

# Synthesis and Characterization of Polymer Electrolyte Membrane Based on Cellulose-Chitosan-Alginate as Li-Ion Battery Separator

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**Abstract** - The current commercial Gel Polymer Electrolyte (GPE) products are generally made of synthetic and non-biodegradable materials. In addition, some of these polymers require toxic reagents and complex synthesis processes. The purpose of this research is to manufacture GPE membrane products using biodegradable raw materials, a combination of Hydroxy Ethyl Cellulose (HEC), Carboxymethyl Chitosan (CMCs), and Sodium Alginate (SA) with lithium salt as the electrolyte source. The methods start from the fabrication/synthesis of biodegradable GPE membranes in various compositions, then LiOH is added as an electrolyte source and glutaraldehyde as a crosslinking agent using a solution casting technique. The mechanical membrane testing (tensile strength and elongation) and characterization were carried out using XRD, SEM, and FTIR. Based on mechanical tests carried out, variations in HEC 50%: SA 50% has the highest tensile strength value of 81.4255 MPa and the lowest elongation value of 11.68%. The results of XRD analysis in the presence of a typical peak in the HEC: SA variation was 11.56°, which could affect the strength of the electrolyte-polymer gel membrane (GPE). The results of SEM analysis proved that the HEC: SA variation has a porous morphology that can affect the ion absorption capacity in lithium-ion battery applications. The results of FTIR analysis proved that there are functional groups S=O, CH, CO, NH, OH, and COC in the three membranes (SA, CMCs, and HEC).

**Keywords**— Cellulose; chitosan; alginate; electrolyte polymer gel; lithium-ion battery.

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## I. INTRODUCTION

Lithium Ion Batteries (LIBs), with their high energy density and operating voltage, good stability, specific capacity, and higher cycle performance make rechargeable lithium-ion batteries have attracted the attention of researchers in recent decades. The use of these LIBs has been explored and widely applied in the field of energy storage and portable electronic devices such as electric vehicles (EV) and hybrid electric vehicles (HEV), cell phones, laptops, digital cameras, etc. [1], [2]. However, using LIBs still has safety hazards such as internal short circuits, liquid leakage, fire, and even explosion [3], [4]. To overcome this problem, researchers in recent years carried out studies to replace organic liquid electrolytes with gel polymer electrolytes

(GPE), which are based on a framework polymer material with a porous structure [5] with a ratio of liquid electrolyte and solid polymer electrolyte. The separator in LIBs affects the safety of LIBs because it isolates the cathode and anode from the contacts, which causes an internal short circuit and plays an important role in ion delivery through microporous cells filled with electrolytes [6]. Until now, commercially produced GPEs are made from synthetic materials such as poly (ethylene oxide) (PEO) [7], [8], poly (acrylonitrile) (PAN) [9,10], poly (methyl methacrylate) (PMMA) [11], poly (vinyl alcohol) (PVA) [12], poly (vinylidene) fluoride (PVDF) [13] and its derivatives. These synthetic polymers have low mechanical strength and high production costs. Another risk of these non-biodegradable GPE products is that they cannot be degraded easily, causing "white pollution" for

the environment. In addition, some of these polymers require toxic reagents and complex synthesis processes. To overcome these problems, developing environmentally friendly GPE needs to be done. Besides being environmentally friendly, some important points and requirements for GPE fabrication are membranes with good ionic conductivity, low cost, good dimensions, and good mechanical stability [14]. Natural polymers that meet the above requirements can be used as electrolyte polymers. Recently, researchers have proposed many natural polymers, such as cellulose and their derivatives [15], starch [16], chitosan [17], and alginate [18], which are suitable for use as host polymers in GPE. The purpose of this research is to make a GPE membrane product using a combination of biodegradable raw materials of cellulose, chitosan, and alginate with lithium salt as the electrolyte source as well as to examine the mechanical and physical properties of the resulting membrane and its characterization includes tensile strength, elongation, thickness, FT-IR, XRD, SEM.

## II. MATERIAL AND METHOD

### A. Materials

The materials used in this study include Hydroxy Ethyl Cellulose (HEC), Carboxymethyl Chitosan (CMCs), and Sodium Alginate (SA). Glutaraldehyde 25% pa, LiOH (Lithium Hydroxide) electrolyte solution, Aqua DM. The equipment used includes FTIR (Fourier Transform Infra-Red) instruments, XRD (X-Ray Diffraction), SEM (Scanning Electron Microscopy), and Tensile Tests.

