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	PHOTOLUMINESCENCE	

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CHAPTER III

PHOTOLUMINESCENCE

3.1 INTRODUCTION

The study of phosphorescence decay provides information about nature of trapping states, energy levels of luminescent material and the type of kinetics involved in the luminescence process. The aim of present investigation is analysis of observations made on CaS: Mn:Sm phosphors at room temperature $(300^{\circ}k)$ so as to get information about following aspects :

- 1) Nature of decay law
- 2) Energy distribution of trap levels
- 3) Kinetics involved in decay mechanism
- 4) The effect of activators on decay behaviour.

3.2 THEORETICAL BACKGROUND

An important step in the theory of phosphorescence was that made by Jablonskii (1) in 1935 for the decay of emission from organic molecules in rigid media. He proposed an energy level scheme for emitting center as shown in fig. 1.4. The fluorescence is given by the transitions between the excited state F and the ground state G, but excited molecules or centers may relax to the metastable state M, transitions M \leftrightarrow G being forbidden. To return to ground state the transition M \rightarrow F must be effected by thermal or optical activation. Thus phosphorescence emission due to $F \rightarrow G$ is conditioned in rate by optical process M \rightarrow F. If the energy required for the latter is E, then the probability per second that it occures is given by

 $p = s \exp(-E/KT)$... (3.1)

where T is the absolute temperature of the phosphor, S is the escape frequency factor and E is the electron trap depth.

In general, a phosphor contains more than a single trap and phosphorescence is known to be a cumulative effect of traps of different depths. Its intensity is dependent on the rate of escape of electrons from traps. The retrapping of electrons in traps also affects the behaviour of phosphorescence.

3.2.1 Decay Laws :

The phosphorescence decay may be either exponential or hyperbolic (2) depending upon the type of kinetics involved in the luminescence process.

1) Exponential decay

In this, as the name suggest, the emission intensity diminishes exponentially with time. The finite delay in emission of absorbed energy is due to life time of the excited state of the emitting atom, ion or molecule. If p is the transition probability for return of the center to the ground state and n centers are excited at any instant, then

$$\frac{dn}{dt} = J - pn \qquad \dots (3.2)$$

where J is the number of excited centers per second by the incident radiation. The transition probability p, according to Randall and Wilkins (3) is given by $eq^n(3.1)$; viz. p = 3 exp.(-E/KT).

The decay of luminescence is given by equation (3.2) with J=0. The equation (3.2) then becomes -

 $\frac{dn}{dt} = -pn$ $\frac{dn}{n} = -p dt$

On integration we get -

n = no exp.(-pt) .. (3.3)

where no is the number of excited electrons at t=0, the time when excitation ceases.

The luminescence intensity is given by

I =
$$-\frac{dn}{dt}$$
 = - (- pn)
= p no exp. (-pt)
... (3.4)

where Io is the intensity at t = 0 and p = $1/\gamma$, γ being the life time of excited state.

The equation (3.4) shows that decay is exponential and the rate of decay depends only upon the transition probability p. The exponential decay occures, when the kinetics of luminescence process is of the first order. i.e. when electron traps are situated quite close to the luminescence centers. The equation (3.4) is useful in evaluating the trap depth.

B) Hyperbolic decay

This type of decay results when luminescence process is of the second order, i.e. when electron traps are located away from luminescence centers.

Let there be n impurity centers with which only n number of electrons combine at a time. If p is the probability with which an electron combines with any of the centers in time dt, then

p = A n dt ... (3.5) where A is a constant known as recombination coefficient. If all the electrons return to the ground state at the centers through the conduction band with the above said probability p, then the rate of decay will be,

$$\frac{dn}{dt} = -A n^2 \qquad \dots (3.6)$$

On integration

$$n = \frac{n_0}{1 + n_0 At}$$
 ... (3.7)

and the luminescence intensity is given by

$$I = -\frac{dn}{dt} = -(-An^{2})$$

= $\frac{n_{0}^{2} A}{(1 + n_{0} At)^{2}}$
= $\frac{Io}{(1 + a t)^{2}}$... (3.8)

where = n_o A is another constant.

For large values of t (t >> 1) the equation (3.8) can be written as

$$I = \frac{IO}{\alpha t^2} \qquad \dots (3.9)$$

Treating more **gamer** generally in the light of different capture cross sections for electrons empty centers and traps, Adirowitch (4, 5) arrived at an approximate Becquerel type relation of the form

$$I = \underline{\text{constant}} \dots (3.10)$$

$$(t + to)^{b}$$

where b depends on the ratio of capture cross-section of traps and empty centers. However, this equation fails in cases where well seperated groups of traps exist.

3.2.2 Effect of steady excitation of a phosphor on the filling of electron traps :

Luminescence intensity of a phosphor reaches to a constant value if it is excited at fix temperature by steady excitation source. This state of phosphor is called equilibrium state, and in this condition the concentration of filled electron traps is in equilibrium. Depending on the relative location and distribution of traps the following cases arises. A) When traps are located close to the luminescence centers (First order kinetics) :

Let J be the intensity of excitation. Then the rate of electron raising to the excited state is proportional to J, and is given by CJ, where C is constant of proportionality. If there are n number of electrons in the N traps, then the rate of capture of excited electrons in traps is CJa (N - n), a being the probability of capture per trap. The rate of escape of electrons from traps is given by ns exp. (-E/KT). Under equilibrium condition, the rate of capture of excited electrons in traps and rate of electrons escaping from traps will be equal, and thus

CJa(N - n) = ns exp. (-E/KT) .. (3.11) from which we get

$$n = \frac{N}{1 + s \exp(-E/KT)} ... (3.12)$$

where A = CaJ is a factor directly proportional to the intensity of excitation.

B) When traps are located away from the luminescence centers(Second order kinetics) :

Let n be the number of trapped electrons and m the number of these in conduction band. The (m+n) is the number of empty luminescence centers and rate of electrons returing from the conduction band is given by,

 $\frac{dm}{dt} = 0 = CJ - \beta \quad m(m+n) + ns exp. (-E/Kt) - mb (N-m) .. (3.13)$ and the rate of electrons returning from the conduction band is given by ,

 $\frac{dn}{dt} = 0 = ns \exp(-E/KT) - mb(N-n) \qquad .. (3.14)$ where β and b are constants and CJ is the rate of electrons raising to the excited state.

If the excitation is not of an appreciably low intensity, the number of electrons in the conduction band is much greater than the number of trapped ones and thus $(m+n) \neq n$.

Hence $\beta m^2 = CJ$... (3.15) and therefore,

$$n = \frac{N}{1 + s \exp(-E/KT)}$$
 ... (3.16)

where $B = b \sqrt{CJ/\beta}$ and thus is proportional to the square root of the excitation intensity.

C) When the traps exist with complex distribution

If the form of specific distribution of traps is known, the derivations of equations (3.12) and (3.16) can be extended further to over complex trap distribution. In most practical cases, the number of traps N of different trap depths does not vary rapidly with E over most of the range of E values. Thus if N is assumed constant or slowly varying, the total number of filled traps from a given excitation intensity can be obtained. Let this number be n_m , then in most cases

$$n_{T} = \int_{0}^{m_{T}} n dE \qquad \dots (3.17)$$

Using the equation (3.12) we obtain

$$n_{T} = \int_{0}^{E_{max}} \frac{N}{1 + \frac{s \exp(-E/KT)}{A}} \dots (3.18)$$

For all practical excitation intensities $A \ll s$ and thus equation (3.18) yields on integration

 $n_T = NKT \log (A^* + 1)$.. (3.19) where $A^* = As^{-1} \exp(-Emax/KT)$ is proportional to the excitation intensity.

