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In the last decades, filler-reinforced thermoplastics especially natural filler-reinforced plastics have been frequently used to improve the physical and thermal properties of polymer materials in plastic industry due to their low density, fully degradable, helpful to reduce the CO_2 emission, free from health hazard and low cost. At current study, wood powder/polypropylene composites (wood/PP) with different filler contents were molded by injection molding process to investigate the effect of filler content on the physical and mechanical properties of the composites. Additionally, the comparison of physical and mechanical properties between talc/PP (which has been widely used in the automotive products) and wood/PP has been carried out based on the tensile, bending, Izod impact tests and the scanning electron microscope observation on the fracture surfaces. Results showed that the highest mechanical property of wood/PP was determined at 30 wt.% wood content. More interesting is that, at the same composite density up to 30 wt.%, the mechanical property of wood/PP was much higher than that of talc/PP. Theoretically, Nielsen equation is often used to predict the elastic modulus of filler reinforced plastics, and in this study, the predicted values were in good agreement with experimental values up to 30 wt.%, after that, they were higher to the experimental values of wood/PP composite. It is considered that is due to the discounted of the distribution and orientation of the filler in the matrix in Nielsen equation.

Keywords: injection molding; filler content; talc; wood; thermal conductivity; modulus prediction; Nielsen equation

1. Introduction

During the last few years, polypropylene (PP) incorporated with particulate fillers has been gained high interests. Since as well known, PP not only exhibits excellent stiffness property and environment adaptability, but also has a good processability allowing accepting different types of fillers. It is known that the fillers are the most often used materials to reduce the production costs and optimize the production properties such as rigidity, strength, dimensional stability, crystallinity, electrical, thermal conductivity, and so on. Although fillers also have a detrimental effect on other properties of filler reinforced thermoplastics, such as the impact properties and deformability, incorporating fillers, can help to increase the application areas of PP. Among all the fillers, talc is one of the widely used to stiffen thermoplastics, typically for PP. Talc can be used in both

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various and small amounts. In various amounts (over 20% by weight), it can improve the heat deflection temperature, thermal conductivity, and stiffness of the thermoplastics. On the other hand, when talc is used under 3% by weight, it is no longer considered as filler but as a nucleating agent, which can increase the starting crystallization temperature, inducing a very short processing time in injection. The main applications for talc filled PP at a comparative highly filled system are automotive parts like heater casing, interior and exterior decoration, fan case and bumper parts, household appliances and engineering plastics. Compared with the polymers, talc has significant higher thermal conductivity, and the heat introduced and generated during processing is transmitted through the composite more quickly. Incorporating talc in PP would increase the thermal conductivity, resulting in faster production rates. However, the heat is also transported out of the composite faster during cooling or heating. As a material which would like to use in the automotive areas, it may decrease the air condition efficiency, and consequent to a low fuel economy.[1–5]

On the other hand, owing to low prices, low density, ecological and economical advantage, less health hazards, and the steadily rising performance of technical and standard plastics, the demand for natural filler incorporate composites has been increased in various application areas such as automotive components, building materials, and electrical engineering. In particularly, more recently, the critical discussion about the preservation of natural resources and recycling has led to a renewed interest concerning natural materials with the focus on renewable raw materials. One of the most common natural fibers used to reinforcing thermoplastics is wood powder. The use of wood as the reinforcement of the composites has received increasing attentions recent years both by the academic sector and by the industry as inexpensive filler to increase the mechanical properties and thermal stabilities of thermoplastic or to reduce raw material costs. Because the wood flour can be derived from sawdust, pulp mill wood residue, bark, nutshell, and straw. Therefore, as potential applications of the composites, wood-reinforced thermoplastic composites are commercially attractive for high-volume applications. In particularly, compared with talc, besides the biomass nature of wood, wood-filled PP is more attractive nowadays not only because the density of wood is lower, but also the thermal conductivity is much lower than talc.[6–11]

