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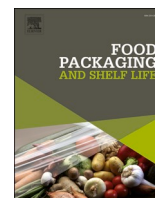
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## Development of a solid food simulant to evaluate migration of chemicals from paper and board food contact materials to moist food

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### ABSTRACT

Food contact materials (FCMs) i.e. materials that food is packaged or handled in, must be safe for their intended use. European FCM legislation uses a risk-based approach, with a cornerstone of FCM's safety evaluation being measurement of migration of substances from FCMs to food simulants. The standard methods mainly developed for plastic FCMs are not always suitable for less inert and moisture sensitive materials such as paper and board. However, these are becoming increasingly common as FCMs e.g. to replace single-use plastics. In addition, there is a drive to further use recycled materials. To support this development, new methods for assessing the safety of these materials are needed. In the present feasibility study, a hydrogel crosslinked through freeze-thawing of poly (vinyl alcohol) was evaluated as a food simulant for moist foods. The migration of surrogate compounds from a spiked paperboard to the hydrogel was determined and compared to the migration to a real moist food (a slice of apple), the commonly used modified polyphenylene oxide (MPPO) and a water extract. Migration of polar surrogates to the hydrogel correlated well with the migration to the apple slice. However, our results indicate that the hydrogel is less suitable as simulant for non-polar surrogates. Overall, the study demonstrates the potential of this hydrogel-based simulant for improving risk assessment of less inert FCMs.

### 1. Introduction

At some point, everything that we eat has been in contact with a material, either during picking, packing, transport, or preparation. These materials are called food contact materials (FCMs) and in Europe FCMs are regulated by a framework regulation, Regulation (EC) No 1935/2004 (European Commission, 2004). The FCM legislation adopts a risk-based approach, unlike for example REACH (Registration, Evaluation, Authorisation and Restriction of Chemicals, (EC) no 1907/2006), which is based on hazard (European commission, 2006). This means that an evaluation of a food contact material considers not only the inherent hazard of a chemical, but the consumer's exposure to that chemical. The exposure is in general assessed by measuring migration from the contact material. Measuring migration from FCM into real food is possible (Bradley et al., 2014; Lorenzini et al., 2013), but it has disadvantages. After the migration, the substance of interest must be extracted from the food; this task will look different with each combination of substance

and food. Another disadvantage is that the food itself is not standardized and might contain background levels of the substance in question or other substances that might interfere with the analysis (Nerín et al., 2022). To standardize the analysis of migration from FCM to food so called food simulants are often used, which are easier to analyse (Baele et al., 2020; Barnkob & Petersen, 2012; Bradley et al., 2014, 2015; Nerín et al., 2022, Urbelis & Cooper, 2021).

Besides the framework regulation for FCMs, Regulation (EC) No 1935/2004, there are specific measures for some materials which define how the framework regulation is to be met within the EU (European Commission, 2004). The most comprehensive specific measure is the one dealing with plastic FCMs, Regulation (EU) no 10/2011 (European commission, 2011). There is no harmonized regulation for paper and board FCMs in the EU; instead, it is common practice to refer to the German recommendation, BfR XXXVI (BfR, 2023). Alike the plastics regulation, this recommendation consists of a positive list of substances allowed to be used in the paper material (BfR, 2023). Some of these

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substances have limitations set for the total content in the material or in extracts (water and/or solvent) of the material. In addition, an increasing number of the listed substances have migration limits (often adopted from the plastic regulation).

Less inert materials such as paper and board have mainly been used in applications for packaging of dry foods, but after the introduction of better barriers and grease-resistant treatments, the applications of paper and board have seen an increased use, for instance, in short term packing applications such as take-away boxes. Furthermore, regulations such as the EU directive on single use plastics, Directive (EU) 2019/904, have resulted in increased use of paper and board as FCM (European commission, 2019). The ongoing drive towards a circular economy require a higher proportion of recycled material in all packaging materials, also in FCM, as long as safety can be demonstrated. The use of recycled material as a component increases the complexity of the starting material for the FCM, such as for instance residue mineral oil. Taken together these developments increases the need for methods to measure migration of paper and board FCM to be able to perform accurate risk-assessment of them (Biedermann-Brem et al., 2016; de Fátima Poças et al., 2011, Lorenzini et al., 2013).

