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Print Durability and Recyclability of Label Paper Equipped with Printed RFID Antenna

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Abstract: Labels are a crucial component of products, offering informational content and attractive visuals; therefore, the durability of the print is an important quality requirement. On the other hand, in accordance with eco-design, the recyclability of printed labels is vital. In our research, the focus was on the assessment of the durability, recyclability, and deinkability of printed label paper equipped with printed RFID tags. The determined color fastness of electrophotographic prints affected by various environmental factors showed good resistance to dry rubbing and, in most cases, light and moist–heat treatment, confirming the applicability of digital printing on self-adhesive biodegradable paper labels. In the second part of this study, recyclability was assessed, and a comparison between the deinkability of the offset and digital prints and two conductive functional inks was conducted. Good deinkability was observed for the printed RFID antennas on both the offset and electrographic prints, with only a small deterioration in optical properties, especially when nano-silver conductive ink was used. The study highlights the importance of the selection of materials and printing techniques when considering the environmental impact of printed electronics. The results showed that INGEDE 11 is a suitable deinking method for printed RFID antennas on offset and electrographic prints.

Keywords: durability; recyclability; deinkability; RFID; CMYK print

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1. Introduction

Labels play a key role in product identification and provide important information about the composition, characteristics, ownership, and purpose of a product. Today, labeling has surpassed its basic function of describing a product and is now a vital sales and marketing tool that contributes significantly to the design of a product or its packaging [1–3]. Among the different types of labels, self-adhesive labels, composed of a liner, adhesive, and face stock, i.e., label paper, are gaining popularity due to their simplicity, user-friendliness, and high-print quality [4]. A label needs to be resistant to various environmental conditions. The permanence and durability of a print constitute an important issue for print quality and the legibility of printed information [5].

Image durability can be defined as the change in or degradation of initial image quality over time and/or in response to a specific factor (light, heat, humidity, and/or air quality). Bugner and Gordon [6] primarily focused on the factors influencing the permanence and durability of commercial prints. Their research covered different printing technologies, emphasizing the role of inks, substrates, and printing systems and highlighting the importance of choosing the right materials based on the intended application [6]. Malenica et al. [7] studied the stability of offset-printed labels created using thermochromic and conventional inks with respect to light, chemicals, and rubbing. Their key findings include the following: substrate type significantly influences print stability and mixing thermochromic ink with conventional ink affects overall stability. Their study also demonstrated good rub resistance but reduced stability against ethanol, water, and UV radiation [7]. A comparison of the image permanence and print durability of digital and offset prints was made by Lindstrom and Bugner [8], assessing the resilience of inkjet, electrophotographic, and traditional offset printing against environmental and physical stress factors. The findings indicate that both digital printing methods approach or even exceed the performance of traditional offset methods used in many commercial printing applications [8]. Enninful et al. [9] focused on identifying factors contributing to image fading in digital prints. Their study showed that paper and ink quality significantly affected print durability [9]. The stability of inkjet-printed labels immersed in fluids was reported by Beiner [10], who focused on the durability of printed labels in fluid-preserved collections. Their study compared the performance of inkjet-, laser-, and thermal-transfer-printed labels under conditions like exposure to ethanol and formaldehyde and reported that inkjet labels performed best [10]. Blaznik et al. [11] investigated the stability of inkjet printing inks under UVC radiation. Their study showed that the presence of oxygen negatively affects ink stability in water solutions and that an inert atmosphere can prolong ink durability [11].

Recently, the environmental aspect has become an important part of every product and manufacturing process. This aspect is even more important for products that have a short lifespan, such as packaging and labels [12]. The design of sustainable products also entails the use of recycled materials and ensuring good recyclability of materials [13,14]. Recycling paper, i.e., all collected paper and cardboard intended for recycling, is the most important raw material (quantitatively) for the European paper industry [15]. It is the most recycled material in Europe and represents a good example of a circular economy. In CEPI countries, that is, members of the Confederation of European Paper Industries representing European nations in the paper industry, the recycling rate has shown a general upward trend over the years, increasing from 51.8% in 2000 to a peak of 73.3% in 2020. The specific rates were 71.9% in 2015, 72% in 2016, 72.4% in 2017, 71.6% in 2018, 72% in 2019, 73.3% in 2020, 71.4% in 2021, and 70.5% in 2022. These data indicate consistent growth in recycling efforts, although there was a slight decline after 2020, with the rate dropping to 70.5% in 2022 [16]. Despite the high rate of recycling, some megatrends have been observed in the last decade that indicate a slight decline in the rate of recycling and the quality of paper for recycling [17–19]. The quality of recycled fibers depends on the type of paper for recycling, the length of the fibers and the amount of non-paper components in the paper for recycling, the ash content, and the process of recovered paper treatment [20]. The constant decline in the use of graphic paper; the greater amount of post-consumer paper collected, which is often mixed with other waste; and the greater presence of packaging made from recycled fibers have a negative effect on quality [21]. One of these trends is the increasing use of digital printing, which is more sustainable compared to offset printing, but the prints are less deinkable [22]. When recycling paper, the ability to remove printing ink is also important for the quality of the recycled fibers. The deinking process is a chemical–mechanical process consisting of removing printing inks from the printed surface of paper [23,24]. In Europe, deinking, which is carried out via the flotation process, is the most important technological process [25,26].

Another trend that could influence the recyclability of paper for recycling in the future is the increasing presence of printed electronics in paper and cardboard products [27,28]. Printed electronics refers to printing passive and active electronic structures on rigid and flexible printing materials such as paper, cardboard, plastic, textiles, etc., using digital and classic printing technologies [29,30]. Adding electronic functions directly onto the packaging material with printing is a very cost-effective production method since printing techniques in the packaging industry have been developed to be cost-efficient at large- and small-product scales [31].

