

ORIGINAL ARTICLE

Study of Lignin Addition as Coupling Agent on Paving Block Properties Made with Polypropylene

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ABSTRACT – Polypropylene is well-known as a type of packaging material and thus results in abundant amount of waste. Several studies have attempted to use plastic as aggregate replacement in paving block as a waste recycling effort. On the other hand, paper industry also produces various byproducts, such as lignin. In this study, lignin was added as a coupling agent in the mixture to enhance the properties of the obtained paving block. The purpose of this study was to investigate the mixing properties of plastics, lignin, and aggregates in paving blocks. This study utilized PP and lignin, with a variation of 0; 0.1; 0.3; 0.5 wt.%. SEM was used to observe morphology, while FTIR was carried out to identify the functional groups of the mixture. Additionally, contact angle test was done to determine the surface tension of each component and showed that PP is compatible with lignin because of their similar properties in hydrophobicity.

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INTRODUCTION

Plastics are ubiquitous in our everyday life, to the point which it is unimaginable today to live without it. The ensuing rapid growth in plastics production is supported by the demand of the community, which surpassing most other man-made materials, particularly as packaging [1], [2]. Notable exceptions are materials that are used extensively in the construction sector, such as steel and cement [3]. However, this fame came with a cost in the form of abundant plastic waste [1].

Waste management has always been a complicated issue in every country. The amount of waste, both organic and inorganic, has increase due to population growth and human activities [4]. In 2025, the amount of waste produced in Indonesia reached 38.62 million tons, of which only 34.25% being managed, according to BPS-Statistics Indonesia 2025 [5]. Approximately 2/3 of total waste is still unmanaged, which is around 25.39 million tons. From those data, 19.5% of those wastes are plastics and 11.1% of it is paper-type waste. Other huge contributors are food waste with 38.3% and wood and branch with 13.2% [5]. Based on those data, plastic and paper waste are top 4 of waste contributor in Indonesia, which rise a profound issue about waste management. Additionally, plastic waste takes ages to decompose and consequently becomes a pollutant in the soil. Not to mention, some plastics emit harmful pollutants to the soil and disrupt environmental health [6].

Therefore, utilization of plastic waste is really needed as an effort to reduce long-term harmful effects of waste. Not only beneficial for environment, but also for economic benefit [7]. It has been efficiently used to manufacture less costly plastic bricks. Its usage as a binder in the production of paver blocks has been tested and successfully applied in everyday usage. In contrast, plastic waste has been used as a viable and prospective replacement of cement [8], coarse aggregates [9], and fine aggregates [10] with its addition in concrete block. Plastic waste can be used as a substitute for aggregate in base and sub-base construction for pavement, which has been known to enhance the pavement's shear, stiffness and bearing capacity [2], [3], [7].

Several studies have been conducted to apply plastic waste to form paving block [6], [8], [9]. However, none of those studies utilized or added lignin in their work. Lignin as one of the byproducts from paper and pulp industry is known as coupling agent and has been applied as such in other studies, particularly as binding agent in construction sector such as concrete [11] and asphalt [3]. Miao et al. [11] concluded that the usage of lignin, hydrolysis- and alkali-based lignin, to develop concrete holds promising potential due to its ability to produce concrete materials with compressive strength from 1.6 – 8.1 MPa, which allowed them to be implemented in non-structural construction application (e.g. pavement, roofing tiles, and non-structural elements) based on ASTM C129 and also reduce global carbon emissions from conventional concrete production. Thus, lignin application in construction materials opens a promising potential in the field of sustainable construction materials [3], [11]. Based on those previous studies, this work utilized lignin as coupling agent in paving block production using polypropylene (PP). This work focused on the effect of lignin on paving block made using plastic waste with variation of lignin concentration of 0; 0.1; 0.3; 0.5 wt.%.

