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Antimicrobial concrete for smart and durable infrastructures: a review

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42 **1. Introduction**

43 Concrete is the most widely used construction material for various infrastructures
44 worldwide. However, concrete structures in certain aggressive environments, such as
45 sewer systems, marine engineering, buildings exposed to high humidity and the like,
46 are easily suffered from microbial attachment, colonization, eventually, deterioration
47 [1-4]. For example, the most typical problem faced by reinforced concrete structures in
48 sewer systems is microbial induced corrosion, which is still commonly referred to as a
49 sulfide (H_2S) gas problem. The process initiated when sulfate-reducing bacteria (SRB)
50 convert sulfate into hydrogen sulfide gas under anaerobic conditions, that is converted
51 into corrosive sulfuric acid by sulfur-oxidizing bacteria (SOB) of the genus
52 *Thiobacillus* [1,5-11]. Some fungi also participate in this activity [12,13]. Concrete
53 structures in the tidal and splash zones of marine concrete engineering are dominantly
54 damaged by *Pseudoalteromonas*, along with *Vibrio*, *Pseudomonas* and *Arthrobacters*,
55 etc. [14, 15]. Bio-deterioration of concrete in irrigation and hydroelectric canals [16],
56 spots or patches covered on concrete walls [17], and biological decay of mortars on

57 building facades [18] commonly result from the growth of algae and cyanobacteria.
58 Algal growth is also quite common on concrete walls of water storage and conveyance
59 structures [19]. Salmonella, an important foodborne pathogen, are easily attached and
60 colonized on surfaces of concrete used in food industry due to their adherence forming
61 biofilms [20]. The propagation and proliferation of microorganisms including bacteria
62 (e.g., pathogens), fungi, and algae alone or together, on and/or in concrete structures,
63 will affect concrete's aesthetic appearance, destroy the internal structure of concrete,
64 degrade mechanical properties and durability of concrete, increasing the cost by
65 rehabilitation and even replacement [2, 16, 21-23]. Therefore, developing antimicrobial
66 concrete for smart and durable infrastructures has become extremely significant and
67 imperative.

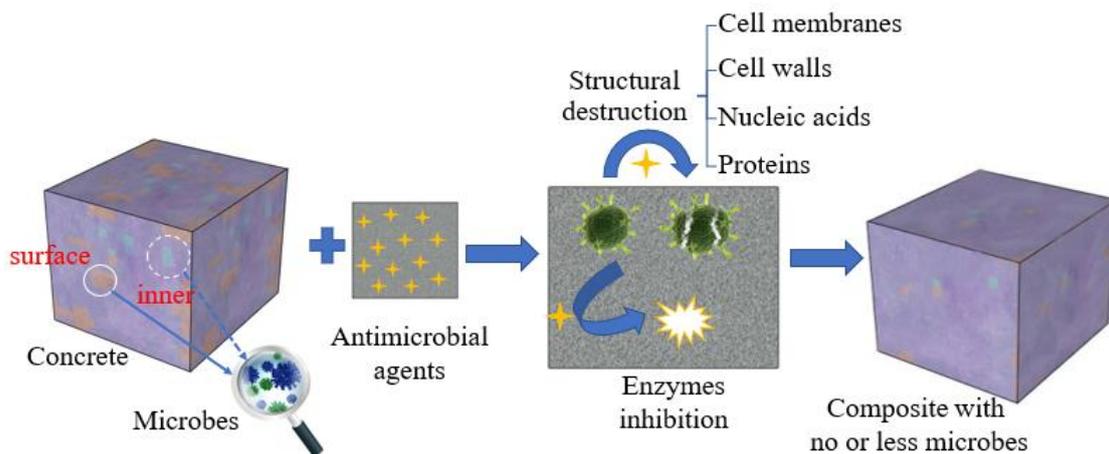
68 Researchers have been attempting to develop antimicrobial concrete (concrete is
69 a collective term referring to concrete, cement mortar and cement paste as well as
70 cementitious/cement-based materials/composites in this paper) by adding some
71 additives having antimicrobial properties for sterilization against a specific
72 microorganism or multiple microorganisms, meanwhile without significantly impairing
73 concrete essential properties such as compressive strength. The last two decades have
74 witnessed an ever-increasing growth in studies on the utilization of functionalized
75 zeolites supporting bactericidal metal ions, such as silver, copper and zinc ions [24-27].
76 Haile et al. [28-30] reported that concrete containing silver bearing zeolite exhibited
77 antimicrobial characteristics against *Acidithiobacillus thiooxidans* (A.thiooxidans), as
78 reflected by inhibition of formation of A.thiooxidans biofilm. Further, Xu [31] and Li
79 [32] reported that concrete added with silver-loading zeolite and polypropylene fiber
80 exhibited obvious bactericidal effect towards *Escherichia coli* (E. coli). Moreover, it is
81 reported that antimicrobial concrete containing Zeomighty (zeolites with silver and

82 copper ions) was introduced on the Japanese market [33]. Quaternary ammonium
83 compounds (Quats) have been used as antimicrobial agents for a long time, and only
84 recently they have been reported to be effective as algaecides [11, 16, 19, 34].
85 Intentionally, considering the severe consequences caused by microbial induced
86 corrosion of concrete, considerable attention has been paid to find effective
87 antimicrobial agents to admix into concrete in order to fight against Thiobacill [3, 23,
88 35, 36]. For instance, Shook and Bell [37] reported that ConShield, added into concrete
89 during the mixing stage, showed high sterilizing rate and stable bactericidal effect
90 against Thiobacillus bacteria. Yamanaka et al. [38] found that calcium formate was able
91 to completely inhibit the growth of both sulfur-oxidizing and acidophilic iron-oxidizing
92 bacteria at concentrations above 50 mM. Some investigators tried to develop
93 antimicrobial concrete by incorporation of nickel, and tungsten specially targeting at
94 SOB which play a dominant role in biogenic corrosion of sewer systems [39-43]. Sun
95 et al. [44] verified the strong bactericidal effect of free nitrous acid (FNA) on
96 microorganisms due to cells in corrosion biofilms of concrete surfaces were killed. In
97 addition, the combination of water repellents (decrease bio-receptivity) plus biocides
98 (decrease biological activity) has been reported to be effectively inhibiting microbial
99 growth in mortars, white concretes and autoclaved aerated concretes [45, 46]. Vaquero
100 et al. [16] proposed a novel cement-based material with biocidal activity that can be
101 used as an overlay of mortar in existing structures, such as canals and pipes.

102 In recent years, with the rapid development of nanotechnology, some researchers
103 have tried to introduce some nano-particles into concrete to inhibit microbial
104 colonization. For example, the research undertaken by Singh et al. [47] indicated that
105 cement-ZnO composite possesses effective antibacterial and antifungal activities under
106 dark and solar light due to the addition of ZnO nano powder. Wang et al. [48]

107 demonstrated that high performance concrete (HPC) incorporated with nano ZnO has
 108 antibacterial ability against *E. coli* and *Staphylococcus aureus* (*S. aureus*). Concrete
 109 fabricated with titanium dioxide nanoparticles has great potential in sterilization under
 110 the light [49]. Ganji et al. [50] found that cement with nano-TiO₂ inhibit the growth of
 111 *E. coli* under UV irradiation. Moreover, Fonseca et al. [18] proposed that anatase can
 112 be an alternative application for preventing bio-deterioration of mortars.

113 This paper is intended to summarize antimicrobial concrete fabricated with
 114 different types of antimicrobial agents, intuitively shown in Fig. 1. First, the
 115 classification of antimicrobial agents and their application methods into concrete are
 116 briefly introduced. Then, the antimicrobial and mechanical properties as well as
 117 mass/weight loss of concrete incorporated with antimicrobial agents are reviewed, with
 118 emphasis on antimicrobial properties. Subsequently, antimicrobial mechanisms of some
 119 inorganic and organic antimicrobial agents were explicated. Finally, applications of
 120 antimicrobial concrete in sewer systems, marine engineering and buildings against
 121 microbial threat are also presented.



122 Fig. 1 Schematic diagram of antimicrobial concrete.

123 2. Classification of antimicrobial agents used for fabricating 124 antimicrobial concrete

125 The antimicrobial property of antimicrobial concrete was attributed to the addition

126 of antimicrobial agent, which is a collective name herein for the mentioned
127 antimicrobial additives facilitating concrete to inhibit and/or kill various microbes
128 including bacteria (e.g., pathogens), fungi, and algae. Antimicrobial compounds
129 including biocides, microbicides, sanitizers, antiseptics and disinfectants characterized
130 by their ability of killing microorganisms and/or inhibiting microbial reproduction, are
131 easily accessible [23,34]. The antimicrobial agents reported to have been added to
132 concrete ingredients can be classified into inorganic and organic antimicrobial agents
133 with respect to their chemical composition as detailed below.

134 2.1 Inorganic antimicrobial agents

135 Inorganic antimicrobial agents that have been reported to be applied in concrete
136 include heavy metals (silver, nickel, tungsten), metal compounds (silver molybdate,
137 copper oxide, zinc oxide, sodium tungstate, sodium bromide), NORGANIX (a silicate
138 concrete sealer), free nitrous acid (FNA), and nano inorganic antimicrobial materials.
139 The antibacterial activity of metal or metal ions is in the order of:
140 $Ag > Hg > Cu > Cd > Cr > Ni > Pb > Co > Zn > Fe$ [22,32,51,52]. Although silver ion
141 antibacterial agent series are effective but considering their high cost, few other
142 alternatives with high bactericidal effect were explored in the literature. For example,
143 Zhang [22] found that cerium nitrate exhibited an excellent antibacterial effect in porous
144 concrete, even with a low content of 1.25%. Furthermore, the use of nanomaterials to
145 control microbial colonization of concrete has considerably increased in recent years
146 [53]. Nanoparticles (NPs) of Cu_2O , $CaCO_3$, TiO_2 , ZnO , CuO , Al_2O_3 , Fe_3O_4 , etc., were
147 reported to exhibit inhibitory effects against a wide range of microorganisms in this
148 field [3, 4, 26, 47,48, 54, 55].

