Advances in Nano Research, *Vol. 15, No. 4 (2023) 285-294*
https://doi.org/10.12989/anr.2023.15.4.285

Effect of the crude oil environment on the electrical conductivity of the epoxy nanocomposites

Seyed Morteza Razavi[1], Soroush Azhdari[2,3] and Fathollah Taheri-Behrooz*[3]

[1]*Department of Mechanical Engineering, University of Science and Culture, Tehran, 14619-68151, Iran*
[2]*The University of British Columbia, Vancouver, Canada*
[3]*School of Mechanical Engineering, Iran University of Science and Technology, Tehran, 16846-13114, Iran*

(*Received September 19, 2021, Revised February 19, 2023, Accepted July 27, 2023*)

Abstract. This study is aimed to investigate the electrically conductive properties of epoxy nanocomposites exposed to an acidic environment under various mechanical loads. For simultaneous assessment of the acidic environment and mechanical load on the electrical conductivity of the samples, the samples with and without carbon nanotubes were exposed to the acidic environment under three different loading conditions for 20 days. Then, the aged samples' strength and flexural stiffness degradation under crude oil and bending stress were measured using a three-point flexural test. The aged samples in the acidic environment and under 80 percent of their intact ultimate strength revealed a 9% and 26% reduction of their electrical conductivity for samples with and without CNTs, respectively. The presence of nanoparticles declined flexural stiffness by about 16.39%. Scanning electron microscopy (SEM) images of the specimen were used to evaluate the dispersion quality of CNTs. The results of this study can be exploited in constructing conductive composite electrodes to be used in petroleum environments such as crude oil electrostatic tanks.

Keywords: carbon nanotubes (CNT); bending load; electrical conductivity; epoxy nanocomposites; stress corrosion; threshold

1. Introduction

The high resistance of polymeric materials against electron transfer or other electrical current carriers has resulted in their application in numerous industries. The insulation of polymers, however, is only sometimes merit. In some applications, conductive polymers could be a leading choice, as conductive polymeric composites are highly popular in corrosive environments. In this regard, remarkable efforts have been devoted to fabricating conductive polymers (Osman *et al*. 2022, Xu *et al*. 2022). Compared to conductive metals, polymeric composites are benefited from low density, ease of formability and flexibility in design, as well as high electrical conductivity range and corrosion resistance (Letti *et al*. 2017, Razavi *et al*. 2019, Arani *et al*. 2021). In combination with carbon fibres, conductive polymers can form composites with high corrosion resistance and mechanical strength which are good candidates to fabricate electrodes. The United States Department of Energy (DOE) necessitates minimum electrical conductivity of 100 S/cm for the electrode sheets (Tabatabaee *et al*. 2019). These electrodes can be used for separating water from oil in the electrostatic desalination reservoirs of crude oil (Razavi *et al*. 2022). Regarding the application of these electrodes in a crude oil environment, it should be indicated that these electrodes can maintain their electrical conductivity and mechanical strength during their operation.

Today, various types of conductive nanoparticles and carbon fibres are among the most common components of conductive polymeric composites (Gu *al*. 2019, Luo *et al*. 2022). The reason is the high tendency of the nanoparticles to form a conductive network due to their chain-like accumulated structure compared to the other conductive additives, such as metallic powders. Carbon fibres can also be regarded as a long chain-like accumulation of carbon particles.

Carbon nanotubes (CNTs) are among polymers' most applied conductive fillers. The electrical conductivity of a composite generally depends on the volume fraction of the filler (Tabatabaee *et al*. 2019, Guo *et al*. 2022). Permeability threshold refers to the critical content of the filler to form a conductive composite based on the diagram of conduction vs. additive content. This concept implies the lowest concentration of the fillers by which the continuous conductive branches or chain will be formed. The type, size, and orientation of the fillers in the polymeric matrix can significantly affect the electrical and mechanical properties of the composites.

Johnson (2009) used polypropylene resin and various fillers such as carbon black, graphite, and CNT (with respective contents of 2.5, 6.5, and 6 wt.%) to prepare composites with electrical conductivity of 91 S/cm. Mighri *et al*. (2004) applied carbon black, graphite, and carbon fibre to enhance the electrical conductivity of poly-propylene resin and polyphenylene sulfide. Their results indicated the best condition (electrical conductivity of 16 S/cm and tensile strength of 84 MPa) for the composite containing 60% filler. Gholami *et al*. (2015) assessed the conduction of polyaniline/zinc oxide nanocomposite. They indicated that the incorporation of ZnO NPs can decline the

*Corresponding author, Professor
E-mail: taheri@iust.ac.ir

http://www.techno-press.org/?journal=journal=anr&subpage=5 ISSN: 2287-237X (Print), 2287-2388 (Online)

conduction of the polyaniline due to the semiconducting nature of ZnO. Using phenolic resin, carbon fibre, and carbon black fillers, Chen *et al.* (2004) made a composite with optimal electrical conductivity and strength of 107 S/cm and 173 MPa, respectively. Wang (2006) used polypropylene, graphite, carbon fibre, and carbon black fillers to fabricate a composite containing 65 wt.% filler. He measured the volumetric electrical conductivity and flexural strength for the optimal condition as 105 S/cm and 47 MPa, respectively. Kakati *et al.* (2010) used phenol resin and a combination of carbon black, graphite and carbon fibre filler (respective contents of 5.5 and 6 wt.%) to prepare a composite with electrical conductivity of 92 S/cm and flexural strength of 55 MPa. Chun-hui *et al.* (2006) used compression moulding to fabricate a conductive composite from sodium silicate. They investigated the effects of graphite content, graphite particle size, and process duration on the electrical conductivity of the composites. Jin *et al.* (2013) achieved an electrical conductivity of 100 S/cm for composites containing 85 wt.% graphite. Dweiri and Sahari (2007) used compression moulding to fabricate poly-propylene/graphite/carbon black composites with electrical conductivity of 7 S/cm. Rhodes *et al.* (2007) tried to improve the electrical properties of resins by forming a carbon fiber network at low concentrations (low penetration threshold). Barton *et al.* (2006) added three different carbon fillers (carbon black, synthetic graphite, and carbon fibre) to the liquid crystal resin of Vectra A950RX. They applied three analytical models to predict the electrical conductivity in the composite systems with filler. All three models led to acceptable results. Liao *et al.* (2008, 2010) in several studies, fabricated polypropylene/ graphite composites with multiwall carbon nanotubes (MWCNTs) by compression molding and melt mixing. They achieved an electrical conductivity of 100 S/cm at 80 wt.% graphite. Du *et al.* (2008) expressed that as the electrical conductivity of filler-free carbon-based composites is about 300 S/cm, expanding graphene could be one of the best-conducting filler options. Xie *et al.* (2009) examined the effect of graphene nanosheets and CNTs on the electrical conductivity of composites. Park *et al.* (2009) experimentally determined the effect of graphite nanofibres on poly nanocomposites and recorded an improvement in the mechanical, thermal, and electrical properties of poly-resin. Shokrieh *et al.* (2013) fabricated conductive composites from vinyl ester, carbon fiber, and carbon black at various weight percentages. These composites can be used as conductive resin composites in corrosive environments. Bourell *et al.* (2011) showed that using nano-sized carbon black could negatively impact the composite sheets with natural graphite as the particles may cover the natural graphite. Moreover, the electrical conductivity and flexural strength may drop under such conditions. Guo *et al.* (2012) improved the conductive graphite composite sheets by selective sintering with the help of a laser. This technique was more cost-effective than conventional injection and compression molding and managed to modify the conductive sheets suitably. Hosseini and Zandi (2016) investigated the electromechanics of reinforced composite sheets. Taherian *et al.* (2011, 2013) also addressed the production of bipolar composites with proper electrical conductivity, mechanical properties, and gas permeability to be employed in resin fuel cells. Modarresi *et al.* (2016) assessed the conductive polyaniline polymers with silica additive.

