

## Flame Retardancy and Mechanical Properties of Kenaf Filled Polypropylene (PP) Containing Ammonium Polyphosphate (APP)

(Kerencatan Nyalaan dan Sifat Mekanik Kenaf Berpengisi Polipropilena yang Mengandungi Ammonia Polifosfat (APP))

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### ABSTRACT

*The effects of ammonium polyphosphate (APP) as flame retardant and kenaf as fillers on flammability, thermal and mechanical properties of polypropylene (PP) composites were determined. Test specimens were prepared by using a co-rotating twin screw extruder for the compounding process followed by injection molding. The flame retardancy of the composites was determined by using limiting oxygen index (LOI) test. Addition of flame retardant into kenaf-PP composites significantly increased the LOI values that indicated the improvement of flame retardancy. Thermogravimetric analysis was done to examine the thermal stability of the composites. The addition of kenaf fiber in PP composites decreased the thermal stability significantly but the influence of APP on thermal properties of the kenaf-filled PP composites was not significant. The flexural strength and modulus of the composites increased with the addition of APP into kenaf filled PP composite. The addition of APP into kenaf filled PP causes increase in the impact strength while increasing the APP content in the kenaf filled PP composite show decrease in impact strength.*

*Keywords: Flame retardancy; mechanical properties; natural fiber polymer composites*

### ABSTRAK

*Kesan ammonia polifosfat (APP) sebagai rencatan nyalaan dan kenaf sebagai pengisi ke atas sifat-sifat kebolehpayaan nyalaan, terma dan mekanik bagi komposit polipropilena (PP) telah dikaji. Spesimen ujian telah disediakan dengan menggunakan alat penyemprit skru berkembar untuk penyebatan dan diikuti dengan alat acuan suntikan. Kerencatan nyalaan bagi komposit juga telah ditentukan dengan menggunakan alat ujian penghad indeks oksigen (LOI). Penambahan perencat kebakaran ke dalam komposit kenaf-PP meningkatkan nilai LOI yang menunjukkan kemajuan dalam keupayaan rencatan nyalaan. Analisis termogravimetri dilakukan untuk memeriksa kestabilan terma bagi komposit. Penambahan serat kenaf ke dalam komposit PP menurunkan kestabilan terma secara ketara tetapi sebaliknya, kesan APP ke atas sifat-sifat terma bagi kenaf berpengisi komposit PP tidak mempunyai perubahan yang ketara. Kekuatan dan modulus lenturan bagi komposit meningkat dengan penambahan APP ke dalam kenaf yang berpengisi komposit PP. Penambahan APP ke dalam kenaf berpengisi PP juga menyebabkan peningkatan dalam kekuatan hentaman manakala peningkatan kandungan APP di dalam kenaf berpengisi komposit PP menunjukkan penurunan di dalam kekuatan hentaman.*

*Kata kunci: Rencatan nyalaan; serat semula jadi komposit polimer; sifat-sifat mekanikal*

### INTRODUCTION

Recently, the acceptance of natural fiber as fillers in the plastic industry has been growing extensively because of their high specific properties, biodegradability, low cost and environment friendly. The increasing utilization of natural fiber composites in various applications such as automotive components and aerospace industry is due to economic advantage as compared with other conventional filler or fiber. Kenaf also known as *Hibiscus cannabinus* is a fast growing annual growth plant cultivated for its fiber has been investigated as filler in thermoplastic composites (Rowell et al. 1999).

