

Effect of Thermal Aging on Electrical Properties of Low Density Polyethylene

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Abstract – The thermal degradation of low density polyethylene (LDPE) will accelerate the production of carbonyl groups (C=O), which can act as the induced dipoles under high voltage. In this paper, we researched the dielectric properties and space charge behavior of LDPE after thermal aging, which can help us to understand the correlation between carbonyl groups (C=O) and electrical properties of LDPE. The spectra results show that LDPE exhibit obvious thermooxidative reactions when the aging time is 35 days and the productions mainly contain carboxylic acid, carboxylic ester and carboxylic anhydride, whose amount increase with the increasing of aging time. The dielectric properties show that the real permittivity of LDPE is inversely proportional to temperature before aging and subsequently become proportional to temperature after thermal aging. Furthermore, both the real and imaginary permittivity increase sharply with the increasing of aging time. The fitting results of imaginary permittivity show that DC conductivity become more sensitive about temperature after thermal aging. On this basis, the active energies of materials calculated from DC conductivity increase first and then decrease with the increasing of aging time. In addition, the space charge results show that the heterocharges accumulated near electrodes in LDPE change to the homocharges after thermal aging and the mean volume charge density increase with the increasing of aging time. It is considered that the overlaps caused by electrical potential area is the main reason for the increase of DC conductivity.

Keywords: Thermal aging, Dielectric properties, Morphology structure, Space charge distribution, Active energy

1. Introduction

Polyethylene is widely used as the insulation materials for the high voltage facilities due to its high DC and AC resistivity, low dielectric loss, and excellent mechanical properties and thermal stabilization [1-3]. Thus, the high temperature of high voltage facilities during operation will accelerate thermal oxidative reactions in polyethylene, and finally leading to the failure of insulation, such as the high temperature (about 90 °C) of cable cores [4-6]. Therefore, it is important to analyze the electrical properties of polyethylene after thermal aging.

To date, there are many researches focused on the mechanical, morphology, dielectric properties, voltage resistivity, and space charge behavior and so on. These researches revealed the relationship between molecular

structures and electrical properties of polyethylene. On this basis, some researches evaluate the remaining lifetime of insulations, which can give us the guidance to maintain the electrical facilities [7-12]. Furthermore, considering the high temperature of facilities during operation, some literatures researched the thermal analysis of polyethylene, which can reveal the thermal degradation of materials. These researches pointed out that the oxygen diffuse coefficient of amorphous area is much higher than that of crystalline area. Namely, the thermal oxidative reactions will mainly occur in amorphous area, which can lead to the β -scission and produce a large amount of free radicals and finally change the mechanical and electrical properties of materials [13-20]. Recently, there are some literatures reported the mechanical and electrical properties of polyethylene after thermal aging and pointed out that the thermal aging will decrease the insulation performance of materials. However, there are few literatures researched the dielectric properties, space charge behavior and the active energy of LDPE after thermal aging. The systematic researches about these properties can help us to analyze the relationship between thermal productions and the insulation properties, showing us more information to assess the statement of insulations in further.

Above all, in this paper we finished the 77 days thermal aging experiment of LDPE and researched the morphology,

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FTIR spectra, dielectric properties and space charge behavior of the samples during different aging time. We used the Arrhenius equation to calculate the DC conductivity and active energy of LDPE and revealed the role of C=O groups produced during thermal aging under high voltage strength.

2. Experiment

2.1 Materials

The base polymer used was an additive-free low density polyethylene (LDPE) with a melt flow index 2.1-2.2 g/min and a density of 0.910-0.925 mg/cm³. The LDPE were press modeled at 393K and at a pressure of 10MPa, to produce films with a thickness of about 150μm. Samples were degassed in a vacuum drier for 7 days at 353K. According to the Chinese Standard GB/T 11017-2002, the temperature of insulation for the high voltage cables can not extend 363K. Therefore, in this paper the samples were heated continuously in a temperature-controlled oven at 363K for 77 days. The properties of samples were tested when the aging time were 0, 14, 35, 56 and 77 days.

2.2 Scanning electron microscope imaging

The sample section surface, fractured in liquid nitrogen,

was observed by scanning electron microscopy (SEM, MERLIN Compact) in order to observe the morphology of LDPE after thermal aging.

2.3 FTIR analysis

FTIR was applied to evaluate the change of functional groups of LDPE after long-term thermal aging. The FTIR transmission spectra were obtained in the range of 400-4000 cm⁻¹ by using Bruker ALPHA FTIR spectroscopy.

