

CHAPTER 1

Review of Iron Pipe Corrosion in Drinking Water Distribution Systems

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ABSTRACT

Iron corrosion is one of the most complicated and costly problems facing drinking water utilities. A large number of parameters affect pipe corrosion, including water quality and composition, flow conditions, biological activity, and corrosion inhibitors. This work synthesizes nearly 100 years of corrosion studies in an attempt to provide the water industry with an updated understanding of factors that influence iron pipe corrosion. In particular, this paper reiterates conclusions of prior studies regarding the Langelier Index—despite its continued widespread use, the Langelier Index does not provide an effective means of controlling iron corrosion. A review of potential implications of upcoming regulations for iron corrosion is also included.

INTRODUCTION

Cast iron pipes have been used to transport potable water for over 500 years,¹ and iron pipe corrosion has been a problem for just as long. The American Water Works Association (AWWA) estimates that it will cost US water utilities \$325 billion over the next 20 years to upgrade water distribution systems.² This AWWA value is built on the U.S. Environmental Protection Agency (USEPA) estimate of \$77.2 billion for service and replacement of transmission and distribution system lines over the next 20 years.³ The majority of distribution system pipes are composed of iron material: cast iron (38%), ductile iron (22%), and steel (5%).⁴ Moreover, a 1997 survey of the 100 largest American Water Works Association Research Foundation (AWWARF) member utilities found that “the most common distribution system problem is corrosion of cast iron pipe.”⁵

Although the body of literature on iron corrosion is quite large, the majority of studies were conducted at experimental conditions inconsistent with the potable water distribution system environment (e.g. brine solutions, oil and gas pipelines, highly acidic conditions, or very

high temperatures). The goal of this literature review is to summarize the results of several hundred articles relevant to the drinking water industry.

Corrosion of iron pipes in a distribution system can cause three distinct but related problems. First, pipe mass is lost through oxidization to soluble iron species or iron-bearing scale. Second, the scale can accumulate as large tubercles that increase head loss and decrease water capacity. Finally, the release of soluble or particulate iron corrosion-byproducts to the water decreases its aesthetic quality and often leads to consumer complaints of “red water” at the tap. The water industry must be concerned with all three of these aspects of corrosion.

FACTORS INFLUENCING IRON CORROSION

Iron corrosion is an extremely complex process. Because of the large variability in distribution system conditions, a particular factor may be critical in one system but relatively unimportant in another system. Moreover, corrosion itself has several different manifestations and can be evaluated in many ways. Previous studies on iron pipe corrosion have focused on different aspects of iron corrosion: pipe degradation (measured by weight loss, oxygen consumption or corrosion current), scale formation (measured by head loss or scale deposition), and by-product release (measured by iron concentration, color, staining, turbidity, or number of customer complaints). Thus, it can be difficult to compare conclusions or theories from different studies. For example, one study found that head loss increased with increasing pH,⁶ whereas another study saw decreased iron by-product release at higher pH.⁷ Although both results could be explained by greater incorporation of corrosion by-products into the scale at higher pH, from a utility perspective the former result is “bad” while the latter is “good.” Thus, this review carefully tracked which aspect of iron corrosion was studied (corrosion rate, scale formation, and iron by-product release). Table 1-1 presents a general overview of the effects of various factors on corrosion. Further explanation and references are provided in the following sections.

Water Quality Effects

Key water quality parameters that are expected to influence corrosion include pH, alkalinity, and buffer intensity.

Role of pH. Weight loss is generally found to increase with increasing pH in the range 7 to 9,⁸ as is the degree of tuberculation.^{6, 8, 9} In contrast, by-product release was decreased at higher pH.⁷ Again, this is consistent with increased corrosion by-products being incorporated into the scale layer. However, one study found that both weight loss and iron concentration decreased as pH was raised from 8.5 to 9.2.¹⁰

Alkalinity. Increasing alkalinity generally leads to lower weight loss^{10, 11} and corrosion rate.^{10, 12} Also, fewer customer complaints of red water were received when the alkalinity was maintained at greater than 60 mg/L as CaCO₃.¹³