The most prominent of physical effects of fillers is the stiffening or modulus increase that they caused in composites. For particulate-filled composites, a number of equations for the prediction of the modulus have been proposed with different methods. Various models for the prediction of elastic modulus have been developed under an ideally uniform distribution of the filler in matrix, and the interface adhesion between filler and matrix is very good. Thus, no filler concentration, distribution, and interfacial condition will be considered in the equations. However, this is not the real case. In reality, no particle would distribute homogenously and own perfect interfacial adhesion between filler and matrix. Especially, for natural filler, the poor adhesion between the hydrophilic natural filler and hydrophobic thermoplastic is always one of the problems researchers considered to overcome to attain high mechanical properties. Therefore, considering the concentration, distribution and interfacial adhesion of the filler in polymer in elastic modulus prediction model can be a major processing challenge.[12–17]

At current study, wood powder reinforced polypropylene composites (wood/PP) with different filler contents were molded by injection molding process to investigate the effect of filler content on the physical and mechanical properties of the composites. Additionally, the comparison of physical and mechanical properties between talc/PP and wood/PP has been carried out. Moreover, the theoretical values of the elastic modulus of the wood/PP composite were calculated by Nielsen equation and compared with the experimental data.

2. Experimental

2.1. Materials

PP pellet, as block polymer, with a melt index of 15 g/10 min (J-784HV, Idemitsu Kosan Co., Ltd) was used as matrix because it is a widely used plastic in the automotive industry that can be easily injection molded. Talc (Asada Seihun, JM-209P) and wood powder that is soft wood type from cryptomeria japonica (from trees in Hiroshima prefecture, Japan) were used as reinforcements to fabricate the talc/PP and wood/PP dumbbell shape specimens by injection molding method. Wood powder was made into pellets with water or some additives by granulation machine supplied by Dalton Co., Ltd. Crystalline polyalpha olefin (CPAO) that works as binder used during wood pellet fabrication process was supplied by Idemitsu Kosan Co., Ltd.

Figure 1 displayed the scanning electron microscope (SEM) observation of wood powder. It is noteworthy that the shape of the wood powder used in this study is not sphere but much like fiber shape. Based on the measurement, the aspect ratio (L/D) of wood powder is 8.22. In addition, the particle size distribution of wood powder was measured by using an ultrasonic vibration particle size distribution sieving machine (Seishin Enterprise Co., Ltd). Result showed the average particle length by mass of the wood powder used in this study was 51.3 µm. Particularly, the particle lengths of more than 33% of the wood powder were less than 38 µm.

CPAO is a kind of olefin, and it is known that olefin which also called alkene is an unsaturated chemical compound containing at least one carbon-to-carbon double bond. Figure 2 showed the chemical structure of CPAO. CPAO gives a new region of polyolefin properties by using new single site catalyst. The melting point of CPAO was controlled in a range of 28–50 °C, and the density was around 900 kg/m^3.

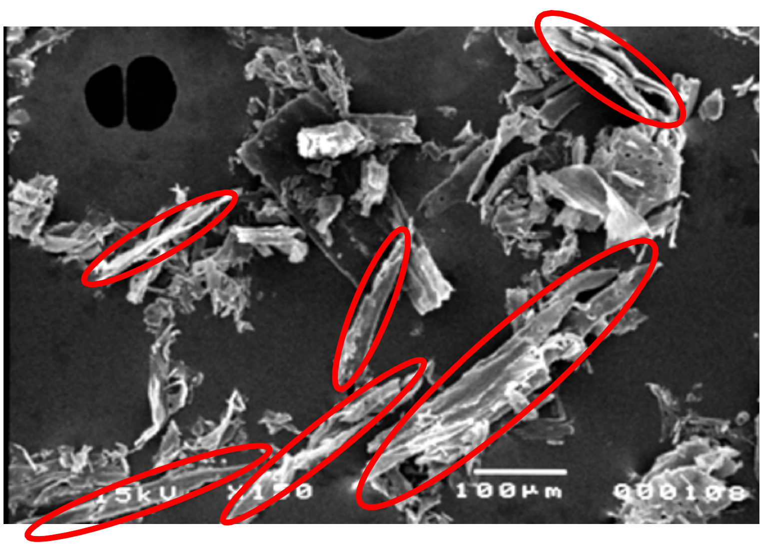


Figure 1. SEM observation of wood powder.

