

Effect of curing temperature on the mechanical properties of coconut shell nano carbon reinforced composites with epoxy matrix

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Abstract. This study aimed to examine the role of curing temperatures at 40, 60, and 80 degrees Celsius with an ageing time of 1.5 hours on the tensile strength, modulus of elasticity and ductility value of coconut shell nanocarbon-reinforced composite materials. The nanocarbon uses a top-down approach with high-energy milling (HEM). This study found that adding coconut shell nano carbon increased the tensile strength by 4.6% from 46 MPa, but the ductility value decreased to 4.2% from 5.28 kJ/m². The curing treatment of nanocarbon composite gives the effect of increasing the tensile strength by 19.7% to 57.5 MPa, the modulus of elasticity increase by around 16.7%, becoming 3.80 GPa and the ductility value increase by 84% to 9.30 kJ/m², that occurs at 80 degrees Celsius temperatures. The curing treatment of epoxy resin gives the effect of increasing the tensile strength by 20% to become 55,2 MPa, the modulus of elasticity growth around 10,5% from 2,96 GPa and the ductility value 11% of 5,28 kJ/m², that occurs at 80 Celsius degrees temperatures.

1 Introduction

The development of materials engineering, especially in composites, has begun to use natural materials as constituent materials. The advantages of composite materials are high strength, lightweight, relatively low production cost and corrosion resistance [1]. Research has been conducted on the effects of adding natural material fillers to composites. One of them is the addition of filler from coconut shell charcoal [2]. The carbon content in coconut shell charcoal is around 88% [3].

Coconut shell charcoal as nano-size reinforcement has yet to be widely used. One study related to using 1000 mesh coconut shell charcoal as a filler in composites with epoxy matrices increased the modulus of elasticity, increasing stiffness, whereas the tensile strength value decreased [4].

The development of materials engineering technology has reached the stage of manufacturing nanomaterials with tiny sizes. Nanomaterials are materials with a size range of 1-100 nm. Nanomaterials have different characteristics, as they have a larger particle surface area than macro sizes. The larger surface area makes nanomaterials more reactive

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and has significant total surface energy. Therefore, the coconut shell charcoal in nano size is used as an alternative filler in composites.

Carbon particles used in nano size significantly affect composites' mechanical properties [5–8]. Coconut shell charcoal particles in nano size have unique characteristics. Nanocarbon has tremendous surface energy and a small fraction of surface atoms that are quickly loose & free.

Carbon particles added to composites with epoxy matrix have a significant effect with increased flexural strength and impact strength [9]. Graphene-type carbon particles (GNP) and multiwall carbon nanotubes (MWCNT) are used as filler materials in epoxy nanocomposites [10]. Adding 1% MWCNT has resulted in a tensile strength increase of about 26% to 46 MPa and a flexural strength increase of about 29% to 129 MPa. Fibre-added composites with epoxy matrix can increase the value of impact resistance (ductility) [11,12].

One of the resins that is widely used as a composite material is epoxy resin. Epoxy resin is a thermosetting polymer with an amorphous structure and requires a curing agent (catalyst) for the polymerization process [13,14]. Epoxy resin molecules contain two elements. Epoxy and amine groups react with hydrogen atoms on the catalyst to produce interconnected cross-links [15,16]. The curing process at room temperature is rarely fully formed, causing the epoxy not to have optimal strength. Studies on the characteristics of pure epoxy resins have been carried out to compare the mechanical properties, tensile strength, strain and elastic modulus of cured epoxy resins.

Some studies show that there are indications that preheating for 15 minutes and curing for 2 hours at 80 degrees Celsius increases the tensile strength of epoxy resins by about 4% [17].

Studies about the curing treatment of epoxy composites with carbon nanoparticle fillers have yet to be conducted. Therefore, this study will examine the effect of curing temperature on epoxy composites reinforced with nanocarbon particles from coconut shell charcoal. Adding carbon to the composite is expected to increase the number of crosslinked epoxy molecules. This crosslink will improve the mechanical properties of nanocarbon-reinforced epoxy composites [18]. Research on the effect of curing temperature variation on carbon nanotube-reinforced composites from coconut shells is expected to form better crosslink bonds to make the composite have more optimal mechanical properties.