Represents as a large fraction of world polymer usage, polypropylene (PP) has been widely used in thermoplastic industry due to its low cost and ease processing. Higher stiffness at lower density and resistance to a higher

temperature when not subjected to mechanical stress are the key properties for PP. Previous study has reported that the addition of fiber reinforcement can enhance the modulus and strength of PP (Sameni et al. 2003). The use of reinforced polypropylene in electrical and automotive engineering has increased due to its excellent high modulus which enables it to replace conventional materials in demanding engineering applications. There are drawbacks in PP, when natural fiber was added in PP due to poor compatibility between fiber and PP chances of agglomeration while processing (Nabi Saheb & Jog 1999). Compatibilizing agent required to be added into composite to increase the interfacial bond between PP and natural fiber. It is due to PP which consists of hydrophobic matrix has less tendency to form interfacial bond with hydrophilic properties of kenaf (Srebrenkoska et al. 2009). The

addition of compatibilizing agent can accommodate polar interaction such as acid-based interaction and becomes a bridge to the hydroxyl groups on a natural fiber.

Nowadays, the demand of flame retardant materials is increasing. PP burns rapidly with a relatively smoke-free flame and without leaving a char residue due to its wholly aliphatic hydrocarbon structure (Zhang & Horrocks 2003). High flammability of PP come from the high self-ignition temperature at 357°C and a rapid decomposition rate compared with wood and other cellulosic materials. Other than that, the main issue of using natural fiber as filler in composite is the high sensitivity to a flame (Abu Bakar et al. 2010). An addition of organic filler such as kenaf increases the flammability properties of the PP composite (Sain et al. 2004). Flame retardant must be added into natural fiber filled PP composite in order to lower the flammability properties. The incorporation of flame retardant can disrupt the burning process at any stages which can lead to the termination of the process before actual ignition occurs. This will result in improvement of the flame retardancy of composite materials to fulfill the safety requirements required in many applications.

Over the last decade, the most commonly used additive types are inorganic compounds, halogenated compounds and phosphorus compound. Halogenated compounds as flame retardant are said to function primarily by a vapour phase flame inhibiting mechanism through radical reaction. As a result, combustion products which can cause corrosiveness and also contain high toxicity were released (Lu & Hamerton 2002). Therefore, the demand to halogenated flame retardant has decreased due to its harmfulness to the environment. The phosphorus compounds which disrupt the flame by increasing the conversion of polymeric materials to a char residue during pyrolysis were used. The phosphorus compounds are very effective in cellulosic materials that form char easily (Zhang & Horrocks 2003). Ammonium polyphosphate (APP) which is one of phosphorus based flame retardants is very commonly used flame retardant in PP. The mechanism of intumescent flame retardant is by the formation of a foam multi-cellular char on the polymer surface that hinders the polymer from burning. The char foam act as an effective barrier against heat and oxygen which slows down the diffusion of gaseous pyrolysis products to the combustion zone.

The objective of this study was to determine the effects of ammonium polyphosphate (APP) as flame

retardant and kenaf as fillers on flammability, thermal and mechanical properties of polypropylene composites. This paper reports on the flammability, thermal and flexural properties of the composites. From this study, it anticipated that thermoplastic composite with good mechanical and flame retardant properties can be used in different applications such as automotive, aerospace and building industries.

#### EXPERIMENTAL DETAILS

Heterophasic PP copolymer (SM-240) supplied by Titan Chemical, Malaysia was used as the polymer matrix. Kenaf fiber was supplied by the Malaysian Agricultural Research and Development Institute (MARDI), Malaysia. Ammonium polyphosphate (APP 750) supplied by Clariant was used as flame retardant. The maleated PP (MAPP) used as compatibilizer to improve the interaction between the filler and polymer, Orevac CA 100 with 1 wt % maleic anhydride was produced by Atofina, France.

Samples of different PP/Kenaf/APP ratios were prepared according to Table 1. Sample containing pure PP acts as the control. Kenaf fibers were ground to powdered form of size <500 µm. The fibers was then oven-dried at 50°C for 24 h before processing to co-rotating twin screw extruder (Brabender PL 2000 Plastic Coder) with a L/D ratio of 36 was used in the compounding process. The extrusion was conducted at a speed of 50 rpm and the temperature profile adopted during compounding were 180°C at the feed section and increased to 200°C at the die head. The extruded strands were then air-dried and pelletized. After being pelletized, the samples were injection-moulded into standard ASTM test specimens using an injection molding machine (HAI TIAN, HTF58X).

