Electrochemical Impedance Spectroscopy of Polyvinylalcohol Based Gel Electrolyte

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Abstract

Research on the effect of electrolyte ammonium salt, concentration electrolyte with plasticizer to ionic and electronic conductivity of polymer gel electrolyte has been conducted with the variations of two electrolyte NH_4Cl-PVA and NH_4SCN-PVA at a concentration of 10, 30, 40 (wt%). The measurement of using ionic conductivity by using impedance spectroscopy method showed that the highest value was 0.0156 Scm^{-1} i.e. in the 54.6% propilen carbonate with 40% NH_4Cl and PVA. The lowest value was 0.009 Scm^{-1} i.e. in the NH_4SCN based electrolyte without propilene carbonate and electronic conductivity showed that the highest value was 0.0156 Scm^{-1} i.e. 40% NH_4Cl-PVA and NH_4SCN-PVA and the lowest value was 0.009 Scm^{-1}, i.e. in the NH_4SCN based electrolyte without propilene carbonate. Factorial analyses showed that the concentrations of electrolyte and the plasticizer affect conductivity value. FTIR (Fourier Transform Infrared) measurement showed that the addition of propilene carbonate did not show new bond formation between electrolyte and PVA. It can be shown in absence of characteristic wave number for propilene carbonate and NH_4^+ ion.

Keywords: P.retrofractum, alkaloids, piperine, structure elucidation

Abstrak (Indonesian)

Penelitian tentang pengaruh elektrolit garam amonium, konsentrasi elektrolit serta plastisizer terhadap konduktivitas ionik dan konduktivitas elektronik gel polimer elektrolit telah dilakukan dengan variasi dua elektrolit NH_4Cl-PVA and NH_4SCN-PVA dengan konsentrasi 10%, 30%, 40% (b/b). Pengukuran konduktivitas ionik dan konduktivitas elektronik menggunakan metode spektroskopi impedansi yang menunjukan nilai impedansi tertinggi sebesar 0,0156 Scm^{-1} yaitu penambahan 2 mL propilen karbonat dengan 40 %NH_4Cl-PVA dan polivinil alkohol. Nilai terendah adalah 0,009 Scm^{-1} yaitu NH_4SCN-PVA dengan konsentrasi paling rendah tanpa penambahan propilen karbonat dan konduktivitas elektronik yang paling tinggi 0,0156 Scm^{-1} yaitu 40% NH_4Cl dan NH_4SCN dan yang paling rendah 0,009 Scm^{-1} yaitu NH_4SCN-PVA dengan konsentrasi paling rendah tanpa penambahan propilen karbonat. Analisa faktorial terhadap nilai konduktivitas memperlihatkan bahwa nilai konduktivitas dipengaruhi oleh konsentrasi elektrolit dan penambahan plastisizer propilen karbonat tetapi tidak dipengaruhi jenis elektrolit. Pada pengukuran menggunakan FTIR menunjukkan bahwa penambahan propilen karbonat tidak terjadi pergeseran bilangan gelombang antara elektrolit dan PVA. Hal ini dapat dilihat dari tidak adanya perubahan pada bilangan-bilangan gelombang yang khas baik untuk propilen karbonat maupun untuk ion NH_4^+.

Keywords: P.retrofractum, alkaloid, piperin, elusidasi struktur

INTRODUCTION

Conductivity occurs due to the movement of ions in the material, so that a material can conduct electricity. In metals, the charge is carried by electrons while in ionics, the charge is carried by the ions. Electrolytes are compounds that can conduct electricity by means of displacement of the ions. Most of the electrolyte solution or the salt is soluble, but some are solids, including crystalline solids. The solid material that has easy moving ion is known as a solid electrolyte, fast ion conductor and superonic conductor. The ability of a material to conduct electricity can be evaluated from its conductivity values.

