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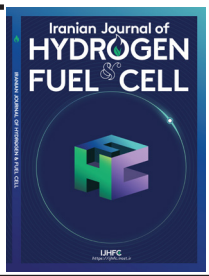

Experimental Analysis of Fiber Laminating Effects on Flexural Rigidity of Composite Endplates for Use in PEM Fuel Cells

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Abstract

One of the most important components of a polymer electrolyte membrane fuel cell is the endplate, which must exert uniform contact pressure distribution on the membrane electrode assembly. Since the endplates must be highly rigid, it is essential to consider the flexural modulus parameter when designing these plates. In this study, the production of lighter-weight endplates with a higher flexural modulus is significantly improved by replacing heavy metallic plates with polymer composite plates. The vacuum bag manufacturing technique was used to create these composite plates from epoxy resin, carbon fibers, and glass fibers, making them compatible with the environment of the fuel cell. The flexural modulus and heat deflection temperature of each sample were evaluated before and after a simulated environment test of the fuel cell. Then, the amount of water absorption for each specimen was calculated. Finally, the composite endplates were fabricated using the two different laminations of fibers to find the optimum fiber lamination to maximize the endplate flexural rigidity. The optimum sample contained carbon fibers with an epoxy resin with 0 degrees arrangement. This specimen has a flexural modulus of about 93.17 GPa, heat deflection temperature of about 261 °C, and water absorption of about 0.86%, which are ideal for fuel cell endplates.

1. Introduction

A large polymer electrolyte membrane fuel cell (PEMFC) stack may consist of hundreds of structural components compressed together by two endplates with a reasonably large clamping force exerted by the tightening screw bolts [1]. In order to support the tightening pressure, stack gravity, vibration, impacting force, and other erratic loads experienced by the stack, the endplates must be sufficiently strong and rigid [2].

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The PEMFC consists of gas diffusion layers (GDLs), membrane electrode assemblies (MEAs), bipolar plates, and gaskets sealed between two endplates. In order to lower the electrical resistance of the cell and prevent fuel leakage, the endplates must exert a significant and consistent amount of pressure on the fuel cell components. In addition, it is essential to provide suitable thermal insulation for low heat loss and appropriate cold start behavior. Therefore, the optimal endplate design must improve fuel cells' power density and reliability [3].

There have been many studies conducted to improve the endplate fuel cell design. For example, Barzegari et al. [4] designed a new PEM fuel cell clamping mechanism to study the contact pressure distribution over the active area of the PEM fuel cell's MEA. The innovative clamping mechanism was compared to the conventional clamping mechanism using simulation, and an experimental investigation supported the numerical results. Cheng et al. [5] suggested that plates with controllable hardness made of polydimethylsiloxane provide were suitable for use as endplates. Barzegari et al. [6] optimized the geometric parameters of the pneumatic clamping system to exert uniform contact pressure distribution on GDLs. During this simulation, they concluded that the weight and efficiency of systems with optimal pneumatic endplates are much better than systems with conventional endplates.

Suvi et al. [7]and consequently has an impact on the mass transfer in the GDL. Thus, the compression pressure distribution on the GDL can have a significant effect on the performance and lifetime of a PEMFC stack. Typically, fuel cell stacks are assembled between two end plates, which function as the supporting structure for the unit cells. The rigidity of the stack end plates is crucial to the pressure distribution. In this work, the compression on the GDL with different end plate structures was studied with finite element modeling. The modeling results show that more uniform pressure distributions can be reached if ribbed-plate structures are used instead of the traditional flat plates. Two different materials, steel and aluminum, were compared as end plate materials. The modeling results were verified with pressure-sensitive film experiments. Recently, using a combination of materials to achieve desirable properties has increased. This class of materials is called composite materials [8]. Composites can be defined as a physical mixture, on a macroscopic scale, of two or more different materials. According to some criteria, the mixture has better mechanical properties than its individual components. Generally, the primary strategy in designing and constructing applicable instruments is to lighten the structure; however, it is impossible to find a material with all the desired properties [9]. One of the most important advantages of composite materials is that their properties can be controlled according to the requirement [10].

