Mechanical and barrier properties of flexible packaging materials after the flexo printing process

P Balaban¹, D Viduka², V Ristic³, M Maksin⁴, V Radic⁵, R Vladisavljevic⁶, M Vulić⁷, M Josimovic⁸ and NZ Radivojevic⁹

¹ Department of Mechanical Engineering, Higher Education Technical School of Professional Studies, Novi Sad, Serbia.
² Department of Economics, Faculty of Economics and Engineering Management in Novi Sad, University Business Academy in Novi Sad, Serbia.
³ MIANU-IRASA International Research Academy of Science and Art, University Singidunum, University Metropolitan, Belgrade, Serbia.
⁴ Institute of Architecture and Urban / Spatial Planning of Serbia, University of Belgrade, Serbia.
⁵ Department of Management, College of Business Economics and Entrepreneurship, Belgrade, Serbia.
⁶ Department of Industrial Engineering/Engineering Management, Faculty of Technical Science, University of Novi Sad, Serbia.
⁷ Department of Economics, Applied Management and Entrepreneurship, Academy at Applied Studies, Kragujevac, Serbia.

Submitted: 05 November 2020; Revised: 20 April 2021; Accepted: 21 May 2021

Abstract: In the flexo printing process, flexible food packaging materials are exposed to various effects (solvents, temperature, transport devices, etc.), that impact the quality of food packaging materials. Although, literature is available on the impact of these factors on the quality of food packaging, there are very few papers examining the impact of the flexo printing process on food packaging materials. Therefore, the aim of this paper is to examine if there are any changes occurring in tensile strength, elongation before tearing and gas permeability after the flexographic printing process regarding the selected flexible packaging materials – oriented polypropylene (OPP) and coated paper/polyethylene (PAP/PE). The measurement of tensile strength and elongation before tearing was carried out according to SRPS G.S2.734, and the measurement of gas permeability according to Lussy Method - DIN 53380. The results presented in this paper show that the tensile strength and elongation in both foil samples increased after the printing process (a higher increase of strength is determined in the PAP/PE foil). The exception is the PAP/PE sample, where increased permeability of N₂ was observed. The reason for such results could be due to different surface characteristics of the tested materials since the process of ink absorption is different. Surface tension tests were conducted by applying measurements of the contact angle. The results show that the process of flexo printing did not change the functional properties of the packaging.

Keywords: Elongation before tearing, flexible packaging materials, flexo printing process, gas permeability, surface tension, tensile strength.

INTRODUCTION

For many years, flexible packaging has been the most rapidly growing segment of the packaging industry (Selke & Culter, 2016). The food industry has been the largest end-user of flexible packaging, and in 2020, it held a 70% share of the global flexible packaging market (Flexible Packaging Market, 2021). The global flexible packaging market is estimated at 31.5 million tonnes in 2021, with an annual average growth of 3.3% (Smithers, 2021).

Flexible packaging includes bags, sacks and bags of various shapes, as well as wrappers made of foil, which, when filled and closed, get a flexible shape (Ebnesajjad, 2012).

Of all flexible packaging materials, particularly polymer mono-materials and polymer-based multilayer...
materials), are extensively used in the packaging industry and have a wide application in food packaging (Palić et al., 2019; Żołek-Tryzniewska et al., 2020). The reason for this is having good properties such as low cost, low weight, tolerability with content (especially foodstuff), appropriate barrier, mechanical and optical properties, as well as suitability for processing by printing and packaging machines (Perić-Maretić et al., 2004; Shagufta Ishteyaq et al., 2019).

Among other characteristics of flexible packaging foils, tensile strength, elongation and permeability, are the key performance requirements and have a significant place within the general characteristics of printed packaging products made of polymeric materials (Ebnesajjad, 2013).

Barrier properties of polymers are low permeation by light, oxygen, moisture, CO₂, aroma, or fat (Desnica et al., 2015). These characteristics are very important because the contact between atmospheric gases and food products can lead to biochemical or physical reactions that can affect the food quality (Fotie et al., 2017). Packaging materials have different barrier properties to gases. Packaging materials are characterized by the property of porosity or permeability. Porosity is a characteristic of paper where the diffusion of gases occurs physically by the transition of molecules between the interpores of the material. Permeability is a physico-chemical process where the passage of gas molecules is done by the influence of chemical affinity or solubility. The molecules absorb on the surface of the material, then diffuse in the direction of the concentration gradient through the foil, and then desorb on the other side.

Tensile strength and elongation are important mechanical properties of foil. These parameters show the suitability of the material for manufacturing during the entire technological process (printing, lamination, packing), as well as resistance during transport, handling and storage. Insufficient strength can lead to cracking of the material and seizing production (Izdebska & Thomas, 2015). Also, just as with other thermoplastic materials, the resistance to elongation of OPP film decreases as the temperature increases (Flexographic Technical Association, 1999).

The packaging should be able to withstand the operations of further production and processing, handling, transport and distribution of food in it (Balaban-Djurdjev, 2006; Izdebska et al., 2015, Marangoni et al., 2020). In previous research conducted by Hertlein (1997); Rubino et al. (2001); Mrkić et al. (2007), and Siracusa (2012), it has been found that processes of production, handling, packaging and printing can affect some characteristics of flexible packaging materials, primarily barrier properties. For example, the absorption of vapours or liquids from the environment can cause a decrease in mechanical properties (Siracusa, 2012; Sangroniz et al., 2019). It has been found that the mechanical stress of the folds causes changes in the properties of the barrier, both monofoil and laminate. In general, the largest change in gas permeability with an increased stress cycle was observed in the film having a metalized layer in the structure (Hertlein, 1997; Mrkić et al., 2007; Siracusa, 2012).

