

# Study on Increasing the Cracking Resistance of Unsaturated Polyester Composites with MMA and Rice Husk Fiber Reinforcement

Nusyirwan Nusyirwan<sup>1\*</sup>, Rully Pratama<sup>2</sup>

\* <sup>1</sup>Department of Mechanical Engineering, University of Andalas, Padang 25163, Indonesia

Received: 14 January 2023, Revised: 2 April 2023, Accepted: 3 September 2023

## Abstract

Polymer composite materials have been widely developed as an alternative material to replace metal materials due to various advantages such as having relatively good mechanical strength, low density, and easy manufacturing process. However, polymers have many disadvantages in that they easily crack when impacted. One of the most widely used materials for composite matrices is unsaturated polyester polymers which are widely used in vehicle, aircraft, and ship hull components as well as vehicle components. The crack study is crucial because it causes the material to no longer be able to support loads below its yield strength, therefore causing failure to occur more quickly. One way to overcome material failure due to cracks in composite materials is to prevent crack propagation by adding reinforcing materials. In this study, a composite material was made using rice husk fiber to increase the crack resistance of the polyester composite matrix. From the results of the crack test, it is known that there is a tendency to increase the percentage variable of adding rice husk by 5%, 10%, 15%, and 20%. The expected value obtained to determine the crack resistance of the material is the value of the stress intensity factor (K1c). The highest K1c value was obtained at a 15% rice husk percentage variable of 1.558 MPa.m<sup>0.5</sup>, and this value could increase the value of the pure polyester stress intensity factor (K1c) by 0.667 MPa.m<sup>0.5</sup>, indicating an increase of 233.58%.

**Keywords:** polyester, rice husk particle fiber, intensity-tension factor

## 1. Introduction

Technological developments in polymer materials, one of the goals is to find alternative materials to replace metal materials for vehicles, ships, and aircraft construction components [1, 2]. This is done because the metal material has a high specific gravity, so it requires considerable driving energy when used for this purpose [1, 3, 4]. One of the materials chosen is polymer to replace metal materials because polymers have low specific gravity, but not all construction components can be replaced by polymers [2, 5, 6]. The problem is that polymers have many weaknesses, including low mechanical strength, easy to crack, and no high-temperature resistance [4, 7]. The polymers that are widely used as matrices in composites are polyester and vinyl ester [5, 8]. But polyester has a relatively low price compared to vinyl ester. Efforts to increase the mechanical strength of polymers have been carried out, including reinforcing them with synthetic fibers or natural fibers [4, 9, 10]. This material has good mechanical properties when combined with fiber as reinforcement to form composite materials [11, 12]. Another method for strengthening polymers is to combine two polymers that have similar properties, for example, mixing polyester with vinyl ester [9, 13, 14]. In its application, composite materials still have many weaknesses, including not being

resistant to collisions which will cause cracks which are the main trigger for failure [15, 16].

One of the things studied in this research is how to increase the crack resistance of unsaturated polyester polymers to various added rice husk fiber (SP) reinforcement mixtures. Previous studies have discussed improving the mechanical properties of polyester against tensile loads, impact loads, and bending loads [3, 8, 9]. But no information discusses the increase in crack resistance of this unsaturated polyester polymer against reinforcement from rice husk fibers against the crack resistance of composites with unsaturated polyester matrices [10, 17, 18]. So, this research will aim to increase the crack resistance of polyester material by reinforcing it with rice husk fiber particles for various percentages of a particular mixture [9, 14].

## 2. Experimental/theoretical method

### 2.1. Material

In this research, strengthening the crack resistance of polyester polymer with reinforcement of rice husk particles was carried out to obtain a composite material that has good crack resistance properties from polyester polymer without reinforcement [6, 9, 19]. Some of the materials used to make composite materials include the following;

\*Corresponding author. Email: nusyirwan1802@gmail.com.

© 2023. The Authors. Published by LPPM ITS.

