Thermal and Mechanical Properties of Hybrid Organoclay/Rockwool Fiber Reinforced Epoxy Composites

M Baiquni1,3, B Soegijono2,* and A N Hakim1

1Graduate Materials Science Program, Department of Physics, University of Indonesia
2Department of Physics, University of Indonesia
3Rocket Technology Center, National Institute of Aeronautics and Space (LAPAN)

*E-mail: naufal@ui.ac.id

Abstract. Hybrid rockwool/clay with polymer-based composites were prepared by varying organoclay contents and fixed rockwool fiber in epoxy resin as a matrix. The epoxy composites were fabricated using hand lay-up method. Characterization of composite materials for thermal and mechanical properties have been done by thermogravimetric analysis (TGA), thermal conductivity meter, universal testing machine (UTM), and impact tester. TGA results confirmed that the decomposition temperature shifted to a lower point. The thermal conductivity increased with the increasing organoclay content. The addition of 1 phr clay increased tensile and impact strength, while the addition of clay above 1 phr decreased the strength. The changes on these properties were due to cross link between intercalated organoclay and epoxy. The addition of 10 phr rockwool also decreased tensile strength, but improved impact strength.

1. Introduction
Epoxy resins have been used in various applications as surface adhesives and coatings for electronics devices and as structural-reinforced composites for automobiles, marine vehicles, and aerospace. However, epoxy resins have limited use at high temperature. Thus, thermal properties enhanced epoxy resins should be investigated for industrial application. Recently, additives or modifiers with inorganic fillers such as nano-powders, nanotubes, and layered silicate clays [1–3] have been generally used to improve their physical and mechanical properties. Layered silicate clays are particularly good candidates in improving the thermal, mechanical, barrier, and flame retardant properties of pure epoxy materials. Epoxy/clay nanocomposites have been studied extensively in recent years [4]. However, the conventional technique usually resulted in an intercalated or intercalated/exfoliated structure rather than a well dispersed and highly exfoliated structure [5–7]. Rockwool, sometimes called stone wool, is a type of inorganic fiber. Rockwool is used primarily for thermal and acoustical insulation, typically in buildings, vehicles, and other industrial equipment. It has a high tensile strength and modulus, high chemical resistance, high dimensional stability, and has excellent insulation properties [8]. Hybrid composite materials with more than one compound of filler are interesting, such as organoclay and rockwool with epoxy resin as a matrix. In this paper, composites epoxy resin with organoclay and rockwool as fillers have been studied to understand the role of organoclay and rockwool on thermal
stability and mechanical properties. Hence, we can use it for alternative material as heat-resistance material for many applications, such as a rocket fin component and rocket nose cone.

2. Experimental Methods

2.1. Raw materials
Diglycidyl ether of bisphenol A (DGEBA) resin was used as a matrix material (E), with a cycloaliphatic amine, EPH 555 as a hardener, was purchased from Justus Kimia Raya, Indonesia. Montmorillonite (MMT) organoclay with an octadecyl ammonium salt was purchased from Nanocor Inc. Rockwool fibers were supplied by PT Insfoil Pradasakti, Indonesia.

2.2. Composite preparations

2.2.1. Epoxy/clay composite preparation
The epoxy-clay composites were prepared using 0, 1, 3, and 5 phr (per hundred resin) of organoclay/MMT (C). The epoxy-clay mixture was stirred for 24 hours at 400 rpm. After homogenization, the mixtures were placed on a vacuum chamber to remove air bubbles for 5 minutes. Then, the curing agent was added and the mixtures was placed in the mold and cured for 24 hours at room temperature.

2.2.2. Rockwool/clay-reinforced epoxy composites preparation
The epoxy-clay mixture containing 1, 3, and 5 phr clay were prepared. Rockwool fiber (R) is hard to process into composite, so the loading of composites was fixed at 10 phr. After addition of the curing agent, the epoxy-clay mixture was poured into rockwool fiber at the mold with hand lay-up method. After degassing at vacuum chamber for 5 minutes, the composites were cured for 24 hours at room temperature.

