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LIGHTFASTNESS OF LITHOGRAPHIC PRIMARY COLOURS COATED WITH NANOCOMPOSITES COMPOSED OF TiO_2 AND WATER-BASED VARNISH

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Abstract: *The UV radiation causes inks to fade. The ink fading will lead to the degradation of the visual appearance of printed imprints making them less attractive to the user. To deal with this that phenomenon, one of the solutions is to create a coating that could challenge these issues. In order to create efficient protective coating against UV induced degradation, nano scaled titanium dioxide (TiO_2) wasl added to the commercial water-based varnish. To determine influence of the amount of TiO_2, was homogenized in various weight ratios. The prepared nanocomposites were applied onto offset (lithography) prints made in accordance with Fogra PSO, i.e. ISO 12647-2:2013 on gloss coated paperboard. The samples have been subjected to artificial UV induced aging for 30 hours. The protective properties of the nanocomposite TiO_2 coating was determined by calculating colorimetric and densitometric change on full tone and determining tone value change of half tones.*

The results showed that the prepared nanocomposite coating has relatively little effect on the printed colour of the samples. However, some coating compositions exceed the allowed tolerance $\Delta E_{ab} > 5$, however in those cases the initial colorimetric value of WB (ΔE_{ab}) was close to the FOGRA PSO border value. The coatings with TiO_2 will increase resistance to accelerated ageing on full and halftone.

To conclude, this research has provided the new perspective on modulation possibilities of commercially available varnishes in order to cope a designated problematics and downsides of coatings which was, in this case, UV induced fading and degradation of visual appearance. The further research should investigate the applicability of this kind of modulated varnish in other coating techniques as well the use of other kind of nano sized compounds.

Key words: Coating, Nanoparticles, Titanium dioxide (TiO_2), Lightfastness, Packaging

1. INTRODUCTION

All industrial areas are generally impacted by two factors, the available technology, and the overall costumer's needs. Printing industry is no exception where some sectors have decreased in volume, leading the overall revenue decrease of the industry in general. Cardboard packaging is no exception where it has a vast and growing share in the packaging industry. Regardless of the grim prognosis, the forecast is that the total growth should reach about 269 billion US by the year 2024 (*Justpaint, n.d*). Furthermore, "green" agenda also pushed the graphic industry even further labelling it as the "forest killer"(Berg & Lingqvist, 2019). Despite these negative influences, the need for printed packaging has a projected and continuously growth worldwide *Statista*, n.d.).

Industries are faced with the new trends, such as eco-friendly and "green" industry development and packing is no exception. Nevertheless, application of these concepts must not degenerate its basic purpose and requirements which are product protection and aesthetics (Makower, n.d.). Paperboard and its derivatives have some weak points and usage of various synthetic (mostly polymer) materials in growing in the general use but those materials do have some disadvantages in terms of sustainability and biodegradation (*Allianz, 2021;* Eu, n.d.).The quality factors of printed packaging can be reduced by an outer influence such as UV induced degradation. The UV radiation (from 100 to 380 nm) not only causes inks to fade (Aydemir & Yenidoğan, 2018), but is also responsible for yellowing and the loss of strength of the material (Jiménez-Reyes *et al.*, 2021) (Brokerhof *et al.*, 2017). The ink fading and yellowing will lead to the loss of the visual appearance of printed imprints making them less attractive to the user. Moreover, the fading of ink can result with losing the products information (i.e., barcodes) that endangering the consumer's rights to information and violate the EU's consumers law (*Consumer rights directive | European Commission*, n.d.).

To diminish that phenomenon, one of the solutions is to create a coating that could challenge these issues. The water-based varnish (denominated as WB) is the most "eco-friendly" coating type on the market that has the industrial, technological, and commercial capability since it does not create hazardous vaporization when handling or curing (*3 Oak News*, n.d.). The WB varnish has some downsides when compared to UV varnish for instance in terms of lightfastness, barrier properties etc (Ragauskas & Lucia, 1998a). Some research has been made with incorporation of nano sized compounds in commercial varnishes that have the upgrade potential (Zvekic *et al.*, 2011; Cigula, Hudika & Vukoje, 2021; Hudika, Cigula & Vukoje, 2021). One of those compounds is a metal oxide, TiO_2, well known for its ability to absorb UV radiation and commonly used as part of inks, colours, coatings, sunscreens, etc (Wang *et al.*, 2009; Hudika *et al.*, 2020).