### B. GPE Membrane Fabrication

The polymer electrolyte membrane fabrication is carried out by the Solution Casting method. The experiment was started by dissolving each ingredient with a total concentration of 3% (3 g in 100 ml of aqua DM), then mechanical stirring while heated using a hot plate at 50°C for 2 hours to obtain a homogeneous solution. Added 0.5 g of Lithium Hydroxide (LiOH) and 1 mL of 6% glutaraldehyde as crosslinking agent..

TABLE I  
GPE MEMBRANE COMPOSITION

Membrane Variations	Hydroxyethyl (HEC) (3% b/v)	Cellulose	Carboxylated (CC) (3% B/v)	Chitosan	Sodium Alginate (SA) (3% b/v)	Glutaraldehyde (mL)	6%	Lithium Hidroside (LiOH) (g)
1	100		0		0	6 mL		0.5
2	0		100		0	6 mL		0.5
3	0		0		100	6 mL		0.5
4	50		50		0	6 mL		0.5
5	50		0		50	6 mL		0.5
6	0		50		50	6 mL		0.5

The homogeneous solution was poured into a glass container, then dried in an oven for 1 hour at 70°C and allowed to dry at room temperature (25±10°C) for later characterization and testing. The composition of the membrane mixture was made in several composition variations, as shown in Table 1.

### C. GPE Membrane Testing and Characterization

1) *GPE Membrane Mechanical Test:* The synthesized GPE membrane was tested using the Tensile Strength, Elongation, and Thickness Test with the ASTM D-638 method for membranes/plastics. Hence, the initial and final length/elongation (cm) and tensile strength values (Kg. F) are obtained.

2) *GPE Membrane Characterization:* The characterization of constituent membrane materials to determine their functional groups was carried out with the FTIR Spectroscopy instrument. The resulting membrane crystallinity was characterized by XRD instrument, and the synthesized GPE membrane morphology was observed using an SEM instrument.

(SA), Carboxymethyl Chitosan (CMCs) and Hydroxyethyl Cellulose (HEC) which can be seen in Table 2.

The manufacture of this GPE membrane is carried out by the solution casting method, by dissolving the raw materials sodium alginate (SA), carboxymethyl chitosan (CMCs), and hydroxyethyl cellulose (HEC) into 100 ml of aqua DM with several variations of 100% sodium alginate (SA), carboxymethyl chitosan (CMCs) 100%, hydroxyethyl cellulose (HEC) 100%, sodium alginate (SA) 50%: carboxymethyl chitosan (CMCs) 50%, carboxymethyl chitosan (CMCs) 50%: hydroxyethyl cellulose (HEC) 50% and sodium alginate (SA) 50%: hydroxyethyl cellulose (HEC) 50%, which is then added with 0.5 g of LiOH electrolyte solution and followed by the addition of 6 ml of 6% glutaraldehyde. The resulting membrane will be brownish-yellow and thickened.







The addition of an electrolyte solution (LiOH) in the manufacture of the membrane aims to increase the value of the ionic conductivity of the lithium battery. This is supported by Hu et al. [19], who said that the concentration and type of lithium salt affect the ionic conductivity of the membrane. The higher the lithium salt concentration, the lower the degree of dissociation. This explains the presence of an optimum salt concentration. The fraction of free ions reaches its maximum point at this optimum concentration. The miscibility of the lithium salt in the polymer solution will cause a difference in the value of the ionic conductivity of the electrolyte membrane. In comparison, adding glutaraldehyde to the membrane aims as a plasticizer that affects the level of elongation (elongation).

## III. RESULT AND DISCUSSION

### A. GPE Membrane Fabrication

The membrane was made at the Integrated Chemistry Laboratory of the Faculty of Mathematics and Natural Sciences, University of Muhammadiyah Riau, Pekanbaru, with several variations in the composition of Sodium Alginate

TABLE II  
PICTURE OF MEMBRANE VARIATION

No	Variation (%)			Resulting Membrane	Information
	HEC	CMCs	SA		
1	100	0	0		Thin sheet, brownish, slightly rough surface (GPE-1)
2	0	100	0		Thin sheet, brownish, slightly rough surface (GPE-2)
3	0	0	100		Thin sheet, brownish, smooth surface (GPE-3)
4	50	50	0		Thin sheet, brownish and slightly rough surface (GPE-4)
5	0	50	50		Thin sheet, brownish, slightly rough surface (GPE-5)
6	50	0	50		Thin sheet, brownish and smooth surface (GPE-6)

### B. GPE Membrane Mechanical Properties Testing

Tests for tensile strength (Tensile), elongation (Elongation), and thickness (Thickness) were carried out at the Research and Development Laboratory of PT. Indah Kiat Pulp & Paper, Perawang. The results of the three test parameters of several variations in the composition of Sodium Alginate (SA), Carboxymethyl Chitosan (CMCs), and Hydroxy Ethyl Cellulose (HEC) are as follows.

