#### THE EFFECTS OF NATURAL AND ARTIFICIAL AGING ON OPTICAL PROPERTIES OF LABORATORY PAPERS WITH TRITICALE PULP PRINTED USING THE UV INKJET TECHNIQUE

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#### Abstract

The fundamental raw material in the paper industry is cellulose. Cellulose fibers are traditionally obtained from wood and are used to produce a wide range of paper substrates. The recent significant increase in the consumption of wood resources is causing a global need for alternative non-wood cellulose fibers in paper production. The straw of various cereals is often used as a source of virgin cellulose fibers. Laboratory paper substrates made of recycled paper and triticale straw pulp are used in this research. This study aimed to explore the effects of artificial and natural aging processes on the optical deterioration of digitally printed laboratory substrates with variable triticale pulp content in recycled wood pulp. The goal was to assess the optical stability of UV inkjet cyan, magenta, yellow, and black prints after two aging process treatments. The optical stability was measured by observing the reflectance in the visible spectrum. The durability of UV inkjet prints and their reproduction quality play an important role in ensuring the accuracy of text and images in graphic products. The substrates.

**Keywords:** *artificial aging, digital printing, natural aging, optical deterioration, triticale pulp, UV inkjet inks* 

### **1. INTRODUCTION**

Paper and paperboard have many applications, from labels, books, newspapers, money to packaging. There are many advantages of paper and cardboard used for packaging, such as sources of raw materials, production principles, environmental protection, and waste management. Paper is a sheet material made of a network of cellulosic fibers that self-bond and form a compact material, which has good physical properties allowing the paper to be printed. The main raw materials, cellulosic fibres, are nowadays produced using wood fibres or recovered wood fibres. The growth in the use of these raw materials reduces the share of carbon dioxide in the atmosphere, thus gradually removing material made from non-renewable sources, such as petrochemical products<sup>1</sup>. Resources of wood, as a traditional raw material in the paper industry, are insufficient today and obviously, the supply of wood for the pulp and paper industry will be even more limited in the future<sup>2</sup>.World production of pulp, paper, and board depends on about 92% of raw wood material either softwood or hardwood<sup>3</sup>. Due to the growing global demand for fibrous material, the global scarcity of forests in many areas, and the growing environmental awareness, different non-wood fibres have become one of the most important alternative sources of fibrous materials in the 21st century<sup>4</sup>.

Today, a very small percentage of paper is derived from non-wood fibers, which are also called "tree-free fiber sources".

In recent years, an increasing number of studies have sought and compared the ability of non-wood fiber alternative fibres in paper production, which are either agricultural residues or primary crops<sup>5</sup>. Alternative sources of fibrous materials have been used depending on the region in which they can be collected in large quantities. In countries such as the Netherlands, Italy, Germany, France, Spain, Greece, Hungary, and Croatia, production in agriculture is relatively high because of adequate climate and fertile soil<sup>6,7</sup>. Therefore, fibres isolated from annual crop residues are an important alternative raw

material for the pulp and paper industry due to their numerous advantages, including cost-effectiveness, abundance, availability, and reproducibility<sup>8-10</sup>.

### 2. MATERIALS AND METHODS

#### 2.1. Papers with triticale straw pulp

Laboratory paper substrates of approx. 42.5 g/m<sup>2</sup> were formed by Rapid-Köthen sheet former (FRANK PTI). Triticale pulp was obtained from triticale straw processed by the soda pulping method (120° C, 16% alkali level for 60 min, 10:1 liquid:biomass ratio)<sup>11</sup>. The paper sheet composition is made either from recycled wood fibers pulp only, or from mixture of recycled wood fibers and triticale pulp in 3 different proportions – 10%, 20% and 30% (1NTR, 2NTR, 3NTR). Paper made only from recycled wood pulp is used as a referent sample (N). Industrial recycled paper (also 42.5 g/m<sup>2</sup>) is used as a control sample (K).

#### 2.2. UV inkjet printing

Full-tone areas of cyan, magenta, yellow and black UV inks were printed on all the laboratory paper substrates. They were printed by a digital EFI Rastek H652 UV curable inkjet printer with the resolution of  $600 \times 600$  dots per inch (dpi) (respectively with high-quality mode 8 pass) and a printing speed of 12.10 m<sup>2</sup>/ h.

