Impact of pH level and magnesium addition on corrosion of re-mineralized seawater reverse osmosis membrane (SWRO) product water on pipeline materials

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HIGHLIGHTS

- pH level did not change the morphology and microstructure of the coupon surfaces.
- Higher Mg/Ca ratio could encourage the precipitate morphology transformation.
- Magnesium reduced CaCO3 precipitant amount and maintained a high alkalinity and hardness level.
- Impact of pH level within the range of pH 7 to pH 8.4 on corrosion was minor and insignificant.
- Intense protection of cement-lined coupon materials was observed in CaMg-7 water.

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ABSTRACT

Remineralization of desalinated seawater has been widely applied to reduce pipeline corrosion problems and to achieve drinking water standards. This study investigated the effects of pH and magnesium addition on corrosion of pipeline materials in stagnant remineralized desalinated seawater under tropical conditions. Three pipeline materials, ductile iron, cement-lined ductile iron and cast iron were examined. The pH of the remineralized reverse osmosis (RO) desalinated seawater ranged from 7.0 to 8.4. Weight loss method was used. The pH variations showed little effect on the corrosion product composition and final water quality for the three pipeline materials. The overall findings suggested that the impact of pH level from pH 7.0 to pH 8.4 on corrosion rates of iron coupon materials was unpronounced. Addition of magnesium changed the morphology and microstructure of CaCO3 precipitant, reduced the amount of the precipitant and maintained a high alkalinity and hardness concentration in the re-mineralized RO permeate. For cement-lined ductile iron coupons, an intense protection was found in water with magnesium addition under pH 7.0 condition.

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1. Introduction

1.1. Background

To meet the growing water demand, desalination has become one of the most important technologies for increasing fresh water supply worldwide in recent years. Although RO desalination has been developed for a few decades, severe corrosion of desalination water distribution pipes is still a critical concern. To counteract corrosion and achieve the drinking water standards, post-treatment of the desalinated seawater is required before distribution [1–3]. Several post-treatment techniques (re-mineralization process), such as direct dosage of chemicals, blending of seawater or brackish water, calcite (CaCO3) dissolution and dolomite (CaMg(CO3)2) dissolution [4–8], were established and compared in previous studies [4–8].

A report from World Health Organization (WHO) [9] addressed the importance of nutritionally desirable component addition as well as corrosion control in desalination water distribution system. Some general water quality parameters, including calcium ion concentration, alkalinity, calcite precipitation index and pH level, were implemented to monitor the water quality of soft water [4,10]. To control both chemical and biological stability of desalinated seawater, and ensure a safe...
drinking water quality, a proposed drinking water standard of post-treated desalinated seawater was released by Israel’s Ministry of Health (Table 1) [11]. Calcium concentration ranging from 80 mg/L to 120 mg/L as CaCO₃ was recommended for remineralization of desalinated seawater. In this study, remineralization of desalinated water followed the established Israeli standard. In addition, magnesium was also recommended for health consideration.

1.2. Literature review

Previous studies have shown the effect of different water quality parameters such as alkalinity, buffer capacity and calcium content on corrosion control [4,5,12]. Our earlier study also demonstrated the positive effect of remineralization of desalinated seawater for corrosion control of pipeline, which was mainly manufactured with the materials of ductile iron, cast iron and cement-lined ductile iron, under stagnant condition [13].

The effect of pH variations on iron pipeline corrosion had been discussed with different conclusions being drawn [10,14–17]. Sarin et al. [14] showed that higher pH led to the formation of an impervious iron membrane on the scaling of iron surfaces, which could reduce iron release and lower pH resulted in the formation of a fibrous porous scale structure, resulting in large amounts of iron released. Lasheen et al. [15] also reported that when pH was less than 6, iron released from pipe increased as pH decreased. Conversely, under high pH condition (>pH 8), increasing water pH level resulted in reduced corrosion rate and a reduction in iron released. In contrast, Birnback et al. [10] found that within the typical pH range of 7.5–8.4, a higher pH resulted in a lower buffering capacity, which was not preferred for corrosion control. Others reported increasing corrosion rates with increasing pH in the range of 6.9 to 8.6 as well [16]. On the other hand, Imran et al. [17] found that high pH levels, in the range of 7.8 to 8.4, had no effect on the rate of corrosion or iron release as long as the water was supersaturated with respect to CaCO₃ precipitation (i.e., LSI > 0).

Effect of pH on deterioration of cement was illustrated in a range of pH 4 to 12 [18–21]. The total porosity and the depth of decalcification were found to increase with the reduction of pH level in comparison with pH 4.5 and pH 8.5 conditions [19,20]. Expansion, which resulted in a reduction in strength of concrete, decreased when pH level of the sulfate solution was reduced from 12 to 7 and increased from 7 to 3 [21]. Expansion rate was higher at higher pH level in comparison between pH 7.0, pH 9.0 and pH 11.0 conditions. The expansion rate was slower under lower pH condition than higher pH condition in pH range of pH 7.0 to 11.0 [18]. However, the comparison of pH impact on cement materials in the past was mostly made between the acidic, neutral or alkaline conditions. Less discussion of pH impact on cement deterioration was brought out within the drinking water pH range 7.0 to 8.4.

