



Article

The Effect of Calcium Stearate Additives in Concrete on Mass Transfer When Exposed to *Aspergillus niger* Fungi

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Abstract

Understanding and predicting the damage to concrete caused by microorganisms in aquatic environments is challenging, highlighting the need for effective, simple, and inexpensive preventative methods. This paper presents the results of a study on the effect of calcium stearate addition on the kinetics of mass transfer processes occurring in cement stone exposed to *Aspergillus niger* fungi under humid conditions. Calcium stearate was added into the cement mix during sample preparation at concentrations of 0.5% and 1% by cement weight. After curing, the cement stone surfaces were inoculated with *Aspergillus niger*. To investigate mass transfer processes during biodegradation, the samples were immersed in water. Calcium leaching from the cement stone was quantified using complexometric titration of the water, while the calcium content within the cement stone was determined by derivatographic analysis. The quantitative indicators of calcium leaching in water from cement stone with calcium stearate additives were 2.5 times lower. The profiles of calcium concentrations in the thickness of cement samples demonstrated an increase in the intensity of mass transfer under the influence of fungi and a significant decrease in the processes in hydrophobic cement stone. The values of the mass conductivity coefficients for fungal-infected samples in water differed by two orders of magnitude from 10^{-9} and 10^{-11} [m²/s] for conventional and hydrophobic concrete. The mass transfer parameters (flow density, mass conductivity coefficients, and mass transfer coefficients) revealed a 3-fold slowdown in mass transfer processes during fungal exposure in cement stone with a hydrophobic additive compared with control samples. A mathematical model of concrete biocorrosion was used to predict the durability of concrete under humid conditions with fungal exposure. The predicted maintenance-free service life of concrete without additives is 15 years, whereas for hydrophobic concrete, it is 25 to 30 years. The research results are used in the design of concrete structures in conditions of high humidity, in the development of new compositions of hydrophobic concretes, to predict the service life of concrete structures, and in the creation of methods for preventing biological damage to concrete structures.



Academic Editor: Yafei Xu

Received: 20 September 2025

Revised: 8 October 2025

Accepted: 10 October 2025

Published: 15 October 2025

Citation: Konovalova, V.S.; Strokin, K.B.; Galtsev, A.A.; Novikov, D.G. The Effect of Calcium Stearate Additives in Concrete on Mass Transfer When Exposed to *Aspergillus niger* Fungi. *J. Compos. Sci.* **2025**, *9*, 569. <https://doi.org/10.3390/jcs9100569>

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Keywords: concrete fungal deterioration; mass transfer in concrete; hydrophobic concrete; intensity of calcium leaching; concrete water resistance; concrete bio-resistance

1. Introduction

Concrete and reinforced concrete are widely used in the construction of residential, industrial, hydraulic, sanitary and other structures under a variety of operating conditions. The required characteristics of concrete and reinforced concrete can be obtained by utilizing various types of cement and employing different technologies [1–4]. Calcium hydrosilicates are the main components of the cement stone structure and chemically interact with substances that penetrate the pores or act on the surface. The aggressiveness of the environment towards concrete is assessed by the amount of reacted and damaged cement stone [5–8]. Therefore, to establish and predict the rate of concrete deterioration, it is advisable to study the mass transfer processes in cement stone and determine the extent of its damage.

Concrete exhibits bioreceptivity, meaning it can harbor microorganisms and allow them to colonize its surface [9–11]. Fungi of the genus *Aspergillus* are among the most common microorganisms found on stone and concrete surfaces [12–16]. Fungi are most active in humid conditions. These fungal microorganisms can cause significant damage to stone and concrete structures due to their complex metabolic activity and its impact on the surface [17–19]. Biological destruction of the binding material in these structures begins with the absorption of calcium, leading to surface erosion [20,21]. During their life processes, *Aspergillus* fungi secrete organic acids that dissolve the calcium-containing components of concrete [17,22]. This acid exposure leads to decalcification of the concrete, resulting in increased porosity and decreased mechanical strength [22–24].