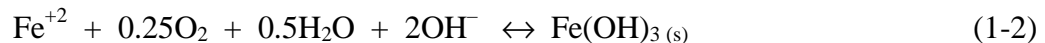
Buffer Intensity. Higher buffer intensity is often associated with increased alkalinity, although the two parameters are not exactly equivalent. However, their effect on iron corrosion seems to be similar. Several studies found the maximum weight loss for cast iron samples occurred at the minimum buffer intensity (pH 8.4) presumably because higher buffer intensity attenuates pH changes due to corrosion reactions at anodic and cathodic areas.^{8, 14-16} However, one study found the opposite effect: weight loss for iron coupons in stagnant water increased with increasing buffer intensity.¹⁷

Dissolved Oxygen

Dissolved oxygen (DO) is an important electron acceptor in the corrosion of metallic iron:



DO can also play a role in the oxidation of ferrous iron (Fe⁺²) or iron scales, for example:



Thus, oxygen concentration can have varying effects on iron corrosion. As expected, the corrosion rate increases with increasing DO.¹ However, effects on iron concentration and tuberculation may be mixed depending on the type of scale formed. Higher turbidity (a surrogate for iron concentration) was seen at lower oxygen saturation,¹⁸ but it is also reported that water free of DO will not tuberculate.¹⁹ DO is also responsible for the ability of buffering ions, including phosphates, to inhibit corrosion.^{8, 20, 21} For example, in water with DO < 1 mg/L,

solutions with phosphates had a higher corrosion rate compared to waters with no phosphates; in water with 1–6 mg/L DO, this trend was reversed.²²

Kuch Mechanism

In the absence of oxygen, it is possible for previously deposited ferric scale to act as an electron acceptor. This Kuch reaction produces ferrous iron and allows the corrosion reaction to continue even after DO is depleted:^{23, 24}



Types of Scale

The rate of corrosion of the iron metal has little relationship to the amount of iron that actually goes into the water, primarily due to the deposition of oxidized iron or other compounds into a scale which serves as a large reservoir of corrosion by-products. Iron scale is typically composed of many different compounds and is thus very heterogeneous (Table 1-2). This scale layer may provide passivation (protection) by limiting the diffusion of oxygen to the metal surface and slowing the corrosion reaction. However, the scale will also contribute iron to the water. This includes both soluble species from scale dissolution as well as scale particles that detach from the surface. Unfortunately, it is difficult to model this scale behavior. The scales are often poorly crystallized and heterogeneous, making solubility predictions very complex; even if solubility was understood, the water may be undersaturated due to kinetic or mass transfer limitations. Particulate detachment mechanisms are also complicated, depending on factors such as scale durability and adherence, water velocity, and temperature changes.

Clearly, scale formation is a complicated process that depends on a variety of physical and chemical conditions in each particular system. However, there are several well-known types of scale that are postulated to affect iron corrosion.

Calcium Carbonate. Precipitation of a thin layer of protective calcium carbonate (CaCO_3 , presumably calcite) was the earliest proposed method for controlling iron corrosion, and it was popular through at least the 1980s. Many papers have been dedicated to extolling the virtues of calcite layers,^{8, 25-32} and many others detail methods to achieve a perfect layer of

protective calcite (see next section). However, few papers ever demonstrated a beneficial role for calcite in controlling iron corrosion.

Siderite. The siderite model postulates that the formation of reduced iron species, especially siderite (FeCO_3), provides a more protective scale than oxidized ferric scales such as goethite (FeOOH) or hematite (Fe_2O_3).^{24, 33} Siderite has been found in many iron scales.^{8, 26, 34-38} However, it is interesting to note a discrepancy regarding siderite in studies of pure iron in high carbonate solutions (not drinking water). Two such studies concluded that siderite was the key to forming a protective scale.^{39, 40} However, two others with similar conditions found that the formation of siderite destroyed the protective nature of other iron oxide films.^{41, 42}

Green Rust. “Green rust” is the generic name given to iron compounds containing both ferrous and ferric iron, as well as other ions such as carbonate, chloride and sulfate.^{43, 44} Green rusts have been identified in the corrosion products on iron and steel,^{45, 46} and it is thought they may act similarly to siderite and form a dense protective film.²⁴

