X-RAY DOSIMETRY AND SOME NEW DOSIMETERS

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Supervisor's Certificate

The study and thesis on "X-ray dosimetry and some new dosimeters" carried out and presented herein by Shri Pratap Singh embodies original investigations. This thesis is submitted in partial,fulfilment of the requirements for the Degree of Doctor of Philosophy in the Department of Physics, Birla Institute of Technology and Science, Pilani (Rajasthan).

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Pratap Singh

(Pratap Singh)

DEDICATED
TO
MY PARENTS

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CHAPTER 1.

INTRODUCTION

INTRODUCTION:

The absorption of monochromatic light by matter sometimes produces unique primary products but this is not the case with ionizing radiations. Here a complex mixture of primary products is always formed[1-4]. Even with a single charged particle, on its first collision, a complex variety of primary products are possible. The reaction sequences are always complex, usually lengthy and often interdependent and in no instance, whether it be of physical, chemical or biological nature, are they adequately understood[5]. There are at least four consecutive stages in the response of a medium to irradiation. In the first physical stage, the absorbed energy is degraded to the atomic level and a number of diversely activated molecules is formed having conspicuously non-uniform spatial distribution. These are called primary products. They are unstable and in the physico-chemical stage, promptly undergo secondary reactions. Ultimately the system attains thermal equilibrium and there follows a chemical stage in which newly formed reactive species such as ions or free radicals react with each other. The biological process covers the response of the organism to chemical products on irradiation. In these primary processes no distinction is made between the effect of the incident charged particles or the effect of X-or gamma ray photons[6]. The quest for knowledge of the primary products rely chiefly on theory. Only scanty experimental informations are available . Practically all methods of observations are too slow to be able to detect the primary products. The quantum theory of stopping power, the Bethe theory, which has been extremely successful in accounting for the penetration in matter of energetic charged particles do not provide this information. The Bethe theory explains what happens to the particle or photon after penetration in the matter and is not concerned with what happens to the matter.

The processes by which high energy radiations and particles interact with matter have been described in detail whether the radiation is electromagnetic (X-rays or gamma rays) or corpuscular[7,8,9] (α or β). The final transfer of energy occurs by way of charged particles or excited molecules. The electromagnetic radiations transfer energy to materials by interaction with atomic nuclei or extranuclear electrons. These particles which interact with atom produce permanent changes in all materials, either directly or indirectly. The radiations that transfer energy to electrons generally do not cause extensive changes in materials such as metals and most inorganic compounds where electron rearrangements are often not stable. This is not the case in organic materials where the valence forces are mostly covalent in character[9].

In organic materials the electron dislocations and subsequent neutralization of the charged particles formed lead to energy rich fragments which react chemically to form compounds that differ from those originally present. Hence most organic materials are much more susceptible to radiation than other materials. These radiations interact with the materials according to the electron density of the substences and this parameter is essentially constant for low atomic-number atoms of which most organic molecules are composed of.

Although the exact mechanism of interaction or the sequence of reactions which follows the initial act is not yet completely understood, it is generally accepted that the following reactions are probably involved[10-14].

$M + \text{radiation} \longrightarrow e + M^{*}$ (excitated or normal) 1.

M^{*} (excited) $\longrightarrow R^{\bullet} + R'^{+}$ 2.

M^{*} (excited or normal) $+ e \rightarrow M$ (excited) 3.

M (excited) $\longrightarrow R^{\bullet} + R'$ 4.

Equation (1) represents the primary act of ionization leading to an excited or a normal ion. The excited ion, can then dismute, by virtue of its excess energy to yield two or more radicals, one of which may be ionized as in eqn.(2) . The excited or normal ion M^{*} will rapidly be neutralized, (eqn. (3)) leading to the original molecule M which is richer in energy by an amount, equal, at least to the charge neutralization energy. This excited molecule can then rupture to give two or more radical fragments as in eqn.(4). The excess excitation energy may be dissipated throughout the molecule and subsequently reconcentrated on any of the bonds present. If the energy can be stored in a relatively stable part of the molecule and slowly dissipated as degraded thermal energy, eqn. (2) and (4), may be greatly repressed. It is also conceded that eqn. (1) through (4) also occur when ionizing radiation interacts with organic materials in the liquid and solid state. The electrons ejected as a result of eqn. (1) are generally energetic enough to cause further ionization. Three cases may happen:

1. The incident photon will generally make only one collision with an electron and is either completely absorbed or is scattered out of the sample and gives rise to an electron which has the kinetic energy of the absorbed photon minus the binding energy of the electron in the orbit from which it was ejected. This type is the predominant process at low photon energies. Such a process is called Thompson Scattering.

2. As the photon energy increases, a small fraction of the photon energy is given to the electron depending upon the angle of scattering and the photon is deflected with the remainder energy. These electrons called Compton electrons have a higher specific ionization than the Thompson electrons.

3. The pair production phenomenon is observed at very high energies where a photon produces an electron-positron pair after annihilation. This does not come in our range of working.

RADIATION EFFECTS:

Radiation effects can again be separated into three categories. (1) Transient, which are due to excited and ionized electrons. (2) Displacement radiation effects, which are manifestation of the atoms displaced from normal lattice sites in crystalline solids and (3) Chemical radiation effects, which are due to molecular rearrangement occuring as a second stage to ionizing interactions.

I. THE TRANSIENT RADIATION EFFECTS:

These are concerned with the excitation including ionization and de-excitation of electrons. Since transient effects are associated with charges in electronic states, they usually produce significant changes only in the electrical and optical properties of the material and in most cases the perturbations are short lived and these are functions of primary radiation dose rate and they disappear soon after the irradiation ceases[15-16]. These effects have become an important problem in connection with electronic circuits which have to operate in space.

The types of perturbations produced in materials by excited electrons include the following:

In semiconductors the densities of majority and minority carriers are changed; this results in conductivity changes, a decrease in reverse impedence of rectifying junctions and generation of photo-voltages at junctions.

In insulators, secondary electrons are emitted from surfaces; these produce net charges on conducting elements and induce an internal space-charge distributions, change in dielectric conductance and polarisation currents. The production of free electron and holes in insulators is similar to that in gases which start conducting currents.

II. DISPLACEMENT RADIATION EFFECTS:

These involve the following physical manifestations[16], (i) increase in the electrical resistivity of metals, particularly at low temperatures as the result of the enhanced concentration of electron scattering centres, (ii) changes in the minority carrier life time, carrier mobility and effective doping of semiconductors as the result of defect stages, introduced in the forbidden energy gap, (iii) changes in mechanical properties of materials due to radiation-induced defects in the lattice and (iv) changes in the thermal conductivity of materials, as the result of lattice defects which act as photon and electron scattering centres.

III. CHEMICAL RADIATION EFFECTS:

These changes are brought about by the interaction of radiation with the atomic electrons producing free electrons and positive ions. The positive ions undergo secondary reactions including charge exchanges and ion exchanges.

The free electrons lose energy by inelastic and elastic scattering and may either recombine with positive ion or attach to neutral or negative molecules to produce negative ions.

If they recombine, the product may be chemically active free radicals, which will participate in secondary chemical reactions.

Many types of reactions can be initiated by radiation, among them are halogenation, polymerization, oxidation and changes in isomerism[10-11]. The reaction yields are called G-products (yield per 100 e.v.). Organic compounds, plastics and polymers which consist of carbon and hydrogen atoms bound together by covalent bonds are disrupted by radiation energy. In this respect they differ from metals which are primarily crystalline and do not generally contain covalent bonds and are not readily affected by radiation. All types of radiation will induce chemical changes in polymers[17-20]. As a result bonds are broken and new bonds are formed. Therefore, most of the radiation effects in these materials are irreversible. Radiation induced changes have their origin in the rupture of covalent bonds in organic molecules. Among radiation induced changes are those in appearance, chemical state, physical state and mechanical properties. Chemical changes include cross- linking, oxidative degradation, polymerization and gas evolution. Physical changes include effects on viscosity, refractive index, solubility, electrical conductivity, optical rotation, fluorescence, melting points, X-ray diffraction and other mechanical properties.

Radiation induced changes can be utilized for measuring radiation doses, which can ultimately be utilized for a quantitative assay of radiation effect. Therefore, a brief survey of earlier work on radiation dosimetry is presented below, covering solid system, liquid system and solid state dosimetry.

SOLID SYSTEM DOSIMETRY:

Solid as well as liquid systems have been applied for radiation dosimetry. The notable examples of solid dosimeters are the polyvinyl-chloride (PVC) plastic dosimeter by Artandi[21] (1960) who derived the absorbed dose from the colour change of the developed PVC after irradiation by measuring the optical density at 396 $m\mu$ on a spectrophotometer. The PMMA dosimeter by Whittaker[22](1967) in which the radiation induced increase in the U.V. absorption of this polymer is utilized for dosimetry[23,24,25] and the cinemoid colour dosimeter by Goldstein[26] (1966) in which the optical density changes in cellulose acetate films were related to the dose absorbed[27]. The other dosimeters in this series were the Thermoluminescence dosimeter of calcium fluoride by Attix[28] (1968) and others[29,30,31], the Hydrogen pressure dosimeter by Sheldon and Morris[32] (1966), photographic film dosimeter by Becker[33] (1966) and Baumagartner[34] (1960) and the 'n' or 'p' colar cell dosimeter by Muller[35] (1964). Amongst these the optical transmission density (O. D.) measurements are the straight forward in carrying out radiation effects dosimetry. These dosimeters use spectrophotometer. The accuracy of such measurements depends on a number of parameters such as the variation of response with radiation type, energy, dose rate, temperature and image stability. The reproducible spectrophotometer measurements depend greatly on calibration of the instrument, which includes accurate determination of wavelength, slit width, absorbance (O. D.) and path length. Measurable changes in plastic films require relatively high radiation doses (10^8 rads) and as such can not be accurately used for measuring X-ray doses which are of lower values. Other limitations are instability and non-linearity of response. Colour changes tend to show a linear optical density versus dose relationship over only a limited range and chemical changes are largely non-linear owing to the continued exhaustion of cross-linkage or cleavable bonds. Most glasses[36] are also used as dosimeters because they get darkened when exposed to high radiation doses and the spectrophotometric readings of this darkening are customarily used to interpret doses from 10^7 rads onwards. The changes in refrective index measurements have also been used to measure X-ray doses.

LIQUID SYSTEM DOSIMETRY:

Multitudes of other dose measuring systems[37-38] for ionising radiation specially for gamma ray dosimetry, have been characterised as chemical dosimeters because the reactions taking place in them result only in changes in the outer electron shells of the atoms involved. Any system in which a measurable change in a chemical property takes place upon irradiation may in principle be termed a chemical dosimeter. The dose absorbed in such a dosimeter is measured by analysing the quantitative change in a given parameter in the system. The Fricke dosimeter[37,38] was developed more than 40 years ago and is based on the oxidation of ferrous ions in a ferrous sulphate solution to ferric ions. The dose is derived from the measure of the optical density changes by a spectrophotometer. This dosimeter is still widely accepted as a standard in radiation dosimetry because of its accuracy and reliability. The other notable dosimeters in this catagory are the Ferrous cupric dosimeter by Bjergabakke[39] (1968), Ceric sulphate dosimeter by Hart[40,41] (1954- 58), Oxalic acid dosimeter by Holm et al[42,43] , Benzene water dosimeter[44,45,46,47] and the Ethanol chlorobenzene dosimeter by Brynjolfsson et al[48]. In these dosimeters the chemical reaction proceed linearly with dose i.e. the primary species from the radiolysis keep reacting with the solutes in the same way as long as the supply of the reacting constituents are not exhausted. Several other chemical dosimeters[49-81] have been developed depending upon the reaction mechanism of the particular dosimeter system. But these systems are far from ideal and so the search continues for the practical panacea for problems of dosimetery, one that is uniform, easily calibrated and capable of measuring absorbed dose, independent of spectrum, dose rate, temperature and other environmental conditions.

SOLID STATE DOSIMETRY:

Semiconductor materials function as electronic devices because of imperfections in their crystal lattice which provide mobile charge carriers when an electric field is applied[82-84]. The excess electrons and holes temporarily created in a semiconductor by exposure to ionizing radiation enhance the conductivity of the bulk material. The process is described as the generation of electron-hole pairs, producing a non-equilibrium state which is eliminated when the excess electrons and holes recombine. Several investigators[85-92] have reported formulations to describe radiation damage in junction transistors, diodes and other devices and bombarding experiments have been performed at several centres in U.S.A. Measurements in selected types of transistors were performed in the Battelle, 2- megawatt pool reactor in Texas. Measurements of electrical parameters were made before, during and after irradiation but no dosimetric study was done at all. Experimental data on diodes and other rectifiers indicated that irradiation invariably increased the forward resistance and decreases the switching time of those diodes. The behaviour in the reverse current usually increased with irradiation. Some 200 devices of the following type were tested:

1N212	(Hoffman and Semcor)
1N617	(Pacific Semiconductors)
1N210	(Hoffman and Semcor)
1N660	(Texas Instruments)
1N538	(General Electric Co.).

The aim was to detect in the device sample, the failure point which is defined as the exposure required to change those devices parameters such that they are outside specific tolerance limits for the particular application. Prelimimary irradiation experiments have been conducted on almost all solid state devices used in space vehicles. A full description has been given in a monogram by F. J. Reid[93]. A recent article by Masafumi and Nagai[94] has given the application of radiation damage to radiation dosimetry. Silicon planer n-p-n transistors of type 2SC33 were used by these physicists for the experiments and they were irradiated by gamma rays with a Co^{60} source of 1000 curies. Common emitter d.c. gain h_{FE} was measured because $1/h_{FE}$ was sensitive parameter to study surface damages of transistors[94,95]. The X-ray dosimeters making use of the changes in some parameters of solid state devices by exposure to X-radiation have not been developed much although a lot of work has been done on X-ray damage effects.

SYNOPSIS OF THE PRESENT WORK:

Organic and semiconductor materials are seriously effected by radiation[6-17]. The radiation effects in these materials may be classified into two types: transient and permanent[84]. Transient effects may be defined as change in operational properties that are noted during an irradiation but which disappear when the radiation field is removed.

Permanent effects begin during an irradiation but persist after it has ended. The scope of the present work is limited to these two effects only.

The second chapter gives the method of calibration of the X-ray unit with the help of the standard Fricke dosimeter[54-58]. The X-ray dose absorbed at a distance of 10 cms from the exit window by Fricke dosimeter is measured to be 10 rads/sec. The absorbed doses in other aqueous solutions of organic materials consisting of H, O, and C. atoms are calculated by Cheek and Linnebom (1960) method[96]. Thus dose- conversion factors for Xylose, Maltose, Galactose, Fructose, Arabinose, Camphoric acid and some Salts of amino-acids have been calculated and tabulated.

The third chapter deals with the effects of X-radiation on some physically measureable parameters of optically active substances[52] referred to in the II chapter. The effect of X-radiation on specific rotation or Molecular rotation of optically active substances has been measured by measuring the change in molceular rotation. The linear relationships between absorbed dose in rads and molecular rotation have been found to hold good in some of the optically active compounds. The values of the constants involved in the linear and sub-linear equations have been calculated and tabulated for the relevent substances. Thus the aim of dosimetric study has been achieved. The subsequent changes in the refractive index of D-fructose and L(+) Arabinose due to X-ray dose were measured and Born's prediction in 1920 that specific rotation was not the fundamental quantity but the relation of the form $\frac{[\alpha]}{(n^2+2)}$ where n is the refractive index was fundamental, is exprimentally proved to be true. The values of constant K for D-fructose and L(+) Arabinose have been worked out.

In the fourth chapter, X-ray dosimetric effects on some cellulosic materials in aqueous solutions have been studied. The very common and widely used celluloses have been selected for irradiation by X-rays. These are the sodium carboxy methylcellulose, methylcellulose, ethylcellulose and sodium alginate. These are used as emulsifying agents, in icecreams, custards, binders and fillers in tablets, emulsion paints, dairy products etc. These were irradiated and changes in flow rate through Ostwald's Viscometer versus irradiation time give a linear log-log graph. The reduced viscosity changes have been found reciprocal with the X-ray dose. The ionization current developed during irradiation has been found to have linear relationship with the dose absorbed by the cellulosic solution. Thus a new class of X-ray dosimeters has been developed.

In the fifth chapter X-ray dosimetric study of non-polar solution of certain tough polymers (Plastics) in organic solvents has been conducted. These are Polyvinylchloride (PVC) in cyclohexane, Polystyrene in carbontetrachloride and PMMA in carbontetrachloride. The study of the radiation induced conductivity in these polymer solutions is an attempt to investigate the relationship between dose, (R), ionization current, (I), produced and the collecting voltage, (V), across the ionization cell. An empirical relation of the form $I = (aV + b) R$, holds good in all these cases. The values of constants a and b have been tabulated. The absorbed X-ray dose (R) has been calculated by Fricke Dosimetry as in chapter II.

The sixth chapter deals with the changes in optical density[52] with X-ray dose of (1M) Levulose, (2.5M) Sucrose and (5M) Xylose at wavelengths 280 mμ , 565 mμ and 555 mμ respectively. All of these substances have proved to be good dosimeters. The dose calculation has been made on the basis of Cheek and Linnenbom method as given in chapter II. The changes in the melting points of certain sugars after irradiation by X-radiation were also measured and on the basis of this type of change, D-fructose, D-galactose and Sucrose were finally picked up for dosimetric study.

The seventh chapter deals with the effect of the irradiation on two thin film capacitors (Tantalum oxide and Mylar). Induced conductivity in solid dielectric films by X-radiation is utilised to develop thin film dosimeters.

The leakage current has been measured and a sublinear relation is found to exist between the dielectric conductance of the capacitors and the dose absorbed. These capacitors recover immediately to normal state after the exposure is closed.

The eighth chapter deals with the X-radiation effects on BEL (Bhart Electronic) and other indigenous solid state devices, with the aim to develop X-ray dosimeters. It has been investigated for the first time only in this work. It is well known that there are changes in the surface states of semiconductors due to X-radiation and this variation in surface states causes degradiation in various parameters of semiconductor devices. In this study, out of a multitudes of devices some diodes and transistors have been ear-marked to be used as very reliable X-ray dosimeters. Reverse biased leakage current was measured in the case of diodes and I_{cBO} and h_{FE} parameters in the case of transistors. DC-10, DR-25, DS-10, CD-26, CD-38 and CD-29 gave linear responses with the dose absorbed in various ranges and DR-10, CD-26 and CD-29 proved to be the best amongst this lot. The recovery in all these cases was immediate. In the case of transistors, the following were irradiated for dosimetric purpose:

AC-125, AC-187, AC-188 (all Germanium PNP), CIL-522, CIL-523, CIL-591, BC-108 and BC-109 (all silicon NPN).

Those were selected out of large number of irradiated samples because they recovered immediately after the exposure was over and gave linear or sublinear response with the X-ray dose at various bias conditions. AC-125 and AC-187 behaved in an unusual way. The sensitive parameter h_{FE} (transistor gain I_c/I_B) increased faithfully with dose while in majority of cases these parameters decreased. This has been explained in the text.. These transistors have proved to be the best amongst this lot of X-ray dosimeters investigated.

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CHAPTER 2.

RADIATION DOSIMETRIC UNITS AND CONVERSIONS

RADIATION DOSIMETRY UNITS:

Any property or effect of radiation physical, chemical or biological, which can be measured and related to an observable change, especially if the relationship is linear, is the basis of a potential dosimeter. Many sources of radiation have been used and various means of measuring radiation doses have been employed in studies on radiation effects. Dosimetric techniques include radiation induced chemical or physical changes and radiation transient effects on solid-state devices. In general two approaches have been made in measuring and reporting radiation exposures i.e. the description of the radiation field in terms of the exposure dose and energy absorbed by a sample- ie absorbed dose.

The exposure dose is measured in Roentgen. The dose which in 1 ml of air at N.T.P. (0.001293 gm) liberates ions carrying 1 e.s.u. of charge of either sign is called one Roentgen. and the absorbed dose of any ionising radiation is the energy absorbed by the material. If 100 ergs are absorbed by 1 g. of the substance, it is called 1 rad.

In the air, 1 Roentagen = 0.87 rads.

There are three commonly used units for measuring the energy absorbed in a given material.

(i) Ergs gm^{-1}

(ii) Rads, defined as 100 ergs gm^{-1}

(iii) ev gm^{-1} (electron volts per gm.)

These units are self explanatory and easily interchangeable. The ergs gm^{-1} and rad are commonly used by those working in the field of radiation effects. The absorbed dose is defined in that it allows one to describe the damage to a material directly in terms of the energy absorbed in the material regardless of the composition of the sample or the type of radiation field.

FRICKE DOSIMETER:

The ferrous ferric system was first proposed by Fricke and Morse[1] they suggested the use of ferrous sulphate, in air saturated 0.4 M H_2SO_4 as a method for measuring X-ray dose. The net effect is the oxidation of ferrous ion to ferric ion. The mechanism of the oxidation is not yet fully understood. Since the inception of the method, a number of modifications have been introduced in regard to the use and analysis of the solutions. Chloride ion was shown to suppress competing reactions involving organic impurities during oxidation of ferrous ion by ionizing radiation. Dewharst[2] subsequently showed that the chloride ions do away with certain added organic impurities in the system. These contributions eliminated the necessity of resorting to ultra-pure solvents and reagents in preparing the ferrous sulphate dosimetric solutions. This dosimeter is still used to standardise and calibrate other dose measuring devices as it is independent of dose rate over a wide range and has small temperature and pressure co-efficients of response with no dependence upon concentration of reactants and products or acidity changes which may occur during irradiation[3,4]. Besides this, the quality of radiation is not a determining factor in the response of the dosimeter.

The procedure for preparing a Fricke dosimeter is divided into the following three steps:

(i) Purification of the solvent (water): The purity of water used as solvent is a major consideration. Ordinary distilled water is redistilled thrice and finally it is redistilled from an acid dichromate solution to reduce the amount of organic impurities present.

(ii) Cleaning of the glassware: The glasswares are freed of organic matter by treatment with a strong acid oxidising agent consisting of concentrated sulphuric acid and some crystals of potassium dichromate and then thoroughly washing with distilled and subsequently with triply distilled water.

(iii) Preparation of the solution: The following quantities of the materials are used.

0.10 gm ferrous ammonium sulphate (A.R.).

0.015 gm sodium chloride (A. R.)

5.5 ml Conc. (98%) sulphuric acid (A. R.)

These chemicals were dissolved in purified water and the total volume was made 250 ml. The solution is then 0.8N (0.4M) with respect to sulphuric acid. To prevent the oxidation of ferrous ions by direct light the measuring flask, containing the above dosimetric solution is wrapped with carbon papers. Ten ml of the solution is filled in the sample tube and the tube is sealed. Ten such sample tubes were prepared for irradiation.

OPERATION OF THE X-RAY UNIT:

The 0.5 hp motor is started to circulate water around the X-ray tube and after some time the main switch is operated to start the current flow in the main circuit. The transformer plug which was originally kept at 0 ma is pressed on and the humming starts. After five minutes, the other plug is switched on and the transformer current is set at 10 mA and the plate voltage at 39 kv. The X-ray ($\lambda = 0.3\,\mathring{A}$) start coming out of the four windows. Initially the windows are kept closed. Sample tube is kept on the stand in front of the slit at a distance of 30 cms from the exit point. The slit is operated and simultaneously the stop clock is started and allowed the rays to fall on the solution for the required time. Thus ferrous ions will be converted into ferric ions according to time of irradiation. All samples were irradiated in turn for different timings and optical density for each sample was measured immediately by a Beckman DU-2 spectrophotometer at λ = 304 mμ keeping non-irradiated solution as a reference in a 0.5 cm cell. If $O.D_1$ is the optical density of the irradiated sample and $O.D_u$ for the unirradiated sample, then accoring to Fricke and Hart[5],

$$\text{Energy absorbed} = 5.253 \times 10^4 \;(O.D_1 - O.D_u)\ \text{Rads.}$$
$$(1\ \text{Rad} = 6.243 \times 10^{13}\ \text{eV/gm})$$

The results are given in the table 1 and are plotted in the graph. This graph gives a complete process of the conversion of Fe^{++} to Fe^{+++} in the presence of air at atmospheric pressure. In the begining for a few minutes the oxidation process is rapid. This is due to the presence of oxygen in the sample tube. But after a certain time the process becomes slow and the absorbed dose rate is linear with the irradiation time till the entire Fe^{++} is converted into Fe^{+++} and the absorbed dose rate becomes constant showing that the total Fe^{++} ions present have been converted to Fe^{+++} ions.