2.3. Characterization
Thermogravimetric analysis (TGA) was used to characterize thermal stability of materials. TGA was conducted using Simultaneous Thermal Analyzer (STA) 6000 (Perkin-Elmer Inc. USA). The test was carried out at 35°C to 800°C with 10°C/min heating rate and 0.1 ml/min nitrogen flow rate. Tensile test was conducted using Tensilon 100KN universal testing machine according ASTM D638. The test was carried out at the constant strain rate of 5 mm/min. Thermal conductivity tests were done using QTM 500 Conductometer. Impact charpy impact tests were done by impact tester according to ASTM E23. The test samples were U-notched.

3. Results and Discussion

3.1. TGA analysis
The thermal decomposition of clay-reinforced epoxy composites are shown in figure 1 and rockwool/clay-reinforced epoxy composites at figure 2. From the observation on figure 1, the composite have three stage of decomposition. First decomposition was at 100°C, which was attributed to degradation of water and other volatile compounds. Then, the epoxy decomposition has onset temperature at 309°C. Due to the addition of organoclay, the onset temperature has shifted to 283-292°C. This was possibly the effect of decreased crosslink density of epoxy resin caused by the addition of organoclay [9]. Thermal decomposition temperature for the composites is at 350°C to 360°C. At this temperature the mass loss rate increased or reached the highest decomposition temperature (Tdmax). The decomposition of pure epoxy ended at 410°C (Tend). After the addition of organoclay the endset temperature increased to about 450°C. This showed that the thermal stability of the composites have increased. Table 1 provides the thermal stability for each composite. The enhanced thermal stability of composite is attributed to the interaction of base polymer and organoclay surface [10]. Furthermore, figure 2 shows TGA curves of rockwool/clay-reinforced epoxy composites. The addition of rockwool increased the thermal stability, which is Td increased to 370°C. The char yields of the composites at 500°C and above shows the rockwool and clay residues.
3.2. Mechanical properties
Figures 3 and 4 show the effect of clay and rockwool addition on tensile strength of epoxy composites. From the observation of figure 3, it can be seen that the composite with 1 phr clay has the highest tensile strength. The increase in tensile strength may be attributed to the formation of exfoliated structure for epoxy nanocomposites [11]. Silicate layer on organoclay exfoliated and dispersed uniformly in the polymer matrix in nanometer size with high aspect ratio. The high aspect ratio of organoclay may also increase the tensile strength by increasing the contact surface area of the filler on the polymer matrix. Large numbers of reinforcing organoclay platelets in the polymer matrix act as efficient stress transfer agents in the composites, inducing plastic deformation into the base polymer, and finally increase the tensile strength [12]. But, the addition of rockwool on epoxy-clay composites decreased the tensile strength due to the brittleness of rockwool itself. Rockwool has high tensile strength and modulus, but low elongation (<5%). The fiber length and orientation in the composites
are in random form. The random orientation of rockwool may responsible for this phenomena. Figure 4 shows the tensile strength for rockwool/clay-reinforced epoxy composites. The addition of organoclay increased tensile strength, but drastically decreased for organoclay addition above 3 phr. This result was attributed to the agglomeration of organoclay.

The impact strength of the rockwool/clay-reinforced epoxy composites increased drastically with the addition of rockwool and 1 phr clay as shown in figure 5. However, the impact strength was slightly higher with the addition of 3 and 5 phr clay compared to the composite without clay addition. The increase in impact strength may be related to the exfoliated silicate layers of clay in the neat epoxy matrix [11]. The organoclays may have a good toughening effect and act as efficient crack stoppers and form a tortuous crack propagation path, resulting in higher impact strength [13]. Furthermore, the intercalated or agglomerated silicate layers of clay were believed to be responsible for the reduction in impact strength for the epoxy composites with clay content above 1 phr. On the other hand, the impact strength of the clay-reinforced epoxy composites increased drastically with the addition of 1 and 5 phr clay and slightly increased with the addition of 3 phr clay. Zhang et al. [14] reported that the impact strength improved with the presence of clay up to 3 wt% and the impact strength drastically reduced in the epoxy/clay nanocomposites with clay content above 3 wt%. However, the addition of 5 phr has the highest impact strength. It may be caused by the non-homogeneous clay during fabrication. The agglomerated clay did not spread evenly in the epoxy mixture.