149 2.2 Organic antimicrobial agents

150 Quats, phthalocyanine compound (including metal organic antimicrobial agent

151 copper phthalocyanine), calcium formate, alkyl nitro-bromide (A II B),
152 isothiazoline/cabamate, ConShield (a highly charged cationic polymer), and ConBlock
153 MIC (whose active ingredient is 3-Trimethoxy silyl propyl dimethyl octadecyl
154 ammonium chloride) are various organic antimicrobial agents used in concrete.
155 Additionally, Freed et al.[56] proposed that fibers incorporated with at least one
156 antimicrobial agent, such as Microban B (a phenol-based antimicrobial agent), were
157 able to inhibit microorganisms. Quats are the most representative organic
158 antimicrobials, e.g., silane quaternary ammonium chloride(SQA)[57], and cetyl-
159 methyl-ammonium bromide [19], which have been widely studied and applied by
160 researchers [23, 51, 58]. Isothiazoline/cabamate is a type of organic antifungal agents,
161 often used to target at *Aspergillus niger* which is easily found in the interiors and
162 exteriors of buildings in damp environment [59]. Uchida et al. [11] stated that water
163 pollution as a result of metal eluted into sewage can be addressed by adding a
164 phthalocyanine compound (a metal phthalocyanine, a metal-free phthalocyanine, and
165 derivatives thereof) into concrete, which will not pollute water and a small amount of
166 inhibitor can prevent deterioration of concrete or mortar due to SOB for a long time.

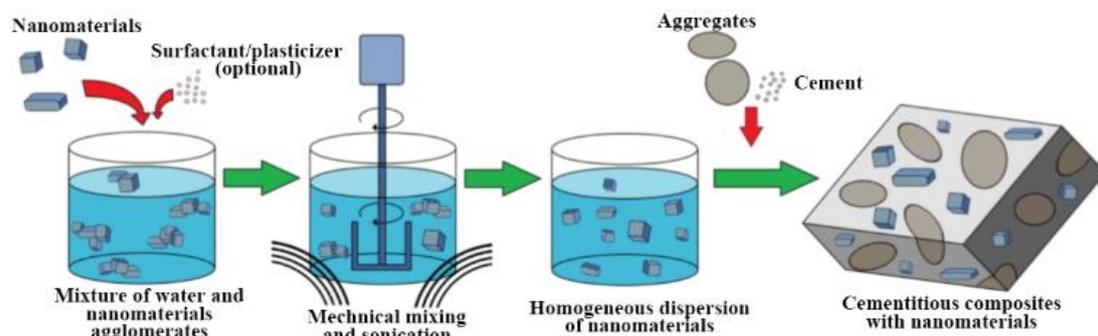
167 Generally, inorganic antimicrobial agents have long service life and high
168 temperature resistance, but have side effects like toxicity. Organic antimicrobial agents
169 possess obvious bactericidal effect in a short term and broad spectrum of killing activity
170 but their temperature resistance is poor [22,31,32,60]. Moreover, most of organic
171 biocides are ultimately ineffective at removing microbes and may eventually lead to a
172 new wave of microbes on the affected surfaces after microbes develop a resistance [34].
173 The following sections will describe these antimicrobial agents and their methods of
174 applications in detail.

175 **3. Methods of applying antimicrobial agents into concrete**

176 Some antimicrobial agents use inorganic or organic cementitious materials as
177 carriers to form protective coatings, with biocidal property on concrete surfaces [23,35].
178 Another method to apply antimicrobial agents into concrete is directly incorporating
179 antimicrobials into concrete mix as functional components after pre-dispersion [23,35].
180 For example, calcium formate was added in the mixture[38], ConShield was
181 incorporated into the mix and the protection was throughout the entire thickness of
182 concrete matrix [37]. The antimicrobial watertight admixture made of fluosilicate salts
183 and antimicrobial compounds (Ni and W) [61] is in liquid state to be homogeneously
184 dispersed in concrete. The phthalocyanine compound [11] can be dispersed uniformly
185 in concrete or mortar by a blending agent selected from a group consisting of an air
186 entraining agent, a water reducing agent, and a viscosity increasing agent. Liquid
187 bactericides like dimethyl benzyl ammonium chloride can be made into powder
188 adsorbed by carrier such as zeolite [23, 62]. In addition, heavy metal antibacterial
189 agents are usually fixed on zeolites by means of adsorption or ion exchange [27,51,63].
190 Known as crystalline pozzolanic aluminosilicate minerals with uniform molecular sized
191 pores, zeolites can be functionalized to exhibit antimicrobial property if calcium and
192 sodium ions in their framework are exchanged by silver, copper or zinc ions, explaining
193 that zeolites are the most common carriers of inorganic metal ions [3, 26, 27, 29, 51,
194 63, 64].

195 Agglomeration due to high activity of antimicrobial nanoparticles in cement
196 matrix is a common concern, significantly decreasing their chemical and physical
197 activities and, hence, affecting their efficiency in cement matrix performance and
198 antimicrobial activity [49, 60]. A dispersion medium (most likely mixing water) and
199 incorporation of organic admixtures and different surfactant types, e.g., plasticizers and

200 superplasticizers, facilitate to address the issue of homogeneous dispersion in the
201 cement matrix, as presented in Fig. 2 [49, 54]. It is also reported that the application of
202 superplasticizer in photocatalytic cement can enhance nano-TiO₂ dispersion in samples
203 by preventing agglomeration of titanium dioxide in cement pastes, which is also
204 conducive to improve the contact between titanium dioxide and bacteria, contributing
205 to better bacterial inactivation [50]. However, in the case of antimicrobial agents being
206 functional components in concrete, the selection of biocide types and contents has not
207 been systematically investigated [35, 65].



208 **Fig. 2** Schematic process of nanomaterials dispersing method commonly used in
209 cement-based composites preparation [54].
210

211 4. Properties of antimicrobial concrete

212 4.1 Antimicrobial property

213 4.1.1 Antimicrobial concrete with inorganic antimicrobial agents

214 The antimicrobial property is the most important assessment factor for
215 antimicrobial concrete, that varies with the addition of different types of antimicrobial
216 agents, as summarized in Table 1. Antimicrobial concrete, with the addition of diverse
217 antimicrobial agents against microorganisms involving in microbial induced corrosion,
218 especially in sewer systems, have been extensively studied in the literature. Nickel and
219 tungsten have been known to protect concrete from microbial corrosion owing to their
220 antimicrobial effect towards causative bacteria, i.e., *Thiobacillus thiooxidans* (T.

221 thiooxidans). Negishi et al. [41] found that the cell growth of *A. thiooxidans*, including
222 strain NB 1-3 (isolated from corroded concrete in Fukuyama, Japan) was strongly
223 inhibited by 20 μM sodium tungstate, and completely inhibited by 50 μM sodium
224 tungstate. Similarly, Sugio et al. [42] reported that cell growth of an iron-oxidizing
225 bacterium, *Acidithiobacillus ferrooxidans* (*A. ferrooxidans*), was strongly inhibited by
226 0.05 mM and completely inhibited by 0.2 mM of sodium tungstate. In the study of
227 Maeda et al. [40], concrete containing 0.1% metal nickel and concrete with 5 mM nickel
228 sulfate were found to completely inhibit the cell growth of strain NB 1-3 of *T.*
229 *thiooxidans* isolated from corroded concrete. Moreover, Kim et al. [61] conducted an
230 investigation to evaluate the antibiosis of antimicrobial ingredients (Ni and W) of
231 antimicrobial watertight admixture mixed in mortar and concrete on *Thiobacillus*
232 *novellus* (*T. novellus*). Broth Microdilution MIC test indicated that *T. novellus* could
233 not survive in the area where the admixture is dropped. As reflected in Table 1, the total
234 colony test numerically shows that *T. novellus* in culture solution with mortar added
235 with the admixture were disappeared after 24 h. The biochemical corrosion simulation
236 test also indicated that the number of *T. novellus* was much lower in the case of mortar
237 mixed with the admixture than plain mortar specimens. The results suggested that the
238 addition of antimicrobial watertight admixture in cement mortar and concrete
239 suppressed the growth of *T. novellus*. Furthermore, Southerland et al. [66] found that
240 tungsten used alone is able to inhibit growth of *T. novellus*, whereas molybdenum,
241 ammonium molybdate or a mixture of ammonium molybdate and tungstate activates
242 growth of the same bacteria. Likewise, it is reported that molybdenum activates growth
243 of *T. novellus* but inhibits growth of *T. thiooxidans*, indicating SOB of the same genus
244 *Thiobacillus* have different growth inhibitory mechanism. It is noteworthy that the
245 antimicrobial property of antimicrobial agent Ni and W is not only largely dependent

246 on their contents, but greatly affected by pH. It is generally recognized that nickel
247 compounds are suitable for neutral environment, while tungsten compounds are more
248 effective in acidic environment [23, 43]. Maeda et al. [40] observed that the amount of
249 nickel contained in the strain NB 1-3 cells treated without nickel, treated with 10 mM
250 nickel sulfate at pH 3.0 and treated with 10 mM nickel sulfate at pH 7.0 was 1.7, 35
251 and 160 nmol nickel per mg protein, respectively. The results indicated that nickel is
252 able to bind to strain NB 1-3 cells, and much more nickel binds to the cells at neutral
253 pH than at acidic pH demonstrated that nickel ions have a better inhibitory effect
254 towards the microbe in neutral environment than in acid environment [40]. The findings
255 of Negishi et al. [41] and Sugio et al. [42], as detailed in Table 1, demonstrated that the
256 antimicrobial property of tungsten is more effective in acidic environment than in
257 neutral environment.