From the linear portion of the graph one can find out the X-ray dose received by the dosimetric liquid in a given time. Using the inverse square law of intensity, the calculated value of the dose received by this dosimetric solution comes to 10 rads per second when the sample is at a distance of 10 cms from the source of X-rays.

CALCULATION OF ABSORBED DOSE IN ONE SOLUTION FROM THAT IN ANOTHER:

The method given by Check and Linnenbom[6] applies to solution of materials composed of the elements H, C, N, O, F, Si, Cl and S.

The following procedure is adopted for calculation of the absorbed dose in an queous solution when the absorbed dose in Fricke dosimeter is known.

The incident photon energy corresponding to the wave length (λ) of the incident photon is calculated. In this case it is 0.0595 MeV.

Corresponding to this incident photon energy the absorbed dose per unit exposure (D_1) is written down from the table given by Check and Linnenbom.

Incident photon energy in MeV	Absorbed dose per unit exposure (D_1) in rads in		
	H	C	O
0.0595	0.916	0.610	0.913

The weight fraction (f_i) of the elements in the sample material in aqueous solution is calculated. Obtain D_i values for the elements in the sample at the incident photon energy from the table and substitute the values and calculate D_m (dose absorbed by sample material) from $D_m = \sum f_i D_i$. In Fricke's ferrous sulphate aqueous dosimeter, the weight fractions of H and O are 0.11 and 0.89. Hence $D_{H2O} = \sum f_i D_i$

$$= (0.11 \times .916) + (0.89 \times 0.913) = 0.901$$

As an illustration let us calculate the dose absorbed in Xylose ($C_5 H_{10} O_5$) if the dose absorbed in the corresponding time under similar conditions in Fricke Dosimetric Solution is known.

$$D_{xylose} = \sum f_i D_i$$

$$=(0.4 \times 0.610)+(0.0667 \times 0.916)+(0.533 \times 0.913)$$

$$= 0.791.$$

$$\frac{D_{xylose}}{D_{H_2O}} = \frac{0.791}{0.901} = 0.877$$

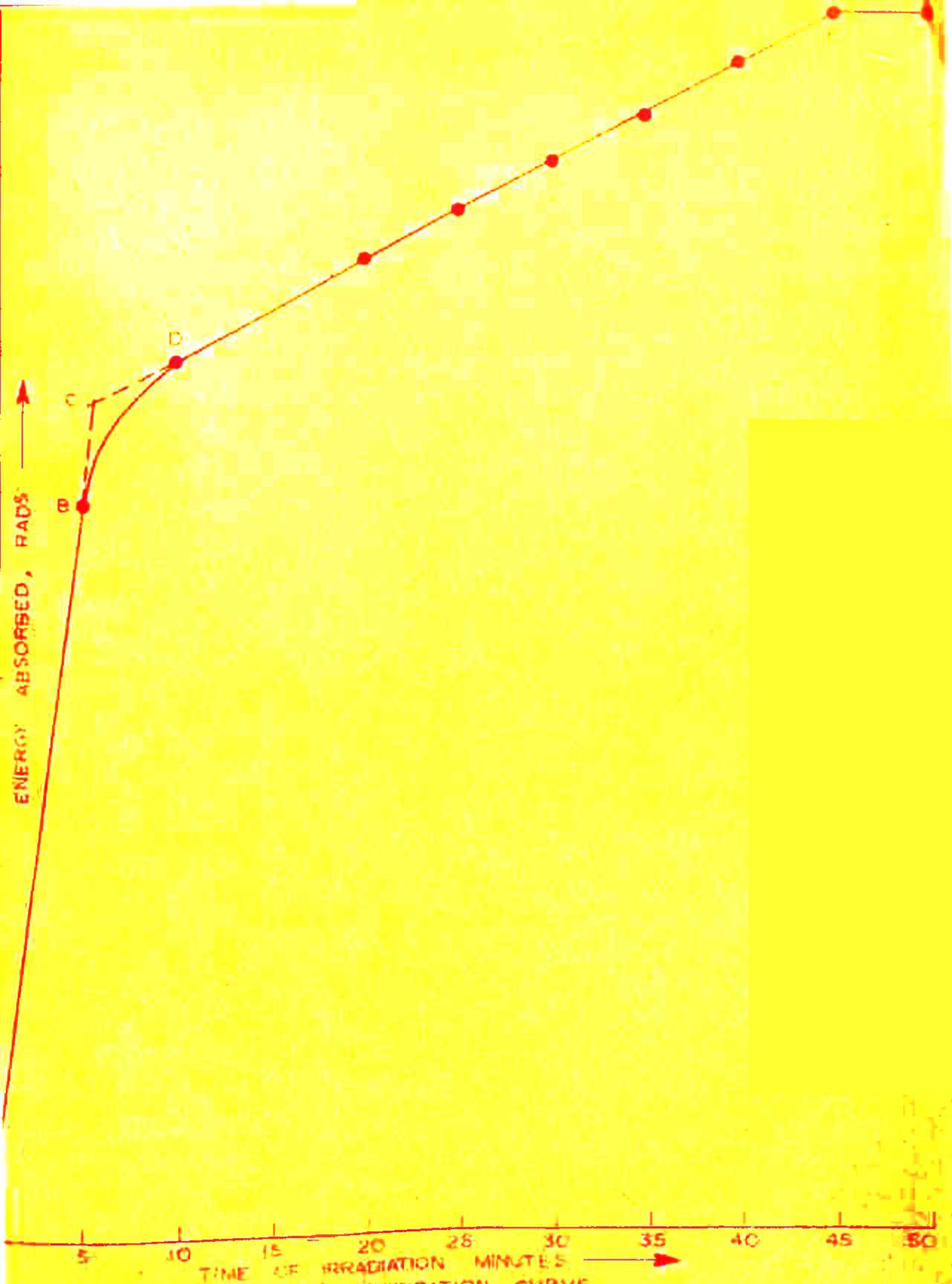
Absorbed dose in xylose ($C_5 H_{10} O_5$) = D_{H_2O} x 0.877

=(Dose measured by Fricke Dosimeter x 0.877)

The absorbed does for the following organic substances in aqueous solutions were calculated and are recorded in the table 2.

TABLE - 1.

Sample No.	Irradiation time in minutes.	$(O.D_1 - O.D_u)$	Dose Absorbed in Rads.	Dose Absorbed in eV/gm
1	5	0.015	787.95	4.918×10^{16}
2	10	0.018	945.72	5.902×10^{16}
3	15			
4	20	0.0200	1050.8	6.560×10^{16}
5	25	0.021	1103.34	6.884×10^{16}
6	30	0.022	1155.88	7.216×10^{16}
7	35	0.023	1208.42	7.539×10^{16}
8	40	0.0240	1260.96	7.870×10^{16}
9	45	0.025	1313.4	8.196×10^{16}
10	50	0.025	1313.4	8.196×10^{16}



X-RAY UNIT CALIBRATION CURVE

TABLE - 2.

Substance in aqueous solution	Multiplying factors to corresponding dose measured by Fricke Dosimeter.
Maltose	0.926
Xylose	0.877
Fructose	0.872
Sucrose	0.866
Galactose	0.872
Arabinose	0.877
Camphoric Acid	1.54
Arginine Monohydrochloride	0.810
Lysine monochloride	0.808

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CHAPTER 3.

X-RAY DOSIMETRIC STUDY OF OPTICALLY ACTIVE SUBSTANCES IN AQUEOUS SOLUTIONS.

X-RAY DOSIMETRIC STUDY OF OPTICALLY ACTIVE SUBSTANCES IN AQUEOUS SOLUTION:

INTRODUCTION:

The optical rotatory properties of molecular systems exhibit extreme sensitivity to very small structural changes[1-3]. The optical rotatory power of an asymmetric carbon atom depends upon the composition, constitution and configuration of its four groups. All carbon compounds which in solution rotate the plane of polarization, possess an asymmetric carbon atom[3-4]. The derivatives of compounds which are active in solution lose their rotatory power when the asymmetry of carbon atoms start disappearing. Hence by exposing the solution of an optically active substance to X-radiation the dissymmetrical order is disturbed, such that one kind of molecules exist in smaller number than the other. Because of the molecular structure the parts of the ions are not short straight lines but short helics twisted in the same direction. The X-radiation induces asymmetry which need not lead to an optical rotation of the same sign. The strengh and stiffness of the bonds between the atoms get modified by ionising radiation by an increase or decrease of rotatory power. The spatial configuration of the molecules also gets modified. There is correspondingly increase in the polarity of the solvent and it was proved by H. G. Rule[5] that the influence of the solvent tends to relate closely to its dipolemoment. It has been observed in most cases (sugars) that an increase in the polarity of the solvent decreases the rotatory power. In the case of the many samples investigation it was found that the specific rotation decreased faithfully with increasing X-ray dose.

Lowry and Dickson[2] proved that it was the polarity of the substituents rather than their masses which determine the magnitude of the molecular dissymmetry and hence degree of rotation. A molecular model of optical activity requires a connection between refractive index&bond structure. A new term electrical polarizability was defined by C. K. Ingold[3] which is the sensitivity to deformation by electrical fields. Molecular refraction, according to him is given by $R = (4\pi N_o/3) P$ where p is the electrical polarizability of a molecule, N_o is the Avagadro's number and R is molecular refraction. Gladeston Dales refraction equation is $R = (\frac{2}{3} \frac{MW}{\rho}) (n - 1)$, where MW is molecular weight. Hence the refractive index (n) is directly linked to electrical polarizibility. By passing ionising radiation into the solution of an optically active substance polarizability is induced and as such there are corresponding changes in the refractive index of the solution[7,8]. It was found that the fundamental quantity describing optical activity was not the specific rotation but as Born suggested[1], a quantity of the form $\frac{[\alpha]}{(n^2+2)}$ where n is the refractive index of the solution. The validity of this relation was tested in arabinose and fructose by passing measured X-radiation dose in the solutions and measuring $[\alpha]$ & n each time.

EXPERIMENTAL:

The optically active substances irradiated during the investigation are given as under. Doubly distilled water was used as the solvent.

L Arginine Monohydrochloride (Rudech and Dolder, Swi-tzerland) 1.93 gm/100 ml.
L(+) Lysine Monochloride, (Merck) 6.4 gm/100 ml.
D(+) Comphoric Acid (Merck) 0.62 gm/100 ml.
D(+) Xylose (Merck) 4 gm/100 ml.
Maltose, (Pifco Lab. Detroit(USA)) 2.5 gm/100 ml.
D(+) Galactose (Merck) 5 gm/100 ml.
D Fractose(Kyowa Hakhi Koayo Co., Ltd., Tokyo) 5 gm/100 ml.
L(+) Arabinose (Merck) 5 gm/100 ml.

Twenty ml of each solution was filled up in each pyrex glass sample tube. Four of the samples of a solution were exposed to 39 kv photon flux X-rays at a time, by fixing all of them in positions 10 cms in front of the windows. The irradiation periods of the relevent samples are given in the table 1. Optical rotation α and refractive index measurements were taken after each cycle of exposure and the same procedure was repeated in all cases. The results have been tabulated in the tables 1 and 2. Table 3 gives the relation between molecular rotation (Y) i.e. $\frac{MW}{100} \times [\alpha]$ and the X-ray dose (X) in kilorads, in the form of sub-linear and linear equations. The values of the various constants involved in the equations have also been tabulated. The measurements were taken on a Carl Zeiss Polarimeter and Refractometer (Jena 291947 Germany).

RESULT:

It is assumed that the changes in the molecular rotation and the subsequent changes in the refractive index of the optically active substances have been brought about by breakage of bonds not involving the asymmetrical carbon centre, thus producing a new optically active molecule[6,7]. These new optically active molecules now have changed induced polarizability. It is the polarity of the substituents rather than their masses which determined the magnitude of the molecular dissymmetry and hence the changes in the molecular rotations and refractive index. The study shows that linear and sublinear relations exist between change in molceular rotation and dose absorbed. L-Lysine, Galactose, Frudtose and Arabinose can very well be used as materials for dosimetric purposes. The validity of relation $\frac{[\alpha]}{(n^2+2)}$ has been tested for L(+) Arabinose and D-Fructose. The values of the constant k for the above two sugars have been calculated.

TABLE - 1.

Molecule	Conc. in water in (gm/100 ml)	Irradiation time in hours	Dose Absorbed in rads (kilo)	Specific rotation	Mol. rotation
L Arginine Monochloride	1.93	0	0	22.40	38.976
		3	87.48	20.15	35.061
		6	174.96	18.00	31.32
		10	291.60	15.23	26.502
		15	437.40	11.81	20.549
L(+)Lysine Monochloride	6.4	0	0	8.11	11.840
		2	58.176	7.656	11.169
		4	116.352	7.242	10.573
		8	232.704	6.463	9.421
		12	349.056	5.617	8.200
D(+)Comphoric	0.62	0	0	47.50	95.109
		2	110.880	44.35	88.80
		4	221.760	41.93	83.95
		6	332.640	40.32	80.73
		8	443.520	39.56	79.21
D(+)Xylose	4	0	0	19.3	28.975
		2	63.144	17.4	26.122
		4	126.288	16.9	25.3719
		6	189.432	16.6	24.9215
		8	252.576	16.35	24.546

Maltose	2.5	0	0	141.0	508.037
		2	66.672	135.0	486.418
		6	200.016	117.0	439.578
		10	333.36	109.0	392.737
		16	933.408	91.6	330.043
D(+)Galactose	5	0	0	79.9	143.856
		2	64.528	79.2	142.614
		4	125.568	78.5	141.372
		8	251.136	76.9	138.564
		12	376.704	76.1	137.016

TABLE - II.

Molecule D-Fructose- Cons. in water, 5 gm/100 ml.

Irradiation time in hours	Dose absorbed in kilo rads (x)	Specific rotation $[\alpha]$	Molecular rotation (Y)	Refractive index(n)
0	0	89.90	161.82	1.3382
2	64.528	88.57	159.426	1.3379
4	125.568	87.20	156.942	1.3376
8	251.136	84.42	151.956	1.3373
12	376.704	81.30	146.358	-

Molecule
L(+) Arabinose-Conc. in water, 5 gm/100 ml.

Irradiation time in hours	Dose absorbed in kilo rads (x)	Specific rotation $[\alpha]$	Molecular rotation (Y)	Refractive index(n)
0	0	108.9	163.485	1.3387
2	63.144	108.2	162.435	1.3385
4	126.288	107.6	161.4	1.3382
8	252.576	106.2	159.33	1.3380
12	378.864	104.8	157.245	-

For D-Fructose $\frac{[\alpha]}{(n^2+2)} = 23.2$

For L(+) Arabinose $\frac{[\alpha]}{(n^2+2)} = 28.5$

TABLE - III.

Molecule	Equation	Value of constants
L Arginine Monohydrochloride	$Y = mx + c$	$m = -4.15 \times 10^{-2}$ $c = 38.6$
L(+) Lysine Monochloride	$Y = mx + c$	$m = -1.01 \times 10^{-2}$ $c = 11.7536$
D(+) Camphoric acid	$Y = c_1e^{c_2x} + c_3$	$c_1 = 23.9$ $c_2 = -2.92 \times 10^{-6}$ $c_3 = 72.21$
D(+) Xylose	$Y = c_1e^{c_2x} + c_3$	$c_1 = 4.91$ $c_2 = -1.6 \times 10^{-5}$ $c_3 = 24.065$
Maltose	$Y = c_1e^{c_2x} + c_3$	$c_1 = 20.4$ $c_2 = -4.99 \times 10^{-6}$ $c_3 = 30.4$
D(+) Galactose	$Y = mx + c$	$m = -2 \times 10^{-2}$ $c = 143.5$
Fructose	$Y = mx + c$	$m = 4.11 \times 10^{-2}$ $c = -162.111$
L(+) Arabinose	$Y = mx + c$	$m = -1.642 \times 10^{-2}$ $c = 163.475$

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CHAPTER 4.

NON - POLAR SOLUTIONS OF CERTAIN PLASTICS IN ORGANIC SOLVENTS AS X-RAY DOSIMETERS.

NON-POLAR SOLUTIONS OF CERTAIN PLASTICS IN ORGANIC SOLVENTS AS X-RAY DOSIMETERS:

INTRODUCTION:

The study of the radiation induced conductivity in non-polar solutions of certain polymers has been done to investigate the relationship between the dose absorbed and ionization current produced, in order to develop a dosimeter. The radiation induced conductivity is an aspect of the energy transfer. The electron vacancy left after excitation or ionization can be taken as mobile and the molecule may be considered to have hole - conduction.

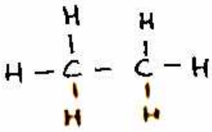
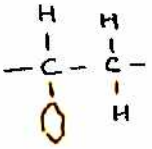
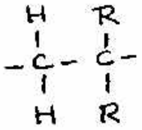
Plastics are primarily organic materials which consist of carbon and hydrogen atoms bound together by covalent bonds that are easily disrupted by the addition of radiation energy. All types of radiation will induce chemical changes in polymers. As a result old bonds are broken and new bonds are formed. Several reactions occur concurrently with radiation but the dominating reactions and the rate at which they proceed depend upon the chemical structure of the materials. The radiation damage to a polymer solution is generally dependent on total dose absorbed regardless of the type of radiation.

When a liquid is ionized which is contained between two electrodes with an electric potential difference applied between them, an ionization current will flow in the external circuit which is several orders of magnitude greater than the 'dark current' i.e. the current that would flow in the absence of ionization. When the ionization ceases this induced current decreases very rapidly at a rate which depends on the transit time of the slowest ions between the electrodes[1-2].

The ionization current versus the collecting voltage in a chamber filled with dielectric liquid after a non-linear intial rise, increases linearly with the applied voltage between electrodes[3]. After the steady state is reached the ionization current density I (amp/cm^2) for the same collecting voltage V, versus the ionizing radiation can be expressed by $I = (aV + b) R$ where R is the radiation intensity and a and b are constants which depend on the radiation kind, on the dielectric used and on the chamber geometry of the ionization cell. The effective values of the ionization current are obtained by subtracting from the measured current the values of the dark current carefully controlled before each measurement[4-5]. The conduction current in a non-polar liquid subjected to an electrical stress is de-pendent on many parameters other than the properties of the liquid itself. This makes it extremely difficult to obtain reproducible data and as a result there is still no general agreement on the mechanisms of charge motion. The principle problem is that of spurious charge generation[6]. Since the current is erratic and unless sufficient waiting periods is allowed before taking the reading, the sublinear relation between current and dose is not available. The small conductance current is enhanced by inducing conduction by irradiating the liquid with X-rays. The problem of measurement of the ionization current becomes difficult because the charge generation due to incident X-ray photons tends to occur in the bulk of the liquid rather than at a specified plane. If sufficient period is given, the current amplitude is found to be a function of the X-radiation dose R for a fixed voltage. As the voltage is increased for different sets of observations the probability of recombination decreases so that the conductance current is enhanced and no saturation is observed[7]. The potential gradient makes it more difficult for the positive and negative charge carriers, continuously supplied by irradiation, to recombine in the space charge region with the result that the over all recombination coefficient decreases and the number of free charge carriers is increased near the electrodes and so the induced current increases with time after application of an electric field[8].

EXPERIMENTAL:

The following polymers were dissolved in the solvents noted against each of them.

Polymer	Repeating unit in the structural formula	Solvent	Density of solution in gm/cc
Polyvenyl chloride (PVC)		Cyclohexane	0.732
Polystyrene		Carbon tetrachloride	1.615
Polymethyl methacrylate (PMMA)		Carbon tetrachloride	0.894

The ionization cell consisted of glass windows and parallel plate electrodes of bronze strips with 0.8 cm spacing between the electrodes. Exact parallelism was maintained by two teflon spacers. A thermocouple was inserted in a corner of the cell to keep guard of the temperature changes if any. The area of the electrodes (2.2 x 3.2) cm^2 was selected after preliminary observation to find the optimum size to avoid adsorption and keeping in mind the mean free path in order to keep track of the collisions. The usual experimental procedure was to connect the electrodes directly through a high resistor and to a high voltage power supply and the desired voltage was kept constant by manual control in the early stages of polymerisation reactions. After the constant value of ionization current at a particular voltage is recorded the voltage is turned off before the next readings. The electrodes are then short circuited because, current is often found to flow out of the sample. These polarization currents are very much existant in non-polar solution and the polarization charge is generally stored in the small volumes of the solution under elecrical stress and is recovered on short circuiting the test cell. The 'dark current' is measured for each set and is subracted from the observed ionization current to give the net ionization current for the voltage applied and for the given X-ray doses. After that the cell is cleaned, dried and filled by the fresh solution and made ready for the next exposure. The electric stress is now changed. The observations are recorded in tables 1 to 3. The values of the constants a and b are given in the table 4. The X-ray dose for any set is calculated by using Adamezewski's[9] formula. The following apparatus was used in this study:

1. Radart High voltage power supply with stabilizer, type 804A (Eastern Electronics, Faridabad).

2. D. C. microvolt meter, Phillips GM-6020
   (Two ranges 1 maga ohms and 10 mega ohms) least count 10^{-12} amperes.

3. X-ray unit strahlen-schutzzulassung PTB-508, Germany at 60 kv, 10 mA.

CONCLUSION:

All the plastics (Polymers) under investigation are being extensively used for radiation protection because they are radiation resistant to a good extent. In these materials the physical or chemical changes are less intense and the energy transfer covers a number of possible modes of reaction, from dissipation of the excitation or ionization energy without chemical change, to simple transfer of radicals. The production of ionization current, though in minute measures, is linearly related to the radiation dose rate for fixed values of the voltages applied. (Table 1, 2 and 3). If the constants a and b in the equation $I = (aV + b)R$ are found out for each polymer as given in table 4, calculation of X-ray dose is very simple. The oxygen dissolved in these solutions has practically no effect on dose absorption and hence these common and easily available polymers in solutions can be used as potential dosimeters for various ranges[9,10,11]. Polyvinylchloride in cyclohexane and polystyrene in carbontetra chloride (tables 1 and 2) are recommended for X-ray dosimetry while PMMA in carbontetrachloride is very well suited to measure very high gamma ray doses. The ionization current developed in this sample is rather insignificant in the case of X-radiation doses (table 3).

TABLE - I.

POLYVENYL CHLORIDE IN CYCLOHEXANE

Set Nos.	Constant voltages across the electrodes in volts	Net value of Ionization current in amp.	Calculated balue of X-ray dose rads/hr.
1	200	0.062×10^{-6}	25514
2	400	0.16×10^{-6}	62093
3	600	0.24×10^{-6}	88133
4	800	0.35×10^{-6}	125440
5	1000	0.43×10^{-6}	142500
6	1200	0.54×10^{-6}	170720
7	1400	0.63×10^{-6}	190360
8	1800	0.82×10^{-6}	237260
9	2000	0.94×10^{-6}	260910

TABLE - II.

POLYSTYRENE IN CARBON TETRA CHLORIDE

Set Nos.	Constant voltage across the electrodes in volts	Net value of ionization current in amp.	Calculated value of X-ray dose in rads/hr.
1	200	1×10^{-8}	2410
2	500	5×10^{-8}	11000
3	800	10×10^{-8}	20400
4	1000	13×10^{-8}	23300
5	1400	19×10^{-8}	33600
6	1500	20×10^{-8}	34600
7	1800	25×10^{-8}	40700
8	2000	27×10^{-8}	42200
9	2500	33×10^{-8}	47000

TABLE - III.

PMMA IN CARBON TETRA CHLORIDE

Set Nos.	Constant voltage across the electrodes in volts	Net value of ionization current in amp.	Calculated value of X-ray dose in rads/hr.
1	100	0.01×10^{-9}	38.7
2	200	0.10×10^{-9}	41.3
3	500	0.15×10^{-9}	48.2
4	600	0.16×10^{-9}	53.6
5	800	0.18×10^{-9}	57.2
6	1000	0.20×10^{-9}	60.5
7	1400	0.24×10^{-9}	66.2
8	1500	0.26×10^{-9}	67.5
9	1700	0.28×10^{-9}	72.4
10	2000	0.32×10^{-9}	77.9
11	2200	0.34×10^{-9}	79.7
12	2500	0.36×10^{-9}	79.8

TABLE - IV.

Solution	Value of a	Value of b
Polyvenyl chloride in cyclohexane	8.6×10^{-17}	3.4×10^{-13}
Polystyrene in Carbontetra-chloride	1.9×10^{-16}	5.44×10^{-13}
PMMA in Carbontetra-chloride	1.3×10^{-16}	3.53×10^{-13}

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CHAPTER 5.

X-RAY DOSIMETRIC STUDY OF SOME CELLULOSIC MATERIALS.

