Effect of curing time on the mechanical properties of composite coconut shell charcoal nanocarbon reinforced epoxy

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Abstract. Developments in composite materials have resulted in many types of compounds that utilize natural materials. One of the natural ingredients used is coconut shell. The result of processing coconut shells which are processed into charcoal can be used as reinforcement in composites. Research on composites with epoxy matrix reinforced with nanocarbon from coconut shell charcoal was given curing and non-curing treatment. Variation of curing holding time is 0.5 hours; 1.5 hours; 2.5 hours; and 3.5 hours at 80oC. Formation of nanosized carbon using a high-energy milling process with a shaker mill machine. The composite is made from a mixture of epoxy resin, hardener, and 400 ppm by weight of coconut shell charcoal nanocarbon. The ratio between resin and hardener is 2:1. The composite that has been formed is then given curing variations and its mechanical properties are observed through tensile and impact testing. The epoxy matrix composite without filler has the highest tensile strength of around 57.9 MPa at a curing time of 2.5 hours and the highest impact toughness is around 5.82 kJ/m2 at a curing time of 1.5 hours. Meanwhile, the nanocarbon-reinforced epoxy matrix composite has the highest tensile strength, which is around 59.3 MPa at a curing time of 2.5 hours and the highest impact toughness is around 9.31 kJ/m^2 at a curing time of 1.5 hours. The addition of nanocarbon particles can increase the highest tensile strength by about 23% at a curing time of 2.5 hours. However, the effect of nanocarbon on impact toughness has the highest increase of about 84.5% at a curing time of 1.5 hours. Nanoparticles and the corresponding curing time greatly affect the value of the impact toughness more significantly than the resulting tensile strength.

1. Introduction

The need for new alternative materials encourages innovation in high-performance materials, especially composites. Composites are a combination of two or more materials with different properties [1], [2]. Composite materials are composed of two phases; one is termed matrix, which is continuous and surrounds the other, often called the dispersed phase [1], [2]. Composite

materials have considerable advantages compared to other materials. It can be made to combine the superiority of the materials while minimizing their weakness. There are many types of composite materials, one of them is polymer nanocomposites.

Polymer nanocomposites use polymer as a matrix and nanoparticles as a reinforcement. On eof the common polymer that is used is the epoxy resin. These composites are generally reinforced using three general nanoparticle types: nanocarbons, nanoclays, and particulate nanocrystals. Among those materials, nanocarbon is the most widely used reinforcement material because of its cost and accessibility. Moreover, nanocarbon can be synthesized using readily available natural materials, which are safer, biodegradable, and more abundant [1–5]. One of the natural materials that can be used as composite reinforcement is coconut shell charcoal-derived nanocarbon.

Characterization of mechanical properties of epoxy nanocomposites has been investigated on multiple occasions. The introduction of nanocarbon filler into the composite proved to enhance the elastic modulus of the composite [6–8]. The research conducted by Chandra et al. (2022) concluded that the addition of 5 wt% of coconut shell charcoal nanoparticles into the matrix of epoxy resin could improve the elastic modulus of the composite by 13% while reducing its ultimate tensile strength by 12%. Other research by Zakaria et al. (2017) also showed that graphene nanoparticle-filled epoxy nanocomposites' tensile strength and elastic modulus improved by up to 11 % and 17%. Moreover, the tensile strength and elastic modulus of multi-walled carbon nanotubes filled with epoxy nanocomposites improved by up to 26% and 29%.

The mechanical properties of epoxy nanocomposites could be further enhanced by curing in elevated temperature [9–12]. Epoxy is a thermosetting polymer that requires a curing process with the help of a curing agent (hardener) to initiate the polymerization process. The molecular structure of epoxy consists of an amine group that can be converted into interconnected crosslinks with the help of a catalyst. This process process is not very effective at room temperature, resulting on a reduction of mechanical strength in epoxy resin [10,13–16].

