The effect of polymer type on electric breakdown strength on a nanosecond time scale

Zhao Liang(赵 亮)[a)b)†], Su Jian-Cang(苏建仓)[b)],
Pan Ya-Feng(潘亚峰)[b)], and Zhang Xi-Bo(张喜波)[b)]

[a)] *Key laboratory of Physical Electronics and Devices of Ministry of Education, Xi'an Jiaotong University, Xi'an* 710049, *China*
[b)] *Science and Technology on High Power Microwave Laboratory, Northwest Institute of Nuclear Technology, Xi'an* 710024, *China*

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Based on the concepts of fast polarization, effective electric field and electron impact ionization criterion, the effect of polymer type on electric breakdown strength ($E_{\rm BD}$) on a nanosecond time scale is investigated, and a formula that qualitatively characterizes the relation between the electric breakdown strength and the polymer type is derived. According to this formula, it is found that the electric breakdown strength decreases with an increase in the effective relative dielectric constants of the polymers. By calculating the effective relative dielectric constants for different types of polymers, the theoretical relation for the electric breakdown strengths of common polymers is predicted. To verify the prediction, the polymers of PE (polyethylene), PTFE (polytetrafluoroethelene), PMMA (organic glass) and Nylon are tested with a nanosecond-pulse generator. The experimental result shows $E_{\rm BD}$ (PTFE) > $E_{\rm BD}$ (PMMA) > $E_{\rm BD}$ (Nylon) > $E_{\rm BD}$ (PE). This result is consistent with the theoretical prediction.

Keywords: electric breakdown strength, polymer type, nanosecond time scale, fast polarization phenomenon

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1. Introduction

Polymers are widely used in high-voltage devices[1−3] and pulsed power systems.[4−7] It is of great importance to research the electrical properties of polymers on a nanosecond time scale. In the 1960s, Martin made some investigations into the volume effect and the pulse number effect on the electric breakdown strength ($E_{\rm BD}$) of polymers, and concluded the famous Martin formulae.[8,9] In the 1970s, Mesyats reported on the pulse width effect and the pulse polarity effect on $E_{\rm BD}$.[10] Other research on the rise time effect,[11] the repetition rate effect[12] and the dielectric thickness effect on $E_{\rm BD}$[13] has also been reported. This research is helpful for the application of polymers on a nanosecond time scale. However, to extend their application and to provide more criteria for designers and scientists, more of the electrical properties of polymers should be studied.

In this paper, the polymer type effect on $E_{\rm BD}$ is firstly investigated and a formula is derived. The theoretical bases for this investigation are introduced in Section 2, and the method for deriving the values of the effective dielectric constant ($\varepsilon_{\rm re}$) and the prediction for the $E_{\rm BD}$ relation of polymers are presented in Section 3. To verify the prediction, experiments on the $E_{\rm BD}$ of four types of polymers are conducted and the experimental results are presented in Section 4. In Section 5, the discussion and conclusion on the derived formula are presented.

2. Theoretical analysis

This section is devoted to the theoretical bases for the polymer type effect on $E_{\rm BD}$ on a nanosecond time scale, which comprises three sub-sections: the fast polarization phenomenon, the concept of the effective electric field ($E_{\rm e}$) and the electron impact ionization criterion.

2.1. The fast polarization phenomenon

There are five basic types of polarization in solids.[14,15] Table 1 lists the mechanisms and the establishment time ($t_{\rm es}$) for each type of polarization. From this table, it can be found that on a nanosecond

†Corresponding author. E-mail: zhaoliang0526@163.com
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Table 1. The five basic types of polarization in solids.