X-RAY DOSIMETRIC STUDY OF SOME CELLULOSIC MATERIALS:

INTRODUCTION:

Cellulose is an organic material which is least resistant to X-radiation. It is a polymer which consists of carbon and hydrogen atoms bound together by covalent bonds, which are easily disrupted by radiant energy[1,2,3]. The increasing interest in reactors and other devices that deal with high energy radiation and the desirability of employing fabricated polymeric materials in such installations demands that a great deal of information regarding the radiation stability of cellulosic materials be studied. The number of cellulose bonds cleaved in the solution increases linearly with dose. Hence cellulosic degradation can be a basis of a good and wide-range dosimeter. With this aim the following cellulosic polymers have been selected for irradiation by X-rays:

1. Carboxy Methyl Cellulose (CMC)
2. Methyl Cellulose (ether)
3. Ethyl Cellulose (ether)
4. Sodium Alginate (Sodium salt of Alginic Acid).

It has been found that effects are small in the case of simple organic compunds but are much more pronounced in polymers and amongst the above category, the bond cleavages have been rather speedy. Some physical changes arising due to the bond rupture such as viscosity, conductivity, and flowrate have been taken up for investigation. These are all due to radiative degradation of these polymers.

1. CARBOXY METHYL CELLULOSE: (CMC)

This cellulose is widely used as a detegrent in textile printing paste, drilling fluid, emulsion paints, oil and grease resistant papers, cotton fabrics of all types, ice- creams, custards, pudding, dairy products and pharmaceuticals. It is the widely used water soluble cellulose derivative and is prepared by the reaction of sodium chloroacetate with alkali cellulose. A number of products of differing properties is obtained depending upon degree of substitution and degree of polymerization or the chain length.

The variation of specific viscosity with $_pH$ for a dilute solution of CMC has been studied by K. Mienes[4], Berlinsteglitz and Swan[5,6] demonstrated the high viscosity in dilute solutions of CMC at $_pH$ 7 by a linear plot of $\eta_{sp/c}$ against very low values of concerntration c. The constants for relating intrinsic viscosity, to molecular weight were obtained by Sitaramaiah and Goring[7]. The effect of temperature on the viscosity of CMC solution has been studied by the Hercules Powder Co., Willington, Del., U.S.A. in 1963.

Here is intended to study the changes in its flow rate through the capillary of an Ostwald viscometer after irradiation with X-rays. As the compound forms part of most finished cotton fabrics, it was thought worthwhile to irradiate it and study its resistance to X-radiation and changes in viscosity, which are the main factors in its use as antiredeposition agent. Here the effect of radiation on polymer solutions in water was studied because the diffusion of adjacent macroradicals is facilitated in a solvent. If the solution is fairly dilute, the average distance between two molecules will be much larger than in the bulk state so that the chances of cross-linking of two different molecules after chain scission are decreased and the relative inportance of degradation will be increased in an irradiated solution of the polymer.

It has been found experimentally that the number of polymer bonds cleaved in a polymer solution are independent of both molecular weight and polymer concentration but increases linearly with dose. Paul Y. Feng[8] has given a relationship among initial and final average molecular weights versus radiation. Accordingly

$$\frac{Mf}{M_o} = \frac{N_o}{N_o + PR}$$, M_f = final average molecular weight.

M_o = initial average molecular weight.

N_o = initial number of polyer molecules.

P = no. of polymer bonds cleaved per unit dose.

R = total radiation dose.

The average molecular weight is also related to the intrinsic viscosity of a polymer

$[\eta_i] = KM^a$ where K and a are constants.

Also $\underset{\text{Limit } c \to 0}{[\eta_i]} = (t/t_o - 1)/c)$ where t_o is the flow time for the solvent in the flow type viscometer and t is the corresponding time for the solution.

Therefore $\eta_{io} = KM_o^a$

and so $\eta_{if} = KM_f^a$

$$= K\ (M_oN_o/N_o + PR)^a$$

$$= \frac{\eta_{io}N_o^a}{(\ N_o + PR)^a}$$

A log- log plot of η_{if} or $\frac{\eta_{if}}{\eta_{io}}$ against $(N_o + PR)$ should be a straight line.

If concentration of the solution remains constant, $[\eta_i]$ becomes approximately proportional to $(\frac{t}{t_o} - 1)$, it follows that if t_r is the flow time for the solution at dose r and t_u is the flow time at zero dose, a log-log plot of $(\frac{t_r}{t_o} - 1 \Big/ \frac{t_u}{t_o} - 1)$ against dose should be linear, where

t_r = flow time for solution at dose r
t_u = flow time for solution at zero dose.
t_o = flow time for water.

As dose is proportional to the irradiation time, the graph plotted converts the whole system to a very good dosimeter.

EXPERIMENTAL:

Five gms of CMC was dissolved in 250 ml of doubly distilled water. Ostwald type viscometer was used for viscosity measurements, the temperature was maintained at 25°C. The viscometers were cleaned with fresh, filtered chromic acid solution and throughly rinsed and dried before each run. When not in use they were kept filled with chromic acid solution. In all five runs were taken for t_r at different times. The X-ray plant was kept at 39 kv. The current was adjusted to 10 mA to avoid unnecessary heating of the plant although water was kept circulating round the plants by a 0.5 H.P. motor. The sample was pipetted in pyrex glass irradiating cells and 5 such samples were put for irradiation. The results of these runs can also be expressed in terms of reduced viscosity.

$$\eta_{(\text{ reduced })} = \frac{1}{C(\eta/\eta_0 - 1)}$$

where C is the concentration of CMC in gm/100 ml of solution, η is the viscosity of the solution and η_o is the viscosity of water, the solvent. The ratio η/η_o is equal to the ratio of the outflow time of the solution to that of the solvent. Finally by using an ionization cell with plain parallel electrodes the ionization currents produced by X-ray doses in the CMC solution at a fixed voltage were measured and making use of I. Adamazeuski[9] relation linking the ionization current with radiation doses, the X-ray unit was calibrated in terms of ionization current versus radiation dose inorder to utilize CMC for radiation dosimetry. Calculated values are due to computer. The values are recorded in table 1 and 2.

RESULT:

The study shows that CMC gets degraded appreciably and the degradation represents cleavages of actual linkages. A graph η (reduced) versus irradiation time (Fig. 2) shows that the reduced viscosity does not go to infinity but approaches a limiting value and the increase in reduced viscosity with increasing radiation dose indicates an accelerating effect on loop- opening reactions. The rupture of many weak linkages starts at mild doses and as the dose increases more and more, the rupture of stronger linkages starts slowly and slowly. The Fig. 1 and 3 show that a linear relationship can be used as dosimeter for X-ray doses.

TABLE - 1.

Carboxy-methyl Cellulose (CMC)

t_u = 57.3 sec. C = 2 gms/100 ml.

t_o = 23 sec. Temperature = 20°C

Sr. No. of runs	t_r in sec.	Log $\left[\frac{\frac{t_r}{t_o}-1}{\frac{t_u}{t_o}-1}\right]$	Irradiation time T in hours	Log T	Calculated value of η (reduced)
1.	55.3	-0.0235	1.3	0.1139	0.356
2.	53.6	-0.0482	2.0	0.3010	0.375
3.	51.3	-0.0820	4.0	0.6021	0.406
4.	48.6	-0.1255	9.5	0.9777	0.449
5.	46.6	-0.1612	20.0	1.3010	0.487

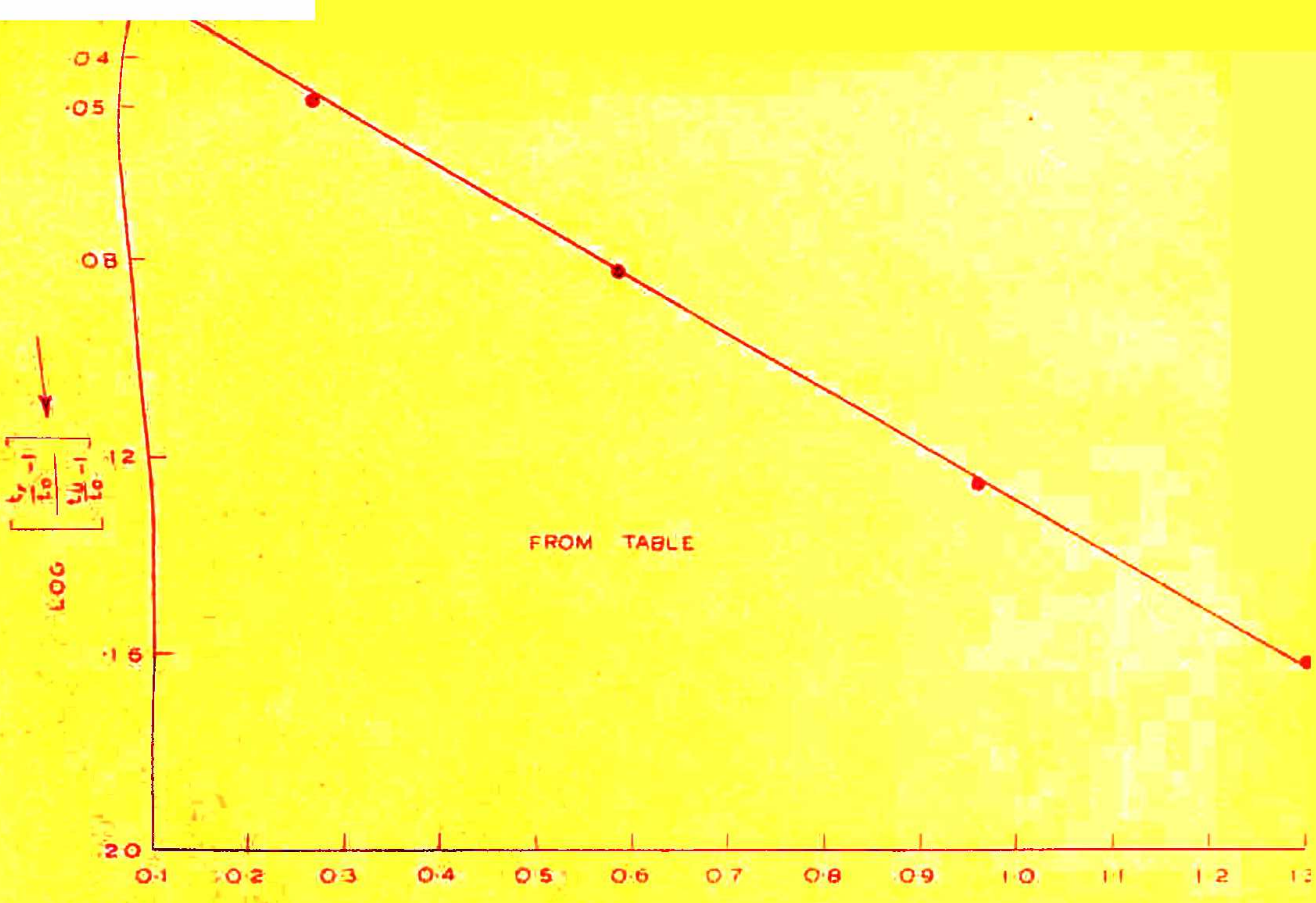

FIG 1 VISCOUS FLOW-RATE CHANGES IN RADIATIVE DEGRADATION OF CARBOXY METHYL CELLULOSE SOLUTION.

TABLE - 2.

Carboxymethyl Cellulose (CMC)

Ionization current in Amperes	Calculated values of X-ray dose in rads per hours.
2×10^{-6}	50.00×10^{4}
5×10^{-6}	125.00×10^{4}
7×10^{-6}	174.00×10^{4}
9.5×10^{-6}	234.00×10^{4}
12×10^{-6}	293.00×10^{4}
15×10^{-6}	364.00×10^{4}
17×10^{-6}	410.00×10^{4}
19×10^{-6}	455.00×10^{4}
21.5×10^{-6}	511.00×10^{4}

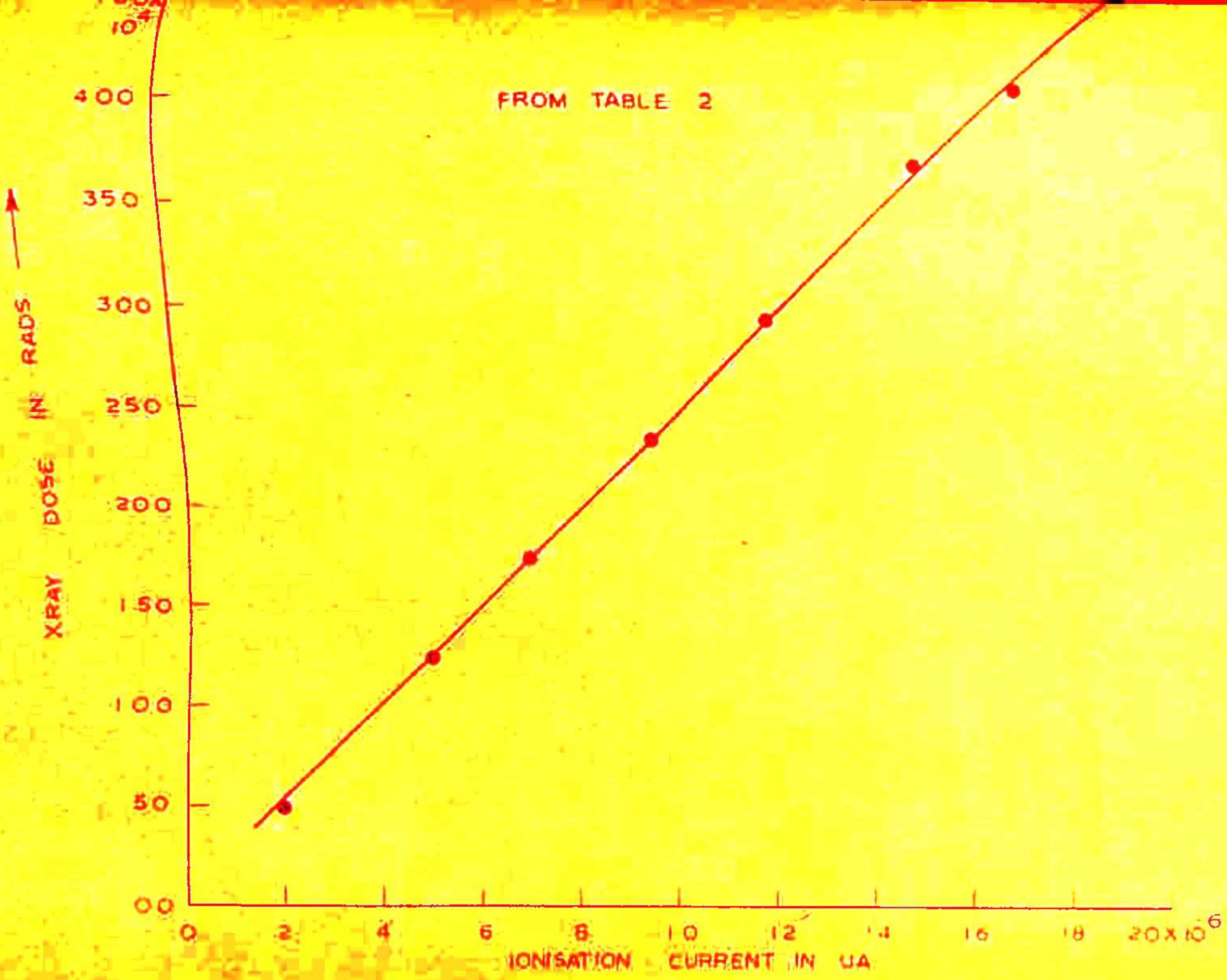

FIG 3 RADIATIVE IONISATION OF CARBOXY METHYL CELLULOSE.

2. METHYL CELLULOSE:

Methyl Cellulose is methyl ether cellulose and is soluble in cold water and disperses in it forming a viscous colloidal solution. It is added to about one third the required amount of boiling water. When the powder is thoroughly hydrated, the remainder of the water, preferably in the forms of ice, is added and stirred until it is homogeneous. It is insensitive to pH.

EXPERIMENTAL:

The same procedure as in the first sample (i.e. caboxymethyl cellelose) was adopted in this case. This time two different sets of observations were taken, first at X-ray plant at 39 kv and the second at 46 kv. The current was kept fixed at 10 mA. Flow readings for t_r were taken after 2, 5, 7 and 9 hours (table 3). The same procedure was repeated with higher doses at 46 kv (table 4.). The irradiation time was also enhanced upto 12 hours. The same ionization cell was used for measuring the ionization currents at constant voltage for different doses (table 5).

RESULTS:

The study of the irradiated methyl cellulose in water shows that this cellulose can very well be used as a

TABLE - 3.

Methyl Cellulose

Temperature 20°C t_u = 69.5 sec. X-ray operation at 39 kv
t_o = 22.5 sec. C = 1 gm/100 ml

Sr. No. of runs	t_r in sec.	Calculated value of $\text{Log}\left[\frac{\frac{t_r}{t_o}-1}{\frac{t_u}{t_o}-1}\right]$	Irradiation time T in hours	Log-T	Calculated value of η (reduced)
1.	58.7	-0.1135	2	0.3010	0.6215
2.	47.2	-0.2789	5	0.6990	0.9109
3.	42.8	-0.3645	7	0.8451	1.108
4.	40.7	-0.4125	9	0.9542	1.230

TABLE - 4.

Methyl Cellulose

Temperature 20°C

t_u = 69.5 sec.

t_o = 22.5 sec.

X-ray operation at 46 kv

C = 1 gm/100 ml.

Sr. No. of runs	t_r in sec.	Calculated value of $\text{Log}\left[\frac{\frac{t_r}{t_o}-1}{\frac{t_u}{t_o}-1}\right]$	Irradiation time T in hours	Log T	Calculated value of η (reduced)
1.	57.8	-0.1238	2	0.3010	0.6373
2.	48.9	-0.2503	4	0.6021	0.8523
3.	41.2	-0.4001	8	0.9031	1.203
4.	38.4	-0.4711	12	1.0792	1.415

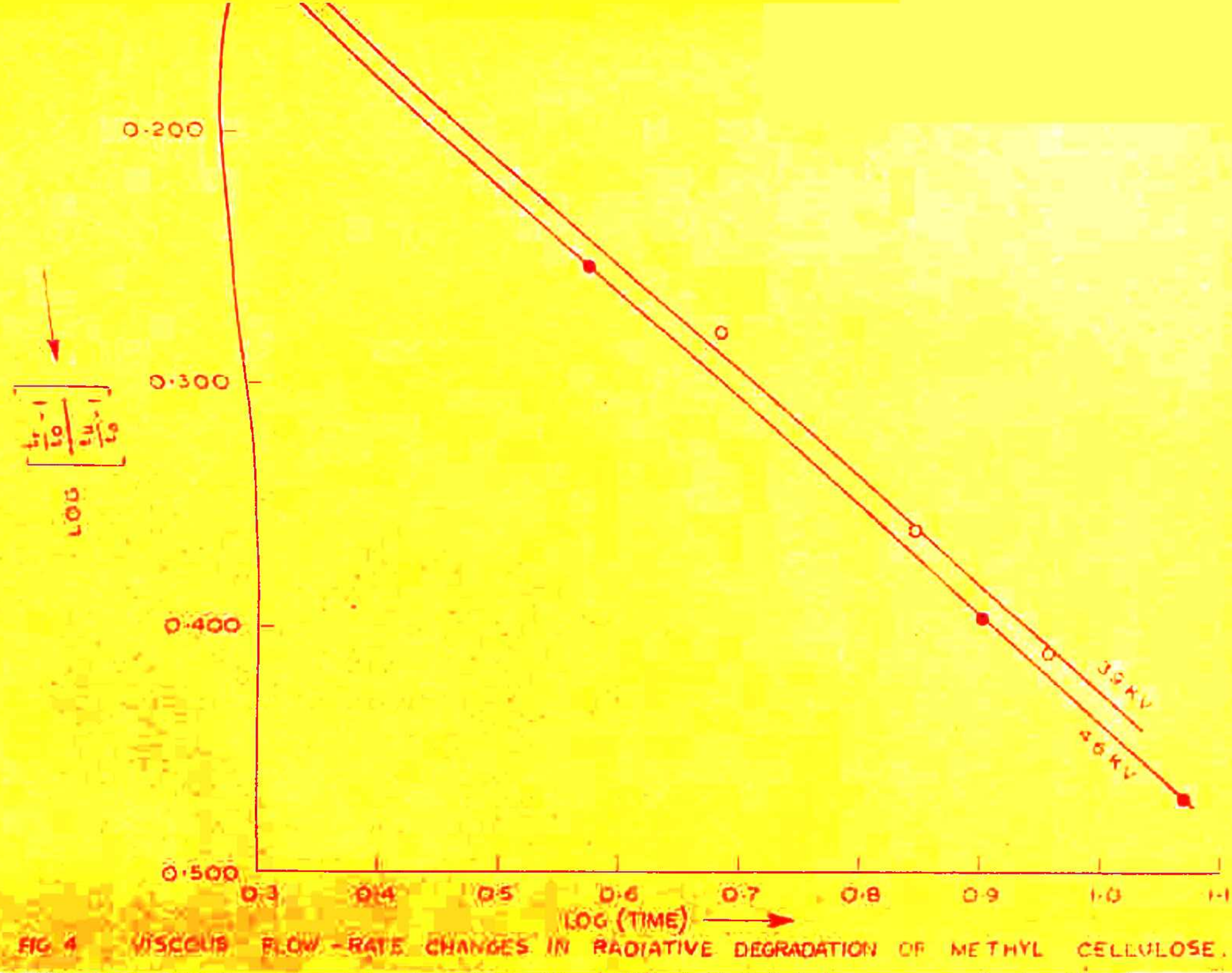

FIG 4 VISCOUS FLOW-RATE CHANGES IN RADIATIVE DEGRADATION OF METHYL CELLULOSE.

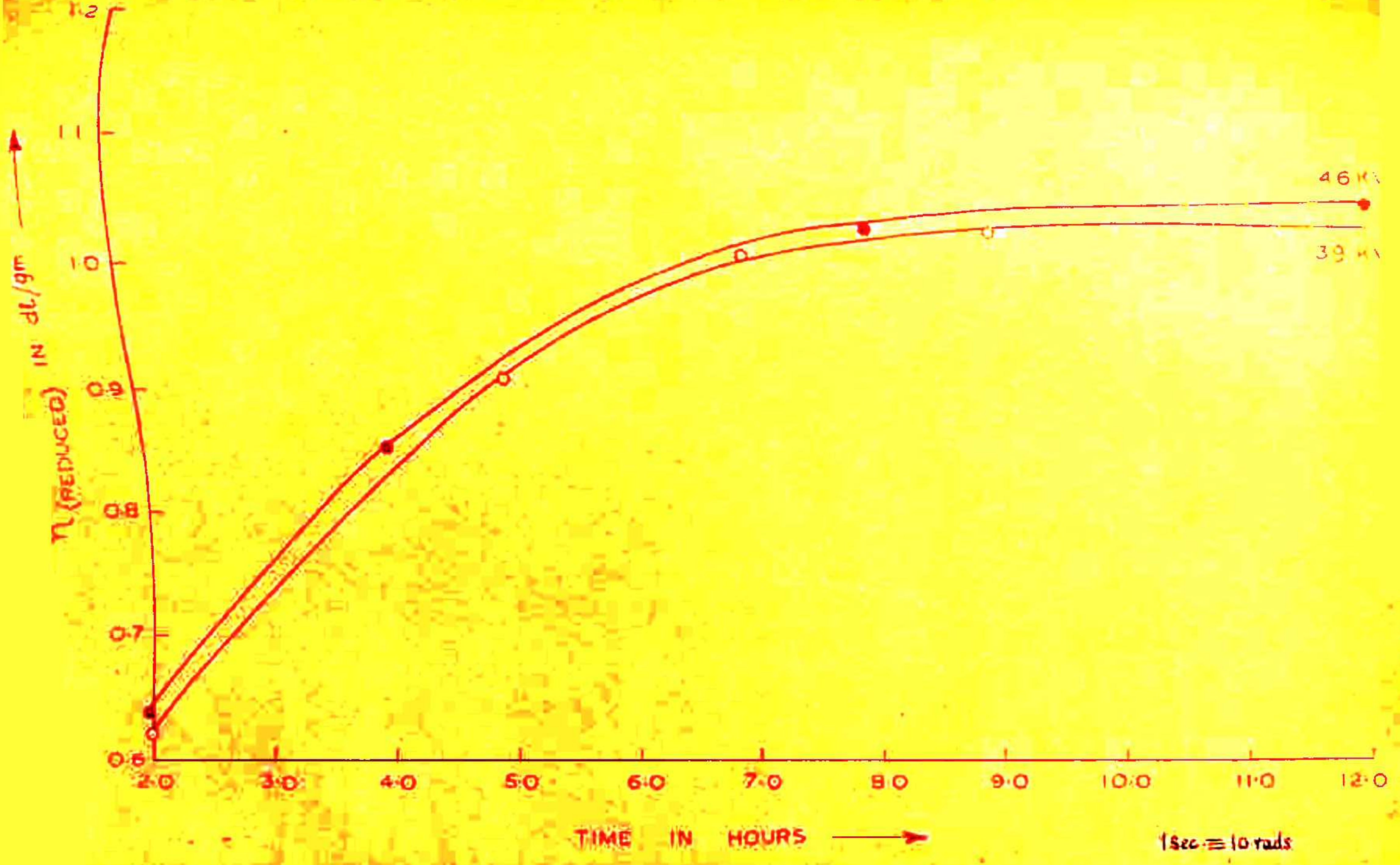

FIG. 5 REDUCED VISCOSITY CHANGES DUE TO RADIATIVE DEGRADATION OF METHYL CELLULOSE.