#### TESTING AND CHARACTERIZATION

The limiting oxygen index (LOI) test was done as per ASTM D 2863. In the test, the sample was held vertically in the transparent chimney, where the flow of oxygen and nitrogen were controlled. The test was repeated under various concentrations of oxygen and nitrogen to determine the minimum concentration of oxygen needed for burning the sample in 3 min.

Flexural test of PP and PP composites was carried out by using Lloyd universal tester according to ASTM D 790 with a cross head speed of 3 mm/min at room temperature. For impact strength, notched Izod impact testing was done

TABLE 1. Sample formulations of PP and PP composites

| Sample code | PP (%) | MAPP (%) | Kenaf fillers (PHR) | APP (PHR) |
|-------------|--------|----------|---------------------|-----------|
| PP          | 100    | -        | -                   | -         |
| PP-KA1      | 94     | 6        | 20                  | 20        |
| PP-KA2      | 94     | 6        | 20                  | 30        |
| PP-A1       | 94     | 6        | -                   | 20        |
| PP-K        | 100    | -        | 20                  | -         |

using an Izod impact tester LS-22005 according to ASTM D 256. Five samples were tested for each batch and the average results was taken and reported.

A Perkin Elmer thermogravimetry analyzer (TGA 7) was used to analyze the thermal and degradation stability of PP and PP based composite samples. Sample weight of around 8 mg was heated from 30 to 900°C at a heating rate of 20°C/min under nitrogen atmosphere. Thermal stability of the composites was determined from the temperature at which 10% weight loss occurred ( $T_{10}$ ).

## RESULTS AND DISCUSSION

The limiting oxygen index (LOI) test measures the minimum oxygen concentration required to support combustion. The LOI of PP and PP composites are given in Figure 1. As expected, the addition of APP into PP and kenaf filled PP composites increased the LOI values significantly,

an indication of increased flame resistance. The LOI values increased from 19 to 26% with incorporation of 30 phr of APP (composition PP-KA2). The incorporation of 20 phr of kenaf fiber did not significantly affect the flame retardant properties of PP. This is unexpected because it has been previously reported that composites filled with natural fiber is more prone to flame (Sain et al. 2004). The addition of MAPP also did not significantly affect the flame retardant properties of PP.

For the kenaf filled PP composites, the addition of 20 phr APP increased the LOI values by 4%. A further addition of APP from 20 to 30 phr marginally increased the flame resistance. This revealed that higher oxygen content was required to initiate and sustain combustion of the samples after the addition of flame retardant into PP composites. Therefore, the overall results obtained from the LOI test indicated that APP is effective as flame retardant PP and kenaf-filled PP composites.

