# FLAME RETARDANT AND ANTIMICROBIAL PAPER COATINGS WITH ROSEMARY OIL AND BARIUM BORATE

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Adding an antimicrobial effect to the papers used in the cardboard packaging industry can extend the life of the packed product. Paper, due to its structure, has the property of being ignited quickly. Enhancing the properties of paper, the combination of antimicrobial protection, low flammability and good printability to the paper will expand its usage area in the packaging industry. In this study, barium borate was synthesized in order to impart low flammability to paper. Different combinations of rosemary oil and barium borate were mixed with binding starch in varying amounts, and formulations were prepared. With these mixtures, 80 g/m$^2$ paper was coated using a laboratory-type paper coating machine. The antimicrobial properties of the paper were investigated by performing the disk diffusion antimicrobial test against *E. coli* and *S. aureus* bacteria. The Limiting Oxygen Index (LOI) test was performed to determine the low flammability of the paper. Color, gloss, contact angle and surface energy tests were performed for the printability properties of the paper. As a result, the coatings containing formulations of barium borate and rosemary oil, applied to the paper surface, succeeded in imparting antimicrobial and low flammability properties to the papers, while also improving the printability features.

***Keywords***: printability, paper coating, antimicrobial coating, flame retardant, active packaging

## INTRODUCTION

Paper has an important place in smart packaging applications. In the printing industry, paper continues to attract attention for its printability properties. Paper constitutes the most important input among recyclable basic raw materials for the printing and packaging industry.[1-3] Increasing the technical properties of paper, without affecting its printability properties, paves the way for expanding its functionality.[4-7] Due to the different printing techniques used in the printing industry, there are also differences in the printing techniques applied on paper. Due to these differences, studies are being carried out to ensure that the high standards achieved in paper production are not affected, and to improve the printability and productivity of paper.[8] Sizing, calendering and coating can be counted among the methods used to improve the printability properties of paper. From these processes, a coating/filling material can be dispersed in the resin and applied to the surface, and as a result of the application, the surface properties of paper can be improved, while also antimicrobial, barrier, contact angle and non-flammability properties can be gained.[9-17] The acquisition of such properties is provided by fillers, resins and additives. Studies on this subject[18-20] have paved the way for the use of smart packaging, especially in the food sector, while research efforts to improve the qualities of paper are still continuing.[21]

Active packaging is a smart packaging application. It is the duty of active packaging to extend the shelf life of the products in the package. Smart packaging gives information about the quality of the product in it, the date of manufacture, the classification of the product and the quality of the product.[22] In a globalizing world, the development of necessary product protection and reliability procedures for manufacturers to market their products on distant markets has accelerated the work on the production and development of smart packaging. The packaging is expected to protect the products, especially the food inside it from the influence of the outer environment, including from microbial attack, which accelerates the degradation of foods after the production process. Smart packaging varies as a function of the nature of the product and its exposure to external influences throughout the production-consumption chain. This determines the nature of the application to be made in the packaging of the product.[23]

Antimicrobial packaging is an application that protects food from environmental effects and extends its shelf life. Antimicrobial agents can be classified into natural, animal-based, plant-based and microbial-based agents.[24] Natural antimicrobial agents have been among the most frequently applied agent types in recent years, due to the absence of negative effects on human health. Plant-based antimicrobial essential oils are generally preferred in the healthcare, food and paper industries. The use of natural products in active packaging is preferred by both the producer and the consumer, as they can decompose in the human digestive system and do not have negative consequences on human health. The highly effective antimicrobial properties of plants, such as clove, garlic, lavender and cinnamon, from which essential oils are obtained, have been determined.[25] Also, antimicrobial active food packaging has been achieved by using Ag nanoparticles in paper coating.[26] Another study stated that the encapsulation of rosemary essential oil is suitable for smart packaging production. Thus, chicken, red meat *etc*. have been packed in smart packaging, and it has been shown that the lifespan of the packed food is extended.[27] Rosemary oil is known for its antimicrobial properties. In the past, it was used to preserve food when refrigeration was not possible.[28] Due to its antipathogenic properties, it is also used in the production of disinfectants.