The main characteristics of unprinted packaging foils are more or less researched and known (Dunn, 2015), and much data on these characteristics are given by the manufacturer of packaging foils to potential buyers. The material datasheet can be a useful tool for assessing the attributes of material in a stable state (Dunno, 2017), but is usually limited in terms of material performance due to, e.g. heat treatment (Ašonja et al., 2013; Han et al., 2018). These and other phenomena can have a negative impact on the performance and shelf life of products packaged in this material (Dunno, 2017).

Flexographic printing is one of the most widely used procedures for printing flexible packaging today (Bolanča et al., 2015). Flexo printing is a technique of direct rotary printing that uses printing plates made of rubber or photopolymer material. Printing plates are mounted on the printing cylinder and inked by the anilox that transfers fast-drying printing ink. It is possible to print on almost all substrates (absorbent and non-absorbent) (Izdebska & Thomas, 2015).

In flexographic printing, changes can occur primarily in the printing machine during the printing process, due to the direct action of machine elements, primarily printing and transport devices and solvents on the foil, or the indirect effect of energy through the medium (in the case of devices for drying printed foils) or the energy field (e.g., in the corona process) (Balaban-Durdev, 2006; Gosh, 2015).

In addition to examining the change of the mechanical and barrier characteristics, the influence of ink acceptance on the printed material was also investigated. One of the factors that significantly affect the quality of the flexo print is wetting of the ink on the substrate, as well as wetting of the ink on ink. Factors that affect the wetting are surface tension of the ink and surface energy of the
substrate. These two factors must remain in a certain ratio in order to avoid poor ink acceptance on the substrate or the ink on the ink (Izdebska & Thomas, 2015; Aydemir et al., 2020). In this paper, the calculation of the surface tension of the tested materials is performed through contact angle measurements.

In practice, as well as in the literature, the question of whether to what extent and why the stated characteristics change in the printing process is often neglected (or only assessments of a qualitative nature are given).

In order to give a solution to the issue, the aim of this study was to examine whether the mechanical and barrier characteristics of flexible packaging foils remain the same after the flexographic printing process and thus determine whether there are quantitative changes that may affect their characteristics. The samples were taken from the industrial flexo printing process.

Packaging materials that were selected for testing are in the production program of one domestic printing facility: monomaterial-oriented polypropylene (OPP) and multilayer foil-coated paper/polyethylene (PAP/PE).

**METHODOLOGY**

**Flexo printing machine**

In general, flexo printing machines consist of four aggregates, which can be with various variations. These are unwinding and rewinding devices, printing and drying devices and devices for transporting and guiding foils. Particularly, important for this work are the phases with a direct interaction between the work process and the printing material (foil) (Balaban-Đurđev, 2006). These are primarily devices for printing, transport and drying of printed foils.

The most important part of the printing machine is the device for dyeing or colour transfer. This colour transfer is done with an anilox roll that transfers a precise amount of ink onto the printing plate. The volume of the ink transferred to the printing plate is determined by the number and size of engraved cells on the surface of the anilox roll. The doctor blades remove the excess ink.

In the process of guiding the foil strip through the printing machine, complex physical operations occur, the consequence of which is the appearance of stress and elongation of the foil. Changes in these characteristics of the foil during printing process is mostly due to the action of temperature and tensile forces, that affect the print quality or the appearance of deviations in the colour register, the length of repetition and the creation of folds in the printing foil.

The drying of printed foils has a significant impact on the quality of printing. At elevated drying temperatures (approx. 100–115 °C), changes in the properties of the foils are possible (excessive elongation of the foil can occur and thus cause the web tension to be incorrect). The possible influence on the tested characteristics of the films is due to intercooling.

Flexo printing predominantly uses solvents of the evaporation range below 100 °C. The drying time of the ink depends on the evaporation temperature of the solvent, on the speed of the foil, the heat input and the turbulence of the air. The time in which the printed ink must dry without problems occurring in the next printing device (e.g., problems with ink trapping) is measured in fractions of a second. On such a short distance between individual printing devices, approx. 80–85% of the solvent from the ink must be removed, and the remaining part is eliminated during the final drying in the channel. The materials referred to in this paper are printed on a ‘flexotechnic’ machine with six dyeing units and a maximum print speed of 140 m/min. The positioning of the inking unit was mechanical.

| Table 1: Printing machine settings during printing |
|---------------------------------|-------------------------------|------------------|---------------------------------|-------------------|-------------------|
| Printing machine               | Speed (m/min)                 | Anilox rolls (lines/cm) | Ink transfer (cm<sup>3</sup>/m<sup>2</sup>) | Anilox cell angle (for all colours in CMYK) | Drying effect | Printing pressure |
| Flexotechnic                    | 100–140                       | 200                           | 5                                 | 45°                      | intermediate drying 30%, final drying 90% | Manually adjusted |
depend on the type of the printed image. Screen ruling in this experiment was 200 L/cm, which is mostly used for printing halftone images (Izdebska & Thomas, 2015). The ink capacity of the anilox roller in the experiment was 5 cm³/m². Solvent printing flexo inks based on nitrocellulose, with a viscosity of 15–20 seconds, were used.

