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DEVELOPMENT AND RECYCLABILITY OF STAINLESS STEEL-BASED NATURAL CONDUCTIVE INK FOR SUSTAINABLE RFID TAG ANTENNAS

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Abstract: *Due to developing economies and increasing fast consumption, products are constantly in motion globally, causing the need to monitor the products from production to sale and to keep track of their stocks regularly. At this point, smart technological products such as RFID tags make tracking easier and contribute to the smooth progress of business by containing product information. However, these tags, attached to almost every product, contain printed electronics and, therefore, valuable metals. These metals, thrown away after the product is purchased, cause e waste and environmental pollution. It may be possible to recycle these metals for reuse at the end of the tags' life. Therefore, in addition to the durability of the printed RFID tag antennas, recyclability also becomes important in sustainability. In this study, a natural conductive ink containing macroparticles of stainless steel and natural resin was developed. RFID tag antennas were printed by screen printing on PE-coated papers using this novel ink. The recycling of metals on the printed tags was studied using the deinking technique Ingede 11. Screen-printed tag antennas on paper were deinked, and the rates of metal particles remaining in paper residues and wastewater were determined. The results indicate that the metals' particle size and the interaction and adhesion of ink to paper are important for a successful deinking process in conductive inks.*

Key words: deinking, printed electronics, sustainability, recyclability, conductive ink, RFID tag

1. INTRODUCTION

Printed electronics have been widely used in sensors, antennas, and radio frequency identification (RFID) tags. RFID tags are the most used printed electronic components in commercial applications. They are widely applied in automotive, logistics, textiles, food and health industries for pallet or item level tracking. In addition, RFID tags are utilized to monitor manufacturing processes from beginning to end in some sectors for inventory management, authentication, and security. Over time, RFID printing technology has significantly advanced. Thus, the development of high-performance and low-cost conductive inks is becoming important for producing RFID tags. Besides, recyclability studies of the metals in conductive inks have been started due to the shortage of metals on earth and environmental issues caused by their wastes. The wastes of these rare earth metals can be recycled and reused as raw resources for new products to achieve a zero-waste economy. Metallic inks, such as silver inks, have been mostly preferred due to their high conductivity (Jansson et al., 2022; Riaz & Riaz, 2017) in RFID tag production. However, the high cost and corrosion problems of silver limit its application and the adoption of silver-containing conductive inks (Zhou et al., 2024). Additionally, a sintering process is needed to improve their conductivity and reinforce the ink-substrate bonding (Pan et al., 2018). Copper and aluminium can be used in conductive inks to obtain high conductivity (Rida et al., 2009). But they still have several negative aspects, like being incompatible with flexible electronics, and the costs of coating and high-temperature sintering to prevent oxidation (Campbell et al., 1999). There are studies for adopting new materials, such as stainless steel, for affordable and high-performance conductive inks to overcome these problems (Duman et al., 2021). However, the additional and essential point that should be considered for these materials is reusability (Hayta & Oktav, 2019). The RFID tags are thrown away after the end of their life cycle. According to IDTechEx forecasts, 39.3 billion passive RFID tags have been sold in 2023, up from 33 billion in 2022, indicating a 20% year-on-year increase (Chang & Das, 2024). When environmental sustainability is considered, the metals on RFID tags make up a respectable amount that can be recycled. Thus, recyclability and biodegradability of the substrate materials have become another important issue for RFID tags. Paper is a good alternative as a substrate for printed electronics because it is a light, low-cost and biodegradable material (Özdemir &