Continued research is recommended to determine the rate of concrete degradation when exposed to various types of microorganisms. This will make it possible to develop procedures and techniques to prevent or mitigate this impact. Consequently, effective and sustainable methods to increase surface resistance to biodegradation should be developed alongside new advancements in nanotechnology.

As a rule, microorganisms attach to hydrophilic surfaces, and their settling on hydrophobic surfaces is difficult. Therefore, volumetric hydrophobization of cement stone can be considered one way to prevent biofouling of concrete and the penetration of aggressive media deep into its structure. Hydrophobic additives are substances that impart hydrophobic (water-repellent) properties to the walls of pores and capillaries in concrete. The mechanism of action of hydrophobic additives involves their precipitation upon contact with cement hydration products, forming tiny droplets that create hydrophobic coatings on the walls of small pores and capillaries.

The use of hydrophobic additives in cement systems contributes to the formation of a dense and homogeneous structure during hardening. This is reflected in a decrease in both the number and size of macropores, as well as in their more uniform distribution throughout the cement stone. The number of macropores in cement systems with hydrophobic additives is 2 to 4 times lower than in systems without additives [25–27].

Hydrophobic additives influence the modification of cement hydration products. The incorporation of complex hydrophobic organomineral additives results in a greater proportion of gel-like fibrous and fine-needle calcium hydrosilicates within the hydrated matrix. This, in turn, enhances the dispersion and homogeneity of the cement stone structure [28–32]. The neoplasms in the structure observed in cement stone with the introduction of an organomineral additive are similar in nature to hydrate formations that occur when using plasticizing hardening accelerators. Furthermore, concretes with complex additives exhibit an increase in average density and strength, attributable to an elevated level of adsorption-bound water.

Salts of fatty acids are used as hydrophobic agents for building mixes. The introduction of zinc stearate, in amounts up to 2% by weight of cement, increases the resistance of

building mortars and concretes to acid attacks [33–35]. Cement mortar with the addition of 5% styrene-acrylic emulsion (SAE) and 1% sodium oleate exhibits better impermeability, hydrophobicity, and compressive strength compared to a solution with only 15% styrene-acrylic emulsion. This is explained by the physical barrier provided by the SAE and the hydrophobic effect of sodium oleate. Increasing the amount of sodium oleate beyond a certain point does not lead to a sustained improvement in water resistance, as it can saturate the concrete with air and increase its porosity [36]. Sodium oleate effectively increases the potential of steel reinforcement in the pore fluid of concrete, thereby inhibiting the initiation of pitting corrosion. It is adsorbed onto the surface of steel reinforcement, forming a dense film that prevents water, oxygen, and aggressive substances from reaching the metal [37]. Sodium oleate significantly improves the properties of lime-based solutions when used in an amount of 0.5% of the total weight of the dry solution [38]; specifically, water absorption decreases, resistance to freeze–thaw cycles increases, and compressive strength improves. Calcium stearate at 0.5% has demonstrated similar effects.

A dosage of 0.2–1 wt.% calcium stearate is recommended for building mixes to impart water-repellent properties [39–42]. Calcium stearate additives in cement mixtures are used to control efflorescence on the surface of materials (Patent US5460648A) [43–46]. The effect of the addition of calcium stearate on increasing both short-term and long-term water resistance of self-sealing concrete was analyzed. The addition of calcium stearate proved to be more effective in slowing the transfer of chlorides than used. The water-repellent layer formed along the capillary pores also significantly reduces capillary water absorption [47].

A complex microdisperse additive, obtained by co-grinding quartz sand, superplasticizer C-3, and calcium stearate, enables the production of fine-grained concrete with a compressive strength up to 50 MPa, a bending strength up to 8.3 MPa, water absorption of 1.4%, and frost resistance exceeding F75 [48].

