The Effects of Rice Straw Weight Fraction and Particle Size on Thermal Conductivity and Mechanical Properties of Polypropylene Composite

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Abstract
Polypropylene is a widely used material for various types of equipment. As a recyclable thermoplastic polymer, polypropylene composite provides an attractive quality of thermal insulation due to its heat-retaining property. The use of rice straw for filler in thermal insulating material is based on its low thermal conductivity, renewable material, and low cost production techniques. The composite were manufactured by melt-mixing of neat polypropylene and filler of various weight fractions, pellet formation, followed by reprocessing with injection molding. The current work focused on determining the effect of filler’s weight fraction, particle size and pretreatment on the thermal conductivity and tensile strength of the composite. The result showed that an increase of filler’s weight fraction up to 30% decreased the composite’s thermal conductivity and tensile strength gradually. Composite of fine particles filler (325 mesh) gave a lower thermal conductivity and higher tensile strength than that of coarse particle (18-35 mesh). Composite with the lowest thermal conductivity of 1.39 W/mK was obtained at 30 w.% filler content and fine particles filler (325 mesh). Improvement on composite properties achieved by acetylated rice straw which enable to reduce straw’s hydrophobicity and enhance adhesion between matrix and filler.

Keyword: acetylation, composite, polypropylene, rice straw, thermal insulation.

1. Introduction
Polypropylene is a superior type of polymer, which is widely used for various products, such as containers, fibers, and composites for outdoor or indoor applications due to its properties of flexibility, hydrophobicity, semi-transparent, and easily dyed. The volume of Indonesia’s waste reached 68 million tons, 14% of which is plastic, which put Indonesia as the second-largest contributor to marine plastic pollution after China [1]. At the UN’s first Ocean Conference (June 2016) Indonesia pledged to reduce plastic debris by 70% by 2025. To overcome the environmental problems caused by polypropylene waste, it is necessary to utilize them into efficient and economically-added products such as thermal insulation composites. The thermal conductivity range of insulation material is 0.034 - 0.21 W/m.K [2], whilst thermal conductivity of Polypropylene at 25 °C is 0.22 W /m.K [3].

The ideal insulation material must have the characteristic to withstand heat and rigidity. To obtain the stiffness and thermal insulation properties of polypropylene it is necessary to add filler material [4]. Fillers that have been used include natural fibers due to their hollow and cellular nature, which perform as acoustic and thermal insulators. In order to obtain composites with superior properties, including strong, lightweight, non-abrasive, able to withstand heat, cheap and environmentally...
friendly it is necessary to select the type and particle size of filler. When cellulosic fiber is compounded with thermoplastic polymers, the main problem encountered is the poor interfacial adhesion between the hydrophobic polymer and hydrophilic filler, poor resistance to moisture absorption and microcracking of composites, resulting in limited composite applications. The adhesion between the reinforcing fibers and the matrix in composite material plays an important role in final thermal properties of the composites. Coupling agents have been added to improve the dispersion, adhesion and compatibility between the hydrophilic cellulose and hydrophobic matrix.

Recently, silica has been explored as filler for polymeric materials and it holds a great potential for improving mechanical properties of polymers. The previous study used modified silica (with organosilane agents) as a nanofiller on neat polypropylene composites, which resulted better surface compatibility and better thermal stability (pure polypropylene fibers decompose at 416 °C, whereas the hydrophobic silica filled fibers begin to decompose at a relatively higher temperature of 430°C) [5].

The paper was focused on the influence of composition, particle size, and pretreatment of rice straw filler on composite’s thermal insulation and mechanical properties.

2. Experimental
2.1 Materials
Neat Polypropylene chips type HI10HO with specific gravity of 0.9, melt flow rate of 10 g/10 min was provided by PT Candra Asri Petrochemical, rice straw was obtained from rice field in Cimahi, West Java. Xylene (technical grade), anhydride acetic acid were obtained from chemical store in Bandung, Indonesia.