#### 1) Tensile Strength Test:

Based on the results of the tensile strength test (Tensile) in Table 2, it can be seen that the highest tensile strength value is 50:50 variation with each material sodium alginate (SA): hydroxyethyl cellulose (HEC) which is 81.4255 Mpa. The high tensile strength value is because the alginate used can bind water better, increasing the percentage of better tensile strength value as well. This is supported by Hu et al. [19], who reported that alginate was able to form a strong polymer matrix and made the intermolecular tensile strength stronger on the membrane. Meanwhile, increasing the hydroxyethyl cellulose (HEC) concentration tends to increase the tensile strength of the membrane.

This is also supported by Luo et al. [20], who reported that this increase in tensile strength occurred because cellulose has a straight and long polymer chain, so it can make the membrane stronger. Then the addition of cellulose due to good interfacial adhesion can form a network of strong hydrogen bonds that occur so that the tensile strength

increases well. Based on this, the best tensile strength value is in the 50:50 variation with each ingredient, sodium alginate (SA): hydroxyethyl cellulose (HEC).

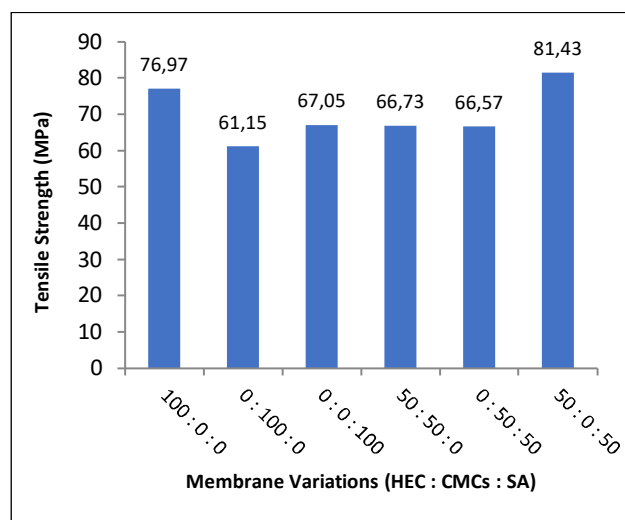


Fig. 1 Tensile Strength of GPE Membrane Variation

#### 2) Percent Elongation Test:

Elongation is the ratio of the increase in the length of the membrane to the initial length. Elongation is carried out to determine the elongation ability of the membrane. The higher the elongation value, the more flexible and plastic the membrane is. In the tests carried out, the highest elongation

value on the membrane with variations of Carboxymethyl Chitosan (CMCs) increased by 25.045%, while the lowest elongation value on membranes with variations of hydroxyethyl cellulose (HEC): sodium alginate (SA) with a value of 11.68%. This is because the addition of hydroxyethyl cellulose (HEC) in the variation will cause a decrease in the elongation value of the membrane.

Based on the theory proposed by Bao et al. [21], it is said that increasing the concentration of membrane-forming materials causes the elongation value at the break to decrease. This shows that the increasing value of elongation at break is inversely proportional to the tensile strength value. Therefore, the greater the tensile strength value, the smaller the elongation value at the break. This is also supported by Bao et al. [21], who reported that the decrease in cellulose content could occur due to the presence of the carbonyl group (COOH) of cellulose. This bond produces high strength and reduces elastic properties because of its cellulose composition; the percent elongation will decrease. This statement is supported by Wichai et al. [22], who state that the increase in elongation value due to the addition of plasticizers can reduce the intermolecular strength of bioplastics between polymer chains and increase the flexibility of bioplastics.

