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Effects of Gamma-Ray Irradiation on Tracking Failure of Polymer Insulating Materials

Boxue Du, Yu Gao and Yong Liu
Tianjin University
China

1. Introduction

Electrical insulation and dielectrics play a key role in the performance and reliability of most electrical systems, where a single-point system failure may prove catastrophic or even fatal to the electrical equipment. Polymers are the most commonly used dielectrics because of their reliability, availability, ease of fabrication and low cost. The selection of the proper polymer dielectric for a desired application depends on the requirements and operating conditions of the system. Voltage surges are known to be one of the factors leading to tracking failure, which often appear in lighting, switching and circuit breaker operation. Tracking failure is a dielectric breakdown phenomenon occurring on polymer surfaces comprising carbonized conductive paths. When a sufficiently intense discharge lasts for a considerable time, the decomposed carbon products, with some parts of the channel carbonized, are progressive and rapidly form on the sample surface. When the carbonized deposits bridge the interval between electrodes, a sudden decrease in the insulating resistance occurs. The importance of this information has been revealed for preventing fires, short-circuit and insulation failure in electrical appliances and devices, and especially the tracking and ignition of polymers have been examined.

Polymer materials are widely used in radiation environments, such as scientific research fields and nuclear power stations, where the high safety and reliability are demanded. The polymer insulating materials are inevitably exposed to various kinds of radiation. The changes in their physical and chemical properties could prematurely terminate the useful life of the dielectric. Outside and inside of the secondary shield in the containment vessel of nuclear power plants, the maximum dose rates of irradiation are 0.01 Gy/h and 1 Gy/h. The dose rate in nuclear power plants varies widely from 10 µGy/h to 10 kGy/h with a potential of total exposure 1000 kGy or greater. The high reliability is based on construction safety and system safety during the use of electrical equipment. Accordingly, it is important to investigate the influence of radiation on polymeric insulating materials used in the radioactive environments. Presently, the researches on irradiation aging are mostly concentrated on electrical and mechanical performance, but the effect of radiation, especially the radiation aging theory is rarely studied. Kuriyama et al have measured the physical and electrical properties of gamma-ray irradiated PVC jacketed cable and concluded that the electrical receptivity of PVC is reduced obviously. We have investigated the tracking resistance of gamma-ray irradiated polyethylene and modified

polycarbonate materials, and the results reveal that the tracking resistance is improved by irradiation for cross-linking type materials, but the conclusions for degradation type materials are opposite.

The polymers used in the field of electrical and electronic engineering are not only subject to an electric field alone, quite often they operate under the influence of both electric and magnetic fields. In addition to shielding effects against space plasma flux, magnetic field changes the electron kinematics and the gas desorption rate, and hence the flashover potential. The effect of a magnetic field parallel to an applied electric field has been considered and reported that a magnetic field can shorten the formative time lag of breakdown. Magnetic field might influence dielectric breakdown in different ways. The basic mechanisms and physical phenomena providing magnetic insulation have been investigated in the past mainly for vacuum-insulator interfaces in high-power transmission lines and vacuum diodes. The influence of magnetic field on the sparking voltage has been examined for a slightly non-uniform electric field applied perpendicularly to a magnetic field, including its effects on the formative time lag and on the cycloidal movement of electrons. Low energy non-disruptive discharge carbonized "track" marks have been identified by a number of users on gas insulating switchgear (GIS) insulating surfaces and the cause of this phenomenon has not yet been identified. However, there have been very few investigations concerning the influence of magnetic field on the tracking failure of polymer materials used in electrical equipments. Therefore, it is necessary to study how their electrical characteristics change under magnetic field.

Polymers are used on space equipment with the examples of these devices working at high dc voltage on satellites such as ionic propulsion systems, photomultiplier tubes, scientific electronic instruments, communication systems and solar cell power supply systems. Energetic electromagnetic radiation and low pressure are two main factors in the space environment. We have reported that surface discharge occurs earlier at low pressures than atmospheric pressure. In fact, surface discharge is one of the principal causes for premature failure in electronic equipments used in low pressure regions. The dielectric properties of the polymers under the environments of low pressure and radiation may change. It is necessary to study the way how their electrical characteristics change under the combined environments.

As technology advances, increasing demands on the reliable operation under various operating and environmental conditions are made on materials and components. Therefore, it is necessary to study how their electrical characteristics change with consideration of radiation, low pressure and magnetic field. However, knowledge of influence of irradiation under combined conditions of radiation, low pressure and magnetic field is limited, and systematic study is desirable.

Polymer materials are widely used in electrical insulation because of their high breakdown strength, high resistivity and low dielectric loss. Polybutylene naphthalate is widely used in aerospace electronics, electric engineering, chemical, metallurgy and medical equipment. Polybutylene terephthalate is usually used for electric coupler, blind plug, switch and insulating cover. Polyethylene terephthalate is one of the most widely used polymers for electrical and electronic industries. Whether the dielectric property of the polymers is changed by irradiation and magnetic field or not are worth investigating. In this chapter, effects of total dose of gamma-ray irradiation on tracking failure of polymer materials are described. The results reveal that the tracking failure properties are greatly changed by gamma-ray irradiation.

2. Experiment setup and procedure

2.1 Test samples

Polybutylene naphthalate (PBN) and polybutylene terephthalate (PBT) are produced with butanediol by reaction with 2, 6-naphthalic acid and dimethyl terephthalate, respectively. Polyethylene terephthalate (PET) is produced with ethylene glycol by reaction with purified terephthalic acid. For comparison, both polycarbonates mixed with 3% polyethylene (M-PC) and polyethylene (PE) samples are also tested.

The samples are irradiated in air up to 100 kGy and then up to 1000 kGy with a dose rate of 10 kGy/h by using a ^{60}Co gamma-source. The thickness and dimensions are 3 mm and 20 mm×20 mm. The sample surface is cleaned with ethyl alcohol and dried in a desiccator in air at room temperature for 24 h or longer before the testing.

2.2 Experimental apparatus and procedure for pulse voltage application

Schematic diagram of experimental set-up is shown in Fig. 1. Test sample is stressed electrically under a HV pulse voltage at the level of 30 kV and the pulse width is 1 ms. The pulse interval is adjusted from 5 ms to 10 ms. The electrode geometry is needle-plate electrode pattern, where the needle electrode is connected to the HV voltage and the plate electrode is grounded through the discharge detecting circuit. The length of each stainless steel electrode is 45 mm, the radius of the semicircle plate electrode is 5 mm and the thickness is 0.5 mm. The diameter of the needle electrode is 0.65 mm and the interval between the electrodes is 3 mm. The magnetic environment is produced by the electromagnet (PEM-50). The magnetic flux density (MFD) is 495 mT and the direction of E×B is 0, 90 and 270 degrees with respect to the sample surface. The vacuum chamber is made from a glass tube with two sheets of acrylic board for the cover and bottom.

(a) Relative angles

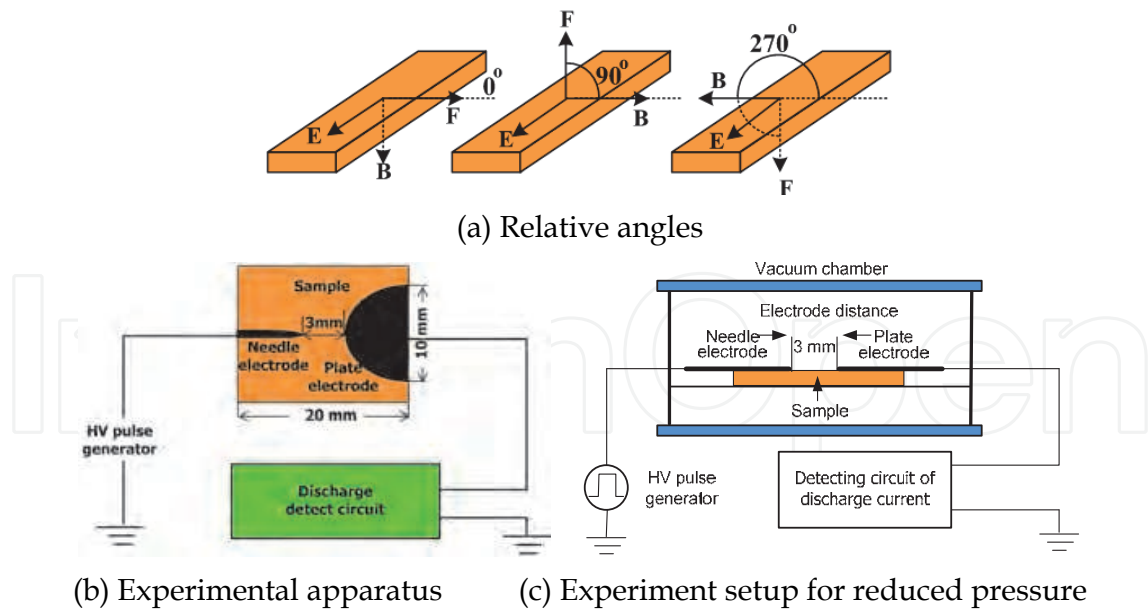(b) Experimental apparatus   (c) Experiment setup for reduced pressure

Fig. 1. Schematic diagram of test system for pulse voltage

Tracking failure is defined as the insulation failure under electric field with consequent lack of dielectric properties, which is determined when the surface is burning or short-circuit of the electrodes has persisted for 2s. The tracking failure is caused by decomposed carbon on the sample surface due to heat generated by the discharge, which starts locally across the

electrodes and forms on the sample surface. A high-speed general purpose AD conversion module (DRF2A) is connected with the experimental circuit in order to measure the discharge current. The sampling time used by this method is 25 µs. The test is repeated 10 times, thus each datum is an average of ten measurements. A monocular-video-zoom microscope with the magnification of 420 is used to acquire the pictures of tracking failure for the detailed analysis to identify the tracking failure phenomenon.

2.3 Experimental apparatus and procedure for IEC 60112 tracking test

The experimental set-up of tracking failure is constructed according to the specifications given in IEC 60112 as shown in Fig. 2. The thickness of each electrode is 2 mm, the length 45 mm, the width 5 mm, and the interval distance between the two electrodes 4 mm. The solution is 0.1% NH_4Cl in de-ionized water, giving a resistivity of approximate 395 Ω cm at the temperature of 23 °C. Droplets are applied at the intervals to keep up the discharge between the electrodes on the sample surface. The droplet size of the electrolyte is 20 mm³ and the time interval between each added droplet is 30 s. Tracking failure is defined as when the sample burns or the current exceeds 0.5 A and persists for 2 s. But because some of the samples do not track evenly at the highest test voltage, repeated discharges gradually cause the erosion of the sample surface between the electrodes. The erosion depth and weight loss are measured after adding 100 droplets.
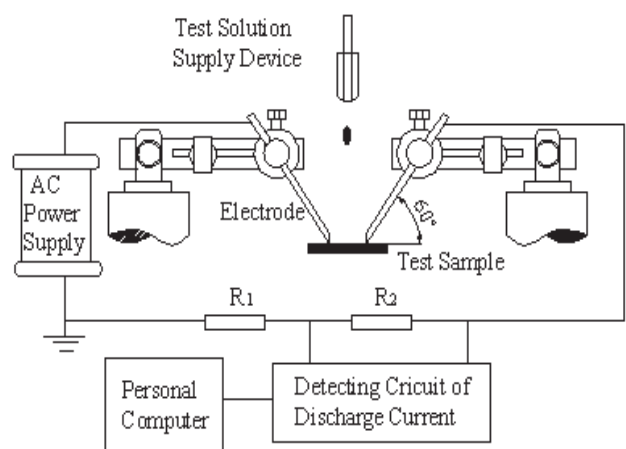
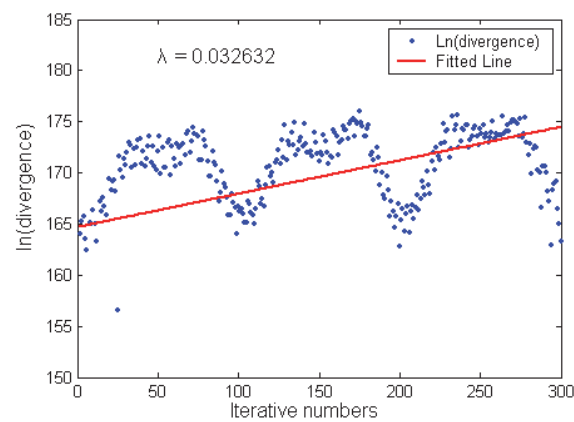Fig. 2. Experimental set-up for IEC 60112 test

Fig. 3. Estimate of largest Lyapunov exponent

3. Methods of data-processing

3.1 The proof for chaos in the discharge process

Detecting the presence of chaos in the discharge process is a precondition for applying the non-linear method to analyze discharge currents. Lyapunov exponents characterize the exponential divergence of neighbor embedding vectors X(i) and X(j) in phase space and one or more positive Lyapunov exponents can provide a necessary and sufficient proof for a dynamic system to be chaotic. By calculating the largest Lyapunov exponent of an experimental time series, the discharge process is proved chaotic. Fig. 3 shows an estimation of the largest Lyapunov exponent by the algorithm given by Rosenstein. The data are sampled at the middle state of the tracking test with unirradiated PBT at 375 V. Here, the largest Lyapunov exponent is calculated as 0.032632 indicating that the process is chaotic.

