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BIODETERIORATION OF CONCRETE

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Abstract
Concrete structures belong to these usually considered as indestructible because of their longer service life as compared with the most constructional products. However, they can get destroyed for a variety of reasons including these of biological origin. Many architectural and other building structures undergo biodeterioration when exposed to contact with soil, water, and sewage, as well as food, agricultural products and waste materials. This review explains the effect of some biogenic corrosive substances produced by the microorganisms on concrete.

Streszczenie
Konstrukcje betonowe należą do tych, które zazwyczaj są uważane jako niezniszczalne ze względu na ich dłuższy okres użytkowania w porównaniu z większością wyrobów konstrukcyjnych. Jednak mogą one ulegać zniszczeniu z różnych przyczyn, włączając te pochodzenia biologicznego. Wiele budowli architektonicznych i innych konstrukcji budowlanych ulega biodeterioracji w przypadku narażenia na kontakt z glebą, wodą i ściekami, jak również produktami i odpadami żywnościowymi i pochodzenia rolniczego. Ten przegląd wyjaśnia działanie na beton niektórych biogennych substancji korozyjnych produkowanych przez mikroorganizmy.

Keywords: **Concrete; Biodeterioration; Microorganisms; Metabolic activity; Biogenic substances.**

1. INTRODUCTION

Concrete is one of the strongest construction materials applied in centuries all over the world. Concrete structures belong to these usually considered as indestructible because of their longer service life as compared with the most constructional products. However, they can get destroyed for a variety of reasons including the material limitations, poor quality design and construction practices, as well as the hard exposure conditions.

Many architectural and other buildings structures undergo biodeterioration when exposed to contact with soil, water, sewage, as well as food, agricultural products and waste materials. Biodeterioration refers to undesirable changes in a material, caused by living organisms. They form specific communities that interact in many different ways with mineral materials and their external environment. This complex phenomenon occurs in conjunction with many physical and chemical destructive processes. Thus, it is difficult to distinguish an extent of the damage caused by biotic factors from that resulting from abiotic ones. However, according to US estimation, the contribution of microorganisms to the deterioration of materials as a whole may be within the range of 30% [1].

Biologically influenced corrosion of concrete has most often been detected in building foundations and walls (Fig. 1), and also in constructions such as dams, harbour and maritime structures, bridges, tanks, pipelines, cooling towers, silos and many others. This type of concrete deterioration occurs often in the food processing and storage works and in the abattoirs and buildings of holdings, in which the different microorganisms including bacteria, microscopic fungi, and algae are usually present at increased concentrations [1]. They colonize the material surface, and its pores, capillaries and microcracks, and cause the concrete damage resulting in aesthetic, functional, or structural problems.

Figure 1.
Concrete foundation covered with different lichens

2. BIODETERIORATION OF CONCRETE

Biodegradability of various mineral building materials including the concrete is mostly due to the increased concentration of carbonates and inorganic sulfur compounds, as well as other chemically aggressive reagents of either abiotic or biotic nature. Their interactions with components of the mineral materials play an essential role in their corrosion induction and process.

It should be taken into account that concrete is a heterogeneous material which is usually composed of the portland cement, aggregates (coarse and fine), water and admixtures (optional). Two main phases of the concrete, i.e. aggregates (59-75% of concrete volume) and hardened cement paste (25-40% of concrete volume) indicate quite different physicochemical properties. However, many physical, chemical and biological factors may affect both the concrete paste and aggregates destruction [2]. The physical factors may be of internal or external origin. Such concrete damages are mostly connected with bad composition of the concrete mixture and inappropriate maintenance of concrete. Both frost and high temperatures are these amongst many physical factors that most often influence concrete deterioration. Water evaporation from the cement paste leads to pores formation allowing the entry of water and aggressive reagents, such as Cl^-, NO_3^-, SO_4^{2-}, and CO_3^{2-}. Besides, some microorganisms seeding on the concrete surface and in its pores excrete the extracellular polymeric substances that change the concrete porosity

Table 1.
Chemicals that promote deterioration of concrete (based on [2])

