Bacterial secondary metabolites as promising "green" microbiologically influenced corrosion inhibitors/biocides: a review

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Abstract. A number of ecological, economic and medical problems arise as a result of damage to building materials by various groups of microorganisms (microbiologically influenced corrosion), as well as due to the treatment of these materials with toxic chemical biocides/inhibitors. Artificial addition of "green" biocides/inhibitors (in particular, of secondary metabolites of heterotrophic bacteria) into corrosive environments is considered as a promising approach to avoid such problems. This review is an attempt to summarize the information on the possibilities of using microbial secondary metabolites as "green" inhibitors/biocides of microbiologically influenced corrosion. The information on the anticorrosive, antimicrobial and/or antibiofilm activity of the secondary metabolites of microorganisms (antibiotics, lipopeptides, exopolysaccharides, siderophores) as well as metallic nanoparticles biosynthesized and conjugated with bacterial secondary metabolites as promising biocides/inhibitors of microbiologically influenced corrosion to protect materials from microbial deterioration, and further prospects of this approach are analyzed. It is emphasized that the inhibition of microbiologically influenced corrosion processes with the help of secondary metabolites is an environmentally safe approach that can be considered from four interrelated points of view: corrosion inhibitors, antimicrobial compounds, antibiofilm compounds, regulators of microbial corrosively active groups.

Keywords: "green" inhibitors/biocides, heterotrophic bacteria, microbial secondary metabolites, microbiologically influenced corrosion, sulfate-reducing bacteria.

1. Introduction

The interaction of microorganisms with construction materials leads to their damage, which results in a number of environmental and economic problems (Lavanya, 2021). Such an interaction is considered as microbiologically influenced corrosion (MIC), which is essentially a bioelectrochemical process occurring in a biofilm on the surface of a material (Andreiuk et al., 2005).

Today, the determining role of sulfidogenic microbial communities in MIC is generally recognized (Beech & Gaylarde, 1999; Marchal, 1999; Andreiuk et al., 2005; Purish & Asaulenko, 2007). In such groups, sulfate-reducing bacteria (SRB) and other sulfate-reducing prokaryotes (Lan et al., 2022) are considered as the main causing agents of MIC, and other groups of bacteria (denitrifying, ammonifying, iron-reducing, mucus-producing, acid-producing) are involved in various mechanisms of MIC (Wakai et al., 2022; Tkachuk & Zelena, 2023), and associated with the development and maintaining of the environment for SRB growth (Andreiuk et al., 2005).

In order to prevent MIC, chemical compounds are used - inhibitors and biocides. Chemically synthesized biocides/inhibitors belonging to the various classes of organic compounds and hazardous to the environment and health are used to protect against MIC (Ashraf et al., 2014; Michalak & Chojnacka, 2014; Scarascia et al., 2016; Vaithiyanathan et al., 2018). Currently, the prevention of microbial damage to materials is possible with the use of "green" biocides/inhibitors (Ashraf et al., 2014; Vaithiyanathan et al., 2018; Fawzy et al., 2023; Verma et al., 2023), in particular secondary metabolites of heterotrophic bacteria (Zuo, 2007; Płaza & Achal, 2020; Wang et al., 2022; Fawzy et al., 2023), under their artificial addition into environments with a corrosion hazard.

There are principles of "green" chemistry, presented in the form of a scheme, according to which compounds can be classified as "green" biocides (Warner et al., 2004; Ashraf et al., 2014).

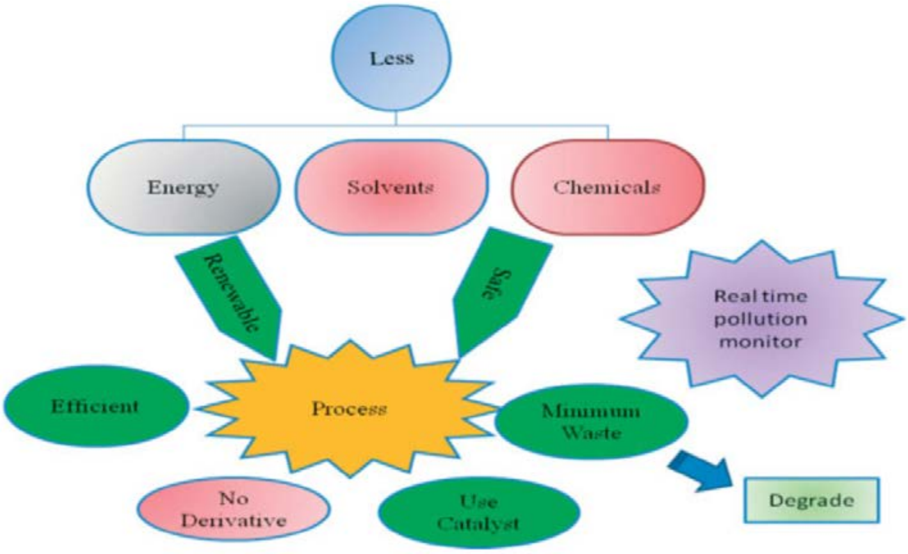

Figure 1. The criteria for being a "green" biocide (Ashraf et al., 2014)

In general, the problem of developing ways of protecting materials from corrosion with "green" inhibitors/biocides is widely discussed in a number of articles (Rani & Basu, 2012; Shehata et al., 2018; Lavanya, 2021; Zhang et al., 2022b; Ghosal & Lavanya, 2023; Wang et al., 2023a). Literature reviews analyzed data on the variety of areas where "green" inhibitors as an effective alternative to chemical compounds can be used, their mechanisms of action, the variety of sources of receipt and their efficacy. At the same time, the issue of the diversity of microbial secondary metabolites as "green" inhibitors/biocides against MIC, variety of organisms that can synthesize them, and the prospects of new applications is insufficiently covered, which determined the aim of this study.

