

Safety of Polycarbonate Water Carboys for Residual and Migration Levels of Bisphenol-A

Özlem Kızıllırmak Esmer¹  ✉ Özlem Çağındı² ¹Ege University, Engineering Faculty, Food Engineering Department 35100 Bornova, İzmir, Turkey²Manisa Celal Bayar University, Engineering Faculty, Food Engineering Department, Muradiye, Manisa, Turkey

Received (Geliş Tarihi): 16.12.2020, Accepted (Kabul Tarihi): 30.12.2020

✉ Corresponding author (Yazışmalardan Sorumlu Yazar): ozlem.kizilirmak@ege.edu.tr (Ö. Kızıllırmak Esmer)

☎ +90 232 311 30 20 📠 +90 248 213 30 99

ABSTRACT

Polycarbonate (PC) materials are widely used for the packaging of drinkable water. The use of PC materials as food packaging raises some concerns for the migration of Bisphenol A (BPA), which is the monomer of PC. Therefore, in this research, it was aimed to determine the quality of new (not used before) and old (life time ended) PC water carboys in terms of residual BPA and BPA migration levels with respect to storage conditions. For this purpose, BPA migration was determined by storing at three different temperatures (5, 20 and 40°C) for seven different storage times (1., 5., 10., 20., 30., 45. and 60 days.) Residual and migrated BPA levels were determined by High Performance Liquid Chromatography with Fluorescence Detector. As a result, PC water carboys contained low levels of residual of BPA, and lower or higher residual BPA levels were detected in old materials compared to new materials. The BPA migration level increased by temperature and time, and storage under sunlight increased the level of BPA migration. BPA migration levels under all storage conditions were below the specific migration limit for BPA. In order to reach the tolerable daily intake of BPA even under the worst storage condition of PC water carboys, daily water consumption was calculated as 16 liters for a person with a body weight of 60 kg. On the other hand, this daily water consumption might not be rational for a person. Therefore, it was concluded that PC water carboys possessed no risk in terms of BPA migration levels under the conditions studied.

Keywords: Bisphenol A (BPA), PC water carboys, migration of BPA, residual BPA

Polikarbonat Su Damacanelerinin Bisfenol-A Kalıntı Miktar ve Migrasyonu Açısından Güvenliği

ÖZ

Polikarbonat (PC) malzemeler, sahip olduğu özellikleri itibarıyla içme sularının ambalajlanmasında oldukça yaygın olarak kullanılan malzemelerdir. PC malzeme; monomeri olan Bisfenol-A (BPA)'dan dolayı gıda ambalajı olarak kullanımına dair BPA migrasyonu açısından bazı endişeler yaratmaktadır. Bu nedenle bu çalışmada yeni (hiç kullanılmamış) ve eski (kullanım ömrünü tamamlamış) PC su damacanelerinin kalıntı BPA miktarları açısından kalitelerinin değerlendirilmesi; depolama sıcaklığı ve depolama süresinin BPA migrasyonu üzerine etkisinin belirlenmesi amaçlanmıştır. BPA kalıntı analizi ve BPA migrasyon analizi Floresans Dedektörlü Yüksek Basınç Sıvı Kromatografisi kullanılarak gerçekleştirilmiştir. Bu amaçla, üç farklı temas sıcaklığı (5, 20 ve 40°C) ve yedi farklı depolama süresinde (1., 5., 10., 20., 30., 45. ve 60. gün) depolama yapılarak BPA migrasyon miktarları belirlenmiştir. Çalışma sonucunda PC damacana örneklerinin BPA kalıntı miktarlarının düşük olduğu ve damacanelerin eski olması durumunda yeni damacanelere göre daha düşük ya da daha yüksek kalıntı BPA miktarlarına sahip olabileceği tespit edilmiştir. BPA migrasyon miktarlarının ise sıcaklığa ve süreye bağlı olarak artış gösterdiği ve damacaneleri güneş ışığı altında depolamanın BPA migrasyon miktarlarını artırdığı tespit edilmiştir. Tüm depolama koşullarında tespit edilen BPA migrasyon miktarlarının, BPA için belirtilen spesifik migrasyon limitinin oldukça altında olduğu belirlenmiştir. 60 kilogram ağırlığındaki bir kişinin BPA'nın tolere edilebilir günlük alım miktarına (4 mikrogram/ 60kg vücut ağırlığı-gün) ulaşmak

için, en kötü depolama koşulunda (40°C/60 gün) bir günde 16 litre su tüketmesi gerektiği hesaplanmıştır. Ancak günlük koşullar dikkate alındığında bu miktarda su içilmesi olası değildir. Bu nedenlerle, gerçekleştirilen bu çalışma sonucunda, belirtilen koşullar altında kullanıldığında polikarbonat su damacanelerinin BPA migrasyonu açısından bir risk teşkil etmediği belirlenmiştir.

Anahtar Kelimeler: Bisfenol A (BPA), Polikarbonat su damaneleri, BPA migrasyonu, kalıntı BPA

INTRODUCTION

Polycarbonate (PC) has an important place among plastics due to its numerous advantageous characteristics such as excellent transparency, impermeability, high thermal stability, and mechanical strength and, therefore, it has a rather broad usage area. PC, which is used commonly in the production of materials contacting foods, especially 5-gallon water bottles and tableware. The key building block of PC is bisphenol A (BPA; 2,2'-bis(4-hydroxyphenyl)propane [1] and BPA may be found as a residue in the materials. Recent studies have brought up that BPA threatens public health by migrating from polycarbonate materials to food products. It is known that BPA migration occurs from PC materials and consumers may be exposed to BPA significantly in their daily diets [1; 2; 3; 4; 5]. Although there are many studies about the negative effects of BPA on health and some concerns expressed in this regard, EFSA CEF Panel concluded that there is no health concern for any age group from dietary exposure and low health concern from aggregated exposure [6]. Indeed, due to the possible negative effects of BPA on health, the use of this monomer in the production of PC materials used for infants and its use as an additive in the plastic materials coming into contact with foodstuffs have been prohibited with the Commission Regulation No 10/2011 [7].

