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บทความวิจัย

Mechanical, Rheological, and Morphological Properties of PP/Al composite modified by PP-g-MAH

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Abstract: This research studied the effects of a coupling agent on materials to improve the compatibility of PP/Al blends using PP grafted with maleic anhydride (PP-g-MAH) as a coupling agent. The research focused on the effects of a coupling agent that affected the composition of the mixtures. The polymer matrix composites with PP-g-MAH were used as a coupling agent and were prepared with an internal mixer at a temperature of 180°C and a speed of 60 rpm, varying Al powder from 0 – 10%wt and fixed PP-g-MAH. The flexural strength and impact strength were determined. The polymer matrix composites (PMCs) showed a positive effect on the mechanical properties. The trend of the melt flow index increased when increasing aluminium powder. The part of the morphology of PMCs with non-PP-g-MAH revealed that the particles of aluminium fine powder dispersed in the matrix phase, but that there were many microvoids between filler and matrix. On the other hand, PMCs with PP-g-MAH caused the microvoids between filler and matrix to reduce.

Keywords: Polymer matrix composites, PP-g-MAH, aluminium, polypropylene

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1. Introduction

A composite material is one that combines at least two different types of components into a single homogeneous substance. The physical and chemical characteristics of each material used will vary in each composite material. Composite materials can improve mechanical, thermal conductivity, electrical conductivity, and natural degradation properties, as well as boost strength, durability, and efficiency. The matrix and reinforcement are the two fundamental components of composite materials. The matrix phase protects the reinforcement from environmental forces acting on the material. As a result, to keep the materials together, a reinforcing agent is employed. The most common type of reinforced material is fibre; however, granules or particles can also be used to strengthen things [1].

At present, polymer matrix composites (PMCs) are widely used in the production of engineering components. It resulted in the invention of new types of materials and the continuous development in engineering. The advantages of PMCs include: lightweight, high strength, high stiffness, good abrasion resistance, good corrosion resistance, and good electrical conductivity [2]. PMCs will have a polymer matrix phase and will be reinforced by adding additional groups of materials. They have been using polypropylene to make a matrix phase and added aluminium as reinforcement such as Bondenne et al. [3] studied the thermophysical properties of polypropylene/aluminium composites using two different types of aluminium (slightly oxidized) filler particles were used. Chavooshi et al. [4] studied the mechanical and physical properties of aluminium powder / MDF dust / polypropylene composites using metal powders and MDF production waste as alternative sources to natural fibers in manufacturing wood-plastic composites (WPCs) and maleic anhydride grafted polypropylene (MAPP) as a coupling agent. Shuai et al. [5] studied the mechanical properties and fracture morphology of $Al(OH)_3$ /polypropylene composites modified by PP grafting with acrylic acid. Osman et al. [6] studied the properties of aluminium-filled polypropylene composites. In addition, various materials such as metal [7-9] and fibers [10-11] were added to polypropylene.

Polypropylene (PP) is a thermoplastic that has morphology and is a semi-crystalline polymer. The properties of polypropylene are colorless, transparent, and translucent. The surface is glossy and resistant to acids, bases, and various chemicals. They show toughness, as electrical insulation and has a good heat resistance. It can be used to make many products such as tubes, children's toys, artificial fertilizer bags, carpets and mat, tarpaulin, rope, straps, packaging, hot bags, beverage bottles, candy bags, tubes, wires and cables, and car interior equipment, etc [12].

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Metals are a group of materials that are mixed with a polymer to create composite materials. One of the most important is aluminium (Al), which is used in engineering today. Also, aluminium is a metal that is found in everyday life and is used in many applications, secondary to iron and copper. For example, it is used to make household kitchen utensils and construction materials. The advantage of aluminium is that the metal is lighter than steel and copper. It is cheap and does not rust [13]. Besides, aluminium also has many forms. The research focused on the aluminum powder form which today uses aluminium powder as a silver coating in paint, such as wood coating (primer), etc.

