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An Investigation on the Properties of Eco-Friendly Alkyd and Alkyd Composite Coatings

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ABSTRACT

Reducing the use of harmful solvents in alkyd paints is a major challenge for researchers. This paper compares some properties of eco-friendly alkyd and alkyd composite coatings. The method used in this work is to compare the properties of Eco-friendly alkyd and alkyd coatings. An eco-friendly alkyd paint was fabricated using an intermediate emulsion made from polysaccharide (LPR) and about 25 weight percent (wt.%) of water. Infrared spectroscopy (FTIR), scanning electron microscopy (SEM), and thermogravimetric analysis (TGA) were used to characterize the material. Impact resistance, flexibility, adhesion, drying time, etc., of an eco-friendly alkyd composite coating were also studied in comparison with those of an alkyd coating. Results showed that both composite coatings performed well in adhesion, flexibility, and impact resistance, with the highest values. Meanwhile, the relative hardness and drying time of the eco-friendly alkyd composite coating were better than those of the alkyd composite coating. After 80 cycles of UV irradiation exposure testing, both composite coatings can be maintained at a high level in the cross-cut test. Meanwhile, the impact resistance of the alkyd coating decreased from 200 kg·cm to 140 kg·cm, and that of the eco-friendly alkyd coating decreased from 200 kg·cm to 180 kg·cm, respectively. This study showed that eco-friendly alkyd coatings could be used instead of conventional alkyd coatings for outdoor applications.

Keywords:
Alkyd Paint, Eco-friendly Coating, Green Coating, Composite Coating, UV Aging, VOC Reduction

1. Introduction

Alkyd paint is the most widely used. It is used to prevent rust and to decorate metal, wood, and other materials, both indoors and outdoors [1-4]. The biggest disadvantage of alkyd paint is much less eco-friendly than other common synthetic paints. Because oil-based paints, such as alkyds, contain volatile organic solvents, these solvents are released into the air when paints are used, posing a risk to people and the environment [5-7]. To reduce the harmful effects of alkyd paints, many authors have been studying on using less solvent. Selim *et al.* [8] synthesized eco-friendly

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hyperbranched alkyd resins from sunflower oil. This kind of alkyd was low-viscosity, low molecular weight, highly branched, highly functional, and abundant in surface functional groups, making it suitable for manufacturing low-VOC coatings. With 0.5 wt.% of nano ZnO, this alkyd resin had significant improvement in its anticorrosive, chemical, and mechanical properties. Sharmin *et al.* [9] studied on environment friendly coating from non-edible, non-medicinal vegetable oil. The coating demonstrated its high solids, water-borne, and UV-curable performance. Ursula Biermann *et al.* [10] investigated eliminating organic solvents by using tung oil- based reactive diluents. The diluents significantly reduced alkyd viscosity, thereby improving film formation and drying performance. Other authors have used wool fibers, polyhydroxyalkanoates, cellulose, chitosan, lignin, rosin, and starch in paints to reduce volatile components and make eco-friendly paints [11-15].

Polysaccharide (LPR) is a vegetable oil-based resin. In which hydroxyl groups (OH) of polysaccharide resin can make hydrogen bonds with carboxyl groups (COOH) of alkyd resin to form a denser, higher molecular weight polymer network [16, 17]. It can be used to reduce volatile organic compounds (VOCs) when added to paints. LPR and water can be added to paint using either the "direct water addition" method or the "Emulsion Intermediate" method. In the "direct water addition" method, LPR and water are added directly to the paint at very high grinding speeds. In the "Emulsion Intermediate" method, an Emulsion Intermediate (EI) can be made by pre-mixing binders, solvents, LPR, and water, then adding EI to paints along with the other components [17-19].

Although polysaccharides have been used in paints to reduce the use of harmful solvents, publications on this topic are limited. This paper introduces an investigation into the properties of alkyd composite coating and an eco-friendly alkyd composite coating made from LPR using the "Emulsion Intermediate" method. To evaluate the properties of the studied eco-friendly alkyd composite coating, physicochemical properties, thermal oxidation resistance, UV-humidity complex stability of eco-friendly alkyd composite coating, and alkyd coatings were investigated so as to compare their quality. The properties of composite coatings were also examined before and after 80 cycles of UV irradiation.

