



Evaluation of Natural Oil Adducts in Alkyd-Based Varnish Emulsion and Effect on Rowan (*Sorbus torminalis*) Wood

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Author's contribution

The sole author designed, analyzed, interpreted and prepared the manuscript.

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ABSTRACT

In these days, preparation of eco-friendly wood surface coating agents has become important topic for environmental concerns. In this study, an alternative surface coating formulations was prepared with adducts of oils of apricot kernel (A), sesame (S) and grape seed (G) were mixed with oil modified alkyd-based synthetic varnish system. These emulsions were applied on Rowan (*Sorbus torminalis*) wood and exposed artificial UV irradiation. However, all emulsion coated samples show lower water sorptions regardless of conditions or level of treatments. Increasing adduct charges from 5.0% to 10% have not considerably effects for water sorptions. The reducing water sorption properties of 35.5%, 35.2% and 39.4% were obtained with 10% sesame oil-varnish coated samples of 10S_I and 10 S_{II} in one- and two-time coatings and three-times coated samples of 5S_{III} in 5.0% sesame oil-varnish emulsion coatings, respectively. Moreover, selected oil adducts into varnish had one and two point lowering effects on coated surface hardness (in 4H-5H levels). The UV exposure seems to not much influence on surface hardness properties. The similar tendency were also observed for cross-cut resistance that marginally similar adhesion (scratch resistance) for both control and UV exposed samples. Increasing coatings and charge of apricot kernel oil in varnish emulsion looks like more effective than other varnish formulations, in terms of gloss stability. However, increasing adducts charges and UV radiation time, have more less similar effects on all coated and UV irradiated samples.

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1. INTRODUCTION

The natural colour of wood are impart warmth and beauty. This provides emotive comfort for humans since ancient times. However, the natural texture of the wood could be protected by surface treatments. These specific agents could be extend its useability of lifespan particularly outdoor conditions [1-4]. Many of these coating agents are petroleum-based and carry some hazardous chemicals resulting a negative environmental impact to ecosystem [5-9]. Moreover, the reduced impact on the environment are associated to type or chemical formulations of agents. In this context, the users and producers have become to avoid the use of toxic chemicals (VOCs) for wood protective applications [8,9]. Therefore, development of new technologies based on low environmental impact and sustainable procedures are emerging issue in recent years. The combined approaches for using natural and chemical components alone and/or together are becoming to be considered as outstanding environmental benefits by many scientists [7-12].

A series of studies for developing more environmentally sound wood protective agents have been conducted by Sadiki and his groups in recent years. Three *Myrtus communis* extractives were found to be effective for improving hydrophobic properties at some level [6]. However, extractives from *Thymus vulgaris* could also be considered as an alternative source of bioactive molecules for limiting and inhibited the adhesion of fungal spores on wood surface [11]. In another study on similar topic, it was speculated that the crude extracts of *Thymus vulgaris* was modify cedar wood surface's physicochemical characteristics [12]. Moreover, the mixtures of natural organic constituents with salts seem to be under encouraging experimentation while the leaching problem of inorganic salts [10]. In addition, change of wood structure through the formation of coatings and the treatment with nanomaterials are promising and might change the performance of wood preservation techniques [7,8].

Preparation of eco-friendly dyestuff for wood substrate has become important issue considering current surface agent's VOC emissions and environmental concerns. The extracts from *Laurel (Laurus nobilis L.)* with alum

and iron mordant mixtures was evaluated for preparing an environmentally sound wood stain. However, the color stability of that emulsion was found to be very effective against UV radiations [13]. Similarly, a dyestuff was prepared from *Oleander (Nerium oleander L.)* extract with in alum and iron mordant mixtures. It was reported that this emulsion was effective for Scots pine (*Pinus sylvestris L.*) and Turkish oriental beech (*Fagus orientalis Lipsky*) wood species in terms of color stability under UV light irradiation [14]. Recently, an alternative dyestuff for wood was developed by extraction from *Pomegranate skin (Punica granatum)* and *Black mulberry (Morus nigra)* with using an ultrasonic method. This dyestuff was tested under the accelerated weathering conditions for evaluating feasibility on wood substrates [15].

