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Tensile properties of kenaf fiber and corn husk flour reinforced poly(lactic acid) hybrid bio-composites: Role of aspect ratio of natural fibers

Hyeok-Jin Kwon [a], Jackapon Sunthornvarabhas [a,b], Ji-Won Park [a], Jung-Hun Lee [a], Hyun-Joong Kim [a,c,*], Kuakoon Piyachomkwan [b], Klanarong Sriroth [d], Donghwan Cho [e]

[a] Lab. of Adhesion & Bio-Composites, Program in Environmental Materials Science, Seoul National University, Seoul 151-921, Republic of Korea
[b] Cassava and Starch Technology Research Unit, National Center for Genetic Engineering and Biotechnology, Bangkok 10900, Thailand
[c] Research Institute for Agriculture and Life Sciences, Seoul National University, Seoul 151-921, Republic of Korea
[d] Kasetsart Agricultural and Agro-Industrial Product Improvement Institute, Department of Biotechnology, Kasetsart University, Bangkok 10900, Thailand
[e] Department of Polymer Science and Engineering, Kumoh National Institute of Technology, Gumi, Gyeongbuk 730-701, Republic of Korea

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ABSTRACT

Kenaf fiber and corn husk flour were used as reinforcement in a novel biodegradable hybrid bio-composite system. It was investigated how the aspect ratios of kenaf fibers measured before and after passing through extrusion process influence the mechanical properties and the improvement of predicted values obtained by the Halpin–Tsai equation. It was found that considering of the aspect ratio of reinforcement obtained after the extrusion process, the difference between theoretical and experimental values of the tensile modulus was not significant, indicating that the aspect ratio determined after extrusion did not influence the predicted values. Therefore it was pointed out that the initial values of aspect ratio determined before extrusion can be used directly. It was also found that a scale ratio between reinforcements of different aspect ratios may play a role as a controlling factor in optimizing the mechanical properties of a hybrid bio-composite.

1. Introduction

Recently, biodegradable polymers, such as PLA, PBS, PHA, and PHB [1–3], have been introduced to replace fossil-based polymers due to many reasons such as shortage of oil supply, global environmental concerns on CO_2emission, and waste problem of plastic from fossil-based products. However, the mechanical properties of most biodegradable polymers are not sufficient to be used as a complete replacement material for fossil-based polymer. It has been known that a combination of natural fibers and synthetic biodegradable polymers may contribute to enhancing the properties and also to compensating their disadvantages by incorporating additives [2,3].

Using natural reinforcement in plastics is a simple but positive approach to improve the mechanical properties of composite materials, which had been established around early 1900s [4]. A variety of cellulose-based natural fibers and chemical treatments including coupling agents were also applied for improving the mechanical properties of biocomposites [5–9]. In the recent years, the development of composite with reinforcing agents of more than one type and shape was proposed to compensate their shortcomings, resulting in a positive hybrid effect [10–12]. A hybrid composite, which consists of two or more reinforcement types with different aspect ratios [10], had been introduced to further increase the mechanical properties of a reinforced composite system. This is because the properties of hybrid composites may surpass those of single reinforced composites [13,14]. Up to date, only a small number of papers have been dealing with hybrid composites consisting of a biodegradable polymer and two different types of natural reinforcement [4,13]. Besides, the prediction of mechanical properties of composites has many advantages when components are changed in volume or sort. Because we do not need additional measurements in order to know their mechanical properties [10]. Consequently, this paper aims to elucidate the effect of aspect ratio on the mechanical properties and the prediction of the mechanical properties by a mathematical approach for the purpose of optimization. In this paper, discussion based on the data collected from literatures was also given in order to draw a brief guideline for future development on hybrid composites.

* Corresponding author at: Lab. of Adhesion & Bio-Composites, Program in Environmental Materials Science, Seoul National University, Seoul 151-921, Republic of Korea. Tel.: +82 10 7123 4784.

