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# Some Imidazole Derivative Compounds and the Investigation of Their Corrosion Inhibition Activities toward Carbon Steel in 1% NaCl Solution Utilizing Tafel Method

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## Abstract

Four imidazole derivative compounds: imidazole, compound 1 (2,4,5-triphenyl-1*H*-imidazole), 2 (2-hexyl-4,5-diphenyl-1*H*-imidazole) and 3 ((*E*)-2-ethyl-4,5-diphenyl-1-(prop-1-enyl)-1*H*-imidazole) have been synthesized utilizing microwave assisted organic synthesis (MAOS) method, in order to investigate their corrosion inhibition mechanism on carbon steel surface. The determination of corrosion inhibition activities of the compounds utilized Tafel plot method. Based on the analysis of Tafel plot data, there was linearity of each Langmuir isotherm adsorptions of each compound, which represent the monolayer formation of each compound on the carbon steel surface. The free Gibbs adsorption energy values,  $\Delta G^0_{ads}$ , for Imidazole, compound 1, 2 and 3 are -28.01, -34.29, -31.68 and -33.26 kJ/mol, respectively, which indicated the spontaneity of adsorption process of each compound on carbon steel surface and

also have the potent to interact with carbon steel through semi-physiosorption or semi-chemisorption.

**Keywords:** imidazole; Langmuir isotherm adsorption; Microwave Assisted Organic Synthesis; Tafel plot; corrosion inhibition activities.

## Introduction

The corrosion process of carbon steel pipelines in gas and oilfield mining induced by the presence of carbon dioxide and water has been a serious problem in the oil and gas industry for decades [1]. One of the most effective ways to protect the corrosion, especially the internal parts of carbon steel pipelines, caused by carbon dioxide and other acidic media is the use of organic corrosion inhibitors [1][2][3][4]. The common organic corrosion inhibitors that widely and commercially used to minimize the corrosion induced by carbon dioxide and acidic media in the gas and oilfield industries are imidazoline derivative compounds. Recently, the studies concerning the correlation between the structure of organic corrosion inhibitors and their corrosion inhibition ability have been quite numerous. There are several studies involving the effect of oxygen, nitrogen and sulphur containing compounds, acyclic and heterocyclic, towards the corrosion inhibition ability [5][6][7][8].

In Indonesia, the gas and oilfield mining is still one of important industries that would give valuable commodity and income to the nation. Therefore, the quest of better performance of organic corrosion inhibitor would be quite crucial considering the effectiveness of production cost. This work was contributed to this sort of quest as well as to the most essential purpose in practicing science methodology. The purpose of this work mainly is to synthesize some imidazole derivative compounds utilizing the novel method in organic synthesis, microwave assisted organic synthesis (MAOS). Many scientists have been engaged in a vast of disciplines that have applied the rapid heating associated with microwave technology to a number of useful processes [9][10][11][12]. This technology opens up new opportunities to the synthetic chemist, in the form of new reactions that are not possible using conventional heating, improved reaction

yields, decreased reaction times and even solvent free reaction conditions [10].

The other aim of this work is to analyze the correlation between the synthesized products structure and their corrosion inhibition activities towards carbon steel in 1% NaCl solution utilizing Tafel plot method. Therefore, in this paper we report some results of the microwave-assisted organic synthesis of some imidazole derivative compounds and the analysis of their corrosion inhibitor corrosion activities toward carbon steel in 1% NaCl solution.

## Experimental

### A. General Procedure

All of reagents used in this research are GR grade. All of solvents were distilled before used. The microwave assisted organic synthesis (MAOS) method was utilizing a GE domestic microwave oven type JEL642WC. The characterization of synthesized products was analyzed using BUCK-IR® at Department of Chemistry ITB for the determination of infrared spectrums, and the determination of the melting point of products utilizing the *Fisher-Johns®* Melt-Temp Apparatus. The structure elucidation was determined utilizing JEOL DELTA NMR 400 MHz (<sup>1</sup>H-NMR) and 100 MHz (<sup>13</sup>C-NMR), using CDCl<sub>3</sub> and acetone-*d*<sub>6</sub> as solvents, at Universiti Kebangsaan Malaysia. The determination of inhibition activity of synthesized products employing *VoltaLab®* apparatus at Department of Chemistry ITB, with carbon steel electrode as working electrode, SCE (Saturated Calomel Electrode) as reference electrode, and platinum electrode as auxiliary electrode.