| No | Sample | $T_{\text{onset}}$(°C) | $T_{\text{decomposition}}$(°C) | $T_{\text{endset}}$(°C) |
|----|--------|-----------------|-----------------|-----------------|
| 1  | E      | 309             | 363             | 410             |
| 2  | E-C1   | 292             | 353             | 445             |
| 3  | E-C3   | 282             | 371             | 447             |
| 4  | E-C5   | 285             | 369             | 448             |
| 5  | E-R10  | 276             | 370             | 450             |
| 6  | E-R10-C1 | 273          | 373             | 450             |
| 7  | E-R10-C3 | 287          | 375             | 453             |
| 8  | E-R10-C5 | 285          | 368             | 453             |

Figure 3. Tensile strength of clay-reinforced epoxy composites

Figure 4. Tensile strength of rockwool/clay-reinforced epoxy composites.
3.3. **Thermal conductivity analysis**

Figure 6 shows thermal conductivity of epoxy, epoxy/clay composites, rockwool/clay-reinforced epoxy composites for various organoclay addition. It can be seen that thermal conductivity increased with the addition of organoclay. Organoclay has higher thermal conductivity than the polymer epoxy. However, the composites with rockwool addition have stable thermal conductivity. TC of composites only increased for the composite with 3 phr clay. TC of composite with 5 phr clay decreased due to voids formation while processing the composites.

4. **Conclusions**

Epoxy-based polymer composites with rockwool and clay reinforcement have been prepared by hand-layup method. The epoxy-clay composite with the addition of 1 phr clay has the highest tensile strength. While the addition of 10 phr rockwool decreases the tensile strength of the composite. The highest impact strength was obtained for the E-C5 composite and the E-R10-C1 composite. The
addition of filler on the composite increased the value of thermal conductivity. The composite with 1 phr clay content (E-R10-C1) shows good thermal stability.

Acknowledgments
This research is supported financially by LAPAN (National Institute of Aeronautics and Space) and “Hibah Publikasi Internasional Terindeks untuk Tugas Akhir Mahasiswa Universitas Indonesia” through letter of decree No:2239/UN2.3.1/HKP.05.00/2018.

References
[1] Yu H J, Wang L, Shi Q, Jiang G H, Zhao Z R, and Dong X C 2006 Prog. Org. Coat. 55 296
[2] Ye Y, Chen H, Wu J, and Ye L 2007 Polymer 48 6426
[3] Kint D P R, Seeley G, Gio-Batta M, and Burgess A N 2005 J. Macromol. Sci. Part B Phys. 44 1021
[4] Ray S S and Okamoto M 2003 Prog. Polym. Sci. 28 1539
[5] Zerda A S and Lesser A J 2001 J. Polym. Sci. Part B Polym. Phys. 39 1137
[6] Miyagawa H, Drzal L T, and Carsello J A 2006 Polym. Eng. Sci. 46 452
[7] Qi B, Zhang Q X, Bannister M, and Mai Y W 2006 Compos. Struct. 75 514
[8] Öztürk, B 2010 Fibers and Polym. 11 464
[9] Brnardić I, Macan, J, Ivanković, H, and Ivanković, M 2008 J. Appl. Polym. Sci. 107 1932
[10] Tortora M, Gorrasi G, Vittoria V, Galli G, and Ritrovati S, and Chiellini, E 2002 Polym. 43 6147
[11] Kusmono, Wildan M W, and Mohd Ishak Z A 2013 Int. J. Poly. Sci. 2013 1
[12] Parija S, Nayak S K, Verma S K, and Tripathy S S 2004 Polym. Compos. 25 646
[13] Inceoglu A B and Yilmazer U 2003 Polym. Eng. Sci. 43 661
[14] Zhang K, Wang L, Wang F, Wang G, and Li Z 2004 J. Polym. Sci. 91 2649