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TL SENSITIVITY CONSTANCY OF QUARTZ UPON UV + (β,γ) IRRADIATION CYCLE: AN IMPROVEMENT ON DATING METHODOLOGY

by

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ABSTRACT

Thermoluminescence (TL) sensitivity of natural quartz (beaches,paleobeaches and fixed dunes) to Beta and Gamma rays has been studied in a temperature range between 250-400C, before and after bleaching by solar irradiation. A first TL glow growth curve was obtained through Beta irradiation of "as found" samples and a second glow growth curve was obtained by bleaching through solar irradiation the TL acquired either through an excitation Gamma dose or through environmental radiation to finally re-irradiate the samples under Beta rays. Experimental results showing the constancy of sensitivity, for doses of until about 10krads, are the basis for a proposal to improve TL dating methodology expanding its present limits.

Key-words: Thermoluminescence; Dating quartz; Solar bleaching.

1 INTRODUCTION

TL dating techniques applied to quartz specimens have been widely used for archaeological and geological purposes[1-6]. In TL dating, minerals are treated as dosimeters. One of the problems is to determine the dose of radiation, wich we will call natural dose, received by the sample during its burial time. A number of TL methods have been proposed; among them, that of Additive Doses[7] is the most diffused one. In this method, known doses of radiation are added to the natural dose, archaeologically deposited in the sample, to construct a plot of TL growth versus dose. Assuming a simple form for the curve usually a straight line, extrapolation to lower values of TL will show the natural dose responsible for the natural TL found (TLN). In geochronological studies, when one has old samples with ages superior to about 30.000 years, linear fitting is not appropriate and extrapolations become unfeasible since a general analytical expression is still not established. A reconstruction of TL growth becomes then very important to find natural doses in samples where linear extrapolation is not possible.

To reconstruct the TL growth it is, first of all, necessary to bleach the TL already existing in the sample (TLN). Thermal bleaching is known to change the TL sensitivity; to account for this, one has to resort to plausible assumptions, like for instance assuming no change in the shape of the growth curve on heating[8] to finally calculate the natural dose through a fitting process.

Solar irradiation is also a bleaching mechanism for quartz[9]. It sets the zero time at the moment of the sample burial, opening in this way the possibility of dating that will determine the time interval between the last exposition to sunlight and today[10,11]. Taking into account the "softness" of solar UV interaction with matter compared with heat treatments that can destroy (annealing) or create (quenching) lattice deffects, it has been possible to verify the immutability of sensitivity of quartz samples after solar bleaching. In this work we propose a method in quartz TL dating, by bleaching the sample TL through solar irradiation to reconstruct the glow growth curves until the TLN value. In the next section we will describe the experiments done to verify the TL sensitivity constancy of quartz after solar bleaching.

2 EXPERIMENTAL MEASUREMENTS AND ANALYSIS

Natural detrital quartz from different geological contexts (beaches, paleo-beaches and fixed dunes) has been purified by: 1) sieving to grain size about 100μm; 2) chemical treatment by HF at 40% in concentration during 40-60 min; 3) new sieving to separate minerals that were not completely eliminated by the acid.

In a first series of measurements, samples found at the surface and bleached by sun light to their residual level (TLR) as verified through ulterior solar irradiation were taken. For each sample two glow growth curves were constructed (e.g. fig 1). The first one represents, in analogy to the natural process, the TL growth through centuries of sample burial and the second one depicts the growth that we could observe today after a new solar bleaching.In order to obtain these curves, samples were divided into two equal parts after the chemical treatment; one of these parts has received several different Beta doses in order to construct the first glow growth curve while the other part has received a single Gamma dose, in order to simulate sample aging. Afterwards a solar bleaching during 2 weeks was able to bring TL of this second part back to TLR .Finally from that part wich had been Solar-Gamma treated it was possible to construct a second growth curve from different Beta doses.To construct the growth curves in fig.1 ,measurements from glow curves were taken at 280C.The heating rate was 5C/s and a MTO H 325c optical filter was placed between sample and the photomultiplier tube (XP2230-PHILLIPS). Comparison between curves' shapes is shown in fig.2 where two curves obtained from the same radiation dose,given before and after solar bleaching ,present a plateau in the 250-400C temperature range. Comparison between glow curves' shapes at lower temperatures should consider the relatively quick thermal bleaching and this was not done in this work. Note that this plateau is related to curves' shapes comparison and should not be confused with TL fading determination plateaus often presented in dating literature.Interpolation between points from first and second glow growth curves in fig.1 and plateaus in fig.2 shows the constancy of TL sensitivity in a wide temperature range (ΔT=150C).

