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Research Article

DETERMINATION OF THE EFFECT ON THE COLOR HOMOGENEITY OF THE USE OF NATURAL DYE STUFF IN PAPER OBTAINED FROM RECYLED PAPER BY ACCELERATED WEATHERING

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ABSTRACT

In the event of reusing the waste paper, which was exposed various contaminants according to its place of use, the stain on it breaks the colour homogeneity. In this study, it was aimed to obtain the colour homogeneity of waste paper with the dyestuff obtained from the wild cranberry (Cornus australis L.). In the study, waste paper groups were formed as mixtures (MP) in the same proportions from offset printed magazine (OPM), newsprint (NP), copy paper (CP), unbleached paper (UP), corrugated cardboard (CB) and paper. In order to investigate the effect of dvestuff, the waste paper groups were produced under groups including the control paper without any additives (I), paper mixed with dyestuff (II) and paper with dyestuff and alum (III). The same conditions were repeated with the imported bleached paper (BP) with the purpose of controlling the hiding power of the dyestuff. The paper produced was subjected to accelerated weathering test for 5-10-25-50 and 100 hours. Wave intensity of 0.65 W/m^2 was chosen as the test condition. At each weathering period, the changes in colour, gloss and opacity values of samples were investigated. According to the results obtained, the highest colour change value was obtained in the sample group I, and the lowest total colour change value was obtained in the sample group III. In Group II, the total colour change was the highest in imported bleached paper. The increase in weathering period did not affect the opacity. During the first 5 hours of weathering, the lowest opacity value was obtained in imported bleached paper. Through weathering method, the maximum decrease in opacity was observed in group II-BP.

Keywords: Waste paper, dyestuff, mordant, accelerated weathering.

1. INTRODUCTION

In the general sense, paper is obtained from fibrous substances by means of water, without using any additives. The waste paper can be converted into pulp in order to use in paper production by turning into fibre with the use of water and a mixer. With the increase in demand for raw materials, the production costs are tried to be reduced in the paper industry as well as in every area by recycling. Considering the recycling processes in other industries, it is quite easy to recycle paper. This case significantly reduces the cost of pulp which is the most important input in

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the paper industry. For instance, it was stated that when the first-hand chemical pulp is used in paper production, the proportion of pulp cost is 95% of the total cost and that with the use of waste paper, the cost of raw materials is reduced by 35-55% which is 65-85% of the total cost (Siewert 1989).

Some disadvantages are encountered in the use of recycled fibres in addition to their economical advantages. Foremost among them is the fact that recycled paper varies depending on the user and the place of use and also contains stain or foreign matter in different qualities and proportions. Not all used paper can be used as raw material in recycling. For instance, paper used for cleaning purposes are not recycled for hygiene reasons. In addition, there may be paper, which is too dirty to be used, among the collected paper. For this reason, the usable amount of paper obtained from recycling is important. The usability ratio of recycled paper depends largely on the awareness of the users. Regardless of the condition of recycled paper, they should go through cleaning and washing operations so that they can be reused. According to the results, which Hubbe (2007) and Sahin (2013) obtained from the comprehensive compiled articles, they reached the conclusion that recycling would be more efficient, decrease would be prevented in resistance and quality paper would be obtained, if recycled paper was to be reused by having knowledge on the pulp of the recycled paper and the method, by which they were produced, and by reclassifying them accordingly.

The most important factor for being successful in recycling paper is the user. In a statistical study conducted by Berger (1997) on 43000 people living in houses in Canada, he stated that the recycling ratio increased depending on their socioeconomic status, education and income. With increasing environmental awareness, not only economic value is considered in recycling, but also the desire of people for protecting nature increases awareness in paper recycling. In the research conducted by Jung et al. (2012), they noted that office workers with environmental consciousness did not pay attention to the negative appearance of the colours on the paper produced from recycled fibres. Tilikidou and Delistavrou (2005) reported that mainly young and educated students and professionals did not find recycled paper poor quality and that they stated that using the paper produced from recycled fibres was a social responsibility in order to protect the trees. They also recommend paper factories to increase the use of recycled fibre and to raise awareness in the social media for this purpose.

