# Influences of Plasma Treatment and $^{60}$Co γ-Ray Radiation on the Over-Voltage Positive Temperature Coefficient of High Density Polyethylene/Carbon Black Nano Composites

*Chia-I Liu,*[1] *Ching-Shan Tsai,*[1] *Keng-Yu Tsao,*[1] *Kan-Nan Chen,*[2] *Jen-Taut Yeh,*[3]
*Chi-Yuan Huang**[1]

**Summary:** Argon plasma-pretreated high-density polyethylene (PHDPE) was blended with the conductive nano carbon black (CB) and inorganic flame retardant (magnesium hydroxide, $Mg(OH)_2$) was added. Influences of the CB content, plasma treatment time, plasma treatment power, and dosage of $^{60}$Co γ-ray radiation on positive temperature coefficient (PTC) behaviors of composite were studied. In addition, the over-voltage resistance behavior of the composites was also investigated. The free radical of PHDPE was measured by 1,1-diphenyl-2-picryhydrazyl (DPPH) method. The gel contents of composite were measured by solvent extraction method. The results showed that the room-temperature volume resistivity and PTC effect of composite were improved significantly with plasma treatment. The PHDPE composite with $^{60}$Co γ-ray radiation eliminated the negative temperature coefficient (NTC) effect in the composites effectively, and the PTC intensity of composite was increased. With increasing of radiation dosage from 20 Mrads to 80 Mrads, the gel content of composites increased up to 83.84% and the response temperature of composites shifted to low temperature (127.5 °C to 114.8 °C). In this work, the composites also successfully passed the over-voltage resistance test, and possessed good reproducibility.

**Keywords:** carbon black; cross-linking; high-density polyethylene; plasma treatment; positive temperature coefficient

## Introduction

Conductive polymer composites were filled with conductive particles to exhibit a positive temperature coefficient (PTC) effect.[1] The PTC phenomenon in polymeric materials was originally discovered by Frydman in 1945,[2] and the theory was reported by Kohler in 1966.[3] The drastic resistivity increase in the temperature range closed to the melting point ($T_m$) of semi-crystalline polymer when the filler concentration achieved a high value.[4] The resistivity had a maximum peak around $T_m$ of polymer, and then it decreased quickly as the temperature increased. The phenomenon was referred to as the negative temperature coefficient (NTC). The cross-linkings of radiative and chemical were both proved to be an effective ways to reduce dramatically the movement of the CB at high temperature and eliminate the NTC effect.[5,6] The plasma treatment was mainly applied in the research of the surface modification of solid materials such as cross-linking, wettability, and adhesion.[7]

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[1] Department of Materials Engineering, Tatung University, No.40, Chung-Shan N. Rd., 3rd Sec., Taipei 104, Taiwan
E-mail: cyhuang@ttu.edu.tw
[2] Department of Chemistry, Tamkang University, No.151, Yingzhuan Rd., Danshui Town, Taipei County 251, Taiwan
[3] Department of Polymer Engineering, National Taiwan University of Science and Technology, No. 43, Sec.4, Keelung Rd., Taipei City 106, Taiwan

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HDPE was treated by plasma to form the uniformly free radical sites on the existing polymer matrix. Finally, the reaction between radicals formed cross-linking chains. In this study, we investigated that the influence of cross-linking by plasma treatment, and dosage of $^{60}$Co $\gamma$-ray radiation on the PTC character of composites. Finally, the structural stability of composites after over-voltage resistance test was also studied.

## Experimental Part

### Materials

The matrix was high-density polyethylene (HDPE, LH606; Taiwan Polyethylene Co., Ltd.). Its density was 0.962 g/cm$^3$ and melting flow index (MFI) was 6 g/10 min. The carbon black (CB) with the commercial name MONARCH 120 was provided by Cabot Corp., USA. It was a lamp type CB which have a particle size of 75 nm, a BET surface area of 25 square meters per gm, and a DBP oil absorption of 64 cc/100 gm. Magnesium hydroxide [$Mg(OH)_2$], SH-2549L with 10 $\mu$m of the average particle size was used as an inorganic flame retardant (Showa Chemical Co., Ltd., Japan). The anti-oxidant was Irganox 1010 (pentaerthrityl tetrakis[3-(3,5-di-ter-butyl-4-hydroxyphenyl) propionate; CIBA GEIGY).

