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Corrosion Mitigation by Planar Benzimidazole Derivatives

*Ambrish Singh, Kashif R. Ansari, Dheeraj S. Chauhan,
Mumtaz A. Quraishi, Savas Kaya, Hua Yu
and Yuanhua Lin*

Abstract

The corrosion has a considerable amount of impact on the economics of every nation, and ultimately it affects the GDP. In the present era, the challenge given by corrosion can be easily mitigated using organic compounds as corrosion inhibitor in different corrosive media. The important property of an inhibitor is the presence of the metal interacting with heteroatoms and a planar structure. In this regard, benzimidazoles (BI) with a fused bicyclic ring consisting of benzene and imidazole moiety in their structural framework making them a potential candidate for anti-corrosion work. In addition to this, benzimidazole derivatives are classified as green inhibitor due to different kinds of biological activities. Their higher potency to mitigate corrosion is because of the planar molecular structure, nitrogen atom and sp^2 hybridized carbon, which provide them an ability to strongly interact with the metal. The focus of this book chapter is to investigate briefly the anti-corrosion ability of benzimidazole (BI) and their derivatives as a potential corrosion inhibitor for various industrially useful metals in different aggressive media.

Keywords: benzimidazole, corrosion, inhibitor

1. Introduction

The costs and the aftermath of corrosion are enormous. There are so many reports identified, resulting in the death and injury of living beings due to the corrosion failure. The easily quantifiable costs are those associated with the repair and replacement of equipment. It is very much difficult to predict the exact loss associated with the corrosion especially when corrosion causes the production unit in the petroleum industry to shut down. The potential method to reduce the corrosion is using the organic compounds in the aggressive media for protecting the metals against corrosion. Among the numerous inhibitor groups, benzimidazole derivatives have gained a more practical application due to their environmentally benign nature, potential ability to strongly interact with the metal surface through the nitrogen atoms and easy methods to modify their molecular structure as per the required need.

The benzo derivative of imidazole is referred to as benzimidazole. Although benzimidazole is the commonest name of the parent compound of the series, other names such as benzimidazole and 1,3-benzodiazole are often used. The main purpose of this book chapter is to introduce briefly the anti-corrosion ability of

benzimidazole and their derivatives as a potential corrosion inhibitor for the various industrially useful metals in different aggressive media.

1.1 A brief overview on biological activities

The molecular structure of benzimidazole arises by fusing the benzene and imidazole rings. It generates a class of bicyclic compounds which have a wide range of biological and pharmacological activities (**Figure 1**).

The molecular structures of some benzimidazole containing nucleus compounds, which are commonly used for various therapeutic applications, are presented in **Figure 2**: (1) benoxaprofen analog (anti-inflammatory), (2) pantoprazole (proton pump inhibitor), (3) candesartan (antihypertensive), (4) albendazole (anthelmintics/antimicrobial), (5) mebendazole (anthelmintics), (6) bilastine (antihistaminic), (7) carbendazim (antifungal), (8) bendamustine (antitumor) and (9) astemizole (antihistaminic) [1].

1.2 Molecular structure

The general molecular structure consists of a bicyclic heteroatomic structure with two nitrogen atoms (**Figure 3**).

The benzimidazole derivatives have different kinds of extensive coupling constant, which leads to generate coupling with strong force constants [2].

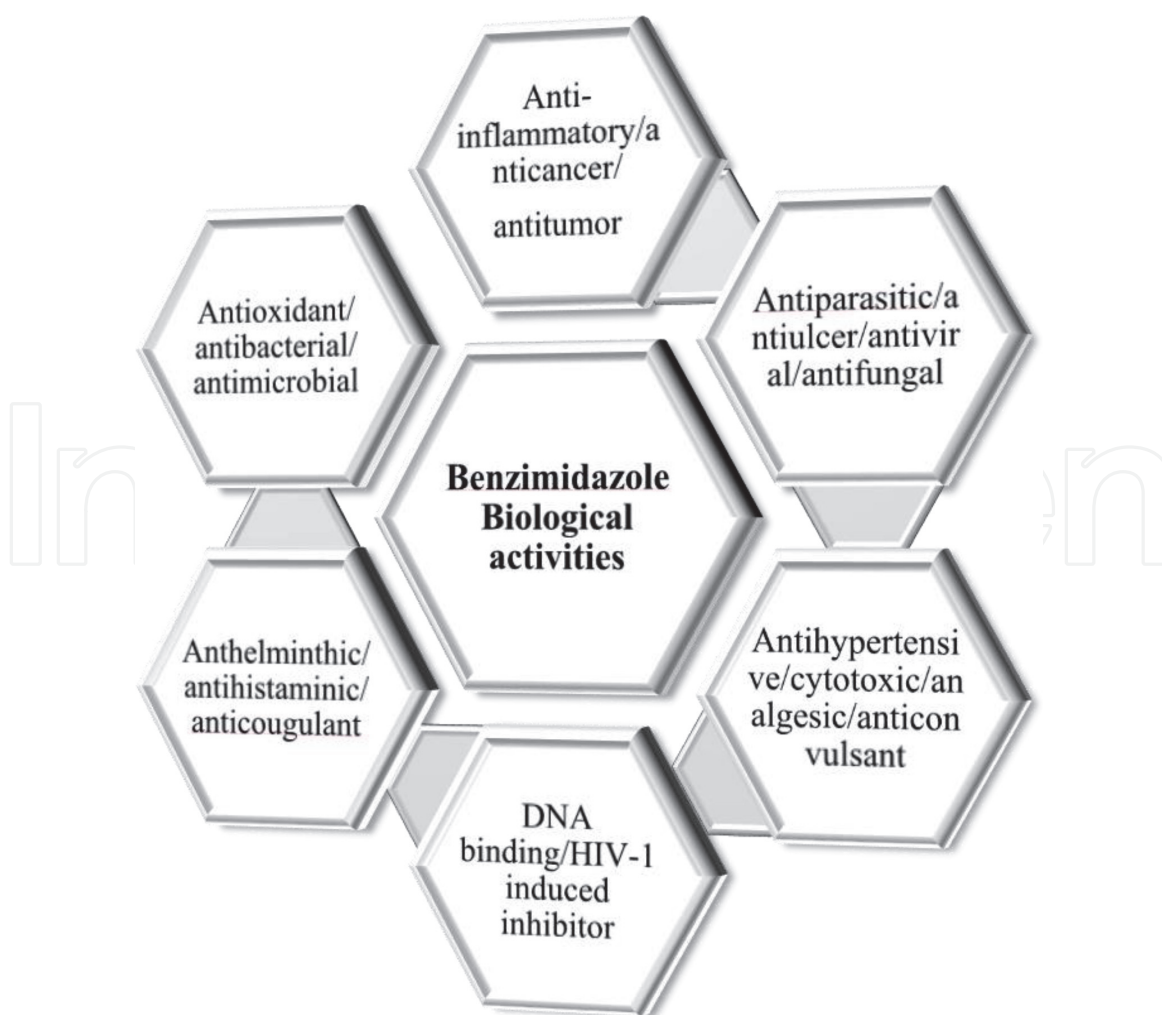


Figure 1.
Biological activities of benzimidazole.

