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EVALUATION OF THE MIGRATION POTENTIAL OF COLORANTS AND ADDITIVES IN PAPERBOARD FOOD PACKAGING MATERIALS

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Abstract: *The increasing implementation of wastepaper collection and recovery has led to an increase in the use of recycled paper and board as food packaging materials. These materials, which are made entirely or partially from recycled fibres, are widely used for food packaging in Europe. However, the potential presence of contaminants in recycled pulp raises concerns about their suitability for food contact applications due to the potential for migration issues.*

In this study, three different paperboard food packaging materials from the Croatian market were tested for their chemical stability and migration of potentially harmful substances. The materials included a Kraftliner sample (made of predominantly virgin fibre pulp), a Brown Testliner sample (top ply made of virgin fibres, bottom ply made of recycled fibre pulp) and a printed French Fries Box (combination of deinked and mechanically recycled pulp). The French Fries Box also contained pigment coating and a partial internal print.

The research focused on testing the migration of colorants (EN 646) and fluorescent whitening agents (FWAs) (EN 648) using food simulants such as olive oil, 3% (m/v) aqueous acetic acid, deionized water, isooctane and saliva simulant. In these test methods the test specimen is placed between two glass fibre papers previously saturated with the test liquids and, after the contact time at room temperature, the bleeding of the colorants or FWAs onto the glass fibre paper is evaluated. In addition, the potential transfer of antimicrobial components, indicating the potential toxicity of the material, was also evaluated by testing the growth inhibition of the bacterium Bacillus subtilis (EN 1104). The results of all analyzes performed were compared with the purity requirements for materials set out in the German national regulation and CEPI's Food Contact Guidelines for the Compliance of Paper and Board Materials and Articles.

The results of migration of colorants were observed only in the Testliner sample in cases where food simulants such as water, acetic acid and saliva solution were used. Unacceptable bleeding of FWAs was confirmed in the French Fries Box sample where the highest level of bleeding was observed in the case when food simulant was water, and to a slightly lesser extent FWAs migrated into glass fibre papers saturated with acetic acid and saliva food simulant. The results of the microbiological tests were positive, as no inhibition zones for bacterial growth were detected in any of the tested paperboards, meaning that none of the samples released antimicrobial substances.

Key words: food packaging, recycled paperboard, colorant migration, FWAs, antimicrobial transfer

1. INTRODUCTION

In the European Union, the growing interest in environmental issues has led to the collection and recovery of increasing amounts of paper waste materials. According to Cepi Key Statistics, in 2022, 70.5% of all paper and board consumed in Europe was recycled, making it one of the most collected and recycled materials in the region (Cepi, 2022). Furthermore, 73.9% of the collected paper for recycling was utilized in packaging paper and board production. Compared to 2020 (73.9%) (Cepi, 2020) and 2021 (72.8%) (Cepi, 2022), the recycling rate has slightly declined, primarily due to reduced paper production and the high costs of electricity and gas, which have negatively impacted paper recycling mills. However, the current recycling rate of 70.5% still places Europe well ahead of the global average recycling rate of 59.9% in 2021 (EPRC, 2022).

In 2021, the European paper value chain pledged to recycle 76% of all paper consumed by 2030, aiming for a 76% recycling rate. The recycling rate is defined as the ratio between the recycling of used paper, including the net trade of paper for recycling, and the consumption of new paper and board. This commitment is outlined in the new European Declaration on Paper Recycling 2021-2030, indicating that the recycling rate will continue to rise with these efforts (EPRC, 2022).

