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Article

Use of Ni/CNT Particles as Additive Fillers in Ebonite Bipolar Plates for Proton-Exchange Membrane Fuel Cells

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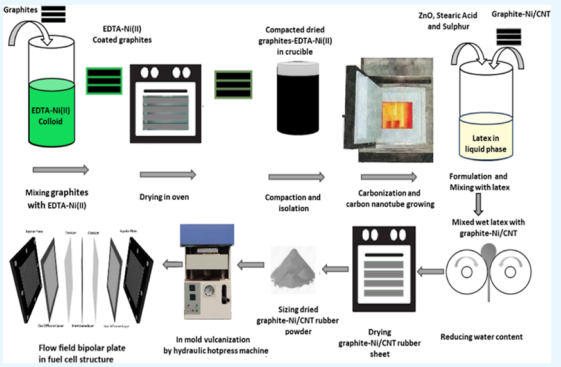
ABSTRACT: Composite-based bipolar plates are a chance to substitute graphite and metal bipolar plates because of their corrosion resistance and better chemical and mechanical properties, but they still have various electrical conductivity properties. One of the problems is the loading of filler that depends on matrix types which will affect the electrical conductivity of bipolar plates; ebonite has potential as a matrix in composite bipolar plates because it is obtained from elastomer or rubber. In this work, nickel and carbon nanotubes (CNT) that have a high electrical conductivity will be investigated as additive fillers on graphite particles to enhance the electrical conductivity of ebonite bipolar plates. The formulation and characterization of ebonite bipolar plates with graphite and graphite Ni/CNT and their various contents are the main objectives of this research. Characterization by scanning electron microscopy (SEM) for identification and morphology of compounds and ebonite bipolar plates and Raman spectroscopy for identification of the type carbon in Ni/CNT was performed. Some tests such as bending/flexural tests, corrosion tests, and resistance testing for interfacial contact resistance were conducted to study the properties and optimum of the composite materials. In this research, Ni/CNT particles were added as additive fillers with graphite to enhance the electrical conductivity of fillers in ebonite bipolar plates of proton-exchange membrane fuel cells and their impacts were studied. By through-plane testing, graphite fillers were added in ebonite bipolar plates with 65−75% w/w content, achieving electrical conductivity values from 22.3 to 34 S/cm. This is still below the technical target set by the US DOE for composite bipolar plates. But by adding 30% Ni/CNT filler contents in ebonite bipolar plates at various filler contents from 65% to 75% w/w, one can achieve electrical conductivities from 104.35 to 165.52 S/cm. Only 65% w/w filler with 30% Ni/CNT can meet the technical targets such as a bending/flexural test value of 25.58 N/mm^2, a corrosion test value of 0.894 μA/cm^2 (I_{corr}), and an interfacial contact resistance value of 3.09 mΩ cm^2. Further improvements are needed based on fuel cell applications, as indicated by some additional data that did not meet technical targets.



1. INTRODUCTION

Increasing the number of electric generators that use fossil fuel energy has become more obvious to meet the requirement for alternatives to the internal combustion engine to decrease the greenhouse effect and be environmentally friendly. The advantage of fuel cells as a power source is a direct conversion from chemical energy to electric energy, which has higher efficiencies than engines. These advantages include high energy efficiency, quiet operation without vibration, portability, almost zero emissions, and suitability for portable electric power, transportation, and stationary uses.[1,2]

Several fuel cell types such as proton-exchange membrane fuel cells (PEMFCs), alkaline fuel cells, solid oxide fuel cells, phosphoric acid fuel cells, molten carbonate fuel cells, and solid oxide fuel cells are classified based on fuel and electrolytes.[3] PEMFCs are the most promising candidates for different applications because they have higher efficiency than internal combustion engines and low operation temperature and produce only water and heat as a tool that can convert directly from hydrogen gas to electric energy. They offer high power density, produce zero emissions, and are scalable; therefore, PEMFCs are considered to be a primary power source for the future.[2]

PEMFCs typically operate between temperatures from 20 to 90 °C.[4] Hydrogen gas is fed as a reactant to the anode side, and oxygen is required as an oxidant to the cathode side in PEMFC applications. A single stack of PEMFCs has three

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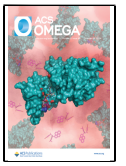

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components, namely, the membrane electrolyte assembly or MEA that consists of the proton electrolyte membrane, an anode and cathode layer of catalyst, and a layer of gas diffuser, seal gasket, and bipolar plate.[5]

Bipolar plates are the primary components of fuel cell stacks; they regulate the anode side's supply of hydrogen gas, the cathode side's air and water (liquid and vapor) drain out, thermal convection to the air, conduct electric current, and serve as the stack's structural framework. Significantly, a fuel cell stacks bipolar plates accounting for 60% of its weight and 30% of its manufacturing costs.[6−9]

Methods of bipolar plate production are classified into three methods, namely, molding, machining, and metal forming processes.[10] Processing by the molding method can be applied in composite bipolar plate production; for thicker graphite and metallic bipolar plates, the machining process can be used, while the metal forming process is required for low-thickness metallic plates.[11]

The Department of Energy (DOE) of the United States released the technical target requirements for properties of bipolar-plate PEMFCs that must be achieved as shown in Table 1. To develop bipolar plate materials, these technical target requirements must be met.