Corrosion Indices

Calcite Indices. The Langelier Index⁴⁷⁻⁵¹ (also called the Saturation Index, or S.I.) has been improperly applied as the cure-all method for solving corrosion problems since it was first proposed in 1936. While this method was successful at some utilities, it is by no means a universal way to control corrosion. To his credit, Langelier never intended the index to be used in this manner, and even pointed out its limitation in certain waters.⁵² Several other studies also found that the S.I. had no correlation with corrosion rate.^{8, 14, 53-55} Despite proposed modifications to the Langelier Index⁵⁶⁻⁵⁸ and its continued use by many utilities, the AWWA manual on corrosion states, “In light of much empirical evidence contradicting the presumed connection between the LI [Langelier Index] and corrosion, this practice should be abandoned.”²⁴ Likewise, other calcite indexes that have been proposed (Ryznar Index (RI),⁵⁹ Aggressiveness Index (AI),⁶⁰ Momentary Excess (ME),⁶¹ Driving Force Index (DFI),⁶² and Calcium Carbonate Precipitation Potential (CCPP);⁴⁸⁻⁵⁰ see reference ⁶² for review of all indices) have only limited use in corrosion control.

Larson Index. The studies of Larson ^{27, 63, 64} found that the ratio of chloride and sulfate to bicarbonate was important, as expressed in the Larson Index:

$$\text{Larson Index} = \frac{2[\text{SO}_4] + [\text{Cl}]}{[\text{HCO}_3]} \quad (1-6)$$

with a higher index indicating a more corrosive water. Several studies qualitatively confirmed these results (although they never directly tested the accuracy of the Larson Index) by reporting that increased chloride^{11, 65} and sulfate^{65, 66} caused increased weight loss. Another paper proposed that the presence of sulfate or chloride in the water leads to “Fe-Cl and/or Fe-SO₄ complexes” in the iron scale that increase Fe⁺² diffusion, causing increased iron concentrations.⁶⁷ However, several other studies found contradictory effects, including that sulfate inhibits dissolution of iron oxides, leading to lower iron concentrations,⁶⁸ the presence of sulfate and chloride caused a more protective scale on steel surfaces,³⁵ and that sulfate and chloride concentrations had no effect on weight loss of cast iron.^{15, 55}

Disinfectant Residual

In general, disinfectant residuals increase corrosion rates.^{14, 69} Monochloramine was found to be less aggressive than free chlorine.⁷⁰ However, if the corrosion is microbially induced, higher disinfectant residuals may decrease corrosion.⁷¹

Phosphate Inhibitors

Phosphate based inhibitors have been added to drinking water since the early 1900’s. Phosphates were first used to prevent excessive calcite precipitation.⁷² The mechanism of this “threshold treatment” was thought to be the sorption onto calcium carbonate nuclei, which prevented the calcite crystals from growing outside of the colloidal range.⁷³ Researchers later discovered that these phosphates could sometimes prevent iron corrosion and red water problems.

Polyphosphates. Condensed chain phosphates, known generally as polyphosphates, were the first phosphorus compounds to be used in corrosion control. The original polyphosphate used was Calgon (known as sodium hexametaphosphate, glassy phosphate, or metaphosphate glass), with the approximate chemical formula of Na₂₂P₂₀O₆₁. Numerous studies found that polyphosphate could prevent corrosion and/or control red water.^{6, 22, 74-91} Corrosion

prevention and red water control are in fact two very different phenomena, but this distinction was sometimes overlooked in these studies. Many reported beneficial results for corrosion are simply due to stabilization of iron particles, causing a decrease in the visual observation of “red water.” This led researchers to claim that iron by-product release had decreased, when in reality the iron concentration and even the corrosion rate might have increased.

The theory of polyphosphate corrosion prevention, if discussed at all in these papers, varies widely. Some researchers claimed that the polyphosphate adsorbed onto the iron surface to form a protective film.⁷⁵ Other studies have stressed the importance of calcium in polyphosphate effectiveness.^{22, 73, 75, 76, 79-82, 87-89, 92-98} This is believed to be due to formation of a thin calcite layer made protective by the adsorption of polyphosphate ions^{81, 87, 88, 99, 100}. Other studies have stressed the importance of flow conditions on the efficacy of polyphosphate action (see following section).

