CHAPTER VII:

THICKNESS DEPENDENCE: PMMA THIN FILMS.
7.1 **INTRODUCTION**

Thickness dependence of the electric strength of a material, helps in predicting the mechanism of breakdown in it.

Low energy as well as high energy criterion does not predict directly any dependence of electric strength upon thickness. Since 'impact ionisation' is responsible for multiplication of electrons in the conduction band according to these criteria, thickness dependence of electric strength is to be expected.

If breakdown results from impact ionisation, the electric strength must increase rapidly, as the specimen thickness approaches the mean free path of conduction electrons. This follows from the fact that not all collisions result in ionisation and in order to produce secondaries, a conduction electron must make several collisions before it is removed at the anode. As thickness is reduced (field strength remaining constant), the number collisions decreases but ionisation can be maintained by increasing the field, because this increases the probability of ionising collision.

According to Prohlich\(^1\), in pure ionic crystals, scattering of electrons is due to temperature dependent
motion of ions and mean free path of electrons is $\approx 10^{-6}$ cm. Evidently, thickness dependence should be appreciable for thin samples.

Austen and Whitehead\textsuperscript{2} reported an increase of dielectric strength of mica flakes by decreasing the thickness.

Plesner\textsuperscript{3} observed an appreciable rise in the electric strength of vacuum evaporated films of KBr and NaF, when thickness was reduced in the range $10^{-4}$ cm to $10^{-5}$ cm. However, for silica (vacuum evaporated films in the thickness range $10^{-6}$ cm to $5 \times 10^{-5}$ cm) and polystyrene (films obtained by evaporation of a dilute solution of polystyrene, in the thickness range ($37 \times 10^{-5}$ cm to $3 \times 10^{-5}$ cm) no thickness dependence was found. It is not contrary to Frohlich's conclusion. As a matter of fact, silica and polystyrene are amorphous, therefore, electronic mean free paths will be extremely small compared to the range of thickness in which studies were made.

Ryu and Kawamura\textsuperscript{4} found electric strength of monocrystalline KCl independent of thickness in the range $1 \times 10^{-3}$ cm to $1.1 \times 10^{-2}$ cm. This result was doubtful on statistical grounds.

Kostygin\textsuperscript{5} found an increase from $2.5$ MV cm\textsuperscript{-1} to $7.5$ MV cm\textsuperscript{-1} in electric strength of single crystal of NaCl as thickness decreased from $16 \times 10^{-4}$ cm to $3 \times 10^{-4}$ cm.
Vermeer for pyrex glass (an amorphous material) found electric strength independent of thickness.

Seitz forty generation theory also yields thickness dependence as given by,

\[ E_b = \frac{1}{\log(d/d_0)} \]  \( (7.1) \)

Where \( d_0 \) is the electronic mean free path and \( d \) is the thickness. Relation (7.1) has been obtained by assuming that (a) an electron having energy just equal to ionising energy can initiate avalanche, and (b) relaxation time of electron is independent of its energy \( E \).

Forlani and Minnaja objecting to Seitz assumptions postulated.

1. For the initiation of an avalanche, impinging electron as well as freed electron must possess energy for further ionisation.

2. The probability per unit time of an electron-phonon interaction changes as long as electron receives energy from the field.

On the basis of these postulates, the logarithmic relationship,

\[ E_b \propto \frac{1}{\log(d/d_0)^{1/3}} \]  \( (7.2) \)

was deduced.
Korn$^8$ prepared amorphous $\text{Al}_2\text{O}_3$ films by thermal decomposition of alluminiun ethylate $\text{(C}_2\text{H}_5\text{O})_3\text{Al}$ or acetylacetonate $\text{(C}_5\text{H}_7\text{O}_2)\text{Al}$ on heated metal substrate and found

$$E_b \propto d^{-\alpha} (\alpha = .45 \text{ to } .85)$$

(7.3)
in the thickness range 800 $\mathring{A}$ to 2000 $\mathring{A}$. On further increasing the thickness, the dependence becomes logarithmic viz.

$$E_b \propto \log d^{-\beta}$$

(7.4)
in the thickness range 2000$\mathring{A}$ to 5000$\mathring{A}$. Beyond this, thickness dependence becomes very weak and practically disappears at thickness $\geq$ 1$\mu$. In all cases the value of exponent $\beta$ does not exceed unity.

