

## MORPHOLOGICAL AND MECHANICAL PROPERTIES OF MT. KELUD VOLCANIC ASH REINFORCED POLYESTER AND EPOXY COMPOSITES

**Henny Pratiwi**

Sekolah Tinggi Teknologi Adisutjipto, Yogyakarta

E-mail: henny\_pratiwi@stta.ac.id

**Abstract** -- *The use of suitable waste products as raw materials has become an interesting matter in composite industry nowadays due to the environmental issues. Volcanic ash is one of the waste materials containing a high number of silica. The aim of this study is to examine the morphological and mechanical properties of Mt. Kelud volcanic ash reinforced polyester and epoxy composites. The volcano ash was dried and sieved into 50 mesh then mixed with polyester or epoxy manually for 10 minutes. The ash added into the matrix was varied by 0%, 10%, 20%, 30% and 40% from matrix volume content. For epoxy matrix, the composite with 40 vol. % particles has the highest tensile strength. However, for the polyester/ash composites, the tensile strength continues to decrease with the addition of particles. There is a significant increasing of 47.04 % for polyester and 5.62 % for epoxy in impact strength when 40 vol. % of volcanic ash added into both polymers. The Scanning Electron Microscopy result shows that there is void and agglomeration contained in epoxy/ash composites and crack propagation along the surface of polyester/ash composites that could be the cause of the failure.*

**Keywords:** *volcanic ash; composite; epoxy; polyester; mechanical properties*

### 1. INTRODUCTION

Nowadays, the use of particulate reinforced composites has been growing interest in many varying applications. The difficulties of the fabrication of fiber reinforced composites which have the high costs sometimes limit their use in many applications. Particulate reinforced composites can be thought as an alternative. Their advantage is the low cost and easily processed that could improve the stiffness, strength and fracture toughness. The addition of solid particles could improve the mechanical properties of the composite material, compared to the polymer material alone. Numerous studies have been published in recent years focusing on the use of silica and other particles as reinforcing agents in polymers [1-5].

Adhesive bonding is one of the most used techniques for joining materials lately. Adhesives can join a wide range of materials (polymers, ceramics, and metals) and the combinations of them [6], [7]. The adhesives are widely used in industry where epoxy resin is the most common structural adhesives used due to their good mechanical, thermal and chemical properties [6]. Because of the high modulus of elasticity and strength, low creep, and good thermal strength, epoxy microstructure is very useful for applications in structural engineering [7], [8]. Nevertheless, the structure of these thermoset polymers also causes brittleness, with a low resistance to the initiation of cracks and their propagation [9], [10]. Toughening of these adhesives has been widely studied in the past forty years, and nowadays represents a large field of scientific and technological concern [11-15]. There are some solutions available to improve the

mechanical properties of brittle adhesives, like the inclusion of particles, inorganic or organic [16], [17].

Since the 19<sup>th</sup> century some particles such as carbon black, silica, clay, etc. have been important reinforcing factors for polymers. Silica has attracted considerable attention recently due to the alternative non-black reinforcing filler that can be used to produce wide range of products with excellent control of vibrations and noise. Silica, however, is less compatible with some polymers and small-sized silica is difficult to disperse. Therefore, an understanding of reinforcing is critical to maximize the potential of silica as reinforcing filler and expand the applications of the polymers [18].

The disadvantage of thermosetting resins is their poor resistance to crack growth, but inorganic particles have been found to be effective in toughen the thermosetting resins. Although they do not increase the toughness as significantly as rubber particle inclusions, they increase the elastic modulus and hardness much better than rubber particles. In contrast, most studies on thermoplastics filled with rigid particulates reported a significant decrease of fracture toughness compared to the neat polymers. Enhancement of impact properties of some pseudo-ductile polymers by the introduction of inorganic particles has also been achieved.

