

**EFFECT OF PAPERBOARD SURFACE MODIFICATIONS ON ELECTRICAL  
CONDUCTIVITY OF PRINTED UHF RFID ANTENNAS**

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**ABSTRACT**

The effect of surface roughness and water contact angle of commercial paperboard before and after surface modification by calendering, coating and calendering and plasma treatment on the functionality of UHF RFID antennas printed with thermal transfer aluminum ribbon was evaluated. A hydrophilic surface was created by coating or plasma treatment, which improved the wettability of the paperboard surface, the spreading of the thermoplastic tie layer and the adhesion of the conductive aluminum layer. A new paper product was created with permanent surface wettability by coating, without the need for plasma treatment before printing. The plasma treatment provided time-limited wettability, needed only during printing, and made it possible to restore the original hydrophobic surface of the paperboard. In addition to the meaning of these surface modifications, the importance and need to reduce the surface roughness was confirmed, as the higher surface roughness of the paperboard limited the effect of the plasma treatment in terms of its printability and the functionality of the printed aluminum antenna. The printability of the paperboard and the functionality of the printed antennas were evaluated using electrical conductivity. The electrical conductivities of the dipole and inductor loop of the UHF RFID antennas printed on modified paperboards varied depending on the antenna design.

**KEYWORDS:** Paperboard, calendering, coating, plasma, thermal transfer printing, UHF RFID antenna, electrical conductivity.

## INTRODUCTION

An RFID antenna printed on paper substrates is an effective solution for low cost and environmentally friendly tag designs because the printed antenna enables RFID tags to be embedded into product packages, which further reduces converting costs during deployments. Printed antennas are usually applied to different plastic films (Chin et al. 2008, Janeczek 2010, Arazna et al. 2017) or paper substrates (Merilampi et al. 2007, Rida et al. 2009, Lakafosis et al. 2010, Xi et al. 2011, Zichner and Bauman 2011, Öhlund and Andersson 2012, Bollström and Toivakka 2013, Kavčič et al. 2014, He et al. 2016, Gigac et al. 2020a,b, Gigac et al. 2021). There are many aspects of paper that make it an excellent candidate for a low cost and environmentally friendly substrate for printed electronics. The different types of paper have various density, coating, thickness, texture and dielectric properties. Paper substrates are becoming preferred to plastic films for RFID antennas as they are more environmentally and economically advantageous. A major challenge to printing antennas on the paper substrates, has been to match the electrical conductivity needed, which is readily obtained on plastic films.

Paper has a rougher surface compared to the plastic film. Irregular surfaces and structural properties of conventional papers require higher ink consumption, allowing them to be used only for electronic components with lower resolution or print quality requirements. In order to improve printability, research of paper substrates is aimed at improving surface smoothness and absorption properties. Coating and smoothing processes in a calender or hot stamping machine can be used to finish the surface of the paper (Gigac and Fišerová 2022). The paper surface is usually smoothed with a dispersion coating consisting of mineral pigments and organic binders. The smoothness of the paper surface depends on composition of coatings, the amount and layers of the coating and the final surface finish. Depending on the composition of the coatings, properties such as smoothness, porosity, permeability and surface energy as well as optical properties (brightness and opacity) can be varied. The surface properties of papers can be adjusted at the same time to achieve the desired functional properties, such as water, oil and grease resistance, low vapor and gas permeability (Long et al. 2015, Brodnjak and Todorova 2017, Ilyas et al. 2018, Kopacic et al. 2018, Gigac et al. 2018), and flame retardation (Sonnier et al. 2018).