3.2 Recurrence plot method

Recurrence plots (RPs), which are used to characterize the degree of aperiodicity of time series, are first introduced by Eckmann. The RP method provides a useful graphical framework for extracting information from time series. By using the RP method, the non-linear characteristics of dynamical systems can be illustrated. The RP method is constructed as follows:

First, according to Takens' embedding theorem, vector X(i) can be reconstructed from a time series x(i) :

$$X_{(i)} = [x(i), x(i+\tau), ..., x(i+(m-1)\tau)], i = 1, 2, 3... \tag{1}$$

Where X(i) is the embedding vector, m is the embedding dimension, and τ is the delay time; x(i) is the time series which stands for the discharge current series in this paper. The embedding dimension m and delay time τ can be chosen by the C-C method. For a given time series x(i), the estimation of τ assures that the coordinates x(i) and x(i+t) are somewhat independent but not completely uncorrelated. The C-C method provides a good criterion for the choice of m and τ and also works well in the presence of noise, especially for noise levels below 30%.

Then the RP can be drawn based on the following matrix:

$$R_{i,j} = H(\varepsilon - ||X_{(i)} - X_{(j)}||_2), X_{(i)}, X_{(j)} \in R^m, i, j \in (1, M) \tag{2}$$

where ε is a predefined threshold, H(x) is the Heaviside function, and M is the number of X(i) vectors constructed from the time series. The value one in this matrix is plotted in black in the RP. The choice of ε is the critical, if it is too big, it will produce irrelevant points, and if it is too small, useful information will be lost. In the investigation, ε is chosen as 0.25σ. Where, σ is the standard deviation of the time series.

The discharge currents sampled directly from the tracking test of IEC 60112 have power frequency components of leakage currents, which have negative effects on non-linear analysis. To filter the power frequency components, wavelet transform of discharge currents are applied and the Morlet wavelet db8 is chosen as the mother wavelet.

Fig. 4 shows an RP example of discharge current with unirradiated PBT at 325 V. Fig. 4a shows an original discharge current and the related RP. It can be concluded that the discharge occurs frequently at the positive and negative peaks of the waveform. Due to the ac voltage, the current waveform has power frequency components. Reflected in the RP, recurrence points are mainly parallel to the diagonal. The distribution of recurrence points of sinusoidal series in RP is determined by the RP method. The periodic characteristic of RP

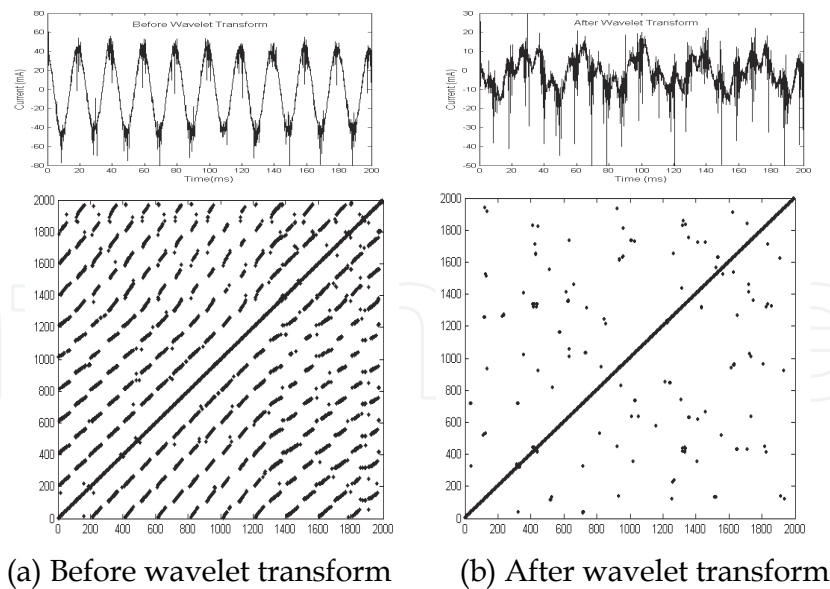(a) Before wavelet transform   (b) After wavelet transform

Fig. 4. Examples of recurrence plots of discharge current

covers non-linear features and acts as adverse factor for non-linear analysis. Fig. 4b shows the treated discharging sequence by wavelet transform and its reconstructed RP. After the data preprocessing, the recurrence points are randomly distributed in the picture and the topological structure can actually reflects the non-linear characteristics. The effects of dosages of gamma-ray irradiation on the resistance to tracking for both PBT and PET are presented by comparing the RP structure, as the above description.

4. Dielectric breakdown and tracking resistance

4.1 Dielectric breakdown by HV pulse voltage application

Fig. 5 shows the photographs of sample surface after the discharge for 20 s at the discharge interval of 10 ms. The color of sample surfaces in both PBN and PBT are gradually dark as the total dose of gamma-ray irradiation increases. The noticeable changes of sample surface in the breakdown process are evidently observed. The repetitive discharges occur before the dielectric breakdown and the quantity of decomposition carbon increases in the area close to the electrodes. As shown in Fig. 5, a carbonization conducting path forms on the surface between the electrodes. The surface is carbonized by the heat energy from the discharge.With the high repetition rate of discharges, there is a high increase of discharge quantity which affects the carbonization progress and the situation of carbon deposition. When the intensive discharge lasts for a certain time, the surface is carbonized first to the area close to the electrodes. Due to the conductive nature of carbonization products, they act as the extension of the electrodes. After a sufficiently long time, the carbonized conduction path finally forms between the electrodes. By comparing Fig. 5a and 5b, the features of dielectric breakdown phenomenon are significantly different between PBN and PBT. The carbonized area decreases with increasing the total dose of irradiation with PBN, but increases with PBT. The difference indicates that the thermal properties are improved with PBN, but worsened with PBT.

Fig. 6 shows the relation between the time to dielectric breakdown and the total dose of the irradiation. For the investigated range of the irradiation, the time to dielectric breakdown

increases with the increase of the total dose with PBN, but decreases with increasing the total dose with PBT. The time to dielectric breakdown of both PBN and PBT decreases with
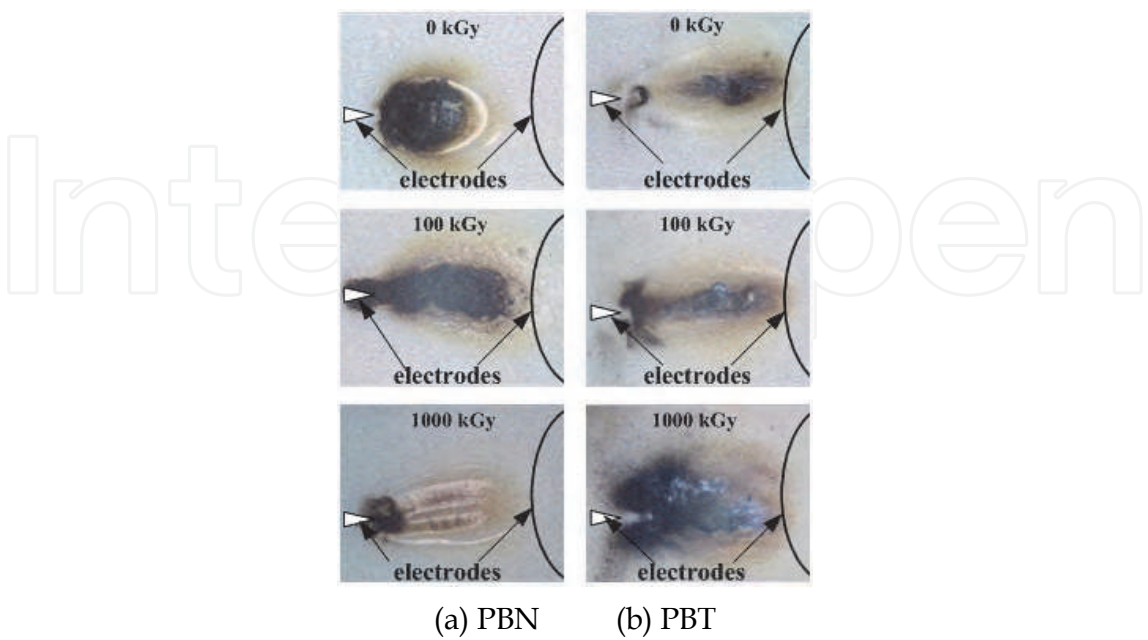

(a) PBN   (b) PBT

Fig. 5. Sample surface after carbonization

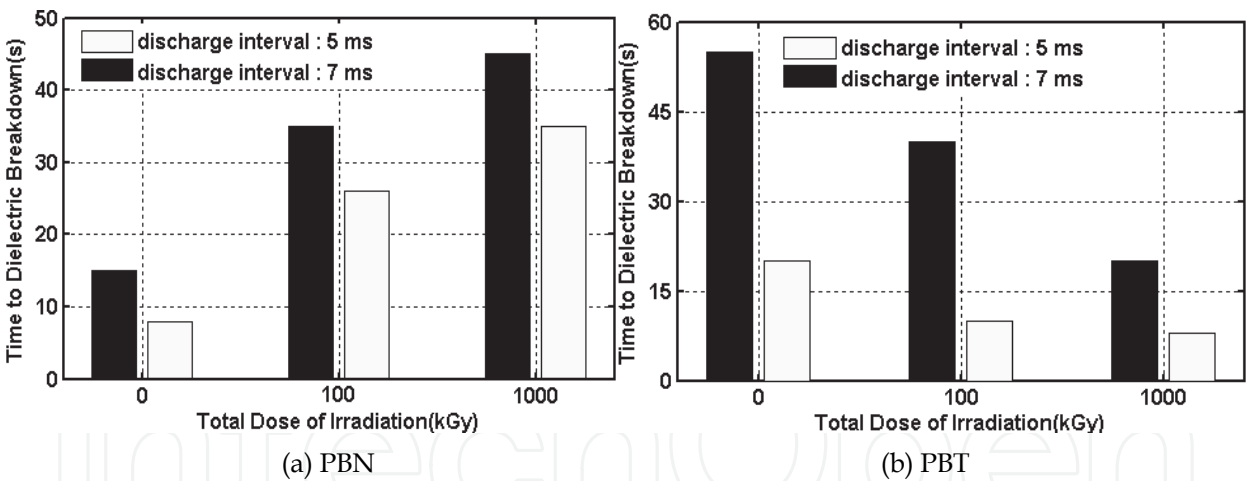(a) PBN   (b) PBT

Fig. 6. Relation between the time to dielectric breakdown and the total dose of irradiation

the decrease of the discharge interval. With the increase of the total dose of irradiation, the time to dielectric breakdown of PBN increases, which indicates that the thermal properties are improved. However, the time to dielectric breakdown of PBT decreases with the increase of the total dose, which indicates that the thermal properties are worsened. The atoms that make up a polymer are bounded together by weak covalent bonds that are disrupted easily by gamma-ray radiation, and as bonds are broken new ones are formed and the structure of the polymer is altered. In practice, cross-linking and degradation reaction often occur simultaneously, the reaction result is determined by the one that is dominant. For PBN, it is supposed that the cross-linking reaction exceeds the degradation reaction and the combination between the molecules extends the three-dimensional networks. For PBT, it is

supposed that the degradation reaction exceeds the cross-linking reaction and the scission of the main chain bonds results in the formation of low-molecular-weight chain fragments.

With the decrease of the discharge interval, the tendency of the time to dielectric breakdown decreases. When the discharge interval decreases, electron emission from the electrode becomes more frequent, the heat caused by the discharge energy which makes the carbon chain of the molecules broken increases, and the carbon conductive path of polymer surface forms more steadily. As a result, the time to dielectric breakdown decreased. By the way, at the discharge interval of 10 ms, the accurate time to dielectric breakdown of the test results does not show in the figure as the dielectric breakdown did not occur until 200 s under the same condition at discharge intervals of 7 ms and 5 ms.

Fig. 7 shows the relation between the discharge quantity and the total dose of irradiation after the discharge for 600 times. The dielectric breakdown is mainly dependent upon the thermal energy formed by the discharge. The discharge quantity is the total integrated charge of the discharge current during the dielectric breakdown period, which is the degree of the heat-durability. If the discharge quantity is smaller, it suggests that the carbon chain is harder to be broken. In other words, the heat-durability of the material is better. With the increase of the total dose, the discharge quantity decreases with PBN, which suggests that the thermal property is improved and the dielectric breakdown becomes not easy. As mentioned in Fig. 6a, the time to dielectric breakdown is delayed with the increase of the total dose. This fact may be due to the occurrence of the cross-linking reaction. With the increase of the total dose of irradiation, the discharge quantity increases with PBT, which suggests that the thermal properties are worsened and the dielectric breakdown becomes faster. As mentioned in Fig. 6b, the time to dielectric breakdown decreases with the increase of the total dose. The reason for this may be due to the degradation reaction by irradiation.