Beta irradiations were performed by a ^{90}Sr source at about 600 rad/min and Gamma irradiations by a ^{60}Co source at about 20 rad/min. Solar bleaching in the next set of experiments,was performed by a 275W General Electric solar lamp placed 30 cm above the samples.

To investigate a possible influence due to dose absorption rate , a second series of measurements on "natural" samples (naturally irradiated at about 100mrads/year during their burial time) was undertaken. In this case Gamma irradiations were not necessary since TLN were already present in the samples.The two above mentioned glow growth curves were obtained by exposing the sample to Beta rays (fig. 3-4);the first one was constructed from TLN and the second one from TLR resulting from the bleaching of TLN by solar lamp irradiation during 13h (e.g.fig.5). Finally displacing the first growth curve to compensate its natural dose (fig.3a-4a), points from the first and second growth curves interpolate smoothly (fig.6). For sample RMG07B this interpolation can be seen even before the displacement (fig.4) due to its small natural dose.In this way it was verified, also in the case of natural irradiation, that there was no change in the quartz sensitivity, in other words,that in applying the proposed method we are,to an excellent approximation,reconstructing TL glow growth curves as it was done by natural processes.

3 DISCUSSION: POTENTIALITY AND LIMITATIONS

In the present work it was shown that until doses of about 10krads no irreversible radiation damage is caused to the sample. Assuming 100mrad/year as annual dose rate at the burial site and 600rad/min our laboratory dose rate, one have 100.000 years as a first approximation of practical time limitations on dating. This limit represents an improvement of about a factor 3 over the Additive Doses Method[7]'s limit. Fig 3 shows that this limit can be above 1 million years if no permanent radiation damage is caused to the sample, but this assumption is not obvious and so a study of this limit is in progress in our laboratory. On the other hand fig 4 shows that the lower limit can be set at least about 1500 years. The displacement one must make in order to find natural dose can be optimized by fitting analytical expressions to experimental measurements. Until 12krads doses it was possible to fit the results to a simple TL=f(D) curve[12], which should represent one trap filling, given by

$$TL/TLsat=1-\exp(-(D+DN)/Dsat)^{\alpha} \qquad \text{where:}$$

DN is the natural dose

TLsat and Dsat are constants linked to saturation values

$\alpha \neq 1$ makes explicit the interdependence between different traps.

To fit the curve at higher doses it seems that one should consider a model with several traps with : different saturation levels; different electron capture cross sections and almost the same or even the same temperature peak position in a TL glow curve. On the other hand, apart from phenomenological interpretations , good fitting to any analytical function could be helpful on natural dose calculations.

Apart from annual dose determination, the main source of errors in sedments dating, for wich the zero time is set by the solar bleaching, is the appropriate evaluation of the residual TL (TLR) wich was already present in the sample at the moment of its burial. Sample COPA has attained its original TLR after 2 weeks of solar irradiation and also after 13h of solar lamp irradiation while RMG04B (fig 5) showed that upon longer irradiation times ,TLR can still decrease to about 50% of this value. This difference has almost no importance in dating older samples (fig 3a) but for more recent ones (fig 4a), TLR has a greater importance.

Recent sample dating must give the same results when done by Additive Doses Method[7], employed by Singhvi et al[11] to date recent sand dunes, or by the procedure described in this work due to TL almost linear growth at low radiation doses, but for older samples (fig 6) , linear extrapolation at the TLN point can lead to false results.

Since all the samples studied during this work (coastal sand from different geological contexts representing also a significant range of ages between zero and about 100.000 years) have yieled good results, namely constancy of sensitivity under the described conditions ,we believe that the presented methodology can be extended also to other samples.

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CAPTIONS

FIGURE 1 TL glow growth versus dose curves from a present-day beach, measured at 280C; heating rate=5C/s; optical filter with passing band at about 325nm. Dashed line, Gamma Excitation Level, shows the aging dose taken before solar bleaching.

FIGURE 2 Comparison between glow curves' shapes obtained before and after solar bleaching.

