COATINGS IN GRAPHIC INDUSTRY

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Abstract: *Contrary to the negative predictions, the graphic arts industry keeps continuing its growth. The modern world is transferring information through digital platforms leading to decrease of the serial publications on paper (newspaper, magazines etc). But on the other hand, packaging industry is increasing its revenue by high numbers. In the same way as for the whole industry, many predicted that paper use will also decrease, as for a long period of time (still partly present) paper was thought of as being killer of trees. Today, along with new findings, paper is becoming more popular with EU Commission banning single use plastics. Paper as a substrate is becoming popular in packaging industry, mainly due to being suitable for both organic and material recycling.*
However, due to lack of some functional properties, materials are often coated. Coating as a process includes covering of a surface by another substance. The coatings processes are present in various industries and are intended to enhance properties of the base materials. In the graphic industry the coating process is often called varnishing, due to the resins used as a coating material. Varnishing is used to improve rub resistance and provide varnished material with special effects (combination of gloss and matte surfaces). Applicability and functionality of the coatings is achieved using various materials, among which are nano-engineered materials. Results of various researchers show benefits of introducing nanocomposites in the packaging industry by improving prints' resistance to degradation by UV irradiation, improving barrier to water vapour and enabling packaging surface to inhibit microbes' growth.
To conclude, coatings development and application plays a significant role in the material development, as it can provide common materials with improved properties, as well as enhanced aesthetics. At the same time, application of coatings could present some obstacles in both materials and organic recycling, and for that reason development of coatings should include evaluation of recyclability of the coated product as well as the characterization of coating's functionality.

Key words: packaging, coatings, varnishing, nanocomposites

1. PACKAGING

The contemporary world is more focused on a quick and widespread information transfer by means of digital media, which led to decrease in the graphic industry focused on the material information transfer (newspaper, magazines etc.). Nevertheless, the printing industry has continuous growth of revenues (LeBlanc, 2019). This growth is mainly related to the packaging sector. Packaging plays different roles, from protecting the product to conveying information to users and facilitating the use of the basic product (Kirwan, 2007). An increased awareness of human impact on the environment is pressuring the use of environmentally friendly materials in terms of their biodegradability and recyclability. In addition to user awareness, the European Union recognized the importance of transforming industries and has set regulations and development guidelines (EU Commission, 2018; *European Partnership under Horizon Europe Processes4Planet*, 2020). These regulations highlighted that although various materials are used for packaging purposes, paper and paper-based materials are highly involved due to good printability, possibility of biodegradation and recyclability (Rastogi & Samyn, 2015). The predictions for paper and paperboard products were very optimistic (*Global Paper and Paperboard Container and Packaging Market to Grow by $ 102.51 Billion During 2020-2024*, n.d.), but due to the obstacles in the global economy (COVID-19 pandemic, war in the Ukraine), these estimations lowered the numbers but are still not in recession (Globenewswire, n.d.; MarketWatch, n.d.).
The packaging is often classified as primary, secondary and tertiary. The primary packaging is in direct contact with the goods and can sometimes have printed outsides (e.g. various chocolates). Secondary packaging in majority of the cases comes in contact with customer, while tertiary packaging is used for storage and transport. Examples of packaging types in the pharmaceutical industry can be seen in Figure 1.

Figure 1: Example of packaging types according to its use (Types of Pharmaceutical Packaging – HVAX | The Pharma-Engineering Blog, n.d.)

Roles and desired properties of the packaging vary depending on its type. Primary packaging is intended to enable protection of the goods from mechanical and, more importantly, chemical threats. Secondary packaging provides mechanical protection and more importantly communicates with the costumer affecting it's purchase desision (Hurley et al., 2013), which is also important for branding (Grundey, 2010). Tertiary packaging is intended to provide a protection during transport and storage. In order to enhance some properties of the packaging materials, the substrates are often coated with other substances.