One type of electrolyte that has many electrochemical applications is gel polymer electrolyte (GPE). Ionic salt with polymer has become such a breakthrough that recently many research focused on the development of GPE for electrochemical energy storage devices pplication, such as fuelcell, supercapacitor and sensors. GPE several advantages compared with such liquid polymer electrolyte is free of leaks, mechanical strength, flexibility, thus enabling miniaturization. However, low ionic conductivity is the barrier for its applications. Therefore, its electrical conductivity is necessary to increase in order to meet the requirement. Various methods have been used to increase the ionic conductivity of the polymer electrolyte such as, by incorporation of inorganic and organic fillers such as ceramics and complex compounds.

Ionic conductivity of GPE is affcted by the various factors such as the crystallinity – amorphous rasio of the polymer, simultaneous cation and anion movement and the formation of ion pairs (anion-cation interactions). The crystallinity of GPE material is grown through the use of salt or acid. Inversely, the increasing of amorphous phase can be done by using the addition of a low molecular weight plasticizer in small quantities. Plasticizer takes the role of the elastic properties, hardness and elasticity. Elasticity of a material is presented by the strain that occurs in a material when a tensile test is applied. The more elastic a material the molecule will be easy to move, making it easier for the ions to perform the movement. Patel et.al. [1] used plasticizers with inorganic salts resulting such degree of crystallinity with higher in ionic conductivity. Liang, et.al. [2] using the plasticizer materials such as propylene carbonate (PC), which results in improving in conductivity of 87.5% - 90%.

Protonated electrolytes, such as NH_4SCN and NH_4Cl are interesting to study because they have fairly good structural and thermal properties in composite [3], [4]. However, their electrical and ionic conductivity have not been studied. In this study, the GPE was prepared with protonated electrolytes (ammonium salts) and plasticizer, i.e PC. The conductivity, functionality and morphology of the GPE was examined by using electrochemial impedance spectroscopy (EIS), Fourier Transform Infrared (FTIR) spectroscopy, and scanning electron microscope (SEM) respectively.

MATERIALS AND METHODS

2.1. Preparation of Gel Polymer Electrolytes (GPE)

Method for preparing GPE can be found elsewhere [5]. A 20-mL beaker glass was filled with 2 grams of with 10% (w/w) NH4SCN in polyvinyl alcohol (PVA). 2 mL of distilled water was added to the mixture and stirred for 15 minutes at 90 - 95 ºC to form gel consistency. A 55% of propylene carbonate was stirred uniformly to the gel, then poured in petri dish and placed in desiccator. The gel was allowed to dry to formed gel polymer electrolyte (GPE). GPE was subjected to vacuum pump for final drying. The same procedure was also carried out on another variation of concentration (30 and 40%), electrolyte (NH4Cl) and with or without plasticizer (propylene carbonate).

2.2. Characterization and Evaluation of Gel Polymer Electrolytes

GPEs were characterized using SEM, and FTIR techniques to study morphologies and surface functionalities, respectively. SEM micrographs were obtained on JEOL JED-2300. FTIR spectra were obtained on Shimadzu IR Prestige 21. Direct measurement of electrochemical impedance spectroscopy (EIS) was performed by using an oscilloscope and function generator [6]. In this measurement, AC voltage was applied to DC circuit and a response voltage was measured. GPE was connected to the EIS instrumentation as illustrated on **Figure 1**. A function generator was used to supplied DC sine waves on various frequencies, i.e 1 Hz, 5 Hz, 10 Hz, 50 Hz, 100 Hz, 500 Hz, 1000 Hz, 5000 Hz, 10000 Hz, 1000000 Hz to the test circuit. Channel 1 and 2 on oscilloscope were functionized to measure applied and responsed voltages. The conductivity was calculated using the following formulas.

$$Z = V_2/V_1 \ x \ R \qquad\qquad (1)$$
$$K = \frac{1}{Z} \qquad\qquad\qquad (2)$$
$$K = K_{ionic} + K_{electronic} \qquad (3)$$

