

PRECIPITATED CALCIUM CARBONATE/LITHOPONE NANOPARTICLES AS SUBSTITUTION OF TiO₂ PIGMENT FOR MATTE-TYPE WATER-BASED PAINT

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ABSTRACT

This study optimized the Precipitated Calcium Carbonate (PCC) and lithopone mixture (50:50) as substitution pigments of titanium dioxide (TiO₂) in matt-type water-based paint. Matt-type water-based paint has been made in two steps; (1) mill base paste preparation and (2) water-based paint production (layout process). Pigment characterization showed that PCC/lithopone mixture produced a white solid of 95.57%, crystallinity degree of 91.5%, and a diameter small size particle of 198.29 nm. Application of PCC/lithopone mixture as pigment into matt-type water-based paints has resulted in good dispersibility and hiding power while exhibiting the same whiteness quality as TiO₂ and glossy better than TiO₂.

Keywords: PCC; Lithopone; TiO₂; Pigment; Water-Based Paint.

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INTRODUCTION

In the paint industry, titanium dioxide (TiO₂) is one of the best white pigments due to its better refractive index, achromatic force, dispersion, and whiteness. However, the production of TiO₂ has been limited due to the acid waste in industrial production, the shortage of titanium resources, and the high price of titanium dioxide. Therefore, finding alternative material sources for pigment function is crucial.¹⁻³ Calcium carbonate (CaCO₃) is an abundant inorganic resource with high whiteness, low hardness, and small energy consumption. CaCO₃ is often used as fillers in composites to improve the composite's physical and mechanical properties.^{4,5} Various researchers have attempted to combine CaCO₃ with TiO₂ through a chemical coating method to form the CaCO₃-TiO₂ pigment. The CaCO₃-TiO₂ pigment has a strong ultraviolet absorption capacity. However, optimal results have not been obtained regarding calcium sources from converting CaCO₃ to calcium oxide (CaO) and precipitated calcium carbonate (PCC).⁶⁻¹⁰ Research on the use of inorganic minerals as pigments and their combination with other minerals such as CaCO₃ in painting applications has been carried out. Karakaş *et al.*,¹¹ studied the stabilization mechanism of several pigments by analyzing the interaction and dispersion between sodium salts and polyacrylic acid with various types of minerals, both single minerals such as TiO₂, calcined kaolin (C.Kaolin), ground calcium carbonate (GCC), or combined minerals such as TiO₂-GCC, TiO₂-C Kaolin, and TiO₂-GCC-C.Kaolin. The results suggest that using the combined mineral is feasible to obtain the desired paint properties. Chen *et al.*,⁶ have developed GCC-TiO₂ (GCTD) with a two-stage coating method: (1) coating GCC with SiO₂, and (2) GCC coated with SiO₂ was re-coated with TiO₂. The GCTD obtained could be used as high-quality coating material. However, its whiteness and brightness are reduced, but its oil absorption value increases compared to GCC.⁶ Karakaş *et al.*,¹ have analyzed the effect of PCC additions on a water-based paint with various pigment volume concentrations (PVC), which indicates that PCC was successfully used to replace the role of TiO₂ pigment in paint formulations. Alvarez *et al.*,¹² studied the effect of the amount and type of functional

