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The Effect of the Ratio of Methyl Metacrylate and Styrene on the Grafting Copolymerization of Natural Rubber Latex as an Emulsion Paint Binder

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Abstract
The emulsion paint industry still relies heavily on conventional synthetic binders, such as acrylic and vinyl resins, which are derived from petrochemicals and pose sustainability and environmental concerns. This study explores the use of natural rubber latex as an alternative, more environmentally friendly binder through a graft copolymerization process with styrene and methyl methacrylate monomers. The reaction was carried out with the addition of 2% sodium lauryl sulfate (SLS) surfactant and 2% benzoyl peroxide (BPO) initiator. The success of the graft was confirmed by the appearance of distinctive peaks in the FTIR spectrum and the increase in the mechanical strength of the resulting binder. Application to emulsion paints demonstrated better performance than pure latex, particularly in adhesion, scrubbing resistance, and water resistance. In addition to improved technical performance, the use of natural rubber also offers advantages over synthetic acrylic binders, including local raw material availability, lower production costs, and a smaller environmental footprint. These results demonstrate that modified natural rubber latex has potential as an environmentally friendly binder for paint, providing a practical basis for the development of sustainable emulsion paint formulations that can support the competitiveness and independence of the national paint industry.

INTRODUCTION

Rubber (Hevea Brasiliensis Mull) is one of Indonesia's leading plantation commodities, cultivated as an annual crop, primarily derived from the sap of the tree trunks obtained through tapping. Indonesia ranks first in the world in terms of rubber plantation area. Based on data from the Directorate General of Plantations, the total area of rubber plantations in Indonesia in 2020 reached 3,726,173 ha. Most of this area is managed by communities through smallholder plantation schemes (90.39%), followed by large private companies (6.04%), and large state-owned companies (3.57%). In Indonesia, there are 301 large rubber companies, consisting of 100 state-owned companies and 201 private companies. Natural rubber has several advantages, one of which is its high tensile strength and excellent elongation, making it a primary ingredient in the manufacture of products such as tires, gloves, and various other rubber-based products. These superior properties can be enhanced through modifications, one of which is by converting it into Natural Rubber Latex (NRL). NRL functions as a compatibility agent, reactive plasticizer, adhesive, and coating, offering the advantages of elasticity, durability, and the ability to return to its original shape more effectively than synthetic latex (Ibrahim et al., 2014; Nawamawat et al., 2011).

Processing NRL into final products requires deproteinization to reduce the protein content, making modification easier. Standard modification methods include epoxidation, vulcanization, depolymerization, and monomer grafting. One application is as a binder in water-based emulsion paints, which has the potential to serve as an alternative to synthetic latex, thereby reducing import dependence and increasing sustainability (Ibrahim et al., 2022). Emulsion paint is a mixture of water and oil, equipped with an emulsifier, consisting of a binder, filler, pigment, solvent, and additives (Kent, 2012). Binders play a crucial role in binding pigments and ensuring adhesion to the surface, typically made from natural or synthetic resins such as alkyd, acrylic, epoxy, and polyurethane (Anugrah, 2009). Graft copolymerization between natural rubber and monomers, especially Styrene, has been widely studied because it enables the improvement of rubber's mechanical and thermal properties due to the presence of aromatic structures in its molecules (Dung et al., 2017). Polystyrene, resulting from styrene polymerization, offers advantages such as high transparency, good viscosity, ease of processing, and good dielectric properties (Zhang et al., 2012). Therefore, the natural rubber latex developed uses methyl methacrylate and Styrene as binders through a graft copolymerization method, where styrene and methyl methacrylate (MMA) are grafted into the natural rubber latex matrix. Styrene and MMA were chosen because they have complementary properties, with Styrene providing mechanical strength and heat resistance (Dung et al., 2017). While MMA can increase adhesion, hardness, and water resistance, it can also produce a more homogeneous and stable binder (Ibrahim et al., 2022; Ibrahim et al., 2022; Zhang et al., 2014). By mixing Styrene - MMA binder in natural rubber latex, it is hoped that it can improve the mechanical properties of natural rubber latex and can be applied as a binder for emulsion paint applications.

