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Reducing Defects in Water-Based Paint with Natural Dyes

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Abstract: Water-based paints with natural dyes are interesting to investigate, in particular with a view to the reduction of defects. This study aims also to report on the properties of water-based paints containing natural dyes of *Ceriops candolleana* Arn. in the context of parameters such as total solids, hiding power, drying time and gloss. The water-based paints were formulated from acrylic resin, water, polyethylene glycol and surfactant. The natural dyes and filler used were *C. candolleana* and TiO_2 powders, respectively. The sizes of the natural dye powders were examined using a particle size analysis instrument. The water-based paints were formulated with various concentrations of Pigment Volume Concentration (PVC) varying from 2.9-22.7%. The Filler Volume Concentration (FVC) used was 0, 1.4 and 2.8%. The results show that the water-based paints formulated using natural dyes from *C. candolleana* suffered defects such as agglomeration, bubbling and cracking. These defects were strongly affected by the concentrations of PVC and FVC. Water-based paints with fewer defects could be achieved, however, by formulating the paint with concentrations of PVC and FVC below 8 and 2.8%, respectively. The total solids, hiding power, drying time and gloss from the water-based paints with *C. candolleana* natural dyes were 40-46%, 5.8-13.7 m^2/L, 40-95 min and 9.5-56.6 GU, respectively and as such satisfied the SNI Standard 3564: 2009 for paints.

Key words: Water-based, paint, natural dye, defects, *Ceriops candolleana*, interesting to investigate

INTRODUCTION

The demand for water-based paints is increasing because in addition to being protective and giving an aesthetically pleasing look to the surfaces they cover, such as wood, iron, copper, paper, canvas, glass and concrete they are also environmentally friendly (Perrin *et al.*, 2009). Water-based paints refer to paints in which water is used as a solvent for dissolving the components and for adjusting the viscosity. The common resins used in water-based paints are latex, acrylic, polyvinyl acetate and styrene resin all of which are soluble in water. The conventional resins for solvent-based paints which include epoxy (Krishnamurti, 1983; Amo *et al.*, 2002; Ahmetli *et al.*, 2012), alkyd, polyurethane (Wang *et al.*, 2013; Papaj *et al.*, 2014), phenolic (Tilak, 1985), polyester, silicates and combinations or hybrids of these compounds (Guyot *et al.*, 2007) are derived from petroleum-based resources (Balgude and Sabnis, 2014) and have low molecular weights. In contrast, resins for water-based paints have a high molecular weight and therefore do not need to be cross-linked in order to develop adequate film properties (Lambourne, 1999). However, the method of cross-linking is very important in the formulation of paints with high Total Solid mass fraction (TS) required for environmentally friendly paints (Attanayake *et al.*, 1993).

The need for environmentally friendly paints has also led to the development of various types of Pigments and Dyes (P&Ds). Some P&Ds that are of good quality but that are environmentally unfriendly are being phased out too slowly. Natural Dyes (NDs) which have been growing in popularity for some time for the colouring of textiles (Baid, 2009; Erkan *et al.*, 2014; Sachdev, 2014; Alkan *et al.*, 2015) have also been successfully developed for Dye-Sensitized Solar Cells (DSSCs) (Zhou *et al.*, 2011; Attanayake *et al.*, 2013; Shahid *et al.*, 2013). A variety of NDs such as indigofera (Erkan *et al.*, 2014), coumarin (Elgemeie *et al.*, 2016), *Caesalpinia sappan* L. (Mulyanto *et al.*, 2016), neem and holy brazil extract (Sachdev, 2014) are well known and are used for colouring fabrics via covalent bond formation. NDs studied for DSSCs are curcumin (Sreekala *et al.*, 2012), black rice (Saehana *et al.*, 2013), tamarillo (Susanti *et al.*, 2014), *Canarium odontophyllum* fruits (Lim *et al.*, 2015), papaya leaf (Lim *et al.*, 2015), purple cabbage, coffee, blueberry and turmeric (Syafinar *et al.*, 2015). Dyes for DSSCs

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(Ludin *et al*., 2014) must have anchoring groups with semiconductors, a wide spectrum absorption of light and a conformity of its energy bands with the semiconductors and electrolytes. Unfortunately, not all natural dyes which succesfully used for fabrics and DSSCs can be directly used for colouring paints.

