

Effect of Ethylene Ionomers on the Properties of Crosslinked Polyethylene

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ABSTRACT: Most premature failure of underground crosslinked polyethylene (XLPE) cables in service, a matter of great concern, is due to aging induced by water treeing. To improve the water-tree resistance, sodium-neutralized poly(ethylene-co-acrylic acid) (EAA-Na) ionomers were blended with XLPE; the EAA-Na ionomers were prepared through the neutralization of sodium hydroxide and poly(ethylene-co-acrylic acid). A series of XLPE/EAA-Na ionomer blends were investigated through the measurement of the water absorption ratio, water treeing, and mechanical and dielectric testing; the results strongly suggested that EAA-Na ionomers could improve the water-tree resistance of XLPE, and the

XLPE/EAA-Na blends retained excellent mechanical properties and dielectric properties. Moreover, through the characterization of XLPE/EAA-Na blends with Fourier transform infrared spectrometry, dynamic mechanical analysis, and scanning electron microscopy, it was found that the neutralization reaction could be achieved completely; the XLPE and EAA-Na ionomers were partially compatible, so the EAA-Na ionomers could be dispersed well in the matrix with the process examined in this study. © 2006 Wiley Periodicals, Inc. *J Appl Polym Sci* 103: 3483–3490, 2007

Key words: blends; ionomers; polyethylene (PE)

INTRODUCTION

Over the last 20 years, crosslinked polyethylene (XLPE) has progressively replaced low-density polyethylene (LDPE) in extruded cable insulation because of its good dielectric properties and adequate mechanical properties at high temperatures at which LDPE becomes too soft.^{1–3} However, mechanical resistance is just one of the many polymer properties to be considered in choosing a material for cable insulation. A very important property for polymer-insulated power cables is the water-tree resistance of the material because polymer-insulated power cables are laid in an underground environment, and water from the soil can slowly diffuse through the protective layers of the cable, forming treelike structures—water trees—that are actually water-filled microcavities.^{1,2} Since the first report on water trees was published nearly 30 years ago, much research has been devoted to the phenomenon.⁴ However, water treeing is still known as the main cause of insulation breakdown in XLPE power cables, especially in medium-voltage cables.^{1,2}

Therefore, because of the importance of water treeing as a cause of insulation breakdown, we need to develop water-tree-resistant materials for power-cable insulation. In response to this requirement, a number of studies have been carried out. For example, Lee and coworkers^{5–8} improved the water-tree resistance of polyethylene by using ethylene vinyl acetate (EVA) and poly(ethylene glycol) (PEG) as additives. However, EVA and PEG are polar polymers that have poor compatibility with polyethylene, so they cannot be well dispersed throughout the matrix. In addition, lipophilic surfactants have been used as additives, improving the water-tree resistance of polyethylene.^{9,10} However, the surfactants are likely to be lost when they come into contact with water, and this will degrade the water-tree resistance of cable insulation. Thus, improved water-tree resistance can be obtained with additives that possess sufficient hydrophilicity to allow polyethylene to link water molecules and prevent them from diffusing into the material and possess sufficient compatibility with polyethylene to prevent the loss or clustering of the additives and keep the diffused water distributed evenly in the mass.¹¹

Because ethylene ionomers are copolymers consisting of methylene sequences as nonpolar segments and small amounts of methacrylic acid or acrylic acid residues as ionic units, which are often used as compatibilizers for polyethylene and polar polymers,^{12–15} poly(ethylene-co-acrylic acid) (EAA) ionomers should be

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used as additives for improving the water-tree resistance of XLPE. However, a systematic study of XLPE modified by ionomers cannot be found in the literature to the best of our knowledge. Hence, the purpose of this work is to study the effects of EAA ionomers on the water-tree resistance of XLPE. To improve the water-tree resistance, a series of sodium-neutralized poly(ethylene-co-acrylic acid) (EAA-Na) ionomers were *in situ* formed in an XLPE matrix through the neutralization of sodium hydroxide (NaOH) and EAA, and the neutralization reaction of NaOH and EAA was investigated. Moreover, the compatibility and morphology of XLPE/EAA-Na blends were also explored. In addition, we studied the effects of the EAA-Na ionomer contents and neutralization degree on the hydrophilicity, mechanical properties, dielectric properties, and water treeing of XLPE/EAA-Na blends.

EXPERIMENTAL

Materials

The polyethylene used in this study was linear low-density polyethylene (LLDPE; LL4004) from Exxon-Mobil (Riyadh, Saudi Arabia), which had a melt flow index (MFI) of 3.6 g/min by ASTM D 1238 and a density of 0.924 g/cm³ by ASTM D 4703/D1505. EAA was ESCOR 5020 from ExxonMobil Chemical (Antwerp, Belgium), which had an MFI of 8 g/min by ASTM D 1238 and a density of 0.933 g/cm³ by ASTM D 4703/D1505. NaOH (analytical reagent) and sodium chloride (NaCl; analytical reagent) were from Shanghai Medicine Chemical Co., Ltd. (Shanghai, China). Dicumyl Peroxide (DCP) (chemical reagent) was obtained from Shanghai Gaoqiao Petroleum Co., Ltd. (Shanghai, China).

Sample preparation

Certain amounts of LLDPE, NaOH, EAA, and DCP were mixed in a Haake RC90 rheometer (Karlsruhe, Germany) at 130°C and at a rotor speed of 60 rpm for 8 min. Afterwards, all samples were compressed at 170°C and 10 MPa for 15 min, and sheets 1 mm thick were obtained.