### B. The Synthesis of Imidazole Derivative Compounds

All of the synthesis procedures of imidazole derivative compounds were summarized as follows (also presented in Scheme 1). This method was the modification of the method used previously by Wolkenberg, et.al. [11] and Usyatinsky, et.al. [12]: 2 mmols of diketones (glyoxal for the synthesis of unsubstituted imidazole; benzil for the synthesis of **compound 1**, **compound 2** and **compound 3**), 2 mmols of aldehydes, 20 mmols of ammonium acetate and 10 mL acetic acid glacial were placed in 100 mL PYREX® Erlenmeyer flask. The

reaction mixtures were stirred well, and then the flask was placed in the GE domestic microwave oven type JEI642WC. The reaction mixtures were irradiated at 700 W of power for several times until the reaction temperature was no longer raised or achieving the stable temperature. The flask was taken out from the oven and was cooled to 40°C. Then the flask was placed in the ice bath. Into the reaction mixtures were added drop wise of saturated ammonium hydroxide solution until the cloudiness appeared. The crude products were filtered out and the residue was washed with aquadest. The crude products were purified utilizing preparative TLC with n-hexane/ethyl acetate = 7:3 (V/V) as eluents. The product were recrystallized in n-hexane/ethyl acetate = 1:1 (V/V).

**Compound 1** (2,4,5-triphenyl-1*H*-imidazole): <sup>1</sup>H-NMR 400 MHz JEOL DELTA (CDCl<sub>3</sub>): δ (ppm) 11.05 (s, broad, 1H); 7.95 – 7.92 (dd *J* = 8 and 2 Hz, 2H); 7.58 – 7.56 (m, 4 H); 7.47 – 7.45 (t, 2H); 7.34 – 7.26 (m, 7H). <sup>13</sup>C-NMR 100 MHz JEOL DELTA (CDCl<sub>3</sub>): δ 145.9, 131.5, 129.6 128.9, 128.6, 127.8, 127.4, 125.3. m/e: **296**; 218; 193; 190; 165; 163; 148; 104; 89; 77; 63; 39.

**Compound 2** (2-hexyl-4,5-diphenyl-1*H*-imidazole): <sup>1</sup>H-NMR 400 MHz JEOL DELTA (Acetone-*d*<sub>6</sub>): δ (ppm) 11.3 (s, broad, 1H); 7.53 – 7.45 (m, 4H); 7.35 – 7.28 (m, 4H); 7.25 – 7.20 (m, 2H); 2.70 (t, 2H); 1.75 (q, 2H); 1.40 – 1.29 (m, 6H); 0.88 (s, 3H). <sup>13</sup>C-NMR 100 MHz JEOL DELTA (Acetone-*d*<sub>6</sub>): δ (ppm) 149.4; 130.5; 129.3; 129.1; 128.9; 128.7; 32.3; 30.4; 30.2; 29.0; 23.2; 14.3. m/e: **304**; 289; 261; 247; 234; 219; 190; 165; 152; 116; 103; 89; 77; 55; 41.

**Compound 3** ((*E*)-2-ethyl-4,5-diphenyl-1-(prop-1-enyl)-1*H*-imidazole): <sup>1</sup>H-NMR 400 MHz JEOL DELTA (CDCl<sub>3</sub>): δ (ppm) 7.93 – 7.91 (dd, 2H); 7.49 – 7.44 (m, 4H); 7.36 – 7.24 (m, 4H); 6.21 (t, 1H); 2.80 – 2.71 (q, 2H); 2.27 – 2.23 (q, 1H); 1.35 – 1.31 (t, 3H); 1.08 – 1.04 (t, 3H). <sup>13</sup>C-NMR 100 MHz JEOL DELTA (CDCl<sub>3</sub>): δ (ppm) 149.4; 148.5; 131.8; 130.2; 129.6; 129.4; 129.3; 129.0; 128.5; 128.2; 125.8; 125.5; 21.9; 21.6; 13.8; 12.7. m/e: **288**; 273; 248; 234; 218; 193; 165; 152; 139; 115; 104; 89; 77; 63; 39.

**Imidazole**: <sup>13</sup>C-NMR 100 MHz JEOL DELTA (CDCl<sub>3</sub>): δ (ppm) 135.12 (1C); 121.85 (2C). m/e: **68**; 41; 28; 14.