258 Furthermore, Kong et al. [62, 65] conducted an investigation to evaluate the impact
259 of adding five bactericides in concrete towards the selected bacteria (as listed in Table
260 1), and to study their applicability for controlling and preventing microbial corrosion
261 of concrete. They reported that concrete with sodium bromide and zinc oxide exhibited
262 excellent antimicrobial property towards the tested bacteria, especially Bacteroidetes,
263 as the number of microbial populations decreased substantially. However, the
264 antimicrobial effect of concrete with a dispersion of sodium tungstate on microbes is
265 worst, as reflected by the lowest bactericidal rate (21.95%), it even promotes the growth
266 and reproduction of Proteobacteria. They also observed the dead and live
267 microorganisms within biofilm with confocal scanning laser microscopy (CLSM), as
268 seen in Fig. 3. The number of live cells within the biofilm all decreased to a certain
269 degree, indicating all the tested bactericides have a certain sterilizing effect. Similarly,
270 Bao [67] obtained that the surface roughness of the control mortars and mortars with

271 sodium tungstate and sodium bromide was 46.65, 14.3 and 9.02 μm after a 3-month
 272 immersion in intensified sewage, respectively. Therefore, they concluded that the
 273 addition of sodium tungstate and sodium bromide could effectively inhibit the growth
 274 and reproduction of microorganisms attached to the surface of cement mortar. In
 275 addition, Sun et al. [44] studied the bactericidal effect of FNA on microbes in sewer
 276 biofilms of two concrete coupons. They observed that, as for the intact corrosion
 277 biofilm, H_2S uptake rates (SUR) were reduced markedly 15 days after FNA spray and
 278 viable bacterial cells severely decreased by over 80% within 39 h (detailed in Table 1),
 279 suggesting that biofilm cells were killed by the treatment. As for a suspended solution
 280 of corrosion biofilms scraped from the concrete coupon, ATP level and ratio of viable
 281 bacterial cells were also severely decreased by the treatment, as clearly seen in Fig. 4,
 282 demonstrating that FNA strongly deactivates bacteria of acidic corrosion biofilm [44].

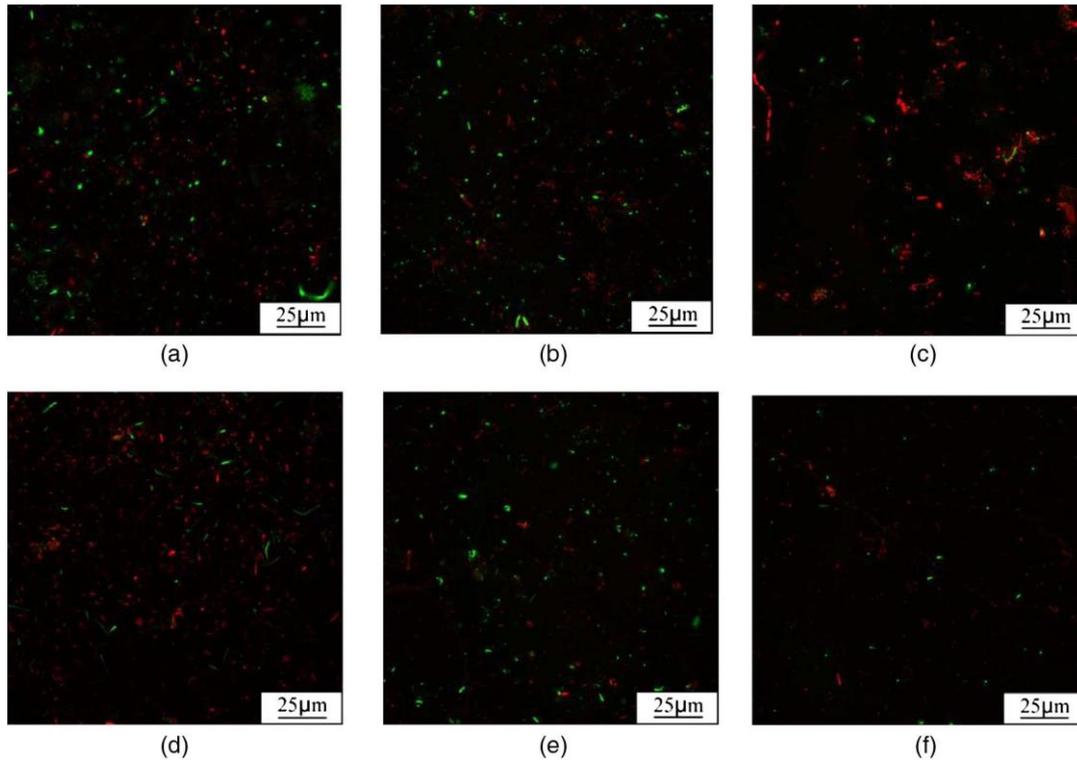
283 Table 1. Summary of different inorganic antimicrobials on antimicrobial property

Antimicrobial	Microorganism	Matrix	Findings
Sodium bromide, zinc oxide, sodium tungstate [65]	Bacteroidetes, Proteobacteria, Firmicutes and Actinomycetes	Concrete	High sterilizing rate of NaBr, ZnO towards Bacteroidetes was 86.80%, 79.19%, respectively Na ₂ WO ₄ showed the lowest bactericidal rate as 21.95% towards all bacteria
Silver-loaded zeolite [30]	A.thiooxidans	Concrete	Growth of planktonic and biofilm populations of A.thiooxidans was inhibited
Zinc and silver loading zeolite [29]	A. thiooxidans	Concrete	Functionalized zeolite coated concrete specimens with epoxy to zeolite weight ratios of 2:2 and 1:3 had negligible biomass growth and acid production rates
Silver/copper zeolite, silver/zinc zeolite [28]	A.thiooxidans	Mortar	Co-cations such as Zn ²⁺ and Cu ²⁺ increases antimicrobial activity of silver bearing zeolite

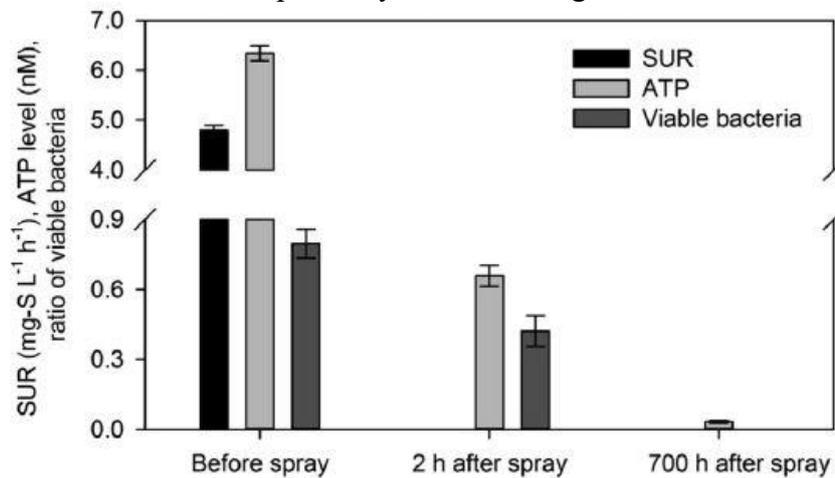
Nano-copper oxide [26]	A.thiooxidans	Concrete	Higher leaching rate of copper from loosely adhered nano-copper oxide film significantly inhibited the activity of A.thiooxidans
Silver copper zeolites [25]	E. coli, Listeria monocytogenes, Salmonella enterica or S. aureus	Mortar	Centration of silver copper zeolites to obtain a bactericidal effect on mortar surfaces is required more than 3%
Zeomighty [33]	Thiobacilli	N.A.	A concentration of metal zeolites of 1% to cement weight is optimum for suppressing the growth of Thiobacilli
Sodium tungstate [41]	A. thiooxidans	N.A.	Approximately 10 times more tungstate bound to the cells of A. thiooxidans at pH 3.0 than at pH 7.0
Sodium tungstate [42]	A. ferroxidans	N.A.	Approximately 2 times more tungsten bound the cells of A. ferroxidans at pH 3.0 than at pH 6.0
Metal (Ni,W) compounds, ZnSiF ₆ [61]	T.novellus	Mortar, concrete	Mortar with antimicrobial watertight admixture had higher pH(6.8) and lower concentration of sulfuric acid(3.78×10^{-8} mol/L) compared to that (6.6 and 2.56×10^{-7} mol/L) of plain mortar
Zinc oxide, sodium bromide, copper slag, ammonium chloride, cetyl-methyl-ammonium bromide [19]	Algae	Mortar	Adding 20 wt.% zinc oxide and 20 wt.% sodium bromide exhibited the most effective algal inhibition under laboratory condition The addition of 20 wt.% sodium bromide and 10 wt.% cetyl-methyl-ammonium bromide (an organic antimicrobial agent) showed highest inhibitory effects at under field condition
FNA [44]	N.A.	Concrete	H ₂ S uptake rate decreased by 84-92% 1-2 months and viable bacterial cells reduced from $84.6 \pm 8.3\%$ to $10.7 \pm 4.3\%$ within 39 h after FNA spray.

Silver molybdate [52]	E. coli and S. aureus	Concrete	The residual colony count of E. coli and S. aureus is 0 cfu/mL by addition of 0.004% silver molybdate
Cerium nitrate [22]	E. coli	Concrete	Bacterial concentration reduced drastically from 7.50 to 0.01,0,0.02 million per ml after 48 h when the content was 1.25,5.00,10.00%, respectively.
Nano sized TiO ₂ , CaCO ₃ [4]	Pseudomonas, Fusarium, algae, blue-green algae and manganese oxidizing bacteria	Mortar	Nano-TiO ₂ modified fly ash mortar and nano-sized TiO ₂ , CaCO ₃ modified fly ash mortar exhibited enhanced antibacterial activities compared to nano-CaCO ₃ modified fly ash mortar
Anatase [18]	Cyanobacteria and chlorophyta species	Mortar	Two types of mortars with different kinds of sand showed the lowest photosynthetic growth ratio (0% and 0.03%, respectively)
SiO ₂ /TiO ₂ nano-composite [68]	E. coli	Cement mortar	Bacteria inactivation after UV light irradiation and without illumination after 120 min was 67% and 42%, respectively.

284 Note: A. thiooxidans: Acidithiobacillus thiooxidans; T. thiooxidans: Thiobacillus
285 thiooxidans; T. novellus: Thiobacillus novellus; A. ferroxidans: Acidithiobacillus
286 ferroxidans; E. coli: Escherichia coli; S. aureus: Staphylococcus aureus; N.A.: not
287 available



288 Fig. 3 CLSM images of the distribution of dead/live cells within biofilm attached to
 289 concretes: (a) plain concrete without bactericide; (b) concrete with dodecyl dimethyl
 290 benzyl ammonium chloride; (c) concrete with sodium bromide; (d) concrete with zinc
 291 oxide; (e) concrete with sodium tungstate; and (f) concrete with copper
 292 phthalocyanine [62]. Note: living and dead cells are displayed in green and red,
 293 respectively, under blue light.