Where Z is the impedance in ohms, V_1 and V_2 were applied and responsed voltages (in volts), respectively, R is the resistance in ohms, K is the total conductivity

in siemens, K_{ionic} is ionic conductivity and $K_{electronic}$ is electronic conductivity.
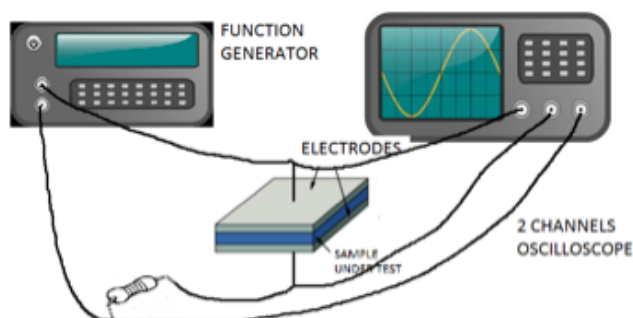

Figure 1. Instrumentation setting for electrochemical impedance spectroscopy measurements

The values for ionic conductivity were derived from measurement data using two different electrodes, such as silver and carbon electrodes, whereas the values for electronic conductivity were derived from measurement data using the same electrode, such as silver [7]. EIS measurement were also capable to derive other parameters, such as bode plot or phase shift [8]

2.3. Factorial Analysis

Factorial experiment design to three design factors, i.e.: 1) electrolytes, 2) concentrations, and 3) plasticizer. The three factors were controllable variables that have a key role to in the performance test, i.e conductivity. Since we have three factors to be considered, the experiment design is called an 18-full factorial design, each with combinations of the three factors across three or two levels of each. According to the general statistical approach for experimental design four replicates were obtained to get a reliable and precise estimate of the effects. The experiment was assumed that the factor is fixed. The design was completely randomized and the usual normality assumptions of the data were satisfied.

RESULT AND DISCUSSION

Awadhia and Agrawal [9] conducted a study on GPE which used protonated electrolyte, i.e. NH4SCN. In that study, the characteristics of the structure, thermal and electrical electrolyte gel but do not study the conductivity. Protonated electrolyte salts such as NH_4Cl which also has not been studied. There is a color distinction between GPEs based on plasticizer addition, i.e the GPEs without the addition of plasticizers has no color (transparent) otherwise white, as shown in **Figure 2**. The addition of the plasticizer in the electrolyte is aimed to improve the properties of the GPE in order to

the higher conductivity. Plasticizers provide elastic properties as indicated by the strain that occurs in a material when a tensile test is done so that the addition of the plasticizer can lower hardness properties of the electrolyte and improve elasticity.

3.1. Ionic Conductivity of Gel Polymer Electrolytes.

The value for ionic conductivity of GPEs were measured at frequency ranging from 5 Hz to 1 MHz with load 100 ohm. Values plot of GPEs ionic conductivity with NH_4Cl were presented in **Figure 3**. The graphs show that GPEs with 10% NH4Cl with or without the addition of plasticizers (propylene carbonate) have lowest conductivity value, i.e 0.009 Scm^{-1}.

Figure 2. GPE test samples (A) without and (B) with plastisizer

The results (**Figure 3**) show that in general the addition of propylene carbonate affect the ionic conductivity of NH_4Cl GPEs. The maximum conductivity can be achieved was 0.0156 Scm^{-1} that derive from 40% NH_4Cl – GPE with 2 mL of propylene carbonate.

The increasing of ionic conductivity value occured due to the higher number of ions and the mobility of ions existed. Osman et al. [10] explained that the ion conductivity of GPE is generally affected by the number of moving ions. However, the addition of plasticizers cause the value decreases as a result of ion density. The decreasing was caused by the distance

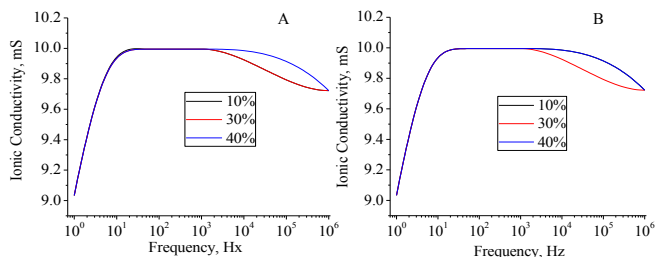between the ions is too close and lead to recombine of ions and form neutral pair that do not contribute to conductivity [11].

