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Novel Potentiometric Screen-printed Carbon Electrodes for Bisphenol S Detection in Commercial Plastic Samples

Hisham S. M. Abd-Rabboh*,† and Ayman H. Kamel****

**Department of Chemistry, Faculty of Science, King Khalid University, Abha 61413, Saudi Arabia*
***Department of Chemistry, Faculty of Science, Ain Shams University, Abbassia 11566, Cairo, Egypt*

Novel miniaturized poly(vinyl chloride) matrix membrane sensors based on screen-printed carbon electrodes and responsive to bisphenol S (BPS) were formulated. Polymeric membranes are based on an ion-pair complex of BPS anion with an Aliquat 336S counter cation. A solid conductive contact of multi-walled carbon nanotubes (MWCNTs) was used on screen-printed carbon platforms. After drop-casting and drying of the MWCNTs on a carbonaceous substrate, it was coated with a layer of polymeric poly(vinyl chloride) PVC sensing membrane containing the recognition complex. Prepared electrodes revealed a near-Nernstian response towards BPS with a -28.2 ± 0.8 mV/decade anionic slope, 0.02 µg/mL detection limit and 2.5×10^{-7} – 1.0×10^{-3} M concentration range ($r^2 = -0.9994$). Signals were recorded in a 30 mM HCO_3^-/CO_3^{2-} buffer, pH 10, with fast response times <10 s. A suggested sensing system was effectively applied in the quantitative determination of diminished BPS levels released from plastic bottle samples, and obtained results were statistically assessed against a chromatographic HPLC independent reference method.

Keywords Bisphenol S (BPS), screen printed carbon electrode, potentiometric sensors, multi-walled carbon nanotubes (MWCNTs), solid-contact sensors

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Introduction

Bisphenol S, BPS, (4,4′-sulfonyl diphenol) is a material that consists of double phenol rings combined by a sulfonyl functional group, which is commonly used as an alternative for bisphenol A, BPA, (4,4′-(propane-2,2-diyl)diphenol) in plastic products. It has higher thermal stability than BPA, making it more applicable in the processes of thermal formulation and manufacturing of plastics.[1] It is also commonly used in curing fast-drying epoxy resin adhesives and cyclic carbonates.[1,2] BPS is widely produced in China for a wide range of applications, such as a chemical additive in pesticides and dyes, a leather tanning agent, a color fast agent, and for the improvement of fibers.[3,4] The vapor pressure of BPS indicates that the chemical compound will not volatilize from dry soil surfaces when present in natural environments. The log K_{OW} of BPS, which is a measure of the partition coefficient of the chemical compound in the octanol/water system, was found to be 1.2, while for BPA it was 3.4, indicating that BPS would be more hydrophilic in nature when compared to BPA (*i.e.* the water solubility of BPS is 1100 mg/L and that for BPA is 300 mg/L).[5] Cao *et al.*[6] found that BPS possesses enhanced mechanical properties and thermal stability for its polymeric products, along with strong resistance towards biodegradation under both aerobic and anaerobic conditions, compared to BPA, making it favorable for replacing BPA in almost all of its plastic products.[7]

Human urine samples collected in many counties around the globe (*e.g.* China, Japan, USA, and Kuwait) were found to contain levels of BPS.[8] The highest detection level of BPS in urine samples was found in Japan, as their mean daily intake value of the chemical was about 3.47 mg/day, followed by USA with a mean BPS daily intake concentration of 1.48 mg/day. The estrogenic behavior of BPS *in vitro* and *in vivo* tests was found to be similar to that of BPA.[9] It was found that both BPA and BPS impact the hypothalamic development in humans.[10] Hence, when both humans and the aquatic environments are exposed to BPS, it can pose a serious threat to the health and the existing ecosystem conditions. Very little literature was available concerning the occurrence of BPS in surface waters. With the substitution of BPS in many polycarbonate plastic products, the point and non-point sources for BPA pollution could be possible pathways for BPS to enter into the surface and groundwater systems.

