


# Food and beverage can coatings: A review on chemical analysis, migration, and risk assessment

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## Abstract

The internal surface of food and beverage cans is generally covered with polymeric coatings to preserve food and protect metal substrate from corrosion. Coating materials are complex formulations that contain different starting substances (e.g., monomers, prepolymers, additives, etc.) and in addition during the manufacture of the material several compounds can be formed (e.g., reaction products, degradation products, etc.). These substances have the potential to migrate into the food. Many of them have not been identified and only some have been toxicologically evaluated. This article aims to provide a comprehensive review on the analytical methods used for the identification of potential migrants in can coatings. The migration and exposure to chemicals migrating from can coatings are also reviewed and discussed so far, which is essential for risk assessment. Moreover, a brief section on the current status of the legislation on varnishes and coatings for food contact in Europe is also presented. Liquid chromatography coupled to diode array and fluorescence detectors and particularly to mass spectrometry and gas chromatography–tandem mass spectrometry seem to be the techniques of choice for the identification of potential migrants in can coatings. Some studies have reported migration levels of BPA (bisphenol A) and BADGE (bisphenol A diglycidyl ether) and derivatives exceeding the specific migration limits set in the European legislation. On the whole, low dietary exposure to migrants from can coatings has been reported. However, it is interesting to highlight that in these studies the combined exposure to multiple chemicals has not been considered.

## KEYWORDS

analytical techniques, exposure assessment, food metal cans, migration, polymeric coatings

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## 1 | INTRODUCTION

Metal food packaging includes a wide variety of products such as cans, closures, lids, tubes, and aerosol containers among others. Cans are by far the most widespread, being canned foods and beverages an important part of the global food supply (LaKind, 2013; Oldring & Nehring, 2007).

The canning of food and beverages is still an important process in food preservation since foods packed in metallic cans have a longer shelf life compared to food packed in other materials.

Polymeric coatings are often applied in the inner surface of food metal cans acting as a barrier between food and the metal surface. They play an essential role in the preservation of food maintaining its quality as well as protecting the metal substrate from corrosion (Cooper & Tice, 2001; Driffield et al., 2018).

Epoxy resins based on bisphenol A diglycidyl ether (BADGE) have been widely used for the inner coating of metal cans because of their excellent mechanical properties and chemical resistance. Although they are still being used, lately due to uncertainties about the potential effects on human health of the endocrine disrupting properties of bisphenol A (BPA), other coatings based on different chemistries are being employed (Driffield et al., 2018). Some examples of these types of resins include polyester, acrylic, and so on.

During the coating manufacture, as a result of an incomplete polymerization process, residual prepolymers and monomers can remain in the final product and are able to migrate to the food. In addition to intentionally added substances, such as prepolymers, monomers, additives, catalysts, and so on, the final material may also contain the so-called nonintentionally added substances (NIAS), such as impurities, reaction products, degradation products, and so on. Thus, a large number of substances can be present in the final materials and have the potential to migrate into the foodstuffs (Aznar et al., 2015; Kato & Conte-Junior, 2021; Nerin et al., 2022). Migration is a mass transfer phenomenon where the substance transferred from the packaging to the food is called migrant. The potential migrants need to be identified to assure the safety of the material. Most of these compounds are unknown; therefore, they have not been evaluated toxicologically, and some of them are highly reactive chemical species (Grob et al., 1999; Paseiro-Cerrato et al., 2017). The analysis of these potential migrants is an extremely difficult task for various reasons, being one the fact that frequently there is not enough information about the composition of the material. On the other hand, the manufacturing process is usually quite complex with several steps and, in many cases is not completely known (Hoppe et al., 2016). Liq-

uid chromatography and gas chromatography coupled to mass spectrometry, particularly to high-resolution mass spectrometry (HRMS), have shown to be powerful tools to accomplish this challenging task (Canellas et al., 2019; Nerin et al., 2013). The strengths and limitations of the analytical techniques used in the analysis of migrants from food contact materials (FCMs) are summarized in a recent review (Nerin et al., 2022).

Hence, to assess the safety of these materials, a number of issues need to be addressed, namely, the identification of potential migrants in the final materials, the determination of their migration in food and beverages, and the risk evaluations through food exposure assessments (Figure 1). The extent of migration and the specific toxicity of a given substance (s) are the two key factors that define the risk to human health that an FCM represents (Poças & Hogg, 2007).

From a food legislation standpoint, varnishes and coatings are one example of a type of FCM for which there is currently no specific harmonized measure at European level; nevertheless, as it is the case for all FCMs, varnishes and coatings must meet the requirements of Article 3 of the Framework Regulation (EC) No. 1935/2004 (European Union, 2004) regarding the inertness criteria as well as the safety requirements for the consumer. On the other hand, national regulations should be followed and CoE (Council of Europe) resolution be used as reference.

This review addresses different aspects of the coatings for food cans. First, a brief overview on the coatings used for food and beverages cans is presented. In the following sections, a compilation of the analytical methods for the identification of potential migrants in polymeric coatings is provided. Next, the migration of chemicals from the can coatings into food as well as the dietary exposure assessment are discussed. Finally, the regulatory aspects of coatings for food contact are commented on.

## 2 | COATINGS FOR FOOD AND BEVERAGE CANS

Coatings can be applied in both the internal and external side of metal cans intended to contain food products and beverages. This review is limited to the coatings used on the food contact side.

Coatings are formed by application of coating materials by different process, generally by either roller coating or spraying before the curing step. The coating application can be performed both on the metal sheet before forming the container and once the object is formed. In most cases, they have to be able to withstand mechanical deformations to which they are subjected. It is quite common to use

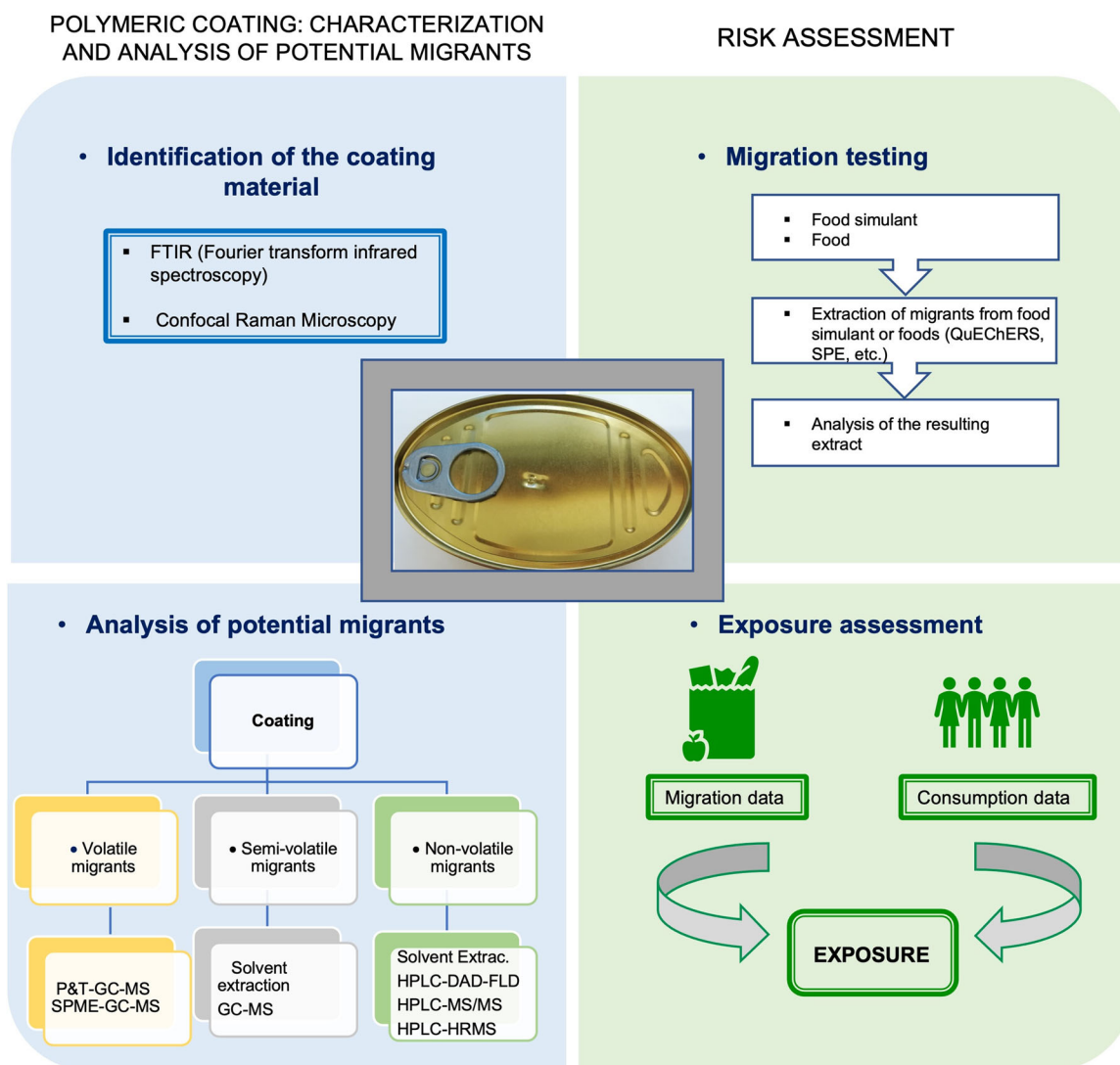


FIGURE 1 Schematic representation of an analytical approach for the chemical safety assessment of food-contact coatings

more than one type of coating material or applying a coating material several times (Oldring & Nehring, 2007). The main components present in can coatings include resins, cross-linking agents, additives, and solvents; the latter are not always present (Oldring & Nehring, 2007). Some examples of compounds that can be present in polymeric can coatings are shown in Table 1.

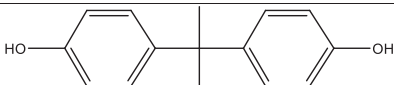

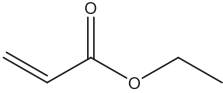
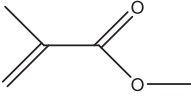
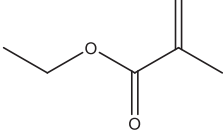
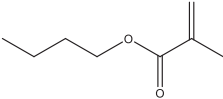
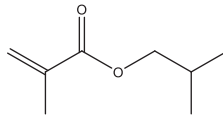

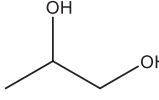
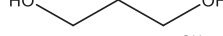
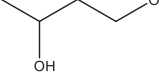
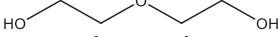

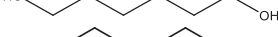


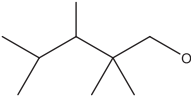
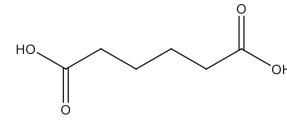
For food contact applications in metal cans, the most commonly resins used include epoxy, phenolic, polyester, acrylic, vinyl and oleoresins, and variations of them (LaKind, 2013; Oldring & Nehring, 2007). There is no single type of resin that serves for all foods, each one is suitable for a certain type of food. Thus, for example, epoxy acrylic resins cross-linked with an amino resin are commonly used for beverage cans; white aluminum-pigmented epoxy resins are suited for fruits and vegetables; and phenolic and epoxy-phenolic blends containing aluminum pigment are widely employed for sulfur-bearing fish and meat (LaKind,

2013; Robertson, 2012; Oldring & Nehring, 2007). A summary of the main chemical and mechanical properties, toxicity, and uses of the coatings used in metal cans is presented in Table 2.

## 2.1 | Oleoresins

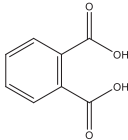
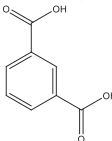
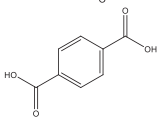
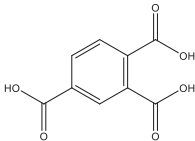
Initially, before the introduction of synthetic resins, oleoresins were extensively used to coat the food contact surface of metal cans. These resins of natural origin are obtained from natural gums and rosins, and then are blended with drying oils such as linseed or tung oil. The oleoresinous-based coatings present a poor adhesion to metal surfaces and require long times of curing. With the exception of specific applications, particularly nonaggressive foods (e.g., dried beans), nowadays their use is very limited due to their poor resistance to corrosion. Besides, it

TABLE 1 Examples of compounds that could be present in polymeric can coatings

Resin	Compound	Chemical Formula	Chemical Structure*	Molecular Weight	CAS Number
Epoxy	Bisphenol A (BPA)	C <sub>15</sub> H <sub>16</sub> O <sub>2</sub>		228.2910	80-05-7
Epoxy	Bisphenol A diglycidyl ether (BADGE)	C <sub>21</sub> H <sub>24</sub> O <sub>4</sub>		340.4190	1675-54-3
Acrylic	Ethyl acrylate	C <sub>5</sub> H <sub>8</sub> O <sub>2</sub>		100.1170	140-88-5
Acrylic	Methacrylic acid methyl ester (MMA)	C <sub>5</sub> H <sub>8</sub> O <sub>2</sub>		100.1170	80-62-6
Acrylic	Methacrylic acid ethyl ester (EMA)	C <sub>6</sub> H <sub>10</sub> O <sub>2</sub>		114.1440	97-63-2
Acrylic	Methacrylic acid butyl ester (BMA)	C <sub>8</sub> H <sub>14</sub> O <sub>2</sub>		142.1980	97-88-1
Acrylic	Methacrylic acid isobutyl ester (iBMA)	C <sub>8</sub> H <sub>14</sub> O <sub>2</sub>		142.1980	97-86-9
Polyester	Ethylene glycol (EG)	C <sub>2</sub> H <sub>6</sub> O <sub>2</sub>		62.0680	107-21-1
Polyester	1,2-Propylene glycol (PG)	C <sub>3</sub> H <sub>8</sub> O <sub>2</sub>		76.0950	57-55-6
Polyester	1,3-Propylene glycol (PG)	C <sub>3</sub> H <sub>8</sub> O <sub>2</sub>		76.0950	504-63-2
Polyester	1,3-Butanediol (BD)	C <sub>4</sub> H <sub>10</sub> O <sub>2</sub>		90.1220	107-88-0
Polyester	Diethylene glycol (DEG)	C <sub>4</sub> H <sub>10</sub> O <sub>3</sub>		106.1210	111-46-6
Polyester	Neopentyl glycol (NPG)	C <sub>5</sub> H <sub>12</sub> O <sub>2</sub>		104.1490	126-30-7
Polyester	1,6-Hexanediol (HD)	C <sub>6</sub> H <sub>14</sub> O <sub>2</sub>		118.1760	629-11-8
Polyester	Tris(hydroxymethyl)propane (HMP)	C <sub>6</sub> H <sub>14</sub> O <sub>3</sub>		134.1750	77-99-6
Polyester	Cyclohexyldimethanol (CHDM)	C <sub>8</sub> H <sub>16</sub> O <sub>2</sub>		144.2140	105-08-8
Polyester	2,2,4-Trimethylpentane-1,3-diol (TMP)	C <sub>8</sub> H <sub>18</sub> O <sub>2</sub>		146.2300	144-19-4
Polyester	Adipic acid (AA)	C <sub>6</sub> H <sub>10</sub> O <sub>4</sub>		146.1420	124-04-9

(Continued)

TABLE 1 (Continued)

Resin	Compound	Chemical Formula	Chemical Structure*	Molecular Weight	CAS Number
Polyester	Phthalic acid (PA)	C <sub>8</sub> H <sub>6</sub> O <sub>4</sub>		166.1320	88-99-3
Polyester	Isophthalic acid (IPA)	C <sub>8</sub> H <sub>6</sub> O <sub>4</sub>		166.1320	121-91-5
Polyester	Terephthalic acid (TPA)	C <sub>8</sub> H <sub>6</sub> O <sub>4</sub>		166.1320	100-21-0
Polyester	Trimellitic acid (TMA)	C <sub>9</sub> H <sub>6</sub> O <sub>6</sub>		210.1410	528-44-9

\*ChemDraw Professional.

has been also reported that they may alter the organoleptic properties of food (LaKind, 2013; Oldring & Nehring, 2007; Robertson, 2012).

## 2.2 | Epoxy-based coatings

Epoxy resins are obtained through a condensation reaction between BPA and epichlorhydrin to yield BADGE. BADGE is the lowest molecular weight resin. These resins are cured with different curing agents mainly amino resins, phenolic resins, and anhydrides, to form a highly cross-linked three-dimensional network in order to provide chemical and pack resistance necessary for the manufacture of food and beverage cans (Oldring & Nehring, 2007; Schaefer & Simat, 2004). Furthermore, BADGE can be also added as stabilizer for organosols acting as a scavenger for hydrochloric acid (El Moussawi et al., 2019; Schaefer & Simat, 2004). However, currently BADGE is not authorized as additive.

Because of their good mechanical properties, high adhesiveness, and chemical resistance, these thermosetting resins are widely employed in internal coatings for metal cans for food and beverages. Moreover, they do not impart flavor to foods (LaKind, 2013).

## 2.3 | Vinyl-based coatings

The vinyl-based coatings are commonly mixed with other type of resins such as epoxy and phenolic in order to

improve their properties. With the exception of vinyl organosol coatings, generally, they are not applied directly on the metal surface because of their limited adhesion to metal substrates (LaKind, 2013).

Vinyl organosols are manufactured by mixing several types of resins including poly(vinyl chloride) (PVC), epoxy, and phenolic and epoxy-phenolic resins (Cottier et al., 1998). PVC is produced by polymerization of vinyl chloride monomer (PVM). The production of PVM involves the reaction between chlorine and ethylene to yield ethylene dichloride, then the dichloride is dehydrochlorinated to give the monomer (Robertson, 2012). Organosols present very good flexibility and pack resistance (Oldring & Nehring, 2007). They can be found in two-piece cans (e.g., canned fish) (LaKind, 2013).

## 2.4 | Polyester-based coatings

Due to the uncertainties regarding the potential adverse effects associated to BPA, polyester-based coatings are being increasingly used as a promising alternative to epoxy resins.

