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Analysis of trap spectroscopy of Ba_2SO_4 in simplified OTOR/GOT equation

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Abstract:

Glow curves of $BaSO_4$ a highly sensitive thermoluminescence (TL) dosimetric material has been analyzed using the recently formulated simplified General One Trap (GOT) equation to determine the trapping parameters. The new simplified GOT equation helps in finding the key trapping parameters E, s, N, γ and α which was not possible with the well known kinetic order formalism. From the analysis it was found that activation energy of the phosphor are in the range 0.66 – 1.15eV and frequency factor ~ 10^8 - $10^{10}s^{-1}$ with lifetime of the decoded five peaks ranging from a few minutes to as high as 39 years depending upon the doses.

Key words: $BaSO_4$, Trapping parameters, activation energy and frequency factor.

1. Introduction

Thermoluminescence (TL) is a temperature stimulated light emission from a solid/amorphous material after removal of excitation, and is probably one of the direct evidences of existence of trapping levels [1]. A complete TL glow curve of a thermoluminescent material may be approximated by a linear combination of glow peak intensities [2]. A glow curve essentially represents the spectroscopy of traps of a solid in

a coded form. Analysis of glow curves is nothing but a sort of decoding the coded glow pattern. Excellent texts as well as a large number of papers have appeared in literature to deal this challenging area [3-7]. Several methods have been developed in decoding the trapping parameters of the glow peaks by many workers [8-10]. With the easy availability of computerized data acquisition and management system, Computerized Glow Curve Deconvolution (CGCD) has been a quite popular to decode a glow curve in the framework of kinetic formalism [11]. This technique is the curve fitting of TL curves consisting of one or more TL peaks and widely used in various area of TL studied and is well documented [12-14]. In the fitting process, the number of minima of the second derivative plot of glow curve can guide the location of the glow peak temperatures. This method can give a mathematical description of thermoluminescence phenomenon and many experimental glow curves can be described with reasonable degree of confidence.

Recently, Lovedy and Gartia [15]. formulated a simplified form of the One Trap One recombination (OTOR) differential equation for routine analysis and they also formulated the lifetime equation for evaluating the lifetime based on the simplified OTOR differential equation [16]. The simplified General one Trap (GOT) differential equation removes the empirical nature involved in the general order kinetics and spans the region from the first order kinetics to second order kinetics. The key feature of the simplified GOT differential equation is the ability to extract not only the trapping parameters namely activation energy (E) and frequency factor (s) but also the other three basic trapping parameters viz. N (number of traps present), α (ratio of the re-trapping probability to the recombination probability) and γ (= N/n_0, n_0 is the number of electrons in the traps).

In the present work, we carried out spectroscopic investigation of the TL glow curves of Ba_2SO_4 by the simplified OTOR differential equation formalism [17]. $BaSO_4$ based phosphors being high sensitive TL materials were used in TL Dosimetry [18]. Manam & Das [19] reported that the TL intensity of $BaSO_4$ phosphor increases by the presence of impurities and could increased by about nine times by the presence of Cu as impurity but about three times by the presence of Mn when compared with that of undoped $BaSO_4$. Due to its high sensitivity and stability, doped $BaSO_4$ with Cu & Mn is of routine use for personal and environmental radiation monitoring.

2. Experiments

TL glow curves of $BaSO_4$ procured from MERC with 99.9% purity are recorded after β-irradiation up to 1, 10, 50 and 100 Gy with heating rate 2°Cs^{-1} in the temperature range from room temperature to 400°C. All the TL curves were measured using commercial TL Reader Model 900I (Neocleonix Systems Pvt. Lt., Hyderabad, India). The different heating rates used in the present analysis were 0.5, 1.0, 2.0 and 5.0°Cs^{-1}. For TL readout 20 mg each of the powder sample were used. A second readout was performed to record the background radiation which includes the black body radiation. The data presented are all with background subtraction. Glow curves of low heating rates namely 0.5 and 1.0°Cs^{-1} were used for suitable correction of thermal lag.