Other studies have been done to optimize the buckling load of composite plates. For example, Woodsenbet et al. [11] investigated some critical points using a simulator model to solve the buckling problems of composite cylinders. Yu et al. [12]two thick steel endplates have been used to maintain a proper contact pressure at the interfaces among gaskets, gas diffusion layer (GDL presented a concept of sandwich-structured composite endplates with pre-designed endplate curvature to improve and homogenize the internal pressure. Following this idea, they designed a sandwich endplate using a pressure distributor for polymer fuel cells to enhance thermal insulation and uniform contact pressure distribution. Fan et al. [13] conducted another research on sandwich plates with a lattice core. In this research, the mechanical properties of these structures were investigated by designing and manufacturing sandwich plates made of two multilayer plates and a mesh core and performing internal pressure, external plane pressure, and three-point bending tests on the manufactured plates. Dai et al. [14]gas diffusion layers (GDLs built an insulating foam–core composite sandwich structure and a pre-curved compliant pressure distributor to design endplates that provide both good insu-

lation performance and uniform pressure distribution on the PEMFC stack. Dai et al. [15]MEA (membrane electrode assemblies also presented a new asymmetric composite sandwich endplate design made of carbon fiber reinforced composite and glass fiber reinforced composite with a pre-curvature generated by residual thermal deformation, which yielded the required pressure distribution in the stack when the endplates were fastened by the clamping device.

Building on the above research, this study aims to design more efficient polymer composite endplates. The primary purpose of this study is to replace metallic endplates with polymer composites in order to reach a much lower weight and better specific mechanical properties. Next, the characterization of crucial parameters for the fabrication of polymer composite PEM fuel cell endplates was studied. Finally, samples were fabricated, and the appropriate lamination of fibers was evaluated according to mechanical tests.

2. Experiments

2.1. Materials and the simulated fuel cell environment

The following items were bought from Aladdin Chemistry Co., Ltd. in China: unidirectional glass fibers 280 g, unidirectional carbon fibers 200 g, and laminating resin epoxy (Tg = 80 °C with 15% hardener). Four composite plates were fabricated using the vacuum bag technique (Fig. 1(a)), calculating and weighing each component before curing. The optimum ratio of resin and fibers (60% fibers and 40% resin) was determined according to Ref. [16]. Additionally, samples with carbon fibers were cured under loading according to the vacuum bag method (Fig. 1(b)). These plates were produced using four separate recipes, each with a unique combination of unidirectional fibers, (0) degrees in one direction and (0,90) degrees in 2 directions. The specimens were labeled CE (carbon fibers and epoxy resin) and GE (glass fibers and epoxy resin), and the specific construction angles for each were given.

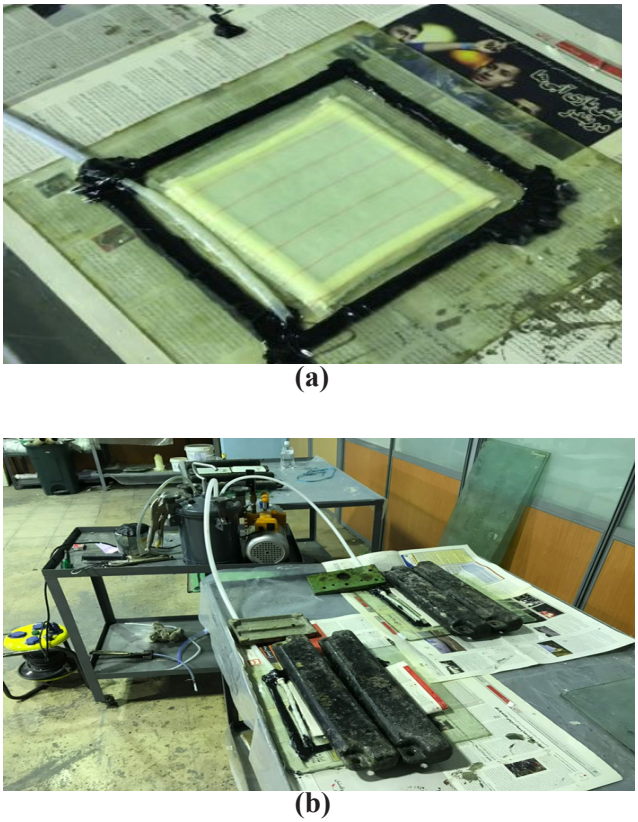


Fig. 1. Vacuum bag method for curing composite panels reinforced with (a) glass fibers in two arrangements and (b) carbon fibers in two arrangements and under loading.

