Effect of NaCl on antimony and phthalate compounds leached from PET bottles

Piyawan Leechart, Duangrat Inthorn and Paitip Thiravetyan

ABSTRACT

Nowadays polyethylene terephthalate (PET) bottles are commonly used as food containers as they are lightweight. PET bottles contain antimony (Sb) and phthalate compounds. In contact with food, antimony and phthalate molecules could migrate from the inner surface of a PET bottle to the food. Therefore, we studied the effect of NaCl concentration in PET bottles on the leakage of antimony and phthalates. It was found that the concentration of antimony leached into the solution was about 6 ng l⁻¹ after 5 days storage at room temperature in the absence of NaCl. Increasing NaCl concentrations to 6% caused a decrease in the amount of soluble antimony in the solution to 2 ng l⁻¹ under the same conditions. In addition, the maximum leakage of phthalate compounds of about 130 ng l⁻¹ occurred after 35 days of storage at 60°C in 0.1% NaCl. It was found that the leakage of phthalate compounds decreased at higher NaCl concentrations (NaCl 0.5%–30%). Higher NaCl concentrations led to a decrease in the migration of antimony and phthalate compounds into the solution. This might be due to the fact that antimony and phthalate compounds form complexes with NaCl. However, the leakage of these compounds was lower than the standard guidelines of the United States Environmental Protection Agency for drinking water.

Key words | antimony, di-2-ethylhexyl phthalate, diethyl phthalate, dimethyl phthalate, NaCl, polyethylene terephthalate

INTRODUCTION

Plastic bottles are commonly used in food manufacturing processes and have replaced glass bottles in many applications, including storing condiments. This is due to their light weight and low costs of production. Plastic bottles are produced using a variety of techniques and materials that depend on the application. Plastic food containers were found to be a source of contamination by the migration of substances, such as additives and residual monomers, which are used as raw materials in plastic production and in the production of decomposition products. Plastic food packages may contain low concentrations of additives such as dyes, pigments, antioxidants, antifogging agents, plasticizers and stabilizers that may migrate into the packaged food (Arvanitoyannis & Bosnea 2004; Lu et al. 2009). Phthalate compounds are widely used as additives in plastic products. These are compounds such as di-2-ethylhexyl phthalate (DEHP) which is primarily used as a plasticizer, and diethyl phthalate (DEP) which is widely used as a solvent to maintain color and scent. Dimethyl phthalate (DMP) is a synthetic chemical. It is mostly used as a solvent and plasticizer in cellulose acetate and cellulose acetate-butyrate manufacturing (Cao 2010). In studies of rodents exposed to certain phthalates, high doses have been shown to change hormone levels and cause birth defects. Phthalate compounds can have adverse endocrine disruptive effects on humans. The United States Environmental Protection Agency (USEPA 2007) has established the maximum contaminant level (MCL) of 6 μg l⁻¹ for DEHP in drinking water and daily levels of exposure to DEHP for the general RfD of 0.02 mg kg⁻¹ day⁻¹ basing on difficulties in reproduction, liver problems, and increased risk of cancer in adult animals. The MCL of DMP for an oral acute exposure is 7 mg kg⁻¹ day⁻¹ based on reproductive effects. A
study on plastic food containers from Brazil found dibutyl phthalate content in the range of <LOQ to 7.5 μg l⁻¹ (Moreira et al. 2013).