In the past decades, the transparent wood surface treatments have become important issue to improve their effectiveness with some adduct of natural plant extracts. Hence, a number of studies have conducted to developed more environmentally sound coating agents [6,9,10-15]. However, understanding of formulation behavior toward wood-emulsion matrix system is paramount important for achieving progress.

The aims of this study is to evaluate coating performance of natural oil adducts containing alkyd-based synthetic varnish that was applied on Rowan wood and exposed UV irradiation. In this sense, oils of apricot kernel, sesame and grape seed were added to oil modified alkyd-based synthetic varnish system, creating a new type emulsion for surface coatings. The performance of these emulsion on wood substrate were evaluated via selected experimental approaches.

2. MATERIALS AND METHODS

Rowan wood (*Sorbus torminalis*) was selected for the investigation. The timbers were cut in the small pieces (5.0x5.0x1.5 cm) from defective free sapwood parts and dried (air-dried, 12%) in laboratory conditions before being subjected to any treatments.

Commercially available oil modified alkyd-based varnish was supplied in a 5 L container. It typically has liquid polymers, antiseptics, 5-15% solvents and plasticizers. The oils from apricot kernel, sesame and grape seed are selected to be adducts into a commercially available this

varnish for evaluating their influences. All three natural oil products (apricot kernel, sesame and grape seed) were obtained from a company, produces by the cold press techniques commercially, in Isparta-Turkiye. The main chemical constituents of these products are as follows. The apricot kernel (*Prunus armeniaca*) contain oil that varies from 27.7 to 66.7%. The contents of the major fatty acids are oleic (58.3–73.4%) and linoleic (18.8–31.7%). However, the major essential amino acids (mmol/100 g) were reported to be arginine (21.7–30.5%) and leucine (16.2–21.6%), with the main non-essential amino acid was glutamic acid (49.9–68.0%) [16].

The typical chemical composition of sesame oil are; Campesterol (18-19%), stigmasterol (6-7%), β -sitosterol (59-62%), Δ^5 -avenasterol (10-11%) and γ - and δ -tocopherols [17]. The grape seed oil (*Vitis vinifera* L.) typically has unsaturated fatty acids constitute (approx. 90%) which particularly has β -sitosterol (60-70%) in the oils of all grape seed varieties [18,19].

In comparative approach, the coatings and property evaluations were performed similarly for coated samples of neat varnish (control) and prepared emulsion with adducts. Six varnish emulsion formulations were prepared to evaluate impact on selected properties. These are based on 5.0 and 10% (volume/volume) adduct of three oils in alkyd-based varnish. It was typically applied on wood samples by simply dipping in one minute of period. After end of dipping procedure, the samples were left to dry freely in the laboratory conditions. The water absorption (WA, %) properties determined with using relevant standard. The UV exposure on all coated samples was conducted in an UV chamber equipped with single UVA-340 lamp. The exposure period was 100 hours in total. The gloss measurements were carried out with a 60° Glossmeter (Pacific Scientific Glossgard II, 60° glossmeter, MI) followed in international standard ISO 2813 [20]. The surface hardness was determined by the pencil hardness (scratch resistance) test according to the standard ASTM D3363 [21]. Adherence of the coating films to the substrate was assessed by a cross-cut test. The aspect of the resulting cut network and the percentage of exfoliated coating film areas are criteria for adherence appreciation on a scale from 0 (maximum) to 5 (minimum), according to the grading details in ISO 2409 [22].

While many combinations were utilized, some code number and abbreviations were established throughout the study given in Figures and Tables. These are; A: Apricot kernel oil added varnish emulsion, G: Grape seed oil added varnish emulsion, S: Sesame oil added varnish emulsion; C: control, 5.0 and 10 are oil concentration in alkyd-based synthetic varnish emulsion (by volume/volume), I, II, III: one-, two- and three-times coated wood samples.

