Ag/AgCl Reference Electrode Based on Thin Film of Arabic Gum Membrane

Sagir Alva[1,*], Aiman Sajidah binti Abdul Aziz[2],
Mohd Ismahadi bin Syono[3], and Wan Adil bin Wan Jamil[4]

[1]Mechanical Engineering Department, Faculty of Engineering, Universitas Mercu Buana,
Jl. Meruya Selatan No. 01, Kembangan, Jakarta-11650, Indonesia

[2]Photonics R&D NST Department, MIMOS Berhad, Technology Park Malaysia,
Kuala Lumpur 57000, Malaysia

[3]Nano, MEMS and IC Design NST Department, MIMOS Berhad, Technology Park Malaysia,
Kuala Lumpur 57000, Malaysia

[4]Product Development Department, MIMOS Berhad, Technology Park Malaysia,
Kuala Lumpur 57000, Malaysia

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ABSTRACT

The Ag/AgCl reference electrode based on the Arabic Gum (AG) hydrogel films was successfully developed. The Arabic Gum concentration used in this study was 10–40% (w/v) with an optimum concentration of 20% (w/v). In this study, reference electrode performance testing is based on ΔmV response of Cl^- ion measurements, stability tests and performance testing against ISE sensors such as K^+, NH_4^+, and NO_3^-. The Ag/AgCl based reference electrode of the Arabic Gum film shows an average ΔmV of 4.0 ± 0.7 mV when response test is performed against a standard Ag/AgCl double junction electrode in various concentrations of Cl^- ion. Meanwhile, instability testing conducted in a 0.01 M KCl solution for 72 h found drift of < 0.6 mV/h. The reference performance test of the Arabic Gum reference electrode with three types of ISE NH_4^+, $K^{+,}$ and NO_3^- commercial sensors obtaining a slope closer to the Nernst value is 54.9 ± 0.9, 52.3 ± 0.5 and -53.2 ± 0.2 mV/dec with all having a linear range of 0.1–10^{-5} M.

Keywords: *solid state reference electrode; Arabic gum; ion-selective electrode; screen printed electrode*

ABSTRAK

Elektroda referensi Ag/AgCl berbasis film hydrogel getah Arabik telah berhasil dikembangkan. Konsentrasi getah Arabik yang digunakan dalam penelitian ini adalah 10–40% (b/v) dengan konsentrasi optimum adalah 20% (b/v). Pengujian performa elektroda Ag/AgCl berbasiskan getah Arabik meliputi pengujian ΔmV berbagai konsentrasi ion Cl^-, pengujian kestabilan serta pengujian performa dengan elektroda ESI komersial seperti K^+, NH_4^+ dan NO_3^-. Elektroda referensi Ag/AgCl berbasiskan film getah Arabik memperlihatkan nilai ΔmV rata-rata ion Cl^- sebesar 4,0 ± 0,7 mV ketika diuji vs elektroda Ag/AgCl dua jembatan standar dalam berbagai konsentrasi ion Cl^-. Sementara itu pengujian kestabilan dalam larutan KCl 0,01 M selama 72 jam menghasilkan pergerakan < 0,6 mV/jam. Pengujian performa elektroda referensi Ag/AgCl berbasiskan getah Arabik dengan menggunakan tiga ESI komersial NH_4^+, K^+ dan NO_3^- menghasilkan nilai yang mendekati bilangan Nernst secara berturut-turut adalah 54,9 ± 0,9, 52,3 ± 0,5 dan -53,2 ± 0,2 mV/dekade dengan keseluruhan mempunyai rentang pengukuran 0,1–10^{-5} M.

Kata Kunci: *elektroda referensi padatan; getah Arabik; elektroda selektif ion; elektroda cetak layar*

INTRODUCTION

Research on the reference electrode in the last few years is growing quickly. This is inseparable from the advancement of electrochemical technology, measurement with an ion-selective electrode (ISE) in particular that led to the miniaturization of technology. One of the widely used reference electrodes in ISE's measurement is Ag/AgCl reference electrode. In the ISE method, the reference electrode serves to establish the potential energy of an electrochemical cell by way of comparing with a working electrode or ISE. A major feature of the reference electrode is that the potential energy reference electrode is worth a fixed, stable and not impressed by the changes in the compactness of the samples [1].