Due to the diverse conclusions drawn by previous reports, this study aimed to examine the effects of pH level on the corrosion of pipes with remineralized SWRO water, especially in tropical countries like Singapore, which less related studies were focused in. In continuous of our previous study [13], we further conducted experiments under stagnant condition to simulate the stagnant condition in intermittent water supply or possible suspension of adsorption processes. According to the standard established by Israel’s Ministry of Health [4,10], pH level of drinking water was recommended to be below 8.5. This study therefore aimed to examine the impact of pH level in the range of pH 7.0 to pH 8.4, which was relevant to drinking water standards.

2. Methodology

2.1. Methods

RO desalinated seawater was collected from the variable salinity plant (VSP) at Tampines, Singapore. The water was transported to the laboratory and stored in a refrigerator at 4 °C before post-treatment. Water quality of the desalinated seawater was analyzed and established in Table 2. Direct dosage of chemicals was chosen as the primary remineralization technique in this study. To avoid addition of extra ionic contents, Ca(OH)₂ and MgCO₃ in two different ratios (100/0 and 60/40) were dosed and mixed homogeneously with the desalinated seawater to achieve 100 mg/L as CaCO₃ hardness and 100 mg/L as CaCO₃ alkalinity. The remineralized water was subsequently adjusted to pH 7.0, pH 8.0 and pH 8.4 by purging carbon dioxide (CO₂) into solution. With un-remineralized desalinated seawater as control, seven types of water were placed in 1 L glass reactors for study. These were conducted in duplicates. Reactors were covered with caps and sealed completely with parafilm. Each reactor was equipped with a PTFE Teflon tube in which one end was immersed in solution while a 0.45 μm pore size syringe filter was attached to the other end which was exposed to air. Remineralized water labels and treatments were elaborated in Table 3.

Table 1

<table>
<thead>
<tr>
<th>Water parameter</th>
<th>Value</th>
<th>Water parameter</th>
<th>Value</th>
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</thead>
<tbody>
<tr>
<td>Calcium carbonate precipitation potential (CCPP)</td>
<td>3–10 mg/L as CaCO₃</td>
<td>Total alkalinity</td>
<td>&gt;80 mg/L as CaCO₃</td>
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<td>pH</td>
<td>&lt;8.5</td>
<td>Calcium</td>
<td>mg/L</td>
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<tr>
<td>Turbidity</td>
<td>&lt;0.5 NTU</td>
<td>Magnesium</td>
<td>mg/L</td>
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Table 2

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<th>Unit</th>
<th>Desalinated seawater</th>
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<tr>
<td>Total alkalinity</td>
<td>mg/L as CaCO₃</td>
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</tr>
<tr>
<td>Total hardness</td>
<td>mg/L as CaCO₃</td>
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<tr>
<td>Calcium</td>
<td>mg/L</td>
<td>0</td>
</tr>
<tr>
<td>Magnesium</td>
<td>mg/L</td>
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</tr>
<tr>
<td>Sodium</td>
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<td>Chloride</td>
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<td>Sulfates</td>
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<td>Phosphate</td>
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<td>mg/L</td>
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<td>Fluoride</td>
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<tr>
<td>Bromide</td>
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<td>pH</td>
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<tr>
<td>Redox potential</td>
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<tr>
<td>Conductivity</td>
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<tr>
<td>Total dissolved solids</td>
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<tr>
<td>Dissolved oxygen</td>
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<tr>
<td>Total organic carbon</td>
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Characteristics of desalinated seawater.
coupons in various remineralized water, the experiment was carried out for 20 days to achieve sufficient weight loss according to ASTM standard [23].

Water quality parameters such as turbidity, alkalinity, hardness, pH, total dissolved solids (TDS), and cation concentrations were measured before and after corrosion experiments. Measuring methods and instruments are listed in Table 4.

Table 3
Alkalinity and hardness content of desalinated seawater and re-mineralized water used in experiments.

