The bipolar plates are key components of Proton Exchange Membrane Fuel Cells (PEMFC). They are responsible for functions of vital importance to the long-term operation of these electrochemical devices. They play major roles in water and gas management, mechanical stability and electrical performance of fuel cells.1–5

Carbon–polymer (P) composites with either thermoplastic or thermoset matrices are advantageous over metallic materials with regard to corrosion resistance, low weight, and easy productability.6–21 These carbon-polymer composites applied for bipolar plates usually contain the thermoset or thermoplastics as matrix and carbon fiber (CF), graphite (G), expanded graphite (EG), and carbon black (CB), carbon nanotube (CNT) as reinforcement.6–9 These fillers differ with together from the viewpoint of morphology and electrical conductivity. Therefore, it is anticipated that the electrical conductivity of their composites differs from each other.1–20

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The electrical conductivity of composite bipolar plates is the most important properties of in DOE. There are some parameters influencing on the electrical conductivity of composites, such as filler content, filler length, filler aspect ratio, filler size, filler orientation, filler surface energy, matrix surface energy, and so on Ref. 7. Moreover, the body temperature of the fuel cell and passing electricity of different layers of the fuel cell during the operation can change the microstructural and electrical properties of composite layers such as bipolar plates and gas diffusion layer (GDL). It has been established that the passage of electrical conductivity and the high body temperature can influence on electrical conductivity of the composites.21–24 thereby PEMFC performance. The effect of temperature on resistivity either have been reported as negative temperature coefficient of resistivity (NTCR effect) or positive temperature coefficient of resistivity (PTCR effect) in literature.

**The effect of temperature.**—In the case of semiconductors, the electrical conductivity has an Arrhenius relation with temperature.21

\[
\sigma = n_0 \mu_e \exp \left( \frac{-E_g}{2k_B T} \right)
\]

$\mu_e$ and $\mu_p$ are the mobility of electrons and holes and $n_o$, $\varepsilon$, $k$, $e$ are constants. In agreement to the above mentioned, Schuartz et al. in 200022 showed that in the case of P/CB, the resistivity of composites decreases exponentially with temperature.

It should be mentioned that in the thermal-activated events accompanied by nucleation and growth processes, such as solidification of metals, diffusion, and heat-treatment, the reaction usually obey an exponential relation with temperature. In the case of electrical conductivity in composites, the increase of temperature leads to increasing electron mobility that leads to increasing electron jump distance and decreasing mean free path thereby increasing electrical conductivity of composites. However, most of the researches disagree with this result. They concluded that heating the composite up to the $T_g$ or $T_m$ leads to decreasing electrical conductivity or increasing resistivity or PTCR. Afterwards, increasing temperature leads to decreasing resistivity or NTCR.23

PTCR for the first time was reported by Frydman in 194825 for P/CB that by heating the composite toward its melting point faced to a sharp PTCR. Afterwards, Kohler in 196623,26 pointed that PTCR mechanism was a function of the difference of the thermal expansion coefficients between the matrix and the filler, so that the sudden expansion, which took place at the melting point of the polymer matrix, resulted in the break-up of the conducting chains with a consequent anomalous increase in the resistivity. However, this model was not able to explain the NTCR effect (the steep decrease in the resistivity above $T_m$ indicated by some researchers), when the volume of matrix continued to increase with temperature.

After that, in 1971 Ohe and Natio26 for the first time applied the tunneling phenomenon to justify PTCR effect. They assumed that there are a uniform distribution of interparticle gaps between fillers so that the gap width is small enough (lower than 10 nm27 or 30 nm2) to allow intensive tunneling to take place at low temperature. They believed that when the temperature nears melting point ($T_m$), the distribution of the gaps become more random and the interparticle gaps are widened so that electron tunneling cannot appreciably be occurred. This leads to eliminating many conductive paths, thereby rising in the resistivity.26,28 However, in this model the NTCR effect, which usually took place immediately above the melting point of the matrix, also remained unexplained.26

Afterwards, Meyer in 197329 for the first time, related the PTCR to glass transition temperature, crystallinity, and Coefficient of Expansion. Meyer28 suggested that the conductive particles collect only in amorphous regions being separated by crystalline films, which has been assumed to be more conductive than amorphous ones. Thus, he suggested that the suddenly rise in resistivity near $T_m$ could be attributed to the melting of the crystals. He also suggested that the NTCR effect might be related to the immersion of carbon black
particles, which resulted in the formation of new conductive chains of the filler particles. However, the Meyer’s model has not mentioned the effect of crystalline change and the NTCR effect. In addition, it was assumed that the conductive particles are only entered into the amorphous regions. This phenomenon is not reasonable.