Types	Polarization mechanisms	t_{es}/s
Electronic polarization /optical polarization	Relative elastic displacement of electron clouds to nucleus	10^{-15}–10^{-14}
Ionic or atomic polarization /vibrational polarization	Relative elastic displacement of negative ions to positive ions	10^{-14}–10^{-12}
Orientational polarization	Uniform alignment of polar molecules	10^{-6}–10^{-2}
Hopping polarization /relaxation polarization	Hopping of localized charges (ions and vacancies, electrons and holes)	10^{-6}–10^{-2}
Space charge polarization /interfacial polarization	Space charge in nonuniform dielectrics	10^{-2}–10^{4}

time scale, only the electronic polarization and the ionic/atomic polarization can be established, but the other three cannot. Moreover, the electronic polarization and the ionic/atomic polarization are both based on the relative elastic displacement of the negative particles to the positive ones, which are also different from the other three types. Based on these two similarities, the electronic polarization and the ionic/atomic polarization are analysed together with a unified name: fast polarization. Furthermore, the relative dielectric constant caused only by fast polarization is called the effective relative dielectric constant, ε_{re}, to distinguish it from the relative dielectric constant ε_r caused by all types of polarization.

When the concept of fast polarization is employed for polymers, it will only denote the electronic polarization, since there are no metallic elements in polymers. Based on the description of the five types of polarization in Ref. [14], Figure 1 shows the basic polarization types and the fast polarization phenomenon in polymers. By taking into account the fact

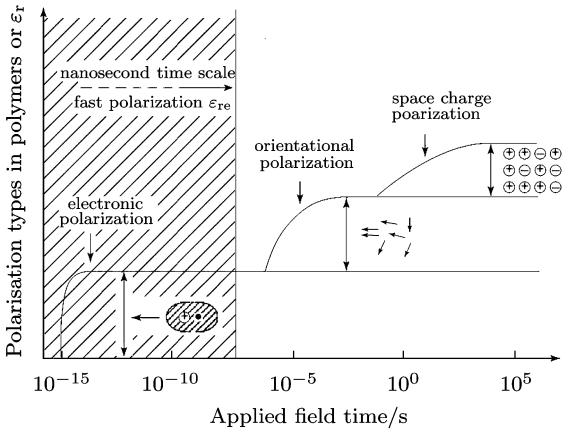


Fig. 1. The polarization types in polymers.

that ε_{re} of a polymer is only determined by its chemical structure, the polymer type can be represented by the value of ε_{re}, and the relation of E_{BD} versus polymer types can be represented by that of E_{BD} versus ε_{re}.

2.2. The concept of E_e

Besides ε_{re}, another concept connected with the relation of E_{BD} versus polymer types is the effective electric field or the local electric field, which is written as E_e. To formulate this concept, the Lorentz model is employed, which is a small sphere of dielectric material removed from around the site of an atom or a molecule to form a cavity (see Ref. [14]).

According to Lorentz,[14] the relation of E_e and the applied field is as follows:

$$E_e = \frac{\varepsilon_r + 2}{3} E_{op}, \tag{1}$$

where E_{op} is the applied field. From Eq. (1), it can be seen that for any ε_r greater than 1, E_e being larger than E_{op} will always hold true. Moreover, if the time is on a nanosecond scale, ε_r should be replaced with ε_{re}.

2.3. Electron impact ionization criterion

The third concept about the relation concerned is the electron impact ionization criterion.[16,17] For breakdown on a nanosecond time scale, since the time scale is so short, the process can be considered as a quasi electron one, which means that the breakdown is caused by the electron impact ionization and multiplication in the dielectrics. Therefore, the electron impact ionization criterion can be employed to describe this process. The basis of this criterion is

$$\vec{e} + A \rightarrow 2e + A^{+}, \tag{2}$$

where $\vec{e}$ and e are the electrons with high and low energy, and A and A^+ are the particles (molecule or

atom) and its positive forms, respectively. The criterion for Eq. (2) is

$$\Delta E = eE_{\rm e}\lambda. \tag{3}$$

Equation (3) means that to ionize an atom or molecule, an electron accelerated in the effective field $E_{\rm e}$ must gain energy equal to or larger than the atom or the molecule's ionization energy ΔE during one free path λ. Inserting Eq. (1) into Eq. (3) gives

$$\Delta E = \frac{2+\varepsilon_{\rm r}}{3} e\lambda E_{\rm op}. \tag{4}$$

For the breakdown on a nanosecond time scale, the parameter $\varepsilon_{\rm r}$ should be replaced by $\varepsilon_{\rm re}$, and $E_{\rm op}$ should be replaced by $E_{\rm BD}$. Transformation of Eq. (4) gives

$$E_{\rm BD} = \frac{3\Delta E}{(2+\varepsilon_{\rm re})e\lambda}. \tag{5}$$

Since the parameter $\varepsilon_{\rm re}$ can represent the polymer type, Equation (5) is considered to be the theoretical expression of $E_{\rm BD}$, dependent on the polymer types on a nanosecond time scale.