2.4 Dielectric properties

The dielectric properties of the materials were measured in the frequency domain from 10⁻¹Hz to 10⁶ Hz at room temperature (25±1 °C) by using a Novocontrol ALPHA-A high resolution dielectric analyzer. Prior to dielectric analysis, gold electrodes were deposited onto both surfaces of the specimens by sputtering. The diameters of sputtered electrodes are 15mm. The results are the average measurements of 5 different specimens for each sample and the error in the measurement is within 2%.

2.5 Space charge measurements

The space charge measurements were carried out with a pulsed electro-acoustic (PEA) system, whose pulse width is 2-5ns, pulse amplitude is 200 V and output voltage is 0-

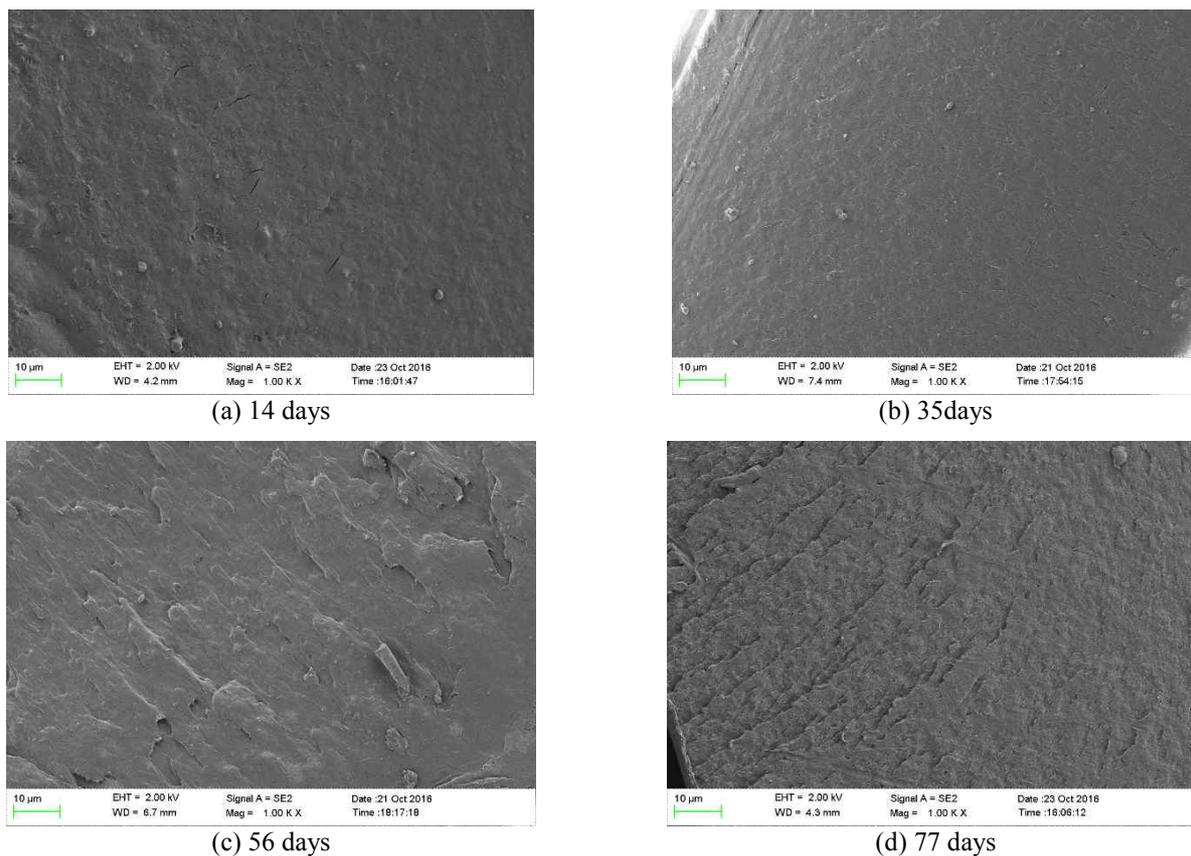


Fig. 1. Sectional morphology of LDPE with different aging time

20 kV. All samples were measured at room temperature (25±1 °C). In this paper, 20kV/mm DC electrical field was applied for 1200s and the space charge formation was confirmed with the polarization of 1200s. The reproducibility of the space charge behavior was confirmed by repeating the experiments five times for each group.

3. Equations and Units

3.1 SEM morphology

Fig. 1 shows the SEM images of LDPE with different thermal aging time. It can be seen that the sectional surfaces of samples are smooth when the aging times are 14 days and 35 days. Thereafter, with the aging time increase to 56 days, the sectional surface becomes rougher, as shown in Fig. 1(c). Additionally, many big cracks are observed when the aging time is 77 days. It can be concluded that the obvious oxidation reactions accelerated by thermal aging occurred when the aging time is 56 days, which may destroy the structure of LDPE.