Polyesters are synthesized by a condensation reaction between polycarboxylic acids and polyols (Pietropaolo et al., 2018; Zhang et al., 2018; Ubeda et al., 2020). Different monomers are employed in their manufacture; some of the more widely used carboxylic acids include phthalic acid, terephthalic acid, isophthalic acid, adipic acid, and trimellitic acid; and among the polyols, some

TABLE 2 Coatings properties, toxicity uses, and applications

Coating	Chemical and mechanical properties	Toxicity	Uses and applications	References
Oleoresins	Flexibility: good Adhesion to metal surfaces: poor Corrosion resistance: limited	Limited information on toxicity is available. Studies have in general reported low toxicity of polymers obtained from natural oils.	Limited uses. Fruits drinks, fruits, and vegetables.	LaKind (2013) Geueke (2016) Sharma and Kundu (2006)
Epoxy	Flexibility: good Adhesion to metal surfaces: good Corrosion resistance: good	BPA: endocrine disruptor; studies shown that it presents reproductive toxicity, genotoxicity, cytotoxicity, mutagenicity, and carcinogenicity. BADGEs: In vitro assays revealed that show endocrine disrupting properties and also exhibit genotoxicity and mutagenicity and cytotoxicity. In vivo assays have shown that BADGEs present adverse effects in the reproductive and endocrine systems.	Wide range of foods and beverages (e.g., meats, fish, juices, etc.).	LaKind (2013) Geueke (2016) Tarafdar et al. (2022) Wang et al. (2021)
Vinyl based resins	Vinyl Flexibility: very good Adhesion to metal surfaces: very poor Corrosion resistance: poor Vinyl organosol Flexibility: very good Adhesion to metal surfaces: good (higher than vinyl) Corrosion resistance: good (higher than vinyl)	Organosol resins are manufactured mixing several types of resins including PVC, epoxy, and phenolic and epoxy-phenolic resins. The toxicity is related to the toxicity of epoxy resins (BPA, BADGE).	Soft drinks (vinyl). Fish (vinyl organosol).	LaKind (2013) Geueke (2016) Oldring and Nehring (2007)

(Continues)

TABLE 2 (Continued)

Coating	Chemical and mechanical properties	Toxicity	Uses and applications	References
Polyester	Flexibility: very good Adhesion to metal surfaces: good Corrosion resistance: poor	Polyester can coatings extracts have been reported to present cytotoxicity.	Nonacid foods.	LaKind (2013) Geueke (2016) Oldring and Nehring (2007) Groh and Muncke (2017)
Phenolic	Flexibility: poor Adhesion to metal surfaces: poor Corrosion resistance: very good	Components of resole have been reported to exhibit estrogenic activity.	Phenolic resins are used as crosslinkers with other resins. Their main applications are canned fish and meat.	LaKind (2013) Geueke (2016) Biedermann and Grob (2006b)
Acrylic	Flexibility: poor Adhesion to metal surfaces: good Corrosion resistance: suitable	Studies have shown that the monomer ethylacrylate used in the manufacture of acrylic resins is nongenotoxic.	Acrylic resins are commonly used as external coatings. When they are used in the food contact surface, the main applications are beverage cans.	LaKind (2013) Geueke (2016) Forrest (2005) Bolognesi et al. (2017)



of the most frequent are ethylene glycol, 1,2-propanediol, 1,3-propanediol, 1,3-butanediol, 2-methyl-1,3-propanediol, neopentyl glycol, 1,6-hexanediol, 2,2,4-trimethylpentane-1,3-diol, trimethylolpropane, and cyclohexyldimethanol (Bradley et al., 2009; Schaefer et al., 2004; Aznar et al., 2019; Ubeda, Aznar, Alfaro et al., 2019; Canellas et al., 2022). This type of resins can be cross-linked with amino resins, phenolic resins, or polyisocyanates (Oldring & Nehring, 2007). In past years, polyester-polyurethane coatings are being developed; these coatings contain in their formulations polyisocyanates to aid the cross-linking of the polymer network during the curing process, resulting in a more inert coating with lower migration characteristics (Bradley et al., 2009).

From the mechanical properties' standpoint, polyester resins exhibit very good flexibility (Oldring & Nehring, 2007). In terms of food contact applications, the polyester-based coatings are not suitable for aggressive and acidic foods since they are sensitive to hydrolytic attack of the ester bond under low-pH conditions (Oldring & Nehring, 2007; LaKind, 2013).

## 2.5 | Phenolic-based coatings

Phenolic resins (called also resoles) are very complex mixtures derived from the reaction products of phenols (phenol, cresol, *tert*-butylphenol) with formaldehyde and low-molecular-weight aliphatic alcohols such as butanol. They are commonly used as cross-linker in combination with other resins such as epoxy resins (Biedermann & Grob, 2006a, 2006b; Oldring & Nehring, 2007). These coatings are often used in food cans and are resistant to aggressive foodstuffs (Oldring & Nehring, 2007). They have good acid and sulfur resistance, but they impart flavor to some foods (Robertson, 2012).

## 2.6 | Acrylic based coatings

In FCMs, acrylic polymers have been used in different applications such as adhesives for food packaging (Nerín et al., 2009; Canellas et al., 2012; Canellas et al., 2017), kitchenware, and coatings on different substrates (Franz & Brandsch, 2013). These types of coatings result from the polymerization of acrylic acid and its derivatives (Piergiovanni & Limbo, 2016). The main monomer used for the production of acrylic resins is ethylacrylate. These coatings are commonly applied in the external surface (LaKind, 2013); however, these coatings also have applications in the easy open ends and bodies of cans intended to contain beverages (Forrest, 2005). These

materials offer heat stability (Piergiovanni & Limbo, 2016).

## 3 | ANALYTICAL METHODS FOR THE DETERMINATION OF MIGRANTS OF POLYMERIC COATINGS FOR FOOD CONTACT

### 3.1 | Determination of migrants in polymeric coatings

The polymeric coatings used in the inner surface of metal cans for food and beverages may contain in their formulations several components such as resins, cross-linking agents, additives including lubricants, wetting agents, and so on, and solvents. Besides these starting substances, other compounds such as oligomers, reaction products, or unreacted compounds resulting from an incomplete polymerization process may be present in the final product and, therefore, have the potential to migrate from the can coating into the packed food (Bradley et al., 2008; El Moussawi et al., 2019).

The identification and quantification of these potential migrants, many times new and unknown, can be extremely difficult and challenging due, among other reasons, to the complexity of the formulations and the lack of commercial standards allowing confirmation.

Chromatographic techniques coupled to different detectors and especially to mass spectrometers have been generally used to achieve this analytical task. Table 3 contains a compilation of analytical methods with their main characteristics for the determination of starting substances, oligomers, and reaction products in polymeric coatings used in metal food and beverage cans.

Some representative examples of analytical methods used for the determination of migrants from different coatings are given next. Both the earliest and the most recent methodologies are discussed. Conventional techniques such as HPLC and GC coupled to different detectors (UV, FLD, FID, etc.) have been commonly used in the analysis of the components of the coatings. Currently, MS technologies coupled to chromatographic techniques or the application of a battery of analytical techniques are the main approaches proposed to characterize the coatings. However, despite the powerful analytical techniques that are being used it is not possible to identify all detected compounds. The limited information about the formulation composition and the manufacturing process makes the identification of unknown migrants an extremely difficult task (Nerín et al., 2022; Hoppe et al., 2016; Bradley et al., 2008).



TABLE 3 Analytical methods used for the identification of potential migrants in polymeric can coatings

Coatings	Potential migrants	Analytical technique detection system	Chromatographic conditions	Sensitivity/validation	Reference
Epoxy	BADGE; BADGE·H <sub>2</sub> O; BADGE·2H <sub>2</sub> O; BADGE·HCl; BADGE·2HCl; BADGE·H <sub>2</sub> OHCl	HPLC–FLD ( $\lambda_{\text{ex}}$ 275 nm, $\lambda_{\text{em}}$ 305 nm) DryLab <sup>®</sup> software	Poroshell 120 SB-C18 (100 × 4.6 mm, 2.7 $\mu\text{m}$ ) column. Mobile phase: ACN and water. Elution mode: gradient Flow: 1.5 ml/min Injection vol.: 10 $\mu\text{l}$ Column temp.: 30°C	Linearity: $R^2 \geq .9995$ LOD: 0.01–0.20 ng/g LOQ: 0.03–0.66 ng/g Recovery: 91.65%–100.76% Reproducibility: RSD < 7.02%	Guo et al. (2020)
Epoxy and phenoxy	BADGE; BADGE·H <sub>2</sub> O; BADGE·2H <sub>2</sub> O; BADGE·HCl; BADGE·2HCl; BADGE·H <sub>2</sub> OHCl; CYDBADGE; BPA; BPB; BPC; BPE; BPF; BPG	HPLC–FLD ( $\lambda_{\text{ex}}$ 225 nm, $\lambda_{\text{em}}$ 305 nm)	Phenosphere 80 Å ODS (150 × 3.2 mm, 3 $\mu\text{m}$ ) column. Mobile phase: water type I and a mixture of ACN:MeOH (50:50, v/v). Elution mode: gradient Flow: 0.5 ml/min Injection vol.: 10 $\mu\text{l}$ Column temp.: 30°C	Linearity: $R^2 \geq .9994$ LOD: 0.005 mg/L LOQ: 0.0125 mg/L Within-day repeatability: RSD < 5%	Lestido-Cardama, Vázquez-Loureiro et al. (2021)
Epoxy	BADGE; BADGE·H <sub>2</sub> O; BADGE·2H <sub>2</sub> O; BADGE·HCl; BADGE·HCl·H <sub>2</sub> O; BFDGE; BFDGE·2HCl, BFDGE·2H <sub>2</sub> O; BADGE·2HCl	LC–MS/MS (ESI+) Parent ion [M+NH <sub>4</sub> ] <sup>+</sup> : BADGE 358.2; BADGE·H <sub>2</sub> O 376.2; BADGE·2H <sub>2</sub> O 394.2; BADGE·HCl 394.2; BADGE·HCl·H <sub>2</sub> O 412.1; BFDGE 330.2; BFDGE·2HCl 402.1; BFDGE·2H <sub>2</sub> O 366.2; BADGE·2HCl 430.1	Synergy MAX-RP (100 × 2.0 mm, 2.5 $\mu\text{m}$ ) column. Mobile phase: 0.01 M ammonium formate solution and MeOH. Elution mode: gradient Flow: 0.2 ml/min Column temp.: 40°C	Linearity: $R^2 > .999$ LOD: 0.5–3.1 $\mu\text{g}/\text{kg}$ LOQ: 1.8–10.3 $\mu\text{g}/\text{kg}$ Recovery: 89.92%–109.51% Intraday repeatability: RSD < 9.5% Interday repeatability: RSD < 12.0%	Miguez et al. (2012)

(Continues)

TABLE 3 (Continued)

Coatings	Potential migrants	Analytical technique detection system	Chromatographic conditions	Sensitivity/validation Reference
Epoxy (epoxy phenolic, epoxy anhydride, water-based modified epoxy spray lacquer)	BADGE; BADGE·BuOH; BADGE·PrOH; BADGE·tBuPhe; DiBADGE; BADGE·BuEtOH/tBuPhe; BADGE·2tBuPhe; Cyclo-Di-BADGE; Tri-BADGE	HPLC-FLD ( $\lambda_{\text{ex}}$ 225 nm, $\lambda_{\text{em}}$ 295 nm) GC-MS (Scan mode)	HPLC-FLD: YMC-Pack CN (250 × 4.6 mm, 5 $\mu\text{m}$ , 120 Å) column with precolumn (Silica, 10 × 4.6 mm). Mobile phase: ( <i>n</i> -propanol/pentane 2/98 [v/v]) and ( <i>n</i> -propanol/MTBE 50/50 [v/v]). Elution mode: gradient Flow: 1 ml/min Injection vol.: 250 $\mu\text{l}$ Column temp.: 40°C GC-MS: PS-255 (8 m × 0.25 mm, 0.2 $\mu\text{m}$ ) column with a precolumn (1.5 m × 0.53 mm, deactivated by coating with a 0.15- $\mu\text{m}$ layer of OV-1701). Temp. program: 80–350°C. Injection mode: on column (100–150 $\mu\text{l}$ ) Syringe: 250 $\mu\text{l}$ Injection rate: 1 $\mu\text{l/s}$	LOD: 4.5 kg/kg (BADGE) LOQ: 9 kg/kg (BADGE) Theobald et al. (2002)
Epoxy	BADGE; BADGE·H <sub>2</sub> O; BADGE·2H <sub>2</sub> O	HPLC-FLD ( $\lambda_{\text{ex}}$ 225 nm, $\lambda_{\text{em}}$ 305 nm) UV (225-nm (for second confirmation))	Spherisorb ODS 2 (150 × 4.6 mm, 5 $\mu\text{m}$ ) column. Mobile phase: ACN and water. Elution mode 1: gradient Elution mode 2: isocratic ACN–water (65:35 v/v) Flow: 1.0 ml/min Injection vol.: 50 $\mu\text{l}$	Linearity: $R^2 > .997$ LOD: 1.5 $\mu\text{g/L}$ (BADGE) Precision: RSD < 3% Paseiro-Losada et al. (1997)

(Continues)

TABLE 3 (Continued)

Coatings	Potential migrants	Analytical technique detection system	Chromatographic conditions	Sensitivity/validation	Reference
Epoxy	BADGE; <i>m</i> -XDA	HPLC-FLD ( $\lambda_{\text{ex}}$ 275 nm, $\lambda_{\text{em}}$ 300 nm) (BADGE) ( $\lambda_{\text{ex}}$ 395 nm, $\lambda_{\text{em}}$ 480 nm) (derivatized <i>m</i> -XDA with fluorescamine)	Pecosphere CRT C <sub>18</sub> RC (150 × 5 mm, 5 $\mu$ m) column. BADGE analysis Mobile phase: ACN and water. Elution mode: gradient. Derivatized <i>m</i> -XDA Mobile phase: phosphate buffer (1/15 M, pH 8)–water–MeOH (16:34:50) Elution mode: isocratic Flow: 1.0 ml/min Injection vol.: 50 $\mu$ l	Linearity: $R^2 \geq .9990$ LOD: 5 $\mu$ g/L (BADGE); 20 $\mu$ g/L ( <i>m</i> -XDA) Recovery: 98.9% $\pm$ 6.5% (BADGE); 89.3% $\pm$ 6.0% ( <i>m</i> -XDA) Precision: RSD < 3.4% (BADGE); RSD < 5.7% ( <i>m</i> -XDA)	Paseiro-Losada et al. (1991)
Epoxy-phenolic	BADGE; BADGE-HCl; BADGE-2HCl; BFDGE ( <i>o,o'</i> , <i>o, p'</i> , <i>p,p'</i> ); BADGE-H <sub>2</sub> O; BADGE-HCl-H <sub>2</sub> O; BADGE-2H <sub>2</sub> O	HPLC-FLD ( $\lambda_{\text{ex}}$ 275 nm, $\lambda_{\text{em}}$ 305 nm)	Betasil C-18 (250 × 4.6 mm, 5 $\mu$ m, 100 Å pore size) column. Mobile phase: water and ACN. Elution mode: gradient Flow: 1.0 ml/min Injection vol.: 10 $\mu$ l Column temp.: 30°C	Linearity: $R^2 > .997$ LOD: 0.004–0.028 mg/L LOQ: 0.013–0.093 mg/L Recovery: 78.9%–128% Repeatability: RSD 0.6%–8.7%	Safta et al. (2018)
BPA-based epoxy	BPS; BADGE-2H <sub>2</sub> O; BPF; BPA; BADGE-HCl-H <sub>2</sub> O; BADGE-H <sub>2</sub> O; BADGE; BADGE-HCl; BADGE-2HCl	LC-MS/MS (ESI+, ESI-) Precursor ( <i>m/z</i> ): BPS 249 [M-H] <sup>-</sup> ; BADGE-2H <sub>2</sub> O 394 [M+NH <sub>4</sub> ] <sup>+</sup> ; BPF 199 [M-H] <sup>-</sup> ; BPA 227 [M-H] <sup>-</sup> ; BADGE-HCl-H <sub>2</sub> O; 412 [M+NH <sub>4</sub> ] <sup>+</sup> ; BADGE-H <sub>2</sub> O 376 [M+NH <sub>4</sub> ] <sup>+</sup> ; BADGE 358 [M+NH <sub>4</sub> ] <sup>+</sup> ; BADGE-HCl 394 [M+NH <sub>4</sub> ] <sup>+</sup> ; BADGE-2HCl 430 [M+NH <sub>4</sub> ] <sup>+</sup>	OTU TriKala C <sub>18</sub> (250 × 3 mm, 5 $\mu$ m) column connected to a C <sub>18</sub> guard column (4 × 3 mm). Mobile phase: water ACN and MeOH. Elution mode: gradient Flow: 500 $\mu$ l/min Injection vol.: 25 $\mu$ l Column temp.: 25°C	Linearity: $R^2 \geq .9989$ Recovery: 75%–118%	Zech et al. (2015)

(Continues)

TABLE 3 (Continued)

Coatings	Potential migrants	Analytical technique detection system	Chromatographic conditions	Sensitivity/validation Reference	
Epoxy	BPA; BPF; BPZ; 2,2'-BP	GC-MS; Target ions ( <i>m/z</i> ): BP 186; BPF 200; BPA 213; BPZ 268	HP-5MS (30 m × 0.25 mm i.d., 0.25 μm) capillary column. Temp. program: 75–275°C. Ion source, transfer line and quadrupole temp.: 230, 300, and 150°C Carrier gas: Helium Flow: 1 ml/min Injection mode: Splitless	Linearity: $R^2 > .99$ LOD: 0.9–2.5 ng/L LOQ: 3.1–8.4 ng/L Intraday repeatability: RSD < 3.59% Interday repeatability: RSD < 5.80% Recovery: 86%–122%	Cacho et al. (2012)
Epoxy and vinyllic organosols	BPA; BPF; BADGE; BFDGE	GC-MS; Selected ions ( <i>m/z</i> ): BPA 228; BPF 200; BADGE 325; BFDGE 312 (for confirmation 207)	DB 5-MS (30 m × 0.32 mm i.d., 0.25 μm) fused silica capillary column. Temp. program: 150–270°C. Injector 260°C and detector 300°C Carrier gas: Helium Flow: 1.1 ml/min	LOD: 0.15–0.86 μg/dm <sup>2</sup> LOQ: 0.51–2.77 μg/dm <sup>2</sup> Repeatability: RSD < 5.0%	Jordáková et al. (2003)
Epoxy	BADGE; BADGE·HCl; BADGE·2HCl; BADGE·H <sub>2</sub> O; BADGE·H <sub>2</sub> O·HCl; BADGE·2H <sub>2</sub> O	RP-HPLC-APCI-MS Full-scan mode Range <i>m/z</i> 100–500 (APCI+) Select ions (+ <i>m/z</i> ) BADGE; 382 [M+CH <sub>3</sub> CNH] <sup>+</sup> BADGE·HCl; 418 [M+CH <sub>3</sub> CNH] <sup>+</sup> BADGE·2HCl; 382 (unknown fragment) BADGE·H <sub>2</sub> O; 400 [M+CH <sub>3</sub> CNH] <sup>+</sup> (APCI-) Select ions (- <i>m/z</i> ) BADGE·H <sub>2</sub> O·HCl; 393 [M-H] <sup>-</sup> BADGE·2H <sub>2</sub> O 375 [M-H] <sup>-</sup>	Kromasil 100 C <sub>18</sub> (15 cm × 0.4 cm, 5 μm) column. Mobile phase: ACN-water (30:70, v/v) Elution mode: gradient Flow: 1.0 ml/min Injection vol.: 50 μl	Linearity: $R^2 \geq .9902$ LOD: 0.05–0.4 mg/L Precision: RSD < 5.0%	Sendón García and Paseiro-Losada (2004)

(Continues)

TABLE 3 (Continued)

Coatings	Potential migrants	Analytical technique detection system	Chromatographic conditions	Sensitivity/validation	Reference
Epoxy and phenoxy	BPF; BADGE·2H <sub>2</sub> O; BPE; BPA; BPB; BADGE·H <sub>2</sub> O; BADGE·H <sub>2</sub> O·HCl; BPC; BADGE; BADGE·HCl; BADGE·2HCl; BPG; CYDBADGE	LC-MS/MS (APCI-) Parent ion [M-H] <sup>-</sup> BPF 198.9; BADGE·2H <sub>2</sub> O 374.8; BPE 212.9; BPA 226.9; BPB 240.9; BPC 254.9; BPG 311.0. BADGE·H <sub>2</sub> O·HCl 283.0 (unknown fragment) LC-MS/MS (APCI+) Parent ion [M+H] <sup>+</sup> CYDBADGE 569.0. [M+CH <sub>3</sub> CNH] <sup>+</sup> BADGE 381.9; BADGE·H <sub>2</sub> O 399.9; BADGE·HCl 417.9 BADGE·2HCl 382.2 (unknown fragment)	Phenosphere 80 Å ODS (150 × 3.2 mm, 3 μm) column with a precolumn. Mobile phase: water and a mixture of ACN:MeOH (50:50, v/v). Elution mode: gradient Flow: 0.5 ml/min Injection vol.: 10 μl Column temp.: 30°C	LOD: 0.5–500 μg/L	Lestido-Cardama et al. (2019)
Epoxy	BADGE·2H <sub>2</sub> O; BADGE·H <sub>2</sub> O; BADGE·HCl·H <sub>2</sub> O; BADGE; BADGE·HCl; BADGE·2HCl; BFDGE·2H <sub>2</sub> O; BFDGE; BFDGE·2HCl	LC-MS/MS heated-electrospray ionization (H-ESI+) Precursor ion (m/z) [M+NH <sub>4</sub> ] <sup>+</sup> : BADGE·2H <sub>2</sub> O 394.2; BADGE·H <sub>2</sub> O 376.2; BADGE·HCl·H <sub>2</sub> O; 412.2; BADGE 358.2; BADGE·HCl 394.2; BADGE·2HCl 430.2; BFDGE·2H <sub>2</sub> O 366.2; BFDGE 330.2; BFDGE·2HCl 402.1	Fused Core™ Ascentis Express C18 (150 × 2.1 mm, 2.7 μm) column. Mobile phase: MeOH and 25 mM formic acid–ammonium formate buffer (pH 3.75) Elution mode: gradient Flow: 600 μl/min Injection vol.: 10 μl Column temp.: 50°C	Linearity: R <sup>2</sup> ≥ .999 LOD: 0.15–8 μg/kg LOQ: 0.5–2.5 μg/kg Run-to-run precision: RSD < 10% Recovery: 70%–95%	Gallart-Ayala et al. (2011a)

