

Research Article

Iron loading effect on graphite/carbon black/polypropylene composite of bipolar plate's mechanical and electrical properties

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Abstract

In polymer electrolyte membrane fuel cells (PEMFC), the materials used for bipolar plates must meet stringent requirements, including high mechanical strength, lightweight, low cost, ease of fabrication, low contact surface resistance, and mechanical stability. Moreover, the performance and properties of these plates are significantly influenced by the materials and their compositions. In this study, all raw materials were used in powder form. The conductive fillers employed were graphite (G), carbon black (CB), and iron (Fe), while polypropylene (PP) was utilized as the polymer binder. The overall composition ratio of fillers to binder was maintained at 80:20 by weight. Within the filler component, the composition was adjusted within the following ranges: 40–50 wt% for graphite, 25 wt% for carbon black, and 5–20 wt% for iron. The fillers were initially blended using a ball milling process to ensure uniform dispersion. Prior to this, compounding of the fillers and the polymer binder was carried out using an internal mixer machine. The compounded material was then processed via hot compression moulding to fabricate the G/CB/Fe/PP composite plates. Subsequently, various performance evaluations were conducted, including in-plane electrical conductivity, mechanical properties (flexural strength and shore hardness), bulk density, and microstructural analysis. Among the samples tested, the composite containing 15 wt% Fe exhibited the most favourable properties, achieving the highest in-plane electrical conductivity of 367.59 S/cm, flexural strength of 44.57 MPa, bulk density of 1.69 g/cm³, and shore hardness of 60.2, respectively.

Keywords: bipolar plate, graphite, carbon black, ferum, conductive filler, and PEMFC

Introduction

The bipolar plate is a critical component in proton exchange membrane (PEM) fuel cell stacks, significantly contributing to both the system's performance and overall manufacturing cost. To ensure optimal functionality and durability, bipolar plates must meet a range of stringent requirements which include high electrical conductivity to minimize ohmic losses, sufficient gas permeability to prevent cross-leakage between reactant gases, and a combination of lightweight characteristics with adequate mechanical strength and rigidity to withstand compressive loads. Additionally, they must exhibit high thermal stability and conductivity to facilitate heat management within the cell, along with superior corrosion resistance to endure the harsh

electrochemical environment during operation. In recognition of these essential performance criteria, the U.S. Department of Energy (DoE) has established specific target requirements for the physical, electrical, and chemical properties of bipolar plates. These standardized specifications are summarized in **Table 1** [1-3].

In general, three types of materials are commonly used for manufacturing bipolar plates: metals, pure graphite, and conductive polymer composites (CPC) [2-5]. Among these, metal bipolar plates are often preferred due to their ability to meet the demands of high-power densities at a relatively low cost. For example, stainless steel has been widely used, for thin metal sheets are not only cost-effective but also

Table 1. Properties of bipolar plates in 2025 [1-3]

Properties	Standard Value
Electrical conductivity	>100 S/cm
Thermal conductivity	>10 W/(mk)
Flexural strength	>25 Mpa
Shore hardness	>50
Weight	<0.4 kg/kW
Bulk Density	<5 g/cm ³
Gas permeability	<2 × 10 ⁻⁶ cm ³ cm ⁻² s ⁻¹ at 80 °C

provide sufficient mechanical strength to meet the standard requirements of bipolar plates [6]. Additionally, the forming processes for metallic plates, such as hydroforming and stamping, are well-established. However, the primary drawback of metallic bipolar plates is their susceptibility to corrosion. According to Heinzl and Mahlendorf (2009), corrosion occurs at both the anode and cathode. On the cathode side, the presence of air and positive potentials promotes the formation of metal oxide layers, which subsequently increases the electrical resistance of the plates. Conversely, the environment reduction at the anode can cause the reduction of oxide layers, formation of metal hydrides, and dissolution of metals into the product water [7, 8]. The activity of the electrode catalysts may be adversely affected by this metal ion release, which may also contaminate the polymer electrolyte membrane [9]. Overall, metallic bipolar plates face several electrochemical corrosion challenges, including chemical instability in the highly corrosive environments of proton exchange membrane fuel cells (PEMFCs). This causes both corrosion and formation of thin oxide layers on their surfaces.

