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Migration of Bisphenol A and Bisphenol A Diglycidyl-ether from Can Coatings into Food Simulants

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The migration and alteration of the key bisphenol-A-related compounds, bisphenol A and bisphenol A diglycidyl-ether, in food simulants (deionized water, 3 % acetic acid and 50 % ethanol) stored in coated cans were investigated. Quantification and confirmation of the two compounds were performed with ultrahigh-performance liquid chromatography-tandem mass spectrometry (UHPLC-MS/MS). Recoveries for bisphenol A and bisphenol A diglycidyl-ether at three concentration levels (10, 25, 250 μg L^{-1}) ranged from 81.8 to 101.5 % and RSDs were 2.61 to 12.52 %. The effects of the initial concentration, heat processing (for sterilization), time and temperature of storage on migration and alteration of bisphenol A and bisphenol A diglycidyl-ether were investigated. The results showed that heat processing contributed greatly to bisphenol release. Besides, storage temperature and time had a significant effect on the migration of bisphenol A , with concentrations increasing gradually with time and temperature. Conversely, the concentrations of bisphenol A diglycidyl-ether decreased during storage because of hydrolysis by the food simulants, especially for aqueous acetic acid.

Keywords: Bisphenol A diglycidyl-ether, Bisphenol A, Can coating, Epoxy resins, Food simulants, Migration.

INTRODUCTION

Epoxy-based lacquer materials, which have excellent flexibility, chemical resistance and adhesion, are commonly used for coating the inside of food cans, large storage vessels and food containers to reduce food spoilage and prevent degradation of the food can[1]. Among these compounds, the epoxy-phenolic resin coatings are mainly derived from bisphenol A and bisphenol A diglycidyl-ether[2] and require BFDGE and bisphenol A diglycidyl-ether as stabilizers during production to neutralize and solidify the hydrochloric acid generated by the reaction to prevent decomposition of the coating.

Migration of potentially toxic compounds in the epoxy resins used for packaging materials, especially for linings in commercial cans, is a critical food safety issue, because of their potential as endocrine disruptors. If the chemical reaction in the production process of the coating is not complete and the cross linking of materials is not sufficient, residues of bisphenols may form. In addition, the coatings can release the bisphenols, as well as oligomers, which can migrate into the packaged foods during processing, sterilization, transportation and storage[3,4]. Such compounds, which can enter the human body through the food chain, affect human health[5,6]. Bisphenol A diglycidyl-ether is genotoxic, mutagenic and has the effect of an antiandrogen, causing abnormalities in the human endocrine system, immune system and nervous system and affecting normal reproductive genetic function[2]. Because of its wide availability in the environment and its estrogenic activity in specific responses *in vitro* and *in vivo*, adverse effects of bisphenol A exposure on human health are possible. It has been hypothesized that exposure to xenoestrogens such as bisphenol A, during early development, may be the underlying cause of the increased incidence of infertility, genital tract abnormalities and breast cancer observed in European and US human populations over the last 50 years[7,8].

Migration and alteration of bisphenol A and bisphenol A diglycidyl-ether is normally studied using food simulants. Lin *et al.*[9] investigated the effect of storage time on the migration of bisphenol-A-related compounds from can coatings into food simulant and oily foods. However, monitoring the migration of compounds from packaging materials into food is complex and requires the use of advanced analytical methods, such as liquid chromatography (HPLC-FLD)[4,10], gas chromatography-mass spectrometry (GC-MS)[11,12] and UHPLC-MS/MS[9]. Migration of bisphenol A has been investigated in different foodstuffs taking into account several parameters, such as time and temperature of storage[4,13,14]. Several studies on hormonal disruptors such as bisphenol A diglycidyl-ether and other derivatives,

including migration, have been performed in aqueous simulants or aqueous canned foods[10,15]. Earlier publications indicated that after migration from packaging into foodstuffs, bisphenol A diglycidyl-ether undergoes various reactions with unidentified food components. Besides migration and alteration of bisphenol A diglycidyl-ether, the compound can undergo hydrolysis as a result of contact with aqueous and acidic foods[3,9]. Given such instability in aqueous media, information about the hydrolysis products and the kinetics of decomposition of bisphenol A diglycidyl-ether is of great interest. In the case of fatty foodstuffs, hydrolysis is not common, although some reports suggest that bisphenol A diglycidyl-ether can react with food components, leading to a decrease of bisphenol A diglycidyl-ether levels depending on the sterilization process and the storage time[2,16].

