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Original Research

Recycled Newspaper Fiber/Polypropylene Composites with Inorganic Flame Retardants: Flame Retardant and Mechanical Characteristics

Haydar U. Zaman[1,2,*], **Ruhul A. Khan**[2]

[1] *Department of Physics, National University of Bangladesh, Gazipur, Dhaka, Bangladesh*

[2] *Institute of Radiation and Polymer Technology, Bangladesh Atomic Energy Commission, P.O. Box-3787, Savar, Dhaka, Bangladesh*

[*] *Correspondence*: haydarzaman07@gmail.com (H.U.Z.);

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Abstract: This study intends to look at ways to increase the flame retardancy of composite materials made of recycled newspaper fiber (RNF) and polypropylene (PP) matrix. A melt-blending approach followed by injection molding was used to create PP/RNF composites, which have various advantages including low cost, the ability to dispose of industrial waste, and good mechanical properties. Different types and concentration of flame retardants, such as magnesium hydroxide ($Mg(OH)_2$) with smaller particle size and zinc borate with antimony trioxide (ZB-AT), were utilized to increase the flame resistance of the composites. The UL94 test was used to assess the composites' flame resistance. In comparison to pure PP, composite blends of PP/RNF/$Mg(OH)_2$ and PP/RNF/ZB-AT are more fire resistant. Depending on the application, the resulting composite may have different amounts of both elements. Although effects on tensile and impact strength have not been discussed, research has been done and publications have been made regarding improvements in the fire retardant properties of such composites. The strength of the PP/RNF/$Mg(OH)_2$ and PP/RNF/ZB-AT composites were discussed in relation to the effects of $Mg(OH)_2$ and ZB-AT concentration. Study and discussion were conducted about the tensile, flexural, and impact properties. With the addition of flame-retardants, a little drop in the mechanical characteristics of the composites was observed.

Keywords: Flame retardancy; Flammability; Mechanical properties; Polypropylene; Recycled newspaper fiber/PP composite.

Introduction

Natural fibers are now being used as reinforcing materials in polymer composites rather than glass fiber as a result of growing environmental concern. Numerous studies have been conducted on the utilization of lignocellulosic fibers, such as wood and cellulose, in polymer composites [1]. Recycling fiber-based items including paper, waste wood, and agricultural leftovers can provide a significant source of lignocellulosic fibers or a comparable feedstock. Newspaper, which contains lignocellulosic material and other inorganic fillers like printing inks and process assistance chemicals, is a good recycling fiber-based product [2]. Both can be utilized in place of inorganic fillers like clay, talc, calcium carbonate, silica, and others when creating polymer composites [3]. Contaminants (inks, coatings, etc.), which have severe negative effects on paper made from wastepaper and recycled, however, have little impact on composites [4]. Natural fiber-reinforced (NFR) composites have a number of benefits over synthetic materials, including low weight, acceptable strength and stiffness, biodegradability, renewable resources, affordability, and environmental friendliness [5-9]. Additionally, when compared to glass fiber reinforced, the NFR lessens tool wear as well as cutaneous and respiratory discomfort during handling [10, 11]. Due to all these advantages, NFR is highly appealing to businesses that produce building materials including siding, decking, fencing, and non-decorative trim. Infrastructure, such as boardwalks, marinas, and guardrails; transportation, such as interior auto panels, truck flooring, and headliners; and industrial and consumer applications, including pallets, playground equipment, and seats, are some more applications [12]. They can also generate a lot of interest when used as reinforcing fillers for thermoplastics, particularly those with a comparatively lower melting point like polypropylene (PP) and high and low density polyethylene (PE) [13].

One of the ubiquitous polymers, PP has a variety of benefits, including ease of processing, resistance to corrosion, mechanical rigidity, low density, and low affordability [14-16]. These encourage the use of PP in a wide range of technological and practical contexts. PP resins are given better mechanical qualities and are less expensive when a cellulosic ingredient is added [17]. One of the cellulosic resources is recycled newspaper fiber (RNF), which is a byproduct of the paper industry. Inorganic materials and paper pulps make up the majority of its composition. PP/RNF composites are quite important, especially in terms of waste recycling. It appears that PP/RNF composites can be used to make construction items like furniture for homes and businesses. However, there is room for improvement with the PP/RNF composites. Their flammability is a disadvantage in such applications. In light of this, it is crucial to increase flame retardancy in PP/RNF composites.

