

Study Of Improved Crack Toughness Of Unsaturated Polymers With Rice Husk Fiber And Sago Flour As Strengthening Materials

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Abstract

The development of environmentally friendly composites from natural fibers is an absolute thing to do to replace non-degradable synthetic composites. Some of the weaknesses of natural fiber composites are low mechanical strength, ease of cracking, no moisture resistance, and high-temperature resistance. One of the things that has been done is to make a combination of synthetic materials as a matrix derived from unsaturated polyester reinforced with natural fibers from crushed rice husk particles and starch from sago flour which is used to reduce the percentage of synthetic materials to be able to form composites that are easily decomposed. From the research, it was found that the strength of crack resistance could be increased with a mixture of polyester reinforced with rice husk fiber and sago flour, obtaining an increase in crack resistance strength until the addition of rice husk with a percentage of 15%. While increasing the RH content above 15%, the crack strength value decreases due to the saturation of the RH molecules in the Unsaturated Polyester (UP) which is no longer bound to the UP molecules. The highest crack strength values occurred for the addition of the percentage of RH and SS to the UP material with the addition of 5% SS, which obtained a cracking force of 550 N while the strength of pure polyester was only 37 N. This shows that RH and SS materials can bond with UP molecules and some molecules of RH and SS can prevent cross-linking of polyester molecules. Meanwhile, the addition of SS up to 10% decreases the fracture force obtained, indicating that not all of the sago starch can bind to the polyester molecules.

Keywords:

Pure-polyester, rice-husk, sago-flour, crack-strength.

1 Introduction

One of the uses of polymer materials is to look for alternative materials to replace metal materials in construction components such as the construction of vehicles, ships, and aircraft[1][2]. This is done because the metal material has a high specific gravity so it requires a large driving energy when used for this purpose[1][3]. One of the advantages of polymeric materials is that they have low specific gravity, are easy to form, and have properties that are resistant to the environment. But not all construction components can be replaced by polymers[1][4][5]. The problem that occurs is that polymers have many weaknesses including low mechanical strength, easy to crack, and cannot withstand high temperatures[6][3][7]. The polymers that are widely used as matrices in composites are polyester and vinyl ester[8][9][10]. But polyester has a relatively cheap price compared to vinyl ester. Attempts to increase the mechanical

strength of polymers have been carried out, including reinforcing by adding synthetic fibers or natural fibers [11][12]. The materials mentioned have good mechanical properties when combined with fibers as reinforcement to form composite materials[13][14].

Another method for strengthening polymers is mixing two polymers that have almost similar properties, for example mixing polyester with vinyl ester[7][1][15]. 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[4][6][3]. In previous studies it has been explained that the addition of rice husk can increase the tensile strength of the polyester composite with rice husk and the addition of sago starch increases the plastic deformation of the composite[1][4][18]. One of the things studied in this research is how to increase the crack resistance of unsaturated polyester (UP) polymers by adding rice husk fiber reinforcement (SP) with different percentages and also adding sago flour (SS) as a filler. Previous studies have discussed improving the mechanical properties of polyester by adding carbon fiber, and natural fibers such as hemp fiber, banana fiber, and so on to form composite materials. The results reported are the mechanical properties of the composite material in the form of tensile loads, impact loads, and bending loads[16][7][8]. However, 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 [3][11][12][17]. So, this research will be aimed at increasing the crack resistance of polyester material by reinforcing it with rice husk fiber particles which are filled with sago flour at various percentages of a certain mixture.

2 Research Methods

This study used composites with fiber-oriented rice husk fiber reinforcement arranged randomly with continuous even distribution. The method used in this study is the hand layup method. The manufacturing process is carried out by pouring the resin by hand into a channel that flows in a fiber-filled mould, then applying pressure while leveling it with a roller and brush. This process is repeated until the required thickness of the composite is 12 mm thick[10][8][18][19].

2.1 Matrix

Polyester is a polymer that is 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[9][1][4][18]. Some of the properties of polyester, among others: polyester has fairly good tensile strength, resistance to stretch, chemicals, and mildew, has very good abrasion resistance, easy maintenance, and polyester has water-repellent properties, and dries quickly. The type of polyester used in this study was unsaturated polyester with Yukalac1560 BL-EX product (Fig. 1). The mechanical properties of polyester can be seen in Table 1.