(a) PBN   (b) PBT
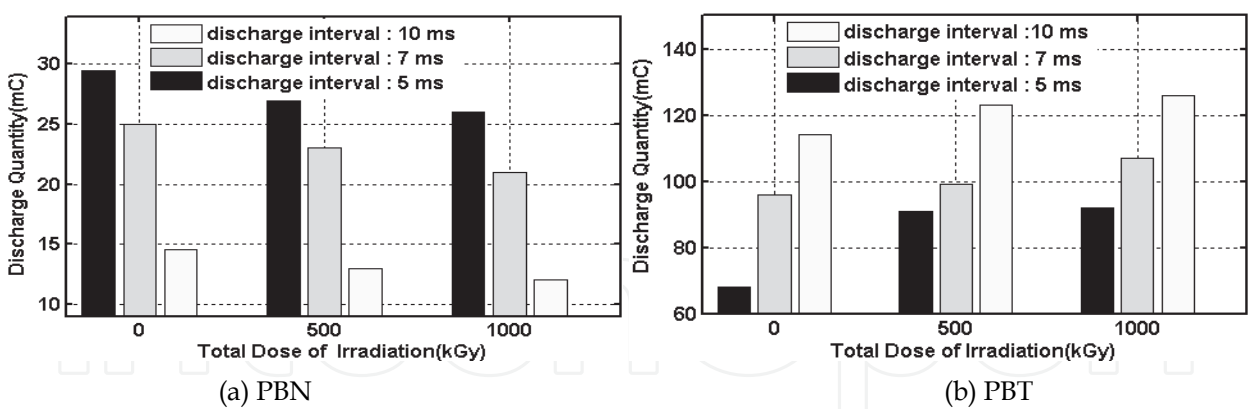
Fig. 7. Relation between the discharge quantity and the total dose of irradiation

From Fig. 7a and 7b, the discharge quantity increases with the decrease of discharge interval for PBN, but the tendency is opposite for PBT. The discharge is much more intensive in the shorter discharge interval because the accumulative energy of consecutive discharge is higher, and the dielectric breakdown occurs more quickly. For PBN, the dielectric breakdown is not easy due to the inherent property and the discharge was continuous. The formation of carbon conductive path is gradual, and the discharge quantity is bigger at the shorter discharge interval. For PBT, the carbonization and dielectric breakdown are faster. The discharge is discontinuous from the visual observation. At the shorter discharge

interval, the carbonization forms more quickly and the brief short circuit occurs more frequently, and the discharges could not completely take place, so the total discharge quantity is smaller as shown in Fig. 7b.

The dielectric properties are improved for PBN but worsened for PBT by gamma-ray irradiation. This difference is attributed to the radiation-induced cross-linking and degradation. The effect of the total dose on electrical properties is markedly different, depending on the chemical structure of the base polymer. The radiation adds a particular dimension to the aging problem, because it interacts strongly with materials in general and brings about structural changes that alter their properties. This is because that it can alter the macroscopic properties of polymeric materials through mechanisms like chain scission, cross linking and oxidation. Comparing the molecule formulas, there are two phenyls in the main chain of PBN but one for PBT. The amount of phenyl in the main chain plays a main role on the result of the radiation reaction. In another word, the dielectric properties of polybutylene polymers are improved for PBN which contains more combined phenyls in the main chain. It is known that to disrupt the two combined phenyls needs more energy than one phenyl. It is supposed that the disruption of the bonds is harder for PBN than PBT after the same dosage of the irradiation. The formation of new bonds does not seem to have much difference because of the same fringe structure. The new main chain is bigger for PBN because of more phenyls. For PBN, it is considered that the effects of the new bonds exceed the disruptive ones. For PBT, it is considered to be opposite to PBN. Therefore, PBN represents cross-linking type results and PBT represents scission type results.

4.2 Effects of tracking resistance by use of IEC60112 method

To estimate the resistance to tracking of polymer material, there are many traditional methods including recording the time to dielectric breakdown, calculating the discharge quantity, measuring the dielectric loss angle and testing the CTI value. Among the methods, the CTI value is extremely important and also considered as an index mark to select insulating materials. The minimal voltage, which could cause tracking failure with the application of 50 drops of electrolyte, is used as a measure of the susceptibility of the material to tracking and is defined as the CTI value according to IEC60112. The example of discharge events is shown in Fig. 8. The test solution is evaporated by Joule heat caused by leakage current which flows between the electrodes across the sample surface. A discharge appears at the dry band. After the appearance of the discharge, the sample surface is eventually dried between the electrodes. The initiation of a carbon deposit is

(a) Scintillation discharge (b) Arc discharge (c) Discharge and carbonization (d) Intense discharge

Fig. 8. Example of discharge events with 100 kGy irradiated M-PC.

closely related to the location where a dry band is formed in the evaporation of solution due to Joule heat, and to heat degradation of the sample surface caused by scintillation discharge across the dry band. With further application of the test solution, the erosion of the sample surface occurs as a result of the tracking test. Fig.9 shows the relation between dosage of the irradiation and the CTI value for both PET and PBT with ac voltage application. As total dosage increases, the CTI value of PET increases, but the CTI of PBT decreases.
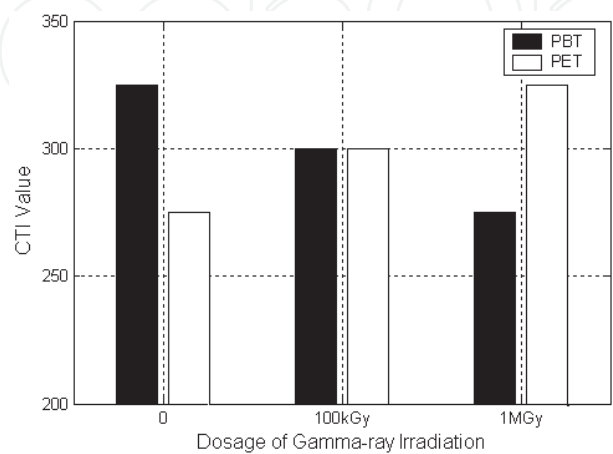
Fig. 9. Relation between the CTI value and the dosage of gamma-ray irradiation

(a) Erosion depth   (b) Weight loss
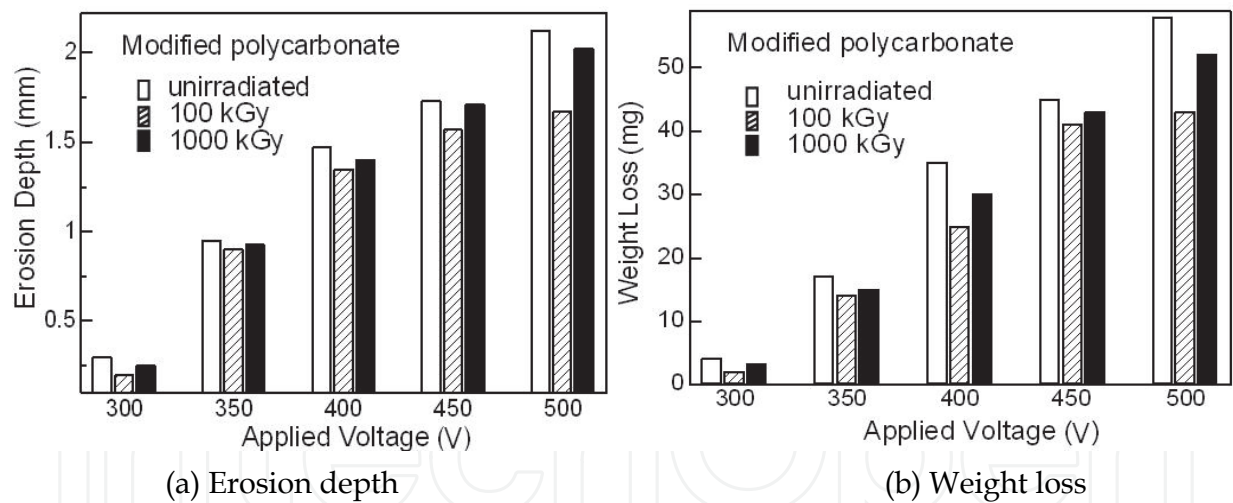
Fig. 10. The changes of tracking resistance on M-PC

Fig. 10 shows the relationship between the biggest erosion depths, weight loss and test voltage with M-PC. For the range of irradiation, both the erosion depths and weight loss are smaller than that of unirradiated samples. It indicated that after the irradiation, the tracking resistance was improved. A cross-linking reaction of an organic material is one main factor for improving tracking resistance and conversely a degradation reaction is conceivable as a factor for decreasing tracking resistance. It is believed that the improvement is due to the result of the cross-linking reaction. However, both the erosion depths and weight loss decreases with increasing the total dose from 0 kGy to 100 kGy, but increases from 100 kGy to 1000 kGy. It indicates that there is a threshold value for the tracking resistance of M-PC

around 100 kGy. When the total dose exceeds the threshold value, the tracking resistance begins to decrease. The decrease might be attributed to the result of the degradation reaction.

The M-PC is mixed with 3% PE. It is known that gamma-ray irradiation caused degradation reaction with PC. Therefore, the mixing of PE maybe one main reason for the improvement of the tracking resistance with irradiated M-PC. In order to confirm the judgment, the tracking resistance of PE after gamma-ray irradiation was investigated.

Fig. 11 shows the relationship between the biggest erosion depths, weight loss and test voltage with PE. For the range of the irradiation, both the erosion depths and weight loss decreased with the increase of the total dose. Accordingly, the trend that the tracking resistance improves through gamma-ray irradiation is assumed. It is also supposed that the heat from irradiation causes the formation of 3-dimensional structures, which strengthens the PE by cross-linking reaction. Therefore, the mixing of PE is probably one main reason for the improvement of the tracking resistance with irradiated M-PC.

(a) Erosion depth   (b) Weight loss
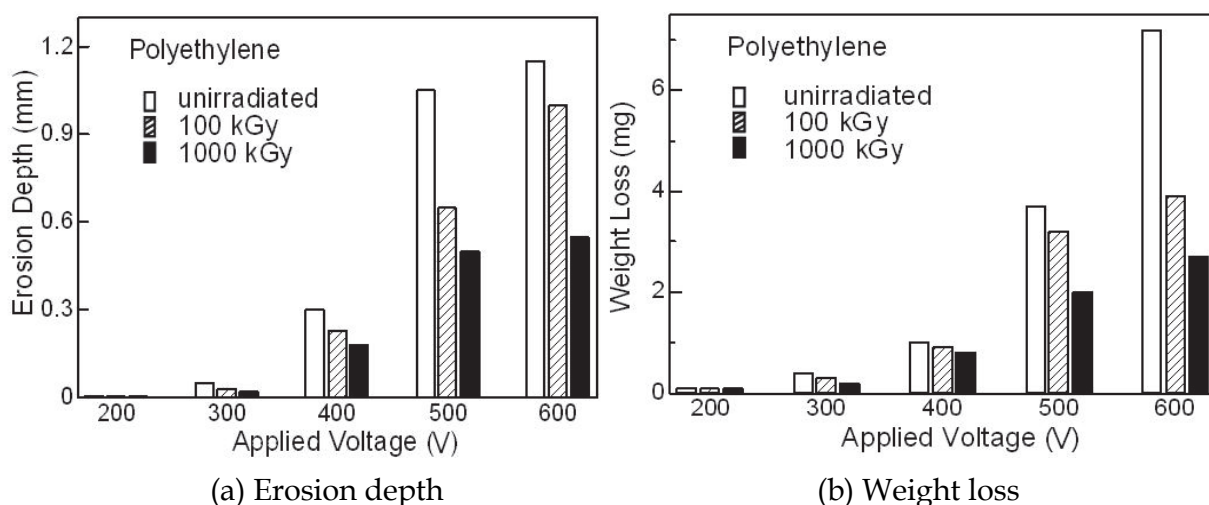
Fig. 11. The changes of tracking resistance on PE

5. Effect of gamma-ray irradiation on tracking resistance under reduced pressure

The relation between the time to tracking failure and the total dose of the irradiation under 100 kPa and 1 kPa are shown in Figs. 12, 13 and 14. With increasing the total dose of irradiation, the time to tracking failure increases with PBN and PET, but decreases with PBT for the investigated range of the irradiation. With the decrease of the atmospheric pressure, the time to tracking failure of all samples increases. With decreasing the pulse interval, the time to tracking failure of all samples decreases.