Fig.2a $TL1=TL(R+\gamma+UV+6Krad\beta)$ and $TL2=TL(R+6Krad\beta)$

fig.2b $TL1=TL(R+\gamma+UV+12Krad\beta)$ and $TL2=TL(R+12Krad\beta)$

fig.2c $TL1=TL(R+\gamma)$ and $TL2=TL(R+\gamma+UV+9Krad\beta)$ where:

R means unbleached residual dose

UV is solar bleaching and

γ is an Gamma aging dose of about 9Krad.

Black body radiation from heating plate is not included in TL1 or TL2.

FIGURE 3 TL glow growth curves versus dose, reported in irradiation time during wich β dose was absorbed at about 600rad/min. Sample RMG04B is quartz from a fixed dune. Measurements were taken at 330C; heating rate=5C/s, optical fiter with passing band at about 380nm. Fig 3a shows the natural dose that this sample must have received in order to present TLN today. Dashed line indicates the dose wich would be absorbed by the sample from environment radiation in 1 million years assuming 100mrads/year as annual dose rate.

FIGURE 4 Idem as in figure 3, but in this case sample RMG07B cames from a recent paleobeach. Fig 4a shows the importance of an appropriate TLR determination.

FIGURE 5 TL versus bleaching time.

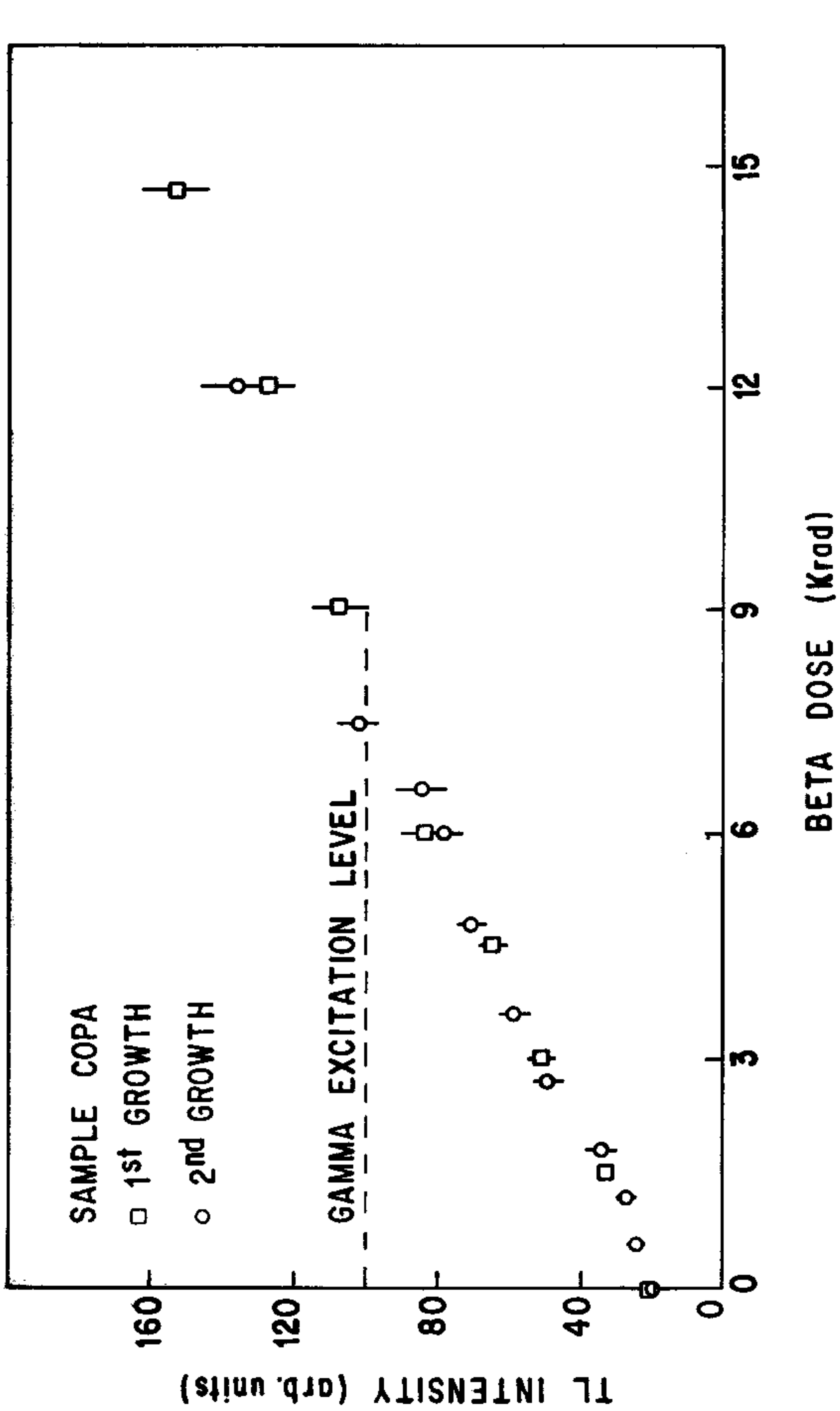
SAMPLE COPA
1st GROWTH
2nd GROWTH
GAMMA EXCITATION LEVEL
TL INTENSITY (arb. units)
BETA DOSE (Krad)
0
3
6
9
12
15
40
80
120
160