Fig. 1. Polyester[18].

Sago flour is purchased from a raw materials store for cake makers where before being made the flour starch is filtered to obtain uniform dimensions. To get refined starch, flour is mixed with room temperature water and then stirred for 30 minutes, then filtered with water, and the solution is precipitated for 2 hours.

Table 1. Polyester physical properties

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 ³	1.0 – 1.45
T _g	°C	100 – 140
CTE	10 ⁻⁶ /°C	55 – 100
Cure shrinkage	%	5 – 12

2.2 Fiber

Rice husk is the outer part of the grain which is a by-product of rice milling (Fig. 2). The chemical composition of rice husk consists of relatively high levels of active silica (SiO₂), namely 94%-96% of husk ash, other components, CaO, MgO, Al₂O₃, and NaO₂. Therefore, the chemical composition of rice husk can be used as a source of raw materials for silica-based materials. The rice husk was crushed using a grinding machine and then filtered to obtain uniform dimensions with a particle size of 100 mesh and a specific gravity of 0.94 g/cm³. To bond well, an alkalization process was carried out in this process, and rice husks were soaked and precipitated with 4% NaOH for 8 hours at room temperature[18].



Fig. 2. Rice husk fiber[18].

2.3 Methyl Methacrylate (MMA)

Methyl methacrylate often referred to as MMA is a polymer material that has biocompatible properties. The advantage of adding MMA to an alloy is that it produces a material that is non-toxic, relatively cheap, easy to process, compatible, and can be used to process materials that have great crack resistance [16][12]. Mixing MMA with thermosetting resins can reduce the viscosity of the polymer mixture [13]. The addition of MMA here is expected to make the polyester network structure homogeneous [15][17].

2.4 Catalyst (MEKP)

The catalyst used is the Mepoxe catalyst produced by PT. Justus Kimiaraya (Fig. 3). The function of the catalyst is as a catalyst to accelerate the drying rate of polyester. The use of a catalyst is 4% for polyester alloys [20].



Fig. 3. Sago starch flour.

2.5 Design of Experiment

In this study, a crack test was carried out on polyester composite materials reinforced with rice husk fiber by giving grooves notches and initial cracks. The test material will be given a fracture load by carrying out vertical tensile loading on both sides which is increased gradually until the maximum load that the material can withstand until it experiences total cracking or maximum fracture toughness of the material, according to ASTM D5405 standard [19]. The magnitude of the stress distribution that occurs at the crack tip for the material given the initial defect is called the stress intensity factor.

2.6 Testing Equipment

Testing with two-sided vertical tensile loading with a withdrawal speed used is 4 mm/minute, because this speed is the standard speed for crack research according to previous studies, if it exceeds the speed of 4 mm/minute it will pose a risk of dynamic effects [19]. In this study, the composite material was made from a polyester matrix by mixing with sago starch and reinforcement from rice husk particles with a composition comparison that can be seen in Table 2 up to Table 4. Each mixture will be compared with the fracture toughness of all the percentages of the mixture made and will be compared to pure polyester without reinforcement. This section describes the steps for making polyester composite specimens of rice husk particles.

Table 2. Compositions of the crack test sample

Sample	UP	RH	UP/RH	SS
	Vol (%)	Vol (%)	Vol (%)	Vol (%)
RH0	100	0	100	0
RH5	95	5	100	0
RH10	90	10	100	0
RH15	85	15	100	0
RH 20	80	20	100	0

Table 3. Composition of the crack test sample

Sample	UP	RH	UP/RH	SS
	Vol (%)	Vol (%)	Vol (%)	Vol (%)
RH0	100	0	95	5
RH5	95	5	95	5
RH10	90	10	95	5
RH15	85	15	95	5
RH 20	80	20	95	5

Table 4. Composition of the crack test sample

Sample	UP	RH	UP/RH	SS
	Vol (%)	Vol (%)	Vol (%)	Vol (%)
RH0	100	0	90	10
RH5	95	5	90	10
RH10	90	10	90	10
RH15	85	15	90	10
RH 20	80	20	90	10

2.6.1 Hot Plat Magnetic

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: Daihan Scientific brand, model MS-H280-Pro, working temperature 25°C–280°C, rotation speed 0rpm–1500 rpm (Fig. 4).

