MECHANICAL AND FLAMMABILITY PROPERTIES OF HYBRID RECYCLED NEWSPAPER / MONTMORILLONITE POLYPROPYLENE COMPOSITES

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To my beloved Mother, Father, brothers, sister and the ones who give me inspiration and support that made this work possible

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ABSTRACT

The objectives of this study are to study the effect of recycled newspaper (RNP) and ammonium polyphosphate (APP) content on mechanical properties and flammability properties of polypropylene (PP) and to determine the effect of APP content on mechanical and flammability properties of PP. In this research, RNP was gathered by collecting unused newspaper and was shredded it into small pieces. Then, it was soaked with NaOH solution (10 wt %) for 24 hours at room temperature. After that, it was rinsed with distilled water for several times. Montmorilloinite (MMT) was used as the other filler in PP. PP composites containing RNP were prepared by melt compounding using twin screw extruder followed by compression molding. The mechanical properties of PP composites were studied through tensile, flexural and impact tests. Limiting oxygen index test was used to investigate flammability properties of PP composites. Tensile strength reduced when RNP content increased while in APP content scope, it remains constant. Tensile modulus increased when RNP content increased and it remains almost constant when in APP content. Elongation at break reduced when RNP content increased while in APP content, it has no significant effects on it. Flexural strength increased when RNP content increased. Flexural strength reduced when APP content increased. Flexural modulus increased when RNP content increased and it remains constant when APP content increased. Impact strength increased when RNP content increased and it reduced when APP content increased. LOI value increased when RNP and APP content increased. From the first batch result, 20 wt% of RNP content formulation was used to produce second batch based on tensile strength and LOI value.

ABSTRAK

Objektif kajian ini adalah untuk mengkaji kesan kandungan akhbar kitar semula (RNP) dan ammonium polyphosphate (APP) pada sifat-sifat mekanik dan sifat kemudahbakaran polipropilena (PP) dan untuk menentukan kesan kandungan APP pada sifat-sifat mekanikal dan kemudahbakaran PP. Dalam kajian ini, RNP diperoleh melalui pengumpulan akhbar lama dan telah dicincang kepada kepingan kecil. Kemudian, ia telah direndam di dalam larutan NaOH (10% kecairan) selama 24 jam pada suhu bilik. Selepas itu, ia telah dibilas dengan air suling untuk beberapa kali. Montmorilloinite (MMT) juga telah digunakan sebagai pengisi di dalam PP. PP komposit mengandungi RNP telah disediakan melalui pengadunan leburan menggunakan alat penyemperit berkembar diikuti dengan pengacuan mampatan. Sifat-sifat komposit PP seperti mekanikal dan sifat kemudahbakaran telah dikaji Sifat-sifat mekanik komposit PP telah dikaji melalui ujian tegangan, lenturan dan hentaman. Ujian pengehad indeks oksigen telah digunakan untuk mengkaji sifat kemudahbakaran komposit PP. Kekuatan tegangan menurun apabila kandungan RNP meningkat. Manakala di dalam bidang APP, kekuatan tersebut tidak berubah. Modulus tegangan meningkat apabila kandugan Pemanjangan putus berkurangan apabila kandungan RNP meningkat dan di dalam bidang APP, pemanjangan tersebut tidak berubah. Kekuatan lenturan meningkat apabila kandungan RNP meningkat dan kekuatan tersebut berkurang apabila kandungan APP meningkat. Modulus lenturan meningkat apabila kandungan RNP meningkat dan di dalam bidang APP, modulus tersebut tidak berubah. Kekuatan hentaman meningkat apabila kandungan RNP meningkat dan kekuatan tersebut berkurang apabila kandungan APP meningkat. Nilai LOI meningkat apabila kandungan RNP dan APP meningkat. Dari hasil kumpulan pertama, formulasi 20 wt% kandungan RNP telah digunakan untuk menghasilkan kumpulan kedua.

