International Journal on Interactive Design and Manufacturing (IJIDeM) (2023) 17:765–773
https://doi.org/10.1007/s12008-022-00980-4

ORIGINAL PAPER

Manufacturing and delamination factor optimization of cellulosic paper/epoxy composites towards proper design for sustainability

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Received: 8 May 2022 / Accepted: 30 June 2022 / Published online: 2 August 2022

Abstract

Determining an appropriate cellulosic based composite type for sustainable products is still challenging for both designers and the industry. Also, enhancing efficient high-quality joining techniques is still motivating the wide applications of composite materials as structural components. This work introduces a potential scheme of recycling accumulated waste copying papers into functional products via paper- epoxy reinforced composite material. Various cellulosic based copying papers were utilized as 15, 25 and 50 papers to fabricate paper/epoxy plate materials. The mechanical properties of the fabricated composites were investigated regarding the tensile strength, tensile modulus and elongation to break. The delamination factors for the drilling operation of the composites were studied and optimized to eliminate the exit (push-out delamination). The operational condition schemes were determined to reduce the delamination factor of the produced holes. Results have revealed that the 50 paper/epoxy composite shows the highest strength with a value of 86 MPa. However, the 25 paper/epoxy case was the best regarding the modulus of elasticity property. The worst delamination factor at 15 paper /epoxy composite occurred at both 6 mm drill diameter at 475 rpm spindle speed and 8 mm drill diameter at 850 rpm spindle speed. The 15 paper/epoxy delamination factor was decreased with increasing the spindle speed at 6 mm drill. The best delamination factor for the 25 paper/epoxy case was obtained with both 6 and 10 mm drill diameter at 1500 rpm spindle speed. However, the 50 paper/epoxy case was capable of reducing the delamination factor for most of the operational conditions.

Keywords Green composites · Paper recycling · Drilling · Sustainability · Failure · Green products

1 Introduction

Technological and commercial interest in green materials and bio-based composites was mainly derived from the fact that their properties are more desirable than that of traditional materials. The implementation of bio-based composite materials is increasing in various engineering sectors including biomedical, pharmaceutical, automotive, defense, aeronautical, construction, energy harvesting, energy storage, electronic and sensors as well as others [1–4]. This is in fact due to their attractive features and characteristics that can expand both sustainable deign possibilities and cleaner production theme. Such desired characteristics of bio-based composites include their excellent specific mechanical properties like strength and stiffness, being light in weight, can be easily manufactured with high level of design specifications, in addition to being ecofriendly, made from renewable sources as well as others [2, 5, 6]. Moreover, better appearance and controlled surface smoothness are possible with certain composite materials. However, they have some limitations like that of being anisotropic, which sophisticates their proper deigns in particular fields. Design for sustainability is currently a growing theme that considers green materials, recyclability and processes to provide essential knowledge to develop sustainable green products [7]. It offers more sustainable green products with several advantages including being environmentally friendly, renewable low-energy consumption, and low cost. To design for sustainability, several activities have to be considered including material selection via several techniques like the optimization, the analytical hierarchy process, manufacturability,

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assemblability, maintainability, recyclability, cost and safety[8].

On the other hand, the need of proper utilization of the available sources as well as waste is highly emphasized worldwide [9–15]. Cellulosic based papers that are used for copying purposes and available in almost every daily activity are usually accumulated. However, it is a valuable biomass product that can be properly recycled in making proper composite materials for several potential applications [16–19]. Such applications required proper mechanical properties as well as joining and assembly where drilling is essential in attaining such objective. The machining of composite materials is also a rising problem in various fields [20–29]. Particularly, the drilling of bio-based composites is not an easy-going task as it is difficult to be controlled and usually leads to delamination in the produced hole due to the layer-structure of the material, which can affect the structural integrity, mechanical properties, and its reliability during operation.