The test specimen will be made according to the dimensions which refer to the standard dimensions following the ASTM D5405 standard as shown in Fig. 5[21].

This study used one type of fiber orientation type, in the form of rice husk particles and the matrix was rice husk mixed with sago flour starch whose fibers were arranged randomly with the mixture being stirred uniformly (Fig. 6). The manufacturing process is done by pouring a uniform mixture into the mold and then pressing it evenly with a roller or brush, this process is repeated until the required thickness is reached.



Fig. 4. Hot plate magnetic stirrer.



Fig. 5. Mold for crack test sample.



Fig. 6. Sample model test result mold crack test.

2.6.2 Crack Testing Machine

The crack testing machine (Fig. 7) 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 Fig.8. 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: brand COM-TEN testing machine 95T series 5K, capacity 5000 pounds, load cell model TSB0050, with monitor display system touch screen or com-touch screen.

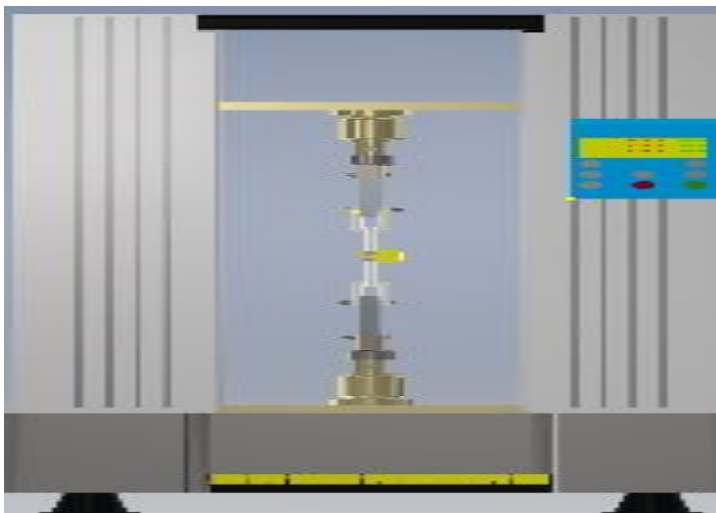


Fig. 7. Crack testing machine.

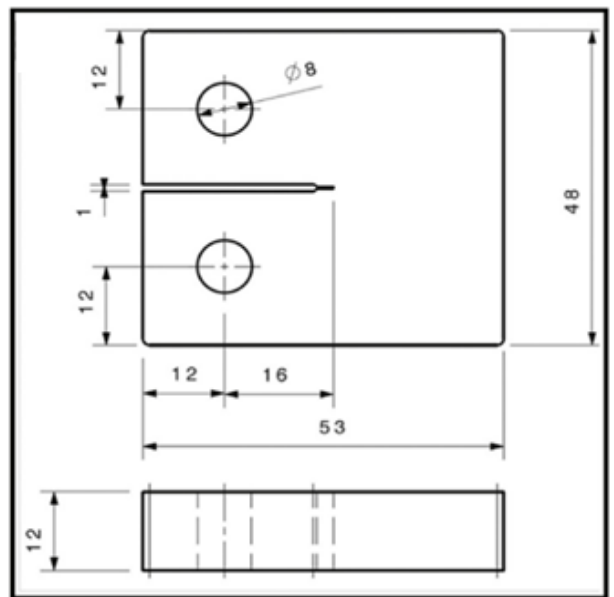


Fig. 8. Dimensions of crack test sample according to ASTM D5405[21].

Crack testing is a material test by providing withdrawal to the material so that the applied load will cause an initial crack to occur in the specimen. The resulting initial crack will cause cracks to propagate in the specimen until the material cannot withstand the given loading. The length of the crack obtained from a measuring instrument that is read on the machine display will be calculated using the mathematical equation in Eq. 1 and Eq. 2 [21][2]. In this study, the dimensions of the test sample referred to the ASTM D 5045 standard which is shown in Fig.8.