E-mail address: hjokim@snu.ac.kr (H.-J. Kim).

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2. Experimental

2.1. Materials

PLA granules with an average diameter of 81 μm and a density 1.24 g/cm^3 were supplied by NatureWorks LLC, USA. The melt-flow rate, melt temperature and glass transition temperature were 4–8 g/10 min (190 °C/2160 g), 140–152 °C and 56.7–57.9 °C, respectively. Kenaf fibers, which were kindly imported from Bangladesh by Sutongsang Co., Korea, were used as reinforcement. Kenaf fibers were finely ground to a size which can pass through a filter of 40 mesh size by a disintegrator (CTM 200 in KUKJE Scientific Instrument., Korea). The 100 meshed corn husk flours were donated by Corn Products Korea, Inc., Korea. Both kenaf fibers and corn husk were dried in an oven at 80 °C for 24 h before compounding to remove water possibly existing in the 'as-supplied' materials and also to improve the blending efficiency before compounding.

2.2. Compounding and extrusion of kenaf/corn husk/PLA pellets

Different types of hybrid bio-composites were fabricated at a fixed ratio of reinforcement and PLA of 30:70 by weight and also at variable ratios of kenaf fiber and corn husk flour of 30:0, 20:10, 15:15, 10:20, and 0:30 by weight. The compounded materials prepared according to the mixing ratios described above were processed using a twin-screw extruder (BA-19 in Bautek Co., Korea). The processing conditions for extrusion were as follows. The barrel temperatures of kneading block zones in the extruder were 175, 175, 185, 185, 185, 175, 155, and 140 °C.The screw speed was 150 rpm. The kenaf/corn husk/PLA extrudates were cooled down to ambient temperature in a water bath. The extrudates were cut by using a pelletizer and sufficiently dried.

2.3. Injection molding to prepare kenaf/corn husk/PLA bio-composites for mechanical tests

Tensile test specimens were prepared from the dried pellets through an injection molding machine (Bautek Co., Korea). The temperature for injection molding was maintained at 190 °C.

2.4. Tensile test

Tensile test was conducted using the universal testing machine (Zwick Co., Germany) according to ASTM D 638-10 at a crosshead speed of 5 mm/min and at room temperature. Five specimens were measured to determine the value of tensile properties and the deviations.

2.5. Frequency distributions of the aspect ratio of natural fillers

An optical microscope (ICAMSCOPE SV-55, SOMETECH, Korea) was used to analyze the length and the diameter of the reinforcements. Before measured, the dried pellets were soaked in chloroform to remove PLA therein, followed by filtration of the diluted solution to retrieve residual reinforcements for analysis. In order to determine the average length and diameter of reinforcements, at least 120 individual specimens were observed.

3. Results and discussion

It has been discussed in author previous work about the effect of fiber loading on the tensile and flexural properties of bio-composites [15]. It was reported that both mechanical properties strongly depended on the kenaf fiber loading. In the present study, investigation was focused on understanding how the aspect ratio of reinforcements incorporated into the PLA matrix affects the improvement of the mechanical properties of bio-composite, by comparing to other hybrid bio-composite cases. Post analysis was conducted to estimate amount of total reinforcement consistency in each resin composite. We expected the loading content of reinforcements was about 30 wt.% in each matrix. Even though an average of loading content from five samples replicate for each condition was close to 30 wt.%, the results showed the loading content of which large variation was up to 12–15 wt.% from randomly selected conditions. The variation in loading content distribution might yield to the limitation of laboratory extruder – small portion of mixing screw length – and processing conditions were also selected based on the performance when making an extrusion process of selected conditions which was determined by cross sectional images of composite resin homogeneity as shown in Fig. 1. While there is the variation of reinforcement loading content

