Properties of silica reinforced unsaturated polyester composite: The effect of filler content

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Abstract. The effect of filler content on the composite properties of unsaturated polyester reinforced with silica filler has been carried out. The content of the silica filler used varied from 10 to 40%. Preparation of composites was carried out by the pressing method using compression molding at a pressure of 125 psi. From the results, it was found that the content of the silica filler was very influential on the properties and characteristics of the composite. Results have shown, the flexural strength decreased with increasing content of filler, while impact strength increased with optimum value in filler content 30% weight which was 11247 J/m². For the water absorption of composite, the increasing content of the filler led to the greater the percentage of composite water absorption. Meanwhile, the thermal stability of silica-filled unsaturated polyester composites was better than that of unsaturated polyester. The results showed that at a heating temperature of 470 °C unsaturated polyester was degraded by 90% while the composite filled with silica was degraded by 60%.

1. Introduction
Polymer matrix composite (PMC) is composite containing polymeric materials which include an embedded reinforcing phase, such as fiber or powder. Nowadays, PMC can be created with superior properties so that the material is very attractive in aviation, building construction, automobiles, sports equipment, furniture, and so on. The advantages of PMC materials include high impact strength and thermal resistance.

Unsaturated Polyester (UPR) is the most widely used thermosetting material because it has low shrinkage properties, can be molded at room temperature, low viscosity, good weather resistance and low cost [1]. However, unsaturated polyester has relatively fragile properties with low impact strength. This is due to the relatively high thermoset molecular weight. Specifically, one way to increase the strength of the material is by adding the dispersed phase in the matrix, so that when the crack starts between the dispersed phase (filler), the crack will elongate, not spread, and muffle which causes increased fracture resistance [2].

The filler material used in the composite matrix can be either organic or inorganic. Several studies on unsaturated polyester composites that use organic fillers have been widely reported. Consideration of using organic fillers in unsaturated polyester is because organic fillers are easy to obtain, and they also have good biodegradability. But in terms of mechanical properties, especially impact strength and also heat resistance and water absorption capacity, organic fillers still give a less satisfying effect. Therefore, the selection of inorganic materials such as silica is worth considering as fillers in unsaturated polyester [3].

Silica is a compound resulting from polymerization of silicic acid, which is composed of a unit chain of tetrahedral SiO₄ with the general formula SiO₂. Silica as a compound found in nature has crystalline structures. Silica has a large surface area and pore volume so it has the ability to absorb various
substances. In its use, silica can increase the mechanical strength of composites, because silica has the ability to absorb water and act as a hardener [4].

2. Method
2.1. Material
Composite preparation was carried out by compression molding method, where unsaturated polyester resin was respectively mixed with silica filler with filler variations of: 0, 10, 20, 30, and 40% of the total weight of the composite. The mixture was also being added with methyl ethyl ketone peroxide (MEKP) catalyst as much as 1.5% by weight of the matrix weight and molded in the mold according to the specified test.

2.2. Characterization
2.2.1. Flexural strength.
These samples were obtained in accordance with ASTM D-790 using instron machine.

2.2.2. Impact strength.
These samples were obtained in accordance with ASTM D-4812-11 using impact test machine.

2.2.3. Water absorption.
The aim is for calculating the amount of water absorption in the composite. At this stage, composite water absorption test was also carried out, where a sample of 5 x 5 cm was soaked in water at room temperature. The sample was removed every 6 hours, dried with tissue and weighed. The treatment was continued until the sample weight was constant. The percentage of water absorption was obtained by calculation:

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\text{\% Water Absorption} = \frac{w_2 - w_1}{w_1} \quad (1)
\]

where, \(w_1\) = weight of sample before soaking
\(w_2\) = weight of sample after soaking

2.2.4. Thermogravimetry analysis.
Thermogravimetry analysis was utilized by using Shimadzu Simultaneous TGA/DTA Analyzer DTG-60 to investigate the thermal characteristics of samples.

2.2.5. Scanning Electron Microscopy (SEM).
The analysed samples were the result of tensile strength test of polyester composite filled with silica with one of the contents that had the best properties among the four variables to observed the morphological changes that occurred in both composites.

3. Results and discussion
3.1 The effect of filler content on the flexural strength of silica filled polyester composites
Figure. 1 presents the effect of the filler content on the flexural strength of a silica filled polyester composite.
Figure 1. The Effect of filler content on the flexural strength of unsaturated polyester composites filled with silica.

The figure above shows that the presence of silica filler has reduced the flexural strength of composite materials. The decline continued from the start of filler addition with a content of 10% to 40%. This was attributed to the addition of the phases in the composite, namely the filler itself and the matrix. The flexural strength of composite is a measurement of the ability of a material against a given transverse load/force [5].

