

**In-Situ Remediation of Small Leaks in Water Pipes: Impacts of Water Chemistry,
Physical Parameters and the Presence of Particles**

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ABSTRACT

Aging and leaking water infrastructure wastes water resources and creates public health risks. Upgrading of potable water systems represents a large financial burden for water utilities and private property owners. The conventional approaches of repair, rehabilitation and replacement are very effective, but will take decades to implement even if a financial commitment to do so was made immediately. A novel approach of in-situ remediation of leaks, achieved by harnessing the ability of water or pipe to repair leaks via clogging, could potentially reduce leak rates and extend the lifetime of existing infrastructure at relatively low cost and inconvenience.

Physical clogging, precipitation and metallic corrosion were identified as major mechanisms of in-situ leak remediation in potable water pipelines. Autogenous repair (i.e., self-repair without added particles) of small leak-holes (150–1000 μm) in copper and iron was validated in the laboratory at water pHs of 3.0–11.0, operating water pressures of 20–60 psi, upward and downward leak orientations, and for a range of water chemistries.

In bench scale experiments, the time to repair of iron pipe leaks increased with leak size to the power of 0.89–1.89, and decreased with pipe wall thickness to the power of -1.9 to -1.0. The time to repair of copper pipe leaks increased with water pressure to the power of 1.7. Additionally, the waters with a higher DO and corrosivity as measured by RSI, significantly decreased the time to repair of carbon steel 400 μm leaks by 50–70%. The presence of chlorine dioxide significantly increased the fraction of repaired 200 μm copper pipe leaks by 3 times when compared to the control without any disinfectant. In the building scale study, the fraction of repaired iron pipe leaks decreased with the logarithmic leak size with a slope of -0.65 after one-year duration of experiments, while leak orientation and water pressure were not influential in time to or likelihood of repair for iron pipe leaks.

Addition of calcium carbonate particles (~8.8 μm), silica particles (~29 μm) and wood ash particles (~160 μm) in Blacksburg, VA tap water at a water pressure of 10 psi increased the fraction of remediated iron pipe leaks of 280–1000 μm diameter sizes. Although the control condition with no added particles for 58 days resulted in remediation of 0/12 leaks, remediation rate increased to 1/12 with calcium carbonate particles, to 10/12 with silica particles and to 10/12 with wood ash particles. Leak size and particle size played an important role in controlling the remediation success rate.

The strength of the in-situ leak repair was sometimes very strong and resilient. The sealing materials of leak-holes repaired at 20–60 psi could sometimes withstand a 100 psi water pressure without failure, demonstrating the potential of the approach to sustain aging and

leaking infrastructure. In-situ leak repair can also occur naturally, and the success rate might be unintentionally altered by adjustment of chemistry or treatments that decrease or increase particulates.

In-Situ Remediation of Small Leaks in Water Pipes: Impacts of Water Chemistry, Physical Parameters and the Presence of Particles

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GENERAL AUDIENCE ABSTRACT

Old and leaking pipes waste water resources and can contaminate water. Upgrading of drinking water systems represents a large financial burden for water utilities and home owners. The traditional approaches to repair or replace the leaking water pipes are very effective, but will take decades to implement even if a financial commitment to do so was made immediately. A new approach of leak remediation, achieved by changing the drinking water chemistry, could potentially reduce leak rates and repair water leaks while in use without digging up the buried water pipes. Therefore, leak remediation could extend the lifetime of existing infrastructure at relatively low cost and inconvenience, and may be necessary if society cannot afford pipe replacement.

Recent field observations indicate that metal corrosion, one type of reaction that eats up water pipes and causes water leaks, could clog the leaks via the corrosion products. And the repair in most cases could last for a long period of time. Our work was the first to reproduce the field observations in the laboratory, revealing that water pipe leaks could be successfully clogged or remediated by natural corrosion reaction products, if the water chemistry is favorable.

Additionally, our work also showed that water leaks could be clogged or remediated by addition of water particles to drinking water, which was practiced by Roman engineers a long time ago. There are legitimate health concerns about particulates because they are indicative of microbial risks, but addition of particulates to water at low levels might heal pipes without such concerns.

We also proved that in some cases the repaired materials clogging the leaks could withstand a very high household water pressure, showing that the sealing materials in water leaks repaired with natural corrosion products or added water particles could create long term repairs. With improved practical understanding this approach might be usefully applied, either intentionally to repair existing leaks, or make sure that changes to water chemistry do not unintentionally make problems worse.

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AUTHOR'S PREFACE

“...When the pipeline is complete, and the water is first let into it, some **wood ash** should be thrown into the tank at the supply end. **This will find its way into any cracks or leaks in the system, and help to clog them up...**” –Marcus Vitruvius Pollio (c. 80-70 BC, *de Architectura*, Book VIII)

This dissertation is organized into six chapters. The first five chapters are in journal article format, with intellectual contributions reflected by the listed authorship.

Chapter 1 is a literature review of *in situ* remediation of water pipe leaks in potable water systems. Three remediation mechanisms (physical clogging, precipitation and metallic corrosion) were identified as a function of both physical and chemical parameters. This work provides a basis for a paradigm shift, in relation to understanding water leakage rates, and also in extending the lifetime of aging infrastructure in the United States. This chapter has been published in *Corrosion Reviews*: Tang, M., Triantafyllidou, S., Edwards, M. (2013) *In Situ Remediation Of Leaks In Potable Water Supply Systems*. *Corrosion Reviews*, 31(3-6), 105-122.

Chapter 2 confirms autogenous repair (i.e., self-repair) of metallic iron and copper pipe leaks (150–400 μm) in potable water for a range of water chemistries at pHs ranging from 3.0 to 11.0 and at water pressures of 20–40 psi in the laboratory for the first time. The strength of the sealing materials was also examined. This study successfully reproduced autogenous repair of iron and copper leaks in the laboratory as it was observed to occur in the field. It further proved that the resulting metallic pipe leak repair via corrosion precipitates was relatively strong and was a function of pH, pipe material and wall thickness. This chapter has been published in *Environmental Science & Technology*: Tang, M., Triantafyllidou, S., Edwards, M. (2015) Autogenous Metallic Pipe Leak Repair In Potable Water Systems. *Environmental Science & Engineering*, 49 (14), 8697-8703.

Chapter 3 extends the study of Chapter 2 and examines the impact of physical parameters (water pressure, leak size, pipe wall thickness and leak orientation) on autogenous repair of iron and copper pipe leaks in both bench-scale and building-scale experiments. The study indicates that smaller diameter leaks in thicker metallic pipes and at lower pressures would have the highest likelihood of repair. The sealing materials in many repaired leaks could withstand a water pressure of up to 100 psi without failure. This chapter has been accepted in *Corrosion*: Tang, M., Edwards, M. (2017) Impact Of Leak Size, Pipe Wall Thickness, Water Pressure And Leak Orientation On Autogenous Metallic Pipe Leak Repair. *Corrosion*, doi: <http://dx.doi.org/10.5006/2323>.

Chapter 4 explores the impact of water chemistry parameters (pH, presence of particles, ionic strength, alkalinity, silica, magnesium, disinfectants, natural organic matter, corrosion inhibitors, dissolved oxygen and Ryznar Stability Index) on autogenous repair of iron and copper pipe leaks in a bench-scale study. This study found that autogenous repair occurred at a wide range of water chemistries, and that corrosion sometimes could have

unexpected benefits, as it could serve to repair leaks as well as cause them in the first place. This chapter will be submitted to *Corrosion* for publication.

Chapter 5 studies physical clogging, another in-situ remediation mechanism, of iron pipes with added calcium carbonate, silica and wood ash particles in Blacksburg, VA tap water in a building-scale experiment. This work demonstrates that the presence of particles could increase the remediation rate (i.e., fraction of remediated leaks), and implied that the modern practice of improving removal of turbidity from potable water might have also reduced the likelihood of natural leak repair in the last few decades. This chapter will be submitted to *Environmental Engineering Science* for publication.

Chapter 6 highlights recommended future work that can help facilitate implementation of autogenous and in-situ leak repair in practice.

EXECUTIVE SUMMARY

WHAT IS THIS DISSERTATION ABOUT?

The objectives of this dissertation were to 1) identify the contributing mechanisms of in-situ remediation of water pipe leaks; 2) examine the impact of physical and chemical parameters on autogenous repair (i.e., self-repair) of metallic pipe leaks as they could influence the formation and size of waterborne and water-formed particles; 3) explore the impact of added water particles on *in situ* water pipe leak remediation; and 4) test the strength of the sealing materials in remediated leak-holes. The results will serve as a basis for in-situ remediation approach to sustain and extend the lifetime of existing water infrastructure in the United States.

A literature review was completed to identify three mechanisms (i.e., metallic corrosion, physical clogging and precipitation) that facilitate successful in-situ remediation of water pipe leaks, along with the impact of both physical and chemical parameters on leak remediation and the expected limitations of this approach (Chapter 1). The concept of autogenous metallic pipe leak repair (i.e., leak remediation without addition of particles) via corrosion precipitates was first validated in a bench-scale study featuring a typical recirculated potable water system (Chapter 2). The study was then extended to examine the impact of physical parameters (leak size, pipe wall thickness, water pressure and leak orientation) on metallic pipe leak repair in both bench-scale and building-scale experiments (Chapter 3). The impact of chemical parameters (pH, presence of particles, ionic strength, alkalinity, silica, magnesium, disinfectants, natural organic matter (NOM), corrosion inhibitors, dissolved oxygen (DO) and Ryznar Stability Index (RSI)) on autogenous iron and copper pipe leak repair was further explored as water chemistry controls the metallic corrosion therefore the likelihood of self-repair via corrosion precipitates (Chapter 4). The impact of added calcium carbonate, silica and wood ash particles that were favored by Roman engineers on iron pipe leak in-situ remediation was lastly tested in a building-scale experiment connected to a Blacksburg, VA tap water outlet (Chapter 5). The strength of the sealing materials in all bench-scale and building-scale experiments was examined via pressure tests to obtain the maximum pressure the sealing materials could withstand before leaking. The future research needed was discussed in relation to the finished work in this dissertation (Chapter 6).

The work in this dissertation was funded by National Science Foundation under grant CBET-1336616. All the experimental work was conducted at Virginia Tech Blacksburg campus.

WHAT RESULTS WERE OBTAINED FROM THE LITERATURE REVIEW?

Water pipe leaks in water mains and premise plumbing systems have very high costs and can also harm public health. If leaks could be remediated *in situ* while a pipeline is in service could at least temporarily, reduce water pipe leaks at costs orders of magnitude lower than conventional approaches of pipe repair, rehabilitation, or replacement. Experiences of Roman engineers and recent field observations suggest that such processes

can occur naturally, or may be engineered, to ameliorate leaks, including those caused by metallic corrosion. Three mechanisms of *in situ* leak remediation (i.e., metallic corrosion, physical clogging, and precipitation) are described in this chapter, in an effort to understand the role of physical factors (e.g., water temperature, water pressure, and leak size) and water chemistry (e.g., pH, alkalinity, corrosion inhibitors, dissolved oxygen, turbidity and disinfectants) in controlling *in situ* remediation for both inert (plastic and aged concrete) and chemically reactive (new concrete, copper, and iron) pipe materials (Table ES1). Although there are possible limitations and uncertainties with the phenomena, including the fraction of pipeline leaks to which it might apply and the durability/longevity of remediation, such approaches may prove useful in economically sustaining some aging drinking water infrastructure assets and reducing future pipe failure rates.

WHAT RESULTS WERE OBTAINED FROM PROOF-OF-CONCEPT EXPERIMENTS?

Conventional copper and iron pipes had a remarkable capability for autogenous repair (i.e., self-repair) of leaks caused by nails, rocks and erosion corrosion in the small bench-scale recirculation potable water system as shown in recent field observations (Table ES2). All (26/26) of the 150 μm leaks contacting representative bulk potable water (i.e., leaks not in contact with low pH and high conductivity pit water) in copper pipes sealed autogenously via formation of corrosion precipitates at 20–40 psi, pH 3.0–11.0, and with upward and downward leak orientations, as indicated by the reduction of leak flow rate to zero. All the same size leaks in carbon steel pipes at 20 psi self-repaired at pH 5.5 (3/3) and 8.5 (3/3), but two leaks did not self-repair permanently at pH 11.0 suggesting that water chemistry may control the durability of materials that seal the leaks and therefore the permanence of repair.

Larger 400 μm holes in copper pipes had much lower (0–33%) success of self-repair at pH 3.0–11.0, whereas all 400 μm holes in carbon steel pipes at 20 psi self-repaired at pH 4.0–11.0. This is possibly because carbon steel pipes corrode 10X faster than copper pipes.

Pressure tests indicated that 5–70% of the repaired carbon steel and copper leaks at 20–40 psi could withstand more than 100 psi without failure. Autogenous repair has implications for understanding patterns of pipe failures, extending the lifetime of decaying infrastructure, and developing new plumbing materials.

Table ES1. Summary of *in situ* remediation mechanisms in water pipes depending on both chemical and physical parameters as they are predicted to influence leak remediation (+ = increases remediation likelihood, - = decreases remediation likelihood, ± = mixed impact on remediation, ? = unknown).

Mechanism Relevant Pipe Materials	Fundamental Remediation Process	Possible Impact of Chemical and Physical Parameters on <i>In Situ</i> Remediation
Physical Clogging All	Waterborne and waterformed particles (e.g., calcium carbonate, silica) can physically block water pipe leak-holes	<ul style="list-style-type: none"> -Higher turbidity = increase presence of particles (+) -pH, ionic strength, phosphate corrosion inhibitors and NOM control surface charge and particle size distribution (±) -Large leak size and high water pressure hinder remediation (-) -Thick pipe wall promotes leak-hole remediation (+) -Leak orientation could influence the deposition of particles (?) -Water temperature affects precipitation, crystallization, diffusion, sedimentation and other aspects of waterborne particles (±)
Precipitation All if the location of precipitation can be controlled. Usually for new concrete pipes	Waterformed solids (e.g., calcium carbonate) or other precipitates (e.g., magnesium silicate, calcium silicate) via precipitation reactions could remediate leak-holes	<ul style="list-style-type: none"> -pH affects the permeability of cracks in concrete (±) -Si, Mg and Ca are constituents for concrete precipitates -Phosphate corrosion inhibitors and NOM inhibit the formation of calcium carbonate (-) -High water velocity, large leak size and high water pressure hinder leak-hole remediation (-) -Thick pipe wall promotes leak-hole remediation (+) -Leak orientation could influence the deposition of precipitations (?) -Water temperature affects the solubility of bonding materials, precipitation kinetics and other water properties (±)
Metallic Corrosion All metal (e.g., iron and copper) except stainless steel	Metallic corrosion can autogenously repair the leak-holes via formed corrosion rust precipitates from electrochemical reactions	<ul style="list-style-type: none"> -DO, pH, ionic strength, phosphate corrosion inhibitors, alkalinity, silica, NOM, water disinfectants (e.g., Cl₂, chlorine dioxide and chloramine) and water temperature affect both metal dissociation rate and rust precipitate formation rate (±) -Sulfate and chloride affect location of corrosion (±) -High water velocity could cause erosion corrosion but could form more corrosion precipitates if high redox (e.g., O₂, Cl₂) is present in water (±) -Large leak size and high water pressure hinder leak-hole remediation (-) -Thick pipe wall promotes leak-hole remediation (+)

WHAT RESULTS WERE OBTAINED FROM EXAMINING THE PHYSICAL PARAMETERS?

The effectiveness of autogenous metallic (iron and copper) pipe leak repair as a function of leak size, pipe wall thickness, water pressure and leak orientation was examined at water pressures up to 60 psi in both bench scale and building scale experiments (Table ES2).

In the bench scale experiments, the time to repair for carbon steel pipe leaks statistically increased with leak size to the power of 0.89–1.89, and decreased with pipe wall thickness to the power of -1.9 to -1.0. The time to repair for copper 150 μm leaks statistically increased with water pressure to the power of 1.7. However, the presence of large particles did not influence the capability or strength of repair for either carbon steel or copper pipe leaks.

In the building-scale experiments with only galvanized iron pipe leaks, water pressure and leak orientation did not influence the time to or likelihood of self-repair, but leak size was a limiting factor as the fraction of the repaired leaks (F) significantly decreased with leak size (L) (i.e., $F = -0.65 \ln(L) + 4.42$) at the end of the 366-day experiment.

The strength of repair in both scales of experiment was relatively strong and resilient, as at least some fraction of leaks autogenously repaired at 20–55 psi could withstand a high household plumbing pressure of 100 psi, indicating that smaller diameter leaks in thicker water pipes and at lower water pressures could naturally repair themselves at a faster rate.

WHAT RESULTS WERE OBTAINED FROM EXAMINING THE CHEMICAL PARAMETERS?

The impact of water chemistry (ionic strength, alkalinity, silica, magnesium, NOM, DO, disinfectants, corrosion inhibitors and corrosion indices) on autogenous repair of 400 μm carbon steel (6.35 mm OD x 890 μm wall) and 200 μm copper (6.35 mm OD x 890 μm wall) leaks were examined in modifications of a potable water at pH 8.5.

Higher DO and RSI significantly decreased the time to repair of carbon steel 400 μm leaks by at least 50–70% while other water chemistry parameters (ionic strength, alkalinity, silica, magnesium, NOM, disinfectants, corrosion inhibitors and Ryznar Stability Index) did not significantly influence the time to repair of the same size carbon steel leaks. The presence of chlorine dioxide resulted in a 300% increase in the fraction of repaired copper leaks with other chemical parameters (corrosion inhibitors, free chlorine, chloramine, DO, silica and NOM) exerting no significant impact on the fraction of repaired copper 200 μm leaks. The presence of particulate metal rust suspended in the water and weight loss of metals were not correlated with the time to repair of carbon steel 400 μm leaks or the fraction of repaired copper 200 μm leaks.

A significant fraction (15–38%) of the leaks repaired at 20 psi could withstand at least 100 psi pressure, confirming the potential for autogenous repair to extend to lifetime of existing water infrastructure in at least some circumstances.

Table ES2. Summary of the examined parameters and key findings.

Work Phase	Key Examined Parameters	Parameter Range		Key Findings
		Copper	Iron	
Chapter 2. Proof-of-Concept	pH	3.0–11.0	4.0–11.0	Leaks of 150–400 μm in copper and iron pipes autogenously repaired at 20–40 psi, pH 3.0–11.0 and with upward and downward leak orientations.
	Water pressure (psi)	20, 40	-	
	Leak orientation	upward, downward	-	
Chapter 3. Physical Parameters	Pipe wall thickness (μm)	360, 890, 1700	890, 1200, 1700	The time to repair for iron pipe leaks decreased with pipe wall thickness to the power of -1.9 to -1.0.
	Leak diameter (μm)	150, 250, 400	150, 280, 400, 700, 1000	The time to repair of carbon steel pipe leaks at a 20 psi water pressure increased with leak size to the power of 0.89–1.89. The fraction of repaired galvanized iron pipe leaks at 20–55 psi water pressures decreased with the ln of leak size with a slope of -0.65.
	Water pressure (psi)	20, 40, 60	20, 40, 55	The time to repair of copper leaks increased with water pressure to the power of 1.7.
	Leak orientation	-	upward, downward	Leak orientation did influence the time to repair or repair likelihood.
Chapter 4. Water Chemistry	pH, ionic strength, alkalinity, silica, magnesium, natural organic matter (NOM), dissolved oxygen (DO), disinfectants (free chlorine, chlorine dioxide and chloramine), phosphate corrosion inhibitors (orthophosphate and polyphosphate), Ryznar Stability Index (RSI)			The waters with higher DO and RSI significantly decreased the time to repair of carbon steel 400 μm leaks by 50–70% and the chlorine dioxide significantly increased the fraction of repaired 200 μm copper pipe leaks by 3 times compared to the control with no disinfectant.
Chapter 5. Presence of Particles	Particles		calcium carbonate, silica, wood ash	Addition of silica particles (~29 μm) and wood ash particles (~160 μm) increased the fraction of remediated iron pipe leaks by 9 times at a water pressure of 10 psi, compared to that with calcium carbonate particles (~8.8 μm).
	Leak diameter (μm)	-	280, 400, 700, 1000	

WHAT RESULTS WERE OBTAINED FROM EXAMINING THE PRESENCE OF ADDED PARTICLES

In situ clogging of leaks by waterborne or water-formed particles was examined in the building-scale experiments at a water pressure of 10 psi (Table ES2). Addition of calcium carbonate particles (~8.8 μm), silica particles (~29 μm) and wood ash particles (~160 μm) that were favored by Roman engineers, increased the fraction of remediated leaks in iron pipes to 1/12, 10/12 and 10/12, respectively, compared to that in the control condition with no added particles (0/12).

The highest remediation rate for wood ash particles, which were 4.5–17.2X larger but at 87–97% smaller particle concentration versus calcium carbonate and silica particles,

indicates that particle size played an important role in controlling iron leak remediation success.

As expected, smaller leaks were repaired with a greater remediation success rate over the 58 day duration of this experiment. Calcium carbonate had 33% success for 280 μm leaks versus 0% success for 400–1000 μm leaks; wood ash had 100% success for 280–700 μm leaks versus 67% for 1000 μm leaks; and silica had 100% success for 280–400 μm leaks versus 67% for 700–1000 μm leaks.

The strength of the leak clogging was strong, since 27–73% of the remediated leaks could withstand a water pressure of 100 psi, depending on particle type.

WHAT FUTURE RESEARCH WILL BE NEEDED?

This work provides a basis for implementing the *in situ* remediation approach in practice by examining various physical/chemical parameters and the presence of added particles. The impact of water temperature, water flow/velocity, pipe wall roughness, shape of leak-holes, leak size ($> 1000 \mu\text{m}$) and metal pipe electrochemistry as influenced by cathodic protection and galvanic coupling on leak remediation is worthy of future study. If possible, examination of *in situ* remediation in a larger pilot scale system and with real potable water could give insight into applying this approach in the field and extending the lifetime of existing water infrastructure.

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CHAPTER 1. *IN SITU* REMEDIATION OF LEAKS IN POTABLE WATER SUPPLY SYSTEMS

Min Tang, Simoni Triantafyllidou, and Marc A. Edwards

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ABSTRACT

Water leaks in distribution system mains and premise plumbing systems have very high costs and public health implications. The possible *in situ* remediation of leaks while a pipeline is in service could reduce leaking at costs orders of magnitude lower than conventional pipe repair, rehabilitation, or replacement. Experiences of Roman engineers and recent field observations suggest that such processes can occur naturally or may even be engineered to ameliorate leaks, including those caused by metallic corrosion. Three mechanisms of *in situ* leak remediation (i.e., metallic corrosion, physical clogging, and precipitation) are described in this paper, in an effort to understand the role of physical factors (e.g., temperature, pressure, and leak size) and water chemistry (e.g., pH, alkalinity, corrosion inhibitors, dissolved oxygen, and turbidity) in controlling *in situ* remediation for both inert (plastic and aged concrete) and chemically reactive (new concrete, copper, and iron) pipe materials. Although there are possible limitations and uncertainties with the phenomenon, including the fraction of pipeline leaks to which it might apply and the durability/longevity of remediation, such approaches may prove useful in economically sustaining some aging drinking water infrastructure assets and reducing future failure rates.

KEYWORDS: *In situ* remediation; leaks; mechanisms; premise plumbing; water mains

INTRODUCTION

A large fraction of the U.S. potable water infrastructure is leaking and is on the verge of failure (an overall condition grade of D), representing one of the most urgent societal infrastructure challenges (U.S. EPA 2010; ASCE 2011, 2013) and the single most challenging issue facing the water industry (Wallis-Lage and Chevrette 2012). The issue profoundly impacts consumers from the perspective of both water mains and premise plumbing piping. Similar problems have been noted in European and Asian distribution systems, where it is reported that leakage rates ranged from 3% to 70% from one country to another over the past 15 years (TEPPFA 2007; Frauendorfer and Liemberger 2010).

Distribution System Water Mains

Distribution system water mains in the United States are typically composed of cementitious pipes (concrete or asbestos cement), plastic pipes [polyvinyl chloride (PVC), chlorinated PVC, or high-density polyethylene], and iron pipes (cast, ductile, or steel) (Tomboulia *et al.* 2004; Folkman *et al.* 2013). Many such systems were installed after World War II and are reaching the end of their design lifetime of 60 to >100 years (U.S. EPA 1982). Water main breaks are currently occurring at a rate of 240,000 per year and rising (Walker and Schaefer 2009; U.S. EPA 2010). Aside from obvious public health implications associated with compromised delivery of uncontaminated drinking water to the tap, failure events can cause extensive property damage and water loss (U.S. EPA 2002). The U.S. Environmental Protection Agency (EPA) estimated that about \$200 billion needs to be invested in existing infrastructure to ensure distribution of safe drinking water in the next 20 years (Shanaghan 2012). The American Water Works Association (AWWA) took into account population growth needs and reported in 2012 that at least \$1 trillion must be invested in drinking water infrastructure through 2035 in the United States (Shanaghan 2012). The American National Standards Institute (ANSI) noted that many of the pipes installed in recent decades have thinner walls than the pipes installed a century ago (PPFA 2002), and as a result, there are many cases in which newer pipes are failing at a faster rate (and sooner) than older pipes. The estimated cost of replacing failing infrastructure mains is \$6300 on average per household in the United States over the next 10 years (AWWA 2001).

Premise Plumbing Systems

An underappreciated problem of similar or perhaps greater magnitude is associated with privately owned potable water plumbing systems in U.S. buildings (Scardina *et al.* 2007), which are mostly composed of copper, galvanized iron, PVC, or cross-linked polyethylene (PEX) pipes (Tomboulia *et al.* 2004). Nonuniform copper corrosion was estimated to cause 750,000 copper pinhole leaks annually in premise plumbing systems, comprising about 50% of the total estimated leaks in premise plumbing materials nationwide (Scardina *et al.* 2007). Plastic plumbing materials' leak rates are similar to those determined for copper pipe, and the overall costs of all leaks to consumers are currently about \$1 billion each year in repair and property damages alone (Bosch and Sarver 2007; Scardina *et al.* 2007). The net present value of premise plumbing in buildings is on the order of that for public water mains, and replacing a premise plumbing system can be an expensive

unforeseen expense to consumers (up to \$4000–\$10,000 or even higher for single family homes) (Edwards 2004; Scardina *et al.* 2007).

As aging potable water distribution systems fall deeper into disrepair and the costs of traditional replacement increasingly dwarf society's ability to invest in upgrades, innovative approaches that can extend the lifetime of existing assets are urgently needed.

Corrosion: Usually Detrimental But Sometimes Beneficial

A major cause of pipe leaks is corrosion (Walker and Schaefer 2009; Folkman *et al.* 2013). Estimates by the AWWA, the American Society of Civil Engineers (ASCE), U.S. EPA, the Water Infrastructure Network, and the National Academy of Corrosion Engineers suggest that direct costs for inspection, repair, or rehabilitation of water pipeline corrosion range between \$8 billion and \$36 billion annually and indirect costs are much higher (U.S. EPA 2002; U.S. FHWA 2002). Since the 1970s, the U.S. water industry has invested heavily in research and application of corrosion control measures (pH/alkalinity adjustment and dosing of certain corrosion inhibitors into the water), with the intent to reduce all types of water main and premise plumbing corrosion to the extent possible (AWWA 2011). A cost-benefit analysis for such efforts suggested that investing each \$1 in corrosion control for water mains reduced costs from corrosion by \$1–\$5 (Ryder 1980; Koch *et al.* 2001). A cost-benefit analysis for premise plumbing corrosion control has been considered even more favorable at about \$5 or \$6 saved for each \$1 invested (Ryder 1980). While understanding of corrosion control is imperfect, much progress has been made in reducing corrosion and controlling costs.

The possibility of benefits sometimes arising from corrosion is emerging. It was recently discovered that existing pipe materials, such as leaking copper pipes, have a remarkable ability to self-remediate in some water chemistries as a result of certain corrosion processes (Scardina *et al.* 2008). Discovery of this heretofore unknown mechanism of pipe remediation, which has now been reported to occur naturally in at least two water systems (Scardina *et al.* 2008; Lytle and Nadagouda 2010), raises prospects of a class of potentially transformative approaches to extending the lifetime of pipeline assets. That is, after more than a century of effort to minimize corrosion at every opportunity, the notion that some types of corrosion might actually be used to repair some existing leaks, or that attempts to control corrosion might sometimes increase the incidence of leaks, is an important consideration and possible paradigm shift. While much more research is needed to document these phenomena and to harness this potential, this paper presents the first comprehensive review of research concepts related to pipeline leak *in situ* remediation.

Concepts And Definitions For Leak Remediation Of Pipelines

The idea of leak remediation of pipes while in service, by manipulating the chemistry of water flowing through pipes, dates back to at least 15 BC, when Roman engineer Vitruvius noted that leaking terracotta pipelines could be permanently sealed by a one-time addition of wood ash to the water (Pollio 15 BC as translated by Morgan 1960). This general concept was seemingly lost for two millennia, until 1836 when it was noted that new cement exposed to water had an ability to heal cracks (Phillips 1925; Turner 1937; Hearn 1998).

Lauer and Slate (1956) defined “autogenous healing” as the ability of cement paste and concrete to heal cracks occurring in the parent material.

The term “*in situ* remediation of leaks” includes both autogenous and nonautogenous repair mechanisms and is defined herein as the ability of any pipe material (plastic, concrete, or metal) to physically-chemically repair or stop existing leaks via interaction with waters while in normal service. *In situ* pipe leak remediation can occur for pipe materials that are relatively chemically reactive with water (e.g., iron, copper, and concrete) or relatively chemically inert (e.g., plastic and stainless steel). Chemically active materials can undergo aqueous chemical reactions such as corrosion or dissolution/precipitation, which can locally alter water chemistry and create solids within existing leak holes, in a manner that can actively promote leak remediation. This type of *in situ* repair is defined as “autogenous” herein. In contrast, chemically inert materials that do not react with water significantly can only be remediated nonautogenously; for example, by dosing or forming high concentrations of particles in the water that can clog leak holes.

The types of leaks encountered also vary with pipeline materials and circumstance. Iron and copper (and sometimes plastic) pipes often initially leak through very small holes termed “pinholes”, whereas concrete pipes (or plastic pipes) often initially leak through very small cracks (Figure 1-1). A primary measure for tracking success of *in situ* remediation is the flow rate through each leak, as per prior studies with concrete materials (Wang *et al.* 1997; Ramm and Biscopig 1998; Edvardsen 1999; Tsivilis *et al.* 2003; Li and Yang 2007), with the ultimate goal of completely stopping leaks and restoring the pipeline integrity.

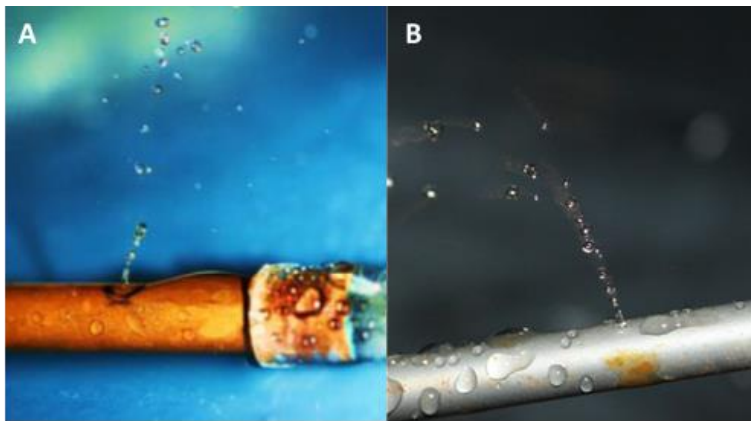


Figure 1-1. Leak types depend on pipe material. Copper (A) (Adapted with permission of Sarver 2010), iron (B) and other metallic materials tend to form holes.

Some Encouraging Exemplars Of *In Situ* Remediation of Leaks

In situ remediation of concrete in water mains

The Delaware Aqueduct is an extreme exemplar of infrastructure challenges associated with many water mains in the United States. The Aqueduct provides half of New York City’s daily water demand via the world’s longest concrete-lined tunnel (Simpson *et al.* 2009). After 65 years of service, very small cracks have formed in the concrete liner of

this engineering marvel, leaking 35 million gallons of water per day at a cost of \$28 million annually for water alone (Kennedy 2001; NYC 2008; Simpson *et al.* 2009). The leaks have also flooded homes, created massive sinkholes, and raised fears of a catastrophic pipeline failure event, which would have devastating economic and public health consequences for the city (Simpson *et al.* 2009). Conventional repair of leaks via construction of a 2.5-mile bypass tunnel for just one part of this pipeline is scheduled to be completed no sooner than 2021 at a cost of \$2.1 billion (NYC 2011).

The inability to easily access the 91–455 m (i.e., 300–1500 ft) deep pipeline or even shut it off for inspection/repair prompted an evaluation of novel approaches to mitigate leaks, reduce the chance of failure, and extend the lifetime of the pipeline. A bench-scale study of *in situ* remediation via temporary formation of suspended calcium carbonate by increasing the pH of the water was undertaken with very promising results (Letterman *et al.* 2008). The change in water chemistry was able to reduce the rate of leaks by 55% in just 2.5 h by clogging simulated cracks (Letterman *et al.* 2008). This novel approach is currently being pilot-tested as a promising stop-gap measure, to buy time and increase the chances that effective repairs can be implemented on a cost, plan, and schedule suitable to ratepayers and the utility. The approach is justified by the short-term benefit alone, but permanent or semi-permanent repair is also a possibility, as was proven by Roman engineers.

In situ remediation of copper pinholes in premise plumbing systems

A case study by Scardina *et al.* (2008) demonstrated that remediation of copper pinholes is sometimes occurring naturally in premise plumbing without our prior knowledge (Figure 1-2). Specifically, two adjacent cities used essentially the same source water, but one had very high incidence of copper pinhole leak damages in buildings while the other did not. Thorough investigation revealed that a similar number of leaks were probably occurring in both cities. The difference was that in one city after a few drops of water leaked the copper pinhole would completely heal itself (Figure 1-2, B) before the consumer even became aware of the problem. This seems to be analogous to autogenous (self) repair that sometimes occurs for concrete, but which occurs as a result of metallic corrosion filling the leaks with scale (rust), as opposed to formation of precipitates in the concrete cracks as a byproduct of concrete dissolution reactions. The only evidence of leaks were visually repaired holes roughly every 0.08 m (1/4") along a 0.9 m (3') copper pipe at a pressure of 344,738 Pa (50 psi) with no resulting water damages or repair costs. This particular case of *in situ* remediation has now been documented to persist for more than a decade. In the adjacent city, the leaks grew rapidly after they first formed (Figure 1-2, A), and each leak created up to thousands of dollars of water damage per incident. One city distributed their water at a slightly different pH and used a different type of corrosion inhibitor. It is therefore speculated that differences in water chemistry likely determined whether the leaks eventually caused the failure or *in situ* remediation, because there were no significant differences in the copper pipes. Preliminary investigation showed that the materials in the sealed holes were mainly copper corrosion precipitates, with some silica, calcium, and magnesium compounds.

Another case study by Lytle and Nadagouda (2010) verified the same observation by studying copper pitting corrosion in buildings of Cincinnati, Ohio. The water dripping rate through certain copper pinhole leaks decreased, due to blue-green corrosion deposits, such as $\text{Cu}_4(\text{OH})_6\text{SO}_4$ and Cu_2O , which formed in the pit structure. Aluminum and silica were also identified in the corrosion deposits. No microbiological activity was identified; thus, water chemistry was demonstrated to play an important role to form the deposits, stopping or reducing the leaks. The water had high pH (~ 8.8), chlorine dioxide ($0.1\text{--}0.37\text{ mg/L}$), low alkalinity ($\sim 50\text{ mg/l as CaCO}_3$), and significant levels of chloride ($\sim 64\text{ mg/L}$) and sulfate ($\sim 120\text{ mg/L}$).

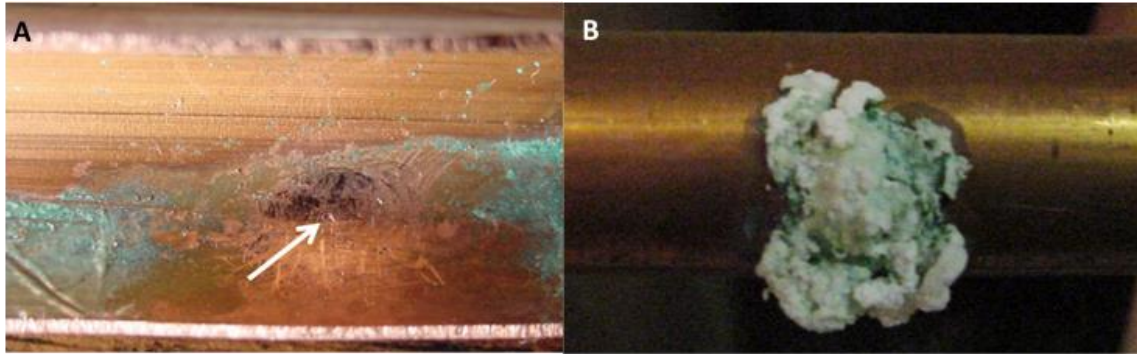


Figure 1-2. Conventional wisdom is that leaks invariably get larger with time (A) (Adapted with permission of Scardina and Edwards 2008), but it has been discovered that in some real-world situations copper leaks can be remediated if appropriate water chemistry is present (B) (Adapted with permission of Scardina *et al.* 2008).

MECHANISMS OF *IN SITU* REMEDIATION

Three mechanisms can induce *in situ* remediation of leaks, including corrosion for metallic pipes, physical clogging for all pipe materials, and active precipitation for concrete (Table 1).

Metallic Corrosion

Electrochemical (reduction-oxidation) reactions in nascent leaks of iron or copper pipes can release iron or copper ions and form corrosion rust [e.g., $\text{Fe}_2\text{O}_3(\text{s})$ or $\text{Cu}_2(\text{OH})_2\text{CO}_3(\text{s})$, respectively] (Table 1-1), which may eventually seal leak-holes because rust solids occupy a greater volume than the metal itself (Figure 1-3) (Evans 1963; Shreir *et al.* 1994; McNeill and Edwards 2001). For instance, each $1 \times 10^{-3}\text{ m}^3$ of iron metal that rusts (i.e., corrodes) can create 2×10^{-3} to $4 \times 10^{-3}\text{ m}^3$ of rust deposit at high pH (Zemajtis 1998). However, if the corrosion products are soluble ions or small colloids, and rusts are not formed that adhere to the pipe, then the hole will not be filled but will instead grow rapidly in size. It is therefore hypothesized that the critical path of a nascent leak to either *in situ* remediation or eventual catastrophic failure depends on the overall balance between the rate at which metals are eaten away and the rate at which scale forms along the wall of holes.

Table 1-1. Summary of metallic corrosion and other mechanisms as contributors to in-situ remediation of leaks in water pipes depending on materials.

Mechanism	Applies to Pipe Materials	Fundamental Healing Process	References
1 Metallic Corrosion	Iron	Iron corrosion rust precipitates in the leaks by electrochemical reaction, e.g.: $4Fe + 2H_2O + 3O_2 = 2Fe_2O_3 \cdot H_2O(s)$ Illustrative half reactions are: Anode (i.e., oxidation): $Fe = Fe^{2+} + 2e^-$ Cathode (i.e., reduction): $O_2 + 2H_2O + 4e^- = 4OH^-$	Evans 1963; McNeill and Edwards 2001; Shreir <i>et al.</i> 1994
	Copper	Copper corrosion rust precipitates in the leaks by electrochemical reaction, e.g.: $2Cu + H_2O + O_2 + CO_2 = Cu_2(OH)_2CO_3(s)$ Illustrative half reactions are: Anode (i.e., oxidation): $Cu = Cu^{2+} + 2e^-$ Cathode (i.e., reduction): $O_2 + 2H_2O + 4e^- = 4OH^-$	Evans 1963; Shreir <i>et al.</i> 1994
2 Physical Clogging	All (plastic, concrete, iron, copper)	Waterborne particles (e.g. bacteria, clay, dust, aluminum hydroxide, sand, etc.) can physically block pipe cracks/leaks	Clear 1985; Hearn 1998; Ramm and Biscopig 1998; Edvardsen 1999; Neville 2002; Letterman <i>et al.</i> 2008
3 Precipitation	New Concrete	Water-formed particles can seal leaks through precipitation reactions, e.g.: <ul style="list-style-type: none"> • Carbonation: $Ca^{2+} + CO_3^{2-} = CaCO_3(s)$ • Hydration: Calcium silicate (or magnesium silicate) precipitation: $Ca(OH)_2 + H_4SiO_4 \rightarrow Ca^{2+} + H_2SiO_4^{2-} + 2H_2O \rightarrow CaH_2SiO_4 \cdot 2H_2O(s)$ 	Munday <i>et al.</i> 1974; Wagner 1974; Clear 1985; Helmuth <i>et al.</i> 1993; Hearn 1997; Hearn 1998; Ramm and Biscopig 1998; Edvardsen 1999; Santhanam <i>et al.</i> 2002; Neville 2002; Li and Yang 2007; Parks <i>et al.</i> 2010

Table 1-2. Metallic corrosion forms commonly occurring in pipelines and their potential effects on existing leaks (Adapted from Schweitzer 2006).

Metallic Corrosion Form	Process	Informed Speculation Regarding Influence on Existing Leaks and In-Situ Remediation
<i>Uniform Corrosion</i>	Even rate of metal loss over the exposed surface	Passive film formed on the surface could potentially prevent leaks or create solids that could detach and/or plug existing leaks.
<i>Galvanic Corrosion</i>	Dissimilar metal corrosion due to the difference of electric potential between two metals	The corrosion rate of the less noble metal is locally accelerated, increasing the likelihood of corrosion and leaks but perhaps also increasing the likelihood of in-situ remediation. Conversely, the more noble metal is protected, with a lesser likelihood of corrosion and leaks, but possibly a lower likelihood of remediation.
<i>Crevice Corrosion</i>	Localized corrosion due to the difference of oxygen concentrations	When exposed to oxygen, species stable in the crevice may rapidly oxidize to form solids (e.g., CuO and Fe(OH) ₃) involved in in-situ remediation.
<i>Pitting Corrosion</i>	Localized loss of metal	Existing leaks would be expanded. Pits usually appear as deep and tiny holes in metallic pipes. Upon pit breakthrough and initial leak, low DO solution and low pH solution contacts air and precipitates may form (e.g. Figure 1-2).
<i>Erosion Corrosion</i>	Deterioration due to cavitation or macro-cell formation	Localized attack might be difficult to remediate, and high velocities through pinholes might dramatically accelerate rate of hole growth.
<i>Biological Corrosion</i>	Deterioration due to corrosive metabolites	Biofilms and biological reaction products might assist in physically blocking holes or accumulating solids.