3 Methods of Analysis

The theoretical derivation of the present work has been described in detail in the recent work of Lovedy and Gartia. [17] Only two equation of importance of (i.e. 22 (a) and (b)) of Lovedy and Gartia [17] are presented below:

$$I_{TL} = \frac{N\alpha s \exp\left(-E/kT\right)}{\beta(1-\alpha)^2 \omega(x(T))(1+\omega(x(T)))} \qquad (1)$$

where $n(cm^{-3})$ is the concentration of electron traps, $N(cm^{-3})$ the concentration of traps of the kind responsible for the peak being considered, α the ratio of the retrapping probabilities to the recombination probabilities, s(sec^{-1}) is the frequency factor, $E(eV)$ the activation energy, β ($^{\circ}Cs^{-1}$) the heating rate, ω the Wright omega function and

$$x(T) = \frac{s}{\beta(1-\alpha)}\int_{T_o}^{T} exp\left(\frac{-E}{kT}\right)dT + \left(\frac{N\alpha}{(1-\alpha)}\right)\frac{1}{n_o} + \log\left(\left(\frac{N\alpha}{(1-\alpha)}\right)\frac{1}{n_o}\right) \quad (2)$$

If we take $\gamma = N / n_o$ then the eqn (2) can be written as

$$x(T) = \frac{s}{\beta(1-\alpha)}\int_{T_o}^{T} exp\left(\frac{-E}{kT}\right)dT + \left(\frac{\gamma\alpha}{(1-\alpha)}\right) + \log\left(\left(\frac{\gamma\alpha}{(1-\alpha)}\right)\right) \quad (3)$$

The model used here for analyzing these TL glow curves assumed a set of discrete electron traps and a set of hole traps (recombination center). All the glow curves of different heating rates are subjected to glow curve deconvolution using simplified GOT differential equations. Computing and fitting of the glow peaks following the simplified GOT differential equations were done from the program developed [20].

The lifetime of the trap electrons are calculated from the equation (i.e. 21) of Lovedy and Gartia [16]

$$\tau_1 = \frac{\alpha \exp(E/kT)}{s}\left(\log(n_o) + \frac{(1-\alpha)}{\alpha} - 1\right) \quad (4)$$

where τ_1 gives the expression for time required for a saturated trap concentration to be reduced to 1.

The goodness of fit of the measured TL glow curve was again tested using $\chi 2$-test of normality [13, 21, 22] which measures the goodness of fit in terms of normality of error distribution (i.e. difference between the observed and calculated intensity). As a cross check, Figure of Merit (FOM) [23-24] was also calculated.

In order to obtain trapping parameters for higher rate of heating, evaluation of the real glow peak temperatures (T_m) is necessary [25] and is calculated using the relation

$$T_m^j = T_m^i - c\ln\left(\frac{\beta_i}{\beta j}\right) \quad (5)$$

where T_m^i and T_m^j are the maximum temperatures of a glow peak with heating rates β_i and β_j, respectively, and c is a constant which is usually evaluated by using two very low heating rates (preferably below 1.00°Cs^{-1}) where the thermal lag (TLA) can be considered as negligible [26]. The effective heating rate (β_{eff}) between the heating element and the thermoluminescent sample during the TL readout in the reader (using contact heating) has been taken into consideration to avoid errors in determining the trapping parameters by glow curve deconvolution is. A simple method of heating rate correction [25] was used to avoid this problem and determine the exact effective heating rate of the TL sample by using the equation:

$$\beta_{eff} = \frac{(T_g - T_0 - \Delta T)}{(T_g - T_0)}\beta = \frac{(T_m - T_0)}{(T_g - T_0)}\beta \quad (6)$$

where $\Delta T = T_g - T_m$, T_g is the observed peak temperature (K) and T_m is the real peak temperature (i.e., with thermal lag correction), T_o is room temperature (25°C).

4. Results and Discussion

TL glow curves of $BaSO_4$ subjected to various doses (1, 10, 50 and 100Gy) of β-irradiation at a constant heating rate of 2°Cs^{-1} is shown in Figure 1. In all cases the same pattern of glow curves are observed with most intense peak at about 190°C. The experimental glow curves show a complex structure indicating a number of TL peaks all over the region. The dose response of the glow curves is shown in Figure 2, which shows that it is linear up to 100 Gy. The glow curves presented in Figure 1 are subjected to glow

curve deconvolution using the simplified OTOR differential equations [15] For correcting the heating rate we followed the works of Kitis and his co-workers [25]. The results of the analysis are presented in Table 1 and some of the fitting are shown in Figs.(3a-3c). The outcome of the analysis show that not only the values of the key trapping parameters namely E, s, n_0, γ and α are in physically realistic range, but also the fitting are extremely good which are also supported by FOM and χ^2 values. In all cases FOM is less than 1% and χ^2 test passes at 5% level of probability. The histogram of error also presented in Figure 3b (inset), which shows the normality of deviation.