The coupling agent is a substance that helps increase contact adhesion between the matrix phase and the dispersed phase, to reduce the gap between the phases, resulting in better transmission of force. The coupling agent is used a lot in composite materials because without compatibility, the matrix and separation phase will be separated, with space between the phase's gaps. These gaps will result in poor transmission and may be the source of the material's failure. As a result, adding the coupling agent will aid in closing the gaps between phases. The materials can be mixed since the coupling agent has sticky components. The compatible side functions as a chain, assisting both phases in sticking together and increasing the contact surface area between them. This results in the materials being stronger and can be utilized to their maximum potential. [14].

Therefore, the goal of this study was to compare materials with and without a coupling agent to determine the properties of polymer matrix composites (PMCs) between aluminium powder and thermoplastic. This encompasses the study of mechanical, rheological, and thermal properties.

2. Materials and Methods

2.1 Materials

The PP grade HP400H, with a density of 0.90 g/cm^2 and a melt flow rate at 230 °C and with a hammer load of 2.16 kg and 2.1 dg/min used in this research was supplied by HMC Polymer, Thailand. Figure 1 shows an irregular shape of aluminium fine powder with an average size of 75 µm, and a density of 3.2 g/cm^3. Al powder was obtained from the HiMedia Laboratories Pvt. Ltd. As a coupling agent, polypropylene graft maleic anhydride (PP-g-MAH) was used. PP-g-MAH was obtained from Aldrich Chemistry, with an average Mw ~9,100 by GPC, average Mn ~3,900 by GPC and maleic anhydride 8-10 wt%.

2.2 Preparation of PMCs

This research investigated and observed the changes in PMCs properties between polypropylene and aluminium powder by using a closed system for the mixing process under the melt mix condition

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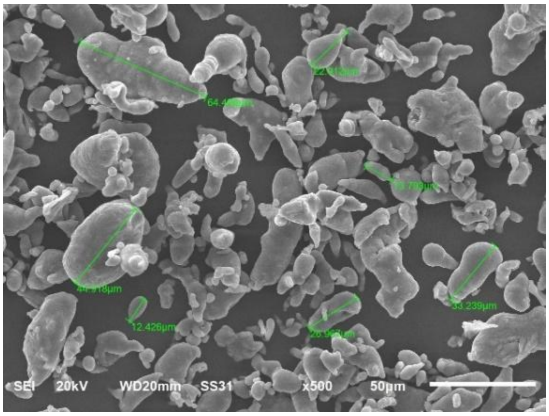

Fig. 1 SEM micrographs of aluminium fine powder at 500x

Table 1 The proportions of compound materials by using the internal mixer

Procedure	Amount (phr)		
	PP	Al	PP-g-MAH
Part I	100.00	-	-
	97.50	2.50	-
	95.00	5.00	-
	92.50	7.50	-
	90.00	10.00	-
Part II	97.50	2.50	1
	95.00	5.00	1
	92.50	7.50	1
	90.00	10.00	1

with the proportions of aluminium powder as follows: The proportions of the mixtures used to make composite materials are shown in Table 1, it is divided into two sections.

Part I, the preparation of composite materials made of polypropylene and fine aluminium powder, began with the drying of PP in a 50°C oven for 24 hours to repel moisture. This was then mixed with an internal mixer (model MX500-TQ, CHAREON TUT CO., LTD) at a temperature of 180°C and a speed of 60 rpm. The mixing process began by adding PP and mixing it for 8 minutes in an internal mixer followed by adding fine aluminium powder and then mixing it for 7 minutes.

Part II, consisted of putting 1 wt% PP-g-MAH in the PMCs. It started by mixing the PP for 8 minutes in an internal mixer, followed by mixing aluminium fine powder for 5 minutes, and 2 minutes of PP-g-MAH.

Furthermore, the specimens of PMCs were prepared for testing the mechanical properties (flexural strength, impact strength and hardness) and rheological property (MFI).

