Proceedings of the 17th
IEEE International Conference on Nanotechnology
Pittsburgh, USA, July 25-28, 2017

Electrical Breakdown of Polymer Nanocomposites Modulated by Space Charges

Daomin Min, Chenyu Yan, Weiwang Wang, Dongri Xie, Michel Fréchette and Shengtao Li

***Abstract*— A method is proposed to calculate the density of deep traps formed in interaction zones based on mesoscopic structure and double electric layer of polymer nanocomposites. Then a space charge modulated breakdown model is utilized to investigate electrical breakdown property and its relation with deep traps in interaction zones. It is found that deep traps formed around independent interaction zones suppress the accumulation of space charges and the distortion of electric field, leading to the improvement of breakdown strength.**

I. Introduction

Polymer nanocomposites have higher electrical breakdown strength than polymer matrices and their microcomposites, which is beneficial for the development of high voltage dc power equipment [1-3]. It has been widely accepted that the enhancement in breakdown strength is originated from interaction zones or interfacial regions formed around nanoparticles [1-3]. Interaction zones can change chain conformation, aggregation structure, morphology of polymer matrices, causing the modifications of trap properties, carrier mobility, and charge injection, and so on. These modifications lead to the suppression of space charges accumulation, the reduction of volume resistivity, and the enhancement of electrical breakdown strength in polymer nanocomposites at low filler contents [1-3].

The dispersion of nanoparticles and the mesoscopic structure of interaction zones determine dielectric properties of polymer nanocomposites. The breakdown field increases firstly and then decreases with an increase in filler content, and it may be saturated at high filler contents [1]. Electrical breakdown strength, volume resistivity and space charge accumulation all depends on filler content, and electrical breakdown strength has positive correlations with volume resistivity and space charge accumulation. It means that the variation in charge transport properties produced by interaction zones can change the accumulation of space charges, which may modulate electrical breakdown strength of polymer nanocomposites.

*Resrach supported by the National Basic Research Program of China under Project with No. 2015CB251003, National Natural Science Foundation of China under Projects with Nos. 51507124 and 51337008, and State Key Lab. of Power System of Tsinghua University (SKLD16KZ04).

D. Min, C. Yan, W. Wang, D. Xie, and S. Li are with the State Key Laboratory of Electrical Insulation and Power Equipment, Xi'an Jiaotong University, Xi'an, Shaanxi, 710049, China (corresponding author to provide phone: +86-29-82663781; fax:+86-29-82668567; e-mail: forrestmin@mail.xjtu.edu.cn).

Michel Fréchette is with Hydro-Hydro-Québec's research institute, 1800, Blv. Lionel-Boulet, Varennes, Canada.

II. Deep Traps and Electrical Breakdown

The density of deep traps may be determined by inter-particle distance, the thickness of bonded and transitional regions, binding strength in the bonded layer, and overlapping of Gouy-Chapman diffuse layers [2, 4], as shown in Figure 1. It is assumed that spherical inorganic nanoparticles with a density of ρ_n and a weight of m_n are incorporated into a semi-crystalline polymer matrix with a density of ρ_p and a weight of m_p to prepare a polymer nanocomposite. The radius of nanoparticles is r_n. They are assumed to be homogeneously dispersed in the polymer matrix with interparticle distance from surface to surface of l_n and are mainly located in amorphous regions [5]. The crystallinity is supposed to be χ_c for polymer nanocomposites. The weight fraction of nanofillers f_m is equal to $m_n/(m_n+m_p)$, while the volume fraction f_v is $m_n\rho_p/(m_n\rho_p+m_p\rho_n)$. Then, the relation between volume fraction f_v and weight fraction f_m is $1/f_v=1/f_m\cdot\rho_n/\rho_p[1-f_m(\rho_p/\rho_n)]$ [3, 6]. The volume fraction of nanoparticles f_v can be expressed by $f_v=(1-\chi_c)\cdot4\pi/3\cdot[r_n/(2r_n+l_n)]^3$ [3, 6]. Then, we can obtain the relation among interparticle distance, weight fraction, the densities of nanoparticles and polymer matrix, and the diameter of nanoparticles [3, 6].

$$l_n=\left[\left[\frac{4\pi(1-\chi_c)}{3}\frac{1}{f_m}\frac{\rho_n}{\rho_p}\left[1-f_m\left(1-\frac{\rho_p}{\rho_n}\right)\right]\right]^{\frac{1}{3}}-2\right]r_n \quad (1)$$

The number density of nanoparticles n_n is inversely proportional to the cubic volume with a length of (l_n+l_{ip}).