monomer on the dispersion of CaCO_3 fillers in acrylic latex coating. One of the interesting and popular minerals to develop is lithopone due to its low production cost and greater coverage. Lithopone is an inorganic compound mixture of zinc sulfide (ZnS) 68-70% and barium sulfate (BaSO_4) 28-30% obtained from the reaction between barium sulfide and zinc sulfate. Lithopone is a white pigment widely used as a basic colorant for water-based paints, inks, plastics, rubber, and various cosmetic products. Most of the role of lithopone as a pigment has been replaced by TiO_2 .¹³⁻¹⁶ Cao *et al.*,¹⁴ had modified pigments sepiolite and lithopone, where the particle size of the modified pigment was comparable with the original pigment but had a tighter diameter distribution and higher homogeneity than the original pigment. It prompted the researchers to mix PCC with lithopone to form a PCC/lithopone mixture which can be used as an alternative to the pigment TiO_2 , which has good ability in terms of dispersibility, whiteness, gloss, and hiding power to the binder/resin. The PCC/lithopone mixture pigment is used as the main pigment for coloring and hiding power in paint manufacturing. Pigments are divided into the main pigment and the extender pigment, which help strengthen the main pigment, increase the strength of the paint layer's viscosity, and prevent sedimentation. In certain types, the extender pigment is used to reduce the gloss of the paint (matte type). A binder is needed to bind the pigment particles with the paint layer so that the paint can form a thin layer and stick to the substrate surface. Water is used as a solvent to dissolve powder materials and disperse pigments, binders, and other additives to allow flow. Additives are used to improve performance and are commonly used in very small amounts. The existence of additives is vital in the final paint result, especially the flow and leveling ability of the paint. The additives used in the manufacture of paints are thickeners, dispersing, coalescing, wetting agents, biocide, ethylene glycol, anti-foaming, and others.^{1,11} The study aims to characterize the nanoparticles of PCC/lithopone mixture as a substitute for TiO_2 in matt-type water-based paint. It is important in the research area of industrial technology, especially the paint industry, which contributes to finding a substitute for TiO_2 with better capabilities.

EXPERIMENTAL

Material

Precipitated calcium carbonate (Precarb 100) was supplied by "Schaefer Kalk" company, Kuala Lumpur, Malaysia. Lithopone (B311) was received from "Anhui Union Titanium Enterprise" company, Shanghai, China. Rutile titanium dioxide (SR-2377) was supplied by "Shandong Dongjia Group" company, Shandong, China. Calcium carbonate (Omyacarb-6 GD) as a filler was obtained from "Camco Omya Indonesia" Company, Jakarta, Indonesia. Styrene acrylic (C-817) binder and Dispersing agent (Chemtex D-710) were received from "The Inawan Chemtex Sukses Abadi" Company, Jakarta, Indonesia. Thickener (Natrosol 250 HBR) was received from "Ashland Global Holding" company, Covington, USA. The wetting agent (Tergitol NP-10) was obtained from "Dow Chemical" Company, Michigan, USA. Anti-foaming agent (Nopco NXZ) was obtained from the "San Chemicals" Company, Osaka, Japan. An anti-microbial agent (Anticide RS) was received from "SpecialChem" Paris, France. Ethylene glycol (EG) was received from "Polychem Indonesia" Company, Jakarta, Indonesia. Coalescent agent (Texanol CS-12) was obtained from "Guangzhou Donghuang Chemical Technology" company, Guangzhou, China.

Preparation of Pigment Mixture

700 g of the PCC and lithopone mixture materials with a composition (50:50) were put into a planetary mixer (KNS-60 LB) and then mixed by rotating in all horizontal and vertical directions for 15 min. The results obtained are the PCC/lithopone mixture.

Characterization of Pigment

The pigments characterized were TiO_2 , PCC, lithopone, and PCC/lithopone mixture. Morphological characterization of the pigment was carried out using Field Emission Scanning Electron Microscope (SEM) - FEI Inspect F50, the chemical composition of the pigment using X-Ray Fluorescence (XRF) spectrometer - PANalytical Epsilon 3, and the crystal structure of the pigment using X-Ray Diffraction (XRD) - PANalytical X'Pert PRO MPD.

Preparation of Mill Base Pasta

Water, thickener, dispersing, wetting agent, biocide, ethylene glycol, and anti-foaming were added successively according to Table-1 into a high-speed mixer, mixed at 2000 rpm for 60 min using a

zirconium ball mill with the composition ratio between the mill base pasta ingredients and zirconium ball mill is (60:40), filtered using a sieve (200 mesh), and cooled at room temperature. The results obtained are in the form of mill base paste.

Application of Nanoparticles as a Pigment in Matt-Type Water-Based Paint

Mill base paste, pigment (TiO_2 , PCC, lithopone, and PCC/lithopone mixture), CaCO_3 , water, and coalescing agent were added successively according to Table-1 into a high-speed mixer, stirred at 800 rpm for 30 min, filtered through a sieve (200 mesh), observed the dispersibility of the pigment. Next, the styrene-acrylic binder was added and stirred again at 100 rpm for 15 min.^{1,11,17} The results obtained were a matt-type water-based paint with various pigments, referred to as the R sample.

