

The effect of CO₂-loaded amine solvents on the corrosion of a carbon steel stripper



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ABSTRACT

The corrosive behaviour of loaded amine solvents was evaluated under stripper operating conditions, for post-combustion carbon capture, to determine the feasibility of using carbon steel in plant construction. In addition to monoethanolamine, three alternative amine solvents: methyldiethanolamine (MDEA), 2-amino-2-methyl-1-propanol (AMP), 1-(2-aminoethyl)piperazine (AEPZ), and the common additive K₂CO₃ were studied when in contact with carbon steel (C1018) over a 28-day period. Corrosive behaviour was evaluated using carbon steel coupons: gravimetric method for weight change, surface imaging (SEM) and analytical techniques (EDX and XRD), and Fe ion concentration in solution (ICP-OES). The results demonstrated that MDEA and AMP as well as K₂CO₃ develop a significant siderite (FeCO₃) layer on the carbon steel surface. The presence of this layer is attributed to the preferred reaction pathway with CO₂ for tertiary and sterically hindered amines. The FeCO₃ layer formed in the case of MDEA provides superior protection from continued corrosion of the carbon steel. By contrast, MEA and AEPZ show significant corrosion to the carbon steel surface. In conclusion, MDEA, AMP, and K₂CO₃ can preferentially produce sufficient surface FeCO₃ layers to reduce corrosion levels in carbon steels for use under stripper conditions in post-combustion carbon capture plants.

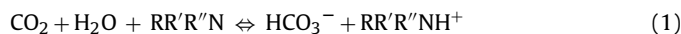
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1. Introduction

A rapid advancement in post-combustion carbon capture (PCCC) technology will result in its inevitable world-wide industrial deployment, if the remaining technical limitations can be overcome. One of the principal concerns is the corrosive nature of aqueous amine solvents, used for PCCC, in the presence of flue gases containing carbon dioxide (CO₂) and a range of impurities. This challenge is a well-known problem in the natural gas sweetening industry (DuPart et al., 1993a,b). To combat this problem all PCCC pilot plants currently in operation use stainless steel to minimize the problem of corrosion, as do industrial-scale natural gas sweetening plants. For carbon capture, this approach is an untenable solution for large-scale PCCC implementation, where the economy of carbon steel utilization is essential.

The first generation of amine carbon capture technology has relied on monoethanolamine (MEA) as the solvent. However, industrialists seeking to improve process efficiency and reduce costs are focused on the use of other amine solvents (Liang et al., 2015). These alkanolamine solvents are selected because of their

ability to preferentially and reversibly react with CO₂. In the presence of CO₂ and water all alkanolamine solvents may undergo a kinetically slow reaction which is limited by the dissociation of carbonic acid to form bicarbonate (Reaction (1)).



However, a kinetically faster reaction is possible for primary and secondary amines due to the presence of at least one hydrogen bonded to the nitrogen atom. In this reaction mechanisms an amine carbamate is formed (Reaction (2)) and followed by a hydrolysis reaction (Reaction (3)).



As a result of the faster kinetic reaction of these amines with CO₂, primary and secondary amines are often preferred industrially. However, the development of multi-component amine mixtures has demonstrated the benefits of using tertiary amines in parallel (Idem et al., 2006).

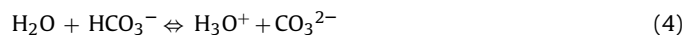
The reaction mechanisms of the amines with CO₂ (Reactions (1) and (2)) are crucial given that the amine solvents themselves are generally relatively inert, with respect to corrosion. However, there is little understanding of the mechanisms by which the

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corrosion itself occurs. Some studies have suggested that the rate of carbamate formation is directly linked to the corrosivity of amine solutions (DuPart et al., 1993a,b; Teevens, 1990). Importantly, the type of amine used has been identified as highly influential in determining the corrosion potential of such solutions (Kittel and Gonzalez, 2014), though only limited research has been reported.

Iron carbonate (FeCO_3) is one of the primary product layers observed during the amine corrosion process. The dissociation of a bicarbonate ion (Reaction (4)) in an aqueous medium with available Fe ions (e.g. Fe^{2+}) (Reaction (5)) results in a straightforward production of FeCO_3 (Reaction (6)).



The formation of Fe carbonate as a protective layer has been investigated in the past (Han et al., 2009, 2011; Tanupabrungsun et al., 2013). However, limited information is known about this process and other corrosion processes with amine solvents, especially their behaviour with carbon steel. Overall, most research reported has demonstrated that increases in temperature and CO_2 loading increase the rate of corrosion (Kittel and Gonzalez, 2014). A few additional studies have focused on the role of impurities, which were generally viewed to have a similarly negative impact. Electrochemical studies of MEA have generally used potentiodynamic polarization with Tafel extrapolations to estimate the corrosion rate, and some investigation of electrochemical impedance (Soosaiprakasham and Veawab, 2008; Wattanaphan et al., 2013; Sun et al., 2011). While such an approach is useful, a traditional and industrially relevant demonstration of corrosion is essential in understanding the full impact likely to be observed in a real industrial scenario (Xiang et al., 2014).