Low-temperature plasma processing of surfaces and interfaces is an interesting option for applications in flexible and printed electronics where surface cleaning, activation or functionalization are required (Vida and Homola 2021). During the plasma treatment variety of active species generated by plasma react with the surface of the treated material. These species introduce new functional groups and/or remove the contaminants from the surface. The mentioned processes could be the reason for the improvement of the surface properties such as wettability, adhesion and printability (Roth 2001). As an effective technique for modifying natural polymers, such as cellulose-based materials, low-temperature plasma has been used, which allows a predictable change in their physical and chemical properties for various practical applications (Assender and Windle 1997, Mukhopadhyay et al. 2002, Laguardia et al. 2005). In particular, plasma processing of paper changes its layer properties (Filatova et al. 2009). Different types of the discharge, as well as treatment conditions on various paper types, have

been reported already in case of atmospheric (Dubreuil and Bongaers 2008, Pykönen 2010, Garcia-Torres et al. 2014) and low-pressure (Vesel et al. 2007, Filatova et al. 2009, Balu et al. 2009, Obeso et al. 2013). Surface modification by cold plasma allows the change in the surface chemistry of paper without altering the bulk material properties. An innovative plasma-activation technology based on a diffuse coplanar surface barrier discharge has been developed and patented (Šimor et al. 2002, 2003, 2004, Černák et al. 2004, 2009, 2011, Skácelová et al. 2013, 2016), which has the potential to move a step closer to the paper and web finishing industry requirements, especially at high processing speed. Low-temperature atmospheric discharge with runaway electrons (ADRE) operating at atmospheric pressure was used for bacteria decontamination on surface of solid states including the inside of tight polymeric packaging (Maltsev 2006) as well as on for preservation of historical documents (Tiño et al. 2021).

Conventional production of RFID antennas was realized by etching of metallized plastic, which is costly and environmentally unfriendly, so there is an effort to produce printed RFID antennas. Different printing technologies are used: flexography, gravure printing, inkjet printing, screen and thermal transfer printing. Different printing technologies enable different accuracy, resolution and conductive layer thickness. Thermal transfer printing with Metallograph® conductive ribbon is a simple, fast and economical method of digital printing for electronic circuits, sensors and RFID antennas (Taylor 2017). Compared to printing techniques such as screen printing, flexography, gravure and inkjet printing, which use pastes and inks containing mostly silver nanoparticles, thermal transfer printing uses aluminum, which is about two orders of magnitude less costly for the equivalent conductivity of silver. Additionally, there are no fluids, no printing set up, no drying and no sintering of the silver nanoparticles. Without these additional steps the process has a very small footprint and takes less than a second to accomplish.

The aim of this work was to compare the effect of surface modification of an commercial paperboard by calendering, coating and calendering, and low-temperature atmospheric discharge with runaway electrons (ADRE plasma) on the surface roughness and initial water contact angle of paperboards as well as on the electrical conductivity of the printed UHF RFID antennas.

## MATERIAL AND METHODS

### Materials

The folding box board (FBB) is a commercial single-sided coated paperboard with a basic weight of 247 g·m<sup>-2</sup> and a specific volume of 1.68 g·cm<sup>-3</sup>. It contains chemical-mechanical pulp and the top layer is bleached chemical pulp, the uncoated bottom has a yellow tinge (Metsä). Melinex ST 505 (DuPont Teijin Films sp.) was used as polyethylene terephthalate (PET) film. Print templates of UHF RFID antennas are shown in Fig. 1. The dimensions of the meander antenna 12 (Talkin' Things) are 70 x 12 mm, the dimensions of the antennas 24 (Talkin' Things) and DB (Avery Dennison Smartrac DogBone) are 94 x 24 mm.

Metallograph® TTR ribbon (SPF-Inc., USA) with an aluminum layer thickness of 260 nm (Gigac et al. 2020b, 2021) contains the five layers-heat-resistant coating, PET carrier film, release layer, vacuum metalized (continuous) layer and thermoplastic tie layer, that bonds

the metalized layer to the substrate when heated in the print head to form the image.



Fig. 1: Designs of symmetric UHF RFID antennas (not in scale).

## Methods

Surface roughness  $OVS_{CLINO}$  (Optical variability of surface) of paperboards was evaluated by the photoclinometric method as optical variability of surface (Kasajová and Gigac 2013). Photoclinometry in the visible range of electromagnetic radiation is a promising method that may be used for on-line measurement of paper roughness. It describes the process of transformation of a 2D surface image into a map of various height levels. Incident light creates shadows (different gray levels). Paper is an anisotropic material, therefore it is necessary to obtain surface images from at least two directions: machine direction (MD) and cross direction (CD). The paperboards surface was scanned using charge-coupled device (CCD) Nikon Coolpix E4500 camera (Nikon Corporation, Japan) by inclined illumination at  $10^\circ$  from MD and CD. Optical variability of surface was calculated from image analysis using the program ImageJ.

