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Full Paper

Electrochemical Determination of Bisphenol A in Polycarbonate Bottles on MWCNTs-Modified Glassy Carbon Electrode in Kenya

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Abstract- Endocrine-disrupting chemicals are pollutants known to disrupt metabolism, and hormone biosynthesis or alter how the endocrine system works. Research has shown that Bisphenol A (BPA) mimics estrogen and thyroid hormone therefore a potential endocrine system disruptor. Exposures to Bisphenol A above threshold levels leading to endocrine disruption are expected when there is persistent use of products made using bisphenol A. Among the key sources of BPA that leach to the container's contents through hydrolysis are polycarbonate containers. In this study, we monitor levels of bisphenol A exposure to users of polycarbonate containers using cyclic and differential pulse voltammetry in Thika town. Results obtained for the bisphenol A levels using voltammetric method for all the new and old sampled bottles subjected to varying conditions like time and temperature were in the range 0.3–1.6 µgmL^{-1} for water samples and 2.2–3.7 µgmL^{-1} for baby milk samples. As temperatures were increased from 25°C to 95°C, the amount of BPA leaching from the polycarbonate containers increased into water and milk. Further, results obtained show that the amount of Bisphenol A leaching increased in all the bottles, both new and used as contact time increased from 1 hour to 12 hours. All the new and old samples overshot the tolerable daily intake for Bisphenol A of 4 µg/kgbw/day allowed by the European Food Safety Authority (EFSA) and this rings an alarm for careful and thorough attention.

Keywords- Bisphenol A; Baby feeding bottles; Water bottles; Polycarbonate plastics; Voltammetry

1. INTRODUCTION

Emerging pollutants are chemicals which have recently been found to be treacherous to the ecosystem including the health of human beings. They have been named "*emerging*" because of the ever-increasing level of concern associated with them. Most surprisingly, the majority of these emerging contaminants have not been modulated under national and international legislation. This poses a much greater risk to the livelihood of human beings. These contaminants include but are not limited to endocrine disruptors, steroids, industrial additives, hormones, antibiotics, drugs, and microplastics [1-4].

In particular, endocrine-disrupting chemicals are pollutants known to interfere with metabolism, and hormone biosynthesis or alter the working of the endocrine system. Bisphenols are predominantly manufactured worldwide. They are known to be among the major sources of pollution in our environment. Bisphenol A (BPA) is one of the main types of Bisphenol found in high quantities globally and has been found to have adverse impact on our ecosystem. Bisphenol A has a chemical name and structure shown in Figure 1. It's mainly used in the production of polycarbonates [5,6] with plastic additives. Polycarbonates are used in the manufacture of water and food contact products like containers [7]. Studies have shown that Bisphenol A leaches from containers like bottles into their contents [8,9].

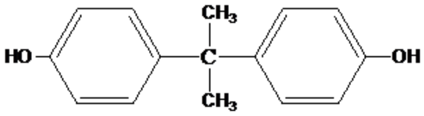


Figure 1. Chemical structure for Bisphenol A whose chemical name is (2,2-bis(4-hydroxyphenyl) propane) [10]

Research has shown that Bisphenol A mimics estrogen and thyroid hormone therefore a potential endocrine system disruptor. By extension Bisphenol A acts as a metabolic and immune disruptor, implying a potential cause of hormonal imbalance in natural systems. In a nutshell, Bisphenol A has a disastrous health effect on humans and the general ecosystem [11]. Bisphenol A has exhibited oestrogenic activity, specifically at tender stages of life. It interferes with foetal development leading to low birth weight and inhibited maturation in body organ systems as a result of interference with organogenesis and histogenesis processes [12]. In fact, more than 40 countries have phased out the use of plastics made using Bisphenol A as baby feed and water storage containers in their countries [13]. Among the exposure routes of Bisphenol A to human beings include inhalation, consumption, and even contact with the skin. Therefore, it's imperative to carry out more studies on the exposure and effects of Bisphenol A as a protection measure for human health and our ecosystem in general.

Unfortunately, in developing countries such as Kenya, polycarbonate plastics are still being used to manufacture polycarbonate products. A popular use of polycarbonate materials is in the manufacture of portable water-carrying bottles and baby food containers of different sizes and shapes [14]. Such usage and the fact that there are no regulatory measures in place exposes innocent polycarbonate users to the dangerous xenoestrogenic compound due to possible leaching into the container contents. Studies have shown that Bisphenol A can leach from polycarbonate materials through hydrolyzation processes [8]. Further, the extent of Bisphenol A leachability depends on the pH, temperature, and contact time [8]. It’s also important to note that polycarbonate portable water-carrying bottles are also used for carrying juices. These practices can easily enhance Bisphenol A leachability. Consequently, it is imperative to monitor levels of exposure of Bisphenol A to polycarbonate plastic users in Kenya depending on the usage of the said bottles and to advise the users accordingly. This study monitors the extent of bisphenol A leachability from polycarbonate plastics into water and baby milk using voltammetric techniques in Kenya.