Table-1: A Matt-Type Water-Based Paint Recipe with Various Pigments

Ingredient	R1 (g)	R2 (g)	R3 (g)	R4 (g)
Water	326.06	326.06	326.06	326.06
Thickener	6.66	6.66	6.66	6.66
Dispersing	1.77	1.77	1.77	1.77
Wetting agent	0.69	0.69	0.69	0.69
Antimicrobial agent	5.43	5.43	5.43	5.43
Ethylene glycol	16.28	16.28	16.28	16.28
Anti-foaming	2.83	2.83	2.83	2.83
Pigment : TiO_2	73.27	-	-	-
Pigment : PCC	-	73.27	-	36.63
Pigment : Lithopone	-	-	73.27	36.63
CaCO_3	439.68	439.68	439.68	439.68
Water	44.62	44.62	44.62	44.62
Coalescing agent	2.72	2.72	2.72	2.72
C-817 binder	80.00	80.00	80.00	80.00
Total	1000.00	1000.00	1000.00	1000.00

Note: R1 = Paint with pigment of TiO_2 (100); R2 = PCC (100), R3 = Lithopone (100), and R4 = PCC/Lithopone mixture (50:50)

Hiding Power, Whiteness, and Gloss Testing of Matt-Type Water-Based Paint

Matt-type water-based paint was applied to the surface of the hiding power panel using a bar coater (100 μ), then left for 24 h. The results obtained were observed visually for hiding power, then whiteness was measured with a portable whiteness meter (BGD 585), and gloss was measured with a gloss meter (BGD 516).

RESULTS AND DISCUSSION

XRF of Pigments

Based on the XRF analysis result of TiO_2 , PCC, Lithopone, and PCC/Lithopone mixture pigments shown in Table-2, rutile has a TiO_2 concentration of 94.57%, PCC has a CaO concentration of 96.49%, and lithopone has a concentration of 91.81%, which consists of BaO, ZnO, and SO_3 compounds. The PCC/Lithopone mixture (50:50) has a concentration of 95.57%, which consists of CaO, BaO, ZnO, and SO_3 . These results indicate that mixing PCC-lithopone at 50:50 compositions is ideal. This composition is considered ideal because the PCC/lithopone mixture (50:50) is close to the concentration of CaO, and the concentration total of BaO, ZnO, and SO_3 is 45.7:49.9. There was no significant difference in the concentration of CaO, BaO, ZnO, and SO_3 , indicating no chemical interaction between the two materials in the mixing process. The concentration of CaO in PCC was not significantly different from previous studies.¹⁸⁻²⁰

Table-2: Chemical Composition of Rutile TiO_2 , PCC, Lithopone, and PCC/Lithopone

Rutile TiO_2		PCC		Lithopone		PCC/Lithopone	
Compound	(wt/%)	Compound	(wt/%)	Compound	(wt/%)	Compound	(wt/%)
TiO_2	94.57	CaO	96.49	BaO	41.55	CaO	45.67

Al ₂ O ₃	1.65	P ₂ O ₅	0.99	ZnO	22.24	BaO	22.35
P ₂ O ₅	1.51	Cl	0.71	SO ₃	28.02	ZnO	14.98
ZrO ₂	0.95	Ag ₂ O	0.70	SiO ₂	2.86	SO ₃	12.57
Ag ₂ O	0.56	Al ₂ O ₃	0.53	Al ₂ O ₃	1.85	P ₂ O ₅	1.62
CaO	0.30	SiO ₂	0.35	P ₂ O ₅	1.69	Al ₂ O ₃	1.28
SiO ₂	0.20	Fe ₂ O ₃	0.11	SrO	0.72	SiO ₂	0.56
Nb ₂ O ₅	0.13	TiO ₂	0.08	CaO	0.56	Ag ₂ O	0.51
CdO	0.06	BaO	0.02	K ₂ O	0.35	TiO ₂	0.25
Fe ₂ O ₃	0.05	SrO	0.02	TiO ₂	0.12	SrO	0.11
ZnO	0.03	ZnO	0.01	Fe ₂ O ₃	0.04	Fe ₂ O ₃	0.09

