NATURAL RUBBER COMPOSITE MIXED WITH POLYETHELENE-ALUMINUM FROM USED BEVERAGE CARTONS WASTES

Walanrak Poomchalit[1], Watthanaphon Cheewawuttipong[2], Jirawat Jai-u[3], Supaaek Pramoonmak[4], Boonsong Chongkolnee[4], Ponlapath Tipboonsri[4] and Anin Memon[4]

[1] Major in Architectural Technology, Faculty of Architecture
Rajamangala University of Technology Thunyaburi, Pathum Thani, 12110, Thailand
[2] Department of Industrial Engineering, Faculty of Engineering,
Rajamangala University of Technology Srivijaya, Songkhla, 90000, Thailand.
[3] Department of Industrial Engineering, Faculty of Industrial Education,
Rajamangala University of Technology Thanyaburi, Pathum Thani, 12110, Thailand.
[4] Department of Industrial Engineering, Faculty of Engineering
Rajamangala University of Technology Thanyaburi, Pathum Thani, 12110, Thailand.

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ABSTRACT

This research focuses on exploring the formulation of rubber composites by adding recycled polyethylene-aluminum composite from used beverage cartons, known as PolyAl, as reinforcing fillers into STR5L natural rubber (NR). The NR-PolyAl compound was mixed in an internal mixer and a two-roll mill. The study includes incorporating various PolyAl reinforcing fillers mixing at 0, 50, 100, and 150 phr in the first part, followed by the comparison of 60 phr PolyAl with carbon black 15 CB and 75 phr PolyAl in the second part. The difference in PolyAl significantly affects the tensile strengths and elongations at the break due to the dispersion of the PolyAl particles in the NR matrix. The PolyAl reinforcing fillers incorporation considerably enhanced the mechanical properties, including 100%modulus, compression set, and hardness. Adding magnesium oxide improved the tension properties of rubber composites. In the second experiment, the combination of PolyAl and CB resulted in 100% modulus, compression set, hardness, and abrasion resistance higher than the effect of 75 phr PolyAl reinforcing fillers. The potential properties of NR-PolyAl composites present a high-quality and feasible option for manufacturing rubber shoe pads, heel pads, insulation pads, and similar products.

1 INTRODUCTION

The beverage cartons are the preferred choice for sterilizing dairy products and foods, resulting in over 3,000 million tons of these cartons being used yearly. These cartons are made of a combination of six thin layers of cardboard. The layers from the outside include (1) polyethylene (PE) for external protection of moisture and antibacteria, (2) paper for stability and package resistance, (3) PE as an adhesion layer, (4) aluminum to prevent oxygen permeation, (5) PE as another adhesion layer, and finally, PE to seal in the product liquid. It is unacceptable that beverage cartons require over 400 years to decompose in landfills and release hazardous smoke when combusting deposal. Recycling beverage cartons is an essential and practical solution to these issues. Consequently, exploring new materials in groups of polymer composites combined with beverage cartons enhances their value and mechanical properties. These advanced materials have a wide range of potential applications, including roofs, tables, chairs, and more [1].

Thailand emerged as the leading exporter of natural rubber in 2022. The country shipped around 157.9 thousand metric tons of black rubber and concentrated latex and ribbed smoked sheet rubber as other exported variants [2]. When producing black rubber composites for tire production, including inner liners, sidewall carcasses, air springs, belts, conveyor wheels, and some vibration isolation devices, carbon black (CB) is commonly used as a reinforced filler. While black rubber with CB filler benefits the manufacturing economy, it is important to note that the carbon black production process has a detrimental effect on air pollution [3,4]. To achieve a sustainable economic solution and increase the value of rubber products, it is important to explore alternative materials for NR composites with different fillers. This approach would provide various benefits such as biodegradability, environmental friendliness, affordability, and reduction of domestic waste [5].

Regarding the aforementioned, this research focuses on studying and designing natural rubber compounds that incorporate recycled beverage cartons (PolyAl) as reinforced fillers with varying phr levels. The objective is to explore the mechanical properties of NR-PolyAl composites, making them suitable for rubber manufacturing.