Thanks to their high corrosion resistance, conductive polymers and composites can be used in diverse applications, such as the electrodes of crude oil electrostatic desalination tanks. In this application, the workpieces are simultaneously exposed to mechanical load and a corrosive environment. So, obtaining the electrical conductivity threshold of these conductive materials and the influence of exposure to an acidic environment and mechanical loads on the electrical conductivity is one of the essential features of this research.

The present study is thus aimed to assess the electrical conductivity and mechanical strength of carbon/ epoxy composites with CNT reinforcement under flexural load exposed to crude oil. To this end, carbon/epoxy samples were prepared with and without CNTs and their optimal electrical conductivity threshold was obtained. Then, the samples were exposed to a three-point flexural load, and the electrical conductivity variation under various flexural loads was assessed within and outside the crude oil environment. The residual mechanical strength of the samples was determined under multiple flexural load conditions upon exposure to the crude oil.

Table 1 Specifications of ML-506 epoxy resin

Parameter	Value
Viscosity (°C25)	1450 cp
Combined time	3 min
Density	11/1 g/m3
Combined weight ratio	15/100 unit
Lifetime of consumption	50 min
Gel time	60 min
Drying time	90 min
Final drying time	Seven day

2. Materials and methods

2.1 Materials

This research used epoxy resin (ML-506) with HA-11 hardener (Mokarrar Engineering Company) as the matrix. This resin is based on bisphenol A epoxy resin, and its hardener is a polyamine. The specifications of the applied epoxy resin based on the manufacturer catalog are listed in Table 1 (Mokarrar Industrial Group. http://mokarrar.com/en. available on 4 September 2018). Various percentages of CNTs were added to increase the electrical conductivity of the resin. The CNT incorporation method can drastically affect the composite’s agglomeration and final properties (Antunes *et al.* 2011, Karimi *et al.* 2017, Feng *et al.* 2021). The stirring method can also affect the composite properties (Yogeswaran and Chen 2008). The specifications of the CNTs (Advanced nano-powder) are presented in Table 2 (Shimimarket. http://www.shimimarket.com/. available on 4

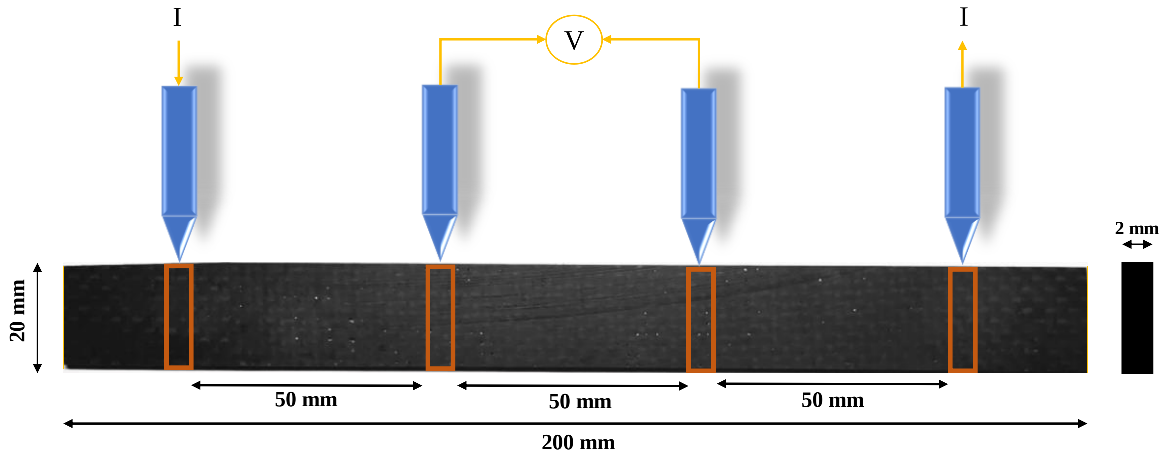


Fig. 1 Schematic of a three-layered beam including a core layer and two piezoelectric layers

Table 2 Physical and apparent characteristics of CNTs

Parameter	Value
Carbon purity percentage	95 %
Electrical Conductivity	> 100 s/cm
Density	1.8 g/cm3
Bulk density	356g/L
Medium diameter	1.1 nm
Length	20 μm
structure	Zigzag

Table 3 Specifications of corrosive environment of crude oil

Parameter	Value
Oil Temperature	65°C
Oil API Degree	29
BS&W	15%
S.G @ 15°C	0.88
Viscosity (cst)	@20°C 17
Inlet Salt Content	20,000-240,000 PPM
H2S Content	S60 W:80 PPM

September 2018). The carbon fabrics prepared by the Torayca company were employed as epoxy resin reinforcement to enhance the mechanical and electrical features of the resin. The fabric is woven plain with a surface density of 1.38 g/cm^2. The electrical conductivity of the fabrics in the fibre direction was 80 S/cm (TORAYCA® | TORAY. http://www.torayca.com/en/. available on 4 September 2018). Crude oil was used as the acidic environment, whose properties are tabulated in Table 3.

2.2 Electrical conductivity evaluation by four-point resistance measurements

The four-point resistance measurement method is common for measuring electrical conductivity (Wafers 2003). Most precise digital multimeters (DMMs) and many source measurement units (SUs) offer four-point resistance

Table 4 Electrical conductivity calculation characteristics

Value	Parameter
R = V/I *Ω*	Electrical Resistance
d = 0.2 cm	Thickness
α = 40.5	Geometric Constant
L = 1 (for d/s < 0.4)	Thickness Constant
T = 1 (at 23 ℃)	Temperature Constant
S ≥ 5 cm	Meter Distance

measurement. The electrical conductivity was measured in this study according to the ASTM F390-98 standard (Pan *et al*. 2000).