The migration from paper and board is today measured mainly using modified polyphenylene oxide (MPPO or Tenax), as a simulant to mimic dry foods (EN 14438:2004). MPPO (food simulant E) is also used for the same purpose in Regulation (EU) No 10/2011 (European commission, 2011). To evaluate contact with moist food, water extraction is used instead (BfR, 2023). For instance, water has been found to be a suitable food simulant to detect biocides in paper and board (Elizalde et al., 2023). However, immersing paper and board in liquids, results in wet extraction, which can be a total or partial extraction depending on which substance that is considered, rather than a measure of migration from the board into the food simulant (Störmer et al., 2024). Dry boards can be described as a multi component system often having an inner core that carries the main part of the migrant (for instance when recycled material is used) and the thinner outer layer from which the migrant desorbs of the surface, but in a wet immersion system the difference between the layers is lost and the board can be seen as a homogenous system (Zülch & Piringer, 2010). Wet extraction of paper and board can therefore also include contributions from the inner core or printing inks on the opposite side of a paperboard, not intended to be in direct contact with food.

The aim of the present study was to create a solid food simulant with a high-water content. A hydrogel, formed by a polymeric network restricting and entrapping the water was therefore tested herein as a food simulant for moist food. This was investigated by measuring migration from a spiked substrate to the gel and comparing with the migration to an apple slice, MPPO and the amounts detected in a water extract.

## 2. Material and methods

### 2.1. Preparation of hydrogels through freeze-thawing

Poly(vinyl alcohol) (PVA) with a molecular weight of 146–186 kDa, 99% hydrolysed, was purchased from Sigma Aldrich (363065, Merck, USA). The high degree of hydrolysis and high molecular weight was selected to maximise the number of hydroxyl groups responsible for the H-bonding to strengthen the network and ensure gelation with only one freeze-thawing cycle. The hydrogels were prepared by mixing a 10% wt. solution of PVA with boiling tap water, then left to cool to 70 °C. Each solution was made by mixing 108 g of water into a Pyrex flask with 12 g of PVA. The PVA-water mixture was stirred on a magnetic heating plate for 15 min, whilst heating at 70 °C. The flasks were wrapped in aluminium foil and placed in an oven at 110 °C for 150 min. The PVA solution was once again placed at the heating plate whilst stirring for 30 min. The PVA solution was poured into round aluminium weighing pans (611–1380, VWR, International) 20 g in each. The moulds were covered

by parafilm and placed in a freezer for 15 h at  $-20$  °C,  $-40$  °C and  $-80$  °C. The frozen PVA gels were left to thaw in ambient conditions. The freeze thawing process resulted in physically crosslinked PVA gels with diameter of 6.5 cm and a thickness of approximately 0.5 cm. The gels were stored in their aluminium pans at 4 °C prior to analysis.

### 2.2. Water activity

The water activity ( $a_w$ ) of an apple slice and the PVA hydrogel prepared at  $-40$  °C were measured using a hygrometer by comparing to water and active coal (Aqualabs, Addium Inc, USA).

### 2.3. Microscopy

The microstructures of the hydrogels prepared at different freezing temperatures and the microstructure of an apple slice were analysed using confocal laser scanning microscopy (CSLM) (Leica TCS SP5, Leica Ltd, Germany), with a 100x/1.4PL APO oil objective. The samples were taken from the bottom of the cast hydrogels and were stained using acridine orange. The light source was an Argon laser using  $\lambda_{\text{ex}} = 488$  nm, and the signal was emitted in the interval 500–600 nm.

### 2.4. Surrogates

Four compounds were selected as surrogates to measure potential migration, see Table 1. Initially two compounds (benzophenone and dibutyl phthalate) with migration limits in the BfR XXXVI recommendation (BfR, 2023) was selected. Acetophenone and n-Heptadecane were added to the list of surrogates to widen the range of molecular weight, boiling point and partition coefficient (Log P, Table 1).

### 2.5. Substrate

A multilayer paperboard with a Cobb 60 value of 30 and grammage of 345 g/m<sup>2</sup> was selected as the substrate for this study. The paperboard was divided into pieces with an area of 0.5 dm<sup>2</sup> (7 × 7 cm). The substrate was spiked with a mix of the surrogates with a concentration of 0.15 or 0.25 mg/L of each surrogate in acetone. The substrates were immersed in the mix for 15 min and then allowed to dry in a fume hood in room temperature for at least 15 min before being used in experiments. The amount of surrogate in the substrate was determined by extracting the paperboard, cut in pieces, with 25 mL of dichloromethane (DCM) through gentle shaking at room temperature for 24 h. A sample of the DCM extract was then used for GC-MS analysis. Triplicate samples were analysed for each migration experiment.

