RUB RESISTANCE OF MULTILAYER PRINTS ON PAPERS PRODUCED WITH ALTERNATIVE FIBER SOURCES

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Abstract

The packaging industry is a rapidly growing industry all over the world and is in constant development following the requirements for more environmentally friendly materials to replace plastic and other non-degradable materials. Paper is a material that can be safely disposed of as a waste or reused as a recycled material. The problem with paper recycling is that the cellulose fibers get shorter with each cycle and thus the properties of the recycled paper deteriorate. Therefore, a certain amount of virgin fibers must be added to the paper pulp to maintain the required quality and strength of the paper used as packaging. Alternative fiber sources, such as rapidly renewable cereal straw, can replace the use of virgin wood fibers which are in short supply and thereby reduce the concerns about deforestation. In this research, laboratory paper substrates were produced from admixture of recycled fibers and virgin cereal straw fibers. The papers produced were printed with three full tone layers of conventional process inks (cyan, magenta and yellow) printed one on top of the other using three different printing techniques (offset, gravure, and screen printing). Since the rub resistance of printed paper is one of the most important factors in determining its suitability for packaging, this study analyzed the rub resistance of multilayer prints on paper substrates with alternative fiber sources. When used as secondary packaging, paper is subjected to various circumstances under which prints can be damaged, such as transport, contact with other packaging material, or handling by the end user. Therefore, the mechanical properties of such prints must be robust and stable in contact with external influences. The quality of the prints after rub resistance tests was determined through measuring the spectrophotometric properties and calculating the color differences between the multilayer prints before and after performed test.

Keywords: alternative fiber sources, gravure, offset, rub resistance, screen printing

Introduction

The packaging and graphic industries have a constant demand for paper products, so the need for rapidly renewable sources of raw materials for papermaking is greatly increasing. Wood is still the main source of cellulose fiber[1] as the most abundant component in paper, but in recent years there has been more scientific research involving alternative sources of cellulose fiber for the same application[2]. Fiber from fast growing annual plants is being considered as a replacement for wood fiber because wood takes more time to grow, and it may be a more valuable material for other industries[3]. It is important to evaluate all possible sources of non-wood cellulose fibers for the paper industry and to obtain information on the quality of the final product such as packaging and printing paper. Recent research shows the great potential of cereal straw as a source of cellulose fibers to be added into the recycled wood pulp to produce paper with adequate qualities for printed secondary packaging[4-6]. This paper examines the rub resistance of laboratory paper substrates containing cellulose fibers obtained from straw of wheat (*Triticum spp.*), barley (*Hordeum vulgare L.*), or triticale (*Triticale sp.*). These papers were printed with three layers of water-based inks using three different printing techniques to determine the most stable printing conditions for multilayer printing. Rubbing off of printed inks is a substantial aspect of determining the printing quality which generally decreases with the amount of ink applied to the paper surface, so it is important to determine the limits of each paper and printing technique and match the compatible inks with paper substrates[7].

Experimental Part

Materials and Methods

The first stage of this research was the production of laboratory paper substrates from 100% recycled wood pulp (as a reference sample) and test samples in which the cereal straw pulp was added. Previous research has shown that the optimal amount of added straw pulp is 30% of the total pulp weight[8]. Straw from wheat, barley and triticale was collected from the Croatian fields after grain harvest, processed by purifying from grain and other residues and cutting. It was, then, converted into straw pulp by the soda method[8]. This straw pulp was added in a 7:3 ratio to the previously prepared recycled wood pulp. The paper sheets were formed in a Rapid-Köthen sheet former (FRANK-PTI) according to EN ISO 5269-2:2004 standard. A total of four types of paper substrates weighing approximately 42.5 g/m^2 were produced for further comparison and testing. Table 1 shows the abbreviations and composition of the paper substrate samples.

