NRC Publications Archive Archives des publications du CNRC

Biodeterioration of asbestos cement (AC) pipe in drinking water distribution systems

Wang, D. L.; Cullimore, R.; Hu, Y.; Chowdhury, R.

This publication could be one of several versions: author's original, accepted manuscript or the publisher's version. / La version de cette publication peut être l'une des suivantes : la version prépublication de l'auteur, la version acceptée du manuscrit ou la version de l'éditeur.

For the publisher's version, please access the DOI link below. / Pour consulter la version de l'éditeur, utilisez le lien DOI ci-dessous.

Publisher's version / Version de l'éditeur:

<https://doi.org/10.1016/j.ibiod.2011.05.004>

International Biodeterioration & Biodegradation, 65, 6, pp. 810-817, 2011-11-01

NRC Publications Record / Notice d'Archives des publications de CNRC:

<https://nrc-publications.canada.ca/eng/view/object/?id=084f0596-f366-44d0-b75f-028fc56a71a0>

<https://publications-cnrc.canada.ca/fra/voir/objet/?id=084f0596-f366-44d0-b75f-028fc56a71a0>

Access and use of this website and the material on it are subject to the Terms and Conditions set forth at

<https://nrc-publications.canada.ca/eng/copyright>

READ THESE TERMS AND CONDITIONS CAREFULLY BEFORE USING THIS WEBSITE.

L'accès à ce site Web et l'utilisation de son contenu sont assujettis aux conditions présentées dans le site

<https://publications-cnrc.canada.ca/fra/droits>

LISEZ CES CONDITIONS ATTENTIVEMENT AVANT D'UTILISER CE SITE WEB.

Questions? Contact the NRC Publications Archive team at

PublicationsArchive-ArchivesPublications@nrc-cnrc.gc.ca. If you wish to email the authors directly, please see the first page of the publication for their contact information.

Vous avez des questions? Nous pouvons vous aider. Pour communiquer directement avec un auteur, consultez la première page de la revue dans laquelle son article a été publié afin de trouver ses coordonnées. Si vous n'arrivez pas à les repérer, communiquez avec nous à PublicationsArchive-ArchivesPublications@nrc-cnrc.gc.ca.

Biodeterioration of asbestos cement (AC) pipe in drinking water distribution systems

Wang, D.L.; Cullimore, R.; Hu, Y.; Chowdhury, R.

NRCC-54544

A version of this document is published in / Une version de ce document se trouve dans:

International Biodeterioration & Biodegradation, 65, (6), pp. 810-817, September 1, 2011,

DOI: [10.1016/j.ibiod.2011.05.004](http://dx.doi.org/10.1016/j.ibiod.2011.05.004)

The material in this document is covered by the provisions of the Copyright Act, by Canadian laws, policies, regulations and international agreements. Such provisions serve to identify the information source and, in specific instances, to prohibit reproduction of materials without written permission. For more information visit <http://laws.justice.gc.ca/en/showtdm/cs/C-42>

Les renseignements dans ce document sont protégés par la Loi sur le droit d'auteur, par les lois, les politiques et les règlements du Canada et des accords internationaux. Ces dispositions permettent d'identifier la source de l'information et, dans certains cas, d'interdire la copie de documents sans permission écrite. Pour obtenir de plus amples renseignements : <http://lois.justice.gc.ca/fr/showtdm/cs/C-42>

Biodeterioration of asbestos cement (AC) pipe in drinking water distribution systems

Dunling Wang^{1*}, Roy Cullimore², Yafei Hu¹, Rudaba Chowdhury¹

¹NRC Centre for Sustainable Infrastructure Research, 3737 Wascana Parkway, Regina, SK S4S 0A2