### 2.6. Migration studies

#### 2.6.1. Comparison of migration to the PVA hydrogel, apple slice and MPPO

The PVA hydrogels produced at  $-40$  °C, apple slices and MPPO were placed on spiked substrates (0.5 dm<sup>2</sup>, spiked in a mix of 0,25 mg/L) in glass Petri dishes. Organic apples bought in a supermarket were divided into slices 4–5 mm thick, with the peel remaining. Both the hydrogels and the apple slices covered an area of 0.33 dm<sup>2</sup> of the substrate. Two

**Table 1**  
Properties of surrogates.

Surrogate	CAS	Molecular weight <sup>a</sup> (g/mol)	Boiling point (°C) <sup>a</sup>	Log P octanol/ water <sup>a</sup>
Acetophenone	98–86–2	120	202	1.5–1.9
Benzophenone	119-61-9	182	305	2.9–3.5
n-Heptadecane	629-78-7	240	302	9.2–9.5
Dibutyl phthalate	84-74-2	278	340	4.5–5.0

<sup>a</sup> Data retrieved from CAS Scifinder database 2023.

grams of MPPO (Tenax TA (refined), 60–80 mesh, Supelco), were spread over an approximately equal area of the substrates. The Petri dishes with the samples were wrapped in aluminium foil and placed at the selected migration parameters: 2 h at 40 °C. Migration experiments were performed in triplicate. After the migration, the hydrogels were divided into eight equally sized pieces and extracted with hexane (50 mL) with gentle shaking at room temperature for 24 h. A range of solvents (acetone, acetonitrile, dichloromethane, ethylacetate and methyl tert butyl ether) and extraction parameters (different extraction times with and without ultra sonic extraction) were evaluated in the optimisation of the extraction protocol. The apple slices were extracted following a published protocol (Bradley et al., 2014). Briefly, the apple slice was homogenized with a mixer and extracted with 30 mL of DCM overnight. The solvent was decanted off and the apple mash was washed with an additional portion of DCM. The portions of solvent were pooled and adjusted to a final volume of 50 mL. MPPO was extracted according to EN 14338:2004 using two portions of acetone (final volume 25 mL) (EN 14338:2004). All extractions were repeated twice with a new portion of solvent. The amount of the surrogates in the extracts was determined by GC-MS analysis.

### 2.6.2. Comparison of migration to the PVA hydrogel and a cold-water extract

Substrates were spiked with a surrogate mix of 0,15 mg/L of each surrogate and used to measure migration to PVA hydrogels. Cold-water extracts according to the standard EN 645:1994 was performed on the same substrates (EN 645:1994). The migration parameters and extraction of gels and reference substrates were performed as above. Migrations and extracts were performed in triplicates. SPE extraction for the determination of acetophenone, benzophenone and dibutyl phthalate in water by GC-MS was performed according to the procedures outlined in EN-ISO 18856:2004 (EN ISO 18856:2004). HyperSep™ C18 cartridge (1000 mg, 6 mL, Thermo Scientific, TN, USA) was employed for SPE process. The SPE cartridges were conditioned sequentially with ethyl acetate and methanol. Subsequently, 10 mL of the water extract was loaded onto the SPE cartridge at a flow rate of 2–5 mL min<sup>-1</sup> using positive pressure. Elution was carried out with ethyl acetate containing an internal standard. The amount of heptadecane was determined after liquid/liquid extraction with dichloromethane. Spiked water extracts were processed the same way and used to calculate recovery rates. Laboratory blanks were processed with Milli-Q water following the same procedure as the hydrogel water extract to assess potential background contamination.

### 2.6.3. Migration to the hydrogels prepared at different temperatures

The migration to the hydrogels after the freeze thawing process at temperatures of –20 °C, –40 °C and –80 °C were compared. The prepared hydrogels were placed on spiked substrates in glass Petri dishes. The Petri dish was wrapped in aluminium foil and kept at the selected migration parameters: 2 h at 40 °C. The migration tests were performed in triplicate. After the migration, the hydrogels were divided in eight equally sized pieces and extracted with hexane (50 mL) for 24 h through gentle shaking at room temperature. The amount of the surrogates in the hydrogels was determined using GC-MS analysis.

