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Anaerobic microbiologically influenced corrosion mechanisms interpreted using bioenergetics and bioelectrochemistry: A review

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ABSTRACT

Microbiologically influenced corrosion (MIC) is a major cause of corrosion damages, facility failures, and financial losses, making MIC an important research topic. Due to complex microbiological activities and a lack of deep understanding of the interactions between biofilms and metal surfaces, MIC occurrences and mechanisms are difficult to predict and interpret. Many theories and mechanisms have been proposed to explain MIC. In this review, the mechanisms of MIC are discussed using bioenergetics, microbial respiration types, and biofilm extracellular electron transfer (EET). Two main MIC types, namely EET-MIC and metabolite MIC (M-MIC), are discussed. This brief review provides a state of the art insight into MIC mechanisms and it helps the diagnosis and prediction of occurrences of MIC under anaerobic conditions in the oil and gas industry.

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1. Introduction

Corrosion causes an annual financial loss of US\$4 trillion globally, half of which is due to corrosion damages while the other half are costs for corrosion protection measures [1,2]. Microbes are ubiquitous in industrial systems. Microbiologically influenced corrosion (MIC), i.e. corrosion resulting from the activities of microorganisms, is a challenging problem in the oil and gas industry and is said to be behind of more than 20% of pipeline corrosion [3–5]. Pipeline leakages and facility failures due to MIC have

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been common occurrences with environmental damages over the past two decades [6–8]. Research activities on biocides, biocide enhancers, antibacterial stainless steels, and antibacterial coatings have been conducted to mitigate MIC [9–12]. MIC problems are more prevalent nowadays due to enhanced oil recovery that relies on seawater injection, which can lead to MIC and reservoir souring [13,14]. Furthermore, the increased awareness of MIC leads to more MIC problems identified.

Microbial contamination also causes biofouling and corrosion to equipment and infrastructure in other industrial settings including underground storage tanks [15], water distribution systems [16], rail systems [17], cooling water systems [18,19], ships [20], medical devices [21], and nuclear waste storage facilities [22]. MIC plays a considerable role in the corrosion and degradation of different materials, including but not limited to carbon steel [23], aluminum alloy [22], stainless steel (SS) [24], duplex SS (DSS) [11,25–28], hyper duplex SS (HDSS) [29], super austenitic SS (SASS) [30], high nitrogen SS (HNS) [31], magnesium [32], and even concrete [33]. Furthermore, MIC is not merely a type of corrosion by itself. It synergistically interacts with other corrosion processes, for instance, stress corrosion cracking (SCC) [34], crevice corrosion [35], fatigue-crack tip embrittlement [36], and under-deposit corrosion [37]. MIC is a persistent issue for corrosion engineers and scientists in various fields.

Many microorganisms are capable of MIC including bacteria, archaea, and fungi. Sulfate-reducing bacteria (SRB) and sulfate-reducing archaea (SRA) have been extensively investigated as the major causative microorganisms in MIC for decades because sulfate is widely distributed in many systems such as seawater, brackish water and agricultural runoff water [9,38]. SRB and SRA are isolated frequently from oil and gas fields with MIC problems [4,39]. A positive correlation among pitting corrosion, sulfide generation, and sulfate consumption is often observed in the presence of SRB [40]. In addition, nitrate-reducing bacteria (NRB) [41], methanogens [40,42], iron-oxidizing bacteria (IOB) [43], manganese-oxidizing bacteria (MOB) [44], and diverse fungal species [45] have also been linked to MIC.

In the natural environment, microorganisms often exist in a community called a biofilm, which is central to the occurrences of biofouling, biodegradation, and biocorrosion. A biofilm consists of extracellular polymeric substances (EPS) and embedded sessile cells. EPS play an important role in biofilm formation, maturation and also maintenance [46,47]. EPS compounds include polysaccharides, proteins, lipids, and nucleic acids, which sometimes form a gel-like slime [48].

Under field conditions, mixed-culture biofilms with a rich microbial diversity are difficult to mitigate [9,49]. Synergistic interactions of different microorganisms in biofilm consortia can cause severe MIC because of nutrient and energy sharing. The symbiotic proliferation of SRB and sulfide-oxidizing bacteria (SOB) was hypothesized to explain severe MIC on 2205 DSS [50]. Fig. 1 shows an example of C1018 carbon steel pitting corrosion by an oilfield biofilm consortium in 7 days in a lab test [49].