2. Materials and Methods

2.1. Chemicals

Alkyd resin, CRESTAKYD 10-1019, was supplied by Scott Bader Company Limited (UK). Polysaccharide Resin LPR 76 was supplied by Lorama Group Inc. (Canada). Methyl Ethyl Ketoxime (MEKO) was supplied by Maldeep Catalysts PVT LTD (India). Xylene, bentonite, lead octoate (32%), carbon black (N220): Industrial products (China). Kerosine was supplied by Petrolimex (Vietnam). TiO_2, Rutile R-996, was a product of Sichuan Lomon Corporation (China).

2.2. Sample Preparation

2.2.1. Emulsion Intermediate Preparation

Raw materials were prepared as in Table 1. Firstly, the alkyd resin was stirred at a speed of 1,400-1,600 rpm, then kerosine and xylene were added gradually. After that, the speed was adjusted to 2,900-3,000 rpm, LPR 76 was added, and stirring was maintained for 20 minutes. Finally, water was added and continued stirring at 2,900- 3,000 rpm for about 20 minutes until a homogeneous Emulsion Intermediate (EI) solution.

Table 1
Constituents of Emulsion Intermediate (EI)

No.	Components	Content (wt. %)
1	Alkyd resin	15
2	LPR 76	12
3	Kerosine	6
4	Xylene	7
5	Water	60

2.2.2. Paint preparation

Raw materials were prepared in Tables 2 and 3. The grinding included 2 stages: (1) *Primary grinding*: 90 wt. % of xylene and all others were added, then stirred at 40-50 rpm for 1 hour. The mixture was kept for 20 hours before moving to the next stage. (2) *Fine grinding*: the mixture was ground at a speed of 1,400- 1,500 rpm, until getting paint fineness of $\leq$ 30 µm. *(3) Preparation:* The rest of the xylene was added and stirred at a speed of 20- 40 rpm for half an hour. A 100 hole/mm^2 mesh was used to remove any dirt and large particles before canning for storage.

Table 2
Constituents of eco-friendly alkyd paint

No.	Components	Content (wt. %)
1	Alkyd resin	20
2	EI solution	42
3	Xylene	11
4	Kerosine	6
5	TiO_2	15
6	N220	4
7	Bentonite	1
8	Lead octoate	0.5
9	MEKO	0.5

Table 3
Constituents of alkyd paint

No.	Components	Content (wt. %)
1	Alkyd resin	26
2	Xylene	35
3	Kerosine	18
4	TiO_2	15
5	N220	4
6	Bentonite	1
7	Lead octoate	0.5
8	MEKO	0.5

2.3. Sample preparation

Composite coatings were deposited on the cleaned panels using a sprayer at 4 kg/cm^2. Coating samples for FT-IR analysis were prepared on glass sheets with a dried thickness of (15±3) µm. Coating samples for morphology were prepared as above with a thickness of (120±7) µm. Coating samples for evaluation of mechanical properties with a thickness of (30±5) µm were prepared on steel panels according to ISO 1514:2004. Samples were kept at room temperature for 7 days before testing. The samples were prepared and kept in a room with a temperature of (25± 2) ^{o}C and relative humidity within the range of 50–60 %.

2.4. Analysis Methods

UV exposure tests of coatings were conducted in a UV/condensation weathering chamber, Atlas UVCON UC-327-2, with UVB-313 fluorescent lamps, in accordance with ASTM G53, and operated under a cycle comprising 8 hours of UV irradiation at 60 °C followed by 4 hours of condensation at 50 °C.

Infrared spectroscopy (FTIR) was conducted on the Fourier FTIR-8700 series converter. The morphology of the coatings was observed using a FESEM Hitachi S4800 at 5,000× magnifications and 5 kV.

