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Research Article

Ethylene Vinyl Alcohol Copolymer (EVOH) as a Functional Barrier against Surrogate Components Migrating from Paperboard

Caroline Maes,[1,2] **Giberto Mitsuyoshi Yuki Junior**,[3] **Cynthia Teniers**,[1] **Wout Luyten**,[1] **Geert Herremans**,[1] **Roos Peeters**,[2] **Pieter Samyn**,[4] **Robert Carleer**,[4] **and Mieke Buntinx**[2]

[1]*Kuraray-Eval Europe NV, Haven 1053 Nieuwe Weg 1, Bus 10, 2070 Zwijndrecht, Belgium*
[2]*Hasselt University, Packaging Technology Center IMO-IMOMEC, Wetenschapspark 27, 3590 Diepenbeek, Belgium*
[3]*Université de Pau et des Pays de l'Adour, École Nationale Supérieure en Génie des Technologies Industrielles, 64000 Pau, France*
[4]*Hasselt University, Applied and Analytical Chemistry IMO-IMOMEC, Agoralaan Building D, 3590 Diepenbeek, Belgium*

Correspondence should be addressed to Mieke Buntinx; mieke.buntinx@uhasselt.be

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Ethylene vinyl alcohol copolymer (EVOH) is a key material of interest as a functional barrier against substances migrating from recycled paperboard, due to its outstanding barrier properties. Three multilayer films containing two different grades of EVOH, L171B (3 μm) and F171B (3 and 5 μm), were benchmarked against a multilayer film containing polyamide 6/6.6 copolymer (PA 6/6.6, 3 μm) and monolayer polyethylene terephthalate (PET, 12 μm). The 5 films were evaluated as barrier materials against 5 surrogate substances simulating different migrants potentially present in recycled paperboard: *n*-heptadecane (C17) as a mineral oil-saturated hydrocarbon (MOSH), 4-methylbenzophenone (MBP) as a photoinitiator, di-*n*-propyl phthalate (DPP) as a plasticiser, and anthracene (ANT) and perylene (PER) as mineral oil aromatic hydrocarbons (MOAHs). The test was accelerated at 60°C for 25 days, which is equivalent to a shelf life of 2 years at 25°C. All films containing 3 or 5 μm EVOH were found to be good barriers, showing no breakthrough values over 1% of the initial concentration found in the paperboard, and they could easily compete with 12 μm PET. The multilayer with 3 μm PA 6/6.6 showed higher breakthrough values for both MBP and DPP than the other materials although still below the 1% threshold value. However, ANT showed substantial breakthrough values of nearly 2%, indicating that PA 6/6.6 might not offer enough protection against low-weight MOAH components.

1. Introduction

Mineral oil hydrocarbons (MOHs) contamination in food has become a major concern in the past decade when it was brought to light that these substances might be potentially harmful for the human health. Both the German Federal Institute for Risk Assessment (BfR) [1] and the European Food Safety Authority (EFSA) [2] published a scientific opinion on the potential health risks of MOHs present in food due to contamination in the food chain.

In terms of MOHs contamination and toxicology, the EFSA [2] distinguishes two main groups: mineral oil-saturated hydrocarbons (MOSHs), which can be *n*-alkanes, branched alkanes and cycloalkanes, and mineral oil aromatic hydrocarbons (MOAHs), which are polyaromatic hydrocarbons, mainly alkylated. MOSHs from C_{16} to C_{35} may accumulate and cause microgranulomas in the lymph nodes, spleen, liver, and other tissues [2]. For MOAHs, there are only few toxicological data available; however, MOAHs with 3 or more nonalkylated and minor alkylated aromatic rings are suspected for being mutagenic and carcinogenic and are therefore of concern [2]. Due to these findings, the EFSA also recommends that the revision of the acceptable daily intake (ADI) of some food-grade MOSHs is necessary.

Migration from recycled paperboard was found to be one of the main sources for this contamination. Several

studies reported severe migration of both MOSHs and MOAHs from paperboard packaging into foodstuff which was not only limited to the primary but also originated from the secondary and even the tertiary packaging [3–7]. Lorenzini et al. [7] found that about 60–80% of the $<n\text{-}C_{24}$ fraction of MOSHs/MOAHs present in the paperboard was transferred to the food inside the package. Vollmer et al. [6] confirmed these findings and also reported that 10–20% of the migrated fraction were MOAHs.

