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Utilization of Laboratory Papers with Non-Wood Fibres as Printing Substrates Observed Through the Maximum Ink Penetration Depth

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Abstract: The use of non-wood fibres for paper production could be one of the most environmentally friendly and economical alternatives. Reducing the consumption of wood pulp in paper and cardboard production by replacing wood pulp with alternative plant biomass could be a viable solution, as the amount of non-wood fibres in biomass is far from being exhausted. In this study, straw from the most commonly grown agricultural crops in Croatia was used as a source of non-wood fibres. Agricultural residues from wheat, barley and triticale were selected as a substitute for wood fibres for the production of laboratory papers with straw fibres. Under laboratory conditions, straw pulp was mixed with recycled wood pulp in a ratio of 30:70 to produce paper sheets that can be printed with different printing techniques. Regardless of the printing technique used, it is desirable that the prints contain a high-quality reproduction of the image and text on the surface of the paper and that the ink does not penetrate completely through the substrate. In this context, this study observed the use of laboratory-made papers with non-wood fibres as the printing substrate by analysing the maximum depth of ink penetration into the printing substrate obtained with two printing techniques - a modern one (digital UV inkjet) and a very high quality conventional one (gravure). It was found that the gravure printing favoured a greater penetration of the UV ink into the substrate with the addition of straw pulp compared to the digital printing technique. However, this is a consequence of the printing technique, as similar ink penetration was also observed on the laboratory substrate made only from recycled fibres. Compared to commercial papers, the ink penetration is slightly higher into the laboratory made printing substrates. It is interesting to note that the printing substrate with the addition of 30% triticale pulp has the lowest ink penetration, especially in multicolour prints produced with the digital UV inkjet printing technique.

Keywords: digital printing; gravure printing; ink penetration; laboratory paper; non-wood fibres; straw pulp

1 INTRODUCTION

Today, most non-wood fibres in biomass remain unused. This represents a massive waste of raw materials every year, but it is possible to add value to it. In India, for example, straw paper is used every day for printing advertising leaflets. This is also the case in Mexico, where paper made from sugarcane fibres is used to print newspapers. However, these types of paper are not widely used in Europe and it is important to find a way to add value to local biomass waste, as straw waste is abundant in Croatia. Straw is in fact the fastest growing annual fibre source, with a lower lignin content than wood with approximately the same cellulose content. Due to the morphology and chemical composition of the plant, non-wood pulp can be produced at low temperatures with a lower chemical charge than wood pulp [1]. Cellulose-based papers are the most important substrate for the printing industry, where the movement of liquid ink in and between cellulose fibres is largely controlled by the hydrophilic nature of the fibres. When using printing techniques based on small drops of ink (the size of a picolitre) dripped onto the paper surface, the ink begins to penetrate the porous paper substrate. When using uncoated paper as a printing substrate, this behaviour is much more pronounced because uncoated paper is an anisotropic porous medium consisting of bundles of fibres that cross each other in a planar orientation. Since the fibres in the paper are usually impregnated with granular mineral substances as fillers, it has been observed that the liquid as ink penetrating the fiber layer first follows the direction of the fibres and moistens them and then fills the pore space between the fibres [2].

However, the addition of fillers (such as calcium carbonate) to the pulp prior to papermaking has been found to have limited effect on reducing ink penetration, although it does significantly increase the opacity of the paper. On the other hand, the addition of chemicals for internal sizing (hydrophobisation) can significantly reduce ink penetration. [3].

In this study, papers with 30% non-wood fibres (barley, triticale and wheat) and 70% recycled wood fibres were produced under laboratory conditions and then printed using a modern technique that is successfully gaining acceptance in the market - digital UV inkjet printing and a high-quality conventional technique - UV gravure printing. The aim of this research was to measure and compare the penetration of the ink into the paper substrate with the above printing methods when it comes to achieving a quality print. It is known that the absorption of ink into the printing substrate depends on the physical and chemical properties of the substrate, on the properties of the ink, but also on the interactions between the substrate and the ink [4]. Although the ink drying method is similar in both processes, the composition of the ink can vary, leading to different ink penetration results. The main advantage of inkjet printing is that it does not require printing plates or engraved cylinders as is the case with traditional printing methods. In fact, the ink droplets are printed directly onto the surface of the substrate following a digital signal, making the digital printing process less fast and mainly used for short runs [5].

