Properties of Unsaturated Polyester Composite Filled Activated Zeolite: The Effect of Filler Addition and Compression

H Nasution, D M Putra, M T Al Fath
Department of Chemical Engineering, Faculty of Engineering, Universitas Sumatera Utara, Padang Bulan, Medan 20155, Indonesia.
E-mail: h_dahliana@yahoo.com

Abstract. The mechanical and physical properties of polyester composites filled with activated zeolites as fillers were investigated. Zeolite is an alumina silicate mineral that has cavity structures. The ability of the cavity structure can be maximized by activation using hydrochloric acid (HCl) 2 M. To achieve this goal, the zeolite was activated and proved it by performing morphological analysis using Scanning Electron Microscope (SEM) to see the surface structure. The aim of this study is to determine the effect of the activated zeolites content and hot press compression on the tensile and water absorption properties of polyester composites. The best tensile properties were found in ratio of unsaturated polyester with zeolite 70:30 (w:w) at a pressure of 125 Psi with a tensile strength of 38.51 MPa. SEM characterization showed a more rough breaking surface, that is in the ratio of polyester with zeolite 60:40 (w:w) at 75 Psi pressure because of the lack wetting between the matrix and activated zeolite. The water absorption of composite has increased as the addition of activated zeolite.

1. Introduction
It has been known that composite is a material composed of a mixture of two or more materials with different chemical and physical properties, and produces a new material that has different properties with its constituent materials. Composite material consisting of polymer resin as matrix and filler as reinforcement. Commonly, the addition of fillers is useful for increasing mechanical properties [1]. Zeolite is one of the minerals found in Indonesia with almost pure shape and has a relatively low price, but its utilization has not been maximized.

In general, zeolite is a natural material that is widely found around the fire mountain. The first zeolite was discovered by Baron Axel in 1756 at nature [2]. Zeolite has a porous micro-porous of aluminosilicate with a crystalline structure in some interconnected cavities [3,4]. Micro porous of zeolite aluminosilicate is composed of the three-dimensional main building of tetrahedral SiO4 and AlO4, pores are widely used because they have high absorption ability, in terms of absorbing water and other polar molecules [5]. The ability of zeolites can be maximized by the activation process. In general, there are three activation processes which are physically heated, chemical activation with acid and chemical activation with bases [6]. Activation of zeolites using HCl solution at various concentrations and temperatures gives the result that activation with HCl causes the zeolites dealumination process [7,8]. Also HCl has ability to dissolve undesired compounds and to exchange
cations in the frame of zeolite. The higher the concentration of acid used, the smaller the adsorption power of zeolite on water. Reports have shown by Ola, et. al. (2013) with activation of zeolites using HCl was better in absorption [9].

The use of zeolite as a filler has been done by researchers, especially to see the ability of zeolite cavities. Therefore researchers want to see the effect of zeolite as a filler in mechanical properties. Based on the explanation above, researchers intested in conducting research with the aim to increase the benefits of zeolite, researchers will research effect of filler addition and hot press compression on composite polyester filled activated zeolite.

2. Method

2.1 Material

The materials used were unsaturated polyester mixed with methyl ethyl Ketone Peroxide (MEKPO) as a catalyst obtained as received. Zeolite used derives from Sukabumi, Indonesia. Zeolite was activated using HCl 2M solution.

2.2 Composite Matrix Preparation

Unsaturated polyester resin was stirred with 1% MEKPO catalyst of the resin. The mixture is stirred for 10-15 minutes until evenly mixed.

2.3 Composite Filler Preparation

Activated zeolite was prepared to be sieved with a 110 mesh sieve, washed with aquadest and then filtered with filter paper and dried with an oven. The main process to make composite is shown in flowsheet below.

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Prepare the materials (zeolite, UPR, HCl, MEKPO)

Zeolite was activated by HCl

Zeolite was heated and dried at furnace

Activated zeolite was mixed by UPR and catalyst

Pour to the mold and cured by hot press

The dry composite was analyzed
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**Figure 1.** Flowsheet of Main Process.

2.4 Zeolite Activation

2 M hydrochloric acid solution was prepared and mixed with zeolite by comparison of zeolite:HCl, 1:10 (w:v). Stirred for 2 hours at 50 rpm and then the hydrochloric acid solution was separated from zeolite with filter paper. Zeolites were then washed with distilled water until the pH of the sample is neutral using litmus paper. Activated zeolite was dried in oven for 2 hours continued with a furnace at 600 °C for 1 hour. Processes of zeolite activation were shown in figures below.
2.5 Composite Casting Process

The unsaturated polyester (matrix) and activated zeolite (filler) with a ratio of 100:0; 90:10; 80:20: 70:30; 60:40% (w/w) were prepared by mixing in a container manually. Each ratio of the mixture was then poured in the mold and press using hot press of 75; 100 and 125 Psi for 8 minutes. After that, the dry composite is removed from the mold and casting repeated up to 3 repetitions. The composite product and hot press machine were shown in figure 3(a) and 3(b).