Table 1: Abbreviations used for marking laboratory made paper substrates

Abbreviation	Pulp composition
N	100% recycled wood pulp
N3W	70% recycled wood pulp + 30% wheat straw pulp
N3B	70% recycled wood pulp + 30% barley straw pulp
N3TR	70% recycled wood pulp + 30% triticale straw pulp

The second stage of the research was the printing on paper substrates using three different printing techniques (offset, gravure, and screen printing) with water-based printing inks which were air-dried after printing.

Offset printing was performed on a Prüfbau multipurpose printability testing machine using SunPak FSP low migration inks (manufacturer Sun Chemicals) at a speed of 0.5 m/s and a pressure of 600N. Gravure prints were obtained on a KPP Gravure laboratory device with a 65 Shore impression roller and ~40 lines/cm (100 lpi) engraving plate (RK Printcoat Instruments Ltd.) at a printing speed of 20 m/min using conventional drying inks Sunprop (Sun Chemicals). Screen printing was carried out at production level on a printing machine Semi-auto Shenzhen Juisun with 120 lines/cm and squeegee with 75 Shore hardness using Epta inks Hi-gloss (KIIAN S.p.A.).

All printing processes were performed under standardized conditions of 23 °C and 50% relative humidity. The printing was performed in three passes with three full tone layers of conventional process inks (cyan, magenta and yellow) printed on top of each other to produce the brown tone. The multilayer prints were produced according to the recommendations of the ISO 12647 standard for different types of printing techniques[9-12].

After printing and drying, the printed paper substrates were cut into round samples with a diameter of 5 cm and subjected to mechanical resistance (i.e., rub resistance) testing on a Hanatek T4 Rub and Abrasion Tester with 20, 40 and 60 circular motions at a speed of one rotation per second. The lightest pressure of 0.23 kg (0.5 lb) was applied to simulate the handling of the printed packaging while in transport or in store conditions. The rub resistance testing was performed under the recommended conditions according to the BS 3110 standard[13].

The evaluation of the obtained results was based on the color difference, ΔE_{00}*, calculated according to Equation 1[14]. The color components of the CIE L*a*b* color space were measured with an X-Rite SpectroEye spectrophotometer under D50 illumination and a 2° standard observer before and after rub resistance tests performed.

$$\Delta E_{00} = \sqrt{\left(\frac{\Delta L'}{k_L S_L}\right)^2 + \left(\frac{\Delta C'}{k_C S_C}\right)^2 + \left(\frac{\Delta H'}{k_H S_H}\right)^2 + R_T \frac{\Delta C'}{k_C S_C}\frac{\Delta H'}{k_H S_H}} \qquad (1)$$

In graphic industry, it has been established that the tolerance for evaluating acceptable print quality by color difference is a calculated value of ΔE_{00}* below 2 [15]. When the color difference reaches values greater than 2, the printed colors are considered unequal, and the color difference is visible to inexperienced observers.

To further examine the color changes of prints after 60 circular motions, the differences between the individual *L*, a** and *b** color components were calculated according to the following equations 2:

$$\begin{aligned} \Delta L^* &= L^*_{60} - L^*_0, \\ \Delta a^* &= a^*_{60} - a^*_0, \\ \Delta b^* &= b^*_{60} - b^*_0 \end{aligned} \quad (2)$$

where ΔL^* defines the difference between the measured lightness after the rub resistance test L_{60}^* and original lightness before the rub resistance test L_0^*. Value of Δa^* defines the difference between the red/green axis after the rub resistance test a_{60}^* and before the rub resistance test a_0^*, and Δb^* defines the difference between the yellow/blue axis after the rub resistance test b_{60}^* and before the rub resistance test b_0^*.

Results and discussion

To evaluate the interaction between printing inks and paper substrates, the spectrophotometric color difference ΔE_{00}^* was calculated from the values of color components measured before and after the rubbing treatment. Table 2 shows the *CIE L*a*b** average values of multilayer prints produced with three different printing techniques (offset, gravure, and screen printing) before the samples were subjected to the rub resistance tests measured on 20 samples.