3. RESULTS AND DISCUSSIONS

Table 1 shows the maximum equilibrium water sorption (WA, %) of Rowan woods which treated in various emulsion formulations, prepared with oils of apricot kernel (A), grape seed (G), and sesame (S) at two concentrations (5.0% and 10%, v/v). All adducts-containing varnishes significantly inhibited water intake when compared with the control varnish. These could be expected, considering the varnish emulsion could relatively effect surface hydrophobic properties, which results are in lowering sorption into woods. In the case of coating numbers, increasing coatings more than two-times have found to be only marginally effective in most cases for all formulations. Similarly, increasing all A, G and S concentrations from 5.0% to 10% in emulsion have also limited effects against water sorptions. The lowest water sorption values of 14.9% and 11.8% were obtained with samples of 10S_I and 10S_{II} in one- and two-time coatings while 11.3% for sample of 5S_{III} in three-time coatings. These values are approximately 35.5%, 35.2% and 39.4% lower than control (neat varnish treated) samples, respectively.

Fig. 1 shows the water sorption differences (Δ ,%) of varnish emulsion coated samples with comparison only neat varnish coated control samples (C_I, C_{II} and C_{III}). It can be clearly seen that a more pronounced positive effect was observed for the sesame oil-alkyd varnish coatings. It has already well proposed that when water contact with wood, it typically caused a pressure (force) on cell wall constituents for binding free hydroxyl. But when these bonding sites covered and/or modified by a hydrophobic substances like hydrophobic agents, the sorption could be reduced at certain level. In this study, it is realized, the sesame oil-alkyd varnish emulsions imparts higher hydrophobic character or resistance against water intake, resulting decrease in the sorption and stability of coatings.

For further evaluating adducts and level of coatings effects on water sorption properties, the measured values were compared with only one time neat varnish coated sample (C_I) (Fig. 2). It was found that all three adducts in alkyd-based varnish are very effective for sorption, while the lowering sorption of all coatings are evident regardless of treatment conditions. It was found to be in range of -21.9% (5A_I) to -36.1% (10S_I) in one-time, -25.8% (10G_{II}) to -49.4% (10S_{II}) in two-times-, and -30.9% (10G_{III}) to -40.3% (5G_{III}) in three-times coatings, according to control (C_I). It is clear that coating numbers and oil additives are both positively effects on lowering water intake in Rowan woods.

The surface hardness properties were comparatively evaluated according to the pencil hardness method (Table 2). Results

clearly showed that the all prepared varnish emulsions had one and two point lowering effects in terms of coated surface hardness (4H-5H levels). However, when same wood samples irradiated under UV radiations, the surface hardness values are only marginally changed and may not be considered any changes occur. These results are very surprising considering ageing effects of UV exposure on wood surfaces. Although it is necessary to expect a more pronounced reduction of the surface hardness properties after UV exposure and in the case of many literature reports [9,13,14], the opposite results observed in this study. It may be speculated that the selected UV irradiation time (100 hours) may not effects on surface coatings while further UV exposure could be need to observe considerably hardness changes.

Table 1. Water sorption (WA, %) properties of Rowan wood samples

| Emulsion | I (coating) | II (coatings) | III (coatings) |
|--|-------------|---------------|----------------|
| Control | 23.1 | 18.2 | 18.8 |
| Adducts concentration in varnish (5.0%) | | | |
| 5A | 18.2 | 15.4 | 11.9 |
| 5G | 16.2 | 16.7 | 13.8 |
| 5S | 18.1 | 11.9 | 11.3 |
| Adducts concentration in varnish (10.0%) | | | |
| 10A | 17.7 | 12.9 | 12.4 |
| 10G | 18.2 | 17.4 | 16.1 |
| 10S | 14.9 | 11.8 | 12.2 |

