

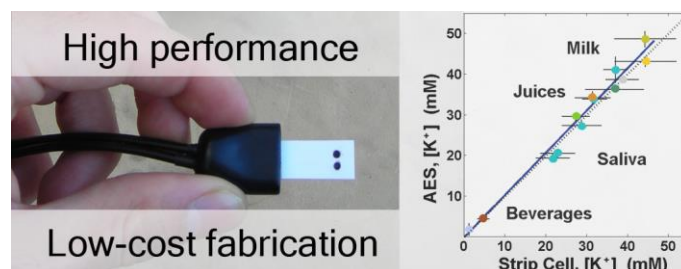
Potentiometric Strip Cell Based on Carbon Nanotubes as Transducer Layer. Towards Low-Cost Decentralized Measurements

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ABSTRACT: In this study, we developed a potentiometric planar strip cell based on single-walled carbon nanotubes that aims to exploit the attributes of solid-contact ion-selective electrodes for decentralized measurements. That is, the ion-selective and reference electrodes have been simultaneously miniaturized onto a plastic planar substrate by screen-printing and drop-casting techniques, obtaining disposable strip cells with satisfactory performance characteristics (i.e. the sensitivity is 57.4 ± 1.3 mV/dec, the response time is ≤ 30 s within the linear range from $\log a_{K^+} = -5$ to -2 , and the limit of detection is -6.5), no need of maintenance during long dry storage, quick signal stabilization and light insensitivity in short-term measurements. We also show how the new potentiometric strip cell makes it possible to perform decentralized and rapid determinations of ions in real samples, such as saliva or beverages.

TOC:



Solid-contact potentiometric ion-selective electrodes (SCISEs) have nowadays similar performance characteristics to conventional inner-solution ion-selective electrodes and offer new and advantageous technical possibilities such as miniaturization to the μm scale, cost-effective fabrication, no need for maintenance, flexibility and multiple shape configurations.^{1,2} That is very relevant from an analytical perspective because there is a high demand for in-situ chemical information (for example, for medical point-of-care testing, measuring hazardous chemicals in the environment, and monitoring the quality of processed food and beverages, among others) which could benefit from the implementation of new solid-contact potentiometric sensors.³

SCISEs have been made possible by the use of solid-contact ion-to-electron transducers such as conducting polymers (CPs)⁴ or carbon nanostructured materials,⁵ which confer the necessary stability to the potentiometric signal.⁶ In particular, single-walled carbon nanotubes (SWCNTs) improve potentiometric stability, they do not show any response to light, and their hydrophobicity avoid the formation of a water layer between the transducer and the selective membrane layers.^{7,8} SWCNTs have been widely used in electroanalytical sensors in the last decade,⁹ and now there are even commercial SWCNT-based potentiometric sensors.¹⁰ Advances in SCISEs have also been made possible by the evolution of ion-selective membrane matrices: low detection limits and high selectivity are now possible thanks to polyacrylic matrices with low ionic diffusion coefficients ($\sim 10^{-11}$ cm²/s) and an optimum control of the membrane ion fluxes.¹¹⁻¹⁴ SWCNTs and polyacrylic membranes have been applied not only to SCISEs, but also to solid-contact reference electrodes (SCREs).^{15, 16}

Screen-printing is a well-established but unique technique for the fabrication of miniaturized solid-contact sensors because of its versatility, reproducibility and cost-effectiveness.¹⁷ The first screen-printed SCISEs were reported in the late 1990s,^{18,19} and since then screen-printed potentiometric cells have been reported mainly for clinical²⁰⁻²² and environmental applications.²³⁻²⁵ Lately, CPs have been used in screen-printed electrodes.²⁶⁻³⁰ However, the hydrophilicity, light or redox sensitivity shown by commonly used Ag/AgCl and some CP solid-contact transducers is a drawback for decentralized applications. Recently, we reported on the use of SWCNTs that had been functionalized with the octadecylamine group (SWCNT-ODA) as an easy to deposit, hydrophobic, redox and light insensitive transducer for disposable electrodes.¹⁶ Therefore, SWCNT-ODA-based disposable sensors could greatly benefit from the unique properties of SWCNT-ODA, especially in decentralized application.

