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REVIEW ARTICLE

MICROBIOLOGICALLY INFLUENCED CORROSION OF CONCRETE: A REVIEW

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ABSTRACT

Corrosion is one of the major issues these days in various sectors of the industries related to construction, sewage disposal, food and manufacturing. Microbiologically induced corrosion (MIC) is a key factor for infrastructure degradation worldwide, which is actually a result of wastewater collection. It is the most common form of corrosion, which is aided by the presence, and activity of microorganisms on the surface of the materials. MIC has hiked demands for sustainable and cost effective construction material on a global level. This review focuses on the reasons and effects of MIC in the environment specifically highlighting the adversity of MIC on cement based structures. Various research works have been discussed for reducing the effect of MIC and thereby increasing the durability of structures.

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INTRODUCTION

Microorganisms present in the wastewater streams interact with surface of materials both chemically and physically depending upon the surrounding conditions. This interaction leads to the corrosion of target materials. So the collection and cost effective treatment of wastewater is the basic step towards maintaining the sanitary standards and reducing corrosive nature of the degraded water (Hvitved-Jacobsen, 2013). Microbial induced corrosion leads to degraded infrastructures as well as numerous diseases in the environment. Most developing countries lack proper networks for sewage and treatment of wastewaters (Hvitved-Jacobsen, 2002). thus they fall prey to the adverse effects of MIC which in turn increases their economic expenses as well. As a matter of fact, MIC is an issue worth many billion dollars annually for oil and gas industries in the states (Setareh, 2000). Table 1 shows some industrial situations where MIC can have its effect. MIC has been stated in many literatures directly focusing on the role of microorganisms in corrosion of metals, alloys and cement based materials surrounded thoroughly by aqueous environment (Peng, 1994). MIC is a very complex process and this results in the failure of materials in justifying their proposed lifetime of operation (De Belie, 2004). In this process microbes slowly attach to the materials thereby forming a biofilm which has the potential to bring in diverse changes in the already existing surface chemistry of the target substrate (Almahamedh, 2008).

Microorganisms of interest include different classes of the Bacteria, the Eucarya and the Archaea out of which the role of Sulphate Reducing Bacteria has been studied thoroughly till date (Almahamedh, 2008). Concrete being widely used around the globe for various construction purposes is more prone to a multistage degradation also called as microbial induced corrosion[6]. Concrete is a composite material having porosity and highly alkaline nature (pH around 13) (Alexander, 2013). It is used in large scale for the construction of sewage pipelines, wastewater collection and treatment infrastructures. Prolonged exposure of such material to wastewaters results into development of biofilms and reduction of sulphates to sulphides which lays the foundation for MIC.

The following steps depict how MIC leads to degradation of exposed concrete materials.

- Initial step includes production of aqueous hydrogen sulphides ($H_2S_{(aq)}$) from sulphates with the help of sulphate reducing bacteria under anaerobic conditions. This process takes place below the waterline.
- Turbulence in the aqueous medium results in the release of $H_2S_{(gas)}$ from the sulphides.
- Above the waterline, the gas liberated partitions with moisture content on the surface and is oxidized to sulphuric acid. The aiding bacteria in this process is usually *Thiobacillus*.

- The acid produced attacks the concrete material thereby decreasing the pH value and results into decalcification of calcium hydroxide a formation of gypsum and other expansive corrosion products.
- As the attack proceeds further, thickness of concrete is reduced and the service life of material is compromised.

Table 1. MIC in Industrial environments

Sr. No	Industries	Areas affected
1	Chemical	Pipelines, Tanks, Condensers, Joints, Heat exchanger
2	Civil	Concrete in marine, fresh water and sub-soil conditions, buildings
3	Aviation	Aluminium fuel tanks
4	Nuclear and thermal	Cooling water tubes and pipes, sub-sea pipe lines, copper-alloys
5	Mining	Underground machinery and engineering materials

The important aspect to be understood is that this series of steps for MIC is preferred for most of the materials but the degree of corrosion strictly depends on the environmental conditions, material characteristics and nature of wastewater used (Hvitved-Jacobsen, 2002). Figure 1 shows the pictorial representation of the above steps.