A number of researches have evaluated the dependence of the mechanical properties of particulate reinforced polymer composites, including strength, toughness, rigidity, wear properties, etc., on the particle size and particle spatial distributions. Recently, a researcher [2] reviewed the effects of the particle size, particle/matrix interface adhesion and particle

loading on the stiffness, strength and toughness of particulate-polymer composites by comparing the published experimental results with theoretical models. They stated that the composite strength and toughness are strongly affected by all three factors: particle size, particle/matrix adhesion, and volume content. The mechanical properties of particulate reinforced elastomer composites are also affected by three factors. Since Hall-Petch [19] reported that the strength of metals and composites was dependent on  $d^{-1/2}$  (where  $d$  is the diameter of particles), many studies have been performed analytically and numerically to predict the strength of particulate reinforced polymer composites [20-22]. In general, hard particles in particulate reinforced polymer composites have two effects: weakening due to stress concentrations and reinforcing due to crack growth barriers. Even though in many studies, a prediction of the strength of these materials is difficult because of a range of parameters, such as interface adhesion, stress concentration and defects or cluster sizes, and their spatial distribution, etc., there is no universally accepted theory to predict the strength of particulate reinforced polymer composites.

Using suitable waste products as raw materials has become an interesting matter in composite industry recently due to the environmental issues, cost reduction and energy saving. Besides, the composition of these waste materials is similar with the natural raw materials when used in composite or ceramic applications [23]. Volcanic ash is one of the waste materials that containing a high number of silica and alumina [24]. Some researchers have reported about the improvement of the addition silica into polymers [25-27]. Therefore, the objective of the present study is to investigate the morphological and mechanical properties of Mt. Kelud volcanic ash reinforced polyester and epoxy composites.

## 2. EXPERIMENTAL METHOD

The volcano ash, which is from the eruption of Mt. Kelud, was dried and sieved into 50 mesh using vibrating screener at Mechanical Engineering Department Gadjah Mada University. The sieved volcanic ash then mixed with polyester or epoxy manually for 10 minutes. For polyester, the ash was blended with the curing catalyst, methyl ethyl ketone peroxide (MEKP), at a concentration of 0.01 w/w of the matrix, supplied by Sigma-Aldrich. For the other type of matrix, Epoxy resin Bakelite EPR 174 and hardener Versamid 140 were supplied by Sinar Kimia Corp., Indonesia with the ratio of 2:1. The mixing process was the same with polyester. The ash added into the matrix was varied by 0%, 10%, 20%, 30% and 40% from matrix vol. content. The mixtures then poured into the mold and were allowed to cure for 15 days at

room temperature inside the mold. The plates were kept in open air for 15 days to obtain a complete polymerization of the polymers. The specimens were then cut from the plates using a saw based. The dimensions of the manufactured plates are  $200 \times 200 \times 4$  mm.

The tensile specimens were evaluated using an ultimate tensile machine (Torse AMU-5-DE) according to the ASTM D638-91 standard at the specified loading rate of 10 mm/min. The composite specimens were tested to failure and, the average value of 5 tensile-tested specimens was reported for each sample. Fracture surfaces of tensile test specimens were examined by scanning electron microscope (JEOL 7800F SEM), operated at 5 kV, to investigate the effects of particles on fiber-matrix interfacial adhesion. The Charpy impact test was conducted to determine the impact resistance of material and the energy absorbed by the material during fracture according to the ASTM D790-02 standard.

## 3. RESULTS AND DISCUSSIONS

There are some studies that have been conducted on the mechanical properties of the particulate-filled polymer composites [16], [18]. Tensile and impact strength can be improved by adding either micro- or nano-particles since rigid inorganic particles generally have a much higher stiffness than polymer matrices. The mechanical properties depend on the particle geometry, shape, size distribution and porosity, as well as on the nature of the polymer/filler interaction. However, strength strongly depends on the stress transfer between the particles and the matrix. For well-bonded particles, the applied stress can be effectively transferred to the particles from the matrix; this clearly improves the strength. It is then expected that high-volume fractions of particles will lead to composites with higher tensile strength, compared with the unfilled material.