Here, we apply SWCNT-ODA as a solid-contact ion-to-electron transducer and polyacrylic membrane in both the SCISE and SCRE to fabricate a disposable potentiometric strip cell at a low cost and with satisfactory performance characteristics. In this way, the new disposable device integrates a well-tested SCRE,¹⁶ polymeric membrane-based SCISE with excellent performance,¹⁴ and SWCNT-ODA as an appropriate transducer layer for decentralized measurements.¹⁶ In order to test the applicability of the new potentiometric cell we determined K⁺ concentration in saliva samples and various beverages. There is a high correlation coefficient between the K⁺ activity present in serum and saliva, so determining K⁺ in saliva can be very valuable for a quick and decentralized assessment of hypertension, renal failure, cardiac distress, disorientation and dehydration.^{31, 32} A disposable potentiometric strip can also be used for monitoring the quality of beverages throughout the production or distribution process. This could increase process efficiency and eventually save large amounts of money.

EXPERIMENTAL SECTION

Reagents. The salts KCl, NaCl, LiCl, NH₄Cl, potassium tetrakis(3,5 bis(trifluoromethyl)phenyl)borate (KTFPB), tetradodecylammonium tetrakis(4-chlorophenyl)borate (ETH500), ionophore

valinomycin, tetrahydrofuran (THF), butyl acrylate (nBA) monomer, cross-linker 1,6-hexanediol diacrylate (HDDA) and photoinitiator 2,2-dimethoxy-2-phenylacetophenone (DMPP) were purchased in puriss p.a. or Selectophore grade from Fluka. MgCl_2 , CaCl_2 , NaH_2PO_4 , Na_2HPO_4 , 2-(*N*-morpholino)ethanesulfonic acid (MES), $\text{K}_3\text{Fe}(\text{CN})_6$ and $\text{K}_4\text{Fe}(\text{CN})_6$ were purchased in analytical grade from Sigma-Aldrich. SWCNTs with >90 % wt. purity were purchased from HeJi Inc. To solubilize the SWCNTs in THF, the long-chain molecule octadecylamine (ODA) was grafted to the SWCNTs to produce SWCNT-ODA.¹⁶ All the solutions were prepared with Milli-Q water (18.2 M Ω ·cm).

Disposable planar electrode development. Screen-printed electrodes were fabricated using a DEK-248 (DEK International) screen-printer. The ink (7102 conductor paste based on carbon and graphite) was provided by DuPont Limited and directly screen-printed onto polyethylene terephthalate (PET) Melinex ST726 film substrate (175 μm thick) provided by Thyssenkrupp Plastic Iberica. The polyurethane squeegee used was provided by DEK International (model SQA152 with a 45° contact angle and a hardness factor of 70). The ink was cured in a Digiheat 150L oven (JP Selecta S.A.) at 120 °C for 10 min. A PET tape coated with acrylic adhesive on one side (Arcare 8565, Adhesives Research Inc) was used to protect the conducting paths. SWCNT-ODA was dissolved in THF (1 mg/mL), and 25 μL (25 drops of 1 μL) was deposited by drop-casting onto each carbon ink sensing area (2.5 mm²). After each drop was drop-casted we let it dry for one minute. The SWCNT-ODA layer was left to dry overnight at room temperature. The K⁺-selective membrane contained 9 mg valinomycin (18.1 mmol/Kg), 4.5 mg KTFPB (11.2 mmol/kg), 4.5 mg ETH500 (8.8 mmol/Kg), 0.6 mg HDDA and 7 mg DMPP in 500 μL nBA monomer. The polyacrylic reference membrane cocktail based on photocured poly(*n*-butylacrylate) (poly(nBA)) was prepared as described elsewhere.¹⁶ One μL of each membrane was drop-casted onto the SWCNT-ODA layer and the membranes were photo-polymerized using a UV lamp (4W) for 5 min under argon flow. Figure 1 depicts the different layers deposited on the planar electrode and the final design of the potentiometric strip cell. The SWCNT-ODA layer is ~ 30 μm thick and the K⁺SCISE and SCRE membrane layers are ~ 240 and ~ 270 μm thick, respectively, as observed by the electronic microscope. The sensing area is 5 mm x 2 mm, comprising the ion-selective and reference membranes area and the area between them. After the potassium SCISE (K⁺SCISE) has been fabricated, an initial membrane conditioning of 3 h in 10⁻³ M KCl is required to avoid super-Nernstian responses and it must subsequently be placed for 30 min in various fresh 10⁻⁸ M KCl to achieve low limits of detection.