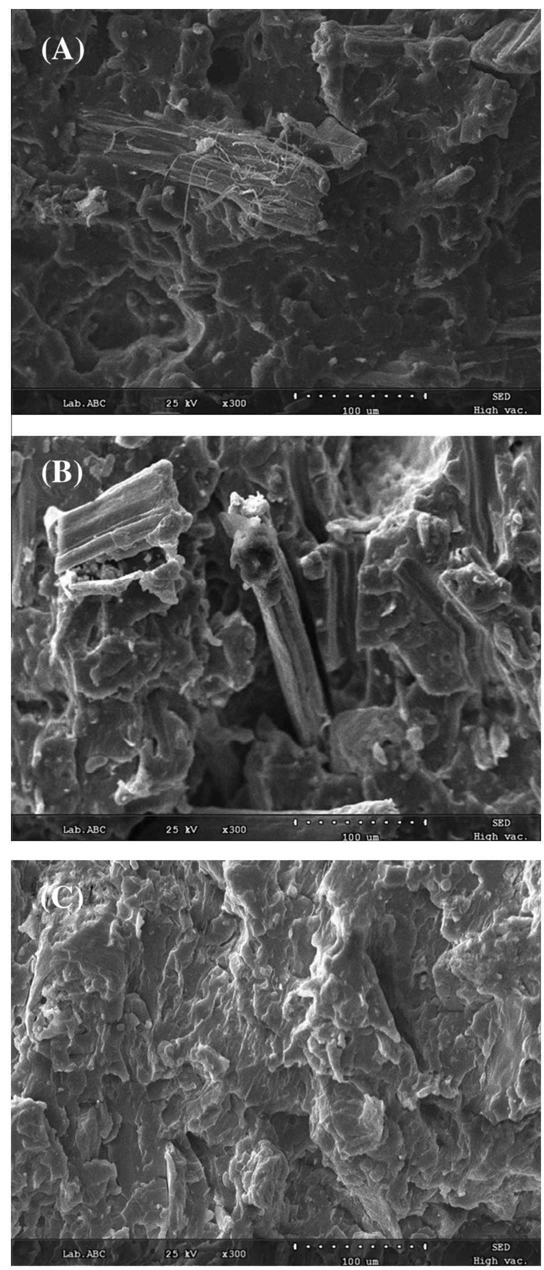

Fig. 1. Cross-sectional micrographs of the extruded PLA pellets with different reinforcement loadings: (A) kenaf 30 wt%, (B) kenaf 15 wt% and corn husk 15 wt%, and (C) corn husk 30 wt%.

in matrix, the mechanical properties has less effect as shown in small variation from measurement.

3.1. Change of the aspect ratio of reinforcements before and after extrusion

The size reduction of reinforcing agents after extrusion process occurs due to shear force [5,10]. The similar case was observed when passing composite of long reinforcing agents through extrusion process [16–18]. The results showed that during the extrusion the reinforcing agents were cut which results in smaller aspect ratio as observed in both kenaf fiber and corn husk flour in this study. The average aspect ratios of reinforcements before and after extrusion process were changed from 10.47 ± 6.52 to 5.37 ± 1.67 for kenaf fibers and from 1.67 ± 0.47 to 1.43 ± 0.27 for corn husk flours, respectively, as shown in Fig. 2. The reduction of the aspect ratio in kenaf was greater than that in corn husk, indicating that the kenaf fibers were subject to the shearing force in the screw barrel. This may be ascribed to the large surface area of kenaf fiber, compared to corn husk flours.