Table 1. Properties of Composites by the US DOE[14]

no	property	target
1	electrical conductivity	>100 (S cm^{-1})
2	thermal conductivity	>10 (W(mK)$^{-1}$)
3	flexural strength	>25 (MPa)
4	hardness	>50 (Shore A)
5	density	<5 (g/cm^3)
6	corrosion resistance	<1 μA cm^{-2}
7	interfacial contact resistance	0.1−0.2 ohm cm^2

The studies of some thermoset resin polymer types can be utilized for composite-based bipolar plates; the advantage of thermosets is their low viscosity; they can easily be added for electrical conductivity fillers in the thermoset material as a matrix at higher temperatures during the manufacturing process of bipolar plates.[12] The characteristics of these materials are low density, high corrosion resistance, and stability in dimensional and thermal that can be applied for manufacturing bipolar plates such as epoxy, phenolic resin, and vinyl ester.[13]

Natural graphite, synthetic graphite, expanded graphite, carbon fiber, carbon nanotubes, and carbon black are the fillers for electrical conductivity that can be mixed to develop a bipolar plate.[15] The thermosets can be used in the liquid phase which must be dissolved by solvents such as vinyl ester and resin or in a powder form such as epoxy and phenolic novolac when they are applied as matrices in bipolar plates.[12,16]

The application of thermoplastics has also been investigated for bipolar plate materials, but at first, these materials appeared less competitive because low fillers as the electrical conductivity filler can be loaded than thermoset resins.[17,18]

Some thermoplastics, such as polyamide, polyethylene, polypropylene, polyvinylidene fluoride, and polyphenylene sulfide, can be combined as matrices in bipolar plates to increase electrical conductivity and particle type selection, and the use of hybrid fillers can also improve the electrical conductivity up to 200 S/cm.[19]

Bipolar plates are made from polyvinylidene fluoride (PVDF) with 5% multiwalled carbon nanotubes (MWCNTs) and 35% graphite fillers that can exhibit excellent electrical conductivity, high flexural or bending strength, good hydrophobicity, and corrosion resistance.[20] Some studies have investigated poly(phenylene) sulfide (PPS), which has better mechanical properties and more fillers that can be loaded.[21] The manufacturing of bipolar plates which uses the injection molding method has a weakness because the maximum filler content that can be loaded to the matrices in the formulation is still lower than the material formulation that uses the compression method.[22,23] The weakness of the loaded filler content is influenced by the presence of nonpolar functional group backbones and the degree of polymer crystallinity.[24]

Thermosetting and thermoplastic polymer matrices that are loaded with graphite fillers are widely used in manufacturing composite bipolar plates. They have various improved properties, such as high electrical and thermal conductivity, corrosion resistance, mechanical strength, high chemical stability, and compatibility. In general, graphite-filled bipolar plates depend on the graphite size, distribution, and capability to mix uniformly with the polymer matrices.[25]

Ebonite is a rubber product and a type of thermosetting polymer that uses natural rubber from latex. In this research, the elastomeric properties of natural rubber can be utilized to facilitate the loading of more filler. Ebonite can be machined and made by adopting common engineering practices such as turning, milling, drilling, etc. Ebonite is an improved natural product that undergoes a vulcanization process to become hard rubber, and it is formulated with more than 30 phr content (parts per hundred rubber) of sulfur. Ebonite has the characteristics of high hardness, which swells minimally when applied to certain liquid media. Subsequently, ebonite has become an essential component of the rubber industry. In spite of hard plastics such as epoxy resin recently substituting for hard rubber, ebonite or hard rubber in the industry still has an important position because of its unique characteristics, mainly its chemical resistance, inertness, high strength, and good appearance; ebonite characteristics and its machinability have led to its wide use and applications in the process industry.[26] Ebonite has some specifications to be considered as a matrix for bipolar plates: a density of about 1.2 gr/cm^3, a shore hardness D of about 85, and a tensile strength between 23 and 35 MPa.[27] Therefore, ebonite can be an alternative material for manufacturing bipolar plates.

Several studies on the development of advanced filler materials have included nanocarbon materials such as carbon black, MWCNTs, single-walled carbon nanotubes (SWCNTs), carbon fiber, and expanded graphite, and their combinations are included to improve the electrical conductivity property of bipolar plates.[28]

The composite bipolar plates based on copper powder as an electrical conductivity filler and low-density polyethylene (LDPE) as a matrix have achieved an electrical conductivity of 0.117 S/cm by using the compression molding method.[29] Adding a little amount of metal powder to the filler with graphite in composite bipolar plates can enhance their electrical conductivity, and also adding carbon or ceramic fiber can increase their mechanical strength.[30] About 5% nanocopper is added to the nanocomposite, which results in good improvement; the density and shape of the nanocomposites regulate and increase the flexural strength. Metal nanoparticles are added to epoxy/graphite composites to

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create nanocomposites that are utilized as PEMFC bipolar plates.[31]