(Continues)

TABLE 3 (Continued)

Coatings	Potential migrants	Analytical technique detection system	Chromatographic conditions	Sensitivity/validation Reference
Epoxy phenolic	BADGE; BADGE·H <sub>2</sub> O; BADGE·HCl; BADGE·H <sub>2</sub> O·HCl; BADGE·2H <sub>2</sub> O; BADGE·2HCl; BADGE·H <sub>2</sub> O·EtOEtOH; Cyclic compound from carnauba wax; BADGE·H <sub>2</sub> O·BuOEtOH; BADGE·2EtOEtOH; BADGE·H <sub>2</sub> O·BuOH; BADGE·BuOEtOH; BADGE·H <sub>2</sub> O·PrOH; linear or cyclic BADGE·BPA; BADGE·EtOEtOH·BuOH; BADGE·H <sub>2</sub> O·BPA; BADGE·MeOEtOH·BuOH; BADGE·MeOMeEtOPrOH; Polyoxyethylene (6) decyl ether; BADGE·EtOEtOH·BuOEtOH BADGE·BuOEtOH·HCl; BADGE·EtOEtOH·BuOH; BADGE·EtOEtOH; BADGE ( <i>n</i> = 2)·H <sub>2</sub> O·BuOEtOH; BADGE·2BuOEtOH; BADGE ( <i>n</i> = 2)·MeOEtOH; BADGE·BuOH· BuOEtOH; BADGE·BPA·BuOEtOH; BADGE·HexOH; BADGE ( <i>n</i> = 2) ·2BuOEtOH	HPLC–FLD ( $\lambda_{\text{ex}}$ 275 nm, $\lambda_{\text{em}}$ 305 nm) LC–TOF–MS (ESI+) (ESI–) Mass range 100–1100 <i>m/z</i> .	HPLC–FLD Sunfire C18 (150 × 4.6 mm, 5 $\mu\text{m}$ ) column. Mobile phase: water and ACN. Elution mode: gradient Flow: 1.0 ml/min Injection vol.: 20 $\mu\text{l}$ LC–TOF–MS Agilent ZORBAX Eclipse XDB–C18 (100 × 2.1 mm, 3.5 $\mu\text{m}$ column) For (ESI+) Mobile phase: 0.1% aqueous acetic acid and ACN. For (ESI–) Mobile phase: 5 mM ammonium formate at pH 5.5 and 0.1% 5 mM ammonium formate at pH 5.5 in ACN. Elution mode: gradient Flow: 0.2 ml/min Injection vol.: 5 $\mu\text{l}$	Bradley et al. (2008)

(Continues)



TABLE 3 (Continued)

Coatings	Potential migrants	Analytical technique detection system	Chromatographic conditions	Sensitivity/validation Reference
Epoxy and organosols	BADGE; Dimer; cyclo-Dimer; Trimer; Tetramer; Pentamer; Hexamer; Heptamer; <i>o,o</i> -BFDGE; <i>p,p</i> -BFDGE; <i>o,p</i> -BFDGE; 3-Ring NOGE; 4-Ring NOGE; 5-Ring NOGE	LC-GC (NPLC-FLD GC-MS) ( $\lambda_{\text{exc}}$ 225 nm, $\lambda_{\text{em}}$ 295 nm)	LC: Cyano phase GromsilCN 2 PR column (25 cm $\times$ 2 mm, 5 $\mu\text{m}$ ). Gradient elution consisted of (pump1) 20% MTBE/pentane, (pump 2) 50% 1-propanol/20% MTBE/30% pentane Flow: 400 $\mu\text{l}/\text{min}$ GC: Capillary column (8 m $\times$ 0.25 mm i.d.) coated with a 0.2- $\mu\text{m}$ film of SOP-50 Temp. program: 80–320°C Injection mode: Large volume on-column	LOD: 5–30 $\mu\text{g}/\text{kg}$ Biedermann and Grob (1998)
Vinyllic organosol	Positional isomers of BFDGE (iBFDGE); BAMGE; positional isomer of BADGE (iBADGE); BADGE; iBFDGE·MeOH; iBFDGE·HCl; BADGE·MeOH; BADGE·HCl	GC-MS: Full scan mode $m/z$ 60–600 LC-MS (ESI+) (APCI+), (ESI+): BADGE 358.1 [M+NH <sub>4</sub> ] <sup>+</sup> ; 363.2 [M+Na] <sup>+</sup> ; 378.9 [M+K] <sup>+</sup> (APCI+): BADGE 382.2 [M+CH <sub>3</sub> CN+H] <sup>+</sup>	GC-MS: DB-5 MS (15 m $\times$ 0.2 mm i.d., 0.2 $\mu\text{m}$ bonded phase) capillary column. Temp. program: 180–280°C. Injector: 260°C. Source and transfer line: 100 and 270°C. Carrier gas: Helium LC-MS: Li-Chrospher C <sub>8</sub> (250 $\times$ 4.6 mm, 5 $\mu\text{m}$ ) column. Mobile phase: ACN–water (0.1 M ammonium acetate) (50:50, v/v). Elution mode: isocratic Flow: 0.5 ml/min	Cottier et al. (1997)

(Continues)

TABLE 3 (Continued)

Coatings	Potential migrants	Analytical technique detection system	Chromatographic conditions	Sensitivity/validation	Reference
Epoxy	BADGE·2H <sub>2</sub> O; BADGE·H <sub>2</sub> O; BADGE·H <sub>2</sub> O·BuEtOH; BADGE·H <sub>2</sub> O·Ph; BADGE·H <sub>2</sub> O·BPA; BADGE·BuEtOH; BADGE·Ph; Cyclo-DiBADGE; BADGE·BPA; BADGE·BPA·BuEtOH; BADGE (n = 1)·BuEtOH; BADGE (n = 1)·BPA; BADGE (n = 1)·BPA·BuEtOH; BADGE (n = 2); BADGE; BADGE·PrOH; BADGE·2BPA; BADGE (n = 2)·H <sub>2</sub> O; BADGE (n = 2)·PrOH; BADGE·2BuEtOH; BADGE·BuEtOH·BuOH; BADGE (n = 1)·2BuEtOH; BADGE (n = 1)·BuEtOH·BuOH	HPLC-MS (ESI+)-UVD-FLD (UV (λ 275 nm) (FLD (λ <sub>ex</sub> 275 nm, λ <sub>em</sub> 305 nm) Scan range m/z 80-1000. Protonated molecular ions (m/z) [M+H] <sup>+</sup> BADGE·2H <sub>2</sub> O 377; BADGE·H <sub>2</sub> O 359; BADGE·H <sub>2</sub> O·BuEtOH 477; BADGE·H <sub>2</sub> O·Ph 453; BADGE·H <sub>2</sub> O·BPA 587; BADGE·BuEtOH 459; BADGE·Ph 435; Cyclo-DiBADGE 569; BADGE·BPA 569; BADGE·BPA·BuEtOH 687; BADGE (n = 1)·BuEtOH 743; BADGE (n = 1)·BPA 853; BADGE (n = 1)·BPA·BuEtOH 971; BADGE (n = 2) 909; BADGE 341; BADGE·PrOH 401; BADGE·2BPA 797; BADGE (n = 2)·H <sub>2</sub> O 927; BADGE (n = 2)·PrOH 969; BADGE·2BuEtOH 577; BADGE·BuEtOH·BuOH 533; BADGE (n = 1)·2BuEtOH 861; BADGE (n = 1)·BuEtOH·BuOH 817	Multospher <sup>®</sup> 100 5C18 column (250 × 3 mm). Mobile phase: 1 mM ammonium formate in water (pH 3) and can. Elution mode: gradient Flow: 0.5 ml/min Column temp.: 25°C	Reproducibility: RSD < 11.50% LOD: 1.0 µg/L	Schaefer and Simat (2004)

(Continues)

TABLE 3 (Continued)

Coatings	Potential migrants	Analytical technique detection system	Chromatographic conditions	Sensitivity/validation	Reference
Polyvinyl chloride	ATBC; DEHA; OA; DBP; DEHP; BGA; BGA-formaldehyde; ELSO	UHPLC-HRMS (ESI+) (APCI+) Full scan $m/z$ 75–1000; 87–1300 Protonated molecular ions ( $m/z$ ) [M+H] <sup>+</sup> ATBC 403.2326; DEHA 371.3156; OA 282.2791; DBP 279.1591; DEHP 279.1591; BBP 313.1434; DEHP 391.2843; BGA 188.0935; BGA-formaldehyde 216.0880; ELSO (APCI) (C <sub>55</sub> H <sub>98</sub> O <sub>10</sub> 919.7233; C <sub>57</sub> H <sub>62</sub> O <sub>15</sub> 1017.6509; C <sub>57</sub> H <sub>96</sub> O <sub>13</sub> 989.6924; C <sub>57</sub> H <sub>98</sub> O <sub>12</sub> 975.7131; C <sub>57</sub> H <sub>100</sub> O <sub>11</sub> 961.7339; C <sub>57</sub> H <sub>102</sub> O <sub>10</sub> 947.7546)	Acquity HSS T3 (100 × 2.1 mm, 1.8 μm) analytical column. Mobile phase: 5 mM ammonium formate with 0.1% formic acid and MeOH containing 0.1% formic acid Elution mode: gradient Flow: 300 μl/min Injection vol.: 5 μl Column temp.: 40°C	Linearity: R <sup>2</sup> > .99 ESI: LOD: 1–5 ng/ml LOQ: 1–10 ng/ml APCI: LOD: 0.5–5 μg/ml LOQ: 1–25 μg/ml Repeatability: <12%	Vaclavikova et al. (2016)
Polyester	Cis-endo-nadic acid; trans-endo,exo-nadic acid; cis-endo-nadic anhydride; cis-exo-nadic anhydride. PA and IPA. PUD	Phthalic acids and nadic acids. LC-MS/MS (ESI-) Selected reaction monitoring (SRM) transitions cis-endo-nadic acid 181.1 > 115.0; trans-endo,exo-nadic acid; 181.1 > 163.0; Phthalic and isophthalic acid 165.0 > 120.9 NAH (nadic anhydride) isomers. GC-TOF-MS Scan range $m/z$ 30–400 exo-NAH and endo-NAH $m/z$ 66.0470 for quantification and $m/z$ 91.0548 for confirmation Oligomers LC-TOF-MS (ESI+) (ESI-) mass range $m/z$ 100–1100	LC-MS/MS: Acquity UPLC HSS T3 (100 × 2.1 mm, 1.8 μm) column. Mobile phase: water with 0.1% acetic acid/0.1 mM ammonium fluoride and MeOH:ACN (1:1). Elution mode: gradient Flow: 0.4 ml/min Injection vol.: 1 μl Column temp.: 40°C GC-TOF-MS: HP-5MS Ultra Inert column (30 m × 0.25 mm i.d., 0.25 μm) column. Temp. program: 40–290°C. Carrier gas: Helium Flow: 1.1 ml/min LC-TOF-MS: Atlantis dC18 (150 × 2.1 mm, 3 μm) column. Mobile phase: 10 mM ammonium acetate in water and MeOH Elution mode: gradient Flow: 0.4 ml/min Injection vol.: 3 μl Column temp.: 30°C	LOD: 0.01–26 μg/kg	Driffield et al. (2018)

(Continues)

TABLE 3 (Continued)

Coatings	Potential migrants	Analytical technique detection system	Chromatographic conditions	Sensitivity/validation Reference
Polyester-polyurethane	Predicted oligomer combinations: Cyclic: 2NPG+2PA; 3NPG+3PA; 4NPG+4PA; NPG+HD+2PA; NPG+2HD+3PA; 2HD+2PA; NPG+3HD+4PA; 2NPG+2HD+4PA; 2NPG+HD+3PA; 3HD+3PA; 4HD+4PA; 3NPG+HD+4PA; DEG+NPG+2PA; 2DEG+2PA; EG+DEG+2PA; EG+2NPG+3PA; DEG+2NPG+3PA; EG+DEG+NPG+3PA; 2DEG+NPG+3PA; DEG+3NPG+4PA; EG+3NPG+4PA; EG+2DEG+3PA; EG+DEG+2NPG+4PA; EG+NPG+2PA; 2EG+NPG+3PA; 2EG+2NPG+4PA Linear: 2HD+PA; NPG+HD+PA	LC-HRMS (HESI+) Full-scan mode $m/z$ 155–1200. Mean measured $[M+NH_4]^+$ $m/z$ Cyclic: 2NPG+2PA 486.21289; 3NPG+3PA 720.30192; 4NPG+4PA 954.39106; NPG+HD+2PA 500.22849; NPG+2HD+3PA 748.33313; 2HD+2PA 514.24422; NPG+3HD+4PA 996.43788; 2NPG+2HD+4PA 982.42209; 2NPG+HD+3PA 734.31758; 3HD+3PA 762.34875; 4HD+4PA 1010.45361; 3NPG+HD+4PA 968.40703; DEG+NPG+2PA 488.19216; 2DEG+2PA 490.17139; EG+DEG+2PA 446.14514; EG+2NPG+3PA 678.25511; DEG+2NPG+3PA 722.28122; EG+DEG+NPG+3PA 680.23438; 2DEG+NPG+3PA	Hypersil Gold (100 × 2.1 mm, 1.9 μm) analytical column. Mobile phase: 10 mM ammonium acetate in water and 10 mM ammonium acetate in ACN. Elution mode: gradient Flow: 0.4 ml/min Injection vol.: 10 μl Column temp.: 40°C	Omer et al. (2018)

(Continues)

TABLE 3 (Continued)

Coatings	Potential migrants	Analytical technique detection system	Chromatographic conditions	Sensitivity/validation	Reference
	Unpredicted compounds;	724.26048; DEG+3NPG+4PA 956.37058;			
	Cyclic: Caprolactam dimer;	EG+3NPG+4PA 912.34420;			
	Caprolactam trimer;	EG+2DEG+3PA 682.21360;			
	Caprolactam tetramer;	EG+DEG+2NPG+4PA914.32351;			
	EG+NPG+PA+SA;	EG+NPG+2PA 444.16589;			
	EG+DEG+PA+SA;	2EG+NPG+3PA 636.20833;			
	EG+DEG+2SA;	2EG+2NPG+4PA 870.29726			
	2NPG+PA+SA;	Linear: 2HD+PA 384.23855;			
	DEG+NPG+PA+SA;	NPG+HD+PA 370.22284			
	2DEG+PA+SA;	Unpredicted compounds:			
	NPG+TEG+2PA;	Cyclic: Caprolactam dimer 227.17565			
	DEG+TEG+2PA;	[M+H] <sup>+</sup> ; Caprolactam trimer			
	EG+2NPG+2PA+SA;	340.25993 [M+H] <sup>+</sup> ; Caprolactam			
	EG+NPG+DEG+2PA+SA;	tetramer 453.34406 [M+H] <sup>+</sup> ;			
	3NPG+2PA+SA;	EG+NPG+PA+SA 480.25989			
	DEG+2NPG+2PA+SA;	[M+NH <sub>4</sub> ] <sup>+</sup> ; EG+DEG+PA+SA			
	C <sub>12</sub> H <sub>24</sub> O <sub>2</sub> +NPG+HD+3PA;	482.23913 [M+NH <sub>4</sub> ] <sup>+</sup> ; EG+DEG+2SA			
	C <sub>12</sub> H <sub>24</sub> O <sub>2</sub> +2HD+3PA;	518.33325 [M+NH <sub>4</sub> ] <sup>+</sup> ; 2NPG+PA+SA			
	4NPG+3PA+SA	522.30681 [M+NH <sub>4</sub> ] <sup>+</sup> ;			
		DEG+NPG+PA+SA 524.28620			
		[M+NH <sub>4</sub> ] <sup>+</sup> ; 2DEG+PA+SA 526.26544			
		[M+NH <sub>4</sub> ] <sup>+</sup> ; NPG+TEG+2PA 532.21841			
		[M+NH <sub>4</sub> ] <sup>+</sup> ; DEG+TEG+2PA 534.19774			
		[M+NH <sub>4</sub> ] <sup>+</sup> ; EG+2NPG+2PA+SA			
		714.34891 [M+NH <sub>4</sub> ] <sup>+</sup> ;			
		EG+NPG+DEG+2PA+SA 716.32826			
		[M+NH <sub>4</sub> ] <sup>+</sup> ; 3NPG+2PA+SA 756.39567			
		[M+NH <sub>4</sub> ] <sup>+</sup> ; DEG+2NPG+2PA+SA			
		758.37498 [M+NH <sub>4</sub> ] <sup>+</sup> ;			
		C <sub>12</sub> H <sub>24</sub> O <sub>2</sub> +NPG+HD+3PA 830.41152			
		[M+NH <sub>4</sub> ] <sup>+</sup> ; C <sub>12</sub> H <sub>24</sub> O <sub>2</sub> +2HD+3PA			
		844.42731 [M+NH <sub>4</sub> ] <sup>+</sup> ; 4NPG+3PA+SA			
		990.48536 [M+NH <sub>4</sub> ] <sup>+</sup>			

(Continues)

TABLE 3 (Continued)

Coatings	Potential migrants	Analytical technique detection system	Chromatographic conditions	Sensitivity/validation Reference
Polyester-polyurethane	2EG+2PA; 2EG+PA+SA; EG+NPG+2PA; EG+DEG+2PA; EG+NPG+PA+SA; EG+DEG+PA+SA; 2NPG+2PA; NPG+DEG+2PA; 2DEG+2PA; NPG+HD+2PA; 2HD+2PA; EG+DEG+2SA; 2NPG+PA+SA; NPG+DEG+PA+SA; 2DEG+PA+SA; NPG+TEG+2PA; DEG+TEG+2PA; IPDI; DPMDI	GC-MS (GC-(EI)qMS; GC-(EI)Orbitrap; GC-(APCI)TOFHRMS) Full-scan mode $m/z$ 50–750; $m/z$ 50–1200. Molecular mass: 2EG+2PA 384.08452; 2EG+PA+SA 420.17842; EG+NPG+2PA 426.13147; EG+DEG+2PA 428.11074; EG+NPG+PA+SA 462.22537; EG+DEG+PA+SA 464.20464; 2NPG+2PA 468.17842; NPG+DEG+2PA 470.15769; 2DEG+2PA 472.13695; NPG+HD+2PA 482.19407; 2HD+2PA 496.20972; EG+DEG+2SA 500.29854; 2NPG+PA+SA 504.27232; NPG+DEG+PA+SA 506.25159; 2DEG+PA+SA 508.23085; NPG+TEG+2PA 514.18390; DEG+TEG+2PA 516.16317	DB-5MS capillary column (30 m × 0.25 mm i.d., 0.25 μm). Temp. program: 120–310°C Injector: 250°C Carrier gas: Helium Flow: 1 ml/min (1.7 ml/min) Injection mode: Splitless Injection vol.: 1 μl	Omer et al. (2019)
Polyester-polyurethane	IPDI; MEKO; 2-Ethylhexanoic acid; Oligomers: 2 PA+2 EG; 2PA+EG+NPG; 2PA+EG+DEG; 2PA+CHDM+EG; 2PA+2NPG; 2PA+CHDM+NPG; 3PA+2EG+NPG or 3PA+3 PG; 3PA+EG+2NPG; 3PA+CHDM+EG+NPG; 3PA+3NPG; 3PA+CHDM+2PG; 3PA+CHDM+2NPG; 3PA+2CHDM+NPG	MEKO-blocked IPDI. LC-MS/MS (ESI+) $m/z$ 928.7 [M+H] <sup>+</sup> (quantification) and $m/z$ 950.68 [M+Na] <sup>+</sup> (confirmation). 2-Ethylhexanoic acid. GC-MS $m/z$ 88 and 116. Oligomers. LC-TOF-MS, mass: 2PA+2EG (Cyclic) 384.0845; 2PA+EG+NPG (Cyclic) 426.1315; 2PA+EG+DEG (Cyclic) 428.1107; 2PA+CHDM+EG (Cyclic) 466.1630; 2PA+2NPG (Cyclic) 468.1784; 2PA+CHDM+NPG (Cyclic) 508.2114; 3PA+2EG+NPG or 3PA+3(PG) (Cyclic) 618.1737; 3PA+EG+2NPG (Cyclic) 660.2238; 3PA+CHDM+EG+NPG (Cyclic) 700.2520; 3PA+3NPG 702.2703; 3PA+CHDM+2PG (Linear) 704.2469; 3PA+2CHDM+NPG (Cyclic) 742.3003; 3PA+2CHDM+NPG (Cyclic) 782.3330	IPDI trimer: Waters Sunfire (C <sub>18</sub> ) column (150 × 2.1 mm, 3.5 μm). Mobile phase: 0.05% formic acid in ACN. Flow: 0.2 ml/min Injection vol.: 20 μl Column temp.: 30°C	Bradley et al. (2009) MEKO-blocked IPDI LOD: 0.01 μg/dm <sup>2</sup> 2-Ethylhexanoic acid LOD: 0.1 μg/dm <sup>2</sup> IPDI trimer LOD: 0.02 μg/dm <sup>2</sup>

