

## DIELECTRIC STRENGTH OF SOME BLENDED POLYMERS PREPARED IN THE FORM OF THICK FILM

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Careful measurements of dielectric strength (DS) of blended polymeric material have been accomplished. For the measurements of DS of the films (1.0 mm thickness) prepared from these blends, McKeown's electrode system with stainless steel electrodes of 25mm diameter was used. Three types of blends were prepared. They were PE-PMA, PS-PI and PS-PE-PI. The film prepared from the blend 100% PMA had IBDS value as 200 kVmm<sup>-1</sup> whereas the additions of PE enhanced IBDS value to 212 and 205 kVmm<sup>-1</sup> respectively at (PE/PMA) 20/80 and 5/95 ratios. 100% PI produced IBDS as 250 kVmm<sup>-1</sup>. The addition of PS decreased the value of IBDS. On the other hand the blend of these polymers (PS-PE-PI) at PS/PE/PI ratio of 70/10/20 gave highest IBDS (263 kVmm<sup>-1</sup>). The break down is initiated by free electrons accelerated by electric field in the largest of the holes present in the amorphous phase of these polymers. These holes were formed during the production of films. Nature, ageing, photodegradation of polymeric material, shape of specimen and good contact of specimen with the surface of electrode influence on the IBDS values and help in excluding the partial discharges.

**Key words:** Low density polyethylene LDPE, Impulse breakdown strength, Blending of polymeric composites, Copolymers, Ethylene methyl acrylate.

### Introduction

The dielectric strength of electrically insulating material is a property of interest for many applications where the electric field is involved. In many cases the dielectric strength of a polymeric material is the determining factor in the design of the apparatus in which it is to be used (ASTM-9 1983). Most of the polymers have amorphous structure either exclusively or at least to a certain degree (Tobolsky 1960) of crystallization. In contrast to the crystalline structure, the volume of the amorphous phase is not completely occupied by the material particles, atoms or molecules. The unoccupied part of the volume or the free volume is constituted by a number of small empty spaces, holes between the molecules (Artbauer 1996). In the amorphous phase a fairly rapid fall in electric strength above the second order transition takes place which is due to a close connection between the mechanical properties and the electric strength (Bueche 1962). Later Garton and Stark (1955) reported their observations that the lowering of the electric strength is due to photo degradation of the polymeric material. Furthermore, Hikita *et al* (1983) suggested that with the increase of temperature, the decrease in dielectric strength may be due to the drop in the mechanical strength and morphology. Nelson (1983) noticed that the results of his

careful measurements of electrical strength of polystyrene, polybutadiene and copolymers are in agreement with the free volume concept. Originally the breakdown starts when the energy of the electrons, accelerated over the largest free path, attains the  $E_i$ , the ionization energy (Artbauer 1996). Besides, according to the results of theory of conduction by Artbauer, breakdown occurs when the energy of the free electrons, accelerated in high field strength and in the holes with the largest free path, attains the energy value needed to jump over the potential barrier in the compact structure. At high field strength, the charge carriers are electrons. They are generated by the ionization of thermally activated neutral centres in the bulk of the dielectric medium (Hikita *et al* 1980). In such field of strength, both the number of electrons and their velocity increase superlinearly. The concentration or the number of electrons per unit volume ' $n_e$ ' depends on the ionization energy  $E_i$ , the temperature ( $T$ ) and the electric field ' $F$ ' which lower the potential barrier. In strong field, the dependence on pool-Frankel Model (Frankel 1938) may be expressed by the equation:

$$n_e \approx N_e \exp[-(E_i - B_{pf} F)^{1/2} / (2KT)]$$

where  $N_e$  is the concentration of ionizable centres,  $K$  is the Boltzman constant,  $E_i$  is the ionization energy and

$$B_{pf} \approx [e^3(\pi \epsilon_1 \epsilon_0)]^{1/2}$$

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where  $Bp_f$  is called the pool Frankel parameter and  $\epsilon$ ,  $\epsilon_0$  are the static permittivity. The conditions whereby polymers are blended, the thermodynamic criteria of miscibility and the properties of blends have been reviewed by Paul and Newman (1978) which help in the determination of dielectric strength. This paper deals with the dielectric strength of some blended polymers and copolymers prepared in the form of thick layers.

## Experimental

**Materials:** Laboratory grade polyethylene: (M.W. 20000), Styrene (E.Merck), methyl acrylate (E.Merck) and commercial grade polyisoprene were used without further purification. All solvents were distilled before use.

**Procedure of preparation of blends:** Blends based on polystyrene (PS) polyethylene (PE) polymethyl acrylate (PM) and polyisoprene (PI) were prepared and films of these blends were cast for the measurement of dielectric strength.

All blends in different monomeric ratios shown in the respective tables were prepared in flanged flask equipped with a condenser, stirrer and thermometer.