The chemical content of the paper is quite different because of the additives added to the special purpose paper. Pivnenko et al. (2015) stated that in addition to hazardous substances such as clay, calcium, carbonate and starch used during paper production, the number of hazardous substances caused by dye and ink used in printing and writing was 133. In the examination conducted on the waste water of recycled paper processing mills, Rigol et al. (2002) determined that these facilities had as much toxic material in their waste water as mills producing paper and pulp. They reported that most of these substances transmitted to paper produced during recycling and because of this reason they were harmful especially in food and hygiene papers.

One of the most significant problems encountered in the paper obtained from the recycled fibres is the colour heterogeneity caused by stain and ink. In order to obtain homogeneous colour in paper, processes such as ink removal, bleaching, dyeing etc. are applied. Darlington (1989) noted that no satisfactory cleaning could be achieved due to the deep penetration of the ink on the toner-printed paper. However, it was reported that when three times-recycling process was performed in bleached Kraft pulp, 5% of sodium hydroxide used to increase whiteness and gloss ratios increased the whiteness by 5.1% and the gloss by 9.1%, compared to control samples (Sutcu and Sahin 2017). However, the fibres suffer some damages during the ink removal and bleaching process of the recycled paper. Waste water pollution caused by the use of chemical substances also arises in these processes. There are also some inconveniences in producing general-purpose paper without any processing the fibres obtained by recycling. One of these inconveniences is the colour differences that vary depending on the writing and printing properties of the waste paper used. Another way of obtaining homogeneous colour is to re-dye the

recycled fibres. Although differ ent synthetic dyestuff is used for this purpose, pollution load increases in waste water of paper mill due to the property of dyestuff. Springer (1993) stated that the dyestuff used conforming with the fibres facilitates the reduction of the dyestuff transmitting to the waste water of paper mill and the recycling of the water.

Beyond its traditional use, paper is used by being dyed in order to gain visual appeal. In addition, the writing and printing process is made by means of dyestuff. The dying of the paper dates back to the Middle Ages. Synthetic dyes began to be used in 1856 after the invention of inorganic pigments such as Prussian Blue and Ultramarine Blue. Since some dyes adhere weakly, their waste water becomes coloured so their treatment is difficult. The mordant should be used to increase adherence (Murray 19XX). Different mordant is used for this purpose and Bechhtold (2009) stated that alum had been an important mordant used for many years. Gencer and Can (2016) stated that permanence was poor when the paper was dyed with organic dyestuff obtained from the seeds of the elderberry (*Sambucus nigra* L.) plant. Gencer et al. (2017) stated that the permanence was increased by the use of alum.

In this study, the dyestuff obtained from the bark of wild cornel (*Cornus australis* L.) wood was used for colouring waste paper. The reason for preferring the cornel bark is to make use of idle bark of cornel wood that have been debarked to be used in making traditional handicrafts 'Devrek Walking Sticks' from cornel wood in Devrek. In order to sustain this handcraft, which contributes to tourism and economics of the region, Devrek Forest Management Directorate of Zonguldak Regional Directorate of Forestry grants permission to local handcraft artisans for logging in certain areas. Since a stool becomes suitable for making a walking stick in 5 to 6 years, logging is made in accordance with the Inventory and Planning of Non-Wood Forest Products and Principles of Sales prepared by General Directorate of Forestry numbered 297 and dated 04.05.2013 for sustainable production. According to this plan, a total of 91.400 stools shall be available to be cut from an area of 22 ha between 2014 and 2019. Devrek Forest Management Directorate carries out this plan successfully.