The rate of metallic corrosion and identity of the corrosion products is controlled by water constituents such as alkalinity, pH, corrosion inhibitors (e.g., polyphosphate), natural organic matter (NOM), dissolved oxygen (DO), and water disinfectants (e.g., free chlorine) (AWWARF 1996). The pressure and resultant velocity of water in holes, temperature, and other physical factors may also likely influence the ability of rust to form and block holes (Wagner 1974; McNeill and Edwards 2001, 2002; Rushing and Edwards 2004a; Scardina *et al.* 2008).

Metallic corrosion is known to occur as either non-uniform corrosion (e.g., galvanic, crevice, pitting, erosion, and biological corrosion) or uniform corrosion (Schweitzer 2006), and each of these forms may have implications for the likelihood of *in situ* remediation (Table 1-2).

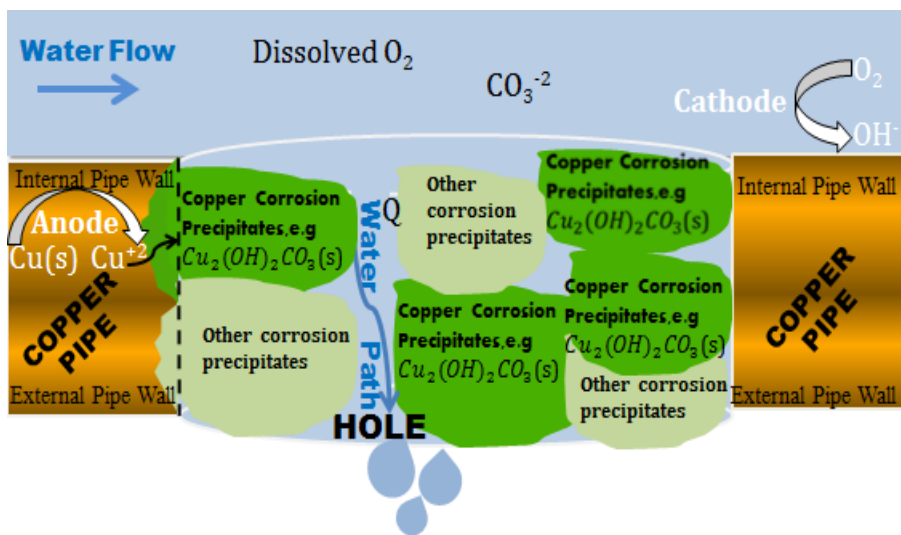


Figure 1-3. Cross section through a conceptual pinhole in a leaking copper pipe. Corrosion reaction precipitates can contribute to remediation of leaks.

Physical Clogging

Water treatment utilities attempt to minimize waterborne turbidity from particles such as soil, bacteria, sand, clay, $\text{Al}(\text{OH})_3(\text{s})$, or $\text{Fe}(\text{OH})_3(\text{s})$. Utilities try to stop these particles from entering distribution systems via coagulation and filtration processes and also to minimize generation of particles in the distribution system such as rust derived from iron pipe. These approaches are credited with protecting public health from waterborne pathogens and improving aesthetics by reducing discolored water complaints. However, practices of Roman engineers indicate that suspended particles may be beneficial by clogging holes, stopping leaks, and perhaps extending the lifetime of water pipes (Table 1-1; Figure 1-4) (Clear 1985; Hearn 1998; Ramm and Biscopig 1998; Edvardsen 1999; Neville 2002). Hearn and Morley (1997) described physical clogging of concrete cracks in infrastructure by entrapment of small particles and formation of a relatively impermeable filter cake. This type of approach has recently been applied to seal leaks in pressurized oil and gas pipelines via injection of discrete particles that seek out and then seal leaks (Ryan *et al.* 2007) and in the practical feasibility studies of crack clogging by suspended calcium carbonate particulates for simulated Delaware Aqueduct water leaks (Snyder 2009).

Clogging of membranes with well-defined micropores (Benjamin and Lawler 2013) provides some insight into leak remediation in pipelines. Specifically, pressure gradient would be the main driving force for particles transport to and then through the leak hole. The likelihood that waterborne particles passing through a small leak would contact the hole wall and contribute to clogging is controlled by sieving, diffusion (Brownian diffusion and shear-induced diffusion), and charge attraction/repulsion between formed particles and the hole wall, which in turn are a strong function of particle size, leak diameter, and surface chemistry. Sieving can play an important role in remediation when the particle diameter is nearly equal to or exceeds the size of leak. In general, smaller particles ($<4 \times 10^{-8}$ m) are likely to collide and attach due to Brownian diffusion, whereas larger particles ($>1 \times 10^{-6}$ m) are likely to attach due to shear-induced diffusion (Benjamin and Lawler 2013). Other than diffusion, the motion of an isolated particle is also a function of drag force, London-van der Waals attractive force, and electrical force.

The likelihood of successful attachment to leak walls (and clogging) may be a particularly strong function of surface charge for both the surface of leak holes and the waterborne particles. If the surface charges, as indirectly measured by zeta potential of the particles and leak walls, are low (often -1.0×10^{-2} to $+1.0 \times 10^{-2}$ V), the likelihood of particle attachment and entrapment in a leak is very high (Tseng *et al.* 2000; Davis *et al.* 2001). Higher surface charge (more than $+1.5 \times 10^{-2}$ V or less than -1.5×10^{-2} V) can prevent attachment and might prevent clogging due to the charge repulsion (Tseng *et al.* 2000; Davis *et al.* 2001). Because the surface charge of waterborne particles suspended in distribution systems is dependent on particle type, water pH, phosphate, NOM concentration, and other water constituents (Naidu *et al.* 1997; Davis *et al.* 2001; Lytle and Snoeyink 2002), water chemistry and particle type will determine whether particles pass through or clog holes in a pipe. These same parameters might be manipulated to engineer improved pipeline leak outcomes and distribution system lifetimes.

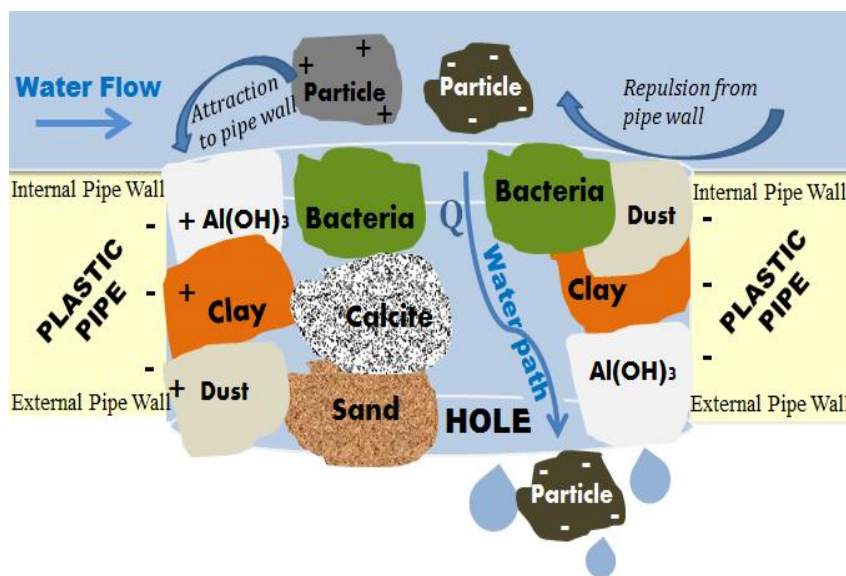


Figure 1-4. Cross section through a conceptual crack of a leaking plastic pipe. Physical clogging by water particles can seal leak-holes and cracks in all types of materials, including inert pipes (e.g., plastic or aged concrete).

Precipitation

Precipitation of carbonation products and hydration products as a result of lime dissolution are considered the main mechanisms of autogenous healing for cracks in new concrete (Table 1-1; Figure 1-5) (Munday *et al.* 1974; Clear 1985; Helmuth *et al.* 1993; Hearn 1998; Ramm and Biscop 1998; Edvardsen 1999; Parks *et al.* 2010). Hydration refers to the reaction of flowing water with unhydrated concrete to form hydrated precipitates (mainly calcium silicate hydrate or C-S-H) that seal cracks (Hearn and Morley 1997). Carbonation refers to precipitation of calcium with carbonate in the flowing water to form calcium carbonate precipitates [CaCO_{3(s)}] (Lauer and Slate 1956; Wagner 1974; Clear 1985; Hearn and Morley 1997; Edvardsen 1999; Neville 2002; Li and Yang 2007). This process requires sufficient carbonate ion present from water, whereas the calcium can be from water or leaching of calcium hydroxide (i.e., lime) from new cement (Neville 2002). Calcium hydroxide was detected in the material clogging leaks in some waters (Neville 2002). Precipitation of magnesium solids is another possibility if sufficient magnesium and silica are present at the high pH present in leak pores (Santhanam *et al.* 2002; Parks *et al.* 2010). However, as concrete “ages” with time, the once reactive concrete pipe becomes relatively unreactive with water constituents and unable to further leach lime and other precipitates, reducing this dimension of remediation (Table 1-1).

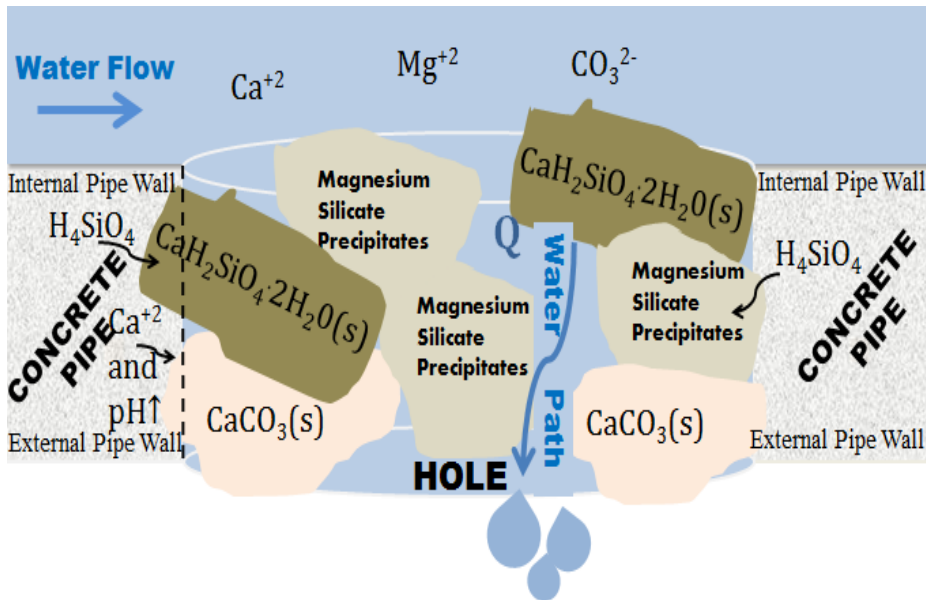


Figure 1-5. Cross section through a conceptual crack in a leaking concrete pipe. Carbonation and hydration reaction precipitates can seal cracks in concrete pipes.

PHYSICAL AND CHEMICAL IMPACTS ON CREATION OF LEAKS IN WATER PIPELINES

Some physical and chemical factors can cause localized corrosion or pinholes depending on circumstances. Specifically, high water velocity can cause erosion corrosion of metallic pipes, occasionally resulting in leaks (Schweitzer 2006). Temperature can cause cracks in concrete pipes due to thermal expansion (Neville 2002). High pH (9.0) and low alkalinity also favor localized pitting corrosion, which may result in pinhole leaks in copper pipes

(Rushing and Edwards 2004b; Marshall and Edwards 2005, 2006; Lytle and Schock 2008; Ha *et al.* 2011; Sarver *et al.* 2011; Sarver and Edwards 2012). NOM in water distribution systems is a carbon source for microbes such as sulfate-reducing bacteria, which can cause microbially induced copper and iron pitting corrosion and pipeline leaks (Hamilton 1985; Jacobs *et al.* 1998; Jacobs and Edwards 2000; Scardina *et al.* 2008). Sulfate can cause pitting corrosion in iron and copper pipes depending on the flow rate and pH condition (Szklaarska-Smialowska 1978; Shemilt *et al.* 1980; Edwards *et al.* 1994a, b, c; Duthil *et al.* 1996). Chloride has complex impacts on metallic pitting corrosion depending on the circumstances (Mattsson and Fredriksson 1968; Edwards *et al.* 1994b, c; Duthil *et al.* 1996). Water disinfectants such as free chlorine could also prompt pitting corrosion (Marshall 2004; Sarver and Edwards 2012).

Physical Factors

Potentially dominant physical factors include water velocity, pressure on leaks, temperature of water, and leak size in the pipeline (Table 1-3).

Table 1-3. Summary of expected effects of physical factors and water chemistry on in-situ remediation of leaks in water pipes.

I = Inert material, R = Reactive material, + = Increases likelihood of remediation, - = Reduces likelihood of remediation, ± = Mixed effects on remediation, ? = Unknown

Parameter		Potential Effect on In-Situ Remediation in				References
		Plastic (I)	Concrete (R when new)	Iron (R)	Copper (R)	
PHYSICAL FACTORS	Water Velocity increase	?	-	±	±	Wagner 1974; Rodolfo <i>et al.</i> 1987; Scardina <i>et al.</i> 2008; Vargas <i>et al.</i> 2010; Taxén <i>et al.</i> 2012; Calle <i>et al.</i> 2007
	Pressure increase	-	-	-	-	Edvardsen 1999; Neville 2002; Scardina <i>et al.</i> 2008
	Temperature increase	-	-	-	±	Neville 2002; McNeill and Edwards 2000; Boulay and Edwards 2001; Lytle and Nadagouda 2010; Obrecht and Quill 1960a, b, c, d, e and f
	Leak Size increase	-	-	-	-	Neville 2002; Li and Yang 2007; Wang <i>et al.</i> 1997
WATER CHEMISTRY	pH increase	?	+	±	±	Ramm and Biscopig 1998; McNeill and Edwards 2001; Sarin <i>et al.</i> 2004; Scardina <i>et al.</i> 2008; Lytle and Nadagouda 2010; Pourbaix 1984; Edwards <i>et al.</i> 1994 b; Edzwald 2011; Moulin and Rogues 2003; Kosmulski 2009; Noh and Schwarz 1989; Montgomery 1985; Naidu <i>et al.</i> 1997
	Carbonic species increase	?	?	±	±	Ha <i>et al.</i> 2011; McNeill and Edwards 2001; Edwards <i>et al.</i> 1996; Broo <i>et al.</i> 1998; Dodrill and Edwards 1995; Rehring and Edwards 1996
	Alkalinity increase	?	?	±	±	Broo <i>et al.</i> 1997; McNeill and Edwards 2001; Sarver and Edwards 2012; Mishra <i>et al.</i> 1992
	Calcium carbonate increase	?	+	?	?	Lauer and Slate 1956; Neville 2002; Hearn 1998; Hearn and Morley 1997; Clear 1985; Parks <i>et al.</i> 2010; Letterman <i>et al.</i> 2008

Natural Organic Matter increase	?	-	±	±	Zhang 2005; Murray-Ramos 2006; Korshin <i>et al.</i> 1996; Lin <i>et al.</i> 2005; Rehring and Edwards 1996; Campbell and Turner 1983; Edwards <i>et al.</i> 1994 b, c; Sarver and Edwards 2012; Stumm and Morgan 1996
Phosphate Inhibitors increase	?	?	±	±	McNeill and Edwards 2000; McNeill and Edwards 2001; Lewandowski <i>et al.</i> 2010; McNeill 2000; Edwards and McNeill 2002; Vargas <i>et al.</i> 2009
Sulfate increase	?	±	±	±	Santhanam <i>et al.</i> 2003; Santhanam <i>et al.</i> 2002; Trautenberg and Foley 1971; Neville 2004
Chloride increase	?	?	±	±	Edwards <i>et al.</i> 1994 b, c; Mattsson and Fredriksson 1968; Elzenga <i>et al.</i> 1987; McNeill and Edwards 2001; Van der Merwe 1988
Magnesium , Silica, Calcium increase	?	+	±	±	McNeill and Edwards 2001; Rushing 2003; Lytle and Nadagouda 2010; Scardina and Edwards 2008; Santhanam 2003; Santhanam 2002; Davis <i>et al.</i> 2001; Parks <i>et al.</i> 2010
Dissolved Oxygen increase	?	?	+	+	McNeill and Edwards 2001; Sarin <i>et al.</i> 2004; Lytle and Nadagouda 2010; AWWARF 1996
Disinfectant (ClO ₂ , Cl ₂ , NH ₂ Cl) increase	?	?	±	±	LeChevallier <i>et al.</i> 1993; Zhang <i>et al.</i> 2008; Zhang 2005; Hallam <i>et al.</i> 2002; Frateur <i>et al.</i> 1999; Sarin <i>et al.</i> 2001; Nguyen 2005; Edwards <i>et al.</i> 2000; Hamilton 1985; Volk <i>et al.</i> 2000; Sarver <i>et al.</i> 2011; Pourbaix 1974; Lytle and Nadagouda 2010; Nguyen <i>et al.</i> 2010
Turbidity increase	+	+	+	+	Hearn 1998; Edvardsen 1999; Clear 1985; Neville 2002; Ramm and Biscopig 1998; Letterman <i>et al.</i> 2008; Morgan 1960
Ionic Strength increase	?	?	±	±	Dunn <i>et al.</i> 2000; Smart and Bockris 1992; Edzwald 2011; Zhang <i>et al.</i> 2002

Water velocity

Water velocity within the pipe can influence many aspects of corrosion, including the type of corrosion (i.e., uniform, erosion corrosion, and cavitation) or the chemical and mechanical conditions at the metal-solution interface, and concentration gradients of dissolved and particulate constituents at the pipe wall (Calle *et al.* 2007; Vargas *et al.* 2010; Taxén *et al.* 2012). Crossflow filtration in membrane gives insight into the potential effect of water velocity on the transport of water constituents within a leak. Specifically, the water velocity is expected to influence diffusion, the shear stress between water flow and the wall, and therefore the potential volume of particles potentially deposited (or eroded from within) on the leaks. As an example of how lower water flow can contribute to autogenous repair, stagnant water allowed healing of cement cracks five times faster compared to water velocity of 0.025 m/s in one study, although differences in the test specimens and crack geometry provide only strong suggestive proof (Wagner 1974).

Water velocity above 3 m/s can cause copper pipe to fail in field studies due to wall penetration from erosion corrosion (Scardina *et al.* 2008). Conversely, higher water velocities may be beneficial when delivering more oxygen to the holes/cracks, allowing formation of more corrosion precipitates, which can potentially seal leaks. One study

showed that more oxide precipitates were formed at higher velocity (1.2 m/s) in mild steel pipes than lower velocity (0.6 m/s) with 9.0 ± 0.1 mg/l DO at 20°C (Rodolfo *et al.* 1987), and similar trends might occur within scale formed within leaks. One experiment with flushing demonstrated that high water velocity (0.167 m/s) caused detachment of more particles from the copper pipes into the water mainly due to the greater shear stress than the lower water velocity (0.022 m/s) (Vargas *et al.* 2010).

Overall, it is expected that lower velocities within leaks might enhance the likelihood of particle attachment and physical blocking of plastic pipe holes by waterborne/water-formed particles. While delivering a larger volume of particulate matter to the proximity of holes would be beneficial, higher scouring rates and reduced attachment would be detrimental. It can also be expected that impacts of velocity would be complex and highly dependent on circumstances.

Pressure

Higher pressure in the pipeline would be anticipated to reduce the likelihood of *in situ* remediation, as it increases the force on deposits that are needed to plug the holes/cracks and the detachment of accumulated particles. Experimental studies showed that four times higher water pressure (97,000 vs. 24,000 Pa; i.e., 14 vs. 3.5 psi) on 2×10^{-4} m wide concrete cracks could lead to 50% less healing, as measured by water flow through the cracks (Edvardsen 1999). Pressure might also be an impediment to *in situ* remediation if the sealed leaks fail to withstand the imposed pressure, and the pressure load versus strength recovery of the sealed cracks/hole may become one key to long-term success of *in situ* remediation (Neville 2002). The capability of sealed cracks or holes to withstand a typical 140,000–420,000 Pa (20–60 psi) pressure in premise plumbing systems, or even higher pressure (420,000–1,400,000 Pa; i.e., 60–200 psi) in water mains (Woodson 2006), might be limiting factors. One field study cited herein demonstrated that remediation of leaks in copper pipes by forming scales could occur even under typical premise plumbing pressures of 350,000 Pa (50 psi) (Scardina *et al.* 2008) and the Roman systems also sometimes operated at high pressure ($>1,400,000$ Pa; i.e., 200 psi; Pollio 15 BC as translated by Morgan 1960). No experimental or other field data are currently available to demonstrate the role of pressure on the three mechanisms of *in situ* remediation.

Temperature

Temperature is a master variable due to its role in solubility, kinetics of precipitation, crystallization, diffusion, sedimentation, and virtually all aspects of *in situ* remediation mechanisms. Temperature can also affect DO solubility, corrosion precipitates' solubility, redox kinetics, and biological activity in water (McNeill and Edwards 2000). Neville (2002) found that lower temperature helped heal cracks in concrete and speculated the enhancement was due to thermal contraction of leaks, whereas high temperature increased the crack size and delayed the remediation of leaks. Temperature has a complicated effect on *in situ* remediation since it would affect the kinetics of the dissolution reaction from metal (tending to increase the hole size) and the corrosion or precipitation reactions (tending to decrease the hole size). The destiny of leaks and possibility of *in situ* remediation would depend on which trend is dominant.

A short-term (4.5 months) experiment with intact iron pipes showed that lower temperature (5°C) created slightly more corrosion products on the pipe surface and more iron leaching into water compared to higher temperature (20–25°C) (McNeill and Edwards 2000). The much bigger volume of the corrosion precipitates created in this study might suggest an improved likelihood of *in situ* remediation. On the contrary, higher temperature would generally accelerate the kinetics of other reactions that enhance growth of leaks for copper including erosion corrosion and release of particulates (Obrecht and Quill 1960a, b, c, d, e, f). For intact copper pipes, a 6-month study indicated that higher temperature (60°C) would release a higher proportion of particulate copper in soft water (5 mg/L alkalinity as CaCO₃) compared to lower temperature (4°C, 20°C, and 24°C) (Boulay and Edwards 2001). Hot temperature could, on the contrary, accelerate the aging of copper pipes and help to develop a stable uniform layer of corrosion products on the pipe surface (Lytle and Nadagouda 2010). As a result, the dominant rate between metal leaching rate and corrosion product formation rate in leaks determines the pathway toward remediation or failure. As was speculated for concrete, any pipe will expand with higher temperature and contract with lower temperature.

Leak size

Larger leaks are expected to be more difficult to seal due to the need to accumulate more adherent precipitates to clog up the leaks, and the greater total force that must be withstood by the area of new deposit in the leaks. Various authors reported that *in situ* remediation of concrete was not possible in reasonable time periods if the crack width exceeded a certain threshold (Wang *et al.* 1997; Neville 2002; Li and Yang 2007). This threshold ranged from 150 µm for cracked engineered cementitious composite specimens in a study by Li and Yang (2007) to 200 µm in research by Wang *et al.* (1997). The reported threshold size of a repairable crack width varies in different studies because of variations in water chemistry and other test conditions as well as differences in the concrete test specimens. Considering the novel concept of *in situ* remediation for metallic and inert pipes, no prior studies have examined the effect of leak size in these materials. It is logical to expect that larger leaks in copper, iron, and plastic pipes would also be more challenging to *in situ* remediation, similar to observations for larger concrete cracks.

Water Chemistry

Water chemistry plays a central role in the corrosion process of all reactive pipe materials, including iron, copper, and concrete. Specifically, pH, alkalinity, carbonic species, calcium carbonate, sulfate, chloride, NOM, corrosion inhibitors, DO, calcium, magnesium, silicate, water disinfectants, and ionic strength affect corrosion in metals and concrete and by extension would probably affect leak *in situ* remediation as well (Table 1-3). Turbidity, a surrogate indicator of particle concentrations in water, is hypothesized to beneficially affect physical clogging for the *in situ* remediation of all pipes including inert materials, but it is uncertain how other water parameters would affect *in situ* remediation of relatively inert plastic pipes (Table 1-3).

pH

pH is also a master variable in drinking water systems, which controls many chemical reactions in water. Both acidic and basic conditions can be corrosive to metallic pipes, but

acidic conditions often tend to favor metallic ions as reaction products (i.e., soluble form), whereas basic conditions tend to form corrosion precipitates (i.e., insoluble form). This trend would be expected to increase the likelihood of success of *in situ* remediation at higher pH. Indeed, in iron pipes, higher pH (pH 7–9) was generally found to develop a greater volume of corrosion products on pipe surfaces (McNeill and Edwards 2001), a necessary step in the remediation of leaks. Another study indicated that higher pH (7.5–9.5) also decreased soluble iron release into the water, minimizing the chance of red water (Sarin *et al.* 2004). At very high pH, however, it is possible that corrosion rates would be substantially reduced, to the point that leak clogging by corrosion precipitates might be impeded.

For copper pipes, some studies identified a difference between the pH of local water near the pinhole (pH 3.5–5.5) and the pH of the bulk water inside the pipes (pH 8.8) (Pourbaix 1984; Edwards *et al.* 1994b; Lytle and Nadagouda 2010). This local pH change might also play a role in preventing remediation of copper pinholes, because the lower pH near the leak would accelerate corrosion and creation of soluble ions. For concrete, a 2-year study on reinforced concrete cracks showed that moderate pH (pH 7) lowered the flow rate through cracks, while lower pH (pH 5.2) allowed more flow through cracks (Ramm and Biscopling 1998). This indicated potential success of *in situ* remediation at higher pH and a path to failure at lower pH.

Aside from effects on reactive pipe materials, pH may influence physical clogging of leak holes in inert pipes. pH determines the point of zero surface charge (PZC) for waterborne particles and plastic pipes in drinking water systems, which in turn influences the size of suspended particles present in water by coagulation processes. The attractive or repulsive force between particles and crack/leak walls will also be highly dependent on the pH for the point of zero charge (pH-PZC) and therefore affects the chance of particles adhering to block pipeline cracks or holes. Thus, at or near the PZC of waterborne or waterformed particles, the likelihood of larger particles and attachment to holes (Figure 1-4) will tend to be maximized. The pH-PZC is controlled by the type of particles (Naidu *et al.* 1997). For example, aluminum hydroxide, calcium carbonate, and silica particles are commonly present in potable water supplies and their pH-PZCs are quite different. The pH-PZC is 8.5 for aluminum hydroxide, 8–10 for calcium carbonate, and 2–4 for silica in relatively pure water (Montgomery 1985; Noh and Schwarz 1989; Moulin and Rogues 2003; Kosmulski 2009; Edzwald 2011). Thus, clogging of leaks in inert materials by waterborne particulates is expected to demonstrate different optimal pH dependent on the particles present.

Carbonic species/alkalinity/calcium carbonate

Calcium and carbonate are two important components participating in *in situ* remediation of concrete materials. Carbonate species participated in self-repair of concrete in experiments, and the material repairing concrete cracks was mainly calcium carbonate (Lauer and Slate 1956; Clear 1985; Hearn and Morley 1997; Hearn 1998; Neville 2002; Parks *et al.* 2010). As a common waterborne particle in natural water sources or by purposeful formation/addition, suspended calcium carbonate was proven capable of physically clogging simulated cracks of aged concrete pipes in a bench-scale study (Letterman *et al.* 2008).

In copper pipes, one study proved that the soluble copper concentration in bulk water leached from copper pipes increased with the free carbon dioxide (Broo *et al.* 1998; Rehring and Edwards 1996). In monitoring for copper to tap water, the highest copper concentration was observed when alkalinity (7.4 mg/L as HCO_3^-) and pH (6.4) were very low (Dodrill and Edwards 1995; Rehring and Edwards 1996; Broo *et al.* 1998), and corrosion rates showed a similar dependence (Broo *et al.* 1998; Rehring and Edwards 1996). Overall, the presence of carbon dioxide and alkalinity can be expected to exert a strong effect on the corrosion rates of copper, copper speciation, and the likelihood for *in situ* remediation.

For cast iron pipes, alkalinity is a controlling factor in multiple steps of the corrosion process and can be expected to influence leak remediation. For example, high alkalinity and higher buffering intensity tend to reduce the corrosion rate (McNeill and Edwards 2001; Sarver and Edwards 2012) and the corresponding volume of corrosion precipitates that form. Carbonate could also help remediation by formation of siderite (FeCO_3) scale, which is believed to be more protective in pipes compared to ferric species, such as goethite (FeOOH) and hematite (Fe_2O_3) (Mishra *et al.* 1992; McNeill and Edwards 2001). Free carbon dioxide, on the contrary, could complex with iron oxides and cause higher soluble ferrous iron in water (McNeill and Edwards 2001) and possibly put leaks on a path to failure.

NOM

NOM is an important constituent of drinking water controlling corrosion rates, colloid stability, and solubility (Rehring and Edwards 1996). NOM interactions are very complex due to its inherently heterogeneous nature, and after decades of research, these reactions are moderately well understood if not perfectly predictable chemically. Thus, leak *in situ* remediation in the presence of NOM may be complicated due to its complex secondary impacts on water chemistry and microbes.

For iron pipes, NOM (0.2 mg/L Aldrich humic acid) in one study had no effect on iron corrosion rates under both saturated and low oxygen conditions when microbes were absent (Zhang 2005). However, in another study, NOM increased the formation of iron oxide scale layers and inhibited iron dissolution (Campbell and Turner 1983), potentially increasing the likelihood of *in situ* remediation. For copper pipes, NOM was an effective corrosion inhibitor when its concentration was above 0.3 mg/L in aggressive water, as defined by Larson's index (Edwards *et al.* 1994b, c; Rehring and Edwards 1996; Murray-Ramos 2006). NOM is also known to reduce leaks by stopping copper pitting corrosion (Edwards *et al.* 1994; Sarver and Edwards 2012). Korshin *et al.* (1996) also showed that NOM could markedly enhance non-uniform corrosion tendencies and potentially form more precipitates in a narrow range of concentrations (0.1–0.2 mg/L), therefore enhancing the likelihood of leak *in situ* remediation.

Calcite (calcium carbonate) precipitation could also be poisoned by NOM (Lin 2005; Lin and Singer 2005; Lin *et al.* 2005) and inhibit self-healing of concrete. NOM can also coat suspended particulates and decrease the zeta potential, thereby stabilizing colloids in

smaller size ranges (Stumm and Morgan 1996), which could tend to prevent clogging of leaks.

Phosphate corrosion inhibitors

Polyphosphate, orthophosphate, zinc orthophosphate, zinc metaphosphate, and bimetallic phosphate are commonly used iron and copper corrosion inhibitors in the United States (McNeill and Edwards 2000, 2001; Lewandowski *et al.* 2010). These inhibitors affect both the metal release rate and scale formation rate within existing leaks, thereby influencing *in situ* remediation. For example, McNeill (2000) showed that the presence of 1 mg/L orthophosphate as P at pH 9.5 decreased iron release from cast iron pipe into water but increased scale buildup on the pipe wall, relative to conditions without phosphate. The greater rate of scale buildup might indicate that phosphates could assist in remediation. Corrosion inhibitors can also slow the copper corrosion rate and reduce the copper release rate by forming a protective insoluble layer [such as $\text{Cu}_3(\text{PO}_4)_2$] outside the pipe (Edwards and McNeill 2002; Vargas *et al.* 2009; Lewandowski *et al.* 2010). This also indicates a potential for *in situ* remediation of copper pipe leaks.

Magnesium, silica, and calcium

Silica is naturally present in water and can be added as a corrosion inhibitor. Silicate was reported to decrease the corrosion rate of iron and is expected to become part of the corrosion products via sorption (Davis *et al.* 2001; McNeill and Edwards 2001). Rushing *et al.* (2003) noted that higher levels of silicate (50 mg/L as SiO_2) would release more iron into water, initially causing a red water problem but in the longer term would then be incorporated into a protective scale layer of the pipe, which might stop leaks. Silica has also been identified in the corrosion products of leaks in copper pipes (Lytle and Nadagouda 2010). In one field study where pits in copper pipes were remediated, the deposits were mainly composed of copper and silica with a small amount of calcium and magnesium (Scardina and Edwards 2008). Calcium silica hydrate and magnesium silica hydrate were also identified in sealed concrete cracks as facilitators of *in situ* remediation (Santhanam *et al.* 2002, 2003; Parks *et al.* 2010). Magnesium, calcium, and silica can therefore contribute to remediation of leaks in a wide range of circumstances. On the contrary, silicate can coat iron hydroxide particulates and create a highly negative surface charge, maintaining smaller colloids and preventing attachment in leaks (Davis *et al.* 2001).

Sulfate/chloride

Sulfate has been identified as aggressive to concrete and cause concrete degradation or expansion in concentrated sodium sulfate and magnesium sulfate solutions (Santhanam *et al.* 2002, 2003; Neville 2004). This trend might reduce *in situ* remediation potential of water by enhancing dissolution rates. Sulfate has mixed effects on iron corrosion depending on pH and flow conditions. One study showed that, at 35°C with pH close to 3 in sulfuric acid solution for 6 h, the weight loss of pure iron coupons increased with sodium sulfate concentrations (from 0.1 to 0.5 mol/L; i.e., from 0.1 to 0.5 M) but decreased thereafter (from 0.5 to 1 M sodium sulfate) (Trautenberg and Foley 1971). That study concluded that sulfate enhanced corrosion rate, but the extrapolation to results at circa neutral pH in potable water is unclear.

Chloride is expected to have a complex influence on *in situ* remediation. For copper, chloride tends to help seal the leaks in the longer term due to stifling reactions and possible formation of CuCl (Mattsson and Fredriksson 1968; Edwards *et al.* 1994b). For iron, one study indicated that chloride would complex with iron in the scale and increase ferrous iron concentration in water that could potentially help remediation by forming more solids (Elzenga *et al.* 1987; McNeill and Edwards 2001). However, Van Der Merwe (1988) showed that chloride had no effect on the weight loss of cast iron, reinforcing the possible complex impact of chloride on *in situ* remediation.

DO

DO is probably the dominant electron acceptor for corrosion in potable water (see cathode reactions in Table 1-1). This might lead to increased rates and concentrations of corrosion scale relative to situations with low DO. Perhaps more importantly, at low DO for iron, dissolved Fe²⁺ species will predominate relative to very insoluble Fe³⁺ species that tend to favor rust formation (AWWARF 1996; Sarin *et al.* 2004; Lytle and Nadagouda 2010). More precipitates will potentially form compared to metal ion leaching into water with increasing DO concentration, thereby reducing leaks. For example, approximately 18 mg/L less iron was leached into the stagnant water compared to the water without any DO (pH 6.2–6.8) in intact iron pipes in 2 h, and iron scales were observed outside pipe surface with the presence of 9.7 mg/L DO (Sarin *et al.* 2004). For copper pipes, little or no corrosion occurs in the absence of oxygen, so at very low DO copper pipe might behave as an inert material (Edwards *et al.* 1994a). Overall, DO could be an important parameter in *in situ* remediation of copper and iron pipes through participation in corrosion reactions but would not be expected to affect remediation of inert concrete and plastic pipes.

Water disinfectant

Free chlorine, monochloramine, and chlorine dioxide are commonly used for drinking water disinfection in the United States. Disinfectants could influence *in situ* remediation by (1) increasing corrosion rates (metal dissociation rate and scale formation rate), (2) promoting the formation of less soluble species, and (3) controlling or minimizing biofilms in water and on pipe surfaces.

By nature, these disinfectants can accelerate corrosion (e.g., copper and iron pipes) by redox reactions (LeChevallier *et al.* 1993; Frateur *et al.* 1999; Sarin *et al.* 2001; Hallam *et al.* 2002; Nguyen 2005; Zhang *et al.* 2008). Several studies have shown varied effects of chlorine and chloramine on corrosion rate in metallic pipes depending on water chemistry (Nguyen 2005; Zhang 2005). Sarver *et al.* (2011) indicated that high levels of chlorine could encourage copper pitting (non-uniform) corrosion, and several researchers (Pourbaix 1974; Lytle and Nadagouda 2010; Nguyen *et al.* 2010) have demonstrated that the concentration of ions within pits was dramatically different than in bulk water, which could affect the reactions contributing to or hindering self-remediation. For example, very low pH <4 within pits could hinder the likelihood of self-remediation after leaks begin, but very high levels of Cu(I) ions, which are easily oxidized to Cu(II) oxide scale, could enhance the likelihood of remediation. Sarver *et al.* (2011) found that chloramine did not contribute to pitting corrosion to the extent that chlorine did, but it nonetheless oxidized copper and contributed to scale formation (Nguyen *et al.* 2010).

These disinfectants can also transform corrosion products and form less soluble species, potentially promoting the chance of *in situ* remediation. Specifically, both iron and copper pipe scale products were shown to react with chlorine dioxide in a first-order rate (4.2×10^{-4} to $1.4 \times 10^{-4} \text{ s}^{-1}$ for iron and 8.7×10^{-5} to $1.0 \times 10^{-4} \text{ s}^{-1}$ for copper), which could oxidize the main scale components Fe_3O_4 or Cu_2O into less soluble ferric or cupric solids (Zhang *et al.* 2008). The ferrous iron, which was commonly found in corrosion products [e.g., FeCO_3 or $\text{Fe}(\text{OH})_2$], could also be oxidized by free chlorine, forming more insoluble Fe (III) oxides, such as Fe_2O_3 or FeOOH (Sarin *et al.* 2001). Free chlorine was also demonstrated to react with copper corrosion precipitates [e.g., Cu_2O or $\text{Cu}(\text{OH})_2$] to form more aged solids, such as tenorite (CuO) (Nguyen 2005).

Disinfectants can control or inhibit microbial activity and biofilms both in water and on pipe surfaces and therefore decrease microbially induced corrosion (MIC) and resulting leaks in pipelines (Hamilton 1985; LeChevallier *et al.* 1993; Edwards *et al.* 2000). However, this process is greatly affected by corrosion byproducts, which could provide a habitat for bacteria and decrease the efficiency of disinfection (Volk *et al.* 2000). In addition, the type of disinfectant would control biofilms, which might have their own propensity to induce leak repair via biofouling. Overall corrosion, disinfectant type, and microbial activity are expected to sometimes work in synergy and sometimes in opposition in relation to *in situ* remediation.

Turbidity

High turbidity is conventionally considered an indicator of poor water quality due to its indication of poor removal of waterborne microorganisms. However, in Roman pipelines in 15 BC, wood ashes were purposefully added to water to help *in situ* remediation of leaks by clogging (Pollio 15 BC as translated by Morgan 1960). Waterborne or water-formed particles have been acknowledged to physically block holes or cracks in concrete materials (Clear 1985; Hearn 1998; Ramm and Biscopig 1998; Edvardsen 1999; Neville 2002; Letterman *et al.* 2008). This indicates that some forms of turbidity could be helpful in *in situ* remediation.

Ionic strength

Ionic strength can affect the stability of waterborne and water-formed particles by compressing the electrical double layer and increasing the likelihood of attachment of particles to surfaces (Edzwald 2011). Thus, ionic strength could affect the type and size of particles present in water and the chance of *in situ* remediation via physical clogging of leak holes.

Ionic strength can also influence corrosion products and corrosion rates (Smart and Bockris 1992; Dunn *et al.* 2000). Higher ionic strength generally has a positive effect on the corrosion rate of metals. Dunn *et al.* (2000) indicated that the corrosion rate for iron at lower ionic strength (0.0028 M Cl^-) was, on average, 74.4% slower (2.1×10^{-4} vs. $8.2 \times 10^{-4} \text{ m/y}$) than that at higher ionic strength (0.028 M Cl^-). Moreover, corrosion products such as lepidocrocite ($\gamma\text{-FeOOH}$) and magnetite (Fe_3O_4) formed in low ionic strength (0.028 M Cl^-) solution versus the more soluble akaganeite ($\beta\text{-FeOOH}$) in high ionic strength (0.028

M Cl⁻) solution. In another study by Smart and Bockris (1992), the corrosion rate of iron linearly increased with increasing ionic strength. In a study by Zhang *et al.* (2002), the corrosion rate of copper increased when ionic strength increased from 0.005 to 0.01 M. The corrosion products [Cu₂O and Cu(OH)₂] were more soluble due to smaller copper ion activity at higher ionic strength. All these studies indicate that ionic strength could change the solubility and type of corrosion products, which could exert considerable influence on leak remediation efforts.

POSSIBLE LIMITATIONS OF *IN SITU* REMEDIATION APPROACHES TO SEAL WATER LEAKS

In situ remediation offers a suite of novel approaches to reduce or stop leaks in drinking water infrastructure, and certain limitations to their successes may be anticipated. First, it is impossible in practice to identify pipelines that have been remediated *in situ*, without removing and forensically investigating underground pipes. This means that real-world cases of remediation might remain undiscovered, reducing our appreciation for its overall importance in pipeline performance and distribution system longevity. Second, if the deposits do not accumulate in leaks, then *in situ* remediation in metallic and concrete pipes may not be possible. Third, it is uncertain if the materials sealing leaks are able to endure high pressures (140,000–1,400,000 Pa; i.e., 20–200 psi) commonly present in both water distribution systems and building plumbing systems. There is no knowledge regarding how long-lasting and durable the repairs can be, although the field studies suggest that remediation of copper leaks can persist for more than 10 years. Fourth, metallic corrosion, scaling, and particle formation in water pipelines, which could produce *in situ* remediation, might also cause high turbidity, colored water, and other consumer concerns. While these issues might limit the longevity and application of leak remediation in practice, the topic is highly deserving of future research given the attractive economics and public health benefits.

SUMMARY AND CONCLUSIONS

In situ leak remediation gives potentially transformative approaches with multiple mechanisms to reducing leaks and extending the lifetime of certain aging U.S. water distribution infrastructure. The phenomenon appears to be occurring in practice without our knowledge in at least some cases. Corrosion is correctly perceived as detrimental to the mechanical integrity of metallic water pipes. However, in at least some cases, corrosion may also provide a pathway to natural *in situ* remediation of some leaks. Two field studies provided practical evidence for the potential of relatively inert (aged concrete) and active (copper) pipes to seal leaks *in situ*, if appropriate and as yet incompletely identified repair mechanisms are enabled.

