Investigation of Tensile and Impact of Composite Materials Reinforced with Natural Materials

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Abstract
This work focuses on the study the effect of Data Seeds (DS) and Olive Seeds (OS) particles on tensile and impact properties of epoxy resin. Olive and dates seeds powder were added to epoxy matrix at weight fraction (0, 8, 13, & 18 wt%) with grain size (300, 450 & 600µm). The composite specimens were prepared by Hand-Lay up technique and cut according to standard test. It's found the higher value of modulus of elasticity and tensile strength happened at (=18 wt %) and grain size (300µm) for specimens reinforced with olive seeds powder, but obtained the lower percentage of elongation at break, impact strength and fracture toughness at this value. Also the mathematical model results show that the weight fraction of powder have higher effect than grain size on properties.

Keywords:- Composite materials, natural materials, mechanical properties, mathematical model.
A composite material is a macroscopic combination of two or more distinct materials, having a recognizable interface between them. Composites are used not only for their structural properties, but also for electrical, thermal, tribological, and environmental applications. Modern composite materials are usually optimized to achieve a particular balance of properties for a given range of applications. However, as a common practical definition, composite materials may be restricted to emphasize those materials that contain a continuous matrix constituent that binds together and provides form to an array of a stronger, stiffer reinforcement constituent. The resulting composite material has a balance of structural properties that is superior to either constituent material alone [1].

There has been an expanding search for new materials with high performance at affordable costs in recent years. In the last decade, a growing interest in the use of vegetable reinforcements in the development of composite materials has gained momentum. Lignocellulosic reinforcements present interesting features such as low cost, low density, and besides they are biodegradable and non-abrasive [2].

The widespread use of plastic materials has created severe environmental, economic, social, and political problems. The availability of landfill space has decreased rapidly and the cost of landfiling plastic wastes has increased enormously. The usage of epoxy resin as a matrix was chosen because it is the standard economic resin commonly used, preferred material in industry and besides, it yields highly rigid products[3]. The other method to tackle this problem is to use natural materials as the reinforcements in polymers [4].

The 1990s witnessed a burgeoning interest in the development of industrial and consumer products that combine lignocellulosic materials and plastic[5].

Olive seeds powder, a lignocellulosic material, is currently used mainly as energy source. Recently, it can be found in the literature references to its use as a raw material to produce activated carbon and furfural and as heavy metal biosorbent. Steam-explosion was also applied to this material in order to separate the main components: cellulose, hemicellulose and lignin [6].

These materials form thus inexpensive “new or secondary resources”, which could make them more valuable for wider utilization. When such materials are used in composites, developing countries, which produce these, become part of global composite industry as developer and manufacturer leading to increasing revenues and the creation of jobs [7&8].

**Abu-Sharkh and et al in (2004)** have studied the stability of the composites made from date palm leaves were compounded with polypropylene (PP) and UV stabilizers in natural weathering conditions of Saudi Arabia and in accelerated weathering conditions was investigated. The composites were found to be much more stable than PP under the severe natural weathering conditions of Saudi Arabia and in accelerated weathering trials [9].

**Jimenez and et al in (2008)** have illustrated a composite material using unsaturated commercial polyester resin (UPE) and olive brush seed (OBS) was prepared. OBS was treated with sodium hydroxide and maleicanhydride (MAN) and subsequently utilized in a proportion of 35 wt% to prepare a composite material. These materials were evaluated in terms of moisture absorption, surface density, and mechanical properties such as flexure and tensile tests [10].
Matosa and et al in (2010) have studied the development of polymer from abundant and renewable biomass resources constitute an attractive choice for polyurethanes and other industrial sectors, where green chemistry and sustainable processes are in focus. Also focus in this study establish the feasibility of converting olive stone (OS) residues by means of both total and partial oxypropylation. The results obtained demonstrated the viability of using this reaction to generate OS-based polymer, thus opening new perspectives for the exploitation of this by-product of the olive oil industry [11].