2. MATERIALS AND METHODS

For this research, lithographic offset printed samples were coated with prepared nanocomposites in various nanoparticle's weight ratios (0.25%, 0.5% and 1%). The base of nanocoating was commercial water-based varnish while chosen nanoparticle was titanium dioxide (TiO_2). The printing for the purpose of this research was done in accordance with the ISO 12647-2:2013 standard, i.e., FOGRA PSO (ISO/TC 130 Graphic technology, 2013). The standards sets the CIE L*a*b* values of each primary colour and defines the tolerance level is ΔE_{ab}=5. The sheetfed offset printing was done with industrial printing press KBA 105 PRO-5+L FAPC (Rapida, n.d.). The printing test form was created which included large patches (50 x 200 mm) with full tone of primary colours and tone value (TV) patches (10 x 10 mm) from 0 – 100% with the step of 2% (0 – 10%) step of 5% (10 – 20% and 80 – 100%) and step of 10%. The four colour CYMK offset printing was done with Novavit F918 Supreme Bio inks made by Flint Group (*Flintgrp, n.d.*). The paper used was UPM Finesse white gloss paper WFC (Woodfree coated) grade with gloss coating and grammage of 300 g/m^2. Paper was conditioned and stored in environment at temperature of (23 ± 1) °C and (50–55) % relative humidity before the printing process. The nanocomposite's base was water-based varnish (denoted as WB) with production name Terra High Gloss Coating G9/285 by Actega USA, used for flexographic printing (coating) technique (*TERRAWET®*, n.d.). The nanoparticles used in this research were TiO_2 was from Sigma Aldrich with the production code EC 2015-282-2 (International organisation for standardisation*,* 2011; Titan dioxide, Sigma A., n.d.). Nanoparticles used in the nanocomposite mixture were weighed using Mettler Toledo XS205DU Dual range Analytic Scale. The varnish was weighted in the mixture cup. Due to the increase in viscosity by adding NPs, in the initial WB a 5% wt of deminiralized water was added. All prepared nanocomposites were applicable in flexography, i.e. viscosity was in range of 0.7 Pa*S. The nanocomposites were prepared by homogenization of nanoparticles into water-based varnish using ultrasound dispenser Hirrlscher UP100H from 20 to 40 minutes depending on the nanoparticles weight ratio (%) in the varnish (Table 1). Time-to-weight ratio of homogenization process derived from previous testing and proved to be the most optimum one with this setup. During the course of homogenization process the mixture was cooled down in cooling console that was immersed into a water bath with the temperature of 5°C.

Table 1: Homogenisation time and denomination

Compound	Denomination	Weight ratio (%)	Homogenization time (min.)
Pure WB	*WB*	-	-
TiO_2	*0.25% TI/NC*	0.25	20
TiO_2	*0.5% TI/NC*	0.5	30
TiO_2	*1% TI/NC*	1	35

To apply the prepared nanocomposite coating, IGT F1 printability tester for flexography was used (IGT, n.d.). The anilox used was IGT 402-258 roller with 90 l/cm screen line and 18 ml/m^2 cell volume. The flexographic polymer printing plate used was Kodak Flexcel NX with DigiCap NX patterning. The printing plate had 100% TV coverage, used for varnishing (Workflowhelp, n.d.).