Another feature that adds value to paper and can expand its usage area is its non-flammability.[29] Since some areas where paper is used (such as laminate flooring, wall covering papers) are at high risk of ignition, non-flammability is a feature that helps protect people and save them from fire. Papers used in such applications are required to ignite late, to allow people living in the respective household time to escape during a possible fire. Apart from this, valuable documents (such as money, checks) also need such features. In this study, it is aimed to provide non-flammability properties to paper by applying coatings. When examining the available literature on imparting non-flammability to paper, it was determined that the effectiveness of the flame retardants added to the coating formulation increased with their homogeneous distribution in the coating. For this reason, acrylate functional flame retardant is added to UV-curable formulations to ensure homogeneous distribution.[30] UV cured paper coatings containing silica nanoparticles and phosphine oxide were made by Ozcan *et al*.[31] Melamine and zinc borate hybrid coated papers were produced by Kandırmaz and the suitability of the flame retardant and printability properties of these papers was stated.[32] In addition, Ural stated that tin oxide has a flame retardant effect on paper surface coatings and improves printability properties.[33]

Many different materials can be used as flame retardants. The disadvantage of halogenated and metal-containing retarders is that they release toxic gas as their ratio in the mixture increases. Because of this, they adversely affect human health.[34] In order to eliminate such risks, in recent years, non-halogen flame retardants have attracted the attention of scientists. Compared to halogenated flame retardants, boron, a natural mineral, has non-toxic non-flammability properties.[35] It is known that the boron mineral is widely used in the production of fireproof clothing. Given that its most important feature is its solubility in water,[36,37] today, this mineral has come to be used in many different sectors, from detergents to space technologies, from healthcare to textiles.[38,39] The fact that the burning material lowers the burning rate, thus reducing the loss of life and property in fires, has triggered the use of boron mineral in plastics, paper, wood products, textiles, pigments *etc*.[40] Boron is preferred in disinfectant production, healthcare, medicine, cleaning and textile sectors, due to its antibacterial properties, in addition to its non-flammability and flame retardant capacity.[41]

In this study, it was aimed to produce paper with good printability, low flammability and antimicrobial properties, by applying coating formulations based on cationic starch, which is a commonly used paper filler, and barium borate and rosemary oil mixtures.

## EXPERIMENTAL

### Materials

Uncoated white 80 g/m$^2$ office paper was used in the study. The basic properties of the paper are given in Table 1. Cationic starch was obtained from BASF. The basic properties of cationic starch are given in Table 2. Rosemary oil (98%) was purchased from a local market. Calcium chloride ($CaCl_2$) and borax ($Na_2B_4O_7.10H_2O$) were obtained from Eti Maden Institute.

Table 1
Technical properties of paper used in the study

| Properties | Standard | Uncoated white paper |
|---|---|---|
| Grammage (g/m$^2$) | ISO 536 | 80 |
| Whiteness (D65/10 (%) | ASTM E313 | 98 |
| Gloss (75°) | ISO 8254-1 Part 1 | 5.8 |
| Yellowness | ASTM E313 | 0.07 |
| Thickness (μm) | TAPPI T411 | 188 |
| Color (L*a*b*) | | 92.72, -3.03, -9.32 |

Table 2
Properties of cationic starch

| Starch (g) | Standard |
|---|---|
| Color | White |
| Fineness | 98% |
| pH | 6.0–8.0 |
| Max. Humidity % | 20 |
| Shape | Powder |
| Impurity | 0.10% |
| Protein | ≤0.35% |

### Methods

***Synthesis of barium borate***

Five g of polyvinylpyrrolidone, which is used as a capping agent to prevent agglomeration, was dissolved in 500 mL of double distilled water. After mixing, 1.5 mol of barium chloride and 1.5 mol of borax dissolved in 250 mL of distilled water were added dropwise at 50 °C. The mixture was stirred for two hours. To separate the precipitates from the solution, the mixture was filtered at the end of the reaction time and dried in a vacuum oven at 50 °C overnight.[42]

***Preparation and application of paper coating formulations***

In the study, 80 g uncoated office paper was used as the control group. The aforementioned base paper was coated with cationic starch (F0). In the coating process, 7.5% cationic starch-water solution was prepared and heated up to 105 °C, and mixed for a while at 500 rpm with a mechanical mixer. The resulting hot mixture was cooled to 60 °C and applied to the paper surface with a laboratory paper coating device, using a Mayer stick 2.[14]