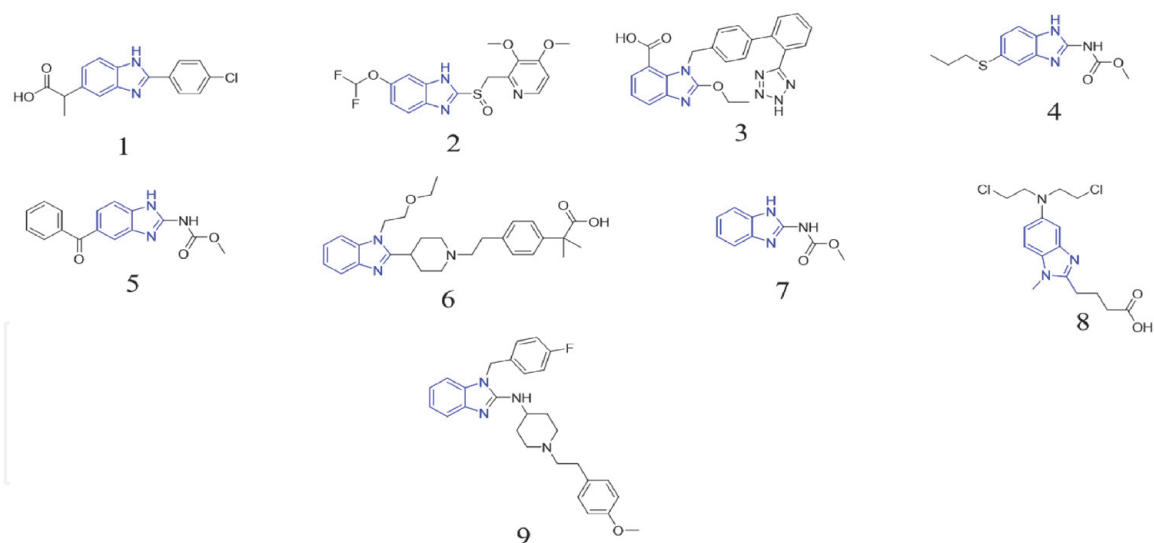


Figure 2.
Therapeutic applications of benzimidazole derivatives.

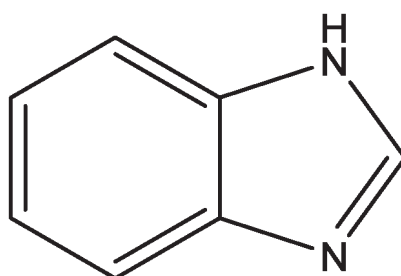


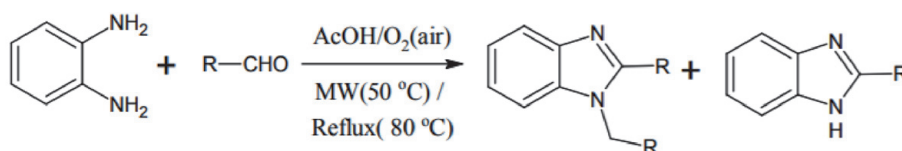
Figure 3.
Molecular structure of benzimidazole.

Benzimidazole derivatives have a greater affinity to produce a variation of chemical species because of the high force constant, and this is a remarkable characteristic of the benzimidazole. The analysis of vibrational property of benzimidazole derivatives is difficult due to the presence of extensive coupling in their molecular structure [3], although scientists were successful to analyse the vibrational properties of simple benzimidazole derivatives by applying density functional theory (DFT).

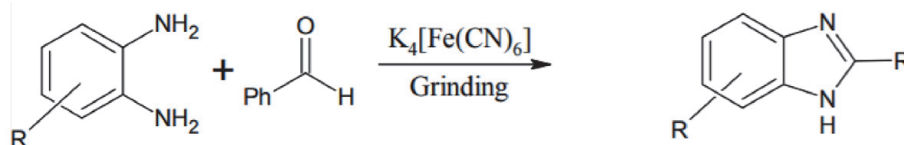
1.3 Synthesis

The synthesis of benzimidazole is very important with respect to their varying potential pharmacological activities. Thus, a unique attention is required to fulfil this process. In literature, various methods are given using different kinds of catalyst. Few of them are as follows:

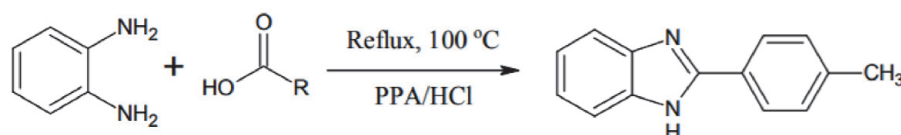
Azarifar et al. [4], synthesized benzimidazole derivatives by condensation of o-phenylenediamine promoted by acetic acid under microwave. They concluded that a mild, manipulatable procedure, eco-friendly and green aspects avoiding hazardous solvents, shorter reaction times and high yields of the products are the advantages of this method.



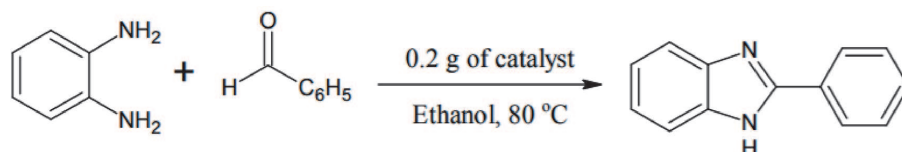
Shaikh et al. [5] have efficiently synthesized benzimidazoles in high yields by treatment of 1,2-diamine with aldehydes using the metal coordinate complex $K_4[Fe(CN)_6]$ as a catalysis.



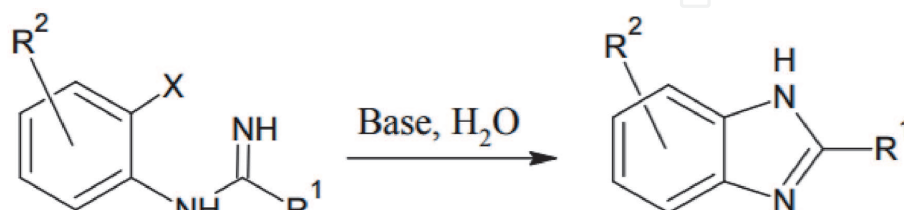
Vaidehi et al. [6] had synthesized a set of 2-substituted benzimidazoles successfully by condensation of o-phenylenediamine with substituted acids in the presence of ring closing agents like polyphosphoric acid/HCl. The present work has demonstrated the use of a simple cyclocondensation method and ring closing agents for synthesis of 2-substituted benzimidazoles.



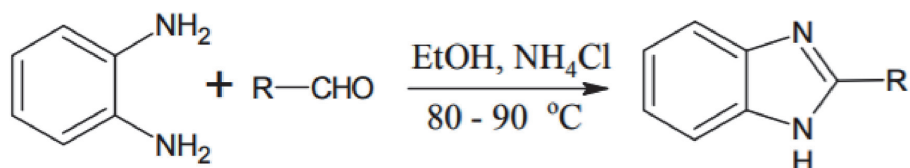
Rekha et al. [7] studied the catalytic activity of alumina, zirconia, manganese oxide/alumina and manganese oxide/zirconia in the condensation reaction between o-phenylenediamine and an aldehyde or a ketone to synthesize 2-substituted benzimidazoles and 1,5-disubstituted benzodiazepines, respectively.



