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A NOVEL EXPLORATION OF QUATERNARY SILANES (QAS) USE IN CONCRETE SEWER STRUCTURES FOR INHIBITING THE GROWTH OF MICROBES RESPONSIBLE FOR MICROBIALLY-INDUCED CORROSION IN CONCRETE (MICC).

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Synopsis

The different stages of *microbially-induced corrosion in concrete* (MICC) attack on concrete are explained to provide an accurate depiction of the problem. The efficacy of Quaternary Silanes against representative organisms responsible for *microbially-induced corrosion in concrete* (MICC) in concrete structures can vary at different pH levels depending on properties such as molecular chain length. An effective solution should combine QAS that offer the most robust efficacy with an optimal concrete compositional design to effectively combat MICC.

1. Microbially-Induced Corrosion of Concrete

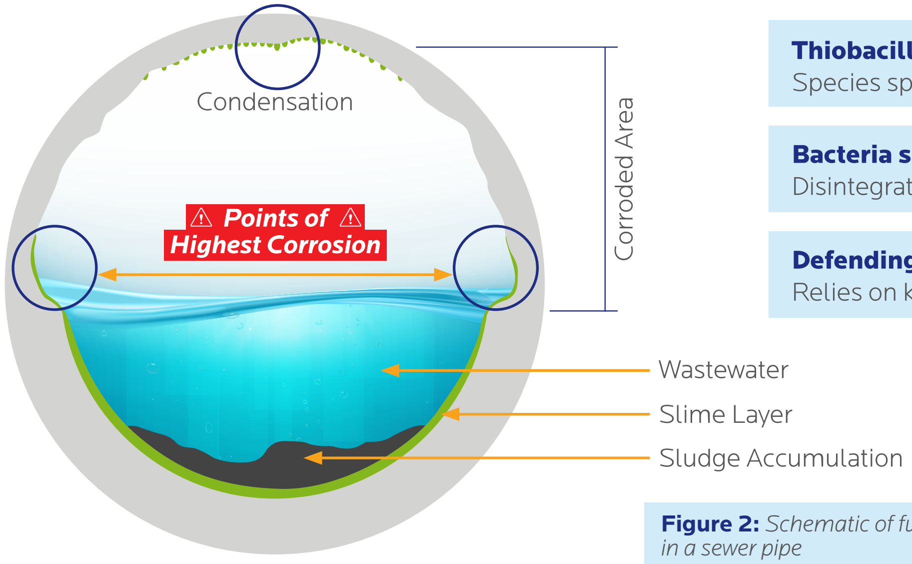
1. Mechanism of Acid Corrosion in Sewer Pipes

Figure 1: *Corroded Concrete manhole access to a sewer line [1]*

Concrete sewer pipes and wastewater infrastructures have been found to deteriorate at unexpectedly rapid rates due to chemical corrosion, as seen in Figure 1. Studies have shown that corrosion is due to high concentrations of sulfuric acid on the surfaces of concrete pipes and structures. This acid is generated by a complex procedure involving a multistage microbial process wherein sulfates in the raw sewage are anaerobically metabolized to generate hydrogen sulfide gas H_2S, which is in turn aerobically metabolized into sulfuric acid H_2SO_4[2]. Once established, the acid corrosion process proceeds as outlined in Figure 2:

1. Anaerobic organisms in the sludge under the sewage's surface metabolize sulfates in the sewage and release hydrogen sulfide (H_2S) into the liquid sewage stream.
2. Aqueous H_2S is released into the gas phase above the water line, where it is absorbed by the moisture layer on the concrete walls. Note: Areas of turbulence or high flow can increase the rate of H_2S release into the gaseous phase.
3. Species of *Thiobacillus* living on the walls metabolize the H_2S to produce sulfuric acid (H_2SO_4).
4. The sulfuric acid attacks (decalcifies) the cement paste component of the concrete to form gypsum and ettringite, both of which are expansive products with little structural integrity.
5. The low strength and expansive nature of the gypsum and ettringite leads to the dislodging of coarse aggregate from the concrete, exposing fresh surfaces to corrosive attack.