2.3 Composite Characterization

2.3.1 Mechanical Properties

The mashed PMCs were dehydrated in an oven for 24 hours at 60°C forming specimens for flexural testing (ASTM D790), and Izod bar (ASTM D256) for impact resistance and hardness test. The preparation of the sample was created by using a hydraulic hot compression. The installation equipment used electric heating and cooling water. The operating condition was set to a load of 100 kg_f/cm^2 and a compression temperature of 180°C. Flexure, impact, and hardness mechanical properties were investigated. Flexural properties were determined using an ASTM D790 [15] by Universal Testing Machine (Model H50KS,

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Tinius Olsen) with a crosshead speed of 1.3 mm/min and a specimen span length of 50 mm. The impact strength test was carried out with a pendulum weight of 9.198 N on a Pendulum Impact Testing Machine (Model IT504), according to the ASTM D256 standard [16]. The hardness of the materials was determined using a hardness testing machine (model GS 120G Type D), following the ASTM D2240 [17]. The reported results were the average of five measurements taken at room temperature for each sample.

2.3.2 Rheological Property

The Melt Flow Index (MFI) was set at 230°C with a hammer load of 2.16 kg in accordance with ASTM 1238 [18]. The PMCs sample was fed into the cylinder for 1 min with a weighted portion according to the expected flow rate. The weight piston was placed in position and then started timing the extruded when the required position for the piston was met. Next, the extruded portion was collected exactly according to the time interval. Then, weighed the extruded PMCs and multiplied the weight by the factor to obtain the flow rate in grams per 10 min.

2.3.3 Morphological Property

The morphological property was investigated with scanning electron microscope (SEM; JEOL Ltd., Tokyo, Japan Model JSM-6610LV). The observation was performed on the fractured surfaces during the impact resistance test. The surface of the samples were covered by gold for 4 mins. The test results of SEM were used to observe the morphology.

3. Results and Discussion

3.1 Mechanical Properties

Flexural, impact and hardness testing results of PMCs are shown in Table 2. It showed that Neat PP, PP with aluminium powder, vary conditions and revealed that by adding PP-g-MAH in PMCs, the value was confirmed by impact strength (IS), flexural strength (FS), flexural modulus (FM) and hardness.

The impact resistance is an indication of the energy that the materials can absorb before a sudden force breaks it. In situations in which the material has high energy absorption values, it will be able to absorb energy well and withstand a lot of impacts. From the results in Fig. 2, we found that the trend of impact resistance decreased when increasing aluminium powder for both non-PP-g-MAH and using PP-g-MAH. This reflects that when adding solid particles to the ductile material, composites changed the behavior of ductile materials to be more hard and brittle materials. A coupling agent was used to improve the adherence of aluminium powder to polypropylene. As a result, the material's behavior altered to a more brittle hard form, and brittle materials lost their capacity to withstand the impact.

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Table 2 Mechanical properties of PMCs

Condition	IS (kJ/mm^2)	FS (MPa)	FM (MPa)	Hardness (Shore D)
Neat PP	3.28 ± 0.21	33.51 ± 1.60	542.49 ± 16.67	65.63 ± 0.41
PP + Al2.5	2.71 ± 0.18	47.39 ± 1.30	672.82 ± 19.38	66.47 ± 0.89
PP + Al5.0	2.64 ± 0.13	51.06 ± 1.00	804.80 ± 15.79	66.97 ± 0.78
PP + Al7.5	2.28 ± 0.10	58.92 ± 2.14	865.19 ± 39.74	68.40 ± 1.00
PP + Al10.0	2.20 ± 0.22	54.27 ± 2.52	998.79 ± 11.14	69.93 ± 1.54
PP + Al2.5 + PP-g-MAH	2.19 ± 0.23	50.12 ± 0.99	724.59 ± 18.90	68.80 ± 1.10
PP + Al5.0 + PP-g-MAH	2.18 ± 0.25	52.73 ± 1.89	855.70 ± 18.49	69.57 ± 1.41
PP + Al7.5 + PP-g-MAH	2.09 ± 0.23	60.76 ± 1.83	885.17 ± 35.10	70.37 ± 1.09
PP + Al10.0 + PP-g-MAH	2.06 ± 0.17	58.74 ± 1.74	1080.77 ± 16.55	71.33 ± 1.27