When calcium stearate is added at a concentration of 1 kg/m³ to self-compacting concrete containing 10% fly ash, the mechanical and physical properties of the concrete are significantly enhanced. This results in improved compressive strength, reduced water absorption, lower permeability to chloride ions, and a diminished degree of corrosion effect [49]. The addition of calcium stearate, up to 2 wt.%, in fresh concrete led to increased strength and decreased chloride ion penetration, thereby enhancing durability [50,51]. The water absorption coefficient of concrete treated with a 2% solution of calcium stearate decreased to 2.5%, 2.3%, and 2.1% at the ages of 7, 28, and 56 days, respectively. For concrete containing 4% calcium stearate, water absorption dropped to 1% after 56 days. However, the use of calcium stearate in combination with other hydrophobic additives in the cement mixture did not exhibit the same level of effectiveness as when calcium stearate was used alone [52].

Calcium stearate significantly enhances the corrosion resistance of steel reinforcement in crack-free concrete exposed to a 3.5% NaCl solution for 60 days. At concentrations of 3% and 5% calcium stearate in the concrete, the rate of corrosion processes was reduced by 88.8% and 91.3%, respectively [53]. Calcium stearate can adsorb onto the surface of steel, forming insoluble hydrophobic salts.

The addition of calcium stearate into concrete effectively prevents corrosion in liquid media [54,55]. Calcium stearate also shows high efficiency in reducing the corrosion of steel reinforcement [53].

The purpose of this study is to investigate the kinetics of mass transfer in the cement stone of concrete containing the hydrophobic additive calcium stearate under fungal action. As a result, the mass transfer parameters are established, which can be used to predict the durability of concrete products. In this context, the kinetics of calcium leaching from cement stone damaged by *Aspergillus niger* fungi were examined, and the calcium content

was analyzed across the thickness of the cement stone. These data were used to calculate the mass transfer parameters, which were then incorporated into a mathematical model of biocorrosion in cement concrete to estimate service life. In this study, durability is assessed based on the intensity of corrosion mass transfer in cement stone, whereas in most studies, durability is characterized by the strength characteristics of concrete. The service life prediction was performed using mass transfer parameters established for the equilibrium state of mass transfer processes during the experimental study of cement stone biodegradation.

The research results will contribute to the characterization of the importance of corrosive groups of microorganisms as a factor of corrosion of concrete and reinforced concrete products. The obtained ideas about the intensity of mass transfer in cement stone during fungal action will complement the overall picture of corrosion phenomena in concrete. The array of data on the mechanisms of destruction of reinforced concrete is expanding, which must be taken into account when managing the life cycle of structures to establish service life and periods of repair and restoration work.

2. Materials and Methods

To study mass transfer in cement stone, samples were created using Portland cement, which were subsequently infected with *Aspergillus niger* fungi after curing. Following biofouling, some samples were placed in distilled water to determine the intensity of calcium leaching from the cement stone. The calcium content in the liquid medium was measured through direct titration of the water samples. Additionally, the calcium content in the cement stone was assessed using derivatographic analysis for fungal-infected samples, both prior to and following immersion in water.

For the production of cement stone samples, Portland cement CEM I 42.5 N, which has a standardized composition and contains no mineral additives, was used as the binder. The chemical and mineralogical compositions of Portland cement CEM I 42.5 N, as detailed in the manufacturer's quality certificate, are presented in Tables 1 and 2.

Table 1. Chemical composition of CEM I 42.5 N brand Portland cement, %.

SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	SO ₃	R ₂ O ^a
21.02	5.42	4.19	65.91	0.40	2.40	0.66

^a R₂O = total alkali (content of Na₂O and K₂O).

Table 2. The content of the main minerals in CEM I 42.5 N brand Portland cement, %.

C ₃ S	C ₂ S	C ₃ A	C ₄ AF
64.31	11.75	7.24	12.76

Cement stone samples were prepared using cement mortar of normal density with a water-cement ratio of W/C = 0.3. The compositions of the samples per 1 kg of cement paste are shown in Table 3. The tests were conducted after curing the samples for 28 days under standard conditions. To study the mass transfer processes in cement stone, the samples were shaped as cubes measuring 3 × 3 × 3 cm.

Table 3. The quantity of components for the production of cement stone samples, g.