Note: The actual corrosion rate of concrete is depends on many factors unique to a given application, including the environmental conditions (which can fluctuate seasonally), sewage characteristics, composition and flow, structural elements of the infrastructure, and the design mix / material properties of the concrete itself[3]. Often, there are significant variations in the rate of corrosion within a single stretch of the same pipe due to localized turbulence in the sewage flow, increasing the rate of H_2S release into the atmosphere[4].



Thiobacillus bacteria
Species specific to MIC, live above waterline

Bacteria secrete sulfuric acid
Disintegrates & further lowers pH of concrete

Defending against MIC
Relies on keeping *thiobacillus* from thriving

Figure 2: *Schematic of fully developed acid corrosion mechanism in a sewer pipe*

1.2 Development of Bacterial Flora responsible for MICC attack

When concrete is freshly installed, none of the elements that support the corrosion process are present. These conditions develop slowly over time. It is important to understand how these elements effectively interfere with, delay, and inhibit the process with a view to maximizing the engineering life of a concrete installation. OPC (Ordinary Portland Cement) Concrete has a very high initial pH of 12.5 to 13.5 which provides intrinsic protection against microbial colonization. This is because MICC-causing organisms cannot tolerate this level of alkalinity. Abiotic chemical processes in the sewer system combine moisture and sewer gases to gradually lowering the pH of the concrete surface until the microbial flora that drives biogenic acid production can get a foothold. The development of the elements of the corrosion process is complex. It may involve more organisms than the *Thiobacillus* genus, but current understanding is that the *Thiobacillus* genus is the major contributor to the issue[5].

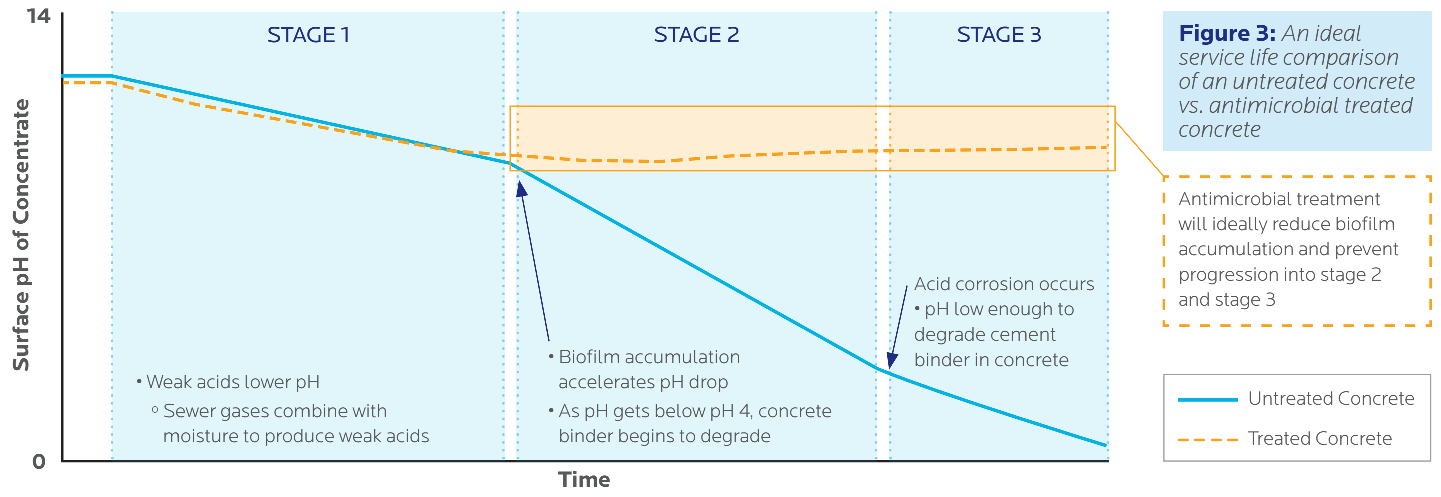


Figure 3: *An ideal service life comparison of an untreated concrete vs. antimicrobial treated concrete*

Antimicrobial treatment will ideally reduce biofilm accumulation and prevent progression into stage 2 and stage 3

To study and understand the development of the corrosion process, a model developed by Weiss and House [6]divides the engineering life of a sewer pipe into 3 distinct stages depending on the surface pH of the concrete (Figure 3):

