



# Ionic liquid as an electrolyte additive for high performance lead-acid batteries

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## HIGHLIGHTS

- The performance of lead-acid battery is improved using ionic liquid (EMIDP).
- EMIDP suppress  $H_2$  gas evolution to very low rate  $0.049 \text{ ml min}^{-1} \text{ cm}^{-2}$  at 80 ppm.
- The battery capacity increases from  $45 \text{ mAh g}^{-1}$  to  $83 \text{ mAh g}^{-1}$  by adding EMIDP.
- SEM-EDX analysis confirms the adsorption of EMIDP on the battery electrode surface.

## ARTICLE INFO

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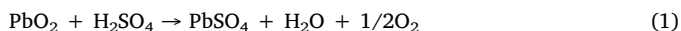
## ABSTRACT

The performance of lead-acid battery is improved in this work by inhibiting the corrosion of negative battery electrode (lead) and hydrogen gas evolution using ionic liquid (1-ethyl-3-methylimidazolium diethyl phosphate). The results display that the addition of ionic liquid to battery electrolyte (5.0 M  $H_2SO_4$  solution) suppresses the hydrogen gas evolution to very low rate  $0.049 \text{ ml min}^{-1} \text{ cm}^{-2}$  at 80 ppm. Electrochemical studies show that the adsorption of ionic liquid molecules on the lead electrode surface leads to the increase in the charge transfer resistance and the decrease in the double layer capacitance. I also notice a noteworthy improvement of battery capacity from  $45 \text{ mAh g}^{-1}$  to  $83 \text{ mAh g}^{-1}$  in the presence of ionic liquid compound. Scanning electron microscopy and energy dispersive X-ray analysis confirm the adsorption of ionic liquid molecules on the battery electrode surface.

## 1. Introduction

The lead-acid battery has been a successful article of commerce for over a century [1]. Lead-acid batteries are successfully used in many applications [2]. Its manufacture and use continue to develop because of new applications for battery power in energy storage.

The lead-acid battery uses  $PbO_2$  as the active material of the positive electrode and metallic Pb as the negative active material [3]. Pb and  $PbO_2$  are thermodynamically unstable in battery electrolyte (5.0 M  $H_2SO_4$  solution) [4]. Oxygen is evolved at the positive electrode and hydrogen at the negative electrode as follows [4]:



The corrosive decay of the negative electrode and hydrogen gas evolution are the most frequent causes of failures of lead-acid batteries [5]. These drawbacks limit its application in large-scale batteries. An attractive approach to circumvent these drawbacks is to use organic

additives in the electrolyte [6–8]. During the past few years, many works have focused on finding a suitable additive to improve the performance of lead-acid batteries [9–12].

Traditional organic additives such as derivatives of benzaldehyde [13], phosphoric acid [14] and amino acids [15], are generally investigated in the literature. The organic additives are affecting the hydrogen gas evolution on Pb by shifting the hydrogen over-voltage to a more negative electrode potential. The primary stage in the suppression of hydrogen evolution in the presence of organic additives is the adsorption of additives on cathodic sites on the Pb surface.

Many former works proposed various surfactants for lead-acid batteries, which greatly promoted their performance through the effect on both hydrogen evolution and the metal corrosion [16]. According to Ghavami the effects of surfactants on the batteries performance based on their concentration and negative or positive charged head groups of surfactants [17].

Some new materials, such as rare earth metals were widely investigated to increase the batteries performance. The main role of rare earth metals is the decrease the resistance of the anodic film, and

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improves the corrosion resistance of Pb electrode [18].

The challenge in using organic additives to improve the performance of lead-acid batteries lies in finding organic compounds that provide good corrosion inhibition, nature-friendly compounds, a long lifespan and a competitive price.

Generally, rare works have been performed on the use of ionic liquids as additives to improve the performance of lead-acid batteries.

Yang et al. [19] prepared a series of composites containing ionic liquid-functionalized graphene oxide to improve the performance of anion exchange membranes for fuel cells.

Salvador et al. [20] found that the gelation of ionic liquid-based solutions with inorganic or organic fillers improves the performance of dye-sensitized solar cells. In this case, the ionic liquid volume ratios represent the key parameters.

Nair et al. [21] demonstrated that the newly elaborated polymer electrolytes that contain a room-temperature ionic liquid represent a promising potential of the materials in storage devices.

Bella et al. [22] suggested that the ionic liquid electrolyte based on iodide/triiodide redox mediator and nanosized silica particles has potential application to prepare highly stable dye-sensitized solar cells.

Nair et al. [23] reported the use of ionic liquid in the preparation of truly solid polymer electrolyte membranes for lithium polymer batteries.

Here we study the influence of 1-ethyl-3-methylimidazolium diethyl phosphate (EMIDP) on the corrosive deterioration of the negative electrode (metallic Pb) and hydrogen gas evolution during the discharge of the lead-acid battery.

EMIDP is imidazolium based ionic liquid containing diethylphosphate as anion (see Fig. 1). Recently, the use of ionic liquids in batteries is receiving increasing attention due to their eminent properties; in addition, they have very low environmental impacts [24]. Therefore, this study offers a new strategic approach to improve the performance of lead-acid battery using ionic liquid as electrolyte additives.