Delamination is of paramount importance for drilling composite materials as it commonly leads to damage in the composite. It is usually occurred at both exit (push-out delamination) and entrance (peel-up delamination) of drilled materials. It was experimentally proven that delamination occurs at the exit of the drilled hole is more severe [30–33]. Delamination can be reduced by proper selection of the drilling parameters such as feed, drill diameter, and spindle speed [34]. The delamination factor is calculated as the ratio between the maximum diameter of the delaminated area around the hole to the hole diameter by the following equation:

$Fd = D_{max} / D$ (1).

For comparison and evaluation purposes, some factors were developed in the literature as an index of delamination [34]. Figure 1 represents schematic illustrations of the delamination of composites.

Several researchers have studied the drilling process of composite materials; however, a few studies are available for the natural fiber composite materials [35–39] and there was no study that presents cellulosic paper/ epoxy composites during drilling. [40] studied the effect of speed and feed on the delamination behavior of glass fiber reinforced polymer composites (GFRP) experimentally. Taguchi method and analysis of variance were utilized to optimize the drilling conditions. It was reported that increasing the spindle speed and reducing the feed rate can reduce the delamination. However, there were limitations on speed and feed rates to reduce the delamination of the produced holes. Moreover, [41] have estimated the critical thrust force and delamination at the exit of the drilled hole by developing an analytical model. They tried to determine the optimal cutting conditions to eliminate the delamination during drilling. [42] have made drilling for Aramid Fiber Reinforced Plastics (AFRPs) to determine the optimal combination of drilling parameters to minimize thrust and torque force to reduce the delamination. Taguchi L54 layout was adopted for optimization. The purpose was also to evaluate the influence of process parameters.

It was reported that drill diameter influences the torque force on the drill bit. Also, a recommendation was reported as the use of high speed and low feed combination with proper drill point angles of 90°–118° can reduce the delamination in AFRP composites. [43] proposed a new method to measure the delamination factor by using image processing to describe the drilling-induced damage based on digital radiography. It was based on reducing the affected area to a quantified damaged area ratio. It was reported that to minimize the quantified damage for the most irregular delamination shapes to a single, unambiguous value makes an explicit delamination factor for measuring drilling-induced damage. [44] studied the drilling of woven carbon fiber reinforced polymeric (CFRP) laminates with four different tool geometries. In addition, the analysis for the effect of the cutting parameters on the cutting forces and delamination damage was performed. [45] used boron nitride nanoparticle in a carbon fiber epoxy nanocomposite to enhance its mechanical and thermal properties. Drilling was performed on the composite showing that the addition of boron nitride nanoparticles assisted in reducing the delamination factor of the epoxy-based composite. On the other hand, few studies have consider the drilling of natural fiber composites as [46] considered the effects of drill diameter, spindle speed, and feed rate on all of force, torque and tool wear in drilling coir polyester composites. The optimal were found using 8 mm drill diameter at 600 rpm spindle speed with feed rate of 0.3 mm/rev. [47] have examined the drilling of woven jute fabric-reinforced polypropylene composites. Different cutting parameters were used. Drill geometry was found significant on the thrust force values and delamination factor. [48] has examined the role of cutting parameters including drill diameters on the delamination of bamboo fiber/polyester composites. The minimum delamination factor was gained with 4 mm drill diameter at 18 mm/min feed rate and 500 rpm spindle speed. Other researches on hybrid green fibers and synthetic ones were reported [49].

It was found from the literature that all studies considered natural fiber polymeric composites in drilling have focused on some key parameters including spindle speed, drilling force, drill diameter and /or geometry, feed rate and the torque. The best combinations of feed, speed and diameters that reduce the force, torque and /or delamination were recommended to be utilized to reduce the delamination in the drilled holes. Other recent works that considered synthetic fiber composites have focused on the prediction