<table>
<thead>
<tr>
<th>Label</th>
<th>Treatment</th>
<th>pH</th>
<th>Alkalinity</th>
<th>Hardness</th>
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<th>Ca²⁺</th>
<th>Mg²⁺</th>
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<td></td>
<td></td>
<td></td>
<td>mg/L as CaCO₃</td>
<td>mg/L</td>
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<td>0</td>
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<td>Ca100-7</td>
<td>Ca(OH)₂ + CO₂</td>
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<td>100</td>
<td>100</td>
<td>100</td>
<td>40</td>
<td>0</td>
</tr>
<tr>
<td>Ca100-8</td>
<td>Ca(OH)₂ + CO₂</td>
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<td>100</td>
<td>100</td>
<td>100</td>
<td>40</td>
<td>0</td>
</tr>
<tr>
<td>Ca100-8.4</td>
<td>Ca(OH)₂ + CO₂</td>
<td>8.4</td>
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<td>100</td>
<td>100</td>
<td>40</td>
<td>0</td>
</tr>
<tr>
<td>CaMg-7</td>
<td>Ca(OH)₂ + MgCO₃ + CO₂</td>
<td>7.0</td>
<td>100</td>
<td>100</td>
<td>100</td>
<td>60</td>
<td>24</td>
</tr>
<tr>
<td>CaMg-8</td>
<td>Ca(OH)₂ + MgCO₃ + CO₂</td>
<td>8.0</td>
<td>100</td>
<td>100</td>
<td>100</td>
<td>60</td>
<td>24</td>
</tr>
<tr>
<td>CaMg-8.4</td>
<td>Ca(OH)₂ + MgCO₃ + CO₂</td>
<td>8.4</td>
<td>100</td>
<td>100</td>
<td>100</td>
<td>60</td>
<td>24</td>
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Table 4
Methodology and equipment used for water quality analyses.

<table>
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<tr>
<th>Water parameter</th>
<th>Method</th>
<th>Instrument</th>
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</thead>
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<tr>
<td>Turbidity</td>
<td>Hach method 8195</td>
<td>Hach 2100N turbidimeter</td>
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<tr>
<td>Apparent color</td>
<td>Platinum–cobalt standard method 8025</td>
<td>Hach DR5000 UV–vis spectrophotometer</td>
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<tr>
<td>pH</td>
<td></td>
<td>Orion pH meter</td>
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<tr>
<td>Total alkalinity</td>
<td>Titration</td>
<td>USEPA burette titration method</td>
</tr>
<tr>
<td>Total hardness</td>
<td>Titration</td>
<td>Burette titration method 8338</td>
</tr>
<tr>
<td>Total dissolved solids (TDS)</td>
<td>Titration</td>
<td>Thermo Scientific Orion 5 Star Multiparameter Meter Kit</td>
</tr>
<tr>
<td>Cation concentration</td>
<td>ICP-OES, Optima 7000 DV, Perkin Elmer</td>
<td></td>
</tr>
</tbody>
</table>

2.2. **Scanning electron microscope (SEM)/energy dispersive spectrometer (EDS)**

After 20-day immersion, coupons were carefully taken out from reactors, dried in an oven at 60 °C and kept in vacuum desiccators. Microstructures of corrosion products on coupon surfaces were observed using a scanning electron microscope (SEM Stereoscan 420, Leica, Fig. 1. Reactors with iron coupons. (A) Ductile iron reactors in SWRO water; (B) ductile iron reactors in Ca100-8 water; (C) ductile iron coupon in SWRO water; (D) ductile iron coupon in Ca100-8 water.

Fig. 2. (A) Ductile iron coupons after experiment in CaMg-8 water; (B) cast iron coupons after experiment in CaMg-8 water; (C) cement-lined coupons after experiment in CaMg-8 water; (D) cement-lined coupons after experiment in Ca100-8 water.
2.3. X-ray diffraction (XRD)

Corrosion products on ductile iron and cast iron coupon surfaces were gently scrapped off by small spades, without damaging the metal surfaces, and ground into powder form for composition analysis [22]. Additionally, water in the reactors was filtered by using 0.45 μm pore size filter paper and corrosion products that had settled at the bottom of the reactor were collected on the filter paper. The dried corrosion products collected from the filter paper were ground into powder form for analysis. Composition of both types of corrosion products was analyzed using the D5005 Bruker powder X-ray diffractometer with copper (Cu) radiation operated at a voltage of 40 kV and current of 40 mA. Full X-ray diffraction patterns were recorded for scan angles (2θ) from 10° to 80° with a step size of 0.02° to identify corrosion products. Individual crystalline compounds were identified from the XRD patterns by using powder diffraction file (PDF) developed by the International Centre for Diffraction Data (ICDD-PDF) database.

3. Result and discussion

3.1. Corrosion phenomenon

Photographs of coupons and reactors were taken for documentation during the course of the experiment. Iron coupons were observed to corrode rapidly after being immersed in the water, with a layer of red deposits forming on the surfaces of the coupons and gradually settling at the bottoms of the reactors. Selected photographs of the reactors, taken on Day 3, are shown in Fig. 1. Suspended corrosion products resulted in the orange coloration of the water in reactors with iron coupons. Orange-colored corrosion products were found covering the iron coupon surfaces. In comparison to reactors with ductile iron coupons in the desalinated seawater and Ca100-8 water, a lighter shade of orange could be observed in the Ca100-8 water, which was due to the lower amounts of corrosion products formed with remineralized water. No obvious difference was found in reactors and coupons at different pH conditions.