To investigate the lightfastness of the prepared samples, accelerated aging (AcA) via UV radiation was used. To create those conditions Solarbox 1500 e chamber was used with the xenon light exposure in the solar chamber with the indoor filter simulates the sunlight rays hitting the surface through a glass or shop window. In this research, an indoor filter S208/S408 (artificial daylight) was used. This was done to assess the level of visual degradation of the samples which were meant to be kept in an indoor environment during their lifespan. Irradiation in the chamber was set to 550 W/m^2 and the temperature of 50 °C (30h

of AcA, i.e., to electromagnetic energy of 59 MJ/m^2). The experiment was done in accordance with the ISO 4892-2 standard (International organisation for standardisation, 2013).
The colour control was conducted by measuring full tone patches with the use of Techkon SpectroDens spectrophotometer (Techkon, n.d.) (D50, 2° standard observer, no polarisation filetr, M1 with calibration on absolute white). There are two important factors that needs to be addressed when considering coatings in general, first being, would and if so in what measure coating influence the original print and second, did the coating achieved and fulfilled its purpose. To tackle the first issue two colorimetric approaches were considered, the ΔE_{ab} which showed the difference between the coated samples and FOGRA PSO (Table 2) designated colour target and ΔE_{00} which provided the difference between aged and unaged samples (Sharma, Wu & Dalal, 2005; Mokrzycki & Tatol, 2011).

Table 2: Fogra PSO tagret values

Colour	*L**	*a**	*b**
C	55	-34	-52
M	47	74	-5
Y	87	-4	90
K	16	0	0

To measure ink density Techkon SpectroDens spectrophotometer was also used (D50, density status E, no polarization filter and calibarion on the paper sample). The equation (1) to describe ink density (D) is:

$$D = log_{10} 1/R \quad (1)$$

Where is: *R* – reflectance.
The ink density was measured before and after UV exposure to better assess the ink fading.

3. RESULTS AND DISCUSSION

After homogenization of TI/NC, coating and curing (24h), colorimetric analysis was conducted in three directions; first determining the difference between sample and colour target set by FOGRA PSO (ΔE_{ab}); second, the density difference. The third part is where the overall efficiency of the prepared TI/NC is measured between the initial unaged sample and sample after 30h AcA (ΔE_{00}). The comparison of the measured ΔE_{ab} values can be seen in the Table 3.

Table 3: Comparison of ΔEab

Sample	C	M	Y	K
Uncoated	1.97	3.74	4.28	3.05
WB	1.98	4.54	2.61	3.57
0.25% TI/NC	1.15	*5.47*	3.73	2.52
0.5% TI/NC	1.32	*6.95*	3.10	2.15
1% TI/NC	1.81	*6.88*	1.42	4.60

To observe the magenta closely, CIELAB coordinates are presented in Table 4. With increase of TiO_2 nanoparticles, a* values decrease while the b* coordinate's values increase.
From these results it could be noted that introduction of nanoparticles into the coating requests primary colours closer to the target CIE LAB values. One can note that the closest colour difference for black (initial value is not close to the allowed colour difference) was achieved on the sample with highest TiO_2 weight ratio. The ΔE_{ab} values rise with WB coating and this is most evident on magenta. The introduction of TI/NC nanoparticles will excel the values over the standard, but one must bare in mind that the initial ΔE_{ab} iof uncoated sample was slightly high (ΔE_{ab}=4.54), although it was in accordance with ISO 12647-2:2013.

Table 4: CIELAB for magenta

Sample	*L**	*a**	*b**
Uncoated	47.49± 0.21	75.81± 0.18	-1.73± 0.21
WB	47.70 ± 0.18	77.01 ± 0.22	-1.64 ± 0.28
0.25% TI/NC	47.53 ± 0.13	76.49 ± 0.11	0.04 ± 0.23
0.5% TI/NC	47.72 ± 0.1	75.90 ± 0.08	2.01 ± 0.17
1% TI/NC	48.06 ± 0.08	74.46 ± 0.16	2.09 ± 0.36

As for the yellow samples coated with TI/NC (Table 5) the values in the b* coordinate rises, i.e. first added weight concentration increased the b* coordinate but further increase of the added nanoparticles caused decrease of the b* coordinate. The overall biggest shift is with the 0.25% TI/NC and with the Hybrid/T. Lowering the b* coordinate can be attributed to the TiO_2's whiteness.