One type of reference electrode miniaturization technology of Ag/AgCl is much developed is with the screen-printed electrode technology (SPE). This technology is changing the shape of the original large-

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* Corresponding author. Tel : +62-85779381297
Email address : sagir.alva@mercubuana.ac.id

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sized electrodes such as rod and pen-shaped into planar electrodes and thin. This change automatically brings out the changing of liquid electrolyte towards a solid form, so this type of electrode is also often called the solid reference electrode. The SPE reference electrode has some advantages such as a simple shape, it can be supplied in bulk, and are suited to integrate with ISEs sensor in the single strip [2-3].

Although there are a few advantages of this SPE technology, this technology also shows some quite significant weaknesses and make the challenge to be a master in the process of making up the reference electrodes. One of the largest challenges was the limited number of electrolytes and rapid changes on the Ag/AgCl surface of the reference electrode. The Ag/AgCl layers of the reference electrode very easily to dissolve into the electrolyte solution of 3 M KCl, that will certainly lead to a reference electrode becomes unstable and had a short lifetime [4].

Therefore, researchers have previously attempted to address this problem, like using some of the hydrophobic membranes such as polyurethane (PU), poly (butyl acrylate), polyvinyl chloride (PVC) mixed with two variations charge of the lipophilic compound [2,5-6].The use of this membrane has a weakness in the control the occurrence of the balance of the charge in the membrane, often one of the salt lipophilic compounds will leach out from the membrane towards the sample. So this will make the charge imbalance in the membrane and cause the reference electrode to become unstable. Other research is to utilize a membrane such as an agarose hydrogel mixed with 3 M KCl and protected with a protective hybrid layer of PVC/Cellulose Nitrate (PVC/SN) [7]. But the presence of 3 M of KCl in the reference electrode system directly on the Ag/AgCl surface cause AgCl dissolution [4].

To overwhelm the above troubles, then demand to wait for another alternative of the fabrics. One of the other options that look potentially utilized the polymer is Arabic Gum (AG). This polymer is a natural hydrogel polymer that consists of complex polysaccharides and glycoproteins. The polymer is known as one of the natural inhibitors. One of the research ever undertaken by Abu-Dalo *et al.* in 2012 [8] reported that the presence of a layer of an AG hydrogel membrane on mild steel has managed to slow the corrosion rate of steel when immersed in acid media. The same phenomenon is also reported in the manufacture of aluminum-air batteries by using NaOH as an electrolyte [9]. Based on the phenomenon, this research was carried out to study the effect of the presence of the AG hydrogel membrane against the performance of the Ag/AgCl reference electrode SPE type.

EXPERIMENTAL SECTION

Materials

The materials used in this study is Arabic Gum membrane obtained from Elnasr Ltd., membranes of cellulose acetate (CA) obtained from Sigma-Aldrich, the organic solvent such as tetrahydrofuran (THF), ethanol, as well as salts such as KCl, KNO_3, NH_4Cl, $FeCl_3$ purchased from Merck.

Instrumentation

Measurements of Nernstian numbers from Ag/AgCl electrodes, performance with ISEs, and also ΔmV measurements of reference electrodes were performed using an ion meter with type a Thermo scientific Orion 4-star plus pH/ISE benchtop Multiparameter. Meanwhile, for measurement with SEM is done by using the JEOL-7600F FE-SEM device. While drift measurement is done by using Potentiostat/Galvanostat Electrochemical Workstation Corrtest with CS-350 model.

Procedure

The SPE of Ag obtained from Scrint Print Malaysia Berhad cleaned with a tissue paper containing ethanol. After clean, the Ag SPE electrode is immersed for 30 seconds in a solution of 0.1 M $FeCl_3$ and formed the blackish-gray layer of AgCl on top of Ag layer and cleaned the SPE with DI water and dry it. Then the SPE of Ag/AgCl was tested against the double junction Ag/AgCl reference electrode standard in the various concentrations of Cl^-.

