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Release of Bisphenol A from Polycarbonate—A Review

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The release of Bisphenol A (BPA) from polycarbonate baby bottles into food and food simulants is reviewed in the perspective of the current intensive discussions on the risks of this substance. Potential factors that have been reported to influence the release of BPA are reviewed. Unlike most polymers polycarbonate is hydrolyzed under alkaline conditions by scale formation, residual alkaline detergents and boiled water. Data suggest that brushing of the bottle did not raise the release of BPA. Claims that used bottles release more BPA than new bottles and that mineral composition of the aqueous food simulant affect release could not be substantiated. There are indications that aminolysis of polycarbonate by milk and ethanolysis of polycarbonate by 50% ethanol might take place under relevant test conditions.
The relatively few migration data following the test conditions of European food contact material legislation, comply with the specific migration limit. Two test conditions were identified that reflect real use and exposure, and might cause higher release of BPA compared to the test conditions of European food contact material legislation. Further detailed studies are necessary to verify whether these two exposure scenarios are more severe.

Keywords Food contact materials, migration, hydrolysis, aminolysis, ethanolysis

INTRODUCTION

Bisphenol A (BPA) is mainly used as monomer for the production of polycarbonate and a precursor of epoxy resins. Consumers may be exposed to BPA via polycarbonate bottles and tableware, such as those used for infant formula milk, and via epoxy resin coatings inside food and beverage containers. More specifically, exposure to BPA can arise under conditions where residual monomer in the polymer migrates into food and beverages, or where the polymer itself hydrolyses, thereby releasing BPA. Other, relatively minor sources of consumer exposure to BPA are dental fissure sealants, epoxy-based surface coatings, adhesives, printing inks, and thermal paper.

Babies are supposed to be the most sensitive group to exposure of BPA. Babies that are not breast-fed, will drink powdered or liquid milk formula. Cans of milk formula may be coated with epoxy resin and may release BPA into the milk formula. Powdered milk formula are normally not packed in epoxy resin-lined cans, whereas the liquid milk formula are for the required heat sterilization. Milk formula may be served in a polycarbonate bottle adding to the exposure to BPA. Breast-fed infants may also receive additional feedings of milk formula or expressed milk.

Several risk assessments of BPA have shown that exposure levels are far below the no-observed-adverse-effect level (Aschberger et al., 2010). A strong scientific debate arose since several toxicological studies were not included in the previous risk assessments. This was because of several short-comings in the methodology of these toxicological studies: small number of animals, fewer or single dose groups, non-oral administration routes. As a consequence of this debate and using the precautionary principle, some countries started to ban BPA releasing articles that are intended for the use by infants and young children below the age of three.

At present the discussion on the risk assessment of BPA focuses on the toxicological part at doses below the current tolerable daily intake. In this respect, it is also important to review the migration of BPA from polycarbonate baby bottles into food and food simulants for exposure scenarios.

RESIDUAL BPA IN POLYCARBONATE

The release of BPA by diffusion is determined by the residual concentration of BPA in polycarbonate. The residual concentration of BPA has been reported to be in the range of 1–140 mg/kg

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(Biles et al., 1997; Howe and Borodinsky, 1998; Kawamura et al., 1998; Sun et al., 2001; Nerín et al., 2003; Wong et al., 2005; Ehlert et al., 2008). Higher residual concentrations for non-complying polycarbonate bottles were reported in the range of 380–600 mg/kg (Kawamura et al., 1998).

Young Park et al. (2006) studied the effect of γ-irradiation on the formation of additional residual BPA in polycarbonate. Their study shows a significant increase of residual BPA from 120 to 470–560 mg/kg. This effect may be relevant for applications where food that is packed in polycarbonate, is preserved by γ-irradiation. Under natural background conditions, γ-irradiation is not expected not generate additional residual BPA.

RELEASE OF BPA FROM POLYCARBONATE

BPA can leach from polycarbonate into liquid foods because of two different processes: diffusion of residual BPA present in polycarbonate after the manufacturing process, and hydrolysis of the polymer (Figure 1) catalyzed by hydroxide in contact with aqueous food and simulants (Ehlert et al., 2008; Mercea, 2009). For dry foods, diffusion is the only relevant process.

Release of BPA from polycarbonate containers into food depends on the contact time, temperature, and type of food. Food simulants are often used in release studies to represent the different types of food, for example, in Europe, 50% of ethanol in water is the food simulant for milk, and 3% of acetic acid in water is the simulant for fruit juice (EEC, 1985).

Tables 1–5 review the scientific literature and recent information from some European National Reference Laboratories on food contact materials (EURL-FCM, 2009). The tables are sorted in the order of contact time followed by contact temperature. Most results are obtained by using HPLC combined with fluorescence detection and therefore the tables indicate only other analytical techniques. Table 1 shows that there are relatively few studies on the release of BPA from polycarbonate products into the real food matrix so far. BPA was not detected in fruit juice (LoD = 0.03 mg/L) at 100°C for 5 min followed by 20°C for 20 min (Mountfort et al. 1997 in ECB 2003). BPA was also not detected in milk formula (LoD = 0.03 mg/L) by either contact at 70°C for 2 h followed by 40° for 24 h or contact at 100°C for 5 min followed by 20°C for 20 min (Mountfort et al., 1997; EURL-FCM, 2009). The concentration of BPA was below the limit of detection of 0.01 mg/L in soup filling at 80°C and leaving it at 20°C for 30 min (Japan, 1998). Lim et al. (2009) studied the migration of BPA into steamed hot rice and cooked hot pork. Filling the polycarbonate containers with the food (note: initial temperature was not reported) and leaving it up to 90 min at room temperature resulted in concentrations below the limit of detection of 0.001 mg/kg. The same was valid for heating the food containing bottles by a water bath (temperatures not reported). Heating the food containing bottles by a microwave (temperatures not reported) resulted in concentrations in rice of 0.006–0.011 mg/kg for 3 min, 0.011–0.014 mg/kg for 6 min, and 0.016–0.019 mg/kg for 9 min. Concentrations in pork were 0.005–0.007 mg/kg for 3 min, 0.011–0.012 mg/kg for 6 min, and 0.014–0.015 mg/kg for 9 min.

Instead, evidence of BPA release comes from the much larger number of studies that were conducted using food simulants and tap water. Table 2 shows that BPA may release substantially into oil based food simulants up to 1.5 mg/L at 65°C for 10 days (Biles et al., 1997; Howe and Borodinsky, 1998; Japan, 1998; Yokohama study in ECB, 2003; Wong et al., 2005).

Biles et al. (1997) showed that the release of BPA increases with the level of ethanol in aqueous solution in the range of 8–50% from 0.87 mg/L to 5.9 mg/L at 65°C after 10 days (Table 3). Less extreme time-temperature conditions resulted in lower release (Howe and Borodinsky, 1998; Japan, 1998; Kawamura et al., 1998; Simoneau et al. (2000) in ECB, 2003; EURL-FCM, 2009; Wong et al., 2005; CSL (2004) in ECB, 2008; EU Member State official control data, 2008–2009; VWA, 2008; Kubwabo et al., 2009).

Most literature studies (Table 4) report a release of BPA into 3% acetic acid below the limit of detection in the range of 0.0005–0.01 mg/L (Howe and Borodinsky, 1998; Japan, 1998; Earls et al. (2000) and Simoneau et al. (2000) in ECB, 2003; VWA, 2005, 2008; Maragou et al., 2008; Sim and Jianhua, 2008; EURL-FCM, 2009) and some above up to 0.0058 mg/L at 100°C for 30 min (D'Antuono et al., 2001; CSL (2004) in ECB, 2008; EURL-FCM, 2009).

