


# A review: microbiologically influenced corrosion and the effect of cathodic polarization on typical bacteria

Meiying Lv · Min Du 

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**Abstract** Microbiologically influenced corrosion is a serious type of corrosion as approximately 20% of the total economic losses. *Sulfate reducing bacteria* and *Iron oxidizing bacteria* are one of the typical representatives of the anaerobic and aerobic bacteria, which are ubiquitous in natural environments and corrode steel structures. Cathodic polarization has been recognized as an effective method for preventing steels from microbial corrosion. Although cathodic polarization method has been widely studied, the specific properties of cathodic current that influences the bacterial removal and inactivation remained largely unclear. This review is to show the main effects of *Sulfate reducing bacteria* and *Iron oxidizing bacteria* on metal decay as well as the inhibition mechanism of cathodic polarization in the study of bio-corrosion.

**Keywords** Microbiologically influenced corrosion · *Sulfate reducing bacteria* · *Iron oxidizing bacteria* · Biofilm · Cathodic polarization

## 1 Introduction

Microbiologically influenced corrosion (MIC) can accelerate deterioration of metal alloys in a wide range of industries such as marine engineering infrastructure, recirculated cooling systems, and oil/gas pipelines, etc. (Jogdeo et al. 2017; Little et al. 1992; Liu and Frank Cheng 2017; Sheng et al. 2007). Microorganisms exhibit extreme tolerance to hostile environments such as acidic and alkaline pH, low and higher temperatures as well as pressure gradients. It is estimated that up to 20% or more of corrosion losses can be attributed to MIC (Fatah et al. 2013). The major characteristics of some anaerobic and aerobic microorganisms are illustrated in Table 1. A number of different groups of microbes carrying out metabolisms such as sulfate reduction (Li et al. 2001), methane production (Boopathy and Daniels 1991), and Fe(III) reduction (Duan et al. 2008) have been implicated in MIC, and these processes can enhance metal dissolution and produce cathodic conditions that cause surface pitting which leads to more rapid corrosion. While the physiological attributes of different microorganism community are generally understood, less is known about the process of MIC. One of the explanations for the difficulty in reaching an adequate understanding of MIC was the lack of an appropriate transfer of knowledge among different areas, including microbiology, metallurgy, electrochemistry, and chemical engineering. Unraveling the

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M. Lv · M. Du (✉)  
The Key Laboratory of Marine Chemistry Theory and Technology, Ministry of Education, College of Chemistry and Chemical Engineering, Ocean University of China, Qingdao 266100, People's Republic of China  
e-mail: ssdm99@ouc.edu.cn; sdm06@126.com

**Table 1** Some anaerobic and aerobic bacteria implicated in MIC

Bacteria	Tem (°C)	pH	Characteristic	Function	References
<i>Sulfate reducing bacteria</i>	20–40	5.0–9.0	Anaerobic	Reduce $\text{SO}_4^{2-}$ to $\text{S}^-$ and $\text{H}_2\text{S}$	Li et al. (2001)
<i>Iron reducing bacteria</i>	10–50	4.0–8.0	Anaerobic	Reduce insoluble Fe(III) to soluble Fe(II)	Duan et al. (2008)
<i>Nitrate reducing bacteria</i>	5–40	5.5–9.5	Anaerobic	Reduce nitrate to ammonium and $\text{N}_2$	Jia et al. (2017a, b)
<i>Methanogenic bacteria</i>	20–40	5.4–7.2	Anaerobic	Produce methane using elemental iron as the sole source of hydrogen, via cathodic depolarization	Boopathy and Daniels (1991)
<i>Acid producing bacteria</i>	20–40	4.5–8.0	Anaerobic	Produce fermentation metabolites, such as volatile fatty acids	Xu et al. (2016)
<i>Iron oxidizing bacteria</i>	20–40	6.0–8.0	Aerobic	Oxidize Fe(II) to Fe(III)	Liu et al. (2016a)
<i>Sulfur oxidizing bacteria</i>	20–40	6.5–7.5	Aerobic	Release aggressive metabolites, such as acetic acid, succinic acid, sulfuric acid	Okabe et al. (2007)

complex interaction between microorganisms, the metal surface they inhabit, the metabolite they produce, and how they affect the corrosion process are all important in MIC research.

Microorganisms associated with corrosion damage are anaerobic and aerobic bacteria. *Sulfate reducing bacteria (SRB)* and *Iron oxidizing bacteria (IOB)* are one of the typical representatives of the anaerobic and aerobic bacteria, which are ubiquitous in natural environments and corrode steel structures (Ashassi-Sorkhabi et al. 2012; Sand and Gehrke 2003). *SRB* are commonly considered the main culprits of MIC because there is the wide availability of sulfate ( $\text{SO}_4^{2-}$ ) in environment (e.g., anoxic seawater) (Abbas et al. 2013; Moon et al. 2007). Anaerobic *SRB* may tolerate exposure to oxygen ( $\text{O}_2$ ) for a considerable time until they adhere to the surface of substrate (Zhang et al. 2015). *SRB* use  $\text{SO}_4^{2-}$  as the electron acceptor and reduce it to  $\text{S}^{2-}$  ions or hydrogen sulfide ( $\text{H}_2\text{S}$ ) (Li et al. 2001). MIC caused by the aerobic bacteria has also been adequately studied (McBeth et al. 2011; Scotto et al. 1985; Yuan and Pehkonen 2007). *IOB* are another major type of corrosive bacteria, causing serious corrosion damages (Maeda et al. 1999; Wang et al. 2014). *IOB* oxidize ferrous ions ( $\text{Fe}^{2+}$ ) to ferric ions ( $\text{Fe}^{3+}$ ) with oxygen ( $\text{O}_2$ ) as the terminal electron acceptor for energy generation in their metabolism (McBeth et al. 2011; McBeth and Emerson 2016). Under biocatalysis by *IOB*, the oxidation rate of  $\text{Fe}^{2+}$  is much faster than the abiotic

chemical oxidation reaction (Liu et al. 2016a). Thus, *IOB* accelerate the dissolution and localized corrosion of steels. *IOB* can deposit ferric hydroxide ( $\text{Fe}(\text{OH})_3$ ) on the steel surface which can lead to crevice attack and can also provide a suitable environment for the anaerobic bacteria in the region beneath the tubercle to accelerate corrosion. The most aggressive MIC occurs with natural populations containing several bacterial species rather than a single species (Liu et al. 2014; Sung et al. 2011). For example, the interaction between *SRB* and *IOB* could together accelerate the pitting corrosion process of steel (Xu et al. 2008). Due to the consumption of  $\text{O}_2$  by aerobic *IOB*, the region within biofilm becomes anaerobic easily and anaerobic *SRB* can proliferate in the locally anaerobic conditions beneath biofilm to accelerate and facilitate corrosion.