2. STUDIES

In the study, recycled paper, offset printed magazine (OPM), non-reference bleached pulp (BP), newsprint (NP), copy paper with a pencil-written text on it (CP), corrugated cardboard (CB), unbleached writing paper with a pencil-written text on it (UP) and a mixture of these paper types (MP). The images of unprocessed recycled paper and reference bleached pulp are as shown in Figure 1.

Reference bleached pulp, used paper and corrugated cardboard samples were individually torn by hand in the size of a palm and they were kept in the beakers for 25 hours by adding water. These samples were pulped by being mixed with a mixer for 10 minutes, provided that they were not exposed to any mechanical effect. The same procedure was repeated for mixed pulps and for white control pulp. The paper obtained from this pulp was classified as paper group 'I'. To obtain dyestuff, 100 g of complete dry barks were boiled in 500 ml of water for 2 hours. Test paper was obtained by adding the dyestuff to the fibre suspension in the fibre mixer. This group paper was classified as paper group 'II'. Also, paper was produced by adding 1% alum suspension to the full dry fibre weight as mordant in order to increase colour permanence. This group paper was classified as paper group 'III'.



(MP)

Figure 1. Recycled paper samples and example of reference bleached pulp.

Accelerated weathering test was carried out in the device of Q-Panel Lab Products, in the laboratory of the Industrial Forest Engineering department of Bartin University. Test parameters in the device are as follows; UV (340 nm, wavelength) at 0,65 W/m2 for 8 h and conditioning at 50 °C for 4 h. Overall, this cycle was operated for 500 h totally (ASTM-G53, 1998).

Test paper was tested for opacity according to TAPPI T 519 om-02.

Colour measurement of the test samples before and after the accelerated weathering were carried out in accordance with ISO 7724 standards by Konica Minolta CD-600 colour meter. On the wood samples, the color measurements from 3 different points were measured and their mean value was calculated for three replicates in each variation (ISO-7724, 1984).

The CIELab (Commission Interational de i'Eclairage) system consists of three variants (ISO 7724). L* refers to Light stability, a* and b* chromotographic coordinates (+a* indicates red, -a* green, +b* yellow, -b* blue). The values of L*, a* and b* were measured on the samples and the color changes were determined according to the following formulas

$$\Delta L^{*}=Lf^{*}Li^{*}$$
(3)
$$\Delta a^{*}=af^{*}-ai^{*}$$
(4)
$$\Delta b^{*}=bf^{*}-bi^{*}$$
(5)

$$\Delta \mathbf{E}^* = (\Delta \mathbf{L}^* 2 + \Delta \mathbf{a}^* 2 + \Delta \mathbf{b}^* 2)^{1/2} \tag{6}$$

One of the first signs of ageing is the gradual loss of gloss of the coating film. Gloss measurements were taken in a KONICA Minolta Multi gloss 268 plus. The angle of incidence of

the radiation was 60°, as defined in ISO 2813. Six measurements were made in each test panel, three parallel to the application direction and three perpendicularly.

3. RESULTS

The colour change values occurred in the samples exposed to 100-hour of weathering are given in Figure 1 as compared with the control samples. In addition, ΔL , Δa , Δb and ΔE values occurring on the sample surfaces are listed in Tables 1-4, respectively.

Images of recycled paper, paper produced from bleached pulp without using dyestuff and alum (paper group I), paper with added dyestuff (paper group II), paper with dyestuff and alum (paper group III) before weathering and after 100 hours of weathering are given in Figure 2.





Figure 2. Colour change occurring on the sample surface after 100 hours of weathering; OPM:offset printed magazine, BP: bleached pulp, NP: newspaper, CP: copy paper, CB: corrugated board, UP; unbleached printing paper, MP: mixed paper

When the Figure 2 is examined, obvious stains appear in some of the paper (group I) obtained from the recycled fibres. While the number and size of stains were observed to be decreased in paper produced with dyestuff (group II), it is seen that stains disappear completely in some of the paper produced with alum and dyestuff (group III). The stains and spots covered with dye and alum addition did not become visible after 100 hours UV application. This shows that the permanence of the dyestuff used for long-term in the paper production is effective. Pictures were chosen one for each in order to represent groups in tens. Since human eye is relative in colour measurement, actual results shall be determined via the measurements made with optical devices. Opacity, gloss and Lab values of the paper produced was given below with the UV times applied.

