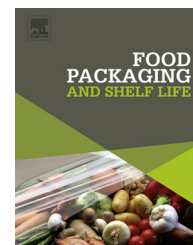


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A study into the potential barrier properties against mineral oils of starch-based coatings on paperboard for food packaging



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ABSTRACT

The aim of this research study is to evaluate the effectiveness of starch-coatings for preventing food contamination from paperboard packaging. Specifically, regarding the migration phenomena of n-alkanes and Mineral Oil Saturated Hydrocarbons (MOSH).

Migration test series were performed under varying conditions (kinetics up to 5 days, at 40 and 60 °C) using spiked model substances (n-alkanes mixture: C₁₀–C₄₀) and Tenax[®] as food simulant. The results obtained for neat paperboard showed that at 40 °C, alkanes from C₁₈ to C₂₆ and at 60 °C from C₁₈ to C₂₈ can migrate almost for 5% of the original spiking. Migration testing of starch-coated paperboard was evaluated, considering the effect of different ingredients and discussed together with the information gained by micro-structural observations. Results for coated paperboards demonstrated significant barrier properties compared to neat materials at the tested conditions. Finally, migration test series with originally contaminated materials were carried out.

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1. Introduction

The European Food Safety Authority (EFSA) published an opinion on mineral oils in food (EFSA CONTAM, 2012), considering available toxicological information and concluding that there is a potential concern associated with their current background exposure. It was stated that migration from paper and board into packed food contributed significantly to the total exposure, due to the presence of recycled pulp and inks. However, it was also highlighted that nearly all

data referred to total Mineral Oil Saturated Hydrocarbons (MOSH) and only little information is currently available on specific sub-classes, such as cyclic, branched or linear alkanes. Moreover, the inclusion of a functional barrier into the packaging assembly was proposed of immediate interest to prevent food contamination.

Several scientific research projects (AGES, 2012; BMELV, 2012; FSA, 2011; IVLV, 2013) and studies (Biedermann et al., 2013; Hauder, Benz, Rüter, & Piringer, 2013; Jung, Simat, Altkofer, & Fügél, 2013; Lorenzini et al., 2013; Zurfluh, Biedermann, & Grob 2013) were already launched and

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Abbreviations: MOSH, Mineral Oil Saturated Hydrocarbons; FB, functional barrier.

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published. The aim was monitoring and characterizing the migration behaviour of mineral oils and other chemicals of concern, from fibre-based materials into simulants or real foodstuffs. Specifically, some studies (Poças, Oliveira, Brandsch, & Hogg, 2011; Zülch & Piringer, 2010) discussed the applicable methodologies for migration modelling. However, in this topic an important gap of scientific knowledge remains for both, the Legislator and the producers. They are currently looking for systematic data which are able to reflect real food packaging systems.

In this situation, barrier materials seem to be the fastest solution for reducing the migration risk. Different private companies have already developed and marketed barrier solutions for mineral oils and simultaneously, some quantitative methods for testing their barrier effect were published in the scientific literature (Ewender, Franz, & Welle, 2012; Fiselier & Grob, 2011).

In 2007, the market of barrier coatings for paper and board was 3778 MD and is expected to climb to 4552 MD by 2014 (PIRA, 2009). Petrol-based polymer materials are already available and mostly used by extrusion-coating techniques or as dispersions onto paper and board. However, the growing request for replacing conventional barriers, such as aluminium and synthetic non-renewable polymers (e.g. polyethylene, polyvinyl alcohol, polyvinylidene chloride), has led to an accelerated development of biopolymers used in food packaging systems. The study of polysaccharides, proteins and lipids for packaging films and coatings is a research field which is rapidly expanding. The benefits of bio-based packaging materials are primarily environmental and related to increased renewability, recyclability and decreased carbon footprint. Other significant advantages are that they could be manufactured from abundant low-cost biomass materials; waste handling costs could thus be decreased. In the past many naturally occurring polysaccharides have been proposed for use in coating and film formulations, including starch, starch derivatives, cellulose derivatives, chitosan, alginates, carrageenan, pectins, mannans, and various naturally occurring gums (Andersson, 2008). Low grease, oxygen, water and water vapour permeability are the most common properties sought after.