Pure graphite bipolar plates are widely recognized for their long service life, particularly in stationary applications where the expected lifespan ranges from 40,000 to 80,000 hours. These plates offer several advantages, including excellent chemical resistance, acceptable thermal and electrical conductivity, and low density. However, the fabrication of complex flow and cooling channels on their surfaces is a challenging and time-consuming process, contributing significantly to the overall production cost [9,10]. Additionally, pure graphite is inherently porous, brittle, and exhibits high gas permeability, necessitating the application of coatings to enhance its permeability [11]. The material's brittleness not only complicates machining but also requires the plates to have a thickness of several millimetres to ensure mechanical stability. This, in turn, increases the overall volume and weight of the fuel cell stack, making it less suitable for applications where compactness and lightweight are critical [3,11].

Conducting Polymer Composites (CPCs) are a class of organic polymers, also known as conducting polymer materials. Due to their unique optical and electrical properties, CPCs have become widely utilized in various applications, demonstrating characteristics similar to those of metals and inorganic semiconductors. At the same time, CPCs retain key polymer properties such as ease of processing, flexibility, and straightforward synthesis. Generally, CPCs are developed by combining conductive fillers with conventional polymers, resulting in composites that have attracted significant attention, particularly in the fabrication of diverse CPC types over the past few decades. CPCs are commonly categorized based on the type of charge carriers responsible for electrical conduction, such as ions, conductive nanomaterials, or delocalized π -electrons [12, 13, 14]. In particular, CPC-based bipolar plates bonded with polymers are well-suited for achieving the desired multifunctional properties. These composites are typically comprised of a high concentration of conductive carbon-based materials including carbon black, carbon nanotubes, and natural or synthetic graphite powders—which enhance electrical conductivity. Polymers, acting as binders, are either thermoplastics or thermosets [15, 16, 17, 18, 19]. CPCs offer an optimal combination of properties, including excellent electrical conductivity, lightweight characteristics, favourable mechanical strength, and cost-effectiveness. A significant advantage of CPCs lies in the ability to tailor their final properties by selecting appropriate conductive fillers and polymer matrices. The choice of conductive filler is critical, as different fillers impart distinct properties, significantly influencing the performance and conductivity of the resulting CPC [20, 21, 22].

One of the most common methods to produce conductive polymer composites (CPCs) is by incorporating hybrid conductive filler materials. In such systems, the CPC consists of more than two different conductive fillers along with a polymer binder, combined at the microscopic scale. Typically, one of the fillers is inorganic while the other is organic in nature. This differs from traditional composites, where the constituents are combined at the macroscopic level (ranging from micrometres to millimetres). Mixing at the microscopic scale, however, results in a more homogeneous material that not only exhibits characteristics intermediate between the original phases but may also demonstrate entirely new properties.

Another approach to enhance the performance of CPCs involves the use of multifilter systems [13, 16, & 23]. Considerable research has been conducted on the effects of combining various types of conductive fillers, such as graphene (G), carbon black (CB), and iron (Fe), as illustrated in **Figure 1**. Studies have