The amount of migrated BPA from PC materials may increase due to PC degradation especially at high temperatures and in contact with water [8; 9; 10; 11; 12; 13; 14; 15; 16; 17; 18; 19] or owing to the impact of environmental conditions or the repetitive processes such as washing, sterilization in the PC materials reused for a long time [1; 10; 20; 21; 22; 23]. As a result, a higher amount of BPA migration may occur compared to the initial amount of residual BPA [12; 20]. In 5-gallon PC water bottles used to store potable water, water can be kept under ambient conditions for a long time and at high temperatures depending on the seasonal conditions. Also, as 5-gallon PC water bottles are reusable materials, they are exposed to the washing process many times. The number of the studies in relation to the BPA migration from the reused food packaging materials is quite limited and no study focusing on the amount of residual BPA in the 5-gallon PC water bottles have been found so far.

This study aimed to determine (i) the amounts of the residual BPA in the 5-gallon PC water bottles provided by several companies in Turkey, (ii) whether the usage period for the end of life samples affected the amount of residual BPA, (iii) whether there was a statistically significant difference between the BPA levels of the upper, the main body, and the bottom parts of the 5-

gallon PC water bottles, and (iv) the effect of the storage temperature, storage period and storage under sunlight on the level of BPA migration.

MATERIALS AND METHODS

Materials

The samples for analysis were provided by five different 5-gallon PC water bottle production companies. The samples for residual analysis coded with N and O meaning new (not used before) and old (life time ended) respectively. The sample having the highest residual BPA concentration was used for the migration analysis.

BPA (Pubchem CID:6623) standard (99% purity) was purchased from Aldrich Chemical Co. (St.Louis, MO). Water, used as a chromatographic solvent, was used for the preparation of the simulant for the migration tests, and additionally for the preparation of BPA standard solutions. It was purified by using a Zener Power 1 (Human Corporation, Seoul-Korea) before the analysis.

The PC water bottle samples were contacted with natural spring water supplied in glass jars from a natural spring water company for the migration.

Methods

Identification of the Material of Water Bottle with FT-IR

The 5-gallon water bottle samples were subjected to infrared analysis to confirm that the material was polycarbonate. Fourier Transform Infrared Spectrophotometer (FT-IR) measurements were carried out by using a Perkin-Elmer Spectrum 100 FT-IR spectrometer (Bucks, UK).

Residual BPA Analysis Method

The residual BPA analysis was conducted according to the procedure performed by Kızılırmak Esmer et al. [20] to determine the total amount of BPA residue in 5-gallon PC water bottles. One gram of polymer taken from the PC water carboy sample by cutting was immersed to 20 mL of dichloromethane in a beaker and placed in an ultrasonic water bath at 20 °C until completely dissolved. The dissolved polymer was titrated with 50 mL of methanol and the precipitate was allowed to rest for 10 min. The supernatant was removed and filtered via 0.22 µm polytetrafluoroethylene (PTFE) filter and the residual BPA was determined using the High-Performance Liquid Chromatography with Fluorescence Detection (HPLC-FLD) procedure described below. All the experiments

were conducted in triplicate runs each with two repetitions.

The validation of the BPA analysis method was carried out in terms of, the limit of detection (LOD), the limit of quantitation (LOQ), the linearity and the repeatability (Rec).

BPA Migration Analysis Method

The migration analysis was performed according to the method of Kızıllırmak Esmer et al. [20], after the 5 - gallon PC water bottles were subjected to the migration test conditions given in the experimental design. The 5 - gallon PC water bottle parts were taken as each part will be as 24 cm² of surface area and each part was immersed in 100 ml of natural spring water in a glass jar in order to ensure the ratio of the surface area to the volume of the water bottle as in the real-use conditions. The surfaces of the samples were carefully cleaned with the paper towels before the analysis. At the end of the contact period, the samples were carefully removed so that the solution on their surface was completely transferred into the jar, and the simulant was filtered through the 0.22 µm PTFE filter, transferred into the vial and the BPA level was determined. Since there is two-sided contact in the immersion method, and the surface area of the sample in contact with the simulant during the migration test was increased by twofold, the migration results were divided by two. However, in real contact-conditions the migration occurs in one-way. Therefore, the calculated migration results for some contact conditions were below the LOD value of the method. All the experiments were conducted in triplicate runs each with two repetitions.

Experimental Design for Migration Analysis

Migration of BPA from the 5-gallon PC water bottles was investigated at three contact temperatures of 5±1°C (D5), 20 ±1°C (D20) and 40±1°C (D40) at dark conditions and

one contact temperature of 20±1 °C (L20) under sunlight filtered through the window for eight storage periods of 1st, 5th, 10th, 30th, 45th and 60th days. The samples under sunlight were put near the window at which room temperature was kept steadily at 20±1°C by the air conditioner. The experimental design for migration analysis was set in triplicate runs with two repetitions for each period.