## 2. Experimental methods

### 2.1. Materials

The working electrode was designed from pure lead 99.99%. The dimensions of lead pieces that used for H<sub>2</sub> evolution measurements are nearly 3.0 cm × 1.0 cm × 0.2 cm. For electrochemical tests, the cylindrical stripe of Pb with surface area of 0.502 cm<sup>2</sup> was prepared.

The cleaning of working electrodes before the testes were carried out according to ASTM G1 - 03(2017)e1 [25].

1-ethyl-3-methylimidazolium diethyl phosphate (EMIDP) (98%) and sulfuric acid (98%) were obtained from Sigma-Aldrich.

### 2.2. H<sub>2</sub> evolution measurements

The experimental setup for H<sub>2</sub> evolution measurements was

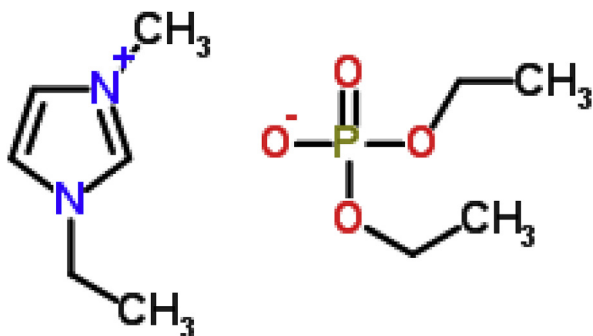


Fig. 1. The molecular structure of 1-ethyl-3-methylimidazolium diethyl phosphate (EMIDP).

described in earlier studies [26]. The volume of the H<sub>2</sub> evolved ( $\Delta V$ ) was determined during the immersion of stripes of Pb in 100 ml 0.5 M H<sub>2</sub>SO<sub>4</sub> solution for 3 h. The rate of hydrogen gas evolved ( $R_{H_2}$ ) during the immersion can be calculated from the following relation [27]:

$$R_{H_2} = \frac{\Delta V(\text{ml})}{\text{immersion time}(\text{min}) \times \text{electrode surface area}(\text{cm}^2)} \quad (3)$$

To get good reproducibility, the measurements are repeated three times. The data obtained are represented using the means ( $\bar{X}$ ) and the standard deviations ( $\sigma$ ) and standard error of the mean (SE).

### 2.3. Electrochemical testing

EIS tests were carried out using a double wall glass cell containing 0.5 M H<sub>2</sub>SO<sub>4</sub> solution as the electrolyte, with a Pt counter electrode and Hg/Hg<sub>2</sub>SO<sub>4</sub> reference electrode. Potentiostat with trade name Gill AC from 947 ACM instruments Company was used to recode all EIS tests. The suggested equivalent circuit and EIS parameters were estimated using Sequencer software. The EIS experiments were carried out by changing the frequency of 5 mV sinusoidal voltage from 30 kHz to 0.01 Hz.

The sensitivity of measuring devices depends on the percent error (Error %). It measures the accuracy of a measurement by the difference between experimental and theoretical values.

### 2.4. Battery test

The battery performance tests were performed using laboratory-made lead-acid cells (2 V/2.8 Ah). This cell consists of one negative, two positive electrodes and poly (vinyl chloride) (PVC) as separator sheets. The battery electrolyte is 5.0 M H<sub>2</sub>SO<sub>4</sub>. The discharge tests were conducted in the absence and presence of additives.

### 2.5. Surface analysis

SEM-EDX analysis was used to study the surface morphology of battery electrode (Pb) after complete discharging time using scanning electron microscopy (JEOL: JSM6510) with an X-ray microanalysis (EDX).

## 3. Results and discussion

### 3.1. The effect of EMIDP on the hydrogen gas evolution

The performance of different concentrations EMIDP on the hydrogen gas evolution during the immersion of Pb electrode in 5.0 M H<sub>2</sub>SO<sub>4</sub> solution was examined by measuring of the rate of hydrogen gas evolved ( $R_{H_2}$ ) with the results shown in Table 1.

The  $R_{H_2}$  results in Table 1 indicate that the EMIDP has a suppression effect on the hydrogen gas evolution. The  $R_{H_2}$  will depend on EMIDP

**Table 1**  
The rate of hydrogen gas evolved ( $R_{H_2}$ ) and the efficiency of EMIDP ( $I_{H_2}\%$ ) during the immersion of Pb electrode in 5.0 M H<sub>2</sub>SO<sub>4</sub> solution without and with EMIDP compound at 298 K.

Solutions	$R_{H_2}$ (mL min <sup>-1</sup> cm <sup>-2</sup> ) $\bar{X} \pm \sigma$	$I_{H_2}\%$
Blank	0.754 ± 0.004	–
20 ppm EMIDP	0.510 ± 0.003	32.36
40 ppm EMIDP	0.360 ± 0.005	52.25
60 ppm EMIDP	0.174 ± 0.003	76.92
80 ppm EMIDP	0.049 ± 0.002	93.50
100 ppm EMIDP	0.052 ± 0.003	93.10
120 ppm EMIDP	0.057 ± 0.002	92.44

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