### 2.7. Gas chromatography mass-spectrometry analysis (GC-MS) and quantification

Separation and determination of compounds was performed on an Agilent 8890 GC coupled to a LECO BT EI time of flight mass spectrometer (LECO, USA). The chromatographic separations were carried out with a 30 m × 0.25 mm × 0.25 µm DB-5MS UI capillary column (Agilent J&W GC columns, USA). The GC operating conditions were as follows: injector temperature 280 °C (splitless mode) oven temperature was held at 50 °C for 1 min, then heated to 280 °C at 10 °C/min and kept at this temperature for 15 min. The carrier gas was helium at a constant

flow rate of 1.0 mL/min. The mass spectrometer was scanned from *m/z* 30 to 550; the ionization was performed by electronic impact and the ion trap temperature was 250 °C, and the electron multiplier voltage was 70 V. The compounds were identified based on total ion spectra and retention time and quantified.

The amount of the surrogates in the solvent extracts was quantified based on *m/z* (acetophenone: 105, benzophenone: 105, heptadecane: 43, 57, 73 and dibutyl phthalate: 149) relative to a calibration curve made from solvent extracts with known amounts of these surrogates and an internal standard (Bicyclohexyl, CAS 92–51-3). Migrating amounts were calculated in mg/dm<sup>2</sup> based on the surface area of the substrates (for amounts before migration and cold-water extracts) or the area of the substrate that was covered by the hydrogel, apple slice or MPPO. Amounts detected in the first and second extraction of the hydrogels, apple slices and MPPO after migration were combined and any low amounts detected in the blank samples were subtracted. Amounts detected in water extracts were corrected for recovery rates based on spiked water extracts (acetophenone 90%, benzophenone 103%, dibutyl phthalate 78% and heptadecane 35%) and any low amounts detected in the blank sample were subtracted.

## 3. Results and discussion

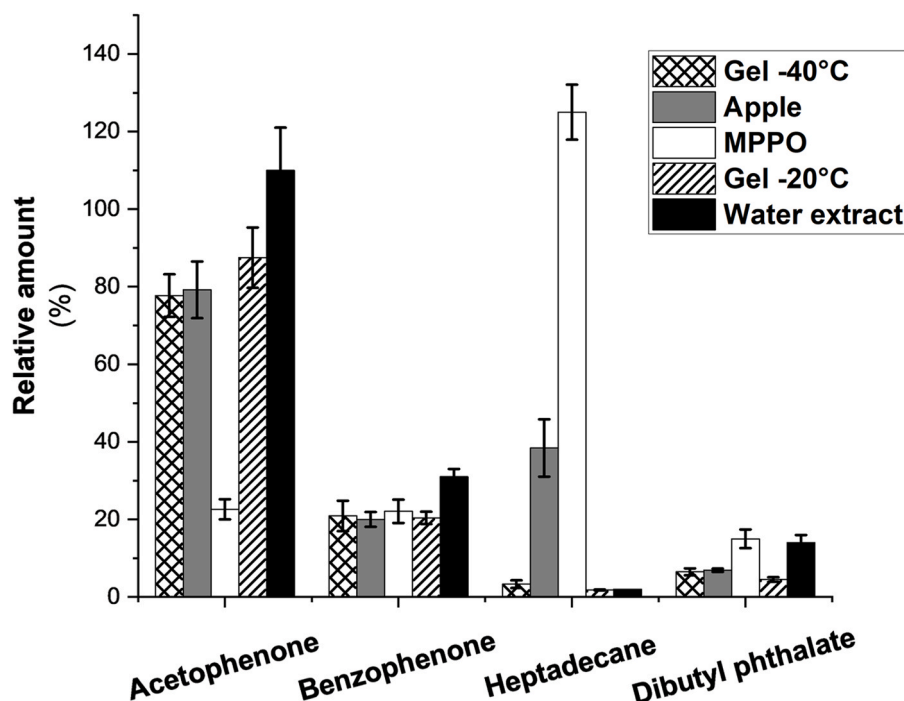
### 3.1. Evaluation of the hydrogels as food simulants for moist food

The synthetic polymer poly(vinyl alcohol) (PVA) was dissolved in water and a hydrogel prepared through one cycle of freezing and thawing of the PVA suspension. Freezing and thawing of the PVA suspension causes physical entanglement of the polymeric chains through the creation of junction points during freezing which remains interlocked after thawing, creating a network structure; this preparation technique is also known as a cryogel (Adelnia et al., 2022; Wan et al., 2014). Freeze thawing PVA is a well-known technique within biomedical applications to create hydrogels without the addition of chemical crosslinking agents (Adelnia et al., 2022; Wan et al., 2014). To minimize the cycles of freezing and thawing and maximise the network strength, a highly hydrolysed PVA with a high molecular weight was selected following previous research (Adelnia et al., 2022). This study hypothesized that these hydrogels can be used as food simulants for moist food such as salad or fresh cut fruit due to their large water content. The aim was to obtain a matching water activity (*a<sub>w</sub>*) to fresh cut fruit to give an indication of the amount of unbound or “free water” in the matrix that controls, among other things, the ability to wet the packaging material (Prabhakar & Mallika, 2014). The *a<sub>w</sub>* of the PVA hydrogel was found to be 0.99 and the apple slice 0.94. It has previously been reported that the water content of a typical apple slice is 85.9%, with an *a<sub>w</sub>* of 0.99 (Kahraman et al., 2021), which is also in the range of other reference values of *a<sub>w</sub>* for fresh fruits (0.95–0.97) (Prabhakar & Mallika, 2014). The water content of the hydrogels used herein was 90 wt%.