### C. The Determination of Corrosion Inhibition Activity

An amount of 2 mgs of products was dissolved in 250 mL 1% (w/v) NaCl solution, to give 8 ppm concentration of sample solutions. The 1% (w/v) NaCl solution was also used as blank solution in each measurement. Into the 400 mL beaker glass equip with magnetic stirrer was introduced 250 mL of blank solution. The working electrode (carbon steel), the reference electrode (SCE), and auxiliary electrode (platinum electrode) were immersed into the electrolyte solution. Carbon dioxide gas was introduced into the electrolyte solution until saturation reached. The measurements utilizing *VoltaLab*<sup>®</sup> and Tafel Method software program until the curve of potential measurement towards time was completely formed well. The measurements of each sample solution must be initiated by the measurement of blank solution. The inhibition activity can be calculated using following equation [6][7][8]:

$$\% \text{EI} = \frac{\text{Blank Corrothion Rate (mm/Y)} - \text{Sample Corrosion Rate (mm/Y)}}{\text{Blank Corrosion Rate (mm/Y)}} \times 100\% \quad (1)$$

Or

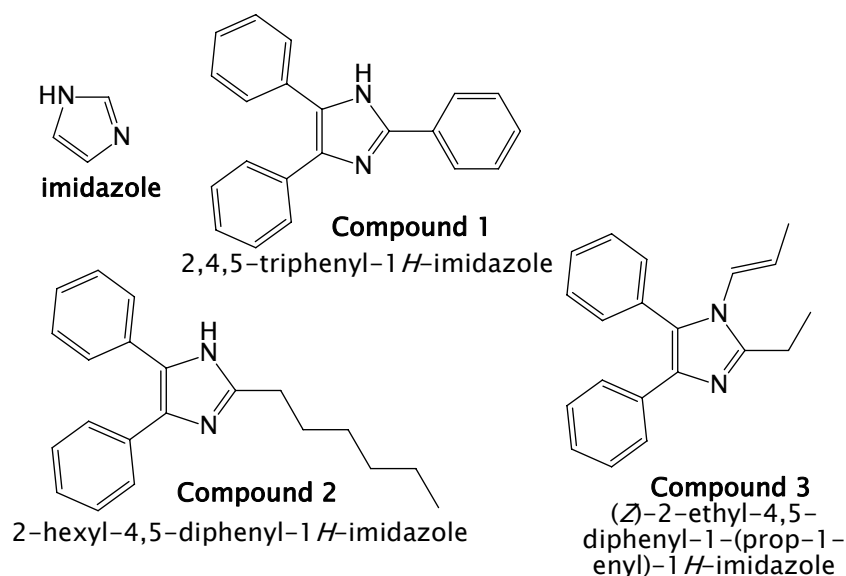
$$\% \text{EI} = \frac{I_{\text{Blank}} \text{ (mA/cm}^2\text{)} - I_{\text{Sample}} \text{ (mA/cm}^2\text{)}}{I_{\text{Blank}} \text{ (mA/cm}^2\text{)}} \times 100\% \quad (2)$$

## Results and Discussion

The synthesis of imidazole derivative compounds was carried out utilizing the MAOS (*Microwave Assisted Organic Synthesis*) method, which was the modification of the methods depicted by Wolkenberg, et.al. [11] and Usyatinsky, et.al. [12]. The results of the synthesis of imidazole derivative compounds were summarized in Table 1. The structures of each product were presented on Figure 1.