²Droycon Bioconcepts Inc., 315 Dewdney Avenue, Regina, SK S4N 0E7

Abstract: Various types of microorganisms have been found to inhabit the inner surfaces of asbestos cement (AC) pipe and their activities can cause significant structural damage. They cause a patina to form on the inside surface of AC pipes as a distinctively continuous coating, commonly 2 to 5 mm in thickness and generally pigmented as yellow, orange, brown or black depending on the metallic cations that have been incorporated into the surface of biofilm (bioaccumulation). Four sublayers can be identified in the patina, from the outer sublayer that directly interacts with the conveyed drinking water to the inner sublayer that is in proximity of the intact cement matrix. The microbes in the outer sublayer are composed mainly of inactive biomass that separates the aerobic environment of the flowing water from the anaerobic conditions inside the patina. The bacteriological community structure shifts from mixed heterotrophic (HAB), iron related (IRB) and slime forming bacteria (SLYM) in the outer layer, to a more diverse community with IRB, acid producing (APB) and SLYM and HAB in the middle sublayer, and further to the SLYM dominated in the inner sublayer. By directly interacting with cementitious materials, including generating organic acids, IRB and APB play important roles in the leaching of free lime and the dissolution of calcium (Ca)-bearing hydrated components of AC pipes, creating porous structure and reducing the pipe strength. Scanning electron microscopy with an energy dispersive X-ray has revealed that bacterial activity on the internal AC pipe wall had resulted in a significant loss of hydrated cement matrix, which can cause pipe failure when stresses imposed on the pipe exceed the remaining pipe strength.

Key Words: biodeterioration, biofilm, asbestos cement (AC) pipe, patina, slime-forming bacteria, acid-producing bacteria, heterotrophic bacteria.

*Corresponding author, phone (306) 780-8698, email Dunling.Wang@nrc-cnrc.gc.ca

INTRODUCTION

Asbestos cement (AC) pipes are made of a mixture of asbestos fibers and Portland cement, with or without silica. During manufacture, a paste of these components is compressed by steel rollers to form a composite material of great strength and density. The pipe was extensively used in the mid-1900s in potable water distribution systems in the United States and western Canada. The Chrysotile Institute of Canada estimates AC pipe lifespan at 70 years (Chrysotile Institute, 2011), but actual service life depends largely on pipe manufacturing quality and working environments.

Traditionally, AC pipe degradation was believed to be a chemical process of free lime leaching due to conveyed water having either an acidic pH or low alkalinity (Millette et al., 1984; AWWA, 2003; AwwaRF and DVGW-Technologiezentrum Wasser, 1996). The loss of free lime in AC pipe leads to pipe softening and a reduction in the effective thickness of the pipe wall, and therefore, a loss of mechanical strength. When stresses imposed on a pipe by external forces exceed the reduced strength, the pipe fails.

Recently, Wang and Cullimore (2010) noted that AC pipes can be degenerated as a result of the actions of various bacteria in drinking water distribution systems. The growth of different species of microbes often occurs within common consortial structures. In drinking water distribution systems, these microorganisms bind with water retaining polymeric matrices to form biofilms and often attach to the surfaces of distribution pipes. The bacteria *Acidithiobacillus* associated with the corrosion of concrete was first noticed more than 50 years ago when it was isolated from corroded cement (Parker, 1954). The microorganisms cause “corrosion” through the excretion of metabolic organic acids and enzymes, and result in bioaccumulation of selected inorganic and organic chemicals travelling in an aqueous phase over the biofilms (Costerton and Lappin-smith, 1989). A 3-5 mm thick patina layer was observed on the inner wall of a broken AC pipe that had been in service for 35 years (Wang and Cullimore, 2010). The patina is a porous layer, mainly composed of microbial biomass along with interwoven asbestos fibers. Several groups of bacteria were identified in the patina, including slime-forming bacteria (SLYM), heterotrophic

bacteria (HAB), iron-related bacteria (IRB) and acid-producing bacteria (APB). Under anaerobic conditions, the AC pipe samples soaked in the bacterial culture of HAB, SLYM and APB for 28 days exhibited various degrees of weight loss, which indicated that these groups of bacteria were very aggressive to the cement matrix of the AC pipes (Wang and Cullimore, 2010).

The deterioration of cementitious pipe matrix occurs in two stages. First, an abiotic process causes pH reduction from >12.4 to around 9 (Islander et al., 1991; Mori et al., 1992). The initial alkalinity of the cement prevents microorganism growth on the surface of AC pipes. However, reactions with conveyed water (normally in a pH range from 6.5 to 8.5) and its dissolved components, such as chloride (Cl^-), sulfate (SO_4^{2-}) and carbonate (CO_3^{2+}), consume the alkalinity and quickly decrease pH value of the cement surface. Once the cement surface pH is reduced to about 9, precursory microorganisms start to inhabit the surface (Zherebyateva et al., 1991). Second, the bioaccumulation of dissolved organic carbon and nutrients from treated drinking water enhance microbial activity and biofilm formation on the pipe surface. Bacterial activities affect the cement-patina interface and make it more reductive. In an anaerobic environment, bacterial fermentation produces short-chain fatty acids, which accelerate the rate of free lime leaching and dissolution of Ca-bearing minerals in the hydrated cement matrix.