2 MATERIALS AND METHODS

2.1 Materials

For this research, to make rubber compounds, STR grade 5L Natural Rubber (NR) was utilized as the NR matrix, along with recycled beverage cartons made of polyethylene and aluminum (PolyAl) as reinforcement fillers. The PolyAl particles were ground and measured to have a size in the range of approximately 864.8 µm (d10 = 630.9 µm, d50 = 891.1 µm, and d90 =1,142.3 µm), as determined by a Laser Particle Size Analyzer. Figure 1 displays the morphology of the PolyAl material.

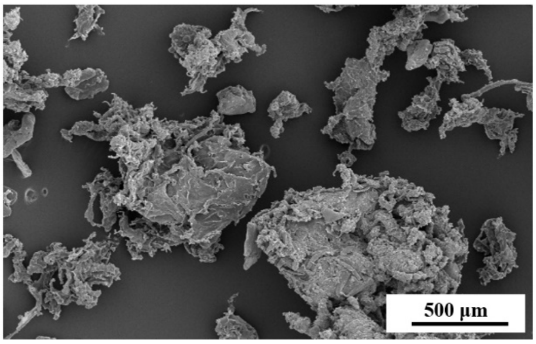

Figure 1: Morphology of PolyAl particles from recycling beverage cartons.

Different chemicals, such as zinc oxide (ZnO), stearic acid ($C_{18}H_{36}O_2$), dibenzothiazyl tetramethyl thiuram disulfide (TMTD), dibenzthiazyl disulphide (MBTS), magnesium oxide (MgO), carbon black (N330), and sulfur (S8), were added for rubber compounds portions.

2.2 Preparation of Rubber Composites

The NR-PolyAl compounds were prepared with two experiments using specific compositions;

For the first exploring composition, STR5L, NR matrix with PolyAl at 0, 50, 100, and 150 phr were mixed in an internal mixer. ZnO and stearic acid were also added during the mixing process to improve the strength and elasticity of the NR-PolyAl compounds. The NR-PolyAl compounds were mixed again with TMTD, MBTS, and sulfur in a two-row mill after stowing for 24 hours. The resulting mixture was then compressed using a hot compression molding machine for vulcanization. As part of this research, various tests were carried out to evaluate the properties of NR-PolyAl. These included tensile testing following ISO 188 and ISO 37, compression set testing following ISO 815, hardness following ISO 7619, and insulation resistance testing adhering to ISO 14309.

In the second exploring composition, the STR5L and the reinforcing fillers of PolyAl at 60 phr with CB at 15 phr and PolyAl at 75 phr were prepared in an internal mixer. In addition to ZnO and stearic acid, MgO and 6PPD were also added during the mixing process with the assumption of increasing more strength and elasticity of the NR-PolyAl compounds. For 24 hours, the materials were stowed prior to vulcanization. TMTD, MBTS, and sulfur were then blended in a two-row mill and compressed. The properties of NR-PolyAl were analyzed. Tensile testing was conducted using ISO 37 and ISO 188, while compression set testing was done with ISO 7743. Hardness testing was carried out using ISO 48-4 and ISO 188, and DIN abrasion testing was performed using ISO 4649.

Table 1 and Figure 2 display the components, apparatus, and preparation procedures of the NR-PolyAl compounds;

Components	Part I (phr)	Part II (phr)
NR (STR5L)	100	100
Reinforcement	PolyAl 0, 50, 100, 150	0, 60PolyAl/15CB, 75PolyAl
Zinc oxide (ZnO)	5	5
Magnesium oxide (MgO)	-	30
6PPD	-	2
Stearic acid ($C_{18}H_{36}O_2$),	2	2
Dibenzo thiazyl Tetramethyl thiuram disulfide (TMTD)	1	1
Disulphide (MBTS),	1	1
Sulfur (S_8)	2	2

Table 1: The composition of the rubber compounds.