To evaluate if the migration to the PVA hydrogels resembled that of moist foods, the migration from spiked substrates were compared to that of an apple slice and MPPO in one experiment and to a cold water extract in another (Fig. 1). The migration parameters utilised in this study (2 h at 40 °C) were chosen based on the intended application of the paper and board. A paper and board for moist food is likely to be used for serving rather than storing food, hence the short contact time of 2 h. MPPO is an accepted simulant for assessing migration at high temperatures (European commission, 2011), the aim of this study was to develop a simulant for mimicking food contact at room temperature during a short time, hence the choice of 40 °C.

Under these test parameters similar migrating amounts were detected to the hydrogel and the apple slice for acetophenone, benzophenone and dibutyl phthalate, whereas the migration of heptadecane was much lower to the hydrogel (Fig. 1). In contrast, the migration to the food simulant MPPO was observed to correlate well with the migration to the apple slice only for benzophenone, while it overestimated the migration





**Fig. 1.** Relative migrating amounts ( $\text{mg}/\text{dm}^2$ ) in the hydrogel compared to an apple, a water extract and the food simulant MPPO compared to the amounts detected in the reference substrates. Error bars represent standard deviation between triplicate samples. The amount detected in the substrates (in  $\text{mg}/\text{dm}^2$ ) before migration were as follows: acetophenone  $0.63 \pm 0.07$ , benzophenone  $0.93 \pm 0.06$ , heptadecane  $0.68 \pm 0.05$  and dibutyl phthalate  $1.12 \pm 0.06$  (for experiment with hydrogel  $-40^\circ\text{C}$ , apple and MPPO) and acetophenone  $0.30 \pm 0.04$ , benzophenone  $0.56 \pm 0.07$ , heptadecane  $0.76 \pm 0.01$  and dibutyl phthalate  $0.65 \pm 0.12$  (for experiment with hydrogel  $-20^\circ\text{C}$  and water extract).

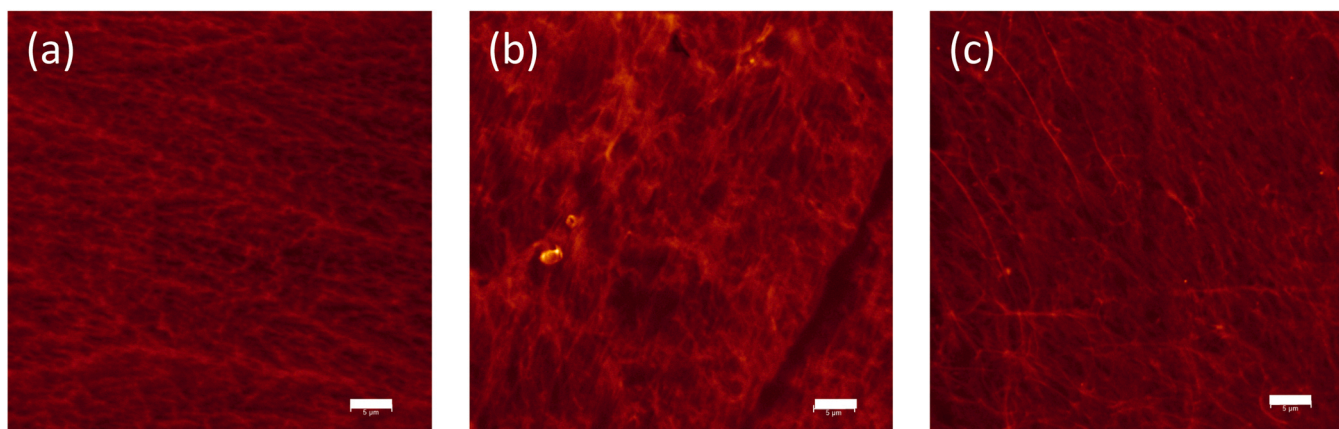
of dibutyl phthalate and heptadecane but underestimated the migration of acetophenone. The amounts detected in the cold-water extract were higher than the migration to the hydrogel for acetophenone, benzophenone and dibutyl phthalate while the amount of heptadecane was similar (Fig. 1). Hence, the water extract slightly overestimated the amounts compared to the migration to the apple for acetophenone and benzophenone and, just as the hydrogel, underestimated the amount of heptadecane that migrated to the apple (Fig. 1). The amount of dibutyl phthalate detected in the water extract was similar to the amount that migrated to MPPO (14 vs. 15% of the amount detected in the spiked substrate, Fig. 1). Overall, this indicates that the hydrogel is a better simulant for moist food than MPPO for polar compounds and can be a good complement to a cold-water extract when one sided migration is of interest. The hydrogel could for instance be used for multilayer materials that contain recycled material or when printing has been done on