The EAA ionomers in all the samples reported in this article are denoted EAA-*xM*, where *x* and *M* are the neutralization degree and metal cation, respectively.

Infrared transmission spectra analysis

A Paragon 1000 Fourier transform infrared (FTIR) spectrometer from PerkinElmer Corp. (Boston, MA) was used to perform infrared transmission spectral analysis on the samples. The samples were pressed into films 100 μm thick.

Dynamic mechanical analysis (DMA)

The dynamic mechanical experiments were performed with a TA Instruments DMA 2890 analyzer. Oscillation tests from -150 to 130°C at a heating rate of 3°C/min were performed with the single-cantilever accessory at a frequency of 1 Hz and an amplitude deformation of 1 μm.

Hydrophilicity

The hydrophilicity of the samples was evaluated by the measurement of the water uptake under a control condition.¹¹ The measurement of the water uptake was carried out with a square sample, with a side length of 50 mm and a thickness of 1 mm, by the immersion/gain method. First, a sample was dried for 72 h at 90°C in an oven, and its original weight (*M*₀) was recorded. Then, the sample was immersed in a sufficient amount of deionized water at 80°C to accelerate the speed of the tests. The weight of the wet sample (*M*_{*t*}) was determined after the removal of the surface water through blotting with filter paper at different times until the absorption equilibrium was reached.

The water absorption ratio (WAR) was calculated as follows:

$$\text{WAR (\%)} = (M_t - M_0)/M_0 \times 100 \quad (1)$$

Mechanical properties

The tensile tests were performed according to ASTM D 638-2003 in an Instron series IX 4465 materials tester, with a grip separation rate of 250 mm/min, with 1-mm-thick specimens.

Dielectric properties

The volume resistivity was measured on a ZC-36 megohmmeter (Shanghai Precision and Scientific Instrument Corp., Shanghai, China) according to ASTM D 6095-2005. The dielectric constant and the dissipation factor were measured on a QS30 high-voltage bridge (Shanghai Huguang Corp., Shanghai, China) according to ASTM D 150-2004. The dielectric strength was measured on an AHDZ-10/100 alternating-current dielectric strength tester (Shanghai Lanpotronics Corp., Shanghai, China) according to ASTM D 149-2004.

Morphology

A Hitachi S520 scanning electron microscope (Tokyo, Japan) was used for morphology observations of the samples. All specimens were prepared by the immersion of test pieces in liquid nitrogen before breaking.

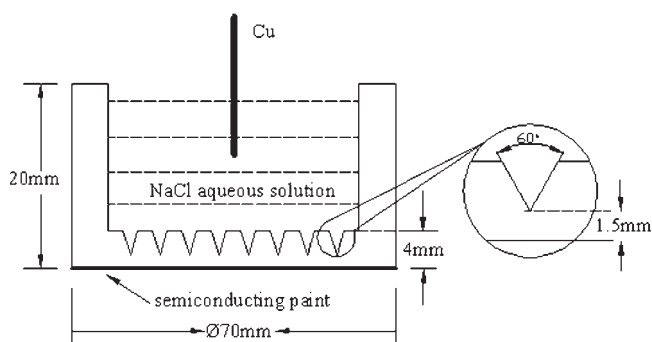


Figure 1 Schematic diagram of the test cell for WTL measurements. The electrolyte was a 1.0M NaCl aqueous solution; the test conditions were 5 kV, 1 kHz, 480 h, and room temperature.

The fractured surfaces of the test pieces were etched to remove the EAA–Na ionomers and coated with gold before the examination.

Water treeing

The cup-shaped samples were employed to carry out the experiment of water treeing. The schematic diagram of the test cell for the measurement of water treeing is shown in Figure 1. The cup-shaped samples with 20 conical defects were prepared through compression molding at 170°C for 15 min with a mold. The distance from the needle tip to the bottom of the cup was 1.5 mm, and the tip radius of each needle was 5 μm . The experiment of water treeing was carried out under the action of the applied voltage of 5 kV at the frequency of 1 kHz and room temperature for 20 days. After the experiment, all samples were stained with methylene blue, and the water-tree length (WTL) was determined by the measurement of the length of the longest branch of the tree. The value averaged from 20 defects was taken as the WTL of the samples.

RESULTS AND DISCUSSION

FTIR analysis of the *in situ* formed EAA–Na ionomers

The *in situ* preparation of the EAA–Na ionomers was confirmed with FTIR spectra [Fig. 2(a–d)]. EAA has a characteristic absorption peak at 1712 cm^{-1} corresponding to the C=O stretching vibration [Fig. 2(b)],¹⁶ so the blend also has an absorption peak at 1712 cm^{-1} when EAA is blended in XLPE [Fig. 2(c)].

When the hydrogen in the —C(=O)OH group is substituted by a cation, the C=O and C–O bonds attached to the same C atom are equalized to

—C(=O)O^- , which has a characteristic absorption peak at 1570 cm^{-1} .^{16,17} Thus, when EAA is neutralized

completely by NaOH in XLPE, the characteristic absorption bands of —C(=O)OH group shift, and the

characteristic absorption bands of —C(=O)O^- appear.

