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WEIGHT LOSS EFFECT AND POTENTIODYNAMIC POLARIZATION RESPONSE OF 1-BUTYL-3-METHYLIMIDAZOLIUM CHLORIDE IONIC LIQUID IN HIGHLY ACIDIC MEDIUM

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Abstract

Ionic liquids are increasingly being used as corrosion inhibitors when oil and gas industries started to give focus on sustainability and green impact in their operations. In this study, 1-butyl-3-methylimidazolium chloride ionic liquid in 2M HCl medium has been investigated on mild steel, stainless steel and aluminium bars by using weight loss technique and potentiodynamic polarization measurement. Results showed that 1-butyl-3-methylimidazolium chloride is able to reduce the weight loss of aluminium metal under acidic corrosive surrounding up to 11% compared to that of without the presence of 1-butyl-3-methylimidazolium chloride. Based on potentiodynamic polarization response, percentage of corrosion inhibition efficiency is found to be up to 99.3%. In summary, 1-butyl-3-methylimidazolium chloride is highly potential to act as an anti-corrosion agent, even in a very low concentration.

Keywords: Ionic liquid, Corrosion, Mild steel, 1-butyl-3-methylimidazolium chloride.

1. Introduction

Most manufacturing industries and industrial processes depend largely on pipelines for transporting feeds, intermediate or final products between process

Nomenclatures

c	Concentration of ionic liquid, ppm
E_{corr}	Corrosion potential, mV
I	Corrosion current density in the absence of ionic liquid, mA cm^{-2}
I_{IL}	Corrosion current density in the presence of ionic liquid, mA cm^{-2}
IE_{corr}	Corrosion inhibition efficiency, %
m_L	Metal weight loss, %
R	Corrosion rate in the absence of ionic liquid, g cm^{-2}hr^{-1}
R_{IL}	Corrosion rate in the presence of ionic liquid, g cm^{-2}hr^{-1}

Greek Symbols

β	Electrochemical polarization slope, mV dec^{-1}
θ	Surface coverage, deg.

Abbreviations

IFT	Interfacial tension
SCE	Saturated calomel electrode
SiC	Silicon carbide

equipment. In particular, for oil and gas industries, pipeline is the heart of transportation process. For example, Russia being the second largest producer of dry gas and the third largest liquid fuels producer in the world is having the world most extensive oil and gas pipeline network at 222,000 km and additional of 20,923 km more by 2018 [1]. However, corrosion is a major threat encountered by pipeline operators [2]. One of the consequences of corrosion phenomenon is the loss in weight of metallic materials. Weight loss of metal leading to loss of mechanical strength that later will result in mechanical structure breakdown. It can also leads to pipeline, pump compressor or vessel blockage due to corrosion precipitates or precipitation that resulted as part of structure breakdown or during the weight loss process [3]. Furthermore, weight loss can lead to pipeline perforation which will make the content inside the pipe escapes and hence can cause an explosion [4]. In oil and gas industries, pipe explosion caused by perforation is very dangerous as it can results in human fatalities [5].

In offshore oil industry, steel jackets have been widely used to support oil platforms. Although jacket structures are required to have a design service life up to 20 years, they are subjected to structural aging [6]. In structural aging, corrosion plays a major role especially when the jacket is exposed to extreme environmental conditions like wave, wind and current loads. The overall strength will degrade with the corrosion growth causing risk in collapse and failure of the whole jacket. Ultimately, this will result in loss of people, property and environment pollution. Therefore, the main motivation for this study is safety.

Prevention methods for corrosion are anodic and cathodic protections, protective coating method and by using corrosion inhibitors [7-11]. Corrosion inhibitors protect metal surface against corrosive attack via (i) physisorption and (ii) chemisorption mechanisms. Mechanism of these adsorptions of heteroatom-containing corrosion inhibitors; such as O, N, S and P, on metal surface depends mainly on physicochemical properties of the inhibitor group, such as electron density at the donor atom (heteroatom), π-orbital character and the electronic structure of the molecule [12].

