NANOCOMPOSITE HEATING ELEMENTS FOR INDUCTION WELDING OF THERMOPLASTIC COMPOSITES

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Keywords: *Nanocomposites, heating elements, joining*

ABSTRACT

This paper presents the preliminary results on the fabrication of conductive nanocomposite films and investigates their potentials and limitations to be used as novel heating elements (HEs) for induction welding of carbon fiber/polyphenylene sulfide (CF/PPS) thermoplastic composites. The nanocomposite films were a mix of a PPS thermoplastic polymer and either multi-walled carbon nanotubes (MWCNTs) or MWCNTs/iron oxide (Fe_3O_4) nanoparticles at different loadings. The nanofillers and the polymer were mixed using either a micro-extruder or a ball-milling machine. The nanocomposite HEs were then fabricated using a hot press and then cut into the desired dimension. Scanning electron microscopy observation revealed that the nanofillers are well dispersed into the polymer matrix. Electrical resistivity measurements showed that the HEs' resistivity decreases with the increase of nanofillers loadings with a maximum value of ~617 S/m for the HEs fabricated in the ball mill machine using 60 wt.% MWCNTs and 10 wt.% Fe_3O_4. Finally, the heating efficiency of the HEs was studied for induction welding of the CF/PPS composites. Preliminary results demonstrated that a maximum temperature of ~180 °C is achieved at the weld interface under the welding conditions which needs to be further improved for the achievement of a high quality welding.

1 INTRODUCTION

Thermoplastic are known as green materials and are widely utilized in various industrial applications such as in aerospace and automotive domains [1]. The ability to soften or melt thermoplastics and their associated fiber-reinforced composites enables joining two thermoplastic components, called adherends, through different welding processes. Joining by welding eliminates the common issues related to adhesive bonding of thermosetting composites such as long curing times and the need for extensive surface preparation as well as possible delamination and stress concentrations that may occur from common mechanical fastening [2].

Induction welding is an efficient process which consists in induction heating the interface between the two adherends above the polymer melting point for semi-crystalline polymers using a heating element (HE) placed at the interface. The HE, which is an electrically conductive or electromagnetic material, provides the required heat for welding when placed within a magnetic field. The HE is used either to heat the interface of two non-conductive adherends or localize the heat at the weld interface for electrically conductive composites. The HE then heats up the resins at its vicinity and the process continues until the resin reaches a desired welding temperature. Upon cooling the joint under an applied pressure, materials consolidation occurs which leads to the achievement of a welded joint [3, 4].

Metallic meshes such as stainless steel meshes are commonly used as efficient HEs [5-7]. However, the meshes revealed issues such as a poor adhesion with the surrounding polymer and possible stress concentration or residual stresses at the interface [8]. Polymer nanocomposites reinforced with electrically conductive or electromagnetic nanofillers such as multi-walled carbon nanotubes (MWCNTs) [9], graphene nanoplatelets [10] and metallic nanowires and nanoparticles [11] have high potential to be used for the fabrication of novel HEs.

Here, we present our preliminary results on the fabrication of nanocomposite conductive films as novel HEs. A polyphenylene sulfide (PPS) thermoplastic polymer was used as the matrix and reinforced with either MWCNTs or MWCNTs/iron oxide (Fe_3O_4) nanoparticles (IPs). The nanofillers were mixed with the polymer matrix using either a micro-extruder or a ball-milling machine. The nanocomposite HEs were then fabricated using a hot press and then cut into the desired dimension. Scanning electron microscopy (SEM) observation and electrical resistivity measurements were conducted on the fabricated HEs in order to gain insight on the nanofillers' dispersion and the HEs' conductivity. The HEs were finally used for induction welding of carbon fiber/PPS composites. The main outcome of this work is to assess potential of nanocomposite films as new HE types by finding the maximum temperature that can be reached.