Among the different printed electronic components, RFID (radio-frequency identification) tags have witnessed widespread commercial use [32–36]. They are mostly used in packaging logistics, primarily at the pallet level in retail applications, though item-level tagging is becoming more popular, too. Besides their use in transportation and logistics, they are also used to track manufacturing processes from start to finish and in the service industry, inventory control, authentication, and security [18]. With the ability to easily interface with additional devices and integrate with different electronic components, such as sensors for strain or crack detection, material corrosion analysis, and food quality and healthcare evaluation, an expansion in the applications of RFID will likely be seen in the future [37,38].

RFID printing technology has evolved remarkably over the years, as evidenced by various research findings from multiple research studies. This advancement is steering towards more environmentally friendly, cost-effective, and versatile applications, from food safety and library management to high-security authentication and IoT implementations. El-Sawy et al. [39] discussed chip-less RFID tags made from copper metal traces on polyamide substrates. Tags, designed for environmental sustainability, leverage a low-cost Bluetooth detector circuit with two detection methods to create unique IDs. Orecchini et al. [40] explored RFID's integration with green technologies. They highlighted the development of inkjet-printed RFID platforms using optimized metallization and diode integration. The study traced RFID's evolution from defense to commercial uses like logistics, pointing towards a future of flexible, eco-friendly RFID devices [40]. Virtanen et al. [41] discussed the development of passive UHF RFID tags using inkjet printing on renewable substrates like wood, paper, and cardboard. These tags, significant for IoT applications, showed varying read ranges based on the substrate, demonstrating the potential of sustainable materials in RFID technology [41]. Ali et al. [42] focused on high-security authentication using chipless RFID technology. They introduced a novel method using the randomness in inkjet printing to create unique EM signatures. These tags showed potential for secure, cost-effective authentication solutions [42]. Kisic et al. [43] presented the design and simulation of cost-effective RFID tags for identification cards. Using ink-jet printing technology and specialized simulation tools like CST Microwave Studio, the study aimed at efficient RFID tag production suitable for various applications. Lee et al. [44] discussed using nano-particle conductive silver ink for printing UHF RFID antennas. The study showed that antennas with this ink have superior radiation efficiency compared to micro-particle inks and are comparable to conventional copper antennas. Zhang and Cui [45] delved into nanometer-scale printing technology for UHF RFID tags. They emphasized the benefits of nano-conductive ink, such as improved production efficiency and cost savings, and explored the application in smart libraries [45]. Machiels et al. [46] focused on the development of high-frequency (HF) RFID antennas using screen printing on fiber-based substrates for creating sustainable and cost-efficient smart packaging solutions. The study involved selecting the best paper substrates based on printability and ink compatibility, designing, and testing RFID antennas, and integrating these antennas into cardboard packaging [46].

Active RFID tags are categorized as electronic devices and therefore fall under the scope of the WEEE Directive (Directive 2012/19/EU) [47], whereas passive RFID tags are not categorized as electronic devices [48,49]. Most of them come in the form of self-adhesive labels attached to consumer-packaged goods. Because they are attached to different types of packaging, after such packaging is disposed of, they are found in various waste treatment streams [50]. Until now, the selective separation of RFID tags from packaging and a special separate printed metallic RFID antenna recycling path has not yet been effectuated. Most RFID tags thus enter the recycling process of the packaging to which they are attached [51–53]. Even if RFID tags do not affect the recycling process, they affect processing costs, material loss, and/or the quality of recycled material [54].

In the process of paper and cardboard recycling, RFID tags attached to packaging enter the recycling process without having been separated in advance. The components of the RFID tag are separated during recycling in the disintegration and mechanical separation processes. Both the chips and the metal parts in the stamped or etched label with a solid-metal antenna, mostly separate from the fibers, remain solid objects in waste. However, some parts can also degrade into smaller pieces that are too small to be screened out. During the deinking process, the components from the RFID tags are then further removed. Similarly, for RFID tags with printed antennas, not all materials will likely be fully separated and removed [18]. Some studies show that the paper recycling process is not greatly disrupted by the presence of printed electronics [54,55]. However, RFID tags can affect the composition and amount of solid and liquid residues present after recycling. Adhesives, together with small plastic and metal parts, can pass through the filter system, increasing the quantity of sticky substances in the recycled fibers and thus deteriorating their quality [19,54].

In a study by Aliaga et al., a pilot-scale recycling test on smart-printed envelopes composed of printed antennae based on nano-silver ink, a silicon-based resistor, and a battery was carried out [28]. The results showed that resistors were retained in the sieves during the screening process, while the battery and the nano-silver functional ink from the antennae were partially dissolved in the process water and consequently had a small effect on the optical and mechanical properties of the recycled paper [28].

A study by the Fiber Box Association assessed the recycling of RFID-tagged corrugated packaging [56]. Two types of tags were tested: one containing a solid metal (copper foil) and the other incorporating an antenna printed with a silver functional ink. The results showed that the laminated solid metal tag was removed intact during screening, while the unlaminated tag broke down into smaller particles, some of which were too small to be screened out readily [56]. For RFID tags with printed antennas, most of the silver from the functional ink ended up in the recycling process [56].

Similarly, research by Huttle showed that silver functional ink disintegrated during the recycling process; most of the silver ended up in the waste sludge, though some particles ended up in the recycled paper [57]. Atkinson investigated the behavior of different metallic conductive inks during the recycling process and found that nickel and silver particles remain in suspension along with the fibers, while nano-silver particles end up in the wastewater [58]. When paper is recycled, the silver from printed antennas ends up to a greater extent in solid or liquid waste and is currently not recovered [27]. In the research by Déprès et al. (2023), it was shown that 63% of the printed material from the waste stream in the recycling of printed electronics on paper was recovered [59]. This indicates the possibility of the effective separation of functional ink from paper and recovering the components of the printed electronics after recycling [59].