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

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

Description:

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)

Vol = Volume

3 Results and Discussion

In this section, the test results for each test will be described, namely, the crack test based on the ASTM D 5045 standard. The length of the crack is obtained from the measuring instrument that is read on the crack testing machine display and then the stress intensity factor value will be calculated using the mathematical equation in Eq. 1 and Eq. 2 [3].

Crack testing is a material test by giving a withdrawal to the material, the dimensions of the test sample refer to the ASTM D 5045 standard. The material for the crack test is a mixture of polyester according to rice husk and without added sago flour filler (SS = 0%) as shown in Table 2. The results obtained are the magnitude of the crack force in each fracture test specimen can be seen in Table 5 and the graph can be seen in Fig. 9 and Fig. 10. The test results show that the magnitude of the fracture force for each UP and RH mixture is greater than the fracture force of pure UP (without mixture) is only 312 N. Furthermore, the addition of rice husk fiber particles to UP can improve the crack resistance of the material. While the value of the maximum fracture force occurs in a polyester mixture of 85% with an RH of 15%, which is an average of 466 N. This shows that the addition of the

percentage of RH to the UP material can increase the crack resistance of the material. While increasing the RH content above 15%, the value of crack strength decreases due to the saturation of the RH molecules in the UP which is no longer bound to the UP molecules.

Table 5. Fracture force of the crack test sample

Sample	UP/RH Vol (%)	SS Vol (%)	Force[N]
RH0	100/0	0	312
RH5	95/5	0	380
RH10	90/10	0	431
RH15	85/15	0	466
RH 20	80/20	0	438

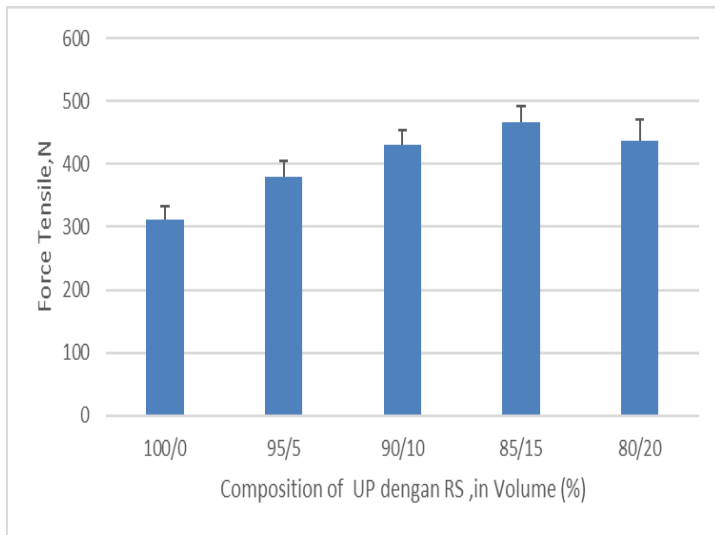


Fig. 9. Fracture force of crack testing machine UP/RS, at SS 0%.

the material's crack resistance. While increasing the RH content above 15% the value of crack strength decreases because it is due to the saturation of the RH molecules in the UP which is no longer bound to the UP molecules.

Table 6. Fracture force of the crack test sample at SS =5 %

Sample	UP/RH Vol (%)	SS Vol (%)	Force[N]
RH0	100/0	5	379
RH5	95/5	5	430
RH10	90/10	5	460
RH15	85/15	5	550
RH 20	80/20	5	445