EXPERIMENTAL METHOD

Materials and Instruments

This study utilized river sand as fine aggregate, PP as a coarse aggregate substitute, lignin as a coupling agent, and cement as a binder. The PP used in this study was virgin plastics, PP RI10HO, the lignin was pure lignin extracted from paper and pulp industry waste, and the cement was Portland cement type I.

The obtained mixture was analyzed using SEM (Scanning Electron Microscope) and FTIR (Fourier-Transform Infra-Red Spectroscopy) instruments. Furthermore, the contact angle between materials was tested with wettability test to investigate suitability of every material used in the study. SEM characterization was performed using Thermo Fisher Scientific Quanta 650 by coating the samples and 10 kV accelerating voltage, while FTIR was conducted using Agilent Cary 630 with ATR method and wavenumber between 4000 – 400 cm^{-1} . SEM was used to analyze the morphology between PP as coarse aggregate and river sand as fine aggregate and the effects of lignin addition on it. FTIR was utilized to investigate functional groups and chemical bonds. The characterization of wettability properties of samples was measured using contact angle values of distilled water and diethylene glycol. Tested samples were attached to a glass slide by double side tape and fixed in the form of a plane. The static sessile drop method was used to measure water contact angles. The test was conducted to observe the hydrophobicity of PP and lignin mix and indicate compatibility between materials. The surface tension of distilled water and diethylene glycol were shown by Table 1.

Table 1. Effect of pressure on flux for different thicknesses

No.	Liquid	Surface Tension (mN/m)
1.	Distilled water – Air	72.8
2.	Diethylene glycol - Air	48.5

Method and Procedure

Sand, PP, cement, and lignin were dry mixed for 15 minutes using dry mixing tool. The ratio of cement to other materials was 1:4. During the mixing process, water was added with water-to-cement ratio (w/c) of 0.4 to hydrate the mixture. Then, the obtained mixture was poured into a mold for paving block. Lignin addition varied into 0, 0.1, 0.3, and 0.5 wt.% and the samples were labeled as PP + 0% Lignin, PP + 0.1% Lignin, PP + 0.3% Lignin, and PP + 0.5% Lignin, respectively.

RESULT AND DISCUSSION

Surface morphology of the materials was observed with SEM as displayed by Figure 1 and 2, with scaling of 100 and 500 μm , respectively. Figure 1a displayed PP + 0% lignin sample, while Figure 1b pictured PP + 0.3% lignin, with black color attributed to PP, gray color acts as indicator for cement, and darker black color assigned to lignin. It was revealed that prior to lignin addition, the mix of PP and sand, Figure 1a, did not mix well which can be seen by different colors on the surface. Consequently, the addition of lignin did not improve the mixture quality of the materials as shown by Figure 1b. Previous studies by Miao et al. [11] and Frasca et al. [12] stated that further processing or conditioning of the materials is needed to really mix and merge the aggregate and aggregate/lignin composite, such as suitable solvent to dissolve the lignin, mixing temperature, and mixing speed. That fact was further confirmed by Ye et al. [13] and Terzopoulou et al. [14] studies which concluded that mixing speed, mixing time, and mixing temperature were directly related to the rate of aggregate and aggregate/lignin dispersion.