2. EXPERIMENTAL SECTION

2.1. Sampling area

Sampling was carried out in Thika town, Kiambu County, Kenya with a catchment area of 839 km^2 and a population of about 279,429 people (From the Kenya National Bureau of Statistics, 2019). The town lies between longitude 37°4'9.59"E and latitude1°1'59.74"S (Figure 2).

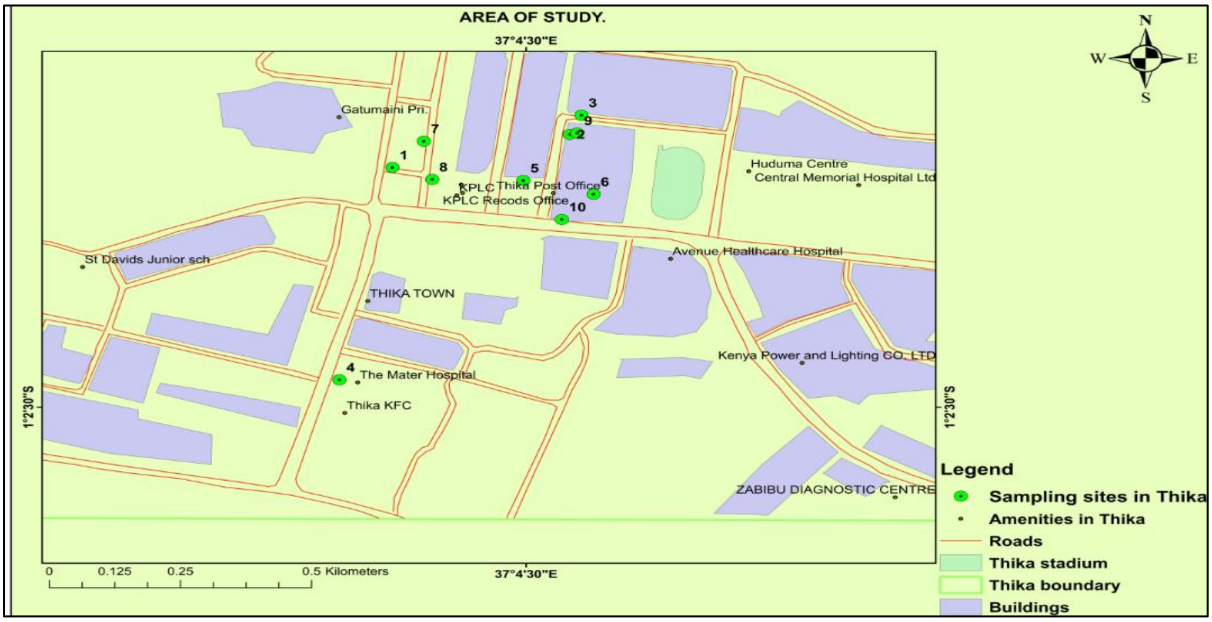


Figure 2. The map of Thika town showing the sampling sites for new and used polycarbonate water and baby milk bottles

Its high population and typical urban set-up mean possibility of many polycarbonate users. Sampling was done in ten randomly selected supermarkets/sampling sites hereby designated as sampling sites 1 -10. All new bottle samples were purchased. At the same time used bottles were also sampled from nearby homes randomly in exchange for new ones.

2.2. Chemicals and reagents

Reagents used in voltammetry are: Potassium ferricyanide ($K_3Fe(CN)_6$, 99.99%) from Fisher Scientific; 99.8% potassium nitrate, potassium dihydrogen phosphate, sodium dihydrogen orthophosphate and disodium hydrogen phosphate from BDH PROLABO; potassium chloride, copper (II) sulphate, lead II nitrate, calcium chloride, magnesium nitrate, sodium nitrate and manganese sulphate all purchased from Sigma Aldrich. 99.9% dimethyl sulfoxide, 99.9% dimethylformamide (DMF) purchased from Alfa Aesar. Double distilled and de-ionized water was used in all studies. All the chemicals and reagents purchased were of analytical grade hence used without any further purification.

2.3. Instruments

The instrument used for all spectroscopic study was Shimadzu IRAffinity-1S - Fourier Transform Infrared Spectrophotometer. All electrochemical experiments were conducted on a Basi Epsilon Eclipse potentiostat composed of a three-electrode system. The three-electrode system comprised of a glassy carbon as the working electrode, a platinum wire as the auxiliary electrode and Potassium-saturated calomel electrode as the reference electrode. All the voltammetric experiments were performed at room temperature in a 10.0mL electrochemical cell. All data were collected and analyzed using Kaleidagraph software, version 4.1.1.