Type of Sample	Cement	Water	Calcium Stearate
Without additives	769.23	230.77	-
0.5 wt.% additive	766.28	229.89	3.83
1 wt.% additive	763.36	229	7.64

To prevent biofouling and the progression of fungal degradation under humid conditions, calcium stearate was added to the cement mixture in amounts of 0.5% and 1% by weight of cement during the manufacturing of cement stone. The amount of calcium stearate was determined based on previous studies aimed at preventing chloride corrosion in concrete using this additive [54]. Additionally, it was found that the hydrophobic additive effectively inhibits the colonization of *Aspergillus niger* fungi on the concrete surface, thereby enhancing its performance [56]. Chemically pure fine crystalline calcium stearate was added into the cement mortar. The uniform distribution of this powder within the cement mix was achieved through mechanical stirring for 5 min.

The samples were coated with bitumen mastic on five sides (Figure 1a), leaving one surface exposed to biofouling. The filamentous fungus *Aspergillus niger*, commonly known as “black mold”, was used to infect the cement stone. This type of fungal microorganism is prevalent in daily life across surfaces made from various materials. Black mold rapidly colonizes and spreads, demonstrating a high survival rate. The fungal strain was provided by a laboratory specializing in the cultivation of industrial microorganisms. The samples were evaluated for their ability to serve as a food source for micromycetes (fungus resistance) and for the presence of fungicidal properties within their components, following the standard method outlined in GOST 9.048-89 “Unified system of corrosion and ageing protection. Technical items. Methods of laboratory tests for mould resistance”. The method involves infecting products, without cleaning of external contaminants, with a suspension of microorganisms and then maintaining them for 28 days under optimal conditions for growth. To infect the surface with fungi, a suspension of fungi in an amount of 1 cm³ with a concentration of 1–2 million fungal spores was sprayed. Two groups of samples were used for testing: test and control. The test samples were employed to assess the intensity of microorganisms’ development and their impact on materials’ properties. The control samples are used to evaluate the properties of products with high humidity, excluding the influence of microorganisms.

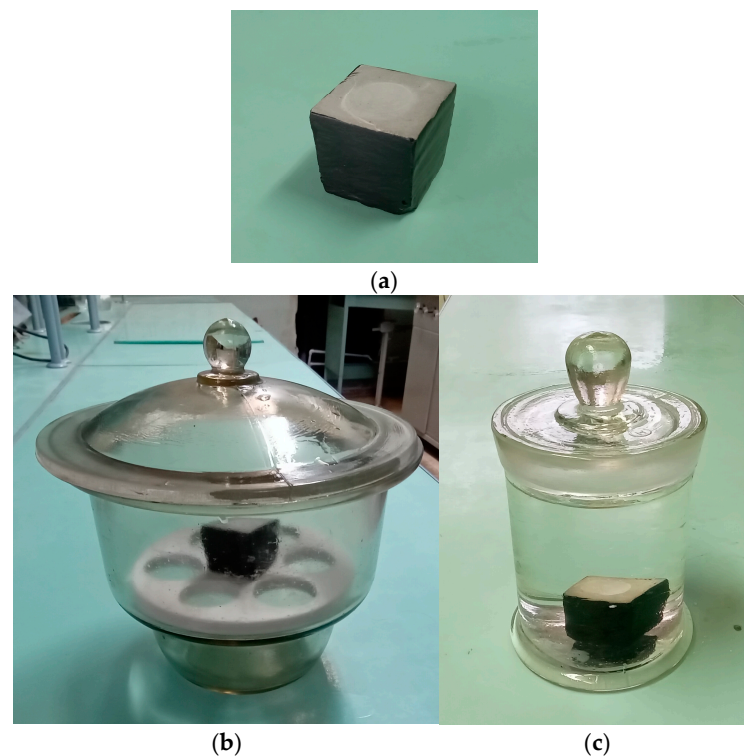


Figure 1. Images of the sample for corrosion resistance tests (a), and of the sample placement in installations for biofouling (b) and for determining the degree of leaching (c).