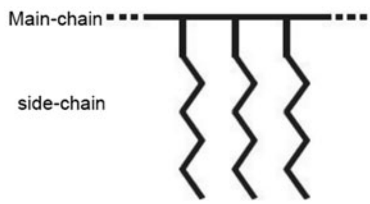


Figure 2. Chemical structure of CPAO.

2.2. *Injection molding*

Nine kinds of dumbbell shaped specimens were prepared as listed in Table 1. In details, three kinds of talc/PP and three kinds of wood/PP with different filler content of 15, 30, and 45 wt.%, respectively; On the other hand, for C1–C3 specimens, wood contents were 51 wt.%, wood was mixed with different CPAO contents during powder to pellet fabricating process. The maleated polypropylene (MAPP, Toyotac H-1000P) was used as coupling agent to improve the interfacial properties of wood/PP in this study. All of the dumbbell test pieces were fabricated by using a 30-ton TOYO PSS TT-30F6 injection molding machine at a barrel temperature between 150 and 210 °C. Here, it should be mentioned that before injection process, wood was dried in an oven set at 90 °C for at least 16 h to remove the moisture.

2.3. *Tensile test*

For tensile test, the gauge length is 115 mm, and strain was measured using extension measurement, which was fixed onto the central surface of the specimens by a rubber band. Tensile test was conducted on an Instron Universal Testing Machine (Type 4206) under a nominal test speed of 1.0 mm/min. Five pieces were repeated.

2.4. *Point bending test*

Three-point bending test was conducted on an Instron Universal Testing Machine (Type 4206) under a nominal test speed of 1.0 mm/min. Five specimens were repeated. The span length of the bending specimen is 48 mm, and the width is 10 mm.

Table 1. Material composition.

Specimen ID	PP (wt.%)	Talc (wt.%)	Wood powder (wt.%)	CPAO (wt.%)	MAPP (wt.%)
HCR-1	85	15	0	0	0
HCR-2	70	30	0	0	0
HCR-3	55	45	0	0	0
HCR-4	83	0	15	0	2
HCR-5	68	0	30	0	2
HCR-6	53	0	45	0	2
C1	47	0	51	0	2
C2	44.3	0	51	2.7	2
C3	41.3	0	51	5.7	2

2.5. *Izod impact test*

Izod impact test was performed on the Impact tester (Toyoseiki) pendulum 5.5 J in accordance with ASTM D 256-05. The size of the specimen was 10 mm × 60 mm, V-notch was used. Five specimens were repeated.

2.6. *Thermal conductivity test*

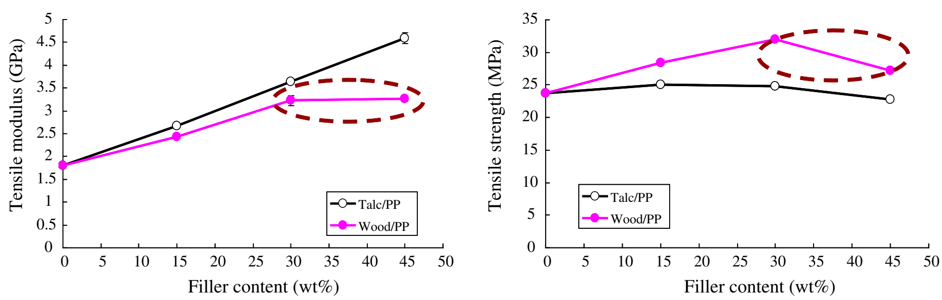
Thermal conductivities of specimens were tested with the TPA-501 thermal conductivity instrument (Kyoto Electronics manufacturing Co., LTD).