The effect of the particle volume fraction on the tensile properties of epoxy/volcanic ash and polyester/volcanic ash is shown in Fig. 1. It is seen that the optimum tensile strength is obtained when 10 vol. % volcanic ash is added into the polyester matrix and 40 vol. % for epoxy matrix. However, the addition of the ash is decreased the tensile strength of polyester matrix. This could be because of some factors, such as the uneven distribution of particles, porosity, void and particle/matrix interface adhesion. For poorly bonded micro-particles, strength reductions occur by adding particles. The voids are weak points where failures initiate. Poorly wetted filler surfaces and air entrapment in the mixture lead to the creation of voids around the particles. When the material is under stress, it will reduce its strength. On the other hand, it could be said that the

epoxy/volcanic ash has better stability than the polyester/volcanic ash in term of the tensile properties. There is slightly different result with the tensile strength of epoxy/ash composites. The strength decreases at first then increases gradually. The improvement of the maximum strength (40% vol. content of ash) is 1.87% if compared with the unfilled materials. It could also

be seen from the figure that the trend between polyester/ash and epoxy/ash is almost the same. When added the 10 vol. % until 30 vol. % particles into both polymers, the strength tends to decrease and increase again when 40 vol. % particles was added.

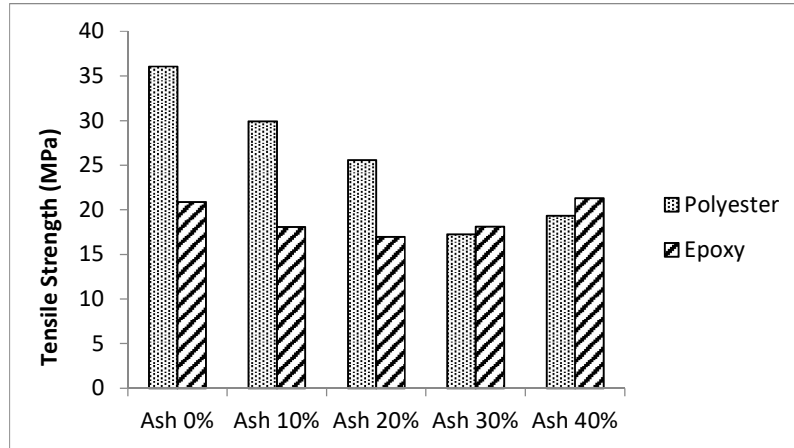


Figure 1. Effect of Mt. Kelud volcanic ash addition on tensile strength of polymer composites

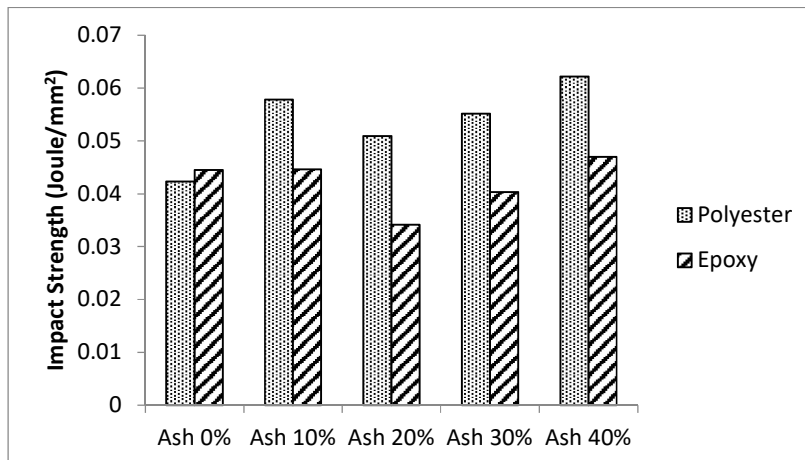


Figure 2. Effect of Mt. Kelud volcanic ash addition on impact strength of polymer composites

The properties of the reinforced adhesive are not only based on the properties of adhesive matrix or reinforcing particles, but also the other parameters that contribute to the toughness process, which largely influence the outcome of the composite material. The parameters considered in this research are the volume fraction (amount) of particles. The amount of particles dispersed in a structural adhesive matrix is a very important parameter in the toughening properties of the adhesive [28-31]. The volume of particles is directly related to the nature of the particles and their mechanical properties, so it is crucial have full knowledge of the particles nature and properties. Typically, for ductile particles, the critical strain energy release rate raise very slowly with the increased volume fraction and then reaches a plateau value [32] and [33].