2 Materials and methods

2.1 Tools and materials

Composite test specimens were made using several tools and materials. The materials used were epoxy resin of Eposchön brand with Bisphenol A -Epichlorohydrin type and catalyst (hardener) of Cycloaliphatic Amine type (EPH 555). Epoxy resin has a 1.20 gr/cm³ density and a shrinkage rate of about 1-5%. Coconut shell charcoal is used as a composite reinforcement material. Coconut shell charcoal was made into nano-sized particles.

The composite specimens were moulded using silicon from Silicone Rubber RTV48. Silicone moulds are used to form specimen patterns for tensile testing and impact testing.

2.2 Nanocarbon manufacturing

The initial material in coconut shell charcoal powder is put into the tube as much as 1/3 volume and added to the ball mill as much as 1/3 volume of the tube container.

Nanocarbon is produced using a shaker mill machine with the High Energy Milling (HEM) method [19]. About two million shaking cycles with a rotation speed of 260 rpm

make carbon into nano-size. The nanocarbon is shown in Figure 1. Nanocarbon should be stored using sealed containers. Carbon in the open state in free air can react with oxygen.



Fig. 1. Nanocarbon from coconut shell.

2.3 Composite manufacturing

The composite manufacturing process uses the hand lay-up method. The volume fraction added to the nanocarbon composite is 400 ppm. It takes about 10 minutes with a stirring rotation speed of around 500-800 rpm for the base solution to mix evenly with the epoxy. After 10 minutes of stirring, the catalyst (hardener) was added to the mixture with a ratio of 2:1 and stirred again for 5 minutes until evenly mixed. The final step of the composite solution is poured into the silicone mould. The shape and size of the mould as shown in Figure 2.

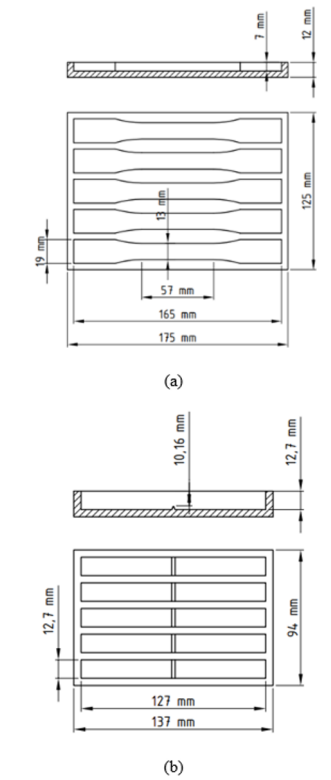


Fig. 2. Shape and size of specimen mould. (a) Tensile test specimen, (b) Impact test specimen.

2.4 Curing Process

Curing heat treatment was carried out using an electric oven with temperature variations of 40, 60, and 80 degrees Celsius for 1.5 hours. The oven was equipped with a thermocouple as a tool to monitor the stability of the oven temperature. The cured specimens are pure epoxy resin specimens without the added filler and the carbon nanocomposite reinforced epoxy matrix from coconut shell.

2.5 Tensile Testing

Tensile test using ASTM D 638-01 standard. The dimensions of the tensile test specimen are shown in Fig. 3. The tensile testing machine used was JTM-UTC 220 Serial 6604 in 2017, owned by the materials laboratory of Sanata Dharma University Yogyakarta.

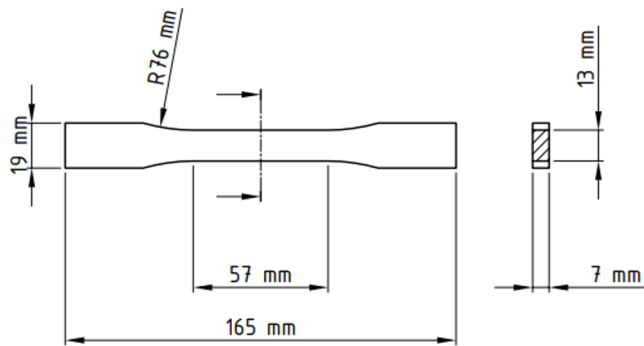


Fig. 3. Shape and size of tensile test specimen.