With the increase of the total dose, the time to tracking failure of PBN and PET increase, which indicates the thermal properties of tracking resistance are improved. However, the time to tracking failure of PBT decreases with the increase of the total dose, which indicates that the thermal properties of tracking resistance are worsened. Evidently, the total dose has different effects on the polymers. A cross-linking reaction is one main factor for improving tracking resistance and conversely a degradation reaction is conceivable as a factor for decreasing tracking resistance. The atoms that make up a polymer are

bounded together by weak covalent bonds that are disrupted easily by gamma-ray radiation, and as bonds are broken, new ones are formed and the structure of the polymer is altered. In practice, cross-linking and degradation reaction often occur simultaneously, and the reaction result is determined by the one that is dominant. For PBN and PET, it is supposed that the cross- linking reaction is superior to the degradation reaction and the combination between the molecules extends the three-dimensional networks. For PBT, it is supposed that the degradation reaction is superior to the cross-linking reaction and the scission of the main chain bonds results in the formation of low-molecular-weight chain fragments.

The time to tracking failure of all samples increases with the decrease of the atmospheric pressure. Under the reduced pressure, the supply of oxygen is not sufficient. The probability of the oxidation reaction on sample surface and the cracked gas decreases with the decrease of the oxygen content. The burning becomes more difficult and the complete carbonized conductive path becomes hard to form. Therefore, the time to tracking failure increases with the decrease of the atmospheric pressure.
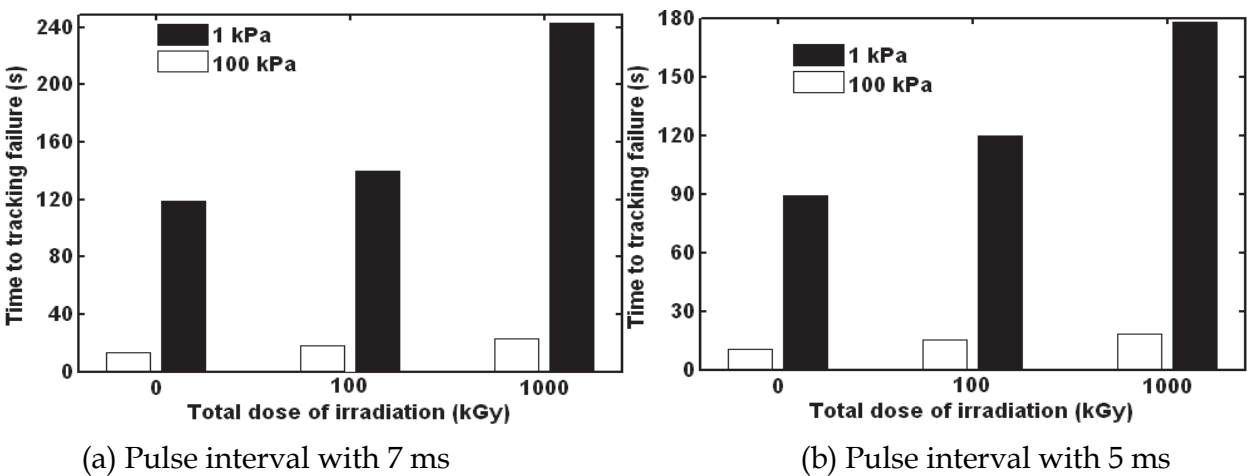
(a) Pulse interval with 7 ms   (b) Pulse interval with 5 ms

Fig. 12. Relation between the time to tracking failure and the total dose of irradiation for PBN

(a) Pulse interval with 7 ms   (b) Pulse interval with 5 ms

Fig. 13. Relation between the time to tracking failure and the total dose of irradiation for PET

(a) Pulse interval with 7 ms   (b) Pulse interval with 5 ms
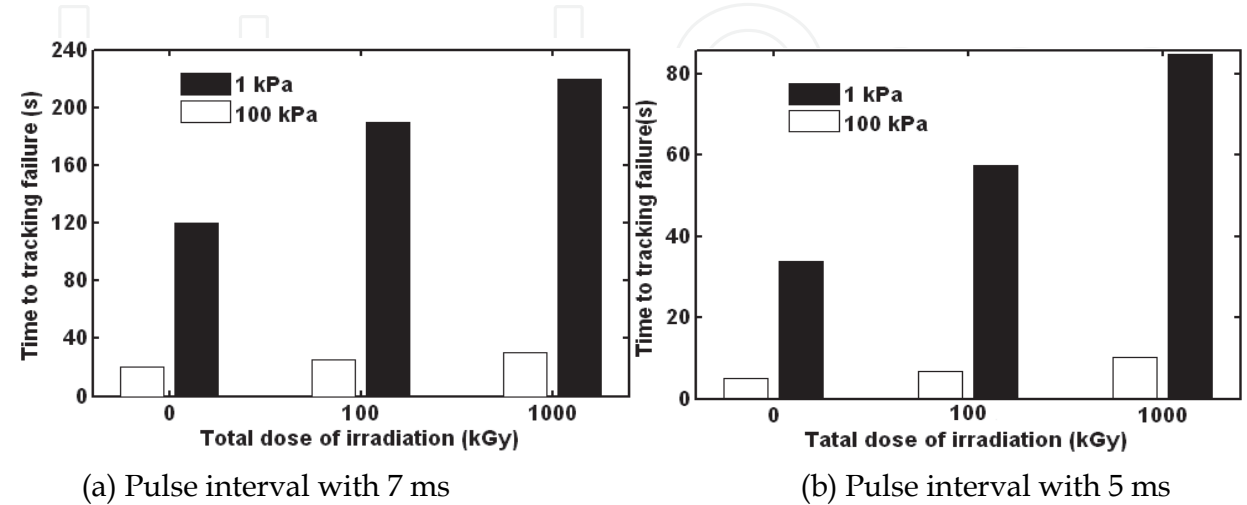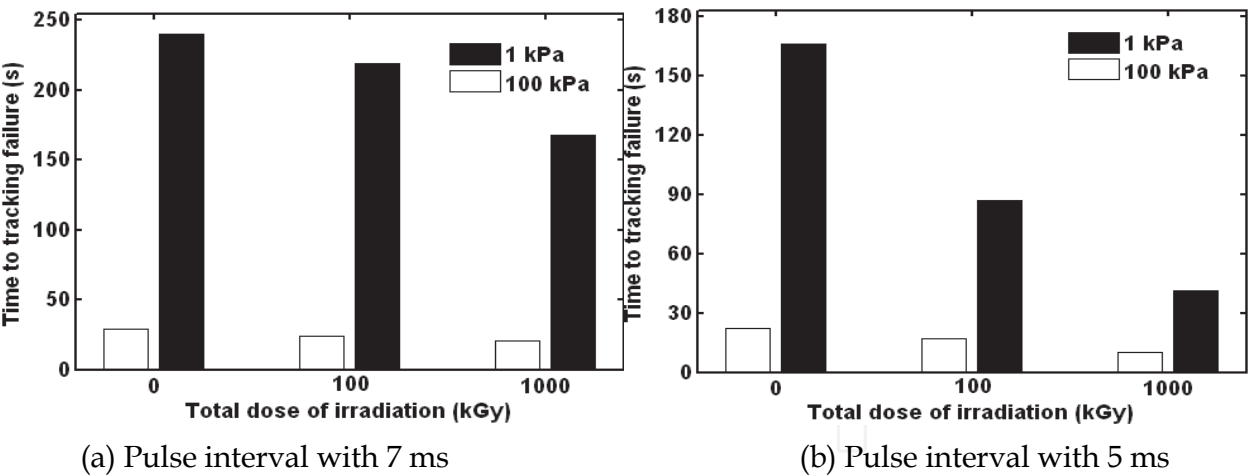
Fig. 14. Relation between the time to tracking failure and the total dose of irradiation for PBT

The time to tracking failure of all samples decreases with the decrease of the pulse interval. When the pulse interval decreases, electron emission from the electrode becomes more frequent, the heat caused by the discharge energy makes the carbon chain of the molecules broken increases, and the carbon conductive path of polymer surface forms more steadily. As a result, the time to tracking failure decreases. In addition, at the pulse interval of 10 ms, the accurate time to tracking failure is not shown from Figs. 12 to 14 as the tracking failure does not occur until 300 s under the same condition as pulse intervals of 7 ms and 5 ms.

The discharge quantity is a token of the heat-durability. The tracking failure mainly depends upon the heat energy formed by the discharge. If discharge quantity is smaller, it suggests that the carbon chain is more difficult to be broken. The relationship between the discharge quantity and the total dose of irradiation before tracking failure for 600 discharges under 100 kPa and 1 kPa are shown in Figs 15, 16 and 17. The discharge quantity decreases with PBN and PET, but increases with PBT with increasing the total dose of irradiation. From Figs. 15 to 17, with the decrease of the atmospheric pressure, the discharge quantity of PBN and PET increase, but decrease with PBT.

(a) Pulse interval with 10 ms   (b) Pulse interval with 5 ms
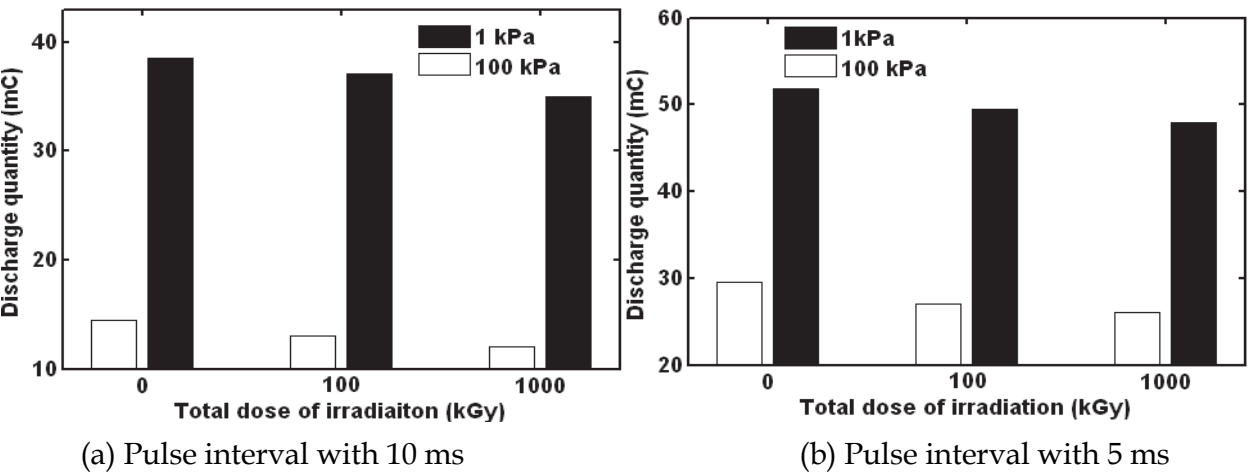
Fig. 15. Relation between the discharge quantity and the total dose of irradiation for PBN.

The discharge quantity of PBN and PET decrease with the increase of the total dose, which suggests that the thermal properties of tracking resistance are improved and the tracking

failure becomes more difficult. This fact is due to the occurrence of the cross-linking reaction. With the increase of the total dose of irradiation, the discharge quantity increase with PBT, which suggests that the thermal properties are worsened and the tracking failure becomes easier. The reason for this is due to the degradation reaction by irradiation.

(a) Pulse interval with 10 ms   (b) Pulse interval with 5 ms
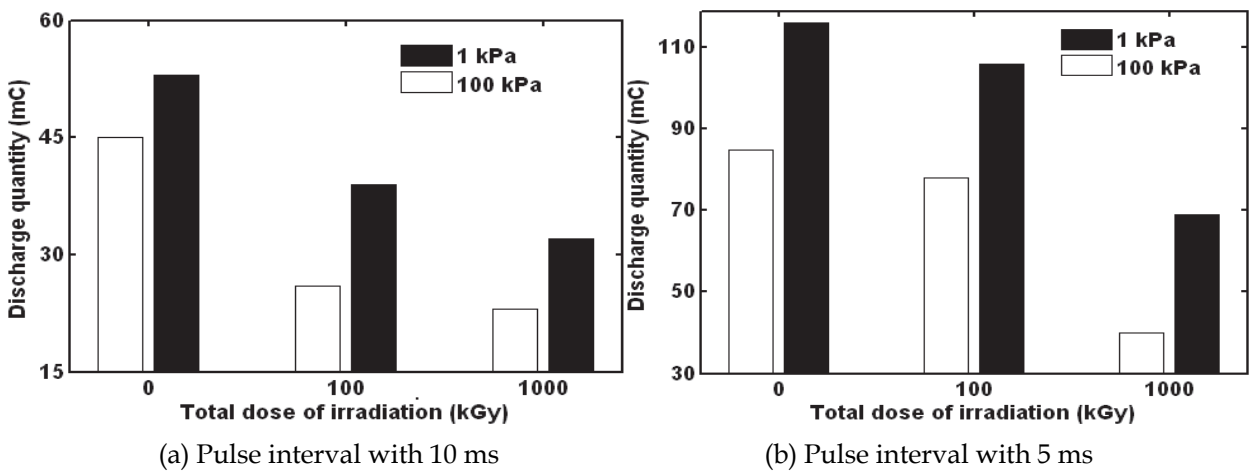
Fig. 16. Relation between the discharge quantity and the total dose of irradiation for PET

The discharge quantity of PBN and PET increase with decreasing the atmospheric pressure. Under the reduced atmospheric pressure, the density of the gases is decreased, the speed of electron becomes fast, and the surface more readily causes the discharging. As a result, the discharge quantity increases with the decrease of the atmospheric pressure. The discharge quantity decreases with decreasing the atmospheric pressure for PBT. It is because under the reduced atmospheric pressure, the probability of the oxidation reaction of PBT surface and the cracked gas decrease with the decrease in the oxygen content. The carbon deposition of PBT by the ignition to the surface increases, and the discharges can not completely take place due to the decomposed carbon, so the total discharge quantity of PBT under the reduced atmospheric pressure is smaller.