Then, three different formulation sets were prepared and applied as paper coatings, which is the other stage of this study. In the first formulation set (F1, F2, F3, F4), 1.0, 2.5, 5.0 and 10 percent of rosemary oil (w/w) was added to the cationic starch formulation. In the second formulation set (F5, F6, F7, F8), barium borate was applied in the same proportions and a different formulation was formed. In the third formulation set (F9, F10, F11, F12), barium borate and rosemary oil were added in equal proportions (1%, 2.5%, 5% and 10%) to the cationic starch formulation. The formed formulations were mixed with a homogenizer for about 10 minutes at 25000 rpm in a beaker. Paper coating formulations were applied to the paper surface under laboratory conditions, using a Mayer rod, in a laboratory paper coating machine. The samples were left to dry for 24 hours at room temperature.

A series of tests were carried out on the surface of the dried samples. The flammability properties of the samples were tested with the LOI. The LOI values of the coatings were measured using a Fire Test Technology (FTT) type device.

***Printability properties of papers coated with barium borate and rosemary oil***

The color properties of the uncoated papers and those coated with barium borate and/or rosemary oil were determined using the CIE L*a*b* color system, according to ISO 12647-5 standards.[43] An X-Rite eXact spectrophotometer was used to measure the CIE L*a*b* values. The spectrophotometer was used in the spectral range of 400-700 nm, using a D50 light source, 2 observers, open polarization filters and 0°/45° geometry.[44]

Table 3
Paper coating formulations

| Formulation sets | Formulation number | Starch (%) | Rosemary oil (%) | Barium borate (%) |
|---|---|---|---|---|
| Control group | Base paper | 0 | 0 | 0 |
| Starch coated | F0 | 100 | 0 | 0 |
| First formulation set | F1 | 99 | 1 | 0 |
| | F2 | 97.5 | 2.5 | 0 |
| | F3 | 95 | 5 | 0 |
| | F4 | 90 | 10 | 0 |
| Second formulation set | F5 | 99 | 0 | 1 |
| | F6 | 97.5 | 0 | 2.5 |
| | F7 | 95 | 0 | 5 |
| | F8 | 90 | 0 | 10 |
| Third formulation set | F9 | 98 | 1 | 1 |
| | F10 | 95 | 2.5 | 2.5 |
| | F11 | 90 | 5 | 5 |
| | F12 | 80 | 10 | 10 |

The obtained coated papers were printed with magenta screen printing ink, using an ARUS semiautomatic printability test device (printing parameters: weaving density of 77 tpc, 75 degree scraping angle and 75 degree shore hardness). The CIE L*a*b* color values of printed papers were measured with an X-Rite eXact spectrophotometer in accordance with ISO 12647-2:2013. The color differences were calculated with the Delta color differences formula given below, according to the CIE ΔE 2000 ISO 13655 standard:

$$\Delta E_{00} = \sqrt{\left(\frac{\Delta L'}{k_L S_L}\right)^2 + \left(\frac{\Delta C'}{k_C S_C}\right)^2 + \left(\frac{\Delta H'}{k_H S_H}\right)^2 + R_T \frac{\Delta C'}{k_C S_C}\frac{\Delta H'}{k_H S_H}}$$

Gloss measurements were made with a BYK-Gardner GmbH gloss meter, with ISO 8254-1:2009 75º geometry. The gloss measurements of the prints were carried out according to ISO 2813:2014, using a BYK Gardner GmbH Micro-Trigloss meter, with 60° geometry. Total surface energy measurements of unprinted papers were calculated over the contact angle measured with the PocketGoniometer PGX+, according to ASTM D5946-17 standard.

***Antibacterial properties of paper coated with barium borate and rosemary oil***

The disk diffusion antimicrobial test is used to determine whether active antimicrobial agents inhibit the growth of certain microbes. Barium borate and rosemary oil used in this study are known antimicrobial agents. The disc diffusion antimicrobial test was used to determine which of the coatings had higher antimicrobial properties.

The activity of paper coated with barium borate and rosemary oil against *E. coli* (gram negative) and *S. aureus* (gram positive) bacteria was determined using the zone of inhibition (disk diffusion) method. For this purpose, two bacterial cultures were activated by inoculation in Tryptic Soy Broth (TSB) at thirty-seven degrees Celsius for one day. Inoculum (0.1 mL) was spread on Mueller-Hinton (MH) agar Petri plates, and 4 mm wide samples cut from coated papers were placed on the plates. These Petri dishes were then incubated for one day and bacterial growth was evaluated. The areas of inhibition around the paper samples in the Petri dish were evaluated qualitatively and quantitatively. Inhibition zone diameters were measured. The void areas around the paper samples were evaluated as an indicator of inhibition of bacterial growth.