FIG. 1

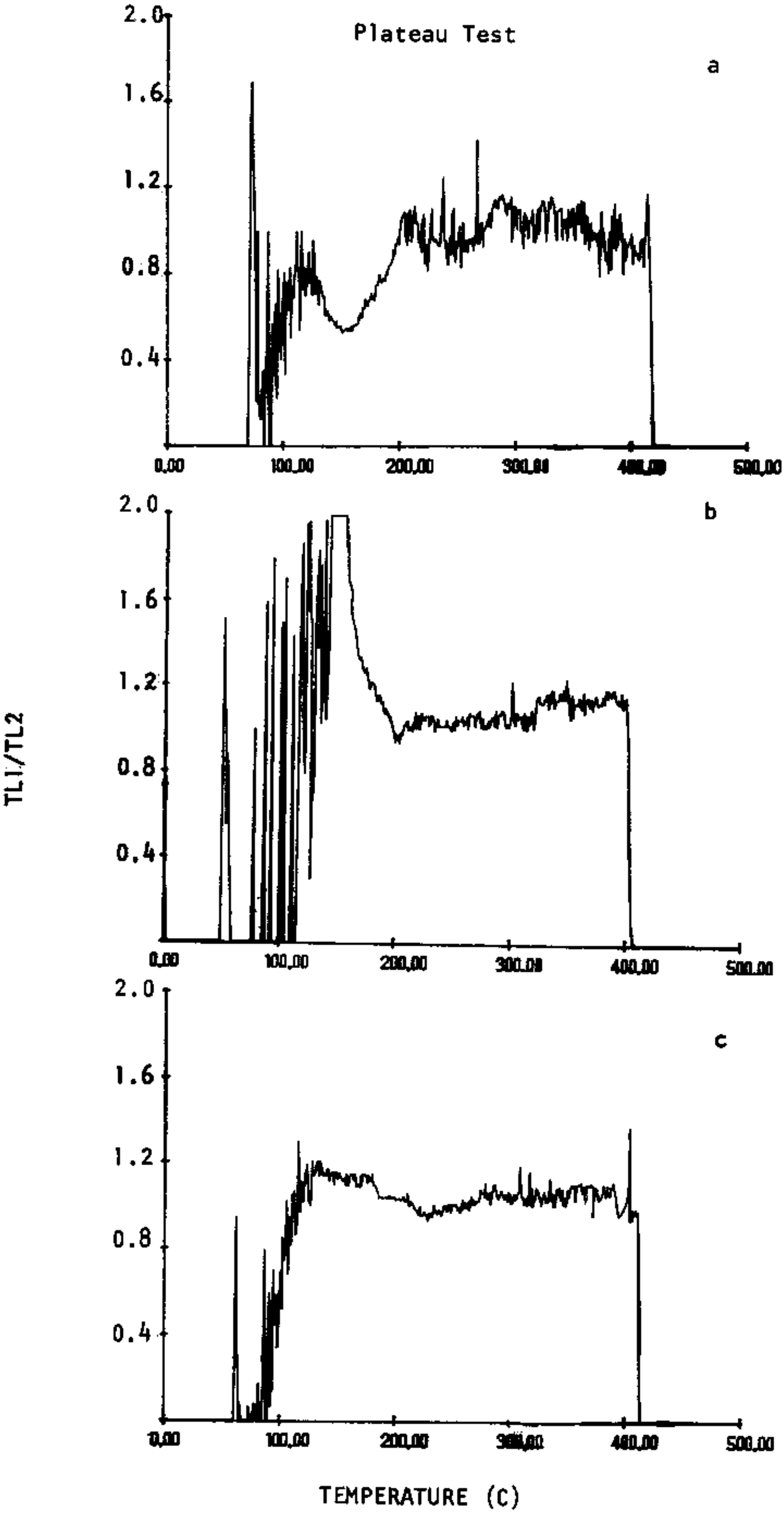
Plateau Test
a
b
c
TL1/TL2
2.0
1.6
1.2
0.8
0.4
0.00
100.00
200.00
300.00
400.00
500.00
TEMPERATURE (C)