(a) Pulse interval with 10 ms   (b) Pulse interval with 5 ms
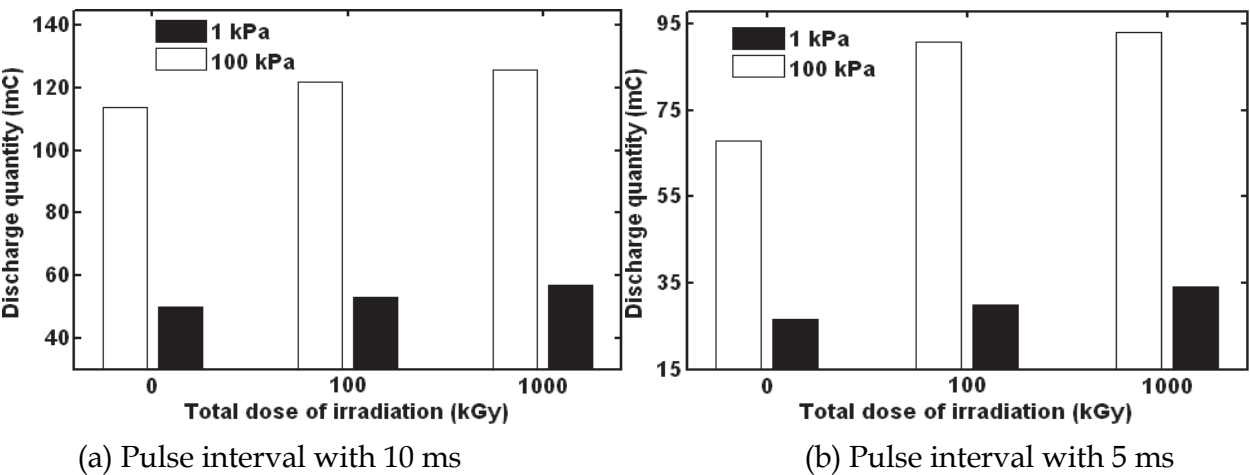
Fig. 17. Relation between the discharge quantity and the total dose of irradiation for PBT

From Figs. 15 to 17, discharge quantity of PBN and PET increased with the decreasing the pulse interval, but the tendency is opposite for PBT. The discharge quantity is bigger in

shorter pulse interval because the higher accumulative heat quantity of consecutive discharge is, the more quickly tracking failure occurred. For PBN and PET, tracking failure is more difficult due to the inherent carbonization property and the discharge is continuous. The formation of carbon conductive path is gradual, and the discharge quantity is bigger at the shorter pulse interval. For PBT, the formation of carbonization and tracking failure are easier. It is observed that short-circuit forms in a very short time, and the discharge is discontinuous. At the shorter pulse interval, the carbonization forms more quickly and the brief short-circuit occurs more frequently, which causes the discharges cannot completely take place, so the total discharge quantity is smaller.

The photographs of sample surface after the discharge for 20 s with the pulse interval of 10 ms under 100 kPa and 1 kPa are shown Tables 1, 2 and 3. The color of sample surfaces is gradually dark according to the total dose of gamma-ray irradiation. The noticeable changes of sample surface in the tracking failure process are observed. The repetitive discharge occurs before tracking failure and the quantity of decomposition carbon increases in the area close to the electrodes. The features of tracking failure phenomenon are different between PBN, PET and PBT. The carbonized area decreases with PBN and PET, but increases with PBT with increasing the total dose of irradiation. The difference indicates that the thermal properties of tracking resistance are improved with PBN and PET, but worsened with PBT.
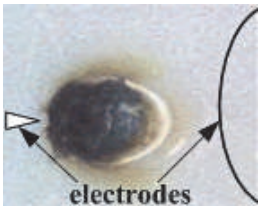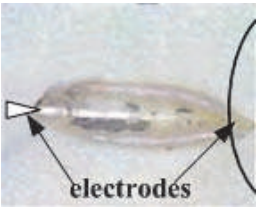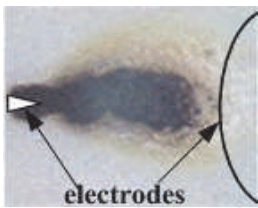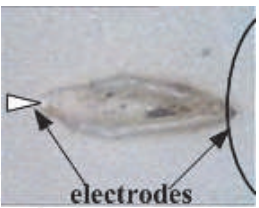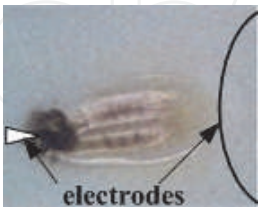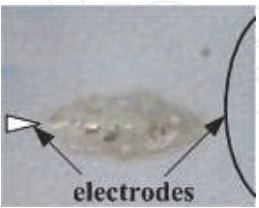
Total dose	100 kPa	1 kPa
0 kGy		
100 kGy		
1000 kGy		

Table 1. Sample surface after the carbonization with PBN

By comparison with the case of 1 kPa, the features of tracking failure phenomenon are different in the case of 100 kPa. The oxidation reaction, which took place on the electrodes, is an exothermic reaction initiated by scintillation discharge and increases the intensity of the discharge. In the case of 100 kPa, there is enough aerial oxygen for the samples to be oxidized completely, which shows more carbonization area in the second columns of Tables 1, 2 and 3. In the case of 1 kPa, there is not enough aerial oxygen for the samples to be

oxidized completely and the oxidation product is largely decreased, which shows less carbonization points in the third columns of Tables 1, 2 and 3. The differences of shape and area of carbonized resultants are independent of the carbonization process with reducing the atmospheric pressure.
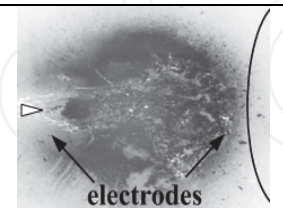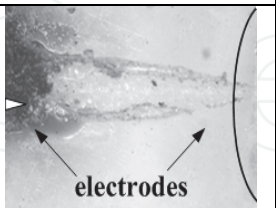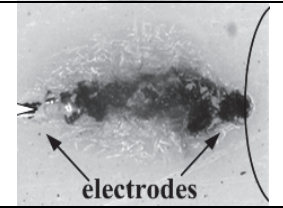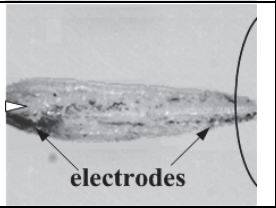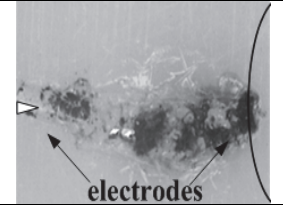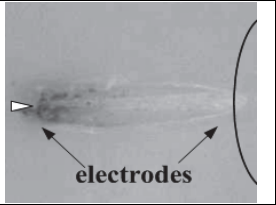
Total dose	100 kPa	1 kPa
0 kGy		
100 kGy		
1000 kGy		

Table 2. Sample surface after the carbonization with PET
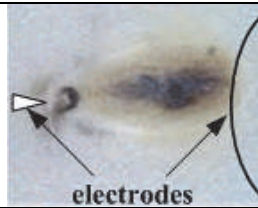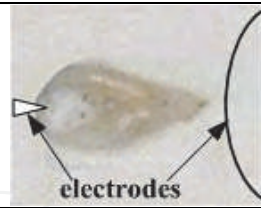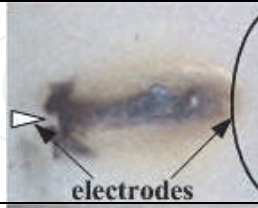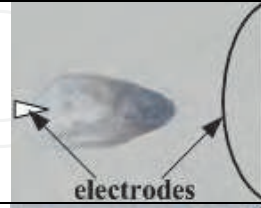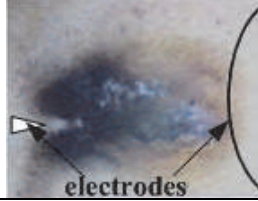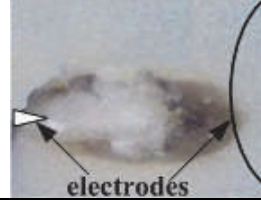
Total dose	100 kPa	1 kPa
0 kGy		
100 kGy		
1000 kGy		

Table 3. Sample surface after the carbonization with PBT

The tracking failure properties are improved for PBN and PET but worsened for PBT by gamma-ray irradiation. This difference is attributed to the radiation-induced cross-linking

and degradation. The effect of the total dose on electrical properties is markedly different, depending on the chemical structure of the base polymer. The radiation adds a particular dimension to the aging problem, because it interacts strongly with materials in general and brings about structural changes that alter their properties. This is because that it can alter the macroscopic properties of polymeric materials through mechanisms like chain scission, cross-linking and oxidation. By the comparison of the molecule formulas of PBN and PBT, there are two phenyls in the main chain of PBN but one for PBT. The amount of phenyl in the main chain plays a main role in the result of the radiation reaction. In another word, the resistance to the tracking failure of polybutylene polymers is improved for PBN which contains more combined phenyls in the main chain. Comparing the molecular structures of PET and PBT, there are four methylene groups in the chain of PBT but two for PET. The four methylenes increase the length of the thinner, less bulky, portion of the molecular chain, resulting in easier bending.

6. Effects of gamma-ray irradiation on tracking failure under magnetic filed

Figs. 18, 19 and 20 show the relation between the time to tracking failure and the total dose of irradiation with and without magnetic field. The time to tracking failure increases with increasing the total dose with PBN and PET, but decreases with PBT. Under the magnetic field, the time to tracking failure of all the samples increases with the relative angles of 0 and 90 degrees, but decreases with the relative angle of 270 degrees.

(a) Pulse interval with 10 ms   (b) Pulse interval with 5 ms
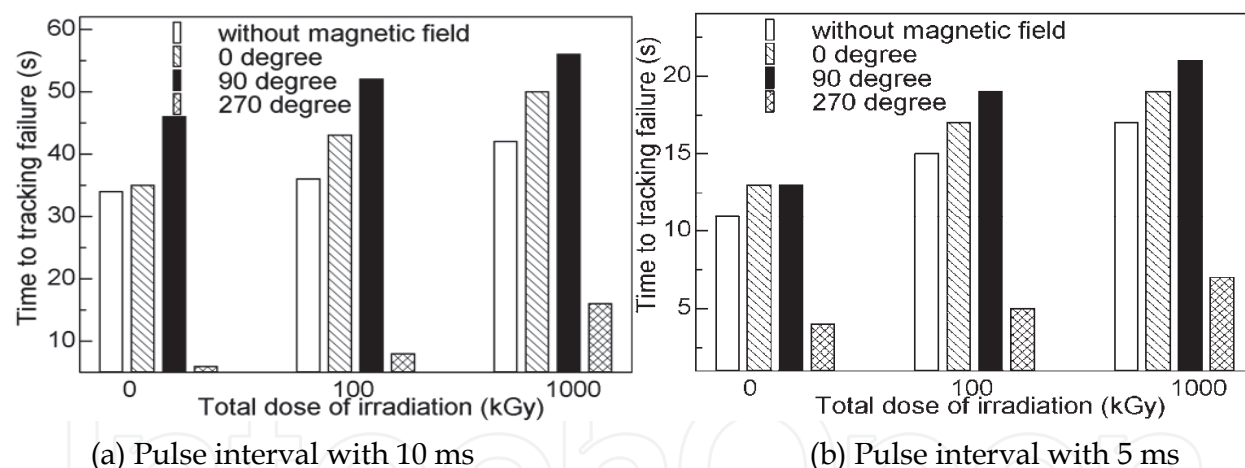
Fig. 18. Relation between the time to tracking failure and the irradiation with PBN

With increasing the total dose, the time to tracking failure of PBN and PET increases, which indicates the properties of tracking resistance are improved. However, the time to tracking failure of PBT decreases with increasing the total dose, which indicates that the properties of tracking resistance are worsened.