At high excitation intensities when $A^* > 1$, the number of filled traps will be a linear function of logarithm of the excitation intensities provided that A << s. When A approaches s,a saturation state will be reached. A similar logarithmic relation can be obtained by using equation (3.16) which includes consideration of retrapping.

3.2.3 Phosphorescence decay for different trap distributions

(Considering retrapping negligible)

In general a phosphor contains traps of various depths. If there are N number of traps with activation energies lying between E and E + dE then assuming retrapping to be negligible, the phosphorescence intensity I at time t after cessation of excitation is given as follows :

$$I = \int Ns \exp(-E/KT) \exp[-st \exp(-E/KT)] dE \dots (3.20)$$

Emin

$$= \frac{Ns}{exp.(s.t)} \int_{Emin}^{Emax} exp.(-E/KT) exp.[exp.(-E/KT)] dE ...(3.21)$$

Considering N to be constant over an energy range of 0 to infinity

the following three cases of interest arises according to the distribution considered.

A) Uniform trap distribution

If a phosphor contains an equal number of traps of all depths in the range of energy from sero to infinity then the distribution of traps is said to be uniform. Then integration of equation (3.21) gives

$$I = \frac{NKT}{t} (1 - exp.(-st)) ... (3.22)$$

If st >> 1, then

$$I = \frac{NKT}{t} ... (3.23)$$

= NKT t⁻¹
= constant t⁻¹ ... (3.24)

Thus the phosphorescence intensity (I) is inversely proportional to decay time (t) and a plot of I versus t^{-1} should be a straight line. Moreover plot of logI against logt should be a straigh line with a slope equal to -1

B) Quasi-Uniform trap distribution

The phosphors having number of traps N, which do not vary too rapidly with values of E in a complete distribution are said to possess quasi uniform trap distribution. Here the intensity of phosphorescence decay at any time t is proportional to the number of electrons in traps of mean life time τ given by

 $\mathcal{T} = s^{-1} \exp(E/KT)$. From equation (3.23) the product I t is found to be proportional to \tilde{E} . Thus a plot of the product I t against log t for a given phosphorescence decay will represent the variation of N with t and gives an approximate idea of the trap distribution.

C) Exponential trap distribution

If we have $N \ll \exp(-\beta E)$ as is found in some experimental cases, then for st >> 1, we have

I = constant $t^{-(\beta KT + 1)}$... (3.25) i.e. power of t varies linearly with the phosphor temperature, the constant β (always positive) can be found experimentally and approximately from the shape of the thermoluminescence curve(3).

When $\beta KT = 1$, the equation (3.25) is correlated with the bimolecular law and when $\beta KT = 0$, the case becomes of uniform trap distribution. i.e. reciprocal law holds good. Many phosphors, at room temperature, shows the decay of the type given by equation (3.25). With any trap distribution covering a wide range, the decay is influenced slightly by variation of temperature because, with rising temperature deeper traps supply electrons at the same rate at which shallow traps supply electrons at alower temperature. At constant temperature, on the other hand, at the time t most of the light emission by aphophor with continuous trap distribution is due to traps in which the electrons will have the mean life time T = t. 3.2.4 Retrapping of electrons during phosphorescence :

It is obvious that the escaping electrons from traps will always have a finite probability of being trapped before recombination with luminescence centres. The process of retrapping causes marked changes in phosphorescence characteristics (6)

Garlick (7), considering the effect of retrapping in monomolecular case obtained an expression for the intensity and is

$$I = \frac{n0^{2}s \text{ exp. (-E/KT)}}{N(1 + \frac{n0}{N} - \text{ St exp. (-E/KT)})^{2}} \qquad \dots \qquad (3.26)$$

Where N is the total number of traps and no is the number of empty centrs or filled traps initially. The equation can be further simplified considering the case no = N i.e. traps are saturated at the commencement of decay. This result in

$$I = \frac{N S \exp(-E/KT)}{[1 + st \exp(-E/KT)]^2} \qquad .. (3.27)$$

At long decay times (t >> 1) the equation (3.26) takes the form

$$I = \frac{N \exp(-E/KT)}{st^2} .. (3.28)$$

Thus for the traps of one depth only, retrapping process causes a fundamental change in the form of decay from exponential to hyperbolic (8). A more general treatment of phosphorescence due to traps when retrapping occures has recently been given by klasens and Wise (3)

3.3 RESULTS AND DISCUSSION :

3.3.1 Decay curves :

Figures 3.1 to 3.3 show some typical decay curves obtained for various samples with different concentrations of Mn and Sm. Initially intensity decreases very fast and then decreases slowly. The nature of the decay curves appears to be of the same general nature as reported for alkaline earth sulphide phosphors (10-14).

The first step in analysis of decay curves is to decide the mode of decay i.e. exponential, hyperbolic or power law decay.

To examine the possibility of simple exponential decay with single trap depth as given by equation (3.4) graphs of log I versus t are plotted. The plots are found to be non linear, indicating that decay is not exponential. However plots of log I versus log t are almost linear. Typical plots are shown in fig 3.4 to 3.8. This suggests that decay may be hyperbolic or power law. Thus the observed decay can be represented by the equation

$$I = Iot^{-D}$$
 .. (3.29)

where I is the intensity at any time t, Io is the intensity at the start of decay and b is the decay constant.

3.3.2 <u>Correlation coefficient</u> :

,

The degree of linearity between log I and log t is confirmed by evaluating the correlation coefficient (r) using the equation

$$\mathbf{r} = \frac{\mathbf{N} \sum \mathbf{x} \mathbf{y} - \sum \mathbf{x} \sum \mathbf{y}}{\left[\mathbf{N} \sum \mathbf{x}^2 - (\sum \mathbf{x})^2\right]^{\frac{1}{2}} \left[\mathbf{N} \sum \mathbf{y}^2 - (\sum \mathbf{y})^2\right]^{\frac{1}{2}}} \qquad (3.30)$$

Where $x = \log t$ and $y = \log I$ and N is number of observations. The magnitude of r signifies the closeness of the relationship while its sign indicates whether y increases or decreases with x. When the relation is linear, the value of r equals unity.

In the present study, the values of r for all samples are calculated (Table 3.1). The value of r for all samples is nearly equal to -1. This indicates that relation between log I and log t is close to linear. The negative sign of r indicates that intensity decreases with time.

3.3.3 Decay Constant :

The value of decay constant is a indicative of decay rate. Faster the decay, higher is the value of decay constant (b) and vice versa. It also provides the information about the relative population of traps at various depths.

The value of decay constant is calculated by the formula of least squares (15) using the relation

$$b = \frac{N \sum xy - \sum x \sum y}{N \sum x^2 - (\sum x)^2} \qquad \dots \qquad (3.31)$$

Where $x = \log t$ and $y = \log I$, N is the number of observations. The values of b calculated by using above equation (3.31) are shown in table 3.1. Values of b obtained from the **xxmux** slopes of log I - log t plots are also given in the same table. It seems that the values of b calculated by using equation (3.31) and that from graph are in good agreement with each other.