Various studies have investigated the curing parameter to determine the optimal temperature and range. Research by Iwan et al. (2024) investigated the effect of curing temperature on the mechanical properties of coconut shell charcoal nanoparticles filled with epoxy resin. This research found that nanocarbon composites experience up to 19% increase in tensile strength, 16% increase in modulus of elasticity, and 84% increase in ductility value by curing it at 80°C for 1,5 hours [17]. Other studies by Saito et al. (2024) showed that the mechanical properties of graphene nanoplatelet epoxy resin composite can be elevated by curing them at 20°C. Other research suggests incorporating preheating the resin for 15 minutes and then curing it for 2 hours at 80°C could improve the performance of the resin by 4% [18]

Nevertheless, there is a paucity of research on the curing holding time of epoxy matrix composites with reinforcing materials from nanosized particles. Therefore, it is necessary to study the effect of curing time on the nanocarbon particle-reinforced epoxy composites. This study will examine the effect of curing time on the nanocarbon particle-reinforced epoxy composites. The addition of nanocarbon to the composite is expected to accelerate the curing process by facilitating more crosslink formation, therefore elevating the mechanical properties of the composite [9,17–19].

2. Material and Methods

This research used epoxy resin, hardener, and nanocarbon from coconut shell charcoal as materials. The epoxy resin employed is an Eposchön brand product comprising Bisphenol A-

epichlorohydrin with cycloaliphatic amine type (EPH 55) epoxy hardener. Both constituents were obtained from PT Justus Kimiaraya, with the ratio of resin and hardener being used is 2:1. The reinforcement comes from coconut shell charcoal nanoparticles, made using the high-energy milling method with a shaker mill machine.[20]

The formation of nanoparticles uses a bottle as the medium. The bottle was filled with a ball mill and coconut shell charcoal refined with a blender. The total volume of the ball mill and coconut shell charcoal is 2/3 of the bottle's volume. Afterward, the bottle was installed on a shaker mill machine to form the nano-size particles. The machine was operated at 200 rpm for 2 million movement cycles [20] Fig. 1 shows the results of carbon particles that have been nanosized.

Figure 1. Nanocarbon particles

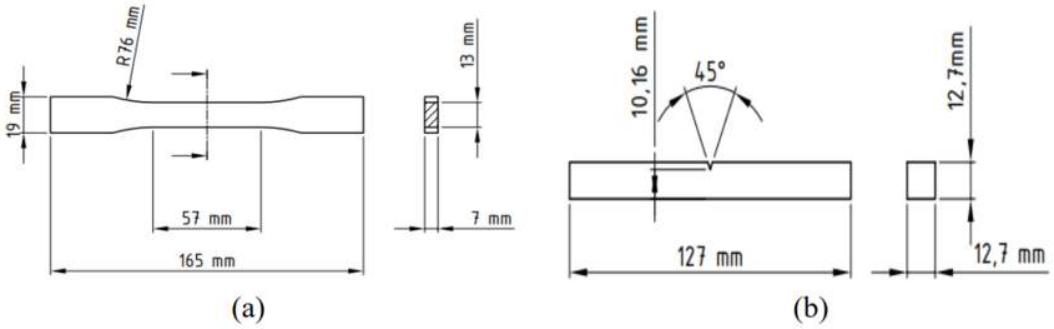

Figure 2. Shape and size of specimen (a) Tensile test, (b) Impact test

The composites were made using silicone-based molding media. The production of composite specimens commences with the fabrication of molds following the tensile test standard, ASTM D638-01, and the impact test standard, ASTM D6110-02 [21,22]The size standard used in the tensile test is ASTM D638-01, while the standard used for the impact test is ASTM D6110-02. Fig. 2 shows the shape and size of the composite specimens.

The composite manufacturing process was initiated with a calculation to determine the volume composition of epoxy resin, hardener, and coconut shell charcoal nanocarbon for tensile and impact test specimens. The next step was to mix the three materials using a stirrer and fill the mixture into the tensile and impact test molds. The composite forming process was carried out for 24 hours at room temperature.