As same as Meyer, Klason et al. in 1975 explained the PTCR and NTCR effects by impressing on crystallinity and the changes taking place in the carbon congregation structure with temperature. The model of Voet in 1980 was similar to Kohler’s model that attributed the PTCR effect to the large volume expansion of the polymer in the melting range.

H. Allak in 1993 similar to Meyer, introduce the crystalline and amorphous phases to explain PTCR effect. However, the Allak’s claim is in contrast with Meyer’s model, which supposed that the CBs enters into the crystalline phase and conductivity, creates from the approaching the crystallitles and tunneling the electron from one crystallite to another. Allack pointed that as the temperature of the crystalline polymer approaches Tm, the crystallities start to become amorphous. However, this process is also accompanied by a sudden and large increase in their volume leading to increasing the intergranular gaps. This will produce more resistive current pathways, and may even decrease in their volume leading to increasing the intergranular gaps. The last will produce more resistive current pathways, and may even decrease in their volume leading to increasing the intergranular gaps.

T. Arfin et al. in 2012 along to the Meyer, Allak and others proposed a model that introduce the crystalline and amorphous phases with different filler concentrations. T. Arfin et al. in 2013 predicted that the relation between electrical conductivity of composites with temperature to be an Arrhenius relation. Chien et al. in 2014 believed that the Joule heating process can change the crystal structure of composite. He tested on PAN-carbon fiber structure and found out with increasing current, the structure was converted gradually to another structure by higher orientation factor, while the d-spacing and crystal size were almost constant at different applied currents.

There are some external forces such as electromagnetic field, shearing force, and electric field induced that are frequently used to prepare polymer composites with aligned fillers. These anisotropic oriented paths has easy manipulation in desired directions assumed the preferential directions for the heat and electrical transport. Duhovic et al. in 2014 pointed that an alternating electromagnetic field operating in the kilohertz to megahertz frequency range, are based on induced eddy currents, so that magnetic polarization effects can induce heating phenomena in ferromagnetic and conductive materials.

Secondary agglomeration.—The heat-treatment and joule heating can lead to Secondary agglomeration. McNally et al. stated that the secondary agglomeration occurs in the dispersion of the Nano scale fillers, such as nanotubes. They stated that in in P/CNT system in the state of secondary agglomeration, the nanotube agglomeration is assumed to result in a denser packing of nanotubes inside the agglomerates and correspondingly in a smaller distance between CNT. McNally et al. states that secondary agglomeration of attractive filler particles is assumed to be thermally activated (Brownian filler diffusion in the melt) and can be accelerated by external mechanical (shear-induced agglomeration). Secondary agglomerated structures can have lower percolation thresholds than dispersed structures. It can be concluded that in order to better make the conductive network, a secondary agglomeration with good dispersion and bad distribution of fillers is necessitated.

Based on the literature review, there is some conflict on the results and proposed mechanisms of authors on the polymer based carbon composites. In addition, most of the researches are related to carbon black composites and there are not enough investigation on the composites containing the other types of carbon fillers such as CF, G, EG. Therefore, the effect of body temperature and joule heating effects with regard to the time have been tested in this research. The different composites containing G, EG, and CF as fillers with different filler loading have been considered in this research.