FIG. 2

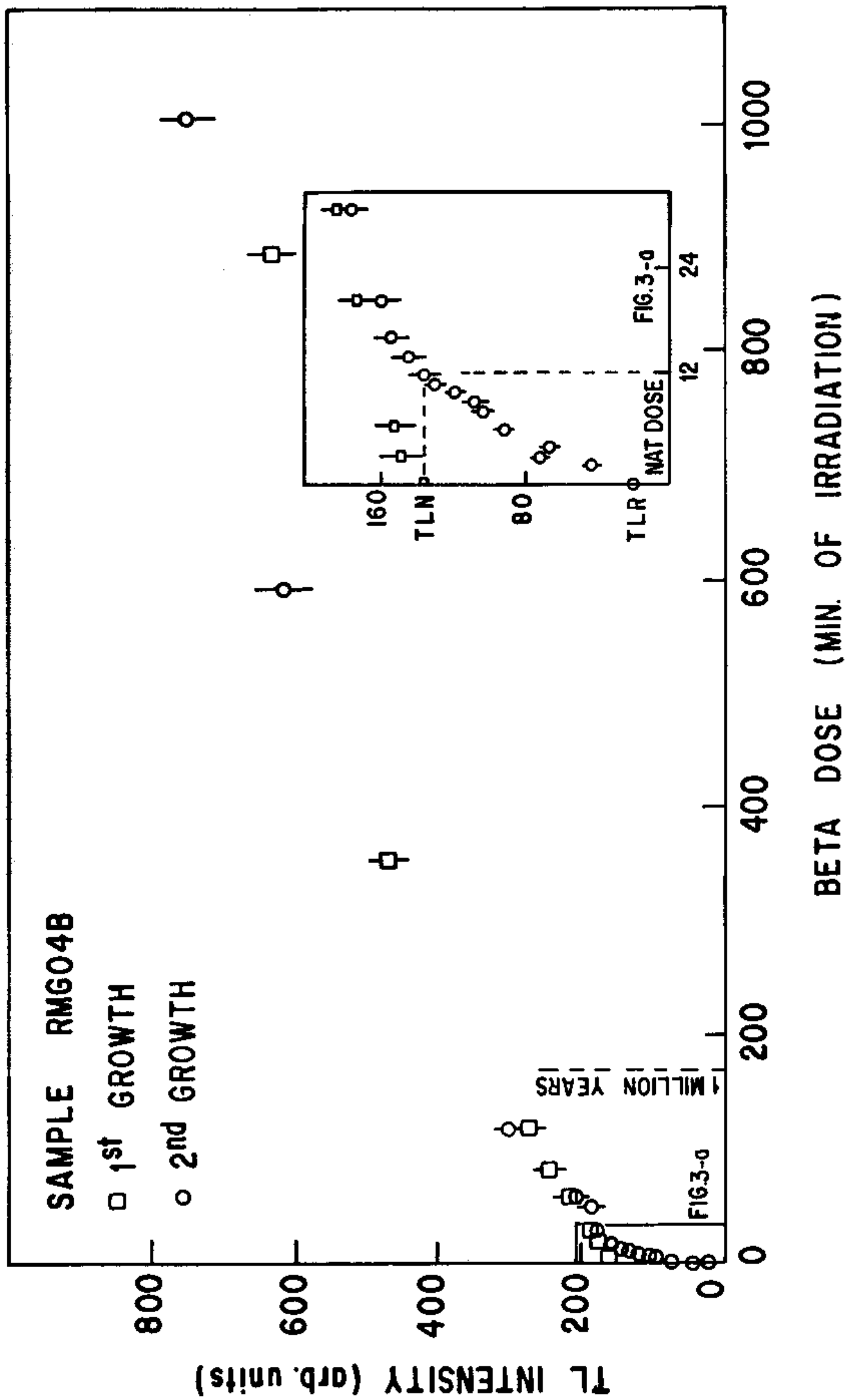
SAMPLE RMG04B
□ 1st GROWTH
○ 2nd GROWTH
TL INTENSITY (arb. units)
BETA DOSE (MIN. OF IRRADIATION)
0
200
400
600
800
1000
1 MILLION YEARS
FIG.3-a
160
TLN
80
TLR
12
24
NAT DOSE