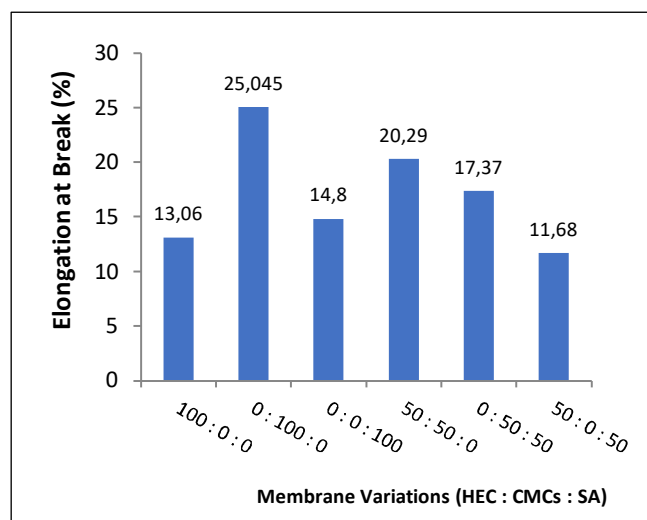


Fig. 2 Elongation at Break of GPE Membrane Variations

### 3) Thickness Test:

The thickness of each gel polymer electrolyte (GPE) membrane in Table 4 was obtained with values between 0.106 mm – 0.236 mm. In this thickness test, the highest value is the variation of sodium alginate (SA): hydroxyethyl cellulose (HEC). This is because the addition of hydroxyethyl cellulose (HEC) and sodium alginate (SA) used in variations can affect the thickness of the electrolyte polymer gel membrane (GPE). This statement is supported by Karzar Jeddı and Mahkam [23], who reported that the concentration of alginate used would affect the thickness of the membrane because the alginate concentration would affect the total solids of the membranes. This total solid will result in a rigid molecular chain. Due to its hydrophilic nature, the polymer is surrounded by mobilized water molecules, causing the alginate membrane to thicken. This total solid will result in a rigid molecular chain. Due to its hydrophilic nature, the polymer is surrounded by

mobilized water molecules, causing the alginate membrane to thicken.

Differences in membrane imprinting can also cause the thickness of the electrolyte polymer gel (GPE) membrane. According to Karzar Jeddı and Mahkam [23], the thicker the membrane, the higher the tensile strength, the lower the elongation value, and the lower the water vapor transmission rate. The thickness results on membranes with variations from SA to HEC: SA in Table 4 are still in the category of fulfilling the requirements based on Japanese Industrial Standard 2-1707 (Edible film standard) with a value of Max. 0.25 mm.