2.6. Tensile Test

The specimens of the composites were selected and cut to form a specimen for tensile strength test ASTM 638. Tensile strength test was performed using a tensometer on each specimen with the thickness of 4 mm. The tensometer was first conditioned at a load of 100 kgf at a speed of 30 mm/min, and then clamped firmly. The engine is turned on and the specimen will be pulled up, until the observed specimen breaks, the maximum stresspoint will be appear.
2.7 Impact Test

Impact Strength was investigated by Standard of ASTM D 4812 -11 with Unnotched Izod method using Impact Test Machine.

2.8 Water Absorption test

Water absorption characteristics of unsaturated polyester and polyester composites are tested by soaking in water at room temperature every 24 hours until the composite material no longer absorbs water (saturated). At each time period of immersion, the sample is taken and cleaned with tissue paper to absorb water. The sample is then weighed and calculated by the equation:

\[ W_a(\%) = \frac{W_e - W_o}{W_e} \times 100\% \]  

Notes:

\( W_a \) = Water absorption percentage of composite  
\( W_e \) = Composite weight after immersion  
\( W_o \) = Composite weight before immersion

2.9 Scanning Electron Microscope (SEM)

The samples analyzed were the results of the tensile strength test of polyester composite filled with activated zeolite filler with one of the compositions having the best properties among variables to observe morphological changes that occurred in both composites.

3. Result and Discussion

3.1 The Effect of Activated Zeolite and Hot Press Compression on the Tensile Strength Composite Polyester

Fig 1. shows the effect of activated zeolite and hot press compression on the tensile strength composite polyester.

Figure 4 shows that the higher of activated zeolite and hot press compression additions, the higher of tensile strength values until reach an optimum value at ratio polyester : activated zeolite 70:30 (w/w) and 125 Psi pressure hot press with a value 38.51 Mpa. It was indicated that the activated zeolite was evenly distributed in matrix unstaurated polyster causing and increase the tensile strength of composite [10,11]. However, there is a slight reduce tensile strength’s value at the ratio polyester:
activated zeolite of 60:40 (w/w) with the same pressure. It was indicated that the activated zeolite was not spread homogenous, so there is no good wetting happen among matrix and filler [12]. The tensile strength values are also affected by the variations of hot press compression, where the higher of hot press compression will increase the tensile strength of composite. This is due to the pressure that was applied to the composite where the chance of the matrix entering activated natural zeolite cavities was easier to occur. These results are supported by scanning electron microscope (SEM) in Figure 5 below.

**Figure 5.** SEM Image of Fractured Samples.  
(a) Activated Zeolite Polyester Composite at a ratio of 70:30, 125 Psi Pressure  
(b) Activated Zeolite Polyester Composite at ratio of 60:40, 75 Psi Pressure

### 3.2 The Effect of Activated Zeolite and Hot press compression on the Impact Strength Composite Polyester

Fig 6. shows the effect of activated zeolite and hot press compression on the impact strength composite polyester.

**Figure 6.** The Effect of Addition of Activated Zeolite Fillers and Hot Press Compression on the Impact Strength of Polyester Composite.

The purpose of impact strength is to measure how much the energy that can be absorbed by sample before it fractures. Figure 3 shows that the addition of filler and hot press compression will increase the value of the composite impact strength until reach an optimum value at ratio polyester : activated zeolite of 70:30 (w/w) and 125 Psi hot press pressure with a value of 8.89 J/m$^2$. It can be indicated the
role of fillers in improving the composite impact resistance, in this case the filler interacts with crack formation and stress transfer medium [13].

At ratio polyester : activated zeolite 60:40 the impact strength was decrease with a value of 7.19 J/m². It can be indicated that fillers were not distributed properly when added more. The composition of activated natural zeolite fillers which exceed the optimum limit will result in wetting not going well in certain areas that will decrease the impact strength value. The impact strength was continue decreased as the filler content increased which makes the concentration of the area around the filler decrease [14]. On the other hand, it can be seen that the effect of hot press compression on the impact strength values have showed that the the higher of hot compression will increase the impact strength of composite. The hot press compression additions make zeolite cavities more easily filled with unsaturated polyester where the composite gets denser and requires more energy to destroy it. According research by Younesi and Bahrololoom (2009), that the higher of hot press compression will increase the value of mechanical properties [15].

3.3 The Effect of Activated Zeolite and Hot Press Compression on Water Absorption Composite Polyester

Fig. 7 shows the effect of activated zeolite and hot press compression on water absorption composite polyester.

![Figure 7](image_url)

**Figure 7.** The Effect of Activated Zeolite and Hot Press Compression on Water Absorption Composite Polyester.

Figure 7 shows that the higher of zeolite and hot press compression, the higher of water absorption values until reach the optimum value at ratio polyester : activated zeolite 60:40 (w/w) and 75 Psi pressure with a value of 5.51 %. It can be concluded that zeolite which is one of the most widely used adsorbents where the structure of the zeolite itself which has empty cavities that have shown in Figure 5. Figure 7 shows that the addition of hot press compression will decrease the water absorption value where the increasing of the pressure could reduce voids and increase the interface bond between the matrix and filler that will increase the density of composite. It can be affected the percentage of composite water absorption decreases with the hot press compression addition [15].

4. Conclusion

The activated zeolite was be one of good reinforcement to composite as filler. The optimum tensile and impact strength was obtained at 125 Psi where the addition of activated zeolite would increase tensile and impact strength. Meanwhile, contrary with the mechanic test, the water absorption was decrease along with hot press compression addition.
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