CHAPTER-III

DIELECTRIC HYSTERESIS

3.1 INTRODUCTION

The dielectric hysteresis is the most important characteristic of ferroelectric materials. The hysteresis loop of these materials enables us to study two outstanding parameters, coercive field (Ec) and spontaneous polarization (P_s). Merz (1954) defined the coercive field as the field for which the probability of nucleating antiparallel domains starts increasing very rapidly. According to Wider (1955) the coercive field is the magnitude of the external applied electric field required for switching of the polarization. The coercive field of $BaTiO_3$ was reported by Janovec (1958). Abe (1960) determined it by assuming theoretically the velocity of forward growth of the domains. Sawada et al(1961) studied the temperature dependence of coercive field of KNO_3 crystal. Bertaut and Lissalde(1967) observed hysteresis loops in ferroelectrics by X-ray intensities and measured true coercive fields in yttrium and rate earth manganates. The second parameter i.e. spontaneous polarization (P_s) can be defined as a polarization shown by the ferroelectric material even in the absence of an external applied electric field. Gavrilyachenko et al (1970) were measured these two parameters from hysteresis loop of lead titanate crystal at $20^{o}C$ (for $1KH_z$). Time effects in hysteresis loop of $(Pb\text{-}Sr)TiO_3$ were studied by Griffith's et al (1972). An investigation was made of non-linear polarization in solid solutions of system $(Ba\text{-}Sr)TiO_3$ and $(Pb\text{-}Sr)TiO_3$ by Gristsberg et al (1977). Kuroda and Kubota (1980) studied the diffuse phase

transition of $(Ba_xSr_{1-x})Nb_2O_6$ doped with Pr^{3+} and Nd^{3+} by measuring the spontaneous polarization. Jamadar et al (1986) were studied dielectric, and hysteresis properties of ferroelectric solid solutions, lead-barium titanate and lead strontium titanate. The dielectric properties of $NaVO_3$, KVO_3 and their solid solutions were studied by Patil et al (1988). Recently, Patil et al (1989) studied dielectric hysteresis and measured coercive fields of undoped and Dy_2O_3 doped KVO_3. Similarly Kashid et al (1989) also studied dielectric properties of undoped and Gd_2O_3 doped KVO_3.

3.2 HYSTERESIS LOOP

The distinguishing feature of the ferrolectrics from pyroelectrics is that the spontaneous polarization that can be reversed with an applied electric field. Valasek (1920) observed polarization reversal by dielectric hysteresis. The hysteresis loop is a common feature of all known ferroelectrics. The shape of the hysteresis loop depends strongly upon purity of the material and magnitude of an external applied electric field. The hysteresis loop can be described by following parameters. (i) The spontaneous polarization P_s, which is the polarization present in the ferroelectric material even in the absence of external field. As temperature decreases, P_s, unusually increases rapidly on crossing the Curie point and reaches a saturation value at low temperature.(ii) The coercive field, Ec, which is already defined it depends not only on temperature but also on the measuring frequency and on the wave form of the applied field.(iii) The slope of the hysteresis loop connecting P & for value E = 0, equals the susceptibility χ obtained by measuring the polarization and applying the field along the ferroelectric axis.

Merz (1954) found the rate of reversal of polarization, which is given by the exponential law,

$$\frac{dP}{dt} = F(P)\, e^{-\alpha/E(t)} \qquad \ldots\ldots 3.1$$

where the function F(P) takes into account the fact that the switching rate depends on the extent to which the ferroelectric has already reversed its polarization. Three parameters usually measured from the hysteresis loop are, the coercive field Ec, the remanent polarization Pr and the saturation polarization P_s. These parameters are functions of frequency and temperature. Specifically, they decrease to zero values as the temperature approaches Tc, where the transition occurs from the ferroelectric to paraelectric phase.

3.3 EXPERIMENTAL DETAILS

The appearance of proper hysteresis loop on the cathode ray oscilloscope (C.R.O) screen is the fundamental criteria for the ferroelectric material. The experimental arrangement for the study of hysteresis loop is a Sawyer Tower (1930) modified circuit as shown in Fig.3.1. The circuit consists of two oil filled condensers(Co) connected in series with the sample (Cx). A step-up transformer gives out put voltage of 700 volts. A gang condenser C is used in series with the sample pellet, which is mounted in a specially designed sample holder. The two sides of the pellets were coated with a thin layer of silver paste for good electrical contact. The temperature of the pellet is varied using electrically heated furnace having a temperature controller arrangement.