Toughness can be described as the resistance of the material to fracture when stressed, or could be said that, the ability of a material to absorb energy and plastically deform without breaking. Toughness is one of the main aspects that determine the strength of materials. The assessment of toughness of an epoxy reinforced with particles can be made by using impact toughness test which results could be seen in Fig. 2. It could be concluded that the optimum particle content for both matrices is 40 vol. %. Impact strength of epoxy/volcanic ash and polyester/volcanic ash is way better than that unfilled materials but the impact strength of polyester is higher than the epoxy. A major percent increase of 47.04 % was achieved when 40 vol. % of volcanic ash particles were incorporated. This behavior was attributed to the

cross-linked effect within the polymer. The mechanical properties of polymers are often linked directly to its crosslink density. Some changes in crosslink density invariably lead to changes in mechanical properties. When perfectly dispersed

and wetted in the matrix, filler particles can maximize the possible attachment points with the polymer, that is the apparent crosslink density of the matrix, and then maximize the hardness and modulus [34].

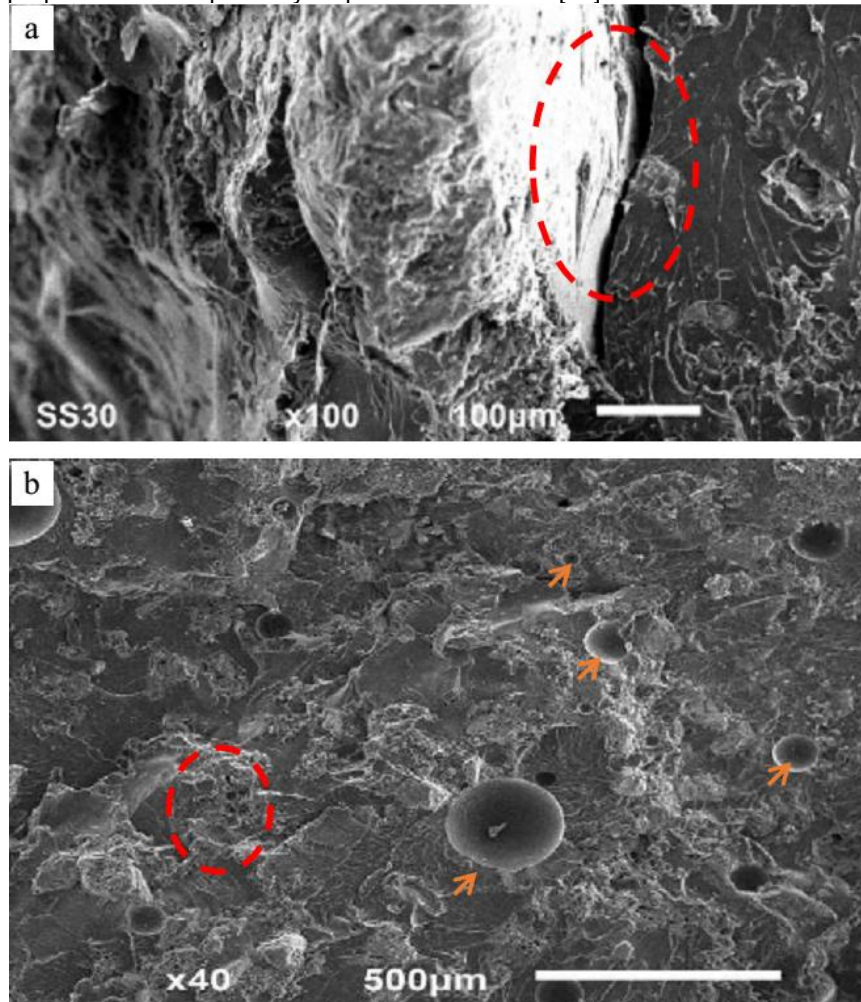


Figure 3. The SEM Micrograph of tensile fracture surface of Mt. Kelud volcanic ash particle composites on: (a) polyester matrix with 10 vol. % volcanic ash, (b) epoxy matrix with 40 vol. % volcanic ash

Scanning electron microscopy (SEM) is the most common methods used in the morphology evaluation. From SEM Micrograph of epoxy/ash and polyester/ash (Fig. 3a), it is seen that there are some voids (arrow sign) contained in the composite of epoxy/ash. This result supports the tensile result that there is the decreasing of tensile properties of material with the addition of the particles. There is also agglomeration (circle sign) found in the surface morphology of epoxy/ash composite. Agglomeration is the particles that stick to each other in adhesive due to chemical binding, surface energy reduction or particle segregation. It leads to a non-homogeneous behavior and lower macroscopic mechanical properties. Particle clusters act as crack or decohesion nucleation sites at stresses lower than the matrix yield strength, causing the composite to fail at unpredictable low stress levels [35]. The

polyester/ash fracture (Fig. 3b) shows that the brittle morphology found in the surface and the crack propagated along the surface. This crack could lead into the material failure if working under stress.