## RESULTS AND DISCUSSION

The coating formulations were prepared successfully according to Table 3. The color and gloss measurement results of the coated papers and the control are given in Table 4.

With reference to the base paper, the a* axis was redshifted when only cationic starch was applied to the surface. The samples onto which coatings containing rosemary oil were applied were slightly blue-shifted, which is consistent with the literature.[27] Only for the mixture of cationic starch and barium borate, the a* axis is shifted towards green. In formulations containing only rosemary oil, the b* axis gradually shifted towards blue, as the proportion of rosemary oil increased. It was determined that, as the mixing ratio of barium borate and rosemary oil increased in the F5-F12 range, the color of the paper shifted towards green. This can be explained by the chromophore groups of rosemary oil. For the F1-F4 formulations containing only rosemary oil, the L value gradually decreased as the ratio of rosemary in the mixture was increased. Delta E is below 3, and it is very difficult to detect with the naked eye.[45] It is also in the appropriate range in the standard. When the gloss values are examined, it is seen that the gloss has increased at least twice. This increase is valid for coated papers. As the roughness of the surface of the coated papers decreases, the amount of reflected light increases, thus increasing the gloss.

The most important of the printability parameters is the relationship between the paper and the liquid. Ink is a fluid liquid material. Analyzing the data from this point of view (Figs. 1 and 2), it appears that the starch applied slightly increased the number of H bonds and decreased the contact angle. Adding rosemary oil to the coating increased the contact angle, as expected. The addition of barium borate significantly reduced the contact angle of the coating. The contact angle values corresponding to the coatings containing both barium borate and oil are between those for coatings containing only rosemary oil and those containing only barium borate. Thus, the problem of non-wetting caused by the oil has been eliminated with the addition of barium borate and the printability of the coated paper has been improved. As the surface energy increased, the contact angle decreased. These findings are consistent with the literature.[46]

Table 4
Color and gloss values of coatings

| Formulation sets | Formulation | L* | a* | b* | $\Delta E_{00}$ | Gloss (75°) |
|---|---|---|---|---|---|---|
| Control group | Base paper | 95.46 | 2.91 | -10.21 | Standard | 5.2 |
| Starch coated | F0 | 93.49 | 3.02 | -10.27 | 0.813 | 9.5 |
| First formulation set | F1 | 93.11 | 2.62 | -10.16 | 0.963 | 11.5 |
| | F2 | 92.82 | 2.72 | -10.45 | 0.964 | 9.5 |
| | F3 | 92.56 | 2.95 | -11.16 | 1.324 | 10.1 |
| | F4 | 91.44 | 3.26 | -12.2 | 2.046 | 9 |
| Second formulation set | F5 | 93.39 | 2.13 | -9.85 | 1.371 | 14.6 |
| | F6 | 93.49 | 2.12 | -10.03 | 1.516 | 15 |
| | F7 | 93.32 | 2.46 | -10.27 | 1.199 | 11.2 |
| | F8 | 93.21 | 2.11 | -9.88 | 1.422 | 9.8 |
| Third formulation set | F9 | 93.47 | 2.07 | -10.36 | 1.706 | 19.1 |
| | F10 | 93.13 | 2.12 | -10.23 | 1.538 | 18.2 |
| | F11 | 92.97 | 2.29 | -10.15 | 1.292 | 10.8 |
| | F12 | 93.48 | 2.1 | -10.48 | 1.744 | 19 |