Furthermore, the evaluation of the performance of grafted NRL binders in standardized emulsion paint formulations with controlled PVC/CPVC parameters remains limited in the literature. To address this limitation, this study was systematically conducted to analyze the effect of varying MMA/Styrene composition ratios on key application performance indicators, specifically washability resistance and drying time, under standardized conditions. This work aims to fill this knowledge gap and provide a scientific basis for the development of more optimal natural rubber-based emulsion paint binders.

Based on the background and research gaps above, this study aims to: (1) Assess the effect of variations in the ratio of MMA and Styrene (10:90, 15:85, 20:80, 25:75, 30:70) on the degree of grafting in natural rubber latex (NRL); (2) Evaluate the effect of the MMA/Styrene ratio on the washability resistance properties of emulsion paint produced according to the ASTM D4828 standard; (3) Analyze the effect of the MMA/Styrene composition on the touch drying time of emulsion paint based on SNI 3564:2009 and ASTM D1640; and (4) Determine the optimal composition of MMA/Styrene for NRL-based emulsion paint binder applications by considering the balance between the degree of grafting, washability resistance, and drying time.

METHOD

Materials

This research utilized deproteinized natural rubber latex (NRL) obtained from rubber plantations in Kampar Regency, Riau, as the base material. The subsequent graft copolymerization process required specific reagents for the synthesis of the binder, namely: Methyl Methacrylate (MMA) and Styrene as the primary monomers; Sodium Lauryl Sulfate

(SLS) as the surfactant; and Benzoyl Peroxide as the initiator. All chemical reagents used for the grafting process (MMA, Styrene, SLS, and Benzoyl Peroxide) were obtained from a local distributor. Although previous studies have tested MMA and Styrene grafting on NRL at various ratios, this study focuses specifically on the effect of varying MMA/Styrene ratios with a dominant Styrene composition (70-90%) on the final paint properties, using a grafting temperature of 60^0C

Methods

This study is divided into two stages: the graft copolymerization stage and the paint formulation stage. The grafting copolymerization stage is carried out using a magnetic stirrer equipped with speed, temperature, and stirring controls. Natural rubber latex is mixed with a combination of Methyl Methacrylate (MMA) and Styrene at a ratio of 85:15 (w/w), with varying MMA-to-styrene compositions of 10:90, 15:85, 20:80, 25:75, and 30:70 (w/w). Sodium Lauryl Sulphate (SLS) surfactant is then added to the mixture as an emulsifier at 2% of the total sample weight. Next, Benzoyl Peroxide initiator is added at a dose of 2% of the total sample weight to accelerate free radical formation, thus facilitating the grafting process in NRL. The process takes place at a constant temperature of 60°C with a stirring speed of 350 rpm for 6 hours to produce modified natural rubber latex.

The next stage is paint formulation. The paint formulation consists of water, hydroxyethyl cellulose, ultramarine blue, alkylphenol ethoxylate, TiO_2, CaO, $CaCO_3$, polypropylene glycol, eastment, dodecylbenzene sulfonate, and modified natural rubber latex. The mixing process is carried out using a mechanical stirrer for 30 minutes. After that, the emulsion paint binder, in the form of modified natural rubber latex, is added in a proportion of 4% of the total paint weight. In the final stage, additives and perfumes, such as ester and dodecylbenzene sulfonate, are added to the paint mixture. Then the paint is ready to be applied, and its physical properties are tested (degree of grafting, washability resistance, and touch drying).

Grafting Degree Analysis

The grafting degree indicates the percentage of monomer mass successfully grafted onto the polymer backbone. This value is determined by oven-drying the sample to constant weight, then refluxing with toluene to dissolve the ungrafted MMA (Bhattacharya & Misra, 2004). This step is followed by the precipitation of the sample using excess acetone, and then refluxing with xylene to dissolve the ungrafted Styrene. Next, the ratio of the mass of the grafted monomer to the mass of the polymer is calculated through acid-base titration, thus obtaining the degree of grafting in the sample (Dung et al., 2017; Eddiyanto, 2007; Ibrahim et al., 2022).