Up to date, ionic liquid has been reported to impede weight loss of metallic material under corrosive surrounding. Ionic liquids are defined as organic salts that melt below room temperature or melt below 100^{o}C. The interest in ionic liquids stems from their potential as 'green solvent' because of their chemical and thermal stabilities, non-flammability, have very low or negligible vapour pressure, high ionic conductivity and a wide electrochemical potential window [13, 14]. Studies have shown that imidazolium-based ionic liquids were able to prevent corrosion from occurring on copper [15, 16], mild steel [17, 18] and aluminium [19] samples.

The mechanism for imidazolium compound to hinder corrosion on metal surfaces is similar to corrosion inhibitors mechanism due to the presence of heteroatom-containing, N atoms, and the π-orbital character as described earlier. Furthermore, the presence of multiple active sites (2N atoms) can be the reason of why imidazolium-based ionic liquid able to form protective layer on metal surfaces thus retards further corrosion. For offshore structures application, typical parts of corrosion attack such as on surface equipment and steel structures are exposed to splash zone and tidal zones (high tide zone and low tide zone). In such cases, ionic liquid is used as electroplating medium during aluminium coating process of the structures [20] or as additive to coating formulation (protective coating method). These inhibitors generally control corrosion by forming synergistic films that modify the degree of corrosivity at the metal surface.

Inhibitors form synergistic films in several ways: by adsorption, the formation of bulky precipitates and/or the formation of a passive layer on the metal surface. Some inhibitors retard corrosion by adsorption to form a thin, invisible film only a few molecules thick. Others form bulky precipitates that coat the metal and protect it from attack. A third mechanism consists of causing the metal to corrode in such a way that a combination of adsorption and corrosion product forms a passive layer. However, there is a lack of published information related to the challenge and limitation of using ionic liquid in the offshore structures application [21]. New technologies are needed since corrosion is such a complex topic, knowledge deficits exist and require additional research [22].

Shi et al. [23] investigated the relationship between corrosion inhibition efficiency and alkyl group which connected to nitrogen atom. They reported that the corrosion inhibition efficiency was increased with increasing carbon chain length of the alkyl group. Zhang et al. [24] conducted another study on the same effect; using the butyl, hexyl and octyl groups; upon aluminium sample in acidic medium. They found an agreement with the former investigation and reported that the corrosion inhibition efficiency is in the order: octyl > hexyl > butyl. This study aims to determine the effect of 1-butyl-3-methylimidazolium chloride ionic liquid on mild steel, stainless steel and aluminium bar in acidic media. The effects are presented in terms of weight loss percentage and potentiodynamic polarization response.

2. Materials Used and Experimental Procedures

We used 1-butyl-3-methylimidazolium chloride (Merck Millipore, CAS No. 79917-90-1) for the ionic liquid in this study with molecular formula of

$C_8H_{15}ClN_2$ and properties of molar mass: 174.68 g/mol and solubility: soluble (20^{o}C). Figure 1 shows the molecular structure of 1-butyl-3-methylimidazolium chloride comprises an imidazole ring, N(1) atom attached to the butyl group and N(3) atom attached to the methyl group. The counteranion is chloride ion. Distilled water was used in all cases.

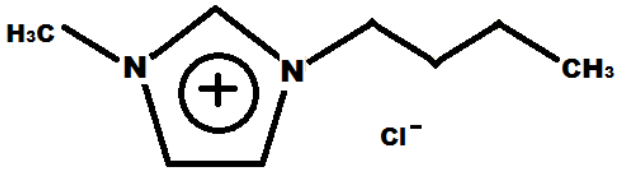


Fig. 1. Molecular structure of 1-butyl-3-methylimidazolium chloride.

Prior to all measurements, the mild steel (C: 0.19%, Si: 0.26%, Mn: 0.64%, S: 0.05%, P: 0.06%, Cr: 0.08%, Mo: 0.02, Cu: 0.27% and the remainder iron), stainless steel, AISI 304 (C: 0.08%, Si: 1.00%, Mn: 2.00%, Ni: 8.00%, Cr: 18.00% and the remainder iron) and aluminium, AL1350 (Fe: 0.40%, Si: 0.10%, Zn: 0.05%, Cu: 0.05%, Cr: 0.01%, C: 0.01% and the remainder aluminium) metal bars were polished mechanically on a polishing machine using SiC foils from 220 to 1000 grit. The metal bars were washed thoroughly with distilled water, cleaned with acetone and dried in air.