The European Union (EU) has established legislation concerning the specific migration limit (SML) for bisphenol A and related compounds in material and articles intended to come into contact with food or food simulants[17]. Regarding legislation, the EU has set a maximum specific migration limit of 9 mg kg^{-1} for bisphenol A diglycidyl-ether and, in the case of bisphenol A, an specific migration limit of 0.6 mg kg^{-1}[18,19]. The presence of BFDGE in food packaging products has been prohibited since 2005[20]. Recently, the French Parliament decreed that the presence of bisphenol A in any food containers is banned from 1 January 2014[18,19].

Many studies have consistently revealed that migration and alteration of bisphenol-A-related compounds gradually increased with increase in the storage temperature and time. However, there is a lack of information concerning migration and alteration of bisphenol A and bisphenol A diglycidyl-ether from can coatings after heat treatment at different temperatures and time.

The aim of this work was to determine the effects of heat processing (at different temperatures) and storage time (up to 35 days) on the migration (and alteration) of bisphenol A and bisphenol A diglycidyl-ether into food simulants when the compounds were used as internal coatings in canned food.

EXPERIMENTAL

A Waters Acquity UPLC-MS/MS system (Waters, Milford, PA, USA) equipped with a Turbo Ion Spray electrospray ionization (ESI) source and triple quadrupole were employed. An ultrasonic bath (AS10200BT, Tianjin Autoscience Instrument Co., Ltd. Tianjin, China), a rotary evaporator (Hei-VAP, Heidolph, Germany), a constant temperature incubator (Heratherm IMH 180-S, Thermo Scientific, Germany) and a Milli-Q water system (Millipore, Billerica, MA, USA) were used for the migration tests.

Dichloromethane (HPLC grade, > 99.9 %, CAS No. 75-09-2) was purchased from Shanghai Anpel Scientific Instrument Co., Ltd. (Shanghai, China). Methanol (LC-MS grade, > 99.9 %, CAS No. 67-56-1) was purchased from Oceanpak Alexative Chemical Co., Ltd. (Goteborg, Sweden). Glacial acetic acid (A.R. grade, > 99.5 %, CAS No. 64-19-7) was purchased from TianJin ZhiYuan Chemical Co., Ltd. (Tianjin, China). Ammonium acetate (A.R. grade, ≥ 98 %, CAS No. 631-61-8), ammonia solution (A.R. grade, 25 %, CAS No. 1336-21-6) and formic acid (HPLC grade, > 99.9 %, CAS No. 64-18-6) were supplied by Xi Long Chemical Co., Ltd. (Shantou, Guangdong, China). A standard solution of bisphenol A diglycidyl-ether (BADGE, CAS No. 1675-54-3) was obtained from Dr. Ehrenstorfer GmbH (Augsburg, Germany). A standard solution of bisphenol-A (BPA, CAS No. 80-05-7) was obtained from AccuStandard Inc. (USA).

Standards and samples: Individual stock solutions, prepared by dissolving 10 mg of each compound in 10 mL of methanol, were stored in Amber brown glass-stoppered bottles at 4 °C. Standard solutions for calibration were prepared on a daily basis by serial dilutions in methanol.

The cans (300 mL; three-piece tin cans with pull-top lids) were purchased from the Kai Bo Food Supermarket (Shenzhen, China). They were pre-cleaned using Milli-Q water and dried before use. Before the migration tests, tested cans were filled with 300 mL of food simulant and sealed with aluminum foil.

