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Determination of threshold electric field for charge injection in polymeric materials

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Accurately determining the threshold electric field at which charge injection from the electrodes starts is important for reliable operation of dielectric materials as the presence of charge in the material can lead to electric field enhancement, resulting in degradation and early failures of the material. The two methods in charge measurements that have been commonly used to find out the threshold field have been compared to the proposed method, which overcomes the drawbacks of the two methods. Such method offers (i) high sensitivity as the effect of capacitive charge has been eliminated and (ii) contributions from both mobile and slow charges; hence, providing a more accurate value for the threshold electric field. Based on the proposed method, it has been found that the threshold field for low density polyethylene is around 8 kV/mm, which is lower than the reported value obtained from the other methods.
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High-voltage direct current (HVDC) transmission is becoming more competitive compared to high-voltage alternate current (HVAC) transmission, owing to its many technical and commercial advantages over HVAC transmission system especially for the long-distance.[1,2] However, one of the drawbacks of the HVDC system is the issue associated with the easy formation of space charge in dielectric. When dc voltage is applied across a dielectric, electrons may overcome a potential barrier between electrode (metal) and the dielectric and enter into the dielectric.[3] The accumulation of those charges will cause local field enhancement in the bulk of the dielectric; hence, accelerate the degradation and ageing processes, resulting in early failure of the dielectric material. With the same dielectric and electrode, the amount of charge injection should be mainly dependent on the voltage level, i.e., the applied field. Therefore, for reliable operation of HVDC systems, it becomes important to accurately identify the applied field value at which the charges start to build up in the dielectric, i.e., the threshold field of charge injection.

For the threshold field of charge injection, An *et al*. suggested that the injected charges could be observed in polyethylene near the cathode even at an electric field around 5 kV/mm.[4] Their sample consists of two dielectric layers and two thin charge blocking layers (fluorinated polyethylene), placed in alternative between the two electrodes. The blocking layers are able to block charge movement, leading to charge accumulation at the interface. These observed charges were suggested to be caused by thermal assisted tunneling. However, this value is significantly lower than the value of 10 kV/mm mentioned in literature for polyethylene.[5,6] As the blocking layer was fluorinated polyethylene, its dielectric properties such as permittivity and conductivity are different from the normal one; therefore, it is possible the charges accumulated at the interface between the two layers may be related to the Maxwell-Wagner polarisation.

Over the years, different techniques have been used to obtain the threshold field including conduction current, electroluminescence (EL), and space charge detection measurement. Among these, space charge measurement is considered to be not only direct but also more accurate method to determine the threshold field. A value of more than 10 kV/mm has been identified for polymeric materials such as LDPE and cross-linked polyethylene (XLPE).[5,6]

Normally, when using space charge detection techniques, two approaches have been frequently used to characterise accumulated space charges in the dielectric: volts-off and volts-on measurements. Volts-off test indicates the process that at each measurement, the power supply is switched off for a short period time (around 5 s) for the purpose of expelling capacitive charges from the electrodes, once the measurement is completed, the power supply should be switched back on immediately, see details in Fig. 1. This method

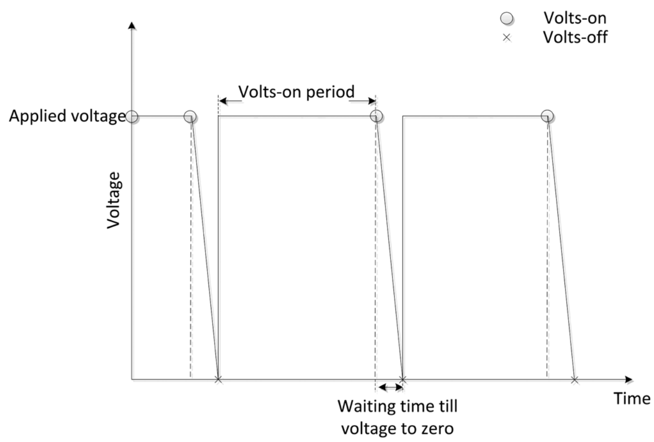

FIG. 1. Schematic diagram showing volts-on and volts-off measurement to detect space charge.