Three mechanisms are described for *in situ* remediation. These include corrosion for metallic pipes, physical clogging for all types of pipes, and precipitation for concrete pipes. Metallic corrosion can seal holes in pipelines with corrosion precipitates. The balance between the rate of hole growth via corrosion and hole shrinkage via scale formation, and the durability of the deposits, will determine the potential for success of remediation in

metallic (iron or copper) pipes. Suspended particles in water can also sometimes be beneficial in terms of *in situ* remediation as opposed to the conventional perception that they are invariably harmful. Physical clogging can be achieved by waterborne or water-formed particles [e.g., sand, bacteria, clay, rust, and $\text{Al}(\text{OH})_{3(s)}$], and its effectiveness is expected to depend on their size and surface charge. Cracks in concrete may also heal due to precipitation reactions, in which calcium carbonate, calcium silicate hydrate, or magnesium silicate hydrate can be the main materials remediating leaks.

The three mechanisms for *in situ* remediation are predicted to be highly dependent on both physical factors and water chemistry. The impact of individual physical or chemical parameters is complex and has not explicitly been studied previously in the context of water leak remediation, with the exception of cracked concrete under conditions not typically found in pressurized potable water pipelines. Additional research work and field observations will be needed to determine if these novel approaches can be applied in practice. Despite potential limitations, *in situ* leak remediation could possibly be used as an economically innovative way to sustain vulnerable drinking water infrastructure – an issue that has been deemed among the most urgent infrastructure problems facing the United States and is also significant around the world.

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CHAPTER 2. AUTOGENOUS METALLIC PIPE LEAK REPAIR IN POTABLE WATER SYSTEMS

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ABSTRACT

Copper and iron pipes have a remarkable capability for autogenous repair (self-repair) of leaks in potable water systems. Field studies revealed exemplars that metallic pipe leaks caused by nails, rocks, and erosion corrosion autogenously repaired, as confirmed in the laboratory experiments. This work demonstrated that 100% (N=26) of 150 μm leaks contacting representative bulk potable water in copper pipes sealed autogenously via formation of corrosion precipitates at 20–40 psi, pH 3.0–11.0, and with upward and downward leak orientations. Similar leaks in carbon steel pipes at 20 psi self-repaired at pH 5.5 and 8.5, but two leaks did not self-repair permanently at pH 11.0 suggesting that water chemistry may control the durability of materials that seal the leaks and therefore the permanence of repair. Larger 400 μm holes in copper pipes had much lower (0–33%) success of self-repair at pH 3.0–11.0, whereas all 400 μm holes in carbon steel pipes at 20 psi self-repaired at pH 4.0–11.0. Pressure tests indicated that some of the repairs created at 20–40 psi ambient pressure could withstand more than 100 psi without failure. Autogenous repair has implications for understanding patterns of pipe failures, extending the lifetime of decaying infrastructure, and developing new plumbing materials.

KEYWORDS: Autogenous repair; metallic pipe leaks; water chemistry; water pressure; physical parameters

INTRODUCTION

The worldwide problem of leaking and aging infrastructure has created a demand for improved plumbing materials and alternative approaches to conventional pipe repair, rehabilitation, and replacement. Leaks in water pipes cause up to half of unaccounted-for water loss, increasing water rates by billions of dollars in the United States (U.S.) alone and squandering precious resources.¹⁻⁴ Water pipes are typically comprised of cementitious, plastic, and iron materials in water mains and of copper, galvanized iron, and plastic materials in premise plumbing system.^{5,6} Recently plastics (including polyethylene chloride (PVC), chlorinated PVC, and cross-linked polyethylene (PEX)) have been more widely used in water mains and house plumbing systems due to its low cost of installation and corrosion resistance.^{7,8} But plastic systems are often joined with brass connectors, and metallic pipe materials are still dominant and include cast iron pipes that comprise approximately one-third of water mains and copper pipes which represent up to 90% of the building plumbing in some cities.⁹⁻¹² Many larger leaks start as small leaks in metallic pipes.¹² The unconventional and potentially transformative approach of autogenous repair, attempts to harness the natural ability of certain pipe materials to self-seal leaks, and might ultimately improve the performance and longevity of existing water distribution system infrastructure via manipulation of water chemistry and other factors.⁵ Autogenous repair of copper pinhole leaks was first reported in consumer homes in 2008, although anecdotally self-repair has long been relied on to stop leaks at new threaded joints in copper alloys.¹³

There are ultimately two pathways to leak initiation in water distribution systems that are of interest, termed type I and type II herein. Type I leaks are initiated as a result of nonuniform corrosion, in which “pit water” (i.e., low pH, low dissolved oxygen, and high conductivity) forms at localized anodic sites and consumes the pipe wall via “pinholes”, as is sometimes observed for copper and iron pipes.¹⁴⁻¹⁹ The initial small volume of acidic pit water is displaced after the wall is penetrated, and the metallic leak holes that are formed under type I conditions then contact the flowing bulk potable water. Type II leaks occur when the metallic leak hole is in contact with flowing bulk water at imperfectly sealed soldered or threaded joints, at external pipe wall penetrations caused by sharp objects such as nails and rocks, or at internal leaks resulting from cavitation or erosion corrosion (Figure 2-1, A-B).^{20,21} Two case studies in the U.S. and Japan suggested that leaks initiated by physical wall penetrations (i.e., not pitting corrosion) were responsible for around 30–40% of copper and iron pipe failures.^{12,21} Moreover, type I leaks might eventually behave like type II leaks after the initial small volume of acidic pore water is displaced.

Although corrosion is appropriately identified as a primary cause of metallic water pipe leaks,^{4,22} copper pipe leaks caused by pitting were reported to self-repair *in situ* via corrosion in the field (Figure A1).^{5,13,17} This fact changes perceptions about the factors controlling field performance of plumbing systems. For instance, a recent review hypothesized that some leaks in copper pipe might be stopped after they start (Figure 2-1), by a sealing process due to accumulation of corrosion rust in the leak hole.⁵ If this were experimentally confirmed for copper and iron pipes, which have long been industry standards for potable water, it could help explain the excellent historical performance of

these materials in practice, improve understanding of pipe failure patterns, and present new possibilities for extending the lifetime of existing infrastructure.

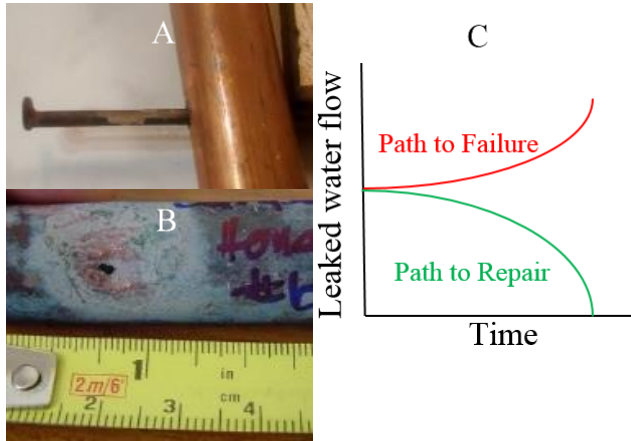


Figure 2-1. Type II leaks can be formed by physical penetration through water pipe wall by sharp objects such as nails (A) and rocks (B). In this case a fully penetrating iron nail through a copper pipe did not noticeably leak for 21 years after installation, and was only discovered during a home remodeling with no observable historical water damage (A). The conventional wisdom of initiated leaks, is that they will inevitably grow into larger leaks and eventually result in a failure (C-red line). However, this work reveals that in-situ autogenous repair can decrease leaked water flow rates to zero (C-green line).

This research was aimed at providing experimental verification of the self-repair ability of metallic iron and copper pipes, via natural reactions between the metal and the bulk water it carries, analogous to autogenous repair (i.e., self-repair) of concrete pipes known to occur in reactions between cracks in cement and the bulk water.²²⁻²⁷ The focus of this study was to document autogenous repair of leaks, in which the leak hole contacts flowing bulk water, as is the case for type II leaks or the latter phases of type I leaks. The strength of the repairs under typical pressure ranges was also examined, along with the role of water chemistry and physical parameters in controlling the likelihood of metallic pipe leak self-repair including the following factors:

Water pH. A wide water pH range of 3.0–11.0 was of interest, because the pH range of distributed bulk water is generally pH 6.5–8.5, but in extreme cases potable water can be as low as pH 4.5 or as high as pH 11.0.²⁸⁻³⁰ Moreover, the pH inside tubercles that initiate leaks in copper or iron was reported in the range of pH 3.0–5.5.^{14,17,18} Though self-repair with this type of water is not the focus of this research, such pHs reflect important boundary conditions worthy of study.

Water Pressure. Water main distribution systems are operated at relatively high pressure (>60 psi).³¹ Because these higher pressures can damage building plumbing, it has become commonplace to install a pressure reducing valve (PRV) before homes to provide consumers with an operating pressure of 20–80 psi (usually less than 50 psi).³¹⁻³³ In buildings, pressure is further reduced by ≈ 5 psi per floor and with water use.^{7,31} In this work, water pressure ranges of 20–40 psi were examined as representative of those found

in premise plumbing, and also because they are within the range of common laboratory pumps. Short-term pressure testing of each leak after complete repair was conducted at pressures up to 100 psi.

Other Parameters. It was hypothesized that other physical parameters including pipe wall thickness, leak size, pipe material, and leak orientation would influence the leak repair rate.⁵

MATERIALS AND METHODS

Experimental Apparatus At 20 Psi

A recirculation system (Figure A2, A) reproduced key features of a leaking potable water system. Water was pumped from a 21 liter reservoir at 0.3 L/min via a peristaltic pump through triplicate 2 in. long (50.8 mm) metallic pipes made from either copper or carbon steel (copper, 5.56 mm o.d. \times 0.36 mm wall thickness; carbon steel, 6.35 mm o.d. \times 0.89 mm wall thickness). Each pipe had a leak created in the middle of its length, by mechanically drilling a 150 or 400 μ m hole in the wall. These two sizes represent a moderate and large size leak for copper, considering the range typically observed in practice for residential copper pipes.¹³ To represent extremes of various leak orientations observed in practice, six copper pipe leaks were oriented upward in one test and all of the other leaks were oriented downward. Constant pressure of 20 ± 5 psi was maintained in the pipes using three 1 m long segments of flexible tubing (15.9 mm o.d. \times 6.35 mm i.d.) to dampen pump output pressure fluctuations, as monitored and confirmed with a pressure gauge (0–60 psi range). Total system pressure was adjusted via a nylon needle valve.

Experimental Apparatus At 40 Psi

To simulate leaking pipes at a higher pressure, a similar recirculation system was equipped with a 5-gallon pressure tank, a stainless steel pressure switch, and a chlorinated poly (vinyl chloride) (CPVC) check valve to maintain the target pressure at 40 ± 5 psi, as confirmed with a pressure gauge (0–100 psi range) (Figure A2, B). Water was pumped from a 19 L reservoir at 0.08 L/min via a chemical dosing pump through one 2 in. long (50.8 mm) copper pipe (type M 15.9 mm o.d. \times 13.8 mm i.d.) with a drilled 150 μ m hole in the center. The test was repeated 5 times, with a single copper pipe tested each time. All of the leak holes were oriented downward. The CPVC pipes and fittings were used to connect the system.

Water Chemistry

A baseline tap water representing typical pit water constituents was synthesized in nanopure water with 15 mg/L Cl^- from NaCl and pH levels ranging from 3.0 to 11.0 without disinfectant added.³⁴ Alkalinity was typically 30 mg/L alkalinity as CaCO_3 from NaHCO_3 as confirmed by EPA Method 310.^{1,35} but pH conditions below 5.5 and at 11.0 did not have added NaHCO_3 , because it is not possible to maintain alkalinity in an open system below pH 5.5 and the condition at pH 11.0 has alkalinity due to OH^- . The dissolved oxygen (DO) was in the range from 5.5 to 8.4 mg/L, as measured by a DO meter (model Oakton DO 110 series). The targeted solution pH was maintained to within ± 0.3 units adjusted with 0.2 M sodium hydroxide solution and 1 M nitric acid solution, as checked by a pH meter (model

Oakton pH 11 series). During pH adjustment, water flow was stopped and the reservoir was thoroughly mixed before turning the pump back on. The reservoir water was replaced with new synthetic tap water once a week.

Hole Leaking/Repair Rates

The fate of individual leaks was tracked by measuring the daily flow rate through the leak hole, as in previous studies tracking repair of leaks in cracked concrete.^{23-26,36,37} In concept if leaks are on a path to failure, the flow rate increases as the hole increases in diameter, whereas autogenous repair is evidenced by decreasing and ultimate cessation of flow rate (Figure 2-1, C). The relationship between leaked water flow, water pressure, and leak size has been described as³⁸⁻⁴¹

$$Q = C_d A (2gP)^{1/2} \quad (1)$$

where Q is the instantaneous leaked water flow rate from a leak hole with cross-sectional area A, C_d is the discharge coefficient, g is the acceleration rate due to gravity, and P is water pressure. The leak rate for each triplicate pipe was quantified by timing the instantaneous fill rate of an appropriately sized graduated cylinder once per day, and all reported measurements of flow rate were obtained at constant pressure of 20 or 40 psi unless stated otherwise. Leaked water passing through the holes was then circuited back into the reservoir. In general, experiments were run until it seemed clear that all triplicate leaks were destined for failure (Figure 2-1, C) or until all triplicate leaks remained sealed completely for at least 72 h. Since the drilled leaks were cylindrical holes in the pipe wall, the change of diameter over time (dL/dt ; unit, micrometers per day) was estimated using the preceding equation based on the measured Q and pressure (Calculation A1). While C_d can change somewhat as a function of Reynolds number,³⁹⁻⁴¹ it was assumed constant herein to provide illustrative calculations of patterns for leak repair (Table 2-1; Table A1).

Repaired Leak Characterization

In cases of complete repair, the corrosion rust/deposits that sealed the leaks were photographed and analyzed for elemental composition via environmental scanning electron microscope with an attached X-ray energy dispersive system (ESEM/EDS).

Pressure Test To Assess Strength of Repair

Each repaired pipe, filled with water and capped closed in one end, was first pumped with nitrogen gas and then put in a tank with water (Figure A2, C). All of the repaired holes, except for those sent for ESEM/EDS and those failing to self-repair, were tested by increasing the pressure in 5 psi increments from 25 psi up to 100 psi. Each tested pressure on repaired pipes lasted approximately 1 min. The maximum pressure the repaired hole could withstand was defined by the point where there was water leaking from the repaired hole.

RESULTS AND DISCUSSION

Autogenous Repair Of Copper Pipe

Autogenous repair of 150 μm predrilled holes in copper pipes was successfully reproduced in the laboratory (Figure 2-2; Table A2).

Repair of leaks facing downward at 20 psi

At pH 11.0 the leaked water flow rate from the triplicate holes dropped to zero evidencing self-repair after 1, 2, and 6 days (on average 3 days). The precise time to seal the hole varied for each replicate, demonstrating a high degree of variability in the autogenous repair process. For instance, one replicate leak rate reduced to zero after a day, one remained steady in the first day but reduced to zero the second day, whereas the third replicate had fluctuating rates for the first 4 days and then reduced to zero (Figure 2-2, A). Even more variability was observed at pH 3.0, where the three holes were repaired after 1, 5, and 14 days (on average 7 days). To increase the statistical power of the approach for one test condition, nine leaks were tested at pH 8.5 and 100% were autogenously repaired from 1 to 34 days (on average 18 days) (Figure 2-2, C). However, due to the high variability of the replicates, the length of time to complete repair was not statistically different at pH 8.5, pH 11.0, and pH 3.0 in these experiments (one-way Anova test; $p > 0.05$). The high variability was also reflected in the calculated leak diameter change rate (dL/dt). For example, the dL/dt for each hole varied from -4.4 to $-150 \mu\text{m}/\text{day}$ at pH 8.5 (Table 2-1). Overall, this experiment provides clear laboratory confirmation of field observations in which copper leaks were hypothesized to self-repair by contacting the bulk tap water (Figure 2-1).⁵ For each repaired pipe leak in the field work and in the laboratory work, there were blue-green corrosion deposits both inside and outside the hole, as revealed by naked eye and by ESEM/EDS (Figure 2-2, B).^{5,13} The composition of the deposits clogging the hole in Figure 2-2, B was approximately 31% copper, 32% carbon, and 37% oxygen by weight, consistent with a typical mixture of copper oxides and carbonates that naturally form on fresh copper pipe in many potable water systems.

Repair at different leak orientations at 20 psi

The average time to autogenous repair of 150 μm holes facing upward was longer compared to holes facing downward at pH 8.5 (Figure 2-2, C). All six upward holes were self-repaired from 2 to 116 days (on average 43 days), and the average calculated diameter reduction rate (average $dL/dt = -16.6 \mu\text{m}/\text{day}$) was about half of the rate for holes facing downward (average $dL/dt = -33.5 \mu\text{m}/\text{day}$). However, again due to the high variability in the repair process for each replicate, the time to complete repair was not statistically different for holes facing upward and downward at pH 8.5 (Wilcoxon test, $p > 0.05$).

Repair At Higher Pressure

All ($N = 5$) of the 150 μm holes at 40 psi were self-repaired from 5 to 87 days (on average 46 days). This was on average about 3 times longer than the time to repair holes at 20 psi (Figure 2-2, C), but the time difference was not statistically significant (Wilcoxon test, $p > 0.05$).

Table 2-1. Calculated leak diameter change rate (dL/dt) on average in each condition (Negative Number = Diameter of holes decreases over time; Positive Number = Diameter of holes increases over time).

dL/dt ($\mu\text{m}/\text{day}$) for holes facing downward @ 20 psi				
Condition	Temporal Average Pipe 1	Temporal Average Pipe 2	Temporal Average Pipe 3	Replicate Temporal Average N=3 or N=9
Cu pH 3.0 150 μm	-11.6	-30.6	-150.0	-64.1
Cu pH 8.5 150 μm	-150.0	-40.0	-75.0	-33.5
Cu pH 8.5 150 μm	-4.8	-5.4	-5.4	
Cu pH 8.5 150 μm	-4.4	-8.3	-8.3	
Cu pH 11.0 150 μm	-19.9	-75.0	-150.0	-81.6
Cu pH 3.0 400 μm	44.0	6.0	7.0	19.0
Cu pH 8.5 400 μm	-6.5	-0.2	-0.7	-2.5
Cu pH 11.0 400 μm	-0.4	1.0	-0.3	0.1
Fe pH 5.5 150 μm	-150.0	-150.0	-150.0	-150.0
Fe pH 8.5 150 μm	-300.0	-60.0	-300.0	-220.0
Fe pH 11.0 150 μm	-3.0	-8.0	-60.0	-23.7
Fe pH 3.0 400 μm *	10.0	-15.0	-11.0	-5.3
Fe pH 4.0 400 μm	-36.1	-15.1	-35.9	-29.0
Fe pH 8.5 400 μm	-8.2	-19.1	-28.7	-18.7
Fe pH 11.0 400 μm	-57.9	-0.9	-66.7	-41.8
dL/dt ($\mu\text{m}/\text{day}$) for holes facing downward @ 40 psi				
Cu pH 8.5 150 μm	-3.2	-	-	N=5 -9.5
Cu pH 8.5 150 μm	-2.0	-	-	
Cu pH 8.5 150 μm	-30.0	-	-	
Cu pH 8.5 150 μm	-3.3	-	-	
Cu pH 8.5 150 μm	-8.9	-	-	
dL/dt ($\mu\text{m}/\text{day}$) for holes facing upward @ 20 psi				
Cu pH 8.5 150 μm	-75	-1.3	-1.3	N=6
Cu pH 8.5 150 μm	-3	-9.4	-9.4	-16.6

Note that the time frame for each calculation was to the end of self-repair or experiment (Table A2).

*Based on day 0 to day 46 since the pressure stayed at zero after day 46.

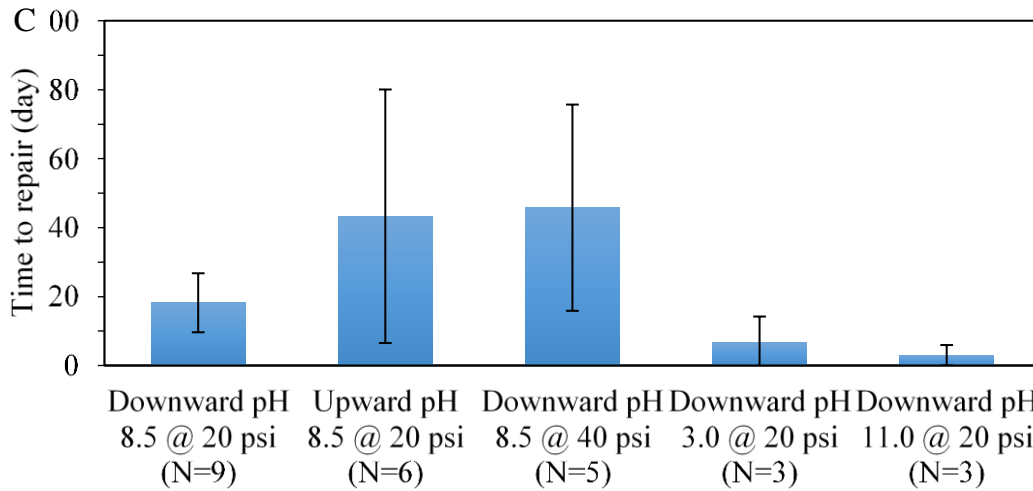
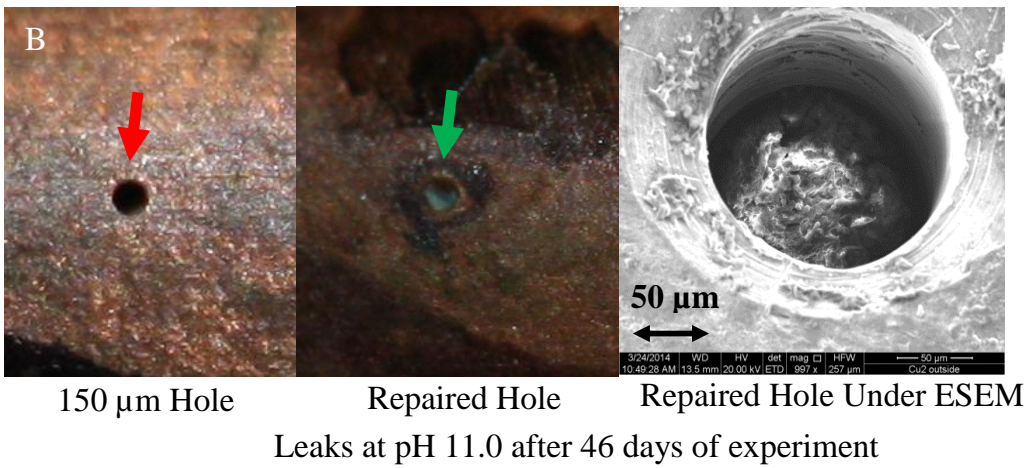
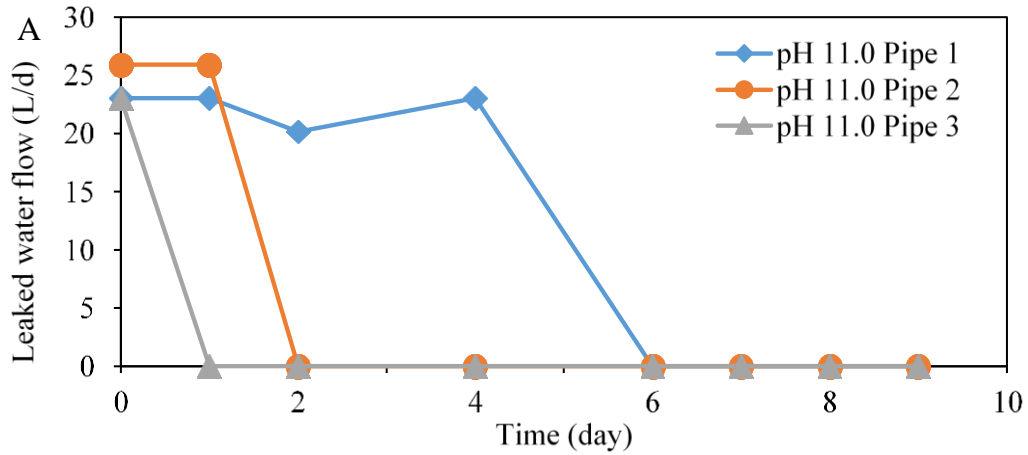


Figure 2-2. In-situ autogenous repair of 150 µm holes in copper pipes with two leak orientations in potable water at two pressures. The leaked water flows were reduced to zero at pH 11.0 (A), thereby resulting in repair of leaks (B). The error bar shows a 95% confidence interval (C).

Autogenous Repair Of Carbon Steel Pipe

Autogenous repair of 150 μm holes in carbon steel pipes was relatively rapid over the range of pH 5.5 to pH 11.0 (Figure 2-3, A; Table A2). It took only 1 day and 2.5 days to repair all of the triplicate holes at pH 5.5 and 8.5, respectively. However, the calculated average diameter reduction rate of holes at pH 8.5 (average $dL/dt = -220.0 \mu\text{m}/\text{day}$) was higher than that at pH 5.5 (average $dL/dt = -150 \mu\text{m}/\text{day}$), since two of the three holes at pH 8.5 took only 0.5 day to repair (Table 2-1).

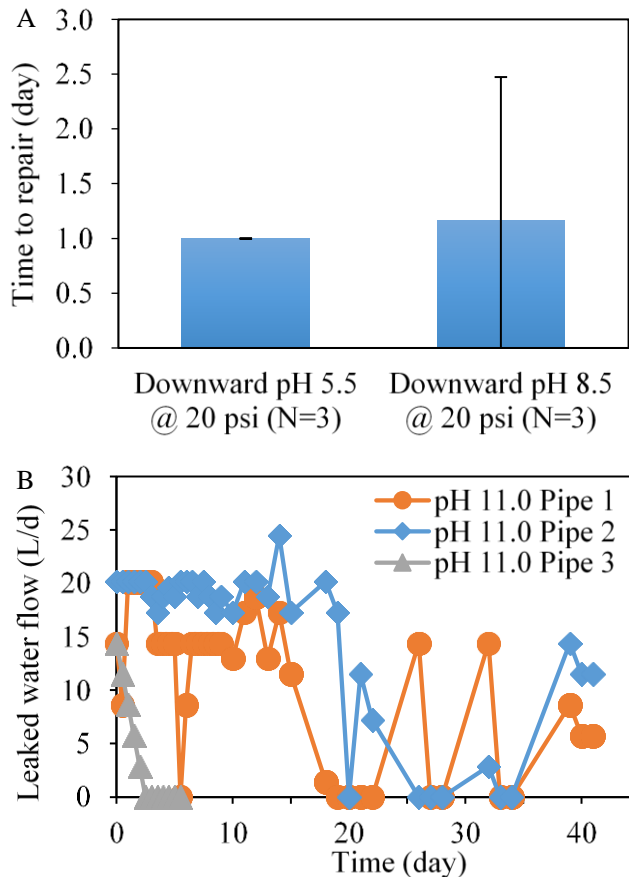
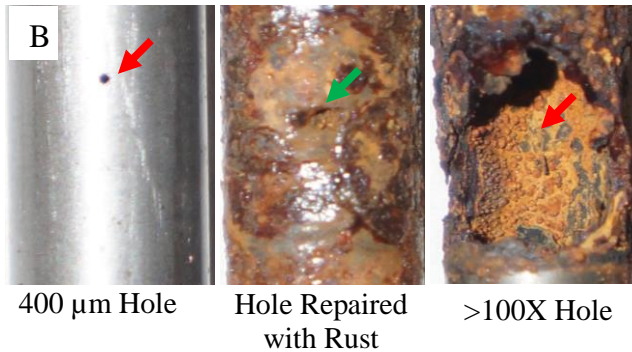
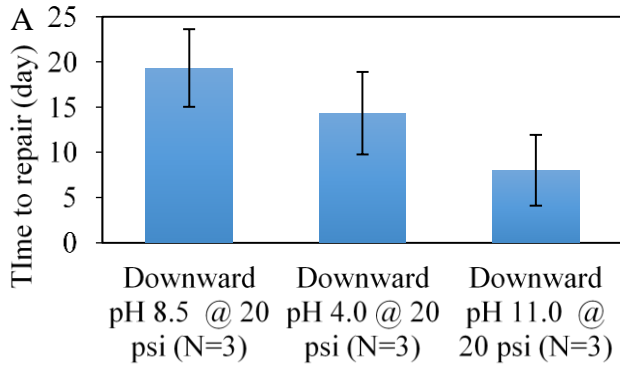


Figure 2-3. In-situ autogenous repair of 150 μm holes facing downward in carbon steel pipes in potable water at 20 psi. The error bar shows a 95% confidence interval (A). Two holes failed to self-repair at pH 11.0 due to impermanent nature of the sealing materials in leak-holes (B).

At pH 11.0 a relatively unusual result was obtained. Leaked water flow of one hole reduced to zero after only 2.5 days and stayed at zero (Figure 2-3, B). The self-repair of the second and third leak hole followed a pattern of healing and reopening. Relative to the 150 μm holes in copper pipes that had self-repaired, the leaked water flow fluctuated dramatically and the repair was transient. The repaired leak hole was covered with rust comprised of 57% iron, 29% oxygen, and 14% carbon by weight under ESEM/EDS, consistent with iron corrosion products on the surface of the pipe (Figure A3).⁴² The results suggest that the temporary nature of the two repaired leaks at this high pH was due to poor durability of the pipe scale clogging the leak.

Repair Of Larger Hole Sizes

There was a mixed result for autogenous repair of 400 μm holes in copper pipes (Figure A4; Table A2). In acidic pH 3.0 or basic pH 11.0 conditions, there was no detectable autogenous repair after 4 months (Figure A4, A). Indeed, the holes grew to 25% larger than the original diameter at pH 3.0, and the target pressure of 20 psi could no longer be maintained due to the limited capacity of the pump, demonstrating that this particular hole was destined to permanent failure and permanent leaking (Figure A4, B). At pH 8.5, one of the replicates self-repaired after 1 month (average $dL/dt = -6.5 \mu\text{m}/\text{day}$) and the other two stayed at about the same size (average $dL/dt = -0.2$ and $-0.7 \mu\text{m}/\text{day}$, respectively) (Figure A4, C).



Holes at pH 3.0 after 90 days of experiment

Figure 2-4. In-situ autogenous repair of 400 μm holes facing downward in carbon steel pipes in potable water at 20 psi. The error bar shows a 95% confidence interval (A). One of the three 400 μm holes grew to more than 100 X larger at pH 3.0, resulting in increased leaked water flow and therefore path of failure (B).

For the larger 400 μm holes in carbon steel pipes, the triplicate holes were all repaired after 12 days at pH 11.0 (on average 8 days) and after 19 days at pH 4.0 (average 14 days) (Figure 2-4, A). However, in this case the pressure at pH 4.0 could not be held steady at 20 psi due to the large initial increase in hole size. It dropped to 7 psi at day 6 but it rose back to 20 psi at day 8, after which time the leak hole began to close and one of the triplicate holes was repaired. As for the holes at pH 8.5, they were all repaired after 22 days (on average 19 days) and had a relatively slower rate of leak closure (average $dL/dt = -18.7 \mu\text{m}/\text{day}$) compared to that at pH 4.0 (average $dL/dt = -29 \mu\text{m}/\text{day}$) and pH 11.0 (average

$dL/dt = -41.8 \mu\text{m/day}$) (Figure 2-4, A; Table 2-1). Examining the calculated leak diameter at 0, 10, 20, and 22 days at pH 8.5, the change rate in the hole diameter (dL/dt) was constant during this experiment at $\sim -16.5 \mu\text{m/day}$ for the first 10 days, $\sim -16.2 \mu\text{m/day}$ for 10–20 days and $\sim -14.9 \mu\text{m/day}$ for the last 2 days. There was also a highly variable result for replicates at pH 3.0, in that two of the three holes were repaired with rust after 36 days and remained sealed for the duration of the test (3 months); however, the third replicate at this condition grew to about 100 times as big as the original hole and was therefore clearly destined to failure (Figure 2-4, B; Figure A5).

Strength Of Repaired Holes

The autogenously repaired leak holes were relatively resilient and durable (Figure 2-5; Table A3). For copper pipe leaks repaired at 20 psi, 25% could withstand 85 psi and 75% stayed repaired at 35 psi. About 5% could withstand more than 100 psi. However, this result may be a lower bound to the actual strength of the holes, since they were dried from the time of repair to the time of testing and there was disturbance to the repaired hole when the apparatus was taken apart, which might have lowered their strength as applied to other tests discussed later. For copper pipe leaks repaired at higher pressure (40 psi), one of five withstood 100 psi without failure. For iron pipes repaired at 20 psi, more than 70% could withstand more than 100 psi without leaking. The later point may be important, since iron pipes are often used in portions of potable water distribution systems with higher pressures, as well as in premise plumbing, and iron might have a higher capacity for self-repair at these higher pressures.

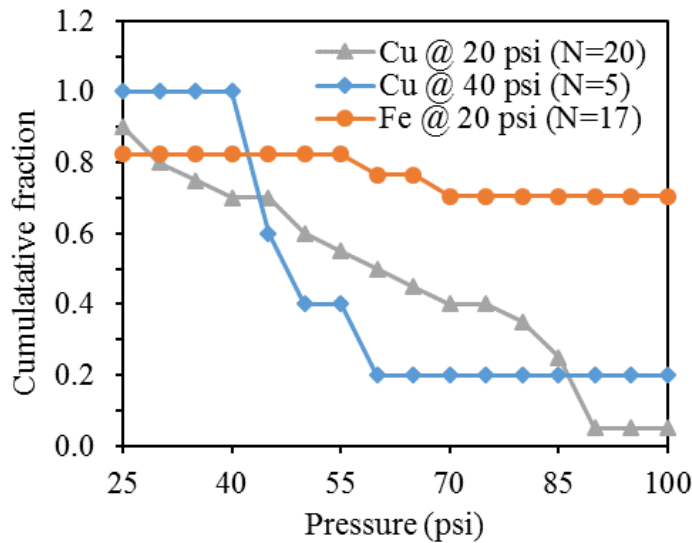


Figure 2-5. Cumulative fraction of the repaired leak-holes that could withstand the tested pressure.

Discussion

The work herein documents a remarkable ability of traditional copper and iron plumbing materials to self-repair or self-heal leaks in the pH range of 3.0–11.0 and ambient pressures of 20–40 psi. Trends in the data support the hypothesis that the likelihood of autogenous leak repair in metallic copper and steel pipes is a function of water pH, leak size, pipe

material, and water pressure among other factors.⁵ At a constant 20 psi pressure and 0.36 mm wall thickness herein, all 150 μm holes facing upward and downward in copper pipes self-healed, suggesting that leak orientation does not strongly influence the leak repair rate at low water pressure systems (< 20 psi). But much lower repair rates of 0–33% occurred at pH 3.0–11.0 with 400 μm holes, indicating that there may be a threshold leak size above which repair is not completely effective or may be destined to fail, similar to prior experience with autogenous repair of cracks in concrete.^{23-24,37}

The 150 μm holes in carbon steel pipes were able to self-repair under all tested conditions (from pH 5.5 to 11.0); however, the repair of holes at pH 11.0 was transient with a cycle of leaking and sealing repeating numerous times. Thus, the durability of the compounds repairing the holes, which is likely influenced by water chemistry, will be an important issue in terms of obtaining semipermanent or permanent repairs.

The enhanced speed of repair for equally sized iron versus copper holes revealed in this work ($\approx 18\text{X}$ faster for 150 μm ; $>5\text{X}$ faster for 400 μm) might reflect the $\approx 10\text{X}$ higher rate of corrosion (and scale formation) typically observed for iron versus copper in potable water systems.^{16,43} Thus, just as higher corrosion rates can increase the rate of failure, they can also increase the likelihood of self-repair in at least some circumstances. Statistically the self-repaired iron pipes were also capable of withstanding higher pressures on average than copper pipes (Wilcoxon test, $p < 0.05$). This difference could be due to the thicker pipe wall of carbon steel ($=0.89$ mm) versus copper ($=0.36$ mm) pipe in this work, or the greater strength of the corrosion deposits for carbon steel, or the more complete coverage by these deposits throughout the depth of the leak observed visually and verified by ESEM/EDS.

This work demonstrates that autogenous metallic pipe leak repair can sometimes reduce or stop water pipe leaks. However, further study is needed to examine a wider range of the physical (e.g., leak size, water pressure, and pipe wall thickness) and chemical (e.g., corrosion inhibitors, disinfectants, and DO) impacts on the effectiveness of autogenous repair. At present it is clear that this process is occurring naturally in some systems, but the extent to which the process can be manipulated to extend the lifetime of water mains and premise plumbing remains to be seen.

ACKNOWLEDGEMENTS

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APPENDIX A

Supporting Information

Autogenous Metallic Pipe Leak Repair In Potable Water Systems

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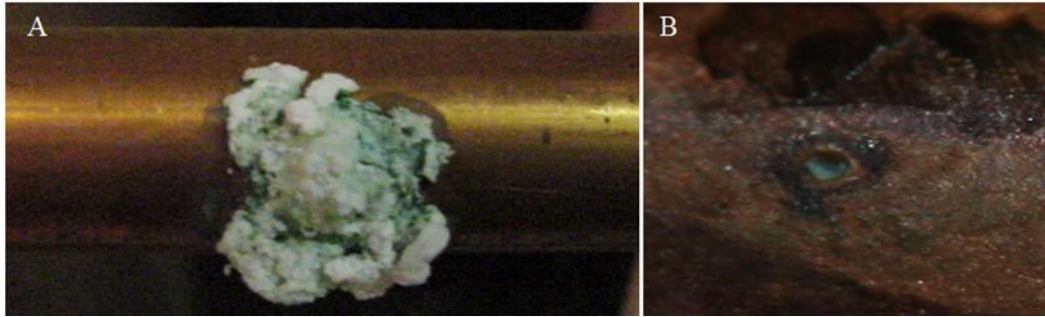


Figure A1. Autogenous repair of leaks in field study (A) (Adapted with permission of Scardina *et al.* 2008) was reproduced in the laboratory (B).

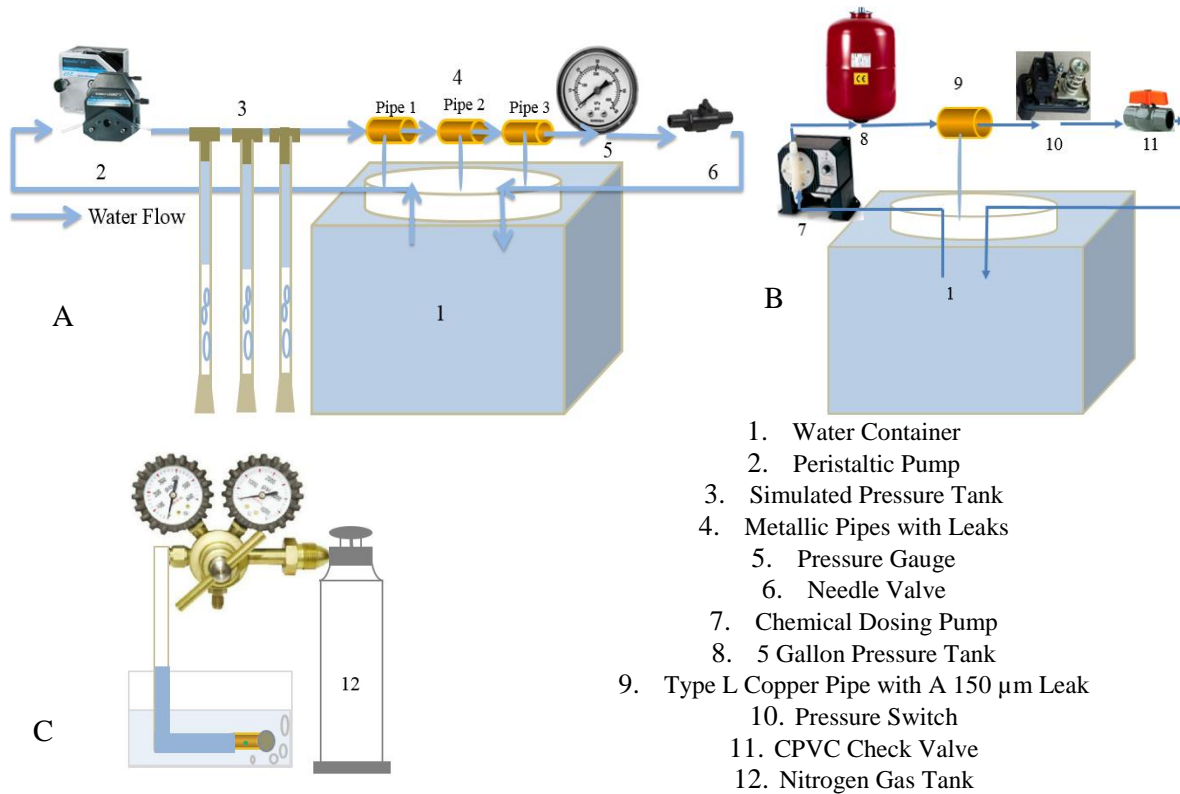


Figure A2. Overview of the recirculation apparatus operated at 20 psi (A) and 40 psi (B). Pressure test was conducted to find the maximum pressure the repaired hole could withstand before leaking (C).



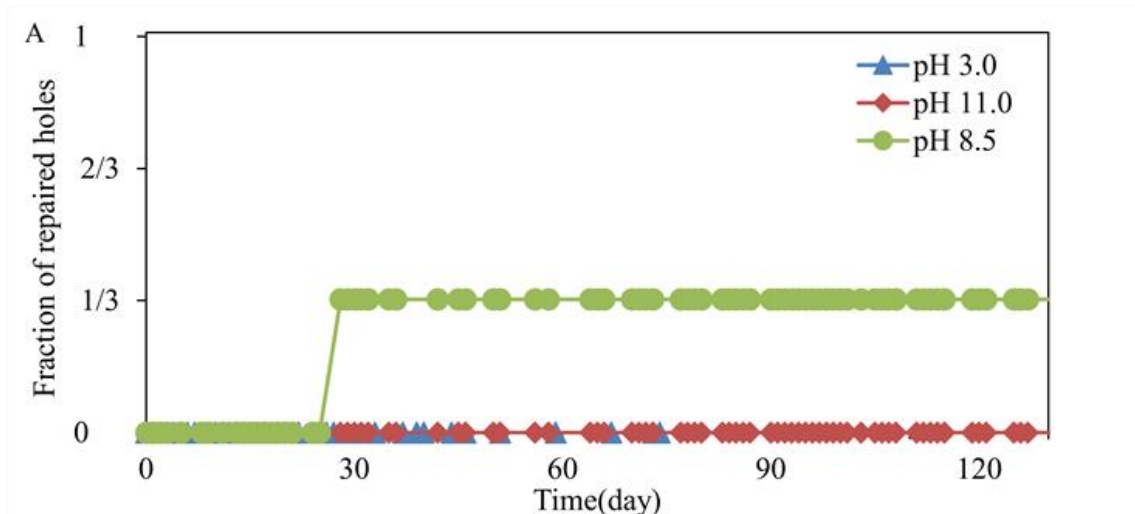
150 μm Hole

Repaired Hole
Covered with Rust

Repaired Hole Under ESEM

Leaks at pH 11.0 after 41 days of experiment

Figure A3. Two 150 μm holes in carbon steel pipes in potable water at 20 psi failed to self-repair at pH 11.0 (left), with the third hole repaired with rust (middle and right).