Similar results were obtained$^9$ for chemically grown $\text{SiO}_2$ amorphous films in the thickness range 400$\mathring{A}$ to 5000$\mathring{A}$.

It is important to mention here that, the transition from power law to logarithmic law, indicates that more than one mechanism of scattering of electrons contribute simultaneously to breakdown. Such a transition occurs for thickness 2000$\mathring{A}$ of chemically grown $\text{SiO}_2$ films.

Weaver and Mclcodd$^9$ experimentally observed a logarithmic behaviour of thickness dependence of electric strength in case of vacuum deposited $\text{NaCl}$, $\text{NaF}$, and Cryolite thin films. It was pointed out that this behaviour is qualitatively of the
type associated with impact ionisation. Time to breakdown was found to be $10^{-6}$ sec. which further supports their conviction.

Vorobev et al.\textsuperscript{10} measured the breakdown voltage of LiF, NaCl and KCl single crystals as function of thickness (0.5 μ to 5 μ). Each halide exhibited a minimum between 2μ and 2.5 μ in the thickness dependence of breakdown voltage. This minimum was regarded as the proof that electric breakdown was caused by impact ionisation.

Oakes\textsuperscript{11} concluded that for polythene |thickness ranges (i) 0.0005" - 0.002" (ii) 0.002" to 0.008", electric strength is independent of thickness for plane recessed samples. Cooper et al.\textsuperscript{12} say that Oakes' result appears to be false. They examined breakdown v/s thickness graph of Oakes and found that there was a difference in the slopes of the lines which best fit the points at the two extremeties of the range of thickness. They further confirmed thickness effect by measuring electric strength of alkathene-7 in thickness range 0.001" to 0.018" using plane recessed specimens prepared by moulding as was done by Oakes. Their measurements satisfied the equation,

$$E_b = 12.8 - 3 \log d \text{ MV cm}^{-1}$$ \hspace{1cm} (7.5)

where $d$ was measured in units of $10^{-3}$ inches. They further pointed out that the intrinsic dielectric strength is supposed
to be independent of thickness, provided this exceeds a few electronic mean free paths, but in fact the possibility of occurrence of a small effect is not excluded by available evidences.

Kolesov-Diefman\textsuperscript{13} measured electric strength of polypropylene, polystyrol and polyethylene films of thickness between 10 μm to 80 μm, and found that as thickness decreases below 20 μm, electric strength of all polymers increases rapidly.

Blinov\textsuperscript{14} gave the relationship,

$$E_b \propto d^{-\frac{1}{2}}$$  \hspace{1cm} (7.6)

for SiO\textsubscript{2} films in the thickness range 0.05 μm to 0.5 μm.

Lomer\textsuperscript{15} obtained for anodized Al,

$$E_b \propto d^{-3}$$  \hspace{1cm} (7.7)

where $B = 0.3$. Relation (7.7) was confirmed by Merril and West\textsuperscript{16}. Experiments of Kawamura and Ryu\textsuperscript{17} on mica layer gave $B = 0.65$.

Hamas\textsuperscript{18} reports that for SiO\textsubscript{2} thin films relation (7.6) holds.

However, the earlier theories exclude electron injection from cathode.
Forlani-Mimaaja\(^7\) considered Fowler-Nordheim emission from cathode and assumed that thickness of the film is much lower than the recombination length and deduced

\[
E_b = \left[\frac{4(2m)^{3/2}}{\phi_{\text{eff}}^{3/2}} \frac{3}{\hbar} \frac{E}{3 \hbar q^2} \right]^{1/2} \quad \text{...(7.8)}
\]

where \(\phi_{\text{eff}}\) is the effective height of the potential barrier at the cathode dielectric interface and \(E\) is the difference between the mean energy of an ionising electron and the mean energy of the electrons emerging from an ionisation event.