Tensile testing produces data on the maximum load received by the specimen and the strain shown on the graph. Based on the test data, the tensile strength and elastic modulus values are obtained. The fracture in the specimen is analyzed by macro photographs of the specimen's cross-section. The analysis is carried out to determine the type of fracture that occurs and examine the characteristics of the test object from the resulting fracture shape.

2.6 Impact testing

The Charpy method used to impact testing refers to the ASTM D 6110-02 standard shown in Fig. 4. The impact test equipment used is GOTECH TESTING MACHINE, Model GT-7045 series 8401159, belonging to the materials laboratory of Sanata Dharma University Yogyakarta.

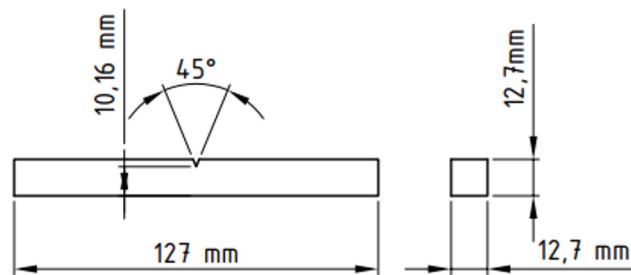


Fig. 4. Shape and size of impact test specimen.

3 Result and discussion

3.1 Tensile test result

The research results related to curing treatment given to pure epoxy resin specimens with temperature variations of 40, 60 and 80 Celsius degrees can be seen in Fig. 5.

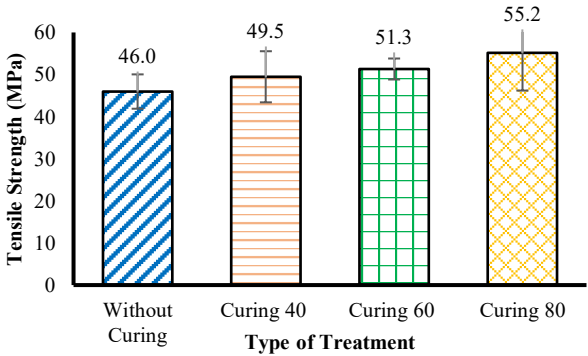


Fig. 5. Tensile Strength of epoxy resin with curing treatment.

The tensile strength increased from 46 MPa in specimens without curing treatment to 48.7 MPa for 40 °C cured specimens. In the 60 °C curing test specimen, the tensile strength value increased to 51.3 MPa, and for the 80 °C curing test specimen, the tensile strength value of 55.2 MPa was the highest. The tensile strength value of pure epoxy resin specimens with curing treatment increases with increased curing temperature. The tensile strength of epoxy resin increases due to the energy added in the curing process. At 80 °C, the tensile strength value increased around 16.7% from 46 MPa of epoxy resin without curing treatment.

The tensile strength value of pure epoxy resin treated with curing increases due to energy through heat given at a specific temperature. Reactions occur between epoxy molecules. Crosslinks are re-formed and improve mechanical properties to be more optimal. The molecules in the epoxy resin undergo the most optimal changes at a temperature of 80 °C. The relatively low curing temperature indicates that the heat energy provided impacts the increased tensile strength value.

Based on previous research, the curing process naturally occurs due to the reaction between the epoxy resin group (Bisphenol A) and the curing agent. The reaction of the molecules forms crosslinks in the polymer chains [20,21]. The arrangement of epoxy groups is broken when reacting with the catalyst. The epoxy groups then form bonds between other epoxy groups. The formation of more crosslinks causes the composite to become stiffer.

The curing reaction mechanism is illustrated in the figure below. Figure 6 is the reaction between epoxy groups with a curing agent (hardener). The reaction produces a crosslink shown on the right side with one chain bond between the epoxy group and the hardener [22]. The curing treatment process at 80 °C for 1.5 hours causes an increase in tensile strength, as seen in Figure 5. The presence of energy through the given 80 °C temperature causes the bond chain arrangement to react again with other bond chain groups. Under these conditions, the most optimal crosslinks formed are illustrated in Figure 6 b, which shows that more crosslinks are formed due to curing.

