Experimental Investigation of Rice Husk Particles as Filler in Hybrid Composites

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Abstract
Natural fiber is being used in various engineering applications to lower the production cost and to apply green technology. This research explores the effective utilization of rice husks (RH) as natural filler in polymer composites. A Hybrid polymer composite was prepared by inserting micro particles of rice husks and fumed silica Nano-powder in an epoxy matrix system. Composites containing different weight fraction of RH were used (5 %, 10%, 15% and 20 %). The variation in some physical and mechanical properties were studied and analyzed. Results showed that tensile properties increase with the amount of the natural filler, plus a slight improvement in hardness tests. Water absorption experiments showed that the diffusion coefficient increased with RH loading. This investigation suggested the possibility of introducing rice husks fine powder obtained from waste agricultural residues as filler in polymer matrix composites.

Keywords: Composite materials; Epoxy resin; Mechanical properties; Rice husk.

1. Introduction
A composite material is a conjunction of two or more chemically and physically different components that yields unique features in the final new formed material. Composites possess many advantages like high specific stiffness and strength, lightweight; good fatigue resistance, and low thermal expansion. These properties make them a suitable material choice for advanced and complicated applications. Despite these great advantages, composites are considered expensive materials.

Epoxy resins were widely used as a matrix in composites due to its good mechanical, outstanding adhesion to different substrates, low shrinkage upon cure, and the ability to be processed under various conditions. The major disadvantage of epoxy resins is brittleness. As a result, a reinforcement or filler materials must be used to carry the load and provide the strength and substantially that cannot be supplied by matrix alone. It has been shown by several investigators that the combination of fillers, such as silica, carbon or graphite fibers and aromatic polyamides can improve the toughness of epoxy resins (Oleksy et al., 2014; Hulugappa et al., 2016). In spite of these advantages, the widespread use of synthetic
fiber-reinforced polymer composite has a tendency to decline because of their high-initial costs and also the production of synthetic composites requires a large energy and causes environment pollution. (Zini and Scandola, 2011) stated that 50% of volume of the glass fibers disposal remains as unburned residues. As a result, it should be dumped or left in landfills due to its accumulation and gathering which causes land pollution (Shubbar and Al-Shadeedi, 2017).

Natural fibers, as reinforcement, have recently attracted the attention of researchers because of their advantages over other established materials. They are environmentally friendly, fully biodegradable, cheap, abundantly available, and have low density (Debnath et al., 2013; Ahmad et al., 2015).

Rice husk (RH) is one of the cheapest natural fibers in the region. It is the outer covering of rice seeds that are usually removed by milling as a by-product and it contains a high percent of cellulosic materials (about 54%) and a high amount of silica (15-20%) that makes it a potential material for many industrial application. One of these applications includes using them as alternative waste materials to substitute plastic products. By using RH as filler we gain two goals: the first is minimizing environmental problems caused by plastic waste and the second reduction the composites production costs (Raju et al., 2012; Kumar et al., 2013).

The main problem with using natural fiber as filler in composites is its water absorption that may cause decrease in mechanical properties after swelling. Chemical treatments can increase the interface adhesion between the fiber and matrix, and decrease the water absorption of fibers (Ismail and Waliuddin, 1996; Li et al. 2007; Alamri and Low, 2012) stated that the addition of Nano silica carbide can decrease the water uptake and enhance the mechanical properties of the polymer.

Applying RH as reinforcing material in polymer composites is being carried out by various researches. (Rozman et al., 2004) studied the effect of glass fiber length on some mechanical properties of rice husk–glass fiber polyester composites. They stated that longer glass fiber had a greater effect on the strength and toughness. The mechanical properties of hybrid composites were investigated by (Mingchao et al., 2009). They found that increasing the amount of fiber increased the flexural modulus. Ofem et al., 2012) focused on preparing cashew nut resin reinforced rice husk composite and noticed that the strength increases as the particle sizes increases. Also there was improvement in mechanical properties when rice husk increased but reduction as the particle sizes increases except for impact. (Hardinnawirda and Aisha, 2012) stated that with the increasing rice husk filler there were a decrease in tensile strength of the unsaturated polyester composite and its water absorption increased. While(Debnath et al., 2014) investigated friction and wear performance of glass fiber reinforced polymer composites filled with rice husk and noticed a decrease in specific wear rate with increasing the applied normal load. (Rout et al., 2012) studied both thermal and mechanical properties of epoxy resin reinforced with glass fiber and filled with rice husk particulates and indicated that micro- Hardness increased while tensile and flexural properties had decreased with addition of rice husk, and coefficient of thermal expansion was decreased noticeably.