2 EXPERIMENTAL

2.1 Fabrication of HEs

In this study, two types of nanocomposite-based HEs were fabricated. The first HE type was prepared by mixing PPS pellets (Sigma Aldrich) and MWCNTs (NanocylTM NC7000). The PPS polymer was used as the matrix in order to be similar to the composite adherends in terms of compatibility and melting temperature. The diameter and length of the MWCNTs were 10-20 nm and 5-30 µm, respectively. The MWCNTs were observed under a field emission scanning electron microscope (FE-SEM, Jeol JSM-7600TFE) and also a transmission electron microscope (TEM, Jeol JEM-2100F). To find out the effect of mixing process on the dispersion and electrical conductivity of the nanocomposites, two methods were used: extrusion method using a twin- screw micro extruder (Xplore®) and ball-mill mixing method using a miller/shaker machine (SPEX SamplePrep 8000M). The desired amounts of MWCNTs were added to the PPS pellets to make 3 gr mixtures with different nanotubes concentrations (i.e., 5, 10, 15, and 20 wt.%). Each mixture was fed into the extruder at 300 °C and mixed for 10 minutes. A temperature of 310 °C was maintained above the melting point of PPS (T_m = 280 °C) at both the inlet and exit of the extruder. The screw speed was 200 rpm. The nanocomposite mixtures obtained in the form of the filaments from the extruder were cut into small pieces using a cutting knife and were stored to be used for the fabrication of the HEs. In the second method, MWCNTs were dispersed into the PPS pellets using the ball-mill mixer in order to make MWCNT/PPS nanocomposites with concentrations similar to those prepared using the extruder. In addition, the simplicity of the ball-mill mixing method allowed us to fabricate nanocomposites with higher nanotube concentrations up to 60 wt.%. The ball-mill mixing time was 20 min.

The second HE type was a hybrid mixture of MWCNT and PPS pellets mixed with magnetic Fe_3O_4 nanoparticles. To achieve very high fillers concentrations (e.g., 70 wt.%), only ball-mill mixing method was used. Based on our experience, ball-mill mixing method is very practical for the synthesis of the nanocomposites with high nanofillers concentrations (>20 wt.%). This method does not deal with the problems associated with the high viscosity of the mixing materials that hinders the extrusion process by clogging the die. After mixing, the nanocomposites in the forms of granules (extrusion mixing) and powders (ball-mill mixing) were used to fabricate films of different thickness by compression molding in a hot press. A desired amount of the nanocomposite granules or powder (e.g., 5 – 10g) was loaded between two stainless steel rectangular plates. For easy removal of the compressed films, two Teflon films were placed to separate nanocomposite materials and the plates. The pressure varied between 2-4 bar while the processing temperature was set at 300 ºC for a constant holding time of 10 min. The plates were taken out of the mold and cooled down to room temperature by placing them in another press without applying any pressure. The heat conduction between the plates and the second press helped fast cooling of the plates and the pressed films (e.g., 5 – 6 min). The compressed sample was then cut into rectangular specimens to dimensions of 12.7 mm × 25.4 mm using a cutting knife and used as new HEs. Note that a nanocomposite with only 70 wt.% IP was also fabricated using the ball-mill mixing method for comparison purposes.

The fabricated HEs were observed under an FE-SEM (SU8230, Hitachi) in order to find out more information about the dispersion of nanofillers within the polymer matrix. The electrical resistivity of the HEs was finally measured using a two-probe multimeter and the resistance was calculated based on the HEs' thickness.

2.2 Induction welding

Thermoplastic composite laminates were fabricated from unidirectional (UD) pre-impregnated plies of CF/PPS (AS4/TC110 from Ten Cate Advanced Composites (CETEX®)). First, composite laminates with quasi-isotropic configuration and a thickness of ~ 2.6 mm were fabricated by stacking 16 layers with a sequence of $[(0/90/\pm45)_2]_s$ inside a metallic mold (30 cm by 30 cm) and pressed using a hydraulic hot press (Accudyne Engineering and Equipment Company). The processing temperature, molding pressure and holding time were 320°C, 0.7 MPa and 20 min, respectively. The average cooling rate was estimated to be 21°C/min. The laminates were then cut into smaller coupons with a dimension of 101.6 mm × 25.4 mm using a water-cooled diamond saw and used as adherends in the induction welding process.