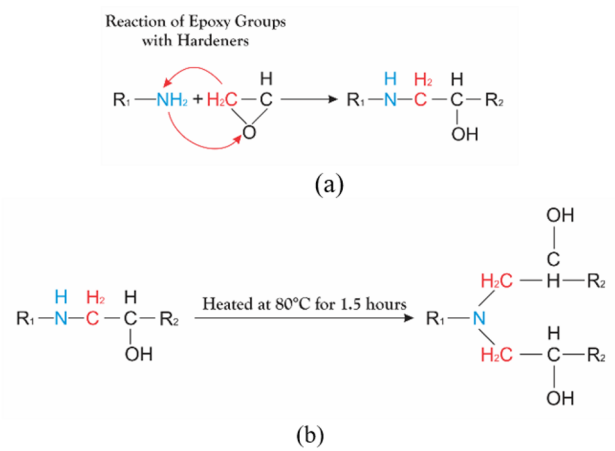


Fig. 6. Illustration of the crosslink of epoxy with curing treatment.

Stiffness is one of the characteristics that a material has to accept stress without deformation, shown through the elastic modulus value. The pure epoxy resin test specimen has an elastic modulus value shown in Figure 7. The elastic modulus is obtained from measuring the ratio between stress and strain in the elastic curve area.

The average elastic modulus diagram of epoxy resin treated with curing shows almost uniform values. The average elastic modulus value with curing treatment has increased by about 10%, from 2.96 GPa to 3.2 GPa. Epoxy resin specimens without curing treatment have an average elastic modulus value of 2.96 GPa. Epoxy resin specimens treated with curing at 40, 60, and 80 degrees Celsius have an average elastic modulus value of about 3.28 GPa, 3.24 GPa, and 3.27 GPa.

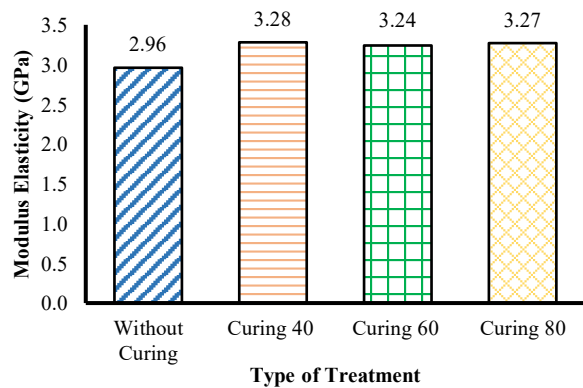


Fig. 7. Modulus of elasticity of epoxy resin with curing treatment.

Figure 8 shows the tensile strength of carbon nano-reinforced epoxy composites with curing treatment. It can be seen that the nano carbon composite without curing treatment has a tensile strength value of 48.1 MPa. The addition of 400 ppm of nanocarbon increases the tensile strength value to 48.1 MPa, about 4.6%. The tensile strength value increased to about 52.3 MPa due to the curing process at 40 °C. In the curing process with a temperature of 60 °C, the tensile strength value is about 51.5 MPa. In the curing treatment with a temperature of 80 °C, the tensile strength value is around 57.5 MPa.

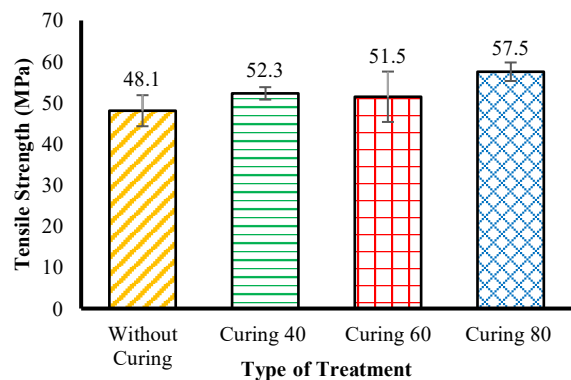


Fig. 8. Tensile strength of nanocarbon-epoxy composites with curing treatment.

Overall, the tensile strength value of nanocarbon-reinforced composite specimens from coconut shells treated with curing increased compared to nanocarbon composite specimens without curing treatment. At 80 °C temperature variation, the strength value of nano carbon reinforced composite specimens from coconut shells increased by around 16.5% to 57.5 MPa.