3.2 FTIR spectra

FTIR spectroscopy analysis curves show the stretching vibrations of molecular bonds, which can reveal the new molecular groups of polymer after thermal aging effectively. Fig. 2 is the FTIR spectra of LDPE with different aging time. In this figure, the strong peaks at 720 cm⁻¹, 1464 cm⁻¹, 2850 cm⁻¹ and 2913 cm⁻¹ represent C-H stretching vibrations. Moreover, in comparison with the unaged samples, three new stretching peaks can be found at 1713cm⁻¹, 1739 cm⁻¹ and 1780 cm⁻¹ when the aging time is over 35 days corresponding to the carboxylic acid, carboxylic ester and carboxylic anhydride produced during thermal aging [2].

In order to assess the thermal aging degree of LDPE, we calculate the carbonyl index by using formula (1) and show the results in Fig. 2[21].

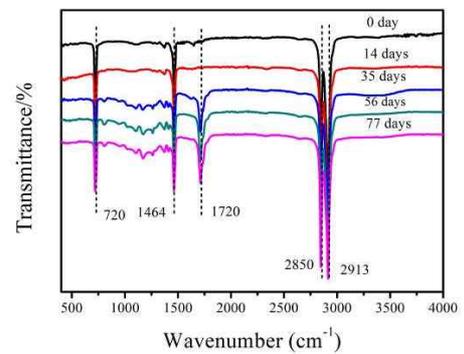
$$\text{Carbonyl index} = \frac{P_c}{P_r} \quad (1)$$

Where, P_c represents the peak value fitted by Lorentz formula, P_r represents the peak value at 1464 cm⁻¹.

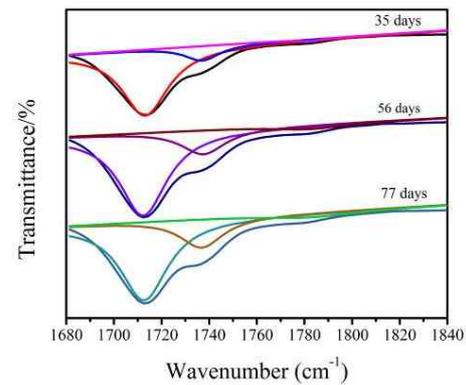
It is noted that the carbonyl index increase with the increasing of aging time. Meanwhile, this value increase dramatically from 35 days to 56 days. However, after that time, this value changes slightly. Namely, the molecular groups distributed in LDPE which are easy oxidized are consumed when the aging time is 56 days and the structure of materials become more stabilization to some certain degree when the aging time increase to 77 days.

3.3 Dielectric properties

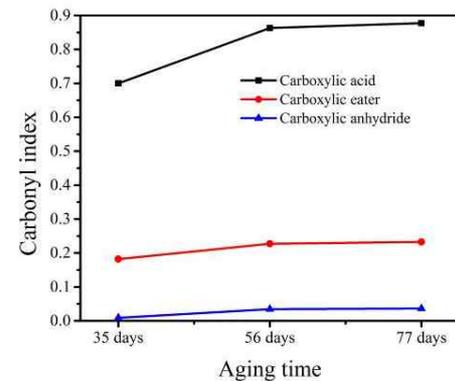
According to the FTIR spectra, the thermal aging degree



(a) FTIR spectra



(b) Fitting results



(c) Carbonyl index

Fig. 2. The FTIR spectra and carbonyl index of LDPE after thermal aging

of samples is the highest when the aging time is 77 days. Therefore, we exhibit the temperature dependence of real permittivity when the aging times are 0 days and 77 days in this paper, which can reveal the changes of real permittivity effectively after thermal aging. In figure 3, we can see the real permittivity of LDPE is about 2.36 at room temperature, and this value increase to 3.5 when the aging time reaches 77 days, which means the thermal aging can increase the real permittivity of LDPE. Furthermore, it is interesting that the real permittivity of LDPE decrease with the increasing of temperature before aging but increase with the increasing of temperature after thermal aging.

Additionally, for the aged samples, the real permittivity at low frequency increase dramatically when the temperature increase to 50°C and this phenomenon become more obvious when the temperature is 60°C. Namely, in comparison with the unaged samples, the aged samples have the more obvious electrode polarization, which may be caused by the hopping of ions.