The gloss value of the coating was determined according to ISO 2813:2014 at an angle of 60 degrees. Adhesion of coating was determined according to ISO 2409:2013, impact resistance of coating was determined by ISO 6272-1:2011, Flexibility of coating was determined according to ISO 1519:2011 and relative hardness of coating was determined according to ISO 1522:2006, Surface drying of coating was determined according to ISO 9117-3:2010, Through drying of coating was determined according to ISO 9117-1: 2009, cross-cut test of coating was determined according to ISO 2409: 2013.

Thermal oxidation resistance: Thermo-gravimetric analysis (TGA) was performed using a NETZSCH TG 209F1 LIBRA with a heating rate of 10 °C/min in air from room temperature to 600 °C.

3. Results and Discussion

3.1. Physicochemical properties of alkyd and eco-friendly alkyd composite coatings

Alkyd and eco-friendly alkyd composite coating samples were applied to standard panels and left for 7 days before testing. Results were shown in Table 4.

Table 4
Physicochemical properties of alkyd and eco-friendly alkyd coatings

No.	Properties	Units	Alkyd coating	Eco-friendly alkyd coating
1	Adhesion	Points	1	1
2	Flexibility	mm	2	2
3	Impact resistance	Kg.cm	200	200
3	Relative hardness	-	0.37	0.42
4	Gloss value at 60^o	%	82	90
5	Surface drying	Minutes	31	23
6	Through drying	Minutes	1,170	1,105

Table 4 shows that both alkyd coating and the eco-friendly alkyd composite coating exhibit good mechanical properties, including adhesion, flexibility, and impact resistance. Surface drying of the eco-friendly alkyd coating was much shorter than that of the alkyd coating. Besides that, the relative hardness of the eco-friendly alkyd coating was higher than that of the alkyd coating. This can be explained by the fact that OH groups of polysaccharides reacted with COOH of alkyd to make higher cross-link and higher Figure 1a of liquid eco-friendly alkyd paint with peaks as 3028 of vC=C (alkene), 2965, 2926, 2857 of vCH (aliphatic hydrogens), 1725 of C=O in ester, 1263 of unsaturated ester, etc. In Figure 1b, peak 3028 was absent, and other peaks, such as 1377, 802, etc., disappeared [19]. Besides that, peaks of fatty acids as 1725,80 and 1724,20 were changed to 1724,25 and 1720,86, or peaks of vinyl groups as 1602,07 and 1598,86 were changed to 1604,12 and 1598,71 on Figure 1c and Figure 1d in comparison to Figure 1a and Figure 1b. It meant that curing molecular-weight polymer chains, so the eco-friendly alkyd composite coating had a shorter drying time and higher relative hardness compared to the alkyd composite coating [16-18].

3.2. *Infrared spectroscopy (FTIR) analysis*

To study the changing of functional groups between liquid eco-friendly alkyd paint, alkyd paint, and those coatings. FTIR spectroscopy was used. Results were shown in Figure 1 and Table 5. The alkyd resin reacted to form the coating. Alkyd resins were dried by both physical (solvent evaporation) and chemical curing (in which oxygen from the atmosphere reacted with unsaturated bonds in the oils to create cross-links via radical reactions). These processes occurred over a very long time [1, 4, 17, 20]. Figures 1c and 1d for alkyd paint and composite coating are the same as for eco-friendly alkyd.

Table 5
Fluctuations in infrared coatings

No.	Typical spectrum	Wavenumbers (cm^{-1})
1	vas(CH_2=), asymmetry in vinyl groups	3028; 3025
2	vas(CH_2=), asymmetry in carbon chains	2965; 2957; 2924; 2923; 2921
3	v (CO) of fatty acids	1725; 1724; 1720
4	v (C=C) in vinyl groups	1604; 1602
5	vCH	1263; 1256; 1266; 1254
6	vC–O stretch, vibrations	1119; 1118; 1117; 1116
7	vaC-O-C (asymmetry)	1070; 1068; 1066
8	δ(CH), oscillation deformation of CH in aromatic rings	768; 740