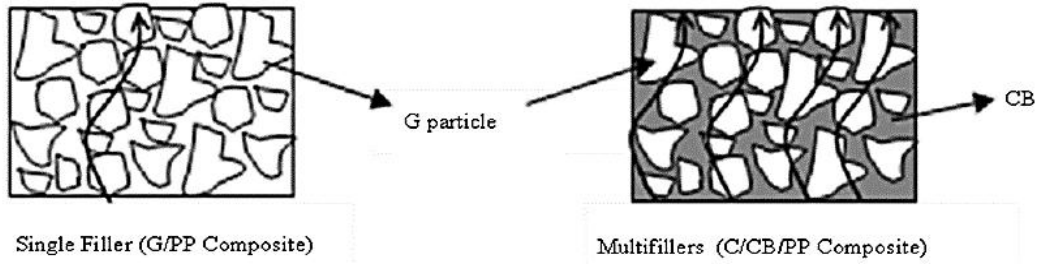


Figure 1. Illustration of single filler and multifiller [24]

shown that the combination of graphene with other conductive fillers is an effective strategy for developing G/CB/Fe polymer composites with enhanced electrical conductivity and improved mechanical properties.

The objective of this study was to investigate the effect of iron (Fe) loading on the mechanical and electrical properties of a composite bipolar plate. The bipolar plate was fabricated using graphite (G), carbon black (CB), iron (Fe), and polypropylene (PP). The study was to evaluate the capability of the developed composite to meet the performance requirements established by the U.S. Department of Energy (DOE) for bipolar plates in proton exchange membrane fuel cells (PEMFCs). To accomplish this, the influence of varying filler loadings on key performance indicators including in-plane electrical conductivity, flexural strength, Shore hardness, bulk density, and microstructural characteristics (examined through scanning electron microscopy, SEM) was systematically investigated and analysed. Based on the findings, the potential of the fabricated bipolar plate to fulfil or exceed the DOE targets for PEMFC bipolar plate applications was assessed.

Materials and Methods

The production process of the G/CB/Fe/PP composite bipolar plates involved several key steps, including raw material pre-mixing, compounding, pulverizing, and compression moulding. The main properties of the raw materials are listed in **Table 2** which were provided by the suppliers of raw materials. **Table 3** presents the compositions of the G/CB/Fe/PP composite. Initially, the fillers such as graphite (G), carbon black (CB), and iron powder (Fe) were pre-

mixed using a ball mill machine for 1.5 hours to achieve a homogeneous mixture. Subsequently, the pre-mixed fillers and polypropylene (PP) powder were compounded in an internal mixer at 200 °C, with a rotor speed of 50 rpm for 15 minutes. This step ensured uniform dispersion of the fillers within the PP matrix. Following compounding, the mixture was pulverized using a pulveriser to obtain a finer powder, deemed essential to prevent agglomeration and ensure uniformity during plate fabrication. The composite plates were then fabricated using the hot compression moulding technique with a 200-Ton hot press machine. Prior to moulding, the press was set to a temperature of 185 °C, and the mould was preheated for approximately 10 minutes. The mixture was then compressed at a pressure of 50 tons for 10 minutes. Finally, the mould was allowed to cool to room temperature before demoulding the fabricated composite plates.

Table 2. Properties of filler materials

Property	G	CB	Fe	PP
Bulk Density (g/cm ³)	1.3-1.95	0.1 - 0.12	7.87	0.91 – 0.92
Melting Point (°C)	3600	3727	1535	169
Boiling Point (°C)	4200	4830	2750	220
Electrical resistivity (ohm-cm)	5x10 ⁻⁶ -30x10 ⁻⁶	0.01 – 0.1	10.1	10 ¹⁴
Average particle size (µm)	52.9	5.48	4.91	2x10 ³

Table 3. The Composition of G/CB/Fe/PP (Based on wt %)

