# Polyvinyl alcohol-cellulose composite: a taste sensing material

SARMISHTHA MAJUMDAR and BASUDAM ADHIKARI\*

Materials Science Centre, Indian Institute of Technology, Kharagpur 721 302, India

MS received 20 May 2005; revised 26 September 2005

Abstract. There are reports of fabrication of taste sensor by adsorbing lipids into Millipore filter paper. With this lipid based sensor, it has been found that the taste sensing efficiency of membrane can be remarkably improved. We have made an attempt to prepare taste sensor material by using functionalized polymer without any lipid. PVA-cellulose composite has been modified to use as the sensor material. The research work covers polymer membrane preparation, morphology study and structural characterization of the membrane and study of the taste sensing characteristics of this membrane for five different taste substances. PVA-cellulose composite membrane was modified by phosphorylation with POCl<sub>3</sub>. FTIR spectroscopic analysis, XRD analysis and SEM were done to get an idea about the structure and morphology of the prepared phosphorylated PVA-cellulose composite membrane. The sensor characteristics like temporal stability, response stability, response to different taste substances, and reproducibility of sensing performance were studied using phosphorylated PVA-cellulose composite membrane. Sensor device prepared with this membrane has shown distinct response patterns for different taste substances in terms of membrane potential. Threshold concentrations of phosphorylated PVA-cellulose composite membrane for HCl, NaCl, Q-HCl, sucrose and MSG are 0.001 mM, 0.001 mM, 0.001 mM, 0.001 mM and 0.009 mM, respectively. The threshold concentrations are below human threshold concentrations. Membranes also showed characteristic response patterns for organic acids like acetic acid, citric acid, formic acid etc, mineral acids like HCl, H<sub>2</sub>SO<sub>4</sub> and HNO<sub>3</sub> salts, bitter substances, sweet substances and umami substances. Sensor device prepared with this membrane has excellent shelf life.

Keywords. Taste sensor; taste substances; functionalized polymer.

#### 1. Introduction

Sense of taste occurs as a result of interaction between taste buds of tongue and taste substance. Different lipid molecules in taste buds of tongue are known to play the key role in sensing tastes of food materials (Miyake *et al* 1976; Kurihara *et al* 1986). Lipid based multichannel artificial taste sensor was constructed for mimicking the taste sensing ability of humans (Hayashi *et al* 1990; Toko *et al* 1990; Kikkawa *et al* 1993; Mikhelson 1994). In these taste sensors various lipids were immobilized, such as *n*-decyl alcohol, oleic acid, dioctyl phosphate etc in plasticized PVC for sensing of sourness, saltiness, bitterness, sweetness and umami.

A taste sensor (Hayashi *et al* 1990) was fabricated consisting of multichannel electrodes with transducers composed of lipid molecules immobilized within plasticized PVC matrix. Eight different types of lipids were used. Such multichannel lipid membrane device has been claimed to have the ability of sensing tastes as that done by human being. The sensor thus made showed low sensitivity to non-electrolytic taste substances compared to electrolytes. Although the sensing function of such membrane is dependent on its hydrophilicity as well as the ionic environment at the vicinity of membrane surface but such membrane surface did not have uniform hydrophilicity since the lipid molecules were dispersed in plasticized PVC. A change in charge density on the membrane surface changes the electric potential of the membrane. The surface electric charge density and the permeability of ions of taste substances to the sensing membrane are altered by the physical adsorption of non-electrolytes, which causes change of electric potential of the membrane (Iiyama et al 1986, 1989; Kurihara et al 1986; Hayashi et al 1989a). By a simple method, Hayashi et al (1995a) adsorbed lipids into the hydrophobic surface of the membrane by hydrophobic effect (Shaw 1980; Tanford 1980). They obtained a uniformly oriented lipid monolayer on the membrane surface, which provided high sensitivity to non-electrolytic taste substances. In both cases PVC was used as a membrane for entrapping lipids used as taste sensing material.

