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Improvement of mechanical properties of polypropylene composite using filler, modifier and reinforcement

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Abstract. This study focuses on improvement of mechanical and physical properties of polypropylene (PP) composite using filler, modifier and jute fiber as reinforcement. Rice husk ash (RHA), low-density polyethylene (LDPE) and jute fiber were used as filler, modifier, and reinforcement in the PP matrix, respectively. A series of test specimens were using various compositions of PP, fillers, modifiers, and reinforcement. Materials were mixed using two roll mixing machine, and specimens were prepared using injection molding machine. The specimens were evaluated on the basis of mechanical (tensile strength, elongation at break) and Physical (water absorption) properties. Effect of RHA was assessed on the basis of mechanical properties. Surface property of the PP composites was studied using Scanning Electron Microscope (SEM). The water absorption property of composites with various compositions was investigated. Further attempts had been made to optimize the three process parameters (RHA, LDPE, jute fiber) in the PP composite with respect to its tensile strength. The optimum parameters for the PP composite were found to be 10 wt% RHA, 10 wt% LDPE and 25 wt% jute reinforcement, respectively in the PP matrix.

1. Introduction

Technological development mostly depends on advancements in the field of engineering materials. Composite materials in this regard represent nothing less than a giant step in the ever constant effort toward optimization in materials. Over past several years, there has been an increasing interest in the use of polymer composite due to their various advantages over other materials. This lightweight type of composites allows for lower fuel consumption which can be used in a broad range of applications, including in the aerospace, automotive, and rail sectors [1].

The use of lignocellulosic fibers as the reinforced materials in thermoplastics has received a lot of interest due to their low densities, low cost, and non-abrasive nature [2], which minimizes the wear of processing equipment and so forth [3-4]. Compared with thermosets, thermoplastic composites materials typically have higher strain to failure, faster to consolidate and retain their ability to be recycled [5]. Among other thermoplastic polymers, Polypropylene (PP) provides most of the advantages with regards to economic (price), ecological (recycling behavior) and other technical requirements [6]. Previously, jute fiber was used as a reinforcing agent in the polymer matrix with various advantages and minimal environmental effect because of its biodegradable properties [7, 8]. Commercially available silica fillers significantly improve composite mechanical properties [9, 10].

In Bangladesh, huge quantities of rice husk ash (RHA) have been generated as waste materials from the rice processing mill. Depending on the combustion conditions, RHA contains a large quantity of silica which is in partly crystalline and amorphous forms, the rest being an amalgamation of carbon
impurity and a small fraction of metal oxide impurity. The composition of the rice husk ash is given below [11].

| Content, wt.% | SiO₂ | Al₂O₃ | Fe₂O₃ | CaO | MgO | K₂O | Na₂O | P₂O₅ |
|---------------|------|-------|-------|-----|-----|-----|------|------|
| 93.4          | 0.05 | 0.06  | 0.31  | 0.35| 1.4 | 0.1 | 0.8  |

In this study, the technical feasibility of RHA as filler materials in the PP matrix has been studied. Effect of RHA, LDPE and jute fiber in the PP matrix has been evaluated separately, and optimum content of the filler, modifier, and reinforcement have been determined. Micro-structural analysis (SEM) and water absorption characteristics of different PP composites have also been reported.

2. Materials and Methods

2.1. Materials

Materials used in this study are Polypropylene (PP) pellets as matrix material, jute fiber as reinforcement and LDPE as modifier and RHA as filler. The PP was supplied by Polyolefin Company Pvt., Ltd., Singapore. The PP has a density of 0.905 g/cm³ at room temperature and the Melting Temperature (Tₘ) and Melt Flow Index are 160°C and 6 g/10 min., respectively. The jute fiber was collected from local market of Gazipur, Dhaka, Bangladesh. LDPE was commercial grade, granular and virgin. It was manufactured by The Polyolefin Company Pvt., Ltd., Singapore. RHA was used as filler in the composite which was bought from local market of Gazipur, Dhaka, Bangladesh.

2.2. Jute preparation

The jute fiber was first washed and then dried in an oven at 40°C for 24h. The dried jute fiber was then cut into 2-3 mm in length for the fabrication of jute-PP composites.

2.3. Rice husk ash (RHA) preparation

Rice husk ash collected from local rice processing station. The blackish rice husk contained carbonaceous material. In view to burn carbonaceous material completely, it was heated at 840°C for an hour in the Muffle furnace. The ash was then collected from the furnace. It was turned into a grayish color which was used as filler in the PP matrix.

2.4. Composite fabrication

In this study, three different types of composite specimens were prepared.

i. RHA blended with PP to prepare RHA-PP composite

ii. Jute fiber reinforced PP composite

iii. LDPE mixed with PP to prepare LDPE-PP composite

PP and the other ingredient was mixed using two roller mixing machine by applying heat at 130-140°C to turn it into a uniform mass. This mixing results in a sheet of PP with other ingredients. The prepared sheet was then cut into small pieces and fed into injection molding machine. In the injection chamber, the molding compound is heated at a temperature about 110°C and therefore it changes into fluid form and after that forced through an orifice into the desired dog bone shaped mold which is kept relatively cool. The composite solidifies in the mold from which it can then be removed for further testing.

2.5. Evaluation of specimens

Mechanical (tensile strength, elongation at break) and physical (water absorption) properties of the polymer composites was evaluated in the study.

Tensile test

The tensile strength of the PP composite specimens was measured following ASTM D638 01 [12] using a Universal Testing Machine (Model DRK101-300; Shandong Drick Instruments Co. Ltd, China) at a cross-head speed of 25mm/min. The size of the sample used for tensile strength test was
150 mm × 10 mm × 3.2 mm. Five specimens of each type of composite were tested and the average values of Tensile Strength and %Elongation were reported by calculating of maximum four values.