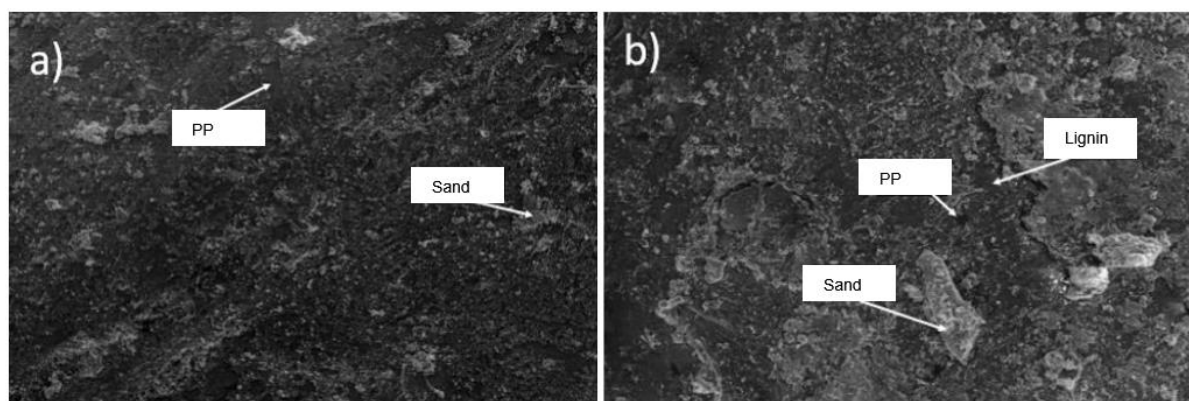


Figure 1. FE-SEM result (a) PP + 0% Lignin (b) PP + 0.3% Lignin with 100 μm scale

This fact was further established when observed on higher scale, 500 μm , on Figure 2. Figure 2a displayed the surface of mixed materials with sand and cement as matrix and PP as coarse aggregate filler. It can be seen that there is clear parting line between matrix and filler. Moreover, Figure 2b with the addition of lignin exhibited similar parting line between PP and the matrix. Those parting lines are gaps between the mixed materials which indicated that the mixed materials did not enhance each other's properties even with the addition of lignin. Compared to other studies about PP

and lignin composite [3], [15], [16], [17], this study showed different results in the mixing of PP and lignin. On the other studies, PP and lignin mix enhanced each other's properties which then improved the function of the material in their respective studies.

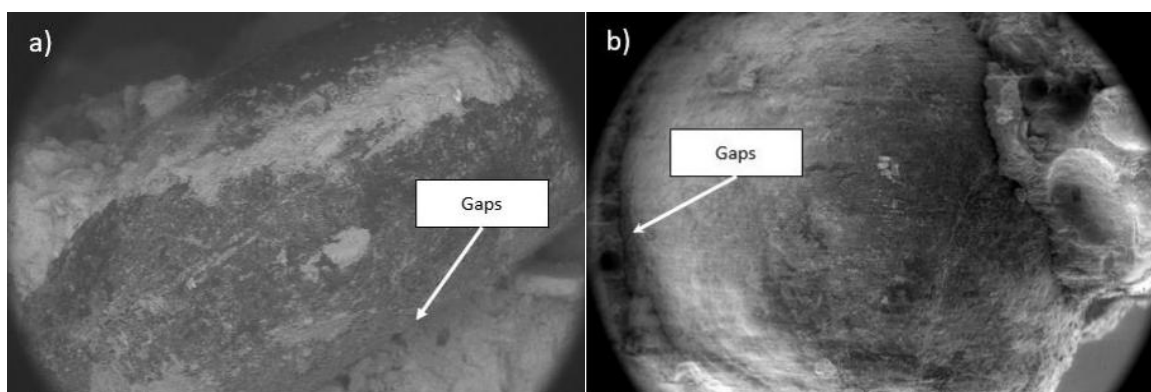


Figure 2. FE-SEM result (a) PP + 0% Lignin (b) PP + 0.3% Lignin with 500 μm scale

Further investigation using FTIR was carried out to examine existing chemical bonds and probability of new bonds in the samples as shown by Figure 3.

