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THE USE OF HERBAL EXTRACTS IN LACQUER PAINT COLORING AND DETERMINATION OF SOME MECHANICAL RESISTANCE PROPERTIES ON WOOD-BASED SURFACES

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ABSTRACT

The aim of this study was to determine the usability of selected natural dye extracts as environmentally friendly colorants that are used in painting of MDF based furniture and evaluate their resistance to scratch resistance, adhesion resistance, and surface hardness properties. In this study, the water-based lacquer coatings were prepared with natural dyes obtained from purple cabbage (*Brassica oleracea*), safflower (*Carthamus tinctorius*), Red beetroot (*Beta vulgaris*) and three synthetic paints that were black, blue and light blue as a comparison. Coated MDF test panels used for evaluation of performances of dyes to abiotic factors that are determined by mechanical tests such as scratch resistance, adhesion resistance, and surface hardness. As a result of the study, it has been observed that, except for scratch resistance, natural dyes perform as well as synthetic ones as alternative colorants in the lacquer coatings. Thus, natural and aesthetic raw materials that are environmentally friendly dyes can be used safely especially in children's furniture and for wood-based products that are especially used indoors.

Keywords: Adhesion resistance, lacquer, natural dye, purple cabbage, Red beetroot, safflower, scratch resistance, surface hardness, MDF.

INTRODUCTION

For the purpose of aesthetics and protection, many synthetic sta-ins, coatings, and paints are used as furniture finishes. Especially, lacquer paint is popular and have some production advantages in the child room furniture finishing and manufacturing. However, these synthetic paints and preservatives contain "Volatile Organic Compounds" (VOCs) (WHO 1987, Ucgun *et al.* 1998, Salthammer *et al.* 2002).

VOCs are organic chemicals that have a high vapor pressure at room temperature. High vapor pressure correlates with a low boiling point, which relates to the number of the sample's molecules in the surrounding air, a trait known as volatility (Koppmann 2007). Products defined as VOCs are generally composed of typical solvents and they have been counted among the most important chemicals ruining the indoor air quality.

Lacquer coated furniture and other colored wood-based products are among the largest sources of VOC's (WHO 1987, Ucgun *et al.* 1998). It is caused by the use of the dangerous pollutants such as ethylbenzene, glycol ethers, methanol, methyl ketone, methyl isobutyl ketone, toluene and xylene to dissolve these paints (Mandiracioglu *et al.* 2011, Singh and Chauhan 2016).

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Indoor air pollution by VOCs, which may cause a hazardous influence on human being such as sick building (sick house) syndrome, has become a serious problem (Tanaka *et al.* 2011, Akkus *et al*. 2019). These compounds can cause a variety of health effects, including eye, nose, and throat irritation; headaches and loss of coordination; nausea; and damage to the liver, kidneys, or central nervous system. Some VOCs are suspected or proven carcinogens (MPCA, 2023).

Long-term exposure to contaminated air, even if only a few parts per million (ppm) of VOCs, can result in long-term health problems (Tang *et al.* 2006). These solvents have been linked to miscarriages in pregnant women, birth deformities, and learning problems in children (Sharon 2021, Lullaby-Paints 2021). Children breathe twice as quickly as adults, and take in more air relative to their body weight. Their respiratory tracks are more permeable and thus more vulnerable. Their immune systems are weaker. Their brains are still developing (UNICEF, 2016).

As a result, rising VOCs emissions, as well as their impact on air quality, are now considered a serious environmental problem. This "going green" trend affects not just individuals' personal lives, but also large institutions such as eco-conscious businesses and cities. Consequently, the need of protecting wood materials from natural sources is growing by the day (Salthammer *et al.* 2002, Bechtold *et al.* 2007).

Natural dyes are gaining popularity across the world as people become more aware of their therapeutic and medical capabilities and the benefits they provide, as well as the known high toxicity of synthetic colorants. In recent years, several efforts have been done related to diminishment and control the indoor air pollution that caused by furniture and wood-based products paints.