(Continues)



TABLE 3 (Continued)

Coatings	Potential migrants	Analytical technique detection system	Chromatographic conditions	Sensitivity/validation	Reference
Polyester-phenol	TPA, IPA, TMA, ADA, EG, DEG, NPG, MPD, BD, TCDDM. Polyester oligomers: 2TPA+2BD; 2TPA+1BD+1TCDDM; 2TPA+2TCDDM; 3TPA+3BD; 3TPA+2BD+1TCDDM; 3TPA+3TCDDM; 2PA+2EG; 2PA+1NPG+1EG; 3PA+3EG; 3PA+2EG+1NPG; 3PA+2NPG+1EG; 3PA+3NPG; 4PA+4EG; 4PA+3EG+1NPG; 4PA+2EG+2NPG; 4PA+3NPG+1EG; 4PA+4NPG; 2PA+2EG; 2PA+2MPD; 2PA+1MPD+1DEG; 2PA+2DEG; 3PA+3MPD; 3PA+2MPD+1DEG; 3PA+2DEG+1MPD; 3PA+3DEG	Polyester acids. HPLC-DAD (scan 200–400 nm) $\lambda$ 210, 220, 240, 275 nm. Polyester polyols GC-MSD (scan $m/z$ 30–150) Polyester oligomers RP-HPLC-DAD-MS (ESI+) ( $\lambda$ 232 nm) Precursor ions [M+H] <sup>+</sup> 2TPA+2BD 441.5; 2TPA+1BD+1TCDDM 547.7; 2TPA+2TCDDM 653.8; 3TPA+3BD 661.8; 3TPA+2BD+1TCDDM 767.9; TPA+2TCDDM+1BD 874.1; 3TPA+3TCDDM 980.3; 2PA+2EG 385.2; 2PA+1NPG+1EG 427.4; 2PA+2NPG 469.5; 3PA+3EG 577.3; 3PA+2EG+1NPG 618.5; 3PA+2NPG+1EG 661.6; 3PA+3NPG 703.8; 4PA+4EG 769.4; 4PA+3EG+1NPG 811.6; 4PA+2EG+2NPG 853.7; 4PA+3NPG+1EG 895.9; 4PA+4NPG 938.0; 2PA+2EG 385.2; 2PA+2MPD 441.4; 2PA+1MPD+1DEG 457.3; 2PA+2DEG 473.2; 3PA+3MPD 661.6; 3PA+2MPD+1DEG 677.5; 3PA+2DEG+1MPD 693.4; 3PA+3DEG 709.3	HPLC-DAD Polaris® C18-Ether column. Mobile phase: 0.1% (v/v) trifluoroacetic acid in distilled water and ACN. Elution mode: gradient Flow: 0.5 ml/min Injection vol.: 10 $\mu$ l Column temp.: 30°C GC-MSD WAXplus® capillary column (30 m $\times$ 0.25 mm i.d., 0.5 $\mu$ m) Temp. program: 60–260°C. Injector temp.: 260°C Transfer line: 260°C Carrier gas: Helium Injection mode: Splitless RP-HPLC-DAD-MS Multospher 120 RP18 HP column (250 $\times$ 3 mm, 5 $\mu$ m). Mobile phase: 0.015% (v/v) formic acid in water and 2-propanol. Elution mode: gradient Flow: 0.4 mL/min Injection vol.: 10 $\mu$ l Column temp.: 60°C Multohigh 100 Phenyl column (250 $\times$ 3 mm, 5 $\mu$ m) Mobile phase: 0.015% (v/v) formic acid in water and ACN. Elution mode: gradient Flow: 0.3 ml/min Injection vol.: 10 $\mu$ l Column temp.: 35°C	Recovery: 80%–120%	Eckardt et al. (2020)

(Continues)

TABLE 3 (Continued)

Coatings	Potential migrants	Analytical technique detection system	Chromatographic conditions	Sensitivity/validation	Reference
Polyester-polyurethane	IPDI trimer	LC-MS/MS (ESI+) IPDI trimer derivatized with dibutylamine. Precursor ion $m/z$ 1078 [M+Na] <sup>+</sup>	Waters Sunfire (C <sub>18</sub> ) column (150 × 2.1 mm, 3.5 μm). Mobile phase: 0.05% formic acid in ACN. Flow: 0.2 ml/min Injection vol.: 20 μl Column temp.: 30°C	Linearity: $R^2 > .9979$ for the quantification channel 1078 > 819 and $R^2 > .9906$ for the confirmation channel 1078 > 949 LOD: 0.02 μg/100 cm <sup>2</sup> Recovery: 87%–106% Repeatability: RSD < 24%	Driffield et al. (2007)

(Continues)

TABLE 3 (Continued)

Coatings	Potential migrants	Analytical technique detection system	Chromatographic conditions	Sensitivity/validation Reference
Polyester-urethane and polyester-phenolic	ADA; IPA; PA; TPA; TMA; dimethyl isophthalate; BHET; 1,3-BD; EG; HD; NPG; 1,2-PG; 1,3-PG; 2,2,4-trimethylpentane-1,3-diol; COH; MPD; Cyclic oligoesters: CYCLO DA (dicarboxylic acid) PO (polyols) (2DA 2PO-1); CYCLO (2DA IPO-1 IPO-2); CYCLO (2DA 2PO-2); CYCLO (3DA IPO-1 2PO-2); CYCLO (2DA 2PO-2 IPO-3); CYCLO (3DA 3PO-2); CYCLO (4DA IPO-1 3PO-2); CYCLO (3DA 2PO-2 IPO-3); CYCLO (4DA 4PO-3); CYCLO (3DA 3PO-1); CYCLO (2DA IPO-1 IPO-4); CYCLO (3DA 2PO-1 IPO-4); CYCLO (2DA 2PO-4); CYCLO (3DA IPO-1 2PO 4); CYCLO (2DA IPO-5 IPO-6); CYCLO (2DA 2PO-5); CYCLO (2DA IPO-3 IPO-5); CYCLO (3DA IPO-3 2PO-5)	Polyols (PO): GC-FID; dicarboxylic acids (DA): HPLC-MS-UV (ESI+); cyclic oligoesters: HPLC-MS-UV (ESI+) and HPLC-ELSD-UV (UV $\lambda$ 232 nm) (Mass range $m/z$ 80–1000)	GC-FID: CP-Pora-Plot Q-HT, capillary column (12.5 m $\times$ 0.32 mm i.d., 10 $\mu$ m). Temp. program: 160–270°C Carrier gas: Nitrogen Injector temp.: 280°C Detector temp.: 280°C HPLC-MS-UV (ESI+) and HPLC-ELSD-UV: For dicarboxylic acids Aqua 125 5C18, column (250 $\times$ 3 mm). Mobile phase: 5 mM ammonium formate in water (pH 3.9) and ACN. Elution mode: gradient Flow: 0.5 ml/min Column temp.: 30°C For cyclic oligoesters Multospher® 100 5C18, column (250 $\times$ 3 mm). Mobile phase: 1 mM ammonium formate in water (pH 3) for MS or bidistilled water for ELS detection and ACN. Elution mode: gradient Flow: 0.5 ml/min Column temp.: 25°C	LOQ: 0.01 mg/dm <sup>2</sup> Schaefer et al. (2004)

(Continues)

TABLE 3 (Continued)

Coatings	Potential migrants	Analytical technique detection system	Chromatographic conditions	Sensitivity/validation Reference
Polyester	Linear oligomers: NA+2EG; PA+MBO; NA+COH; PA+2MBO; PA+COH; 2PA+MBO; NA+COH+MBO; PA+MBO+COH; NA+PA+2MBO; 2PA+2MBO; NA+PA+COH; NA+2COH; 2PA+COH; NA+PA+3MBO; PA+NA+COH+MBO; PA+tBuPh+COH; 2PA+MBO+COH; 2PA+2MBO+COH; 2PA+2COH; 3PA+2COH+MBO; 3PA+2MBO+COH; 2PA+2COH+MBO. Cyclic oligomers: 3PA+3MBOc; 2NA+MBO+COHc; 3PA+2MBO+COHc; 2PA+COH+MBOc; 2NA+PA+3MBOc; PA+NA+COH+MBOc; NA+PA+2COHc	HPLC-DAD/MS (ESI+) (ESI-) ( $\lambda$ ) 200–400 nm, 225, 254, 280, 288, and 330 nm) Full-scan mode $m/z$ 100–1000 HPLC-DAD/CAD UHPLC-HRMS (ESI+, ESI-) Full-scan $m/z$ 100–800 (ESI+) Full-scan $m/z$ 100–1000 (ESI-) Exact mass ( $m/z$ ): Linear oligomers: NA+2EG 293.0999 [M+Na] <sup>+</sup> ; PA+MBO 237.0771[M-H] <sup>-</sup> ; NA+COH 331.1519 [M+Na] <sup>+</sup> ; PA+2MBO 333.1212 [M+Na] <sup>+</sup> ; PA+COH 291.1242 [M-H] <sup>-</sup> ; 2PA+MBO 385.0935 [M-H] <sup>-</sup> ; NA+COH+MBO 403.2095[M+Na] <sup>+</sup> ; PA+MBO+COH 387.1781[M+Na] <sup>+</sup> ; NA+PA+2MBO 497.1790 [M+Na] <sup>+</sup> ; 2PA+2MBO 457.1511[M-H] <sup>-</sup> ; NA+PA+COH 474.2126 [M+NH <sub>4</sub> ] <sup>+</sup> ; NA+2COH 457.2566 [M+Na] <sup>+</sup> ; 2PA+COH 463.1363 [M+Na] <sup>+</sup> ; NA+PA+3MBO 569.2364 [M+Na] <sup>+</sup> ; PA+NA+COH+MBO 551.2259 [M+Na] <sup>+</sup> ; PA+tBuPh+COH 425.2302 [M+H] <sup>+</sup> ; 2PA+MBO+COH 511.1979 [M-H] <sup>-</sup> ; 2PA+2MBO+COH 607.2521[M+Na] <sup>+</sup> ; 2PA+2COH 565.2450 [M-H] <sup>-</sup> ; 3PA+2COH+MBO 787.3306 [M+H] <sup>+</sup> ; 3PA+2MBO+COH 750.3124 [M+NH <sub>4</sub> ] <sup>+</sup> ; 2PA+2COH+MBO 639.3170 [M+H] <sup>+</sup> . Cyclic oligomers: 3PA+3MBOc 678.2500[M+NH <sub>4</sub> ] <sup>+</sup> ; 2NA+MBO+COHc 544.2913[M+NH <sub>4</sub> ] <sup>+</sup> ; 3PA+2MBO+COHc 715.2726 [M+H] <sup>+</sup> ; 2PA+COH+MBOc 517.1840 [M+Na] <sup>+</sup> ; 2NA+PA+3MBOc 710.3172 [M+NH <sub>4</sub> ] <sup>+</sup> ; PA+NA+COH+MBOc 511.2331[M+H] <sup>+</sup> ; NA+PA+2COHc 565.2804[M+H] <sup>+</sup>	HPLC-DAD/MS Agilent Polaris 3 (C18) column (150 × 2.0 mm). Mobile phase: water 0.1% formic acid and ACN 0.1% formic acid. Elution mode: gradient Flow: 0.3 ml/min Injection vol.: 10 $\mu$ l Column temp.: 25°C HPLC-DAD/CAD Agilent Zorbax Extend C18 column 80 Å (2.1 × 150 mm, 3 $\mu$ m). Mobile phase: water and ACN. Elution mode: gradient Flow: 0.3 ml/min Injection vol.: 10 $\mu$ l Column temp.: 25°C UHPLC-HRMS Acquity UPLC BEH C18 column (150 × 2.1 mm, 1.7 $\mu$ m). Mobile phase: 5% ACN in water 0.1% formic acid and 5% water in ACN 0.1% formic acid Elution mode: gradient Flow: 0.2 ml/min Injection vol.: 1 $\mu$ l	Paseiro-Cerrato, MacMahon et al. (2016)

(Continues)

TABLE 3 (Continued)

Coatings	Potential migrants	Analytical technique detection system	Chromatographic conditions	Sensitivity/validation Reference
Polyester	Linear oligomers: IPA+2 HD; IPA+2NPG; IPA+2MPD; TPA+2HD.	HPLC-MS (ESI+) Precursor ion <i>m/z</i> [M+H] <sup>+</sup>	HPLC-MS: Atlantis T3 C <sub>18</sub> column (150 mm × 4.6 mm, 5 μm).	Linearity: R <sup>2</sup> > .996 LOD:
	Cyclic oligomers: 2TPA+2HD; 2IPA+2HD; 2IPA+2MPD; 2TPA+2MPD; IPA+TPA+2NPG; 2IPA+HD+MPD; 2IPA+HD+NPG; 3IPA+3HD 2DPA+2HD; (internal standard)	IPA+2NPG 339.2; TPA+ 2HD 367.2; IPA+2HD 367.2; IPA+2MPD 311.1; 2TPA+2HD 497.2; 2IPA+2HD 497.2; 2IPA+2MPD 441.2; 2TPA+2MPD 441.2; IPA+TPA+2NPG 469.2; 2IPA+HD+MPD 469.2; 2IPA+HD+NPG 483.2; 3IPA+3HD 745.3; 2DPA+2HD 499.2	Mobile phase: ultrapure water with 0.1% trifluoroacetic acid and MeOH. Elution mode: gradient Flow: 1.0 ml/min HPLC-HRMS/MS: Halo C18 column (100 × 0.5 mm, 2.7 μm). Mobile phase: ammonium acetate (1.0 mM) in water and ammonium acetate (1.0 mM) in MeOH. Elution mode: gradient Flow: 15.0 μl/min Injection vol.: 300 nl Column temp.: 45°C	0.003–0.30 ng/ml LOQ: 0.010–1.00 ng/ml Intraday: RSD < 8.2% Interday: RSD < 13.3% Recovery: 55.1–68.4%
Acrylic resins	MMA; EMA; BMA; iBMA	GC-FID	DB VXR column (30 m × 0.32 mm i.d., 1.8 μm) and DB 624 column (30 m × 0.32 mm i.d., 1.8 μm). Temp. program: 35–260°C Transferline: 150°C Detector: 260°C	Linearity: R <sup>2</sup> > .996 LOD (Water): 20–80 pg/ml LOD (Tenax): 0.25 μg /ml LOD (Miglyol 840): 0.2–1.5 ng/ml Recovery (Water): 88%–104% Recovery (Tenax): 87%–100% Recovery (Miglyol 840): 91%–99%

(Continues)

TABLE 3 (Continued)

Coatings	Potential migrants	Analytical technique detection system	Chromatographic conditions	Sensitivity/validation	Reference
Acrylic-phenolic	BPA, BADGE-2H <sub>2</sub> O; BGA	BPA: HPLC-MS/MS (ESI-) MS/MS transitions: 227.3 <i>m/z</i> to 212.3 and 133.1; BADGE-2H <sub>2</sub> O: HPLC-DAD (λ 225 nm); BGA: UHPLC-HRMS (ESI+) [M+H] <sup>+</sup> 188.0935	Agilent Polaris 3 C18 column (150 × 2.0 mm). Mobile phase: water and ACN. Elution mode: gradient Flow: 0.1 ml/min Injection vol.: 5 μl Column temp.: 35°C BGA: Acquity HSS T3 column (100 mm × 2.1 mm and 1.8 μm). Mobile phase: 5 mM ammonium formate with 0.1% formic acid and methanol containing 0.1% formic acid Elution mode: gradient Flow: 300 μl/min Injection vol.: 5 μl Column temp.: 40°C	LOD: BADGE-2H <sub>2</sub> O: 50 ng/ml; LOD BGA: 0.2 ng/ml Recovery: 80%–110%	Paseiro-Cerrato et al. (2017)

(Continues)



TABLE 3 (Continued)

Coatings	Potential migrants	Analytical technique detection system	Chromatographic conditions	Sensitivity/validation Reference
Phenolic resins (phenol-based resole)	Phenol (P); Methylol (M); Butyl (B); Hemiformaldehyde (F); Methylene (m). P; PM; PMB; P2MB; P2M2B; P2MBF; P2M2BF; P3MB; P3M2B; P3M3B; P3M2BF; P3M3BF; (PmP)2M2B; (PmP)3M2B; (PmP)3M3B; (PmP)4M3B; (PmP)4M4B; (PmP)4M4BF	GC-MS; GC×GC-FID; NPLC; SEC (ELSD, FLD $\lambda$ 225/295 nm) The samples were trimethyl silylated with N,O-bis-(trimethylsilyl)-trifluoroacetamide before the GC analysis. GC-MS (electron impact EI, chemical ionization CI) Molecular mass ( $M^+$ ) P 166; PM 268; PMB 252; P2MB 354; P2M2B 338; P2M2BF 384; P2M2BF 368; P3MB 456; P3M2B 440; P3M3B 424; P3M2BF 470; P3M3BF 454; (PmP)2M2B 516; (PmP)3M2B 618; (PmP)3M3B 602; (PmP)4M3B 704; (PmP)4M4B 688; (PmP)4M4BF 718	GC-MS: Capillary column (30 m × 0.25 mm i.d., 0.15 $\mu$ m PS-255) equipped with an uncoated precolumn (20 cm × 0.53 mm i.d., deactivated with diphenyl tetramethyl disilazane). Temp. program: 50–350°C Transfer line: 300°C Source: 200°C Carrier gas: Helium Injection mode: on-column Injection vol.: 1 $\mu$ l GC×GC-FID: Capillary column (30 m × 0.25 mm i.d., 0.15 $\mu$ m PS-255) coupled with a (1.5 m × 0.1 mm i.d., 0.1 $\mu$ m SOP-50 [50% phenyl silicone]) second dimension column. Temp. program: 50–360°C Carrier gas: Hydrogen Injection mode: Splitless NPLC: Grom-Sil 100 Cyano-2 PR column (25 cm × 2 mm i.d., 5 $\mu$ m). Mobile phase: 1% 1-propanol/hexane and 50% 1-propanol/dichloromethane Elution mode: gradient Flow: 400 $\mu$ l/min Injection vol.: 3 $\mu$ l SEC: Phenogel 5 mm/500 Å (300 × 7.8 mm i.d.) column and (50 × 7.8 mm i.d.) guard column. Mobile phase: redistilled THF Flow: 800 $\mu$ l/min	Biedermann and Grob (2006a)

