СЕКЦИЯ 3. МОДИФИКАЦИЯ СВОЙСТВ МАТЕРИАЛОВ SECTION 3. MODIFICATION OF MATERIAL PROPERTIES

EFFECT OF ELECTRON BEAM IRRADIATION ON SURFACE DIELECTRIC PROPERTIES OF POLYMERIC MATERIALS

Chengyan Ren^{1), 2)}, Duo Hu^{1), 2)}, Chuansheng Zhang²⁾, Tao Shao^{1), 2)}, Ping Yan^{1), 2)} ¹⁾Key Laboratory of Power Electronics and Electric Drive, Institute of Electrical Engineering, Chinese Academy of Sciences, 100190 Beijing, China ²⁾University of Chinese Academy of Sciences, 100049 Beijing, China rcy@mail.iee.ac.cn, huduo@mail.iee.ac.cn, zhangchuansheng18@mails.ucas.ac.cn, st@mail.iee.ac.cn, pingyan@mail.iee.ac.cn

The polymeric materials could degrade due to the electron irradiation, photo-irradiation and heat effect following with the surface flashover in vacuum. On the other hand, the surface insulating performance could be improved by electron irradiation. In order to study the effect of electron irradiation on the insulating materials, the surface parameters were measured and the flashover experiment was performed in vacuum after electron beam irradiation. First, several typical polymeric samples, such as polytetrafluoroethylene (PTFE), polymethylmethacrylate (PMMA) and polyamide (PA6), were exposed by different energy electron beam from 10 keV to 30 keV and in different radiation times. Then the surface parameters, such as surface topography, chemical group and surface roughness, were measured for insulating samples in different processing conditions. Since the surface trap distribution is closely related to the microscopic structure and defects, the trap parameters of PA6 samples were deduced based on the surface potential data. In this measurement, the polymeric surface was charged by needle-to-plane corona discharge, and then the surface potential was measured by Kelvin electrostatic probe. The surface flashover experiment in vacuum was performed using finger-type plane-electrodes, and the relationship between electron beam energy, irradiation time, and flashover voltage was analyzed. The experiment results indicate that the surface trap energy level is deeper and the trap density is greater after electron irradiation, and the surface flashover voltages for several materials are also improved to different degrees. Combined the macroscopic dielectric parameters with the microscopic one, the effect of electron irradiation on surface insulating property was analyzed.

Keywords: electron irradiation; surface properties; trap; vacuum flashover; polymer.

Introduction

The changes in dielectric insulating properties affect reliable operation and service life of electrical equipments in an irradiation environment. The research fields of dielectric properties after irradiation include irradiation crosslinking [1], space environment [2], material aging [3], and so on. In the case of spacecraft, surface or deep discharge phenomena are caused by charging of the dielectric in the space environment, resulting in abnormal operation of the spacecraft, such as the internal charge accumulation of the high-power cable insulation, the electromagnetic interference caused by the electrostatic discharge to the electronic device, the flashover across the insulation between the conductive rings of the driving mechanism, and the surface discharge between the battery cells [4], etc. The direct internal damage events of spacecraft are rare since the high energy (MeV or higher) electron and ion density is low in the space environment [5]. Therefore, it is important to pay attention to the surface insulating properties of the dielectric after irradiation.

It is widely concerned about the electrical properties of insulating dielectric after irradiation. Hegeler et al. reported that the effects of different ultraviolet irradiation positions on vacuum flashover with polycarbonate and borosilicate glass. They found that the surface withstand voltage was reduced when irradiated on the electrode position; however, the flashover voltage had a 10% increase when irradiated on the dielectric surface [6]. Fujii et al. found that the flashover voltage of lead-containing glass decreased after 100 eV low-energy electron beam irradiation, and it became more obvious with increasing electrode gap [7]. Liu et al. observed that under the irradiation of low energy electron beam (keV), the vacuum flashover voltage increased first and then decreased with the

increase of electron beam energy, but it was still higher than the unirradiated sample [8]. Li et al. reported that flashover voltage was jointly affected by electron beam energy and incident angle on sample surface [9].

In addition, more researches have focused on changes in the intrinsic properties of dielectrics. Lin et al. reported that epoxy micro-composite was treated by electron beam irradiation in vacuum environment, and the shallow trap density decreased, the deep trap density and energy level increased with increase of the irradiation energy [10]. Zheng et al. found that the ability to resist radiation and store charge is related to material type during high energy electron beam irradiation (MeV). They also showed that the relative dielectric constant of materials is mainly influenced by the number of molecules per unit volume and the molecular polarizability [11, 12]. Thus, the existing research mostly focuses on the effects of irradiation energy, angle, and irradiation position. Some experimental results are inconsistent. Therefore, it is necessary to study the changes of the surface insulation properties of different materials after irradiation and at the same time take into account the effects of different irradiation times.