2.4. Sample collection and pretreatment

During sampling, ‘suspected’ polycarbonate bottles were purchased from the various supermarkets/sampling sites. Most of the bottles did not have a polycarbonate label and selection was done on generally known characteristic of polycarbonate materials such as; strength, stiffness, transparency, rigidity amongst others [15]. In total twelve (12) samples were purchased of which six were baby feeding and six were portable water carrying bottles. At the same time used bottles were also sampled from nearby homes randomly selected. In this study, bottles which were more than two months in usage were regarded as “*used bottles*” and were obtained in exchange for new ones. In total twenty-four (24) ‘suspected’ polycarbonate bottles were sampled of which twelve (12) were drinking water bottles composed of six used and six new bottles. The other twelve (12) were baby feeding bottles composed of six used and six new bottles. Each sample purchased or/and collected from the mothers within Thika was stripped with labeling masks and were labelled A, B, C, D, E, F, G, H, I, J, K and L and transported to the physical chemistry laboratory of the department of Chemistry at the University of Nairobi for analysis.

The sampled baby feeding bottles (BFBs) and drinking water bottles (DWBs) obtained from the local supermarkets were cut into tiny pieces with scissors. The polymer identification of the cut pieces was analyzed by Fourier-transform infrared FTIR spectroscopy. Before each measurement the ATR crystals were cleaned using ethanol. A background spectrum was run on the cleaned ATR crystals to ascertain complete evaporation of the solvent. Reference spectra were used to accomplish polymer identification of the samples. From the analysis, all the 24 samples were confirmed to be of polycarbonate material.

2.5. Preparation of baby milk

The purpose of this part was to prepare the baby milk for protein precipitation. First, 3 spoons (12.0 g) of the baby formula were added to 100 ml of boiling water in a polycarbonate baby feeding bottle and the mixture was stirred well then left to stand for 2 hours. After that, 100 µL of concentrated hydrochloric acid was added to acidify the mixture. From the acidified solution, 5mL was taken and poured into 10 mL of acetonitrile in a 20 mL conical flask. The expected precipitate that formed was centrifuged at 4000 rpm for 10 minutes. Finally filtered using Whiteman filter papers [16].

2.6. Preparation of phosphate buffer solution

0.1 M phosphate buffer solution was prepared by dissolving 9.453 g disodium hydrogen phosphate (Na_2HPO_4) and 5.21 g of sodium dihydrogen phosphate ($NaH_2PO_4.2H_2O$) separately in 800 mL distilled water. This salt solution was mixed in accordance to the Henderson-Hasselbalch equations to attain the required pH range of 7.0. The pH of the phosphate buffer solution was maintained by storing it in a refrigerator at 4 ºC.

2.7. Preparation of Bisphenol A standard solution

A stock solution of 500 µM Biphenol A standard was prepared by adding 11.41 mg of the Bisphenol A standard in 100 ml of methanol. The working standard solutions designated as 10 µM, 20 µM, 30 µM, 40 µM and 50 µM were prepared from the 500 µM standard stock solution.

2.8. Preparation of interferent solutions

To prepare interferant solutions, a measured amount (Table 1) of salt was weighed and dissolved in 25 ml of distilled water while stirring. The concentrations of all interferent salt solutions were 0.1 M. Then diluted later accordingly.

Table 1. Preparation of interferant solutions from salts of interest

Type of salt	Mass of salt dissolved (grams)
Magnesium chloride	0.5000
Sodium nitrite	0.1725
Lead nitrate	0.8280
Potassium dihydrogen orthophosphate	0.3400
Calcium chloride	0.2775
Magnesium nitrate	0.6410
Zinc chloride	0.3400
Potassium chloride	0.1864
Sodium nitrate	0.2120
Copper sulphate	0.6242
Potassium nitrate	0.2528
Sodium dihydrogen orthophosphate	0.3900

2.9. Preparation of the MWCNTs-modified glassy carbon electrode

The surface of the bare glassy carbon electrode was modified with multi-walled carbon nanotubes by drop casting method. A bare glassy carbon electrode (GCE) was polished with alumina powder of different sizes to produce a surface that is mirror-like, rinsed using de-ionized water then dried at room temperature before use. 1.0 mg of multiwalled carbon nanotubes (MWCNTs) were dispersed in 1.0 mL of dimethylformamide to make a suspension. A 5 µL aliquot of the MWCNTs suspension was casted on the GCE surface then air dried for 1 hour at room temperature. The dried electrode was designated as MWCNTs/GCE and used for voltammetric measurements without further treatments.

2.10. Voltammetric Analysis of BPA

Bisphenol A is an electroactive compound that gives an oxidation peak at around 0.60 V vs. SCE on a polished glassy carbon electrode (GCE) [17]. The Bisphenol A leaching studies were investigated using both cyclic and differential pulse voltammetry by scanning from -1.0 V to +1.0 V potential window. By using different concentrations of Bisphenol A in the electrolytic cell, the peak current dependence on the concentration of Bisphenol A was monitored by cyclic and differential pulse voltammetry within a potential window of -1.0 V to +1.0 V. The known Bisphenol A concentrations versus the voltammetric currents were used to make calibration plots from which sample concentrations were determined. The key variables considered were contact time (1 hour, 6 hours, and 12 hours) and temperatures (25°C, 50°C, and 95°C).

3. RESULTS AND DISCUSSION

3.1. FT-IR Identification of the bottle samples as Polycarbonate

It was important first to confirm that all the bottles sampled were of polycarbonate. This was done by comparing their FT-IR spectra with the known polycarbonate standard spectra. Figure 3 shows the FT-IR signatures for both the samples and polycarbonate standard.