(Continues)

TABLE 3 (Continued)

Coatings	Potential migrants	Analytical technique detection system	Chromatographic conditions	Sensitivity/validation Reference
Phenolic resins (Resoles based on cresol/phenol mixtures or tert. butyl phenol)	Phenol (P); Cresol (C); tert. Butyl phenol (T); Methylol (M); Butyl (B); Hemiformaldehyde hemiacetals (F); Methylene (m). Mono-methylene ether (Om)	GC-MS; GCXGC-FID; NPLC; SEC (ELSD, FLD $\lambda$ 225/295 nm). The samples were trimethyl silylated with N,O-bis-(trimethylsilyl)-trifluoroacetamide before the GC analysis. GC-MS (electron impact EI, chemical ionization CI) Molecular mass ( $M^+$ ) o-Cresol 180; m-Cresol 180, p-Cresol 180; o-PM 268, p-PM 268, CM 282; PMB 252; CMB 266; P2M 370; C2M 284; P2MB 354; C2MB 368; (PmP) 344; C2M2B 352; (CmP) 558; (CmC) 372; P3MB 456, (CmP)M 460; (PmP)M 446; (CmC)M 474; (CmP)MB 444; (CmC)MB 458; (CmP)2M 562; (CmC)2M 576; (CmP)2MB 546; (CmC)2MB 560; (CmP)3M 664; (CmC)3M2B 544; (CmP)3MB 648; (CmP)3M2B 632; CMF 312; PMBF 282; CMBF 296; P2M2B 338; C2M2BF 382; P3M2B 440; P3M2BF 470; C3M2B 454; P3M3B 424; C3M3B 438	GC-MS: Capillary column (30 m $\times$ 0.25 mm i.d., 0.15 $\mu$ m PS-255) equipped with an uncoated precolumn (20 cm $\times$ 0.53 mm i.d. deactivated with diphenyl tetramethyl disilazane). Temp. program: 50–350°C Injection mode: on-column Injection vol.: 1 $\mu$ l GCXGC-FID: Capillary column (30 m $\times$ 0.25 mm i.d., 0.15 $\mu$ m PS-255) coupled with a (1.5 m $\times$ 0.1 mm i.d., 0.1 $\mu$ m SOP-50 [50% phenyl silicone]) second dimension column. Temp. program: 50–360°C Injection mode: Splitless NPLC: Grom-Sil 100 Cyano-2 PR column (25 cm $\times$ 2 mm, 5 $\mu$ m). Mobile phase: 1% 1-propanol/hexane and 50% 1-propanol/dichloromethane Elution mode: gradient Flow: 400 $\mu$ l/min SEC: Phenogel 5 mm/500 Å (300 $\times$ 7.8 mm) column and (50 $\times$ 7.8 mm) guard column. Mobile phase: redistilled THF Flow: 800 $\mu$ l/min	Biedermann and Grob (2006b)

Abbreviations: ACN, acetonitrile; ADA, adipic acid; APCI, atmospheric pressure chemical ionization; ATBC, acetyl tributyl citrate; B, butyl; BADGE, bisphenol A diglycidyl ether; BADGE-H<sub>2</sub>O, bisphenol A (2,3-dihydroxypropyl) glycidyl ether; BADGE-2H<sub>2</sub>O, bisphenol A bis(2,3-dihydroxypropyl) ether; BADGE-HCl, bisphenol A bis(3-chloro-2-hydroxypropyl) glycidyl ether; BADGE-2HCl, bisphenol A bis(3-chloro-2-hydroxypropyl) glycidyl ether; BADGE-H<sub>2</sub>O-HCl, bisphenol A (3-chloro-2-hydroxypropyl) ether; BAMGE, bisphenol A monoglycidyl ether; BPF, bisphenol F bis [2, 3 dihydroxypropyl] ether; BFDGE-2H<sub>2</sub>O, bisphenol F bis [2, 3 dihydroxypropyl] ether; BGA, benzoguanamine; BHET, bis-hydroxyethylene terephthalate; BMA, methacrylic acid butyl ester; BP-biphenol; BPA-bisphenol A; BPB, bisphenol B; BPC, bisphenol C; BPE, bisphenol E; BPF, bisphenol F; BPG, bisphenol G; BPS, bisphenol S; BPZ, bisphenol Z; BuOEtOH, butoxyethanol; BuOH, butanol; C, cresol; CAD, charged aerosol detector; CHDM, 1,4-bis(hydroxyl-methyl) cyclohexane; Cl, chemical ionization; COH, 1,4-cyclohexanedimethanol; CYDBADGE, cyclo-di-BADGE; DA, dicarboxylic acid; DBP, dibutyl phthalate; DEHA, diethylhexyl adipate; DEHP, bis(2-ethylhexyl) phthalate; DEG, diethylene glycol; DiBP, diisobutyl phthalate; DPMDI, 4,4'-diphenylmethane diisocyanate; EG ethylene glycol; EI, electron-impact; ELSD, evaporative light scattering detector; ELSO, epoxydised linseed oil; EMA, methacrylic acid ethyl ester; ESI, electrospray ionization; EtOEtOH, ethoxyethanol; F, hemiformaldehyde; GC-FID, gas chromatography-flame ionization detector; GC-MS, gas chromatography-mass spectrometry; GC-TOF-MS, gas chromatography-time of flight-mass spectrometry; HD-1,6-hexanediol; HESI, heated-electrospray ionization; HexOH, hexanol; HPLC-DAD, high-performance liquid chromatography-diode array detector; HPLC-FLD, high-performance liquid chromatography-fluorescence detector; iBMA, methacrylic acid isobutyl ester; IPA, isophthalic acid; IPDI, 1-iso-cyanato-3-isocyanatomethyl-3,5-trimethyl cyclohexane homopolymer; LC-GC, liquid chromatography-gas chromatography; LC-MS/MS, liquid chromatography-tandem mass spectrometry; LC-TOF-MS, liquid chromatography-time of flight-mass spectrometry; M, methylol; m, methylene; MEKO, methyl ethyl ketone oxime; MeOH, methanol; MeOEtOH, methoxyethanol; MMA, methacrylic acid methyl ester; MPD/MBO, 2-methyl-1,3-propanediol; NA, nadic acid; NOGE, novolak glycidyl ether; NPG, neopentyl glycol; NPLC, normal phase liquid chromatography; OA, oleamide; Om, mono-methylene ether; P, phenol; PA, phthalic acid; Ph, phenol; PG, propanediol; PO, polyols; ProH, propanol; PUD, polyester polyurethane diol; SA, sebacic acid; SEC, size exclusion chromatography; SRM, selected reaction monitoring; T-/tBuPhe, tert-butylphenol; TCDDM, tricyclodecanedimethanol; TEG, triethylene glycol; TMA, trimellitic acid; UHPLC-HRMS, ultrahigh performance liquid chromatography-high-resolution mass spectrometry; UVD, ultraviolet detection; m-XDA, m-xylol/enediamine.

### 3.1.1 | Analysis of bisphenols, BADGEs, and derivatives in epoxy resins

High-performance liquid chromatography with fluorescence detection (HPLC–FLD) has proven, since the 90s, to be a sensitive, inexpensive, and valuable technique for the determination of bisphenol-related compounds, BADGEs, and its derivatives from epoxy resins (Paseiro-Losada et al., 1991; Paseiro-Losada et al., 1997; Nerín et al., 2002). Even today, it is still used due to their known advantages (Guo et al., 2020; Lestido-Cardama, Vázquez-Loureiro et al., 2021). As can be inferred from Table 3, in terms of sensitivity, most of the newly published methods present a limit of quantification that allows to detect the analytes at the regulatory level, when it has been established. Chromatographic separation is generally performed on C-18-type columns such as Phenosphere 80 Å ODS, Betasil C-18, Poroshell 120 SB-C18, Fused Core™ Ascentis Express C18, and so on, columns with a particle size < 3 μm considerably shorten analysis time. The gradient elution system usually is composed of water and an organic solvent such as acetonitrile (ACN) or methanol. Recently, Lestido-Cardama, Vázquez-Loureiro et al. (2021) developed a reversed-phase HPLC–FLD method for the simultaneous determination of 13 compounds, including bisphenol analogues and BADGEs in beverage can coatings. The analysis was completed in 28 min. The authors pointed out its appropriateness as a multiresidue method. In another study, Guo et al. (2020) proposed a method for the determination of BADGE and its hydrolyzed and chlorinated derivatives using a column based on superficially porous particles or core–shell particles, which consisted of a solid core and a thin porous shell that surrounds it. This type of column presents higher speed and higher efficiency. The chromatographic conditions, for example, gradient time, composition of the mobile phase, and column temperature, were optimized by using a simulation software (DryLab®). Under the selected conditions, the separation was achieved in 5 min.

Although conventional reversed-phase analytical columns are the most commonly used in HPLC analysis of bisphenols and BADGEs, normal stationary phases have also shown to be suitable. For example, Biedermann and Grob (1998) used a column packed with a cyano phase GromsilCN 2 PR (25 cm × 2 mm i.d., 5 μm) and a mobile phase consisted of a gradient of 20% MTBE/pentane and 50% 1-propanol/20% MTBE/30% pentane to determine oligomers of BADGE and Novolak compounds from epoxy resins and organosols. Fluorescence detection was used at an excitation and emission wavelengths of 225 and 295 nm, respectively. Under these conditions, the limits of detection (LODs) obtained were 10 μg/kg for BADGE and

BFDGE and 300 μg/kg for NOGE. The major component was identified as cyclo-di-BADGE. The authors claimed that the use of normal phase chromatography allows the direct analysis of oily extracts without previous clean-up treatment. A similar method with a slight modification is used by Uematsu et al. (2001) to analyze chlorohydrins of BADGE and BFDGE in canned vegetables and ready-to-drink canned coffee. In the study, 400 μl of the extract was injected on a (25 cm × 4 mm i.d.) column packed with the cyano phase Lichrospher 100 CN. Other studies have also reported the advantage of cyano- and silica-based chromatographic columns to overcome the problem of matrix interference when analyzing BADGE in oily foods (Rauter et al., 1999).

Less frequent is the use of diode array detector for the analysis of these compounds. In a recent study, El-Kosasy et al. (2018) reported an HPLC method with photodiode detection to determine BADGE, BADGE·H<sub>2</sub>O, BADGE·HCl·H<sub>2</sub>O, and BADGE·2HCl. In developing the method, different mobile phase compositions and several stationary phases, namely, Agilent C<sub>18</sub>, Targa C<sub>18</sub>, and Synergi Hydro C<sub>18</sub>, were tested. The separation of BADGE·H<sub>2</sub>O and BADGE·HCl·H<sub>2</sub>O turned to be critical due to the similar chemical structure of both compounds. A binary gradient composed of water and ACN and the Synergi Hydro-RP C18 column (250 × 4.6 mm, 4 μm) were found as the best conditions to achieve a suitable chromatographic separation. The analytes were monitored at a wavelength of 227 nm. The method had enough sensitivity to detect the analytes at a concentration below the specific migration limit (SML) and also provided excellent precision Relative standard deviation (RSD) < 2.52% and good accuracy with recovery values ≥96.31%. Performance parameters are comparable to those reported for fluorescence detection.

Due to its high selectivity, sensitivity, and confirming capability, liquid chromatography coupled to mass spectrometry (HPLC–MS) has gained increasing application in the analysis of epoxy resins (Míguez et al., 2012). With respect to the ionization mode, electrospray ionization (ESI) has been widely used and to a lesser extent atmospheric pressure chemical ionization (APCI). Bisphenols are generally analyzed using ESI in the negative mode that produces the deprotonated molecular ions [M–H]<sup>–</sup> and, on the contrary, BADGEs, BFDGEs (bisphenol F diglycidyl ether), and related compounds give either a weak signal or no signal in negative mode; however, in positive mode they exhibit high tendency to form adducts of [M+Na]<sup>+</sup>, [M+K]<sup>+</sup>, [M+NH<sub>4</sub>]<sup>+</sup>, and [M+ACN]<sup>+</sup> ions (Gallart-Ayala et al., 2011a; Alabi et al., 2014). In two different studies, Míguez et al. (2012) and Gallart-Ayala et al. (2011a) reported the development of an LC–MS/MS

method using an ESI source for the determination of BADGE, BFDGE, and their hydrolyzed and chlorinated derivatives using the adduct of ammonium as a precursor ion. In another study, the same authors explored the fragmentation pathway of BADGEs, BFDGEs, and derivatives by combining multiple-stage mass spectrometry and accurate mass measurements, using ESI in positive and negative modes. Only the hydrolyzed derivatives could be ionized in negative mode. The authors concluded that the fragmentation involves first the loss of one phenylglycidyl ether group followed by the  $\alpha$ -cleavage of the remaining ether group. These fragmentations yield a common product ion at  $m/z$  135 for BADGEs and at  $m/z$  107 for BFDGEs. This last product ion was also observed in the fragmentation pattern of BADGEs as a result of the further loss of the two methyl groups (Gallart-Ayala et al., 2010). In another method reported by Schaefer and Simat (2004), HPLC coupled to an ultraviolet light detector (UVD), FLD, and MS (ESI) detector was used to analyze three different epoxy-based coatings including epoxy anhydride and epoxy phenolic. Beside BADGE and its hydrolyzed derivatives, the authors also identified reaction products of BADGE with chain stoppers (e.g., *tert*-butylphenol), solvents (alcohols and glycols), and phenolic monomers. The chromatographic separation was achieved on a reversed-phase column (Multospher<sup>®</sup> 100 5C18, 250  $\times$  3 mm) and using a gradient composed of 1 mM ammonium formate in water (pH 3) and ACN. In a latter work, Bradley et al. (2008) also identified these compounds in an epoxy-phenolic coating using a time-of-flight mass spectrometer (TOF-MS) in ESI (+) and ESI (-) modes. In addition, the power of TOF-MS enabled the identification of new unknown compounds (e.g., BADGE $\cdot$ HexOH; BADGE $\cdot$ BuOEtOH $\cdot$ HCl); however, it is important to note that there still remain some compounds in the coating extract whose identification was not possible. The chromatographic conditions were slightly different to those reported by Schaefer and Simat (2004); a ZORBAX Eclipse XDB-C18 (100  $\times$  2.1 mm, 3.5  $\mu$ m) column was used as a stationary phase and a gradient consisted of 0.1% aqueous acetic acid and ACN for positive mode and 5 mM ammonium formate at pH 5.5 and 0.1% 5 mM ammonium formate at pH 5.5 in ACN for negative mode, as mobile phase.

LC-APCI-MS was employed by Sendón and Paseiro Losada (2004) to determine BADGE and its hydrolysis and chlorohydroxy derivatives. The analyses were performed in positive and negative mode; thus, BADGE, BADGE $\cdot$ HCl, BADGE $\cdot$ 2HCl, and BADGE $\cdot$ H<sub>2</sub>O were detected in positive mode, while BADGE $\cdot$ H<sub>2</sub>O $\cdot$ HCl and BADGE $\cdot$ 2H<sub>2</sub>O were detected in negative mode. The method showed good linearity and precision and was suitable to quantify the analytes in different samples. In a recent study published by Lestido-Cardama et al.

(2019), LC-APCI-MS in positive and negative modes was used for the simultaneous determination of BADGEs and bisphenol analogues. The proposed method presented low LODs and could be a useful tool for confirmation purposes.

Bisphenols and BADGEs have also been analyzed using GC-MS. With these approaches, the bisphenol analysis generally involves a derivatization step, although not rigorously required, which is suggested in order to increase the volatility of the compounds and improve the sensitivity of the method. Different derivatization agents have been used (Cacho et al., 2012; Gallart-Ayala et al., 2011b; Alabi et al., 2014). Cacho et al. (2012) investigated two derivatization reactions namely *in situ* acetylation with acetic anhydride and *in tube* silylation with BSTFA (bis(trimethylsilyl)trifluoroacetamide) coupled to SBSE (stir bar sorptive extraction). The authors concluded that the derivatization increased markedly the sensitivity compared with the underivatized compounds, and on the other hand, a higher sensitivity was achieved for the acyl compared to silyl compounds. The derivatizing agent BSTFA was also employed by Cottier et al. (1997) to determine BADGE, BFDGE, and their derivatives from a vinylic organosol varnish using GC-MS after the silylation reaction.

### 3.1.2 | Analysis of polyester monomers and oligomers

Different analytical approaches to determine monomers and oligomers migrating from polyester-based coatings have been reported in the literature. For instance, Schaefer et al. (2004) developed an analytical methodology to identify and quantify migrants from polyester-urethane and polyester-phenolic coatings. After the hydrolysis of polyester extracts into their monomers, that is, polyols and polyvalent carboxylic acids, they were analyzed by gas chromatography-flame ionization detector (GC-FID) and by HPLC-ESI-MS/UVD, respectively. The cyclic oligoesters were determined by HPLC-ESI-MS/UVD and high-performance liquid chromatography-evaporative light scattering HPLC-ELSD/(UV), and their quantification was performed by using the correlation between the amount of the benzoic rings (chromophore) and the UV response using a calibration obtained with commercial standards. Regarding the ELSD, the authors concluded that it overestimates the amount of oligomers with a higher molecular weight than the cyclic oligoester formed by three molecules of isophthalic acid and three molecules of ethylene glycol. A similar approach was suggested by Eckardt et al. (2020). Pietropaolo et al. (2018) investigated the oligomers in two different



polyester resins; one of them was synthesized by condensation of isophthalic acid and terephthalic acid with 1,6-hexanediol and neopentylglycol, and for the second isophthalic acid, terephthalic acid, 1,6-hexanediol, and 2-methylpropanediol were used as monomers. The analysis of the migrating extracts by HPLC–ESI–MS disclosed the presence of several cyclic and linear oligomers. In the samples analyzed, cyclic oligomers predominated compared to linear ones. For a confident identification, selected oligomers, precisely four linear and 11 cyclic oligomers, were synthesized and characterized by  $^1\text{H}$  and  $^{13}\text{C}$  NMR spectroscopy. These synthesized standards were used to quantify the oligomers in the resin samples by micro-HPLC–HRMS. The method was validated, and the authors reported good linearity ( $R^2 > .995$ ), satisfactory sensitivity with LODs ranging from 0.003 to 0.30 ng/ml, and suitable repeatabilities (intraday RSD [ $n = 7$ ]  $< 8.2\%$  and interday RSD [ $n = 35$ ]  $< 10\%$ ); the authors also highlighted the synthesis of standards to allow an exact quantification.

Paseiro-Cerrato, MacMahon et al. (2016) reported the use of several techniques, namely, FTIR, GC–MS, HPLC–DAD/MS, HPLC–DAD/CAD (charged aerosol detector), and UHPLC–HRMS, for the characterization and analysis of compounds from polyester coatings. First, an FTIR analysis was performed to identify the polymeric coating. In a second step, resins were extracted with two different solvents (ACN and ethyl acetate) and the resulting extracts were later analyzed by different analytical techniques. Several volatile and semivolatile compounds were detected by GC–MS; however, no good library matches were observed for none of them. Nonvolatile compounds were analyzed by HPLC–DAD/MS, HPLC–DAD/CAD, and UHPLC–HRMS. Twenty-nine oligomers were identified, of which 22 were linear oligomers and seven cyclic. Most of them were detected in positive mode and only six of the identified oligomers exhibited higher intensity in negative than in positive mode. One of the detected compounds, specifically 3-(((4-(hydroxymethyl)cyclohexyl)methoxy)carbonyl) bicyclo[2.2.1]hept-5-ene-2-carboxylic acid (Na+COH), was synthesized and used as standard to confirm the identification. Finally, in an attempt to simplify the experimental procedure, direct analysis in real time coupled to high-resolution mass spectrometry (DART–HRMS) was applied to detect directly the previously identified compounds. Promising results were reported with this latest analytical technique; 23 of the 29 oligomers identified by UHPLC–HRMS were detected directly in the can coating in a very short time of analysis and without sample preparation.