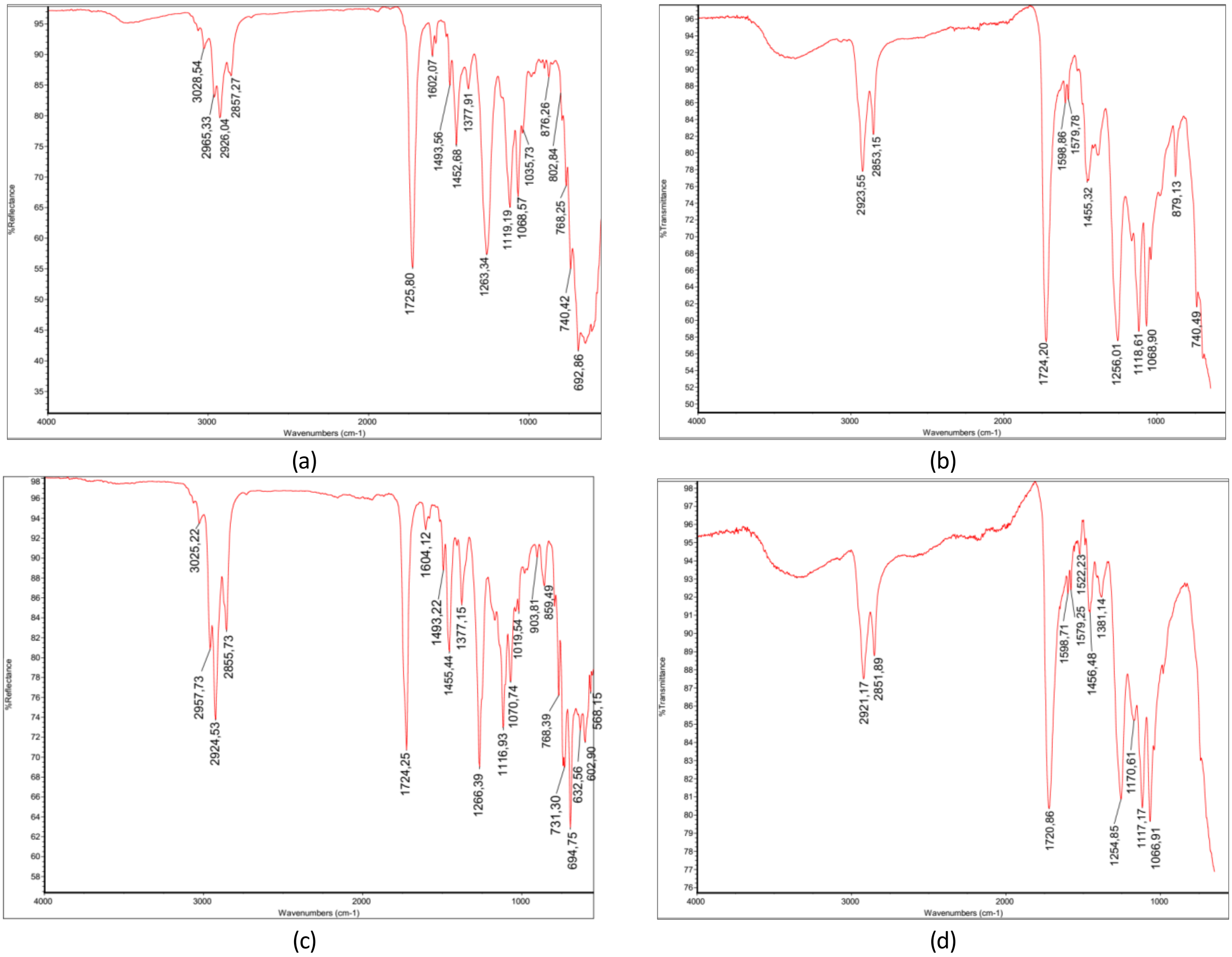


Fig. 1. (a) IR of eco-friendly alkyd paint, (b) eco-friendly alkyd coating, (c) alkyd paint, and (d) alkyd coating

3.3. UV exposure stability of composite coatings

To investigate the effects of UV radiation on coatings, alkyd and eco-friendly alkyd coating samples were applied to steel standard panels and left for 7 days before testing with 80 cycles of UV irradiation. Results were shown in Figures 2 and 3, Figure 4, Table 6, and Table 7.