TABLE - 5.

Methyl Cellulose

Ionization current in Amp.	Calculated value of X-ray dose in rads per hour.
29×10^{-6}	7.43×10^{6}
51×10^{-6}	12.90×10^{6}
70×10^{-6}	17.7×10^{6}
90×10^{-6}	22.6×10^{6}
100×10^{-6}	26.9×10^{6}
120×10^{-6}	31.0×10^{6}
150×10^{6}	37.0×10^{6}

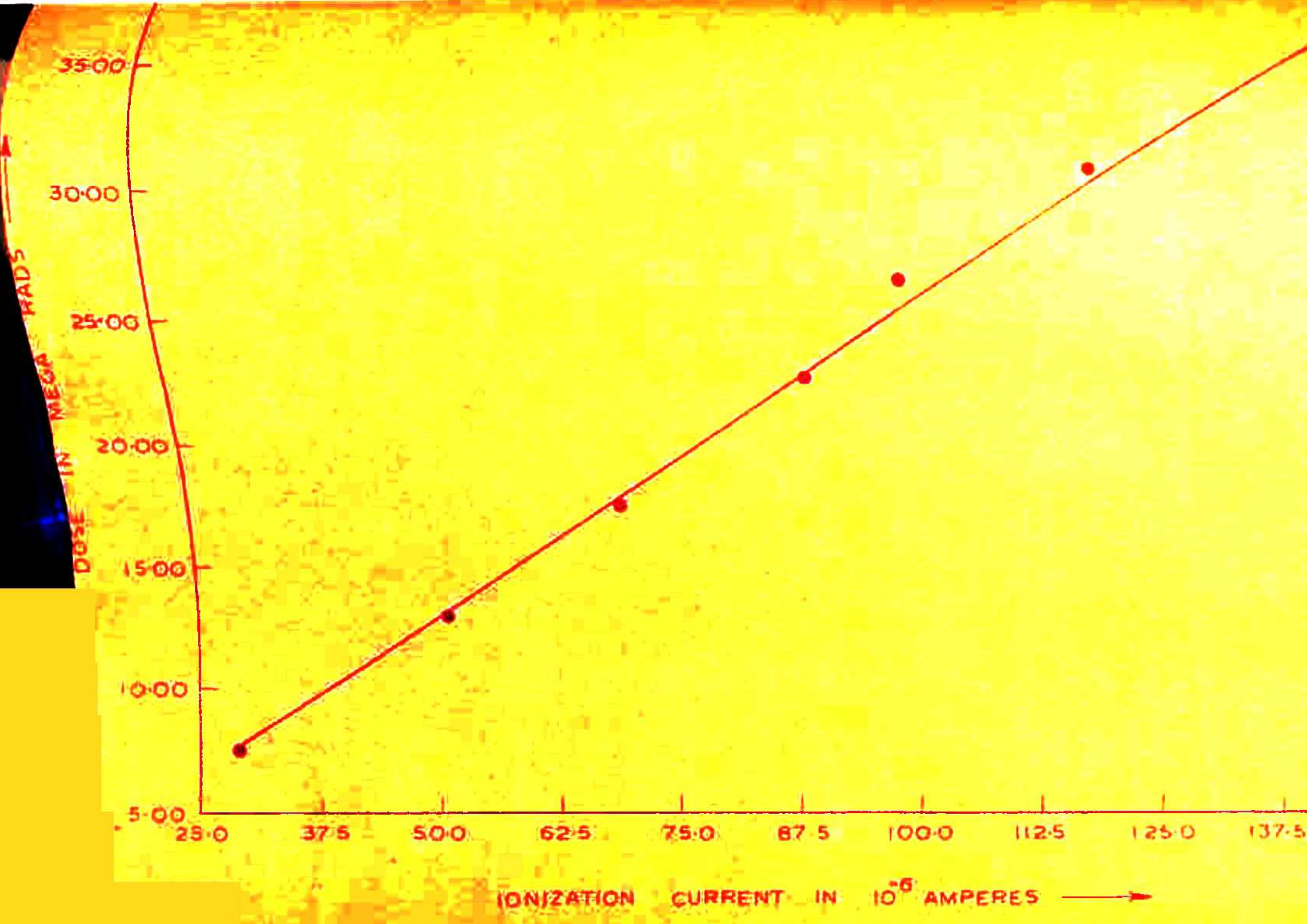

FIG.-6 RADIATIVE IONISATION OF METHYL CELLULOSE.

substance for dosimetric purposes (Fig. 4 and 6). The increase in reduced viscosity (Fig. 5) with increasing radiation dose indicates an accelerating effect on denaturation reaction[7]. For dosimetric purpose the dose absorbed can be calculated for the time it has been given to the solution as in chapter II. A linear plot between current and dose helps in developing a methyl cellulosic dosimeter. By increasing the intensity of the X-rays, the physical effects are only enhanced (Fig. 4 and 5).

3. ETHYL CELLULOSE:

Ethyl cellulose is a thermoplast water soluble cellulose ether. The X-radiation ruptures weak bonds of ethyl cellulose rapidly and stronger bonds slowly. The viscosity reaches a limiting value. There is a linear relation between the dose absorbed and ionization currents, which can be utilized for a dosimeter. Ethyl cellulose is popularly used as a binder and filler in dry vitamin preparations and also as a protective coating for vitamins and mineral tablets. Since polymers are affected by radiation[1-6], the power of protection of this compound against ionizing radiations was tested with the aim of finding the possibility of some linear relationship between changes in any of its physical properties and the radiation dose absorbed by it.

TABLE - 6.

Ethylcellulose

Dose absorbed in M-rads.	Calculated value of viscosity in CP.	Log (viscosity)
0.5	1487.13	3.1724
0.7	928.7	2.9678
1.0	548.75	2.7394
1.5	324.0	2.5105
2	200.9	2.3010
2.5	169.3	2.2287
3	134.2	2.1277
4	91.14	1.9597
4.5	79.11	1.8983
5.0	58.28	1.7682
7.0	34.35	1.5372
9.0	70.98	1.2549

TABLE - 7.

Ethyl cellulose.

Ionization current in micro amp.	Calculated dose in M-rads.
1.5	0.5
3.5	1.0
5.5	1.5
7.5	2.0
11.0	2.8
18.8	5.0
29.0	7.4
51.0	12.9
70.0	17.7

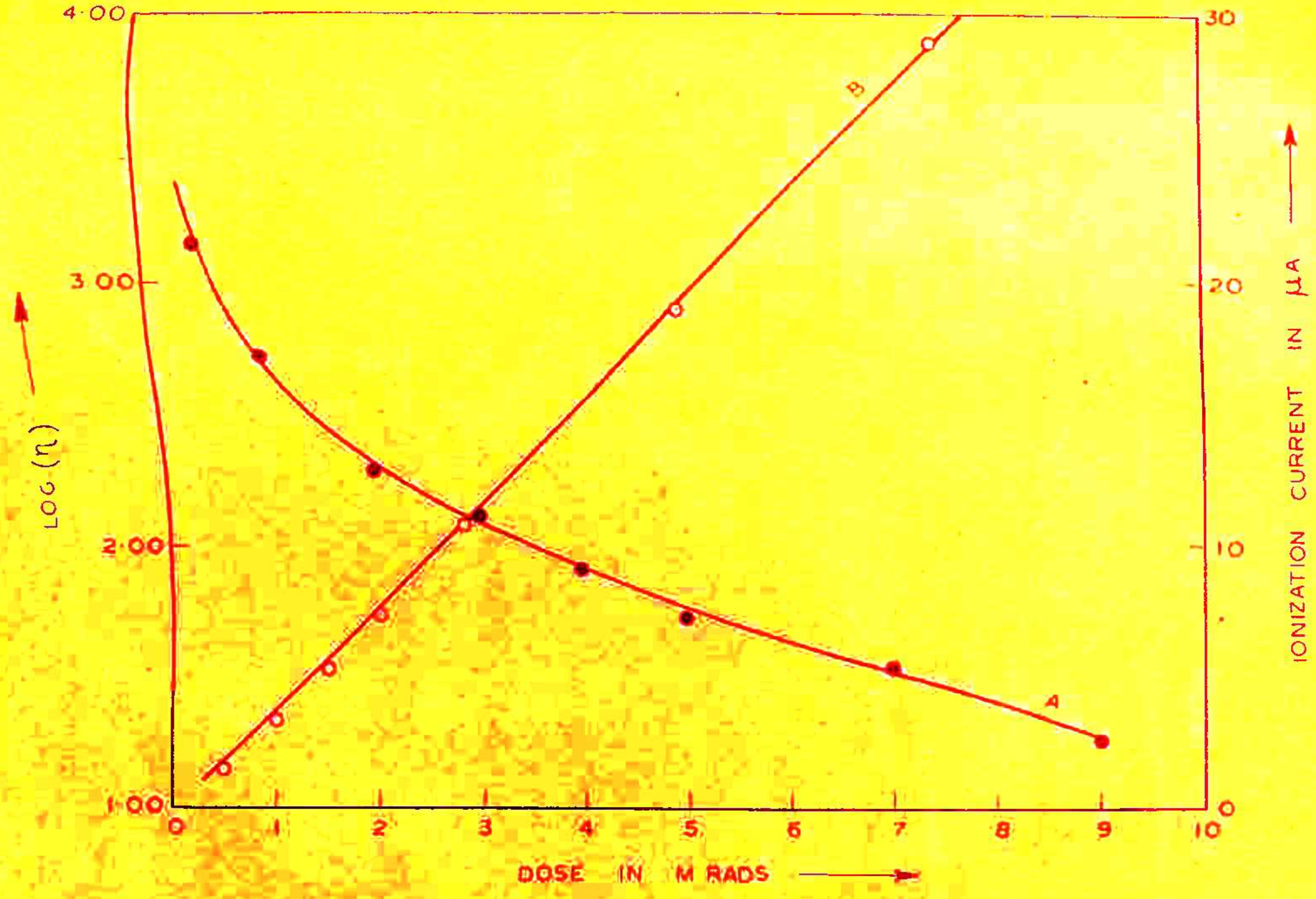

FIG. 7 (A) VISCOUS DEGRADATION OF ETHYLCELLULOSE BY X RADIATION.
(B) RADIATIVE IONIZATION OF ETHYLCELLULOSE.

Five percent solution of ethyl cellulose using toluene- ethanol (60: 40) as solvent was prepared and 20 samples in exactly identical pyrex glass cells were made ready for exposure. Four samples at a time could be exposed by fixing them in positions against the 4 windows of the X-ray (39 kv) plant. The time of irradiation was uniformly increased in all cases and the viscosity was measured by an Ostwald viscometer. Finally the ionization currents developed were measured and the dose absorbed in each case was calculated. All observations were made at the room temperature ($20^{o}C$) and are recoreded in tables 6 and 7.

The value of viscosity decreases quite rapidly with dose which indicates the rupture of weak bonds. (Fig. 7). This rate of change decreases with time showing the slow rupture of stronger bonds. Ultimately viscosity approaches a limiting value and even stronger doses are not much effective. The linear relationship between the ionization current and dose absorbed prove that ethyl cellulose can be used for dosimetric measurements (Fig. 7).

4. SODIUM ALGINATE:

In this study sodium alginate, a sodium salt of alginic acid, is selected. It is slowly soloublo in water forming a viscous solution and is used as a suspending and emulsifying agent and in icecreams. It is also used as a binding agent in the manufacture of tablets. Therefore the viscosity changes in the acqueous solution of sodium alginate due to X-ray doses of 39 kv photon flux was studied to develop an X-ray dosimeter. The changes in flow rate and reduced viscosity changes have also been related to the absorbed dose.

EXPERIMENTAL:

Stir 1.0 gm of the sample with 25 gms of cold water. Add 70 ml of boiling water, stir for a few minutes, cool, dilute to 100 ml with water and allow to stand for 24 hours. Take 25 ml of the solution in a pyrex tube and irradiate the sample. Four such samples were taken at a time and the 4 windows of the X-ray plant were used for irradiation. The X-ray working voltage was kept fixed at 39 kv and 10 mA. The four pyrex glass sample tubes were exactly identical. The sample were kept 5 cms away from the windows so that each could face the full photon flux. The samples were irradiated for 2 hours, 4.5 hours, 9 hours and 16 hours respecively. A fifth sample was irradiated in exactly similar conditions for 44 hours. After respective irradiations, 10 ml of each sample was pipetted out in Ostwald's viscometer and the flow time was noted in each case. The viscometer was cleaned with fresh and filtered chromic acid solution each time, rinsed again with distilled water and dried. The flow time for unirradiated solution and fresh water (solvent) was also noted. At least 10 runs for each set were taken and the mean flow time is recorded in the table 8.

RESULTS:

A graph η (reduced) versus irradiation time shows (Fig. 9) that reduced viscosity does not go to infinity but approaches a limiting value and the increased in reduced viscosity with increasing dose indicates the rupture of many weak linkages at the start and as the dose increases more and more with time of irradiation, the rupture of stronger bonds also starts but at an extremely slow rate. The linear relationship shows that this system can be used as a X-ray dosimeter (Fig. 8).

TABLE - 8.

Sodium Alginate

t_u = 77 sec.

t_o = 22.5 sec.

T = Irradiation Period

Temperature = 22°C

C = 0.4 gm/100 ml

S. No.	t_r in sec.	Calculated value of $\text{Log}\left[\frac{\frac{t_r}{t_o}-1}{\frac{t_u}{t_o}-1}\right]$	Irradiation time T	Log T	Calculated value of η (reduced)
1.	73.6	-0.028	2 hours	0.3010	1.10
2.	68.5	-0.0737	4.5 ,,	0.6532	1.22
3.	63.4	-0.1247	9 ,,	0.9540	1.375
4.	60.8	-0.1550	16 ,,	1.2041	1.462
5.	55.0	-0.2246	44 ,,	1.6435	1.730

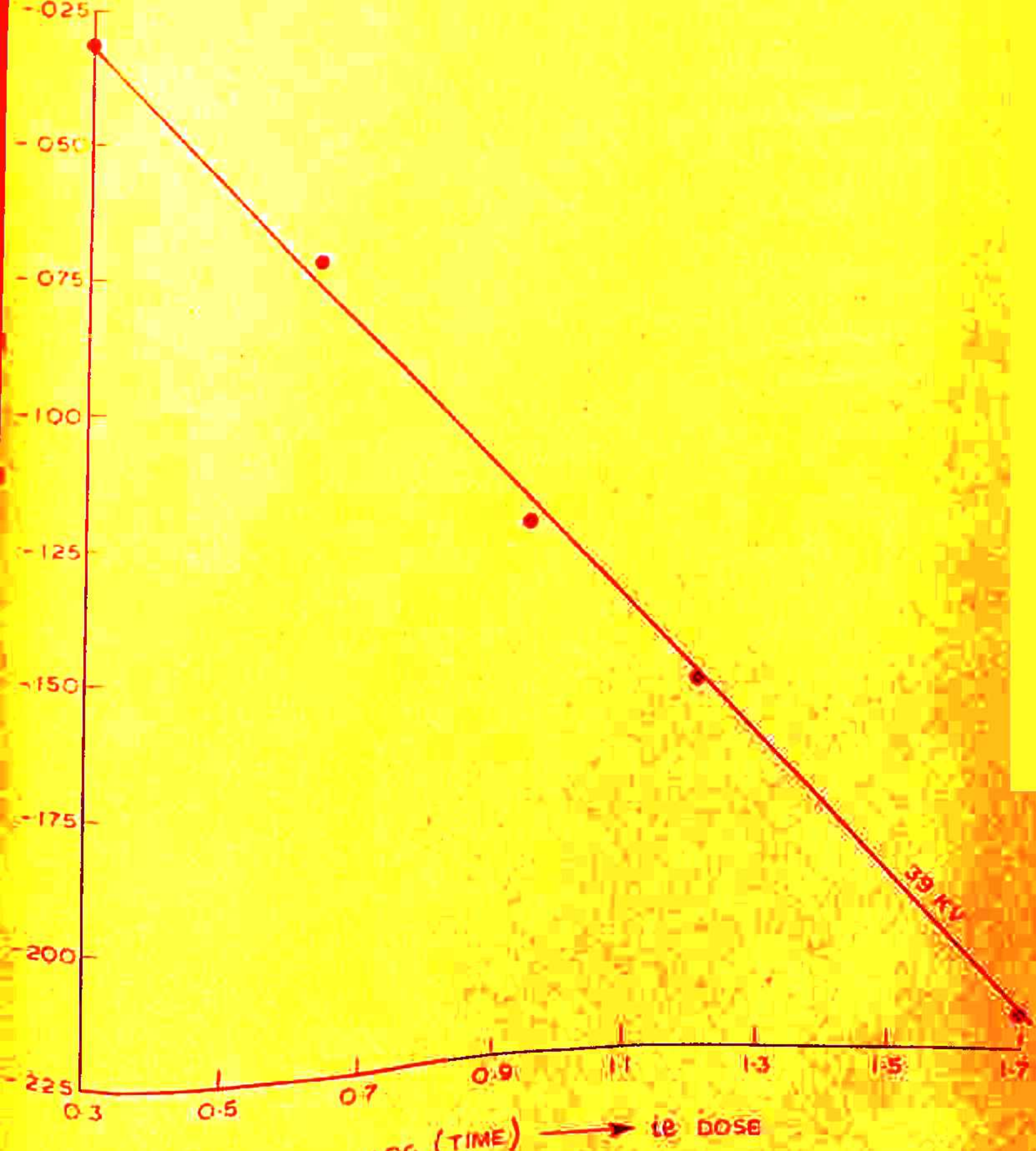

FIG. 8 VISCOUS FLOW RATE CHANGES IN RADIATIVE DEGRADATION OF SODIUM ALGINATE.

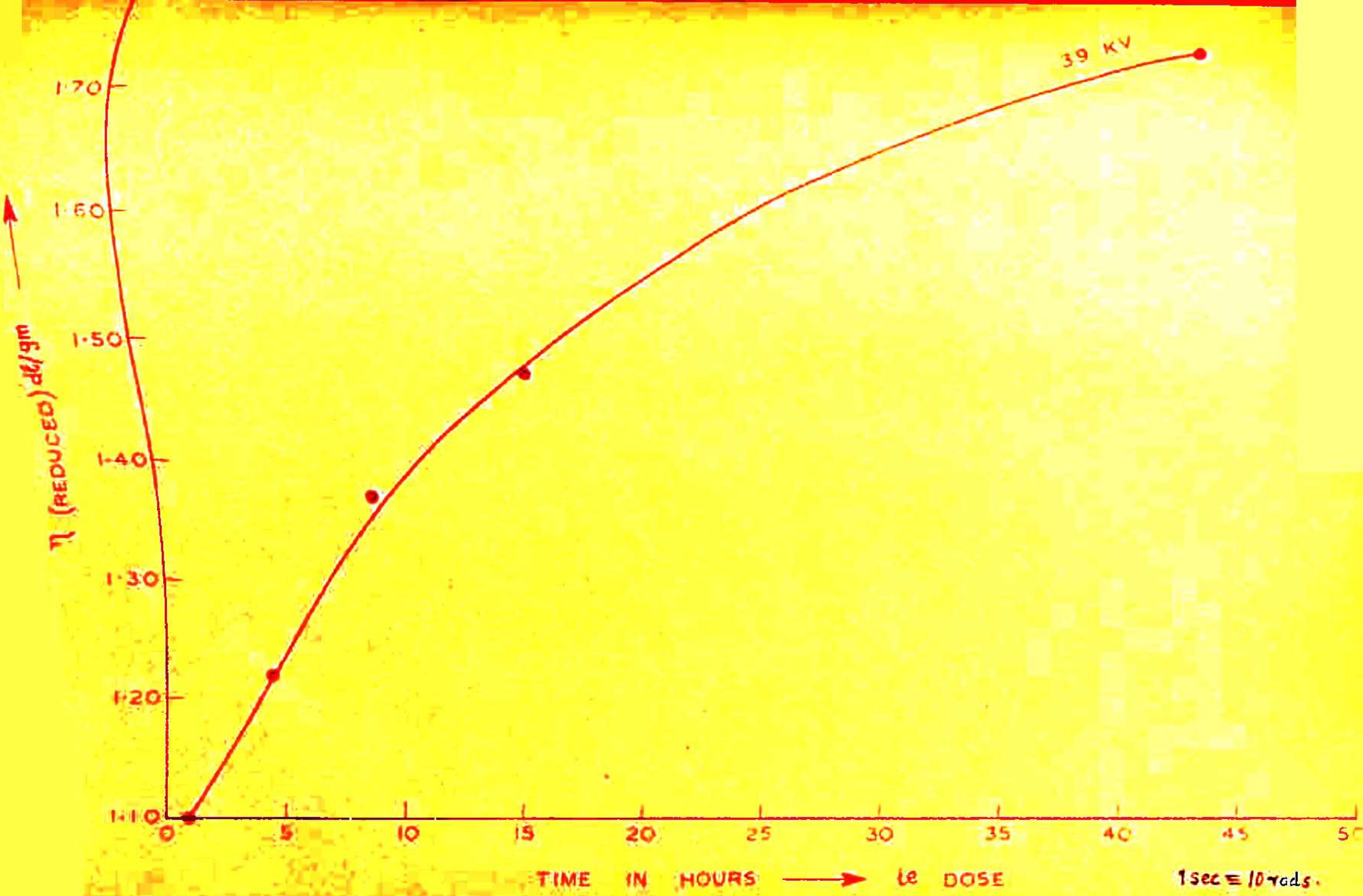

FIG 9 REDUCED VISCOSITY CHANGES IN THE RADIATIVE DEGRADATION OF SODIUM ALGINATE

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CHAPTER 6.

DOSIMETRIC STUDY BY MEASURING THE CHANGES IN OPTICAL DENSITY AND MELTING POINTS OF SOME SUGARS AFTER IRRADIATION BY X-RAYS

DOSIMETRIC STUDY BY MEASURING THE CHANGES IN OPTICAL DENSITY AND MELTING POINT OF SOME SUGARS AFTER IRRADIATION BY X-RAYS.

INTRODUCTION: (Change in optical density)

The optical density changes due to Gamma ray irradiation of certain samples (solids and liquids) have been used to measure Gamma ray doses by many investigators$^{1-5}$. In the present study aqueous solutions of some organic substances (sugar) of various molar strengths have been tried for X-ray dosimetric purposes.

If I_o is the intensity of the incident light and I_t is the fraction of the incident light transmitted by the thickness t of the medium, then I_t/I_o is called the transmittence. The optical density (O.D)of the medium is given as $O.D. = \log \frac{I_o}{I_t}$ for the given wavelength. Thus a medium of optical density 1, for a given wave-length, transmits 10% of the incident light. It is, therefore, necessary first to fixup that wave-length at which optical density is the maximum for the sample. This is called the peak wavelength and is specific for the particular sample. Optical density variations are then measured at this peak wavelength corresponding to the X-ray dose absorbed.

EXPERIMENTAL:

The Beckman Model DU-2 Ultraviolet Spectrophotometer has been used to measure optical density. It offers greater precision and improved operating convenience in the wavelength range from 190 to 1000 millimicrons. The optical density also changes with temperature and in this appratus the temperature may be maintained at any desired level within the range from 0°C to 100°C by a Thermospacer Set. The absorption spectra of 5M Xylose, 2.5M Sucrose and 1M levulose are plotted.(Tables 1, 2 and 3 and Fig. 1, 2 and 3). From these the peak wavelengths have been found to be 555 $m\mu$ for Xylose, 565 $m\mu$ for Sucrose and 280 $m\mu$ for levulose. The samples were then irradiated for definite periods of time and the doses absorbed were calculated as in chapter II. The corresponding optical density changes in the various samples were measured at the relevent peak wavelength and are tabulated (Table 4, 5 and 6).

RESULT:

The graphs (Fig. 1, 2 and 3) , X-ray dose versus optical density show linear relationship and hence this study may be used for dosimetric purposes.

