Tensile Behavior of Zeolite-filled High Density Polyethylene Annealed Composite

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Abstrak-Penelitian ini difokuskan pada penyelidikan perilaku tarik dari komposit zeolit alam- *high density polyethylene* (HDPE) yang dianil. Proses anil komposit dilakukan pada suhu 90^{o}C selama 12 jam. Pengujian tarik uniaksial dilakukan pada *cross head speed* konstan 4 mm/menit pada suhu kamar (25^{o}C). Hasil penelitian menunjukkan bahwa perpanjangan putus, rasio daktilitas dan *yield stress* menurun dengan meningkatnya konsentrasi zeolit dalam matrik. Sementara itu, kekuatan tarik dan modulus Young meningkat dengan meningkatnya kandungan zeolit hingga 5 wt.%, dan nilai-nilai itu menurun pada penambahan kandungan zeolit lebih lanjut. Hal ini disebabkan mekanisme transfer tegangan dari matriks ke partikel terjadi secara efisien dalam kandungan zeolit 5 wt.%.

Kata kunci: tensile strength, zeolite, HDPE, mechanical properties

Abstract-This work is focused on an investigation of tensile behavior of natural zeolite-high density polyethylene (HDPE) annealed composites. Annealing process of injection molded composite was performed at 90^{o}C for 12 h. Uniaxial tensile test was conducted at constant cross head speed of 4 mm/minat room temperature (25^{o}C). Results showed that elongation at break, ductility ratioandyield stressdecreasedwith increasingconcentration of zeolite in host matrix. While, tensile strength and Young'smodulusincreasedwith the increasing zeolite content up to 5 wt.%, and they decreased in further additions. This is due to the mechanism of stress transfer from the matrix to the particles occurs efficiently in zeolite content of 5 wt.%.

Keywords:tensile strength, zeolite, HDPE, mechanical properties

1. Introduction

Mechanical properties of engineering materials such as tensile strength, ductility, toughness, and fatigue strength are frequently found to posses great mechanical properties so then they are suitable for implementation. In uniaxial tensile test, percentage reduction in elongation obtained is taken to be a measure of ductility. As a candidate materials for skull implant reconstruction, zeolite/HDPE composites has all the requirements such as biocompatible (Bedi et.al, 2012), bioactive (Kaali & Czel, 2012; Bedi et.al, 2012; Sanchez et.al, 2015) and their ability to protectthe matrix degradation due to ultraviolet radiation (Pavelic & Hadzija, 2003). Many studies on the mechanical properties of bone implants have been conducted through characterization of the stiffness and the tensile strength (Taurio & Tormala, 1991; Wang & Bonfield, 2001; Couves et.al, 1993; Purnomo et.al, 2015). Since the low mechanical properties, the material can only be used for low load-bearing application (Woodcocket.al, 1999). However, good strength and good stiffness are important if an implant material is used for load-bearing application such as skull reconstruction implant (Marinkovicet.al, 2004), so the research that aims to improve these properties is very important to be done.

This paper presents the results on the tensile behavior of zeolite/HDPE annealed composites. The tests were carried out in room temperatures. The variation of composite stiffness with loading of zeolite was also investigated.

2. Research method

The material used is the same as previous studies (Purnomo et.al, 2014, 2015, 2016, inpress) namely commercial grade of HDPE as matrixand natural zeolite powder as fillers.The composites with zeolite weight concentration of 0 wt.%, 2.5 wt.%, 5 wt.%, and 7.5 wt.% was annealed for 12 h with heating rate of 1 ^oC/min and cooling rate of 0.5 ^oC/min.Tensile properties of zeolite/HDPE annealed composites were examined by tensile test on dumbbell specimens. The tests were performed according to ASTM D638 at room temperature using a universal testing machine at a constant cross head speed of 4 mm/min. All samples were subjected a tensile load till the samples totally rupture and the specimen elongation was calculated from the crosshead displacement.

3. Result and Discussion

Plots of zeolite concentration against the elongation at break and ducyility ratio, i.e., the ratio of the elongation at break (x_b)tothe maximum load (F_{max}), were shown in Fig. 1. Apparently that the ductility of composites decreased which characterized by decreasing the elongation at break and elongation per load applied with zeolite content increased. This means that the increased of zeolite concentration caused the behavior gradual transition to brittle. However, their value was higher than that in previous study [Purnomo et.al, 2015]. This is due to effect of annealing treatments process before specimen tested.