1) Stage 1 is where freshly made OPC concrete is very alkaline (pH > 12), providing an inherent protection period against microbial attack. Sewer gases (mainly CO_2 and H_2S) combine with moisture to form weak acids that gradually neutralize the concrete surface, lowering the pH sufficiently to allow colonies of microorganisms such as *Thiobacillus* spp. to grow. Different concrete formulations offer varying levels of protection at this stage, as some have a relatively lower pH (i.e., an initial pH of ~10 rather than 12+).In such cases, the time span of Stage 1 protection may be drastically shortened, hastening the onset of Stage 2. The rate of pH decrease on the concrete surface during Stage 1 correlates strongly with initial pH, environmental conditions, and the sewage composition.

2) Stage 2 is characterized by an accelerating decline in the pH of the concrete. A complex and changing ecosystem of microbes inhabits the concrete surface as the pH continues to drop, hastened by the increasing amounts of sulfuric acid generated by these microbes' metabolic activities. However, the pH is not yet sufficiently low to cause concrete corrosion; only the pH reduction is occurring.

Species	Preferred pH Growth Range
T. thioparus	9 → 5
T. novellus	8 → 2.5
T. intermedius	8 → 2.5
T. neapolitanus	7 → 3
T. thiooxidans	3 → 0.5

Table 1: *The preferred pH ranges for Thiobacillus species*

The pH reduction is caused by a succession of *Thiobacillus* species (the main actors listed in Table 1), which prefer to grow at different pH values. At a higher pH, the neutrophilic (neutral pH-loving) organisms grow and produce sulfuric acid, which further lowers the pH until they die off and the acidophilic (acid-loving) species start to grow and further lower the pH[7].

3) Stage 3 is where the pH becomes low enough to actually lead to corrosion of the concrete, with failure becoming imminent and unpreventable. The sulfuric acid produced by the organisms converts the cement binder in concrete into non-load-bearing gypsum and ettringite. *T. thiooxidans* is the organism that is most damaging at this stage.

1.3 Common Approaches to Combat MICC

Most traditional approaches to combat MICC focus on fortifying the concrete against acid attack. These methods ignore the protection provided by the initial alkaline pH, which prevents biofilm establishment. True and comprehensive protection against microbial attack should also take into consideration the prolonging of Stages 1 and 2 to maximize inherent protection; by Stage 3, the concrete is actively corroding, and it is too late to abort the process. Traditional methods to fortify the concrete include adding densifiers such as GGBS (Granulated Ground Blast-furnace Slag) or silica fume to the concrete[8], which makes it stronger and more resistant to the ingress of water. However, densifiers do not necessarily make concrete more resistant to acid attack [9], as the pozzolanic reaction that incorporates the additives into the concrete binder system scavenges free $Ca(OH)_2$ out of the finished concrete. This can drastically lower the initial inherent pH and reduce the length of Stage 1. This result hastens the introduction of Stage 2 organisms and accelerates the structure towards Stage 3 corrosion.