TABLE - 1.

Absorption Spectrum of Sucrose

Wavelength in $m\mu$	Optical density
345	0.05
375	0.07
420	0.12
440	0.15
475	0.23
500	0.27
530	0.32
550	0.34
565	0.38
600	0.34
640	0.30
680	0.25

TABLE - 2.

Absorption Spectrum of Xylose

Wavelength in mμ	Optical density
345	0.065
375	0.085
420	0.13
440	0.18
475	0.25
500	0.31
550	0.37
555	0.41
600	0.32
640	0.25
680	0.20

TABLE - 3.

Absorption Spectrum of Levulose

Wavelength in mμ	Optical density
250	0.52
255	0.62
260	0.78
265	0.94
270	1.10
275	1.242
280	1.30
285	1.27
290	1.135
295	0.91

TABLE - 4.

SCUROSE (2.5 M)

Absorption Spectrum at λ = 565 mμ

Dose in Kilo Rads	Optical density
0.	0.180
23.220	0.192
46.440	0.205
69.660	0.219
92.880	0.232

TABLE - 5.

Xylose (5M)

Absorption Spectrum at λ = 555 mμ

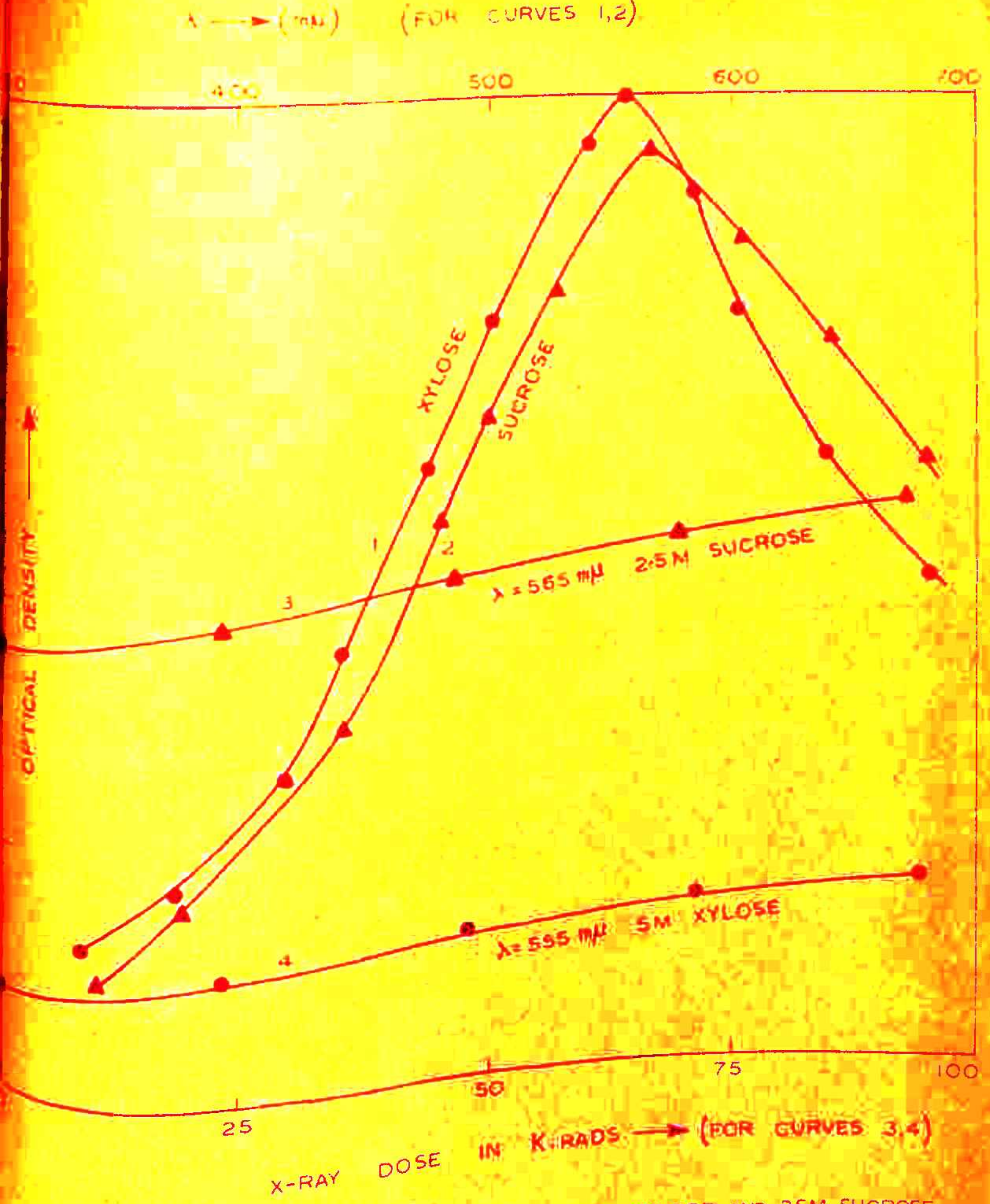
Dose in Kilo Rads.	Optical density
0	0.0452
23.678	0.0525
47.358	0.0601
71.037	0.0680
94.716	0.0755

TABLE - 6.

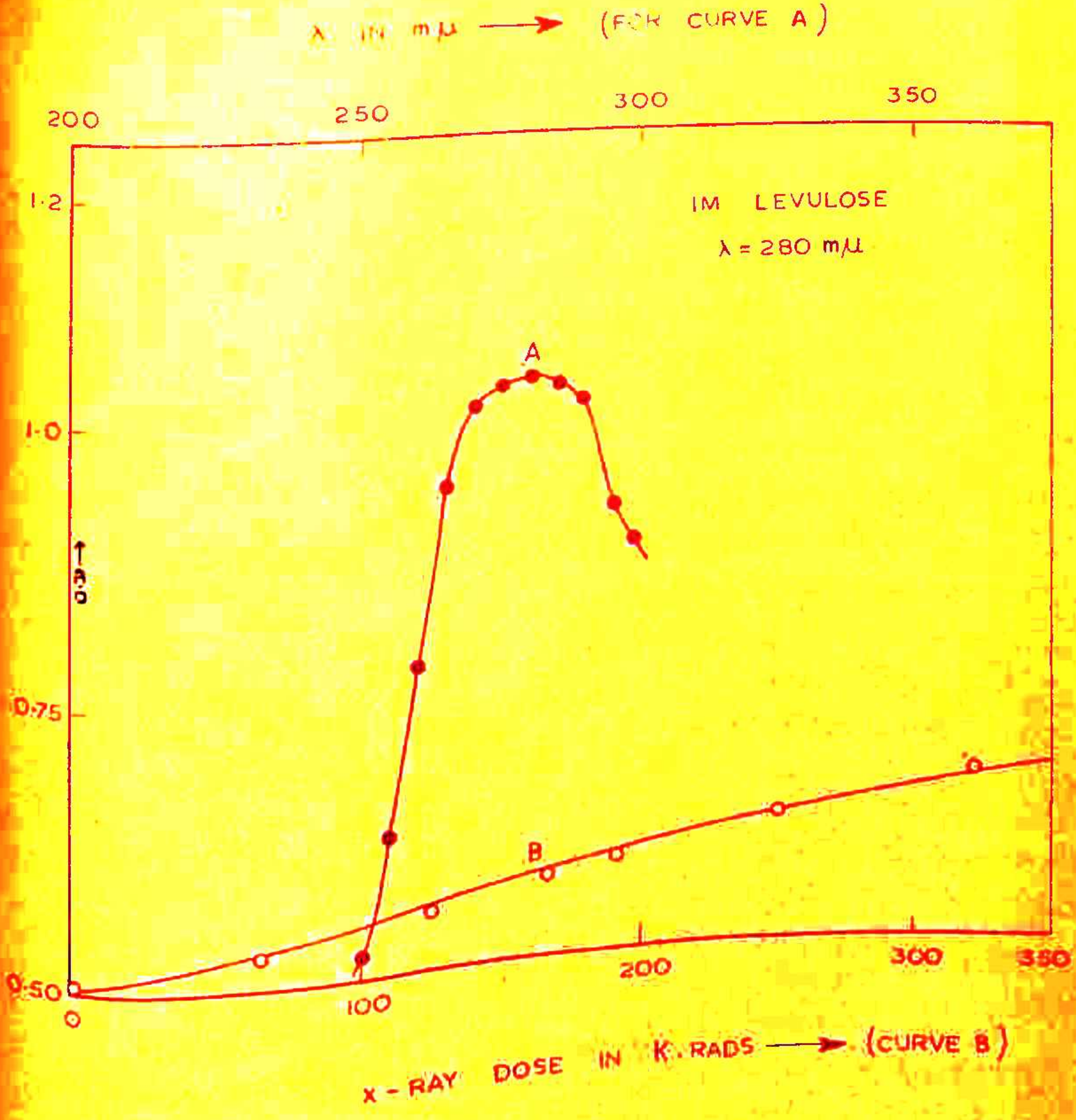
Levulose (1 M)

Absorption Spectrum at λ = 280 mμ

Dose in Kilo Rads.	Optical density
0.	0.501
64.528	0.532
125.568	0.561
193.584	0.591
251.136	0.620
322.640	0.650



8) ABSORPTION SPECTRA OF IRRADIATED 5M XYLOSE AND 2·5M SUCROSE. OPTICAL DENSITY VARIATIONS OF 5M XYLOSE ($\lambda = 555 m\mu$) & 2·5M SUCROSE ($\lambda = 565 m\mu$) WITH X-RAY DOSE.



G. 3

A :- ULTRA VOILET ABSORPTION SPECTRUM OF IRRADIATED IM LEVULOSE

B :- OPTICAL DENSITY VARIATIONS OF IM LEVULOSE SOLUTION WITH X - RAY DOSES AT 280 mμ.

INTRODUCTION: (Change in melting point)

One of the basic applications of the melting point is its use as a criterion of purity. If a substance is pure, the melting point will usually be sharp. It has become the most important physical property for characterization and indentification and estimation of purity in a cysthesized compound. When a pure crystalline solid melts, the molecules arranged in a high degree of order in a crystal lattice are separated by thermal forces (temporature) to form the liquid state in which they are in complete disorder due to an abrupt increase in entropy. Thus the phenomenon of melting a crystalline solid (eg. sugars) involves the transition of the molecules from an ordered, crystalline lattice in which each atom or molecule occupies a definite mean position, to a disordered liquid.

If such a crystalline substance is irradiated by X-rays the crystalline lattice is disturbed due to bond fractures. Thermal vibrations of smaller intensity may overcome the intermolecular forces holding the molecules in the solid lattice. As such the melting point of the irradiated crystalline solid may decrease and this decrease in the melting point will obviously be proportioned to the X-ray dose absorbed by the substance.of course care has to be taken about the availability of purest form of the sugar sample for irradiation. The degradation of the parent sample into fragments of lower moleculer weights adds an element of impurity into the sample which also helps in decreasing the melting point of the substance because any foreign material, even of the same catagory but of different molecular weight, acts as an impurity in the parent substance.

EXPERIMENTAL:

For this type of investigations, D-Fructose, D-Glucose and Sucrose samples were taken in 1.0 mm capillary tubes in turn and exposed to 39 kv X-ray flux from a distance of 1 cm from the exit window. In this case the samples received 3.6×10^6 rads per hour (Ref. Chapter II) equivalent in water. Certain precautions should be observed in constructing and filling the capillary tubes. The tubing from which the capillaries are made should be washed and dried before they are drawn. They should be sealed at both ends to keep them clean until they are put to use. The substance under examination be well packed into the bottom of the tube to ensure the maximum contact between the sample and the walls of the tube. The sugar samples under investigation were of the purest form available from Merck. The irradiation scheme is given in tables Nos. 1, 2 and 3.

For measuring the melting point just after the irradiation of the sample for specific time and for a definite quota of dose, the following apparatus was used.

Capillary Melting Point Apparatus
Arther H. Thomas Comp.
Philadelphia PA U.S.A.

In this apparatus the capillary tube containing the sample is immersed in a both (Silcone oil) adjacent to the thermometer. The temperature is gradually raised and the temperature is noted when the crystaline sample just seems to melt. The bath is automatically well stirred&is heated at an even rate about 1°C per minute in the vicinity of the melting point. The apparatus ensures unrestricted visibility of the capillary tube and the thermometer. The rate of heating should be kept low for maximum accuracy in the melting point.

INFERENCE:

If Y represents the melting point and X represents the X-ray dose in M. rads. , Then the following empirical relations may be established:

Sucrose : $Y = -0.775X + 187$
D-Glucose: $Y = -0.235X + 142$
D-Fructose: $Y = -0.333X + 109$

The fig. shows that the linear relationship between dose absorbed and melting point can be utilised to develop dosimeters.

RADIATIVE DEGRADATION IN THE MELTING POINTS

(1) SUCROSE

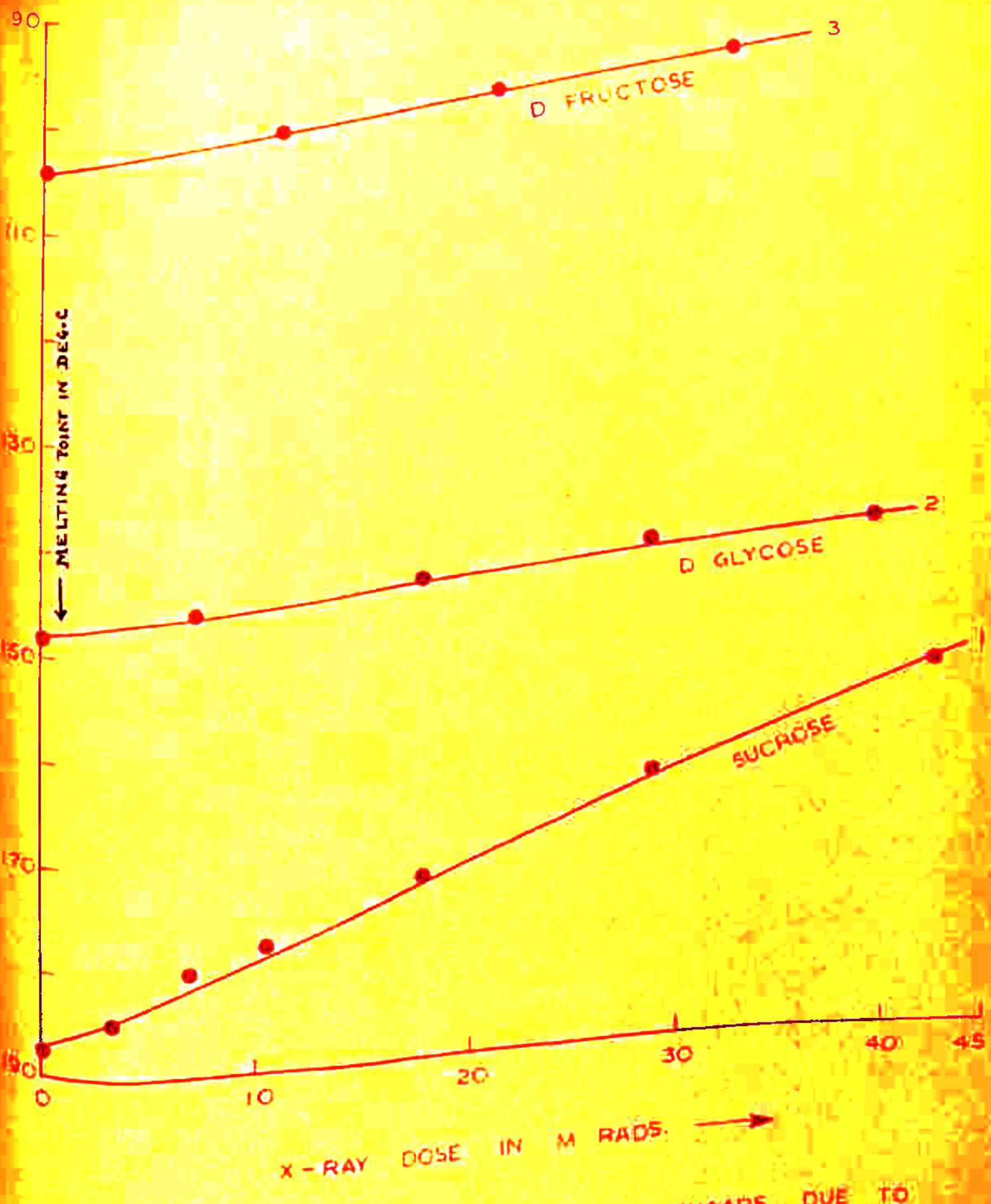
Irradiation time in hour	Dose absorbed in Rads	Melting Point
0	0	188°C
1	$3.6x10^{6}$	185°C
2	$7.2x10^{6}$	181.9°C
3	$10.8x10^{6}$	178.8°C
5	$18.0x10^{6}$	173.0°C
8	$28.8x10^{6}$	163.9°C
12	$43.2x10^{6}$	154.4°C

(2) D-FRUCTOSE

0	0	104°C
3	$10.8x10^{6}$	100.4°C
6	$21.6x10^{6}$	96.9°C
9	$32.4x10^{6}$	93.2°C
13	$46.8x10^{6}$	88.5°C

(3) D-GLUCOSE

0	0	148°C
2	$7.2x10^{6}$	146°C
5	$18.0x10^{6}$	144.2°C
8	$28.8x10^{6}$	141.9°C
11	$39.6x10^{6}$	139.8°C



DEGRADATION OF MELTING POINT OF SUGARS DUE TO IRRADIATION BY X-RAYS.

REFERENCE:

1. Armstrong, W. A. and Grant, G. A., Can. J. Chem. Vol. 36, 1398 (1958).

2. Aitken, P. B., Dyne, P. J. and Trapp, E. C., Nucleonics, 15(1) 100, (1957).

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CHAPTER 7.

DOSIMETRIC STUDY OF THE CHANGES BROUGHT ABOUT BY X-RADIATION DOSES IN THE DI-ELECTRIC CONDUCTANCE OF THIN FILM CAPACITORS.

DOSIMETRIC STUDY OF THE CHANGES BROUGHT ABOUT BY X-RADIATION DOSES IN THE DIELECTRIC CONDUCTANCE OF THIN FILM CAPACITORS.

INTRODUCTION:

Induced conductivity by X-radiation in solid dielectric films is utilised to develop X-ray dosimeters. In these materials the conductivity may be increased by several orders of magnitude by ionizing radiation and the return to normal value does not take much time. A sublinear relation exists between induced dielectric conductance and the dose absorbed, thus, making this study useful for dosimetric purposes. This chapter describes the results of experiments performed to study the effects of exposure to X-radiation on $10\,\mu f$ tantalum oxide and $6\,\mu f$ mylar capacitors.

The basic processes occuring in insulators are very similar to those in semiconductors involving the production of electrons and holes which attain constant mobility[1-2] and subsequently recombine via defect states. The X-radiation can cause changes in the electrical properties of capacitors by changing the properties of the dielectric. The radiation loses energy as it passes through the capacitor. These energy losses produce free electrons in the capacitor dielectric. The charge carriers drift in the presence of an applied field and increase the current in the dielectric. The induced conductivity takes some time to build up to the equilibrium level. The induced conductivity results from absorption of energy radiation, excitation of charge carriers from non-conducting states to conducting states and return of charge carriers from conducting states to non-conducting states[3-4]. Fowler J. F[5], assumes that ionizing radiation produces free electrons and an equal number of holes through out the volume of the substance. Since less then 1 in 10^3 of these will be fast electrons produced by primary ionization, the majority of the free electrons have thermal energy. The drift of these electrons when an electric field is applied is measured as induced current. Ionic conductance is negligible since it requires very high activation energies. The existence of any energy band structure is not obvious in these materials[5-6]. However if an electron is given sufficient energy by the ionizing radiation, it may become free to move through the substance and many be said to exist in a conduction band. The term dielectric conductance is used to distinguish this quantity from metallic conductances and it sounds more logical to speak of dielectric conductance in materials like Tantalum Oxide and Mylar which are normally thought to be insulating. It is the ratio of the current measured at a specified dose to the steady dc voltage applied to the specimen throughout the irradiation period.

DESCRIPTION OF THE THIN FILM CAPACITORS:

This is a thin film of Tantalum Oxide (4000 x 10^{-8} cm) on Tantalum deposited by anodization method. The Tantalum Oxide is then flushed with silver oxide which gives rise to an ohmic contact on the plate. The thicker the film, the more uniform the density distribution and the more sensitive and reproducible the dosimeter. Mylar is a crystalline polyester with wide commercial applications under such trade names as dacron, terylene etc. These capacitors are superior in characterstics and easy to manufacture. Their stability and performance are unaffected at as high as 60 kv doses. Even after repeated voltage and dose cycling, the reproducibility of the same measuring condition remains. These capacitors recover after some time to the original value, once the X-ray unit is closed. The applied voltage on these capacitors was 1.25 x 10^5 volts/cm.

EXPERIMENTAL:

The method depends upon the measurement of the leakage current through the specimen when a constant X-ray photon flux at 53 kv is incident on it. The specimen is connected to the circuit after applying the fixed bias voltage. The X-ray unit is started and as the window is opened, the measurement of current with time is made after adjusting the gain as near full scale deflection as possible and noting the current at the end of the electrification period. The dc dielectric conductance is then computed for various periods. The dose absorbed in a particular time period by the thin film capacitor is calculated as equivalent to absorbed by water in a Fricke Dosimeter. Care must be taken to ensure that stray and unwanted currents do not influence the measurement except the dark current which is already taken care of before exposing the sample to radiation. As the currents involved in this measurement were of the order of 10^{-10} ampere, the connections to the measuring instruments were shielded by thick ebonite planks against posibility of charges being induced in the measuring circuit by moving objects. All measurements were made at room temperature (25^{o}C) as measurements below room temperature present special problem because of the possibility of condensation of moisture on the surface of the specimen. The results are given in Table 1 and 2.

DISCUSSIONS:

There is no direct method of calculating the X-ray dose absorbed by thin film capacitors because (1) it is extremely difficult to ensure uniform thickness of the films

(2) in the case of Mylar the structural formula as well as the percentage of different functional groups is never known. Therefore the best way to use such capacitors for dosimeter purposes is to note the different periods of irradiation of the capacitors for which the ionization current is accurately measured at a fixed voltage. The dose absorbed by the thin film is then calculated in terms of its equivalent in water (Fricke Dosimeter Chaper II) for which the X-ray unit is already calibrated. The induced dielectric conductance k has been found to be a function of dose absorbed by the dielectric materials under investigation and can be empirically related by $k = KR^{\delta}$ where K and δ are the material constants. The logarithm of induced dielectric conducatnce versus logarithm of dose absorbed, give a straight line graph. The slopes of the straight lines for two materials under study are 1.8 and 2.4 for Tantelum Oxide and Mylar respectively. Thus the study is useful for developing X-ray dosimeters. The Tantalum Oxide and Mylar dosimeters have ranges from 50 K rads to 100 K rads and from 100 K rads to 300 K rads respectively. Above these ranges the dosimeter readings become unstable and hence unreliable.

TABLE - 1.