Rapid deterioration of concrete	Moderate deterioration of concrete	
Aluminum chloride	Aluminum sulfate*	Mustard oil*
Calcium bisulfite	Ammonium bisulfate	Perchloric acid, 10%
Hydrochloric acid (all concentrations)*	Ammonium nitrate	Potassium dichromate
Hydrofluoric acid (all concentrations)	Ammonium sulfate*	Potassium hydroxide (>25%)
Nitric acid (all concentrations)	Ammonium sulfide	Rapeseed oil*
Sulfuric acid, 10-80 percent*	Ammonium sulfite	Slaughterhouse waste[2]
Sulfurous acid	Ammonium superphosphate	Sodium bisulfate
	Ammonium thiosulfate	Sodium bisulfite
	Castor oil	Sodium hydroxide (>20%)
	Cocoa bean oil*	Sulfite liquor
	Cocoa butter*	Sulfuric acid, 80% oleum*
	Coconut oil*	Tanning liquor (if acid)
	Cottonseed oil*	Zinc refining solutions[3]
	Fish liquor[1]	

* Sometimes used in food processing or as food or beverage ingredient.

[1] Contains carbonic acid, fish oils, hydrogen sulfide, methyl amine, brine, other potentially active substances.

[2] May contain various mixtures of blood, fats and oils, bile and other digestive juices, partially digested vegetable matter, urine, and manure, with varying amounts of water.

[3] Usually contain zinc sulfate in sulfuric acid. Sulfuric acid concentration may be low (about 6% in "low current density" process) or higher (about 22-28% in "high current density" process).

and permeability. Where porous concrete is in contact with water or saturated ground, the water phase is continuous on the material surface and various ions can readily enter the concrete matrix.

Many chemical substances including acids, alkalis, gases, oils and fats, sugars, and many others are very corrosive agents that cause a chemical corrosion of the concrete (Table 1).

The acids (organic and mineral), salts (mostly sulfates), hydrogen sulfide, ammonia and carbon dioxide can derive from aggregates, air, combustion gases, waters, deicing resources and industrial impurities. These substances can attack the cement paste, the aggregate, or both. It has been observed that natural pebble aggregates (gravels, base coals, sands) and aggregates from broken magmatic rocks (granites, basalts) in general do not corrode under chemical action. The chemical corrosion resistance of some artificial aggregates (pure furnace clinkers, ceramsite) is also well known. The natural broken calcareous rocks and the aggregates from furnace slag are corrodible in acid environments. Such environments are corrosive also for the cement paste.

The cement hydration results in formation of the calcium hydroxide $Ca(OH)_2$ which reacts with carbon dioxide (CO_2):

$$Ca(OH)_2 + CO_2 \rightarrow CaCO_3 + H_2O. \quad (1)$$

Resulting calcium carbonate $CaCO_3$ is sparingly soluble and its formation causes tightening of the cement paste and increase in the concrete compressive strength. This process called concrete carbonation is considered favorable for this material properties.

However, carbonates formation leads to the concrete shrinkage increase during drying, thus promoting the material cracking. Besides, concrete carbonation leads to its lower alkalinity and subsequently to corrosion of steel reinforcement.

Both calcium hydroxide and carbonate react with different chemical substances, especially with acids (both organic and mineral) and with some salts. Aluminates present in the concrete easily react with sulfates. Products of these reactions are strongly hydrated swelling salts expanding inside the concrete, thus forcing it to crack from inside.

The metabolic activity of microorganisms causes liberation of many acids as well as hydrogen sulfide and other corrosive reagents into environment (Table 2).