Initial and dynamic contact angle (CA) of paperboards was measured using the OCA 35 optical tensiometer (DataPhysics Instrument, Germany). Contact angle was measured by sessile drop method. Wetting time was recorded by a CCD camera at the sequence  $20 \text{ frames s}^{-1}$  from the first contact of the liquid drop with the paperboard surface (50 ms) through 5 s. Contact angle was calculated as the average of 10 parallel measurements.

Calendering of paperboards was performed using a Kleinewefers laboratory calender FUS 80 (Kleinewefers GmbH, Germany) by two passes between paper and metal cylinder at a linear load in the nip of  $260 \text{ kN.m}^{-1}$ , a temperature of  $80^\circ\text{C}$  and a speed of  $5 \text{ m.min}^{-1}$ . The coated side was in contact with the heated metal cylinder.

Coating of paperboards was performed using a #2.5 metering rod and drying at  $120^\circ\text{C}$  in an oven. The coating colour contained silica pigment (Sipernat 310) and polyvinyl alcohol in a ratio of 60/40 with the addition of a wetting agent (Dynol 810) to increase the surface energy and hydrophilicity of coating. The basis weight of the coating was  $1.5 \text{ g.m}^{-2}$ .

Plasma treatment of paperboards and PET film was done by low-temperature ADRE (Atmospheric discharge with runaway electrons) plasma (Tiño et al. 2021). The effect of the plasma treatment strongly depended on the conditions used during the exposure of the paperboards to the plasma. The plasma conditions and process parameters of EST Ltd. device (Tomsk, Russia): atmospheric air pressure; power  $0.9 \text{ J.s}^{-1}$ ; voltage 230 V; interelectrode distance 2.5 cm; energy of electrons 125 keV; frequency 2000 Hz; exposure time 3 min, which was optimal for the examined paperboards.

Printing of UHF RFID antennas on paperboards and PET film was performed in a SATO CL4NX thermal transfer printer (Japan) with Metallograph® ribbon (SPF-Inc., USA) at a printhead pressure (left and right side) 5, darkness range A10, darkness adjust to 75% and printing speed  $50 \text{ mm.s}^{-1}$ .

Electrical conductivity of printed antennas was calculated as the inverse of the electrical resistance. Electrical resistance of printed antennas was measured using a multimeter UNIT-T, Model UT70B separately on the two parts of the antenna, on the dipole and on the inductor loop, which differ in the width and length of the conductive trace. The dipole UHF RFID antennas (Fig. 1) consist of the following main parts: the dipole, the inductor loop and the connecting part. The dipole allows communication between the reader and the RFID tag. The inductor loop adjusts the capacity of the RFID chip and transfers energy from the dipole to the chip. The connecting part connects the dipole to the inductor loop and transfers energy between them. In Fig. 2, the points of measurement of the electrical resistance of the dipole (1 and 2) and the inductor loop (3 and 4) on the antenna 24 are shown. Using electrical conductivity, the printability of the paperboard and the functionality of the printed antenna were evaluated.

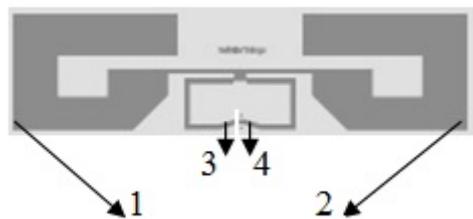


Fig. 2: Measurement of the electrical resistance of the dipole (points 1 and 2) and the inductor loop (points 3 and 4) of the UHF RFID antenna 24.