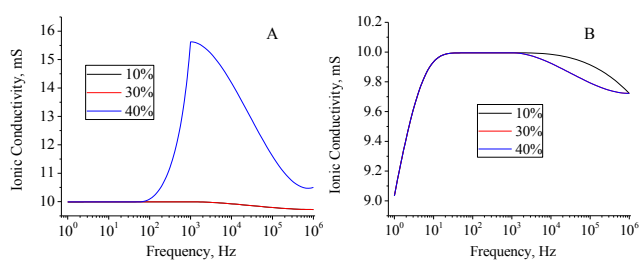Figure 3. Plots of ionic conductivity for 10-40% NH4Cl GPE (A) without and (B) 66.8% plasticizer

Figure 4. Plots of ionic conductivity for 10-40% NH4SCN GPE (A) without and (B) with 66.8% plasticizer
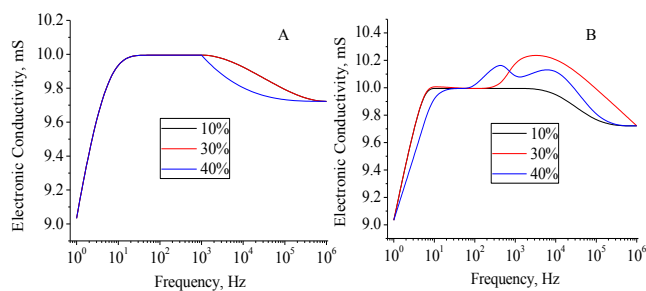

Figure 5. Plots of electronic conductivity for NH$_4$Cl GPE (A) without and (B) with 66.8% plasticizer
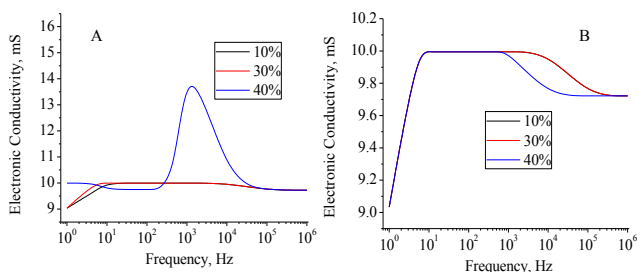
Figure 6. Plots of electronic conductivity for NH$_4$Cl GPE (A) without and (B) with 66.8% plasticizer

Values plot of GPEs ionic conductivity with NH$_4$SCN were presented in **Figure 4**. The results shows that 40% NH4SCN - GPE without the addition of PC has the lowest conductivity value, i.e 0.009 Scm^{-1}. It can be seen that the highest ionic conductivity (0.0156 Scm^{-1}) of the GPE was present in NH4SCN 40% - GPE with PC in the frequency below 1000 Hz.

3.2. Electronic Conductivity of Gel Polymer Electrolytes.

Figures 5 and **Figures 6** show the result plots of both NH$_4$Cl – GPE and NH$_4$SCN – GPE with or without the addition of plasticizers PC. It can be seen that the lowest value for electronic conductivity of both NH$_4$Cl – GPE and NH$_4$SCN – GPE are the same, i.e 0.009 Scm^{-1}. These results show no effect of PC addition on electronic conductivity at a concentration of 10% and 30%. Most of NH$_4$Cl – GPE with the addition of PC has 0.01025 Scm^{-1} of electronic conductivity. Inversely, 40% NH$_4$SCN – GPE with PC increased the electronic conductivity as 0.0156 Scm^{-1}.

