Study of the Mechanical Properties of HDPE Matrix Composites Reinforced with Rice Hulls Particles

دَرَاسَة الخواصَ الميكانيَكية لَمَوادَ المركَبة المُتَمَكِّنة من البولي اثيلين عالي الكثافة كأساس مع قشور الرز ككَتَاقَ قوية

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Abstract:
The present article summarizes an experimental study on the mechanical behavior of high density polyethylene (HDPE)/ rice hulls (RH) composites. The mean goals of this investigation were using of rice hulls (RH) as a filler (reinforcer), effectiveness of hulls flour fiber with different size (10-30 mesh as fine) and (30-50 mesh as coarse) on mechanical properties of composites and effect of maleic anhydride (MA) as a coupling agent treatment is performed to improve interfacial adhesion between the particles and HDPE matrix. Generally, (16) treatment were conducted as (8) treatment of HDPE with 5, 15, 25, 35% coarse rice hulls with (0.5, 1.0, 1.5, 2.0) MA, 8 treatment of (HDPE) with 5,15, 25, 35% fine rice hulls (RH) with (0.5, 1.0, 1.5, 2.0) MA, (HDPE), (RH). This study focused upon tensile strength, flexural, impact strength and hardness of reinforced polyethylene. The results showed that increase rice hulls rate to 25% in both fine and coarse, improved [tensile strength, hardness, flexural and impact strength] values, Pine (RH) particles sample showed then, better coarse (RH) when addition for HDPE as matrix to produced composite material. The optimum formulation for the (HDPE and RH) composites on the mechanical proerty is (25wt.%RH as fine-HDPE-1.5wt.%MA).

الخلاصة:

الدراسة الحالية تعتبر محاولة لدراسة سلوك الخواص الميكانيكية لمادة مركبة ناتجة من بولي اثيلين عالي الكثافة كأساس اساس مع قشور الرز ككَتَاقَ قوية، أن الهدف المتحقق من استخدام هذا نوع من الاليف Rice Hulls ذات الاحجام المختلفة (30-50 mesh) (mesh size) من النوع الناعم هو تحصين الخواص الميكانيكية للمادة المركبة الناتجة وبخاصّة بين النوع الناعم والخشن، كذلك تم استخدام عامل الربط Maliec Anhydride (Coupling Agent) من نوع معين في تحسين التلاصق بين البولي اثيلين عالي الكثافة وقشور الرز. ستة عشرة عاملة تم القيام بإجراءها ثمانية لقياس الناية والخشب، حيث استخدمت نسبة ووزينة مختلفة (0.5, 1.0, 1.5, 2.0їل MA) ونسب وزينة مختلفة أيضاً (0.5, 1.0, 1.5, 2.0їل MA). تم دراسة مقاومة الناية (Tensile Strength) ومقاومة الشدة (Impact Strength) ومقاومة الصدمة (Hardness) والصالة Flexural Strength لدراسة المادة المركبة الناتجة. أفضل خواص ميكانيكية تم الحصول عليها عند نسبة الوزينة (25%) في كلا الحجَمَ النايم وقشور الرز. بكل ظروف الدراسة اتبات ان الحجَمَ النايم بعضاً خواص ميكانيكية أفضل من الحجَمَ الخشن، كذلك تم التوصل إلى ان أفضل خواص حصلت في التركيب: 25%wt.RH fine-HDPE-1.5%wt. MA.

Introduction:
New properties, lower prices and reuse of polymers are needed to meet demands of today's society and therefore of the polymer industry. Polymer is the technical name for what is more generally known as plastic, [1]. Plastic are used by almost everybody since (for example) most domestic machines have a housing, which is made out of it [1,2]. Fiber reinforced thermoplastic composites have emerged as a major class of structural material in the field of aerospace, automobiles, construction, etc. These material are characterized by easy processibility, good dimensional stability, and excellent mechanical performance [3,4].