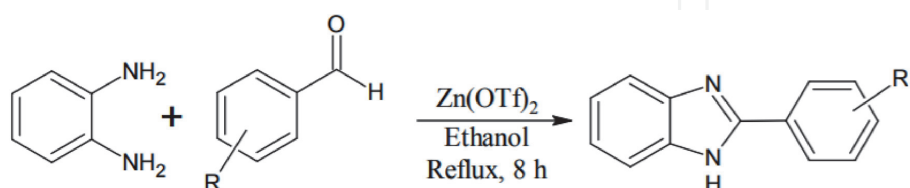
Chunxia et al. [8] have developed a straightforward method for the synthesis of the benzimidazole ring system through a carbon-nitrogen cross-coupling reaction in the presence of K_2CO_3 in water at 100 °C for 30 h; the intermolecular cyclization of N-(2-iodoaryl) benzamidine provides benzimidazole derivatives in moderate to high yields.



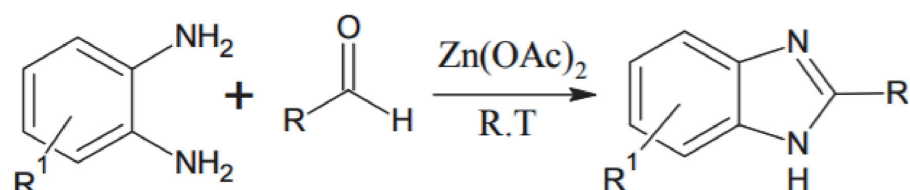
Kathirvelan et al. [9] synthesized various 2-substituted benzimidazoles in moderate to good yields in a one-pot reaction by condensation of o-phenylenediamine and an aldehyde in the presence of ammonium chloride as a catalyst at 80–90 °C.



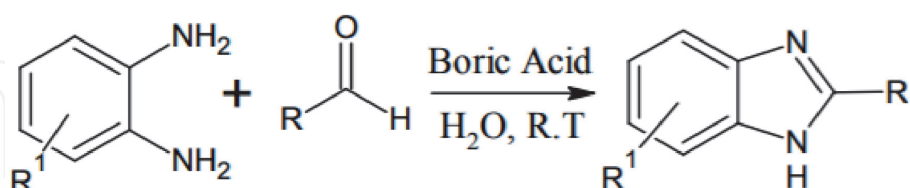
Srinivasulu et al. synthesized benzimidazole derivatives using zinc triflate as an efficient catalyst in a one-pot synthesis of 2-substituted benzimidazole derivatives from o-phenylenediamine and substituted aldehydes in ethanol solvent at reflux temperature. They concluded that zinc triflate was found to be an efficient catalyst for the formation of benzimidazole from aldehydes and o-phenylenediamine [10].



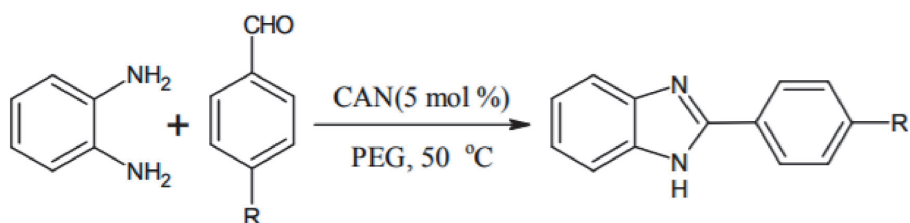
Patil et al. [11] have shown that benzimidazole derivatives have been synthesized by reacting substituted o-phenylenediamine with aldehyde derivatives using a catalytic amount of zinc acetate at room temperature with excellent yields.



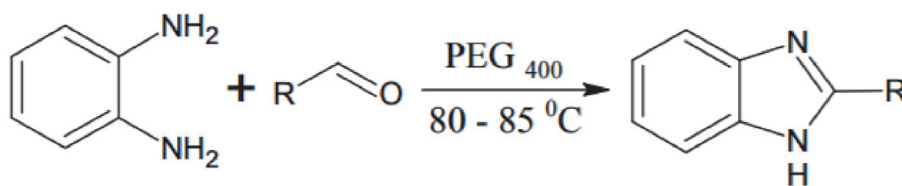
Karimi-Jaberi et al. [12] had synthesized 2-substituted benzimidazoles in a one-pot reaction from o-phenylenediamine and aldehydes in the presence of boric acid in water at room temperature.



Kidwai et al. [13] synthesized benzimidazole derivatives from o-phenylenediamine and aldehydes in PEG as a solvent with ceric ammonium nitrate (CAN) as a catalyst. This method provides a novel route for the synthesis of benzimidazoles in good yields with little catalyst loading.



Khunt et al. [14] have synthesized the benzimidazole by reacting o-phenylenediamine with several aldehydes using a green solvent PEG400 and got good yields.



2. Corrosion inhibition by benzimidazole derivatives

The inhibition ability of the organic compounds containing heteroatoms depends upon the rate of the molecular adsorption at the metal/solution interface which in turn depends upon the molecular structure/symmetry and charge density presenting on the inhibitor molecules [15]. Benzimidazoles are very good corrosion inhibitors because of their unique molecular structure and solubility in aqueous media. As we have seen in **Figure 3**, the molecular structure of benzimidazole has an aromatic property due to the presence of the fused six-membered benzene ring with the imidazole ring. The presence of planar benzene ring with sp² hybridized carbon atoms and lone pair of electrons presenting on the nitrogen atoms of the imidazole ring makes the whole molecular system as an anchoring site for molecular adsorption at the metal surface [16, 17].

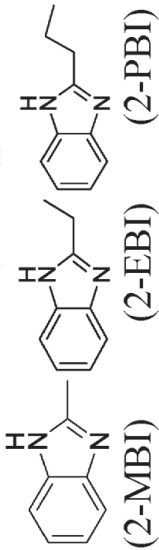
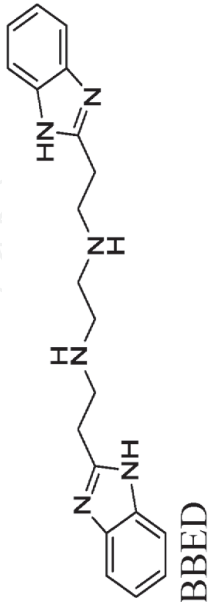
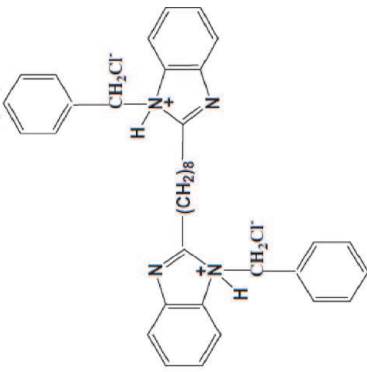
2.1 Metal protection by benzimidazole in aqueous media