3.3.4 Variation of decay constant with activator concentration :

The variation of decay constant with activator concentration is shown in fig 3.9. From this figure is shown that value of decay constant. is almost constant and is not found to vary much with concentration. This indicates that activators probably do not create new traps but modify the relative importance of traps contributing to the phosphorescence decay. This observation is supported by TL studies (Chapter IV).

3.3.5 'Peeling off' of decay curves :

In the present investigation the decay can be represented by the equation $I = Io t^{-b}$. For hyperbolic decay b should be equal to 2 and for power law decay b should be equal to 1 for t >> 1. The value of b is neither 2 nor 1 but it is nearly equal to 1. This excludes the possibility of hyperbolic decay and power law decay resulting from traps of single depth. However the observed power law decay can be explained on the basis of monomolecular superposition theory suggested by Randall and Wilkins (3) and followed by other workers (14, 16, 17, 18). Such a decay results due to superposition of various exponentials corresponding to different traps and is expressed by the equation

$$I = Io t^{-b}$$

= Io₁ exp. $I_{-p_1}t$) + Io₂ exp. $(-p_2t)$ +
--- + Io_n exp. $(-p_nt)$... (3.32)

where Io_1 , Io_2 ,---, Io_n are the starting contributions to luminescence intensity by first, second,---, nth exponential component and $p_1 = S \exp(-E1/KT)$, $p_2 = S \exp(-E2/KT)$, ----, $Pn = S \exp(-En/KT)$ are the transition probabilities of an electron escaping from traps of depth E1, E2, ---- En respectively.

It is possible then to split each decay curve into a set of exponentials by the method suggested by Bube (19) and followed by others (14, 16, 18). In the present investigation all the decay curves were split in to three exponentials_ (fig 3.11 to 3.15). The activation energies corresponding to these exponentials are calculated by using the slopes p_1 , p_2 , p_3 of straight line on semialog plot between I and t. The values of E thus evaluated for slowest, middle and fastest exponentials are shown in tabke 3.2. The value of 3 used in above calculations is taken from the thermoluminescence studies carried on same samples.

3.3.6 Effect of decay activator concentration on trap depths:

From table 3.2 it may be seen that there is no significant change in trap depth with activator concentration. This suggestes that the activators do not introduce any new traps. The relatively small and unsystematic variation observed in different samples may be due to perturbation of trapping states (20, 21). The conclusion is consistant with that drawn from variation of b with activator concentration.

3.3.7 Distribution of traps :

The distribution of traps may be uniform, quasi uniform or exponential (22). For uniform trap distribution over a wide range, the relation between phosphorescence intensity I and time t for st >> 1 is given by

$$I = fo t^{-1}$$
 ... (3.33)

This relation is similar to $I = Io t^{-b}$. Thus for uniform trap distribution over a wide range should be unity. In the present investigation the observed values of b are close to unity (table 3.1). This indicates that trap distribution may be uniform trap distribution. For exponential trap

distribution b > 1 (23). In the present study be is nearly equal to 1. This rules out the possibility of exponential trap distribution.

The plot between I and t^{-1} is a straight line (fig 3.16). This also suggest that trap distribution may be uniform. The above conclusion is also supported by a plot between I.t and log t. This graph is also a straight line which is nearly parallel to log t axis. (fig 3.17).

3.3.8 Kinetics of luminescence :

The kinetics involved in the decay process may be either monomolecular (first order), bimolecular (second order) or intermediate. For a decay resulting from traps of single depth, the mnonmolecular process involves the exponential decay. The bimolcular process involves the hyperbolic decay. While power law decay results when process is neither first order nor second order. (24). For first order kinetics plot of I and t on semilog paper must be a straight line. For second order kinetics plot of recipocal of square root of intensity and time must be a straight line. Further for second order and intermediate order the slope of graph between log I and log t assumes the values of -2 and -1 respectively at large values of t. In the present investigation graph between I and t on semilog paper is not a straight line (fig 3.11 to 3.15) This indicates that kinetics is not monomolecular. Further the plot of $1/\sqrt{1}$ versus t is also not a straight line (fig 3.10) and slope of log I - log t is not equal to -2. Hence kinetics

is not bimolecular. However the decay can be represented by a power law equation of the type $I = Io^{-b}$. But the value of b is not exactly equal to as required by intermediate kinetics, which excludes the possibility of intermediated kinetics. On the other hand, the observed power law decay could be well explained on the basis of monomolecular superposition theory. This suggests that kinetics involved is likely to be monomolecular.

3.4 SUMMARY :

From the above study following inferences may be drawn :

- 1. The nature of decay for various emples is similar and may be expressed by an equation of the form $I = I_0 t^{-b}$
- Nature of decay is of power law type and can be explained in terms of superposition of various exponentials of monomolecular type.
- 3. The value of decay constant is nearly equal to unity. The distribution may be uniform.
- 4. The trap depths of effective levels are relatively insensitive to the concentration of activators (Mn, Sm)
- 5. The probable kinetics of decay process is likely to be monomolecular.

	Correlation	Decay Cons	
	coefficient (-ve)	by calculation (-ve)	from graph (-ve)
SD 12	1.007	0.9732	1.04
SD 13	1.005	1.0278	1.1
SD 14	0 .997	1.043	1.16
SD 15	0 .997	0.846	0.84
SD 16	0.999	1.114	1.1
SD 17	1.029	1.189	1.18
SD 18	1.06	1.377	1.24
SD 19	1.26	1.34	1.1
SD 20	0.99	0.871	0.88
SD 21	0.9	0.9948	1.0
SD 22	0.9947	1.0355	1.04
SD 23	0.9822	1.08	0.92
SD 24	1.018	1.04	1.02
SD 25	1.11	1.11	1.02
SB 26	1.005	0.954	0.96
SD 27	1.037	0.9217	0.92
SD 28	1.009	0.9313	0.95

<u>Table 3.1</u>: showing correlation coefficient and decay constant -obtained for various phosphors.