2. COATINGS

Coating is a widely used term, described by Cambridge dictionary as "a layer of a particular substance that covers a surface" (*COATING | Meaning, Definition in Cambridge English Dictionary*, n.d.). This can be applied to vast variety of industrial applications. The coatings are almost always used to provide or increase some kind of functionality of the basic material, for example; coating of the metal to enables anticorrosive behaviour (Li et al., 2018; Wang et al., 2019; Zhang et al., 2019), applying coating onto wood can increase its resistance to irradiation and humidity or as a flame retardant (Chen et al., 2022; Nair et al., 2018; Teaca et al., 2019), etc., fabrics enhanced by its waterproofness (Bramhecha & Sheikh, 2021; Luftinor et al., 2022; Pongsathit et al., 2019), etc.

In graphic industry coating process is usually related to application of the overprint coating or a primer to the printing substrate. Printing on a paper substrate often presents a challenge due to paper's porosity. The porous surface enables absorption of the small particles into the paper and diminishes their functionality, for example. conductive ink (Agianniotis, n.d.). Therefore, primers are applied to provide a paper substrate surface properties with good printability, i.e. primer is a coating which is applied onto the paper surface planned to be printed on (Havenko et al., 2020; Khadzhynova, 2020; Mendez-Rossal & Wallner, 2019; Morić et al., 2019).

On the other hand, basic roles of overprint coatings are to provide surfaces with some aesthetics due to the high gloss or matte finish and enhanced rub resistance. Although there are many overprint varnishes/coating present on the market, they are usually divided into three groups depending on the composition and/or application method (Hook, 2018). In offset printing, varnish is applied the same way as is the ink. In that case the applying is performed inline and can be spot printed or on the whole surface. For the aqueous or water-based coating the presses often have a special printing unit which is designed for coating. These are often used to cover the whole image. Both varnish and water-based coatings are curing without additional drying equipment. On the other hand, UV coating is cured by UV radiation. The UV coating is often being printed off-line, due to the fact that majority of the printing presses are not equipped with UV drier. Additionally, there is a strong movement towards UV LED technology due to the lower costs, environmental friendliness and shorter starting time (UV lamp need to heat up before curing process can start) (Milmo, 2020). Nevertheless, UV coating technology can provide higher gloss values and better rub resistance. On the other hand, although coating is intended to be transparent and should not change colour of the prints, previous research have confirmed colour changes (Hoffstadt, 2004; Simonot & Elias, 2004).

The research of MEGA research group also provided an insight into the colour change due to application of the commercial coatings (Figure 2) (Cigula, Hudika, & Donevski, 2021).