It is difficult to predict the relation of $E_{\rm BD}$ to polymers only using Eq. (5), since the ΔE and λ for each type of polymer are not equal to those of others. To solve this problem, two 'bold' conjectures are introduced. One is that all the polymers have the same ionization energy, and the other is that the average inter-atomic distance for each type of polymer is basically the same as the others. Using these two conjectures, one can make a prediction that the larger the $\varepsilon_{\rm re}$ for a polymer, the smaller the $E_{\rm BD}$ for this type of polymer.

It should be noted that these two conjectures are basically acceptable. The reason for this lies in the fact that even if polymers are different from type to type, the main atoms comprising the polymers (such as C, H and O) are the same. This results in the average ionization energy for atoms or the average energy gaps for covalent bonds like C–H, C–O, O–H and C–C being the same. Moreover, since the number of atoms per unit volume in dielectrics is very large (10^{23} cm^{-3}), the average interatomic distance can also be considered to be the same.

It is also noted that this prediction of the relation between $E_{\rm BD}$ and polymer types is a qualitative one. This is because that for polymers, there are always defects or pores in them. The existence of defects will import impurity energy levels into the intrinsic energy levels, which will cause the fluctuation of ΔE. The existence of pores will introduce low-density domains into the polymers, which will cause the variation in λ. Both these effects will lead the prediction to be a qualitative one.

3. Derivation of $\varepsilon_{\rm re}$

To predict $E_{\rm BD}$ related to polymers using Eq. (5), the values of $\varepsilon_{\rm re}$ for different types of polymers should be derived.

3.1. Method to calculate $\varepsilon_{\rm re}$

The Clausius–Mossotti equation (K–M equation) is employed to calculate $\varepsilon_{\rm re}$ as[14,15]

$$\frac{\varepsilon_{\rm re}-1}{\varepsilon_{\rm re}+2} = \frac{N\alpha_{\rm s}}{3\varepsilon_0}, \tag{6}$$

where N is the number of atoms or molecules per unit volume, $\alpha_{\rm s}$ is the sum of the polarizability of electronic polarization, and ε_0 is the dielectric constant of the polymers in vacuum. Since N is an indirect physical parameter, it is replaced by the following equation:

$$n_{\rm m} = \frac{N}{N_{\rm A}} = \frac{\rho}{M}, \tag{7}$$

where $n_{\rm m}$ is the mole number, $N_{\rm A}$ is Avogadro's number, M is the atomic or molecular weight, and ρ is the density of the polymer. Inserting Eq. (7) into Eq. (6) gives

$$\frac{\varepsilon_{\rm re}-1}{\varepsilon_{\rm re}+2}\frac{M}{\rho} = \frac{N_{\rm A}\alpha_{\rm s}}{3\varepsilon_0}. \tag{8}$$

The left part of Eq. (8) is called the molar polarizability (R), which represents the macroscopic polarizability in light frequency or on a 10^{-15} s time scale. For the value of R, since the polymers are composed of covalent bonds such as C–H, C–O and C–C, it can be derived via summing up the molar polarizability of each small covalent bond as

$$R = \sum_i r_i, \tag{9}$$

where r_i represents the molar polarizability of the ith covalent bond in a polymer. The values of r_i for common covalent bonds are listed in Table 2.[18] Combining Eqs. (8) and (9) gives

$$\varepsilon_{\rm re} = \frac{M + 2\rho\sum\limits_i r_i}{M - \rho\sum\limits_i r_i}. \tag{10}$$

Equation (10) is the formula for calculating $\varepsilon_{\rm re}$ for different types of polymers.