400 μm Hole

500 μm Hole

Holes at pH 3.0 after 141 days of experiment



400 μm Hole

Repaired Hole

Holes at pH 8.5 after 141 days of experiment

Figure A4. In-situ autogenous repair of 400 μm holes facing downward in copper pipes in potable water at 20 psi. The fraction of holes to heal was based on the triplicate holes in each condition.

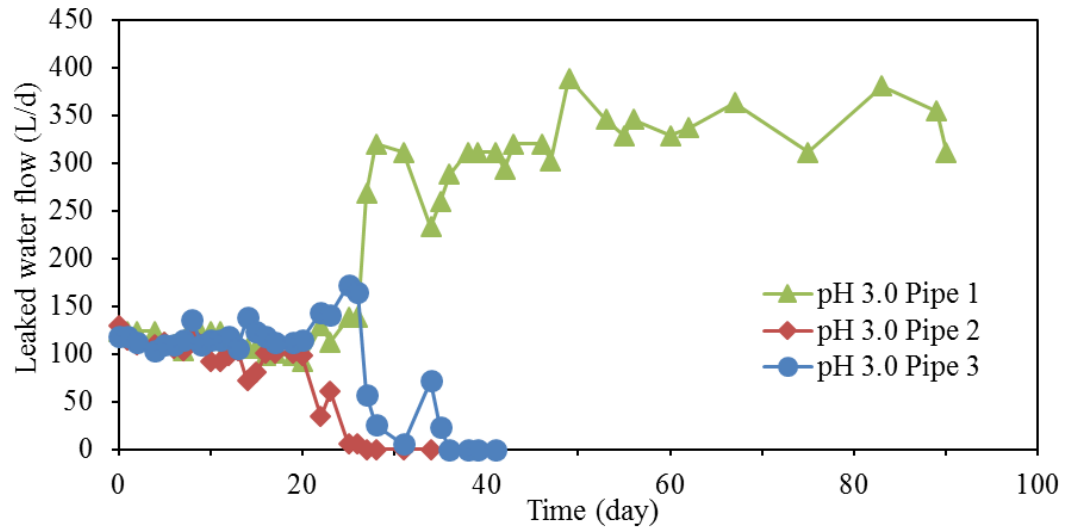


Figure A5. Leaked water flow of 400 μm holes facing downward in carbon steel pipes at pH 3.0 in potable water at 20 psi.

Table A1. Representative calculation of diameter change rate over time for 400 μm holes in copper pipes at pH 8.5.

Time (day)	Leaked Water Flow (L/d)				Pressure (psi)	Pipe 1		Pipe 2		Pipe 3	
	Pipe 1	Pipe 2	Pipe 3	average		Constant C=	0.00017	Constant C=	0.00016	Constant C=	0.00017
						Leak Diameter L (μm)	dL/dt ($\mu\text{m}/\text{d}$)	Leak Diameter L (μm)	dL/dt ($\mu\text{m}/\text{d}$)	Leak Diameter L (μm)	dL/dt ($\mu\text{m}/\text{d}$)
0.0	135.4	126.7	135.4	132.5	23.5	400.0	-	400.0	-	400.0	-
1.0	123.8	121.0	126.7	123.8	17.0	414.9	14.9	423.8	23.8	419.7	19.7
2.0	144.0	144.0	138.2	142.1	19.0	435.1	20.2	449.7	25.9	426.3	6.6
3.0	149.8	144.0	149.8	147.8	20.0	438.0	3.0	443.9	-5.7	438.0	11.8
5.0	138.2	135.4	158.4	144.0	19.0	426.3	-5.9	436.0	-4.0	456.3	9.1
8.0	132.5	123.8	129.6	128.6	18.0	423.0	-1.1	422.7	-4.4	418.4	-12.6
9.0	135.4	126.7	135.4	132.5	19.5	419.1	-3.9	419.1	-3.6	419.1	0.7
11.0	135.4	126.7	138.2	133.4	20.0	416.5	-1.3	416.5	-1.3	420.9	0.9
13.0	138.2	129.6	135.4	134.4	19.5	423.5	3.5	423.8	3.7	419.1	-0.9
14.0	132.5	126.7	132.5	130.6	18.5	420.1	-3.4	424.7	0.8	420.1	1.0
15.0	132.5	123.8	129.6	128.6	20.0	412.0	-8.1	411.7	-13.0	407.5	-12.6
16.0	138.2	129.6	138.2	135.4	21.0	415.8	3.8	416.1	4.4	415.8	8.3
17.0	126.7	121.0	132.5	126.7	18.0	413.7	-2.1	417.7	1.7	423.0	7.2
18.0	144.0	135.4	144.0	141.1	19.0	435.1	21.4	436.0	18.2	435.1	12.1
19.0	141.1	129.6	144.0	138.2	19.5	427.9	-7.2	423.8	-12.1	432.3	-2.8
20.0	144.0	132.5	144.0	140.2	20.0	429.5	1.6	425.8	2.0	429.5	-2.7
21.0	132.5	132.5	141.1	135.4	19.0	417.3	-12.2	431.3	5.5	430.7	1.2
24.0	138.2	132.5	146.9	139.2	20.0	420.9	1.2	425.8	-1.8	433.8	1.0
25.0	132.5	129.6	138.2	133.4	20.5	409.5	-11.4	418.6	-7.2	418.3	-15.5
28.0	0.0	135.4	149.8	95.0	20.0	0.0	-136.5	430.4	3.9	438.0	6.6
29.0	0.0	135.4	141.1	92.2	20.0	0.0	0.0	430.4	0.0	425.2	-12.8
30.0	0.0	123.8	138.2	87.4	20.0	0.0	0.0	411.7	-18.7	420.9	-4.4
31.0	0.0	141.1	152.6	97.9	21.0	0.0	0.0	434.2	22.5	436.9	16.0
32.0	0.0	129.6	146.9	92.2	20.0	0.0	0.0	421.2	-13.0	433.8	-3.1
35.0	0.0	138.2	172.8	103.7	20.0	0.0	0.0	435.0	4.6	470.5	12.2
36.0	0.0	135.4	144.0	93.1	20.0	0.0	0.0	430.4	-4.6	429.5	-41.0
42.0	0.0	138.2	146.9	95.0	20.0	0.0	0.0	435.0	0.8	433.8	0.7
45.0	0.0	126.7	141.1	89.3	20.0	0.0	0.0	416.5	-6.2	425.2	-2.9
46.0	0.0	135.4	141.1	92.2	20.0	0.0	0.0	430.4	14.0	425.2	0.0

50.0	0.0	146.9	146.9	97.9	20.0	0.0	0.0	448.4	4.5	433.8	2.1
51.0	0.0	135.4	135.4	90.2	20.5	0.0	0.0	427.8	-20.6	413.9	-19.9
56.0	0.0	135.4	149.8	95.0	20.5	0.0	0.0	427.8	0.0	435.4	4.3
58.0	0.0	135.4	138.2	91.2	19.5	0.0	0.0	433.2	2.7	423.5	-5.9
64.0	0.0	138.2	149.8	96.0	20.0	0.0	0.0	435.0	0.3	438.0	2.4
65.0	0.0	135.4	146.9	94.1	20.0	0.0	0.0	430.4	-4.6	433.8	-4.2
66.0	0.0	138.2	149.8	96.0	20.0	0.0	0.0	435.0	4.6	438.0	4.2
70.0	0.0	141.1	146.9	96.0	20.0	0.0	0.0	439.5	1.1	433.8	-1.1
71.0	0.0	141.1	144.0	95.0	20.0	0.0	0.0	439.5	0.0	429.5	-4.3
72.0	0.0	146.9	144.0	97.0	23.5	0.0	0.0	430.6	-8.8	412.6	-17.0
73.0	0.0	141.1	146.9	96.0	22.5	0.0	0.0	426.7	-3.9	421.2	8.7
77.0	0.0	138.2	141.1	93.1	19.5	0.0	0.0	437.7	2.8	427.9	1.7
78.0	0.0	135.4	138.2	91.2	21.5	0.0	0.0	422.7	-15.0	413.3	-14.6
79.0	0.0	123.8	132.5	85.4	20.5	0.0	0.0	409.2	-13.5	409.5	-3.9
80.0	0.0	132.5	135.4	89.3	21.0	0.0	0.0	420.7	11.5	411.4	1.9
83.0	0.0	138.2	144.0	94.1	19.0	0.0	0.0	440.6	6.6	435.1	7.9
84.0	0.0	141.1	141.1	94.1	20.0	0.0	0.0	439.5	-1.1	425.2	-9.9
85.0	0.0	126.7	132.5	86.4	21.0	0.0	0.0	411.4	-28.1	407.0	-18.2
86.0	0.0	132.5	135.4	89.3	20.5	0.0	0.0	423.2	11.8	413.9	6.9
87.0	0.0	132.5	135.4	89.3	19.5	0.0	0.0	428.5	5.3	419.1	5.2
90.0	0.0	129.6	135.4	88.3	20.0	0.0	0.0	421.2	-2.5	416.5	-0.9
91.0	0.0	121.0	135.4	85.4	20.5	0.0	0.0	404.4	-16.8	413.9	-2.6
92.0	0.0	121.0	126.7	82.6	19.5	0.0	0.0	409.5	5.1	405.5	-8.4
93.0	0.0	129.6	132.5	87.4	20.5	0.0	0.0	418.6	9.1	409.5	4.0
94.0	0.0	129.6	132.5	87.4	19.5	0.0	0.0	423.8	5.3	414.6	5.2
95.0	0.0	138.2	138.2	92.2	20.0	0.0	0.0	435.0	11.1	420.9	6.2
96.0	0.0	126.7	138.2	88.3	20.0	0.0	0.0	416.5	-18.5	420.9	0.0
97.0	0.0	129.6	135.4	88.3	20.0	0.0	0.0	421.2	4.7	416.5	-4.4
98.0	0.0	126.7	132.5	86.4	20.0	0.0	0.0	416.5	-4.7	412.0	-4.5
99.0	0.0	136.8	148.3	95.0	21.5	0.0	0.0	424.9	8.5	428.1	16.1
100.0	0.0	123.8	135.4	86.4	21.0	0.0	0.0	406.7	-18.2	411.4	-16.7
101.0	0.0	138.2	144.0	94.1	20.0	0.0	0.0	435.0	28.3	429.5	18.1
103.0	0.0	123.8	135.4	86.4	20.0	0.0	0.0	411.7	-11.6	416.5	-6.5
105.0	0.0	126.7	129.6	85.4	19.5	0.0	0.0	419.1	3.7	410.1	-3.2
106.0	0.0	126.7	135.4	87.4	20.5	0.0	0.0	413.9	-5.2	413.9	3.8
107.0	0.0	126.7	129.6	85.4	29.5	0.0	0.0	377.9	-36.0	369.8	-44.1

108.0	0.0	129.6	123.8	84.5	19.5	0.0	0.0	423.8	45.9	400.9	31.1
111.0	0.0	129.6	136.8	88.8	19.5	0.0	0.0	423.8	0.0	421.3	6.8
112.0	0.0	132.5	135.4	89.3	21.0	0.0	0.0	420.7	-3.2	411.4	-9.9
113.0	0.0	129.6	138.2	89.3	18.5	0.0	0.0	429.5	8.8	429.1	17.7
114.0	0.0	123.8	132.5	85.4	20.0	0.0	0.0	411.7	-17.8	412.0	-17.1
115.0	0.0	135.4	141.1	92.2	20.0	0.0	0.0	430.4	18.7	425.2	13.2
119.0	0.0	126.7	126.7	84.5	20.0	0.0	0.0	416.5	-3.5	402.9	-5.6
120.0	0.0	132.5	135.4	89.3	21.0	0.0	0.0	420.7	4.2	411.4	8.5
121.0	0.0	123.8	126.7	83.5	19.5	0.0	0.0	414.3	-6.3	405.5	-5.9
125.0	0.0	129.6	141.1	90.2	19.5	0.0	0.0	423.8	2.4	427.9	5.6
126.0	0.0	138.2	144.0	94.1	19.0	0.0	0.0	440.6	16.8	435.1	7.2
127.0	0.0	129.6	138.2	89.3	20.0	0.0	0.0	421.2	-19.4	420.9	-14.2
132.0	0.0	135.4	141.1	92.2	20.0	0.0	0.0	430.4	1.9	425.2	0.9
133.0	0.0	126.7	126.7	84.5	20.0	0.0	0.0	416.5	-14.0	402.9	-22.3
134.0	0.0	129.6	129.6	86.4	20.0	0.0	0.0	421.2	4.7	407.5	4.6
135.0	0.0	135.4	135.4	90.2	20.0	0.0	0.0	430.4	9.3	416.5	9.0
136.0	0.0	138.2	144.0	94.1	20.0	0.0	0.0	435.0	4.6	429.5	13.1
139.0	0.0	138.2	138.2	92.2	19.5	0.0	0.0	437.7	0.9	423.5	-2.0
140.0	0.0	129.6	129.6	86.4	20.0	0.0	0.0	421.2	-16.6	407.5	-16.0
141.0	0.0	123.8	132.5	85.4	20.0	0.0	0.0	411.7	-9.5	412.0	4.5
						Sum	-123.5		-18.4		-58.6
						Average	-6.5		-0.2		-0.7

Calculation A1. Example of dL/dt calculation with listed assumptions

Assumptions

- 1) C_d is assumed constant in each pipe experiment
- 2) The cylindrical hole has initial diameter of 400 μm and is repaired uniformly

$$Q = C_d \times A \times (2gP)^{1/2}$$

$$A = \pi r^2$$

$$Q = C_d \times \pi L^2 \times (2gP)^{1/2} / 4$$

$$\text{Assume Constant } C = C_d \times \pi \times (2g)^{1/2} / 4$$

Thus $Q = C \times L^2 \times P^{1/2}$ among which L is the leak diameter in μm , r is the leak radius in μm , P is in psi and Q is in L/d

Take Pipe 1 as an example:

$$C = \frac{Q_0}{L^2 \times P^{1/2}} = \frac{135.4}{400 \times 400 \times 23.5^{1/2}} = 0.00017$$

$$\text{At day 0: } L = \left(\frac{Q_0}{C \times P^{1/2}} \right)^{1/2} = \left(\frac{135.4}{0.00017 \times 23.5^{1/2}} \right)^{1/2} = 400$$

$$\text{At day 1: } L = \left(\frac{Q_1}{C \times P^{1/2}} \right)^{1/2} = \left(\frac{123.8}{0.00017 \times 17.0^{1/2}} \right)^{1/2} = 414.9$$

dL/dt for day 1 and day 0:

$$\frac{dL}{dt} = \frac{L_1 - L_0}{t_1 - t_0} = \frac{414.9 - 400}{1 - 0} = 14.9$$

Which suggests the hole size on average increased by 14.9 μm for the first day.

The sum dL/dt for Pipe 1 was the sum from day 0 to day 29, whereas for pipe 2 and pipe 3 was from day 0 to day 141.

The average dL/dt was the average value for the calculated dL/dt from day 0 to day 29 for pipe 1, whereas from day 0 to day 141 for pipe 2 and pipe 3.

So on average the leak diameter of pipe 1 decreased by 6.5 μm per day over the 141 days of experiment, whereas pipe 2 (dL/dt= -0.2 $\mu\text{m}/\text{day}$) and pipe 3 (dL/dt= -0.7 $\mu\text{m}/\text{day}$) did not decrease significantly.

Table A2. Summary of the time to self-repair for 150 μm or 400 μm holes in copper and carbon steel pipes for different conditions.

Time to self-repair (day) for holes facing downward at 20 psi				
Condition	Pipe 1	Pipe 2	Pipe 3	Average
Cu pH 3.0 150 μm	14	5	1	7
Cu pH 8.5 150 μm	1	4	2	18
Cu pH 8.5 150 μm	31	28	28	
Cu pH 8.5 150 μm	34	18	18	
Cu pH 11.0 150 μm	6	2	1	3
Cu pH 3.0 400 μm	74	74	74	NA
Cu pH 8.5 400 μm	28	141	141	NA
Cu pH 11.0 400 μm	141	141	141	NA
Fe pH 3.0 400 μm	90	27	36	NA
Fe pH 4.0 400 μm	12	19	12	14
Fe pH 8.5 400 μm	22	15	21	19
Fe pH 11.0 400 μm	6	12	6	8
Fe pH 5.5 150 μm	1	1	1	1
Fe pH 8.5 150 μm	0.5	2.5	0.5	1
Fe pH 11.0 150 μm	41	41	2.5	NA
Time to self-repair (day) for holes facing upward at 20 psi				
Cu pH 8.5 150 μm	2	116	60	43
Cu pH 8.5 150 μm	50	16	16	
Time to self-repair (day) for holes facing downward at 40 psi				
Cu pH 8.5 150 μm	71	NA		46
Cu pH 8.5 150 μm	87			
Cu pH 8.5 150 μm	5			
Cu pH 8.5 150 μm	20			
Cu pH 8.5 150 μm	46			

Number in green means the hole repaired at the specified days; Number in red means the hole did not repair at the specified days or the length of experiment.

Table A3. Summary of the maximum pressure the repaired hole could withstand before leaking.

Maximum pressure (psi) for holes facing downward at 20 psi			
Condition	Pipe 1	Pipe 2	Pipe 3
Cu pH 3.0 150 μm	25	90	25
Cu pH 8.5 150 μm	90	90	90
Cu pH 8.5 150 μm	65	>100	60
Cu pH 8.5 150 μm	85	50	30
Cu pH 11.0 150 μm	40	NA	30
Cu pH 3.0 400 μm	NA	NA	NA
Cu pH 8.5 400 μm	NA	NA	NA
Cu pH 11.0 400 μm	NA	NA	NA
Fe pH 3.0 400 μm	>100	25	NA
Fe pH 4.0 400 μm	>100	>100	>100
Fe pH 8.5 400 μm	60	25	25
Fe pH 11.0 400 μm	>100	>100	70
Fe pH 5.5 150 μm	>100	>100	>100
Fe pH 8.5 150 μm	>100	>100	>100
Fe pH 11.0 150 μm	NA	NA	NA
Maximum pressure (psi) for holes facing upward at 20 psi			
Cu pH 8.5 150 μm	35	85	55
Cu pH 8.5 150 μm	80	50	70
Maximum pressure (psi) for holes facing downward at 40 psi			
Cu pH 8.5 150 μm	>100	NA	
Cu pH 8.5 150 μm	60		
Cu pH 8.5 150 μm	45		
Cu pH 8.5 150 μm	50		
Cu pH 8.5 150 μm	45		

NA Represents the repaired holes that were not sent for pressure test due to leak failure or ESEM test. The cumulative fraction calculated for Figure 2-5 excludes these holes without available maximum pressures.

CHAPTER 3. IMPACT OF LEAK SIZE, PIPE WALL THICKNESS, WATER PRESSURE AND LEAK ORIENTATION ON AUTOGENOUS METALLIC PIPE LEAK REPAIR

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ABSTRACT

The effectiveness of autogenous metallic (iron and copper) pipe leak repair as a function of leak size, pipe wall thickness, water pressure and leak orientation was examined at water pressures up to 60 psi. The time to repair for carbon steel pipe leaks statistically increased with leak size to the power of 0.89–1.89, and decreased with pipe wall thickness to the power of -1.9 to -1.0. Additionally, water pressure and leak orientation did not influence the time to or likelihood of self-repair of galvanized iron coated pipe leaks, but leak size was a limiting factor as the repair likelihood decreased with the ln of leak size with a slope of -0.65. In contrast, the time to repair 150 μm leaks in copper statistically increased with water pressure to the power of 1.7.

KEYWORDS: Metallic pipe leak repair, leak size, water pressure, pipe wall thickness, leak orientation

INTRODUCTION

Leaking and aging potable water infrastructure is estimated to cause 7 billion gallons of daily water loss,¹ and leaks can also allow for water contamination, property damage and flooding.²⁻⁴ Conventional repair, rehabilitation and replacement approaches are effective, but they are very costly and time-consuming and pose economic challenges for some municipalities and building owners.⁵ The American Water Works Association (AWWA) estimated that about a trillion dollars are needed over the next 20 years to restore existing drinking water systems.⁶⁻⁷ Consequently, alternative approaches to reduce water pipe leak incidence that are at lower cost and are easier to implement, would be attractive to extend the lifetime and performance of these critical assets.⁵

It was recently discovered that water leaks in metallic pipes such as copper and iron could naturally repair themselves via formation of corrosion precipitates (Figure 3-1, A-B).^{5,8-10} This phenomenon of “autogenous leak repair” or self-repair was reproduced in the laboratory for carbon steel and copper pipe leaks at water pHs from 3.0–11.0 and water pressures from 20–40 psi (Figure 3-1, C).^{5,10} That earlier work found that corrosion could sometimes have beneficial impacts by causing leak repair—improved understanding and exploitation of this phenomenon could extend the lifetime of existing infrastructure.^{5,10} It was hypothesized that the leak size, leak orientation, water pressure, and pipe wall thickness could impact the trajectory of leaks towards repair or failure.^{5,10}

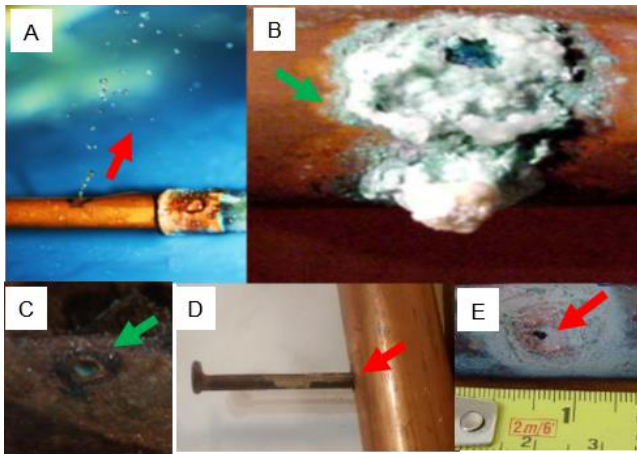


Figure 3-1. The copper pipe leaks (A) in contact with bulk water autogenously repaired (i.e., self-repaired) in a field study (B) as reproduced in the laboratory (C). Such leaks can be formed by physical penetration through water pipe wall by sharp objects such as nails (D), rocks (E), or after pit water is displaced from pinhole leaks (Adapted with the permission of Tang *et al.* 2015, Copyright 2015 American Chemical Society).

Common pinhole leaks in copper pipes found in premise plumbing systems are usually smaller than 1 mm² as observed in field studies,⁸ whereas the leak size observed in failed iron pipes from water mains ranges from 1–31,400 cm² as controlled by failure modes that include circumferential crack, longitudinal crack, leaking joints, corrosion pits and blow out holes.¹¹⁻¹³ Presumably, many of the iron pipe leaks observed during repair started from much smaller sizes.¹⁰ Field observations and laboratory studies also indicated that water

pipe leaks could occur in all orientations,^{8,14} but in other cases they tend to be on the bottom of pipes and preliminary work indicates repair incidence is not a function of orientation.¹⁰

The water pressure in mains is 20–200 psi or even higher,⁴ and pressure reduction has been a means of reducing leakage rate or failures.^{4,15-17} By installing a pressure reducing valve (PRV) before homes to provide customers with 20–80 psi water pressure (usually less than 50 psi),^{4,10,18-19} water conservation and longer plumbing life may be realized, and in some situations pressure in homes drops below 20 psi.^{4,10,20}

The wall thickness of metallic pipes in distribution systems has steadily decreased over the past century to reduce cost due to improved strength of materials.^{5,21-22} For example, the wall thickness of cast iron pipe (7" or 180 mm ID) built in the 1800s was around 26 mm and could withstand up to 360 psi,²³ whereas the similar size pipe in the high pressure category in 1920 was less than 15 mm,²⁴ and wall thickness has continually decreased with time.²¹ Wall thickness of copper pipes (0.6–5 mm) and galvanized iron (1.5–10.3 mm) in household plumbing system is much thinner than that in mains (up to 43 mm).^{24-26y}

Earlier testing examined autogenous repair of leaks up to 400 μm diameter, pressures up to 40 psi, and pipe wall thicknesses up to 890 μm .¹⁰ This work extended these preliminary results to more typical distribution system pressures of 20, 40 and 55 psi, leak sizes of 280, 400, 700 and 1000 μm and wall thicknesses of 360, 890, 1200 and 1700 μm for both copper and iron pipes. Both upward and downward orientations were tested. As corrosion precipitates could form in water and in some cases corrosion rust on the interior pipe surface could detach to the water resulting in clogging of leaks,^{5,27} the impact of large particles was also examined.

EXPERIMENTAL PROCEDURE

Two phases of experiments were conducted: 1) a small scale recirculation potable water system operated at 20–60 psi with synthesized water to examine leak size, pipe wall thickness, and water pressure impact on autogenous metallic pipe repair; 2) similar testing at building scale in Blacksburg, VA tap water at 20–55 psi, simulating a leaking premise plumbing potable water system.

Phase 1. Small Scale Recirculation Potable Water System

The experimental apparatus, water chemistry, procedures for measuring leakage rate, characterization of minerals clogging the leaks and pressure testing of leak repair strength are detailed in Tang *et al.* 2015 and summarized herein.

Experimental Apparatus at 20 psi. Water from a 21 liter reservoir was recirculated through triplicate 2" long (50.8 mm) metallic pipes at a pressure of 20 psi (Figure 3-2). A leak hole was drilled in the center of each pipe and all the holes were oriented downwards. The leaked water from each leak-hole was collected and routed back to the 21 liter reservoir to maintain constant volume. The drilled holes simulate the 30–40% existing leaks in potable water pipelines that do not result from pitting or non-uniform corrosion.^{10,28-29} In such cases the leak-holes are in contact with the bulk water from the start of the leak. The leaks also might represent those resulting from non-uniform pitting after the acidic pit solution has

Table 3-1. Key parameters examined in the small scale recirculation potable water system and large scale Blacksburg, VA tap water system.

Phase	Parameter	Pipe Wall Thickness ³				Leak Diameter ⁴				Water Pressure ⁵					
		W, μm	Unfiltered		Filtered		L, μm	Unfiltered		Filtered		P, psi	Unfiltered		Filtered
			N	D, day	N	D, day		N	D, day	N	D, day		N	D, day	
Phase 1. Small scale recirculation potable water system ¹	Material: Copper	360	3	180	3	180	150	9	47	3	20	20	9	47	NA
		890	3	104	3	104	250	3	180	3	180	40	5	98	
		1700	3	180	3	180	400	3	141	3	141	60	3	219	
	Material: Carbon Steel	890	6	148	3	34	150	3	15	3	15	NA			
		1200	6	54	3	74	400	3	24	NA	NA				
		1700	6	32	3	23	700	6	148	3	34				
						1000	3	42	3	81					
Phase 2. Large scale Blacksburg, VA tap water system ²	Material: Galvanized Iron Pipe	Orientation	N	D, day		L, μm	N	D, day		P, psi	N	D, day			
		Upward	36			280	18			20	24				
	Downward	36	366		400	18			40	24					
						700	18								
					1000	18	366		55	24	366				

¹Synthetic tap water composition: 15 mg/L Cl⁻, 30 mg/L alkalinity as CaCO₃, saturated DO, no disinfectant, pH 8.5

²Typical Blacksburg, VA tap water composition: pH 7.8, 3 mg/L chloramine, zinc phosphate (310 ppb as P), 40 mg/L alkalinity, 10.7 mg/L calcium, 14.5 mg/L chloride.²⁹

³Copper: 6.35 mm OD x 250 μm leak size; carbon steel: 6.35 mm OD x 700 μm leak size; water pressure = 20 psi

⁴Copper: 6.35 mm OD x 360 μm wall; carbon steel: 6.35 mm OD x 890 μm wall; water pressure = 20 psi

⁵Type M 1/2" copper pipe x 150 μm leak size.

W= pipe wall thickness; L=leak diameter; P=water pressure; N=total leak number;

D=experimental duration; NA=not available; Unfiltered/Filtered=conditions untreated/treated with a 1 μm filter

been displaced by bulk water flowing through the leak.¹⁰ Leak-holes without pitting corrosion can occur at imperfectly sealed soldered or threaded joints, or are formed by external pipe wall penetrations caused by sharp objects such as nails and rocks or at internal leaks resulting from cavitation or erosion corrosion (Figure 3-1, D-E).¹⁰ Copper and carbon steel pipes, which represent 33–90% of the premise plumbing water pipes in a typical water distribution system, were examined with pipe wall thickness up to 1700 μm and leak diameter size up to 1000 μm (Table 3-1).^{10,26,28-29} To increase the statistical power of the results, some conditions were repeated two or three times, resulting in a total of six or nine leak-holes for these conditions. To examine the effect of suspended particulate matter that might be formed as a result of pipe corrosion in potable water systems, a 1 μm pore size filter was sometimes installed at the end of the recirculation system to remove suspected

water particles (Figure 3-2). As the experiments were generally ended after all the triplicate leaks were completely repaired for at least 72 hours or at least one leak was destined for failure, the experimental duration of each condition varied between 15–148 days for iron and between 20–180 days for copper (Table 3-1).

Experimental Apparatus at 60 psi. To simulate leaking pipes at a common house plumbing system pressure of 60 psi, the 40 psi recirculating experimental apparatus in Tang *et al.* 2015 was used for each 150 μm leak drilled in the 2" (50.8 mm) long $\frac{1}{2}$ " type M copper pipe (12.7 mm OD x 11.4 mm ID) and the chlorinated polyvinyl chloride (CPVC) check valve was adjusted to achieve a 60 psi water pressure.²⁰ The test was repeated three times with a single copper pipe leak-hole tested each time. All the leak-holes were oriented downwards.

Water Chemistry. The reservoir was filled with a synthesized water comprised of 15 mg/L Cl^- from sodium chloride and 30 mg/L alkalinity as CaCO_3 at pH 8.5, a chemistry that has a high likelihood of autogenous leak repair.¹⁰ No disinfectant was added. The dissolved oxygen (DO) was always 5.3–8.6 mg/L.

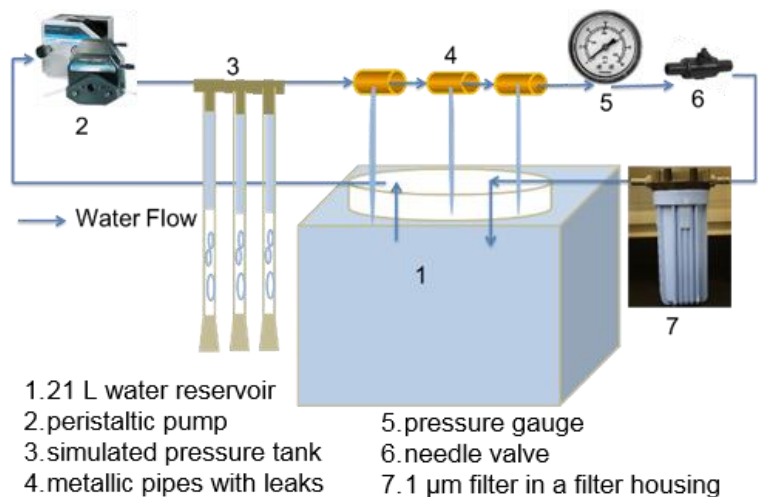


Figure 3-2. Experimental apparatus of the small scale recirculation potable water system with a 1 μm filter (Adapted with the permission of Tang *et al.* 2015, Copyright 2015 American Chemical Society).

Hole Leakage/Repair Rates. The leakage rate for each hole was tracked by quantifying the instantaneous fill rate of a graduate cylinder on a daily basis at a constant pressure of 20 psi or 60 psi unless stated otherwise.¹⁰ The conclusion of autogenous repair was defined as the time when the leakage rate was reduced to zero.

Strength/Characterization of Repaired Leaks. The strength of each repaired leak was tested by increasing the water pressure in 5 psi increments from 25 to 100 psi.¹⁰ The maximum pressure each repaired leak could withstand, was defined by the pressure point of failure at which water started leaking from the repaired holes. For leaks that did not fail at 100 psi, the sealing materials in the leaks were photographed and characterized using elemental

composition via environmental scanning electron microscope with an attached X-ray energy dispersive system (ESEM/EDS).

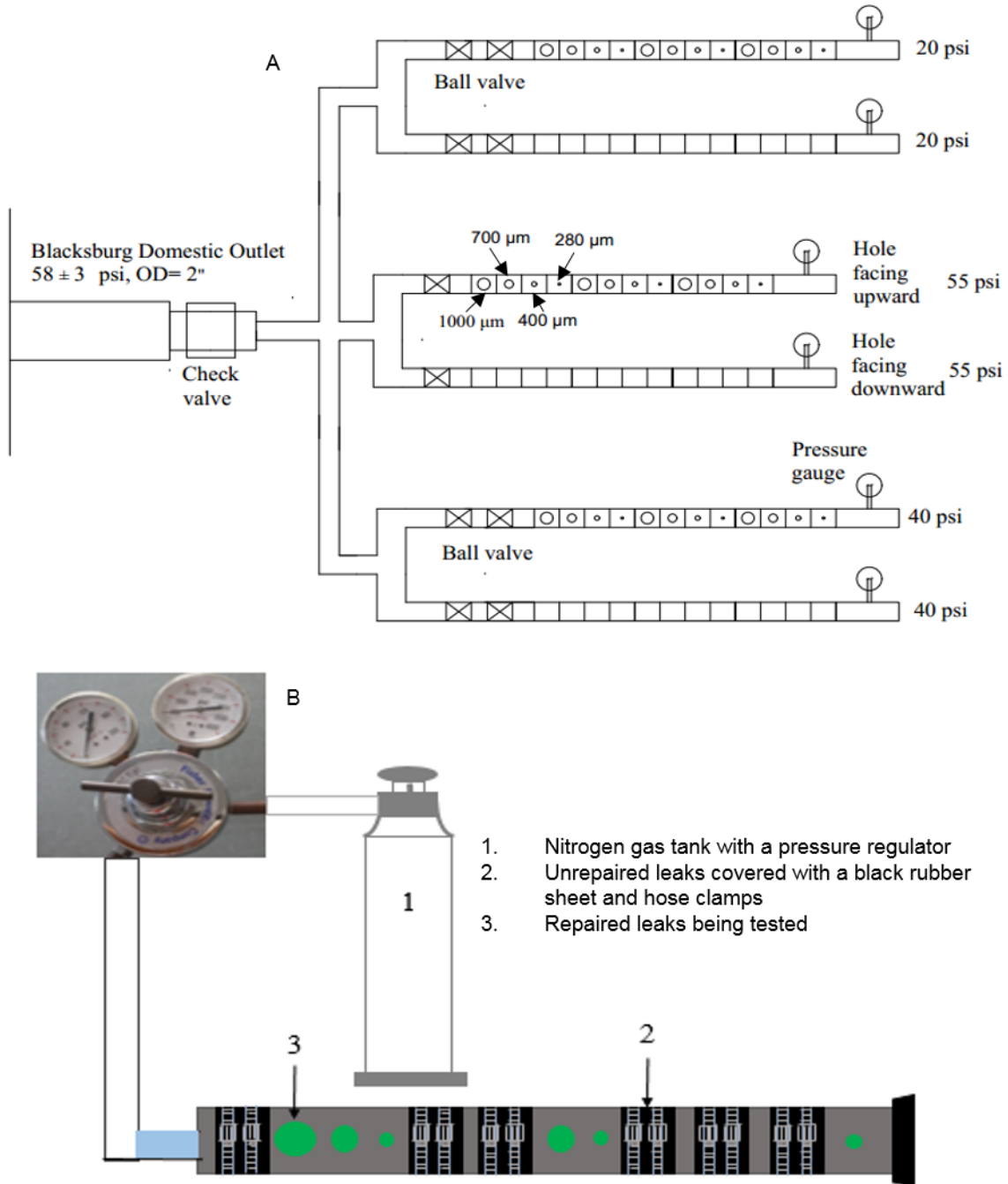


Figure 3-3. Schematic diagram (top view) (A) of the experimental apparatus (B) for the large scale Blacksburg, VA tap water system. Six 3/4" (19 mm) galvanized iron coated pipes with upward and downward leaks (1000, 700, 400 and 280 μm) in triplicate were examined at 20, 40 and 55 psi. Pressure testing was performed to examine the maximum pressure the repaired leaks could withstand before leaking (B).

Phase 2. Large Scale Blacksburg, VA Tap Water System

Experimental Apparatus. A system comprised of six 3/4" (19 mm) galvanized iron coated pipes, was connected to a domestic Blacksburg, VA tap water outlet at pressure of 58 ± 3 psi (Table 3-1; Figure 3-3, A). A one way check valve was installed after the domestic outlet, followed with three branches of galvanized iron coated pipes operated at three pressures. At each pressure (20, 40 or 55 psi), two hole orientations were examined including holes facing upward and downward, to represent the orientation extremes as in a previous study.¹⁰ For each pipe, each leak diameter size (1000, 700, 400 or 280 μm) was tested in triplicate and was drilled every 1 ft (30.5 cm) in the pipe, with the largest 1000 μm leak closest to the domestic outlet. Leak-holes were drilled every 3 in (7.6 cm). Two ball valves were used to reduce the pressure to targets of 40 or 20 psi as monitored by the pressure gauge (0–100 psi range).

Blacksburg Tap Water Chemistry. The typical Blacksburg, VA tap water has a neutral to basic pH of around 7.8, low alkalinity of 40 mg/L as CaCO_3 , calcium concentration of 10.7 mg/L, chloride of 14.5 mg/L, chloramine residual of 3 mg/L and corrosion inhibitor of 310 ppb zinc phosphate as P (Table 3-1).³⁰

Hole Leakage/Repair Rates. The success of repair was monitored by tracking the leakage rate as in the small-scale recirculation potable water system (Phase 1) for a year (366 days).¹⁰ The leakage rate from each leak was monitored three times per week for the first 4 months, once per week for another 2 months, and then every two weeks thereafter.

Strength/Characterization of Repaired Leaks. After the one-year (366 days) experiment, the unrepaired leaks in each pipe were covered with a black rubber sheet and hose clamps, the end of each pipe was sealed, filled with water and connected to a nitrogen gas pressure tank (Figure 3-3, B). A pressure test was then conducted by increasing examined pressure from 20 to 100 psi in 5 psi increments. Similar to Phase 1, the maximum repair pressure was defined as the point where water first leaked.

Statistical Comparisons. As the data were not normally distributed, non-parametric statistics including Wilcoxon test and Kruskal-Wallis test were used to compare the time to repair.³¹ Non-linear regression was employed to develop the relationship between the time to repair and the leak size, the wall thickness and the water pressure. The calculated p value of < 0.05 based on an alpha (α) value of 0.05 indicated that the coefficients or means were significantly different.

RESULTS AND DISCUSSION

Phase 1. Small Scale Recirculation Potable Water System

Both carbon steel and copper pipe leaks of up to 1000 μm in a pipe wall of up to 1700 μm and at water pressures up to 60 psi could repair themselves as indicated by reduction of leakage to zero (Figure 3-4, 3-5, 3-6, 3-7, 3-8 and 3-9). For example, one 700 μm leak in a pipe with an 890 μm wall thickness, had an initial leakage rate of 54.7 L/d and reduced to zero from 3–8 days (Figure 3-4). But the leakage rate increased to 104 L/d after 8 days and increased to 363 L/d after 64 days, before finally decreasing to zero after 72 days and staying sealed to the end of the experiment. All the leaks were repaired via rust clogging

the leak hole, as illustrated by one repaired 400 μm leak examined under ESEM/EDS (Figure 3-5). The repaired materials in the leak (Figure 3-5, A and C) were comprised of 9% carbon, 27% oxygen and 64% iron on a normalized mass basis (Figure 3-5, B and D), which was the same as iron rust formed on the pipe surface.²⁷

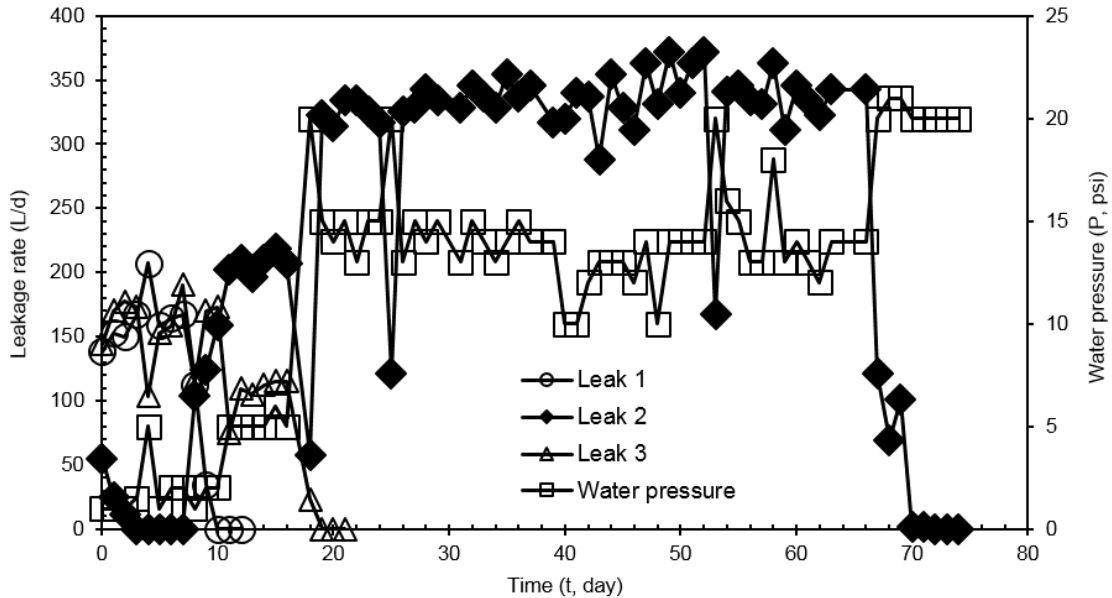


Figure 3-4. The leakage rate was reduced to zero indicating success of repair for 700 μm carbon steel (6.5 mm OD x 890 μm wall thickness) leaks in the unfiltered condition. When the leak size was larger than 700 μm , the system water pressure was not able to achieve the 20 psi target for this apparatus.