To transfer fungi to the surface of the samples, a nutrient medium is prepared consisting of 30 g/L malt extract, 1 g/L peptone, 20 g/L agar-agar, and distilled water up to a final volume of 1 L. The fungal suspension is prepared immediately before application. It is sprayed onto the surface of the cement stone. The infected samples are dried at a temperature of 25 °C and relative humidity of 70–90% until the drops evaporate, but not for more than 60 min. Petri dishes containing the samples are then placed in a desiccator, where distilled water is added to the bottom to create a high-humidity environment essential for microorganism development (Figure 1b). The distance from the walls of the desiccator should be at least 50 mm. The tests are conducted at a temperature of (29 ± 2) °C and a relative humidity of over 90% for 28 days. Every 7 days, the desiccator covers are opened for 3 min to allow air circulation.

The sample groups for each type of test consisted of 10 units without additives and 10 units with the addition of calcium stearate. The results were averaged over at least 5 measurements differing from the others by more than 5%.

The water tightness of the cement stone was determined by the “wet spot” method according to GOST 12730.5–2018 “Concretes. Methods for determination of water tightness”. The method determines the grade of concrete by water resistance using the “wet spot” technique as follows. The water pressure is increased incrementally by 0.2 MPa, maintained at each increment for 4 h, and applied for 1–5 min. The test continues until water filtration is observed on the upper surface of the sample, appearing as droplets or a wet spot. The water tightness of a series of samples is evaluated based on the maximum water pressure at which at least four of the six samples show no signs of water filtration.

Derivatographic analysis was performed using a standard technique on a Q-1500D derivatograph (MOM Szerviz Kft, Budapest, Hungary). Samples were prepared by dividing the cement stone into three 1 cm thick sections, and the analytical sample was taken from the center of the section. Heating was carried out up to 1000 °C at a rate of 10 °C/min. The obtained TG curves were used to calculate CaO mass during the thermal decomposition of Ca(OH)_2 and CaCO_3 . Next, the recorded amounts were recalculated by the mass of the sample plate.

To determine the degree of fungal influence on the rate of calcium leaching from concrete cement stone, samples were individually placed in containers with 1 L of distilled water (Figure 1c). Quantitative determination of the calcium cation content in liquid media was performed using complexometric titration. This method involves direct titration of the test solution with a standard solution of EDTA (complexon III) using either chromogen dark blue acid or murexide as an indicator. These indicators form a red complex compound with calcium ions. For the analysis, 25 mL of the solution was transferred to a flask, and 50 mL of distilled water was added. Subsequently, 25 mL of a 20% sodium hydroxide solution and 2–3 drops of the indicator were added. A 0.1 N EDTA solution was then added dropwise to the flask with continuous stirring until the red color of the liquid changed to purple or blue.

The calculation of the calcium cation concentration in the solution is performed using the following equation:

$$g_A = \frac{E_A \times N_B \times V_B}{1000} \quad (1)$$

where g_A is the total content of calcium cations in solution, [g]; E_A is the equivalent of calcium cations; N_B is the concentration of standard EDTA solution, [eq/L]; V_B is the volume of standard EDTA solution spent on titration, [mL].

The general scheme of the experiment is shown in Figure 2.