### Argon Plasma Treatment

The self-designed rotary glow discharge reactor used in this study, and was described in our previous study.[8,9] HDPE particles were put into the rotating inner cylinder, and subjected to have an Argon plasma treatment. Before the plasma started, the pressure of inside chamber was reduced to 150 mTorr by vacuum pump, and then the pure Argon (Ar, 99.999%) gas was introduced into it that maintained at 250 ± 2 mTorr by adjusting of micro-throttle valve on the Ar-inlet tube. When the pressure was stable, RF powers generator (13.56 MHz) operated in various plasma-treated conditions, respectively. PHDPE particles were exposed in air for 10 min in order to generate the peroxide.[10] Because the plasma-treated HDPE, the free radicals formed on the surface of HDPE, and it would cause chemical reaction and form hydrogen peroxide as soon as contact to the air.

### Free Radical Measurement of PHDPE by DPPH Radical Method[11]

PHDPE particles were put in a $1.0 \times 10^{-4}\,\mathrm{mol \cdot L^{-1}}$ deaerated benzene solution of 1,l-diphenyl-2-picrylhydrazyl (DPPH), and kept at 70 °C for 6 h to decompose the formation of peroxides on the HDPE particles surfaces. The DPPH molecules consumed were measured from the difference in transmittance of UV-VIS Spectrophotometer JASCO V-560 at 520 nm between the virgin and PHDPE particles. The absorption coefficient of DPPH at 520 nm was $1.08 \times 10^4\,\mathrm{L\,mol^{-1}\,cm^{-1}}$.

### Composite Preparation

Plasma treated HDPE, CB, and other additives were fabricated by melt blending in a Brabender Plasti-Corder Torque Reometer PLE-330 (Brabender, Germany) for 10 min at 180 °C. The compounding process of composites was compressed into 2 mm thick sheet by hot-compression-molded at 180 °C under the pressure of 10 MPa for 6 min and then the specimens were cooled down with water cooling system. In order to reduce the contact resistance, the copper foil was served as electrode (SD 100 3 OZ (A); produced by Co-Tech Copper Foil Corporation) that was laminated to the melt-pressed slab under heat and pressure. The specimens were cut about $40 \times 40 \times 2\,\mathrm{mm}^3$ in size. To reduce the residual stress of specimens, we proceeded with the annealing treatment of specimens. The specimens put into oven at 130 °C for 1 h and then natural cooling to room temperature. The abbreviations of samples were denoted in the Table 1.

### $^{60}$Co $\gamma$-Ray Radiation

In order to increase the cross-linking degree of samples, the samples were irradiated by $^{60}$Co $\gamma$-ray in air at room temperature. The irradiated dosages were 20 Mrads, 40 Mrads, and 80 Mrads, respectively. All of

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***Table 1.***
The abbreviations of samples.

| Sample | CB (wt%) | $Mg(OH)_2$ (wt%) | Plasma treatment conditions | | Dosages of radiation (Mrad) |
|---|---|---|---|---|---|
| | | | Power (W) | Time (min) | |
| H(0)C25M28 | 25.0 | 28.0 | 0.0 | 0.0 | 0.0 |
| H(0)C30M28 | 30.0 | 28.0 | 0.0 | 0.0 | 0.0 |
| H(0)C33.7M28 | 33.7 | 28.0 | 0.0 | 0.0 | 0.0 |
| H(0)C37.1M28 | 37.1 | 28.0 | 0.0 | 0.0 | 0.0 |
| H(0)C40M28 | 40.0 | 28.0 | 0.0 | 0.0 | 0.0 |
| H(0)C43M28 | 43.0 | 28.0 | 0.0 | 0.0 | 0.0 |
| H(0)C45M28 | 45.0 | 28.0 | 0.0 | 0.0 | 0.0 |
| PH(201)C33.7M28 | 33.7 | 28.0 | 20.0 | 1.0 | 0.0 |
| PH(203)C33.7M28 | 33.7 | 28.0 | 20.0 | 3.0 | 0.0 |
| PH(207)C33.7M28 | 33.7 | 28.0 | 20.0 | 7.0 | 0.0 |
| PH(401)C33.7M28 | 33.7 | 28.0 | 40.0 | 1.0 | 0.0 |
| PH(403)C33.7M28 | 33.7 | 28.0 | 40.0 | 3.0 | 0.0 |
| PH(407)C33.7M28 | 33.7 | 28.0 | 40.0 | 7.0 | 0.0 |
| PH(601)C33.7M28 | 33.7 | 28.0 | 60.0 | 1.0 | 0.0 |
| PH(603)C33.7M28 | 33.7 | 28.0 | 60.0 | 3.0 | 0.0 |
| PH(607)C33.7M28 | 33.7 | 28.0 | 60.0 | 7.0 | 0.0 |
| PH(201)C33.7M28-20 | 33.7 | 28.0 | 20.0 | 1.0 | 20.0 |
| PH(201)C33.7M28-40 | 33.7 | 28.0 | 20.0 | 1.0 | 40.0 |
| PH(201)C33.7M28-80 | 33.7 | 28.0 | 20.0 | 1.0 | 80.0 |