3.2. Aspect ratio of the reinforcement to predict the mechanical properties

A mathematical model to predict the mechanical properties of hybrid composites has often been utilized to anticipate composite behavior. A Rule of hybrid mixture, ROHM, and Halpin-Tsai equation has been used in many studies on hybrid composite. The ROHM is the simplest form of model prediction for composite while another version of model prediction is the Halpin–Tsai equation. The Halpin–Tsai model was calculated by follow as:

$$E_{random} = 3/8E_{11} + 5/8E_{22} \quad (1)$$

where E_{11}, E_{22} were calculated according to the following equations:

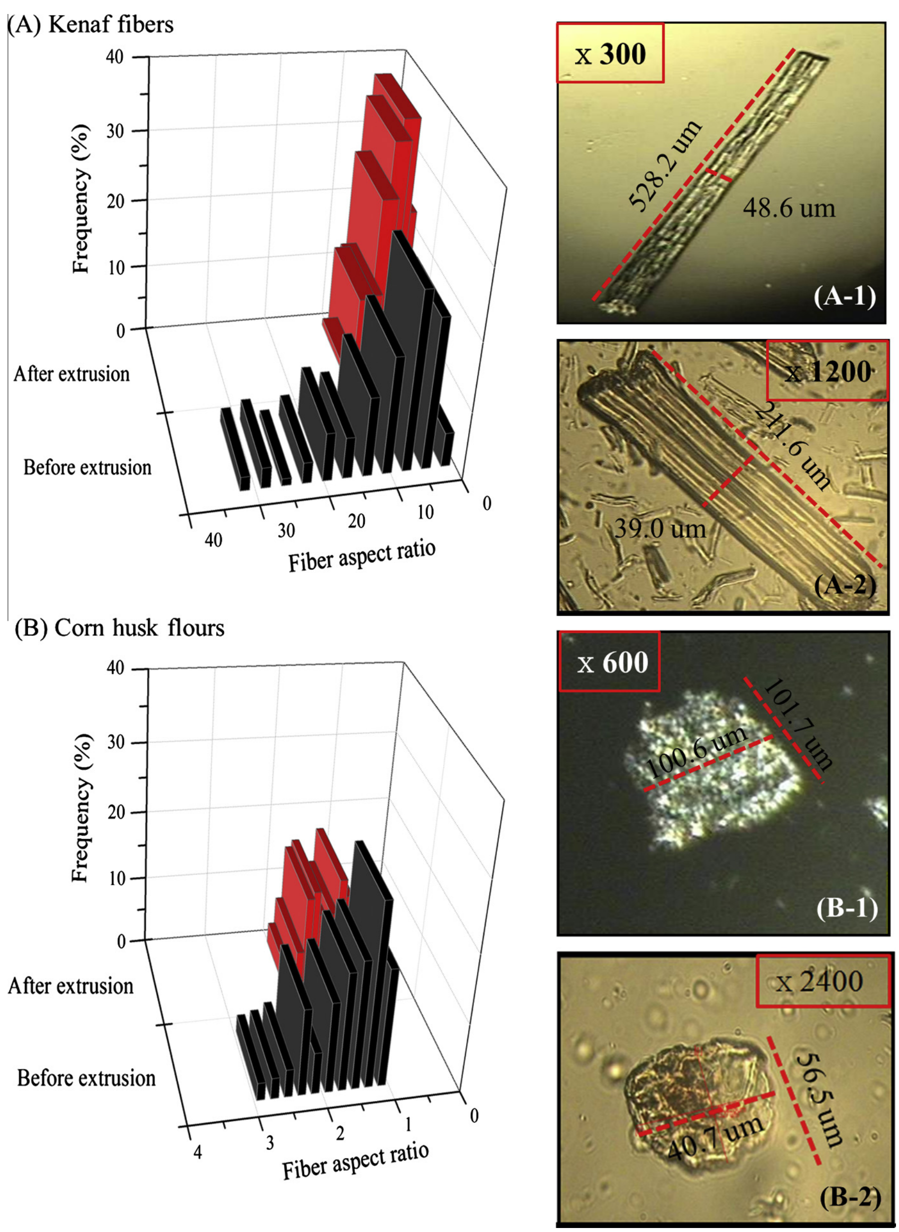

Fig. 2. Variations of the aspect ratio of (A) kenaf fibers and (B) corn husk flours before and after extrusion process; A-1 and B-1 before extrusion, A-2 and B-2 after extrusion.