The colour change values in the test and control samples were calculated using the L* (light intensity), a* and b* chromatographic coordinates (+a red, -a green, +b yellow, -b blue) determined according to the CIELab system. According to the results obtained, with the increase in ΔL , group I increased negatively except I-CB, and groups II and III increased positively. Darkening is expected on the sample surfaces in-group I, which represents the group without dyestuff and alum. The lowest ΔL values were obtained in OPM in-group I and in NP in groups II and III. In addition, the highest ΔL values were observed in the I-CB, II-BP and III-CP sample groups. When the groups II, III, and I were compared with each other, the highest change was obtained in the III-CP samples with the value of 6.17. The ΔL values tended to be positive with the use of dyestuff and alum.

When Δa values (Table 2), which represents the red-green colour coordinates of the samples, were examined, it was negative (green) in groups I-II-III CP and CB positive (red) in the other variations. In the variations generated, while UP had the highest Δa value, I-CB, II-III BP had the lowest Δa value. Among all variations, the highest Δa was obtained in I-UP with the value of +3.97. This is due to the structure of UP. It is because of the high content of lignin in unbleached pulps (UP) and the fact that the lignin changes colour by oxidation.

			ΔL		
	5 hours	10 hours	25 hours	50 hours	100 hours
I-OPM	-1,97 (1,46)	-2,84 (1,38)	-4,20 (0,04)	-4,91 (0,14)	-5,34 (1,20)
I-BP	-0,34 (0,21)	-0,41 (0,18)	-0,47 (0,20)	-0,32 (0,30)	-0,31 (0,15)
I-NP	-1,12 (0,19)	-1,50 (0,13)	-2,17 (0,21)	-2,47 (0,17)	-2,89 (0,12)
I-CP	-0,15 (0,40)	-0,22 (0,21)	-0,29 (0,17)	-0,34 (0,17)	-0,42 (0,28)
I-CB	0,09 (0,44)	0,54 (0,83)	1,15 (0,45)	2,03 (0,57)	2,68 (0,78)
I-UP	-1,00 (0,13)	-1,59 (0,04)	-2,01 (0,13)	-2,34 (0,18)	-2,80 (0,13)
I-MP	-0,56 (0,21)	-0,71 (0,10)	-1,15 (0,23)	-1,02 (0,23)	-1,06 (0,11)
II-OPM	-1,59 (0,37)	-1,77 (0,27)	-1,69 (0,30)	-1,46 (0,26)	-1,27 (0,19)
II-BP	-0,52 (0,10)	-0,36 (0,05)	0,74 (0,14)	2,15 (0,11)	3,32 (0,11)
II-NP	-0,34 (0,14)	-0,41 (0,25)	0,13 (0,18)	0,52 (0,30)	1,03 (0,06)
II-CP	-0,56 (0,10)	-0,31 (0,42)	0,33 (0,18)	1,28 (0,12)	2,48 (0,19)
II-CB	-0,85 (0,11)	-0,35 (0,29)	0,67 (0,29)	1,47 (0,14)	2,50 (0,03)
II-UP	-0,66 (1,36)	-0,59 (1,07)	-0,76 (0,77)	-0,63 (0,92)	-0,99 (1,16)
II-MP	-0,93 (2,47)	-0,75 (2,45)	-0,24 (2,32)	0,39 (0,45)	1,01 (1,79)
III-OPM	-0,57 (0,29)	-0,61 (0,40)	0,12 (0,36)	1,18 (2,37)	1,80 (0,82)
III-BP	0,11 (0,32)	0,75 (0,51)	1,52 (0,95)	3,10 (0,96)	4,55 (0,49)
III-NP	-0,59 (0,20)	-1,08 (0,27)	-0,68 (0,64)	-0,30 (0,50)	-0,19 (0,35)
III-CP	-0,19 (0,95)	0,20 (0,13)	1,32 (1,20)	3,04 (0,77)	6,17 (0,55)
III-CB	-0,39 (0,56)	0,39 (0,29)	1,14 (0,70)	2,57 (0,43)	3,93 (0,47)
III-UP	-1,63 (0,35)	-1,53 (0,18)	-0,88 (0,29)	-0,35 (0,29)	0,31 (0,31)
III-MP	-0,76 (0,41)	-1,24 (1,23)	-1,23 (1,35)	-0,68 (0,88)	-0,03 (0,29)