**Table 1.** The synthesis of imidazole derivative compounds utilizing the MAOS method at 700 W irradiation powers.

$  \begin{array}{c}  \text{R}^1 \\    \\  \text{O}=\text{C}-\text{C}=\text{O} \\    \quad   \\  \text{R}^1 \quad \text{R}^1 \\  \text{diketone, 2mmol}  \end{array}  +  \begin{array}{c}  \text{O} \\     \\  \text{R}^2 \\  \text{Aldehyde, 2 mmol}  \end{array}  \xrightarrow[\text{microwave irradiation at 700 W}]{+ 20 \text{ mmol NH}_4\text{OAc in 10 mL HOAc}}  \text{Product}  $							
Entry	-R <sup>1</sup>	-R <sup>2</sup>	Reaction Times (seconds)	Temperature (°C)	Melting Points (°C)	Yields (%)	Product
1	-H	-H	65	117	75-77	91.32	imidazole
2	-C <sub>6</sub> H <sub>5</sub>	-C <sub>6</sub> H <sub>5</sub>	40	110	271-272	88.24	2,4,5-triphenyl-1 <i>H</i> -imidazole
3	-C <sub>6</sub> H <sub>5</sub>	-C <sub>6</sub> H <sub>13</sub>	50	112	253-255	91.21	2-hexyl-4,5-diphenyl-1 <i>H</i> -imidazole
4	-C <sub>6</sub> H <sub>5</sub>	-C <sub>2</sub> H <sub>5</sub>	150	132	216-218	87.25	( <i>E</i> )-2-ethyl-4,5-diphenyl-1-(prop-1-enyl)-1 <i>H</i> -imidazole



**Figure 1.** The structures of imidazole derivative compounds based on spectroscopy data analysis

The investigation of the corrosion inhibition activities of imidazole, compound 1 (2,4,5-triphenyl-1*H*-imidazole), 2 (2-hexyl-4,5-diphenyl-1*H*-imidazole) and 3 ((*E*)-2-ethyl-4,5-diphenyl-1-(prop-1-enyl)-1*H*-imidazole) toward carbon steel in 1% NaCl solution were determined utilizing the Tafel plot method. The results were

summarized in Table 2. The efficiency inhibition activities of each compound were calculated based on equation (1) and (2).

**Table 2.** The efficiency inhibition activities (%EI) of synthesized imidazole derivative compounds towards carbon steel in 1% NaCl solution utilizing Tafel method

Sample	$I_{corr}$ of 1% NaCl solution (mA/cm <sup>2</sup> )	$I_{corr}$ of sample in 1% NaCl solution (mA/cm <sup>2</sup> )	% EI (Efficiency Inhibition)
Imidazole	0.1326	0.0984	27.65
Compound 1	0.1235	0.0591	52.15
Compound 2	0.1309	0.1061	18.95
Compound 3	0.1636	0.1044	36.19

From Table 2 it was observed that the corrosion inhibition activities of imidazole derivative compounds depend on the structure and substituents attached to the imidazole ring framework. Compound 1 (2,4,5-triphenyl-1*H*-imidazole) gave the highest corrosion inhibition activities than unsubstituted imidazole, compound 2 and 3. This corrosion inhibition activity should be related to the presence of three aromatic benzene rings attached to imidazole ring framework, which have the potent to interact with iron of carbon steel through their phi's ( $\pi$ ) electrons as Lewis base. The planarity of compound 1's structure should be an additional account to the readily close packed in the interaction process with carbon steel surface. The additional phi's ( $\pi$ ) electron at the N(1) position and the ethyl substituent at C(2) position of imidazole ring of compound 3 ((*E*)-2-ethyl-4,5-diphenyl-1-(prop-1-enyl)-1*H*-imidazole) contributed to the better corrosion inhibition activity of compound 3 compare to unsubstituted imidazole. The electron-donating group of ethyl substituent on compound 3 increased the basicity of imidazole ring framework; therefore it would be a better Lewis base to interact with the acid Lewis carbon steel. On the other hand, the similar electron donating property of hexyl substituent on compound 2 (2-hexyl-4,5-diphenyl-1*H*-imidazole) did

not increase its corrosion inhibition activity compare to unsubstituted imidazole. The basicity of compound **2** should be higher than unsubstituted imidazole; however the steric and bulky structure caused by the presence of long alkyl group (hexyl group) would prevent the close packed interaction between compound **2** and carbon steel surface. Therefore the preventive layer made by compound **2** did not achieve the better protection on carbon steel surface.

In order to investigate the corrosion inhibition mechanism we analyzed the relationship between the various concentrations of corrosion inhibitors and their corrosion efficiency inhibition percentages. The analysis of the adsorption mechanism of imidazole derivative compounds towards carbon steel can be derived by the determination of the degree of surface coverage ( $\theta$ ) according to the following equation [6][7],

$$\theta = 1 - \left( \frac{I_{inh}}{I_{uninh}} \right) \quad (3)$$

with  $I_{inh}$  and  $I_{uninh}$  is the corrosion current density (in mA/cm<sup>2</sup>) of sample solution with and without inhibitor, respectively. The data of the degree of surface coverage,  $\theta$ , at various concentrations of compound **1b**, **2b** and **3b** at 27 °C was also presented on Table 3.