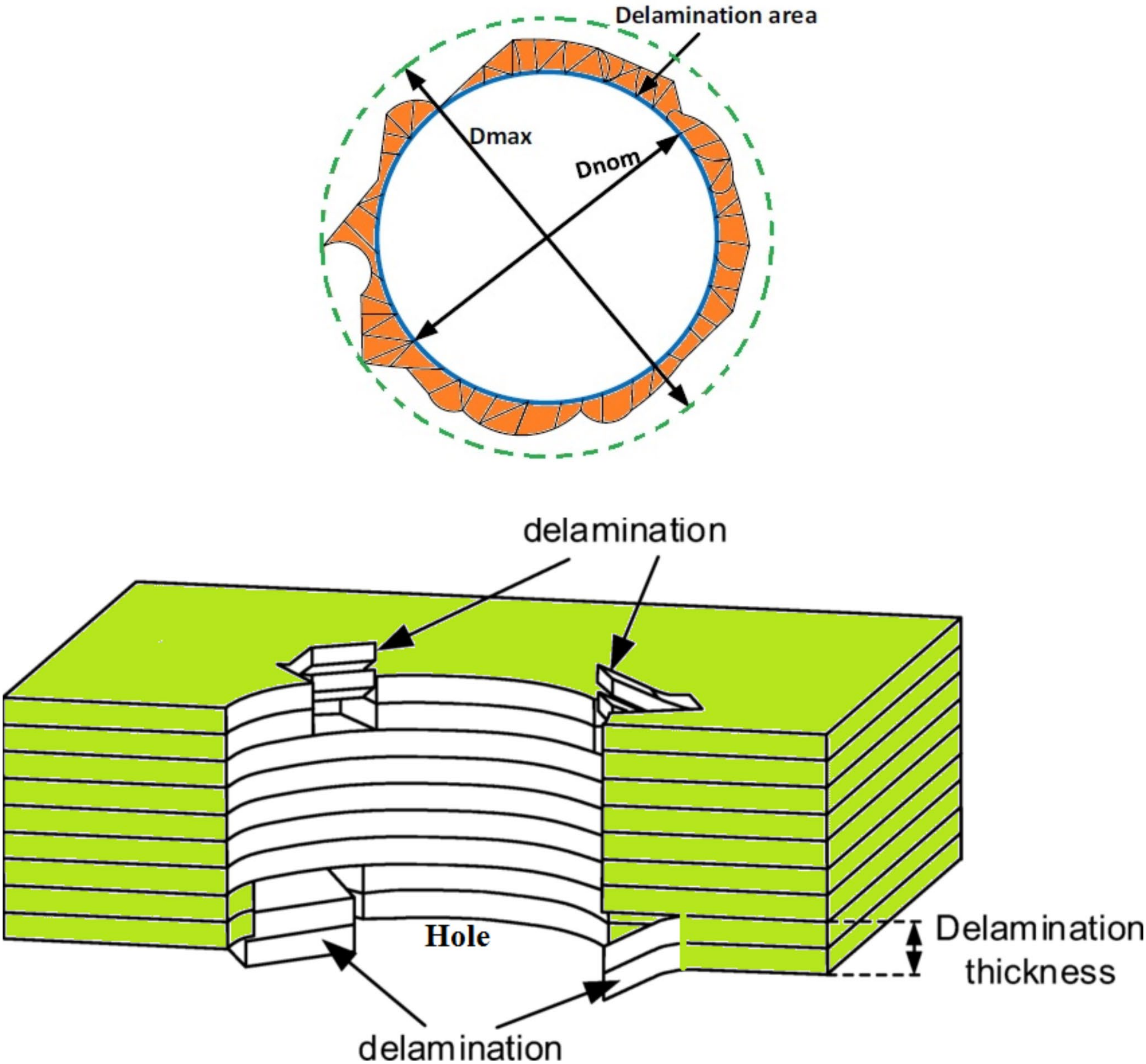

Fig. 1 Schematic representations of delamination in a drilled hole

of delamination during drilling. However a few studies are available for the natural fiber composite materials [35–37] and there was no study presents cellulosic paper/ epoxy composites during drilling.

Consequently, this work aims to present a novel cellulosic paper /epoxy composites and to investigate their mechanical properties to enhance their reliable use in various applications. Moreover, the work studies the cellulosic paper /epoxy composite behavior during drilling to optimize the best reinforcement conditions that are capable of reducing the delamination of the produced holes. It also aimed to determine the proper process parameter combinations to present a potential recycling approach of the available accumulated waste papers and to enhance their reliability for various potential applications.