Chromatographic Parameters

The quantitative determination of BPA in the samples was achieved by using Agilent 1200 Series HPLC system (Santa Clara, United States) with fluorescent detection at Ex: 285 nm and at Em:315 nm and autosampler system. A 5 µm Zorbax Eclipse XDB C18 (150mm x4.6mm i.d.) was used. Mobile phase was methanol/water (70:30, v/v) in isocratic elution at 0.4 mL/min at 25°C, and the injection volume was 10 µL for all the solutions.

Statistical Analysis

The data of residual content of BPA were statistically analyzed and presented as the mean ± standard deviation (SD). The coefficient of determination (R²) was determined by regression/ correlation analysis in the SPSS software [24]. Statistical significance was performed using one-way ANOVA and Duncan's multiple range tests (p<0.05).

RESULTS and DISCUSSION

Identification of the Material of Water Bottle with FT-IR

The FTIR spectrum of PC water carboys was shown in Figure 1. The characteristic absorption peaks of Bisphenol-A based Polycarbonate are suitable with the results of Hacıoğlu [40] and Parshin et al., [41] which shows the material is PC.

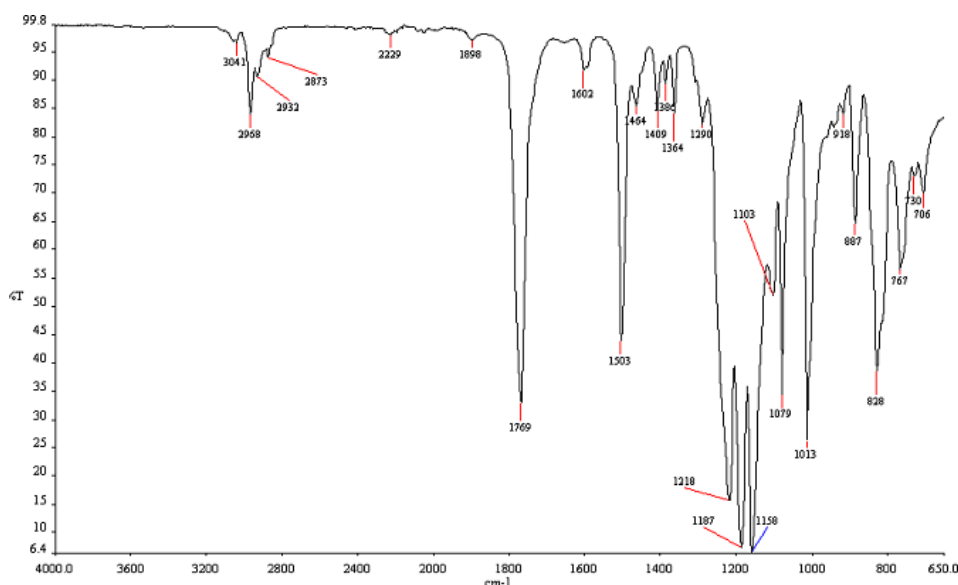


Figure 1. FTIR spectrum of a PC water carboy sample

Method Validation Results

Residual BPA Analysis

It was determined that the limit of detection (LOD), and limit of quantification (LOQ) were 6.39 ppb and 9.30 ppb respectively and the recovery was 84.2%. Calibration curves were obtained by measuring the standard solutions at five levels in the range of 200-2000 ppb with three replicates per concentration. It was found that the coefficient of determination (R^2) is greater than 0.99 in linear regression analysis performed by using the analyte peak area vs. analyte concentration.

BPA Migration Analysis

It was determined that the LOD and the LOQ were 1.14 ppb, and 1.48 ppb respectively and the recovery was 103%. The recovery analysis was carried out by contacting the PC water bottle samples with both solution A and solution B for 40°C/10 days and then the migration amounts were calculated. In this analysis, solution A was composed of natural spring water spiked with BPA standard to attain 20 ppb of BPA concentration, whereas

solution B was just natural spring water. The % recovery was calculated by using the equation of;

$$\% \text{ Recovery} = (M_A - M_B) / 20 * 100$$

M_A : BPA amount in solution A (ppb)
 M_B : BPA amount in solution B (ppb)

Calibration curves were obtained by measuring standard solutions at five levels in the range of 0-30 ppb with three replicates per concentration. It was found that the coefficient of determination (R^2) is greater than 0.99 in linear regression analysis performed by using the analyte peak area vs. analyte concentration.

Residual BPA Amount

As depicted in Table 1, it was found that the highest amount of residual BPA in the samples was 3.50 ± 0.14 ppm, the lowest amount of residual BPA was 0.71 ± 0.13 ppm. The amounts of residual BPA in total 30 water carboys samples including 5 new ones and 5 old ones from three different parts were over 2 ppm only in 8 samples and between 1-2 ppm in 18 samples, below 1 ppm in 4 samples. The amount of residual BPA was over 2 ppm in all of the samples of the trademark no 2.

Table 1. Residual BPA amounts in the PC water carboys

Sample Code	Age	Residual BPA amounts for different parts of water bottles (ppm)*			Average BPA amounts (ppm)
		Upper part	Medium part	Lower part	
1	New	1.66±0.00Aa	1.86±0.03Ba	1.45±0.09Ca	1.66±0.04a
1	Old	1.12±0.17Aa	0.95±0.008Ab	1.00±0.20Aa	1.02±0.13b
2	New	2.50±0.05Aa	3.02±0.30Aa	2.55±0.25Aa	2.69±0.20a
2	Old	3.50±0.14Ab	3.29±0.16Aa	2.23±0.31Ba	3.00±0.20a
3	New	2.03±0.06Aa	1.77±0.05Ba	1.72±0.04Ba	1.84±0.05a
3	Old	0.96±0.10Ab	1.21±0.20Aa	1.18±0.00Ab	1.12±0.10b
4	New	1.72±0.05Aa	1.37±0.11Aa	1.44±0.13Aa	1.51±0.10a
4	Old	2.57±0.14Ab	1.73±0.13Ba	1.03±0.20Ba	1.78±0.16a
5	New	1.58±0.13Aa	0.71±0.13Ba	0.99±0.04Ba	1.09±0.10a
5	Old	1.26±0.00Ba	1.77±0.14Ab	1.13±0.12Ba	1.39±0.09a

*: Means with different capital letters show significant differences in the same line ($p \leq 0.05$). Means with different lowercase letters show significant differences between new and old samples of the same coded samples ($p \leq 0.05$).