Table 2: *CIE L*a*b** values of multilayer prints made using three different printing techniques measured before the rub resistance test

	L*			a*			b*		
	OFFSET								
N	40.73	±	1.25	-3.50	±	1.61	0.63	±	2.52
N3W	38.48	±	0.91	-1.94	±	1.29	-5.62	±	1.04
N3B	37.03	±	0.99	-6.61	±	1.22	-0.24	±	0.85
N3TR	37.72	±	1.33	-4.11	±	0.83	0.01	±	1.00
	GRAVURE								
N	24.40	±	2.13	12.61	±	1.25	1.46	±	2.60
N3W	24.99	±	1.37	11.53	±	1.05	0.58	±	1.92
N3B	26.30	±	1.94	12.32	±	1.00	2.70	±	2.55
N3TR	26.39	±	2.38	12.24	±	1.12	2.96	±	3.39
	SCREEN								
N	20.44	±	0.64	10.87	±	0.43	-4.04	±	0.43
N3W	21.64	±	0.84	10.77	±	0.51	-2.72	±	0.61
N3B	22.12	±	0.72	10.99	±	0.48	-2.04	±	0.49
N3TR	21.62	±	1.08	10.99	±	0.49	-2.37	±	0.49

From the L^* component, which represents the lightness of the measured color, it is visible that the offset prints (values ranging from 37.03 to 40.73) had a greater lightness than the gravure prints (values ranging from 24.40 to 26.39) or screen prints (values ranging from 20.44 to 22.12). The a^* axis which represents chromacity from red to green indicates that the offset prints had a mild inclination towards the green axis (negative values), while gravure and screen prints show a stronger tendency towards the red axis (positive values) with similar measured values. On the b^* axis that indicates yellow and blue hues, all average values gravitate around zero with the largest negative inclination in offset prints on paper substrate with wheat straw pulp (b^* = -5.62) showing a bluish hue, while gravure prints showed a slight tendency toward a yellow hue. Overall, offset multilayer prints, although having the lightest L^* values, had the most neutral gray appearance of all prints obtained by three chosen printing techniques with the values of color components a^* and b^* closer to zero. The gravure and screen prints had similar, darker, values of lightness L^*, while the largest deviations in the a^* axis were observed in direction of the red hue.

The rubbing treatment was performed in three iterations with 20, 40 and 60 rotations of the device. After each iteration cycle, spectrophotometric measurements were performed and the color difference ΔE_{00}^* was calculated to show the best mechanical stability of printing inks applied on the tested paper substrates with and without straw pulp using three different printing techniques. Figure 1 compares the ΔE_{00}^* color difference for all paper substrates and printing inks after 20 rotations on the rub resistance device.

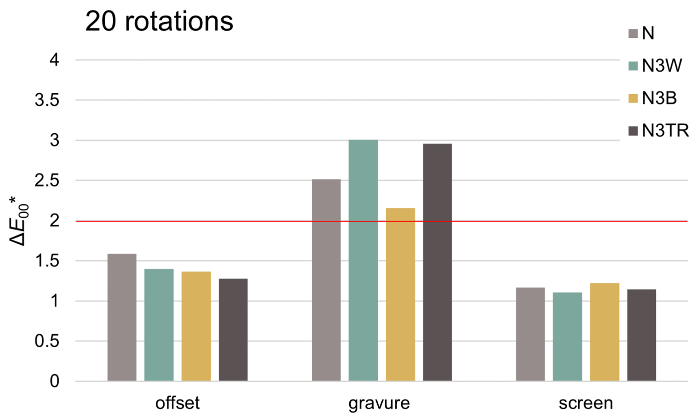


Figure 1: Comparison of color difference ΔE_{00}* of multilayer prints after 20 rotations of rub resistance test

After the first iteration, it appeared that the most stable prints with the lowest ΔE_{00}* values were obtained with the screen printing technique. Offset prints also showed a satisfactory color difference with the values of ΔE_{00}* below value of 2, which means that the untrained observer cannot perceive the difference in color. Gravure prints however, showed values of ΔE_{00}* above 2, already at 20 rotations, indicating that an untrained observer can perceive color differences, marking such prints as unstable.