Orthophosphate. Unlike polyphosphates, orthophosphates are added for scale formation but not for iron sequestration.^{20, 89, 101-104} One often overlooked fact is that polyphosphates revert to orthophosphate with time,⁹³ so the results of many polyphosphate studies could be confounded by the presence of orthophosphate.

Bimetallic (Zinc) Phosphate. In the 1960's, manufacturers began blending poly- and orthophosphates with 5 – 25% zinc to form bimetallic phosphates, claiming either that the presence of zinc accelerated polyphosphate film formation or that a zinc ortho- or zinc polyphosphate film was superior to regular phosphate films for inhibiting corrosion. These compounds were reported to decrease corrosion compared to regular poly- or orthophosphates.^{94-96, 105-109} However, several studies found no benefit of zinc phosphates compared to regular phosphates.^{89, 90, 101, 108, 110-112} Moreover, recent concerns over zinc loading to wastewater plants has somewhat curtailed the usage of these zinc phosphates.²⁴

Other Mechanisms. There are several other phosphate effects that are traditionally not considered in the context of pipe corrosion. Phosphate sorption may restabilize iron scales as colloids.⁹² Polyphosphate has been shown to induce ligand-promoted dissolution of iron oxides, leading to higher iron concentrations in water (although very high phosphate concentrations may

actually cause a decrease in iron concentration.¹¹³) Orthophosphate can either inhibit or enhance ligand-promoted dissolution depending on pH.^{68, 114} Orthophosphate also dramatically decreases reductive dissolution of iron oxides.¹¹⁵

Silicates

Silicate compounds were first used as a coagulation aid because of their ability to adsorb onto particles and decrease surface charge.^{116, 117} These properties were soon applied to sequestration (dispersion) of iron particles in distribution systems.¹¹⁸⁻¹²² An early study by Riddick⁶⁶ found that natural silica present in the water was concentrated in the relatively protective iron scale. Addition of silicate-based inhibitors has also been found to reduce iron corrosion rate.^{12, 90, 123} Finally, silicates are reported to decrease the oxidation of Fe^{+2} because ferrous iron diffusion through the silicate film is slower than through iron oxide scale.¹²⁴

Silicates appear to form a self-limiting film on the corroded iron surface that will break down if silica dosing is stopped. It has been proposed that the metal must already be somewhat corroded because the protective film must consist of ferric oxide and silicate;²⁴ similarly, iron must be oxidized in order to be sequestered by the silicate.^{119, 121, 122} Addition of silicates can also raise the pH, which is generally beneficial towards iron corrosion. Silicates are naturally present in many waters, and may have effects similar to those of added silicate inhibitors.

Pipe Age

The length of time the pipe has been aged dramatically affects its corrosion. In general, both iron concentration and the rate of corrosion increase with time when a pipe is first exposed to water, but then gradually both are reduced as the scale builds up. However, the location of this “cross-over” point varies widely with water quality and other conditions.

Water Velocity

There are very mixed results for the effect of flow velocity (see reference ¹²⁵ and ⁷⁷ for a review). It is thought that two factors are dominant: increased flow provides more oxygen for the corrosion reaction, but it can also hasten the precipitation of a protective layer. For example, a study of mild steel found that the weight loss increased with increasing water velocity when the DO was saturated.^{1, 14} However, other studies found a more dense protective layer at higher

water flow rates.³⁸ Also, if the velocity is very high, the water can scour away the protective scale.

There is anecdotal evidence that phosphate inhibitors perform poorly at low flow or stagnant conditions^{74, 75, 78, 85, 123}, and several studies found that phosphate inhibitors had no effect or actually increased iron corrosion under such conditions^{6, 77, 112, 126}.

Stagnation Time

Iron concentrations were shown to increase with longer stagnation times.¹²⁷ One study from The Netherlands¹²⁸ found that turbidity in a cast iron distribution system peaked during the night, then decreased in the early morning as demand increased and stagnation time decreased.

