Research Article

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Mixed gel electrolytes: Synthesis, characterization, and gas release on PbSb electrode

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Abstract: Mixed gel electrolytes were fabricated chemically at a temperature range of 2–5°C using some additives comparing with traditional electrolytes (1.26 g/cm³ H₂SO₄). The polyacrylamide (PAM) content is kept constant (0.2 wt%), while the propylene glycol (PPG) and nano fumed silica (NFS) contents varied. The material characteristics of samples were assessed by physical appearance observation, electrochemical measurements, and physicochemical methods. The results showed that a hard gel without separation of solutions was found in the case using a PAM/PPG/NFS ternary additive (0.2/0.1/0.6 wt%). The morphological structure (<50 nm) of mixed gel electrolytes was in the range of nanometer. The gas release on the PbSb electrode was strongly inhibited (92.2–93.7% for O₂ and ~55% for H₂ at the 120th cycle), which will both contribute to reducing the pressure as well as limiting water loss in the sealed lead acid battery, compared with that in the sulfuric acid medium.

Keywords: mixed gel electrolyte, additives, ionic conductivity, diffusion coefficient, gas release

1 Introduction

Currently, mixed gel electrolytes are of concern for scientists in the field of sealed lead acid batteries [1–4] because there is no need for maintenance, and acid vapors that can damage the surroundings are also controlled. To improve them, various additives as gelling agents were used. For example, vanillin is referred to as a gelling agent that enhances battery deep discharge but reduces capacity after a storage period of only 3 months [4]. Some polyols such as propanetriol, butanetriol, pentitol, and pentaerythritol can facilitate the gelation process due to the ability to repel water molecules from the hydrate layer surrounding SiO₂ [5]. However, their electrochemical properties are also affected by the length of the carbon branch and the number of OH groups. PAM is an additive that both reduces the gelling time and increases the hardness of gelled electrolytes, which can increase the discharge capacity of batteries [4,6,7]. PPG is a well-expanding polymer with natural hydrophobic properties, which are used for the synthesis of organo-modified SBA-15 [8]. However, it is not yet used as an additive in the manufacture of gel electrolytes for the battery. According to the previous research, mixed gel electrolytes could retain more sulfuric acid compared with that using a single gelling agent [9]. The charge transfer resistance at the open circuit potential (OCP) can also be reduced because of their three-dimensional structures, leading to an improved initial capacity of the gel battery. Several oxides such as TiO₂, Al₂O₃, and B₂O₃ have been studied for fuming silica (up to 6%)-based gel electrolyte system [3]. However, its viscosity increased due to the presence of B₂O₃, and the three-dimensional space structure decreased because Al₂O₃ was not well connected with the isolated silanol groups, except for TiO₂ because of its ability to form a three-dimensional space like fuming silica.

This article presents some results about mixed gel electrolytes containing PAM, PPG, and NFS (in various

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weight percent) compared with traditional electrolytes. The aim of the study is to synthesize mixed gel electrolytes, to evaluate the properties of materials, and to investigate the gas release at PbSb electrodes in different electrolytes. The relationship between the material properties and the diffusion capacity of the HSO_4^- ion in mixed gel electrolytes will be elucidated.

2 Materials and methods

Chemicals used in this study were provided by Sigma Aldrich (NFS 7 nm; PAM), Merck (PPG, H_2SO_4), and China (alcohol; $\text{Na}_2\text{SiO}_3 \cdot x\text{H}_2\text{O}$ (1.54 g/cm³)).

2.1 Sample manufacturing

The temperature was kept in the range of 2–5°C for material synthesis. H_2SO_4 solution (1.37 g/cm³) and $\text{Na}_2\text{SiO}_3 \cdot x\text{H}_2\text{O}$ (1.54 g/cm³) were used in the volume ratio of 3.2/1.0 for sample preparation. A fresh 5% NFS suspension was used as one of the additives whose content was (in weight percentage) compared with the mass of sulfuric acid. The content of PAM was constant (0.2 wt%), while the content ranges of PPG and NFS were 0.0–0.4 wt% and 0.2–0.8 wt%, respectively. After synthesis, the samples were washed with alcohol to remove water from the gel. They were then dried at 40°C for a while to provide for measuring infrared spectroscopy measurement, X-ray diffraction, SEM imaging, and thermal analysis.