In general, materials may be selected and specified different properties. Here, though, we are mainly interested in composites for structural applications. The two properties of particular
relevance are stiffness and strength, preferably combined with low weight [5]. Fiber reinforcement may be used in several different forms or arrangements, depending on the application and manufacturing route. They are categorized firstly, in terms of length: (I) Short (less than 10mm)-used mainly in thermoplastic moulding compounds. (II) long (10-20mm)-used in chopped strand mat and in thermosetting moulding compounds, and (III) continuous particles, it offer the highest mechanical properties, and give the possibility of using specific orientations to give the composite directional properties. The are available as lengths of fabric in many different woven, knitted or stitched forms, all of which have different properties, processing characteristic and costs. These include: Unidirectional, biaxial, multiaxial and random. Generally, both the fibres and the form of the reinforcement contribute to the properties of the composite, while the manufacturing, cycle is primarily determined by the matrix [6].

The growing interest in using natural vegetable particles as a reinforcement of polymeric based composites is mainly due to their renewable origin, relative high specific strength and modulus, light weight and low price [7]. Recent developments in natural particles such as jute, sisal, coir, flax, rice hulls, etc, have shown that it is possible to obtain well performing materials, using environment freindly reinforcements [8]. The mechanical properties of natural fiber-reinforced composites can, in fact, be further improved by chemically promoting a good adhesion between the matrix and the fiber. Other advantage of utilizing natural particles are related to their cycle of production that is economical and their ease of processing which demands minor requirements in equipment and safer handing and working conditions with respect to glass particles. In any case, the most interesting feature coming from the employment of natural particles is the extremely favorable environmental impact, due to the fact that natural particles are produced from a renewable sourse and are biodegradable [9]. On the other hand , low thermal stability, high moisture up take, and limited fiber lengths, represent some of the disadvantage related to the utilization of natural fiber composites [10].

To enhance the compatibility of the two phase in such composites, a compatibilizer or coupling agent is normally added, to the Mixture. Many researchers have reported improvements in mechanical properties when a compatibilizer was used or the particles were chemically modified prior to mixture [11,12].

In the present investigation , the mechanical characteristics of various rice hulls (RH)-high density polyethylene (HDPE) composites have been investigated. The effect of coupling agent maleic anhydride (MA) treatment on the mechanical properties of composites have been studied.

Materials and Methods

Raw Materials

(a) High density polyethylene (HDPE), powder was supplied by Basrah Petrochemical General Company with a density 950 kg/m³. Its melting temperature (132°C) and melt flow index (MFI) 6g/10 min, was used as the base polymer matrix.

(b) Rice hulls (RH) particles, having a different size (10-50 mesh) obtained from (Al-Tajiah fields near Babylon University), was used as the reinforcing fiber.

(c) Maleic anhydride (MA), supplied by Univoyal chemicals, MB226D. was used as a coupling agent.

Particle Treatment

The Particles, scoured in detergent at 65°C for 30 min. to remove dirt and core material followed by washing with distilled water, were dried in oven at 60°C. The dried particles were cut and grinding to suitable mesh size.

Composites Preparation

Polymer, particles and coupling agent were initially weighed and bagged according to the various fiber contents indicated in Table (1). The composition of each formulation is also shown.
The compounds were prepared by means of hot rolls at a temperature 160°C, which is above the melting point of the polyethylene, for 30 minutes. The rice hulls (RH) previously cut and grindind to (10-50 mesh) size, and dried in an oven at 60°C for 5 hours ensure that moisture contents were blow 0.5%. Once the polymer was melted, the appropriate percentage of particles was added to the polymer. Immediately after mixing, the material was extruded [single screw extruder, C.W. Brabender. I. P. with a screw L/D of 30:1 of 60 rpm] into granules, and then injection molded to obtain standardized specimens. Using compression molded (3 ton) to produce sheet 2mm and 3mm thickness for various testing.

**Conditioning**

After preparation, all specimens were conditioned in a humidity controlled room at 25°C and 50% relative humidity for at least 48 hours prior testing.

**Composite Characterization**

- Tensile testing, sample were cut according to ASTM D638. the machine that was used for the testing of tensile properties is Universal Testing Machine (PHYWE). The test was conducted at velocity 5mm/s at ambient temperature.
- Flexural test was also using Universal Testing Machine (PHYWE) according ASTM D790. for testing, the support span was fixed at 100mm and the rate crosshed motion at 3 mm/min.
- The Izod impact machine was used for this testing. The dimension of the sample specimen conform to ISO179.
- Hardness Test was used Universal testing Machine (PHYWE), the samples were cut according to ASTM D785.