Table 1. ΔL values of test and control samples

In parentheses: SD

			$\Delta \mathbf{a}$				
	5 hours	10 hours	25 hours	50 hours	100 hours		
I-OPM	-0,05 (0,19)	0,29 (0,16)	1,04 (0,03)	1,62 (0,13)	2,31 (0,19)		
I-BP	-0,22 (0,15)	-0,16 (0,12)	-0,12 (0,14)	-0,03 (0,11)	0,02 (0,11)		
I-NP	0,05 (0,06)	0,40 (0,05)	1,28 (0,06)	1,97 (0,04)	2,72 (0,09)		
I-CP	-0,55 (0,02)	-0,71 (0,04)	-0,98 (0,03)	-1,07 (0,05)	-1,09 (0,02)		
I-CB	-0,75 (0,13)	-0,97 (0,13)	-1,13 (0,26)	-1,30 (0,28)	-1,21 (0,24)		
I-UP	0,40 (0,04)	0,86 (0,04)	1,95 (0,06)	2,86 (0,11)	3,97 (0,09)		
I-MP	-0,22 (0,07)	-0,23 (0,03)	0,25 (0,16)	0,55 (0,10)	0,89 (0,04)		
II-OPM	0,75 (0,11)	1,09 (0,49)	1,11 (0,05)	1,37 (0,08)	1,50 (0,04)		
II-BP	0,54 (0,12)	0,56 (0,10)	0,07 (0,09)	-0,57 (0,01)	-1,05 (0,10)		
II-NP	0,54 (0,09)	0,84 (0,10)	1,25 (0,11)	1,46 (0,07)	1,80 (0,05)		
II-CP	0,45 (0,03)	0,32 (0,07)	-0,09 (0,10)	-0,36 (0,11)	-0,73 (0,09)		
II-CB	0,09 (0,13)	-0,11 (0,18)	-0,30 (0,07)	-0,54 (0,18)	-0,71 (0,13)		
II-UP	0,42 (0,63)	0,89 (0,54)	1,72 (0,34)	2,41 (0,51)	3,37 (0,68)		
II-MP	0,62 (0,54)	0,68 (0,55)	0,95 (0,40)	1,02 (0,18)	1,11 (0,22)		
III-OPM	0,73 (0,14)	0,96 (0,04)	0,96 (0,27)	1,10 (0,48)	1,18 (0,18)		
III-BP	0,34 (0,13)	0,18 (0,18)	0,01 (0,32)	-0,50 (0,39)	-0,95 (0,17)		
III-NP	0,77 (0,10)	1,15 (0,15)	1,22 (0,26)	1,76 (0,19)	2,19 (0,29)		
III-CP	0,47 (0,07)	0,56 (0,03)	0,51 (0,09)	0,19 (0,17)	-0,61 (0,17)		
III-CB	0,08 (0,09)	0,19 (0,18)	0,23 (0,07)	0,03 (0,18)	-0,36 (0,21)		
III-UP	1,34 (0,11)	1,84 (0,09)	2,25 (0,42)	3,03 (0,23)	3,74 (0,09)		
III-MP	0,08 (0,10)	0,26 (0,23)	0,55 (0,34)	0,77 (0,21)	1,03 (0,05)		
In parentheses: SD							