In the presented review, we tried to decipher some peculiarities of “green” biocides associated with ecological aspects:

1) the potential impact of secondary metabolites produced by microorganisms on MIC sites;

2) the diversity of microorganisms producing secondary metabolites with antimicrobial, anticorrosive, antibiofilm properties for protection against MIC;

3) the chemical nature of such compounds;

4) influence of metabolites with anticorrosive properties and their complexes with metal nanoparticles on environments.

2. General characteristics of secondary metabolites

Secondary metabolites include compounds synthesized by some species of microorganisms, which are mainly formed after the cessation of growth in the form of a complex of similar compounds (Kondrashevska et al., 2018). Often, the ability to synthesize them is lost due to mutations or in the process of storing bacterial producers. Secondary metabolites are characterized by the diverse chemical structures and demonstrate a wide range of biological activities. In particular, secondary metabolites are represented by antibiotics, exopolysaccharides, enzyme inhibitors, immunosuppressants, polyketides, terpenes, terpenoids (Kondrashevska et al., 2018). Secondary metabolites of microorganisms also include biosurfactants (Zahed et al., 2022).

The following classification of biosurfactants produced by microorganisms was proposed (Płaza et al., 2014): 1) hydroxylated and cross linked fatty acids (for examples, mycolic acids); 2) glycolipids (for examples, rhamnolipids, trehalolipids, mannosylerythritol lipids and sophorolipids); 3) lipopolysaccharides; 4) lipoproteins-lipopeptides (cyclic lipopeptides are classified into four families: the surfactins, the iturins, the fengycins or plipastatins and the kurstakins); 5) fatty acids, phospholipids and neutral lipids; 6) polymeric biosurfactants (for examples, emulsan, liposan, alasan, lipomanan and other polysaccharide-protein complexes). Secondary metabolites play roles in nutrient acquisition, metabolism, and inhibition or other interactions with surrounding organisms or the environment (Sharrar et al., 2020).

Therefore, secondary metabolites are compounds of different chemical nature, characterized by biological activity and synthesized by microorganisms-producers. They attract the attention of MIC researchers as potential biocides/inhibitors because they are considered eco-friendly (“green”) compounds.

3. Bacterial secondary metabolites with anticorrosive, antimicrobial and/or antibiofilm activity in the context of MIC processes

To prevent MIC, it is important to remove corrosive substances through microbial activity, for example, microbes consuming reactive oxygen through aerobic respiration; to provide the formation of a protective layer, which could be established by overproduction of extracellular polymeric substance (EPS) by non-damaging microbes; destroy/suppress participating microorganisms and/or prevent/eliminate biofilm formation produced by them (the latter one can be achieved through antimicrobial production by non-corrosive microorganisms) (Zuo, 2007; Videla & Herrera, 2009).

A variety of bacterial secondary metabolites exhibit these properties, which allows them to be considered as “green” inhibitors/biocides for protection against MIC. In particular, there are reports on the effectiveness of antibiotics, biosurfactants, siderophores to prevent MIC, including those synthesized by genetically modified bacterial strains.

3.1. Antibiotics

Antibiotics refer to a group of antimicrobial compounds that are able to kill and/or prevent the growth of microorganisms, providing prevention or treatment of infections (Di Martino, 2022). Due to their antimicrobial properties, antibiotics have proven to be effective compounds for the prevention of MIC. The antibiotics indolicidin, bactenecin, and probacterin released by biofilms of genetically engineered *B. subtilis* strain BE1500 and *B. subtilis* strain WB600 were able to inhibit the growth of *Desulfovibrio vulgaris* and *D. gigas* (SRBs that cause corrosion), and significantly reduced corrosion in continuous culture (Jayaraman et al., 1999a). Inhibition of SRB colonization and reduction of steel corrosion were also observed for the gramicidin S peptide naturally produced by the biofilm-forming strain *Bacillus brevis* (*B. brevis*) strain 18 (Jayaraman et al., 1999b). If the antibiotic ampicillin was added before SRB colonization, it only inhibited SRB growth. However, the associative culture of S-gramicidin-producing *B. brevis* strain 18 with SRB completely blocked the growth of the latter (Jayaraman et al., 1999b). Today it is known that norfloxacin and clindamycin hydrochloride at a concentration of 40 mg/l effectively inhibit MIC caused by anaerobic microbial consortium (Liu et al., 2024). It turned out that antibiotics synthesized by microorganisms are effective not only against MIC. Antibiotics show high anti-corrosion properties in acid solutions (Shukla et al., 2009; Fouda et al., 2016; Kamel et al., 2021).

However, it is known that the use of antibiotics leads to the formation of antibiotic-resistant strains of microorganisms that are difficult to fight (World Health Organization, 2014). Therefore, their use for the prevention of MIC carries the risk of the formation of antibiotic-resistant, corrosion-active strains and the strengthening of the corrosion process. In addition, copper and lead corrosion products increased antibiotic resistance, although iron corrosion products had minimal impact on antibiotic resistance (Folvarska et al., 2024). Thus, the use of antibiotics to prevent MIC requires further in-depth research.