The results above are in line with several studies that have been conducted previously. Adding multi-walled carbon nanotube (MWCNT) as much as 0.75% increased tensile strength, toughness and elastic modulus values [23]. Adding 1.5% multi-walled carbon nanotube (MWCNT) to epoxy resin composites increased the tensile strength by 21% [24]. Using carbon nanotube (CNT) and carbon fibre as much as 0.5wt% increases the value of tensile strength and elastic modulus [25].

The increased tensile strength of carbon nano-reinforced composites is because of the carbon particles used in the nano-size [26]. Carbon from coconut shell charcoal has good thermal properties [27]. With heat energy provided through the curing process, nano-sized carbon particles can insert into the crosslinking of epoxy resin. Carbon nano has the characteristics of binding and producing bonds with other groups, as illustrated in Figure 9. The presence of carbon particles and curing treatment makes the composite crosslink chain more and more, thus giving an influence, namely the increasing tensile strength value of nanocarbon reinforced composites.

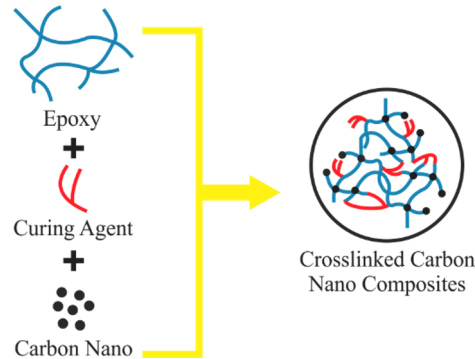


Fig. 9. The illustration of crosslinking of nanocarbon composite.

The modulus of elasticity is obtained by measuring the ratio between stress and strain in the elastic curve area. Coconut shell nano carbon reinforced epoxy matrix composites have elastic modulus values shown in Figure 10.

The average elastic modulus diagram of coconut shell nano carbon reinforced epoxy matrix composites shows similar values. The composite without curing treatment has an

average elastic modulus value of around 3.30 GPa. In contrast, the carbon nanocomposite treated with curing at 40 °C, 60 °C and 80 °C has modulus elasticity values of 3.36 GPa, 3.69 GPa, and 3.85 GPa, respectively.

Materials have basic properties that show their characteristics. Epoxy resin has good basic properties of stiffness. Research on the mechanical properties of nanocarbon-reinforced composites from coconut shells with epoxy matrices tends to be dominated by polymers (epoxy resins). The addition of nanocarbon as reinforcement is relatively small at 400 ppm, so the composite material has the basic properties of epoxy resin. The increase in the elastic modulus value of the effect of curing treatment on nanocarbon-reinforced composite material from coconut shell with epoxy matrix produced is still within the range of elastic modulus of epoxy resin.

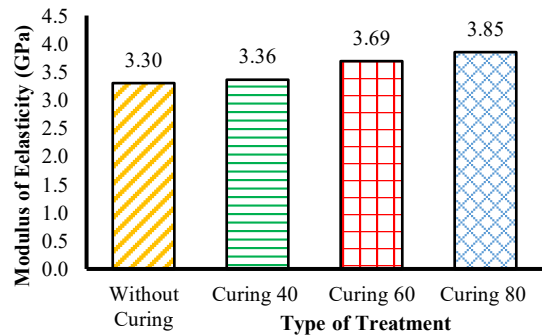


Fig. 10. Modulus of Elasticity composite of nanocarbon-reinforced epoxy matrix.

3.2 Impact test result

The curing treatment process with temperature variations on pure epoxy resin test specimens can be seen in Figure 11. Overall, the ductility value of epoxy resin with curing treatment has increased slightly with increasing temperature. Pure epoxy resin without curing treatment has a ductility value of 5.28 kJ/m². At curing temperature variations of 40 °C, 60 °C and 80 °C, the ductility value of epoxy resin increases to 5.40 kJ/m², 5.70 kJ/m² and 5.85 kJ/m².

The increase in the ductility value of epoxy resin is due to the energy provided through the curing process using temperature. At a temperature of 80 °C, the energy is getting bigger; these conditions cause the molecules that make up the epoxy resin to stretch easily, moving with each other to rearrange the cross-links flexibly. The increase in temperature has an impact on the curing process. Namely, there is energy to make the material re-form more cross-links, causing the material to have more optimal mechanical properties.