Regulation (EC) No. 1935/2004 [8] states that food contact materials (FCMs) should not transfer any of their constituents to food in quantities that may endanger human health and bring about unacceptable changes in the composition of the food or a deterioration in the organoleptic characteristics thereof. Unfortunately, so far, no harmonised European Regulation for paperboard packaging exists. Recently, Commission Recommendation (EU) 2017/84 [9] has been issued to monitor the presence of MOHs and their sources in 2017 and 2018. The results of this study are expected in 2019. Several countries already took the lead by establishing their own recommendation or regulation; for instance, in Austria, the Federal Ministry of Health states in Recommendation BMG-75210/0018-II/B/13/2012 [10] that if food is packaged in recycled paperboard, it should be protected by a functional barrier. Switzerland has a similar approach in its "Bedarfsgegenständeverordnung" 817.023.21 [11], which states that recycled paperboard should not be in contact with food, unless measures are taken, e.g. by means of a barrier layer.

Even though there is no specific regulatory obligation on the European level, it is now generally recommended to minimise the transfer of MOHs and other harmful components migrating from paperboard packaging [1, 2, 12, 13]. The most widely accepted solution is the integration of a functional barrier in the primary packaging concept, where it can limit the migration from primary, secondary, and tertiary packaging as well as the cross-contamination from other products and the environment [5]. A functional barrier can either limit the maximum migration into food to a quantity below the specific migration limit (SML) or can prolong the time of breakthrough when migrated components are first detected, to a time beyond the shelf life [14].

Ethylene vinyl alcohol copolymer (EVOH) (see the structure in Figure 1) is a key material of interest due to its outstanding barrier properties as reported by Maes et al. [15]. The random copolymer consists of ethylene units, which contribute to processability and flexibility, and vinyl alcohol units, which are responsible for the high barrier properties. EVOH has one of the lowest oxygen permeability rates (P_{O_2}) amongst polymers, and it shows excellent barrier properties against other gases, such as nitrogen and carbon dioxide. In addition, it also prevents the scalping of aromas and flavours in food packaging [16–18]. EVOH also offers a good barrier against fuels and other chemicals, such as benzene, toluene, ethylbenzene and xylene isomers (BTEX), agrochemicals including fumigants, and others. In the automotive sector, EVOH is already widely used in applications like fuel tanks [15].

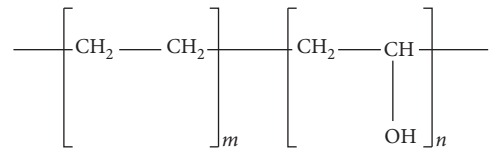


Figure 1: Structure of ethylene vinyl alcohol copolymer (EVOH).

Maes et al. [19] already demonstrated that EVOH offers good protection against mineral oil migration. The breakthrough time of a mineral oil mixture (Gravex 913) was determined for two EVAL™ EVOH grades, E105B and F171B, using the method developed by Fiselier and Grob [14]. The multilayer films containing 5.2 and 4.4 μm EVOH, respectively, showed breakthrough times of, respectively, >7.1 and >10.6 years [19].

More recent studies are focussing on the use of surrogate substances instead of a mineral oil mixture to assess the functional barrier against mineral oil and other contaminants originating from recycled paperboard [20, 21]. This approach allows careful evaluation of known components with different polarity and boiling points instead of total migration. Based on these studies, the Schweizerisches Verpackungsinstitut (SVI) published a guideline for the evaluation of the barrier efficiency of internal bags for food packaging in recycled paperboard [22]. Because the shelf life of most dry foods is several years, a test at regular storage conditions (25°C) would take too long; however, the test can be accelerated by increasing the temperature. The acceleration factor can be calculated using the Arrhenius equation (Equation 1) as described in Commission Regulation (EU) No. 10/2011 [9], which suggests an activation energy (E_a) of 80 kJ/mol as a worst-case scenario. A test performed at 60°C would be accelerated by factor 30 compared to a test at 25°C, reducing the test time for a shelf time of two years to 25 days:

$$t_2 = t_1 \cdot e\left[-\frac{E_a}{R} \cdot \left(\frac{1}{T_1} - \frac{1}{T_2}\right)\right]. \quad (1)$$

In this paper, EVOH was evaluated as a functional barrier against 5 surrogate substances simulating a MOSH, two MOAHs, a plasticiser, and a printing ink component, migrating from recycled paperboard.

