Comparison of Silica and Zeolite as Fillers on Unsaturated Polyester Resin (UPR) Composites: The Effect on Tensile Properties

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Abstract: The effect of unsaturated polyester resin (UPR) composite filled with silica or zeolite on the tensile properties were investigated. The composites were prepared by hand-lay up process with the variations of filler content viz. 0, 10, 20, 30, and 40% by weight. The properties and characterization were carried out using tensile properties and scanning electron microscopy. The results showed in the lower content of filler the tensile properties of composite filled with zeolite were higher than filled with silica. The presence of pores and tunnels in zeolite structure have increased the addition of the matrix-filler interface. The results were also confirmed by image on morphology of the materials by scanning electron microscopy.

1 INTRODUCTION

Unsaturated polyester resin (UPR) is the most widely used thermoset material because it has low shrinkage properties, low viscosity, low cost, can be molded at room temperature, and good weather resistance (Prasad, Rao and Nagasrinivasulu, 2009; Deepa et al., 2011; Waigonkar et al., 2011; Ray and Rout, 2005). However, UPR tends to have relatively brittle properties with low impact strength due to the relatively high 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), it will be elongated, not spread, and muffled which cause increased fracture toughness (Jesson and Watts, 2012).

The filler used in the composite matrix can be organic or inorganic. Several studies on UPR composites using organic fillers have been widely reported (Gao et al, 2011; Du et al., 2010; Adenkunle et al., 2011; Khalil, Bhat and Sartika, 2010). Consideration of the use of organic fillers in UPR, among others, is because of the easily attainable organic filler and also the good biodegradation ability. However, in terms of mechanical properties, especially tensile strength, impact and also resistance to heat and water absorption, organic filler still gives less satisfactory effect. Therefore, the selection of inorganic materials such as silica and zeolite is feasible to be considered as fillers in UPR matrix.

Silica is a polymerization compound of silicic acid, composed of a tetrahedral SiO₄ chain with a general formula SiO₂. Silica as a compound found in nature has crystal structure. Silica has large surface area and pore volume, hence it has the ability to absorb various substances. In the usage, silica can increase the mechanical strength of composites, because silica has the ability to absorb water and as a hardener. On the other hand, zeolite is porous hydrated alumina silicate crystals mineral having a three-dimensional skeletal structure formed of tetrahedral [SiO₄]₄ and [AlO₄]₄ which form a network of tunnel and cavities or pores (Pecover, 1987). By the presence of the pores/cavities/tunnel structure, the wetting of fillers by the matrix is better and will result in more perfect interface interaction. The composite produced will not need any additives such as compatibilizer or coupling agent. The advantage of using zeolite as a filler in the preparation of composite material is that the matrix will be absorbed by zeolite through surface absorption process and trapped in the tunnel of zeolite structure, based on this absorption mechanism it is expected that the matrix can be bound in zeolite so that there is a good interaction between matrix and filler. From the above-stated, the research on the study of the addition of silica powder and zeolite in the preparation of UPR based composites...
2 METHODS

2.1 Materials

Materials unsaturated polyester resin (UPR) as matrix, silica and zeolite as fillers and methyl ethyl ketone peroxide (MEKP) as catalyst/hardener were supplied by chemical store and used as received.

2.2 Preparation of Composite Materials

Composites preparation were done using hand layup method, in which unsaturated polyester resin (UPR) was mixed with silica respectively with filler variations: 0, 10, 20, 30, and 40% of composite total weight. The mixtures were also added with methyl ethyl ketone peroxide (MEKP) catalyst of 1.5% by weight of the matrix weight and molded in the mold according to the predefined test. Similar preparations were also done for the UPR composite filled with zeolite.

2.3 Tensile Properties

Tensile test was performed refers to the standard ASTM D 638 Type IV using an instrument of Instron machine AI700. The results of tensile test were obtained in the form tensile strength and elongation at break.

2.3.1 Tensile Strength

Tensile strength is a measure of maximum load that a composite material can withstand without fraction. Tensile strength is measured in force per unit of cross-sectional area.

2.3.2 Elongation at Break

Elongation at break is the ratio between increased length and initial length after breakage of the tested composite. It measures how much bending and shaping a material can withstand without breaking.

2.4 Morphological Analysis

Scanning electron microscopy (SEM) analysis were performed to observe the morphological changes that occur in the unsaturated polyester resin (UPR), UPR filled silica composite as well as UPR filled zeolite composite. SEM analysis were done using SEM EVO MA 10 ZEISS.

3 RESULT AND DISCUSSION

3.1 The Effect of the Filler Composition on the Tensile Strength of the Unsaturated Polyester Composite Filled with Silica and Zeolite

Figure 1 below shows the effect of filler content on tensile strength of unsaturated polyester filled with silica and zeolite composite.