the side not intended for contact with food.

### 3.2. Evaluation of hydrogels produced at different temperatures

To assess the impact of freezing temperatures on the microstructure of the produced PVA hydrogel, confocal laser scanning microscopy (CLSM) was conducted after the different freezing temperatures of  $-20^\circ\text{C}$ ,  $-40^\circ\text{C}$  and  $-80^\circ\text{C}$  (Fig. 2).

The CLSM micrographs of the hydrogels showed a very fine-stranded network and an effect of the different freezing temperatures (Fig. 2). The freezing temperature regulates the rate of freezing and a low freezing temperature means that the sample spends less time in temperature interval below zero where ice crystals are formed and vice versa. It was shown that the hydrogels prepared at the highest temperature,  $-20^\circ\text{C}$  (Fig. 2(a)) had a finer network with a more aligned structure compared



**Fig. 2.** Confocal laser scanning microscopy (CLSM) of the microstructure of the PVA hydrogels after preparation at different freezing temperatures, (a)  $-20^\circ\text{C}$ , (b)  $-40^\circ\text{C}$  and (c)  $-80^\circ\text{C}$ . All scale bars are  $5\ \mu\text{m}$ .

to the others. The aligned structure could be due to propagation of the ice front in combination with the long growth time of the ice crystals. As the temperature was reduced, the network structures were less aligned (Fig. 2b and c) due to the decreased time allowed for the growth of ice crystals. The junction points in the PVA network are reported to be formed by the proximity of the polymeric chains during freezing (Adelnia et al., 2022), when the hydrogel is thawing the chains are further entangled and a physical network is formed.

In contrast, the micrographs of the apple tissue (Fig. 3) show that the pore structure of the apple is considerably larger than the pore size of the hydrogels (Fig. 2). Note that the scale bars for the micrographs of the apple are 100  $\mu\text{m}$  compared to 5  $\mu\text{m}$  for the hydrogels (Figs. 2 and 3). In addition, it can be observed that the porosity of the apple varies more than that of the hydrogels. The apple has a cellular structure, where the tissue is built up by strong cell walls compared to the fine stranded network of the hydrogel. The microstructures shown here resemble micrographs found in the literature on apple tissue (Kahraman et al., 2021). Whilst apples have a high water activity it is also composed of a higher % of dry weight (44–53% for immature apples) (Stevenson et al., 2005), which could contribute to the difference in migration of the non-polar substance (heptadecane).

To assess if the migration to the hydrogels was affected by the microstructural differences observed (Fig. 2) between PVA hydrogels produced at  $-20\text{ }^{\circ}\text{C}$ ,  $-40\text{ }^{\circ}\text{C}$  and  $-80\text{ }^{\circ}\text{C}$  they were evaluated in a migration experiment (Fig. 4). The migration of the surrogates from the spiked substrate was observed to be similar irrespective of the hydrogel freezing temperature. This indicates that the change in microstructure observed in Fig. 2 by freezing the hydrogels at different temperature did not alter the migration from the substrate.

### 3.3. Surrogate properties and their migration

In general, the migration of the surrogates from the substrates to the gels followed their polarity (indicated by Log P, Table 1). The highest migration was observed for the most polar compound acetophenone, even though it was present in a lower amount in the substrate before the migration. The next highest migration was observed for benzophenone followed by dibutyl phthalate and heptadecane (Fig. 4). The same trend was observed for the amounts detected in the water extract (Fig. 1).