If the effective height of the potential barrier is very low, then

\[
E_b \approx \frac{1}{d^{3/4}} \quad \text{...(7.9)}
\]

For Schottky emission from cathode -

\[
E_b = \frac{\phi_{\text{eff}}}{kT} \frac{E}{q} \cdot \frac{1}{d} \quad \text{...(7.10)}
\]

which yields that breakdown voltage is independent of dielectric thickness or breakdown strength is inversely proportional to thickness.

Pock\(^{19}\), Klein and Gafni\(^{20}\) attribute breakdown to a thermal effect i.e. the current density even without an avalanche multiplication, generates more heat into the dielectric than the dielectric is able to dissipate to the surroundings. On the basis of thermal breakdown concepts O'Dwyer showed that the breakdown field is proportional to
the inverse of the square root of thickness, while Klein and Gafni deduced that such a field varies as the reciprocal of the logarithm of the film thickness.

Thickness dependence arising from thermal instability has also been discussed in Chapter I.

7.2 EXPERIMENTAL

Thickness dependence of electric strength of PMMA thin films was studied at room temperature using d.c. voltage.

Experimental conditions in brief were:

1. Type II (circular capacitors) samples were employed.
2. Type A as well as type B breakdown studies were carried out.
3. In case of type A breakdown samples were studied in air and in case of type B breakdown, sample were immersed in transformer oil.
4. Thickness of samples were calculated from capacitance measurements. Dielectric constant of PMMA was taken equal to 4 at 1 KHz and room temperature.

For type A breakdown, samples of thickness 1.36\mu, 1.7\mu and 2.8\mu and for type B breakdown, samples of thickness 1.7\mu, 2.8\mu, 3.3\mu and 4.9\mu were taken.

5. Circuit for type A breakdown is given in Figure 3.1.
while for type B breakdown, electronic monitor was removed from the circuit.

15 samples of each thickness were tested in type B breakdown.

Observations have been represented graphically as follows:

In case of type A breakdown, breakdown voltages for samples of each thickness is found from test number v/s breakdown voltage curves (Fig. 7.3.1). Then the graph between thickness and breakdown voltage is plotted (Fig. 7.3.2).

Each point on thickness v/s breakdown voltage graph for type B breakdown represents an average of 15 tests and each bar represents the standard deviation (Fig. 7.3.3).

7.3 RESULTS AND DISCUSSION

Graphs 7.3.2 and 7.3.3, show that electric strength decreases with increase in thickness. The shape of the curves show an inverse power law of thickness dependence i.e.

\[ F_b \propto d^{-\alpha} \]

Values of \( \alpha \) calculated for different parts of the curve 7.3.2 vary between .43 and .62.
FIG. 7.3.1- ELECTRIC STRENGTH AS A FUNCTION OF TEST NUMBER FOR PMMA THIN FILMS OF DIFFERENT THICKNESS.
FIG. 7.3.2 - THICKNESS DEPENDENCE OF ELECTRIC STRENGTH OF PMMA THIN FILMS AT ROOM TEMPERATURE.
FIG: 7.3.3 - ELECTRIC STRENGTH AS A FUNCTION OF THICKNESS FOR PMMA THIN FILMS.
It appears that d.c. breakdown in PMMA thin films is electronic, controlled by Fowler-Nordheim emission from cathode as suggested by Forlenzi and Minnaja\(^7\).

The polarity effect in PMMA thin films also indicates electron injection from cathode. Further, present investigation on temperature dependence of electric strength of these films, indicate that electron injection process should be temperature independent i.e. Fowler-Nordheim emission.

Inverse power relation is also obtained in case of thermal breakdown. The temperature insensitive behaviour of breakdown field of PMMA thin films, rules out the possibility of thermal breakdown mechanism.
REFERENCES