Qutaiba in (2011) has studied of a new range of sustainable reinforced polymer composite materials using powdered olive pits as a novel filler material to be used with epoxy resin. Also the influence of the untreated and treated powder loading (weight fraction) on the void content and the mechanical properties of the composites was examined. The results show significant improvements in mechanical properties for composites reinforced with treated olive pits than composites reinforced with untreated olive pits [12].

Rodrigo and et al in (2012) have studied Valorization of some lignocellulosic agro-industrial residues to obtain biopolyols, The valorization of renewable and abundant resources (date seed, olive stone, corncob, rapeseed cake and apple pomace) from agro-industrial activities was performed by mild liquefaction using polyhydric alcohols to obtain biopolyols that constitute an attractive choice for polyurethanes and other industrial sectors. The results obtained demonstrated the viability of using polyhydric alcohol liquefaction to generate bio-based polyols, thus opening new avenues of exploitation of these by-products. [13].

Koutsomitopoulou and et al in (2013) have illustrated the effect of olive pits powder reinforced PLA-matrix. This study is focused on recycling potential of some waste materials, such as olive pits, i.e. the solid phase derived from an olive oil mill, blended with thermoplastic polymers and used for the production of new materials applied in manufacturing containers and formworks. The olive pit powders are described and characterized. Then the powder is introduced in a bio-based and biodegradable matrix (polylactic acid, PLA) at various percentages [14].

Hamma and et al in (2013) have studied the effects of date stone flour (DSF) on morphology, thermal, and mechanical properties of polypropylene (PP) composites in the absence and presence of ethylene-butyl acrylate-glycidyl methacrylate (EBAGMA) used as the compatibilizer. The study showed through scanning electron microscopy analysis that (EBAGMA) compatibilizer improved the dispersion and the wettability of DSF in the PP matrix. Thermogravimetric analysis (TGA) indicated a slight decrease in the decomposition temperature at onset (T onset) for all composite materials compared to PP matrix, whereas the thermal degradation rate was slower. Differential scanning calorimetry (DSC) data revealed that the melting temperature of PP in the composite materials remained almost unchanged. The nucleating effect of DSF was however reduced by the compatibilizer. Furthermore, the incorporation of DSF resulted in the increase of stiffness of the PP composites accompanied by a significant decrease in both the stress and strain at break. The addition of EBAGMA to PP/DSF composites improved significantly the ductility due to the elastomeric effect of EBAGMA [15]. The aim of this research was to study the influence of weight fraction of powder on mechanical properties of epoxy reinforced by olive and dates seeds.
Theoretical Analysis

The modulus of elasticity for the composite material determined in terms of the properties of the powders and the matrix and in terms of the relative volumes of powder and matrix. The upper bound modulus of elasticity is [16]:

$$E_{c(u)} = E_p V_p + E_m V_m$$  \hspace{1cm} (1)

Also the lower bound modulus of elasticity is:

$$E_{c(L)} = \frac{E_p E_m}{E_p V_m + E_m V_p}$$  \hspace{1cm} (2)

Where:

- $E_p$ and $E_m$ = Young modulus for powder and matrix respectively.
- $V_p$ and $V_m$ = volume fraction for powder and matrix respectively.

Impact resistance is calculated for samples from the following relationship [17]

$$G_c = \frac{U_c}{A}$$  \hspace{1cm} (3)

Where

- $G_c$: impact strength of material (J/m²).
- $U_c$: impact energy (J).
- $A$: cross-sectional area of specimen (m²).

Fracture toughness can be expressed as [18].

$$K_c = \sqrt{G_c E}$$  \hspace{1cm} (4)

Where:

- $K_c$: fracture toughness of material (MPa.m¹/²).
- $E$: elastic modulus of material (MPa).

Mathematical Model

Response surface method (RSM) is a collection of mathematical and statistical techniques that are useful for modeling, analysis, and optimizing the process in which the response of interest is influenced by several variables and the objective. In many engineering fields, there is a relationship between an output variable of interest $z$ and a set of controllable variables $\{x, y, \ldots, N_n\}$. In some systems, the nature of the relationship between $y$ and $x$ values might be known. Then, a model can be written in the form [19]:

$$z = f(x, y, \ldots, N) + \epsilon$$  \hspace{1cm} (5)

The variables $(x, y$ and $N)$ are independent variables where the response ($z$) depends on them. The experimental error term, denoted as $\epsilon$.