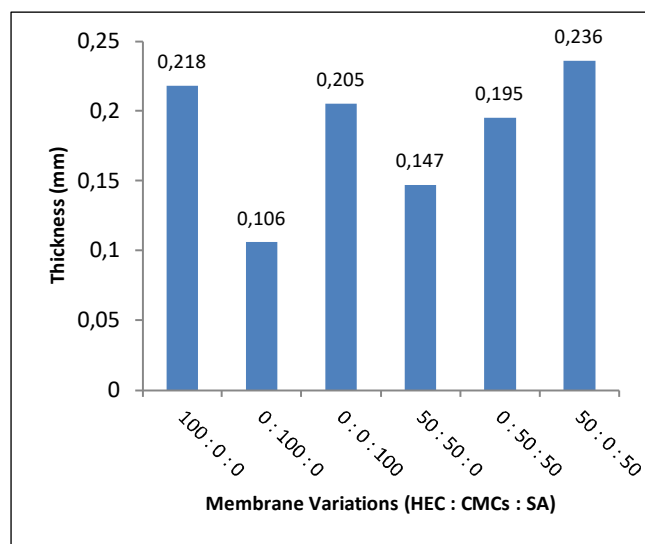


Fig. 3 Thickness of GPE Membrane Variation

### C. GPE Membrane Characterization

#### 1) X-Ray Diffraction (XRD):

XRD (X-Ray Diffraction) testing aims to determine the crystal structure formed on membranes based on alginate, carboxymethyl chitosan, and hydroxyethyl cellulose. Through testing with XRD, it can also be seen that the hardness of the membrane has increased by looking at the highest peak intensity at the time of testing [24]. XRD diffraction patterns of alginate, carboxymethyl chitosan, and hydroxyethyl cellulose samples were obtained under operating conditions involving Cu radiation at 30 mA, 40 kV. Relative intensities were recorded at angles ( $2\theta$ ) from  $5^\circ$  to  $80^\circ$ . XRD instrument tests used sample variations at 100% SA, 100% HEC, and 50% HEC: 50% SA. This variation was obtained from the best results after testing the physical and mechanical properties of the electrolytic polymer gel membrane (GPE). The XRD analysis showed a peak in the sample code C at angles  $2\theta = 21.64^\circ, 29.87^\circ, 30.89^\circ,$  and  $32.11^\circ$ . The HEC sample code produces peaks at  $21.37^\circ, 23.44^\circ, 29.49^\circ, 30.56^\circ, 31.81^\circ, 34.09^\circ, 36.16^\circ, 37.05^\circ, 39.81^\circ, 43.54^\circ$  and  $48, 83^\circ$ .

The results of XRD analysis on combining materials with sample code HEC: SA produced peaks of  $11.56^\circ, 21.41^\circ, 23.45^\circ, 29.52^\circ, 30.61^\circ, 31.82^\circ, 34.17^\circ, 35.08^\circ, 36.17^\circ, 36.96^\circ$  and  $39.66^\circ$  which indicate that the three materials with sample code SA100%, HEC 100% and HEC 50%: SA 50% are crystalline which means they have an orderly arrangement of particles against the electrolyte polymer gel membrane (GPE). The HEC: SA variation sample has a typical diffraction peak



at  $11.56^\circ$ . This characteristic peak is owned because the two materials can form a strong network. This statement is also related based on the value of the tensile strength in the HEC: SA variation; high results are obtained. This is also supported by Kang et al. [25], who state that the interaction between two polymer types will form a strong network with good mechanical properties, but it is inefficient as a water vaporizer because of its low elongation rate.

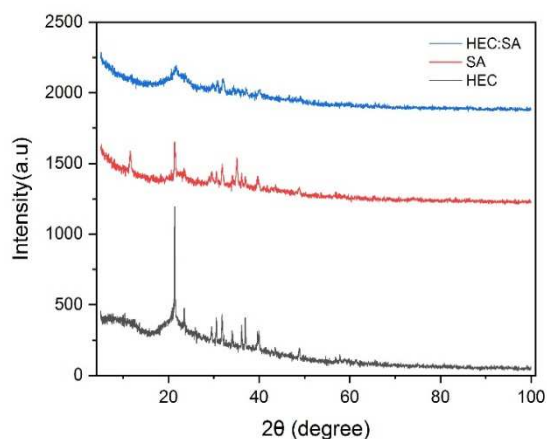


Fig. 4 X-Ray Diffraction of GPE Membrane

### 2) Scanning Electron Microscope (SEM):

SEM (Scanning Electron Microscopy) analysis aims to determine the morphological structure of the membrane. SEM analysis was carried out at 1000x and 2500x transverse magnification. The results of the SEM analysis can be seen in Figure 5. morphological differences between edible films

HEC: SA, HEC: CMCs, and SA: CMCs. In figure 5 (a), the surface of the membrane in the HEC: SA variation shows that clumps and visible crystal granules appear on the membrane, which is estimated to be LiOH salts scattered in the membrane matrix.

Figures 5 (b) and (c) are membranes with variations of HEC: CMCs and SA: CMCs. It can be seen that the resulting matrix is hollow and porous. This is probably formed due to the presence of air bubbles trapped in the membrane. This is supported by Li, Ding, and Han [26], stating that the clumps and empty spaces are due to the weakening of molecular bonds, so it will decrease the density and affect the porosity properties of the membrane matrix.

The results of cross-sectional SEM analysis showed differences in the membrane microstructure according to Chitra et al. [27], stating that a high pore density indicates many pores with a large surface area. The less-dense the membrane structure, causes more water is absorbed, and this supports the process of ion exchange mechanisms that occur in the membrane [28]. From this statement, the morphological results on the membrane can affect the physical and mechanical properties of the polymer electrolyte gel membrane (GPE) regarding the value of tensile strength and elongation, that the denser the matrix on the membrane, the fewer cavities are produced, and the elongation value is small as shown in Figure 5 (a) HEC: SA variation. While in Figures 5 (b) and (c), variations of HEC: CMCs and SA: CMCs have low tensile strength values but high elongation values and have pores in the membrane matrix which indicate that these membranes have high water absorption and will affect the ion exchange that occurs in lithium battery applications [29].