Oktav, 2023). Little research has been done on recycling RFID tags to reuse the metals in producing new tags (Atkinson, 2017; Déprès et al., 2023; Huttle, 2009; Pogačar, Bolanča-Mirković & Gregor-Svetec, 2024; Southwell, 2023). All were applied to silver-printed tags, and most of the silver from the ink ended up in recycled paper residues or wastewater. The effectiveness of the deinking was not good enough due to the silver particle size. To recycle the metals, the deinking process was applied to the paper-based printed RFID tags. First, the samples were prepared and aged in the oven to conduct the deinking process. Then, a pulp was produced by adding the deinking chemicals and defibrated. The fibres swelled in the solution, and then paper fibre residues were collected using a loose filter. Afterwards, a magnet was immersed in the filtered water solution to collect the stainless steel particles passed through the filter. For a successful deinking process, an electromagnet can be used not to lose any metal particles when removing them from the magnet. Additionally, the hydrophobicity of the ink, the particle size of the conductive materials inside the ink, and the ink's interaction with the substrate are also important. In screen printing, the absorption and penetration of ink into the paper structure causes the conductive ink to adhere to paper surfaces (Kavčič, Karlovits & Zule, 2020). This research aims to analyze the recyclability of paper-based printed RFID tags using a conductive ink containing stainless steel macroparticles. The INGEDE 11 method was used for deinking the screen-printed RFID tags. Stainless steel ink particles in paper residues were analyzed using their zoomed images, and their extracts in wastewater were determined. Possible health risks of using stainless steel as a conductive ink should be considered; some particles can be released into the water in industrial recycling and deinking practices within the e-waste sector.

2. MATERIALS AND METHODS

2.1 Printing

In the presented work, RFID tag samples were screen printed using stainless steel spherical powder as a conductive element to examine the recyclability of metals in RFID tags. Type 316 Stainless Steel powder with 5 µm particle size and 99.8% purity and natural pine tree resin (Hayta & Oktav, 2020) were mixed to obtain a natural conductive ink and used with 40% ratios of stainless steel in weight due to our previous work (Duman et al., 2021). Type 316 steel contains 2-3% molybdenum, increasing corrosion resistance and strength at high temperatures, and 0.08% carbon. PE-coated standard bulk paper (285 g/m^2) was used as a substrate to substitute the RFID tags. The screen mesh with 77 mesh count was used to screen print the RFID tags, which was made of polyester fabric with a printable antenna design. A 15 cm rubber squeegee with 70 Shore A hardness, 75o and 3 bars pressure was used, and 26 N/cm screen tension was applied for the screen in printing. Before printing, the prepared ink with stainless steel and pine resin was heated at 40°C for 10 minutes to obtain homogeneously dispersed steel particles in the ink. The printed RFID tag antennas had a line thickness of 0,35 mm and were printed in one layer using a semi-automatic screen-printing machine. Then, the samples were aged at 60°C for 72 hours in an oven, simulating 3-6 months in a normal environment according to INGENE Method 11:2018 used for recycling (Figure 1).

2.2 Adhesion

Zoomed images are obtained on stainless steel containing ink printed on PE-coated paper to determine conductive ink adhesion on the paper substrate and the paper fibre residues after deinking.

2.3 Deinking

The efficiency of deinking was evaluated with the help of the INGEDE Method 11. The samples were cut into pieces after the ageing process. A solution was prepared from tap water and mixed with deinking chemicals, as shown in Figure 1. The paper samples were immersed in the solution and then defibrated in a laboratory defibrillator for 20 min at 45°C by keeping the pH value at 9.0. Afterwards, the mixture was filtered using a coarse paper filter. The zoomed images of deinked paper residues, captured with a scanner (300 PPI), were used to evaluate their surface properties. The amount of stainless steel particles was measured in the deinked fibre residue and the related water extract.