**Table 1.** Polyester Mechanical Properties [9]

Item	Unity	Value
Tensile strength	MPa	20-100
Tensile modulus	GPa	2.1-4.1
Ultimate strain	%	1-6
Poisson's ratio	-	-
Density	g/cm <sup>3</sup>	1.0-1.45
Tg	°C	100-140
CTE	10 <sup>-6</sup> /°C	55-100
Cure Shrinkage	%	5-12

**Figure 1.** Rice Husk Particles

### 2.1.1. Polyester

Polyester is a polymer commonly used as a matrix to form composite materials when mixed with rice husk fiber particles to increase the expected crack resistance properties. The mechanical properties possessed by polyester are relatively good, and the material is inexpensive [4,5]. The following are some properties of polyester, among others: polyester has fairly good tensile strength, resistance to stretch, chemicals, and mildew, has excellent abrasion resistance, easy maintenance, has water-repellent properties, and dries quickly. The type of polyester used in this study was unsaturated polyester with Yukalac 1560 BL-EX product. The mechanical properties of polyester can be seen in Table 1.

### 2.1.2. Rice Husk Particle

Rice husk is the outer part of the grain, which is a by-product of rice milling. One of this research's objectives is to use rice husks to increase the strength of cracking resistance of polyester composite materials. The chemical composition of rice husk consists of relatively high levels of active silica (SiO<sub>2</sub>), namely 94-96% of husk ash, other components, CaO, MgO, Al<sub>2</sub>O<sub>3</sub>, and NaO<sub>2</sub>, so that from the chemical composition of rice husk, it can be used as a source raw materials for silica-based materials. Apart from being abundant, another supporting factor is the ease with which silica can be extracted simply and at a relatively low cost, namely using alkaline extraction or ashing, as shown in Table 2.

**Table 2.** Composition of Rice Husk [9]

No	Component	Content Percentage (%)
1	Water content	9.02
2	Crude protein	3.03
3	Fat	1.18
4	Coarse fiber	35.68
5	Ash	17.71

**Figure 2.** Hot plate magnetic stirrer

### 2.1.3. Rice Methyl Methacrylate (MMA)

Methyl methacrylate, often referred to as MMA, is a polymer material that has biocompatible properties, which makes MMA a research material in the study of biomedical material literature [20]. The MMA molecules that are joined in the bond chain lead to the distance between the bonds in the polyester bond, reducing the stiffness of the polyester network structure [7]. MMA is a colorless polymer material with a relatively low cost and excellent fracture resistance [21]. Mixing MMA with thermosetting resins can reduce the viscosity of the polymer blend [22]. Adding MMA here is expected to make the network structure of polyester homogeneous [3]. In previous studies, the impact value was increased with the addition of 10% MMA [3].

### 2.1.4. Catalyst MEKP

The catalyst used is the Mepoxe catalyst produced by PT. Justus Kimiaraya. The function of the catalyst is as a catalyst to accelerate the drying rate of polyester. The catalyst is 4% for polyester alloys [3].

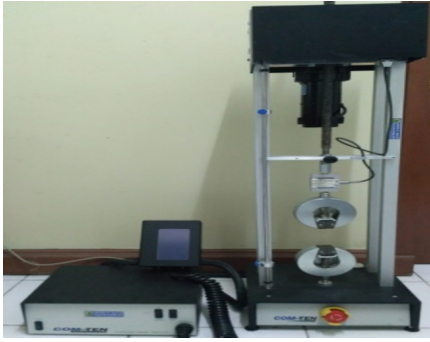


Figure 3. COM-TEN 95T Series 5 K Crack Testing Machine

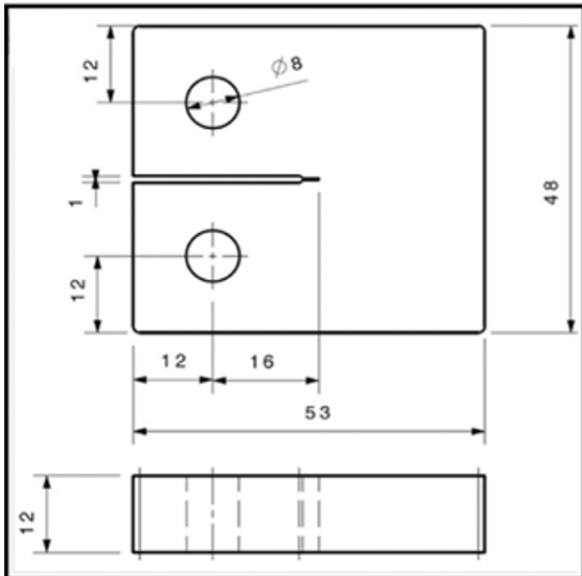


Figure 4. The dimensions of the test specimen are appropriate ASTM D5405 standards