When comparing the ink stability in relation to the type of paper substrate used, offset and screen multilayer prints showed similar color differences on paper substrates made only from recycled wood pulp (reference sample N) and paper substrates with added cereal straw pulp (N3W, N3B and N3TR). The type of cereal pulp used in the production of paper substrates (wheat, barley, or triticale) did not significantly affect the calculated ΔE_{00}* values. Gravure prints showed greater differences in relation to the type of cereal straw used in the paper substrate where N3B paper substrates with barley pulp showed the smallest color difference among the observed multilayer prints, but still the ΔE_{00}* value was larger than the tolerance value of 2.

Further iterations of rub resistance test with 40 rotations were performed to determine the color difference on the printed paper substrates after prolonged exposure to rubbing (Figure 2). As expected, all prints had increased ΔE_{00}* values, but also showed that the greatest effect on mechanical stability occurred during the first rubbing iteration.

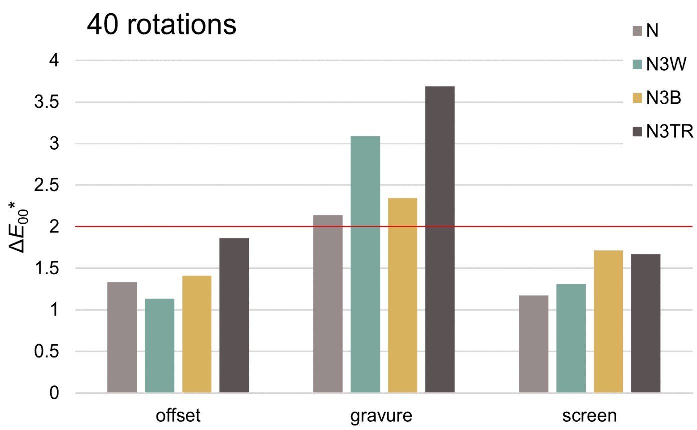


Figure 2: Comparison of color difference ΔE_{00}* of multilayer prints after 40 rotations of rub resistance test

Screen prints on papers with wheat straw pulp (N3W) showed a similar color difference as paper substrates without cereal straw pulp (N), while prints with added barley or triticale straw pulp (N3B, N3TR) showed slightly higher ΔE_{00}* values. Offset prints showed that papers with barley or triticale straw pulp had a greater color difference after 40 rotations of rub resistance test than papers without straw pulp (N) or with added wheat pulp (N3W). Gravure prints showed an even greater increase in ΔE_{00}* with the worst values (ΔE_{00}*> 3.5) on paper substrate with triticale straw pulp (N3TR), followed by prints on paper substrate with the addition of wheat pulp (ΔE_{00}* > 3).

When the rubbing tests were repeated with additional cycles on printed paper substrates (amounting to 60 rotations), the overall color difference did not triple for all multilayer prints as would be expected. Figure 3 shows the largest increase in color difference ΔE_{00}* for offset prints on paper substrates with wheat straw pulp (N3W) and screen prints with barley (N3B) or triticale (N3TR) straw pulp papers which exceeded the ΔE_{00}* value of 2. The gravure prints didn't show greater fluctuations at rubbing test with 60 rotations compared to previous rubbing iterations.