As shown in Figure 2(d), the presence of the absorption peak at 1570 cm^{-1} in the spectrum of the XLPE/EAA–1.0Na blend indicates that the hydrogen in the —C(=O)OH group is substituted by a cation, and

—C(=O)OH is changed to —C(=O)O^- ; this suggests that EAA reacts with NaOH and forms EAA–1.0Na ionomers.

DMA

The compatibility behavior of melt-mixed blends is often judged by the glass-transition temperature,^{18,19} so DMA is one of the best methods at present for characterizing the compatibility. As shown in Figure 3, both XLPE and the XLPE/EAA–1.0Na blend exhibit three loss peaks, which are designated α , β , and γ in decreasing order of temperature; however, EAA–1.0Na exhibits two loss peaks corresponding to β and γ , respectively. The high-temperature α relaxation is frequently attributed to the vibration or orientational motion within the crystallites, and γ relaxation is first due to the crankshaft movements of polymethylene chains.^{20–25} The β relaxation should be assigned to a greatly broadened glass–rubber transition in the amorphous phase.^{23–25} For XLPE/EAA–1.0Na blends, the β loss peak of EAA–1.0Na disappears, and the β loss peak of XLPE is broadened, although it does not move; this is because the

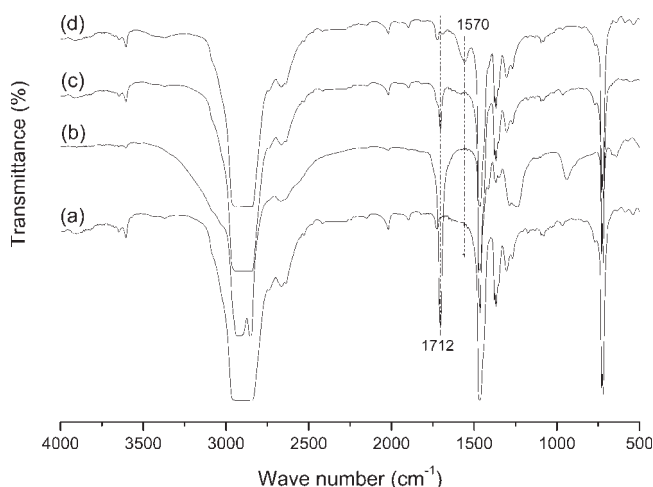


Figure 2 FTIR spectra of (a) XLPE, (b) EAA, (c) an XLPE/EAA (100/5.0) blend, and (d) an XLPE/EAA–1.0Na (100/5.0) blend.

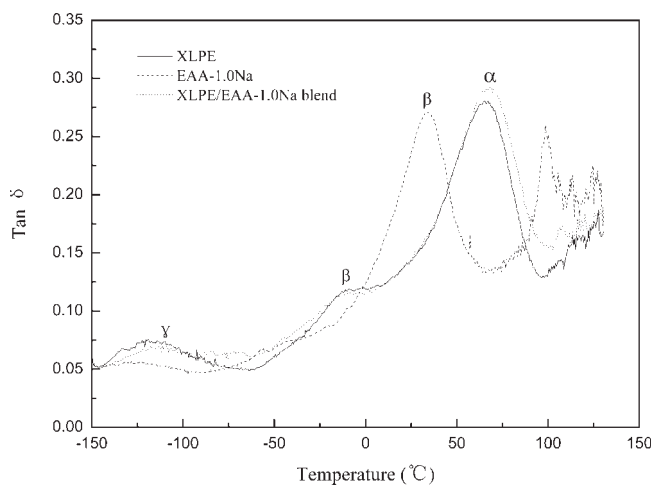


Figure 3 Temperature dependence of the loss tangent ($\tan \delta$) for XLPE, EAA-1.0Na, and an XLPE/EAA-1.0Na (100/5.0) blend.

amount of the EAA-1.0Na dispersing phase is much less than that of the XLPE continuous phase. Therefore, EAA-1.0Na is not enough to change the β loss peak of XLPE but just broadens it; this indicates that the XLPE/EAA-1.0Na blend system is partially compatible.^{21,22}

Hydrophilicity

Shown in Figures 4 and 5 are the effects of the content and neutralization degree of EAA-Na ionomers on the WAR of the samples. The WAR of the samples increases with an increase in the EAA-Na ionomer content and neutralization degree, and this is attributed to the stronger hydrophilicity of the $-\text{COONa}$ group versus that of the $-\text{COOH}$ group. The $-\text{COONa}$ groups in the samples are hydrophilic groups, and the $-\text{COONa}$ groups in the

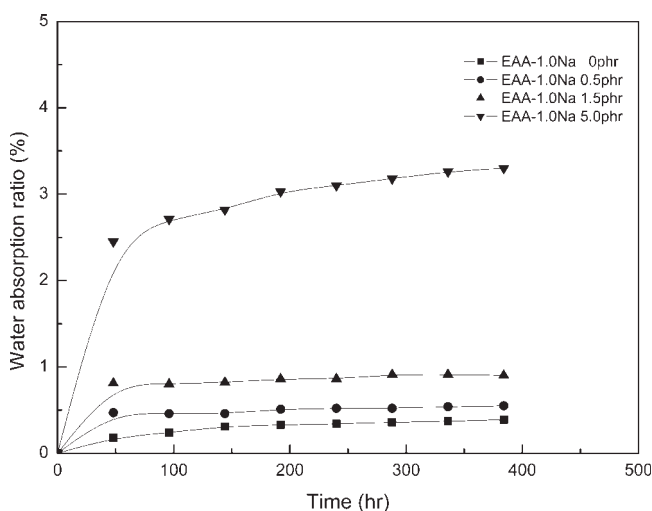


Figure 4 Dependence of the hydrophilicity on the EAA-1.0Na contents in the XLPE/EAA-1.0Na blends.