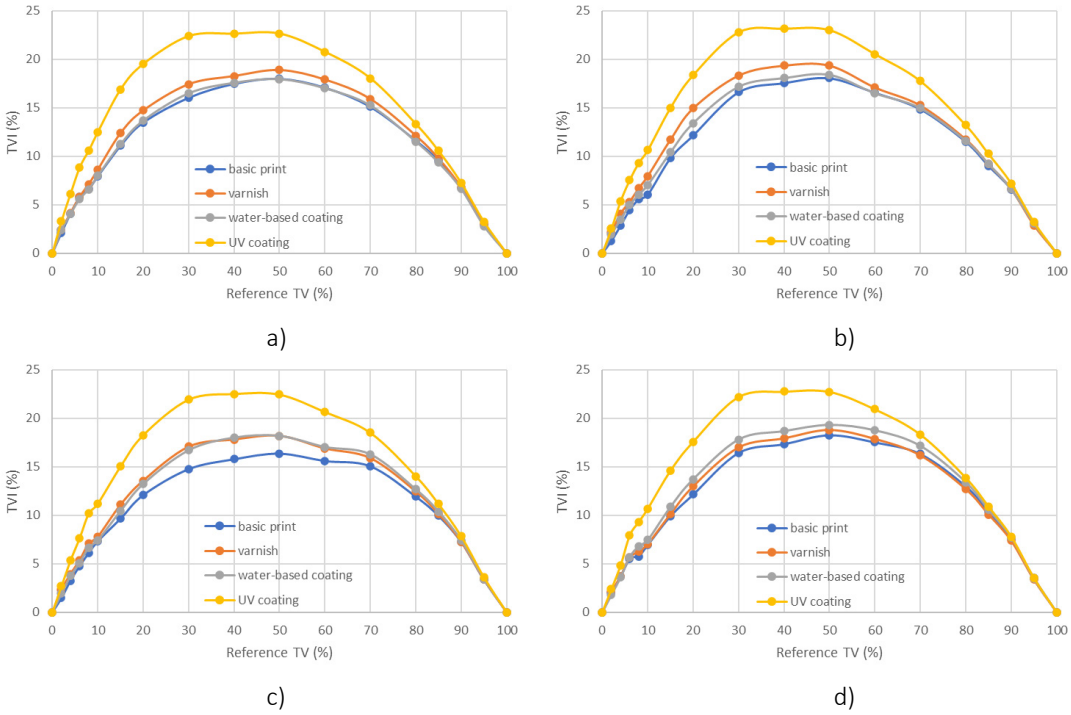


Figure 2: Tone value increase (TVI) of coated prints a) black, b) cyan, c) magenta, d) yellow

As it can be seen Figure 2, the water-based varnish has less influence on the tone values of all applied coating (with the exception of influence on magenta). On the other hand, applying UV varnish will result in significant change of the tone value and darker appearance of the print. Please note that in this research, UV varnish was applied off-line with the screen printing unit, i.e. it had thicker layer than the others.

Beside mentioned, development of nanotechnology have introduced nanomaterials to the coating industry as well. The nanocomposites include some nanosized compound which is incorporated into the polymer matrix and give added functionality to the basic coating. There are numerous papers related to this thematic, for example corrosion protection (Deyab et al., 2021; Kasar et al., 2020; Peng et al., 2020), UV protection (Nuraje et al., 2013; Zeljko et al., 2021), creating superhydrophobic surfaces (Ghashghaee et al., 2019; Ibrahim & Sultan, 2019; Sutar et al., 2020), antimicrobial behaviour (Kumaravel et al., 2021; Nakhaie et al., 2022).

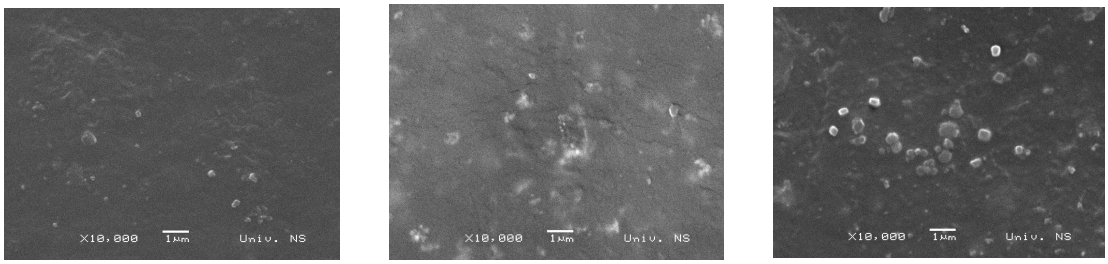
For that reason, the results section will present research covering the topic of nanocomposite coatings in the packaging industry.

3. RESULTS OF NANOCOMPOSITE AS OVERPRINT COATING

In the last few years, we have conducted research and development of new coatings, which would provide some additional features to the packaging material (paperboard, cardboard, labels). The results of those efforts are showed below, with references where the specific experimental details are shown in full.

For these coatings research team included nanoparticles of ZnO, TiO_2 or SiO_2 and mixed them into polycaprolactone (PCL) and polylactic acid (PLA). This proved to be challenging, as both of the biopolymers are soluble in organic solutions (ethyl-acetate and chloroform for PCL and PLA, respectively). For that reason, PLA was excluded from further research as it did not provide better functionality than PCL. All coatings were prepared by first dissolving biopolymer granules in a defined solvent (mixing in a closed glass container using magnetic stirrer). In the second step, defined mass of the nanoparticles was added and homogenized by ultrasonic homogenizer. Coatings were then applied with a laboratory coater in a defined wet layer thickness. After drying, the prepared print samples were analysed to reveal how the applied layer had influenced their properties.