## RESULTS AND DISCUSSION

### Functional properties of paperboards

Commercial paperboard is not suitable for electronics printing because it has a porous and rougher surface than the plastic film commonly used for this purpose. Therefore, the surface of the commercial paperboard was modified by calendering, coating and calendering, and plasma treatment. Functional properties surface roughness  $OVS_{CLINO}$  and initial water contact angle WCA of commercial (FBB), calendered (FBB-CL), coated and calendered (FBB-CO-CL) paperboards without and after 3 min plasma treatment are given in Tab. 1. Commercial paperboard (FBB) had the surface roughness of 6.8% and the initial water contact angle of  $96^\circ$  because it has a hydrophobic coating, to be resistant to water. For printing electrical and electronic components, the surface roughness should be below 6% (Gigac et al. 2020a), which corresponds to the printing roughness of PPS (Parker print surf) below  $0.8 \mu\text{m}$  (Gigac and Fišerová 2022), and the initial water contact angle should be around  $62^\circ$ . After plasma treatment, the surface roughness of commercial paperboard (FBB) increased from 6.8% to 7.6%, while the contact angle decreased to  $60^\circ$ , thus its surface became hydrophilic. The surface roughness of commercial paperboard after calendering (FBB-CL) was 4.5% and the initial water contact angle  $93^\circ$ , so that its surface remained hydrophobic. After plasma treatment, the surface roughness of the calendered paperboard increased to 5.2%, while the initial water contact angle decreased to  $66^\circ$ . The surface roughness of commercial paperboard after coating and calendering (FBB-CO-CL) was 5.3% and the initial water contact angle  $64^\circ$ . After plasma treatment, the surface roughness of the coated and calendered paperboard increased to 5.8%, while the initial

water contact angle decreased to 49°.

*Tab. 1: Functional properties of commercial paperboard (FBB), after calendering (FBB-CL), coating and calendering (FBB-CO-CL) without and after 3 min plasma treatment.*

Paperboard	Plasma treatment time			
	0 min		3 min	
	Surface roughness OVS <sub>CLINO</sub> (%)	Initial water contact angle WCA (°)	Surface roughness OVS <sub>CLINO</sub> (%)	Initial water contact angle WCA (°)
FBB	6.8	96	7.6	60
FBB-CL	4.5	93	5.2	66
FBB-CO-CL	5.3	64	5.8	49

The surface roughness of commercial paperboard decreased by 33.8% after calendering, while after coating and calendering decreased only by 22.1%. The initial water contact angle of commercial paperboard did not change significantly after calendering, while after coating and calendering decreased by 33,3% due to the presence of a wetting agent in the coating colour. Plasma treatment for 3 min increased the surface roughness of commercial paperboard by 11.8%, while the initial water contact angle decreased by 37.0%. The surface roughness of calendered paperboard increased by 15.6% after plasma treatment, while the initial contact angle decreased by 29.0%. Plasma treatment had a similar effect on coated and calendered paperboard, the surface roughness of which increased by 8.6% after plasma treatment, while the initial water contact angle decreased by 23.4%.

After plasma treatment of the paperboards, the surface roughness increased and the initial water contact angle decreased, indicating increased surface wettability in accordance with the literature (Wenzel 1936). The results of the work (Pykönen et al. 2008) showed that the reduction of the contact angle due to plasma treatment cannot be explained only by the roughening of the surface. Only a part of the wettability change in plasma-treated samples is probably due to morphological changes in addition to chemical modification.

### **Electrical conductivity of UHF RFID antennas printed on paperboards**

Electrical conductivities of the dipole and inductor loop of aluminum UHF RFID antennas 12, 24 and DB printed on commercial (FBB), calendered (FBB-CL), coated and calendered (FBB-CL-CO) paperboard were compared with the electrical conductivities of the antennas printed on the PET film as well as after plasma treatment of paperboards and PET film. The electrical conductivity of UHF RFID antennas printed on PET film was used as a standard in evaluating the conductivity of antennas printed on paperboards, because the PET film has a low surface roughness and the initial water contact angle was 64°, which allows perfect printing with an aluminum thermal transfer ribbon.

#### *UHF RFID antenna 12*

In Fig. 3 are the electrical conductivities of the aluminum UHF RFID antenna 12 printed on commercial, calendered, coated and calendered paperboard and PET film. The antenna printed on the PET film had the electrical conductivity of the dipole 46 mS and inductor loop 169 mS. The antennas printed on the commercial (FBB) and calendered (FBB-CL) paperboard were electrically non-conductive. The electrical conductivity of the dipole and conductor loop of

the UHF RFID antenna 12 printed on the coated and calendered paperboard (FBB-CO-CL) was the same as the electrical conductivity of the antenna printed on the PET film.