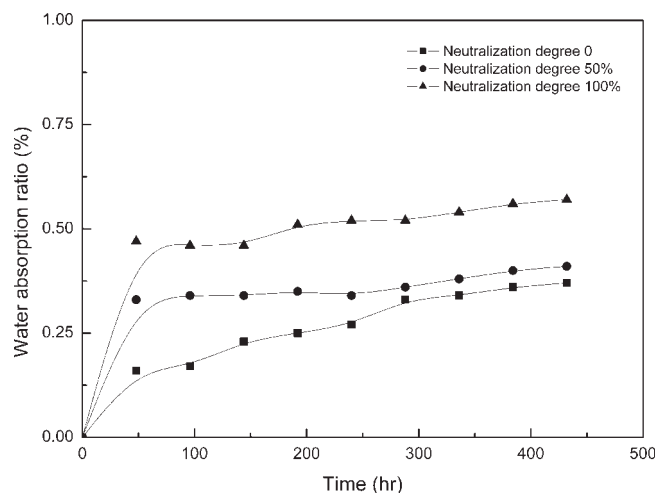


Figure 5 Dependence of the hydrophilicity on the neutralization degree of EAA-Na in the XLPE/EAA-Na (100/0.5) blends.

EAA-Na ionomer molecular chains partially ionize and become carboxylate anions ($-\text{COO}^-$) and mobile counterions (Na^+) when the samples are in contact with water.²⁶ Both the $-\text{COO}^-$ groups and the dissociated ions are capable of attracting water molecules, but pure XLPE almost does not absorb water. Thus, the hydrophilicity of the samples has been improved by the addition of the EAA-Na ionomers; moreover, the samples possess stronger hydrophilicity with an increase in the neutralization degree and content of the EAA-1.0Na ionomers.

Water treeing

The water-tree resistance of the samples has been evaluated with the WTL, and this is shown in Figures 6 and 7. The WTL values of the samples decrease with an

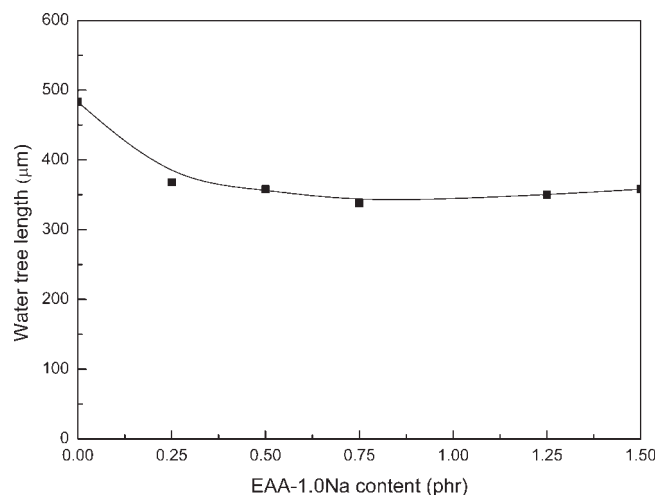


Figure 6 Dependence of the WTL on the EAA-1.0Na contents in the XLPE/EAA-1.0Na blends.

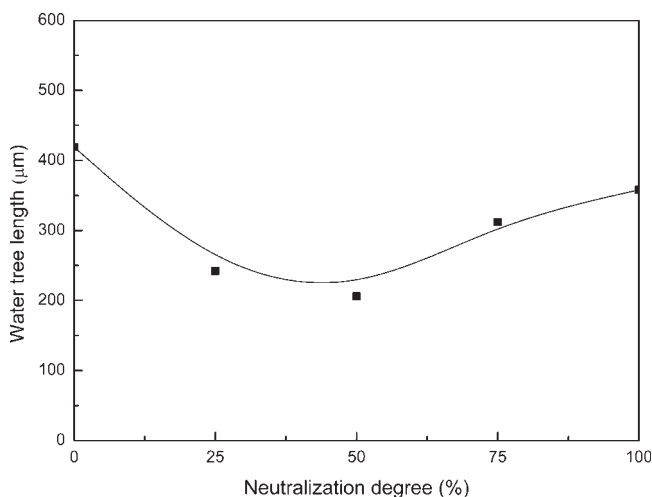


Figure 7 Dependence of the WTL on the neutralization degree of EAA-Na in the XLPE/EAA-Na (100/0.5) blends.

increase in the EAA-1.0Na content (Fig. 6). When the EAA-1.0Na content reaches 0.75 phr, the WTL of the samples decreases to 338 µm from 483 µm, and this indicates that the water-tree resistance of the samples has been improved. The results should be attributed to the improvement of the hydrophilicity for the samples.