The definition of flammability is the ease with which anything will burn, ignite, or cause combustion. In order to meet the safety criteria for composite goods, it has become more vital to increase the flame retardant characteristic of composite materials because polymers and fibers are particularly susceptible to flame as organic materials [18]. There are five key processes in the burning process: heating, decomposition, ignition, combustion, and propagation [19, 20]. The burning process can be interrupted at any of these points, which can result in the process ending before actual ignition, to produce flame retardancy. The fastest way to achieve flame retardancy is to incorporate flame-retardants that can hinder combustion during a certain stage of the process such that the finished system exhibits acceptable flame retardancy [19]. Inorganic substances, halogenated substances, and phosphorous substances are the most often employed additive types of flame-retardants [21]. As additional flame retardant chemicals, boric acid, ammonium phosphates and borates, ammonium sulphate and chlorides, zinc chloride and borate, antimony oxide, sodium borate, and dicyanodiamide have all been employed [10-13, 22-24]. To obtain the desired fire resistance, fire retardants are chemically or physically integrated into the composites. There is growing interest in inorganic substances such metallic hydroxide additions in the search for fire retardants that are safe for the environment and human health while maintaining improved fire performance. The fire retardants employed in this work are magnesium hydroxide, and zinc borate with antimony trioxide co-additive. In the polymer composite, various amounts of fire retardants are added in various ratios. The most used metal hydroxide flame retardant ingredient for polymers is magnesium hydroxide [$Mg(OH)_2$]. $Mg(OH)_2$ has the benefit that processing it into plastics like polypropylene (PP) is possible since its decomposition into magnesium oxide, MgO, and water begins at a somewhat higher temperature (300-320^oC) [25].

$$\mathrm{Mg(OH)_2 \rightarrow MgO + H_2O} \qquad \Delta H = 328\ \mathrm{cal/g}$$

According to modern fire standards, zinc borate is a highly efficient inorganic flame retardant with the distinctive qualities of suppressing smoke and encouraging charring. In order to increase the FR qualities of other halogenated or halogen free flame retardant solutions, zinc borate is frequently utilized as a multifunctional flame retardant. The use of a polymer and the type of halogen source (aliphatic versus aromatic) determine how effective it is. In terms of fire retardancy, zinc borate and antimony oxide typically show synergistic effects [26]. The characteristics and attributes of zinc borate are displayed in Table 1. Depending on its structure, antimony trioxide (AT) can be either colorless or white. AT was slightly dissolved in water and also dispersed in potassium hydroxide, diluted hydrochloric acid, and many organic acids [27]. Table 2 displays the attributes of AT [28]. Figure 1 depicts AT's chemical composition. The flammability of composites has received very little attention, despite the fact that the flammability of polymers has received a great deal of attention [29, 30]. The goal of this paper is to investigate how $Mg(OH)_2$ and ZB-AT affect the mechanical and flammability characteristics of PP/RNF composites.

Table 1: Characterization and attributes of zinc borate [27].

Mol wt.	pH	Density g/cm^3	Melting point (°C)	Appearance
434.62	7.6	3.64	980	White Crystalline

Table 2: Antimony trioxide characteristics [28].

Density (g/cm^3)	Boiling Point (°C)	Melting Point (°C)	Property
5.67	1425	656	Value

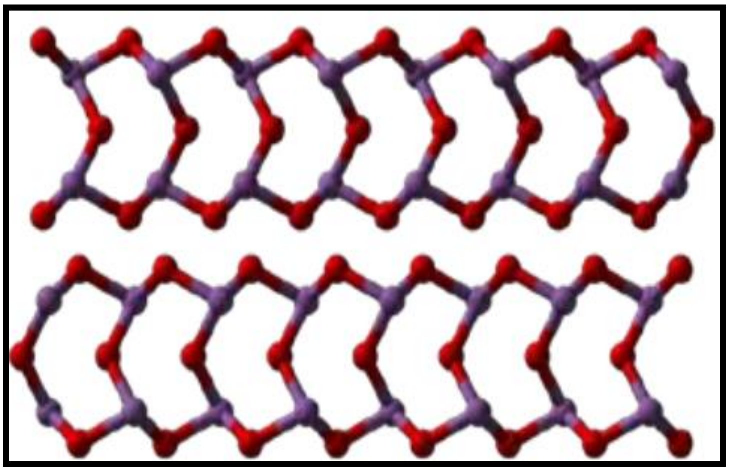

Figure 1: AT's chemical make-up.