#### 4. CONCLUSIONS

In general, the mechanical properties of epoxy or polyester resin increased by adding volcanic ash that contains silica particles. For tensile test, the epoxy/ash composites with 40 vol. % particles has the highest tensile strength compared with the other variations. However, for the polyester/ash composites, the tensile strength continues to decrease with the addition of particles, which could be caused by some factors: uneven distribution of particles, porosity, void and the surface adhesion between particles and matrices.

It is different with the results of the impact test. There is a significant increasing of 47.04 % for polyester and 5.62 % for epoxy in impact strength when 40 vol. % of volcanic ash added into both polymers. This behavior was attributed to the cross-linked effect within the polymer that could enhance its properties. The SEM Micrograph result shows that there are voids and agglomeration contained in the epoxy/ash composites that could be the cause of the failure. There is also crack propagation along the surface of polyester/ash composites, which contributes to the failure mechanism of polyester reinforced volcanic ash particles composites.

#### ACKNOWLEDGEMENT

The author would like to thank the Adisutjipto College of Technology for funding this project. The authors would like to thank Agus Rianto and Djoko Kurniawan for their support to this work.

#### REFERENCES

- [1]. Danusso F, Tieghi G. Strength versus composition of rigid matrix particulate composites in *Polymer Journal*, pages 1385–1390. 1985.
- [2]. [2] Shao-Yun Fu, Xi-Qiao Feng, Bernd Lauke, Yiu-Wing Mai. Effects of particle size, particle/matrix interface adhesion and particle loading on mechanical properties of particulate-polymer composites in *Composites Part B: Engineering*, pages 933 – 961. 2008.
- [3]. Nafar Dastgerdi, G. Marquis, S. Sankaranarayanan, M. Gupta. Fatigue crack growth behavior of amorphous particulate reinforced composites in *Composite Structures Journal*, pages 782 – 790, 2016.
- [4]. Dong-Joo Lee. Fracture mechanical model for tensile strength of particle reinforced elastomeric composites in *Mechanics of Materials Journal*, pages 54 – 60. 2016.
- [5]. Debnath S, Ranade R, Wunder SL, McCool J, Boberick K, Baran G. Interface effects on mechanical properties of particle-reinforced composites in *Dental Materials Journal*, pages 677 – 686. 2004.
- [6]. D.E. Packham. *Handbook of adhesion*. John Wiley & Sons, Ltd, England, 2005.
- [7]. [7] R.D. Adams. *Adhesive bonding - science, technology and applications*. Woodhead Publishing Limited, Cambridge, 2000.
- [8]. A. Kinloch, J.H. Lee, A.C. Taylor, S. Sprenger, C. Eger, D. Egan. Toughening structural adhesives via nano-and micro-phase inclusions in *The Journal of Adhesive*, pages 867–873, 2003.
- [9]. C.B. Bucknall. *Toughened plastics*. Springer Science+Business Media, London, 1977.
- [10]. Y. Huang, D.L. Hunston, A.J. Kinloch, C.K. Riew. Mechanisms of toughening thermoset resins in *Toughened plastics I: Science and engineering*, American Chemical Society, Washington, pages 1–35. 1993.
- [11]. V.D. Ramos, H.M. Costa, V.L.P. Soares, R.S.V. Nascimento. Modification of epoxy resin: a comparison of different types of elastomer in *Polymer Testing Journal*, pages 387–394. 2005.
- [12]. B. Cardwell, A.F. Yee. Toughening of epoxies through thermoplastic crack bridging in *Journal of Material Science*, pages 5473–5484. 1998.
- [13]. G.P. Tandon, G.J. Weng A theory of particle-reinforced plasticity in *Journal of Applied Mechanics*, pages 126–135. 1988.
- [14]. A.D. Jenkins. *Advanced routes for polymer toughening*. Polymer Science Library, Elsevier, Amsterdam. 1996.
- [15]. G. Gkikas, N.M. Barkoula, A.S. Paipetis. Effect of dispersion conditions on the thermo-mechanical and toughness properties of multi walled carbon nanotubes-reinforced epoxy in *Composite Part B: Engineering Journal*, pages 2697–2705. 2012.
- [16]. A.Q. Barbosa, L.F.M. da Silva, M.D. Banea, A. Öchsner. Methods to increase the toughness of structural adhesives with micro particles: an overview with focus on cork particles in *Materialwissenschaft und Werkstofftechnik Journal*, pages 307–325. 2016.
- [17]. G.J. Withers, Y. Yu, V.N. Khabashesku, L. Cercone, V.G. Hadjiev, J.M. Souza, et al. Improved mechanical properties of an epoxy glass-fiber composite reinforced with surface organomodified nanoclays in *Composite Part B: Engineering Journal*, pages 175–182. 2015.
- [18]. Marjetka Conradi. Nanosilica-reinforced polymer composites in *Materials and Technology Journal*, pages 285 – 293. 2013.
- [19]. Anderson, T.L., *Fracture Mechanics*, 2nd Edition, CRC Press, Boca Raton, FL, 1995.
- [20]. P.A. Tzika, M.C. Boyce, D.M. Parks. Micromechanics of deformation in particle-toughened polyamide in *Journal of Mechanical Phys. Solids*, pages 1893–1929. 2001.
- [21]. M. Quaresimin, M. Zappalorto Salviato. A multi-scale and multi mechanism approach for the fracture toughness assessment of polymer nanocomposites in *Composites Science and Technology Journal*, pages 16 – 21. 2014.
- [22]. J Cho, M.S. Joshi, C.T. Sun. Effect of inclusion size on mechanical properties of polymeric composites with micro and nano particles in *Composites Science and Technology Journal*, pages 1941–1952. 2006.
- [23]. M.F. Serra, M.S. Conconi, G. Suarez, E.F. Aglietta, N.M. Rendtorff. Volcanic ash as flux in clay based triaxial ceramic materials, effect of the firing temperature in phases and