Gu et al. (1998) observed that certain bacteria (*i.e.*, *Acidithiobacillus*) grown in the biofilms excreted sulfuric acid. Organic compounds and sulfuric acid could act simultaneously to release calcium by chelation and dissolution with their exopolymeric substances. A recent research indicated that *E.coli* and synthetic organic acid can cause seriously damage to structure of hardened cement paste (Magniont et al., 2011). Several groups of environmental bacteria, such as sulfate reducers, ammonia and nitrite oxidizers, denitrifies and cyanobacteria, could cause the deterioration of cementitious materials (Wang and Cullimore, 2010; Zherebyateva et al., 1991).

The primary objective of this study was to assess the impacts of microbial activity inside biofilms on the deterioration of AC pipes in drinking water distribution systems. Biodeterioration of AC pipes in a

drinking water distribution network was evaluated through an investigation of bacterial activity, changes of bacterial types across sublayers of pipe walls, and the examination of the cement matrix microstructure using scanning electron microscopy.

MATERIALS AND METHODS

Pipe Samples

Two AC Pipe samples were collected from the drinking water distribution system in the City of Regina, SK, Canada (Table 1). One pipe sample, referred to as C202, was installed in 1956 and the other, C311, was installed in 1957. Pipe C202 failed in 2008 and C311 failed in 2009 as a result of ground shifts caused by expansive clay. Both pipes had similar accumulations of biofilms on the internal pipe surfaces, and phenolphthalein staining tests indicated that the leaching of free lime extended to a depth of 3.1 to 4.6 mm, which was considered to be a significant patina layer.

The treated drinking water in Regina has a pH value range from 7.2 to 7.5, and an alkalinity between 105 and 160 mg/L. Total hardness varies from 165 to 260 mg/L, with calcium concentrations ranging from 35 to 60 mg/L. Conductivity varies from 470 to 650 mg/L and total dissolved solids ranges from 320 to 500 mg/L. Normally, the low concentrations are observed in summer and the high concentrations occur during winter as a result of precipitation patterns in the region. The treated drinking water has a total organic carbon in a range from 0.3 to 4.5 mg/L, a nitrate concentration <0.1 mg/L and total phosphorus <0.05 mg/L. The sulfate content ranges from 100 to 200 mg/L, which is not considered to be aggressive to AC pipes. The analysis of the soils collected from pipe sample locations indicated that pipe C202 was surrounded by an expansive clayey soil, with a pH value of 7.8, and sulfate content of 2,030 mg/Kg. Pipe C311 was also buried in a clayey soil, which had a pH of 8.2 and sulfate content of 1,616 mg/Kg.

The Identification of Bacterial Group and Community Structure

The identification of the bacterial communities was based on the biological activity reaction tests (BARTTM), manufactured by Droycon Bioconcepts Inc., Regina, Canada (Cullimore, 1993). The tester is designed to provide a range of environments from very reductive in the base to very oxidative around the floating ball, therefore creating a vertical gradient in the tester that encourages many different types of bacteria to grow. For this purpose, a crystallized selective medium pellet is placed in the base of the tester. The nutrients in the selective medium diffuse up and allow only the bacteria groups being investigated to grow and trigger reactions. The BART testers are categorized into several product groups that look for bacterial activity of specific groups. In general, anaerobic bacteria often start multiplying from oxygen-depleted zone at the bottom of reactor and gradually move upward; while aerobic bacteria always grow from well-aerated zone around the floating ball and move down. The lag time from start date to completion of the reaction is then used to calculate the approximated activity. A shorter lag time reflect a higher population of active bacteria. As a result, the tester measures activities of a group of bacteria that thrive in similar conditions and have similar functions towards their impact on the environment, rather than a differentiation of specific species or strains. The BART tests only measure the population of active bacteria in a sample not including those in sleep mode or inactive because it will take a longer time for them to become active. The traditional colonies formation unit (cfu) is not used because bacterial activity is recorded as time lags, which is then used to calculate the predicted active cells.

The investigation of bacterial community structure focused on the patina coating on internal walls of the pipes. Four patina samples were taken from the porous layer inside wall of each AC pipe at different positions, and each further divided into 4 sublayers based on the physical appearance of the porous layer. sublayer A represents the deterioration frontier deep inside the patina in direct contact with the intact portion of AC pipe wall, and D corresponds to the thin outer surface stratum interacting with flowing water; sublayers B and C are between A and D. The composite sublayer samples were used for active bacterial population and community structure tests with BART testers.

