

# The Effect of Curative Concentration on Thermal and Mechanical Properties of Flexible Epoxy Coated Jute Fabric Reinforced Polyamide 6 Composites

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## ABSTRACT

Many researchers have shown interest in the reinforcement of commodity thermoplastic with natural fibers. However, the drawback of natural fibers is their low thermal processing temperatures, that border around 200°C. In this investigation, we tried to improve the thermal stability of natural fibers with the use of flexible epoxy surface coating that could facilitate processing with engineering thermoplastics. Jute fabric and Polyamide 6 (PA6) composites were prepared by compression molding. The thermal decomposition characteristics of the jute fabric were evaluated by using thermo gravimetric analysis (TGA). Mechanical analysis was conducted to evaluate tensile test and three point bending test of composite. It was found that thermal degradation resistance of jute fabric was improved by coating with flexible epoxy resin. Moreover, the flexural modulus improved with increasing curative concentration. The interfacial interaction between the epoxy and PA6 was clearly indicated by the photo micrographs of the polished cross sections of the coated and uncoated jute fabric/PA6 composites.

**Keywords:** Natural Fiber (Jute); Polyamine 6; Flexible Epoxy; Composites; Thermal Degradation; Mechanical Properties

## 1. Introduction

In recent years, natural fibers such as sisal, hemp, abaca, ramie, and jute have attracted the attention of researchers because of the advantages that these fibers provide over conventional reinforcement materials [1-5]. Natural fiber exhibit many advanced properties such as low density, low cost, high specific properties, recyclability and biodegradability [6,7]. Thus, natural fiber reinforced composites have been show to yield properties suitable for low stress application such as automotive, some part of the air plan.

However, the disadvantages of natural fibers in thermoplastics are incompatibility between the hydrophilic natural fiber and hydrophobic polymer [8-12]. Some researchers have reported improvements in the mechanical properties when compatibilizer used on the fiber were chemically modified prior to mixing [13-16]. Rashed *et al.* studied about jute fiber reinforced polypropylene. The tensile strength was increased after alkalized treated jute fibers [17]. Cao *et al.* reported 13% improvement in tensile strength, 14% in flexural strength and 30% in impact

strength after fibers were treated with 1% NaOH solution [18].

Another drawback seems to be the low thermal stability of natural fibers, where the first degradation of natural fiber occurs at temperatures above 200°C [19]. Natural fibers are therefore typically used with commodity thermoplastic matrix such as polypropylene (PP), polyethylene (PE), poly vinyl chloride (PVC) and polystyrene (PS) with melting points that are lower than the degradation temperature of the natural fibers [20].

S. C. Jana and A. Prieto [21,22] study about the development of wood flour composites with high temperature thermoplastic polymers as the matrix resin. Wood flour was miscible blend with curing agent for improving thermal resistance of it. In the addition, poly (phenylene ether) (PPE) was miscible blend with epoxy to reduce the melting temperature of the latter prior to incorporation of wood flour through twin-screw extrusion. The composite exhibited an improvement in mechanical performance since low processing temperatures were used to prevent extensive degradation to the wood flour whereas the matrix itself exhibited high strength and stiffness. Thermo-

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setting polymers are among the most important materials in many diverse industries and are being used increasingly in structural engineering applications. However, these PPE composites indicated the lower thermal properties as compared with original PPE injection molding parts. On the other hand, we recently reported jute-fabric reinforced engineering thermoplastic sandwich composites [23], and the jute fabric was pre-coated with thermosetting resin to improve their thermal resistance. As a result, we have successfully fabricated the high thermal resistance natural fiber/engineering thermoplastics composites.

In this study, we try to enhance the thermal resistance of jute fabric in order to facilitate their usage with engineering thermoplastics, *i.e.* polyamide 6. The jute fabrics were coated with flexible epoxy resin to improve their thermal resistance prior to molding of the composites. In addition, the effect of the amount of curing agent to react with jute fabric and epoxy resin was discussed. The thermal decomposition characteristics of the jute fabrics before and after coating were evaluated by using thermal gravimetric analysis (TGA) while mechanical tests were also performed to evaluate the performance of the composites.

## 2. Experimental

### 2.1. Materials

The matrix resins used in this study was Polyamide 6 (PA6) (grade CM 1056) supplied by Toray Co., Ltd. The flexible epoxy resin (grade PB 3600) and curing agent (grade B 0231) were supplied by Daicel Chemical Co., Ltd. Recycled woven jute coffee bags with a thickness of approximately 0.5 mm provided by a coffee company in Japan were used as the reinforcement.