Table 2. Δa values of test and control samples

In parentheses: SD

When Δb value (Table 3), which represents the yellow-blue colour coordinates of the samples, were examined (Table 3), the lowest value was obtained in BP while the highest value was obtained in UP among all groups of I, II and III. Among all variations, the highest Δb value was obtained in I-UP with 17,30 and the lowest in III-BP with -4,41. That is to say, the tendency was observed towards yellow colour in I-UP and towards blue colour in III-BP. Since the lignin content in the unbleached pulp is high, the yellow colour tendency is increased due to the weathering effect and the oxidation occurred in the lignin. There is a minute amount of lignin in bleached pulp, because of this there is no tendency to become yellow. As the bleaching substances are actually blue in colour, tendency towards blue is increased by oxidation.

When all variations were examined, the lowest total colour change (ΔE) was observed in CB samples. The highest colour change was obtained in I-NP, II-III UP samples. Adding dyestuff and alum into the CB did not have a significant effect on total colour change. Total colour change was 3.30 in I-CB, 3.14 in II-CB and 4.37 in III-CB. This may be caused by the inadequate hiding power of the dyestuff due to the production of CBs from non-bleached pulp. This situation is supported by the fact that the colour change in II-CB and III-CB group paper is higher than the paper with non-additives (I-CB).

When the groups I, II and III were evaluated in themselves, the highest total colour change (ΔE) after 100 hours weathering process was obtained in samples numbered I, and the lowest total colour change was obtained in samples numbered II. The fact that sample group I did not have dyestuff and alum caused ΔE value to be high. While the lowest ΔE value for the first 25 hours was obtained by the use of alum, which enables the dyestuff adherence on the paper, the effect of

alum disappeared with the increase of the weathering period and ΔE values in samples numbered III became higher than those in group II.

$\Delta \mathbf{b}$								
	5 hours	10 hours	25 hours	50 hours	100 hours			
I-OPM	6,97 (0,5)	9,84 (0,28)	12,99 (0,45)	14,35 (0,10)	15,26 (0,38)			
I-BP	2,10 (0,43)	2,06 (0,47)	1,92 (0,35)	1,42 (0,43)	0,95 (0,24)			
I-NP	8,44 (0,16)	11,10 (0,08)	14,68 (0,07)	16,09 (0,22)	16,92 (0,15)			
I-CP	3,50 (0,44)	4,73 (0,21)	6,90 (0,29)	8,16 (0,42)	9,16 (0,10)			
I-CB	0,89 (0,43)	1,14 (0,19)	2,04 (0,50)	1,98 (0,28)	1,88 (0,41)			
I-UP	7,63 (0,18)	10,56 (0,09)	14,10 (0,23)	16,09 (0,39)	17,30 (0,23)			
I-MP	4,36 (0,84)	6,29 (0,69)	8,56 (0,78)	9,68 (0,79)	9,57 (0,69)			
II-OPM	4,16 (0,10)	5,49 (0,08)	7,18 (0,09)	7,87 (0,14)	7,68 (0,16)			
II-BP	0,86 (0,40)	0,84 (0,14)	0,00 (0,24)	-1,64 (0,18)	-3,81 (0,40)			
II-NP	4,67 (0,15)	6,34 (0,35)	8,44 (0,18)	8,96 (0,49)	9,60 (0,17)			
II-CP	2,10 (0,10)	2,81 (0,36)	3,12 (0,42)	2,83 (0,36)	1,74 (0,56)			
II-CB	0,76 (0,17)	1,22 (0,16)	2,04 (0,06)	1,97 (0,28)	1,76 (0,17)			
II-UP	6,17 (1,35)	8,43 (0,95)	11,46 (0,47)	12,63 (1,65)	13,66 (1,85)			
II-MP	2,65 (1,02)	3,62 (0,99)	5,00 (0,73)	5,17 (0,65)	5,06 (0,38)			
III-OPM	3,79 (0,17)	5,31 (0,08)	6,44 (0,49)	6,69 (0,65)	6,33 (0,34)			
III-BP	0,78 (0,49)	0,30 (0,70)	-0,13 (0,82)	-1,99 (1,11)	-4,41 (0,63)			
III-NP	4,52 (0,37)	6,23 (0,36)	7,87 (0,64)	9,25 (0,81)	9,95 (0,94)			
III-CP	1,25 (1,08)	1,84 (0,19)	2,40 (1,26)	2,00 (0,47)	-0,19 (0,77)			
III-CB	0,50 (0,31)	1,31 (0,18)	2,00 (0,31)	2,15 (0,42)	1,83 (0,39)			
III-UP	4,21 (0,49)	6,21 (0,28)	7,94 (0,70)	9,87 (0,48)	11,29 (0,16)			
III-MP	3,56 (0,22)	4,92 (0,35)	6,86 (0,48)	7,88 (0,23)	8,50 (0,23)			
	In paranthacas: SD							