About 1.5 g of solid sample was used to estimate active cells of the interested bacterial groups. The addition of 15 ml of sterilized distilled water to the BART tester allowed the selective medium to develop oxidation reduction potential (ORP) gradients. The standard operating practice was modified to allow for the placement of a 1.5 g solid porous patina sample. For this investigation, time lapse was recorded with an automatic digital reader. The testers selected for this study included SRB, IRB, SLYM, HAB, and APB.

Staining Test, Scanning Electron Microscopy (SEM) with Energy Dispersive X-ray

The staining test employs phenolphthalein solution that is mild acidic and colorless, but turns into dark pink when reacts with strongly basic materials. The test starts with spraying the solution onto a freshly cut AC pipe sample. The phenolphthalein turns dark pink where the pipe has not been subjected to deterioration and free lime in the hydrated cement matrix preserves a strong alkaline environment. When the pipe is deteriorated, free lime is removed from the hydrated matrix and the deteriorated layer become pH neutral or slightly acidic, phenolphthalein solution remains colorless. Therefore, the staining test differentiates the areas where lime leaching occurs. Deterioration depths are directly measured on the tested pipes at four equally spaced points along the circumferential positions, and deterioration rate is calculated based on the number of years that the pipe has been in service. Figure 1 shows a grinded pipe cross section and the coloring of cross section after staining.

Thin section samples were cut from the AC pipe, polished and mounted on glass slides for analysis of surface morphology by a Jeol JSM 6360 Scanning Electron Microscope (SEM) equipped with a Noran-System 7 EDS spectrometer. A series of images (at 100X, 500X and 1000X magnifications) were taken from the outside to the inside of the pipe samples. Each image covers approximately a 1-2 mm width. Microanalysis of the elemental distribution was performed on selected SEM image areas, and the following elements were measured as a percentage of total weight: oxygen (O), calcium (Ca), silicon (Si), magnesium (Mg), iron (Fe), aluminum (Al), sulfur (S), and chloride (Cl).

RESULTS

Free Lime Leaching and AC Pipe Deterioration Rate

The inner and outer pipe walls have been interacting with completely different environmental conditions, and therefore different deterioration rates have been observed. For the outer surface of AC pipes, contact with varying groundwater and soils has caused different degrees of leaching of free lime, sulfate reaction and carbonation on pipe wall. Due to uneven moisture conditions around the pipes, the free lime leaching depth from the outer surface can be significantly different. The free lime leaching depths of the pipe C202 varied from 2.0 to 5.0 mm. The deterioration rates are calculated in a range of 0.038 to 0.097 mm/yr. In comparison, the pipe C311 leaching depths range from 1.5 to 4.8 mm, with the calculated deterioration rates from 0.029 to 0.092 mm/yr (Table 1). This variation in the same pipe can be related to the soil and ground water conditions at the particular locations where the pipes were exhumed. However, due to these two pipes had been conveying the same treated water for 52 years, they have similar internal lime leaching depths and deterioration rates. The pipe C202 free lime leaching depths range from 3.6 to 4.6 mm, and the calculated average deterioration rate is 0.083 mm/year from inner surface. Similarly, the pipe C311 has a leaching depth range from 3.1 to 4.4 mm, with an average deterioration rate of 0.072 mm/year (Table 1). A pH value higher than 7 and elevated levels of total alkalinity and hardness of the treated drinking water has kept the pipe deterioration rates at a reasonably low.

Cultural Identification of Bacterial Communities within Patina

Of the total active microbial population recovered from the four sublayers of the patina, SLYM bacteria accounted for 95.5%, HAB 2.4%, IRB 1.6%, APB 0.4% and no SRB detected (Table 2). Bacterial activity and composition in each of the four sublayers showed that 68.7% of the total active bacteria in the patina occurred in sublayer A, where these bacteria directly interacted with the hydrated cement matrix near the intact portion of AC pipe wall. Among those active bacteria, the SLYM group comprised 99.5%,

with the rest 0.5% belong to IRB (Table 3). In comparison, the D sublayer that openly interacted with the conveyed drinking water accounted for only 1.7% of the total active population in the patina, even though it constituted a significant biomass, apparently a big portion of the biomass in this sublayer is inactive bacteria and organic materials from microbial metabolisms. Sublayer B, within the void-developing zone where loss of free lime and other hydrated calcium silicates were observed, comprised 28.4% of the total active population; the majority of bacteria was SLYM (93.5%) with the increased numbers of IRB (1.5%) and HAB (5%). In sublayer C, approximately 1.2% of the recovered bacterial population occurred in this sublayer. It should be noted that the majority of bacteria in this sublayer were aggressive to AC pipes, such as APB (30.8%), IRB (42.0%) and HAB (8.9%), and the large voids developed in this sublayer become clearly visible (Figure 2).