5 Conclusion

Trapping parameters obtained from the analysis of $BaSO_4$ glow curves irradiated at different dose of β-irradiation show activation energy in the range 0.68 – 1.2 eV and frequency factor in the range 10^8 – 10^{11} s^{-1}. Each glow curves can be fitted by five constituent's peaks.

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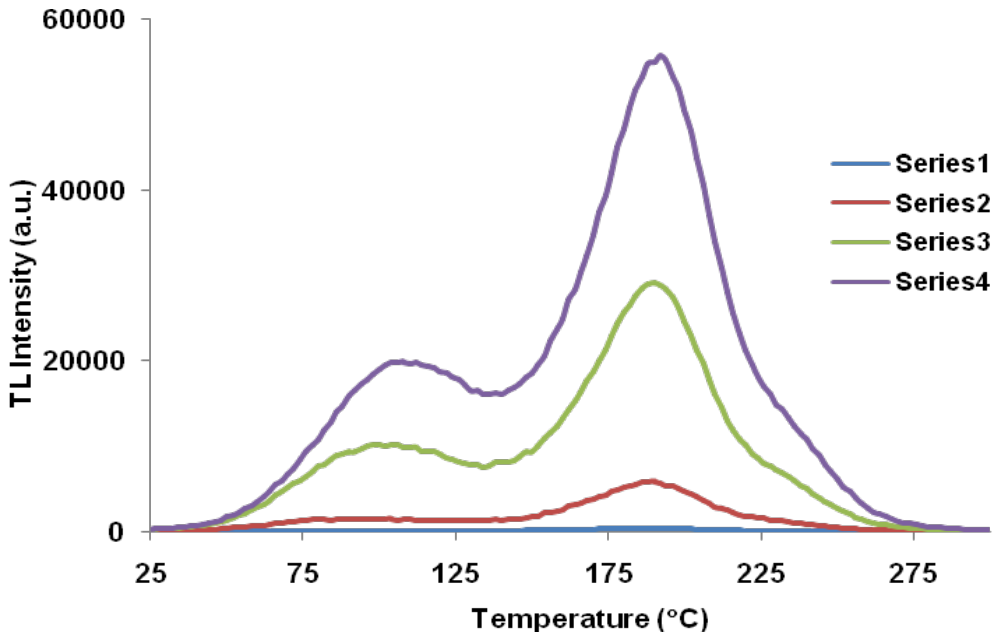

Figure 1. Glow curves of $BaSO_4$ subjected to different dose of β-irradiation with constant heating rate 2°Cs^{-1}. (Series 1, 2, 3 & 4 corresponds to 1, 10, 50 & 100 Gy respectively)

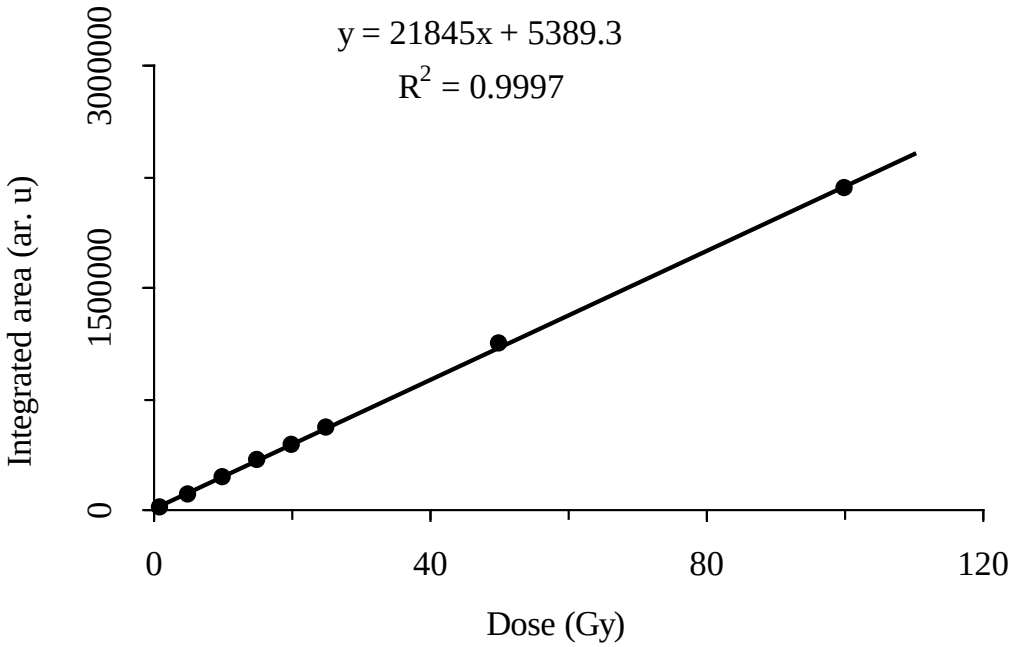

Figure 2. Dose Response curve of β - irradiated $BaSO_4$.