Four-point resistance measurement is often used to measure resistances below one kΩ. The applied four-point resistance measurement device includes four equally distanced metallic measurers made from tungsten with a limited tip diameter. Any of these measures is fixed by some springs to decline the damage during the tests. These metallic measurers are components of an automatic mechanical base moving upward and downward during the measurement. A current source with high impedance was used to supply the two outer measurement bases. A voltmeter also measures the voltage between the two internal measurement bases to determine the sample resistance, as shown in Fig. 1.

As the current can be drastically high in this method, controlling the voltage by the current is practically difficult. Thus, a capacitor is used with proper capacitance. Finally, the specific electrical resistance and conduction were determined by applying various voltages and currents based on Eqs. (1) and (2).

$$\rho(\Omega.\text{cm}) = \alpha \times T \times L \times d \times R \quad (1)$$

$$\beta\ (\text{s/cm}) = 1/\rho \quad (2)$$

where, ρ, β, R, d, L, T, and α denote specific electrical resistance, electrical conductivity, electrical resistance, thickness, and constant values, respectively, as listed in Table 4.

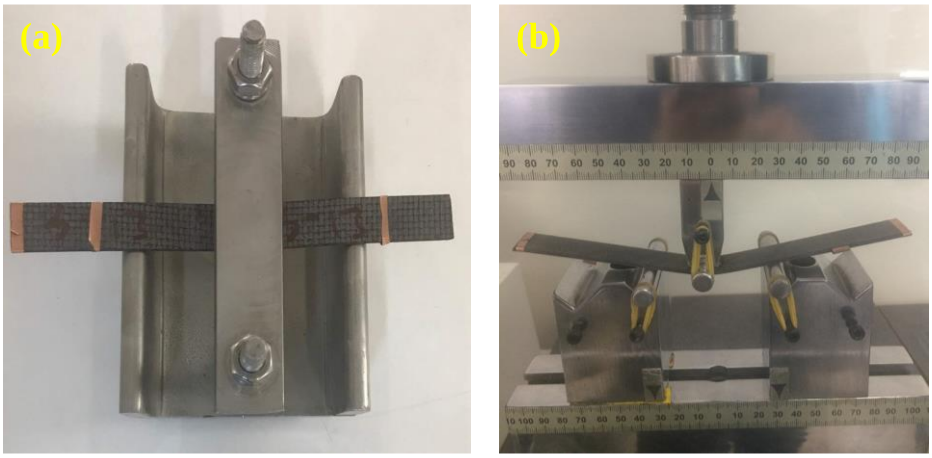


Fig. 2 (a) Loading fixture with the sample, (b) Three-point bending test fixture

2.3 Flexural strength of the sample

The constant displacement method was utilized to measure the mechanical response of the composite samples in ordinary and corrosive environments. The specific three-point bending fixture was developed according to Fig. 2(a) to study the mechanical load's effect on the conductive composites' electrical conductivity. As seen, the specimens were placed in the fixture. After firming the screws of the clamp and placing the measurement device between the two openings, the transverse deformation of the sample was zero. Then, the desired displacement was applied to the samples using regulation bolts. Then, the fixture was entered into the crude oil and kept there for 20 days. After 20 days, the fixture was removed, washed with distilled water, and dried at ambient temperature. Finally, to measure the flexural strength, the samples were tested according to the ASTM-D7264 flexural test standard (Testing and Materials 2015) using a 5-ton SANTAM apparatus (STM50), as depicted in Fig. 2(b).

3. Sample preparation

3.1 Carbon/epoxy samples

Carbon/epoxy samples were fabricated using ten layers of woven carbon fabric and epoxy resin using the hand layup method. The samples were first cured at room temperature for 24 h, followed by 6 hours of complete curing at 80 ℃. The initial samples had a dimension of $200\times 20 \times 2$ mm^3. The fibre volume fraction was measured based on ASTM D3171-06 (ASTM 2015), equal to 53.5%.

3.2 Epoxy Resin samples filled with CNT

To determine the optimum filler content to fabricate conductive epoxy resins, epoxy samples were prepared with 5, 10, 15, and 25 wt.% CNTs. To this end, the resin and desired level of CNTs were mixed by an electric mixer (POLYTRON, PT1200C) for 10 minutes, followed by 45 minutes of sonication (BANDELIN, with US70/T probe) at 40 ℃ at the power of 80 W. As CNTs were highly concentrated, the samples were diluted by heating to 90 ℃ for 5 min in 5-minute intervals. Then, the hardener was added to the sonicated mixture, stirring the produced solution for 5 minutes. After casting the resin in the molds, the samples were degassed for 20 minutes using a vacuum pump. As air is electrically insulated, this process can significantly enhance conduction. Finally, the samples were completely cured in an oven (STUART SCIENTIFIC) for two hours at 80 ℃.

3.3 Carbon/epoxy composite samples filled with CNT

After determining the optimum CNT content in the epoxy resin for the electrical purpose, the carbon/epoxy composite samples containing the optimum CNT content were fabricated, and their electrical conductivity was assessed under mechanical loads in the crude oil environment. The samples with a final dimension of $200\times 20 \times 2$ mm^3 were fabricated using ten layers of unidirectional carbon fabric with optimum CNT content. Based on the resin vendor's instruction, the samples were cured at room temperature for 24 h, followed by 2 hours of final curing at 80 ℃. Fig. 3 shows the scanning electron microscopy (SEM) images for the carbon/ epoxy composite filled with CNT. Nanoparticles are evenly distributed as fine worm-shaped particles on carbon fibres and inside the resin. The orientation of the nanotubes inside the resin depends on the stirring method and the length-to-diameter ratio of the filler. As SEM images show, CNTs in the carbon/epoxy samples have a random orientation.