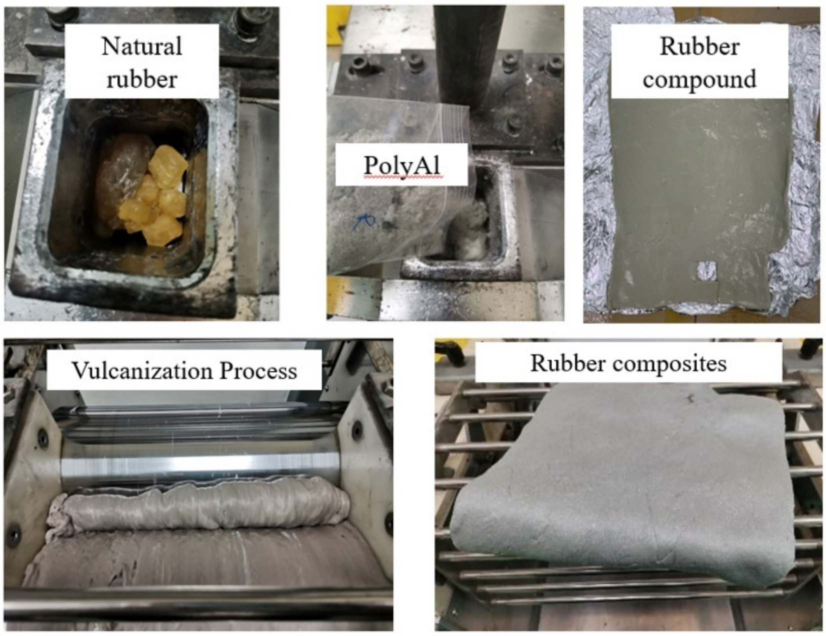

Figure 2: Rubber composites were mixed in an internal mixer and a two-roll mill

2.3 Analysis of Rubber Composites

2.3.1 Analysis Technique and Methods

In this research, the Laser Particle Size Analyzer (LPSA2) is a commonly used method for measuring particle size. It works by analyzing the angle of light scatter as particles pass through a laser beam. For the material morphology, the FEI Quanta 400 SEM scanning electron microscope was used to examine the adhesive bonding between the matrix and reinforced filler crystals present at the interface.

2.3.2 Mechanical Testing

The universal testing machine (Zwick Roell, Germany) was carried out to determine tensile properties according to ISO 37 and ISO 188 and investigated the compression properties of thermoplastic rubber using ISO 7743.

The compression set tester (Yasuda JIS A5756, Japan) was used to determine the compression characteristics of vulcanized rubber and vulcanized thermoplastic rubbers according to ISO815.

The DIN abrasion tester was used for evaluating the abrasion resistance of vulcanized and thermoplastic rubber according to ISO 4649.

2.3.3 Physical Properties

Hardness (Shore A) was determined using Bareiss, Germany according to ISO 7619, ISO 48-4, and ISO 188.

The insulation resistance was measured using the Hikoki SM8220, Japan which under measuring resistance range of $5.0x10^4$ to $2.0x10^{16}$ ohm and voltage ranges of 10, 25, 50, 100, 250, 500, and 1000 V.

3 RESULTS AND DISCUSSION

3.1 Mechanical and Physical Properties of NR-PolyAl Composites

The NR-PolyAl composites incorporated NR matrix with PolyAl reinforced fillers from ground beverage carton wastes were determined in various dimensions of mechanical and physical properties.

In Figure 3, the 100% Modulus, tensile strength, and elongation at the break were analyzed through tensile tests conducted on PolyAl fillers at 0, 50, 100, and 150 phr. These experiments were carried out at room temperature and after thermal aging at 70°C for 168 hours.

The NR-PolyAl composites demonstrated the lowest 100% modulus at room temperature but possessed the highest tensile strength and elongation at the break. Increasing PolyAl improves the 100% modulus in all cases, both at room temperature and in thermal aging states. Among the specimens tested, those containing PolyAl at 150 phr showed the highest 100% modulus, measuring 95.7% at room temperature and 96.5% for thermal aging. Meanwhile, the PolyAl at 100 phr showed the second-highest tensile strength and elongation at break. Conversely, the PolyAl at 50 phr demonstrated the lowest results under both states.