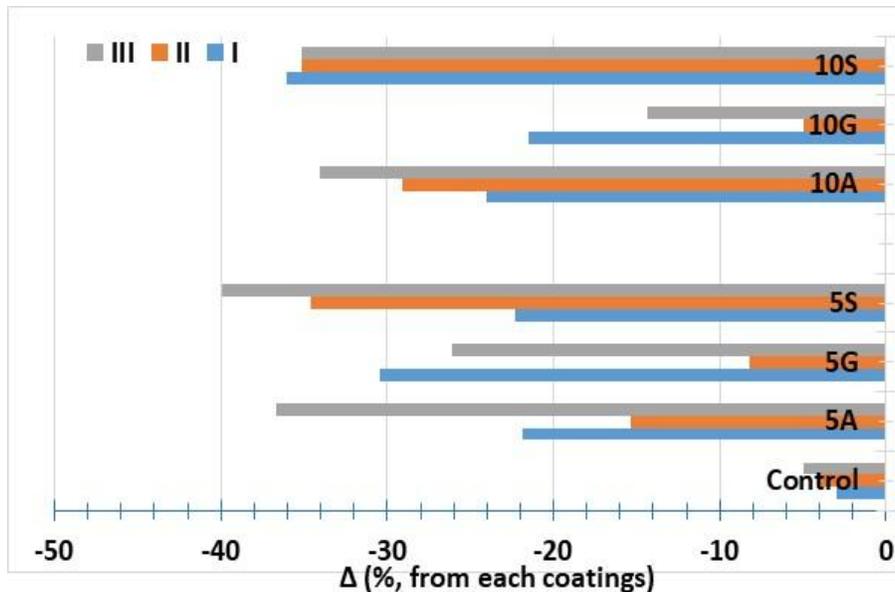


Fig. 1. Water sorption properties of samples (Δ % from C_I, C_{II} and C_{III})

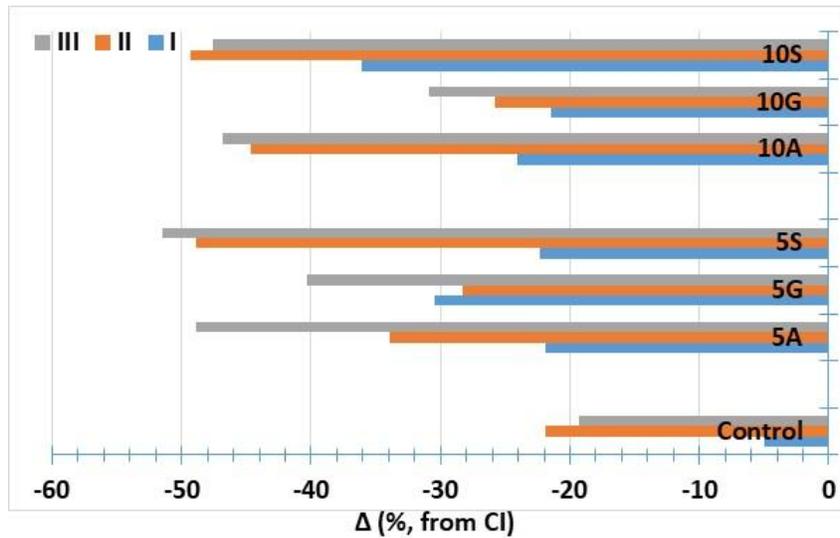


Fig. 2. Water sorption properties of samples (Δ % from C1)

Table 2. Surface hardness (pencil hardness) properties of samples

| | Control | | | UV exposure (100 hours) | | |
|-----|-------------|---------------|----------------|-------------------------|---------------|----------------|
| | I (coating) | II (coatings) | III (coatings) | I (coating) | II (coatings) | III (coatings) |
| C | 6H | 6H | 6H | 6H | 6H | 6H |
| 5G | 5H | 5H | 5H | 6H | 6H | 6H |
| 5S | 5H | 5H | 4H | 5H | 5H | 4H |
| 5A | 4H | 4H | 4H | 4H | 4H | 4H |
| 10G | 5H | 5H | 5H | 6H | 6H | 5H |
| 10S | 4H | 4H | 5H | 4H | 5H | 4H |
| 10A | 4H | 4H | 4H | 4H | 4H | 4H |