Elemental Analysis with Scanning Electron Microscopy (SEM)

The SEM image and elemental distribution images indicated that the chemical weathering in the outer layer is directly related to soil conditions and groundwater characteristics. For the given pH values and sulfate concentrations of the two locations, there was a clear loss of calcium content, but the magnitude was relatively small in comparison with the inner layer (Table 4), and the original minerals and aggregates were still held in place without significant deformation (Figure 2). In contrast, the leaching from the inner surface layer became more severe in comparison with the outer surface layer. SEM analysis has shown that a significant amount of the hydrated cement materials had been dissolved, left considerable amount of porosity and some large voids (Figure 2). These voids seemed to extend and develop along the structural weak zone between those original layers created during the manufacture process. Elemental compositions in this deteriorated layer showed a complete removal of Fe. A loss of 86.3% (from 21.2% to 2.9%) and 71.8% (from 18.0% to 5.08%) of Ca was observed in the deteriorated layer of sample C202 and C311, respectively (Table 4). In the meantime, 10% enrichment of Mg took place in the deteriorated layer of both pipes. This relative enrichment was directly resulted from the loss

of Ca-bearing minerals in the hydrated cement matrix, as white asbestos ($\text{Mg}_3\text{Si}_2\text{O}_5(\text{OH})_4$), which is extremely resistant to weathering, was used for manufacturing these AC pipes. Similarly, small increases in elements of Si and Al in the deteriorated layer reflect relative enrichment of quartz and aluminum oxides after the Ca bearing-minerals is weathered and left the matrix.

DISCUSSION

The AC pipes installed in Regina drinking water distribution systems are classified as Type II, which contains about 15 to 20% of asbestos, 45 to 51% Portland cement and 32 to 34% of silica. When these components mixed and hydrated, about 1% of free lime was produced. Initial pH of the AC pipe is greater than 12.4, due to presence of free lime. Degradation of AC pipe is a combination of biological and chemical processes and these processes often facilitate each other. For example, the chemical process in the beginning caused reduction in pH of inner pipe wall from greater than 12 to about 9, which made it possible for bacterial communities to establish on the pipe surface (Bastidas-Arteaga et al., 2008). Once these microbes established, they started to acquiring nutrients from the flowing drinking water and from pipe surface to multiplying in population. SLYM bacteria may have first harbored and thrive as biofilm on the pipe surface around the voids and cracks. As biofilm further developed, other bacteria in the flowing water can be intercepted and joined in the biofilm. Together, they form a community as patina developed. Within this community, each group would play specific functions. After more than many years, this bacterial community became mature and stable, and played important roles in the destruction processes of AC pipes.

Functionally, bacteria in the sublayer D directly interact with the treated drinking water, battle with disinfectants, intercept nutrients and restrict free oxygen flow into the deep layer. While the bacteria in the sublayer A will have to face strong alkaline conditions, and continue to move the deterioration front further into the intact cement matrix. In the sublayer B and C, HAB, IRB and APB thrive in a relatively

favorable environment that created by the bacteria in the sublayers A and D, which has an anaerobic and appropriate pH conditions. These bacterial activities produce organic compounds and short-chain fatty acids that not only caused the dissolution of Ca-bearing minerals in the hydrated cement matrix, but also provided necessary organic substrates for the bacteria in adjacent sublayers and help overcome the strong alkaline conditions in the intact cement matrix.

Fermentation under anaerobic conditions often generates organic compounds that include short-chain fatty acids. The production of these compounds causes local pH to decrease and further facilitates the weathering and dissolution of the acid-receptive minerals in hydrated cement matrix, thus, creating pitting and voids within a pipe wall. In the meantime, the increased porosity allows dissolved Ca ions to be diffused into the bulk water to remain the pore water in these voids in a state of unsaturation. This unequilibrium will maintain dissolution process to continue. A significant dissolution and loss can be observed close to the inner surface of pipe walls (Figure 3). The lost components are mainly Ca-bearing free lime, C2S, C3S because these minerals become unstable in acidic conditions. Microbial activities inside patina continue to produce acids and maintain the environment in acidic range.