3.2. Biosurfactants

The researches who studied MIC have paid a considerable attention to the possibility of using biosurfactants to prevent corrosive processes (Płaza & Achal, 2020; Sivakumar et al., 2024). Synthesis of extracellular molecules such as biosurfactants or proteins should have a major effect on bacterial adhesion. These molecules can adsorb on surfaces and change their energy characteristics (Garry et al., 1998). The aim of the review by Malik et al. (2011) was to delineate the ability of surfactants to inhibit corrosion on different metal surfaces. Different potential applications and properties of different types of surfactants have also been discussed. Various parameters like effect of surfactant concentration, temperature and the mechanism of corrosion inhibition and mode of adsorption were also discussed in the review article (Malik et al., 2011). Biosurfactants are proven to be one of the best eco-friendly anticorrosion substances to inhibit the biocorrosion process and protect materials against corrosion.

Biosurfactants have recently became one of the important products of bioeconomy with multiplying applications, while there is scare knowledge on their using in biocorrosion treatment. In the review (Płaza & Achal, 2020), the recent findings on the application of biosurfactants as eco-friendly and innovative biocides against biocorrosion were highlighted. It has been shown that lipopeptides produced by *B. subtilis* (surfactin, iturin and fengycin) were able to modify the surface hydrophobicity of stainless steel and Teflon and, as a result, affect microbial adhesion to their surface (Shakerifard et al., 2009). The authors showed that these effects depend on the type of lipopeptide, concentration and substrate. At the same time, surfactin and fengycin increased the hydrophobicity of steel, while iturin had no effect. Surface hydrophobicity can be modified by microbial surfactants and is a novel strategy to reduce bacterial adhesion to surfaces (Shakerifard et al., 2009). Biosurfactants block the active sites and retard the anodic and cathodic processes and act as mixed-type inhibitors (Verma et al., 2023).

Bacillus sp. strain F7 produces a biosurfactant, that is a main component of a new antimicrobial substance is being developed for the prevention and destruction of biofilm and is a promising candidate for a new anticorrosion agent (Purwasena et al., 2019). The specified biosurfactant is capable of inhibiting the attachment of *Pseudomonas* sp. strain 1 and *Pseudomonas* sp. strain 2 to ST37 carbon steel surfaces and is also able to remove pre-formed biofilm on steel surfaces. This study showed a reduction in the corrosion rate of ST37 carbon steel as a result of the treatment of the material with biosurfactants (Purwasena et al., 2019).

Thus, the effect of aluminum-chelating anionic peptides, which are released by biofilms of natural and genetically modified *Bacillus*, on pitting corrosion of aluminum alloy in continuous reactors was studied by Ornek et al. (2002). At the same time, a 90% reduction of the corrosion rate of aluminum alloy 2024 was revealed due to the biofilm of *Bacillus licheniformis* (*B. licheniformis*) strain WB600/pBE92-Asp, which formed γ-polyglutamate. The authors noted a significant reduction in aluminum corrosion by the *Bacillus subtilis* (*B. subtilis*) biofilm compared to the sterile control (without bacterial/bacilli biofilm). One of the peptides secreted by the genetically engineered biofilm only slightly reduced pitting compared to the biofilm on the control material.

Lipopeptide biosurfactants (surfactin, fengycin and iturin), produced by *Bacillus* sp. with a high level of similarity to *B. subtilis, B. amyloliquefaciens* and *B. pumilus*, showed high antifungal properties when applied to objects of cultural heritage (Silva et al., 2017). The control of biodegradation processes in the context of heritage using green compounds was considered as an important perspective (Caldeira, 2021).

Amino acid-based surfactants (biosurfactants), including sodium N-dodecyl asparagine, sodium N-dodecyl tryptophan, sodium N-dodecyl histidine indicated their potential the copper corrosion inhibition efficacy in H_2SO_4 (1.0 M) solution at 298 K) and antibacterial properties (Fawzy et al., 2023).

Therefore, the production by microorganisms of biosurfactants with antibacterial properties, the ability to modify the surface of the material, is the most studied issue regarding the prevention of MIC by secondary metabolites. This area of research is interesting and important, especially given the growth of research using chemically pure biosurfactants.

3.3. Microbial diversity of MIC biocides/inhibitor producers

The involvement of microorganisms of various species in MIC processes attracts attention from the point of view of both their interaction and the development of the corrosion process, as well as the possibility of using artificially added species (or their metabolites) to prevent microbial deterioration of materials.

Since the secondary metabolites of some microorganisms are able to influence the existence of others, the question arises of the functioning of corrosively active groups in natural conditions with different secondary metabolic profiles in them. Considerable attention is focused on primary metabolites using metabolomic surveys (Beale et al., 2015). Limited information is available about secondary metabolites from MIC sites. In particular, the studies of Bonifay et al. (2017) found that in the “high corrosion” system secondary metabolism is represented by 110 putative compounds represent secondary metabolism, and a total of 735 putatively identified metabolites were not associated with any specific metabolic pathway in the Kyoto Encyclopedia of Genes and Genomes (KEGG) database. Therefore, the question of the role of these compounds in the functioning of the corrosively active group arises, and further researches will provide an answer to it.

3.3.1. Potential of *Bacillus velezensis* to prevent MIC

B. velezensis strains are known for their biocontrol properties when applied in agricultural technologies (Chowdhury et al., 2015; Khan et al., 2016; Fazle Rabbee & Baek, 2020). Significant anti-biofilm/anti-biofouling properties of *B. velezensis* have been noted by a number of researchers. It should be noted that the species name *B. amyloliquefaciens* subsp. *plantarum*, *B. methylotrophicus*, "*Bacillus oryzicola*" and *B. methylotrophicus* subsp. *plantarum* are modern heterotypic synonyms of *Bacillus velezensis* (*B. velezensis*) (NCBI, Taxonomy Browser; Dunlap et al., 2016).