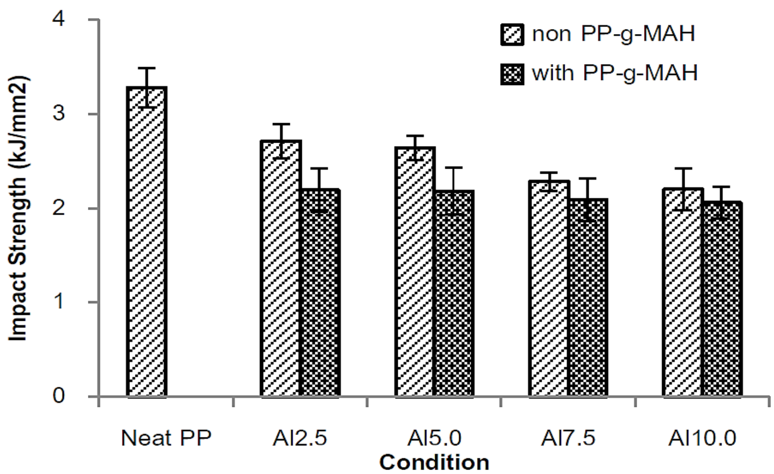

Fig. 2 Impact strength of PP/Al composites with different Al powder contents

Flexural strength (FS) and Flexural modulus (FM) refers to a material's ability to withstand bending stresses applied perpendicular to its longitudinal axis [19-20]. The addition of filler to composites enhances flexural strength, as seen in Fig. 3 (a) and flexural modulus, as seen in Fig 3 (b). The FS is expected to increase further with the addition of Al powder loading if and only if the interfacial adhesion between Al and the polymer matrix is strong enough. The strength of the contact is determined by the surface energy of the components. Aluminum has a rather significant interaction with the matrix and filler due to its high surface energy. The quality of the interface in composites has a substantial influence on the flexural properties, according to Mirjalili et al. [20].

From the results of the flexural strength test, it was found that the flexural strength shown in

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Fig. 3 (a) and flexural modulus shown in Fig. 3 (b) of the PMCs without PP-g-MAH was higher than that of neat polypropylene, and the value increased when the amount of aluminum powder increased. Due to the addition of fine aluminum powder, the aluminum powder was well dispersed in the polypropylene, and aluminum powder was harder than polypropylene. When aluminum powder was mixed with polypropylene, the composite had both higher flexural strength and flexural modulus. But when increasing the amount of aluminum powder with more than 10 wt.%, it was found that flexural strength and flexural modulus reduced due to the adhesion of aluminum powder (Agglomerate). The results of these tests were consistent with the research of Shuai et al. [5] that studied the mechanical properties and fracture morphology of Al(OH)3 /polypropylene composites modified by PP grafting with acrylic acid, it was found that when adding a small amount of $Al(OH)_3$, it was well dispersed in the polyethylene body. It did not clump together, but when more of $Al(OH)_3$ was added, they clumped together and created a stress concentration. For the tensile test, Prathumrat et al. [21] studied the effects of PMCs added polypropylene graft maleic anhydride (PP-g-MAH) in PMCs (PP/Al), the amount of Al was 0, 2.5, 5, 7.5 and 10%wt. Tensile strength was in the range of 20-30 MPa and young's modulus in the range of 400 – 500 MPa. The tensile strength tended to be close to the flexural test, i.e., the highest value was at 7.5%Al and at 10%Al there was a decrease in tensile strength. The compatibilizer (PP-g-MAH) influenced PMCs' ultimate tensile strength. When compared to samples without compatibilizer, the compatibilizer enhanced the tensile strength trend and the Young's Modulus of PMCs. This phenomenon's cause has been well described by them.

From the results of the experiment of the hardness test in Fig 4, it was found that when comparing the hardness of neat polypropylene and those by adding aluminium powder in various proportions, the average hardness value increased when increased with aluminium powder. Due to the insertion of aluminium powder into the matrix of polypropylene in a solid form it created materials with a higher hardness than polypropylene, thus making composite materials more rigid when the amount of aluminium powder inserted increased. When comparing PP-g-MAH and non-PP-g-MAH, it showed that the trend of hardness was increased because of the influence of a coupling agent, resulting in the strong interaction between the aluminium powder and the polypropylene that reduced the gaps between the phases. The distribution of particle groups was further separated, so the binder made the hardness of the composite materials higher.