Table 6
Gloss and mechanical properties of aged alkyd and aged eco-friendly alkyd coatings

No.	Properties	Units	Alkyd coating	Eco-friendly alkyd coating
1	Adhesion	Points	2	2
2	Flexibility	mm	4	3
3	Relative hardness	-	0.52	0.49
4	Gloss value at 60 °	%	70	82

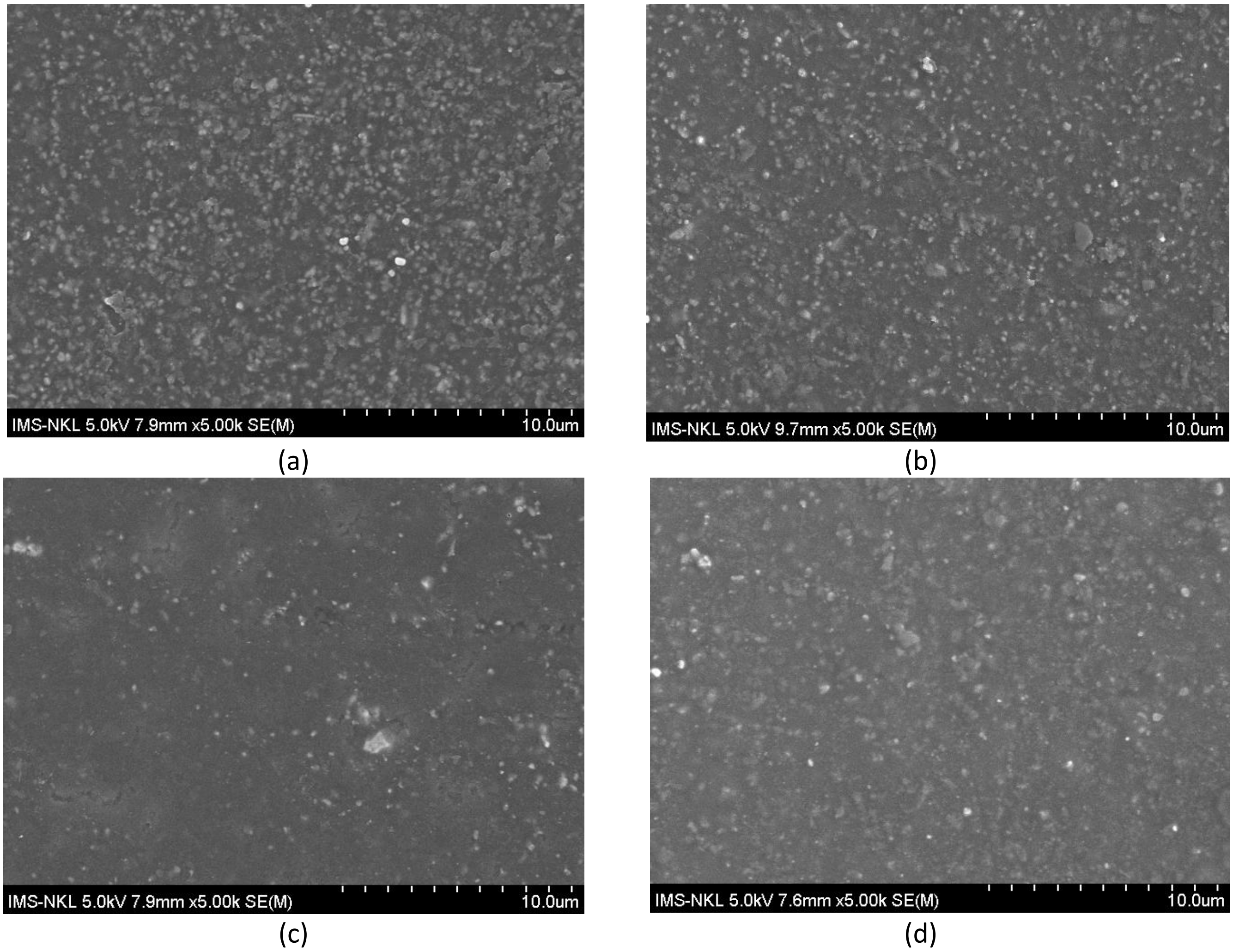


Fig. 2. SEM of initial coatings' surfaces of alkyd coating (a), eco-friendly alkyd coating (c), and aged coatings' surfaces of alkyd coating (b), aged eco-friendly alkyd coating (d)