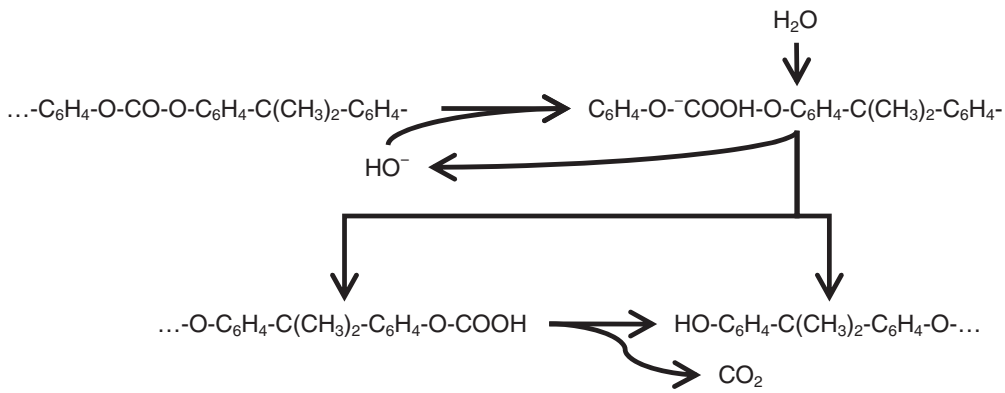

Figure 1 Degradation of polycarbonate into BPA and carbon dioxide catalysed by hydroxide.

Table 1 Overview of data of migration of BPA into food and other food simulants

Ref.	Samples	Pretreatment	Time/temperature	Simulant	Migration method	LoD (mg/L)	Results (mg/L)
Biedermann-Brem et al. (2008)	4 brands	n.r.	1 h @ 80°	3% citric acid (pH = 2) 3% phosphate/acetate (pH = 8) 3% sodium bicarbonate (pH = 9) 1% sodium carbonate (pH = 11) 0.1% sodium hydroxide (pH = 12)	100 mL, contact to all bottle surface by inclinated rotation in water bath	0.00001	0.0013 0.006 0.025 1.6 1.0
Maia et al. (2009)	1 brand (?)	Rinsing with distilled water and sterilization	1 h @ 120°C	Detergent solution (10 g/L)	Pieces (S/V = 20/dm); oven; rinsing of pieces with distilled water between migration tests	0.005	0.11 (1) 0.024 (2) 0.030 (3) 0.018 (4)
Mountfort (1997) in ECB (2003)		2 h sterilization, 3× rinsing	0.5 min @ 100 + 20 min @ 20°C	Fruit juice	250 mL; microwave (750 W)	0.03	<0.03 (3–20 cycles)
Japan (1998)	15 tableware	n.r.	1 h @ 60°C	n-heptane	n.r.	0.0005	<0.0005–0.001
Mountfort et al. (1997)		2 h sterilization, 3× rinsing	0.5 min @ 100 + 20 min @ 20°C	Milk formula	250 mL; microwave (750 W)	0.03	<0.03 (3–20 cycles)
EURL-FCM (2009)	11 brands	Hand washed with water + domestic detergent, rinsed with tap water, left to dry	2 h @ 70°C + 24 h@ 40°C	Milk formula	Preheated simulant used	0.03(LoQ)	<0.03 (1) <0.03 (2) <0.03 (3)
Bayer (1999) in ECB (2003)	24 bottles	n.r.	±30 days @ 2°C–8°C	Milk	n.r.	0.001	<0.001
Japan (1998)	30 tableware	n.r.	Filled @ 80°C + 30 min @ 20°C Filled @ 60°C + 30 min @ 20°C	Soup	n.r.	0.01	<0.01 <0.01

Numbers in brackets in the column results indicate n^{th} successive migration. LoD, limit of detection; LoQ, limit of quantification; n.r., not reported; S/V, surface-to-volume ratio.

Table 2 Overview of data of migration of BPA into oil or similar simulants

Ref.	Samples	Pretreatment	Time/temperature	Simulant	Migration method	LoD (mg/L)	Results (mg/L)
Wong et al. (2005)	28 brands	n.r.	8 h @ 100°C 3 days @ 100°C 10 days @ 100°C	corn oil	Rim (S/V = 6.45/dm); oven	0.0008 mg/dm^2	<0.005–0.26 <0.005–0.48 <0.005–0.65
Howe and Borodinsky (1998)	Test piece	n.r.	6 h @ 100°C 10 day @ 49°C	Miglyol®	Disc; S/V = 6.45/dm; oven	0.0008 mg/dm^2 (LoQ)	<0.005 <0.005
Biles et al. (1997)	1 brand	n.r.	10 day @ 65°C	Miglyol®	Test pieces; S/V = ±10/dm; oven; no shaking	n.r.	±0.03 (no shaking) ±1.5 (shaking)
Japan (1998)	30 tableware	n.r.	Filled with 60°C followed by 30 min @ 20°C	Olive oil	n.r.	0.05	<0.05
Yokohama study in ECB (2003)	1 brand	n.r.	24 h @ 50°C	Olive oil	Filled with half volume; water-bath horizontal shaker at 140 cycles/min	0.01	<0.01

LoD, limit of detection; LoQ, limit of quantification; n.r., not reported; S/V, surface-to-volume ratio.