Bacillus sp. strain H2O-1 was active against sulfate-reducing bacteria and had 99.8 and 99.5% 16S rRNA gene sequence similarity to the type strains of *Bacillus amyloliquefaciens* (*B. amyloliquefaciens*) and *Bacillus methylotrophicus* (*B. methylotrophicus*), respectively (Korenblum et al., 2012). It was shown that *Bacillus* sp. strain H2O-1 (isolated from the connate water of a Brazilian reservoir) produces an antimicrobial substance (provisional designation AMS H2O-1) active against sulfate-reducing bacteria. AMS H2O-1 was found to be a mixture of four surfactin-like homologues, and its biocidal activity and surfactant properties indicated a possibility to control SRB growth. The authors highlight a potential alternative of using such a compound to prevent SRB growth in the oil industry versus chemical biocides or surface coatings that are currently in use (Korenblum et al., 2012).

The high activity of proteases isolated from *B. velezensis* strain SM-1 against marine fouling was revealed (Wang et al., 2019). The authors note that this may be the basis of a modern ecological approach to the development of antifouling materials. The potential of *B. methylotrophicus* strain WY to control membrane biofouling was detected based on the study of the ability of the bacterium to quench quorum sensing (Khan et al., 2016). Thus, this strain has a significant effect on the degradation of acylhomoserinlactone (AHSL), which is a kind of signaling molecule necessary for the development of biofilm.

The inhibitory activity of *B. velezensis* strain K68 was investigated against *Streptococcus mutans* biofilm on polystyrene (Yoo et al., 2019). The antibiofilm properties of the culture liquid of *B. velezensis* strain NUChC C1 and strain NUChC C2b were demonstrated in the experiment with sulfate-reducing bacteria *Desulfovibrio oryzae* and the material polyethylene terephthalate (Tkachuk et al., 2021a). It was established that the studied strains of *B. velezensis* have active genes responsible for the synthesis of the siderophore bacillibactin (Tkachuk et al., 2021a).

Various strains of *B. velezensis*, in addition to bacillibactin (Mongkolthanaruk, 2012; Lamb, 2015; Silva et al., 2019; Fazle Rabbee & Baek, 2020), are able to synthesize a number of other compounds with antimicrobial properties: bacilysin (Chen et al., 2009; Özcengiz & Öğülür, 2015; Khan et al., 2016; Silva et al., 2019; Fazle Rabbee & Baek, 2020), difficidin (Chen et al., 2009; Silva et al., 2019; Fazle Rabbee & Baek, 2020), fengycin (Mongkolthanaruk, 2012; Silva et al., 2019; Fazle

Rabbee & Baek, 2020; Yang et al., 2020), bacillaene (Butcher et al., 2007; Silva et al., 2019), macrolactin (Silva et al., 2019; Fazle Rabbee & Baek, 2020), surfactin (Vollenbroich et al., 1997; Silva et al., 2019; Fazle Rabbee & Baek, 2020; Yang et al., 2020), plantazolicins (Kalyon et al., 2011; Silva et al., 2019), mersacidin (Brötz et al., 1998; Sahl & Bierbaum, 1998; Kruszewska et al., 2004; Appleyard et al., 2009; Silva et al., 2019), subtilin (Mongkolthanaruk, 2012; Silva et al. 2019), locillomycins (Luo et al., 2019; Silva et al., 2019), bacillomycins (Mongkolthanaruk, 2012; Pan et al., 2017; Suneeta et al., 2018; Silva et al. 2019; Balderas-Ruíz et al., 2020; Fazle Rabbee & Baek, 2020).

Therefore, secondary metabolites of *B. velezensis* should be considered as the promising “green” biocides/MIC inhibitors (Khan et al., 2016; Wang et al., 2019; Yoo et al., 2019; Tkachuk et al., 2021b; Tkachuk & Zelena, 2022). Besides, siderophores synthesized by other members of the genus *Bacillus* are also attracting attention as “green” inhibitors/biocides, and this issue is under discussion (Tkachuk & Zelena, 2021). Generally, siderophores are considered as the environmentally friendly compounds with high protective properties against corrosion of metals and steel (Little & Mansfeld, 1995; McCafferty & McArdle, 1995; Little et al., 2008; Rajala, 2017; Javaherdashti & Alasvand, 2019; Pérez-Miranda et al., 2020; Zanna et al., 2020). They are “green” corrosion inhibitors, as are macromolecules, bacteria, alkaloids, and polyphenols (Pérez-Miranda et al., 2020). The issue of using siderophores to protect materials from MIC is open and requires further research (Tkachuk & Zelena, 2021).