Next on the surface of the selected of Ag/AgCl SPE coated with varying concentration of AG membrane between 10–40% (w/v) containing 3 M of KCl. After that, the AG membrane was dried for one night at room temperature. Once the dried of AG membrane form, the membrane surface is protected by the CA 5% (w/v) dissolved in THF solvent. The membrane CA is allowed to dry for one hour at room temperatures. After the CA membrane dry, the SPE electrode was tested to measure the mV changing of the Cl^- against the double junction of Ag/AgCl reference electrode standard in various concentrations of Cl^- solution within the range of 0.1–10^{-5} M to choose the optimal concentration of AG membranes.

After the optimal concentration of membranes obtained, repeat the experiment by the coating the Ag/AgCl surface of the SPE with the selected concentration of an AG membrane bare and left one night at room temperature to dry. After drying, the AG membrane surface was coated with a 5% CA (w/v) membrane and set aside to dry for 1 h. Later on, the layer of CA membrane dry, above the surface are mounted with a selected AG membrane containing 3 M of KCl and again it is gone forth to dry for overnight at room temperatures. Once dry, above its surface, mounted with CA as protective and left to dry for one hour. After the protective layer of the CA membrane was dry, the Ag/AgCl reference electrode SPE type was performed with ΔmV of Cl^- measurement, stability testing, and performance testing by using the commercial of ISE's sensors such as K^+, NH_4^+, and NO_3^- .

RESULT AND DISCUSSION

Ag/AgCl Electrode Characterization

Ag/AgCl electrode that has been constructed to prove its quality before it is mounted with the AG and the CA membrane. This is done to reduce the effect of the quality of electrode Ag/AgCl against the results of this research. Thus, the effect of the AG membrane coating against the performance of the SPE reference electrode was developed can be learned to better.

In this study, testing the quality of Ag/AgCl electrodes is carried out using two approaches, by looking at the thickness of the layer of AgCl formed above the surface of the Ag electrode. The suitable thickness of the AgCl layer is in the micrometer range (≈ 10^3 nm). The AgCl layer dissolves easily in the 3 M of KCl electrolyte solution on the reference electrode, and it makes to have a short life period [4]. In this study, the thickness of AgCl measured using SEM equipment, where the results of the AgCl layer thickness was formed around 24–34 μm, and forms the ununiform aggregate of crystals as shown Fig. 1.

In addition to looking at the thickness of AgCl are formed, testing the quality of Ag/AgCl electrodes can be done by testing the number of the Nernstian number of variations of Cl^- concentration between $0.1–10^{-4}$ M. Ag/AgCl electrodes are very sensitive to changes in Cl^- concentration in the sample. The nature of the sensitive surface of the electrode Ag/AgCl can be explained through the following mechanisms:

In electrochemistry, the surface of AgCl will be according to the following reaction:

$$AgCl(s) + e^- \rightarrow Ag(s) + Cl^-(aq) \qquad (1)$$

In chemical equilibrium and is expressed through Nernst, then the Eq. 1 can be composed as follows:

$$E = E^o - \frac{2.303RT}{nF} Log\left[\frac{a_{Ag}a_{Cl^-}}{a_{AgCl}}\right] \qquad (2)$$

where E^o is the potential energy standard, R is the ideal gas constant (8.314 J K^{-1} mol^{-1}), T is the absolute temperature (K), n is the number of moles of the displacement of electrons (n = 1), F is the Faraday numbers (96485 C mol^{-1}), and the a_x is a coefficient activity of x.

It should be mentioned, that the coefficient activity of the chemical species in the solid phase is one of integrity and sense of balance in the organization also involves a little to moderate activity coefficient of Cl^-, so that Eq. 2 can be expressed equally:

$$E = E^o - \frac{2.303RT}{nF} Log\left[\frac{|AgCl||Cl^-|}{|AgCl|}\right] \qquad (3)$$

Referable to the slightly-soluble AgCl in aqueous media, thus AgCl concentration should ideally be a constant (k) so that Eq. 3 can be composed as follows:

$$E = E^o - \frac{2.303RT}{nF} Log\left[\frac{Cl^-}{k}\right] \qquad (4)$$

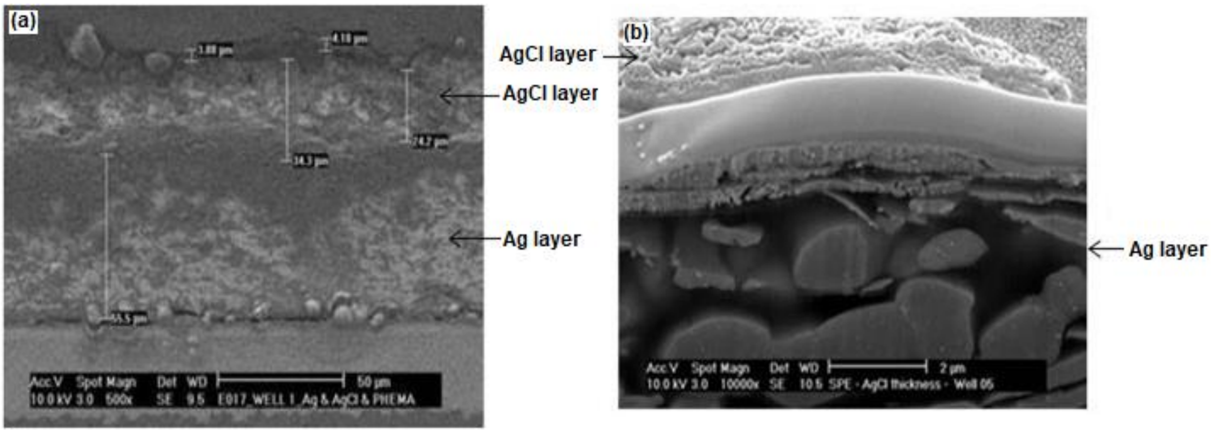

Fig 1. SEM image of the AgCl formation on top of an Ag SPE (a) the thickness of AgCl layer (b) the crystal aggregation of AgCl layer

With a substituted the T value in the temperature of 25 °C into the Eq. 4, it will be retrieved the next equation:

$$E = E^{o} - 59.6\ \text{mV Log}\left[Cl^{-}\right] + 59.16\ \text{mV Log}\left[k\right] \quad (5)$$

Since k is constant, then the Eq. 5 can be simplified into:

$$E = E^{o} - 59.16\ \text{mV Log}\left[Cl^{-}\right] \quad (6)$$

Based on Eq. 6, then any increase 10 times in the concentration of Cl^- will cause a decrease in the potential energy of 59.16 mV [10]. In general, the number obtained is amounting to Nernstian (-52)–(-49) mV/dec [4]. In this work, the Nernstian number founded closely with the Nernstian as displayed in Table 1. This demonstrates that the quality of electrode Ag/AgCl has produced already meet the banner as a transducer electrode Ag/AgCl.

ΔmV Cl^- Ion Test

The principal characteristics of the reference electrode are the potential energy of the reference electrode is unchanged despite changes in the compactness of the sample result. This is a reference to the characteristics of the electrode can be applied to evaluate the potential difference in potentiometry technique. For it is very important in this research to measure the difference potential of Ag/AgCl SPE reference electrode against the double junction of Ag/AgCl reference electrode standard in the varying concentration of the Cl^- solution [2-3].

Testing is performed by using a variation of the solution sample ions Cl^- concentrations between 0.1–10^{-5} M. Testing is done by using two kinds of SPE reference electrode. The first type is the SPE reference electrode that uses only two layers of membrane, i.e., one layer an AG membrane containing 3 M KCl and single layer membrane CA 5% (w/v) as the protector as shown Fig. 2. The purpose of protective coating caused by AG membrane which is very soluble in the water. The selection of a CA as a protective layer of the AG membrane due to the nature of the membrane CA which is mechanically very strong, are not water soluble and have enough pores to allow the occurrence of ion movement in two directions from either the reference or the electrodes from the sample solution [11].

In the first type of SPE reference electrode, membrane concentration of AG used between 10–40% (w/v) containing KCl 3 M. The selection of membrane AG in this study is based on the properties of hydrophilic membrane, so that the membrane can be present in the form of hydrogel and still be able to allow the movement of the ion in two directions [12]. In addition, some of the previous reports, the AG membrane is known as the nature anti-corrosion. The AG membrane is a complex a polysaccharide containing glycoproteins and is rich in functional groups OH that is able to withstand the movement of Cl^- to be less free, so the corrosion rate on the surface of the SPE reference electrode can be slowed [8].