In order to determine whether several parts of PC water carboys were statistically different in terms of the amounts of residual BPA, the amounts of the residual BPA between the same samples were assessed by applying one-way analysis of variance and Duncan's test. The difference was statistically significant in 6 samples ($p \leq 0.05$) whereas the difference was not statistically important for the other 4 samples as shown in Table 1. Several studies have indicated that PC materials may be degraded especially due to the effect of different ambient conditions such as high temperature, alkaline environment, sunlight or different repeating processes such as washing with detergent solution, sterilization, and there may be an increase in the amounts of residual BPA in the material or amounts of BPA migration to the food [1; 10; 12; 20; 21; 22; 23; 25]. In our study, as shown in Table 1, the residual BPA amounts of old (life time ended) PC water carboys were higher than that of the new (not used before) samples in three of the five samples and one of these differences was statistically important ($p \leq 0.05$). For the other two samples the residual BPA amount of old

samples were lower than that of the new samples and both of these differences were significantly important ($p \leq 0.05$). Although the higher BPA residual amounts were expected in all of the old samples since exposure to the environmental and usage conditions, the residual BPA amounts of old samples may be lower than the new ones. This may be due to that, even if the material degrades depending on the conditions of use, the increase in residual BPA amount can not be detected since it may migrate to the water contacted during the next usage period since these materials are reusable. Indeed, in a study, it was determined that there was no significant difference between the brand-new PC water bottles and the PC water bottles used (1-9 year) by consumers at home under ordinary usage conditions in terms of BPA migration and they stated that consumers washed PC water bottles with hot water at 50°C at home and, therefore, this process did not affect PC material in terms of the amount of residual BPA [3]. As a result of the literature review about the residual BPA amounts of PC materials, the amounts of residual BPA in the PC

materials used for the different purposes may vary from 5-10 ppb to 599 ppm as seen in Table 2 and it was remarkable that the results were generally at ppm-level.

For this reason, it is considered that the amounts of residual BPA of the samples of PC water carboys produced in Turkey were at a low level.

Table 2. Residual BPA amounts of different PC materials in previous studies

PC material	Residual BPA amounts	Reference
Baby bottle	0.60-6.23 ppm	[20]
Plate	8-13 ppm	
Glass	7-10 ppm	
DVD	80 ppm	[12]
Baby bottle	5-40 ppm	
Bowl	15 ppm	
Baby bottle	16.2-17.6 ppb	[13]
Water galloy	2.5-70 ppm	
Bottle	2.1-25 ppm	
Film	4-5 ppm	
Plate	2.5-9.0 ppm	[27]
Bar	9.0-9.3 ppm	
Disc	10-15 ppm	
Baby bottle	1.4-35.3 ppm	[36]
Baby bottle	6-25 ppb	[22]
Baby bottle	<1 ppm-500 ppm	[28]
Baby bottle	4.01-141 ppm	[37]
Food stock box from PC waste	347 ppm	[38]
Ricebowl	379 ppm	
Mug	599 ppm	
Soup cup	596	[8]
Dish	431	
Baby bottle	<1 ppm-20 ppm	
Baby bottle	7-58 ppm	[11]
Baby bottle	4-139 ppm	[39]

BPA Migration Analysis Results

Typical BPA chromatograms are shown in Figure 2. BPA migration results for different contact conditions are demonstrated in Figure 3. Also, Table 3 shows the statistical comparison of the results. According to these results, the BPA migration increased with the contact period and the contact temperature and these increases were statistically important ($p < 0.05$). Based on the migration theory, the concentration of the migrating compound is directly proportional to the square root of the contact time [18; 19; 26]. As the contact period increases the migration amount of a substance will increase, too. Moreover the migration of substances is accelerated by heat. So the migration will occur faster if the temperature is raised. Our results were in accordance with this correlation. As depicted in Figure 4, it was observed that the increase of BPA migration with square root of time was exponential based on D20, D40, and L20 contact conditions. The findings of this research are in agreement with the literature. In a study conducted to determine the BPA migration from PC materials contact with water at 40°C, 60°C and 80°C in different periods up to 360 hours, it was determined that with increasing the contact temperature, the migrated amount of BPA and the rate of the migration increased with contact period [27].