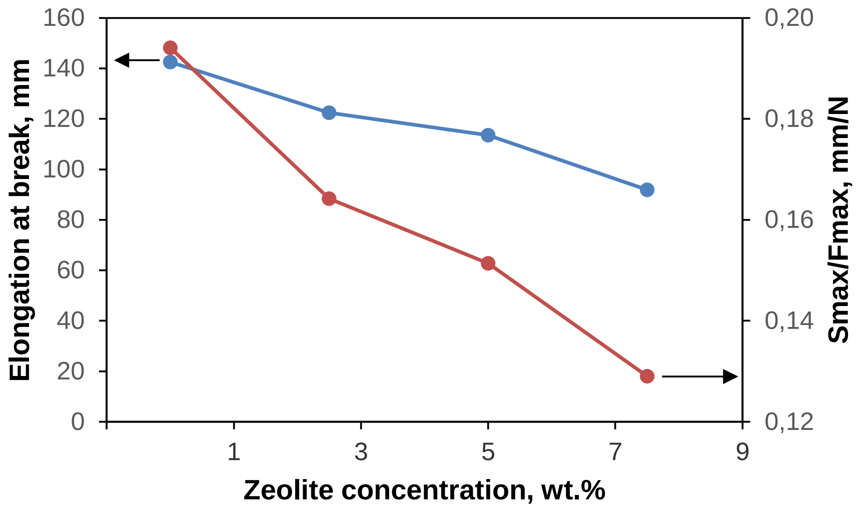

Figure 1. The zeolite concentration effect on the elongation at break and x_b/F_{max} ratio

Plotting of tensile strength against zeolite concentration was shown in Fig. 2. Apparently that the tensile strength of annealed composites increased with increasing the zeolite concentration up to 5 wt% in host matrix. However, increasing zeolite concentration above 5 wt% brings decrease in tensile strength. This result confirmed the previous study [Purnomo et.al, 2015] despite each value in same composition in this study increase in compared with value in previous study.The influence of zeolite concentration on the yield stresse of composite can be seen in Fig.3. Apparently, incorporation of zeolite in host matrix led to decrease gradually in yield stress of composites. This phenomenon is different with the results of previous studies [Purnomo et.al, 2015] which had been reported that the yield stress decreased sharply by incorporation of zeolites in host matrix.

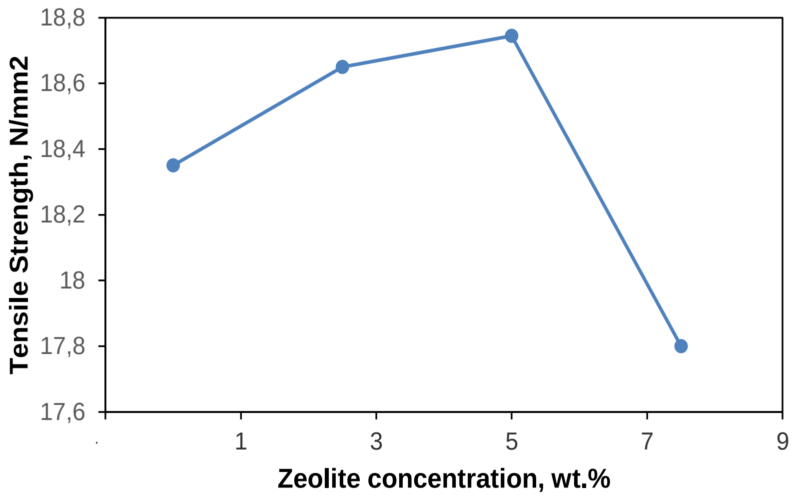

Fig.2. The influence of zeolite content on the tensile strength of composite

Figure 3 also show the effect of zeolite content on the Young'smodulus ofannealed composites. It can be seen that the modulus increased with incorporation of zeolite up 5 wt.%, while it was decreased with zeolite contents further up.

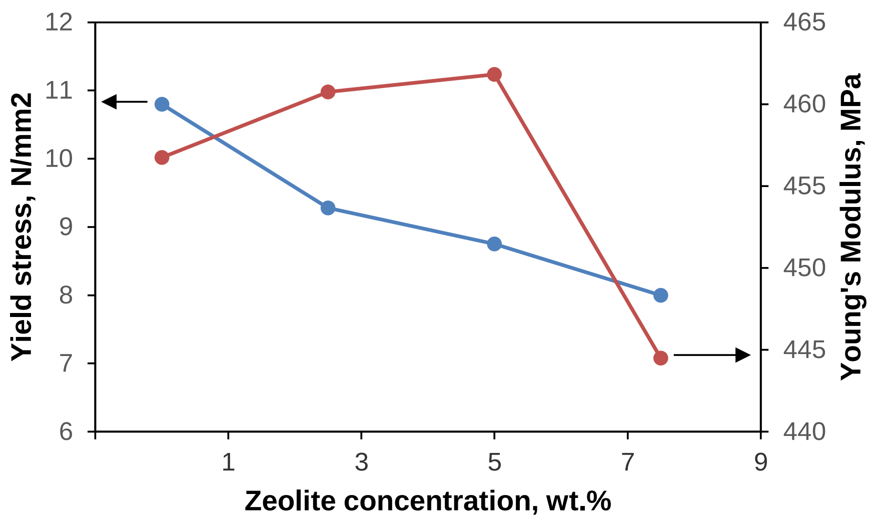

Fig.3. The yield stress and Young’s modulus of composites against zeolite concentration.