2 Materials and methods

2.1 Materials

Epoxy resin was prepared by thoroughly mixing the NORYL 7310 resin epoxy and hardener. It was purchased from SABIC company, Saudi Arabia. The yield tensile stress according to ASTM D638 was 54 MPa, and tensile stress at break was 45 MPa. The tensile modulus was 2400 MPa.

Table 1 The cutting condition combinations

Experiment #	Number of papers	Spindle speed (rpm)	Drill diameter (mm)
1	15	475	6
2	15	850	6
3	15	1500	6
4	15	475	8
5	15	850	8
6	15	1500	8
7	15	475	10
8	15	850	10
9	15	1500	10
10	25	475	6
11	25	850	6
12	25	1500	6
13	25	475	8
14	25	850	8
15	25	1500	8
16	25	475	10
17	25	850	10
18	25	1500	10
19	50	475	6
20	50	850	6
21	50	1500	6
22	50	475	8
23	50	850	8
24	50	1500	8
25	50	475	10
26	50	850	10
27	50	1500	10

The melt flow rate according to ASTM D1238 was 11 g/10 min[50]. Different numbers of copy A4 papers were utilized as 15, 25, and 50 to design low, medium and high thickness of composites.

2.2 Methods

The utilization of the proper manufacturing process as well as composite constituents can affect the overall performance of the green product. Here, the design for sustainability was enhanced via proper selection and recycling of the accumulated waste copying papers as a low-cost reinforcement to produce new materials with desired properties capable of being utilized for various industrial applications including assembly processes. The epoxy resins with hardener were infused into the papers by vacuum-assisted resin infusion molding process. The composite plates were then allowed to cure at room temperature for 24 h and then post-cured in an oven for 2 h at 110 ◦C. Samples were then prepared for the tensile test to evaluate the mechanical properties of the epoxy resin and paper plates. The test was performed according to the ASTM D 3039/D 3039 M standard using a universal testing machine at a loading rate of 5 mm/min [51, 52]. The drilling operation was performed using the high-speed steel drill bits of 118° tip angle with different diameters of 6 mm, 8 and 10 mm as such diameters are usually utilized for assembly purposes. Three different spindle speeds were utilized as 475 rpm, 850, rpm, and 1500 rpm to investigate the effect of spindle speed on the delamination of the drilled holes as these speed levels are commonly used for various commercial purposes. Drilling was performed dry without coolant at a feed rate of 50 mm/min as it was recommended in several studies for polymeric based composites. The cutting conditions are shown in Table 1. Steel support was used under the composite parts to reduce the bending of the workpieces to eliminate the influence of this structural parameter during the study. The operation of drilling was performed in a single step. The determination of the delamination factor in the case of drilling was based upon the ratio between the actual hole diameter (D_{max}) observed by optical microscope and the diameter (D) of the drill bit used. The delamination factor was calculated as the ratio between the maximum diameter of the delaminated area around the hole to the hole diameter.

3 Results and discussion

Mechanical characterization of the fabricated paper/epoxy composites are investigated and discussed here. Whole diameters and delamination factors with the drilling operational conditions and reinforcement environment are also discussed to determine the behavior trends of the produced holes and the optimal operational condition to minimize the delamination during drilling.

3.1 Mechanical properties

The ultimate tensile strength and breaking mechanical properties of the fabricated cellulosic paper/epoxy composites as well as the neat epoxy are presented in Fig. 2. It can be demonstrated that the ultimate tensile strength was enhanced with using higher number of papers in the composite. The tensile strength for the 15 paper /epoxy reached 76.6 MPa comparable to the 62.7 MPa of the neat epoxy. The highest value of this property was obtained at 50 paper/epoxy composite with about 37% enhancement of the epoxy resin utilized. This was due to the reinforcement of the cellulosic papers that enhanced the stress transfer efficiency of the composites with increasing the fillers inside the epoxy resin [53–57]. However, the strain at break property was reduced compared to the epoxy resin itself as the reinforcement enhanced the brittleness behavior of the composites. This is due to the fact that the cellulosic paper elongation to