Bradley et al. (2009) also proposed the use of several techniques, specifically FTIR, HS–GC–MS, and LC–TOF–MS, for the characterization of polyester–polyurethane can coatings. The authors mentioned that only two among the

detected semivolatile compounds presented acceptable library matches, butyl octadecanoate, and 2-[2-[4-(1,1,3,3-tetramethylbutyl)-phenoxy]ethoxy]ethanol. Regarding nonvolatile substances, one linear and 12 cyclic oligomers, with a molecular weight below 800, were identified. In this regard, it is worth commenting that in some cases assigning properly the identity of the chromatographic peaks can be extremely difficult such as in the case of oligomers with the same formula but different identity. In a later work, the same authors used gas chromatography coupled to time-of-flight mass spectrometry (GC–TOF–MS) to investigate nadic anhydrides in a polyester coating (Driffield et al., 2018). Ubeda, Aznar, and Nerín (2019) proposed the use of both GC–MS and APGC–QTOF for a complete characterization of volatile and semivolatile compounds in a biopolymer based on PLA and polyester.

Other interesting strategy for the elucidation of oligomers migrating from polyester–polyurethane coatings was developed by Omer et al. (2018). The strategy was based on an untargeted approach using LC–HRMS to characterize the coatings, the data generated were processed using a specific data processing (open-source R-environment: *xcms* and *CAMERA* packages), and the identification of the detected compounds was performed by comparing with a homemade database based on the predicted polyester oligomer combinations. Furthermore, to increase the level of confidence in the identification, the chromatographic behavior as well as the fragmentation pattern were considered. The authors reported the presence of 14 and 17 cyclic oligomers in the two lacquers studied, respectively, and also suggested unpredicted migrants, for example, caprolactam oligomers. In line with this, Ubeda et al. (2017) detected caprolactam oligomers (dimer, trimer, tetramer, pentamer) in the migration extract from a multilayer film (PET/PA/PPP).

Recently, confocal Raman microscopy has been reported as a valuable tool to identify the polymeric coatings. The technique allows a complete characterization of the coating by imaging the different layers. The novel approach has been successfully applied to characterize polyester-based coatings for food contact (Lestido-Cardama et al., 2022).

### 3.1.3 | Analysis of acrylic monomers and other coatings components

There are very few works in the literature reporting analytical methods for the determination of components released from acrylic based resins. Franz and Brandsch (2013) developed a method based on GC–FID for the analysis of acrylic monomers in acrylic polymer resins. In the

study, the authors determined acrylic acid methyl ester, methacrylic acid ethyl ester, methacrylic acid butyl ester, and methacrylic acid isobutyl ester in four acrylic resins. Samples were analyzed by multiple headspace extraction sampling with six sequential extractions before the GC–FID determination. Quantification was performed by the external standard method and appropriate linearity ( $R^2 > .998$ ) was reported. The release of the monomers in different food simulants was also investigated and, in general, low diffusion coefficients were obtained compared with other polymers. More recently, Paseiro-Cerrato et al. (2017) investigated potential migrants from an acrylic–phenolic coating using HPLC coupled to different detectors, for example, DAD, CAD, MS, and HRMS. Several migrants including BPA, BADGE·2H<sub>2</sub>O, and BGA (benzoguanamine) and seven unknown compounds that showed good response with DAD were detected in ACN extracts of acrylic–phenolic can coating as well as in food simulants (aqueous food simulants, 50% ethanol [v/v]). However, the authors indicated that it was not possible to identify acrylic–phenolic derivatives. Mainly, higher concentrations of migrants in food simulants than in ACN extracts were found.

### 3.1.4 | Analysis of phenolic resins components

Phenolic resins or resoles are complex mixtures commonly employed as a cross-linkers for other resins. Due to the complexity of its nature, they are difficult to characterize, and it is to be expected that different techniques will be needed to address the study of their composition. An analytical methodology that involves the use of a battery of instrumental techniques, including GC–MS, comprehensive two-dimensional GC (GC×GC), normal-phase high-performance liquid chromatography (NPLC), and size exclusion chromatography (SEC), for the determination of resole components has been proposed by Biedermann and Grob (2006a). Thus, GC–MS was used as a very useful tool in terms of identification of chromatographic peaks; GC×GC provided additional information, an improved resolution, and also allowed to group peaks belonging to the same type of compounds. NPLC was used as a complementary technique, namely, as a pre-separation step of samples before GC analysis, and SEC provided information about the distribution of the molecular weight of the compounds in the mixture. The method was applied to characterize a commercial resole based on phenol. Samples were trimethyl silylated before the GC analysis. Monophenolic, bisphenols, and polyphenols resole components were identified. The authors highlighted the valuable information provided by each technique for a

complete analysis. In the second part of the work (Biedermann & Grob, 2006b), the methodology was applied to two resoles based on cresol/phenol mixtures and a resole manufactured from *tert*-butylphenol. Resole components including mono-cresols, mono-*tert*-butylphenols, cresol-phenols, bis-cresols, bis-*tert*-butylphenols, and three-ring compounds were identified. The resole obtained from *tert*-butylphenol was mainly composed by components with one to three rings.

## 3.2 | Determination of migrants transferred from coatings in foods

The analysis of the migrants in the foodstuff usually involves the extraction of the analytes from the matrix prior to the chromatographic analysis.

Several sample preparation treatments have been used for bisphenols, BADGE, and derivatives, as reported in Table 4; solvent extraction and solid-phase extraction (SPE) are the most predominant. Other extraction techniques including solid-phase microextraction (SPME), on-line SPE, supramolecular solvent-based microextraction (SUPRASS), Quick, Easy, Cheap, Effective, Rugged, & Safe (QuEChERS), and molecularly imprinted polymers, among others, have been also applied. The SUPRAS-based microextraction has demonstrated to be a suitable technique to extract bisphenols and BADGEs from a diversity of canned foods including vegetables, fish, and meat products. Several variables, namely, solvent composition and volume, pH, and time, were tested during the optimization of SUPRAS extraction. Tetradecanol-based SUPRAS was reported to be the most suitable and a good extraction efficiency for the target analytes was achieved (Alabi et al., 2014). An interesting alternative to the conventional SPE is the molecularly imprinted polymers-solid phase extraction (MISPE). Due to its inherent selectivity, it generally provides greater effectiveness. MISPE cartridges have been successfully applied for the purification of bisphenols and BADGEs from canned beers and energy drinks (Cirillo et al., 2019; Gallo et al., 2017). A new single-step extraction and clean-up method based on alkylcarboxylate-coated magnetic nanoparticles to extract BPA from soft drinks has been proposed by Yazdinezhad et al. (2013). The authors highlighted that the method was rapid and simple and showed lower solvent consumption compared to other conventional procedures. In the case of polyester monomers and oligomers, there are very few examples in the literature. The extraction procedures reported comprise a solvent extraction followed by a clean-up step with SPE or QuEChERS (Paseiro-Cerrato et al., 2019a; Driffield et al., 2018).

TABLE 4 Migration values of chemical components from can coatings into food

Coating	Migrant	Food	Extraction procedure	Analytical technique	Migration results (Restrictions)	Reference
Polyester coating	TA, IPA, NA, and oligomers (PA+2MPD, PA+CHDM+MPD)	Chicken noodle soup, chicken cream, sauerkraut, mushrooms, green beans, tomatoes, pumpkins, coconut milk, and chicken rice no salt	Solvent extraction (ACN with 0.1% formic acid) and SPE	UFLC-MS/MS for monomers, and LC-DAD for oligomers UHPLC-HRMS for confirmation	<sup>a</sup> IPA: 5.1–69 µg/kg; TA: nd–61 µg/kg; NA: <LOD–21 µg/kg; PA+2MPD: nd–324 µg/kg; PA+CHDM+MPD: nd–454 µg/kg	Paseiro-Cerrato et al. (2019a)
Polyester can coatings based on NAH	PA, IPA, NA isomers (cis-endo-NA, trans-endo-exo-NA), NAH isomers (endo-NAH, exo-NAH), and oligomers <1000 Da	Green beans, tomato sauce, diced tomatoes, wax beans, peas, whole kernel corn, crema style corn, corn, spilt pea and ham soup, chicken broth, pumpkin, refried beans, potatoes, pork and beans, black beans, ketchup, and jalapeno peppers	Solvent extraction (ACN) (oligomers) and QuEChERS (monomers)	LC-MS/MS for monomers, GC-TOF-MS for NAHs, and LC-TOF-MS for oligomers	<sup>a</sup> IPA: <20 µg/kg; PA: <20 µg/kg; cis-endo-NA: <0.01–17 µg/kg; trans-endo-exo-NA: <0.02–2.2 µg/kg; <sup>d</sup> endo-NAH: <0.77–42 µg/kg; <sup>d</sup> exo-NAH: <0.78–4.6 µg/kg; oligomers (<1000 Da): <LOD–943 µg/kg	Driffield et al. (2018)
Epoxy resin	BPA, BPF, BPE, BPB, BPS	Soft drinks (soda, beer, cola, tea, and energy drinks)	On-line SPE	LC-MS/MS	<sup>a</sup> BPA: nd–607 ng/L; BPF: nd–218 ng/L; BPS, BPE, BPB: nd	Gallart-Ayala et al. (2011b)

(Continues)

TABLE 4 (Continued)

Coating	Migrant	Food	Extraction procedure	Analytical technique	Migration results (Restrictions)	Reference
Epoxy resin	BADGE.2H <sub>2</sub> O, BPF, BPE, BPA, BADGE.H <sub>2</sub> O.HCl, BADGE.H <sub>2</sub> O, BPB, BADGE.2HCl, BADGE.HCl, BADGE, BFDGE, BFDGE.2HCl	Vegetables (mushroom, red pepper, olive, green beans, asparagus), legumes (chickpeas, lentils), fruits (pineapple, peach), fish and other seafood (mackerel, mussels, tuna, cockles), meat products (tripe, meat ball), and grains (sweet corn)	Microextraction with a SUPRASS made up of inverse aggregates of tetradecanol	HPLC-FLD	<sup>b</sup> BADGE.2H <sub>2</sub> O: nd-959 µg/kg; BPF, BPE: nd; <sup>a</sup> BPA: nd-241 µg/kg; BADGE.H <sub>2</sub> O.HCl: nd-533 µg/kg; BADGE.H <sub>2</sub> O: nd-179 µg/kg; BPB: nd-40 µg/kg; BADGE.2HCl: nd-215 µg/kg; BADGE.HCl: nd-21 µg/kg; BADGE: nd-7.1 µg/kg; BFDGE: nd-314 µg/kg; BFDGE.2HCl: nd-120 µg/kg	Alabi et al. (2014)
Epoxy resin	BPA, BPB	Vegetables (maize, green beans, peas, mushroom, soybean, tomato, spinach, and carrot) and fruits (pineapple, peaches, lychees, pears, mango, papaya, passion, and fruit salad)	QuEChERS and DLLME using ACN as dispersive solvent, tetrachloroethylene as extractive solvent, and anhydride acetic as derivatizing reagent	GC-MS	Fruits <sup>a</sup> BPA: nd-10.2 µg/kg BPB: nd-3.4 µg/kg Vegetables BPA: nd-265.6 µg/kg BPB: nd-3 µg/kg	Cunha and Fernandes (2013)
Epoxy resin	BPA, BPB, BPF, BADGE, and BFDGE	Energy drinks	MIP (Affinimip <sup>®</sup> BPA SPE cartridge)	UPLC-FLD	<sup>a</sup> BPA: <LOQ-3.3 ng/ml; BPB: <LOQ; BPF: <LOQ-1.3 ng/ml; <sup>b</sup> BADGE: <LOQ-19.4 ng/ml; BFDGE: <LOQ-0.6 ng/ml	Gallo et al. (2017)

(Continues)



TABLE 4 (Continued)

Coating	Migrant	Food	Extraction procedure	Analytical technique	Migration results (Restrictions)	Reference
BPA-based epoxy resin	BPA, BPF, BADGE, BADGE.2H <sub>2</sub> O, BADGE.H <sub>2</sub> O, BADGE.HCl.H <sub>2</sub> O, BADGE.2HCl, and BADGE.HCl	Beer (Pilsner, wheat beer, black beer, and others)	LLE	LC-MS/MS	<sup>a</sup> BPA: <0.1–2.54 µg/L; BPF: nd–0.3 µg/L; <sup>b</sup> BADGE: nd–<0.05 µg/L; BADGE.2H <sub>2</sub> O: <0.5–14.3 µg/L; BADGE.H <sub>2</sub> O: nd; BADGE.HCl.H <sub>2</sub> O: nd–0.65 µg/L; BADGE.2HCl: nd–0.55 µg/L; BADGE.HCl: nd–<0.1 µg/L	Zech et al. (2015)
Epoxy resin	BADGE, BADGE.H <sub>2</sub> O, BADGE.2H <sub>2</sub> O, BADGE.HCl, BADGE.2HCl, BADGE.HCl.H <sub>2</sub> O, BFDGE, BFDGE.2HCl and BFDGE.2H <sub>2</sub> O	Fish products (mussels, European anchovy, sardines, mackerels, and tuna)	Solvent extraction (1:1, ACN-hexane)	LC-MS/MS	<sup>b</sup> BADGE, BADGE.HCl, BADGE.2HCl, BFDGE, BFDGE.2HCl, BFDGE.2H <sub>2</sub> O: nd; BADGE.H <sub>2</sub> O: nd–120 µg/kg; BADGE.2H <sub>2</sub> O: nd–625 µg/kg; BADGE.HCl.H <sub>2</sub> O: nd–87 µg/kg	Miguez et al. (2012)
Amino-based cross-linking resins	Melamine	Three food types (acidic, fatty, and fish/meat)	Solvent extraction (ACN and water)	LC-MS/MS	<sup>c</sup> Melamine: <23–152 µg/kg	Bradley et al. (2011)
Epoxy resin	BPA	Beverages, fruits, vegetables, and fat-containing foodstuffs	Sol-gel immunoaffinity chromatography	HPLC-FLD	<sup>a</sup> BPA: nd–43 ng/g in tuna	Braunrath et al. (2005)
Epoxy resin	BPA	Pineapple, tuna, and mushrooms	MAE and MISPE	LC-ESI/MS	<sup>a</sup> BPA: 7.3 ng/g in pineapple to 42.3 ng/g in tuna	Maragou et al. (2020)

(Continues)

TABLE 4 (Continued)

Coating	Migrant	Food	Extraction procedure	Analytical technique	Migration results (Restrictions)	Reference
Epoxy-phenolic and organosol coatings	BADGE and BFDGE	Seafood products (sardines, tuna, mackerel, mussels, cod) and covering sauces	Solvent extraction: <i>n</i> -heptane and ACN 90%	HPLC-FLD	The highest values were 0.34 mg/kg for <sup>b</sup> BADGE and 0.74 mg/kg for BFDGE in mackerel with red pepper sauce	Cabado et al. (2008)
Epoxy lacquers	BPA, BPF, BPZ, and BP	Beverages and filling liquids of canned vegetables	Derivatization and SBSE	TD-GC-MS	Soft drinks: <sup>a</sup> BPA: nd-0.68 µg/L; BPF: nd-0.26 µg/L; BPZ: nd-0.09 µg/L; BP: nd Filling liquids: BPA: nd-13.98 µg/L; BPF: nd-7.07 µg/L; BPZ: nd-0.92 µg/L; BP: nd-0.58 µg/L	Cacho et al. (2012)
Epoxy resin	BADGE, BADGE.H <sub>2</sub> O, BADGE.2H <sub>2</sub> O, BADGE.HCl, BADGE.2HCl, BADGE.HCl.H <sub>2</sub> O, BFDGE, BFDGE.2H <sub>2</sub> O, and BFDGE.2HCl	Soft drinks and aqueous-based canned foods (fruits and vegetables)	Soft drinks: SPE; canned food: ethyl acetate	LC-MS/MS	<sup>b</sup> BADGE, BFDGE, BFDGE.2H <sub>2</sub> O, BFDGE.2HCl: nd; BADGE.H <sub>2</sub> O: nd-53 µg/kg; BADGE.2H <sub>2</sub> O: 2.1-675 µg/kg; BADGE.HCl: nd-11 µg/kg; BADGE.2HCl: nd-2.8 µg/kg; BADGE.HCl.H <sub>2</sub> O: nd-274 µg/kg	Gallart-Ayala et al. (2011a)
Organosol and epoxy can coatings	BPA	Tuna fish	Solvent extraction (MeOH, hexane and ACN)	RP-HPLC-FLD	<sup>a</sup> BPA: <LOQ to 102.7 µg/kg	Munguia-López et al. (2005)

(Continues)

TABLE 4 (Continued)

Coating	Migrant	Food	Extraction procedure	Analytical technique	Migration results (Restrictions)	Reference
PVC organosol	BADGE.2HCl, BFDGE.2HCl, BADGE.2H <sub>2</sub> O, BFDGE.2H <sub>2</sub> O, BADGE.HCl.H <sub>2</sub> O, and BFDGE.HCl.H <sub>2</sub> O	Ready-to-drink canned coffee and canned vegetables (corn, tomatoes, and others)	Solvent extraction (15% DCM/pentane)	HPLC-FLD and GC-MS for confirmation	<sup>b</sup> BADGE.2HCl: <0.02–0.14 mg/kg; BFDGE.2HCl: <0.04–0.42 mg/kg; BADGE.2H <sub>2</sub> O: nd–4.03 mg/kg; BFDGE.2H <sub>2</sub> O: nd–0.7 mg/kg; BADGE.HCl.H <sub>2</sub> O: nd–0.48 mg/kg; BFDGE.HCl.H <sub>2</sub> O: nd–1.08 mg/kg	Uematsu et al. (2001)
Epoxy phenolic coating	BPA	Canned foods (green beans, tomatoes, peas, pineapple, ravioli, chili, fish, soups, etc.) separating the solid food from the liquid supernatant	Solvent extraction (ACN)	HPLC-MS/MS	<sup>a</sup> BPA: <2–790 ng/g, showing that BPA concentrations are higher in the solid food	Noonan et al. (2011)
Epoxy resins, organosol, and polyester coatings	Cyclo-di-BADGE	2010: canned fish 2012: canned fish in oil and meat, chocolate cream, soup	Solvent extraction (EtOH and hexane)	HPLC-FLD	CYDBADGE: (2010) <25–1980 µg/kg (2012) <25–2600 µg/kg	Biedermann et al. (2013)
Epoxy-based lacquers	BPA	Vegetables, fruit, fish, soup and sauces, meat, spaghetti and baked beans, infant foods, beverages, and miscellaneous	Solvent extraction (ACN or ACN- trimethylpentane for samples with >1% fat) and derivatization (acetic anhydride)	GC-MS	<sup>a</sup> BPA: <10–192 µg/kg	Thomson and Grounds (2005)

(Continues)

TABLE 4 (Continued)

Coating	Migrant	Food	Extraction procedure	Analytical technique	Migration results (Restrictions)	Reference
Epoxy-based coating	BPA	Canned tomatoes (peeled, cherry, and concentrated paste)	SPE	HPLC-DAD-FLD	<sup>a</sup> BPA: Mean concentrations range: 0.31–3.80 µg/kg in peeled tomatoes, 0.90–2.21 µg/kg in cherry tomatoes, 0.39–1.82 µg/kg in concentrated tomato paste	Errico et al. (2014)
Epoxy can coatings	BPA	Jalapeño peppers	Solvent extraction (MeOH)	HPLC-FLD and confirmation by GC-MS	<sup>a</sup> BPA: <LOQ–5.59 µg/kg	Munguía-López et al. (2002)
Epoxy resin	BFDGE	Fish in oil (sardine, tuna, anchovy, mackerel, mussel, oyster, etc.)	LLE (hexane and ACN)	HPLC-FLD and confirmation by GC-MS	<sup>b</sup> BFDGE: nd–4.2 mg/kg	Theobald et al. (2000)
Epoxy-phenolic coating	BPA, BPF, BADGE, BADGE.2H <sub>2</sub> O, BADGE.2HCl, BFDGE, BFDGE.2H <sub>2</sub> O, and BFDGE.2HCl	Vegetables including fava beans, red beans, chickpeas, and okra	Solvent extraction (MeOH) and SPE	UHPLC-FLD and UHPLC-TOF-MS for confirmation	BPF, BADGE, BADGE.2HCl, BFDGE; BADGE.2HCl, BFDGE.2H <sub>2</sub> O, and BFDGE.2HCl: nd <sup>a</sup> BPA: 12.8–54.6 µg/kg and BADGE.2H <sub>2</sub> O: 101–146 µg/kg were confirmed by MS	El Moussawi et al. (2019)
Gold epoxy-phenolic lacquer or BADGE enamel	BPA, BPB, BADGE, BFDGE	Tuna fish (in oil or aqueous medium)	Tuna: solvent extraction ( <i>n</i> -hexane/ACN) Oil: solvent extraction ( <i>n</i> -hexane) and SPE Aqueous medium: directly	LC-FLD and confirmation by LC-MS/MS	Tuna <sup>a</sup> BPA: nd–187 ng/g; BPB: nd–145.9 ng/g; <sup>b</sup> BADGE: nd–91.1 ng/g; BFDGE: nd–38.5 ng/g Medium BPA: nd–16.2 ng/ml; BADGE: nd–66.9 ng/ml; BPB, BFDGE: nd	Fattore et al. (2015)