2.2 Research methods

First, the physical appearance of samples was observed visually after fabrication. Their ionic conductivity was then measured on electrochemical equipment (IM6, Zahner-Elektrok, Germany) using the EIS method. Two stainless steel electrodes were used for the cell in EIS measurements at OCP (frequency: 8 MHz–1 kHz; amplitude: 5 mV). The resistance of electrolytes is the value of the intersection between the Nyquist line and the horizontal axis following Thales Software [10]. The ionic conductivity χ (in S/cm) can be determined by Eq. 1, in which l is the length of electrolyte block (in cm), A is the cross section of this block (in cm²), and R_Ω is the electrolyte resistance (in Ω).

$$\chi = \frac{l}{AR_\Omega}. \quad (1)$$

The HSO_4^- ions play an important role in the electrode reactions of lead acid batteries, so their diffusion coefficient should be considered in this study. It can be detected by the cyclic voltammetry (CV) method (scanning speed: 30–100 mV/s; potential range from –1.5 to –0.5 V; PbSb material as a working electrode; platin sheet as a counter electrode; and $\text{Hg}/\text{Hg}_2\text{SO}_4/\text{sat.K}_2\text{SO}_4$ as a reference electrode). The plot is constructed from the Randles–Sevcik equation at 25°C to determine the diffusion coefficient D_o [11] following Eq. 2:

$$i_p = 2.69 \times 10^5 n^{3/2} A D_o^{1/2} C_o v^{1/2} \quad (2)$$

where i_p is the oxidation peak (in A/cm²), n is the exchange electronic number, A is the electrode surface area (in cm²), D_o is the diffusion coefficient (in cm²/s), C_o is the initial concentration of the oxidant (in mol/cm³), and v is the scan rate (in V/s).

The stability of the mixed gel electrolyte and the gas release were also observed by CV measurements with the same three-electrode cell. One hundred twenty CV cycles were carried out at a scan rate of 100 mV/s (potential range: from –1.5 to 2.1 V). The amount of electricity devoted to the oxygen release is calculated by the peak integral in the oxygen draining zone thanks to Thales Software [10].

The infrared spectrum was measured in the wavelength range from 4,000 to 400 cm^{–1} on the Spectrum Two FTIR Spectrometers by Perkin Elmer (USA). The crystal structures of samples were analyzed on X-ray diffractometer D5000-Siemens (Germany) in the interval of 5–115°. The morphology of the samples was assessed by SEM images taken on the FE-SEM Hitachi S-4800 (Japan) with 5 kV voltage acceleration. The thermal stability of the materials was examined using a Setaram (France) simultaneous differential thermal analyzer (DTA/DSC/TGA) apparatus (Labsys Evo S60/58988) under a 120 mL/min flow of argon atmosphere at a heating rate of 10°C/min from room temperature to 800°C.

3 Results and discussion

3.1 Physical appearance of mixed gel electrolytes

The results in Table 1 show the significant influence of additives on the physical appearance of fabricated samples. The state of the sample was found to exist in its normal form without solution separation at the PAM

Table 1: Physical appearance of mixed gel electrolyte containing different additives

Sample no	Content of additives in mixed gel electrolyte (wt%)			Physical appearance of mixed gel electrolyte
	PAM	PPG	NFS	
1	0.2	0.0		Normal gel
2	0.2	0.1		Normal gel
3	0.2	0.2		Solution splitting
4	0.2	0.3		Solution splitting
5	0.2	0.4		Solution splitting
6	0.2		0.2	Poor gel
7	0.2		0.4	Poor gel
8	0.2		0.6	Soft gel
9	0.2		0.8	Poor gel
10	0.2	0.1	0.2	Normal gel
11	0.2	0.1	0.4	Normal gel
12	0.2	0.1	0.6	Hard gel
13	0.2	0.1	0.8	Normal gel

content of 0.2 wt%. However, when adding PPG in combination with PAM, this state is only achieved if the PPG content does not exceed 1% by weight. The reason may be due to the hydrophobic nature of PPG [8], which can cause water to be easily separated from the gel if the PPG content exceeds 1 wt%.