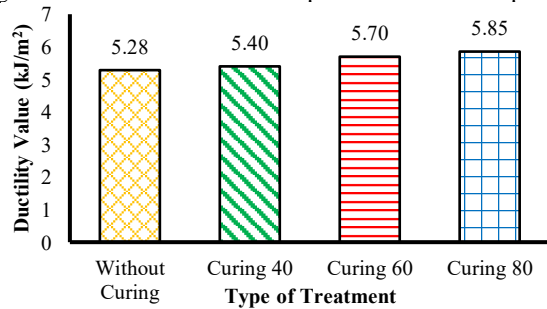


Fig. 11. Average ductility e of pure epoxy resin.

In the nanocarbon-reinforced composite material treated with curing, data on the ability of nanocarbon composite test specimens to absorb energy shown by the increasing value of composite ductility can be seen in Figure 12. The ductility value of nano carbon reinforced composites from coconut shell with epoxy matrix treated with curing as a whole increased as the curing temperature used increases.

The nanocarbon-reinforced composite specimen without curing treatment has a ductility value of 5.05 kJ/m². The addition of carbon in nano size as much as 400 ppm decreases the ductility value from 5.30 kJ/m² to 5.05 kJ/m² or around 4.2%. In nano carbon composite specimens treated with curing temperature variations of 40 °C, 60 °C and 80 °C, the ductility value of epoxy resin increases to 6.15 kJ/m², 7.34 kJ/m² and 9.31 kJ/m².

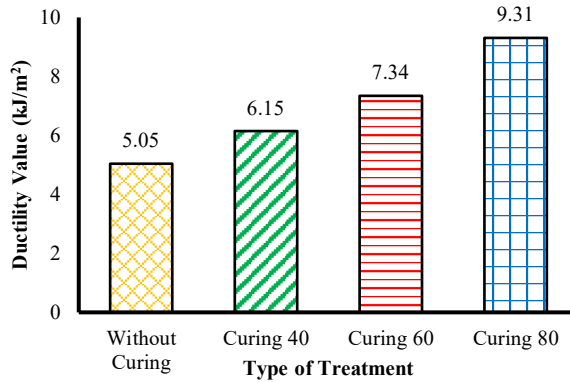


Fig. 12. Average ductility of nanocarbon composites.

The addition of carbon in nano size initially caused a decrease in the ductility value of the material. However, the ductility value of the nanocarbon-reinforced composite from coconut shell with epoxy matrix increased due to the energy provided through the heat in the curing process. Carbon in nano size can respond to the heat energy provided to move into the crosslink and bind the epoxy group so that the ductility value of the material increases. The curing process naturally occurs when the epoxy resin reacts with the curing agent (hardener); when the reaction takes place, there is an increase in the density of the crosslinks, which gradually increases until it finally becomes solid. The curing process using temperatures above room temperature provides heat energy that makes the material molecules of epoxy resin and (atomic) carbon as filler move to become flexible and then bond to re-form the crosslinks to be more optimal.

3.3 Fracture analysis of tensile test

The failure of the nanocarbon-reinforced composite material is further investigated by looking at the shape of the fracture resulting from the tensile test. The shape of the fracture is seen and compared between the shape of the fracture of the nanocarbon composite specimen that is not treated with curing and the specimen that is treated with curing. The fault shape visually has a relatively flat surface and forms a pattern. There is a shiny surface as if the centre point of the fault occurs there. Based on these characteristics, the fracture shape of the nanocarbon composite specimens, whether treated with curing or not, is a brittle fracture. The fracture failure of the specimens occurs quickly, and there is no plastic deformation.

The shape of the fracture is reviewed more specifically using macro photographs. The results of macro photographs of carbon nano-reinforced composite specimens without curing treatment are shown in Figure 13 a.