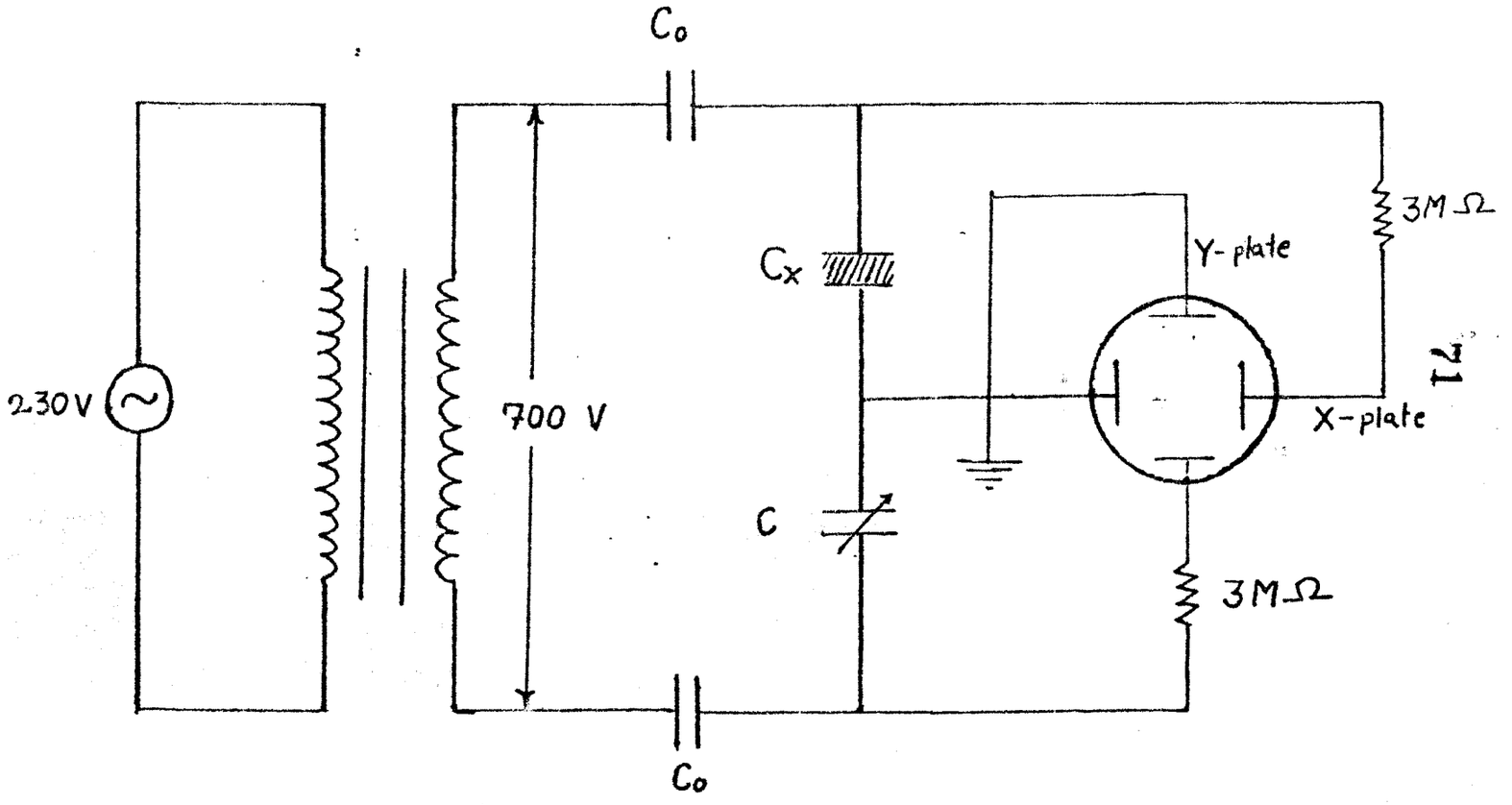


Fig.3·1 - MODIFIED FORM OF SAWYER-TOWER CIRCUIT FOR THE DISPLAY OF THE HYSTERESIS LOOP.

The damage of C.R.O can be avoided by making use of two high resistances each of 1 MΩ . The voltage across the sample pellet is 1kv/cm and frequency of an applied field as 50 H_z. The experimental set-up to observe the hysteresis loop on C.R.O is shown in Fig. 3.2. Initially, the scale of C.R.O is calibrated to measure polarization P on vertical axis and applied electric field along horizontal axis. P and E are more meaningful parameters for the study of dielectric hysteresis. The half width measurement of the hysteresis loop directly gives coercive fields at various temperatures. Similarly, vanishing of hysteresis loop at a particular temperature gives a Curie temperature of that sample. In brief, using this experimental set-up we have measured coercive fields of different samples alongwith their Curie temperatures.

3.4 HYSTERESIS LOOP OF UNDOPED KVO_3

The ferroelectric hysteresis loop of undoped KVO_3 was observed on the screen of C.R.O, at various temperatures by heating the sample in a pellet form inside a furnace. Fig.3.3(a,b,c,d) show the hysteresis loops for undoped KVO_3 at various temperatures. It is observed that the hysteresis loss is small at room temperature i.e. loop width is small at room temperature and as temperature increases the loop width gradually increases and quite above the room temperature practically it remains constnat. For further increase of temperture the loop width decreases and finally at a ferroelectric Curie temperature of pure KVO_3 it vanishes. The observed value of ferroelectric Curie temperature of pure KVO_3 is 322.5 C which is in agreement with the reported value of Curie temperature of pure KVO_3 by earlier workers.

