rapidly in air.

TL measurements were carried out at the Centre for Scientific and Industrial Research in Pretoria, South Africa, using a RISO luminescence reader (model TL/OSL-DA-15) at various heating rates between 30 and 500ºC. The signal was detected by a bialkali photomultiplier tube (Thorn-EMI 9235 QB) through a Schott BG-39 filter (transmission band 340–620 nm FWHM). All the measurements were made in a nitrogen atmosphere. For each sample, two measurements were made one after the other and each from 30ºC. In each case, the second measurement is subtracted from the first in order to correct for the effect of blackbody radiation from the sample holder.

For each of the measurements reported in this work about 10 mg of the sample was thinly spread on an aluminium disc and held in place by a thin layer of silicon grease (Silicon spray, Den Braven Sealants).

In order to investigate the dose response of the quartz, sets of 16 samples each were irradiated to doses in the range 0.88–1057 Gy. The irradiation, at 10.573 Gy/min, was done using the in-built $^{90}\mathrm{Sr}\,\beta$-source in the RISO luminescence reader.

To estimate the linearity of the dose response, the linearity index $f(D)$ proposed by Chen and Mckeever (1994) was used:

$$f(D) = \left[\frac{S(D) - S_0}{D}\right]\left[\frac{S(D_1) - S_0}{D_1}\right]^{-1}, \tag{1}$$

where in a graph of TL response (i.e. intensity) against dose, S_0 is the intercept on the TL intensity axis as extrapolated from the linear region of the graph. $S(D)$ is the sample TL response corresponding to dose D, and D_1 is a dose in the linear region. $S(D_1)$ is the sample TL response corresponding to dose D_1. By this definition $f(D)>1$, $f(D)=1$, and $f(D)<1$ respectively imply supralinearity, linearity and sublinearity.

3. Results and discussion

3.1. TL glow curves

Fig. 1 shows a TL glow curve of the quartz irradiated to a dose of about 10.57 Gy and readout at a heating rate of 1 $^{\circ}\mathrm{Cs}^{-1}$. The glow curve shows four clear peaks around 82, 148, 200 and 303ºC, which henceforth are referred to as peak I, II, III and IV, respectively. This shows that there are at least four different charge traps in this phosphor. The peak temperature of peak I shifts to 101º when the heating rate is increased to 2 $^{\circ}\mathrm{Cs}^{-1}$ This compares well with the values from Arkansas quartz (106.2 ± 5.5º C), BDH quartz (102.4 ± 5.5ºC) , Norwegian quartz (104.8 ± 2.6º) and SOT quartz (102.3 ± 5.1ºC) all of which were also obtained at 2 $^{\circ}\mathrm{Cs}^{-1}$ (de Lima et al., 2002). Similarly, Yazici and Topaksu (2003) reported a glow peak at 96º for a synthetic quartz readout at a heating rate of 1 $^{\circ}\mathrm{Cs}^{-1}$as for peak I in this work. Glow curves of different quartz (Arkansas, Norwegian and Sea sand) obtained at a heating rate of 2 $^{\circ}\mathrm{Cs}^{-1}$ (Pagonis et al., 2003), showed glow peaks around 170, 220 and 320ºC. These glow peaks thus occur at positions similar to those of peak II (170ºC), peak III (226ºC) and peak IV (324ºC) obtained at a heating rate of 2 ºCs^{-1}with our sample. The glow peaks around 193 and 296ºC reported for synthetic quartz at a heating rate of 1 ºCs^{-1} by Yazici and Topaksu (2003) are also close to 200 and 303ºC obtained in the Nigerian quartz at the same heating rate. In general, the glow curve structure of the Nigerian quartz closely resembles those of BDH, Norwegian, and SOT quartz as presented by Adamiec (2005).

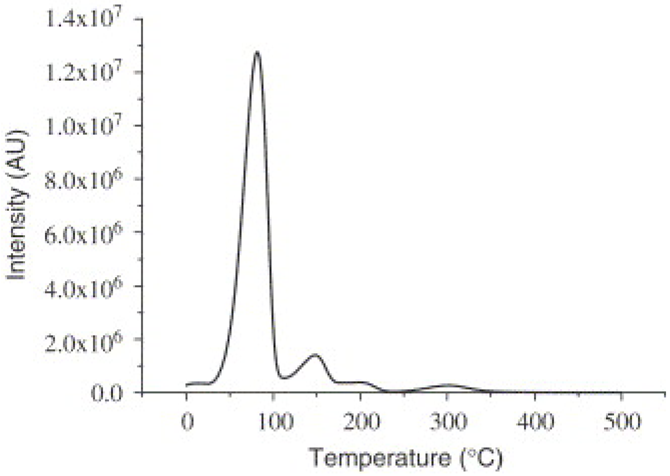

Fig. 1. TL glow curve of the quartz irradiated to a dose of 10.57 Gy and readout a heating rate of 1 ºCs^{-1}.