A bipolar plate with 30% nickel powder as an electrical conductivity filler and a high-density polyethylene (HDPE) matrix has achieved an electrical conductivity of about 83 S/cm.[32] The compression molding method for making composite bipolar plates based on graphite-nanocopper fillers and epoxy has increased the electrical conductivity by about 0.833 S/cm.[31] Using the injection molding method in making composite bipolar plates has been investigated, and an electrical conductivity of 112 S/cm is achieved with 30% copper−graphite and HDPE as a matrix.[33] Bipolar plates based on PP polymer as a matrix and a combination of carbon and Pb as filler materials achieved an electrical conductivity of about 8.42 S/cm.[34] A combination of 42% natural flake graphite, 10% carbon black, and 8% copper fiber that is filled to polymer composites of bipolar plates with epoxy resin as a matrix was developed to improve the properties of electrical conductivity, exhibited electrical conductivity of about 169 S/cm.[35]

The application of metal matrix nanocomposites demonstrates the significant benefit of fusing matrices and reinforcing material features to create a sophisticated material with extraordinarily high-level qualities specifically suited to the desired function. But it is important to recognize that a number of variables may affect the characteristics of the nanocomposite, including the method of manufacture, the effectiveness of the reinforcement's dispersion, the way the components interact, and the combination of the matrices and reinforcement materials.[36]

The growth of MWCNTs is investigated using two chemical vapor deposition (CVD) methods: ambient pressure thermal CVD and low-pressure plasma-accelerated CVD. Hydrocarbon or other carbon-bearing precursors are employed in the CVD process for the mass manufacture of CNTs using a catalyst with any of the transition metals, such as iron pentacarbonyl, ferrocene, Mo, Co, or Ni.[37]

EDTA or ethylenediaminetetraacetic acid is a synthetic compound mostly utilized as a chelating agent to form complexes with metal ions due to its multiple amine and carboxylate groups; it is useful in a variety of applications because it can bind to metal ions.[38] In this research, Ni particles bound in EDTA become chelate EDTA-Ni (II) for coating graphite; their combination is dried in an oven, calcined, and pyrolyzed at 900 °C in a furnace. In this process, nickel particles are used as catalysts for growing carbon nanotubes and EDTA as a carbon source. They will be used as additive fillers with graphite as an electrical conductivity filler for application in bipolar plate fuel cells. The graphite-Ni/CNT particles are utilized as electrical conductivity fillers with ebonite as a binder/matrix to become ebonite bipolar plates with graphite-Ni/CNT. The purpose of graphite coating is to facilitate the loading of graphite into the composite matrix and also to enhance the electrical conductivity properties of the bipolar plate.

2. EXPERIMENTAL SECTION

2.1. Materials. The commercial G-Seal*graphite was purchased from Mi SWACO; technical-grade EDTA-4Na ex BASF and NaOH (sodium hydroxide) ≥ 99% were obtained from Merck, and nickel(II) chloride hexahydrate or $NiCl_2 \cdot 6H_2O$ 99.9% was obtained from Sigma-Aldrich. In addition, other materials used include latex with 60% dry rubber content from PT. Inti Permata Restu, ZnO from Merck, stearic acid from Merck, and sulfur powder 99.9% with mesh 325 from MIDAS ex Korea.

2.2. Ebonite Formulation. Charles Goodyear invented the ebonite formula that the sulfur contents are set to about 30−80 phr, the higher sulfur content will more bond in rubber compounds, which results in increased density and hardness. In simple terms, the basic formula of an ebonite bipolar plate compound contents needs

- 167 phr of latex composed based on 100 phr of natural rubber,
- 5 phr of ZnO,
- 2 phr of stearic acid, and
- 50 phr of sulfur as a curing agent.

Natural rubber can be obtained from liquid latex, which contains dry rubber content/DRC 60%, as the specification set by the standards SNI ISO 2004:2017.[39]

A filler is involved in rubber compounds because it is intended to improve the technical capabilities of rubber products, for example, hardness, tensile strength, tear resistance, chemical resistance, material costs, and others (usually carbon black, calcium carbonate, etc.).

2.3. Making the Graphite-Ni/CNT Filler. The colloid of EDTA-Ni (II) was used for coating the graphite particles to make graphite-Ni/CNT particles; they were then dried, sized to form EDTA-Ni (II) powder, compacted, and isolated to remove excess oxygen in a crucible, respectively, finally followed by drying or pyrolysis treatment. This process resulted in Ni/CNT-layered graphite powder when nickel nanoparticles catalyzed the carbon source from EDTA-Ni (II) to grow nanocarbon in the pyrolysis process at 900 °C.[40]

2.4. Graphite-Ni/CNT as Fillers for Ebonite Bipolar Plates. Ebonite bipolar plates with graphite or graphite-Ni/CNT fillers can be manufactured through some stages, namely,

a. Formulation for graphite and graphite-Ni/CNT ebonite bipolar plates

The ebonite bipolar plate formula with various variations of graphite with filler contents of 65, 70, and 75% is shown in Table 2.

The ebonite bipolar plate formula with various variations of graphite-Ni/CNT with a filler content of 65% is shown in Table 3.