MIC is mainly caused by the formation of biofilm (Flemming 2002; Sheng et al. 2007; Videla and Herrera 2005; Wang et al. 2004). Once established on the substratum surface, biofilms are often difficult to control and are almost impossible to eliminate with conventional microbiological control strategies with antimicrobial agents and disinfectants (Costerton et al. 2003). For this reason, it is generally recognized that one of the most effective strategies to control biofilms formation is to prevent adhesion of bacteria at initial steps (Bos et al. 1999). Cathodic polarization (CP) is an effective and commonly used method to prevent metallic facilities from corrosion by applying

impressed current to a metallic structure and allowing only the cathodic oxygen reduction reaction to occur. The CP method used for controlling bacterial adhesion is considered to be environmental friendly because it uses electrons as the nontoxic reaction mediator. This method can be widely used in aqueous systems especially useful in marine environments or certain soils where the medium conductivity is high enough to obtain a uniform current distribution over the metal surface for protection. Some investigations suggest that CP can inhibit MIC (Edyvean et al. 1992; Liu and Cheng 2017; van der Borden et al. 2004). However, there is still much controversy with respect to the possible inhibitory mechanisms of CP on MIC (Istanbulu et al. 2012; Liu and Cheng 2017; Pérez et al. 2009).

This review describes the current understanding of MIC mechanisms caused by *SRB* and *IOB*, the process of biofilm formation and the role of the bacterial biofilm in both corrosion and mitigation response, and finally the inhibitory effects of CP on MIC.

## 2 The corrosion mechanisms of typical microorganisms

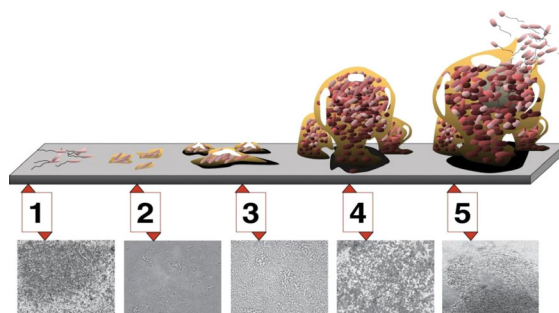
### 2.1 The formation process of biofilm and its effect on steel corrosion

Abiotic corrosion can be divided into physical, chemical and electrochemical corrosion. MIC is defined by no single characteristic. It is a complex biogeochemical process initiated and/or exacerbated by the presence and activity of bacteria and their associated biofilms (Castaneda and Benetton 2008; Stadler et al. 2010). Thus, MIC is considered biocorrosion, and the terms “microbes” and “bacteria” are used interchangeably. Bacteria in the environment exist in the planktonic state, which tends to attach to material surfaces and form a biofilm layer, which consists of a community of bacteria, water, extracellular polymeric substances (EPS), corrosion products, etc. (Alabbas et al. 2013). The initial event of bacterial attachment is important since it lays the foundation for the formation of heterogeneous biofilms. Many surface characteristics such as free energy, charge and roughness can modulate bacterial adhesion (Rosenberger and Kraume 2002). Both reversible and irreversible attachment are possible. Figure 1 shows

a schematic process of biofilm development (Chitra et al. 2014): (1) initial attachment of planktonic cells to the metal surface, (2) irreversible attachment of bacterial cells through formation of EPS, (3) formation of a biofilm layer, (4) maturation of a steady-state biofilm layer, (5) detachment of bacterial cells from the biofilm outer surface.

In some instances, a compact microbial biofilm can act as a diffusion barrier for reactants such as oxygen, aggressive anions and cations (Chongdar et al. 2005; Grooters et al. 2007; Little and Ray 2002). But, the dynamic development of biofilm may also increase corrosion. Biofilm mediate interactions between metal surfaces and the liquid environment, leading to major modifications of the metal–solution interface by drastically changing the composition and concentration of chemical species, pH, and oxygen levels, etc. (Little et al. 1992; Vastra et al. 2016). As a consequence of these changes, the electrochemical behavior of the metal can be modified initiating and enhancing MIC.

In the initial stages of biofilm growth, the distribution of cells and EPS is likely to be heterogeneous, which may result in the areas of the metal substratum being covered by biofilm and adjacent area being exposed to the surrounding environment. The metabolic activity of cells within the biofilm will lead to low oxygen concentrations, and so form an oxygen concentration cell between those regions under the biofilm (anode), and the uncolonized areas exposed to maximal oxygen concentration (cathode) enhancing corrosion (Hamilton 2003). Castaneda and Benetton (2008) investigated that *SRB*-biofilm heterogeneities caused local gradient differences and increased the bio-catalytic active sites where corrosion processes take place, increased the corrosion rate. Moreover,



**Fig. 1** Stages of biofilm development (Chitra et al. 2014)

some studies have proposed that EPS isolated from biofilm have a nonnegligible effect on the corrosion process. Beech and Sunner (2004) proposed that metal ions bound by EPS accelerated the ionization of the metal by chelation, resulting in metal dissolution. Dong et al. (2011) found that excessive EPS stimulated anodic dissolution of carbon steel because  $\text{Fe}^{2+}$  from metal dissolution were bound by the polysaccharides in the EPS.

Central to the phenomenon of MIC is the formation of biofilm on the metal surface. The morphology of the biofilm has a significant influence on the corrosion effect, either in a detrimental or beneficial way. Usually, the formation of biofilm is known as the initial stage of MIC.

## 2.2 Anaerobic bacteria: *SRB*

*SRB* is a typical example of anaerobic MIC (Lee et al. 1995). Some researchers do insist that *SRB* are worthy of studying so much and regard their unique importance in MIC as “Myths of MIC” (Javaherdashti 1999). The corrosion mechanism of metal in the presence of *SRB* is a complicated biogeochemical process. Various mechanisms are proposed for the corrosion occurrence of *SRB*.

### (1) The cathodic depolarization theory

“Cathodic depolarization theory” (CDT) was the first proposed mechanism for MIC in 1934 by Von Wolzogen Kuhr and Van der Vlugt (1934). According to the classical theory, *SRB* consume cathodic hydrogen through consuming an enzyme called “hydrogenase” for the reduction of  $\text{SO}_4^{2-}$  as given below:

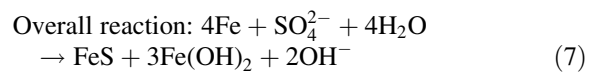
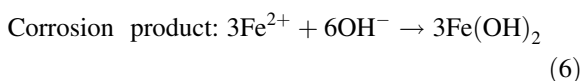
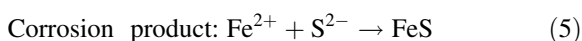
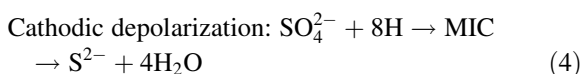
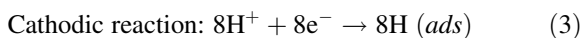
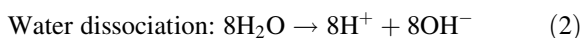