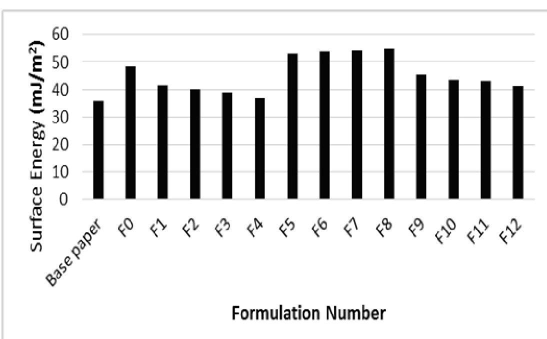


Figure 1: Surface energy values of coated and uncoated papers

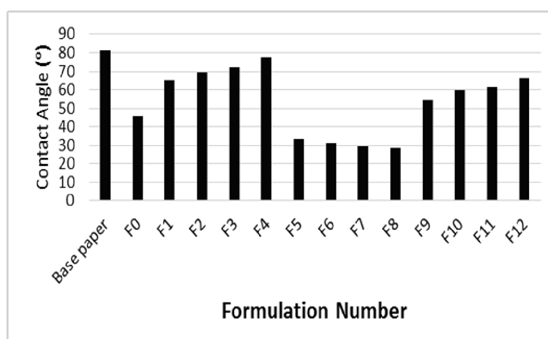


Figure 2: Contact angle values of coated and uncoated papers

The measured LOI values of coated papers are given in Figure 3. It is seen that the cationic starch coating (F0) applied to the surface of the base paper undertakes a protection task, making ignition more difficult compared to the base paper. When the results were examined, it was determined that all the coatings ignited later than the base paper, but the best flame retardancy feature was provided by the presence of barium borate. As the amount of barium borate in the formulation gradually increased, the coating became a significant flame retarding barrier, improving the flame retardant property of the paper. When examining samples F5-F8, it is seen that F8, which has the highest content of barium borate in the mixture, has a LOI values of 25.4%. The dehydration reaction of barium borate acts as a strong protective layer against oxygen penetration, leading to high flame retardancy. Since rosemary oil left a light oily layer on the surface, it had little effect on ignition. The LOI value of the F1-F4 set increased from 21.2 to 21.75, only due to the increase in the amount of rosemary oil. A similar increasing trend is observed for the increase in the rosemary oil content in the presence of barium borate. The reason why rosemary oil adds a light flame retardancy feature is that it creates an oily layer on the surface and reduces oxygen transmission. When Figure 3 is examined, it can be noted that rosemary oil had a little effect lowering ignition in the F1-F4 formulations, in which rosemary oil was used, compared to only starch-coated paper (F0). This suggests that the oily surface increases the burning resistance of the paper coated with cationic starch. In addition, when the formulations (F9-F12), in which rosemary oil and barium borate are used together, are examined, it is seen that as the mixing ratios increase, the low ignition rates also increase the LOI value a little, compared to the samples coated with barium borate alone. The data obtained are in line with the literature.[47]

The antibacterial test was applied to the coated papers to see whether the coatings provide protection against gram negative and gram positive bacteria (*E. coli* and *S. aureus*). The main compounds responsible for the antimicrobial activity in rosemary oil are α-pinene, bornyl acetate, camphor and 1,8-cineole. These limit bacterial growth. Boron compounds have antimicrobial properties since they can pass through the cell membrane.

The obtained results are given in Figure 4. When examining the figure, it was determined that both rosemary oil and barium borate were effective against both bacterial species. It was determined that the coatings containing only rosemary oil were more effective against *E. coli*, and coatings with only barium borate were effective against *S. aureus*. When samples coated with barium borate or rosemary oil were compared, barium borate produced a greater inhibition diameter than rosemary oil did for both bacterial species. It was determined that coatings containing both active substances were more effective than all other coatings against both bacterial groups. The results are compatible with the literature.[48,49]