### 2.2. Sample Preparation Methods

#### 2.2.1. Preparation of Matrix Resin Sheet

PA6 pellets were dried at 120°C for 8 hours. These pellets were then used to form matrix sheets by compression molding at 270°C under a constant pressure of 10 MPa. The pellets were pre-heated for 3 min while molding time was 2 min. The molded sheets were then cooled for 3 min prior to the release of pressure.

#### 2.2.2. Surface Coating by Using Flexible Epoxy Resin

The jute fabrics were dried at 80°C for 24 hours prior to immersion in flexible epoxy. The flexible epoxy was dissolved in acetone before using. The volume ratio between flexible epoxy and acetone was 1:1. The concentrations of curing agent for the flexible epoxy were varied at 0, 5, 10, 15, and 20 wt%. After 2 minutes immersion,

the jute fabric were hung up and allowed to cure at 80°C for 2 hours in the oven. The thickness of the resin coating was determined to be approximately 0.15 mm. The weight content of each material was showed in **Table 1**.

#### 2.2.3. Composites Fabrication

TGA measurements were carried out on the uncoated and coated jute fabrics by using a thermo gravimetric analyzer (TGA) (TA Instruments 2950) over temperature range of 40°C to 600°C at a heating rate of 50°C/min. Measurements were performed in air. The weight of all samples was maintained around 5 mg.

### 2.3. Characterization

#### 2.3.1. Thermogravimetric Analysis (TGA)

TGA measurements were carried out on the uncoated and coated jute fabrics by using a thermo gravimetric analyzer (TGA) (TA Instruments 2950) over temperature range of 40°C to 600°C at a heating rate of 50°C/min. Measurements were performed in air. The weight of all samples was maintained around 5 mg.

#### 2.3.2. Flexural Testing

The specimens were cut into strips in order to perform 3-point flexural test (ASTM D790) by using an Instron 4206 universal testing machine at 28°C. Specimens of 125 mm length, 10 mm wide and 3 mm thickness were cut from the composite such the jute warp oriented along the length of the specimen. The flexural tests were conducted at a crosshead speed of 1 mm/min. Span length was 48 mm. Tensile test was performed by the Instron 4206 universal testing machine at a crosshead speed of 1 mm/min. At least five composite specimens were test for each condition.

#### 2.3.3. Tensile Testing

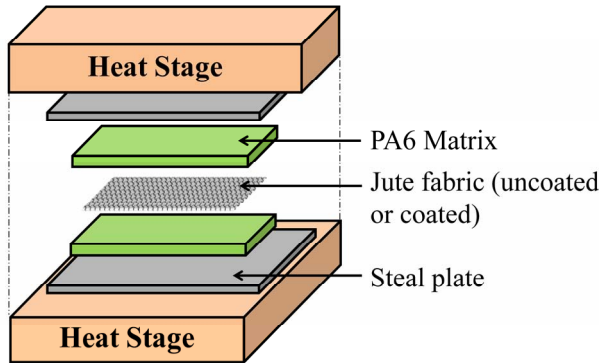
Tensile testing was carried out with an Instron 4206 machine at 28°C, according to ASTM D 3039, at a cross head speed of 1 mm/min. At least five composite specimens were test for each condition.

#### 2.3.4. Morphology Studies

A scanning electron microscope (JEOL, JSM5200) was used for morphology studies of the gold sputtered fracture surfaces of the composites. The polished cross-sections of the composites were observed by using a digital microscope (Keyence VH-S30). The specimens were finely polished to a mirror finish by gradually changing the roughness of the polishing medium from coarse to fine (*i.e.* polishing paper index from 200 to 400, 800, 1200, and 2000) with constant water flow over the specimen and polishing papers to prevent the specimens

**Table 1. Weight content of each material.**

Materials	Weight content (wt% of composites )
Jute Fabric	10.73
Flexible Epoxy	8.72
PA 6	80.53