**Blend of PE-PMA:** A weighed quantity of PE was dissolved in xylene in the reaction flask and known quantity of methyl acrylate monomer and benzoyl peroxide (BP) (0.5g) was added to it. The whole contents of the reaction flask were stirred and heated at 120-130°C till a viscous paste was obtained.

**Blend of PS-PI:** The known quantity of PI was digested in the xylene and the reaction flask of weighed quantity of styrene and BP (.5 g) was added to it. The contents were stirred and heated at 130°C to get a viscous paste.

**Blend of PS-PI-PE:** Weighed quantity of PI and PE were digested in xylene. The known quantity of styrene and BP (0.5g) was added to it. The reactants were stirred and heated at 130°C till a highly viscous paste was obtained.

**Preparation of polymeric films:** The measurement of the dielectric strength of the films of the pastes were cast in a die (25x25x1mm<sup>3</sup>). The paste was spread on the die and the solvent was evaporated. It was then hydraulically pressed at a pressure 100 Kg in<sup>-2</sup> at room temperature.

**Measurement of dielectric strength:** For the measurement of dielectric strength of polymeric films, the Mckeowns electrodes system (Fig 1) with stainless steel electrodes of 25mm diameters was used. A negative pulse of 40 Sec (micro sec) impulse normalized wave was applied on the opposite faces of each sample for three times with a difference of 5kV in stepping up order, starting from 70% of the expected breakdown voltage of the individual specimen. After each

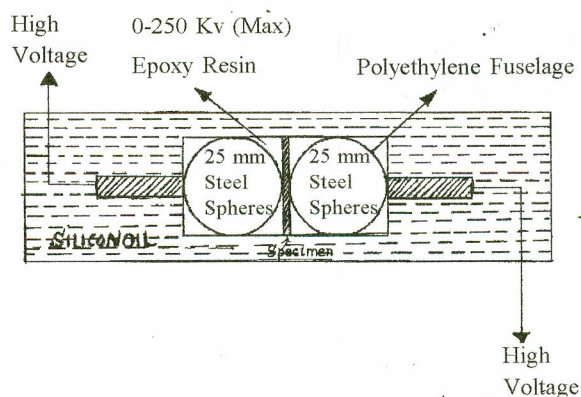


Fig 1. Modified Mckeowns electrode system for the measurement of dielectric strength.

measuring, the surface of the electrode was polished and kept free from irregularities. Seals were applied around the electrodes in contact with the specimen. The test voltage was applied on the opposite faces of test electrodes. The impulse breakdown strength for each of the prepared sample was recorded by a voltmeter using a Fluke, 0-350kV stabilized power supply and thus was calculated per mm thickness. The mean value of the three concurrent readings was obtained for obtaining the Y-values (63.3%) of accumulated breakdown probability with the X-values of blending ratio. Since such polymer samples have high breakdown strength greater than 250 kV, all the tests were performed in any mineral oil chamber e.g. silicon oil. All the metallic parts were grounded for the safety purpose.

## Results and Discussion

Table 1-3 summarize the data regarding the dielectric strength of films prepared from the blends of PE-PMA, PS-PI and PS-PE-PI. The blends of PE and PMA in different monomeric ratios were prepared and from these blends the films of 1mm thickness were cast as shown in Table-1. The film prepared from the blend containing 100% PMA gave dielectric strength value as 200 kV mm<sup>-1</sup> whereas the addition of PE in the blend gave the highest value of dielectric strength as 212 and 205 kV mm<sup>-1</sup> respectively at (PE/PMA) 20/80 and 5/95 ratios. The lowest dielectric strength was received (160 kV mm<sup>-1</sup>) at (PE/PMA) 40/60 ratio. This change may be due to the free electrons accelerated by the electric field in the largest holes present in the amorphous phase of PE (C<sub>4</sub>H<sub>4</sub>) and PMA (C<sub>4</sub>H<sub>6</sub>O<sub>2</sub>). Polyethylene is a thermoplastic polymer of ethene (CH<sub>2</sub> = CH<sub>2</sub>) an electrone donating monomer and is known as the elastomer of superior electrical properties at very high radio frequencies whereas PMA is an electron accepting monomer. The interaction of these two monomers creates holes that increase or decrease the break down value in the form of dielectric strength. In reality the measured values of break-

**Table 1**  
Dielectric strength of films prepared from the blend of polyethylene-polymethyl acrylate (PE-PMA)

S.No.	Blending ratio %		IBDS kV mm <sup>-1</sup>
	PE	PMA	
1	0	100	200
2	5	95	205
3	10	90	190
4	20	80	212
5	30	70	175
6	40	60	160

**Table 2**  
Dielectric strength of films prepared from the blend of poly (styrene) - poly (isoprene) rubber

S.No.	Blending ratio %		IBDS kV mm <sup>-1</sup>
	PS	PI	
1	0	100	250
2	10	90	245
3	15	85	235
4	20	75	233
5	25	70	225
6	30	75	229
7	40	60	222