One alternative to approach this problem is using natural dyes. Because natural color pigments derived from flora and wildlife are non-toxic, non-carcinogenic, and biodegradable; thus, they are considered to be safe. Some research has been done on developing environmentally friendly wood stains made from plants with natural dyes. For this purpose, Goktas *et al.* (2008a) treated wood material with laurel (*Laurus nobilis*), extracts and reported that good color stability were obtained in UV weathering tests.

Another study found that wood samples treated with walnut shell (*Juglans regia*) colorants had increased fungal resistance and showed good color resistance in UV weathering tests (Goktas *et al.* 2008b). In two separate studies; it was determined that Madder root (*Rubia tinctorium*), (Goktas *et al.* 2009a, Oleander (*Nerium oleander*) (Goktas *et al.* 2009b), extracts were applied to wood samples and UV weathering tests were performed, colored samples were showed less color change compared to the control group.

Yeniocak *et al.* (2015) investigated red beetroot (*Beta vulgaris*) for suitability with wood products, color stability on wooden samples, and reported that beetroot extracts may be used for indoor applications and toys. Goktas *et al.* (2015) investigated the leachability of walnut husk (*Juglans regia*) on wood substances and found that mordanted walnut husk extracts reduced desorption in leaching.

Currently, there is limited information about the plant-based colorants in lacquer coating and about theirs scratch resistance, adhesion strength and surface hardness properties. For this purpose, as a natural dye source Red beetroot (*Beta vulgaris*), safflower (*Carthamus tintorius*), and purple cabbage (*Brassica oleracea*) were chosen. In this study, experiment was carried out by comparing the efficiency of mechanical properties of natural and synthetic colorants that were applied on Medium Density Fiberboard MDF panels.

MATERIAL AND METHODS

MDF

In the study, E1 quality MDF (Çamsan, Türkiye) that is widely used for furniture production was used as a substrate. MDF samples for all tests were prepared in dimensions of 100 mm x 100 mm x 10 mm.

Natural colorants

Red beetroot

Red beetroot (*Beta vulgaris* L.) is a plant species belonging to the Amaranthaceae family and is among the 10 vegetables with high antioxidant effects. The antioxidant effect in red beet is due to the phenolic components and especially the betalains. Betalains are water-soluble and nitrogen-containing color pigments that give red beet its distinctive color. They are divided into two main groups, betacyanins and betaxanthins.

Betacyanins have red - purple color pigments, while betaxanthins have yellow - orange color pigments. The reason for this difference is due to the different chemical structures and ways of formation. Due to their high antioxidant effect, they play an effective role in the retention of free radicals and the prevention of diseases related to oxidative stress. In addition, betalains can also be used for food colorings owing to the fact that they are suitable for use in a wide pH range (3 - 7) and they can provide the desired color naturally (Ayan 2018). Natural colorants as Red beetroot was procured from local market from Mugla, Türkiye.

Safflower

Safflower (*Carthamus tintorius* L.) is an annual oil plant also known as false saffron, parrot food, or painter's safflower, is an herbaceous plant belonging to the genus Carthamus. Safflower, which can be 60 cm to 70 cm tall, blooms yellow, red and orange flowers in July and September, depending on the variety (Karadag 2007). It contains a dyestuff called Carthamin, which used as herbal medicine, food, paint, varnishes, and cosmetics industries (Yeniocak 2013). Natural colorants as safflower was procured from local market from Mugla, Türkiye.

Purple cabbage

Purple cabbage (*Brassica oleracea*), is a cabbage variety from the cruciferous family with large and thick layers of leaves, grown as an autumn vegetable, and its leaves are red and purple. It usually turns blue when cooked. In order to preserve its red color, vinegar or an acidic fruit is placed in it. Needing enough moisture and fertilized soil, red cabbage is a seasonal plant. It is planted in the spring and harvested in the fall. Purple cabbage is good for health as well as a suitable food colorant. Purple cabbage anthocyanins exhibit a wide spectrum of colors, ranging from red to blue depending on the pH of the medium (Ahmadiani *et al*. 2014). Natural colorants as Purple cabbage was procured from local market from Mugla, Türkiye.