3. Experimental results and discussion

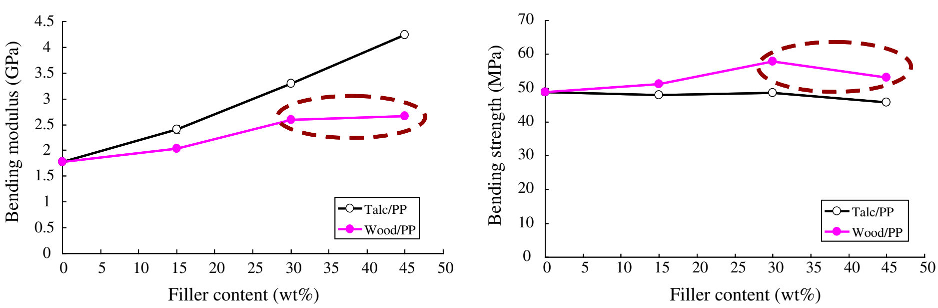
3.1. *Comparison between talc/PP and wood/PP*

3.1.1. *Mechanical properties of talc/PP and wood/PP*

The comparison of mechanical properties between talc/PP and wood/PP with different filler contents was graphically presented in Figure 3. It is noteworthy that for talc/PP, both tensile and bending modulus increased significantly with the increasing of talc content, this is caused by the fact that the reinforcing filler usually displays considerably more rigid phase compared with the polymer matrix. While for the tensile and bending strength, it was found that increasing the talc content resulted in a slight decrease in the strength. That was considered due to the weak adhesion between PP matrix and talc filler. On the other hand, for wood/PP, both tensile and bending properties increased with the increasing in wood content up to 30 wt.%. After then, the tensile and bending properties started to decrease especially the strength. It is well known that the type of



(a) Tensile properties



(b) Bending properties

Figure 3. Mechanical properties of talc/PP and wood/PP as a function of the filler content.

filler, distribution of filler and interaction between matrix and filler are the most important factors affecting the mechanical properties of filler reinforced composites. In this study, the agglomeration of the wood powder, poor dispersion of the wood powder in the matrix is considered the main reasons which led to the decrease in mechanical properties in high wood content.

It is known that the researches on using natural filler-reinforced thermoplastics instead of synthetic filler-reinforced thermoplastics have been received increasing attentions recent years, the optimizations of using natural filler are the weight savings, decrease the cost without reducing the rigidity of the composites and so on. As products used in automotive or aerospace application areas, the lighter the material will be, the higher fuel efficiency will attain. Although wood/PP did not show advantages when compared with talc/PP at the same filler content, the differences are clear when compared at the same composite density. The changes in the mechanical properties of talc/PP and wood/PP as a function of the composite density were shown in Figure 4. Here, the density of the composite has been calculated by using the density of each component. Densities of talc, wood and PP were 2.7, 1.4 and 0.9 g/cm^3, respectively. It is interesting that both tensile and bending properties of wood/PP showed higher values compared to talc/PP at the same composite density below 30 wt.% filler content. That is to say, for two composites with the same tensile property, the weight of wood/PP composite will be much lighter than talc/PP.

Izod impact properties of talc/PP and wood/PP as a function of the filler content were presented in Figure 5. The Izod strength for both filler reinforced composites decreased with the increasing in the filler content. After incorporating 15 wt.% filler,