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TABLE OF ABBREVIATION / SYMBOLS

ρ	Density	kg/m^3
μ	Fluid viscosity	kg/m.s
ρ_1	Liquid density	kg/m^3
ΔH	Enthalpy changes, duty	kJ/h
ΔH_l	Enthalpy for liquid	kJ/h
ΔH^o_f	Standard heat of formation	kJ/mol
ΔH^o_{vap}	Standard heat of vaporization	kJ/mol
ΔH_v	Enthalpy for vapor	kJ/h
ΔH_{vap}	Heat of vaporization	kJ/mol
ΔP	Pressure differential or pressure drop	N/m^2
ΔT_{LM}	Log mean temperature differences	^oC
A	Cross sectional area	m^2
A	Heat transfer area	m^2
A_c	Cross-sectional area	m^2
A_d	Down-corner cross sectional area	m^2
A_t	Heat transfer area	m^2
C_{BM}	Bare module cost	US $
C_d	Experimentally determined drag coefficient	Dimensionless
C_o	Orifice coefficient	Dimensionless
C_p	Vapor heat capacity	J/g mol K
d	Diameter	m
D	Vessel diameter	m
D_c	Column diameter	m
D_i	Inside diameter	m
D_m	Mean diameter	m

D_o	Outside diameter	m
η	Efficiency	Dimensionless
F	Molar flow rate	lbmol/h
F^a_{BM}	Bare module factor	Dimensionless
F_M	Materials factor	Dimensionless
g	Gravity acceleration	m/s^2
H	Enthalpy	kJ/kmol
H	Height of the column	m
MW	Molecular weight	Dimensionless
P	Pressure	kPa, atm
Q	Duty or heat transferred per unit time	kW
Q	Volumetric flow rate	m^3/s
R	Universal gas constant	Dimensionless
SG	Specific gravity	Dimensionless
T	Temperature	K, ^oC
U	Overall heat transfer coefficient	$W/m^{2o}C$
u_f	Vapor velocity at flooding point	m/s
V	Volume	m^3
V	Volumetric flow rate	m^3/h
W_v	Total vapor flow rate	kg/h
X	Conversion of component	Dimensionless

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CHAPTER 1

INTRODUCTION

1.1 Background of Study

The usage of synthetic fiber has been widely implemented in polymer industry since few years ago. Among the reasons of using synthetic fiber are high strength and high specific modulus of reinforcing fiber. Synthetic fiber also must have high or excellent in one of these properties: tensile strength, operating temperature, heat resistance, and flammability properties (Thomas *et al*., 2012). Although this criteria is among the key points of selecting filler, there is another issue which is environmental issue where it must be considered. The usage of natural filler such as recycled newspaper (RNP) helped to reduce this problem where it concerns the society about the environment. The usage of common polyolefin such as polypropylene (PP) brought advantage when RNP and PP was fused together to form PP composites.

The usage of using RNP as reinforcing filler in composites was found in 20th century where Sanadi *et al*., 1994, has used RNP to reinforce PP composites. In this research, it stated that there are several points needed to be considered in order to optimize the composites properties: fiber aspect ratio, uniformity of dispersion of the fibers in the matrix, and the ability of the interface to transfer stress to the fiber. The usage of chemical compound coupling agents helped the composites to achieve better

strength. Further investigation needed to be done such as the use of lubricants, processing equipment, and usage of impact modifiers could produce composites with better properties. Due to the low strength and high water absorption properties of RNP, the addition of montmorillonite (MMT) will increase the strength of the composites. On the other hand, the amount of MMT needed is small but the output strength is high. The hybrid filler will be produced and thus, composites with better properties can be produced.

Flame retardant application has been widely increased in these days. As reported by Zhang *et al*., 2003, the first flame retardant PP was developed in 1954 where Blatz used flame retardant by the incorporation of thermoplastic melt with halogen compound (1,2-dibromo-4-(a,b-di-bromoethylcyclohexane) and metal oxide (Sb_4O_6), a free radical initiator. On the other hand, the presence of ammonium polyphosphate (APP) in polypropylene has increased its oxygen index from 18.5 to maximum value of 21.5 when the weight percentage of APP reached 25% (Granzow *et al*., 1980)

Composites are made from the mixing of polymer matrix and filler. Filler can be ranged from synthetic filler to natural filler. RNP is under the category of natural filler. It contains lignocellulosic material and other inorganic fillers, such as printing inks and process aids. It also can be used as a substitute for inorganic fillers such as clay, talc, calcium carbonate, and silica (Ismail *et al*., 2008). Hybrid filler has also has been used in PP composites to increase the strength of PP. This has been discussed in the paper published by Panthapulakkal *et al*., 2006. In this paper, the usage of hybrid filler between glass fiber and hemp fiber were used to increase the mechanical properties of PP matrix. The researchers found that incorporation of glass fibers has improved tensile, flexural and impact properties of the original short hemp fiber composites. The strength increased when glass fiber content increased. On the other hand, the water absorption properties of short hemp fiber composites reduced when glass fiber and hemp fiber worked together as hybrid filler.