Sample	G %	CB %	Fe %	PP %
1	50	25	5	20
2	45	25	10	20
3	40	25	15	20
4	35	25	20	20

Properties testing and analysis

Several tests were conducted to determine the properties of the bipolar plates. Electrical conductivity was measured at nine points on both the top and bottom surfaces of each specimen. Using the Jandel Multiheight Microposition Probe technology and a constant current of 1 mA applied during testing, measurements were averaged to determine the final value. The in-plane conductivity was calculated according to the method described in Equation 1. Sample density was evaluated with an electronic densimeter following the dry bulk density measurement method described in ASTM C559. Each test was repeated three times for specimens with dimensions of 1 cm × 1 cm. Meanwhile, Flexural strength was determined using an INSTRON Universal Testing Machine in accordance with ASTM D790-03. Specimens (13 cm × 1.3 cm × 0.3 cm) were fixed in a custom jig. Three repetitions were performed for each sample. The shore hardness was measured using an analog TECLOCK GS-702G durometer according to ASTM D2240D. Each sample was tested five times. Finally, the microstructure was examined using a JEOL JSM-840 scanning electron microscope (SEM) operating at 15 kV, with magnifications ranging from 500× to 3000×. A backscattered electron imaging mode was used to analyze surface morphology. Energy-dispersive X-ray spectroscopy (EDX) was employed to determine elemental composition. SEM images revealed fine topographic details not visible to the naked eye, while EDX provided compositional information.

$$S \text{ cm}^{-1} = \frac{1}{2\pi S(v)(0.6336)} \quad (1)$$

where, S = 0.1 cm (distance of JMHPFP point)
0.6336 = factor of thickness per diameter of specimen.
v is data from test.

Results and Discussion

Four samples of blank bipolar plates were fabricated using G/CB/Fe/PP composites with varying Fe weight percentages (wt%). A comprehensive series of tests was conducted to evaluate their performance, including in-plane electrical conductivity and mechanical properties such as flexural strength, Shore hardness, and bulk density. Additionally, microstructural analyses were performed using Scanning Electron Microscopy (SEM) and Energy-Dispersive X-ray Spectroscopy (EDX). The obtained results were systematically analysed and discussed to determine the optimal composition that offered the best balance of electrical, mechanical, and structural properties for application as bipolar plates.

Electrical conductivity

Figure 2 presents the in-plane electrical conductivity

of average value of 18 points of four CPC samples. According to the U.S. Department of Energy (DoE) standards, CPC bipolar plates must exhibit conductivity of at least 100 S/cm. All samples tested surpassed this threshold. The incorporation of Fe at concentrations ranging from 5 wt% to 15 wt% led to a significant improvement in electrical conductivity, increasing from 247.07 S/cm to a peak value of 367.59 S/cm for the sample containing 15 wt% Fe. This enhancement was attributed to the small particle size of Fe (4.91 μm), which promoted the formation of effective conductive pathways among the graphite (G), carbon black (CB), and Fe networks. The small Fe particles effectively filled the interstitial spaces between the larger graphite particles (52.9 μm), thereby enhancing the overall conductivity of the composite. This behaviour aligns with the percolation threshold theory, wherein the addition of conductive fillers increases the probability of forming continuous conductive networks. The introduction of Fe as a third conductive filler, alongside G and CB, significantly improved the composite's conductivity up to 15 wt% Fe content. However, when the Fe content increased to 20 wt%, a decline in conductivity was observed, dropping to 158.7 S/cm. This reduction was attributed to the disruption of the conductive network due to poor dispersion and agglomeration of Fe particles at higher concentrations. The excessive presence of Fe interfered with the effective interaction between G, CB, and Fe particles, leading to reduced contact points and an overall deterioration in the conductive network. Additionally, the agglomerated Fe particles negatively affected the bonding with the polymer matrix and other conductive fillers, as confirmed by the SEM microstructural observations shown in **Table 5**. Thus, while Fe acts as an effective conductive filler at optimal levels (5 wt% to 15 wt%), exceeding this range leads to reduced dispersion quality, agglomeration, and diminished electrical conductivity. These results are in line with prior study findings [23, 24, 25] and are corroborated by more recent studies that draw attention to the problems associated with high filler content in CPC composites [26, 27, 28, 29, 30].