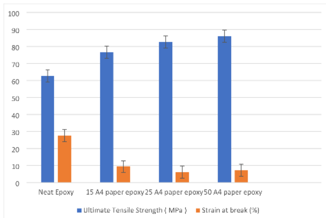

Fig. 2 Tensile strength and strain at break properties of the composites

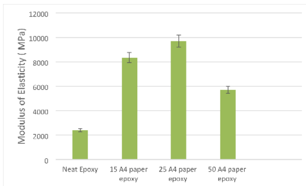

Fig. 3 Modulus of elasticity property of the composites

break was much lower than that of the epoxy resin. It can be seen that the elongation to break (%) for the epoxy was 27.6 while the 15 paper /epoxy composite had only 9.3, and the 25 paper/ epoxy composite had 6.1 then increased a bit in the 50 paper/epoxy case to be 7.1%. This was due the fact that the 50 paper/ epoxy composite had much higher epoxy comparable to the other reinforcement conditions due to the higher number of papers that required to be wetted with the resin.

On the other hand, the modulus of elasticity property of the composites is demonstrated in Fig. 3. It can be seen that all reinforcement conditions enhanced this mechanical property. Its maximum value was obtained at 25 paper/ epoxy combination with a value of 9703.512 MP comparable to 2400 MPa for the neat epoxy resin. This means that utilizing the cellulosic papers in epoxy had increased its modulus of elasticity property more than 300% demonstrating its effectiveness for recycling waste papers into potential new materials for various applications. It can also be noted here that the modulus of elasticity of the composite was reduced at 50 paper /epoxy case due to the higher number of papers leading to increase the possibility of improper adhesion between layers inside the composites comparable to other reinforcement conditions.

3.2 Drilling hole and delamination factor analysis

The diameters of the produced holes with 6 mm diameter drill at various spindle speeds with reinforcement conditions are demonstrated in Fig. 4 and 5. It can be shown that all diameters were more than 6 mm. The diameters of the produced holes were increased with increasing speeds at all reinforcement conditions except that at 50 papers/ epoxy condition where spindle speed of 850 rpm and 1500 rpm were almost close. It is obvious that at speed of 475 rpm, the

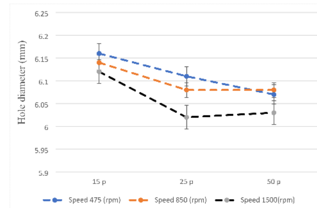

Fig. 4 Hole diameter with speed and reinforcement at 6 mm drill diameter

Table 2 Summary of the hole diameters of the produced holes

15 p		Φ 6 mm	Φ 8 mm	Φ 10 mm
	Speed 475 (rpm)	**6.16**	**8.13**	**10.11**
	Speed 850 (rpm)	**6.14**	**8.21**	**10.1**
	Speed 1500 (rpm)	**6.12**	**8.16**	**10.05**
25 p		Φ 6 mm	Φ 8 mm	Φ 10 mm
	Speed 475 (rpm)	**6.11**	**8.1**	**10.1**
	Speed 850 (rpm)	**6.08**	**8.15**	**10.07**
	Speed 1500 (rpm)	**6.02**	**8.09**	**10.03**
50 p		Φ 6 mm	Φ 8 mm	Φ 10 mm
	Speed 475 (rpm)	**6.07**	**8.06**	**10.08**
	Speed 850 (rpm)	**6.08**	**8.04**	**10.05**
	Speed 1500 (rpm)	**6.03**	**8.02**	**10.02**

drilled diameters were reduced with enhancing reinforcement conditions as higher number of papers develops better resistance to spindle chattering and reduces the deviation in drilled diameters. However, at higher spindle speeds all of 25 and 50 paper/ epoxy composites had almost consistent hole diameters as at higher spindle speeds, chattering is reduced. Thus, the reinforcement is capable of resisting such chattering leading to almost consistent whole diameter. A summary of the average hole diameters of the produced holes are demonstrated in Table 2.