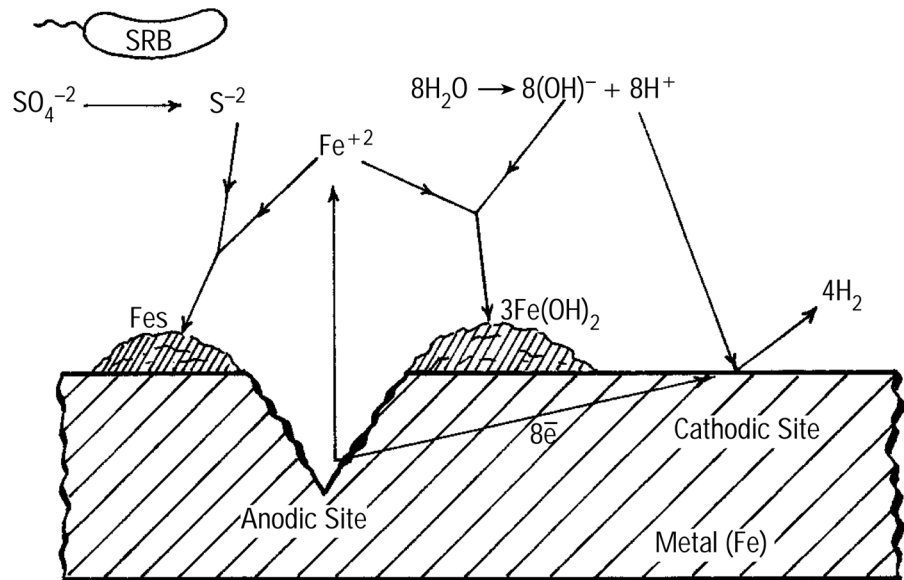
Figure 2 schematically summarizes the cathodic depolarization theory of *SRB* activity. The basic idea of the CDT was that the removal of hydrogen from the cathodic area on the iron surface by the hydrogenases of the bacteria, coupled to the reduction of  $\text{SO}_4^{2-}$ . The resulting corrosion products would thus be FeS and  $\text{Fe}(\text{OH})_2$ . *SRB*, by using the adsorbed hydrogen in the  $\text{SO}_4^{2-}$  reduction, catalyze the recombination of the adsorbed atomic hydrogen into hydrogen gas ( $\text{H}_2$ ), and increase the rate of cathodic reaction. Thus, the corrosion reaction would be indirectly accelerated by depolarization of the cathodic reaction.

Although CDT could explain MIC by *SRB* on the basis of electrochemistry, it suffers from much controversy in the subsequent decades (Tiller and Booth 1962). The idea that microbial  $\text{H}_2$  scavenging would accelerate corrosion, proposed by CDT, is very hard to accept. Costello (1974) demonstrated that  $\text{H}_2\text{S}$  from  $\text{SO}_4^{2-}$  reduction was a cathodic active compound; Hence, much of the electrochemical evidence for the “CDT” became disputable. On the basis of CDT, the ratio of corroded  $\text{Fe}^{2+}$  to FeS should be 4:1. However, in fact, the ratio varies from 0.9:1 to 1:1 (Javaherdashti 2011). In addition, the CDT was the lack of information on the role of sulfides that are themselves cathodic to iron and thus cause anodic dissolution (Xu et al. 2012). The complexity of biological environments involved with *SRB* activity makes it very difficult to assess any microbiological effect by means of electrochemical methods, because the physical and chemical parameters at the metal interface are continuously varying due to microbial metabolism. Drawbacks like these have prepared the ground for “new theories”.

### (2) MIC by microbes that secrete corrosive metabolites

*SRB* catalytic activity can promote the corrosion rate via the formation of aggressive corrosive metabolite  $\text{H}_2\text{S}$  (Abbas et al. 2013; Jia et al. 2018). This acid attack metals such as  $\text{Fe}^0$  extracellularly by removing electrons from  $\text{Fe}^0$  without bio-catalysis. Therefore, acid corrosion can occur without a biofilm in conventional chemical corrosion. In fact, Romero et al. (2005) reported that the bacteria in the *SRB* biofilm can reach  $10^8$  cfu/cm<sup>2</sup> to produce enough corrosive

**Fig. 2** Schematic of the cathodic depolarization theory of *SRB* activity (Javaherdashti 1999)



H<sub>2</sub>S. The introduction of H<sub>2</sub>S reduces the local pH in the biofilm due to H<sub>2</sub>S dissolution, thereby causing serious local corrosion. But in the whole process, the biofilm slows down the leak of H<sub>2</sub>S due to its diffusion barrier action.

FeS is considered the main corrosion product when steels are exposed to *SRB* environments (Fatah et al. 2013; Sheng et al. 2007). The physicochemical properties of the FeS film can affect steel dissolution, which depends on the ferrous ion/sulfide anions ratio, the presence of *SRB* and how the biofilm has covered the metal surface. However, in bacteria containing media, the sulfide films are usually unstable (Castaneda and Benetton 2008). They are disrupted by bacteria metabolic actions (Abbas et al. 2013). Therefore, the integrity of the protective film will be then degraded and leads to active corrosion cells between the FeS film (cathode) and the nearby metal substrate (anode) accelerating the corrosion rate significantly (Hamilton 2003).

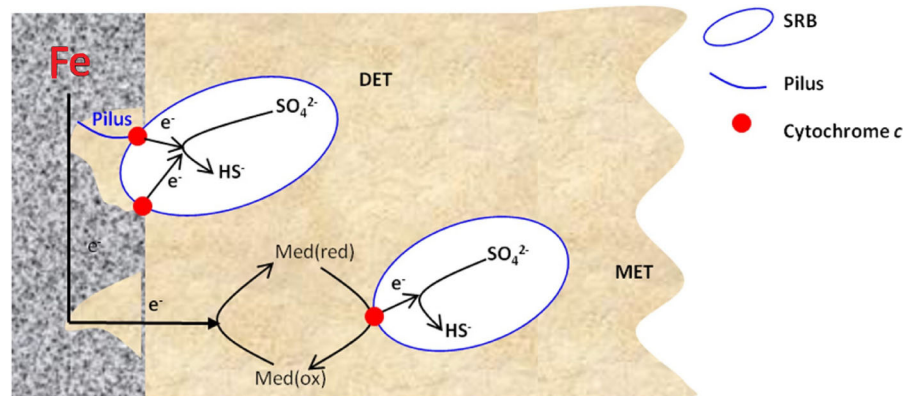
### (3) Extracellular electron transfer

Advances in microbiology, electrochemistry and surface analysis techniques have revealed the existence of biochemical pathway associated with MIC. “Extracellular electron transfer” (EET) theory offers better understanding for the study of metal–microbe interactions. Organic carbon molecules (e.g. lactate and acetate) can diffuse into the cytoplasm inside *SRB* cells to be oxidized. They donate electrons in the