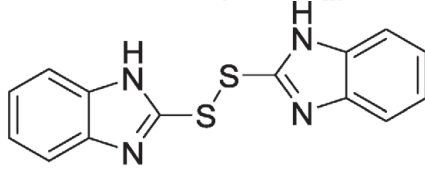
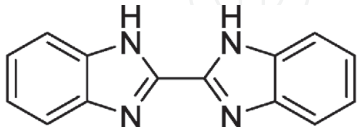
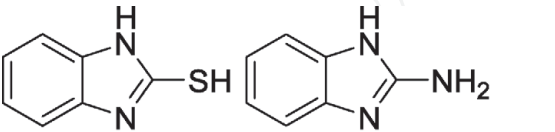
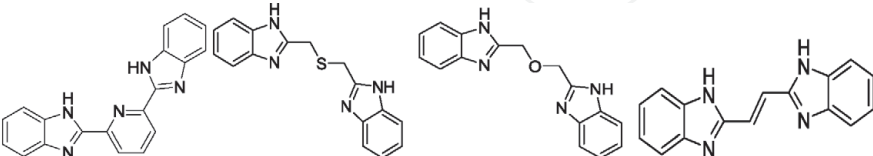
Benzimidazole and its derivatives prevail themselves as potential corrosion inhibitor candidates. They prove the ability to reduce the corrosion phenomenon in aqueous media for various metals like mild steel, copper, brass and zinc. In this chapter, I would like to emphasize only on these metals. The molecular structures of benzimidazole derivatives studied in this book chapter are tabulated in **Table 1**.

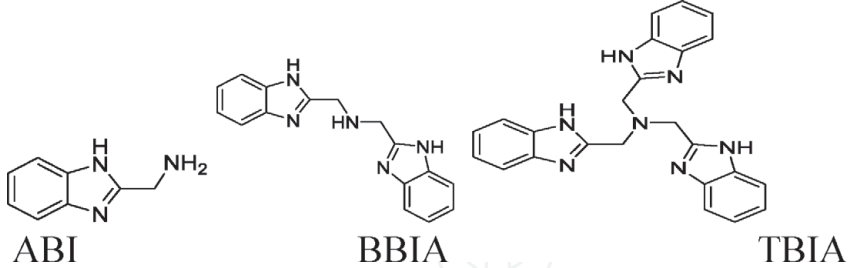
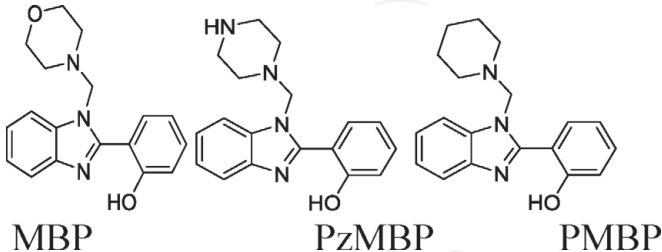
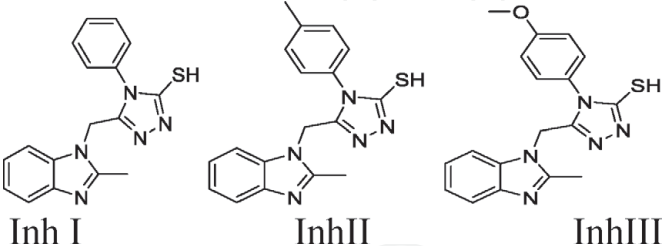
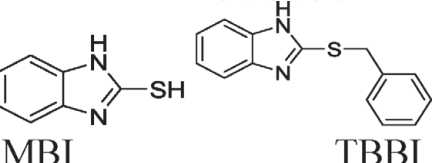
2.1.1 Experimental results

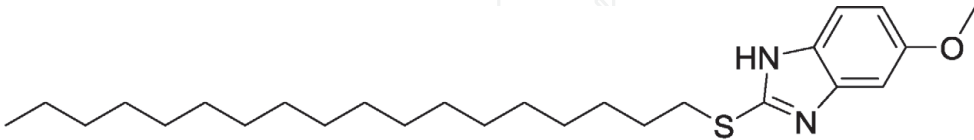
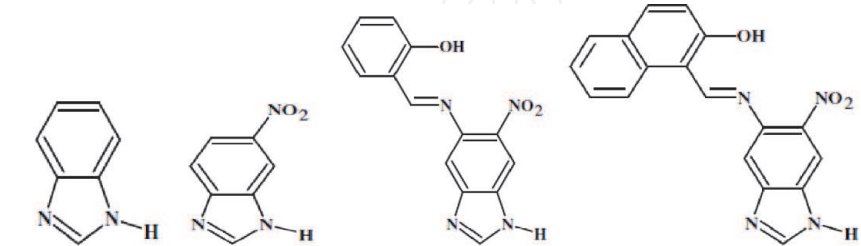
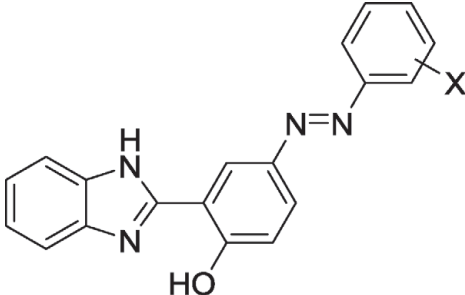
Ramya et al. [18] have studied the synergistic hydrogen-bonded interaction of alkyl benzimidazole derivatives (2-MBI, 2-EBI, 2-PBI), and 1,2,3-benzotriazole (BTZ) and its corrosion protection properties on mild steel in hydrochloric acid at different temperatures have been studied using polarization, EIS, adsorption, surface studies and computational methods. They observed that benzimidazole derivatives and BTZ molecules are effective inhibitors and their inhibition efficiency increases when these two inhibitors are used in combination.

In 2011 Guadalupe et al. [19] analysed the corrosion inhibition properties of BBED via electrochemical (polarization curves and electrochemical impedance spectroscopy) and DFT techniques. Electrochemical impedance data demonstrate that the interface charge capacitance of the electrode with the BBED solution affects the time of charge/discharge of the interface, facilitating the formation of adsorption layer over the iron surface. They also estimated the fractal dimension of the electrode surface in order to understand the nature of the electrode surface. DFT results clearly show that BBED possess corrosion inhibition properties by having a delocalization region (N1=C1=N2) in the benzimidazole ring that gives up its p

Structure	Metal/medium	Reference
 (2-MBI) (2-EBI) (2-PBI)	Mild steel/1 M HCl	[18]
 BBED	Carbon steel/1 M HCl	[19]
 CBO	Mild steel/0.1–1 M HCl	[20]

Structure	Metal/medium	Reference
 <p>BIMDS</p>	Mild steel/1 M HCl and 0.5 M H ₂ SO ₄	[21]
 <p>2,2'-bis(benzimidazole)</p>	Mild steel/1 M HCl	[22]
 <p>2-SH-BI 2-NH₂-BI</p>	Mild steel/1 M HCl	[23, 24]
 <p>BBP BBMS BBMO</p> <p>BBE</p>	Mild steel/1 M HCl	[25]

Structure	Metal/medium	Reference
 <p>ABI BBIA TBIA</p>	Mild steel/1 M HCl	[26]
 <p>MBP PzMBP PMBP</p>	N80 steel/15% HCl	[27]
 <p>Inh I Inh II Inh III</p>	N80 steel/15% HCl	[28]
 <p>MBI TBBI</p>	Cu/1 M HNO ₃	[29]