Washability Resistance Analysis

Washability resistance analysis is performed to assess the resistance of emulsion paint to water-induced abrasion. The higher the resistance, the better the quality of the paint. The test is performed by applying a layer of paint to a waterproof paper surface, then drying it for 24 hours at room temperature. Next, the washability of the paint layer is tested using a BGD 526 Wet Abrasion Scrub Tester according to ASTM D4828 standards.

Touch Drying Time Analysis

Touch drying time analysis was performed to determine the duration required for emulsion paint to reach a touch-dry state. Measurements were made using the Set-to-touch drying method with a glass panel, as per ASTM D1640 standards, with test time references based on SNI 3564:2009.

RESULT AND DISCUSSION

Grafting Degree

Modification of natural rubber latex (NRL) as an emulsion paint binder was carried out through a grafting method using Methyl Methacrylate (MMA) and Styrene monomers. The success of the grafting process was evaluated by comparing the sample mass before and after grafting to calculate the grafting yield, which is expressed as a percentage and known as the grafting degree (Eddiyanto, 2007). To identify functional groups in the binder and verify the success of grafting, analysis was performed using Fourier Transform Infrared Spectroscopy (FTIR), a method for compound characterization based on interaction with electromagnetic radiation (Alauhdin et al., 2001). The FTIR spectrum results of the NRL binder are shown in Figure 1.

The NRL grafting process with MMA and Styrene is said to be successful if the FTIR spectrum of the NRL binder shows peaks indicating the carbonyl group (C=O) of MMA and the benzene ring (C=C) of Styrene. The isoprene group is characterized by a C-H bond at wavenumbers of 3,000–3,500 cm⁻¹. In MMA, two typical groups can be detected, namely C=O (1765–1720 cm⁻¹) and C-O-C (1,180–1,290 cm⁻¹). Meanwhile, in Styrene, the typical groups include C=C (1,400–1,700 cm⁻¹) and C-H (2,700–3,000 cm⁻¹). (Suhardjo et al., 2011). Based on Figure 1, several peaks were identified in the FTIR spectrum indicating the presence of functional groups in the NRL binder, including peaks at 3,376.45 cm⁻¹ (C-H from isoprene), 2,954.99 cm⁻¹ (C-H from Styrene), 1,446.08 cm⁻¹ (C=C from Styrene), and 1,238 cm⁻¹ (C-O-C from MMA). The success of grafting on natural rubber is indicated by the presence of C=O groups from MMA and C=C groups from Styrene (Suhardjo et al., 2011).

The FTIR analysis results of the emulsion paint binder showed that Styrene was successfully grafted onto NRL, indicated by the appearance of aromatic C=C groups at 1,446.08 cm⁻¹. The C-O-C groups of MMA were detected at 1,238 cm⁻¹, but the carbonyl (C=O) groups typical of MMA that should appear in the range of 1,720-1,730 cm⁻¹ were not detected in the FTIR spectrum. This indicates that MMA was not chemically grafted onto the NRL backbone at the grafting temperature of 60°C used in this study. This finding is consistent with the literature, which states that MMA requires a higher grafting temperature (approximately 90°C) to achieve effective grafting (Siregar et al., 2015). Therefore, it can be concluded that in this study, only Styrene monomer was successfully chemically grafted onto the modified NRL as an emulsion paint binder. At the same time, MMA likely remained in the system as a physical mixture or formed an interpenetrating polymer network (IPN) without covalent chemical bonds with the NRL backbone.