XRD Analysis of Pigments

The XRD pattern is presented in Fig.-1. The characteristic peaks of TiO₂ at 2θ are 27.43°, 36.08°, 39.21°, 41.25°, 44.08°, 54.36°, 56.67°, 62.81°, 64.11°, 69.06°, 69.86°, 82.39°, 89.62°, 95.24°, and 96.04°. This result corresponds to the characteristic peak of rutile (01-087-0920). It is compatible with previous reports,⁶ which has a crystal size of 29.77 nm with a tetragonal shape and a crystallinity degree of 24.0 %.

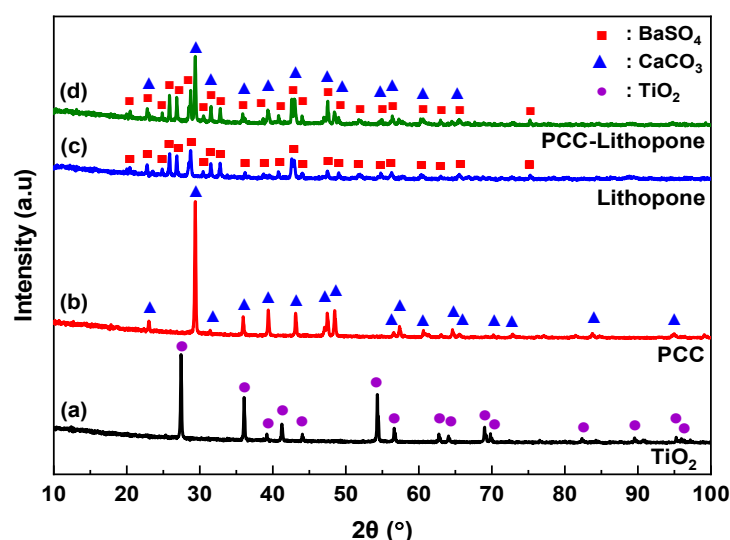


Fig.-1: XRD of (a) TiO₂; (b) PCC; (c) Lithopone; and (d) PCC/Lithopone

The characteristic peaks of PCC at 2θ are 23.03°, 29.37°, 31.41°, 35.96°, 39.39°, 43.14°, 47.51°, 48.49°, 56.57°, 57.39°, 60.64°, 64.66°, 65.58°, 72.89°, 83.79°, 94.85° and this corresponds to the characteristic peak of calcite (01-076-2712) and is aligned with prior works,¹⁹⁻²³ which has a crystal size of 21.48 nm with a rhombohedral shape and a crystallinity degree of 37.5%. The characteristic peaks of lithopone at 2θ are 20.46°, 22.76°, 24.87°, 25.84°, 26.85°, 28.70°, 30.46°, 31.53°, 32.77°, 36.37°, 38.73°, 40.78°, 42.74°, 44.04°, 47.51°, 49.04°, 51.78°, 54.77°, 56.28°, 60.28°, 65.52°, and 75.30°. This result corresponds to the characteristic peak of BaSO₄ (00-024-1035) and agrees with previous reports,¹⁴ which has a crystal size of 18.21 nm with an orthorhombic shape and a crystallinity degree of 78.4%. The characteristic peaks of the PCC/lithopone mixture at 2θ are 20.46°, 22.87°, 24.83°, 25.84°, 26.91°, 28.67°, 29.37°, 30.53°, 31.52°, 32.83°, 35.88°, 39.34°, 40.71°, 42.57°, 43.04°, 44.04°, 47.55°, 48.47°, 56.40°, 57.28°, 60.36°, 65.61°, and 75.28°. These correspond to characteristic peaks of CaCO₃ (01-076-2712) and BaSO₄ (00-005-0448), which have a crystal size of 29.12 nm with two types of crystal forms: rhombohedral and orthorhombic,^{14,20,21} and has a crystallinity degree of 91.5%. The mixing of PCC-lithopone pigments has slightly increased crystal size compared to the rutile TiO₂.