As we known, the thermal expansion of LDPE will increase the volume of samples under high temperature, which may decrease the number of dipoles per unit volume with the increasing of temperature and subsequently present the decrease of real permittivity with the increasing of temperature, as shown in Fig. 3(a). Thus, with the appearance of thermal oxidative in LDPE, the β-scission of polymer chains will produce smaller molecular chains and free radicals. These productions may act as induced dipoles affected by electrical strength, which can increase the real permittivity of samples. Additionally, with the increase of temperature, these induced dipoles can move more easily, exhibiting the increase of real permittivity with the increasing of temperature as shown in Fig. 3(b).

Imaginary permittivity can reveal the dielectric loss of polymer, which is an important element for the electronic

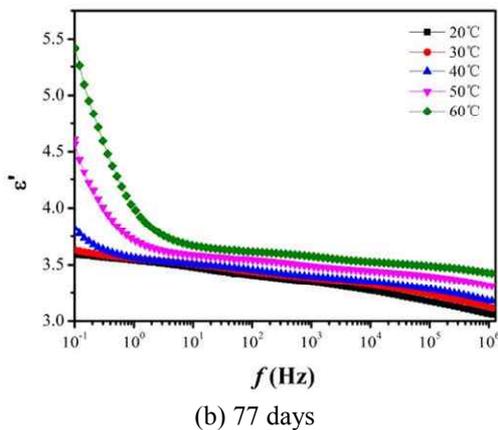
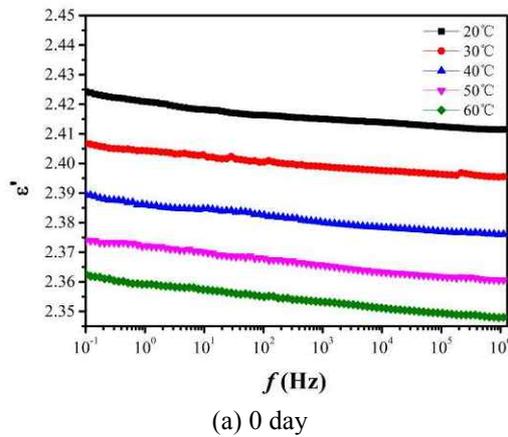


Fig. 3. The real permittivity of LDPE dependent on the temperature

equipment, energy storage device and also high voltage facilities. In this paper, Fig. 4 shows the temperature dependence of imaginary permittivity of LDPE before and after thermal aging.

According to the Fig. 4, it is obvious that imaginary permittivity of LDPE has two relaxation peaks. We labeled one of them around 10² Hz as A relaxation peak. Because the maximum frequency is 10⁶ Hz in this paper, we can only observe a part of another relaxation peak and we labeled this one as B relaxation peak. It is noted that the A relaxation peak shift towards to higher frequency with the increasing of temperature, which means the higher temperature gives the dipoles more energy to move with the change of electrical field. By comparing all the samples, it can be seen that the imaginary permittivity become more and more easily to be affected by temperature when the aging time is over 14 days. As temperature is 60°C, the imaginary permittivity at 10⁻¹ Hz increases to 7.8 when the aging time is 77 days. Based on the results of FTIR spectra, showing that the oxidative reactions happened when the aging time is over 14 days, we can conclude that the increasing of imaginary permittivity is contributed by the thermal degradation of LDPE.

In order to analyze the dc conductivity and the relaxation peaks of materials, we use formula (2) to fit the imaginary permittivity. In this formula, it contains three different phenomena, namely, DC conduction, relaxation due to hopping of ions, and A, B relaxation peaks[22].

$$\begin{aligned} \epsilon''(\omega) = & \frac{\sigma_{dc}}{\epsilon_0 \omega} + \frac{\Delta\epsilon_\alpha (\omega\tau_\alpha)^{1-\gamma_\alpha} \cos(\gamma_\alpha \pi / 2)}{1 + 2(\omega\tau_\alpha)^{1-\gamma_\alpha} \sin(\gamma_\alpha \pi / 2) + (\omega\tau_\alpha)^{2(1-\gamma_\alpha)}} \\ & + \frac{\Delta\epsilon_\beta (\omega\tau_\beta)^{1-\gamma_\beta} \cos(\gamma_\beta \pi / 2)}{1 + 2(\omega\tau_\beta)^{1-\gamma_\beta} \sin(\gamma_\beta \pi / 2) + (\omega\tau_\beta)^{2(1-\gamma_\beta)}} \\ & + \frac{\Delta\epsilon_{ion} (\omega\tau_{ion})^{1-\gamma_{ion}} \cos(\gamma_{ion} \pi / 2)}{1 + 2(\omega\tau_{ion})^{1-\gamma_{ion}} \sin(\gamma_{ion} \pi / 2) + (\omega\tau_{ion})^{2(1-\gamma_{ion})}} \end{aligned} \quad (2)$$

Where, ϵ_0 is the permittivity of vacuum in Fm-1. $\Delta\epsilon_\alpha$ and $\Delta\epsilon_\beta$ are the increase in imaginary permittivity due to A relaxation peak and B relaxation peak respectively. τ_α , γ_α and τ_β , γ_β are the time constant and the parameter showing the deviation of A relaxation peak and B relaxation peak from a Debye type, respectively. $\Delta\epsilon_{ion}$, τ_{ion} and γ_{ion} are those for hopping of ions.