Table 2. Formulation of Ebonite Bipolar Plates with Graphite Fillers

materials	formulation (phr)[a]		
	65G	70G	75G
Matrix Content, 35%			
natural rubber (latex, 60% DRC)	100	100	100
ZnO	5	5	5
stearic acid	2	2	2
sulfur	50	50	50
subtotal of matrices (phr)	157	157	157
Graphite Filler Contents			
graphite	291.6	366.3	471
total	448.6	523.3	628.0

[a]Here 100 phr of natural rubber = 60% latex mass; 65G, 70G, and 75G indicate that the graphite contents are 65, 70, and 75%, respectively.

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Table 3. Formulation of Ebonite Bipolar Plates with Graphite-Ni/CNT Fillers

materials	formulation (phr)[a]			
	65N1	65N2	65N3	65N4
Matrix Content, 35%				
natural rubber (latex, 60% DRC)	100	100	100	100
ZnO	5	5	5	5
stearic acid	2	2	2	2
sulfur	50	50	50	50
subtotal of matrices (phr)	167			
Filler of Graphite-Ni/CNT contents, 65%				
graphite	262.4	233.3	204.1	174.9
Ni/CNT	29.2	58.3	87.5	116.6
subtotal of fillers (phr)	291.6			
total	448.6	448.6	448.6	448.6

[a]Here 65N1, 65N2, 65N3, and 65N4 values explain that the filler of the ebonite bipolar plate has about 65% content with 10, 20, 30, and 40% of Ni/CNT.

b. Making the ebonite bipolar plate compound

In manufacturing the compound, ZnO powder, stearic acid, sulfur, and the filler were mixed with latex in the wet/liquid phase.[41] Vulcanization is a crucial critical process in manufacturing of rubber compounds, where a cross-linking reaction occurs between rubber molecules and the sulfur vulcanization agent. There are several stages in this process:

- mixing the powder of ZnO, stearic acid, sulfur, and graphite as fillers based on Table 2 for graphite-ebonite bipolar plates and graphite-Ni/CNT as fillers based on Table 3 for graphite-Ni/CNT-ebonite bipolar plates;
- mixing with latex in the wet/liquid phase with 60% dry rubber content;
- removing the water in the compound by compression with a rolling open mill; (d) drying in an oven at 80 °C for at least 5 h;
- sizing to make the compound powder;
- compressing the compound powder in a mold;
- finally, vulcanization by hot-pressing at 20 MPa for 1 h.
- Making specimens for testing and characterization

Every type of specimen will be attempted to meet the requirement for testing and characterization of the adopted standards.

2.5. Characterization of the Graphite-Ebonite Bipolar Plates. Scanning electron microscopy is a technique studied in materials science to obtain the detailed microstructure and composition images of the surfaces of samples at the microscopic level (JSM-6510 JEOL), and Raman spectroscopy is utilized to investigate carbon nanoparticles using a HORIBA LabRAM HR Evolution Raman microscope instrument.

Measurement of the electrical resistance of the bipolar plate uses a milli/micro-ohmmeter low-resistance tester instrument using the in-plane measurement method. Based on the electrical protocol for composite materials, the US Fuel Cell Council (USFCC), resistance measurements can be carried out using a through-plane test.[42] The instrument for this measurement is YAOREA YR2050.

The electrical conductivity is the inverse of the resistivity ($\sigma = 1/\rho$). The resistivity results from measuring the material's resistance and is calculated based on Ohm's law.

$$\sigma = \frac{1}{\rho}, \qquad \text{where } \rho = R\frac{A}{l} \tag{1}$$

The bending/flexural test visually determines the quality of the material. In addition, the bending test is used to measure the strength of the material based on loading and the elasticity of the specimen. The test standard is ASTM D 790 with a 5 kN UTM tool, Shimadzu AG-X.

The electrical contact resistance of the stack's power terminal electrical contact and the polymer electrolyte membrane fuel cell (PEMFC) bipolar plate are important components to optimize. The conductivity and roughness of the two surfaces in contact have an impact on the contact resistance.[43,44]

The interfacial contact resistance (ICR) test is based on the electrical protocol for composite materials by the US Fuel Cell Council (USFCC). The sample with dimensions of 2.54 cm × 2.54 cm × 0.2 cm is sandwiched between a carbon paper plate as a gas diffusion layer and a copper electrode, then treated with a constant pressure of 6.895 MPa (1000 psi), and measured using an ohm micro/milli-scale ohmmeter. The ICR measurement between bipolar plate samples and carbon paper is approximated by eq 2[45]

$$R_{c/p} = \frac{1}{2}(R_1 - R_2) \tag{2}$$

The PEMFC environment simulated in this research is 0.5 M H_2SO_4 + 2 ppm of HF solution, which is quite aggressive, representing the environmental conditions of PEM fuel cells with a pH between 3 and 4.[46−48]

The fillers of ebonite bipolar plates consisting of both graphite and nickel undergo corrosion in the operation of PEMFCs. In aqueous environments in the presence of certain electrolytes, graphite can undergo electrochemical reactions that cause material degradation. The mechanism of carbon corrosion is given in eq 3[49]

$$C(s) + 2H_2O \rightarrow CO_2(g) + 4H^+ + 4e^- \tag{3}$$

Acidic conditions in PEMFCs also can lead to chemical corrosion, degrading the graphite structure. The instrument for corrosion testing is a potentiostat/galvanostat/EIS, Analyzer test equipment AMETEK PARSTAT 4000A. The density of corrosion current was identified by Tafel extrapolation in the polarization curves. By extrapolating linear sections based on the log current vs potential plot, corrosion current density can be determined on a bipolar plate.[50] Metal (nickel) corrosion is a process in which metals deteriorate as a result of chemical reactions with their environment (PEMFCs).