Table 2.
Biochemical processes essential for the mineral materials deterioration influenced by microorganisms indicating especially high corrosive aggressiveness (based on [3])

Biochemical process	Corrodible mineral materials	Microorganisms that cause biodeterioration
oxidation $NH_4^+ \rightarrow NO_2^-$	natural stones, brick, concrete, mortars	bacteria: *Nitrosomonas, Nitrosococcus, Nitrosospira, Nitrosolobus*
oxidation $NO_2^- \rightarrow NO_3^-$	natural stones, brick, concrete, mortars	bacteria: *Nitrobacter, Nitrococcus*
reduction $NO_3^- \rightarrow NO_2^-$	natural stones, brick, concrete, mortars	bacteria: *Escherichia, Pseudomonas*
reduction $NO_3^- \rightarrow NH_4^+$	concrete	bacteria: *Clostridium*
reduction $SO_4^{2-} \rightarrow H_2S$	natural stones, brick, concrete, mortars	bacteria: *Clostridium*; sulfate reducing bacteria (**SRB**): *Desulfovibrio, Desulfotomaculum, Desulfomonas*
oxidation H_2	brick, concrete, mortars	bacteria: *Alcaligenes, Micrococcus, Desulfovibrio*
oxidation S^o, RSC* $\rightarrow H_2SO_4$	natural stones, brick, concrete, mortars	sulfur oxidizing bacteria (**SOB**): *Acidithiobacillus, Thiobacillus, Sulfobacillus, Sulfolobus*
oxidation $Fe^{2+} \rightarrow Fe^{3+}$	natural stones, brick, concrete, mortars	bacteria: *Crenothrix, Gallionella, Leptothrix, Sphaerotilus, Sulfobacillus, Acidithiobacillus ferrooxidans*
oxidation $Mn^{2+} \rightarrow Mn^{4+}$	natural stones, brick, concrete, mortars	bacteria: *Bacillus, Gallionella, Pseudomonas, Sphaerotilus*
fermentative processes → organic acids; CO_2	natural stones, brick, concrete, mortars; glass (fungi)	**bacteria:** *Achromobacter, Bacillus, Clostridium, Eschericha, Flavobacterium, Lactobacillus, Proteus, Pseudomonas, Salmonella* **fungi:** *Aspergillus, Ceratostomella, Cladosporium, Fusarium, Hormoconis, Hormodendrum, Penicillium, Spicardia, Trichosporon*

* RSC – reduced sulfur compounds (inorganic)

The intensive growth of microorganisms is cause of accumulation of the corrosive biogenic substances on different construction and building materials including concrete [3-6]. Microorganisms can be present on the surface, in cleavages and fissures or within the material or interstitial space of the material. Their interaction with the material and its environment can give rise to biodeterioration. The effects of microbial metabolism are primarily of chemical nature, and later on, they may influence physical processes. All types of microorganisms (bacteria, algae, lichens, yeasts and fungi) can be involved in cases of biodeterioration, where they can act separately or jointly in complex interactions.

Bacteria and microscopic fungi are the microorganisms that mostly influence the concrete biodeterioration [7-13]. Extremely high corrosive towards the concrete are some biogenic organic acids (acetic, lactic, butyric and the like) and carbon dioxide, which are produced by many various microorganisms. Very corrosive are also biogenic mineral acids, especially the nitric acid produced by nitrification bacteria, as well as the sulfuric acid produced by sulfur oxidizing bacteria (SOB). They oxidize also the biogenic hydrogen sulfide (H_2S) which is a gas liberated into the environment by sulfate reducing bacteria (SRB). All these chemical substances of biological origin easily react with components of the concrete (Table 3) and cause its deterioration [14].

The concrete matrix disintegration by biogenic corrosive reagents is more intensive in comparison to the single application of corrosive substance on concrete surface. Moreover, the rate of attack is difficult to predict as various microorganisms indicate different metabolic activities. Continuous production of microbial metabolites at the site of attack causes its great intensification. Thus, microorganisms act as

Table 3.
Biogenic substances and chemical reactions essential for the microbiologically influenced deterioration of concrete (based on [3])