Materials and Methods

Materials Utilization

In this experiment, the matrix was Polypropylene (R520Y) provided by SK Corporation, which has a melt flow index (MFI) of 1.8 g/10 min (ASTM D1238) and a density of 0.9 g/cm^3. This polymer with a low MFI was chosen because it could provide improved mechanical performance. The material for the reinforcing fibers was recycled newspaper. They were gathered locally and primarily consisted of The Ittefaq, a daily newspaper published in Bangladesh. The RNF, which has a particle size of 23 μm, a density of 1.8 g/cm^3, and approximately 97% organic components (cellulose, hemicellulose, and lignin), was employed. Table 3 provides specifics regarding the RNF's chemical components. The probable fire retardants employed in this phase were magnesium hydroxide and zinc borate with antimony trioxide (ZB-AT), which were provided by Fisher Scientific and US Borax Inc.

Table 3: X-ray Flourescence Spectrometer Rigaku RIX 3000 used for semi-quantitative RNF analysis.

Component	Wt%
Na_2O	0.45
MgO	0.093
Al_2O_3	0.87
SiO_2	0.91
SO_3	0.014
K2O	0.2
CaO	0.44
TiO_2	Trace
Fe_2O_3	Trace
LOI (Organic)	97.0

Fiber Preparation

The Ittefaq newspapers were acquired as sheets. The newspapers were submerged in a NaOH (10 weight percent) solution at room temperature for 24 hours before being repeatedly washed with distilled water. The newspapers were rinsed, dried in a vacuum oven at 80 °C for 24 hours to remove any remaining moisture, and then ground into a finer powder. The fiber was sieved into the desired size, or 23 μm, using an Endecott's sieve. The produced RNF must be kept in a desiccator at room temperature prior to mixing in order to prevent moisture absorption.

Procedures for Mixing

The composite is made up mostly of PP, RNF, and $Mg(OH)_2$/ZB-AT. To explore the impact of $Mg(OH)_2$/ZB-AT on the composite, different concentrations of $Mg(OH)_2$/ZB-AT were added to PP to create various compositions. First, the PP, RNF, and $Mg(OH)_2$/ZB-AT were combined in a single bag and thoroughly shaken to blend. A twin-screw co-rotating intermeshing extruder (Bau-Tech, Korea) was used to combine the mixture. Its screw diameter was 19 mm, and its L/D ratio was 40. The distance between the screw axes was 18.4 mm. As shown in Figure 2, it was equipped with a modular screw arrangement that included various combinations of right-handed and left-handed screws, neutral kneading disk elements, and one reserve-pumping screw element. The cylinder temperature was maintained at 150, 165, 175, and 180 °C from the hopper to the die while the screw speed was kept at 150 rpm. The melted compounds were allowed to cool to room temperature, and then palletized using a granulated grinder. The granulates were then injection molded into tensile, flexural, and impact test specimens using an Engel Injection Molder (Model ES-28) fitted with a typical ASTM mold at 204oC. For experimental testing, dog-bone-shaped specimens with the necessary dimensions are produced by the injection molding technique. Figure 3 explains the flow chart for the manufacturing process, and Table 4 displays the blend proportions of the four components.

Figure 2: Used screw configuration for PP/RNF blending tests.

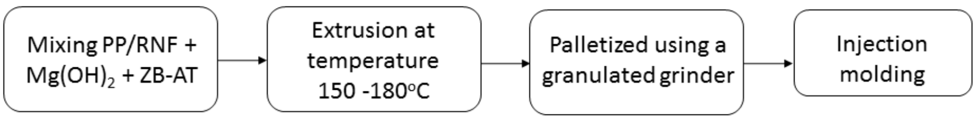

Figure 3: Flow chart for the production process.

Flammability Assessments

The flammability of the specimens was investigated using the ASTM D 635 horizontal burning test and the ASTM D 2863 limiting oxygen index test, respectively. The sample was held horizontally for the horizontal burning test, and one end of the sample was lit using a flame powered by natural gas. A measurement was made of the amount of time it took the flame to go from the first reference point, which was 25 mm from the end, to the second reference mark, which was 100 mm from the end. The sample was held vertically in the glass chamber for the limiting oxygen index test, which involves a regulated flow of oxygen and nitrogen. The sample was lit from the top, and the amount of time it took for 50 mm of the sample to burn was recorded. To find the minimal oxygen concentration required to burn the sample, the test was repeated under varied oxygen and nitrogen concentrations.