In this paper, several typical polymers were used, such as polytetrafluoroethylene (PTFE), polymethyl methacrylate (PMMA), and polyamide (PA6), which are widely used in electrical equipments. The dielectric surface was firstly irradiated with different energy and different time electron beams. Then the surface parameters of the samples, such as surface morphology, surface roughness and trap distribution, were measured. At last the surface flashover experiment was developed to obtain the surface insulating properties of the irradiated samples in vacuum.

¹³⁻я Международная конференция «Взаимодействие излучений с твердым телом», 30 сентября - 3 октября 2019 г., Минск, Беларусь 13th International Conference "Interaction of Radiation with Solids", September 30 - October 3, 2019, Minsk, Belarus

Experimental Setup

The samples used in the experiment were PTFE, PMMA and PA6, with a size of $50 \times 50 \times 2$ mm, dried for 5 hours at 70 °C in a vacuum drying box. The electron gun in scanning electron microscope (JSM-6360LV) was used to irradiate the surface of the samples. The irradiating conditions were shown in Table 1.

Tab. 1. Treatment of samples by electron beam irradiation

Energy (keV)	10	15	20	30
l ime (min)				
10				✓
20				\checkmark
40				\checkmark
60	✓	\checkmark	\checkmark	\checkmark

In the vacuum flashover experiment, finger-shaped electrodes are used to ensure that the flashover occurs in a certain area. A lower voltage is firstly applied on the sample surface. If there is no flashover within 20 s, 0.5 kV will be increased. This process of gradually increasing voltage will be repeated until flashover occurs. Five flashover experiments were performed on each sample. The vacuum is maintained at $(2~6)\times10^{-4}$ Pa during the whole flashover experiment. The schematic diagram of vacuum flashover experiment is shown in Figure 1.



Fig. 1. Schematic diagram of flashover experimental system

Experimental Results

The surface morphology of PTFE after irradiation under different conditions was observed by SEM, which is shown in Figure 2. It can be seen that some micropores begin to appear on the surface of the material after irradiation with certain conditions. When irradiation energy is 10 keV, the sample surface is partly layered and fewer micropores are also observed, but the above phenomenon is not increased when the energy is increased to 15 keV. When the energy is increased to 20 keV, many small and dense pores appear on the surface of the sample except the above micropores and folds. As the energy is increased to 30 keV, the pore size of the sample surface also increases. A similar situation exists in PMMA, but no micropores appear on the surface of PA6.

Since the SEM observation range is $21 \times 17 \ \mu m^2$, the irradiated area of the sample is about tens of mm², and the minimum spacing of the electrodes is mm in the flashover experiment, we measured the surface roughness *Ra* by roughness meter (TR210). The result is shown in Figure 3. The increase of *Ra* for PTFE is relatively small compared to non-irradiation sample, and the increment is maximum at 20 keV. The surface

roughness of PMMA and PA6 also increases, but the increment is smaller than PTFE.



Fig. 2. Morphology of samples radiated by electron beam with different conditions



Fig. 3. Surface roughness of samples after irradiation

Figure 4 shows the infrared spectrum of the samples after irradiation with electron beam energy of 30 keV for different times. The characteristic peaks of PTFE, PMMA and PA6 in different irradiation times are basically coincident. That is, electron beam irradiation under vacuum does not cause new chemical groups

¹³⁻я Международная конференция «Взаимодействие излучений с твердым телом», 30 сентября - 3 октября 2019 г., Минск, Беларусь 13th International Conference "Interaction of Radiation with Solids", September 30 - October 3, 2019, Minsk, Belarus

on the surface of polymeric materials due to chemical reaction.



Fig. 4. Infrared spectrum of samples after irradiation

The surface trap of insulating material is related to the accumulation and dissipation of surface charge, which affects the local electric field. Its influence on vacuum flashover has attracted extensive attention [13]. The surface trap parameters of PA6 were calculated by Isothermal Surface Potential Decay (ISPD). The trap parameters can be calculated as follows: [13, 14]:

$$E_T = k_B T \ln \left(v_{ATE} t \right) \tag{1}$$

$$Q_{s}(t) = t \frac{\varepsilon_{0}\varepsilon_{r}}{q_{e}L} \frac{d\phi_{s}(t)}{dt}$$
(2)

Where E_T is the trap level, Q_s is the trap charge density, k_B is the Boltzmann constant, T is the sample temperature, v_{ATE} is the escape frequency of trap charge (4.17x10¹³ s⁻¹), *L* is the sample thickness, $\Phi s(t)$ is the surface potential value.