As the collection and recycling of waste paper has been introduced on a larger scale, the use of recycled paper and board as packaging material for food has also increased. In many European countries, paper and board made partly or entirely from recycled fibres are used as primary or secondary packaging for food. However, depending on the origin of the recycled fibres and the way they are prepared and treated, it is possible that even after recycling, potentially harmful substances may remain in the recycled pulp in concentrations that can lead to unacceptable migration, making the recycled paper and board unsuitable for food contact applications (Jamnicki et al., 2015; FoodDrinkEurope, 2016). In recent studies on paper food packaging, various substances such as phthalates, solvents, primary aromatic amines and mineral oils have been detected in recycled pulp (Boccacci Mariani, Chiacchierini & Gesumundo, 1999; Summerfield & Cooper, 2001; Binderup et al., 2002; Biedermann, Uematsu, & Grob, 2011; Suciu et al., 2013; Pivnenko, Eriksson & Astrup, 2015; Vápenka, Vavrouš, & Votavová, 2016; Boccacci Mariani et al., 2015; Geueke, Groh & Muncke 2018). To ensure food safety in packaging made from recycled fibres, the recovered paper grade must be carefully selected, and the material must undergo appropriate treatment and cleaning processes. This ensures that the finished product complies with the general requirements of the EU Framework Regulation (EC) No. 1935/2004 (European Commission, 2004) and Regulation (EC) No. 2023/2006 on Good Manufacturing Practices (GMP) (European Commission, 2006). These regulations stipulate that materials in contact with food must not transfer harmful substances, alter the composition of the food, or impair its sensory properties (Boccacci Mariani et al., 2015; European Commission, 2004; European Commission, 2006).

However, aside from the general requirements outlined in these regulations, there is currently no harmonized legislation in the EU for the use of recycled paper in food packaging. As a result, some Member States have implemented their own laws. National regulations often define the chemicals permitted in the manufacture of paper and board and set limits for various contaminants in finished products (Von Wright, 2007).

The recommendation of the German Federal Institute for Risk Assessment (BfR), known as the *BfR Recommendations on Food Contact Materials* (BfR, 2023), is regarded as the most reputable regulation in Europe for paper packaging (Vápenka, Vavrouš & Votavová 2016). In the absence of uniform legislation across Europe, to help manufacturers of paper and board navigate the various regulations in force, the European paper and board industry, along with converting sectors and other associations, has published a guideline document entitled *Food Contact Guidelines for the Compliance of Paper and Board Materials and Articles* (CEPI/CITPA, 2019). This document, formally referred to as the *CEPI Industry Guideline*, along with the BfR recommendations, specifies migration limits for contaminants commonly found in recycled paper and board intended for food contact applications. Although these limits are not mandatory, paper manufacturers are encouraged to comply voluntarily if they wish to achieve higher safety standards for food packaging containing recycled fibres.

In this research, three different paperboard food packaging materials were subjected to testing for chemical stability in terms of not releasing colorants from recycled and/or coloured paper and board. Additionally, the potential migration of fluorescent whitened agents (FWAs) was tested, as well as the possible transfer of antimicrobial constituents indicating the potential toxicity of the tested samples. The results of the conducted analyses were compared with the quantitative restrictions laid down in the German (BfR) regulation (BfR, 2023) and CEPI Industry Guideline (CEPI/CITPA, 2019).

2. MATERIALS AND METHODS

Three different paperboard packaging materials were chosen for this study (Table 1) taken from Croatian market – a Kraftliner sample containing mostly virgin fibre pulp, a Brown Testliner sample having the top ply made of virgin fibres, and the bottom ply made of recycled-fibre pulp. Both linerboards were unprinted. The third simple was a printed French Fries Box used for fast food applications. French Fries Box was made of a combination of deinked and mechanically recycled pulp. The outer surface of the box was coated with pigment coating and contained a printed design on top. In addition, the inner, food contact side of the box, was also partially printed with yellow ink that covered approximately 40% of the inner surface.