Samples containing 0.2 wt% PAM and NFS (0.2–0.8 wt%) showed that the majority of the resulting products were generally less durable. One of them was found to be stable in a soft gel state for NFS at 0.6% by weight. When PPG (0.1 wt%) was added to them, the physical state of samples improved significantly. In which the state of the sample using 0.6 wt% NFS changed from a soft gel to a hard gel. This means that if the NFS is used inadequate (<0.6% by weight) or excess (>0.6% by weight) in the presence of PPG (0.1% by weight), then the hydrogen bonds in water in the Si–O–Si network will become loose, resulting in only normal gel formation and less stiffness.

3.2 Ionic conductivity

Figure 1(a–c) shows the Nyquist diagrams measured in different mixed gel electrolytes. An electrical equivalent circuit (EEC) was found (d) with five elements by simulation following the Thales Software [10]. These include L , R_Ω , C_d , R_{ct} , and W , which refer to the inductive element (relaxation impedance), the resistance of gel electrolyte, a double-layer capacitance, the resistance of charge transfer, and a Warburg diffusion element, respectively.

The fitting error less than 10% shows that the EEC is quite consistent with the experimental results.

The ionic conductivity (see Table 2) is calculated using Eq. 1, in which the resistance R_Ω is the intersection value between the Nyquist line and the horizontal axis with an error of less than 5%. Its value ranges from 0.40 to 0.56 S/cm, in which the lowest one (0.40 S/cm) belongs to the sample using ternary additive (PPG, PAM, and NFS with 0.1, 0.2, and 0.8 wt%, respectively), and the highest one belongs to the sample using binary additive (PAM/PPG (0.2/0.3 wt%)).

After studying the physical appearance and the ionic conductivity, four representative samples 1 and 2 (normal gel for single PAM and binary PAM/PPG additives, respectively), 8 (soft gel for binary PAM/NFS additive), and 12 (hard gel for PAM/NFS/PPG ternary additive) were selected for further study. The reason for this choice is that the samples should be gel like without solution separation. This is related to the ionic conductivity and diffusion process of HSO_4^- inside of the gel. Furthermore, it is very necessary because oxygen needs to be transported through the pores in the mixed gel electrolytes from the positive plate to the negative plate to recombine it into water (see reactions 3–6; Eq. 3–6) during a sealed lead acid battery charged [5,12].

The total reaction is as follows:

3.3 Diffusion coefficient of HSO_4^- ions

From observing the physical state of the electrolyte (see Table 1), four representative samples were selected to take CV curves for studying the diffusion of HSO_4^- ions in mixed gel electrolytes. Observing them in Figure 2 shows that oxidation peak appears with an increasing height as the scanning rate changes, but the position of the peak remains virtually unchanged (approximately –0.96 V). This peak shows that an oxidation occurs on the research electrode according to the following reaction:

Eq. 7 shows that the diffusion coefficient in Eq. 2 is considered for HSO_4^- ions. The results from Figure 3 and

Figure 1: Nyquist diagrams in different mixed gel electrolytes using: (a) various content of PPG adding PAM (constant content), (b) various content of NFS adding PAM (constant content), (c) various content of NFS adding PAM & PPG (constant content); and (d) electrical equivalent circuit (EEC).

Table 2: Determination of ionic conductivity of mixed gel electrolytes

Content of additives in mixed gel electrolytes (wt%)			R_{Ω} (Ω)		Ionic conductivity (S/cm)	Main physical state
PAM	PPG	NFS	Value	Error (%)		
0.2	0.0		11.11	3.58	0.47	Normal gel
0.2	0.1		9.93	3.59	0.52	Normal gel
0.2	0.2		9.43	3.67	0.55	Solution splitting
0.2	0.3		9.21	3.39	0.56	Solution splitting
0.2	0.4		10.44	3.76	0.50	Solution splitting
0.2		0.2	10.66	0.71	0.49	Poor gel
0.2		0.4	9.98	0.60	0.52	Poor gel
0.2		0.6	9.60	2.08	0.54	Soft gel
0.2		0.8	9.83	1.49	0.53	Poor gel
0.2	0.1	0.2	11.58	3.61	0.45	Normal gel
0.2	0.1	0.4	12.78	4.75	0.41	Normal gel
0.2	0.1	0.6	12.36	4.38	0.42	Hard gel
0.2	0.1	0.8	12.98	4.21	0.40	Normal gel

Figure 2: Voltammograms of the PbSb electrode at different scan rate in various mixed gel electrolytes using (a) 0.2 wt% PAM, (b) PAM and PPG (0.2 and 0.1 wt%, respectively), (c) PAM and NFS (0.2 and 0.6 wt%, respectively), and (d) PAM, PPG, and NFS (0.2, 0.1, and 0.6 wt%, respectively).