Food simulant: Based on the EU regulation no. 10/2011 on plastic materials and articles intended to come into contact with food[21], distilled water, 3 % (w/v) acetic acid and 50 % (v/v) ethanol were selected as food simulants. Acetic acid (15.0000 g) was weighed accurately into a volumetric flask and diluted with Milli-Q water to 500 mL to obtain a 3 % (w/v) acetic acid solution. For 50 % (v/v) ethanol, 250 mL of absolute ethanol was transferred to a volumetric flask and diluted to 500 mL with Milli-Q water.

Chromatographic conditions: A waters acquity UPLC BEH C18 column (50 mm × 2.1 mm i.d., 1.7 μm particle size) was used for separations. Column temperature was kept at 35 ± 0.5 °C and the injection volume was 5 μL for mixed standards and samples. Elution was carried out in binary gradient mode. The mobile phase for the determination of bisphenol A diglycidyl-ether in ESI^+ mode was 0.1 % formic acid in 5 mM ammonium acetate solution (A) and methanol (B). Before analysis, the column was conditioned for 15 min with A:B (70:30) to obtain a stable baseline. The binary gradient program employed in ESI^+ mode was the following: 0-0.5 min, 30 % B; 0.5-2 min, 30-70 % B; 2-2.5 min, 70 % B was maintained; 2.5-3 min, 70-95 % B; 3-5 min, 95 % B was maintained; 5-6 min, 95-30 % B. The flow rate of the mobile phase was 0.25 mL min^{-1}. The mobile phase in ESI- mode for the determination of bisphenol A was 0.2 % ammonia solution (A) and methanol (B). The binary gradient program employed in ESI- mode was the following: 0-0.5 min, 30 % B; 0.5-2 min, 30-70 % B; 2-3.5 min, 70 % B was maintained; 3.5-5 min, 70-30 % B. The flow rate of the mobile phase was 0.25 mL min^{-1}.

Mass spectrometry conditions: Mass spectrometry data were acquired and processed using MassLynx 4.1 software. Analytes were detected in multiple reaction monitoring mode (MRM) using ESI in positive or negative ion mode and using a single chromatographic run per sample. Two reaction fragmentations were scanned for each analyte. The MS source parameters were as follows: ion source temperature 120 °C, desolvation temperature 350 °C, desolvation flow rate 700 L h^{-1}, cone gas flow rate 50 L h^{-1} and capillary voltage 3.2 kV. The gas used in the mass spectrometer was high purity (99.99 %) nitrogen. Argon was used as collision gas at 1.5 mtorr and the optimum cone voltage (V) and collision energy (eV) selected for each transition are indicated in Table-1.

TABLE-1
MRM ACQUISITION PARAMETERS

Compound	Precursor ion (*m/z*)	Quantitation: Product ions (*m/z*)	Cone (V)	Collision energy (eV)
Bisphenol A diglycidyl-ether	358.4[a]	191.2*	28	15
		135.2	28	30
Bisphenol A	227.2[b]	211.3*	45	20
		133.1	45	20

*First ion indicated for each analyte was employed for quantification purposes while the second ion was used for confirmation.
[a]$[M + NH_4]^+$ adduct as precursor ion; [b][M-H]-adduct as precursor ion

Determination of concentration of bisphenol A and bisphenol A diglycidyl-ether in can coating: Three hundred mL of dichloromethane was poured into the metal can and sealed with aluminum foil, followed by ultrasonic extraction (frequency: 40 KHz; power: 300 W; 30 °C) for 25 min. Thirty mL of the extract were taken and evaporated to near dryness at 40 °C with a rotary evaporator. The residue was dissolved in methanol and the solution made up to 10 mL. Finally, 1 mL of the solution was passed through a 0.22 μm nylon membrane filter prior to analysis by UHPLC-MS/MS.