Figure 1 schematically illustrates the welding process and the coupons assembly. The welding set-up is composed of several components mainly: 1. Power supply (Ambrell Easy Heat machine, 10 kW) to provide the required energy for heating with a frequency ranging from 150 kHz to 450 kHz. The power supply delivers a current that can vary from 0 to 700 mA and the frequency is managed automatically by the system and is kept constant at 268 kHz, 2. A hairpin type copper coil as inductor fed by the power supply to create the magnetic field, 3. A home-made pneumatic device to apply a pressure required for the welding, and 4. A water-cooling system to cool down the welding assembly. The HE was assembled between two identical composite adherends in a lap shear configuration according to the ASTM D5868 standard. The specimen (i.e., the adherends/HE assembly) was located under the coil so that the welding area was centered with the coil. Ceramic blocks were also used to fix the specimens at a desired location and apply pressure. A magnetic flux concentrator was also used to increase the magnetic field intensity [2]. The distance between the coil and the specimen varied between 2 – 5 mm using ceramic spacers. A wide range of current between 400 – 700A was applied. The increase of welding temperature as a function of time was acquired at the weld area and also at the upper surface of top adherends using two thermocouples and a temperature acquisition system (Graphtec). A constant welding pressure of 200 kPa was applied for all the tests. Applying higher pressures led to the creation of cracks in the HEs before the welding process begins, mostly for the HEs made of high fillers concentrations (e.g., 40 wt.%).

3 RESULTS AND DISCUSSION

3.1 Morphological and electrical characterization of HEs

Figure 2a and 2b show typical SEM and TEM images of the MWCNTs, respectively. According to the supplier, the MWCNTs are purified and seen as entangled materials (**Figure 2a**), that is a typical appearance of MWCNTs. However, these materials need to be properly dispersed and distributed within the matrix without damaging their aspect ratio (i.e., length/diameter) and their structural integrity (e.g., defects created during purification) in order to be effectively used for the enhancement of the electrical conductivity [12]. As it is seen in the TEM image (**Figure 2b**), the nanotubes have a diameter in the 10 – 20 nm range and lengths reaching up to few tens of microns.

Figure 3 represents SEM images of the nanocomposites with different loadings of nanofillers or mixing methods. **Figure 3a** shows an SEM image of the nanocomposites containing 5 wt.% MWCNTs, prepared using the micro-extruder. The nanotubes are clearly seen as the bright spots that are fairly dispersed into the PPS resin, as it is difficult to spot clusters of MWCNTs. The good dispersion of the nanotubes is still observed at a higher filler loading in **Figure 3b** for the nanocomposite containing 15 wt.% MWCNTs. Based on all the above observations, the fairly uniform dispersion of the nanotubes for both cases can be attributed to the effectiveness of the high shear mixing of the nanocomposite using the micro-extruder. The morphology of the nanocomposites prepared using the ball-mill mixing method is entirely different, as it is shown in **Figure 3c and d**. **Figure 3c** is an SEM image of a ball-mill mixed nanocomposite pellet (i.e., cluster) with nanotube concentration of 15 wt.%. As it can be seen in **Figure 3b and 3d**, for the nanocomposites prepared by the extrusion method, the nanotubes are well dispersed and embedded into the resin while the nanotubes seem to form a coating-like structures on the PPS pellets (or clusters) for the ball-mill mixed nanocomposites. The formation of PPS clusters might be attributable to the thermal energy caused by ball hitting that possibly caused PPS sintering.

Figure 4 shows the electrical conductivity of the nanocomposite HEs as a function of CNT concentration. It is observed that by increasing the MWCNT concentration from 5 to 20 wt.%, the electrical conductivity of the nanocomposites prepared by the extrusion mixing method increased by an order of magnitude from ~ 1.7×10^{-2} to ~ 0.45 $S.cm^{-1}$. Considering the inherent non-conductive characteristic of the PPS, the addition of MWCNTs into the matrix is believed to result in the formation of conductive networks throughout the nanocomposite, thus leading to the significant increase of the overall conductivity of the nanocomposites. The increase of viscosity which caused processing difficulty was the main issue that prevented the preparation of nanocomposites in the extruder at higher nanotube concentrations (i.e., >20 wt.%). The ball-mill mixing method, however, enabled the fabrication of nanocomposites with very high fillers concentrations (e.g., up to 70 wt.%). As it is shown in **Figure 4**, the electrical conductivity of the ball-mill mixed nanocomposites increased by almost three orders of magnitude from ~ 9.1×10^{-3} to ~ 4.2 $S.cm^{-1}$ by the addition of nanotubes from 10 wt.% to 60 wt.%. Due to uncertainty of the results, it is not possible to have a fair conclusion on the effect of the mixing methods on the nanocomposites' electrical conductivity at a given nanotube concentration (e.g., 10 and 15 wt.%). Based on the SEM observation (**Figure 3**), the nanotubes are well dispersed into the matrix in the extrusion method while the ball-mill mixed nanocomposites seem to be composed of a polymer core coated by the nanotubes. Although the good dispersion of nanofillers are a curial factor for the improvement of nanocomposites mechanical properties, the electrical conductivity is based on the creation of percolation pathways which is expected to occur more for the ball-mill mixed nanocomposites at high fillers concentrations [13]. The addition of Fe_3O_4 nanoparticles to make hybrid nanocomposite had a slight effect on the electrical conductivity of the nanocomposites. For example, the conductivity of the nanocomposite containing 30 wt.% MWCNTs increased from ~6.8×10^{-1} to ~1.96 $S.cm^{-1}$ when 37 wt.% IP was added. The IPs might place between nanotubes and thus interfere with the nanotubes percolation pathways, although it is only a hypothesis and was not experimentally verified here.