The whiteness of the PCC-lithopone pigment is not significantly different from the TiO₂ pigment. A fairly high crystallinity degree of PCC-lithopone can potentially increase the gloss of water-based paints compared to TiO₂ pigments. The presence of CaCO₃ resulted in water-based paint, a matt-type (not glossy). The presence of lithopone impacts the performance of the paint in terms of whiteness, gloss, and

hiding power in water-based paint. However, it is unknown whether it is caused by BaSO_4 or ZnS because there is no special treatment for the lithopone.

SEM Analysis of Pigments

Based on the results of morphological analysis using SEM (Fig.-2), the surface structure of the pigment particles is presented with a magnification of 10,000 times ($10\ \mu\text{m}$) and a magnification of 100,000 times ($1\ \mu\text{m}$). The particles of these pigments are fine, uniform in shape, and have a particle diameter size of less than $200\ \text{nm}$.^{1,6,24}

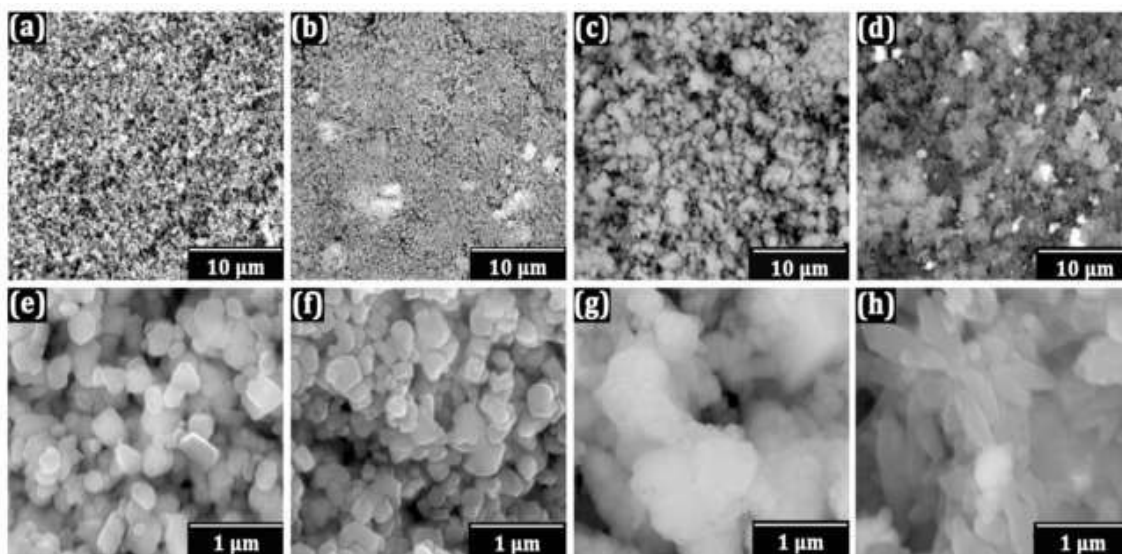


Fig.-2: SEM Images of (a,e) TiO_2 ; (b,f) PCC; (c,g) Lithopone; and (d,h) PCC-Lithopone