In our research, the focus was on the assessment of the durability, recyclability, and deinkability of printed label paper equipped with printed RFID tags. Color printing typically uses ink of four colors: cyan, magenta, yellow, and key (black) (CMYK). The color fastness of prints using the CMYK color model, which is commonly employed in color printing, was determined after exposure to different environmental factors. A comparison of the deinkability of the offset and digital prints equipped with printed RFID tags was made. The recyclability of the printed electronics was assessed. Our main interest lies in determining the deinkability of printed RFID antennas and, secondly, comparing the deinkability of functional inks with micro- and nano-silver particles. To compare the deinkability of functional inks with micro- and nano-silver particles, only offset printed label paper was used to exclude the influence of printing ink as much as possible.

2. Materials and Methods

2.1. Materials

The labels tested in our research are commercial self-adhesive labels made from FSC-certified paper with a basis weight of 80 g/m^2, an acrylic-based adhesive, and paper liner. The face stock of Sample 1 is one-side-coated, woodfree paper, and that of Sample 2 is one-side-coated art paper. Sample 2 is one of the most used self-adhesive labels for printing in Slovenia. It is recyclable but not fully biodegradable, whereas Sample 1 has been declared a biodegradable, compostable commercial self-adhesive label.

CMYK (cyan, magenta, yellow, black) color patches with 100% coverage were printed on both samples using electrophotography. This printing technique was chosen because of its popularity in the industry. Since electrophotographic printing is cost-effective and more sustainable than offset printing and can be utilized on a large scale, it is increasingly being used in the packaging industry to print labels on various products.

To evaluate recyclability, coated label paper with a basis weight of 80 g/m^2 was used as a printing substrate. The paper was printed with magenta (M) ink using two printing techniques, electrophotography and offset printing. The coverage with the ink was 70%. In the next step, a commercial UHF RFID antenna was printed on the back, unprinted side of the paper. For printing, a RokuPrint SD 05 semi-automatic screen-printing machine (RokuPrint GmbH, Dornstadt, Germany) was used. The printed antennas were dried at 100 °C in 5 passes (one pass = 45 s) in a drying tunnel to achieve the appropriate conductive properties. To obtain a working RFID tag, chips were glued on the printed antennas with Epo-tek e2101 glue (Epoxy Technology, Inc., Billerica, MA, USA) and dried for 10 min at 150 °C between two cardboard sheets. For the production of printed electronics, commercially available functional inks from well-known producers of inks (Sun Chemical and Agfa) were chosen. Two conductive functional inks were used to print the commercial UHF antenna: Conductive Ag ink SunChemical CRSN2442 (Sun Chemical Corporation, Parsippany, NJ, USA) and Agfa Nanosilver ink ORGACON SI-P2000 (Agfa-Gevaert. N.V., Mortsel, Belgium). Conductive Ag ink SunChemical CRSN2442 was used to print antennas on both the electrographically and offset-printed samples, whereas Agfa Nanosilver ink ORGACON SI-P2000 was only printed on the offset-printed sample.

The flowchart of sample preparation is given in Figure 1.

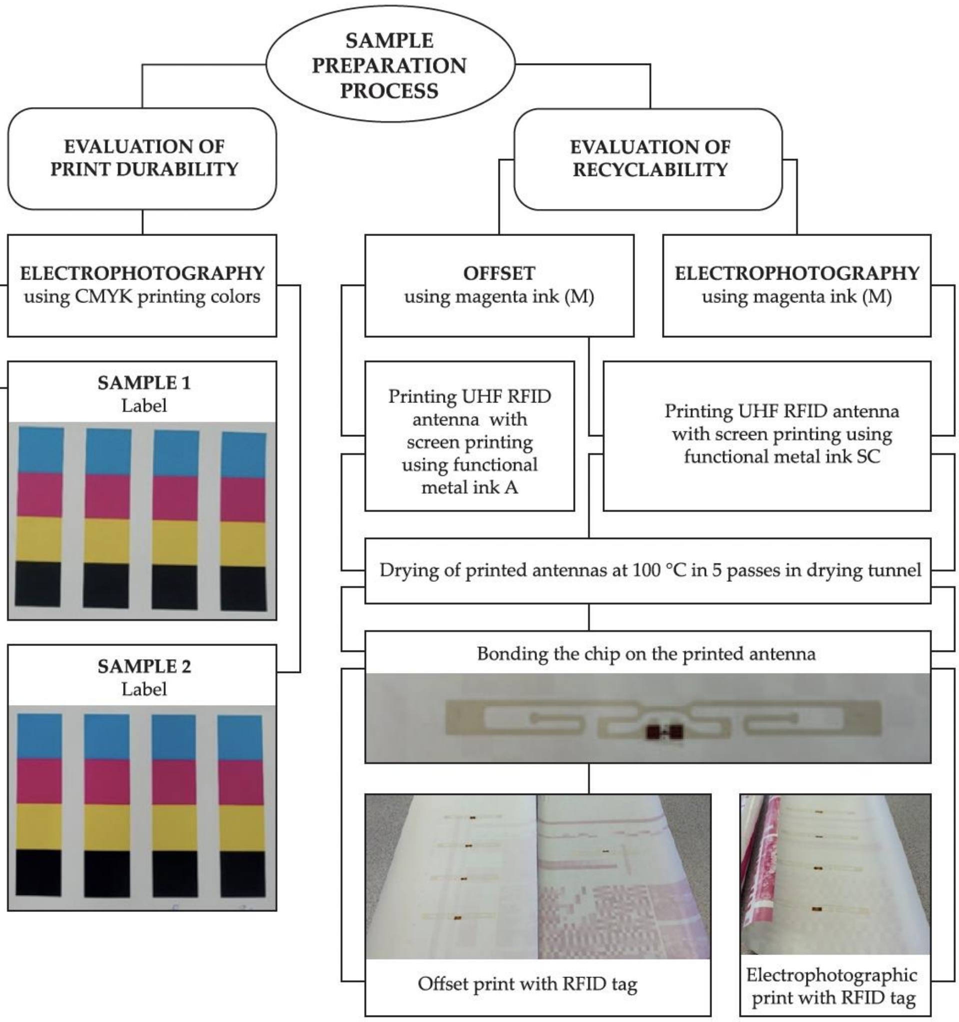


Figure 1. Flowchart of sample preparation for evaluation of durability of prints and recyclability of prints.