**Printing materials**

In the production of polymer films, macromolecules in a melt are arranged randomly and in a ‘relaxed’ state in all three directions, homogeneously, such as in cast or blown films. The distribution of macromolecules in a melt is always isotropic. If the film stretches at an elevated temperature in a certain direction, the macromolecules will be under tension. If such a film cools to room temperature, the stresses and positions of the macromolecules freeze, and the macromolecules are fixed in their new dimension. The dimensions of such a film are constant even if heated for a long time at an elevated temperature (Gosh, 2015).

Oriented films are obtained by stretching previously produced films in one (longitudinal) direction or in both (longitudinal and transverse) directions. This process improves the quality characteristics of the produced films. Compared to non-oriented, oriented films, which increase the tensile strength and elongation at break in the direction of stretching, more transparent and higher smoothness films are obtained, and to some extent, the barrier characteristics for gases and water vapour are improved. In biaxially oriented films, the macromolecules are stretched in the longitudinal and transverse directions (the ratios of stretching are adjustable).

In this paper, polyethylene (PE) does not appear as a monofoil but as a component of a multilayer PAP/PE foil, made by extrusion process with a slightly coated paper surface. Coating and roughness tolerances are not controlled. For the examination of the characteristics of the foils in this study, samples of unprinted foils were used followed by the examination of the printed foils.

The properties of tested OPP and PAP/PE foil were:

- Oriented polypropylene film (OPP) with a thickness of 20 µm, and unit weight of 18.20 g/m²;
- Multilayer material with a coated paper as a layer and polyethylene as an inner layer (PAP/PE) which is in contact with the product, with a thickness of 72 µm and unit weight of 69.25 g/m² (paper 47.85 g; polyethylene 21.40 g).

**Surface tension**

In the case of non-polar materials that include the tested polypropylene, the surface of the foil to be printed is pretreated in order to increase the polarity of the surface. This surface treatment of the foil (mainly by the corona process through electrical discharge) achieves the effect of surface oxidation, and the existing non-polar structure is converted into polar groups (Aydemir et al., 2021). This significantly improves the print quality, adhesion and chemical affinity, which are a condition for the optimal process of printing, laminating and coating (Aydemir et al., 2021).

The foils tested in the study come to the printing facility after pre-processed by the corona process and therefore, not additionally processed (by the corona process) before printing.

In flexo printing houses, the usual surface tension measurement is performed using a liquid with known surface tension. The surface tension of the foil then corresponds to the surface tension of the test liquid. With this method, it is only possible to determine the interval in which the surface tension of the foil is located, which is usually 2 mN/m. Using this test, as well as the test with a special pen with a surface tension of 38 mN/m (normally used in operating conditions), it is only possible to approximately determine the surface tension.

The surface tension of solids can only be measured indirectly. The most common method is based on the measurement of the contact angle. The determination of the surface tension of solids is based on the fact that the shape of a drop of a liquid on a solid body depends on the surface tension of the materials in contact. The surface tension calculation is based on the Young-Dupre equation, where \( \sigma_s \) = surface free energy, \( \sigma_{dl} \) = interfacial tension between the liquid and solid, \( \sigma_l \) = surface tension of the liquid (Krüss Application Report, 2014):

\[
\sigma_s = \sigma_{dl} + \sigma_l \cos \theta
\]  

\( \ldots(1) \)

The contact angle \( \theta \) and the surface tension of the liquid \( \sigma_l \) are measurable. The quantities \( \sigma_s \) and \( \sigma_{dl} \) cannot be determined experimentally. However, the surface of a solid body can be indirectly characterised by measuring or calculating the size of the contact angle. The problem is solved with the help of Fowkes’ theoretical setting, which defines the surface tension of a solid (\( \sigma_s \)) and a liquid (\( \sigma_l \)) as the sum of the dispersive (non-polar) - \( \sigma_d \) and polar part - \( \sigma_p \):

\[
\sigma_s = \sigma^p_l + \sigma^d_l
\]  

\( \ldots(2) \)
\( \sigma_s = \sigma_s^p + \sigma_s^d \) \hspace{1cm} \text{(3)}

The Owens, Wendt, Rabel and Kaelble (OWRK) is a method that uses the values of contact angle of two test liquids with a known polar and dispersive part. It is a standard method for calculating the surface free energy of a solid from the contact angle with several liquids (Aydemir et al., 2019). In this way, the surface free energy is divided into polar and non-polar parts.

By combining or arranging the equations of Young, Owens, Wendt and Rabel, the equation is as follows:

\[
1 + \cos \theta = \frac{\sigma_i}{\sqrt{\sigma_i^d}} = \sqrt{\sigma_i^p} \cdot \sqrt{\sigma_i^d} + \sqrt{\sigma_i^a} \cdot \sqrt{\sigma_i^d} \hspace{1cm} \text{(4)}
\]

This expression represents the equation of the straight line:

\[ y = ax + b \hspace{1cm} \text{(5)} \]

Where,

\[
x = \frac{\sqrt{\sigma_i^p} - \sigma_i^d}{\sigma_i^d} \hspace{1cm} y = \frac{1 + \cos \theta}{2} \frac{\sqrt{\sigma_i^p}}{\sqrt{\sigma_i^d}}
\]

\[
a = \sqrt{\sigma_i^p} \hspace{1cm} b = \sqrt{\sigma_i^d} \hspace{1cm} \text{(6)}
\]

If the values of the total surface tension \( \sigma_s \) and of the polar and dispersion part \( \sigma_s^p \) and \( \sigma_s^d \) are known for different liquids with which the test is performed, as well as the corresponding contact angles of the droplet. Then, using the above expressions, the line can be determined (by linear regression), on which the values can be read directly.