**Table 3**  
Dielectric strength of films prepared from the blend of polystyrene-polyethylene with polyisoprene rubber (PS-PE-PI)

S.No.	Blending ratio %			IBDS kV mm <sup>-1</sup>
	PS	PE	PI (Rubber)	
1	70	10	20	263
2	60	20	20	258
3	50	30	20	252
4	40	40	20	249
5	30	50	20	245
6	20	60	20	241
7	10	70	20	233

down field are not the inherent electric strength because the weak point by which they are determined are not a part of the polymer structure. Weak pathways are cavities which are

developed on the boundaries of crystalline regions during production and processing (Artbauer 1996).

Table-2 includes the data regarding dielectric strength of the films prepared from the blends PS-PI. Dielectric strength at 100% concentration of PI was determined as 250 kV mm<sup>-1</sup>. The addition of PS in the blend decreases progressively the dielectric strength values. The dielectric strength was determined as 245 kV mm<sup>-1</sup> at (PS/PI) 10/90 ratio whereas it was 222 kV mm<sup>-1</sup> at (PS/PI) 40/60 ratio. Isoprene monomer contains conjugated double bonds (CH<sub>2</sub>=C (CH<sub>2</sub>) CH=CH<sub>2</sub>) whereas styrene monomer bears resonance structure. On polymerization the molecular interaction of these monomers results in a change in the molecules. It must be remembered that the holes in amorphous structure are an inherent part of the polymer. The dimensions are of molecular order. Furthermore, empty spaces or holes are not the voids known as weak points that lead to break down in technical insulation. The latter are defects or imperfections which are also certainly due to manufacturing material or damages. The blends prepared from freshly prepared polymeric material may give better results. Photodegradation of polymeric material can not be neglected as one of the factor in the change of dielectric strength.

In the Table-3, the data regarding the dielectric strength of films prepared from the blend of PS-PE-PI in different monomeric ratios is presented. In these blends, a fixed quantity of PI (20%) was blended with different ratios of PS & PE. The highest value of dielectric strength was obtained as 263 kV mm<sup>-1</sup> at (PS/PE/PI) 70/10/20 ratio. It decreases progressively with the increasing quantity of PE (see Table-3). The lowest value of dielectric strength was obtained as 233 kV mm<sup>-1</sup> at (PS/PE/PI) 10/70/20 ratio. The drop in dielectric strength may be due to PE as an electron donor (its monomer). These results also indicate that the change in dielectric strength is not only due to the inherent holes of the material but the nature and ageing of polymeric material also play an important role in the change of break down field.

In brief, when an electric current is applied through the specimen, the breakdown is initiated by free electrons accelerated by electric field in the largest of the holes or cavities present in the amorphous phase of these polymers. The holes are formed during the production of films prepared from the blends of polymeric material. Actually when films are cast, during cooling, the imperfections on boundaries are evolved which act as holes and affect the dielectric strength values. Though the holes are inherent in the amorphous phase of polymeric material but nature, ageing, photodegradation of polymeric material influence the dielectric strength values. Besides these parameters, the shape of the specimen, material

of the electrode, good contact of specimen with the surface of the electrode to exclude partial discharges and the way in which the voltage is changed upto breakdown play significant role in getting the highest and accurate dielectric strength values. Furthermore, for getting accurate results the surface of the electrode should not be irregular and should be polished.

### References

- ASTM-9 1983 Section-9 *Designation-Rubber D-149-81*, 25-37.
- Artbauer J 1996 Electric strength of polymers. *J Phys D App Phys* **29**, 446-456.
- Bueche F 1962 *Physical Properties of Polymers* Inter Science publications, Chapter No.2 (86-99).
- Garton C G, Stark K H 1995 Electric strength of irradiated polyethene. *Nature* **176** 1225-1226.
- Frankel J 1938 On prebreakdown phenomena in insulators and electronic semi conductors. *Physical Review* **54** 647-648.
- Hikita M, Nagao M, Sawa G, Leda M 1983 Electrical breakdown solid structure in polyvinylidene fluoride. *J Phys D Opp Phys* **16** 157-161.
- Hikita M, Nagao M, Sawa G, Leda M 1980 Dielectric behaviour and electrical conduction of polyvinylidene fluoride at high temperature region. *J Phys D App Phys* **13** 661-666.
- Nelson J K 1983 *Breakdown of solids* Philadelphia PA, (John Wiley & Sons, NY) Chapter No.1 and Chapter No.IX, IInd Edn.
- Paul DR, Newman S 1978 *Polymer Blends* (I), Academic Press NY, Chapter III (104-113).
- Tobolsky A V 1960 *Properties and Structure of Polymers*, John Wiley and Song, NY, Chapter No.2 (43-57), IInd Edn.