Extraction processing for migration tests: Determination of the amounts of bisphenol A and bisphenol A diglycidyl-ether in the food simulants under the various temperature-time conditions was performed as follows: each can investigated was opened and 100 mL of food simulant from the can was placed in a glass bottle (250 mL). Next, 30 mL of dichloromethane was added to the glass bottle, which then underwent ultrasonic extraction (Frequency: 40 KHz; Power: 300 W; 30 °C) for 10 min. The extract in the dichloromethane layer was then collected in a 300 mL round-bottom flask. The liquid-liquid extraction procedure was repeated and the total combined dichloromethane extract was evaporated to near dryness at 40 °C with a rotary evaporator. The residue was dissolved in methanol and the solution made up to 10 mL. Finally, 1 mL of the solution was passed through a 0.22 μm nylon membrane filter prior to analysis by UHPLC-MS/MS.

Migration tests: Heat treatment was applied to six groups of cans (each group in triplicate), each can containing 300 mL of 3 % acetic acid for testing at the different temperatures and times. Group 1 was processed at 121 °C and migration times were 5, 10, 15, 20, 30, 40, 50 and 60 min. Migration tests for group 2 occurred over 0, 5, 10, 15, 20, 25, 30 and 35 days with storage at ambient temperature (25 ± 2 °C) following heat treatment at 121 °C for 0.5 h, as practiced in an industry. To investigate the impact of different heat treatment processes on the migration of bisphenol from the can coating, migration tests for groups 3, 4, 5 and 6 were performed for processing at 60 °C/10 min, 60 °C/0.5 h, 100 °C/10 min and 100 °C/0.5 h, respectively, with migration periods (at 25 ± 2 °C) of 0, 5, 10, 15, 20, 25, 30 and 35 days.

No heat treatment was applied to the other five groups of cans, each group consisting of sextuplets. Migration tests for three groups were performed at 4 °C, 25 ± 2 °C and 45 °C and migration times were 2, 5, 10, 15, 20, 25, 30 and 35 days. The remaining two groups of cans, one filled with deionized water and the other filled with 50 % ethanol, were stored at 25 ± 2 °C and cans were analyzed after 2, 5, 10, 15, 20, 25, 30 and 35 days.

RESULTS AND DISCUSSION

Validation of method: For validation of the analytical method, the limit of detection (LOD), the limit of quantitation (LOQ), recovery and repeatability for bisphenol A and bisphenol A diglycidyl-ether determinations in food simulants were checked. Standard solutions (2, 5, 10, 20, 50, 100, 200 and 500 μg L^{-1}) for bisphenol A and standard solutions (5, 10, 20, 50, 100, 200, 500 and 1000 μg L^{-1}) for bisphenol A diglycidyl-ether were prepared by diluting stock standard solutions (10 mg L^{-1}) with methanol. The limit of detection was defined as the lowest concentration giving a signal response equal to three times the average of the baseline noise level. The limit of detections for bisphenol A and bisphenol A diglycidyl-ether were 0.2 and 0.5 μg kg^{-1}, respectively. The limit of quantitation was defined as the lowest concentration giving a signal response equal to 10 times the average baseline noise level. The limit of quantitations for bisphenol A and bisphenol A diglycidyl-ether were 0.8 and 1.2 μg kg^{-1}, respectively (Table-2).

The repeatability of the method was verified by spiking food simulants with bisphenol A and bisphenol A diglycidyl-ether at three concentration levels (10, 25, 250 μg L^{-1}) in six replicate experiments. The recoveries and relative standard deviations (RSDs) in deionized water were 93.9-102 and 2.61-7.32 % for bisphenol A and 87.3-94.3 and 4.57-6.77 % for bisphenol A diglycidyl-ether, respectively. The recoveries and RSDs in 3 % acetic acid were 91.4-97.6 and 4.46-9.23 % for bisphenol A and 90.8-94.9 and 4.84-10.83 % for bisphenol A diglycidyl-ether, respectively. In 50 % ethanol, the recoveries and RSDs were 83.9-91.2 and 4.35-8.76 % for bisphenol A and 81.8-87.3 and 5.62-12.52 % for bisphenol A diglycidyl-ether, respectively (Table-3).