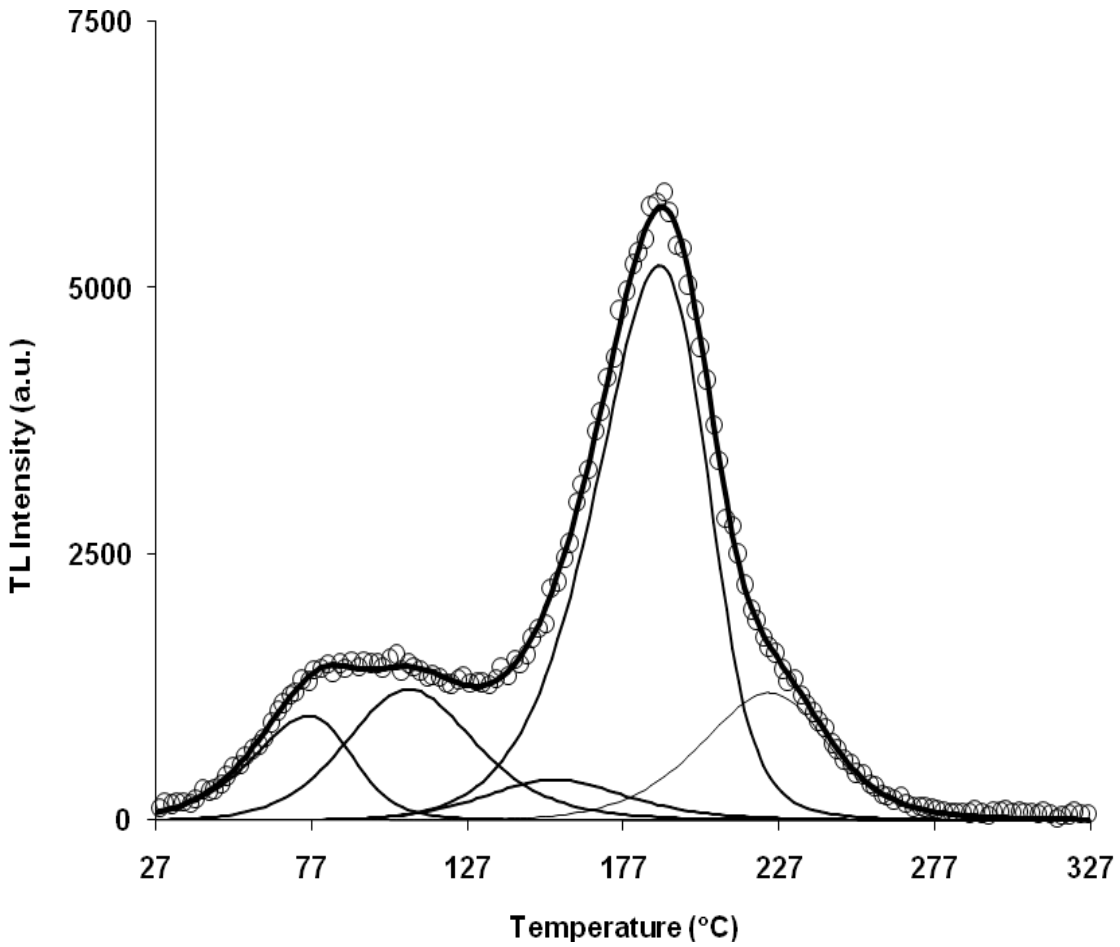

Figure 3a. CGCD of $BaSO_4$ irradiated at 10Gy. (Heating rate = 2°Cs^{-1})

oooooo Experimental curve

——— Numerically generated curve

▬▬▬ Sum of the numerically generated best curves.