When the irradiation energy is 30 keV, there is a positive correlation between the irradiation time and the trap energy level, as shown in Fig. 5. Meanwhile there is also a positive correlation between the irradiation energy and the trap level.



Fig. 5. Trap parameter of PA6 after irradiation

The vacuum flashover experiments were carried out on the samples irradiated by different electron beams to study the surface insulating property. Figure 6 shows the surface flashover voltage of several materials in vacuum. In general, the flashover voltage of several materials is on the up trend after diffrent electron beam irradiations. There are some differences in the change trend for different materials.



Fig. 6. Flashover voltage of samples in different conditions of irradiation

In Figure 6, the flashover voltage of PA6 increases with the irradiation time and energy, but the surface morphology does not change significantly. The surface trap parameter results show that the trap density and

¹³⁻я Международная конференция «Взаимодействие излучений с твердым телом», 30 сентября - 3 октября 2019 г., Минск, Беларусь 13th International Conference "Interaction of Radiation with Solids", September 30 - October 3, 2019, Minsk, Belarus

energy level increase after electron beam irradiation, which is similar to the results of ducument [10].

Conclusions

The typical polymer samples, PTFE, PMMA, and PA6, were irradiated under different conditions of electron beams. Some surface parameters were measured and analyzed. The micropores appear on the surface of PTFE and PMMA, but the chemical groups have no obvious changes for three materilas. The surface roughness has minor increase for three materilas.

The surface trap level is deeper and the trap density is higher after electron beam irradiation, and the surface flashover voltage of several materials also increases approximately. The surface insulating performance is improved mainly by the trap distribution change after electron beam irradiation.

References

- 1. Ma Y., Zhang C., Zhang Z., Xiao R., Song Y., Yang Y., et al. Electron beam irradiation dose effects on crossinglinking degree of PAN/PEO. *Nuclear Physics Review* 2014; 31(1): 86-91.
- 2. Li S., Nie Y., Min D., Pan S. Research process on vacuum surface flashover of solid dielectrics. *Transactions of China Electrotechnical Society* 2017; 32(8): 1-9.
- 3. Zhang Y., Gong Z., Wang L., Wang L., Yun H., Zheng Z. Thermal and irradiation aging detection technology of cables based on dielectric properties. *Insulating Materials* 2017; 50(9): 62-67.
- 4. Li S., Li G. Dielectric charging research status. *Chinese Science Bulletin* 2017; 62(10): 990-1003.

- 5. Shu T. L. Fundamentals of spacecraft charging: spacecraft interactions with space plasmas. New Jersey: Princeton University Press; 2012. 7 p.
- Hegeler F., Krompholz H., Hatfield I.L., Kristiansen M. Dielectric surface flashover in a simulated low earth orbit environment. *IEEE transactions on plasma science* 1997; 25(2): 300-305.
- Fujii H., Hiro S. Surface flashover of dielectrics during lowenergy electron irradiation in vacuum. Proceedings of the 18th International Symposium on Discharges and Electrical Insulation in Vacuum; 1998. p. 808-811.
- Liu C., Zheng Y., Yang P., Zheng X., Li Y., Bian E. The DC surface flashover performance research of polyimide under low-energy electron irradiation environment. *IEEE Transactions on Plasma Science* 2016; 44(1): 85-92.
- 9. Li S., Pan S., Li G., Min D., Wang W., Li S. Influence of electron beam irradiation on DC surface flashover of polyimide in vacuum. *IEEE Transactions on Dielectrics and Electrical Insulation* 2017; 24(2): 1288-1294.
- Lin S., Huang Y., Min D., Li Z., Xie D., Li S., et al. Influence of electron beam irradiation on surface trao properties and surface conduction of epoxy resin microcomposites. *Insulating Materials* 2016; (12): 55-61.
- 11. Ma Y., Zheng X., Bie C. Influence of electron beam radiation on electrical properties of PTFE. *Insulating Materials* 2009; 42(4): 53-56.
- Zheng X., Liu X. Zhang Y., Ma L. Influence of high energy electron beam radiation on electrical characteristic of dielectric materials. *Proceedings of the CSEE* 2007; 27(30): 32-37.
- Liu M., Zhou F., Chen Z., Li J., Min D. Characterizing trap distribution in LDPE and HDPE based on isothermal surface potential decay measurement. *Proceeding of the CSEE* 2016; 36(1): 285-291.
- 14. Gao Y., Li Y., Cui J., Du B. Effect of gamma-ray irradiation on surface trap distribution of epoxy resin. *Transactions of China Electrotechnical Society* 2012; 27(12): 264-269.