Polyethylene terephthalate (PET) resin typically contains between 100 and 300 mg Sb kg⁻¹ plastic (Duh 2002). Antimony is regulated as a drinking water contaminant because it can cause negative health effects, such as nausea, vomiting, and diarrhea, when exposure exceeds the MCL of 6 μg l⁻¹ in relatively short periods. Long-term exposure can lead to increased blood cholesterol and decreased blood sugar (Gebel 1997). In the United States, the Environmental Protection Agency (USEPA) has established the MCL of 6 μg l⁻¹ for antimony, which is the same as the limit set by Health Canada. The German Federal Ministry of Environment has set the limit of 5 μg l⁻¹, while the Japanese drinking water standard requires the level of antimony to be below 2 μg l⁻¹ (Shotyk & Krachler 2007). Previous studies reported that the leaching of antimony in PET plastics, used for storing bottled water in Europe and Canada, was higher than the limit for 16 out of 55 brands of bottled water (Naohara 1998; Shotyk et al. 2006). Nishioka et al. (2002) investigated antimony concentrations in PET bottles manufactured in Japan. They found a bimodal distribution of concentrations, with some bottles having antimony concentrations between 170 and 220 mg kg⁻¹ plastic, while in other bottles, antimony concentrations were below the limit of detection (<0.1 mg kg⁻¹). In another study from Japan, PET bottles from 80 different brands were collected and the predicted model for Sb migration suggested that storage conditions of all PET drink products should be below 70 °C for a maximum of 72 days to avoid the Sb levels exceeding the recommended value of 5 μg l⁻¹, which was based on the European standards (Rungchang et al. 2013). Sb concentration increased marginally with the storage time but still remained below the various guidelines for Sb levels in drinking water (Brandao et al. 2014). The influence of storage time and temperature of PET bottles on the mineral water was studied and the migration studies showed that water stored at 4 and 20 °C was not subject to Sb migration (Carneado et al. 2015). At 40 °C there was a significant increase in Sb concentration. However, it still did not exceed the limit established by the European Union (5 μg l⁻¹), whereas at 60 °C samples were subject to considerable Sb migration after 30 days of storage. It exceeded the maximum limit established by the European Union (Carneado et al. 2013). Sánchez-Martínez et al. (2013) studied antimony migration into food simulants and vinegars from PET bottles produced in Spain. The result showed that the values of antimony that leached from reused bottles ranged from 0.5 to 1.2 μg l⁻¹. It was less than the USEPA guideline for drinking water, according to which it should have been less than 6 μg l⁻¹. Several investigators have demonstrated significant levels of antimony in bottled water in PET containers.

The packaging itself can therefore represent a source of contamination through the migration of substances from the packaging material into the food. The migration of additives or contaminants from polymers in the food containers to the food may be divided into three different stages, as follows. (1) Diffusion within the polymer: the diffusion of the migrant that is the Brownian motion of individual migrant molecules in the polymer lattice. The molecular transport of migrants is described by Fick’s laws of diffusion and the rate of migration varies with the thickness of the polymer container. (2) Solvation on the polymer–food surface: the migrant partitions are more soluble in food than in the polymer, which facilitates the rate of migration to food. On the other hand, if the migrant partitions are poorly soluble in food, retardation of the migration rate into the food might occur. (3) Dispersion into bulk food: the solvated migrant molecules diffuse away from the interface and move into bulk food. The solubility and diffusion of migrants govern the dispersion of migrants into the food, and the coefficients are the prime factors affecting the rate of migration as a whole (Lau & Wong 2000). Factors such as humidity, temperature, sunlight, trace metal ions, moisture and acid/alkaline impurities can play a significant role in the degradation process of PET in the environment. These factors affect hydrolytic degradation, photodegradation and thermal degradation.

PET plastic bottles are not only used for drinking water, but also as food containers. People in Southeast Asia and the coastal regions of East Asia use condiments during cooking. They are usually used in moderation because of their intense flavors. These condiments include fish sauce, soy sauce, oyster sauce, chili sauce and ketchup. They are kept in glass and plastic bottles depending on the quantity and utilization by customers. Condiments contain salt. Fish sauce has a particularly high salt content (between 25%
and 28%) (Tungkawachara 2003). Thus, the objective of this paper was to investigate the effect of NaCl concentration in food containers (condiment bottles). The effects include for example chloride conditions in food products to examine the leaching of antimony and phthalate compounds from PET plastic bottles.

MATERIALS AND METHODS

Collection of plastic bottles

Condiment bottles containing fish sauce, distilled vinegar, tomato ketchup and chili sauce were used as the materials for antimony detection. They were collected from the supermarkets and convenience stores in Thailand. Twenty-four brands of commercial condiments were purchased with consideration of the type of plastic and different types of condiments. Condiments were purchased, considering their shelf life, that were more than 6 months before the expiration date of the food packaging. The test bottles were washed many times with clean water and dried at room temperature. Antimony in the test bottles was digested by microwave digestion (Anton Paar Multiwave 3000 in the Rotor 16MF100). We collected six samples for each brand and triplicate samples were analyzed for each sample. Condiment bottles were cut into small pieces (∼5 × 5 mm², film thickness 0.33–1.0 mm) and weighed 0.25 g. The samples were mixed with 10 ml of HNO₃ and 2 ml of HCl and subjected to microwave digestion. The sample solutions were diluted to 50 ml with deionized water and were added to the solution (concentration: 1.0 μg l⁻¹) as an internal standard for the measurement by inductively coupled plasma mass spectrometry (ICP-MS, Agilent 7500a series). The limits of detection and method reporting levels were 0.00196 μg l⁻¹ and 0.012 μg l⁻¹, respectively.