From the figure above it can be observed that in the addition of silica filler as much as 10%, the composite tensile strength decreased from 40.5 MPa to 22.7 MPa. The presence of silica as filler in a polyester matrix has created a new phase. This leads to increased matrix and filler interface tension. The small composition of silica has not dispersed evenly in the polyester matrix, and thus, its presence actually makes the composite become weak. However this changed in the addition of silica by 20% (26.6 MPa) and continued with 30% (33.4 MPa), where the tensile strength of composite materials was increased. In the addition of 20% followed with 30% silica, the polyester will be absorbed by the silica through surface absorption process. Silica has a large surface area and pore volume that has the ability to absorb the matrix. Based on this absorption mechanism, the matrix trapped in the pores of silica could be bound so that there is a good interaction between the matrix and the filler. However, the addition of 40% silica again reduced the tensile strength of composite materials to 27.6 MPa. This is due to the presence of a relatively large amount of fillers, so that not all
fillers could be moistened by the matrix perfectly and also the agglomeration potential of the filler. For zeolite filler, a continuously declining trend of tensile strength in composite materials was observed, where the addition of 10% - 40% of fillers has decreased the tensile strength properties from 36.3 MPa - 20.9 MPa. This shows that incorporating zeolite filler to polyester matrix has not been able to increase composite tensile strength. The presence of zeolite fillers having a hollow and tunnel-like structure is likely to have caused a large number of interface phases that weaken the interaction between matrix and filler. This weak interaction caused high interface tension of matrix and filler as well as weakening the attachment between the two phases. However, in addition to the 10% and 20% fillers, the tensile strength value exhibited by the zeolite-filled polyester composite was better than the silica filled. This indicates that the presence of a relatively small amount of zeolite filler was more effective than the silica filler. The presence of pores and tunnels within the zeolite filler structure in optimum amount (10-20%) gave a better effect on the tensile properties of the composite material than the pore structure in the silica filler.

3.2 The Effect of Filler Composition on Elongation at Break of the Unsaturated Polyester Resin Composite (UPR) Filled with Silica and Zeolite

Figure 2 below shows the effect of the filler content on the elongation at break of unsaturated polyester composite filled with silica as well as zeolite.

![Graph showing the effect of filler content on elongation at break](image)

The figure above shows that both the silica and zeolite filler have lower the elongation at break of the composite material. The decrease occurs for each addition of the filler composition ranging from 10% to 40%. This shows that the presence of such fillers has lowered the elasticity of the polyester matrix. In this case, the interaction between the polyester matrix with the silica and zeolite fillers has resulted in a more limited movement of the polymer chain causing the elongation at break in the resulting composite to decrease. Furthermore, from the figure can also be observed that the composite materials filled with zeolite were more elastic than those containing silica filler for the addition of 10-20% filler. In this case, the tunnel and pores present in the silica structure and absorbing the matrix can still maintain the elasticity of the composite material.

3.3 The Morphological Characteristics of Unsaturated Polymers Resin (UPR) and UPR Composites Filled with Silica and UPR Composites Filled with Zeolite

The images of surface morphology of unsaturated polyester resin (UPR) and UPR composites filled with silica as well as UPR composite filled with zeolite can be seen in Figure 3 below.

![Image showing surface morphology](image)
Figure 3: Characteristics of surface morphology (a) unsaturated polyester resin (UPR); (b) UPR filled with silica; (c) UPR filled with zeolite.

The figure above shows the surface morphology of unsaturated polyester resin (UPR), UPR composite filled with silica as well as filled with zeolite. In the figure of UPR (Fig. 3a) the surface image appeared to have rigid structure, some empty fractions (voids), and brittle. Whereas for the morphology of UPR composite filled with silica (Fig. 3b) indicated that the matrix has been absorbed into the silica filler. However, in some regions it was observed that the fillers were localized, marked with uneven clumps. Compared with the morphological characteristics of the UPR (Fig. 3a), it can be observed that the presence of the silica filler in the unsaturated polyester matrix (Fig 3b) has increased the matrix-filler interface tension. This causes the lower composite tensile strength than that of a UPR. Meanwhile, for the zeolite-filled composite (Fig 3c) it appeared that an UPR matrix has also been absorbed into the zeolite filler. However, when compared to the silica-filled composite (Fig 3b), this composite showed a slightly different morphology. The surface was observed to contain tunnels that came from the filler structure, and dispersion of the filler were observed to be more uniform. This showed that the porous and tunnel form zeolite filler structure was more compatible in the matrix absorption, so that the surface tension between the matrix-filler can be reduced.

4 CONCLUSION

Unsaturated polyester resin (UPR) composite filled with silica as well as zeolite still showed a lower tensile strength value compared to polyester material. Meanwhile, the most optimum silica filler composition was 30% by weight, while for the zeolite filler was 10% by weight. In lower composition, the ability of zeolite as filler was better than silica. In this case, this showed that the porous zeolite structure and the tunnel form were more compatible in the matrix absorption, so that the surface tension between the matrix-filler can be reduced.

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