Synthetic colorants

For this study, we also used three synthetic colorants for the comparison. One of black acrylic paint (Cassati Co.) and the others blue and light blue synthetic paint (İzosan Co.). Natural colorants were used for coloration of a synthetic white water-based lacquer paint (Hydrolack-Dewilux DYO Co., Türkiye) for top surface coating.

Mordant agents

Mordant agents were ferrous sulphate ($Fe_2(SO_4)_3.7H_2O$ (technical grade 96 percent purity (Merck)), aluminum sulphate ($Al_2(SO_4)_3.18H_2O$ (puriss. p.a. Fluka / Kimetsan Co.) and grape vinegar (CH_3COOH / Fersan Co).

Preparation of dyestuff

The preparation of the dyestuff was carried out according to the optimum conditions during the experimental process by authors. A weighed amount of dry plant material was extracted with distilled water in a pot with boiling method. In the standard procedure the mass ratio of plant material to the volume of liquid was 1:10 g (plant/water); extraction was performed for approximately 60 min at 100 °C. Due to the high liquor ratio, manual stirring during the extraction time was adequate to spread the plant material throughout the liquid. To compensate for volume loss due to evaporation, water was added at the end of the extraction phase to restore the original volume.

Application conditions of colorants and ratios are given in Table 1. Aqueous solutions were mordaned with 3 % ferrous sulphate, 5 % aluminum sulphate and 10 % grape vinegar to stabilize the color of extracted dyes, guarantee that they stuck to the applied substrate (to enhance retention quantity), and generate color options (Figure 1).

Table 1: Application ratios of dyes and mordants.

Colorants	Mordants Ratios (%)		Lacquer Paint (g)	Dye Ratio (%)
Red beetroot (*Beta vulgaris*)	Control	0	100	20
	Ferrous sulphate	3		
	Aluminum sulphate	5		
	Vinegar	10		
Safflower (*Carthamus tinctorius* L.)	Control	0		
	Ferrous sulphate	3		
	Aluminum sulphate	5		
	Vinegar	10		
Purple (*Brassica oleracea)*	Control	0		
	Ferrous sulphate	3		
	Aluminum sulphate	5		
	Vinegar	10		
Synthetic paint (black)	-	-		
Synthetic paint (light blue)	-	-		
Synthetic paint (Blue)	-	-		

Figure 1: Color options.

Dyeing test samples

Obtained natural dye solutions and synthetic colorants were mixed with water-based lacquer paint. Then mordants were added to mixtures for final application on to MDF test samples. For the comparison of mordant efficiency, some MDF samples were coated with paint that was without mordants as control. Natural dyes and mordants mixtures have been added to water-based lacquer paint then applied to MDF test samples by air pressure spray gun (Figure 2).

Figure 2: Colored test samples.

Mechanical tests

Scratch resistance test

The lacquered surface was tested in the scratch resistance test on a total of 75 painted test pieces measuring 100 mm x 100 mm x 10 mm, starting at 0,5 N and in increments of 0,5 N after 1 round of circular rotation according to the TS EN 15186 (2012) standard. The test was continued by reducing the load with a load sensitivity of 0,1 N and the scratch resistance of the painted surface was determined from Newton's value. Tests were performed under a load of between 0,5 N to 7 N, with 0,5 N increments. During the evaluation of the surface, the most significant consideration in this test is to distinguish properly between the trace generated by crushing the paint film and the trace formed by tearing the paint film.

Pendulum hardness test

The specimens that used for pendulum hardness test were conditioned for moisture content in accordance with ASTM D3924 (2019) standard. The hardness values of samples coated with both synthetic and natural dyes that characterize the painted layer's resistance to external variables were determined using a pendulum hardness measurement with the Köning method, in accordance with the principles of ASTM D4366-95 (2003). It determines the layer hardness according to the movements of the pendulum oscillating with the ball. Surfaces with a high number of oscillation readings for on the device screen are considered hard, while surfaces with less hardness give lower number of readings.