Table 1. The Nersnstian number of the Ag/AgCl SPE in various concentration of Cl^- solution (0.1–10^{-4} M) (n = 3)

SPE Ag/AgCl	Slope (mV/dec)	SD	RSD (%)	R^2
1	-55.2	0.5	0.97	0.9994
2	-54.4	0.8	1.55	0.9995
3	-55.1	0.1	0.11	0.9993

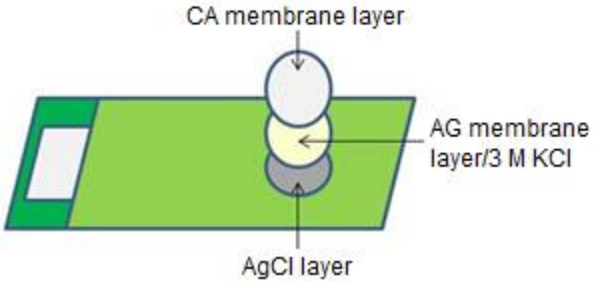

Fig 2. The first type of structure of SPE reference electrode

The problem of corrosion is one contributor declining the performance of a reference electrode Ag/AgCl through a mechanism as follows:

A layer of AgCl on the surface will be broken up with the bearing of the KCl solution in high concentrations [4].

$$AgCl \rightarrow Ag(aq) + Cl^{-}(aq)$$

The next layer of Ag electrode reaction with oxygen and carbon dioxide trapped in the membrane and the aqueous sample wherein the carbon dioxide change to carbonate acid form. The reaction occurs to form a new layer of silver oxide or silver carbonate can block the transfer of electrons in the system reference electrode [1,13].

$$4Ag(s) + O_2 \rightarrow 2Ag_2(s)$$

$$H_2O + CO_2 \rightarrow H_2CO_3$$

$$2Ag(s) + H_2CO_3 \rightarrow Ag_2CO_3(s) + H_2$$

The use of concentration variation of the hydrogel can affect the performance of the reference electrode, so the concentration of hydrogel need to be optimized to obtain the best performance [1,14]. The results of ΔmV Cl^- measurements of the SPE electrode reference first type shown in Table 2 and Fig. 3. Seen that ΔmV of Cl^- occurs at the optimum concentration of membrane AG 20% (w/v) with a value of 7.3 ± 0.8 mV. Meanwhile, at a concentration of 10% obtained grades ΔmV Cl^- 10.5 mV. This happens because of the water

Table 2. ΔmV Cl^- measurements of the SPE reference electrode the first type (one layer of AG membrane) (n = 4)

Log[Cl^-]	10% AG (mV)	20% AG (mV)	30%AG (mV)	40%AG (mV)
-1	151.7	143.7	139.5	137.9
-2	155.9	146.8	143.2	140.3
-3	162.2	151.0	149.3	149.7
-4	159.4	147.6	147.8	147.8
-5	158.9	146.9	147.7	148.0
ΔmV	10.5	7.3	9.8	11.8
SD	1.5	0.8	1.3	1.6

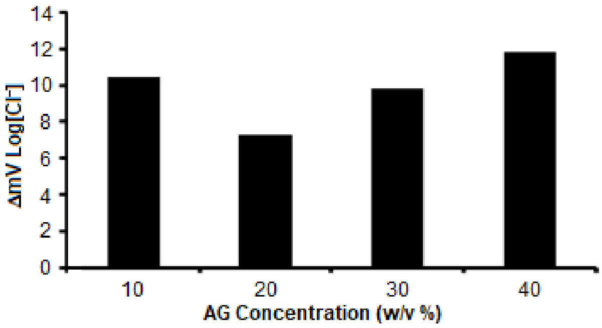

Fig 3. Graph of ΔmV Cl^- vs Ag/AgCl standard reference electrode (one layer of AG membrane)

content in the membrane concentration by AG 10% still high enough so that movement of Cl^- is still free enough and this contributed to accelerating the onset of corrosion processes on the surface of the SPE reference electrode. Meanwhile, at the concentration of AG membranes at 30 and 40% (w/v) occur rebound ΔmV Cl^- i.e., greater than 7.3 mV. This rebound occurs because of the volume of water at a concentration of 30 and 40% (w/v) of the AG membrane lower than the AG membrane with 20% concentration, so the SPE of the reference electrode dried. This circumstance makes the ion movement in two directions a little stunted.