(Continues)

TABLE 4 (Continued)

Coating	Migrant	Food	Extraction procedure	Analytical technique	Migration results (Restrictions)	Reference
Epoxy-based coating	BPA	Canned foodstuffs (peas, rice pudding, toffee pudding, beans, spaghetti Bolognese, etc.)	Solvent extraction ( <i>n</i> -heptane and ACN) and derivatization	GC-MS	<sup>a</sup> BPA: 8.6–129.5 µg/kg	Goodson et al. (2004)
Epoxy lacquers	BPA, BPS, and BP	Peas, peas with carrots, sweet corn, artichoke, mushroom, bean shoot, and mixed vegetables	SPME and derivatization (acetic anhydride or BSTFA)	GC-MS	Food <sup>a</sup> BPA: nd–77.7 ng/g; BPS: nd–36.1 ng/g; BP: nd–40.1 ng/g Supermatant BPA: 11.7–321 ng/ml; BPS: nd–175 ng/ml; BP: 17.2–365 ng/ml	Viñas et al. (2010)
Epoxy-phenolic and organosol coatings	BADGE	Tuna, sardines, anchovies, mackerel, corned beef, ham/chopped ham and pork, pork luncheon meat, spreads, milk, infant formulae	Solvent extraction (DCM, hexane and ACN) and SPE	HPLC-FLD and confirmation by GC-MS	<sup>b</sup> BADGE <0.02–62 mg/kg	Summerfield et al. (1998)
Epoxy resin	BPA and BADGE	Fish, meats, fruits and vegetables, soups and sauces, coconut milk	Solvent extraction (ACN, water) and SPE	LC/ECD or LC/MS, LC/MS/MS for confirmation	<sup>a</sup> BPA: nd–842 ng/g; BADGE: nd	Sajiki et al. (2007)
Epoxy resin	BPA, BPB, BPF, BADGE, and BFDGE	Beers	SPE (MIPs)	UPLC-FLD	<sup>a</sup> BPA: <LOQ–0.8 ng/ml; BPF: <LOQ–2.5 ng/ml; BADGE: <LOQ–1.2 ng/ml; BPB and BFDGE: <LOQ	Cirillo et al. (2019)

(Continues)

TABLE 4 (Continued)

Coating	Migrant	Food	Extraction procedure	Analytical technique	Migration results (Restrictions)	Reference
Organosol (PVC type), epoxy lacquers	BADGE, BADGE.H <sub>2</sub> O, BADGE.2H <sub>2</sub> O, BADGE.HCl, BADGE.2HCl	Fish in oil, in tomato/dill sauce, or in vinegar, vegetables, pasties, and meat sauce	Solvent extraction (ACN:hexane and ethyl acetate)	HPLC-FLD	<sup>b</sup> BADGE was found in concentrations up to 5.1 mg/kg, BADGE.2H <sub>2</sub> O up to 2.6 mg/kg, BADGE.H <sub>2</sub> O was not detected, BADGE.HCl up to 2.4 mg/kg and BADGE.2HCl up to 8.3 mg/kg	Hammarling et al. (2000)
Epoxy resin	BPA	Soft drinks	Hemicellular magnetic SPE	LC-MS/MS	<sup>a</sup> BPA: 0.066–1.08 ng/ml	Yazdinezhad et al. (2013)

Abbreviations: ACN, acetonitrile; BADGE, bisphenol A diglycidyl ether; BADGE.H<sub>2</sub>O, bisphenol A (2,3-dihydroxypropyl) glycidyl ether; BADGE.2H<sub>2</sub>O, bisphenol A bis(2,3-dihydroxypropyl) ether; BADGE.HCl, bisphenol A (3-chloro-2-hydroxypropyl) glycidyl ether; BADGE.2HCl, bisphenol A bis(3-chloro-2-hydroxypropyl) ether; BADGE.H<sub>2</sub>OHCl, bisphenol A (3-chloro-2-hydroxypropyl) (2,3-dihydroxypropyl) ether; BFDGE, bisphenol F diglycidyl ether; BFDGE.2HCl, bisphenol F bis [3 chloro-2 hydroxypropyl] ether; BFDGE.2H<sub>2</sub>O, bisphenol F bis [2, 3 dihydroxypropyl] ether; BFDGE.H<sub>2</sub>O.HCl, *p,p* bisphenol F (3-chloro-2-hydroxypropyl) ether; BP, bisphenol; BPA, bisphenol A; BPB, bisphenol B; BPE, bisphenol E; BPF, bisphenol F; BPS, bisphenol S; BPZ, bisphenol Z; BSTFA, N,O-bis(trimethylsilyl)trifluoroacetamide; CHDM, 1,4-cyclohexanedimethanol; CYDBADGE, cyclo-di-BADGE; DLLME, dispersive liquid-liquid microextraction; ECD, electrochemical detection; ESI, electrospray ionization; GC-MS, gas chromatography-mass spectrometry; GC-TOF-MS, gas chromatography-time of flight-mass spectrometry; HPLC-FLD, high-performance liquid chromatography-fluorescence detector; IPA, isophthalic acid; MISPE, molecularly imprinted polymer-solid-phase extraction; LC-DAD, liquid chromatography-diode array detector; LC-MS/MS, liquid chromatography-tandem mass spectrometry; LC-TOF-MS, liquid chromatography-time of flight-mass spectrometry; LLE, liquid-liquid extraction; LOD, limit of detection; LOQ, limit of quantification; MAE, microwave assisted extraction; MIP, molecularly imprinted polymer; MPD, 2-methyl-1,3-propanediol; NA, nadic acid; NAH, nadic anhydride; nd, not detect; PA, phthalic acid; PVC, polyvinyl chloride; QuEChERS, Quick, Easy, Cheap, Effective, Rugged, & Safe; SBSE, stir bar sorptive extraction; SPE, solid-phase extraction; SPME, solid-phase microextraction; SUPRAS, supramolecular solvent; TA, terephthalic acid; TD-GC-MS, thermal desorption-gas chromatography-mass spectrometry; UFLC-MS/MS, ultrafast liquid chromatography-tandem mass spectrometry; UHPLC-HRMS, ultrahigh-performance liquid chromatography-high-resolution mass spectrometry.

<sup>a</sup>Regulation (EU) No. 10/2011 Specific Migration Limit (SML) in plastic materials; SML BPA 0.6 mg/kg; SML BPS 0.05 mg/kg; SML IPA 5 mg/kg; SML TA 7.5 mg/kg; SML melamine 30 mg/kg. Regulation (EU) 2018/213 SML BPA 0.05 mg/kg.

<sup>b</sup>Regulation (EC) No. 1895/2005 Sum of the migrations of BADGE+BADGE.H<sub>2</sub>O+BADGE.2H<sub>2</sub>O 9 mg/kg; BADGE.HCl+BADGE.2HCl+BADGE.H<sub>2</sub>O.HCl 1 mg/kg. BFDGE "The use and/or presence in the manufacture of materials and articles are prohibited."

Commission Directive 2002/16/EC: Sum of the migration levels of BADGE+BADGE.H<sub>2</sub>O+BADGE.HCl+BADGE.2HCl+BADGE.2HCl+BADGE.H<sub>2</sub>O.HCl 1 mg/kg. Sum of the migration levels of BFDGE+BFDDGE.H<sub>2</sub>O+BFDDGE.HCl+BFDDGE.2HCl+BFDDGE.H<sub>2</sub>O.HCl 1 mg/kg. No longer in force, date of end of validity: December 31, 2005.

<sup>c</sup>Commission Regulation (EU) No. 1282/2011 SML melamine 2.5 mg/kg.

<sup>d</sup>EFSA CEF Panel (EFSA Panel on Food Contact Materials, Enzymes, Flavorings, and Processing Aids), 2014. Scientific Opinion on the safety assessment of the substance, 5-norbornene-2,3-dicarboxylic anhydride, CAS No 826-62-0, for use in food contact materials. Migration of 5-norbornene-2,3-dicarboxylic anhydride and its hydrolysis products should in total not exceed 50 µg/kg food.

## 4 | MIGRATION OF CHEMICALS FROM CAN COATINGS

Components of the can coatings can migrate into the food and may adversely affect its quality and safety. In order to assess the safety of the coatings and to estimate the exposure to substances migrating into the food, migration tests into foodstuffs or food simulants should be performed.

The migration is a predictable phenomenon that usually obeys Fick's laws of diffusion. The key parameters that define the migration kinetics are the partition coefficient and the diffusion coefficient. The partition coefficient ( $K_{P/F}$ ) indicates the relative solubility of the migrant between the polymer and the food when the equilibrium is reached, and thus  $K_{P/F} > 1$  indicates that the migrant is found in higher concentrations in the polymer than in the food. And the diffusion coefficient describes the kinetics of the migration process (Tehrany & Desobry, 2004; Castle, 2007; Maia et al., 2016; Nerín, 2016; Canellas et al., 2010; Rodríguez-Bernaldo de Quirós et al., 2019).

Migration of chemicals from food packaging materials is affected by several factors. The migrant (size/shape), the thickness of the film, the degree of cross-linking, the time of exposure, and, most crucial, the temperature are some of the factors reported by Wagner et al. (2018) that influence the migration from an epoxy-phenolic can coating. The results of the study showed that the apparent diffusion coefficients decreased as the cross-link density increased, whereas they increased as the film thickness increased. Other important factors that also affect the migration are the nature of food or food simulant as well as the type of contact (Castle, 2007). Some of these parameters have been evaluated in different studies. For example, Cabado et al. (2008) studied the migration of BADGE and BFDGE in canned seafood with various covering sauces and manufactured under different conditions, that is, different sterilization conditions (115°C/45 min; 121°C/30 min) and different time and temperature storage conditions. It was found, on one hand, that the migration of BADGE and BFDGE depended on the specie of the seafood and the composition of the sauce; thus, migration occurs to a greater extent in fatty foods. The highest values of BADGE and BFDGE were found in mackerel-red pepper sauce, which was the highest fat content. However, it was not affected by the sterilization conditions. On the other hand, the BFDGE migration also depended on the storage time; the migration levels after 12 and 18 months of storage were significantly different than those found at 6 months. Munguía-López et al. (2005) reported a marked effect of the heating process on the migration of BPA into a fatty food simulant. However, in a previous study, the same authors found a minimal effect of heat treatment on BPA migra-

tion into an acidic food simulant. In another work, Errico et al. (2014) reported an increase in the migration of BPA in canned tomatoes with the storage temperature and also observed higher migration levels in damaged cans compared to intact ones. In the opposite way, Goodson et al. (2004) found no appreciable effect on BPA migration in damaged cans. The authors also investigated the effect of storage and concluded that about 80%–100% of the BPA present in the coating migrated during the can processing stage and this level was not increased during the storage (9 months).

In a series of studies reported in the literature, long-term migration tests were evaluated. Paseiro-Cerrato, Noonan et al. (2016) investigated the migration of polyester monomers and oligomers from polyester coatings into food simulants at 40°C for 515 days. The authors reported an increase in the concentration of the monitored migrants during the assay, particularly in ethanol simulants, and also concluded that 3% acetic acid (w/v) is not an appropriate simulant because of rusting. In a later work, the same authors evaluated the migration from two different coatings, an epoxy and an acrylic-phenolic resin, and several migrants such as BPA, BADGE, BADGE derivatives, and so on were monitored. For water, no variation in concentration during the test was observed; in the case of 50% ethanol (v/v), an increase in concentration was observed until equilibrium was reached (90–305 days). This increase occurred after 10 days, which is the time conventionally used in migration tests (Paseiro-Cerrato et al., 2017). Subsequently, the authors carried out the tests under accelerated conditions, and concluded that the migration tests performed at 60°C in short periods of time (4 h to 30 days) may simulate migration tests performed at 40°C over long periods of time (1.5 years) (Paseiro-Cerrato et al., 2019b).

In order to reduce the migration from polymeric coatings, newer materials are being developed. Polyester-based coatings are growing continuously. In this context, polyester-polyurethane coatings are a promising alternative. These coatings present lower migration properties due to the formation of urethane linkages by reacting with polyisocyanate (Driffield et al., 2018; Bradley et al., 2009).

Migration levels in canned food and beverages are discussed next and summarized in Table 4. For BPA, the concentrations detected ranged from not detected to 842 ng/g. The highest concentrations have been reported in brown sauce with 842 ng/g (Sajiki et al., 2007); refried beans with 790 ng/g (Noonan et al., 2011); mushroom slices with 265 µg/kg (Cunha & Fernandes, 2013); and red pepper with 241 µg/kg (Alabi et al., 2014). The samples of brown sauce and refried beans exceeded the previous established SML of 0.6 mg/kg (Commission Regulation [EU] No. 10/2011 [European Union, 2011]), and the



samples of mushrooms and red pepper would exceed the updated SML of 0.05 mg/kg (Regulation [EU] No. 2018/213 [European Union, 2018]). Regarding other bisphenols, BPF has been detected in soft and energy drinks, beers, and filling liquids of canned vegetables at concentrations ranging from 0.10 to 7.07  $\mu\text{g/L}$  (Gallart-Ayala et al., 2011a; Gallo et al., 2017; Zech et al., 2015; Cirillo et al., 2019; Cacho et al., 2012). Cunha and Fernandes (2013) determined BPB in canned fruits and vegetables (nd to 3.4  $\mu\text{g/kg}$ ). Others such as BPZ, BP, and BPS have been analyzed in different canned foods and beverages. Thus, for example, BPZ has been determined in sport drinks and filling liquids of olives and pineapple (0.09–0.92  $\mu\text{g/L}$ ), BP and BPS have been analyzed in vegetable canned foods in both the food and the supernatant, and the reported levels varied between nd to 365 and nd to 175 ng/ml, respectively. The concentrations of BADGE and its hydrolyzed and chlorinated derivatives reported generally were below the SML (the sum of BADGE and its hydroxyl derivatives: 9 mg/kg; the sum of its chlorinated derivatives: 1 mg/kg) (Commission Regulation [EC] No. 1895/2005 [European Union, 2005]). Nevertheless, for instance, Hammarling et al. (2000) detected levels up to 12.6 mg/kg (BADGE+BADGE·2H<sub>2</sub>O+BADGE·HCl+BADGE·2HCl) in a sample of mackerel in tomato sauce, and Summerfield et al. (1998) reported a concentration of BADGE of 62 mg/kg in the covering oil of a sample of canned anchovies. High concentrations of BFDGE (up to 4.2 mg/kg) in canned fish were reported by Theobald et al. (2000) as part of a European survey on the migration of components from epoxy resins in canned fish. It is worth to comment that, although since 2006 its use is prohibited, several studies have reported its presence in canned food (Alabi et al., 2014; Gallo et al., 2017; Cabado et al., 2008). It is also interesting to highlight the relatively high levels of cyclo-di-BADGE (<25–2600  $\mu\text{g/kg}$ ) found in canned fish and meat products (Biedermann et al., 2013). Polyester monomers and oligomers have been determined in different canned foods. For example, Paseiro-Cerrato et al. (2019a) analyzed three monomers and two oligomers, namely, TA, IPA, NA, PA+2MPD, and PD+CHDM+MPD in several samples from the U.S. market. The concentrations of monomers measured did not exceed the SML, and the concentration of oligomers ranged from not detected to 324  $\mu\text{g/kg}$  and from not detect to 454  $\mu\text{g/kg}$ , respectively. Driffield et al. (2018) evaluated the migration of selected monomers (IPA, PA, *cis*-endo-NA, *trans*-endo-exo-NA, endo-NAH, and exo-NAH) and oligomers from polyester can coatings based on nadic anhydride. The IPA and PA monomers were below the SML and reported an estimated concentration of total oligomers ranging from <LOD to 943  $\mu\text{g/kg}$ .

## 5 | DIETARY EXPOSURE ASSESSMENT

Consumer exposure to chemicals from FCMs can occur as a result of the migration from the food packaging materials into the food. Exposure assessment is one of the essential elements in the risk assessment process (Poças & Hogg, 2007). To estimate the exposure to migrants from FCMs, several approaches have been applied. In the European Union, a conservative assumption is adopted, namely, a person of 60 kg body weight (bw) consumes daily 1 kg of food packed in a cubic container of 6 dm<sup>2</sup>, and it also assumes that the migration occurs at the maximum permitted level. Although, in general, this assumption leads to an overestimation of the exposure, in some cases this may lead to an underestimation (Holmes et al., 2005). Another approach also deterministic is the one used in the United States by the Food and Drug Administration (US FDA), which combines migration data with the factors known as “consumption factors” and “food-type distribution factors” (FDA, 2007; Duffy et al., 2007). More refined approaches have also been used, and thus in the framework of the European project FACET (Flavors, Additives, and Food Contact Materials Exposure Task), a probabilistic modeling tool to estimate the exposure to migrants from FCMs was developed. This model allows the exposure assessment of migrants by using information on packaging (use, composition), consumption data, and so on (Oldring et al., 2014). Dietary exposure can also be assessed more realistic by combining data on food consumption obtained from consumer surveys and the migrated substance concentration (Lestido-Cardama, Vázquez-Loureiro et al., 2021; Lestido-Cardama, Sendón et al., 2021).

Limited exposure data on substances migrating from polymeric coatings are available. For BPA, due to possible endocrine disrupting effects, tolerable daily intakes (TDIs) were established in order to protect the consumer health. In the opinion published by the European Food Safety Authority (EFSA) in 2015 on the risk to public health related to the presence of BPA in foodstuffs, the TDI was reduced from 0.05 mg/kg bw per day established in its opinion of 2006 to a temporary tolerable daily intake (t-TDI) of 4  $\mu\text{g/kg}$  bw per day. The CEF panel concluded that there is no health concern for any age group from dietary exposure (EFSA, 2015). Also, a reference dose of 0.05 mg BPA/kg bw/day was established by the U.S. Environmental Protection Agency (U.S. EPA, 1988). And on the other hand, a TDI for BADGE and its hydrolysis products of 0.15 mg/kg bw was established by EFSA in its opinion of 2004. With respect to the toxicity of BADGE and its chlorohydrins, EFSA concluded that they were not a cause of concern for carcinogenicity



and genotoxicity *in vivo* (EFSA, 2004). However, a recent study published in the literature highlighted the higher cytotoxicity of BADGE and BADGE·2HCl compared to BADGE·H<sub>2</sub>O. This study also evidenced endocrine disrupting potential of these compounds and has showed the alterations in placenta cell lipid produced after the exposure to BADGE derivatives. Due to the toxicity exhibited by these compounds, the authors claimed that the SML and TDI need to be reviewed and their exposure monitored (Marqueño et al., 2019). In addition, BADGE and its derivatives BADGE·H<sub>2</sub>O and BADGE·2H<sub>2</sub>O have been described as responsible for reproductive failures (Nerín et al., 2014). In this context, it is interesting to comment that several studies have reported the very low toxicity of tetramethyl bisphenol F and even its reaction products with epichlorohydrin have shown low toxicity in *in vitro* assays. Tetramethyl bisphenol F has been proposed as an alternative to replace epoxy-resins based on BADGE (Zhang et al., 2020). Regarding the toxicity of bisphenols, the toxicological effects of BPA have been widely studied; however, less information has been available for other bisphenol analogues. Thus, for example, BPB, BPE, and BPF have exhibited moderate to slight acute toxicity. None have shown mutagenicity and all of them presented estrogenic activity. It is interesting to note that the molecules with lower polarity presented higher estrogenic activity (Chen et al., 2002). On the other hand, several studies have shown that BPA, BPS, and BPF induced oxidative stress (Maćczak et al., 2017; Huang et al., 2020).