### 2.3. Artificial aging

Artificial aging treatment of printed UV inkjet substrates was derived in a SunTEST XLS+ test chamber by the standard method ASTM D 6789-02<sup>12</sup>. The test chamber is supplied with a xenon-arc lamp and a daylight filter which emits visible and near ultraviolet electromagnetic radiation (from 290 nm to 800 nm) with a light intensity of 765  $\pm$  50 W/m<sup>2</sup> under elevated temperature inside the test chamber up to 39.5° C (Figure 1). In the chamber the light from the xenon-arc lamp simulates natural daylight passing through windows on a printed paper surface.

All printed substrates with dimensions of 20 mm x 50 mm were treated with two cycles of 48 hours. Along with the printed substrates, the unprinted substrates were also treated in the same way.



Fig. 1. Treatment of printed substrates in a SUNTEST XSL+ test chamber

### 2.4. Natural aging

To analyze the impact of natural aging without the influence of electromagnetic radiation, the printed substrates were stored during a period of  $2 \times 365$  days (two years) side by side in a black bookcase in a dark and dry place.

### 2.5. Optical properties

The influence of two artificial aging process treatments and natural aging processes on the optical stability of UV inkjet cyan, magenta, yellow, and black printed laboratory substrates with variable

content of triticale pulp was observed on the basis of modification in spectral reflection. Reflectance spectra measurements of all observed substrates were processed using a Spectroeye, X-rite spectrophotometer with standard illuminate D50 and 2° observer, in the interval of the wavelengths from 400 nm to 700 nm for every 10 nm. The reflectance values (R) were determined before and after aging processes and repeated 10 times on each substrate.

## 3. RESULTS

The measured reflection values of the printed substrates without (K, N) and with 10%, 20%, and 30% triticale pulp (1NTR, 2NTR, 3NTR) before the aging treatment are presented in Figure 2.

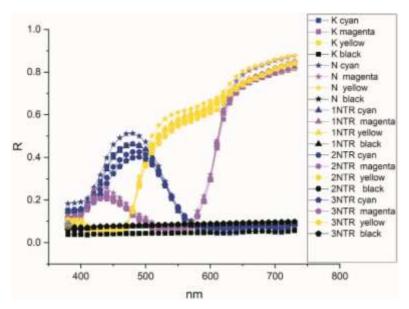


Fig. 2. Spectral reflections of unaged laboratory substrates printed with cyan, magenta, yellow, and black inks

The optical degradation of all printed substrates without (K, N) and with 10%. 20% and 30% triticale pulp (1NTR, 2NTR, 3NTR) was observed based on the difference in reflection before and after the aging process,  $\Delta R$  (natural or artificial aging of 48 hours or artificial aging of 96 hours) (Figure 3-13). All samples containing triticale pulp were compared with laboratory reference sample (N) and control sample, industrial paper of 42.5 g/m<sup>2</sup> (K) separately for cyan, magenta, yellow, and black inks.

Differences in the spectral reflections of substrates printed with cyan ink before and after the natural aging process are presented in Figure 3, while differences in the spectral reflections of the same samples before and after artificial aging processes are presented in Figure 4 for the 48-hour aging process, and in Figure 5 for the 96-hour aging process.

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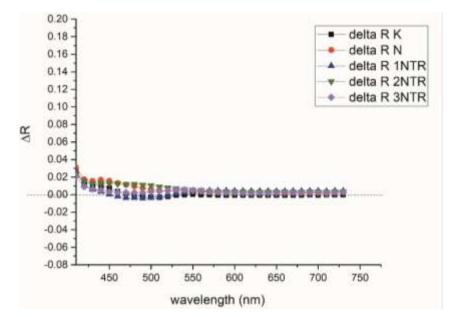


Fig. 3. Reflection difference of printed substrates with cyan ink before and after the natural aging process

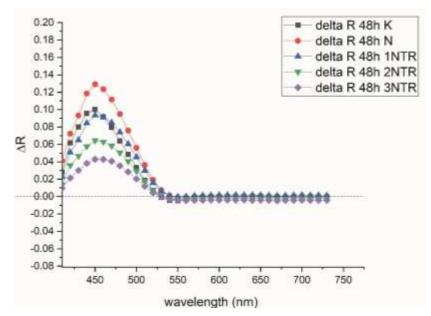


Fig. 4. Reflection difference of printed substrates with cyan ink before and after the artificial aging process of 48 hours