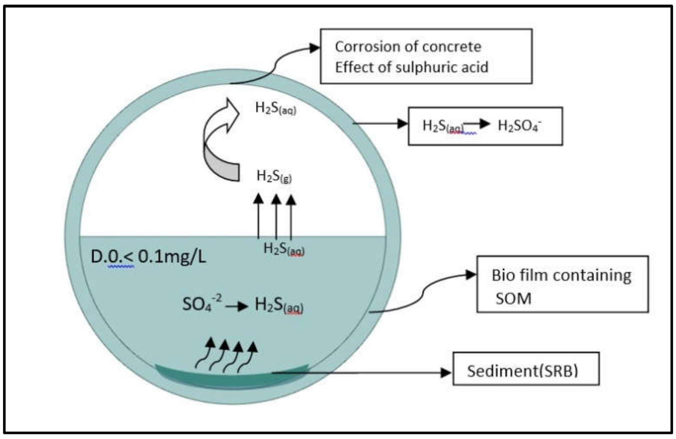

Figure 1. Steps involved in MIC degradation

Regardless of all the studies done till date, the exact mechanism of MIC is yet to be defined. Some mathematical models have been devised to study concrete system undergoing corrosions but they are too limited to certain categories and geometries of the infrastructures. The complex nature of MIC makes it vulnerable to shortcomings in laboratory imitations of the same. As a result, the methods become time consuming, less effective and costly. Lack of sufficient knowledge on the initial process of microbial colonization is reason for failure in producing a sustainable product till date. So moreover, simplified approaches are followed resulting in to inconsistent and manipulated results. This paper aims on putting forward a review in context to the process of MIC along with its causes, after effects and mitigation techniques. The paper focuses broadly on the effect of MIC on concrete based wastewater systems.

Generation of sulphides: The generation of sulphides is a necessary step for the initiation of MIC. There must be presence of dissolved sulphides in the wastewater streams. Aqueous hydrogen sulphides are produced by the activity of anaerobic SRB species like *Desulfovibrio* and *Desulfomaculum* (Barton, 2009). These species are present in the biofilms made up of extracellular polymeric substrates[13]. It is actually a result of accumulation of microbes within a thickness ranging from 0.3mm to 1.0mm for a prolonged time period on a surface[14]. Majority of the reduction takes place in these films and presence of both organic substrates (electron donor) along with sulphates (electrophile) is required for the generation of sulphides. Equation 1 refers to the reduction of sulphates by SRB.

$$SO_4^{2-}(aq) + 2C + H_2O \rightarrow 2HCO^{3-}(aq) + H_2S(aq) \qquad (1)$$

The dissolved oxygen diffuses into the biofilm which is further taken up by the sulphate oxidising molecules. This build a gradient of DO vanishing near the wall of concrete system. Now this facilitates anaerobic environment feasible for generation of sulphides with the aid of SRB. Factors affecting the sulphide content in waste water include.

- **DO**- As the amount of DO decreases towards the walls, activity of SRB species increases since they work efficiently in anaerobic conditions. Corrosion is less where DO is present in appreciable amount[15].
- **Turbulence**- Some turbulence is necessary as it boosts re-aeration resulting into release of sulphide gas. Increased turbulence leads to increased liberation of the gas.
- **Temperature**- Increased temperature reduces amount of DO and builds up preferable environment for the activity of SRB species.
- **BOD and Sulphate content**- More the amount of sulphate content present in waste water, more will be the amount of sulphide generated as well as high BOD will encourage colonization of microbes.

The flow of the stream also plays role in formation of biofilms. Networks which are devoid of abrasions and weathering due to weak or irregular flow of streams have relatively thicker biofilm.