**Table 1: Composition of Evaluated Formulation (wt.%)**

<table>
<thead>
<tr>
<th>Formulation</th>
<th>Specimen Code*</th>
<th>Resin Content</th>
<th>Coupling agent**</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>HDPE-RH-5(fine)</td>
<td>95</td>
<td>-</td>
</tr>
<tr>
<td>2</td>
<td>HDPE-RH-15(fine)</td>
<td>85</td>
<td>-</td>
</tr>
<tr>
<td>3</td>
<td>HDPE-RH-25(fine)</td>
<td>75</td>
<td>-</td>
</tr>
<tr>
<td>4</td>
<td>HDPE-RH-35(fine)</td>
<td>65</td>
<td>-</td>
</tr>
<tr>
<td>5</td>
<td>HDPE-RH-5(coarse)</td>
<td>95</td>
<td>-</td>
</tr>
<tr>
<td>6</td>
<td>HDPE-RH-15(coarse)</td>
<td>85</td>
<td>-</td>
</tr>
<tr>
<td>7</td>
<td>HDPE-RH-25(coarse)</td>
<td>75</td>
<td>-</td>
</tr>
<tr>
<td>8</td>
<td>HDPE-RH-35(coarse)</td>
<td>65</td>
<td>-</td>
</tr>
<tr>
<td>9</td>
<td>HDPE-RH-25(fine)</td>
<td>74.5</td>
<td>0.5</td>
</tr>
<tr>
<td>10</td>
<td>HDPE-RH-25(fine)</td>
<td>74</td>
<td>1.0</td>
</tr>
<tr>
<td>11</td>
<td>HDPE-RH-25(fine)</td>
<td>73.5</td>
<td>1.5</td>
</tr>
<tr>
<td>12</td>
<td>HDPE-RH-25(fine)</td>
<td>73</td>
<td>2.0</td>
</tr>
<tr>
<td>13</td>
<td>HDPE-RH-25(coarse)</td>
<td>74.5</td>
<td>0.5</td>
</tr>
<tr>
<td>14</td>
<td>HDPE-RH-25(coarse)</td>
<td>74</td>
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<tr>
<td>15</td>
<td>HDPE-RH-25(coarse)</td>
<td>73.5</td>
<td>1.5</td>
</tr>
<tr>
<td>16</td>
<td>HDPE-RH-25(coarse)</td>
<td>73</td>
<td>2.0</td>
</tr>
</tbody>
</table>

* HDPE= high density polyethylene; RH= rice hulls; fine (10-30 mesh); coarse (30-50 mesh);
** coupling agent = Malic anhydride.