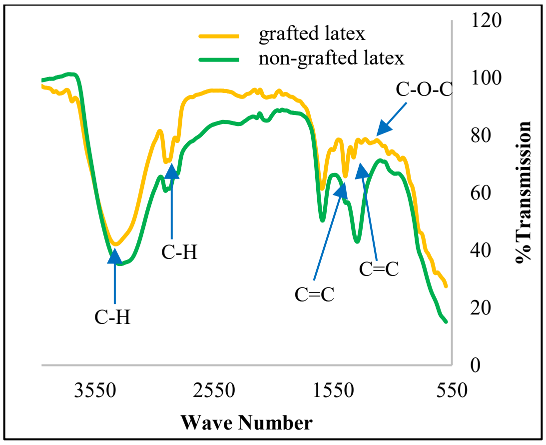

Figure 1. FTIR spectra for NRL with grafting and without grafting

The success rate of NRL grafting as an emulsion paint binder can be evaluated through the grafting degree percentage shown in Figure 2. The highest value was obtained at a 10:90 MMA and Styrene composition, which was 48%, while the lowest value was recorded at a 30:70 ratio with a percentage of 26.5%. These results indicate that variations in the MMA and Styrene ratio affect the grafting degree. The higher the proportion of Styrene in the mixture, the greater the grafting degree value produced. The use of Styrene as a comonomer in natural rubber modification can provide higher grafting efficiency compared to other monomers (Angnanon et al., 2011).

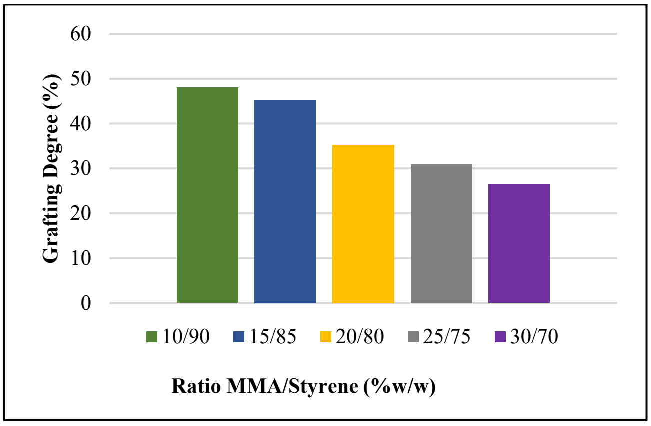

Figure 2. NRL Binder Grafting Degree

The results of the study showed that increasing styrene levels were directly proportional to the degree of grafting. This occurs because higher amounts of Styrene influence the deactivation and transfer of radical molecular chains, where styrene molecules detach from the main chain and then graft onto the NRL chain (Dung et al., 2017). The study also confirmed that the amount of Styrene used as the NRL grafting monomer had a significant effect on the degree of grafting (Dung et al., 2017). MMA not grafting to NRL is thought to be due to the grafting temperature. The grafting temperature used, 60 °C, was chosen based on research. Susmanto et al. (2021) showed that this temperature is the optimal condition for grafting styrene monomers onto natural rubber latex, with the highest grafting efficiency reaching 87.98%. At this temperature, the initiator benzoyl peroxide (BPO) decomposes at a rate suitable for the formation of free radicals that activate Styrene grafting. However, a temperature of 60°C is not optimal for MMA grafting, as Siregar et al. (2015) suggest that temperatures around 90°C are required to achieve effective grafting. This limitation was identified in this study and is an important consideration for further development, so the primary focus of grafting optimization is on the styrene monomer, with MMA acting as a comonomer that contributes to the binder properties through mechanisms other than chemical grafting. The choice of 60°C as the temperature is also based on considerations of the stability of natural rubber latex, where heating above 60°C can cause premature coagulation or damage to the latex structure. (Susmanto et al., 2019). Temperatures above this limit can damage the cross-linking of natural rubber molecules, thus inhibiting the perfection of the grafting process. Therefore, this study can be concluded that only Styrene monomer was successfully grafted onto the modified NRL as an emulsion paint binder.

Although MMA was not chemically grafted onto the NRL backbone (as evidenced by the absence of the C=O peak at 1,720-1,730 cm^{-1}), the presence of MMA in the emulsion system can still affect the physicochemical properties of the binder through several mechanisms: (1) the formation of an interpenetrating polymer network (IPN) where the MMA polymer chains are physically intertwined with the NRL-g-Styrene chains without chemical bonds; (2) homopolymerization of MMA in the emulsion phase resulting in a polymethyl methacrylate (PMMA) phase dispersed within the NRL matrix; and (3) modification of the morphological structure of latex particles that affects the paint film formation process. Thus, the variation in the MMA/Styrene ratio in this study primarily reflects the variation in the degree of Styrene grafting and the relative proportion between NRL-g-Styrene and the ungrafted MMA component in the binder system.