3.2 Heating efficiency of HEs

The capability of the fabricated HEs was evaluated for the induction welding of the composite adherends. **Table 1** lists the detailed processing conditions and properties of four different HE types used. **Figure 5** shows representative temperature-time curves obtained for the four HEs with the processing conditions shown in **Table 1**. The temperature-time curves are based on the data that was collected by the two thermocouples placed at the weld interface. The HEs revealed an initial sharp increase of temperature followed by a slower increase with the time. The heating behavior of the HEs was then quantified as average heating rates (i.e., increase of temperature over time). The average values of heating rates (averaged between 0 and 70 sec) provided by the HEs, the maximum temperature at the weld interface and the HEs conductivity are listed in **Table 1**. The average heating rates and the maximum interface temperature achieved were found to be dependents of the electrical conductivity as well as type of the HEs. The minimum average heating rate and lowest value of the maximum interface temperature were obtained for the nanocomposites containing 60 wt.% MWCNTs. The nanocomposite filled with 70 wt.% IPs showed higher values, although its electrical conductivity was lower than the MWCNTs-filled nanocomposite. This might be due to their different heating mechanisms. It is believed that, for the first HE type, the Eddy currents generate heat through Joule losses in the MWCNTs. Junction heating might be another heating source that can occur between the neighboring and in-contact MWCNTs. For the second HE type, the hysteresis loss might also contribute to the heating because of the magnetic properties of IPs [14]. This source of heating is generated when a magnetic particle vibrates and the energy is lost due to friction [14].

The hybrid nanocomposites showed better results when compared with the first two HE types discussed above. The best results were achieved for the nanocomposite containing 30 wt.% MWCNTs and 37 wt.% IPs showing a maximum interface temperature of 183 ºC and an average heating rate of 2.04 ºC/min. The achievement of the highest value in this case can be attributed to the synergetic effect of the hybrid nanocomposites by the combination of several heating mechanisms related to the nanotubes and IPs.

Even with all these achievements, the induction welding of the composite adherends was unsuccessful under any processing conditions using the present induction welding setup. For a successful induction welding of the composite adherends used in this study, the temperature required at the weld interface has to be around 320 ºC (above the melting point of the PPS), which is far above the maximum temperature achieved using our nanocomposite HEs.

4 CONCLUSION

In the present study, various nanocomposite films were fabricated and tested as new HE types for induction welding of thermoplastic composites, i.e., UD CF/PPS stacked in a quasi-isotropic configuration. The main goal was to gain an insight about the heating behavior of the new nanocomposite-based HEs as well as finding the influencing processing parameters and the issues towards the efficient utilization of the nanocomposite films as new HEs. It was found that the average heating rate and the maximum temperature at the weld interface depend on the electrical resistivity of the HEs and their types. The HE made of a hybrid nanocomposite containing 30 wt.% MWCNTs and 37 wt.% IP showed the highest values for the maximum temperature at the interface (i.e., 183 ºC) and the average heating rate (i.e., 2.04 ºC/min) under similar testing conditions. The maximum temperature achieved was far below the temperature required (i.e., 310 ºC) for the induction welding of such a thermoplastic composite, and therefore led to an unsuccessful welding. Future works will focus on finding the best welding conditions as well as the improvement of the electrical conductivity of the HEs. The latter will be achieved by a systematic investigation on the nanocomposite mixing strategies and the utilization of other types of nanofillers such as metal-coated nanotubes or nickel nanostrands. The development of such heating elements is foreseen to address the current issues existing with metallic susceptors sometimes used in the induction welding process.