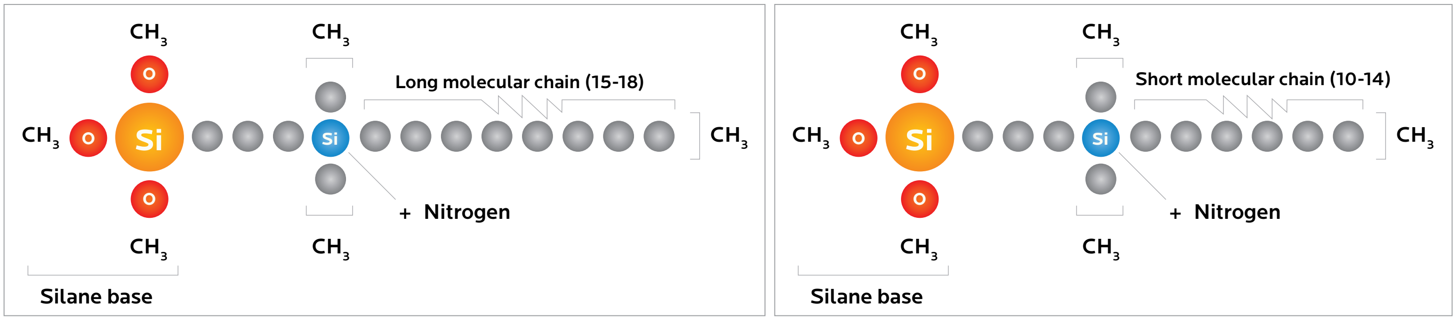
2. Antimicrobial Choice

2.1 Description of Quaternary Ammonium Chemistries

Quaternary Ammonium Compounds are well established in the antimicrobial industry. Due to their relatively low toxicity, good stability, and compatibility with other chemistries, quaternary ammonium compounds (QACs) have been used extensively for many decades for surface protection in a wide range of applications, such as disinfectants, textiles, hand washes and soaps, and swimming pool chemistries. Additionally, QACs are valued because many exhibit broad-spectrum effectiveness against bacteria, fungi, and selected viruses [10]. The efficacy of such chemistry against microorganisms is attributed to the positive charge centered around the nitrogen atom within the QAC molecule. Bacteria typically have a negatively charged cell wall. This can be readily compromised by the positive charge center of a QAC molecule, leading to leakage of essential cell metabolites and ultimately to its death. Mammalian cells are not susceptible to this type of attack; hence, quaternary ammoniums are frequently used in soap formulations for homes, institutions, and hospitals.

Quaternary ammonium silanes (QASs) are a specific type of QAC that incorporates a silane base, providing additional chemical benefits in specific applications. While QASs have been explored in depth in other applications, their benefits in concrete have not been thoroughly examined. For example, QASs can be added to concrete to improve corrosion resistance in a simulated marine environment [11]. Still, the intricacies of how their properties affect performance are not well modelled or quantified.

Figure 4: *Depicts an image of a long-chain versus a short-chain alkane chain*



Two main attributes make Quaternary silanes particularly suitable for concrete use:

1. The unique silane-based functionality at one end of the molecule is highly compatible with silicates and cementitious compositions. Under the right conditions the silane functional group binds and associates itself to concrete and aggregates, increasing the durability of such chemistries with respect to time.

2. Hydrocarbon chains can add a degree of hydrophobicity, which helps the molecule impart greater water resistance and thus enhances the anti-corrosion performance of the overall system. Differing chain lengths can influence efficacy depending on the application.

3. Evaluation of Microbial Effectiveness

3.1 Evaluation via C1904-2020 Test Method

In order to practically evaluate the efficacy of any antimicrobial in concrete,   one needs a testing protocol that can simulate many years of sewer service to effectively simulate the loss of the intrinsically high pH protection of Stage 1 concrete lifecycle. Most microbes, including *Thiobacillus*, are sensitive to the pH of their environment. As stated, the pH of many freshly-made concrete samples is high enough to prevent organisms from growing. This may lead to a false positive in a test, when bacterial kill is incorrectly attributed to the antimicrobial rather than to a high intrinsic pH. When preconditioning a concrete sample to lower its surface pH to support *Thiobacillus* growth, it is often observed that the pH quickly recovers and returns to its original high values during the course of testing. This is because such preconditioning only affects the concrete's surface, while the sample's core remains very alkaline. Over time (especially with moisture), the surface pH will rise again.

ASTM C1904-20, Standard Test Method for Determination of the Effect of Biogenic Acidification on Concrete Antimicrobial Additives and/or Concrete Products, establishes a laboratory procedure for evaluating how antimicrobial-treated concrete materials perform when exposed to microbially induced acidification. The method is designed to simulate real-world conditions in which microorganisms, particularly acid-producing bacteria, colonize concrete surfaces and generate acidic byproducts that can deteriorate the cement matrix. This controlled environment allows engineers and scientists to understand how well antimicrobial additives or treatments suppress microbial activity and slow the rate of degradation due to microbially induced acidification, while accurately mimicking actual applications.

The ASTM C1904-20 test method exposes concrete specimens, with and without antimicrobial additives, to microorganisms known to produce biogenic acids. Over a prescribed exposure period, microbial activity, surface pH, mass loss, visual degradation, and other chemical changes are monitored to quantify the extent of acidification and its effects on the concrete. The procedure ensures consistent inoculation, incubation, and measurement practices ensuring comparable results across materials and formulations. This provides a reliable means of determining whether antimicrobial technology effectively inhibits microbial acid production compared with untreated controls.