the samples were held for 1 day after radiation cross-linking to eliminate post-radiation effect.

### Electric Resistance Measurement

In order to measure the resistance of composites at progressively elevated temperature, we connected a computerized system that comprising a computer, a multimeter (KELTHLEY 7002 switch system and 2400 source meter), and a programmable oven (DELTA 9039). The resistance of composites along the thickness direction as a function of temperature was measured at a heating rate 4 °C/min. To prepare the test specimens, the two sides of the sample were laminated with copper foil, and cut into a circular form which diameter of sample was 5.0 mm and thickness of sample was 2.0 mm. According to ASTM D 4496, samples were measured the volume resistivity ($\rho_v$) of specimens, and then were calculated by following equation 1:

$$\rho_v = (AR_v/L) \tag{1}$$

where A, $R_v$, and L represent the area of the effective electrode ($cm^2$), measured resistance (ohm), and distance between electrodes (cm), respectively.

### Scanning Electron Microscopy

Scanning electron microscopy (SEM) observations of composites were performed on a JEQL JSM-6300 model. The fractured surface of composites was prepared in liquid nitrogen followed by a coating with a thin layer of gold (coating 3 min). The morphology was determined using an accelerating voltage of 10 kV.

### Gel Content

The percentage of insoluble material (gel) after irradiation was measured with a Soxhlet apparatus. Approximately 200 mg of sample wrapped in nickel mesh was exposed to refluxing xylene until the sample attained constant weight, which usually took 24 h. After that, the sample was dried in a vacuum oven at 90 °C overnight, and the weight was measured. The degree of cross-linking (DOC) was calculated by equation (2):

$$g\,\% = (w_1 - w_c)/(w - w_c) \tag{2}$$

where g% was the gel fraction, $w_1$ was the weight of the sample after refluxing, w was

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the weight of the sample before refluxing, and $w_c$ was the weight of CB.

### Over-Voltage Resistance Test

According to the above results, the composites proceeded the over-voltage resistance test. According to the test standard (YD/T 741-95),[12] the test conditions were voltage of 250 V, electric current of 3 A, and frequency of 10 cycles. The test standard (YD/T 741-95) was specification of Positive Temperature Coefficient (PTC) thermistors for the over-current protection of communications installations. The multiple-channel automated current shock withstanding tester was used to proceed the over-voltage resistance test. The test voltage was set in 250 V, and then adjusted the variable resistances that made the electric current to reach 3 A. In this condition, the samples tested the frequency of 10 times in succession.

### Reproducibility

In addition to over-voltage resistance test, the samples also needed to proceed the reproducibility test. The test condition was heating from room temperature to 140 °C, and then natural cooling was from 140 °C to room temperature. The test process went on the frequency of 5 cycles.