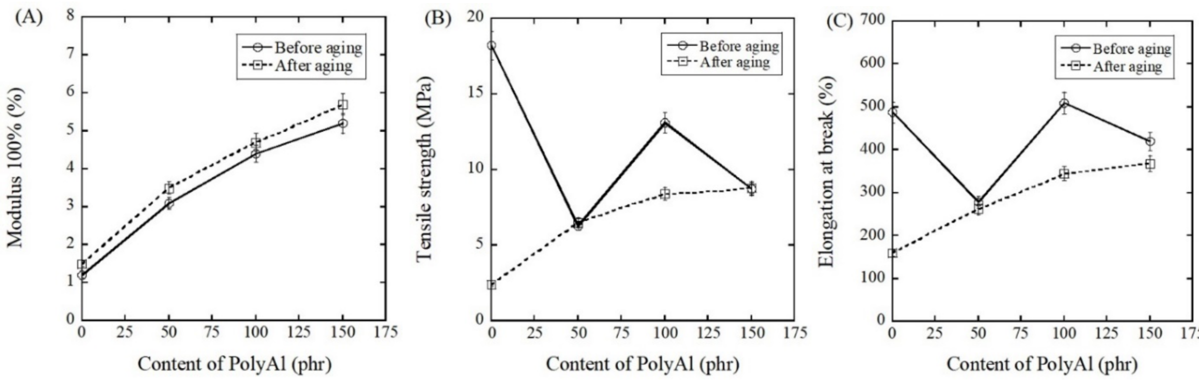

Figure 3: Tensile properties of NR-PolyAl composites :
(A) 100% Modulus, (B) Tensile strength, and (C) Elongation at the break.

The use of PolyAl led to an increase in the compression properties as shown in Figure 4, with higher amounts resulting in improved elastic properties of the NR-PolyAl composite. The compressive stress behaviors were positively impacted as well. Specifically, the 150 phr PolyAl led to the highest compression of 48.2%, which is 295.40% greater than the NR-neat result.

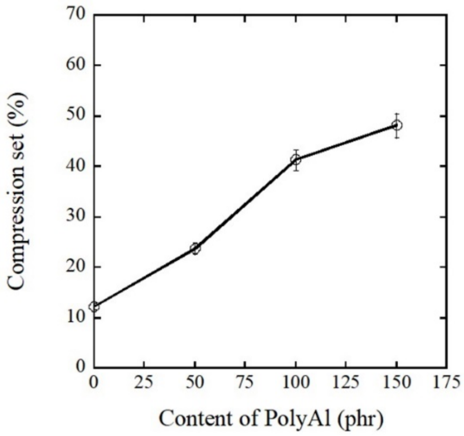

Figure 4: Compression characteristics of rubber composites reinforcing PolyAl.

Based on the results shown in Figure 5, it can be observed that the hardness tendency increased as the PolyAl reinforced fillers were increased. The composite of NR-PolyAl at 150 phr demonstrated the highest hardness level of 90.6 shore A, which is 84.5% greater than NR. However, the insulation properties showed a decrease with an increase in PolyAl. The NR-PolyAl composite at 50 phr had the lowest insulation resistance, measuring 2.81x1017 ohm.m which is 65.98% lower than NR, as depicted in Figure 6.

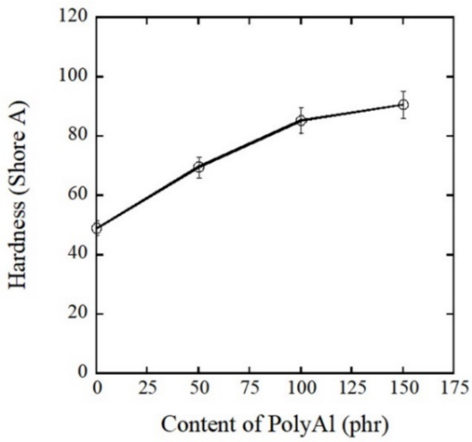

Figure 5: Hardness results of rubber composites reinforcing PolyAl.