Based on the increased production and use of BPS, it was used as a model organic pollutant, and many analytical procedures were introduced for its sensitive assay. These approaches include UV-spectrophotometry,[11] high-pressure liquid chromatography (HPLC)[12] and ultra performance/pressure liquid chromatography with mass spectrometric detection (UPLC-MS/MS).[13] Aside from their sensitivity, all of the preceding approaches have the disadvantages of expensive instrumentation, long experimental times, and the need for expertise personnel. On the other hand, potentiometric ion-selective electrodes (ISEs) introduce fast, low-cost, portable, sensitive and selective alternatives to such laborious techniques.[14] Accordingly, the fabrication of a BPS-selective sensor would provide a reliable tool that could replace such techniques in the sensitive and selective determination of BPS in plastics.[15,16]

† To whom correspondence should be addressed.
E-mail: habdrabboh@kku.edu.sa; hasalah@hotmail.com

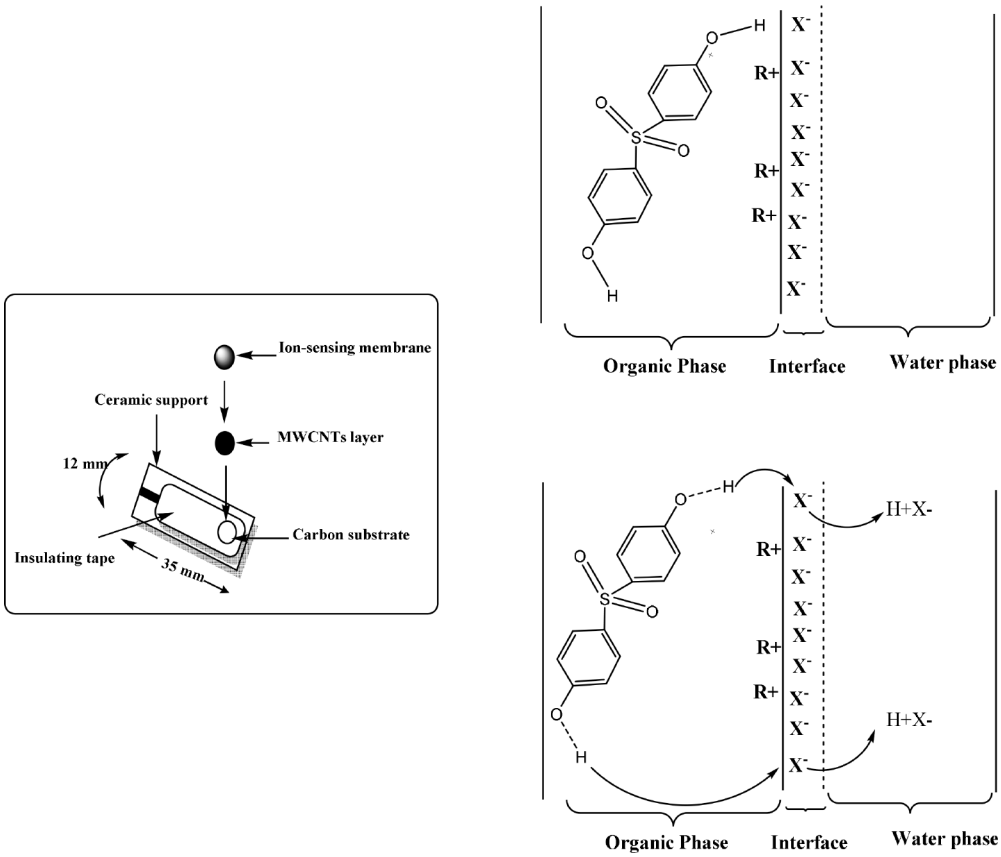

Fig. 1 Schematic representation of the proposed sensor and its operation mechanism.

Recent approaches in the field of ISEs revealed a tremendous enhancement in the sensors' detection limit, lowering it down to the sub-nanomolar level of concentration.[17] Newly developed ISEs are being used extensively in several fields of application, such as environmental monitoring, pharmaceutical and medicinal analysis, biosensing, as well as in food and water industries.[18-23] Fortunately, only one recent publication had reported an all-solid-state BPS potentiometric sensor based on the known molecular imprinting approach.[24]

In the present work, we introduce a novel, all-solid-state potentiometric ion-selective electrodes (ISEs) for the sensitive and selective detection of BPS in commercial samples. The sensing element, Aliquat 336S-BPS ion association complex, was formulated within a PVC-based polymeric membrane for the determination of BPS. A thin layer of the polymeric membrane was applied over a film of multi-walled carbon nanotubes (MWCNTs), which was *in situ* formed on the conductive solid-contact carbonized screen-printed platform. It is thus the first time to introduce a screen-printed ISE with ion-to-electron transducer for BPS detection.