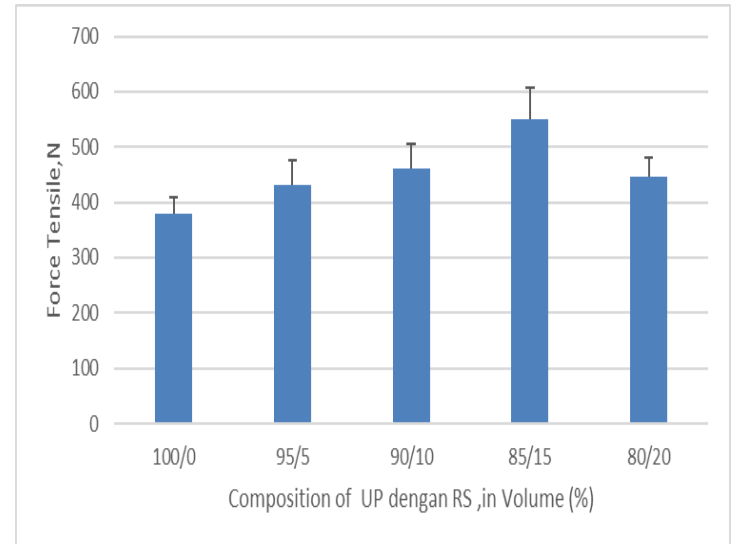


Fig. 11. Fracture force of crack testing machine UP/RS, at SS5%.

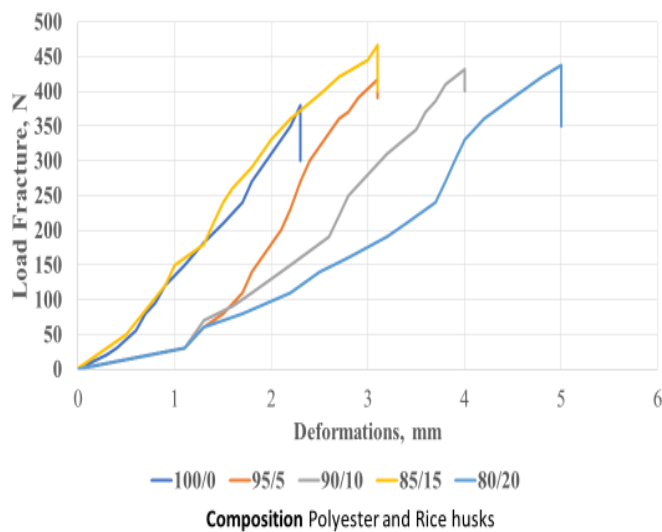


Fig. 10. Fracture force of crack testing machine UP/RS, at SS 0%.

Furthermore, crack testing on crack test specimens with a mixture of polyester and rice husk with the addition of sago flour as a filler (SS = 5%) is shown in Table 3. The dimensions of the test sample refer to the ASTM D 5045 standard. The results obtained are the magnitude of the fracture force on each fracture test specimen can be seen in Table 6 and the graphs can be seen in Fig. 11 and Fig. 12. The test results show the magnitude of the fracture force of each mixture of UP and RH is greater than the fracture force of pure UP (without mixture) of only 379 N. Furthermore, the addition of RH to UP can increase the crack resistance of the material. While the value of the maximum fracture force that occurs in the UP mixture of 85% while the RH is 15%, which is an average of 550 N. This shows that the addition of the percentage of RH and SS in the UP material can increase

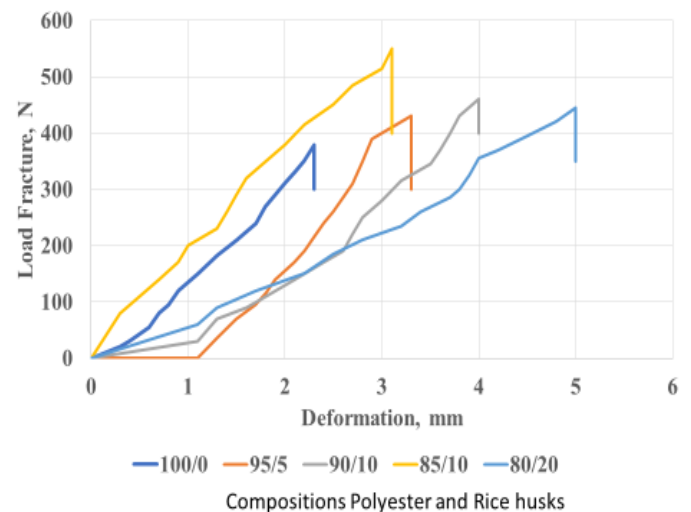


Fig. 12. Fracture force of crack testing machine UP/RS, at SS 5 %.