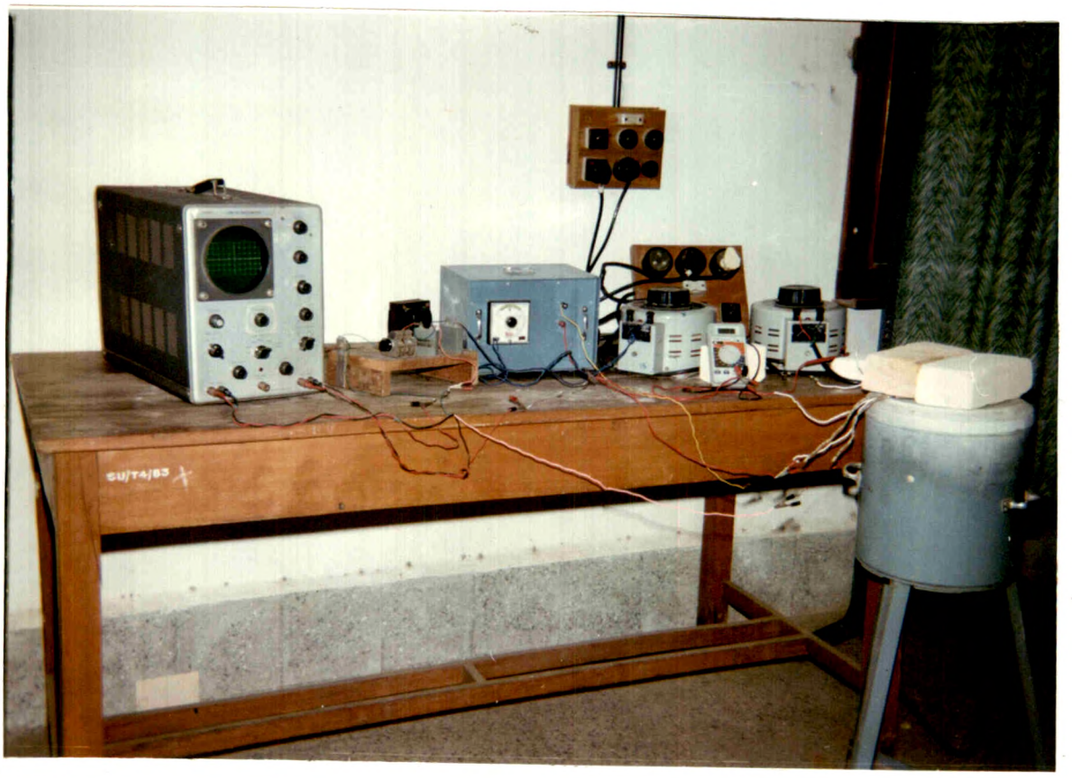


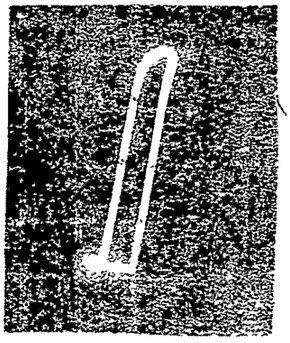
Fig. 3.2 :Experimental set-up for studies of the hysteresiss loop and coercive field(E_c).

3.5 HYSTERESIS LOOP OF Er_2O_3 DOPED KVO_3 SAMPLES

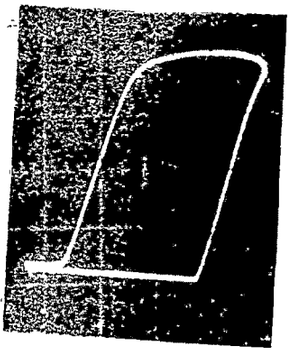
Ferroelectric hysteresis loops at various temperatures of KVO_3 doped with different concentrations (0.025 to 3 mol%) of Er_2O_3, were observed on the screen of C.R.O. The experimental arrangement for the same is shown in Fig. 3.1. The temperture dependent hysteresis loops for different samples are shown in Fig . 3.4(a, b,c,d) to Fig.3.9 (a, b,c,d). The shape of , hysteresis loop for these doped samples is also same as that of undoped KVO_3, however, hysteresis loops are vanishing at different temperatures. Figs. 3.4 to 3.9 show that the Curie temperature decreases with the increase of Fr_2O_3 content in KVO_3.

3.6 COERCIVE FIELD MEASUREMENT

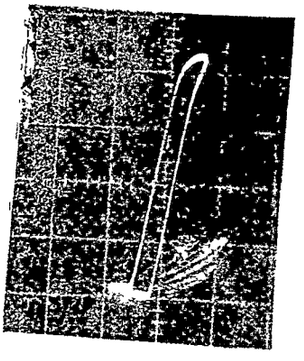
The study of the shape of hysteresis loop on C.R.O. screen enables us to measure the coercive field at various temperatures. Fig 3.3 to Fig.3.9 show the temperature dependent coercive fields for different samples of KVO_3 doped with Er_2O_3 having 0.025 mol% to 3 mol% concentratioon. The half-width measurement of the hysteresis loop gives directly coercive field of the sample. The observed coercive fields are different for different concentrations of Er_2O_3. It is seen that at room temperature the coercive field is small for all samples, further, as temperature increases coercive field slowly increases and quite above room temperature it remains constant. Then further as temperature increases coercive field slowly decreases and finally it falls to zero at Curie temperature as shown in Fig.3.10. Fig. 3.11 reveals us that Curie temperature of KVO_3 doped with Er_2O_3 decreases as Er_2O_3 content increases from 0.025 to 3 mol%. The peak values of coercive fields and Curie temperatures from Fig.3.10 and Fig. 3.11 are summarised in Table No.3.1