Immersion test was performed on the sample bars with a rectangular form (50mm×25mm×1mm) in 2 M HCl solution, with and without the addition of different concentrations of 1-butyl-3-methylimidazolium chloride ionic liquid. Each sample bar was weighed using an electronic balance (Shimadzu AY220, σ≤0.1 mg), suspended by using nylon thread through a hole and then was immersed in a 400 mL acid solution. The sample bar was removed after 24 h for corrosion rate determination. After that, the surface of the sample bar was cleaned with 5% HCl solution and the hard deposits were removed by a soft wire brush. The sample bar was rinsed with distilled water and air dried. For each experiment, a freshly prepared solution was used, the immersion test was performed three times and average values are used to calculate the corrosion rate. The corrosion rate, *R* is determined by using Eq. (1):

$$R\ (g.cm^{-2}hr^{-1}) = \frac{(weight\ loss\ in\ grams)}{(area\ in\ squared\ cm)(time\ of\ immersion\ in\ hour)} \quad (1)$$

Potentiodynamic polarization measurements were carried out using Volta lab (PGP 201) electrochemical work station and controlled by corrosion analysis software model (Voltamaster 4). Electrochemical experiments were performed in a standard electrochemical cell at 303 K under atmospheric condition, with a saturated calomel electrode (SCE) electrode as a reference electrode, a platinum electrode as an auxiliary electrode and a specimen bar as a working electrode. Potentiodynamic polarization studies were performed with a scan rate of 1 mVs^{-1} and all potentials were recorded with respect to the SCE.

3. Results and Discussion

3.1. Weight loss effect

Effect of 1-butyl-3-methylimidazolium chloride ionic liquid at different concentrations on mild steel, stainless steel and aluminium bar in 2 M HCl solution is shown in Fig. 2(a). In the absence of the ionic liquid, weight loss is found to be 93%, 88% and 37% for mild steel, stainless steel and aluminium, respectively. Since the 2 M HCl solution is corrosive, the reduction of the total mass of material make up the sample is caused by the interaction between the corrosive medium and the sample surfaces. At 6,250 ppm, the weight loss is reduced to 37%, 40% and 28% for mild steel, stainless steel and aluminium, respectively.

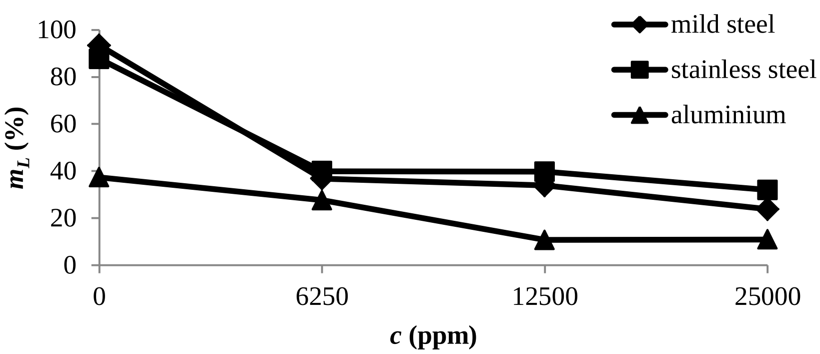
The study found that increasing the ionic liquid concentration doubled from its initial values gave no significant change on the weight loss. At the highest ionic liquid concentration of 25,000 ppm, the weight loss is 24%, 32% and 11% for mild steel, stainless steel and aluminium, respectively. Based on the highest calculated total mass reduction in percent, the samples are ranked as follows: stainless steel > mild steel > aluminium. The weight loss percent is elucidated by calculating the percentage of corrosion inhibition efficiency. Herein, Eq. (2) [25] is used:

$$IE_{corr} = \frac{R - R_{IL}}{R} \times 100 \quad (2)$$

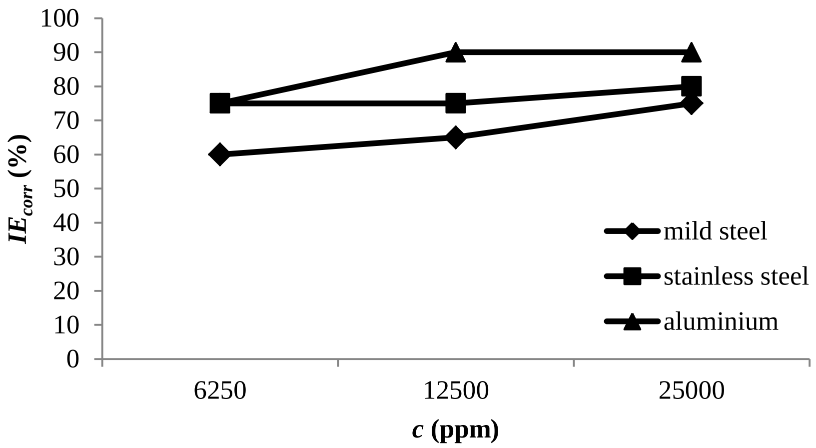
where R and R_{IL} are the corrosion rates ($g.cm^{-2}hr^{-1}$) in the absence and presence of the ionic liquid, respectively.

Figure 2(b) showed the IE_{corr} versus 1-butyl-3-methylimidazolium chloride concentration. It is found that the highest anti-corrosion behaviour is shown by aluminium, which is 90% at 12,500 ppm and 25,000 ppm. Mild steel showed the lowest IE_{corr} which is 60% at 6,250 ppm. Degree of 1-butyl-3-methylimidazolium chloride adsorption on metal surface is reflected by the magnitude of gradient of the lines. Magnitude of gradient for stainless steel is 3 while the magnitude of gradient for mild steel and aluminium is 8. Since the magnitude of all gradients is more than zero, this means that the degree of adsorption of 1-butyl-3-methylimidazolium chloride is sensitive on concentration. Different intensification of the magnitude of gradient shown in Fig. 2(b) suggests that the adsorption of 1-butyl-3-methylimidazolium chloride concentration is sensitive upon different type of metal surface.

When imidazolium-based ionic liquids adsorption behaviour was investigated [26] by using interfacial tension (IFT) measurement, it was found that the IFT of the system (n-butyl acetate + water system) significantly decreased due to the imidazolium-based ionic liquid excellent surfactant nature and hydrophobicity. IFT reductions were up to 75%. For modeling, the Szyszkowski adsorption equation was used to reproduce the experimental data. The consistent fittings have confirmed the ideal adsorption of the ionic liquid molecules at the interface. In summary, 1-butyl-3-methylimidazolium chloride had shown an anti-corrosion property with IE_{corr} higher than 60% on mild steel, stainless steel and aluminium bars.



(a) Metal weight loss.



(b) Corrosion inhibition efficiency.

Fig. 2. The effect of 1-butyl-3-methylimidazolium chloride on stainless steel, mild steel and aluminium bar at different concentrations according to weight loss and corrosion inhibition efficiency methods.

Langmuir adsorption isotherm was employed in order to evaluate experimental data from this study and to understand the mechanism of corrosion inhibition of metals. Assuming that the adsorption of 1-butyl-3-methylimidazolium chloride was mainly due to a monolayer adsorption and ignoring the interaction between the adsorbed molecules, then the Langmuir adsorption isotherm can be employed. The surface coverage (θ) was calculated as *IE/100* for the different concentrations of 1-butyl-3-methylimidazolium chloride. When $\log \theta/(1-\theta)$ vs $\log c$ was plotted (as in Fig. 3), a straight line was obtained which revealed the relationship between the fraction of the surface covered and the concentration of 1-butyl-3-methylimidazolium chloride. The linear correlation coefficient was used to choose the isotherm that fits the best of the experimental data. It should be noticed that the data fit the straight line with linear correlation coefficient higher than 0.999 indicates that 1-butyl-3-methylimidazolium chloride adsorb on mild steel, stainless steel and aluminium bar according to the Langmuir adsorption isotherm [27, 28]. The θ values obtained from the weight loss technique are in good agreement and all obey the Langmuir adsorption isotherm.