Polysaccharides generally possess good film-forming ability, which can lead to high or medium grease and oxygen barrier properties (Hartman, Albertsson, Söderqvist Lindblad, & Sjöberg, 2006; Nisperos-Carriedo & Myrna, 1994). Important properties for a barrier layer include not only low gas permeability, but also good mechanical strength properties and a suitable degree of flexibility (Arvanitoyannis & Biliaderis, 1999). Therefore, a plasticizer is often added to achieve sufficient film flexibility and to facilitate molecular motion and decrease internal frictions within the biopolymer matrix.

Bio-coated paperboards have already been designed to protect and preserve packed food from external influences (Andersson, 2008; Khwaldia, Arab-Tehrany, & Desobry, 2010). However, as regards their effectiveness in preventing food contamination from chemicals, only few data are currently available. In our recent studies (Guazzotti, Tasca, Piergiovanni, & Limbo, 2012), we have evaluated the barrier potential of different bio-coatings onto paper against the migration of a polar (benzophenone) and a non-polar (di-isobutyl phthalate) chemical. Starch-coated paperboard revealed to be the most

promising choice. Other authors (Hartman et al., 2013) tested a hydroxypropylated birch xylan layer towards mineral oil migration.

In the present study the specific aim was to evaluate the barrier properties of starch coated paperboard against migration of n-alkanes and mineral oils. Different type of starches and the presence of sorbitol as plasticizer were tested. Comparative data between migration from neat paperboard, spiked or real samples (originally contaminated material) were additionally obtained and discussed.

2. Materials and methods

2.1. Materials

2.1.1. Paperboard samples

In this study, the substrate used for coating with biopolymers was a 100% virgin kraft grade paperboard, unprinted and suitable for dry food, provided by an Italian industrial supplier. It was one side calendered with a thickness and a grammage respectively of 0.300 mm and 135 g m⁻².

Additionally, to perform migration experiments with real samples (originally contaminated material), a fully coated white lined chipboard with kraft back, made by 95% of recycled fibres with a thickness of 0.425 mm and a grammage of 430 g m⁻² was used. It was obtained from an International packaging company.

The two types of paperboard samples will be identified later on in this paper as sample MO-0080 (the 100% virgin one) and MO-0079 (the one made of 95% recycled fibres).

2.1.2. Solvents and reagents

All the solvents and reagents used in this study were of highest purity available, purchased from Sigma Aldrich-Germany, Restek-USA, Fluka and VWR International Ltd.

Poly 2,6-diphenyl-p-phenylene oxide (60–80 mesh), a standard food simulant for dry fatty foods also known as Tenax[®], was obtained from Grace Alltech Grom GmbH and cleaned up before usage, using an Accelerated Solvent Extractor (ASE) 300 DIONEX. As solvent ethanol and a mixture of hexane/ethyl acetate 56:44 were chosen. Oven temperature was maintained at 110 and 100 °C, respectively for the different solvents, flush time was 120 s, purge time was 100 s, 3 cycles were performed at a pressure of 100 bar. After this cleaning Tenax[®] was collected in an Erlenmeyer flask and allowed to dry in a ventilated oven at 160 °C for 6 h. The method parameters used (polarity of the chosen extraction solvents, temperatures and pressure) were optimized to obtain an efficient extraction. Obtained results were compared with the procedure described in UNI-EN1186-13:2003. Gas chromatograms obtained from the extracts have shown no detectable levels of impurities.

2.1.3. Starches and sorbitol used for coating

Starch coatings were obtained from maize cationic waxy starch (HI-CAT 21370[®], Roquette, Italy) or maize cationic starch (HI-CAT 260[®], Roquette, Italy) or cationic starch mixture with high amylose content based on cereal and tuber starch (HI-CAT 5283A[®], Roquette, Italy). Additionally an

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