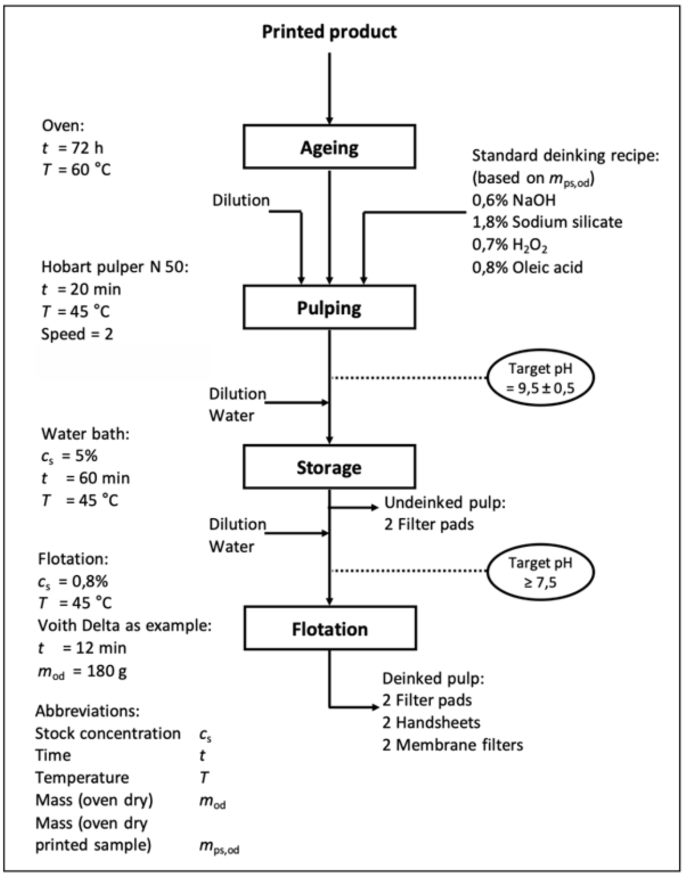

Figure 1: The standard procedure of the Ingede Method for testing the deinkability of printed papers (Ingede Method, 2018)

3. RESULTS AND DISCUSSION

The adhesion of conductive ink to the substrate is a key factor that can significantly affect deinking, and the homogenous distribution of the metals is vital for strong signals for RFID tags. As shown in Figure 2, the stainless steel particles were homogeneously distributed as desired in the printed samples on paper. The ink was heated before printing and samples were cured after printing to eliminate agloramation of metal particles. After printing the samples, the pulp is prepared, defibrated using the Ingene method and filtered to remove the paper fibre residues from the solution, as seen in Figure 3. The stainless steel particles were settled down in the remaining solution.

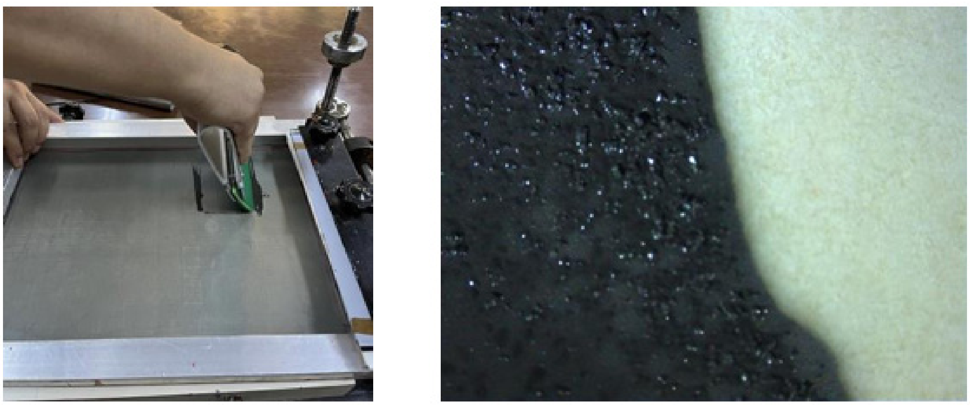

Figure 2: The photo of the screen-printing process for sample printing and the zoomed image of the printed sample with stainless steel containing conductive ink, respectively, from left to right

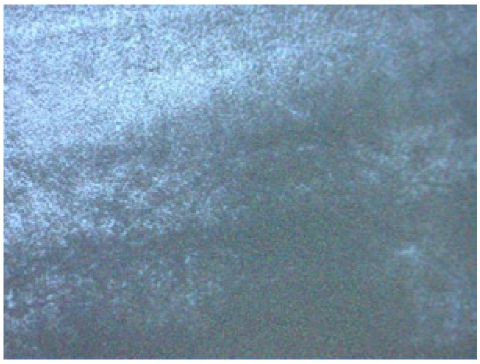

Figure 3: Filtration process of the pulp and stainless steel particles settled down in the wastewater after filtration of paper fibres, respectively, from left to right