Washability Resistance

Based on Figure 3, the highest washability resistance value was obtained with a 10:90 MMA-to-styrene ratio, reaching 20 cycles, while the lowest value was found with a 30:70 ratio, reaching 14 cycles. These test results indicate that ungrafted NRL has a lower washability resistance value than NRL grafted with Styrene and MMA. This suggests that the grafting process has a significant impact on the resistance of emulsion paint to water abrasion.

The observed increase in washability resistance is directly proportional to the increase in styrene content and the decrease in MMA content (Arjmandi et al., 2024). This relationship is driven by the monomer composition of the binder, which plays a crucial role in determining its hydrophilicity. Styrene is water-insoluble (hydrophobic), while ungrafted MMA is slightly soluble in water (more hydrophilic) (Zainudin et al., 2021, Nguyen et al., 2020). Since only Styrene was grafted onto NRL in this study, an increase in the Styrene ratio (a corresponding decrease in the MMA ratio) indicates an increase in the number of hydrophobic segments chemically bonded to the NRL backbone (Wildeson et al., 2008).. This chemical bonding enhances the binder's resistance to water penetration. In contrast, increasing the content of ungrafted MMA tends to decrease the abrasion resistance of the paint due to its more hydrophilic nature and its presence as a separate phase that is less integrated in the binder structure. The quality of emulsion paint is highly dependent on the binder's characteristics, which must include being waterproof (Nguyen et al., 2020).

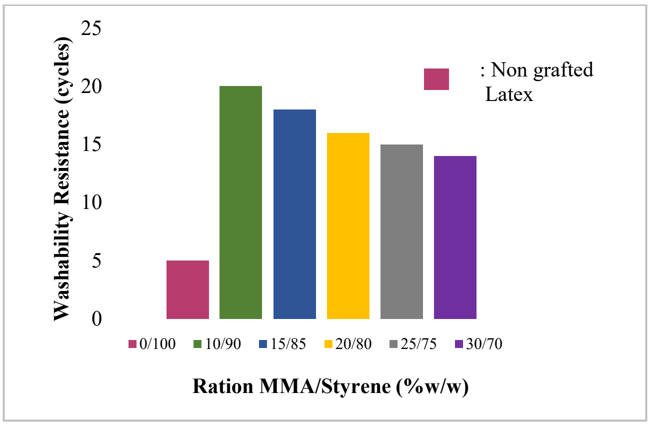

Figure 3. Washability Resistance Test Results of Emulsion Paint with NRL Binder

In this context, NRL (Natural Rubber Latex) serves as a grafting medium, featuring unsaturated bonds that play a crucial role in determining the adhesion properties of natural rubber and its derivatives. Grafting of MMA and Styrene into NRL was carried out specifically to increase the adhesion of NRL as a binder, while also improving the resistance of emulsion paint to water (Zainudin et al., 2021).

Furthermore, the amount of binder used also affects film formation and resistance to water abrasion. After the paint is applied, water from the dispersion evaporates, causing the polymer particles to become denser and coalesce to form a dense film layer without cavities (Arjmandi et al., 2024). This film formation process provides the paint with the strength to withstand abrasion throughout its service life. Consequently, emulsion paint with a good film layer will have high washability resistance (El-Wahab et al., 2019). Several studies also show that high binder content increases the paint's resistance to water abrasion in scrub and abrasion tests (Bahruddin et al., 2025). Washability resistance, the paint's ability to withstand abrasion caused by water, is a crucial factor affecting the quality of emulsion paint . (Lusiana & Cahyono, 2014). The higher the paint's resistance to water abrasion, the better the quality of the emulsion paint. This abrasion resistance is determined through scrub and abrasion tests, which aim to ensure the paint, can withstand the frictional force of water throughout its useful life (Glover, 2021)

Touch Dry Time

The test results shown in Figure 4 indicate that grafting MMA and Styrene onto the NRL binder affected the paint's drying time. Ungrafted NRL exhibited a longer drying time than grafted NRL. Specifically, a 10:90 MMA and Styrene composition produced the longest drying time, at 43 minutes and 12 seconds, while a 30:70 MMA and Styrene ratio provided a faster drying time, at 28 minutes and 2 seconds. Based on SNI 3564:2009, the maximum allowable touch drying time is 30 minutes. Therefore, the 30:70 MMA and Styrene composition met this quality standard and demonstrated exemplary performance in terms of paint drying.