4. Results

The carbon/epoxy composites with and without CNT fillers were tested in the current research. The former group was labelled by C, indicating the presence of CNT, while the latter was marked by E, denoting epoxy. The following results are the mean value of the three samples tested in each condition. In each series of the samples, one sample is regarded as the reference and was not placed in the

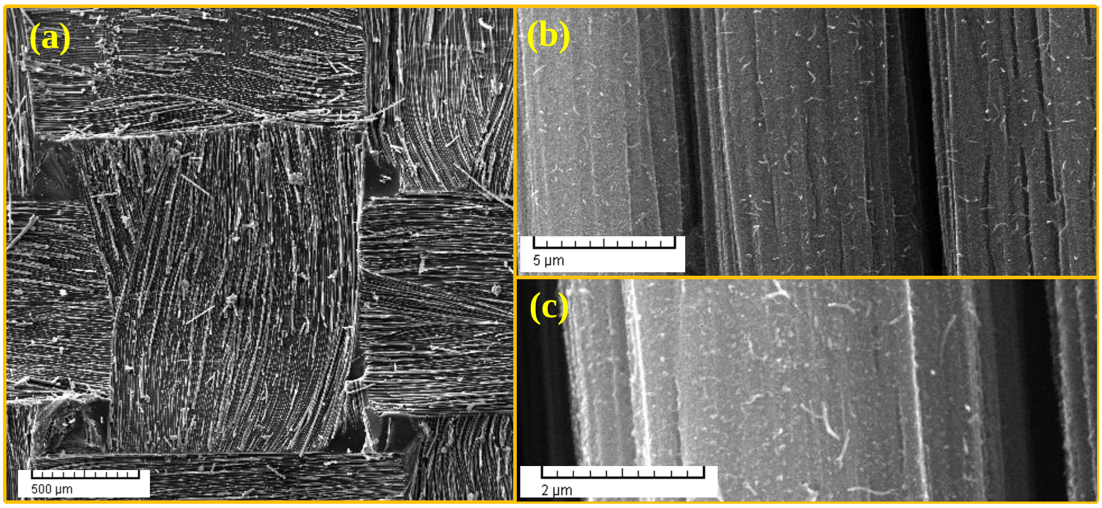


Fig. 3 Different resolution SEM images of the carbon/epoxy sample filled with CNT

Table 5 The details of the carbon /epoxy specimens with/ without CNTs coding in acidic and normal environment under different transverse displacements.

Material	Transverse displacement	Positioning duration in crude oil (day)	Sample code
Carbon/ Epoxy /CNT	-	-	C-C-0
Carbon/ Epoxy/ CNT	Failure displacement 40 %	-	C-C-40
Carbon/ Epoxy/ CNT	Failure displacement 80%	-	C-C-80
Carbon/ Epoxy	-	-	E-C-0
Carbon/ Epoxy	Failure displacement 40 %	-	E-C-40
Carbon/ Epoxy	Failure displacement 80%	-	E-C-80
Carbon/ Epoxy/ CNT	-	20 day	C-A-0
Carbon/ Epoxy/ CNT	Failure displacement 40 %	20 day	C-A-40
Carbon/ Epoxy/ CNT	Failure displacement 80%	20 day	C-A-80
Carbon/ Epoxy	-	20 day	E-A-0
Carbon/ Epoxy	Failure displacement 40 %	20 day	E-A-40
Carbon/ Epoxy	Failure displacement 80%	20 day	E-A-80

corrosive environment, denoted by C. Those samples exposed to the corrosive environment are represented by A. The specimens placed in crude oil for 20 days and under the bending loads of 0, 40, and 80% of failure displacement were denoted by 0, 40, and 80 codes, respectively. The sample coding system is presented in Table 5.

4.1 Results of optimal CNT content

Fig. 4 depicts the electrical conductivity of resin samples containing different amounts of CNT fillers. An increase in the filler content enhanced the electrical conductivity. This incremental trend was first linearly followed by a constant trend, reflecting the threshold of the electrical conductivity permeability, which is 10% for the CNT filler (C-C). Worth to mention this value of electrical conductivity is in the DOE-acceptable range.

High filler percentages can affect the composites' mechanical and electrical properties in two ways. First, there will be less resin to wet the fillers, and second the higher probability of fillers agglomeration. The strength of

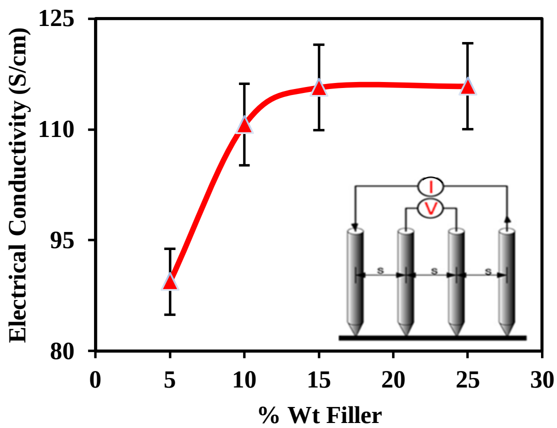


Fig. 4 Electrical conductivity threshold of the resin samples containing 5, 10, 15, and 25 wt.% CNT

the filler and matrix can effectively enhance the strength of the composite if the load can be transferred from the matrix to the fillers. This requires high bonding strength at their interface to transfer the stress. The electrical conductivity

Table 6 Carbon/epoxy samples with /without CNT in the different environments under mechanical loading

	Electrical Conductivity (S/cm)				
Sample Code	1	2	3	Average	STD
C-C-0	141.66	148.02	151.74	147.21	5.12
C-C-40	140.11	145.33	150.15	145.19	5.02
C-C-80	139.51	144.11	148.95	144.19	4.72
E-C-0	32.46	32.22	33.79	32.79	0.85
E-C-40	21.11	21.18	21.11	21.13	0.04
E-C-80	21.01	20.12	19.85	20.32	0.61
C-A-0	136.83	145.30	148.56	144.17	90.5
C-A-40	134.58	140.83	145.07	140.16	5.28
C-A-80	127.49	133.42	133.53	131.48	3.46
E-A-0	31.81	31.67	33.19	32.23	0.84
E-A-40	17.11	16.35	16.66	16.70	0.38
E-A-80	12.12	11.13	13.84	12.36	1.37

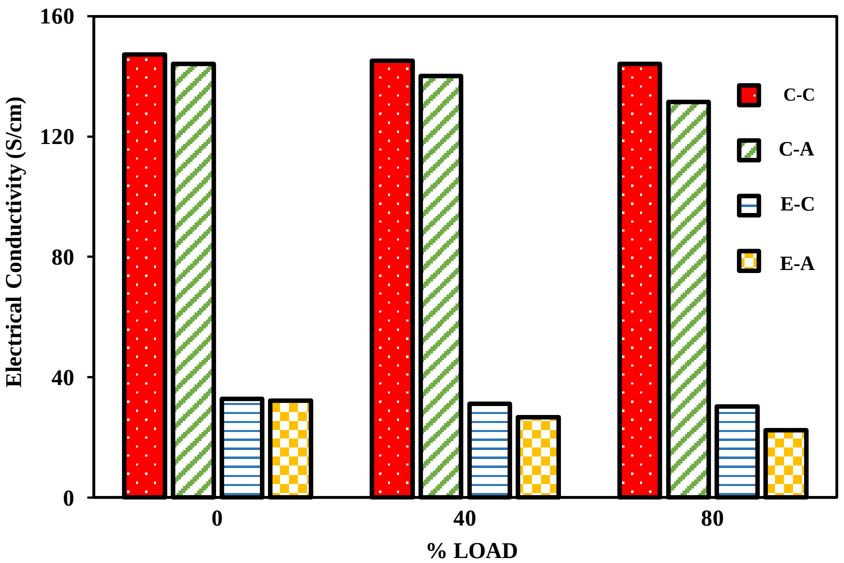


Fig. 5 Electrical conductivity vs. transverse displacement for the carbon/epoxy composites with/without CNT in an acidic environment

threshold was also measured by adding ten layers of carbon fabrics which was shown that adding carbon fabric layers did not affect the electrical conductivity threshold. The electrical conductivity threshold of the samples was in the DOE-acceptable range.

