Journal of Alloys and Compounds 712 (2017) 121–133

Contents lists available at ScienceDirect

Journal of Alloys and Compounds

journal homepage: http://www.elsevier.com/locate/jalcom

Electrochemical, surface and quantum chemical studies of novel imidazole derivatives as corrosion inhibitors for J55 steel in sweet corrosive environment

Ambrish Singh [a, b, *], K.R. Ansari [c], Ashok Kumar [d], Wanying Liu [a], Chen Songsong [a], Yuanhua Lin [a, b, **]

[a] School of Materials Science and Engineering, Southwest Petroleum University, Chengdu, 610500, Sichuan, China
[b] State Key Laboratory of Oil and Gas Reservoir Geology and Exploitation, Southwest Petroleum University, Chengdu, Sichuan, 610500, China
[c] Department of Chemistry, Indian Institute of Technology, Banaras Hindu University, Varanasi, 221005, U.P., India
[d] Department of Chemistry and Biochemistry, Arizona State University, Tempe, AZ, 85287-1604, USA

ARTICLE INFO

Article history:
Received 28 January 2017
Received in revised form
7 March 2017
Accepted 7 April 2017
Available online 10 April 2017

Keywords:
Imidazole
J55 steel
XPS
EIS
SECM
AFM

ABSTRACT

The corrosion inhibition performance of three novel imidazole derivatives namely 2-(4-methoxyphenyl)-4,5-diphenyl-imidazole (M-1), 4,5-diphenyl-2-(*p*-tolyl)-imidazole (M-2) and 2-(4-nitrophenyl)-4,5-diphenyl-imidazole (M-3) for J55 steel in CO_2 saturated brine solution was studied by weight loss method, electrochemical impedance spectroscopy (EIS), potentiodynamic polarization, scanning electrochemical microscopy (SECM), contact angle, scanning electron microscope (SEM), atomic force microscopy (AFM), X-ray photoelectron spectroscopy (XPS) and quantum chemical calculation. M-1 exhibited the best inhibition efficiency of 93% at 400 mg/L concentration. The adsorption of the imidazole derivatives obeyed the Langmuir adsorption isotherm. Contact angle measurement reveals the hydrophobic nature of J55 steel in presence of inhibitors. Quantum chemical calculation well supports the experimental results.

1. Introduction

Corrosion by carbon dioxide in oil and gas industry is the major factor for the failure of pipelines in deep and ultra-deep wells and thus it has considered as the hot spot research area for decades. At present, many pipelines used in petroleum industry are made of low alloy steels due to economic reasons [1]. However, they are very prone to undergo corrosion due to the formation of weak carbonic acid by the reaction of the carbon dioxide with dissolves saltwater [2]. There are many methods to minimize the corrosion by carbon dioxide, but out of these use of corrosion inhibitor is considered as the most cost effective [3,4].

Imidazoline/imidazole and their derivatives are considered to be the most effective organic inhibitors [5,6], and have been widely used against CO2 corrosion in pipelines [7–13]. Because of their many biological, chemical and pharmaceutical properties imidazole derivatives are considered as"green" corrosion inhibitors [14]. In past years imidazole derivatives have been used as corrosion inhibitor in hydrochloric acid [15–22]. The inhibition effectiveness of imidazole derivatives is mainly attributed to the adsorption of the inhibitor on metallic surfaces. The inhibitor adsorbs on the metal surface and suppresses the electrochemical reactions of corrosion processes [23].

The present work describes the synthesis and characterization of three novel imidazole derivatives namely 2-(4-methoxyphenyl)-4,5-diphenyl-imidazole (M-1), 4,5-diphenyl-2-(*p*-tolyl)-imidazole (M-2) and 2-(4-nitrophenyl)-4,5-diphenyl-imidazole (M-3) for J55 steel in CO_2 saturated brine solution using weight loss method, electrochemical impedance spectroscopy (EIS), potentiodynamic polarization, scanning electrochemical microscopy (SECM), contact angle, scanning electron microscope (SEM), atomic force microscopy (AFM), X-ray photoelectron spectroscopy (XPS) and quantum chemical calculation.

* Corresponding author. School of Materials Science and Engineering, Southwest Petroleum University, Chengdu, 610500, Sichuan, China.
** Corresponding author. School of Materials Science and Engineering, Southwest Petroleum University, Chengdu, 610500, Sichuan, China.
E-mail addresses: vishisingh4uall@gmail.com (A. Singh), yhlin28@163.com (Y. Lin).

http://dx.doi.org/10.1016/j.jallcom.2017.04.072
0925-8388/

Our motivation to study imidazole derivative as corrosion inhibitor for J55 steel was due their environmentally benign nature.