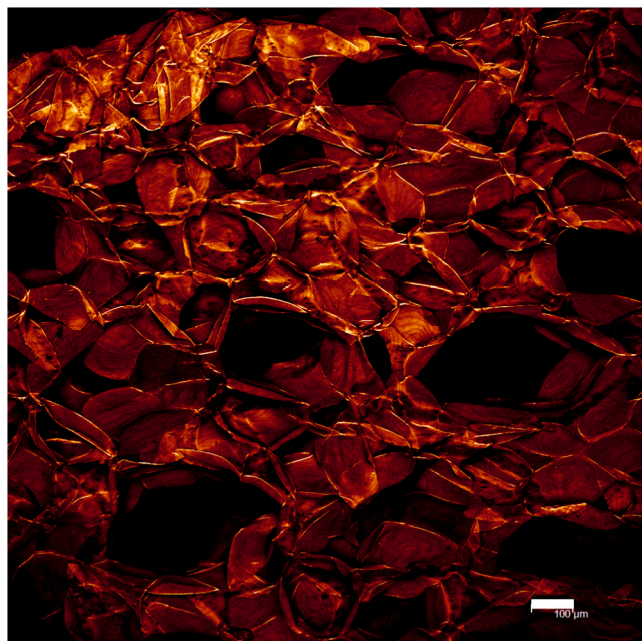


Fig. 3. Confocal laser scanning microscopy (CSLM) of the microstructure of the apple. The scale bars are equal to 100  $\mu\text{m}$ .

Similarly, Bradley et al. attributed the migration results to different fruits and vegetables observed in their study to polarity, apart from the extent of indirect and direct contact between the substrate and the different food types included in their study (Bradley et al., 2014), factors that can be excluded in our study since we only compared to one food type. It has previously been reported that polar compounds may migrate to a larger extent with increasing humidity (Urbelis & Cooper, 2021). The hydrogels, which resemble foods with high  $a_w$ , would increase the humidity at the surface of, or in, the paperboard, which could alter the migration by enhancing capillary flow as has previously been reported for benzophenone (Barnkob & Petersen, 2012). In this study, the hydrogel wet the surface but did not wet the substrate further than the boundaries of the gel during the 2 h migration study. The contact of the wet surface to the paper board may aid the migration of polar components. It is proposed that the polarity of the migrants can play a role due to its interaction with the surface of the paper fibre (Elizalde et al., 2020).

To the best of our knowledge few previous studies have been published on comparison between cold water extracts, migration to MPPO and migration to food. A study by Elizalde et al. has found that a cold-water extract performs better in respect to MPPO when it comes to evaluation of board containing biocides in close contact with vegetables (Elizalde et al., 2023). Migration from paper and board to MPPO or dry food has previously been described to be mainly affected by molecular weight and volatility (Urbelis & Cooper, 2021). In general, medium-size migrants has the highest migration values since the most volatile compounds are lost through evaporation, while migrants with a high molecular weight have lower diffusion coefficient (Urbelis & Cooper, 2021). This correlates well with the high migration observed of heptadecane to MPPO in this study.

For acetophenone, the migration to MPPO was four times lower than that of the apple. Bradley et al. previously observed an overestimation of the migration of acetophenone to MPPO compared to mushrooms, apples and potatoes for three out of four substrates investigated in their study (Bradley et al., 2014). However, direct comparisons are not feasible due to their use of whole apples with intact peel as well as different migration parameters. Our results indicate that care must be taken before using MPPO to measure migration of acetophenone. Further studies are needed to determine if this also includes other volatile polar compounds.

While the hydrogel could be used as a simulant for the more polar substances investigated in this study, the results show that it is a poor simulant for heptadecane. The migration to the hydrogel was less than one tenth of the migration to the apple of heptadecane (1,8–3,3% vs. 38,4% of the amount in the substrate, Fig. 1). In addition, the same underestimation of the migration (as seen with the hydrogel) of heptadecane was observed with the water extract (2% of the amount in the substrate, Fig. 1). In contrast, the migration to MPPO overestimated the migration of heptadecane more than three times (Fig. 1). The hydrogel consists of 10% dry weight of a linear polymer where 99% of all non-polar side groups have been hydrolysed and is therefore very polar, not able to retain any non-polar substances in the fine stranded network. The apple slice on the other hand has a more heterogenous composition, a higher dry weight with rigid cell walls containing starch and cellulose which may be able to retain non-polar components.

The difference in migration between the hydrogel, water extract, MPPO and the apple for heptadecane indicates that further studies are needed to develop food simulants suitable for measuring non-polar substances.