It is inaccurate to believe that all biofilms degrade the integrity of metals. *Pseudomonas aeruginosa* biofilms increased the corrosion rate of a nickel–copper alloy but protected a nickel–zinc alloy [51]. *Escherichia coli* and *Geobacter sulfurreducens* biofilms formed a protective barrier on a SS surface enhancing its corrosion resistance [52,53]. Aerobic biofilms can serve as an oxygen barrier slowing down oxygen permeation and thus possibly retarding the corrosion. Biofilms at each stage affect a corrosion process differently. A biofilm sample isolated from a drinking water system accelerated corrosion during the initial 7-day incubation but it offered protection to the pipe after incubating for 30 days [54]. Therefore, a clear link should be established between the microbial community and its influence on the corrosion behavior of different materials.

Table 1
Redox potential^a (E°).

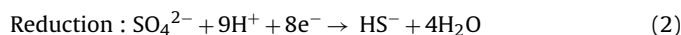
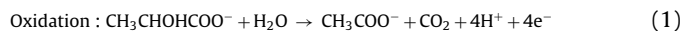
Redox couple	E° (mV)
Fe ²⁺ /Fe ⁰	–447
CO ₂ + acetate/lactate	–430
CO ₂ /formate	–432
CO ₂ /methanol	–370
SO ₄ ^{2–} /HS [–]	–217
CO ₂ /CH ₄	–244
NO ₂ [–] /NH ₃	+330
NO ₃ [–] /NH ₃	+360
2NO ₃ [–] /N ₂	+760

^a Data from [41,65]. “o” in bioelectrochemistry indicates the condition with 1 M solutes (1 bar gases) except for H⁺, 25 °C and pH 7.

Various mechanisms have been proposed to explain MIC mechanisms. In the classical cathodic depolarization theory (CDT), SRB cells use hydrogenase enzymes to lower the activation energy of the H atom desorption process [55]. This is regarded as the rate-limiting step for MIC by SRB. The CDT can only explain MIC caused by hydrogenase-positive SRB, but it is not applicable to SRB lacking the enzyme. Many MIC researchers embraced this early theory [56–59]. However, the understanding of MIC is hampered by the absence of a clear explanation of the bioelectrochemical processes occurring at the interface between biofilm and metal matrix. To make a step ahead, the mechanism of metal MIC is elucidated from a multidisciplinary perspective in this review. The reasons regarding why and how MIC occurs are explained based on bioenergetics and extracellular electron transfer (EET). Thus, this review provides useful information for corrosion management and risk assessment.

2. Bioenergetics of MIC

Microorganisms require the following principal components to provide energy for their metabolism: an electron donor (energy source) and an electron acceptor [60]. Hydrocarbons and fatty acids (e.g., formate, pyruvate, acetate, methanol and lactate) usually serve as organic carbons for SRB growth [61]. They provide energy and carbon for growth. For hydrogenase-positive SRB, hydrogen gas is an alternative electron donor. SRB usually use sulfate as the terminal electron acceptor, reducing it finally to HS[–] [62,63]. In addition to sulfate, some SRB can also use sulfite, thiosulfate, and elemental sulfur. In addition, some SRB strains may switch to nitrate or nitrite as the terminal electron acceptor [64,65]. The oxidation and reduction reactions for SRB metabolism in the presence of carbon source (using lactate as an example) and sulfate are listed below [65].



Using the redox potentials shown in Table 1, the reaction combining the two reactions above yields a change in the Gibbs free energy (ΔG°) of –164 kJ/mol of sulfate under standard condition. Therefore, sulfate reduction using lactate is a thermodynamically favorable process producing energy for microbial growth.

The E° value of the couple Fe²⁺/Fe⁰ is close to that of the couple CO₂ + acetate/lactate. Consequently, the coupling of iron oxidation with sulfate reduction is thermodynamically favorable. Thus, elemental iron is an electron donor (i.e., energy source) for SRB metabolism.