## Results and Discussion

### Effect of Carbon Black Content on PTC Effectiveness

Figure 1 depicted the relationships between room-temperature resistivity and average PTC intensity in composites with various CB contents. At low CB contents, the gap between CB aggregates, where the electrons were transmitted, was very large and the resistivity of the specimen was approximately that of the polymer matrix. As increasing of CB content, CB particles fused into primary aggregates, and Van Der Waals forces caused these aggregates to join in more loosely assembled agglomerates,[13] a percolation threshold or critical content was reached where the resistivity started to decrease abruptly as a function of CB content. This phenomenon was explained by percolation theory.[14] The entire dramatic resistivity decreasing region was called the percolation region. The range had an advantage to ensure high PTC intensity. The PTC intensity, defined as the ratio of resistivity at the peak position ($\rho_{max}$) to that at room temperature ($\rho_{RT}$). P [the average PTC intensity (order) = log ($\rho_{max}$ / $\rho_{RT}$)]. The optimum P was 4.3 order in the percolation threshold concentration (33.7 wt%). Concerning the occurrence

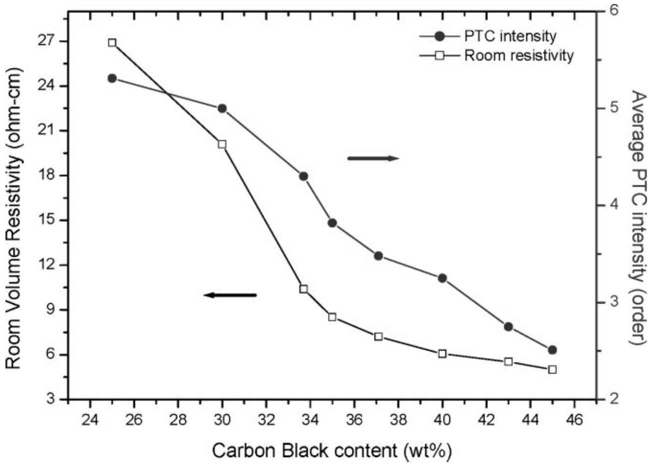

*Figure 1.*

The relationships between room-temperature volume resistivity and average PTC intensity in composites with various CB contents.

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of NTC effect, we suggested that it was related to the re-agglomeration of CB particles at high temperature. It had been shown[15] that the interaction between PE and CB was weak because PE was a non-polar polymer. So the CB particles could re-agglomerated easily in the molten matrix resulting in the formation of a new conductive network. Consequently, the resistivity of HDPE/CB/$Mg(OH)_2$ composite will start to decrease giving rise to the NTC effect. To be directed against this problem, we tried to increase the cross-linking structure of HDPE, and then improved the NTC effect.

### Effect of Plasma Treatment on PTC Effectiveness

During this exposedness in air, most free radicals in the surface region of the HDPE particles must be converted to peroxides. For the determination of peroxides quality, the DPPH method was found to be effective for determining the concentration of peroxides formed. The principle of this method is to measure the consumption of DPPH when the plasma-treated HDPE particles are put in a benzene solution containing a given amount of DPPH and kept at 70 °C to decompose the peroxides. Figure 2 showed the free radical contents calculated from the DPPH consumption as a function of the heating time at 70 °C for the HDPE particles exposed to Ar plasma for various plasma treatment conditions. It could be found that the free radicals were produced on the surface of PHDPE and the content of free radicals was decreased with increasing of plasma treatment time and plasma treatment power. The longer plasma treatment time and larger plasma treatment power did not help the formation of larger amounts of peroxide. It should be noted that low free radical contents at longer plasma treatment time and larger plasma treatment power were due to inter-reaction of the excess peroxides. There was an optimum plasma treatment condition (the plasma treated power of 20 W and the plasma treated time of 1 min) to obtain the maximum free radical content of $1.14 \times 10^{-6}$ mole $g^{-1}$. The result was coincided with the result of Suzuki's study.[11]

Figure 3 depicted the influence of plasma treatment conditions on the gel content of composites. It was obvious that the gel content (%) of PHDPE increased with various plasma treatment conditions. The gel contents (%) of PHDPE were 42.53% ~ 45.02%. The cross-linking of plasma-treated PE was based on free radical mechanism.[15] The free radicals generated on plasma treatment can attack