The curing treatment process uses an oven equipped with a temperature control. The temperature used for curing treatment is 80°C, with four variations of holding time: 0.5 hours, 1.5 hours, 2.5 hours, and 3.5 hours. Subsequently, the specimens are tensile and impact tested. Tensile testing was conducted using a 2017 JTM-UTC 220 serial 6604 tensile testing machine. The test specimen was held with a grip that was given an abrasive coating to prevent slipping during the test. The resulting test data is then analyzed to obtain the tensile strength of the specimens. Impact testing was performed using a 1995 GT-7045 8401159 impact testing machine manufactured by Gotech Testing Machines Inc. The impact test specimen features a notch angle of 45°, evaluated through the Charpy method. The test data is then analyzed to obtain the ductility value of the specimens.

3. Results and Discussion

The tensile strength of all specimens obtained by tensile test is presented as a bar chart. The tensile strength of the specimens without and with filler are shown in Fig. 3. It can be seen that up to a variation of 2.5 hours of curing time, there is an increase in tensile strength. The tensile strength is decreased for a more extended curing time variation, 3.5 hours. The tensile strength of the specimens that were not given curing treatment is about 45.1 Mpa. After being cured for 0.5 hours, the tensile strength of the specimen increased to 47.6 MPa. A similar trend is observed for the specimens cured for 1.5 and 2.5 hours. Each experienced increased tensile strength by about 9% to 52.0 MPa for 1.5 hours and about 22% to 57.9 MPa for 2.5 hours of curing time variation compared to the specimen that is not cured. However, the tensile strength of the specimen cured for 3,5 hours decreased by about 4% to 55.5 MPa compared to the specimen cured for 2.5 hours.

The rearrangement of the molecular bond in the composite causes the increase in tensile strength observed [10,13,19]. The curing treatment of the specimens provides energy to rearrange the crosslink bonds. The increase in tensile strength occurs due to the hardener group and the epoxy group forming crosslinks that form bonds with other polymer chains, as shown in the Fig. 4. The number of crosslink bonds formed is increasing according to the time given [17,19]. This is in accordance with the test results shown in Fig. 3, where the tensile strength increases as the curing holding time. The specimen cured for 3,5 hours has lower tensile strength than the specimen cured for 2.5 hours. This phenomenon happened due to the breaking of the main polymer chain of the resin caused by excessively long curing time. Consequently, the tensile strength value of the specimen decreased [17,19].

A similar trend is also observed in the nanocarbon-reinforced specimens. Fig. 3 also presents the tensile strength of the nanocarbon-particle-reinforced specimens. The tensile strength of the specimens increases up to the specimen cured for 2.5 hours. However, the addition of carbon nanoparticles increased the tensile strength by about 6.2% to 48.1 MPa compared to the specimens without filler that were not given curing treatment.

Nanocarbon-reinforced specimens not given curing treatment had tensile strength values around 48.1 MPa. After being cured for 0.5 hours, the tensile strength of the specimen increased to around 54.0 MPa. The specimens cured for 1.5 and 2.5 hours are also observed to have higher tensile strength than non-cured specimens. The tensile strength of 1.5 hours hours-cured specimen increased to around 57.5 MPa. The tensile strength of the 2.5 hours-cured specimens also increased to around 59.3 MPa. Still, a decrease in tensile strength of around 53.2 MPa occurred at a 3.5 hours-cured specimen.