Figure 2 shows that, in general, after testing, both coating surfaces were rougher than before. But there were no cracks, blisters, or surface changes in the samples investigated for both eco-friendly and alkyd coatings. This meant that both composite coatings could be subjected to complex combinations of UV, thermal, and humidity conditions for extended periods. Table 6 shows that after 80 aging cycles, the adhesion and flexibility of both coatings had not decreased significantly. Besides that, the relative hardness and gloss of the alkyd composite coating became much worse than those of the eco-friendly alkyd composite coating. In which the relative hardness of alkyd composite coating increased from 0.37 to 0.52, flexibility reduced from 2 mm to 4 mm, and gloss reduced from 82 % to 70 %. Meanwhile, the relative hardness of the eco-friendly composite coating increased from 0.42 to 0.49, flexibility decreased from 2 mm to 3 mm, and gloss decreased from 90 % to 82 %. This meant that, compared with the eco-friendly alkyd composite coating, the alkyd composite coating became more brittle and lost more adhesion and gloss. It can be explained that the tighter molecular packing and longer polymer chains of the eco-friendly composite coating prevented polymer chains from cutting or breaking, thereby reducing the UV-induced aging of this coating compared to the alkyd composite coating [18, 20].

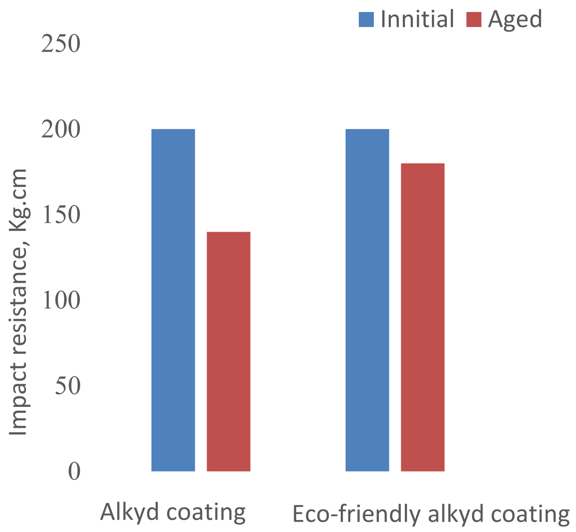


Fig. 3. Impact resistance of coatings before and after 80 cycles of UV irradiation exposure test

Table 7
Cross-cut classification and impact resistance results for alkyd and eco-friendly alkyd coatings

Samples	Cross-cut classification		Impact resistance (Kg.cm)	
	Before aging	After aging	Before aging	After aging
Alkyd coating	0	1	200	140
Eco-friendly alkyd coating	0	0	200	180