Structure	Metal/medium	Reference
 <p>MOTBI</p>	Cu/0.02 M NaCl	[30]
 <p>BI 5-NO₂BI BSB NSB</p>	Cu and brass/0.4 M NaCl + 0.1 M NaOH	[31]
 <p>X=o-OCH₃, p-OCH₃, o-CH₃, m-CH₃, m-OCH₃, p-NO₂, m-NO₂ (BPP-1) (BPP-2) (BPP-3) (BPP-4) (BPP-5) (BPP-6) (BPP-7)</p>	Cu/2 M HNO ₃	[32]

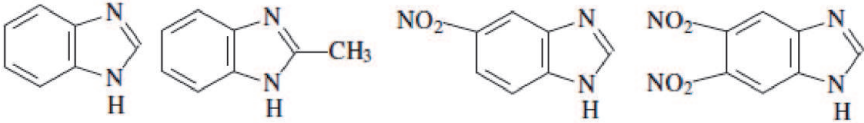
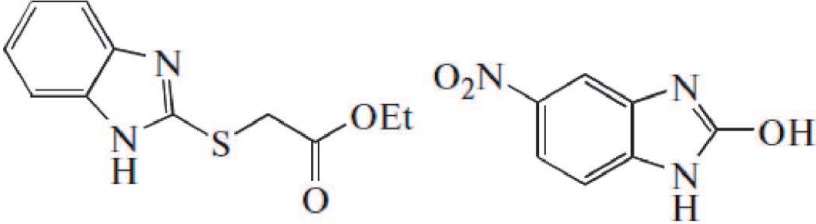
Structure	Metal/medium	Reference
<div><p>BIM 2-CH3BIM 5-NO2BIM 5,6-di NO2BIM</p></div>	Copper, zinc, brass/0.5 M NaCl	[33]
<div><p>MBEA HBI</p></div>	Zn/0.1 M HCl	[34]

Table 1.
The molecular structure of benzimidazole derivatives.

electron density through its HOMO orbital to the metal LUMO to form an adsorption layer over the metallic surface.

The influences of a benzimidazole derivative, namely, 1,8-bis(1-chlorobenzyl-benzimidazolyl)-octane (CBO), on the corrosion behaviour of mild steel in different concentration HCl solutions was studied by Wang et al. [20]. The authors conclude that CBO acted as an excellent mixed-type inhibitor and strongly adsorbed via chemical bond formation. The data obtained from weight loss and electrochemical measurements have shown that the CBO has the excellent inhibiting properties for mild steel in HCl solution.

Ahamad and Quraishi [21] studied the corrosion inhibition effects of a water-soluble benzimidazole derivative, namely, bis(benzimidazol-2-yl)disulphide (BIMDS) on mild steel 1.0 M HCl and 0.5 M H₂SO₄. The results reveal that BIMDS showed a better performance in 0.5 M H₂SO₄ solutions than in 1.0 M HCl. Polarization measurements indicated that BIMDS is a mixed-type inhibitor in both acid media. The inhibitor obeyed the Langmuir adsorption isotherm model in both acid media.

Abboud et al. [22] studied the 2,2'-bis(benzimidazole) as a corrosion inhibitor for mild steel in 1 M HCl. The analysis of polarization curves showed that the mechanism of corrosion inhibition remains unchanged after the addition of the inhibitor and the inhibitor acts as a mixed type. The corrosion inhibition of the inhibitor can be interpreted by a simple blocked fraction of the electrode surface related to the adsorption of the inhibitor species according to a Langmuir isotherm on the steel surface.

Popova et al. and Mahdavian et al., respectively, in 2004 and 2010 studied the corrosion inhibition property of a series of benzimidazole derivatives [23, 24]. The results of Popova et al. reveal that the inhibition efficiency increased with the increase of organic substrate concentration, while the adsorption followed the Frumkin isotherm. They also suggest that there was no correlation observed among the studied various parameters of the electronic structure and the corrosion inhibition efficiency. Likewise Mahdavian results reveal that the studied inhibitor reduced both the cathodic and anodic polarization curves and behaved as a mixed-type inhibitor. The change in the values of the impedance parameters suggests the adsorption of the inhibitor on the metal surface. In surface analysis like SEM/EDX, the presence of sulphur confirmed the adsorption of the inhibitor on the mild steel surface.

Dutta et al. [25] in 2015 studied the corrosion inhibition of bis-benzimidazole type derivatives for mild steel with an immersion of 4 days in 1 M HCl. The results of the potentiodynamic polarization studies reveal that inhibitors produce a mixed-type protection but mostly affect the cathodic reaction.

2.1.2 Computational results

The approach of DFT results were used to probe into the interaction modes for the inhibitor molecules on the steel surface. The MDS was used to analyse the configuration of equilibrium adsorption of inhibitors. All the interaction energies between the inhibitors and the metal surface were deduced in the aqueous phase.

Cao et al. [26] performed the comparative theoretical study using DFT and MSD for 2-aminomethyl benzimidazole (ABI), bis(2-benzimidazolylmethyl) amine (BBIA) and tri-(2-benzimidazolylmethyl) amine (TBIA) on the mild steel surface. They concluded that the three-inhibitor molecules showed similar ability for the electron donation, while the difference in the electron-accepting ability makes them give a different inhibition performance. MD simulations suggest that the steric effect among the inhibitor molecular structures significantly affects the adsorptive

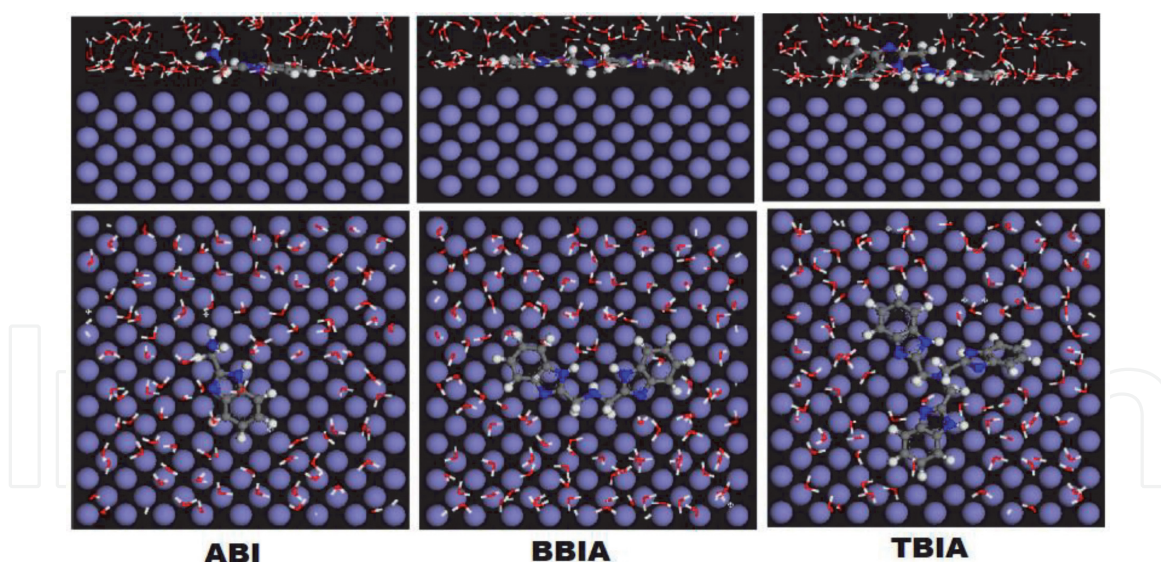


Figure 4.
Equilibrium configurations of ABI, BBIA and TBIA molecules.

configurations over the Fe (1 0 0) surface. **Figure 4** represents the equilibrium adsorption configurations of inhibitors on Fe (1 0 0) in the aqueous phase.