Table 3 Overview of data of migration of BPA into ethanol

Ref.	Samples	Pretreatment	Time/temperature	Simulant	Migration method	LoD (mg/L)	Results (mg/L)
Biles et al. (1997)	1 brand	n.r.	10 day @ 65°C	8% ethanol	Test pieces; S/V = ±10/dm; oven; shaking	n.r.	±0.87
			30 min @ 100°C	10% ethanol	Test pieces; S/V = ±120/dm; oven; shaking		±2.5 (1) ±0.84 (2) ±0.48 (3) ±0.48 (4)
CSL (2004) in ECB (2008)	2 brands	Sterilization	1 h @ 70°C	10% ethanol	n.r.	n.r.	<0.0011–0.0045 (1) <0.0011 (2) <0.0011 (3)
Howe and Borodinsky (1998)	Test piece	n.r.	6 h @ 100°C	10% ethanol	S/V = 6.45/dm; oven	0.0008 mg/dm^2	<0.005
Wong et al. (2005)	28 brands	n.r.	8 h @ 70°C	10% ethanol	Rim (S/V = 6.45/dm); oven	0.0008 mg/dm^2	<0.005–0.058
Simoneau et al. (2000) in ECB (2003)	1 brand	n.r.	24 h @ 50°C	10% ethanol	Filled with half volume; water-bath horizontal shaker at 140 cycles/min	0.01	<0.01
Wong et al. (2005)	28 brands	n.r.	3 days @ 70°C	10% ethanol	Rim (S/V = 6.45/dm); oven	0.0008 mg/dm^2	<0.005–0.062
Howe and Borodinsky (1998)	Test piece	n.r.	10 days @ 49°C	10% ethanol	S/V = 6.45/dm; oven	0.0008 mg/dm^2 (LoQ)	<0.005
Biles et al. (1997)	1 brand	n.r.	10 days @ 65°C	10% ethanol	Test pieces; S/V = ±10/dm; oven; no shaking	n.r.	±0.91
Wong et al. (2005)	28 brands	n.r.	10 days @ 70°C	10% ethanol	Rim (S/V = 6.45/dm); oven	0.0008 mg/dm^2	<0.005–0.19
D'Antuono et al. (2001)	4 brands	Rinsing distilled water twice	2 h @ 80°C	15% ethanol	n.r.; S/V = 10/dm	0.0002 (HPLC-VD)	0.0025
Kawamura et al. (1998)	2 rice bowl	n.r.	30 min @ 60°C	20% ethanol	150 mL	0.0005	0.0017–0.0029
Japan (1998)	15 tableware	n.r.	30 min @ 60°C	20% ethanol	n.r.	0.0005	<0.0005–0.0006
EURL-FCM (2009)	6 bottles	n.r.	2 h @ 70°C	50% ethanol	n.r.	0.02	<0.02
	17 brands	n.r.	2 h @ 70°C		n.r.	n.r. (LC-MS-MS)	0.0012–0.0032 (3) or <0.0002–0.0004 mg/dm^2
	11 brands	Hand washed with water and domestic detergent, rinsed with tap water, left to dry	2 h @ 70°C + 24 h @ 40°C		Preheated simulant used	0.033 mg/L (LoQ)	<0.033–0.045 (1) <0.033–0.035 (2) <0.033 (3)
Kubwabo et al. (2009)	12 brands	Rinsing 3* with HPLC-grade water	8 h @ 40°C 24 h @ 40°C	50% ethanol	Oven	0.00000004 (GC-MS-MS)	0.00017 0.0015
VWA (2008)	30	n.r.	24 h @ 40°C	50% ethanol	EN 14350-2	0.0021	<0.0021
Kubwabo et al. (2009)	12 brands	Rinsing 3* with HPLC-grade water	10 day @ 40°C	50% ethanol	Oven	0.00000004 (GC-MS-MS)	0.0024
Biles et al. (1997)	1 brand	n.r.	10 day @ 65°C	50% ethanol	Test pieces; S/V = ±10/dm; oven; no shaking	n.r.	±5.9
Simoneau et al. (2000) in ECB (2003)	1 brand	n.r.	24 h @ 50°C	95% ethanol	Filled with half volume; water-bath horizontal shaker at 140 cycles/min	0.01	<0.01–0.11
Biles et al. (1997)	1 brand	n.r.	7 day @ 65°C	95% ethanol	Test pieces; S/V = ±10/dm; oven; no shaking	n.r.	±2.2

Numbers in brackets in the column results indicate nth successive migration. LoD, limit of detection; LoQ, limit of quantification; n.r., not reported; S/V, surface-to-volume ratio; VD, voltametric detector.

Table 4 Overview of data of migration of BPA into 3% acetic acid

Ref.	Samples	Pretreatment	Time/temperature	Simulant	Migration method	LoD (mg/L)	Results (mg/L)
EURL-FCM (2009)	7 brands	Washing with tap water and detergent (pH = 6.7), brushing, sterilization 10 min @ 100°C	30 min @ 100°C	3% acetic acid	Hot filling to capacity	0.0001	0.00028 (1) 0.0012 (2) 0.0058 (3)
Maragou et al. (2008)	6 brands	12 cycles of brushing with detergent; sterilization for 10 min @ 100°C	filled @ boiling + 45 min @ ambient	3% acetic acid	120–250 mL	0.0018	<0.0018
CSL (2004) in ECB (2008)	2 brands	Sterilization	1 h @ 70°C	3% acetic acid	n.r.	n.r.	<0.0003–0.0007 (1) <0.0003 (2) <0.0003 (3)
Maragou et al. (2008)	2 brands	10 cycles of machine dishwashing (60°C) with detergent; rinsing with distilled water; sterilization for 10 min @ 95°C	2 h @ 70°C	3% acetic acid	120–250 mL	0.0018	<0.0018
		4 cycles of hand dishwashing with brush and detergent; rinsing with distilled water; sterilization for 10 min @ 95°C					<0.0018
		1 cycles of rinsing with distilled water					<0.0018
EURL-FCM (2009)	5	n.r.	2 h @ 70°C	3% acetic acid	Between migrations cleaning with brush and sterilization 1 h @ 100°C	n.r.	<0.003 (2 samples) <0.03 (2 samples) <0.01 (1 sample)
	11 brands	Hand washed with water and domestic detergent, rinsed with tap water, left to dry	2 h @ 70°C + 24 h @ 40°C		Preheated simulant used	0.03 mg/L (LoQ)	<0.03 (1) <0.03 (2) <0.03 (3)
D'Antuono et al. (2001)	4 brands	Rinsing distilled water twice	2 h @ 80°C	3% acetic acid	n.r.; S/V = 10/dm	0.0002 (HPLC-VD)	0.0022
Earls et al. (2000) in ECB (2003)	21 new and 12 used bottles	Steam sterilization	filled @ boiling + 24 h @ 5°C + ?? @ 40°C	3% acetic acid	100 mL; refrigerator; boiling water bath (40°C)	0.01 (HPLC-DA)	<0.01 (new) <0.01–0.05 (used)
VWA (2008)	30	n.r.	24 h @ 40°C	3% acetic acid	EN 14350-2	0.0039	<0.0039
VWA (2005)	22	n.r.	24 h @ 40°C	3% acetic acid	EN 14350-2	0.0039	<0.0039 <0.0039 (used)
Sim and Jianhua (2008)	2	n.r.	24 h @ 40°C	3% acetic acid	EN 14350-2	0.03	<0.03
EURL-FCM (2009)	10	n.r.	24 h @ 40°C	3% acetic acid	EN 14350-2	0.01 (GC-MS)	<0.01
Simoneau et al. (2000) in ECB (2003)	1 brand	n.r.	24 h @ 50°C	3% acetic acid	Filled with half volume; water-bath horizontal shaker at 140 cycles/min	0.01	<0.01
Howe and Borodinsky (1998)	Test piece	n.r.	10 day @ 49°C	3% acetic acid	S/V = 6.45/dm; oven	0.0008 mg/dm^2 (LoQ)	<0.005
Japan (1998)	15 tableware	n.r.	30 min @ 60°C	4% acetic acid	n.r.	0.0005	<0.0005

Numbers in brackets in the column results indicate nth successive migration. DA, diode array detector; LoD, limit of detection; LoQ, limit of quantification; n.r., not reported; S/V, surface-to-volume ratio; VD, voltametric detector.