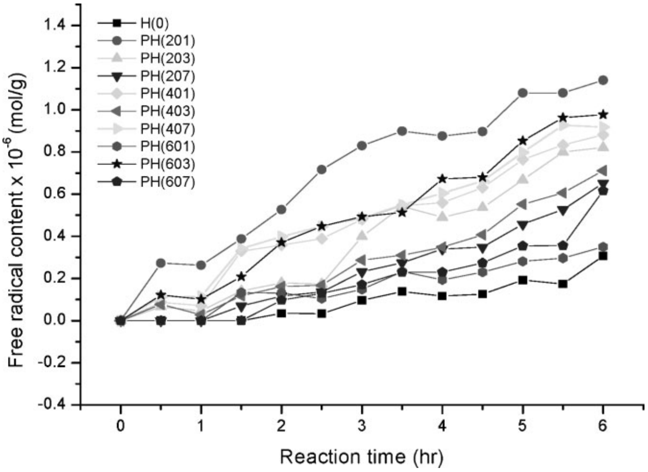

***Figure 2.***
The free radical contents calculated from the DPPH consumption as a function of the heating time at 70 °C for the HDPE particles exposed to Ar plasma for various plasma treatment conditions.

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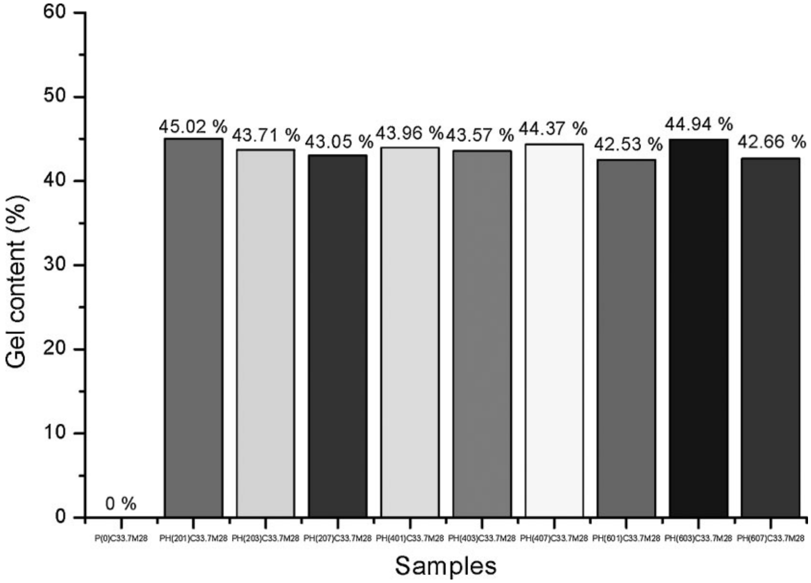

***Figure 3.***
The influence of plasma treatment conditions on the gel content of composites.

to the molten PE free chains and cross-linking of the polymer chains may occur. Figure 3 could be found that the PHDPE (20 W, 1 min) had the highest gel content (45.02%) of all samples. Besides, this result coincided with the result of Figure 2.

Figure 4 showed the volume resistivity of composite with various plasma treatment conditions as a function of temperature. It was found that the room-temperature volume resisitvity of plasma-treated composite was reduced. The free radicals formed on the surface of PHDPE, it will cause chemical reaction and form hydrogen peroxide as soon as contact to the air. The CB dispersion in composite was associated with the free radicals of PHDPE. The CB could be dispersed uniformly and the CB

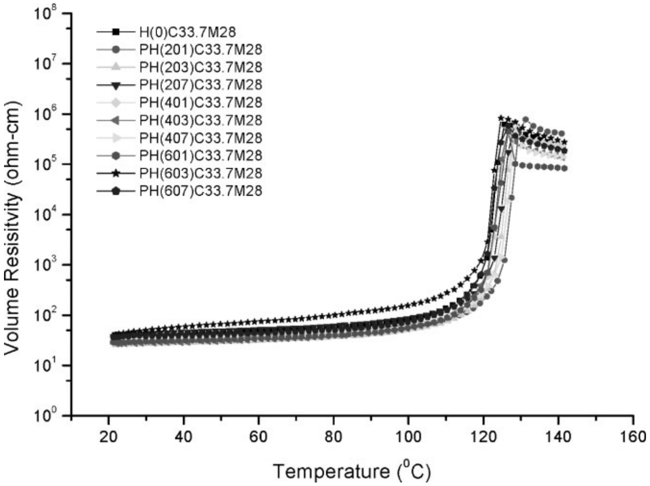

***Figure 4.***
The volume resistivity of composite with various plasma treatment conditions as a function of temperature.