The dimensions of the test sample refer to the ASTM D 5045 standard. The results obtained are the magnitude of the fracture force on each fracture test specimen can be seen in Table 6 and the graphs can be seen in Fig. 11 and Fig. 12. The test results show the magnitude of the fracture force of each mixture of UP and RH is greater than the fracture force of pure UP (without mixture) of only 379 N. Furthermore, the addition of RH to UP can increase the crack resistance of the material. Furthermore, the crack test on the crack test specimen with a mixture of polyester and rice husk with the addition of sago starch (SS = 10%) is shown in Table 4. The dimensions of the test sample refer to the ASTM D 5045 standard. The results obtained are the magnitude of the crack force on each. The crack test specimens for each can be seen in Table 7 and the graphs can be seen in Fig. 13 and Fig. 14. The test results showed that the magnitude of the fracture force for each UP and

RH mixture was greater than the pure UP fracture force (without mixture) of only 329 N. Furthermore, the addition of RH to the UP can increase the crack resistance of the material. While the value of the maximum fracture force occurs in the UP mixture of 85% while the RH is 15%, which is an average of 454 N. This shows that the addition of the percentage of RH and SS in the UP material can increase the material's crack resistance. While increasing the RH content above 15% the value of crack strength decreases because it is due to the saturation of the RH molecules in the UP which is no longer bound to the UP molecules. For the addition of SS up to 10%, the cracking force decreases from the addition of 5% SS, this means saturation of the SS mixture as a filler in the mixture occurs.

Table 7. Fracture force of the crack test sample UP/RS, at SS 10%

Sample	UP/RH Vol (%)	SS Vol (%)	Force[N]
RH0	100/0	10	329
RH5	95/5	10	400
RH10	90/10	10	430
RH15	85/15	10	454
RH 20	80/20	10	400

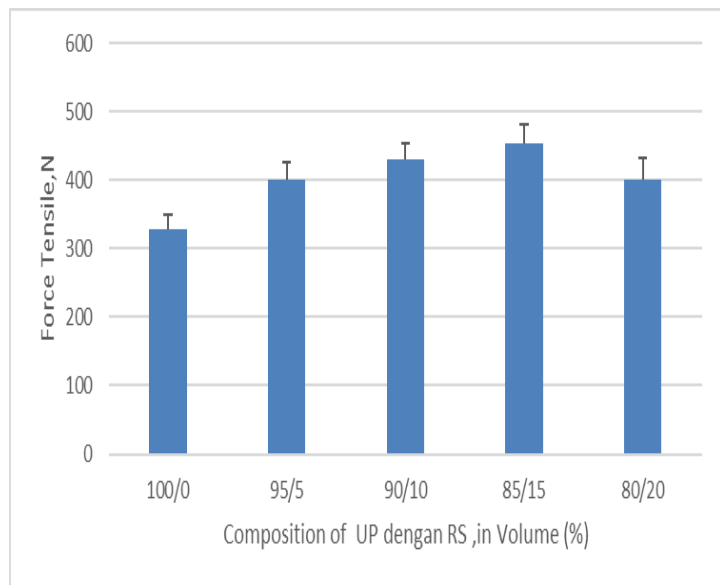


Fig. 13. Fracture force of crack testing machine UP/RS, at SS 10%.

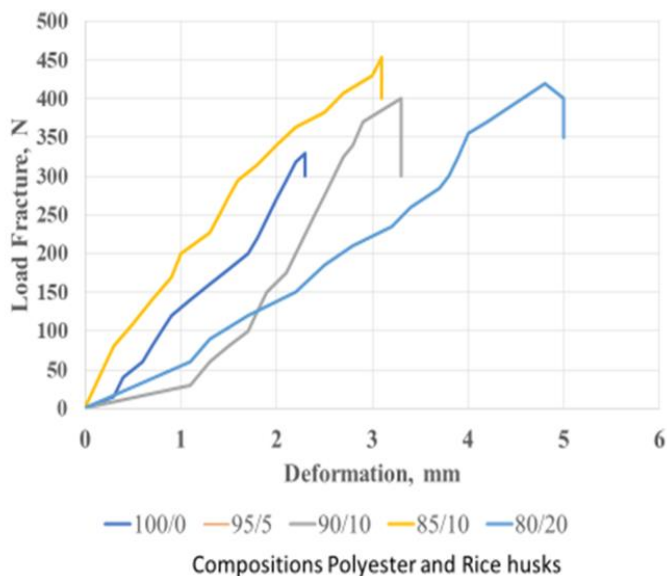


Fig. 14. Fracture force of crack testing machine UP/RS, at SS 10%.