Gravure printing, on the other hand, requires an engraved copper cylinder, which is very expensive, and print runs must be very large (millions of copies) to be profitable.

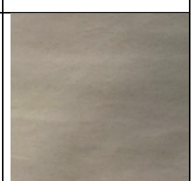
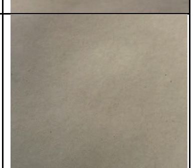
In this study, a method combining microscopy and image analysis was used to study the penetration of ink into paper substrates. Digital and gravure prints on paper substrates of different compositions (made from recycled pulp or with the addition of straw pulp) were microtomed, scanned with an optical microscope and analysed by image processing. The depth of penetration and the distribution of the ink in the paper were then determined by statistical analysis to evaluate the possibility of using papers with non-wood fibres as a printing substrate.

2 EXPERIMENTAL PART

2.1 Laboratory Papers

Laboratory papers weighing approximately 42.5 g/m^2 were prepared using a Rapid-Köthen sheet former (FRANK-PTI) according to the standard EN ISO 5269-2:2004 [6]. For this purpose, crop residues left in Croatian fields after the harvest of wheat, barley and triticale were collected and cut by hand into pieces up to 3 cm long. The straw, cleaned of grain and impurities, was converted into a semi-chemical pulp using the soda pulping method [7]. The resulting unbleached pulp was blended with a 70% recycled wood fibre pulp, and four types of laboratories produced papers were produced as shown in Tab. 1. In addition to the laboratory produced papers where N was produced from the recycled pulp only (as a reference sample) and 3NW, 3NB and 3NTR were produced with 30% straw pulp, the commercial paper labelled K served as a control sample.

Table 1 Mark and composition of papers used as printing substrates.

Printing substrate				
Mark	Composition: Straw pulp	Composition: Recycled pulp	Production	Photography
K	0%	100%	commercial	
N	0%	100%	laboratory	
3NW	30% wheat	70%		
3NB	30% barley	70%		
3NTR	30% triticale	70%		

As the commercial paper K is made only from recycled wood fibres, a specific method of ash determination was carried out to estimate the content of mineral salts and other inorganic substances contained in the paper. It is applied to all types of pulp. Whilst most inorganic fillers used in pulp and paper industry do not decompose below 900 °C, calcium carbonate ($CaCO_3$) is an exception, losing carbon dioxide at temperatures above 525 °C. Therefore, if the ash is determined at two different temperatures (900 °C and 525 °C according to TAPPI standards T 413 om-11 and T 211 om-2 respectively) [8,9] it is possible to calculate the percentage of calcium carbonate and clay in paper according to equations (1-2) [10].

$$\mathrm{CaCO_3},\,\% = \left(Ash_{525} - Ash_{900}\right)\times\frac{100}{44} \quad (1)$$

$$\mathrm{Clay},\,\% = \left(Ash_{525} - \mathrm{CaCO_3}\right)\times 1.13 \quad (2)$$

The percentages of calcium carbonate and clay content in commercial paper K were calculated on oven dry-basis, so before sample ignition the moisture content in accordance with the TAPPI standard T 550 om-08 was determined [11].

2.2 Gravure Prints

Gravure printing enables high-quality and consistent reproduction of the finest multicolour details, even in the smallest images, on thin and flexible printing materials in large print runs. This printing process achieves the highest, most consistent print quality with continuous reproduction compared to other printing techniques [12].

In this study, all prepared substrates (Tab. 1) were printed in full tone by KPP Gravure system with 65 Shore impression roller and a 100 lines/inch engraving plate (RK Rint Coat Instrument Ltd) with Solarflex UV-curable inks from Sun Chemicals at a temperature of 23 °C and a relative humidity of 50 %. After printing, the gravure prints were dried using a Technigraf Aktiprint L 10-1 continuous dryer with a light source output of 120 W/cm and an intensity of 60%.