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could directly give accumulated space charges in the bulk and therefore, the threshold field could be determined.[5,6]

For volts-on test, the charge dynamics are recorded without turning off power supply. The peak values of signal, corresponding to positive/negative charges at electrodes, are plotted against the applied electric field. In the absence of space charge, a linear relationship between the peak values and the applied field exists. The presence of space charge in the sample will lead to a deviation from the straight line. The electric field value at which the deviation starts is identified as the threshold for charge injection. Fig. 2 shows schematically the principle of the method. Chen *et al*. have extended such method to draw X-plot, which gives threshold field of injected charges accumulation under ac condition.[7] However, for the accurate determination of threshold field for charge injection, there are some shortcomings of these two methods. Simply said, volts-off measurements may not be able to capture fast/mobile charges, and volts-on method may have problems associated with low sensitivity and requiring normalization. Therefore, in this letter, another method has been proposed to overcome those problems, and the method gives a more accurate field value at which charge injection occurs in polymeric materials.

In the present research, to demonstrate the effectiveness of the proposed method of identifying the threshold field, the pulsed electro-acoustic technique was employed to measure space charge in films of normal additive-free low-density polyethylene (LDPE) with thickness of $165 \pm 10\,\mu$m. The PEA is the most popular technique for measuring space charge in polymeric materials, since it was developed in 1980 s. The applied electric field started from 6 kV/mm and raised to 40 kV/mm. At each applied field, space charge profiles in LDPE were observed up to 1800 s. And within 1800 s, charge profiles were taken at 60 s, 180 s, 360 s, 600 s, 900 s, 1200 s, 1500 s, and 1800 s, respectively. For the sake of clarification in figures, only data recorded at 60 s, 600 s, and 1800 s will be shown in the results. To eliminate the influence of pre-stressing after each measurement, the used sample was replaced with another fresh one. The external voltage had to be adjusted in accordance with thickness variation of each sample to achieve the intended field.

Observation of space charges in volts-off intervals could be the most direct way to determine the threshold field of charge injection. To determine the threshold field value, the charge amount in the bulk Q_{abs} could be expressed as

$$Q_{abs} = \left(\int_0^{l_{neg}} |\rho_{neg}(x)| dx + \int_{D-l_{pos}}^{D} \rho_{pos}(x) dx \right) \cdot S, \quad (1)$$

where $\rho_{neg}(x)$ and $\rho_{pos}(x)$ are charge density distributions of negative and positive charges, l_{neg} and l_{pos} are thicknesses of negative and positive charge layer, respectively, and D is the thickness of the sample. It has been noted that negative charge injection occurs early and positive injection from the anode does not show up till a higher field. For example, for the present volts-off measurements, obvious positive charge accumulation is observed to occur above 25 kV/mm, see Fig. 3. Hence, before that, only the negative space charge peak near the cathode was counted. Plotting the calculated charge amount (using Eq. (1) on volts-off data) versus the applied electric fields, the threshold field could be determined around 12 kV/mm, see Fig. 4.

The peak values during volts-on measurements can also be used to determine the threshold field. However, this is not feasible for present experiment data. The reason is that the sample was replaced by another fresh one after each measurement to eliminate the effect of pre-stressing. Consequently, the peak values will be considerably affected by slight difference in thicknesses of LDPE films and possibly also a little difference in mechanical pressure exerting on each sample. This dilemma can be resolved by using reference data of each test for normalization. As illustrated in Fig. 5, maximum absolute values of reference signal's positive and negative peaks at reference voltage V_{ref} were denoted as A_{ref_+} and A_{ref_-}. Accordingly, the maximum absolute value of both peaks at applied voltage V_{app}, as A_{app_+} and A_{app_-}, could be normalized as $i_\pm$