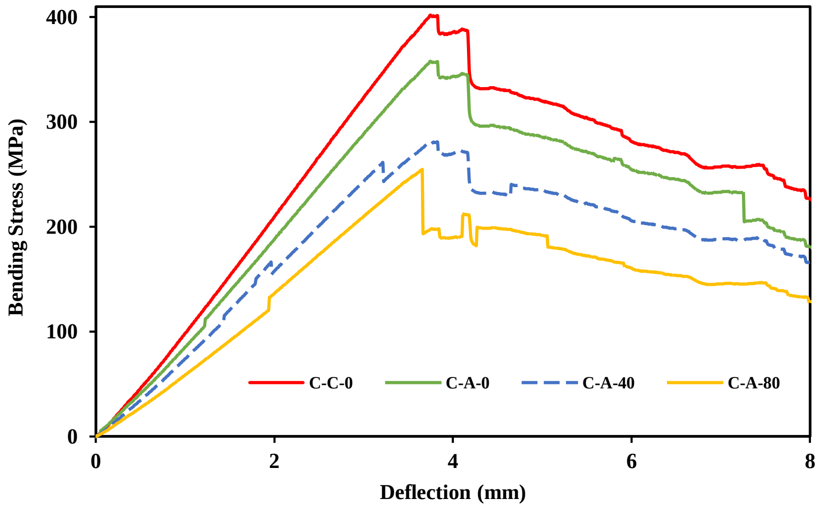
4.2 Electrical conductivity and flexural test results of the samples

Table 6 shows electrical conductivity measured in the carbon/epoxy samples with and without CNTs (CC-0, EC-0) as well as those exposed to 40% and 80% transverse displacement relative to the ultimate displacement (CC-40, CC-80, EC-40, EC-80).

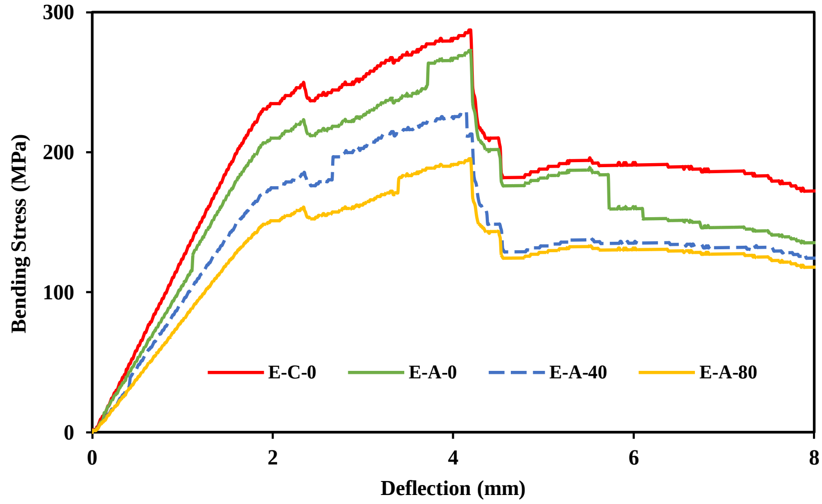
As shown in Table 6, the flexural load-induced displacement reduced the electrical conductivity. The higher the displacement, the more significant the decline in the electrical conductivity. Upon flexural load application, the inter-particle distance increased due to the strain, which declined the conductivity. Moreover, with increasing flexural load, the rate of decrease in electrical conductivity was higher in the samples without CNT. In other words, the presence of nanoparticles is effective in reducing electrical conductivity by increasing the flexural load.

4.3 Electrical conductivity and flexural test results of carbon/epoxy with and without CNT exposed to the acidic environment

Table 6 shows the electrical conductivity of the specimens subjected to an acidic solution for 20 days (CA-0, EA-0) and loaded at 40% and 80% of their ultimate displacement (CA-40, CA-80, EA-40, EA-80). As suggested by Table 6, in the neat carbon/epoxy samples, the electrical conductivity was reduced by exposure to an acidic solution which is due to the damage to the constituent



(a) The CNT-filled carbon/epoxy samples in a normal and acidic environment



(b) The neat carbon/epoxy specimens in a normal and acidic environment

Fig. 5 Electrical conductivity vs. transverse displacement for the carbon/epoxy composites with/without CNT in an acidic environment

crystal structure (Kim *et al*. 2005). Furthermore, exposure to an acidic solution and different load levels had little impact on the reduction of the electrical conductivity, where the slight decrease is attributed to the stretch of the resin between the fibres upon bending. In this case, the electrical conductivity is very low compared to the sample filled with carbon nanotubes. According to the results, exposure to an acidic solution causes damage to CNT particles. It reduces electrical conductivity, but this damage is not enough to ignore the role of CNT particles in electrical conductivity in an acidic solution (Kim *et al*. 2005). As shown in Fig. 5, the electrical conductivity further decreased by increasing the flexural load in an acidic environment, which was more significant in the neat carbon/epoxy samples.