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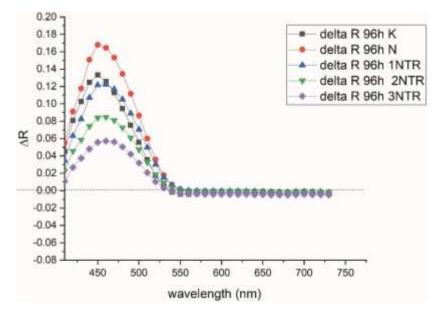


Fig. 5. Reflection difference of printed substrates with cyan ink before and after the artificial aging process of 96 hours

Differences in the spectral reflections of substrates printed with magenta ink before and after the natural aging process are presented in Figure 6, while differences in the spectral reflections of the same samples before and after artificial aging processes are presented in Figure 7 for the 48-hour aging process, and in Figure 8 for the 96-hour aging process.

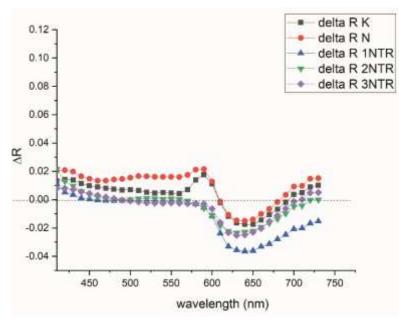


Fig. 6. Reflection difference of printed substrates with magenta ink before and after the natural aging process

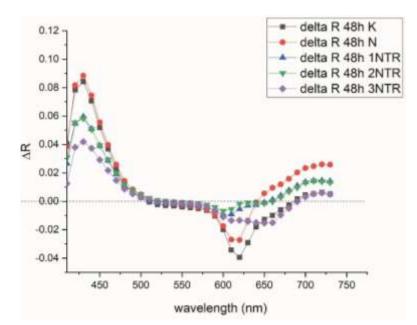


Fig. 7. Reflection difference of printed substrates with magenta ink before and after the artificial aging process of 48 hours

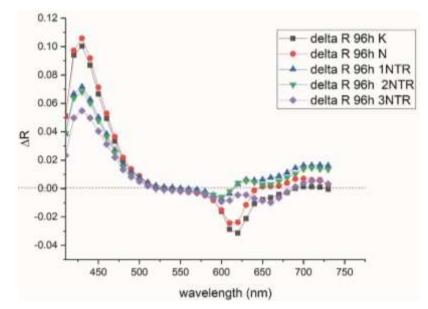


Fig. 8. Reflection difference of printed substrates with magenta ink before and after the artificial aging process of 96 hours

Differences in the spectral reflections of substrates printed with yellow ink before and after the natural aging process are presented in Figure 9, while differences in the spectral reflections of the same samples before and after artificial aging processes are presented in Figure 10 for the 48-hour aging process, and in Figure 11 for the 96-hour aging process.

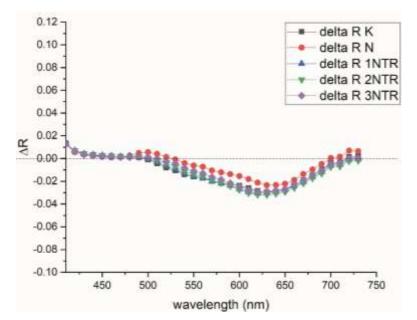


Fig. 9. Reflection difference of printed substrates with yellow ink before and after the natural aging process

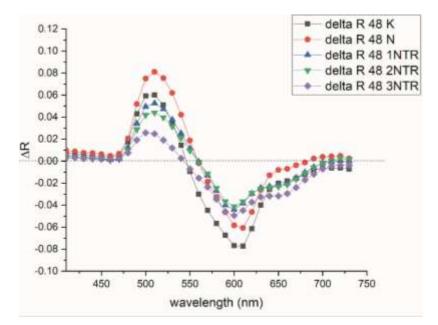


Fig. 10. Reflection difference of printed substrates with yellow ink before and after the artificial aging process of 48 hours

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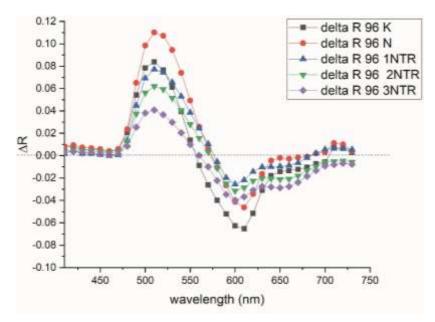


Fig. 11. Reflection difference of printed substrates with yellow ink before and after the artificial aging process of 96 hours

Differences in the spectral reflections of substrates printed with black ink before and after the natural aging process are presented in Figure 12, while differences in the spectral reflections of the same samples before and after artificial aging processes are presented in Figure 13 for the 48-hour aging process, and in Figure 14 for the 96-hour aging process.