Partitioning of aqueous H_2S into air: For the release $H_2S_{(aq)}$ into gas phase, the main factors responsible is the pH of the aqueous medium along with temperature and equilibrium condition between the phases. Equation 2 shows relevant dissociation behaviour during phase transfer from liquid to gas-

$$H_2S_{(aq)} \rightarrow HS^-_{(aq)} + H^+ \qquad \text{…………….(2)}$$

Figure 2 shows that when pH starts falling below 7 i.e. towards the acidic side, $H_2S_{(aq)}$ dominates all other species[17,18]. Concentration acts as a driving force in the transition of phases which is governed by Henry's law

$$S_g = kP_g, \qquad \text{………………(3)}$$

where S_g is the molar concentration of H_2S present in solution, k represents applicable Henry's Law constant and the term P_g represents the partial pressure of the H_2S liberated over the solution. Figure 3 gives a detailed idea about equilibrium concentration of gaseous H_2S in a closed system as a function of temperature. The main idea derived from the graph is that when temperature is increased the solubility of gas decreases appreciably and the concentration of gas in the system increases.

Another factor that influence release of H_2S gas into the sewer system is turbulence which can be result of high velocities, poor sewer constructions, sudden drops. It increases the surface area of the water-air interface thereby promoting the liberation of the gas. Systems having higher turbulence experience more corrosion than those which are stable in flow. Ventilation also effects the liberation of gas but at the same time may not encourage gad build-up in a specific volume.

Conversion of H_2S to sulphuric acid: After the release of H_2S gas in the open space of the sewer, it interacts with the moisture films present on the surface of water. $H_2S(aq)$ acts as a precursor in production of sulphuric acid irrespective of the path chosen to produce sulphuric acid. The $H_2S(aq)$ can undergo both biotic and abiotic conversion during oxidation ultimately producing sulphuric acid as the end product. Local pH also influence the oxidation states of sulphur and alters activity of SOM (Islander, 1991). At very low pH direct elemental sulphur is obtained from $H_2S(aq)$ whereas sulphate (SO_4^{2-}) is obtained at relatively higher pH value with thiosulphate ($S_2O_3^{2-}$) acting as the intermediate product of the conversion. A scientist named Parker in 1947 discovered the SOM molecule i.e. *Thiobacillus Concretivorous*, majorly contributing to the production of sulphuric acid as a side result if which pH of the targter surface fell as low as 1. The reduction of pH to such levels indicates the presence of more than one SOM species on the substrate. Such SOMs has been notably confirmed in different research works published over the years (Davis, 1998). Table 2 shows some variations of *Thiobacillus* species their preferred pH range and substrate[21]. It is also a matter of fact that *Thiobacillus* bacteria are not the only species residing on a concrete surface rather there is a group of fungi, lichens, aerobic heterotrophs also found on such surfaces.

Table 2. The preferred substrates and pH range for growth of different SOM

Sr. no	Species	Preferred substrate	Preferred pH range for growth
1	*T. thioparus*	H_2S, S^0, S_2CO^{3-}	5-9
2	*T. novellus*	S_2CO^{3-}	2.5-8
3	*T. intermedius*	S_2CO^{3-}	2.5-8
4	*T. neapolitanus*	S^0, S_2CO^{3-}	3-7
5	*T. thiooxidants*	H_2S, S^0	0.5-3

The abiotic lowering of concrete surface is necessary step since the fresh concrete surface is highly alkaline and has a pH range moreover in between 12.5-13.5[22]. Also to what extent the initial pH value gets reduces is still a debatable topic. For better understandings, Carbonation is actually given credits for the initial lowering in the pH value. Colonization starts only when pH value slips down to 9. Bacteria like *T. thioparus* also called neutrophilic sulphur oxidising microorganisms (NSOM) associated to mild degrees of corrosion (Davis, 1988) whereas on the other hand acidophilic SOM like *T. thiooxidants* produce a very acidic environment and results in intense corrosion along the walls of sewer pipes.

Acid Degradation: No concrete material till date has fully withstand the acidic outburst on its surface due to MIC since the chemical composition of the Portland cement makes it highly prone to acidic degradation (Duchesne, 2013). Volume composition of the paste of Portland cement is given in Table 3 (Mehta, 1993).