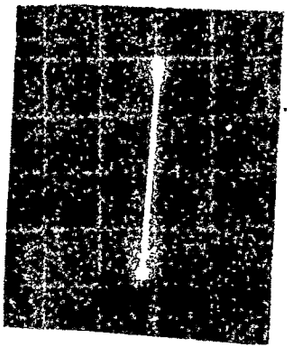
(a) Room Temperature



(b) 200°C

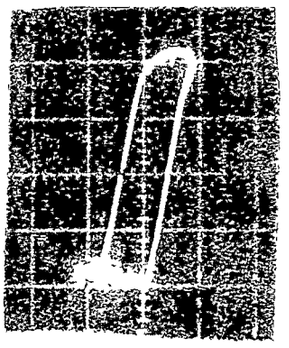


(c) 300°C

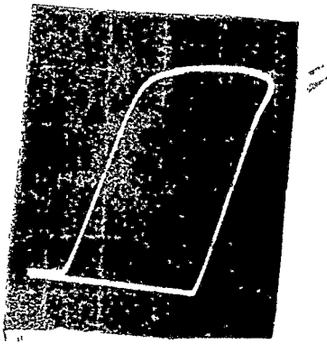


(d) 322.5°C

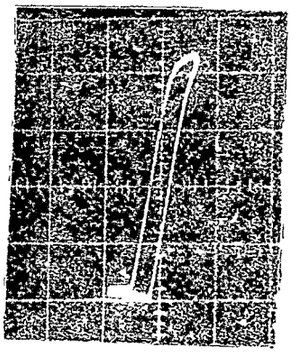
FIG. 5.3 : Ferroelectric hysteresis loop of undoped KVO_3. at different temperatures.



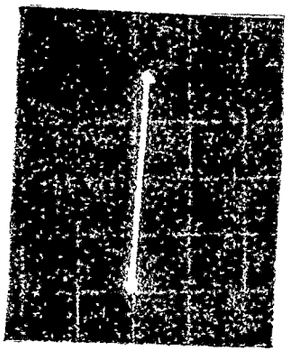
(a) Room Temperature



(b) 200°C



(c) 300°C



(d) 317.5°C

FIG. 3.4 : Ferroelectric hysteresis loop of 0.025 mol % Er_2O_3 doped KVO_3 at different temperatures.

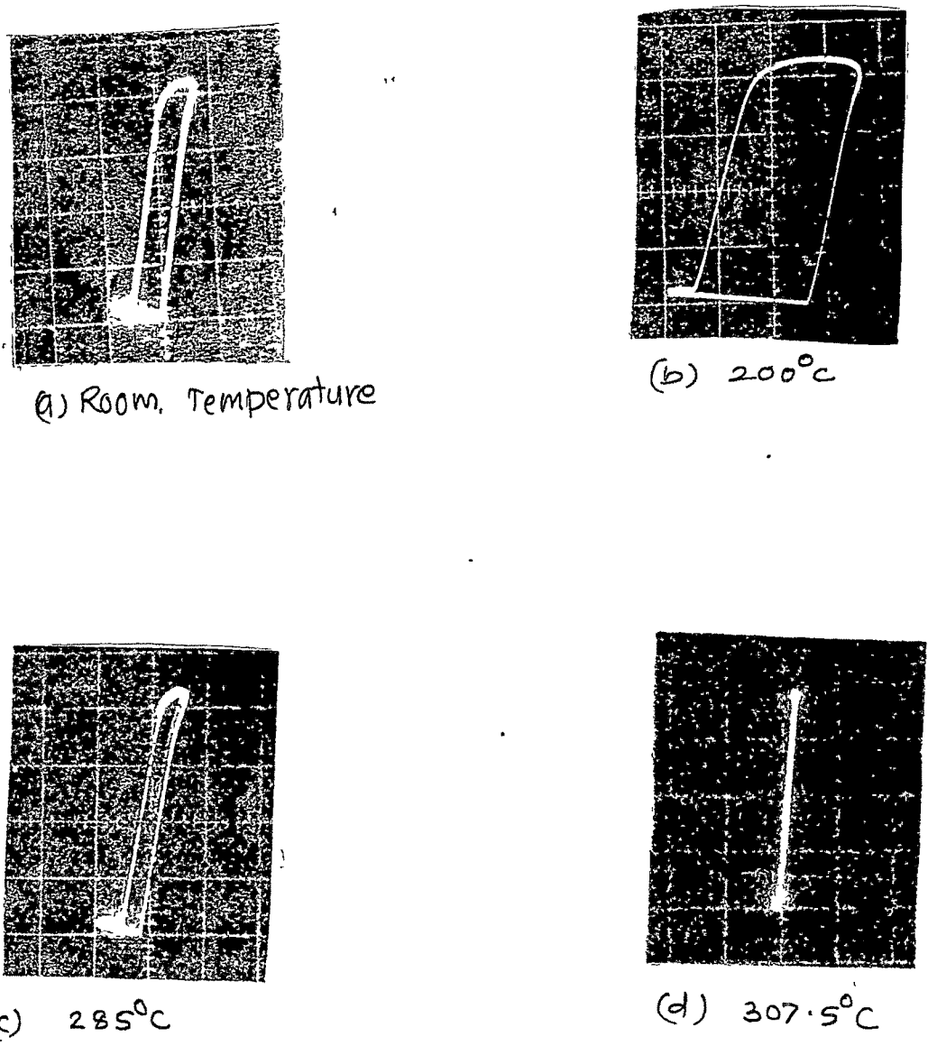
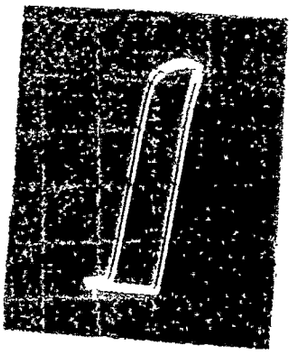
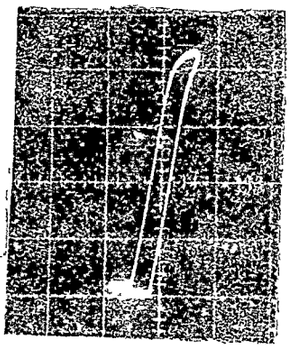