Under the magnetic field, the time to tracking failure of the three samples are delayed with the relative angles of 0 and 90 degrees, but shortened with the relative angle of 270 degrees. The tracking failure is caused by the decomposed carbon on the sample surface, which is precipitated due to heat generated by the discharge between the electrodes. Free electrons which are emitted from electrode dissociate polymer molecules by the rupture of C-H bond. When magnetic field is applied, an electromagnetic force, the direction of which is decided by the direction of E×B, will affect the frequency of electron collision and the formation of

decomposed carbon. As a result, the dielectric performance is changed by the magnetic field. With the relative angles of 0 and 90 degrees, the charge carriers are deflected to one side and upward away from the surface, respectively. As a result, there is a decrease in collision frequency and the time to tracking failure increases. When the relative angle is 270 degrees, the electrons are deflected towards the sample surface because of the electromagnetic force. The C-H bonds are ruptured and the carbon is separated more readily. Therefore, the time to tracking failure is shortened with the relative angle of 270 degrees.
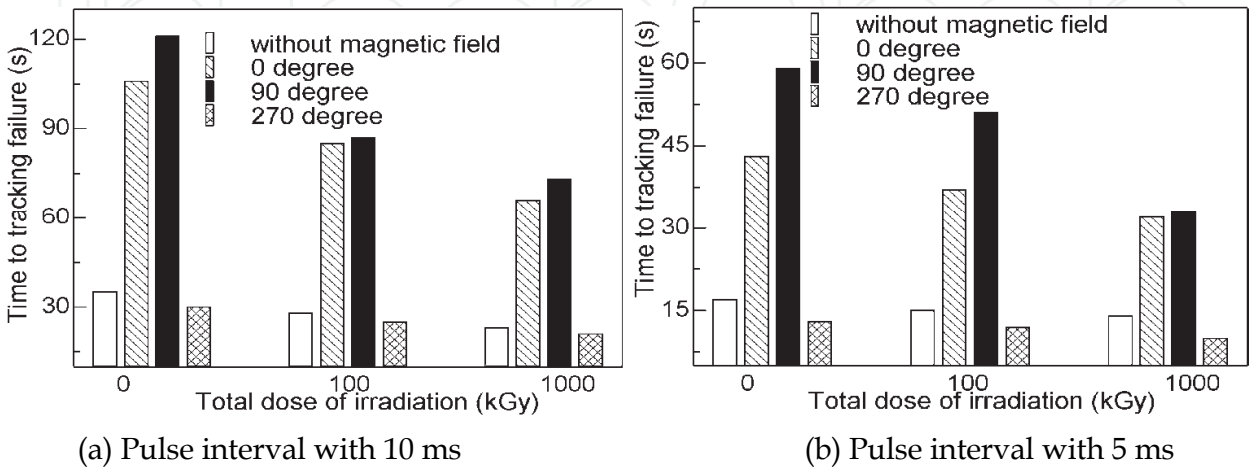
(a) Pulse interval with 10 ms   (b) Pulse interval with 5 ms

Fig. 19. Relation between the time to tracking failure and the irradiation with PBT

(a)Pulse interval with 10 ms   (b) Pulse interval with 5 ms
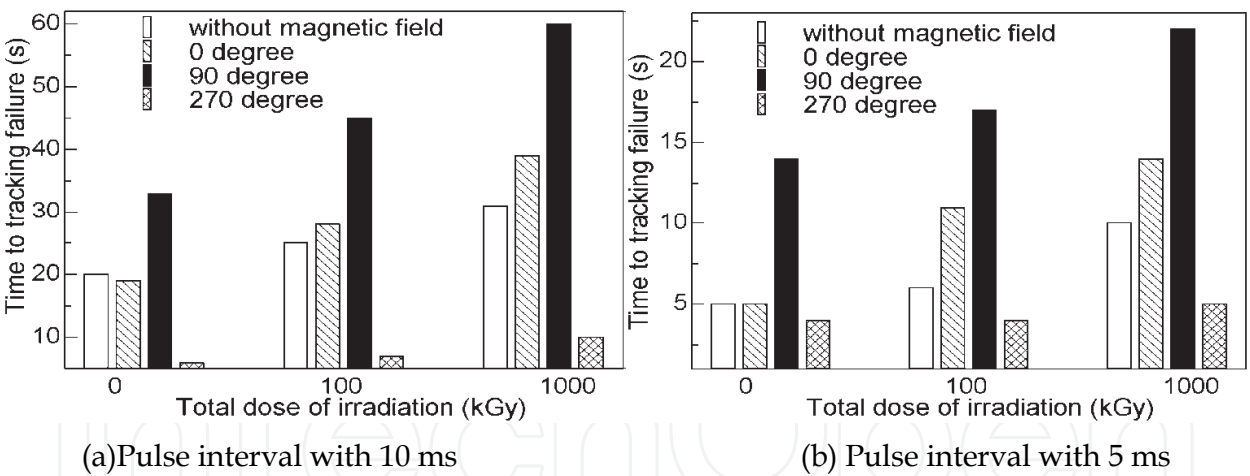
Fig. 20. Relation between the time to tracking failure and the irradiation with PET

Figs. 21, 22 and 23 show the relation between the discharge quantity and the total dose of irradiation. With the increase of the total dose of irradiation, the discharge quantity decreases with PBN and PET, but increases with PBT. Under magnetic field, the discharge quantity of the samples increases with the relative angles of 90 and 270 degrees, but decreases with the relative angle of 0 degree. The discharge quantity increases with the relative angles of 90 and 270 degrees, but decreases with the relative angle of 0 degree. In addition, it decreases with the relative angle of 90 degrees for PBT. When the relative angle is 0 degree, the electromagnetic force is parallel to the sample surface, which makes the electrons deviate to the surface. The tracking failure is suppressed, since the probability of the electron collision is decreased. Therefore, the discharge quantity is smaller.
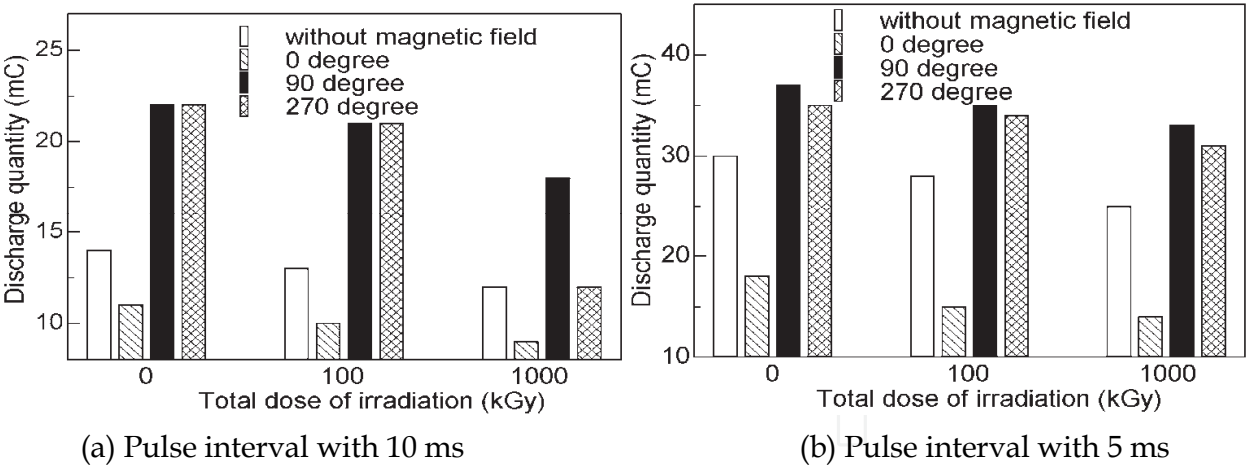
(a) Pulse interval with 10 ms   (b) Pulse interval with 5 ms

Fig. 21. Relation between the discharge quantity and the total dose of irradiation with PBN

(a) Pulse interval with 10 ms   (b) Pulse interval with 5 ms

Fig. 22. Relation between the discharge quantity and the total dose of irradiation with PBT

(a) Pulse interval with 10 ms   (b) Pulse interval with 5 ms
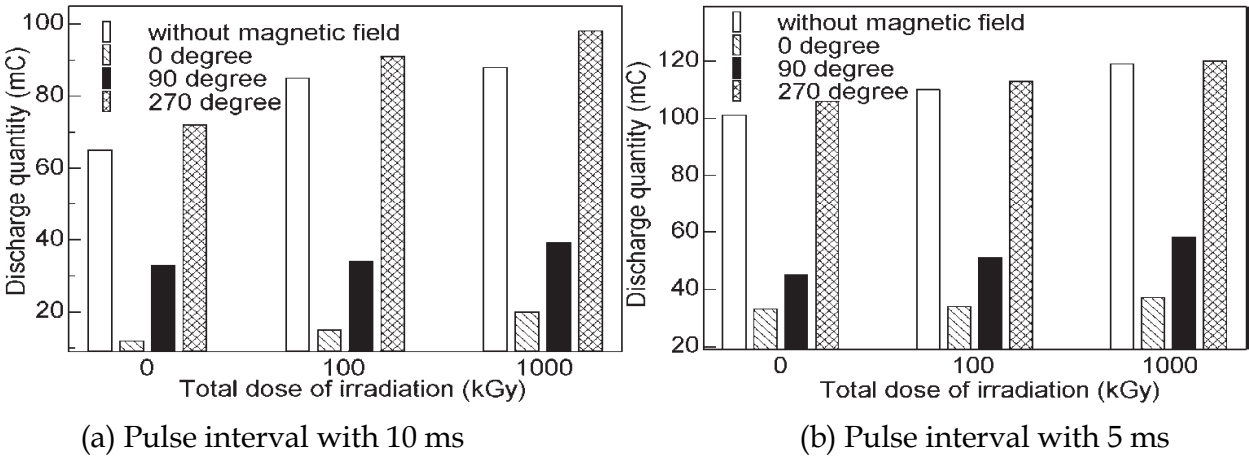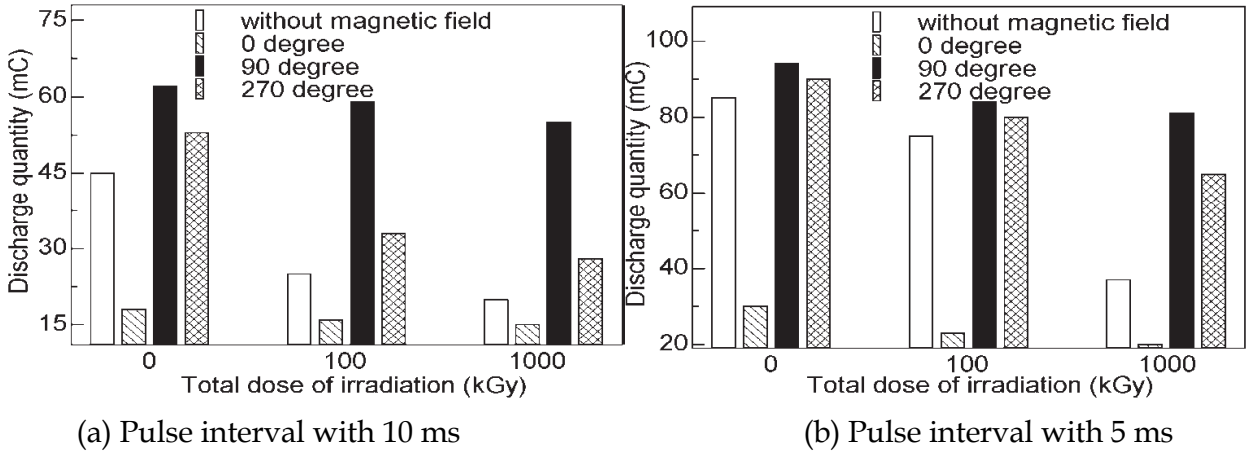
Fig. 23. Relation between the discharge quantity and the total dose of irradiation with PET

Conversely, with the relative angle of 270 degrees, the electromagnetic force is downward to the sample surface, the electron collision is increased on the sample surface and the discharge quantity increases. The electromagnetic force is upward to the sample surface

with the relative angle of 90 degrees, which makes the electrons pass the sample surface more easily. As a result, the electromagnetic force is upward to the sample surface and the discharge quantity is larger for PBN and PET. For PBT, the tacking failure is mostly decided by the carbonization. The electrons upward to the sample surface inhibit the carbonization process, and the discharge quantity decreases.