Interestingly, NDs have begun to be developed in the field of paint formulation (Pawlak *et al*., 2006; Abidin *et al*., 2013a, b; Shahid *et al*., 2013; Usop *et al*., 2016) and NDs which are commonly explored for paint production include curcumin for red (Abidin *et al*., 2013a), anthocyanin for red, purple and blue (Lee *et al*., 2015) and Streptomyces for brown, red, yellow and black shades (Sastry *et al*., 2016); *Thymus serpyllum* (Cakmakci *et al*., 2013) and lawsone have also been explored for brownish shades (Abidin *et al*., 2013b). The main problem with NDs when formulated in paints is conformity with the other components in the paint. However, many components for paints are available on the market and on the one hand these offer alternatives in formulation but on the other hand they may possibly introduce defects.

A good paint has to meet quality standards in terms of Volatile Organic Compounds (VOCs), hiding power, drying time, gloss, toughness and weather resistance. Some additives are also used to give resistance to bacteria, fungi, Ultraviolet (UV) light and fire (Slawson, 1996; Abidin *et al*., 2013a, b; Lee *et al*., 2015; Trouillas *et al*., 2016). In general, paints are formulated by mixing (Gurses *et al*., 2016) of the main four components, comprising resins, solvents, additives and pigments or dyes. These components, besides having special properties should also be compatible with one another, such that defects are avoided. Usually, defects occur simultaneously and this hinders the determination of their causes (Khataee *et al*., 2016). Unfortunately, the compatibility of water-based resins, surfactants and additives in paints which use Natural Dyes (NDs) is still seldom addressed, especially in the context of minimizing defects. Therefore, this study reports an investigation of the defects found in water-based paints with *Ceriops candolleana* including the effects of the Pigment Volume Concentration (PVC) and Filler Volume Concentration (FVC) together with several other factors.

MATERIALS AND METHODS

The materials used were acrylic resin (PT. Lautan Luas tbk), polyethylene glycol (PG, PT. Agung Jaya), TiO_2 (PT. Agung Jaya) and polycarboxylate polymer type surfactant (POIZ 530, PT KAO Indonesia Chemicals). The materials were used without any further treatment.

The natural dyes used were extracted from the wood of *Ceriops candolleana* Arn. Samples of *C. candolleana* wood which were bought from a local market were cut into chips with a size of 3-4 cm. The wood chips were macerated for 4 h in water at a temperature of 80°C (Mulyanto *et al*., 2016). Afterwards, the dyes dissolved in water were cooled and filtered to separate the fibres and other solids. The extracted dyes were then formed into powders by use of a spray dryer operated at a temperature of 110°C. The size of the dye powders was examined using a Particle Size Analysis machine (PSA, Delsa Nano C Beckman Coulter).

The water-based paints were formulated with various volume concentrations of pigment and filler. PVC was calculated from the ratio of the mass of dyes to the total mass of paint and FVC was calculated from the ratio of the mass of filler (TiO_2) to the total mass of paint. FVC was varied from 2.9-22.7% and FVC was varied at 0, 1.4 and 2.8%. The concentrations of water, surfactants and Propylene Glycol (PG) in paints were 7, 4.3 and 14%, respectively. All the components were mixed at 1000 rpm for 3 h.

The paints were then tested for TS, hiding power, drying time, glossiness and defects. Testing for TS was conducted by coating the paints onto the substrate and heating in a furnace at a temperature of 110±5°C for 60 min. The residue was considered to be the TS. The drying time was measured by lightly touching the paints coated on the substrate at intervals of 60 sec. The paint layers were considered to be dry when they did not leave marks on the fingers when touched at the same observation area. Meanwhile, glossiness was tested using a Gloss meter ETB-0686. In addition, the hiding power was measured according to how much paint was used to cover an area of 1 m^2.

The dry paint was examined with a microscope XSZ-107BN. The amount of agglomeration and other defects was calculated according to the ratio of the defect area to the area of observation in the images from the microscope which was calculated by using ImageJ software.