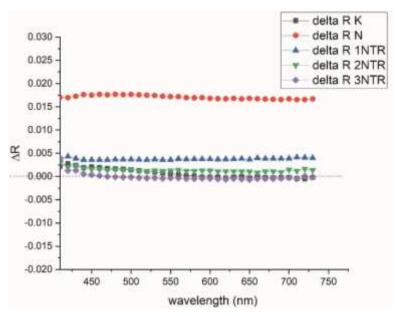


Fig. 12. Reflection difference of printed substrates with black ink before and after the natural aging process

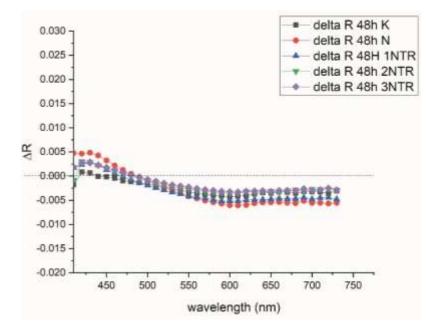


Fig. 13. Reflection difference of printed substrates with black ink before and after the artificial aging process of 48 hours

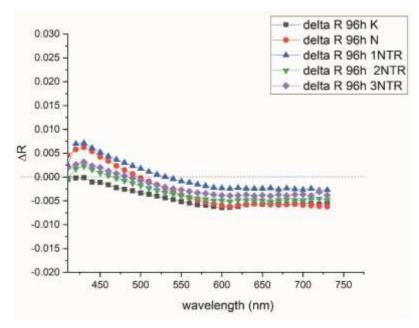


Fig. 14. Reflection difference of printed substrates with black ink before and after the artificial aging process of 96 hours

# 4. DISCUSSION

Prior to any aging treatment, each printing ink reflects visible light at specific wavelength intervals reaching a specific reflection maximum (Figure 2). The reflections measured for each of the 5 paper substrates have noticeable minimal differences only in cyan and yellow prints where the prints on the laboratory paper substrate with recycled wood fibers (N) have the greatest reflection, while other prints contain approximately equal specific reflection values, regardless of the printing substrates composition.

After natural aging for a period of two years, the cyan prints did not show significant color changes on any of the observed paper substrates (Figure 3), while the effect of artificial aging resulted in significant changes (after 48 hours  $\Delta R_{max(N)}$ = 0.129 on 450 nm; after 96 hours  $\Delta R_{max(N)}$ = 0.168 on 450 nm) what can be seen in Figures 4 and 5. After the artificial aging processes, the largest changes in the cyan prints occur on laboratory substrate N (reference sample). Approximately the same changes are observed in prints on K substrates (industrial paper) and substrates with a content of 10% triticale pulp (1NTR), while cyan samples which are printed on substrates with 20% (2NTR) and 30% (3NTR) proportion of triticale pulp show less and fewer changes in reflection after both cycles of artificial aging processes (after 48 hours  $\Delta R_{max(3NTR)}$ = 0.043 on 450 nm; after 96 hours  $\Delta R_{max(3NTR)}$ = 0.057 on 460 nm).

As a consequence of natural aging, magenta prints have slightly larger changes in reflection on substrates containing recycled wood pulp in the interval from 400 nm to 600 nm, for example,  $\Delta R_{max(N)} = 0.022$  on 590 nm and  $\Delta R_{max(K)} = 0.018$  on 590 nm, while UV inkjet prints performed on substrates with triticale pulp contain no changes in this interval (Figure 6). In the interval from 600 nm to 700 nm, little oscillations in  $\Delta R$  occur in the reflection part of the spectrum. On the other side, after both cycles of artificial aging processes, reflection differences in the blue part of the spectrum are significant on all the substrates, but the largest differences are visible on substrates containing only recycled wood fibers K and N ( $\Delta R_{96max(N)} = 0.106$  on 430 nm and  $\Delta R_{96max(K)} = 0.100$  on 430 nm) (Figures 7 and 8). In the red part of the spectrum, there are oscillations in reflection differences similar to those in naturally aged samples, but again the largest oscillations are visible on substrates K and N.