oxidation process for SO<sub>4</sub><sup>2-</sup> reduction. Fe<sup>0</sup> in an iron (or steel) matrix is insoluble and thus Fe<sup>0</sup> cannot enter the cytoplasm. In MIC caused by *SRB*, Fe<sup>0</sup> oxidation occurs extracellularly while SO<sub>4</sub><sup>2-</sup> reduction occurs intracellularly with enzyme catalysis. This means the extracellular electrons released by Fe<sup>0</sup> oxidation must be transported across the cell wall into the *SRB* cytoplasm (Li et al. 2015). This kind of EET is achieved by electrogenic biofilm (Venzlaff et al. 2013; Yu et al. 2013). There are two primary categories for the mechanisms of EET. One is the direct electron transfer (DET) using cell-membrane bound redox proteins such as c-type cytochrome, conductive nanowires (pili) (Mehanna et al. 2009; Reguera et al. 2005; Sherar et al. 2011) and the other mediated electron transfer (MET) using electron carriers such as hydrogen or electron transfer mediators such as riboflavin and flavin adenine dinucleotide (FAD) (Jia et al. 2017a, b; Zhang et al. 2015). Figure 3 shows a schematic diagram for the mechanism of MIC by *SRB* due to cross-cell wall electron transfer involving DET and MET (Li et al. 2015). Based on EET mechanism for MIC, Xu et al. (Xu and Gu 2014) showed that starved *SRB* biofilm is more corrosive against carbon steel because they utilize extracellular electrons released by Fe<sup>0</sup> oxidation as an electron donor when there is a lack of organic carbon in the culture medium. Zhang et al. (2015) investigated that electron transfer is a bottleneck in this type of MIC by *SRB* because



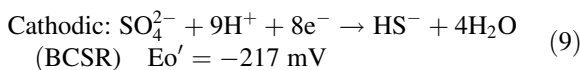
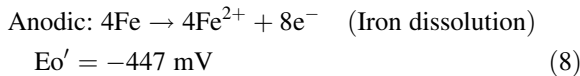
**Fig. 3** Schematic illustration for the mechanism of MIC by *SRB* due to cross-cell wall electron transfer involving direct electron transfer (DET) and mediated electron transfer (MET) (Li et al. 2015)



adding an electron mediator to the culture medium to enhance electron transfer accelerated corrosion.

#### (4) Biocatalytic cathodic sulfate reduction theory

Gu et al. (2009) proposed a new MIC theory called “biocatalytic cathodic sulfate reduction theory” (BCSR) based on bioenergetics. When there is a lack of food source (e.g. lactate) due to diffusional limitation, the sessile cells under an *SRB* biofilm will switch to elemental iron as an alternate electron donor for the reduction of  $\text{SO}_4^{2-}$  to harvest energy for metabolic activities. The following equations can be used to explain *SRB* corrosion in BCSR (Li et al. 2015):

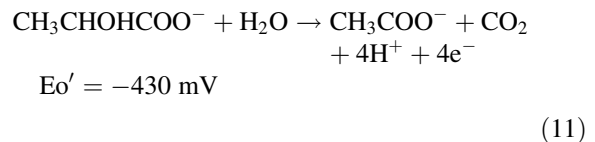


The cell potential for the redox reaction coupling Reactions (8) and (9) is  $\Delta E_o' = +230 \text{ mV}$  versus the standard hydrogen electrode (SHE) [calculated from  $-217 \text{ mV}$  to  $(-447 \text{ mV})$ ] under the conditions of  $25^\circ\text{C}$ , pH 7, 1 M concentration for solutes and 1 bar partial pressure for gases. This positive cell potential corresponds to a Gibbs free energy change of reaction  $\Delta G_o' = -178 \text{ kJ/mol SO}_4^{2-}$  based on the following equation, in which  $n$  is the number of electrons for  $\text{SO}_4^{2-}$  reduction ( $n = 8$ ) and  $F$  is the Faraday constant ( $F = 96,485 \text{ C/mol}$ ):

$$\Delta G_o' = -nF\Delta E_o' \quad (10)$$

The negative  $\Delta G_o'$  suggests an exergonic reaction that it is highly favorable thermodynamically and that corrosion of steels can occur spontaneously.

When lactate is used as the organic carbon source, the following oxidation reaction occurs in the cytoplasm (Xu and Gu 2014):



In fact,  $\text{Fe}^0$  is slightly more energetic than lactate because the  $\Delta E_o'$  value for  $\text{Fe}^{2+}/\text{Fe}^0$  ( $-447 \text{ mV}$ ) is slightly more negative than that for  $\text{CO}_2 + \text{acetate}/\text{lactate}$  ( $-430 \text{ mV}$ ). The equilibrium potentials above are used to calculate the thermodynamic driving force of MIC. However, favorable driving force does not mean that corrosion will occur if kinetics is retarded.  $\text{SO}_4^{2-}$  reduction has a high activation energy. It will proceed in the *SRB* cytoplasm via the adenosine phosphosulfate (APS) pathway catalyzed by multiple enzymes. Thus, Gu et al. (2009) proposed the BCSR theory by treating  $\text{Fe}^0$  oxidation as the anodic reaction and biocatalytic  $\text{SO}_4^{2-}$  reduction as the cathodic reaction. The word “cathodic” is used in BCSR merely to suggest that it is a reduction reaction in the corrosion mechanism, thus providing energy for *SRB* metabolic activities such as active transport, signal amplification and organic synthesis (Xu and Gu 2014). There is actually no physical cathode at the site where  $\text{SO}_4^{2-}$  reduction occurs because it happens in the *SRB* cytoplasm.

Put simply, there is not a single mechanism to explain all of the *SRB*-related corrosion on different

materials. The mechanisms are quite complex involving several bio- and physico-chemical parameters. Apart from the chemistry changing of the metal–solution interface and/or the metabolites in corrosion, the investigation of electrogenic biofilm of bacteria is still very useful in MIC. The new trends in research should focus on bio-electrochemical phenomena at the metal surface.

### 2.3 Aerobic bacteria: *IOB*

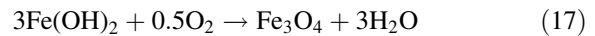
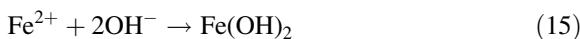
*IOB* are bacteria that can harness energy from the oxidation of Fe(II) to Fe(III) for growth, which are ubiquitous in various environments such as marine environment (Moradi et al. 2011) and oil field (Liu et al. 2016b). Fe(II) can function as an electron donor and O<sub>2</sub> can function as a terminal electron acceptor under aerobic conditions in the presence of *IOB* (Emerson and Moyer 1997). Under biocatalysis by *IOB*, the oxidation rate of Fe(II) to Fe(III) seem to be much faster than that of abiotic chemical oxidation. Thus, *IOB* accelerate the dissolution of metal and the development of localized corrosion (Liu et al. 2016a). Compared to potential inorganic energy sources for *IOB*, the oxidation of Fe(II) to Fe(III) yields the lowest Gibbs free energy ( $\Delta G^0 = -29 \text{ kJ mol}^{-1}$ ,  $\text{Fe}^{2+} + 0.25\text{O}_2 + \text{H}^+ \rightarrow \text{Fe}^{3+} + 0.5\text{H}_2\text{O}$ ) for cellular metabolism. Furthermore, if the Fe(III) is precipitated as an iron oxide-hydroxide [ $\text{Fe}^{2+} + 0.25\text{O}_2 + 2.5\text{H}_2\text{O} \rightarrow \text{Fe}(\text{OH})_3 + 2\text{H}^+$ ], the energetic yield is estimated to be double (Emerson et al. 2010).