Table 1: Properties of tested paperboard samples

	Basis weight (g/m^2)	Moisture Content (%)
Kraftliner	139	6.3
Brown Testliner	129	4.0
Printed French Fries Box	252	5.3

2.1 Determination of bleed fastness of FWAs and colorants

The transfer of fluorescent whitened agents (EN 648) (European Standard, 2006b) and colorants (EN 646) (European Standard, 2006a) from the paperboard samples were tested with test fluids such as rectified olive oil, 3% (m/v) aqueous acetic acid, deionized water, iso-octane and saliva simulant. In these test procedures, the sample is brought into contact with glass-fibre papers previously saturated with a test fluid (food simulant) and placed under a 1 kg load for 24 h. For each sample, the test was performed with one replication and the results are presented as the arithmetic mean of two measurements. Both sides of the paperboard were subjected to the tests. The staining of the glass fibre paper was evaluated comparatively with a series of fluorescent whitened papers under the UV light in the UV-A range (for the detection of FWAs) or against a grey scale (for colour fastness analysis). For both test procedures no staining of the fibre glass paper should be obtained, meaning that the value 5 on the evaluation scale must be reached – according to the German (BfR) regulation and CEPI Industry Guideline. This is especially important in case of packing the moist and/or fatty foods (CEPI/CITPA, 2019).

2.2 Determination of the transfer of antimicrobial constituents

The selected paperboard samples were tested for their effect on the growth of the bacterium *Bacillus subtilis*. The method outlined in EN 1104 (European Standard, 2005) is widely used for testing the antimicrobial activity of materials. This method is based on detecting the inhibition zone in bacterial growth, which results from the diffusion of antimicrobial agents in agar plates. In this test procedure, Petri dishes were prepared with *Bacillus subtilis*, and three test pieces from each paperboard sample were placed on the semi-solid nutrient medium. The plates were then incubated for 3 days at 30°C to allow bacterial growth. After incubation, the presence of an inhibition zone (an area with no bacterial growth) indicated the release of antimicrobial constituents. The test determines whether the paperboard samples release antimicrobial substances in quantities sufficient to inhibit bacterial growth, thereby reflecting the toxicity of the tested papers. Each sample was tested in duplicate. According to the German BfR regulation and CEPI Industry Guideline "the finished paper or paperboard must have no preserving effect on the foodstuffs with which they come into contact". This requirement applies to all types of foods (CEPI/CITPA, 2019). Samples with no evidence of an inhibition zone are considered to lack water-soluble antimicrobial substances. Therefore, overgrown samples (without an inhibition zone) are evaluated as suitable for food contact.

3. RESULTS AND DISCUSSION

The bleeding of colorants was evaluated by measuring the lightness difference (ΔY_{tristm}) of glass fiber filter paper before and after the migration (exposure) test. The surface of the filter paper, saturated with the prescribed food simulant solution and in contact with the test piece during the test, was measured with a spectrophotometer both before and after the migration test (measuring only the tristimulus value Y). The resulting lightness difference was then compared to a defined lightness scale, known as the Grey Scale (ΔY_{tristm}), in accordance with the EN 20105-A03 standard (European Standard, 2003). The results of the evaluation of colorant migration from one side (A) and the other side (B) of the tested samples are shown in Tables 2-4.

Table 2: Results of colorants migration from the Kraftliner sample

Kraftliner	H_2O	CH_3COOH	Saliva simulant	Iso-octane	Olive oil
(A) $\Delta Y_{tristm.}$	0.20	0.53	0.55	0.33	2.98
GRADE	5	5	5	5	4-5
(B) $\Delta Y_{tristm.}$	0.56	0.40	0.44	0.39	1.18
GRADE	5	5	5	5	5

Table 3: Results of colorants migration from the Brown Testliner sample

Testliner	H_2O	CH_3COOH	Saliva simulant	Iso-octane	Olive oil
(A) $\Delta Y_{tristm.}$	10.08	9.33	11.06	1.38	3.48
GRADE	3	3	2-3	5	4-5
(B) $\Delta Y_{tristm.}$	7.13	9.60	8.92	0.92	1.41
GRADE	3	3	3	5	5