Biogenic organic acids	
$Ca(OH)_2 + 2\ C_2H_4(OH)COOH + 3\ H_2O \rightarrow Ca[C_2H_4(OH)COO]_2 \cdot 5\ H_2O$ (lactic acid → calcium lactate)	(2)
$Ca(OH)_2 + 2\ CH_3COOH \rightarrow Ca(CH_3COO)_2 \cdot H_2O + H_2O.$ (acetic acid → calcium acetate)	(3)
Biogenic carbon dioxide	
$Ca(OH)_2 + 2\ CO_2 \rightarrow Ca(HCO_3)_2$	(4)
$CaCO_3 + CO_2 + H_2O \rightarrow Ca(HCO_3)_2.$	(5)
Biogenic nitric acid	
$2\ NH_4^+ + 3\ O_2 \xrightarrow{Nitrosomonas} 2\ NO_2^- + 2\ H_2O + 4\ H^+$	(6)
$2\ NO_2^- + O_2 \xrightarrow{Nitrobacter} 2\ NO_3^-$	(7)
$Ca(OH)_2 + 2\ HNO_3 \rightarrow Ca(NO_3)_2 + 2\ H_2O$	(8)
Biogenic hydrogen sulfide and sulfuric acid	
$SO_4^{2-} + 2\ H^+ + 4\ H_2 \xrightarrow{SRB} H_2S\uparrow + 4\ H_2O$	(9)
SRB – sulfate reducing bacteria	
$H_2S + 2O_2 \xrightarrow{SOB} H_2SO_4$	(10)
SOB – sulfur oxidizing bacteria	
$Ca(OH)_2 + H_2SO_4 \rightarrow CaSO_4 \cdot 2\ H_2O.$ (gypsum)	(11)
$3CaO \cdot Al_2O_3 \cdot 3CaSO_4 \cdot nH_2O + 3(CaSO_4 \cdot 2H_2O) + H_2O \rightarrow 3CaO \cdot Al_2O_3 \cdot 3CaSO_4 . 32H_2O$ (gypsum → ettringite)	(12)

primary point sources for incessant supply of corroding agents.

2.1. Biogenic organic acids

Many biogenic organic acids that attack concrete originate from acid-containing or acid-producing substances, such as silage, fruit juices, sour milk and animal wastes. The concrete corrosion influenced by biogenic organic acids follows due to their reaction with basic components of the cement paste, i.e. calcium hydroxide and carbonate. The primary effect of any type of acid attack on concrete is the dissolution of the cement paste matrix. Fermentative microorganisms excrete into the environment many organic acids, including the lactic, acetic and butyric acids. These acids corrosive aggressiveness is relative to their kind and strength. Resulted salts: lactate, acetate, butyrate and others are water-soluble and rinsed out the concrete surface [4, 11]. The concrete biodegradability influenced by biogenic organic acids depends mostly on the cement and aggregate types [2, 5]. The acid attack on concrete surface with limestone and dolomite aggregates (composed of calcium, and calcium and magnesium carbonates, respectively) causes the aggregate dissolving at a rate similar to that of the cement paste, thus leading to the surface uniform deterioration. Deterioration of the concrete with siliceous gravel, granite or basalt aggregates usually appears in the aggregates salience from the concrete matrix. As the primary effects of the concrete biodeterioration due to acid attack, the concrete higher humidity and acidity (reflected by pH-decrease to values lower than 5), as well as simultaneously decrease in the material compressive strength and bending strength are usually observed [10, 15, 16].

2.2. Biogenic carbon dioxide

The corrosive aggressiveness of some waters and humid grounds may result from presence in them of aggressive carbon dioxide (CO_2) which may originate (among other things) from biochemical processes carried out by many (micro)organisms present in these environments. The lower is the water hardness, the higher its corrosive aggressiveness. The increased concentration of CO_2 in waters penetrating through the ground causes its weakly acid reaction due to formation of the carbonic acid. This acid reacts with the calcium hydroxide to form the water-soluble calcium hydrogen carbonate $Ca(HCO_3)_2$ which is easily eluted from the concrete. In compacted concretes, this effect is insignificant even at increased concentrations of CO_2 because of slower absorption of water as compared with the ordinary concrete [12].