Table 5 Overview of data of migration of BPA into various types of water

Ref.	Samples	Pretreatment	Time/temperature	Simulant	Migration method	LoD (mg/L)	Results (mg/L)
Biedermann-Brem and Grob (2009)	bottle	n.r.	1 min @ 100°C	Tap water (pH 7.5; 37°f)	200 mL; microwave heating	0.0005	<0.00005
			1 min @ 100°C	Boiled tap water (pH 7.5; 37°f)			0.01
			2.5 min @ 100°C	Tap water (pH 7.5; 37°f)			<0.00005
			2.5 min @ 100°C	Boiled tap water (pH 7.5; 37°f)			0.022
			5 min @ 50°C	Tap water (pH 7.5; 37°f)			<0.00005
			5 min @ 50°C	Boiled tap water (pH 7.5; 37°f)			0.002
			5 min @ 55°C	Tap water (pH 7.5; 37°f)			<0.00005
			5 min @ 55°C	Boiled tap water (pH 7.5; 37°f)			0.004
			5 min @ 65°C	Tap water (pH 7.5; 37°f)			<0.00005
			5 min @ 65°C	Boiled tap water (pH 7.5; 37°f)			0.0075
			5 min @ 75°C	Tap water (pH 7.5; 37°f)			<0.00005
			5 min @ 75°C	Boiled tap water (pH 7.5; 37°f)			0.015
			5 min @ 100°C	Tap water (pH 7.5; 37°f)			0.0006–0.0015
			5 min @ 100°C	Tap water (pH 7.7; 35°f)			0.0025–0.0029
			5 min @ 100°C	Boiled tap water (pH 7.5; 37°f)			0.033–0.042
Ehlert et al. (2008)	18 brands	Immersion in boiling water for 5 min; Drying for 3 h	1 min after reaching 100°C + 8 min @ 20°C	HPLC-grade water	100 mL or 200 mL (bottles >200 mL) washing of emptied bottle 3 × 50 mL water for 15 sec + drying for 3 h between successive migration	0.0001 (SPE-GC-MS)	<0.0001–0.00049 (1) <0.0001–0.00073 (2) <0.0001–0.00030 (3)
Lim et al. (2009)	container	n.r.	Filled @ 100°C + 10 min @ 20°C	Boiled tap water	200 mL	0.001	0.0013
EURL-FCM (2009)	bottle	n.r.	10 min @ 100°C	Tap water (pH 8; 20°f–30°f)	microwave heating	0.00002	<0.00002–0.00038
Biedermann-Brem and Grob (2009)	bottle	n.r.	10 min @ 100°C	Tap water (pH 7.7; 35°f)	200 mL; microwave heating	0.0005	0.0075–0.0011
				Tap water (pH 7.5; 37°f)			0.023
				Boiled tap water (pH 7.5; 37°f)			0.14
Lim et al. (2009)	container	n.r.	Filled @ 100°C + 20 min @ 20°C	Boiled tap water	200 mL	0.001	0.0018
			Filled @ 100°C + 30 min @ 20°C				0.0022
Takahashi (1998) in ECB (2003)	2 bottles	n.r.	30 min @ 50°C	Water	200 mL	n.r.	0.0000008–0.000018
	1 mug		30 min @ 95°C				0.000056
Japan (1998)	15 tableware	n.r.	30 min @ 60°C	Water	n.r.	0.0005	<0.0005
	30 tableware		Filled @ 80°C + 30 min @ 20°C				<0.0005
Sun et al. (2000)	2 brands	n.r.	30 min @ 95°C	Boiling water		0.00038 (HPLC-CL)	0.00059–0.00075 (1) <0.00038 (2–4)
Yoshida et al. (2003)	4 brands	n.r.	30 min @ 95°C	Boiling milliQ water		0.0000005 (HPLC-CL)	0.000008–0.00019 (1) 0.000003–0.000052 (2) 0.000001–0.000018 (3) <0.0000005–0.000004 (4)
Kawamura et al. (1998)	4 bottles	n.r.	30 min @ 95°C	Water	100 mL; S/V = 9.1–12/dm 200 mL; S/V = 9.2–15	0.0005	<0.0005–0.0005

Kawamura et al. (1998)	3 mug				150 mL; S/V = 8.7/dm		<0.0005–0.026 (1) 0.0034 (2) 0.0024 (3) 0.0017 (4) 0.0015 (5)
	Measuring cup				500 mL; S/V = 5.6/dm		<0.0005
	2 rice bowl				150 mL; S/V = 6/dm		0.0032–0.0046
Takahashi (1998) in ECB (2003)	2 bottles 1 mug	n.r.	30 min @ 95°C	Water	200 mL	n.r.	0.0008–0.002 0.00076
Takao et al. (1998)	n.r.	n.r.	30 min @ 95°C	Water	n.r.	n.r. (SPME-GC-MS)	0.000001–0.0000035 (new) 0.000001–0.0000065 (old)
Chang et al. (2005)	3 bottles	n.r.	30 min @ 95°C + cooling to 20°C	milliQ water	150 mL	n.r. (SPME-GC-MS)	0.0006–0.0041
D'Antuono et al. (2001)	4 brands	Rinsing distilled water twice	30 min @ 100°C	Distilled water	n.r.; S/V = 10/dm	0.0002 (HPLC-VD)	0.0012
EURL-FCM (2009)	7 brands	Washing with tap water and detergent (pH = 6.7), brushing, sterilization 10 min @ 100°C	30 min @ 100°C	Deionized water	Filling to capacity	0.0001	0.00049–0.0091 (1) 0.00033–0.0061 (2) 0.00017–0.0058 (3) 0.00048–0.0066 (4)
Lázaro Martínez et al. (2009)	1 brand	Rinsing distilled water twice	30 min @ 100°C	Distilled water	250 mL; oven?	0.00014 (HPLC-ED)	0.0017 (1) 0.00052 (2)
				Distilled water?	250 mL; after 10 use cycles with boiling tap water (10 min), brushing and rinsing; oven?		0.0036
				tap water (pH 6.9; total alkalinity 59 mg/L)	250 mL; oven?		0.0037
				tap water(pH 7.6; total alkalinity 499 mg/L)	250 mL; oven?		0.0091
Krishnan et al. (1993)	1 brand	n.r.	30 min @ 120–125°C; total cycle time 75 min	Distilled water	500 mL; autoclave	n.r. (RA)	0.003
Maragou et al. (2008)	6 brands	5 cycles of brushing with detergent; sterilization for 10 min @ 100°C	filled @ 100°C + 45 min @ ambient	MilliQ water	120–250 mL	0.0024	0.0039–0.014 (1) 0.0026–0.013 (2) <0.0024–0.014 (3) <0.0024–0.013 (4) 0.0024–0.0044 (5) 0.0031–0.0053 (6) <0.0024–0.0039 (7) <0.0024–0.0048 (8) <0.0024 (9)
Biedermann-Brem et al. (2008)	4 brands	n.r.	1 h @ 80°	Tap water (pH 6)	100 mL in bottle, contact to all bottle surface by inclinated rotation in water bath	0.00001	0.020

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Table 5 Overview of data of migration of BPA into various types of water *(Continued)*

Ref.	Samples	Pretreatment	Time/temperature	Simulant	Migration method	LoD (mg/L)	Results (mg/L)
Lim et al. (2009)	container	n.r.	Filled @ 100°C + 1 h @ 20°C	Boiled tap water	200 mL	0.001	0.0025
Brede et al. (2003)	12 brands	Rinsing with boiling water	1 h @ 100°C	milliQ water	200 mL; in oven	n.r. (SPE-GC-MS)	0.00011–0.00043
Biedermann-Brem and Grob (2009)	bottle	n.r.	1 h @ 100°C	Tap water (pH 7.5; 37°f)	in bottle; heating in microwave	0.0005	0.086
EURL-FCM (2009)	17 brands	n.r.	1 h @ 100°C	Water		LC-MS-MS LOD = n.r.	0.006–0.078 mg/ L (3) or 0.0007–0.0092 mg/dm^2
	1 brand (?)	Rinsing with distilled water and sterilization	1 h @ 120°C	Distilled water	Pieces (S/V = 20/dm); oven; rinsing of pieces with distilled water between migration tests	0.005	0.11 (1) 0.024 (2) 0.030 (3) 0.018 (4)
Maragou et al. (2008)	2 brands	10 cycles of machine dishwashing (60°C) with detergent; rinsing with distilled water; sterilization for 10 min @ 95°C	2 h @ 70°C	MilliQ water	120–250 mL	0.0024	<0.0024
		4 cycles of hand dishwashing with brush and detergent; rinsing with distilled water; sterilization for 10 min @ 95°C					<0.0024
		1 cycles of rinsing with distilled water					<0.0024
D'Antuono et al. (2001)	4 brands	Rinsing distilled water twice	2 h @ 80°C	Distilled water	n.r.; S/V = 10/dm	0.0002 (HPLC-VD)	0.0011
Lim et al. (2009)	container	n.r.	Filled @ 100°C + 2 h @ 20°C	Boiled tap water	200 mL	0.001	0.0026
Biedermann-Brem and Grob (2009)	bottle	n.r.	2 h @ boiling	Tap water (Berlin)	200 mL; microwave heating	0.0005	0.088–0.16
Lim et al. (2009)	container	n.r.	Filled @ 100°C + 3 h @ 20°C	Boiled tap water	200 mL	0.001	0.0026
EURL-FCM (2009)	6 brands	Washing with tap water and detergent (pH = 6.7), brushing, sterilization 10 min @ 100°C	4 h @ 100°C	Deionized water	Filling to capacity	0.0001	0.0061–0.026 (1) 0.0094–0.061 (2) 0.0094–0.059 (3)
Hanai (1997) in ECB (2003)	6 brands	n.r.	5 h @ 26°C	Purified water		0.002 (GC-MS)	<0.002
Howe and Borodinsky (1998)	Test piece	n.r.	6 h @ 100°C	Water	S/V = 6.45/dm; oven	0.0008 mg/dm^2 (LoQ)	<0.005
Kubwabo et al. (2009)	12 brands	Rinsing 3* with HPLC-grade water	8 h @ 40°C	HPLC-grade water	Oven	0.00000004 (GC-MS-MS)	0.00011