Table 3. Composition of Portland cement

Sr. no.	Constituents	Volume percentage (%)
1	Calcium silica hydrate	50-60
2	Calcium hydroxide	20-25
3	Calcium sulfoaluminates	15-20
4	Cement Grains	Varying proportions

The attack of acid results into decalcification of the structures resulting into increased porosity and decreased mechanical strength (Pavlík, 1994a). This degradation is actually affected by the type of acid attack the surface has to bear. Moreover, the resistant capability of a surface towards such acid attacks is majorly dependent on the porosity, acid neutralization value and composition of hydrated products. To understand the effect and intensity of acid attacks by the SOMs, they are purely classified as Active and Passive acid attacks. The active attacks are a result of prolonged and continuous exposure of the surface to acid. Such is the impact that alkalinity present in the substrate isn't enough to neutralize the acid. Example include degradation of sewer pipe where generation of acid is a continuous process. Figure 3 shows the process of such attacks. On the other hand, periodic or occasional exposure of substrates to a finite acidic environment in various industries is an example of passive attack. In passive attacks, the rate of corrosion is controlled by the product layer which limits the diffusion of acid towards the where reaction is yet to take place (Pavlík, 1994b). Figure 4 shows the pictorial representation of passive attacks. The role of this product layer is changed in active attacks. While the normal diffusion of acid towards unreacted zone does takes place, there exists evidences of the growth of SOM through the thickness of the layer (Davis, 1998). The layer comprises of ASOM throughout the thickness and NSOM merely present in the outer regions. The strength of acid also influences the MIC process. Mild acidic solutions lead to leaching of calcium ions over a long period of time. This results into decreased structural integrity and mechanical aspects (Carde, 1996). When conditions are highly acidic, dissolution of calcium hydroxide takes place along with the decalcification. So it can be understood that weak acids produce insoluble salts where as strong acids produce more soluble salts (Pavlík, 1997). In case of sulphuric acids, gypsum and ettringette have been reported as the end products (Monteny, 2000). These produce cracks within the matrix of concrete system.

MIC: A Three-Stage Process: For simplifying the concept of MIC, the process has been divided into three separate based on a theoretical model[32]. Figure 5 shows the relevant information. The first stage includes the abiotic lowering of the pH of concrete substrate. The pH reduction occurs as a result of carbonation and attacks of acids produced due to oxidation of H_2S[33]. There is no eminent loss of material at this stage. In the second stage, the concrete substrate experiences further lowering of pH at the surface. This is basically due to the attachment and growth of *Thiobacillus* species on the surface. The rate of colonization is directly accelerated if the level of $H_2S_{(g)}$ is increased. The third stage refers to the condition where the decrement in surface pH levels reach to such lows that concrete may experience acid attacks. This is the stage where massive loss of material with a corrosion rate > 10mm/year (Grengg, 2015). Such severe corrosion takes place in presence of H_2S gas and there is a strong belief that the gas plays a vital role in controlling the rates of corrosion.

Table 4. Use of metals as antimicrobial agent

Sr. no.	Material used	Metal addition	Setup	Effect	Reference
1	Ordinary Portland Cement	Ni	In situ experiments	Significant inhibitory effects on various strains of neutrophilic bacteria.	(Negishi et al., 2009)
2	Zeolites	Ag, Zn, Cu	Laboratory testing	Binary system of Cu/Zn , same bacteriostatic efficiency as Ag	(Kaali et al., 2011)
3	Geopolymer	Ag	Laboratory testing	99% mortality of gram positive/negative bacteria	Adak et al., 2015)

Though, not much has been reported on the exact relationship of concentration of H_2S gas with the corrosion rate.

MIC mitigation: Various processes have been opted to reduce or control the process of MIC. They are

- Physical methods
- Electrochemical methods
- Chemical methods
- Other biological treatments

Physical methods: This includes removal of biofilms using mechanical forces. It may not result into complete destruction of the MIC base.

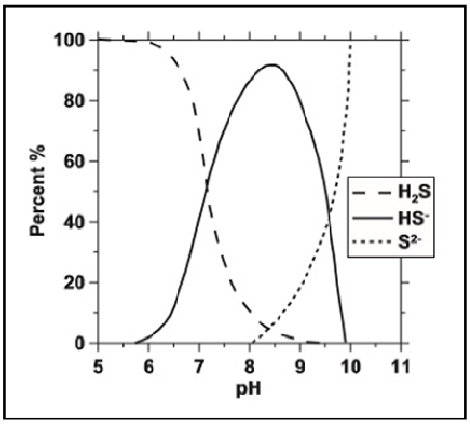

Figure 2. Equilibrium speciation of aqueous hydrogen sulphide as a function of pH (after U.S. center for environmental research information, 1985)