Figure 3: The linear plots of i_p vs $v^{1/2}$.

Table 3 also show that the height of the peak well linearly depends on the square root of the potential scanning speed because of R^2 in the range of 0.96–0.99. This reflects the electrochemical process that happens to be reversible. The diffusion coefficient D_o can be calculated from the slope K ($= 2.69 \times 10^5 n^{3/2} A D_o^{1/2} C_o$). In which n , A , and C_o are constants with a constant value, except D_o . The bigger the K is, the bigger D_o is. So instead of comparing the value of D_o , we can use the K value to compare the diffusion capacity of HSO_4^- ions in different electrolytes. The results showed that the lowest K value (0.1217 mol/s^{1/2}) belongs to the sample containing the single PAM (0.2 wt%) and the highest one (0.2625 mol/s^{1/2}) indicates the combination of ternary additive (PAM, NFS, and PPG with 0.2, 0.8, and 0.1 wt%, respectively). The results also showed that the diffusion capacity of HSO_4^- ions in the mixed gel electrolyte was improved better by adding NFS whether PPG is present. This is probably

Table 3: The parameters determined from the plots in Figure 3

Additives (wt%) in mixed gel electrolytes	Linear equations	R^2	K (mol/s ^{1/2})
PAM (0.2)	$y = 0.1217x - 0.0025$	0.9701	0.1217
PPG and PAM (0.1 and 0.2, respectively)	$y = 0.1858x - 0.0209$	0.9906	0.1858
PAM and NFS (0.2 and 0.6, respectively)	$y = 0.2463x - 0.0196$	0.9776	0.2463
PPG, PAM, and NFS (0.1, 0.2, and 0.6, respectively)	$y = 0.2625x - 0.0377$	0.9614	0.2625

due to the formation of the three-dimensional structure thanks to NFS.

3.4 FTIR studies

FTIR spectra shown in Figure 4 and data presented in Table 4 indicate that the peak at near 3,500 cm⁻¹ belongs to the Si-OH and O-H groups linked to molecular water hydrogen as well as the N-H group due to the presence

of PAM. The peak appears in interval 1,500–1,700 cm⁻¹ explains the stretching vibration of the SiO₂ network or the C=O and C-N resonances of PAM. The peak around 1,055 cm⁻¹ is assigned to the asymmetrical stretching oscillation of the SiO₂ network. The peak in interval 900–850 cm⁻¹ describes the symmetrical stretching oscillation of Si-O-Si bonds corresponding to a ring structure. The peaks are located at under 600 cm⁻¹ thanks to the bond bending vibration of Si-O-Si. The obtained data are agreed with the previous studies [13,14].

The results showed that the presence of PPG beside PAM (b) almost did not change the position as well as the intensity of adsorbed peaks compared without PPG (a). The intensity of the adsorption peak decreased by the presence of NFS next to PAM (c); however, the peak position did not seem to change. Especially in the presence of all of them (d), including PAM, PPG, and NFS, the adsorption peak intensity continued to decrease slightly. In which the peak representing the OH group (3,428 cm⁻¹) was shifted to a lower wavenumber compared with that in the case of the separated PAM (a, 3,442 cm⁻¹). Thus, a slight change in this peak is related to the composition used for the preparation of mixed gel electrolytes. However, in addition to this peak, there is a peak appearing at wavenumber above 1,600 cm⁻¹ (vibration of SiO₂ network) related to the ionic conductivity due to proton transfer in their three-dimensional network. This can be explained that the silica particles are exposed during gelling and bonding into a three-dimensional lattice in which the H⁺ ion can jump from an H₃O⁺ ion (formed during dissociation of H₂SO₄ in water) to a neighboring H₂O molecule as shown in Eq. 8 [15].