294 Fig. 4 Levels of SUR, ATP and the ratio of viable bacteria measured on reactor
 295 solutions containing the suspended corrosion biofilm scraped from a concrete coupon
 296 after 40 months of exposure prior to and after FNA treatment. The ratio of viable
 297 bacteria was not determined after 700 h of FNA treatment as cells could not be
 298 extracted from the reactor solution [44]. Note: SUR means H₂S uptake rate.
 299 Zeolite containing metal ions has been investigated a lot to be used in concrete due

300 to its excellent antimicrobial property. For example, Haile et al. [28] evaluated the

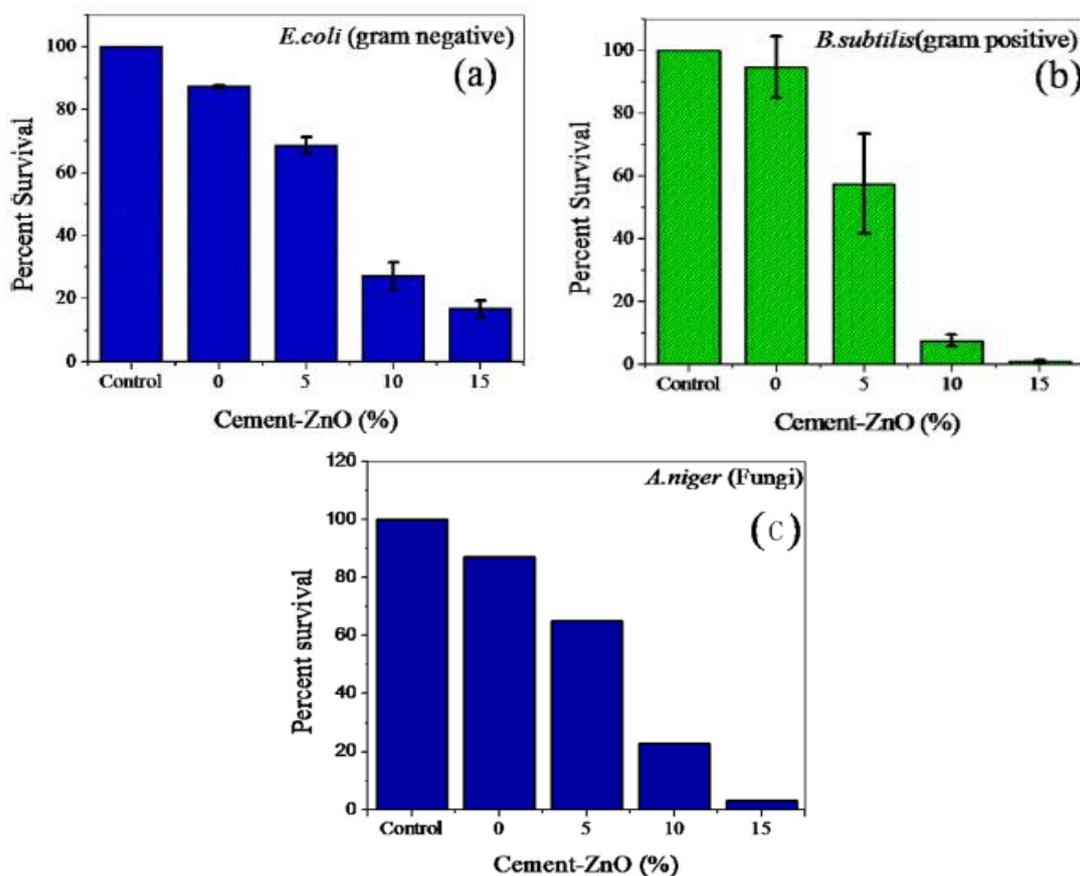
301 antimicrobial characteristics of mortar specimens coated with silver bearing zeolite
302 with *A. thiooxidans*. They observed that biomass concentration of *A. thiooxidans* dry
303 cell weight (DCW) of control specimens (236 mg TSS/L and 181 mg TSS/L) was as
304 much as 2-fold higher compared to the mortars coated with silver-loaded zeolite (125
305 mg TSS/L and 80 mg TSS/L). The reduced microbial numbers evidenced that the mortar
306 specimens coated silver bearing zeolite exerted antimicrobial characteristic on
307 *A. thiooxidans* and inhibited bacterial growth. They also found that bacteria were not
308 affected in the nutrient solution indicated that the antimicrobial characteristics of
309 zeolitic coatings were only apparent on solid surface particles [28]. Moreover, Haile et
310 al. [30] discovered that no biomass growth was observed upon exposure of the
311 bacterium to silver-loaded zeolite coated concrete specimens, and there was no oxygen
312 uptake measured, meaning no viability of *A. thiooxidans* cells for the silver-loaded
313 zeolite coated concrete specimens. The research results confirmed that zeolite
314 containing 5 wt.% Ag is inhibitory to planktonic and biofilm of *A. thiooxidans* [30].
315 Similarly, De Muynck et al. [69] observed that mortar specimens with silver-copper
316 zeolites (zeolites contain 3.5% silver and 6.5% copper) obtained a 12-fold decrease of
317 ATP content after 24 h, while inhibition of antimicrobial fibers on bactericidal activity
318 was limited, indicating biocidal effect towards SOB was limited in the case of
319 antimicrobial fibers and that of antimicrobial zeolites was much better. Moreover, De
320 Muynck et al. [25] investigated the antimicrobial effectiveness of silver copper zeolites
321 against *E. coli*, *Listeria monocytogenes*, *Salmonella enterica* or *S. aureus* in a
322 quantitative way. A clear decrease in the total ATP content was observed for mortar
323 specimens containing silver copper zeolites, indicating the occurrence of antimicrobial
324 activity by the presence of silver and copper ions. Furthermore, they concluded that the
325 concentration of silver copper zeolites is required to be more than 3% so as to obtain a

326 bactericidal effect on mortar surfaces [25]. In the experiment of Haile et al. [70], cellular
327 ATP in concrete contained 2.6 wt.% silver-loaded chabasite declined to zero with a
328 corresponding DCW value of 35 mg, indicating there was no growth after bacteria were
329 exposed to 2.6 wt.% silver-loaded chabasite, whereas the biomass was 51 mg DCW and
330 cellular ATP was 0.21 mg for concrete coated 18 wt.% silver-loaded chabasite. The
331 results indicated that antibacterial characteristics of concrete specimens coated with 2.6
332 wt.% is superior to the specimens with 18 wt.% silver-loaded chabasite. The results of
333 the experiment conducted by Xu and Meng [64] indicated that the content of *E. coli* in
334 concrete incorporating silver-bearing zeolite and polypropylene fiber was reduced
335 compared to the control samples, demonstrating that silver-bearing zeolite and
336 polypropylene fiber play a bactericidal role and reduce the breeding of *E. coli*. Likewise,
337 Li [32] discovered that concrete specimens added with 0.5% silver-loading zeolite and
338 polypropylene fiber had the most pronounced bactericidal effect towards *E. coli*, as
339 reflected by the greatest OD value (the greater the OD value, the lower the bacterial
340 concentration of the concrete samples) according to antibacterial test results. While
341 antimicrobial effect of concrete specimens incorporated with fly ash and mineral
342 powder was not evident.

343 Researchers have paid much attention to the effect of antimicrobial nanoparticles
344 on antimicrobial property of concrete. Singh et al. [47] admixed ZnO nano powder into
345 cement composite and evaluated the antimicrobial effect of the formed cement-ZnO
346 composites against two bacterial strains *E. coli*, *Bacillus subtilis* and fungal strain
347 *Aspergillus niger*. As shown in Fig. 5, the antibacterial and antifungal effects of cement-
348 ZnO composite increased as the ZnO concentration increased in the range of 0,5, 10,
349 15 wt.%. Moreover, it was also noted that both antibacterial and antifungal activities of
350 cement-ZnO composite was enhanced under sunlight compared to dark condition. In

351 addition, Wang et al. [48] conducted a research to study the antimicrobial effect of high-
352 performance concrete (HPC) added with nano ZnO against *E. coli* and *S. aureus*. The
353 results showed that the antibacterial rate of the two groups of antibacterial concrete
354 against *E. coli* reached 100%, however the antibacterial rate against *S. aureus* was 54.61%
355 and 99.12%, respectively. Through SEM observations, it is found that nano ZnO and
356 its resulting compounds precipitation adhered to surface of cement hydrate, thus
357 inhibited the growth of bacteria, accounting for the significant antibacterial effect of
358 HPC [48]. Sikora et al. [54] conducted a series of tests to evaluate the antimicrobial
359 effect of four metal oxide nanoparticles (Al_2O_3 , CuO, Fe_3O_4 , ZnO) used in cement-
360 based composites. They discovered that all the studied nanoparticles inhibited microbial
361 growth, and the growth kinetics showed that the highest inhibitory effect on *E. coli*
362 ATCC[®] 8739[™] and *E. coli* MG 1655 was Fe_3O_4 nanoparticles, ZnO nanoparticles,
363 respectively. The biofilm formation assay indicated that the tested nanoparticles were
364 able to reduce the formation of bacterial biofilms, *E. coli* ATCC[®] 8739[™] biofilms
365 were inhibited by all nano-oxides, ZnO nanoparticles significantly affected the
366 formation of *P. aeruginosa* and *S. aureus* biofilms. However, the viability of *P.*
367 *aeruginosa* cells in sample with Al_2O_3 was significantly higher compared to the control
368 sample. Similarly, Dyshlyuk et al. [71] evaluated antibacterial and fungicidal properties
369 of ZnO, TiO_2 and SiO_2 nanoparticle solution by interaction with eight types of
370 microorganisms commonly causing bio-damage to buildings and concrete structures.
371 They found that ZnO nanoparticles of 2-7 nm in size with a suspension concentration
372 of 0.01-0.25% displayed the most noticeable antimicrobial properties against the tested
373 strains, decreasing microorganisms by 2-3 orders of magnitude. They also revealed that
374 ZnO nanoparticles interacted specifically to a microorganism type, leading to a
375 decrease in the number of *Bacillus subtilis* B 1448 bacterium by 2 orders of magnitude,

376 and that of fungi of *Penicillium ochrochloron* F 920 by 3 orders of magnitude. However,
377 TiO_2 and SiO_2 nanoparticles exhibited a low antimicrobial activity. Nano- TiO_2 , with its
378 excellent photocatalytic effect, has aroused much interest of many researchers in the
379 aspect of microorganism inactivation. For instance, Ganji et al. [50] investigated the
380 antimicrobial performance of cement samples containing 1,5 and 10 wt.% nano- TiO_2
381 against *E. coli* under UV irradiation. They found that bacterial inactivity enhanced as
382 the amount of TiO_2 nanoparticles in cement samples increased, however, the
383 inactivation effect was not obvious even the amount of TiO_2 nanoparticles further
384 increase to 10 wt.%. Therefore, 5 wt.% TiO_2 is proposed to be the most proper content
385 in cement samples for inactivation of *E. coli* taking into account both the photocatalytic
386 inactivation and cost. Linkous et al. [72] employed nano- TiO_2 in concrete to inhibit the
387 attachment and growth of oedogonium. They discovered that concrete containing 10
388 wt.% TiO_2 nanoparticles obtained a 66% reduction in the growth of oedogonium.