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contact increased. Thus, the plasma-treated composite reduced the room-temperature resistivity. In addition, the HDPE which pretreated with plasma in 20 W for 1 min had the lower room-temperature volume resistivity (32.94 ohm-cm), and the better PTC intensity (4.37 order). The surface of HDPE beads successfully formed cross-linking by the plasma treatment.[16] Cross-linking of composites made CB to fix on network structure, and then the PTC intensity of composite increased. Besides, this result also coincided with the results of Figure 2 and Figure 3.

### Effect of $^{60}$Co $\gamma$-Ray Radiation Dosages on PTC Effectiveness

The strategy of radiation cross-linking was found to eliminate the NTC effect that could be resulted the more stable conductive behavior in those materials.[17,18] Figure 5(a) depicted the volume resistivity temperature behaviors of composites by irradiating of various $^{60}$Co $\gamma$-ray dosages. It was observed significantly that the PTC intensity and NTC effect of composite were obvious improved, especially the NTC effect of composite was completely eliminated. It indicated that the cross-linking density of composite with $^{60}$Co $\gamma$-ray radiation increased obviously, and the rigid cross-linking structure could be limited the move of molecular chains effectively in high temperature. As compared with Figure 5(a), we found the special phenomenon that the response temperature of composite could be shifted to low temperature (127.5 °C to 114.8 °C) with increasing of the $^{60}$Co $\gamma$-ray radiation dosage. It indicated that the molecular chains of composite were broken to form the tiny high cross-linking density molecular chains with increasing of the $^{60}$Co $\gamma$-ray radiation dosage. Figure 6(b) depicted the relationships between average PTC intensity and average NTC intensity of composites by irradiating of various $^{60}$Co $\gamma$-ray dosages. The $^{60}$Co $\gamma$-ray radiation dosage rose from 0 to 20 Mrads, the PTC intensity of composites increased from 4.19 orders of magnitude to 4.58 orders of magnitude. Furthermore, with increasing of $^{60}$Co $\gamma$-ray radiation dosage, the PTC intensity of composites remained unchanged up to 20 Mrads and then decreased. Because the radiation cross-linking of composite restricted the movement of CB particles, so the NTC effect of composites could be eliminated fully. On the other hand, the higher cross-linking density also can limited the expansion of composites, so the PTC intensity of irradiated composites in 40 Mrads ~ 80 Mrads were lower in 20 Mrads. Therefore, the composite with $^{60}$Co $\gamma$-ray radiation dosage of 20 Mrads could be got the best PTC intensity and eliminated the NTC effect.

Figure 6 depicted the SEM photographs of the fractured morphology of composites by irradiating of various $^{60}$Co $\gamma$-ray

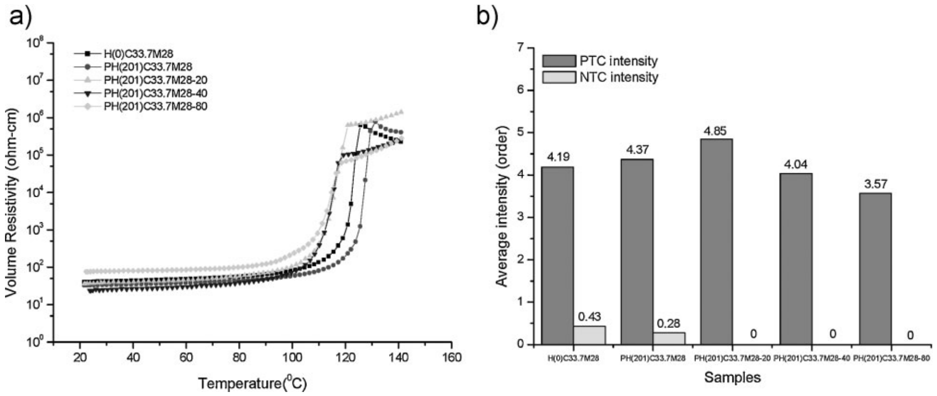

***Figure 5.***
The PH(201)C33.7M28 composites by irradiating of $^{60}$Co $\gamma$-ray dosages. (a) the volume resistivity temperature behaviors; (b) the relationships between average PTC intensity and NTC intensity.