(Sachudhanandan et al., 2015) examined tensile strength and flexural strength of rice and aramid reinforced vinyl ester composite, and noticed that there is significant increase in the tensile strength and flexural strength of the composite.
they stated that Hybridization results a higher figures in tensile strength. (Prithivirajan et.al.,2015) examined the addition of rice husk and coir pith particulates to reinforce the epoxy composite, and noticed a considerable increase in mechanical properties of the composites when adding fillers. (Bisht and Gope, 2015) stated that although adding rice husk to epoxy resin reduces some mechanical properties, but it’s a better reinforcement when compared to wood dust.

Since sustainability concept became an important goal, the objective of the current work is to determine the effect of using different percentage of rice husk fine powder as cheap natural filler in epoxy matrix material and to evaluate some mechanical and physical properties of the related composite.

2. Materials and Methods
2.1. Materials
2.1.1. Epoxy resin
Modified low viscosity Epoxy resin Araldite CY 223 and hardener based on poly amidoamine HY 956 were used as the matrix materials. These resins were supplied by Ciba Geigy, having excellent adhesion to different materials, high dimensional stability, excellent mechanical properties, and negligible shrinkage.

2.1.2. Rice husk
Rice husk was obtained as a by-product from a local rice mill in Najaf. To reduce its moisture absorption it was chemically treated by soaking in a solution of 4% sodium hydroxide solution for about three hours. Then it was washed well with water. After this, the husk was dried in oven for (5-6 hours) at 100°C to remove any remaining humidity present in it. Then it was grounded in a laboratory grinding mill followed by screening it in a sieving shaker to obtain particles less than 90 μm in diameter as shown in Figure 1. The final step was putting the powder in desecrator to insure remaining dry.

Figure 1. Rice husk before (on the left) and after (on the right) grinding and sieving

2.1.3. Fumed silica
Fumed silica AEROSIL are non-porous amorphous nano silica particles (less than 20 nm in diameter) which has been used successfully as thickening filler in
epoxy systems was used to prevent the filler sedimentation during curing process and to ensure uniform distribution of the particles in molded samples.

2.2. Preparation of composites

The composite were fabricated by hand lay-up technique. Molds used for fabricating the composite were made up of 4 mm thick Perpex sheets, and the cavities were done by a CNC machine. The mold was positioned on a plate of clear glass, and to ensure easy removal of samples both the mold and glass were sprayed well with releasing agent as shown in Figure 2.

The milled rice husk were mixed with epoxy resin by varying the percentage of RH (0, 5, 10, 15 and 20 wt. %). Then fumed silica was added in a fixed ratio to all samples (0.25 wt. %), and they were mixed with a high speed mechanical mixer for about 5 minutes, then containers were left to settle down to ensure it has no air bubbles inside it, then the hardener was added with the recommended ratio (3:1 by weight) with a gentle and short time mixing to avoid epoxy crosslinking. The casting of samples was done slowly in order to prevent bubbles and flaws formation in the samples. The setup is left to cure completely for 24 hours at room temperature. Table 1 illustrates the sample composition.

<table>
<thead>
<tr>
<th>Composite sample code</th>
<th>Composition</th>
</tr>
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<tbody>
<tr>
<td>R_0</td>
<td>Pure Epoxy</td>
</tr>
<tr>
<td>R_1</td>
<td>Epoxy (94.75%) + 0.25% fumed silica + Rice Husk (5%)</td>
</tr>
<tr>
<td>R_2</td>
<td>Epoxy (89.75%) + 0.25% fumed silica + Rice Husk (10%)</td>
</tr>
<tr>
<td>R_3</td>
<td>Epoxy (84.75%) + 0.25% fumed silica + Rice Husk (15%)</td>
</tr>
<tr>
<td>R_4</td>
<td>Epoxy (79.75%) + 0.25% fumed silica + Rice Husk (20%)</td>
</tr>
</tbody>
</table>

Figure 2. Casting process for the composite in the mold
2.3. Characterization of the Composites

2.3.1. Tensile test

This tensile test was conducted with a computer controlled universal testing machine at cross-head speed of 2 mm/min. The test was carried out in accordance with ASTM D3039 with the average results obtained from five dumbbell-shaped samples were reported for each sample.

2.3.2. Hardness test

The hardness test was accomplished by a Durometer that is commonly used for measuring the indentation hardness of epoxy composites according to (ASTM D-2240-68). The device is manufactured by ZWICK Co. Germany. The type used was Shore D that can be defined as resistance of a material against the penetration by a cone. The readings were taken after 1 sec of applying the load (5kg).