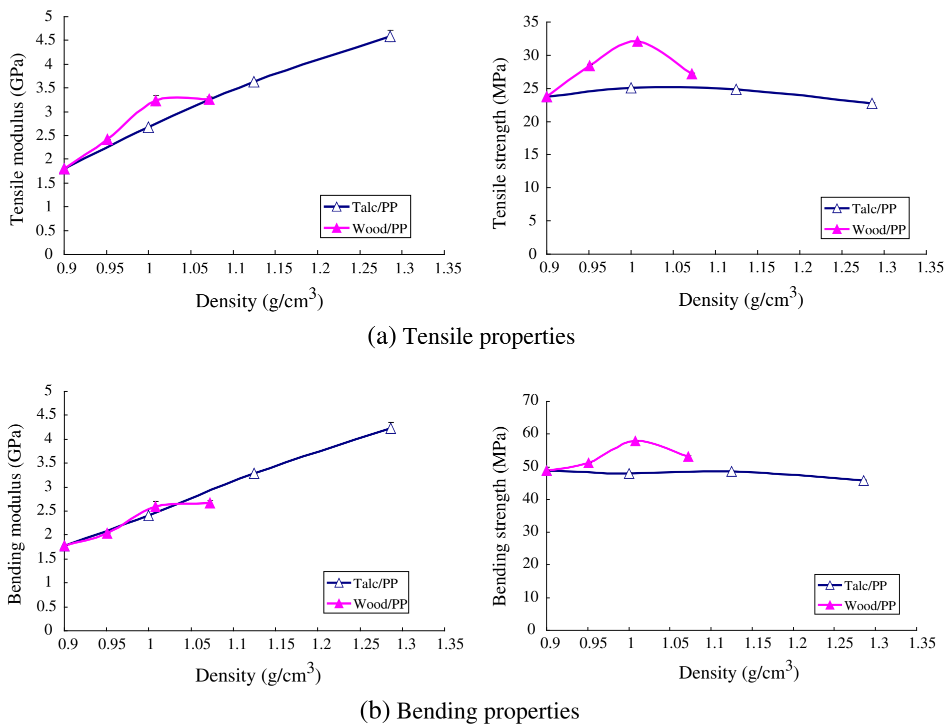


Figure 4. Mechanical properties of talc/PP and wood/PP as a function of the composite density.

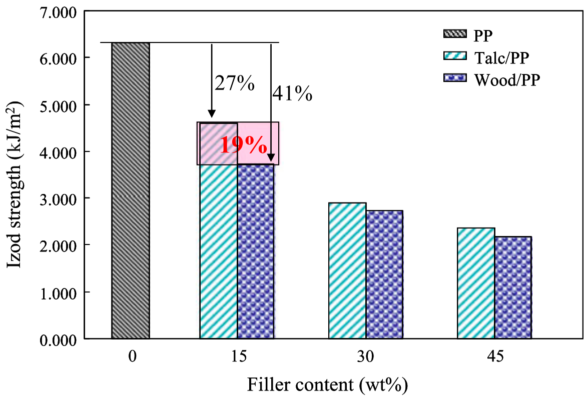


Figure 5. Izod impact properties of talc/PP and wood/PP as a function of the filler content.

compared with the virgin PP, the Izod strength of talc/PP and wood/PP decreased 27 and 41%, respectively. It is apparent that the toughness of the composite decreased after incorporating the filler. The immobilization of the matrix by the filler so that the latter fails to deform before failure is considered as one of the reasons leading to the decreasing of the Izod strength. Additionally, poor wetting of the powder by PP, giving rise to poor interfacial adhesion between the filler and the polymer matrix resulting in weak interfacial regions is also considered one of the reasons. From the results, it also can be seen that the decreasing for the Izod strength of wood/PP composite is much severer compared with talc/PP. It is noted that at 15 wt.% filler content, wood/PP showed 19% lower Izod strength than that of talc/PP. That maybe due to the higher tendency of wood to form the filler agglomerates, resulting in a poor dispersion of the fillers in the matrix compared with talc and weaker interfacial adhesion between wood powder and wood powder and polypropylene because the hydrophilic nature of the wood powder.

SEM observations were carried out to examine the wood powder distribution state in the composite. In order to give a more distinct detection of the wood powder, etching process was performed before SEM on the polished specimens. CRP-MARS etchant provided from Okuno Chemical Industries Co., Ltd was used as etching solution. The fracture surface observations after etching process of wood/PP with different wood contents were shown in Figure 6, it was obviously that after etching, the wood powder was etched and left behind lots of voids on the fracture surface. Those voids distribution of the specimen after etching was considered equal to wood distribution. It is obviously that at high wood powder content composite (45 wt.%), wood powder agglomerated in some regions and the distribution of the powder in matrix became worse and worse with increasing of the wood powder content. These observation results were equal to the consideration leading to the decreasing of the mechanical properties of composite at high wood filler content.