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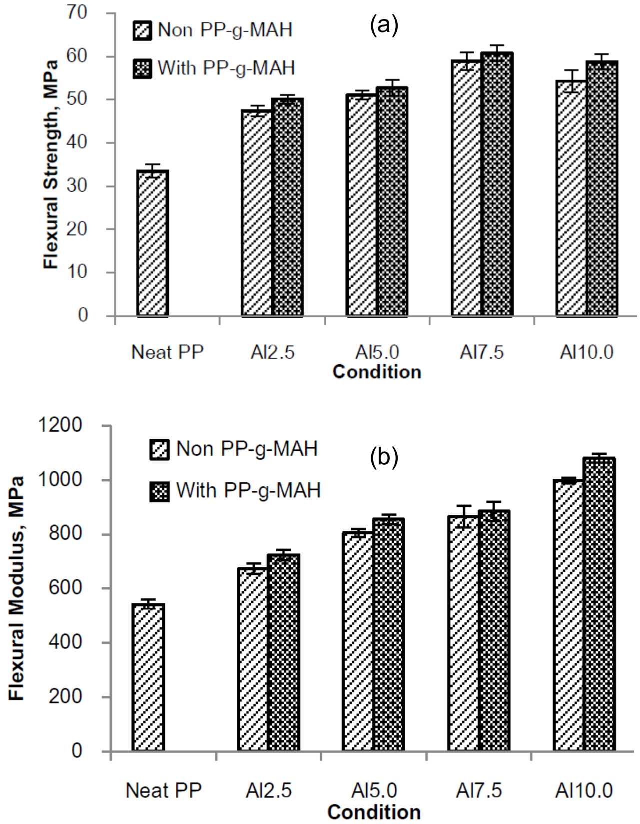

Fig. 3 Flexural strength PP/Al composites with different Al powder contents

(a) Flexural strength and (b) Flexural modulus

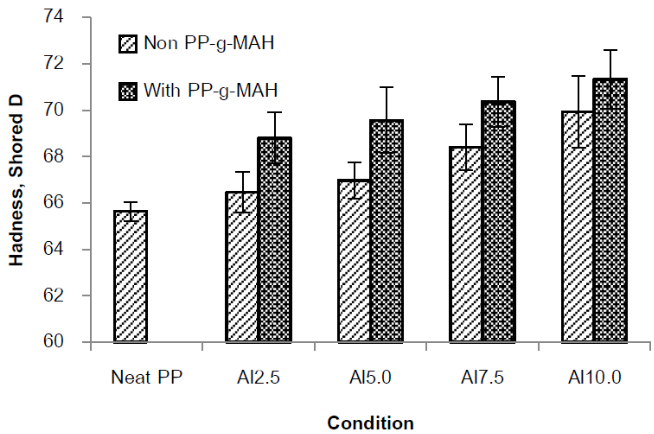

Fig. 4 Hardness of PP/Al composites with different Al powder contents

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3.2 Rheological Property

In Fig. 5, the melt flow index (MFI) of a PMCs between polypropylene, aluminium powder and non-PP-g-MAH, showed that the neat PP flow index is 22.04. However, when mixing it with aluminium powder it was found that the trend of melt flow index increased. It may be because aluminium has heat conductivity when the aluminium is heated for the test. The heat was distributed to the polypropylene. As a result, aluminium powder interfered with the slipping of the polymer chain, making PP easier to melt. When adding PP-g-MAH, the melt flow index increased and PP-g-MAH accelerated the deterioration of the polypropylene chain resulting in a chain that was easier to pass through and the trend of the melt flow index was higher. But when adding aluminium powder at 10%wt, the melt flow index value decreased because of the excessive amount of aluminium powder causing it to obstruct the flow in the hole of the tester.