The fitting results of imaginary permittivity when the aging times are 0 day and 77 days are shown in Fig. 5 and

Table 1. The DC conductivity calculated by fitting results

	0 day	14 days	35 days	56 days	77 days
20°C	1.28E-15	1.35E-15	1.43E-14	1.01E-13	1.61E-13
30°C	4.18E-15	3.88E-15	9.37E-14	3.84E-13	9.15E-13
40°C	1.27E-14	1.33E-14	4.87E-13	2.24E-12	4.35E-12
50°C	3.55E-14	3.77E-14	2.71E-12	9.73E-12	1.36E-11
60°C	9.45E-14	9.99E-14	7.79E-12	2.98E-11	3.78E-11

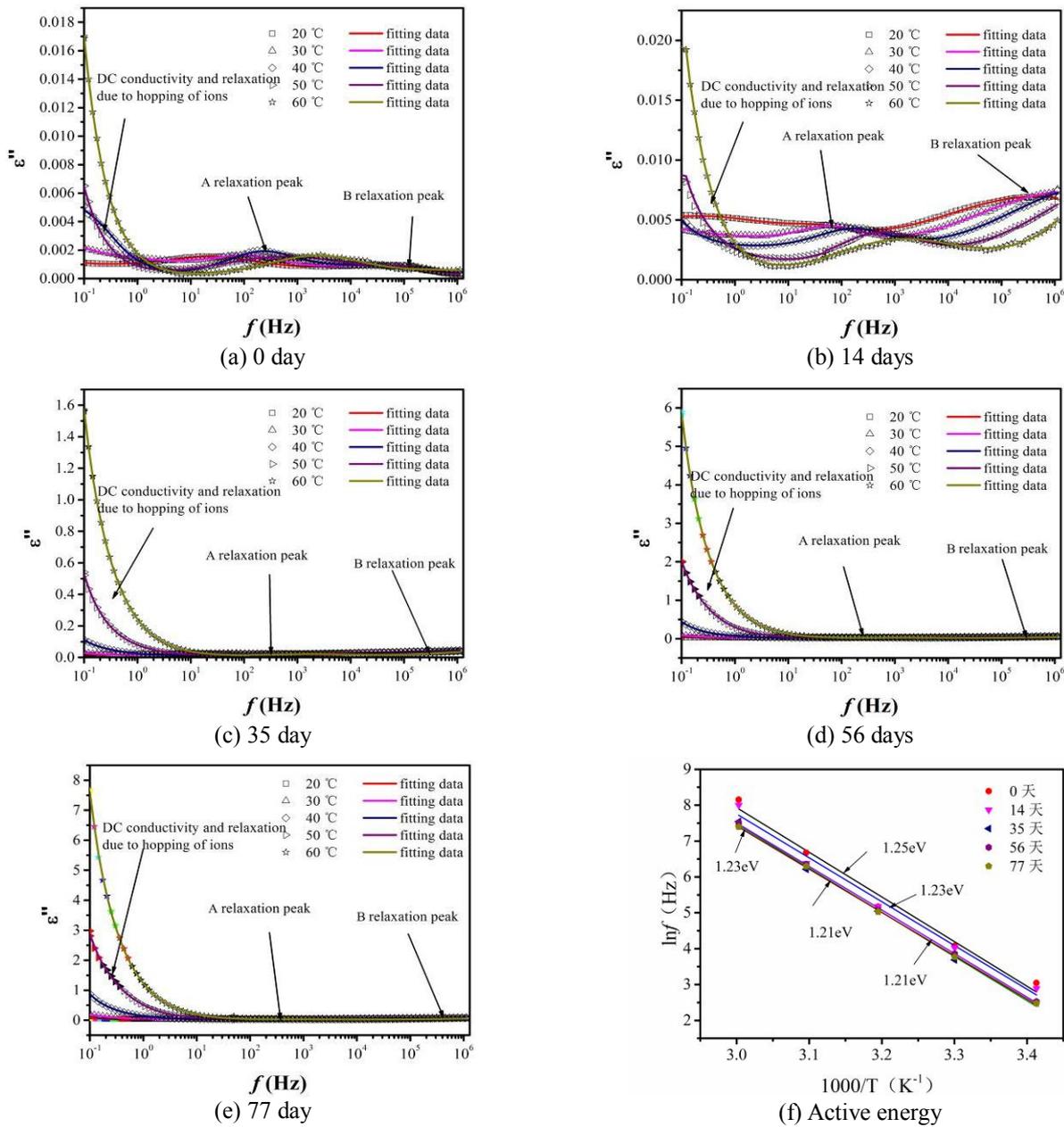


Fig. 4. The imaginary permittivity and the active energy of LDPE with different aging time

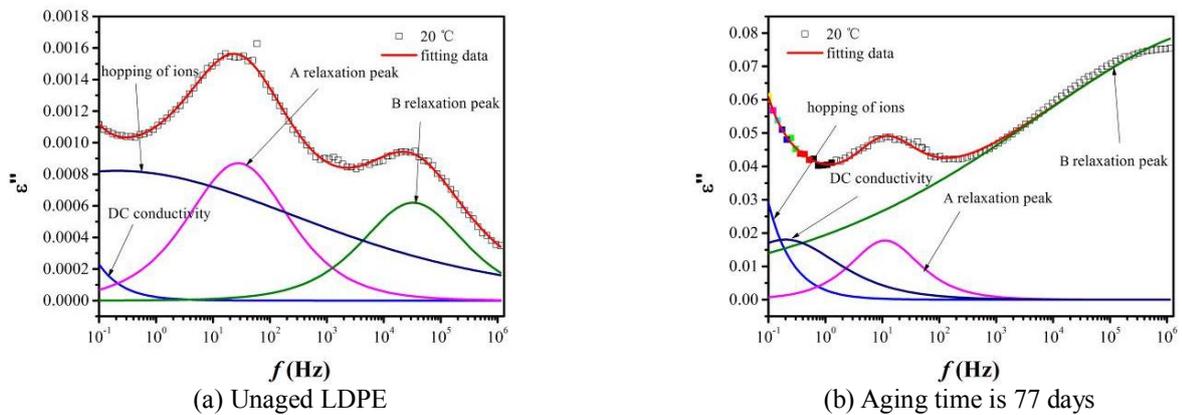


Fig. 5. The fitting results of imaginary permittivity

the DC conductivity are shown in Table 1. From this figure, we can see the contribution of the above mentioned four parts to the imaginary permittivity directly. According to the Table 1, it is noted that the DC conductivity of samples increase with the increasing of aging time. When the aging time is 77 days, the DC conductivity has the highest value. Furthermore, we use the Arrhenius equation to describe the relationship of DC conductivity and temperature[23].

$$\sigma_{dc} T \propto \exp(-E_{dc} / kT) \quad (3)$$

Where, σ_{dc} is the DC conductivity. T is the temperature. E_{dc} is the active energy and k is the Boltzmann constant.

The results are shown in Fig. 4(f), exhibiting the active energy of LDPE with different aging time. It can be seen that the active energies of LDPE decrease with the increasing of aging time. As we know, the active energy reflects the chemical active of materials. Thus, we can see that the chemical active of LDPE decrease with the increasing of aging time.