According to the initiation and propagation mechanisms of water trees, water and Maxwell stresses are the most crucial factors for water-tree formation.^{11,27-29} XLPE is a hydrophobic, nonpolar polymer, so the electromechanical condensation of water occurs easily in electrically irregular points (voids, contaminants, etc.). Because of the inhomogeneous electric field of the defects in the polymer, water drops tend to move to the points of the highest stresses. Then, crazing of the polymer takes place under the action of the alternating Maxwell stresses and compressive stresses of the diffusion of water. Finally, with the growth of the craze, the crazelike structure of the polymer leads to the formation of treelike microchannels or tubes, which are water trees.^{11,30,31} Therefore, when the hydrophilic $-\text{COO}^-$ or $-\text{COOH}$ groups of the EAA-Na ionomers in the samples are in contact with water, the water molecules are prevented from diffusing in the samples because of the strong interaction between the $-\text{COO}^-$ or $-\text{COOH}$ groups and water molecules. On the other hand, the condensation of water in electrically irregular points is also prevented because the $-\text{COO}^-$ or $-\text{COOH}$ groups are scattered throughout the samples. Finally, the propagation of water trees is retarded by the addition of the EAA-Na ionomers in the samples.

Figure 7 shows that the WTL values of the samples decrease with an increase in the neutralization degree of the EAA-Na ionomers and reach the minimum at a neutralization degree of 50%, suggesting that the EAA-Na ionomers result in a better improvement of

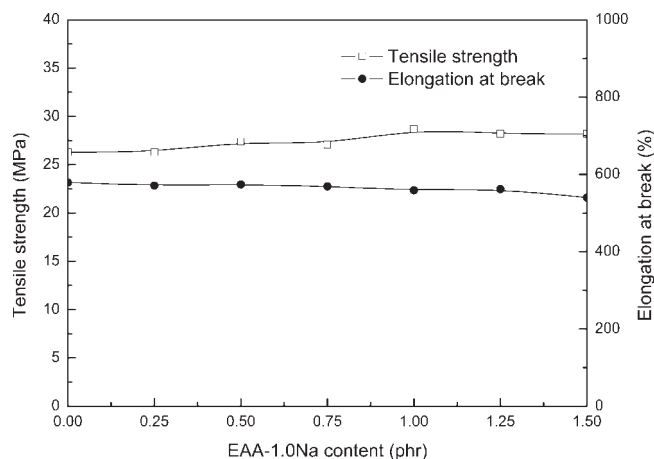


Figure 8 Dependence of the mechanical properties on the EAA-1.0Na contents in the XLPE/EAA-1.0Na blends.

the water-tree resistance for the samples versus EAA. The results should be attributed to the stronger hydrophilicity of the $-\text{COO}^-$ group of the EAA-Na ionomers versus that of the $-\text{COOH}$ group of EAA. However, when the neutralization degree of the EAA-Na ionomers exceeds 50%, the WTL values of the samples increase compared with that at the neutralization degree of 50%. This is possibly due to the fact that the size of the ionic aggregations of the EAA-Na ionomers increases with an increase in the neutralization degree, so the water concentration of the areas around the ionic aggregations increases gradually, and then the areas themselves become new defects.^{11,32,33}

Mechanical properties

Figures 8 and 9 show the effects of the EAA-1.0Na content and the neutralization degree of the EAA-Na ionomers on the mechanical properties of the

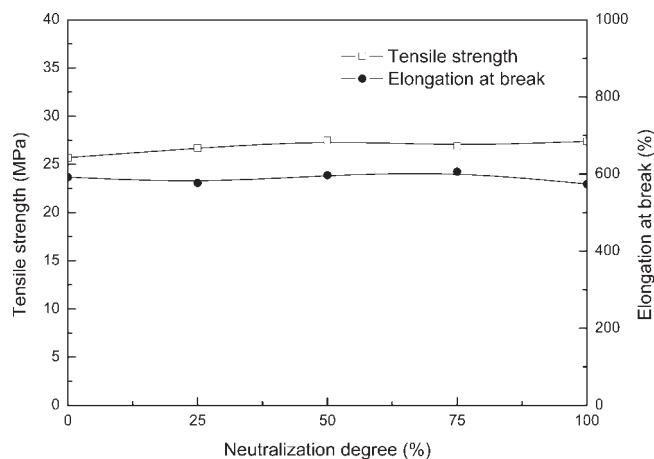


Figure 9 Dependence of the mechanical properties on the neutralization degree of EAA-Na in the XLPE/EAA-Na (100/0.5) blends.

samples. As shown in Figure 8, the tensile strength has only a slight enhancement with an increase in the EAA-1.0Na content in the samples, and this is mainly due to the physical crosslinks through the ionic aggregates of the EAA-1.0Na ionomers.³⁴ However, the EAA-1.0Na content has almost no effect on the elongation at break of the samples, and this suggests that the EAA-1.0Na content is still low and cannot influence the elongation at break. We can also see in Figure 9 that, although the increase in the neutralization degree of the ionomers can cause an enhancement of the tensile strength and a decrease in the elongation at break,³⁴ the neutralization degree of the EAA-Na ionomers nearly has no effect on the mechanical properties of the samples because of the small amounts of the EAA-Na ionomers. Hence, this reflects the fact that EAA-Na ionomers can be used to impart water-tree resistance to XLPE with excellent mechanical properties.