The coefficient of line \( a \) and section \( b \) is calculated according to the following equations:

\[
a = \frac{\sum x_i \cdot y_i - \sum x_i \cdot \sum y_i}{n} \hspace{1cm} \text{(7)}
\]

\[
b = \frac{\sum x_i \cdot (\sum y_i)}{n} - \frac{\sum x_i \cdot (\sum y_i)}{n} \hspace{1cm} \text{(8)}
\]

\( x_i, y_i \) - the coordinates of the points calculated for the individual test fluids according to the above equations.

According to the above equations (Owens & Wendt, 1969),

\[
\sigma_s^p = a^2 \text{ (polar part)} \hspace{1cm} \text{(9)}
\]

\[
\sigma_s^d = a^2 \text{ (polar part)} \hspace{1cm} \text{(10)}
\]

\[
\sigma_s = \sigma_s^p + \sigma_s^d \text{ (total surface tension)} \hspace{1cm} \text{(11)}
\]

The calculation of the surface tension of the foils in this paper was performed using the sessile drop method based on the values of the measured contact angles using the DSA 25 device from Krüss with the integrated image processing (Software Zur Tropfenkonturanalyse, 2012). The principle of measuring the contact angle is based on the fact that a drop of the test liquid is dosed on the sample. The drop is recorded with a camera, and the video is analysed. Based on the mean values of five measured contact angles and known surface tension of the test liquids, the total surface tension of the films has been calculated.

**Tensile strength and elongation**

Tensile strength and elongation before tearing were determined by the method of SRPS G.S2.734. Both materials were tested on an Instron machine (Instron Universal Testing Instrument, Model No 4301). Samples of packaging materials were cut into dimensions 200 mm \( \times \) 15 mm. The initial distance of the clamps was 100 mm, and the crosshead speed was 400 mm/min. The tensile strength is calculated according to the formula:

\[
\sigma_s = \frac{F_x}{A} \text{ [N/mm}^2] \hspace{1cm} \text{(12)}
\]

\( F_x \) - value of the breaking force [N]

\( A = b \cdot d \) - the smallest initial cross-sectional area of the foil, mm\(^2\)

\( b \) - 15mm, foil width

\( d \) - foil thickness, mm

The relative elongation at break (\( \varepsilon \)) is the ratio of the absolute elongation of the foil sample \( (l_f - l_i) \) and its length before testing \( (l_i) \):

\[
\varepsilon = \frac{l_f - l_i}{l_i} \cdot 100 \% \hspace{1cm} \text{(13)}
\]

\( l_f \) - the length of the foil sample at the moment of tearing.
The values of tensile strength and elongation are expressed as the mean value of five individual measurements in both the longitudinal and transverse directions. Standard deviation and coefficient of variation are also given. In the case of combined foils, such as the tested foil PAP50 / PE20, the strength of their individual components is different. Hence the increase in strength of this film after printing is expressed with a tearing force of N/15 mm (15 mm-foil width).

**Gas permeability**

Measurement of gas permeability was performed by the Lussy method according to DIN 53380, on a Lyssy GPM-200 apparatus, with the corresponding Gasokuro Kogyo GC-320 gas chromatography and Hewlett-Packard 3396 A integrator (Lyssy, 1984).

Gas permeability was determined isostatically in a chamber divided by the sample of the tested packaging into two units. In a one chamber, there was pure helium, and in the other, a mixture of gases: oxygen, carbon dioxide, nitrogen (1:1:1). The pressure on both sides of the film was identical (0.2 bar). During the analysis, it gradually came to the saturation of helium, depending on the permeability of the packaging material. The concentration of measured gases in the helium chamber was detected in a chromatogram with an integrator. The retention time is given with the area under the peak.

In the case of combined foils, such as the tested foil PAP50 / PE20, the strength of their individual components is different. Hence the increase in strength of this film after printing is expressed with a tearing force of 1 bar.

**RESULTS AND DISCUSSION**

**Surface tension**

In order to calculate the surface free energy (SFE) of a solid, using the OWRK method, it is necessary to have at least two liquids with known disperse and polar parts, wherein at least one of the liquids must have a polar part greater than 0 (this choice is made in order to better determine the polar and dispersed fractions), (Kruess Application Report, 2014). The list of the most common measuring liquids can be found in (Kopczynska & Ehrenstein, 2017).

The surface tension components of the fluids used for calculation are found in Table 2.

| Liquid      | \( \sigma_l \) (mN/m) | \( \sigma_d \) (mN/m) | \( \sigma_g \) (mN/m) |
|-------------|------------------------|------------------------|------------------------|
| Water       | 73.0                   | 26.0                   | 46.8                   |
| Ethylene glycol | 47.7                   | 26.4                   | 21.3                   |
| 1,4 – dioxane | 33.0                   | 33.0                   | 0.0                    |

Table 3 shows the measured contact angles and the calculated values of the total (\( \sigma_l \)) and polar (\( \sigma_d \)) parts of the tested materials. The surface tension of solvent-based inks (that are used for printing the tested samples) ranges from 20 to 30 mN (Aydemir et al., 2021). According to some studies, the surface energy of polymeric materials should be 10 mN/m higher than the surface tension of the ink, although in some studies, this is not decisive (Aydemir et al., 2021). Untreated polymeric materials have non-porous surface, hydrophobic characteristics and show a low value of surface energy and low polarity. In order to achieve adequate ink reception, the required surface energy of plastic materials on a printing press should be greater than 42 mN /m (Morsy et al., 2006; Brandt, 2015). As it can be seen from the results in the table, the calculated surface tension is slightly lower than the recommended. The reason for this may be that during the storage surface, free energy decreases (Izdebska & Thomas, 2015). Therefore, corona treatment is recommended just before printing.

