

Review

Influence the Fillers Orientation: A Short Review

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Abstract: Bipolar plates significantly contribute in the development of the polymer electrolyte membrane (PEM) fuel cells technology due to their ability to produce high electrical conductivity based on type of materials used. Mismatching of inappropriate materials and manufacture may lead to the inferior performance of PEM fuel cells. Hence, material development was determined crucial to balance the overall performance of PEM fuels including the mechanical properties and electrical conductivity of the materials. Studies on conductive polymer composites (CPCs) offered filler orientation as an alternative method to enhance the overall performance of bipolar plate. Filler orientations permit an excellent conductivity network formation while controlling the filler alignment based on required applications. This paper reviewed various studies of filler orientations including materials used and methods of manufacture of CPC materials for the effective development of bipolar plate.

Keywords: conductive polymer composite; orientation; electrical conductivity.

1. Introduction

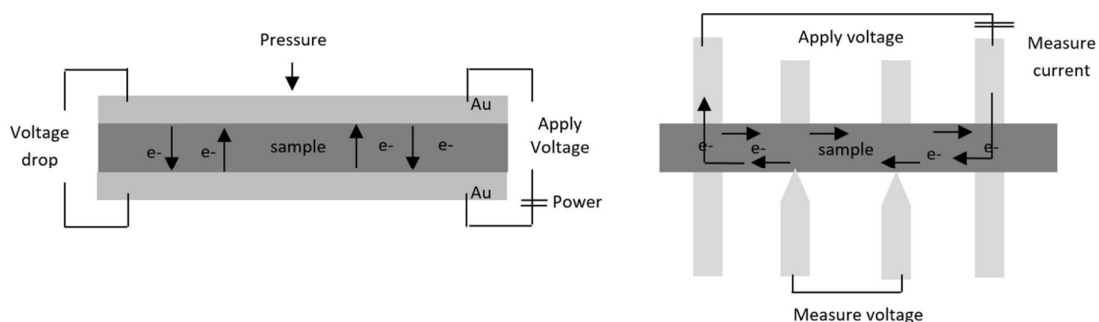
Polymer electrolyte membrane fuel cell (PEMFC) is a potential power source for various applications due to its low operating temperature, high efficiency, high power density and good impermeability [1,2]. A significant component of the PEMFC is the bipolar plates which account for 80 % of the total stack weight and 45 % of stack cost [3]. Generally, there are two types of bipolar plates used in PEMFCs including metal based bipolar plate and carbon based bipolar plate [4,5]. However, the major limitation of metal based bipolar plate is the lack of corrosion resistance which promotes degradation while carbon-based bipolar plate is lack in mechanical properties as the material become more brittle, less durable and more permeable to gases [3,4,6–8].

There are various materials available to develop the bipolar plate including traditional graphite, metallic alloy and graphite-polymer composite [3,6]. However, these materials have a major drawback which deteriorates the performance of bipolar plate as a result of imbalance between the electrical conductivity and mechanical properties [3]. Thus, current research trends tend to use conductive polymer composite (CPC) as an alternative material due to its ability to enhance electrical conductivity while maintaining the mechanical properties of the material [9–12]. CPC materials are obtained by mixing the insulating material such as thermoplastic or thermoset material with conductive fillers such as carbon black, carbon nano-tubes (CNTs) and carbon fibre [3,13]. The unique morphology and structure of the conductive pathways within the composite materials are the main parameters that determine the electrical properties of CPCs [14]. However, due to numerous choices of materials, there are still lots of improvement on the bipolar plate performance are required. Commercial bipolar plate is usually developed using graphite (G) materials which have the

disadvantage in brittleness and difficulty in machining [3,7,8,15,16]. As result, a thick bipolar plate is produced which in turn increases the weight of fuel cell stack [3,8].

High electrical conductivity is the key requirement for an excellent bipolar plate. The electrical conductivity is usually measured as a through-plane and in-plane electrical conductivity. However, the obtained electrical conductivity remains inadequate as the through-plane electrical conductivity is generally lower than in-plane electrical conductivity as shown in numerous studies [7,10,17]. For example, a study using CNTs/G reinforce epoxy (EP) showed that the electrical conductivities of the through-plane and in-plane conductivity were 180 S/cm and 65 S/cm, respectively [7]. The same trend was also reported using the CF/SG/EP composite materials where the through-plane conductivity (50.34 S/cm) was determined lower than in-plane conductivity (69.79 S/cm) [10]. These results explained the deficiency of the overall performance of bipolar plate as a result of low through-plane conductivity. Figure 1 illustrates the mechanism of electron flow in the bipolar plate which determined the through-plane and in-plane conductivities of the plate [7,17]. Yet, none of the studies has achieved these conditions due to some deficiency in conductivity network formations.