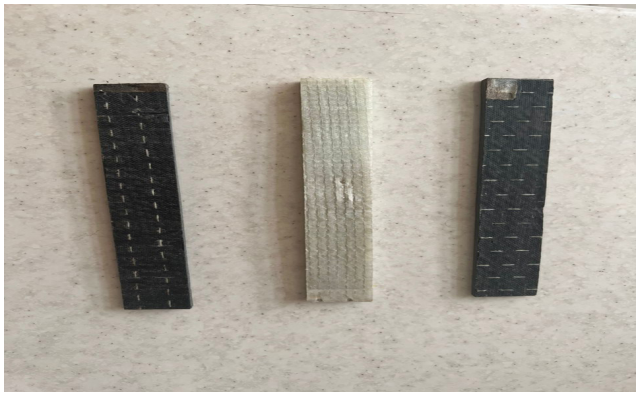
This study used a solution similar to the real PEM fuel cell environment to simulate the fuel cell environment. It is composed of 48% HF and 98% H_2SO_4 dissolved in balance reagent grade water. The final composition of the solution was 12 ppm H_2SO_4, 1.8 ppm HF with reagent grade water having 18 Mega Ohm resistances as the exposure medium. The pH value for the solution was measured as 3.35 at room temperature (25 °C) using a digital pH meter. Test temperatures were selected at 80 °C and 60 °C, which are close to the operating temperatures of actual PEM fuel cells.

2.2. Flexural test

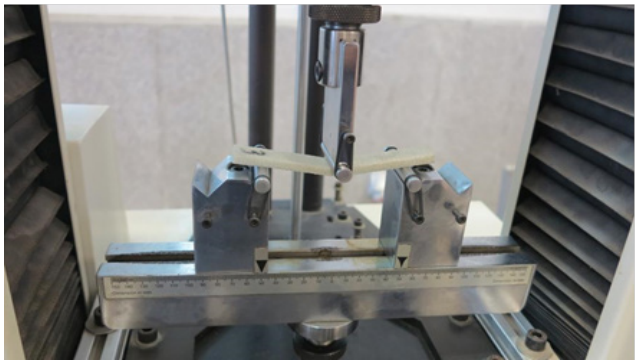
The flexural test measures the force required to bend a

beam under three-point loading situations. The data is often used to select elements for parts that will support loads without inflection. The flexural modulus is used as an indication of a material's stiffness when inflected. Since the properties of many elements (especially thermoplastics) can vary depending on ambient temperature, it is appropriate to test materials at temperatures that simulate the real environment.

The samples required for the three-point bending test were cut from the primary plates of GE and CE with the standard dimensions of the test, demonstrated in Fig. 2(a). Then, bending tests were conducted using samples with dimensions of 120×20×4 mm in accordance with AStM D790 at a speed of 3 mm/min and an 80 mm span length (Fig. 2(b)). To guarantee the results' reproducibility, at least five samples were used in each test.



(a)



(b)

Fig. 2. (a) Standard specimens prepared from GE and CE plates for the three-point bending test. (b) Three-point bending test of a sample.

A three-point flexural test is a mechanical test that calculates the values of flexural elasticity modulus E_f, flexural stress σ_f, flexural strain ε_f, and the flexural stress-strain response of the material.

The following equation is used to determine the flexural modulus of composites and plastics:

$$E = \frac{F \times L_0^3}{4 \times C \times D^3 \times \gamma} \quad (1)$$

Where F, L_0, C, D, and γ stand for the peak deflection, length of the sample, sample width, sample thickness, and the deflection of the sample, respectively.

2.3. Water absorption test

The resistance of composites to water absorption was determined by placing pre-weighed samples in boiling water for different times, removing the sample, wiping it with dry paper, and immediately weighing the sample with an accuracy of 0.001. For the water absorption test, AStMD570, the samples were weighed dry and then completely immersed in water. Then after 1 hour, the samples were gently dried with paper towels and re-immersed in water for a subsequent test period. After, they were removed from the water and weighed again. The test was stopped when the samples reached equilibrium, that is until they absorbed enough water that their mass did not increase any further. This test was conducted for 1100 hours. The absorbed water content (% TA) of the samples was calculated from the ratio of dry sample mass (Ms) and sample weight after immersion (Mi) at different test times. Four samples were tested, and the average was used to show the results.

$$TA(\%) = \left(\frac{M_i - M_s}{M_s} \right) \times 100 \quad (2)$$

2.4. Aging test in the simulated fuel cell environment

First, each sample was inserted into the prepared solution to prove that the samples operate well in the PEM fuel cell environment. The canisters were then heated to 80 °C in a circulating oven for 400 hours. Each specimen underwent another three-point bending test 400 hours later. The pH value in the real fuel cell conditions is 3.5-5, so to bring the test results closer to the real conditions, we tried making a stronger acidic environment and reducing the test time.