Table 2. Polarizability for common covalent bonds.

Covalent bond	Molar polarizability	Covalent bond	Molar polarizability
C–H	1.70	C=O	3.42
H–H	2.08	C–OH	3.23
C–C	1.21	C–NH–C	5.13
C=C	4.15	C–F	1.83
C–O	1.42	C–Cl	6.57

3.2. Calculation of $\varepsilon_{\rm re}$ for polymers

With the data in Table 2 and the density of the polymers, the key parameter $\varepsilon_{\rm re}$ for different types of polymers can be calculated. The calculation process is presented by taking PMMA (organic glass) as an example. Figure 2 shows the chemical structure of PMMA. Based on this structure, the molecular weight and the number of various covalent bonds in a single PMMA unit are summed up, as listed in Table 3. In this table, it is found that $M = 100.12$ and $R = 24.7$. By taking into account that ρ (PMMA) = 1.18–1.19 g/cm^3, $\varepsilon_{\rm re}$ (PMMA) is calculated to be 2.23–2.24.

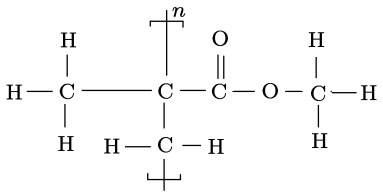


Fig. 2. The chemical structure of PMMA.

Table 3. R and M for PMMA.

Number of covalent bond	Σr_i	Species of atom	$\Sigma n M_{\rm e}$
4C–C	4×1.21	5C	5×12.0107
8C–H	8×1.70	8H	8×1.00794
2C–O	2×1.42	2O	2×15.9994
1C==O	1×3.42		
$\Sigma r_i = R = 24.7$		$\Sigma n M_{\rm e} = M = 100.1158$	

Note: n represents the atom number in a single unit for a type of polymer, and $M_{\rm e}$ represents the weight of this kind of atom. For example, for C its atom number is 5 and $M_{\rm e}$ is 12.0107.

Similarly, $\varepsilon_{\rm re}$ for the other common polymers used in high-voltage devices are also calculated, as listed in Table 4. As a comparison, the normal dielectric constant $\varepsilon_{\rm r}$ for these polymers are also listed. From Table 4, one can predict the $E_{\rm BD}$ relations for common polymers. For example, the $E_{\rm BD}$ relations for PE (polyethylene), PTFE (polytetrafluoroethelene), PMMA and Nylon are

$$E_{\rm BD}\,({\rm PTFE}) > E_{\rm BD}\,({\rm PMMA}) > E_{\rm BD}\,({\rm Nylon}) > E_{\rm BD}\,({\rm PE})\,. \quad (11)$$

Table 4. The parameters $\varepsilon_{\rm re}$ and $\varepsilon_{\rm r}$ for common polymers.

Polymers	$\varepsilon_{\rm re}$	$\varepsilon_{\rm r}$
PTFE	1.77–1.87	2.0–2.2
PMMA	2.23–2.24	3.6
PP	2.26	2.0–2.6
Nylon1010	2.28–2.32	3.0–3.6
PE	2.28–2.34	2.2–2.4
PVC	2.38	4–6
PS	2.39–2.42	2.4–2.65

4. Experiments to verify the $E_{\rm BD}$ relations

To verify the prediction in expression (11), some experiments were designed and conducted.

4.1. Experimental setup and procedure

The schematic diagram of the experimental setup can be found in Ref. [13], and consists of a nanosecond pulse generator,[19–21] a sample tank, a load tank and a diagnostic system. The electrode in the sample tank is sphere-plane shaped with a diameter of 60 mm to form a quasi-uniform electric field. The test samples are made of PE, PMMA, PTFE and Nylon (1010), all of which are cylindrical with a diameter of 40 mm and a thickness (d) ranging from 0.5 to 3.5 mm. Both the electrode and the test sample are immersed in transformer oil to avoid surface flashover. The typical output voltage waveform for this setup is trapezoidal with a pulse width of 8.5 ns, a rise and fall time of 1.5 ns and a maximum amplitude of 300 kV.