Table 4: The blend proportions of the four components.

Ingredients	Mix number			
	A	**B**	**C**	**D**
PP	100	50	50	50
RNF	-	50	25	25
$Mg(OH)_2$	-	-	25	20
ZB-AT	-	-	-	5

Mechanical Testing

A typical computerized testing apparatus (Sintech Model 20) with a cross head speed of 12.5 mm/min was used to conduct a tensile test on the PP/RNF/$Mg(OH)_2$ or PP/RNF/ZB-AT composite specimen. For tensile tests, the specimen geometry and experiment were set up in accordance with ASTM standard D638. Figure 4 depicts the dog-bone-shaped specimen for the tensile specimen.

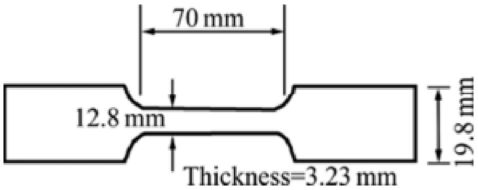

Figure 4: Tensile test sample made from a dog bone.

A standard computerized testing apparatus has been used to conduct the three-point bending test in accordance with the recommendations in ASTM standard D790 (Sintech Model 20). Figure 5 illustrates the dimensions and orientation of the rectangular bending specimen, where L is the span length between the two supports, d is the depth, and b is the thickness. The span length of 50.8 mm existed between the two supporting nodes.

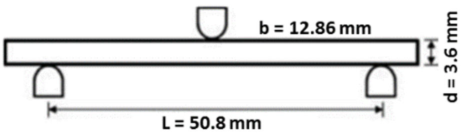

Figure 5: Test configuration for three-point bending.

Using a Pendulum type Beam Impact machine, impact tests were conducted in accordance with ASTM D 256. For each sample, at least six specimens were evaluated in order to determine an accurate average and standard deviations for every mechanical property. At room temperature, all mechanical characteristics were tested.

Results and Discussion

The Composites' Flammability

The horizontal burning test results for the PP and filled composites (Mixes A–D) are shown in Figure 6. The burning rate of a 50% filled RNF-PP composite is higher than that of a virgin polymer, demonstrating the composite's high flammability sensitivity. According to reports [31, 32], $Mg(OH)_2$ has the ability to operate as a flame retardant for plastics and is effective at larger loadings. When 25% $Mg(OH)_2$ is added to the composite (Mix C), the burning rate drops to even lower levels than the PP alone, or about half that of Composite B. (Mix A). This suggests that, at a lower loading, $Mg(OH)_2$ can be employed as a flame retardant for RNF composites. Compared to the composite including magnesium hydroxide (Mix C), the combination of ZB-AT and magnesium hydroxide in the ratio of 20:5 (Mix D) burns at a faster rate, although it burns at a lower rate than the composite B. This suggests that these flame retardants have a retarding effect rather than a synergistic effect, which may be explained by the identical flame retarding behavior of $Mg(OH)_2$ and ZB-AT. $Mg(OH)_2$ is well recognized for having a superior endothermic flame retarding response (approximately 1450 J/g) [33] compared to that of any boron compounds; as a result, replacing a portion of it with ZB-AT causes the $Mg(OH)_2$ impact to be lessened.

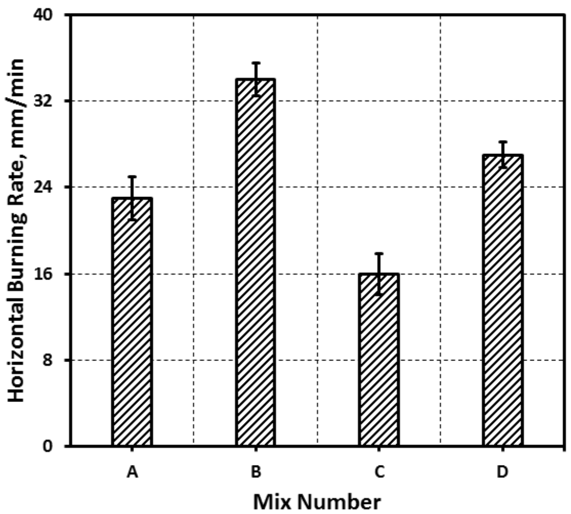

Figure 6: Horizontal burning rates for mixtures A through D.