FIG. 3.5 : Ferroelectric hysteresis loop of 0.05 mol % Er_2O_3 doped KVO_3 at different temperatures.

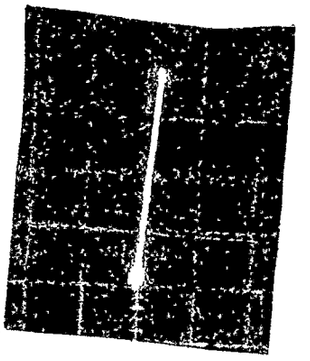


(a) Room Temperature

(b) 200°C



(c) 285°C



(d) 297.5°C

FIG.3.6 : Ferroelectric hysteresis loop of 0.1 mol % Er_2O_3 doped KVO_3 at different temperatures.

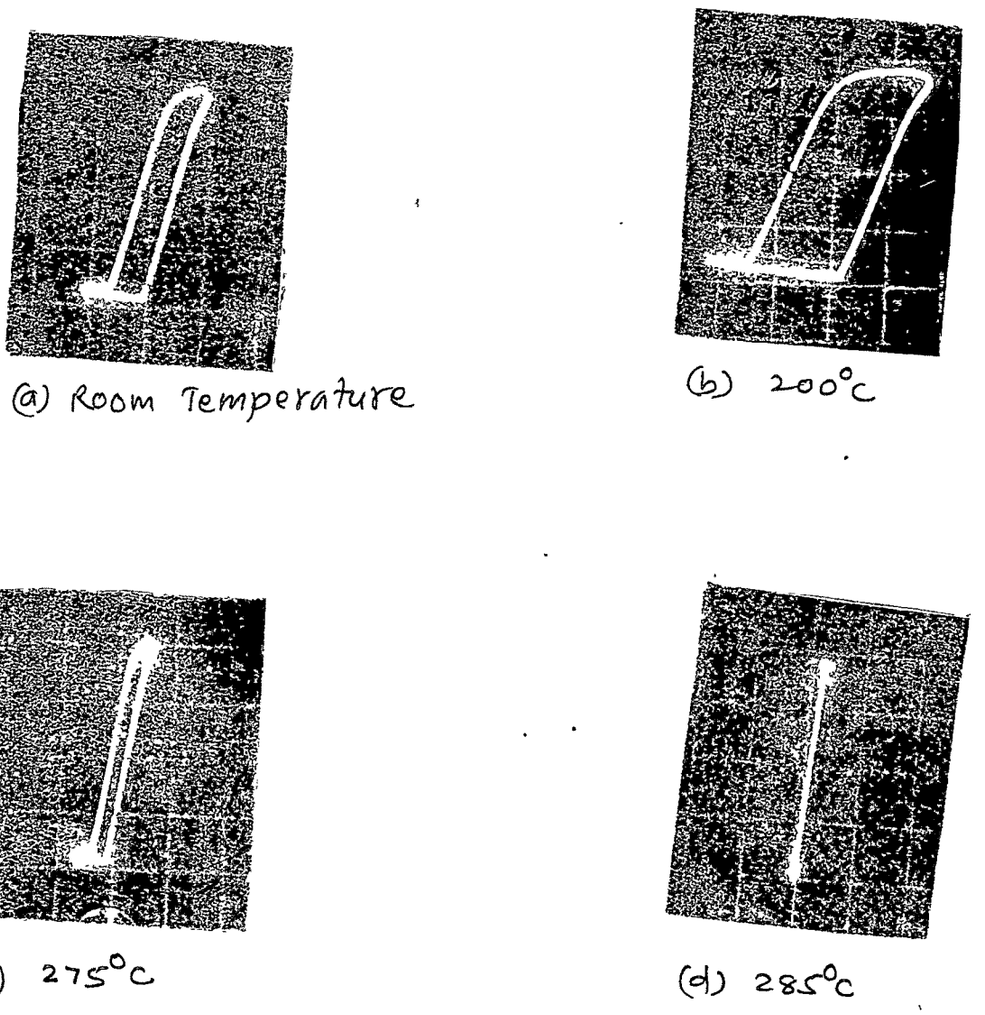
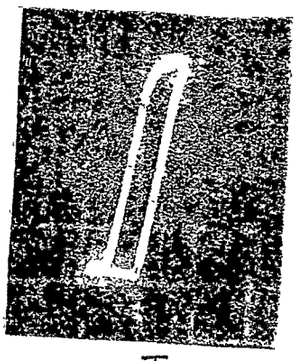
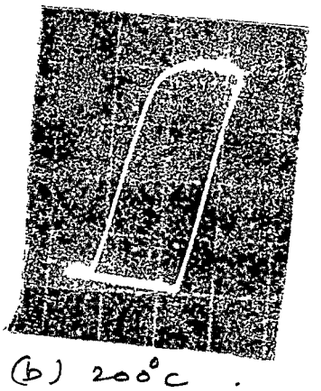