By using hybrid filler, the improvement of composites was truly remarkable. The usage of RNP and other kind of filler such as carbon black (CB) and silica increased the tensile strength and elongation at break of the composites increased. On the other hand, the Young's modulus decreased with increasing CB and silica in the weight ratio of RNP / CB and RNP / silica to PP composites (Ismail *et al*., 2008). In this paper, Increasing of tensile strength can be seen for both hybrid-filler filled PP/NR composites, with the RNP/CB hybrid showing higher tensile strength than the RNP/silica hybrid for all weathering conditions. The author also suggested that the replacement of RNP with CB is sufficient to achieve improvement of weatherability for hybrid composites.

1.2 Problem Statements

The issue of using synthetic filler such as glass fiber, aramid, and silica has brought concerns in the society. The usage of these fillers can cause damage to the environment. As an alternative, the usage of natural filler such as recycled newspaper (RNP) filler can reduce the possibility of damaging the environment. However, only little research is available and published related in studying the effects of RNP content in polymer and rubber matrix.

A part from this study, this research is investigating the effects of RNP content in polypropylene (PP) composites. RNP is generally considered as waste for example where old and unused newspaper was sent to the landfill. By using RNP as filler, it can reduce environmental problem by reducing numbers of newspapers to the landfill. From the data gathered from Alam Flora Sdn. Bhd., one tonnes of paper can saves:

- 17 trees
- 2 barrels of oil
- 4,100 kilowatts of energy

Another statement made by Ream Paper Marketing Sdn. Bhd., 2007, approximately over 57,000 tonnes of waste paper per month, which occupies 456,000 cubic meters of landfill space, was thrown into landfills in Malaysia. The area in landfill is almost similar to an average large warehouse. In addition, it is also similar to chopping down 680,000 of trees. This problem can cause diseases such as dengue due to stagnant pools exhibit in the landfill. On the other hand, waste to wealth concept can be implemented due to the cheap source of raw material.

Research has being done on fibers and RNP hybrid filler reinforced in polymer. Recent work published by Shakeri *et al*., 2009, the main purpose of this research is to investigate the behavior of thickness swelling and water absorption properties of glass fiber / RNP PP composites. In their research, it stated that when RNP filler content increase, the thickness swelling and water absorption properties increased slightly. However, these properties reduce when glass fiber content increased. The presence of coupling agent and glass fiber has greatly improved the properties of PP composites by reducing thickness swelling and water absorption properties.

There is no current research that is related to this topic under scope of RNP / MMT hybrid filler. The investigation of this research is new because commonly only one type of filler either MMT or RNP was used as filler to produce the composites. On the other hand, the investigation of flammability properties of polymer is also not being done under scope of RNP.

1.3 Objectives

The overall objective of this study is to develop a new type of formulation based on MMT/RNP filled PP composites. In this research, PP composites were prepared by blending PP, RNP and MAPP in the initial stage. In the next stage, MMT and APP was added together with materials in the initial stage to produce PP composites.

The overall objective can be categorized into:

- To study the effect of RNP content on mechanical properties and flammability properties of PP

- To determine the effect of APP content on mechanical and flammability properties of PP

1.4 Significance of Study

From the research, the usage of RNP as filler in PP composites will bring advantage because of the high availability of RNP in the world. The cost effective composites will be developed through this research and it has high marketability potential.

1.5 Scope of Study

In order to achieve the objectives of the research, the activities below were carried out:

1. Sample Preparation
    a. RNP was bleached with NaOH solution to purify it.
    b. Twin screw extruder to blend PP, compatibilizer, RNP, MMT and APP.
    c. Compression molding to prepare specimen according to specific dimension

2. Mechanical Properties Study
    a. Tensile Test
    b. Flexural Test
    c. Izod Impact Test

3. Flammability Properties Study
    a. Limiting Oxygen Index Test

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