Moreover, the diameters of the produced holes with 8 mm diameter drill at various spindle speeds with reinforcement conditions are demonstrated in Fig. 4. It can be illustrated that at all levels of spindle speeds, hole diameters were reduced with increasing the number of utilized papers inside the epoxy resin due to the higher resistance in the drill chattering. It can be noticed that at speed of 850 rpm, hole diameters were higher than other speeds except of 50 paper/ epoxy composite as it was capable of reducing the drill chattering leading to reduce the drill diameter. In addition, the diameters of the produced holes with 10 mm diameter drill are shown in Fig. 6 with various spindle speeds and reinforcement conditions. The drill diameters of the produced holes were shown to be decreased with increasing the number of utilized papers at all spindle speeds when utilized 10 mm diameter drill. The larger diameters of the produced holes were occurred at speed of 475 rpm and the smaller diameters were achieved at speed of 1500 rpm. It can also be shown that speed of 850 rpm did not produced the worst

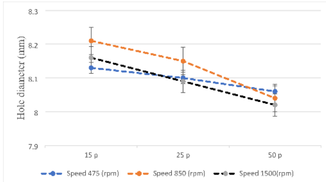

Fig. 5 Hole diameter with speed and reinforcement at 8 mm drill diameter

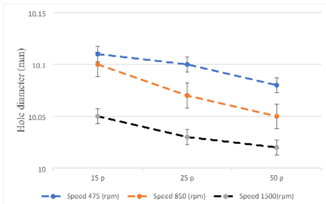

Fig. 6 Hole diameter with speed and reinforcement at 10 mm drill diameter

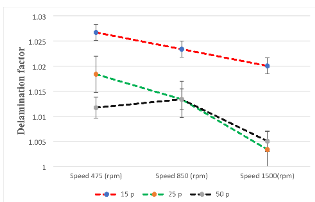

Fig. 7 Delamination factor with speed and reinforcement at 6 mm drill diameter

hole diameters as the drill diameter of 10 mm was capable of reducing the chattering during drilling leading to reduce the hole diameters at all reinforcement conditions.

The delamination factor trends of the produced holes inside the composites with 6 mm drill diameter are demonstrated in Fig. 7 with various spindle speeds and reinforcement conditions. It can be shown that the delamination factor was reduced with increasing spindle speed at both 15 and 25 paper/epoxy composites, but slightly enhanced in case of 50 paper/ epoxy at 850 rpm speed due to the higher chattering occurred. Drilled diameters were found to be reduced with enhancing the reinforcement conditions as the higher number of papers is capable of developing better resistance to spindle chattering. However, at higher spindle speeds all of 25 and 50 paper/ epoxy composites had almost small delamination factors due to the fact that at higher spindle speed, chattering is reduced and composites were found capable of resisting such chattering.

The delamination factor at 8 mm drill with various speeds and reinforcement conditions are illustrated in Fig. 8 and 9. It is shown that the delamination factor was reduced with increasing the number of papers in the matrix at all spindle speeds and was the worst at speed of 850 rpm at both 15 and 25 paper/ epoxy cases due to the higher chattering of the drill. However, the 50paper/epoxy was capable of reducing

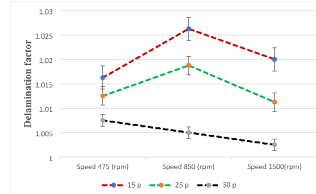

Fig. 8 Delamination factor with speed and reinforcement at 8 mm drill diameter

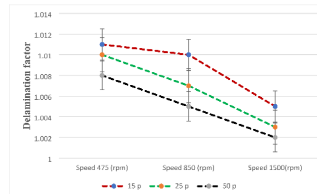

Fig. 9 Delamination factor with speed and reinforcement at 10 mm drill diameter

the delamination factor at this particular speed due to its capability to reduce such drill chattering. Figure 10 presents the delamination factor with speed and reinforcement at 10 mm drill diameter. It can be seen that the delamination factor was reduced with increasing papers inside the matrix at all spindle speeds. It is worthy notice here that at 10 mm drill the reduction in the delamination factor was reduced dramatically with increasing spindle speed as both higher drill diameter and spindle speed can reduce the chattering during the process.