FIG. 3

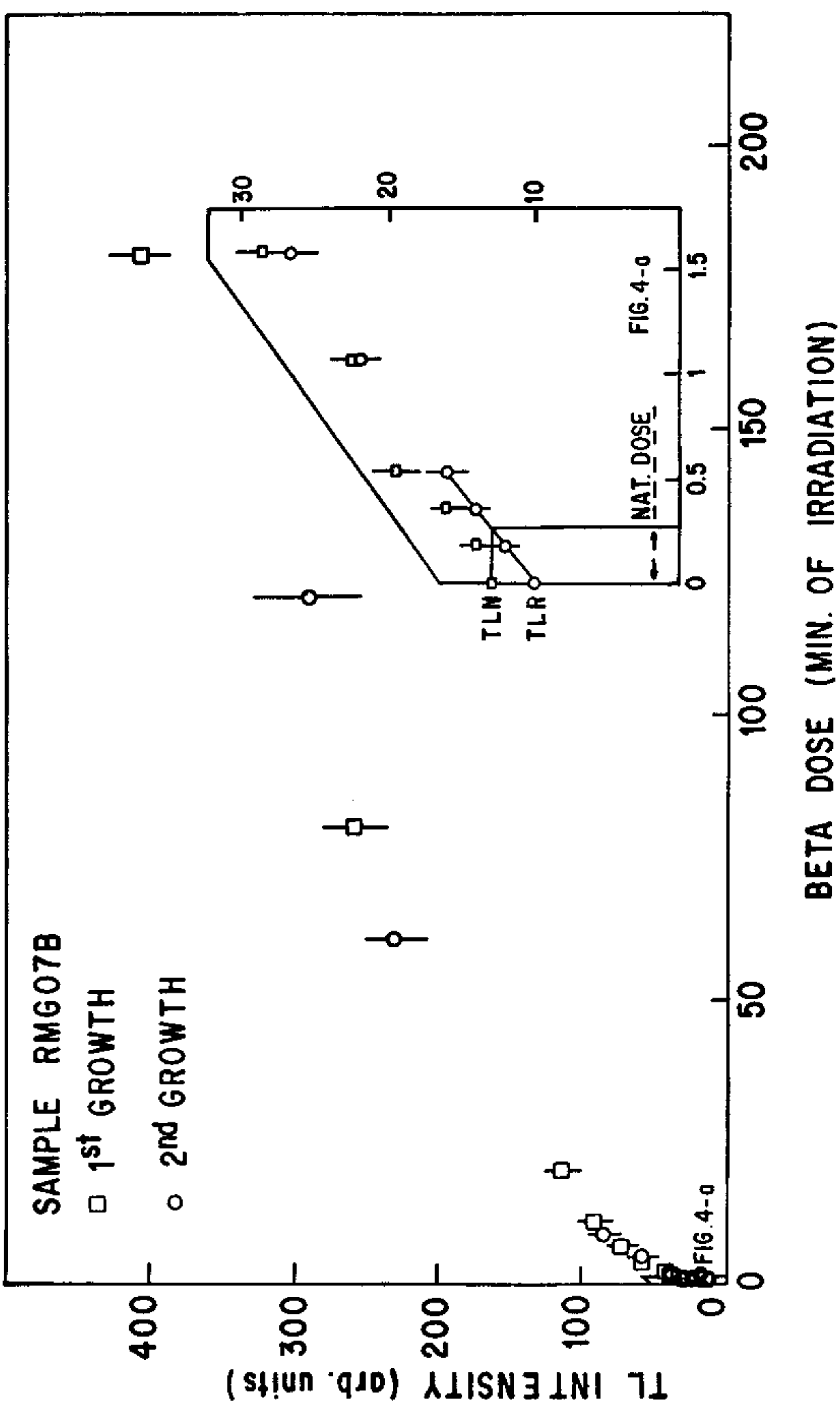
SAMPLE RMG07B
1st GROWTH
2nd GROWTH
TL INTENSITY (arb. units)
BETA DOSE (MIN. OF IRRADIATION)
0
100
200
300
400
0
50
100
150
200
TLN
TLR
NAT. DOSE
FIG. 4-a
0
0.5
1
1.5
10
20
30