When we examined the BPA migration values, there was no BPA migration at a detectable level at 5°C of contact temperature during the entire storage period. At 40°C of contact conditions, the maximum BPA migration was 14.91±0.58 ppb at the end of the storage period of 60 days. To determine the effect of sunlight, the samples

were stored under dark and sunlight conditions at 20°C of contact conditions. The BPA migration statistically changed according to the availability of sunlight ($p \leq 0.05$). It increased the BPA migration to exposure to sunlight and at the end of storage period of 60 days at 20°C under sunlight, the maximum BPA migration was 12.91±0.55 ppb whereas the maximum BPA migration was 9.93±0.65 ppb for the same contact conditions in dark. And these differences were statistically important ($p \leq 0.05$). As stated by some researchers, this situation might have occurred due to the fact that polymeric material may be degraded when PC materials were exposed to the external environmental conditions such as sunlight [12; 28-30]. As a result of the degradation of the material, there was an increase in the amount of the residual BPA, and this situation may lead to a rise in the amount of the BPA migration level. In a study, the researchers analyzed the BPA migration from new PC water bottles to water at room temperature (22°C) under the 1, 3, 5 and 7 days contact conditions. They found that the BPA migration values were 0.08-0.36 ppb at the end of the 1st day and 0.73-1.33 ppb at the end of the 7th day [3]. Similar results were obtained in the present study as a result of the storage at room temperature. Indeed, in the study conducted by [31], to determine the migration of BPA under severe conditions of contact temperature of 70°C, they found that the amount of BPA migration was in the range of 32-54.7 ppb at the end of the first day in PC baby bottle of three trademarks and the reusable PC water bottle of two trademarks in contact with water at 70°C and the amount of BPA migration increased in time and it reached to 228-528 ppb range on the 6th day.

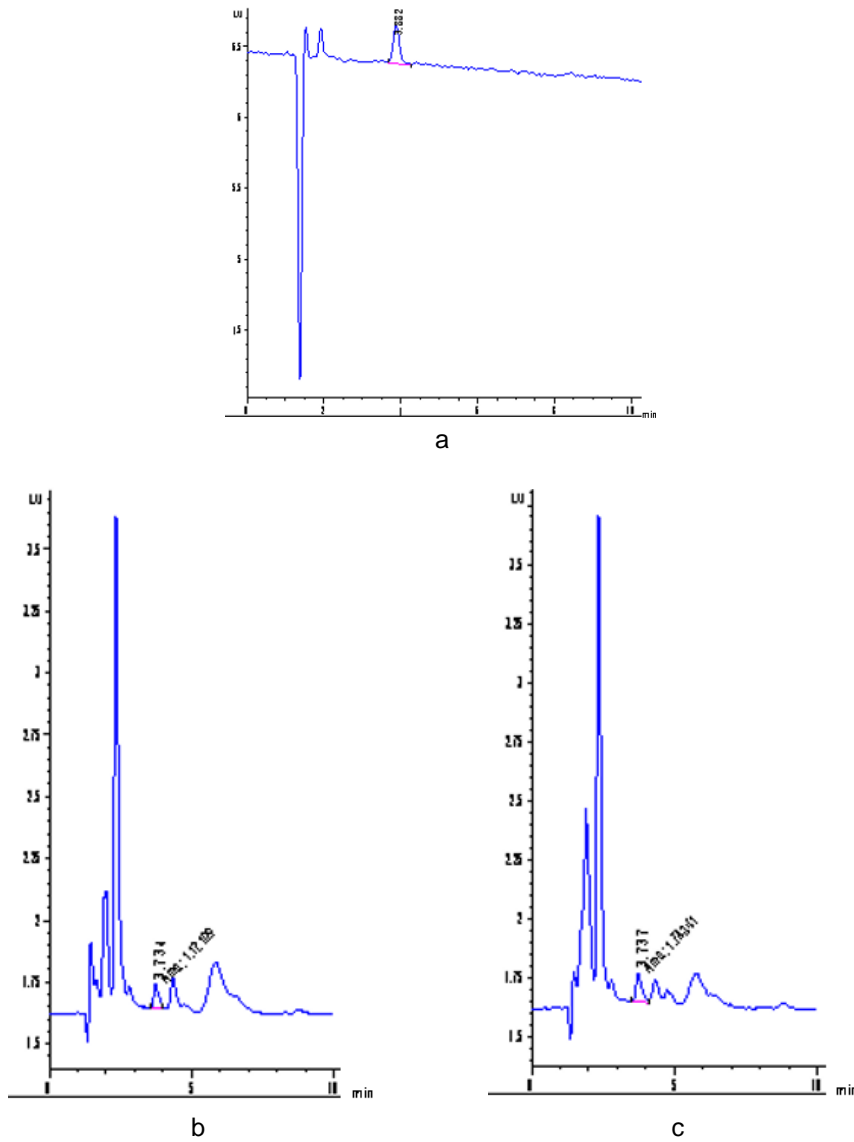


Figure 2. BPA chromatogram a) of 10 ppb of BPA standard b) at contact conditions of 20°C/45 days c) of the sample spiked with 20 ppb BPA

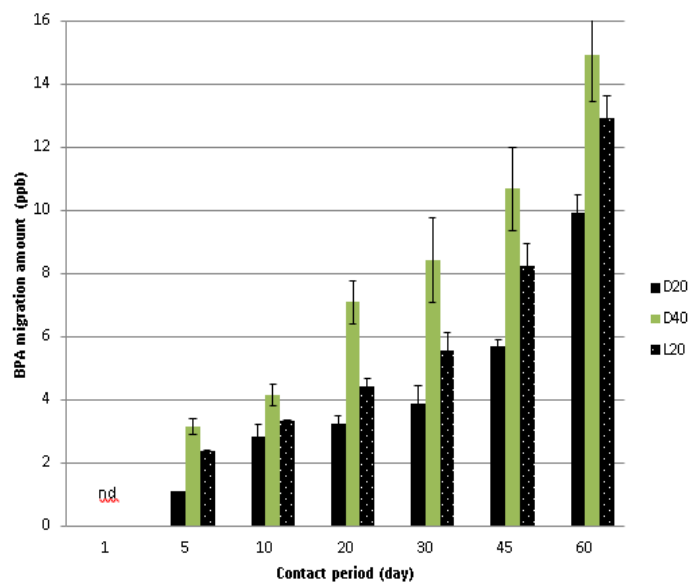


Figure 3. BPA migration amounts at different contact temperature and contact periods.