Experiment conditions

The bottle that had the highest antimony concentration leaching from the bottle was used as the test material in this study. The conditional treatment was focused on the NaCl solution test. Plastic bottle samples were added to 300 ml of distilled water (control sample), and to a prepared NaCl solution (treated sample). The test of the compounds and intermediate compounds occurring in NaCl solution was prepared by varying the concentration of NaCl from 0.1% to 50.0%. Plastic container samples were kept at 60 °C for 7, 14, 21, 28 and 35 days. Five millilitres of a sample for the measurement was collected and acidified with 5% HNO₃ before the ICP-MS analysis. One-millilitre volumes of samples were collected using a syringe and filtered with a 0.2 μm syringe filter (diameter 13 mm, pore size 0.2 μm). Antimony concentrations were determined by using ICP-MS.

The high-performance liquid chromatography-UV (HPLC-UV) system has an LC-10AD pump and a SPD10-Avp UV detector and the results were analyzed with CLASS-VP software (Shimadzu, Kyoto, Japan). The separations were performed in an ODS-3-C18 column (250 × 4.6 mm i.d., particle size 5 μm, Inertsil, Japan). The mixture of the mobile phase used the ratio 50:50 of acetonitrile and potassium phosphate buffer (pH 2.5) (v:v) for DMP and DEP detection. The DEHP detection used the ratio 70:30 of acetonitrile and deionized water (v:v) as a mobile phase. The 226 nm wavelength of the UV detector was selected for all phthalate compounds. The injector volume was 40 μl and the peak’s height was used as the analytical measurement. All of the reagents used in the phthalate compounds experiment were of HPLC grade. Standard solutions of phthalate compounds were prepared by dissolving in ethanol and were used within 24 h. Three hundred millilitres of a sample solution was extracted with 100 ml of hexane. After phase separation, the extracted solution was collected and concentrated to 10 ml by evaporating under a hood instrument (at room temperature) and kept in a glass tube. One-millilitre volumes of samples were collected by using a syringe and filtered with a 0.2 μm syringe filter (diameter 13 mm, pore size 0.2 μm). Phthalate compounds were measured using HPLC. The changes in pH were measured by a pH meter (Mettler Delta 340).

RESULTS AND DISCUSSION

Antimony concentrations extracted from plastic bottles

Twenty-four bottles of condiments from commercial brands were collected from supermarkets and convenience stores.
Twenty-three brands were labeled as type 1 plastic (PET), and one brand was labeled as type 7 plastic (OTHER). The symbols that appeared on all bottles are also known as the resin identification codes. These symbols indicate the material from which the bottle is made. Type 1 plastic (PET) is the esterification reaction of a copolymer of ethylene glycol with terephthalic acid or the transesterification reaction of a copolymer of ethylene glycol with dimethyl terephthalate. Antimony trioxide (Sb$_2$O$_3$) has been widely used as a clarificant, pigment, flame retardant and catalyst. In the polyester process of PET production, antimony trioxide is used as a homogeneous catalyst in the polymerization of ethylene glycol and terephthalic acid. Type 7 plastic (OTHER) is made from another type of resin than the following six types of resin: PET, high density polyethylene, polyvinyl chloride, low density polyethylene, polypropylene and polystyrene. It can also be made from more than one resin and used as a multi-layer combination. The properties of plastic depend on the resin or the combination of resins (American Chemistry Council 2007).