Several studies suggested that the electrical conductivity of composite materials often affected by the structural properties of the fillers used in the insulating resin matrix including filler aspect ratio, shape factor and fillers orientations [3,18,19]. Filler orientations seem to be an alternative way to improve the electrical conductivity of bipolar plate. For instance, studies based on the multiwalled carbon nano-tubes (MWNTs) revealed the occurrence of network formations at low MWNT content in the bipolar plate due to excellent nano-tubes dispersions [20]. Since 1995, Lux et al. have described the importance of filler distributions, orientation and processing technique in insulating matrix as the orientation of fillers along the basal plane is able to produce higher electrical conductivity in comparison to the perpendicular to basal plane [21,22]. The properties of graphite and CNTs fillers, which have the high aspect ratio exhibited the anisotropic electrical properties that guarantee the high performance in the parallel axial direction [22–26]. However, fillers not necessary have to be in highly anisotropic conditions as studies using the carbon black (CB) particles reported that the electron tunnelling was formed as the CB tends to aggregate [14]. Yet, the electrical conductivity of the composite is usually lower compared to the filler with anisotropic behavior. These results reported while using the CB particles that eventually decreases the anisotropic of the composite and the overall performance on the composite [22]. Moreover, the efficient and accurate predictions of the filler orientations also very useful to control and design the manufacturing process as inhomogeneity of fillers often leads to lower materials performance [27].



[Figure 1 Illustrations of (a) through-plane conductivity and (b) in-plane conductivity.]

2. Development in the processing of composite materials

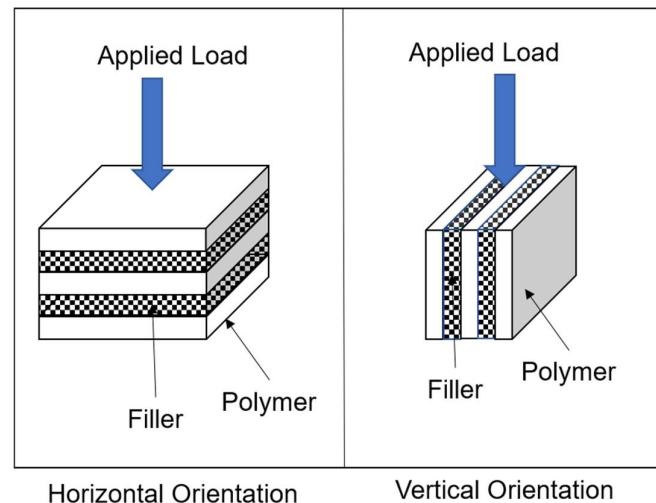
The manufacturing process of bipolar plate is the most important factors as it affects both mechanical properties and the electrical conductivity of composite materials. These factors related to the filler orientation, dispersion within the polymer matrix and conductivity network formations in composite materials [3]. In the current research trend, compression moulding or injection moulding technique is usually used to fabricate the bipolar plate. Injection moulding process offers high productivity that aid in massive productive rates and cost reductions. However, the viscosity of the composites increases due to increase in filler loading which harden the manufacturing process.

Therefore, compression molding seems to be an efficient alternative to produce high CPC materials [8,12]. Compression molding enables the fillers to retain within the polymer matrix while producing a laminate structure that maximizes the mechanical strength of the composite materials [28]. Studies using the GF/WPVC showed that compression molded sample had better mechanical properties compared to that produced by extrusion process due to low shear stress and low deformations experienced by the composite materials [29]. The high shear stress resulted in higher fibre breakage which lowers the mechanical strength of composite materials. The severe results obtained by Wang et al. using GF/PP composite materials undergoing injection molding process showed that the long fibre (LF) experienced more fibre breakage compared to short fibre. This phenomenon is difficult to compromise as it leads to deterioration in the mechanical properties of composite materials even though shear stress experienced by the fibre enhance the fibre orientations and tensile performance [30]. However, different studies using both long and short fibre of CF/PP composite reported that the longer CF was able to enhance the electrical conductivity of composites due to the earlier formation of conductive pathways [31]. Taipalus et al. explained in detail that the fibre with high aspect ratio was able to form end-to-end contact and led to an excellent conductive network formation. This phenomenon showed that the fibre loading is the key factor in developing excellent conductive pathways since at higher filler loading fibre tends to experience more filler breakage compared to low fibre loading [3,17].