### Experimental

**Materials.—** The resin used for manufacturing composite was phenolic resin, the type of Novalac that containing Hexamethylene tetramine and formaldehyde. This resin was purchased from Resitan Co. This polymer is powder shape, yellow-color with the average powder size about 60 μm and the bulk density of 1.1 g/cm³. The reinforcements are carbon fiber, synthesis graphite, and expanded graphite that their specifications have been brought in Table I. The carbon fiber, graphite, and expandable graphite were purchased from Highborn International Co., Ltd, Merk Co., and Qingdao Yanxin Graphite Products Co., Ltd.

| Filler type | Carbon fiber | Graphite | Expanded graphite |
|-------------|--------------|----------|-------------------|
| Carbon purity (%) | 95 | 99.9% | 99.5 |
| Color, apparent shape | fiber | Gray, flaky powder shape | Silvery-gray to black, Wormy and honeycomb shape |
| Aspect ratio dimension | 410 | 0.1 | |
| relative density (g/cm³) | 7.3 μm Diameter, 3 mm length | μm | μm |
| Electrical conductivity (S/cm) | 100 μm diameter, 300 μm length | |
| Through-plane | 1.9 | 2.2 | 2.2 |
| In-plane | 598 | 6000 | |
| Thermal conductivity (W/mK) | 10.46 | 0.2 | 0.05 |
| Through-plane | 0.03 | 111 | 96 |
| In-plane | 0.120 | 0.05 | |
| Percolation threshold (vol.%) | 1.0 | 0.2 | 0.05 |
| Max. electrical conductivity in 0.7 vol.% of filler (S/cm) | 3800 | weak | Very weak |
| Tensile strength (MPa) | | | |
Composite preparation.—At first the polymer as matrix and CF, EG, and G as reinforcements are weighted. Then, a high-energy mixer mixes those. EG powder is added at the end, because its structure is very fragile; retaining the wormy and honeycomb structure of EG is very important in enhancing electrical conductivity of its composites. The final mixture is poured into the three-part die cavity. The die is heated by an element and is soaked at 200 °C for 45 min in 200 bar to cure the polymer, completely. The pressure is preserved until cool the die under 80 °C (the glassing temperature of the polymer). The produced disk is cut into the dimension: 10 mm × 10 mm × 2.8 mm.

Procedure.—The aim of this research is that the investigation on the effect of “joule heating” (the heat induced by passing the electrical current from composite) and the “heat treatment” (heating the composite samples by soaking in the furnace) on the change of electrical conductivity of different composites. In order to achieve this aim, the research procedure is divided into two parts; the joule heating part and heat-treatment part. In both parts, the samples include of the phenolic resin as the polymer and the different types of fillers with different percentages. The composition of these composites is constant in either the joule heating part or heat-treatment part as follows:

1) Composites reinforced by G: P/20G, P/40G, P/70G
2) Composites reinforced by EG: P/20EG, P/40EG, P/70EG
3) Composites reinforced by CF: P/20CF, P/40CF, P/70CF
4) Final composite: P/45G/10EG/5CF that a layer of carbon fiber paper has been inserted in the middle of composite

Joule heating part procedure.—The sample was inserted between two-polished copper surfaces under the constant pressure 60 N.cm⁻². It means the electrical conductivity of composite was determined under the defined stress and after the inducing the electrical field. All 10 types of composite were tested by three powers (1.5, 5, and 10). Thus, for each composite, three samples were prepared. A medium voltage power source, Dazheng DC power supply ps-305D, is used to produce the power of 1.5, 5, and 10. The resistance of the circuit, thereby the sample resistivity was performed by milliohmmeter Lutron MO-2013. The temperature of samples during joule heating was measured with a contact thermometer. The average temperature of all samples in 1.5, 5, and 10 W was recorded 50 °C, 100 °C and 150 °C, respectively. In some samples, due to extra heating, instead of 5 W, the value of 3 W was used that is equal to 75 °C

Based on the above mentioned, overall, 30 samples were provided to determine the effect of changing a power value on the electrical conductivity of composite. In these 30 samples the measurements were performed at 4 levels by an interval of 1 min. It means, for example P/20G in power 1.5, the measurements were performed in: 0 (without passing electrical current), 1 min, 2 min, and 3 min.

\[ P = R I^2 \] (this is the power produced by the power source apparatus)

\[ R = \frac{V}{I} \] (This is the circuit resistivity measured by milliohmmeter)