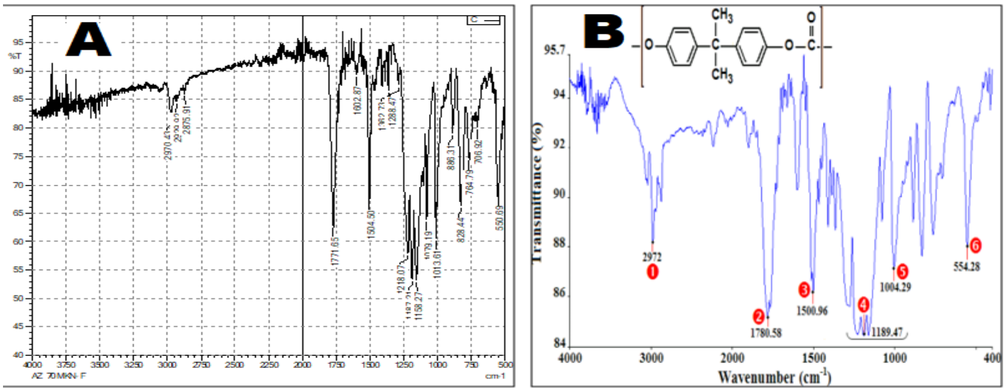


Figure 3. (A) The FT-IR spectrum of polycarbonate water bottle used in the study; (B) FT-IR spectrum obtained from the literature for polycarbonate [18]

The prominent peaks in the polycarbonate infra-red spectrum shown in Figure 3(A) are as result of the following characteristic vibrations: at 1770 from carbonyl group (C=O) vibration, at 1504 for C=C vibration rings, at 2970 to the stretching vibration of C-H aromatic ring, at 1079 attributed to CH_3 vibrations and at 1187 and 1013 from the two phenol rings corresponding to stretching deformations of asymmetric and symmetric O-C-O carbonate group. These results for polycarbonate plastics shown in Figure 3(B) are consistent with observations by Ghorbel et al. [18]. The FT-IR analysis results confirmed that all the water and baby milk bottles used in this work were of polycarbonate.

3.2. Electrochemical Characterization of MWCNTs/GCE Electrodes

Conventional bare electrodes have various limitations including low sensitivity and selectivity towards specific analytes. Introduction of multifunctional materials such as carbon nanotubes and/or polymers with high specific surface area and good electrical conductivity can help overcome these challenges. Potassium ferricyanide is well behaved electrochemically [19,20]. Hence, widely used as a model for characterization of electrochemical reactions. Thus, using potassium ferricyanide as the electroactive probe, cyclic and differential pulse voltammograms were obtained for the bare glassy carbon (GC) and the MWCNTs-modified GC electrodes shown in Figures 4(A) and 4(B). From cyclic voltammetry, it was observed that the current increased from 38.60 μA of a bare glassy carbon electrode to 42.02 μA of

MWCNTs-modified glassy carbon electrode. While from differential pulse voltammetry, the current increased from 77.0325 µA of a bare glassy carbon electrode to 81.5338 µA of the MWCNTs-modified glassy carbon electrode. These current increases can be attributed to the presence of multi-walled carbon nanotubes which possibly enhanced the sensitivity and the conductivity of the glassy carbon electrode surface. These results are consistent with earlier observations by Guto, P.M. [19,21].

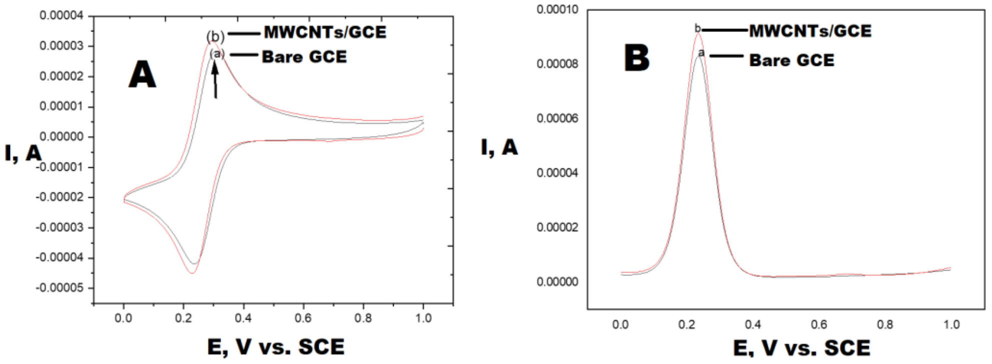


Figure Error! No text of specified style in document.. (A) Cyclic voltammograms (B) Differential pulse voltammograms of bare GC electrode and MWCNTs-modified GC electrodes in 0.1 M KNO_3 with 3.0 mM $K_3Fe(CN)_6$

3.3. Studying the catalytic activity of MWCNTs/GCE towards Bisphenol A

The electroactivity of Bisphenol A was tested using cyclic voltammetry (CV) on bare and modified glassy carbon electrode. The cyclic voltammograms of BPA (Figure 5) were obtained in 0.5 mM Bisphenol A in pH 7.0 phosphate buffer at potential range of -1.0 V to +1.0 V and scan rate of 50 mV/s.