389 Fig. 5 Effect of different concentrations of cement-ZnO composite on various
390 microorganisms [47]: (a) *E. coli*, (b) *Bacillus subtilis* and (c) *Aspergillus niger*.
391

392 Besides above, researchers have also investigated antimicrobial effects of
393 antimicrobial concrete towards some other commonly microbes threatening concrete.
394 For example, Umar et al. [36] evaluated the antimicrobial activity of four types of semi-
395 circular modified cement composite specimens using *Serratia marcescens* collected
396 from seashore and then isolated from microbe samples. The results showed that cement
397 composites admixed with sodium nitrite-based inhibitor performed better with the least
398 percentage increment of total viable count at the end of 144 h as compared to the cement
399 composite with styrene acrylate copolymer, with acrylic polymer, and cement
400 composite without any admixture, respectively. This can infer that cement composite
401 with sodium nitrite-based inhibitor exhibited noticeably improved ability to suppress
402 the growth of *Serratia marcescens* in marine environment. NORGANIX [73] is able to
403 endue concrete with powerful antimicrobial property to eliminate *Salmonella*, *Listeria*,
404 *E. coli*, *Clostridium*, and mold spores not just on the surface but deep within the
405 concrete. Moreover, antimicrobial concrete with NORGANIX can prevent microbes
406 from re-entering concrete from any directions because NORGANIX will hydrate with
407 the unused Portland cement within the concrete to generate new cement, thereby sealing
408 the capillary system. Paiva et al. [20] determined the antimicrobial efficiency of
409 BioSealed for Concrete™, a hydro-silicate catalyst in a colloidal liquid base, to prevent
410 *Salmonella* spp. attached on concrete bricks in food industry. They found that concrete
411 bricks treated with BioSealed for Concrete™ after inoculation, before and after

412 inoculation had immediate bactericidal effects towards the tested five strains of
413 Salmonella in contrast with bricks not treated with BioSealed for Concrete™ and bricks
414 treated with BioSealed for Concrete™ before inoculation, as observed by significantly
415 lower viable counts of Salmonella.

416 4.1.2 Antimicrobial concrete with organic antimicrobial agents

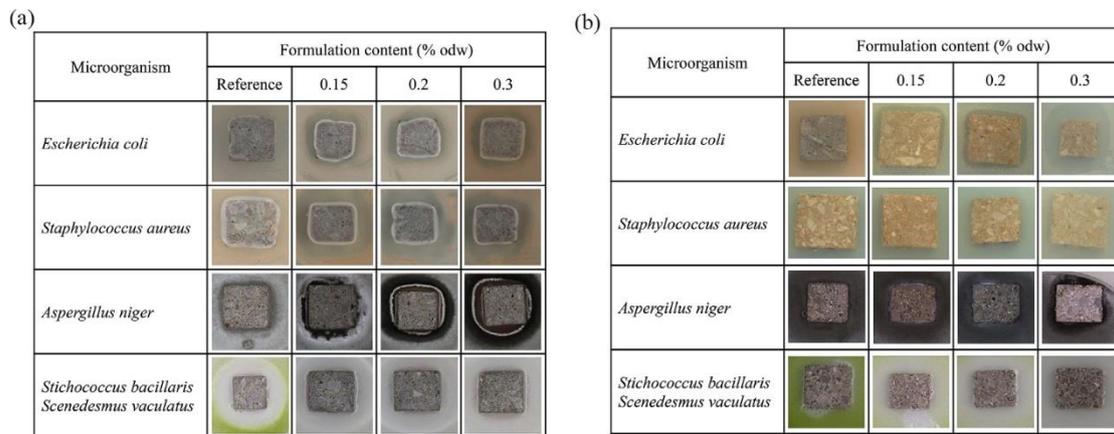
417 Yamanaka et al. [38] studied the inhibitory effects of formate on the growth of
418 bacteria causing concrete corrosion in sewerage systems. They found that the growth
419 of SOB isolated from corroded concrete were completely inhibited by 10 mM calcium
420 formate for 18 days, while the growth of acidophilic iron-oxidizing bacteria was
421 inhibited by 10 mM calcium formate during 34 days. This finding shows that even the
422 same antimicrobial agent has different inhibitory effect on different microbes. In
423 addition, they also observed that the formation of ATP in bacterial cells was ceased after
424 the addition of calcium formate into concrete test pieces. Erbehtas et al. [57] evaluated
425 the antimicrobial efficacy of silane quaternary ammonium chloride (SQA) aqueous salt
426 solution against planktonic *Halothiobacillus Neapolitanus* and *A.thiooxidans*. They
427 found that the antimicrobial efficacy directly related to bacterial population and activity,
428 and indirectly depends on pH. Furthermore, antimicrobial effectiveness occurs when
429 the pH is greater than 4. In the research undertaken by Do et al. [59], cement mortars
430 with isothiazoline/cabamate exhibited a good antifungal effect against *Aspergillus niger*,
431 while mortars with nitrofurantoin did not show inhibitory effect even the content of
432 nitrofurantoin was up to 5 wt.%. Moreover, the antifungal effect of cement mortar
433 containing isothiazoline/cabamate on *Aspergillus niger* enhanced almost linearly as the
434 content increases (0%,0.3%,0.5%,1%,2% and 5% by mass to cement). According to
435 [74], researchers of former Soviet Union tested mortar samples with alkyl nitro-

436 bromide (A II B) stored for 6 years. The results indicated that the microbial retention
437 rate on the surface of mortar specimens was merely 0.6% and 0.1% when the content
438 of A II B is 0.025 wt.% and 0.05 wt.%, respectively, after 5 hours of irradiation,
439 confirming the strong and long-lasting antimicrobial ability of A II B.

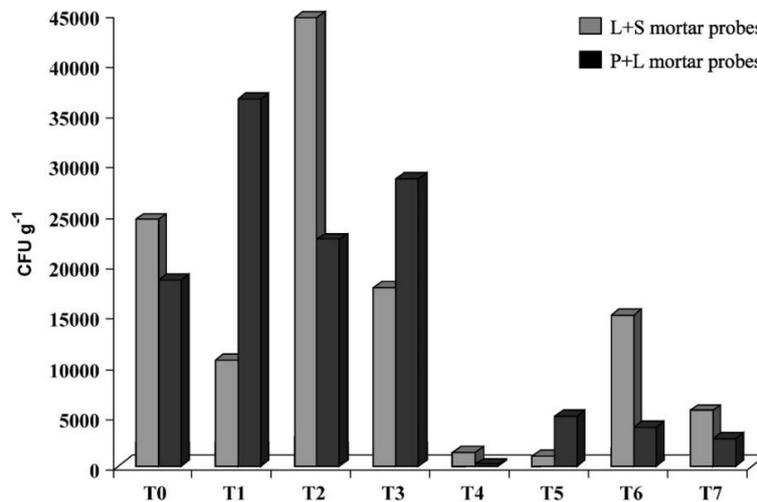
440 It is worthwhile noting that some organic antimicrobial agents are extremely
441 suitable to add into concrete due to their antimicrobial power to combat against diverse
442 microbes, rather than only a single type of microbe. For example, Kong et al. [62] found
443 that concrete added with copper phthalocyanine exhibited outstanding antimicrobial
444 effect with high bactericidal rates towards Bacteroidetes (90.82%) and Proteobacteria
445 (64.25%), and the bactericidal rate towards all tested microbes is as high as 82.59%.
446 The number of live cells within the biofilm attached to concrete added with copper
447 phthalocyanine showed a significant drop, and the content of live cells was only 12%
448 of that attached to plain concrete. A large number of dead microbes was observed, as
449 seen in Fig. 3 (f). Vaquero et al. [16] studied the bactericidal ability of 15 commercial
450 bactericides blended into concrete against microbial induced corrosion by culturing
451 microbes and evaluating the antimicrobial efficiency. Research results indicated that
452 the multicomponent formulation PL-UV-H-2 B was the sole formulation to succeed in
453 all the evaluation process among all formulations. Concrete samples fabricated with
454 PL-UV-H-2 B, of which the actives are 30% 2-octyl-2H-isothiazol-3-one + Terbutryn
455 and 15% 2,4,4'-trichloro-2'-hydroxy-diphenyl ether (calcium filler as a dispersive
456 matrix), exhibited high effectiveness in antimicrobial tests against algae (*Scenedesmus*
457 *vaculatus* and *Stichococcus bacillaris*), fungus (*Aspergillus niger*), and bacteria (*S.*
458 *aureus* and *E.coli*), both before and after accelerated aging processes, as exhibited in
459 Fig 6. They also paid special attention to the reasons responsible for failure of some
460 biocide formulations and concluded that the water-soluble bactericide showed a lower

461 retention rate in concrete and thus plays a poor role in protecting concrete in the long
462 term [16]. Urziet al. [45] evaluated the efficiency of three water-repellent compounds
463 and two biocide compounds, i.e. ALGOPHASE and the new water miscible formulation
464 ALGOPHASE PH 025/d having the same active ingredient 2,3,5,6-tetrachloro-4-
465 methylsulfonyl-pyridine, against microbial colonization of mortars both in laboratory
466 conditions and outdoors. They observed that the application of water-repellent alone
467 was insufficient to prevent biofilm growth on the surface, whereas the combined
468 application of water repellents and biocides in a single step prevent microbial growth,
469 reflecting by complete absence of bacterial colonization, absence of algal colonization,
470 dramatically reduced colonization by fungi on the surface of mortars (seen the
471 representative samples T4 and T5 shown in Fig. 7). Single-step application of biocide
472 and water repellent exhibits excellent performance due to biocide compound randomly
473 distribute below, between and above the hydro-repellent film. In this way, the biocide
474 has the ability to remove the remains of old colonization below, and stop new microbial
475 colonization on the surface [45]. Shook and Bell [37] evaluated the antimicrobial effect
476 of ConShield using wafers of concrete mortar incubated with a bacterial suspension of
477 T.thiooxidans, T. thioparus, and T. denitrificans. The results indicated that the viable
478 bacterial count of concrete wafers treated with ConShield is zero, suggesting that
479 ConShield killed all of the tested bacteria with a complete 100% kill after 24 hours.
480 Moreover, it is reported that ConBlock MIC [75], whether integrated throughout the
481 matrix of concrete when used as an admixture and/or directly applied to concrete as a
482 surface treatment, it inhibits the growth of bacteria, fungi, mold, and algae. Freed et al.
483 [56] evaluated the efficacy of concrete reinforced with fibers incorporating Microban
484 B. The inhibition zone of concrete treated with polypropylene fibers containing
485 Microban B towards E. coli, S. aureus, and mixed mold(fungi) was 3,4, and 2 mm,

486 respectively, indicating fibers carrying Microban B could kill microorganisms.