Unlike the iron coupons, no orange-colored corrosion products were observed in reactors with cement-lined coupons, whereas white precipitants were observed on the reactor walls and at the bottoms of all reactors filled with remineralized water at all pH levels tested. When the
white precipitants were dissolved in hydrogen chloride acid, numerous bubbles were formed on the precipitants, which implied the possible existence of CO$_3^{2-}$ or HCO$_3^-$ in the precipitants. Analysis by ICP-OES revealed that the main component of metal cation in the dissolved precipitants was calcium. The results verified that the dominant compound which constituted the white precipitants was calcium carbonate. This white precipitant CaCO$_3$ was not found in the reactors with desalinated seawater without remineralization.

Coupons were dried in an oven at 60 °C and their surface properties were analyzed. Ductile iron and cast iron coupons in CaMg-8 water are shown in Fig. 2(A) & (B) where uniform orange-colored corrosion products coated large proportions of the coupon surface, while white precipitants could be observed at the edges of the coupons. In contrast, no corrosion products were noticed in images of cement-lined coupons in both CaMg and Ca100 water (Fig. 2(C) & (D)). Instead, few white dotted crystalline precipitants were found attached onto the cement-lined coupon surfaces.

### 3.2. Corrosion product analysis

#### 3.2.1. SEM results

The surfaces of different materials were examined under SEM. Uniform flower-like aggregated crystalline microstructures were observed on all iron coupons (Fig. 3(A) & (B)). Similar microstructures of iron oxides were also noted by Swietilik et al. [24].

Formation of rhombic-like (calcite) or needle-like (aragonite) microstructures was clearly observed on ductile iron coupons in both Ca100 and CaMg remineralized water (Fig. 3(C) & (D)). The role of magnesium in modulating the morphology and structure of calcium carbonate was illustrated in various papers [25–27]. With the ratio of [Mg$^{2+}$]/[Ca$^{2+}$] higher than 0.5 and at a temperature of 25 °C, the development of aragonite, characterized by needle-like microstructure, was dominant over calcite. On the other hand, calcite, which was characterized by rhombic-like microstructure, was formed in the environment of low [Mg$^{2+}$]/[Ca$^{2+}$] ratio. This concurred with the observation that in Ca100 water (Fig. 3(C)) with [Mg$^{2+}$]/[Ca$^{2+}$] ratio of around 0.02, both calcite and aragonite crystalline microstructures were found. However, only aragonite crystalline microstructures were present in high Mg$^{2+}$ concentration CaMg water with [Mg$^{2+}$]/[Ca$^{2+}$] ratio of around 0.45 in the original stock water and around 0.9 at the end of experiment (Fig. 3(D)).

Photomicrographs of cement-lined coupon surfaces under SEM examination are presented in Fig. 3(E) & (F), in both Ca100 and CaMg water. In contrast to the iron coupons, no iron oxides were found on the cement-lined coupon surfaces despite the fact that dotted white crystals were observed in the photographs taken (Fig. 2(C) & (D)). In addition, there was no significant difference in all cement-lined surfaces after immersion in the different types of water.

#### 3.2.2. XRD results

As presented in Section 3.2.1, SEM provided only information on microstructure and morphology of the corrosion products and precipitants. In addition, composition of exact compounds of the corrosion products and precipitants was identified by X-ray diffraction (XRD) [28]. Typical compounds that are regularly found in iron corrosion scales included goethite (α-FeOOH), lepidocrocite (γ-FeOOH), magnetite (Fe$_3$O$_4$), maghemite (α-Fe$_2$O$_3$), ferric hydroxide (Fe(OH)$_3$), ferrous oxhide (FeO), siderite (FeCO$_3$), ferrous hydroxide (Fe(OH)$_2$), ferrhydrite (5Fe$_2$O$_3$·9H$_2$O), and calcium carbonate (CaCO$_3$) [28–32].

Detailed XRD analysis (Table 5) suggested that the corrosion products were a mixture of different compounds. The primary components of the corrosion products were lepidocrocite (γ-FeOOH), lepidocrocite (Fe$_2$O$_3$·H$_2$O), goethite (α-FeOOH), iron hydroxide (Fe(OH)$_3$), goethite (Fe$_2$O$_3$·H$_2$O·xH$_2$O), iron oxide (Fe$_2$O$_3$) and calcium carbonate (CaCO$_3$).

The difference in corrosion product compositions between ductile and cast iron was observed when both coupons were immersed in CaMg water, while the compound compositions between two iron materials were similar in other types of water. No lepidocrocite (γ-FeOOH) was formed on cast iron coupons in water with magnesium contents added. A comparison of corrosion products collected on coupon surfaces and at the bottom of all reactors revealed that CaCO$_3$ was observed only on the coupon surfaces rather than in the sediments. The precipitants which formed on the coupon surfaces adhered strongly onto the coupon surfaces and were not readily dislodged.