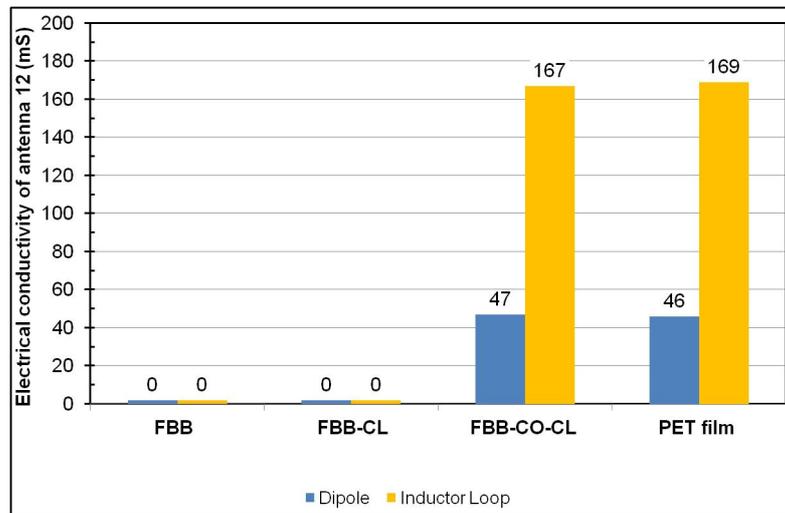


Fig. 3: Comparison of the dipole and the inductor loop electrical conductivity of the aluminium UHF RFID antenna 12 printed on commercial (FBB), calendered (FBB-CL), coated and calendered (FBB-CO-CL) paperboard and PET film.

Plasma treatment of commercial, calendered, coated and calendered paperboards had a significant effect on their surface roughness and initial water contact angle (Tab. 1). Therefore, the 3 min effect of plasma treatment on the electrical conductivity of the dipole and inductor loop of the UHF RFID antenna 12 printed on these paperboards and PET film was investigated (Fig. 4). The UHF RFID antenna 12 printed on plasma-treated commercial paperboard (FBB) was electrically non-conductive. After plasma treatment of the calendered paperboard (FBB-CL), the electrical conductivity of the dipole and inductor loop of the printed UHF RFID antenna 12 was the same as that of the antenna printed on the PET film without and with plasma treatment. After plasma treatment of coated and calendered paperboard (FBB-CO-CL), the electrical conductivity of the printed UHF RFID antenna 12 was unchanged. It has the same electrical conductivity of the dipole and inductor loop as the antenna printed on PET film without and with plasma treatment. It follows that after the plasma treatment of calendered paperboard (FBB-CL) the same electrical conductivity of the printed UHF RFID antenna was achieved as on the coated and calendered paperboard (FBB-CO-CL) without plasma treatment.