P: power, R: Circuit resistance and I: electrical current, σ: electrical conductivity, l: sample thickness, A: the sample surface area

Heat-treatment part procedure.—The heat-treatment test was performed by using of 8 samples. Here, instead of inducing different voltages, soaking the samples in the furnace with a specific temperature influence the electrical conductivity of composite. The temperatures of the furnace were considered 50, 100, and 150 °C. The temperature was selected based on low-temperature and high-temperature PEMFCs. In each sample, the measurements were performed at four temperatures by an interval of 5 min. It means, for example, in the sample of P/20G, the electrical conductivities of the sample were measured after soaking at 25 °C, 50 °C for 5 min, 100 °C for 5 min, and 150 °C for 5 min.

SEM analysis.—in order to consider the topography of composites, scanning electron microscope Philips was used.

TGA-DSC analysis.—the apparatus used for thermal analysis has performed the thermogravimetric analysis (TGA) and differential scanning calorimetric analysis (DSC), simultaneously. This set was STA-1500, rheometric scientific, USA. This analysis specifies the different critical temperatures during the curing of polymers by determining the mass loss, value and varying the heat flow value.

Test station.—The bipolar plate was made of P/45G/10CF/5EG in middle a carbon loth layer, containing the depth and support width equal to 1 mm. The channel type was serpentine, and the gasket material was silicone type. The membrane was made of nafion 112 coated by Pt and Teflon. Before fabricating the electrode layer on the membrane, Nafion 112 (DuPont, DE, USA) membrane was pretreated with H₂O₂ and H₂SO₄ solutions step-by-step to remove the remaining organic and inorganic contaminants. GDL type was carbon cloth. Twenty wt% of Pt/C electrocatalyst (Johnson Matthey, MI, USA) and 5 wt% of Nafion solution were used to make anode and cathode electrode. The prepared membrane electrode assembly has platinum loadings of 0.4 mg.cm⁻². The oxygen and hydrogen used as reactant gases in cathode and anode side contain 99.999% and 99.9% purity, respectively. Humidification percentage was preserved at 95% during the test. Humidification percentage was selected 95%. H₂-inlet-temperature and O₂-inlet-temperature were at 70 °C. The standard deviation of the data is assumed 3%.

Results and Discussion

Based on DOE guideline, bipolar plates in PEM fuel cell should have some properties such as high electrical and thermal conductivity. During operation, due to crossing electricity and body temperature bipolar plates can be exposed to high temperature e.g. 150 °C or upper. Therefore, it can be predicted the conductivity of PEMFC devices change. These researches show that with different types of carbon fillers in P/carbon composite bipolar plates, how the electrical conductivity of composite changes. For this aim, the samples were exposed to the processes of thermal treatment and joule heating. At first, it is better to characterize the effect of heat-treatment and joule heating on the value of electrical conductivity of composites in literature.

In addition to some internal factors affecting electrical conductivity of composites, such as filler loading percentage, filler type, mixing method, etc. There are some external factors affecting on composite conductivity such as joule heating, heat-treatment, delaying time of the process, etc. In this research, some internal and external factors will be discussed: filler type, filler loading percentage as internal factors and joule heating, heat-treatment as external factors. In the joule heating the induced power and the delaying time and in the heat-treatment process the temperature value will be the variables.

The effect of internal factors on the electrical conductivity.—It should be emphasized that for providing electrical conductivity in the composite, in addition to the high carbon purity of fillers, the fillers should have good dispersion and bad distribution. It means that in order to form a conductive network of fillers within a polymer matrix, it is better the fillers to be slightly agglomerated. There are some internal factors that influence on electrical conductivity of composites: the filler electrical conductivity, filler value, filler aspect ratio, the difference value between surface energies of filler and matrix, mixing method, any process influencing on surface morphology such as functionalization process. All the mentioned factors have been the subject of some researches; however, the fact is that hardly can change one of the above factors without changing the others. For example, there is some difference
between the carbon filler types such as graphite, expanded graphite and carbon fiber. The Table I has been indicated some specifications of these fillers. The expanded graphite is the best filler for electrical conductivity of composite, because either the carbon purity of EG is 99.9%, thereby the electrical conductivity of that is high, or the filler morphology is warm like and honey-comb like so that this warm-like shape increase the filler aspect ratio and better making the conductive network. The carbon fiber has a higher aspect ratio, but the low purity of that results in a lower conductivity in the composite in comparison to EG. Graphite which also consist of discrete flakes ranging in size from 50 to 800 micrometers in diameter and 1–50 micrometers so to EG. Graphite which also consists of discrete flakes ranging in size from 50 to 800 micrometers in diameter and 1–50 micrometers thick86 has high carbon purity, however flake-like structure of that make a low aspect ratio. Based on the results of researches, the percolation threshold more depends on the aspect ratio than filler purity86,70. Taipalus71 reported that the increase of CF length leads to decreasing the percolation threshold and increasing the maximum electrical conductivity.