### 3.4. Conclusion

Our results show that PVA hydrogels can potentially be used as simulants to mimic high moisture foods like fresh-cut fruit products or salads. Most importantly, it can be used to measure one-sided migration from non-inert, water sensitive materials, such as paper and board. This

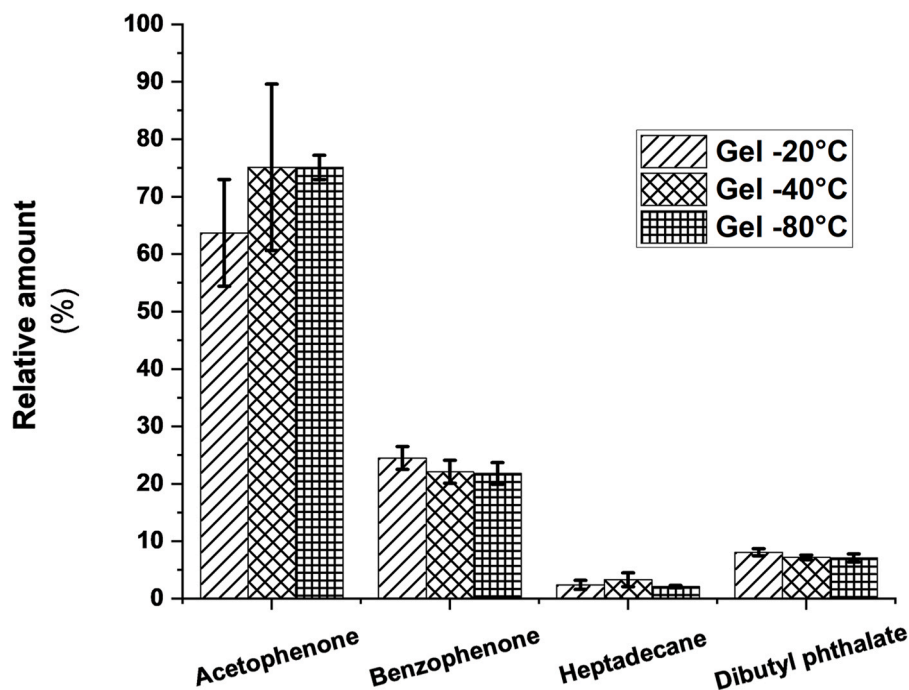


Fig. 4. Relative migrating amounts ( $\text{mg}/\text{dm}^2$ ) in hydrogels produced at different freezing temperatures compared to the amounts detected in the reference substrates. Error bars represent standard deviation between triplicate samples. The amount detected in the substrates (in  $\text{mg}/\text{dm}^2$ ) before migration were as follows: acetophenone  $0.43 \pm 0.07$ , benzophenone  $0.88 \pm 0.01$ , heptadecane  $0.84 \pm 0.04$  and dibutyl phthalate:  $1.04 \pm 0.02$ .

may be particularly valuable for food contact articles made of several layers, for instance with recycled material in the core. Further development of the gels is needed to be able to measure migration of non-polar substances. The PVA-hydrogels are easily prepared and robust, i. e. the migration was shown not to be affected by preparation conditions. In this feasibility study we evaluated four surrogate compounds and compared the migration to one food type under one test condition, more experiments with a variety of food types and test conditions are needed to define the limits and full potential of this potential novel food simulant. In addition, future experiments are needed to cross validate the results with other labs.

This new food simulant can help to fill the gap in available standard methods to measure migration from paper and board FCM and thereby improving the risk assessment of these material as a complement to wet extracts. This is particularly important as paper and board are increasingly used as FCM. For instance, the directive on single use plastics and the expanded possibilities to use paper and board FCM with more types of food following development of barriers and grease-resistance treatment enhance this development. Furthermore, the push for a circular economy will mean an increased use of recycled material in FCM. This will also increase the complexity of risk assessment and risk management and further increase demand for novel methods to measure migration.

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#### CRedit authorship contribution statement

**Astrid Ahlinder:** Writing – review & editing, Writing – original draft, Validation, Methodology, Investigation, Conceptualization. **Jenny Lindh:** Writing – review & editing, Writing – original draft,

Validation, Project administration, Methodology, Investigation, Funding acquisition, Conceptualization. **Mats Stading:** Writing – review & editing, Conceptualization. **Susanna Andersson:** Writing – review & editing, Project administration, Funding acquisition, Conceptualization. **Camilla Öhgren:** Writing – original draft, Investigation, Formal analysis. **Hans Steijer:** Writing – review & editing, Conceptualization.

#### Declaration of Competing Interest

None.

#### Data Availability

Data will be made available on request.

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#### Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at [doi:10.1016/j.fpsl.2024.101340](https://doi.org/10.1016/j.fpsl.2024.101340).

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