A model membrane obtained by Kurihara *et al* (Miyake *et al* 1975; Kurihara *et al* 1978) developed taste sensing with lipids extracted from bovine tongue epithelium and Millipore filter. Lipids were adsorbed into Millipore filter paper for this purpose. It was observed (Iiyama *et al* 1986; Toko *et al* 1986) that the DOPH-adsorbed membrane changed its oscillatory amplitude and frequency in

<sup>\*</sup>Author for correspondence (ba@matsc.iitkgp.ernet.in)

the presence of taste substances. In its non-oscillatory state, the effect of taste substances on its resting potential showed a good agreement with the tendency observed in biological systems regarding the order of the sensing threshold to such chemicals as quinine (bitter), NaCl (salt) and HCl (sour) (Iiyama *et al* 1987). Hayashi *et al* (1989b) investigated the effect of taste substances for a lipid membrane by casting complexes of synthetic lipid (double-chain ammonium salt) and sodium polystyrenesulphonate on a silicon wafer with a single minute pore. Reproducibility of the electric properties was improved remarkably by the use of the present construction method for the lipid membrane.

Hayashi et al (1995b) examined lipid and ion exchange cellulose for transducer materials of a taste sensor. The membrane was constructed with lipid, protein and glycocalyx. These transducers can detect all basic tastes, excepting sweet substances. Ethanol was also detected with the lipid/cellulose membrane. The self-assembled artificial membrane of poly (o-ethoxyaniline) with sulfonated lignin was satisfactorily used in the distinction of four brands of coffee with a.c. measurements (Firmino et al 2002). Chitosan was alternated with sulfonated polystyrene (PSS) to build layer-by-layer (LBL) films that are used as sensing units in electronic tongue (dos Santos et al 2003). Using impedance spectroscopy as the principal method of detection, an array using chitosan/PSS LBL film and a bare gold electrode as the sensing unit was capable of distinguishing the basic tastes (salty, sweet, bitter and sour) to a concentration below human threshold.

Riul *et al* (2002) constructed an electronic tongue composed of polyaniline oligomers (16-mer) and polypyrrole (Ppy), which were able to distinguish salt, sweet, bitter and acidic solutions. The sensor was also able to distinguish between different brands of mineral water, tea and coffee and could differentiate tastants below the human detection threshold, proving that conducting polymers are useful sensing materials and transducers for this sort of application.

Novelty of this work lies in the use of functionalized membrane to act as taste sensing material without the help of lipid molecules. Cellulose was soaked with PVA to form a composite and the membrane was phosphorylated to generate the active functional groups for sensing the basic tastes, viz. sweetness, sourness, saltiness, bitterness and umami. Taste sensing performances of this membrane are reported in this communication.

#### 2. Materials and methods

## 2.1 Materials

Polyvinyl alcohol (PVA) was obtained from Fluka AG (Switzerland). Phosphorus oxychloride (POCl<sub>3</sub>) was procured from Spectrochem Pvt Ltd., Mumbai (India). Triethylamine  $[(C_2H_5)_3N]$  was purchased from Qualigens Fine Chemicals, Mumbai (India). Tetrahydrofuran (THF) was taken from E. Merck (India) Limited. Ash less filter paper was procured from Whatman International Limited, England.

## 2.2 Membrane preparation

2.2a Soaking thin cellulose sheet with polyvinyl alcohol: Ash less filter paper, a source of cellulose was soaked in PVA solution containing 10 g PVA in 100 ml water for 24 h. After draining out the excess polymer solution, the membrane was dried in air. The dried membrane was stored in a vacuum desiccator.

2.2b *Phosphorylation of PVA soaked cellulose*: The PVA soaked cellulose was phosphorylated by refluxing with POCl<sub>3</sub> in THF at 70°C using triethylamine as catalyst. PVA soaked cellulose was phosphorylated for 1-2 h. Finally, the phosphorylated membrane was washed with distilled water and dried.