3.3 Morphological Property

From the morphology study of polymer composite materials between polypropylene and aluminium powder with non-PP-g-MA, it showed that PMCs are heterogeneous. In Fig. 6, there is a phase separation of aluminium particles scattered on the polypropylene. It can be seen from the shadows on the aluminium powder particles, which reflect the incompatibility between the polypropylene and aluminium powders. That is the result of aluminium having a polar and polypropylene is non-polar. On the other hand, when using PP-g-MAH in PMCs in Fig. 7, it was found to be more homogeneous than non-PP-g-MAH. Because of this, it didn't show gaps in the aluminium particles between the phases occurring. This shows that the coupling agents help to increase the bond strength between polypropylene and aluminium powder for better adhesion.

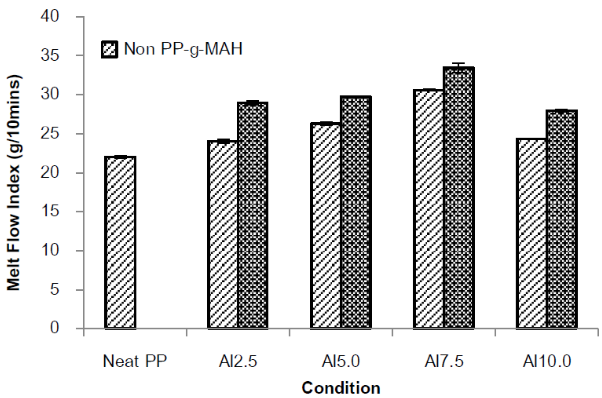

Fig. 5 Melt Flow Index (MFI) of PP/Al composites with different Al powder contents

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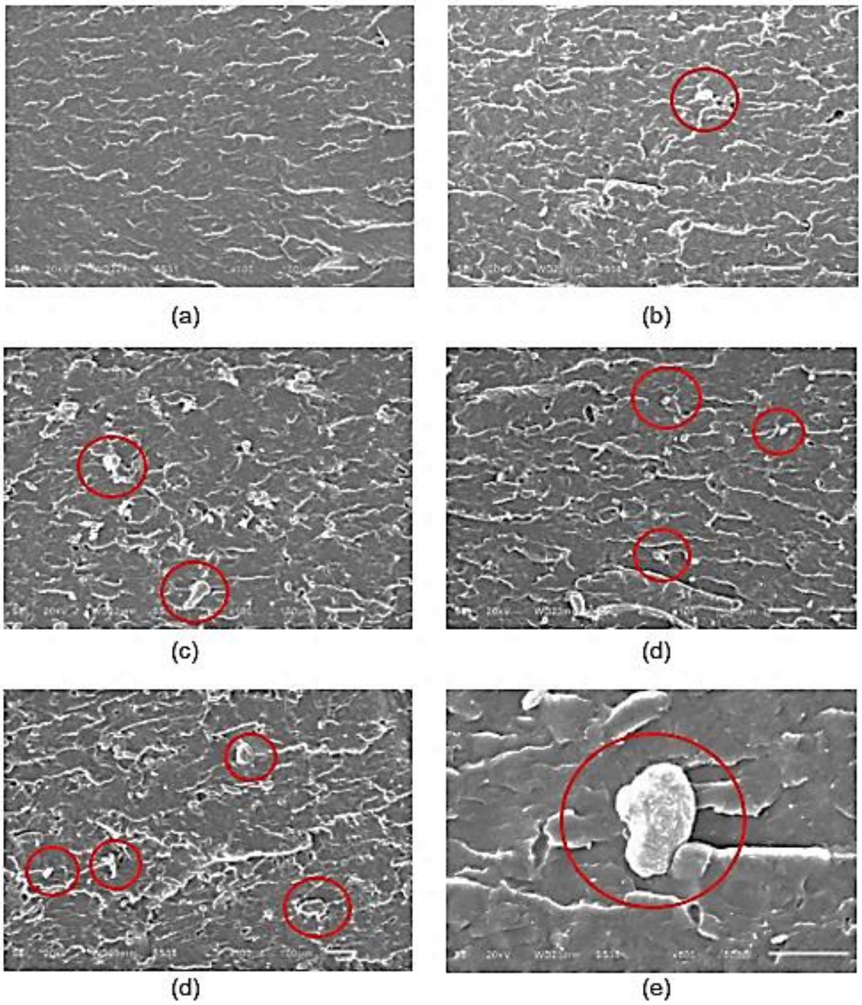