The dose response (maximum intensity) of peak I in the Nigerian quartz was relatively high compared to that of the other peaks and thus suppresses the intensities of the other peaks. In comparison, the glow peaks of synthetic quartz reported by Yazici and Topaksu (2003) and of Arkansas quartz reported by Kitis et al. (2003) as well as of Norwegian, BDH, SOT and Arkansas quartz reported by Adamiec (2005) and of Brazillian quartz reported by de Lima et al. (2002) show similar intensity characteristics. The ratio of maximum intensity of peak I to each of the other peaks in Arkansas, BDH, Norwegian and SOT quartz were compared with that of peak I in the Nigerian sample of this study. The comparisons show that the dose response of the Norwegian sample closely matches that of the Nigerian quartz. Except for the Nigerian sample, the ratios were determined by reading off the maximum intensities of the peaks from the glow curves published by Adamiec (2005). In the latter, the 360 and 550 nm TL emissions were detected separately whereas in our work the filter used (Schott BG-39) allows both emissions to be detected simultaneously by the TL reader. The maximum intensities in the work of Adamiec (2005) were therefore deduced to be the sum of maximum intensities for 360 and 550 nm TL emissions. The ratios are presented in Table 1. The comparisons show that the TL properties of the Nigerian quartz are consistent with the published properties of the selected quartz.

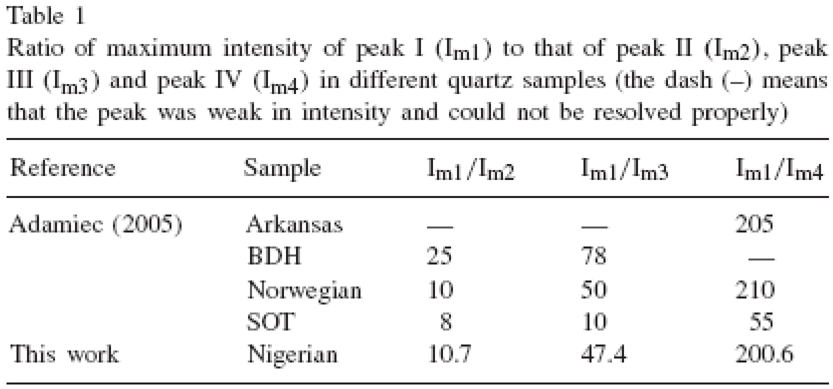

Table 1
Ratio of maximum intensity of peak I (I_{m1}) to that of peak II (I_{m2}), peak III (I_{m3}) and peak IV (I_{m4}) in different quartz samples (the dash (–) means that the peak was weak in intensity and could not be resolved properly)

Reference	Sample	I_{m1}/I_{m2}	I_{m1}/I_{m3}	I_{m1}/I_{m4}
Adamiec (2005)	Arkansas	—	—	205
	BDH	25	78	—
	Norwegian	10	50	210
	SOT	8	10	55
This work	Nigerian	10.7	47.4	200.6

3.2. Shift of peak temperature with dose

The glow curves obtained at three different doses and presented in Fig. 2 show the effect of increasing excitation dose on the peak positions for peaks I–IV. The glow curves were obtained using a heating rate of 1 ºCs^{-1}. The peak temperatures of each of peak I–III were found to be essentially constant at 82.0±0.0, 148.0±0.4 and 200 ± 0.8ºC, respectively, for all doses in the range 0.88–1057 Gy. On the other hand, the peak temperature of glow peak IV was found to shift with dose. The peak temperature increased with dose, reached a maximum at about 335ºC for a dose of 177 Gy and decreased thereafter. The detailed dose-dependent shift in position for the peak is shown in Fig. 3. This kind of shift in glow peak position has not been reported for quartz before. However, behaviour similar to this has been reported by Rodine and Land (1971) for some glow peaks in ThO_2. Later numerical work by Kristianpoller et al. (1974) showed that in ThO_2 this kind of peak temperature shift with dose is possible. In arriving at this conclusion they assumed the presence of a second trap (competitor trap) competing with the recombination centre. The competitor trap is such that electrons are not released from it during heating in the temperature range of the luminescence trap being considered. Other assumptions made include the following: (1) trapping in the competitor is much faster than both recombination in the recombination centre and retrapping into the luminescence trap (2) the retrapping probability of the luminescence trap is very small compared to the rate of release of carriers. Li et al. (2004) have also reported a shift in peak position of a glow peak to higher temperature with increase in dose in BaB_4O_7:Dy phosphor but did not give a detailed explanation for this anomalous behaviour, only basing it on assumption of second-order kinetics.