| Specimen    | PAP/PE \( \Theta [\text{'}] \) | OPP \( \Theta [\text{'}] \) |
|-------------|---------------------------------|-----------------------------|
| Contact angle | \( \Theta [\text{'}] \)       | \( \Theta [\text{'}] \)     |
| Water       | 80.5                            | 77.2                        |
| Ethylene glycol | 54.4                            | 46.9                        |
| 1,4 - dioxane | 28.37                           | 30.5                        |
| \( \sigma_l \) | 33.85                           | 35.6                        |
| \( \sigma_d \) | 4.8                             | 6.58                        |

Using this method, the surface tension of the polar and non-polar parts of the test materials can be determined. The greater the similarity between liquid and solid material in terms of its polar and dispersed proportions, the better the wetting must be. In general, for good wetting, the surface free energy and its polar interactive component must be increased.
The results presented show a slightly higher polar component of the OPP sample (Kopczynska & Ehrenstein, 2017). However, in order to determine whether the cause of sorptive behaviour is energy (contact angle) or geometry (capillarity/porosity) in the examined paper sample, it would be necessary to do Washburn measurements (Zilles, 2000). It can also be seen from the results that the coated paper showed a higher value for contact angle with water, indicating that it has a hydrophobic nature. It also has a lower polar fraction value.

**Tensile strength and elongation**

Measurements have shown that the tensile strength and elongation before tearing of the OPP and PAP/PE foil increase after the printing process (Tables 4 and 5). When it comes to the OPP foil, the results show that this increase is 3.48% longitudinally, 5.84% transversely, and for the PAP/PE foil, the increase is 6.80% longitudinally and 8.55% transversely.

The results show a higher increase in tensile strength after printing on the PAP/PE foils compared to the OPP foils. The reason could be different surface characteristics of the tested materials and, therefore, different ink transfer. In general, ink transfer takes place in three steps: contact and wetting between the substrate and the ink, immobilisation of part of the ink layer and ink splitting (Thorman, 2018). The interaction between the ink and polymeric materials is a very complex process that depends on the physical and chemical properties of both the ink and the substrate. When applying solvent-based ink, a solid film of ink is created by evaporating the solvent at room or slightly elevated temperatures.

The combination of coating, paper/polyethylene material has a cellulose-based surface. In general, the penetration of liquids into porous materials is regulated by two main properties of the system: geometry (capillarity/porosity) and energy (contact angle) (Zilles, 2000). It is generally accepted that liquid absorption into porous networks can depend on wetting, liquid properties and the pore structure. Namely, cellulose fibres in contact with water and cause structural change (Aydemir, 2019). The reason for the increase of the values of tensile strength and elongation could be a stronger penetration of the binder from the ink and fixation in the capillary structure of the paper part of the foil during drying, due to PAP/PE porous structure. However, due to the fact that porosity is not measured in this study additional tests should be done.

Whether the above explanation also applies to other flexible packaging materials could not be reliably assessed by this investigation. However, this may be the reason for the stated, although slightly smaller, increase in the tensile strength of the tested OPP foils.

The results of the elongation before tearing off of the foils show that the PAP/PE foils have a negligible stretching potential, longitudinally 2.65% before printing, and 2.95% after printing and transversely 9.63% before printing and 6.24% after printing. The tensile strength is higher in the longitudinal direction and the elongation in the transverse direction, which is a consequence of the orientation of the cellulose fibres in the paper.

For the OPP films, the relative elongation was 33.40% longitudinally before printing and 27.94% after printing, and transversely 121.09% before printing and 127.05% after printing. The test results obtained for of oriented PP films are were in accordance with the theoretical explanation of the relationship between their mechanical characteristics in the longitudinal and transverse directions. The film had a higher tensile strength in the longitudinal direction and less elongation in the transverse direction, and the same behaviour was shown after the printing process. The elongation property is especially important in polymeric films that show properties of viscoelastic behaviour (Izdebska & Thomas, 2015) because under unchanged load, the material is constantly elongated. It does not return to its original length but remains elongated. If the printed film stretches permanently after passing through the printing device due to excessive stress, e.g. in the winding device and/or due to overheating in the drying device, then, in the end, the repetition length may be longer than the format length, which can affect the print quality.

However, according to some literature sources (Schröder, 2012), more attention should be paid to the starting point of the tensile test in the measuring range of the so-called secant modulus at an elongation of 1–2%. The linear-elastic, as well as linear-viscoelastic area are especially important because it is in these areas with mild deformations (e.g., 1–2%) that the foil is loaded. For printing and packaging machines, the deformation by stretching the foil is less than 1% (Schröder, 2012).