Fig. 6 SEM micrographs x500 PP/Al composites with different Al powder contents non-PP-g-MAH (a) PP100% 100X, (b) PP/Al2.5% 100X, (c) PP/Al5% 100X, (d) PP/Al7.5% 100X, (e) PP/Al10% 100X and (f) PP/Al10% 500X

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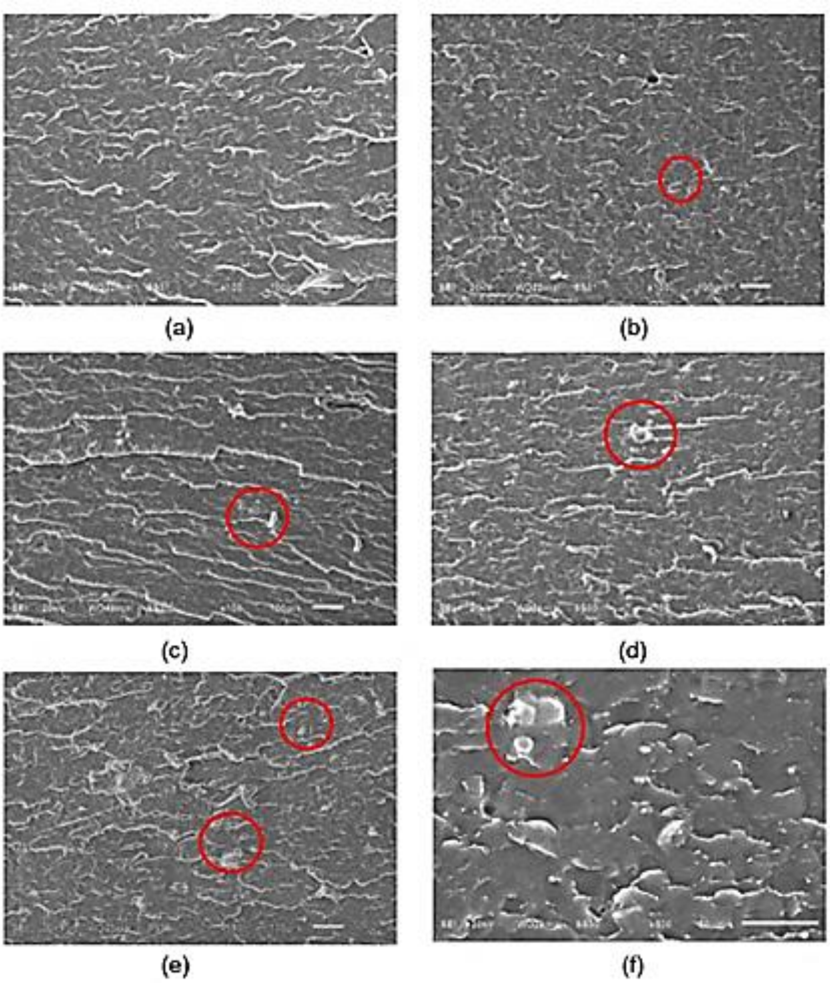

Fig. 7 SEM micrographs x500 PP/Al composites with different Al powder contents adding PP-g-MAH (a) PP100% 100X, (b) PP/Al2.5% 100X, (c) PP/Al5% 100X, (d) PP/Al7.5% 100X, (e) PP/Al10% 100X and (f) PP/Al10% 500X

Since the distribution of aluminium powder particles in the polypropylene surface is not visible from SEM, the EDX was tested to see the particles of aluminium powder inserted in the polypropylene as shown in Fig. 8, it was found that the aluminium powder particles were infiltrated and distributed in the polypropylene which was influenced by the addition of the coupling agent. It is possible to confirm that the coupling agent can increase the adhesion between the polypropylene phase and the aluminium powder.