Based on Table 3 below, the most suitable relationship between concentration of corrosion inhibitor compounds,  $C_{inh}$ , and the degree of surface coverage,  $\theta$ , is the Langmuir isotherm adsorption in the simplest form, which is the linear relationship between  $C_{inh}$  and  $C_{inh}/\theta$ , according to the following equation [6][7],

$$\theta = \frac{bC_{inh}}{1 + bC_{inh}} \quad (4)$$

with  $b$  is the adsorption coefficient;  $C_{inh}$  is the concentration of corrosion inhibitor compounds and  $\theta$  is the degree of surface coverage. Table 3 represents the data of each compound at concentration of 8, 16 and 32 ppms of sample in 1% NaCl solution.



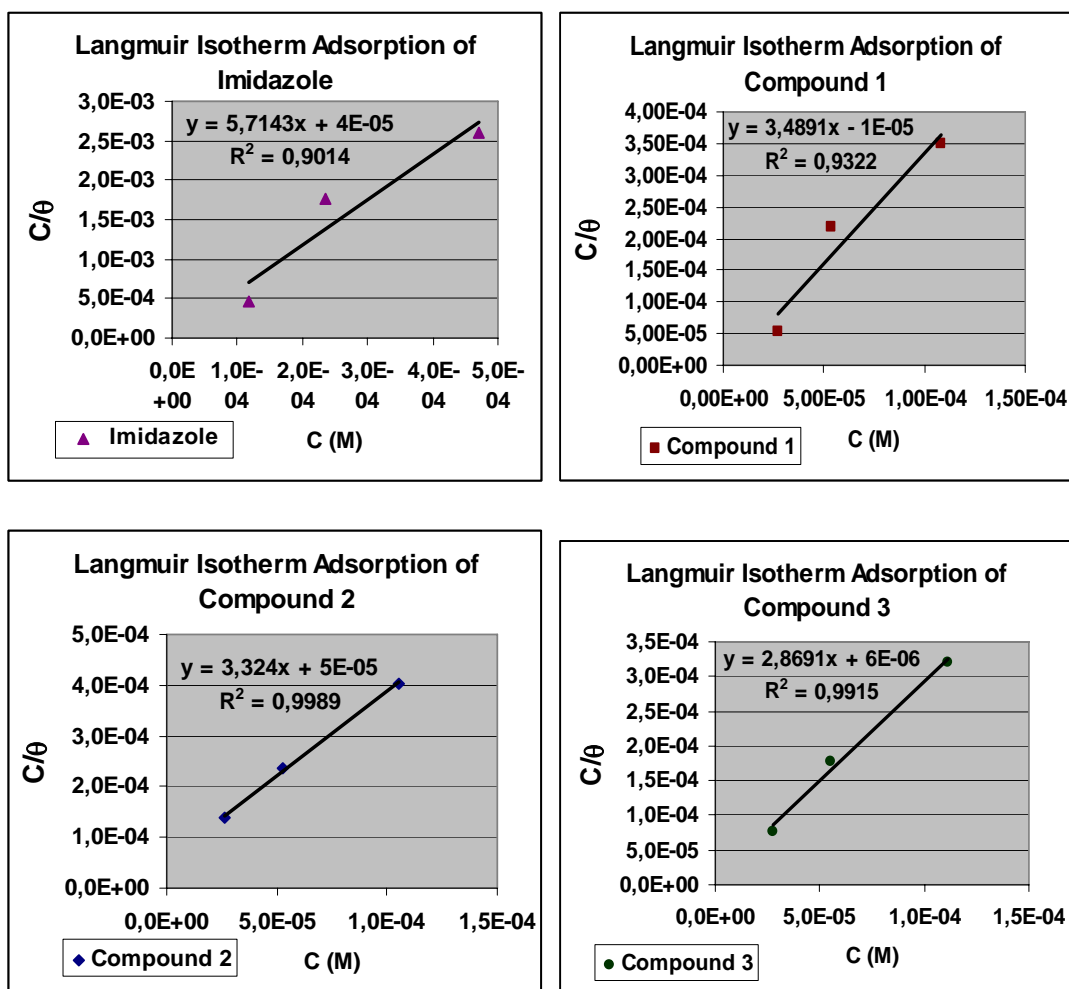
**Table 3.** The correlation between %EI of imidazole derivative compounds and the degree of surface coverage,  $\theta$ , on carbon steel