Various condiment bottles were digested by microwave digestion, and antimony concentrations from digested solutions were analyzed by an ICP-MS. The antimony concentration from 24 brands of plastic bottles ranged from 0.00986 ± 0.00 to 63.10 ± 0.46 mgSb kg$^{-1}$ plastic (Table 1). The result revealed that the PET bottles (type 1 plastic) had higher antimony concentration than type 7 plastic bottles. It was found that antimony was used as a catalyst in the production process of the PET bottles. Therefore, condiment bottles made from type 1 plastic were used in a further study on the effect of NaCl concentration on antimony and phthalate compounds leaching from PET containers.

### Effect of NaCl solutions on antimony and DMP leached from plastic bottles

NaCl solutions at various concentrations (from 0.1% to 6.0%) were put into condiment bottle samples and kept at room temperature (30 °C) for 5 days. It was found that increasing NaCl concentration caused a decrease in the concentration of DMP and antimony in the solution (Figure 1). Hypochlorite radicals (OCl$^-$) have an effect on the oxidative degradation of polymer surfaces which led to phthalate compounds leached from the plastic bottles surface into the solution. Moreover, the excess of OCl$^-$ could be reacted with phthalate compounds to form other compounds. It was proposed that the presence of NaCl could generate OCl$^-$, which are reactive Cl compounds. The mechanism of OCl$^-$ generation is not known (Sajiki & Yonekubo 2002; Duvall & Edwards 2009). The concentration of antimony that leached into the condiment bottles decreased when NaCl concentration increased. According to various studies, antimony can be transported in the solution through a variety of aqueous complexes. For example, in chloride-rich solutions, antimony speciations can form complexes with chloride (SbCl$_2$$^+$, SbCl$_3$$^+$, SbCl$_4$$^+$ and SbCl$_4$) (Pantani & Desideri 1959; Ovchinnikov et al. 1982; Wood et al. 1987). Chloride enrichment has been shown to significantly reduce the concentration of dissolved antimony that migrates from the surface of PET polymer.

<table>
<thead>
<tr>
<th>Sample brand ID</th>
<th>Sb concentration (mgSb kg$^{-1}$ plastic)</th>
<th>Plastic type</th>
</tr>
</thead>
<tbody>
<tr>
<td>01</td>
<td>51.67 ± 0.12</td>
<td>1</td>
</tr>
<tr>
<td>02</td>
<td>48.68 ± 0.39</td>
<td>1</td>
</tr>
<tr>
<td>03</td>
<td>49.47 ± 0.30</td>
<td>1</td>
</tr>
<tr>
<td>04</td>
<td>56.73 ± 0.31</td>
<td>1</td>
</tr>
<tr>
<td>05</td>
<td>49.67 ± 0.49</td>
<td>1</td>
</tr>
<tr>
<td>06</td>
<td>56.60 ± 0.40</td>
<td>1</td>
</tr>
<tr>
<td>07</td>
<td>56.46 ± 0.31</td>
<td>1</td>
</tr>
<tr>
<td>08</td>
<td>55.07 ± 0.50</td>
<td>1</td>
</tr>
<tr>
<td>09</td>
<td>56.43 ± 0.52</td>
<td>1</td>
</tr>
<tr>
<td>10</td>
<td>63.10 ± 0.46</td>
<td>1</td>
</tr>
<tr>
<td>11</td>
<td>31.43 ± 0.40</td>
<td>1</td>
</tr>
<tr>
<td>12</td>
<td>56.40 ± 0.40</td>
<td>1</td>
</tr>
<tr>
<td>13</td>
<td>58.56 ± 0.32</td>
<td>1</td>
</tr>
<tr>
<td>14</td>
<td>57.27 ± 0.29</td>
<td>1</td>
</tr>
<tr>
<td>15</td>
<td>55.14 ± 0.31</td>
<td>1</td>
</tr>
<tr>
<td>16</td>
<td>37.78 ± 0.33</td>
<td>1</td>
</tr>
<tr>
<td>17</td>
<td>0.00986 ± 0.00</td>
<td>7</td>
</tr>
<tr>
<td>18</td>
<td>58.53 ± 0.08</td>
<td>1</td>
</tr>
<tr>
<td>19</td>
<td>43.14 ± 0.64</td>
<td>1</td>
</tr>
<tr>
<td>20</td>
<td>53.46 ± 0.40</td>
<td>1</td>
</tr>
<tr>
<td>21</td>
<td>47.10 ± 0.26</td>
<td>1</td>
</tr>
<tr>
<td>22</td>
<td>44.45 ± 0.47</td>
<td>1</td>
</tr>
<tr>
<td>23</td>
<td>51.47 ± 0.24</td>
<td>1</td>
</tr>
<tr>
<td>24</td>
<td>37.00 ± 0.36</td>
<td>1</td>
</tr>
</tbody>
</table>
PET bottle polymers could have occurred by a diffusion within the polymer, solvation at the polymer–food interface and dispersion into the bulk food. The packaging itself could represent the source of contamination through the migration of substances from the packaging material to the food.