Then the corrosion rate in years can use Faraday's law as shown in eq 4

$$\text{Corrosion Rate(mmpy)} = 3.27 I_{corr}(\text{EW})/d \tag{4}$$

where I_{corr} is in mA/cm^2 unit, EW is the equivalent of weight (atomic weight/valence number), and d is the density.

3. RESULTS AND DISCUSSION

3.1. Scanning Electron Microscopy. Figure 1 shows the planar graphites coated by agglomerated particles of Ni/CNT; their size as fillers is about <50 μm. The average size of the ebonite bipolar plate compound particles is around 100 μm, as shown in Figure 2. Figure 3 shows that a transition process has occurred in the form of the material from the compound to the vulcanizate of ebonite bipolar plates by hot-press vulcanization.

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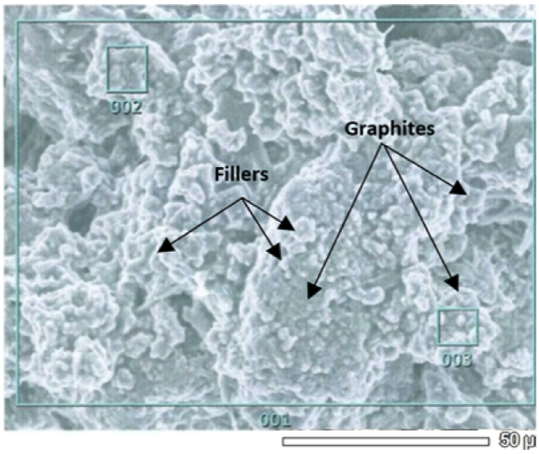

Figure 1. SEM of coated graphite by Ni/CNT.

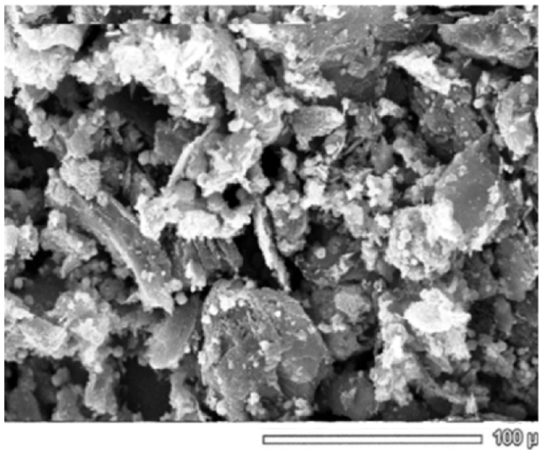

Figure 2. Compound powder of ebonite bipolar plates.

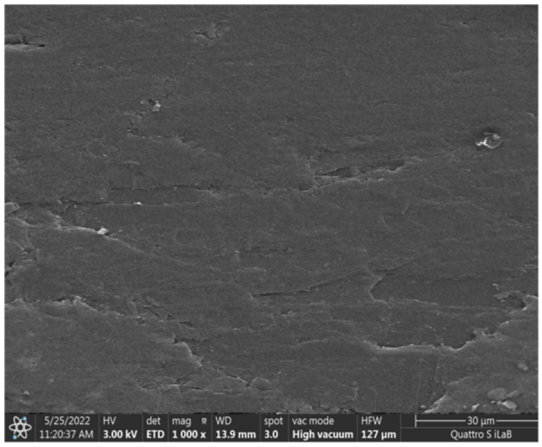

Figure 3. Vulcanizate of ebonite bipolar plates.

3.2. Raman Spectroscopy. Figure 4 displays the Raman spectra. It shows that the peak location RBM (cm^{-1}) at 308 cm^{-1} is closely related to the diameter d (nm) of the SWCNT of Ni/CNT particles.

The peak of SWCNTs is in along the low-wavenumber region of radial breathing mode or RBM (100−300 cm^{-1}). SWCNTs expand and contract in a diametric way in the RBM mode. On the sample, the desired SWCNT diameter can be assessed. It is calculated that the SWCNT diameter in the Ni/

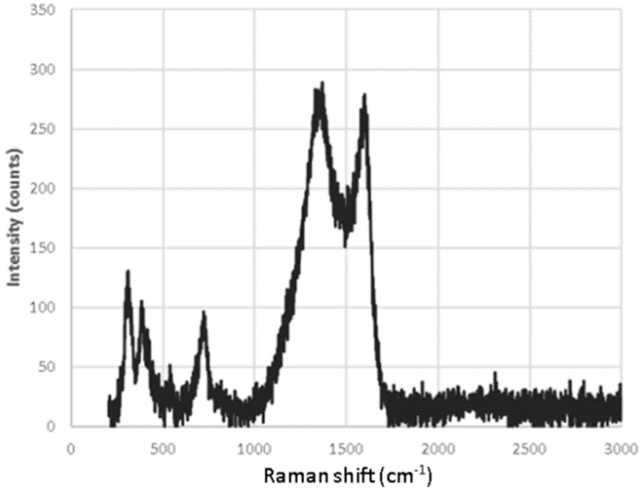

Figure 4. Raman spectroscopy of Ni/CNT.