2.3 Digital UV Inkjet Prints

All print substrates were printed using an EFI Rastek H652 digital UV-curable inkjet printer with a resolution of 600 × 600 dots per inch (dpi) (with high quality mode 8 pass) and a print speed of 12.10 m^2/h. This printer uses two double-intensity ultraviolet (UV) lamps with a power of 700 W, which cure UV ink through the shortest curing times and exceptional polymerisation [13].

Inkjet printing is a non-contact technology in which the printing process is based on the ink droplets sprayed from the printhead nozzles. The data of the digital print job is transmitted directly to the inkjet system, which transfers the ink via nozzles to the substrate [14].

In both printing techniques, each printing substrate was printed to achieve a full tone with either: a) one layer of ink (cyan (C), magenta (M), yellow (Y)) or b) two layers of ink, where one ink is printed over the other, resulting in three different colours: Magenta + Yellow = Red (R), Cyan + Yellow = Green (G), Cyan + Magenta = Blue (B) or c) three layers of ink (Cyan + Magenta + Yellow) (marked as S).

The method of drying UV-curable inks is similar for both processes, with the drying process involving UV radiation to cross-link the organic molecules (monomers) or curing by radiation. The ink used and its interaction with the print substrate determine the thickness of the ink layer on the substrate and thus the overall print quality.

2.4 Preparation of Samples for Microscopic Analysis

After all substrates were printed using both printing techniques, they were cut into 10 × 30 mm strips and placed in an epoxy resin that is a mixture of 88% Epofix resin (containing bisphenol-a-diglycidyl ether) and 12% Epofix hardener (containing triethylenetetramine). The resins were then dried with the test strips for 24 hours at room temperature before being ground and polished with a Buehler grinder and a Struers DAP -V polisher.

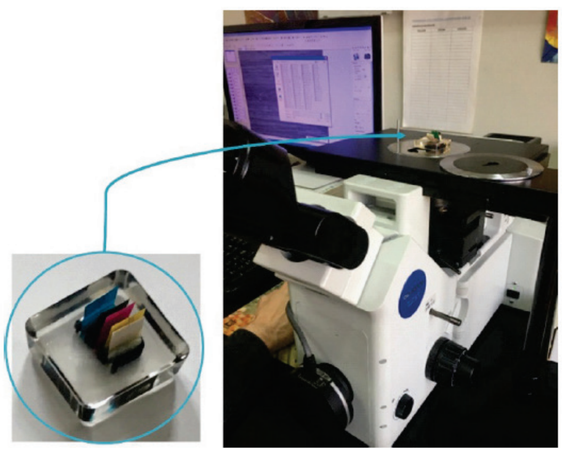


Figure 1 Samples placed in an epoxy resin and examined through the Olympus microscope GX 51 for analysis of the cross section [*15*].

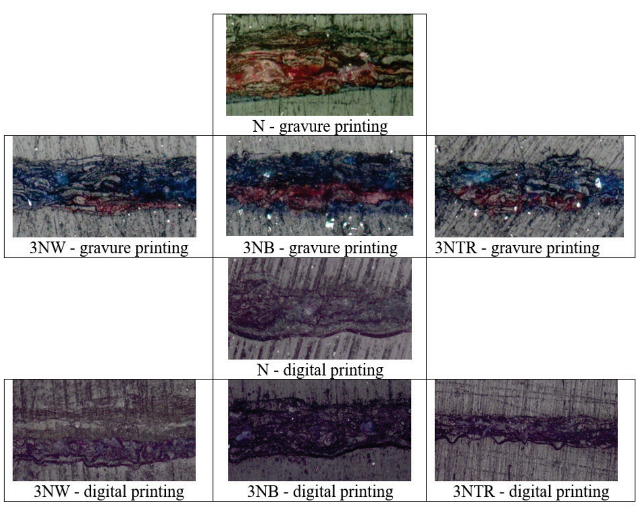


Figure 2 Microscopic images of the penetration of cyan+magenta (B) inks into laboratory papers (N, 3NW, 3NB, 3NTR) printed by gravure and digital printing processes.