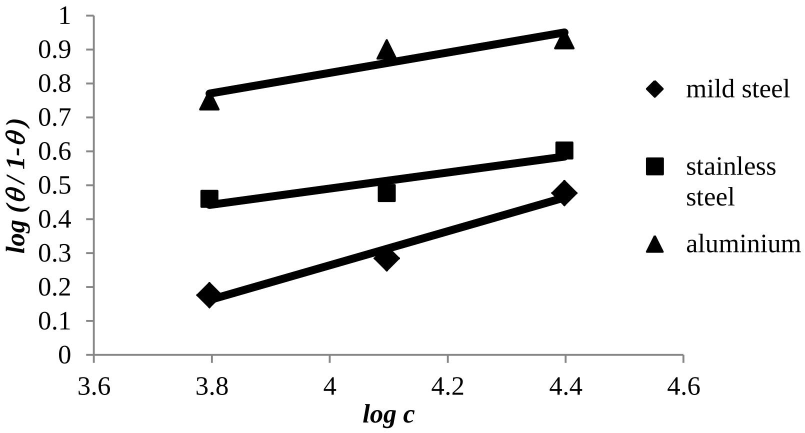


Fig. 3. Langmuir isotherm for the adsorption of 1-butyl-3-methylimidazolium chloride on stainless steel, mild steel and aluminium bar surfaces by using surface coverage values calculated by weight loss result.

3.2. Potentiodynamic polarization response

Potentiodynamic polarization curves for mild steel, stainless steel and aluminium at different ionic liquid concentrations are shown in Figs. 4(a)-(c). It could be observed that the presence of ionic liquid caused more negative shift in corrosion potential (E_{corr}) especially at 25000 ppm. The corrosion potential (E_{corr}), corrosion current density (I) obtained from the Tafel extrapolation of the polarization curves, and both the cathodic and anodic slopes (β's) are given in Table 1. The IE_{corr}% (Table 2) from potentiodynamic polarization response was calculated by using Eq. (3) [29]:

$$IE_{corr} = \frac{I - I_{IL}}{I} \times 100 \quad (3)$$

where I and I_{IL} are the corrosion current density without and with addition of ionic liquid, respectively. In general, the ionic liquid had shown an anti-corrosion property with IE_{corr} in the range of 41.7 – 99.3% on mild steel, stainless steel and aluminium bar by using potentiodynamic polarization measurement. The calculated results in Table 2 also showed that when ionic liquid concentration is increasing, the IE_{corr} increases whereas the I_{IL} decreases. In the basis of ionic liquid adsorption ability on metal surfaces, the adsorption process is enhanced at higher ionic liquid concentration. These results are in good agreement with those obtained from the weight loss technique. Among the two tests conducted, the difference of the obtained IE_{corr} for each concentration is no more than 18%. This indicates the stability of the ionic liquid to protect the metal surface. When compared to previous studies, the ionic liquid showed similar anti-corrosion property with IE_{corr} of 81.9% on aluminium [24] and 85.9% on mild steel [30].