Experimental

Reagents and chemicals

The reagents and chemicals used all over this work were of analytical grade, and solutions were prepared using deionized water. Tricaprylmethylammonium chloride (Aliquat 336S), tetradodecylammonium tetrakis(4-chlorophenyl) borate (ETH 500), multi-walled carbon nanotubes (MWCNTs), *o*-nitrophenyloctyl ether (*o*-NPOE) and tetrahydrofuran (THF) were products of Sigma-Aldrich (www.sigmaaldrich.com). Poly(vinyl chloride) with high molecular weight (PVC) was a product of Fluka (www.sigmaaldrich.com).

BPS, in its anionic form, prepared in a 30 mM $NaHCO_3$/Na_2CO_3 buffer solution of pH = 10 were used for a 0.01 M stock solution. Working BPS standard solutions covering the concentration range 10^{-3} – 10^{-8} M were daily prepared in the $NaHCO_3$/Na_2CO_3 buffer solution.

Apparatus

A measuring cell was constructed, consisting of screen-printed carbon electrode as a working electrode and a reference electrode (Ag/AgCl/Cl^-) (Model 9002, Thermo-Orion, USA) with 1 M CH_3COOLi (LiOAc) as an outer filling solution. Potential and pH measurements were recorded by the aid of a Thermo-Orion pH/mV meter (Model SA 720, Thermo Scientific, USA) at 25 ± 1°C, equipped with a Ross Sure-Flow combination pH electrode (Model 8172BNWP, Thermo-Orion, USA). Both chronopotentiometric and electrochemical impedance spectroscopy (EIS) experiments were carried out on an Autolab potentiostat/galvanostat from Metrohom (Model 204, Metrohom Instruments, Switzerland), supplied with a 3-electrode conventional cell containing a Ag/AgCl/3 M Cl^-/1 M AcO^- reference electrode, an auxiliary electrode made of platinum and a working electrode, which is the proposed BPS sensor.

Electrode potential-determining phase

Aliquat-BPS ion association complex was prepared by mixing 50 mL of an Aliquat 336S solution in chloroform (6% w/v) with 20 mL of a 10^{-2} M BPS solution (adjusted to pH = 10 using NaOH solution) in a separating funnel. The mixture was shaken

for 10 min and the aqueous phase was discarded. The collected organic phase was de-hydrated by passing through an anhydrous Na_2SO_4 column, and the solvent was then left to evaporate at ambient temperature in order to collect the dry Aliquat-BPS complex.

Sensor fabrication

Screen-printed carbon electrodes were products of Gwent Ltd. (Lancster, UK), where the electronic conductor is a carbon disc with a 2-mm diameter (Fig. 1). The BPS sensors were fabricated by initial casting of the solid contact conducting substrate, followed by screen printing of a polymeric membrane film.[25,26] The conducting disc on the screen-printed electrode (SPE) was homogeneously covered with a mixture of 10 mg ETH 500/2 mg MWCNTs. The electrode is then heated under an infrared radiation for 10 s to melt the ETH 500 in the mixture completely, then cooled down to ensure strong adherence on the SPE surface. The ion-sensing membrane (ISM) is prepared by mixing 100 mg of membrane components (2 mg Aliquat-BPS, 49 mg *o*-NPOE and 49 mg PVC) then dissolving the mixture in 2 mL THF. Then, 10 µL of the polymeric membrane cocktail was spread on the surface of the electronic conductor and solvent was allowed to dry at ambient temperature for 2 h. Electrodes without MWCNTs were prepared by the same previously mentioned steps for comparison. Sensors were pre-conditioned in a 1.0×10^{-4} M BPS solution, pH 10, and stored in the same solution between measurements and when not used.