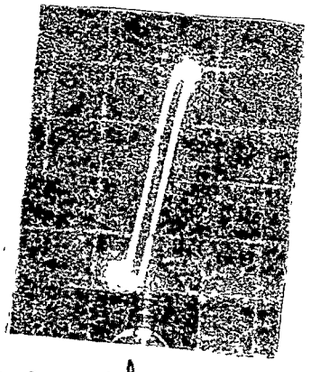
FIG . 3.7 : Ferroelectric hysteresis loop of 0.5 mol % Er_2O_3 doped KVO_3 at different temperatures.



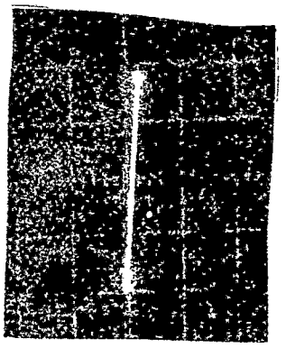
(a) Room Temperature



(b) 200°C

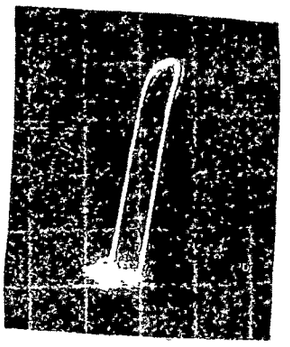


(c) 260°C

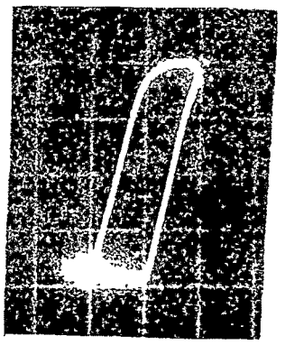


(d) 272.5°C

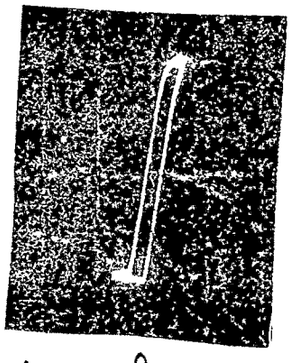
FIG.3.8 : Ferroelectric hysteresis loop of 1.0 mol%Er_2O_3 doped KVO_3 at different temperatures.



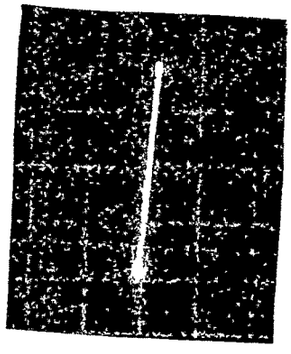
(a) Room Temperature



(b) 200°C



(c) 250°C



(d) 260°C

FIG. 3.9 : Ferroelectric hystersis loop of 3.0 mol % Er_2O_3 doped KVO_3 at different temperatures.