2.2. Evaluation of the Durability of Prints

The durability of the prints was determined via color fastness testing. This test was employed as an accelerated aging method for assessing identical areas of the individual CMYK prints following exposure to different factors using standard methods. The color fastness of the prints was determined after moist–heat treatment in climate chamber Binder KMF (Binder) based on the SIST ISO 5630-3 standard (80 °C and 65% relative humidity) [60]. The light fastness of the prints was determined after aging using a xenon lamp in a Xenotest (Alpha) apparatus based on the ISO 12040 standard (a chamber temperature of 35 °C, a black standard temperature of 50 °C, 35% relative humidity, and 42 W/m^2 irradiation intensity) [61]. In both cases, the exposure times were 72 and 144 h.

The dry rub fastness test was performed using a rubbing device, namely, Param RT-01 (Labthink Instruments Co., Medford, MA, USA), according to the ASTM D5264-98 standard (mass of weights: 2 kg; speed: 106 cpm). The CMYK prints were rubbed against the unprinted paper in 500 and 1000 strokes [62]. The color change was determined using spectrophotometric measurements. Ink transfer or bleeding from the printed test piece to the receptor surface (unprinted paper) was also evaluated visually and with densitometry.

Optical density (D) was measured on the CMYK prints using a reflection densitometer, eXact Standard (X-Rite Inc., Grand Rapits, MI, USA) (measurement geometry: 0/45; light source: D50; calibration conducted on unprinted paper). The difference in optical density ΔD was determined by subtracting the measured values of the CMYK prints for each CMYK color, before and after exposure to different factors. The reference in this case consisted of the CMYK prints before treatment.

Spectrophotometric measurements were conducted on the CMYK prints before and after exposure to the previously mentioned treatments. Each CMYK print was measured three times at three different positions on the printed area. Altogether, nine measurements were made on each tested area for each CMYK color. CIELAB is the most widely used color evaluation system, wherein color is defined by color values: here, L* denotes lightness; a completely black color corresponds to L* = 0, while a completely white color corresponds to L* = 100. Additionally, there are coordinates a* and b*, where a* indicates green/red, and b* indicates the blue/yellow coordinate axis. In the L*C*h color space, C* represents chroma, and h is the hue angle. Chroma and hue are calculated from the a* and b* coordinates. Measurements were performed based on the standard using an X-Rite Eye-One i1Pro (X-Rite, Grand Rapits, MI, USA) spectrophotometer (measurement geometry: 45/0, observation angle: 10°, and light source: D65). The color differences, ΔE*ab between the prints before and after the conducted treatments, were calculated using the equation $\Delta E^{*}ab = \sqrt{(\Delta L^{*})^{2} + (\Delta a^{*})^{2} + (\Delta b^{*})^{2}}$. The colorimetric values of the CMYK prints before treatment were taken as a reference.

2.3. Evaluation of Recyclability

The recycling of prints was conducted according to the INGEDE method 11 [63]. This method, on a laboratory scale, defines the essential steps of the deinking process: pulping and flotation. The individual stages of the process are shown in Figure 2. After the accelerated aging of the prints, their dissolution in an alkaline medium, storage in a water bath, and flotation followed. Flotation was carried out in a laboratory flotation cell. Laboratory paper handsheets were prepared from the suspension of recycled fibers after flotation using the Rapid-Köthen sheet former (Frank-PTI GmbH, Birkenau, Germany).

On the handsheets, their optical properties were determined using a Technidyne Color Touch 2 Spectrophotometer. Standardized methods were used to determine ISO Brightness (ISO 2470) [64], Luminosity, and L*, a*, b* color values (ISO 5631-2) [65]. L* represents lightness from black to white on a scale of zero to 100, while a* (the red/green coordinate) and b* (the yellow/blue coordinate) represent chromaticity with no specific numeric limits. Effective residual ink concentration (ERIC) and ink elimination (IE) values were determined according to INGEDE Method 2. ERIC is a dimensionless ratio of the light absorption coefficient of pulp-containing ink to the light absorption coefficient of the ink itself, both being determined at a wavelength of 950 nm. Ink elimination (IE) is obtained from the light absorption coefficient of the undeinked, deinked, and unprinted samples.