Thus, in order to balance the overall performance of composite materials, current studies applied various manufacturing methods including method to disperse the fibre within the polymer matrix. For instance, Suherman et al. and Kakati et al. used several steps to ensure a homogenous mixture including ball milling technique [22,23]. However, studies reported in 1996 showed that the mechanical properties of composites were improved while using the extrusion process as the fibre tends to orientate its align to the desired orientation [32,33]. This indicated that extrusion is an alternative mixing method which can minimise the processing cost and time. In 2008, Barton et al. applied the extrusion process as the pre-mixing process before undergoing the injection molding process on G/CF reinforced liquid crystal polymer (LCP). This demonstrated that the extrusion process is able to obtain a homogenous mixture even with multiple fillers [34]. Moreover, studies using MWNT/PC showed that the mixing process via extrusion is able to produce a uniformly disperse mixture [20]. It is crucial to determine the suitable mixing and manufacturing processes to acquire maximum electrical conductivity while maintaining the mechanical properties of composite materials.

3. Fillers orientations effects on CPC materials performance

Previous studies reported that the filler orientations aid in the enhancement of mechanical properties and electrical conductivity at the preferential direction. Studies using GF/Wood-PVC composites showed that the align fibre tended to have a higher flexural modulus and flexural strength of 20% increment compared to the random fibre alignment due to the continuity of fibre to bearing of the applied loads. The experiment reported that the fibre which orientates at 0° experienced the highest tensile strength compared to fibre that orientates at 45° and 90° [29]. Meanwhile, studies using the cotton fibre reinforced geopolymer composites explained that the fibre in horizontal orientations exhibited a higher flexural strength, fracture toughness and compressive strength in contrast to the fibre in vertical orientations as shown in Figure 2 [35]. These are due to the fact that as in vertical orientations, the cotton fibre suffered from detachments and delamination which lowered the composites strength. Moreover, Alomayri et al. (2014) reported the existence of the crack-bridging in the vertical orientation which further lowered the composite strength [35]. Besides, the fibre orientation along the parallel directions showed excellent anisotropic mechanical properties rather than, in the perpendicular direction when using the graphite composites materials [36]. These indicated that the fibre orientations are crucial to determine optimum mechanical properties which can be developed by using the mixing process including an extrusion process.



[Figure 2 Schematic drawing of the cotton fibre orientation with respect to applied load.]

Kakati et al. reported that the electrical conductivity of the composite materials changed by the fibres orientation [3,22]. These were showed as at higher CF loading, within 5% to 9%, the electrical conductivity started to deteriorate due to agglomeration of the fibre in the NG-CB-CF-PF systems [22]. These were due to the fact that, CF has higher aspect ratio compared to other fillers in the system as a result of the fillers orientations in the matrix. The same pattern of electrical conductivity enhancement was reported using synthetic graphite (SG) fillers in liquid crystal polymer (LCP) compared to CF and CB fillers due to the fact that the fillers with high aspect ratio tended to orientate accordingly within the polymer matrix to provide a good electrical pathway [19]. The latest research reported that the fillers with high aspect ratio were used as the secondary fillers as their ability to fill the empty space of the primary fillers within the polymer matrix while developing an electrical network simultaneously [9]. These showed that the high aspect ratio fillers have the

capability to enhance the electrical conductivity of composite materials in many ways. Meanwhile, studies using the MWNTs in polycarbonate (PC) matrix reported that there was no evidence of fibres orientation as MWNT orientated randomly in both perpendicular and parallel to the extrusion directions [20]. Researchers reported that this phenomenon was due to the high aspect ratio and extreme geometry structure of the fibre. Besides, low shear rate and viscosity during the melting and mixing process also the main cause of the fibre to orientate randomly, as low shear stress was transferred onto the fibre. However, these randomly orientated MWNTs seem to be practical in certain applications, in order to enhance the percolation threshold of the composite materials up to 1.5 wt.%, as WMNTs were dispersed evenly in the polymer matrix [20]. These explained that the fibre orientation has the ability to control the mechanical properties and the electrical conductivity based on the applications needs.