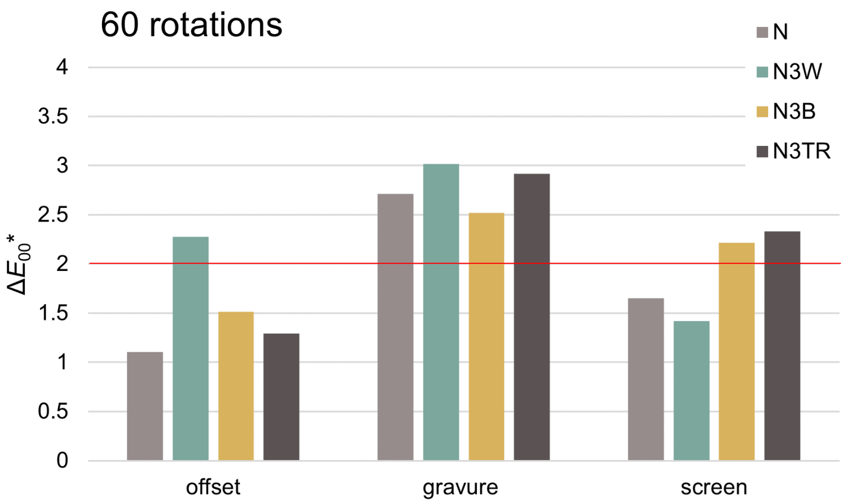
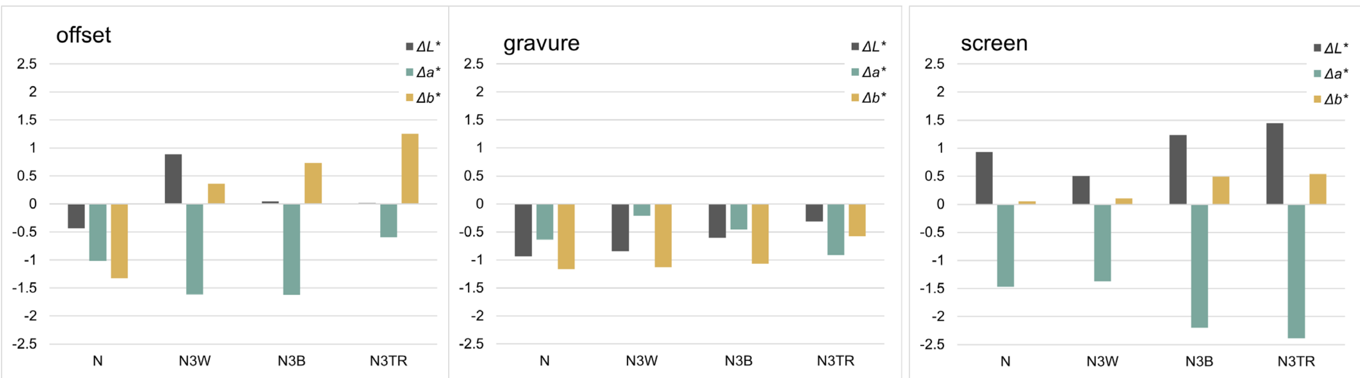


Figure 3: Comparison of color difference ΔE_{00}* of multilayer prints after 60 rotations of rub resistance test

The color change of multilayer prints was additionally examined by calculating the differences of individual *CIE L*a*b** components, which indicated in which component of the color changes occurred the most (Figure 4).



a) b) c)

Figure 4: Difference ΔL*, Δa* and Δb* before and after 60 rotations of rub resistance test for a) offset prints, b) gravure prints and c) screen prints

For offset prints (Figure 4a), minor or negligible changes were seen in the lightness *L** component, and most changes were observed in the *a** and *b** hue axis. All paper substrates, regardless of their composition, increased values in the green axis (negative Δa*). While paper substrates with added cereal straw pulp increased the yellow hue (positive Δb*), the reference paper substrate N without cereal straw pulp changed towards the blue axis (negative Δb*).

For gravure prints (Figure 4b), the calculated lightness difference ΔL* of all tested paper substrates was negative, indicating that the color became slightly darker, while the *a** component changed towards the green hue, and the b* component changed towards the blue axis.