2.3. Biogenic nitric acid

The biogenic ammonia present in water environments stimulate the concrete corrosion caused by the biogenic nitric acid. The ammonization bacteria (urobacteria) decompose urea and cause the ammonia formation. Bacteria belonging to *Nitrosomonas, Nitrosococcus, Nitrosospira* and *Nitrosolobus* genera oxidize the ammonia to nitrous acid subsequently oxidized to nitric acid by *Nitrobacter* and *Nitrococcus* bacteria. This acid reacts with calcium compounds in the hardened cement paste to form the water-soluble calcium nitrate, easily rinsed from the concrete structure [17]. It has been shown that the biogenic nitric acid produced by the nitrifying bacteria causes severe corrosion of the mineral building materials. High cells numbers of the nitrifying bacteria were present on both historical sandstone buildings as well as on modern concrete buildings [8, 9].

2.4. Biogenic hydrogen sulfide and sulfuric acid

The biogenic hydrogen sulfide (H_2S) is the end metabolic product of SRB living in anaerobic zones of the soil, polluted waters and sewage. The presence of H_2S and sulfuric acid is usually a reason of the corrosion of concrete-sewer pipes in which the bad ventilation and a small sewage flow occur [12, 18]. Such conditions are favorable for the SRB development in the slime, especially in the bottom layer. The SRB metabolic activity leads to production of H_2S then oxidized by SOB to sulfuric acid (Fig. 2).

The biogenic sulfuric acid concentration in concrete pore solution may rich a level as high as 10% that corresponds to a pH value less than 1.0. In some cases, the pore solution indicated a pH as low as 0.6 because of microbiologically induced attack, although having an initial alkalinity as high as a pH of 13. The sulfuric acid causes the sulfate attack on the cement paste leading to formation the ettringite $(CaO)_6(Al_2O_3)(SO_3)_3 \cdot 32H_2O$ (called Candlot's salt), and sometimes also the thaumasite $CaSiO_3 \cdot CaCO_3 \cdot CaSO_4 \cdot 15H_2O$ [20, 21]. During crystallization, these salts enlarge their own volume about twice as compared with the initial substrata volume. It makes for the concrete expansion leading to its cracking and disintegration.

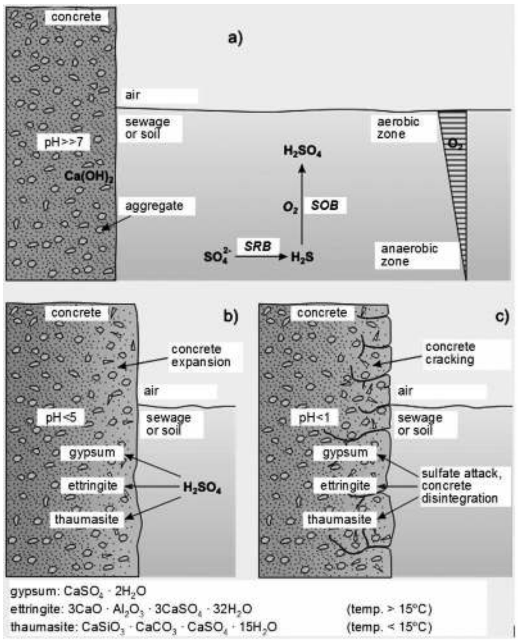

Figure 2.
Concrete biodeterioration influenced by sulfate reducing bacteria (SRB) and sulfur oxidizing bacteria (SOB) (picture adapted from [19], with modification)

The concrete biodeterioration is mostly due to metabolic activity of SOB belonging to *Acidithiobacillus* and *Thiobacillus* genera, especially of *A. thiooxidans* species (formerly *T. thiooxidans*). These bacteria were a major cause of demolition of the sewer canals in Hamburg [22]. The mass-losses in concrete specimens treated with liquid salt medium inoculated with *A. thiooxidans* bacteria were even 30-times higher as compared with samples simultaneously treated with a sterile medium. Besides, the decrease in the concrete specimens bending strength was three times greater, too. The corrosion rates of concrete may be as high as 1/2” to 3/4” (1.27-1.9 cm) per year, depending on concrete quality.