**Figure 1. Schematic representation of the compression molding process.**

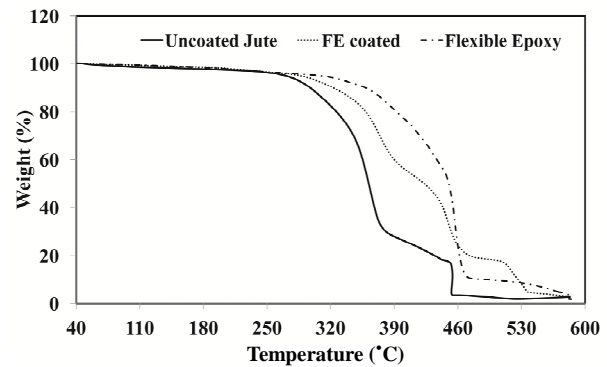
from being damaged by heat and also to flush away the debris. Further polishing using graded alumina suspensions in water was performed. The alumina practice size was gradually changed from 1.0 to 0.1 and finally 0.05  $\mu\text{m}$  (3 min for each polishing stage). The polished specimens were thoroughly washed to discard any residue by immersing the specimens into an ultrasonic cleanser filled with clean water for 15 minutes.

### 3. Result and Discussion

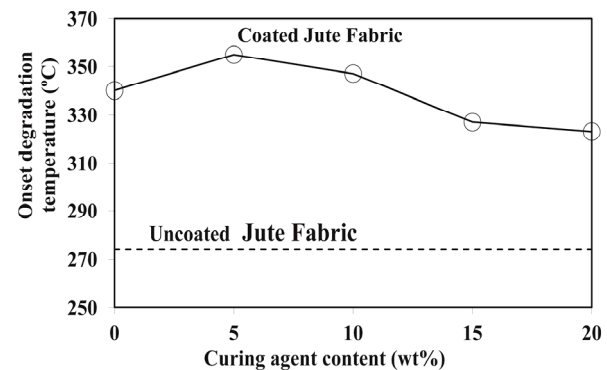
#### 3.1. Thermal Degradation Resistance of Jute Fabric

**Figure 2** shows the TGA thermograms of uncoated and coated jute fabrics containing 5 wt% of curing agent. It can be seen that uncoated jute fabrics started to degrade considerably after 270°C. Meanwhile, the fabrics coated with flexible epoxy resin recorded an onset degradation temperature of 355°C. This indicates that the thermal degradation resistance of jute fabric can be significantly improved with the flexible epoxy coating.

The effect of curing agent concentration on thermal degradation resistance of flexible epoxy coated jute fibers is shown in **Figure 3** Although the thermal degradation resistances of all epoxy coated jute fibers were higher than the uncoated fibers, the onset degradation temperature decreased with increasing curing agent content. Thus indicating that 5 wt% of curing agent is the optimum concentration as thermal degradation resistance decreased at higher curing agent concentrations. The lower degradation temperature at high curing agent contents could be due to the presence of flammable residue from the curing



**Figure 2. TGA thermograms of flexible epoxy, uncoated and flexible epoxy coated jute fabrics.**



**Figure 3. Onset degradation temperatures of uncoated and coated jute fabrics with flexible epoxy.**

agent. However, the incorporation of curing agent is unavoidable since it is important for the epoxy coating on the jute fibers to solidify prior to composite fabrication. Therefore, it is thought that the usage of 5 wt% curing agent is sufficient for curing of the resin as it provides good thermal stability to the jute fibers.

#### 3.2. Effect of Surface Coating on Mechanical Properties

The flexural moduli of composites of uncoated and coated jute fabrics as compared to neat PA6 are elaborated in **Figure 4**. The flexural modulus of the matrix and uncoated jute fabric are found to be 2.35 and 2.65 GPa, respectively. For the coated jute fabric, the flexural modulus increased with increasing curing agent content. This indicates that the flexural modulus can be improved when curing agent content increased due to the increased stiffness of the fabric after coating with flexible epoxy.

**Figure 5** shows the flexural strength of the composites as compared with neat PA6. The flexural strength of neat PA6 and uncoated jute composites are similar, with the coated composites showing lower to higher values with increasing curative concentration. The Low flexural

strength of coated jute composites at lower curative content could be due to incomplete curing of the epoxy. However, the flexural strength increased with increasing curing agent concentration that could be attributed to high crosslinking of flexible epoxy resulting in improved strength of the fabric and consequently affected on reinforcement of the matrix.

Figure 6 elaborates the tensile modulus of composites

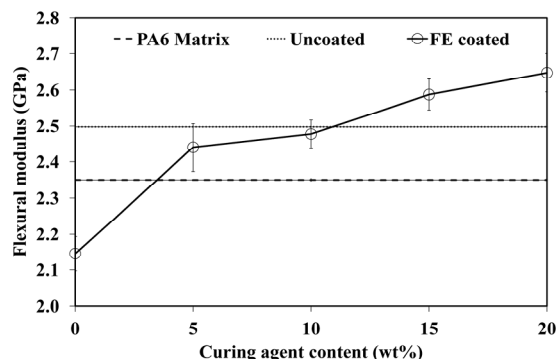


Figure 4. Effect of curing agent concentration on the flexural modulus of PA6 reinforced with flexible epoxy coated and uncoated jute fabric.