CNT particles is approximately 0.805 nm by using the $d = 248/\omega_{RBM}$ approach.

The G band arises from the in-plane vibration of sp^2-bonded carbon atoms. It is a primary feature in the Raman spectra of graphitic materials, as shown in Figure 5. The Raman spectra

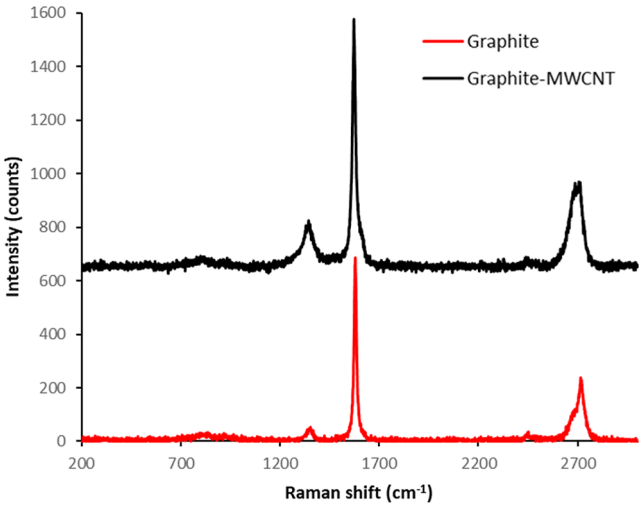

Figure 5. Raman spectroscopy of graphite and graphite-30% Ni/CNT.

of graphite-Ni/CNT particles in Figure 5 showed that they contained three peaks that corresponded to the carbon nanotubes: 1353 cm^{-1} for the D band, 1578 cm^{-1} for the G band, and 2697 cm^{-1} for the 2D band. The D band in MWCNTs indicates the presence of defects or impurities, which is the ratio of the intensity of the G band to the intensity of the D band. A low degree of crystallinity or defects was indicated by the ID/IG value of 0.114, which is the ratio of the intensity of the G band to the intensity of the D band.

The difference between graphite and graphite-CNT spectra is that the 2D band of graphite in highly ordered graphite has a single sharp peak, and the 2D band of graphite-30% Ni/CNT is broader due to the curvature and multiwalled structure.

3.3. Electrical Conductivity. The first step for characterization is the electrical conductivity property. The content of Ni/CNT that will be selected must meet the requirement for the properties of the composite by the US DOE as shown in

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Table 1. The electrical conductivity of graphite-ebonite bipolar plates with various graphite filler contents of 65, 70, and 75 is still unable to exceed the technical target of bipolar plate electrical conductivity which is set by the US DOE.

Increasing the graphite filler content influenced the electrical conductivity of graphite-ebonite bipolar plates. Comparisons of ebonite with composite graphite bipolar plates that were tested by the through-plane method are shown in Table 4.

Table 4. Ebonite Is Compared with Composite Bipolar Plates

matrices	electrical conductivity	references
ebonite	34.04	
PP/G	4.76	51
phenolic resin	29.00	52
FE-PP	42.40	53
PPS	54.00	54
epoxy	19.00	55

Although their electrical conductivity was below the technical target of the US DOE, the composite bipolar plates were commercialized.

Then, bipolar plates were made using 65% filler of graphite with various Ni/CNT filler additives, which was determined based on the lower content in this research. The electrical conductivity measurement increases electrical conductivity as adding Ni/CNT content is shown in Table 5.

Table 5. Electrical Conductivity of Ebonite Bipolar Plates with Various Ni/CNT Contents

no	content of Ni/CNT (%)	electrical conductivity, S/cm
1	10	7.75
2	20	39.37
3	30	104.35
4	40	110.95

Figure 6 shows that the electrical conductivity for ebonite bipolar plates in proportion tends to increase the Ni/CNT content. The content of 30% and 40% Ni/CNT showed their electrical conductivity that exceeded 100 S/cm of the technical target of the US DOE. So, the content of 30% Ni/CNT can be used to investigate the effect of graphite-Ni/CNT variation from 65% to 75% filler content.

The electrical conductivity of the ebonite-bipolar plate with various contents of graphite-30% Ni/CNT filler can be seen in Table 6.

Table 6. Electrical Conductivity of Ebonite-Bipolar Plates with Graphite-30% Ni/CNT Fillers

sample	graphite-30% Ni/CNT filler	σ (S/cm)
65N3	65%	104.35
70N3	70%	145.45
75N3	75%	165.52

Comparisons of composite graphite bipolar plates with CNT/metal mesh additives that were tested by the through-plane method can be seen in Table 7.

Table 7. Ebonite-Graphite Bipolar Plates with 30% Ni/CNT and Composite Graphite with Additive Fillers

matrices	additive filler	electrical conductivity		
		through-plane	in-plane	references
ebonite	Ni/CNT	104.35		
epoxy	CNT	70.00	120	18 and 56
phenolic resin	CNT	30.00	165	57
polypropylene	CNT	21.00	340	58 and 59
hybrid polymer	Cu mesh	36.70	156>	60
hybrid polymer	Al mesh	22.90	156>	60
epoxy	copper fiber		111	35

The ebonite bipolar plate with graphite-30% Ni/CNT is better than the composite graphite bipolar plate with additive CNTs because of the combination of Ni/CNT; each particle has high electrical conductivity.