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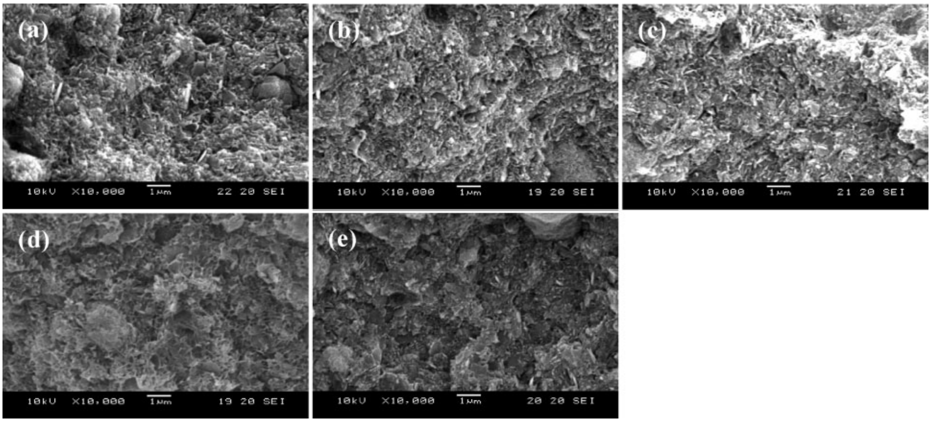

***Figure 6.***
SEM photographs of the fractured morphology of PH(201)C33.7M28 composites by irradiating of $^{60}$Co γ-ray dosages. (a) H(0)C33.7M28 10000x; (b) PH(201)C33.7M28 10000x; (c) PH(201)C33.7M28-20 10000x; (d) PH(201) C33.7M28-40 10000x; (e) PH(201)C33.7M28-80 10000x.

dosages. Figure 6(a) showed the surface structure of composite was inordinate. In Figure 6(b) ~ Figure 6(e), the CB particles which dispersed in the cross-linking networks could be observed because the irradiated composites produced the inseparable cross-linking structure. The size of the cavities of the irradiated composite was close as increasing of irradiated dosages. It suggested that only part of the CB aggregates was bound to the cross-linking networks when the radiation dosage was lower than 20 Mrads. Above the $T_m$ of polymer, the compressed CB particles in the original amorphous region started to migrate to the new amorphous area with the result that the NTC effect was observed. However, with increasing radiation dosage, many more CB particles were bound in the cross-linking networks and prevented the migration of CB particles. Consequently, the NTC effect decreased and the PTC intensity increased (as shown in Figure 5(a)).

Figure 7 depicted the influence of various $^{60}$Co γ-ray radiation dosages on the gel content of composites. It could be

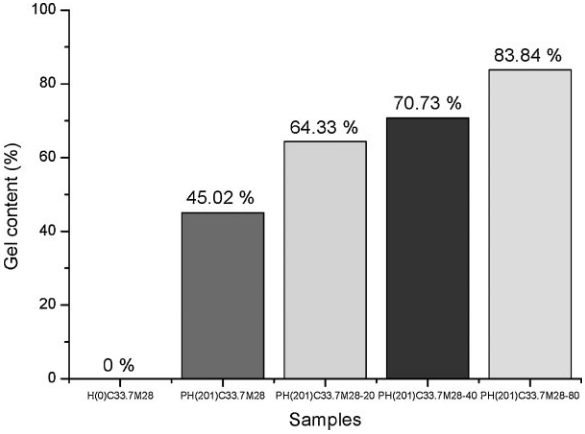

***Figure 7.***
The influence of various irradiated dosages on the gel content of composites.

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***Table 2.***

The results of over-voltage resistance test for PH(201)C33.7M28 composites by irradiating of $^{60}$Co γ-ray dosages.