The photographs of sample surface after tracking failure with the pulse interval of 10 ms at the relative angles of 0, 90 and 270 degrees are shown in Tables 4, 5 and 6. The color of sample surface is gradually dark according to the total dose of gamma-ray irradiation. The repetitive discharge occurs before tracking failure and the quantity of decomposition carbon increases in the area close to the electrodes. As shown in the Tables, the surface is carbonized by the heat energy from the discharge. With the high repetition rate of discharges, there is a high increase of discharge quantity that affects the carbonization progress and the situation of carbon deposition. When the intensive discharge lasts for a period of time, the surface is carbonized first to the area close to the electrodes. Due to the conductive nature of carbonization products, they act as the extension of the electrodes. After a sufficiently long time, the carbonized conduction path is finally formed over the short-circuited path.
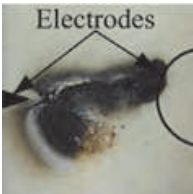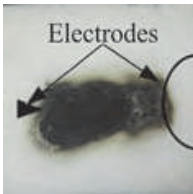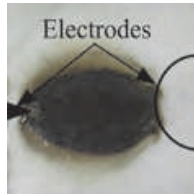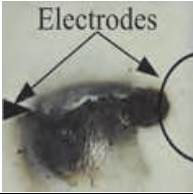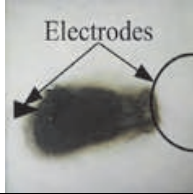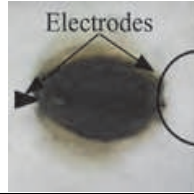
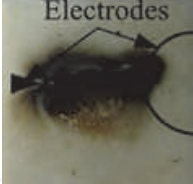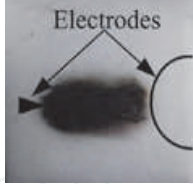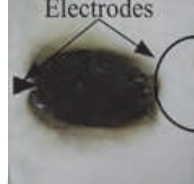| Dosage	0	90	270
0kGy			
100kGy			
1000kGy			

Table 4. Sample surface after the carbonization with PBN

The features of tracking failure phenomenon are different among PBN, PET and PBT. The carbonized area decreases with PBN and PET, but increases with PBT with increasing the total dose of irradiation. By comparing the cases of 0, 90 and 270 degrees, the features of tracking failure phenomenon are different. The carbonization path is downside to the sample surface with the relative angle of 0 degree. The carbonization path of 90 degrees is narrow and the carbonization path of 270 degrees was almost circular.

The oxidation reaction, which takes place on the electrodes, is an exothermic reaction initiated by scintillation discharge and enhanced the intensity of the discharge. When the relative angle is 0 degree, the electromagnetic force is parallel to the sample surface, which makes the electrons deviate to the surface of the samples. The decomposition carbon separates out by the energy of electrons. As a result, the carbonization path is downside to the sample surface. When the relative angle is 90 degrees, the electromagnetic force is

upward to the sample surface. There is a decrease in collision frequency of the electrons and the sample surface. The discharge energy is smaller and the carbonization progress is difficult. As a result, the growth of carbonization path is inhibited. The shape is narrow and the carbonized area decreases. When the relative angle is 270 degrees, the electromagnetic force is downward to the sample surface. The electron collision is increased on the sample surface and the discharge energy increases. The carbonization growth is easier and the carbonization paths extend more easily. Accordingly, the carbonization path of 90 degrees is almost circular and the carbonized area increases. The carbonized area of 0 degree is biggest and that of 90 degrees is smallest for PBT and PET, but that of 270 degrees is biggest for PBN. For PBN, the development of carbonization is not complete.
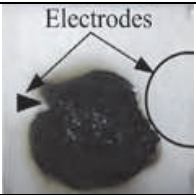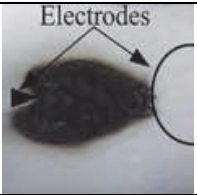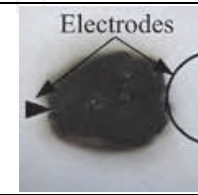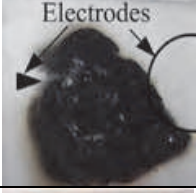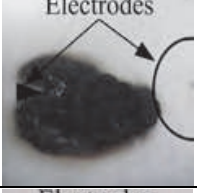
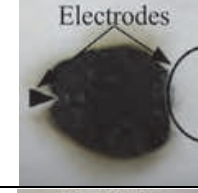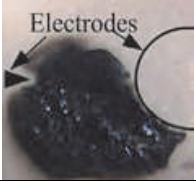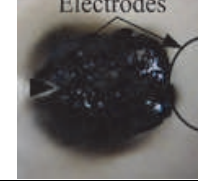| Dosage	0	90	270
0kGy			
100kGy			
1000kGy			

Table 5. Sample surface after the carbonization with PBT
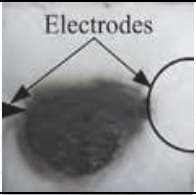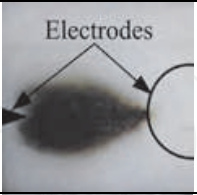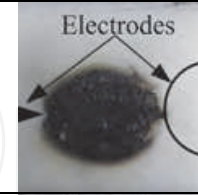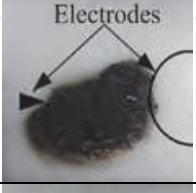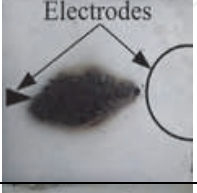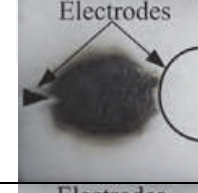
Dosage	0	90	270
0kGy			
100kGy			
1000kGy			

Table 6. Sample surface after the carbonization with PET

7. Recurrence plots of discharge current in tracking test

Fig. 24 shows the variation of RP topological structure at different states of discharge process for PBT with 100 kGy dosage at 350 V. Fig. 24a shows the initial state of the test. The sparse recurrence point density indicates a low correlation between embedding vectors $X(i)$ and $X(j)$ in m-dimension space. It also shows that the beginning of the tracking test is an u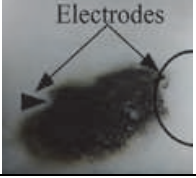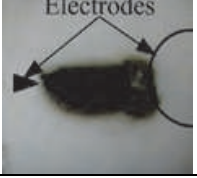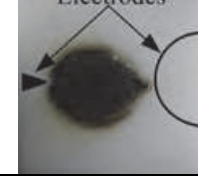nstable state due to incomplete evolution of the solution on the sample surface, which results in leakage currents and the occurrence of discharge between tiny droplets. Fig. 24b shows the middle state of discharge process. It shows a stationary state which is manifested in a comparatively regular and dense recurrence point distribution. This indicates that some embedding vectors are the same or very similar to other embedding vectors in m-dimension phase space. It can also be considered to correspond to the state of the discharge process when the discharge mainly occurs between the deposited carbon patches and the process is in a comparatively stationary state. Fig. 24c shows the state of the discharge process just before tracking failure. The topological structure is completely different from that shown in Fig. 24a or Fig.24b. From vector $X(0)$ to $X(4500)$, the main segments of the map indicates a stationary state with an exception of a short unstable one that produced the white cross blank area between vector $X(4500)$ to $X(5000)$. This also indicates that the discharge state will switch to the next state which indicates the imminence of the breakdown.
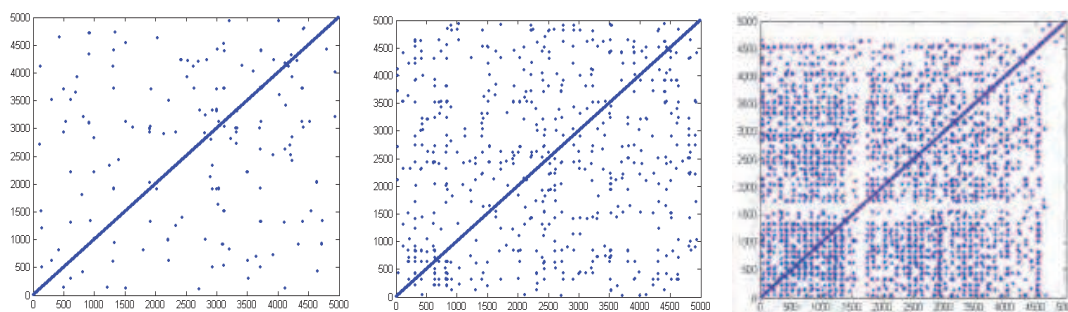
(a) At the initial stage   (b) In the middle stage   (c) Just before tracking failure

Fig. 24. Recurrence plots for three different stages in the discharge

The variations of topological structures for PBT at 375 V and for PET at 450 V with the increase of irradiation dosage are shown in Fig. 25. In Fig. 25a, the point density of PBT decreases with the increase of the irradiation dosage. By contrast, in Fig. 25b, the point density of PET increases with the increase of irradiation dosage. The two opposite tendencies of recurrence point density indicate that gamma-ray irradiation has completely different effects on the resistance to tracking for PBT and PET.

In order to understand the underlying mechanism of the confusing discharge process, the RP has been made to evaluate the tracking resistance. Fig. 26 shows the RPs of the discharge resulting from the application of 400V with M-PC. The structures of the irradiated samples are different from that of the unirradiated ones and there are bigger white space segments in the plot of the unirradiated sample. The white space segments stand for inflation processes due to the high amplitude transients, which indicate that there are high amplitude changes in the discharge currents of unirradiated sample and the fluctuation is more strenuous than that of the total dose at 100 kGy and 1000 kGy. The discharge processes change due to the

increase of the irradiation. There is almost no obvious difference in the white space, but the point density increases with the irradiation dosage from 100 kGy to 1000 kGy, which indicates that the embedding vectors are very similar to the neighbor vectors. It is suggested that the discharge process at 1000 kGy is more intensive than that at 100 kGy. This results of is related to the phenomenon in which as total dose increases, the tracking resistance of M-PC is improved compared with unirradiated samples and the tracking resistance decreases with the total dose from 100 kGy to 1000 kGy.

| (i) 0 kGy | (ii) 100 kGy | (iii) 1 MGy |

(a) PBT, the applied voltage was 375 V

| (i) 0 kGy | (ii) 100 kGy | (iii) 1 MGy |

(b) PET, the applied voltage was 450 V
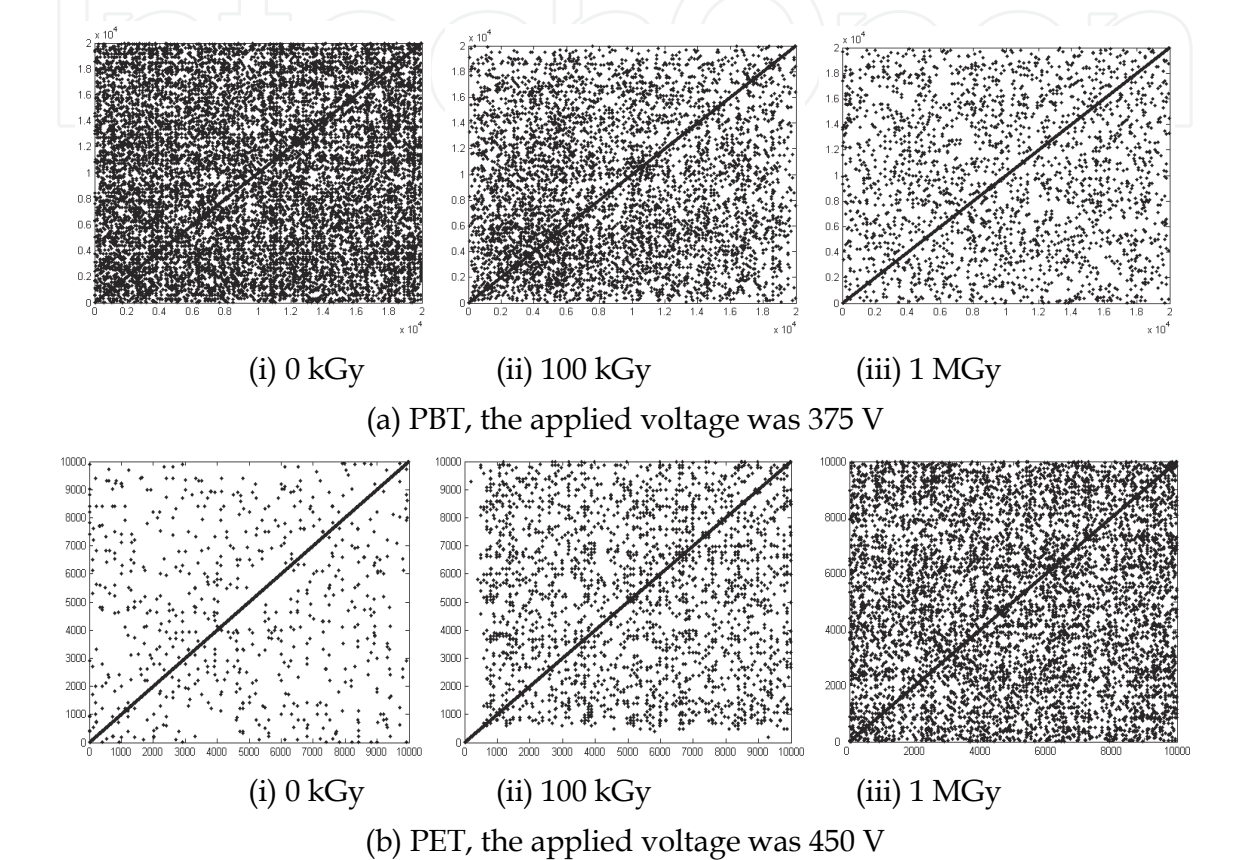
Fig. 25. Relation between the RP and the dosage of gamma-ray irradiation