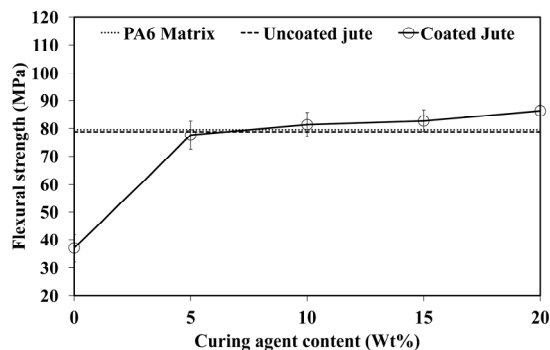


Figure 5. The effect of curing agent concentration on the flexural strength of PA6 reinforced with flexible epoxy coated and uncoated jute fabric.

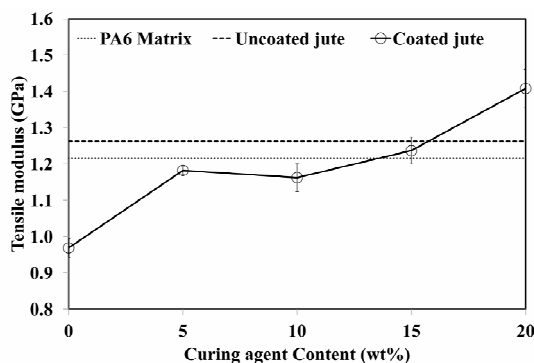


Figure 6. Effect of curing agent concentration on the Tensile modulus of PA6 reinforced with flexible epoxy coated and uncoated jute fabric.

of uncoated and coated jute fabric as compared to neat PA6. The tensile modulus of the uncoated jute composite and neat PA6 are found to be 1.38 and 1.21 GPa, respectively. It is obvious that the uncoated jute fabric shows superior tensile modulus as compared to the coated composites with the exception of the 20 wt% concentration. It should be noted that PA6 is not a hydrophobic matrix [6-10] and therefore good interfacial interaction is expected to arise from hydrogen bonding between jute surface and PA6. For the coated jute fabric, the tensile modulus of the composites is lower than the uncoated jute albeit the tensile moduli of coated jute increased slightly with increasing curing agent content. It can be seen that tensile modulus can be slightly improved by flexible epoxy when curing agent content increased.

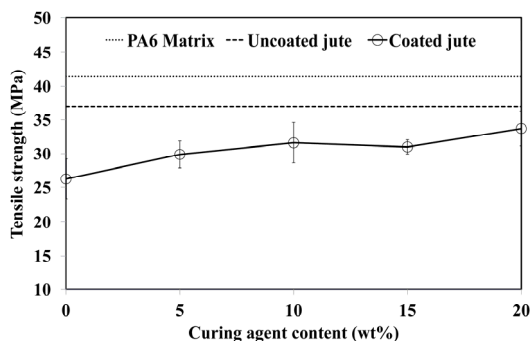
Moreover, tensile strengths of coated jute composites as shown in Figure 7 are all weaker than the uncoated jute composite. This result is contrary to expectation as natural fibers are known to have good interfacial interaction with epoxy [6-10] while the epoxy is expected also to serve as a good compatibilizer between jute and PA6 matrix since it is situated in between them and it interacts positively with both. This contradiction in tensile properties can be explained by the fact that the epoxy has been cured prior to the composite fabrication stage. As epoxy is a thermoset that becomes intractable when cured, substantial chemical interaction failed to occur during the composite fabrication so the interface between the flexible epoxy and PA6 is weak and that could be the source of the weakness of the coated composites. This notion is clarified by the polished cross sections of the composites as discussed later.