#### 2.3 Membrane characterization

2.3a Phosphorus estimation: Since the PVA-cellulose composite membrane was converted to phosphate ester by phosphorylation, phosphorus content of the membrane was estimated by Heraeus Schöniger Combustion Apparatus, Germany, following the method of Schöniger (1955). A small piece (~ 5 mg) of phosphorylated membrane was wrapped in ash less filter paper and mounted onto the platinum holder of the Schöniger combustion apparatus. About 10 ml distilled water was taken in the flask for absorption of the combustion products. Then the flask was filled with oxygen and closed with the platinum holder stopper assembly containing the sample. Through some electrical contact the filter paper along with the sample was ignited for complete combustion in oxygen atmosphere. After combustion, the flask was allowed to stand for 15 min for absorbing the gaseous products thus formed out of combustion. The platinum holder was rinsed with distilled water. The absorption solution thus obtained was heated to 80°C and titrated with 0.005 M Ce<sup>3+</sup> ion solution prepared by reducing ceric ammonium nitrate (CAN) solution with hydroxylamine. Eriochrome Black T was used as indicator with hexamine as buffer. End point was detected by change of colour from blue to red. From the titre value, wt% P in the sample was calculated using the formula:

% Phosphorus=
$$\frac{\text{Titre} \times f \times 15.49}{\text{Wt of sample}}$$
.

2.3b *Water absorption*: To perform as taste sensor, the membrane surface should be wetted by the aqueous solution of the substance whose taste is to be assessed. In

order to judge the hydrophilicity of the membrane surface, the polymer membrane was immersed in water for 24 h at 25°C and increase in weight of the membrane was recorded and the result was expressed as % water absorption.

2.3c *Moisture absorption*: Moisture absorption behaviour of the cellulose and modified cellulose membranes were assessed by exposing the membranes to laboratory environment within a range of 58-80% RH at  $30-33\cdot5^{\circ}$ C. Weight gain of the membranes was measured at an interval of 24 h up to 7 days.

2.3d *FTIR study*: For structural analysis, FTIR study of cellulose, PVA–cellulose composite and phosphorylated PVA–cellulose composite, were done using Thermo Nicolet, NEXUS 870 FTIR spectrophotometer. The study was done in absorbance mode. The polymer membranes were finely chopped and KBr pellet was prepared. The FTIR spectrum was taken in the frequency range 4000–500 cm<sup>-1</sup>.

2.3e *XRD analysis*: XRD analysis was performed for the phosphorylated PVA–cellulose composite film with PW 1710 X-ray diffractometer with Cu-target (l = 1.5418 Å) and Ni-filter between 10° and 60° (2*q*).

2.3f *Scanning electron microscopic analysis*: Study of surface morphology of cellulose, PVA–cellulose composite and phosphorylated PVA–cellulose composite was done using JEOL-JSM 5800 scanning electron microscope. The polymer films were gold coated before the study. Photographs were taken at 500 magnification.

# 2.4 Sensor set up

Figure 1(a) shows the experimental set up for the measurement of tastes of five basic taste substances, for example, NaCl for saltiness, HCl for sourness, quinine-HCl for bitterness, sucrose for sweetness and monosodium glutamate (MSG) for umami. As shown in figure 1(b) the membrane electrode device was fabricated by mounting phosphorylated PVA–cellulose composite membrane over a circular cavity on a perspex block. The cavity was filled with 100 mM KCl solution (Hayashi *et al* 1990, 1995a) through a narrow hole and one Ag/AgCl electrode was inserted into the cavity. The reference electrode device was constructed of Ag/AgCl enclosed in a glass tube filled with 100 mM KCl and 1% agar (Hayashi *et al* 1990, 1995a). The two electrode terminals were connected to a digital multimeter for measuring the electric potential across the polymer membrane.

The membrane electrode was preconditioned in 1 mM KCl solution for 30 min. The effect of change in concentration of taste solutions on the electric potential was measured. Taste substances were dissolved in 1 mM KCl solution (Hayashi *et al* 1995a). All experiments were carried out at room temperature ( $25^{\circ}$ C).