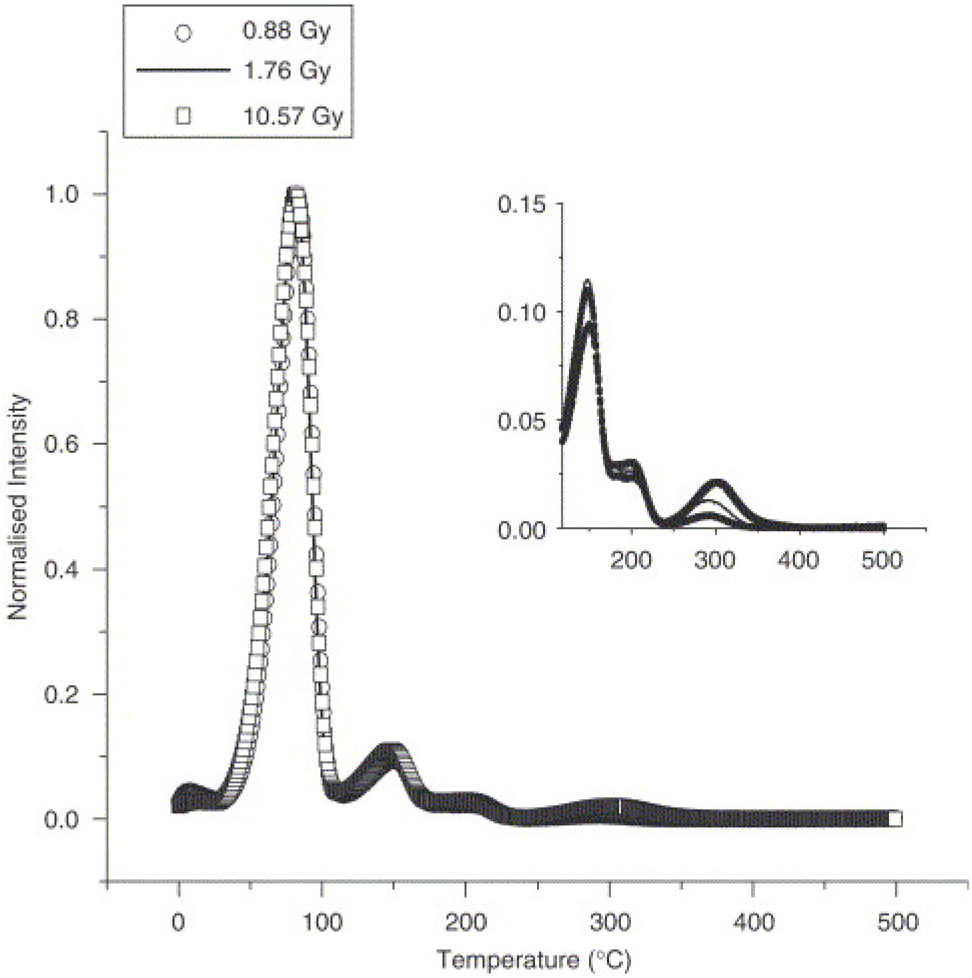

Fig. 2. TL glow curves of the quartz at three different doses all measured at 1 °C/s.

The stability of the peak positions of peaks I–III with change in dose is consistent with what has been reported for glow peaks in both synthetic quartz (Yazici and Topaksu, 2003) and natural quartz (Nakagawa and Hashimoto, 2003). That the peak is constant with dose indicates that peaks I–III can be described by first-order kinetics. From their previous studies on quartz, Durrani et al. (1977) proposed an hypothesis that radiation affects each of the traps in quartz differently. The work of Sawakuchi and Okuno (2004) on Brazillian quartz also indicated this possibility. The behaviour of peak IV, which is different from that of the other three peaks in the present work, is a further support for this hypothesis.