XRD results showed that the addition of Ca$^{2+}$ into desalinated seawater brought about the formation of CaCO$_3$ (Table 5). This could

### Table 5

<table>
<thead>
<tr>
<th>Ductile Iron</th>
<th>SWRO</th>
<th>Ca100-7</th>
<th>Ca100-8</th>
<th>Ca100-8.4</th>
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− FeOOH Fe$_2$O$_3$·H$_2$O CaCO$_3$ Fe$_3$O$_4$ Fe(OH)$_3$
explain the presence of white crystalline precipitants observed on the coupon surfaces and calcite- or aragonite-like crystalline microstructures seen during SEM examination. XRD spectrums of both ductile iron and cast iron corrosion products in both types of remineralized water are presented in Fig. 4. With addition of magnesium content, the components of iron oxide mixtures in corrosion products remained unchanged. However, calcite vanished from the main component of calcium carbonate precipitant. Instead, aragonite was the only component of white crystalline precipitant in CaMg water. This result also complied with the SEM results that higher Mg/Ca ratio could encourage the transformation of precipitant morphology.

In Fig. 5, the comparison of XRD patterns of corrosion products on ductile iron coupon surfaces in Ca100 water with pH level from 7.0 to 8.4 is shown. The alteration of pH level did not change the composition of corrosion products and precipitant formation during the experiment as well. Analysis of corrosion products suggested that the impact of pH level on iron materials and their resultant corrosion products was insignificant. Complied with SEM observation, calcium carbonate precipitants closely attached on the coupon surfaces in remineralized water, and magnesium addition encouraged the precipitant morphology transformation as well.

3.3. Water quality

3.3.1. Iron coupon

The effect of magnesium addition on water quality is presented in Fig. 6. After 20-day immersion, alkalinity and hardness concentration of the remineralized water declined significantly compared with stock water alkalinity and hardness concentration for reactors with both ductile iron and cast iron coupons. As shown in Fig. 6(A), alkalinity concentration reduced from 102.4 to 46 mg/L as CaCO3 (about 53% reductions) in Ca100 water. With magnesium addition, a lower reduction in alkalinity concentration from 106.8 to 80 mg/L as CaCO3 (about 25% reduction), was observed. This resulted in a higher alkalinity concentration in CaMg water after experiment. With respect to hardness concentrations at the end of the experiment, similar effect was also noted in water supplemented with magnesium (Fig. 6(B)). Reduction of hardness concentration in Ca100 water was around 53%, while in CaMg water, the hardness reduction was only 31%. The decline of alkalinity and hardness concentration could be explained by the precipitation of calcium carbonate as discussed previously.

Reduction of magnesium concentration was around 5% after experiment for both ductile and cast iron (Fig. 6(D)). The partial alkalinity and hardness attributed to magnesium content did not vary too much in CaMg water, total alkalinity and hardness were comparably higher in CaMg water than in Ca100. The role of magnesium content in maintaining a comparably high alkalinity and hardness attributed to the minor decline of magnesium ion concentration. This finding was in line with the SEM and XRD results, that no magnesium precipitant was found in water or on coupon surfaces. Magnesium content remained in a soluble ionized form in neutral pH level range, while pH varied from pH 7.5 to pH 8.0 after immersion of iron coupons in our study.

In addition, calcium ion concentration in Ca100 water reduced by around 65%, which was higher than the 54% decline observed in CaMg water (Fig. 6(C)). By studying calcium balance in the previous studies [13], almost all the reduction of calcium ion concentration translated to the formation of calcium carbonate precipitant. With magnesium addition in CaMg water, the lower reduction of calcium content could result in lower amount of calcium carbonate formation. Likewise, the presence of magnesium did not increase the amount of calcium carbonate precipitant. Several studies [26,33–36] have shown that the presence of magnesium ion would retard the precipitation process of calcium carbonate. The retardation might be due to several reasons, (i) MgCO3 aquo-complex formation, which reduces free CO32− ions, (ii) covering active surface sites of CaCO3 nuclei and crystals with Mg2+, and (iii) incorporation of Mg2+ in CaCO3, which induces changes in calcite solubility [26]. This inhibition of calcium carbonate precipitation may be related to the high hydration energy of Mg2+ and to the calcite structure which allows incorporation of Mg2+, and at the same time
time, the solubility constant of calcite increased with increasing Mg$^{2+}$ [33,36,37]. In conclusion, the presence of magnesium did not increase the amount of calcium carbonate precipitant but rather, altered the precipitant morphology and structure.

The effects of different pH on water quality parameters (alkalinity, hardness, calcium ion concentration) are illustrated in Fig. 7. The extents of reduction in both alkalinity and hardness concentrations were similar under different pH conditions, which were 52% to 54%, respectively for ductile iron coupons. A similar proportional decline of calcium concentration with pH level variation could also be observed in Fig. 7(C). The calcium concentration reduced from a range of 35.9–37.5 mg/L to a range of 12.6–15.2 mg/L (~ranging from 60 to 65% reduction) in reactors with ductile iron coupons. Regardless the initial pH levels of 7, 8 and 8.4, pH levels attained at the end of all experiments were between pH 7.8 and pH 7.9 (Fig. 7(D)).