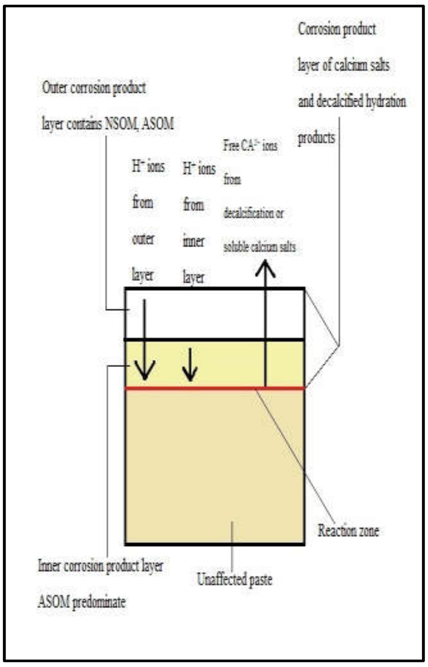

Figure 3. Active Acid attacks on a concrete paste by SOM

Pigging: Cleaning and inspection of pipelines of gas and oils with the help of a pig is known as pigging. It is notanideal method as it does not completely scrap out the biofilm and in various situations these aren't able to reach sites due to shape change or unfavourable geometry of the substrate.

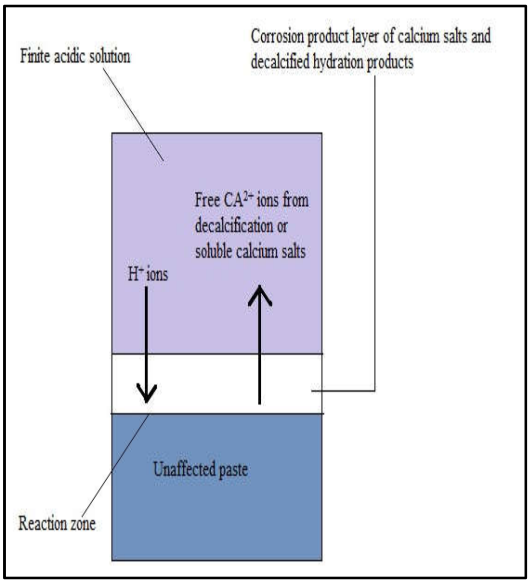

Figure 4. Passive attack on cement paste

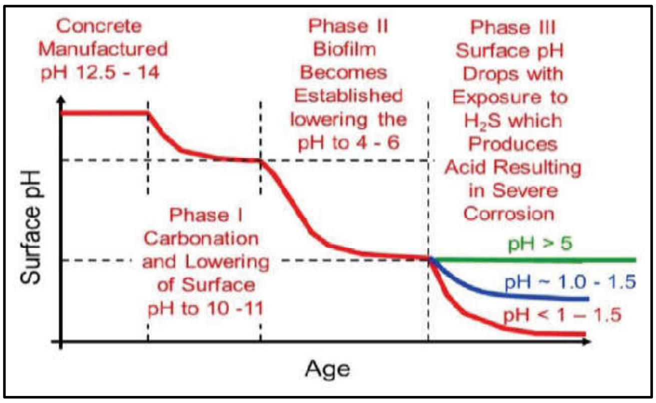

Figure 5. Three-stage MIC process

Ultrasonic treatment: Ultrasonic destroys the membranes of the cell which forms the building block of the mechanism. This inhibits any further growth of the microbes and kills the existent ones present on the surface of the target substrates. The basic principle of ultrasonic treatment includes application of acoustic pressure and developing a bubble with appreciable capacity. The demerit includes damage to the surface itself (Pound, 2005).

Chemical methods: Chemical compounds that can kill microorganisms present on the surface are used. They are called as biocides and can be of two types, oxidizing and non-oxidizing biocides.

Example of oxidizing is chlorine and non-oxidizing is aldehydes. These bear the capability to penetrate and kill the cells of microbes. These compounds can be expensive and may cause corrosion to the surface itself.