2.3.3. Water Absorption test

Long term water immersion method was used to determine the absorption behavior of the prepared hybrid composites. The water absorption test of the composites was carried out according to ASTM D570-98 with repeated immersions in distilled water at room temperature. It was measured by soaking the samples in a beaker of water at 25°C for different periods (up to 7 days). The first step was putting all the samples in an oven at 105°C overnight to ensure its dryness. After waiting it to cool down in a desecrator, its weight was measured to get the dry mass of the sample. Then it was immersed in distilled water and after certain period of times (24 hour), samples were taken out from the water, wiped, and then weighted. The mass gained was measured by subtraction of initial weight from final weight. The percentage of water absorption was determined by using the formula:

\[ MC\% = \frac{(m_2 - m_1)}{m_1} \times 100\% \]

where \( m_2 \) = the weight after immersion, \( m_1 \) = the dry weight before immersion.

3. Results and discussions

3.1. Tensile strength

The tensile strength at break for rice husks composites were presented graphically in Figure 3. \( R_0 \) represents the sample with pure resin; in the next sample, \( R_1 \), Nanoparticle content increased from 0% to 0.25wt. % and rice husk micro particles were added by 5wt. %. An escalating increase in tensile strength was seen in this stage, followed by a gradual increase in tensile strength with increasing rice husk content. The initial increase by adding 5wt. % RH was about 50% is probably due to the action of both fumed silica and rice husk filler to plain epoxy, which finally improves the load carrying capacity of the composite. The addition of rice husk particles in fumed silica-epoxy composites by 20 wt. % improved the tensile strength by 79%. The reason for this increasing may be a result of the crystalline structure of rice husk, which improve the load carrying efficiency of the composites. Another reason for the increase in tensile strength as filler loading increase is attributed to the prior chemical treatment of RH, which removes impurities and roughens its surface.
features in order to perform a good adhesion between the matrix and filler (Bisanda and Ansell, 1991). These results coincide with Dhakal et al. (2007), where they reported that by adding filler to the polymer material, the strength of the composite could be enhanced.

![Figure 3. Tensile strength values for the prepared composite](image)

### 3.2. Hardness test

The effect of rice husk content on hardness of the composite is shown in Figure 4. It shows that hardness of the composite is a function of (rice husk) content. The hardness of the RH composite is found to lie in the range of 65 and 75 Shore D. The hardness of plain unfilled epoxy shows remarkable increase (about 39%) with 5% rice husk addition. The reason is due to the presence of two types of filler incorporated to pure epoxy, including rice husk and the Nano fumed silica, which fills the voids that may be present in epoxy matrix and increase the hardness. Then the increasing rate of hardness became much less after increasing RH content alone. The hardness improvement was 10% when raising the RH from 5 to 20% in the composite. This improvement is because Nano particles of fumed silica enrich the bonding strength between rice husk and the epoxy resin and provide resistance against penetration, especially on the surface, during hardness test.

![Figure 4. Shore D hardness as a function of RH content in epoxy composite](image)
3.3. Water Absorption

The effect of immersion time on water absorption of RH composites prepared with different wt. % of RH is shown in Figure 5. It can be seen that in the case of R_0, the rate of water absorption is very low with time, but with adding RH there were an increase in water uptake with raising the filler content in composite.

The water absorption in composites is mainly due to that most of natural cellulose fillers and RH is one of them, are lignocellulosic, polar and hydrophilic in nature, due to their chemical structure, that provides active sites for the adsorption of water. So when the RH filler composites were soaked in water, they swelled and caused minor cracks in the brittle epoxy matrix. That cracking generates capillarity that causes swelling and water absorption, leading to an increase in weight. The high cellulose of the rice husks contribute to additional water penetration into the interface through micro-cracks induced by swelling of the filler, which also creates stress and ultimately, leads to failure of the composite (Dhakal et.al., 2007).

![Figure 5. Effect of immersion time on water absorption of RH epoxy composite](image)

4. Conclusions

According to results of present work, the following can be concluded:

Based on the results for a hybrid of micro particles of rice husks and fumed silica Nano-powder, in which epoxy was used as a matrix, it was shown that there was a good improvement in tensile strength and hardness with increasing the natural filler content. The water absorption also increased slightly with increasing rice husk particles. So, micro particles of rice husks and fumed silica Nano-powder can represents an effective material for production of environment friendly composites for moderate applications.

References