2.4a *Temporal stability*: In order to judge the time required for obtaining stable response of electric potential (temporal stability) across phosphorylated PVA-cellulose composite membrane, the membrane electrode device was dipped in 1 mM KCl solution and the potential was measured using a Ag/AgCl reference electrode immediately after dipping at an interval of 1 min and continued till stable response (Hayashi *et al* 1990).

2.4b *Response stability of membrane*: In order to judge the time required for obtaining stable response of electric potential (temporal stability) across phosphorylated PVA– cellulose composite membrane, the membrane electrode device was dipped in 1 mM KCl solution and the potential was measured using a Ag/AgCl reference electrode immediately after dipping at an interval of 1 min and continued till stable response (Hayashi *et al* 1990).



Figure 1. (a) Experimental set up for measurement and (b) membrane electrode device used in the set up.

2.4c *Changes in response with repetitive use*: The change in response in terms of electric potential of phosphorylated PVA–cellulose composite membrane to each taste substance was studied in three consecutive cycles of use. After each cycle of measurement the membrane device was rinsed with water and kept immersed in 1 mM KCl solution for 5 min prior to next cycle of measurement.

## 3. Results and discussion

#### 3.1 Membrane preparation

One of the main objectives of this investigation is to develop functionalized polymer, which will mimic lipids of the biological system of taste sensing. Lipids adsorbed in Millipore (cellulose acetate) have been found to respond to such taste substances (Miyake et al 1975; Kurihara et al 1978). In this investigation instead of using lipids we have soaked ash less filter paper, a source of cellulose with PVA and phosphorylated the polymer soaked membrane to mimic the phospholipids. Table 1 shows the physical properties of cellulose, PVA-cellulose composite and phosphorylated PVA-cellulose composite membranes. In PVA soaked membranes, polymer loading was 38%. It has been found that phosphorus content increases from 1.04-1.24% with increase in duration of phosphorylation from 1-2 h. For measurement of taste sensing in terms of membrane potential, wetting of the membrane surface by the taste solution is essential. So, water absorption study of the phosphorylated PVA-cellulose composite film was done. As shown in table 1, water absorption of cellulose (66%) increases with incorporation of PVA followed by phosphorylation. It was also found that water absorption decreases from 93.57-79.5% with increase in duration of phosphorylation from 1-2 h. This may be due to formation of phosphate esters with increase in duration of phosphorylation.

It was anticipated that the physical properties of the prepared membrane might be affected by absorption of moisture during storage. In order to know the extent of moisture absorption the membranes were kept exposed to ambient atmosphere for a period of 7 days under varying conditions of 58-80% relative humidity (RH) at 30-33.5°C. The results of moisture absorption of cellulose, PVA-cellulose composite and phosphorylated PVA-cellulose composite membranes are shown in table 2. The moisture absorption value ranges from 1.13-3.21% in case of cellulose, 1.79-4.34% in case of PVA-cellulose composite, 1.7-4.26% in case of PVA soaked and 1 h phosphorylated cellulose and 1.54-3.59% in case of PVA soaked and 2 h phosphorylated cellulose. It is also observed from table 2 that in the first 48 h the moisture absorption of the membranes was low, which increased with the increase in relative humidity to 76% in the next 72 h and again decreased with decrease in relative humidity to 62%. It is seen from table 2 that soaking with PVA increases moisture absorption of cellulose whereas the phosphorylation of PVA soaked cellulose reduces moisture absorption. Moisture absorption also decreases with increase in duration of phosphorylation from 1-2 h, i.e. with the increase in phosphorous content from 1.04-1.24%. Increase in phosphate diester linkages between two hydroxyl groups resulted in a decrease in the number of hydroxyl groups in the membrane, which decreased the moisture absorption. Thus water absorption and moisture absorption studies show sufficient hydrophilicity of the membrane for taste sensor property study in aqueous medium.