5 ACKNOWLEDGEMENTS

The authors acknowledge the financial support for postdoctoral fellows (postdoctoral fellowship award) from Natural Sciences and Engineering Research Council of Canada (NSERC). The authors also acknowledge the financial support from NSERC (Discovery Grant Program) by which the materials were purchased and the testing expenses were covered. The authors also would like to thank Professor Daniel Therriault for some experiments that were performed in his research labs at Polytechnique Montreal.

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Table 1. Welding parameters and materials used for the samples shown in Figure 5.

Sample No.	HE	Conductivity (S/cm)	Coil/sample distance (mm)	Input current (A)	Max. Temp. (°C)	Average heating rate (ºC/min)
1	60 wt.% MWCNTs	5.35	~2	700	133	1.04
2	60 wt.% MWCNTs+10 wt.% IP	6.17	~2	700	152	1.57
3	30 wt.% MWCNTs+37 wt.% IP	1.95	~2	700	183	2.04
4	70 wt.% IP	3.0×10^{-3}	~2	700	150	1.38

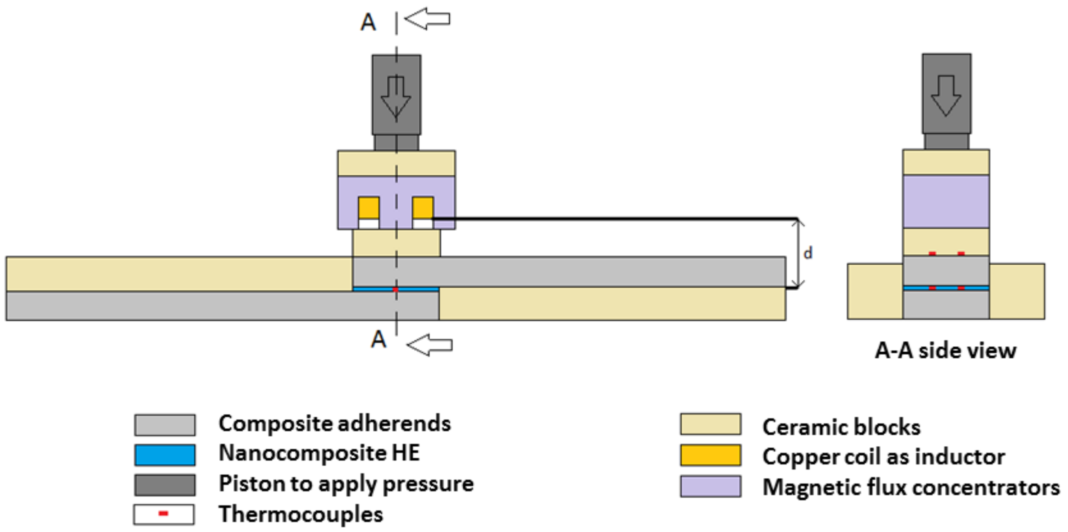

Figure 1: Schematic representation of the induction welding setup.