Biedermann-Brem and Grob (2009)	bottle	n.r.	Overnight @ ambient	Tap water (pH 7.4; 22°f)	200 mL preboiled water in bottle	0.0005	0.006
Hanai (1997) in ECB (2003)	6 brands	n.r.	Filled @ 95°C + overnight @ ambient	Purified water		0.002 (GC-MS)	0.0031–0.055
Earls et al. (2000) in ECB (2003)	21 new and 12 used bottles	Steam sterilization	filled @ boiling + 24 h @ 5°C + ?? @ 40°C	Water	100 mL; refrigerator; boiling water bath (40°C)	0.01 HPLC-DA	<0.01 (new) <0.01–0.05 (used)
Le et al. (2008)	1 brand new + old bottles	Rinsing with 1 l of distilled water; Brushing with alkaline detergent; 6 rinses with 1 l of distilled water; 2 rinses with 100 mL HPLC-grade water; 3 rinses with 100 mL methanol	24 h @ 22°C Bottle heated to 100°C + filled @ 22°C + 24 h @ 22°C filled @ boiling + 24 h @ 22°C	HPLC-grade water	100 mL; horizontal rotation of bottle (S/V = 48/dm)	0.00005 (ELISA)	0.00008–0.00036 0.0023–0.0046 (new) 0.00066 (used) 0.0038–0.0077 (new) 0.0019 (used)
Cao and Corriveau (2008b)	5 brands	Rinsing with water	filled @ boiling + 24 h @ 22°C	Boiling tap water	Filled to max. capacity	0.0005 (SPME-GC-MS)	0.0017–0.0041
Li et al. (2010)	4 brands	n.r.	24 h @ 24°C	milliQ water	240 mL; water bath; AL-foil against photolysis	0.0000007 (SPE-GC-MS)	<0.0000007–0.00027
Tan and Mustafa (2003)	? brands	Rinsing with distilled water	24 h @ 25°C filled @ 80°C + 24 h @ 25°	Distilled water	100 or 250 mL	0.001 (GC-MS)	dl-0.0000011 mg/ dm^2 (new) 0.000002–0.0010 mg/dm^2 (used) dl-0.00013 mg/ dm^2 (new) 0.000011–0.0026 mg/dm^2 (used)
VWA (2005)	22	Instruction	24 h @ 40°C	Water	EN 14350-2	0.0025	<0.0025 <0.0025–0.0050 (used)
Sim and Jianhua (2008)	2	n.r.	24 h @ 40°C	Water	EN 14350-2	0.03	<0.03
EURL-FCM (2009)	6 brands	Washing with tap water and detergent (pH = 6.7), brushing, sterilization 10 min @ 100°C	24 h @ 40°C	Deionized water	EN 14350-2	0.0001	<0.0001–0.00014
Kubwabo et al. (2009)	12 brands	Rinsing 3* with HPLC-grade water	24 h @ 40°C	HPLC-grade water	Oven	0.00000004 (GC-MS-MS)	0.00012
Li et al. (2010)	4 brands	n.r.	24 h @ 40°C	milliQ water	240 mL; water bath; AL-foil against photolysis	0.0000007 (SPE-GC-MS)	0.000009–0.00099
ECB (2003)	1 brand	n.r.	24 h @ 50°C	Water	Filled with half volume; water-bath horizontal shaker at 140 cycles/min	0.01	<0.01

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Table 5 Overview of data of migration of BPA into various types of water *(Continued)*

Ref.	Samples	Pretreatment	Time/temperature	Simulant	Migration method	LoD (mg/L)	Results (mg/L)
Kubwabo et al. (2009)	12 brands	Rinsing 3* with HPLC-grade water	filled @ boiling + 24 h @ 60°C	HPLC-grade water	Oven	0.00000004 (GC-MS-MS)	0.0018 (range 0.0005–0.0065)
Cao and Corriveau (2008a)	3 baby bottles 2 reusable water bottles	rinsing with boiling water	filled @ boiling + 24 h @ 70°C	Tap water (pH ±7)	in oven	0.0005 (SPME-GC-MS)	0.032–0.055
Li et al. (2010)	4 brands	n.r.	2 h @ 100°C + 24 h @ 24 °C	milliQ water	240 mL; water bath; AL-foil against photolysis	0.0000007 (SPE-GC-MS)	0.000034–0.0045
			2 days @ 24°C				0.0000019–0.000084 (1) <0.0000007–0.000033 (2) <0.0000007–0.000051 (3)
			2 days @ 40°C				0.000024–0.00024 (1) 0.000038–0.00025 (2) 0.000001–0.00016 (3)
Cao and Corriveau (2008a)	3 baby bottles 2 reusable water bottles	rinsing with boiling water	filled @ boiling + 2 days @ 70°C	Tap water (pH ±7)	in oven	0.0005 (SPME-GC-MS)	0.061–0.10
Le et al. (2008)	1 brand new and old bottles	Rinsing with 1 l of distilled water; Brushing with alkaline detergent; 6 rinses with 1 l of distilled water; 2 rinses with 100 mL HPLC-grade water; 3 rinses with 100 mL methanol	3 days @ 22°C	HPLC-grade water	100 mL and horizontal rotation of bottle (S/V = 48/dm)	0.00005 (ELISA)	0.00025–0.00079
Cao and Corriveau (2008a)	3 baby bottles 2 reusable water bottles	rinsing with boiling water	filled @ boiling + 3 days @ 70°C	Tap water (pH ±7)	in oven	0.0005 (SPME-GC-MS)	0.099–0.21
Le et al. (2008)	1 brand new and old bottles	Rinsing with 1 l of distilled water; Brushing with alkaline detergent; 6 rinses with 1 l of distilled water; 2 rinses with 100 mL HPLC-grade water; 3 rinses with 100 mL methanol	5 days @ 22°C	HPLC-grade water	100 mL and horizontal rotation of bottle (S/V = 48/dm)	0.00005 (ELISA)	0.00028–0.00072
Cao and Corriveau (2008a)	3 baby bottles 2 reusable water bottles	rinsing with boiling water	filled @ boiling + 6 days @ 70°C	Tap water (pH ±7)	in oven	0.0005 (SPME-GC-MS)	0.22–0.52