**Results and Discussion**

Figures (1-4) illustrates the characteristic (mechanical properties –rice hulls) curves on the studied fiberous reinforcement. It is observed that the mechanical properties such as tensile strength, flexural strength, hardness, and impact strength of HDPE-rice hulls (RH) composites
increased linearly with increase in fiber loading from 5 to 25%. An approximate increase of 39, 44, 223, and 60% in tensile, flexural, hardness and impact strength respectively (fine rice hulls) was observed with 30% rice hulls (RH) -filled composites as compared with the virgin matrix. A similar improvement in the mechanical properties where increase of 35, 34, 213 and 43% in tensile, flexural, hardness and impact strength respectively (coarse rice hulls). This behavior is primarily attributed to the reinforcing effect of the particles leading to a uniform stress distribution from the continuous polymer matrix to the dispersed fiber phase. However, beyond 25% of fiber loading there was a notable reduction in mechanical properties of composites. This decrease at high fiber loading implied poor particle-matrix adhesion, which promoted micro crack formation at the interface as well as nonuniform stress transfer due to fiber agglomeration within the matrix [4]. On the other hand for high concentration (35% in weight), the polymer matrix was not continuously distributed and most rice hulls particles directly contacted one another, thus resulting in poor adhesion at the interface. This situation was improved when the amount of thermoplastic larger than (up 60%) that of rice hulls (RH) particles in weight, most particles were enveloped by the polymer matrix when polymer phase were abundant in composite, this was due that the total surface contact area of individual particles with the matrix was close to the maximum value, and the dispersion resistance reached the maximum value [13]. In all figures (1-4) shows that the fine rice hulls flour particle samples showed better in mechanical properties compared with coarse rice hulls flour particle, it is evident that this behavior is primarily attributed to improved fiber – matrix interfacial adhesion, which increases the friction between the polymer and the particles thereby resulting in mechanical properties [4]. On the other hand, the small diameter can not contain significantly laug cracks, so the material is much stronger [13]. The mechanical properties of the rice hulls (25% by weight fine and 25% by weight coarse)-reinforced high density polyethylene HDPE composites, as a function of maleic anhydride MA weight are shown figures (5-12). From all figures, it is evident that the mechanical properties of the composite increase with increase the coupling agent (MA) from 0.5-1.5%. this mainly due to the reaction of (MA) with radicals for med by the functionalisation of HDPE leading to the grafting (of MA) on to HDPE. On the other hand the rice hulls (RH) was randomly distributed and separated in a continuous thermoplastic matrix. It was encapsulated or enveloped by the thermoplastic matrix mainly with mechanical connection, without the coupling treatment, the interfacial region was weakly linked, under loading, composite were mainly damaged along the loose and weak interfacial connections between rice hulls particles and thermoplastics and followed a cohesive mode. For composites with coupling treatment, most rice hulls particles were combined with thermoplastics through covalent bonding or strong interfacial bounding and the interface was strengthened with coupling agents [14]. The composites prepared using 1.5% MA by weight exhibited optimum mechanical properties. However, with a further increase in MA weight from 1.5-2%, a marginal decrease in mechanical properties was observed, this behavior may be attributed to the migration of excess MA around the particles, causing self-entanglement among themselves rather than the polymer matrix resulting in slippage [13].
Fig. (1): Influence of rice hulls (rh) particle weight percentage on tensile strength of rh fiber – hdpe composites.

Fig. (2): Influence of rice hulls (rh) particle weight percentage on flexural strength of rh fiber – hdpe composites

Fig. (3): Influence of rice hulls (rh) particle weight percentage on brinell hardness of rh fiber – hdpe composites

Fig. (4): Influence of rice hulls (rh) particle weight percentage on impact strength of rh fiber – hdpe composites
Fig. (5): Influence of maliec anhydrid (ma) coupling agent weight percentage on tensile strength of rice hulls (25% fine) particle – hdpe composites.

Fig. (6): Influence of maliec anhydrid (ma) coupling agent weight percentage on flexural strength of rice hulls (25% fine) particle – hdpe composites

Fig. (7): Influence of maliec anhydrid (ma) coupling agent weight percentage on brinell hardness of rice hulls (25% fine) particle – hdpe composites

Fig. (8): Influence of maliec anhydrid (ma) coupling agent weight percentage on impact strength of rice hulls (25% fine) particle – hdpe composites
Fig. (9): Influence of maliec anhydrid (ma) coupling agent weight percentage on tensile strength of rice hulls (25% coarse) particle – hdpe composites

Fig. (10): Influence of maliec anhydrid (ma) coupling agent weight percentage on flexural strength of rice hulls (25% coarse) particle – hdpe composites

Fig. (11): Influence of Maliec Anhydrid (MA) Coupling Agent Weight Percentage on Brinell Hardness of Rice Hulls (25% coarse) Fiber – HDPE Composites.

Fig. (12): Influence of maliec anhydrid (ma) coupling agent weight percentage on impact strength of rice hulls (25% coarse) particle – hdpe composites

**Conclusion:**
I. The overall trend shows a marked increase in a mechanical properties of composites [HDPE-RH] with increasing of rice hulls (RH) content (10-30% wt.).
II. Coupled composites have higher mechanical properties values than did uncoupled composites. This was attributed to the improved interface quality due to the presence of the compatibilizer.
III. Low cost of rice hulls (RH) particle can be used as an effective reinforcement with a fiber loading of 25% to prepare high performance RH-HDPE composites.
IV. Generally the composites content rice hulls particle fine [10-30 mesh] introduced mechanical properties better than composites content rice hulls particle coarse [30-50 mesh].
References: