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Elucidating the corrosion of carbon steel in hybrid monoethanolamine solutions containing methanol or n-methyl-2-pyrrolidone

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Abstract

Corrosion has been a persistent issue in carbon dioxide (CO_2) absorption unit that needs to be resolved in order to prolong the life span of the absorber unit. In this regard, understanding the corrosion behaviour, particularly in hybrid monoethanolamine (MEA) solutions, is crucial. This study aims to give detail information on the role of different process parameters, encompassing types of amine solution, temperature, and CO_2 loading on the corrosion behaviour of carbon steel in system containing hybrid solution of monoethanolamine (MEA) and methanol (MeOH) or N-methyl-2-pyrrolidone (NMP). Here, the corrosion extent of the carbon steel was evaluated using gravimetric experiments. The results indicated that the corrosion rate of carbon steels, when submerged in various amine solutions, increased with higher solution temperatures and the presence of CO_2. In line with the Raman spectroscopy results and the surface morphology analysis, results also showed that the carbon steel coupons immersed in MEA+MeOH had the lowest corrosion rate.

1. Introduction

Carbon dioxide (CO_2) is one of the key acid gases that need to be extracted from gas streams like natural gas in order to meet industrial sales specifications. Additionally, removing CO_2 from flue gas is essential for controlling greenhouse gas emissions, which contribute to climate change [1-3]. Various strategies, including both chemical and physical absorption, have been used to remove CO_2 gas; chemical absorbents are more frequently employed than physical absorbents because they excel in CO_2 absorption, even under low CO_2 partial pressures [4,5]. Common alkanolamine used for this purpose includes monoethanolamines (MEA), diethanolamine (DEA), methyldiethanolamine (MDEA), and di-2-propanolamine (DIPA) [3]. Nevertheless, the utilization of alkanolamine may subsequently lead to material corrosion. In fact, corrosion would happen in the tubes and vessels when they are in contact with these chemical solutions [6]. Due to its economic viability, carbon steel has been usually employed as the construction material for various industrial facilities, including chemical absorption units [7]. Carbon steel represents nearly 85% of the global annual steel output, largely because of its lower cost compared to other alloy types [8].

Carbon steel corrosion is an undesirable phenomenon that leads to the deterioration of unit operations and pipelines in plants. Since investment in plant equipment or systems usually involves substantial amount of expenditure, this makes understanding the corrosion behavior of carbon steel as crucial. Different forms of corrosion could occur in amine-based CO_2 absorption units, and the presence of H_2S may create an even more complex corrosive environment [9]. Among the corrosion type which could occur are intergranular corrosion, uniform corrosion, pitting corrosion, stress corrosion cracking (SCC), erosion-corrosion, and crevice corrosion [10,11]. It would pose significant detrimental effects on the costing, as well as the health and safety of the workers. In particular, equipment downtime could cause plants to experience production losses hence leading to revenue losses, incur repair costs for corroded components, and corroded equipment may lead to a shortened lifespan [11,12].

In recent years, extensive research has been conducted on a hybrid formulation to enhance the CO_2 absorption efficiency of the chemical absorption process. It acts as an alternative to the conventional alkanolamine aqueous solvent and may even facilitate the removal of multiple acid gases at the same time. A hybrid solvent combines chemical and physical absorbents wherein its ability to capture CO_2 is enhanced [13]. Hybrid solvents encompassing MEA, dimethylethanolamine (DMEA) with acetal compounds, or methyl-1-propanol (AMP), have been conducted for CO_2 absorption. The 2,5,7,10-tetraoxaundecane (TOU), one of the acetal compounds, has been utilized as the physical absorbent and found to significantly enhanced the CO_2 absorption performance [14]. In another study, it was reported that CO_2 removal from natural gas using MEA aqueous solvent can be enhanced by introducing N-methyl-2-pyrrolidone (NMP) into the solution [15].

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In addition, the CO_2 absorption performance can be enhanced through the uses of hybrid solvent of diethanolamine (DEA) and methanol (MeOH) [16]. Based on those findings, MEA is considered the first-generation standard and most frequently used amine in hybrid systems designed for CO_2 absorption. High reactivity, good thermal stability, ease of process operation, and fast absorption rate are among the key advantages offered by the alkanolamine (or MEA specifically), making it widely used in CO_2 absorption applications. Nevertheless, MEA is corrosive especially after certain amount of CO_2 is captured [17,18]. Although hybrid solvents have demonstrated improved CO_2 absorption performance, the elevated corrosion potential of MEA may restrict its use in capturing CO_2. Hence, it is imperative to understand their corrosion behavior in relation to carbon steel before further expanding the usage of MEA hybrid. The extent of corrosion in carbon steel can be determined using the gravimetric method, where the percentage of weight loss is calculated based on the difference between the weight of the fresh and corroded carbon steel [19].

Past studies have demonstrated the corrosion effects on carbon steels immersed in alkanolamine solutions by investigating the impact of factors such as CO_2 loading, water content, and process temperature [20,21]. However, there is a lack of information on the corrosion behaviour of alkanolamine hybrid solvents in the literature, which serves as the research gap. Closing this research gap is imperative for future development of engineering materials that can withstand such corrosive conditions during post-combustion carbon capture and natural gas purification processes. The main goal of this study is to disclose the corrosion behaviour of carbon steel coupon upon its exposure to alkanolamine hybrid solvent. Here, the solvents consist of either MeOH or NMP combined with the commonly used MEA. It is expected that NMP can improve the removal of CO_2 from the acid gases [15]. On the other hand, the presence of MeOH may enhance the CO_2 diffusivity and solubility in the solvent as the deprotonation rate reduced [22]. The extent of corrosion was determined through gravimetric method and its mechanism was correlated to the physico-chemical properties of the carbon coupons obtained through Raman spectroscopy and scanning electron microscopy analyses.

2. Experimental

2.1 Materials

Standard carbon steel coupons of C1018 mild carbon steel used in this study was supplied by Metal Samples (USA). The surface area of the coupon was 21.8 cm^2. For the preparation of MEA solutions, MEA (purity: 99.9% Merck), NMP (purity: 99.98%, Merck), MeOH (purity 99.98%, Merck) and distilled water were used. Water was distilled according to the type IV Specification D 1193 [23] for the blending of MEA solution. CO_2 tank (purity >99%, Air Product) was used to bubble the CO_2 into the solutions for experiments with CO_2 loaded solutions.

2.2 Solution composition

In this study, five different MEA solutions, namely 20 wt% MEA + 80 wt% H_2O (MEA + H_2O solution), 20 wt% MEA + 40 wt% NMP + 40 wt% H_2O (MEA + NMP + H_2O solution), 20 wt % MEA + 80 wt% NMP (MEA + NMP solution), 20 wt% MEA + 40 wt% MeOH + 40 wt% H_2O (MEA + MeOH + H_2O solution) and 20 wt% MEA + 80 wt% MeOH (MEA + MeOH solution) were prepared as the solvents to be investigated.

2.3 Process parameters

The effect of three parameters, i.e. immersion time (7, 14, 21, and 28 day), temperature (25°C, 50°C, and 60°C) and the presence of CO_2 loading in solutions were investigated to determine their respective impacts on the corrosion of carbon steel when different MEA solutions were used. For each condition, duplicate carbon steel corrosion testing was performed, and the average corrosion rate was calculated.

2.4 Surface morphology analysis

Fresh carbon steel coupon and carbon steel coupons which had been immersed in the 5 types of MEA solution for 28 day were subjected to surface morphology analysis to investigate the changes in the carbon steel microstructure before and after it contacted with the MEA solution. The surface morphology of carbon steel coupons was characterized by Field Emission Scanning Electron Microscopy (FE-SEM) (Helios NanoLab G3, FEI, Australia) under the condition with 1 kV beam energy with current of 13 pA. The overall spectra were recorded in the energy range of 10 keV to 0 eV.

2.5 Corrosion rate analysis

American Society for Testing & Materials (ASTM) method G31-72 [23] was adopted as the standard methodology for corrosion testing. The immersion corrosion analysis was performed to access the corrosion rate of carbon steel coupons. The ASTM standard defined the procedures including the carbon steel coupons preparation, test condition, apparatus and material, method of cleaning immersed coupons, technique for evaluating and quantifying of corrosion rate based on gravimetric techniques.

Prior to the experiment, all the standard corrosion testing carbon steel coupons were checked to make sure they were in good condition, correctly tagged and identified. The physical characteristics of each specific carbon steel coupon such as initial mass, dimensions, and initial surface condition were recorded. Acetone was used to remove the grease on the surface of the carbon steel coupons. The coupons were dried with lint free tissue and an analytical balance (ME104T/00, Mettler Toledo, United States of America) was utilized to determine the final mass of the carbon steel coupons. The final dimension of the carbon steel coupons was also determined using a calliper tool. The total surface area was then determined for corrosion rate calculation. Equation (1) was used to compute the corrosion rate (MPY):

$$\text{Corrosion rate (MPY)} = \frac{(M_o - M_i)}{\rho \times A} \times K \qquad (1)$$

where M_o is the initial weight (g) of carbon steel coupon before the experiment while M_i is the weight (g) of the carbon steel after the experiment. K is factor constant, 3.45 $cm^2 \times 106\ cm^2$; ρ is carbon steel coupoun's density, 7.86 $g{\cdot}cm^{-3}$ and A is the exposed area of carbon steel coupon [23].

2.6 Raman spectroscopy analysis

All five types of MEA solution, with and without CO_2 loading were analysed using Raman spectroscopy (Thermo Fisher Scientific, DXR Smart Raman Spectroscopy, United States of America) to analyse the changes in chemical component which could occur in the solution after the bubbling with CO_2 as this might potentially affect the carbon steel corrosion behaviour.

3. Results and discussion

3.1 Role of immersion time and MEA types on the corrosion behaviour

Prior to the surface morphology analysis, the corrosion rate of carbon steel coupons immersed in the five types of MEA solutions was investigated. This was done to obtain a preliminary idea of the corrosion behaviour of carbon steel in contact with the MEA-based solution (without taking into account process parameter variations like CO_2 loading and temperature). Figure 1 shows the corrosion rate after 7, 14, 21, and 28 day of immersion at room temperature.

The results indicated that at the interval of 7 day, the average corrosion rate of coupon immersed in MEA + H_2O was 2.010 mils per year (MPY) whereas the corrosion rate of coupon immersed in MEA + NMP + H_2O was 1.990 MPY. Meanwhile, the corrosion rate of carbon steel coupons immersed in MEA + NMP, MEA + MeOH + H_2O, and MEA + MeOH was 1.920 MPY 0.150 MPY, and 0.120 MPY, respectively. Ultimately, at the interval of 28 day, the corrosion rate of the coupons immersed in MEA + H_2O had the most significant corrosion rate which was 2.440 MPY. Corrosion rate of coupons in MEA + NMP + H_2O was 2.300 MPY while in MEA + NMP, the corrosion rate was 2.140 MPY. However, minor changes of corrosion rate were observed after 28 day in the carbon steel coupons immersed in MEA + MeOH + H_2O and MEA + MeOH, with corrosion rates of 0.222 MPY and 0.150 MPY, respectively.

The immersion of carbon steel coupons in MEA aqueous solution were observed to consistently resulting in the highest corrosion rate in each immersion period, while coupons immersed in MEA hybrid solution consisting of MeOH experienced the lowest corrosion rate. The results implied that the existence of H_2O could act as an electrolyte to promote the corrosion process. This result is within expectation as water is the most common electrolyte associated with corrosion [24]. However, when carbon steel coupons were exposed to an aqueous solution after a certain period of time, corrosion products would form on the surface. These corrosion products could prevent further corrosion by acting as a barrier between the environment and surface of the carbon steel coupons, especially when they were insoluble in the solution [25]. Therefore, carbon steel coupons immersed in MEA + H_2O and MEA + NMP + H_2O experienced a noticeable increasing corrosion rate from the interval between 7 day to 14 day. However, from the period of 14 day to 28 day, slower corrosion rates were observed in these solutions. Immersion of the carbon steel in MEA + MeOH + H_2O led to insignificant corrosion between the period of 7 day to 28 day. Hence, it is deduced that the presence of organic solvent, MeOH, could significantly reduce the carbon steel coupon corrosion.

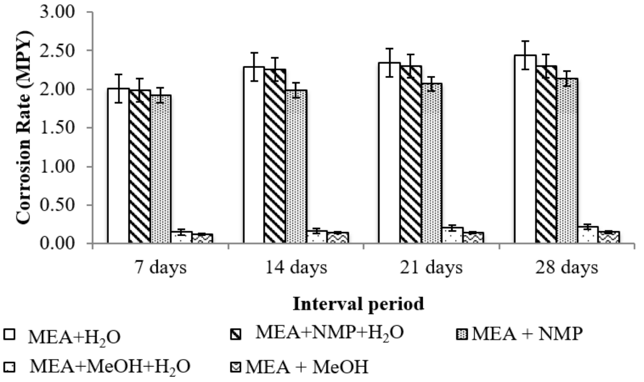

Figure 1. The rate of corrosion observed on carbon steel coupons immersed in various types of MEA-based solutions.

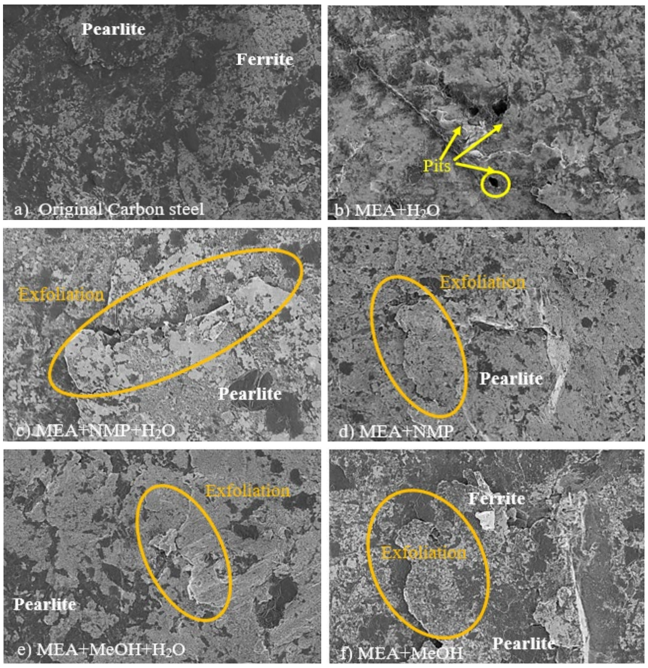

Figure 2. FESEM images at 1000x magnification showing carbon steel coupons: (a) original; post 28 day immersion in (b) MEA + H_2O; (c) MEA + NMP + H_2O; (d) MEA + NMP; (e) MEA + MeOH + H_2O; and (f) MEA + MeOH.

The evolution of carbon steel microstructure played a vital role, especially when corrosion had taken place. Figure 2 shows the surface morphologies of carbon steel coupons immersed in five types of MEA solution for 28 day. Carbon steel consists of ferrite and pearlite [26]. Figure 2(a) shows the smooth surface of carbon steel before the immersion in any of the MEA solution. As shown in Figure 2(b-e), exfoliated surfaces were observed on carbon steel coupons immersed in MEA solutions for 28 day. Exfoliation is a form of intergranular corrosion that spreads laterally from the initiation sites along planes parallel to the surface. These formed corrosion products had greater volume than the parent metal. This phenomenon forced the layers apart and caused the metal to exfoliate or delaminate by giving rise to a layered appearance. In addition, it was found that the surface of the carbon steels shown in Figures 2(b-f) is thicker and rougher compared to the fresh carbon steel (Figure 2(a)), indicating the deposition of iron oxide. However, the thickness of the deposition is difficult to be assessed due to the top view of the SEM image and surface irregularities.

Although the microstructures of the immersed carbon steel coupon were almost identical to the corrosion product formed on the surface of the coupon, the thickness of the corrosion product might impact the corrosion rate, depending on the exposure time [27]. It was because these corrosion products would deposit on the carbon steel surface, resulted in lower corrosion rate significantly [28]. It could be seen from Figure 2(b) that corrosion pits formed on the carbon steel surface after immersion in MEA + H_2O solution. The pits would enlarge over time, aggravating the corrosion; this aligns with the increases of corrosion rates from 2.010 MPY to 2.440 MPY (21.4% increment) from 7 day to 28 day of immersion, as shown in Figure 1. Hence, porous corrosion products were observed on the surface of the coupon. The occurrence would usually go through the three major stages, namely pit nucleation, pit growth, and passivation through formation of insoluble corrosion products. Yet, the robustness of the passivation film would determine the corrosion behaviour, such as whether it could stop continuous corrosion or cause the corrosion condition to worsen [29]. Meanwhile, there was no pit spotted in Figures 2(c-f) for carbon steel immersed in MEA hybrid solutions. Nonetheless, corrosion scales were observed to develop on the surface of the coupons. These corrosion scales could reduce the acceleration of the corrosion reaction by slowing down the solution diffusion. Owing to this, the corrosion rate appears to be lower when the carbon steel was immersed in MEA + NMP + H_2O, MEA + NMP, MEA + MeOH + H_2O, and MEA + MeOH, as delineated in Figure 1. The porosity of the corrosion scales was the key to determine the rate of solution transport across the carbon steel to induce corrosion [27].

MEA solutions containing MeOH resulted in significantly lower corrosion rate compared to other solutions because MeOH is a protogenic solvent which could auto-disassociate in equilibrium as follows:

$$2CH_3OH \leftrightarrow CH_3(OH)_2^+ + CH_3O^- \qquad (2)$$

The polar methanol molecule would adsorb to the surface of the carbon steel [39]. The stability of the protective layer on the metal surface increased with the increased amount of methoxyl (CH_3O^-) formation. Mixing alcohol and water would also entail the two molecules to combine to one another [30]. The structure of liquid MeOH under room temperature was like a chain made up of a combination of 6 or 8 MeOH molecules. When water molecules were present, the MeOH chain interacted with water molecule and formed clusters of different sizes. These clusters were held together by hydrogen bonds and more importantly, could act as a protective structure, adhering on the carbon steel surface. Hence, this explained a significantly low corrosion rate for carbon steel coupons immersed in the solutions containing MeOH.

3.2 Role of temperature on the corrosion behaviour

Figure 3 shows the corrosion rate of carbon steel immersed in the five types of MEA solution for 7 day at 25°C, 50°C and 60°C. The corrosion rate of the carbon steel coupons increased when the temperature of the MEA solutions was increased from 25°C to 50°C. At 25°C, the corrosion rate of the carbon steel coupons immersed in MEA + H_2O, MEA + NMP + H_2O and MEA + NMP were 2.010 MPY, 1.990 MPY and 1.920 MPY, respectively. By increasing the medium temperature to 50°C, the corrosion rate was found to increase to 2.881 MPY, 2.582 MPY and 2.414 MPY, respectively. However, there is no significant increase in the corrosion rate when the temperature was raised from 50°C to 60°C. For the carbon steel immersed in MEA + MeOH and MEA + MeOH + H_2O, a slight increase of corrosion rate was found when the medium temperature was increased from 25°C to 60°C. These values are lower than those reported in the literature [31], which found that the corrosion rates of MEA and its mixtures with methyldiethanolamine (MDEA) or 2-amino-2-methyl-1-propanol (AMP) may range from 77.6 MPY to 136.4 MPY. This difference is primarily attributed to higher operating temperatures and higher CO_2 content used in the study as compared to the present work. Despite that, the results shown in Figure 3 are aligned with previously reported findings which indicating that blended amine solutions may exhibit lower corrosion rates compared to single amine solution [32,33].

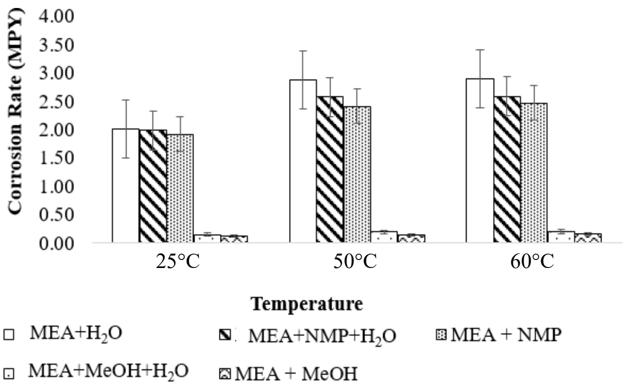

Figure 3. Corrosion rate versus different types of solution at different temperatures.

Based on the results in Figure 3, it is observed that there was a tendency for the rate of corrosion reaction to increase when the temperature was raised above the ambient temperature, which is consistent with the Arrhenius law. Previous research found that the temperature ranges of –20°C to +10°C had a linear relationship with corrosion rate, and that the reaction rate increased by about 10 times with increasing temperature. However, further increasing the temperature from 10°C to 20°C increased the corrosion rate by 7 times [34,35]. This demonstrated that the quantum of increase in corrosion rate with increasing temperature did not follow a linear relationship. It was observed that raising the ambient temperature from 15°C to 20°C led to an approximate 50% increase in the corrosion rate [36]. Hence, the increase of corrosion rate observed in this study when the temperature was increased from ambient temperature was aligned with the observation by other researchers.

In this study, two different situations were observed. Firstly, the corrosion rates of carbon steel coupons immersed in MEA + MeOH and MEA + MEA + MeOH + H_2O were slightly increased from 25°C to 60°C, whereas the corrosion rate of carbon steels immersed in MEA + H_2O, MEA + NMP + H_2O and MEA + NMP increased initially when temperature was increased from 25°C to 50°C, but the corrosion rate remained constant when the temperature was further elevated to 60°C. Based on the Arrhenius' law, increasing the temperature could increase the molecules collision between the solutions and carbon steel coupons. This consequently increased the corrosion rate of the carbon steel coupon. Therefore, it was not surprising that the corrosion rate increased when the temperature was increased from 25°C to 50°C. However, since the corrosion product could have formed and acted as a protective film on the surface, it eventually placed the carbon steel surface in a passive state. This could be one of the plausible explanations for the insignificant corrosion when the temperature was further increased beyond 50°C.

Secondly, it could be observed that the carbon steel immersed in MeOH containing solutions did not experience significant corrosion compared to the other three types of MEA solution. A recent study had established that the methanolic compound had the ability to reduce the carbon steel corrosion rate. It was believed that the corrosion behaviour was attributed to the covered surface area of the carbon steel by the adsorbed methanolic compound. As a result, direct contact between carbon steel and the environment was alleviated [37]. Therefore, the corrosion inhibition was found to be effective within the temperature range studied in this study. The temperature increased from 25°C to 60°C only increased the rate of corrosion product formation, which could potentially act as a protective film on the carbon steel surface, but not to the extent that the carbon steel deteriorated. Overall, the corrosion results of carbon steels in MeOH-containing solutions showed the Arrhenius behaviour, in which the corrosion rate increased with increasing temperature, but not as significantly as the other three types of MEA solution.

3.3 Role of CO_2 loading on the corrosion behaviour

To investigate the effect of CO_2 presence in the solution on carbon steels, the process conditions with or without CO_2 loading were carried out at a temperature of 25°C. Figure 4 illustrates the corrosion rate of carbon steel in the presence and absence of CO_2 loading in solution at 25°C after a 28 day immersion period. It is observed that the corrosion rate of carbon steel coupons immersed in MEA + H_2O was 9.42 MPY with CO_2 loading in the solution. This indicates that the presence of CO_2 posed a severe impact on the carbon steel coupon in MEA + H_2O solution. However, the corrosion rate of carbon steel coupons immersed in MEA + NMP and MEA + NMP + H_2O hybrid solutions with CO_2 loading were 2.77 MPY and 2.43 MPY, respectively, while the corrosion rate of carbon steel in MEA-MeOH hybrid solutions appeared to be negligible.

To further decipher the corrosion behaviour, Raman spectroscopy analysis was conducted to analyse the changes in the MEA solutions with the presence and absence of CO_2 loading. The Raman spectrums of the MEA solutions with the presence and absence of CO_2 loading are shown in Figure 5-9. Based on the Raman spectrum, CO_2, carbamate, and carbonate were observed to be present in all the MEA solutions after CO_2 loading. Additionally, bicarbonate ions were also found in MEA + H_2O after the loading of CO_2. The additional components of CO_2, carbamate, carbonate, and bicarbonate detected in the MEA aqueous solution after loaded with CO_2 could lead to the possible development of additional reaction associated with the protonation of MEA-H_2O-CO_2.

These components could accelerate the corrosion of carbon steel in amine solution [38]. Nevertheless, the MeOH-containing MEA solutions were found to have the least corrosion degradation effect on carbon steels. These findings were intriguing, especially given that no significant difference in corrosion rate was observed when carbon steels were immersed in MEA + MeOH and MEA + MeOH + H_2O solutions. It is deduced that methanol could be decomposed into certain intermediates and promoted methoxylation. This could promote formation of a stable $FeOCH_3$ layer to protect the carbon steel surface from CO_2 and MEA [39].

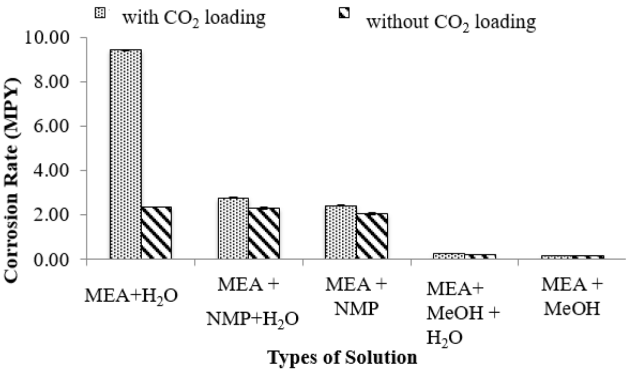

Figure 4. The corrosion rate of the carbon steel in the presence and absence of CO_2 loading.

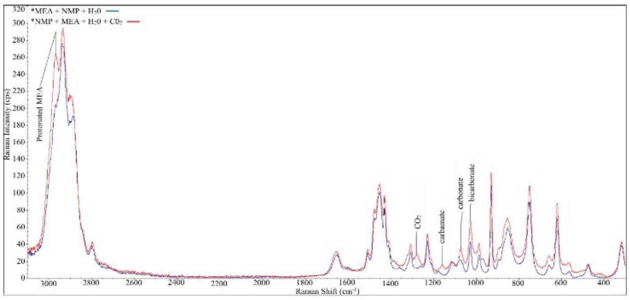

Figure 5. Raman spectrum analysis of MEA + H_2O before and after CO_2 loading.

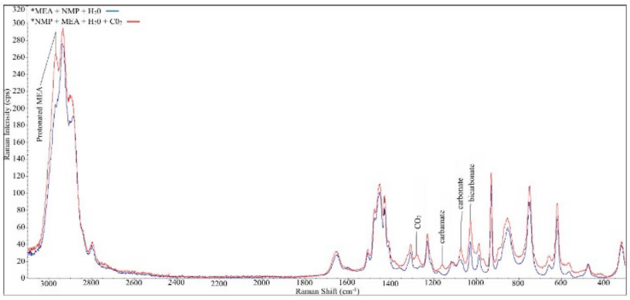

Figure 6. Raman spectrum analysis of MEA + NMP + H_2O before and after CO_2 loading.

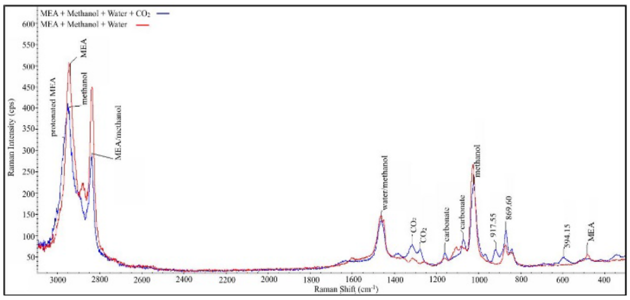

Figure 7. Raman spectrum analysis of MEA + MeOH + H_2O before and after CO_2 loading.

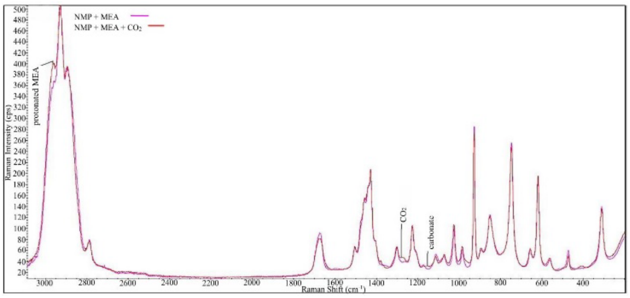

Figure 8. Raman spectrum analysis of MEA + NMP before and after CO_2 loading.

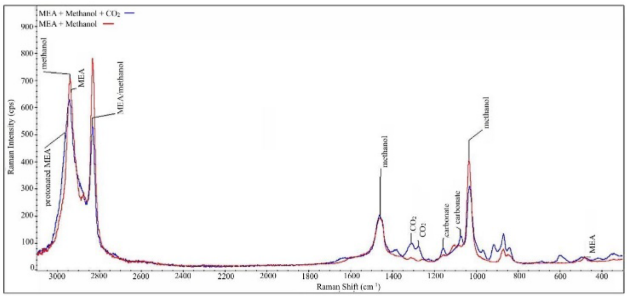

Figure 9. Raman spectrum analysis of MEA + MeOH before and after CO_2 loading.

4. Conclusions

This study elucidated that carbon steel coupons immersed in MEA + MeOH had the lowest corrosion rate among all the solutions across all the interval periods investigated in this study. It was found that raising the temperature expedited the corrosion of the carbon steel coupon immersed in all solutions due to the increased kinetic energy in the molecules or atom of carbon steel coupon and solution. The immersion of carbon steel coupon in MEA + H_2O was observed to result in the highest corrosion potential, which might be due to the initiation of film formation, such as iron oxide deposit on the surface. For the carbon steel immersed in MEA + MeOH, a passivating layer may have been created on the surface of the carbon coupon to protect it from corrosion. In terms of CO_2 loading, carbon steel coupons in MEA aqueous solutions corroded at a significantly higher rate than solutions without CO_2 loading. In short, organic solvent was found to have a positive impact on the corrosion resistance of carbon steel in MEA solution in the presence of CO_2. For future studies, it is recommended to conduct Electrochemical Impedance Spectroscopy (EIS) analysis to reveal the underlying mechanisms driving the different corrosion behaviours observed when carbon steel is immersed in the hybrid solvents.

Declaration of competing interest

The authors declare that they have no financial or personal conflicts of interest that could have affected the work reported in this study.

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