This procedure enabled a satisfactory distribution of the nanoparticles within the nanocomposite, and consequently, on the applied layer on the paperboard prints Figure 3.



a) b) c

Figure 3: SEM images of coatings on printed coated paperboard: a) PCL-0, b) PCL + 0.5 % ZnO, c) PCL + 0.5 % TiO_2

The visible particles on the surface of the samples originated from Ca (Figure 4), which is commonly used as anti-set-off powder (preventing the prints to stick to each other during the printing process).

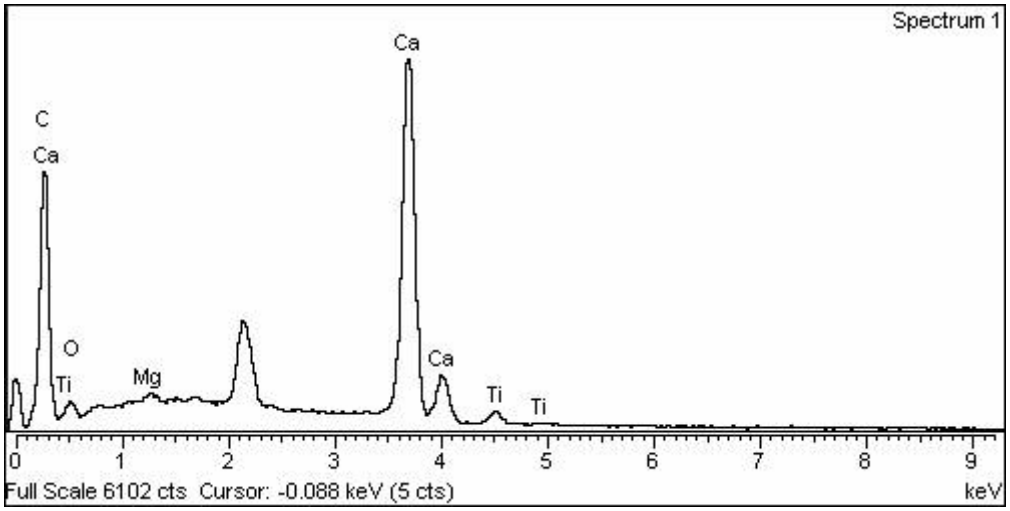


Figure 4: EDS spectra of the PCL-0.5%TiO_2 nanocomposite on coated paperboard print

Nevertheless, coatings have provided basic prints with better lightfastness and also improved barrier to water vapour, but had not influenced colour that well. Research have been undertaken on both coated and uncoated paperboard.

In Table 1 and Table 2 one can see the colour difference ΔE_{00} of nanocomposites including SiO_2, ZnO and TiO_2 (Cigula, Hudika, & Tomasegovic, 2021; Majetić, 2020). It can be noted that colour differences are visually unnoticeable. The results were obtained during previous research (Cigula, Hudika, & Tomasegovic, 2021; Majetić, 2020).

Black ink was influenced the most, which is expected as prints generally darken when coated, as previously shown in Figure 2.

Table 1: ΔE_{00} between uncoated print and samples with applied nanocomposite coating on a coated paperboard

	Cyan			Magenta			Yellow			Black		
	SiO_2	ZnO	TiO_2	SiO_2	ZnO	TiO_2	SiO_2	ZnO	TiO_2	SiO_2	ZnO	TiO_2
PCL	0.19			0.48			0.25			1.43		
0.10%	0.35	0.63	0.95	1.18	1.12	0.54	0.38	1.08	0.40	2.34	1.10	1.39
0.25%		0.58	0.53		1.10	0.58		1.01	0.14		1.02	1.34
0.50%	0.85	0.63	0.38	1.26	1.40	0.50	0.57	1.04	0.63	2.68	0.48	2.58