The cross-section of the samples was viewed at 200× magnification using an Olympus GX 51 light microscope and analysis software (Fig. 1). The images taken under the microscope (Fig. 2) were further analysed using ImageJ software to measure the maximum penetration depth of inks into laboratory papers with non-wood fibres. The maximum ink penetration depth, referred to as *Hp* in this paper, was calculated according to equation 3 from 20 measured cross-sections of the maximum ink penetration value obtained from each microscopic image. The thickness of the print substrates was also determined from the images taken.

$$Hp = \frac{l}{d} \times 100 \quad (3)$$

Where: l represents the maximum ink penetration value and d is the local thickness of the paper in the measuring section.

3 RESULTS AND DISCUSSION

From a macroscopic point of view, there are differences between printing on commercial and laboratory papers and between printing on papers made from recycled fibres and papers with added non-wood fibres. In fact, wheat, barley and triticale fibres are wider than the recycled fibres, resulting in high local roughness and making it difficult for inks to cover these areas. In recent research [7], it was found that the addition of straw fibres to recycled wood fibre pulp results in papers with a rougher surface by up to 30% (the arithmetic means surface roughness, *Ra*, is 10% higher than papers made from recycled fibres alone).

As the paper used as a control in this research was commercial paper (K) made only from recycled wood fibres, ash i.e. filler content was determined since their presence in printing substrate have significant influence on ink penetration. Results of chemical composition analysis of control paper K is presented in Tab. 2.

Table 2 Chemical composition of control paper substrate K.

w, %	Substrate K
Moisture	3.12 ± 0.38
Ash_{525}	14.12 ± 0.21
Ash_{900}	9.68 ± 0.17
$CaCO_3$	10.10 ± 0.18
Clay	4.54 ± 0.20

First, a comparison was made between commercial paper (K) and laboratory papers with non-wood fibres, considering only prints in one ink layer. Then the same comparison was made, but this time with prints on a reference sample, i.e., laboratory paper made from recycled fibres (N).

In order to achieve a high-quality multicolour reproduction, the prints must be printed in several ink layers. Therefore, an additional comparison was made with multiple ink layers printed according to the recommendations of ISO 12647: Cyan+Magenta (B), Cyan+Yellow (G), Magenta+Yellow (R) and three of these inks (S).

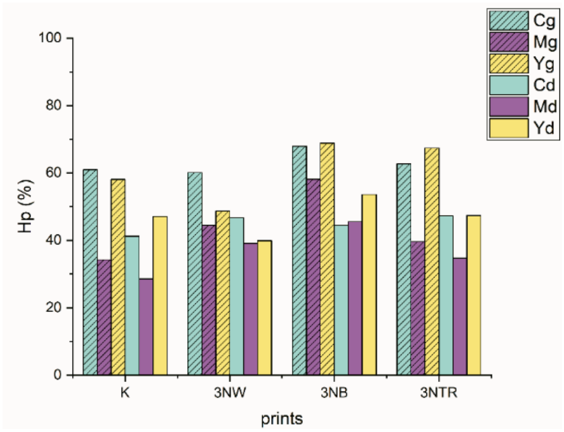


Figure 3 Comparison of the maximum penetration depth of process inks on the control sample (K) and the laboratory papers with non-wood fibres (3NW, 3NB, 3NTR) in gravure and digital inkjet printing processes.

Fig. 3 shows that substrate 3NW (with 30% wheat pulp) has the lowest ink penetration, especially with digital inkjet printing technology. Furthermore, the 3NTR substrate shows similar results to the control paper (K) for both printing techniques, but especially for the digital inkjet technique. The 3NB printing substrate has the highest ink penetration rate of up to 70% achieved in gravure printing, and this high value results in the ink being seen on the other side of the paper. This phenomenon could lead to a reduction in the number of applications for this substrate.