487 Fig. 6 Effectiveness of concrete incorporated with PL-UV-H-2 B formulation against
 488 different microorganisms: (a) before and (b) after the accelerated aging process [16].
 489



490 Fig. 7 Enumeration of fungi (CFU g⁻¹) colonizing mortar probes after 15 months of
 491 outdoor exposure. L +S=lime +sand and P +L=Pozzolana +lime. T0 represents
 492 untreated mortars; T1, T2, T3 represents mortar samples treated with different water-
 493 repellent alone; T4, T5, T6 represents mortar probes treated both with water-repellent
 494 and biocide; T7 represents mortar probes treated with biocide alone. T4, T5, and T7
 495 treated with ALGOPHASE, and T6 treated with ALGOPHASE PH 025/d [45].
 496

497 Above investigations have indicated that antimicrobial agents could endow
 498 concrete with antimicrobial property to varying degrees. Antimicrobial properties of
 499 antimicrobial concrete is largely depending on respective intrinsic natures, types and
 500 contents of antimicrobial agents. However, the existing researchers paid little attention
 501 to the impact of the addition of antimicrobial agents on the microstructure of concrete.
 502 It is necessary to establish the underlying connections between different properties as

503 well as the microstructure of concrete after adding antimicrobial agents. Moreover, high
504 retention rate of antimicrobial agents in concrete is required in order to maintain the
505 long-lasting inhibitory or killing effect towards microbes, while the long-term retention
506 rate of a biocide and its influence on the other properties of concrete are poorly
507 understood [35, 65].

508 4.2 Mechanical properties

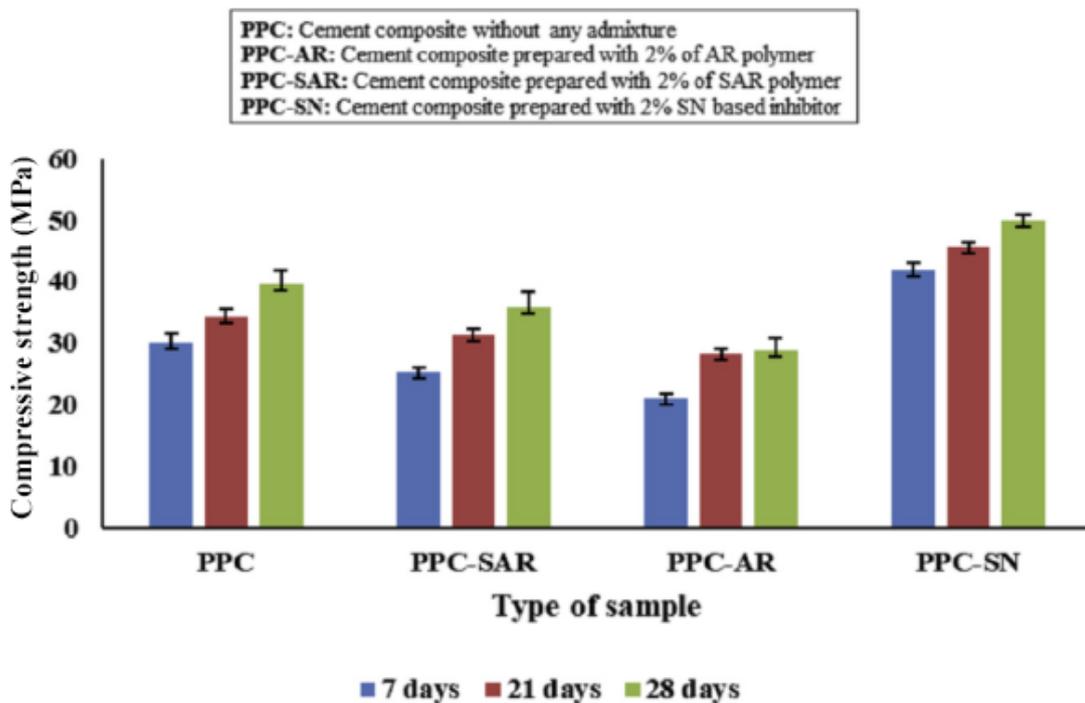
509 Antimicrobial concrete exhibited different mechanical properties for various types
510 and quantities of antimicrobial agents added. Kim et al. [61] reported that compressive
511 strength of concrete with antimicrobial watertight admixture, of which antimicrobial
512 ingredients are nickel and tungsten compounds, was decreased at an early age but the
513 long-term compressive strength was increased. De Muynck et al. [25] observed a small
514 decrease in compressive strength of mortar specimens added with the highest
515 concentration of zeolites (4.65%), i.e. 41.1 ± 0.8 MPa as compared to 49.0 ± 3.4 MPa for
516 control specimens. Kong and Zhang et al. [65,76] tested the 7 days, 28 days and 56 days
517 compressive strength of concrete added with different types and contents of bactericide.
518 They observed that the 28 days compressive strength of concrete adding with copper
519 phthalocyanine (CP) was enhanced by 60% with the dosage of 0.1%, which indicated
520 that CP not only increased the fluidity of concrete, but also accelerated the hydration of
521 cement, thus promoted the strength development by dispersing cement. Meanwhile, the
522 enhancement of compressive strength also makes some contribution to maintain the
523 surface pH of concrete added CP as high as 10.6. However, the strength of concrete will
524 be impaired when the contents of zinc oxide and dodecyl dimethyl benzyl ammonium
525 added in concrete are more than 0.05% [65,76]. Umar et al. [36] investigated the
526 strength development of four types of cement composite modified with polymer/added
527 inhibitor at the age of 7, 21, and 28 days. The results showed that compressive strength

528 of cement composite admixed with sodium nitrite-based inhibitor is increased by 26%
529 (28 days) with respect to that of cement composite without any admixture, and higher
530 than cement composite prepared with styrene acrylate copolymer and acrylic polymer,
531 as shown in Fig. 8. Vaquero et al. [16] obtained that the 28 d compressive strength of
532 concrete mixed with multicomponent formulation PL-UV-H-2 B was 37.1, 36.9, 35.7,
533 and 34.9 MPa when the content is 0, 0.15, 0.2, and 0.3%, respectively, and the 28 d
534 flexural strength was 9.4, 8.6, 8.2, and 8.5 MPa when the content is 0, 0.15, 0.2, and
535 0.3%, respectively. Consequently, they concluded that the addition of PL-UV-H-2 B in
536 concrete only slightly decreased the compressive strength and flexural strength as
537 compared to those of control samples [16]. Moreover, Do et al. [59] observed that
538 compressive and flexural strengths of cement mortar containing the antifungal agent of
539 isothiazoline/cabamate were almost equal to those of non-added cement mortar; hence,
540 they concluded that the addition of isothiazoline/cabamate has a very little adverse
541 impact on compressive and flexural strengths of cement mortar and is negligibly
542 insignificant.

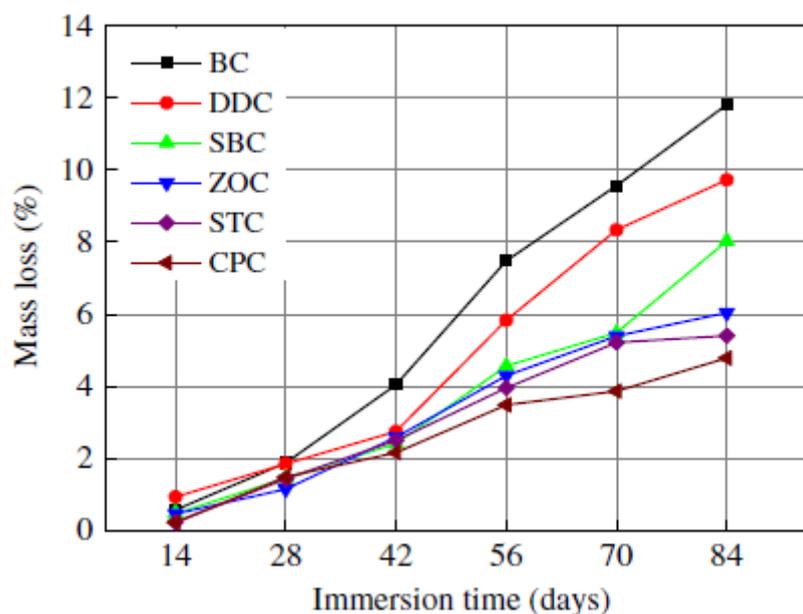
543 4.3 Mass/weight loss

544 Researchers not only investigated the antimicrobial and mechanical properties of
545 antimicrobial concrete, but also paid attention to its mass/weight loss. For example,
546 Negishi et al. [41] obtained that the weight loss of cement specimens without
547 antimicrobial agents, with 0.075% metal nickel, and with 0.075% metal nickel plus
548 0.075% calcium tungstate was 10, 6, and 1%, respectively, after being exposed to a
549 sewage treatment plant containing 28 ppm of H₂S for 2 years. The least weight loss of
550 nickel modified samples after adding calcium tungstate was due to the higher binding
551 tendency of tungsten to *A. thiooxidans*. As it can be seen in Fig. 9, there is an apparent
552 difference in mass losses in specimens with various bactericides and without adding

553 any bactericide, the mass loss rate of concrete specimen with copper phthalocyanine
 554 was the lowest (4.78%) as compared to other specimens, providing evidence that copper
 555 phthalocyanine has the best effect on resistance to the microbial induced corrosion of
 556 concrete [62]. Bao [67] reported that the mass loss of reference mortars and mortars
 557 added with mineral powder and fly ash was 1.26, 0.44 and 0.47% after an immersion
 558 in intensified sewage for 5 months, respectively. While the mass loss of mortar samples
 559 with antimicrobial agent sodium tungstate and sodium bromide reached 0.57% and
 560 0.6%, which indicated that incorporation of admixture has a better improvement effect
 561 than antimicrobial agents from the perspective of reduced mass loss. In addition, Shook
 562 and Bell [37] conducted an in-situ field test using concrete samples from concrete pipe
 563 in a sewer manhole which had evident corrosion occurring and an obviously high H₂S
 564 concentration. They obtained that concrete samples treated without ConShield had a
 565 great weight loss of 3.44%, whereas the concrete samples treated with ConShield
 566 showed a significantly lower weight loss of 0.32% after 3 months.