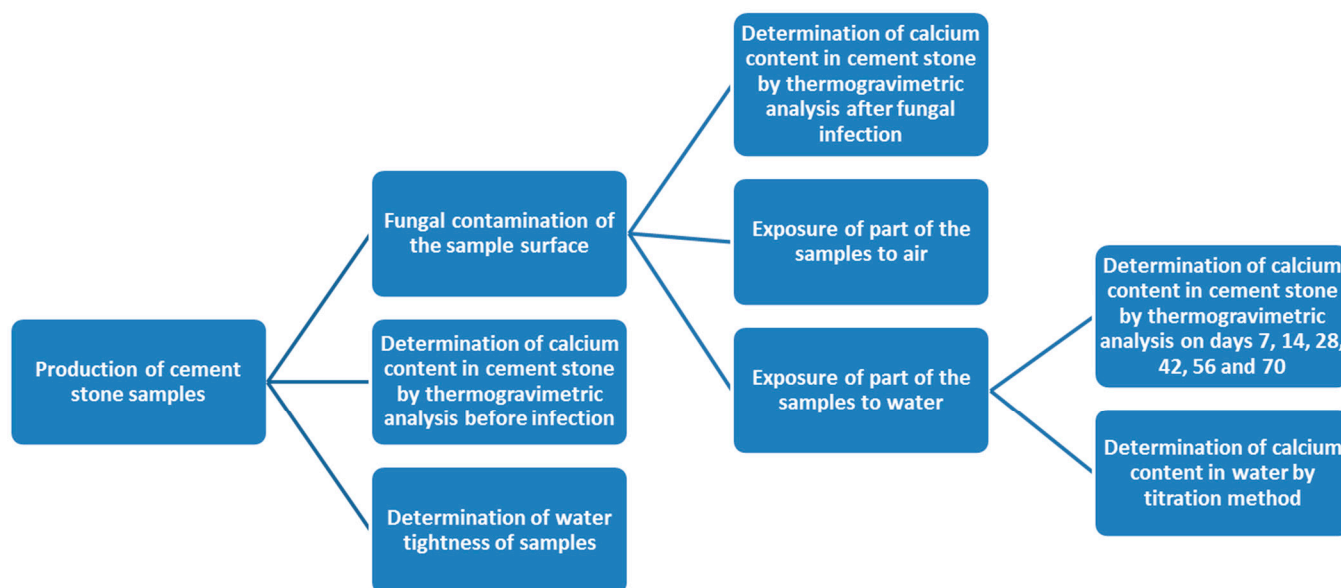


Figure 2. The scheme of the experiment.

The mass transfer characteristics (mass flow density of calcium hydroxide q , mass conductivity coefficient k and mass transfer coefficient β) were calculated using the following equations:

$$q = \frac{\Delta C_{liq}}{S \times \tau} \quad (2)$$

where ΔC_{liq} is the mass of the substance transferred to the liquid medium from the cement stone, [kg]; S is the area of the corroded sample surface, [m²]; τ is process time, [sec].

$$k = \frac{q}{\rho_0 \times \frac{dC}{dx}} \quad (3)$$

where q is mass flow density due to chemical reactions, [kg/(m²·s)]; ρ_0 is density of the solid phase, [kg/m³]; $\frac{dC}{dx}$ is concentration gradient of the transferred component over the sample thickness, [kg/m].

$$\beta = \frac{q}{\Delta C} \quad (4)$$

where q is diffusion flow, [kg/(m²·s)]; ΔC is concentration difference, [kg/m³].

3. Results

Using the “wet spot” method, it was determined that the water resistance of sample with an addition of 0.5% calcium stearate by weight of cement corresponds to class W6, while the addition of 1% corresponds to class W10.

The results of the calcium cation content determination in the liquid phase, using the method of complexometric titration, are presented in Figure 3.

Figure 3 shows that after 70 days of immersion in water, there is no change in the concentration of calcium ions in the liquid phase, as indicated by the plateaus on the kinetic curves. This state is regarded as close to equilibrium, where the calcium content in the cement stone remains constant. Consequently, the variation in calcium content along the thickness of the cement stone for a group of samples was assessed using a derivatograph over the 70-day period. The profiles of calcium hydroxide concentrations along the thickness of the cement stone are presented in Figure 4. These concentration

profiles clearly illustrate the changes in the intensity of mass transfer processes in the cement stone over time.