Effect of Leak Size

Leaks with unfiltered water. The time to repair for both copper and iron pipes was limited by leak size when no filter was implemented (Figure 3-6, A; Figure 3-7). The time to repair of carbon steel pipe leaks increased with leak size to the power of 1.85 ($t=10^{-4} L^{1.85}$, t in days, L in μm ; $p < 0.05$ in non-linear regression). Specifically, all of the 150, 400, 700 and 1000 μm leaks repaired themselves after 2 (on average 1 day), 22 (on average 19 days), 146 (on average 49 days) and 37 days (on average 23 days), respectively.¹⁰ The 1000 μm leaks had a faster average repair time but two of the six 700 μm leaks self-repaired after only 10 days, which was about half of the average time needed to repair all the 1000 μm leaks. The power of leak size in the developed equation was still statistically different from zero despite the anomalous result at 700 μm .

For leak sizes $> 700 \mu\text{m}$, the peristaltic pump system output was not capable of maintaining a targeted 20 psi initially. For example, water pressure started at around 1 psi for triplicate 700 carbon steel pipe leaks in 890 μm pipe wall, and pressure progressively increased as holes filled or were partly clogged (Figure 3-4). With a 1000 μm leak size, the water pressure could be maintained at 20 psi after at least two of the three leaks were repaired (data not shown). This limitation necessitated development of a much larger rig connected

to distribution system higher pressures and flow volumes to study the effects of ambient pressure on leak repair rates.

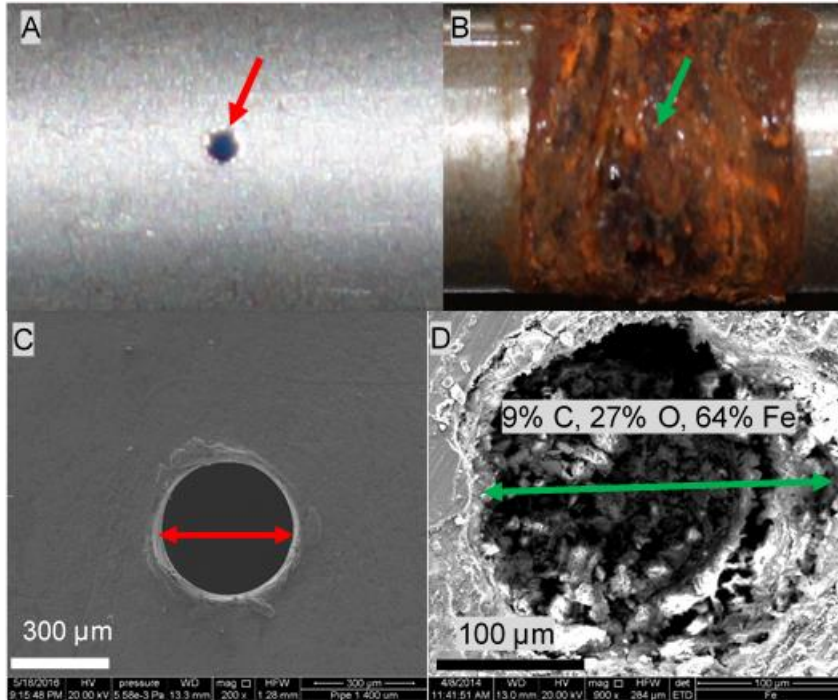


Figure 3-5. Carbon steel leaks were repaired with iron rust as identified under ESEM/EDS. One 400 µm leak (A, C) (pipe size: 6.35 mm OD x 890 µm wall) was shown as an example and the sealing materials (B) were composed of 9% C, 27% O and 64% Fe on a normalized mass percent basis (D)

Compared to the leak repair in carbon steel pipes, a lower repair likelihood was observed for copper pipe leaks as 0–100% of the leaks repaired themselves depending on the wall thickness and leak size (Figure 3-7). A lower success rate of autogenous repair was observed with larger leak size. For instance, all (9/9) smaller 150 µm leaks were repaired with copper rust after 34 days (on average 18 days)¹⁰ whereas none (0/3) of the 250 µm leaks after 180 days and 33% (1/3) of the 400 µm leaks sustained repairs after 141 days (Figure 3-8). One of the nine repaired 150 µm leaks was sealed with a deposit of 37% C, 38% O, 3% Cl and 22% Cu on a normalized mass percent (Figure 3-8, A-B) whereas one of the two unrepaired 400 µm leaks remained open at the end of 141-day experiment (Figure 3-8, C-D). No numerical relationship could be developed between time to repair and leak size.

Leaks with filtered water. The removal of large suspended water particles via a 1 µm filter did not influence the self-repair capability of both carbon steel and copper pipes (Figure 3-6, B; Figure 3-7). When the filtered water was flowing the carbon steel pipes with an 890 µm wall, a similar trend was observed as in the unfiltered condition. The time to repair of each carbon steel leak increased with the hole size to the power of 0.89 ($t=0.06 L^{0.89}$, t in day, L in µm) ($p < 0.05$ in non-linear regression) (Figure 3-6, B).

In the case of leaks in copper pipe with a 360 μm thick wall, the 1 μm filter treatment also yielded similar results to the unfiltered conditions (Figure 3-7, A-B). All smaller 150 μm leaks were repaired after 18 days (on average 16 days) whereas only 33–67 % of the larger 250–400 μm leaks repaired themselves at the end of the 6-month experiment.

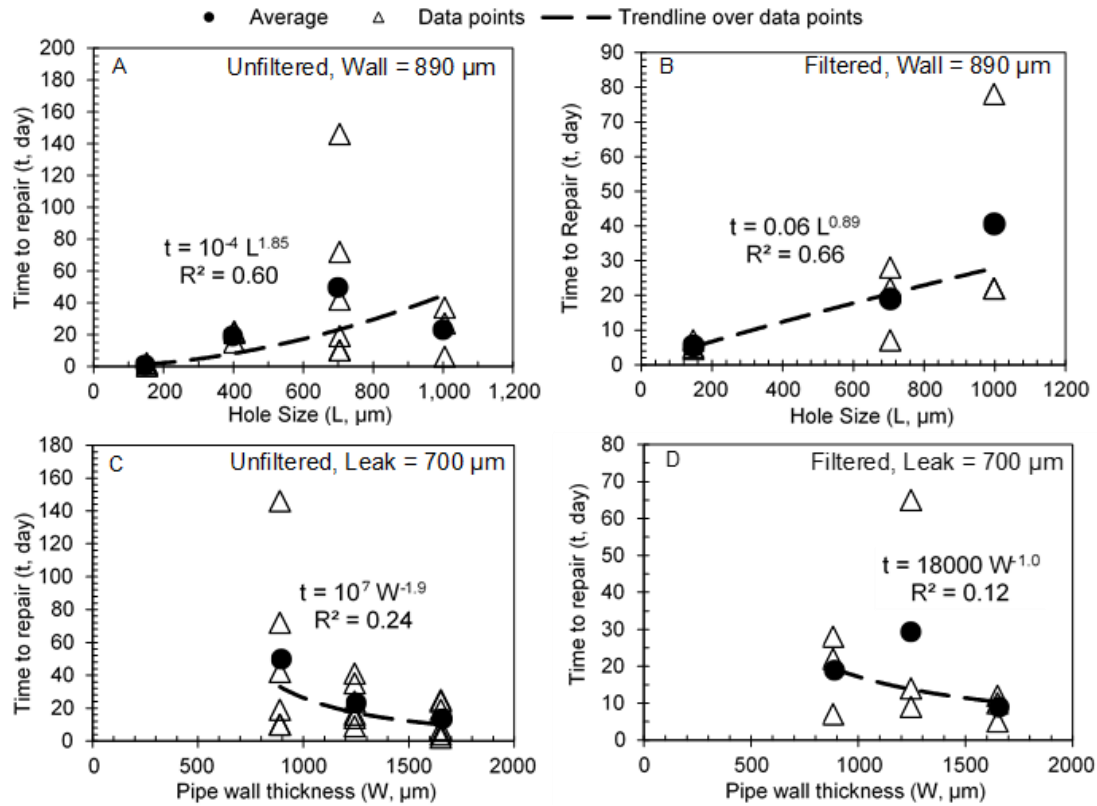


Figure 3-6. Leak size and pipe wall thickness impact on the time to repair of carbon steel pipe leaks in unfiltered (A, C) and filtered (B, D) conditions at 20 psi and pH 8.5.

Effect of Pipe Wall Thickness

Leaks with unfiltered water. The time to repair was a function of wall thickness for both carbon steel and copper pipe leaks (Figure 3-6, C; Figure 3-7). The time to repair of carbon steel 700 μm leaks decreased with the pipe wall thickness to the power of -1.9 ($t=10^7W^{-1.9}$, t in days, W in μm) ($p < 0.05$ in non-linear regression). Specifically, all six 700 μm leaks in 890, 1200 and 1700 μm thick walls self-repaired after 146 (on average 50 days), 41 (on average 23 days) and 25 days (on average 14 days), respectively.

For 250 μm leaks in copper pipe, no relationship was developed between the time to repair and pipe wall thickness but a higher likelihood of self-repair was identified at the wall thickness of 890 μm among the three examined wall thicknesses (Figure 3-7, A-B). All three 250 μm leaks in an 890 μm thick wall self-repaired after 87 days (on average 45 days) whereas 0% (0/3) of the leaks in the thinner wall of 360 μm and 67% (2/3) in the thicker wall of 1700 μm autogenously repaired after 6-month experiment.

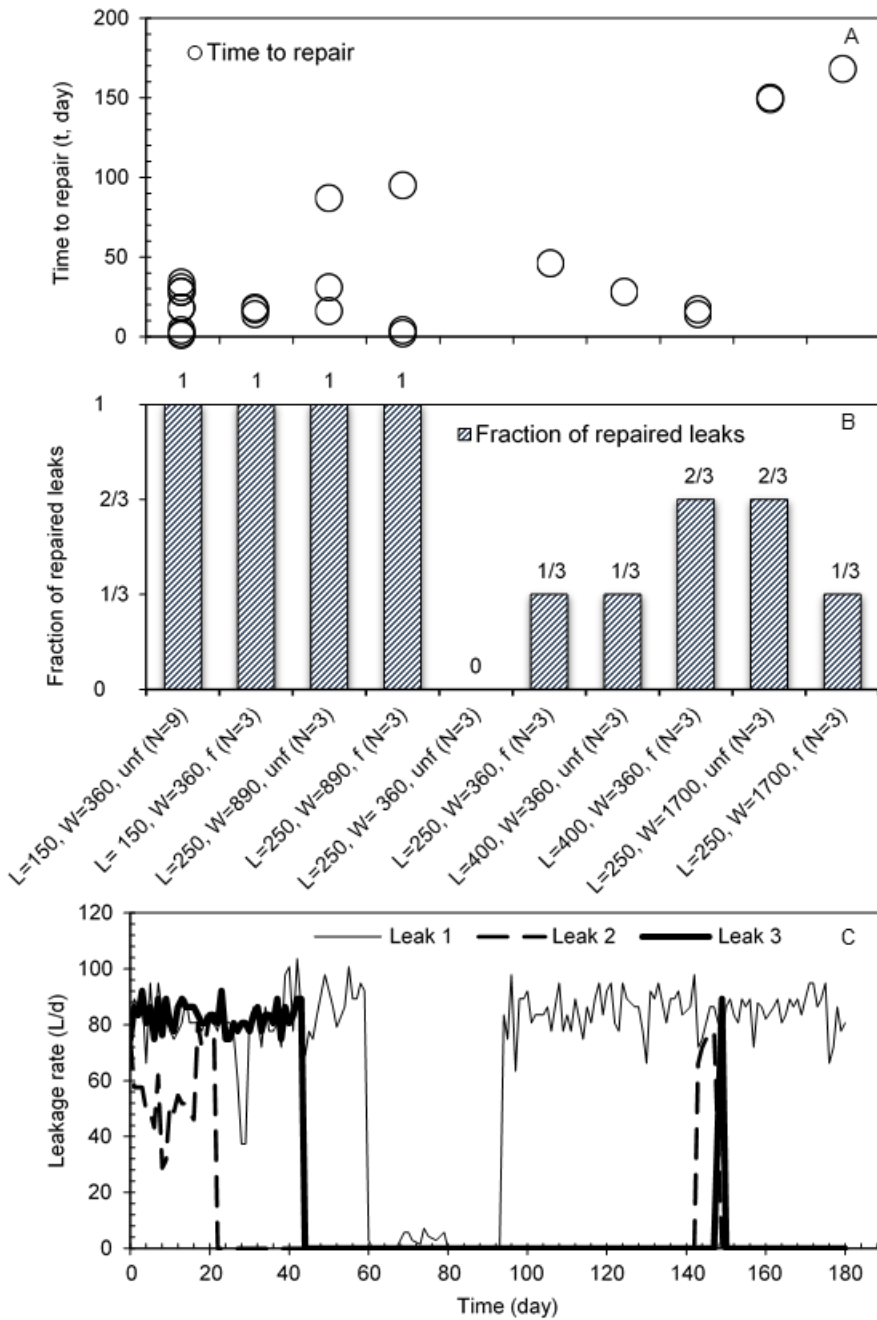


Figure 3-7. Autogenous repair of copper pipe leaks in unfiltered and filtered conditions at 20 psi and pH 8.5 (L=leak diameter in μm , W=wall thickness in μm , unf = unfiltered, f = filtered, N=total leak number). The time to repair was only shown for repaired leaks (A) and fraction of repaired leaks was calculated for all examined conditions (B). Exemplary case of one leak that repaired and reopened for 250 μm leaks in 1700 μm thick copper pipes in unfiltered conditions, whereas two out of three leaks self-repaired permanently (C).

A non-durable repair was observed for 250 μm leaks in a 1700 μm thick wall, in that a sealing and breaking process occurred multiple times (Figure 3-7, C). As an example, one 250 μm leak (leak 2 in Figure 3-7, C) repaired itself after 22 days and then stayed repaired for 120 days. It then broke after 143 days and stayed open for 5 days at which point it was repaired again until the end of the experiment. The strength of the minerals sealing the holes under these conditions, is hypothesized to play a key role in the longevity of repair for copper pipe leaks.

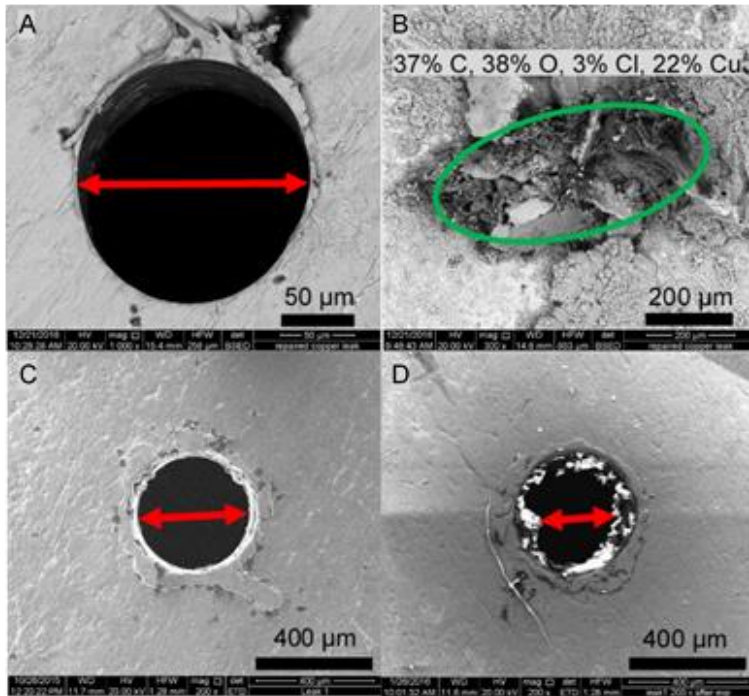


Figure 3-8. Smaller 150 μm leaks (A) in copper pipe (6.35 mm OD x 360 μm wall thickness) autogenously repaired with copper rust (37% C, 38% O, 3% Cl and 22% Cu on a normalized mass percent basis) as identified under ESEM/EDS at pH 8.5 and 20 psi (B). In contrast, one larger 400 μm leak (C) in same size copper pipe failed to repair itself (D).

Leaks with filtered water. Similarly, removal of large particles via implementing a 1 μm filter did not attenuate the repair capacity of both carbon steel and copper pipe leaks (Figure 3-6, D; Figure 3-7). The time to repair of carbon steel 700 μm leaks decreased with a power of -1.0 as the pipe wall thickness increased ($t=18000W^{-1.0}$, t in days, W in μm) ($p > 0.05$ in non-linear regression). A large variability was observed among the duplicate samples in filtered conditions such that the correlation coefficients were not statistically different from zero. The comparatively high repair rate in iron pipe walls examined in this work, implied that the thicker pipes in practice might possibly have a much higher repair rate for similar size leaks.²³⁻²⁴

Similar observations were made for the 250 μm leaks in copper pipes in the filtered conditions (Figure 3-7). The 890 μm copper pipe wall induced higher likelihood (100%)

of self-repair for 250 μm leaks versus 33–67% for 360 and 1200 μm thick walls. The non-durable repair was also observed in the complementary filtered condition as sealing and breaking processes repeatedly occurred in the 1700 μm thick wall (data not shown).

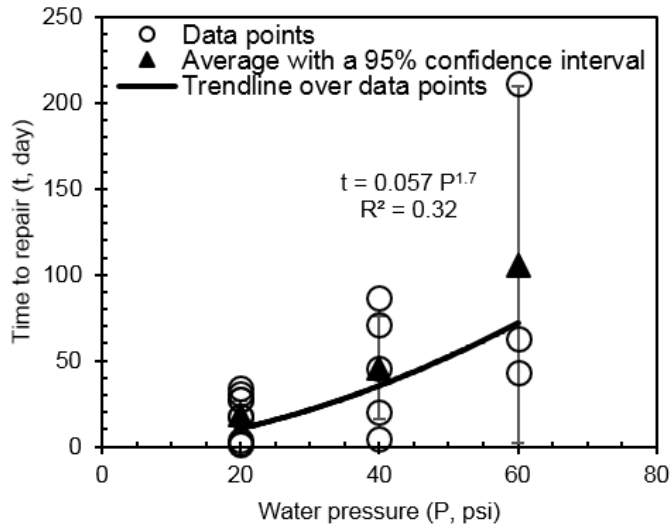


Figure 3-9. The time to repair increased as water pressure increased for copper pipe 150 μm leaks in the unfiltered condition at pH 8.5 in the small recirculation potable water system.

Effect of Water Pressure

The seventeen 150 μm leaks in copper pipes had a 100% repair rate at water pressure ranging from 20–60 psi in unfiltered conditions (Figure 3-9).¹⁰ The time to repair increased as the water pressure increased to the power of 1.7 ($t=0.057P^{1.7}$, t in day, P in psi) ($p < 0.05$). Though the time to repair did not have any statistical difference for 150 μm leaks between 20 psi and 40 psi (Wilcoxon test, $p > 0.05$),¹⁰ all the same size leaks at 60 psi repaired themselves after 211 days, which on average was statistically longer than the time to repair at 20 psi (Wilcoxon test, $p < 0.05$).

Maximum Pressure of Repaired Leaks

Strength of repair in copper pipes at 20 psi. The autogenously repaired copper leaks were relatively strong (Figure 3-10). For the copper pipe leaks repaired at 20 psi with filtered water flowing through the pipe, about 25% could withstand 55 psi which is a common house plumbing system pressure²⁰ and 12% could withstand more than 100 psi which is considered a high pressure in building plumbing systems.⁴ When the bulk water was not treated with a 1 μm filter, about 50% of the repaired leaks could withstand 55 psi but only 8% of them could withstand more than 100 psi.¹⁰ No statistical difference was observed for the strength of repaired leaks between the filtered and unfiltered conditions in copper pipes (Wilcoxon test, $p > 0.05$).

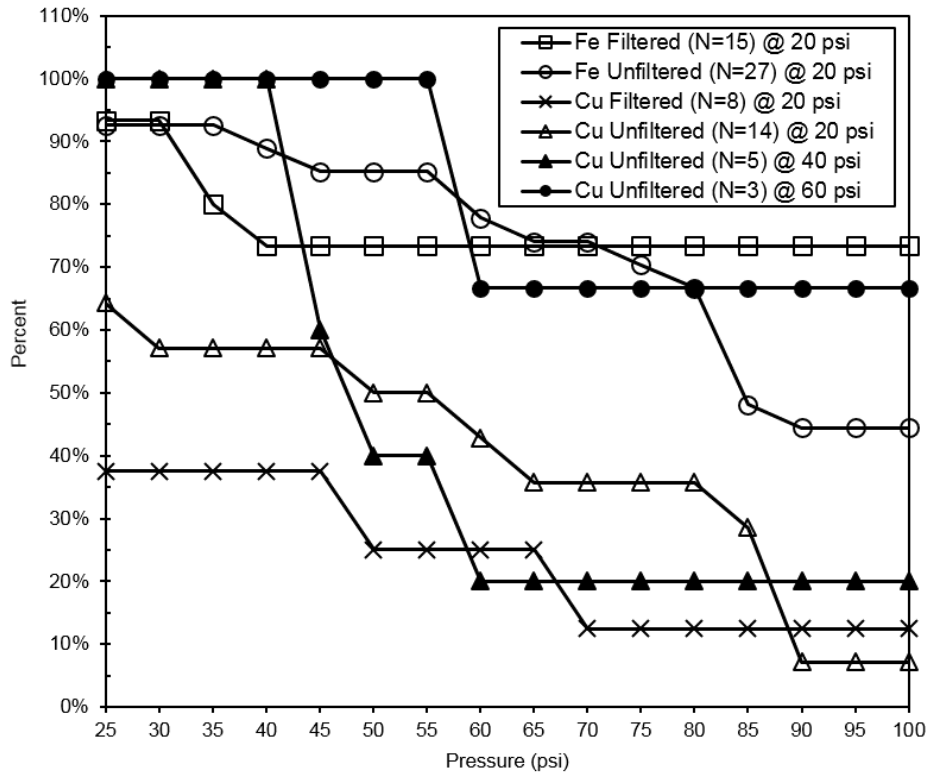


Figure 3-10. Percent of the repaired leak-holes in the small scale recirculation potable water system that did not fail at the examined pressures (N = total repaired leak number).

Strength of repair in copper pipes at higher pressures. Compared to leaks repaired at 20 psi, 20% (1/5) of the repaired copper pipe leaks at 40 psi could withstand more than 100 psi (Figure 3-10).¹⁰ About 67% (2/3) of the repaired leaks at 60 psi could withstand more than 100 psi, suggesting that copper pipe leaks autogenously repaired at higher pressure might be stronger.

Strength of repair in carbon steel pipes at 20 psi. Stronger repair was observed for carbon steel pipe leaks at 20 psi versus copper pipe leaks (Figure 3-10).¹⁰ At the examined water pressure of 55 psi, 74% stayed repaired with filtered water and higher percent of 85% remained repaired with unfiltered water. At 100 psi, 74% stayed repaired with filtered water and 45% remained clogged with unfiltered water. Similar to observations in the repaired copper pipe leaks, there was no statistical difference for the strength of carbon steel pipe leak repair between the filtered and unfiltered conditions (Wilcoxon test, $p > 0.05$), indicating that the presence of large particles did not influence the strength of the repair.

The repair in carbon steel pipe leaks was stronger than the copper pipe leak repair (Wilcoxon test, $p < 0.05$). About 45–74% of the carbon steel pipe leaks repaired at 20 psi could withstand 100 psi, which was at least 3 times higher than that of the copper pipe leaks. It is possible that the 10X higher corrosion rate of carbon steel pipe versus copper, potentially increased the density and strength of repair.^{10,32-33}

The strong repair of medium to large size leaks at water pressure of 20–60 psi indicates, that some fraction of leaks in copper and iron pipes could potentially repair themselves and the repair could last to sustain the leaking water infrastructure.

Phase 2. Large Scale Blacksburg, VA Tap Water System

Effect of Leak Orientation

The leak orientation did not make any statistical difference in the likelihood of self-repair or the time to repair (Kruskal-Wallis test, $p > 0.05$). For the 27 repaired leaks after the 366-day experiment, 48% (13/27) of them were facing upward whereas 52% (14/27) were facing downward (Table 3-2). Additionally, they were repaired at a similar speed due to the large variability of the time to repair in both orientations. For instance, the thirteen repaired upward leaks were repaired after 11 to 352 days and the fourteen downward repaired leaks had a complete repair after 21 to 311 days. The repaired leaks were also sealed with corrosion rust starting from inside the pipe as verified under ESEM/EDS, taking an upward 400 μm leak as an example (Figure 3-11).

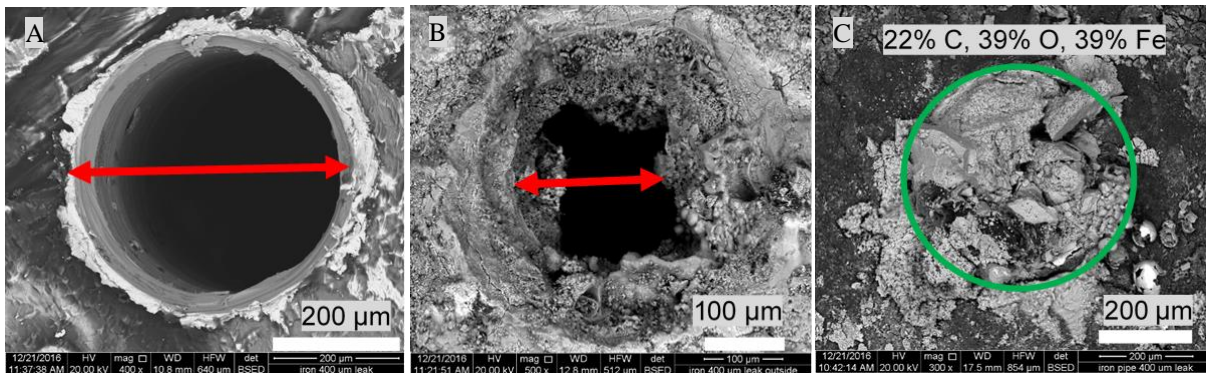


Figure 3-11. The first upward original 400 μm leak (L1) (A) from the domestic outlet autogenously repaired at 40 psi in the large scale Blacksburg, VA tap water system. The leak was not completely covered with rust from outside (B) but the hole was blocked with rust (22% C, 39% O and 39% Fe on a normalized mass percent basis) formed from inside the pipe as identified under ESEM/EDS (C).

Effect of Water Pressure

The inability of the laboratory peristaltic pump to produce sufficient output pressure to maintain 20 psi for holes larger than 700 μm was overcome in the large Blacksburg, VA tap water testing system. The testing system connected to the water distribution system, could maintain a constant pressure of 20, 40 or 55 psi for each pipe as required.

Surprisingly, the water pressure did not make any difference in the repair likelihood or time to repair (Kruskal-Wallis test, $p > 0.05$). Regardless of the leak orientation which did not influence the autogenous repair likelihood and rate, similar fractions of leaks self-repaired at a similar speed for the three examined pressures after a one-year experiment in the building plumbing configuration (Table 3-2). Specifically, nine out of 24 leaks were self-

repaired after 11–352 days at 20 psi and eleven were repaired after 44–352 days at higher pressure of 40 psi, whereas seven repaired themselves after 21–296 days at 55 psi.

Table 3-2. Summary of the time to repair for leaks in the large scale Blacksburg, VA tap water system after one-year (366 days) experiment.

Leak Diameter (µm)	Time to Repair (t, day)																	
	Water Pressure = 20 psi						Water Pressure = 40 psi						Water Pressure = 55 psi					
	Downward			Upward			Downward			Upward			Downward			Upward		
	L1	L2	L3	L1	L2	L3	L1	L2	L3	L1	L2	L3	L1	L2	L3	L1	L2	L3
1000	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
700	114	-	-	-	352	-	226	-	-	-	-	-	-	-	-	-	-	-
400	-	-	-	93	-	-	86	65	-	44	352	163	-	296	163	-	-	213
280	53	56	311	98	60	11	311	37	311	58	-	163	86	21	-	-	268	184

Note that - represents the leaks did not self-repair at the end of experiment. L1=Leak 1 which was the leak of that size closest to the domestic outlet; L3=Leak 3 which was the leak of that size furthest from the domestic outlet (Figure 3-3).

Effect of Leak Size

Since leak orientation and water pressure had no significant impact, data from these different conditions were pooled to examine the effect of leak size. Though leak size did not make a statistical difference in the time to repair as the repair time varied dramatically for each of the four leak sizes examined across the three pressures (Kruskal-Wallis test, $p > 0.05$), a logarithmic relationship was developed between the fraction of repaired leaks (F) and the leak diameter size (L) after 3, 6, 9 and 12 months (Table 3-2; Figure 3-12).

At the end of the one-year experiment, fifteen 280 µm leaks, nine 400 µm leaks, three 700 µm leaks and none of the 1000 µm leaks repaired themselves. Compared to that of 3 months, the slope of the equation decreased from -0.34 to -0.65 and the intercept increased from 2.29 to 4.42 showing a stronger negative correlation between the ln of leak size and the fraction of repaired leaks over time ($p < 0.05$ in non-linear regression).

The logarithmic relationship developed from the large scale Blacksburg, VA tap water system was also applicable to 150 µm leaks with a 100% repair rate from the small scale recirculation potable water system (phase 1) (Figure 3-12).¹⁰ The calculated fractions from 9 and 12 months in Figure 3-10 were both close to the actual 100% repair rate in the small scale recirculation system (Figure 3-12, C-D). It further implies that the iron leak repair was limited by leak size.

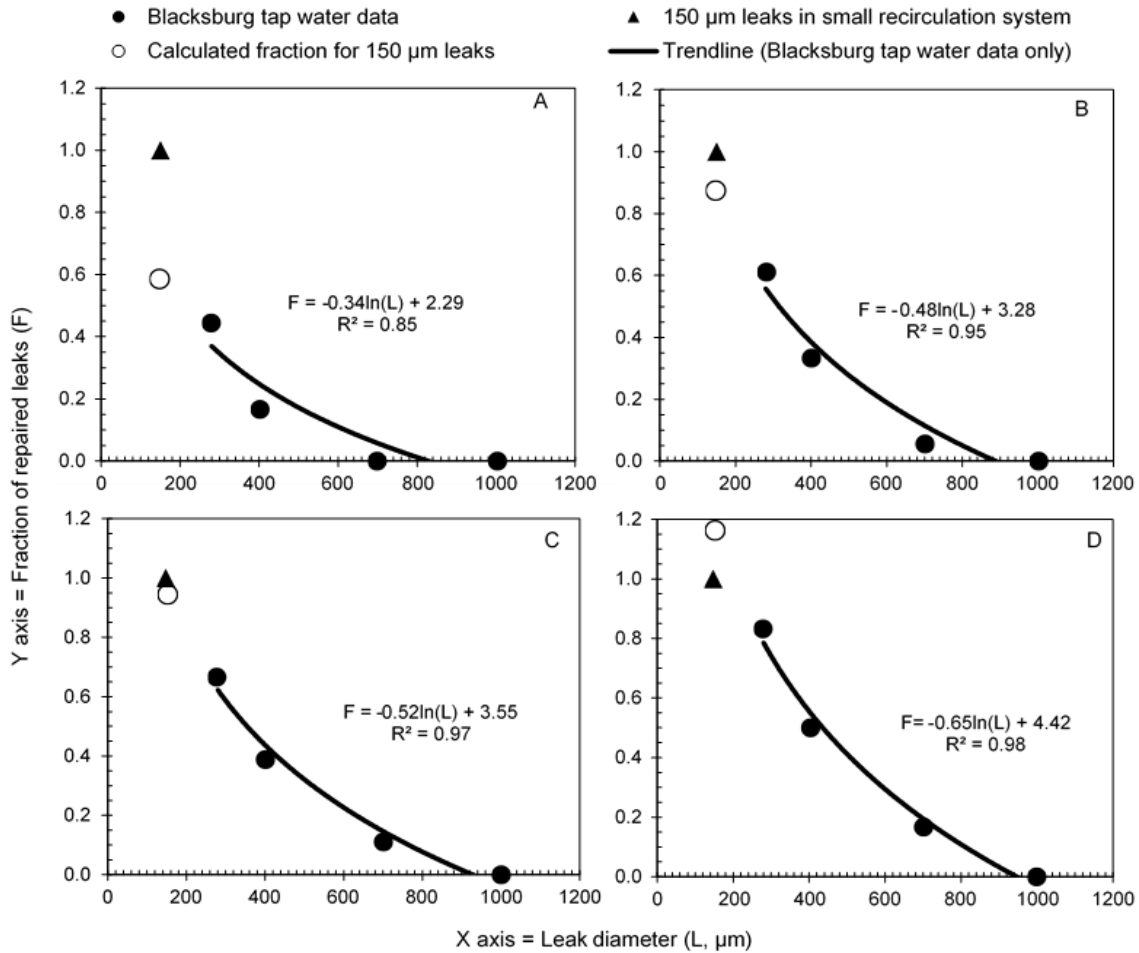


Figure 3-12. The fraction of repaired leaks as a function of leak diameter size in the large scale Blacksburg, VA tap water system after 3 months (A), 6 months (B), 9 months (C) and 12 months (D). The 100% repair rate of 150 μm carbon steel pipe (6.35 mm OD x 890 μm wall thickness) leaks in Phase 1 was compared to the calculated fraction of repaired 150 μm leaks using the developed equations (A-D) from Phase 2.

Strength of Repaired Leaks

A few repaired holes failed at a lower pressure than the experimental pressure. For instance, all (4/4) the repaired downward leaks at 20 psi were either broken or failed at 20 psi, resulting in the much lower bound of the maximum pressure in pressure testing. It is likely that some of the repaired leaks were broken or partially damaged before pressure testing from the unavoidable and unusual disturbance during disassembly of the leak test apparatus or assembly of the pressure test apparatus.¹⁰

But overall the strength of repair was strong (Figure 3-13). For example, about 60% (3/5) of the repaired upward leaks at 20 psi could withstand 100 psi which is considered as a high house plumbing pressure.⁴ About 40% (2/5) of the repaired upward leaks at 40 psi and 50% (2/4) of downward leaks repaired at 55 psi could withstand 100 psi.

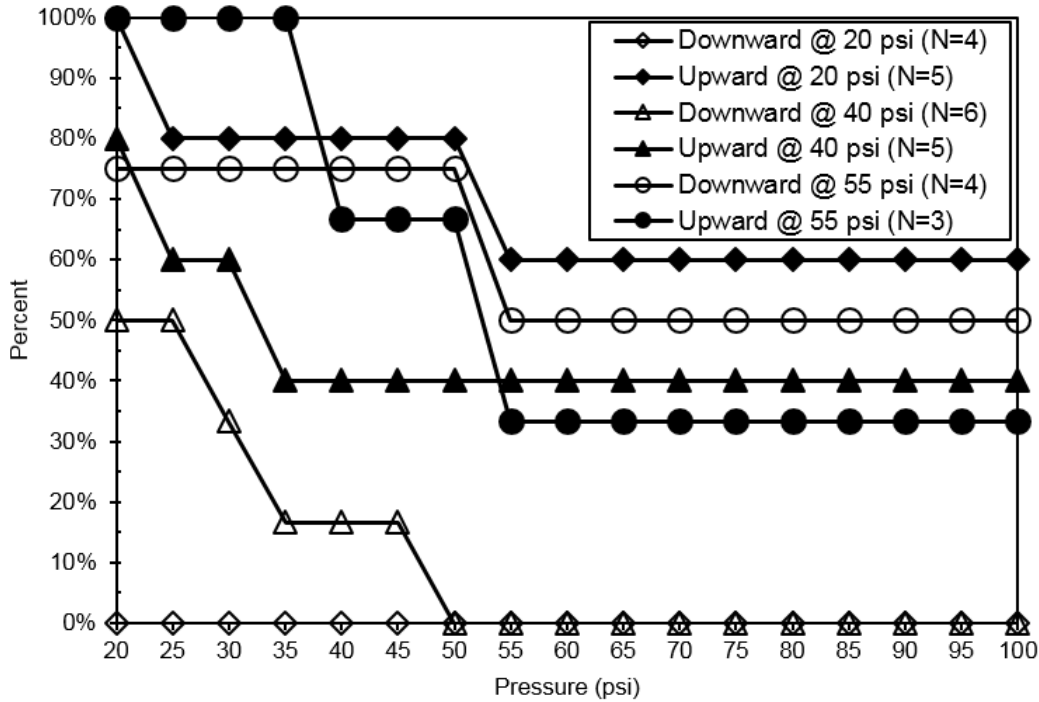


Figure 3-13. Percent of the repaired leaks in the large scale Blacksburg, VA tap water system that did not fail at the examined pressures (N = total repaired leak number).

CONCLUSIONS

Autogenous repair of copper and iron pipe leaks via corrosion rust occurred over a wide range of pipe wall thicknesses, leak sizes, water pressures and leak orientations. In the small scale recirculation potable water system (phase 1) at 20 psi and pH 8.5:

- ❖ The time to repair (t) of carbon steel leaks increased as the leak size (L) increased, according to the equations: $t = 10^{-4}L^{1.85}$ ($p < 0.05$) for the unfiltered condition and $t = 0.06L^{0.89}$ ($p < 0.05$) for the filtered condition.
- ❖ The time to repair (t) of carbon steel leaks increased as pipe wall thickness (W) decreased, as described in the equations: $t = 10^7W^{-1.9}$ ($p < 0.05$) for the unfiltered condition and $t = 18000W^{-1.0}$ ($p > 0.05$) for the filtered condition.
- ❖ The time to repair (t) of copper 150 μm leaks increased as the water pressure (P) increased, based on the developed equation: $t = 0.057P^{1.7}$ ($p < 0.05$).
- ❖ The presence of large particles did not influence the repair capacity and repair strength for both carbon steel and copper pipe leaks.
- ❖ Stronger repair was observed in carbon steel pipe leaks versus copper leaks ($p < 0.05$).

When galvanized iron coated pipe leaks were examined in the large scale Blacksburg, VA tap water system at water pressure from 20 – 55 psi for 366 days:

- ❖ The leak orientation and water pressure was not influential in the repair likelihood and the time to repair ($p > 0.05$).
- ❖ The leak size was a limiting factor for self-repair in that the fraction of repaired leaks linearly decreased with the ln of leak size ($p < 0.05$).

ACKNOWLEDGEMENTS

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CHAPTER 4. WATER CHEMISTRY IMPACT ON AUTOGENOUS METALLIC PIPE LEAK REPAIR IN SIMULATED POTABLE WATER SYSTEMS

Min Tang, Marc Edwards

ABSTRACT

The impact of water chemistry (ionic strength, alkalinity, silica, magnesium, NOM, DO, disinfectants, corrosion inhibitors and corrosion indices) on autogenous repair of 400 μm carbon steel (6.35 mm OD x 890 μm wall) and 200 μm copper (6.35 mm OD x 890 μm wall) leaks were examined in modifications of a potable water at pH 8.5. Higher DO and RSI significantly decreased the time to repair of carbon steel 400 μm leaks by at least 50–70% and chlorine dioxide produced a 300% increase in the fraction of repaired 200 μm copper leaks. Weight loss of metals and presence of particulate metal rust in the water were not correlated with the time to repair of 400 μm carbon steel leaks or the fraction of repaired 200 μm copper leaks. A significant fraction (15–38%) of the leaks repaired at 20 psi could withstand at least 100 psi water pressure, confirming the potential for autogenous repair to extend to lifetime of existing water infrastructure in at least some circumstances.

KEYWORDS: Water chemistry, autogenous repair, iron pipe leaks, copper pipe leaks, potable water

INTRODUCTION

Aging and leaking water infrastructure in the United States causes 7 billion gallons of lost water daily and property damage, and it creates health concerns.¹⁻³ Traditional approaches of pipe repair, rehabilitation and replacement are effective but costly, and will require decades to implement even if society makes water infrastructure upgrades a priority. New approaches that can reduce or stop water pipe leaks, and extend the lifetime of aging water infrastructure and reduce adverse consequences, are therefore of significant interest.⁴

Autogenous repair (i.e., self-repair) of leaks, either occurring naturally or which might be engineered to occur more frequently, represents an inexpensive approach to reducing or stopping some water pipe leaks in potable water systems.⁴⁻⁵ Ironically, metallic corrosion which is a major cause of pipeline leaks in the first place, can sometimes self-seal leaks by clogging holes with corrosion rust.^{4,6-8} Tang *et al.* (2015) successfully reproduced autogenous repair of 150–400 μm leaks in the copper and iron materials that typically comprise 33–90% of water pipelines, in tests with up to 40 psi water pressure and pH 3.0–11.0 in bench scale studies.⁵ Tang and Edwards (2017) then extended these results to pressures up to 55 psi and leak sizes up to 1000 μm diameter using off the shelf pipe materials in both bench and building scale studies.⁸ The leaks repaired at 20–55 psi, could withstand at least 100 psi in many circumstances, verifying field observations that autogenous repairs could be long-lasting.⁵⁻⁹ Smaller leaks in thicker walled pipe at lower water pressures could usually repair themselves more frequently and at faster rates, compared to leaks in thin walled pipes at higher pressures.⁸ The overall results indicate promise for the approach to be implemented in practice, only if the chemistry conditions favoring autogenous repair could be better understood.^{4,10-12} Even if chemistry was not intentionally manipulated to improve outcomes, it is desirable to ensure that engineering practices do not unintentionally undermine the ability of pipes to self-repair in practice.

Autogenous metallic pipe leak repair has been hypothesized to be a function of water chemistry, including water pH, ionic strength, alkalinity, silica, magnesium, natural organic matter (NOM), dissolved oxygen (DO), disinfectants, corrosion inhibitors and Ryznar Stability Index (RSI) (Table 4-1).⁴ The possible impact of each parameter on autogenous metallic pipe leak repair was described in Tang *et al.* 2013.⁴

Briefly, water chemistry such as ionic strength and water pH could influence autogenous metallic pipe leak repair by changing the size of water-formed rust particles and particle surface charge.^{4,5,13-15} In addition, NOM and corrosion inhibitors (e.g., orthophosphate and tripolyphosphate) could also be influential, by decreasing the formation of scale or rust that clogs leak holes, or by altering the size of water suspended rust particles.^{4,6,16-26} For autogenous repair of concrete, concentrations of silica, magnesium and bicarbonate were important as they became components of materials clogging leaks—effects could occur for rust compounds that can clog metallic pipe leak holes.^{4,6-7} Higher levels of DO and disinfectants including chlorine, chloramine and chlorine dioxide create an oxidizing environment, which can both increase the rate of metal corrosion and leak hole formation on the one hand, but also increase the rate of rust formation that could ultimately cause self-repair on the other.^{4,27-36} Similarly, Ryznar Stability Index (RSI) is developed based on corrosion rate and film formation in iron pipes,³⁷⁻³⁸ and a higher RSI might ultimately

indicate both more nascent leaks and a greater likelihood of clogging repairs—the net effect might depend on which of these two effects is dominant.