In the present work we offer an explanation for the observed anomalous shift in the peak temperature of peak IV, that is, the characteristic of the glow peak going through a maximum as dose is increased. We suppose that although the peak appears as one, it may actually be a complex one, consisting of multiple first-order glow peaks each with different TL dose behaviour. Thus, if it is assumed that the higher-temperature components of the complex peak are lower in intensity at small doses but grow faster as the dose increases than the lower-temperature components, then the net result will be a shift toward higher temperature. The later decrease in the peak temperature at very high doses can be explained if it is further assumed that the higher-temperature components saturate faster than the lower-temperature components. Quantitatively this means the lower-temperature components begin to grow faster in intensity than the higher-temperature components from the dose where saturation begins, hence the observed decrease in peak temperature at high doses. This argument is supported by the fact that the peak temperature begins to decrease from around 200 Gy where saturation begins for this peak (see Fig. 3, Fig. 4 and Fig. 5). A similar argument has been used by Kitis et al. (2005) to explain a shift to lower temperature of the peak position of the high-temperature glow peak in a Paprika sample with increase in dose.

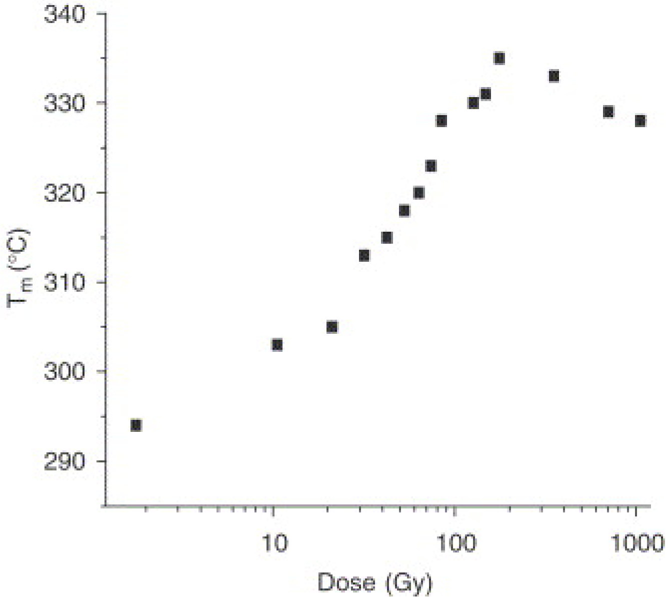

Fig. 3. Variation of peak temperature with dose for glow peak IV. All measurements were made at 1 °C/s.

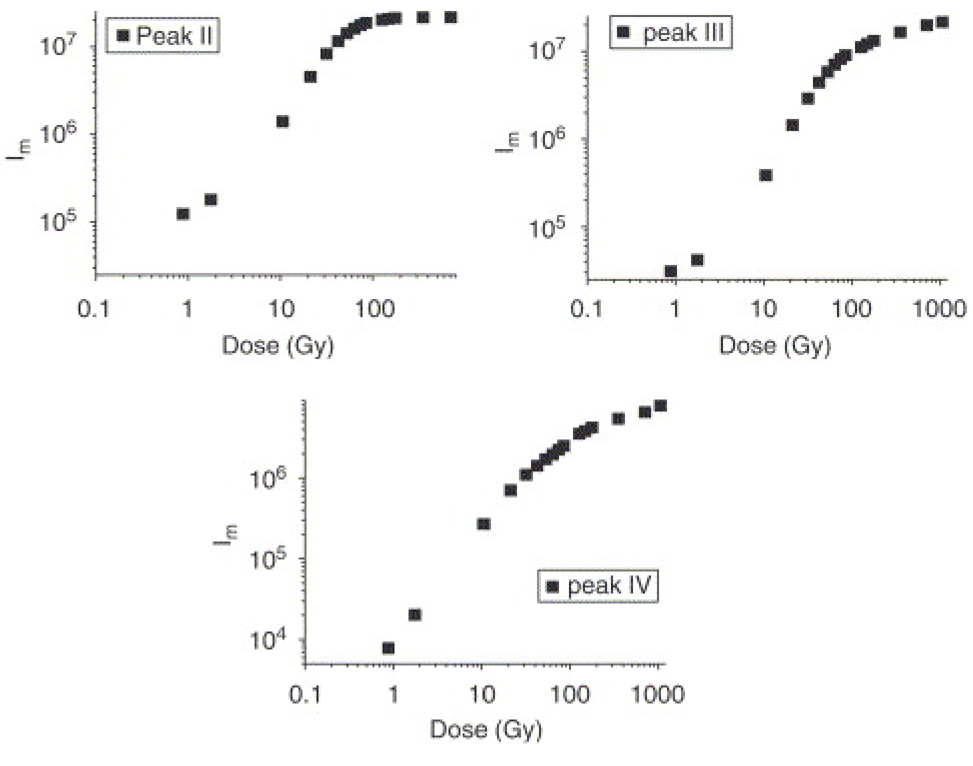

Fig. 4. Plot of maximum peak intensity against dose for peaks II–IV.