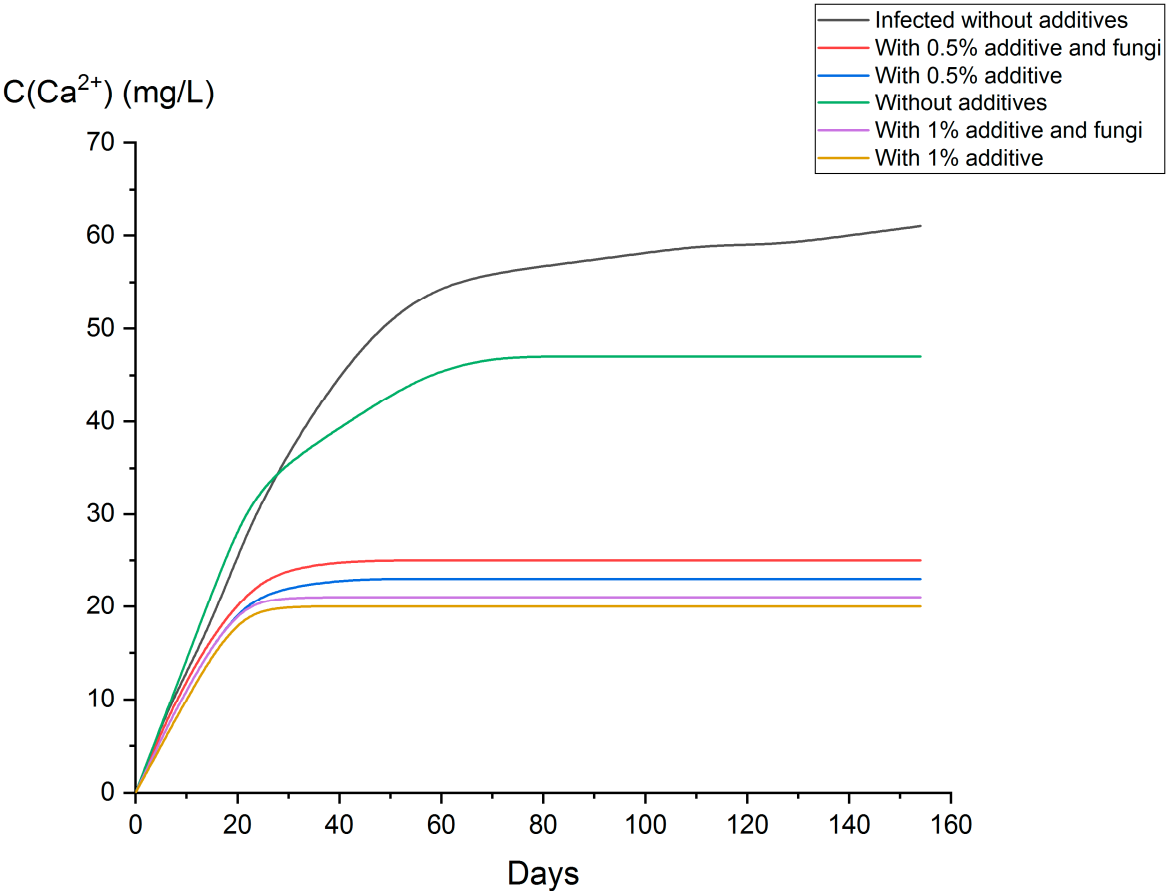


Figure 3. Kinetics of calcium leaching with water from cement stone samples with additives of calcium stearate and under the action of *Aspergillus niger* fungi.

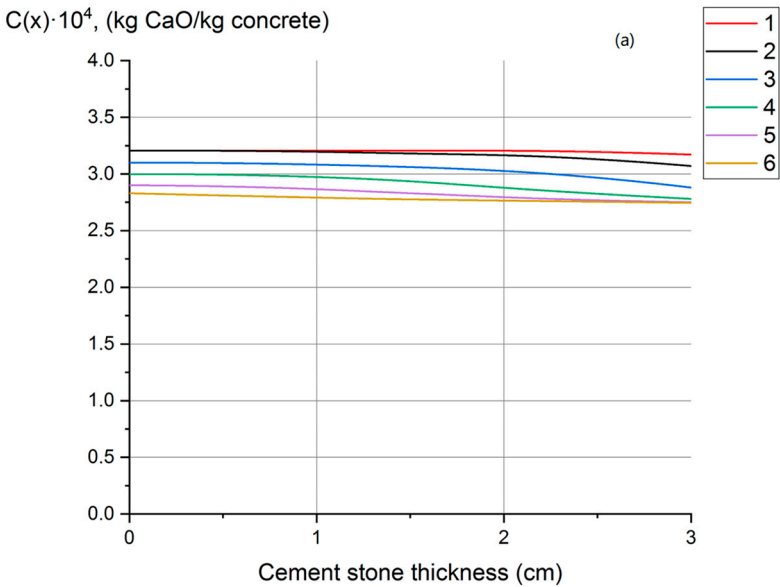


Figure 4. Cont.