Deterioration rates were relatively uniform along the internal circumference of the pipe and became more severe in comparison with the outer surface layer (Table 1). This can be resulted from following reasons: (1) flowing water inside the pipe carries away the dissolved components, such as Ca^{2+} , and maintains deterioration processes continuing, while the outer pipe surface experienced periodical dry-wet and unsaturation and oversaturation cycles; (2) the development of a patina on inner surface creating an anaerobic environment, in which microbial fermentation produced organic acids and caused the local pH to decrease. However, the out pipe surface faced changing environments that also created uneven conditions around the pipes. Similar internal leaching deterioration rates from both AC pipes seemed directly related to the same drinking water they conveyed (Table 1).

There was a reduction of ORP across the patina at the sublayer D as indicated by the bacterial growth reactions in the BART testers. IRB test with sample from sublayer D showed a black growth started

around the floating ball and moving downwards, pointing to a mixed community dominated with aerobic including pseudomonad bacteria. In comparison, the SLYM test with sample from sublayer A produced a foamy reaction around the floating ball, indicating that the sample is reductive and anaerobic fermentative bacteria are dominating. This observation is expected as the free oxygen movement through sublayer D is restricted due to bacterial activities. It is also evidenced that the sublayers B and C were reductive and associated with fermentative bacterial community, such as HAB and APB. The activity of HAB and APB is most likely to be acid-generating, which could then trigger dissolution of Ca-bearing minerals and push the deterioration processes further into the deep layer towards the centre. The IRB group included both iron-oxidizing and iron-reducing bacteria that use iron in their metabolism. Type II AC pipes originally contains about 0.9 to 1% Fe in the forms $\text{Ca}_4\text{Al}_n\text{Fe}_{2-n}\text{O}_7$. This Fe was completely removed from the deteriorated layers as a result of IRB activities.

Asbestos fibers and quartz particles are extremely resistant to weathering and normally remain intact, therefore, the relative content of these two become elevated in the deteriorated pipe.

Deterioration rates depend on the ability of the growing patina to utilize nutrients entrapped in the patina from the conveyed water. Nutrients may also be assimilated from cement matrix itself. The quality of treated drinking water including pH level, content of organic carbon substrates, alkalinity and sulfate concentrations, also affects biological processes. Acidic water facilitates the weathering and transformation of minerals in the hydrated cement and can significantly accelerate the deterioration rates. The content of disinfectant in the drinking water influences the activity level of bacteria, particularly in the sublayer D. However, biodeterioration always occurs in drinking water distribution systems, not just limited to those that convey acidic treated water. The Regina's drinking water has a pH value in a range from neutral to slightly alkaline with moderate alkalinity, hardness, and a relatively low sulfate concentration, therefore, the deterioration rate is relatively low. But an active patina development was clearly exhibited in the inner layer of both pipes samples. Similarly, the soils around the two samples

were considered to be low to moderate in terms of aggressiveness to AC pipes. Therefore, AC pipes maintain their relatively good conditions.

The primary bioaccumulative interface with the flowing water is traditionally regarded as biofilms. Beneath the interface, the patina supports much of the microbiological activity, retained with the tangled asbestos fibers exposed by the reductive acidic deterioration of the hydrated cement matrix. In a fast growing corrosive process, the fibers may form a support stratum, while a considerable amount of microbiological activity continues to occur in the deep layer at the interface of the intact cement matrix. Biodeterioration primarily occur as radial pitting and quickly become circumferential along the structural weakness of the original cement layers. Organic carbon substrates would be of most concern in the drinking water. These substrates, upon reductive degradation, would generate the shorter-chained, fatty acids that cause localized drops in pH into the acid range, and thereby trigger further dissolution of hydrated Ca-bearing cement at greater depths.

From these investigations, it can be summarized that the biodeterioration of AC pipe occurs in four major stages (Figure 4). First, conveyed water reduces the surface pH, allowing bacteria to grow on the inner surface of the pipe wall. The chemical processes can be achieved very quickly, but the establishment of biofilm may take a longer time. Second, active development of biofilm causes formation of a patina and the dissolution of Ca-bearing hydrated cement in the surface layer; mineral loss becomes clearly parallel to the surface along the structural weakness inherited from pipe manufacturing. Third, the patina further extends to deep layers along cracks and continues the corrosion processes. Fourth and last, extensive dissolution of Ca-bearing minerals within the patina caused by more aggressive bacterial groups including HAB, IRB and APB significantly increases porosity. The recalcitrant asbestos fibers within the deteriorated cement matrix become an interwork of asbestos fibers and quartz particles. At this stage, the AC pipe loses its structural strength and becomes fragile. A minor disturbance, such as pressure changes from water hammer, can result in a release of biofilms and asbestos fibers into the drinking water, causing

a health concern. With this being a common concern in AC water distribution pipes, feasible processes that either stabilize the patina or prevent further development of the patina would be desirable. Traditional biofilm removal by physical scratches is not recommended for AC pipe systems.