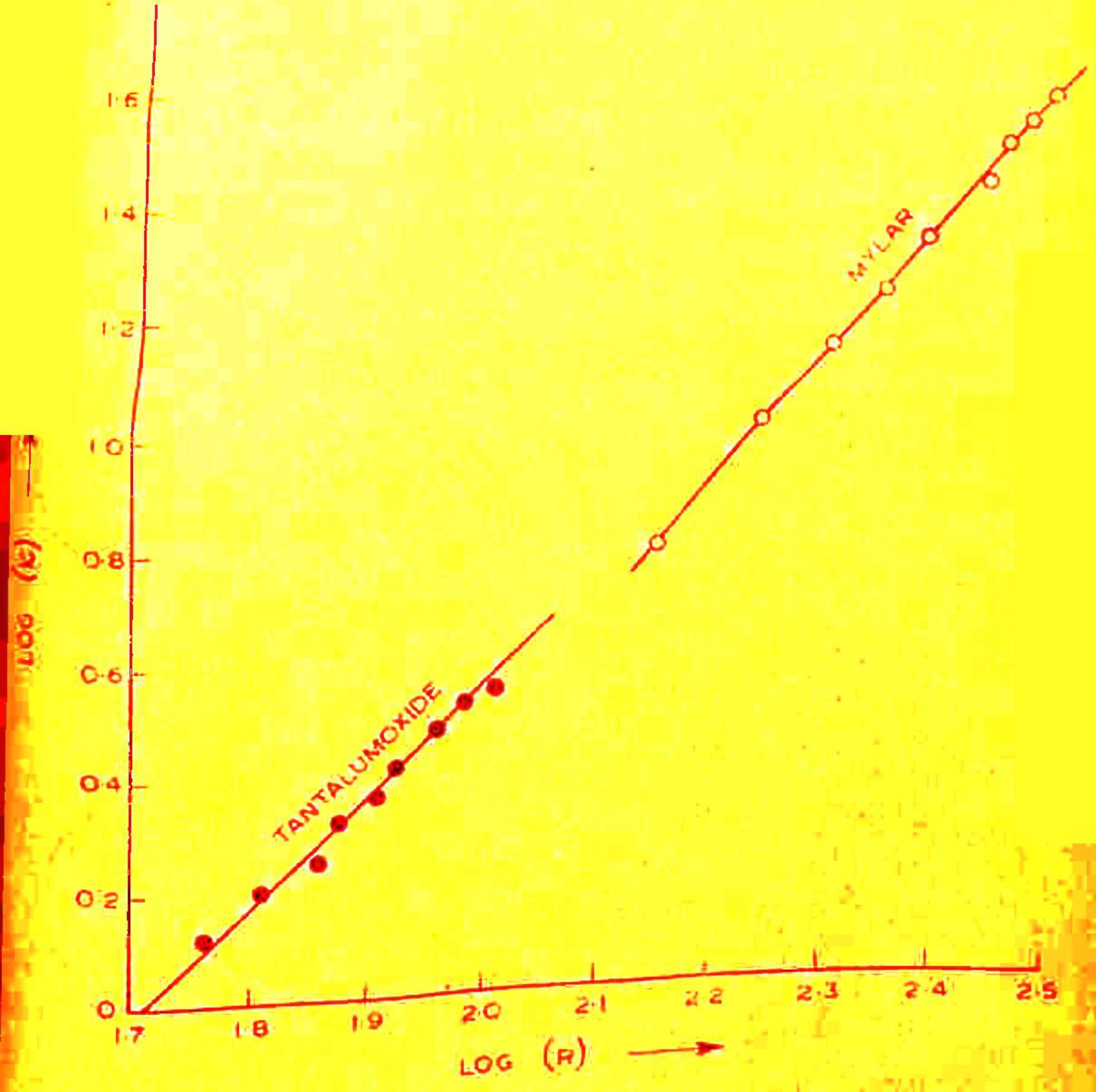
Tantalum Oxide Capacitor

Dielectric conductance in ohms^{-1}	Dose (Equivalent in water) in Kilorads.
1.31×10^{-9}	57.900
1.60×10^{-9}	64.800
1.80×10^{-9}	72.000
2.11×10^{-9}	75.600
2.30×10^{-9}	81.000
2.60×10^{-9}	84.960
3.01×10^{-9}	91.800
3.30×10^{-9}	97.200
3.50×10^{-9}	102.600

TABLE - 2.

Mylar

Dielectric conductance in ohm^{-1}	Dose (equivalent in water) in Kilo rads
6.1×10^{-9}	144.0
10.2×10^{-9}	178.1
14.0×10^{-9}	204.9
17.9×10^{-9}	227.6
22.1×10^{-9}	247.4
27.9×10^{-9}	278.6
32.2×10^{-9}	289.2
35.95×10^{-9}	303.8
40.0×10^{-9}	317.4



LOGARITHM OF DIELECTRIC CONDUCTANCE (K) V/S LOGARITHM OF DOSE R.

REFERENCE:

1. Kooi, E., Philips Research Report Vol. 20, p. 306, June (1965).

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CHAPTER 8.

X-RAY DOSIMETRIC STUDY OF SOME SOLID STATE DEVICES.

X-RAY DOSIMETRIC STUDY OF SOME SOLID STATE DEVICES.

(DIODES)

INTRODUCTION:

Transient radiation effects are defined to be manifestations of electrons excited in materials[1] and many of these disappear with very short relaxation time. These effects are distinct from (i) displacement radiation effects, which are the manifestation of atoms that have been displaced from their normal positions in crystalline lattices and (ii) chemical radiation effects, which are due to the rearrangements of molecular bonds. Surface effects become noticeable at radiation dose of 1000 Rads, as compared to 10^7 Rads for bulk effects[2]. The most radiation sensitive parameters have been found to be the reverse-bias leakage currents for diodes and I_{CBO} and h_{FE} for transistors. For diodes the leakage current may increase as much as several orders of magnitudes and may or may not saturate. The degraded characteristics show partial and sometimes complete recovery. Recovery is promoted by baking, forward biasing and exposure to radiation without bias. The surface effects of ambient are also significant. The magnitude of the reverse current also depends upon the relative humidity.

A majority of the tests during this investigation was performed on group of diodes. Junction capacitances were monitored during irradiation for several bias conditions. The amount of degradation produced by X-radiation was dependent on electrical bias, with the degradation in reverse-biased junction being the most severe. The impedence of a rectifying junction in semiconductor devices is even more susceptible to transient radiation effects than the bulk conductance because the properties of these junctions[4] especially when biased in the reverse direction are determined not so much by the majority carrier concentration but by the very much smaller minority carrier concentration[5]. Hence very lou radiation dose rates are capable of changing significantly the minority carrier concentration.

MEASUREMENT OF X-RAY DOSE:

The problem of describing and measuring radiation in units that are proportional to the amount of ionisation is solved when the unit of a rad is adopted[6]. Any material has recieved one rad of exposure when 100 ergs are deposited in one gramm of the material. The simple relationship of silicon, that a rad creates 4×10^{13} hole-electron pairs per cm^3, permits a direct conversion between radiation and carrier generation in a silicon device. The current generated by a diode when exposed to an ionising radiation of R rads/sec is

$$I = e\, g_0\, R\, V_{eff}$$

where $g_0 = 4 \times 10^{13}$ hole-electron pairs per cm^3 and V_{eff} is the effective volume calculated and for the silicon samples it is equal $13.5 \times 10^{-6}\ cm^3$, e is the charge of an electron. Hence if the generated reversed biased current is measured the corresponding radiation dose in rads can be calculated.

EXPERIMENTAL:

The X-ray unit was switched on at 53 kv, 10 mA and the diodes were exposed to X-radiation, the focussing distance always being 10 cms from the window. Each diode was put into the circuit under various reverse bias voltages successively. The following diodes with the reversed -bias noted against each were ultimately selected for dosimetric purposes after many trials of reverse voltage and dose cycling:

CD - 38	Silicon	Reversed	Voltage	20 volts.
CD - 29	,,	,,	,,	120 volts.
CD - 26	,,	,,	,,	15 volts.

DR - 25	Germanium (non-passivated)	Reversed voltage	10 volts.
DR - 10	,,	,, ,,	10 volts.

The reverse voltage was supplied by Systronics transistor power supply, type 611, S. No. 6076, and the leakage current was measured by DC micro voltmeter, Phillips GM. 6220 (two ranges: 1 megaohm and 10 mega ohms) least count 10^{-12} ampere. The X-ray unit strahlenschutzulassung PTB 508 (Germany) was operated on the copper target.

RESULTS:

The diodes CD-38 and CD-36 are (Fig. 1) not recommended for dosimetric purposes because the rate of recombination of electrons and holes is very large even at lower doses. This may be due to impurity states in the device. At higher doses the saturation in the leakage current starts. Of course CD-36 is a bit better than CD-38. DR-10 (Germanium) can be used to measure very small X-ray doses although saturation of the leakage current starts soon after (Fig. 2). CD-26 is extremely reliable in the range 700 rads to 2500 rads (Fig. 3). CD-29 gives a linear relation in the range 200 rads to 50000 rads and is the best amongst its family (Fig. 4). In the

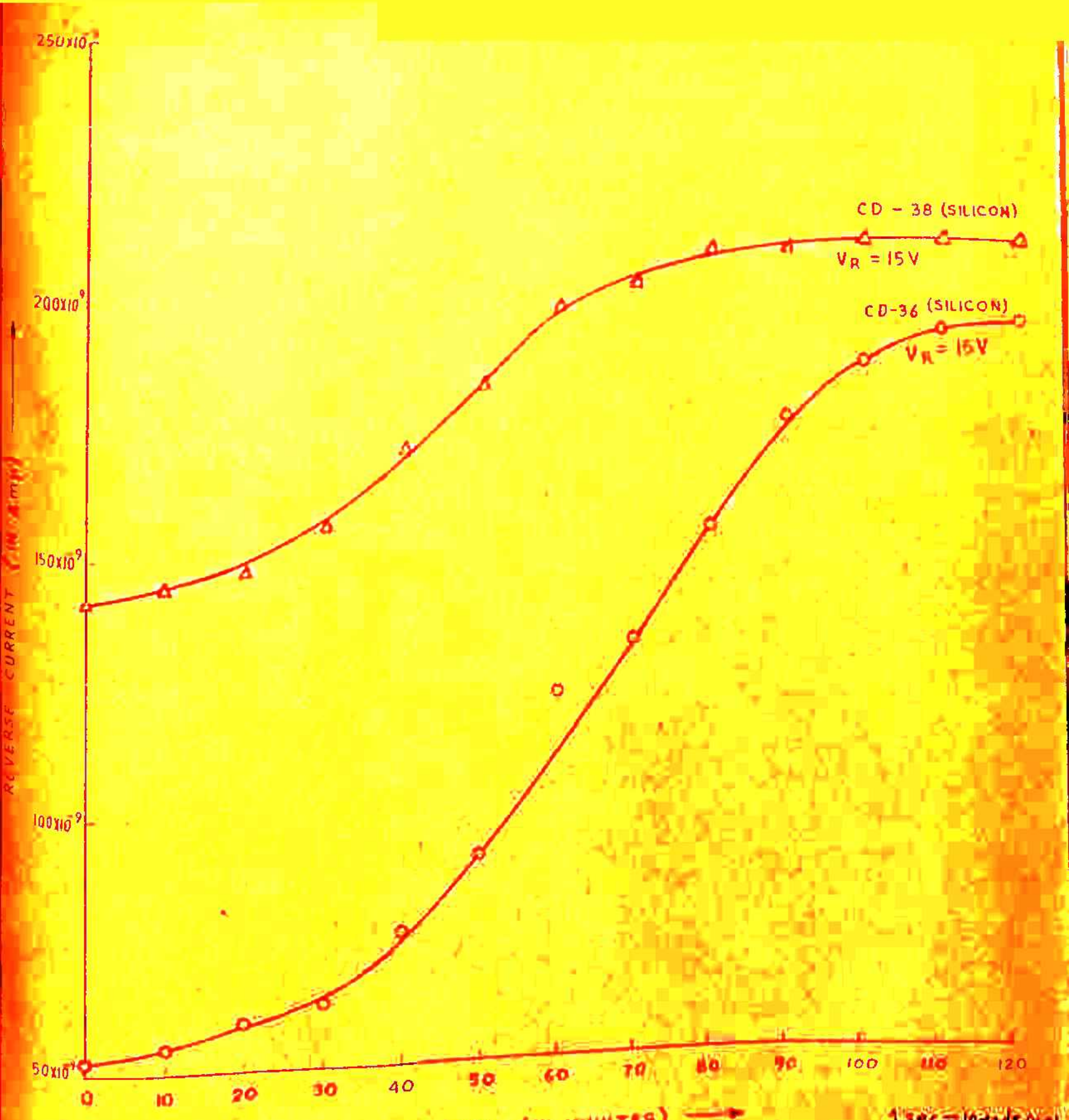


FIG.1 REVERSE CURRENT DEGRADATION IN DIODES.

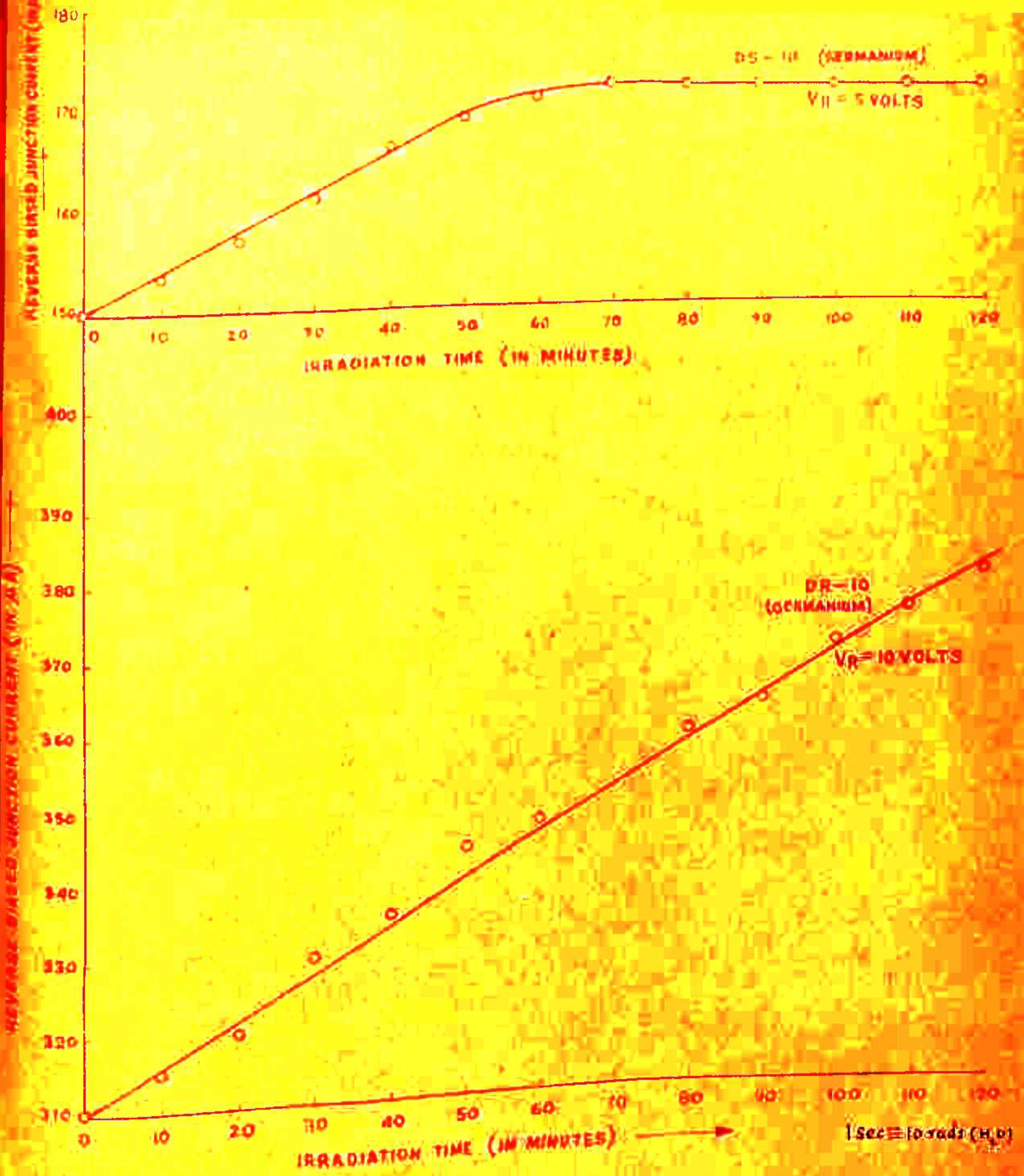


FIG. 2 REVERSE CURRENT DEGRADATION IN DIODES

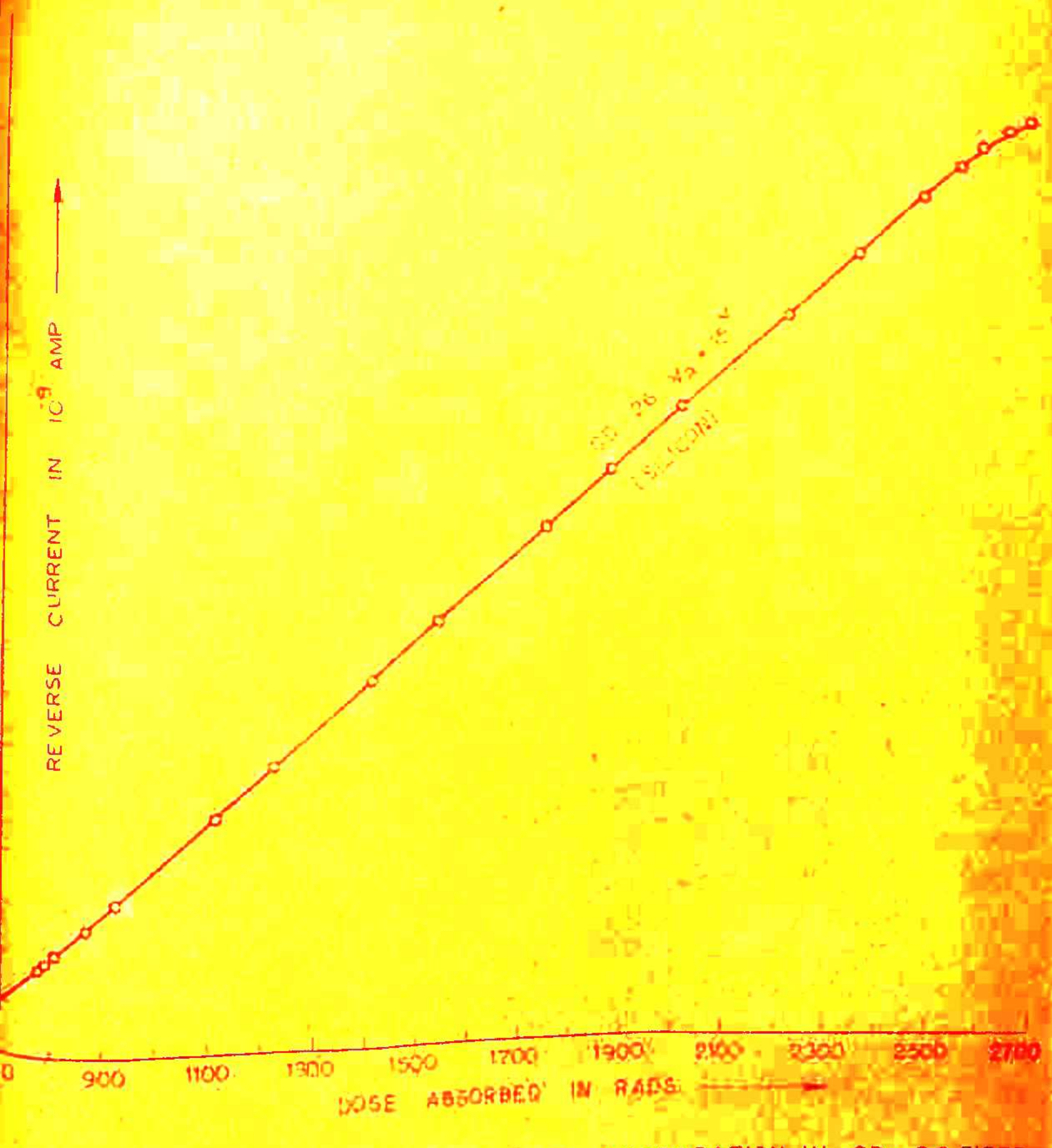


FIG 3 REVERSE CURRENT DEGRADATION IN CD-26 DIODE.

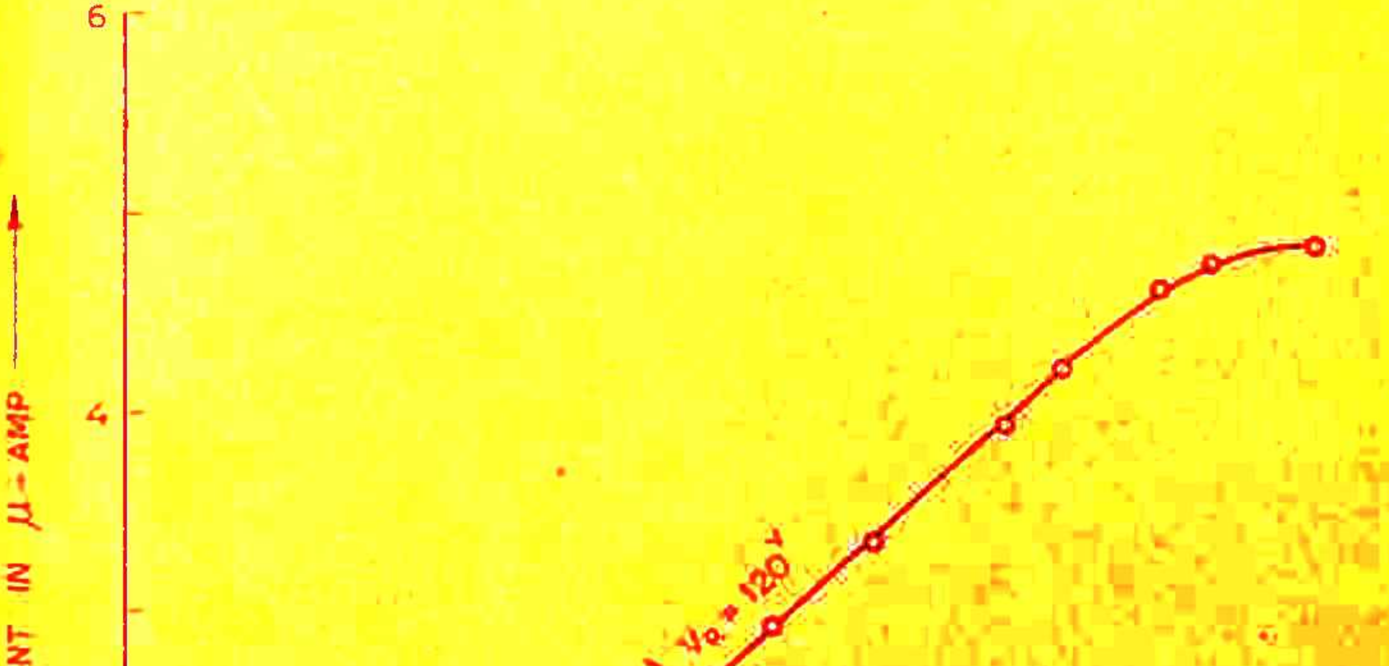


FIG. 4 REVERSE CURRENT DEGRADATION IN CD-29 DIODE

Germanium family DR - 10 is very usefull as a dosimeter upto 70000 rads (Fig. 2). All the above diodes recover immediately after the X-ray plant is switched off.

IRRADIATION OF TRANSISTORS:

An ionisation chamber is generally used as a radiation dosimeter, but it is very difficult to handle it. Here the study of the quasipermanent effects of the irradiated semiconductor devices and observations on the variation of electrical parameters due to irradiation of transistors are presented. AC 187 (pnp) Germanium, AC 125 (npn) Germanium, and other transistors (Table 1), were irradiated by 46 kv photon flux X-rays from Strahlenschutzzulassing PTB 508 (Germany) X-ray plant. Masafumi Yamaguchi and Osamu Nagai[10] examined the application of radiation damage of silicon planar npn transistors of type 2SC 33. Common emitter dc gain h_{FE} was measured because $1/h_{FE}$ was sensitive parameter. At lower dose $\triangle(1/h_{FE})$ was not proportional to radiation dose since changes of surface states were dominant. But at large doses, the lattice damage became dominant and $\triangle(1/h_{FE})$ was proportional to number of recombination centres induced by irradiation.

In the present study BEL transistors were mostly irradiated for dosimetric study under various bias conditions depending on the specifications given by the manufacturers. (Table 1). In the case of AC-187 (npn) Germanium and AC-125 (pnp) Germanium, the observations were very exciting in the sense that the transistor gain h_{FE} increased linearly with time (i.e. dose) which is very rare phenomenon in transistors[8], but a very useful phenomenon for developing X-ray dosimeters. These transistors recover immediately after the irradiation is stopped. In the case of AC-188 the increase in h_{FE} with X-ray dose was slow (Fig. 5). BC-108, BC-109, CIL-522, CIL-523 and CIL-591 showed a usual decrease in the h_{FE}. In the case of BC-108 and BC-109 (Fig. 6) the fall in h_{FE} with irradiation time was quite rapid but not strictly linear. CIL-522, CIL-523 and CIL-591 showed very slow degradation in h_{FE} (Fig. 7) and are recommended for Gamma-ray dosimetry. Regarding the change in the parameter I_{CBO}, the AC-187, AC-125 and AC-188 gave linear relationships (Fig. 8) with X-ray dose, but in different ranges of irradiation time and can be used as dosimeters. Amongst the other, transistors, CIL-522, CIL-591 and CIL-523 show approximately linear responses after a time gap (Fig. 9) but BC-108 and BC-109 do not come upto the expectations (Fig. 10).