Electrochemical and selectivity measurements

All solid-contact BPS ISEs were calibrated in 10^{-2} M $NaHCO_3/Na_2CO_3$ buffer solution, pH 10.0. An indicator electrode, together with the reference electrode (Ag/AgCl/Cl^-/AcO^-), were doped in 9 mL buffer solution contained in a 50 mL glass beaker. Solution was stirred, and aliquots (1.0 mL) of standard BPS solutions (1.0×10^{-8} to 1.0×10^{-4} M) were sequentially added every 3 min. Potential readings were recorded every 3 min and calibration graphs were plotted between log[BPS] and the potential readings. Selectivity coefficients for BPS sensors were calculated using the modified separate solution method, where BPS sensors were calibrated separately in each interfering ion.[27]

BPS assessment in baby bottles

Proposed BPS-based potentiometric sensors were used to determine the leached BPS levels from commercial plastic baby bottles. Different BPA-free baby bottles were collected from the local market and cut into small pieces (5 mm diameter), placed in 50 mL de-oxygenated water and heated under reflux at for 24 h at 100°C to leach BPS. Resultant solution with BPS extract was assayed using the standard addition method by the proposed ISEs. For accuracy illustration, the comparison between the proposed sensor and the high-performance liquid chromatography (HPLC) method was performed. In brief, the mobile phase (20% acetonitrile and 80% ultrapure water) was injected into the HPLC column C-18 (LiChrospher) with a 1.0 mL/min flow rate by the aid of an auto sampler. The components were exposed to UV/vis light as they exit the HPLC column, and the absorbance of the separated components was measured at 234 and 258 nm by a spectrophotometric detector.

Results and Discussion

ISEs characteristics

Screen-printed carbon electrodes (3 cm long and 10 mm width) containing carbon orifice with an area of 3.1-mm^2 covered with MWCNTs layer acting as an ion-to-electron transducer, and a thin ion-selective polymeric membrane containing Aliquat 336S as the electroactive material were prepared and characterized as solid-contact ISEs responsive to BPS in its anionic form. BPS is known as a weak acid with a $pK_a = 8.2$ and is readily in its anionic form in aqueous solutions at pHs > 8.2. So, the BPS anion can thus be detected by the proposed ISE in 30 mM $NaHCO_3/Na_2CO_3$ buffer solution of pH 10. The ion-sensing membranes containing 2 wt% aliquat-BPS, 49 wt% PVC and 49 wt% *o*-NPOE revealed the best short and stable response for BPS. Figure 2 shows calibration plot for the proposed sensor, and Table 1 summarizes its performance characteristics calculated after 12 calibration experiments, each in triplicate, over a time span of 6 weeks. The sensor revealed a Nernstian response with an anionic slope of –28.2 ± 0.8 mV/decade, a detection limit of 0.02 µg/mL, and a linear range of 2.5×10^{-7} - 1.0×10^{-3} M ($r^2 = -0.9994$). Developed BPS sensors had achieved their steady-state potential equilibrium in fast response times, <10 s. Negligible drift in the potential signals of the sensors were noticeable within the measurement period of 6 weeks, indicating the sensors' long-term stability and long life-time.

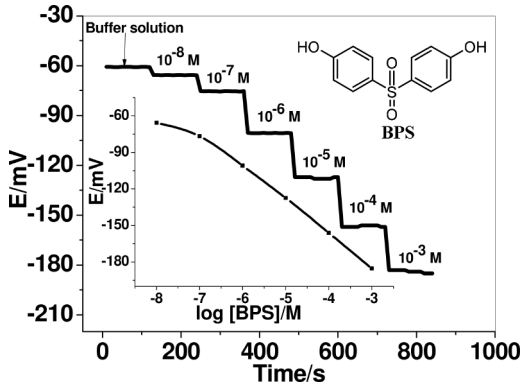

Fig. 2 Time traces and calibration curves of BPS/MWCNTs-ISE.