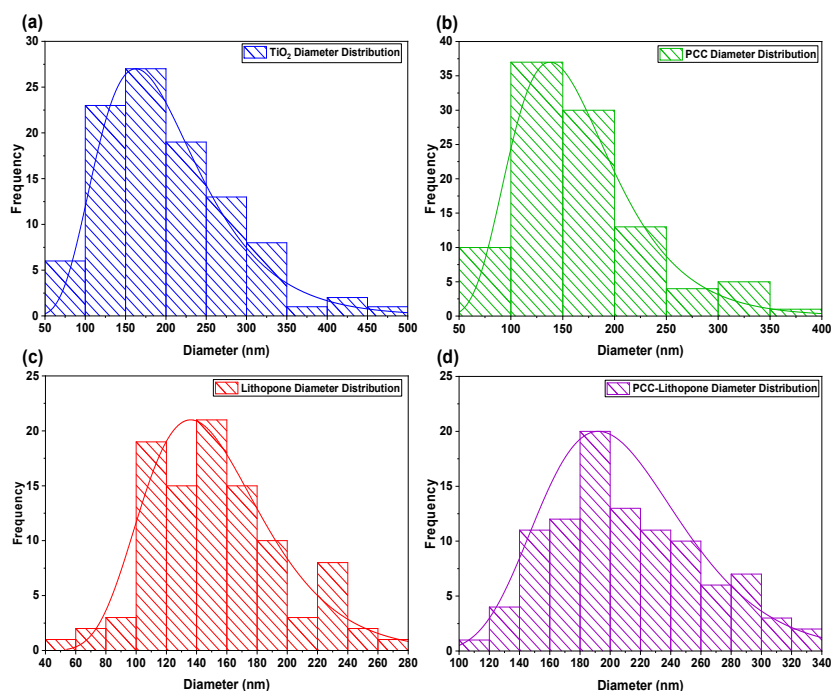


Fig.-3: Diameter Distribution of (a) TiO_2 ; (b) PCC; (c) Lithopone; and (d) PCC-Lithopone

The diameter distribution of particles was measured based on SEM results (Fig.-3). The average particle diameter of TiO_2 is $182.07\ \text{nm}$, PCC is $146.38\ \text{nm}$, lithopone is $144.57\ \text{nm}$, and PCC-lithopone is $198.29\ \text{nm}$. The particle diameter of all pigments is smaller than the sieve of mesh 200 ($74\ \mu\text{m}$). The small particle size is suitable for the pigment in matt-type water-based paint characterized by evenly dispersed

particles without any pigment remaining on the sieve. The morphology result showed that the dispersibility of the water-based paint is quite good.^{1,11}

Dispersibility Pigments, Hiding Power, Whiteness, and Gloss of Matt-Type Water-Based Paint

Based on Table-3, all pigment materials (TiO_2 , PCC, lithopone, and PCC-lithopone) were good dispersibility. The dispersion results of the pigment in the matt-type water-based paint showed that all samples (R1, R2, R3, and R4) have passed through a sieve of mesh 200. The absence of pigment particles left on the sieve has indicated that the pigment has been evenly dispersed (not agglomerated).²⁵

Table-3: Testing of Matt-Type Water-Based Paint with Various Pigments

Recipe Name	Whiteness	Gloss			Hiding Power	Dispersibility
		20°	60°	85°		
R1	79.53	1.33	2.03	3.27	Good*	Good**
R2	67.70	1.27	2.03	4.17	Good*	Good**
R3	68.57	1.23	2.07	3.33	Good*	Good**
R4	75.67	1.33	2.13	4.47	Good*	Good**

Notes:

* Evaluation of hiding power is carried out based on comparing the paint coating result against the standard white and black colors on the base of the paper panel.

**Evaluation of dispersibility is carried out based on the number of particles left on the sieve of mesh 200.

All samples have good hiding power quality in terms of hiding power, as shown in Fig.-4. These results indicate that PCC, lithopone, and PCC-lithopone pigments are good enough to be used as a pigments substitute for TiO_2 in matt-type of water-based paints. Based on previous reports, PCC, lithopone, and mixtures can substitute for TiO_2 in water-based paints.^{1,11,13,14} Based on the whiteness (Fig.-5a), the R4 sample has good whiteness qualities but is not significantly different from samples R1 with TiO_2 pigments.²⁵ The whiteness percentage of the R4 sample is 4.8% lower than the R1 sample. Meanwhile, the R2 and R3 samples had a whiteness percentage of 13.8% and 14.8%, which was lower than the R1 sample. These results indicate that individual PCC or lithopone pigment shows improved whiteness properties compared to TiO_2 pigment, while the PCC/ lithopone mixture shows no improvement in whiteness.