In 2009, Gu et al. [66] proposed the biocatalytic cathodic sulfate reduction (BCSR) theory, which is the first theory that utilizes bioenergetics to explain the motive behind SRB MIC against carbon steel. The BCSR theory assumes that the electrons released by iron oxidation are transported across the SRB cell wall and finally uti-

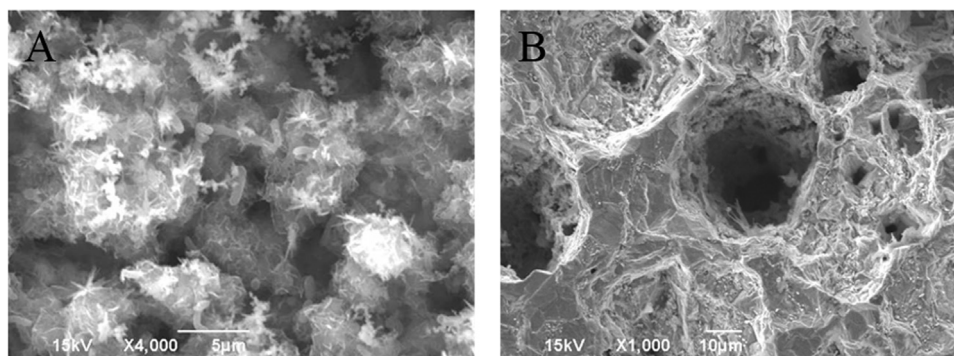


Fig. 1. Scanning electron microscopy (SEM) images of a biofilm (A) and corrosion pits (B) (after biofilm removal) on C1018 carbon steel after 7 days of incubation at 37 °C in ATCC 1249 culture medium inoculated with a seed culture from an oilfield biofilm consortium [49].
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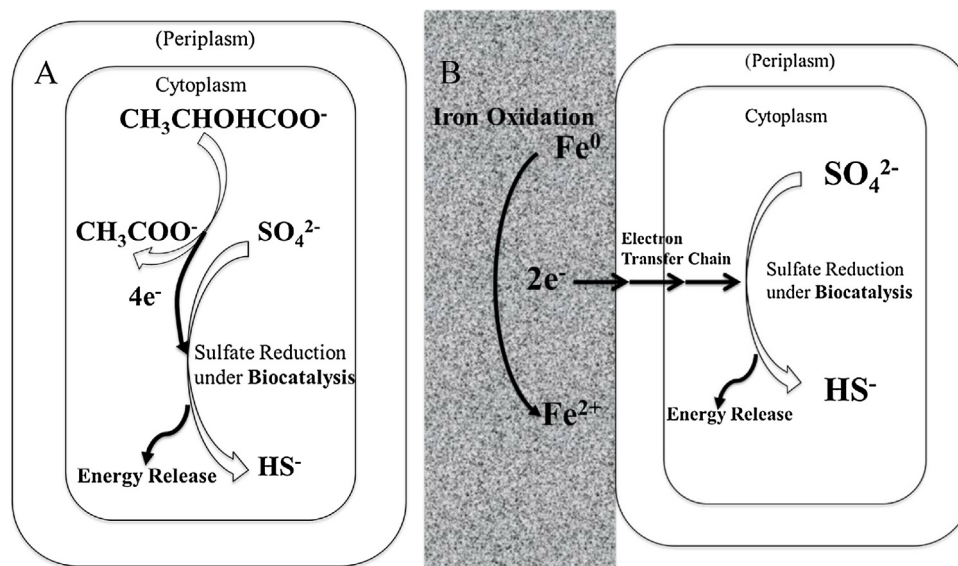
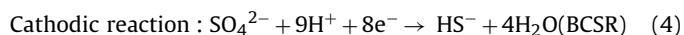
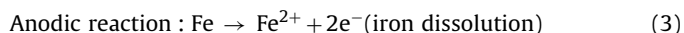


Fig. 2. Schematic drawings of: (A) organic carbon-sulfate respiration and (B) BCSR with iron as the electron donor (Only gram-negative SRB have a periplasm.).

lized for sulfate reduction in the cytoplasm of SRB, which results in corrosion as shown in Reactions 3 and 4 below.



Instead of a physical cathode, SRB serves as some sort of a biocathode, which is a concept in the research of microbial fuel cell (MFC) technology for wastewater treatment and bioenergy production [67]. The coupling of Reactions 3 and 4 yields a change in the Gibbs free energy of -178 kJ/mol of sulfate, which implies that iron dissolution coupled with sulfate reduction is thermodynamically favorable or energy producing for SRB growth. However, without biocatalysis of SRB, sulfate reduction has a kinetically negligible reaction rate, which means that SRB biofilms are needed to corrode steel. The biocatalysis accelerates the kinetics, resulting in the occurrence of MIC.