The composition of the materials used in GPEs contributed to the ionic conductivity. In general, ionic conductivity resulting from the addition of plasticizers higher than without the addition of plasticizers. The increase in ionic conductivity showed higher number of ions in a polymer matrix. Plasticization process by using a low molecular weight, such as propylene carbonate increases the value of ionic conductivity and lower crystallinity mixture is formed.

3.3. Effects of electrolytes, concentrations and plastisizer on conductivity

The effect of electrolytes, concentrations and plastisizer on conductivity was analyzed by using full factorial method in Minitab®. Minitab is statistical package that assists in experimentation design and data analysis. Interaction and main effects plot are obtained to examine effect of factors on parameter. Design of experiment can be implemented by considering various input factors. For n factors and p variations p n trials are required to be carried out. Hence selection of n and p is very important in effectiveness of this method. For linear parameter variation p can be taken as 2 and to reduce the complexity, n is taken as 3. The usage of three variation (level) can be considered if there is a non-linierity in the system. Therefore, there are 18 experiments conducted for measuring ionic and electronic conductivity. Variations of these factors and the calculations results after processing measurement data are given in **Table 1**. Factorial analysis of all experiments is presented as plots in **Figure 7**.

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It can be shown in **Figure 7A** that concentration has highest effect on ionic conductivity. Some authors suggested that the materials used in the preparation of GPE have high effect on the ionic conductivity. This research suggests that, higher ionic conductivity can be obtained with the addition of plasticizers. Plasticization process by using a low molecular weight, such as propylene carbonate decrease the GPE density. Therefore, low crystallinity of GPE is formed.

The electrolytes (NH_4SCN and NH_4Cl) have only slightly effect on ionic conductivity. Both salts are relatively easy to hydrated compare to other salts such as nitrate, bromate and sulphate [12]. However, for the two protonated type electrolytes, NH4SCN produced more conductive GPE. According to Vanysek, et al [13] SCN^- ions was more easily hydrated and more mobile than the Cl^- ions so that the conductivity is greater. Inversely, th**e** high concentration of SCN^- have conductivity values lower than Cl^-. It is clearly shown that the concentration provided effect on the ionic conductivity.

Table 1. Variations of electrolytes, concentration and plastisizer in GPE and the results of calculations

#	Factors			Parameters (10^{-3} S)	
	Electrolytes	Concentrations	PLaPPlastisizer	K_{ionic}	$K_{electronic}$
1	NH_4Cl	10%	54.60%	10	10
2	NH_4Cl	10%	66.80%	10.72	10.25
3	NH_4Cl	10%	0%	10	10
4	NH_4Cl	30%	0%	10	10
5	NH_4Cl	30%	66.80%	10	10.25
6	NH_4Cl	30%	54.60%	10	10
7	NH_4Cl	40%	54.60%	15.62	15.62
8	NH_4Cl	40%	66.80%	10.62	10.25
9	NH_4Cl	40%	0%	10	10
10	NH_4SCN	10%	0%	10	10
11	NH_4SCN	10%	66.80%	10	10
12	NH_4SCN	10%	54.60%	10	10
13	NH_4SCN	30%	54.60%	10.25	10.25
14	NH_4SCN	30%	66.80%	10	10.25
15	NH_4SCN	30%	0%	10	10
16	NH_4SCN	40%	54.60%	10.25	10.25
17	NH_4SCN	40%	66.80%	10	10
18	NH_4SCN	40%	0%	15.62	15.62

Futhermore, the addition of propylene carbonate as plasticizer also affected ionic conductivity. The addition of 54.6% plasticizer slightly increased the conductivity and decrease significantly on the addition of 66.8%. It was obtained that optimum conditions for addition of plasticizers is 54.6%.