567 Fig. 8 Comparison of compressive strength of (SAR denotes styrene acrylate
 568 copolymer, AR denotes acrylic polymer and SN denotes sodium nitrite) [36].



569 Fig. 9 Effect of various bactericides on mass loss of concrete immersed in sewage
 570 [62]. DDC, SBC, ZOC, STC, CPC, and BC represent concrete incorporated with
 571 dodecyl dimethyl benzyl ammonium chloride, sodium bromide, zinc oxide, sodium
 572 tungstate, copper phthalocyanine, and plain concrete without bactericide, respectively.
 573

574 5. Antimicrobial mechanisms of antimicrobial agents

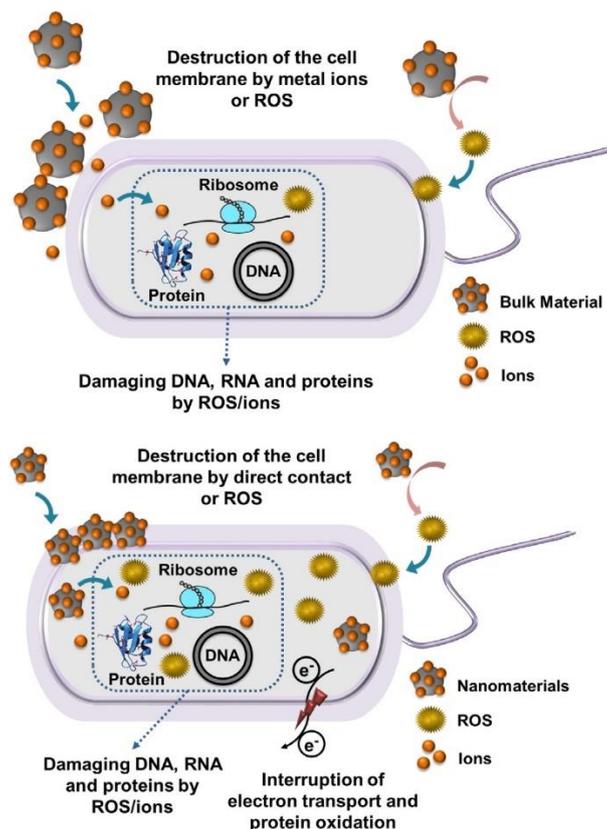
575 5.1 Antimicrobial mechanisms of inorganic antimicrobial agents

576 The antimicrobial mechanisms of heavy metal antibacterial agents towards
 577 microorganisms attached to and/or penetrated into concrete is generally considered to
 578 follow the reactions below. During the action of antibacterial agents, metal ions
 579 gradually dissolve and react with thiol group (-SH), amino group (-NH₂) and other
 580 sulfur nitrogen-containing functional groups existing in proteins and nucleic acids of
 581 bacteria, which inhibit or inactivate some necessary enzymes, and disturb the osmotic
 582 stability of the cell, thus achieving the antibacterial purpose [34,51,77]. More
 583 specifically, the action of silver ion released from the zeolite matrix in concrete and
 584 reactive oxygen species (ROS) generated from silver within the matrix are considered
 585 as the mechanisms of bactericidal action of silver-loaded zeolites, and it has been
 586 reported that either the silver itself or the ROS must interact with biological
 587 macromolecules like enzymes and DNA by an electron release mechanism to maintain

588 long-lasting antibacterial effect [63, 70]. It is assumed that nickel does not attack on
589 bacteria themselves, but binds to an enzyme of bacteria to exhibit growth inhibitory
590 effect [43]. Nogami et al. [39] concluded that nickel ions incorporated into concrete
591 bind to the plasma membrane and inhibit the activity of sulfur dioxygenase and sulfite
592 oxidase of *T. thiooxidans* to exert its inhibitory effect. Maeda et al. [40] also stated that
593 nickel binds to *T. thiooxidans* cells and inhibits enzymes involved in sulfur oxidation
594 of the bacterium, consequently inhibiting cell growth and sulfuric acid generation.
595 Similarly, tungsten exerts its antimicrobial effect on *A. thiooxidans* by binding to *A.*
596 *thiooxidans* cells and inhibiting the sulfur oxidation enzyme system, such as sulfur
597 oxidase, sulfur dioxygenase and sulfite oxidase of cells [41]. Sugio et al. [42] also
598 studied the mechanism of growth inhibition by tungsten in *A. ferrooxidans*, concluding
599 that tungsten binds to cytochrome c oxidase in plasma membranes and inhibits
600 cytochrome c oxidase activity, stopping cell growth from oxidation of Fe^{2+} . Moreover,
601 Kim et al. [61] ascribed the antimicrobial mechanism of antimicrobial metals (Ni and
602 W) to the destruction of cell membrane or internal protein tissue of microbe by Ni and
603 W according to simulation tests.

604 Significantly increased surface area-to-volume ratio of nanoparticles contributes
605 to greater interaction with microorganisms and enhances the release of toxic ions,
606 assisting nanoparticles to achieve excellent antimicrobial properties [3,78]. The
607 multiple bactericidal mechanisms of nanomaterials, such as copper oxide and zinc
608 oxide nanoparticles, have been attributed to damage of the cell membrane by either
609 direct contact with nanoparticles or photocatalytic production of ROS; release of toxic
610 ions; interruption of electron transport, protein oxidation, and modification of
611 membrane charges. Degradation of DNA, RNA and proteins by ROS, and lowering the
612 production of ATP due to acidification and ROS production also accounts for

613 bactericidal properties of nano-sized materials [3,79]. Fig. 10 illustrates a comparison
 614 of antibacterial mechanisms between antimicrobial nanomaterials and their bulk
 615 counterparts. In addition, the two major explanations to the photo-sterilization
 616 mechanism of concrete involving nano-TiO₂ under light are the attack of chemical
 617 species leading to the death of microorganisms or the biological structure destruction
 618 causing the inactivation of microorganisms [55].



619 Fig. 10 Illustration of possible bactericidal mechanism of nanomaterials (bottom)
 620 compared to their bulk form (top) [3].

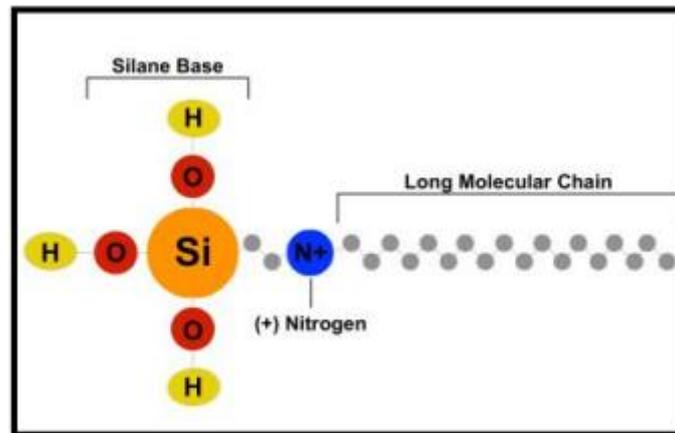
621 5.2 Antimicrobial mechanisms of organic antimicrobial agents

622 Generally, organic antimicrobial agents inhibit the growth and reproduction of
 623 microorganisms by destroying cell membranes, denaturing proteins, or disrupting
 624 metabolic processes. The phthalocyanine compound contained in concrete or mortar
 625 can be easily introduced into the cell of SOB, inhibiting enzyme reaction within the cell
 626 and, eventually, killing SOB [11]. In terms of copper phthalocyanine [62,65,76], its

627 high bactericidal property towards bacteria is mainly provided by copper ions. Copper
628 ions could interfere with the metabolic process of bacterial cells or interfere with the
629 function of various enzymes, losing their biological functions and eventually leading to
630 the death of cells [62,65,76]. Quats, like dodecyl dimethyl benzyl ammonium chloride
631 [62,65], the positively charged organic cations can be selectively adsorbed by the
632 negatively charged bacteria contacting with concrete. They could enter into the cell
633 membrane by permeation and diffusion, thus impede the semi-permeation action of cell
634 membranes and then inhibit the generation of enzyme to achieve the sterilization effect
635 [80]. McDonnel et al. [81] proposed that Quats target the cytoplasmic membrane and
636 damage the phospholipid bilayer. Additionally, the cellular membrane of bacteria will
637 be pierced by the long molecular carbon chain of silane quaternary ammonium chloride
638 (SQA) [57] and cell destruction will be triggered by ions exchange between the
639 positively charged ammonium cation of SQA and ions within cell membranes, are two
640 major hypotheses accounting for the antimicrobial working mechanisms of SQA. The
641 antimicrobial mechanism of concrete with ConBlock MIC [75] is the active ingredient
642 in ConBlock MIC 3-Trimethoxy silyl propyl dimethyl octadecyl ammonium chloride
643 has a positive charged nitrogen atom (as shown in Fig. 11), electrostatically attracting
644 many bacteria to the molecule. The molecular chain of carbon 18 atoms long pierces
645 the cellular membrane of bacteria and the outer cell is punctured upon reaching the
646 nitrogen atom. Consequently, it creates an uninhabitable environment for the
647 microbiological organisms on the surface of concrete [75]. As for ConShield, it endues
648 concrete with excellent antimicrobial effect through molecularly bonding to the
649 ingredients of concrete mix, then, providing hundreds of microscopic spikes over an
650 area of a single bacterium, which puncture the fragile single cell of bacteria [82, 83].