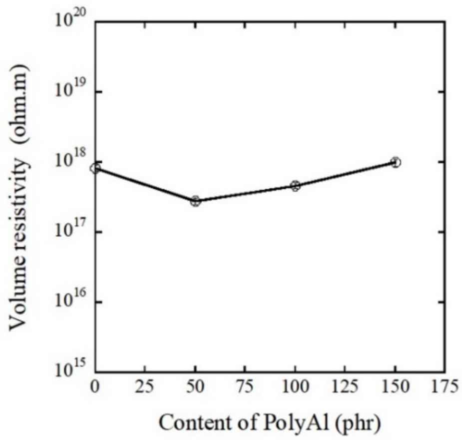

Figure 6: Volume resistivity of rubber composites reinforcing PolyAl.

In Figure 7, a comparison is presented on the impact of including magnesium oxide (MgO) at 30 phr and carbon black (CB). The experiment tested with different compositions: 1) NR-neat, 2) NR-m, 3) NR-m with 60PolyAl/15CB, and 4) NR-m with 75PolyAl. Based on the results, it was found that the NR-m composites showed the greatest tensile strength and elongation at the break. After analyzing the different reinforced fillers between NR-m with 60PolyAl/15CB and a single PolyAl, it was discovered that NR-m with 75PolyAl resulted in composites with higher tensile strength and elongation at break compared to NR-m. Based on Figures 8, 9, and 10, it is evident that the hybrid fillers, NR-m with 60PolyAl/15CB and NR-m with 75PolyAl, have significantly enhanced the compression. According to the data presented, the composites exhibited a strength of 24.9 N/mm^2 and were 857% higher than that of the Nr-m composites. Additionally, the NR-m with 75PolyAl showed a hardness of 89.6 shore A and abrasion of 279 mm^3, which was 57.6% greater than that of the Nr-m composites in both categories.

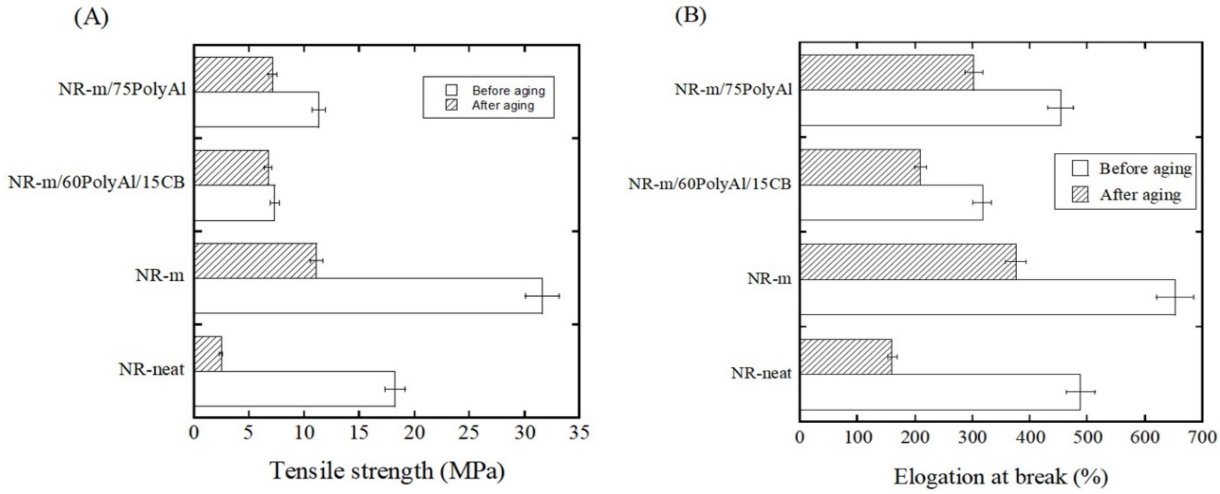

Figure 7: Tensile properties of rubber composites as reinforcing fillers at 75 phr;
(A) Tensile strength, and (B) Elongation at the break.

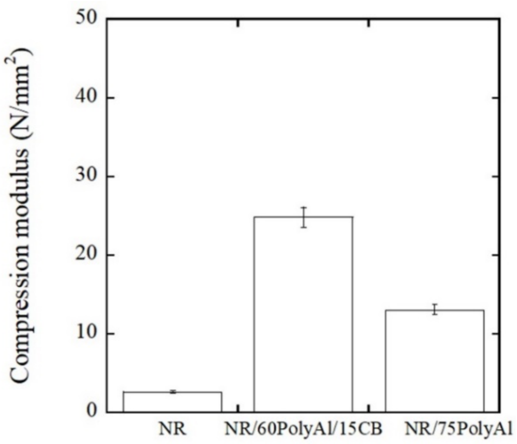

Figure 8: Compression properties of rubber composites as reinforcing fillers at 75 phr.