Table 4: Results of colorants migration from the Printed French Fries Box sample

Printed Box	H_2O	CH_3COOH	Saliva simulant	Iso-octane	Olive oil
(A) $\Delta Y_{tristm.}$	0.00	0.13	0.22	0.33	2.98
GRADE	5	5	5	5	4-5
(B) $\Delta Y_{tristm.}$	0.26	0.00	0.84	0.59	1.01
GRADE	5	5	5	5	4-5

Table 5 shows the criteria used to evaluate the migration of colorants from the tested samples onto filter paper. Based on the defined lightness difference ($\Delta Y_{tristm.}$), the tested sample is assigned values ranging from 1 to 5 (in 9 degrees, including half steps). According to the German BfR regulation and CEPI Industry Guideline value 5 on the evaluation scale should be obtained, meaning there is no migration of colorants to the foodstuff. Test pieces having a value less than 5 are declared unsuitable for contact with certain types of food, depending on the food simulant for which the negative rating was obtained.

Table 5: Comparison of the range of lightness difference values (ΔY_{tristm}) with the corresponding rating on the Grey Scale (EN 20105-A03)

Fastness grade	$\Delta Y_{tristm.}$	Tolerance
5	0.0	0.2
4-5	2.2	±0.3
4	4.3	±0.3
3-4	6.0	±0.4
3	8.5	±0.5
2-3	12.0	±0.7
2	16.9	±1,0
1-2	24.0	±1,5
1	34.1	±2.0

When reviewing the results for all tested samples, it is evident that only the Brown Testliner sample showed unacceptable results in the case of water, acetic acid, and saliva food simulants. This means that such a paperboard should not be used in contact with moist food, nor with food that has an acidic character. For all other tested samples, positive results were obtained for all standard food simulants, indicating that there is no risk of colorant migration from such materials.

Tables 6 and 7 show the results of the migration of fluorescent whitening agents (FWAs) from one side (A) and the other side (B) of the tested paperboard samples. The staining of the glass fibre papers on the side that was in contact with the sample was compared with a series of comparison fluorescent papers under a UV lamp. UV light in the UV-A range was used to illuminate the samples. The comparison filter papers were prepared as follows: One gram of the fluorescent whitening agent (Fluorescent Whitener 28, content 90%, C.I. 40622, CAS number: 4193-55-9) was dissolved in one litre of deionized water to prepare stock solution (1000 mg/l), which was then diluted with water in four different ratios. Specifically, four different volumes of the stock solution (125 ml, 31 ml, 8 ml and 3 ml) were taken and each diluted with one litre of deionized water. The prepared diluted solutions were dripped onto glass fibre papers, which were then air-dried without exposure to light and used as comparison standards after drying. The evaluation criteria are listed in Table 8 (European Standard, 2006b). According to the German BfR regulation and the CEPI Industry Guideline, a score of 5 is considered positive, while lower results (4, 3, 2 and 1) are considered negative, indicating that the tested sample releases fluorescent whitening agents into certain types of food in quantities exceeding the permitted limits. Food simulants used in this test procedure were deionized water, 3% (m/v) aqueous acetic acid, saliva simulant and olive oil.

Table 6: Results of the migration of FWAs measured on side A of the samples

Side A of the sample	Kraftliner	Brown Testliner	Printed Box
Food simulant			
H_20	5	5	3
CH_3COOH	5	5	4
Saliva simulant	5	5	4
Olive oil	5	5	5

The results presented in Tables 7-8 show that significant migration of FWAs only occurred in the printed French Fries Box when water was used as a food simulant. To a slightly lesser extent, FWAs also migrated into glass fibre papers saturated with acetic acid and saliva simulants. This suggests that such packaging should not be used in direct contact with aqueous or acidic foods. Notably, stronger migration of FWAs from this sample was observed on the "A" side (the inner side of the box) when exposed to the aqueous medium, while the outer "B" side showed a more moderate migration (rating 4). As this carton is used for the packaging of French fries, which have a high fat content, the migration into the glass fibre paper saturated with olive oil received an acceptable score of 5. This indicates that this type of packaging can be safely used in direct contact with fatty foods as intended. Previous studies (MAFF, UK, 1995) have shown that FWAs are soluble in water and aqueous solutions, but not in the oily or fatty components of food.