Biological Activity

Microbes are present in many distribution systems,^{46, 71, 129-137} and they can influence iron corrosion in a number of ways. Bacteria have been found in iron tubercles.^{46, 132, 134, 136, 138} Growth of a bacterial biofilm on a pipe wall may serve as a barrier to corrosion,^{130, 139} but biofilms can also produce a differential aeration cell, leading to localized changes in oxygen concentration and electrical potential.¹³¹ The biopolymers in the biofilm may also uptake soluble metals.⁴⁶ Various bacteria can affect iron speciation by reducing Fe^{+3} or oxidizing Fe^{+2} .¹⁴⁰⁻¹⁴⁵ Bacteria also may consume oxygen,¹²⁹ cause localized pH gradients,⁴⁶ and produce corrosive metabolites such as H_2S ^{46, 135} or iron phosphide.¹⁴⁶⁻¹⁵² Thus, the role of biological activity in a water pipe can be mixed, but is generally considered to be detrimental to most aspects of iron corrosion. In cases where such activity is dominant, it is not surprising that biocides such as chlorine effectively reduce overall corrosion problems despite their oxidative properties.

Temperature

The effect of temperature on iron corrosion is often overlooked. Many parameters that influence corrosion can vary with temperature: dissolved oxygen (DO) solubility, solution properties (e.g. viscosity and ion mobility), ferrous iron oxidation rate, thermodynamic properties of iron scale (leading to formation of different phases or compounds), and biological activity. Moreover, heterogeneous iron scale formed on pipes may have large differences in

physical properties such as scale density (as described by the Pilling-Bedworth ratio PBR) and coefficients of thermal expansion.¹⁵³ If the scale is exposed to temperature gradients or cycling, these differences can cause mechanical stresses in the scale, leading to spalling or crack formation.

Only a few studies have examined the role of different temperatures in distribution system corrosion. In one study, weight loss decreased for iron samples held at 13°C versus 20°C.³⁸ Other studies found lower iron concentrations¹¹¹ and fewer customer complaints of red water¹³ during the colder winter months. Finally, two pilot scale studies saw peaks in turbidity¹⁵⁴ and metal concentrations¹⁵⁵ that appeared to correlate with temperature changes.

Effect of Other Compounds

Dissolved Copper. Two studies found that dissolved copper as low as 0.01 mg/L catalyzed iron corrosion rates.^{156, 157}

Natural Organic Matter. Several studies^{33, 158, 159} found that natural organic matter (NOM) decreased the corrosion rate of both galvanized steel and cast iron. NOM was also found to encourage a more protective scale¹⁶⁰ and alter the redox chemistry by reducing Fe^{+3} colloids to soluble Fe^{+2} .¹¹⁴ Finally, NOM can complex metal ions.²⁴

“Free CO₂”. Several studies stressed the importance of “free CO₂”, which is the sum of carbonic acid and dissolved carbon dioxide. These studies found that the free CO₂ forms surface complexes on the iron oxide, increasing the solubility of ferrous iron and thus increasing iron concentration and corrosion rates.^{17, 161-163}

Magnetic Effects

Some iron oxides are magnetic, such as magnetite, while others such as hematite are not. One final interesting theory is that precipitated ferric scale is attracted back to the pipe surface by magnetic force, so the amount of scale built up depends on its magnetic properties.¹⁹

IMPLICATIONS OF UPCOMING REGULATIONS

The drinking water industry faces numerous impending or recently promulgated regulations. Implementation of these new regulations may require significant changes to

treatment processes and finished water quality, thereby creating the potential for secondary effects on iron corrosion.¹⁶⁴

Lead and Copper Rule (LCR)

The LCR, first implemented in 1991, requires utilities to control lead and copper levels in tap water.¹⁶⁵ In order to comply with the LCR, most utilities either changed their finished water quality or began dosing phosphate inhibitors. Typical water quality adjustments are raising pH and lowering dissolved inorganic carbon. According to Table 1-1, raising pH is detrimental to iron corrosion rate and scale build-up, but can reduce by-product release.