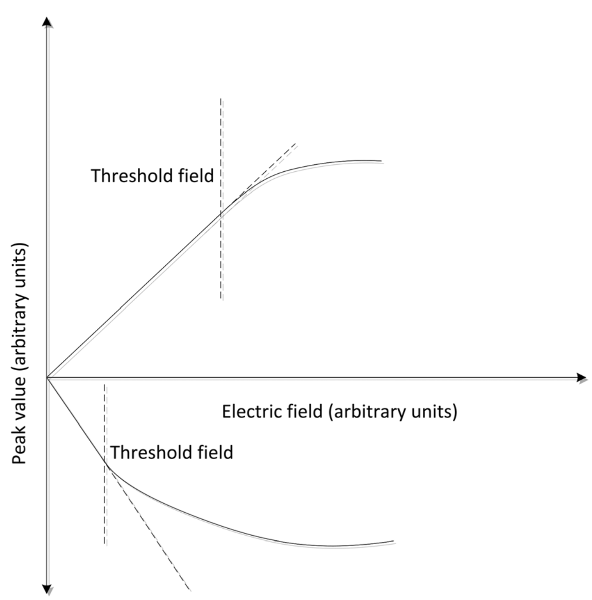

FIG. 2. Illustration of using the maximum peak value to determine threshold field under dc condition for positive and negative accumulated charges, respectively.

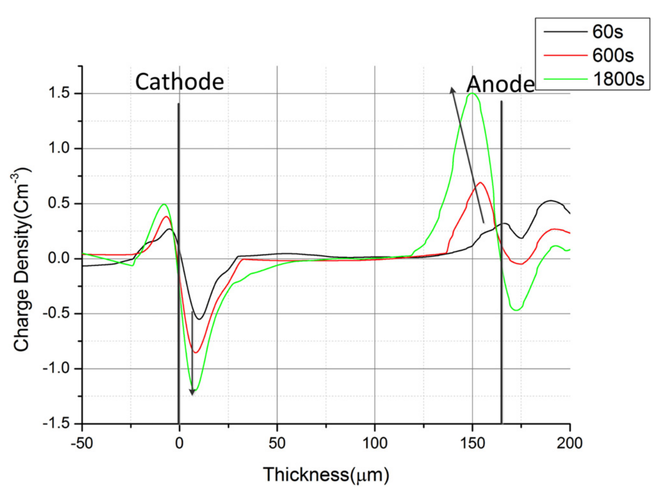

FIG. 3. Space charge dynamics from volts-off measurements applied field of 25 kV/mm on 165 μm LDPE film, showing bipolar injected charges.

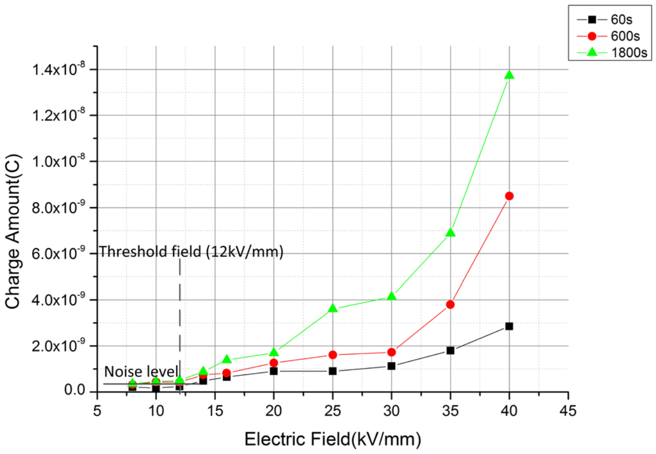

FIG. 4. Plot of charge amount versus applied electric fields ranging from 8 kV/mm to 40 kV/mm within 1800 s, using data of volts-off measurement, where threshold field of space charge injection was determined around 12 kV/mm.

$$i_{\pm} = \frac{A_{app\pm}}{\frac{V_{app}}{V_{ref}} A_{ref\pm}}. \quad (2)$$

Theoretically, if the value of i_+ or i_- equals 1, it should suggest zero space charge accumulation of such charge polarity in the bulk. Once the value falls below 1, i.e., $i_+ < 1$ or $i_- < 1$, it indicates positive or negative injected charges gathering in the vicinity of the anode or cathode.

Applying Eq. (2) to the data from volts-on measurement, the relationship between normalized peak values $i_{\pm}$ with the applied electric field could be found as obtained in Figs. 6 and 7. In these two plots, once i_+ or i_- is below the normalized threshold value 1, the corresponding electric field could be treated as the threshold of charge injection of such polarity. Considering the effect of noise, the threshold field corresponding to $i = 0.96$ has been selected. It can be seen from Fig. 6, positive charges' injection start to take place at 16 kV/mm, where $i_+ \approx 0.96$. Likewise, in Fig. 7, the threshold of negative charges' injection is about 10 kV/mm. As expected, the threshold field is also time-dependent. The longer the applied electric field, the lower the threshold field value for charge injection, therefore, it can be summarized that starting from about 10 kV/mm to 14 kV/mm, negative injected charges is predominant in the bulk, and after 16 kV/mm, the bipolar charges start to accumulate. Furthermore, the turn-back of i_- above 30 kV/mm, see Fig. 7, is ascribed to the positive charge domination, which induces negative image charges at the cathode, cancelling previous positive image charges.