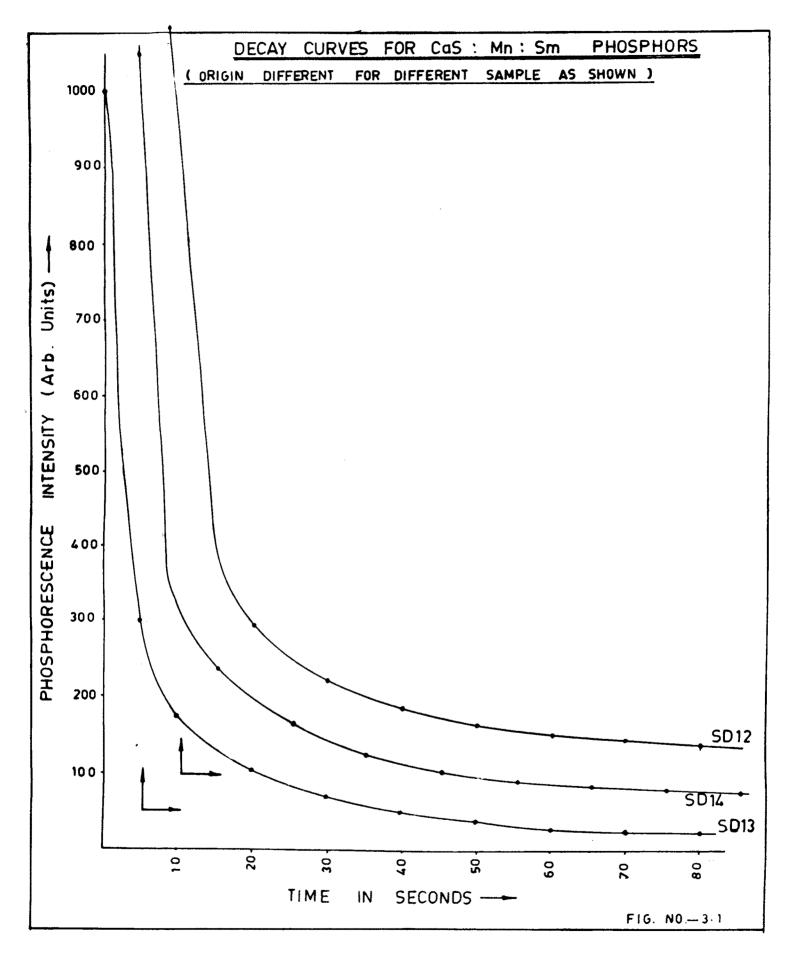
Sample no.	Values of trap depth from peeling off of decay curves		
	Slowest exponential E1	II exponential ^E 2	Fastest exponentia] E3
SD 12	0,5598	0.4631	0.4239
SD 13	0.565	0.4821	0.4321
SD 14	0 .5645	0.479	0.4324
SD 15	0 .5587	0.491	0.4252
SD 16	0.5589	0.5002	0.4292
SD 17	0.5587	0.4740	0.4255
SD 18	0.5589	0.482	0.4243
SD 19	0.5716	0.5071	0.4237
SD 20	0.5303	0.4533	0.4229
3D 21	0.5721	0.4846	0.4258
SD 22	0.5542	0.4630	0.4252
SD 23	0.5724	0.4655	0.4272
SD 24	0.5542	0•4783	0 .4258
SD 25	0 .5648	0.4821	0.4237
SD 26	0.5713	0.4817	0.4230
SD 27	0 .5643	0.4947	0.4242
D 28	0.5683	0.4885	0.4249

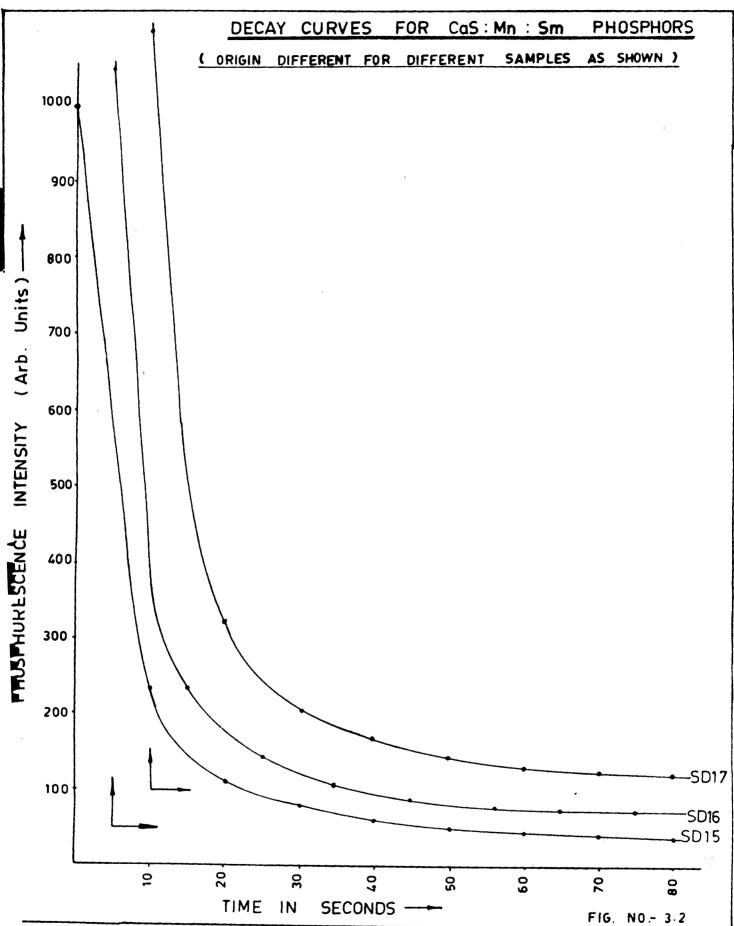
<u>Table 3.2</u> ; Showing activation energies calculated from peeling off of decay curves for different samples.