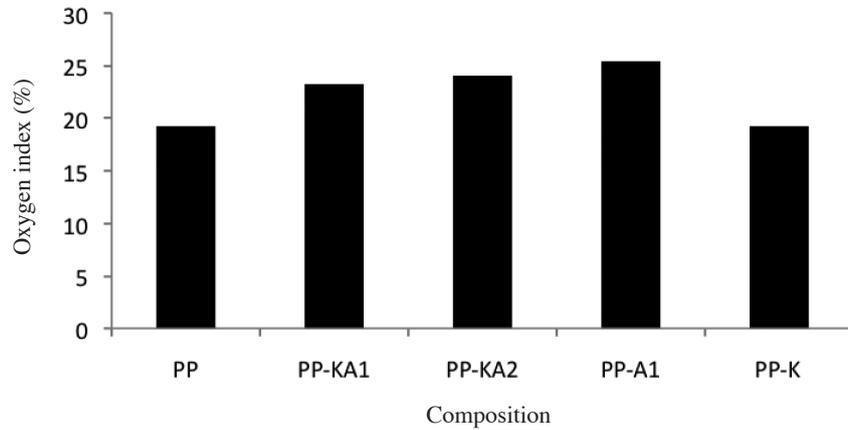


FIGURE 1. Limiting oxygen index of PP and PP different composites

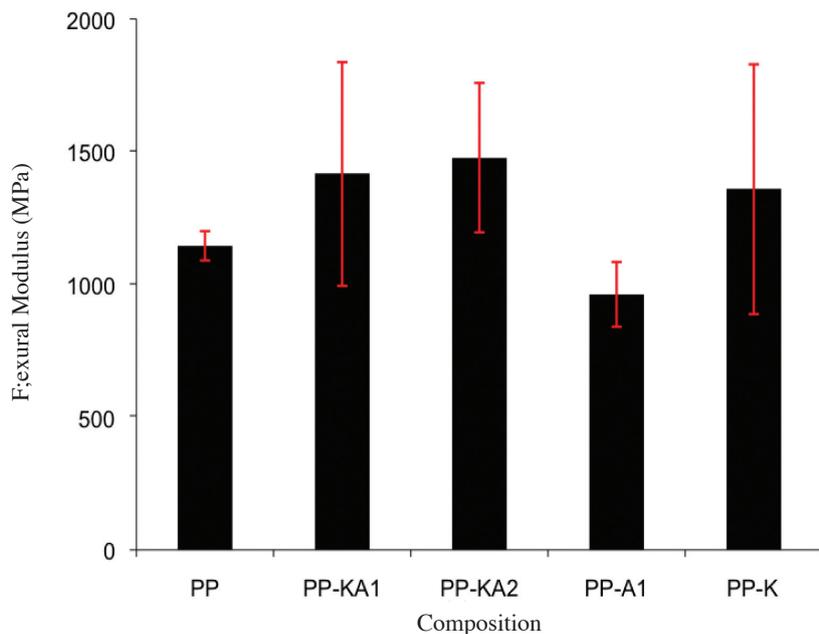


FIGURE 2. Flexural modulus of PP and different PP composites

Figure 2 shows that in general, the addition of APP and kenaf increased the flexural modulus of the PP composites. The flexural modulus of PP increased significantly with incorporation of 20 phr kenaf into PP. According to Abu Bakar et al. (2010), the natural fiber in composites acted as rigid fiber reinforcements, therefore increased the modulus of PP composites. The flexural modulus of the composites slightly increased when APP content increased from 20 to 30 phr. The improvement in flexural modulus of composites is due to the improvement of APP particles dispersion in the composite (Abu Bakar et al. 2010). PP-KA2 composite showed the highest flexural modulus among the PP composite samples.

As illustrated in Figure 3, the flexural strength decreased significantly with the incorporation of kenaf fillers in PP matrix and the addition of APP in kenaf filled PP composites. A greater decrement of flexural strength (36%) was observed when 20 phr kenaf was incorporated into PP compared with the incorporation of 30 phr APP. As

polar material, kenaf fiber has a tendency to agglomerates among them rather than to interact with nonpolar PP matrix. Besides that, the poor compatibility between APP and PP matrix also resulted in the decline of flexural strength of composites. Previous studies have reported that deterioration of mechanical properties of composites with addition of flame retardant (Abu Bakar et al. 2010; Sain et al. 2004), however, increase of APP content in the kenaf-filled composite from 20 to 30 phr resulted in an increase of flexural strength.

Figure 4 shows that the impact strength increased with the incorporation of kenaf fibers into PP matrix and the addition of 20 phr APP in kenaf filled composites. The increase in the content of APP from 20 to 30 phr in kenaf filled composite resulted in a decrease of impact strength. Study done by Zhang et al. (2012) also showed that the impact strength of composites decreased with increasing filler loading. Typically, composite with high filler content has less ability to absorb impact energy because of