3.4. Density. The density of the graphite-ebonite bipolar plate increased with the addition of a graphite filler, as shown in Figure 7. The density of the ebonite bipolar plate with graphite-Ni/CNT also increases when the filler content is added. The density of the graphite-30% Ni/CNT bipolar plate was higher than the density of the graphite-ebonite bipolar plate because of the content of Ni particles.

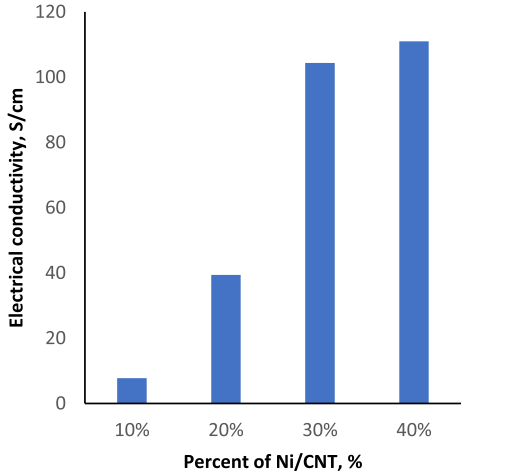

Figure 6. Electrical conductivity of ebonite bipolar plates with 65% filler of graphite upon varying the Ni/CNT content.

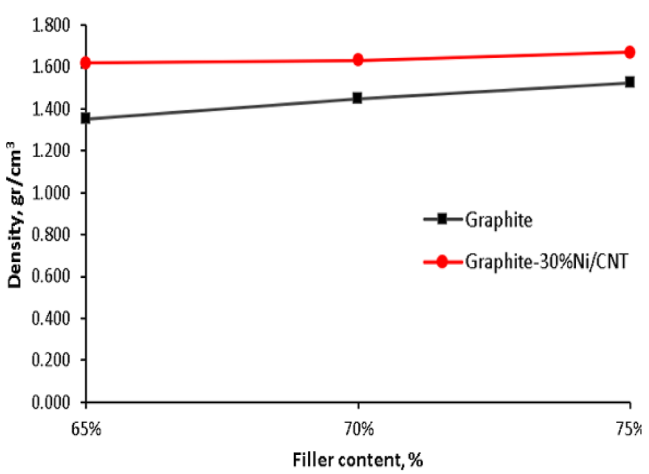

Figure 7. Variation in the density of graphite/graphite-30% Ni/CNT ebonite bipolar plates.

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3.5. Hardness. There are tests for the hardness of ebonite bipolar plate filler-type graphite and graphite with 30% Ni/CNT based on the ASTM D2240 standard using a Durometer hardness tool. The results of hardness testing of the ebonite bipolar plate that was filled with both graphite and graphite with 30% of Ni/CNT fillers have achieved >50 shore A from the US DOE technical target.

3.6. Interfacial Contact Resistance Bipolar Plates. The results of resistance testing for calculating the interfacial contact resistance of ebonite bipolar plates have not met the technical target set by the DOE of 10 mΩ cm^2 for ebonite bipolar plate samples with the graphite filler, as shown in Tables 8 and 9 for ebonite bipolar plate samples with the graphite-Ni/CNT filler.

Table 8. Interfacial Contact Resistance Properties of Ebonite Bipolar Plates Using Graphite Fillers

sample	ICR	
	Ω cm^2	mΩ cm^2
65G	0.1290	129.0
70G	0.0210	21.0
75G	0.0155	15.5

Table 9. Interfacial Contact Resistance Properties of Ebonite Bipolar Plates with Graphite-30% Ni/CNT Fillers

sample	ICR	
	W cm^2	mW cm^2
65N3	0.00309	3.09
70N3	0.00282	2.82
75N3	0.00092	0.92

Furthermore, the ICR of the ebonite bipolar plate with graphite-30% of Ni/CNT fillers is shown in Table 9. The resistance of ebonite bipolar plates with 65, 70, and 75% content of graphite-30% Ni/CNT fillers met the technical target of the US DOE.

3.7. Bending/Flexural Strength. Bending test results of the ebonite bipolar plate with graphite and graphite-30% Ni/CNT filler types shown in Table 10 indicate that a composite material, such as a polymer matrix filled with particles, can influence and alter the properties of stress and strain when the filler content is increased. Fillers can increase the stiffness (elastic modulus) of the composite. A stiffer material can lead to a reduction in strain for a given load, which might reduce the overall stress on the matrix itself.

Table 10. Bending Test Results of Ebonite Bipolar Plates with Graphite and Graphite-30% Ni/CNT Fillers

samples	Max_Force, *N*	N/mm^2
65G	55.97	48.81
70G	52.14	39.88
75G	37.99	27.67
65N3	35.03	25.58
70N3	53.91	25.10
75N3	28.44	16.38

The following is a bending/flexural test graph according to Figure 8, indicating that the samples of 65G to 75G met the US DOE technical target flexural above 25 N/mm^2, and in Figure 9, only samples of 65N3 and 70N3 with the addition of particle 30% Ni/CNT met the US DOE technical target.