Table 3. BPA migration amounts from water carboy samples into the water under different contact conditions

Contact period	Amount of BPA migration (ppb)			
	D5	D20	D40	L20
1 st day	Nd	Nd	Nd	Nd
5 th day	Nd	1.09±0.54Aa	3.14±0.47Ac	2.38±0.38Ab
10 th day	Nd	2.80±0.34Ba	4.14±0.53Bb	3.33±0.56Ba
20 th day	Nd	3.22±0.56Ba	7.08±0.48Cb	4.40±0.79BCa
30 th day	Nd	3.86±0.74Ba	8.43±0.38Cc	5.57±0.63Cb
45 th day	Nd	5.67±0.92 Ca	10.18±0.74Dc	8.22±0.32Db
60 th day	Nd	9.93±0.65 Da	14.91±0.58Eb	12.91±0.55Eb

*: Means with different capital letters show significant differences in the same column (p≤0.05). Means with different lowercase letters show significant differences in the same line (p≤0.05).

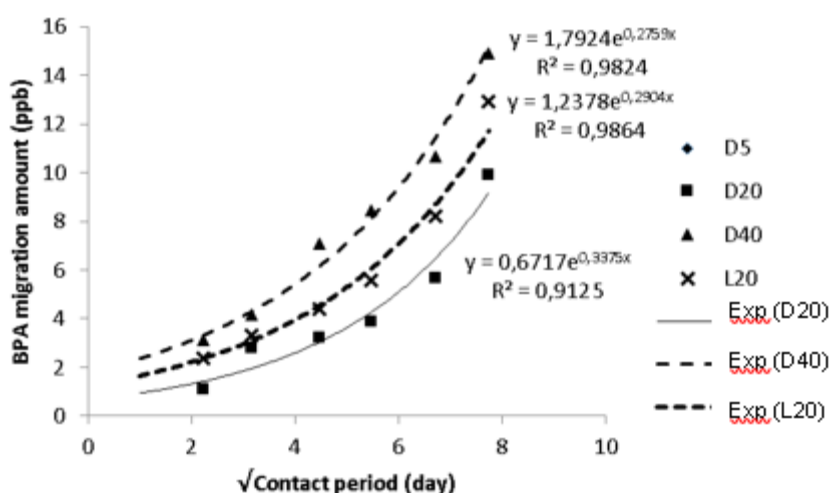


Figure 4. The change in migration of BPA with square root of time at different temperatures

In general, it was remarkable that the determined BPA migration amounts were rather lower compared to the residual amounts of BPA. This was considered to be due to the fact that the solubility of BPA in water is low [32-34]. In many situations, the "migration from polymers" term is defined as a mass transfer related to the diffusion managed by the predictable physical processes included in a polymer network [35]. Although it has been stated that the BPA migration from PC materials to aqueous foods is also caused by the BPA released as a result of the hydrolysis occurring at the surface of the material as well as the residual BPA remained in the material after production, it is known that PC materials are more susceptible to hydrolysis in contact with water especially at the temperatures over 60°C [10, 12-16]. The carbonate linkages in the PC material are subjected to hydrolytic attack at high temperatures [13]. As the hydrolysis does not occur when the water in the PC water bottle is not stored at high temperatures such as 60°C, it is considered that the amounts of the BPA migration were at low concentrations.

Assessment of Amounts of BPA Migration Based on TDI Value and Specific Migration Amount

The tolerable daily intake (TDI) value for BPA has been changed by EFSA within years and the value has been decreased. The last TDI value is 4 µg/kg body weight/day [6]. The amount of BPA daily tolerable for a human being

with an average body weight of 60 kg is 240 µg. In this case, in order to reach this amount based on the amounts of BPA migration determined according to different environmental conditions:

The human body will be able to tolerate the amount of BPA taken;

- When 203 L of water is consumed daily from a 5-gallon PC water bottle stored at the D5 storage conditions for 60 days. (1.14 ppb of LOD was taken as the maximum BPA migration amount)
- When 24 L of water is consumed daily from a 5-gallon PC water bottle stored at the D20 storage conditions for 60 days.
- When 16 L of water is consumed daily from a 5-gallon PC water bottle stored at the D40 storage conditions for 60 days.
- When 19 L of water is consumed daily from a 5-gallon PC water bottle stored at the L20 storage conditions for 60 days.

The specific migration limit (SML) for BPA was 0.6 mg/kg food in 2002, this value was decreased to 0.05 mg/kg food (50 µg/kg food=50 ppb) in 2018 according to the Commission Regulation No 2018/213 [42]. In other words, the amount of BPA to migrate from the 5-gallon PC water bottles to the water included in them is 50 µg for 1 L of water. When we compared this value with our

results even for 60-days storage we concluded that; the migration amounts are quite lower than the SML value.

CONCLUSION

It was determined that the 5-gallon PC water bottles produced in Turkey had a standard quality in terms of the amount of residual BPA and these amounts were lower compared to the residual BPA values in the literature. Although it has been stated in the current literature that there may be an increase in the amount of residual BPA as a result of the degradation of the PC material due to the processes applied during the usage periods and the conditions it was exposed to, the increase in the residual BPA amounts may not be noticeable by residue analysis, as the material is a reusable and may migrate into the water contacted during each usage period. Therefore, lower or higher residue amounts can be detected in old materials compared to new materials.

When we evaluate the BPA migration amounts on the basis of TDI value, the amount of water should be drunk is 16 L at the worst conditions of 40°C/60 days. It is not practicable to drink such amounts of water. Overall it is possible to conclude that PC water carboys are safe for the BPA migration.