Water Absorption
Specimens of the composites were immersed in the static water glass beaker at room temperature for different time periods (up to 72 hours). Before immersion, the weight of the samples was taken. At certain time intervals, samples were taken out from the beaker and wiped smoothly with tissue paper and after that weighed promptly. Percent of water uptake was then calculated using the following equation:

$$W_{up} (%) = \frac{(W_f - W_i)}{W_i} \times 100\% \quad (1)$$

Where, $W_{up}$ = water absorption, wt%
$W_i$ = weight of the sample before immersion in water
$W_f$ = weight of the sample after immersion

2.6. Micro-structural analysis
Micro-structural analysis of PP composites with varying compositions was conducted using Scanning Electron Microscope (SEM, Model: JSM-6701F) supplied by JEOL Company Limited, Japan.

3. Results and Discussion
3.1. Tensile strength
Effect of RHA content on the tensile strength is presented in Figure 1. The figure shows that addition of RHA up to 10 wt% in the PP matrix improves the tensile strength; further addition of RHA reduces the strength. The initial increase in strength of the composite is due to good filler-matrix interactions, which also enable more stress to be transferred from the matrix to the fillers during external loading. Above 10 wt% of RHA, with further increase in filler content, the matrix-filler aggregates deform easily in the composite due to the reduction of binder matrix (PP) and an increase of filler material, which tend to reduce the strength of the composite.

![Figure 1. Tensile strength as a function of RHA content, wt%](image)

Variation in the tensile strength of PP composite with jute fiber content is presented in Figure 2. The Figure shows that the tensile strength of jute-PP composite increases up to 25 wt% jute fiber content in the PP matrix, thereafter strength decreases with the further addition of jute fiber content in the matrix. The initial increment in strength can be explained by the interfacial bond formation between polymer and jute. After the optimum composition of PP matrix with 25 wt% jute fiber loading, the figure shows a reduction in strength because of improper mixing and the weak interfacial area between the fiber and matrix. A large content of jute fiber prevents the polymer melt from wetting the jute fiber during the molding.
Figure 2. Tensile strength as a function of jute content, wt%
Blends of PP and LDPE contribute to making recycling more economically attractive. Effect of LDPE content on the tensile strength of PP composite is presented in Figure 3. The figure shows that PP composite containing 10 wt% of LDPE shows optimum tensile strength. The effect of LDPE in the PP matrix is as similar as RHA. Addition of LDPE up to 10 wt% improves the tensile strength; further incorporation of LDPE in the PP matrix reduces the strength. At lower filler content, matrix-filler interactions are quite good and the tensile strength is better in that region. After the optimum point, amount of binder matrix material continually decreases as the LDPE content increases which is the reason behind matrix-filler deformation and lower tensile properties.

Figure 3. Tensile strength as a function of LDPE content, wt%

3.2. Elongation at break
Figure 4 shows variation in elongation of PP-RHA composite with RHA content in the composition. The figure indicates that addition of RHA in the PP matrix decreases elongation of the PP composite. As tensile strength increases with increasing filler content, the elongation decreases simultaneously and vice versa.
Variation in elongation behavior with jute fiber content in the PP matrix is presented in Figure 5. The figure shows that elongation decreases with the increased jute fiber content in the PP matrix. The cause behind the decrease is the cross-linking between the neighboring jute with Polypropylene chain which increases the rigidity and toughness of the composite material but decreases its ductility.

The elongation at break for the different content of LDPE in the PP matrix is presented in Figure 6. It is found that PP composite with 10 wt% LDPE shows higher elongation at break.

The water absorption capacity of PP composites with different RHA content is presented in the Fig. 7. The figure shows that water absorption increases with increasing filler RHA content. This result is expected since RHA is hydrophilic in nature, the amount of water uptake increases significantly with increasing RHA content.
Figure 7. Water absorption capacity as a function of RHA content, wt%.

Figure 8 shows the water absorption capacity of PP composite as a function of jute fiber content. The figure shows that water absorption capacity increases with increasing jute fiber contents in the matrix. The reason is that the PP matrix is hydrophobic and a layer of PP always repels water to penetrate it into the composite. Since the jute is highly hydrophilic in nature and as the jute content increase in the composite, some of the jute pulls out from the PP matrix and absorb water and this leads to the further uptake of water into the composite matrix.

Figure 8. Water absorption capacity as a function of jute content, wt%.

3.4. Micro-structural analysis

Figure 9. SEM micrograph (a) PP with 10 wt% CaCO$_3$ and (b) PP with 10 wt% RHA.
Micro-structure of PP-CaCO$_3$ and PP-RHA composite have been analyzed using SEM. Figure 9 clearly indicates improved crystallinity in the RHA-PP composite than the CaCO$_3$-PP composite. So, RHA can be a good replacement of conventional CaCO$_3$ filler in the composite without degrading the composite mechanical properties.

4. Conclusions
Rice husk ash was used as alternative fillers in replace of conventional filler calcium carbonate in the PP matrix. The tensile strength and elongation at break of the RHA blended PP composites was found maximum at 10 wt% RHA content in the composite. The SEM micrograph of RHA-PP composite showed improved crystallinity than CaCO$_3$-PP composite. Therefore, RHA can be used as an alternative of CaCO$_3$ in PP composite which is environment friendly, biodegradable and cheap filler. Water absorption properties of the different specimen showed that water absorption capacity increases with both RHA and jute content in the PP matrix. Based on mechanical properties, the optimum parameters for the PP composite was found to be 10 wt% RHA, 10 wt% LDPE and 25 wt% jute reinforcement, respectively and in the PP matrix.

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