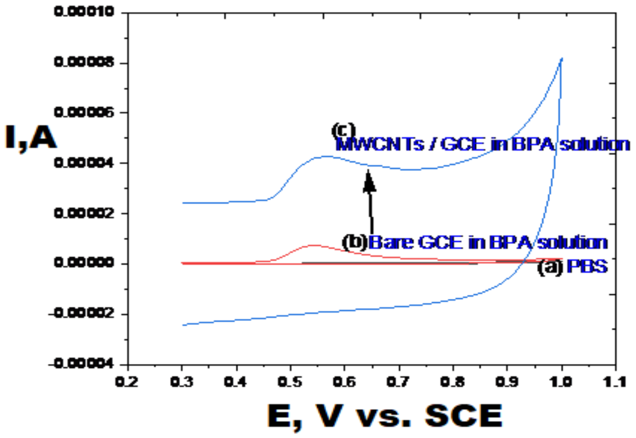


Figure 5. Cyclic voltammograms of (a) phosphate buffer, pH 7.0 (b) 20 µL of 0.5 mM Bisphenol A at bare GC electrode and (c) 20 µL of 0.5 mM Bisphenol A at MWCNTs-modified GC electrode. The scan rate was 50 mV/s.

It was observed that the oxidation peak of Bisphenol A was at +0.55 V. This result was consistent with results obtained by Gugoasa, et al. [22,23]. It's important to note that modifying the glassy carbon electrode with MWCTs increased the oxidation peak current by about 100% as shown in Figure 5 for Bisphenol A. Generally, modifying the glassy carbon electrode with MWCNTs leads to increased sensitivity for Bisphenol A oxidation hence enhanced peak current and possible reduction of surface fouling.

The results obtained from differential pulse voltammetry (Figure 6) are similar to those from cyclic voltammetry because there is increase in the voltammetric current. Results from both cyclic voltammetry and differential pulse voltammetry suggests that modifying the glassy carbon electrode with MWCNTs contributes to improvement of electrochemical electrode response towards Bisphenol A in terms of electron-transfer rate. Similar improvement of electrode signal has been observed previously by Han and co-workers [24].

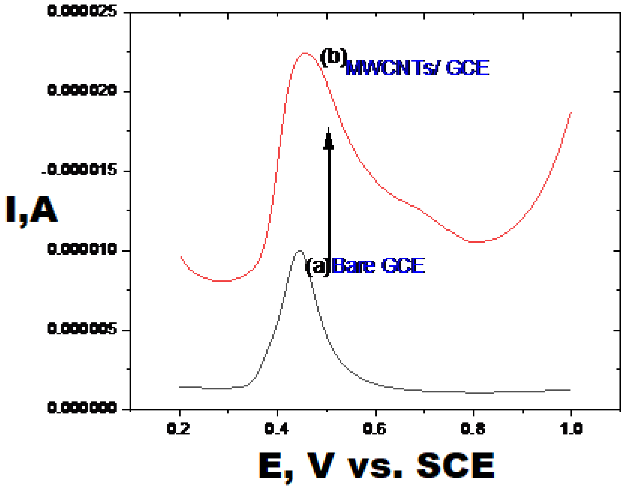


Figure 6. Differential pulse voltammetry of 20 µL of 500 µM Bisphenol A on bare GC electrode (a) and 20µL of 500 µM Bisphenol A at MWCNTs/GC electrode (b); supporting electrolyte was 0.1 M phosphate buffer, pH 7.0

3.4. Scan Rate study of Bisphenol A

Bisphenol A is an electroactive molecule. The electrochemical behavior of Bisphenol A is typically an irreversible electrode reaction similar to other phenolic compounds [17,25]. Ordinarily, scan rate and voltammetric peak current relationships provide valuable information involving electrochemical mechanism of the system under investigation. Cyclic voltammograms were taken for 0.5 mM Bisphenol A for scan rates from 10 mV/s to 60 mV/s in solution using the MWCNTs-modified electrode as shown in Figure 7A. It's important to note that the low scan rates selected allows more time for the analyte to diffuse to the electrode surface for detection. The anodic peak currents (I_{pa}) were found to increase with rise in scan rate (Figure 7B). This observation generally suggests that the electro-oxidation of Bisphenol A on the MWCNTs-modified glassy carbon electrode was a surface-controlled electrode process [26, 27].