2. Materials and Methods

2.1. Chemicals and Materials. Methyl tert-butyl ether (MTBE, 100%) and methanol (MeOH, 98.5%) were obtained from VWR International S.A.S. (Fontenay-sous-Bois, France). Anthracene (ANT, 99%) and bicyclohexyl (CyCy, 99%) were from Acros Organics (Geel, Belgium). Di-*n*-propyl phthalate (DPP, 98%), *n*-heptadecane (C17, 99%), 4-methylbenzophenone (MBP, 98%), and Sudan red II (1(2,4-dimethylphenylazo)-2-naphthol) all were supplied by Alfa Aesar-Thermo Fisher GmbH (Kandel, Germany). Finally, perylene (PER, synthesis grade) was obtained from Merck KGaA (Darmstadt, Germany).

For this study, five different film samples were evaluated. Four of these films were multilayer structures which were

produced on a Dr. Collin 5-layer blown film line by the TS Department of EVAL Europe N.V., and one was a monolayer polyethylene terephthalate (PET) film (12 μm) obtained through the EVAL R&D Department of Kuraray Co. Ltd. in Japan. The general structure of the multilayer films was LDPE/tie/barrier/tie/LDPE. The barrier layer was either EVAL™ EVOH-grade F171B (32 mol% ethylene) of either 5 or 3 μm thickness, 3 μm EVAL™ EVOH-grade L171B (27 mol% ethylene), or 3 μm polyamide 6/6.6 copolymer (PA 6/6.6). The different samples are shown in Table 1. Both structures of PA 6/6.6 and PET are given in Figure 2.

The paperboard used as a donor was of virgin grade (290 g/m^2) which was provided by Smurfit Kappa Van Mierlo Offset Packaging (Turnhout, Belgium). The receptor material was obtained from BGB Analytik AG (Boeckten, Switzerland) and was made of copy paper (115 g/m^2) soaked in 20% polydimethylsiloxane (PDMS), which was cured and cleaned for 8 h at 130°C and 0.1 bar.

2.2. Spiking of the Donors. The spike solution was prepared using *n*-heptadecane (C17), 4-methylbenzophenone (MBP), and di-*n*-propyl phthalate (DPP) as surrogate components and Sudan red II for visual control of equal distribution in the paperboard as listed by Biedermann-Brem et al. [21] and in the guideline provided by the SVI [22]. As shown in Table 2, C17, MBP, and DPP, respectively, represent an MOSH, photoinitiator, and plasticiser, respectively. Yet MOAH components are of the greatest concern; therefore, anthracene (ANT) and perylene (PER) were added to simulate MOAH components (Table 2). All components were dissolved in MTBE at a concentration of 250 mg/L.

The donor paperboard (A5) was soaked in this solution until it was completely saturated; afterwards, it was dried by holding it in a vertical position underneath the fume hood to allow the MTBE to evaporate. Several boards were then tightly wrapped together in aluminium foil and placed in an oven at 40°C for 2 weeks for conditioning. This condition was chosen based on experiments of Biedermann-Brem et al. to allow the polar components to penetrate the paperboard more deeply. This allowed a more gradual release of the components from the paperboard mimicking a more realistic migration scenario [21, 23].

2.3. Preparation of the Test Packs. After conditioning, the donor paperboard was sampled for the determination of the initial concentrations by removing a piece of 9 cm × 2 cm for extraction and subsequent analysis by gas chromatography using a quadrupole mass spectrometer (GC-MS) as detector. The remaining donor was then covered in aluminium foil on the top side and folded around the border with approximately 1 cm overlap on all edges of the bottom side. This resulting donor pack was then taped on top of the test film sample, which had larger dimensions. To ensure the donor was wrapped tightly, the excess of the film sample was folded around the donor pack and taped on top. Finally, the receptor (A5) was secured on the other side of the film sample (Figure 3). Afterwards, the test pack was completely covered in aluminium foil. To ensure the receptor was free of possible contaminations, it was first heat-treated at 150°C for 30 minutes prior to using it in the test pack.

All test packs were put in the oven simultaneously and loaded with a heavy object (e.g. a glass plate) to decrease the air exchange between the packs. The test packs were kept in the oven for 25 days at 60°C, which covers a period of 2 years of shelf life at 25°C, using an acceleration factor of 30 as calculated by the Arrhenius equation. The receptor was sampled at regular intervals by cutting a piece of 9 cm × 2 cm keeping a distance of 1 cm from the border and the previous sampling to avoid the effect of sideways migration due to the absence of the receptor material in the vicinity area. Even though this effect was not found to be considerable, it was found to be a best practice [21]. Each film sample was tested in duplicate.