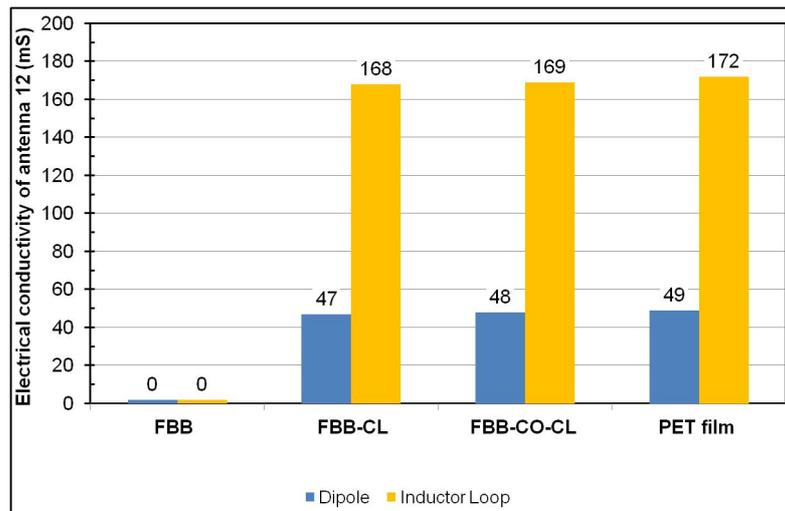


Fig. 4: Effect of plasma treatment (3 min) of commercial (FBB), calendered (FBB-CL), coated and calendered (FBB-CO-CL) paperboard and PET film on the electrical conductivity of the dipole and inductor loop of printed aluminium UHF RFID antenna 12.

#### UHF RFID antenna 24

The electrical conductivities of the dipole and inductor loop of the aluminum UHF RFID antenna 24 printed on commercial, calendered, coated and calendered paperboard and PET film are presented in Fig. 5. The antenna printed on the PET film had the electrical conductivity of dipole 128 mS and inductor loop 102 mS. The antennas printed on the commercial (FBB) and calendered (FBB-CL) paperboard were electrically non-conductive. The electrical conductivity of the dipole and inductor loop of the antenna printed on the coated and calendered (FBB-CO-CL) paperboard was the same as the electrical conductivity of the antenna printed on the PET film.

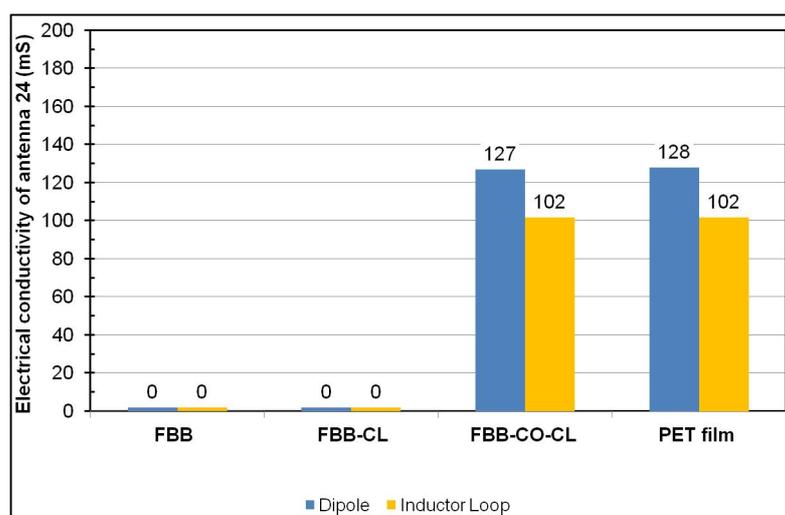


Fig. 5: Comparison of the dipole and the inductor loop electrical conductivity of the aluminium UHF RFID antenna 24 printed on commercial (FBB), calendered (FBB-CL), coated and calendered (FBB-CO-CL) paperboard and PET film.

The effect of the 3 min plasma treatment on the electrical conductivity of the dipole and inductor loop of the UHF RFID antenna 24 printed on commercial, calendered, coated and calendered paperboards and PET film was investigated (Fig. 6). The UHF RFID antenna 24 printed on plasma-treated commercial paperboard (FBB) was electrically non-conductive. After plasma treatment of the calendered paperboard (FBB-CL), the electrical conductivity of the dipole and inductor loop of the printed UHF RFID antenna 24 was the same as that of the antenna printed on the PET film without and with plasma treatment. After plasma treatment of coated and calendered paper paperboard (FBB-CO-CL), the electrical conductivity of the printed UHF RFID antenna 24 was unchanged. It has the same electrical conductivity of the dipole and inductor loop as the antenna printed on PET film without and with plasma treatment. It follows that after the plasma treatment of calendered paperboard (FBB-CL) the same electrical conductivity of the printed UHF RFID antenna was achieved as on the coated and calendered paperboard (FBB-CO-CL) without plasma treatment.