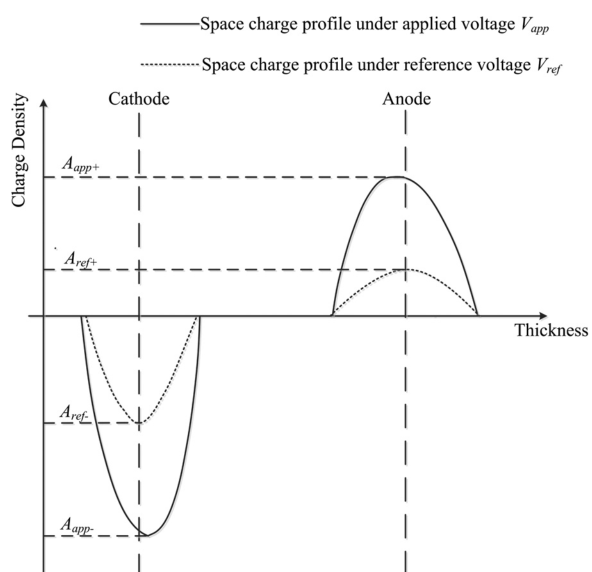

FIG. 5. Schematic representation of a typical space charge signal with bipolar injection to explain $A_{ref\pm}$ and $A_{app\pm}$.

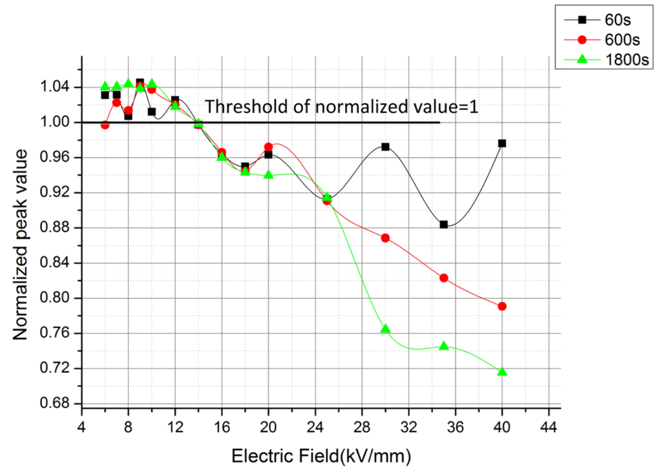

FIG. 6. Plot of normalized value i_+ versus electric field.

For the volts-off measurement, the multiple switching-offs during the measurement might lead to inaccurate result, as some space charges of higher mobility may not be included. The charges with high mobility can move out of the dielectric during the volts-off period. This has been proved by previous charge decay measurements on LDPE samples,[8,9] which showed rapid changes in charge amount immediately after turning off the power supply.

It is clear that peak values of volts-on data could not be directly used for determination of threshold field, as the sample has to be changed after measurements at each voltage level. Normalization has to be made on the data. Also, such method could not give the threshold field of total charges'

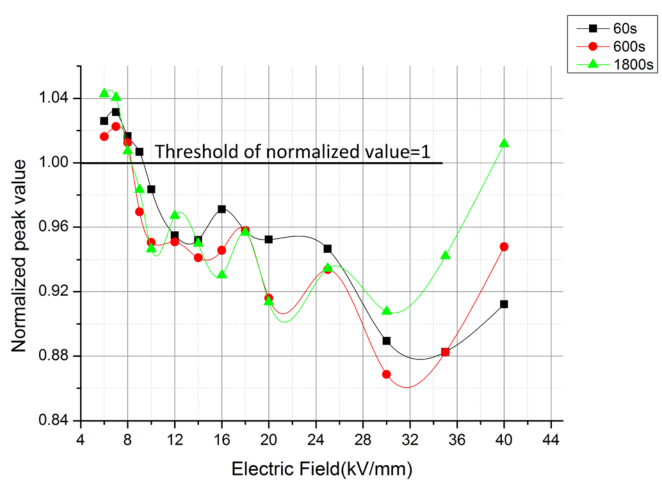

FIG. 7. Plot of normalized value i_- versus electric field.