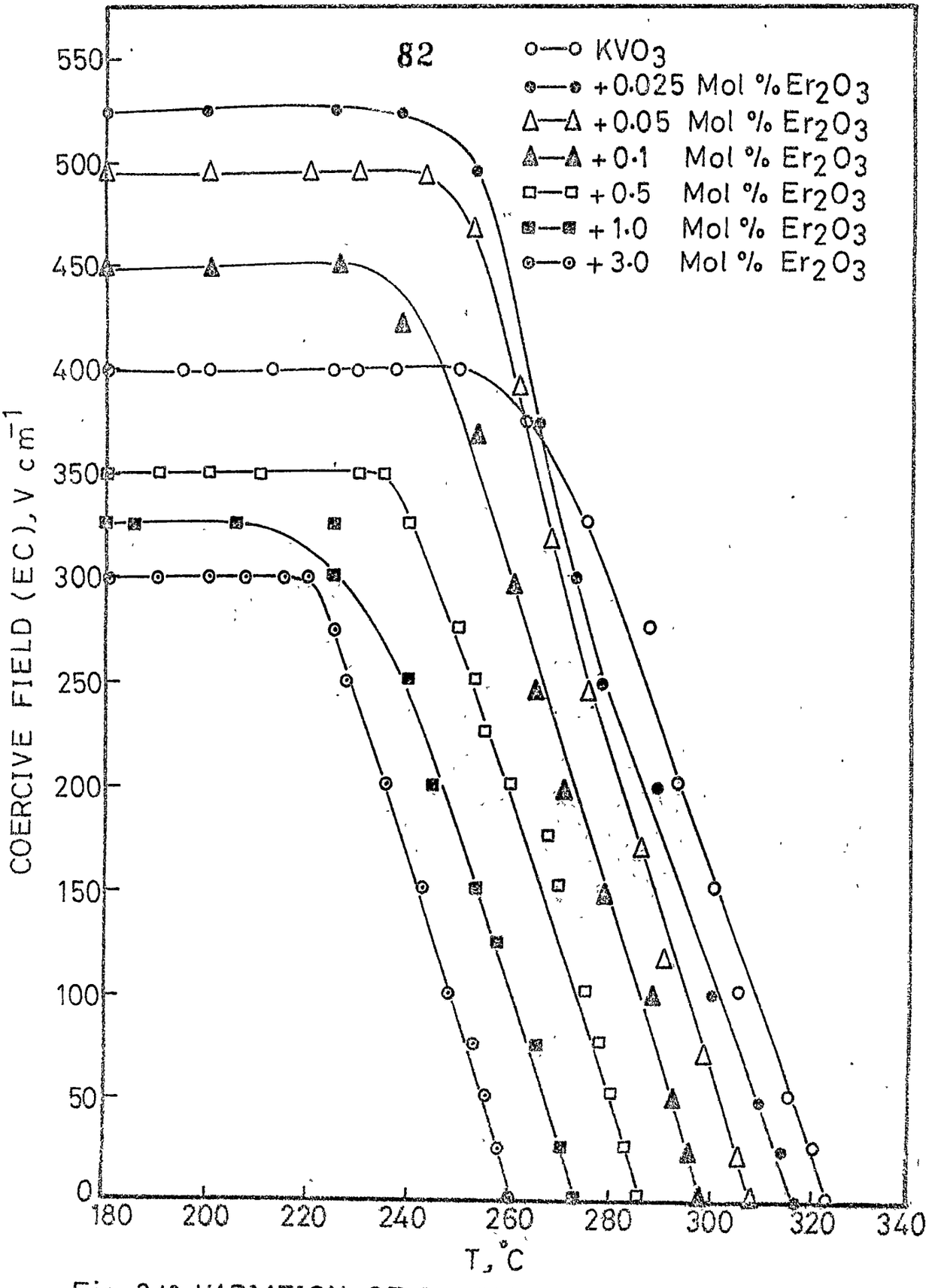


Fig. 3.10 VARIATION OF COERCIVE FIELD WITH TEMPERATURE FOR UNDOPED AND DOPED KVO_3 WITH Er_2O_3.

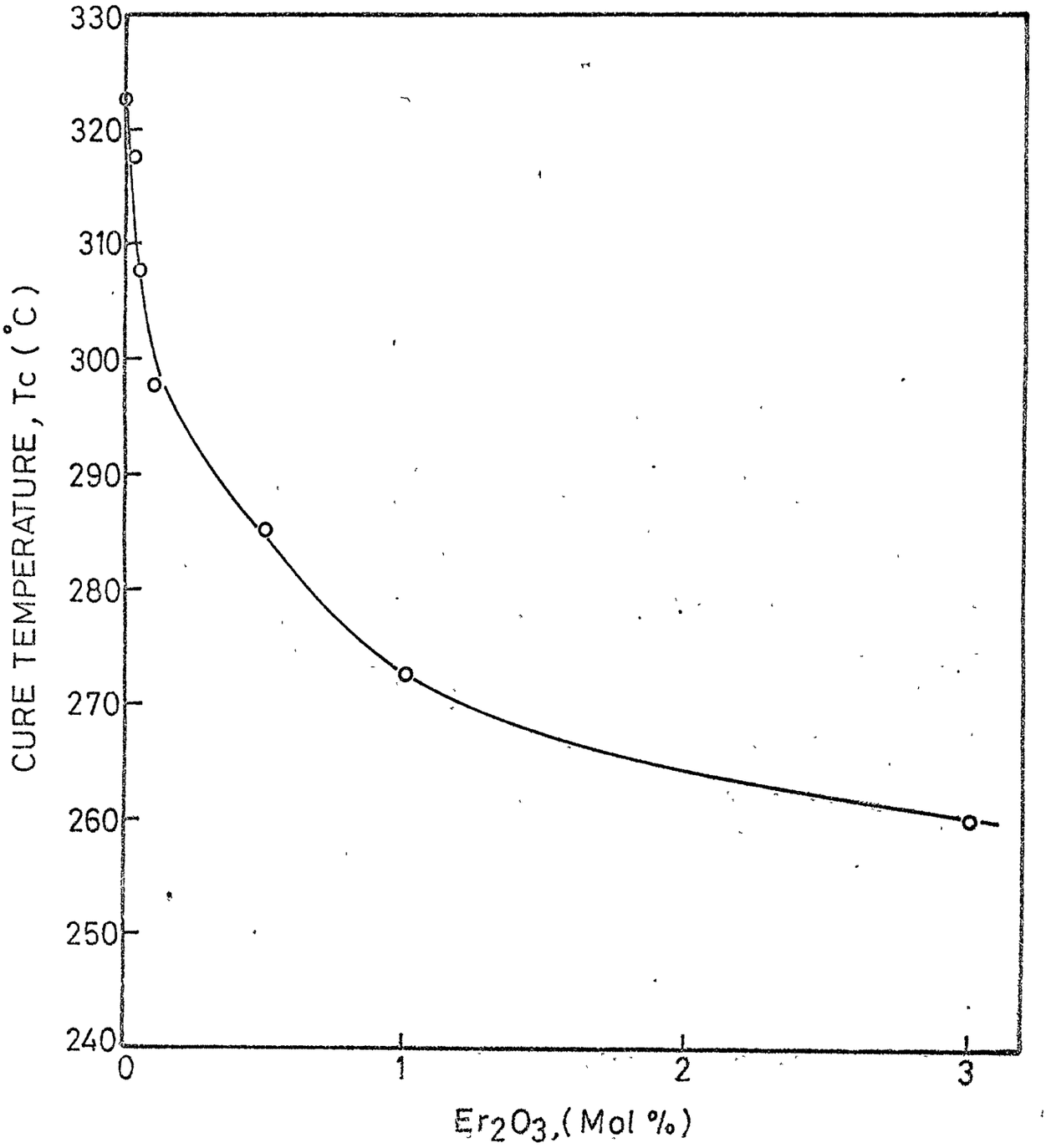


Fig. 3-11 PLOT OF CURIE TEMPERATURE VS Er_2O_3,(Mol %) IN KVO_3.