Potentiometric measurements. Electromotive forces (emf) were measured at room temperature (22 ± 2 °C) with an EM16 Lawson Labs, Inc. high-input (10¹⁵ Ω) impedance potentiometer and 2-way or 8-way edge connectors. Potentiometric measurements were performed by immersing the electrodes in stirred solutions, using a different beaker per calibration or sample solution. When indicated, experiments were recorded against a double-junction Ag/AgCl/KCl (3 M) RE (type 6.0729.100, Metrohm AG) containing a 1 M LiOAc electrolyte bridge. In this case, emf values were corrected for liquid-junction potentials using the Henderson equation. Activity coefficients were calculated by the Debye-Hückel approximation. The

potentiometric performance characteristics were calculated following the IUPAC criteria.^{33, 34} All experiments were performed with at least three electrodes.

Electrochemical impedance spectroscopy. Electrochemical impedance spectroscopy (EIS) was performed with an Autolab general purpose electrochemical system (AUTOLAB, Eco Chemie, B.V.). All electrodes under study had an area of 2.5 mm² and were connected as the working electrode in a one-compartment cell in 0.1 M KCl at room temperature (22 ± 2 °C). The RE was an Ag/AgCl/KCl (3 M) single junction (Model 6.0733.100, Metrohm), and the auxiliary electrode was a glassy carbon rod. The impedance spectra were recorded in the frequency range 100 kHz-10 mHz at open circuit potential and an amplitude of 100 mV.

Saliva and beverages measurements. Saliva samples were collected on different days and stored using a standard method.³⁵ Non-alcoholic beverages, fruit juices, baby food, soy milk and cow milk were bought from a local shop. Before any measurements were taken, saliva samples and various natural beverages were diluted (1:100) in 1.7 mM phosphate buffer solution (PBS, pH=7.2) or 50 mM MES with 0.01 M NaCl (pH=5). Buffer solutions were used to equalize the ionic strength and pH of standard samples with real samples. When high ionic strength buffers were used, calculated activities were approximated to concentration values. We used potentiometric strip cells which had been conditioned 3h in 10⁻³ M KCl and stored in dry conditions. No additional conditioning was used before saliva and beverages measurements. Atomic emission spectrometry (AES) (Unicam Solaar 969) was used to generate alternative reference values to which the potentiometric measurements were compared. We used a calibration plot generated with the measurement of 5 standard solutions to calculate the K⁺ concentration of the tested samples by AES.

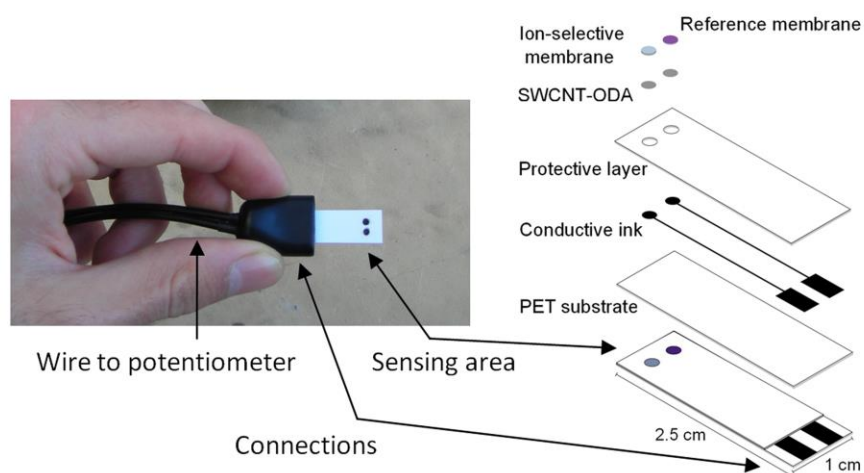


Figure 1. Photograph, scheme of the different layers, and final design (bottom) of the potentiometric strip cell based on SWCNT-ODA and polyacrylic membranes.

RESULTS AND DISCUSSION

The new disposable potentiometric strip cell consists of a SWCNT-ODA-based K⁺SCISE and a recently reported SWCNT-ODA-based SCRE.¹⁶ The same materials and fabrication procedure has been used for both the K⁺SCISE and SCRE (see Figure 1). Thus, the strip cell have been batch produced in the

laboratory at a low cost (see Table S1 in Supporting Information) and can easily be transferred to a high throughput, highly parallel and mass manufacturing process, which would reduce costs and increase reproducibility. In this technical note, we report first the analytical performance of the new disposable SWCNT-ODA-based K^+ SCISE, and then of the complete SWCNT-ODA-based potentiometric strip cell.