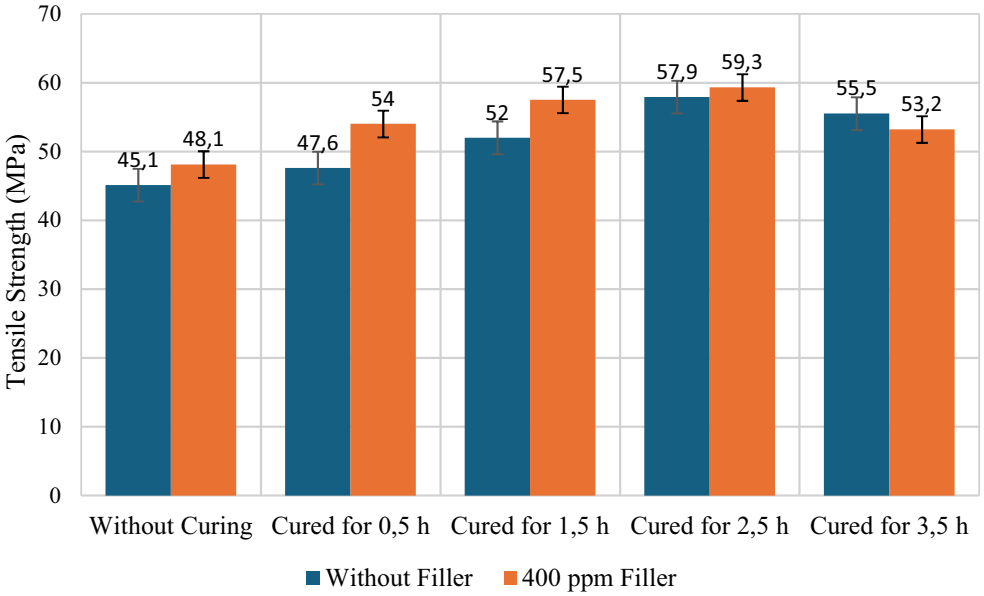

Figure 3. Tensile strength of specimens without and with nanocarbon filler.

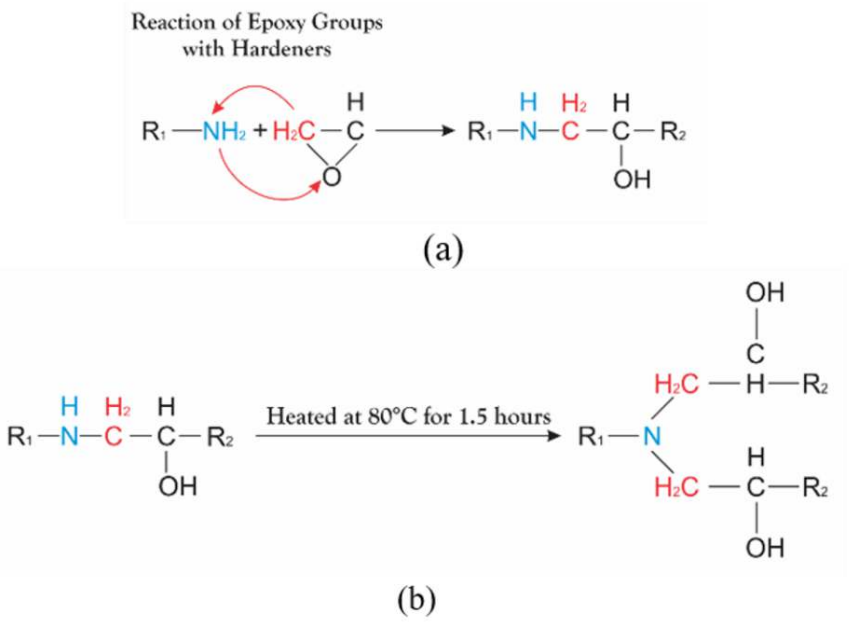

Figure 4. Illustration of crosslink formation.

The tensile strength of nanocarbon-filled specimens without curing treatment is 23% lower than the 2.5 hours-cured specimen. This happened because the nanocarbon reacted with the epoxy, which acted as a catalyst, to form a crosslinking bond. In addition, the insertion of nano-sized carbon into the cross-linking of the epoxy groups also strengthens the bond. The curing

treatment provides energy to rearrange and multiply the cross-link bonds linked to the main chain of the polymer, increasing its strength [17,19].