In the BCSR theory, sulfate is the terminal electron acceptor. Oxidation of insoluble iron occurs outside SRB cells while sulfate reduction occurs inside SRB cells. The electrons released by elemental iron oxidation are transferred from outside the cells across the cell wall to the SRB cytoplasm where sulfate reduction takes place. Organic carbon such as lactate is soluble. Its biocatalyzed oxidation occurs in the SRB cytoplasm. Thus, the released electrons do not need to be transported across the SRB cell wall. Fig. 2 shows the

similarities and differences between organic carbon-sulfate reduction and BCSR with iron as the electron donor. As shown in Fig. 2B, EET in the form of an electron transfer chain is required for SRB to use elemental iron as the electron donor.

Biofilms act as mass transfer barriers to carbon source diffusion. The top layer of a biofilm can consume a certain amount of carbon source leaving less organic carbon for the sessile cells in the bottom layer of the biofilm, leading to a local starved environment for sessile cells near or directly on the metal surface. When there is a lack of carbon sources (electron donor), starved SRB will switch to elemental iron as the electron donor to produce energy for maintenance. Carbon starvation experimentally has been shown to accelerate C1018 carbon steel corrosion, despite a lowered sessile cell count as a result of starvation. With less carbon in the culture medium, sessile SRB cells have been reported to obtain electrons from iron, causing severe corrosion, in contrast to SRB biofilms in full-strength nutrient media [68]. Most recently, Jia et al. also confirmed that sessile *P. aeruginosa* cells, if cultured as an NRB, became more corrosive under organic carbon starvation [69]. The vulnerability of a metal material to a biofilm in this case depends on whether it can be used as an electron donor for microbial metabolism [70].

Parallel to BCSR, BCNR (biocatalytic cathodic nitrate reduction) can be used to explain NRB MIC. Like sulfate reduction, nitrate reduction can also lead to MIC. For instance, *Bacillus licheniformis* grown as NRB created a $14.5\text{-}\mu\text{m}$ pit on a C1018 carbon steel in one

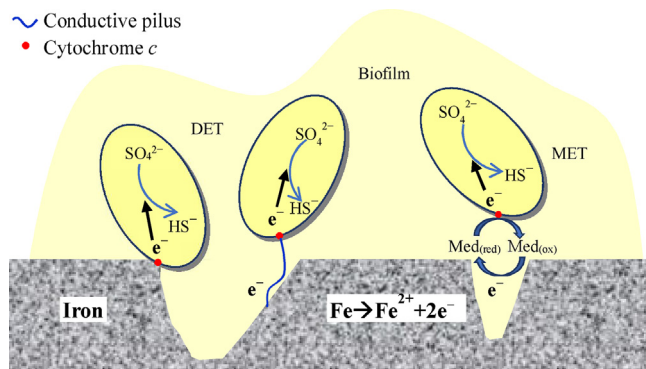
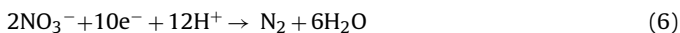
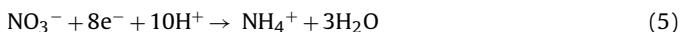


Fig. 3. Schematic illustration of DET and MET in MIC by sessile SRB cells, “Med_(red)” denotes the reduced form of an electron mediator after electron uptake and “Med_(ox)” the oxidized form after electron loss.

week [41]. Table 1 data shows that both nitrate reduction and nitrite reduction can be coupled with iron oxidation to yield thermodynamically favorable corrosion processes with energy generation for microbial growth or maintenance. Nitrate reduction and nitrite reduction have much higher reduction potentials than sulfate reduction (see Table 1).



3. EET in MIC

While bioenergetics explains why MIC occur, the concept of EET was introduced to MIC research involving SRB and NRB by Gu and Xu in 2010 [68,71,72] to explain how the corrosion process happens. They used the concept of EET in bioelectrochemistry to study the cross-cell wall electron transfer in MIC by SRB and NRB. This concept was adapted from MFC research, which relies on EET for power generation. Extracellular electrons released from iron oxidation are used by biofilm. Without a biofilm, the electrons cannot be utilized by planktonic cells suspended in the liquid because electrons, unlike ions, cannot “swim” in the liquid.