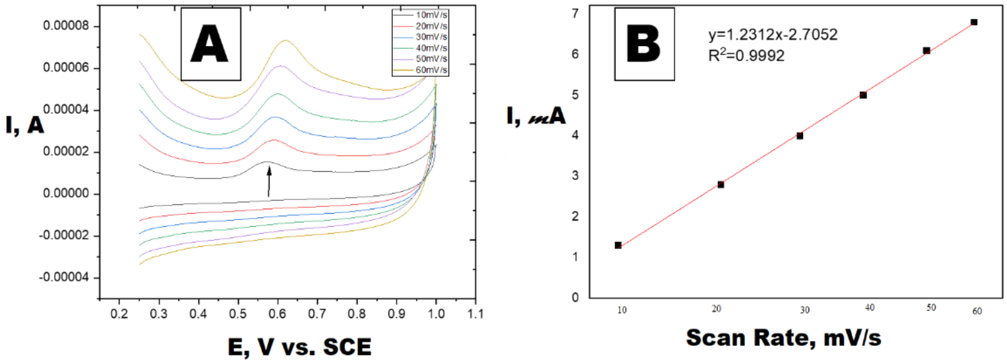
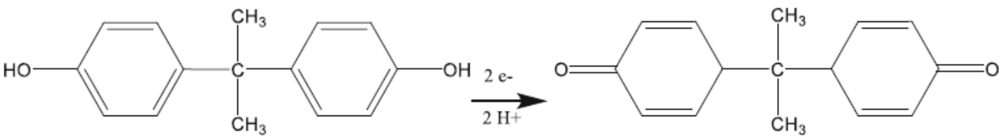


Figure 7. (A) An overlay of cyclic voltammograms of 0.5 mM Bisphenol A in 0.1 M phosphate buffer, pH 7.0 on MWCNTs-modified glassy carbon electrode at scan rates from 10 mV/s to 60 mV/s; (B) A plot of voltammetric peak currents against scan rate

To understand the mechanism of Bisphenol A oxidation better, we investigated the number of electrons, *n*, involved in the electro-oxidation process. Plots of potential, E, versus log[i/(i_d-i)] were made (equation 1) whose slopes were used to obtain the number of electrons, *n* [28].

$$E = E_{1/_2} - \frac{0.0591}{n} log\left(\frac{i}{i_d - i}\right) \qquad (1)$$

Where E is the potential at any point on the wave, $E_{1/2}$ is the half-wave potential, i is the current at any point on the wave, i_d is the peak current and *n* is the number of electrons exchanged in the oxidation process. Consequently, *n* was found to have a value of 2 for the oxidation of Bisphenol A on MWCNTs-modified glassy carbon electrode. Bisphenol A's oxidation mechanism can be linked to the hydroxyl functional group on the aromatic ring in Bisphenol A [29]. The electrode process involves two-electron and two-proton (Scheme 1).



Scheme 1. Electro-oxidation mechanism of Bisphenol A

3.5. Interference study

The impact of numerous substances which are likely to interfere with Bisphenol A determination were investigated using cyclic voltammetry. The interferants were chosen on the basis that they could occur in the specimen matrices in baby feeds and water. Furthermore, they are electrochemically active and it was important to ensure their electrochemical signals did not interfere significantly with Bisphenol A detection. Using cyclic voltammetry, known concentration of interferants were added to known concentration of Bisphenol A. The mixture was scanned from -1.0 V to +1.0 V potential window at a scan rate of 50 mV/s. The resulting currents and oxidation potentials were observed and recorded as shown in Table 2.

Table 2. The potential and oxidation currents of interferants in phosphate buffer, pH 7.0

Interferent	**Potential(V)**	**Current(µA)**
Control	0.687	5.371
Magnesium chloride	0.688	5.337
Sodium nitrite	0.689	5.957
Lead nitrate	0.689	5.168
Potassium dihydrogen orthophosphate	0.680	5.167
Calcium chloride	0.685	5.421
Magnesium nitrate	0.685	5.535
Zinc chloride	0.688	6.597
Potassium chloride	0.676	6.407
Sodium nitrate	0.688	5.167
Copper sulphate	0.689	7.274
Potassium nitrate	0.684	5.535
Sodium dihydrogen orthophosphate	0.684	6.265

Results in Table 2 shows that the selected interferants had negligible interference in the 50-fold concentrations of Ca^{2+}, Mn^{2+}, Pb^{2+}, Mg^{2+}, Zn^{2+}, Cu^{2+}, SO_4^{2-}, K^+, Na^+, Cl^-, NO_3^-, and PO_4^{3-} ions. Moreover, there is no significant change in Bisphenol A oxidation potential, as the mean was 0.6855 and a standard deviation of 0.0039 resulting in a relative standard deviation of 0.57%.

3.6. pH optimization for Voltammetric determination of Bisphenol A

The pH of an electrolyte solution has a significant effect on the acid-base dissociation of Bisphenol A influencing directly on its oxidation potential and current. Therefore, the effect of pH on the oxidation peak current and peak potentials of Bisphenol A on MWCNT-modified GC electrode was investigated from pH 4 to pH 12 by cyclic voltammetry (Figure 8A). It was observed that the peak currents of Bisphenol A gradually increased with increasing pH value up to pH 7, then started to decrease as shown in Figure 8B. Thus, the pH 7.0 was chosen as the optimal pH for the detection of Bisphenol A.