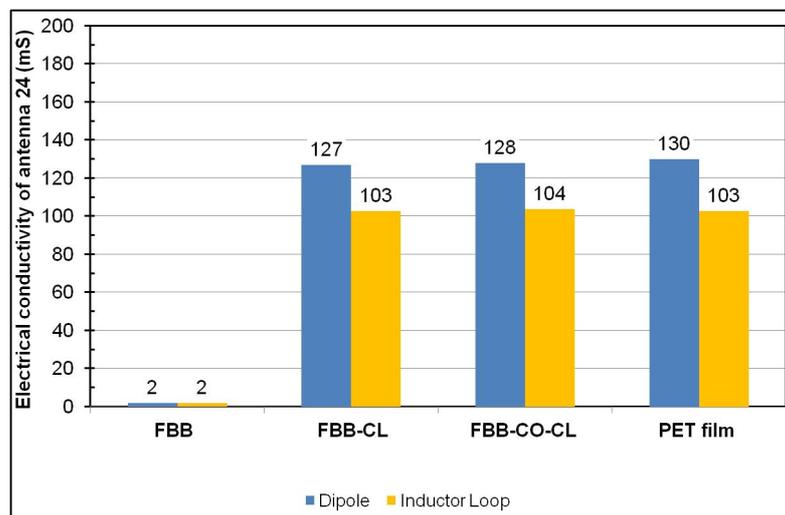


Fig. 6: Effect of plasma treatment (3 min) of commercial (FBB), calendered (FBB-CL), coated and calendered (FBB-CO-CL) paperboard and PET film on the electrical conductivity of the dipole and inductor loop of printed aluminium UHF RFID antenna 24.

#### UHF RFID antenna DB

In Fig. 7 are the electrical conductivities of the aluminum UHF RFID antenna DB printed on commercial, calendered, coated and calendered paperboard and PET film. The antenna printed on the PET film had the electrical conductivity of the dipole 222 mS and inductor loop 85 mS. The antennas printed on the commercial (FBB) and calendered (FBB-CL) paperboard were electrically non-conductive. The electrical conductivity of the dipole and inductor loop of the UHF RFID antenna DB printed on the coated and calendered paperboard (FBB-CO-CL) was the same as the electrical conductivity of the antenna printed on the PET film.

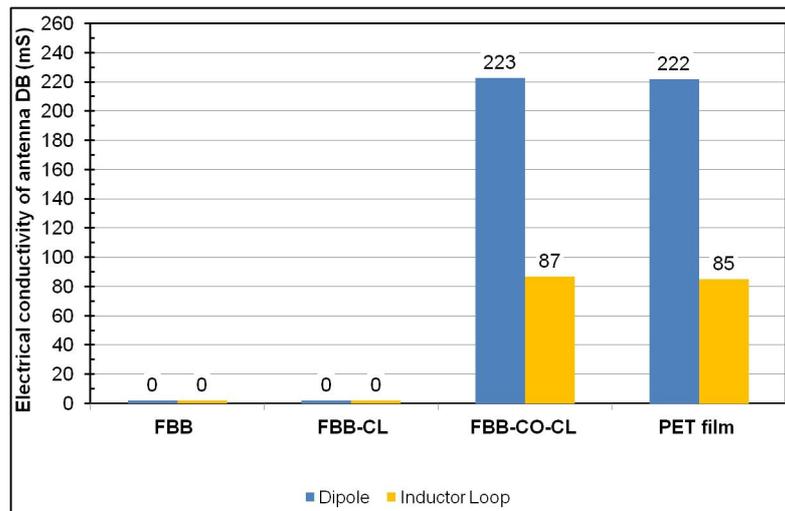


Fig. 7: Comparison of the dipole and the inductor loop electrical conductivity of the aluminium UHF RFID antenna DB printed on commercial (FBB), calendered (FBB-CL), coated and calendered (FBB-CO-CL) paperboard and PET film.

The effect of plasma treatment (3 min) on the electrical conductivity of the dipole and inductor loop of the UHF RFID antenna DB printed on commercial, calendered, coated and calendered paperboards and PET film was investigated (Fig. 8).