As the stagnant experiments were conducted in reactors that were exposed to the environment, pH level was not controlled during the experiment studies, which could be observed in Fig. 7(D). Fig. 8 presents the pH variations during the experiment under each pH level condition. Initial pH levels of pH 7.0 and pH 8.4 conditions gradually approached pH 8.0 at the end of the experiment. Meanwhile, the experiment conducted at the initial pH value of pH 8.0 remained at a steady level throughout the entire experimental duration. It was concluded that regardless of the inconsistency in stock water quality, water within each iron coupon reactor tends to achieve similar final water quality after 20-day immersion. The pH level had little effect on the water quality in reactors with iron coupons.

### 3.3.2. Cement-lined coupons

In reactors with cement-lined coupons, alkalinity, hardness, calcium concentration and pH level increased significantly in all types of remineralized water. The comparison of water quality change in Ca100-8 and CaMg-8 was established in Fig. 8. The increment of alkalinity and hardness was around 65% to 70% in both types of remineralized water after experiment (Fig. 9(A) & (B)). Shown in Fig. 9(C), calcium concentration increased from 35.9 to 72.7 mg/L in Ca100 water and 23.2 to 70.9 mg/L in Mg supplemented water. The increased calcium concentration was the consequence of both leaching and carbonation of cement-lining.

It is widely accepted that leaching of cementitious materials is one of the most significant elements which affect the durability of cement lining [38]. The contact of cementitious materials with deionized water or low conductivity water would create concentration gradients. As a result, calcium ions in the pore solution would diffuse outwards into the aggressive solution environment.

On the other hand, the carbonation process comprises of chemical reactions involving calcium hydroxide and calcium silicate hydrates with carbon dioxide, resulting in the initial matrix leaching, and the formation of calcium carbonates and water [39–41].

The overall carbonation reaction can be described as:

$$\text{Ca(OH)}_2 + \text{H}_2\text{O} + \text{CO}_2 \rightarrow \text{CaCO}_3 + 2\text{H}_2\text{O}. \quad (1)$$

Though, carbonation could be seen in a long run, only dotted white precipitant on cement-lined coupon surfaces could be observed in this 20-day experiment. However, excess calcium content in the water resulted in calcium carbonate precipitant on the reactor walls and bottoms. The leaching of calcium hydroxide also raised the pH level from pH 8.0 to around pH 11.35 as shown in Fig. 9(D).

Fig. 10 shows the changes in water quality at pH 7.0, 8.0 and 8.4 after cement-lined coupons were immersed in CaMg water for 20 days. Magnesium concentration dropped from around 10 mg/L to zero (Fig. 10(D)). Magnesium content was recovered from the filter
membrane paper after filtration of the water sample and at the reactor bottom after experiment. Almost all the magnesium content transformed from dissolute ion to suspended solid form at the high pH level of 11.3 after experiment. Magnesium ions reacted with cement-lining to form insoluble form Mg(OH)$_2$ (brucite) in accordance to the following reaction equation [42].

\[
\text{Ca(OH)}_2 + \text{Mg}^{2+} \rightarrow \text{Mg(OH)}_2 + \text{Ca}^{2+}
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(2)

In reactors with iron coupons, water quality parameters, such as hardness, calcium concentration and alkalinity reduced to a similar level despite the pH differences. In comparison, in reactors with cement-lined coupons, alkalinity, hardness and calcium concentration increased due to the leaching of cement-lining as discussed previously. The extent of increment was not the same under different pH conditions for each water quality parameter. There was no notable relationship found between pH level and the changes in Ca$^{2+}$ concentration, hardness and alkalinity at the end of the experiment. In reactors with...
cement-lined coupons, the increment of total alkalinity and hardness ranged from 65% to 160% under different pH conditions. However, the calcium content leaching in water involved in complex mechanisms of combined diffusion–dissolution/precipitation process. The amount of release was related to the preparation process, porosity, lining thickness, density of cement-lining etc. [27,43,44]. Therefore, the extent of increment of calcium concentration, total alkalinity and hardness was unpredictable and inconsistent in this study. Any correlation between pH level and water quality parameters such as alkalinity, hardness and calcium concentration could not be ascertained as yet.

Fig. 11 presents the pH variations during the entire experimental period under each pH condition with cement-lined coupons immersed in Ca100 water. pH levels under all three initial pH conditions rose rapidly from the initial value in the first day and gradually slowed to a taper, approaching a plateau towards the end of the 20 day experiment. It took a longer duration for the water originally at pH 7.0 to approach a steady pH level as compared to the other two original pH conditions of pH 8.0 and pH 8.4. At all times during the course of the 20 day experiment, the water with original pH of 7.0 was slightly lower than the other two experimental pH whereas the difference in pH levels for water at original pH of 8.0 and pH 8.4 was not obvious. Nevertheless, the final pH attained in all reactors with cement-line coupons was approximately 11.3 regardless of the initial pH levels. All three conditions with initial pH levels at pH 7.0, 8.0 and 8.4 did not affect the final pH level after experiment in cement-lined coupon reactors. This phenomenon was the same for that with iron coupons.