One of the important factors affecting the stability of a coating is the bonding force between the molecules of agent (cohesion) and the wood surface (adhesion). In this regards, the surface coating layer stability or adhesion properties were determined by creating a cut network (cross-cut) procedure and results (assessment are given ranks in numbers) are presented in Table 3. Interestingly, like surface hardness, marginally similar surface adhesion properties between varnish formulations and wood surfaces were observed for both control and UV exposure samples. However, the wood surfaces were showed lower adhesion initially (ranked 4-5) and UV irradiation typically not much effect on adhesion, regardless of type of varnish formulations or coating procedures. It appears that these oil adducts into alkyd-based varnish emulsion only one level (increase

or decrease) influence on surface cross-hatch properties. It is also important to note that there is no correlations found between type of coating formulations and UV ageing.

However, the surface agent-wood interactions (i.e. dye, varnish, lacquer) are very complex phenomenon and numerous variables effects on that context. The different mechanisms should control the surface adhesions. It could be suggested that the modification of alkyd varnish system with selected oil adducts has no visible improvement effects on coating layer adhesion properties.

In order to evaluate the surface hardness (pencil hardness) and adhesion properties (cross-cut) together with control and UV exposed samples, the measured values were plotted (Fig. 3). More less similar plot shapes were observed for both

control (non-UV irradiated) and UV exposed samples. However, it is realized that UV exposed samples showed higher hardness initially and further coatings (three-times) had lowering effects not only for UV exposed samples but also controls. One could be suggested that up to two-times coatings resulted in positive effects on both control and UV irradiated samples and further treatments has even lowering effects for both hardness and adhesion some level. Although typically wood surfaces like each other, but in detail, many phenomenological properties were reported for wood substrates and the quantification of all those are very complicated and need further chemical investigations [9].

Amount of light reflected from the surface could be measured numerically with the help of special devices (Glossmeter) and materials can be classified accordingly [23]. The measured numerical value is determined as glossiness

value (GU). For evaluating UV radiations and coating variables affecting on surface gloss properties, finding gloss values were plotted; as a correlation between coating numbers and oil adduct concentrations (Fig. 4a), UV irradiation time and coating numbers (Fig. 4b), and UV irradiation time and oil adduct concentrations (Fig. 4c), respectively.

It could be seen that increasing coatings and concentration of apricot kernel oil in varnish emulsion looks like more effective than other formulations, in terms of gloss (Fig. 4a). In contrast, increasing UV irradiation time and sesame oil concentration seemed to be more effective for getting higher gloss values (Fig. 4b). Interestingly, increasing oil content (G, S and A) and UV radiation time have more less similar effects on all samples, regardless of coating formulations (Fig. 4c) while apricot kernel oil-varnish emulsion treated samples show 1-2 points lower gloss values (GU) than others.

Table 3. Cross-cut properties of wood samples

| | Control | | | UV exposure (100 hours) | | |
|-----|--------------|---------------|----------------|-------------------------|---------------|----------------|
| | I (coatings) | II (coatings) | III (coatings) | I (coatings) | II (coatings) | III (coatings) |
| C | 5 | 4 | 5 | 5 | 4 | 5 |
| 5G | 4 | 5 | 5 | 4 | 4 | 5 |
| 5S | 4 | 5 | 5 | 4 | 4 | 5 |
| 5A | 4 | 4 | 4 | 5 | 5 | 4 |
| 10G | 4 | 4 | 5 | 4 | 5 | 5 |
| 10S | 5 | 4 | 5 | 5 | 4 | 5 |
| 10A | 5 | 4 | 4 | 4 | 4 | 5 |