The decreasing flexural strength of a material was caused by the limited transfer of load from the matrix to the filler. Although structurally the silica filler has a pore that can absorb the matrix (unsaturated polyester), but in fact, the presence of the pore increased the contact surface area and as well increase the interface tension of the filler and matrix. This condition increased the failure rate of composite materials when subjected to transverse forces [6]. Unlike the case with unsaturated polyester material without filler, where phase uniformity made no interface tension. In this condition, when given a transverse force in the form of a flexural test, unsaturated polyester material showed better strength (101 MPa).

3.2 The effect of filler content on the impact strength of unsaturated polyester composites filled with silica

Figure 2 below presents the effect of the content of the filler on the impact strength of polyester composite filled with silica.

Figure 2. The effect of silica filler content on the impact strength of unsaturated polyester composites filled with silica.
The tendency of positive trend was obtained from the impact strength of composite materials filled with silica. The figure above shows that the presence of fillers has increased the impact strength of unsaturated polyester composites filled with silica. It can be seen that the impact strength of composite materials continued to increase in filler contents from 10% (5810 J/m$^2$) to 30% (11247 J/m$^2$). This was related to the impact test mechanism carried out during testing, fillers in the composite act as the former of the starting point of the crack formation and stress transferring medium, whereas the reinforcement mechanism can occur due to the localization of cracks which began with the concentration of stress (load) around the silica particles, followed by cavity growth in unsaturated polyester (matrix) [6]. In this case, silica fillers containing pores filled with unsaturated polyester matrix successfully absorbed higher impact energy. The impact strength of unsaturated polyester composites has decreased in the filler content of 40%. This is because the content of the filler is too large so that the matrix cannot wet the entire surface and pore of the filler. The unevenness of wetting caused the energy absorbed was also not uniform so that the impact strength decreases [7]. The positive trend shown in the tensile strength of unsaturated polyester composites filled with silica can be seen from the morphological characteristics of the break in Fig. 3. It shows the pores of the silica filler has been filled with matrix so that when the collision occurred, the energy can be absorbed by the filler properly.

![Figure 3. Morphology of the fractured surfaces of unsaturated polyester composites filled with modified silica.](image)

### 3.3 Effect of silica filler content and soaking time on composite water absorption

Figure 4 shows the percentage of water absorption of unsaturated polyester composites filled with silica at several soaking times. It can be observed that the longer the soaking time, the greater the percentage of composite water absorption. This happened for all silica filler content given. Furthermore, it shows the water absorption of the composite remained constant at 144 hours (6 days) of soaking time for each different content of silica. This is due to the decreased ability of the composite to absorb water until finally reaching the saturation point of water absorption (stop absorbing water) until 8 days of soaking (192 hours) [8].
Figure 4. The effect of silica filler content and soaking time on composite water absorption.

From the figure above, the relationship between the water absorption of composite and the content of silica filler can be seen that the greater the content of filler, the greater the percentage of water absorption for a certain time. This condition was related to the porous structure and water absorption ability of silica, so that the greater the silica content in the composite, the more pores were available. From the figure above, it can also be seen that at the filler content of 30%, water absorption was only slightly higher than that of the composite containing 20% filler. This shows that the porous absorption of the unsaturated polyester matrix was more optimal, so the free space was less available [9].

3.4 Thermal characteristic of composite

Fig. 5 below shows the thermal characteristic by using thermogravimetric analysis (TGA) for unsaturated polyester, silica, and unsaturated polyester composite filled with silica.

Figure 5. Changes in the mass of each component to the increase in temperature by using TGA.

For unsaturated polyester and composite (curve 1 & 2) it can be seen that the first weight reduction occurred at a temperature of 45 °C to a temperature of 300 °C with a percentage of about 10%. In this
phase the removal of water molecules and volatile materials happened [10]. Furthermore, for the second phase the weight reduction of UPR (1) occurred at a temperature of 300 °C to 450 °C with a percentage of about 15%. In this phase the degradation was caused by the decomposition of aromatic rings, the release of methane, CO and CO₂ [11]. Furthermore, above 450 °C the UPR has decomposed to ash. For silica (2), the decomposition process almost was not occurred because the melting point and boiling point of silica reached 1610 and 2230 °C, respectively. For unsaturated polyester composites filled with silica (3), it can be seen that the heat resistance was higher than that of unsaturated polyester materials. This can be seen from the position of the curve where at a temperature of about 470 °C the weight of unsaturated polyester without silica has been reduced by as much as 94%, while the composite filled with silica was degraded by only about 60% [12].

4. Conclusion
The content of silica fillers was very influential on the properties of unsaturated polyester composite. It shows flexural strength decreases with increasing filler content, while impact strength increases with an optimum value of 30% filler content. In addition, an increase in the filler content was also followed by a large percentage of the ability of the composite to absorb water. For thermal stability of materials, silica-filled composites have better heat stability than the unsaturated polyester itself.

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