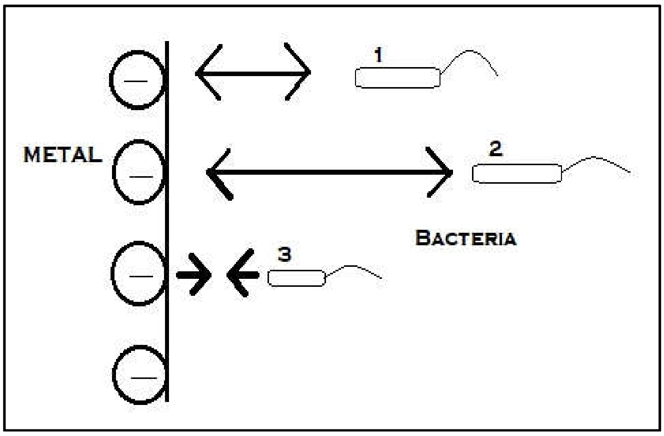

Figure 6. Electrostatic method for MIC mitigation

Electrochemical methods: One of the most common methods to prevent corrosion on alloys and various metals. The structure material is made as the negative terminal i.e. cathodic protection. According to the electrostatic chemical theory, negative potential is applied on the metal surface, which repels the incoming negatively charged bacteria. Figure 6 shows the same. Along with that the release of hydroxyl ion into the solution due to the cathodic reaction increases the alkalinity of the system inhibiting the growth of microorganism (Javaherdashti, 2008).

Biological treatments: This is actually a recent scheme in order to reduce microbiologically induced corrosion. The proposed mechanisms focus on use of other bacterial species against the primary bacteria involved in MIC. Some studies have mentioned use of nitrate reducing bacteria in order to eliminate SRB.

Antimicrobial agents: Use of antimicrobial agent to reduce the effect of MIC has been well documented in various research works. In the recent times, antimicrobial agents have been exclusively used in commercial concretes to increase durability and resistance towards the growth of SOBs. The most widely used approaches include the use of heavy metals like Co, W, Al, Fe, Mn, Mo, Zn, Cu, V, Cr as antimicrobial agents due to their inhibitory efficiency, affinity towards sulphur, and interaction with microorganisms present on the surface of the target surface. The heavy metals react with the cell wall of bacteria which is actually negatively charged. This leads to formation of complex compounds within the bacterial membrane. Such reactions reduce the sensitivity of enzymes responsible for microbial growths and as a result rupture the periphery leading to leakage of intracellular matrix. The application of zeolites with exchange of cations has shown promising results for concretes exposed to aggressive wastewater environement. The binary system constituting Cu/Zn has a greater efficiency to enhance antimicrobial activities of materials exposed to attack of microorganisms. Table 4 below shows the effective use of heavy metals as antimicrobial agents and their applications regarding the same. Variousstudies have pour into a common thought of focusing more on the growth control of NSOBs since they are more responsible to appreciable decrease in the pH from 9.5 to approximately 4 during MIC propagation. Along with that, the other effect of heavy metal on the concrete should also be considered for future research purpose.

Coatings: Every pipeline set under the ground for sewer or other uses is coated with some kind of protective layers. When these coatings are destroyed due to numerous reasons like physical abrasions, high flow leads to the onset of corrosion. Recent developments in order to make these coatings more effective and tolerable include the use of adherent, continuous and mechanically strong polymer material (Javaherdashti, 2008). Some examples of highly stable polymer are polystyrene, epoxy resin, polyethylene.

Conclusion

Microbiologically induced corrosion is a serious threat to the infrastructures all around the world. Concrete, metal and alloy surfaces when exposed to wastewaters corrode due to the activity of microorganisms. The effects of MIC can be prominently seen in industries like civil, chemical, aviation and many others involving heavy infrastructures exposed to aggressive environments on regular basis. No commercially available structure is efficient enough to completely stand against microbial degradation. Lack of proper knowledge on the exact mechanism of MIC has restricted new updates in the target surface characteristics. Recent techniques to treat MIC aren't fully efficient as well. Future research should focus more on understanding the microbial ecology and deducing techniques to reduce the concrete-microorganism interaction.

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