3.1.2. Thermal conductivities of talc/PP and wood/PP

Figure 7 showed the thermal conductivity properties of both talc/PP and wood/PP composites. As can be seen from the figure, thermal conductivity for talc/PP composite increased with the increasing of the talc content. However, for wood/PP composite, the wood content didn't affect the thermal conductivity of the composite dramatically. That is considered due to the significantly higher thermal conductivity of the talc

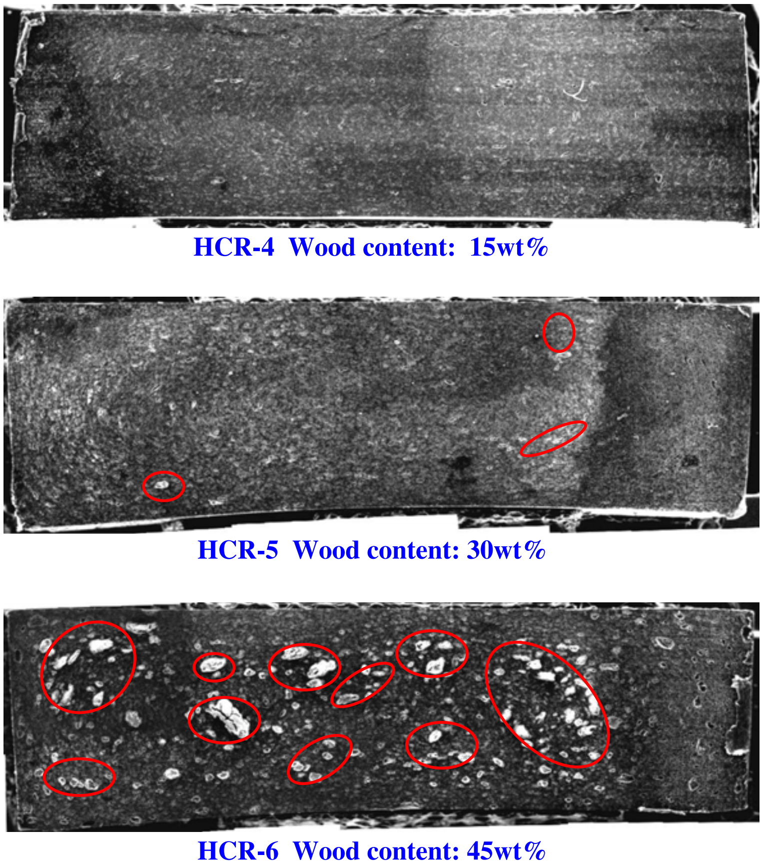


Figure 6. Fracture surface observations after etching process of wood/PP with different wood content.

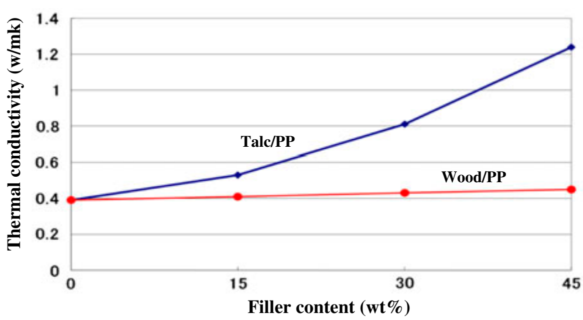


Figure 7. Thermal conductivity properties of both talc/PP and wood/PP composites as a function of filler content.

compared to the wood powder. Low thermal conductivity at high filler content, that means, the heat is transported out of the composite slowly, when the production is used in automotives, the air condition efficiency is increased, and consequents to a high fuel economy. That is also one of the advantages by using natural fillers instead of synthetic fillers.

3.2. Modulus prediction of wood/PP by using Nielsen equation

In this study, the elastic modulus of the wood/PP composite was predicted by using Nielsen equation which was shown as follows:

$$\frac{M}{M_1} = \frac{1 + ABV_f}{1 - B\psi V_f}$$

$$A = K_E - 1$$

$$B = \frac{M_2/M_1 - 1}{M_2/M_1 + A}$$

$$\psi = \left(\frac{1 - P_f}{P_f^2}\right) V_f$$

M, M_1, and M_2 are modulus of the composite, matrix, and filler respectively; K_E is the Einstein coefficient; V_f is the fractional filler volume; and P_f is the packing fraction of the filler at maximum filler concentration. The constant K_E accounts for the shape of the filler particles and Poisson's ratio of the matrix. In this study, $K_E = 3.9$ and $P_f = 0.53$.