In our daily life we should not use the materials having any migration risk of especially toxicologically important substances at high temperature and long contact times.

ACKNOWLEDGEMENTS

The authors would like to thank the Ege University Scientific Research Projects Commission (2015-MUH-019) for financial support.

REFERENCES

- [1] Maia, J., Cruz, J.M., Sendón, R., Bustos, J., Sanchez, J.J., Paseiro, P. (2009). Effect of detergents in the release of bisphenol A from polycarbonate baby bottles. *Food Research International*, 42(10), 1410–1414.
- [2] Kızılırmak Esmer, Ö., Üçüncü, M., Saygılıer, E. (2010), Ambalajlardan gıdalara geçen bisfenol-A'ya ilişkin risk değerlendirmeleri, VI. *International Packaging Congress*, İstanbul.
- [3] Le, H.H., Carlson, E.M., Chua, J.P., Belcher, S.M. (2008). Bisphenol A is released from polycarbonate drinking bottles and mimics the neurotoxic actions of estrogen in developing cerebellar neurons. *Toxicology Letters*, 176(2), 149–156.
- [4] Itoh, H., Iwasaki, M., Hanaoka, T., Sasaki, H., Tanaka, T., Tsugane, S. (2007). Urinary Bisphenol-A Concentration in Infertile Japanese Women and Its Association with Endometriosis: A Cross-Sectional Study. *Environmental Health and Preventive Medicine*, 12(6), 258–264.
- [5] Kang, J.-H., Katayama, Y., Kondo, F. (2006). Biodegradation or metabolism of bisphenol A: from microorganisms to mammals. *Toxicology*, 217(2), 81–90.
- [6] EFSA (European Food Safety Authority). (2015). Scientific Opinion on the risks to public health related to the presence of bisphenol A (BPA) in foodstuffs: Executive summary. EFSA panel on food contact materials, enzymes, flavourings and processing aids.
- [7] Commission Regulation. No 10/2011. (2011). Plastic materials and articles intended to come into contact with food. Official Journal of the European Communities, L 12/1.
- [8] Kawamura, Y., Koyano, Y., Takeda, Y., Yamada, T. (1998). Migration of Bisphenol A from Polycarbonate Products. *Food Hygiene and Safety Science (Shokuhin Eiseigaku Zasshi)*, 39(3), 206-212_1.
- [9] Maragou, N.C., Makri, A., Lampi, E.N., Thomaidis, N.S., Koupparis, M.A. (2008). Migration of bisphenol A from polycarbonate baby bottles under real use conditions. *Food Additives & Contaminants: Part A*, 25(3), 373–383.
- [10] Brede, C., Fjeldal, P., Skjevraak, I., Herikstad, H. (2003). Increased migration levels of bisphenol A from polycarbonate baby bottles after dishwashing, boiling and brushing. *Food Additives & Contaminants*, 20(7), 684–689.
- [11] Biles, J.E., McNeal, T.P., Begley, T.H., Hollifield, H.C. (1997). Determination of Bisphenol-A in Reusable Polycarbonate Food-Contact Plastics and Migration to Food-Simulating Liquids. *Journal of Agricultural and Food Chemistry*, 45(9), 3541–3544.
- [12] Pedersen, D.A., Hvilsted, S., Petersen, J.H. (2015). Migration of bisphenol-A from polycarbonate plastic of different qualities, Environmental project No. 1710. http://orbit.dtu.dk/files/110762088/BPA_MST_project_No_1710_2015.pdf
- [13] Nam, S.-H., Seo, Y.-M., Kim, M.-G. (2010). Bisphenol A migration from polycarbonate baby bottle with repeated use. *Chemosphere*, 79(9), 949–952.
- [14] De Coensel, N., David, F., Sandra, P. (2009). Study on the migration of bisphenol-A from baby bottles by stir bar sorptive extraction-thermal desorption-capillary GC-MS. *Journal of Separation Science*, 32(21), 3829–3836.
- [15] Kubwabo, C., Kosarac, I., Stewart, B., Gauthier, B.R., Lalonde, K., Lalonde, P.J. (2009). Migration of bisphenol A from plastic baby bottles, baby bottle liners and reusable polycarbonate drinking bottles. *Food Additives and Contaminants - Part A Chemistry, Analysis, Control, Exposure and Risk Assessment*, 26(6), 928–937.
- [16] Hu, L.-C., Oku, A., Yamada, E. (1998). Alkali-catalyzed methanolysis of polycarbonate. A study on recycling of bisphenol A and dimethyl carbonate. *Polymer*, 39(16), 3841–3845.