Disposable K^+ SCISE characterization. Figure 2 depicts the potentiometric response of the disposable SWCNT-ODA-based K^+ SCISE against a commercial double-junction RE. K^+ SCISE showed a near to Nernstian sensitivity (57.2 ± 1.2 mV/decade a_{K^+}) in the linear range from $\log a_{K^+} = -6$ to -2 , and a limit of detection (LOD) of -6.6 . The inset in Figure 2 shows the K^+ SCISE short-term stability of 0.3 mV/min at 10^{-4} M KCl without spurious noise, and the fast response time ($t_{0.3\text{mV}/\text{min}} \leq 10$ s) within $\log a_{K^+} = -5$ to -2 . The medium-term stability of SWCNT-ODA based K^+ SCISEs over 24 h in 10^{-3} M KCl was 0.19 ± 0.05 mV/h ($n=3$) (see Figure S2 in Supporting Information). The selectivity coefficients ($\log K_{KJ}^{\text{pot}}$) calculated using the experimental slope obtained by the separate solution method³⁶ for activities between 10^{-3} and 10^{-2} M ($n = 4$) are $\log K_{KLi}^{\text{pot}} = -5.9 \pm 0.1$, $\log K_{KNa}^{\text{pot}} = -5.0 \pm 0.1$, $\log K_{KNH_4}^{\text{pot}} = -2.0 \pm 0.1$, $\log K_{KMg}^{\text{pot}} = -6.7 \pm 0.1$, and $\log K_{KCa}^{\text{pot}} = -5.7 \pm 0.1$ (additional data is available in Table S2 in Supporting Information). These analytical performance characteristics are comparable to the best ones reported in the literature for rod-type macro K^+ SCISE fabricated either with carboxylated SWCNTs or CPs and polyacrylate membranes.^{7, 13, 14} This shows that the materials and fabrication techniques used here can produce miniaturized low-cost K^+ SCISE with the highest achievable performance.

Screen-printing and drop-casting techniques produced K^+ SCISEs with close emf values (standard deviation = 9.2 mV at 10^{-4} M KCl, see Figure 2) in nine different K^+ SCISEs (fabricated in three batches of three K^+ SCISEs each, fabricated over nine months). Potential differences are possibly due to variations in the SWCNT-ODA and membrane deposition, so an automated fabrication process would decrease the emf differences.³⁰

After the initial conditioning, the K^+ SCISE were stored in dry conditions. On contact with 10^{-4} M KCl solution the potentiometric signal stabilized in 3 min, after which a stability of 0.14 mV/min was observed over the first 30 min (see Figure S3 in the Supporting Information).

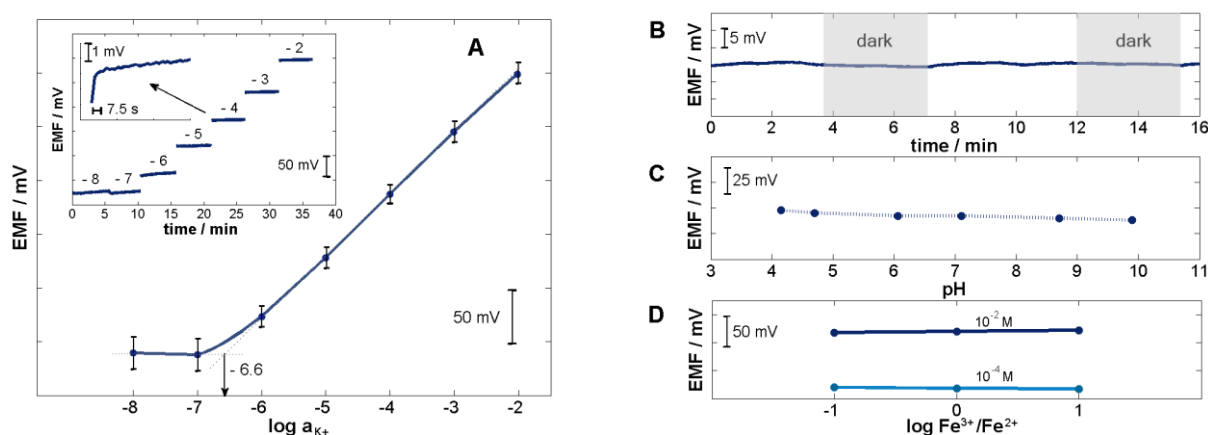


Figure 2. (A) Characterization of K^+ SCISE analytical performance: emf dependence on $\log a_{K^+}$ and time (inset) for disposable planar K^+ SCISEs based on SWCNT-ODA against a commercial double-junction RE. The error bars

correspond to the calculated standard deviation for each point ($n = 9$); (B) Light sensitivity (dark area shows the change in emf when the potentiometric cell is covered with an opaque box), (C) pH sensitivity on the addition of HCl or NaOH, and (D) 10^{-4} and 10^{-2} M $K_3Fe(CN)_6/K_4Fe(CN)_6$ redox species sensitivity in 10^{-2} M KCl.