CONCLUSION

Microbial activities caused patina formation on the inner pipe surface and played an important role in the deterioration of AC pipes in drinking water distribution systems. The SLYM bacteria are directly in contact with the hydrated cement materials, creating an environment for other types of bacteria to establish inside the patina. The activities of HAB, IRB and APB make a local environment anaerobic and acidic, which greatly accelerated the leaching of free lime and dissolving Ca-bearing minerals in hydrated cement matrix. Losses of free limes and Ca-bearing silicates left significant voids and porosity, reduced the effective wall thickness of the cement pipe. Microbial growth on the pipe surface attributed to the development of patina layer in the inner surface of the AC pipe, which resulted in a reduction of pipe strength.

Acknowledgments: The authors would like to thank Communities of Tomorrow and the Water Research Foundation for financial support for this investigation. Contributions from the City of Regina by collecting sample pipes and Dr. Ian Coulson for help in SEM analysis are also appreciated.

REFERENCES

American Water Works Association (AWWA), 2003. Standard for asbestos cement distribution pipe 4” through 16” for water and other liquids. AWWA C400-03, Denver, CO.

- American Water Works Association Research Foundation (AwwaRF) and DVGW-Technologiezentrum Wasser. 1996. Internal corrosion of water distribution system. 2nd Edition, AwwaRF, Denver, CO.
- Bastidas-Arteaga, E., M. Sanchez-Silva, A. Chateauneuf and M.R. Silva, 2008. Coupled reliability model of biodeterioration, chloride ingress and cracking for reinforced concrete structure. *Structural Safety* 30, 110-129.
- Chrysotile Institute, 2011. <http://www.asbestos-institute.ca/specialreports/acpipes/acpipes.html>.
- Costerton, J.W. and H.M. Lappin-Smith, 1989. Behavior of bacteria in biofilms. *American Society of Microbiological News* 55, 650-654.
- Cullimore, D.R., 1993. "Practical Manual of Groundwater Microbiology", Lewis Publishing, Chelsea, Michigan, USA.
- Gu, J. D., T. E. Ford, N. S. Berke, and R. Mitchell, 1998. Biodeterioration of concrete by the fungus *Fusarium*. *International Biodeterioration and Biodegradation* 41, 101-109.
- Hu, Y. D.W. Hubble, 2007. Factors contributing to the failure of asbestos cement water mains. *Canadian Journal of Civil Engineering* 34, 1-14.
- Islander, R.L., J.S. Devinney, F. Mansfeld, A. Postyn, H. Shih, 1991. Microbial ecology of crown corrosion in sewers. *Journal of Environmental Engineering* 117, 751-770.
- Millette, J.R., G.S. Logsdon, P.J. Clark, and R.N. Kinman, 1984. Evaluating the condition of asbestos cement pipe. *Corrosion 84: the International Corrosion Forum Devoted Exclusively to the Protection and Performance of Materials*, New Orleans, Louisiana, USA April 2-6.
- Mori, T., T. Nonaka, K. Tazaki, M. Koga, Y. Hikosaka, and S. Nota, 1992. Interactions of nutrients, moisture, and pH on microbial corrosion of concrete sewer pipes. *Water Resources* 26, 29-37.
- Parker, C.D., 1945. The corrosion of concrete. 1. The isolation of a species of bacterium associated with corrosion of concrete exposure to atmospheres containing hydrogen sulfide. *Australian Journal of Experimental Biology and Medical Sciences* 23, 81-90.

- Wang, D. and D.R. Cullimore, 2010. Bacterial challenges to asbestos cement water distribution pipelines. *Journal of Environmental Science* 22, 1203-1208.
- Zherebyateva, T. V., E. V. Lebedeva, and G. L. Karavaiko, 1991. Microbiological corrosion of concrete structures of hydraulic facilities. *Geomicrobiology Journal* 9, 119-127.