It was reported that the greater the surface-to-volume ratio of the filler particle, the more likely is inter-particle contact.72 The research showed that the increasing aspect ratio in CF leads to increasing the mobile carrier concentration.73 Rosca et al.74 reported that the increasing aspect ratio of CB leads to decreasing percolation threshold and increasing the slope of conductivity/filler curve in P/CB in respect to P/G. of course, the aspect ratio of fillers is not usually as high as the real value that is as a result of the bending and entanglement of fillers.

In addition to the mentioned parameters, the mixing method that determines the kind of shear force on the fillers and can be effective in the distribution, dispersion, and agglomeration percentage of filler substantially influences on the electrical conductivity of composites. Anyway, it is clear that all these parameters are effective on composite conductivity; however, the effect value is not clear and needs to more researches.

Based on the previous works, the curves of composite conductivity via vol.% of fillers were drawn and the results of percolation threshold values and the maximum values of electrical conductivities of composites in about 0.7 vol.% have been reported as was pointed in the table. As it was anticipated CF has the least percolation threshold due to higher aspect ratio (the length of CF was 3 mm here), but its maximum Conductivity is the lowest, because of the low carbon purity and low conductivity of this filler. The expanded graphite either has a low percolation threshold or has the highest conductivity of the maximum point. The warm-like structure of EG increase the aspect ratio and decreases the percolation threshold and the high carbon purity of EG increases the maximum point of the curve of conductivity via vol.%. the more discussion on internal factors have been explained in previous articles.75

The effect of external factors on the composite conductivity.—In addition to internal factors, the external forces such as electrical field, magnetic field, chemical environment, thermal processing can considerably influence on the microstructure and fillers orientation, thereby composite properties. It can be anticipated that during the flowing the electrical field through the composites, within different filler networks a solenoid to be formed, thereby inducing an electromagnetic field and enhancing the filler orientation.

The effect of external forces on electrical conductivity of composites can be performed via the following changes: changing the orientation of fillers, changing dispersion and distribution of fillers, decomposition of polymers between fillers, thereby influencing on tunneling effect.

Optical and SEM images.—Fig. 1 shows the optical micrographs of the carbon structure in carbon fiber (a, b), expanded graphite (c, d), and graphite (e, f), respectively. The (a), (c), and (e) figures have a different cross section with (b), (d), and (f) been specified in the figures. The SEM figures indicate the aspect ratios of P/CF, P/G, P/EG, and P/G/CF/EG. The last composite is the optimum composition of composite bipolar plate attained in another research.5 The left hand side is related to forward view and the right hand side are related to lateral view. It can be seen that there are many differences between forward side and lateral side of P/CF and P/EG. That is as a consequence of the carbon fiber and expanded graphite have a high aspect ratio, thereby are extremely oriented, while the aspect ratio of graphite is near 1. Percolation threshold of the composites in the high oriented structures of fillers are much more than that in the particular structures of fillers.8 Fig. 2 shows the SEM images of P/CF, P/EG, P/G and P/G/EG/CF that verified the above mentioned.
Figure 3. DSC (a) and TGA (b) analyses of pure cured polymer, P/CF and P/EG.