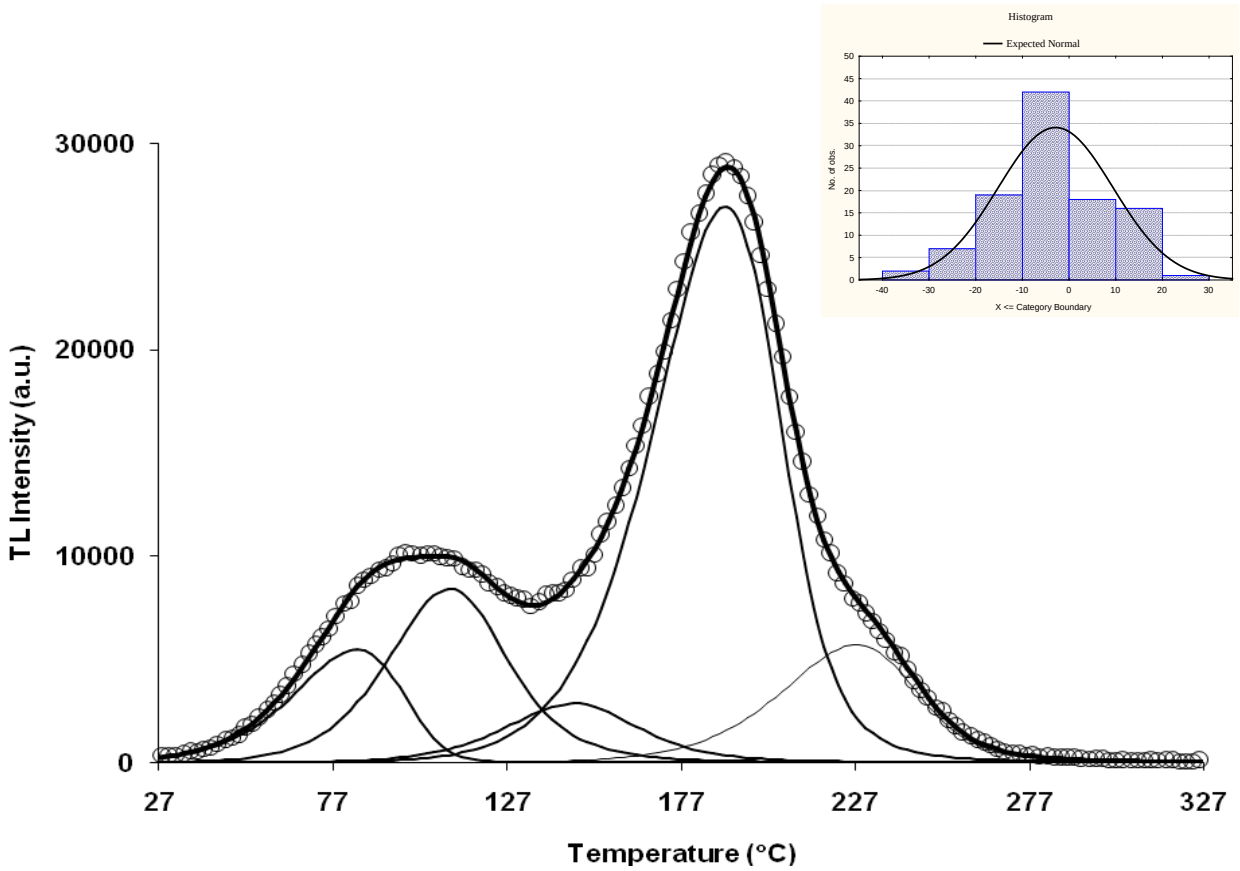

Figure 3b. CGCD of BaSO4 irradiated at 50Gy. (Heating rate = 2°Cs^{-1})

oooooo Experimental curve

——— Numerically generated curve

▬▬▬ Sum of the numerically generated best curves.

(Inset : The histogram of error)