Dielectric properties

The dielectric properties of samples loaded with various amounts of EAA-Na ionomers and different neutralization degrees of EAA-Na ionomers were measured, and the results are shown in Figure 10(a-d). The EAA-Na ionomer content has only a slight influence on the dielectric properties of the samples [Fig. 10(a,b)], and this is probably due to the polar EAA-1.0Na ionomers. It is well known that a polar impurity has an effect on the dielectric properties of a matrix, including a decrease in the volume resistivity and an increase in the dissipation factor, dielectric strength, and dielectric constant.^{35,36} In addition, Figure 10(c,d) shows that the neutralization degree of the EAA-Na ionomers has almost no effect on the dielectric properties of the samples because of the small amounts of the EAA-Na ionomers. Therefore, the results reveal that the samples still present the typical characteristics of dielectric behavior for pure

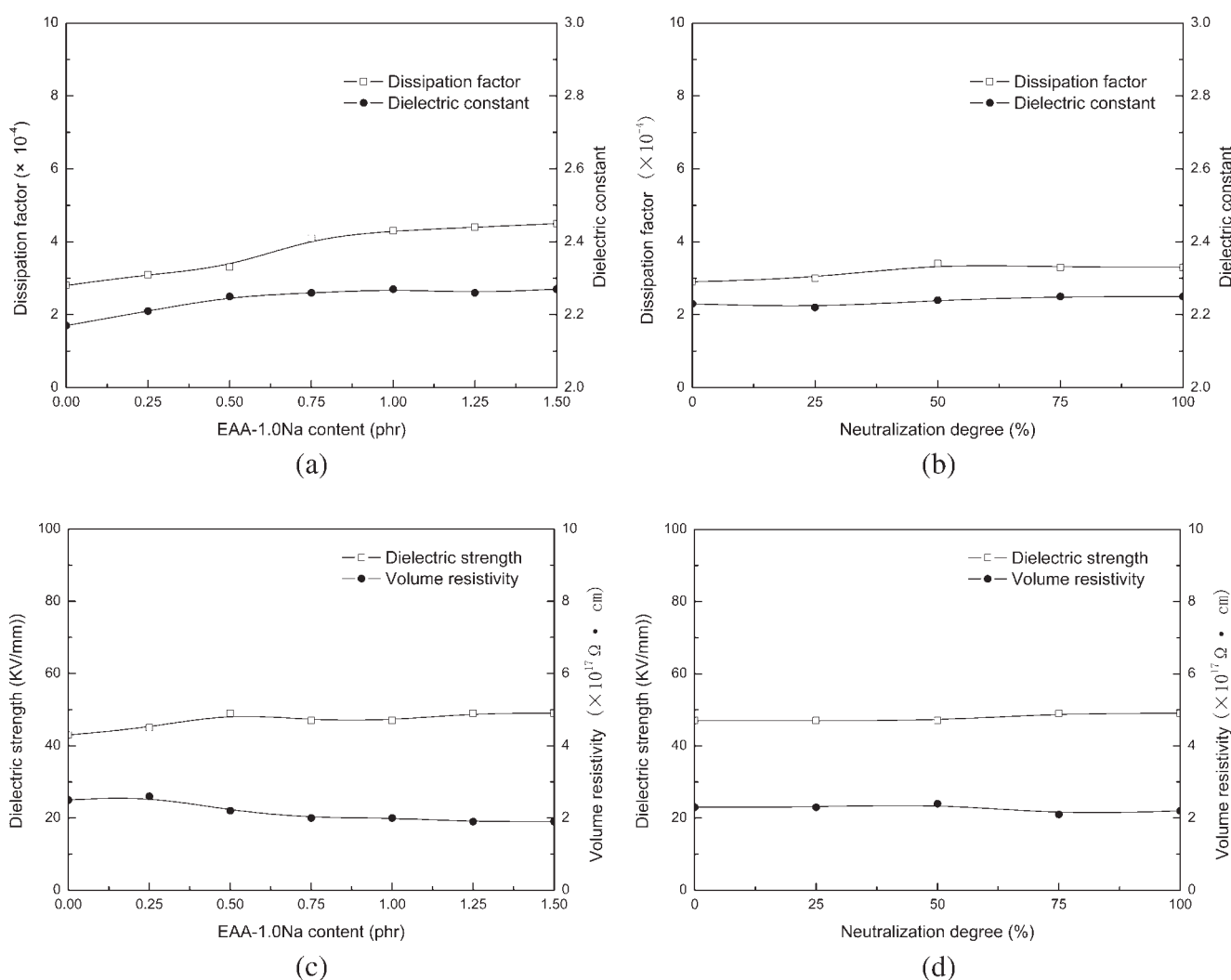


Figure 10 Dependence of the dielectric properties on (a,b) the contents and (c,d) the neutralization degree of EAA-Na ionomers in the XLPE/EAA-Na (100/0.5) blend.