The SEM image as shown in Fig. 4 taken on the strain region show the voids grow in the tensile stress direction (Fig.4a). In further deformation of matrix caused matrix decohesion and void growth to coalescence (Fig. 4b) before rupture.

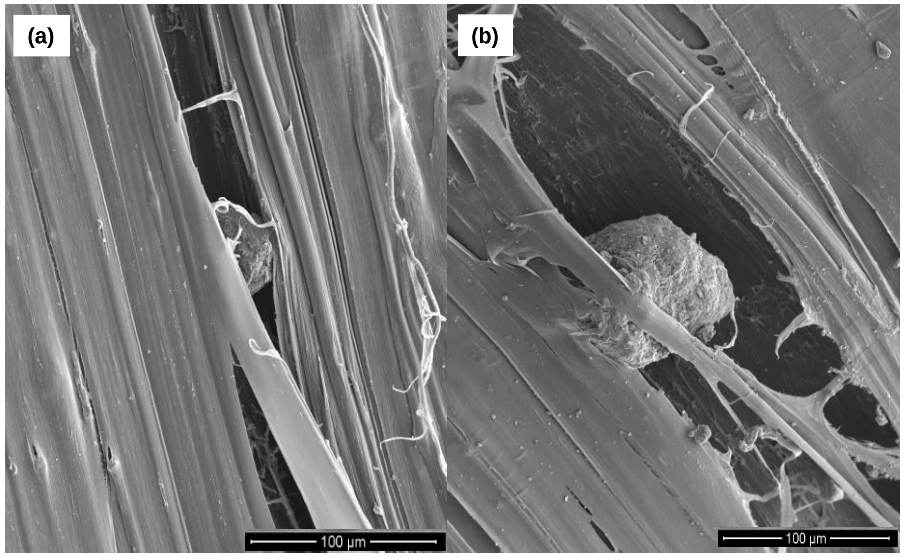

Fig. 4. SEM images taken in stain region of 5 wt.% zeolite/HDPE

In case of composites with zeolite content of 5 wt.%, the good interface adhesion between HDPE matrix and zeolite particles lead to stress in an efficient transfer mechanism from the matrix to the particle. Beyond zeolite concentration of 5 wt.%, incorporation of zeolite promotes the aggregates formation of zeolite particles in host matrix. The aggregates, associated with the material filling the cracks, are poor bonded so that the particles will undergo debonding and not capable to carry the load. Therefore, the bonding strength of the material decreases with increasing content of zeolite.The yield stress depends on adhesion between the filler and the matrix. Yield stress of composite is equal to yield stress of unfilled polymer if the adhesion between the particles and the matrix as strong cohesion in the matrix. Decrease of adhesion cause a reduction in the yield stress.

There are two models exist to predict the stress required to initiate particles-matrix debonding, i.e. developed by Vollenberg et.al (1987, 1988) and Pukánszky & Voros (1993). According to both models initiation stress dependent on the interfacial strength of particles-matrix bonding, on the matrix stiffness and on the particles size. Relationship the related parameters can be expressed as follows (Pukánszky& Voros, 1993):

$$\sigma_B = -C_1\sigma_{Th} + C_2\sqrt{\frac{W_A E_M}{R}} \tag{1}$$

where σ_B is debonding stress, σ_{Th} is thermal stress, W_A is the reversible work of adhesion, E_M is the Young's modulus of matrix, R is the radius of the particles, while C_1 and C_2 are constants. Thus, greater stress initiation is required to initiate debonding. This stress increase with increasing the adhesion at the matrix-particle interface. The influence of particle size and interfacial adhesion on debonding stress have been widely studied in the past, and it clearly proves the validity of the model (Pukánszky et.al, 1993, 1994). According to Pukánszky& Voros (1993) and Vollenberg et.al (1988), debonding stress is comparable to the E_M and W_A, which means the debonding stress increased with increasing the reversible work of adhesion. While, Nilsen and Landel (1974) and Kolarik et.al (1987) reported that the composite stiffness is comparable to the matrix stiffness.

4. Conclusion

Annealing the zeolite-HDPE composites have an impact on changes in the trend value of tensile strength and Young's modulus. In the zeolite content higher than 5%, the tensile strength was increased but Young's modulus decreased sharply lower than that of neat HDPE. Composites with the best properties is achieved on the composition of 5 wt.% zeolite and 95 wt.% HDPE.