3. SOME CASES OF CONCRETE BIODETERIORATION

The history of research on the concrete acid and sulfate corrosion due to biogenic sulfuric acid attack goes back to the 1940s [14]. The first reports referred to this type of concrete damage described results of the microbiologically influenced corrosion of concrete gravity sewer pipes in countries with hot climate (Australia, Republic of South Africa). However, this problem appeared also in Scandinavian countries, despite the cold climate prevalent there. Warscheid et al. [23] have stated, that tropical climates favor the destructive activity of corrosion-influencing microorganisms, whilst air-pollution supports biodeterioration of mineral construction materials in moderate zones. In many concrete structures biodeterioration effects have been evidenced in Germany in the 1970s and the 1980s [22]. Essential increase in concentration of SOB, mainly of *A. thiooxidans* species usually caused the strong corrosion of concrete sewer pipes due to considerable acidifying of the concrete, in which the pH of the pore solution decreased to 1-2. The moderately – corroded concrete indicates the pH 3-5. The SOB quantity is usually negligible in the non-corroded concrete indicating pH>7. Correlation between the pH-changes in pore solution and the number of *A. thiooxidans* bacteria present in the concrete point to biological origin of this acidifying effect. However, pH-changes do not always reflect an extent of bacterially influenced attack on the concrete due to buffering properties of some components. Nowadays it is widely accepted that the concentration of *A. thiooxidans* bacteria in concrete surface layer may be the best indicator of its biodeterioration intensity.

Ważny and Czajnik made the interesting finding in 1963, during examination of the concrete experimental tunnel in Warsaw metro (after [16]). Surprisingly, the microscopic fungi *Coniophora puteana* and *Serpula lacrymans* were present in corroded concrete matrix although these microorganisms are usually responsible for the wood deterioration. Further laboratory research of these fungi influence (for six months) on the concrete samples made of the Portland cement 450 showed their humidity increase up to 18-25%, the pH decrease from value 12 to values 5-7, and 5-20% decrease of their bending strength (one sample infected by *Serpula lacrymans* indicated even 80% lower bending strength). These findings proved a thesis that microscopic fungi may strongly influence the physical, chemical and mechanical proprieties of the concrete due to its contact with the infected wood [16]. These observations are compatible with those presented by Paajanen et al. [15]. Other experiments indicated an important hazard for the concrete due to *Fusarium* fungi and *Acidithiobacillus* bacteria synergistic action leading to the calcium ions liberation from the concrete structure and their complexing by extracellular polymers [11].

4. CONCRETE PROTECTION FROM BIODETERIORATION

Generally, the methods commonly used for the concrete structures protection from biodeterioration include modifications of concrete mix design; coatings that may be sprayed, painted or rolled onto the concrete surface; and liners. The concrete mix modification usually involves increasing the alkalinity, since the corrosion rate is inversely proportional to concrete alkalinity. The concrete that we expect to expose on the biological attack should indicate the water-to-cement ratio $w/cm \leq 0.45$, and depth of the water penetration < 2.0 cm, and should contain specific additives including polypropylene or other fibers, and biocides [24]. The additives may be prepared as bacteriostatic composite systems protecting concrete for a long time [25-27]. Biocides selection must always depend on the microorganisms that will settle the concrete stone [17]. Simultaneous usage of the biocide and the protective coating as well as the biocides addition to the coating are more frequently recommended [28].

5. CONCLUDING REMARKS

Although the microbiologically influenced deterioration (MID) of mineral materials including the concrete occurs simultaneously with many physical, chemical and electrochemical processes of abiotic character, MID cannot be identified with these type processes. MID is the process resulting from attack of biogenic substances of large corrosive aggressiveness, which are the products of metabolic activity of multiplying microorganisms [1, 29, 30]. Their intensive growth (exponential) causes production of more and more quantities of enzymes that are biocatalysts of all metabolic processes. Taking into account that microorganisms cause both the initiation, as well as the intensification of corrosion processes, and on the other hand the fact that any construction effective protection against the MID requires usage of specific antimicrobial methods and/or biocides, the MID discrimination seems to be advisable as the special type of materials corrosion.

It is worth to noticing that many problems related to the biologically influenced deterioration of materials have not been solved until now, in spite considerable progresses in recognition of the MID kinetics and mechanisms. Thus, further research on biodeterioration processes and methods of protections against these processes are necessary.

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