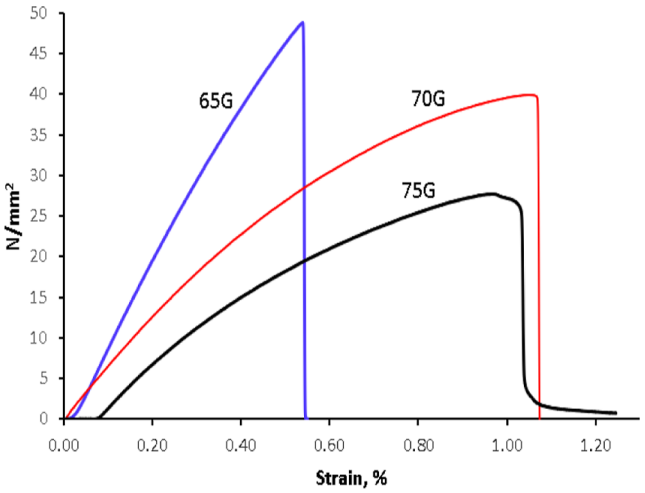

Figure 8. Effect of the graphite filler content on the flexural/bending property of ebonite bipolar plates.

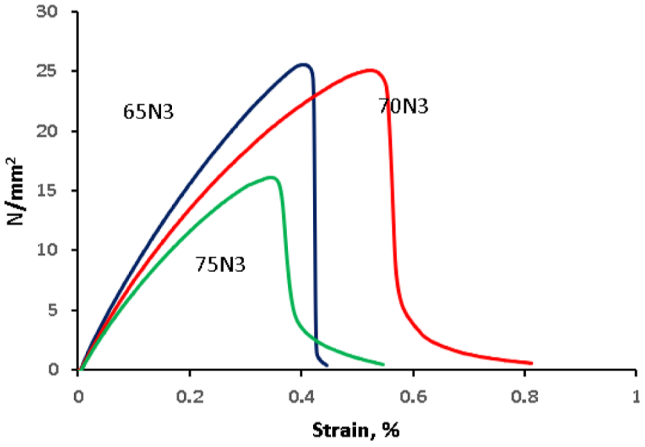

Figure 9. Effect of graphite with 30% Ni/CNT filler content on the flexural/bending property of ebonite bipolar plates.

3.8. Corrosion Test. The testing results show that the ebonite bipolar plate samples with graphite filler types 65G and 70G have met the US DOE technical target for corrosion resistance. To obtain the corrosion rate, as used in eq 4, the ebonite bipolar plate with the graphite filler or sample 65G has 0.001993214 mmpy (mm per year) and 70G has 0.003157036 mmpy, respectively.

The corrosion resistance of ebonite bipolar plates made from the graphite filler also shows its ability to be comparable to the corrosion resistance of composite bipolar plates, as mentioned in Table 11.

In this research, due to the highly hydrophobic nature of carbon nanotubes, the polymer/CNT nanocomposites also have high chemical stability to improve corrosion resistance.[65] The corrosion resistance of the ebonite bipolar plate with graphite-30% Ni/CNT fillers is shown in Table 12.

Only the ebonite bipolar plate sample 65N3 with graphite-30% Ni/CNT filler type met the technical target. It is suspected that the significant 35% matrix content of ebonite can play a role in protecting the filler of graphite-30% Ni/CNT from the oxidation process.

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Table 11. Corrosion Resistance of Some Composite Bipolar Plates

matrices	I_{corr}, $\mu A/cm^2$	reference
ebonite	0.89	
resin	0.69	[A[61,62]]
PVDF	0.97	[A[62,63]]
ABS	0.50	[A[63,64]]
PEEK	1.05	[A[64,65]]

Table 12. Corrosion Test Results of Ebonite Bipolar Plate with Graphite-30% Ni/CNT Filler

samples	I_{corr}, $\mu A/cm^2$
65N3	0.894
70N3	1.122
75N3	1.349

The corrosion rate of ebonite bipolar plates with graphite-30% Ni/CNT filler or sample 65GN3 is 0.009624161 mmpy.

4. CONCLUSIONS

After a series of tests and discussions were carried out, the following conclusions were drawn:

- Thegraphite filler in ebonite bipolar plates is not able to meet the technical target for electrical conductivity.
- Ni/CNT particles as additive fillers with about 30% content are able to enhance the electrical conductivity properties of ebonite bipolar plates beyond the technical target of composite bipolar plate electrical conductivity.
- At least 30% of Ni/CNT of filler contents in ebonite bipolar plates at various filler contents from 65% to 75% w/w can achieve electrical conductivities from 104.35 to 165.52 S/cm.
- At least 65% w/w filler with 30% of Ni/CNT can meet the technical targets such as a bending/flexural test value of 25.58 N/mm^2, a corrosion test value of 0.894 $\mu A/cm^2$, and an interfacial contact resistance value of 3.09 $m\Omega\ cm^2$.
- Some additional data did not meet technical targets; hence, ebonite bipolar plates with graphite Ni/CNT filler can be improved to find better bipolar plates.

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Notes

The authors declare no competing financial interest.

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