The overall delamination factor at various spindle speeds, drill diameters, and paper content are demonstrated in Fig. 10. It is noticeable that the highest values of the delamination factor at 15 paper /epoxy composite were occurred at both 6 mm drill diameter at 475 rpm spindle speed and 8 mm drill diameter at 850 rpm spindle speed, while the best delamination factor was obtained with 10 mm diameter at 1500 rpm spindle speed. However, the best delamination factor for the 25 paper/epoxy case was obtained with both 6 and 10 mm drill diameter at 1500 rpm spindle speed. In addition, the 50 paper/ epoxy case was capable of reducing the delamination factor for most of the operational conditions compared to other composite cases. The best value was occurred at 1500 rpm spindle speed with 10 mm drill diameter. On the other hand, comparison for the trends of delamination factors of the fabricated composites is demonstrated in Fig. 11. The 15 paper/ epoxy delamination factor was decreased with increasing the spindle speed at 6 mm drill and continued to reduce at 450 rpm speed with 8 mm drill and increased in case of 8 mm drill at 850 rpm speed to reach it maximum value, then reduced for the rest of operational conditions. Similar trend was obtained for the case of 25 paper/ epoxy composite with 6 mm drill to reach its best value at 1500 rpm spindle speed then increased to its

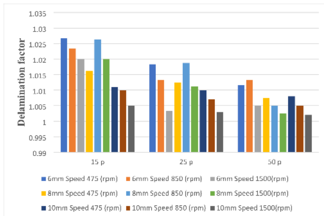

Fig. 10 Overall delamination factors at various operating conditions and reinforcement

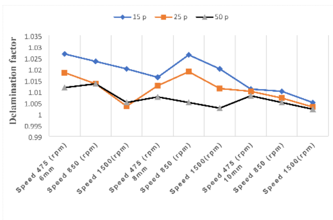

Fig. 11 Trend comparisons of delamination factors of the composites

maximum value at 850 rpm spindle speed with 8 mm drill diameter then reduced after that. However, the 50 paper / epoxy case was more consistent in the delamination factor comparable to the other cases. Its worst value was at 850 rpm speed with 6 mm drill diameter, but it was capable of maintaining proper delamination factors in all cases comparable to the other fabricated composites.

4 Conclusions

Green materials are of paramount importance for greener products. Utilizing the available cellulosic waste to produce functional materials is still limited to its possibility to be joined and assembled. The available waste of copying papers can be usefully recycled into polymeric based composites. This would enhance the design for sustainability theme. The mechanical properties of the prepared composites were found potential. The delamination factor for the 50 paper/epoxy composite was almost small at high spindle speeds. The drill diameter was found critical for reducing the delamination factor of the produced holes as higher drill diameter was capable of reducing the chattering during drilling process. The highest delamination values were found at 15 paper /epoxy composite at both 6 mm drill diameter at 475 rpm spindle speed and 8 mm drill diameter at 850 rpm spindle speed. The best delamination factor for the 25 paper/epoxy case was obtained with both 6 and 10 mm drill diameter at 1500 rpm spindle speed. However, the 50 paper /epoxy case was more consistent in the delamination factor compared to the other cases. Its worst value was at 850 rpm speed with 6 mm drill diameter, but it was capable of maintaining proper delamination factors in all cases compared to the other fabricated composites.

Acknowledgements The anonymous reviewers are thankful for their supportive comments and suggestions that have enriched this work during the peer review process.

Funding No funding was received to perform this work.

Data Availability Not applicable.

Data Availability The raw/processed data required to reproduce these findings cannot be shared at this time as the data also forms part of an ongoing study.

Declarations

Ethics approval and consent to participate Not applicable.

Consent for publication Not applicable.

Competing interests The authors declare that they have no competing interests.

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