Measurement of ΔmV Cl^- is also exercised on the SPE reference electrode type two, which consists of four layers of the membrane that is two layers of AG membrane and double layer CA membrane as shown in Fig. 4. The concentration of the AG membrane is 20% (w/v), where for the first layer of the AG membrane is a bare the second layer of AG membrane contains 3 M KCl. Meanwhile, the inner layer of the membrane CA serves as a separator between the first and second layers of AG membrane, while the outermost layers of the CA function as a protector.

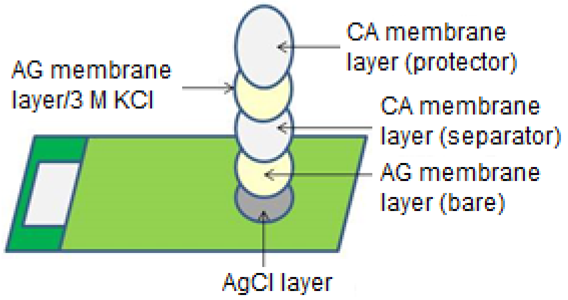

Fig 4. Structure of SPE reference electrode second type

The results of the measurements ΔmV of Cl^- the SPE reference electrode acquired for the second type of 4.0 ± 0.7 mV as shown in Table 3 and Fig. 5. The results showed the value lower than the first type. This is made possible by the existence of the first layer of the AG membrane that does not contain an electrolyte KCl 3 M and the existence of a layer separator CA membrane successfully to slow down the Cl^- a movement to quickly come to the surface of Ag/AgCl. This helps to inhibit the process of dissolving of AgCl and formation of new layers of Ag_2CO_3 and Ag_2O on the SPE reference electrode.

Table 3. Comparison of ΔmV $Cl^-$of the SPE reference electrode the first type (one layer of 20% AG membrane) and the second type (two layers of 20% AG membrane) (n = 4)

Log[Cl^-]	1 Layer AG (20%)	2 Layer AG (20%)
-1	143.7	172.9
-2	146.8	173.3
-3	151.0	176.9
-4	147.6	174.1
-5	146.9	173.1
ΔmV	7.3	4.0
SD	0.8	0.7

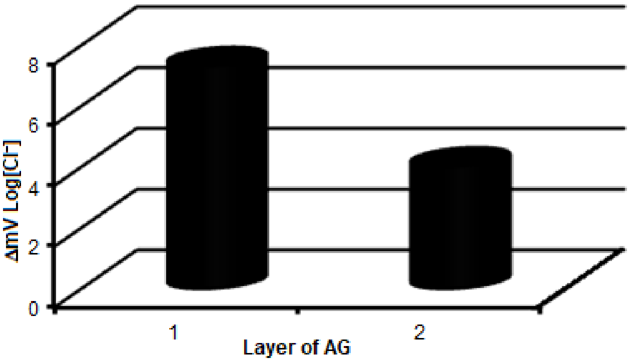

Fig 5. Comparison of ΔmV Cl^- vs Ag/AgCl standard reference electrode between SPE reference electrode first and the second type

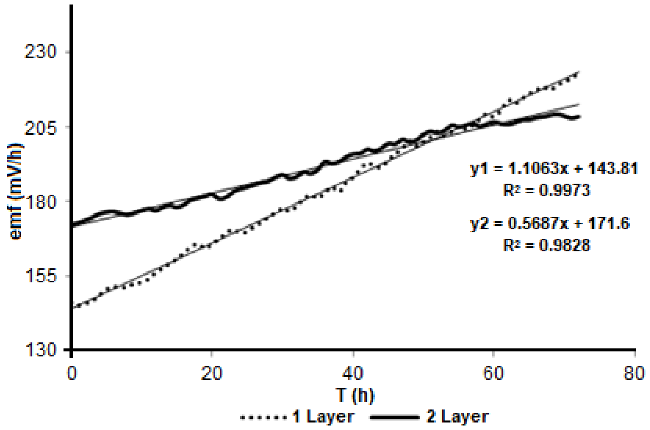

Fig 6. Stability test of the SPE reference electrode first and the second type in 0.01 M KCl for 72 h