The total weight of paper residues and stainless steel particles obtained as a result of the deinking process, as seen in Figure 4, is presented in Table 1. Approximately 90% of stainless steel particles are recycled by deinking printed samples. By deinking, the PE coating of the paper was removed together with the ink from the paper. Paper fibres in the pulp residues contain a few stainless steel particles, as seen in Figure 5, which can hardly be seen on the surface of the paper fibre residues. This can be attributed to the fact that ions in stainless steel bind more strongly to coatings than fibres.

Table 1: Stainless steel content of un-deinked and deinked printed PE-coated paper samples

Sample	Total Sample Weight (g)	Total Stainless Steel Particle Weight (g)
Unprinted paper	0.47	0
Un-deinked paper	0.69	0.09
Deinked paper residues	0.66	0.08

Figure 4: Wastewater, paper fiber residues and stainless steel particles obtained by the deinking process, respectively, from left to right

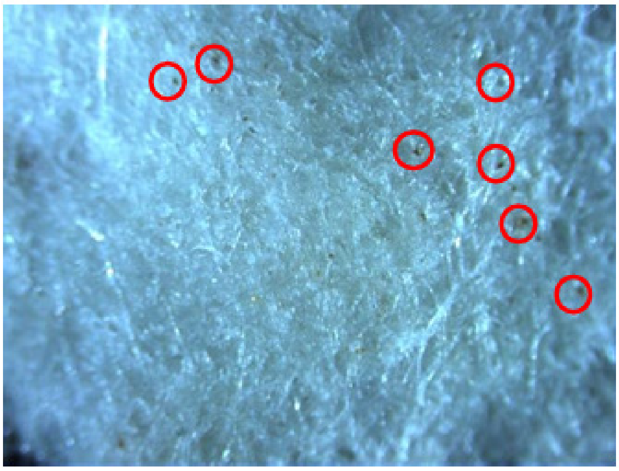

Figure 5: Stainless steel particles, circled in red, on the filtrated paper fiber residues

Stainless steel is an iron-based alloy that contains nickel (Ni), chromium (Cr), and iron (Fe) and is characterized by high corrosion resistance. When exposed to oxygen or water, a chromium-rich oxide layer forms on the surface of stainless steel, preventing the material's leaching and corrosion. Additionally, nickel improves the re-passivation of the surface oxide on stainless steel, limiting the release of metals from the alloy, even in aggressive environments depending on pH and temperature (Taxell & Huuskonen, 2022). Stainless steel accumulation in city water could harm living organisms if they enter the food chain. Modern deinking plants typically recycle a significant amount of their process water, which could lead to the accumulation of stainless steel if it leaches from paper materials. There are various methods, such as membrane filtration, ion exchange, and adsorption, to eliminate stainless steel efficiently from the water (Kavčič, Karlovits & Zule, 2020).

4. CONCLUSIONS

The research demonstrated that paper with printed conductive inks can be successfully deinked, though the process's effectiveness can be influenced by both the substrate and ink composition. Additionally, the potential release of stainless steel particles in municipal water systems during deinking should be taken into account to avoid environmental issues. It is evident that the presence of fillers and coatings on the paper impacts the removal of stainless steel particles from the fibre structure. The samples were screen-printed with stainless steel based natural ink and dried in the oven to evaporate the solvent and increase the penetration of the ink. Then, they were deinked using the procedure given in the Ingede method to recycle the conductive metals in the printed ink. The results show that the deinking process was efficient with the ink containing stainless steel particles printed on a PE-coated paper substrate. More than 90% of stainless steel particles were removed with the deinking process due to poor adhesion to paper fibres. Although the stainless steel content in the printed sample was higher than the deinked residues, it can be explained by the adhesion of conductive stainless steel particles to the materials used to collect them due to static electricity.

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