Table 1 Performance characteristics of BPS/MWCNTs-ISE

Parameter[a]	BPS/MWCNTs-ISE
Slope/mV decade^{-1}	–28.2 ± 0.8
Correlation coefficient, r^2	–0.9994
Lower detection limit/µg mL^{-1}	0.02 (8×10^{-8} M)
Linear range/M	2.5×10^{-7} - 1.0×10^{-3}
Working acidity range/pH	10
Response time/s	<10
Life span/week	6
Precision, %	1.7
Accuracy, %	98.8
Standard deviation, σ_{mV}	0.6

a. Mean of 6 measurements.

The detection limit (LOD) of BPS was calculated from the intersection of extrapolated linear segments of the calibration graph according to IUPAC guidelines, and were found to be 8.0×10^{-8} M (0.02 µg/mL) PBS^- with a linear range of 2.5×10^{-7} - 1.0×10^{-3} M ($r^2 = -0.9994$).

The precision and accuracy of the proposed method were examined by using 6 replicate measurements of 5 µg/mL BPS

Table 2 Selectivity values for BPS/MWCNTs-ISE

Interfering ion, j	Log $K^{Pot}_{BPS,J}$ [a]
BPA	−0.8 ± 0.09
TBBP	−4.9 ± 0.5
1-Naphthol	−3.6 ± 0.2
2-Nitrophenol	−2.5 ± 0.6
DMP	−5.2 ± 0.1
NO_3^-	−3.5 ± 0.2
Cl^-	−4.4 ± 0.3
PO_4^{3-}	−6.3 ± 0.9
F^-	−5.6 ± 0.3
Citrate	−5.1 ± 0.2
Tartrate	−5.5 ± 0.9
$HCOO^-$	−5.4 ± 0.7

a. Mean of three measurements.

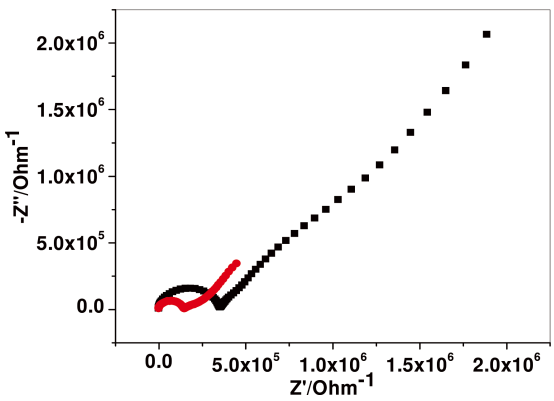

Fig. 3 Impedance spectra for (●) BPS/MWCNTs-ISE and (■) BPS-ISE recorded in a 1.0×10^{-4} M BPS solution.

as an internal quality control sample. Method precision (relative standard deviation, RSD) and accuracy were calculated using:

$$\text{Accuracy, \%} = (x/\mu) \times 100, \quad (1)$$

$$\text{Precision (RSD), \%} = (S/x) \times 100, \quad (2)$$

where x, μ, and S are the average measured analyte concentration, the reference concentration, and standard deviation, respectively (Table 1).

The closeness of the agreement between mutually independent repetitive test results obtained with 5 µg mL^{-1} internal quality control BPS sample was measured using the proposed sensor during short intervals of time within one working day (within day reproducibility), significantly small variation (±2.2%) from the final mV readings was noticed with a relative standard deviation (RSD) 1.7%. The reproducibility of the results (day-to-day response variations) was also tested by measuring a 5 µg/mL internal quality control DMA sample over 5 consecutive days using different batches of the reagents and being daily recalibrated. A small variation of the results compared to those obtained for repeatability experiments was obtained. The RSD was found to be 2.3%. These data indicate a good response stability of the proposed MIP based membrane sensor. With all sensors examined, the detection limits, response times, linear ranges and calibration slopes were reproducible over a period of at least 8 weeks.