4.4 Mechanical strength of the samples

Three-point bending tests were performed on normal specimens as well as those exposed to an acidic environment for 20 days. Three samples were tested for each condition, and the average is reported in Table 7. The failure load, along with the failure transverse displacement, were obtained from the three-point flexural test, as reported in Table 7. Based on Table 7, the ultimate failure load of the

Table 7 Failure load and failure displacement values of the carbon/epoxy samples with and without CNTs in normal and acidic environment under variable transverse displacements

Sample Code	Failure Load (N)	Failure displacement (mm)
C-C-0	213.28	3.75
C-A-0	189.82	3.75
C-A-40	149.30	3.75
C-A-80	135.08	3.65
E-C-0	152.06	4.19
E-A-0	144.46	4.19
E-A-40	121.02	4.15
E-A-80	103.40	4.19

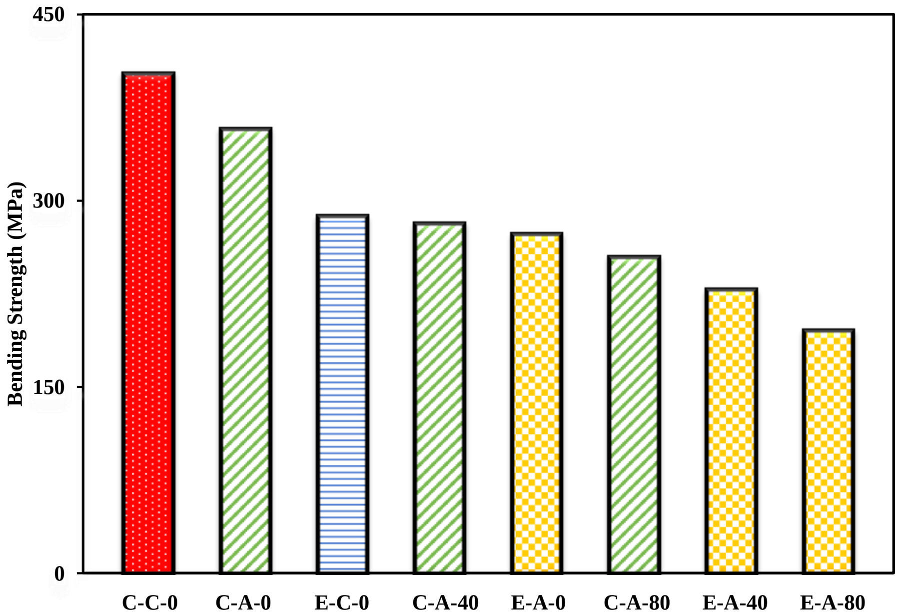


Fig. 7 Flexural strength of the carbon/epoxy samples with/without carbon nanotubes in normal and an acidic environment with/without applying transverse displacements

samples decreased by increasing loading in an acidic environment. The failure load of the carbon/epoxy composites filled with CNT was higher than neat carbon/epoxy composites. In other words, the strength of the samples declined by increasing mechanical load in an acidic solution which can be due to the penetration of acidic solution between nanoparticles and fibres. Moreover, stress corrosion can also explain such a drop in properties under load in an acidic environment. The transverse displacement associated with the fracture point was lower in the samples with the nanoparticle.

According to Tables 7 and 8, failure displacement did not exhibit substantial changes with increasing load in the acidic environment. Significant changes in the acidic environment are related to the samples' electrical conductivity and failure load.

The stress on the center of the beam was determined at any point on the force-displacement diagram during the flexural test. The stress-displacement diagram of the CNT-filled carbon/epoxy and the neat carbon/epoxy samples are shown in Figs. 6(a) and (b), respectively.

As shown in Fig. 6(a), the flexural strength and overall stiffness of the specimens exhibited a reduction by increasing the load in the acidic environment. The flexural strength in the neat carbon/epoxy specimens also decreased by increasing the load in the acidic environment. Note that these values are less than their corresponding values in the CNT-filled samples. By comparing Figs. 6(a) and (b), it was shown that the flexural strength reduction in neat samples was about 30-45% of the CNT- filled samples.

Fig. 7 shows the bar chart of the flexural strength of the carbon/epoxy samples with/without carbon nanotubes. Based on Fig. 7, exposure to an acidic solution decremented flexural strength. Moreover, increasing the load in the acidic environment led to an extra reduction in the flexural strength of the samples. This is because of the oxidation phenomenon due to corrosion stress. Table 8 lists the flexural strength and the percentage of loss in flexural strength for each sample. Due to being in an acidic environment, the damage to CNT particles reduces flexural strength. Also, an increase in the applied mechanical load

Table 8 Flexural strength of the carbon/epoxy samples with/without CNT in the normal and acidic environment under various transverse displacements

Sample Code	Flexural strength (MPa)	Percentage drop (%)
C-C-0	402.06	----
C-A-0	357.83	11.00
C-A-40	281.44	30.00
C-A-80	254.57	36.68
E-C-0	287.18	----
E-A-0	272.82	5.00
E-A-40	228.49	20.43
E-A-80	195.28	32.00

Table 9 Flexural stiffness of the carbon/epoxy filled with CNTs and without CNTs samples in normal and acidic environment under variable transverse displacements

Sample Code	Flexural stiffness (GPa)	Percentage drop (%)
C-C-0	103.41	----
C-A-0	92.03	11.00
C-A-40	77.40	25.15
C-A-80	66.56	35.63
E-C-0	86.46	----
E-A-0	77.38	10.50
E-A-40	66.21	23.56
E-A-80	54.11	37.41

reduces the mechanical strength of the samples.

The flexural stiffness of all specimens and the percentage of loss relative to the reference sample were calculated and tabulated in Table 9. The flexural stiffness was determined by measuring the linear slope of the flexural stress-transverse displacement diagrams.

As shown in Table 9, the flexural stiffness reduction was almost similar in the neat and CNT-filled carbon/epoxy samples. As it is clear from Tables 8 and 9, the flexural strength and stiffness are higher in the samples containing nanoparticles. The flexural strength and stiffness of the samples decrease when placed in an acidic environment due to the acidic solution penetrating them.

Also, as it is clear in Tables 8 and 9, when the samples with and without nanoparticles are exposed to simultaneous mechanical loading and an acidic environment, there is a drop in the flexural strength and stiffness value, which is more in the samples with nanoparticles. The reason for this drop could be related to the agglomeration of nanoparticles. In order to meet the electrical conductivity of the specimens, a high volume percentage of the CNTs (about 10%) was used to fabricate test specimens.

5. Conclusions

The main objective of this research is to develop a conductive carbon/epoxy composite and investigate its electrical conductivity under mechanical loading exposed to acidic environments. The electrical conductivity of the samples was measured by a four-point method. To assess the simultaneous effect of the acidic environment and mechanical load on the electrical conductivity of the samples, the specimens with/without carbon nanotubes were placed in the acidic environment for 20 days under three different mechanical loads.

It was shown that the incorporation of 10% nanoparticles increased the electrical conductivity of the epoxy carbon composite above the acceptable DOE standard for electrodes.

For the samples exposed to an acid environment without applying mechanical load, the electrical conductivity showed a 2 and 3% reduction in the samples with and without carbon nanotubes, respectively. Also, for the samples subjected to the acidic environment and 40% of mechanical load, the electrical conductivity exhibited a 3.5 and 14% reduction in the samples with and without carbon nanotubes, respectively. For the samples exposed to 80% load and the acidic environment, the electrical conductivity was decreased by 9 and 26% in samples with and without carbon nanotubes, respectively. These results reveal the effect of nanoparticles in reducing the electrical conductivity of the samples exposed to the mechanical load and acidic environment.