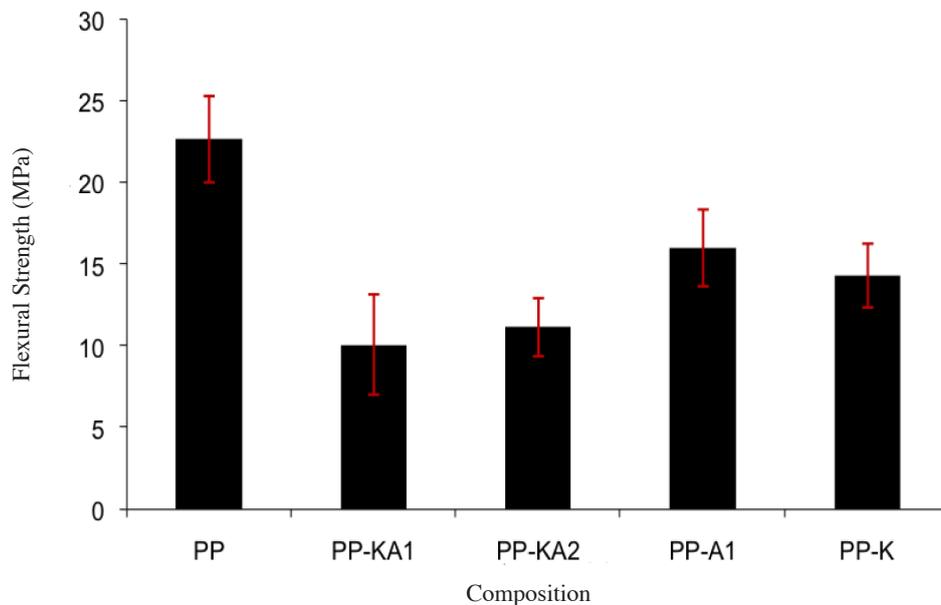


FIGURE 3. Flexural strength of PP and different PP composites

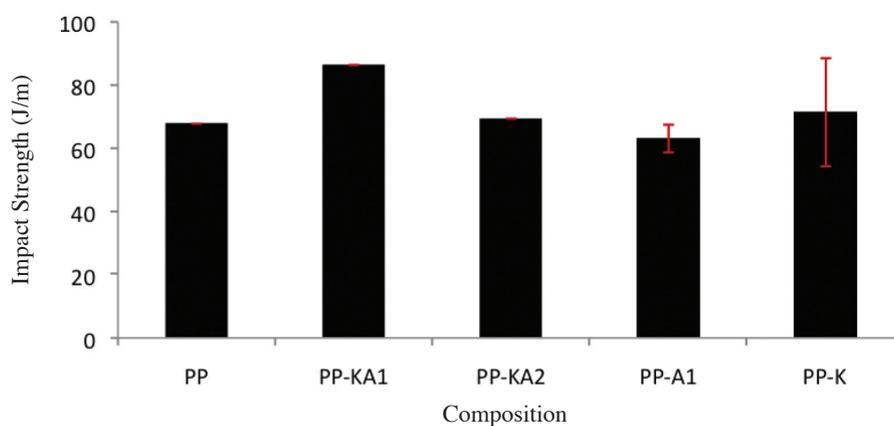


FIGURE 4. Impact strength of PP and different PP composites

interference from fillers on matrix continuity thus acted as micro crack initiator.

Thermal behaviors of PP and kenaf filled composites were examined by using TGA and thermal curve as shown in Figure 5. In this study, the initial thermal decomposition temperature ( $T_d$ ) is defined as 10% weight loss occurs to observe the differences. The figure shows a single mass-loss step of thermal decomposition. The presence of APP, kenaf and MAPP decreased  $T_d$  of the composites. The reduction of thermal stability is observed to be greater in PP containing APP composites as compared with PP. Increasing APP content in the kenaf filled PP composites did not influence the thermal stability of PP. Addition of APP into PP promoted char formation of the composites. APP degrades to form phosphorus acid derivatives that can be used in the phosphorylation of cellulose, especially the reactive C-6 primary hydroxyl. The formation of a phosphorus ester at C-6 inhibits depolymerization and promotes char formation by blocking this site. In addition, phosphorus acids formed by the thermal degradation of a phosphorus salt or organophosphorus compound catalyze the dehydration and further promote char formation and reduce fuel-gas generation. It is expected that the amount of residue is dependent on the amount of APP in the PP composites. Based on Table 2, the remaining residue is almost the same with APP content added. For PP-KA2, the

amount of residue is more than APP content because some of the kenaf is promoted as char residue. The char residues from kenaf come from lignin which consist 20-30 wt% of kenaf (Abu Bakar et al. 2010).