The migrated antimony in different NaCl solutions could have reacted with chloride to form chloride complexes such as SbCl\(_{2}^{2+}\), SbCl\(_{3}^{2+}\), SbCl\(_{5}^{5-}\) and SbCl\(_{4}^{4-}\). The pH values in the system accounted for 6.30 ± 0.05–6.90 ± 0.05. Trivalent antimony ions can react with chlorine ions to create a series of complex ions in the chloride solution. The complex reactions and equilibrium constants are shown in Equation (1) where \( K_i \) is the cumulative stability constant of the complex ion at all levels (Poter & Jacobson 1970; Lothenbach et al. 1999; Chen et al. 2009):

\[
\text{Sb}^{3+} + i\text{Cl}^- = \text{SbCl}_{i}^{2+}, \quad K_i = \frac{[\text{SbCl}_{i}^{2+}]}{[\text{Sb}^{3+}][\text{Cl}^-]^i},
\]

\( i = 1, 2, 3, 4, 5, 6 \)

There are a lot of chloride molecules that could have reacted with migrated antimony from the surface of the PET polymer and formed chloride complexes at higher NaCl concentrations. Therefore, the detection of the remaining migrated antimony in chloride-rich solutions is lower than the migrated antimony in low chloride solutions. According to the results, the highest values of migrated antimony were found in distilled water because it did not contain chloride molecules which could react with antimony migrated from the surface of the PET polymer (Figure 1). The amount of antimony which was contained in drinking water was about 6 ng L\(^{-1}\), which is lower than the USEPA standard (6 μg L\(^{-1}\)) (USEPA 2007). In addition, phthalate compounds might form complexes with NaCl simultaneously. Consequently, this also caused the decrease of phthalate compounds at higher concentrations of NaCl (Table 1).

**Phthalate compounds and intermediate compounds occurring in NaCl solutions**

HPLC analysis could detect the peak of phthalate compounds and intermediate compounds leaching from PET bottles when the storage time increased at the temperature of 60 °C. Tables 2–4 show different phthalate compounds such as DMP, DEP and DEHP that occurred in NaCl solutions (from 0.1% to 50.0%) that were kept for 35 days at 60 °C, under no light. The concentrations of DMP, DEP and DEHP in 0.1%, 0.5% and 1.0% NaCl solution were higher than in distilled water, in the absence of NaCl. Longer storage times could enhance the leaching of phthalate compounds from PET bottles. The results suggested that the increase in NaCl concentration in the water led to the decrease in the concentration of phthalate compounds leaching from the PET bottles. The concentrations of DMP, DEP and DEHP that leached from the PET bottles decreased in 5.0%, 10.0%, 20.0% and 30.0% NaCl solutions.

The study of NaCl solutions (from 0.1% to 6.0%) on DMP leached from plastic bottles showed that the increasing NaCl concentration caused a decrease in the concentration of
Antimony, DMP, DEP and DEHP leached from PET bottles. The amount of leached compounds depended on the degradation of the plastic and the tolerance of PET packaging materials to NaCl concentration in the solution and the storage temperature. High temperature enhanced the degradation of plastic and accelerated the speed of diffusion of additives or contaminant molecules from the container’s polymer wall to the food. In addition, higher NaCl concentration also enhanced the decrease in the amounts of antimony and phthalate compounds in the solution. This might be due to the formation of antimony and phthalate compounds with NaCl. Although the concentrations of antimony and phthalate compounds that leached from PET bottles were not high, we need to be careful using PET bottles and we should not keep them under sunlight or at high temperatures because this will enhance the leakage of these contaminating compounds to the water. Our results implied that fish sauces in PET bottles might contain fewer antimony and phthalate compounds than does distilled water because high amounts of salt (23%–28%) caused antimony and phthalate compounds to form complexes with NaCl. In addition, mineral water and sport drinks, which contained low concentrations of NaCl, can cause higher leaching of phthalate compounds in PET bottles than does distilled water.