Selectivity behavior of BPS membrane based sensor

The selectivity of BPS/MWCNTs-ISE over other phenols and other anions was determined through calculating the values of the selectivity coefficients (log $K^{pot}_{BPS,J}$) using the modified separate solution method (MSSM).[27] The obtained results, summarized in Table 2, showed that BPS/MWCNTs-ISE revealed the highest selectivity towards BPS compared to other common interfering anions such as NO_3^-, Cl^-, PO_4^{3-}, F^-, citrate, tartrate and $HCOO^-$. The selectivity behavior of the BPS sensors against other interfering phenols (bisphenol A (BPA), 1-naphthol, 2-nitrophenol, 2,6-dimethylphenol (DMP) and tetrabromo bisphenol A (TBBP)) was also tested. At pH = 10, all of the examined phenolic compounds were present in their ionized forms as their pK_a values range between 7.8 – 10.5. The obtained results show that the order of selectivity for BPS/MWCNTs-ISE is: BPS (pK_a = 8.2) > BPA (pK_a = 9.6) > 1-naphthol (pK_a = 9.3) > 2-nitrophenol (pK_a = 7.2) > TBBP (pK_a = 9.4) > DMP (pK_a = 10.6). This selectivity order is affected by the acidity, as well as the lipophilicity of the examined phenolic species. It is then anticipated that the higher is the lipophilicity and the stronger is the acidity of the anionic phenolic derivative, the stronger is its potentiometric response by the BPS sensor. As a result, DMP revealed a smaller anionic response than other tested phenolic derivatives. The selectivity behavior of the other phenolic compounds can be attributed to their lipophilicities.

EIS measurements

The impedance spectra of BPS/MWCNTs-ISE and BPS-ISE (without MWCNTs), obtained under the conditions of a 10^{-4} M BPS working solution, 10 mV amplitude of excitation, and 100 kHz – 0.01 Hz frequency are shown in Fig. 3. All impedance spectra for both BPS/MWCNTs-ISE and BPS-ISE are displayed as semicircles with high-frequencies corresponding to the resistance of the bulk membrane (R_b) together with the interfacial resistance between the electronic conductor or solid contact layer and the polymeric membrane. It is clear that the obtained resistance of the BPS/MWCNTs-ISE (0.13 MΩ) looks much smaller compared to that of the BPS-ISE (0.34 MΩ). This can be explained as meaning that the solid-contact transducer, MWCNTs, facilitates the charge transfer process across the interfaces. In addition, BPS/MWCNTs-ISE showed a smaller low-frequency semicircle than the BPS-ISE. These findings indicate that the presence of MWCNTs enhanced the low-frequency capacitance (C_L), and supported ion-to-electron transduction between the solid contact and the sensing polymeric film.

Chronopotentiometry measurements

ISEs short-term potential signal stability was examined by reversal chronopotentiometry with a constant current.[28] Figure 4 represents chronopotentiograms obtained using BPS/MWCNTs-ISE and BPS-ISE. According to the $\Delta E/\Delta t$ ratio, BPS/MWCNTs-ISE showed a potential drift (1.88 µV/s) lower than that of the BPS-ISE (93.7 µV/s) under the same conditions. The double-layer capacitances of BPS/MWCNTs-ISE and BPS-ISE were calculated and their values were 531.4 ± 2.7 and 10.6 ± 0.3 µF, respectively. The obtained results indicate that when the MWCNTs layer is casted above the surface of the electronic conductor and underneath the BPS sensing film, an enhanced stability of the ISEs potential signals is obtained. This enhancement of signal stability could be a result of the high capacitance of the MWCNTs double-layer.

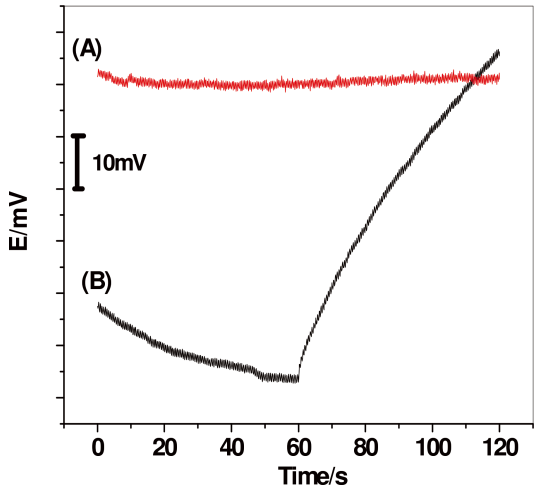

Fig. 4 Chronopotentiograms for (a) BPS/MWCNTs-ISE and (b) BPS-ISE recorded in a 1.0×10^{-4} M BPS standard solution, with ±1 nA applied current for 60 s.