Ultimately, ASTM C1904 serves as a performance-based tool for assessing durability improvements in concrete materials used in microbially active environments such as wastewater systems, agricultural facilities, and industrial drainage structures. By quantifying the degree to which antimicrobial additives mitigate biogenic acidification, the method helps product developers, engineers, and municipalities determine the suitability of new formulations and guide material selection for long-term structural protection. The test results support informed decision-making by demonstrating how an additive performs under conditions that mimic the biochemical interactions causing real-world concrete degradation.

3.2 Evaluation of Quaternary Silane Chain Length on Microbial Efficacy in Concrete Recipes

Quaternary ammonium silanes (QAS) are widely used antimicrobial agents whose performance is strongly influenced by molecular structure, particularly the length of the attached hydrocarbon chain. In applications such as sanitation, disinfection, and hard-surface treatment, longer alkyl chains are associated with enhanced hydrophobic interactions and increased persistence on treated substrates. Within industries that desire quick efficacy (sanitation and disinfection) it has been found that varied chain lengths offer different performance, with the most desirable chain length being around 16 carbon chains [12]. However, the impact of QAS chain length has not been explicitly examined within concrete formulations, where high initial alkalinity, internal porosity, and biogenic acidification create a distinct and complex chemical environment. Understanding how chain length affects antimicrobial efficacy in cementitious materials is therefore essential for developing more robust additives suited to concrete exposure conditions.

3.3 Effect of Chain Length on pH-Dependent Performance

Experimental evaluation demonstrates that hydrocarbon chain length significantly influences the pH range within which the QAS remains active in concrete. As surfaces undergo biogenic acidification, as driven by microbial communities that generate acids, the local pH can decline from the initial alkaline conditions towards pH ranges that are more aggressively acidic. Fortunately, companies in the industry are advancing efforts to understand this process better. MarMac recently submitted candidate formulations to Oregon State University for evaluation via the C1904-20 test method to gain insight into the impact that the antimicrobial, specifically QAS, has on the concrete performance. The testing candidates included 1-alpha (short-chain analog) and 5-alpha (long-chain analog), with performance assessed relative to an untreated control.

Experimental results demonstrate that the longer-chain QAS maintains antimicrobial activity down to approximately pH 4, whereas 1-alpha (a shorter-chain analog) shows a similar pH response to the untreated control (Figure 5).

At pH 5, it can be seen that 5-alpha shows significantly improved pH response at pH 5 (Figure 6). The long chain QAS maintains a higher pH throughout the duration of the test, demonstrating stronger prevention of microbially induced corrosion conditions.

This performance differential suggests that extended hydrophobic chains enhance both molecular stability and retention within the concrete matrix. Stronger adsorption to pore surfaces and reduced susceptibility to acid-induced deactivation likely contribute to the broader effective pH window observed for long-chain variants.

3.4 Implications for Concrete Durability and Additive Selection

The expanded working pH range of long-chain QAS has direct implications for improving the durability of concrete exposed to microbially active environments. Antimicrobial additives that are capable of maintaining efficacy deeper into the acidification cycle can help delay microbial colonization and reduce the onset of biochemical mechanisms leading to concrete deterioration. As a result, hydrocarbon chain length becomes an important design parameter for antimicrobial formulations intended for cementitious systems. Longer-chain QAS provides enhanced resilience under acidic stress, making them better suited for applications where materials must withstand the conditions demonstrated in ASTM C1904-20 environments.