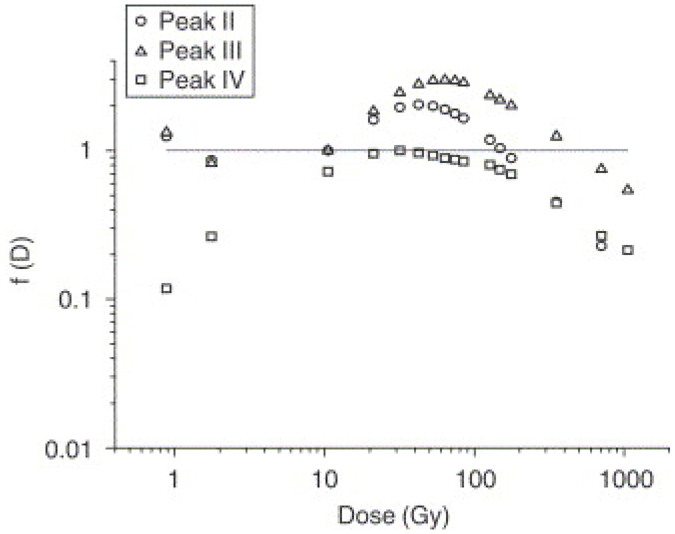

Fig. 5. Plot of the linearity index function *f*(*D*) against dose using (a) total intensity and (b) maximum TL intensity as the response of the sample.

3.3. Dose response

Fig. 4 shows the variation of maximum TL intensity with dose for peaks II–IV with dose. Details for peak I have not been reported because the response for the peak exceeds the detection limit of the TL reader at doses greater than 10.57 Gy. The maximum TL intensity for each of the peaks II–IV varies in the same manner with dose. The dose dependence of the maximum TL intensity of all the peaks is assumed to obey the relation

$$S(D)=\alpha D^{\delta} \qquad (2)$$

as given in Chen and Mckeever (1997) where $S(D)$ stands for maximum TL intensity, D is the excitation dose, α and δ are fitting parameters. The linear region of the dose response for each of the peaks was located by fitting Eq. (2) to the curves shown in Fig. 4. The linear region was taken to be one where δ is closest to 1. In this way, the linear region for each of peaks II and III was determined to be from 0.88 to 10.57 Gy with δ=1.009 for peak II and δ=1.060 for peak III. For peak IV, the linear region is between 21 and 53 Gy with δ=1.003. Also for peak IV, δ was obtained to be 1.427 by fitting the responses at doses below 21 Gy and 0.500 by fitting the responses above 53 Gy to Eq. (2). The plots in Fig. 4 suggest the presence of high supralinear dose response for the peaks in the present quartz. It is notable that Charitidis et al., 1999 and Charitidis et al., 2000 have also reported supralinearity in the dose response for the ‘110ºC’ glow peak in synthetic quartz. A further analysis of the dose response behaviour of the peaks was carried out using the linearity index function in Eq. (1). The results of the analysis are presented in Fig. 5. From the figure, three regions can be identified for each of the peaks. For all the peaks, region 1 is up to a dose of about 11 Gy. In this region responses of peaks II and III are linear whereas that of peak IV is sublinear. Region 2 is from 11 Gy up to about 127 Gy for peak II; up to about 352 Gy for peak III and up to about 53 Gy for peak IV. In the second region, peaks II and III are supralinear while peak IV is linear. In region 3, the response of each of the three peaks is sublinear.

4. Conclusion

The TL properties of a Nigerian quartz have been investigated. An anomalous shift in the peak position of peak IV in the quartz with change in dose was observed. The position of the peak increases with dose to a maximum at about 335ºC for 177 Gy and then decrease when the dose is further increased. This has been explained by assuming that peak IV is a composite peak consisting of multiple overlapping peaks. Linearity index analysis showed that the responses of peaks II and III are linear at low doses, grow supralinear as dose is increased and then become sublinear at high doses. In contrast, the response of peak IV is sublinear at low doses, followed by a linear region with further increase in dose, then sublinearity at high doses.

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