3.4 Space charge distribution

Space charge will accumulate in polymer insulations under high DC voltage, which may distort the electrical strength and hence accelerate degradation. PEA method can reveal the amount and the location of accumulated space charge in materials and help us to understand the space charge behavior in LDPE after thermal aging. Fig. 6(a) shows the space charge distribution in LDPE with the different aging time. The curves in the figure represent the space charge accumulation of material when the polarization time is 1200s.

From the figure 6, we can see heterocharges are accumulated near cathode electrode, which may be caused by the decomposition of residues in LDPE under high voltage [12]. However, when the aging time is 14 days, the homocharges occurred near electrodes and whose amount increase with the increasing of aging time. When the aging

time reaches 77 days, the homocharges near electrodes have the highest amount. Furthermore, in order to analyze the space charge amount directly, we use formula (4) to calculate the mean volume charge density of LDPE [24].

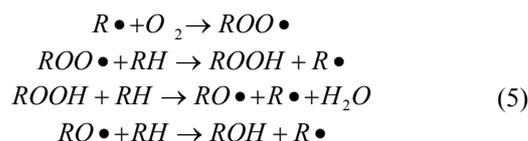
$$q(t; E_p) = \frac{1}{x_1 - x_0} \int_{x_0}^{x_1} |q_p(x, t; E_p)| dx \quad (4)$$

Where, x_0 and x_1 are the positions of anode and cathode respectively. t is the depolarization time, E_p is the polarization voltage, and $q_p(x, t; E_p)$ is the charge density in the samples. Induced charges at the electrodes are not taken into account.