Table 1. Physical properties of cellulose (C), PVA-cellulose (PVA-C) and phosphorylated PVA-cellulose (PVA-C-P) membranes.

Polymer membrane	Phosphorylation time (h)	PVA (%)	Colour	Phosphorus (%)	Water absorption (%)	Thickness (µm)
С	0	_	White	0.0	66	180
PVA-C	0	38	White	_	PVA dissolves	200
PVA-C-P	1.0	38	Light brown	1.04	93.57	210
PVA-C-2P	2.0	38	Light brown	1.24	79.5	215

**Table 2.** Moisture absorption characteristics of cellulose (C), PVA–cellulose (PVA–C) and phosphorylated PVA–cellulose (PVA–C–P) membranes after exposure to 58–80% relative humidity at 30–33.5°C up to 7 days.

	Moisture absorption (%)*							
Polymer membrane	24 h	48 h	72 h	96 h	120 h	144 h	168 h	
C PVA-C PVA-C-P	2.72 3.98 3.68	2.62 3.52 3.53	3·21 4·34 4·26	2.9 3.53 3.36	3·21 3·72 3·71	1.13 1.79 1.70	1.98 2.58 2.56	

\*Relative humidity was about 73% up to first 48 h and later increased to 76% in the next 72 h and again decreased to 62% in the last 48 h.

# 3.2 Membrane characterization

3.2a FTIR study: FTIR analysis was done to study the change in structures of cellulose after soaking with PVA and also that of PVA-cellulose composite after phosphorylation. FTIR spectra of cellulose, PVA-cellulose composite and phosphorylated PVA-cellulose composite membranes are shown in figure 2. Assignments to FTIR peaks of cellulose membrane, PVA-cellulose composite membrane and PVA-cellulose composite membranes phosphorylated for 1 h and 2 h are shown in table 3. Cellulose membrane, PVA-cellulose composite membrane and PVAcellulose composite membranes phosphorylated for 1 h and 2 h show sharp peaks in the range 3310-3340 cm<sup>-1</sup> for hydrogen bonded -O-H stretching. Intensity of peak decreases and broadens with soaking of cellulose with PVA. This may be due to intermolecular hydrogen bonding between PVA and cellulose. After phosphorylation the peak intensity increases. This may be due to decrease in intermolecular hydrogen bonding due to phosphate ester formation. Cellulose membrane, PVA-cellulose composite membrane and PVA-cellulose composite membranes phosphorylated for 1 h and 2 h show sharp peaks for asymmetric stretching of  $-CH_2$ - in  $-CH_2OH$  of cellulose. PVA-cellulose composite membrane and PVA-cellulose composite membranes phosphorylated for 1 h and 2 h show peaks in the range 1735–1745 cm<sup>-1</sup> for > C=O stretching of acetate ester group in PVA. PVA-cellulose composite membranes phosphorylated for 1 h and 2 h show peaks at 1021 cm<sup>-1</sup> for P-O-alkyl stretching. PVAcellulose composite membranes phosphorylated for 1 h and 2 h also show peaks at 713 cm<sup>-1</sup> and 707 cm<sup>-1</sup>, respectively for P-O stretching. These observations support the formation of intermolecular phosphate diester linkage between two polymer chains.

3.2b XRD analysis: X-ray diffractograms of cellulose and phosphorylated PVA-cellulose composite membranes are shown in figure 3. It is observed that the intensity of the peaks at  $14.75^{\circ}$  (2 q) and  $22.65^{\circ}$  (2 q) in cellulose decreased after soaking with PVA followed by phosphorylation indicating a little decrease in crystallinity. The crystallinity calculated from figure 3 for cellulose and 2 h phosphorylated polyvinyl alcohol soaked cellulose were 65.5% and 59%, respectively. The crystallinity of a cast PVA membrane was found to be 24%. The decrease in crystallinity of the composite membrane might be due to the intermolecular phosphate diester linkage



**Figure 2.** FTIR spectra of cellulose, PVA–cellulose composite and phosphorylated PVA–cellulose composite membranes in the range  $3700-650 \text{ cm}^{-1}$ .