$$E_{11} = [E_m\{1 + 2(L_{f1}/d_{f1})\eta_{L1}V_{f1}\}/(1 - \eta_{L1}V_{f1})] + [E_m\{1 + 2(L_{f2}/d_{f2})\eta_{L2}V_{f2}\}/(1 - \eta_{L2}V_{f2})] \quad (2)$$

$$E_{22} = [E_m(1 + 2\eta_{T1}V_{f1})/(1 - \eta_{T1}V_{f1})] + [E_m(1 + 2\eta_{T2}V_{f2})/(1 - \eta_{T2}V_{f2})] \quad (3)$$

where η_{L1}, η_{L2}, η_{T1}, η_{T2} were calculated according to the following equation:

$$\eta_{L1} = \{(E_{f1}/E_m) - 1\}/\{(E_{f1}/E_m) + 2(L_{f1}/d_{f1})\} \quad (4)$$

$$\eta_{L2} = \{(E_{f2}/E_m) - 1\}/\{(E_{f2}/E_m) + 2(L_{f2}/d_{f2})\} \quad (5)$$

$$\eta_{T1} = \{(E_{f1}/E_m) - 1\}/\{(E_{f1}/E_m) + 2\} \quad (6)$$

$$\eta_{T2} = \{(E_{f2}/E_m) - 1\}/\{(E_{f2}/E_m) + 2\} \quad (7)$$

where m is matrix, $f1$ is first fiber (kenaf fiber), $f2$ is second fiber (corn husk flours), L_f is length of fiber and D_f is diameter of fiber [15].

The Halpin–Tsai equation is based on the self-consistent field method developed by Hill. But Halpin and Tsai make it simpler analytical form. This equation can be used to predict of elasticity of composite material based on the geometry and orientation of the filler and the elastic properties of the filler and matrix [19].

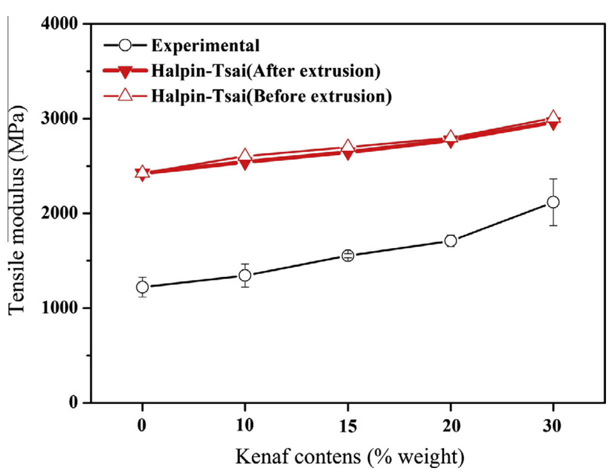

Fig. 3. Comparisons of the predicted tensile modulus before and after extrusion.

The present result exhibited that the ROHM can predict the mechanical properties better than the Halpin–Tasi [15], similarly found in other study [10].

To improve the prediction value by the Halpin–Tsai equation, the aspect ratios of reinforcements used here were re-evaluated as a potential factor because they changed largely after the extrusion process. Fig. 3 compares the predicted values by the Halpin–Tsai equation before and after the extrusion. It was found that there was no significant difference in the predicted values. Therefore it was concluded that the aspect ratio before or after extrusion did not change the prediction so that the initial value of aspect ratio can be used directly to predict the mechanical properties of the present hybrid bio-composite.

3.3. Scale ratio of reinforcing agent aspect ratios to magnitude of mechanical properties

As discussed earlier about the effect of aspect ratio before and after the extrusion process as the possibility to improve the predicted value from Halpin–Tsai equation. Another view of aspect ratio of loaded reinforcing agents will be discussed in this section as the potential value to determine the effect to hybrid composite mechanical properties. To understand the effect of such factor, diverse types of matrix and reinforcements are required.