3.3.2. Metabolites of *Streptomyces* spp. and other bacteria as biocides/inhibitors of MIC

A number of other species of bacteria and their secondary metabolites are also being actively studied as “green” inhibitors/biocides of MIC. Secondary metabolites with antimicrobial/anticorrosive properties are also produced by members of the Actinobacteria genus *Streptomyces*. Thus, studies of Okon (2010) showed that albomycin (a product of fermentation *Streptomyces griseus*) is a good adsorption inhibitor of zinc corrosion in a solution of sulfate acid (Okon, 2010). It was found that *Streptomyces lunalinharesii* (*S. lunalinharesii*) strain 235 inhibits the growth of *Bacillus pumilus* strain LF-4 and *Desulfovibrio alaskensis* strain NCIMB 13491, which are participants in the formation of biofilms and the process of biocorrosion (Pacheco da Rosa et al., 2013). Prevention of SRB biofilm formation by this strain is due to antimicrobial substances that a strain synthesized (Rosa et al., 2016). The protective ability of an intact non-viable biofilm was evaluated by adding kanamycin to kill biofilm bacteria *in situ*, and non-biofilm-forming *Streptomyces lividans* was used to assess the role of metabolites in corrosion inhibition. The ability of a fully developed biofilm to inhibit corrosion with mild agitation was also probed (Jayaraman et al., 1997). It is shown that marine *Streptomyces* sp. effectively inhibit SRB-induced corrosion (Wang et al., 2023b). Green defense for MIC inhibition in this case is considered from the point of view of “bacteriostasis with bacteria”. The property of *Streptomyces gardneri* to prevent biofilm formation of heterotrophs was shown in the study of Tkachuk and Zelena (2022). Antibiofilm properties may be caused by the action of secondary metabolites of this strain of streptomycetes.

In the study by Jayaraman et al. (1997), protective biofilms for corrosion prevention were generated with stationary and mildly shaken cultures of *Pseudomonas fragi* and *Escherichia coli* strain DH5a. The main objectives of this study were to investigate the mechanism responsible for corrosion inhibition and to assess the potential of a biopolymer film for corrosion inhibition. This is the first report of corrosion inhibition with these strains (Jayaraman et al., 1997). It was showed that *Pseudomonas putida* strain RSS biopassivates of mild steel for long term corrosion inhibition (Suma et al., 2019). This is due to the formation of a strong and stable iron-extracellular polymeric substance coating over the concrete bacterial phosphate layer. The developed biofilm remained adhered on the surface of the mild steel surface even after the death of bacterial cells, and it conferred further protection. The corrosion resistance of mild steel surface after mechanically removing biofilm was also investigated. Results of electrochemical studies showed no traces of corrosion even after 12 months of immersion with negligible corrosion rate of 3.01×10^{-2} mm/year. The developed surface biopassivation system can be employed for long term corrosion inhibition of steel structures in aquatic systems (Suma et al., 2019). EPSs are mostly composed of polysaccharides (exopolysaccharides) and proteins (Flemming et al., 2000; Donlan, 2002). Polysaccharides (EPS), especially homopolysaccharides produced by lactic acid bacteria *Lactobacillus delbrueckii* strain K27, *Lactobacillus delbrueckii* strain B8, *Lactobacillus delbrueckii* strain KO43, *Lactobacillus delbrueckii* strain K3, *Lactobacillus delbrueckii* strain K15 and *Lactobacillus delbrueckii* strain K17 showed anti-corrosive properties (Ignatova-Ivanova & Ivanov, 2016). Exopolysaccharides produced by other lactic acid bacteria *Lactobacillus mesenteroides* have shown their electrochemical and physical properties and suitability as anti-corrosion coatings (Finkenstadt et al., 2017). The mechanism of anticorrosive action of bacterial polysaccharide coatings in relation to low carbon steel was established, which consisted in reducing ionic diffusion rates and maintaining a relatively passive metal-coating interface (Finkenstadt et al., 2017). It is also noted that EPS produced by *Vibrio neocaledonicus* strain MS1 (Moradi et al., 2018) and *Marinobacter aquaeolei* (Khan et al., 2020) provide corrosion inhibition. Green methodologies for corrosion protection by EPS are provided (Wang et al., 2022).

Therefore, the search for effective strains-producers of secondary metabolites, which can be potentially used as “green” inhibitors/biocides could be carried out among bacterial species: *Bacillus* spp. (in particular, *B. velezensis*), *Streptomyces* spp., *Lactobacillus delbrueckii*.

Summarizing information about the effect of bacterial secondary metabolites on MIC processes is shown in the Table 1.

Table 1. The effect of bacterial secondary metabolites on MIC processes.

Secondary metabolite	Bacteria-producers	Effects	References
Exopolysaccharides (EPS)	*Lactobacillus delbrueckii* strain K27, *Lactobacillus delbrueckii* strain B8, *Lactobacillus delbrueckii* strain KO43, *Lactobacillus delbrueckii* strain K3, *Lactobacillus delbrueckii* strain K15 and *Lactobacillus delbrueckii* strain K17	Inhibition of corrosion	(Ignatova-Ivanova & Ivanov, 2016)
	Lactobacillus mesenteroides	Inhibition of corrosion	(Finkenstadt et al., 2017)
	Vibrio neocaledonicus strain MS1	Inhibition of corrosion	(Moradi et al., 2018)
	Pseudomonas putida strain RSS	Inhibition of corrosion	(Suma et al., 2019)
	Marinobacter aquaeolei	Inhibition of corrosion	(Khan et al., 2020)
γ-polyglutamic acid	*B. licheniformis* strain WB600/pBE92-Asp	Formation of a protective biofilm, inhibition of corrosion	(Ornek et al., 2002)
Antibiotics indolicidin, bactenecin, probacterin	*B. subtilis* strain BE1500 and *B. subtilis* strain WB600	Suppression of the growth of SRB, inhibition of corrosion	(Jayaraman et al., 1999a)
Peptide gramicidin S	*B. brevis* strain 18	Suppression of the colonization of	(Jayaraman et al., 1999b)