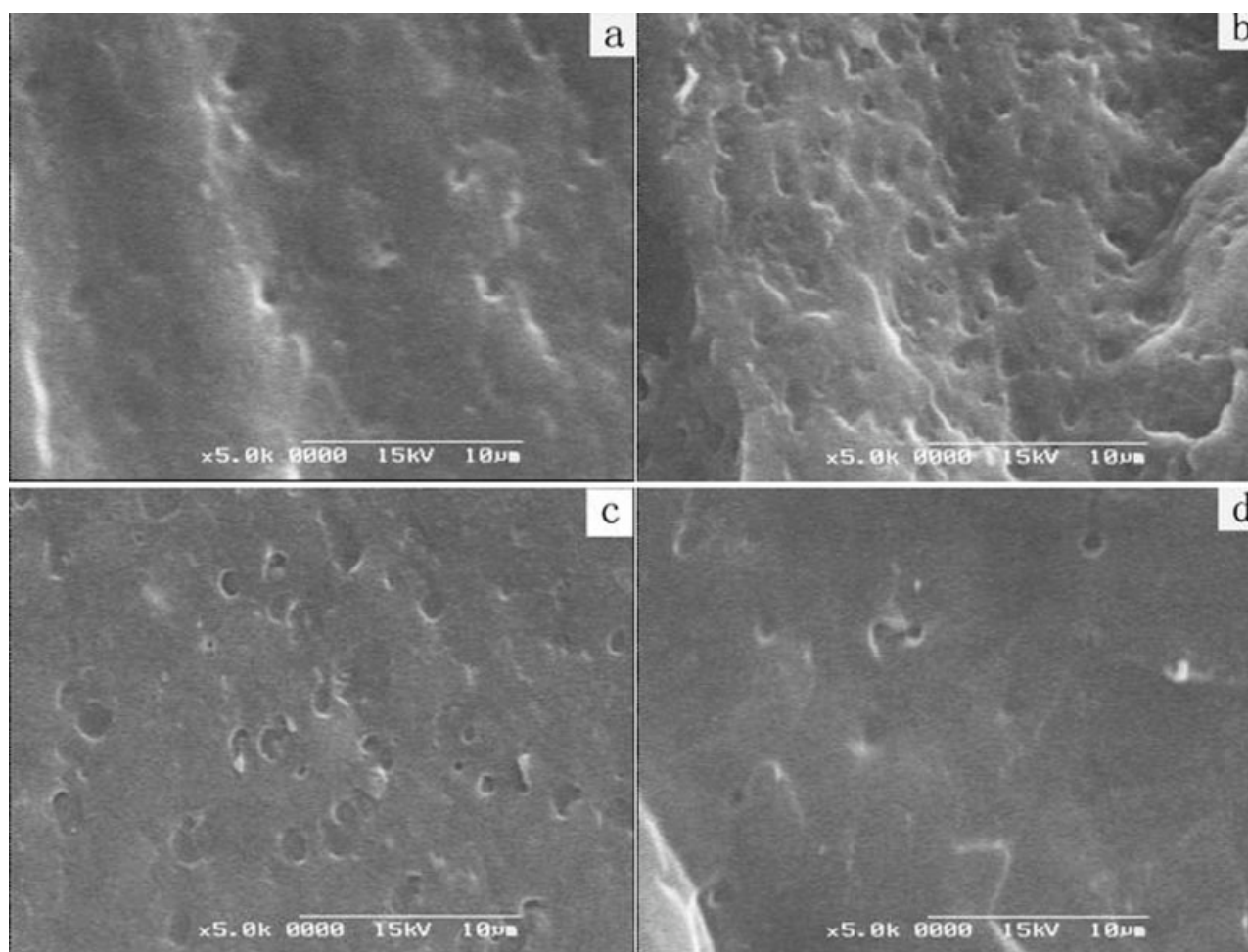


Figure 11 SEM micrographs of the samples: (a) an XLPE/EAA (100/5.0) blend, (b) an XLPE/EAA-0.5Na (100/5.0) blend, (c) an XLPE/EAA-1.0Na (100/5.0) blend, and (d) an XLPE/EAA-1.0Na (100/1.5) blend.

XLPE when the amount of the EAA-Na ionomers is small.

Morphology observation

From this analysis, the conclusion can be drawn that, despite the introduction of EAA-Na ionomers for the improvement of the water-tree resistance, the mechanical properties of XLPE still remain excellent. Nevertheless, the mechanical properties are related to the microstructure of the materials. Hence, Figure 11 presents the morphologies of fractured surfaces of XLPE/EAA-Na blends by scanning electron microscopy (SEM). The SEM micrographs reveal clearly that the EAA-Na ionomers disperse evenly in the XLPE matrix to form a dispersing phase, and XLPE still keeps a good continuous phase; this indicates that the XLPE/EAA-Na blends are partially compatible. Thus, the mechanical properties of the samples are not reduced; on the contrary, they are slightly enhanced because of the physical crosslinks through the ionic aggregates of the EAA-1.0Na ionomers.³⁴

In addition, the even dispersion of the EAA-Na ionomers in the XLPE matrix, which keeps the diffused water distributed evenly in the mass, should be one of the reasons that the water-tree resistance of the blends is improved.

CONCLUSIONS

The results have allowed the conclusion that EAA-Na ionomers employed as additives enhance greatly the water-tree resistance of XLPE.

At low EAA-1.0Na ionomer concentrations, such as 0.75 phr, the water-tree resistance of the samples can be improved. In particular, when the neutralization degree of EAA-Na ionomers is 50%, the water-tree resistance of blends will result in a better enhancement than that of the EAA-1.0Na ionomers. In addition, with the addition of EAA-Na ionomers, the dielectric and mechanical properties of the samples are not changed remarkably. The tensile strength has only a slight enhancement, which is attributed to the ionic crosslinking of EAA-Na ion-

omers, with an increase in the EAA–1.0Na ionomer content.

FTIR spectroscopy has revealed the *in situ* formation of EAA–Na ionomers through the neutralization of NaOH and EAA. Moreover, the XLPE/EAA–Na ionomer blend system is partially compatible, as indicated by DMA and morphology observations. In addition, the testing of the water absorption indicates that the hydrophilicity of the blends is improved with the addition of EAA–Na ionomers.