Table 3. Δb values of test and control samples

In parentheses: SD

I-OPM 7,33 (0,69) 10,30 (0,63) 13,69 (0,44) 14 I-BP 2,14 (0,47) 2,11 (0,50) 1,99 (0,38) 1 I-NP 8,51 (0,15) 11,21 (0,09) 14,89 (0,07) 16 I-CP 3,56 (0,45) 4,79 (0,22) 6,98 (0,29) 8 I-CB 1,16 (0,35) 1,58 (0,24) 2,68 (0,46) 3 I-UP 7,71 (0,17) 10,72 (0,10) 14,37 (0,24) 16 I-MP 4,41 (0,78) 6,33 (0,17) 8,65 (0,22) 9 II-OPM 4,52 (0,20) 5,88 (0,16) 7,46 (0,04) 8 II-BP 1,15 (0,39) 1,07 (0,14) 0,78 (0,13) 2 II-NP 4,71 (0,14) 6,41 (0,35) 8,53 (0,19) 9 II-CP 2,22 (0,11) 2,87 (0,40) 3,14 (0,42) 3 II-CB 1,16 (0,13) 1,30 (0,17) 2,18 (0,09) 2	5,25 (0,12) 16 ,49 (0,42) 1 5,39 (0,19) 17 ,23 (0,43) 9 ,25 (0,41) 3 5,51 (0,43) 17 ,75 (0,37) 9	00 hours 5,35 (0,74) ,02 (0,18) 7,38 (0,15) ,24 (0,11) ,30 (0,54) 7,97 (0,24) ,67 (0,42)
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II-OPM 4,52 (0,20) 5,88 (0,16) 7,46 (0,04) 8 II-BP 1,15 (0,39) 1,07 (0,14) 0,78 (0,13) 2 II-NP 4,71 (0,14) 6,41 (0,35) 8,53 (0,19) 9 II-CP 2,22 (0,11) 2,87 (0,40) 3,14 (0,42) 3 II-CB 1,16 (0,13) 1,30 (0,17) 2,18 (0,09) 2		, , ,
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II-CP2,22 (0,11)2,87 (0,40)3,14 (0,42)3II-CB1,16 (0,13)1,30 (0,17)2,18 (0,09)2	,76 (0,14) 5	,16 (0,35)
II-CB 1,16 (0,13) 1,30 (0,17) 2,18 (0,09) 2	,10 (0,49) 9	,82 (0,16)
	,13 (0,35) 3	,15 (0,31)
II-UP 6,33 (1,50) 8,53 (1,05) 11,63 (0,56) 12	,53 (0,21) 3	,14 (0,11)
	2,89 (1,75) 14	4,14 (2,02)
II-MP 3,48 (1,52) 4,27 (1,31) 5,47 (0,84) 5	,30 (0,63) 5	,51 (0,16)
III-OPM 3,91 (0,21) 5,44 (0,06) 6,52 (0,52) 7	,18 (0,39) 6	,73 (0,27)
III-BP 0,98 (0,24) 1,12 (0,17) 1,76 (0,81) 3	,75 (1,38) 6	,41 (0,79)
III-NP 4,63 (0,37) 6,43 (0,39) 8,02 (0,66) 9	,44 (0,80) 10	0,20 (0,97)
III-CP 1,74 (0,65) 1,94 (0,19) 2,87 (1,56) 3	,71 (0,45) 6	,23 (0,59)
III-CB 0,84 (0,14) 1,41 (0,19) 2,36 (0,56) 3	,36 (0,55) 4	,37 (0,41)
III-UP 4,73 (0,42) 6,65 (0,30) 8,30 (0,80) 10		1,90 (0,15)
III-MP 3,66 (0,27) 5,20 (0,17) 7,10 (0,15) 7	0,34 (0,52) 11	,57 (0,22)