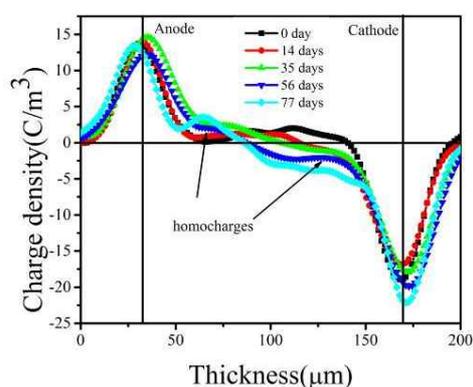
The results calculated by formula (4) are shown in Fig. 6(b). It is noted that the mean volume charge density of LDPE decrease first when the aging time is 14 days and then increase dramatically with the increasing of aging time. Especially, when the aging time is 77 days, the value of the charge density grows to 3.5 C/m^3 .

4. Discussion

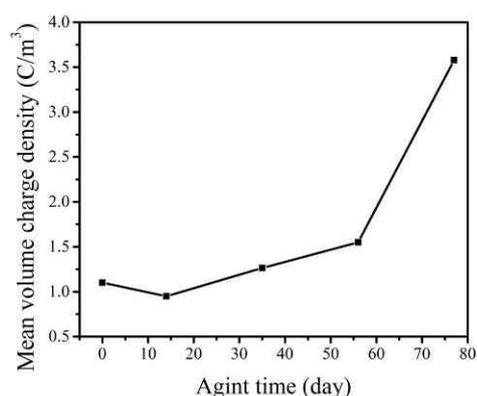
Based on the past researches, oxygen can react with the tertiary carbon atoms and then produce alkyl hydroperoxide and free radicals and also lead to the β -scission of polymer chains. The main processes are showing as follows:



From these formulas, we can see that the free radicals can obtain the hydrogen atom from alkyl hydroperoxide to produce alkoxy radicals, alkyl radicals and the water and also lead to the β -scission of polymer chains. Thus, the free radicals are the key productions during the thermal aging of LDPE. Additionally, the increase of free radical will also



(a) Space charge distribution



(b) Mean volume charge density

Fig. 6. Space charge distribution and mean volume charge density of LDPE with different aging time