Le et al. (2008)	1 brand new and old bottles	Rinsing with 1 l of distilled water; Brushing with alkaline detergent; 6 rinses with 1 l of distilled water; 2 rinses with 100 mL HPLC-grade water; 3 rinses with 100 mL methanol	7 days @ 22°C	HPLC-grade water	100 mL and horizontal rotation of bottle (S/V = 48/dm)	0.00005 (ELISA)	0.00034–0.0013
Mountfort (1997) in ECB (2003); Mountfort et al. (1997)	24 brands	2 h sterilization, 3× rinsing	10 days @ 40°C	Distilled water	250 mL; microwave (750 W)	0.03	<0.03
Kubwabo et al. (2009)	17 brands	Rinsing 3* with HPLC-grade water	10 days @ 40°C	HPLC-grade water	Oven	0.00000004 (GC-MS-MS)	0.0019–0.0047
Howe and Borodinsky (1998)	Test piece	n.r.	10 day @ 49°C	Water	S/V = 6.45/dm; oven	0.0008 mg/dm^2 (LoQ)	<0.005
Biles et al. (1997)	1 brand		10 day @ 65°C	Water	Test pieces; S/V = ±10/dm; oven	n.r.	±0.23 (no shaking) ±1.0 (shaking)
	5 gallon carboy (±19 L)	n.r.	21 days @ ambient		no shaking		0.0000001–0.0000002
Mercea (2009)	8 bottles	n.r.	54 days @ 54°C	Deionized water	n.r.; heating in oven	0.001	0.10–0.15
Biles et al. (1997)	5 gallon carboy (±19 L)	n.r.	84 days @ ambient 273 days @ ambient	Water	no shaking	n.r.	0.0000004–0.0000005 0.0000046–0.0000047
Szymañski et al. (2006)	3 bottles	No	Ambient during storage	Mineral water		n.r.	0.00040–0.00052

Numbers in brackets in the column results indicate nth successive migration. CL, chemiluminescence detector; DA, diode array detector; ED, electrochemical detector; ELISA, enzyme-linked immunosorbent assay; LoD, limit of detection; LoQ, limit of quantification; n.r., not reported; (n), RA, radioreceptor assay; SPE, solid phase extraction; SPME solid phase microextraction; S/V, surface-to-volume ratio; VD, voltametric detector.

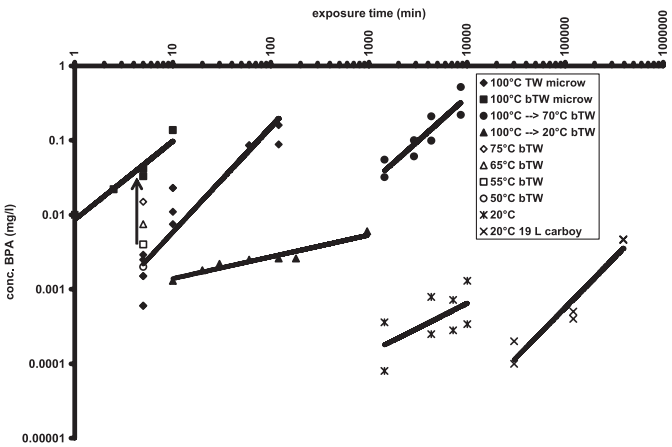

Figure 2 Release of BPA from baby bottles into various types of water as function of the contact time (Biles et al., 1997; Le et al., 2008; Cao and Corriveau, 2008a; Biedermann-Brem and Grob, 2009; Lim et al., 2009). bTW, boiled tap water; microw, microwave; TW, tap water.

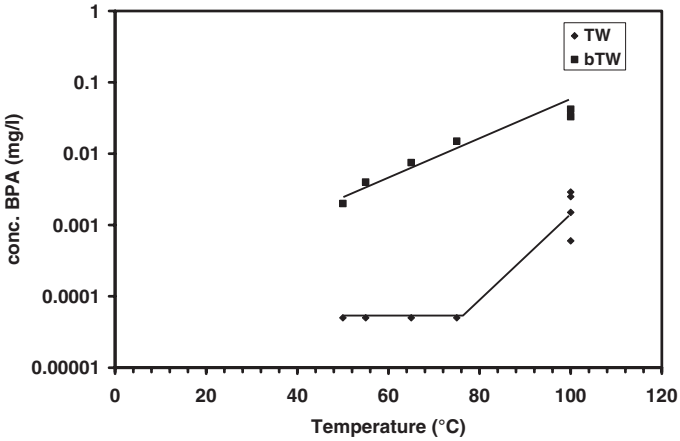

Figure 3 Release of BPA from baby bottles into various types of water as function of the temperature for 5 min in a microwave (Biedermann-Brem and Grob, 2009). bTW, boiled tap water; TW, tap water.

The release of BPA from polycarbonate into deionized water and tap water was studied in detail by many research groups (Table 5) (Krishnan et al., 1993; Biles et al., 1997; Mountfort et al., 1997; Howe and Borodinsky, 1998; Japan, 1998; Kawamura et al., 1998; Takao et al., 1998; Sun et al., 2000; D'Antuono et al., 2001; Brede et al., 2003; ECB, 2003; Tan and Mustafa, 2003; Yoshida et al., 2003; Chang et al., 2005; VWA, 2005; Szymanski et al., 2006; Biedermann-Brem et al., 2008; Ehlert et al., 2008; Le et al., 2008; Maragou et al., 2008; Sim and Jianhua, 2008; Cao and Corriveau, 2008a, 2008b; Biedermann-Brem and Grob, 2009; EURL-FCM, 2009; Kubwabo et al., 2009; Lázaro Martínez et al., 2009; Lim et al., 2009; Maia et al., 2009; Mercea, 2009). The highest concentration reported was 1 mg/L at 65°C for 10 days (Biles et al., 1997). Figure 2 shows the available data on the influence of the contact time. The figure shows that microwave heating enhances the release of BPA compared to heating in an oven. Using preboiled water instead of cold tap water in a microwave for boiling also increases the release of BPA. The effect of the temperature on preboiled water in a microwave is shown too. The effect of surface-to-volume ratio is shown by the comparison between baby bottles and carboys of 19 L.

Very few studies have analyzed release at more than two temperatures. Biedermann-Brem and Grob (2009) systematically studied the effect of temperature on the release of BPA into tap water and boiled tap water of the same water supply by heating in a microwave for 5 min. The concentration of BPA in tap water increased from <0.0001 mg/L at 50°C to 0.0006 mg/L at boiling temperature whereas the concentration of BPA in boiled tap water having a pH of about 9.5 increased from <0.002 mg/L at 50°C to 0.033 mg/L at boiling temperature (Figure 3).

The increase of the release of BPA by increasing the pH is clearly shown in Figure 4 (Biedermann-Brem et al., 2008). The effect of pH on the release of BPA is independent of other physical considerations such as exposure time, temperature and heating mode (microwave/thermal oven). This has special relevance if one considers that some food preparation processes may cause an increase of the pH above 8, which is normally the highest value for food. An example is boiling tap water in a pan or microwave for several minutes during which carbon dioxide can be released, consequently increasing the pH.

Biedermann-Brem and Grob (2009) and Mercea (2009) also indicated that the mineral content may also influence release. However, they failed to show clear evidence that the effect was really caused by the mineral composition and not by the pH.