Chronopotentiometry tests confirmed that the stability of the K^+ SCISEs was due to the presence of the SWCNT-ODA layer: when currents of ± 1 nA were applied, SWCNT-ODA based K^+ SCISE showed a 0.13 mV/s drift, whereas coated-wire type electrodes (i.e. membrane deposited onto the conductive ink) showed a 7.6 mV/s drift (see Figure S4 in Supporting Information). The results of electrochemical impedance spectroscopy (EIS) also showed a clear difference between SWCNT-ODA based K^+ SCISEs and coated wire-type K^+ ISEs. The Nyquist plot for SWCNT-ODA based K^+ SCISEs showed a high frequency depressed semicircle assigned to the bulk membrane resistance ($R_b \approx 12.5$ M Ω) in parallel to the geometric capacitance ($G_g \approx 4.6$ pF, calculated using the Warburg equivalent circuit ¹⁶), and a medium-to-low frequency diffusion line assigned to the diffusion of ions in the bulk membrane (Figure 3 inset). The coated wire-type electrode showed a high frequency depressed semicircle ($R_b \approx 70$ M Ω) followed by a low frequency semicircle produced by a deficient ion-to-electron signal transduction in the membrane/conductive ink interface (Figure 3).³⁷ EIS and chronopotentiometric tests show that SWCNT-ODA needs to be used to obtain a good solid contact and stable potentiometric readings in disposable planar electrodes. Note that the membrane resistance in the SWCNT-ODA based K^+ SCISE is significantly lower than in the coated-type electrodes. This may be due to the partial dispersion of SWCNT-ODA in the as-deposited liquid poly(nBA), then forming a very well established contact between the membrane and the SWCNT-ODA layer.

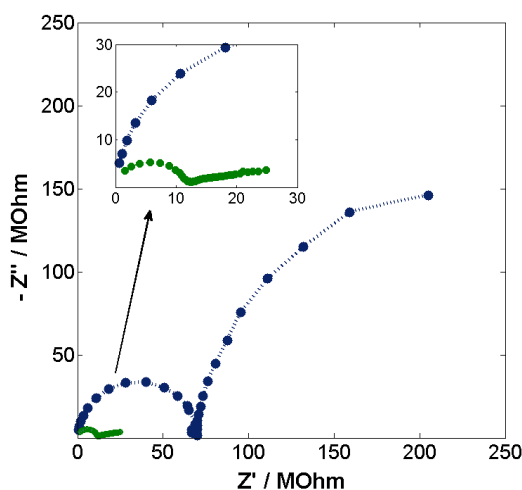


Figure 3. Nyquist plots recorded for SWCNT-ODA based K^+ SCISE (green, inset shows detail) and coated-wire type K^+ SCISE (blue) in 0.1 M KCl solution.

Potentiometric strip cell characterization. We joined the disposable K^+ SCISE and SCRE to obtain a disposable potentiometric strip cell (Figure 1) based on SWCNT-ODA and a poly(nBA) membrane. The disposable SCRE showed insensitivity to K^+ , Na^+ , Cl^- and other common chemical species present in clinical and beverage samples (slopes below 2 mV/dec for analyte concentrations ranging from 10^{-7} to 10^{-1} M, and a

standard deviation of the emf signal between 15 analytes of 4.3 mV).¹⁶ To achieve these results, we have recommended elsewhere that the SCREs be conditioned for 12 h in 10^{-3} M KCl.¹⁶ However, to shorten the conditioning time, here we opted to use non-conditioned SCRE, which stabilized its emf signal on contact with an aqueous solution within 3 min, showing low emf variation in different concentrations of KCl, NH_4Cl , NaNO_3 , and insensitivity to light and redox species, but sensitivity to changes in pH.¹⁶