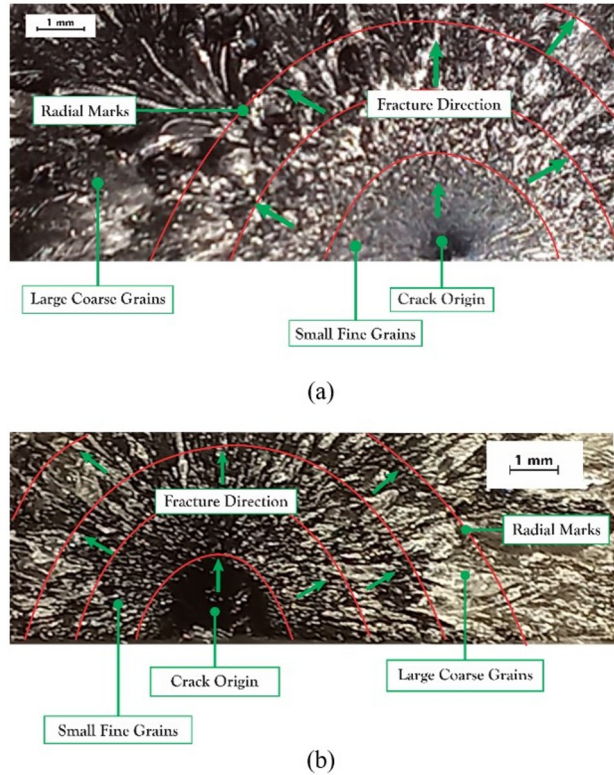


Fig. 13. Fracture shape of nanocarbon-epoxy composite tensile specimens. (a) without curing treatment (b) with curing treatment.

Macro photographs of the nanocarbon-reinforced composite specimens without curing treatment were then identified for the characteristics of the faults formed. Observations from macro photos look like a centre point which is the initial position of the fault. In the area near the centre of the fault, it looks like refined grains are getting out away from the centre of the fault area; the pattern looks like coarse grains. Refined grains on the fracture of carbon nanocomposite specimens without curing treatment appear more dominant than the shape of nanocarbon composite specimens treated with curing, as shown in Figure 13 b.

The characteristics of the fault shape that occurs in the nanocarbon-reinforced composite material treated with curing are identified to show the possible area as the fault's starting point, the direction of the fault propagation, and the stress mechanism that occurs in the material. The resulting fault shape resembles a rosette pattern; a centre point indicates the initial fault and then spreads outward.

In that area, the highest stress occurs, which develops into the starting point of the fracture and propagates to the surface. The position of the fault centre is slightly shifted from the centre point of the cross section due to a slight tilt in the installation of the tensile testing machine. In the area near the fault centre point, small and refined grains appear to form a pattern with boundaries resembling radial marks. The further away from the centre point of the fault, the larger and coarser the grains appear. The shape of the fault that occurs in the outer area of the fault centre looks like a chunk or shard. The shape of the large fragments indicates that the composite material has brittleness on the surface of the composite material. The fracture pattern formed shows a slight radial mark that follows the direction of the fracture towards the surface, showing its ductility. Carbon nanocomposite material treated

with curing has a little ductility indicated by the shape of the fracture pattern, such as a rough curved radial shape at the centre point of the fracture following the plane of the pattern formed from the fracture.

4 Conclusion

Following the discussion that has been carried out and looking back at the problem formulation that has also been written in the introduction, two conclusions can be drawn. First, the curing treatment of epoxy resin material without adding filler can increase the material's tensile strength, elastic modulus and ductility value. The most optimal increase in tensile strength occurred at a curing temperature of 80 °C, about 20% of the 45.9 MPa average tensile strength value of pure epoxy resin that was not treated with curing. The modulus of elasticity of the epoxy resin increased by about 10.5% from 2.96 GPa after curing. The increased ductility value of epoxy resin treated with curing at 80 °C is the highest, about 11% of 5.28 kJ/m².

Second, the addition of carbon derived from coconut shell in nano size has an impact on increasing the tensile strength by about 4.6%, from 45.9 MPa, while the ductility value of nano carbon reinforced composite material has decreased by about 4.2% from 5.28 kJ/m². The curing treatment process carried out on carbon nano-reinforced composite materials increases the tensile strength, modulus of elasticity and ductility value of the material. The most significant increase in tensile strength value occurs in curing treatment at 80 °C temperature, about 19.7%. The elastic modulus of epoxy resin increased by around 16.7% to 3.85 GPa after curing. The increase in the ductility value of carbon nano-reinforced composites treated with curing at 80 °C is the highest, about 84% to 9.31 kJ/m².

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