**Figure 3.** X-ray diffractograms of cellulose and phosphorylated PVA–cellulose composite membranes.

Table 3. FTIR peak assignment for cellulose (C), PVA-cellulose (PVA-C) and phosphorylated PVA-cellulose (PVA-C-P) membranes.

	v	Wavenumber (cm	-1)	
Cellulose	PVA-C	PVA-C-P	PVA-C-2P	Peak assignment
3340	3417	3410	3410	H bonded –O–H stretching
2905	2924	2911	2917	Asymmetric stretching of -CH <sub>2</sub> - in -CH <sub>2</sub> OH of cellulose
_	1745	1732	1738	> C=O stretching of ester group in PVA
_	_	1021	1021	P–O–alkyl stretching
-	_	713	707	P–O stretching



Figure 4. Scanning electron micrograph of the surface of (a) cellulose, (b) PVA-cellulose composite and (c) phosphorylated PVA-cellulose composite membranes.

between hydroxyl groups of cellulose and PVA as well as between the hydroxyl groups of PVA.

3.2c Scanning electron microscopy: Figure 4 shows the scanning electron micrograph of cellulose, PVAcellulose composite and phosphorylated PVA-cellulose composite membranes. The study of surface morphology by scanning electron microscopy revealed presence of cellulose fibre entanglements on the surface of the membranes. Figure 4(a) shows cellulose membrane before soaking with PVA and figures 4(b) and (c) show the surface morphology after PVA soaking and phosphorylation, respectively. After soaking, cellulose fibres become coated with polymer, as a result the fibres become thicker and also the fibres in the inner layers become less visible in figures 4(b) and (c). Although some diffused features are visible in the uncoated cellulose membranes due to the presence of some open fibrils, but the fibres are apparently smoothened due to coating of the open fibrils with PVA.

# 3.3 Taste sensor property study

3.3a *Temporal stability of membrane*: Temporal stability of the potential of the phosphorylated PVA–cellulose composite membrane in 1 mM KCl is shown in figure 5. The potentials were measured immediately after immersion of the membrane electrode device in 1 mM KCl solution and the membrane potential was measured at intervals of 1 min. As revealed in figure 5, PVA soaked and phosphorylated cellulose membrane shows stable response after 30 min. Based on this observation the membrane electrode device was preconditioned for 30 min in 1 mM KCl solution before measuring the response of taste solutions.

3.3b *Response stability of membrane*: In order to judge the response stability of the membrane in a particular taste solution the membrane electrode device was dipped in a taste solution of 1 mM concentration containing 1 mM KCl and the potential was measured up to 5 min at an interval of 30 s. Figure 6 shows the stability of response with time to HCl, NaCl, QHCl, sucrose and MSG for phosphorylated PVA-cellulose composite membrane. This observation indicates that the membrane is suitable for sensing tastes of different substances. Since the magnitude of response potential is different for different taste substances the membrane is able to recognize different tastes.

3.3c Responses to taste substances: The responses in terms of electric potential of phosphorylated PVA-cellulose composite membrane to five different taste substances were studied for a concentration range of 0.001– 100 mM solution and the results are shown in figure 7. Due to presence of phosphate group, membrane develops some charge. This leads to the development of some charged layers near the membrane surface. Phosphorylating the PVA soaked cellulose by POCl<sub>3</sub> converts PVA to phosphoric acid diester through which the PVA is crosslinked. A simultaneous phosphorylation of cellulosic hydroxyl groups cannot be ruled out. However, a probable structure of PVA–phosphate diester (A) is shown as:



708

From structure 'A', it is understood that the surface of the phosphorylated PVA–cellulose composite membrane would be anionic. This anionic surface of the membrane in contact with aqueous 1 mM KCl forms an electrical double layer, which is responsible for the observed potential across the membrane. On addition of taste solution, which may be either electrolytic or non-electrolytic, to this KCl alters the charge distribution on the membrane surface as well as in the bulk of the solution due to the dissociation of 'A' and the electrolytic taste substance. Such alteration in charge distribution causes the change of potential across the membrane. As a result, membrane potential decreases with increase in concentrations of HCl, NaCl, Q-HCl and MSG. Different slopes of the curves are due to different dissociation constants of the taste sub-