EET has three main ways: direct contact, conductive pilus, and electron mediator [73,74]. The first two approaches are called direct electron transfer (DET), and the third one is called mediated electron transfer (MET), as illustrated in Fig. 3. In DET, a direct contact between microorganisms and the iron surface is needed, while MET involves soluble redox mediators, which are secreted by microbes or pre-existing in the solution [75,76].

In general, cytochromes such as cyt *c*, conductive pili (nanowires), and iron-sulfur proteins on or in cell membranes influence electron transfer in DET [77]. A group of researchers showed that in a culture medium without an organic carbon source, SRB cells formed pili to attach to the iron surface [78] to harvest electrons (Fig. 4). These pili were absent if the SRB were grown with organic carbon in the culture medium. Xu et al. interpreted that the SRB cells used these pili to transfer electrons from the carbon steel surface for sulfate reduction to survive carbon source starvation [41].

Gu and Xu stated that SRB MIC can also involve MET [72,79]. With the addition of electron mediators to the medium, SRB cells caused more severe MIC and deeper pits on C1018 carbon steel and stainless steel [24]. With the addition of electron mediators, NRB also became more corrosive and caused more severe MIC attack [80]. In both SRB-MIC and NRB-MIC processes, EET was the rate limiting process.

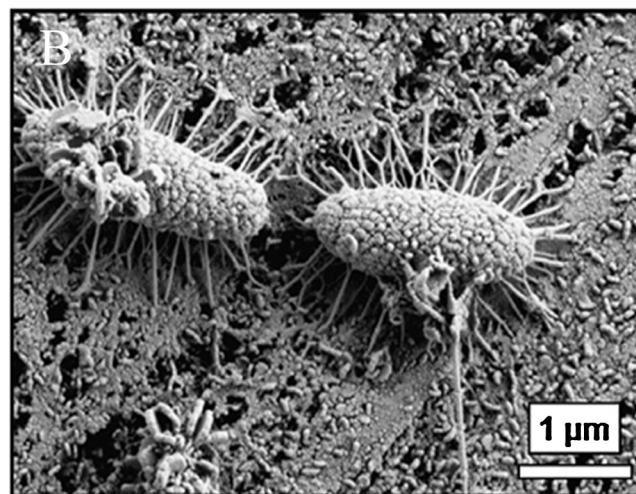
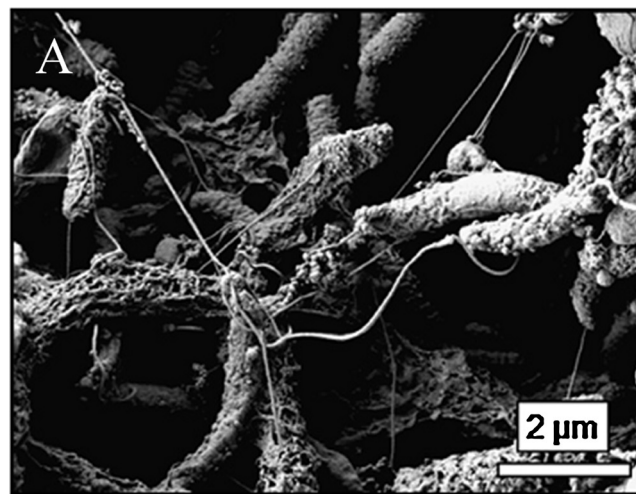


Fig. 4. SEM images of SRB cells growing in modified Postgate media lacking carbon. Source: (A) in a colony and (B) individually in a culture medium lacking organic carbon [78]. (Reproduced with permission from Elsevier.)

4. MIC classification

Apart from SRB and NRB, other microorganisms also can cause anaerobic corrosion. Gu [81,82] classified MIC into three types. Type I MIC involves respiration using sulfate or nitrate as electron acceptors. Cross-cell wall EET is required for Type I MIC. Thus, Type I MIC is called EET-MIC [83,84]. Microbes capable of EET are involved in MFC because they are electrogenic when EET direction is reversed to transport electrons from organic carbon oxidation in cytoplasm to an external anode. These electrogenic microbes can cause MIC, if there is a local shortage of organic carbon (electron donor). SRB-MIC and NRB-MIC mechanisms as discussed above belong to EET-MIC.