In this study, multiple polynomial (least square fitting) regression analysis is used to establish a mathematical model among the experimentally obtained parameters. Multiple regression analysis techniques are applied to relate the weight fraction and grain size of powder for two type of powder (dates seed and olive seed),
the best form of the relationship between the property, weight fraction and grain size of powder parameters is chosen in the form of [20]:-

$$z = a_0 + b \times x + c \times y + d \times x^2 + e \times y^2 + g \times x \times y \quad \cdots (6)$$

The analysis of variance (ANOVA) is also called coefficient of multiple determination referred to ($R^2$) measure the proportionate reduction of total variation in associated with use of the set of predictors in the model (is used to check the validity of the model) it is defined in terms of SST, SSR, and SSE as [21]:-

$$R^2 = \frac{SSR}{SST} = 1 - \frac{SSE}{SST} \quad \cdots (7)$$

Where:-
- SSE: the sum of squared error.
- SSR: the regression sum of squares.
- SST: the total correct sum of squares.

**Experimental Work**

Basically two main tasks were carried out to achieve the objectives of study represented by preparation of composite material by combining the epoxy with olive seeds and date seeds powder with different weight fraction of powder (0, 8, 13 & 18 wt%) separately and grain size (300, 450 & 600 µm). Then it was continued by performing the mechanical test carried out to determine the characteristics of the studied composite. The type of epoxy resin is (swiss cherr) provided from the Saudi Arabia Company in the form of transparent viscous liquid at room temperature. Epoxy and hardener used in this study in ratio of 3:1.

**Preparation of Composites**

The composite specimens were fabricated by using hand lay-up technique. Composites having different powders content were prepared by varying the type and weight two for olive seeds and dates seeds powders. In the first process of preparing the composite specimens’ preparation process is to set the weight and grain size of powders content in the composite. The amount of resin needed for each category of composite was calculated after that. Then the resin was mixed uniformly with hardener, the mixture was poured carefully into the moulds made of stainless steel and left in the mould for 24 hours. After the composites were fully dried, they were separated off from the moulds, and then put the specimens in oven at (55°C) for (1 hrs) [22].

Specimens are prepared after the composites are ready. The geometry of the specimens is set by referring to ASTM standard D-638 at room temperature [23].

**Tensile Test Results**

Tensile test is done by using universal testing machine type (LARYEE) with capacity (50 KN) applied load and strain rate (0.5 mm/min).

Izod impact test machine used for testing polymeric materials. This test is performed according to standard (ISO-180) at room temperature [24].

**Results & Discussions**

**Tensile Test Results**

Figures (1, 2, 3, 4, 5 & 6) show the stress-strain curves for composite specimens. It is clearly from these figures it is clearly evident that characteristic
stress-strain results are trend growing from linearity and eventually develops into nonlinear. The stress and tensile strength increases with increasing weight fraction, the reason behind such behavior is explained as due to the nature of bonding force between the matrix and powder which is strong bonding that does not allow forming internal defects (cracks) in quick manner and in turn the composite material will have high tensile strength. Also, from these figures can be seen that the strain decrease with increasing weight fraction of powder, and the reason behind such behavior that due to reduction in the internal-particles spacing so the polymer chains may not have the full freedom of movement that they would have in the bulk polymer. This results agree with reference [25]. The best value of stress obtained when the composite reinforced with olive seeds and particle size (300µm) with weight fraction (18 wt%) is (45.6 MPa).