4 Conclusion

The test results show that the magnitude of the fracture force of each mixture of polyester (UP) and rice husk (RH) can increase the magnitude of the fracture force of pure polyester (without mixture) to only 329 N. Furthermore, the addition of rice husk to polyester can increase the crack resistance of the material. The value of the maximum cracking force occurs in the UP mixture of 85% while the RH is 15%, that is, an average of 454 N increases by 137%. This shows that the addition of the percentage of RH and SS in the UP material can increase the crack resistance of the material. While increasing the RH content above 15% the value of crack strength decreases because it is due to the saturation of the RH molecules in the UP which are no longer bound to the UP molecules. The highest value of crack strength occurred for the addition of the percentage of RH and SS to the UP material with the addition of 5% SS, which obtained a cracking force of 550 N. This shows that RH and SS materials can bond with UP molecules and some molecules of RH and SS can prevent bonding crosslinked polyester molecules. Whereas for the addition of SS up to 10%, the fracture force obtained decreased, indicating that not all of the sago starch could bind to the polyester molecules. From the results of research, it is shown that the crack test based on the ASTM D 5045 standard for each sample shows a linear relationship with mechanical strength. The results were obtained by the tensile test at reference [18].

References

- [1] N. Adnan, H. Abrial, D. H, and E. Staria, "Identification of Mechanical Strength for Mixture of Thermoset Polyester with Thermoset Vinyl Ester due to Bending Load," *JMPM (Jurnal Mater. dan Proses Manufaktur)*, vol. 6, no. 1, pp. 19–25, 2022, doi: 10.18196/jmpm.v6i1.14450.
- [2] P. K. Naik, N. V. Londe, B. Yogesha, L. Laxmana Naik, and K. V. Pradeep, "Mode I Fracture Characterization of Banana Fibre Reinforced Polymer Composite," *IOP Conf. Ser. Mater. Sci. Eng.*, vol. 376, no. 1, 2018, doi: 10.1088/1757-899X/376/1/012041.
- [3] C. V. Opelt, G. M. Cândido, and M. C. Rezende, "Fractographic study of damage mechanisms in fiber reinforced polymer composites submitted to uniaxial compression," *Eng. Fail. Anal.*, vol. 92, no. June, pp. 520–527, 2018, doi: 10.1016/j.engfailanal.2018.06.009.
- [4] D. Frómata *et al.*, "Identification of fracture toughness parameters to understand the fracture resistance of advanced high strength sheet steels," *Eng. Fract. Mech.*, vol. 229, no. February, p. 106949, 2020, doi: 10.1016/j.engfracmech.2020.106949.
- [5] A. Mahyudin, S. Arief, H. Abrial, Emriadi, M. Muldarisnur, and M. P. Artika, "Mechanical properties and biodegradability of areca nut fiber-reinforced polymer blend composites," *Evergreen*, vol. 7, no. 3, pp. 366–372, 2020, doi: 10.5109/4068618.
- [6] M. T. Albdiry and B. F. Yousif, "Toughening of brittle polyester with functionalized halloysite nanocomposites," *Compos. Part B Eng.*, vol. 160, no. October 2018, pp. 94–109, 2019, doi: 10.1016/j.compositesb.2018.10.032.
- [7] H. Ardhyanta *et al.*, "Mechanical and Thermal Properties of Unsaturated Polyester/Vinyl Ester Blends Cured at Room Temperature," *IOP Conf. Ser. Mater. Sci. Eng.*, vol. 202, no. 1, 2017, doi: 10.1088/1757-899X/202/1/012088.
- [8] 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," *Polym. Compos.*, vol. 40, no. S2, pp. E1512–E1524, 2019, doi: 10.1002/pc.25065.

- [9] A. A. Betelie, Y. T. Megera, D. T. Redda, and A. Sinclair, "Experimental investigation of fracture toughness for treated sisal epoxy composite," *AIMS Mater. Sci.*, vol. 5, no. 1, pp. 93–104, 2018, doi: 10.3934/matensci.2018.1.93.
- [10] 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," *Compos. Part B Eng.*, vol. 198, no. May, p. 108156, 2020, doi: 10.1016/j.compositesb.2020.108156.
- [11] H. N. Dhakal and S. O. Ismail, *Unsaturated polyester resins: Blends, interpenetrating polymer networks, composites, and nanocomposites*. Elsevier Inc., 2019. doi: 10.1016/B978-0-12-816129-6.00008-9.
- [12] M. Santiam, R. Drainage, and W. Cascades, "The Applicability of Linear Elastic Fracture Mechanics to Compressive Damage of the Carbon Fiber Reinforced Plastic Matrix," 2019.
- [13] Z. Yang, H. Peng, W. Wang, and T. Liu, "Crystallization behavior of poly(ϵ -caprolactone)/layered double hydroxide nanocomposites," *J. Appl. Polym. Sci.*, vol. 116, no. 5, pp. 2658–2667, 2010, doi: 10.1002/app.
- [14] H. Abrial *et al.*, "Improving impact, tensile and thermal properties of thermoset unsaturated polyester via mixing with thermoset vinyl ester and methyl methacrylate," *Polym. Test.*, vol. 81, no. August 2019, p. 106193, 2020, doi: 10.1016/j.polymertesting.2019.106193.
- [15] M. T. Albdiry, B. F. Yousif, and H. Ku, "Fracture toughness and toughening mechanisms of unsaturated polyester-based clay nanocomposites," *13th Int. Conf. Fract. 2013, ICF 2013*, vol. 5, pp. 3446–3455, 2013.
- [16] C. Miao *et al.*, "Superior crack initiation and growth characteristics of cellulose nanopapers," *Cellulose*, vol. 27, no. 6, pp. 3181–3195, 2020, doi: 10.1007/s10570-020-03015-x.
- [17] B. B. Rath and J. J. Vittal, "Mechanical Bending and Modulation of Photoactuation Properties in a One-Dimensional Pb(II) Coordination Polymer," *Chem. Mater.*, vol. 33, no. 12, pp. 4621–4627, 2021, doi: 10.1021/acs.chemmater.1c01124.
- [18] Nusyirwan, H. Abrial, M. Hakim, and R. Vadia, "The potential of rising husk fiber/native sago starch reinforced biocomposite to automotive component," *IOP Conf. Ser. Mater. Sci. Eng.*, vol. 602, no. 1, 2019, doi: 10.1088/1757-899X/602/1/012085.
- [19] H. Abrial *et al.*, "Nanovoids in fracture surface of unsaturated polyester/vinyl ester blends resulting from disruption of the cross-linking of the polymer chain networks," *IOP Conf. Ser. Mater. Sci. Eng.*, vol. 1062, no. 1, 2021, doi: 10.1088/1757-899X/1062/1/012051.
- [20] and D. H. H. Abrial, R. Fajrul, M. Mahardika, "Improving impact, tensile and thermal properties of thermoset unsaturated polyester via mixing with methyl methacrylate and thermoset vinyl ester," 2019.
- [21] a Standard, "Standard Test Methods for Plane-Strain Fracture Toughness and Strain Energy Release Rate of Plastic Materials," *Annu. B. ASTM Stand.*, vol. 99, no. Reapproved, pp. 1–9, 1996, doi: 10.1520/D5045-99R07E01.2.
- [22] M. Hughes, C. A. S. Hill, and J. R. B. Hague, "The fracture toughness of bast fibre reinforced polyester composites: Part 1 Evaluation and analysis," *J. Mater. Sci.*, vol. 37, no. 21, pp. 4669–4676, 2002, doi: 10.1023/A:1020621020862.