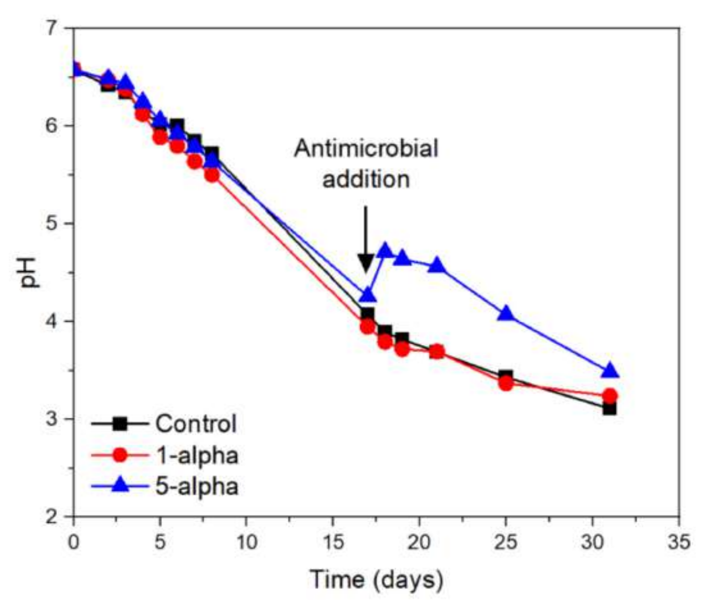


Figure 5: *C1904 Response - Antimicrobial Addition at Concrete pH of 4*

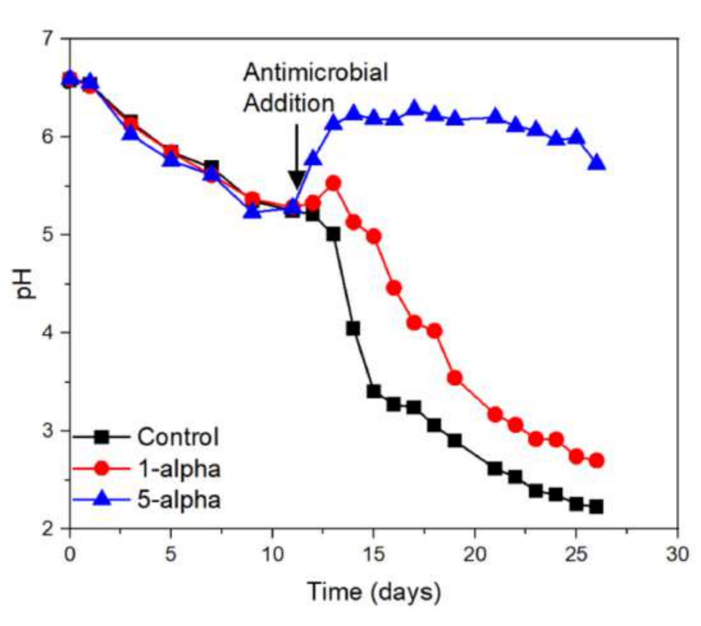


Figure 6: *C1904 Response - Antimicrobial Addition at Concrete pH of 5*

4. Concluding Remarks

The proper selection of concrete composition and MICC protection technology to be deployed in the MICC-resistant concrete layer is essential. Based on the preceding discussion, the following attributes are highly desirable in materials and technology selection:

1. **Select a concrete composition that offers superior Phase 1 protection.** i.e., this means that the concrete should have a high starting pH, and be able to retain that high intrinsic pH for a long period of time. A pH of 12 or higher is suggested to maximize this property. Concrete formulations with high intrinsic pH provide valuable protection against MICC, which can remain effective for significant periods of time. Conversely, concrete with a lower initial pH, or concrete that is easily neutralizable, is at risk of premature microbial attack. It is strongly believed that a high intrinsic pH should be the paramount selection criteria for MICC-protected concrete, rather than infusing concrete with additives to impart increased strength or increased densification of the concrete, as these measures do not directly address this critical stage.
2. **Infuse the concrete with an appropriate antimicrobial that can offer Phase 2 protection.** Here, the choice of an antimicrobial must be properly vetted through a systematic process, as discussed in the preceding paragraphs: (a) an initial determination through a standard Minimum Inhibitory Concentration test as proof positive of antimicrobial efficacy, and (b) an appropriate antimicrobial test that compares a concrete composition treated with the antimicrobial against a concrete with a similar composition without the antimicrobial agent, to be done under test conditions with low enough pH to support the growth of the relevant test organism. The ideal antimicrobial shows efficacy across a broad pH range. This allows for efficacy to be maintained over a longer duration. The exact correlation between microbially induced corrosion of concrete and the overall lifespan of the concrete structure will require future studies for a deeper understanding. However, the literature suggests that, in applications such as marine concrete structures, the expected service life of 100 years can be reduced to as low as 30 years without such protection [13].

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