**DSC-TGA analysis.**—Fig. 3 shows the results of DSC and TGA analyses of the pure polymer, P/EG and P/CF and two double-filler composites with different contents of expanded graphite and carbon fiber, respectively. The curves indicate to the reactions occurring during the heating of the some composites. It can be seen that in the cured polymer, the dissociation reactions start sooner than the other ones. Moreover, dissociation reactions, including structure decomposition and atomic decomposition occurring in high temperature, perform at a single step in the P/CF and P/EG while performing at two steps in pure polymer. In addition, it can be observed that the P/EG has the highest decomposition temperature in comparison to other ones.

**Joule effect test.**—Fig. 4, Fig. 5, and Fig. 6 present the results of Joule heating tests, that are related to P/CF, P/EG, P/G, and P/G/EG/CF composites, respectively. The starting points in the different powers in a type of composite Slightly differ. It has resulted from this fact that the composites were produced as a plate and it may the different parts of the plate not to be homogenous to have similar electrical conductivity. There are several notable cases that are followed:

1) The first notable case in the Fig. 4, 5 and 6 is that by enhancing the induced power from 1.5 to 10 W, the slope of the curve increases.

2) The increase in electrical conductivity in part 1 basically is at the limit of the first 60 s and afterwards the slope of the curve decreases.

3) With regard to 1.5, 5, and 10 W are equal to 50°C, 100°C, and 150°C, therefore it can be stated that the increase of temperature due to joule heating process leads to increasing the electrical conductivity of samples (Table II) or shows an NTCR effect.

The above observations can be justified as follows:

There are two Hypotheses can be used for these observations:

1- Indeed, in the composite from the microscopical viewpoint, the fillers cannot adhere together and always there are some space between fillers. In these spaces usually polymer infiltrates. By inducing voltage differential the electrons pass from one particle to another filler. When the space between two fillers is less than 10 nm, tunneling effect occurs and the electrons jump from one particle to another one. Here, the resistance between the particles increases, thereby the sample is heated. As it was mentioned the surface temperature of the samples was recorded by a probe. But it should be emphasized that the surface temperature in joule heating always is less than bulk temperature, because in joule heating the heat is produced from the inside of the sample. It can be predicted that the actual temperatures inside the sample would be much higher than the temperatures ascribed to the powers of 1.5, 5, and 10 (50, 100, and 150, respectively). These temperatures based on Fig. 3 are enough to attain to the dissociation temperature of the phenolic resin (≈480°C) and converting to carbon or other conductive compositions. Probably this dissociation basically performs in the first 60 s. By this theory it can be predicted that by increasing the power, the conductivity increases because this helps to more dissociation. With regard to the results of the samples in Table II, it can be concluded that these samples presented as NTCR effect. This
The conductive pathway formed by conductive particles is a twisted path, so that it can be assumed that a solenoid is formed in the composite and crossing the electron within these solenoid can induce a magnetic field in the composite. This magnetic field can align the fillers in the direction of passing electrons. There are many researches that confirm this effect of magnetic field on fillers such as carbon nanotube. The other effect of magnetic field is the production of the secondary agglomeration phenomenon that can enhance
Table II. The changes of conductivity of samples during the joule heating process.

| Power | Sample   | P/CF40 | P/CF70 | P/EG20 | P/EG 40 | P/G40 | P/G70 | P/G/EG/CF |
|-------|----------|--------|--------|--------|---------|-------|-------|-----------|
| 1.5 W | ≈ 50° C  | 0.0068 | 0.027  | 0.0175 | 0.2115  | 0.0033| 0.066 | 0.037     |
| 3 W   | ≈ 75° C  |        |        |        | 0.343   |       |       |           |
| 5 W   | ≈ 100° C | 0.0709 | 0.138  | 0.075  |         | 0.010 |       | 0.028     |
| 10 W  | ≈ 150° C | 0.125  | 0.1956 | 0.058  | 3.958E-05|       |       | 0.204     |

The electrical conductivity of composite. There are many researches related to secondary agglomeration in the composites.