*IOB* can promote oxidation of Fe(II) and induce different types of steel corrosion, especially localized corrosion (Moradi et al. 2011; Starosvetsky et al. 2008). The main corrosion products induced by *IOB* are iron oxides such as FeOOH, Fe<sub>2</sub>O<sub>3</sub> and Fe<sub>3</sub>O<sub>4</sub>, the main reaction process is shown in reaction (12)–(18) (Jin et al. 2015):

Anodic reaction:

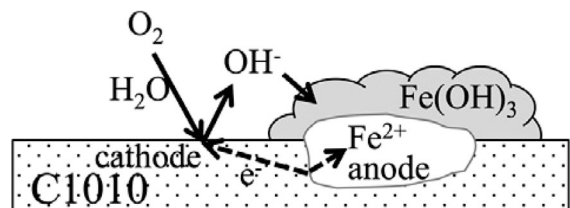


Cathodic reaction:



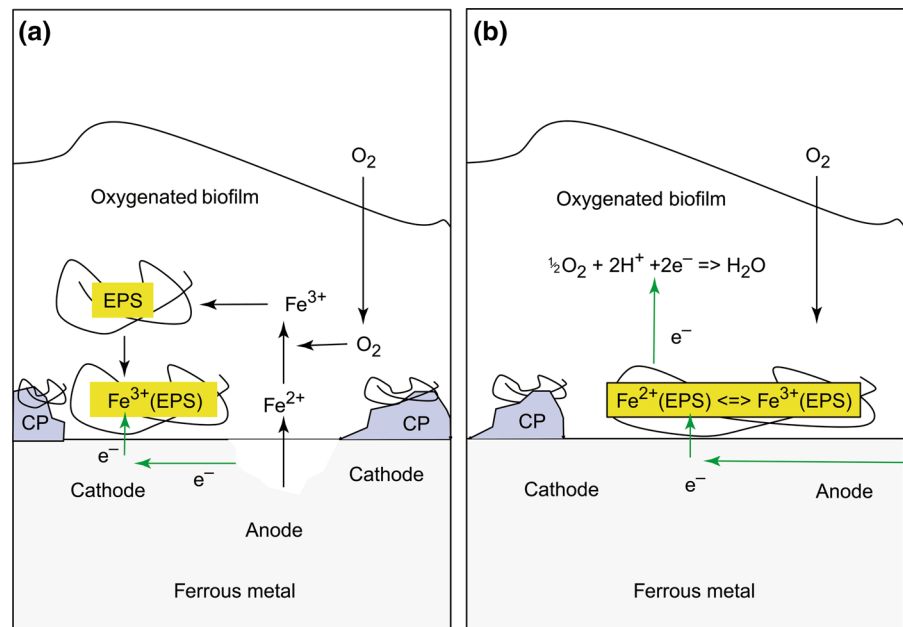
Wang et al. (Wang et al. 2014) have put forward “crevice corrosion induced by iron oxide” for aerobic *IOB*-induced MIC mechanisms. Figure 4 is the mechanism schematic of the crevice corrosion initiated by the Fe(OH)<sub>3</sub> layer under aerobic conditions. Under aerobic conditions, the carbon steel surface beneath the iron oxide layer could form small anodic sites to transfer electrons from Fe<sup>0</sup> to oxygen at cathodic sites to form hydroxyl, which could continuously oxidize Fe<sup>0</sup> to Fe<sup>2+</sup> as energy source for cells and form iron oxide precipitation and lower the local pH. At the anodic site, Fe<sup>0</sup> oxidation continued, ultimately forming a pit on the carbon steel surface.

The corrosion process induced by *IOB* is also owing to the presence of biofilm formed on surfaces of metallic materials. The development of a biofilm is facilitated by the production of EPS which play a key role in MIC. In addition to promoting the growth of sessile bacteria, EPS can also to bind metal ions considerably modifying the redox reaction pathways in the biofilm/metal interface (as shown in Fig. 5a), such as direct electron transfer from a metal surface (Beech and Sunner 2004). Some electroactive EPS-bound metal ions in a biofilm can act as electron “shuttles” to transport electrons to the terminal electron acceptor O<sub>2</sub> (as shown in Fig. 5b), which accelerates corrosion. Nevertheless, EPS at later incubation stages can act as an oxygen barrier and thereby inhibit corrosion (Jin and Guan 2014). Liu et al. (2017) studied the effect of EPS on the corrosion behavior and mechanism of carbon steel, and found that 240 mg L<sup>-1</sup> of 7 day-old EPS secreted by *IOB* could inhibit corrosion due to the formation of a compact adsorbed EPS film.



**Fig. 4** Schematic of the crevice corrosion initiated by the Fe(OH)<sub>3</sub> layer under aerobic conditions (Wang et al. 2014)

**Fig. 5** Schematic of corrosion reactions involving EPS-bound metal ions in oxygenated biofilm: **a**  $\text{Fe}^{3+}$  is bound by EPS and the  $\text{Fe}^{3+}$ -EPS complex is deposited on the metal surface, **b** In the presence of oxygen, acting as terminal electron acceptor,  $\text{Fe}^{2+}$  in EPS is reoxidised to  $\text{Fe}^{3+}$  (Beech and Sunner 2004)



The corrosion mechanism of *IOB* is much less comprehensively understood than the corrosion mechanism of *SRB*. It remains to be further strengthened, using the advanced molecular biology technology and microelectrode technology, revealed from the microcosmic *IOB* corrosion process so as to provide theoretical support for better corrosion prevention.