Table 1 AC pipe samples from the drinking water distribution system in the City of Regina, Canada

	C202-2	C311-2
Pipe diameter (mm)	150	150 mm
Service period	1957 – 2009 (52 yrs)	1956 – 2008 (52 yrs)
Patina development (mm)	0.3 – 0.5	0.3 – 0.5
Pipe thickness (mm)	18.24	17.25
Internal free lime leaching depths (mm)	3.6 – 4.6	3.1 – 4.4
Average internal deterioration rate(mm/yr)*	0.083	0.072
External free lime leaching depths (mm)	2.0 – 5.0	1.5 – 4.8
Average external deterioration rates (mm/yr)	0.038 – 0.097	0.029 – 0.092

*The leaching rate is estimated by dividing the free lime depth by the service years. The Free lime depth is the distance from the edge to the beginning of the purple zone (phenolphthalein test), measured at four equally spaced points along the circumference of the sample section.

Table 2 Distribution of detected active bacteria in the five groups and the four patina sub-layers

	A	B	C	D	Total
Cumulative population* ($\times 10^3$) (%)	55,800 (68.7%)	23,000 (28.4%)	970 (1.2%)	1,400 (1.7%)	81,170 (100.0%)
SLYM	68.4%	26.5%	0.2%	0.4%	95.5%
IRB	0.3%	0.4%	0.5%	0.4%	1.6%
HAB	ND	1.4%	0.1%	0.9%	2.4%
APB	ND	ND	0.4%	ND	0.4%
SRB	ND	ND	ND	ND	ND

* Cumulative populations were estimated from the predicted active cells per gram based on time lapses of the BART testers;
ND: Not detected.

Table 3 Bacterial community structure in the each patina sub-layer

	A	B	C	D
SLYM	99.5%	93.5%	18.9%	20.6%
IRB	0.5%	1.5%	42.0%	25.4%
HAB	ND	5.0%	8.9%	53.9%
APB	ND	ND	30.8%	ND
SRB	ND	ND	ND	ND

Table 4 Weight percentage of element distribution in the outer surface layer, central layer and inner surface layer of the AC pipe samples.

Element		C202-2			C311-2		
		Outer layer ¹ Wt. %	Central layer ² Wt. %	Inner layer ³ Wt. %	Outer layer Wt. %	Central layer Wt. %	Inner layer Wt. %
Calcium	Ca	16.5	21.2	2.90	19.8	18.0	5.08
Silica	Si	17.7	19.7	24.0	17.9	21.9	24.7
Alumina	Al	1.47	1.45	2.01	1.27	1.50	1.64
Iron	Fe	ND	1.00	ND	ND	0.90	ND
Magnesium	Mg	6.34	3.88	14.9	5.19	5.07	14.7
Potassium	K	ND	ND	ND	0.06	0.13	ND
Oxygen	O	58.0	52.3	55.8	55.8	52.5	53.9

¹ Average of two measurements (about 4.0 mm in thickness) from pipe outer surface;

² Average of two measurements (about 4.0 mm in thickness) from central zone with no obvious deterioration; and

³ Average of two measurements (about 4.0 mm in thickness) from pipe inner surface.

ND: Not detectable.

Figure 1 Phenolphthalein staining test measures deterioration depth

Figure 2 SEM photos of the outer, central and inner layers from the AC pipe sample

Figure 3 A SEM photo and four element distribution images of internal layers of a deteriorated AC pipe

Figure 4 Progression of AC pipe deterioration under the influence of microorganisms' growth.

Stage A: Conveyed water reduces the surface pH, allowing bacteria to grow on the inner surface of the pipe.

1. Free lime combines all components together.
2. Original calcium silicate hydrates (C2S and C3S).
3. Asbestos fibers.

Stage B: Active biological activity causes formation of patina and calcium-bearing mineral dissolution.

4. Active microbial activities decrease reduction-oxidation potential and pH, causing dissolution of Ca-bearing minerals, such as C2S and C3S, had horizontal voids.
5. Lime leaching zone.
6. Cracks caused by the formation of expandable minerals.

Stage C: The patina further extends to deep layers and surface layers become porous.

7. Dissolution continues moving as microbial activity extends to deep layers.
8. New cracks in deep layers.
9. Microorganism active zone.

Stage D. Severely deteriorated surface layer with concentrated asbestos fiber and increased porosity.

10. Asbestos fibers holding remaining particles together.
11. Porous layer with quartz particles;
12. Mineral weathering zone.