Studies indicated that aging in an acidic environment with mechanical load prepared the stress corrosion conditions to the specimens and reduced their flexural stiffness and strength.

The results of this study can be exploited in constructing conductive composite electrodes to be used in petroleum environments such as crude oil electrostatic tanks.

References

Antunes, R.A., De Oliveira, M.C., Ett, G. and Ett, V. (2011), "Carbon materials in composite bipolar plates for polymer electrolyte membrane fuel cells: A review of the main challenges to improve electrical performance", *J. Power Sources*, **196**(6), 2945-2961. https://doi.org/10.1016/j.jpowsour.2010.12.041

Arani, A.G., Farazin, A. and Mohammadimehr, M. (2021), "The effect of nanoparticles on enhancement of the specific mechanical properties of the composite structures: A review research", *Adv. Nano Res.*, **10**(4), 327-337. https://doi.org/10.12989/anr.2021.10.4.327

Barton, R., King, J. and Keith, J. (2006), "Development and modelling of electrically conductive carbon filled liquid crystal polymer composites for fuel cell bipolar plate applications", *Proceedings of the Conference: The 6. International Symposium on New Materials for Electrochemical Systems*, Montreal, Canada, July.

Bourell, D., Leu, M.C., Chakravarthy, K., Guo, N. and Alayavalli, K. (2011), "Graphite-based indirect laser sintered fuel cell bipolar plates containing carbon fiber additions", *CIRP Annals*, **60**(1), 275-278. https://doi.org/10.1016/j.cirp.2011.03.105

Chen, S., Bourell, D.L. and Wood, K.L. (2004). "Fabrication of PEM fuel cell bipolar plates by indirect SLS", *Proceedings of the 2004 International Solid Freeform Fabrication Symposium*. http:// doi.org/10.26153/tsw/6990

Du, L. and Jana, S.C. (2008), "Hygrothermal effects on properties of highly conductive epoxy/graphite composites for applications as bipolar plates", *J. Power Sources*, **182**(1), 223-229. https://doi.org/10.1016/j.jpowsour.2008.03.071

Dweiri, R. and Sahari, J. (2007), "Computer simulation of electrical conductivity of graphite-based polypropylene composites based on digital image analysis", *J. Mater. Sci.*, **42**(24), 10098-10102. https://doi.org/10.1007/s10853-007-2092-x

Feng, T., Liu, N., Wang, S., Qin, C., Shi, S., Zeng, X. and Liu, G. (2021), "Research on the dispersion of carbon nanotubes and their application in solution-processed polymeric matrix composites: A review", *Adv. Nano Res.*, **10**(6), 559-576. https://doi.org/10.12989/anr.2021.10.6.559

Gholami, H., Shakeri, A. and Moosavi, S.H. (2015), "Preparation and properties investigation of conductive Polyaniline-Zinc Oxide nanocomposites", *J. Sci. Technol. Compos*, **2**, 7-12.

Gu, H., Zhang, H., Ma, C., Xu, X., Wang, Y., Wang, Z., Wei, R., Liu, H., Liu, C. and Shao, Q. (2019), "Trace electrosprayed nanopolystyrene facilitated dispersion of multiwalled carbon nanotubes: Simultaneously strengthening and toughening epoxy", *Carbon*, **142**, 131-140. https://doi.org/10.1016/j.carbon.2018.10.029

Guo, J., Chen, Z., Xu, X., Li, X., Liu, H., Xi, S., Abdul, W., Wu, Q., Zhang, P. and Xu, B.B. (2022), "Enhanced electromagnetic wave absorption of engineered epoxy nanocomposites with the assistance of polyaniline fillers", *Adv. Compos. Hybrid Mater.*, 1-9. https://doi.org/10.1007/s42114-022-00417-2

Guo, N. and Leu, M.C. (2012), "Effect of different graphite materials on the electrical conductivity and flexural strength of bipolar plates fabricated using selective laser sintering", *Int. J. Hydrogen Energy*, **37**(4), 3558-3566. https://doi.org/10.1016/j.ijhydene.2011.11.058

Hosseini, M. and Zandi Baghche Maryam, A. (2016), "Electromechanical response analysis of a rotating piezoelectric cylinder with functionally graded material under thermo-magnetic fields", *J. Sci. Technol. Compos.*, **3**(1), 59-72.

ASTM (American Society for Testing and Materials) (2015), *ASTM D3171-15-Standard Test Methods for Constituent Content of Composite Materials*, ASTM International West

Conshohocken, Filadelfia, Pennsylvania, U.S.A.

Jin, J., Lin, Y., Song, M., Gui, C. and Leesirisan, S. (2013), "Enhancing the electrical conductivity of polymer composites", *Eur*. *Polym*. *J*., **49**(5), 1066-1072. https://doi.org/10.1016/j.eurpolymj.2013.01.014

Johnson, B.A. (2009), "Thermally and electrically conductive polypropylene based resins for fuel cell bipolar plates", Doctoral dissertation, Michigan Technological University

Kakati, B., Sathiyamoorthy, D. and Verma, A. (2010), "Electrochemical and mechanical behavior of carbon composite bipolar plate for fuel cell", *Int*. *J*. *Hydrogen Energy*, **35**(9), 4185-4194. https://doi.org/10.1016/j.ijhydene.2010.02.033

Karimi, M., Ghajar, R. and Montazeri, A. (2017), "Investigation of nanotubes' length and their agglomeration effects on the elastoplastic behavior of polymer-based nanocomposites", *J*. *Sci*. *Technol*. *Compos*., **4**(2), 229-240.

Kim, Y.J., Shin, T.S., Do Choi, H., Kwon, J.H., Chung, Y.-C. and Yoon, H.G. (2005), "Electrical conductivity of chemically modified multiwalled carbon nanotube/epoxy composites", *Carbon*, **43**(1), 23-30. https://doi.org/10.1016/j.carbon.2004.08.015

Letti, C.J., Costa, K.A., Gross, M.A., Paterno, L.G., Pereira-da-Silva, M.A., Morais, P.C. and Soler, M.A. (2017), "Synthesis, morphology and electrochemical applications of iron oxide based nanocomposites", *Adv*. *Nano Res*., **5**(3), 215. http://doi.org/10.12989/anr.2017.5.3.215

Liao, S.H., Hsiao, M.C., Yen, C.Y., Ma, C.C.M., Lee, S.J., Su, A., Tsai, M.C., Yen, M.Y. and Liu, P.L. (2010), "Novel functionalized carbon nanotubes as cross-links reinforced vinyl ester/nanocomposite bipolar plates for polymer electrolyte membrane fuel cells", *J*. *Power Sources*, **195**(23), 7808-7817. https://doi.org/10.1016/j.jpowsour.2009.10.020