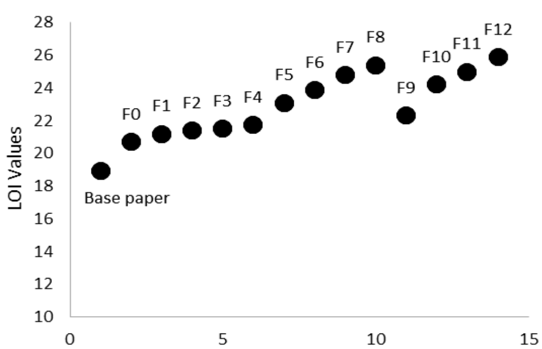


Figure 3: LOI values of coated and uncoated papers

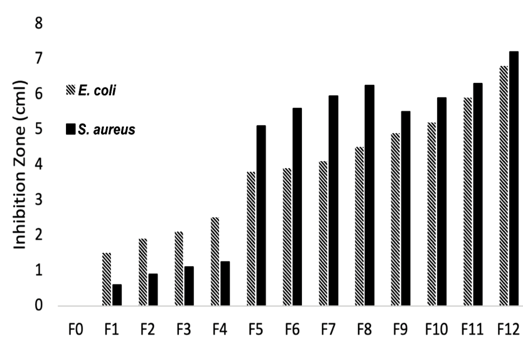


Figure 4: Antibacterial properties of coated papers

Screen printing test prints were made on the prepared papers. The color and gloss values of the printed papers are shown in Table 5. In the formulations containing rosemary oil, the color of the print has shifted to a light number, because of the oil's own color. The data obtained are compatible with the literature.[27] In formulations containing barium borate, the a* value shifted slightly to red and the b* value shifted slightly to blue. There was no significant color difference in a* value for the mixtures containing both rosemary and barium borate. Barium borate slightly increased the L* value of the print. In general, the printing color values do not show significant differences. When the gloss values in Table 5 were examined, parallel results were obtained in the coatings. Compared to the base paper, the gloss of the prints made on all coated papers is higher. This can be explained by the fact that the gaps between the cellulose fibers are filled by the polymeric carrier layer, increasing the gloss.[44]

Table 5
Color and gloss properties of printed samples

| Formulation sets | Formulation | L* | a* | b* | $\Delta E_{00}$ | Gloss (60°) |
|---|---|---|---|---|---|---|
| Control group | Base paper | 49.2 | 71.1 | 41.7 | Standard | 8.1 |
| Starch coated | F0 | 49.77 | 71.09 | 42.6 | 0.78 | 13.7 |
| First formulation set | F1 | 51.19 | 73.48 | 37.28 | 2.75 | 14.8 |
| | F2 | 50.98 | 72.74 | 37.44 | 2 | 8.4 |
| | F3 | 51.68 | 74.42 | 29.18 | 7 | 12 |
| | F4 | 52.44 | 75.51 | 30.24 | 5 | 11.1 |
| Second formulation set | F5 | 51.64 | 74.68 | 37.81 | 5.5 | 13.5 |
| | F6 | 52.95 | 76.65 | 23.8 | 9.4 | 14.1 |
| | F7 | 52.37 | 75.47 | 24.98 | 7.8 | 10.2 |
| | F8 | 51.58 | 74.49 | 30.31 | 4.98 | 13.6 |
| Third formulation set | F9 | 52.4 | 75.33 | 30.18 | 5.9 | 13.3 |
| | F10 | 51.31 | 74.2 | 36.73 | 3.2 | 17.7 |
| | F11 | 53.32 | 76.13 | 25.81 | 8.9 | 10.1 |
| | F12 | 52.88 | 76.45 | 24.45 | 8.21 | 14.3 |

## CONCLUSION

In the present study, barium borate was successfully synthesized and paper coatings containing barium borate and rosemary oil were developed and applied on paper. The findings have shown that the color difference among the coated papers is too small to be distinguished by the naked eye, and all the coatings have inhibitory properties against *E. coli* and *S aureus.* Moreover, all the coatings have made the paper flame retardant, but barium borate imparts better flame retardancy. When examining the contact angles, it has been established that rosemary oil makes the starch coating slightly hydrophobic, while barium borate makes it hydrophilic, and thus, the printability of the paper surface is improved when using both components in the coating formulation. All coated papers were successfully printed with screen printing.

## REFERENCES


[1] E. A. Kandirmaz, H. Birtane, A. Beyler Cigil and A. Ozcan, *Flavour. Fragr. J.*, **35**, 174 (2020), https://doi.org/10.1002/ffj.3549

[2] A. C. Atti-Santos, M. Rossato, G. F. Pauletti, L. D. Rota, J. C. Rech *at al.*, *Braz. Arch. Biol. Technol.*, **48**, 1035 (2005), https://doi.org/10.1590/S1516-89132005000800020

[3] A. Bahrami, R. Delshadi, E. Assadpour, S. M. Jafari and L. Williams, *Adv. Colloid Interface Sci.*, **278**, 102140 (2020), https://doi.org/10.1016/j.cis.2020.102140

[4] S. El-Sherbiny, F. Morsy, M. Samir and O. A. Fouad, *Appl. Nanosci.*, **4**, 305 (2014), https://doi.org/10.1007/s13204-013-0196-y

[5] K. Ghule, A. V. Ghule, B. J. Chen and Y. C. Ling, *Green Chem.*, **8**, 1034 (2006), https://doi.org/10.1039/B605623G