DISCUSSION:

Ionization due to X-radiation creates localized charged energy states on semiconductor surfaces. As a result of changes in surface potential, the surface recombination may be increased causing device degradation[7-11]. Channel formation may occur at any pn junction. When a diode is under reverse bias, the reverse current increases because the channel increases the effective area of junction[7-8]. The I_{CBO} for the transistor behaves in a similar way to diode-junction reverse current. Hence the I_{CBO} is also increased by the formation of a channel across the base region so as to provide leakage path from emitter to collector.

The current gain ($h_{FE} = \frac{I_c}{I_b}$) of transistor is affected by channels as well as by surface recombinations in the region of the emitter base junction. The common emitter gain h_{FE} is influenced through I_b. I_b is increased to supply majority carriers for recombination because the base transport factor of a transistor is decreased due to increased surface recombination at the base surface[9-11]. Hence h_{FE} is decreased . But if the channel extends across the entire base surface from the collector to the emitter, the surface recombination will be reduced due to reduction in the concentration of holes[9]. Although a large increase in I_{CBO} would result but under these conditions the h_{FE} may actually increase since the increase in I_{CBO} would appear as an increase in I_c without any increase in I_b. This has happened with devices AC-187 (npn) Germanium and AC-125 (pnp) Germanium (Fig. 5). In the case of CIL-522 (npn) silicon and others (Fig. 6, 7) the h_{FE} decreased as usual and I_{CBO} increased more or less linearly with dose after 10 minutes of irradiation (Fig. 8, 9, 10). It was also observed that Germanium AC-125 (pnp) showed saturation of I_{CBO} while silicon CIL-522 did not. The I_{CBO} in the case of silicon transistors goes on increasing linearly with dose[11].

RESULTS:

The h_{FE} improvements in transistors AC-187 and AC-125 and I_{CBO} changes in all samples under study except BC-108 and BC-109 after irradiation and have been recommended for X-ray dosimetric purposes.

TABLE - 1.

Bias Conditions during the measurements of I_{CBO} and h_{FE}

Parameter	Device	Bias Conditions
h_{FE}	AC 125	V_{cE} = 1 volt I_c = 5 mA
	AC 187	V_{cE} = 1 volt I_c = 5 mA
	AC 188	V_{cE} = 1 volt I_c = 5 mA
	CIL 522	V_{cE} =10 volts I_c = 10 mA
	CIL 523	V_{cE} =10 volts I_c = 10 mA
	CIL 591	V_{cE} = 5 volts I_c = 10 mA
	BC 108	V_{cE} = 5 volts I_c = 5 mA
	BC 109	V_{cE} = 5 volts I_c = 5 mA
I_{CBO}	AC 125	V_{cB} =20 volts
	AC 187	V_{cB} =15 volts
	AC 188	V_{cB} =20 volts
	CIL 522	V_{cB} =25 volts
	CIL 523	V_{cB} =15 volts
	CIL 591	V_{cB} =10 volts
	BC 108	V_{cB} =25 volts
	BC 109	V_{cB} =25 volts

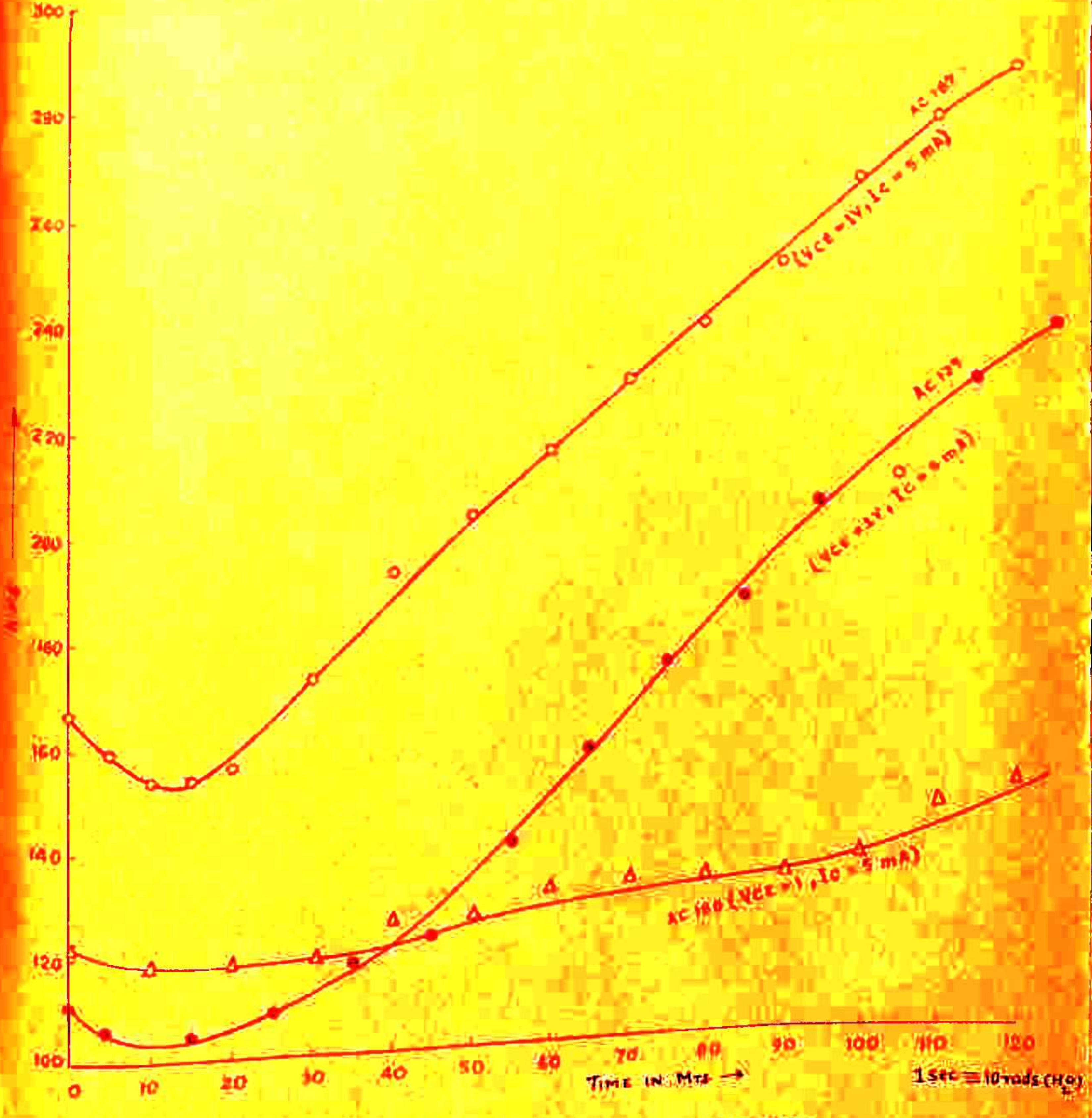


FIG. 5 hFE IMPROVEMENT IN TRANSISTORS

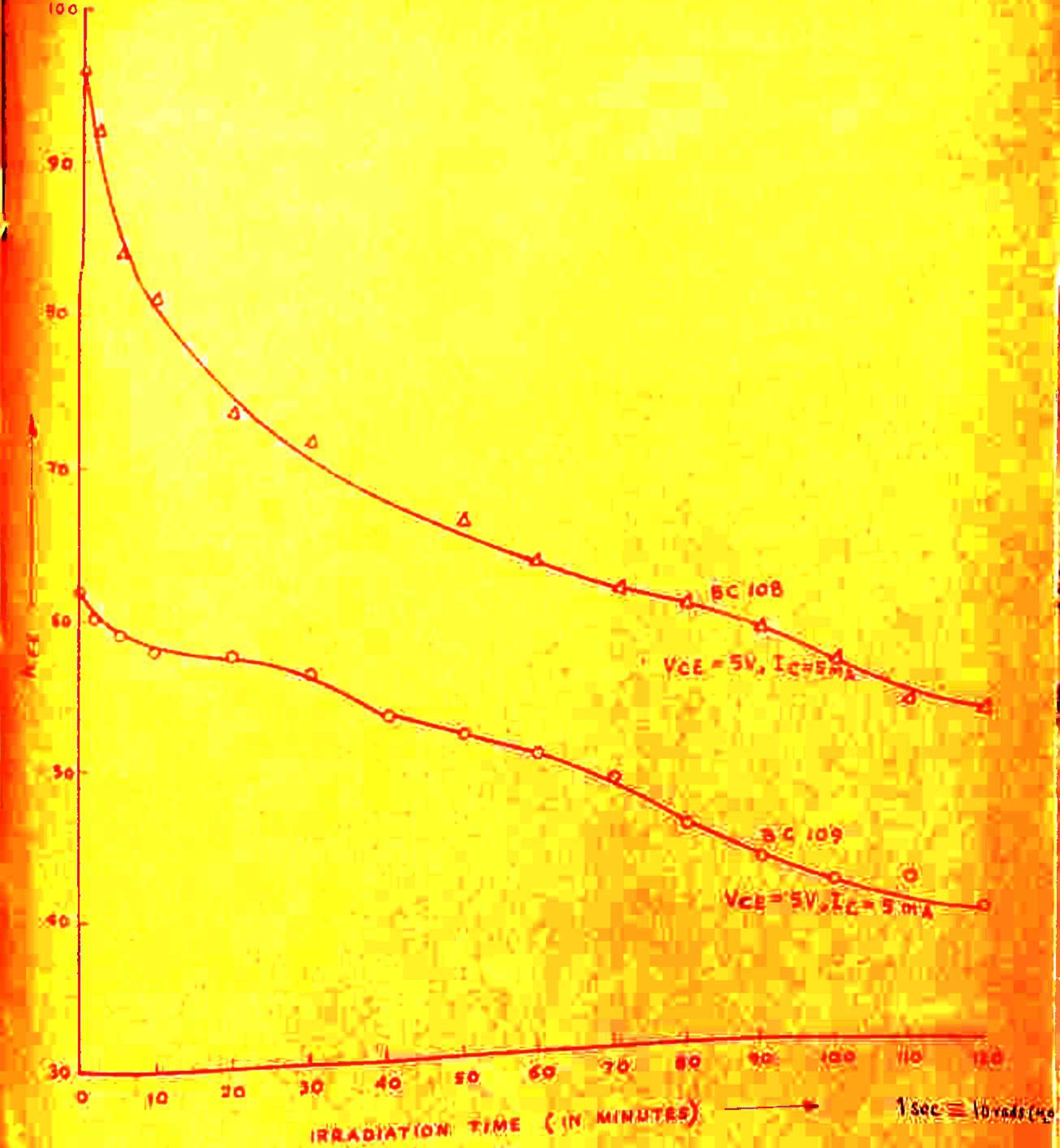


FIG. 6   hFE DEGRADATION IN TRANSISTORS.

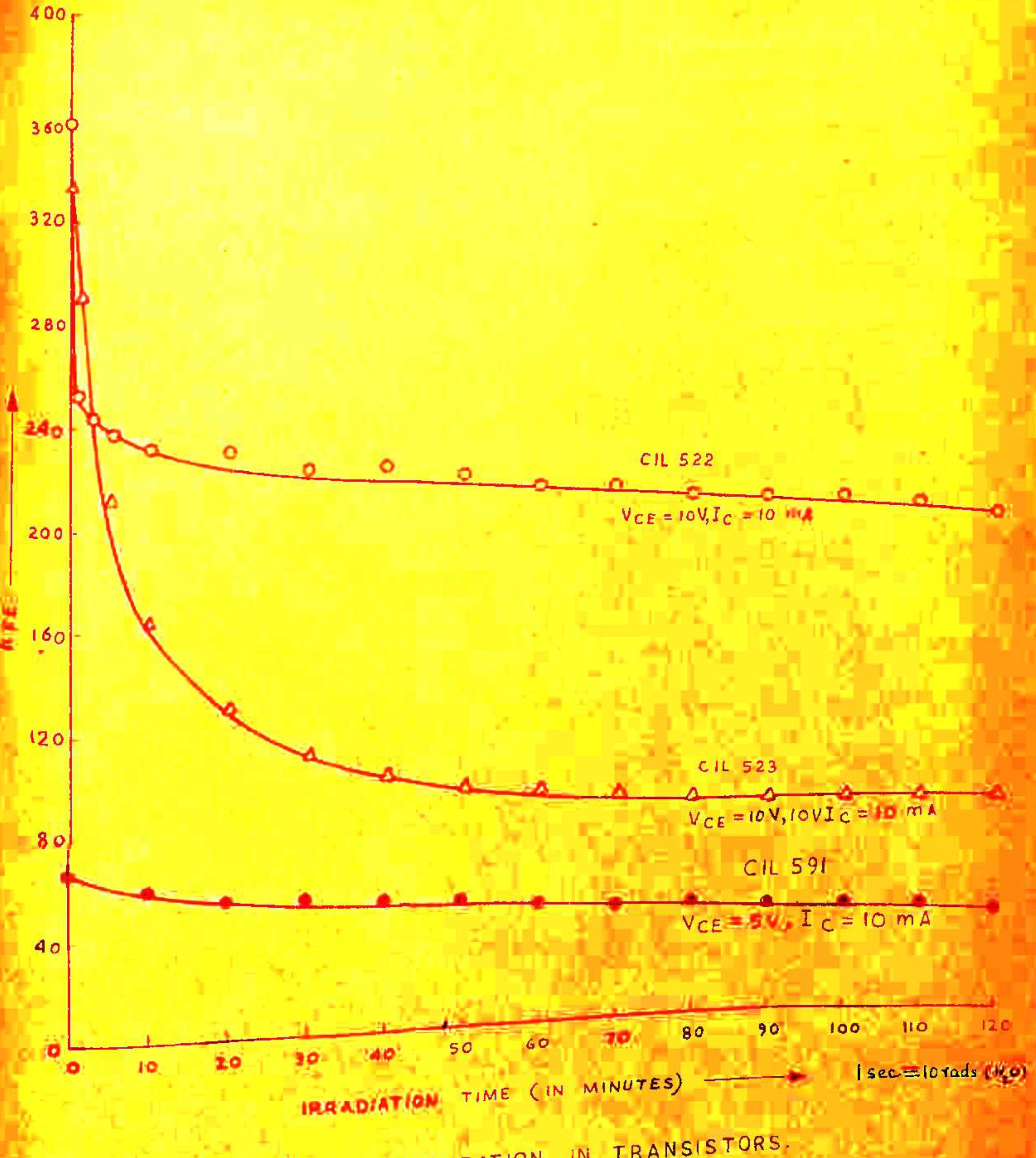


FIG. 7 h_{FE} DEGRADATION IN TRANSISTORS.

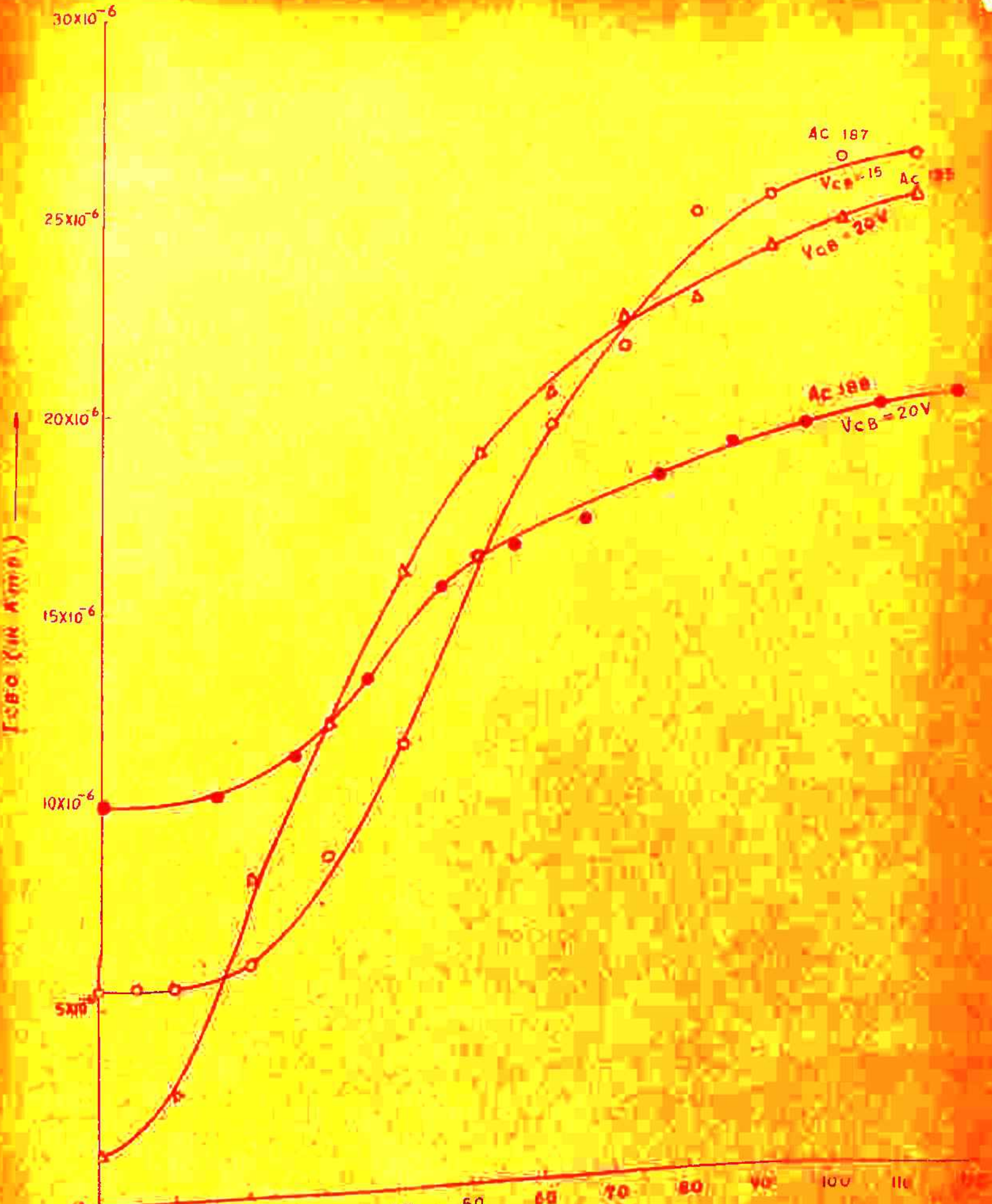


FIG. 8 I_{CBO} DEGRADATION IN TRANSISTORS.

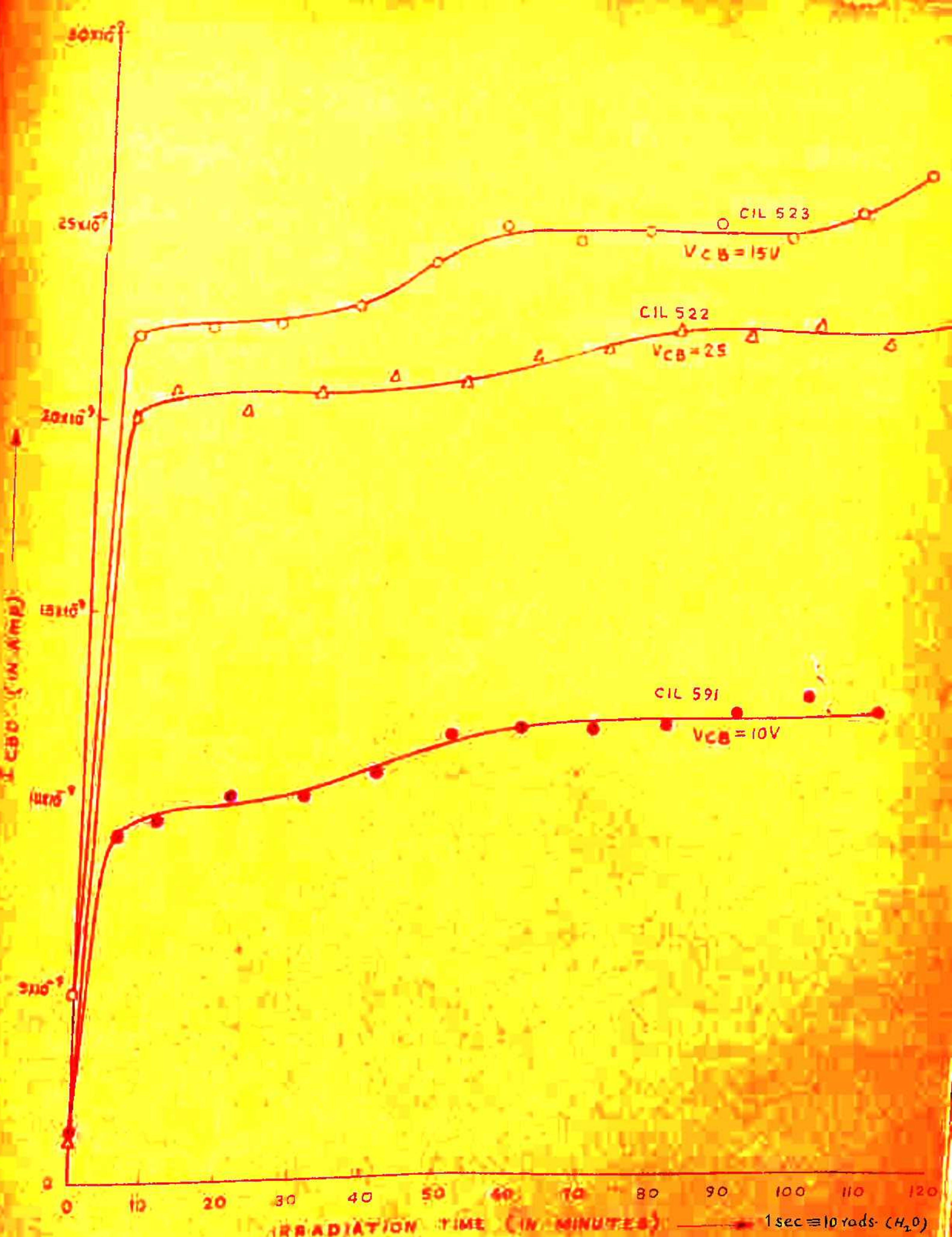


FIG. 9 I_{CBO} DEGRADATION IN TRANSISTORS

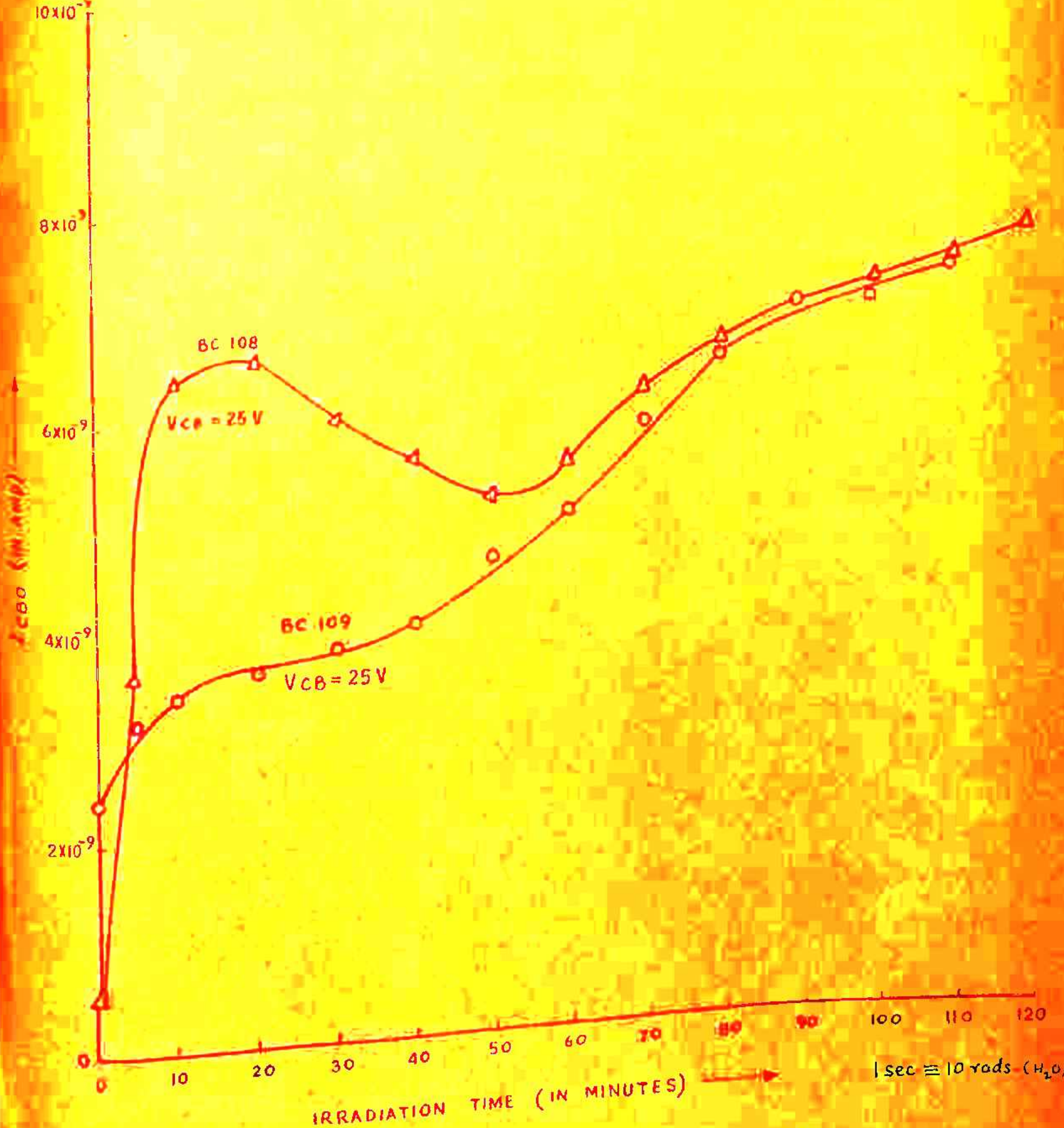


FIG. 10 I_{CBO} DEGRADATION IN TRANSISTORS.