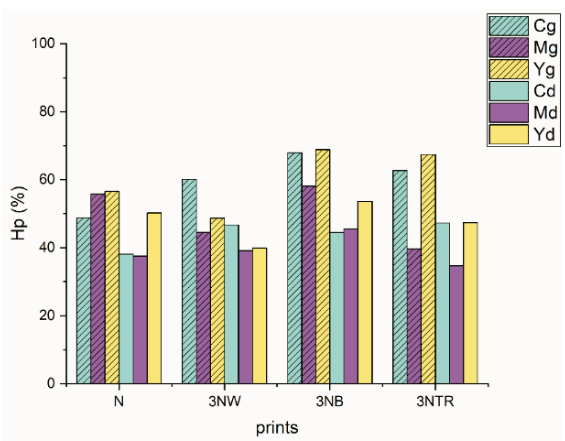


Figure 4 Comparison of the maximum penetration depth of process inks on reference samples (N) and laboratory papers with non-wood fibres (3NW, 3NB, 3NTR) obtained with gravure and digital inkjet printing processes.

The reference paper (N) showed similar Hp values to the laboratory paper with wheat pulp (3NW) for both printing techniques (Fig. 4). In terms of printed ink, the magenta ink penetrates the least through the observed papers (regardless of the manufacturing method). The range of penetration of magenta ink is from 27% to a maximum of 60%, while the range for other inks such as yellow ink is from 39% to a maximum of 76%. The graph also shows that the 3NW paper substrate is better than the 3NB and 3NTR substrates as there is a lower average penetration depth. In the packaging industry, most multicolour prints consist of four primary inks, in some cases even up to 8 inks (CMYK+Pantone). Therefore, it is very important to analyse the ink receptivity on the ink in layers.

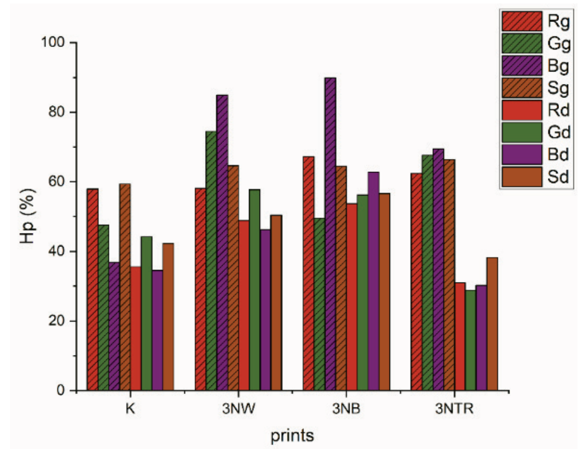


Figure 5 Comparison of the maximum ink penetration depth in two/three layers on the control sample (K) and the laboratory papers with non-wood fibres (3NW, 3NB, 3NTR) obtained with gravure and digital inkjet printing processes.

Fig. 5 shows that the 3NTR print substrate again has an Hp value closest to that calculated for the commercial paper (K). Furthermore, the ink penetration depth achieved on a 3NTR print substrate appears to be constant for all inks applied with the same printing technique. In addition, the Hp values obtained from inkjet ink penetration measurements in the 3NTR printing substrate are lower than those obtained on the commercial paper (K).

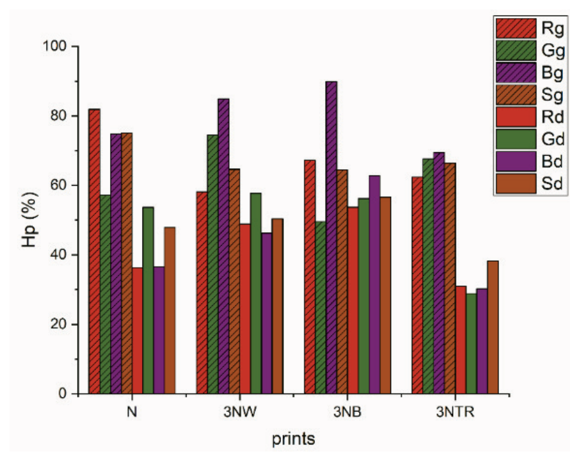


Figure 6 Comparison of the maximum ink penetration depth in two/three layers on the reference sample (N) and the laboratory papers with non-wood fibres (3NW, 3NB, 3NTR) obtained with gravure and digital inkjet printing processes.