**Figure 5.** Temporal stability of phosphorylated PVA–cellulose composite membrane.



**Figure 6.** Stability of response potential of phosphorylated PVA-cellulose composite membrane in 1 mM taste solutions (HCl, NaCl, Q-HCl, sucrose and MSG).

stances. In case of sucrose, being non-electrolytic, there is negligible change in potential.

For phosphorylated PVA–cellulose composite membrane detection threshold values for HCl, NaCl, Q-HCl, sucrose and MSG are 0.001 mM, 0.001 mM, 0.001 mM and 0.009 mM, respectively. These threshold concentrations were below those of human threshold concentrations, which are 0.9 mM, 30 mM, 0.03 mM, 170 mM and 1.6 mM for HCl, NaCl, Q-HCl, sucrose (Pfaffmann 1959) and MSG (Yamaguchi 1967), respectively. The errors (%) defined by the standard deviations divided by the average values were 0.30, 1.52, 0.58, 1.16 and 1.43 for HCl, NaCl, Q-HCl, sucrose and MSG, respectively at 1 mM concentration in each case.

3.3d *Changes in response with repetitive use*: In order to see the reproducibility of response pattern of the membrane to a particular taste solution the membrane electrode device was tested for repeatability by three consecutive measurements of response potential in each taste solution for the concentration range 0.001–100 mM. Figure 8 shows reproducibility of response by phosphorylated PVA-cellulose composite membrane to five different taste substances. It was found that PVA soaked and phosphorylated cellulose membrane shows almost identical response patterns on repeated measurements though there is little drift of about 1 mV.

3.3e Response to organic acids, mineral acids, salts, bitter, sweet and umami substances: Although the phosphorylated PVA-cellulose composite membrane senses the tastes of saltiness, sweetness, sourness, bitterness etc but it is essential to know how this membrane behaves towards different chemical substances in an identical taste category. For this reason we have studied the res-



**Figure 7.** Responses of phosphorylated PVA–cellulose composite membrane to HCl, NaCl, sucrose, monosodium glutamate (MSG) and quinine–HCl (QHCl).



**Figure 8.** Repeatability of phosphorylated PVA–cellulose composite membrane for (a) HCl, (b) NaCl, (c) Q-HCl, (d) sucrose and (e) MSG solutions at an interval of 5 min.

ponse patterns of the prepared membrane for different compounds of each taste category. Phosphorylated PVA– cellulose composite membrane shows characteristic responses to organic acids like acetic acid, citric acid, formic acid, oxalic acid and lactic acid, respectively as shown in figure 9a. Responses of phosphorylated PVA–cellulose composite membrane to mineral acids like hydrochloric, sulphuric and nitric acids are shown in figure 9b. Responses for salts (NaCl, KCl, KBr, NaNO<sub>3</sub>, Na<sub>2</sub>SO<sub>4</sub>, NaAc, NaHCO<sub>3</sub>), bitter substances (Q-HCl, MgSO<sub>4</sub>, MgCl<sub>2</sub>), sweet substances (sucrose, glucose, fructose, glycine) and umami substances (MSG, IMP, GMP) are shown in figures 9c, 9d, 9e and 9f, respectively. The variation in slopes of the curves is due to their difference in dissociation constants. From figures 9a–f it can also be said that the prepared membrane in a suitable sensor device is able to recognize taste substances as well as differentiate among substances in a particular taste category.

# 4. Conclusions

Our main objective of this work was to develop taste sensing membrane material using tailor-made polymer. The taste sensing property of phosphorylated PVA-cellulose composite membrane thus prepared was evaluated by studying the change in membrane potential with concentration of taste substances. The membrane shows characteristic curve pattern with each of the taste substances.