[6] M. Sharma, R. Aguado, D. Murtinho, A. J. Valente, A. P. M. De Sousa *at al.*, *Int. J. Biol. Macromol.*, **162**, 578 (2020), https://doi.org/10.1016/j.ijbiomac.2020.06.131

[7] H. Kjellgren, M. Gällstedt, G. Engström and L. Järnström, *Carbohyd. Polym.*, **65**, 453 (2006), https://doi.org/10.1016/j.carbpol.2006.02.005

[8] J. Ju, X. Chen, Y. Xie, H. Yu, Y. Guo *at al.*, *Trends Food Sci. Technol.*, **92**, 22 (2019), https://doi.org/10.1016/j.tifs.2019.08.005

[9] N. Lavoine, I. Desloges, B. Khelifi and J. Bras, *J. Mater. Sci.*, **49**, 2879 (2014), https://doi.org/10.1007/s10853-013-7995-0

[10] P. Sakare, A. K. Bharimalla, J. Dhakane-Lad and P. G. Patil, *J. Nat. Fibers*, **18**, 1974 (2021), https://doi.org/10.1080/15440478.2019.1710652

[11] S. Lohmander and M. Rigdahl, *Nord. Pulp Pap. Res. J.*, **15**, 231 (2000), https://doi.org/10.3183/npprj-2000-15-03-p231-236

[12] P. Vernhes, M. Dubé and J. F. Bloch, *Appl. Surface Sci.*, **256**, 6923 (2010), https://doi.org/10.1016/j.apsusc.2010.05.004

[13] P. Vernhes, A. Blayo, J. Bloch and B. Pineaux, *Appl. Optics*, **47** 5429 (2008), https://doi.org/10.1364/AO.47.005429

[14] A. Ozcan, *J. Appl. Biomater. Funct. Mater.*, **17**, (2019), https://doi.org/10.1177/2280800018816012

[15] A. Gimat, A. Michelin, O. Belhadj, E. Pellizzi, P. Massiani *at al.*, *Cellulose*, **28**, 2419 (2021), https://doi.org/10.1007/s10570-020-03655-z

[16] C. J. Ridgway and P. A. Gane, *Nord. Pulp Pap. Res. J.*, **18**, 24 (2003), https://doi.org/10.3183/npprj-2003-18-01-p024-031

[17] G. Engström, in *Procs. 13th Fundamental Research Symposium*, 2005, p. 1011, https://doi.org/10.15376/frc.2005.2.1011