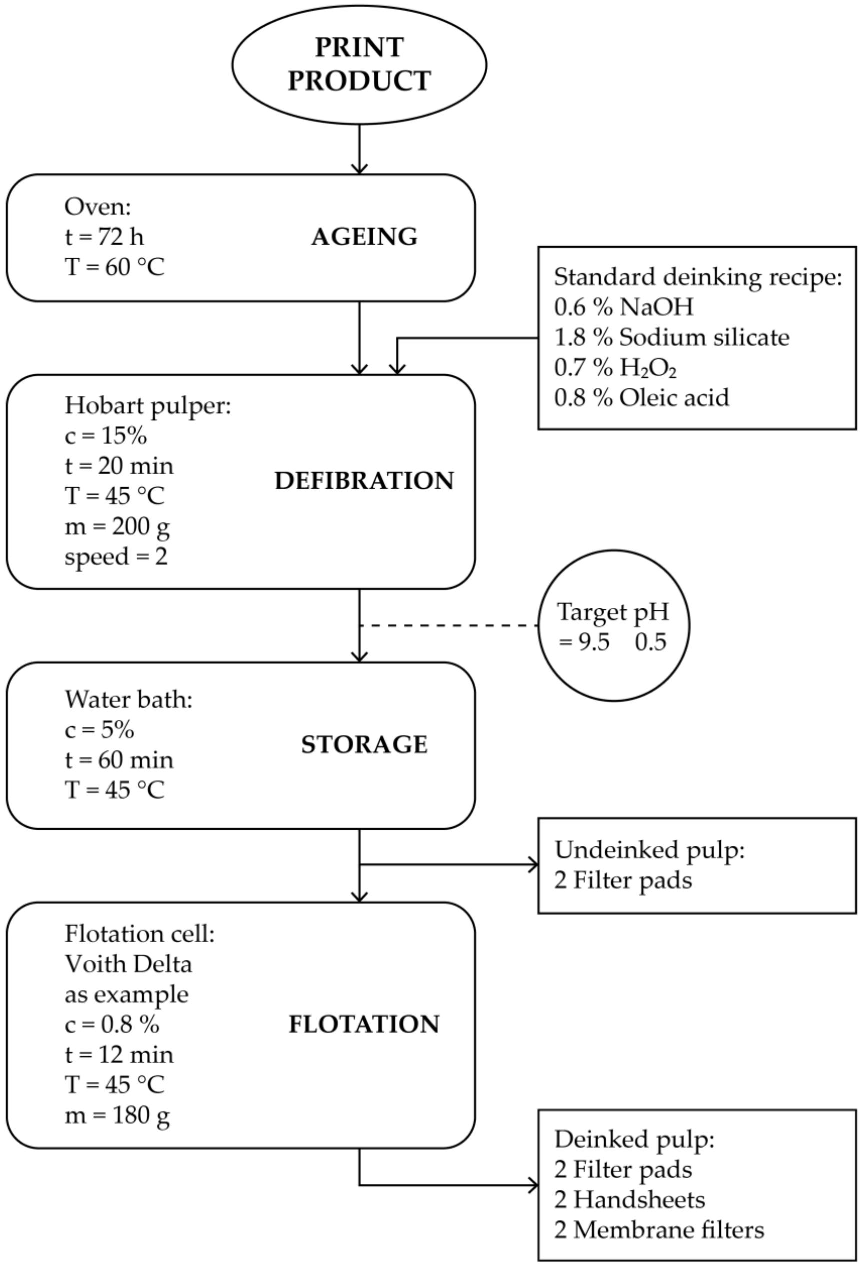


Figure 2. Procedure for testing recyclability/deinkability according to INGEDE Method 11 [63].

The number of dirt specks, i.e., residual ink particles, and their total area in the handsheets were determined via image analysis and data processing using Spec Scan 2000 (Apogee Systems Inc., Cape Canaveral, FL, USA) according to ISO 15755 [66]. The handsheets were scanned with a resolution of 600 dpi. The image scanned in grayscale mode was converted to black and white with a manual threshold adjusted to 100 (pixels under 100 were set to black, and those above 100 were set to white).

In Table 1, the samples that were recycled and analyzed are given, i.e., prints on label paper made via electrophotography (digital) and offset printing, without and with printed RFID antennas and with RFID antennas printed on the label paper itself.

Table 1. The designation and description of analyzed samples.

Designation	Description
toner	recycled digital print
toner + rfid SC	recycled digital print with RFID tag printed with SunChemical ink
ofset	recycled offset print
ofset + rfid SC	recycled offset print with RFID tag printed with SunChemical ink
ofset + rfid A	recycled offset print with RFID tag printed with Agfa ink
rfid SC	recycled unprinted paper with RFID tag printed with SunChemical ink
rfid A	recycled unprinted paper with RFID tag printed with Agfa ink

3. Results and Discussion

3.1. Durability

To evaluate the durability of CMYK prints, their optical density and color differences were determined. The test specimen was exposed to accelerating levels of exposure to an environmental factor (light, moisture, heat, and rubbing) for a fixed duration, and the results were compared to an internal benchmark. The changes in density and colorimetry relative to those measured for an untreated sample were calculated.

In Table 2, the measured colorimetric values and optical density of the CMYK prints and the colorimetric values of both label papers (Sample 1 and Sample 2) are presented. The color difference between the label papers is very small (ΔE*ab = 1.3), with Sample 2 being slightly lighter and brighter than Sample 1. Such a small difference cannot be seen by the untrained eye. Though the difference is small, it still has some effect on the colorimetric values of the CMYK prints. The color difference for the CMYK prints is between 3.5 and 6, meaning that an obvious difference in all four CMYK colors could be seen on the printed labels. The CMY prints on Sample 2 are lighter and brighter, and the K print is darker and duller than the prints in Sample 1.

Table 2. Colorimetric values L*, a*, b*, C*, h and optical density (D) of label papers (Sample 1 and Sample 2) and CMYK-color-printed label papers (CMYK prints Sample 1 and CMYK prints Sample 2) determined for each color.

			L*	a*	b*	C*	h	D
Label paper	Sample 1		91.64	−1.26	−21.87	21.91	267	/
	Sample 2		92.77	−0.74	−22.31	22.32	268	/
CMYK prints	Sample 1	C	49.97	−24.44	−62.74	67.34	249	1.607
		M	42.55	70.95	−16.84	72.92	347	1.495
		Y	83.08	−12.37	81.11	82.05	99	1.398
		K	14.74	−1.21	−6.51	6.62	259	1.706
CMYK prints	Sample 2	C	52.80	−27.22	−64.29	69.82	247	1.686
		M	43.30	74.50	−20.19	77.19	345	1.510
		Y	84.90	−12.66	84.41	85.35	99	1.439
		K	9.29	−1.46	−4.82	5.03	253	1.957

As shown in Table 3, the differences in the optical density (ΔD) of the CMYK prints before and after treatment are small, and in cases where the values are <0.05, they are negligible. The changes in optical density were expected to be greater after a longer exposure time (144 h) and a higher number of rubbing strokes (1000). As shown in Table 3, this outcome was confirmed in most cases. After the CMYK prints were exposed to light for a longer period, the change was slightly greater, with the greatest change being observed for Sample 2. The biggest difference after light treatment was exhibited by the yellow print. Almost all the CMYK prints experienced a decrease in optical density after exposure to the light, whereas after the moist–heat treatment, optical density increased in all cases. The largest increase was exhibited by the black print, whereas the smallest was observed for the yellow print. Higher temperatures can lead to the thermal expansion of materials, while moisture can lead to swelling, leading to more interaction, resulting in thicker and more even ink surface coverage, which affects reflection and induces an increase in optical density. The change was greater for Sample 1 and with a longer duration of treatment, whereas, for Sample 2, a longer treatment duration had almost no effect on the print. The optical density of the CMYK prints decreased minimally after 500 rubs; also, after 1000 rubs, the changes were still very small. Also, the difference between label papers was small; the changes in optical density were slightly smaller for Sample 1 compared to Sample 2. For both labels, there was practically no color transfer to the unprinted paper, meaning that good adhesion between the label paper and printing ink existed, resulting in excellent resistance to dry rubbing.