Figure 9. Responses in terms of membrane potential of phosphorylated PVA-cellulose composite membrane to different (a) organic acids, (b) mineral acids, (c) salts, (d) bitter substances, (e) sweet substances and (f) umami substances.

Threshold values of membrane response for different taste substances were below human threshold values. The membranes showed good stability of response in 1 mM taste solutions. Repeatability in response is quite good with small drift in potential (1-2 mV). Phosphorylated PVA–cellulose composite membrane also shows characteristic response patterns for organic acids, mineral acids, salts, bitter substances, sweet substances and umami substances.

#### References

- dos Santos D S Jr, Riul A Jr., Malmegrim R R, Fonseca F J, Oliviera O N Jr. and Mattoso L H C 2003 *Macro. Biosci.* **3** 591
- Firmino A Jr., Antonio R, Venancio E C, Fonseca F and Mattoso L H C 2002 Development of taste sensors using natural polymers for evaluation and classification of coffees, in Natural polymers and composites IV, Proceedings from the international symposium on natural polymers and composites, Sao

Pedro, Brazil, (eds) C Mattoso et al (Sao Carlos, Brazil: Embrapa Instrumentacao Agropecuaria) p. 55

- Hayashi K, Matsuki Y, Toko K, Murata T, Yamafuji Ke and Yamafuji Ka 1989a Sens. Mater. 1 321
- Hayashi K, Yamafuji K, Toko K, Ozaki N, Yoshida T, Iiyama S and Nakashima N 1989b *Sens. Actuators* **16** 25
- Hayashi K, Yamanaka M, Toko K and Yamafugi K 1990 Sens. Actuators B Chem. 2 205
- Hayashi K, Toko K, Yamanaka M, Yoshihara H, Yamafuji K, Ikezaki H, Toukubo R and Sato K 1995a Sens. Actuators B Chem. 23 55
- Hayashi K, Wakita Y and Moriyama K 1995b Kagoshima Daigaku Kogakubu Kenkyu Hokoku **37** 27
- Iiyama S, Toko K and Yamafuji K 1986 Agric. Biol. Chem. 50 2709
- Iiyama S, Toko K and Yamafuji K 1987 *Maku (Membrane)* **12** 231(in Japanese)
- Iiyama S, Toko K, Hayashi K and Yamafuji K 1989 Agric. Biol. Chem. **53** 675
- Kikkawa Y, Toko K, Matsuno T and Yamafuji K 1993 Jpn J. Appl. Phys. **32** 5731

- Kurihara K, Kamo N and Kobatake Y 1978 Adv. Biophys. **10** 27 Kurihara K, Yashii K and Kashiyayanagi M 1086 Comp. Bio
- Kurihara K, Yoshii K and Kashiwayanagi M 1986 Comp. Biochem. Physiol. A85 1
- Mikhelson K N 1994 Sens. Actuators B Chem. 18 31
- Miyake M, Kamo N, Kurihara K and Kobatake Y 1975 J. Membr. Biol. 22 197
- Miyake M, Kamo N, Kurihara K and Kobatake Y 1976 *Biochim. Biophys. Acta* **436** 856
- Pfaffmann C 1959 *The sense of taste*, in *Handbook of physiology*, *neurophysiology* (ed.) J Field (Washington, DC: American Physiological Society), **Vol. 1**, p. 507
- Riul A Jr. et al 2002 Langmuir 18 239
- Schöniger W 1955 Mikrochim. Acta 123
- Shaw D J 1980 Introduction to colloid and surface chemistry (London: Butterworth)
- Tanford C 1980 The hydrophobic effect (New York: Wiley)
- Toko K, Tsukiji M, Iiyama S and Yamafuji K 1986 *Biophys. Chem.* **23** 201
- Toko K, Hayashi K, Yamanaka M and Yamafuji K 1990 Tech. Digest. 9th Sens. Symp. p. 193
- Yamaguchi S 1967 J. Food. Sci. 32 473