The same tendency of the results in relation to the comparison of commercial (K) and laboratory-produced paper (N) can be seen in Fig. 6. In contrast to the gravure prints, laboratory substrates printed with digital inkjet technology show a lower penetration depth of the ink into the substrate, which is most evident in the prints on substrates with barley and wheat pulp. When looking at the inks, it can be seen that the deepest penetration of the ink is achieved by a combination of magenta and cyan ink (marked B), which is up to 90% for prints on substrate 3NB, i.e., in these prints the ink can be seen on the back of the paper.

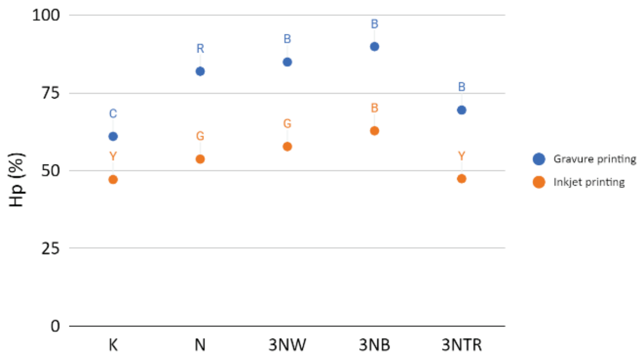


Figure 7 Maximum ink penetration depth for each substrate achieved with a given ink in both printing techniques.

Regardless of the composition and production method of the paper as substrate, the highest value for the penetration depth of the ink is achieved in gravure printing (Fig. 7). From the aspect of ink, in general, the highest penetration, regardless of the type of printing technique and printing substrate, was shown by inks applied in two layers. However, the highest penetration of all inks is observed for cyan+magenta (B) ink into laboratory papers with straw pulp (3NW, 3NB, 3NTR) printed by gravure technique. This can be explained as a consequence of the viscosity of the ink, since inkjet ink is more fluid (10-20 mPa·s) than gravure ink (0.05-0.2 Pa·s) [12]. Furthermore, these results can be explained by the fact that gravure requires a significant amount of pressure (from 0.345 MPa to 0.690 MPa) to be applied to the substrate during printing, in contrast to inkjet printing where the ink penetrates more deeply into the substrate [16].

Furthermore, compared to commercial paper (K), the 3NTR laboratory paper produced with an addition of 30% triticale pulp achieves similar or even better results in multicolour printing in terms of ink penetration for both printing processes. The average ink penetration for the 3NTR paper printed with the digital inkjet printing technique is almost the same as that of the control paper (K), while the lowest average penetration of all the papers examined was achieved with gravure printing on the commercial paper.

These results can be explained by the fact that wheat and barley straw have similar fibre lengths and their distribution is almost equal, whereas this is not the case with triticale straw. The triticale fibres have a much wider length range (from 0.27 mm to 2.63 mm) than the other two straw fibres [17]. Precisely because of the wide distribution of fibre length from short to very long in this type of raw material, there is a reduction in ink penetration through the substrate containing triticale fibres. In commercial paper, many additives are added to the pulp and to the paper surface after sheet formation that reduce ink penetration. For example, fillers such as calcium carbonate, barium sulphate or titanium dioxide are used to make the paper whiter and brighter, and these particle forms can reduce ink penetration. Barium sulphate, for example, has a large and flat shape (similar to a flake) that makes it harder for ink to penetrate the paper.

4 CONCLUSION

Based on the research conducted, we can draw several conclusions:

- The differences between 100% recycled papers and papers made with addition of 30% virgin straw fibres were extremely small from the aspect of ink penetration during printing.
- A modern method of printing, digital UV inkjet, achieves a lower penetration of ink into the printing substrate due to the elimination of high pressure that unavoidable in the conventional printing process.
- Prints on laboratory paper containing 30% triticale pulp, achieve the lowest maximum ink penetration regardless of the printing technique used, proving that this substrate can be used for both monocolour and multicolour printing.
- Although prints on laboratory paper with 30% barley pulp have the highest ink penetration depth, this substrate can be used for single-sided printing without affecting the visibility of the ink on the other side of the paper, e.g., for labels or flyers.

From a development point of view, it would be interesting to carry out analyses on papers made from other biomass-derived fibre materials, such as sugar cane or bamboo fibres, to investigate whether the maximum ink penetration depth is even closer to papers produced in paper mills.

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