Table 1 summarizes various hybrid bio-composite systems consisting of different aspect ratios of reinforcing natural fibers with different polymer matrices at various fiber/matrix contents. Here, the scale ratio is defined as the ratio between the aspect ratios of reinforcing fibers with two different dimensions. Table 2 compares their tensile moduli at different loadings in the bio-composites.

Aspect ratio significantly affects the mechanical properties of hybrid composites, because high aspect ratio effectively transfers stress to matrix [21].As shown in Fig. 1, the aspect ratio of kenaf fiber was higher than that of corn husk flour regardless of extrusion process. As a result, the mechanical properties of composites were increased when the amount of kenaf fiber compared to corn husk flour was largely incorporated in composites, as shown in Table 1. Moreover, this phenomenon was observed in not only in the experimental results but also in the prediction model by the Halpin–Tsai equation. The predicted values by the Halpin–Tsai equation exhibited an increase in the modulus with increasing amount of kenaf fiber in composites. The result indicated that one natural fiber having a higher aspect ratio than another played a role as primary reinforcement, and the scale ratio between the high and low aspect ratios of reinforcements incorporated into

Table 1
Hybrid bio-composites with various cellulose-based natural reinforcements.

Polymer matrix type	Fiber/matrix content[a]	Natural fiber with large aspect ratio	Dimension (μm)	Natural fiber with small aspect ratio	Dimension (μm)	Scale ratio[b]	Novelty	Reference
Polypropylene	40/60	Kenaf fiber	177–400 (opening)	Wood flour	149–250 (opening)	1.45	Coupling agent and initiator were used in this composite	[10]
Polypropylene	30/70	Hemp fiber	300–700 (opening)	Hemp powder	45–180 (opening)	4.44	Both large and small agents came from the same material	[13]
Epoxy LY 556	40/60	Sisal fiber	11,000 (length)	Banana fiber	5000 (length)	2.20	Experimental values showed optimum point	[14]
Polyester resin HSR 8131	30/70	Sisal fiber	205 (diameter)	Banana fiber	120 (diameter)	1.71	Layering pattern affect optimum loading to mechanical properties	[20]
Poly(lactic acid)	30/70	Glass fiber	3170 (length)	Cellulose fiber from newspaper	20 (diameter) 850 (length)	3.73	Use of recycled newspaper as a source of cellulose fiber	[5]
Poly(lactic acid)	30/70	Kenaf fiber	49 (diameter) 528 (length)	Corn husk flour	101 (diameter, length)	6.27		Author's own work

[a] Ratio between amount of reinforcing agent loaded and matrix by weight.
[b] Estimated values between dimension of large and small loaded agents based on reported dimension; opening size, diameter, and length.

Table 2
Comparisons between the mechanical properties of hybrid bio-composites and their scale ratios[a].

Matrix material		Reinforcing agents				Scale ratio	Reference
Type	Tensile modulus (MPa)	Type (s)	100% Large object[b](MPa)	50%:50% (MPa)	100% Small object (MPa)		
Polypropylene	820	Kenaf fiber/wood flour	3240 (3.95)[c]	2900 (3.54)	2660 (3.24)	1.45	[10]
Polypropylene	334	Hemp fiber/hemp powder	551 (1.65)	591 (1.77)[d]	571 (1.71)	4.44	[13]
Epoxy LY 556	N/A	Sisal fiber/banana fiber	345 (N/A)	375 (N/A)	288 (N/A)	2.20	[14]
Polyester resin HSR 8131	N/A	Sisal fiber/banana fiber	1185 (N/A)	1443 (N/A)	1312 (N/A)	1.71	[20]
Poly (lactic acid)	2700	Glass fiber/cellulose fiber from newspaper	6700 (2.48)	N/A	5300 (1.26)	3.73	[5]
Poly (lactic acid)	1156	Kenaf fiber/corn husk flour	2117 (1.83)	1547 (1.34)	1221 (1.06)	6.27	Author's own work

[a] All mechanical properties were interpreted from literature.
[b] Reinforce only with fibers with large aspect ratio.
[c] Tensile modulus ratios – ratio between tensile modulus of hybrid composite and matrix material – are shown in bracket.
[d] Compositions are at 60% large object volume fraction.

the hybrid bio-composite needs to be considered to optimize the mechanical properties of composites.