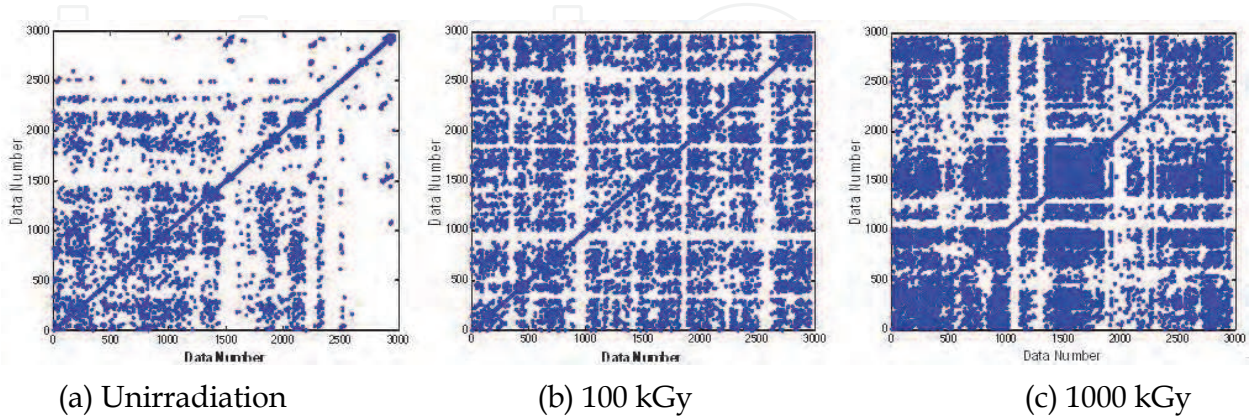| (a) Unirradiation | (b) 100 kGy | (c) 1000 kGy |

Fig. 26. RPs of discharge currents with M-PC

Fig. 27 shows the recurrence plots of the discharge currents with PE at 400V. The point density and the white spaces all decrease with the increase of the irradiation dosage, which

indicates the intensity and the high amplitude transients of the discharge currents decrease. These results are related to the phenomenon where as total dose increases the tracking resistance of PE is improved. It is also confirmed that the point density and the white space are directly related to the features of the discharge process.

Fig. 28 shows the relationship between the quantitative indicators and the gamma-ray radiation. The RR increases for M-PC but decreases for PE with the increase of the radiation dosage, while the DET of PE increases. The DET of M-PC increases with the total dose from 0 kGy to 100 kGy and decreases from 100 kGy to 1000 kGy. For M-PC, the increasing of RR indicates that the intensity of the discharge currents increases. The DET increases first and then decreases indicate that the deterministic ingredients of the discharge currents increases first and then decreases with increasing the radiation dosage. For PE, the decreasing of RR and the decreasing of DET indicate that the discharge is more stable and regular.

(a) Unirradiation   (b) 100 kGy   (c) 1000 kGy
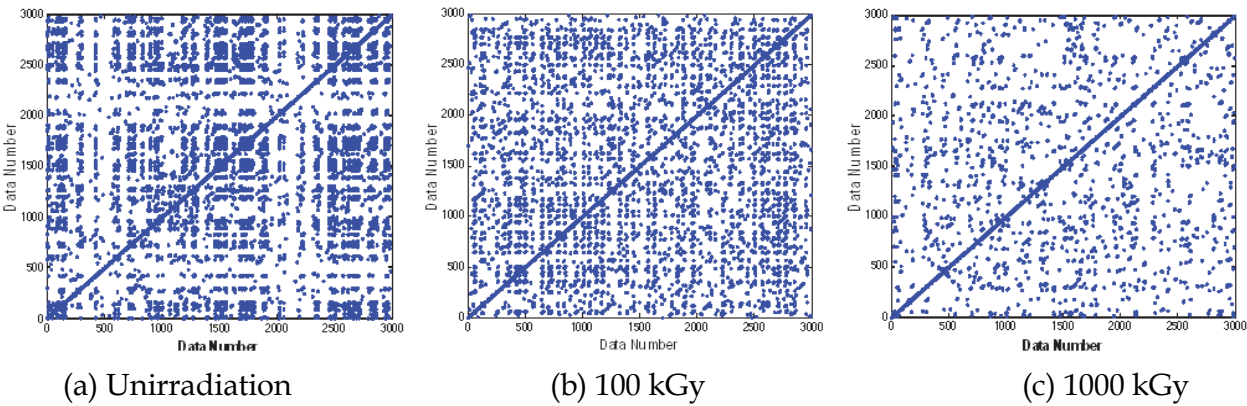
Fig. 27. RPs of discharge currents with PE

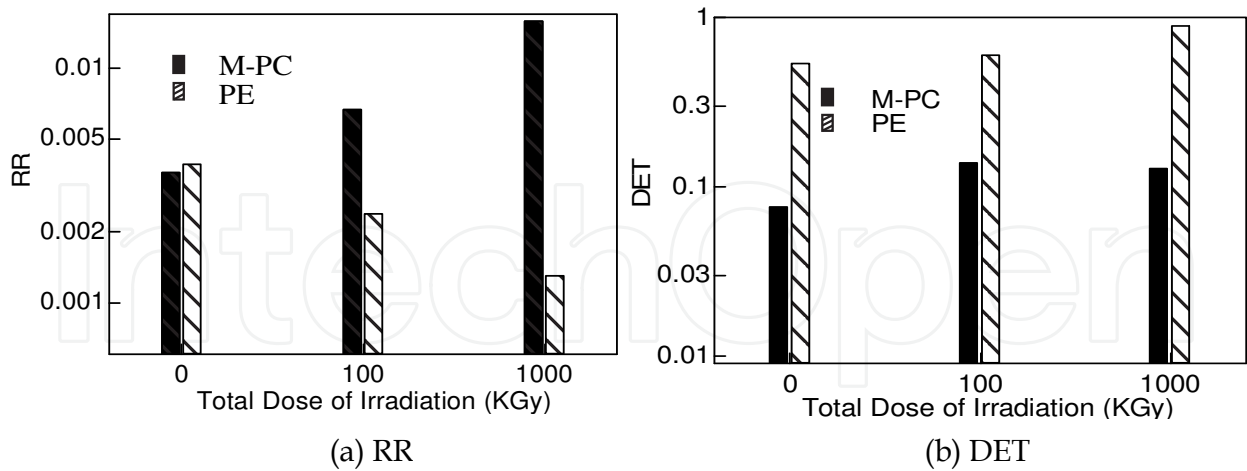(a) RR   (b) DET

Fig. 28. Relationship between the quantitative indicators and the gamma-ray radiation

8. Conclusions

The effects of gamma-ray irradiation on dielectric breakdown have been studied by applying a dc pulse voltage. The experiments reveal that the dielectric properties of polybutylene polymers are obviously changed by the gamma-ray irradiation. With the

increase of the total dose of irradiation, the time to dielectric breakdown increases with PBN, but decreases with PBT, the discharge quantity decreases with PBN, but increases with PBT. It indicates that the total dose of irradiation has a converse effect on the discharge quantities. The discharge quantity increases with the decrease of the discharge interval with PBN, but the tendency is opposite with PBT. Time to dielectric breakdown for both PBN and PBT all decreases with the decrease of the discharge interval. The dielectric properties of PBN are improved, but worsened for PBT by gamma-ray irradiation. Dielectric properties are improved by irradiation for polybutylene polymers which contain more combined phenyls in the main chain.

The tracking resistance of gamma-ray irradiated polycarbonate mixed with polyethylene by use of IEC 60112 is studied. With the increase of the total dose of Gamma-ray irradiation, both the erosion depth and weight loss are smaller than that of unirradiated samples, which show the tracking resistance is improved. Both the erosion depth and weight loss decrease with the total dose increasing from 0 kGy to 100 kGy, but increase from 100 kGy to 1000 kGy. There is a threshold value for the tracking resistance near 100 kGy. When the total dose exceeds the threshold value, the tracking resistance begins to decrease.

The effect of gamma-ray irradiation on tracking failure of PBN, PBT and PET by applying a dc pulse voltage under reduce atmospheric pressure is studied. With the decrease of the atmospheric pressure, the time to tracking failure and the discharge quantity all increase with PBN and PET. The time to tracking failure is delayed with PBT, and the discharge quantity decreases with decreasing the atmospheric pressure. It is found that the irradiation and reduced pressure stress response of PBN, PET and PBT are variable: the resistance to tracking failure of PBN and PET are improved, but worsened for PBT by gamma-ray irradiation and the resistance to tracking failure of all them are influenced obviously by the reduced atmospheric pressure.

The effects of gamma-ray irradiation and magnetic field on tracking failure of PBN, PBT and PET by applying a HV pulse voltage are studied. Under the magnetic field, the time to tracking failure of all the samples are delayed with the relative angles of 0 and 90 degrees, but decrease with the relative angle of 270 degrees, the discharge quantity of the samples increase with the relative angles of 90 and 270 degrees, but decrease with the relative angle of 0 degree. In addition, it decreases with the relative angle of 90 degrees for PBT. The carbonization path is downside to the sample surface with the relative angle of 0 degree. The carbonization path of 90 degrees is narrow and the carbonization path of 270 degrees is almost circular. The carbonized area decreases from 0 to 90 degrees and then increases from 90 to 270 degrees for all the samples. The carbonized area of 0 degree is biggest and that of 90 degrees is smallest for PBT and PET, but that of 270 degrees is biggest for PBN.

Recurrence plots are employed to evaluate the resistance to tracking of polymer samples with gamma-ray irradiation. The results obtained show that it is useful to visualize the state of discharge process. The recurrence plots confirmed by the CTI can successfully indicate the resistance to tracking of polymer material with gamma-ray irradiation. Recurrence plots can serve as a visual method to identify different dynamic systems and reveal time-domain features of the discharge. The recurrence point density decreases with the increase of the dosage of irradiation for PBT. This can be explained by the resistance to tracking of PBT decreasing by increasing the dosage of gamma-ray irradiation. On the other hand, with the increase of the dosage of gamma-ray irradiation, the recurrence point density increases for PET, indicating less complex discharge process. This can be explained due to cross-linking

being the predominant process through irradiation. The improvement in mechanical and thermal performance through irradiation leads to less easy carbonization. In the dc tracking test of gamma-ray irradiated polycarbonate, the topological structures of the recurrence plots with the irradiated samples are different from that of the unirradiated sample. There are bigger white space segments, which indicate that there are high amplitude transients in the discharge currents of unirradiated sample. The recurrence point density increases from 100 kGy to 1000 kGy, which suggests that the discharge processes of the total dose at 1000 kGy irradiated samples is more intensive than that at 100 kGy.

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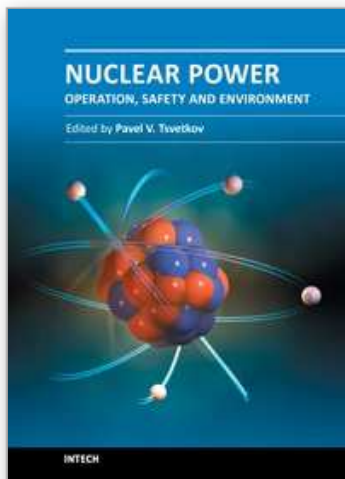
Nuclear Power - Operation, Safety and Environment

Edited by Dr. Pavel Tsvetkov

Today's nuclear reactors are safe and highly efficient energy systems that offer electricity and a multitude of co-generation energy products ranging from potable water to heat for industrial applications. At the same time, catastrophic earthquake and tsunami events in Japan resulted in the nuclear accident that forced us to rethink our approach to nuclear safety, design requirements and facilitated growing interests in advanced nuclear energy systems, next generation nuclear reactors, which are inherently capable to withstand natural disasters and avoid catastrophic consequences without any environmental impact. This book is one in a series of books on nuclear power published by InTech. Under the single-volume cover, we put together such topics as operation, safety, environment and radiation effects. The book is not offering a comprehensive coverage of the material in each area. Instead, selected themes are highlighted by authors of individual chapters representing contemporary interests worldwide. With all diversity of topics in 16 chapters, the integrated system analysis approach of nuclear power operation, safety and environment is the common thread. The goal of the book is to bring nuclear power to our readers as one of the promising energy sources that has a unique potential to meet energy demands with minimized environmental impact, near-zero carbon footprint, and competitive economics via robust potential applications. The book targets everyone as its potential readership groups - students, researchers and practitioners - who are interested to learn about nuclear power.

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