Table 7: Results of the migration of FWAs measured on side B of the samples

Side B of the sample	Kraftliner	Brown Testliner	Printed Box
Food simulant			
H_20	5	5	4
CH_3COOH	5	5	4
Saliva simulant	5	5	4
Olive oil	5	5	5

Table 8: Evaluation criteria (EN 648)

Evaluation grade	FWA-solution	FWA mg/l
1	125 ml stock solution diluted to 1 l	125
2	31 ml stock solution diluted to 1 l	31
3	8 ml stock solution diluted to 1 l	8
4	3 ml stock solution diluted to 1 l	3
5	blank with distilled water	0

The results of testing the transfer of antimicrobial substances from paperboard samples showed that none of the analyzed samples released substances with antimicrobial properties, as no inhibition zones for the growth of bacterial cultures were detected (Figures 1-3). From a microbiological point of view, all analyzed samples can therefore be considered safe for direct contact with food, as the bacterial culture *Bacillus subtilis* was observed to have overgrown the paperboard samples in all tested cases.

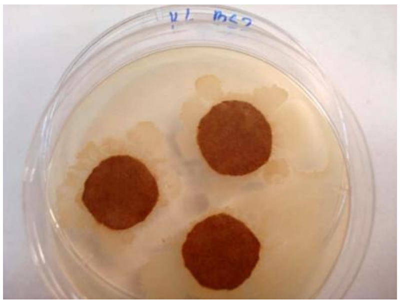

Figure 1: Overgrown cultures of Bacillus Subtilis on Kraftliner samples

Figure 2: Overgrown cultures of Bacillus Subtilis on Brown Testliner samples

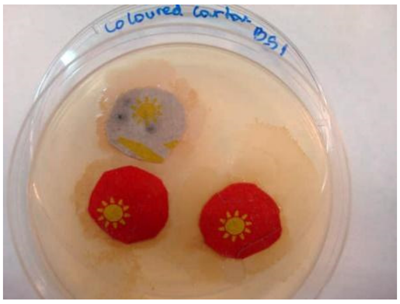

Figure 3: Overgrown cultures of Bacillus Subtilis on Printed French Fries Box samples

5. CONCLUSIONS

In this study, the chemical stability and migration of potentially harmful substances from three different paperboard food packaging materials available on the Croatian market, consisting of recycled and virgin fibres, were evaluated. Based on the results, the following conclusions can be drawn:
Migration of colorants was only observed in the Testliner sample when exposed to aqueous, acidic, and saliva food simulants. This indicates that such materials should not be used in direct contact with moist or acidic foods due to potential contamination risks. Other tested samples showed no colorant migration, making them suitable for various food packaging applications.
Significant migration of Fluorescent Whitening Agents was observed in the printed French Fries Box when it came into contact with water and to a lesser extent with acetic acid and saliva simulants. This suggests that the French Fries Box should not be used for packaging aqueous or acidic foods. However, it showed acceptable results when tested with olive oil, confirming its suitability for packaging fatty foods like French fries.

None of the paperboard samples released antimicrobial substances that could inhibit bacterial growth, as no inhibition zones were detected in any of the tests. From a microbiological point of view, all tested materials can therefore be considered safe for direct contact with food.
Materials made from virgin fibres or properly treated recycled fibres comply with safety guidelines and can be considered safe for food packaging based on the results of the food contact analyzes performed in this study, provided they are used with appropriate foods, especially with regard to moisture and acidity. To definitively prove that these materials are truly suitable for food contact, further testing is required, e.g. testing the concentration of certain chemical contaminants commonly found in paper packaging.
These results support the careful selection of paperboard materials for specific food contact applications, especially when recycled fibres are involved, to ensure food safety and compliance with industry regulations.

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