References

Bedi R.S., Beving D.E., Zanello L.P., Yan Y, 2012, Biocompatibility of corrosion-resistant zeolite coatings for titanium alloy biomedical implants,*Acta Biomaterialia*,Vol. 5. Pp. 3265-3271.

Bedi R.S., Chow G., Wang J., Zanello L., Yan Y.S., 2012, Bioactive Materials for Regenerative Medicine: Zeolite-Hydroxyapatite Bone Mimetic Coatings, *Advanced Engineering Materials*, Vol.14, no. 3, pp. 200-206.

Couves J. W., Jones R. H., Parker S. C., Tschaufeser P., Catlow C. R. A., 1993, J. Phys.: Condens. Matter 5, L329.

Kaali P.& Czel G, 2012,Single, Binary and Ternary Ion Exchanged Zeolite as an Effective Bioactive Filler for Biomedical Polymer Composites,*Materials Science Forum*, 729, 234.

Kolarík J., Lednický F., Pukánszky B., 1987. Ternary Composites Polypropylene/ Elastomer/filler: Structure and Elastic Properties, Proceedings of 6th ICCM/2nd ECCM, London, United Kingdom, Vol. 1, 452–461

Marinkovic B.A., Jardim.,A. Saavedra, L.Y. Lau, C. Baehtz, R.R. de Avillez, F. Rizzo, 2004, Negative thermal expansion in hydrated HZSM-5 orthorhombic zeolite. *Microporous and Mesoporous Materials*, Vol. 71, pp. 117–124.

Nilsen L. E., Landel R. F., 1974. Mechanical Properties of Polymers and Composites, Marcel Dekker, New York

Pavelic K., Hadzija M., 2003, Medical Applications of Zeolites. in Hand Book Of Zeolites Sciences And Technology. Marcel Dekker, Inc, New York.

Pukánszky B., Vörös G., 1993, Mechanism of interfacial interactions in particulate filled composites. *Composite Interfaces*, Vol. **1**, pp. 411–427.

Pukánszky B., van Es M., Maurer F. H. J., Vörös G., 1994, Micromechanical deformations in particulate filled thermoplastics- volume strain measurements. *Journal of Material Science*, Vol. **29**, pp. 2350–2358.

Purnomo, Soenoko, R., Irawan, Y. S. And Suprapto, A, 2014, Fracture behavior of Zeolite-filled High Density Polyethylene Based on Energy Partitioning Work of Fracture,*International Journal of Applied Engineering Research*, Vol. 9, No. 24, pp. 28737-28747.

Purnomo, Soenoko, R., Irawan, Y. S. and Suprapto, A, 2015, Morphological and Mechanical Properties of Natural Zeolite-High Density Polyethylene Composite, *International Journal of Applied Engineering Research*, Vol. 10, No. 11, pp. 28001-28012.

Purnomo, Soenoko, R., Suprapto, A. and Irawan, Y. S., 2016, Impact Fracture Toughness Evaluation by Essential Work of Fracture Method in High Density Polyethylene Filled with Zeolite, *FME Transactions*, Vol. 44, pp. 180-186.

Purnomo, Soenoko, R., Irawan, Y. S. and Suprapto, A, in Press, Deformation Under Quasi Static Loading In High Density Polyethylene Filled With Natural Zeolite, *Journal of Engineering Science and Technology*, inPress

Sanchez M.J., Gamero P., Cortes D, 2012, Bioactivity assessment of ZSM-5 type zeolite functionalized with silver or zinc,*Journal of Material Letters*,Vol. 74, pp. 250-253.

Taurio R, Tormala P., 1991, Coral based and sintered hydroxylapatite blocks reinforced with "brous cage-like polylactide composite: a comparative study. In: Bonfield W, Hastings GW, Tanner KE, editors. Bioceramics 4. Oxford: Butterworth Heinemann Ltd. p. 287-294.

Vollenberg P., Heikens D., Ladan H. C. B., 1988, Experimental determination of thermal and adhesion stress in particle filled thermoplasts. *Polymer Composites*, Vol. **9**, pp. 382–388.

Vollenberg P. H. T., 1987, The mechanical behaviour of particle filled thermoplastics. PhD Thesis, Eindhoven University of Tec hnology.
Wang M., Bonfield W, 2001, Chemically coupled hydroxyapatite-polyethylene composites: structure and properties. *Biomaterials*, Vol. 22, pp. 1311-1320.
Woodcock D. A., Lightfoot P., Villaescusa L. A., Diaz-Cabanas M. J., Camblor M.A., Engberg D, 1999,*Chem. Mater*. 11, pp. 2508.