Values of reflection difference caused by natural aging are very low on yellow prints regardless of the used paper substrates (Figure 9). Larger changes are observed in prints after both cycles of artificial aging processes, where the yellow prints on substrates containing only recycled wood pulp (N and K) are the most unstable ( $\Delta R_{96max(N)}$ = 0.110 on 510 nm and  $\Delta R_{96max(K)}$ = 0.084 on 510 nm), while a yellow print obtained on a substrate with 30% triticale pulp (3NTR) has the smallest changes in reflection differences ( $\Delta R_{96max(3NTR)}$ = 0.040 on 510 nm (Figure 10-11).

Each particular ink has a specific wavelength in the dominant reflective interval, where reflectance values achieve their maximum. Black ink absorbs electromagnetic radiation throughout the measurement spectrum, so there will be color changes of almost  $\Delta R \approx 0$  for the whole measurement interval (Figure 12-14).

The highest degree of optical deterioration after artificial aging concerning the used printing inks is observed in cyan prints, while the lowest change is obtained in black prints.

Changes in reflection due to the process of two-year natural aging are negligible, while the result of electromagnetic radiation is significantly noticeable.

# 5. CONCLUSIONS

Taking into account all the results obtained after natural and artificial aging processes, the following could be concluded:

- Electromagnetic radiation applied during artificial aging has a greater effect on changes in the colour of the print than the natural aging of two years.
- Printed samples containing triticale pulp are optically more stable than the reference and control samples due to the partial use of primary fibers instead of secondary ones.
- The reference laboratory sample shows the worst optical stability, so the addition of primary fibers and fillers could improve its properties.

The obtained results of the optical properties of laboratory papers with triticale pulp printed using the UV inkjet technique indicate that addition of triticale pulp could improve stability of recycled paper as a printing substrate.

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### REFERENCES

- 1. Kirwan, MJ 2013, Handbook of Paper and Paperboard. Packaging Technology. Second Edition, Wiley-Blackwell, UK
- 2. Ince, PJ 1996, *Recycling of wood and paper products in the United States*, U.S. Department of Agriculture. Forest Products Laboratory, Madison, USA; General Technical Report FPL-GTR-98, p. 10.
- 3. Fahmy, Y; Fahmy, TYA; Mobarak, F; El-Sakhawy, M; Fadl, MH 2017, "Agricultural Residues (Wastes) for Manufacture of Paper, Board, and Miscellaneous Products: Background Overview and Future Prospects" *International Journal of ChemTech Research*, vol. 10, no. 2, pp. 425-448.
- 4. Ashori, A 2006, "Nonwood Fibers A Potential Source of Raw Material in Papermaking", *Polymer-Plastics Technology and Engineering*. vol. 45, no.10, pp.1133–1136.
- 5. Evans, P; Sherin, A; Lee, I 2013, *The Graphic Design Reference Specification Book Everything Graphic Designers Need to Know Every Day*, Rockport Publishers, p.320.
- Youngquist, JA; Krzysik, AM; English, BW; Spelter, HN; Chow, P 1996; "Agricultural fibers for use in building components. The use of recycled wood and paper in building applications," *Proceedings of a 1996 Symposium*, Madison, WI, pp. 123-134.
- 7. Spöttle, M; Alberici, S; Toop, G; Peters, D; Gamba, L; Ping, S; Van Steen, H; Bellefleur, D 2013, "Low ILUC potential of wastes and residues for biofuels," *Straw, forestry residues, UCO, corn cobs, ECOFYS Netherlands B.V.*, p. 168
- 8. Leponiemi, A 2008, "Non-wood pulping possibilities A challenge for the chemical pulping industry", *Appita J.* vol. 61, no. 3, pp. 234-243.
- Jahan, MS; Gunter, BG; Rahman, AFMA 2009, "Substituting wood with nonwood fibers in papermaking: A win-win solution for Bangladesh," BDRWPS Working Paper No. 4. DOI: 10.2139/ssrn.1322292
- 10. Sridach, W 2010, "The environmentally benign pulping process of non-wood fibers," *Suranaree J. Sci. Technol.* vol.17 no. 2, pp. 105-123.
- 11. Plazonic, I; Bates, I; Barbaric-Mikocevic, Z 2016, "The Effect of Straw Fibres in Printing Papers on Dot Reproduction Attributes, as Realized by UV Inkjet Technology" *BioResources*, vol. 11, no. 2, pp. 5033-5049.
- 12. ASTM D 6789-02, Test Method for Accelerated Light Ageing of Printing and Writing Paper by Xenon-Arc Exposure Apparatus (2002).