FIG. 4

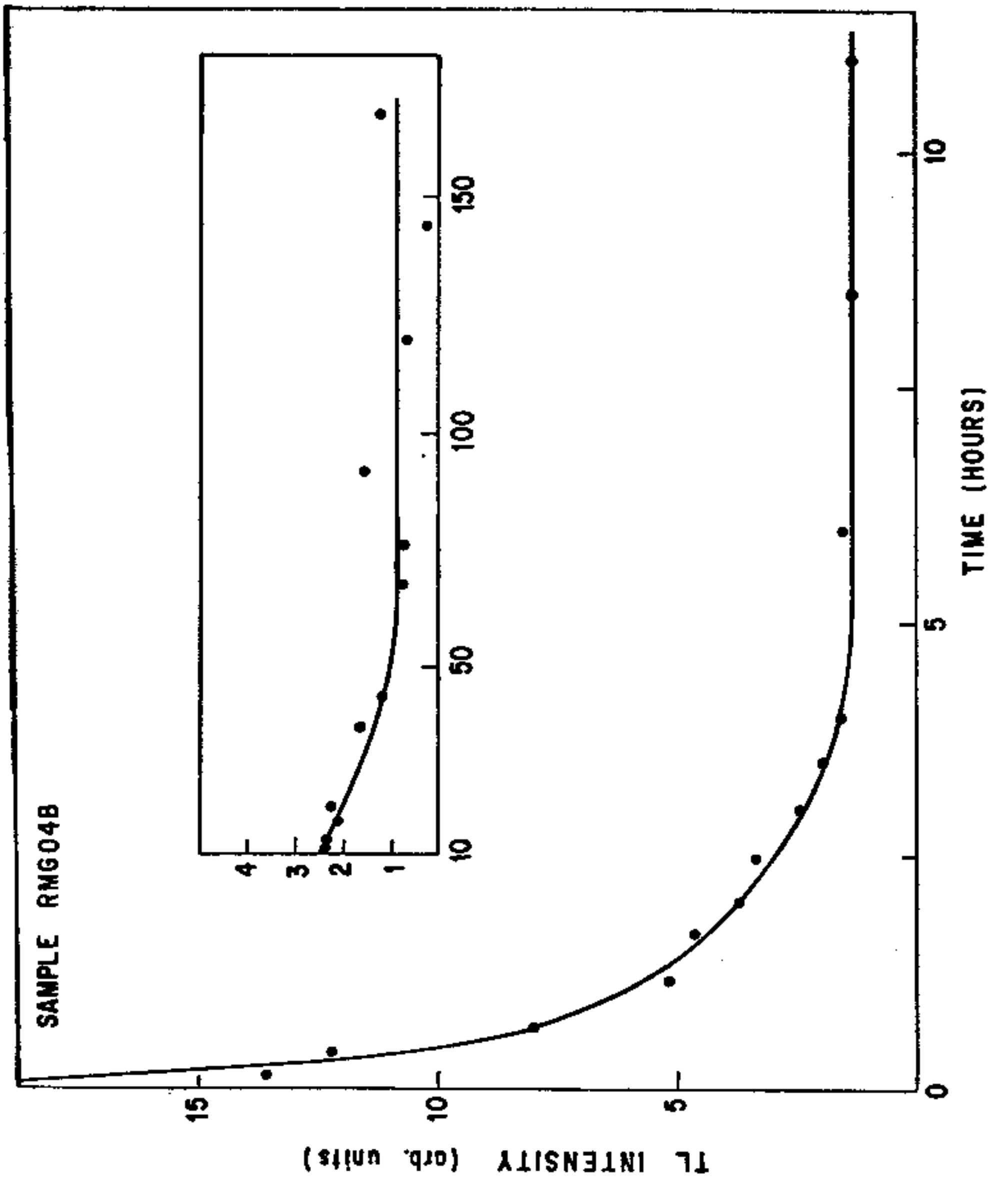
SAMPLE RMG04B
TL INTENSITY (arb. units)
TIME (HOURS)
0
5
10
15
10
50
100
150
1
2
3
4