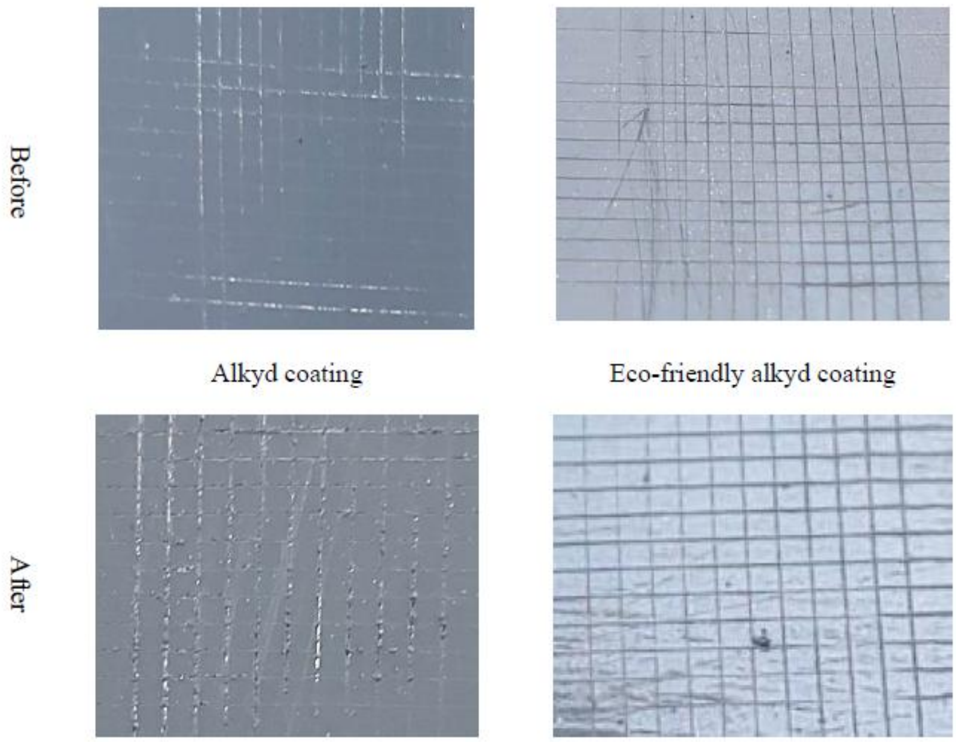


Fig. 4. (a) Optical photographs on the surface of coatings examined by crosscut before and after 80 cycles of UV irradiation exposure test for alkyd, and (b) eco-friendly alkyd

Figures 3 and 4 and Table 7 show that after 80 cycles of UV irradiation testing, the impact resistance of the alkyd composite coating decreased more than that of the eco-friendly alkyd composite coating. The impact resistance of the alkyd composite coating decreased from 200 kg·cm to 140 kg·cm; meanwhile, it decreased from 200 kg·cm to 180 kg·cm for the eco-friendly alkyd one.

To examine the interface between the substrate and coatings during UV irradiation testing, a cross-cut test was performed. Before the UV irradiation exposure test, both coatings had good adhesion, rated level "0". After 80 cycles of UV irradiation exposure, coatings could still reach level "1" for alkyd coating and level "0" for eco-friendly alkyd coating, respectively. The results demonstrated that alkyd and eco-friendly alkyd coatings could protect substrate from UV irradiation for long periods of time (80 cycles of UV exposure test in a UV/condensation weathering chamber with UVB-313 fluorescent lamps and operation was processed under conditions of a cycle comprising 8 hours UV irradiation at 60 °C then following by 4 hours of condensation at 50 °C) and those coatings could be used for outdoor purposes [21-23].

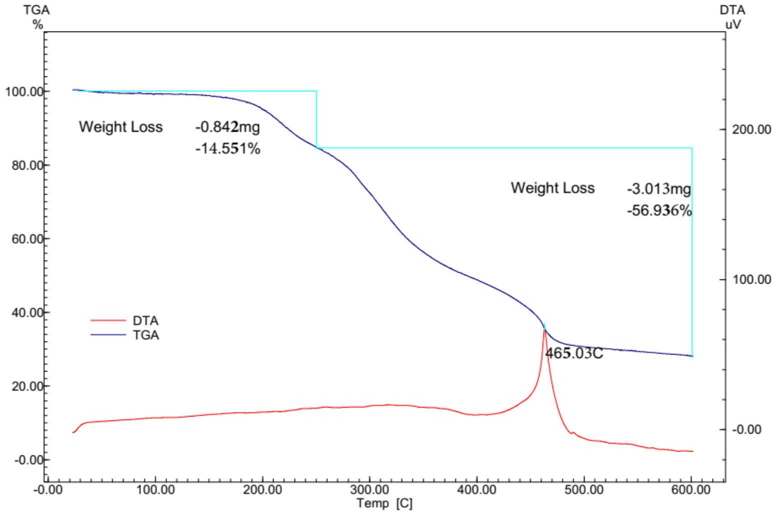
3.4. Thermal oxidation resistance of composite coatings

To study the thermal oxidation resistance of alkyd coatings, including eco-friendly alkyd coatings, Thermogravimetric analysis (TGA) was used. Results were shown in Table 8 and Figure 5.