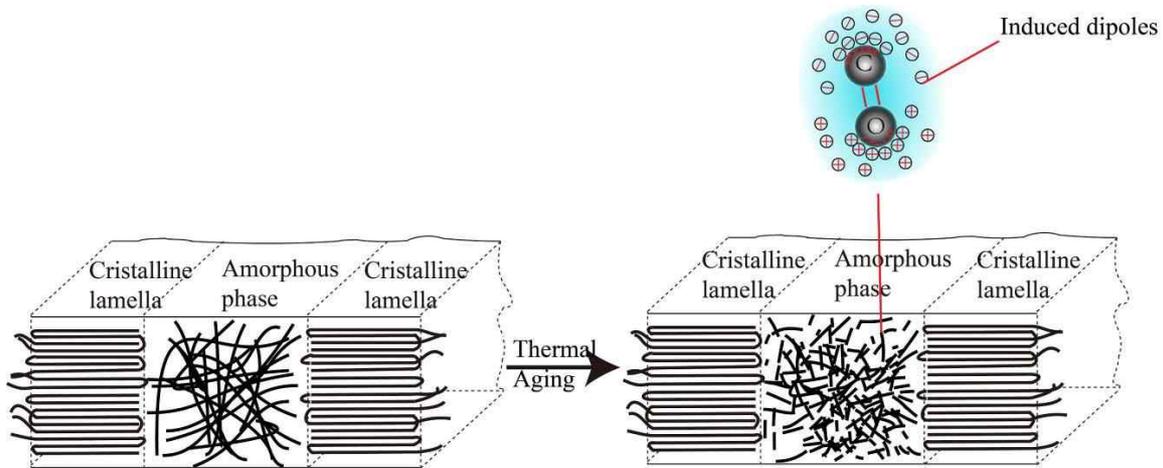


Fig. 7. Illustration of LDPE before and after thermal aging

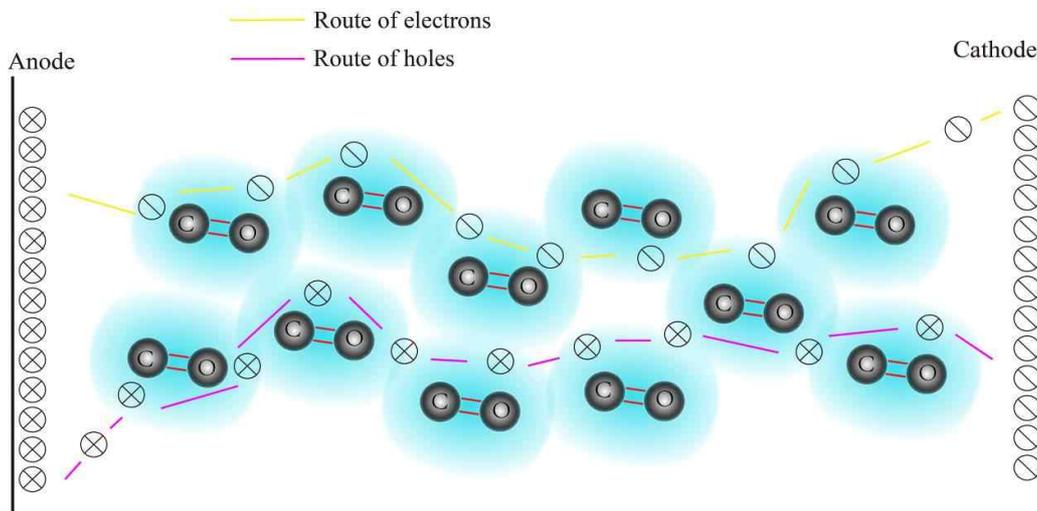


Fig. 8. Illustration of movement of carriers in induced electric field produced by C=O dipoles

lead to the increase of carbonyl groups (C=O).

As we know, the space charge accumulation is caused by the charge traps distributed in the materials, which can trap carriers injected from electrodes. As we see from the Fig. 6, the space charge behavior of LDPE shows obvious homocharges near electrodes after thermal aging. This may be caused by two reasons: 1. During thermal aging, the surfaces of the samples are exposed in air, leading to the thermal-oxidative aging and hence destroy the structure of the surface. Namely, the amount of defects distributed in this area will increase after thermal aging. These defects can act as charge traps and thus increase the homocharges near electrodes. 2. According to the research of T. Takada [25], the C=O groups can act as induced dipoles under high voltage strength, whose electrical potential area can trap carriers surrounded them. Namely, the C=O groups can be thought as charge traps, which lead to the homocharges after thermal aging. Fig. 7 shows the change of structure of LDPE after thermal aging. The long polymer chains become to smaller one because of the β -scission of polymer chains.

Furthermore, based on the fitting results of permittivity, we can see that the DC conductivity of LDPE increase with the increasing of aging time. As we mentioned above, T. Takada[25] proved that the C=O groups can be thought as the charge traps. And the calculation results from their research show the charge depth contributed by C=O groups is about 0.45 eV, which means the C=O groups can be thought as shallow charge traps. Namely, the carriers can move by trap and detrap to across their electrical potential area. Therefore, with the increasing of C=O groups during thermal aging, the overlaps of electrical potential area can prove the route for carrier, which can be thought as the “quasi- conductivity area” and lead to the increase of DC conductivity finally, as shown in Fig. 8. Furthermore, the homocharges accumulated near electrode will enhance the electrical strength in the samples, which can also contribute to the increase of DC conductivity to some certain degree. Additionally, due to the increasing of DC conductivity after thermal aging, the homocharges near electrodes extended into materials.

5. Conclusion

This paper researched the morphology, FTIR spectra, dielectric properties and space charge behavior of LDPE after thermal aging. The main conclusions are as following:

1. The thermooxidative reactions are observed in sample when the aging is over 35 days and the main productions are carboxylic acid, carboxylic ester and carboxylic anhydride, whose amount increase with the increasing of aging time. Meanwhile, the thermal aging may destroy the morphology structure of LDPE leading to the cracks on the sectional surface.
2. The real permittivity of LDPE is proportional to temperature before aging but it becomes inversely proportional to temperature after thermal aging. Thermal aging contribute to the increase of real and imaginary permittivity. Furthermore, the DC conductivity of LDPE may increase with the increasing of aging time based on the fitting results of imaginary permittivity. This phenomenon may be caused by the overlaps of electrical potential area provided by C=O groups, which can be thought as "quasi-conductivity area".
3. The heterocharges accumulated near electrodes in LDPE change to homocharges after thermal aging. The mean volume charge density decrease first and then increase with the increasing of aging time. The charge traps caused by the C=O groups and defects may be the main reason for the increase of homocharges after thermal aging.

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