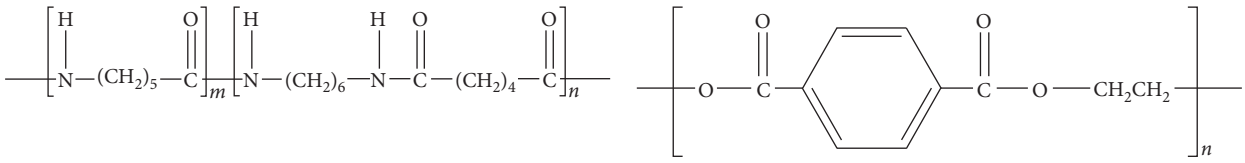
2.4. Extraction and GC-MS Analysis. The samples were extracted using a MTBE/MeOH (95/5 v/v) solution. The 9 cm × 2 m samples of the donor and receptor were cut into smaller pieces and then extracted with, respectively, 10 and 2 mL of the extraction solution. To each of these extracts, 10 μL of the internal standard solution was added, prepared by dissolving 100 mg CyCy in 10 mL MTBE of which 300 μL was then diluted in 10 mL MTBE. The film samples were extracted in the same way as the receptor to check whether there were possible interferences with the analysis of the surrogate substances. The vials containing the samples with extraction solution were put in an oven overnight at 40°C to facilitate the extraction process after the addition of CyCy as the internal standard. Biedermann-Brem et al. found that the extraction was most challenging for the most polar component (MPB) after it has penetrated the board deeply. However, after a night at 40°C, the extraction was virtually complete [23].

The extracts were analysed by injecting 1 μL into a Trace Ultra 1310 GC coupled to an ISQ LT Quadrupole MS in a splitless mode at a constant helium flow of 1.2 mL/min. The column used was a 30 m × 0.25 mm i.d. × 0.25 μm film thickness DB5-MS capillary column. The oven temperature was first kept at 30° C for 5 minutes after injection. Then, at a 10°C/min ramp, the temperature was raised till 330°C, which was maintained for 2 minutes. 6 calibration solutions were prepared to which 10 μL of the internal standard was added to determine the exact amount of the surrogate components in the sample extracts. For each test pack, the amount of migration for each of the 5 substances that passed through the barrier into the receptor was determined. This amount was expressed as a percentage breakthrough value of the initial concentration that was found in the donor paperboard.

2.5. O_2GTR Measurements. The oxygen transmission rate (O_2GTR) of all five film samples was determined using a MOCON OX-TRAN® 2/21 at 20°C and 65% RH in accordance with ASTM F1927. For the PET and PA 6/6.6 film, the use of a mask was necessary to reduce the test surface area from the standard 50 cm^2 to 5 cm^2 to remain within the detection range of the coulometric detector.

Table 1: Film samples and their characteristics.

Barrier material	Ethylene content (mol%)	Average layer distribution LDPE/tie/barrier/tie/LDPE (μm)	Oxygen barrier at 20°C and 65% RH	
			O_2GTR ($cm^3/(m^2 \cdot day \cdot atm)$)	P_{O_2} ($cm^3 \cdot \mu m/(m^2 \cdot day \cdot atm)$)
F171B	32	21/4/5/5/20	0.7	3.2
		21/5/3/5/20	1.7	5.4
L171B	27	22/5/3/5/21	0.7	2.0
PA 6/6.6	/	20/5/3/4/18	479	1629
PET	/	12	91	60



(a) (b)

Figure 2: Structure of polyamide 6/6.6 copolymer (PA 6/6.6) (a) and polyethylene terephthalate (PET) (b).

3. Results and Discussion

3.1. Breakthrough of Surrogate Components. In Figure 4, the breakthrough values of the five surrogate components are given in percent of the initial concentration found in the spiked donor for all films after 25 days at 60°C, which translates into a shelf life of 2 years at 25°C. The guideline provided by the Swiss Packaging Institute states that the barrier is good when the values remain below 1% of the initial concentration at the end of the shelf life [22]. This criterion was based on GC analysis of recycled paperboard extracts initiated by the German Federal Ministry of Food, Agriculture and Consumer Protection (BMELV) and its outcome that no nonevaluated substances in the critical volatility range (C_{14}–C_{25}) exceeded 10 mg/kg paperboard. The BMELV specified a threshold of toxicological relevance of 0.01 mg/kg in food, which corresponds to the detection limit that is widely applied by the EU [24]. Taking into account the typical mass ratio of food to paperboard of 10, this leads to the threshold value of 1% of the substances initially found in the paperboard [23].