Sample	C (ppm)	%EI	$\theta$	C/ $\theta$	Log C	Log( $\theta/1-\theta$ )
Imidazole	1.2E-04	25.79	0.26	4.6E-04	-3.93	-0.46
	2.4E-04	13.29	0.13	1.8E-03	-3.63	-0.81
	4.7E-04	18.09	0.18	2.6E-03	-3.33	-0.66
Compound 1	2.7E-05	52.15	0.52	5.2E-05	-4.57	0.04
	5.4E-05	24.62	0.25	2.2E-04	-4.27	-0.49
	1.1E-04	30.94	0.31	3.5E-04	-3.97	-0.35
Compound 2	2.6E-05	18.95	0.19	1.4E-04	-4.58	-0.63
	5.3E-05	22.42	0.22	2.4E-04	-4.28	-0.54
	1.1E-04	26.12	0.26	4.0E-04	-3.98	-0.45
Compound 3	2.8E-05	36.19	0.36	7.7E-05	-4.56	-0.25
	5.6E-05	31.25	0.31	1.8E-04	-4.26	-0.34
	1.1E-04	34.71	0.35	3.2E-04	-3.95	-0.27

Figure 2 represents the linear relationship according to Langmuir adsorption isotherm of the synthesized products. The value of  $b$ , the adsorption coefficient, which is equal to the adsorption equilibrium constant ( $K_{ads}$ ), of each compound was presented on Table 4. The value of free Gibbs adsorption energy ( $\Delta G^0_{ads}$ , in kJ/mol) of each compound, which is also presented on Table 4, can be determined using the following equation [6][7],

$$K_{ads} = \frac{1}{55} \exp\left(-\frac{\Delta G^0_{ads}}{RT}\right) \quad (5)$$

with  $R$  = ideal gas constant = 8.314 J/mol.K and  $T$  is temperature in K.



**Figure 2.** The linear relationship between concentration of corrosion inhibitor compounds,  $C_{inh}$ , and  $C_{inh}/\theta$ , according to Langmuir adsorption isotherm of imidazole, compound 1, 2 and 3.

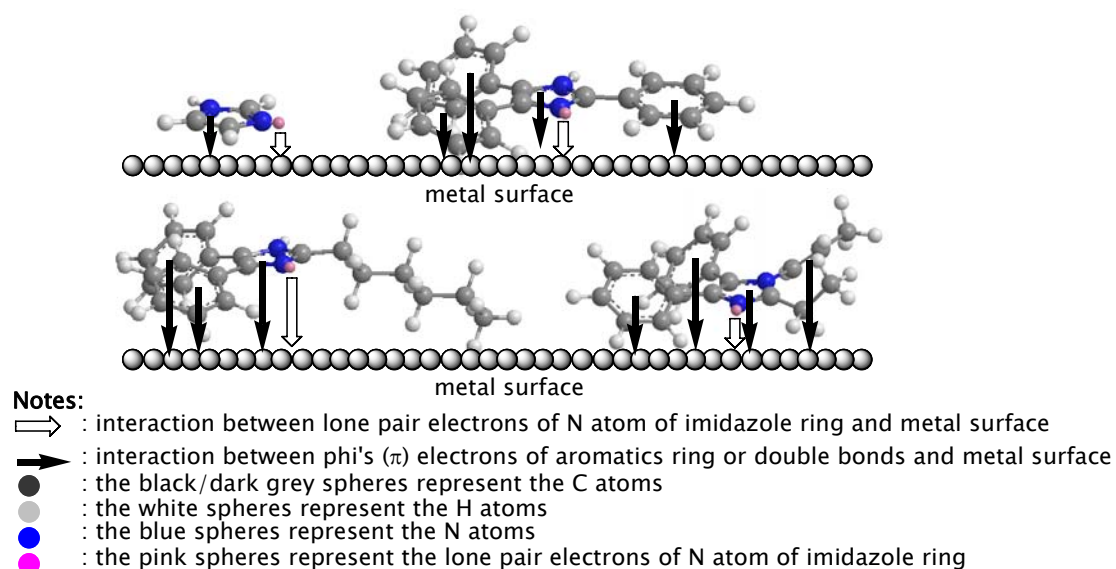
The linearity of each Langmuir isotherm adsorptions of each compounds, as shown on Figure 2, represent the monolayer formation of each compound on the carbon steel surface. Based on Table 4 we can see that all of free Gibbs adsorption energy values,  $\Delta G^0_{ads}$ , for each compound are negatives, which indicated the spontaneity of adsorption process of each compound on carbon steel surface. The more negative of  $\Delta G^0_{ads}$  value the more spontaneous its adsorption process on metal surface, therefore the corrosion inhibition activity would increase. The value of  $\Delta G^0_{ads}$  up to  $-20$  kJ/mol is consistent with the physical adsorption (physisorption), whether the value of  $\Delta G^0_{ads}$ , which is above  $-40$  kJ/mol, is consistent with the chemical adsorption (chemisorption) [6][7]. Therefore, imidazole, compound 1, 2 and 3 have the potent to interact with carbon steel through semi-