Interaction between factors analysis plots are given in **Figure 7B**. It is clearly shown that the ionic conductivity was affected by the interaction between electrolyte and concentration as well as interaction betweeen electrolyte and plastisizer. Interaction between concentration and plastisizer had highest effect on ionic conductivity only in concentration of 40%. Interestingly, this interaction had no effect on ionic conductivity due to lower concentration effect.
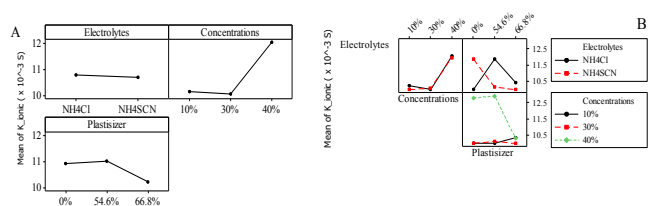

Figure 7. Factorial analysis plot of electrolyte, concentration and plastisizer on ionic conductivity of GPE (A) main factor and (B) interaction between factors
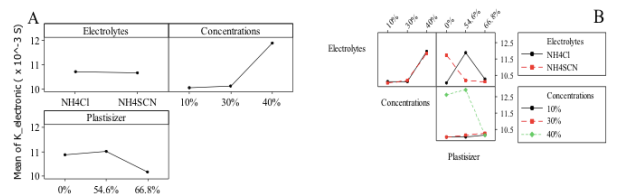

Figure 8. Factorial analysis plot of electrolyte, concentration and plastisizer on electronic conductivity of GPE (A) main factor and (B) interaction between factors

It can be seen in **Figure 8A** that the type of electrolytes slightly affect the value of the electronic conductivity. The electrolyte concentration in the ammonium salt 40% turned out to greatly affect the value of the electronic conductivity. The use of electrolyte is more than 30% will increase the electrical conductivity of the GPE as high as 1.2 mS. While the of the plasticizer as high as 54.6% slightly affect the value of conductivity with the low increasing of electronic conductivity. The more addition of plasticizer reduced the value. Factors interactions more likely affected the conductivity (**Figure 7B** and **8**B). It can be seen that the factor interaction can reduce or increase the value more less 2 mS. Interestingly, both type of GPEs had the same tendency due to the changes of three factors.

3.4. Surface Features of Gel Polymer Electrolytes

Fourier Transform Infrared (FTIR) spectra for GPE with and without plasticizer are shown in **Figure 9 and 10**. It can be shown that OH and NH_2 groups of each GPE derived from PVA and the ammonium salts (NH_4SCN and NH_4Cl). The amina group was observed at 3115 cm^{-1} and 3024 cm^{-1}. The increasing of amina due to the addition of ammonium salt to the GPE affects the increasing of intensity of the peaks in those wave number. According to Kadir et al. [14], the recombination of ions to form neutral or non-neutral ion pair causing the conductivity increased with the addition of 40% (w/w) ammonium salt.

It can be shown in **Figure 9** that the conduction in GPE clearly derived from the movement of proton from ammonium ion. The ions can move freely due to the absence of plasticizer; therefore it can be move from one bond to another bond and fill the empty area. In the

other hand, the addition of plasticizer has no effect on the functionality of organics in GPE (**Figure 10**). It can be seen that relatively, there is much change in intensity or shifting in wavenumber or in the other words, there is no formation of a new bond between the PVA-electrolyte (NH_4Cl and NH_4SCN).
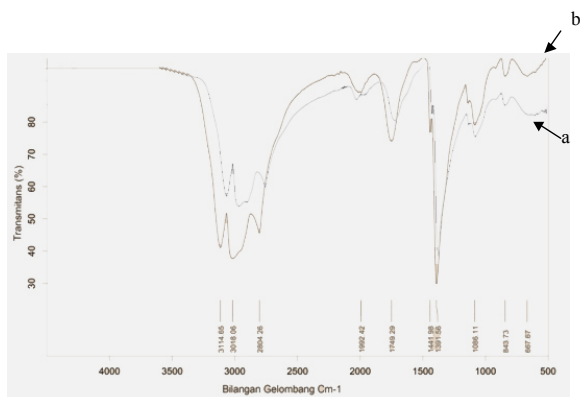