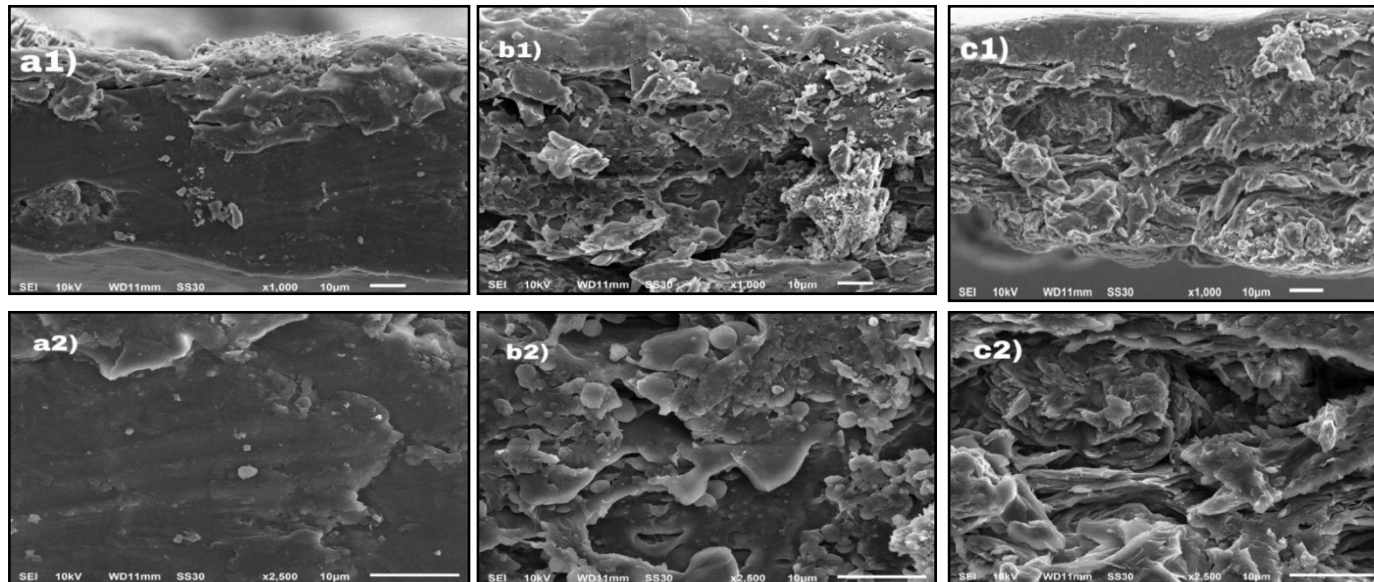


Fig. 5 SEM Micrograph of GPE Membrane a) HEC: SA variation, b) HEC: CMCs variation, and c) SA: CMCs variation in magnification 1.000x (1) and 2.500x (2)

### 3) Fourier Transform Infra-Red (FTIR):

The FTIR (Fourier Transform Infrared) test is used to see the functional groups on the polymer gel membrane (GPE). The new functional groups formed on the membrane indicate chemical interactions while combining functional groups between components on the membrane indicates physical mixing [30]. Based on the tests carried out on alginate, the

results of the FTIR analysis for alginate showed that the absorption band at  $1287.54\text{ cm}^{-1}$  was a typical area of the CO group. The presence of the CO group in the absorption band comes from the carboxyl group that makes up the structure of the alginate compound. Another spectrum appears in the fingerprint region of  $1391.37\text{ cm}^{-1}$ , which is the O-Na absorption band of Na in the alginate structure [31]. Another absorption band appears at wave number  $2940.61\text{ cm}^{-1}$  with

medium intensity indicating the presence of a CH group. In the hydroxyethyl cellulose spectrum, the appearance of a spectral peak at wave number  $3032.32\text{ cm}^{-1}$  indicates a stretch in the CH group. The indexed functional groups were the CO functional groups at wavelengths of  $1004.96\text{ cm}^{-1}$ ,  $1037.75\text{ cm}^{-1}$ ,  $1101.40\text{ cm}^{-1}$ , and  $1203.63\text{ cm}^{-1}$ . The absorption at the number  $1585.55\text{ cm}^{-1}$  is the absorption of NH.

Furthermore, the CO group has a wavelength of  $1102.37\text{ cm}^{-1}$ . The functional group detected was the COC functional group at a wavelength of  $1028.10\text{ cm}^{-1}$ , which shows that Schiff's base reaction happened between the hydroxyethyl cellulose aldehyde oxidation group and the carboxymethyl chitosan amino group. Based on the FTIR spectrum, the

functional groups contained in hydroxyethyl cellulose are OH, CO, and COC [32].