2.1.5. Research Method

This study conducted a crack test on polyester composite materials reinforced with rice husk fiber by giving grooves notches and initial cracks. The test material will be subjected to a vertical tensile loading on both sides which gradually increases until the maximum load that the material can withstand is reached until it experiences a total crack or maximum fracture toughness of the material, following ASTM D5405 Standard [23]. The stress intensity factor is the magnitude of the stress distribution that occurs at the crack tip for the material given the initial defect. The magnitude of the stress intensity factor of a material with specific dimensions against a given load until the material experiences total cracking is called the critical stress intensity factor. This term is known as fracture toughness, denoted by ( $K_{Ic}$ ).

2.1.6. Preparation of Test Samples

This study conducted a crack test on polyester composite materials reinforced with rice husk fiber by giving grooves

notches and initial cracks. The test material will be subjected to a vertical tensile loading on both sides which gradually increases until the maximum load that the material can withstand is reached until it experiences a total crack or maximum fracture toughness of the material, by ASTM D5405 Standard [23]. The stress intensity factor is the magnitude of the stress distribution that occurs at the crack tip for the material given the initial defect. The magnitude of the stress intensity factor. In this study, the composite material was made from a polyester matrix with reinforcement of rice husk particles with the ratio of polyester to rice husk particles as follows 100%: 0%, 95%: 5%, 90%: 10%, 85%: 15% and 80%: 20% of each mixture will be compared to the fracture toughness of all the percentages of the mixtures made and will be compared to pure polyester without reinforcement.

2.1.7. Testing Procedure

The hot plate magnetic stirrer is used as a stirrer for the mixed matrix and reinforcement with a temperature that can be adjusted as desired so that the composite material is well formed. The specifications of the hot plate magnetic stirrer are as follows. brand Daihan Scientific, Model MS-H280-Pro, Working Temperature 25–280°C, rotation 0 – 1500 rpm.

The crack test tool is used for crack testing of composite samples that have been made according to the referenced standard, namely ASTM D5405. The dimensions can be seen in Figure 2. This tool will be able to input material specification data needed for analyzing the crack strength properties of composite material samples. The specifications of the crack testing machine are as follows; Brand COM-TEN testing machine 95T Series 5K, Capacity 5000 Pounds, load Cell Model TSB0050, with a touch screen monitor display system or com-touch screen.

After carrying out the composite material crack testing process, then the measurement results will be entered into the following equations 1 and equation 2:

$$K_{Ic} = \frac{P}{BW^{\frac{1}{2}}} \cdot f\left(\frac{a}{w}\right) \tag{1}$$

$$f\left(\frac{a}{w}\right) = \frac{(2 + \frac{a}{w})\{0.886 + 4.64(\frac{a}{w}) - 13.32(\frac{a}{w})^2 + 4.64(\frac{a}{w}) - 13.32(\frac{a}{w})^2\} + 14.72(\frac{a}{w})^3 - 5.6(\frac{a}{w})^4}{(1 - \frac{a}{w})^{3/2}} \tag{2}$$

interpreted as:

- $K_{Ic}$  = Stress intensity factor (MPa.m 0.5)
- P = Maximum load (kN)
- B = Specimen thickness (cm)
- W = Specimen width (cm)
- a = Crack length (cm)

**Table 3.** Mixed composition of composite

Sample No.	UP (wt %)	SP (wt %)	MMA (wt %)	MEKP (wt %)
1	100	0	10	4
2	95	5	10	4
3	90	10	10	4
4	85	15	10	4
5	80	20	10	4



**Figure 5.** Casted polyester and rice husk composite specimens

### 3. Results and Discussion

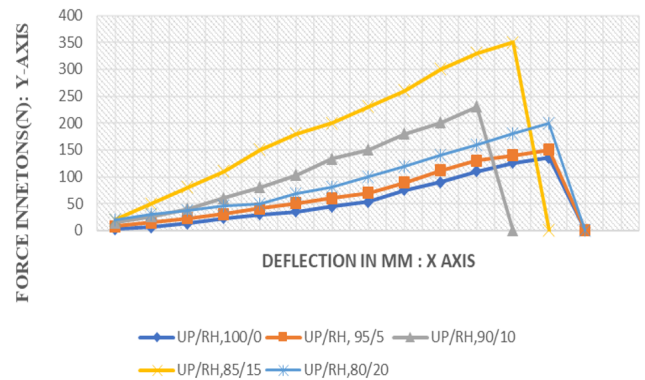
The main work in this research was to produce a polyester composite mixture (UP) with rice husk particles (SP) and MKEP and MMA catalysts according to the percentage of each mixture and freeze the material at room temperature according to the mixture composition shown in Table 1. The composite mixture was molded and dried for three days to produce a transparent, dense, hard, stiff material, as shown in Figure 5. from the cured mixture. Drying time increases with increasing vinyl ester composition in the mixture.