**Table 2** | DMP concentrations leached from PET bottles under various NaCl concentrations for a storage time of 35 days (at 60 °C and no light)

<table>
<thead>
<tr>
<th>Sample solution</th>
<th>7 days</th>
<th>14 days</th>
<th>21 days</th>
<th>28 days</th>
<th>35 days</th>
</tr>
</thead>
<tbody>
<tr>
<td>DW</td>
<td>61 ± 1&lt;sup&gt;a&lt;/sup&gt;</td>
<td>62 ± 1&lt;sup&gt;a&lt;/sup&gt;</td>
<td>61 ± 0&lt;sup&gt;a&lt;/sup&gt;</td>
<td>72 ± 0&lt;sup&gt;a&lt;/sup&gt;</td>
<td>72 ± 0&lt;sup&gt;a&lt;/sup&gt;</td>
</tr>
<tr>
<td>NaCl 0.1%</td>
<td>91 ± 1&lt;sup&gt;a&lt;/sup&gt;</td>
<td>91 ± 1&lt;sup&gt;a&lt;/sup&gt;</td>
<td>102 ± 0&lt;sup&gt;b&lt;/sup&gt;</td>
<td>102 ± 0&lt;sup&gt;b&lt;/sup&gt;</td>
<td>130 ± 0&lt;sup&gt;b&lt;/sup&gt;</td>
</tr>
<tr>
<td>NaCl 0.5%</td>
<td>91 ± 1&lt;sup&gt;a&lt;/sup&gt;</td>
<td>91 ± 0&lt;sup&gt;a&lt;/sup&gt;</td>
<td>110 ± 0&lt;sup&gt;b&lt;/sup&gt;</td>
<td>110 ± 0&lt;sup&gt;b&lt;/sup&gt;</td>
<td>121 ± 0&lt;sup&gt;b&lt;/sup&gt;</td>
</tr>
<tr>
<td>NaCl 1.0%</td>
<td>100 ± 1&lt;sup&gt;a&lt;/sup&gt;</td>
<td>101 ± 0&lt;sup&gt;a&lt;/sup&gt;</td>
<td>110 ± 0&lt;sup&gt;b&lt;/sup&gt;</td>
<td>110 ± 0&lt;sup&gt;b&lt;/sup&gt;</td>
<td>121 ± 0&lt;sup&gt;b&lt;/sup&gt;</td>
</tr>
<tr>
<td>NaCl 5%</td>
<td>ND</td>
<td>30 ± 1&lt;sup&gt;c&lt;/sup&gt;</td>
<td>51 ± 0&lt;sup&gt;c&lt;/sup&gt;</td>
<td>72 ± 0&lt;sup&gt;c&lt;/sup&gt;</td>
<td>82 ± 1&lt;sup&gt;c&lt;/sup&gt;</td>
</tr>
<tr>
<td>NaCl 10%</td>
<td>ND</td>
<td>13 ± 2&lt;sup&gt;c&lt;/sup&gt;</td>
<td>20 ± 1&lt;sup&gt;c&lt;/sup&gt;</td>
<td>31 ± 1&lt;sup&gt;c&lt;/sup&gt;</td>
<td>30 ± 1&lt;sup&gt;c&lt;/sup&gt;</td>
</tr>
<tr>
<td>NaCl 20%</td>
<td>ND</td>
<td>ND</td>
<td>12 ± 1&lt;sup&gt;c&lt;/sup&gt;</td>
<td>20 ± 0&lt;sup&gt;c&lt;/sup&gt;</td>
<td>20 ± 0&lt;sup&gt;c&lt;/sup&gt;</td>
</tr>
<tr>
<td>NaCl 30%</td>
<td>ND</td>
<td>ND</td>
<td>ND</td>
<td>12 ± 0&lt;sup&gt;c&lt;/sup&gt;</td>
<td>21 ± 0&lt;sup&gt;c&lt;/sup&gt;</td>
</tr>
</tbody>
</table>

**Table 3** | DEP concentrations leached from PET bottles under various NaCl concentrations for a storage time of 35 days (at 60 °C and no light)