TABLE-2
PERFORMANCE PARAMETERS FOR BISPHENOL A AND BISPHENOL A DIGLYCIDYL-ETHER DETERMINATIONS (n = 6)

Abbreviation	Retention time ± SD (min)	Linearity range: 2-1000 μg L^{-1}*: Slope ± SD	Intercept ± SD	Determination Coefficient (R^2)	LODI, LODM (μg L^{-1}, μg kg^{-1})	LOQI, LOQM (μg L^{-1}, μg kg^{-1})
BPA	2.67 ± 0.01	15.7 ± 0.2	18.0 ± 1.8	0.9965	0.2, 0.4	0.8, 1.5
BADGE	3.55 ± 0.02	106.7 ± 0.4	-46.6 ± 1.1	0.9981	0.5, 0.8	1.2, 2.0

Note: LODI, LODM: the limit of detection of the instrument and the LOD of the method; LOQI, LOQM: the limit of quantitation of the instrument and the LOQ of the method; *2-500 μg L^{-1} for BPA and 5-1000 μg L^{-1} for BADGE

TABLE-3
RECOVERIES AND RSDs FOR THE SPIKED FOOD SIMULANTS (n = 6)

Food simulant	Compound	Spiked concentration of Bisphenol a and bisphenol A diglycidyl-ether					
		10 (μg L^{-1})		25 (μg L^{-1})		250 (μg L^{-1})	
		Mean recovery (%)	RSD (%)	Mean recovery (%)	RSD (%)	Mean recovery (%)	RSD (%)
Deionized water	BPA	93.9	6.13	96.6	7.32	101.5	2.61
	BADGE	87.3	6.05	91.3	4.57	94.3	6.77
3 % Acetic acid (w/v)	BPA	97.6	4.46	91.4	9.23	96.3	5.27
	BADGE	94.9	5.61	90.8	4.84	93.3	10.83
50 % ethanol (v/v)	BPA	83.9	5.41	88.4	4.35	91.2	8.76
	BADGE	81.8	7.88	83.9	12.52	87.3	5.62

Concentrations of bisphenol A and bisphenol A diglycidyl-ether in can coating: The proposed method using dichloromethane as extractant coupled with UHPLC-MS/MS was applied to determine bisphenol A and bisphenol A diglycidyl-ether in the can coating. It was found that the concentrations of bisphenol A and bisphenol A diglycidyl-ether were 0.46 ± 0.03 and 39.73 ± 2.06 mg kg^{-1}, respectively.

Effect of time and temperature: Different heating regimes were applied to the food simulant solutions. Fig. 1 shows the changes in the concentrations of bisphenol A and bisphenol A diglycidyl-ether in 3 % acetic acid at 121 °C. The heat processing contributed greatly to bisphenol release and the heat processing time had a significant effect on the migration of bisphenol A. The bisphenol A concentration increased gradually during heating and then leveled off after about 60 min. In the case of bisphenol A diglycidyl-ether at 121 °C, the compound migrated rapidly from the coating into the simulant over the first 5 min (start point of the line in Fig. 1), reaching a concentration of 865 μg kg^{-1}; thereafter, the hydrolysis rate for bisphenol A diglycidyl-ether became greater than the rate of migration. Thus there was a rapid decrease in bisphenol A diglycidyl-ether levels after 10 min and after 60 min, concentrations were close to the limit of detection, *i.e.*, 0.5 μg kg^{-1}. It was, therefore, concluded that the removal and decomposition of bisphenol A diglycidyl-ether was due to hydrolysis as a result of contact with acetic acid. Furthermore, the longer the duration of high-temperature processing, the faster was the rate of hydrolysis. In addition, the reduction in the bisphenol A diglycidyl-ether (cf signal response at about 3.55 min in Fig. 2) concentration during storage might contribute to an increase in the concentration of derivative products (*cf* signal response at about 4.15 min in Fig. 2).