Yadav et al. [27, 28] group studied the corrosion inhibition of 2-(1-(morpholinomethyl)-1Hbenzo[d]imidazol-2-yl)phenol (MBP), 2-(1-((piperazine-1-yl)methyl)-1H-benzo[d]imidazol-2-yl)phenol (PzMBP), 2-(1-((piperidine-1-yl)methyl)-1H-benzo[d]imidazol-2-yl)phenol (PMBP), 4-(phenyl)-5-[(2-methyl-1H-benzimidazol-1-yl)methyl]-4H-1,2,4-triazole-3-thiol (Inh I), 4-(4-methylphenyl)-5-[(2-methyl-1H-benzimidazol-1-yl)methyl]-4H-1,2,4-triazole-3-thiol (Inh II) and 4-(4-methoxyphenyl)-5-[(2-methyl-1H-benzimidazol-1-yl)methyl]-4H-1,2,4-triazole-3-thiol (Inh III) on N80 steel in 15% HCl solution using weight loss measurement, potentiodynamic polarization and electrochemical impedance spectroscopy (EIS) techniques. They observed that inhibition efficiency of all the inhibitors increases with increase in concentration and decreases with increase in temperature. Furthermore the steel surface was analysed by SEM and AFM techniques. The density functional theory was employed for the theoretical evaluation of the studied inhibitors. **Figures 5** and **6** represent the AFM and SEM micrograph of the N80 steel surface.

The inhibitive actions of 2-mercaptobenzimidazole (MBI) and 2 thiobenzylbenzimidazole (TBBI) on copper corrosion in 1 M HNO₃ medium were studied, using weight loss method, at 25–65°C and in the concentrations of the range of 5×10^{-5} M to 10^{-3} M by Niamien et al. group in 2011 [29]. They reported the inhibition efficiencies of 90.0% for TBBI and 87.7% for MBI at 25°C. Negative values of changes in free energies indicate the spontaneous adsorption process with a combination of both physical and chemical processes. They have done the correlation between theoretical data and experimental results using DFT/B3LYP/6-31G (d,p) method.

Rao et al. [30] demonstrated the self-assembled (SAM) monolayer formation of 5-methoxy-2-(octadecylthio)benzimidazole (MOTBI) on fresh copper surface. The MOTBI SAM on copper surface was characterized by contact angle measurements, X-ray photoelectron spectroscopy and reflection absorption FTIR spectroscopy, and it is inferred that chemisorption of MOTBI on copper surface is through nitrogen. The corrosion protection ability of MOTBI SAM was evaluated in aqueous NaCl solution using impedance, electrochemical quartz crystal nanobalance, potentiodynamic polarization and weight loss studies.

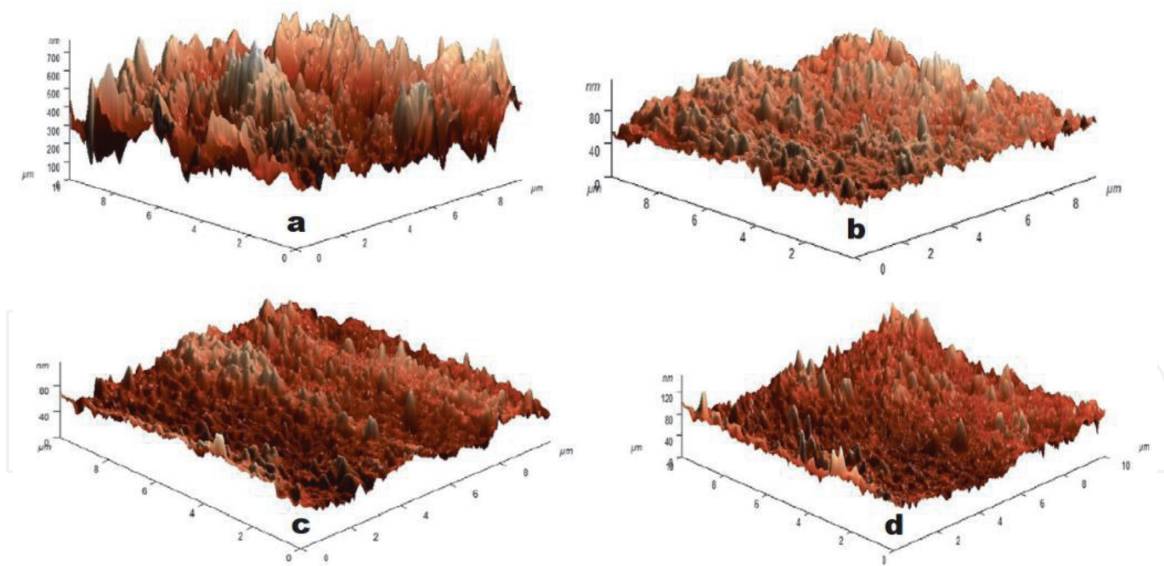


Figure 5.
AFM micrograph (a) without inhibitor, (b) with PzMBP, (c) with MBP and (d) with PMBP.