RESULTS AND DISCUSSION

Figure 1 shows images of water-based paints with dyes from *C. candolleana*. These paints were widely perceived to be of a red and red-brown colour. The greater the PVC, the deeper the colours of the paints appeared. A higher FVC resulted in lighter red colours. The paints with very high FVC and PVC were of poor quality.

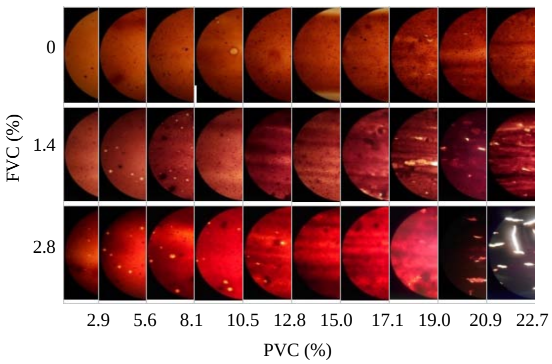

Fig. 1: Images of water-based paints

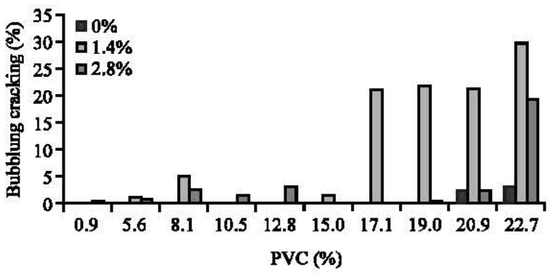

Fig. 2: Bubbling and cracking defects occurring in water-based paints with *Ceriops candolleana* natural dyes

The poor quality of the paints produced at very high volume concentrations of filler and pigment was due to defect formation in the form of agglomeration, bubbling and cracking. The defects due to bubbling and cracking had a random pattern as shown in Fig. 2. Bubbling defects are frequently seen in paints (Fitzsimons and Parry, 2010) and consist of small domes that rise to the intact or damaged surface and may also leave craters. Bubbling defects may be caused by air trapped in the layers of paint before the surface dries. In addition, the difference between the surface tension of the paint and that of the substrate may also cause this type of defect. Moreover, this type of defect may also occur on contaminated surfaces with contaminants such as particles or oil. However, in general, the defects occur because there are areas experiencing stress concentration (Americus, 1982).

In addition to bubbling and cracking defects in the water-based paints with NDs from *C. candolleana* there were also defects in the form of dye agglomeration. The agglomeration of dyes can be caused by low amounts of surfactant and tends to be worse when the paints are mixed with dyes. As a result, the dispersion force of the dyes decreases. Young's equation indicates that:

$$\gamma_s = \gamma_{sl} + \gamma_l \cos\theta \quad (1)$$

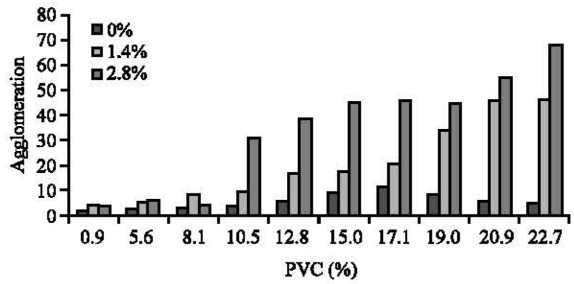

Fig. 3: The agglomeration found in water-based paints with *Ceriops candolleana*

Where:

γ_s = The free surface energy of the solid
γ_{st} = The solid-liquid interfacial energy
γ_l = The surface tension of the liquid
θ = The contact angle at the solid-liquid interface

The main parameter determining the dispersion ability of the dyes in solution is the wetting quality. A wetting quality is achieved if the surface tension of the liquid (γ_l) is lower than that of the solid. If there are filler particles of TiO_2 in the paint that have $\theta = 0$ then the surface tension of the liquid (γ_l) will take a value which is good for dispersion; however, the addition of TiO_2 filler to water-based paint with *C. candolleana* was not able to improve the dispersion of the dyes which was characterized by an increase in agglomeration with rising filler concentration as shown in Fig. 3.