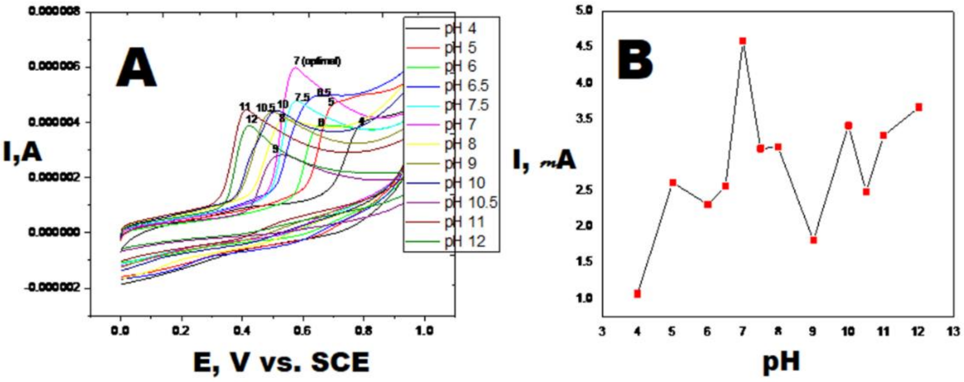


Figure 8. (A) Cyclic voltammograms obtained for MWCNTs-modified GC electrode against 0.5 mM Bisphenol A at different pH values; (B) Voltammetric peak currents obtained from Figure 8(A) versus pH values

3.7. Determination of BPA in Water and Milk samples using Differential pulse voltammetry

To determine the presence and concentration of Bisphenol A in baby milk and water samples, first a calibration plot of Bisphenol A was made. This was done by spiking Bisphenol A concentrations in the range of 0-50 µM on the MWCNTs-glassy carbon electrode in pH 7.0 phosphate buffer. The resultant differential pulse voltammetric responses after addition of each aliquot of the Bisphenol A standard were recorded (Figure 9A) and used to construct the calibration curve given in Figure 9B. Consequently, the calibration curve was then used to obtain Bisphenol A amounts in baby milk and water samples.

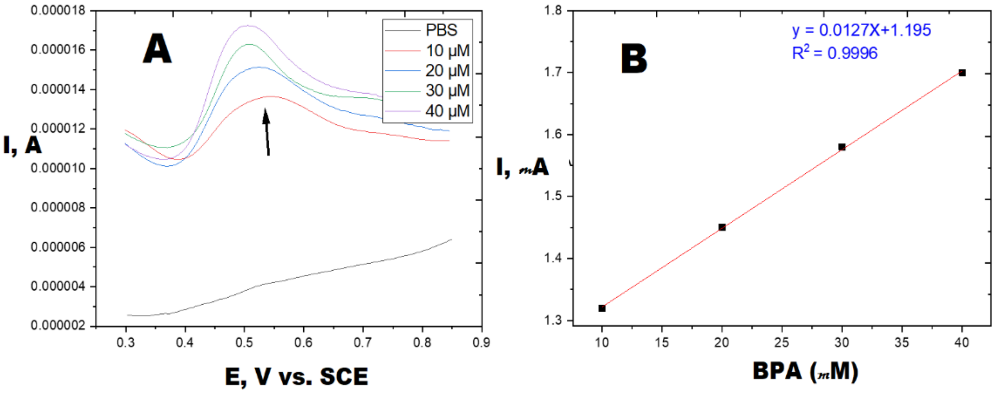


Figure 9. (A) Differential pulse voltammograms of Bisphenol A at different concentrations on MWCNTs/GC electrode in pH 7.0 phosphate buffer solution; (B) Plot of differential pulse voltammetric Peak currents versus concentrations of Bisphenol A. The current readings were obtained from Figure 9(A).

Differential pulse voltammetry (DPV) was used to determine Bisphenol A in the baby milk and water bottle samples. Table 3 shows that there is more leaching of Bisphenol A from used bottles compared to new bottles at 25ºC, 50ºC and 95ºC for water and milk samples. It's important to note that used bottles are normally subjected to harsh conditions during washing and sterilization normally practiced by mothers before using the bottles to feed the baby. It's important to note that the washing, brushing and sterilizing loosens the Bisphenol A polymer bonds on the container material surface. Consequently, this action causes free Bisphenol A to leach into the bottle contents. This would mean where no polycarbonate bottles alternatives exist, then the age of the bottle should be put into consideration. Besides, alternative cleaning methods involving no use of heat treatment or harsh chemicals should be desired. It's also evident from Table 3 that leaching of Bisphenol A increased with increasing temperature, that is, from 25ºC to 95ºC for both new and used bottles. Increased Bisphenol A leaching at high temperatures maybe as result of accelerated hydrolysis of carbonate linkages as observed earlier by Vilarinho and co-workers [30].