As shown in Fig. 4, each bar represents ratio of modulus values of specific scale ratio at different proportion of large reinforcing object in composite [2–4]. For all the case of large object composition, as scale ratio decrease or the difference between dimensions of reinforcing object becomes small, increase of mechanical properties as compared to bare matrix is greatly pronounced. While at large scale ratio, improvement of modulus did not significant. As mentioned earlier about the effect of reinforcing agent that higher aspect ratio can shift stress into matrix better than reinforcing agent with small aspect ratio. This justified the hypothesis in this study that a hybrid composite consisting of reinforcements with similar dimensions, resulting in a low scale ratio, can transfer external force more effectively and exhibit higher modulus, regardless of the polymer matrix and the reinforcement type used. Consequently, the present study provides a guideline that use of reinforcements with a similar scale ratio may be desirable for improving the mechanical properties of a hybrid bio-composite.

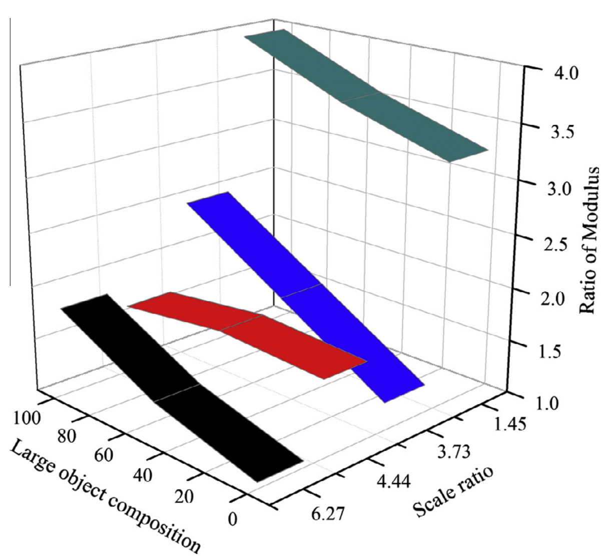

Fig. 4. Variations of the ratio of the tensile modulus of different bio-composites as functions of the large object composition and the scale ratio. (blank stripe – Author's own work, red stripe – Ref. [4], blue stripe – Ref. [2], and greenshied blue – Ref. [3]). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

4. Conclusion

Prediction model is assumed that no interaction in each other in hybrid composite, but the difference of Young's modulus of fibers can practically influence the stress transfer from matrix to fiber as found. So, there is difference between theoretical and experimental values. To improve the prediction value, the aspect ratios of reinforcements were re-evaluated as a potential factor because we expected that they changed largely after the extrusion process. But it was found that using aspect ratio of reinforcements after extrusion process could not significantly reduce gap between mathematical prediction and experimental modulus of hybrid composite. Therefore, to predict hybrid composite properties of this system, the aspect ratio of reinforcements before extrusion process can be used directly. Descriptive conclusion pointed out a trend of new factor a scale ratio – proportion between sizes of reinforcements – exhibited the potential to be an important factor when optimizing hybrid composite properties despite the fact that such conclusion was got from separated resources.

In a future study, it would be fascinating to see a more detailed study on the effect of different portion and sizes of reinforcements as in scale ratio to the overall mechanical behavior of biodegradable hybrid composite, such that a systematically comparison and use as a guideline for optimizing hybrid composite properties.

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