Table 3. The difference in optical density (ΔD) of CMYK colors after different treatments of CMYK prints (72 or 144 h; 500 or 1000 strokes).

		Light		Moist-Heat		Dry-Rub	
		ΔD—72	**ΔD—144**	**ΔD—72**	**ΔD—144**	**ΔD—500**	**ΔD—1000**
CMYK prints Sample 1	C	0.005	0.020	0.193	0.194	−0.017	−0.007
	M	−0.020	−0.029	0.118	0.136	−0.006	0.003
	Y	−0.049	−0.063	0.042	0.064	−0.016	0.008
	K	0.0006	0.006	0.299	0.306	−0.007	−0.003
CMYK prints Sample 2	C	−0.005	−0.021	0.026	0.024	−0.033	−0.064
	M	−0.015	−0.057	0.054	0.057	−0.022	−0.026
	Y	−0.017	−0.080	0.042	0.029	−0.029	−0.039
	K	0.0066	−0.011	0.092	0.090	−0.045	−0.037

Exposure to light had a significant effect on both label papers: the color difference was already over 8 after 72 h of treatment. Such a high value indicates a very obvious difference in the color of the paper and could influence the color of the print. In Figure 3, the color difference (ΔE*ab) of the CMYK colors obtained after the accelerated aging of CMYK prints with a xenon lamp after 72 and then 144 h is presented. The greater color change of label paper Sample 1 is reflected in the greater color change of the yellow print (Y), which can be categorized as an obvious difference. Also, the cyan print (C) showed an obvious difference after only 72 h of exposure to the light on both printed samples. The magenta print (M) showed a medium-level difference, whereas no visible difference was seen for the black print (K). The change in chroma was highest for the yellow print (Y), which became duller. A very small difference between colors can be perceived when the values are between one and three, and a color difference of up to three is acceptable for the printing industry. Though both labels performed well, the CMYK prints on Sample 2 showed better light fastness.

The colorimetric values of the CMYK prints exposed to elevated humidity (65%) and temperature (80 °C) are given in Figure 4. After moist–heat treatment, an obvious color difference was noticeable in most cases. In the case of the cyan print (C), the color difference was very obvious for both labels, whereas for the black print (K), it was only noticeable for Sample 1, which later also showed an obvious difference in the case of the magenta print (M). The smallest color change was evident for the yellow print (Y) on both labels: the change in chroma showed that they became duller, whereas other colors became brighter.

From Figure 5, it can be gleaned that most values are lower than 3, indicating the acceptable color change and good rub resistance of the prints. The minimum ΔE*ab for seeing a difference is about 2, and it is higher for saturated colors. Sample 1 showed a greater color difference after rubbing than Sample 2, especially in magenta and yellow prints, where a medium-level difference was obtained.

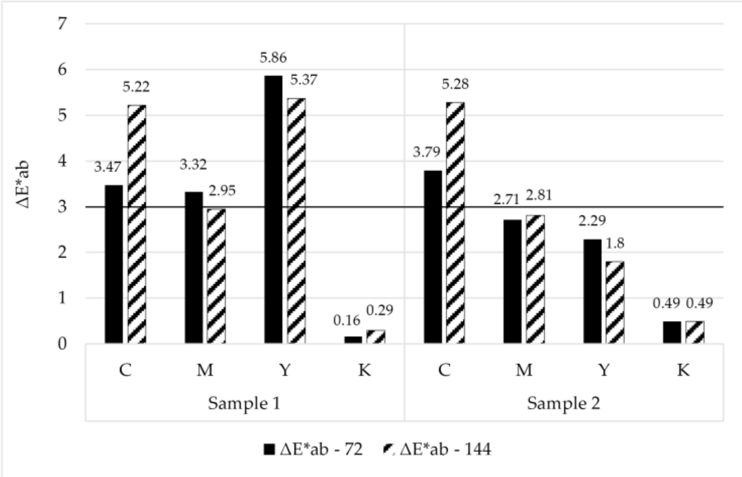


Figure 3. Color difference, ΔE*ab of CMYK colors, determined on CMYK prints (Sample 1 and Sample 2) after exposure to light for 72 and 144 h.

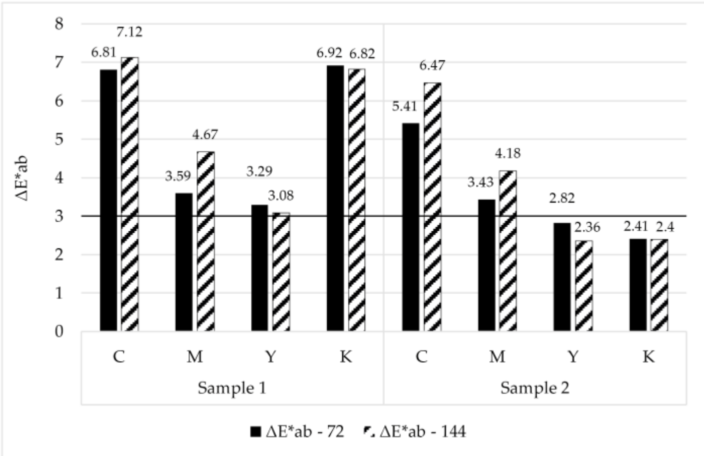


Figure 4. Color difference, ΔE*ab of CMYK colors, determined on CMYK prints (Sample 1 and Sample 2) after moist–heat treatment for 72 and 144 h.