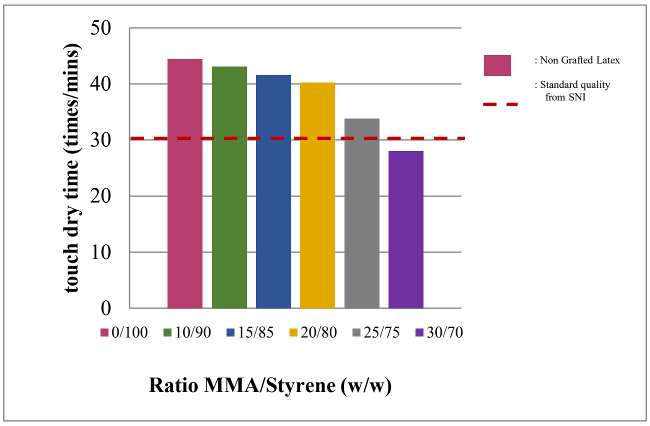

Figure 4. Touch Drying Time Test Results of Emulsion Paint with NRL Binder

The properties of each monomer can explain this difference in drying time. The core factor affecting touch dry time is the binder, as it plays a crucial role in forming the paint film. Styrene, being an oil-like liquid, takes longer to evaporate, while MMA has properties that tend to accelerate film formation, thus speeding up the paint drying process (Angnanon et al., 2011; Butler et al., 2003). Increasing the NRL content in the binder tends to prolong the touch-dry time. This is due to the nature of modified NRL, which has a relatively high water content retained within its polymer chains. This water content inhibits the water evaporation process, thus slowing the paint drying time (Ibrahim et al., 2022). Thus, increasing the MMA content and decreasing the styrene content in the grafted NRL binder consistently accelerated the touch drying time of emulsion paint (Ibrahim et al., 2022; Ibrahim et al., 2022)

These findings are corroborated by another study that also tested the touch dry time of wall paint with a mixture of NRL binders grafted with different ratios of MMA and Styrene (Ibrahim et al., 2022). That study revealed that the lower the styrene content and the higher the MMA content, the faster the paint drying process occurs. In their test, the composition of MMA and Styrene at a ratio of 65:35 produced the longest drying time, namely 104 minutes, while the ratio of 80:20 gave the fastest drying time, namely 92 minutes. These findings confirm that changes in the monomer composition of the binder can affect the drying speed of the paint, which is important for achieving optimal performance in wall paint applications. The faster the paint dries, the lower the risk of contamination that could damage the paint layer. However, testing the touch-dry time is subjective and highly dependent on the measurement method used, so there is potential for human error in determining accurate results. (McNally, 2011).

CONCLUSION

Modification of natural rubber using a grafting method with methyl methacrylate (MMA) and Styrene at 60°C successfully produced a binder suitable for application in emulsion paints1. Based on FTIR analysis, only styrene monomer successfully chemically grafted onto the NRL backbone, while MMA did not graft under the temperature conditions used. This finding is consistent with the literature, which shows that Styrene exhibits higher grafting efficiency at 60°C. Variations in the composition of MMA and Styrene in the emulsion paint binder affected various physical properties measured in this study. The primary impacts observed were that increasing the grafted Styrene content improved the paint's washability resistance, likely due to the hydrophobic nature of the polystyrene segments bonded to the NRL backbone. Conversely, increasing the MMA content consistently accelerated the touch-dry time due to faster evaporation and more efficient film formation in systems with lower styrene content.

These findings indicate a critical trade-off between the paint's key performance metrics. Overall, increasing the styrene content in the emulsion binder composition improves washability resistance, but also slows down the drying time of the paint. Conversely, increasing the MMA content accelerates the touch-dry time, but can also decrease washability resistance. This balance suggests that the resulting product has potential for economy-grade interior paint applications or exterior paints with lower washability requirements. Therefore, this trade-off between durability and drying time must be carefully considered in designing optimal natural rubber-based emulsion paint binders.

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