Figure 4: FTIR spectra of gelled electrolytes using (a) 0.2 wt% PAM, (b) PAM and PPG (0.2 and 0.1 wt%, respectively), (c) PAM and NFS (0.2 and 0.6 wt%, respectively), (d) PAM, NFS, and PPG (0.2, 0.6, and 0.1 wt%, respectively).

The presence of PPG together with NFS and PAM (d) has significantly reduced the peak intensity, reflecting less hydroxyl groups resulting in a decrease in ionic conductivity than the sample containing both NFS and

Table 4: Data of FTIR analysis from Figure 4 for mixed gel electrolytes

Wavenumber ν (cm^{-1})				Binding
(a)	(b)	(c)	(d)	
3,442	3,444	3,435	3,428	Si–O–H group, O–H group of PPG or residual water, N–H stretching of PAM
1,634	1,638	1,639	1,637	C=O and C–N resonance of PAM; combination of vibration of the SiO_2 network
1,055	1,055	1,055	1,058	O–Si–O (asymmetrical stretching oscillation of SiO_2 network)
881	883	882	882	Si–O–Si (symmetrical stretching oscillation to ring structure)
591, 459	584, 456	592, 459	592, 457	Si–O–Si (bond bending vibration)

PAM (c). It means that a small portion of NFS in this case would have been involved in the mechanical reinforcement process if it was used in conjunction with PPG.

3.5 X-ray diffraction

All the X-ray spectra in Figure 5 show a wide peak in the interval from 15° to 45° (see peak 1) corresponding to the presence of SiO_2 with the same amorphous structure similar to the previous studies [16,17]. An unclear peak appears at the near position of 11° (see peak 2) explains a contribution of PAM. However, this peak almost disappeared when adding PPG and PAM without the presence of NFS (b). The aforementioned results also show that the presence of additives did not affect the amorphous structure of silica.

Figure 5: X-ray spectra of gelled electrolytes using (a) 0.2 wt% PAM, (b) PAM and PPG (0.2 and 0.1 wt%, respectively), (c) PAM and NFS (0.2 and 0.6 wt%, respectively), (d) PAM, NFS, and PPG (0.2, 0.6, and 0.1 wt%, respectively).

3.6 SEM image analysis

Four samples, including numbers 1, 2, 8, and 12 representing different physical states (see Table 1), were selected for SEM imaging and comparing their morphological structure with each other. The results show that the presence of NFS in mixed gel electrolytes contributes to bigger material particle size than without NFS (see Figure 6). It can be explained by an agglomeration of the crystals thanks to the three-dimension structure formed by the presence of NFS. However, observing the SEM images found that all the samples contained crystalline particles in the nano-region (<50 nm).

3.7 Thermal analysis

The graphs shown in Figure 7 show three distinct weight loss intervals. The first stage below 100°C reflects desorption of physico-adsorbed water and evaporation of water from sulfuric acid inside the gel. The second stage was recorded at a temperature below 350°C , indicating a dehydroxylation process of silanol (Si–OH) groups [18] to form siloxane (Si–O–Si) bonding of polysiloxane. At higher temperatures, the material is further degraded by the reduced polysiloxane to SiO_2 , and it seems to stop at about 500°C . The data presented in Table 5 show the exothermic temperatures and the weight loss percent at 200°C , 350°C , and 800°C . From about 100 – 200°C , a first exothermic appearance is likely due to the formation of new cross-links in the three-dimensional network when the silanol bonds are broken [19]. In the period of less than 350°C , there was a second heat emission, indicating the decomposition of functional groups of organic additives [20]. The percentage reduction in weight of the gel sample using ternary additive (PAM/PPG/NFS) was the least, whereas the rest of the samples lost not only more weight but also the same degree of reduction. According to the previous study [9], the more the weight of material loses, the more the gel become durable and porous. This

Figure 6: SEM images of mixed gel electrolytes containing different additives.

Figure 7: TGA curves of mixed gel electrolyte using different additives.

explained that the gel sample using all three additives formed with a three-dimensional structure less porous than using the single PAM and binary additives (PAM/PPG or PAM/NFS). This is consistent with the results of the SEM image analysis and conductivity measurement in the previous sections.