Figures (7, 8, 9 & 10) show the variation of young's modulus and tensile strength respectively for composite reinforced with olive and dates seeds powder. It is observed a progressive increase in young's modulus and tensile strength with increasing the amount of powder because of the powder particles exert the expected reinforcing effect leading to an increase in stiffness. Also from these figures can be seen that the value of modulus of elasticity and tensile strength decreases with increasing the particles size this may be attributed to weak bond between the particle and matrix and less interaction between them lead to formation the weak structures and may be due to non-wetting of the powder particles with the matrix and also due to the non-uniform distribution of the particles. As compared the results between composite reinforced with olive and dates seeds its found the higher value of modulus of elasticity and tensile strength in reinforced with olive seeds powder and particle size (300µm) with value (2700 MPa &45.5 MPa) respectively.

Figures (11 & 12) show the relationship between the percentage elongation at break value for composite specimens. It can be seen from these figures the value of percentage elongation decreases with increasing weight fraction of powder and increase with increase particle size. This is because of increasing the weight fraction, which will act as localized stress concentration regions, so the percentage elongation at break will be decreased. Also percentage elongation properties decreased with the addition of powder indicating interference by the powder in the mobility or deformability of the matrix and in turn lead to strain reduction with increasing weight fraction.

Also from these figures can be seen the percentage elongation for all composite specimens increase with increasing particle size. This may be attributed the large particles have low surface area and weak interface between particles and polymer.

Impact Test Results

Figures (13, 14, 15 & 16) show the variation between the impact strength and fracture toughness with weight fraction of the olive and dates seeds powder. Results had revealed that the impact strength decreases with increasing weight fraction of powder particles for all groups of composite materials compared to the impact strength of the matrix. This is because of the powder particles, which may represent points for a localized stress concentration, from which the failure will begin, or this is mainly due to the reduction of elasticity of material due to powder addition and thereby reducing the deformability of matrix and in turn the ductility in the skin area.
Also, an increase in concentration of powder reduces the ability of matrix to absorb energy and thereby reducing the toughness, so impact energy decreases [26].

Also this result for all specimens is attributed to lower powder dispersion and poor powder-matrix interaction and is consistent with morphological olive and dates seeds. The formation of agglomerates and other defects are responsible for the mechanical failure of the composite materials through propagation of the micro cracks during impact acting as stress concentration [27&28].

Mathematical Model Results

The experimental results are modeled using RSM. Table (1&2) show the summary of models and coefficient multiple determinations ($R^2$) of the properties ($z$) as function of ($x =$ weight fraction of powder) and ($y =$ particle size). It can be seen from these models that the weight fraction of powder have greater effect than the particle size on the properties.

Conclusions

The main conclusion of result was:-
1. The composite specimens reinforced with olive seeds powder gives high mechanical than composites specimens reinforced with dates seeds powder.
2. Higher value of modulus of elasticity and tensile strength were obtained at ($=18 \text{ wt\%}$) and grain size ($=300\mu\text{m}$) for two types of powder.
3. The value of percentage elongation, impact strength and fracture toughness decreases with increasing weight fraction and grain size of powder.
4. Mathematical model results show that the weight fraction of powder have higher effect than grain size on properties.

Table (1): Results for mathematical model for epoxy reinforced by olive seeds powder.

<table>
<thead>
<tr>
<th>property</th>
<th>Mathematical model and coefficient of multiple determination</th>
<th>Figure</th>
</tr>
</thead>
<tbody>
<tr>
<td>Modulus of elasticity</td>
<td>$Z=3819.28+203.473\times x+(-13.3813)\times y+(-1.296)\times x^2+(0.0137)\times y^2+(-0.274)\times x\times y$</td>
<td>$R^2 = 0.9687$</td>
</tr>
<tr>
<td>Tensile strength</td>
<td>$Z=27.247+(1.713)\times x+(-0.0358)\times y+(0.0252)\times x^2+(0.00036)\times y^2+(-0.00245)\times x\times y$</td>
<td>$R^2 = 0.996$</td>
</tr>
<tr>
<td>Percentage elongation</td>
<td>$Z= 8.2774+(-0.77)\times x+(0.00383)\times y+(0.0174)\times x^2+(-0.0000041)\times y^2+(0.0000325)\times x\times y$</td>
<td>$R^2 = 0.997$</td>
</tr>
</tbody>
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</thead>
<tbody>
<tr>
<td><strong>Impact strength</strong></td>
<td>$Z = 16394 + (-1914.3) * x + (4.233) * y + (43.905) * x^2 + (-0.0106) * y^2 + (1.4532) * x * y$</td>
<td>$R^2 = 0.995$</td>
</tr>
<tr>
<td><strong>Fracture toughness</strong></td>
<td>$Z = 4076.083 + (-5.94) * x + (-1.0063) * y + (1.364) * x^2 + (-0.00021) * y^2 + (-0.0876) * x * y$</td>
<td>$R^2 = 0.796$</td>
</tr>
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Table (2): Results for mathematical model for epoxy reinforced by dates seeds powders