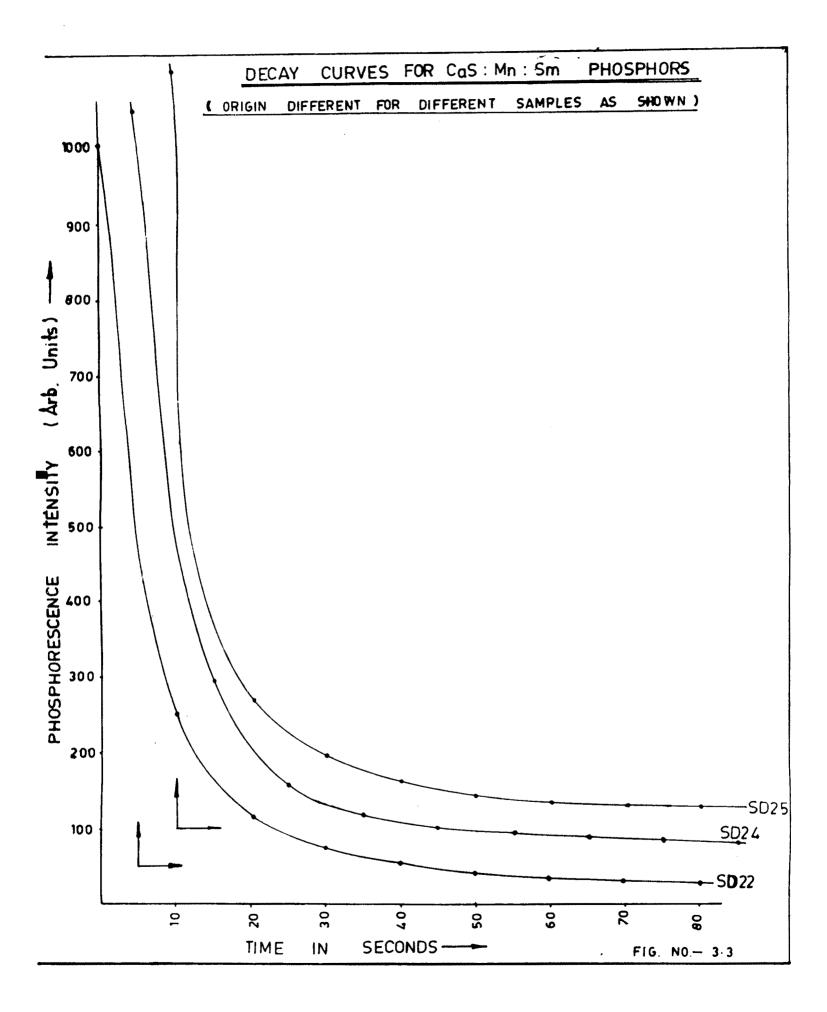
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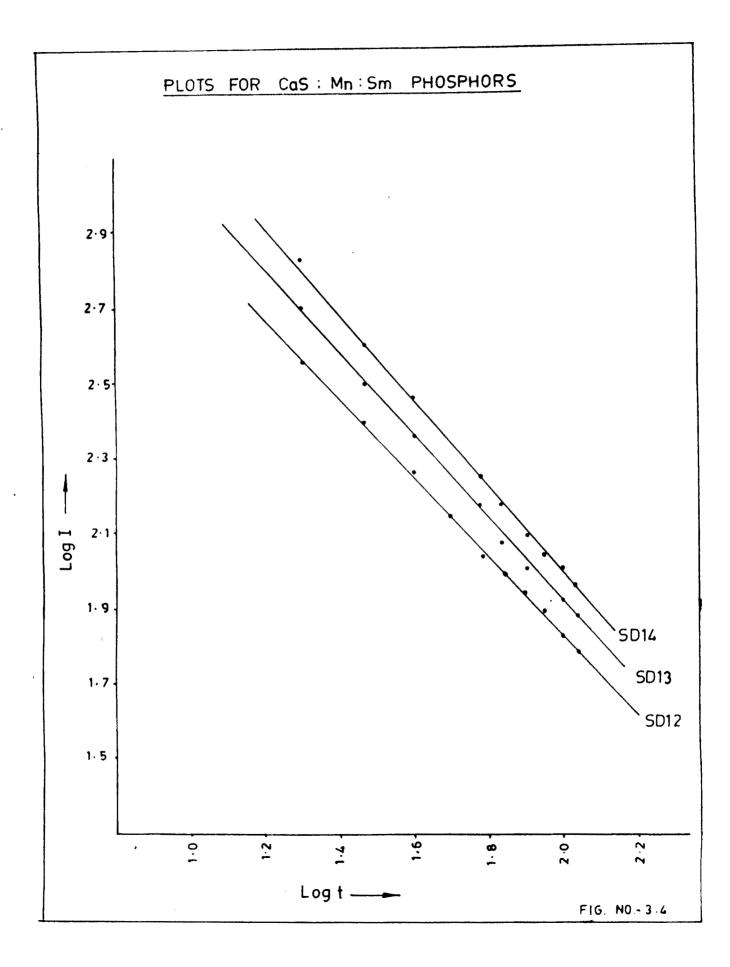
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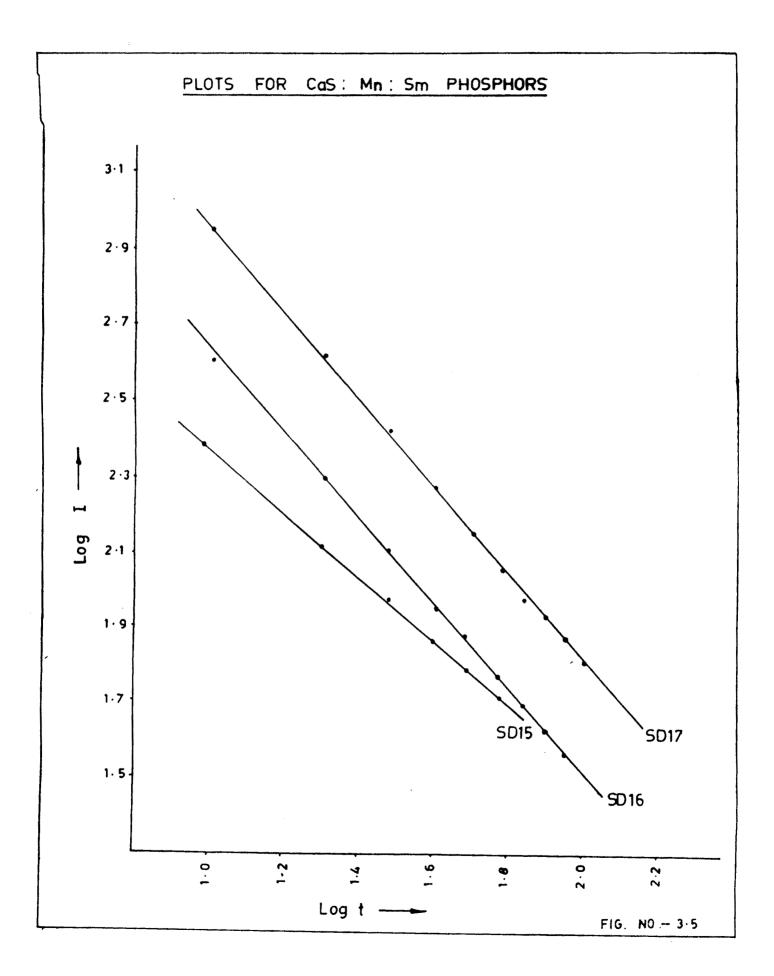
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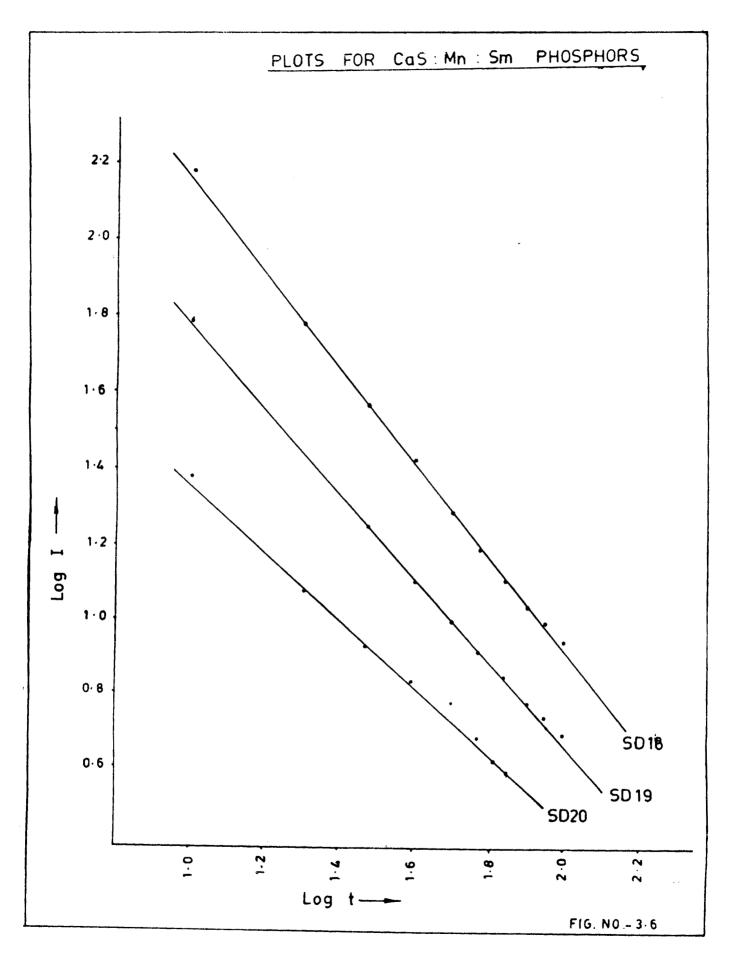