The first assumption that the printing process, solvent, and drying temperature could cause a decrease in mechanical properties of the tested packaging material was not confirmed by these tests.
Table 4: Tensile strength and elongation of unprinted and printed OPP foils

| No | Unprinted | Printed | Unprinted | Printed | Unprinted | Printed | Unprinted | Printed |
|----|-----------|---------|-----------|---------|-----------|---------|-----------|---------|
|    | Fs [N]    | ε [%]   | Fs [N]    | ε [%]   | Fs [N]    | ε [%]   | Fs [N]    | ε [%]   |
| 1  | 74.71     | 32.28   | 31.54     | 36.86   | 43.6      | 126.53  | 129.12    |
| 2  | 80.92     | 33.28   | 38.70     | 38.04   | 43.9      | 127.56  | 130.16    |
| 3  | 83.37     | 32.54   | 47.41     | 42.80   | 40.9      | 119.91  | 105.17    |
| 4  | 81.48     | 35.34   | 37.42     | 41.22   | 38.6      | 106.78  | 110.08    |
| 5  | 84.03     | 33.58   | 29.70     | 39.80   | 43.3      | 124.67  | 150.70    |
| s  | 3.70      | 2.16    | 6.973     | 2.376   | 2.268     | 8.52    | 18.165    |
| Kν [%] | 4.57    | 6.01    | 18.87     | 5.98    | 5.39      | 7.00    | 14.52     |
| $\bar{\sigma}_y$ [N/mm²] | 273.67 | 279.10 | 132.47 | 140.47 |

Table 5: Tensile strength and elongation of unprinted and printed PAP/PE foils

| No | Unprinted | Printed | Unprinted | Printed | Unprinted | Printed | Unprinted | Printed |
|----|-----------|---------|-----------|---------|-----------|---------|-----------|---------|
|    | Fs [N]    | ε [%]   | Fs [N]    | ε [%]   | Fs [N]    | ε [%]   | Fs [N]    | ε [%]   |
| 1  | 51.83     | 2.54    | 2.66      | 24.33   | 26.56     | 9.13    | 6.92      |
| 2  | 54.90     | 2.91    | 2.71      | 25.02   | 26.34     | 9.63    | 5.81      |
| 3  | 53.25     | 2.57    | 3.00      | 24.36   | 26.07     | 9.55    | 6.24      |
| 4  | 50.74     | 2.67    | 2.92      | 24.75   | 28.41     | 9.45    | 5.93      |
| 5  | 52.26     | 2.59    | 3.47      | 25.45   | 27.12     | 10.38   | 6.32      |
| $\bar{x}$ | 52.60   | 2.65    | 2.95      | 24.78   | 26.90     | 9.63    | 6.24      |
| s  | 0.57      | 0.15    | 0.11      | 0.447   | 0.99      | 0.46    | 0.43      |
| Kν [%] | 3.00    | 4.15    | 5.66      | 1.80    | 3.65      | 4.79    | 6.92      |

Gas permeability

The results of gas barrier characteristics of the unprinted and printed OPP and the PAP/PE foils are presented in Figures 1 and 2, respectively.

By comparing the mean measured values of the permeability of the unprinted and printed samples of OPP, a decrease in the permeability of the foils after printing was noticed. In the case of CO₂, this reduction is 9.54%, for O₂ 18.02%, for N₂ 15.03% and 16.66% for air. In the case of tested PAP/PE foils, the permeability after printing is even higher in some gases: CO₂ - permeability reduced by 47.38 %, O₂ - permeability reduced by 20.64% and air - permeability reduced by 5.93%. The measurements showed an increase in permeability only for N₂ by 5.43%.

The results obtained can be compared only with available data by Rubino et al. (2001) and Yahya and Khalifa (2016) (Table 6).

Research done by Rubino et al. (2001) concluded that permeation could be influenced by the type, but not simply by the presence of printing ink on the film, and that barrier properties imparted by the film coatings were more important in oxygen transmission than the presence or type of the ink.
The explanation for a higher permeability reduction in the tested printed PAP/PE foils compared to the printed OPP foils could be similar to the increased tensile strength after printing. The material made from paper and polyethylene, due to its structure, is more susceptible to colour penetration during the drying process of the printed foil. In this process, one part of the binder from the ink penetrates into the surface structure of the foil and solidifies in it, which fills the micropores. Another part forms a layer of ink tied to the surface of the foil. Both cases could contribute to the reduction of permeability in relation to the permeability of the foil before printing, but the first case is primary for a larger decrease in permeability in the PAP/PE foil.

A research done by Bohlin (2013) on the porous structure of pigment coatings on paperboard revealed that a more porous coating structure increases ink penetration. However, to verify this statement in this paper, measurement of porosity of the tested materials should be done.

In another research done by Aydemir et al. (2019), an explanation for the increase of the tested characteristics after the printing process can also be found. In this work, the surface energy on matte and glossy coated papers were examined after printing with offset printing ink. It was found that on both papers, the surface energy decreased, although more on the glossy coated papers. The reason for that was that paper fibres absorb the liquid phase of the ink (because of the capillarity action), become saturated with liquids and thus become hydrophobic, which increases the contact angle. That is, wettability decreases.

It should be emphasised that these tests are not to determine the absolute values of permeability of the tested foils and their comparison but to compare the change in permeability after the printing process.
CONCLUSION

In this paper, it has been analysed whether changes in the mechanical and barrier characteristics of the two flexible packaging materials (OPP and PAP/PE) occur after the flexographic printing process in industrial conditions. In order to get a clear idea of the interaction between the tested substrates and the ink, the surface energies of the substrates were also obtained by measuring the contact angle via the OWRK method.