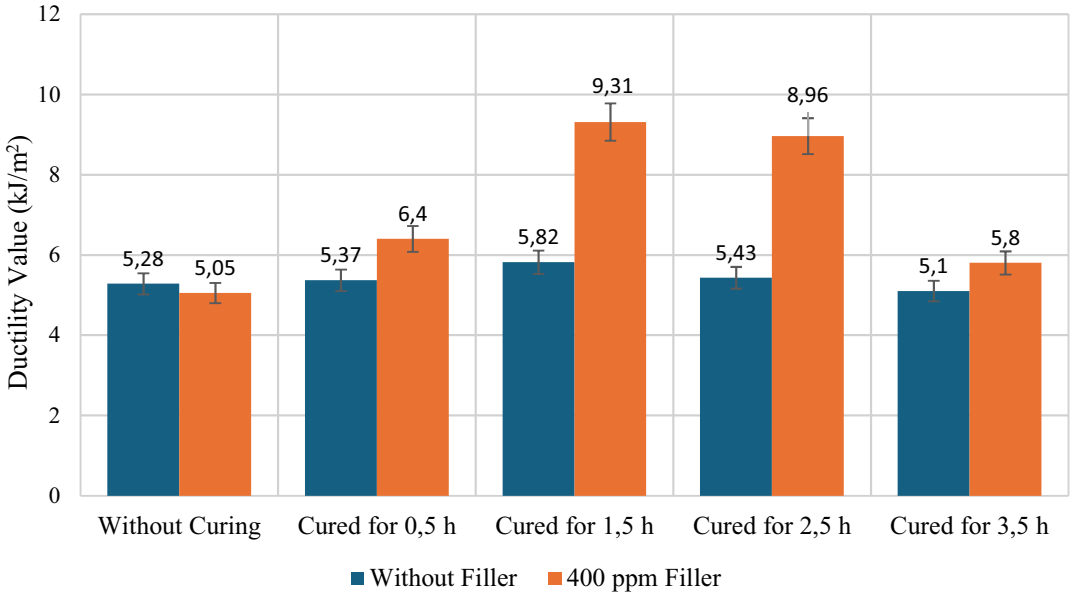

Figure 5. Impact test diagram of specimens without and with nanocarbon filler.

The impact toughness of the specimens with and without filler are given in Fig. 5. The impact toughness increased for the specimens cured for 0.5, 1.5, and 2.5 hours. The 3.5 hours-cured specimens experience a decrease in impact toughness (ductility) value. Specimens without fillers not given curing treatment had an impact toughness value of about 5.28 kJ/m². Then, after being cured for 0.5 hours, the impact toughness value of the specimen increased to about 5.37 kJ/m². The increase in impact toughness value by 10.2% to around 5.82 kJ/m² also occurred in the specimens cured for 1.5 hours. The impact toughness value decreased in the 2.5 and 3.5 hours-cured specimens. The impact toughness values of 2.5 and 3.5 hours-cured specimens are about 5.43 kJ/m² and 5.10 kJ/m² respectively. The decrease in toughness value at curing times above 2.5 hours indicates the breakage of crosslink bonds in the main chain due to excessive curing [17,19].

Fig. 5 also presents the bar chart of the impact toughness of the nanocarbon-reinforced specimens with curing time variations. The impact toughness value of the non-cured specimen is around 5.05 kJ/m2. Compared to the noncured nonreinforced specimen, the noncured reinforced specimen had lower impact strength (about 4.4% lower), which is about 5.05 kJ/m2 compared to 5.05 kJ/m2 in the noncured nonfilled condition.