#### 3.1. Mechanical Properties of Composite of Polyester and Rice Husk

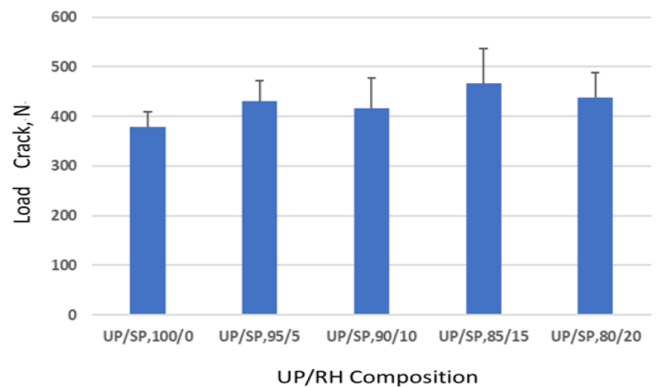
The main work in this research was to produce a composite of polyester (UP), rice husk particles (RH), and MKEP and MMA catalysts according to the percentage of each mixture and freeze the material at room temperature according to the mixture composition shown in Table 1. The composite mixture was molded and dried for three days to produce a transparent, dense, hard, stiff material, as shown in Figure 3. from the cured mixture. Drying time increases with increasing vinyl ester composition in the mixture.

The printed polymer mixture samples' stress intensity factor ( $K_{Ic}$ ) was tested against the crack load using a crack testing machine (CT). As shown in Figure 4, the CT specimens were prepared according to the standard dimensions of the crack test according to ASTM D5045 [23]. The velocity of the forced movement on the sample is 5

mm/min, and the magnitude of the load on the deformation that occurs is shown in Figure 5. The figure shows that the load-cracking curve obtained from the cracking test results of various percentages of vinyl ester mixtures on polyester polymers has been described in the picture above. All samples experienced different crack propagation depending on the percentage of the mixture. Before the maximum load, the curve tends to form a straight line, as illustrated in Figure 5 above. Table 2 shows the results of the mechanical properties of the UP/RH mixture. The figure shows that pure polyester produces relatively high tensile strength, but little deformation is formed, following the chemical structural network of polyester, which is cross-linked to each other so that it has little movement and is very stiff. The addition of rice husk composition to polyester increased the tensile strength of the UP/RH mixture by up to 15% by weight of RH, and plastic deformation occurred in the mixture according to the amount of ester rice husk added. The increase in crack strength is due to the RH of the cross-chain structure, in which rigidification bonds become slightly disturbed and cause movement of the polyester. The printed polymer mixture sample's intensity factor ( $K_{Ic}$ ) against the re-cracking test load (CT).



**Figure 6.** The typical loads versus crack opening displacement (COD) curves



**Figure 7.** Burden Causes Cracks from a mixture of Polyester and Rice Husk

**Table 4.** Critical Stress Intensity Factor of a mixture of polyester with rice husk

No	Material	Fracture toughness MPa.m <sup>0.5</sup>
1	UP/RH,100/0	0.667
2	UP/RH, 95/5	1.227
3	UP/RH,90/10	1.260
4	UP/RH,85/15	1.558
5	UP/RH,80/20	1.270

As shown in Figure 4, the CT specimens were prepared according to the standard dimensions of the crack test according to ASTM D5045 [12]. The force motion on the sample is 5 mm/min, and the magnitude of the load on the deformation that occurs is shown in Figure 6. The curve shown in the figure shows that the load-cracking curve obtained from testing various proportions of the RH mixture on the polymer has been described in Figure 6 above. All samples experienced different crack propagation depending on the proportion of the mixture. Before maximum load, the curve tends to form a straight line, as illustrated above [6, 9, 24]. Table 3 shows the results of the mechanical properties of the UP/RH mixture. The figure shows that pure polyester produces relatively high fracture strength, but minor deformation is formed following the chemical structural network of RH, which is cross-linked so that it has little movement and is very stiff. Adding the RH composition to the polyester increases the tensile strength of the UP/RH mixture up to 15% by weight of RH and the occurrence of plastic deformation in the mixture according to the amount of RH added. The

## References

- [1] N. Hiremath, S. Young, H. Ghossein, D. Penumadu, U. Vaidya, and M. Theodore, "Low cost textile-grade carbon-fiber epoxy composites for automotive and wind energy applications," *Composites Part B: Engineering*, vol. 198, p. 108156, 2020.
- [2] M. Davallo, H. Pasdar, and M. Mohseni, "Mechanical properties of unsaturated polyester resin," *International Journal of ChemTech Research*, vol. 2, no. 4, pp. 2113–2117, 2010.
- [3] H. Abral, R. Fajrul, M. Mahardika, D. Handayani, E. Sugiarti, A. N. Muslimin, and S. D. Rosanti, "Improving impact, tensile and thermal properties of thermoset unsaturated polyester via mixing with thermoset vinyl ester and methyl methacrylate," *Polymer Testing*, vol. 81, p. 106193, 2020.
- [4] D. Frómeta, S. Parareda, A. Lara, S. Molas, D. Casellas, P. Jonsén, and J. Calvo, "Identification of fracture toughness parameters to understand the fracture resistance of advanced high strength sheet steels," *Engineering Fracture Mechanics*, vol. 229, p. 106949, 2020.
- [5] N. Adnan, H. Abral, H. Dahlan, and E. Staria, "Identification of mechanical strength for mixture of thermoset polyester with thermoset vinyl ester due to bending load," *JMPM (Jurnal Material dan Proses Manufaktur)*, vol. 6, no. 1, pp. 19–25, 2022.
- [6] N. Adnand, R. Mutya, F. Ridwan, H. Abral, H. Dahlan, and E. Satria, "Pengaruh variasi persentase campuran polymer polyester dan vinyl ester terhadap kekuatan tegangan lentur," *Metal: Jurnal Sistem Mekanik dan Termal*, vol. 5, no. 2, pp. 126–131, 2021.
- [7] P. K. Naik, N. V. Londe, B. Yogesha, L. L. Naik, and K. Pradeep, "Mode I fracture characterization of banana fibre reinforced polymer composite," in *IOP Conference Series: Materials Science and Engineering*, vol. 376, p. 012041, IOP Publishing, 2018.
- [8] H. Ardhyanta, F. Puspawati, S. Wicaksono, A. Wibisono, B. Kurniawan, H. Ismail, A. Salsac, et al., "Mechanical and thermal properties of unsaturated polyester/vinyl ester blends cured at room temperature," in *IOP Conference Series: Materials*

increase in stress caused by the repair of the cross-chain, which is regulated, becomes slightly disturbed and causes movement of the polyester. The addition of the SP mixture to a content of 20% decreased the crack strength due to exceeding the saturation level of the rice husk particles, which no longer bonded well with the polyester matrix.

## 4. Conclusions

This study reports the successful determination of the proper composition of a polymer mixture made of unsaturated polyester matrix by adding rice husk fiber particles to make a tough composite and crack resistant. With this research, engineering in the engineering field is beneficial, especially in raw materials for vehicle components, tourist boat bodies, and fishing boats. This unsaturated polyester blend with 15% SP and 10% MMA has the highest critical stress intensity factor. With the highest performance, this material can withstand good fracture strength, making it good and useful for engineering applications. This research increased pure polyester materials' toughness and crack resistance by adding SP and MMA mixtures on pure unsaturated polyester base materials. The fracture toughness of  $K_{Ic}$  increased from 0.667 MPa.m<sup>0.5</sup> for pure polyester to  $K_{Ic} = 1.558$  MPa.m<sup>0.5</sup> with the addition of a mixture of 15% rice husk and 10% MMA by weight of the process polymer mixture. This achievement shows the highest fracture toughness value for the right mixture.

## Acknowledgments

This research was funded by the Research Grant Fund of the Department of Mechanical Engineering, Faculty of Engineering, Andalas University, Fiscal Year 2022.