		SRB, reduction of corrosion	
Biosurfactants lipopeptides surfactin, iturin A, fengycin	*B. subtilis*	Change in microbial adhesion to the surface of the material	(Shakerifard et al., 2009)
	Bacillus sp. (with a high level of similarity with *B. subtilis, B. amyloliquefaciens* and *B. pumilus*)	Antifungal	(Silva et al., 2017)
Biosurfactant	*Bacillus* sp. strain F7	Antimicrobial, antibiofilm, anticorrosive	(Purwasena et al., 2019)
Biosurfactants (a mixture of four surfactin-like homologues)	*Bacillus* sp. strain H2O-1 (99.5% sequence similarity of the 16S rRNA gene to *B. velezensis*)	Antimicrobial	(Korenblum et al., 2012)
Bacillibactin	*B. velezensis* strain NUChC C1 and *B. velezensis* strain NUChC C2b	Antibiofilm	(Tkachuk et al., 2021a)
Not specified	*B. methylotrophicus* strain WY (synonym of *B. velezensis*)	antibiofouling (quorum quenching properties)	(Khan et al., 2016)
	B. velezensis strain K68	Antibiofilm	(Yoo et al., 2019)
	S. lunalinharesii strain 235	Antimicrobial, antibiofilm	(Pacheco da Rosa et al., 2013; Rosa et al., 2016)

3.4. Metallic nanoparticles synthesized by bacteria as promising biocides/inhibitors of MIC

In the last decade, the biological properties of nanoparticles have been actively studied. They are used in health, agriculture, chemical, food, feed, space, cosmetic industries (Marslin et al., 2018;

Bahrulolum et al., 2021). The impact of nanoparticles on the processes of MIC and bacteria associated with it is also being investigated. It should be noted that the biosynthesis of metal and metal salt nanoparticles has been recognized as fast, harmless, ecological and economical process (Gahlawat & Choudhury, 2019; Bahrulolum et al., 2021).

The process of biosynthesis with microorganisms' participation is based on the mechanism of reducing the toxicity of metal salts, if they get into the environment, converting them into the less toxic nanoparticles with the involvement of primary and secondary metabolites of microorganisms (Singh et al., 2018; Messaoudi & Bendahou, 2020).

In particular, with the involvement of secondary metabolites, as the researchers note, extracellular biosynthesis took place: 1) silver nanoparticles: bacteria *Rhodococcus rhodochrous* (*R. rhodochrous*) and *Streptomyces* sp. (Alam et al., 2021), *Pseudomonas aeruginosa* (*P. aeruginosa*) strain DM1 (Kumari et al., 2017), strain BS-161R (Ganesh et al., 2010; Kumar & Mamidyala, 2011), strain OBP1 (Saikia et al., 2013), *B. subtilis* strain T-1 (Płaza et al., 2016; Mendrek et al., 2017; Yu et al., 2021), *Streptomyces ghanaensis* (*S. ghanaensis*) strain VITHM1 (Abirami & Kannabiran, 2016), *Cytobacillus firmus* (*C. firmus*) (Sudarsan et al., 2021), *Streptomyces griseoplanus* (*S. griseoplanus*) strain SAI-25 (Vijayabharathi et al., 2018), *Brevibacterium casei* (*B. casei*) strain MSA19 (Kiran et al., 2010), fungus *Nigrospora* sp. (Arrieta et al., 2017), fungi (Guilger-Casagrande & de Lima, 2019), fungus *Talaromyces purpureogenus* (Sharma et al., 2022); 2) gold nanoparticles: thermophilic filamentous fungi (Molnár et al., 2018), the tropical marine yeast *Yarrowia lipolytica* strain NCIM 3589 (Apte et al., 2013); 3) zinc oxide nanoparticles: fungi (Kamaruzaman et al., 2022); 4) ZnS nanoparticles: *P. aeruginosa* strain MTCC 2297 (Narayanan et al., 2010), *P. aeruginosa* strain BS01 (Hazra et al., 2013); 5) silver and gold nanoparticles: *P. aeruginosa* (Patil et al., 2022), *B. subtilis* strain BBK006 (Reddy et al., 2009); 6) Cu/Zn nanoparticles: *Aspergillus iizukae* (Noman et al., 2019); 7) cobalt nanoparticles: yeast *Starmerella bombicola* (Kasture et al., 2007).

Recently, classification of chemical compounds of plant secondary metabolites conjugated with nanoparticle biosynthesis was presented by Marslin et al. (2018). However, not much is known about the chemical nature of secondary metabolites of microorganisms conjugated with green biosynthesis of nanoparticles. Thus, among them are terpenoids and phenols (Sharma et al., 2022), lipopeptide (Yu et al., 2021), rhamnolipids (Ganesh et al., 2010; Narayanan et al., 2010; Saikia et al., 2013), glycolipid (Kiran et al., 2010), sophorolipids (Kasture et al., 2007), a peptide-based siderophore pyoverdine (Kumari et al., 2017; Patil et al., 2022), a blue-green pigment pyocyanin (Patil et al., 2022), 4-phenylbutanal, *N*-(2-(1*H*indol-3-yl)ethyl)-3-chloropropanamide and 3-benzylhexahydropyrrolo[1,2-a]pyrazine-1,4-dione (Abirami & Kannabiran, 2016), amines, carboxylic acids and aromatic phenols/alcohols (Vijayabharathi et al., 2018), melanin (Apte et al., 2013). Therefore, most researchers indicate the use of biosurfactants in the conjugated biosynthesis of nanoparticles.