injection; instead, threshold field of positive or negative charges injection separately. Additionally, as injected charges at lower voltage level are much smaller with respect to capacitive charges on the electrode, the sensitivity of peak value method might become a problem. In other words, injected charges might only cause little changes on the peak values, which might be harder to follow by comparison with direct observation of space charge in the bulk.

Here, we propose a subtraction method which can be employed to remove the capacitive charges from the electrodes. In this method, the reference data is multiplied by the ratio between the applied voltage and reference voltage, and this can be considered as the charge density data at applied voltage without effect of space charges in the bulk. The actual measured charge density data obtained from the volts-on measurement minus the multiplied will reveal the injected charges in the sample and its induced image charge at the electrodes. Mathematically, it can be expressed as

$$\rho_{acc}(x) = \rho_{app}(x) - \frac{V_{app}}{V_{ref}}\rho_{ref}(x), \quad (3)$$

where V_{ref} and V_{app} denote reference and applied voltages, $\rho_{acc}(x)$ represents space charge density after subtraction, and $\rho_{app}(x)$ and $\rho_{ref}(x)$ represent charge density at applied voltage and reference voltage, respectively.

The validity of Eq. (3) could be justified by applying a voltage multiple times higher than reference voltage on polymethyl methacrylate (PMMA) material, which has little charge injection below 30 kV/mm. It is expected that the subtracted results should be zero when Eq. (3) applies. Presently, a commercially purchased PMMA film with a thickness of 180 μm was used for the verification measurement. The reference signal was collected at 1 kV, then using the same sample, signal at 1.5 kV and 2 kV is also collected, see Fig. 8. Applying Eq. (3), the subtracted data of 1.5 kV and 2 kV were shown in Fig. 8 as well. It can be seen that the subtracted results for both voltages turn out to be nearly zero. This satisfies the expectation and proves the proposed subtraction method can be used as an effective way to find out accumulation of space charge in the sample; hence, the threshold field.

As mentioned previously, volts-off measurements may not able to capture some fast/mobile charges, and this can be verified by collecting charge data (volts-on) before turning off the power supply at each reading time and applying the subtraction method to get charge profiles without capacitive charges. Comparing the subtracted charge profiles of volts-on data to those directly collected during volts-off intervals, it has been found that injected space charges peaks obtained from volts-on data are generally higher than that from volts-off periods, as shown in an example in Fig. 9, where the applied field is 30 kV/mm. The reason for the difference in charge magnitude is the subtraction method includes the contribution from fast/mobile charges.

To conclude, if we apply the subtraction method on volts-on data, threshold field of charge injection could be determined with a better accuracy. By employing the subtraction method, i.e., Eq. (3), the charge amount at each field value can be estimated using Eq. (1). The relationship of charge amount against electric fields is illustrated in Fig. 10. Taking zoom-in from 6 kV/mm to 14 kV/mm, the threshold field is found to be 9 kV/mm for data collected at 60 s. If the data from 1800 s is used, the threshold field moves to 8 kV/mm, see details in zoom-in plot of Fig. 10. Moreover, from the zoom-in plot of Fig.10, even under low field from 6 kV/mm to 8 kV/mm, which is supposed to be little charge injection, the subtracted charge amount still increase with the voltage application time. It indicates that even under low fields, with the application time prolongs, charges continue to accumulate in the bulk little by little. This observation supports that the threshold injection field is also related to the applied voltage duration, especially at low fields. When the time of voltage application prolongs, charges continue to accumulate in the bulk little by little. This explains the observed increase in charge amount for 600 s and 1800 s below 8 kV/mm.