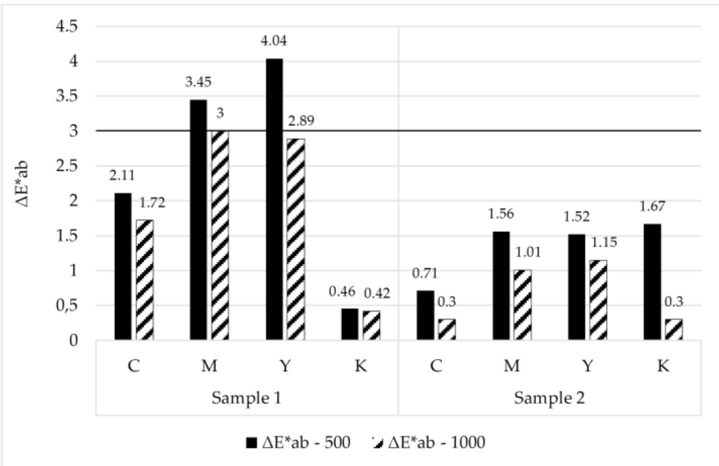


Figure 5. Color differences, ΔE*ab of CMYK colors, determined on CMYK prints (Sample 1 and Sample 2) after dry-rubbing treatment with 500 or 1000 rubbing strokes. The line at ΔE*ab = 3 indicates an acceptable color change.

These analyses have shown that the color fastness of CMYK prints can be declared as very good after most treatments for Sample 2, as values of ΔE*ab below 3 indicate a visible but small color difference, which is acceptable. Sample 1 has very good resistance to rubbing, whereas with light and moist–heat treatment, a clearly visible color change was seen for most colors.

3.2. Recyclability

To evaluate the recyclability of printed electronics, an analysis of the deinkability of printed label paper equipped with RFID tags was performed. The RFID antennas were printed using a conductive functional ink composed of micro- or nano-sized silver particles on printed and unprinted label paper.

The prints made using offset- and electrophotographic printing techniques were compared. These two techniques were selected because they both yield excellent print quality and are often used in label printing. Offset printing is one of the most cost-effective methods for printing in high volumes, whereas electrophotography is a better option for lower-cost low-volume printing. Both techniques, offset, and electrophotography, are also suitable for flotation deinking and yield good deinkability results.

Recyclability was evaluated using the following parameters:

- ISO Brightness, a diffuse blue reflectance factor (ISO brightness) of pulps, papers, and boards, which indicates the amount of blue light reflected from the surface of the paper;
- Luminosity, describing the average spectral sensitivity of the human visual perception of brightness;
- L*a*b* values;
- ERIC value—an effective residual color concentration is a measure of the darkening effect of the remaining ink in a paper due to residual color;
- Ink elimination, IE, which indicates the ink removal rate;
- Number of dirt particles and their total surface area (ink residues).

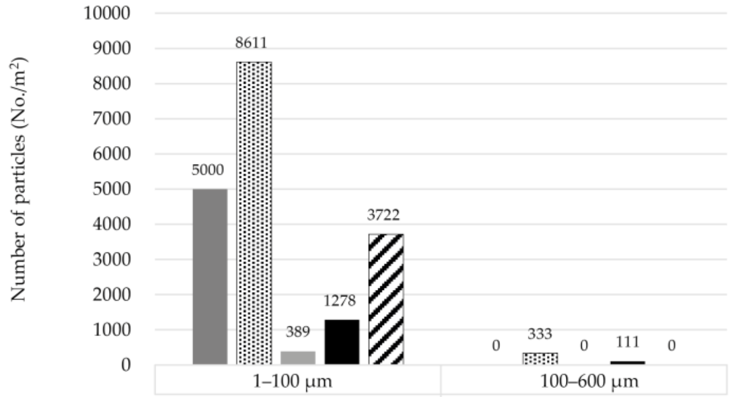
In Table 4, the measured parameters for the assessment of the printed papers' recyclability are presented. A comparison of the recyclability parameters and ink removal efficiency showed that there were only minor differences between the deinked digital and offset prints in terms of optical properties, as the difference in ISO brightness, luminosity, and lightness was 0.4–1.3% or lower. For the recycled offset prints, a higher percentage of ink elimination and a slightly lower ERIC value were determined.

Table 4. Assessment parameters for determination of printed papers' recyclability.

	Luminosity (%)	ISO Brightness (%)	L* (/)	a* (/)	b* (/)	ERIC (ppm)	IE (%)
toner	64.7	82.8	84.4	1.14	−3.11	553	22.7
toner + RFID SC	62.6	80.4	83.2	1.07	−2.93	638	37.1
offset	64.4	81.7	84.1	2.36	−3.62	503	30.7
offset + RFID SC	61.1	80.7	82.3	1.83	−3.49	685	27.6
offset + RFID A	79.9	82.1	91.6	8.25	−1.64	90	44.4
RFID SC	81.1	83.4	92.3	0.78	−1.66	88	6.0
RFID A	80.7	83.5	89.0	0.59	−1.78	105	5.4