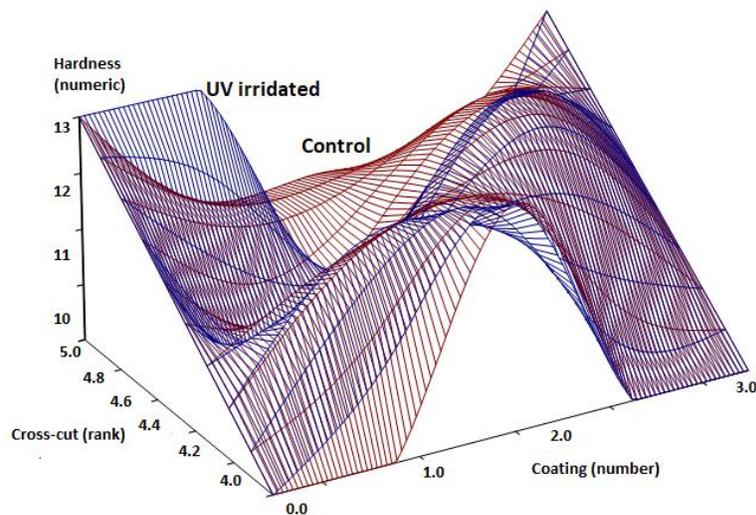


Fig. 3. UV radiations effects on hardness and adhesion properties of woods

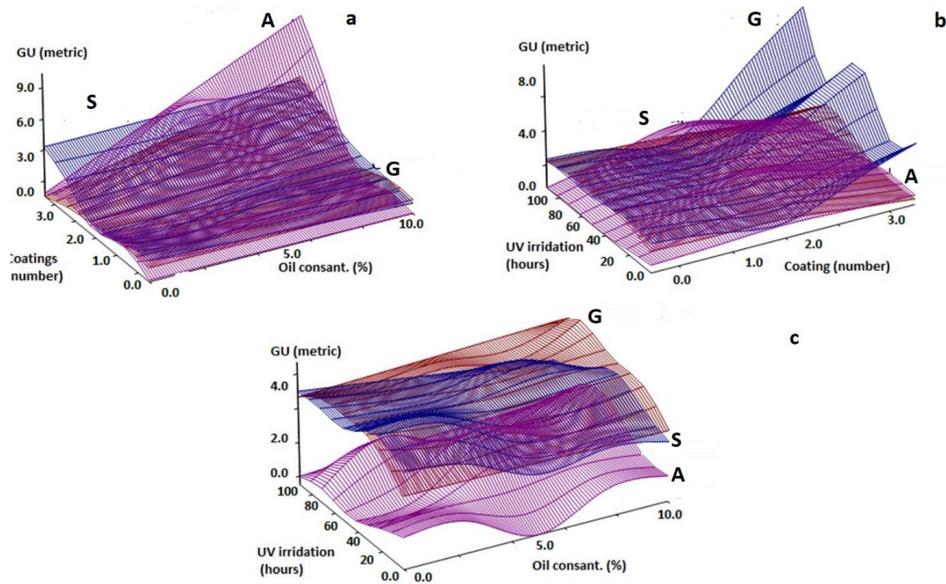


Fig. 4. UV radiations effects on gloss properties of woods (A,G, S: apricot kernel-, grape seed- and sesame oil added varnish emulsion; a: coating number and oil concentration, b: UV irradiation time and coating numbers, c: UV irradiation time and oil concentration)

4. CONCLUSIONS

Wood-based material's service life can be increased by application of a top hydrophobic layer coatings, which is in line with numerous previous literature reports. The apricot kernel oil, sesame oil and grape seed oil adducts were dispersed into the neat formulation in alkyd-based synthetic varnish at loadings of 5.0 and 10% (by volume). In comparison to the control samples (neat varnish), the presence of adducts clearly impact on water absorption of Rowan woods. All adducts were found to impart hydrophobic properties probably due to hindering the mobility of macromolecular chains at the interface around the particles. However, there is no clear improvement were found for surface hardness and scratch resistance (cross-cut) properties in both UV-irradiated and non-irradiated samples. Depends on experimental variables, increasing coating number and apricot kernel oil concentration in varnish emulsion looks like some level more effective than other varnish formulations, in terms of gloss properties.

These selected properties are of great importance in the woodworking industry where coating surfaces have to retain good properties for many years. Although evaluation of a wood-coating agent interactions is quite complex phenomenon and need further experiments, the

data obtained in this study are important in terms of providing a basis and guiding in similar studies in future.

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COMPETING INTERESTS

Author has declared that no competing interests exist.

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