## CONCLUSION

The effects of kenaf and APP on flammability, mechanical and thermal properties of PP composites have been investigated. APP has shown to be effective as flame retardant and has increased the flammability resistance of PP and kenaf-filled PP composites. The incorporation of kenaf fiber into PP did not reduce the LOI index significantly. The addition of APP and kenaf increased the flexural modulus of the PP composites. The flexural modulus increased significantly with incorporation of 20 phr of kenaf into PP and APP filled PP composite. Sample with 20 phr kenaf and 30 phr APP showed the highest flexural modulus among the PP composites. Incorporation of kenaf into PP and APP filled PP composites decreased the flexural strength significantly but increased the impact strength of the composites. A decrease in impact strength was observed with increasing APP content in PP and kenaf filled PP. The TGA has shown that kenaf has reduced the thermal stability of PP and APP filled PP composites. The increase of APP in the kenaf-filled PP composites did not influence the thermal

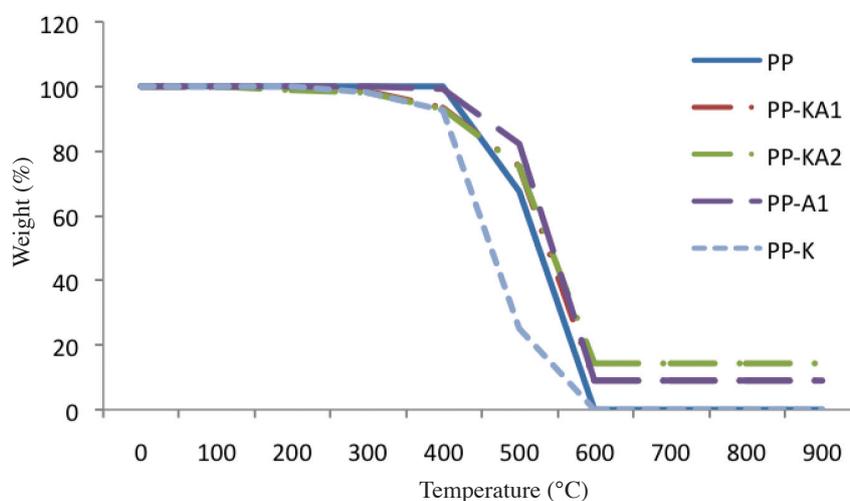


FIGURE 5. TGA of PP and PP composites at heating rate 10°C/min

TABLE 2. Thermal behaviors of PP composites

| Composition | * $T_d$ (°C) | *Residue (%) |
|-------------|--------------|--------------|
| PP          | 476.2        | 0            |
| PP-KA1      | 440.9        | 14.8         |
| PP-KA2      | 441.7        | 36.4         |
| PP-A1       | 483.3        | 29.2         |
| PP-K        | 460.0        | 0            |

\* $T_d$  at 10% weight loss

stability of PP composites significantly. Overall, the study has shown that kenaf-filled PP composites containing APP have better flammability resistance, flexural stiffness and strength and toughness as compared with PP, but lower thermal stability.

#### ACKNOWLEDGEMENTS

The financial supports from the Fundamental Research Grant Scheme (FRGS Vot: 78354, Sub Code: 3F302), Ministry of Higher Education (MOHE) and UTM are acknowledged.

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Received: 3 June 2011

Accepted: 20 January 2012