Table 3. The effect of temperature on the amount of Bisphenol A leaching into water and baby milk monitored by DPV

No.	**Sample**	**Bottle Type**	**Temp. ºC**	**BPA Conc., µgmL^{-1}**
1.	Water	New	25	0.3
		Old	50	0.4
		New	95	0.8
		Old	25	1.2
		New	50	1.3
		Old	95	1.6
2.	Baby Milk	New	25	2.2
		Old	50	2.7
		New	95	2.7
		Old	25	3.2
		New	50	3.0
		Old	95	3.7

Generally, many studies have reported increasing concentrations of Bisphenol A leached when storage temperatures increase. Mai et al. [31] in Riyah, Saudi Arabia found that the mean concentrations of BPA in drinking water kept indoors were 4.03 ng/L while those kept outdoor exposed to natural sunlight were 7.5 ng/L. In packaged drinking water, Neha and Pandya [32] in India found that the range of BPA concentrations were from 0.25 to 2.25 ng/L. Interestingly, in Jeddah city Saudia Arabia, Lina et al. [33] found that the following bottled drinking waters - at room temperature, exposed to sunlight and another with boiled water had 9.46 ng/L, 16.13 ng/L and 14.7 ng/L BPA concentrations, respectively.

Results of this study are very similar to what was reported by Zhang et al. [34] where Bisphenol A migrated in the ranges of 0.50–0.84 µgmL^{-1} in four different polycarbonate containers. The current results are also comparable to results obtained by Bashir and Audu [16] where Bisphenol A migration in hot water and milk stored in polycarbonate bottles were in the range of 0.40-0.52 µgmL^{-1} for three hot water samples and 3.85-1.78 µgmL^{-1} for the milk samples respectively. Another study conducted by Xu and colleagues also reported Bisphenol A concentrations in water and milk stored in polycarbonate bottles ranged from 0.56 – 0.68 µgmL^{-1} and 0.13–0.27 µgmL^{-1} respectively. Bisphenol A concentrations they found in milk were much lower than the present study probably due to different sources of the milk as milk powder was used in their study whereas fresh milk was used in the present study.

Contact time is one of the key determinants on the amount of Bisphenol A leaching from polycarbonate bottles as demonstrated earlier by Yun et al. [35]. Table 4 shows Bisphenol A amounts monitored by differential pulse voltammetry over a period of 12 hours. More Bisphenol A was detected in used polycarbonate bottle contents compared to new bottle contents. Also, there was observable increase of Bisphenol A amounts in water as contact time increased from 1 hour to 12 hours. These results show that polycarbonate bottles leach Bisphenol A into its contents and the situation is worsened when the contents are stored for long hours.

Table 4. The effect of contact time on Bisphenol A leaching in water monitored by DPV

No.	Sample	Bottle Type	Time (Hours)	BPA Conc. µgmL^{-1}
1.	Water	New	1	1.3
		Old	1	1.8
		New	6	2.4
		Old	6	2.8
		New	12	3.1
		Old	12	3.5

The trend of this results is similar to what was observed by Kubwabo et al. [36] who demonstrated that increasing storage duration increased concentration of Bisphenol A leaching from polycarbonate water bottles into water. On the other hand, Yun et al. [35] from Selangor, Malaysia reported increased concentration of Bisphenol A leaching from polycarbonate water bottles with prolonged durations.

The recommended safe maximum known as the tolerable daily intake (TDI) of Bisphenol A allowed by European Food Safety Authority (EFSA) of 2023 is 4 µg/kgbw/day [37]. The average migration value of Bisphenol A in water and milk samples obtained in this study at 25°C from new bottles were 0.3 µgmL^{-1} and 2.2 µgmL^{-1} respectively while from used bottles were 0.4 µgmL^{-1} and 2.7 µgmL^{-1} respectively. The estimated daily intake from these results assuming an infant of 5kg consuming at least 250 ml of water and milk in a day, would be 15 µg/kgbw/day and 110 µg/kgbw/dy respectively for the new bottles and 20 µg/kgbw/day and 135 µg/kgbw/dy respectively for the used bottles. This indicates that the average daily intake of Bisphenol A in water and milk in the present study exceeds the set tolerable daily intake of 4 µg/kgbw/day.

It's evident that use of polycarbonate feeding and water bottles in Kenya cannot be regarded as safe. The level of Bisphenol A consumed at every feeding of the infant exceeds the tolerable daily intake allowed by the EFSA.

4. CONCLUSION

With differential pulse voltammetry, Bisphenol A leachability for all the water and baby milk samples were between 0.3–1.6 µg/mL and 2.2–3.7 µg/mL respectively. Results from differential pulse voltammetry confirmed that as the temperatures increased from 25ºC to 95ºC, the amount to BPA leaching from the polycarbonate containers increased into water and milk. Moreover, there is more leaching of Bisphenol A from used bottles compared to new bottles at 25ºC, 50ºC and 95ºC for both water and milk samples. It's evident that the amount of Bisphenol A leaching increased in all the bottles, both new and used as contact time increased from 1 hour to 12 hours. All the samples analyzed by differential pulse voltammetry surpassed the Bisphenol A tolerable daily intake (TDI) of 4 µg/kgbw/day recommended by EFSA. Therefore, it's possible to mention that polycarbonate bottles currently used in Kenya are not safe for storing baby feeds and drinking water.

Declarations of interest

The authors declare no conflict of interest in this reported work.

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