[18] S. Bhardwaj and N. K. Bhardwaj, *Nord. Pulp Pap. Res. J.*, **34**, 343 (2019), https://doi.org/10.1515/npprj-2018-0063
[19] R. Bown, in "Paper Chemistry", edited by J. C. Roberts, Springer, 1996, p. 194, https://doi.org/10.1007/978-94-011-0605-4_11
[20] A. Ozcan and M. Oktav, *Asian J. Chem.*, **23**, 2685 (2011)
[21] R. Irkin and O. K. Esmer, *J. Food Sci. Technol.*, **52**, 6095 (2015), https://doi.org/10.1007/s13197-015-1780-9
[22] I. Majid, G. A. Nayik, S. M. Dar and V. Nanda, *J. Saudi Soc. Agric. Sci.*, **17**, 454 (2018), https://doi.org/10.1016/j.jssas.2016.11.003
[23] P. S. Taoukis and T. P. Labuza, *J. Food Sci.*, **54**, 783 (1989), https://doi.org/10.1111/j.1365-2621.1989.tb07882.x
[24] J. Ju, Y. Xie, Y. Guo, Y. Cheng, H. Qian *at al.*, *Crit. Rev. Food Sci. Nutr.*, **60**, 2825 (2018), https://doi.org/10.1080/10408398.2018.1503590
[25] S. Prabuseenivasan, M. Jayakumar and S. Ignacimuthu, *BMC Compl. Altern. Med.*, **6**, 1 (2006), https://doi.org/10.1186/1472-6882-6-39
[26] E. A. Kandirmaz and A. Ozcan, *Nord. Pulp Pap. Res. J.*, **34**, 507 (2019), https://doi.org/10.1515/npprj-2019-0034
[27] E. A. Kandırmaz, *Nord. Pulp Pap. Res. J.*, **36**, 323 (2021), https://doi.org/10.1515/npprj-2021-0014
[28] N. Bousbia, M. A. Vian, M. A. Ferhat, E. Petitcolas, B. Y. Meklati *at al.*, *Food Chem.*, **114**, 355 (2009), https://doi.org/10.1016/j.foodchem.2008.09.106
[29] M. Shau, P. Tsai, W. Teng and W. Hsu, *Eur. Polym. J.*, **42**, 1899 (2006), https://doi.org/10.1016/j.eurpolymj.2006.02.016
[30] H. Lu, L. Song and Y. Hu, *Polym. Adv. Technol.*, **22**, 379 (2011), https://doi.org/10.1002/pat.1891
[31] A. Özcan, N. Kasikovic, E. A. Kandırmaz, S. Durdevic and S. Petrovic, *Polym. Adv. Technol.*, **31**, 2647 (2020), https://doi.org/10.1002/pat.4991
[32] E. A. Kandirmaz, *J. Graph. Eng. Design*, **11**, 47 (2020), http://doi.org/10.24867/JGED-2020-1-047
[33] E. Ural, *Cellulose Chem. Technol.*, **56**, 141 (2022), https://doi.org/10.35812/CelluloseChemTechnol.2022.56.13
[34] W. El Khatib, B. Youssef, C. Bunel and B. Mortaigne, *Polym. Int.*, **52**, 146 (2003), https://doi.org/10.1002/pi.1009
[35] Y. L. Chang, Y. Z. Wang, D. M. Ban, B. Yang and G. M. Zhao, *Macromol. Mater. Eng.*, **289**, 703 (2004), https://doi.org/10.1002/mame.200400064
[36] E. Tsantaki, T. Velegraki, A. Katsaounis and D. Mantzavinos, *J. Hazard. Mater.*, **207**, 91 (2012), https://doi.org/10.1016/j.jhazmat.2011.03.107
[37] W. G. Woods, *Environ. Health Perspect.*, **102**, 5 (1994), https://doi.org/10.1289/ehp.94102s75
[38] E. H. Ezechi, M. H. Isa, S. R. Kutty and N. B. Sapari, in *Procs. 2011 National Postgraduate Conference*, 2011, p. 1, https://doi.org/10.1109/NatPC.2011.6136374
[39] M. Kabu and M. S. Akosman, *Rev. Environ. Contam. Toxicol.*, **225**, 57 (2013), https://doi.org/10.1007/978-1-4614-6470-9_2
[40] C. R. Rejeesh and K. K. Saju, *J. Wood Sci.*, **64**, 697 (2018), https://doi.org/10.1007/s10086-018-1747-3
[41] D. P. Singh, J. Beloy, J. K. McInerney and L. Day, *Food Chem.*, **132**, 1161 (2012), https://doi.org/10.1016/j.foodchem.2011.11.045
[42] S. Meier, F. Moore, A. Morales, M. E. González, A. Seguel *at al.*, *Plant Physiol. Biochem.*, **151**, 673 (2020), https://doi.org/10.1016/j.plaphy.2020.04.025
[43] ISO 12647-5:2015 Graphic technology – Process control for the manufacture of half-tone colour separations, proof and production prints – Part 5: Screen printing
[44] E. Ural and E. A. Kandirmaz, *J. Appl. Biomater. Funct. Mater.*, **16**, 23 (2018), https://doi.org/10.5301/jabfm.5000378
[45] W. S. Mokrzycki and M. Tatol, *Mach. Graph. Vis.*, **20**, 383 (2011)
[46] A. Ozcan, E. A. Kandirmaz, P. Hayta and B. Mutlu, *Cellulose Chem. Technol.*, **53**, 307 (2019), https://doi.org/10.35812/CelluloseChemTechnol.2019.53.30
[47] M. Kilinc, G. O. Cakal, G. Bayram, I. Eroglu and S. Özkar, *J. Appl. Polym. Sci.*, **132**, 42016 (2015), https://doi.org/10.1002/app.42016
[48] R. D. Houlsby, M. Ghajar and G. O. Chavez, *Antimicrob. Agents Chemother.*, **29**, 803 (1986), https://doi.org/10.1128/AAC.29.5.803
[49] A. Jafari-Sales and M. Pashazadeh, *Int. J. Life Sci. Biotechnol.*, **3**, 62 (2020), https://doi.org/10.38001/ijlsb.693371