The heat-treatment test.—Figs. 7a–7d shows the thermal treatment test of the P/CF, P/EG, P/G, and P/G/EG/CF composites, respectively. Based on the curves, the temperature has a different effect on each composite. In the P/40CF composite, there is a PTCR effect during heating, while for P/70CF there is no considerable effect by heating. In P/70EG and P/40EG have an inverse effect and by increasing the temperature, a NTCR can be seen, while in P/20EG, the temperature has no considerable effect. The behavior of P/70G is as similar to P/40CF and shows a PTCR effect, while the temperature has not significant effect on P/40G. In the P/G/CF/EG the behavior is NTCR.

In the joule heating process two hypotheses have been introduced to justify the NTCR. However, in the case of P/G and P/CF the PTCR effect and in the case of P/EG and P/G/C/EG the NTCR effect has been observed. These facts can be justified as follows: as it was stated there are some polymer branches among filler particles such as CF and G. Therefore, by increasing temperature the branches expand and leads to getting away the fillers from each other. But why this temperature cannot dissociate the polymer located among the fillers or induce a magnetic field as joule heating process. The response is that unlike the joule heating, in the heat-treatment process the surface of the sample is hotter than the central part. Therefore, the central part of the sample does not reach to the temperature upper than 150° C. This temperature is not even near the dissociation temperature of polymer (480° C). In addition, in the heat-treatment process, there is not any ΔV during the process. Therefore, the magnetic field cannot be formed. Heating of the sample from 50° C to 150° C only can lead to expanding the polymer branches, thereby providing a PTCR effect.

However, in the case of P/EG that the NTCR has been observed, it can be stated that this fact is related to the inherent nature of expanded graphite. Natural graphite or graphite flake is oxidized by H₂SO₄ or other acids to exfoliate the layers and intercalate some of the organic groups such as OH⁻, S²⁻, ...⁸⁶ By increasing the temperature up to 150° C, these weak organic groups can remove from the middle of the layers and leads to enhancing the carbon purity in EG particles, thereby increasing electrical conductivity of composite or NTCR effect.

Figure 7. The conductivity-temperature curves of heat-treatment test for different composites.
The I-V curves of PEM fuel cell containing P/G/CF/EG composite bipolar plate and P/CF cloth as GDL.

**Test station.**—Fig. 8 indicates the I-V curves of the P/G/EG/CF composite as bipolar plates. The hydrogen and oxygen were used as reactant gases. The effect of body temperature on performance of PEM fuel cell was investigated. There are three types of polarization of PEM fuel cells: activation, concentration, and ohmic polarizations. According to Fig. 8, the ohmic polarization influences on the slope of the middle part of the performance curve. This ohmic polarization can include the resistivity of the bipolar plates (P/G/CF/EG), GDL (P/CF), and the metallic parts as well as the interface resistance between these layers that two later items are neglected. It means that it is supposed that the increasing of body temperature of PEMFC only differ the ohmic resistance of P/CF composite of GD and P/G/CF/EG composite of bipolar plates. The other conditions related to reactant gases were considered constant to remove other polarizations. Fig. 8 shows the I-V curves related to different temperatures. It can be seen that the performance of the PEMFC increases with the increase of temperature. Decreasing the performance of PEMFC at temperatures above 80°C (Fig. 8) can be a result of drying of the membrane.

**Conclusions**

The DSC-TGA analyses indicated that dissociation reactions occurring in high temperature, perform at a single step in the P/CF and P/EG while performs at two steps in pure polymer. In addition, it can be observed that the P/EG has the highest decomposition temperature in comparison to other ones. The joule effect test showed that in all composites, especially for the first 60 s, NTCR effect has been observed and by enhancing the induced power, the slope of the curve increases. This fact may be as a result of dissociation of polymer among the carbon particles or is due to alignment of fibers and particles along of the path of electron passage. In addition, The result of heat-treatment test showed the P/EG and P/CF s have a PTCR effect and P/EG and P/G/CF/EG has a NTCR effect. The PTCR effect is as a result of expanding the polymer branches among the G or CF particles and getting away the particles from each other, thereby increasing electrical resistivity. However, the NTCR effect of P/EG and P/G/CF/EG can be related to removing the organic functional groups such as $S^2-$ and $OH^-$ from the interface of graphite layers and increasing the carbon purity, thereby increasing electrical conductivity of composite.

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