Adhesion resistance test

For determination of adhesion resistance of samples coated with both synthetic and natural dyestuff and dried samples were conditioned with 23 ºC ± 2 °C temperatures and 50 % ± 5 % relative humidity for a period of 16 hours according to ASTM D3924 (2019). In accordance with standard ASTM D4541-17 (1995), stainless steel cylinders (20 mm in diameter) were glued to the wood surfaces with double-competent epoxy glue and left to dry for 24 h under 20 °C temperature to perform a pullout according to standard. After drying, the paint surface was cut circularly over the rollers with a special scraping knife until reaching the substrate's bare/unapplied surface. The rollers were pulled out from the paint surface with a hydraulic adhesion force test device, and the resistance of the paint to separation from the lower layers was determined in the device indicator by N.

The adhesion (X) was calculated in terms of MPa using the Equation 1:

$$X = \frac{4F}{\pi \times d^2} \quad (1)$$

Where; F = Rupture force (Newton); d = Diameter of the experiment cylinder (mm)

Statistical evaluation

The measurements were analyzed with SPSS (Statistical Package for the Social Sciences) statistical soft-

ware program. Variance analysis was performed to determine the effects of the main factors and interactions. Duncan test was applied at 95 % statistical confidence level. The homogeneity groups (HG) of the experimental results were used for evaluation. Different letters in HG indicate the difference that can be considered statistically significant.

RESULTS AND DISCUSSIONS

The descriptive statistical values of the scratch resistance of natural colorants, synthetic colorants and mordants that were applied on MDF panels are given in Table 2.

According to data in Table 2, the highest scratch resistance mean value (6,72) was obtained from synthetic light blue paint and the lowest mean value (4,26) was obtained from Red beetroot with aluminum sulfate mordant samples. In general, values of scratch resistance decreased for all the three species of natural dyes and mordant mixture. In this study, the low scratch resistance of water-based natural paints is similar to the low scratch resistance of water-based synthetic paints that applied on MDF (Akkus *et al.* 2019), and water-based varnish applied on solid wood (Çakıcıer *et al.* 2011) in the literature.

The lowest values of scratch resistance could have been due to the low dry film thicknesses of the water-based paint. Previous research found that the hardness values of water-soluble varnishes were not high and therefore, the impact, abrasion, and scratch resistance values of these varnish layers were low (Sönmez *et al.* 2004, Akkus *et al*. 2019). The binding agent (resin) of the paint has a decisive effect on the mechanical properties of the coating including hardness, abrasion and scratch resistance (Veigel *et al.* 2014).

Compared to synthetic colorants, it is a normal phenomenon that the plant-based natural colorants we used in this study show lower scratch resistance due to the absence of any binder or hardener. 20 % water-based natural colorants that we mixed into the white lacquer coating layer adversely affected the scratch resistance of the top layer. Akkus *et al.* (2019) attributed the low scratch resistance values of water-based paint to the dry thickness of the film and the use of the water as the resin solvent.

It is estimated that the plant-based colorants, mixed with lacquer paints in powder form without dilution with water, will positively affect the scratch resistance. The scratch resistance values of control samples (without mordants) colored with all natural dyes were acceptable. This means that, except color options, there is no need to use mordants for enhancing the scratch resistance of samples that are colored with safflower, purple and red beetroot.

Another positive outcome is the performances of vinegar mordant that is a natural mordant. Vinegar mordant has been as effective as ferrous and aluminum sulfates. Thus, it could be claimed that vinegar can be used for color options.

Table 2: The performance of dyes and mordants to scratch resistance.

Mechanical test	Dye	Mordant	N	Mean	S.D.	X min	X max
Scratch Resistance Test	Safflower	C	5	$5,10^{ef}$	0,1	4,9	5,9
		FS	5	$5,16^{ef}$	0,1	5,1	5,2
		AS	5	$5,08^{ef}$	0,2	4,9	5,4
		V	5	$6,04^{b}$	0,1	5,9	6,2
	Purple cabbage	C	5	$5,38^{cde}$	0,1	5,2	5,5
		FS	5	$5,22^{e}$	0,6	4,4	5,9
		AS	5	$5,60^{cd}$	0,5	4,8	6
		V	5	$5,32^{de}$	0,2	5,2	5,6
	Red beetroot	C	5	$4,34^{h}$	0,3	3,8	4,6
		FS	5	$4,84^{fg}$	0,1	4,7	4,9
		AS	5	$4,26^{h}$	0,3	4,1	4,8
		V	5	$4,60^{gh}$	0,1	4,5	4,7
	SP (black)	-	5	$4,38^{c}$	0,5	3,5	4,8
	SP (light blue)	-	5	$6,72^{a}$	0,1	6,7	6,8
	SP (Blue)	-	5	$5,70^{bc}$	0,1	5,6	5,8