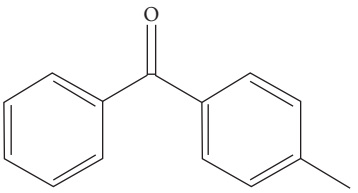
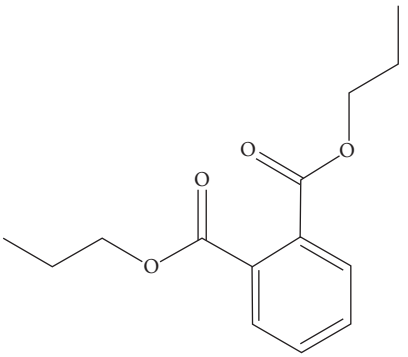
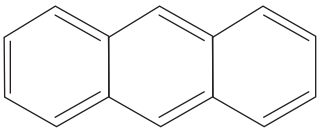
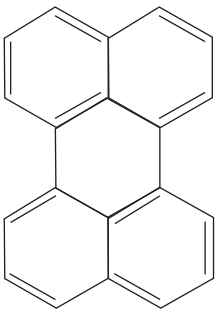
The results in the graph clearly show that the multilayer films with the two different EVAL™ grades, 5 and 3 μm F171 and 3 μm L171, are sufficient and can easily compete with a monolayer 12 μm PET film. For the EVAL™ and PET films, C17 shows the highest migration potential, which is likely due to its low boiling point (302°C) and selectivity of the polymer matrix. Biedermann-Brem and Grob found that polyvinyl alcohol had a higher selectivity for C17 than for MBP and DPP, making it less resistant to C17, resulting in a higher breakthrough percentage than the other two components [20].

The multilayer film containing 3 μm of PA 6/6.6, however, shows higher values for MBP and DPP compared to the other films, but these values are still below the 1% threshold value. It is likely that these components might have caused a plasticising effect on the PA 6/6.6 matrix, which could be less present in PET and EVOH due to stronger inter- and intramolecular bonds caused by the presence of more functional groups [25, 26]. PA 6/6.6 has longer polar chains and is therefore to be expected that the free space between the chains allows larger molecules to pass through. C17 did not show higher breakthrough values than the other two materials, which indicates a strong change in selectivity as was shown by Biedermann-Brem and Grob [20]. ANT is an apolar molecule that shows breakthrough values of nearly 2% after 25 days at 60°C. The threshold value of 1% for ANT was reached after only 5 days at 60°C in the PA 6/6.6 film, which is approximately 150 days at 25°C, indicating that 3 μm PA 6/6.6 does not provide sufficient protection against low-weight MOAH components for products that require a shelf life over 150 days. This effect was possibly strengthened by the plasticising effect of the polar components or interaction between the components DPP, MBP, and ANT such as copermeation. Zhou et al. reported this effect for limonene and ethyl butyrate, where the presence of ethyl butyrate increased the permeation of limonene through a high-density polyethylene (HDPE) film [27]. But further research is needed to confirm these findings.

The component PER did not break through in any of the films, which is most likely due to its higher boiling point of 494°C opposed to C17 (302°C), MBP (326°C), DPP (340°C), and ANT (340°C), making it less susceptible for migration from the cardboard, as transport of the molecules occurs in the gas phase and requires a minimum vapour pressure [7, 14].

3.2. Relationship Breakthrough Values and O_2GTR. The O_2GTR values are given in Table 1 as well as the P_{O_2} normalised to a barrier thickness of 1 μm. The films containing EVOH have very low O_2GTR values, which are two orders of magnitude lower than the PET film, which is also still considered as a good barrier. The PA 6/6.6 has an O_2GTR value significantly higher than that of the other two materials, and this was also reflected in the breakthrough values of

Table 2: Components used in the spike solution and their physical and chemical properties. For each substance, the structure, CAS Registry Number, polarity, partition coefficient for octanol/water (log $P_{o/w}$), molar weight (Mw), boiling point (Bp), and simulant group are given.