$$n_n=(1-\chi_c)\frac{1}{(2r_n+l_n)^3} \quad (2)$$

It is suggested that deep traps are formed in the bonded and transitional regions [1, 3]. Consequently, the density of deep traps is influenced just by volume fraction of bonded and transitional regions. Assumed average distance between two deep traps in bonded and transitional regions to be λ_{DT}, the number of deep traps ξ generated in an interaction zone can be calculated by the following equation:

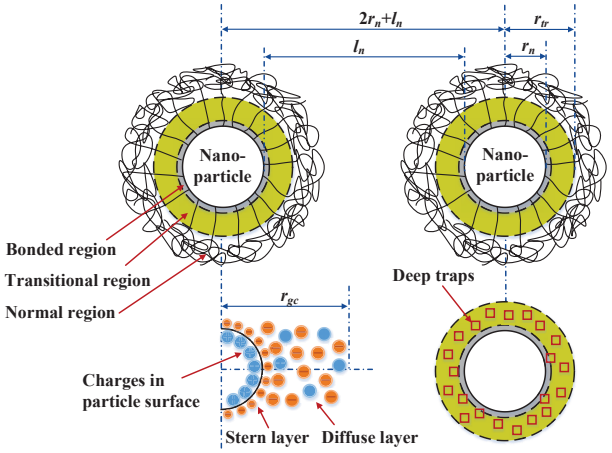

Figure 1. Schematic of mesoscopic structure of and charge distribution in interaction zone. Deep traps are mainly formed in bonded and transitional regions.

$$\xi = \frac{4\pi}{3} \cdot \frac{\left(r_{br}^{\ 3} - r_n^{\ 3}\right)}{\lambda_{DT}^{\ 3}}, \tag{3}$$

where r_{br} is the radius of transitional region.

It is possible for Gouy-Chapman layers to overlap with each other at high filler contents. The overlapping probability may increase dramatically with an increase in filler content. Accordingly, the possibility for independent interaction zones decreases strikingly.

$$P_i = \left[1+\left(\frac{32}{3}\pi n_n \frac{r_{gc}^{\ 6}}{r_n^{\ 3}}\right)^{\beta}\right] \exp\left[-\left(\frac{32}{3}\pi n_n \frac{r_{gc}^{\ 6}}{r_n^{\ 3}}\right)^{\beta}\right] \tag{4}$$

where r_{gc} is the radius of the Gouy-Chapman layer, while β is an exponent for the stretched exponential function.

The density of deep traps in interfacial regions also depends on the type of nanoparticles, surface treatment, charge distribution in Gouy-Chapman diffuse layer, binding strength in the first layer, and so on. Therefore, the density of deep traps can be written as $N_{DT}=\xi n_n P_i$.

Then a space charge modulated electrical breakdown model [7] is utilized to simulate the breakdown strength of LDPE/Al_2O_3 nanocomposites at various filler contents. Electrons and holes are injected into the material from cathode and anode, respectively, by Schottky thermionic emission [8, 9]. Charge injection into and transport in an insulating material are governed by a set of self-consistent equations, including equations of charge injection, charge continuity, and transport, and Poisson's equation [8, 10-15].

LDPE/Al_2O_3 nanocomposites with various filler contents ranging from 0 wt% to 2 wt% were used for dc breakdown simulations. The nanocomposite films were 200 μm in thickness. In the dc breakdown simulations LDPE/Al_2O_3 nanocomposite films were discretized into 500 elements and each element was 0.4 μm in length. The computation time step Δt was 1 ms. Deep traps in polymer nanocomposites consist of

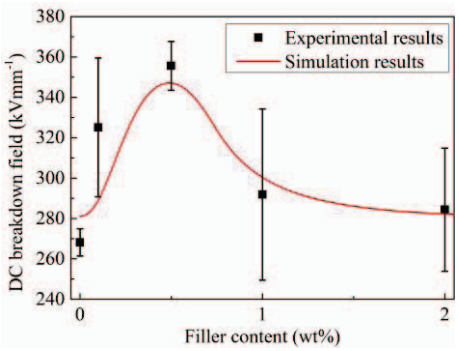

Figure 2. DC breakdown field as a function of filler content of polymer nanocomposites.

those in interfacial regions with a density of N_{Tn} and in the polymer matrix with a density of N_{Tp}. It is assumed that deep traps in interfacial regions and those in the polymer matrix are located in the same energy level. Consequently, the density of deep traps in polymer nanocomposites N_T equals to the summation of N_{Tn} and N_{Tp}, namely, $N_T = N_{Tn}+N_{Tp}$.