Type II MIC is caused by secreted metabolites of microbes. Thus, Type II MIC is called metabolite MIC (M-MIC) [83,84]. Corrosive metabolites are oxidants such as protons, organic acids, and sulfides. Examples are MIC of steel by acids secreted by acid producing bacteria (APB) in biofilms, and copper MIC by H₂S excreted by SRB [85]. In the absence of an externally supplied oxidant, microbes such as APB can perform anaerobic fermentation, which often produces organic acids. With a sufficiently acidic pH under an APB biofilm, proton reduction can be coupled with iron oxidation to yield a thermodynamically favorable corrosion process. Like EET-MIC, M-MIC is also an electrochemical corrosion process, because iron oxidation and proton reduction are two separable electrode reactions. The word “metabolite” is used in M-MIC instead of the

Table 2
Comparison of EET-MIC and M-MIC.

EET-MIC	M-MIC
Anaerobic respiration	Anaerobic fermentation or respiration
Intracellular reduction of oxidant	Extracellular reduction of oxidant
Biocatalysis needed for reduction	Biocatalysis not needed for reduction

word “chemical,” because “chemical corrosion” refers to the direct reaction of a metal with an oxidant, usually at high temperatures, without separable oxidation and reduction reactions, unlike in an electrochemical corrosion process. Unlike EET-MIC, M-MIC does not need biocatalysis. For example, proton is secreted by APB and it attacks metal extracellularly without enzymes. Because of this, M-MIC has abiotic counterparts. For example, acetic acid corrosion can be either due to APB excreting acetic acid or due to abiotic acetic acid. Apart from APB, fungi such as *Aspergillus niger* secrete acidic metabolites (e.g., oxalic acid) that cause M-MIC [22].

A recent report by Jia et al. found that a low H₂S concentration in the culture medium caused less toxicity to sessile cells and also led to a thinner partially protective sulfide film than that with a high H₂S concentration, which resulted in increased EET-MIC. They concluded that biogenic H₂S corrosion is not a major contributor to carbon steel MIC by SRB [83]. This means that M-MIC was not the main factor in carbon steel MIC by SRB, but rather EET-MIC. Interestingly, Fu et al. [85] found that copper was not energetic enough as an electron donor for direct sulfate reduction by SRB. However, copper is corroded by SRB. Their thermodynamic analysis showed that the corrosion was caused by sulfide and proton with Cu₂S as the corrosion product. Thus, copper corrosion by sulfate-respiring SRB belongs to M-MIC. This means that for SRB, both EET-MIC and M-MIC can occur depending on the metal type.

In EET-MIC, SRB and NRB corrode carbon steel intentionally for energy harvesting. In the M-MIC process, fermentative bacteria and fungi in biofilms secrete organic acids, which cause M-MIC. The corrosion process results in H₂, which is an energy source for those microorganisms possessing hydrogenase. It is still unknown whether microbes cause M-MIC for the purpose to harvest energy. H₂ can also cause physical/electrochemical corrosion like fatigue cracking and hydrogen embrittlement. Furthermore, acidification of the culture medium will enhance the rate of crack growth [36,86].

A comparison of EET-MIC and M-MIC characteristics is presented in Table 1. The characteristics of EET-MIC and M-MIC mechanisms provide useful insights into MIC by SRB, NRB, and APB, which dominate anaerobic MIC processes (Table 2).

In addition to Type I MIC and Type II MIC, Type III MIC can be classified as the biodegradation of organic materials such as organic polymers by microorganisms [68]. Microbes, especially fungi in a warm and humid environment, excrete enzymes to degrade large molecules to produce utilizable small organic molecules. Type III MIC can damage polymer insulation of electrical systems, etc. [87]. It is not the focus of this review.

5. Conclusion

In this multidisciplinary review, different mechanisms of MIC are discussed using the concept of bioenergetics, electron transfer theories, and respiration types. In an environment lacking carbon sources and other electron donors, which are necessary for the energy metabolism of microorganisms, microbes can switch to the utilization of elemental iron or other energetic metals as electron donors, leading to MIC. Two forms of EET are involved: DET and MET. The two main types of anaerobic MIC of metals are classified according to microbial respiration types and EET involvement. This review can help researchers and field engineers to judge why

and how an MIC process occurs depending on local environmental conditions.

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