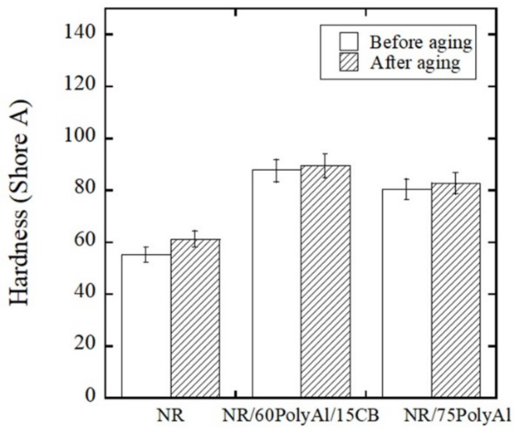

Figure 9: Hardness results of rubber composites as reinforcing fillers at 75 phr.

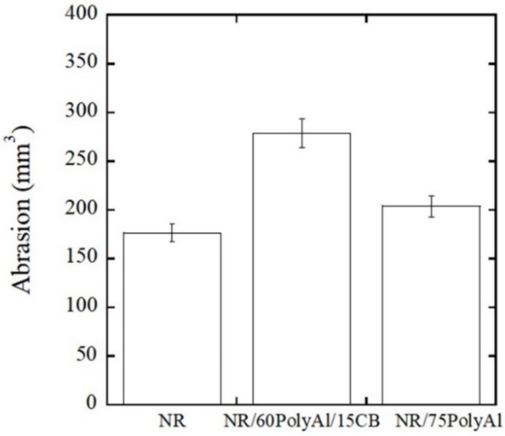

Figure 10: Abrasion results of rubber composites as reinforcing fillers at 75 phr.

3.2 The Morphology Properties of NR-PolyAl Composites

In Figure 11, the dispersion behaviors of PolyAl reinforcing fillers in the NR matrix are displayed at 100 and 500 µm. The study analyzed different compositions, including (A) and (B) of NR/50PolyAl, (C) and (D) of NR/100PolyAl, and (E) and (F) of NR/150PolyAl. As the amount of PolyAl increased, the distance between its particles decreased, which was observed through a comparison. The dispersion behavior of NR/50PolyAl depicted in Figure 11 (A) and (B) revealed the highest dispersion between PolyAl particles. The increase in PolyAl had an impact on the stress between the interfaces of the matrix and reinforced fillers, resulting in a tendency to decrease the tensile strength and elongation at break. Figure 11(C) to (F) demonstrate that the dispersion of PolyAl in the NR matrix is good at 100 and 150 phr. Adding a significant amount of PolyAl particles has a positive impact on improving the 100% modulus, tensile strength, elongation at break, compression set, and hardness.