<table>
<thead>
<tr>
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<th>Mathematical model and coefficient of multiple determination</th>
<th>Figure</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Modulus of elasticity</strong></td>
<td>$Z = 80.0898 + (86.358) * x + (3.1783) * y + (1.2327) * x^2 + (-0.0033) * y^2 + (-0.17415) * x * y$</td>
<td>$R^2 = 0.9817$</td>
</tr>
<tr>
<td><strong>Tensile strength</strong></td>
<td>$Z = 26.482 + (0.7267) * x + (-0.0335) * y + (0.0172) * x^2 + (0.0000353) * y^2 + (-0.00133) * x * y$</td>
<td>$R^2 = 0.985$</td>
</tr>
<tr>
<td><strong>Percentage elongation</strong></td>
<td>$Z = 9.831 + (-0.6284) * x + (-0.00702) * y + (0.0139) * x^2 + (0.0000131) * y^2 + (0.004052) * x * y$</td>
<td>$R^2 = 0.989$</td>
</tr>
<tr>
<td><strong>Impact strength</strong></td>
<td>$Z = 17622.1 + (-1701.603) * x + (-6.5927) * y + (48.0603) * x^2 + (0.00814) * y^2 + (0.5629) * x * y$</td>
<td>$R^2 = 0.9928$</td>
</tr>
<tr>
<td><strong>Fracture toughness</strong></td>
<td>$Z = 3519.737 + (-120.534) * x + (0.1521) * y + (5.2944) * x^2 + (-0.000051) * y^2 + (-0.0749) * x * y$</td>
<td>$R^2 = 0.994$</td>
</tr>
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</table>
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Figure (1): Stress-strain curve for composite reinforced with olive seeds powder & grain size (300µm).

Figure (2): Stress-strain curve for composite reinforced with olive seeds powder & grain size (450µm).

Figure (3): Stress-strain curve for composite reinforced with olive seeds powder & grain size (600µm).
Figure (4):- Stress-strain curve for composite reinforced with dates seeds powder & grain size (300µm).

Figure (5):- Stress-strain curve for composite reinforced with dates seeds powder & grain size (450µm).

Figure (6):- Stress-strain curve for composite reinforced with dates seeds powder & grain size (600µm).
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Figure (7):- The relationship between modulus of elasticity & wt% for composite reinforced with olive seeds powder.

Figure (8):- The relationship between modulus of elasticity & wt% for composite reinforced with dates seeds powder.

Figure (9):- The relationship between tensile strength & wt% for composite reinforced with olive seeds powder.
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Figure (10):- The relationship between tensile strength & wt% for composite reinforced with dates seeds powder.

Figure (11):- The relationship between % elongation & wt% for composite reinforced with olive seeds powder.

Figure (12):- The relationship between % elongation & wt% for composite reinforced with dates seeds powder.
Figure (13):- The relationship between Impact strength & wt% for composite reinforced with olive seeds powder.

Figure (14):- The relationship between Impact strength & wt% for composite reinforced with dates seeds powder.

Figure (15):- The relationship between fracture toughness & wt% for composite reinforced with olive seeds powder.
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Figure (16):- The relationship between fracture toughness & wt% for composite reinforced with dates seeds powder.

References
[28] Law, T. T. & Mohd Ishak, Z. A. J., "Water Absorption and Dimensional Stability of Short Kenaf Fiber-Filled Polypropylene Composites Treated with Maleated