The test was performed at two temperatures, at room temperature (25 °C) and in an oven (80 °C). The 80°C oven temperature was chosen to approximate the operating temperature of a fuel cell and to investigate the effect of temperature on the reaction rate between two materials (composite and acid). First, the mechanical properties of composites mentioned in the experiment section were measured before starting the test and recorded as the initial value of the composites. Then, at regular intervals, the properties of the composites were measured, and their changes were calculated and compared to the initial value.

The purpose of conducting an aging test was to check each selected material's durability in the desired fuel cell environment. The test lasted for 400 hours at a temperature of 80 degrees when the samples were placed in the oven.

2.5. Heat deflection temperature test

Heat deflection temperature (HDT) was measured using an Instron HDT/Vicat tester in accordance with AStM D648. Studies were carried out following ISO 75 using samples that measured 130 x 13 x 60 mm and a force of 1.8 MPa. Utilizing the HDT technique, the composites' deflection up to 1mm was measured. The HDT was measured using at least two specimens at a heating rate of 2 °C/min and a span width of 64 mm.

3. Results and discussion

A three-point bending test was taken from 4 composites, and the flexural modulus was calculated for each specimen before and after the aging test (Table 1). The composites were considered more effective due to their lower density and weight compared to metals with similar mechanical properties. Moreover, samples with 0-degree lamination showed better flexural properties due to the alignment of all fibers in one direction.

Table 1. Properties of each sample after the three-point bending test.

Sample	Force at peak (N)	Deflection at peak (mm)	Flexural Modulus (Gpa)	Flexural Modulus After Aging (GPa)
GE (0)	2883.6	8.58	22.02	18.38
GE (0,90)	660.2	3.79	13.56	11.86
CE (0)	2032.1	4.61	93.17	76.79
CE (0,90)	1306.7	6.51	33.86	30.70
SS316	18763	23.58	33.66	31.8

In addition, the composite plates have good resistance against acidic and environmental conditions according to the selected matrix. For this reason, the epoxy matrix is resistant to acidic conditions, and there were minimal changes in the final flexural modulus after placing the samples in the acidic conditions of the fuel cell.

Nevertheless, the effect of weak acidic conditions on composite plates also needs to be investigated in this design. Therefore, we examined the modulus calculated before applying the environmental conditions of the fuel cell. Results showed that modulus values and mechanical properties resulting from the formulations used in this design were higher than metals and previous patents.

The flexural modulus obtained from the epoxy resin and glass fiber samples in two laminations of a (0,90) degree and (0) degree porcelain layer were calculated as 13.6 GPa and 22 GPa, respectively. Also, samples made of carbon fibers and epoxy resin had the best performance in terms of flexural properties. The reported modulus of these plates using the flexural modulus equation was reported as 33.8 and 93.1 GPa for (0,90) and (0) lamination, respectively.

Bending test diagrams before the fuel cell environment simulation test for the CE sample and GE sample are shown in Fig. 3 and Fig. 4. After placing the samples in the environmental conditions of the fuel cell for 400 hours, each of the samples was completely dried in the oven. Then, a three-point bending test was taken to observe the changes in modulus and flexural properties after 400 hours, and its results were discussed.

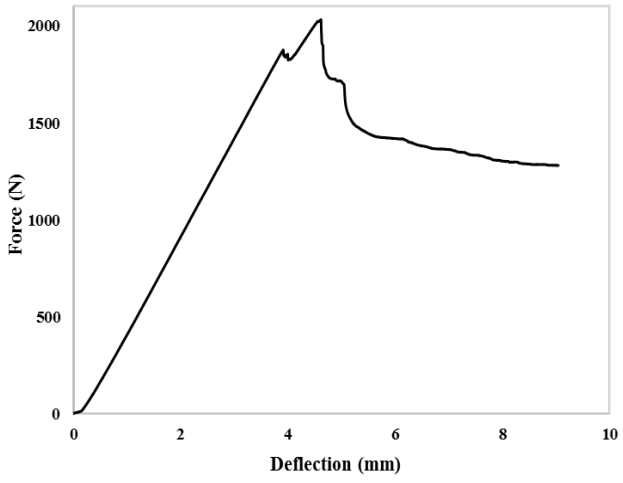


Fig. 3. Bending curve for CE sample with lamination (0).