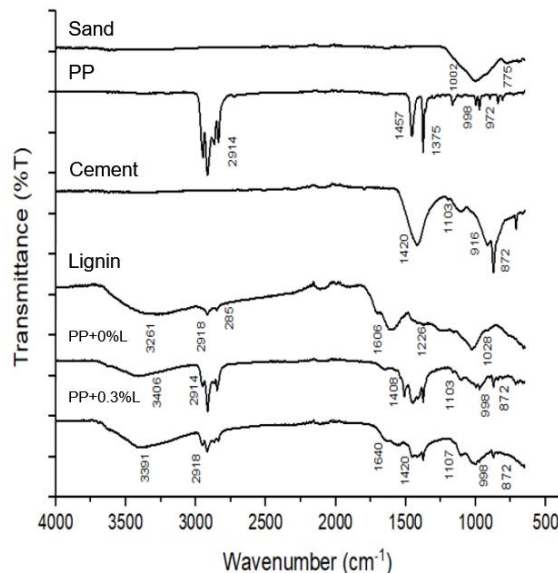


Figure 3. FTIR Graph of Each Component and PP-Lignin Mix

Observed peak at 1002 cm^{-1} is attributed to stretching vibration of Si-O-Si functional group, while low intensity spectra at 775 cm^{-1} is assigned to rocking vibration of Si-O group in sand [18]. Peak at 2914 cm^{-1} represents stretching vibration of C-H functional groups in the polymer parent chain. Spectra at 1457 and 972 cm^{-1} indicate the bending and rocking vibration of C-H in PP chain, respectively. Observed spectra at 1375 and 998 cm^{-1} are attributed to the bending and rocking vibration of C-H, respectively [1]. Moreover, peaks at 1420 and 872 cm^{-1} are assigned to vibration of CO₃²⁻ group from CaCO₃. Spectra at 1130 cm^{-1} is attributed to SO₄²⁻ bond in CaSO₄. Peak at 916 cm^{-1} represents asymmetric stretching vibration of Si-O group [19].

Broad spectra at 3261 cm^{-1} corresponds to stretching vibration of -OH from phenolic functional group in monolignol lignin. Peak at 2918 cm^{-1} indicates stretching vibration of C-H bond in aromatic ring. Spectra at 2851 cm^{-1} is attributed to stretching vibration of C-H bond [1]. Peak at 1606 cm^{-1} is assigned to stretching vibration of C=C functional group from aromatic ring. Observed peak at 1226 cm^{-1} represents stretching vibration of C-O bond in aliphatic ring from monolignol syringyl and guaiacyl. Spectra at 1028 cm^{-1} indicates stretching vibration of C-O-C group from ether in aliphatic ring [20].

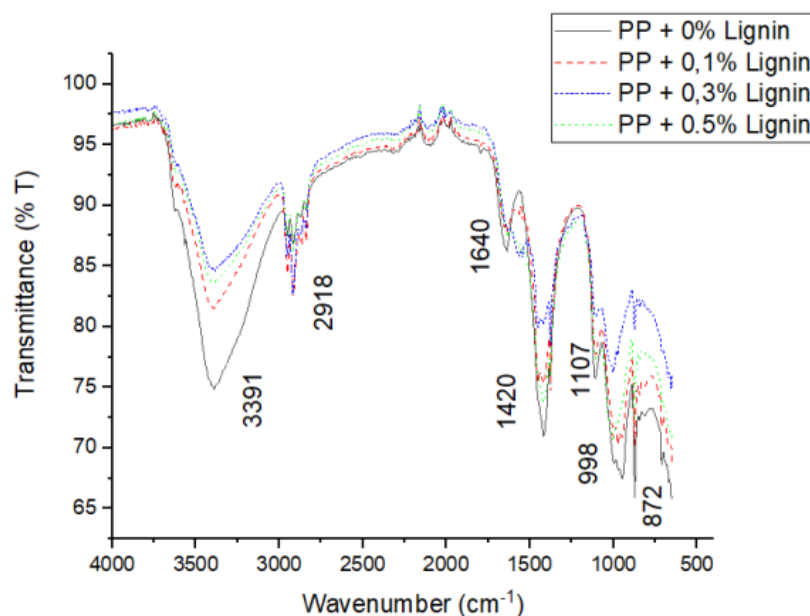


Figure 4. FTIR Graph of Mixed PP and Lignin Variations

After the components were dry mixed together, some functional groups and chemical bonds in the mix were identified using FTIR. Broad spectra at 3391 cm^{-1} represents -OH groups from water when hydration process occurred during the mixing process. Peak at 2918 cm^{-1} indicates C-H group from alkane [1]. Spectra at 1640 cm^{-1} is attributed to C=C group in aromatic ring [20]. Peaks at 1420 and 872 cm^{-1} are assigned to CO₃²⁻ group from CaCO₃, while spectra at 1107 cm^{-1} is ascribed to SO₄²⁻ group in CaSO₄ [19]. Spectra at 998 and 872 cm^{-1} are credited to C-H from PP [1].