FIG. 5

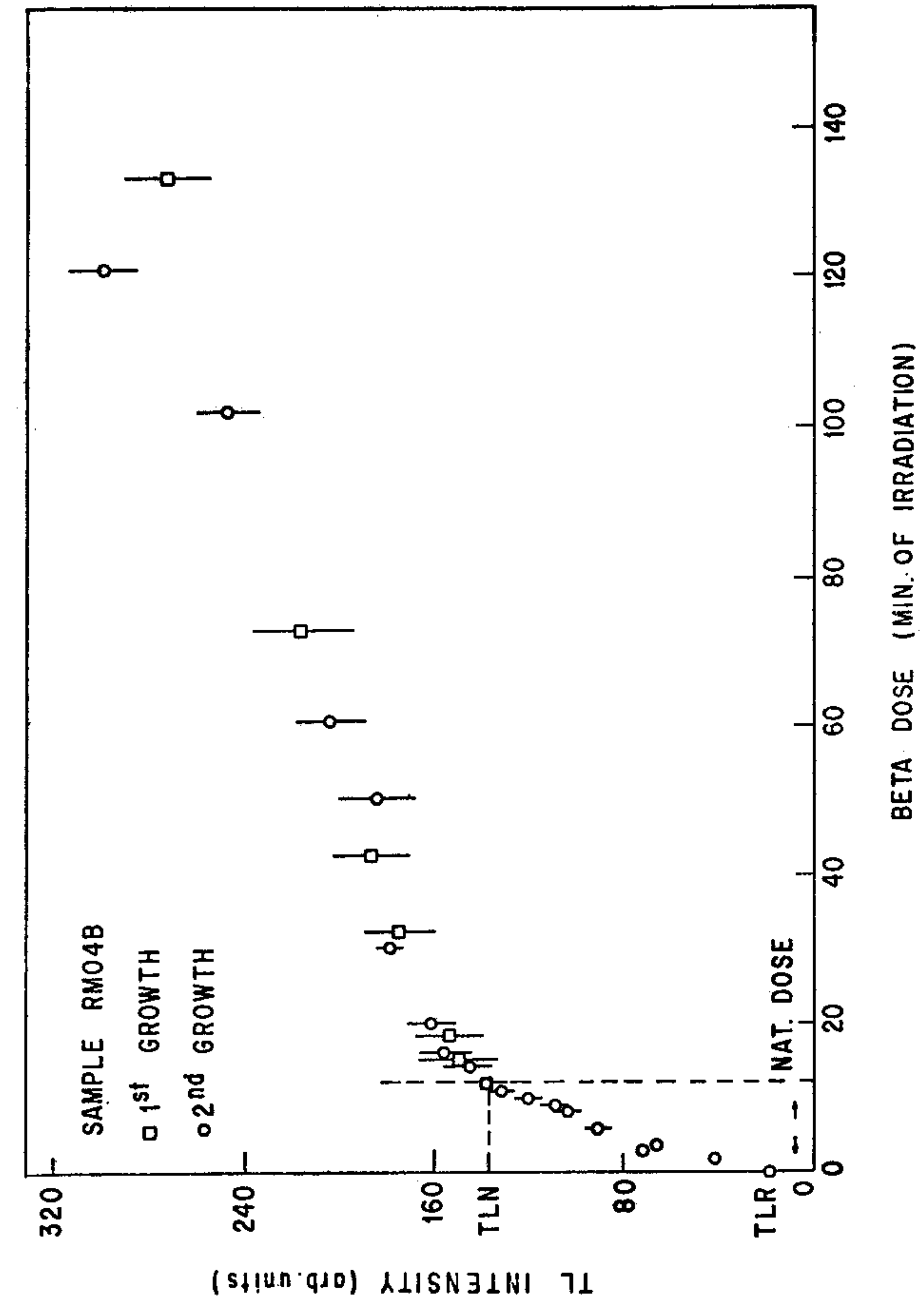
SAMPLE RM04B
□ 1st GROWTH
○ 2nd GROWTH
NAT. DOSE
BETA DOSE (MIN. OF IRRADIATION)
0
20
40
60
80
100
120
140
TL INTENSITY (arb.units)
TLR
0
80
TLN
160
240
320

FIG. 6

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