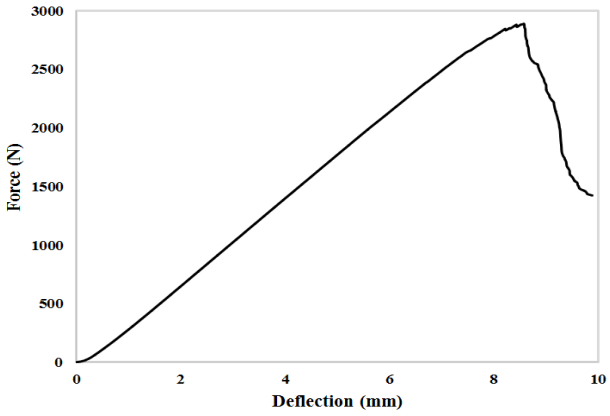


Fig. 4. Bending curve for GE sample with lamination (0).

The highest amount of flexural strength against the least displacement was found in the CE (0) sample, Fig. 5, resulting in the maximum flexural modulus in the manufactured samples. Figs. 5 and 6 show the three-point bending test diagram for the two manufactured samples after 400 hours of aging in the fuel cell simulation environment. After 400 hours, a drop of about 10-15% was observed in the flexural modulus of the samples, but no significant changes were observed in the weight before and after being placed in the acid solution. Also, the dimensional appearance and overall shape of the samples remained completely unchanged. The main cause of modulus drop is the penetration of acidic solution between the layers of fibers, which causes holes and degradation in the samples.

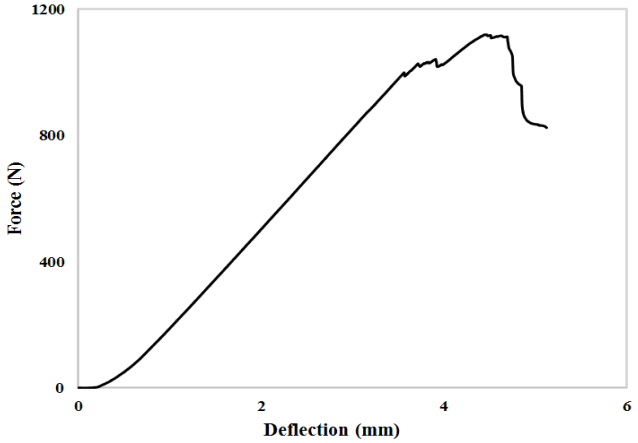


Fig. 5. Bending curve for CE (0) after aging.

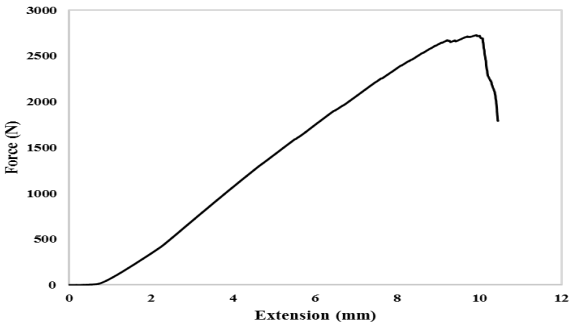


Fig. 6. Bending curve for GE (0) after aging.

Also, some of the resin was damaged in the acidic environment. The diffusion of acidic solution in the samples with 0 ° formulation was slightly higher than in the samples with 0 and 90 ° formulations due to the more space between fiber layers. Because carbon fibers are complete resistance to acid, it is concluded that the samples' loss of flexural properties after aging is due to the degradation of the resin or the reaction of the hydrophilic groups of the glass fibers with the acidic environment. As seen in the samples' bending test diagram, the flexural strength of the samples has also decreased for the same reasons mentioned for the flexural modulus decrease [17].

In summary, after 400 hours of aging, the flexural modulus and flexural strength decreased, and the extension of samples increased.

Because polymer composites are water absorbent and soluble, they have little water absorption. As a result, it takes a long time for the samples to dry completely [18]. So it is important to note that the water absorption in the composite samples is limited; if they remain longer in the solution, they will not be further damaged or decrease the flexural properties. The purpose of this test was to achieve the final water absorption value and measure the properties of the composite in that range and working conditions.

Each sample contains the minimum water absorption (under 1%) attained following a single absorption stage. It is observed that CE (0) provides far superior water absorption protection, about 0.86%, then the other specimens. Each sample was weighed before and after the water absorption test, see Fig. 7.