The specified secondary metabolites have antimicrobial properties, except 4-phenylbutanal and N-[2-(1H-indol-3-yl)ethyl]-3-chloropropanamide, for which there are no publications on the study of antimicrobial properties. Thus, antimicrobial properties are known for terpenoids (Guimarães et al., 2019; Mahizan et al., 2019), phenols (Cueva et al., 2010), lipopeptides (Meena & Kanwar, 2015), in particular surfactin (Reddy et al., 2009), pyoverdines (Liu et al., 2021), pyocyanin (Raji El Feghali & Nawas, 2018), carboxylic acids (Vázquez et al., 2011), aromatic alcohols (Lucchini et al., 1990; Fraud et al., 2003), microbial surfactants (Paraszkiewicz et al., 2021), melanin (Zerrad et al., 2014), which can be used in the biosynthesis processes of nanoparticles with simultaneous enhancement of their antimicrobial properties against corrosively active microorganisms. 3-benzyl-hexahydro-pyrrolo[1,2-a]pyrazine-1,4-dione showed significant anti-quorum sensing activity against a model quorum sensing bacterium strain *P. aeruginosa* PAO1 and a clinical isolate *P. aeruginosa* PAH (Singh et al., 2019).

It was noted that the antimicrobial effect of nanoparticles in combination with biosurfactants has not yet been clarified (Płaza et al., 2014) and a nonspecific synergistic effect was observed (Chojniak et al., 2018). There are no other reports on the combined action of nanoparticles and secondary metabolites of bacteria in the currently available publications. It is postulated that chemical surfactants (Skoglund et al., 2017) and biosurfactants (Płaza et al., 2014; Płaza et al., 2016) are capable of preventing the aggregation of silver nanoparticles, thus preserving their high antimicrobial properties (Płaza et al., 2014). Rhamnolipids were also used for stabilization processes in syntheses of silver nanoparticles (Xie et al., 2006), NiO nanorods (Palanisamy, 2008) and NiO nanoparticles (Palanisamy & Raichur, 2009).

Bacterial secondary metabolites conjugated with nanoparticle biosynthesis are summarized in Table 2.

Table 2. Bacterial secondary metabolites conjugated with nanoparticle biosynthesis.

Bacteria conjugated with biosynthesis	Secondary metabolite conjugated with biosynthesis of nanoparticles	Nanoparticles	References
R. rhodochrous	Not specified	silver	(Alam et al., 2021)
Streptomyces sp.	Not specified	silver	(Alam et al., 2021)
P. aeruginosa strain DM1	A peptide-based siderophore pyoverdine	silver	(Kumari et al., 2017)
P. aeruginosa strain BS-161R	Rhamnolipids	silver	(Ganesh et al., 2010; Kumar & Mamidyala, 2011)

P. aeruginosa strain MTCC 2297	Rhamnolipids	ZnS	(Narayanan et al., 2010)
P. aeruginosa strain BS01	Rhamnolipids	ZnS	(Hazra et al., 2013)
P. aeruginosa strain OBP1	Rhamnolipid	silver	(Saikia et al., 2013)
B. subtilis	Lipopeptide	silver	(Yu et al., 2021)
	Biosurfactant	silver	(Mendrek et al., 2017)
	Biosurfactants surfactin and iturin	silver	(Płaza et al., 2016)
S. ghanaensis strain VITHM1	4-phenylbutanal, *N*-(2-(1*H*indol-3-yl)ethyl)-3-chloropropanamide and 3-benzylhexahydropyrrolo[1,2-a]pyrazine-1,4-dione	silver	(Abirami & Kannabiran, 2016)
C. firmus	Not specified	silver	(Sudarsan et al., 2021)
S. griseoplanus strain SAI-25	Amines, carboxylic acids and aromatic phenols/alcohols	silver	(Vijayabharathi et al., 2018)
P. aeruginosa	A peptide-based siderophore pyoverdine and a blue-green pigment pyocyanin	gold and silver	(Patil et al., 2022)
B. subtilis strain BBK006	Lipopeptide surfactin	gold and silver	(Reddy et al., 2009)
B. casei strain MSA19	Glycolipid	silver	(Kiran et al., 2010)

However, the toxic properties of metal nanoparticles are known. Today the dependence of the toxicity of nanoparticles on their physicochemical properties (chemical composition, size, shape, surface chemistry) is discussed (Zhang et al., 2022a).

Therefore, the issue of MIC processes prevention using metal nanoparticles synthesized by microorganisms in combination with secondary microbial metabolites is still largely unexplored. Although similar researches have been performed with plants (Royani et al., 2023).

Thus, this problem in the study of MIC looks attractive, taking into account the information about the antibacterial, antibiofilm, anticorrosive properties of both groups of compounds. It is necessary to focus on research and toxic properties of these compounds, their impact on the ecosystem where construction materials are exploited.

4. Progress, Challenges and Future Direction for This Work

To provide safety, environmental friendliness and health when solving the issue of MIC, “green” inhibitors/biocides have significant prospects. Among the secondary metabolites of microorganisms, biosurfactants and exopolysaccharide complexes (namely EPS) are the most studied “green” inhibitors/biocides of MIC. At the same time, the number of studies on the antimicrobial/antibiofilm and anticorrosive effects of microbial siderophores – environmentally safe metal chelators – is increasing.