- Science and Engineering*, vol. 202, p. 012088, IOP Publishing, 2017.
- [9] H. Abral, M. Hakim, R. Vadia, *et al.*, "The potential of rising husk fiber/native sago starch reinforced biocomposite to automotive component," in *IOP Conference Series: Materials Science and Engineering*, vol. 602, p. 012085, IOP Publishing, 2019.
- [10] A. T. Seyhan, M. Tanoğlu, and K. Schulte, "Tensile mechanical behavior and fracture toughness of mwcnt and dwcnt modified vinyl-ester/polyester hybrid nanocomposites produced by 3-roll milling," *Materials Science and Engineering: A*, vol. 523, no. 1-2, pp. 85–92, 2009.
- [11] Z. Yang, H. Peng, W. Wang, and T. Liu, "Crystallization behavior of poly( $\epsilon$ -caprolactone)/layered double hydroxide nanocomposites," *Journal of Applied Polymer Science*, vol. 116, no. 5, pp. 2658–2667, 2010.
- [12] H. Abral, R. Fajrul, M. Mahardika, D. Handayani, E. Sugiarti, A. N. Muslimin, and S. D. Rosanti, "Improving impact, tensile and thermal properties of thermoset unsaturated polyester via mixing with thermoset vinyl ester and methyl methacrylate," *Polymer Testing*, vol. 81, p. 106193, 2020.
- [13] A. Budiman and S. Sugiman, "Karakteristik sifat mekanik komposit serat bambu resin polyester tak jenuh dengan filler partikel sekam," *Dinamika Teknik Mesin*, vol. 6, no. 1, 2016.
- [14] H. Abral, R. Fajrul, M. Mahardika, D. Handayani, E. Sugiarti, A. N. Muslimin, and S. D. Rosanti, "Nanovoids in fracture surface of unsaturated polyester/vinyl ester blends resulting from disruption of the cross-linking of the polymer chain networks," *IOP Conference Series: Materials Science and Engineering*, vol. 1062, p. 012051, feb 2021.
- [15] S. Jeyanthi, J. J. Rani, *et al.*, "Improving mechanical properties by kenaf natural long fiber reinforced composite for automotive structures," *Journal of Applied Science and Engineering*, vol. 15, no. 3, pp. 275–280, 2012.
- [16] Q. Meng and T. Wang, "An improved crack-bridging model for rigid particle-polymer composites," *Engineering Fracture Mechanics*, vol. 211, pp. 291–302, 2019.
- [17] H. N. Dhakal and S. O. Ismail, "Unsaturated polyester resins: Blends, interpenetrating polymer networks, composites, and nanocomposites," in *Unsaturated Polyester Resins*, pp. 181–198, Elsevier, 2019.
- [18] K. Liu, S. He, Y. Qian, Q. An, A. Stein, and C. W. Macosko, "Nanoparticles in glass fiber-reinforced polyester composites: comparing toughening effects of modified graphene oxide and core-shell rubber," *Polymer Composites*, vol. 40, no. S2, pp. E1512–E1524, 2019.
- [19] N. Nusyirwan, "Metode pengering gabah aliran massa kontinu dengan wadah pengering horizontal dan pengaduk putar," *MECHANICAL*, vol. 6, no. 2, 2015.
- [20] S. Jaiswal, P. Dutta, S. Kumar, J. Koh, and S. Pandey, "Methyl methacrylate modified chitosan: Synthesis, characterization and application in drug and gene delivery," *Carbohydrate polymers*, vol. 211, pp. 109–117, 2019.
- [21] U. Ali, K. J. B. A. Karim, and N. A. Buang, "A review of the properties and applications of poly (methyl methacrylate)(pmma)," *Polymer Reviews*, vol. 55, no. 4, pp. 678–705, 2015.
- [22] S. Kalia and S. Vashistha, "Surface modification of sisal fibers (agave sisalana) using bacterial cellulase and methyl methacrylate," *Journal of Polymers and the Environment*, vol. 20, pp. 142–151, 2012.
- [23] A. International, "Standard test methods for plane-strain fracture toughness and strain energy release rate of plastic materials," *ASTM D5045-99*, 2007.
- [24] R. Masoodi, R. Hajjar, K. M. Pillai, A. Javadi, and R. Sabo, "An experimental study on crack propagation in green composites made from cellulose nanofibers and epoxy," in *Int. SAMPE Tech. Conf.*, no. 2011.