TABLE NO.3.1

FERROELECTRIC CURIE TEMPERATURE AND COERCIVE FIELDS OF DIFFERENT SAMPLES.

Er_2O_3 Content (mol%)	Curie Temperature T_c, ^0c	Peak value of E_c volt/cm
0.000	322.5	400
0.025	317.5	525
0.050	307.5	500
0.100	297.5	450
0.500	285.0	350
1.000	272.5	325
3.000	260.0	300

Table 3.1 indicates that peak values of coercive field for different samples are different and Curie temperatures are also different. It is also observed that as Er_2O_3 in KVO_3 increases, Curie temperature decreases from 322.5 ^0c to 260.0^0c.

3.7 RESULTS AND DISCUSSION

The observed hysteresis loop is rectangular in shape and area of the loop gives the energy loss during a complete cycel. Figs. 3.3 to 3.9 show that the shape of hyseresis loop depends strongly on temperature and hence coercive field is also temperature dependent. As temperature decreases loop width becomes zero indicating the Curie temperature of the sample. The Curie temperature of undoped KVO_3 is 322.5^0c which is quite in agreement

with that reported by Sawada et al (1951), Chavan et al (1986), Patil et al(1989) and Kashid et al (1989). The Curie temperature of the sample decreases as Er_2O_3 content in KVO_3 increasaes.It is observed that Curie temperature decreases from 322.5^0c to 260^0c as Er_2O_3 increases from 0.025 mol% to 3 mol% as shown in the Fig.3.11. It is also found that the peak value of coercive field of sample containing 0.025 mol% Er_2O_3 is maximum and it is due to high densification of the sample. The sample containing 0.025 mol% Er_2O_3 indicates the substitution of K^{+1} by Er^{+3}, because of their comparable ionic radii and hence the sample shows high density. The further doping of Er_2O_3 represents decrease in peak value of coercive field,indicating the defects created in the ceramic. Similar results regarding hysteresis behaviour were reported by Sawada et al (1951) for $NaVO_3$ crystal and by Ismilazade et al (1981) for rare earth vanadates.

From the hysteresis behaviour of our samples, the following conclusions could be drawn.

1. The hysteresis loss is very small at room temperature and is pronounced at a temperature quite above the room temperature.
2. The hysteresis loop vanishes at a particular temperature indicating ferroelectric Curie temperature of the sample.
3. The Curie temperature of KVO_3 depends on Er_2O_3 concentration.
4. Curie temperature of KVO_3 deceases from 322.5^0c to 260^0c as dopant concentration of Er_2O_3 increase in KVO_3.
5. The peak values of coercive fields are different for various samples.
6. The peak value of coercive field is maximum for sample having 0.025 mol% Er_2O_3 in KVO_3 and it decreases as Er_2O_3 content in KVO_3 increases.

REFERENCES

Abe R.	(1960), J.Phys. Soc.Jpn. 15,795.
Bertaut E.F. and Lissalde F.	(1967),Solid State Comm. 5, 173.
Chavan S.H. and Suryavanshi S.G.	(1985), Ind.J.Phys. 59A, 555.
Griffiths G.R. and Ruzell R.	(1972), J. Am. Ceram. Soc. 55, 110.
Gristsberg V,Gaevskis A.P. and Grinvald G.	(1977). Izv. Vuz. Fiz.USSR, 1, 103.
Ismailzade I.H. and Others	(1981), Ferroelectrics. 31, 45.
Jamadar V.M. Patil T.A. and Chavan S.H.	(1986), Ind. J.Pure and Appl. Phys.24, 561.
Janovec V.	(1958), Czechoslov. J.Phys. 8,3.
Kashid A.P., Patil V.V. and Chavan S.H.	(1989), Bull.Mat. Sci.Vol-12.461.
Kuroda S.and Kubota K.	(1980)(1980), J.Phys. Chem. Sol. 42, 573.
Merz W.J.	(1954), Phys. Rev. 95, 890.
Patil T.A. Jamadar V.M. and Chavan S.H.	(1988), Ind. J.Pure and Appl.Phys.Vol-26 456-458.
Patil N.B., Patil R.T. and Chavan S.H.	(1989), Bull.Mat.Sci. 129-132.
Sawada s. and Nomura s.	(1951), J.Phys. Soc. Jpn. 6, 192.
Sawyer C.B. and Tower C.H. and Yohoko A.	(1930),Phys.Rev. 35, 269.
Yalasek J.	(1920), Phys.Rev. B-12, 3755.
Weider H.H.	(1955), J.Appl.Phys. 26, 1479.