The findings of the testing of the mixes A–D are depicted in Figure 7 and are based on oxygen index measurements, which determine the minimal oxygen concentration necessary to support combustion. The oxygen concentration required to ignite a sample with high flame retardancy is higher. The most oxygen is needed for composite C to burn, whereas composite B only needs a small amount of oxygen to burn and is most like virgin polymer. When $Mg(OH)_2$ is employed in conjunction with ZB-AT, oxygen index tests similarly demonstrate a reduction in the flame retarding impact rather than a synergetic effect, similar to the horizontal burning experiments.

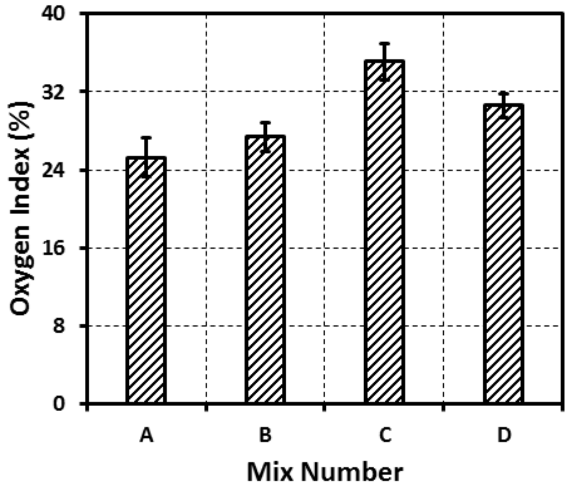

Figure 7: Oxygen index for mixtures A through D.

Study of Mechanical Features

The mechanical characteristics of the PP and RNF filled composites (Mixes A–D) are summarized in Table 5. Tensile and flexural properties of the composites are negatively impacted by the addition of flame retardants, with reductions of 16 and 14%, respectively, in comparison to the composite without flame retardant. This might be explained by the poor compatibility between the polymer with the additional flame retardants. In contrast to pure PP, these blends perform better. Additionally, the composite's tensile and flexural moduli both exhibit a slight decline in comparison to those of the composite lacking $Mg(OH)_2$. Some employees have reported that the use of flame-retardants causes the mechanical properties of filled and unfilled plastics to degrade [34, 35]. The combined effects of flame retardants exhibit roughly the same tensile and flexural values as composite C. The inclusion of flame-retardants does not significantly change the impact properties of the composites.

Table 5: Mechanical characteristics of PP and composites.

Properties	**Mix number**			
	A	**B**	**C**	**D**
Tensile Strength, MPa	28.3 ± 0.4	38.4 ± 0.5	32.4 ± 0.3	31.9 ± 0.2
Tensile Modulus, GPa	0.98 ± 0.2	2.8 ± 0.1	2.5 ± 0.2	2.3 ± 0.1
Flexural Strength, MPa	35.6 ± 0.4	47.3 ± 1.1	38.2 ± 0.4	37.6 ± 0.3
Flexural Modulus, GPa	1.1 ± 0.2	2.9 ± 0.3	2.5 ± 0.2	2.3 ± 0.1
Impact Strength, J/m				
Notched	690 ± 65	60 ± 3	39 ± 2	45 ± 3
Unnotched	895 ± 83	139 ± 18	122 ± 4	156 ± 5

Conclusion

The entire outcome of this work emphasizes the composition's effects and fresh knowledge to help composite materials enhance their fire retardancy performance while providing technology for yet another efficient method of environmental protection. A fabrication attempt of the PP/RNF composite was made, along with an investigation of the effects of utilizing $Mg(OH)_2$ and ZB-AT as fire retardant materials on the mechanical behavior of the composite, with a suggestion for their use. Magnesium hydroxide at a 25% concentration can successfully cut the flammability of the filled composite to 50% of the composite without flame retardant. The flame retardant feature of $Mg(OH)_2$ is shown to have a retarding impact when partially replaced with ZB-AT, as opposed to having a synergetic effect. The mechanical properties of the flame retardant filled composites exhibit somewhat worse characteristics compared to the composites without a flame retardant while exhibiting superior characteristics compared to virgin PP.

Disclosure Statement

The author(s) did not report any potential conflict of interest.

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