Flexural strength

Based on the flexural strengths of the CPC bipolar plates shown in **Figure 3**, it can be observed that increasing the Fe content in the G/CB/Fe/PP composites from 5 wt% to 15 wt% led to an improvement in flexural strength, yield strength, and flexural modulus. However, when the Fe content was further increased to 20 wt%, a decline in flexural strength was noted. This reduction was attributed to poor surface interactions between the filler particles and the PP matrix, as previously reported [21, 23, 31]. The observed sudden increase in flexural strength at certain compositions was likely due to the non-

homogeneous mixing of the filler materials with the binder. Overall, the flexural strength decreases when the Fe content exceeds 15 wt% [16-18]. Despite these variations, the flexural strength of all CPC bipolar plates fabricated from the G/CB/Fe/PP composites satisfies the flexural strength requirement set by the US Department of Energy (DOE), which specifies a minimum flexural strength of 25 MPa.

Density test

The density of each sample was measured three times, and the average value was determined. As shown in **Figure 4** (Bulk Density), the results indicated that for CPC bipolar plates composed of G/CB/Fe/PP composites with CPC content ranging from 5 wt% to 25 wt%, the variation in density was minimal, showing no significant difference among the samples. Additionally, all CPC samples had measured densities below 5 g/cm³, which is the standard set by the U.S. Department of Energy (DOE) target (2017).

Shore Hardness test (Shore-D)

Hardness measurements were conducted at five different locations on the plate, with each location

tested more than three times to ensure accuracy. The average hardness for each point was calculated, and the measurement locations were documented. As shown in **Figure 4**, the plate fabricated with the G/CB/Fe/PP composition exhibited the highest hardness value of 56.13 Shore-D. All CPC samples tested surpassed the minimum hardness requirement established by the US Department of Energy (US-DOE), which specifies a Shore-D hardness of at least 40. However, the addition of Fe as a filler material resulted in a decrease in hardness compared to compositions without Fe. This indicated that the presence of Fe adversely affected the interfacial bonding between the filler materials and the polymer binder.

EDX and surface morphology analysis

Table 4 presents the SEM analysis at 9k magnification, illustrating the microstructure with distinct contrast: the bright areas represent iron (Fe), the dark areas correspond to carbon black (CB), and the grey areas indicate graphite (G) [13, 21]. EDX analysis further confirmed that the white regions were rich in Fe, as detailed in **Table 4**.