C: Control, FS: Ferrous sulphate, As: Aluminum sulphate, V: Vinegar, SP: Synthetic Paint

a,b,c,d,e,f values having the same letter are not significantly different and vice versa (for Duncan test).

SD: Standard deviation, X min: Minimum value, X max: Maximum value. ANOVA: Significance level $p<0,05$.

The performances specimens that were coated with natural dyestuffs and synthetic paints in pendulum hardness tests given in Table 3. The highest value for pendulum hardness was obtained from synthetic paint (black 48,6). In this study, the low scratch resistance of water-based natural paints is similar to the low surface hardness of water-based synthetic varnish applied on solid wood (Çakıcıer *et al.* 2011) in the literature. The lowest values of surface hardness could have been due to the low dry film thicknesses (Erdinler *et al.* 2019) of the water-based paint. Previous research found that the hardness values of water-soluble varnishes were not high and therefore, the impact, abrasion, and scratch resistance values of these varnish layers were low (Sönmez *et al.* 2004, Akkus *et al.* 2019). The binding agent (resin) of the paint has a decisive effect on the mechanical properties of the coating including hardness, abrasion and scratch resistance (Veigel *et al.* 2014). This situation is considered to be related to the binders in the paint. However, the fact that the lowest hardness value was obtained from light blue paint (29,2) seemed to be a contradiction. This situation may have occurred because of dilution conditions of the synthetic light blue paint.

Table 3: The performance of dyes and mordants to pendulum hardness.

Mechanical test	Dye	Mordant	N	Mean	S.D.	X min	X max
Pendulum Hardness	Safflower	C	5	$33,8^{cde}$	0,45	33	34
		FS	5	$35,8^{b}$	1	35	37
		AS	5	$34,6^{bcd}$	0,71	34	36
		V	5	$33,2^{def}$	2,17	31	36
	Purple	C	5	35^{bcd}	1	34	36
		FS	5	$32,6^{def}$	2,05	31	35
		AS	5	$35,6^{b}$	2,17	33	38
		V	5	$33,2^{ef}$	0,55	32	33
	Red beetroot	C	5	35^{bcd}	0,84	34	36
		FS	5	36^{a}	0,55	35	36
		AS	5	$31,8^{f}$	2,05	30	35
		V	5	$34,8^{bcd}$	0,89	34	36
	SP (black)	-	5	$48,6^{a}$	0,55	48	49
	SP (light blue)	-	5	$29,2^{g}$	2,77	25	32
	SP (Blue)	-	5	35^{bcd}	0,71	34	36

C: Control, FS: Ferrous sulphate, As: Aluminum sulphate, V: Vinegar, SP: Synthetic Paint

a,b,c,d,e,f values having the same letter are not significantly different and vice versa (for Duncan test).

SD: Standard deviation, X min: Minimum value, X max: Maximum value. ANOVA: Significance level $p<0,05$.

The mean, adhesion resistance test values of specimens that were colored by three types of synthetic paints and natural dyes and mordant mixes are shown in Table 4.

Table 4: The performance of dyes and mordants to adhesion resistance (MPa).