Table 5: CIELAB for yellow

Sample	*L**	*a**	*b**
Uncoated	88.22 ± 0.17	-4.08 ± 0.16	94,44 ± 0.13
WB	88.24 ± 0.18	-4.49 ± 0.43	92.33 ± 1.02
0.25% TI/NC	88.20 ± 0.21	-4.15 ± 0.02	95.23 ± 0.5
0.5% TI/NC	88.56 ± 0.1	-4.49 ± 0.36	92.68 ± 0.46
1% TI/NC	88.15 ± 0.72	-4.26 ± 0.11	90.60 ± 0.31

To get a density value assessment, measurements were performed on the wedge patches of different nominal tone values before and after 30h AcA. The first measurement set was performed on 100% TV and second one on half tones. Tone values (TV) were calculated using Murray-Davies equation (4). In Table 8, density changes of the full tone or ΔD between 0h and 30h AcA are presented for TI/NC, this was done using equation 5. It can be noted that yellow samples have the biggest density difference. Although the difference is notable, it lowers with increase of TiO_2 nanocomposite weight ratio (%). As for the cyan and black samples, they proved to be the stable, i.e., almost no change can be noticed. Densitometry was also used to investigate the dot gain (also known as tone value increase, TVI), which is defined as the difference between the actual printed dot and nominal (ideal) digital dot (%), i.e., a dot could indicate 50% TV, but after printing it measures 65%. For the tone value control Murray-Davies equation (*Cmykhistory*, n.d.) (4) was used to calculate tone values (G). To investigate the density change (ΔD), equation 2 was used:

$$\Delta D = D_b - D_{AcA} \quad (2)$$

Where are: ΔD – density difference, D_b – density before AcA, D_{AcA} – density after 30h AcA.

Table 6: ΔD between 0h and 30h AcA for TI/NC

Sample	C	M	Y	K
Uncoated	-0.09	0.14	0.38	-0.05
WB	-0.02	0.09	0.31	-0.03
0.25% TI/NC	0.00	0.11	0.32	0.03
0.5% TI/NC	0.01	0.10	0.29	0.04
1% TI/NC	-0.03	0.10	0.22	0.02

To investigate the half-tone values, the results for yellow are presented…. The yellow has proven to be most sensible to change, one for being the brightest process ink and due to its pigment which has the lowest lightfastness from the process inks (Ragauskas & Lucia, 1998b). It can be noted that with increase of nanoparticle weight ratio (%) overall density lowers. The 1% TI/NC proved to be the most resistant to change in the half-tones.

Table 7: ΔTV of yellow for TI/NC sample

Sample	20%	30%	40%	50%	60%
Uncoated	31.46	33.7	27.84	28.61	26.31
WB	24.69	24.26	26.94	22.50	19.32
0.25% TI/NC	20.16	20.15	16.56	14.57	11.27
0.5% TI/NC	5.22	8.25	11.48	11.77	7.14
1% TI/NC	-0.93	3.29	6.14	5.93	5.31

The colorimetric difference, ΔE_{00} between samples non-AcA samples and 30h AcA samples in comparison to WB can be seen in the Table 6. Most notable difference can be seen on the yellow samples. With increase of nanoparticles in the nanocomposite coating, the effect of UV exposure decreases. The TiO_2 nanoparticles can absorb the part of UV radiation. The gap band of TiO_2, is up to 329 nm. This leads to ink samples coated with TiO_2 to appear darker, as less light is reflected (Vukoje et al., 2022).