Table 4-1. The potential impact of the tested water chemistry parameters on autogenous repair of copper and carbon steel pipe leaks (\pm = mixed impact on repair; + = increase of repair likelihood; ? = unknown).

Water parameter ²	Potential Impact on autogenous leak repair		Test condition	References
	CS	Cu		
pH	\pm	\pm	15 mg/L NaCl as Cl ⁻ , 30 mg/L alk, 7.5 mg/L DO, pH 3.0 to 11.0, w/o a 1 μ m filter ¹	5, 7, 25-26
Ionic strength (IS)	\pm	\pm	IS 2.6 mM: 20 mg/L NaCl as Cl ⁻ , 100 mg/L alk, 7.5 mg/L DO	15
			IS 5.0 mM: 75 mg/L Cl ⁻ , 100 mg/L alk, 7.5 mg/L DO (Control) ¹	
			IS 14.1 mM: 280 mg/L Cl ⁻ , 100 mg/L alk, 7.5 mg/L DO	
			IS 26.9 mM: 650 mg/L Cl ⁻ , 100 mg/L alk, 7.5 mg/L DO	
Alkalinity (Alk)	\pm	\pm	50 mg/L alk: 110 mg/L Cl ⁻ , 50 mg/L alk, 7.5 mg/L DO	18-19
			200 mg/L alk: 10 mg/L Cl ⁻ , 200 mg/L alk, 7.5 mg/L DO	
Silica (Si)	\pm	\pm	4 mg/L Si: 60 mg/L Cl ⁻ , 4 mg/L Si, 100 mg/L alk, 7.5 mg/L DO	17-20
			12 mg/L Si :30 mg/L Cl ⁻ , 12 mg/L Si, 100 mg/L alk, 7.5 mg/L DO	
			20 mg/L Si: 8 mg/L Cl ⁻ , 20 mg/L Si, 100 mg/L alk, 7.5 mg/L DO ¹	
Magnesium (Mg)	?	+	1 mg/L Mg: 70 mg/L Cl ⁻ , 1 mg/L Mg ²⁺ , 100 mg/L alk, 7.5 mg/L DO	6-7
			10 mg/L Mg: 60 mg/L Cl ⁻ , 10 mg/L Mg ²⁺ , 100 mg/L alk, 7.5 mg/L DO	
			18 mg/L Mg: 53 mg/L Cl ⁻ , 18 mg/L Mg ²⁺ , 100 mg/L alk, 7.5 mg/L DO	
			28 mg/L Mg: 80 mg/L Cl ⁻ , 28 mg/L Mg ²⁺ , 100 mg/L alk, 7.5 mg/L DO	
Natural organic matter (NOM)	\pm	\pm	2.5 mg/L NOM: 85 mg/L Cl ⁻ , 2.5 mg/L NOM, 100 mg/L alk, 7.5 mg/L DO ¹	16, 21-24
Dissolved oxygen (DO)	+	+	0.5 mg/L DO: 75 mg/L Cl ⁻ , 100 mg/L alk, 0.5 mg/L DO ¹	4, 26, 29, 32
Disinfectant	\pm	\pm	0.8 mg/L chlorine dioxide (ChD): 75 mg/L Cl ⁻ , 0.8 mg/L ChD, 100 mg/L alk, 7.5 mg/L DO ¹	31-36
			4 mg/L chloramine (ChM): 75 mg/L Cl ⁻ , 4 mg/L ChM, 100 mg/L alk, 7.5 mg/L DO ¹	
			4 mg/L free chlorine (Ch): 75 mg/L Cl ⁻ , 4 mg/L Ch, 100 mg/L alk, 7.5 mg/L DO ¹	
Corrosion inhibitor	\pm	\pm	1 mg/L orthophosphate (OP) as P: 75 mg/L Cl ⁻ , 1 mg/L OP as P, 100 mg/L alk, 7.5 mg/L DO ¹	17, 20, 25-26
			0.5 mg/L tripolyphosphate (PP) as P: 75 mg/L Cl ⁻ , 0.5 mg/L PP as P, 100 mg/L alk, 7.5 mg/L DO ¹	
Ryznar Stability Index (RSI)	+	+	RSI 9.8: 100 mg/L Cl ⁻ , 13 mg/L Ca ²⁺ , 26 mg/L alk, 0.5 mM HCO ₃ ⁻ , 20.2°C	37-38
			RSI 7.9: 350 mg/L Cl ⁻ , 81 mg/L Ca ²⁺ , 54 mg/L alk, 1.0 mM HCO ₃ ⁻ , 20.3°C	
			RSI 7.3: 750 mg/L Cl ⁻ , 150 mg/L Ca ²⁺ , 116 mg/L alk, 2.0 mM HCO ₃ ⁻ , 20.3°C	

Alk = alkalinity in mg/L CaCO₃, CS = carbon steel.

¹Examined with both carbon steel and copper pipe leaks, with other conditions examined with only carbon steel pipe leaks.

²Only water pH examined in Phase 1 in which 150 and 400 μ m leaks in both carbon steel and copper pipes were tested at a 20 psi water pressure, with other water chemistry parameters examined in Phase 2 in which copper pipe 200 μ m leaks and carbon steel 400 μ m leaks were tested at also 20 psi.

The objectives of this study were to 1) examine the impact of various water chemistry parameters (pH, presence of particles, ionic strength, alkalinity, silica, magnesium, NOM, DO, disinfectants, corrosion inhibitors and RSI) on the time it takes to seal pre-formed carbon steel and copper pipe leaks; 2) study the impact of particulate corrosion rust suspended in the water, on the time it takes to repair pipe leaks or the likelihood of self-repair; and 3) test the strength of the materials sealing the leak hole after it has clogged.

EXPERIMENTAL PROCEDURES

Experimental Apparatus

The experimental apparatus, experimental protocol and maintenance procedures in Tang *et al.* 2015 and Tang and Edwards (2017) were used, with modifications described below.

Specifically, synthesized potable water was pumped from a 21 liter reservoir through triplicate 2" (50.8 mm) long carbon steel or copper pipes in a recirculation system at a minimum water distribution pressure of 20 psi (Figure 4-1).⁵ A leak-hole was pre-drilled in the center of each pipe and all the holes were oriented downwards, to represent the 30–40% water leaks that are in contact with bulk drinking water.^{5,8,12,39} The leaked water from each leak-hole was routed back to the reservoir to maintain a constant water volume of 21 liters.⁵ A simulated pressure tank, a pressure gauge and a needle valve were utilized to maintain the system at a minimum water distribution system pressure of 20 ± 5 psi.^{5,8-9}

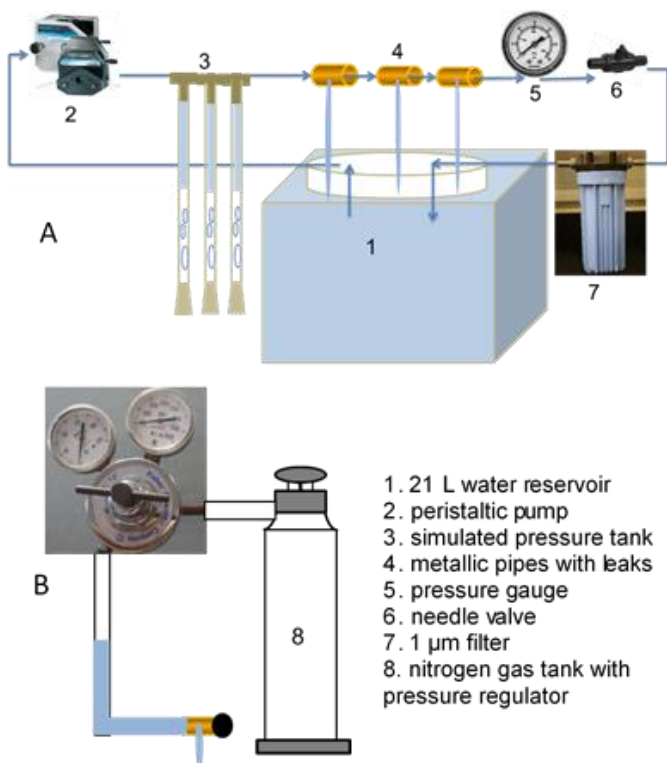


Figure 4-1. Experimental apparatus of the recirculation potable water system with a 1 μm filter (A) and the pressure test apparatus (B) (Adapted with the permission of Tang *et al.* 2015, Copyright 2015 American Chemical Society).

Water Chemistry Parameters

Phase 1. pH

The reservoir water had a baseline water chemistry composed of 15 mg/L Cl^- from sodium chloride (NaCl) and 30 mg/L alkalinity as CaCO_3 at water pHs of 3.0–11.0 as in previous studies described in Tang *et al.* 2015 (Table 4-1).⁵ The target pH was maintained within ± 0.3 units with 0.2 M sodium hydroxide solution and 1 M nitric acid solution, as measured by a pH meter (Model Oakton 11 series).^{5,8} The alkalinity was confirmed by titration per EPA Method 310.1.⁴⁰ Conditions below pH 5.5 in an open system were not able to keep the target alkalinity above the threshold due to stripping of CO_2 , and the condition above pH 11.0 achieved the target alkalinity without addition of sodium bicarbonate due the presence of hydroxide ions.⁵ The water had a relatively high level of DO (7.5 mg/L DO) as confirmed by a DO meter (model Oakton DO 110 Series), and the baseline condition had no disinfectants or phosphate based corrosion inhibitors. The carbon steel and copper pipes leaks (150 and 400 μm diameter) were chosen to represent the medium to large leaks in small diameter building plumbing system tubes observed in field studies.⁵⁻⁶ In some tests a 1 μm filter was installed at the end of the recirculation system to examine the impact of constantly removing large waterformed or waterborne particles which might help clog the leaks (Figure 4-1).⁸ The water was changed on a weekly basis. A pH that could induce a high likelihood of autogenous metallic pipe leak repair was selected as the baseline water chemistry for Phase 2 experiments quantifying other chemical effects.

Phase 2. Other water chemistry parameters

Carbon steel pipe (6.35 mm OD x 890 μm wall) with 400 μm diameter drilled leaks representing a large building plumbing nascent leak size that induces high likelihood of self-repair were tested in a wide range of water chemistries (Table 4-1).^{5-6,8,41} The control water was composed of 75 mg/L Cl^- from NaCl, 100 mg/L alkalinity as CaCO_3 and 7.5 mg/L DO at pH 8.5, without any other added water constituents. The copper pipe (6.35 mm OD x 890 μm wall) with a 200 μm diameter size were examined only in selected conditions based on the carbon steel pipe results.⁸ The water was changed on a weekly basis.

The ionic strength of the control and other waters was kept at ~ 5.0 mM unless stated otherwise, as confirmed by measuring electrical conductivity (EC) according to Standard Method 2510 (EUTECH Instrument waterproof model ECTestr 11+).⁴² The NOM was prepared as described in Zhang (2015) and confirmed by a total organic carbon (TOC) analyzer (Sievers InnovOx TOC Analyzer with a GE Autosampler).²² Chloride, silica, magnesium and phosphorus target concentrations were confirmed on a Thermo Electron X-Series Inductively Coupled Plasma-Mass Spectrometer (ICP-MS) by collecting 10 mL aliquots from the 21 liter water reservoir after complete mixing. The 10 mL aliquots were acidified with 2% v/v concentrated nitric acid (~ 16 N) according to Standard Method 3125-B.⁴²

The chlorine dioxide stock solution of ~ 500 mg/L was generated by adding 2 L deionized water into a 2L500 SelectiveMicro Pouch. Commercial bleach was used to prepare the free chlorine stock solution of 5000 mg/L. The ammonia stock of 14,000 mg/L was prepared from concentrated (~ 14.8 M) ammonia. The chloramine was generated with a 1:1 molar

ratio of ammonia and free chlorine. The actual concentrations of chlorine dioxide (0.8 ± 0.5 mg/L), chloramine (4 ± 0.5 mg/L), free chlorine (4 ± 0.5 mg/L) in the 21 liter water reservoir were confirmed using a HACH instrument (DR 5000 UV-Vis Spectrophotometer).⁴²

Tests with a low level of DO (< 0.5 mg/L) in the water reservoir confirmed by a DO meter (model Oakton DO 110 Series) after addition of 16 grams of sodium sulfite (Na_2SO_3) oxygen scavenger every week. In this case the ionic strength of the reservoir water increased to ~ 21.2 mM. The RSI of water was calculated with water temperature as measured by a thermometer, alkalinity, calcium concentration and total dissolved solids according to Ryznar (1944) (Equation 1).³⁷⁻³⁸

$$RSI = 2pH_s - pH \quad (\text{Equation 1})$$

Where:

pH_s =calculated pH of saturation for calcium carbonate= $(9.3+A+B)-(C+D)$; pH =actual water pH; $A=\log(\text{TDS}-1)/10$; $B=-13.12\log(T+273)+34.55$, temperature T in °C; $C=\log(\text{calcium hardness})-0.4$, calcium hardness in mg/L as CaCO_3 ; $D=\log(\text{alkalinity})$, alkalinity in mg/L as CaCO_3 .

Hole Leakage And Repair Rates

The leakage rate from each triplicate leak-hole was tracked by measuring the instantaneous fill rate of an appropriately sized graduate cylinder on a daily basis.^{5,8} A leakage rate of zero maintained for at least 72 hours was used to indicate the success of the autogenous repair.⁴

Particulate Metal In Water

The total and dissolved ($0.45 \mu\text{m}$) metal (iron or copper) released to each water reservoir as part of the corrosion product was quantified by extracting two 10 mL aliquots from the completely mixed water reservoir immediately before the water change. The metal concentration in both samples was measured by ICP-MS as described above. The difference between the total and the dissolved concentrations calculated the suspended particulate metal concentration from the water-formed corrosion rust particles.

Strength And Characterization Of Repaired Leaks

After a leak was completely repaired, the materials which sealed some leaks were photographed and characterized for elemental composition via environmental scanning electron microscope with an attached X-ray energy dispersive system (ESEM/EDS) (FEI Quanta 600 FEG). The pressure test of the repaired leaks was conducted by increasing the pressure in 5 psi increments from 25 to 100 psi with details described in Tang *et al.* (2015) (Figure 4-1).^{5,8} The maximum pressure that the repaired leak could withstand was designated as the pressure when the water started leaking from the repaired holes.

Weight Loss Rate And Rust Formation Rate

Weight loss of metal pipe and the rust formed on pipe surfaces represent two important manifestations of the corrosion process. To track the weight loss rate and rust formation rate of each metal pipe: 1) the dried (at 103°C for 12 hours) weight of each metal pipe

before the experiment was measured; 2) the dried (at 103°C for 12 hours) weight of each metal pipe after the recirculation experiment and pressure testing was quantified; 3) the rust on interior and exterior pipe surfaces was then removed carefully and the cleaned pipe was dried (at 103°C for 12 hours) and weighed. The weight loss of each metal pipe was the difference between step 1 and step 3, and the rust formed on each pipe surface was the difference between step 2 and step 3. The weight loss rate and rust formation rate were calculated using the following equation (Equation 2):³⁰

$$\text{Corrosion Rate} \left(CR, \frac{\text{mils}}{\text{year}} \right) = \frac{\text{Weight loss or Rust formed (g)} \times k}{\text{Alloy density} \left(\frac{\text{g}}{\text{cm}^3} \right) \times \text{Exposed area (A)} \times \text{Exposure time (hr)}}$$

In which:

$k = 3.45 \times 10^6$; $A = 7.3 \text{ cm}^2$; Alloy density = 8.96 g/cm^3 for copper and 7.85 g/cm^3 for carbon steel; Exposure time represented the experimental time.

Statistical tests

Parametric statistics including a one-way Anova test were used to compare the time to repair for normally distributed data whereas non-parametric statistics including Wilcoxon test and Kruskal Wallis test were used for non-normally distributed data.⁴³⁻⁴⁴ The proportion test was employed to compare the fraction of repaired leaks and the Spearman's test to examine the correlation between time to repair or fraction of repaired leaks and particulate metal concentration, weight loss rate and rust formation rate. A p value of < 0.05 with an alpha value (α) of 0.05 was selected to determine the statistical significance.

RESULTS AND DISCUSSION

Phase 1. pH

Carbon steel pipe leaks. When water was not passed through a 1 μm filter, carbon steel pipe leaks of 150–400 μm diameter had a 100% repair rate at pH from 4.0–11.0, as indicated by measured leak flow reduction to zero (Figure 4-2, A and C).^{5,8} All the 150 μm leaks at pH 5.5–11.0 autogenously had repaired with corrosion rust within 18 days whereas the triplicate 150 μm leaks at pH 4.0 self-repaired after 115 days (Figure 4-2, A).⁴⁻⁵ The repair took longer in the higher acidity water as expected. Due to the large variability of time to repair between replicates, the repair speed at each pH was not statistically different (Kruskal Wallis test, $p > 0.05$). However, the same size leaks at pH 2.5 were on a path to failure and one leak diameter became $\sim 20 \text{ X}$ the initial size within 5 days, indicating that pH 4 could be a threshold pH below which autogenous repair might not occur (Figure 4-2, E).^{5,8} All of the 400 μm leaks at pH 4.0–11.0 were also repaired after 22 days and the time to repair was not statistically different at each pH (Wilcoxon test, $p > 0.05$), and the only exception was one leak at pH 3.0 which became 25% larger after 3 months.^{5,8}

When water recirculated through the pipes was passed through a 1 μm pore size filter, all of the 150–400 μm leaks autogenously repaired after 33 days at water pH from 4.0–11.0 (Figure 4-2, B, D).⁸ Though the time to repair in some filtered conditions increased by up to 6.7 times versus the unfiltered conditions such as at pH 11.0, the absence of large water-formed particles was not statistically different from when they were present (Wilcoxon test, $p > 0.05$), consistent with previous observations.⁸

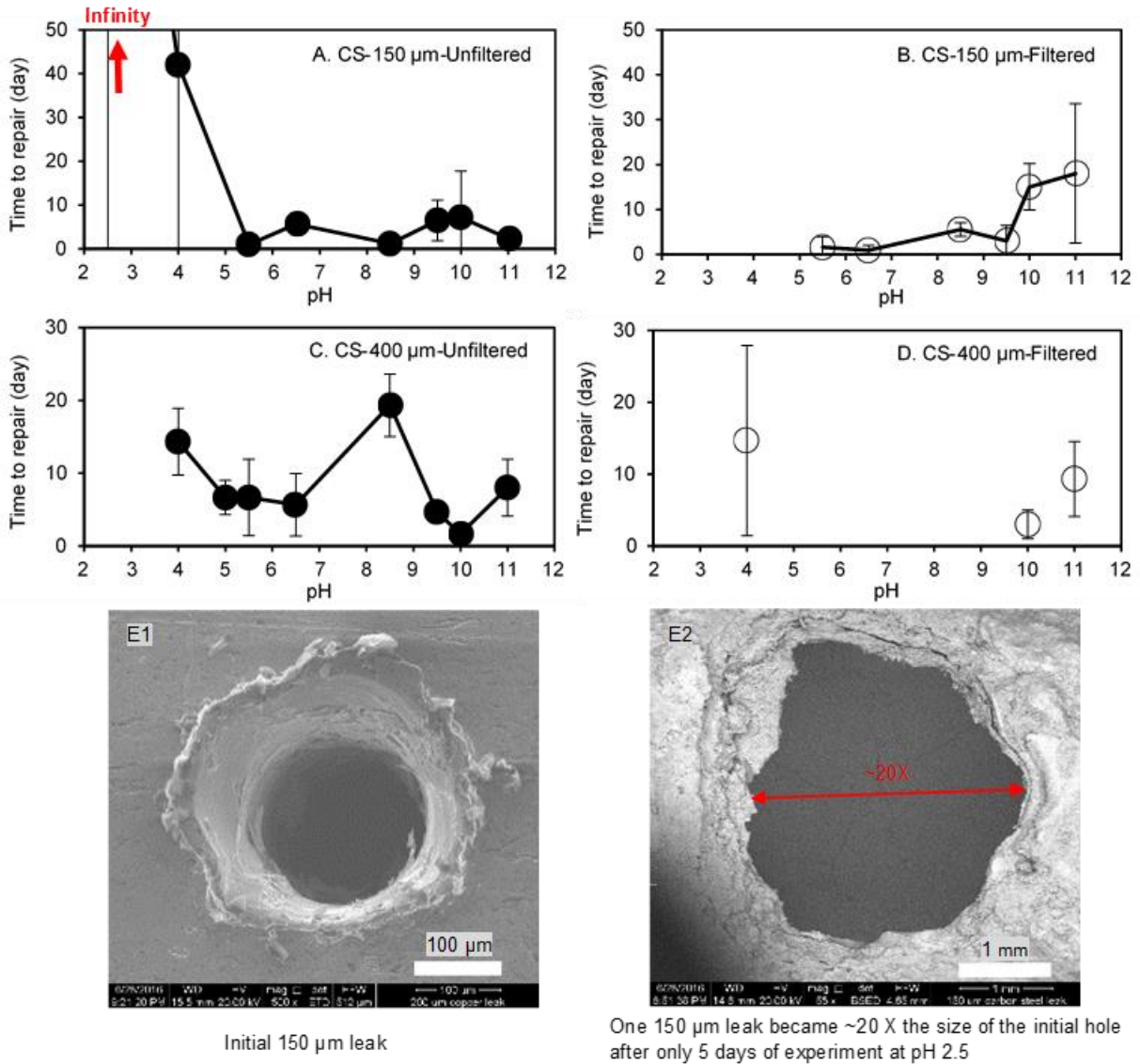


Figure 4-2. The time to repair of 150 and 400 μm leaks in carbon steel (CS) pipes in both unfiltered (A, C) and filtered conditions (B, D) as a function of pH. The error bars show 95% confidence intervals. The triplicate 150 μm leaks (E1) at pH 2.5 all went on a path to complete failure after only 5 days of experiment. One 150 μm leak diameter enlarged ~20X and was shown as an example (E2) (total leak number N=3 for all examined conditions).

Copper pipe leaks. When water was not passed through a 1 μm filter, copper pipe 150 μm leaks had a 100% repair rate at pH from 3.0–11.0. In addition, the repair speed at each pH was statistically the same (Kruskal Wallis test, $p > 0.05$) (Figure 4-3, A).^{5,8} However, the larger leaks of 400 μm had a much smaller repair likelihood over the duration of this work (Figure 4-3, B). Only two out of 21 leaks were successfully repaired for the examined conditions after a 141-day experiment.^{5,8} Specifically, one 400 μm leak at pH 8.5 and one at pH 10.0 self-repaired. Compared to the same size carbon steel leaks, the repair rate (9.5%) of copper 400 μm leaks was 90% smaller and the repair of copper pipe 400 μm

leaks was much slower during the 141-day experiment. This indicated that leak size was a major limiting factor for autogenous repair of copper pipes leaks as indicated in Tang and Edwards (2017).^{5,8}

The time to repair for copper 150 μm leaks in filtered conditions was statistically higher than that in unfiltered conditions (Wilcoxon test, $p < 0.05$) (Figure 4-3, A).⁸ On average, about 1–5.5 times more time was needed for 150 μm leaks to self-repair in filtered conditions versus unfiltered conditions. Therefore, the presence of large particles in water was beneficial in the copper pipe repair of 150 μm leaks, possibly due to a 10X faster corrosion rate of iron versus copper mitigating the impact of large particles on small iron pipe leak repair.^{5,8,13,47}

Compared to 150 μm leaks all of which self-repaired at pH 5.5–11.0 at similar speed (Wilcoxon test, $p > 0.05$), the repair rate was smaller for larger leaks of 400 μm (Figure 4-3, B).^{5,8} Only four out of 15 leaks in the higher size range were successfully repaired, with two at pH 8.5, one at pH 9.5 and one at pH 10.0.

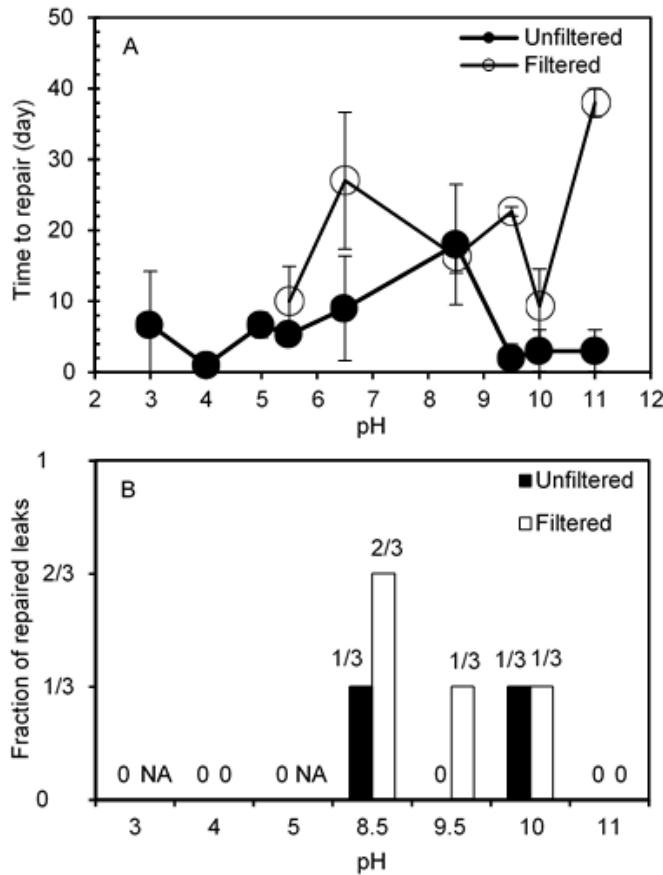


Figure 4-3. The time to repair of 150 μm leaks (A) and fraction of repaired 400 μm leaks (B) in copper pipes in both unfiltered and filtered conditions as a function of pH. The error bars show 95% confidence intervals (total leak number $N = 9$ for 150 μm leaks at pH 8.5 in unfiltered conditions and 3 for other examined conditions; NA=not available).

Selection of water pH. All the carbon steel 150–400 μm and copper 150 μm leaks self-repaired at pH 8.5.^{5,8} In addition, the 400 μm copper pipe leaks in both filtered and unfiltered conditions had the highest repair rate of 33–67% at pH 8.5.^{5,8} Thus, water pH of 8.5 was selected for Phase 2.

Phase 2. Other Water Chemistry Parameters

Autogenous repair of carbon steel pipe leaks

Time to repair. The time to repair carbon steel 400 μm diameter leaks varied amongst different examined water chemistry parameters as described in the following paragraphs (Figure 4-4, A).⁴⁻⁵

Control. The average time to repair the triplicate 400 μm leaks was 15 days and the sealing materials in these repaired leaks were mainly composed of iron corrosion rust, as indicated by ESEM/EDS and previous studies (Figure 4-5).^{5,7-8} For example, the sealing materials of one repaired leak was composed of 24% oxygen and 76% iron on a normalized mass percent basis, representing a typical iron rust composition.²⁷

Ionic strength. The average time to repair of the 400 μm leaks at the lowest water ionic strength of 2.6 mM was 19 days, 4 days more than that at ionic strengths of 5.0 mM (control) and 26.9 mM, and 9 days more than that at ionic strength of 14.1 mM. The time to repair among the four examined ionic strength levels was not statistically different (One-way ANOVA test, $p > 0.05$), due to the large variations in time to repair for amongst the triplicate leaks at each condition.

Alkalinity. Alkalinity had mixed effects on the average time to repair. The average time to repair of 400 μm leaks at alkalinity of 100 mg/L as CaCO_3 was 15 days (control), 8 days more than that at alkalinity of 50 mg/L as CaCO_3 and 10 days more than that at alkalinity of 200 mg/L as CaCO_3 . The time to repair among the three examined alkalinity levels was not statistically different (Kruskal Wallis test, $p > 0.05$).

Silica. Silica had mixed effects on average repair speed of carbon steel pipe 400 μm leaks. Compared to the control condition (no silica added) of 15 days, the time to repair of three 400 μm leaks decreased by 6 days when a concentration of 4 mg/L silica was added to water, but increased by 4 days when a concentration of 12 mg/L silica was dosed. At the highest level of 20 mg/L silica, the average time to repair was 13 days, 87% of that in the control condition. The time to repair among the four examined silica levels was not statistically different (Kruskal Wallis test, $p > 0.05$).

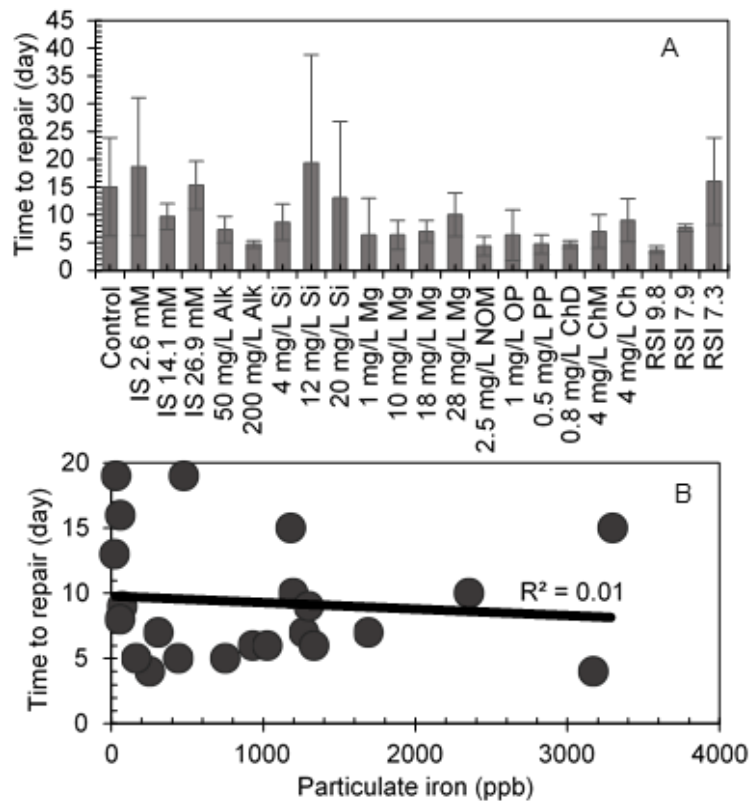


Figure 4-4. The time to repair (A) of 400 μm leaks in carbon steel pipes as a function of different water chemistry parameters was not correlated with waterformed corrosion rust suspended in the water (B). The error bars show a 95% confidence intervals (total leak number $N = 6$ for control and 3 for other examined conditions; IS = ionic strength; alk = alkalinity; OP=orthophosphate; PP=tripolyphosphate; ChD=chlorine dioxide; ChM=chloramine; Ch= free chlorine).

Magnesium. The addition of magnesium to water promoted average autogenous repair speed. Specifically, the time to repair of all 400 μm leaks at magnesium levels ranging from 1–28 mg/L decreased by 53–67% or 5–9 days compared to the control (no magnesium) time of 15 days. But the time to repair among the five examined magnesium levels was not statistically different (Kruskal Wallis test, $p > 0.05$).

NOM. The average time to repair decreased by 73% or 11 days with the presence of 2.5 mg/L NOM in water compared to that in the control condition (no NOM), but the time to repair of the two examined NOM levels was not statistically different (Wilcoxon test, $p > 0.05$).

Corrosion inhibitors. The addition of 1 mg/L orthophosphate or 0.5 mg/L polyphosphate as P enhanced the speed of repair. Specifically, the average time to repair decreased by 58–69% or 9–10 days with the addition of corrosion inhibitors versus the control (no corrosion inhibitors) time of 15 days. But the time to repair among the three examined corrosion inhibitor levels was not statistically different (Kruskal Wallis test, $p > 0.05$).

Disinfectants. The addition of disinfectants decreased the time to repair versus the control (no disinfectants) time of 15 days. The time to repair decreased by 67% with the addition of 0.8 mg/L chlorine dioxide, 53% with 4 mg/L chloramine and 40% with 4 mg/L free chlorine. However, the time to repair among the four disinfectant levels was not statistically different (Kruskal Wallis test, $p > 0.05$).

DO. When a low level of < 0.5 mg/L DO was maintained in the reservoir water, three out of six leaks autogenously repaired after 72 days during the 104-day experiment (data not shown). In contrast, all six 400 μm leaks in water with 7.5 mg/L DO (control) repaired themselves with an average repair of 15 days (Figure 4-4, A). The time to repair at 7.5 mg/L DO was statistically faster than at < 0.5 mg/L DO (Wilcoxon test, $p < 0.05$), indicating that DO was a critical factor in autogenous repair of carbon steel pipe leaks.⁴

RSI. The leaks with more corrosive water as indicated by higher RSI,³⁷⁻³⁸ autogenously repaired more quickly on average (Kruskal Wallis test, $p < 0.05$). The average time to repair of all three 400 μm leaks at RSI of 7.3 (significantly corrosive) was the highest of 16 days, and it decreased to 8 days at RSI of 7.9 (heavy corrosion), and to 4 days at RSI of 9.8 (intolerably corrosive). This affirms that higher corrosivity can sometimes have slight benefits, as it could partly serve to repair pipe leaks. In some previous field studies, pipes observed to be riddled with fully penetrating pinholes, were never detected as leaking due to very high incidence of rapid self-repair.⁴⁻⁷

In summary, the repair rate of carbon steel 400 μm leaks for a wide range of water chemistries was relatively high in that 96% (72/75) of the leaks were autogenously repaired at pH 8.5. The only exception was the three leaks at low levels of < 0.5 mg/L DO. While DO and RSI were two statistically important factors influencing the speed of autogenous repair, the presence of the other examined chemical constituents did not statistically affect the time to repair of carbon steel pipe 400 μm leaks significantly.

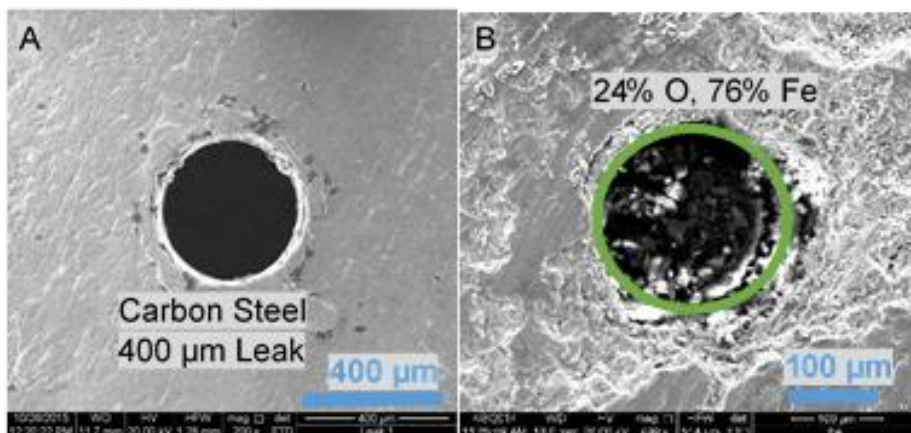


Figure 4-5. The sealing materials of a 400 μm leak (A) that autogenously repaired in the control condition were composed of corrosion rust (24% oxygen and 76% iron on a normalized mass percent basis) (B).

Association between time to repair and particulate iron in water. The water-formed particulate iron from corrosion rust varied among the examined water chemistries as measured by ICP-MS (Figure 4-4, B).

More water-formed particulate iron was associated with higher ionic strength and lower alkalinity.^{15,18} Specifically, the particulate iron levels increased from 480 to 3,290 ppb as ionic strength increased from 2.6 to 26.9 mM. In contrast, the particulate iron levels decreased from 1,260 to 440 ppb when the alkalinity was increased from 50 to 200 mg/L as CaCO₃.

The presence of silica, NOM or tripolyphosphate resulted in smaller amount of water-formed iron particles.^{16-17,21-23} Compared to the 1,170 ppb particulate iron in the control condition, only 22 ppb (1.9% of the control) particulate iron were present with 20 mg/L silica in water, 250 ppb (21.2%) with 2.5 mg/L NOM and 740 ppb (63.2%) with 0.5 mg/L tripolyphosphate. However, the presence of 1 mg/L orthophosphate as P resulted in a 14% increase in iron particle concentrations compared to the control (1,170 ppb).

The presence of magnesium had mixed effects on the particulate iron levels in water. Compared to the 1,170 ppb in the control condition, the particulate level of 250 ppb (72.8% lower) was the lowest with 18 mg/L Mg and the highest at 1,200 ppb (3.0% higher) with 28 mg/L Mg.

Disinfectants increased the pipe corrosion rate as expected,³¹⁻³⁶ since total iron released to water with disinfectants increased by 14–86% compared to the control. However, disinfectants had mixed effects on particulate iron formed in water. Specifically, chloramine and free chlorine increased the particulate iron by 12–45% versus the control while chlorine dioxide decreased the particulate iron by 86%.

Higher DO increased the water-formed iron rust via redox reactions,^{26,29,32} as the particulate iron in water with < 0.5 mg/L DO decreased by 69% versus water with 7.5 mg/L (1,170 ppb).

The more corrosive water as indicated by larger RSI increased the iron rust formed in water. Specifically, the higher RSI of 9.8 led to more particulate iron of 3,160 ppb, 49 times that at RSI of 7.9 and 52.5 times that at RSI of 7.3.

However, the average time to repair of carbon steel pipe 400 µm leaks was not correlated with the average water-formed particulate iron in water (Spearman's, $\rho = -0.03$; DO data excluded as only 3/6 leaks were repaired) (Figure 4-4, B). It indicated that the abundance of the water-formed particulate iron in water as influenced by various water chemistries did not influence the speed of repair.

Association between time to repair and weight loss rate. As the average time to repair of the carbon steel pipe 400 µm leaks was not correlated with water-formed particulate iron, only pipe leaks examined in a few water chemistries known to inhibit or increase iron pipe corrosion were examined for metal release rate and rust formation rate, including corrosion inhibitors, disinfectants and NOM (Figure 4-6).^{21-23,25-28}

The average pipe weight loss in control conditions was 38.8 mils/year, 2.1% lower than that with 4 mg/L free chlorine, but 0.2–220% higher than that in other examined conditions (Figure 4-6, A-B). Despite the variations among the seven conditions, the weight loss of each examined parameter was not statistically different from that in the control condition (Wilcoxon test, $p > 0.05$).

In addition, the time to repair was not correlated with the weight loss rate (Spearman's, $\rho = 0.27$), indicating that the metal loss as influenced by water chemistry did not affect the time to repair for 400 μm size leaks in carbon steel pipes significantly.

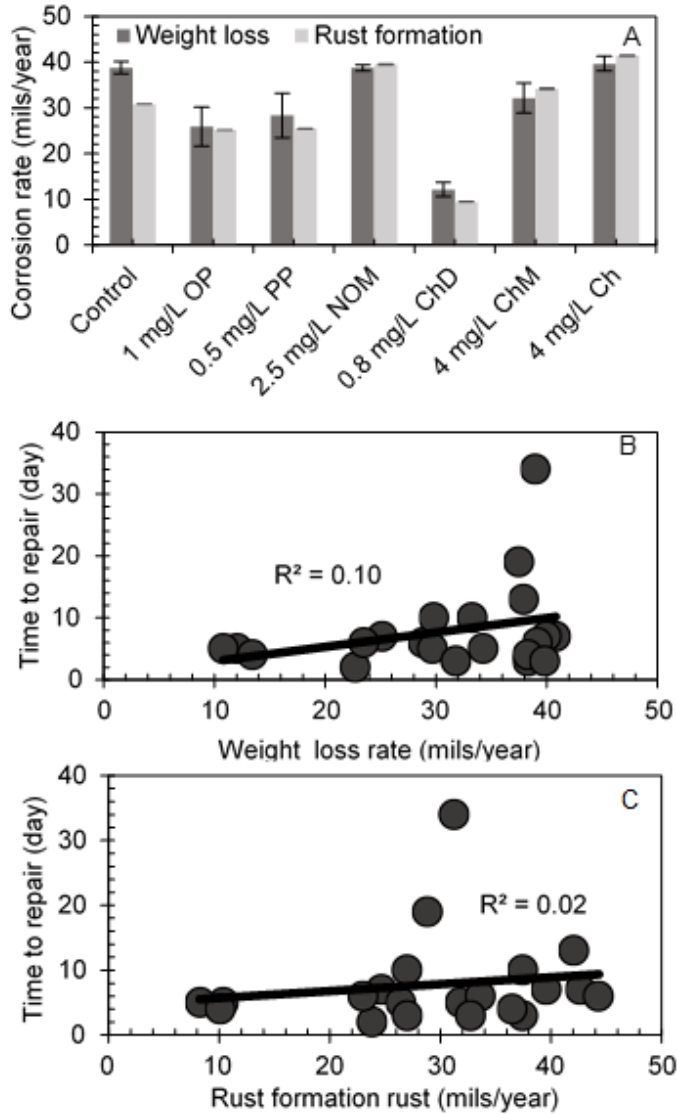


Figure 4-6. The 400 μm leaks in carbon steel pipes in selected water chemistries were examined for weight loss and gained rust rate (A) and the time to repair was not correlated with weight loss rate (B) and rust formation rate (C). The error bars show 95% confidence intervals (OP=orthophosphate; PP=tripolyphosphate; ChD=chlorine dioxide; ChM=chloramine; Ch= free chlorine).

Association between time to repair and rust formation rate. The rust formation rate on pipe surfaces was of a similar magnitude to the pipe weight loss rate, which ranged from 9.5–41.4 mils/year (Figure 4-6, A). The rust formation rate decreased by 17.5–69.1% in the orthophosphate, tripolyphosphate and chlorine dioxide conditions versus the 30.8 mils/year in the control condition. In contrast, the rust formation rate of the remaining three conditions (i.e., NOM, chloramine and chlorine dioxide) increased by 10.9–34.4%. However, the rust formation rate of each examined water chemistry condition again was not statistically different from the control condition (Wilcoxon test, $p > 0.05$). Thus, the time to repair was not correlated with the rust formation rate as influenced by water chemistry (Spearman's, $\rho = 0.30$) (Figure 4-6, C), demonstrating that the formation of rust on pipe surfaces did not speed up the remediation of the 400 μm size leaks in carbon steel pipes.