Table 4. Total colour change (ΔE) values of the test samples

In parentheses: SD

	0 hour	5 hours	10 hours	25 hours	50 hours	100 hours
I-OPM	99,02	99,03	98,87	98,86	99,21	99,14
I-BP	88,72	89,05	89,29	89,42	89,12	89,02
I-NP	99,86	99,86	99,85	99,96	99,76	99,84
I-CP	96,26	96,69	97,02	96,79	96,81	96,78
I-CB	99,31	98,71	99,05	99,97	99,72	98,91
I-UP	99,73	99,88	99,62	99,14	99,63	99,55
I-MP	99,30	99,63	99,66	99,61	99,54	99,51
II-OPM	99,42	99,66	99,87	99,96	99,88	99,76
II-BP	91,74	90,05	90,30	89,76	89,29	88,90
II-NP	99,51	100,00	99,99	99,99	99,98	99,87
II-CP	99,06	98,25	98,32	98,26	98,15	97,88
II-CB	98,48	99,66	99,82	99,69	99,51	99,41
II-UP	99,10	99,71	99,51	99,69	99,56	99,51
II-MP	99,74	99,78	99,79	99,79	99,77	99,68
III-OPM	99,72	99,51	99,42	99,78	99,56	99,88
III-BP	89,65	92,18	92,08	95,64	91,24	90,53
III-NP	99,85	99,75	99,56	99,10	99,51	99,63
III-CP	98,63	99,13	99,01	98,59	98,61	97,04
III-CB	98,59	97,90	99,54	99,39	98,61	98,78
III-UP	99,68	99,49	99,36	96,13	99,35	99,91
III-MP	99,72	99,89	99,74	99,73	99,83	99,78

Table 5. Opacity values of paper samples before and after weathering test

In parentheses: SD

The increase in weathering period did not change the opacity value. Iosip (2010) stated that if the ratio of recycled non-bleached wrapping paper and cardboard was high in the paper to be newly produced, despite the fact that its ink had been removed, it reduced the gloss and increased the number and sizes of stains. However, in the study conducted by Gulsoy and Erenturk (2017), they noted that the value of gloss increased with the addition of waste paper to virgin pulps obtained by Kraft method from pine wood. This may be due to the low ink content in recycled paper or the higher gloss value than the pulp obtained by the Kraft method. Generally, the high colour change with UV in the paper obtained from non-bleached pulps is not related to the dyestuff used but it is directly proportional to the ratio of lignin remaining in the pulp.

4. CONCLUSION

The paper, which is obtained from the colorant obtained from wild cranberry stains and dirt, were significantly reduced compared to the control samples. It was also found that some of the aluminum used samples were completely lost. After 100 hours of UV treatment the stains and spots did not become clear again. it can be said that the covering of the stain obtained in this case is sufficient.

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