Today, in the issue of microbial-induced corrosion, the issue of biodiversity of microorganisms producing secondary metabolites with antimicrobial, anti-corrosion, anti-biofilm properties for protection against MIC remains insufficiently studied. Also, the attention of researchers should be focused on the study of the chemical nature of bacterial secondary metabolites with the above properties for protection against MIC, the possibilities of practical application of secondary metabolites of microorganisms to prevent MIC, considering the disadvantages and advantages of this approach. Considering the effectiveness of the use of “green” inhibitors only under the laboratory experiments, and not in field conditions with complex ecological factors and organisms’ interactions (Little et al., 2020), perhaps efforts should be targeted on researching their use at cultural heritage sites. Research on the antimicrobial, anticorrosive, and antibiofilm properties of metal nanoparticles synthesized by microorganisms in combination with secondary microbial metabolites for protection against MIC should be initiated. At the same time, attention should be paid to the possible toxicity of such complexes and their possible negative impact on ecosystems. In addition, further researches should be related to the analysis of secondary metabolic profiles in MIC sites and the assessment of the ability of these compounds to influence the mechanisms of functioning of microbial corrosively active groups.

5. Conclusions

Various “green” inhibitors/biocides have their own advantages and disadvantages but all of them should fulfill principles of “green” chemistry and been characterized according to their activity, toxicity, low-cost, environmental friendliness, decomposition rate and total effectiveness. Secondary metabolite of plants, fungi and microorganisms are extensively studied as the perspective “green” biocides both as only component and in combination with other compounds for solving wide spectrum of issues, in particular mitigation and prevention of MIC. Secondary metabolites of microorganisms are studied in MIC processes with the practical goal of using them as “green” inhibitors/biocides to protect against biodamage/biofouling of materials. Microbial secondary metabolites are environmentally safe compounds and are promising for the use in MIC sites: lipopeptides, exopolysaccharides (EPS), siderophores, (Fig. 2). At the same time, one should be careful with the use of antibiotics and complexes of microbial secondary metabolites with biosynthesized nanoparticles to prevent MIC, since such compounds can negatively affect the microbial community, changing its structure (metal nanoparticles) and/or forming an antibiotic-resistant microbiome (antibiotics). Research in this area should be concentrated on further evaluation and analysis of antimicrobial, antibiofilm and anticorrosion activity of biosurfactants, siderophores and metal nanoparticles, in particular, their complex action.

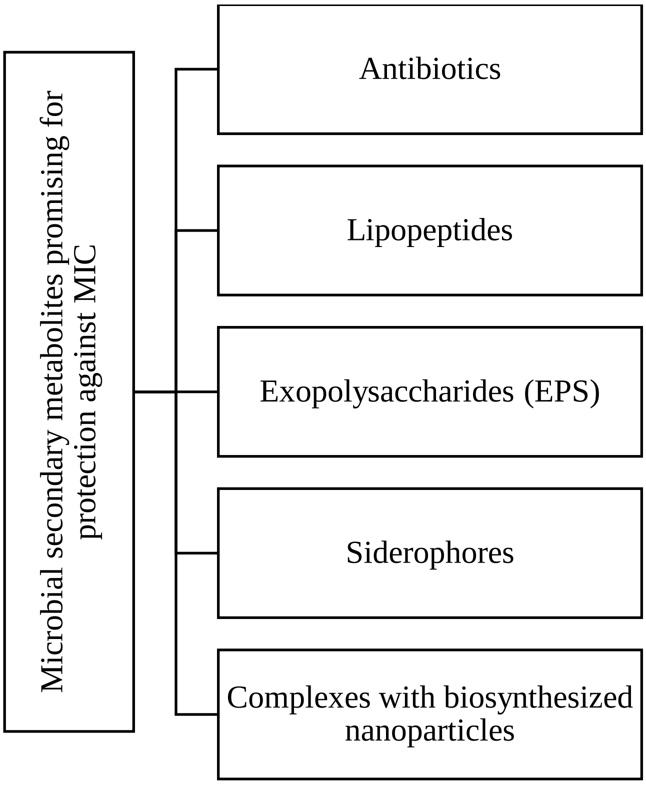

Figure 2. Microbial secondary metabolites promising for protection against MIC

At the same time, secondary metabolites are compounds that can affect the functioning of microbial groups, in particular, corrosive ones, which is a complicated open question. Totally, the role of microbial secondary metabolites in MIC is summarized in Figure 3 as interrelated functions and they could be considered for selection of compounds as environmentally friendly approach against MIC.

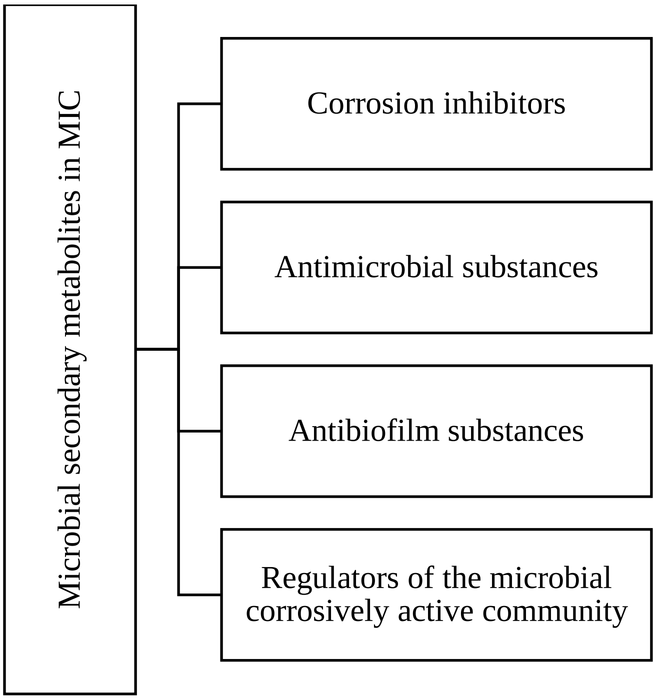

Figure 3. The value of microbial secondary metabolites for inhibition of MIC processes

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