Screen prints (Figure 4c) showed greater fluctuations in individual color components with lightness ΔL* change towards a lighter color (positive ΔL*). The value of Δa* component changed towards the green axis (negative Δa*) and the Δb* with a minor or negligible change towards the yellow axis (positive Δb*).

Conclusion

This research tested the mechanical resistance (i.e., rub resistance) properties of paper substrates containing cellulose fiber which was sourced from recycled wood pulp and alternative sources, namely pulp from wheat, barley and triticale cereal straw collected in the fields after the grain harvest. Rub resistance tests were performed in three iterations simulating packaging transportation, handling, and close contact with other packaging. The following was observed:

- The initial iteration of 20 rotations of the rub resistance test had the greatest impact on the color difference of multilayer prints, while the additional two iterations increased the color difference to a lesser extent.
- The best interaction of inks and paper substrate was observed in multilayer on screen and offset prints with the smallest calculated color difference which slightly exceeded the tolerance value only in some paper substrates after the prolonged rub resistance test of 60 rotations.
- The color difference of gravure prints already showed values above the tolerance already after the first rub resistance iteration but did not change significantly after additional second and third iterations.
- The printing technique has a great influence on the changes in the values of individual components of *CIE L*a*b**: Offset printing was affected mainly on the green axis, and less on the lightness; Gravure printing had larger variations in all three color components, and screen printing showed significant changes in the green axis, but also influenced the lightness component with the least changes in the yellow axis.

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References

1. CEPI, Key Statistics 2022, European pulp & paper industry (2023) Available online: https://www.cepi.org/wp-content/uploads/2023/07/2022-Key-Statistics-FINAL.pdf (26.7.2023)
2. M. Eugenia Eugenio, D. Ibarra, R. Martín-Sampedro, E. Espinosa, I. Bascón, and A. Rodríguez, *Cellulose. IntechOpen*, (2019) doi: 10.5772/intechopen.90041.
3. E. Hurmekoski, R. Jonsson, J. Korhonen, J. Jänis, M. Mäkinen, P. Leskinen, and L. Hetemäki. *Canadian Journal of Forest Research*. 48(12), 1417-1432 (2018) doi.org/10.1139/cjfr-2018-0116
4. M. Rudolf, I. Plazonić, K. Petric Maretić, and I. Bates, *Processes*, 11(5), 1465, (2023), doi: 10.3390/pr11051465.
5. I. Bates, I. Plazonić, V. Radić Seleš, K. Petric Maretić, and M. Rudolf, *Sustainability*, 15(6), 5378, (2023), doi: 10.3390/su15065378.
6. S. P. Bangar, P. Kajla, T. Ghosh, *International Journal of Biological Macromolecules*, 227(10), 762-776 (2022).
7. I. Bates, I. Plazonić, K. Petric Maretić, and M. Rudolf, *Hem Ind*, 77(2), 167–176, (2023)
8. I. Plazonić, I. Bates, Ž. Barbarić- Mikočević, *BioResources*, 11, 5033 – 5049 (2016)
9. ISO 12647-8 Graphic technology — Process control for the production of half-tone colour separations, proof and production prints — Part 8: Validation print processes working directly from digital data (2021)
10. ISO 12647-4 Graphic technology — Process control for the production of half-tone colour separations, proof and production prints — Part 4: Publication gravure printing (2014)
11. ISO 12647-5 Graphic technology — Process control for the manufacture of half-tone colour separations, proof and production prints — Part 5: Screen printing (2015)
12. ISO 12647-2 Graphic technology — Process control for the production of halftone colour separations, proof and production prints — Part 2: Offset lithographic processes (2013)
13. BS 3110, Methods for measuring the rub resistance of print, (2017)
14. G. Sharma, W. Wu, E.N. Dalal, *Color Res Appl*. 30(1): 21-30. (2004) https://doi.org/10.1002/col.20070
15. W.S. Mokrzycki, M. Tatol, *Mach. Graph. Vis.* 20, 383–411 (2011)