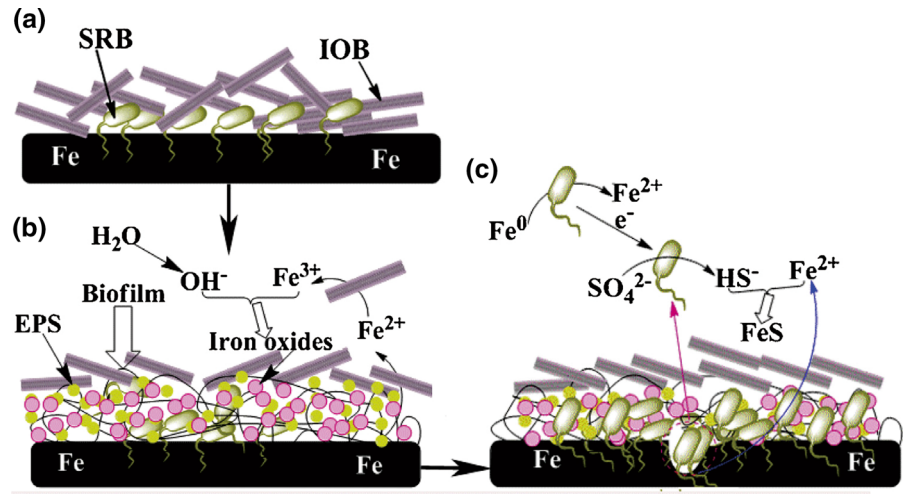
#### 2.4 MIC in the mixed culture of *SRB* and *IOB*

Bacteria are major colonizers on a substrate surface in almost all environments (An et al. 2010; Ilhan-Sungur and Çotuk 2010; Quan et al. 2010). Thus, the majority of MIC studies have investigated the effects of pure or mixed culture bacteria on corrosion behavior of a variety of metals and alloys such as iron, copper, aluminum (Agwa et al. 2017; Guan et al. 2017; Huttunen-Saarivirta et al. 2017). The main categories of bacteria typically linked to corrosion in terrestrial and aquatic habitats are *SRB* (Jia et al. 2018), *Sulfur oxidizing bacteria* (Xin et al. 2009), *IOB* (Emerson and Moyer 1997), *Iron reducing bacteria* (Herrera and Videla 2009), *Nitrate reducing bacteria* (Jia et al. 2017a, b), and *Acid producing bacteria* (Xu et al. 2016). These bacteria can coexist and adhere in biofilm, forming complex consortia on corroding metallic surfaces (Beech and Sunner 2004). For example, anaerobic *SRB* and aerobic *IOB* are

sometimes found together in the biofilm (Liu et al. 2015). In an aerobic environment, *IOB* oxidize ferrous ions ( $\text{Fe}^{2+}$ ) to ferric ions ( $\text{Fe}^{3+}$ ), where the electrons released are used as energy sources for growth. Subsequently, these ferric ions ( $\text{Fe}^{3+}$ ) precipitates as iron hydroxide complexes, which can cause a blockage by the formation of tubercles in the pipeline. With the mixed culture system, anaerobic *SRB* may propagate rapidly and attack metals in the locally anaerobic conditions beneath the iron-rich tubercles in the presence of *IOB*. The interaction between *SRB* and *IOB* accelerated the corrosion process of metal specimens (Xu et al. 2007). Xu et al. (2008) investigated the localized corrosion behavior of 316L stainless steel in the presence of anaerobic *SRB* and aerobic *IOB*. They found that the synergies between the metal surface, abiotic corrosion products, bacterial cells and their metabolic products increased the corrosion rate and accelerated pitting propagation. Rao et al. (2000) suggested that aerobic iron bacteria and *SRB* induced corrosion of carbon steel in the cooling water system, and beneath the tubercles, *SRB* induced significant pitting corrosion. Eun-Hae Sung et al. (2011) assessed the effects of microbial growth on metal corrosion. With the mixed culture system, the number of *SRB* in the presence of *IOB*, was 66 times higher than that of the sole *SRB* culture, which was attributed to the potential synergistic effects. It



**Fig. 6** The formation propagation mechanism of pitting corrosion in the mixture of *SRB* and *IOB*: **a** initial period, **b** formation of biofilm, and **c** formation and propagation of pitting corrosion (Liu et al. 2015)



increased the degrees of corrosion by factors of 2–7 compared with that of the single *IOB* and *SRB* cultures. Meanwhile, in the mixed culture, black sulfide formed by *SRB* was observed beneath the tubercles formed by *IOB*. *SRB* can also exist in aerobic conditions (Beech and Sunner 2004). Liu et al. (2015) investigated corrosion behavior of carbon steel in the presence of *SRB* and *IOB* with 4.2 mg/L dissolved  $O_2$ . The results showed that the synergistic effect of *SRB* and *IOB* played a key role for the pitting corrosion of carbon steel. They illustrated the pitting corrosion schematic of carbon steel in the presence of *SRB* and *IOB* (Fig. 6a). In the first step, planktonic cells attach on the coupon surface (Fig. 6a). Then, a large amount of metabolites were produced, accompanying with the formation of biofilm. In the third step, a steady-state biofilm layer was formed with a local anaerobic environment which will promote the growth of *SRB* (Fig. 6c). On the one hand, in the presence of *SRB* and *IOB*, *IOB* could oxidize  $Fe^{2+}$  to  $Fe^{3+}$ , and  $S^{2-}$  produced by *SRB* could react with  $Fe^{2+}$ , which would promote the formation of pitting corrosion. On the other hand, *SRB* switched to elemental Fe to obtain sulfate-reduced electrons due to local shortage of organic carbon underneath biofilm.

The importance of microbial synergy has recently been reconfirmed in studies of steel corrosion. The corrosion rate obtained with the combination of *SRB* and *IOB* is considerably higher than those measured in pure cultures. The research on synergistic corrosion caused by anaerobic and aerobic bacteria can help to

reveal the MIC mechanism under operating conditions and provide theoretical support for MIC control.

### 3 The influence of cathodic polarization on MIC

Cathodic Polarization (CP) has been recognized for a long time as a reliable, effective and economic method to prevent corrosion of pipelines, marine structures, tanks (Gurrappa 2005). It is a proven technique which is able to arrest ongoing corrosion and induce and sustain steel passivity (Christodoulou et al. 2010). According to the National Association of Corrosion Engineers (NACE) standard RP-0169, the criteria of CP is applying an impressed current or potential to induce negative steel polarization, in other words to drive the steel potentials more cathodically than  $-770$  mV versus saturated calomel electrode (SCE), where corrosion process is thermodynamically impossible to occur (Pourbaix 1996). Under these conditions the steel will be immune to corrosion. The influence of CP on bacterial activity is important in modifying the economics of protection and the occurrence of corrosion. Edyvean et al. (1992) found that, whether for stainless steel or carbon steel, CP can inhibit the settlement and attachment of aerobic microorganisms to the surface of the electrode. Researchers have controlled the current or potential of metal surface to prevent biofilm growth. Shirliff et al. (2005) provided a constant flux of  $370 \mu A/cm^2$  on a stainless-steel electrode surface. The results suggested that current application was unable to reduce

biofilm and even increased the number of biofilm bacteria. Similarly, Borden et al. (2004) applied a current between 2.86 and 4.76  $\mu\text{A}/\text{cm}^2$  to induce bacterial detachment of growing *Staphylococcus epidermidis* biofilm from surgical stainless steel. Other researchers have controlled the potential of electrode surfaces to prevent cell growth. For example, Edyvean et al. (1992) poised a stainless steel at negative potentials and observed a decrease in bacterial settlement and attachment. Little and Wagner's work (Little et al. 1997) also investigated the interaction of CP and the biofilm formation on steels. They confirmed that, when the CP current was intermittent, the corrosion attack due to the microorganisms became more aggressive.