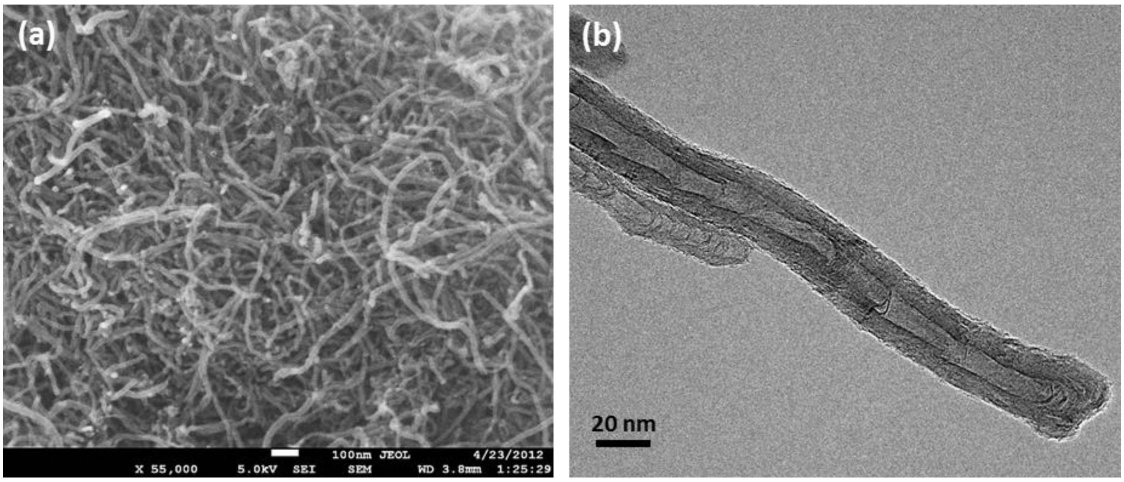

Figure 2: SEM (a) and TEM (b) micrographs of MWCNTs used in this study.

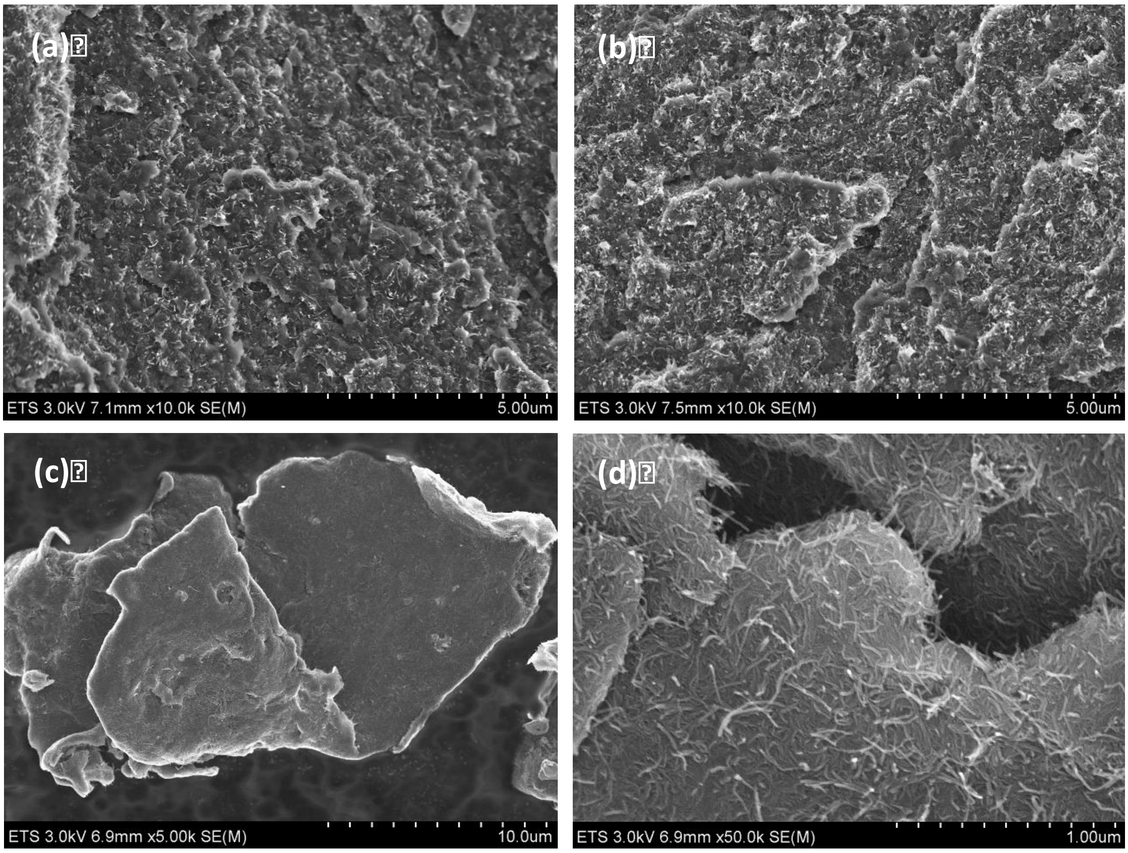

Figure 3: SEM micrographs of (a) nanocomposites containing 5 wt.% MWCNTs and (b) nanocomposite containing 15 wt.% MWCNTs, both prepared using the micro-extruder, (c) a ball-mill mixed nanocomposite pellet (i.e., cluster) with nanotube concentration of 15 wt.% and (d) a higher magnification image of (c).