	Name	Structure	CAS	Polarity	Log $P_{o/w}$	Mw (Da)	Bp (°C)	Simulant group
C17	*n*-Heptadecane		629-78-7	Nonpolar	9.8	240	302	MOSH
MBP	4-Methyl benzophenone		134-84-9	Polar	3.4	196	326	Photoinitiator
DPP	Di-*n*-propyl phthalate		131-16-8	Polar	3.8	250	340	Plasticiser
ANT	Anthracene		120-12-7	Nonpolar	4.56	178	340	MOAH
PER	Perylene		198-55-0	Nonpolar	5.74	252	494	MOAH
—	Sudan red II		3118-97-6	—	—	—	—	Visual inspection
MTBE	Methyl tert-butyl ether		1634-04-4	—	—	—	55.2	Solvent

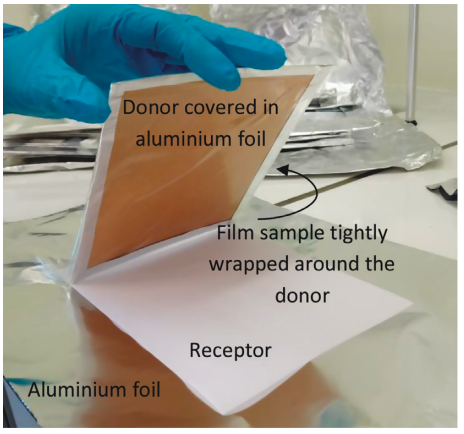


Figure 3: Test pack before being enclosed in aluminium foil.

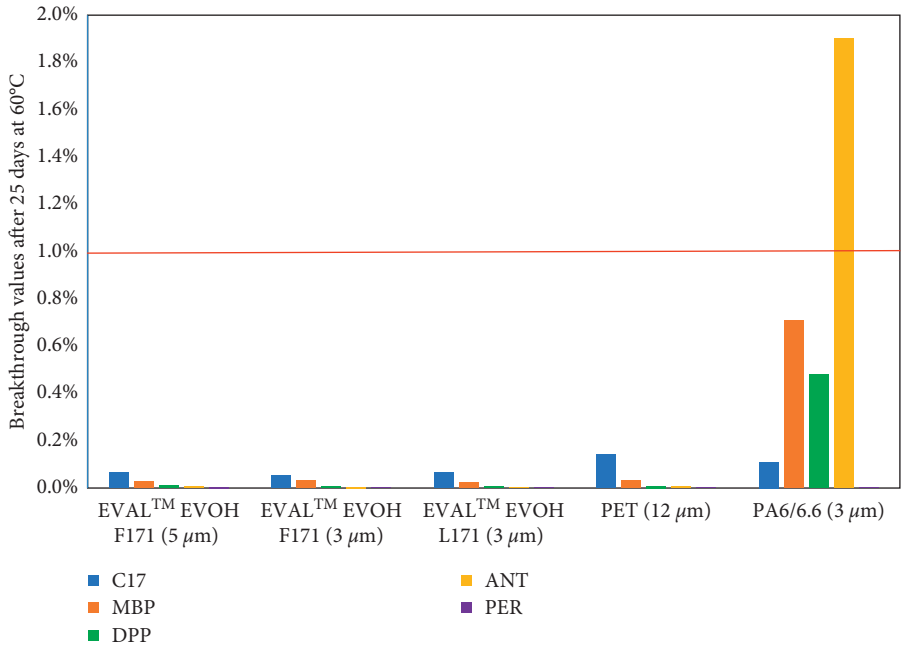


Figure 4: Breakthrough values of *n*-heptadecane (C17), 4-methyl benzophenone (MBP), di-*n*-propyl phthalate (DPP), anthracene (ANT), and perylene (PER) after 25 days at 60°C.

MBP, DPP, and ANT. However, a clear correlation between O_2GTR and the migration of the surrogate components was not visible, as the PA 6/6.6 had similar breakthrough values for C17 as PET and EVOH, which both outperformed the PA 6/6.6 as an oxygen barrier.

4. Conclusion

The results show that both EVAL™ F171B (3 and 5 μm) and L171B (3 μm) have good barrier properties against the surrogate components migrating from recycled paperboard. They can easily compete with PET (12 μm), while clearly outperforming PA 6/6.6 at the same thickness.