Therefore, the wetting ability of the dyes does not lead to stable emulsions with good dispersion. However, the smaller the sizes of the dye particles, the more easily are the dyes dispersed in the emulsion. The magnitude of the attraction Force (F_A) is expressed by Doroszkowski (1999):

$$F_A \simeq -\frac{Ar_1r_2}{6d(r_1+r_2)} \quad (2)$$

Where:

A = Hamaker's constant
r_1 and r_2 = The radius of the 1st and 2nd dyes
d = The distance between the dyes

Here, the role of the repulsion-attraction forces among particles of dye which have sizes bigger than 10 nm is significant. Therefore, the role of the surfactant decreases because the average diameter of *C. candolleana* powders is 378.7 nm as can be seen in Fig. 4. In addition to the particle size of the dyes, the magnitude of the attraction force is also influenced by the distance between the dye particles resulting from the value of the PVC. Therefore, the quality of the water-based paints with *C. candolleana* was mainly

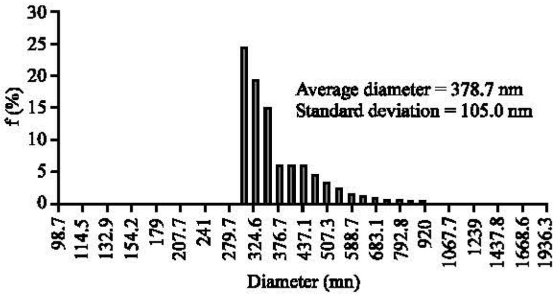

Fig. 4: Particle size of *Ceriops candolleana* powders

influenced by the PVC and FVC. The higher the PVC and FVC, the higher the degree of agglomeration was achieved, especially with PVC of more than 10% as shown in Fig. 3.

The distribution of the dye particles in the paint is strongly influenced by the properties of the dye, the solvent, the surfactant and the filler. A good distribution of dye particles can be maintained in the absence of aggregates, flocculation, agglomeration or clustering in the paints; however, agglomeration is more pronounced in water-based gloss paints than in solvent-based paints (Tiarks *et al*., 2003).

The mechanism for the formation of colour in paints is different from that in textiles and DSSCs which requires that solvent-soluble dyes are used. Bonding to textile dyes and DSSCs generally occurs covalently (Shahid *et al*., 2013). With paints, the dyes are generally dispersed in solvents and resins. In order to provide a visually pleasing result in the form of colour and gloss, the dyes should be well dispersed and there should be no agglomeration.

The function of the dye is to give a definite colour to the paint. Therefore, the natural dyes should have good optical properties, be resistant to light and heat and be resistant to other substances that are acidic or alkaline. Therefore, the quality of the water-based paints is not only determined by the resin/solvent but is also determined by the properties of the dyes used.

Low TS results in a high volatile content that can evaporate and pollute the environment. The use of water as a substitute for petroleum-based solvents should also be controlled, although, water is more environmentally friendly. According to the required standards, the TS in paint is required to be more than 40% and the water-based paint with *C. candolleana* already has a TS value above 40% as shown in Fig. 5. The higher the PVC and FVC, the higher is the TS content. On the other hand, too high a TS content can cause low fluidity of the paints.

The hiding power of the water-based paint containing natural dye from *C. candolleana* showed a downward trend with increasing PVC as shown in Fig. 6. The use of

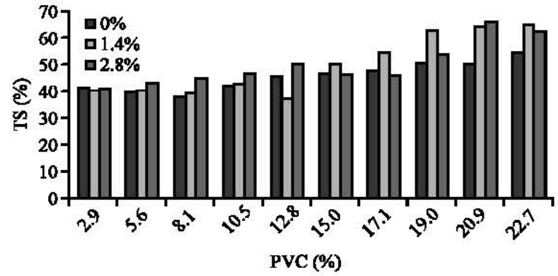

Fig. 5: The concentration of total solid in water-based paints with *Ceriops candolleana*