3.4. Corrosion index

3.4.1. Langelier saturation index (LSI)

The corrosion index of Langelier saturation index (LSI) is an equilibrium model derived from the theoretical concept of saturation and provides an indicator on the degree of saturation of water with respect to calcium carbonate. With the increase in pH level from 7.0 to 8.4, LSI of Ca100 stock water increased from negative value to positive value (Fig. 12(A)). This implied the transformation of water from a condition of ‘no-scale’ formation environment at pH 7.0 to a preferred scale-forming environment at higher pH level. Similar as in Ca100 water, increase of pH level would also result in increased LSI to positive values in CaMg stock water (Fig. 12(B)). After immersion of iron coupons, LSI of water with ductile and cast iron coupons was relatively similar in a slightly negative range in Ca100 water due to the precipitation of calcium carbonate in water. Variation of LSI value after experiment with respect to the change of pH level was small and negligible compared with the difference of LSI values of stock water. The minor difference of LSI values suggested that the initial pH level in the range of 7.0 and 8.4 did not have any significant impact on escalating the possibility of scale-formation. In contrast, due to calcium leaching of the cement-lining, alkalinity, hardness, pH and calcium concentration increased, which hence resulted in an increased LSI value to positive value in Ca100 water with cement-lined coupons. The calculation of LSI has provided for the justification on the tendency of scale-formation in water containing cement-lined coupons.

3.4.2. Calcium carbonate precipitation potential (CCPP)

Calcium carbonate precipitation potential (CCPP), which is the quantity of calcium carbonate that can theoretically be precipitated from oversaturated waters or alternatively dissolved into unsaturated waters, was calculated for the remineralized water. A value of CCPP being larger than zero indicates that a system is supersaturated with calcium carbonate and it may help to prevent corrosion due to the formation of scales. Conversely, a CCPP value of less than zero indicates that the water is undersaturated with calcium carbonate and there is an increased tendency for corrosion to occur. A value of −5 to 0 mg/L as CaCO₃ indicates passive, −10 to −5 mg/L as CaCO₃ indicates mildly...
dissolving and below −10 indicates that the water would dissolve calcium carbonate [45].

As presented in Fig. 13, CCPP value of Ca100 stock water increased from −24.1 in pH 7.0 water to 4.3 in pH 8.0 water and to 5.7 mg/L as CaCO₃ in pH 8.4 water. Such an observation indicated a higher tendency for saturation of calcium carbonate and hence, corrosion prevention with rising pH level. With pH ≥ 8, CCPP value fell into the recommended range (3–10 mg/L as CaCO₃) of remineralization water quality in Table 1. In CaMg stock water, CCPP value was slightly lower than in Ca100 water at same pH level, while calcium carbonate supersaturated condition was also preferred in pH 8.0 and pH 8.4 water.

After immersion, CCPP values of Ca100 water with iron coupons under varied pH level conditions ranged from −2 to −4 mg/L as CaCO₃, which indicated undersaturated and passive calcium carbonate scaling condition. With magnesium addition, CCPP value ranged from −2 to 0.2 mg/L as CaCO₃, which created a neutral or slightly passive calcium carbonate scaling condition due to the higher alkalinity of CaMg water after experiment.

With cement-lined coupon, CCPP value of both types of water under all pH level conditions increased to extremely high value of around 200 to 500 mg/L as CaCO₃, which was due both to the dissolution of calcium content from cement-lining and rising pH level during the experiment. The high CCPP value was in agreement with LSI results, which indicated the high tendency of calcium carbonate scaling. It suggested that higher pH level in stock water increased the possibility of scale-formation. However, as the pH changed gradually under stagnant condition during the experiment, the impact of pH variations was not significant. With the presence of magnesium content, CCPP maintained at a higher level as compared with Ca100 water.

3.5. Corrosion rate

Comparing the corrosion rates between different materials, corrosion of cement-lined ductile iron was the lowest; corrosion rate was around 3.58 to 3.76 mpy in Ca100 water and 1.04 to 3.12 mpy with magnesium added. The most heavily corroded material was the cast iron,
which ranged from approximately 5.36 to 5.54 mpy in Ca100 water and from 5.21 to 5.47 mpy in CaMg water.

With magnesium addition, reduced corrosion rates could be observed at each pH level for all coupon materials. With iron coupons, the reduced corrosion rates of approximately 5%–9% were observed in CaMg water as compared with Ca100 water at each pH level. The reduction of corrosion rates of cement-lined coupons in CaMg water as compared with Ca100 water was distinguished with the reduction extent varied from 17% for pH 8.0 to more than 70% for pH 7.0 conditions. This result complied with previous studies that the addition of magnesium would effectively control the corrosion of pipeline material, especially cement-lined ductile iron material [13].