Some scientists studied the difference between release of BPA by new and by used baby bottles and came to contradictory conclusions (Tan and Mustafa, 2003; Le et al., 2008; Mercea, 2009). Furthermore, the authors did not report whether the used bottles were of the same production lot as the new ones, so their experiments cannot be taken as conclusive on whether the release of BPA would decrease or increase with age of the bottle.

Repeated testing with the same bottles clearly demonstrated that the release decreased or at least remained constant during successive release experiments with water (Figure 5). The same observation was made with 10% aqueous ethanol solutions (Biles et al., 1997; CSL (2004) in ECB, 2008; EURL-FCM,

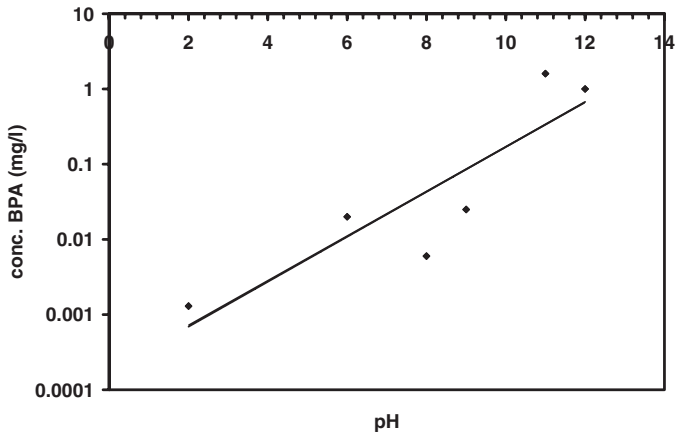

Figure 4 Release of BPA from baby bottles into water as function of the pH at 80°C for 60 min in an oven (Biedermann-Brem et al., 2008).

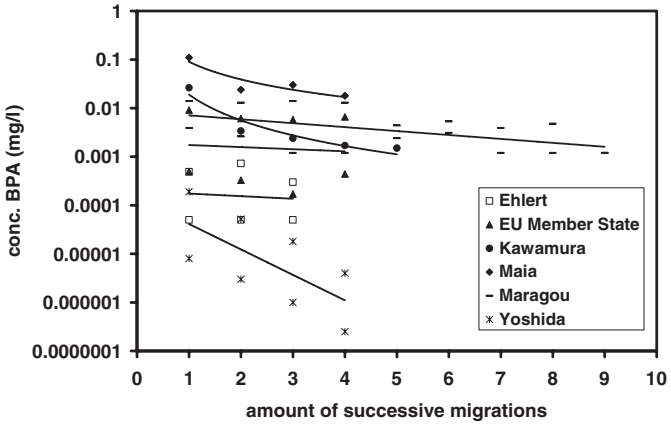

Figure 5 Release of BPA from baby bottles into water as function of the amount of successive migrations at various conditions of temperature and contact time (Kawamura et al., 1998; Yoshida et al., 2003; Ehlert et al., 2008; Maragou et al., 2008; EURL-FCM, 2009; Maia et al., 2009).

2009). One Member State reference laboratory of the European Union (EU) (EURL-FCM, 2009) observed an increase of the release of BPA into 3% acetic acid during successive experiments at 100°C for 30 min.

Washing the bottles also induces chemical ageing of the internal surface, but experimental results on this topic are not homogeneous. Both Biedermann-Brem et al. (2008) and Mercea (2009) observed that the bottles released lower amounts of BPA into aqueous food simulants after washing. In contrast, Brede et al. (2003) observed an increase. To explain the discrepancies, it has been speculated that the pH of the detergent solution could be crucial, but the rinsing after washing before drying the bottle may also have an effect. Manual brushing for cleaning the bottles does not raise the release of BPA (Maragou et al., 2008).

Philo et al. (1994) studied the stability of BPA at a level of 6 mg/L in distilled water, 3% acetic acid, 15% ethanol, and olive oil. BPA seems not to be stable in 3% acetic acid at 100°C for 1 h, however, it should be noted that the quantification method for BPA is not clear from the paper.

DISCUSSION ON EUROPEAN LEGISLATIVE ASPECTS AND EXPOSURE

According to the European Commission Directive 2002/72/EC relating to plastic materials and articles intended to come into contact with foodstuffs (and its amendments), test results of fillable articles that have a volume below 500 mL should be expressed as mg/dm^2 (EC, 2002). In case such articles are intended for infants (1–12 months) and young children (1–3 years), the results need to be expressed in mg/kg food. Baby bottles are repeated use articles. The migration test shall be carried out three times on a single bottle using fresh food or food simulant on each occasion. The bottle is compliant for BPA when the migrated concentration in the third test is below the migration limit of 0.6 mg/kg food. No second and third tests are necessary if there is conclusive proof that the level of the migration does not increase in the second and third tests and if the migration limit is not exceeded by the first test.

Table 6 Reported release of BPA (mg/kg food) from baby bottles tested according conditions set in European food contact material legislation

Food simulant	2 h @ 70°C	10 days @ 40°C
Water	<0.0024 [$]	0.0019–0.0047 [$]
3% acetic acid (juices)	<0.0018 [$]	<0.005 [$,*]
50% ethanol (liquid milk)	0.0012–0.0032 [&]	0.0024[$]
Reconstituted milk powder	<0.03 [$,#]	n.r.

[$]first migration; [&]third migration; *49°C instead of 40°C; [#]2 h @ 70°C + 24 h @40°C; n.r., not reported in literature.

Baby bottles are also a special case since they come in contact with food at temperatures in the range of 70–100°C for periods shorter than 15 min, so called "hot fill" conditions. In that case, the bottle can be tested at 70°C for 2 h according to amended Directive 82/711/EEC (EEC, 1982). However, if the material or article is intended to be used also for storage at room temperature, the above mentioned test is replaced by a test at 40°C for 10 days, which is considered to be more severe. It should be noted that the severity of the test conditions as mentioned in the Directive 82/711/EEC is based on knowledge on migration by diffusion. However, this statement may not be valid for other types of release such as hydrolysis of the polymeric structure and as such also not for the release of BPA from polycarbonate.

Table 6 shows the very few data that are available for the test conditions of 2 h at 70°C and 10 days at 40°C (Howe and Borodinsky, 1998; Maragou et al., 2008; EURL-FCM, 2009; Kubwabo et al., 2009). Only one data set shows the results of the third migration. All data in Table 6 comply with the European migration limit of 0.6 mg/kg. EU Member State official control data often have a relative high limit of detection in the range of 0.01–0.03 mg/L because the purpose of their analysis is to check the compliance of the polycarbonate bottle against the migration limit.

In the past, some EU official control laboratories and private test laboratories have used the European Norm EN 14350-2 (2004) covering drinking equipment for child use and care articles (VWA, 2005, 2008; Sim and Jianhua, 2008; EURL-FCM, 2009). This standard has no legal basis in the EU because the General Product Safety Directive (EC, 2001) states that "where products are subject to specific safety requirements imposed by Community legislation, this Directive shall apply only to the aspects and risks or categories of risks not covered by those requirements." It is interesting to note that this norm EN 14350-2 (2004) tests against a migration limit of 0.03 mg/L of BPA, which is a 20-fold lower value than the current migration limit in the amended Directive 2002/72/EC (EC, 2002). The latter (0.6 mg/kg) is based on the average body weight of adults of 60 kg, the consumption 1 kg of food and a tolerable daily intake of 0.01 mg/kg bw (EC, 2002). It seems plausible that the lower migration limit took into account a body weight of a newborn of about 3 kg instead of that of an adult.