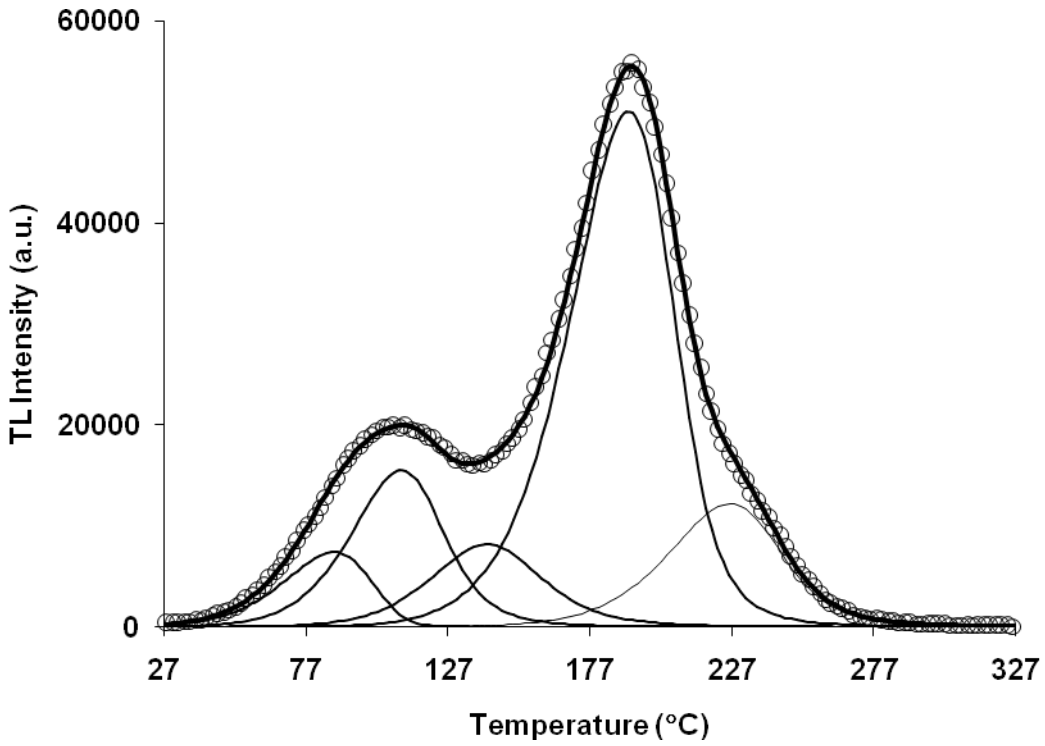

Figure 3c. CGCD of $BaSO_4$ irradiated at 100Gy. (Heating rate = 2°Cs^{-1})

oooooo Experimental curve

——— Numerically generated curve

━━━ Sum of the numerically generated best curves.

Table 1: Trapping parameter as obtained using CGCD of the glow curves with different dose of β-irradiation with constant heating rate 2°Cs^{-1}

Dose (Gy)	T_m (°C)	E (eV)	s(sec^{-1})	n_0	α	γ (= N/n_0)	τ	FOM (%)	χ2 (d.f.)
1	88	0.66	4.75×10^{8}	3852.554	0.0114	88.10	5.02 min	1.75	1.07 (2)
	116	0.83	2.42×10^{10}	3790.545	0.0129	182.23	1.22 hr		
	152	0.96	3.82×10^{10}	2854.252	0.0020	165.82	4.58 day		
	188	1.02	1.86×10^{10}	20849.687	0.0009	443.52	2.49 month		
	226	1.01	4.36×10^{10}	4677.146	0.0208	172.27	1.84 yr		
10	76	0.66	4.75×10^{8}	41902.469	0.0114	8.10	5.15 min	1.37	0.51 (3)
	108	0.82	4.81×10^{10}	68526.885	0.4636	10.08	1.92 hr		
	154	0.92	3.07×10^{10}	23372.444	0.1619	20.25	2.62 day		
	188	1.01	1.33×10^{10}	248782.701	0.0013	37.17	3.19 month		
	224	1.14	4.36×10^{10}	65666.829	0.0208	12.27	11.81 yr		
50	84	0.68	4.75×10^{8}	223296.053	0.0051	1.52	9.17 min	0.72	7.50 (3)
	110	0.88	4.81×10^{10}	388062.360	0.2636	1.78	11.07 hr		
	146	0.94	3.07×10^{10}	145628.308	0.1619	3.25	6.32 day		
	190	1.02	4.32×10^{10}	1315398.720	0.0108	7.03	3.78 month		
	228	1.15	4.36×10^{10}	294062.774	0.0401	2.74	23.53 yr		
100	86	0.70	9.40×10^{8}	297728.070	0.0061	1.14	13.04 min	0.71	5.18 (3)
	110	0.85	1.57×10^{10}	677206.863	0.1971	1.02	8.66 hr		
	140	0.93	2.27×10^{10}	426389.189	0.4481	1.11	10.73 day		
	190	1.02	1.35×10^{10}	2492521.020	0.0202	3.71	4.63 month		
	226	1.15	4.36×10^{10}	649783.871	0.1277	1.24	39.19 yr		

Calculated χ^2-values are accepted at 5% level of probability.