In summary, a subtraction method has been proposed to determine the threshold field of space charge injection in

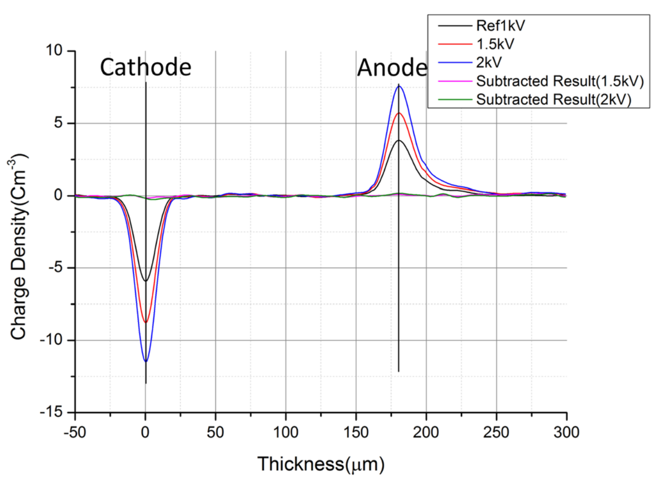

FIG. 8. Space charge profiles with external voltages applied at 1 kV (reference signal), 1.5 kV, and 2 kV on 180 μm the PMMA sample, and also the subtracted results, respectively, of 1.5 kV and 2 kV.

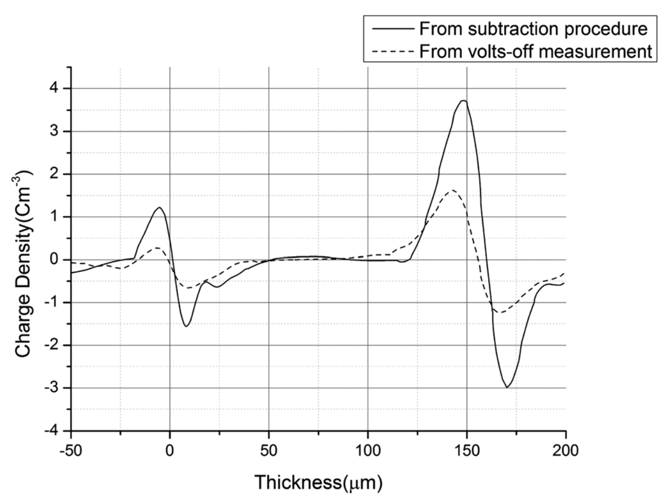

FIG. 9. Space charge dynamics with external field applied at 30 kV/mm at 1800 s on 170 μm LDPE film, where solid and dashed lines stand for charge profiles from the subtraction procedure and volts-off measurements, respectively.

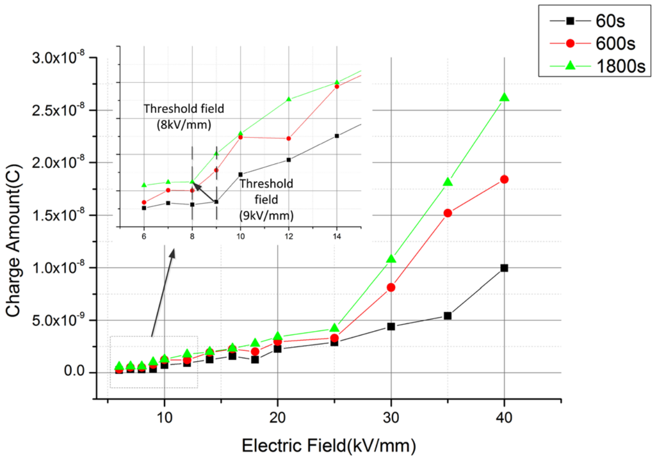

FIG. 10. Plot of charge amount versus applied electric fields from 6 kV/mm to 40 kV/mm, using subtraction method applied on volts-on measurement data with zoom-in plot of selected fields' range from 6 kV/mm to 14 kV/mm. With the increasing of application time of external voltage, threshold field moves from 9 kV/mm to 8 kV/mm within 1800 s, as implied by the arrow.

dielectrics. Compared to the commonly used methods, it offers significant advantages: (i) high sensitivity, as it eliminates the influence of capacitive charges on the signal and (ii) including all the charge contributions, as it uses the data from volts-on measurements.

The method has been validated by applying it to a PMMA film, where no charge injection occurs below 30 kV/mm. When it is applied to LDPE, a threshold field of 8 kV/mm is determined, which is lower than the reported value obtained from conventional methods, suggesting the method is more sensitive.

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