The whole SWCNT-ODA-based disposable potentiometric strip cell showed a close to Nernstian response (57.4 ± 1.3 mV/dec), a relatively fast response time ($t_{0.3\text{mV}/\text{min}} \leq 30$ s) within the linear range from $\log a_{\text{K}^+} = -5$ to -2 , and a LOD of -6.5 . As a whole, the disposable potentiometric strip cell showed comparable performance characteristics to the disposable K^+SCISE and a commercial double-junction RE (see above). Such a disposable potentiometric strip cell would be very useful in decentralized measurements, where ambient factors such as light intensity, pH and redox species can affect performance. The SWCNT-ODA-based K^+SCISE showed no significant response to changes in ambient light (Figure 2A), a linear response of -1.5 mV/pH unit from pH 4 to 10 (Figure 2B), and ± 2 mV response to different ratios of $\text{K}_3\text{Fe}(\text{CN})/\text{K}_4\text{Fe}(\text{CN})$ in either 10^{-2} or 10^{-4} M total concentration (Figure 2C). The potential shift observed between 10^{-2} and 10^{-4} M, and the negative slope at 10^{-4} M $\text{K}_3\text{Fe}(\text{CN})/\text{K}_4\text{Fe}(\text{CN})$ is attributed to the different K^+ concentration in the solutions prepared. Only the slight positive slope (2 mV/dec) observed at 10^{-2} M is due to the redox species effect. The non-conditioned disposable SCRE shows insensitivity to light and to $\text{Fe}(\text{CN})^{3-}/\text{Fe}(\text{CN})^{4-}$ redox species (see Supporting Information in ref. 16). Variations in CO_2 and O_2 concentration in solution can also affect the signal of the potentiometric strip cell.⁶ Indeed, when CO_2 or air was bubbled into a 10^{-3} M KCl solution in which the electrodes were immersed, the potential of bare conductive ink/SWCNT-ODA electrodes negatively drifted until it reached a steady state (see Figure S5 in Supporting Information). However, K^+SCISE signal was not significantly affected. SCREs responded to a change in pH produced by an increase in CO_2 in solution, and also to air as the gas percolated across the membrane and interacted with Ag/AgCl salts and the SWCNT-ODA layer.

After 8 months of dry storage, the disposable SCRE is still insensitive to KCl.¹⁶ The disposable K^+SCISE showed a close to Nernstian response (54 ± 2 mV/dec) in the linear range from -5 to -2 $\log a_{\text{K}^+}$ and a LOD of -5.7 after 8 months of dry storage. Therefore, the potentiometric strip cell can be stored in dry conditions for prolonged periods with no significant loss of performance characteristics.

K^+ measurement in diluted saliva and beverages. The long lifetime of the SWCNT-ODA-based potentiometric strip cell in dry storage, together with its rapid stabilization time when it comes into contact with an aqueous solution and its rapid response time are performance characteristics that enable the disposable sensor to be used in quick and decentralized measurements. However, the strip cell still needs to be calibrated because of the variable standard potential of the disposable SCRE and, to a lesser extent, of the K^+SCISE . Here, we performed a one point calibration procedure for the quick and decentralized measurement of saliva and beverages. First, we calibrated a strip cell from 10^{-6} to 10^{-2} M KCl in 1.7 mM PBS (pH=7.2), and 50 mM MES with 0.01 M NaCl (pH=5) (see Figure 4 A and C). From the calibration plot

we obtained the LOD (close to 10^{-5} M for both buffer solutions), the sensitivity (51.3 and 57.6 mV/dec for pH 7 and 5, respectively), and the linear range (from 10^{-4} to 10^{-2} M KCl). These performance characteristics were extrapolated to all strip cells. Note that the analytical performance characteristics found made it possible to quantitatively determine K^+ concentration in diluted (1:100) saliva samples and most natural beverages, since K^+ concentration in these undiluted samples range between 1 and 100 mM. After dry storage (1-7 days), the strip cells were immersed in 10^{-4} M KCl standard solution. The signal stabilized in a short time (~6 min, see Figure 4 B and D). Immediately after signal stabilization, the strip cells were immersed in diluted saliva or beverage samples for 2 min. The last emf value obtained for each sample segment was standardized with the 10^{-4} M KCl emf value and used to calculate the K^+ concentration. This procedure enabled K^+ concentration to be directly measured in diluted (1:100) saliva and various beverages, requiring only simple data treatment.