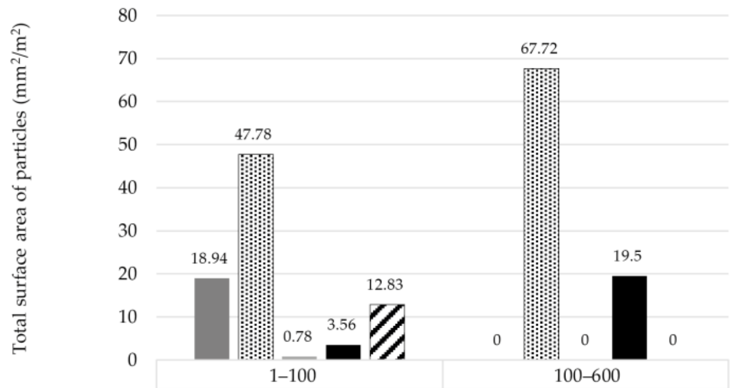
The measurements of ISO brightness, luminosity, and colorimetric properties show that most of the printing ink was removed and that the presence of the functional conductive ink from RFID antennas had no major effect on optical properties. Between the recycled samples wherein an RFID tag was present and the samples without an RFID tag, the differences in ISO brightness, luminosity, and colorimetric components were small, ranging up to 5%, except for the recycled offset print with an RFID tag printed with nano-silver conductive ink. In this case, the differences were larger, ranging up to 24%. Comparison of the deinkability of offset-printed label paper using micro- and nano-silver conductive ink showed higher luminosity, ISO brightness, and lightness, as well as greater ink elimination and lower ERIC values, but a higher number of small dirt particles for offset-printed label paper with RFID tag printed with nano-silver conductive ink.

The presence of dirt specks, particles from printing, and functional ink was evaluated according to the number of particles in several size classes and their total surface area. In the case of the recycled print samples with an integrated RFID tag, there were more impurities, i.e., ink residues, with a greater total surface area. With electrophotography, there were 5000 particles, and there were 8944 with the added RFID tag, with an area of 115.50 mm^2/m^2. With offset printing, there were 389 particles, and there were 1389 particles when an RFID tag was incorporated, with an area of 23.06 mm^2/m^2. More particles (3722 no./m^2) with a greater particle surface area (12.83 mm^2/m^2) were present in the recycled offset print incorporating an RFID tag and printed with nano-silver conductive ink (Figures 6 and 7).



	1–100 µm	100–600 µm
Toner	5000	0
Toner+rfid SC	8611	333
Ofset	389	0
Ofset+rfid SC	1278	111
Ofset+rfid A	3722	0

Figure 6. Number of particles, i.e., ink residue in deinked samples.



	1–100	100–600
Toner	18.94	0
Toner+rfid SC	47.78	67.72
Ofset	0.78	0
Ofset+rfid SC	3.56	19.5
Ofset+rfid A	12.83	0

Figure 7. The total surface area of particles, i.e., ink residues in deinked samples.

ERIC is a measurement of the overall optical effect that residual ink, mostly of sub-visible size, has on the appearance of a sheet. As shown in Table 4, the effective residual ink concentration (ERIC) is higher in the recycled samples wherein functional ink was also present (553, 638, 503, and 685 ppm), except for the case where nano-silver functional ink was used for printing (90 ppm).

The percentage of removed color (IE) was also higher for the recycled prints with an incorporated RFID tag. The presence of impurities, particles from printing, and functional ink was evaluated according to the number of particles in several size classes and their total surface area. Most of the particles present were small, with a diameter of up to 100 µm, and only a negligible number of larger particles were present. This is in line with the optical properties of the recycled samples, where, despite small differences, these properties were slightly worse for the prints incorporating an RFID.

4. Conclusions

Our comprehensive study investigated the durability, recyclability, and deinkability of printed label paper, particularly focusing on self-adhesive labels equipped with an RFID tag. In this research, we examined the impact of various environmental factors on the color fastness of CMYK prints and assessed the compatibility of RFID with recycling practices.

Labels give us a first impression of a product, so they must have good print resistance to various factors. Color fastness is crucial. The increasing use of electrophotography in label printing due to its high quality—which approaches the performance of offset printing in terms of durability, efficiency, and versatility, connected with low costs at low volumes and more sustainable printing compared to traditional printing techniques—was the main reason that we chose electrophotography for label printing. The durability tests conducted on CMYK prints, especially Sample 2, demonstrated very good resistance to environmental stresses like light, moisture, heat, and physical rubbing. Very good resistance to rubbing was also seen for Sample 1, indicating good print quality of both labels. As Sample 1 demonstrated an obvious color change after light and moist–heat treatment, the replacement of the most-used self-adhesive label (Sample 2) employed by a Slovenian company with a fully biodegradable, compostable novel commercial label (Sample 1) would only be possible in less demanding applications because it cannot guarantee the same print quality.

On the other hand, the removal of printing ink in the recycling process is crucial for obtaining recycled fibers of good quality. The second goal of our research was to assess the recyclability of printed label paper equipped with printed electronics. Recyclability was assessed, and a comparison between the deinkability of offset and digital prints and two conductive functional inks was made. Our experiment showed that the recycling process was not greatly disrupted by the presence of RFID tags. The deinkability of both electrographic and offset prints was good, with minor differences in optical properties. Most printing inks could be effectively removed, with offset prints showing higher ink elimination percentages. Good deinkability was also determined for the printed RFID antennas on both offset and electrographic prints, with only a small deterioration in optical properties. Better optical properties after deinking were obtained when nano-silver conductive ink was used to print RFID antennas, the sample is brighter and lighter. This suggests that printed antennas can be integrated into labels without greatly compromising recyclability. However, the increase in dirt specks in the recycled fibers due to the presence of Ag particles in conductive ink is a concern that needs to be addressed.

The applicability of this research is evident in the selection of materials and printing techniques. As a printing substrate, classic wood-free label paper was selected instead of special papers for printed electronics. Both printing techniques, offset and electrophotography, are mature, widely used printing techniques, and so is screen printing for printed electronics. The selection of widely used materials and printing techniques ensures low prices and broadens the interest expressed by the industry in the use of printed electronics. It should be noted that in the future it is planned to expand the research to other combinations of printing substrates and inks to expand knowledge in this scientific field.

Further, the use of a recycling test that simulates process steps in an industrial deinking plant at the laboratory level can provide important information to the industry about the recyclability of printed RFID antennas. Good recyclability/deinkability complies with a circular economy and sustainability.

Our research emphasizes the potential of incorporating RFID technologies into label printing. While advancements in label technology offer significant benefits for product tracking, they also pose challenges concerning recycling and environmental sustainability. This balance is crucial for the sustainable evolution of the packaging industry.

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