- [17] Anonymous, "Baby's toxic bottle- Bisphenol-A leaching from popular baby bottles, A report prepared by Work Group for Safe Markets," 2020 May 2008. [Online]. Available: <https://www.cleanwateraction.org/files/publications/national/babystoxicbottle-final.pdf> /.
- [18] Castle, L. (2007). Chemical migration into food: an overview. In K.A. Barnes, C.R. Sinclair & D.H. Watson (Eds.) *Chemical migration and food contact* (p. 1-12). Woodhead Publishing Limited, Abington Hall, Abington Cambridge CB21 6AH, England . ISBN-13: 978-1-84569-029-8
- [19] Vom Bruck, C.G., Bieber, W.D., Figge, K. (1986). Interaction between food and packaging materials and its consequence on migration. *Food Packaging and Preservation. Theory and Practice* (ed. M. Mathlouti), Elsevier Applied Science, London, 39-66.
- [20] Kızılırmak Esmer, Ö, Çağındı, Ö., Şahin, B. (2017). Does the realistic contact and daily use conditions limit the use of polycarbonate baby bottles for migration and residue level of Bisphenol-A. *Journal of Food and Health Science*, 3(4), 150-160.
- [21] Biedermann-Brem, S., Grob, K. (2009). Release of bisphenol A from polycarbonate baby bottles: Water hardness as the most relevant factor. *European Food Research and Technology*, 228(5), 679–684.
- [22] Biedermann-Brem, S., Grob, K., Fjeldal, P. (2008). Release of bisphenol A from polycarbonate baby bottles: Mechanisms of formation and investigation of worst case scenarios. *European Food Research and Technology*, 227(4), 1053–1060.
- [23] Howdeshell, K.L., Peterman, P.H., Judy, B.M., Taylor, J.A., Orazio, C.E., Ruhlen, R.L., vom Saal, S.F., Welshons, W.V. (2003). Bisphenol A is released from used polycarbonate animal cages into water at room temperature. *Environmental Health Perspectives*, 111(9), 1180–1187.
- [24] IBM Corp. (2013). IBM SPSS Statistics for windows, Version 22.0. Armonk, NY: IBM Corp.
- [25] Takao, Y., Lee, H.C., Ishibashi, Y., Kohra, S., Tominaga, N., Arizono, K. (1999). Fast screening method for Bisphenol A in environmental water and in food by solid-phase microextraction (SPME). *Journal Of Health Science*, 45(1), P39-39.
- [26] Arvanitoyannis, L., Bosnea, L. (2004) Migration of substances from food packaging materials to foods. *Critical Reviews in Food Science and Nutrition*, 44 (2), 63-76. |
- [27] Mercea, P. (2009). Physicochemical processes involved in migration of bisphenol A from polycarbonate. *Journal of Applied Polymer Science*, 112(2), 579–593.
- [28] Diepens, M., Gijsman, P. (2010). Photodegradation of bisphenol A polycarbonate with different types of stabilizers. *Polymer Degradation and Stability*, 95(5), 811–817.
- [29] Diepens, M., Gijsman, P. (2009). Photostabilizing of bisphenol A polycarbonate by using UV-absorbers and self protective block copolymers based on resorcinol polyarylate blocks. *Polymer Degradation and Stability*, 94(10), 1808–1813. 008
- [30] Diepens, M., Gijsman, P. (2007). Photodegradation of bisphenol A polycarbonate. *Polymer Degradation and Stability*, 92(3), 397–406.
- [31] Cao, X.-L., Corriveau, J. (2008). Migration of Bisphenol A from Polycarbonate Baby and Water Bottles into Water under Severe Conditions. *Journal of Agricultural and Food Chemistry*, 56(15), 6378–6381
- [32] Corrales, J., Kristofco, L.A., Baylor Steele, W., Yates, B.S., Breed, C.S., Spencer Williams, E., & Brooks, B.W. (2015). Global assessment of bisphenol a in the environment: *Review and analysis of its occurrence and bioaccumulation. Dose-Response*, 13(3), 1–29
- [33] Özdal, T., Yeşilcubuk, N.Ş. (2014). Toxicity of Bisphenol-A: Effects on Health and Regulations. *International Journal of Agricultural and Biosystems Engineering*, 8(6), 553–557.
- [34] Bhunia, K., Sablani, S.S., Tang, J., Rasco, B. (2013). Migration of chemical compounds from packaging polymers during microwave, conventional heat treatment, and storage. *Comprehensive Reviews in Food Science and Food Safety*, 12(5), 523–545.
- [35] Piringer, O.G., Baner, A.L. (2008). *Plastic packaging*, (2nd ed.). Weinheim,: Wiley-VCH Verlag GmbH & Co. KGaA.
- [36] Ehlert, K.A., Beumer, C.W.E., Groot, M.C.E. (2008). Migration of bisphenol-A into water from polycarbonate baby bottles during microwave heating. *Food Additives and Contaminants, Part A*, 25 (7), 904-910.
- [37] Wong, K.O., Leo, L.W. & Seah, H.L. (2005). Dietary exposure assessment of infants to bisphenol A from the use of polycarbonate baby milk bottles. *Food Additives and Contaminants Part A*, 22(3), 280-288.
- [38] Yamamoto, T., Yasuhara, A. (1999). Chlorination of bisphenol A in aqueous media: formation of chlorinated bisphenol A congeners and degradation to chlorinated phenolic compounds. *Chemosphere* 46, 1215–1223.
- [39] Mountfort, K.A., Kelly, J., Jickells, S.M., Castle, L. (1997). Investigations in to the potential degradation of polycarbonate baby bottles during sterilization with consequent release of Bisphenol A. *Food Additives and Contaminants*, 14 (6-7), 737-740.
- [40] Hacıoğlu, F. (2017). Degradation of polycarbonate, bentonite, barite, carbon fiber and glass fiber filled polycarbonate via gamma irradiation and possible use of polycarbonate in radioactive waste management. PhD Thesis of the Graduate School of Natural and Applied Sciences of Middle East Technical University, Ankara.

[41] Parshin, A.M., Gunyakov, V.A., Zyryanov, V.Y., Shabanov, V.F. (2013). Domain structures in nematic liquid crystals on a polycarbonate surface. *International Journal of Molecular Sciences*,14, 16303-16320.

[42] Commission Regulation. (2018). The use of bisphenol A in varnishes and coatings intended to

come into contact with food and amending Regulation (EU) No 10/2011 as regards the use of that substance in plastic food contact materials Official Journal of the European Communities, L 41, 6-12.