The test conditions in amended Directive 82/711/EEC might not reflect the realistic worst-case release of BPA from polycarbonate, since they do not account for releases owing to hydrolysis in presence of hydroxide. For this purpose, one has to compare the release of BPA using the legal compliance test conditions with those of all possible exposure scenarios for babies fed using polycarbonate bottles. One can:

(1) pour boiling water into the sterilized bottle to which powdered infant formula is added; leave the bottle to cool down for 15 min.
(2) boil water in a sterilized bottle up to 5 min in a microwave, add infant formula and leave it to cool down for 15 min.
(3) sterilize the bottle filled with water up to 5 min in a microwave, add infant formula after adjusting the volume of the hot water in the bottle and leave it to cool down for 15 min.

The three exposure scenarios can also be performed with water that was already boiled for a previous milk preparation having a higher pH. From an exposure point of view, scenarios 2 and 3 are not different. Scenario 3 is time saving.

For exposure scenario 1, there are no data available for the milk simulant 50% ethanol. There are data available for water, which was the milk simulant until 2007. Present data seem not show the importance of the length of the cool down period. Maragou et al. (2008) observed concentrations in the range of 0.0039–0.014 mg/L (first migration), 0.0026–0.013 mg/L (second migration), and <0.0024–0.014 mg/L (third migration) for a cool down period of 45 min, whereas Le et al. (2008) and Cao and Corriveau (2008b) found concentrations in the range of 0.0017–0.0077 mg/L (first migration) for a cool down period of 24 h. The maximum concentration of the third migration is 0.014 mg/L and is higher than the concentration observed using the test conditions set under EU food contact material legislation in Table 6.

For exposure scenario 2 and 3 there are no experiments in literature that simulate this contact. Biedermann-Brem and Grob (2009) boiled tap water and previously boiled tap water as food simulant during 5 min in a microwave resulting in concentrations in the range of 0.0006–0.0029 and 0.033–0.042 mg/L, respectively (Biedermann-Brem and Grob, 2009). Using preboiled tap water results in 10-fold higher concentrations than the concentration observed for the test requirements set in European food contact material legislation in Table 6. For 50% ethanol it is difficult to estimate the migration for 5 min at 100°C in a microwave since there are no relevant data available.

The results for exposure scenario 1 indicate that the European "hot fill" derogation, that is, the actual contact time of the article at 70°C–100°C is less than 15 min and therefore, the article can be tested at 70°C for 2 h, might not be valid for polycarbonate (because of diffusion + hydrolysis). In that case, the article needs official testing at 100°C for 30 min. D'Antuono et al. (2001), EURL-FCM (2009), and Lázaro Martínez et al. (2009) observed concentrations in water in the range of 0.0005–0.0091 mg/L for the first migration. EURL-FCM (2009) found 0.0005–0.0058 mg/L for the third migration. The maximum concentration in water of the third migration is 0.0058 mg/L and is slightly higher than the concentration observed for the test requirements set in European food contact material legislation in Table 6.

Boiling 10% ethanol for 30 min results in a concentration of 0.48 mg/L for the third migration (Biles et al., 1997). Since these data are based on an unrealistic surface-to-volume (S/V) ratio of 120/dm, this concentration needs to be corrected for a more realistic S/V for baby bottles of about 8/dm, being roughly 0.03 mg/L. Biles et al. observed that the concentration increases with increasing ethanol content of the simulant. The maximum concentration in 50% ethanol of the third migration will be much higher than the concentration observed for the test requirements set in European food contact material legislation in Table 6.

Recently, Maia et al. (2010) reported the aminolysis of polycarbonate into BPA by some amines. It may be relevant process for biogenic amines present in milk. Solutions of 600 mg/L of 1,4-diaminobutane and 1,3-cyclohexane-bis(methylamine) showed clear aminolysis at 121°C for 1 h, whereas trimethylamine and 1,3-xylylenediamine were less effective. The pH effect (10.4–11.4) was excluded by comparing with relevant sodium hydroxide solutions. During contact at 25°C for 5 days aminolysis was not detectable.

Aminolysis of polycarbonate is also possible with 50 mM solutions of glycine and methionine adjusted to pH 11 by sodium hydroxide at lower temperatures of 37°C (Sajiki and Yonekubo, 2004). The migration rates are in the range of 0.38–0.49 mg/L/day and, corrected for the realistic surface-to-volume ratio of baby bottles of about 8/dm, 0.11–0.14 mg/L/day. These migration rates are much lower than those that can be calculated from Maia et al. (2010) that are in the range of 48–12,000 mg/L/day or corrected for S/V, 19-5000 mg/L/day. The data of Sajiki and Yonekubo indicate that amino acids may result in about 0.01 mg/L of BPA at pH 11 and 37°C for 30 min. contact.

The few data on migration of BPA into milk that exist, were below the limit of detection of 0.03 mg/L. It cannot be concluded from present data whether aminolysis contributes to the release of BPA into milk or not. However, it is clear that only a few substances might have an effect and the question is whether these substances are present in milk at relevant concentrations.

Another relevant depolymerization process that might influence the release of BPA from polycarbonate during migration testing with 50% ethanol is ethanolysis. Methanolysis is an important process to recycle BPA under mild conditions (Liu et al. 2009, 2010). Under supercritical conditions (243°C and 6.4 MPa) polycarbonate is converted by ethanol into BPA and diethyl carbonate (Jie et al., 2006). Glycol is able to depolymerize polycarbonate at 180°C using sodium hydroxide as catalyst (Oku et al., 2000). The question arose whether ethanol is also able to degrade polycarbonate under milder conditions and whether 50% ethanol is a suitable food simulant of milk for polycarbonate. Fact is that the release of BPA increases with the

level of ethanol in aqueous solution in the range of 8%–50% (Biles et al., 1997). This can indicate that BPA dissolves better by increasing ethanol concentration, that ethanol depolymerizes polycarbonate or that both processes occur in parallel.

CONCLUSIONS

Our review on the release of BPA from polycarbonate and our considerations on exposure and European legislative aspects related to food contact materials, leads us to the following conclusions:

- Release of BPA from polycarbonate into aqueous liquids is caused by diffusion and hydrolysis of polycarbonate catalyzed by hydroxide;
- Main parameters affecting the release of BPA are contact time, temperature, and pH of the food simulant, all having a positive correlation;
- Dissolution of scale during boiling of water in the polycarbonate bottle may raise the concentration of BPA due to the rise of the pH;
- Brushing of the bottle does not seem to raise the release of BPA;
- The effect of aging is difficult to estimate, as studies suggesting this effect were not based on experiments with new and used bottles of the same production lot;
- Residual alkaline detergent remaining on the surface of the baby bottle after dishwashing may increase the release of BPA;
- Some food preparation processes cause an increase of the pH above the normal pH of food;
- The possible effect of the mineral composition of the aqueous food simulant on the release of BPA is not clear to date;
- There are very few migration data of BPA available for the legal European test conditions. They all comply with the migration limit of the European Directive;
- Release data of BPA seems to indicate that the European "hot fill" derogation may not be applicable to polycarbonate;
- Use conditions that reflect more realistic worst-case use of polycarbonate baby bottles compared to the test conditions set in European food contact material legislation were identified. This review suggests further investigations on migration data confirming and establishing realistic worst-case exposure concentrations;
- A study on the composition of milk combined with migration tests should reveal whether aminolysis might be relevant for compliance testing of polycarbonate and whether food similants can still replace milk;
- A study on ethanolysis should reveal whether 50% ethanol can be used as food simulant in contact with polycarbonate.

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