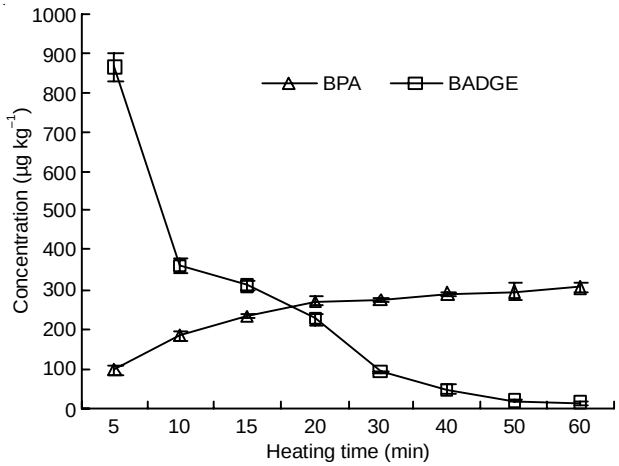

Fig. 1. Changes in concentration of bisphenol A and bisphenol A diglycidyl-ether for 3 % acetic acid at 121 °C (n = 3)

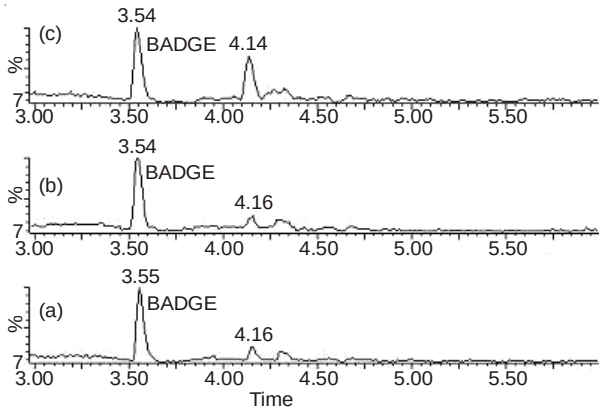

Fig. 2. Total-ion chromatograms of isphenol A diglycidyl-ether in 3 % acetic acid without heat processing and with storage at room temperature (25 ± 2 °C): (a) 5 days, (b) 10 days, (c) 15 days

The bisphenol A and bisphenol A diglycidyl-ether contents in 3 % acetic acid after heat processing (121 °C, 0.5 h) and storage at room temperature (25 ± 2 °C) are shown in Fig. 3. Levels up to about 400 μg kg^{-1} of bisphenol A were found after 30 days' storage at 25 ± 2 °C. In contrast, the bisphenol A diglycidyl-ether concentrations decreased during storage and were not detected after 25 days.

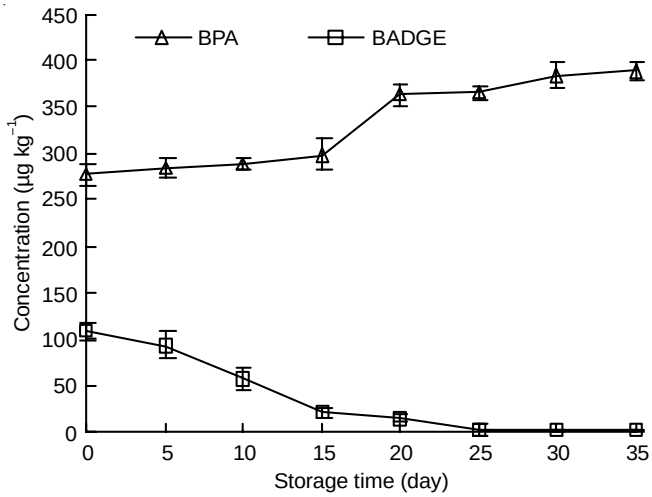

Fig. 3. Changes in concentration of bisphenol A and bisphenol A diglycidyl -ether in 3 % acetic acid after heat processing (121 °C, 0.5 h) and storage at room temperature (25 ± 2 °C) (n = 3)

Fig. 4 shows the changes in the concentrations of bisphenol A (a) and bisphenol A diglycidyl-ether (b) in 3 % acetic acid after different heat treatments and storage at room temperature. As was expected, higher heating temperatures