The experimental procedures are designed as follows: (i) choose 6–10 samples of one kind of polymer with the same thickness to obtain one $E_{\rm BD}$ versus d datum; (ii) change the polymer thickness and repeat procedure (i) five times to obtain one set of $E_{\rm BD}$ versus d data; (iii) change the polymer types three more times and repeat procedures (i) and (ii) to obtain the other three sets of $E_{\rm BD}$ versus d relations.

4.2. Experimental results

The experimental results are shown in Fig. 3. To show the regularity clearly, the standard error of each set of data is removed and the results are expressed

in a log–log coordinate system.[13,22] According to this figure, it is found that

$$E_{\rm BD}\,({\rm PTFE}) > E_{\rm BD}\,({\rm PMMA}) > E_{\rm BD}\,({\rm Nylon}) > E_{\rm BD}\,({\rm PE})\,. \quad (12)$$

Expression (12) is consistent with the theoretical prediction shown in expression (11).

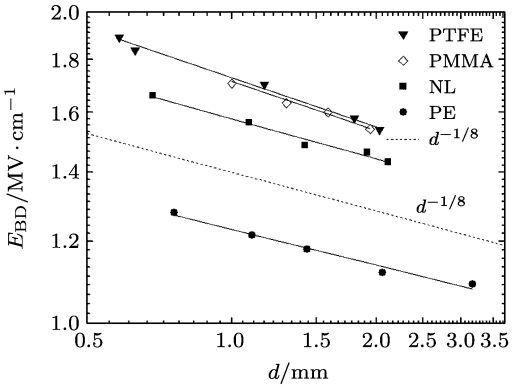


Fig. 3. The dependence of $E_{\rm BD}$ on the dielectric thickness of four types of polymer.

Besides the aforementioned experimental results, the experimental result in Ref. [10] also supports the polymer-type effect on $E_{\rm BD}$ expressed in Eq. (5). This set of experiments gives the results

$$E_{\rm BD}\,({\rm PMMA}) > E_{\rm BD}\,({\rm PVC}) > E_{\rm BD}\,({\rm PS})\,, \quad (13)$$

where PVC is polyvinyl chloride and PS is polystyrene. According to Table 4, the relation of $\varepsilon_{\rm re}$ between the three types of polymers is

$$\varepsilon_{\rm re}\,({\rm PMMA}) < \varepsilon_{\rm re}\,({\rm PVC}) < \varepsilon_{\rm re}\,({\rm PS})\,. \quad (14)$$

Expressions (13) and (14) agree with each other.

5. Conclusion and discussion

The polymer-type effect on $E_{\rm BD}$ on a nanosecond time scale has been investigated in this paper, and a qualitative formula to characterize the dependence of $E_{\rm BD}$ on polymer type is derived. It is concluded that the smaller the effective relative dielectric constant of a polymer, the larger the $E_{\rm BD}$ of this polymer. It is believed that this conclusion presents a feasible criterion for choosing polymers for scientists and engineers when designing insulating structures for high-voltage devices, especially pulsed power systems.

It is worth mentioning that before applying this formula and the conclusion, some preconditions should be clarified.

1) The formula are based on the electron impact ionization mechanism, which is an avalanche breakdown process belonging to electrical breakdown theory. If the breakdown mechanism is thermal, mechanical or chemical,[23] then the formula will not hold true.

2) The conclusion is established on a nanosecond time scale. When the time is extended to microseconds (μs), milliseconds (ms) or the ac/dc scale, the conclusion will not be applicable. The reasons for this lie in the fact that when the time scale gets bigger, there will be more polarization mechanisms. To solve this problem, the $\alpha_{\rm s}$ parameter in Eq. (6) should be added into the corresponding polarizability of the relevant polarization mechanisms.

3) The conjectures that ΔE and λ for each type of polymer are approximately the same require that the polymers should be perfect or quasi perfect. If there are too many defects or pores in the polymers, they will change the approximate values of ΔE and λ, which will also cause the inapplicability of the conclusion.

Future work is needed to improve the formula and the conclusion obtained in this paper.

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