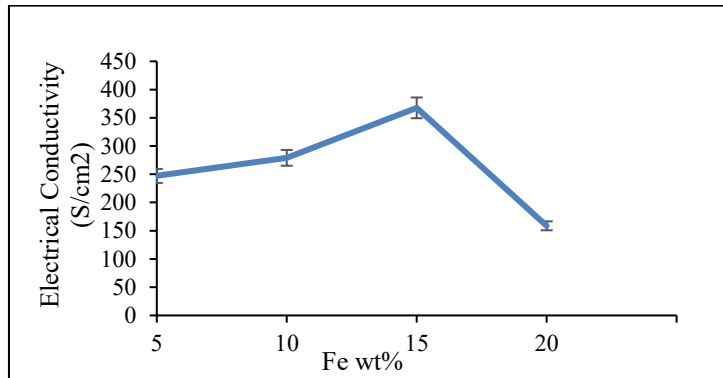


Figure 2. Graph of electrical conductivity

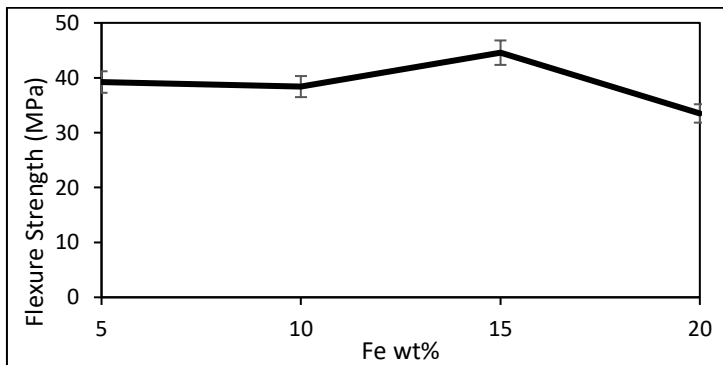


Figure 3. Graph of flexural strength

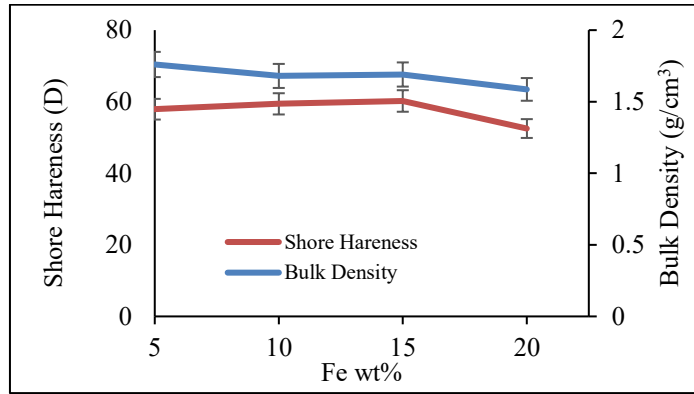
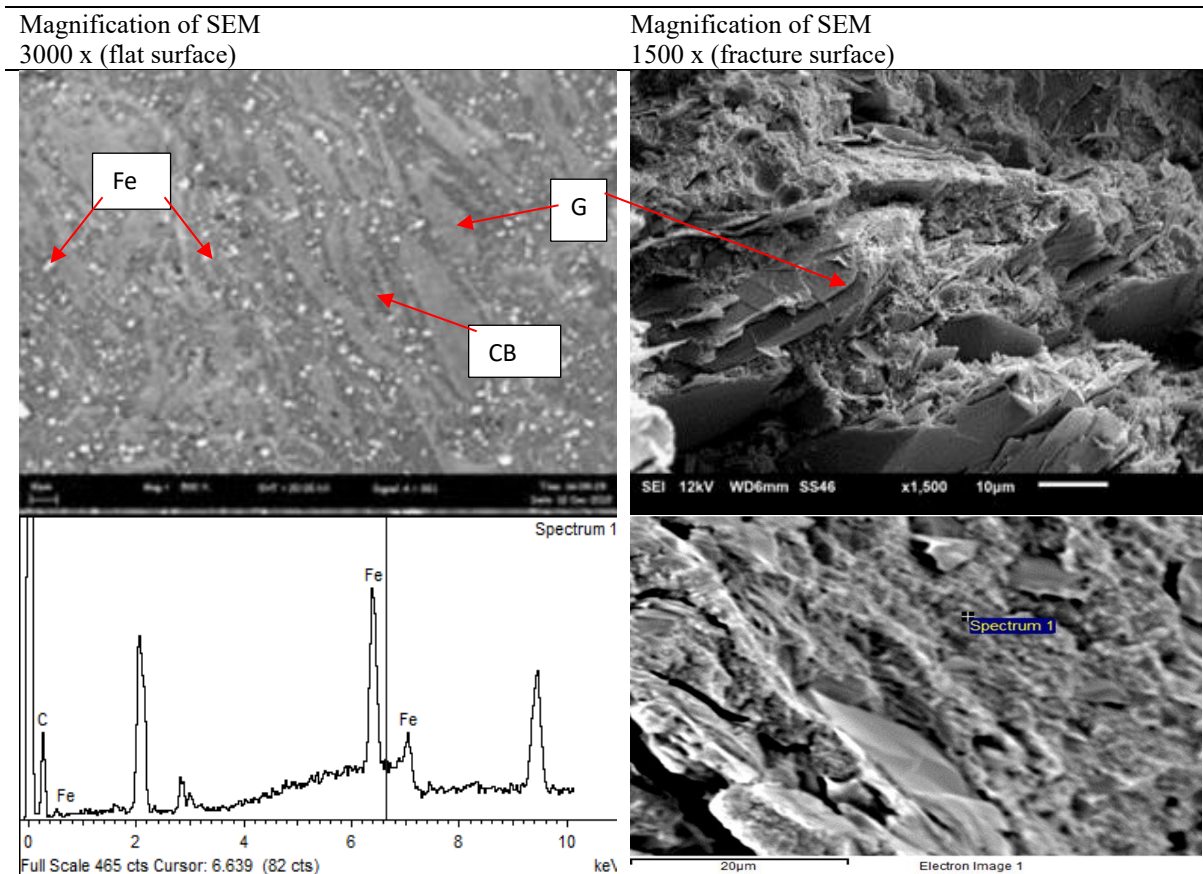