Liao, S.H., Hung, C.H., Ma, C.C.M., Yen, C.Y., Lin, Y.-F. and Weng, C.C. (2008), "Preparation and properties of carbon nanotube-reinforced vinyl ester/nanocomposite bipolar plates for polymer electrolyte membrane fuel cells", *J*. *Power Sources*, **176**(1), 175-182. https://doi.org/10.1016/j.jpowsour.2007.10.064

Luo, X., Yang, G. and Schubert, D.W. (2022), "Electrically conductive polymer composite containing hybrid graphene nanoplatelets and carbon nanotubes: Synergistic effect and tunable conductivity anisotropy", *Adv*. *Compos*. *Hybrid Mater*., **5**(1), 250-262. https://doi.org/10.1007/s42114-021-00332-y

Mighri, F., Huneault, M.A. and Champagne, M.F. (2004), "Electrically conductive thermoplastic blends for injection and compression molding of bipolar plates in the fuel cell application", *Polym*. *Eng*. *Sci*., **44**(9), 1755-1765. https://doi.org/10.1002/pen.20177

Modarresi-Alam, A.R., Soleimani, M., Pakseresht, M., Farzaneh-Jobaneh, E., Zeraatkar, V., Tabatabaei, F.A., Shabzendedar, S. and Movahedifar, F. (2016), "Preparation of new conductive nanocomposites of polyaniline and silica under solid-state condition", *Polym*. *Int*, **29**, 387-398.

Osman, A., Elhakeem, A., Kaytbay, S. and Ahmed, A. (2022), "A comprehensive review on the thermal, electrical, and mechanical properties of graphene-based multi-functional epoxy composites", *Adv*. *Compos*. *Hybrid Mater*., 1-59. https://doi.org/10.1007/s42114-022-00423-4

Pan, Y.X., Yu, Z.Z., Ou, Y.C. and Hu, G.H. (2000), "A new process of fabricating electrically conducting nylon 6/graphite nano-composites via intercalation polymerization", *J*. *Polym*. *Sci*. *Part B Polym*. *Phys*., **38**(12), 1626-1633. https://doi.org/10.1002/(SICI)10990488(20000615)38:12<1626: :AID-POLB80>3.0.CO,2-R

Park, S.M., Jung, D.H., Kim, S.K., Lim, S., Peck, D. and Hong, W.H. (2009), "The effect of vapor-grown carbon fiber as an additive to the catalyst layer on the performance of a direct methanol fuel cell", *Electrochimica Acta*, **54**(11), 3066-3072. https://doi.org/10.1016/j.electacta.2008.11.066

Razavi, M., Ghomi, M.T., Taheri-Behrooz, F. and Liaghat, G. (2019), "Effect of bending load on the electrical conductivity of carbon/epoxy composites filled with nanoparticles", *Iran*. *J*. *Polym*. *Sci*. *Technol*., **32**, 79-92.

Razavi, S.M., Sadollah, A. and Al-Shamiri, A.K. (2022), "Prediction and optimization of electrical conductivity for polymer-based composites using design of experiment and artificial neural networks", *Neural Comput*. *Appl*., **34**(10), 7653-7671. https://doi.org/10.1007/s00521-021-06798-7

Rhodes, S.M., Higgins, B., Xu, Y. and Brittain, W.J. (2007), "Hyperbranched polyol/carbon nanofiber composites", *Polymer*, **48**(6), 1500-1509. https://doi.org/10.1016/j.polymer.2007.01.038

Shen, C.H., Mu, P. and Yuan, R.Z. (2006), "Sodium silicate/ graphite conductive composite bipolar plates for proton exchange membrane fuel cells", *J*. *Power Sourc*., **162**(1), 460-463. https://doi.org/10.1016/j.jpowsour.2006.06.095

Shokrieh, M.M., Esmkhani, M., Vahedi, F. and Shahverdi, H.R. (2013), "Improvement of mechanical and electrical properties of epoxy resin with carbon nanofibers", *Iran*. *Polym*. *J*., **22**(10), 721-727. https://doi.org/10.1007/s13726-013-0170-2

Sun, L., Cui, R., Jalbout, A., Li, M., Pan, X., Wang, R. and Xie, H. (2009), "LiFePO4 as an optimum power cell material", *J*. *Power Sources*, **189**(1), 522-526. https://doi.org/10.1016/j.jpowsour.2008.10.120

Tabatabaee, M., Taheri-Behrooz, F., Razavi, S.M. and Liaghat, G.H. (2019), "Electrical conductivity enhancement of Carbon/ Epoxy composites using nanoparticles", *J*. *Sci*. *Technol*. *Compos*., **5**(4), 605-614.

Taherian, R., Golikand, A.N. and Hadianfard, M.J. (2011), "The effect of mold pressing pressure and composition on properties of nanocomposite bipolar plate for proton exchange membrane fuel cell", *Mater*. *Des*., **32**(7), 3883-3892. https://doi.org/10.1016/j.matdes.2011.02.059

Taherian, R., Hadianfard, M.J. and Golikand, A.N. (2013), "Manufacture of a polymer-based carbon nanocomposite as bipolar plate of proton exchange membrane fuel cells", *Mater*. *Des*., **49**, 242-251. https://doi.org/10.1016/j.matdes.2013.01.058

Testing, A.S.F. and Materials (2015), "Standard test method for flexural properties of polymer matrix composite materials", ASTM D7264.

Wafers, S. (2003), "Sheet resistance of thin metallic films with a collinear four-probe array 1", *Measurement*, **98**, 1-4.

Wang, Y. (2006), *Conductive thermoplastic composite blends for flow field plates for use in polymer electrolyte membrane fuel cells (PEMFC)*, Master's thesis, University of Waterloo, Waterloo, Canada.

Xu, X., Yao, F., Ali, O.A.A., Xie, W., Mahmoud, S.F., Xie, P., El-Bahy, S.M., Huang, M., Liu, C. and Fan, R. (2022), "Adjustable core-sheath architecture of polyaniline-decorated hollow carbon nanofiber nanocomposites with negative permittivity for superb electromagnetic interference shielding", *Adv*. *Compos*. *Hybrid Mater*., **5**(3), 2002-2011. https://doi.org/10.1007/s42114-022-00538-8

Yogeswaran, U. and Chen, S.M. (2008), "Multi-walled carbon nanotubes with poly (methylene blue) composite film for the enhancement and separation of electroanalytical responses of catecholamine and ascorbic acid", *Sensors Actuat*. *B Chem*., **130**(2), 739-749. https://doi.org/10.1016/j.snb.2007.10.040

CC