| Sample | $\rho_{RT}$* (ohm) (before) | $\rho_{RT}$* (ohm) (after) | Pass | PTC intensity (order) | NTC intensity (order) |
|---|---|---|---|---|---|
| PH(201)C33.7M28-20 | 74.9 | 141.1 | ˇ | 4.53 | 0 |
| PH(201)C33.7M28-40 | 154.5 | 180.0 | ˇ | 3.86 | 0 |
| PH(201)C33.7M28-80 | 374.2 | 712.3 | ˇ | 3.31 | 0 |

*$\rho_{RT}$ was the room-temperature resistance value.

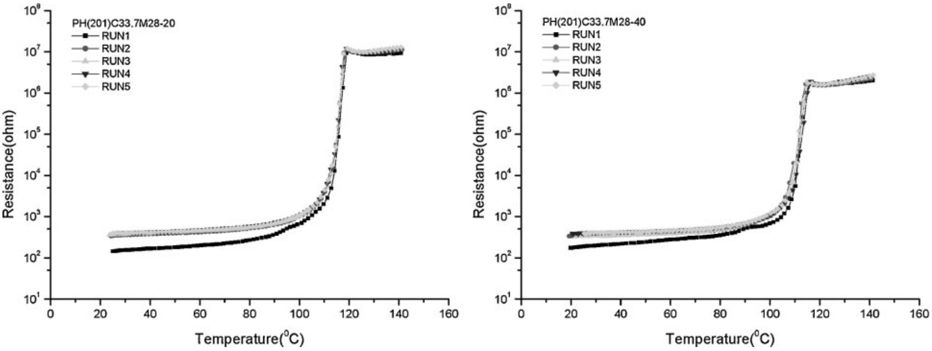

***Figure 8.***

The electrical reproducibility of the passed over-voltage resistance test of PH(201)C33.7M28 composites.

found that the gel content increased up to 83.84% with increasing of irradiated dosages from 20 Mrads to 80 Mrads. When the compound was cross-linking with the irradiation source, a shorter time was required to accomplish the radiation modification of the polymers or its composites at a higher radiation dosage. Therefore, $^{60}$Co γ-ray irradiation induced cross-linking networks of the polymer at a high absorbed dosage limited the movement of CB particles. The composites were irradiated with $^{60}$Co γ-ray above 20.0 Mrads, the PTC/NTC effects of composite were improved, demonstrating that the cross-linking network was completely formed at this time and thus can effectively stabilize the PTC effects of composites.

### Over-Voltage Resistance Test

According to the above results, the composites with irradiating of various $^{60}$Co γ-ray dosages proceeded the over-voltage resistance test. The composites passed the over-voltage resistance test, and the result was showed in Table 2.

### Reproducibility

Figure 8 depicted the reproducibility of the PH(201)C33.7M28 composites as a function of temperature. The trapping of CB within the cross-linking composite not only made the redistribution of CB particles during the expansion of the composite at high temperature, but also took them back substantially to their original positions when the composite was cooled. Figure 8 could be observed the structural stability of composite which did not destroyed in the over-voltage resistance test. The good reproducibility of composite was successfully modified by collocating of plasma treatment, and $^{60}$Co γ-ray radiation.

## Conclusion

There were some advantages by plasma pretreated as listed below: (1) After plasma disposing, the surface of HDPE formed free radical, which improved the dispersion of CB in PHDPE, resulted a lower room temperature resistance. (2) It also affected

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cross-linking degree of PHDPE, and made CB to fix on network structure of PHDPE, and then the intensity of PTC increased. The optimum plasma treatment condition was 20 W, 1 min by the free radical analysis, which the maximum free radicals content was $1.14 \times 10^{-6}$ mol g$^{-1}$. The cross-linking of $^{60}$Co γ-ray radiation was an effective way to eliminate the NTC effect of composites. With increasing of radiation dosage, the gel content of composites increased up to 86.81%, and the response temperature of composites shifted to low temperature (127.5 °C to 114.8 °C). Good reproducibility in PH(201)C33.7M28-20 of composition was proved by thermal cycles of 5 times, and successfully passed the over-voltage resistance test.

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