Figure 4. Graph of Shore Hareness and Bulk Density

Table 4. SEM and EDX analysis G/CB/Fe/PP (15 wt% Fe)

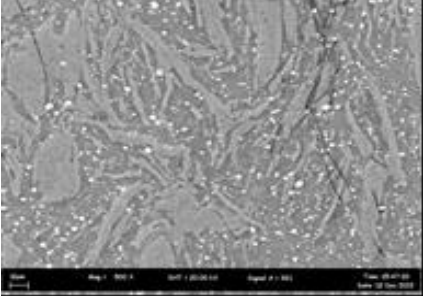
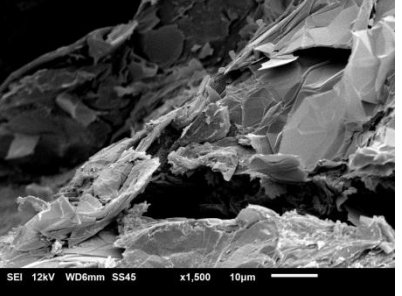
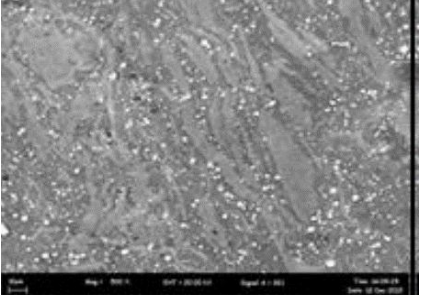
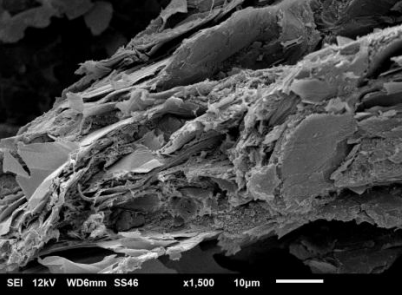
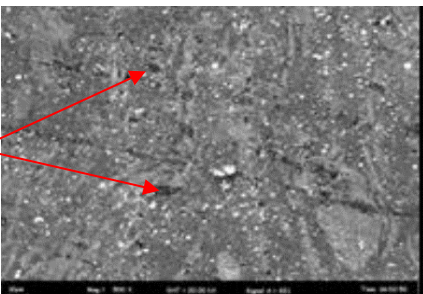
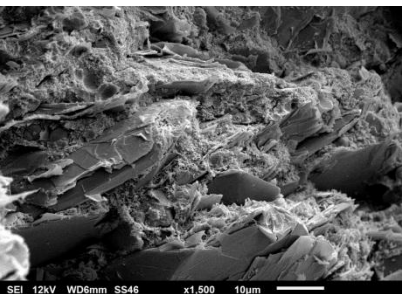
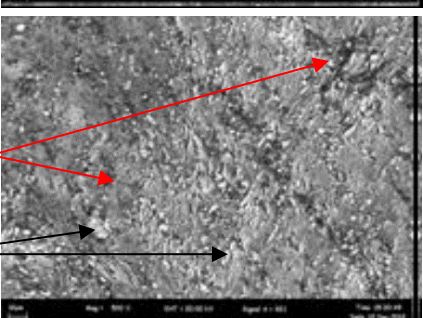
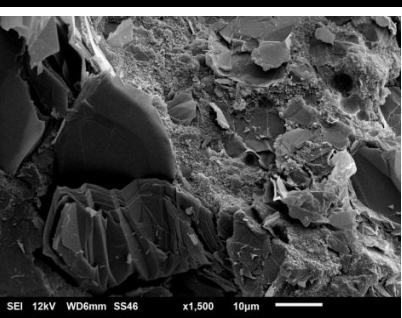