Many utilities also began adding phosphate inhibitors to meet the LCR, often without regard to how the inhibitors would impact iron corrosion. In fact, from 1992 to 1994 the percentage of large utilities dosing inhibitors doubled from 30 to 60%¹⁶⁶ and may be even higher today. While these phosphate inhibitors can be effective in decreasing lead and copper corrosion under some conditions, they can also be quite detrimental to iron corrosion.¹¹²

Disinfectants and Disinfectant Byproduct (D/DBP) Rule

Implementing the D/DBP rule¹⁶⁷ can impact iron corrosion due to changes associated with disinfection and increased organic matter removal. As discussed above, decreased residuals generally decrease corrosion rates. However, decreased residual may increase corrosion if it is related to microbial activity. Many utilities may change to a different disinfectant (e.g. from chlorine to chloramine), which can also impact iron corrosion.

The second aspect of the D/DBP rule is the requirement for increased removal of organic matter (as measured by total organic carbon, TOC) through enhanced coagulation or enhanced softening. Organic matter is thought to be beneficial for iron corrosion rate and scale build-up, but detrimental to by-product release, so the overall effect of increased TOC removal is unclear. Other potential impacts of enhanced treatment have been documented¹⁶⁸. Briefly, enhanced coagulation can be associated with lower pH and alkalinity and higher sulfate and chloride concentrations due to increased coagulant doses. Enhanced softening may be associated with higher pH, lower alkalinity, and decreased hardness. According to Table 1-1, each of these factors may have mixed consequences for iron corrosion.

Enhanced Surface Water Treatment Rule (ESWTR), Groundwater Disinfection Rule (GWDR), and Total Coliform Rule

In contrast to the D/DBP rule, the primary concern for utilities trying to meet the ESWTR,¹⁶⁹ GWDR, or Total Coliform Rule¹⁷⁰ will be increased disinfection and disinfectant residual requirements. In addition, the Total Coliform Rule may require changes in circulation and flow patterns in the distribution system in order to decrease “dead ends.” This may be beneficial to corrosion, because these dead end areas are often the source of iron corrosion problems due to low flow conditions and decreased dissolved oxygen.

SUMMARY

Iron pipe corrosion is extremely complicated and is affected by practically every physical, chemical, and biological parameter in water distribution systems. This work provides a summary of key factors that utilities must evaluate in order to mitigate iron corrosion problems. Utilities should also consider potential secondary impacts on corrosion due to compliance efforts for new regulations.

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Table 1-1: Summary of expected effects of various factors on iron corrosion.
 “+” = beneficial effect, “-” = detrimental effect, “±” = mixed results, “?” = not known.

Factor	Potential Effect on...		
	Corrosion Rate	Scale Build-up	By-product Release
pH increase	-	-	+
Alkalinity increase	+	-	+
Buffer Intensity increase	+	?	?
Dissolved Oxygen increase	-	±	±
Kuch Reaction	-	?	-
Calcite Scale	+	-	±
Siderite Scale	+	?	+
Green Rust Scale	+	?	+
Langelier Index	Should Not Be Used!		
Larson Index increase	±	±	±
Disinfectant Residual increase	-	?	-
Phosphate Inhibitors	±	±	±
Silicates	+	+	±
Pipe Age increase	+	-	+
Water Velocity increase	-	±	±
Stagnation Time increase	?	?	-
Microbial Activity	-	-	-
Temperature increase	-	?	-
Dissolved Copper	-	?	?
NOM	+	+	-
“Free CO ₂ ”	-	?	-

Table 1-2: Selected iron solids

Name	Chemical Formula	Fe oxidation state
Ferrous hydroxide	Fe(OH) ₂	II
Ferric hydroxide	Fe(OH) ₃	III
Wustite	FeO	II
Goethite	α-FeOOH	III
Akaganeite	β-FeOOH	III
Lepidocrosite	γ-FeOOH	III
Hematite	α-Fe ₂ O ₃	III
Maghemite	γ-Fe ₂ O ₃	III
Magnetite	Fe ₃ O ₄ (FeO•Fe ₂ O ₃)	II and III
Ferric oxyhydroxide	FeO _x (OH) _{3-2x}	III
Siderite	FeCO ₃	II
Iron hydroxycarbonate	Fe _x (OH) _y (CO ₃) _z	III
“Green rust”	Fe(III) _{x1} Fe(II) _{x2} (OH) _y (CO ₃ ,SO ₄) _z	II and III
Vivianite	Fe ₃ (PO ₄) ₂ • 8H ₂ O	II
Strengite	FePO ₄	III
Schreibersite	Fe ₄ P	?