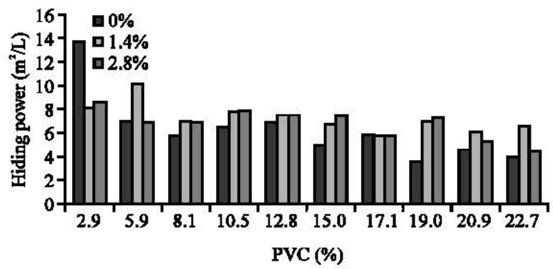

Fig. 6: Hiding power of water-based paints containing *Ceriops candolleana* natural dyes

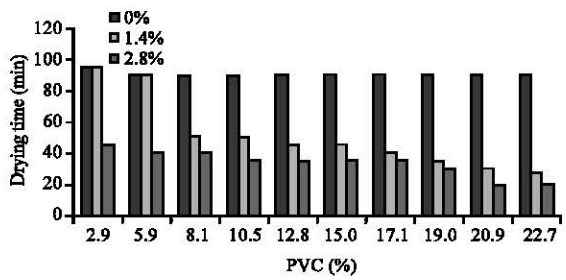

Fig. 7: Drying time of water-based paints containing *Ceriops candolleana* natural dyes

FVC with TiO_2 improved the hiding power slightly. A hiding power >8 m^2/L can be achieved with a FVC of 1.4% and a PVC of up to 10%.

The drying time of water-based paints without FVC was very slow and was only slightly influenced by the PVC. Interestingly, the addition of filler can significantly reduce the drying time as can be seen in Fig. 7. In the water-based paints, the drying time is not only influenced by the evaporation of water because the various ingredients of the paints may form cross-links (Gurses *et al*., 2016). Moreover, acrylic resins serve not only as a binder but also as a booster for the drying rate of water-based paints. Acrylic resins also have a strong impact on properties such as gloss, flexibility, toughness and resistance to degradation under UV irradiation (Midtdal and Jelle, 2013).

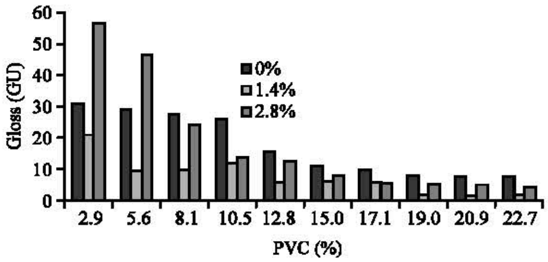

Fig. 8: Glossiness levels of water-based paints containing *Ceriops candolleana* natural dyes

Dyes not only affect the colours but also have a major influence on the visual appearance of paints, especially the glossiness level. Gloss is one of the key properties and is of course very important to visual appearance (Tiarks *et al*., 2003). Figure 8 shows that an increase in concentration of the pigment in paints leads to a lower gloss because of the presence of a larger amount of pigment on the surface. Since, the pigment has a higher refractive index than the resin, the gloss decreases with an increasing amount of dye in paints. Therefore, the even distribution of dye in paint is critical and directly affects the properties of gloss and hiding power (Doroszkowski, 1999; Tiarks *et al*., 2003). On the other hand, the addition of filler at 1.4% can lead to a decrease of the glossiness level of water-based paints containing *C. candolleana*. The addition of filler at concentrations of up to 2.8% quite sharply increases the glossiness level because TiO_2 has a high refractive index (nD = 2.5-2.7).

CONCLUSION

Water-based paints have been successfully formulated using natural dyes from *C. candolleana*. There are defects in water-based paints containing *C. candolleana* such as agglomeration, bubbling and cracking which are affected by the PVC and FVC. Water-based paints with low defects can be achieved by formulating the paint with a PVC and FVC below 8% and 2.8%, respectively. The total solid, hiding power, drying time and gloss for the water-based paints containing *C. candolleana* were 40-46%, 5.8-13.7 m^2/L, 40-95 min and 9.5-56.6 GU, respectively and therefore satisfied the quality standards for paints. Therefore, this formulation is important for coating industries to better understand their formulation of water-based paints using natural dyes and how to minimize the defects such as agglomeration, bubbling and cracking. Indeed, the free defects in water-based paints with natural dyes are an interesting topic for further research.

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