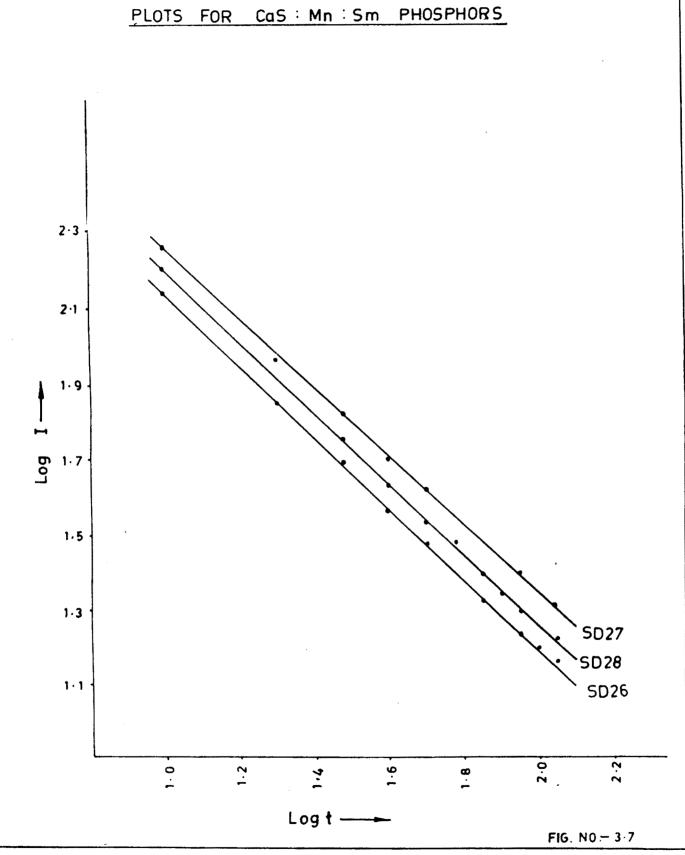


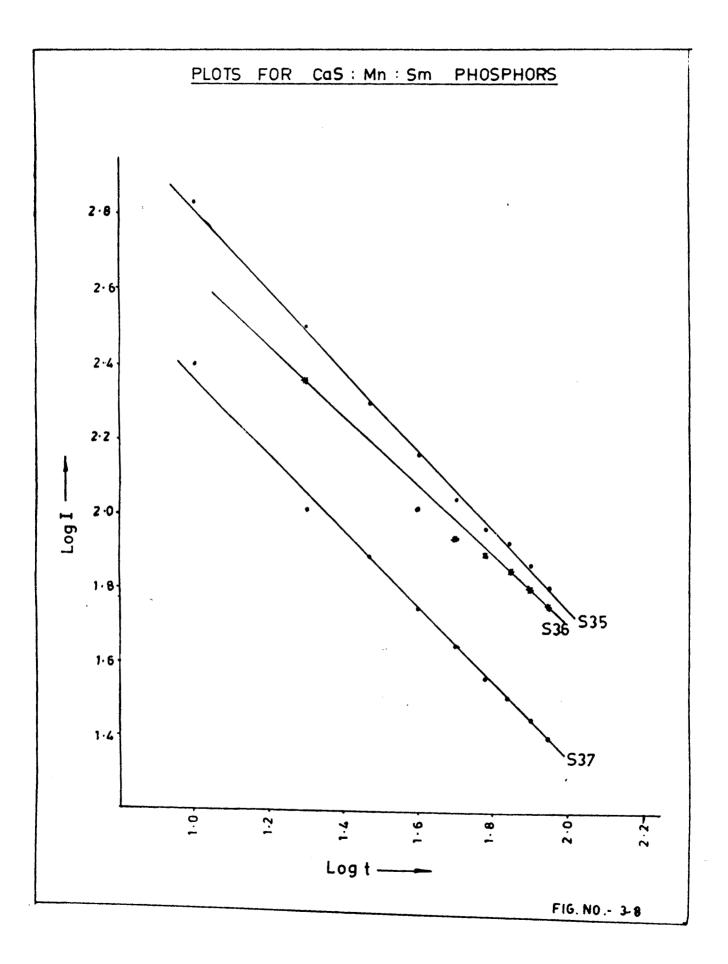






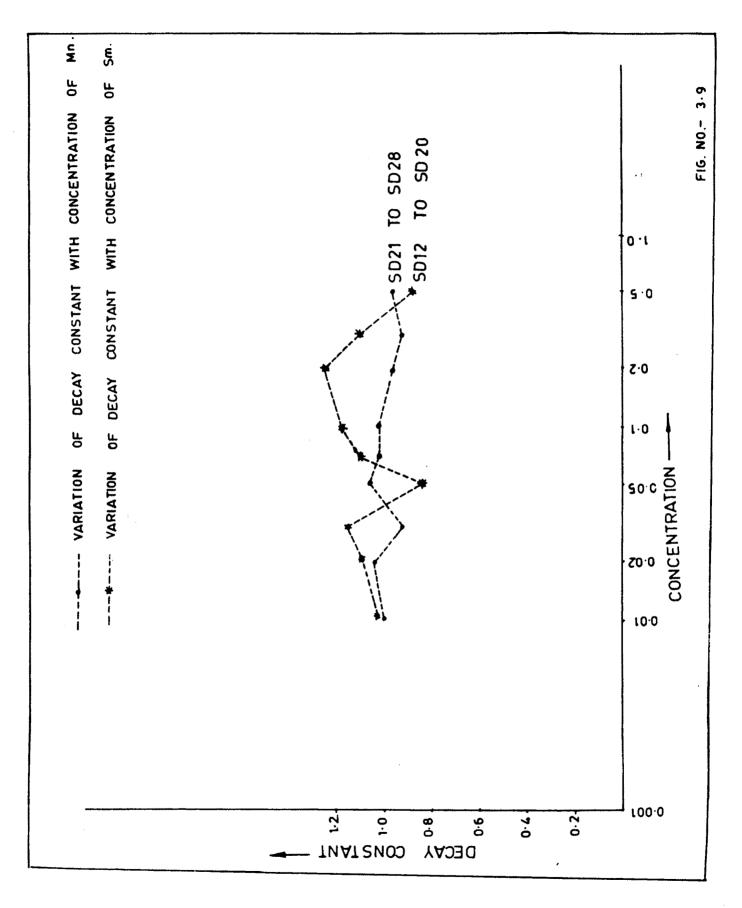


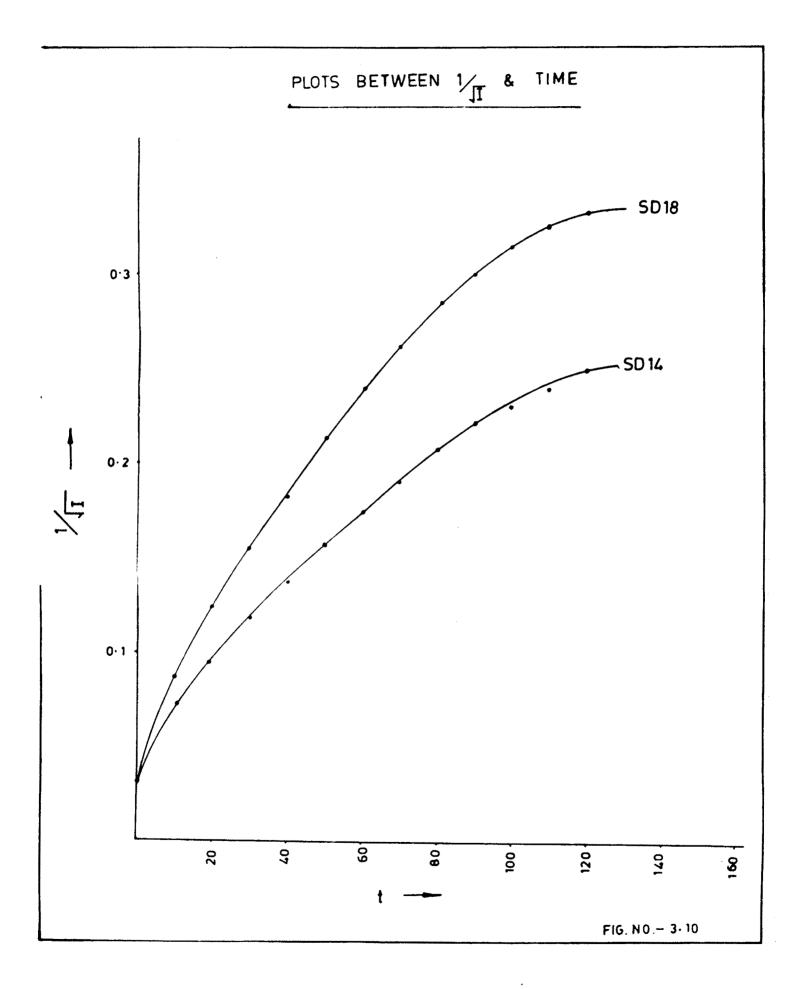


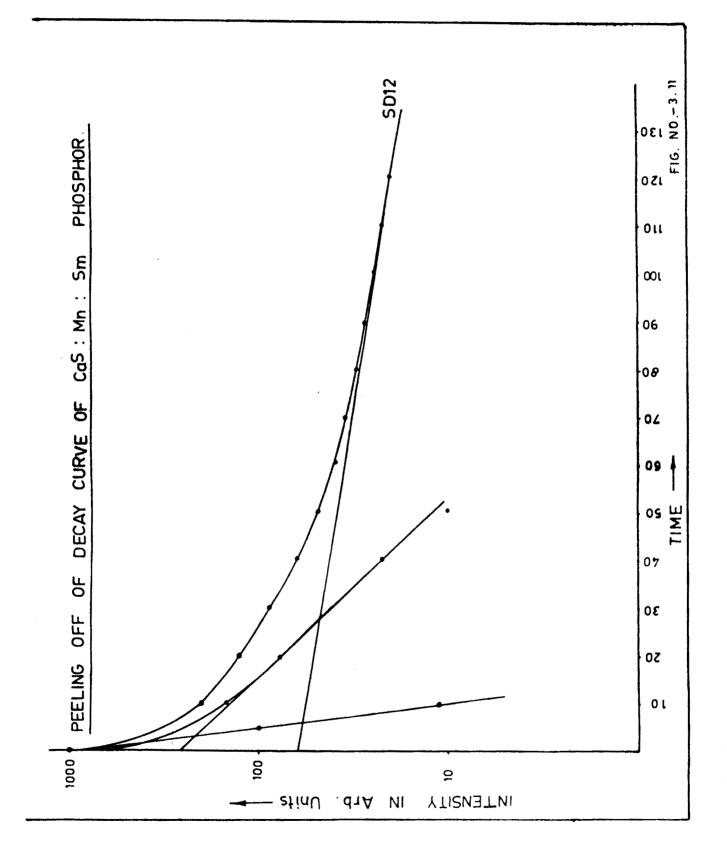