- mechanical properties in *Ceramic International Journal*, pages 6169 – 6177. 2015.
- [24]. Mohammed Seddik Meddah. Durability performance and engineering properties of shale and volcanic ashes concretes in *Construction and Building Materials Journal*, pages 73 – 82. 2015.
- [25]. Youngchan Shin, Deokkyu Lee, Kangtaek Lee, Kyung Hyun Ahn, Bumsang Kim. Surface properties of silica nanoparticles modified with polymers for polymer nanocomposite applications in *Journal of Industrial and Engineering Chemistry*, pages 515 – 519. 2008.
- [26]. Alisa Boonyapookana, Anchalee Saengsai, Supachai Surapunt, Kohsoku Nagata, Yoshiharu Mutoh. Time dependent fatigue crack growth behavior of silica particle reinforced epoxy resin composite in *International Journal of Fatigue*, pages 288 – 293. 2016.
- [27]. Rubens Bagni Torres, Júlio Cesar dos Santos, Túlio Hallak Panzera, André Luis Christoforo, Paulo H. Ribeiro Borges, Fabrizio Scarpa. Hybrid glass fibre reinforced composites containing silica and cement microparticles based on a design of experiment in *Polymer Testing Journal*, pages 87 – 93. 2016.
- [28]. Z. Minfeng, S. Xudong, X. Huiquan, J. Genzhong, J. Xuewen, W. Baoyi. Investigation of free volume and the interfacial, and toughening behavior for epoxy resin/rubber composites by positron annihilation in *Radiation Physics and Chemistry Journal*, pages 245 – 251. 2008.
- [29]. Y. Huang, A.J. Kinloch. The toughness of epoxy polymers containing microvoids in *Polymer Journal*, pages 1330 – 1332. 1992.
- [30]. A.J. Kinloch, D. Hunston. Effect of volume fraction of dispersed rubbery phase on the toughness of rubber-toughened epoxy polymers in *Journal of Materials Science*, pages 137 – 139. 1987