Table 5: Some parameters in thermal analysis of mixed gel electrolytes from Figure 7

Parameter	(a)	(b)	(c)	(d)
$T_{\text{Exotherm}} (^{\circ}\text{C})$	108.41	111.75	116.67	124.16
$m_{\text{loss at } 200^{\circ}\text{C}} (\%)$	45.32	43.02	41.21	37.37
$m_{\text{loss at } 350^{\circ}\text{C}} (\%)$	57.36	56.38	56.46	47.25
$m_{\text{loss at } 800^{\circ}\text{C}} (\%)$	69.38	66.90	68.36	51.32

3.8 Gas release on PbSb electrode

The main idea in studying CV spectra in this section is to consider the escape of gas, including hydrogen and oxygen, which occurs at two potential ranges from -1.2 to -1.5 V and from 1.6 to 2.1 V, respectively. First, we see that this gas release takes place strongly in sulfuric acid solution (see Figure 8e), especially the peak of oxygen release reaching 200 mA/cm^2 at the 120th cycle. In addition, we can see that the more the scanning, the higher the rate of oxygen release.

Figure 8: CV curves of the PbSb electrode at 100 mV/s in different mixed gel electrolytes using (a) PAM (0.2 wt%), (b) PAM and PPG (0.2 and 0.1 wt%, respectively), (c) 0.2 wt% PAM adding 0.6 wt% NFS, (d) PAM, PPG, and NFS (0.2, 0.1, and 0.6 wt%, respectively) compared with that in (e) 1.26 g/cm³ H₂SO₄ solution.

In contrast, in mixed gel electrolyte (see Figure 8a–d), the release of gas is much lower (for hydrogen <40 mA/cm² and oxygen <15 mA/cm²) at the 120th cycle comparing with those in H₂SO₄ solution (90 and 200 mA/cm², respectively, Figure 8e). It indicates that more than 90% of

oxygen and 55% of hydrogen are inhibited thanks to mixed gel electrolytes. The amount of electricity (see Table 6) required to release oxygen in mixed gel electrolytes is generally much lower than in acidic solutions. At the 120th cycle, it requires only 354 μ C for oxygen release

Table 6: Amount of electricity (in μC) for oxygen release in different electrolytes for various CV numbers determined from Figure 8

Number of CV	Mixed gel electrolyte				Acid sulfuric solution (E)
	(A)	(B)	(C)	(D)	
1	795.3	338.4	686.2	809.6	2503.0
30	233.0	267.6	280.4	335.7	1135.0
60	237.8	280.0	310.5	357.3	1924.0
90	301.5	322.2	332.3	392.7	3843.0
120	366.9	366.0	354.0	438.4	5612.0

in mixed gel electrolytes containing PAM and NFS (0.2% and 0.6% by weight, respectively), much lower compared to that in the H_2SO_4 solution (5,612 μC). It clearly explains that 93.7% oxygen was inhibited. This is very beneficial when using mixed gel electrolytes in the manufacturing of sealed lead acid batteries.

In addition, we can see that the height of peak for oxygen release sharply decreases in mixed gel electrolytes from about 40 mA/cm^2 to less than 15 mA/cm^2 after the first cycle, and it is almost stable in subsequent cycles. Reducing the rate of gas release, especially oxygen (see Eq. 9), will reduce water loss, so the corrosion of electronic devices around the battery will also decrease as the acid vapors are controlled.

This suggests that the mixed gel electrolytes are beneficial in the manufacturing of sealed lead acid batteries because the pressure in the battery is reduced and requires less maintenance due to limiting water loss.

4 Conclusion

The mixed gel electrolytes have been synthesized using a number of additives (PAM, NFS, and PPG). They were characterized by observing their physical shape and electrochemical measurements (EIS and CV) as well as some physicochemical methods (FTIR, X-ray, SEM, and TGA). The outstanding results of this study are as follows: (i) using the ternary additives (PAM/PPG/NFS) provided a mixed hard gel electrolyte with the three-dimensional network structure in the range of nano size, which improved the diffusion of HSO_4^- ion inside the gel; (ii) the mixed gel electrolyte contributes to the reducing gas release (92.2–93.7% O_2 and ~55% H_2 at 120th CV) on the PbSb electrode, which is beneficial for making the sealed lead acid battery.

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