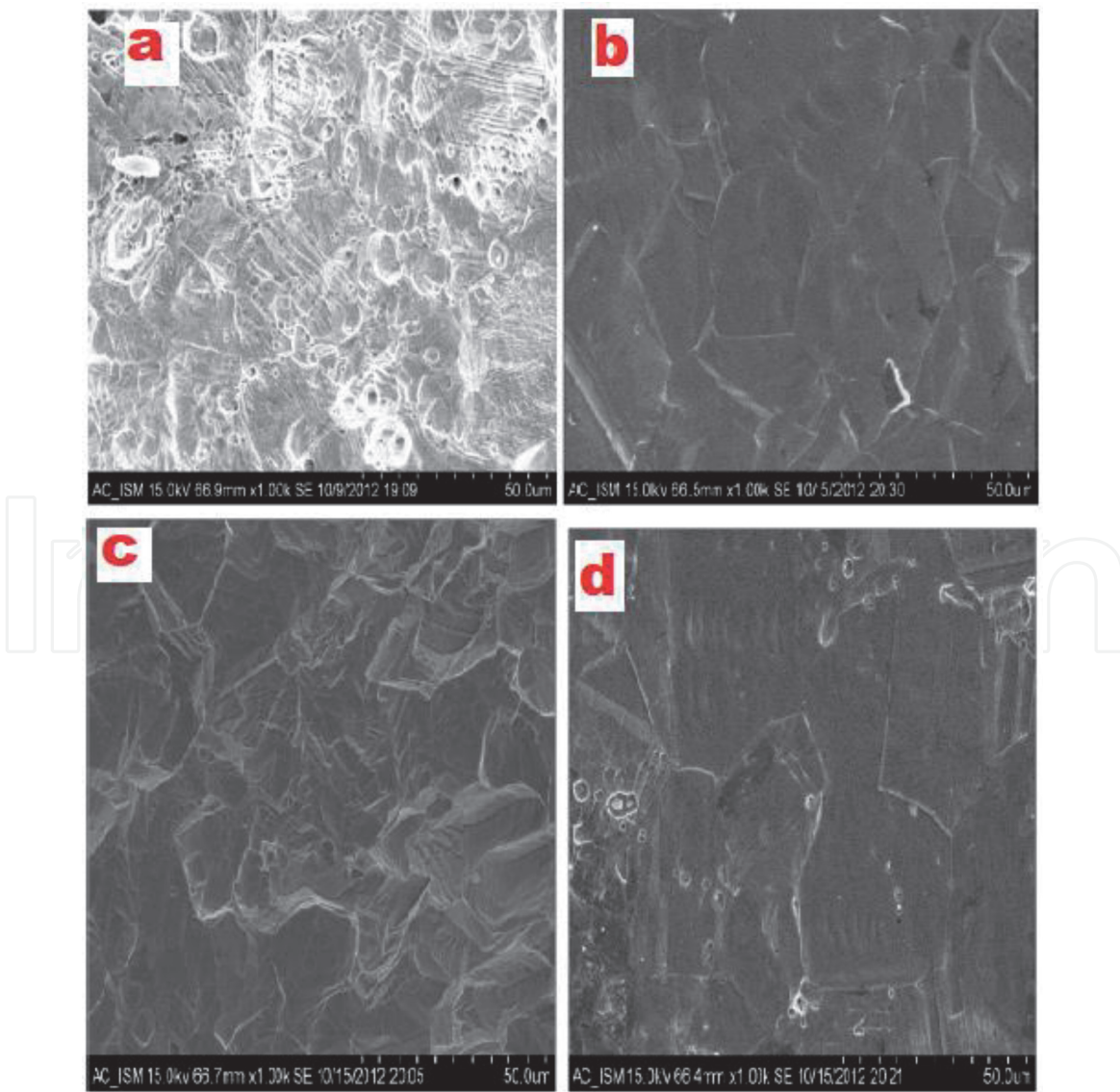


Figure 6.
SEM micrograph (a) without inhibitor (b) Inh III, (c) Inh II and (d) Inh I.

In 2014, Özbay et al. [31] have carried out the inhibitive action of newly synthesized ortho-hydroxy Schiff bases of 5-amino-6-nitro-1H-benzimidazole against the corrosion of copper and brass in alkaline medium using potentiodynamic polarization and electrochemical impedance spectroscopy. The results showed an inhibition efficiency of 91.0% and 97.4% for copper and brass, respectively.

Madkour and Elsham [32] studied the inhibitive performance of seven synthesized 2-(2-benzimidazolyl)-4 (phenylazo) phenol (BPP_1–7) derivatives which were investigated experimentally on the corrosion of copper in 2.0 M HNO_3 acid using mass loss and thermometric and DC potentiodynamic polarization techniques. Quantum chemical calculations were investigated to correlate the electronic structure parameters of the investigated benzimidazole derivatives with their inhibition efficiencies values.

Yanardag et al. [33] have studied the corrosion inhibition property of 1H-benzimidazole, (BIM), 2-methyl-1H-benzimidazole (2- CH_3BIM), 5-nitro-1H-benzimidazole [5(6)- NO_2BIM] and 5(6)-dinitrobenzimidazole (5,6-di NO_2BIM) for copper, zinc and brass in alkaline and neutral media. The results under the experimental conditions suggest the efficiency order of the inhibitors is $\text{BIM} > 5(6)\text{-NO}_2\text{BIM} > 5,6\text{-diNO}_2\text{BIM} > 2\text{-CH}_3\text{BIM}$ in alkaline media (pH: 13) and $2\text{-CH}_3\text{BIM} > 5(6)\text{-NO}_2\text{BIM} > \text{BIM}$ in the 0.5 M NaCl solution.

The corrosion inhibition characteristics of 2-mercaptobenzimidazole (MBI), 2-mercapto benzimidazolyl-ethyl acetate (MBEA), 2-hydroxy benzimidazole (HBI) and 2-hydroxy 5-nitro benzimidazole (HNBI) on zinc corrosion in 0.1 M hydrochloric acid were investigated by weight loss and potentiodynamic polarization techniques by the group of Shanbhag et al. [34]. The inhibition efficiency

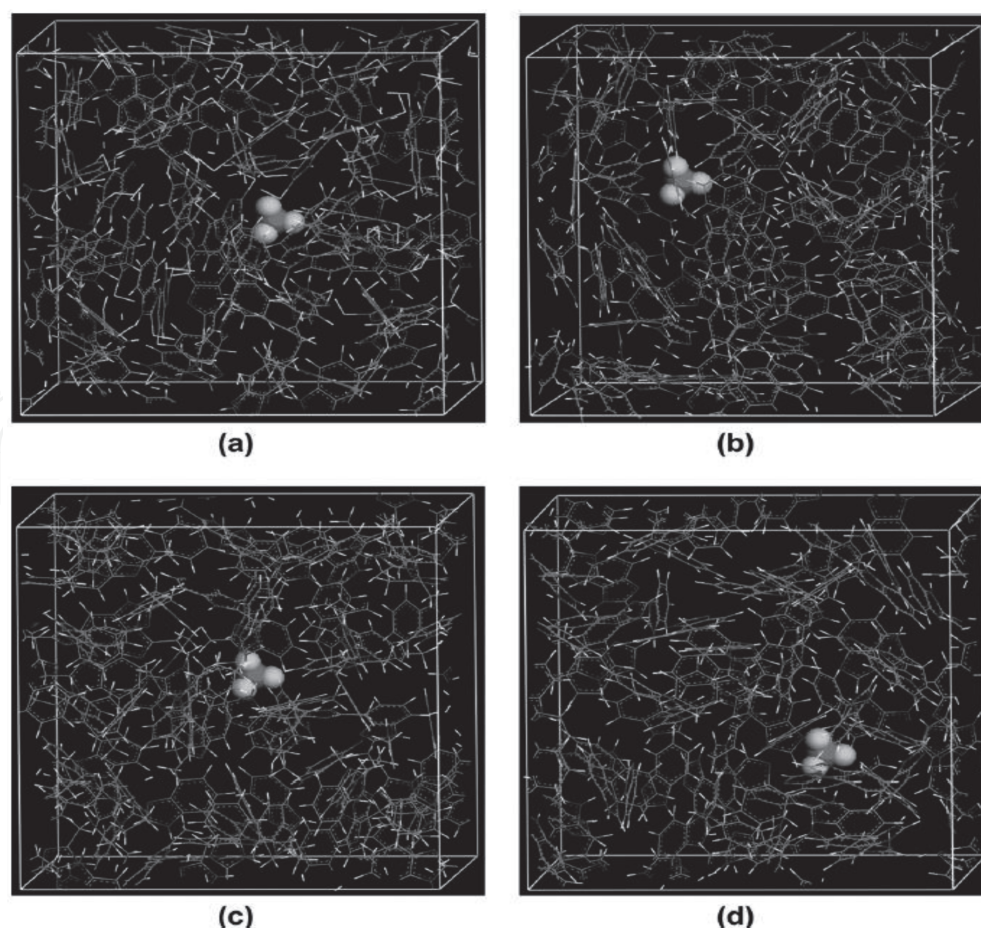


Figure 7.
Simulation models of H_3O^+ (shown in CPK) in various inhibitor films (shown in line): (a) 2-SH-BI, (b) 2- $\text{NH}_2\text{-BI}$, (c) 2- $\text{CH}_3\text{-BI}$ and (d) BI.

increased with an increase in inhibitor concentration but decreased with an increase in temperature. The adsorption of MBI and MBEA obeyed Langmuir's adsorption isotherm, but HBI and HNBI followed Temkin's adsorption isotherm. The existence of inhibitor film on metal surface was established by scanning electron microscopy (SEM) images.