Various lignin additions effect on the mix was investigated with FTIR as displayed by Figure 4. As the concentration of lignin increased, there was only an increase in the intensity of the spectra without newly formed bonds. The observed peaks without the addition of lignin, PP + 0% Lignin, also displayed similar peaks even after lignin addition which indicated that lignin did not contribute as bridging component or binder in the mixed material. This result explained further the SEM result where there are clear parting lines between the matrix and filler which indicates that there were no uniting bonds in the mixed samples that enhance the obtained materials and each materials act as stand alone.

Wettability test results conducted on each material resulted in surface energy value as shown by Table 2.

Table 2. Surface Energy of Each Component at 25°C

No.	Component	Surface Energy (mN/m)
1.	Lignin	24.47
2.	PP	29.01

Surface energy of materials indicates the adhesive ability of materials, their ability to form bonds, and their nature to water [21]. Solids with surface tension >45 mN/m is hydrophilic, while those <45 mN/m are hydrophobic [22]. Based on the results on Table 2, both lignin and PP are hydrophobic materials and thus compatible with each other. However, due to lower surface energy of lignin, 24.47 mN/m, compared to PP, 29.01 mN/m. The adhesion of lignin is significantly minimal thus could not act as an enhancing component in the resulting composite, which is shown by SEM and FTIR result of this study.

In contrast, other studies have shown that PP and lignin were compatible with each other and can enhance each other properties [3], [15], [16], [17]. Different results between this study and the others might be related to different materials used during the study, this study used virgin plastic and pure lignin extracted from paper industry waste. Virgin plastic is more inert compared to plastic waste which might contribute to the higher surface energy of PP [3]. Additionally, the pure lignin was not further processed to increase its surface energy which also contributed to the success in forming plastic-lignin composite [11], [21], [23], [24], [25]. Moreover, water as solvent might also contribute to the mixture of quality due to degree of insolubility of the materials used in this study [11]. This conclusion is in line with previous studies by Miao et al. [11] and Frasca et al. [12] which processed or conditioned the aggregate and lignin in their studies by classifying the type of lignin in the mix to understand the chemical specification, utilized alternate solvent other than water, and mixed their samples with specific mixing temperature. Utilization of lignin in construction material holds immense potential in order to reduce carbon emissions from conventional construction material processing [11], [13], [14].

CONCLUSION

Plastic waste has become an alarming issue, which motivated several studies to utilize it as construction material to reduce emissions. Lignin as binder material has potential to enhance the obtained construction material and further reduce emissions from conventional construction material processing. This fact is supported by the similar surface energy

between lignin and PP as hydrophobic material with 24.47 and 29.01 mN/m, respectively. However, the surface energy value becomes a challenge during the mixing process due to lignin lower surface tension, which means that lignin unable to act as a binder or bridging agent between PP as filler and sand and cement as matrix in the mixed materials. Thus, it was unable to enhance the properties of paving block which also supported by SEM and FTIR analysis. However, other studies have shown that these two materials are compatible and able to enhance each other's properties. The different results in this study were due to the different processing and conditioning of the materials which affected the mixture quality compared to other studies. Thus, further research by actively altering material properties, such as determine suitable processing of aggregate and lignin, dictate the mixing speed, mixing temperature, and mixing time of the materials, and utilize alternative solvents other than water are believed to be a solution to develop this study into a successful result. Addition of mechanical performance testing is advised to be performed when similar study showed successful result.

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