In both the tested flexible materials (OPP and PAP/PE), there was an increase in tensile strength and elongation of the foils after printing. The results show a higher increase in strength after printing on PAP/PE foils compared to OPP foils. As the coated paper is more porous than the polymer foil, the reason for this could be a stronger penetration of the binder from the ink and fixation in the capillary structure of the paper part during drying. However, for further clarifications of the sorptive behaviour on porous materials, it would be necessary to make additional measurements (e.g., the capillarity and porosity).

The permeability of the foils in both foil samples decreased after printing. The reason for the higher reduction of permeability in the tests printed PAP/PE foils compared to the printed OPP foils, especially for CO2 and O2 gases, could be the same as for the increase in tensile strength (different surface structure of the material and consequently different ways of ink transfer during drying).

The research has shown that the printing process has an impact on changing certain characteristics of the film, but without a significant effect on the functional properties of packaging. Since the foils were tested as used in production and with tolerances given by the manufacturer, the reliability of the results after printing should be respected within these tolerances.

For more reliable results and assessment of the impact of flexo printing on the functional properties of flexible packaging materials, additional research should be done with other flexible materials and inks as well as with a defined amount of transferred inks.

REFERENCES

Aydemir C., Karademir, A., Imamoglu S., Nazli Altay B., Fleming P. & Tutak D. (2019). Investigation of the evolution of hydrophobicity and wettability of paper in multi-color printing process. Cellulose Chemistry and Technology 53(7-8): 787-794. DOI:https://doi.org/10.35812/CelluloseChemTechnol.2019.53.77

Ašonja A., Adamović Ž. & Jevtić N. (2013). Analysis of reliability of cardan shafts based on condition diagnostics of bearing assembly in cardan joints. Journal Metalurgia International 18(4): 216-221.

Balaban-Durdev P. (2006). Influence of flexo printing process on characteristics of polymer packaging foils. Masters Thesis, Faculty of Technical Sciences, University of Novi Sad, Serbia.

Bohlin E. (2013). Surface and porous structure of pigment coatings-Interactions with flexographic ink and effects on print quality. Dissertation, Karlstad University Studies, Sweden.

Bolanča S., Majnarić, I. & Golubović K. (2015) Packaging printing today. Acta Graphica 26(4): 27–33.

Brandt N. (2015). Geeignete Testmethoden zur Charakterisierung von Flexodrucksubstraten für eine verbesserte Druckfarbenhaftung und Bedruckbarkeit. PTS-Forschungsbericht IGF 69 EBR.

Desnica E., Ašonja A., Mikić D. & Stojanović B. (2015). Reliability of model of bearing assembly on an agricultural of Cardan shaft. Journal of the Balkan Tribological Association 21(1): 38-48.

Dunn T. (2015). Manufacturing Flexible Packaging. Elsevier, Netherlands.

Dunno K. (2017). Effects of transportation hazards on high barrier flexible packaging films. Journal of Applied Packaging Research 9(1): 1–7.

Ebnesajjad S. (2013). Plastic Films in Food Packaging, pp. 53. Elsevier, Netherlands.

DOI: https://doi.org/10.1016/C2012-0-00246-3

Flexible Packaging Market - Global Outlook and Forecast 2021–2026 (2021). Available at https://www.arizton.com/market-reports/flexible-packaging-market-industry-report, accessed 1 April 2021.

Flexographic Technical Association (1999). Flexography, Principles & Practices, volume 1, 5th edition, pp. 19. Foundation of Flexographic Technical Association, USA.

Fotie G., Rampazzo R., Ortenzi M.A., Checcia S., Fessas D. & Piergiovanni L. (2017). The effect of moisture on cellulose nanocrystals intended as a high gas barrier coating on flexible packaging materials. Polymers 9: 415. DOI:https://doi.org/10.3390/polym9090415

Gosh A. (2015). Technology of Polymer Packaging, pp. 47. Carl Hanser Verlag, Munich, Germany.

DOI: https://doi.org/10.3139/9781569905777

Han J.-W., Ruiz-Garcia L. & Qian J.P. (2018). Food packaging: a comprehensive review and future trends. Comprehensive Reviews in Food Science and Food Safety 17(4): 860–877. DOI: https://doi.org/10.1111/1541-4337.12343

Hertlein J. (1997). Untersuchung über Veränderung der Barriereeigenschaften metallisierter Kunststofffolien
beim maschinellen Verarbeiten. *Dissertation*. Technical University of Munich, Germany. (In German)

Izdebska J. & Thomas S. (2015). *Printing on Polymers, Fundamentals and Applications*. Elsevier, Netherlands.

Kopczyńska A. & Ehrenstein G W. (2017). Oberflächen spannung von Kunststoffen. Messmethoden am LKT. Friedrich-Alexander-Universität Erlangen-Nürnberg. Available at [https://www.lkt.fau.de/files/2017/06/Oberflaechenspannung.pdf](https://www.lkt.fau.de/files/2017/06/Oberflaechenspannung.pdf), accessed 5 April 2021.

Krüss Application Report (2014). Warum Testtinten nicht die ganze Wahrheit über die freie Oberflächenenergie sagen. Available at [https://www.kruss-scientific.com/de-DE/know-how/applicationsberichte/ar272-warum-testtinten-nicht-die-ganze-wahrheit-ueber-die-freie-oberflaechenergie-sagen](https://www.kruss-scientific.com/de-DE/know-how/applicationsberichte/ar272-warum-testtinten-nicht-die-ganze-wahrheit-ueber-die-freie-oberflaechenergie-sagen).

LYSSY A. (1984). *Analytical Gas Permeability Tester GPM-200, Operation Manual*, pp. 1–24.