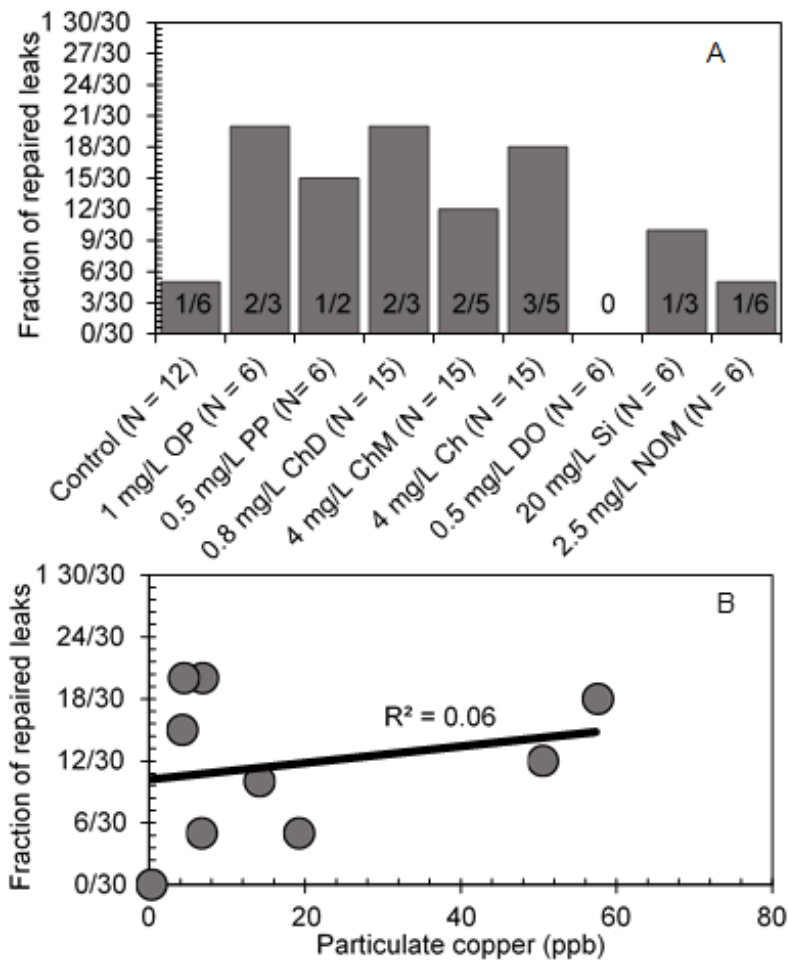


Figure 4-7. The time to repair of 200 μm leaks in copper pipes as a function of different water chemistry parameters (A) was independent of waterformed corrosion rust (B) (N = total leak number; OP=orthophosphate; PP=tripolyphosphate; ChD=chlorine dioxide; ChM=chloramine; Ch= free chlorine).

Autogenous Repair Of Copper Pipe Leaks

Since the time to repair of 400 μm leaks in carbon steel pipe was not strongly influenced by various water chemistry parameters except DO and RSI, a few parameters that were known to inhibit or promote copper pipe corrosion were selected to examine their effect on copper pipe 200 μm leaks including corrosion inhibitors, disinfectants, DO, silica and NOM.^{7,19-20,24,30-31}

Fraction of repaired leaks. As at least some of the 200 μm leaks in copper pipes for each examined condition were not repaired as indicated by the reduction of leakage rate after 2 months of experiment (data not shown),⁴⁻⁵ it was not possible to calculate the average time to repair, and the fraction of repaired leaks was employed to represent differences in the remediation rate in the nine examined chemistries (Figure 4-7, A).

Control. Out of the twelve examined 200 μm leaks, only two were successfully repaired after 2 month of experiments.

Disinfectants. The presence of disinfectants increased the fraction of repaired leaks. Six out of fifteen 200 μm leaks autogenously repaired with 4 mg/L chloramine in water, which was 1.4 times more than that (2/12) in the control condition. But the increase was not statistically significant as determined by a proportion test ($p > 0.05$).

Interestingly, three extreme cases as indicated by the change in leakage rate, were observed in the presence of 4 mg/L chloramine (Figure 4-8, A). One 200 μm leak (Figure 4-8, B1-B2) was remediated with a typical copper corrosion rust composition (28% C, 11.3% O, 1.1% Al, 1.2% Si and 58.4% Cu on a normalized mass percent basis) as indicated by the reduction of leakage rate from 45.7 L/d to zero after 23 days. However, one leak (Figure 4-8, B3) became approximately 1.1 times larger in size as the leakage rate increased from 51.8 to 158.4 L/d. One leak was partially repaired with copper rust as the leakage rate was reduced by 72% (Figure 4-8, B4). Thus, in this condition, the destiny of an individual leak, even in terms of growing or sealing the leak-hole, was higher variable.

Additionally, nine and ten out of fifteen 200 μm leaks in water with 4 mg/L free chlorine and 0.8 mg/L chlorine dioxide autogenously repaired, respectively. They were statistically the same as the fraction of repaired leaks for the control (2/12) (Proportion test, $p > 0.05$).

Corrosion inhibitors. A higher fraction of repaired leaks was observed in the presence of corrosion inhibitors versus the control. Specifically, 67% (4/6) of the 200 μm leaks self-repaired with 1 mg/L orthophosphate as P and 50% (3/6) repaired themselves with 0.5 mg/L tripolyphosphate as P, 4 and 3 times the fraction in control, respectively. The presence of corrosion inhibitors did not yield a statistically significant difference from the control (Proportion test, $p > 0.05$).

NOM. The presence of 2.5 mg/L NOM did not influence the fraction of repaired leaks as the fraction of repaired leaks was the same as that in the control condition (Proportion test, $p > 0.05$).

Silica. A higher fraction of repaired leaks was observed in water with a higher level of silica. Two out of six 200 μm leaks were repaired when 20 mg/L silica was added to water, two times the level in control, but it was not significantly different from the control (Proportion test, $p > 0.05$).

DO. The presence of DO increased the fraction of repaired leaks. Two out of twelve 200 μm leaks were repaired with 7.5 mg/L DO whereas none of the six 200 μm leaks was repaired with < 0.5 mg/L DO in water. Again, the increase was not statistically significant (Proportion test, $p > 0.05$).

In summary, the addition of free chlorine, chloramine, corrosion inhibitors, silica, NOM, and DO increased the fraction of repaired leaks by 1–3 times. However, the increase compared to the control was not statistically significant. In contrast, the presence of chlorine dioxide significantly increased the fraction of repaired copper pipe 200 μm leaks by 3 times.

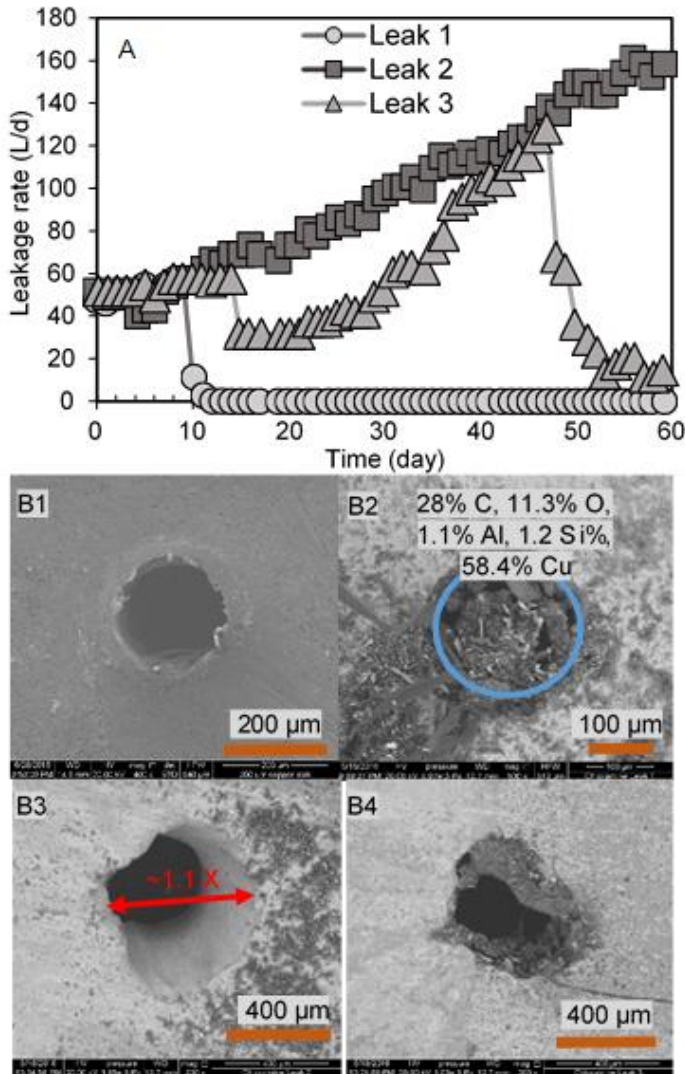


Figure 4-8. In the examined chloramine condition (A), one 200 μm leak (B1) was repaired with copper corrosion rust (28% carbon, 11.3% oxygen, 1.1% alumina, 1.2% silica and 58.4% copper on a normalized mass percent basis) as indicated by reduction of leakage rate to zero (B2, leak 1) while one failed as the leak increased by ~ 1.1 times in size (B3, leak 2) and one leak was only partially repaired (B4, leak 3).

Association between fraction of repaired leaks and particulate copper. The particulate copper in water varied from 4.1–57 ppb among the nine water chemistry conditions (Figure 4-7, B). Similar to carbon steel 400 μm leaks, the fraction of repaired copper pipe 200 μm leaks was not correlated with the particulate copper formed in water (Spearman's, $\rho = -0.13$). Thus, the remediation rate or fraction of repaired copper pipe 200 μm leaks was not influenced by the water-formed copper rust as water chemistry varied, possibly due to their low concentrations.

However, the enhanced fraction ($\sim 2.1\text{X}$) of repaired leaks for carbon steel leaks (30/33) versus copper pipe leaks (37/87) might be attributed to the 7.8–1,650 times more particulate metal formed from the faster metal corrosion in the carbon steel conditions, as it created a better likelihood of leak clogging.^{27,47}

Association between fraction of repaired leaks and weight loss rate. After 2 months, the weight loss rate of the copper pipe leaks varied from 0.26–1.34 mils/year (Figure 4-9, A-B). The average weight loss rate (0.29 mils/year) with 1 mg/L orthophosphate as P was the same as that (0.37 mils/year) in the control condition (Wilcoxon test, $p > 0.05$) while the rate (0.26 mils/year) with 20 mg/L silica was 30% lower (Wilcoxon test, $p < 0.05$). In contrast, the presence of polyphosphate, NOM, disinfectants and DO increased the weight loss rate significantly compared to the control (Wilcoxon test, $p < 0.05$).

These results showed that silica inhibited copper pipe corrosion effectively by reducing copper loss, contrary to the trend for orthophosphate, polyphosphate and NOM.^{12,27} Disinfectants increased copper loss as they are chemically reactive with copper pipes as an oxidizing agent in water.^{24,26,32,34} Interestingly, the average weight loss (0.64 mils/year) in water with < 0.5 mg/L DO was statistically higher than that with 7.5 mg/L DO (Wilcoxon test, $p < 0.05$). It might be due to the fact that the addition of sodium sulfite to water to maintain the low level of DO dramatically increased the ionic strength of water, and therefore the corrosivity of water to copper.^{4,21}

However, the fraction of repaired leaks was not highly correlated with the weight loss rate of copper pipe (Spearman's, $\rho = 0.37$). This implies that the remediation rate of copper pipe leaks was not influenced by the pipe weight loss rate as controlled by water chemistry.

Association between fraction of repaired leaks and rust formation rate. The average rust formation rate on copper pipe surfaces ranged from 0.15–1.03 mils/year and was of similar magnitude to the weight loss rate (Figure 4-9, A, C). The rust formation rate with disinfectants (0.52–1.03 mils/year) was statistically higher than that of the control condition (0.20 mils/year) (Wilcoxon test, $p < 0.05$). In contrast, the average rust formation rate in all the other examined parameters (corrosion inhibitors, NOM, DO and silica) ranged from 0.15–0.22 mils/year and was statistically the same as that of control (Wilcoxon test, $p > 0.05$). Therefore, disinfectants reactive with copper pipes promoted corrosion rate, resulting in an increase of the rust formation, with other parameters (i.e., corrosion inhibitors, NOM, low DO and silica) uninfluential in rust formation.^{24,26}

Similarly, the fraction of repaired leaks was not correlated with the copper pipe rust formation rate (Spearman's, $\rho = 0.22$). It is speculated that the rust formed in leak-holes due to the change of water chemistry was not significant such that the fraction of repaired was not affected.

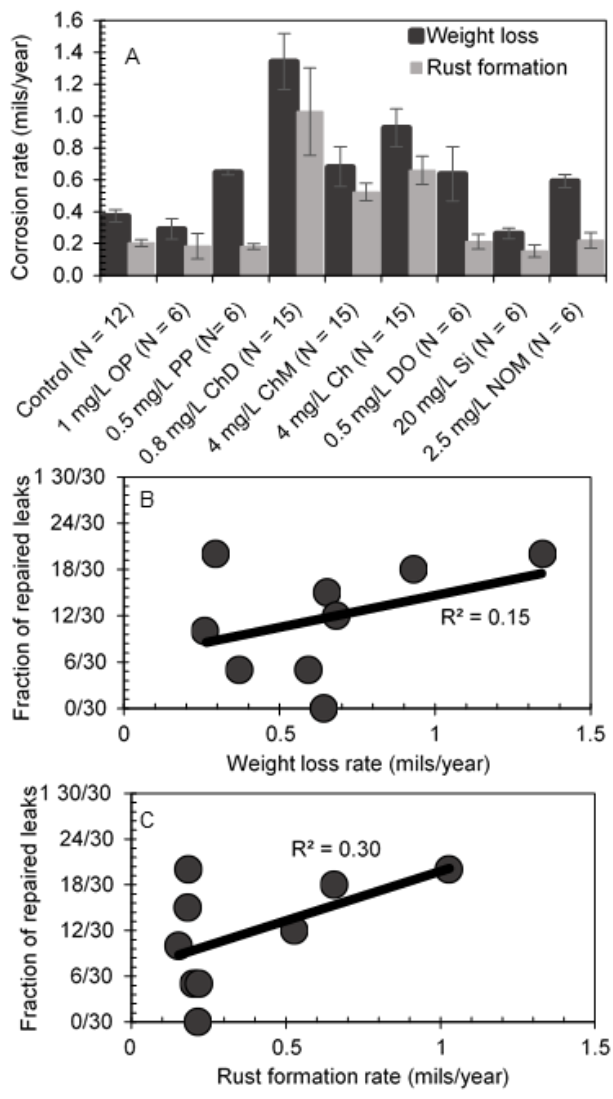


Figure 4-9. The 200 μm leaks in copper pipes in selected water chemistries were examined for weight loss and rust formation rate (A) and the time to repair was not correlated with weight loss rate (B) and rust formation rate (C). The error bars show 95% confidence intervals (N = total leak number; OP=orthophosphate; PP=tripolyphosphate; ChD=chlorine dioxide; ChM=chloramine; Ch= free chlorine).

Strength of Repaired Leaks

Strength of repaired copper pipe leaks. The sealing materials in copper pipe leaks were relatively strong and resilient, consistent with previous field and research studies.⁵⁻⁶ Specifically, 37% of the copper leaks that were repaired at 20 psi could withstand a household plumbing pressure of 55 psi which is deemed typical.^{5,48} About 15% of the repaired copper pipe leaks could withstand a considerably high household plumbing pressure of 100 psi.^{5,9} However, 49% of the repaired leaks could only withstand 25 psi. This might be due to the fact that the strength of the sealing materials was compromised

by the disassembly or installation of the pressure testing apparatus, which was also previously noted to occur during the testing of carbon steel pipe leaks.^{5,8}

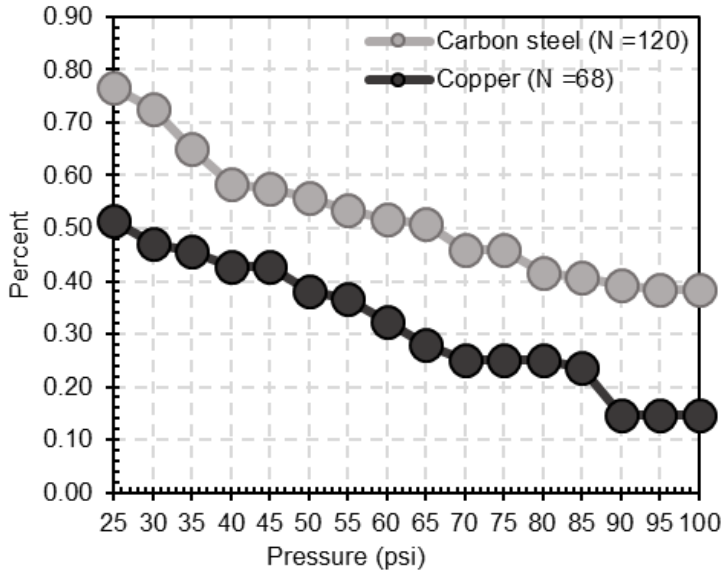


Figure 4-10. The percent of repaired leaks in carbon steel and copper pipes that withstood the examined pressure before leaking (N = total repaired leak number examined in pressure testing).

Strength of repaired carbon steel pipe leaks. The strength of repaired carbon steel pipe leaks was even stronger, in that 53% of the leaks repaired at 20 psi could withstand a typical household plumbing pressure of 55 psi and only 23% of the repaired leaks failed at 25 psi (Wilcoxon test, $p < 0.05$).^{5,48} In addition, 38% of the repaired carbon steel leaks could withstand a considerably high plumbing pressure of 100 psi.^{5,9}

The weaker sealing materials in copper pipe leaks might reflect the fact that the magnitude of the particulate iron concentration was 7.8-1,650 times more than the particulate copper at the same water chemistry conditions. Moreover, the weight loss rate or the rust formation rate of carbon steel pipe was 8–180 times more than that of copper pipe. This indicates that, although the corrosion of both copper and iron pipe as influenced by various water chemistries did not affect the time to repair or the fraction of repaired leaks with statistically significant differences, the stronger repair of carbon steel leaks might be associated with a larger volume of corrosion rust formed in leak-holes due to the enhanced corrosion rate of carbon steel pipes.^{5,27,48}

CONCLUSIONS

Studies of autogenous leak hole repair, considered representative of leaks occurring in smaller diameter building plumbing systems, yielded the following conclusions:

- The presence of large ($> 1 \mu\text{m}$) suspended particles of corrosion by-products significantly decreased the time to repair of copper 150 μm leaks at pH from 3.0–11.0.
- Ionic strength, alkalinity, magnesium, NOM, silica, disinfectants and corrosion inhibitors did not significantly influence the time to repair of carbon steel 400 μm leaks at pH 8.5, whereas higher DO and RSI significantly decreased the time to repair by 50–70%.
- DO, corrosion inhibitors, silica, NOM, free chlorine and chloramine were not influential in altering the fraction of repaired copper pipe 200 μm leaks at pH 8.5. The only exception was chlorine dioxide, which significantly increased the fraction of repaired leaks by 3 times.
- The time to repair of carbon steel 400 μm leaks or the fraction of repaired copper pipe 200 μm leaks at pH 8.5 was not correlated with the particulate metal concentration, weight loss rate or rust formation rate.
- The strength of sealing corrosion rust in some autogenously repaired copper and carbon steel pipes was strong, confirming the great potential of the autogenous repair approach to extend the lifetime of aging water infrastructure.

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CHAPTER 5. IN-SITU REMEDIATION OF IRON PIPE LEAKS WITH CALCIUM CARBONATE, SILICA AND WOOD ASH PARTICLES IN POTABLE WATER SYSTEMS

Min Tang, Fei Wang, Marc Edwards

ABSTRACT

Our aging and leaking potable water infrastructure creates water loss, property damage and public health risks. In-situ clogging of leaks by waterborne or waterformed particles might be used to extend the lifetime of these vital assets. Addition of calcium carbonate particles (~8.8 μm), silica particles (~29 μm) and wood ash particles (~160 μm) that were favored by Roman engineers, increased the fraction of remediated leaks in iron pipes to 1/12, 10/12 and 10/12, respectively, compared to that in the control condition without any particles (0/12). Smaller leaks were remediated with a higher success rate over the 58 day duration of this experiment, as indicated by calcium carbonate which had 33% success for 280 μm leaks versus 0% success for 400–1000 μm leaks; silica which had 100% success for 280–400 μm leaks versus 67% for 700–1000 μm leaks; and wood ash which had 100% success for 280–700 μm leaks versus 67% for 1000 μm leaks. The strength of the leak clogging was strong, since 27–73% of the remediated leaks could withstand a 100 psi pressure depending on particle type.

KEYWORDS: Physical clogging; iron pipe leak remediation; silica; calcium carbonate; wood ash

INTRODUCTION

Much of the water infrastructure in the United States is on the verge of failure and is presently leaking about 7 billion gallons of water per day (ACSE, 2009; Folkman *et al.*, 2012). The American Water Works Association believes that at least 1 trillion dollars will be needed in the coming decades to upgrade our existing water infrastructure and ensure safe drinking water (Shanaghan, 2012). The traditional approach of pipe repair, rehabilitation and replacement are effective, but they are very costly and will take decades to implement (Tang *et al.*, 2013). New transformative approaches are needed to remediate water pipe leaks and/or extend the lifetime of our water distribution systems. *In situ* remediation of potable water pipe leaks is a possible low cost and convenient approach to reducing water pipe leaks (Shanaghan, 2012; Tang *et al.*, 2013; Tang *et al.*, 2015; Tang and Edwards 2017).

Clogging Of Leaks By Waterformed/Waterborne Particles

Successful clogging of pipe leaks with waterformed or waterborne particles such as calcium carbonate or silica in potable water systems (Figure 5-1) (Hearn and Morley, 1997; Hearn, 1998; Letterman *et al.*, 2008; Parks *et al.*, 2010; Tang *et al.*, 2013; Tang *et al.*, 2015), originated in Ancient Rome (\approx 15 BC), where wood ash particles permanently sealed leaks in new terracotta pressure pipelines (Pollio 15 BC as translated by Morgan, 1960; Etiegni and Campbell, 1991; Misra *et al.*, 1993; Pitman, 2006; Tang *et al.*, 2013). The approach was lost for almost 2000 years, before Letterman *et al.* (2008) and others re-discovered the idea, and explored its possible application for repair of concrete pipe cracks in the Delaware Aqueduct.

Aside from the purposeful use of particles to actively clog leaks, it is hypothetically possible that some of our problems with declining and leaking water infrastructure, may be an unintended consequence of well-intentioned efforts to improve public health and reduce aesthetic concerns, by minimizing the amount of particles entering water distribution systems (Table 5-1) (EPA, 1986; Tang *et al.*, 2013). Various primary regulations of the United States Environmental Protection Agency (U.S. EPA) now limit turbidity in potable water (EPA, 1986)—compliance with such regulations might reduce the likelihood of leak repair that once occurred from particles naturally present in our potable water systems.

A range of particles might be naturally present in, or purposely introduced to, potable water systems and physically clog leaks (Table 5-1). Calcium carbonate (CaCO_3) particles are naturally present in drinking water and may also form in water mains and household plumbing system when water chemistry is favorable or water temperatures are increased (Montgomery, 1985; Edzwald, 2011; Brazeau and Edwards, 2011). Letterman *et al.* (2008) introduced CaCO_3 particles to reduce the water leak rate through simulated concrete crack leaks by 55% within 2.5 hours in their bench scale study. Silica (SiO_2) particles are also naturally present in drinking water and might contribute to physical clogging (Rushing *et al.*, 2003; Tang *et al.*, 2013), and treatment processes such as ballasted sedimentation and sand filtration can contribute silica fines to drinking water (Davis *et al.*, 2001; Edzwald, 2011). Other naturally occurring particles, such as wood ash favored by Romans, are

generated by combustion and are composed of lime, calcite, portlandite and calcium silicate - the percentage of each compound varies dependent on circumstance (Etiegni and Campbell, 1991; Misra *et al.*, 1993).

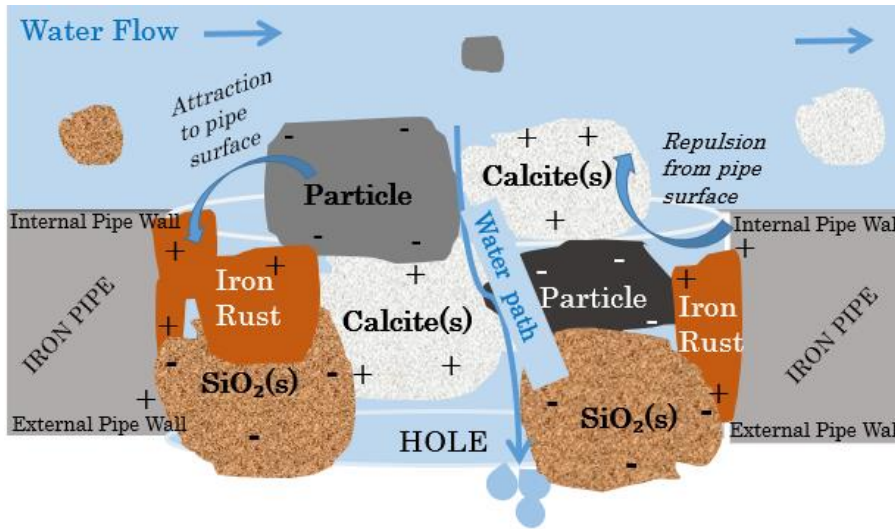


Figure 5-1. Physical clogging via sieving, diffusion and charge attraction/repulsion could remediate iron water pipe leaks by waterborne and waterformed particles in potable water systems, such as iron rust, silica ($\text{SiO}_2(\text{s})$) and calcium carbonate ($\text{CaCO}_3(\text{s})$) (Adapted with permission of Tang *et al.* 2013).

Physical And Chemical Parameters Affect Leak Remediation

Particle size is expected to affect the likelihood of repair via physical clogging, which involves entrapment of particles by sieving, diffusion and charge attraction/repulsion forces as in the case of filtration (Figure 5-1) (Hearn and Morley, 1997; Benjamin and Lawler 2013; Tang *et al.*, 2013). For example, larger particles are more likely to be trapped in leaks via sieving, resulting in improved blockage and leak remediation. Wood ash particles tend to swell and hydrate in water, potentially increasing the likelihood of remediation (Etiegni and Campbell, 1991).

Table 5-1. Summary of three representative particles in drinking water.

Particle type	Common source in drinking water	Particle concentration/turbidity (g/L/NTU) ¹	Particle diameter ±std (µm) ¹	Zeta potential±std (mV) ¹	Particle stability in water ¹
Calcium carbonate ($\text{CaCO}_3(\text{s})$)	Natural; drinking water treatment (DWT) (softening or pH control); water heating	0.00082	8.8±4.4	+2.0±2.3	Very likely to coagulate (Very unstable)
Silica ($\text{SiO}_2(\text{s})$)	Natural; DWT (ballasted sedimentation and sand filtration)	0.0036	29±27	-15.2±4.9	Likely to coagulate
Wood ash	Not normally present	0.0029	160±140	-11.9±3.4	(Unstable)

(References: Montgomery, 1985; Etiegni and Campbell 1991; Misra *et al.*, 1993; Taylor, 1995; McNeill and Edwards, 2001; Edzwald, 2011; Brazeau and Edwards, 2011; Tang *et al.*, 2013)

¹Measurements and predictions for particles in Blacksburg, VA tap water.

Likewise, water chemistry affects particle size and surface charge, which in turn affects the likelihood of capture and leak blockage as in the case of granular media filtration (Somasundaran and Agar, 1967; Kosmulski, 2009 a and b; Hanaor *et al.*, 2012; Benjamin and Lawler 2013; Tang *et al.*, 2013; Tang *et al.*, 2015). For instance, water at pH_{PZC} might induce higher likelihood of remediation as particles tend to be unstable and grow larger, or tend to attach to the walls of leak holes. The reported pH_{PZC} ranges from 8–10 for CaCO_3 , 2–4 for silica and is around 8 for aged iron pipe surfaces (Taylor, 1995; Kosmulski, 2009 a and b; Edzwald, 2011), whereas no experimental data for pH_{PZC} of wood ash particles could be found. Similarly, the presence of corrosion inhibitors is sometimes influential in leak remediation as they could adsorb to calcium carbonate particles and prevent them from agglomerating or precipitate further (Lin and Singer, 2005).

This study evaluated *in situ* leak remediation via physical clogging by adding calcium carbonate, wood ash, and silica particles to Blacksburg, VA tap water flowing in iron pipes, which are widely used for water mains and service lines. These representative waterborne particulates had a range of zeta potentials and particle sizes, which could affect their ability to repair leaks. The effect of initial leak size (280–1000 μm) on the likelihood and speed of leak repair, and the strength of clogged leaks were also examined.

MATERIALS AND METHODS

Materials

Blacksburg, VA tap water

Blacksburg, VA tap water served as the bulk solution in each pipe of our experimental setup (Figure 5-2). The water typically has a pH of 7.8, alkalinity of 40 mg/L as CaCO_3 , calcium of 10.7 mg/L, chloride of 14.5 mg/L, disinfectant residual chloramine of 3 mg/L and the corrosion inhibitor zinc phosphate with a concentration of 310 ppb as phosphorus (Clark *et al.*, 2013).

Particles

The calcium carbonate particles (> 98% CaCO_3) were purchased from Duda Energy and were at food grade (Figure 5-3). The silica particles (99.5% SiO_2) were provided by U.S. Silica and were produced in Berkeley Springs, WV, as SIL-CO-SIL 75 ground silica. The wood ash was collected from an oak wood campfire at Blacksburg, VA and the wood ash particles were collected through an 840 μm sieve.

Methods

Experimental set-up of galvanized iron coated pipes

Three $\frac{3}{4}$ " OD (19 mm) galvanized iron coated pipes (Allied Tube and Conduit Corporation) were connected to a domestic Blacksburg tap water outlet at pressure of 58 ± 3 psi (Figure 5-2). A one way check valve was installed after the Blacksburg tap water outlet and a ball valve was used to reduce the water pressure to a target pressure of 10 psi, as determined by a pressure gauge (0–100 psi). In each pipe, there were triplicates for all four leak sizes (1000, 700, 400 and 280 μm). All the pre-drilled leaks were oriented upwards, presumably the most difficult leak orientation to clog by particles that are heavier than water, and leaks were placed every 3 inches (7.6 cm) apart (Table 5-1) (Scardina *et*

al., 2008; Tang *et al.*, 2015; Tang and Edwards, 2017). Each leak size was drilled every 1 ft (30.5 cm), with the largest leak size closest to and the smallest leak size furthest from the domestic Blacksburg tap water outlet. All the connecting elbows, couplings and pipes were composed of polyvinyl chloride (PVC) material.

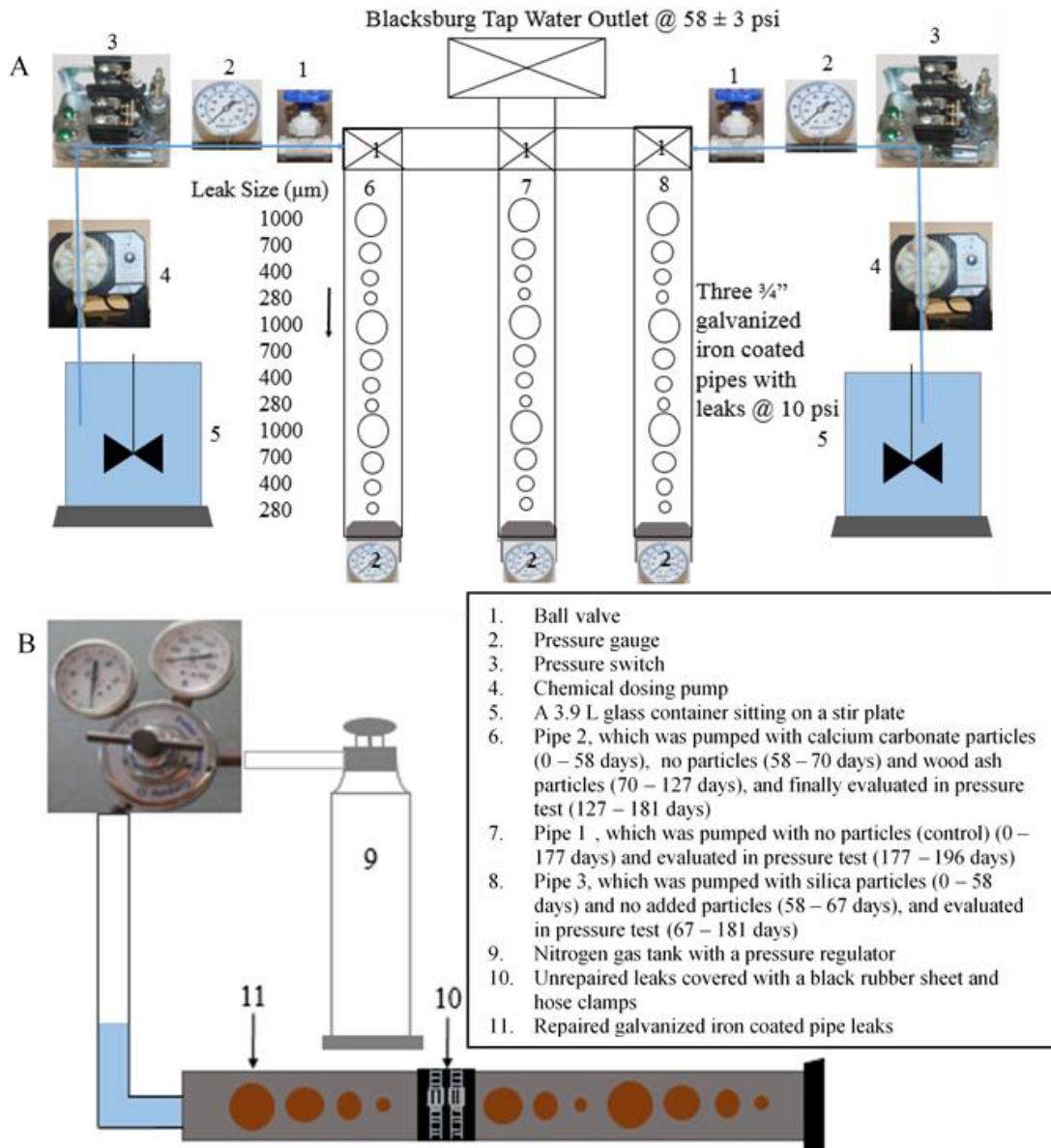


Figure 5-2. Experimental apparatus of three galvanized iron coated pipes with upward leaks that were in-situ remediated with calcium carbonate, wood ash and silica particles at a water pressure of 10 psi (A). The strength of the sealing materials was then examined in a two-phase pressure test: 1) the first phase was conducted in the experimental setup (A) by increasing water pressure up to 60 psi, via adjustment of the ball valve connected to the domestic Blacksburg tap water outlet; 2) the second phase examined the maximum pressure the repaired leaks could withstand using a separate setup (B), by increasing water pressure up to 100 psi.

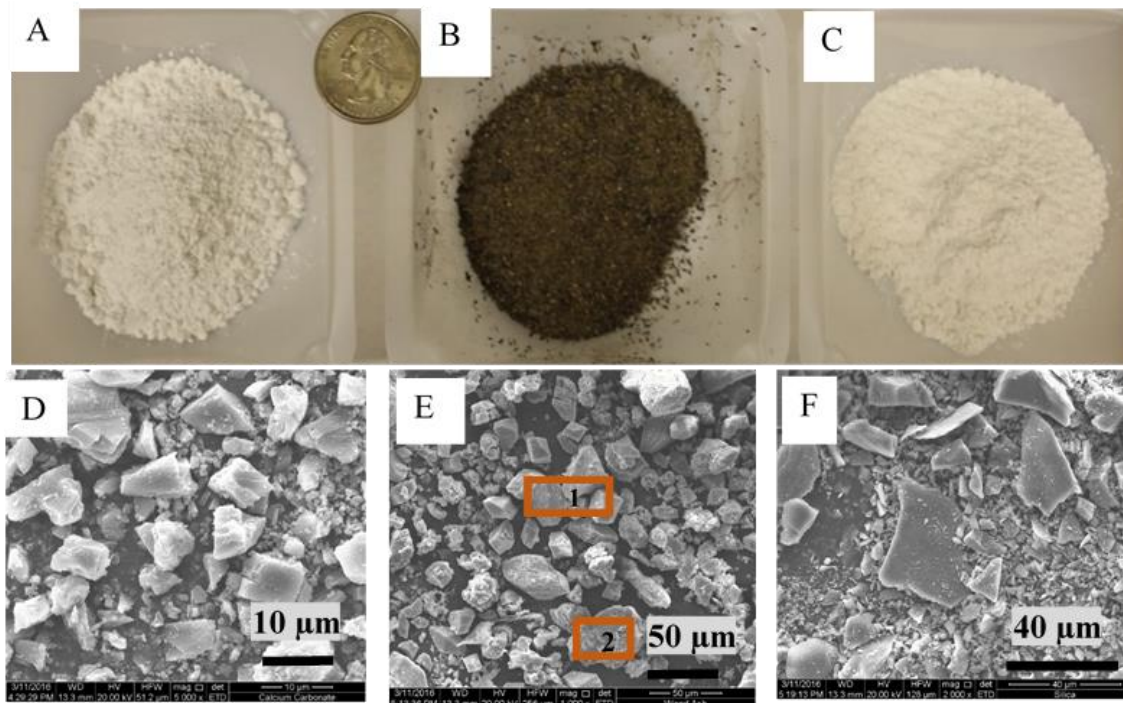


Figure 5-3. Photos and microscope images of the calcium carbonate (A, D), wood ash (B, E) and silica (C, F) particles added to Blacksburg tap water. The composition of wood ash was characterized using ESEM/EDS on a normalized mass percent basis. The wood ash in spot 1 was composed of 8% C, 51% O and 41% Ca and that in spot 2 was composed of 40% O, 2% Mg, 10% Al, 26% Si, 2% P, 11% K and 9% Ca.

Particle feeding system

The calcium carbonate, silica and wood ash particles (Figure 5-3) were mixed with the Blacksburg tap water in the reservoir, before being delivered to two galvanized iron coated pipes (pipe 2 and pipe 3 in Figure 5-2) via a chemical dosing pump (Hanna Instruments BL5-1 Blackstone). As a control condition, no particles were added to the third pipe (pipe 1 in Figure 5-2). To represent potable water with extremely high turbidity, 30–200 grams of calcium carbonate or silica particles were added to 3.9 liter of Blacksburg tap water in a 3.9 liter glass container every day (Figure B1). In contrast, to represent potable water with moderately high turbidity, only 0–15 grams of wood ash particles were added each day to 3.9 L Blacksburg tap water. The particle solutions were constantly mixed using a stir bar, resulting in an average water pH of 8.4, 8.1 and 8.2 for calcium carbonate, silica and wood ash particle solutions, respectively.

For pipe 1, the Blacksburg tap water was pumped through the pipe without any added particles for 177 days, as a control condition (Figure 5-2). For pipe 2, calcium carbonate particles were added during the first 58 days, and then the pipe was flushed with only Blacksburg tap water for 12 days to clean the particulate debris. Afterwards, it was pumped with wood ash particles from day 70–127. For pipe 3, silica particles were pumped from

day 0–58. The particle feeding system also stayed at a water pressure of 10 psi, as confirmed by a pressure switch and a pressure gauge connected to the domestic outlet.

Particle size, crystal composition and zeta potential

The particle size distribution of calcium carbonate, silica and wood ash particles in Blacksburg tap water was analyzed by a particle sizer (Model HORIBA LA 300). The zeta potential of particles in Blacksburg tap water was measured by a Zetameter (Model Zeta-Meter System 3.0) (Table 5-1). The crystal composition of calcium carbonate, silica and wood ash was analyzed by X-Ray Diffraction (Model Panalytical X'Pert 3 Powder). The surface morphology of these particles was also examined using an environmental scanning electron microscope with an attached X-ray energy dispersive system (ESEM/EDS) (FEI Quanta 600 FEG).

Turbidity in water

The turbidities of the water leaking from the leak-holes that were closest to and furthest from the domestic Blacksburg tap water outlet were measured daily using a Turbidimeter (HACH Model 2100N Turbidimeter), and were denoted as the influent turbidity and effluent turbidity, respectively.

Leakage rate

The leakage rate (L/day) from leak-holes was measured in a graduated cylinder on a daily basis (Tang *et al.*, 2015; Tang and Edwards, 2017). The success of in-situ remediation was defined as the point that the leakage rate reduced to zero (Tang *et al.*, 2013; Tang *et al.*, 2015; Tang and Edwards, 2017).

Pressure test

Two phases of pressure tests were conducted after completion of leak repair (Figure 5-2). In Phase 1, the pressure test was conducted by increasing water pressure in 10 psi increments every 3 days up to 60 psi (Figure 5-2, A). The pressure test was conducted from day 177 to 196 for the control (pipe 1), from day 127 to 181 for the pipe remediated with calcium carbonate and wood ash particles (pipe 2) and from day 67 to 181 for the pipe remediated with wood silica particles (pipe 3).

In Phase 2, all three pipes were disconnected from the domestic Blacksburg tap water outlet and then connected to a separate pressure test apparatus (Figure 5-2, B). The unrepaired leaks and the broken leaks in the first phase of the pressure test were covered with black rubber and stainless steel hose clamps. Then, these pipes were filled with water and capped at one end. The pressure test was conducted by pumping nitrogen gas from the other end of the pipes and gradually increasing pressure from 10 psi up to 100 psi in 5 psi increments. The maximum pressure the repaired leaks could withstand was defined as the pressure at which water started leaking from the repaired holes in both phases.

Characterization of remediated leaks

After two phases of pressure testing, the remediated leaks that did not fail at 100 psi were photographed and the sealing materials were examined under ESEM/EDS (FEI Quanta 600 FEG) as in Tang *et al.* (2015) and Tang and Edwards (2017).

RESULTS AND DISCUSSIONS

Remediation of Galvanized Iron Coated Pipe Leaks

Natural Remediation of Iron Pipe Leaks Without Added Particles (control condition). The fraction of remediated leaks (i.e., remediation rate) for the control condition was a function of reaction time between the leak and water flowing in the iron pipes at a water pressure of 10 psi (Figure 5-4, A; Figure B2). None of the twelve leaks was remediated after 58 days, while seven of twelve leaks remediated themselves at the end of 177-day experiment as defined by the point at which reduction of leakage rate fell to zero (Figure 5-4, A). For instance, the leakage rate of a 280 μm leak started from 43 L/day and gradually decreased to zero after 87 days. Similarly, the leakage rate of a 400 μm leak was initially 72 L/day and reduced to zero after 107 days (Figure 5-4, B). This confirms prior research demonstrating that iron pipes have a natural capability for self-repair even without particulates (Tang *et al.*, 2015; Tang and Edwards, 2017). In contrast, one 700 μm and one 1000 μm leak were only partially remediated during the 177-day experiment (Figure 5-4, B).