SEM photomicrographs of the composites with Fe content ranging from 5 wt% to 20 wt% revealed that samples containing 5 wt% to 15 wt% exhibited a more homogeneous and refined microstructure. However, at higher Fe contents (20 wt% to 25 wt%), the images displayed the emergence of voids and dimples associated with Fe distribution. Specifically, the fracture surface of the 5 wt% Fe sample demonstrated a brittle fracture mode with minimal plastic deformation and no evidence of necking. **Table 5**

highlights the increasing presence of voids and dimples as the Fe content rose, indicating a transition toward more brittle fracture behaviour. This brittleness adversely affected the mechanical properties, particularly the flexural strength, which declined with increased Fe content.

Furthermore, higher magnification SEM images of the fracture surfaces revealed a transgranular fracture mode, characterized by numerous voids of varying

Table 5. SEM for all composition G/CB/Fe/PP

Composition of G/CB/Fe/PP	Magnification of SEM 3000 x (flat surface)	Magnification of SEM 1500 x (fracture surface)
5 Fe wt%		
10 Fe wt%		
15 Fe wt%		
20 Fe wt%		

sizes and shapes interspersed with shallow dimples. **Table 5** provides a comprehensive illustration of the microstructures for all composite formulations at magnifications of 1500x and 5000x, showing the distribution of G, CB, and Fe within the blank bipolar plates.

In addition, Table 5 offers a clearer visualization of the filler and binder dispersion across the different G/CB/Fe/PP composite compositions. The SEM photomicrographs revealed that Fe particles tended to agglomerate at specific locations within the matrix. This agglomeration significantly impacted the final properties of the CPC (carbon-polymer composite) bipolar plates by contributing to micro or nano porosity, which in turn affected the mechanical and electrical performance of the composites. Moreover, the agglomeration may impair the wetting behaviour of the binder material, further degrading the composite's properties, including electrical conductivity, flexural strength, and hardness [27, 31, 32]. From an electrical perspective, the presence of

small Fe particles facilitates the formation of conductive pathways (tunnels) between G and CB particles, thereby enhancing the overall electrical conductivity of the composite, as depicted in **Figure 4**. However, the mechanical analysis of the fracture surfaces confirmed the predominance of brittle fracture, with minimal plastic deformation and the absence of necking. Increased Fe concentration caused voids and dimples to proliferate, as **Table 5** illustrates, which negatively impacted the composites' flexural strength and hardness, as illustrated in **Figures 3 and 4**.

Conclusion

Based on the results, Fe content has a significant influence on both the properties and fabrication process of the CPC bipolar plate. All samples of the G/CB/Fe/PP composite have successfully met the desired properties including in-plane electrical conductivity, flexural strength, bulk density, and Shore hardness within the targets set by the Design of Experiments (DoE). Among the samples tested, the composite containing 15 wt% Fe exhibit the most favourable overall performance, indicating that the incorporation of Fe into the G/CB/PP composite notably enhances its properties, resulting in a material that is lightweight, strong, cost-effective, easy to fabricate, possesses low contact surface resistance, and exhibits good mechanical stability. These attributes highlight the potential of the developed composite to serve as a promising material for future bipolar plate applications.

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