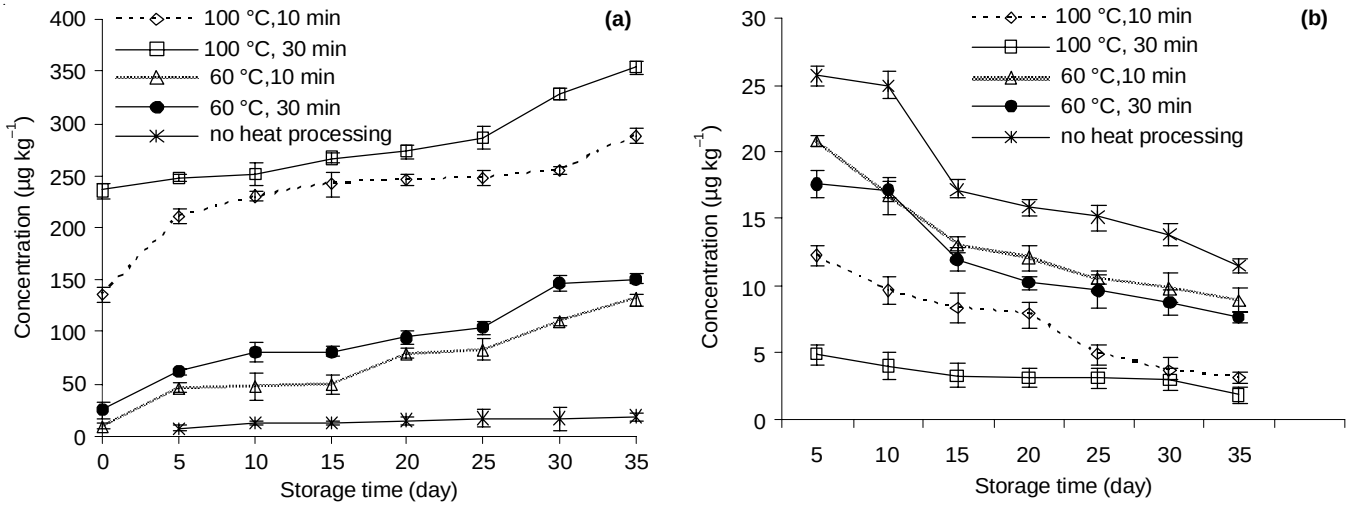

Fig. 4. Changes in concentration of bisphenol A (a) and bisphenol A diglycidyl-ether (b) in 3 % acetic acid after different heat treatments and storage at room temperature (25 ± 2 °C) (n = 3)

and heating times led to an increase in bisphenol A migration from the can coating, which contrasted with that for bisphenol A diglycidyl-ether. In the case of bisphenol A diglycidyl-ether, after heat treatments (60 °C/10 min, 60 °C/0.5 h, 100 °C/10 min, 100 °C/0.5 h) and immediate measurement, the concentrations were 155, 178, 222 and 249 µg kg^{-1}, respectively. Thereafter, concentrations all decreased rapidly to a maximum of only 20 µg kg^{-1}. In the case of the NHT (no heat processing) group, the concentrations for bisphenol A diglycidyl-ether also dropped to 26 µg kg^{-1} in 5 days (start point of lines in Fig. 4). After 35 days' storage, the bisphenol A diglycidyl-ether concentrations were less than 15 µg kg^{-1} in all tests. This indicated that the bisphenol A diglycidyl-ether concentrations were not significantly different between the heat treatment groups and the no heat treatment groups after a certain storage time because of the compound undergoing hydrolysis.

It can be concluded that the migration of bisphenol A from can coatings after heat processing was much larger than that for cans not receiving heat processing. Also, because of the hydrolysis of bisphenol A diglycidyl-ether, heat processing will not have a significant impact on the concentrations of bisphenol A diglycidyl-ether in canned food stored for relatively long times, for example more than 30 days.

Effect of storage time and temperature: In general, there was an increase in the migration of bisphenol A from the can coating to 3 % acetic acid at higher storage temperatures and longer storage times [Fig. 5 (a)]. In contrast, the concentrations of bisphenol A diglycidyl-ether decreased under the same conditions. This is well illustrated for the test at 45 °C with storage for 20 days [Fig. 5 (b)].