4. Methods to induce fillers orientations

Since 1997, filler orientation in composite materials has been investigated to enhance materials performance. However, studies often focused on the mechanical properties rather than the overall performance of the composite materials. Thus, current trends showed that filler orientations are able to boost the electrical conductivity while maintaining the mechanical properties of the materials [37]. There were a few methods used to induce the fibre into certain orientations including shear rate, die geometries and filler aspect ratio [24,31,38]. Fibre aspect ratio often related to the filler distributions within the polymer matrix. At low filler loading, filler aspect ratio is the key of filler distributions affecting the conductivity network formations [39]. Moreover, high aspect ratio fillers tended to orientate more compared to low aspect ratio fillers [31].

It has been demonstrated that by introducing a breaker plate as the pre-orientation medium led to higher degrees of fibre orientation compared to traditional slit convergent dies [40]. In 1996, studies reported that the classical dies, later known as convergent dies produced a longitudinal orientation while divergent dies produced elongational deformations which allow the fibre to orientate to the desired orientations [32]. However, near the die walls, the shear deformation force of the fibre to align in longitudinal orientations. These explained the importance of die geometry in order to produce adequate fibre orientations. A study on glass fibre reinforced polypropylene indicated that the introduction of a pre-shearing using a slit die was able to orientate the glass fibre at 0.1 s^{-1} pre-shearing for 60 min [41]. However, different phenomenon were reported when using series sizes of the die gage on WMNT/vinyl ester composites. Fan et al. reported that there are similarities of WMNTs alignment in every micro channel, nano channel and cross section of the cylinder despite differing in gauge sizes due to non-Brownian motion effect to randomise the fibre orientations [24]. Tungjitpornkull et al. (2009) demonstrated that the compression molding was able to orientate fibre in 0° compared to extrusion technique using the twin-screw extruder. The studies reported that the fibre tended to align 0° while giving maximum mechanical properties [29].

Besides, studies using the PP/wood-fibre composites reported that the screw speed of 150 rpm produced higher mechanical properties and narrow size distributions compared to 100 rpm and 200 rpm. These are due to the better dispersion of the wood-fibre in the composite materials [42]. This strongly explained by the morphology structure of the composite PP/wood-fibre, where the best reaction occurred at 150 rpm. Studies using polyethylene glycol as a lubricant able to induce the short fibre into a correct fibre orientation throughout all the die thicknesses influenced by the shear force

[20,24,32,41]. Moreover, Fan et al. reported that the orientations of MWNT remain unchanged after applying the shear force for 24 hours suggesting that the shear flow were permanently generated [24]. The shear stress not only does develop better orientations but also affect the fibre microstructure. Studies using the long glass fibre reinforced polypropylene (LFPP) reported that LFPP experienced fibre breakage which deteriorated the mechanical properties including the tensile properties of composite materials [30]. Besides, an increase in the generated shear force increased the melt viscosity, which in turn increased the fibre addition and resulted in fibres orientation [43,44]. However, a composite mixture with high viscosity may lead to difficulties in processing and formation of voids in the final composite materials [7]. Huang et al. proposed a study on different orientation angles which indicated that at an angle of 0° has the highest shear stress and normal stress compared to 30°, 60° and 90° of fibre orientations [45]. This demonstrated that the fibre orientation strongly related to shear stress and viscosity of the composite materials.

5. Measuring the filler orientations

Measuring the filler orientation is very critical as studies showed that the angle of filler orientation was used in predicting the electrical conductivity of composite materials [31,37]. There are various methods available in measuring the filler orientations in composite materials including direct methods and indirect methods. Direct methods such as SEM, FESEM, AFM, X-CT and more are usually used to visualise the fibre orientations especially for micron size fibres as these are the easiest methods to determine the morphology changes in composite materials [24,30]. For instance, Potschke et al. used the direct methods of SEM, TEM and AFM to observe the fibre alignment in cross section area, perpendicular and parallel to extrusion direction [20]. In additions, the dispersion of fibre in the nano-fibre materials are usually observed using TEM and FESEM. In this observation, the sample is sliced into thin film [46–48]. On the other hand, Dweri et al. used the digital image analysis (MatLab) in cooperating with the direct method of VPSEM to measure the electrical conductivity of the composite materials [49,50]. This indicates that the variety of direct methods can be used to observe the filler orientations in the composite materials. Details of fibre orientation measuring methods are summarized in Table 1.