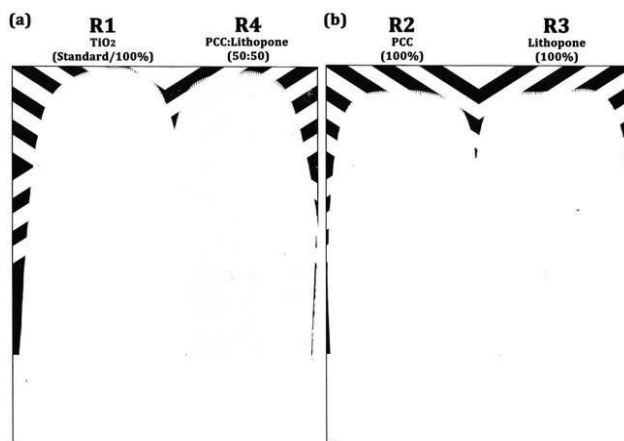


Fig.-4: A Hiding Power Test of (a) R1 (TiO_2) & R4 (PCC/Lithopone); and (b) R2 (PCC) & R3 (Lithopone)

Based on the gloss (Fig.-5b) at a reflection angle of 20°, R1 and R4 samples have the same gloss value of 1.33. At 60°, the R4 sample achieved a gloss value of 2.13, better than all samples. At 85°, the R4 sample achieved a gloss value of 4.47, the highest compared to all samples.^{11,25}

The dispersibility, hiding power, whiteness, and gloss of the R4 sample using PCC-lithopone pigment have shown the same quality as the R1 sample using TiO_2 pigment. However, it has a slightly lower (not more than 4.8%) whiteness than the R1 sample. The presence of PCC-lithopone pigment has improved the glossy value of the matt-type water-based paint.

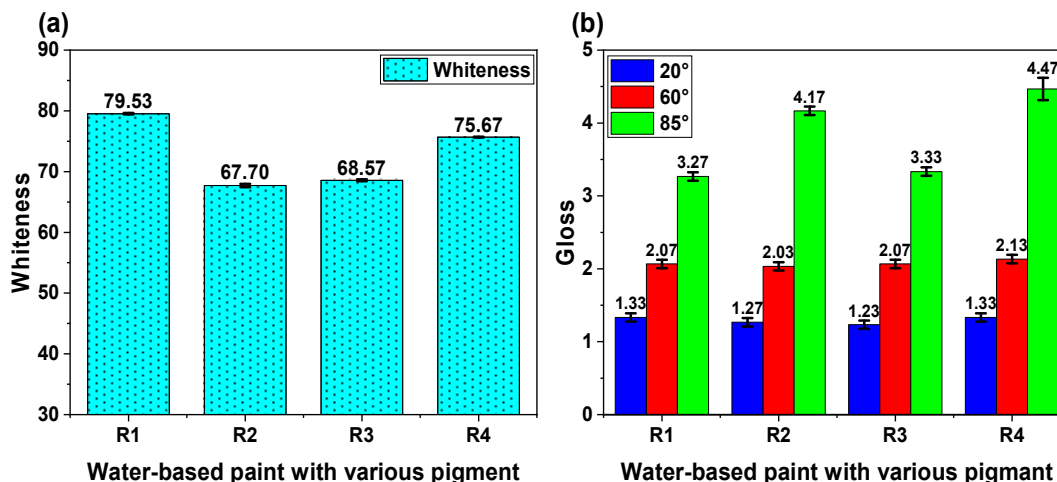


Fig.-5: A (a) Whiteness Test and (b) Gloss Test of A Matt-Type Water-Based Paint with Various Pigments

CONCLUSION

The function of PCC-lithopone as substitution pigments of TiO_2 was successfully optimized by mixing PCC and lithopone at 50:50 compositions, which was applied to a matt-type water-based paint. The application of PCC-lithopone to the water-based paint showed good hiding power and evenly dispersion due to the PCC/lithopone nanoparticles having a very fine, small particle size of 198.29 nm in diameter. The whiteness and gloss of PCC-lithopone originate from CaO , BaO , ZnO , and SO_3 compounds, which are 95.57%. The presence of ZnO compounds in PCC-lithopone resulted in a fairly high gloss, increasing PCC-lithopone's crystallinity degree by 91.5%.

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