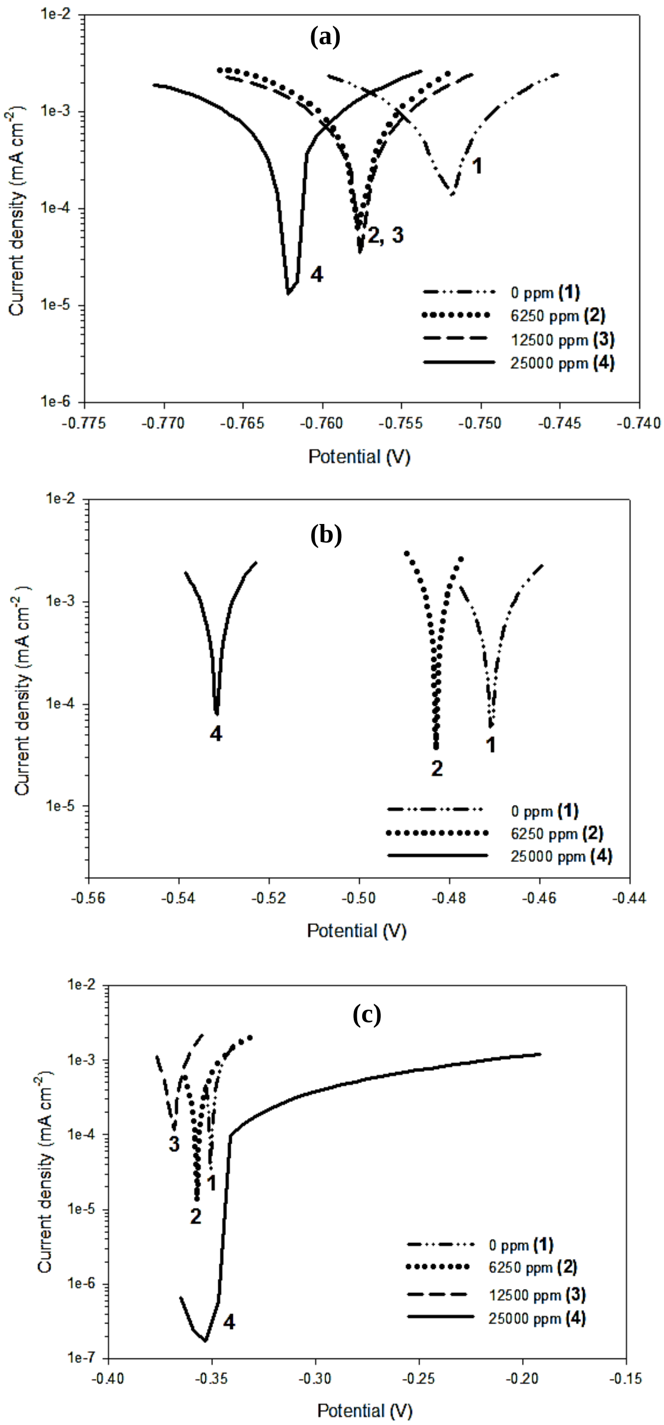


Fig. 4. Effect of 1-butyl-3-methylimidazolium chloride on the potentiodynamic polarization response for (a) aluminium bar, (b) mild steel, and (c) stainless steel.

Table 1. Electrochemical polarization parameters in the absence and presence of 1-butyl-3-methylimidazolium chloride.

Sample	c (ppm)	$-E_{corr}$ (mV)	$-\beta_c$ (mV dec^{-1})	β_a (mV dec^{-1})
aluminium	0	752.0	28.3	43.3
	6250	757.9	60.0	36.0
	12500	757.7	29.2	27.2
	25000	762.0	21.9	36.5
mild steel	0	471.0	84.0	42.0
	6250	483.0	146.5	196.5
	25000	531.5	32.8	164.0
stainless	0	357.0	28.8	43.3
steel	6250	369.0	166.2	249.3
	12500	359.0	54.0	162.0
	25000	-	0.0094	1.0514

Table 2. The corresponding corrosion inhibition efficiencies in the absence and presence of 1-butyl-3-methylimidazolium chloride.

Sample	c (ppm)	i_{corr} (mA cm^{-2})	IE_{corr} (%)
aluminium	0	15.0	*n.a*
	6250	6.0	60.0
	12500	3.7	75.3
	25000	1.5	90.0
mild steel	0	6.0	*n.a*
	6250	3.5	41.7
	25000	0.8	86.7
stainless	0	3.5	*n.a*
steel	6250	1.5	57.1
	12500	1.4	60.0
	25000	0.0240	99.3

4. Conclusions

The presence of 1-butyl-3-methylimidazolium chloride in 2 M HCl solution prevents weight loss of steel/metal up to 11%. Based on potentiodynamic polarization responses, 1-butyl-3-methylimidazolium chloride was found to have up to 99.3% corrosion inhibition efficiency. The percentage of corrosion inhibition efficiency increases when the 1-butyl-3-methylimidazolium chloride concentration is increased. The adsorption of the 1-butyl-3-methylimidazolium chloride molecules on the steel/metal surface from 2 M HCl solution obeys Langmuir adsorption isotherm. In summary, 1-butyl-3-methylimidazolium chloride showed good corrosion inhibition property even at a very low concentration.

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