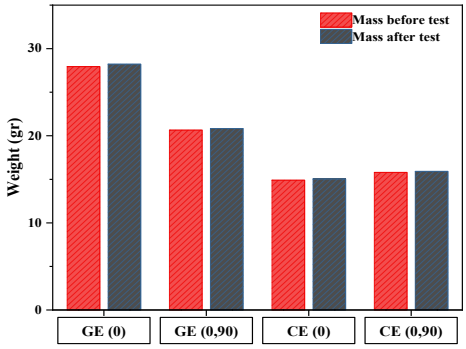


Fig. 7. The weight of each sample before and after the water absorption test.

The amount of water absorption for the GE (0,90) sample is the highest, with the percentage of gained water being 1.05. Also, a lower water absorption rate was observed in the samples with a layering angle (0) because of less space between the fibers and holes [19]. With a lengthy immersion duration, the sample mass was seen to grow steadily. A statistical study demonstrates the significance of the specimen weight change. It appears that the short-term water intake used in this study gradually increased with more immersion days.

The exact numerical values and percentage of water absorption are reported in Table 2. Long-term submersion in water has the potential to saturate the sample. The hydrophilic groups in the glass fiber would attract water molecules when the composites were submerged in water.

Table 2. The results of the water absorption test for the samples.

Sample	Initial weight (gr)	Weight after test (gr)	Water absorption (%)
GE (0)	27.94	28.235	0.9
GE (0,90)	20.653	20.836	1.05
CE (0)	14.907	15.042	0.86
CE (0,90)	15.795	15.916	0.96

After a longer period of submersion, the hydrophilic groups of the glass fiber would undergo chemical interactions with the water molecules, potentially producing soluble compounds. As a result, the weight would continuously decrease until the mass of the composites fell below its original mass. In this instance, the specimen's change in weight results from two effects: extracting materials and absorbing water. On the other hand, capillarity would draw water molecules into the material, and the holes and fractures in composite materials are perfect locations for the water to settle [20].

The HDT test can be defined as the temperature at which a particular test bar deflection is produced under a continuous normal load (the test bar rests on two supports of a given span) [21]. According to this definition, HDT is a straightforward assessment of the top limit of a plastic material's dimensional stability under a normal load and heat effect [22]. The HDT test was conducted to ensure that the composite plates would perform as anticipated at a temperature of roughly above 80 °C (fuel cell operating temperature), when the resin degrades based on its data sheet. The characteristics of the samples were sorted in decreasing order, much like the flexural modulus. In Fig. 8, the HDT for each sample, both before and after aging, is displayed.

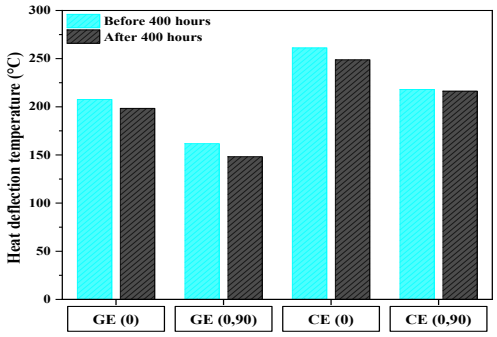


Fig. 8. The results of the HDT test for each sample before and after 400 hours of aging.

The maximal heat deflection temperature for the CE (0) sample was around 261 °C, which decreased only slightly to 248 °C after 400 hours of aging. This could be brought on by the acidic atmosphere causing the epoxy resin to degrade. Additionally, temperatures of 207.6 °C and 198.4 °C were recorded for the sample GE (0) before and after it was put in the acidic environment of the fuel cell, respectively. These findings show that the prepared end plates' mechanical characteristics will not be impacted by the working temperature of the cell, which is between 80 and 100 °C. Composites have a high heat deflection temperature due to the integration of fibers with excellent thermal characteristics.

4. Conclusion

This study aimed to make polymer composite plates with a spedific flexural modulus higher than metals. As a result, this research produced much lighter endplates with lower manufacturing costs and an easier machining process. Moreover, the lower density and weight of these plates greatly increase the efficiency of the fuel cell. In addition, the effects of the fibers' lamination were investigated to obtain better mechanical properties. According to the results, the optimum sample was CE (0) with a flexural modulus of about 93 GPa, heat deflection temperature of about 261 °C, and water absorption of about 0.86%, relatively higher than steel endplates.

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