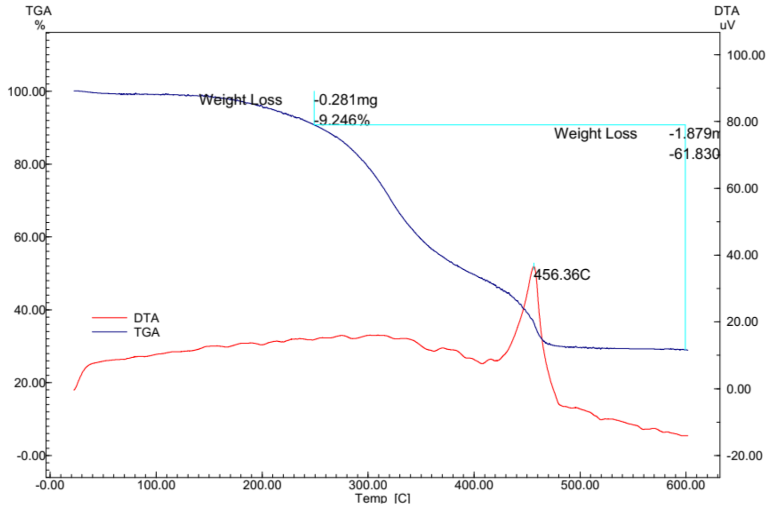
Table 8 and Figure 5 showed that, at different temperatures, the TGA curves of the alkyd coating and the eco-friendly alkyd coating differed. Results showed that when the temperature was below 200 °C, decomposition of low molecular substances and residual solvents, and weight loss of coating samples were about 4-10 %. From 200 ^{o}C to 400 ^{o}C, the eco-friendly alkyd coating showed lower weight loss than the alkyd coating. Figure 5 also showed differences in the TGA curve slopes between Figure 5a and Figure 5b. Figure 5b had a smaller slope than Figure 5a, which meant that the thermal oxidation stability of the eco-friendly alkyd coating was higher than that of the alkyd coating. This can be explained by the fact that the eco-friendly alkyd coating had a higher cross-link density and higher molecular weight than the alkyd coating, so its tighter molecular structure prevented oxygen from penetrating the coating and improved its thermal oxidation resistance. Besides that, for eco-friendly coating, the polymer network structure of OH groups in polysaccharides reacts with the COOH groups of alkyd, forming higher cross-link density and higher molecular-weight polymer chains, creating stronger, entangled networks that restrict chain mobility. It needed more energy to break bonds that why the coating was more stable, durable, and resistant to high temperatures [17, 24].

Table 8
Thermal oxidation resistance of composite coatings

Samples	Weight loss (%)		
	300 ^{o}C	400 ^{o}C	500 ^{o}C
Alkyd coating	25.72	53.69	72.46
Eco-friendly alkyd coating	19.44	50.26	71.08



(a)



(b)

Fig. 5. (a) TGA of alkyd coating, (b) eco-friendly alkyd coating

4. Conclusions

An eco-friendly alkyd coating with about 25% water in the paint has good mechanical properties and better relative hardness and drying time than the alkyd one. The thermal oxidation resistance of eco-friendly alkyd paint is higher than that of alkyd coating. After 80 cycles of UV irradiation exposure test, the adhesion and flexibility of both coatings did not decrease remarkably. Relative hardness of alkyd coating increased from 0.37 to 0.52 (about 40 %), flexibility reduced from 2 mm to 4 mm, and gloss reduced from 82 % to 70 % (about 15 %). The relative hardness of the eco-friendly coating increased from 0.42 to 0.49 (about 15 %), flexibility decreased from 2 mm to 3 mm, and gloss decreased from 90 % to 82 % (about 9 %). Surface drying was reduced by about 25%, and through drying by about 5%. The study's results showed that eco-friendly alkyd coating can be used instead of conventional alkyd coating for outdoor applications.

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