The predicted results for elastic modulus by using Nielsen equation were shown in Figure 8. It is found that the predicted elastic modulus were in good agreement with experimental values up to 30 wt.%. However, after that, as filler content increased, Nielsen equation tends to overestimate the elastic modulus of the composite. That is because Nielsen equation appears to be more relevant to materials with low concentrations of noninteractive spheres. Therefore, it is considered the inappropriate predicting modulus at high filler content is due to the aggregation and poor distribution of the wood powder in the PP matrix.

In order to prove the above consideration, CPAO was used in this study. In the previous research,[18] it was found that the distribution of wood powder was improved

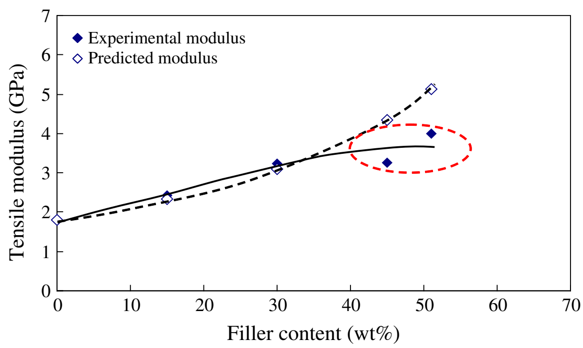


Figure 8. Correlation between experimental and prediction elastic modulus by using Nielsen equation.

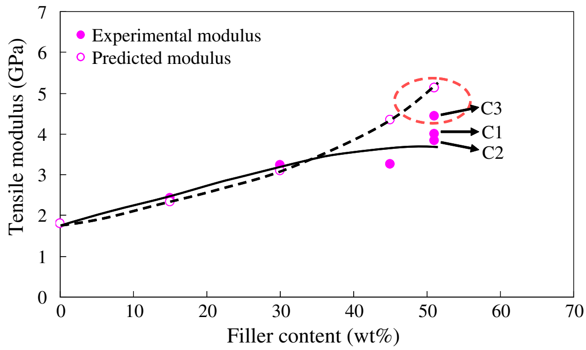


Figure 9. Comparison between the experimental results of 51 wt.% wood powder-reinforced wood/PP with different CPAO content and prediction elastic modulus by using Nielsen equation.

with increasing the CPAO content. Furthermore, the size of the wood powder was also much better uniform of composite with 5.7 wt.% CPAO. Figure 9 showed the comparison between the experimental results of 51 wt.% wood powder reinforced PP with different CPAO content and prediction elastic modulus by using Nielsen equation. It is noted that with the better distribution of wood in PP matrix, the predicted elastic modulus tends to approach the experimental result. Especially for the composite with 5.7 wt.% CPAO, the predicted result is in good agreement with the experimental result. That is to say, Nielsen equation is suitable in predicting the modulus of filler reinforced composite only when the filler distributed well in the matrix.

4. Conclusion

At current study, wood/PP composites with different filler contents were molded by injection molding process to investigate the effect of filler content on the physical and mechanical properties of the composites. Additionally, the comparison of physical and mechanical properties between talc/PP and wood/PP has been carried out. Moreover, the theoretical values of the elastic modulus of the wood/PP composite were calculated by Nielsen equation and compared with the experimental data.

(1) The tensile and bending modulus for both talc/PP and wood/PP increased with the increasing of the filler content. However, for wood/PP composite, at high wood content, the tensile and bending properties started to decrease. The agglomeration of the wood powder is considered the main reason led to the decrease.
(2) Both tensile and bending properties of wood/PP showed higher values compared with talc/PP at the same composite density below 30 wt.% filler content. That is to say, at the same tensile modulus or strength, the weight of wood/PP composite will be lighter.
(3) The Izod strength for both talc/PP and wood/PP composites decreased with the increasing of the filler content. It is apparent that the toughness of the composite decreased after incorporating the filler and thereby led to the decrease of the Izod strength.

(4) Thermal conductivity for talc/PP composite increased with the increasing of the talc content. However, for wood/PP composite, the wood content did not affect the thermal conductivity of the composite significantly.
(5) It is noted that the distribution of the wood in PP matrix is important during predicting the modulus of wood/PP composite. Nielsen equation is suitable in predicting the modulus of filler reinforced composite only when the filler distributed well in the matrix.

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