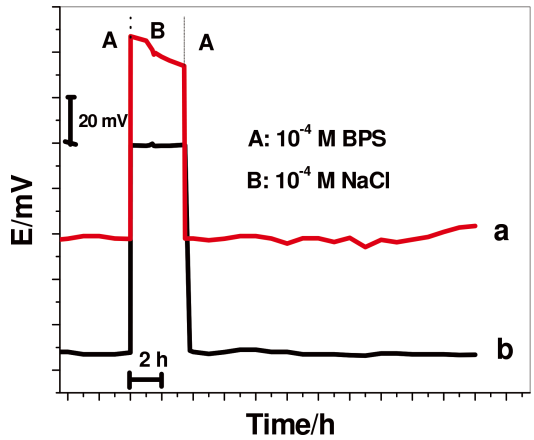

Fig. 5 Water layer tests for (a) BPS-ISE and (b) BPS/MWCNTs-ISE.

Water layer test

A long-term potential instability for the BPS/MWCNTs-ISE can be explained by the generation of a layer of water within the MWCNTs and the polymeric membrane layers. The presence of such layers can change the membrane composition because of the re-equilibration of ions across the sensing membrane.[29] In order to study this effect, a test for the presence of water layer was carried out. Figure 5 represents the potential responses of BPS/MWCNTs-ISE and BPS-ISE measured in 10^{-4} M BPS (A), then in 10^{-3} M NaCl (B), and finally in 10^{-4} M BPS (A) again. It is apparent from the figure that the BPS-ISE (without MWCNTs) acquired a negative drift within the NaCl interfering solution, and when reverting to the BPS solution a slight positive potential drift is noticed. This recorded potential drift can be a result of the formation of a layer of water within the MWCNTs and the ISE layers. On contrary, no observed drifts in the recorded potential signals were noticed for the electrode containing MWCNTs as a conducting transducer. This reflects the high lipophilicity of MWCNTs and its capability to prevent the formation of the undesirable water layer.

Table 3 Analysis of BPS in BPA-free baby bottles

Sample	BPS amount[a]/μM				Recovery, %
	BPS/ MWCNTs-ISE	HPLC	Spiked amount	Found	
Sample 1	0.23 ± 0.04	0.27 ± 0.02	0.2	0.42 ± 0.03	97.7
			0.5	0.76 ± 0.01	104.1
			1.0	1.21 ± 0.04	98.4
Sample 2	0.18 ± 0.03	0.15 ± 0.01	0.2	0.40 ± 0.06	105.3
			0.5	0.65 ± 0.02	95.6
			1.0	1.17 ± 0.07	99.1

a. Average of five measurements ± standard deviation.

Determination of BPS in commercial samples

The feasibility and applicability of the proposed ISEs in real sample analysis for BPS determination were examined. The sensors were subjected to BPS monitoring in BPA-free baby bottles. The BPS levels in the separated extract were determined by the potentiometric standard addition method, and the results were compared with the standard HPLC method (results are represented in Table 3). Data reveal that BPS recovery measurements in baby bottle samples ranged between 95.6 – 105.3%, where the means and variances are not significantly different from those obtained with the HPLC technique within a 95% confidence level.

Conclusions

Novel solid-contact ISEs for the detection of bisphenol S (BPS) were introduced for the first time in a screen-printed carbon electrodes format, where MWCNTs acted as the ion-to-electron transducer and Aliquat-BPS ion association complex was used as a sensory element to monitor BPS. The sensors revealed the following response characteristics: a wide linear range covering concentrations of 2.5×10^{-7} to 1.0×10^{-3} M; a slope of –28.2 ± 0.8 mV/decade; a linear relationship with a correlation coefficient (r^2) of –0.9994 and a low limit of detection equals 0.02 μg/mL. The proposed ISEs acquired no potential drifts with time, which could be attributed to the lipophilic MWCNTs that prevents the generation of an interfacial layer of water between it and the sensing membrane, and hence an enhanced potential signal stability. These miniaturized sensors are easy to handle and potentially cheap sensor devices for the electrochemical detection of BPS having low levels in plastic samples, with high accuracy, remarkable selectivity and announced reproducibility.

Acknowledgements

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