Izdebska J., Tryznowska Z. & Swietonski A. (2015). Correlation between plastic film properties and flexographic prints quality. *Journal of Graphic Engineering and Design* 6(2): 19–25.

Marangoni Junior L., De Oliveira L.M., Balardin H.D.F., Cristianini M., Padula M. & Anjos C.A.R. (2020). Influence of high-pressure processing on morphological, thermal and mechanical properties of retort and metallized flexible packaging. *Journal of Food Engineering* 273: 109812. DOI: [https://doi.org/10.1016/j.jfoodeng.2019.109812](https://doi.org/10.1016/j.jfoodeng.2019.109812).

Morsy F.A., Elsayad S.Y., Bakry A. & Eid M.A. (2006). Surface properties and printability of polypropylene film treated by an air dielectric barrier discharge plasma. *Surface Coatings International Part B: Coatings Transactions* 89: 49–55. DOI: [https://doi.org/10.1016/BF02699614](https://doi.org/10.1016/BF02699614).

Mrkić S., Galić K. & Ivanović M. (2007). Effect of temperature and mechanical stress on barrier properties of polymeric films used for food packaging. *Journal of Plastic Film and Sheet ing* 23(3): 239–256. DOI: [https://doi.org/10.1177/8756087907086102](https://doi.org/10.1177/8756087907086102).

Owens D.K. & Wendi R.C. (1969). Estimation of the surface free energy of polymers. *Journal of Applied Polymer Science* 13(18): 1741–1747.

Palić N., Slavković V., Jovanović Ž., Živić F. & Grujović N. (2019). Mechanical behaviour of small load bearing structures fabricated by 3D printing. *Applied Engineering Letters* 4(3): 88–92. DOI: [https://doi.org/10.18485/iaeletters.2019.4.3.2](https://doi.org/10.18485/iaeletters.2019.4.3.2).

Perić-Maretić K., Bates I. & Modrić D. (2014). Comparison of colorimetric values of prints made with cyan ink on different polymer materials. *Procedia Engineering* 69: 1556–1561. DOI: [https://doi.org/10.1016/j.proeng.2014.03.155](https://doi.org/10.1016/j.proeng.2014.03.155).

Rubino M., Tung A.M., Yada S. & Britt I.J. (2001). Permeation of oxygen, water vapor, and limonene through printed and unprinted biaxially oriented polypropylene films. *Journal of Agricultural and Food Chemistry* 49(6): 3041–3045. DOI: [https://doi.org/10.1021/jf001427s](https://doi.org/10.1021/jf001427s).

Sangroniz A., Zhu J., Tang X., Etteberria A., Chen E.Y.X. & Sardon H. (2019). Packaging materials with desired mechanical and barrier properties and full chemical recyclability. *Nature Communications* 10: 3559. DOI: [https://doi.org/10.1038/s41467-019-11525-x](https://doi.org/10.1038/s41467-019-11525-x).

Schroder K. (2012). Spezifikationen für folienverpackungen teil 4: dynamische folienprüfungen, *Imno-Letter*. Available at [https://expertecke1.rssing.com/chan-14308869/all_p1.html](https://expertecke1.rssing.com/chan-14308869/all_p1.html).

Shagualstheyaq A., Asianelam, Omm-e-hany & Syed J.M. (2019). Physical properties and biodegradable study of metallized and non-metallized polypropylene (pp) films: a comparative research. *Advances in Biotechnology & Microbiology* 12(3): 555838. DOI: [https://doi.org/10.19080/AIBM.2019.12.555838](https://doi.org/10.19080/AIBM.2019.12.555838).

Selke E.M.S. & Culter J. (2016). *Plastics Packaging*, 3rd edition, Hanser Publishers, Munich, Germany.

Siracusa V. (2012). Food packaging permeability behaviour: a report. *International Journal of Polymer Science* 2012: 30202. DOI: [https://doi.org/10.1155/2012/302029](https://doi.org/10.1155/2012/302029).

Smithers (2021). Flexible packaging market to grow at 3.4% across 2021–2026. Available at [https://www.smithers.com/resources/2021/feb/flexible-packaging-market-grow-3-4-to-2026](https://www.smithers.com/resources/2021/feb/flexible-packaging-market-grow-3-4-to-2026), accessed 2 April 2021.

Software zur Tropfenkonturanalyse (2012). Benutzerhandbuch Teil 1 u. 2, Krüss GmbH, Hamburg, Germany. (In German)

Thorman S. (2018). Where did the ink go? The effect of liquid absorption on ink distribution in flexography. PhD thesis, Karlstad University Studies, Sweden. DOI: [https://doi.org/10.31803/ihg-20190226231838](https://doi.org/10.31803/ihg-20190226231838).

Yahya I. & Khalifa M. (2016). Effect of the printing remedies and lamination techniques on barrier properties ‘WVTR and OTR value’ for polypropylene film. *EC Nutrition* 5(2): 1089–1099.

Zilles J.U. (2000). Wettabilities and surface tensions of different paper types. *KRÜSS Application/Technical Notes 2001*: 1–6.

Żolek-Tryznowska Z., Rombel M., Petriaszwili G., Dedijer S. & Kašiković N. (2020). Influence of Some Flexographic Printing Process Conditions on the Optical Density and Tonal Value Increase of Overprinted Plastic Films. *Coatings* 10: 816. DOI: [https://doi.org/10.3390/coatings10090816](https://doi.org/10.3390/coatings10090816).