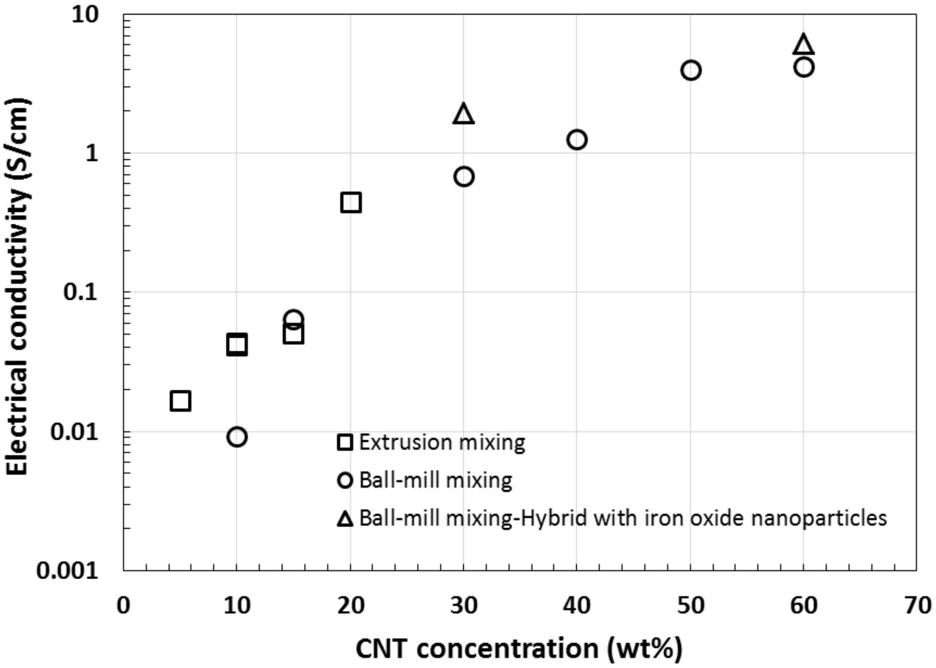

Figure 4: Electrical conductivity of the nanocomposite HEs as a function of MWCNT concentration. The hybrid nanocomposites contain 37 wt.% (for the left point) and 10 wt.% (for the left point), respectively.

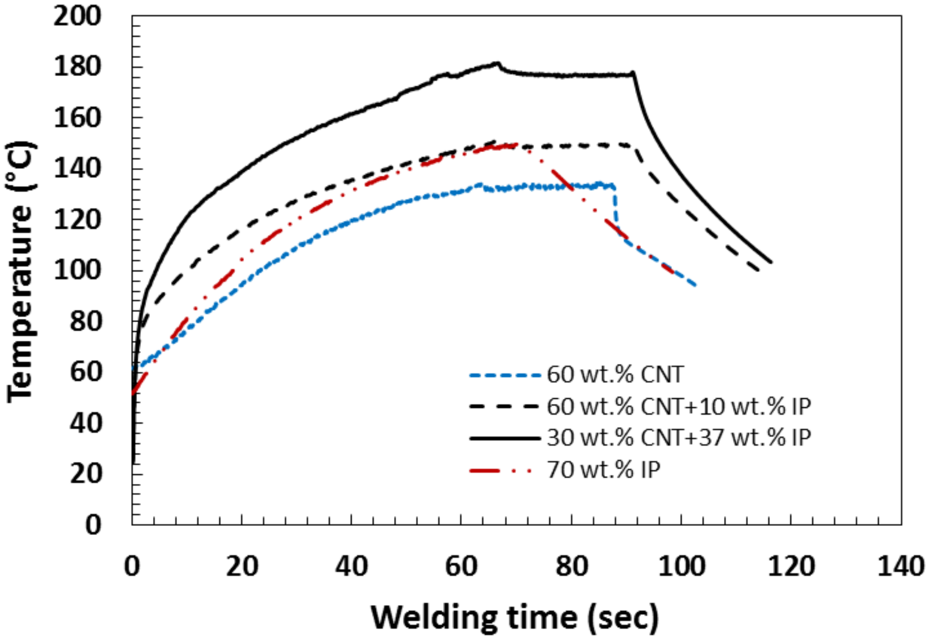

Figure 5: Heating behavior of the fabricated nanocomposite HEs as temperature variation at the weld interface as a function of welding time at the input current of 700A.