Overall, smaller leaks (280 and 400 μm) had a larger remediation rate compared to larger leaks (700 and 1000 μm). At the end of 177-day experiment, all the 280 (3/3) and 400 μm (3/3) leaks self-remediated but only one out of the three 700 μm leaks and none of the 1000 μm leaks were in-situ remediated.

The reduction in the leakage rates for all leak sizes during the 177-day experiment was probably caused by the increased water turbidity due to particles released from iron pipe corrosion, which has been previously validated for sealing metallic pipe leaks (Scardina *et al.*, 2008; Lytle and Nagadouda, 2010; Tang *et al.*, 2013; Tang *et al.*, 2015; Tang and Edwards, 2017). This was also suggested by the fact that the leaks of the same size further from the Blacksburg outlet always self-remediated faster than those closer to the outlet. The average influent turbidity of the Blacksburg tap water before the iron pipes was 0.18 NTU, which is less than EPA regulations that require less than 1 NTU under all circumstances and 95% of the water samples below 0.3 NTU (EPA, 1986). But as the tap water flowed through the pipe, more particles were present in the water after it contacted with the pipe as indicated by the roughly doubled effluent turbidity of 0.39 NTU.

Remediation with calcium carbonate particles. When calcium carbonate or calcite particles with a mean diameter of 8.8 μm (Figure B3; Figure B4; Table 5-1) were added for 58 days, the average effluent turbidity of the Blacksburg tap water was 344–369 NTU, 69–74 times higher than the immediate EPA action level of 5 NTU and 717–1845 times higher than the control condition (Figure 5-5, A).

Interestingly, this high level (~0.3 g/L at the measured influent/effluent turbidity) of suspended calcite particles did not improve the fraction of remediated leaks significantly versus the control condition after 58 days of experiment (Figure 5-4; Table 5-1). Only one leak (in the size of 280 μm) out of twelve leaks (8.3%) was remediated after 26 days with calcite particles, which was not significantly different from the control condition where none of the twelve leaks was repaired (Figure 5-5, B).

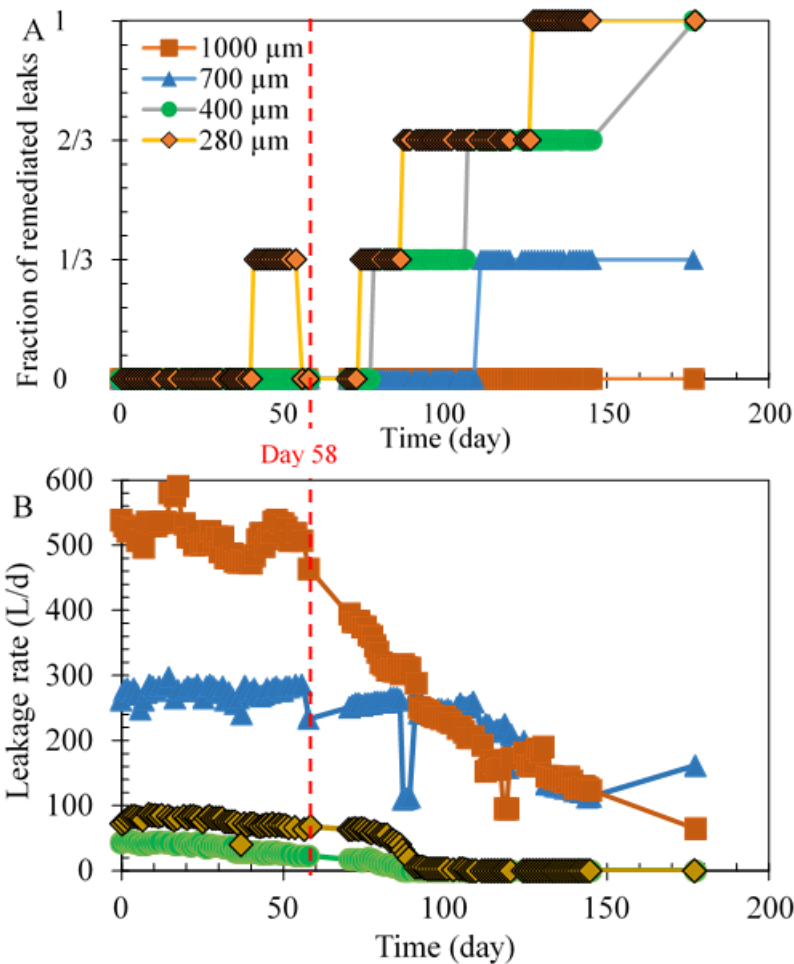


Figure 5-4. The leak repair of four sizes (280, 400, 700, and 1000 μm) in the 177-day control experiment without any added particles (pipe 1). The fraction of remediated leaks of four leak sizes was a function of the reaction time (A) and the leakage rate for all four representative leaks reduced with reaction time (B).

Remediation with silica particles. When larger silica or alpha quartz particles with a mean diameter of 29 μm were continuously dosed to Blacksburg tap water for 58 days, the average zeta potential of silica was -15.2 mV making the particles less likely to coagulate to form large particles, compared to calcium carbonate particles of 2.0 mV (Figure B3; Figure B4; Table 5-1) (Hanaor *et al.*, 2012). However, the leak remediation with silica particles occurred faster and at a significantly higher rate than the control and the condition with calcium carbonate particles during the 58 days of experimentation even though a high average turbidity of 363–383 NTU was measured in water flowing through the pipe (Figure 5-5; Figure B5).

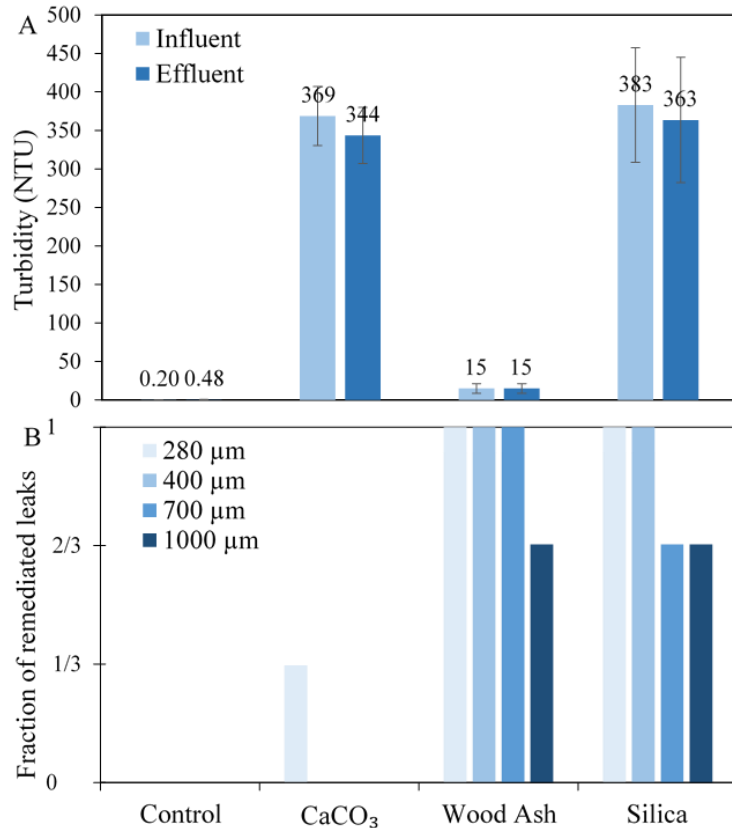


Figure 5-5. The turbidity (A) and the fraction of remediated leaks (B) after adding three types of particles for 58 days. The error bars indicate 95% confidence intervals and the number shown was the average turbidity.

During the first 58 days of the experiment, while none of the twelve leaks was remediated in the control condition, all three 280 μm leaks were remediated after 16 days with silica particles, which was 0.7–12 times faster than the one 280 μm leak remediated with calcium carbonate particles. In addition, all three 400 μm leaks were completely sealed after 9 days. One leak reappeared after 15 days, but was soon permanently re-sealed after another 4 days. Two 700 μm and two 1000 μm leaks also clogged in 30 days.

Consequently, ten out of twelve leaks (83.3%) were successfully remediated with silica particles, 10 times the remediation rate of the calcium carbonate condition (Figure 5-5). The faster and higher remediation rate with silica particles versus calcium carbonate particles is likely due to both the 2.3 times larger particle size and the 3.3 times larger particle concentration (~1.3 g/L) in water, which increased the likelihood of leak blockage (Benjamin and Lawler 2013; Tang *et al.*, 2013).

Remediation with wood ash particles. As only one 280 μm leak was successfully remediated with calcium carbonate particles after 58 days of addition, which was not different from the control (Figure 5-2; Figure 5-5), the success rate of wood ash particles utilized by Roman engineers was examined thereafter using this same apparatus after flushing the apparatus with Blacksburg tap water from day 58 to day 70. The wood ash

particles were fed from day 70 to day 127 and had an average size of 160 μm in Blacksburg tap water (Table 5-1; Figure B4).

The wood ash in this work was composed of mainly calcite and some minor silica particles (Figure B3). Wood ash particles in Blacksburg tap water had an average zeta potential of -11.9 mV, making the wood ash particles more resistant to coagulation and growth than the calcium carbonate (Hanaor *et al.*, 2012) (Table 5-1). Even though the turbidity was 4% of that for the silica and calcium carbonate resulting in a wood ash particle concentration of ~ 0.04 g/L, the remediation capability for wood ash was the highest among three particles (Figure 5-5; Table 5-1).

For example, after only two days of adding wood ash particles to pipe with eleven open leaks (two 280 μm , three 400 μm , three 700 μm and three 1000 μm), two 280 μm , one 400 μm and one 1000 μm leaks were completely sealed (Figure B5). After 23 days of adding wood ash particles, all leaks were sealed except for one 1000 μm leak. After 43 days, the only unrepaired 1000 μm leak was also remediated but another repaired 1000 μm broke open and stayed open until the end of 58-day experiment. Overall, 83.3% (10/12) of the iron leaks were remediated with wood ash particles, 10 times the rate (1/12 or 8.3%) when the same pipe leaks were remediated with calcium carbonate particles and the same as that of the pipe leaks remediated with silica particles. The high remediation rate for all leak sizes with 4.5–17.2X larger but 85–97% fewer wood ash particles versus calcium carbonate and silica, indicates that particle size is crucial in leak blockage especially for larger leak sizes (Benjamin and Lawler 2013; Tang *et al.*, 2013).

Pressure Test

Strength of repair when no particles were added (control, pipe 1). Most remediated leaks remained sealed throughout the test, when the strength of remediated leaks was examined by increasing water pressure in 10 psi increments every three days from 10 psi to 60 psi (the first phase of pressure test), while others broke shortly after the pressure was raised (Figure 5-2; Table B1). For example, the leakage rate for a remediated 280 μm leak remained zero from day 177 to 196 (Figure B6). In contrast, a remediated 400 μm leak broke at 40 psi whereas the un-remediated 700 μm and 1000 μm leaks had increased leakage rate as water pressure increased. In summary, six of the seven repaired leaks stayed sealed in the first phase of pressure test and the only exception was the 400 μm leak which failed at 40 psi.

When the strength of remediated leaks in the same pipe was examined in another pressure test apparatus by increasing water pressure in 5 psi increments up to 100 psi (the second phase of pressure test), one of the six remaining repaired leaks broke due to disturbance of the apparatus (Tang and Edwards, 2017). It also occurred in the pressure test of the other two pipes, and the maximum pressure of this leak was denoted as more than 60 psi.

The repair was relatively strong, as indicated by the results from both phases of pressure test (Figure 5-6, A; Table B1). About 43% (3/7) of the repaired leaks could withstand more than 100 psi, a high home plumbing pressure (LeChevallier *et al.*, 2014). And 86% (6/7) of the leaks could withstand a typical household plumbing pressure of 55 psi (Joyce, 2011).

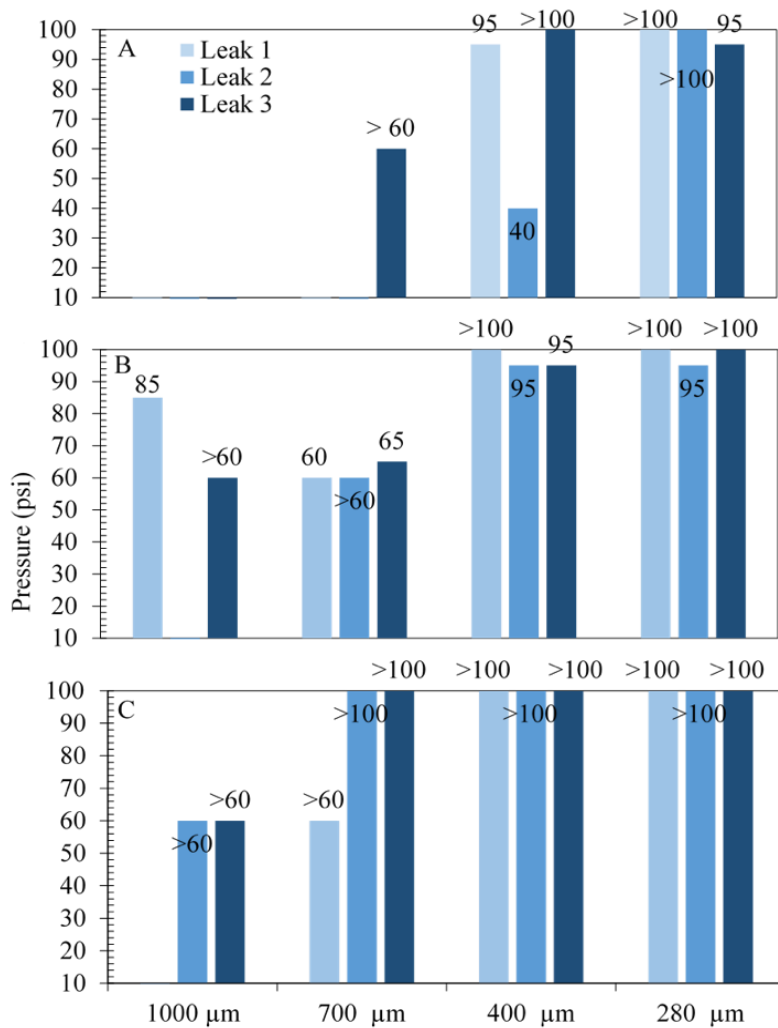


Figure 5-6. The maximum pressure the repaired leaks could withstand before leaking again after the first phase and second phase of pressure test. [A = pipe 1 as control condition; B = pipe 2 with calcium carbonate and wood ash particles; C = pipe 3 with silica particles; >100 psi represents that the leak did not fail at 100 psi after both phases of pressure test; >60 psi represents that the leak did not fail at 60 psi but it failed due to the disturbance of the pipe while preparing it for the pressure test.]

Strength of repair when calcium carbonate and wood ash particles were added (pipe 2).

The strength of repaired materials when calcium carbonate and wood ash particles were added was also relatively strong and resilient (Figure 5-6, B). About 27% (3/11) of the repaired leaks could withstand a high household plumbing pressure of 100 psi and 100% (11/11) of them could withstand a typical household plumbing pressure of 55 psi (Joyce, 2011; LeChevallier *et al.*, 2014).

Strength of repair when silica particles were added (pipe 3). Overall, the sealing materials remediated with silica particles were strong. All eleven repaired leaks (11/11 or 100%)

could withstand a typical household plumbing pressure of 55 psi, and eight of the repaired leaks (8/11 or 73%) could withstand a high house plumbing pressure of 100 psi (Figure 5-6, C) (Joyce, 2011; LeChevallier *et al.*, 2014). The strength of repair in leaks with silica particles was not statistically different from that in the other two pipes possibly due to the limited sample number (Wilcoxon test, $p > 0.05$)

It is worth noting that even though the conventional wisdom suggests that leakage rate from leak-holes increases as water pressure increases (Hiki, 1981; Lambert, 2001; Greyvenstein and Van Zyl, 2005), one un-remediated 700 μm leak with silica particles was remediated at a water pressure of 60 psi via the natural repair process of iron corrosion during the first phase of pressure test (Tang *et al.*, 2013; Tang *et al.*, 2015) (Figure B6). The repaired leak sustained 60 psi and was still fully repaired at the end of the 6-month experiment.

Surface Morphology of the Repaired Leaks

The repaired leaks in all three pipes were covered with rust on both the inside and outside of leaks (Figure 5-7). For example, in the control condition without any added particles, both the outside and inside of a 280 μm leak was covered with rust, and the inside of the leak was blocked with such massive rust that it could not be identified visually (Figure 5-7, A1-A4). After the rust on the inside pipe surface was carefully scratched off by sandpaper, the sealing materials in the leak were identified as a typical iron corrosion rust under ESEM/EDS (Figure 5-7, A4). Specifically, the rust was composed of 21% iron, 72% oxygen and 7% zirconium on a normalized mass percent basis with zirconium as a background element due to coating.

For the pipe remediated with calcium carbonate and wood ash particles, the visual appearance of sealing materials for a 280 μm leak was similar to the control condition, with abundant rust on both inside and outside pipe surfaces (Figure 5-7, B1-B4). The sealing materials in one 280 μm leak after scratching off from the inside pipe surface were composed of 8% carbon, 29% oxygen, 1% silica, 2% calcium and 60% iron on a normalized mass percent basis (Figure 5-7, B2). Thus, the clog itself was mostly iron rust, and it had only a minor amount of either wood ash or calcium carbonate as a constituent.

Similarly, when the iron pipe leaks were remediated with silica particles, the inside scale of a 280 μm leak consisted of 33% oxygen, 4% silicon and 63% iron on a normalized mass percent basis, showing that the remediation was again completed with mostly rust products and some minor silica particles (Figure 5-7, C).

Given the dramatic enhancement of the leak repair rate with either the wood ash or silica, it was somewhat surprising that the leak hole was dominated by iron. It indicates that waterborne particles act to enhance the natural autogenous repair mechanism attributable to iron rust.

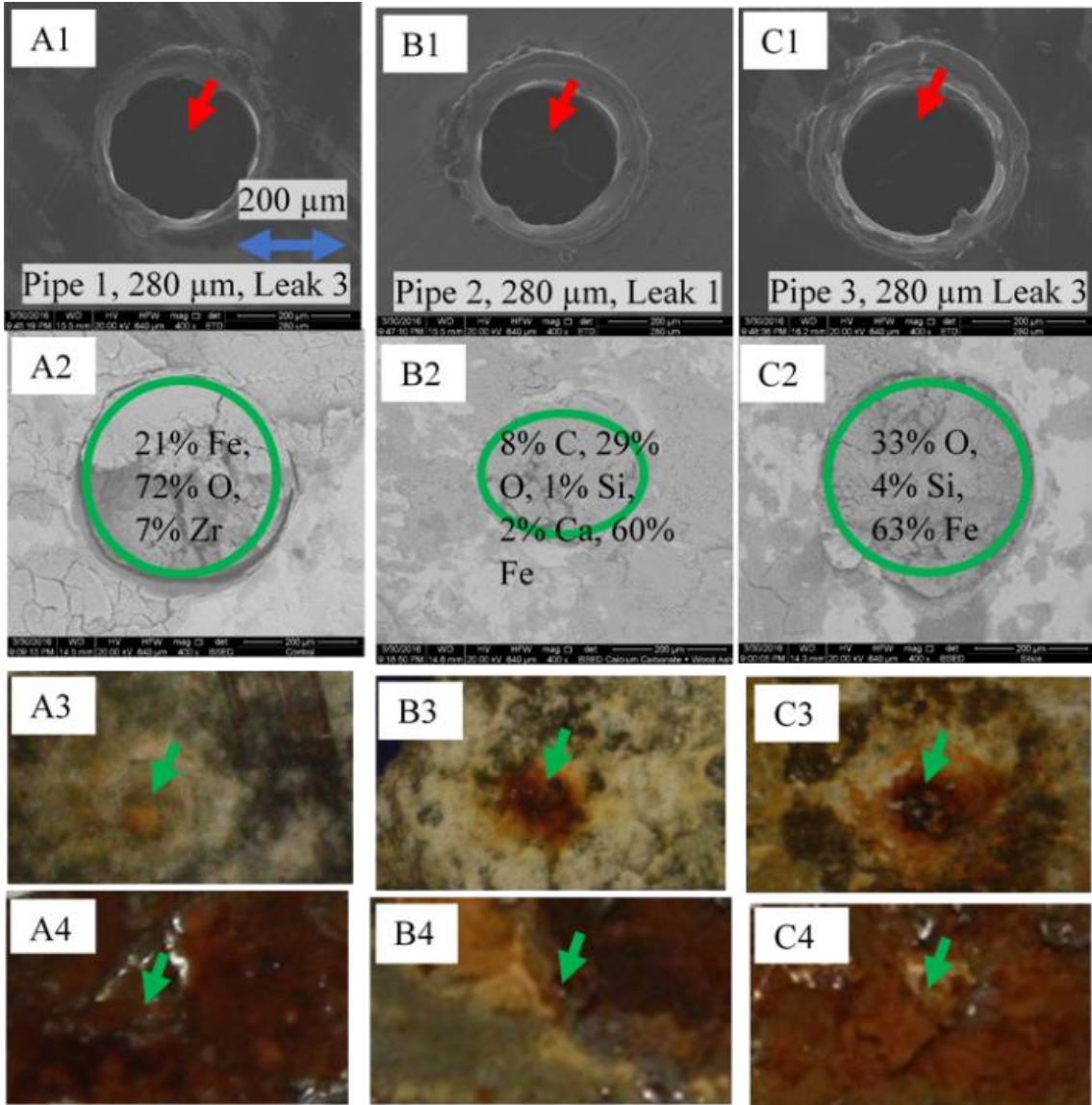


Figure 5-7. ESEM/EDS and camera photos of three representative 280 μm leaks from three different pipes. The initial (A1, B1, C1) and final (A2, B2, C2) microscope images showed that leaks were repaired, and the composition of the sealing materials shown in each photo was on a normalized mass percent basis. The rusts on the outside surface (A3, B3, C3) were more abundant than the inside surface by eyes (A4, B4, C4). The sealed materials in the leaks were rusts or added particles as examined under ESEM/EDS (A4, B4, C4) after removal of inner surface layers of rusts (A = 280 μm leak 3 in the control pipe; B = 280 μm leak 1 in the pipe remediated with calcium carbonate and wood ash particles; C = 280 μm leak 3 in the pipe remediated with silica particles).

Discussions

Zeta potential did not play a significant role in the remediation of leaks. Calcium carbonate, silica and wood ash particles in Blacksburg tap water had a zeta potential of 2.0 mV, -15.2 mV and -11.9 mV respectively, making calcium carbonate particles more likely to grow and coagulate compared to the other two particles (Hanaor *et al.*, 2012). However, the remediation rate (1/12) with calcium carbonate particles was only slightly better than that (0/12) in the control condition, whereas it was much worse compared to that (10/12) with wood ash particles and to that (10/12) with silica particles.

Particle size was crucial in controlling the success of iron leak remediation. Overall, larger particles tend to clog the same size leaks at a greater success rate. Though the wood ash particle concentration (~0.04 g/L) in water was about 15% of that with the added calcium carbonate particles, the 17.2 times larger wood ash particles versus calcium carbonate led to 9 times larger remediation as indicated by the fraction of remediated leaks. Likewise, with only 3.1% of the silica particle concentration (~1.3 g/L), the 4.5 times larger wood ash particles achieved the same remediation rate via physical clogging as the silica particles.

The leak size was another important factor for determining the success rate of leak clogging (Tang and Edwards, 2017). In the case of control condition with no added particles, smaller leak size increased the remediation rate when the reaction time was sufficient. Specifically, none of the twelve leaks (280–1000 μm) was remediated during the first short reaction time of 58 days whereas all smaller leaks (280–400 μm) and 0–33% of the larger leaks (700–1000 μm) self-remediated at the end of 177-day experiment. When silica and wood ash particles were added to Blacksburg tap water for 58 days, all smaller leaks (280–400 μm) and 67–100% of larger leaks (700–1000 μm) were successfully remediated. When calcium carbonate particles were dosed for 58 days, only one of the smallest 280 μm leaks was remediated.

The above observations indicate that addition of particles in a short term could enhance leak remediation rate. Consequently, efforts over the last century, and especially the last few decades, to minimize turbidity of effluent filtered water, might have reduced the likelihood of leak repair that once might have occurred naturally.

CONCLUSIONS

The particle analysis, leakage rate measurement, pressure test and characterization of sealing materials yielded the following conclusions:

- Without particles, leak repair occurred naturally in galvanized iron pipes with the remediation rate increasingly with time.
- The addition of calcium carbonate, silica and wood ash particles for 58 days increased the remediation rate from 0% (0/12) in the control condition to 8.3% (1/12) for calcium carbonate, to 83.3% (10/12) for silica and to 83.3% (10/12) for wood ash.
- Larger particle size remediated the iron pipe leaks with a greater remediation rate.
- The zeta potential did not influence the iron leak remediation rate significantly.

- Smaller leak sizes could be remediated with added particles at a higher remediation rate.
- The strength of leak repair was relatively strong and resilient in all three tested pipes. About 27–73% of the repaired leaks could withstand a high household plumbing pressure of 100 psi and 86–100% of them could withstand a typical household plumbing pressure of 55 psi.
- The iron corrosion rust was a dominant component in clogging materials even when particles were introduced, indicating that addition of calcium carbonate, silica or wood ash enhanced the natural ability of iron pipe to remediate itself.

ACKNOWLEDGEMENTS

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APPENDIX B

Supporting Information

Chapter 5. In-Situ Remediation Of Iron Pipe Leaks With Calcium Carbonate, Silica And Wood Ash Particles In Potable Water Systems

Min Tang, Fei Wang, and Marc A. Edwards

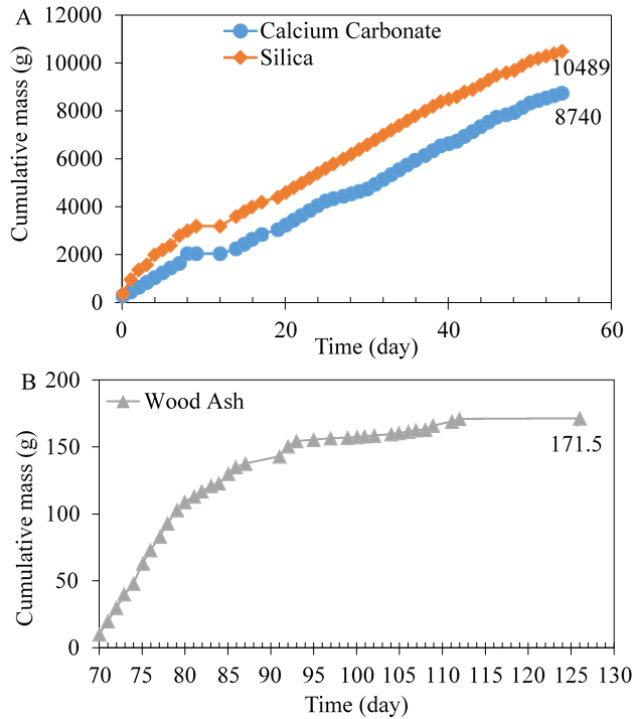


Figure B1. The cumulative mass of calcium carbonate, silica and wood ash particles added to pipe 2 and pipe 3 as a function of time. About 10,489 grams of calcium carbonate were added to pipe 2 and 8,740 grams of silica were added to pipe 3 after 58 days of experiment (A). Afterwards, about 171.5 grams of wood ash were added to pipe 2 from day 70 – 127 (58 days) (B).

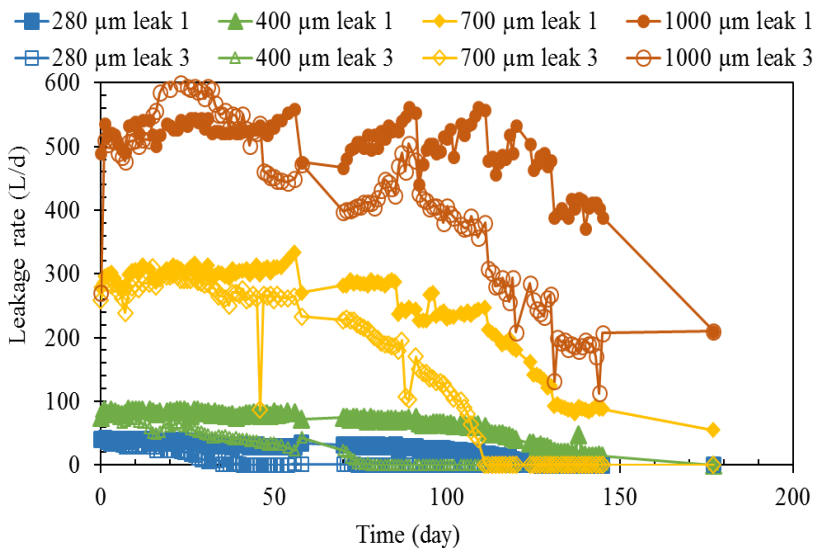


Figure B2. The leakage rate of each leak size in pipe 1 at a water pressure of 10 psi was a function of time when no particles were added as a control condition (leak 1 = the leak closest to the domestic Blacksburg tap water outlet; leak 3 = the leak furthest from the outlet).

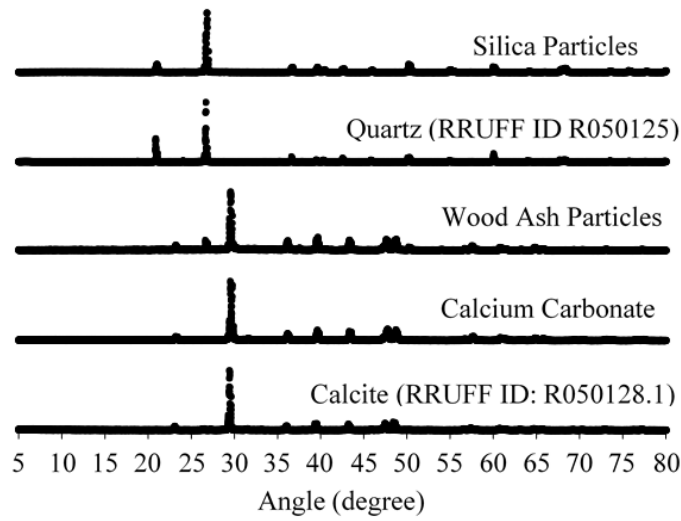


Figure B3. The X-Ray Diffraction (XRD) patterns for calcium carbonate, silica and wood ash particles (Anthony *et al.*, 1990).

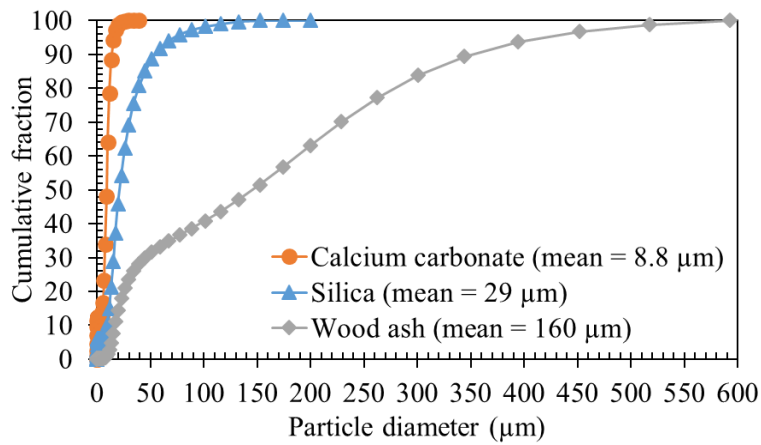


Figure B4. Cumulative fraction of calcium carbonate, silica and wood ash particles in Blacksburg tap water that passed the examined diameter. The mean (\pm std) diameter size of calcium carbonate, silica and wood ash particles in Blacksburg tap water was 8.8 (\pm 4.4), 29 (\pm 27) and 160 (\pm 140) μm respectively.

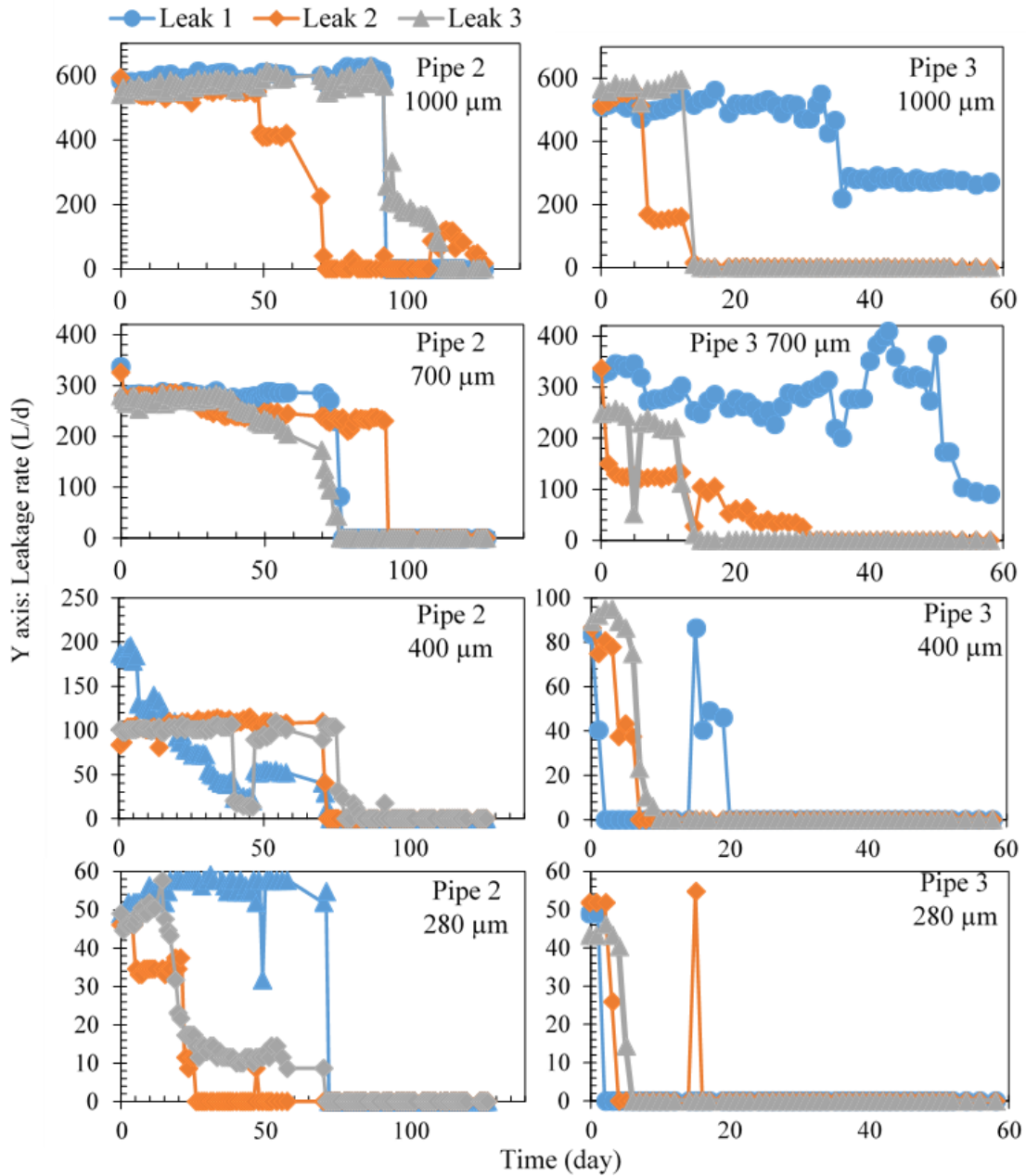


Figure B5. Leakage rate versus time for leaks in waters with calcium carbonate and wood ash particles (Pipe 2, left column) and silica particles (Pipe 3, right column) (leak 1 = the leak closest to the domestic Blacksburg tap water outlet; leak 3 = the leak furthest from the outlet).

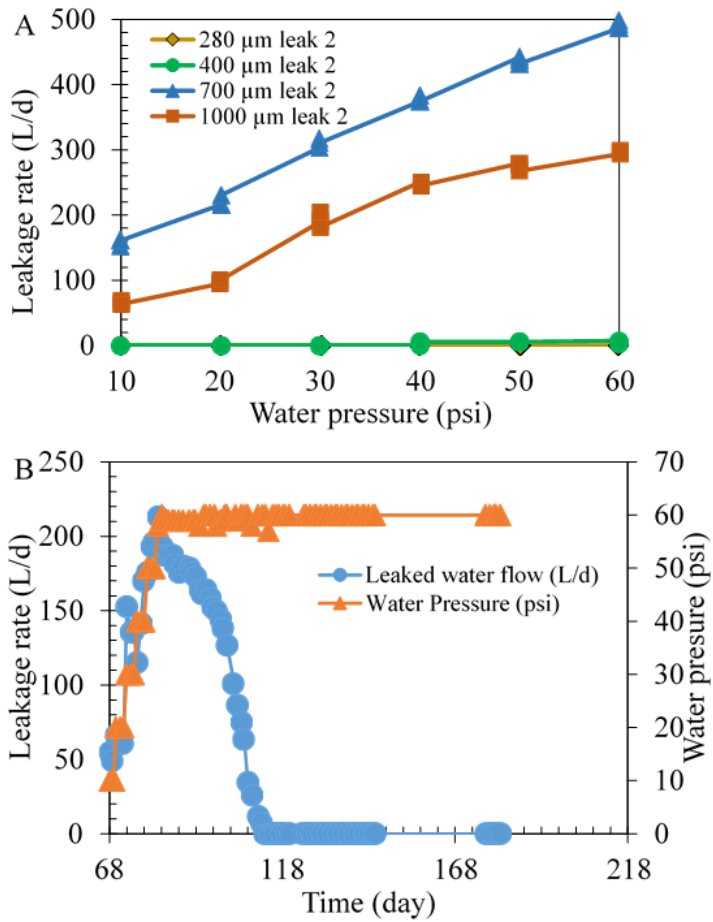


Figure B6. The leakage rate of four representative leaks was a function of water pressure in the first phase of pressure test for the control condition (A). In the first phase of the pressure test, a previously unrepaired 700 μm leak (or leak 1 in pipe 3 with silica particles) was completely remediated at a water pressure of 60 psi, as indicated by the reduction of leakage rate to zero (B).

Table B1. The maximum pressure the leak could withstand before leaking again in two phases of pressure test.

Pipe ID	Maximum Pressure That The Leak Could Withstand Before Leaking (psi)								
	Pressure Testing in	Phase 1				Phase 2			
Pipe 2	<i>Leak Size (μm)</i>	<i>1000</i>	<i>700</i>	<i>400</i>	<i>280</i>	<i>1000</i>	<i>700</i>	<i>400</i>	<i>280</i>
	Leak 1	> 60	> 60	> 60	> 60	85	60	> 100	> 100
	Leak 2	NA	> 60	> 60	> 60	NA	25	95	95
	Leak 3	> 60	> 60	> 60	> 60	55	65	95	> 100
Pipe 1	Leak 1	NA	NA	> 60	> 60	NA	NA	95	> 100
	Leak 2	NA	NA	40	> 60	NA	NA	40	> 100
	Leak 3	NA	> 60	> 60	> 60	NA	30	>100	95
Pipe 3	Leak 1	NA	> 60	> 60	> 60	NA	20	> 100	> 100
	Leak 2	> 60	> 60	> 60	> 60	45	> 100	> 100	> 100
	Leak 3	> 60	> 60	> 60	> 60	45	> 100	> 100	> 100

NA = Unrepaired leaks at water pressure of 10 psi; leak 1 = the leak closest to the domestic Blacksburg tap water outlet; leak 3 = the leak furthest from the outlet; Number in bold and red color = Repaired leaks in Phase 1 were partially damaged in phase 2 due to disturbance of taking-apart or installation of new apparatus process, and therefore the pressures these repaired leaks could withstand in phase 1 of pressure test were listed; > 60 = Repaired leaks stayed repaired at water pressure of 60 psi in the first phase of pressure test; > 100 = Repaired leaks stayed repaired at water pressure of 100 psi in the second phase of pressure test

CHAPTER 6. OVERVIEW AND FUTURE WORK

OVERVIEW

This dissertation represents the first study of *in situ* leak remediation for potable water pipes, which is facilitated by waterborne or water-formed particulates. Key conclusions included the following: 1) metallic corrosion, precipitation and physical clogging are the main mechanisms of *in situ* remediation (Chapter 1); 2) the metallic pipe leaks could autogenously repair via formation of corrosion rust at a wide range of water chemistries (Chapter 2); 3) the success and speed of autogenous metallic pipe leak repair was influenced by physical parameters, including leak size, water pressure, leak orientation and pipe wall thickness (Chapter 3); 4) the success and speed of autogenous metallic pipe leak repair could be manipulated by changing water chemistry (ionic strength, alkalinity, silica, magnesium, dissolved oxygen, corrosion inhibitors, disinfectants and Ryznar Stability Index) (Chapter 4); and 5) the presence of large particles in the water could physically block leaks and enhance the likelihood of leak repair (Chapter 5). The sealing materials were photographed, characterized and tested under pressure, and in many cases were strong and resilient, indicating the potential of *in situ* remediation to sustain failing infrastructure in some cases.

FUTURE WORK

Parameters including water temperature, water flow/velocity, pipe wall roughness, the shape of leak-holes and leak size ($> 1000 \mu\text{m}$) representing a worse case in field studies are worthy of additional study, as they could affect the formation and deposition of water-formed or waterborne particles in leak-holes.

The electrochemical potential of iron or copper pipes can be altered by cathodic protection and galvanic couplings (Clark *et al.* 2013), which in turn, could influence the formation of leaks in the first place. The potential impact of these factors on the likelihood of *in situ* repair should be further examined.

One pilot study of *in situ* leak repair was already conducted under conditions deemed likely to heal the Delaware Aqueduct (Letterman *et al.* 2008) and it seemed promising. Some full-scale studies would be necessary to prove the viability of the approach in practice, since water distribution systems are inherently more complex than the systems tested either at pilot or bench-scale herein.

Even if purposeful *in situ* remediation is not implemented or tested, retrospective studies should examine the hypothesis that thinner walled pipes, reduced loading of particulates and alternative disinfectants may have reduced the likelihood of *in situ* repair that once occurred naturally. Water pipes installed after World War II had a relatively short lifetime compared to many of the thicker walled materials installed earlier, and some consideration should be given to the idea that this might be partly due to a reduced natural frequency of *in situ* leak remediation. Likewise, some studies have found that higher pressures can increase the incidence of pipe leaks. The work cited herein, provides a possible explanation

for this observation, in that lower pipeline pressures can enhance the likelihood of leaks healing themselves.

REFERENCES

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