[Table 1 Fibre orientation measuring methods.]

Matrix	Filler	Composite Processing method	Orientation measurement	Reference
Polybutadiene (PB)	MWNT	Rheology	Shear light-scattering/microscopy (optical measurement)	[51]
Polypropylene	Graphite Carbon black	Compression molding	VPSEM	[49,52]
Polypropylene	Short fibre	Extrusion	SEM (photograph)	[32]
Phenol formaldehyde	Graphite	Compression molding	SEM (data image processing)	[23]
Polycarbonate (PC)	MWNTs	Extrusions	SEM AFM TEM	[20]

Vinyl ester	Oxidized MWNT	Micro and nano-sized channels	TEM Second order tensor	[24]
Polyetherimide (PEI)	Carbon fibre	Plain weave (adhesive wear study) Compression molding	SEM	[28]
Sheet molding compounds (SMC)	Glass fibre	Pyrolysis	Transmission of visible light X-ray imaging	[46]
Polypropylene	Glass fibre (short fibre and long fibre)	Compression molding	X-ray computed tomography (X-CT)	[41]
Poly-ester Poly-epoxy	Short carbon fibre	Compression molding	SEM	[13]
Polyacrylamide	Short glass fibre	-	SEM (2D-image analysis) Fibre orientation distribution (FOD)	[53]
Isophthalic polyester	Glass fibre	Twisted fibre yarns (stitching)	SEM	[54]
Polymers	Short fibre	-	Fibre length distribution (FLD) Fibre orientation distribution (FOD)	[55]
Isotactic polypropylene	Glass fibre	Extrusions Injection molding	Optical microscope (OM) observation SEM Micro-FTIR	[30]
Wood/poly(vinyl chloride)	E-glass fibre	Extrusions Compression molding	SEM	[29]
Rubber	MWNTs	Mechanical mixing Extrusions	FESEM WAXD	[48]
Thermoplastic	Short fibre	Injection molding	Second order orientation tensor	[27]
Polypropylene	Glass fibre	Extrusions	Transputer controlled image analysis	[40]
Epoxy	Milled carbon fibre	Mechanical mixing Compression molding	SEM	[10]

Nylon 6,6 Polycarbonate	Carbon fiber	Extrusions Injection molding	Electrical conductivity model	[18]
Vectra liquid crystal polymer (LCP)	Carbon fibre	Extrusions Injection molding	Optical microscope (OM)	[56]
Geopolymer matrix	Cotton fabric	Layer	SEM	[35]
Polypropylene	Single wall carbon nanotube (SWCNT)	Tensile stress- strain	Finite element method (PATRAN)	[45]

Meanwhile, indirect method seems to be an effective method to quantify the amount of fibre alignment produced and to determine the fibre misalignment in the composite materials [38]. X-ray diffraction (XRD) is used extensively for this purpose where the graphitic plane azimuthal diffraction is measured in degrees and used to characterize the fibre orientation. For instance, the fibre alignment was reported as a vapour grew carbon in the preferred direction (0°). It is critical to note that the intensity of the diffraction pattern at high angles is generally very low while the intensity at 0° is generally very high as shown in a few studies [38,57]. In addition, several studies investigated the fibre orientation behaviour in composite materials using the wide-angle X-ray diffraction (WAXD) [48,56]. For example, Kim et al. determined that the CNTs were aligned in the x-direction as expected when measured using this method. Moreover, WAXD is able to determine the degree of orientation at different CNT loading. For instance, at 5 wt%. the degree of orientation is lower compared to 30 wt%. [48]. This behaviour is due to the fact that at higher filler loading a better network of conductivity was formed resulting from better CNT orientation within the polymer matrix. This demonstrated that the selection of technique for measuring the fibre orientation is very crucial for effective investigation of the composite materials.

6. Conclusions

The development of conductive polymer composite (CPC) materials is very important due to the fact that these materials directly influenced the performance of PEM fuel cell. Fibre orientations are the key factor to enhance both the mechanical properties and electrical conductivity of CPC materials. Extrusion process is the best method to induce the fibre into the desired orientations. Based on the discussion above, the extrusion process is able to induce orientations as it possesses an adequate shear rate and controllable die geometries. Meanwhile, the scanning electron microscope is a common method in determining the fibre orientation compared to indirect methods.

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