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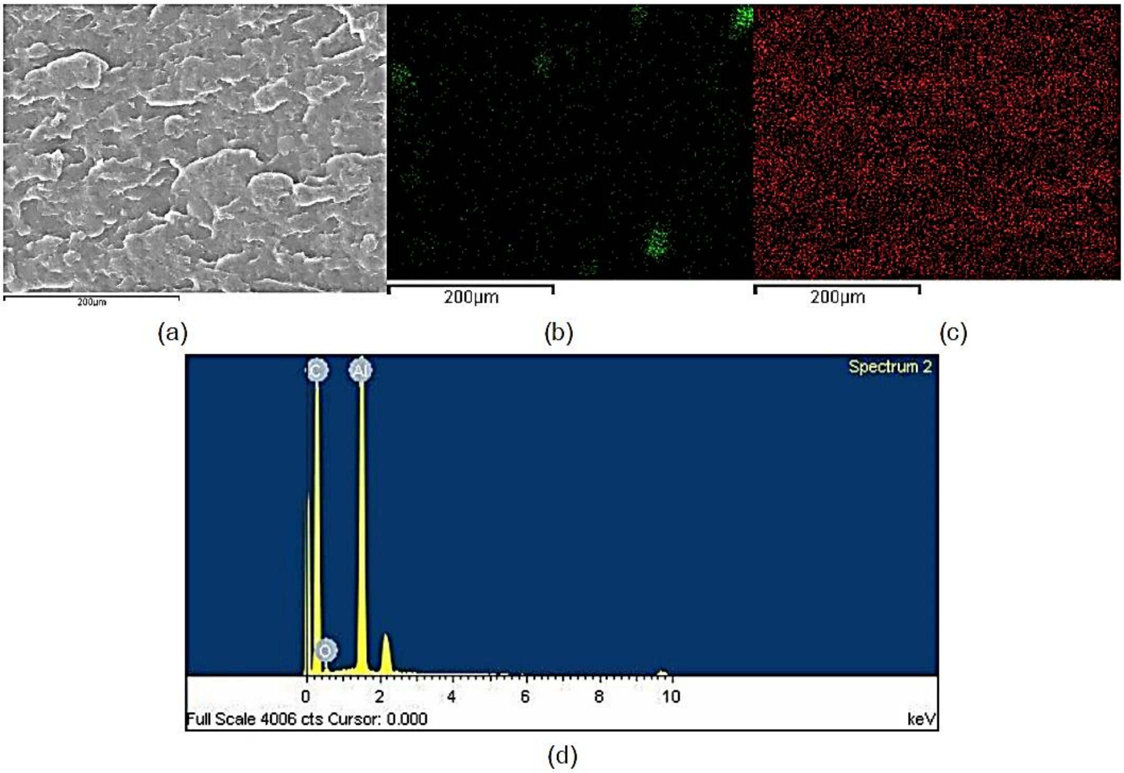

Fig. 8 SEM-EDX of PMCs based on PP/Al composites: (a) SEM and (b) EDX of aluminium, (c) EDX of carbon and (d) spectrum of PMCs with PP-g-MAH

4. Conclusions

The influence of coupling agents on the properties of PMCs (PP/Al) were investigated. The first portion was mixed into the polypropylene with aluminum powder with a particle size of less than 75 µm, and the second part was added to the composites with the coupling agent by utilizing polypropylene graft maleic anhydride (PP-g-MAH). Using an internal mixer for mixing and a compression machine to form a test specimen.

From the results of the melt flow index (MFI), mechanical properties and morphology investigations of composites, revealed that the PMCs without the coupling agent, the melt flow index, flexural strength, flexural modulus, and hardness increased when compared to neat polypropylene. While the impact resistance decreased. The morphology of PMCs was examined at the interface between the aluminum powder and the polypropylene matrix. It was found to have large gaps, and there was an incompatibility between polypropylene and

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aluminum powder in that there was an agglomeration of aluminum powder.

When PP-g-MAH was introduced to PMCs (PP/Al), the results were better than when no PP-g-MAH was used. When utilizing PMCs with PP-g-MAH to analyze morphology, it was discovered that there were no interphase gaps in the area around the aluminum powder particles. This demonstrates that the binder improves the bonding strength between polypropylene and aluminum powder, resulting in improved adhesion. When the coupling agent was used to polarize polypropylene, it interacted strongly with the polarized aluminum particles. As a result, the space between the phases narrowed, resulting in improved properties.

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