The sterically-hindered primary amine 2-amino-2-methyl-1-propanol (AMP) demonstrates a range of positive features in comparison to MEA. Two reports on the corrosive nature of AMP, using electrochemistry, highlight that in an aqueous solution the corrosion rate is an order of magnitude lower under than MEA under lean conditions of CO_2 loading and comparable under rich CO_2 loading conditions (Veawab et al., 1999a,b). Again using electrochemistry, a second study compared various amines to indicate their relative corrosivity (Gunasekaran et al., 2013), at 80°C with CO_2 saturation for a solution of 5 kmol m^{-3} (with the exception of AMP at 4 kmol m^{-3}). The corrosion rate in decreasing order is suggested: $\text{MEA} > \text{AMP} > \text{diethanolamine (DEA)} > \text{piperazine (PZ)} > \text{methyldiethanolamine (MDEA)}$.

Aside from MEA, MDEA is the most widely studied amine with early research, focused on natural gas sweetening applications, indicating the reduced corrosion behaviours observed. (DuPart et al., 1993a,b) An early investigation highlights that MDEA may form a protective scale under some circumstances. (Tomoe et al., 1996) The apparent reduced corrosive effect of MDEA has been probed, almost exclusively, using electrochemical methods (Khorrami et al., 2008; Guo and Tomoe, 1999). One report (Xiang et al., 2015) includes traditional approaches to observing corrosion, highlighting the critical nature of using this approach in addition to electrochemical reports.

The work reported herein focuses on assessing the corrosion behaviour which would occur under process conditions found in a CO_2 stripper column fabricated from carbon steel. This study is essential to fully understanding previous electrochemical reports and industrially relevant observations. In particular, with a very low oxygen concentration, thermal degradation of the solution at elevated temperatures will occur. This industrially relevant research evaluates the feasibility of using carbon steel infrastructure in the stripper for aqueous amine solutions. The corrosive effect on the

carbon steel at elevated temperatures was monitored for 28 days to provide a more comprehensive view of the corrosion chemistry which occurs. This study includes a direct comparison of an aqueous MEA solution with three other popular alternative amine solvents (AMP, MDEA, and 1-(2-aminoethyl)piperazine (AEPZ)), as well as the common additive potassium carbonate (K_2CO_3).

2. Materials and methods

The chemicals MEA (Sigma $\geq 98\%$), AMP (Sigma $\geq 90\%$), AEPZ (Sigma $\geq 99\%$), MDEA (Sigma $\geq 99\%$), and K_2CO_3 (Sigma) were used as received. Solutions (5 M) for all amine solvents of interest were made with DI water and the solution containing K_2CO_3 to 0.5 M using DI water.

Carbon steel coupons (Type C1018: Fe: 98.85%, C: 0.17%, Mn: 0.80%, Cr: 0.04%, Al: 0.04%, $\leq 0.02\%$: P, S, Si, Cu, Ni, Sn, N, V, B, Ti, Co) (Alabama Specialty Products) with the dimensions $73\text{ mm} \times 22\text{ mm} \times 2.2\text{ mm}$, two holes for attachment (radius of 6.75 mm), and a machine finish were used in custom made glass reactors.

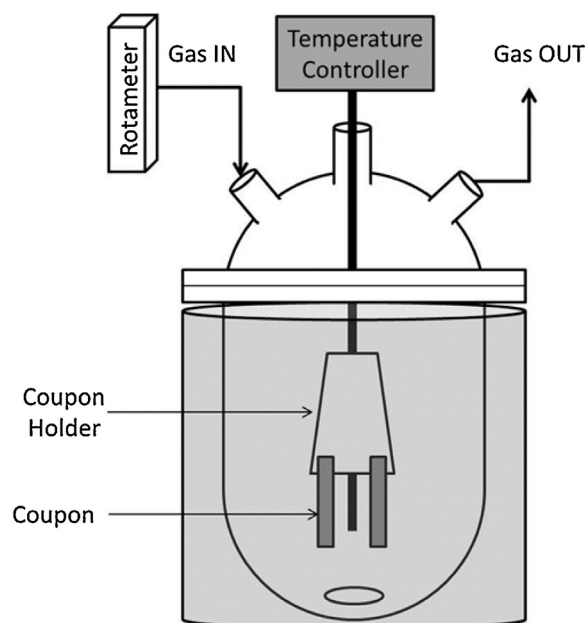


Fig. 1. Corrosion vessel.

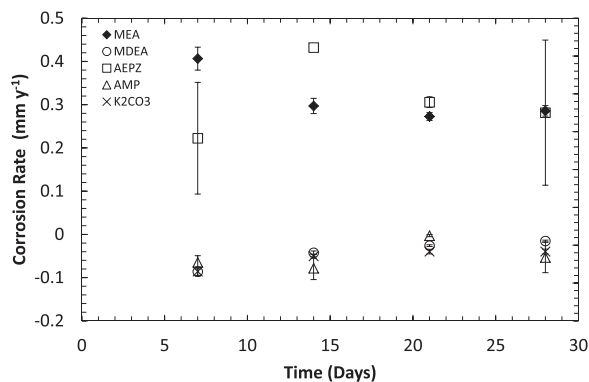


Fig. 2. Corrosion rates based on mass loss following the weekly removal of mild steel coupons from 5 M solutions of MEA, MDEA, AEPZ, and AMP and a 0.5 M solution of K_2CO_3 .

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