EFFECT OF X-RADIATION ON STORAGE TIME OF DIODES:

The storage time, t_s is an important parameter in transient and high frequency performance of diodes[12]. Therefore, the study of this parameter also becomes important inorder to know the behaviour in fast electronic circuitry put into radiation environment. Silicon planar diodes CD 29 and CD 38 were selected for this study. The circuit used for this is shown in Fig. 11. By adjusting E_1 and potentiometer P_1, a forward current in the test diode D is adjusted. E_2 and P_2 along with R_2 are used to balance out the initial thermionic emission current of the vacuum diodes. On top of this d.c. bias, the diode is driven by a square wave capable of reversebiasing the diode. When the diode D is forward biased, all the vacuum diodes conduct, shorting the output resistance R_3 and allowing the forward current to pass through the vacuum diodes. When the test diode D is reverse biased, the vacuum diodes are cut off and the current through the diode, D,can be measured across R_3 directly. Thus the constant current period t_s can be measured,(Fig. 12). The photographs of the C.R.O. patterns are taken by high speed polaroid camera used at a desired time. The storage time, t_s is measured from the photographs. The calculated values are given in Tables 2 and 3.

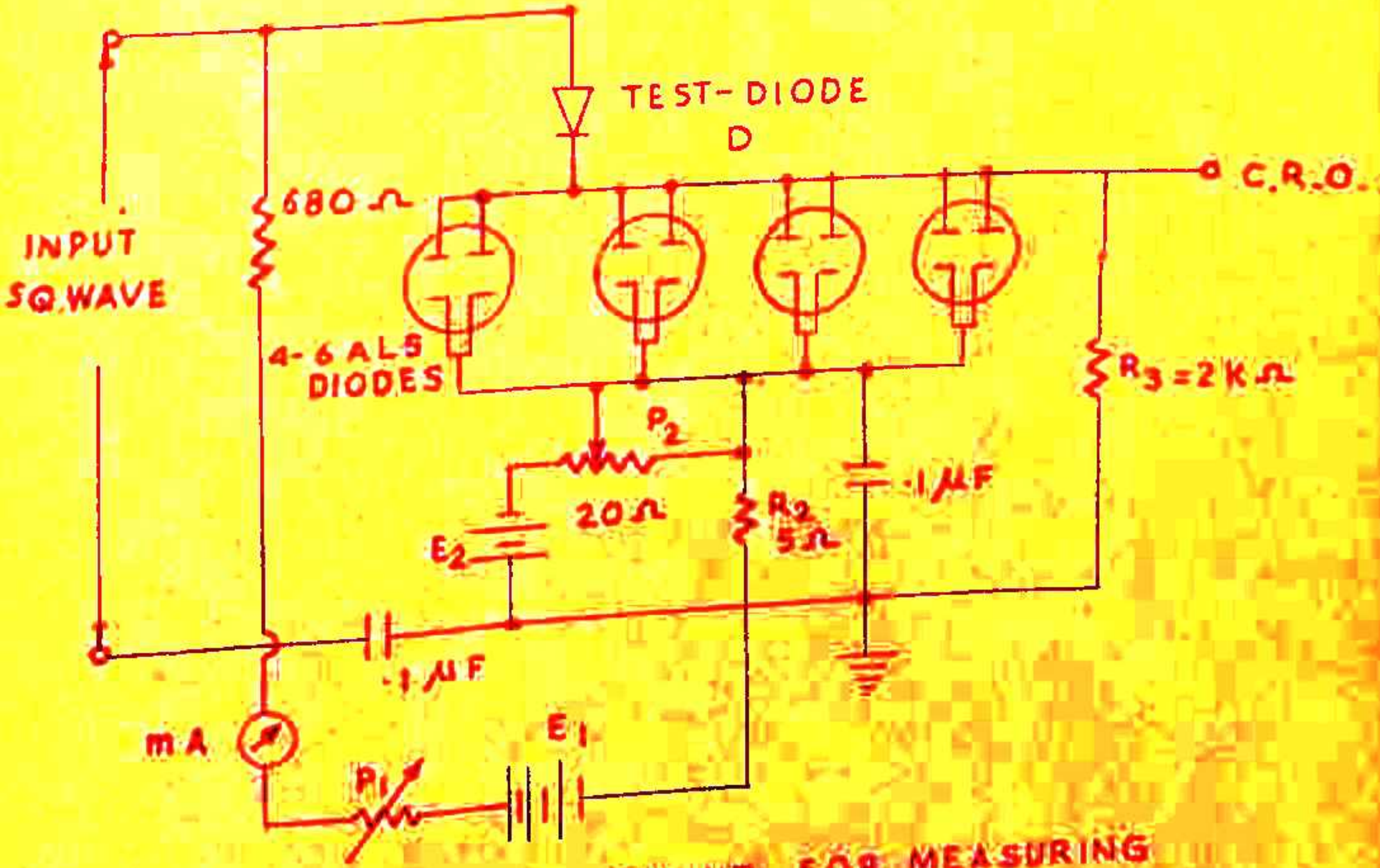


FIG. 11 EXPERIMENTAL SET-UP FOR MEASURING RECOVERY-TIMES.

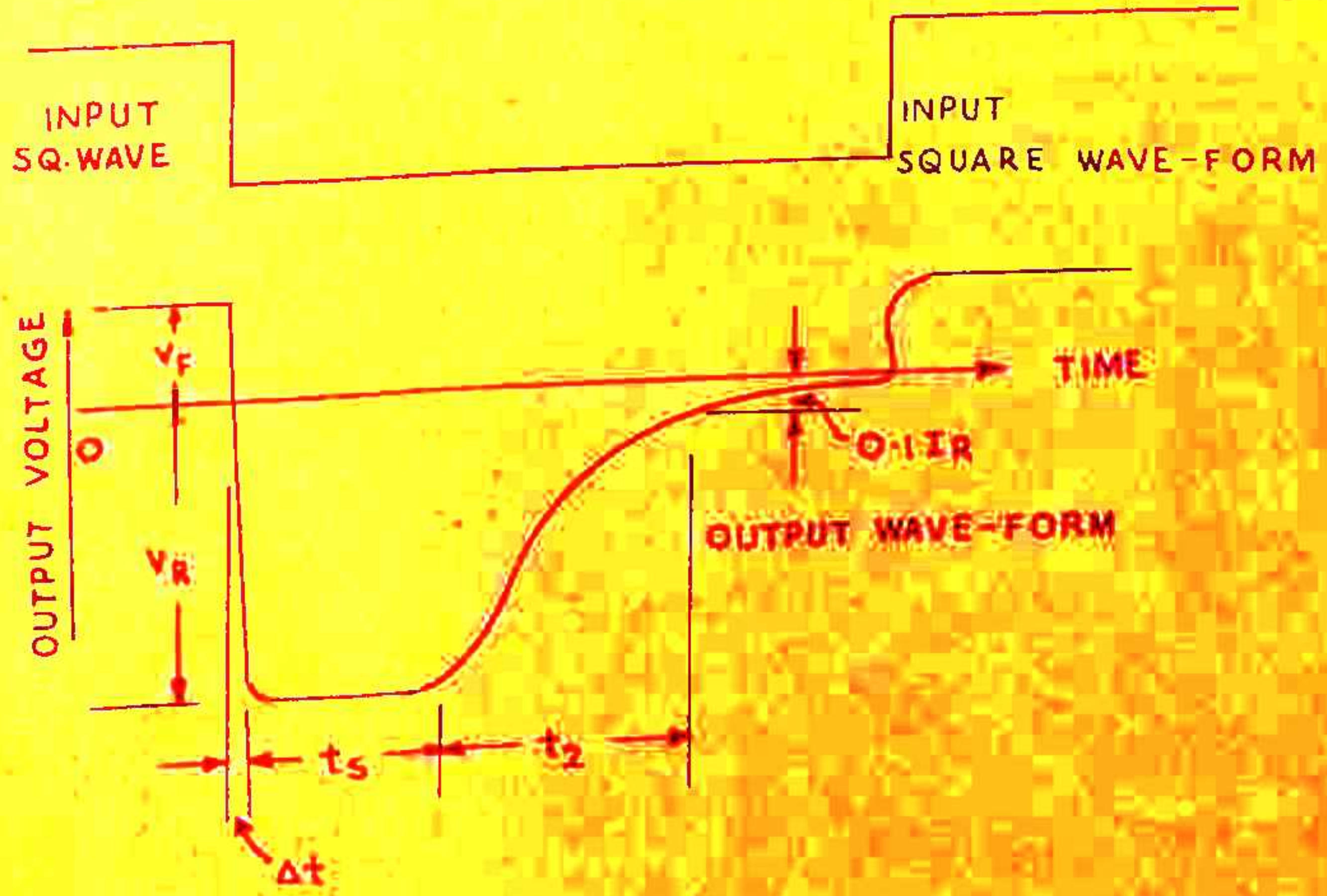


FIG. 12 WAVE-FORM IN CIRCUIT OF FIG. 11.

DISCUSSION:

When the device is irradiated by X-rays, generation of electron-hole pair can occur within the silicon diode. Under the influence of junction field, there will be separation of charges resulting in the inversion of the surface of the cathode region near the junction. The junction between p-type channel and n-type cathode is formed. The channel on the surface of the base is connected ohmically to the diffused emitter junction. It is shown by Reddi[14] that the application of forward bias can result in the injection of carriers not only from the anode region, but also from the channel. This concept can be made use of in identifying the cause for an increase of ts in X-ray radiations as follows: when a reverse voltage, V_R is applied to a diode, junction remains under forward bias until the stored charge in the n-region is removed such that the edge concentration falls to zero. During the process of recovery, there is no injection from the anode. However, the injection of minority carriers from the channel continues which gives excess carriers and thereby delaying the process of recovery. This mechanism accounts for the increase in storage time t_s.

TABLE-2.

Diode CD 38

Time Scale in Tektronics oscilloscope: 1 cm = 0.25 micro-se

Irradiation time in minutes	Base length in cms.	Storage time t_s in micro-sec.
0	2.9	0.725
30	3.3	0.825
60	3.4	0.850
90	3.5	0.875
120	3.6	0.900
150	3.7	0.925

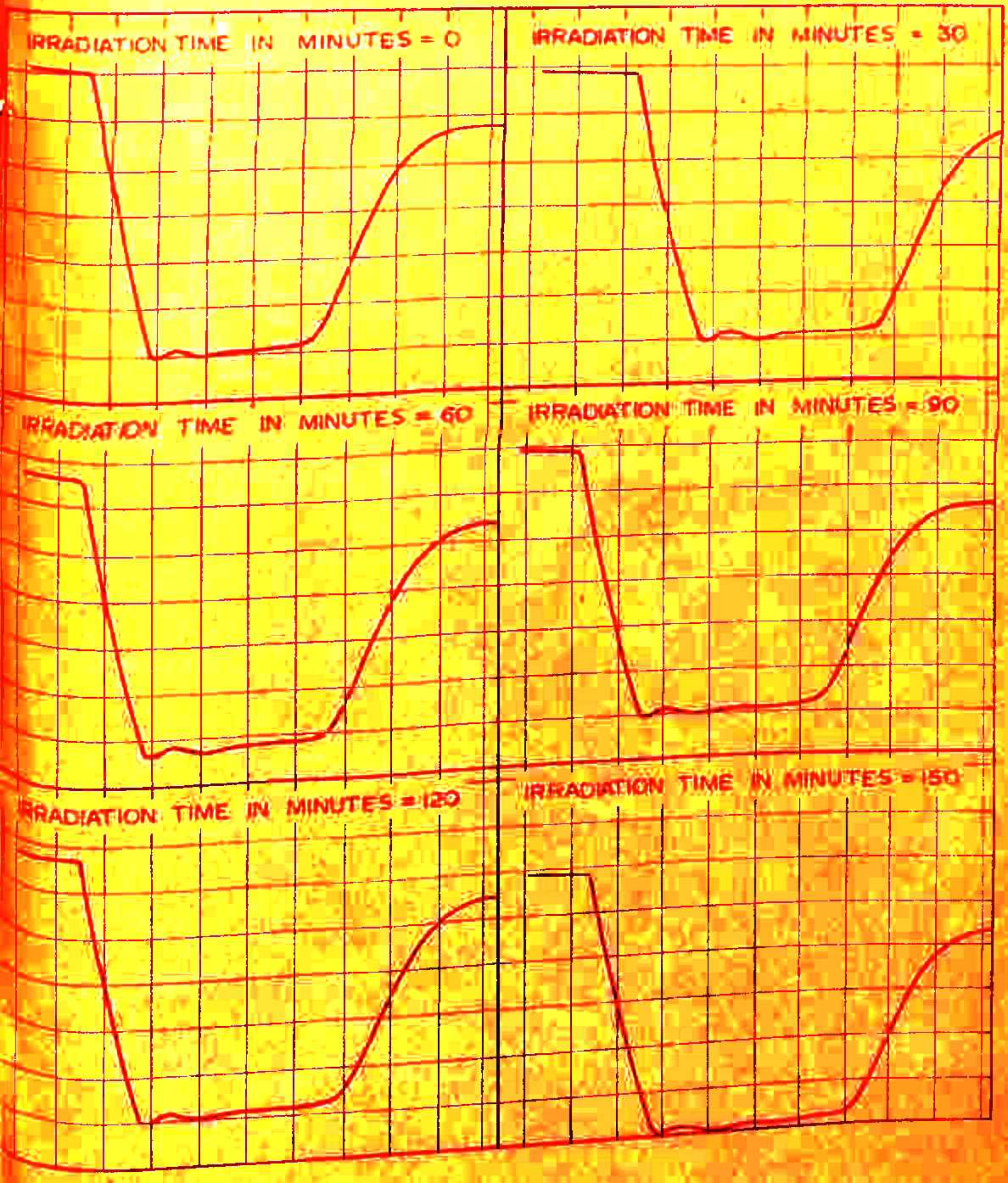
TABLE - 3.

Diode CD 29

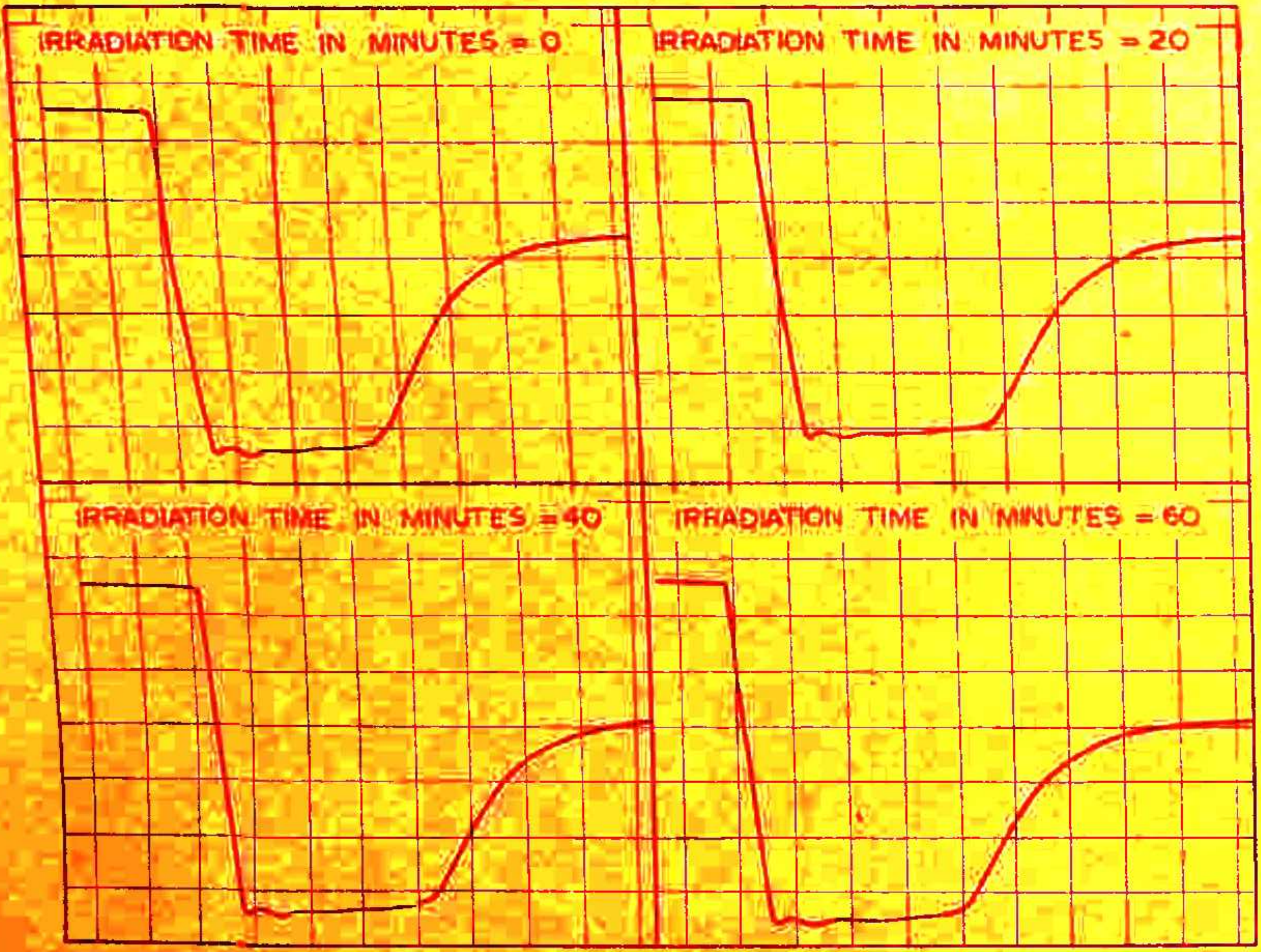
Time Scale, 1 cm = 0.25 micro-sec.

Irradiation time in minutes.	Base length in cms	Storage time t_s in micro-sec.
0	2.55	0.637
20	2.80	0.700
40	2.90	0.725
60	3.00	0.750

DIODE CD - 38 TIME SCALE, 1 CM = 0·25 MICRO-SEC



DIODE CD-29 TIME SCALE, 1 CM=0·25 MICRO-SEC


IRRADIATION TIME IN MINUTES = 0

IRRADIATION TIME IN MINUTES = 20

IRRADIATION TIME IN MINUTES = 40

IRRADIATION TIME IN MINUTES = 60

INSTRUMENTS USED FOR THIS STUDY:

1. Electronic Volt-ohmmeter Phillips GM6001.
2. D.C. Micro-Voltmeter Phillips GM6020.
3. Systronics transistor power supply, Ahmedabad.
4. Type 502A double beam oscilloscope (Tektronics Inc. Portland Oragan, U.S.A.).
5. Oscilloscope camera C-27 (Tektronics).
6. Polaroid land film type 107.
7. 211B, Square wave generator (Hewlet Packard).

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CHAPTER 9.

SUMMARY OF RESULTS

SUMMARY OF RESULTS

One of the most important problems dealt with here is the proper choice of a dosimetry system. For absorbed dose measurements, the dosimetric characteristic which is of greatest importance is its X-ray energy response. Other characteristics which are of secondary importance are the size, space and its compatibility with the environment in which the dosimeter is to be used. In many instances a dosimeter must be able to withstand highly corrosive and erosive environment and extremes in temperature and humidity. Besides that in deciding what to record and what not to record the aim has been to measure only those parameters which come within the capabilities of available instruments. During this study the intention had been to develop some new X-ray dosimeters such that the response to dose is mostly linear. Although many dosimeters have come up to this expectations and even those that give sub-linear but steady response have been equally appreciated. The solid state dosimeters (diodes and transistors) that have also been selected will sustain their reputation for medium dose ranges. It is certain that they are going to be very useful in the space research programmes, atomic power stations and radiology. A summary of the various dosimeters investigated in this work is given ahead in tabular form.

Characteristics of X-ray dosimeters developed and described in this work

System	Radiation effect	Method of analysis	Normal range in rads.	Dose relation-ships	Comments
Salts of Amino acids (L Arginine and L(+)Lysine)	Induced asymmetry due to changes in the stiffness of bonds	Measurement of changes in optical rotation	50 K rads to 400 K rads	Relationship between dose absorbed and molecular rotation is linear.	Very stable dosimeters.
D(+) Camphoric acid	"	"	100 K rads to 400 K rads	Sublinear relationship	"
Maltose	"	"	50 K rads to 900 K rads	"	Stable and cheap
D(+) Xylose	"	"	50 K rads to 250 K rads.	"	"
D(+) Galactose	"	"	50 K rads to 350 K rads	Linear relation	Sensitive to temperature changes
D Fructose	Change in refractive index	Measurement by refractometer	50 K rads to 250 K rads	Linear relation	"
L(+) Arabinose	"	"	"	"	"

Polymers (Plastics in organic solvents)

PVC in cyclohexane	Induced conductivity	Measurement of ionization current	25 K rads to 250 K rads	Current increases with dose $I=(aV+b)R$	Very stable useful for stronger dose
Polystyrene in CCl_4	,,	,,	2.5 K rads to 45 K rads	,,	,,
PMMA in CCl_4	,,	,,	Very much resistant to X-radiation	,,	Recommended for gamma rays dosimetry

Cellulosics as aqueous dosimeters

CMC	Change in viscosity and feeble ionization	Flow rate measurement through Ostwald's viscometer and measurement of ionization current	500 K rads to 5000 K rads	Linear response	Suitable for very high doses.
Methyl cellulose	,,	,,	7000 K rads to 35000 K rads	,,	,,
Ethyl cellulose	,,	,,	1 M rad to 15 M rads.	,,	,,
Sodium Alginate	,,	,,	,,	,,	Suitable for high X-ray doses only. Dissolved air has no effect

Optical Density Dosimeters

Sucrose (2.5 M) Aqu. sol.	Change in optical density	Measurement of O.D. at λ=565 Mμ	From 0 to 90 K rads	Linear relation	Degree of accuracy is very high
Xylose (5M)	"	" λ=555 Mμ	"	"	"
Levulose (M)	"	" λ=280 Mμ (u.v. region)	0 to 300 K rads	"	"

Degradation in melting point (dosimeters)

Sucrose (Crystals)	Degradation in melting point	Measurement of changes in melting point by Capillary appa.	0 to 40 M rads	Linear relation	Not very reliable for want of 100% pure samples
D Fructose D Glucose	"	"	"	"	Quite encouraging study.

Thin Film Capacitors

Tantalum oxide	Increase in dielectric conductance	Measurement of leakage current at constant voltage	50 K rads to 100 K rads	Sublinear relation	Very reliable, Sensitive but susceptible to humidity
Mylar	"	"	Up to 300 K rads	"	"

Solid State Dosimeters.

Diodes CD 29 (Silicon) CD 28 ,, DR 25 (Germanium) DR 10 ,,	Transient changes in junction capacitances	Measurement of reverse biased leakage current	1) CD 29, from 0.2 K rads to 80 K rads	Linear relation	Good dosimeter Immediate recovery very suitable for low ranges
			2) CD 28, from 0.7 K rads to 2.5 K rads	,,	,,
			3) DR 25, 0 to 70 K rads	,,	Very reliable dosimeter.
			4) DR 10, 0 to 70 K rads	,,	Best amongest the diode dosimeters
Transistors AC 187 (npn, Ger.) AC 125 (pnp, Ger.)	Variation in electrical parameters	Measurement of changes in I_{CBO} and h_{FE}	AC187, 0 to few AC125, kilorads	Linear relation	Extremely good dosimeters for medium ranges.
CIL 522 CIL 501 CIL 523	,,	Measurement of changes in I_{CBO}	0 to few kilorads	,,	,,

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