Table 2: ΔE_{00} between uncoated print and samples with applied nanocomposite coating on an uncoated paperboard

	Cyan			Magenta			Yellow			Black		
	SiO_2	ZnO	TiO_2	SiO_2	ZnO	TiO_2	SiO_2	ZnO	TiO_2	SiO_2	ZnO	TiO_2
PCL	0.19			0.48			0.25			1.43		
0.10%	0.33	1.09	0.43	0.26	0.64	1.02	0.21	0.88	0.24	1.61	0.86	1.87
0.25%		1.06	1.02		0.69	1.36		1.00	0.56		1.48	2.65
0.50%	0.16	0.94	0.60	0.50	0.62	1.58	0.33	0.96	0.50	1.52	0.92	1.99

An application of biocomposite coatings reduced water vapour transfer rate (initial paperboards without coatings were 0.52 g/m^2*day and 1.44 g/m^2*day for coated and uncoated paperboard respectively), although it was evident that addition of nanoparticles gave worse results that the neat PCL coating.

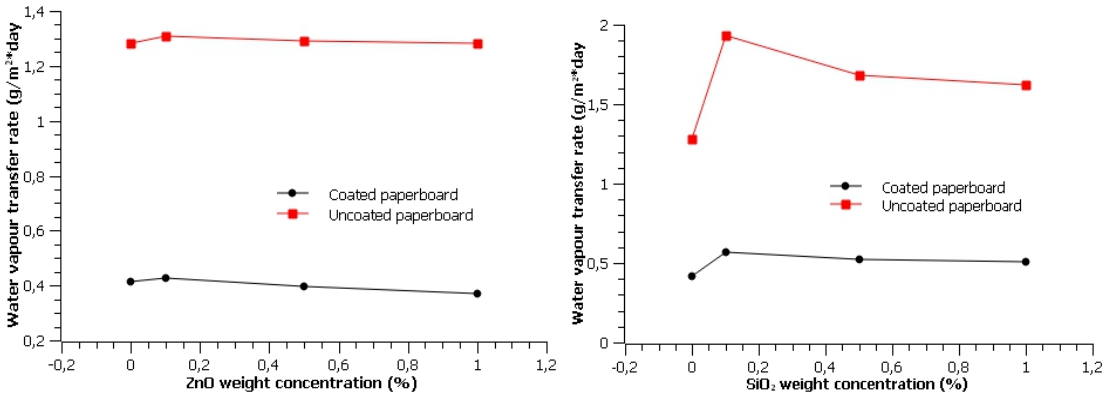


Figure 5: Water vapour transfer rate of SiO_2-PCL (right) and ZnO-PCL (left) nanocomposite

The second property investigated was the resilience of prints to the accelerated ageing by exposing the samples to the UV light source under controlled conditions (Cofomegra Xenon chamber 1500e, using indoor filter). The test was performed on coatings containing ZnO and TiO_2, as they were known UV absorbers (Ghamsari et al., 2016; Tsuzuki & Wang, 2010; Wang et al., 2009). Due to being most sensitive to UV radiation, the results shown include observing the chromatic change of yellow ink only. Further information is available in (Cigula, Hudika, & Tomasegovic, 2021).