We assessed the accuracy of the potentiometric strip cell method against an alternative method based on AES by analyzing the K^+ concentration in 13 samples (five saliva samples, two non-alcoholic beverages, two fruit juices, two types of milk, and two types of baby food). We were able to test K^+ concentrations in a large variety of samples with the strip cell because it incorporated a SCRE insensitive to a wide variety of species.¹⁶ Table 1 shows the values obtained with both methods. We compared the two sets of values using the joint confidence interval for the slope and the intercept of the regression line.³⁸ The regression line ($y = 1.04 \cdot x - 0.3$, see Figure 5A) was calculated taking into account the individual errors associated to each experimental point of the potentiometric strip cell and of the AES standard method (see Table 1). The errors are significantly higher for the potentiometric strip cell measurements than for AES. This is because the potentiometric measurements were performed with four strip cells twice on two different days ($n=8$) while the AES measurements were performed with the same instrument twice on two different days ($n=4$), and because the potentiometric method with one calibration point is a quick and decentralized method (also known as vanguard method³⁹) whereas AES is an alternative laboratory method. Figure 5B shows the joint confidence interval for the slope and the intercept of the regression line between the results obtained using the potentiometric strip cell and the AES alternative method. Since the theoretical point (intercept = 0 and slope = 1) falls within the experimental joint confidence interval we can conclude that there are no statistically significant differences between the two methods at a significance level of 5% throughout the interval of samples tested. These results show that SWCNT-ODA-based SCRE and SCISE integrated in a disposable strip cell is an excellent device for the measurement of complex samples in low-cost decentralized measurements.

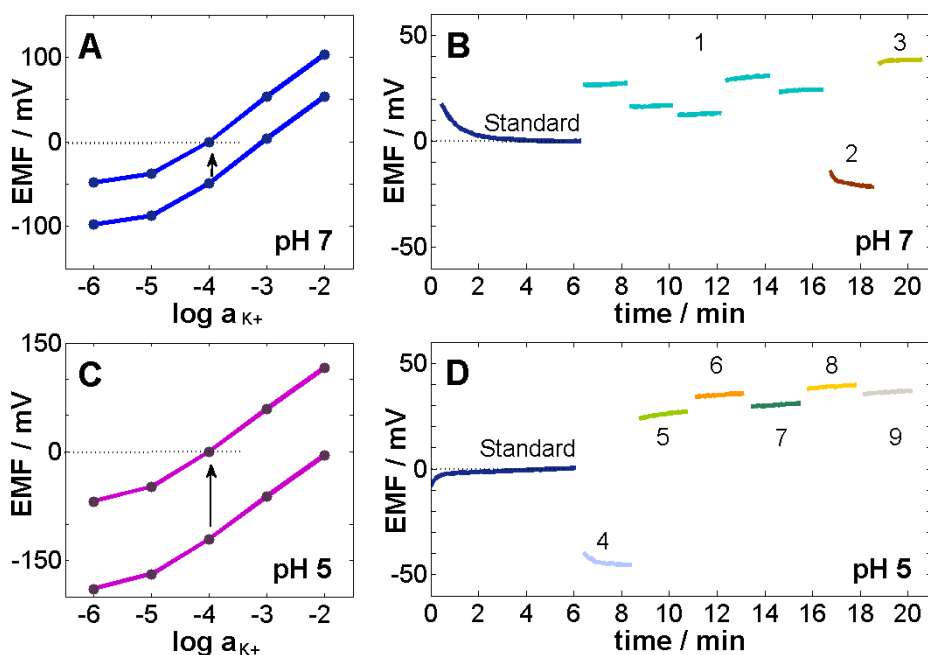


Figure 4. The graphics on the left show the original and the standardized calibration plots for K^+ in 1.7 mM PBS (top, pH 7) and 50 mM MES with 0.01 M NaCl (bottom, pH 5). The graphics on the right show emf values for K^+ measurements in 10^{-4} M KCl (standard), five saliva samples (1), Nestea (2), soy milk (3), Aquarius (4), apple juice (5), pear and pineapple juice (6), apple and pear baby food (7), apple and apricot baby food (8), and cow milk (9) using the disposable potentiometric strip cell.

Table 1. K^+ concentration measurements in saliva and beverage samples using the new potentiometric strip cell and AES. The \pm corresponds to the standard deviation of the associated results. Potentiometric measurements were repeated twice on two different days using four strip cells ($n=8$). AES measurements were repeated twice on two different days using the same instrument ($n=4$).