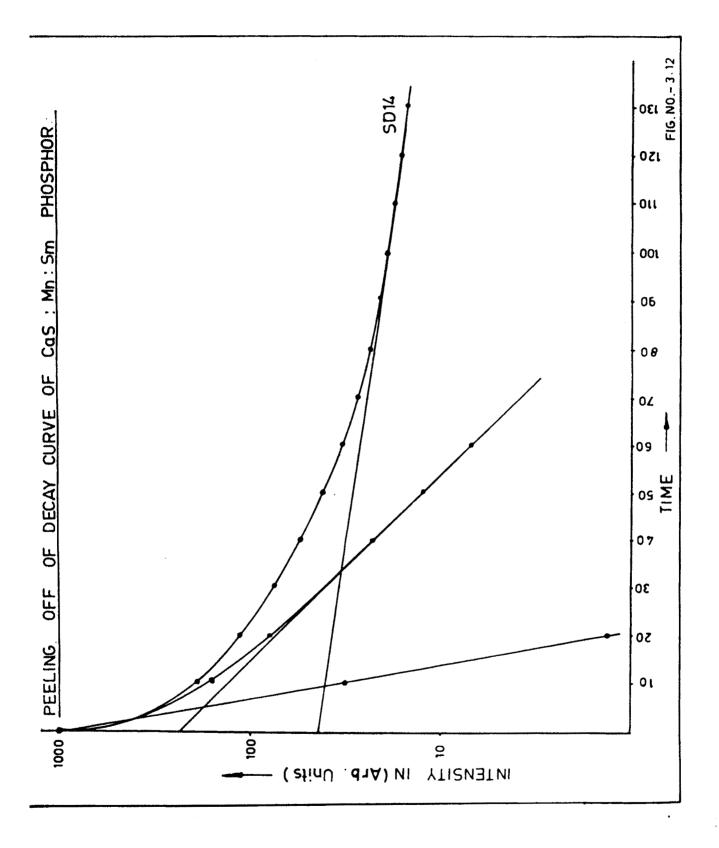
96

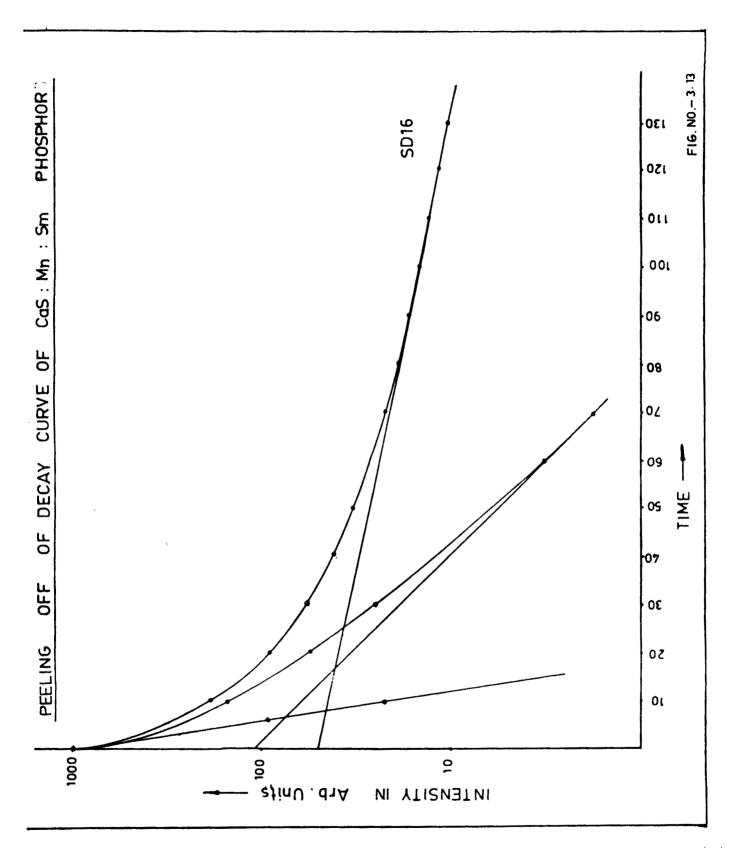
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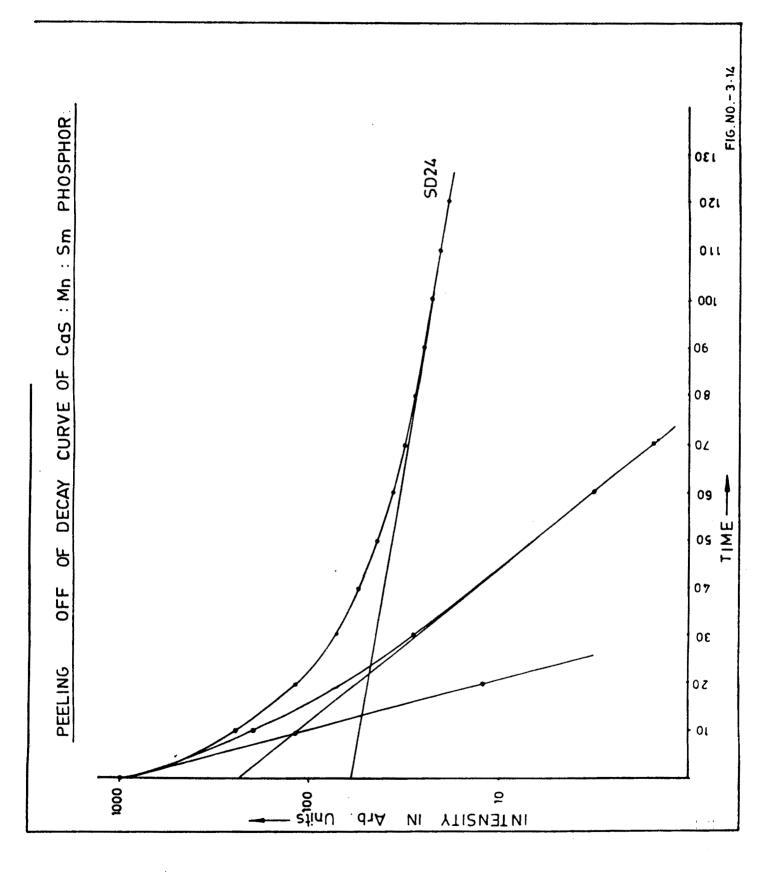