Based on the FTIR spectrum, the functional groups contained in carboxymethyl chitosan are CH, OH, CO, and NH. At a wavelength of  $3430.34\text{ cm}^{-1}$ , it shows the stretching vibration of OH. According to Perumal et al. [33], the vibration peak,  $1155\text{ cm}^{-1}$ , is the absorption peak of -OH on the C6 position of chitosan. Compared with the CS infrared spectrum, strong new absorption peaks at  $1596\text{ cm}^{-1}$  and  $1410\text{ cm}^{-1}$  on the infrared spectrum of CMCS are COO-symmetrical stretching vibration and asymmetric stretching vibration absorption peaks, respectively, indicating the carboxymethyl group was successfully introduced on chitosan.

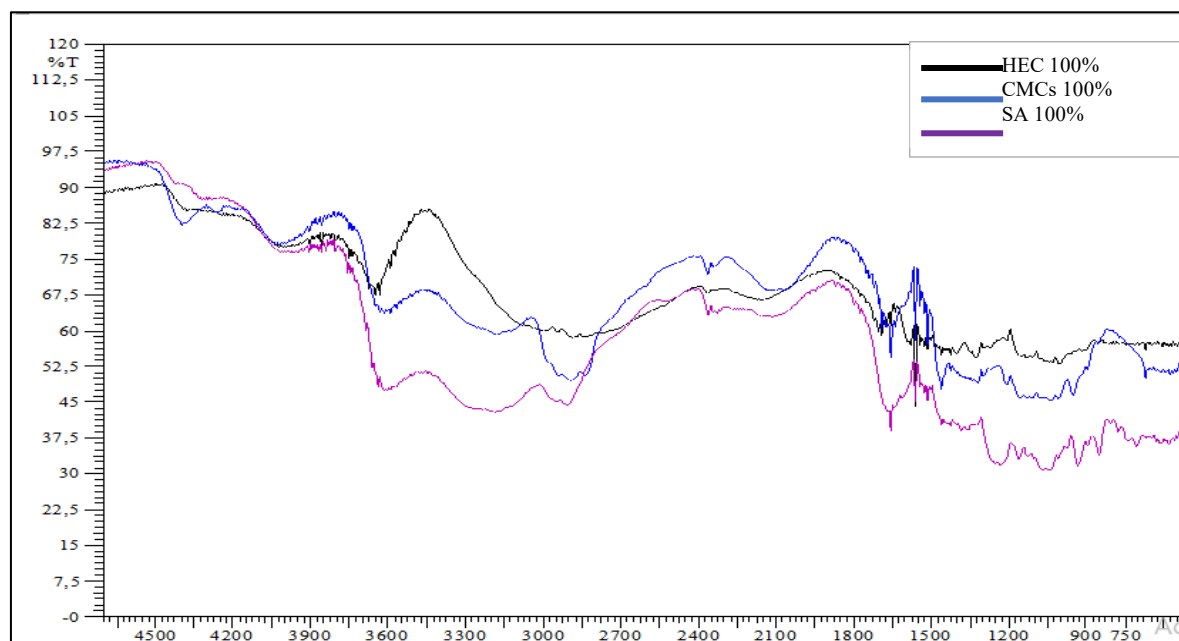


Fig. 6 FTIR Spectroscopy of Membranes

#### IV. CONCLUSION

Based on the research and discussion that has been described, it can be concluded that: The results of the FTIR analysis prove that the functional groups are S=O, CH, and CO in alginate, and there are CH, OH, CO, and NH functional groups in carboxymethyl chitosan and OH, CO, and COC functional groups in the hydroxyethyl cellulose sample. The results of XRD analysis resulted in a typical peak in the HEC: SA variation of  $11.56^\circ$ , which could affect the strength of the electrolyte-polymer gel membrane (GPE). The results of SEM analysis prove that the HEC: SA variation has a tight matrix and a porous morphology which can affect the ion absorption capacity in lithium battery applications. Based on mechanical tests, variations in Alginate (SA) 50%: Hydroxy Ethyl Cellulose (HEC) 50% has the highest tensile strength value of  $81.4255\text{ MPa}$ , and the lowest elongation value is  $11.68\%$ .

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