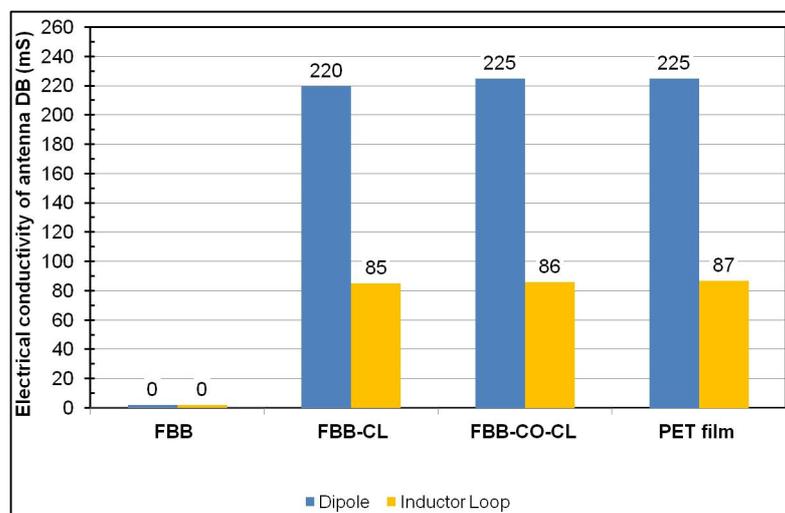


Fig. 8: Effect of plasma treatment (3 min) of commercial (FBB), calendered (FBB-CL), coated and calendered (FBB-CO-CL) paperboard and PET film on the electrical conductivity of the dipole and inductor loop of printed aluminium UHF RFID antenna DB.

The UHF RFID antenna DB printed on plasma-treated commercial paperboard (FBB) was electrically non-conductive. After plasma treatment of the calendered paperboard (FBB-CL), the electrical conductivity of the dipole and inductor loop of the printed UHF RFID antenna DB was the same as that of the antenna printed on the PET film without and with plasma treatment. After plasma treatment of coated and calendered paperboard (FBB-CO-CL), the electrical conductivity of the printed UHF RFID antenna DB was unchanged. It has the same electrical

conductivity of the dipole and inductor loop as the antenna printed on PET film without and with plasma treatment. It follows that after the plasma treatment of calendered paperboard (FBB-CL) the same electrical conductivity of the printed UHF RFID antenna was achieved as on the coated and calendered paperboard (FBB-CO-CL) without plasma treatment.

## CONCLUSIONS

The surface modification of commercial paperboard had an effect on the surface roughness and the initial water contact angle. After calendering, the surface roughness of commercial paperboard decreased by 33.8%, while after coating and calendering it decreased by only 22.1%. The initial water contact angle of commercial paperboard did not change significantly after calendering, while after coating and calendering decreased by 33,3% due to the presence of a wetting agent in the coating colour. After plasma treatment, the surface roughness of commercial paperboard increased by 11.8%, calendered paperboard by 15.6%, coated and calendered paperboard by 8.6%. After plasma treatment, the initial water contact angle of commercial paperboard decreased by 37.0%, calendered paperboard by 29.0%, coated and calendered paperboard by 23.4%.

The UHF RFID aluminum antennas printed on commercial and calendered paperboard were electrically non-conductive due to non-spreading of the thermoplastic tie layer and poor adhesion of the conductive aluminum layer. Antennas printed on plasma-treated commercial paperboard were electrically non-conductive despite improved wettability, because excessive surface roughness (7.6%) caused defects in the aluminum layer only 260 nm thick. Defects in the printed aluminum layer caused interruption of the electrical conductivity and functionality of the printed antenna in terms of its communication between the reader and the tag as well as the energy transfer from the dipole to the tag chip.

The required surface modification of the calendered paperboard for printing electrically conductive antennas was achieved only after plasma treatment, which increased the wettability of the paperboard, spreading of the thermoplastic tie layer and conductive aluminum layer adhesion. The electrical conductivity of the UHF RFID antennas printed on the coated and calendered paperboard was the same as on the PET film, after the plasma treatment the electrical conductivity of the antennas has not changed. The results show that after surface modification by coating and calendering commercial paperboard, the same electrical conductivity of printed UHF RFID antennas was achieved as after surface modification by calendering and plasma treatment of commercial paperboard.

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