Table 5 summarizes studies on the dietary exposure to chemicals migrating from metal food and beverage cans. All of them are focused on bisphenol-related compounds and BADGE and derivatives. In general, the exposure data reported in the literature indicate low exposure to bisphenols, BADGE, and derivatives with values, in all cases, below the established TDIs. For example, Bemrah et al. (2014) used a probabilistic approach to estimate the dietary exposure to BPA in the French population. Different population groups, namely, infants under 36 months, children and adolescents, adults, and pregnant women, were considered. In infants and young children, the exposure increased with the age, and the authors attributed this increase to the introduction of common foods in the diet of young children; on the contrary, in the group of children and adolescents, the exposure decreased with the age, which was attributed to an increase in the body weight. In all groups, low exposure to BPA (0.038–0.14  $\mu\text{g}/\text{kg}$  bw/day) was observed. In another interesting study conducted by Simoneau et al. (1999), the exposure to BADGE from canned fish in the European and Swiss population was investigated. The results reported showed that Italy was the country with the highest exposure levels (724  $\mu\text{g}/\text{person}/\text{year}$ ), and the anchovies were the food

product that showed a higher potential for exposure to BADGE. Although the exposure was low, the authors could not conclude if these exposure levels present a risk for the consumer since, when the study was carried out, a TDI had not yet been established. A two-dimensional probabilistic approach to estimate the dietary exposure of U.K. consumers to BADGE was used by Oldring et al. (2006). The model scenarios used included different age groups. For all of them, estimated values (97.5th percentile 0.41–0.83  $\mu\text{g}/\text{kg}$  bw/day) were below the TDI of 0.15 mg/kg bw for BADGE and the hydrolysis products. Beverages and aqueous and acid foods turned out to be the foodstuffs with the great contribution to the exposure due to the high consumption of these products. More recently, Lestido-Cardama et al., reported a study on the dietary exposure of the Spanish adult population to bisphenol-related compounds migrating from metal food and beverage cans (Lestido-Cardama, Vázquez-Loureiro et al., 2021; Lestido-Cardama, Sendón et al., 2021). The estimated exposure levels were also below the TDIs. Within the beverage group, natural mineral water was the main contributor to the exposure (0.05  $\mu\text{g}/\text{kg}$  bw/day), and in the canned food group, the authors highlighted the high concentration of cyclo-di-BADGE found in a sample of mussels (3590  $\mu\text{g}/\text{kg}$ ).

Examining the data reported in the literature, low dietary exposure to bisphenols, BADGE, and derivatives have been reported. However, it is important to highlight that, in human risk assessment cumulative exposure, that is, “combined exposure to multiple chemicals including all routes, pathways, and sources of exposure to multiple chemicals,” and aggregate exposure, that is, “exposure to a single substance from multiple sources and by multiple pathways and routes,” should be considered (EFSA, 2013), and these studies have not taken into account the exposure to multiple chemicals.

## 6 | REGULATORY ISSUES

In Europe, there is not currently a specific harmonized legislation for varnishes and coatings. However, specific regulations exist in several EU Member States. An overview of the national measures (positive or negative list, end use if applicable, restrictions residual, quantity in material, overall migration limit, SML, and test conditions of any) of the Member States is presented in the JRC Science for Policy report (Simoneau et al., 2016). An updated revision on the coating evaluation and the current situation in Europe is presented in a report of EFSA Task Force on varnishes and coatings for FCMs (EFSA, 2019). Like other FCMs, varnishes and coatings are covered by the Framework Regulation (EC) No. 1935/2004 (European Union,

TABLE 5 Dietary exposure data of migrants transferred from polymeric can coatings

Migrant	Source	Exposure methodology	Country/population	Exposure data/results (TDI)	Reference
BPA	Foods (pasta, rice and wheat products, meat, poultry and game, fish, vegetables, potatoes, fruits, desserts, etc.) and beverages (milk, coffee, alcoholic drinks, etc.)	Probabilistic approach	French population/Infants (<36 months), children and adolescents (3–17 years), adults (>18 years) and pregnant women	<sup>a</sup> Results: 0.12–0.14 $\mu\text{g}/\text{kg}$ bw/day for infants, 0.05–0.06 $\mu\text{g}/\text{kg}$ bw/day for children and adolescents, 0.038–0.040 $\mu\text{g}/\text{kg}$ bw/day for adults, and 0.05–0.06 $\mu\text{g}/\text{kg}$ bw/day for pregnant women	Bemrah et al. (2014)
BADGE	Foods in contact with coated light metal food packaging (tuna, mackerel, sardines, anchovies, vegetables, soups, desserts, pasta, fruit, meat, beverages)	Two-dimensional probabilistic model (Monte-Carlo)	U.K. population for different ages and genders	<sup>b</sup> Results: 0.69 $\mu\text{g}/\text{kg}$ bw/day for youth, 0.4 $\mu\text{g}/\text{kg}$ bw/day for adult, and 0.39 $\mu\text{g}/\text{kg}$ bw/day for senior. The 97.5th percentile level: 0.41–0.83 $\mu\text{g}/\text{kg}$ bw/day	Oldring et al. (2006)
BPA, BPAF, BPAP, BPB, BPF, BPP, BPS, BPZ	Canned beverages, fats and oils, fish and seafood, meat and meat products, fruits, vegetables, and “others”	Multiplying the measured concentrations by the average daily ingestion rates of the corresponding food item by each age group	United States/Infants (<1 year), toddlers (<6 years), children (6 to <11 years), teenagers (11 to <21 years), and adults ( $\geq 21$ years)	<sup>a</sup> Estimated daily dietary intakes of bisphenols: 243, 142, 117, 63.6, and 58.6 ng/kg bw/day for toddlers, infants, children, teenagers, and adults, respectively	Liao and Kannan (2013)
BPA, BPS, BPF, BPB	Canned cereal, canned meat (beef), canned fish (tuna)	Multiplying the average contamination concentrations and daily recommended intakes	Adult residents (18–45 years old) in Zhejiang Province (China)	<sup>b</sup> Daily dietary intakes of bisphenols: BPA: 3.8–8.8; BPS: 0.0–0.032; BPF: 0.16–1.2 mean (ng/kg bw/day); BPA: 13.6–24.8; BPS: 0.0–0.16; BPF: 0.56–2.9 95th (ng/kg bw/day)	Zhou et al. (2019)
BADGE	Canned food (pasta, peaches in syrup, etc.)	Two-dimensional probabilistic approach (Monte Carlo)	U.K. population	<sup>a</sup> Results: 0.137 $\mu\text{g}/\text{kg}$ bw/day for youth, 0.065 $\mu\text{g}/\text{kg}$ bw/day for adult, and 0.050 $\mu\text{g}/\text{kg}$ bw/day for senior population	Holmes et al. (2005)

(Continues)

TABLE 5 (Continued)

Migrant	Source	Exposure methodology	Country/population	Exposure data/results (TDI)	Reference
BADGE	Canned fish in oil	The results of consumption were used together with the values obtained for the samples analyzed in the European survey on the quantification of BADGE	Each Member State of European Union and Switzerland	<sup>b</sup> France: nd–212 µg/person/year; Germany: nd–37.4 µg/person/year; Italy: nd–72.4 µg/person/year; Luxembourg: nd–355 µg/person/year; Spain: nd–59 µg/person/year; Switzerland: nd–9 µg/person/year	Simoneau et al. (1999)
BPA	Canned vegetables, fruit, meat (including pork, beef, and poultry), fish, dairy	A “forward” approach combining BPA concentrations in food with daily consumption rates, and a “backward” approach using the measurement of BPA in urine	General adult population in Dallas, Texas (United States)	<sup>a</sup> Overall dairy intakes from canned foods 12.4 ng/kg/day, which was lower than the determined from urine. Canned vegetables accounted for 11.9 ng/kg/day, with canned meat second at 0.4 ng/kg/day	Lorber et al. (2015)
BPA	Different food items from scientific literature published in peer-reviewed journals and from EFSA’s call for data	Source-to-dose modeling (forward calculation) for different age groups and compared with estimates based on urinary biomonitoring data (backward calculation)	EU population/Infants (0–6 months, 6–12 months), toddlers (1–3 years), children (3–10 years), adolescents (11–18 years), adults (18–45 years), 46–65 years, and elderly (66 years and above)	<sup>a</sup> Average internal dietary exposure: 165 ng/kg bw/day for infants, breast-fed (0–3 months), 375 ng/kg bw/day for toddlers (1–3 years), 290 ng/kg bw/day for children (3–10 years) and 132 ng/kg bw/day for adult women (18–45 years); average internal exposure by backward modeling: <10 ng/kg bw/day for infants (0–1 months), 107 ng/kg bw/day for toddlers (3–5 years), 49 ng/kg bw/day for children (5–10 years) and 36 ng/kg bw/day for adult women (18–52 years)	Von Goetz et al. (2017)

(Continues)

TABLE 5 (Continued)

Migrant	Source	Exposure methodology	Country/population	Exposure data/results (TDI)	Reference
BADGE.2H <sub>2</sub> O and BADGE	Epoxy-coated beverage and food metal cans	Product of the concentration and the total food intake (1.5 and 60 kg/person)	Korea population	<sup>b</sup> Results: $7.1 \times 10^{-4}$ $\mu\text{g}/\text{kg}$ bw/day for BADGE.2H <sub>2</sub> O and $2.8 \times 10^{-4}$ $\mu\text{g}/\text{kg}$ bw/day for BADGE	Hwang et al. (2020)
BADGE and BFDGE	Canned liquid infant formula	Product of the concentration and the intake during the specific growth period	Canada/Infants from premature to 18 months	<sup>b</sup> Results BADGE: 0.08–22 $\mu\text{g}/\text{kg}$ bw/day for the 12–18 months; BFDGE: 0.067–3.4 $\mu\text{g}/\text{kg}$ bw/day	Cao et al. (2009)
Cyclo-di-BADGE	Canned fish	Assumptions using consumption statistics	Swiss population	There is no significant health concern for the Swiss consumers	Biedermann et al. (2013)
BPA	Tuna fish, canned in either oil or aqueous medium	On the basis of measured concentrations and general daily ingestion rate	Italian population	<sup>a</sup> Results: 0.007 $\mu\text{g}/\text{kg}$ bw/day	Fattore et al. (2015)
BPA, BPB, BPC, BPE, BPF, BPG, BADGE, BADGE.H <sub>2</sub> O, BADGE.2H <sub>2</sub> O, BADGE.H <sub>2</sub> O.HCl, BADGE.HCl, BADGE.2HCl, cyclo-di-BADGE	Beverage samples, including alcoholic drinks (beer, vodka), energetic drinks, soft drinks (tonic, cola), and mineral water	Estimation using the measured concentrations and the consumption data	Spanish adult population (18–74 years)	<sup>a,b</sup> Low dietary exposure data: 0.01–0.02 $\mu\text{g}/\text{kg}$ bw/day. The highest estimated dietary exposure in the 95th percentile was 0.05 $\mu\text{g}/\text{kg}$ bw/day in natural mineral water drink	Lestido-Cardama, Vázquez-Loureiro et al. (2021)
BPA, BPB, BPC, BPE, BPF, BPG, BADGE, BADGE.H <sub>2</sub> O, BADGE.2H <sub>2</sub> O, BADGE.H <sub>2</sub> O.HCl, BADGE.HCl, BADGE.2HCl, cyclo-di-BADGE	Canned food samples: fish (tuna, sardines), seafood (clams, mussels), vegetables (olives, asparagus, tomato), grains (sweet corn), and fruit (peach in syrup)	Estimation using the measured concentrations and the consumption data	Spanish adult population (18–74 years)	<sup>a,b</sup> Results: 0.003–0.985 $\mu\text{g}/\text{kg}$ bw/day. The highest mean concentration was observed for cyclo-di-BADGE in a sample of pickled mussels	Lestido-Cardama, Sendón et al. (2021)

(Continues)

TABLE 5 (Continued)

Migrant	Source	Exposure methodology	Country/population	Exposure data/results (TDI)	Reference
BPA	Canned beverages: Water, sparkling, cola, cola lemon, cola diet, cola diet caffeine-free, energy drink, beer, beer diet cherry flavored, sport drink orange, sport drink lemon, sport drink peach, iced tea, iced tea mango, iced tea lemon, green tea citrus, apple juice, orange juice, vegetables juice, juice tropical, tonic water, soda orange, soda lemon, soda citrus, soda tropical, soda lemon-lime Canned foods: fruit mix, applesauce, black olives, carrots, carrots and peas, corn, haricot beans in tomato sauce, peeled tomatoes, bamboo, mushrooms, chicken cream soup, vegetable soup, tomato-vegetable soup, tuna in oil, tuna in water, salmon, anchovy, sausages, ravioli	Exposure was assessed through canned food and beverages consumption (Belgian food consumption survey) (Intake was calculated using food and urinary detection)	Belgian adult population	<sup>a</sup> Mean: 0.015 µg/kg bw/day 95th percentile: 0.086 µg/kg bw/day (average adult bw 70 kg)	Geens et al. (2010)

Abbreviations: BADGE, bisphenol A diglycidyl ether; BADGE.H<sub>2</sub>O, bisphenol A (2,3-dihydroxypropyl) glycidyl ether; BADGE.2H<sub>2</sub>O, bisphenol A bis(2,3-dihydroxypropyl) ether; BADGE.HCl, bisphenol A (3-chloro-2-hydroxypropyl) glycidyl ether; BADGE.2HCl, bisphenol A bis(3-chloro-2-hydroxypropyl) ether; BADGE.H<sub>2</sub>O.HCl, bisphenol A (3-chloro-2-hydroxypropyl) (2,3-dihydroxypropyl) ether; BFDGE, bisphenol F diglycidyl ether; BPA, bisphenol A; BPAF, bisphenol AF; BPAP, bisphenol AF; BPPB, bisphenol B; BPC, bisphenol C; BPE, bisphenol E; BPF, bisphenol F; BPG, bisphenol G; BPP, bisphenol P; BPS, bisphenol S; BPZ, bisphenol Z; bw, body weight

<sup>a</sup>TDI BPA 0.05 mg/kg body weight per day from 2015 a t-TDI 4 mg/Kg bw/day. Opinion of the Scientific Panel on Food Additives, Flavorings, Processing Aids and Materials in Contact with Food on a request from the Commission related to 2,2-BIS(4-HYDROXYPHENYL)PROPANE (Bisphenol A) The EFSA Journal 2006;428:1 of 75, and SCIENTIFIC OPINION Scientific Opinion on the risks to public health related to the presence of bisphenol A (BPA) in foodstuffs: Executive summary. EFSA Journal 2015;13(1):3978.

<sup>b</sup>TDI BADGE and its hydrolysis products 0.15 mg/kg bw The Scientific Panel on Food Additives, Flavorings, Processing Aids and Materials in Contact with Food (AFC) on a request from the Commission related to 2,2-bis(4-hydroxyphenyl)propane bis(2,3-epoxypropyl)ether (Bisphenol A diglycidyl ether, BADGE). The EFSA Journal 2004;86:1-40.



2004), whereby they must fulfill the general requirements (Article 3) for all FCMs. Varnishes and coatings are included in Annex I of the aforementioned regulation, which specified the list of groups of materials and articles for which specific measures may be established.

On the other hand, there is a resolution of the Council of Europe (CoE, 2004) on coatings intended to come into contact with foodstuffs that includes general guidelines and a list of substances used in the manufacture of coatings intended to come into contact with foodstuffs.

Notwithstanding the above, the Regulation (EC) No. 1895/2005 (European Union, 2005) restricts the use of certain epoxy derivatives in materials and articles intended to come into contact with food. Specifically, it establishes a maximum SML of 9 mg/kg for the sum of BADGE and their hydrolyzed derivatives, that is, BADGE·H<sub>2</sub>O and BADGE·2H<sub>2</sub>O, and also an SML of 1 mg/kg for the sum of their hydrochloric derivatives, that is, BADGE·HCl, BADGE·2HCl, and BADGE·H<sub>2</sub>O·HCl. Moreover, the use or presence of BFDGE and NOGE (novolac glycidyl ether) in the manufacture of materials is prohibited. And, with respect to the use of BPA in varnishes and coatings intended to come into contact with food, Regulation (EU) No. 2018/213 (European Union, 2018), an amendment of Regulation (EU) No. 10/2011 (European Union, 2011) regarding the use of that substance in plastic FCM, set the SML of BPA at 0.05 mg/kg. Hence, varnishes and coatings should be in compliance with these substance-specific measures, and with the Framework Regulation (EC) No. 1935/2004 (European Union, 2004), and furthermore, they must also comply with national legislations when available.

## 7 | CONCLUSIONS

The inner surface of food metal cans is usually coated with a polymeric coating. Currently, coatings based on different chemistries such as epoxy, polyester, acrylic, and so on are being applied. The polymeric coatings are very complex formulations that contain not only starting substances (e.g., monomers, prepolymers, additives, etc.) but also unknown compounds (e.g., degradation and reaction products, oligomers, etc.). These substances can migrate into the food and may constitute a risk for the consumers' health; therefore, it is necessary to analyze them properly. Sample preparation previous to instrumental analyses usually comprises solvent extraction followed by a clean-up step. SPE and QuEChERS are the most commonly used sample pretreatment methods. For complex matrix, molecularly imprinted polymers cartridges instead of the conventional SPE cartridges have been used due to their high selectivity. For separation and detection, liquid chro-

matography and gas chromatography coupled to different detectors and particularly to mass spectrometry are the techniques of choice to accomplish this task. For the unknown compounds, HRMS has demonstrated to be a powerful tool that provides accurate mass measurements. Recent applications have shown DART-HRMS as a complementary and efficient analytical tool that allows to complete the analysis in few minutes. Canned food can be an important source of exposure to different chemicals, for example, bisphenols, BADGEs, and so on; therefore, the study of the migration and dietary exposure is required for the risk assessment and the successive establishment of legislation. Although a considerable number of studies on the migration from metal cans have been reported in the literature, most of them are focused on the migration of monomers and other starting substances; however, information about oligomers and other NIAS is limited. Hence, more studies are needed to identify and evaluate the migration of these substances to ensure the safety of the canned food. As an essential step in the risk assessment, the exposure to chemicals migrating from coatings is assessed, but up to now scarce data are available and much effort is still needed to investigate human exposure to these migrants. Future works should consider the combined exposure to multiple chemicals. From the regulatory perspective, at present there is not a harmonized legislation in the European Union for varnishes and coatings for food contact; they must comply with the Framework Regulation No. 1935/2004 (European Union, 2004) applicable to all FCMs. It would be of value to have an harmonized European legislation for varnishes and coatings in order to support the safety of this group of FCMs.

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## AUTHOR CONTRIBUTIONS

**Antía Lestido-Cardama:** Conceptualization; Investigation; Methodology; Writing—original draft. **Raquel Sendón:** Conceptualization; Methodology; Project administration; Supervision; Writing—review & editing. **Juana Bustos:** Conceptualization; Methodology; Supervision; Writing—review & editing. **María Teresa Nieto:** Methodology; Writing—review & editing. **Perfecto Paseiro-Losada:** Conceptualization; Methodology; Supervision; Writing—review & editing. **Ana Rodríguez-Bernaldo de Quirós:** Conceptualization; Funding acquisition; Investigation; Methodology; Supervision; Writing—original draft.

## CONFLICTS OF INTEREST

The authors declare no conflicts of interest.

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