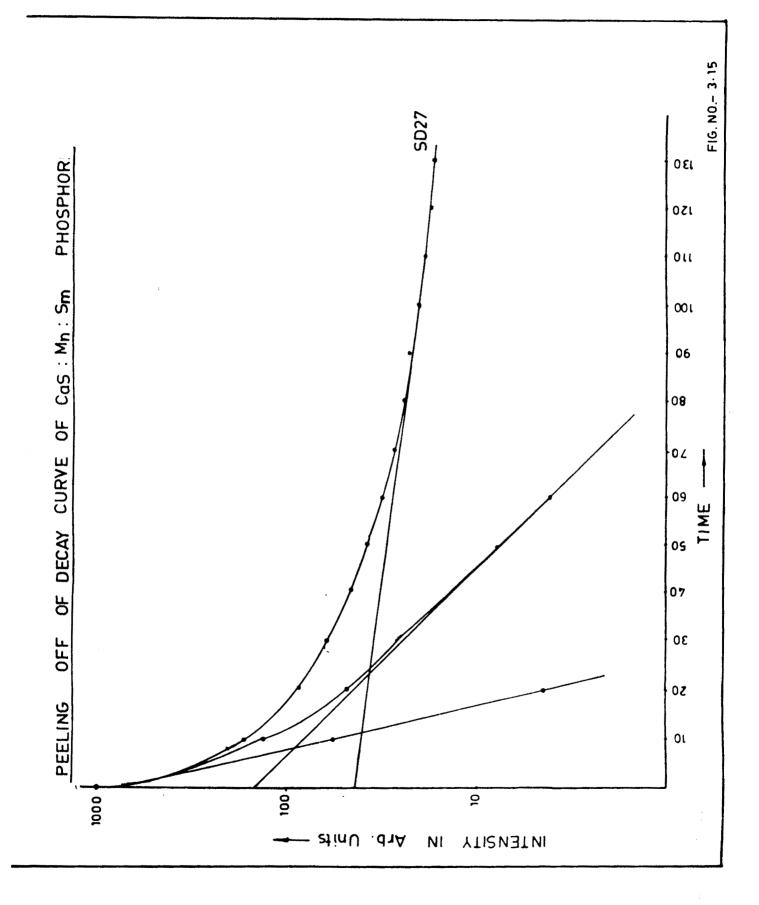


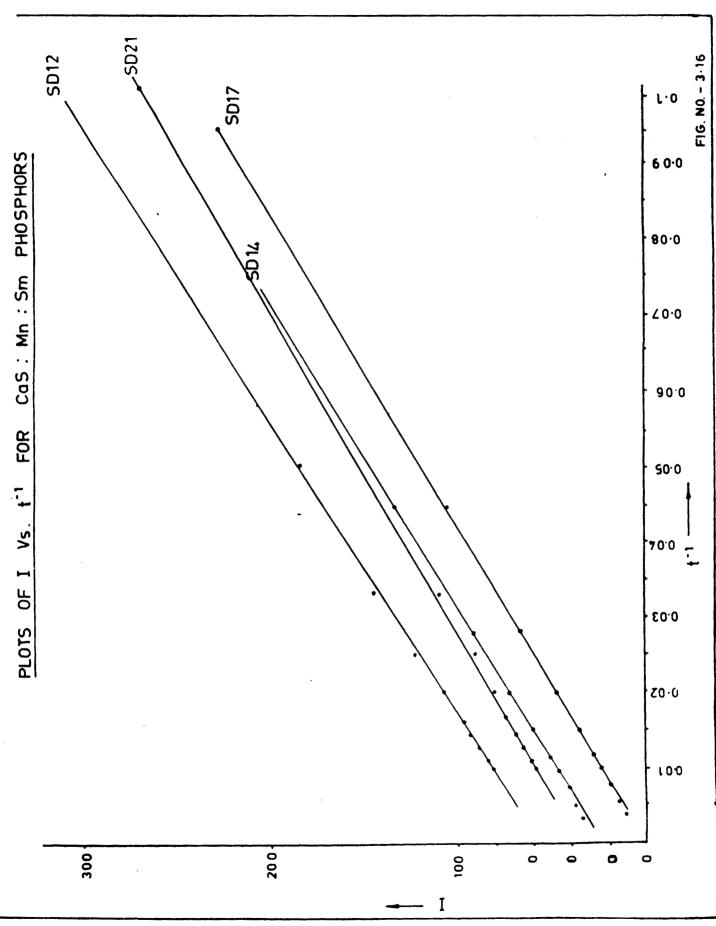












2400 ŞÐ21 INTENSITY X TIME 1000 1000 4 . . 2000 • SDIE 1200 800 400 1.6-1.8 60 Ö 1.0 1.2 -2.0. 1.4 . Log t -FIG.NO.- 3.17