<table>
<thead>
<tr>
<th>Sample solution</th>
<th>7 days</th>
<th>14 days</th>
<th>21 days</th>
<th>28 days</th>
<th>35 days</th>
</tr>
</thead>
<tbody>
<tr>
<td>DW</td>
<td>53 ± 1&lt;sup&gt;a&lt;/sup&gt;</td>
<td>60 ± 1&lt;sup&gt;a&lt;/sup&gt;</td>
<td>60 ± 1&lt;sup&gt;a&lt;/sup&gt;</td>
<td>71 ± 0&lt;sup&gt;a&lt;/sup&gt;</td>
<td>70 ± 0&lt;sup&gt;a&lt;/sup&gt;</td>
</tr>
<tr>
<td>NaCl 0.1%</td>
<td>92 ± 1&lt;sup&gt;a&lt;/sup&gt;</td>
<td>92 ± 1&lt;sup&gt;a&lt;/sup&gt;</td>
<td>102 ± 0&lt;sup&gt;b&lt;/sup&gt;</td>
<td>112 ± 0&lt;sup&gt;b&lt;/sup&gt;</td>
<td>111 ± 1&lt;sup&gt;b&lt;/sup&gt;</td>
</tr>
<tr>
<td>NaCl 0.5%</td>
<td>92 ± 0&lt;sup&gt;a&lt;/sup&gt;</td>
<td>92 ± 0&lt;sup&gt;a&lt;/sup&gt;</td>
<td>92 ± 0&lt;sup&gt;a&lt;/sup&gt;</td>
<td>102 ± 0&lt;sup&gt;b&lt;/sup&gt;</td>
<td>102 ± 0&lt;sup&gt;b&lt;/sup&gt;</td>
</tr>
<tr>
<td>NaCl 1.0%</td>
<td>92 ± 1&lt;sup&gt;a&lt;/sup&gt;</td>
<td>102 ± 0&lt;sup&gt;b&lt;/sup&gt;</td>
<td>102 ± 1&lt;sup&gt;b&lt;/sup&gt;</td>
<td>112 ± 0&lt;sup&gt;b&lt;/sup&gt;</td>
<td>111 ± 0&lt;sup&gt;b&lt;/sup&gt;</td>
</tr>
<tr>
<td>NaCl 5%</td>
<td>ND</td>
<td>22 ± 0&lt;sup&gt;c&lt;/sup&gt;</td>
<td>30 ± 0&lt;sup&gt;c&lt;/sup&gt;</td>
<td>60 ± 0&lt;sup&gt;c&lt;/sup&gt;</td>
<td>70 ± 0&lt;sup&gt;c&lt;/sup&gt;</td>
</tr>
<tr>
<td>NaCl 10%</td>
<td>ND</td>
<td>11 ± 0&lt;sup&gt;c&lt;/sup&gt;</td>
<td>11 ± 0&lt;sup&gt;c&lt;/sup&gt;</td>
<td>21 ± 0&lt;sup&gt;c&lt;/sup&gt;</td>
<td>30 ± 0&lt;sup&gt;c&lt;/sup&gt;</td>
</tr>
<tr>
<td>NaCl 20%</td>
<td>ND</td>
<td>ND</td>
<td>11 ± 0&lt;sup&gt;c&lt;/sup&gt;</td>
<td>21 ± 0&lt;sup&gt;c&lt;/sup&gt;</td>
<td>30 ± 0&lt;sup&gt;c&lt;/sup&gt;</td>
</tr>
<tr>
<td>NaCl 30%</td>
<td>ND</td>
<td>ND</td>
<td>ND</td>
<td>20 ± 1&lt;sup&gt;c&lt;/sup&gt;</td>
<td>21 ± 0&lt;sup&gt;c&lt;/sup&gt;</td>
</tr>
</tbody>
</table>

**Table 4** | DEHP concentrations leached from PET bottles under various NaCl concentrations for a storage time of 35 days (at 60 °C and no light)