References

1. Ciuprina, F.; Teissedre, G.; Filippini, J. C. *Polymer* 2001, 42, 7841.
2. Gulmineand, J. V.; Akcelrud, L. *J Appl Polym Sci* 2004, 94, 222.
3. Sarathi, R.; Das, S.; Kumarand, C. R. A.; Velmurugan, R. *J Appl Polym Sci* 2004, 92, 2169.
4. Crine, J. P.; Jow, J. *IEEE Trans Dielectr Electr Insul* 2005, 12, 801.
5. Lee, J. H.; Suh, K. S.; Kim, S. J.; Jeong, D. W.; Han, M. K. *Proceeding of the 4th International Conference on Conduction and Breakdown in Solid Dielectrics*, IEEE, Sestri Levante, Italy, 1992; p 451.
6. Han, M. K.; Lee, J. H.; Suh, K. S.; Choi, Y. H. *Electr Insul Dielectr Phenom Annu Rep* 1992, 638.
7. Turbett, R. J. U.S. Pat. 4,440,671 (1984).
8. Topcik, B. U.S. Pat. 4,812,505 (1989).
9. Sckii, Y.; Momose, N.; Takatori, K.; Tadao, G. *Conference on Electrical Insulation and Dielectric Phenomena*, IEEE, Victoria, Canada, 2000; p 347.
10. Sckii, Y.; Momose, N.; Takatori, K.; Tadao, G. *Proceeding of the 7th International Conference on Properties and Applications of Dielectric Materials*, IEEE, Victoria, Canada, 2003; p 927.
11. Peruzzotti, F.; Castellani, L.; Dacal, L. J.; Garcia, V. U.S. Pat. 6,521,695 (2003).
12. Kazuhiro, K.; Horii, F. *J Polym Sci Part B: Polym Phys* 2002, 40, 1142.
13. Willis, J. M.; Caldas, V.; Favis, B. D. *J Mater Sci* 1991, 26, 4742.
14. Mascia, L.; Valenza, A. *Adv Polym Technol* 1995, 14, 327.
15. Mascia, L.; Bellahdeh, F. *Adv Polym Technol* 1994, 13, 99.
16. Ren, W. T.; Peng, Z. L.; Zhang, Y.; Zhang, Y. X. *J Appl Polym Sci* 2004, 92, 1804.
17. Norihornme, B.; Crine, J. P.; Bulinsky, A. T.; Densley, R. J. *Electr Insul Dielectr Phenom Annu Rep* 1991, 386.
18. Maria, J. C.; Jose, L. A.; Amelia, L. *Macromol Chem Phys* 2005, 206, 680.
19. Chen, S. C.; Chiu, H. T.; Ye, C. P. *J Appl Polym Sci* 2002, 86, 3740.
20. Niaounkis, M.; Kontou, E. *J Polym Sci Part B: Polym Phys* 2005, 43, 1712.
21. He, P. X.; Huang, H.; Xiao, W. D.; Huang, S. Q.; Cheng, S. Y. *J Appl Polym Sci* 1997, 64, 2535.
22. Xu, S. Q.; Tan, T.; Huang, B. T.; Yin, C. M. *Acta Chim Sinica* 1998, 56, 445.
23. Ward, I. M.; Sweeney, J. *An Introduction to the Mechanical Properties of Solid Polymers*; Wiley: West Sussex, England, 2004.
24. Cerradaa, M. L.; Benaventea, R.; Peñab, B.; Péreza, E. *Polymer* 2000, 41, 5957.
25. Hitorhi, T.; Hisaaki, H.; Eieaku, H.; Shoichi, K.; Kenji, T.; Shiniichi, Y. *Macromolecules* 1993, 26, 752.
26. Ren, W. T.; Zhang, Y.; Peng, Z. L.; Zhang, Y. X. *Polym Test* 2004, 23, 809.
27. Filippini, J. C. *Proceedings of the IEEE International Symposium on Electrical Insulation*, IEEE, Toronto, Canada, 1990; p 183.
28. Ross, R. *IEEE Trans Dielectr Electr Insul* 1998, 5, 660.
29. Fan, Z. H.; Yoshimura, N. *IEEE Trans Dielectr Electr Insul* 1996, 3, 849.
30. Takahashi, T.; Niwa, T.; Suzuki, J. *Proceedings of the Properties and Applications of Dielectric Materials, Second International Conference on Properties and Applications*, IEEE, Beijing, China, 1988; p 486.
31. Wael, M. A. H.; Ahmed, A. H. E. *Conference Record of the IEEE International Symposium on Electrical Insulation*, IEEE, Pittsburgh, PA, 1994; p 253.
32. Hitorhi, T.; Hisaaki, H.; Eieaku, H.; Shoichi, K.; Kenji, T.; Shiniichi, Y. *Macromolecules* 2000, 33, 3818.
33. Tsujita, Y.; Yasuda, M.; Takei, M.; Kinoshita, T.; Takizawa, A.; Yoshimizu, H. *Macromolecules* 2001, 34, 2220.
34. Swapan, K. G.; Bhattacharya, A. K.; De, P. P.; Khastgir, D.; De, S. K. *Rubber Chem Technol* 2001, 74, 883.
35. Gu, Z. J.; Wang, S. T. *Polymer Electrics and Magnetism*; Shanghai Jiao Tong University Press: Shanghai, 1990.
36. He, M. J.; Chen, W. X.; Dong, X. X. *Polymer Physics*; Fudan University Press: Shanghai, 1991.