In addition, the studies indicated that migration of bisphenol A and bisphenol A diglycidyl-ether from the can coating to 50 % ethanol with storage at room temperature was higher than for the other two food simulants. The bisphenol A concentration increased from 25.4 to 129 µg kg^{-1} and the bisphenol A diglycidyl-ether concentration decreased from 3263 to 1375 µg kg^{-1} in 50 % ethanol after 35 days of storage at room temperature (25 ± 2 °C). Also, there was not a significant increase in the bisphenol A concentration (0.83-2.84 µg kg^{-1}) or a significant decrease in the bisphenol A diglycidyl-ether concentration (7.98-1.20 µg kg^{-1}) in deionized water with storage for 35 days at room temperature (25 ± 2 °C). These results are consistent with recent literature where substantial migration of bisphenol-A-related compounds in fatty foodstuffs with lipophilic properties was reported[18,10].

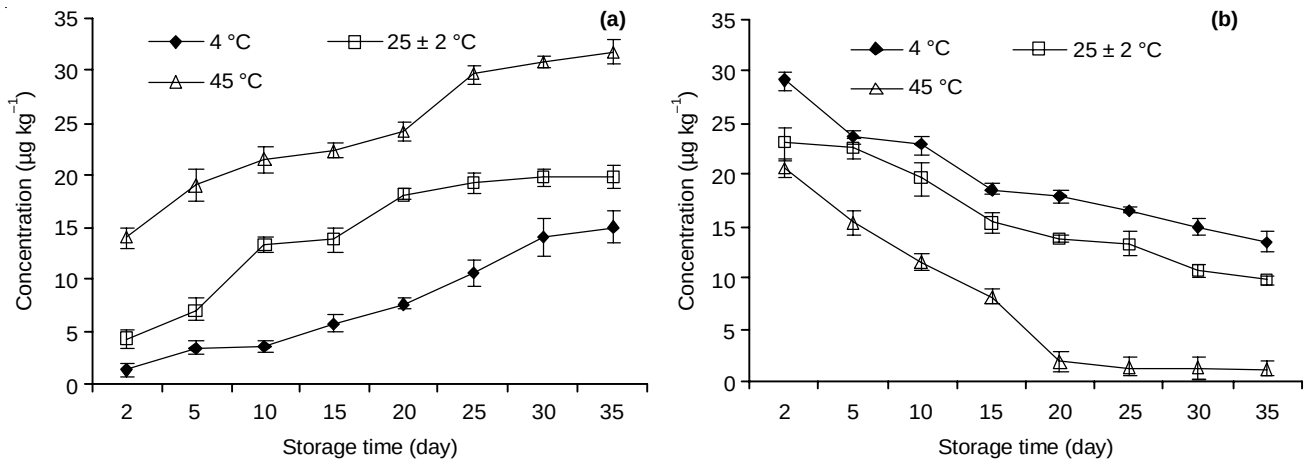

Fig. 5. Changes in concentration of bisphenol A (a) and bisphenol A diglycidyl-ether (b) in 3 % acetic acid without heat treatment (n = 3)

There were other compounds (possibly derivatives of bisphenol A diglycidyl-ether) in the can coating capable of migration into the food simulants (Fig. 2). An investigation of possible relationships between the concentrations of such derivatives and bisphenol A diglycidyl-ether concentrations is needed.

Conclusion

Heat processing enhanced the rate of migration of bisphenol A and bisphenol A diglycidyl-ether from the can coating into the food simulants. Storage temperature and storage time both showed a significant effect on the migration of bisphenol A from the can. Migration and decomposition of bisphenol A diglycidyl-ether was affected by its propensity to undergo hydrolysis, especially in acidic food simulants. The concentration levels of bisphenol A and bisphenol A diglycidyl-ether for migration under different test conditions ranged from 0.8-389 and 1.2-3263 μg kg^{-1}, respectively. The concentrations determined for each compound did not exceed EU legal limits. This study has supplemented valuable information concerning the migration of bisphenol-A-related compounds. Further work on bisphenol A diglycidyl-ether derivatives and their conversion during storage is planned.

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