2.2 Methods
The rice straw fiber was dried in sun light, immersed in water, redried in an oven at 105°C to reduce water content to 5%w, then it was cut to 1cm, being ground and sieved to two different sizes (18-35 mesh or 325 mesh or 45 μm). The compounding process used for preparing rice straw (RS) fiber/PP composites included two steps. First step, weighed RS particles and PP/xylene (with ratio of 1g PP to 5ml xylene) were melt-mixed at 160°C for 30 minutes in oil bath, until homogeneity was obtained, then the hot agglomerate were transferred to the shallow cast (2mm deep), then cooled down to room temperature in the fume hood, and let the remaining xylene to evaporate, then the dry sheet was cut into pellets of 2mm . In the second step the composite pellet was injection molded (pressure of 5bar, temperature of 170-190°C, for 15 minutes) to form specimens of a certain shape and dimension according to the testing standard. Cooling of the specimen was done at room temperature, then the specimen was removed from the mold. The specimen was then tested for thermal conductivity, tensile properties and morphology using Scanning Electron Microscopy.

In order to investigate the improvement of interfacial adhesion between matrix and filler, the rice straw was treated by acetylation prior to composite mixing. Acetylation process was started by immersing rice straw in demineralized water for an hour, then dried in oven for 1.5 hour at 105°C. The straw was then reacted with acetylation reaction showed in Figure 1.
2.3 Characterization

2.3.1 Thermal conductivity. The specimen thermal conductivity was measured by using thermal conductivity analyzer. Value of heat that absorbed by composite was calculated by formula:

\[ Q_{\text{cond}} = -kA \frac{\Delta T}{x} \]

where

- \( Q_{\text{cond}} \): heat that absorbed by composite (Watt)
- \( x \): thickness of composite (m)
- \( k \): thermal conductivity of composite (W/m.\( ^\circ \)C)
- \( A \): area of heat transfer (m\(^2\))
- \( \Delta T \): Temperature difference (\( T_{\text{composite}} - T_{\text{room}} \))

Testing was conducted for cylinder specimen with diameter of 5 cm and thickness of 5 mm.

2.3.2 Tensile Test. The tensile strength was measured on a caest pendulum impact based on ASTM E8/E8M-09 standard.

2.3.3 Scanning Electron Microscope (SEM). The as prepared composites were morphologically inspected by using SEM in order to observe filler distribution and adhesion between filler and matrix.

3 Results and Discussion

3.1 Effect of Filler Content on Thermal Conductivity of Composite

Figure 3 shows the thermal conductivity curves with rice straw weight content. It is observed that incorporation of microparticles with relatively low thermal conductivity into polypropylene matrix resulted in gradual reduction of composite thermal conductivity. These results are in agreement with those obtained by previous study as the filler material (fly ash) is added to the composite the thermal conductivity of the composite reduces as compared to that of the pure matrix (epoxy) [6].
3.2 Effect of Filler Size on Thermal Conductivity of Composite

The effect of filler particle size on thermal conductivity of composite was conducted by particle sizes of 325 mesh and 18-35 mesh with filler content of 10% w.

Figure 4 shows that composite with coarser filler (18-35 mesh) has higher thermal conductivity than that of fine filler (325 mesh). The small reduction on the thermal conductivity due to the fact that coarser filler was isolated (no inter-filler networks was formed). The fine filler give a better effect due to better particle distribution in the matrix.

3.3 Effect of Filler - Matrix Adhesion on Thermal Conductivity of Composite

The effect of filler-matrix adhesion on thermal conductivity of composite was conducted by acetylation of rice straw which is then milled to 325 mesh.

Figure 5 shows that composite with acetylated rice straw filler has lower thermal conductivity than that of untreated filler (filler content of 10% w). The filler - polypropylene matrix adhesion improved remarkably due to acetylation, which change the rice straw fiber surface morphology (it is proven by SEM morphological test of the composites).
Figure 5 Effect of filler-matrix adhesion on Thermal Conductivity of Composite

3.4 Effect of Filler Content on Tensile Strength of Composite

The effect of filler content on tensile strength of Composite was conducted with filler particle size of 325 mesh.