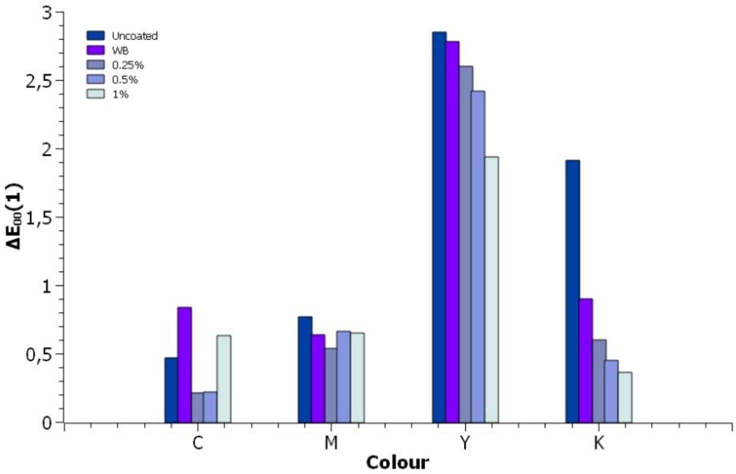

Figure 1: Colour difference (ΔE00) of TI/NC coated samples

The yellow samples had the biggest chromatic change as well as they had on ΔE_{00} (Table 7). This leads to conclusion that with the increase of nanoparticles weight ratio chromatic value difference decreases. Meanwhile, the ΔC for cyan dropped belove 1 for 1% od added TiO_2, meaning that the colour became more intensive. This can be attributed to TiO_2 compound's colour of the natural state. TiO_2 as mentioned before is used also as white pigment but in its natural state it is blue (Krishnan & Shriwastav, 2021).

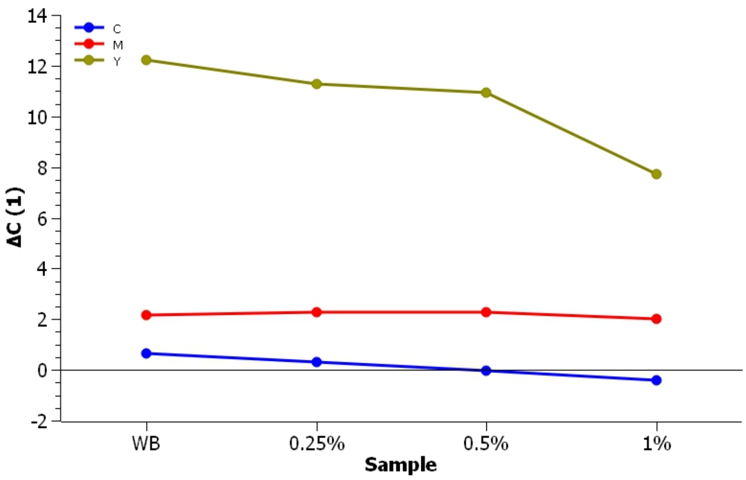

Figure 2: Chroma difference (ΔC) of TI/NC coated samples

4. CONCLUSION

The aim of this research was to compose a nanocomposite coating which will upgrade existing varnish and enable transfer of the visual message and function of the packaging throughout the product's lifespan. To do so, the newly formed nanocomposite compositions was created with commercially available water-based varnish as a base that was enriched with TiO_2 and SiO_2 nanoparticles in various weight ratios. The research provided the results from it can be concluded that the prepared nanocomposites can protect the printed paperboard packaging, with a relatively small effect on the colour. Although on some samples primary colours caused colour difference to the colour targets set by Fogra PSO above ΔE_{ab}=5, it must be considered that the initial values were close to the tolerance range (i.e., magenta was above ΔE_{ab} 4.5). Nevertheless, that means that usage of the proposed nanocomposites leads to better control of the printing and setting the primary colours closer to the colour targets, if printing in standardized mode. The results proved that TI/NC did increase the resistance to AcA induced by the UV radiation, i.e. reduced ink's fading. After tested for 30h AcA, TI/NC overall showed resistance to UV radiation, and it can be noted that increase of the nanoparticle's weight ratio will increase preservation of colour in the AcA process.

The Hybrid/T nanocomposite did upgrade the WB performance in terms of resistance to the AcA, where it showed great resistance to the lightfastness caused by the UV radiation. For instance, when compared to WB, it upgraded the system substantially, where in terms of magenta and yellow ink Hybrid/T performed 15 and 60% better.

This research provided a new method of possibility of varnish modification via added nanoparticle that can upgrade the protective benefits against outer environmental influence such as UV radiation. The newly formed nanocomposite coatings protected the printed image while having limited effect on the colorimetric values of printed inks.

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