In studying the effect of pH level in corrosion control, graphs of average corrosion rate at various pH levels are demonstrated in Fig. 14. A similar tendency of corrosion rate was found for all the three materials. The corrosion rate constantly increased from pH 7.0 to pH 8.0 and reduced from pH 8.0 to pH 8.4 for all three materials investigated. Taking ductile iron in Ca100 water as an example, with calcium content added only, the corrosion increased from 4.74 to 4.90 mpy (from pH 7.0 to pH 8.0). When pH level increased to 8.4, the corrosion rate reduced to 4.82 mpy, with the standard deviation of the corrosion rates ranging from 0.29 to 0.38. The variance of the increase in corrosion rate, from 4.74 to 4.90 mpy and reduction from 4.90 to 4.82 mpy, was within the range of around 3%. While for cement-lined coupon, the difference between the highest and lowest average corrosion rate was around 5% at pH 8.0.

In CaMg water, a similar phenomenon on corrosion rates was observed for ductile iron and cement-lined coupons except for cast iron coupons. However, variation due to the pH change was extremely low for iron coupons compared with the cement-lined coupons. With pH level increased from 7.0 to 8.0, the corrosion rate increased by three times from 1.04 to 3.12 mpy. The corrosion rate reduced by 8% from 3.12 mpy at pH 8.0 to 2.87 mpy at pH 8.4, while the standard deviation ranged from 0.42 to 1.75.

Results of this study have shown a decreasing, followed by increasing corrosion rates from pH 7.0 to pH 8.4 in Ca100 water for all pipeline materials except cast iron in CaMg water. However, the level of significance of effects of the pH variations on corrosion of the various materials was different. Such differences could be attributed to the complex relationship between pH and pipeline materials.

Variation in corrosion rates of iron coupons was unpronounced and insignificant despite pH changes within the range of 7.0 to 8.4. pH levels investigated in this study were within the drinking water standard range, and therefore narrow and closely associated with other parameters, such as buffer capacity and LSI value. Conclusion from various past studies suggested that higher pH could result in a comparatively higher protection layer for iron materials which benefited corrosion control,
while it could also complement the lower buffering capacity at a narrow range which promoted corrosion control. The combined effects of pH within the range of 7.0 to 8.4 could bring out the little variation in corrosion rates.

With regard to the cement-lined coupons, variation in corrosion rates with pH change was negligible in CaMg water. An intense protection could be observed to the cement-lined coupons at pH 7.0 in CaMg water while no obvious difference was found between pH 8 and pH 8.4 conditions. Reduction of corrosion rate was primarily due to the leaching of free lime in cement-lining. The role of magnesium ion on the attack of concrete or cement paste was addressed in Alexander et al.’s study [18]. The formation of brucite with magnesium addition on the leaching of free lime in cement-lining provided an extraordinary protection to the iron coupons. No obvious iron oxide could be found on the cement-lined coupon surfaces. However, the orange-colored iron oxides and white crystal precipitant were found on both ductile iron and cast iron coupon surfaces in remineralized water. The main components of the iron oxides were lepidocrocite (γ-FeOOH), lepidocrocite (FeO·H₂O), goethite (α-FeOOH), iron hydroxide (Fe(OH)₃), goethite (Fe₂O₃·H₂O-xH₂O), iron oxide (Fe₂O₃) and calcium carbonate (CaCO₃). CaCO₃ was found adhered to the coupon surface and with Mg²⁺/Ca²⁺ larger than 0.5 at 25 °C, the aragonite, needle-like crystals were dominant over the calcite which were in rhombic shape crystal microstructures. However, within the same type of water, the variation of pH level had insignificant effect on the formation and composition of corrosion products.

In reactors with iron coupons, the addition of magnesium content not only changed the morphology and microstructure of calcium carbonate precipitation, but also reduced the amount of calcium carbonate precipitation and maintained a high alkalinity and hardness concentration compared to that when no magnesium was added. In comparison, leaching of calcium hydroxide from cement-lined coupons raised the pH level from pH 8.0 to around pH 11.3. At high pH level, magnesium content transferred from dissolved ion form to precipitant form. Within the proposed drinking water standard pH range of 7.0 to 8.4, the variations of pH level had little effect on the final water quality with all types of pipeline materials under stagnant condition.

The impact of pH level within the range of pH 7.0 to pH 8.4 on corrosion control of iron coupon materials was minor and insignificant. On the other hand, no obvious difference of corrosion rate was found between pH 8.0 and pH 8.4 conditions for cement-lined ductile iron coupon materials. An intense protection of cement-lined ductile iron coupon materials was observed in neutral pH 7.0 water with magnesium addition. The results suggested that within the recommended pH level range of distributed remineralized desalinated seawater, magnesium mineral addition would give better protection to all types of pipeline materials. It would be recommended that the relative low pH within drinking water pH range of magnesium added remineralized desalinated seawater would minimize the corrosion of the cement-lined ductile iron pipeline.

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References


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