![Thermal Conductivity Graph]

Figure 6. Effect of Filler Content on Tensile Strength of Composite

Addition of rice straw filler tends to decrease tensile strength of the composites, the higher the filler content the weaker bond between matrix and filler. Decreasing of tensile strength with addition of rice straw filler was conducted with research conducted by Grozdanov et.al (2006) about polypropylene composite with rice straw filler [4]. These results are also in agreement with those obtained by Shao et.al (2008) over the same range of filler size, that the composite strength is reduced with increasing particle loading [7].

Decreasing tensile strength of composites with filler content of 10% w (deviates from trend) caused by air bubble trapped in the composite. An ideal composite is massive, with no bubble inside (Figure 7.a). The bubbles were formed (Figure 7.b) causing weak spot inside the composite, and decrease the effective cross section of specimen which in turn decrease the tensile strength.

![Composite with no Bubble and Composite with Bubble]

Figure 7 (a) Composite with no Bubble, (b) Composite with Bubble
3.5 Effect of Filler Particle Size on Tensile Strength of Composite

The effect of filler particle size on tensile strength of composite was conducted with filler content of 10% w (particle of 325 mesh and 18-35 mesh).

![Figure 8. Effect of Particle Size on Tensile Strength of Composite](image)

Figure 8 shows that composite with coarser filler has higher tensile strength than that of fine filler. The previous study suggest that PP composites with microparticles show a decrease in strength as the particle size increases [7]. The anomaly accured because the composite with fine filler was contaminated by neat polypropylene during molding process.

3.6 Effect of Rice Straw Acetylation on Tensile Strength of Composite

Figure 9 shows that composite with acetylated rice straw has a higher tensile strength than that of untreated rice straw. Tensile strength enhancement of the composite due to good interface adhesion between polypropylene matrix and rice straw filler. It is fit with Bledzki et.al, research that showed the effect of acetylated rice straw on adhesion improvement between polypropylene and rice straw in composite [8]. These results are also in agreement with those obtained by Abdallah et al. (2009) that PP composite filled with silane-treated cork has a higher tensile modulus than that of untreated cork [9]. Whilst Çelebi (2017) found that PP Composites filled with silane-treated Hollow Glass Microsphere enhance the interfacial region between HGM and polymer matrix [10].

![Figure 9. Effect of Rice Straw Acetylation on Tensile Strength of Composite](image)
3.7 Composite Morphology Analysis
Morphology test of composite was conducted by means of Scanning Electron Microscope (SEM) to observe filler particle distribution, and the influence of filler acetylation on interfacial adhesion between matrix and filler.

3.7.1 Effect of Particle Size on Filler Particle Distribution in Composite. Filler particle distribution for rice straw with size 18-25 mesh and 325 mesh in filler proportion 10% showed in Figure 10.

From the SEM micrographs on Figure 10.a, it is evident that composite with filler size of 18-35 mesh (with 80 X magnification -in red circle) has bigger void than that of filler size of 325 mesh (Figure 11.b with 2000 X magnification). The bigger the void decrease tensile strength of composite. It was also observed that the smaller-sized filler are distributed more uniformly in the matrix than the larger particles.

3.7.2 Effect of Straw Acetylation on Adhesion Between Filler and Matrix. Morphological analysis of composite with acetylated and non acetylated rice straw was conducted with filler particle size of 325 mesh.

Figure 11 shows that composites with acetylated straw fillers has intensive interface adhesion, where there is no void between the straw and polypropylene, whereas in non-acetylated straw (b) void was formed between the straw and the polypropylene. The strong adhesion between polypropylene and straw increases the tensile strength and decreases the thermal conductivity of composite.
4. Conclusion
The rice straw loading in polypropylene resulting in a decreasing thermal conductivity of composite, which range was considered as thermal insulator. The higher the straw loading the lower the thermal conductivity. Composite with finer filler size has higher tensile strength and lower thermal conductivity than that of coarser filler size. The composite thermal insulating property could be improved by acetylated straw filler, which has a higher tensile strength and lower thermal conductivity than that of non acetylated straw.

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