Curing the specimen for 0.5 hours, the impact toughness value increases to 6.40 kJ/m². The increase in impact toughness value by 84.5% to 9.31 kJ/m² occurs in 1.5 hours-cured specimens, which is very significant. The impact toughness values for 2.5 and 3.5 hours-cured specimens plummeted to around 8.96 kJ/m² and 5.80 kJ/m², respectively. In conclusion, shorter curing of nanocarbon-filled composites enhanced the impact toughness of the composite. The curing causes the specimens to be more capable of absorbing the energy provided and creating more crosslink bonds to resist cracks when exposed to impact loads. Nonetheless, longer curing will promote more breaking of the crosslink bonds eventually reducing the composites' impact toughness value.

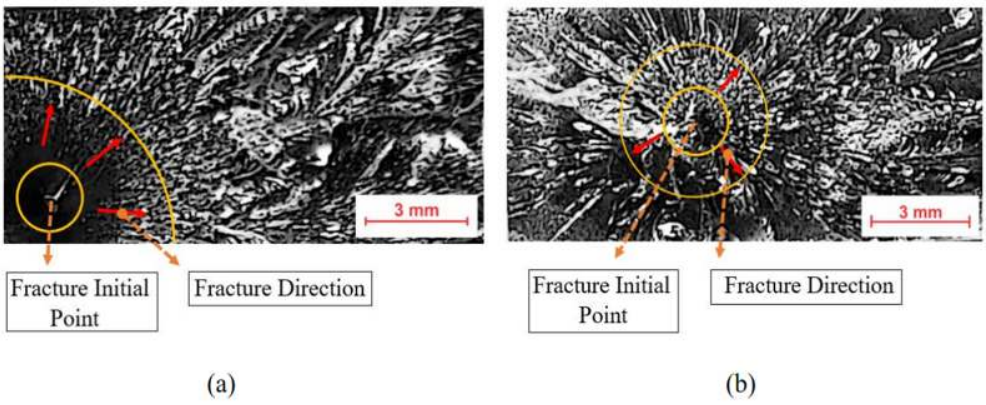

Figure 6. Macro photos of tensile test specimens at curing (a) 0.5 hours, (b) 2.5 hours

Fig. 6(a) showed the fracture shape of the nanocarbon-reinforced specimen with a curing time of 0.5 hours, which had undergone tensile testing. Meanwhile, Fig. 6(b) presented the fracture shape of a nanocarbon-reinforced tensile specimen with a curing time of 2.5 hours, which had undergone tensile testing. The specimen in Fig. 6(b) had higher tensile strength than in Fig. 6(a). It can be seen from Fig 6(a) that the fracture starts from the edge of the specimen's cross-section and causes the tensile stress in the specimen to be unequal. The black area at the beginning of the fracture point indicated that the fracture process takes place very quickly with a smooth and small fracture surface. The direction of crack propagation has a radial spread pattern starting from the beginning of the crack towards the outer side, as shown by the red arrow. The spread closer to the outside creates a coarser and larger crack.

Fig. 6(b) presented the fracture shape of the nanocarbon-reinforced tensile specimen with a curing time of 2.5 hours, which had a higher tensile strength. The fractures tend to show brittle properties like any other thermoset fracture. This can be seen from the crack growth direction, which forms a radial spread from the inside to the outside, as shown in the red arrow. The crack starts from the center point, which is a point with more stress concentration. The tensile stress is retained more evenly by the cross-section of the specimen, which causes greater tensile strength at failure. The crack at the beginning has a white area that tends to be rough because the specimen can resist the stresses. After that, the crack spreads to the outer area of the specimen, which causes the fracture surface to have a large white area.

4. Conclusion

The curing treatment has significantly impacted the mechanical properties of both non-filled and nanocarbon-filled composites. The addition of nanocarbon particles increases the tensile strength and impact toughness value of the composites for the same curing time. Moreover, addition of the nanocarbon particles reduces the curing time needed for the composite to reach the same value of the tensile strength and impact toughness. The combination of nanocarbon and curing treatment resulted in various strengths, but the increase in impact test values was more significant because the ability of the specimens to resist cracks after loading was better compared to the tensile test values, which were vulnerable to cracks because they were clamped on two sides.

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