The understanding of the mutual interaction of CP and MIC is vague. Some of the inhibitory mechanisms of CP on MIC has been interpreted in terms of (1) electrophoretic force and electrostatic repulsive force (Hong et al. 2008), (2) The effect of oxygen reduction reaction (Istanbullu et al. 2012; Pérez et al. 2009) and (3) the excessive electrons supplied by CP power source used as the electron donor by bacteria (Liu and Cheng 2017). However, none of these mechanisms is widely accepted because of conflicting results in the literature (Edyvean et al. 1992; Little et al. 1997) and the impossibility of comparing the different electrochemical systems.

### 3.1 Electrostatic repulsive force and electrophoretic force

On an electrically conductive surface over which current or potential is applied, adhesion of the bacterial cells may be influenced by electrostatic force and electrophoretic force. Wilson et al. (2001) stated that, bacterial surfaces appear to be negatively charged by virtue of ionized phosphoryl and carboxylate substituents on outer cell envelope surfaces under normal conditions. A cathodic polarized surface is a negatively charged region and would therefore repel approaching negatively charged bacterial cell during the process of attachment (Heckels et al. 1976).

Under these scenarios, there are overwhelming studies on the mutual effect of CP and microbial activity. de Saravia et al. (1997) studied the influence of mixed anaerobic bacterial biofilm on cathodically protected 304L stainless steel. The results showed that there was a decrease in the number of attached cells

when CP was applied in the initial stages of biofilm formation, which may be influenced by electrostatic forces. And this decrease was greater at more negative potentials. But, CP did not have an effect when a stationary biofilm was formed. Busscher and Weerkamp (1987) has reported that the adhesion of bacteria to the solid surface would be influenced by electrostatic repulsion. Busalmen and de Sanchez (2001) have contrasted the adhesion of *Pseudomonas fluorescens* to nonpolarized and negatively polarized thin films of gold. When imposed a strong negative charge to the metal surface ( $-0.5$  and  $-0.2$  V vs. Ag/AgCl), bacterial adhesion was strongly inhibited. There was a repulsive electrostatic interaction with the negatively charged bacterial cells that depended on the electrical potentials of the surfaces. Hong et al. (2008) clarified the specific role of electric currents (cathodic and anodic) in order to develop the optimal strategy for controlling bacterial adhesion. They applied a 15  $\mu\text{A}/\text{cm}^2$  cathodic current and found that the applied current promoted 80% detachment of bacteria from the electrode surface, 20% of bacteria still remained on the electrode surface despite the presence of electrostatic and electrophoretic repulsive forces. In contrast, when an anodic current was applied, the bacteria that remained on the surface became inactive with time, although bacterial detachment was not significant. Poortinga et al. (2001) found that the electrophoretic force and electrostatic repulsion resulting from a impressed cathode current stimulated desorption of adhering bacteria.

### 3.2 The effect of oxygen reduction reaction

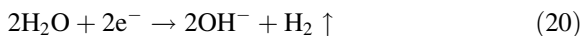
When applying a CP to a metallic structure, CP increases the hydroxyl ion ( $\text{OH}^-$ ) concentration and the partial reduction of oxygen ( $\text{O}_2$ ) leads to hydrogen peroxide ( $\text{H}_2\text{O}_2$ ) production. At the same time, the consequent pH increase diminishes the solubility of calcium magnesium at the metal surface favoring the precipitation of a calcareous scale. There is evidence that the electrochemical changes CP produces on the surrounding environment influence the settlement of fouling organisms.

#### (1) Increased interfacial pH values

Under CP, the electrochemical reactions that occur on the surface of metallic structures are the reduction of  $\text{O}_2$ .



and the reduction of water leading to hydrogen evolution for more negative cathodic potentials.

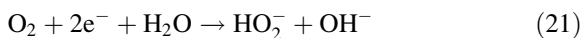


Recent results have demonstrated that CP inhibited microbiological growth and attachment because of the increased pH values generated in the vicinity of the protected steel surface. Pérez et al. (2009) investigated the influence of cathodic currents on biofouling processes in metal/anticorrosive paint/seawater systems. The results indicated that an increased interfacial pH as a consequence of the cathodic currents cause a marked decline of biofouling attachment. Del Pozo et al. (2009) afforded a potential means to significantly reduce the number of viable bacteria by prolonged exposure of biofilm to electrical current alone, and to call this phenomenon as the “electricidal effect”. The mechanism of the “electricidal effect” could relate to the pH changes.

That is to say, biofilm are able to modify metal–solution interface by drastically changing the physical, chemical and hydrodynamic conditions (de Saravia et al. 1997). But, owing to the buffering capacity of seawater and the microbial activity within the biofilm, the interfacial pH value was lower than the expected theoretical value.

### (2) The production of $\text{H}_2\text{O}_2$

According to Eq. (19), oxygen is reduced to  $\text{OH}^-$  through a four-electron pathway on metal surfaces. However, at the same time, the partial reduction of oxygen leads to  $\text{H}_2\text{O}_2$  production through a two-electron pathway (Babić and Metikoš-Huković 1993).

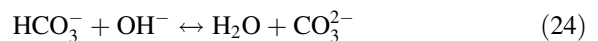
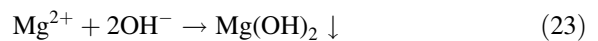


The mechanism of the antibacterial activity of electrical current has been suggested to result from the generation of  $\text{H}_2\text{O}_2$ . Istanbullu et al. (2012) found that  $\text{H}_2\text{O}_2$  was produced near 316L SS surfaces when a negative potential was applied.  $\text{H}_2\text{O}_2$  produced on a SS surface was able to prevent biofilm growth. Dhar et al. (1982) applied an electrochemical study on the prevention of bacterial attachment and found that the applied biofilm growth was inhibited. They thought that this was due to the production of  $\text{H}_2\text{O}_2$ . Del Pozo

et al. (2009) applied a current between  $1.626 \times 10^{-3}$  and  $1.626 \times 10^{-6} \mu\text{A cm}^{-2}$  on electrode surface to prevent planktonic bacterial attachment and did not detect  $\text{H}_2\text{O}_2$ . This is most likely because  $\text{H}_2\text{O}_2$  was produced at a low concentration near the electrode surface and could not be detected in the bulk solution.