Zhang et al. [35] studied the diffusion of corrosive particles inside inhibitor films consisting of 2-mercaptobenzimidazole (2-SH-BI), 2-aminobenzimidazole (2-NH₂-BI), 2-methylbenzimidazole (2-CH₃-BI) and benzimidazole using molecular dynamics simulation (MDS). Diffusion coefficients of various corrosive particles in the films were calculated, and their order is 2-SH-BI < 2-NH₂-BI < 2-CH₃-BI < BI. Fractional free volume, interaction between corrosive particles and films and mobility of films were also investigated to illustrate the microscopic diffusion mechanism. As a result, it can be inferred that the order of inhibition efficiency is 2-SH-BI > 2-NH₂-BI > 2-CH₃-BI > BI. The simulation models of H₃O⁺ in various inhibitor films are shown in **Figure 7**.

Recently Singh et al. [36] have studied the corrosion inhibition effect of three synthesized benzimidazole derivatives, namely, 2-(3,4,5-trimethoxyphenyl)-1H-benzo[d]imidazole (TMI), 2-(3,4-dimethoxyphenyl)-1H-benzo[d]imidazole (DMI) and 2-(4-methoxyphenyl)-1H-benzo[d]imidazole (MMI), for J55 steel saturated with CO₂ in a 3.5% NaCl solution. The author has analysed the corrosion inhibition property using weight loss, impedance spectroscopy (EIS) and potentiodynamic polarization methods. The metal surface was examined by scanning electron microscope (SEM) and X-ray photoelectron spectroscopy (XPS). They have justified the experimental results using the DFT and MD studies. Their investigations both by experimental and theoretical analyses suggest that as the number of methoxy groups increases, the corrosion protection ability of the inhibitors increases, and thus TMI is the best inhibitor. The results of the Tafel curves are presented in **Figure 8**. As observed from the curves, inhibitor TMI exhibited a

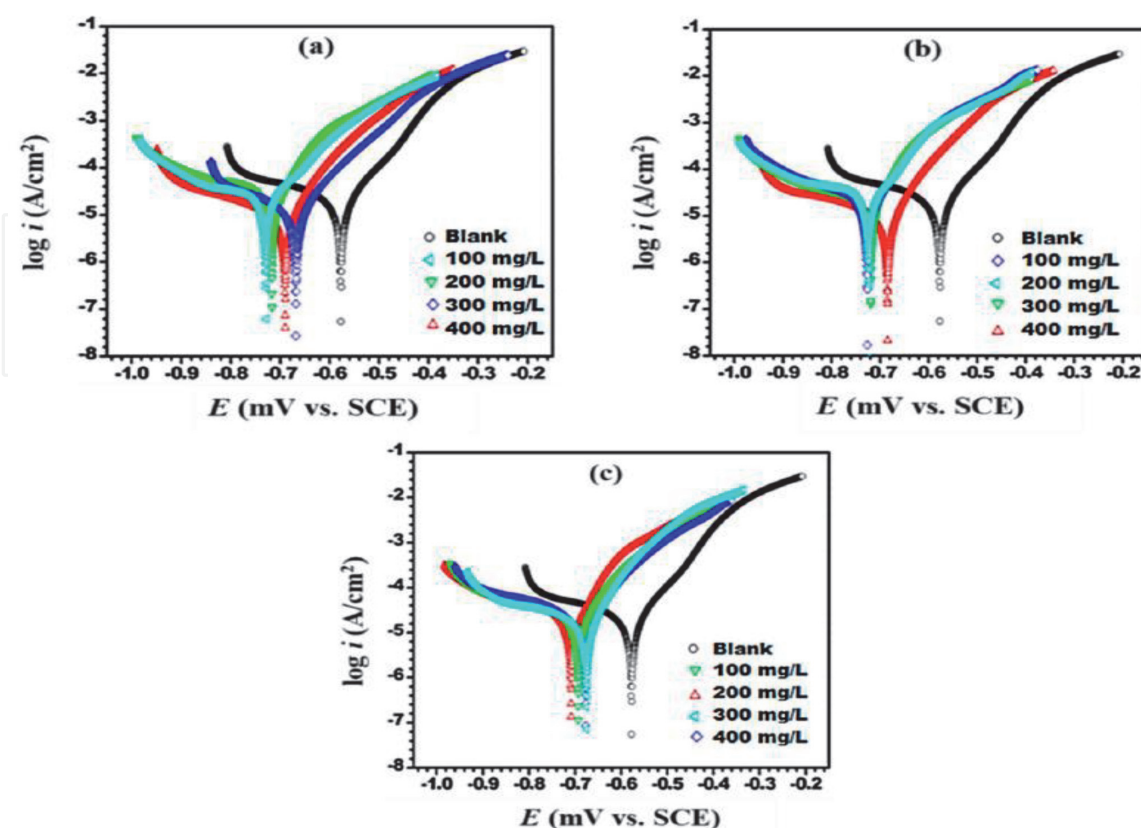


Figure 8.

Tafel curves in without and with different concentrations of inhibitors: (a) TMI; (b) DMI; (c) MMI.

superior inhibition as compared to DMI and MMI, and the adsorption of these molecules on the metal surface affected both anodic and cathodic reactions. The three studied compounds can significantly reduce the corrosion current density values, and they exhibit mixed-type inhibition characteristics. The values of corrosion current density (i_{corr}) without addition inhibitors are $104.4 \mu\text{A cm}^{-2}$, and the addition of inhibitors reduced it to $5.5 \mu\text{A cm}^{-2}$ (TMI), $13.2 \mu\text{A cm}^{-2}$ (DMI) and $21.5 \mu\text{A cm}^{-2}$ (MMI).

3. Conclusion

The book chapter gives an overview of the role of benzimidazole and their derivatives in the corrosion mitigation of industrially important alloys such as mild steel, N80 steel, aluminium, zinc, copper and brass in various aggressive media. Here we have also tried to explore the molecular structure, synthesis protocol and some important biological activity of benzimidazole derivatives. In view of the literature analysis, it could be concluded that benzimidazole derivatives have the potential ability to act as a suitable additive for various corrosive solutions.

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Author details

Ambrish Singh^{1*}, Kashif R. Ansari², Dheeraj S. Chauhan², Mumtaz A. Quraishi², Savas Kaya³, Hua Yu⁴ and Yuanhua Lin¹

¹ School of New Energy and Materials, Southwest Petroleum University, Chengdu, Sichuan, China


² Centre of Research Excellence in Corrosion, Research Institute, King Fahd University of Petroleum and Minerals, Dhahran, Saudi Arabia

³ Department of Chemistry, Sivas Cumhuriyet University, Turkey

⁴ Institute of Photovoltaics, Southwest Petroleum University, Chengdu, Sichuan, China

*Address all correspondence to: vishisingh4uall@gmail.com

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