### 3.3. Morphology Studies

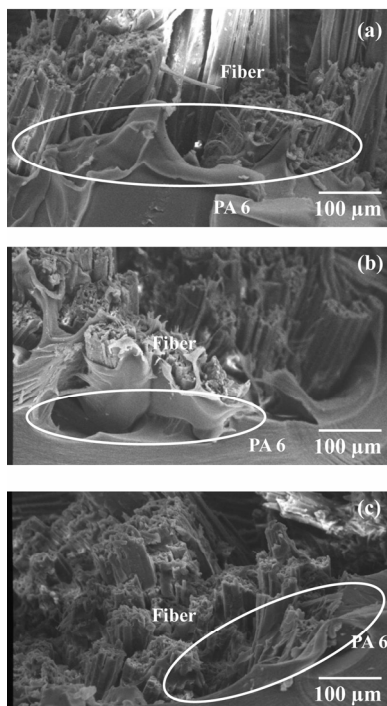
Figures 8(a)-(c) show the SEM micrographs of the tensile fracture surfaces of uncoated and coated jute fibers. It can be seen from Figure 8(a) that the fiber/matrix interfacial bonding between jute and PA6 is quite strong. This is indicated by the lack of fiber pull out and the fact that the matrix and fibers fractured on the same plane. The strong interfacial interaction could be due to hydrogen bonding between the jute fibers and PA6. Similarly strong interfacial bonding is displayed by the flexible epoxy coating and jute fibers in Figures 8(b) and (c) for the 0 wt% and 20 wt% curative contents respectively. This is expected as epoxy is known to have good interfacial interaction with natural fibers. However, the poor mechanical properties of the epoxy coated jute/PA6 composites could be traced to the poor interfacial interaction between the fully cured epoxy and PA6 as revealed by the photo micrograph of the polished cross section of the epoxy coated jute/PA6 composites.

Figure 9(a) elaborates the photo micrograph of the

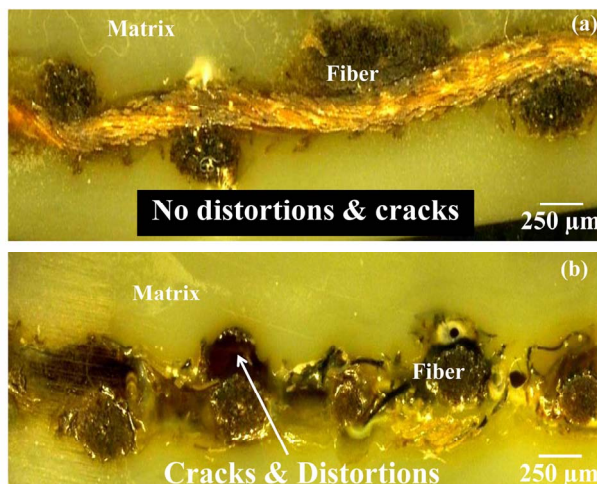
polished cross section of the uncoated jute fabric/PA6 composite. The fiber bundles in the warp and weft directions are clearly indicated with adequate wetting by the matrix and free from imperfections. However, **Figure 9(b)** shows a lot of distortions and cracks along the interface between flexible epoxy and the PA6 matrix. Even though positive interaction is predicted between the epoxy and PA6 matrix, imperfections arose from the inability of the already cured epoxy to adequately interact with PA6. Being a thermoset, epoxy becomes intractable when cured and so it could not melt and interact with PA6 during the composite fabrication stage. This accounts for the poor interfacial interaction between epoxy and PA6 and



**Figure 7.** Effect of curing agent concentration on the tensile strength of PA6 reinforced with flexible epoxy coated and uncoated jute fabric.



**Figure 8.** SEM micrograph of the tensile fracture surface of jute fabric/PA6 composite; (a) Uncoated; (b) Curing agent 0 wt%; (c) Curing agent 20 wt%.



**Figure 9.** Photo micrograph of the polished cross section of jute fabric/PA6 Composite; (a) Uncoated jute fabric; (b) Coated jute fabric.

hence the poor tensile properties of the epoxy coated jute/PA6 composites.

#### 4. Conclusion

This paper investigation intended to improve the thermal stability of natural fibers with the use of surface treatments *i.e.* flexible epoxy so that the natural fiber can be processed with high temperature engineering thermoplastics. It was found that the flexible epoxy coating was able to improve the thermal degradation resistance of jute fibers with the best result given by 5 wt% curative concentration. However, flexural properties improved continuously with increasing curative concentration. The epoxy coated jute fabric composites showed inferior tensile properties as compared to the uncoated jute fabric composite albeit the properties increased with increasing curative content. SEM studies revealed strong fiber/matrix interfacial bonding for both coated and uncoated jute fabric composites. The inferior tensile properties of the resin coated jute composites could be attributed to the poor interfacial interaction between the flexible epoxy coating and the PA6.

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