651 However, majority of the antimicrobial mechanisms mentioned above are relevant

652 to inhibiting or killing bacteria, the antifungal and algaecidal mechanisms of
653 corresponding antimicrobial agents used in concrete are rare, requiring further
654 investigations.



655 Fig. 11 Molecular structure of 3-Trimethoxy silyl propyl dimethyl octadecyl
656 ammonium chloride [75].
657

658 6. Applications of antimicrobial concrete

659 Concrete is the most abundant material in wastewater systems but at the greatest
660 risk for corrosion. Despite most of the findings are based upon laboratory testing, there
661 still exist some findings from practical applications of antimicrobial concrete.
662 Considering the superior antimicrobial property of concrete imparted by some typical
663 antimicrobial agents, one of the major applications of antimicrobial concrete is to
664 mitigate and control microbial corrosion caused by microbial metabolism in sewer
665 systems, such as concrete sewage pipes, sewer manholes, wastewater collection
666 systems and treatment plants, etc. For instance, in order to combat the growth and
667 proliferation of Thiobacilli in sewer systems, new sewer construction in Atlanta has
668 been utilizing concrete admixed with ConShield since 1997, and rehabilitation works
669 of concrete manholes in Columbus, OH, Oskaloosa Co., FL, Mt. Prospect, IL, Miami,
670 FL, and Corsica, TX have adopted the same material [37]. The results shown in Fig. 12
671 (a) and (b) clearly demonstrated the long-term protection due to the addition of

672 ConShield into concrete against microbial induced corrosion in the Maline Drop Shaft
673 [82]. Owing to the proved high antimicrobial effectiveness, ConShield has a wide range
674 of industrial applications in concrete structures mainly including two aspects: one is
675 new and rebuilt concrete structures subjected to highly concentrated sulfide conditions
676 like concrete pipe and manholes (Fig. 12 c), wet wells, lift stations, WWTP head works,
677 clarifiers, and the like. Another one is rehabilitation of heavily corroded manholes,
678 pipelines and tunnels in place via shotcrete (Fig. 12 d) [83]. Similarly, with excellent
679 antimicrobial power and long-lasting antimicrobial effect, concrete incorporated with
680 antimicrobial additive Zeomighty (zeolite-supported silver and copper) was popular in
681 the Japanese market. The practical applications of antimicrobial concrete with
682 Zeomighty include secondary concrete products such as Hume pipes, manholes, and
683 box culverts, cast-in place concrete structures for sewer and treatment facilities and
684 other premix mortar, etc., as shown in Fig. 13 [33]. Kurihara et al. [84] invented an
685 antibacterial agent composed of a silver compound (selected from silver carbonate,
686 silver oxide and silver phosphate), a copper compound (selected from copper carbonate,
687 copper oxide, copper phosphate and copper hydroxide) and an ion-retaining compound,
688 and concrete containing the antibacterial agent exhibits outstanding antibacterial effect
689 against SRB, SOB, and carboxylic acid-producing bacteria particularly in sewage
690 treatment plants. Uchida et al. [11] disclosed that the addition of phthalocyanine
691 compound (a metal phthalocyanine, a metal-free phthalocyanine, and derivatives
692 thereof) in concrete or mortar can be easily introduced into a cell of SOB, thus inhibit
693 and/or kill SOB via inhibiting enzyme reaction within the cell of SOB. Consequently,
694 the deterioration inhibitor with the effective component, phthalocyanine compound,
695 showed ability to mitigate the deterioration of concrete or mortar. Antimicrobial
696 concrete fabricated with copper phthalocyanine [62,65] has the merits of excellent

697 bactericidal performance, high retention rate of bactericide and low cost. Moreover, the
698 addition of copper phthalocyanine does not affect the performance of concrete.
699 Consequently, such antimicrobial concrete can be widely used in the construction of
700 municipal sewage facilities [85]. Moreover, it is stated that the antimicrobial additive
701 ConBlock MIC can be applied in new concrete infrastructure and cementitious
702 infrastructure repair products, for example, concrete pipe, manhole and septic tanks, or
703 for ready mixed concrete or cementitious mortars and liners [75]. With the advantages
704 of long-lasting bactericidal effect on SOB (one to several years), the low cost and
705 environmentally friendly chemical (i.e. nitrite), FNA spray [44] is a promising practical
706 technology for mitigate and control of microbially induced concrete corrosion.



(a) Corrosion prior to repair in 1999



(b) Shaft after repair in 2009



(c) New concrete pipe and manholes with ConShield precisely metered into the mix at the plant



(d) Rehabilitation of pipelines and tunnels in place by shotcrete added with ConShield

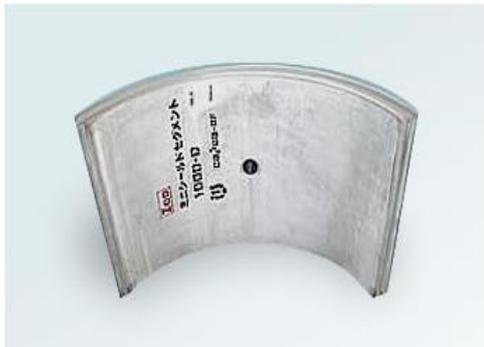
707 Fig. 12 Comparison before (a) and after (b) adding ConShield of the Maline Drop
708 Shaft [82], and (c) and (d) are the examples of industrial use of ConShield [83].



(a) pipe for a trenchless construction method



(b) manhole



(c) mini-shield segment before construction execution



(d) mini-shield segment after construction execution

Fig. 13 Examples of actual applications of antimicrobial concrete with Zeomighty [33].

710
711
712

713 In addition, according to [86], concrete added with copper oxide (methyl cellulose
714 as dispersant) and zinc oxide (fly ash as dispersant) was proved to be able to protect
715 marine ecological engineering construction from microorganism attack. Compared to
716 the untreated concrete columns with a number of plaques found on the surface, no
717 evidence of plaque was found on the surface of three treated concrete columns after 18
718 months. Similarly, concrete with TiO_2 , utilizing the light-induced bactericidal activity
719 of TiO_2 , can be employed to control microbiological growth on concrete surfaces, thus
720 enhancing the durability of concrete in ocean engineering. The same concrete can be
721 also used as exterior wall materials of buildings, achieving sterilization function by
722 decomposing bacteria attached on surface [49, 87]. Janus et al. [88] proposed that

723 concretes admixed with modified titania, with enhanced antibacterial properties, can
724 have a wide application in places demanding high sterilization levels, such as hospitals,
725 institutions, school and water storage tanks. In addition, Freed et al. [56] disclosed that
726 antimicrobial concrete reinforced with fiber carrying antimicrobial agents, such as
727 Microban B, has the ability to protect concrete from biological attack. The antimicrobial
728 agent is first incorporated into or coated onto fibers and then the treated fibers are
729 admixed with concrete. Such antimicrobial concrete, with the ability to inhibit growth
730 and contact of microorganisms such as bacteria, fungi, mold, etc., aims to be employed
731 in areas requiring extraordinary cleanliness such as food processing plants, hospitals,
732 kitchens, locker rooms, and the like.

733 **7. Summary and prospects**

734 Microbial attachment, colonization and eventually deterioration have been a great
735 threat to concrete structures in sewer systems, marine environments, buildings exposed
736 to high humidity and the like. Antimicrobial concrete, with the addition of inorganic or
737 organic antimicrobial agents, exhibits excellent antimicrobial effect against specific
738 microorganism and helps to address such issues caused by microorganism metabolism.
739 Also, the appearance of antimicrobial concrete makes infrastructures smarter and more
740 durable, prolongs the service life of infrastructures and lowers the huge cost by
741 rehabilitation and even replacement.

742 Despite many investigations have been conducted in this area in the past decades,
743 there still remains some key issues to be addressed. The relationship between
744 antimicrobial property and various affecting parameters (including contents, retention
745 rate and dispersion, etc.) should be further comprehensively investigated so as to
746 effectively enhance the antimicrobial effect of antimicrobial concrete. Combining
747 different antimicrobial agents to form biocide formulation according to their respective

748 intrinsic properties may be a promising strategy to boost antimicrobial efficiency. The
749 toxicity due to the release of some active ingredients into the environment during the
750 entire service life of inorganic antimicrobial agents such as nanoparticles and generally
751 temporary effectiveness for organic antimicrobial agents are impediments to the
752 widespread application of antimicrobial concrete. Moreover, the resistance of
753 microorganisms to antimicrobial agents has to be considered in developing
754 antimicrobial concrete.

755 Currently, most researches are restricted to the laboratory stage, practical
756 applications are few and field trials are still highly required to verify the feasibility of
757 antimicrobial concrete with aforementioned antimicrobial agents. The development of
758 antimicrobial concrete is based on the advancement of antimicrobial agents. In future,
759 it is expected to provide novel, high-efficiency, long lasting, broad-spectrum and
760 environmental-friendly antimicrobial agents for fabricating antimicrobial concrete. In
761 addition, antimicrobial concrete with its exceptional antimicrobial performance may
762 have an extended application in the field of fighting against viruses. Especially, the
763 world is in novel coronavirus pandemic now. Countries around the world are building
764 new hospitals or improving the facilities of existing hospitals to better treat infected
765 patients. Additionally, following its detection in the sewers in Massachusetts, the novel
766 coronavirus was also reported to be found in the non-potable water system used for
767 cleaning streets and watering parks in Paris. If the infrastructures such as hospitals and
768 sewage systems have the ability to kill viruses, it is beneficial for preventing the spread
769 and reproduction of viruses. Furthermore, the combination of new technologies may
770 promote the development of antimicrobial concrete, such as nanotechnology,
771 geopolymer technology, 3D printing/digital production technology, biotechnology, self-
772 assembly technology, damage and failure evaluation technology, organic-inorganic

773 composite technology and multiscale simulation technology [89-100].

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