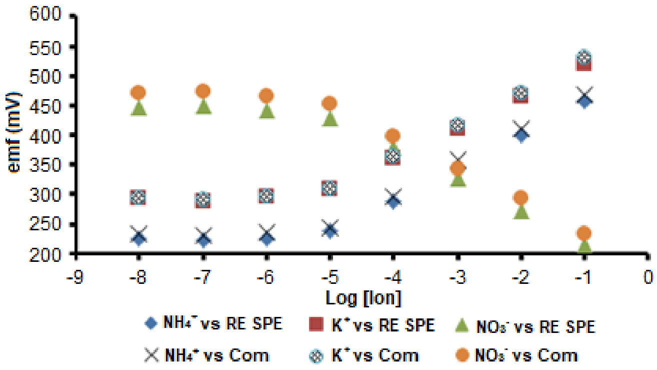

Fig 7. Performance test of the SPE reference electrode and a commercial reference electrode coupled with ISE's commercial

Stability

The stability of a reference electrode is one very important factor to take note in the reference electrode fabrication. The stability of the reference electrode will help produce a result of the ISE method became more accurate and precise especially for continuous measurement. Besides that, the stability of the reference electrode can also predict the lifetime of the reference electrode [1]. In this work, the stability studies of both SPE reference electrode were performed through the measurement of the different potential when paired with the double junction Ag/AgCl reference electrode from ORION. The measurement is performed by applying a solution of KCl, 0.01 M with the longest measurement for 72 h or three days by continuous measurement.

Fig. 6 shows the generated measurement reference electrode stability SPE the first type (one layer of AG) and the second type (two layers of AG). In general, both the SPE reference electrode shows good stability to 1.1 mV/h for which the first type and < 0.6 mV/h for the second type. However, the second type to look more stable. The stability of solid reference electrode Ag/AgCl in miniature size is generally influenced by three main factors, namely the first volume limited electrolyte, so rapidly depleted due to the occurrence of leach out salt electrolyte into the sample. The leach out process leads to the instability of the ions in the reference electrode [15]. The second factor is the dissolve of AgCl layer by the electrolyte, and this accelerated the formation of the oxide on the surface of the reference electrode, this causes the potential value of the reference electrode is changed [4]. The third occurrence the dilution process of the electrolyte in the reference electrode caused the small amount of water from the sample solution immersed and trapped in the membrane of solid-state reference electrode [2-3]. The third factor, particularly the second factor give a large part of why on the first type of the SPE reference electrodes have lower stability compared with the second type. In the first type, the AG membrane containing 3 M of KCl solution and placed just above the surface of the electrode Ag/AgCl without any separator layer. This circumstance makes the process of dissolving, and the formation of a layer of Ag_2CO_3 and Ag_2O on AgCl surface becomes faster than the SPE reference electrode type two [13].

Performance Tests with Commercial ISE

The last test of this research is looking at the performance of the SPE reference electrode vs. commercial ISE. The testing with ISE commercial to make sure that the SPE reference electrode based on AG membrane can be properly functionalized when coupled with the ISE sensor [2-3]. In this study, the selected of SPE reference electrode to be paired with the ISE sensor is the second type of the SPE reference electrode. The selection of the second type of the SPE reference electrode is based on the performance shown on the test was like ΔmV of Cl^- and the stability testing. The ISE commercial used in this research is NH_4^+, K^+, and NO_3^-. All's this ISE sensor is the most usable in various applications such as the environment, agriculture, food, industry, and health [16-18].

Testing with commercial ISE was done by using a various concentration of ion NH_4^+, K^+, and NO_3^- within the range 0.1–10^{-8} M. The test results with the coupled ISE shows the response that approximates the value of the Nernstian factor as displayed in Table 4 and Fig. 7. This result proves that the second type of SPE reference electrode can be used as a reference electrode and show the major feature that is not affected by changes in the concentration of the sample solution. The Nernstian factor also confirmed that the results do not have much difference with the coupled of

ISE commercial vs. the double junction of the Ag/AgCl reference electrode standard.

CONCLUSION

The SPE reference electrode Ag/AgCl based on AG membrane successfully developed and established the characteristics as a reference electrode. The test result is marked by ∆mV Cl^- measurement, stability test and performed by ISE commercial. In this study, the optimum concentration of an AG membrane generated is 20% (w/v) with two layers of the AG membranes.

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