Sample	K^+ concentration (mM)	
	Potentiometric strip cell ^a	AES ^b
Saliva 1	31.95 ± 2.99	33.76 ± 1.05
Saliva 2	22.90 ± 4.18	20.44 ± 0.91
Saliva 3	21.77 ± 4.13	19.32 ± 1.00
Saliva 4	28.75 ± 4.82	27.32 ± 1.03
Saliva 5	37.07 ± 2.92	41.10 ± 3.22
Nestea	4.76 ± 1.45	4.34 ± 0.45
Soy milk	44.25 ± 7.53	48.62 ± 1.99
Aquarius	1.36 ± 0.13	1.75 ± 1.50
Apple juice	27.36 ± 3.38	29.53 ± 0.94
Pear and pineapple juice	31.48 ± 4.37	34.16 ± 1.52
Cow milk	38.66 ± 3.98	38.60 ± 0.64
Apple and pear baby food	36.81 ± 7.15	36.13 ± 0.47
Apple and apricot baby food	44.34 ± 7.53	43.08 ± 1.24

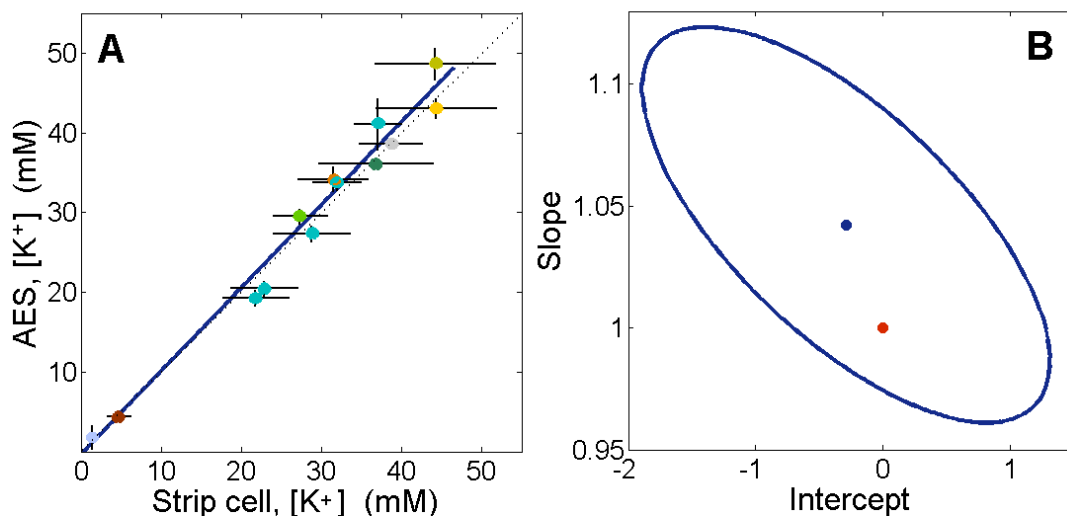


Figure 5. A) Regression line between the K⁺ concentrations (mM) in the samples listed in Table 1 determined by the potentiometric strip cell or the AES standard method. Each sample point is represented with the same color as in Figure 4. The horizontal and vertical bars associated with each experimental point are the squared standard deviations of each measurement (either the potentiometric strip cell or the AES standard method). B) Joint confidence interval for the slope and the intercept of the regression line.

CONCLUSIONS

We report on a new disposable potentiometric strip cell with SWCNT-ODA as the ion-to-electron transducer layer, and poly(nBA) matrix for both the SCISE and SCRE membranes. The fabrication procedure was designed so that it can be easily transferred to a high throughput, highly parallel and mass manufacturing process, thus reducing the costs and increasing reproducibility. In this manner, we have developed a disposable potentiometric strip cell selective for K⁺ ions which shows a close to Nernstian response (57.4 ± 1.3 mV/dec), a relatively fast response time ($t_{0.3mV/min} \leq 30$ s) within the linear range from $\log a_{K^+} = -5$ to -2 , and a LOD of -6.5 . The new disposable potentiometric strip cell can be stored for more than 8 months in dry conditions without any maintenance, and a stable emf signal and Nernstian sensitivity are attained 3 min after contact with an aqueous solution. Furthermore, the quick response time, the high selectivity for K⁺, the lack of response to interfering ambient agents such as light or redox systems and the use of only simple measuring equipment, makes this strip cell ideal for decentralized measurements. We tested the applicability of the disposable potentiometric strip for determining K⁺ in saliva samples and various beverages, and we found that results were comparable to the AES alternative method. Therefore, the new disposable potentiometric strip cell is a step towards implementing potentiometry in decentralized measuring systems, ultimately making it more accessible to general users. However, disposable solid-contact potentiometric systems still need to address a number of challenges: for example, the need for calibration or sample pretreatment, and more robust (i.e. pH insensitive) reference electrodes.

ACKNOWLEDGMENTS

We would like to thank the Spanish Ministry of Science and Innovation (MICINN) for supporting this work under project grant CTQ2010-18717. FXR-R also thanks the Universitat Rovira i Virgili for its economic support and NT Sensors S.L. for the facilities provided.

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