Mechanical tests	Dye	Mordant	N	Mean	S.D.	X min	X max
Adhesion resistance	Safflower	C	5	$0,0095^{a}$	0,37	0.0077	0,0107
		FS	5	$0,0069^{abcd}$	0,66	0,0037	0,0092
		AS	5	$0,0068^{e}$	0,62	0,0047	0,0087
		V	5	$0,0074^{bcd}$	0,38	0,0059	0,0087
	Purple	C	5	$0,0082^{abc}$	0,17	0,0076	0,0089
		FS	5	$0,0087^{abc}$	0,34	0,0069	0,0099
		AS	5	$0,0065^{de}$	0,18	0,0058	0,0071
		V	5	$0,0091^{ab}$	0,17	0,0083	0,0096
	Red beetroot	C	5	$0,0087^{abc}$	0,16	0,0081	0,0093
		FS	5	$0,0079^{abcd}$	0,58	0,0052	0,0099
		AS	5	$0,0057^{e}$	0,11	0,0055	0,0063
		V	5	$0,0074^{bcd}$	0,69	0,0042	0,0095
	SP (black)	-	5	$0,0076^{bcd}$	0,61	0,0044	0,0094
	SP (light blue)	-	5	$0,0081^{abcd}$	0,34	0,0070	0,0098
	SP (Blue)	-	5	$0,0091^{ab}$	0,25	0,0083	0,0103

C: Control, FS: Ferrous sulphate, As: Aluminum sulphate, V: Vinegar, SP: Synthetic Paint

a,b,c,d,e, values having the same letter are not significantly different and vice versa (for Duncan test).

SD: Standard deviation, X min: Minimum value, X max: Maximum value. ANOVA: Significance level $p<0,05$.

According to data in Table 4, the highest adhesion resistance mean value (0,0095) was obtained from safflower's control (without mordant) samples and the lowest mean value (0,0057) was obtained from red beetroot with aluminum sulfate mordant samples. In this study, the low adhesion resistance of water-based natural paints is similar to the low adhesion resistance of water-based paints that applied on MDF in the literature (Akkus *et al.* (2019). However, our adhesion strength results are lower than those that obtained with water-based paints obtained by Akkus *et al*. (2019). Compared to synthetic colorants, it is a normal phenomenon that the plant-based natural colorants we used in this study show lower adhesion resistance due to the absence of any binder or hardener. Because the 20 % water-based natural colorants that we mixed into the white lacquer coating layer adversely affected the adhesion resistance of the top layer. It is reported that increasing the dry film thickness improves the surface properties (Erdinler *et al.* 2019). Akkus *et al.* (2019) attributed the low adhesion resistance values of water-based paint to the dry thickness of the film and the use of the water as the resin solvent. It is estimated that the plant-based colorants, mixed with lacquer paints in powder form without dilution with water, will positively affect the adhesion resistance. In our study, the adhesion strength of the test samples that were without mordant (control) were generally higher than the samples containing mordants. These results are important in showing that we do not necessarily have to use mordants such as iron sulfate, aluminum sulfate and vinegar when using natural colorants for lacquer coating. However, using metal mordants as ferrous sulfate and aluminum sulfate and natural mordant as vinegar are also preferable in terms of color variety.

CONCLUSIONS

The effects of natural colorants and mordants in lacquer coating for MDF on the surface scratch resistance, pendulum hardness and adhesion resistances were investigated. Safflower, Purple and Red beetroot were used as natural colorants and black, blue and light blue as synthetic paints were used as synthetic colorants. The findings of this work are useful in furniture manufacturing for selecting the environmentally friendly colorants for lacquered products. The specific conclusions of this study are presented as follows:

In general, scratch resistance and surface hardness decreased for all the three species of natural dyes and mordants mixture. It is estimated that the most important reason for these results was the dilution of natural colorants with water. One of the positive outcome is the performances of vinegar mordant that is a natural mordant. Vinegar mordant has been as effective as ferrous and aluminum sulfates. Generally, except color options, there is no need to use mordants for enhancing the mechanical properties of lacquer coatings are colored with safflower, purple and red beetroot as natural plant-based colorants.

For further studies, it is estimated that, if the plant-based colorants can be mixed with lacquer paints in powder form that are without dilution with water, they will positively affect the mechanical properties of coatings. As a result, we can recommend the plant-based colorants in lacquer coatings especially for children room and indoor furniture.

AUTHORSHIP CONTRIBUTIONS

O. G.: Conceptualization, design, methodology, investigation, formal analysis, writing - original draft, visualization; Y. T. B.: Collected and analyzed data, formal analysis, project management; M. Y.: Study design, collected and analyzed data, methodology, investigation, writing - review and editing.

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