2. Experimental

2.1. Synthesis of inhibitors

Imidazole derivatives were synthesized in the laboratory by using the known procedure [14]. A mixture of benzil (1 mmol), aromatic aldehyde (1.1 mmol) and ammonium acetate (2 mmol) was poured into 6 ml of glycerol, and stirred at 90° C for 40–90 min under ambient conditions. After completion of the reaction (monitored by TLC) the reaction mixture was poured into ice water. The separated solid was filtered and the product was crystallized using ethanol. Table 1 represents the molecular structure, abbreviations and analytical properties of all imidazole derivatives. The scheme of synthesis is shown in Fig. 1. The detail characterization is as follows:

(a) **2-(4-methoxyphenyl)-4,5-diphenyl-imidazole (M-1)**
^{1}HNMR (300 MHz, DMSO-d_6): 12.50 (s, 1H), 6.53–7.57 (m, 14H), 3.82 (s, 3H)

(b) **4,5-diphenyl-2-(*p*-tolyl)-imidazole (M-2)**
^{1}H NMR (300 MHz, DMSO-d_6): 12.59 (s, 1H), 7.12–7.48 (m,14H), 2.35 (s, 3H)

(c) **2-(4-nitrophenyl)-4,5-diphenyl-imidazole (M-3)**
^{1}HNMR (300 MHz, DMSO-d_6): d 12.81 (s, 1H), 6.15–8.34 (m, 14H)

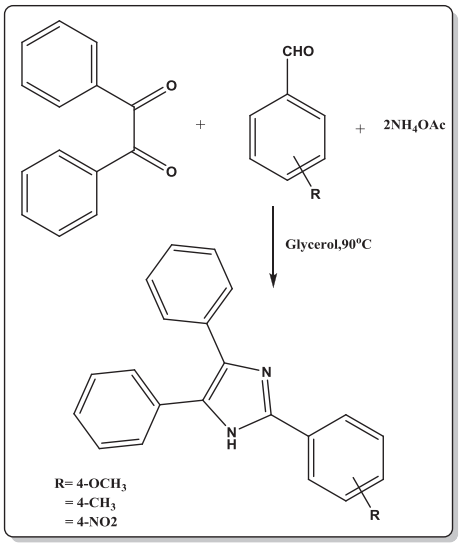

Fig. 1. Synthetic scheme of inhibitors.

2.2. Materials and solutions

J55 steel composition are as follows (wt.%): C 0.24; Si 0.22; Mn 1.1; P 0.103; S 0.004; Cr 0.5; Ni 0.28; Mo 0.021; Cu 0.019; Fe remainder. The steel coupons are flat with a dimension of 5.0 cm × 2.5 cm × 0.2 cm and 2.0 cm × 1.0 cm × 0.025 cm used for gravimetric and electrochemical experiments respectively. Only one end face (1.0 cm^2) was exposed and the rest was sealed by epoxy resin. All the steel coupons were abraded through 600, 800 and 1200 grit silicon carbide metallurgical papers, degreased in acetone, washed with anhydrous ethanol, and then dried at room temperature and finally kept in the desiccators. In the prepared 3.5% NaCl solution CO_2 gas was passed for 120 min at pressure of 6 MPa till the pH of the solution becomes 4 ± 0.05 and when necessary the pH was adjusted with small amounts of $NaHCO_3$ or HCl. The 3.5% NaCl solution was continuously saturated with CO_2 throughout the experiment and also nitrogen gas is passed for minimizing the oxygen concentration. All the experiments were performed in static, unstirred solutions.

2.3. Methods

3.5% NaCl solutions were prepared with double distilled water. Different concentration (25, 50, 100, 200, 300, 400 mg/L) of imidazole derivatives were dissolved in the corrosive media i.e. 3.5% NaCl saturated with CO_2. The corrosion behaviors of J55 steel in 3.5% NaCl saturated with CO_2 with and without inhibitors were

Table 1
Molecular structure and analytical data of inhibitors.

Inhibitor	Structure		Analytical data
2-(4-methoxyphenyl)-4,5-diphenyl-imidazole (M-1)	**2-(4-methoxyphenyl)-4,5-diphenyl-imidazole (M-1)**	Mp. 230-232°C. **^{1}HNMR** (300 MHz, DMSO-d_6): 12.50 (s, 1H), 6.53-7.57 (m, 14H), 3.82 (s, 3H)	Mp. 230–232 °C. **^{1}HNMR** (300 MHz, DMSO-d_6): 12.50 (s, 1H), 6.53–7.57 (m, 14H), 3.82 (s, 3H)
4,5-diphenyl-2-(*p*-tolyl)-imidazole (M-2)	**4,5-diphenyl-2-(p-tolyl)-imidazole (M-2)**	Mp. 236–238°C. **^{1}H NMR** (300 MHz, DMSO-d_6): 12.59 (s, 1H), 7.12-7.48 (m,14H), 2.35 (s, 3H)	Mp. 236–238 °C. **^{1}H NMR** (300 MHz, DMSO-d_6): 12.59 (s, 1H), 7.12–7.48 (m,14H), 2.35 (s, 3H)
2-(4-nitrophenyl)-4,5-diphenyl-imidazole (M-3)	**2-(4-nitrophenyl)-4,5-diphenyl-imidazole (M-3)**	Mp 241-242°C. **^{1}HNMR** (300 MHz, DMSO-d6): d 12.81 (s, 1H), 6.15-8.34 (m, 14H)	Mp 241–242 °C. **^{1}HNMR** (300 MHz, DMSO-d_6): d 12.81 (s, 1H), 6.15–8.34 (m, 14H)