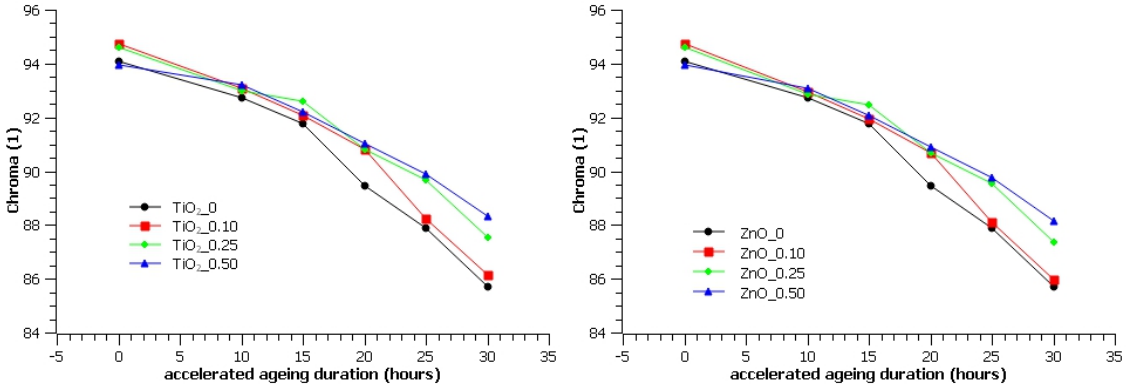


Figure 6: Yellow colour chroma change of prints on coated substrate due to accelerated ageing. Samples coated with PCL-TiO_2 (left) and PCL-ZnO (right)

It can be seen in Figure 4 that both nanoparticles had performed similar in the accelerated ageing tests and diminished chroma change. Additionally, it is visible that in the first 15 hours of accelerated ageing there is almost no change in chroma regardless to the nanocomposite composition i.e. weight ratio of the incorporated nanoparticles in coating

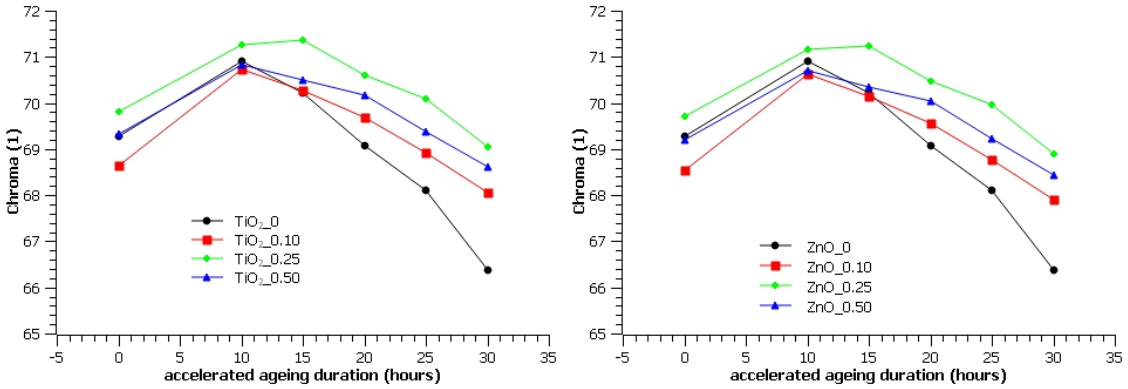


Figure 7: Yellow colour chroma change of prints on uncoated substrate due to accelerated ageing. Samples coated with PCL-TiO_2 (left) and PCL-ZnO (right)

Opposite to the prints on coated substrate, chroma of the samples on uncoated paperboard is significantly lower and the accelerated ageing has lower influence in general (probably due to the partial protection of printing ink by paper fibres). Influence of the nanoparticle's concentration is more visible on these samples. At the beginning of the accelerated ageing process, it is increase of the yellow chroma, most probably due to the yellowing of the paperboard and higher amount of the optical brighteners in the sample paperboard.

4. CONCLUSION

This paper was prepared to give some insight in the coatings in the graphic industry. Furthermore, some results of the research including biopolymer based nanocomposites and their influence on some aspects of the packaging material improvement. These results have proved that procedure for coatings' mixing and homogenization results with satisfactory distribution of nanoparticles. Application of biopolymer based nanocomposites had provided some improvements in the lightfastness of the samples without causing significant colour change.

To conclude, development of coatings and their application is widespread activity over numerous industries. It plays a significant role in the material development as it can provide basic material with enhances properties, but enhanced aesthetics as well. But with all benefits, application of coatings could provide some obstacles in materials and organic recycling due to the antimicrobial potential of some added particles and possibly, their retention in wastewater. For that reason, development of coatings should include determination of recyclability of the coated product as well as the characterization of coating's functionality.

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