A ramp voltage with a rising rate of 500 Vs^{-1} was applied on the nanocomposite films. An intrinsic breakdown strength was set for LDPE nanocomposites, which means that dc breakdown simulations stop when the internal local electric field reaches 365 kV/mm. Then the breakdown field was calculated from the applied voltage divided by sample thickness. Figure 2 shows a comparison between simulation and experimental results of dc breakdown field as a function of filler content in a range of 0 wt% to 2 wt% in LDPE/Al_2O_3 nanocomposites. The simulation results are in good agreement with experiments. The dc breakdown field obtained by simulation increases firstly and then decreases with an increase in filler content. The breakdown field reaches its maximum at a filler content of about 0.5 wt% and gets saturated above 2 wt%.

I. Conclusion

The density of deep traps formed in polymer nanocomposites was calculated by considering the mesoscopic structure of and double electric layer around interaction zones. The estimation method of deep trap density was involved into a space charge modulated breakdown model to calculate breakdown strength of polymer nanocomposites. The relation between breakdown strength and filler content was obtained and the simulation results were in good agreement with experiments. Deep traps formed around independent interaction zones at relatively low filler content suppress the accumulation of space charges and the distortion of electric field, leading to the improvement of breakdown strength. The proposed model is advantageous to explore the influencing mechanism of interaction zone on electrical property of polymer nanocomposites.

REFERENCES

[1] S. Li, G. Yin, G. Chen, J. Li, S. Bai, L. Zhong, Y. Zhang and Q. Q. Lei, "Short-term breakdown and long-term failure in nanodielectrics: A review", IEEE Trans. Dielectr. Electr. Insul., vol. 17, no. 5, pp. 1523-1535, 2010.

[2] T. Tanaka, "Dielectric nanocomposites with insulating properties", IEEE Trans. Dielectr. Electr. Insul., vol. 12, no. 5, pp. 914-928, 2005.

[3] T. Tanaka, M. Kozako, N. Fuse and Y. Ohki, "Proposal of a multi-core model for polymer nanocomposite dielectrics", IEEE Trans. Dielectr. Electr. Insul., vol. 12, no. 4, pp. 669-681, 2005.

[4] J. K. Nelson, *Dielectric polymer nanocomposites*, Springer: Spring, New York, USA, pp. 1-285, 2010.

[5] T. J. Lewis, "Charge transport in polyethylene nano dielectrics", IEEE Trans. Dielectr. Electr. Insul., vol. 21, no. 2, pp. 497-502, 2014.

[6] P. O. Henk, T. W. Kortsen and T. Kvarts, "Increasing the electrical discharge endurance of acid anhydride cured DGEBA epoxy resin by dispersion of nanoparticle silica", High Perform. Polym., vol. 11, no. 3, pp. 281-296, 1999.

[7] S. T. Li, Y. W. Zhu, D. M. Min and G. Chen, "Space charge modulated electrical breakdown", Sci. Rep., vol. 6, no. 32588, pp. 1-4, 2016.

[8] S. Le Roy, P. Segur, G. Teyssedre and C. Laurent, "Description of bipolar charge transport in polyethylene using a fluid model with a constant mobility: Model prediction", J. Phys. D: Appl. Phys., vol. 37, no. 2, pp. 298-305, 2004.

[9] C. Laurent, G. Teyssedre, S. Le Roy and F. Baudoin, "Charge dynamics and its energetic features in polymeric materials", IEEE Trans. Dielectr. Electr. Insul., vol. 20, no. 2, pp. 357-381, 2013.

[10] J. M. Alison and R. M. Hill, "A model for bipolar charge transport trapping and recombination in degassed crosslinked polyethylene", J. Phys. D: Appl. Phys., vol. 27, no. 6, pp. 1291-1299, 1994.

[11] F. Boufayed, G. Teyssedre, C. Laurent, S. Le Roy, L. A. Dissado, P. Segur and G. C. Montanari, "Models of bipolar charge transport in polyethylene", J. Appl. Phys., vol. 100, no. 10, pp. 104105(10 pp), 2006.

[12] S. Le Roy, G. Teyssedre, C. Laurent, G. C. Montanari and F. Palmieri, "Description of charge transport in polyethylene using a fluid model with a constant mobility: Fitting model and experiments", J. Phys. D: Appl. Phys., vol. 39, no. 7, pp. 1427-1436, 2006.

[13] G. Chen, "A new model for surface potential decay of corona-charged polymers", J. Phys. D: Appl. Phys., vol. 43, no. 5, pp. 055405 (7pp), 2010.

[14] D. M. Min, "*Investigation into charge trapping transport properties and mechanisms in polymeric insulating materials*", Ph.D. dissertation, Xi'an Jiaotong University, Xi'an, 2013.

[15] D. M. Min, W. W. Wang and S. T. Li, "Numerical analysis of space charge accumulation and conduction properties in LDPE nanodielectrics", IEEE Trans. Dielectr. Electr. Insul., vol. 22, no. 3, pp. 1483-1491, 2015.