When applying the 1% threshold value for breakthrough, both EVOH grades can comfortably protect the food for a shelf life of over 2 years at 25°C against mineral oil migration. Additionally, EVOH also offers good barrier properties against oxygen, nitrogen, carbon dioxide, odours, aroma, flavours, and others, making it a multifunctional solution.

Data Availability

The data used to support the findings of this study are available from the corresponding author upon request.

Conflicts of Interest

This study was done via a collaboration between University Hasselt and Kuraray EVAL Europe, which was partly supported by the agency Flanders Innovation & Entrepreneurship via a VLAIO Baekeland mandate (PhD in collaboration with the industry). The main author was employed at EVAL Europe during her PhD and is affiliated to both University Hasselt and Kuraray EVAL Europe.

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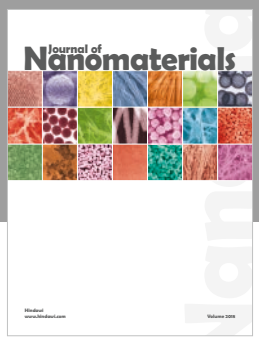

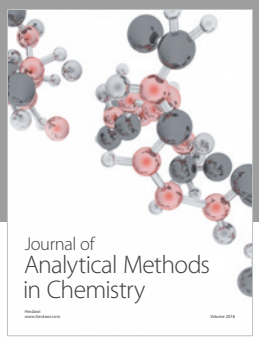

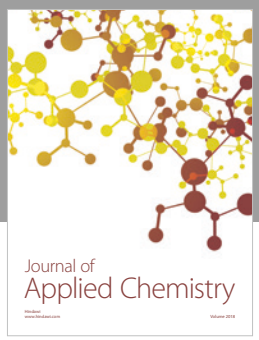

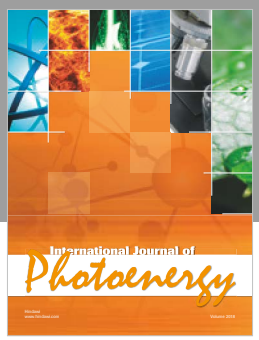

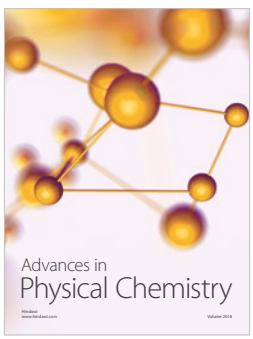

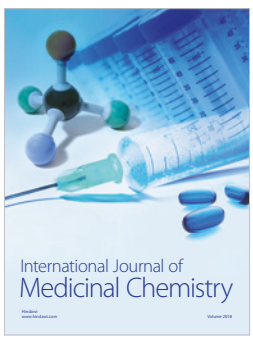

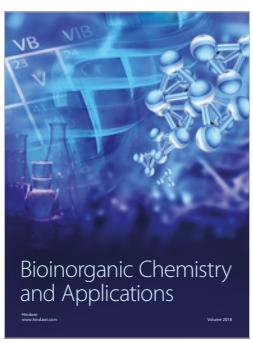

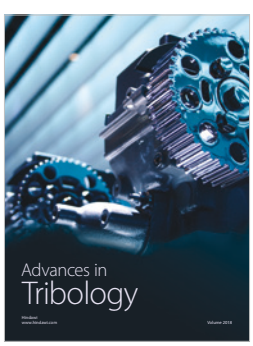

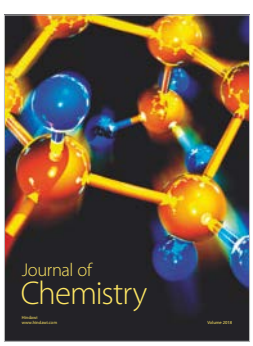

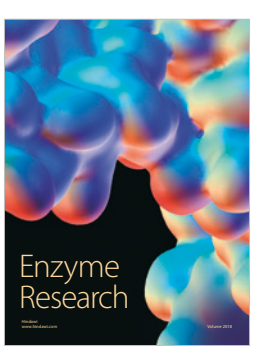

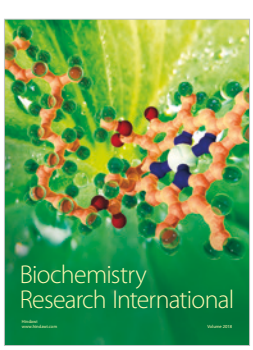

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