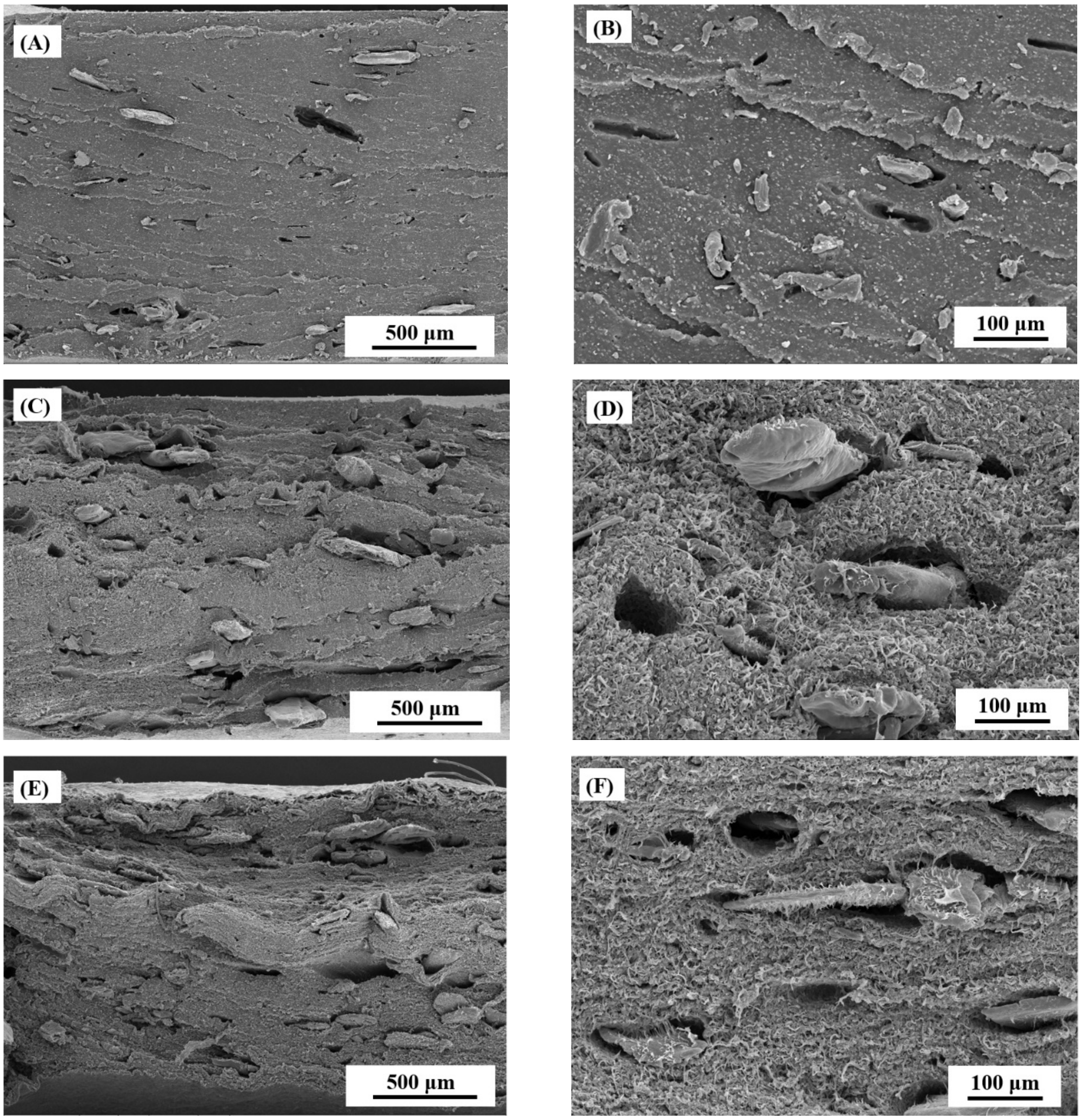

Figure 11: SEM images of NR-PolyAl composites with
(A) 50 phr, (B) 100 phr, and (C)150 phr of PolyAl.

4 CONCLUSIONS

This research found that the PolyAl reinforcing fillers were very effective in improving tensile strength and elongation at break. After thermal aging, the hybrid fillers showed even greater improvements in these areas compared to NR. After conducting tests, it was found that NR combined with 150PolyAl showed improved 100% modulus, compression set, and hardness. However, it was observed that increasing the amount of PolyAl in NR-PolyAl composites resulted in the decreased of insulation properties. Upon analyzing the morphology with SEM, it was determined that increasing PolyAl from 100 to 150 phr resulted in enhanced particle dispersion within the NR matrix. This improved dispersion ultimately led to better mechanical properties of NR composites. Incorporating magnesium hydroxide led to NR-m composites displaying superior mechanical properties in terms of tensile strength and elongation at break when compared to NR composites. When comparing hybrid fillers of NR with PolyAl and CB against a single filler of PolyAl at 75 phr, it was observed that the 60PolyAl/15CB combination demonstrated the highest 100% modulus, compression set, hardness, and abrasion. However, a single PolyAl at 75 phr displayed greater tensile strength and elongation at break than the hybrid fillers.

5 ACKNOWLEDGEMENTS

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