physiosorption or semi-chemisorption because of their  $\Delta G^0_{ads}$  values are in between - 20 kJ/mol and - 40 kJ/mol.

**Table 4.** The coefficient adsorption values ( $b$ ) and free Gibbs adsorption energy ( $\Delta G^0_{ads}$ , in kJ/mol) of the synthesized imidazole derivative compounds at 27°C (300K)

Sample	$b$ (M <sup>-1</sup> )	$\Delta G^0_{ads}$ , (kJ/mol)
Imidazole	1358.23	-28.01
Compound 1	16834.86	-34.29
Compound 2	5910.58	-31.68
Compound 3	11127.99	-33.26

Figure 3 represents the simulation of the arrangements of imidazole, compound 1, 2 and 3 on metal surface. Each structure has already been in its minimized energy utilizing the MM2 program in Chem3D Ultra 8.0 of ChemOffice Cambridgesoft® software.



**Figure 3.** The description of three-dimensional simulation, which illustrates the interactions of imidazole derivative compounds on the

metal surface, utilized the Chem3D Ultra 8.0® software. Metal surface is indicated by the arrangements of round balls under each molecule.

As we can see from Figure 3 that the adsorption sites of compound 1 are at lone pair electrons of imidazole ring and phi's ( $\pi$ ) electrons of three aromatic benzene rings and imidazole ring itself. The adsorption sites of compound 1 are more various in amount and quality than imidazole, compound 2 and 3 resulting the highest in its corrosion inhibition activity and the more negative of its free Gibbs adsorption energy. Thus compound 1 has the most spontaneous adsorption process on metal surface than others. On the other hand, unsubstituted imidazole has the least interaction than the others that caused the smallest efficiency inhibition and the least adsorption spontaneity on metal surface. The steric hindrance of hexyl group of compound 2 has made the interaction between its Lewis bases sites and the Lewis acid metal surface became weaker than compound 1 and 3 that caused the low efficiency corrosion inhibition.

## Conclusions

The synthesis of imidazole derivative compounds utilizing the MAOS method produced four types of compounds: imidazole, compound 1 (2,4,5-triphenyl-1*H*-imidazole), 2 (2-hexyl-4,5-diphenyl-1*H*-imidazole) and 3 ((*E*)-2-ethyl-4,5-diphenyl-1-(prop-1-enyl)-1*H*-imidazole). Compound 1 has the highest corrosion inhibition activity towards carbon steel (52.12%) at 8 ppm concentrations in 1% NaCl solution. Based on the analysis of Tafel plot data, the most suitable relationship between concentration of corrosion inhibitor compounds,  $C_{inh}$ , and the degree of surface coverage,  $\theta$ , is the Langmuir isotherm adsorption in the simplest form. The linearity of each Langmuir isotherm adsorptions of each compounds represent the monolayer formation of each compound on the carbon steel surface. All of free Gibbs adsorption energy values,  $\Delta G^0_{ads}$ , for each compound is a negative, which indicated the spontaneity of adsorption process of each compound on carbon steel surface. Imidazole, compound 1, 2 and 3 have the potent to interact with carbon steel through semi-physiosorption or semi-chemisorption because of their  $\Delta G^0_{ads}$  values are in between - 20 kJ/mol and - 40 kJ/mol.

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