Figure 9. FTIR spectrogram of GPE, 40% NH4SCN (a) without and (b) with 54,6% plastisizer

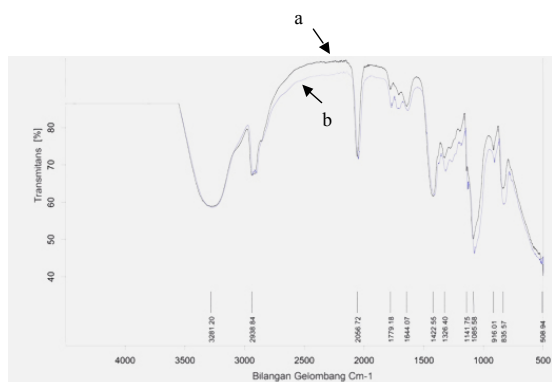Figure 10. FTIR spectrogram of GPE, NH4Cl (a) without; (b) and with 54,6% plastisizer

Kamisan, et al. [15] stated that the new bond of propylene carbonate and ammonium salts can be seen on 1000-1900 cm^{-1} and 700-800 cm^{-1} of wave number where the peak of C = O bond centered at 1785 cm^{-1} or shifted at 1780 cm^{-1} when bonded with ammonium salts. The bonding occured between the C = O of propylene carbonate and the lone pair of ammonium ion (NH_4^+) and formed the complex of $NH_4^+ \leftarrow OC$. In addition, the peak of NH_4^+ absorption in 1634 cm^{-1} shifted to 1626 cm^{-1} due to the addition of at least 0.7M salt. The existence of new absorption peaks as a result of the addition of propylene carbonate, i.e. at 1195.4 cm^{-1}, indicated the absorption of CH_2 of propylene carbonate. It can be inferred that the addition of propylene carbonate caused the GPE not rigid and relatively elastic. Therefore, two characters contributed the mechanical strength of GPE.
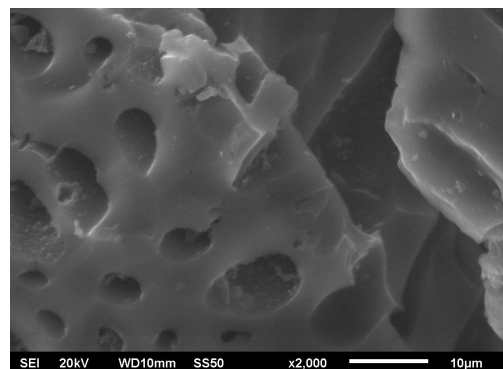

Figure 10. SEM image of GPE, 30% NH₄SCN without plastisizer

Figure 10 show the microstuctures of GPE-NH_4SCN without plasticizer. The GPE exhibit non-uniform pore typically in the range of 2 – 10 μm. It can be seen that the structure is uniquely rigid.

CONCLUSION

The measurement of using ionic conductivity by using impedance spectroscopy method showed that the highest value was 0.0156 Scm^{-1} i.e. in the 54.6% propilen carbonate with 40% NH_4Cl and PVA. The lowest value was 0.009 Scm^{-1} i.e. in the NH_4SCN based electrolyte without propilene carbonate and electronic conductivity showed that the highest value was 0.0156 Scm^{-1} i.e. 40% NH_4Cl-PVA and NH_4SCN-PVA and the lowest value was 0.009 Scm^{-1}, i.e. in the NH_4SCN based electrolyte without propilene carbonate. Faktorial analyses showed that the concentrations electrolyte and the plasticizer affect conductivity value. FTIR (Fourier Transform Infrared) measurement showed that the addition of propilene carbonate did not show new bond formation between electrolyte and PVA. It can be shown in absence of characteristic wave number for propilene carbonate and NH_4^+ ion.

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