### (3) The formation of calcareous deposits

Both reactions (19) and (20) can generate  $\text{OH}^-$  ions near the surface of metallic structures resulting in the precipitation of calcium carbonates ( $\text{CaCO}_3$ ) and magnesium hydroxides ( $\text{Mg}(\text{OH})_2$ ) on the cathodically protected surfaces according to the following reactions (Sun et al. 2012):

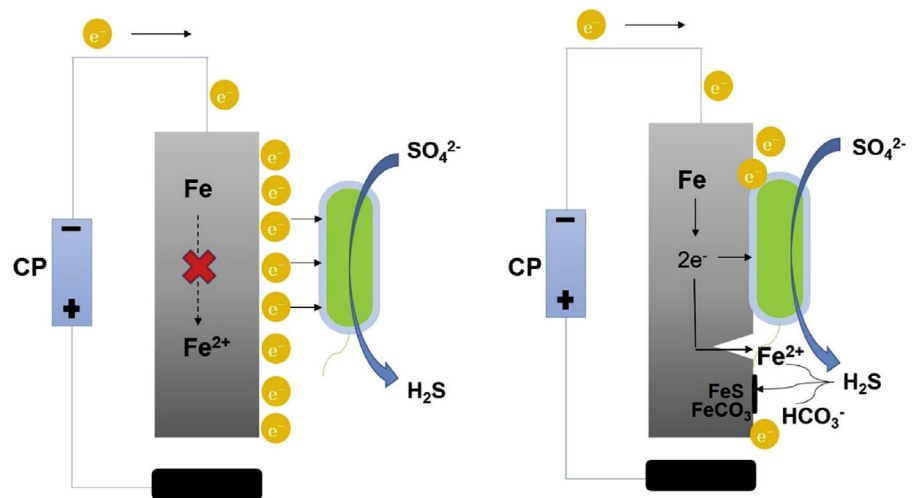


The importance of calcareous deposits to the effectiveness and efficiency of cathodic protection in submerged ocean situations is generally recognized (Guezennec 1994). These calcareous deposits can act as a physical barrier to general corrosion attack, impede solute diffusion (e.g. oxygen), reduce the current density required to maintain a given potential, and consequently decrease the cost of cathodic protection. It is widely believed that bacterial cells are able to attach to negatively charged surfaces by using divalent cations such as  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  as a “bridge”. The precipitation of calcium and magnesium ions results in a decrease in their local concentration and thus they would be unavailable to the bacteria (Edyvean et al. 1992). Eashwar et al. (2009) stated that calcareous deposits developed quite rapidly on polarized SS cathodes and lead to a significant reduction of bacterial numbers.

### 3.3 The excessive electrons supplied by CP

However, some findings were opposite to those listed above. Some studies have found that hydrogen produced by the cathode can encourage growth of hydrogenase-containing bacteria such as *SRB* (Guezennec 1994). Nekoksa and Gutherman’s work (1991) showed that the CP application enhanced the incubation and growth of microbes, with more bacteria settling on a cathodic protected stainless steel than on

**Fig. 7** Schematic diagrams illustrating **a** *SRB* use electrons supplied by CP power source, and **b** sessile *SRB* use electrons from the steel directly (Liu and Cheng 2017)



unpolarized ones. Similar results were obtained by Olivares et al. (2003), who found that the *SRB* population on CP-applied ( $-850$  mV vs. Cu/CuSO<sub>4</sub>) pipeline steel was twice of the microorganisms on the coupons without CP application. In the latest report, Liu et al. (2017) found that the CP facilitated the bacterial attachment to the steel, and the number of sessile cells on the steel increases as the CP potential becomes more negative. They proposed that if the CP potential is sufficiently negative, the *SRB* use the electrons from the CP source. The excessive electrons supplied by the CP power source that accumulated on the steel surface are used as the electron donor by *SRB* in their metabolism. Fe<sup>0</sup> would not act as the electron donor, as schematically shown in Fig. 7a. However, when the potential is not sufficient locally due to potential fluctuations caused by the shielding effect of the biofilm, *SRB* would use electrons from the steel directly initiating pitting corrosion (Fig. 7b).

Some of reported results of relevant literature on this topic are controversial to each other. Undoubtedly, not enough is known about the relationship between cathodic polarization and microbial attachment. CP has been recognized for a long time as an effective way to prevent MIC. Chen et al. (2015) have reported that the pH in *SRB*-containing medium was higher than that in the *SRB*-free medium. And owing to the formed biofilm of bacteria, the cathodic protection potential for carbon steel is more negative under biotic conditions. In *SRB*-containing aqueous solutions, Esquivel et al. (2015) suggested that the applied CP potential should be shifted to  $-950$  mV

versus Cu/CuSO<sub>4</sub> electrode or even more negative values in order to achieve good control on the MIC. But, the negative CP potential would stimulate the cathodic hydrogen generation which would promote the stress corrosion cracking (SCC) susceptibility of the steel (Wu et al. 2015). Therefore, the investigation of appropriate CP potential and the mutual effect between CP and microbial attachment are stronger required.

#### 4 Future perspectives

MIC of steels is a great issue in many industries such as gas/oil pipelines, storage tanks, residual water treatment systems, steel pilings in marine applications, and, more recently, offshore wind farms. MIC is rarely linked to a single mechanism or to a single species of microorganisms. It results from the synergistic effect between microbes, metals, metabolites, and their environment. Despite considerable research efforts into the MIC phenomenon, there is still ongoing dispute with respect to which component, biotic or abiotic, is of greater relevance to MIC process. Advances in microbiology, electrochemistry and surface analysis techniques allow for better understanding the dynamic processes of biological and abiotic reactions in space and time and how these, in turn, affect mechanisms and accelerate rates of electrochemical reactions leading to corrosion. The author believes that a better understanding of the corrosion caused by communities of bacteria is envisioned to

ultimately aid in the design of better MIC prevention and mitigation strategies for a variety of steel constructions. Conventional ways to combat MIC are well-known physical and chemical methods such as pigging, applying antimicrobial agents and disinfectants, but, especially from an environmental point of view, it is necessary to find new ways to control microbial deleterious effects through environmentally friendly approaches.

## 5 Conclusion

MIC of steels caused by aerobic and anaerobic bacteria in several industrial environments are discussed: (1) generally, biofilm formation is the initial stage of MIC, and biofilm can also influence the physical and chemical parameters at metal–solution interface, modifying the electrochemical behavior of the metal in a detrimental or beneficial way. (2) Mechanisms of anaerobic SRB and aerobic IOB in MIC are analyzed. Owing to the complexity of the microbe-metal interaction, the underlying mechanism of MIC allows for multi-mechanisms explanation for corrosion damage of steels. (3) The synergistically aggressive roles between SRB and IOB are responsible for severe pitting corrosion of metal specimens. (4) CP has been recognized as an effective way to prevent MIC. When CP is applied, microbial adhesion is affected by the imposed current over relatively extended periods of time. However, there is still confusion and limitations about the interaction between microbial activity and the process of preventing corrosion by CP in published literature. To investigate MIC, a clear understanding of its mechanisms is needed to guide field operators on how to mitigate and inhibit MIC.

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### Compliance with ethical standards

**Conflict of interest** The authors declare that they have no conflict of interest.

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