<table>
<thead>
<tr>
<th>Sample solution</th>
<th>7 days</th>
<th>14 days</th>
<th>21 days</th>
<th>28 days</th>
<th>35 days</th>
</tr>
</thead>
<tbody>
<tr>
<td>DW</td>
<td>50 ± 0&lt;sup&gt;a&lt;/sup&gt;</td>
<td>61 ± 0&lt;sup&gt;a&lt;/sup&gt;</td>
<td>70 ± 0&lt;sup&gt;a&lt;/sup&gt;</td>
<td>70 ± 0&lt;sup&gt;a&lt;/sup&gt;</td>
<td>70 ± 0&lt;sup&gt;a&lt;/sup&gt;</td>
</tr>
<tr>
<td>NaCl 0.1%</td>
<td>90 ± 0&lt;sup&gt;a&lt;/sup&gt;</td>
<td>101 ± 0&lt;sup&gt;b&lt;/sup&gt;</td>
<td>101 ± 0&lt;sup&gt;b&lt;/sup&gt;</td>
<td>110 ± 0&lt;sup&gt;b&lt;/sup&gt;</td>
<td>110 ± 0&lt;sup&gt;b&lt;/sup&gt;</td>
</tr>
<tr>
<td>NaCl 0.5%</td>
<td>90 ± 0&lt;sup&gt;a&lt;/sup&gt;</td>
<td>101 ± 0&lt;sup&gt;b&lt;/sup&gt;</td>
<td>101 ± 0&lt;sup&gt;b&lt;/sup&gt;</td>
<td>110 ± 0&lt;sup&gt;b&lt;/sup&gt;</td>
<td>110 ± 0&lt;sup&gt;b&lt;/sup&gt;</td>
</tr>
<tr>
<td>NaCl 1.0%</td>
<td>90 ± 0&lt;sup&gt;a&lt;/sup&gt;</td>
<td>101 ± 0&lt;sup&gt;b&lt;/sup&gt;</td>
<td>101 ± 0&lt;sup&gt;b&lt;/sup&gt;</td>
<td>111 ± 0&lt;sup&gt;b&lt;/sup&gt;</td>
<td>120 ± 0&lt;sup&gt;b&lt;/sup&gt;</td>
</tr>
<tr>
<td>NaCl 5%</td>
<td>ND</td>
<td>20 ± 0&lt;sup&gt;c&lt;/sup&gt;</td>
<td>30 ± 0&lt;sup&gt;c&lt;/sup&gt;</td>
<td>30 ± 0&lt;sup&gt;c&lt;/sup&gt;</td>
<td>110 ± 0&lt;sup&gt;b&lt;/sup&gt;</td>
</tr>
<tr>
<td>NaCl 10%</td>
<td>ND</td>
<td>ND</td>
<td>20 ± 0&lt;sup&gt;c&lt;/sup&gt;</td>
<td>30 ± 0&lt;sup&gt;c&lt;/sup&gt;</td>
<td>30 ± 0&lt;sup&gt;c&lt;/sup&gt;</td>
</tr>
<tr>
<td>NaCl 20%</td>
<td>ND</td>
<td>ND</td>
<td>ND</td>
<td>20 ± 0&lt;sup&gt;c&lt;/sup&gt;</td>
<td>20 ± 0&lt;sup&gt;c&lt;/sup&gt;</td>
</tr>
<tr>
<td>NaCl 30%</td>
<td>ND</td>
<td>ND</td>
<td>ND</td>
<td>10 ± 0&lt;sup&gt;c&lt;/sup&gt;</td>
<td>21 ± 0&lt;sup&gt;c&lt;/sup&gt;</td>
</tr>
</tbody>
</table>

DW: distilled water.
ND: not detected (detection limit ≥ 2 ng l<sup>–1</sup>).
<sup>a</sup>, <sup>b</sup>, <sup>c</sup>: Values in the same row with the same letter are not significantly different (α = 0.05).
ACKNOWLEDGEMENTS

This research is supported by the grant under the program Strategic Scholarships for Frontier Research Network for the Joint PhD Program Thai Doctoral Degree from the Office of the Higher Education Commission, Thailand.

REFERENCES

CID=1102&DID=4645&DOC=FILE.PDF (accessed 10 October 2010).
Duh, B. 2002 Effect of antimony catalyst on solid-state polycondensation of poly(ethylene terephthalate). Polymer 43 (11), 3147-3154.


First received 25 November 2014; accepted in revised form 18 February 2015. Available online 4 March 2015