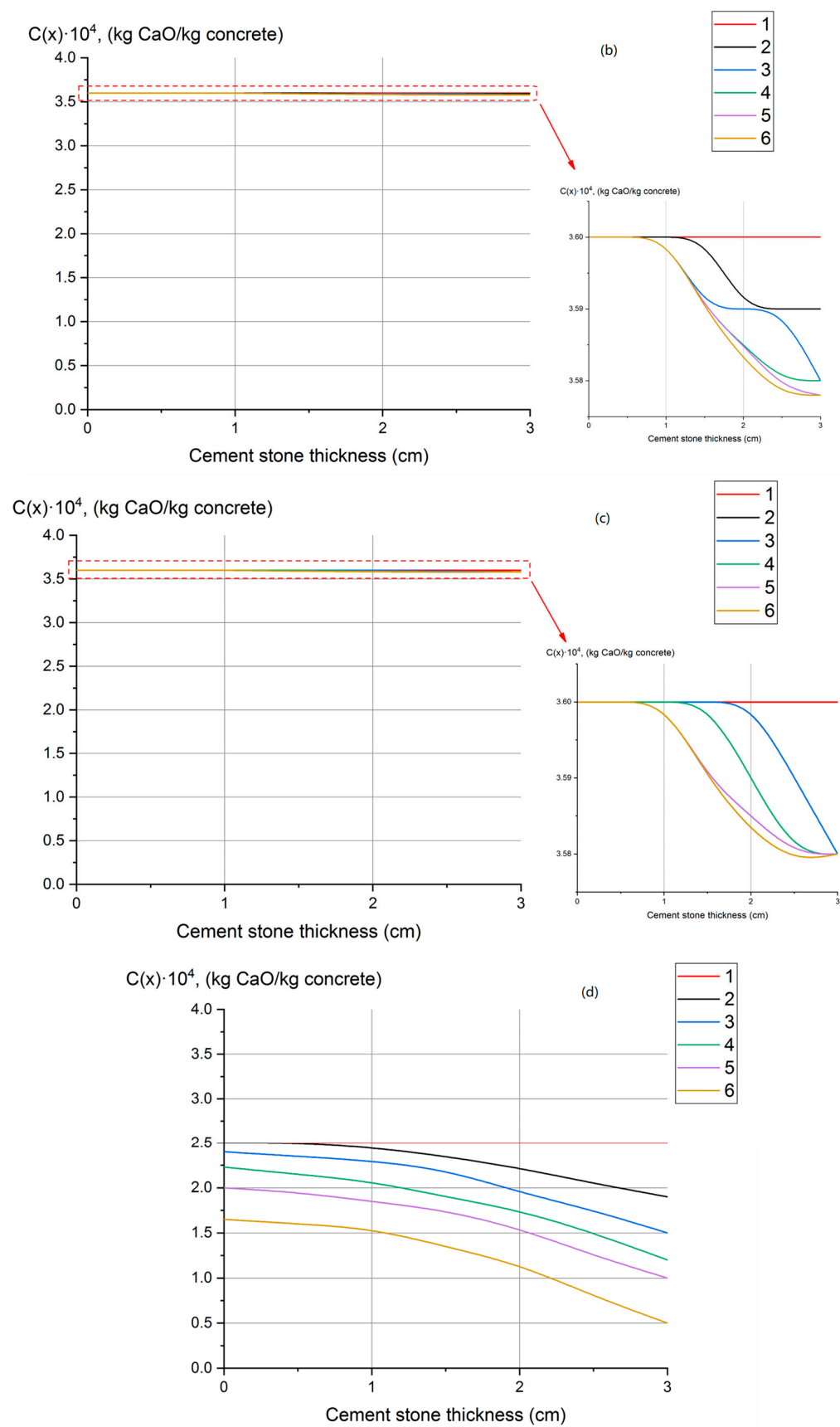


Figure 4. Cont.

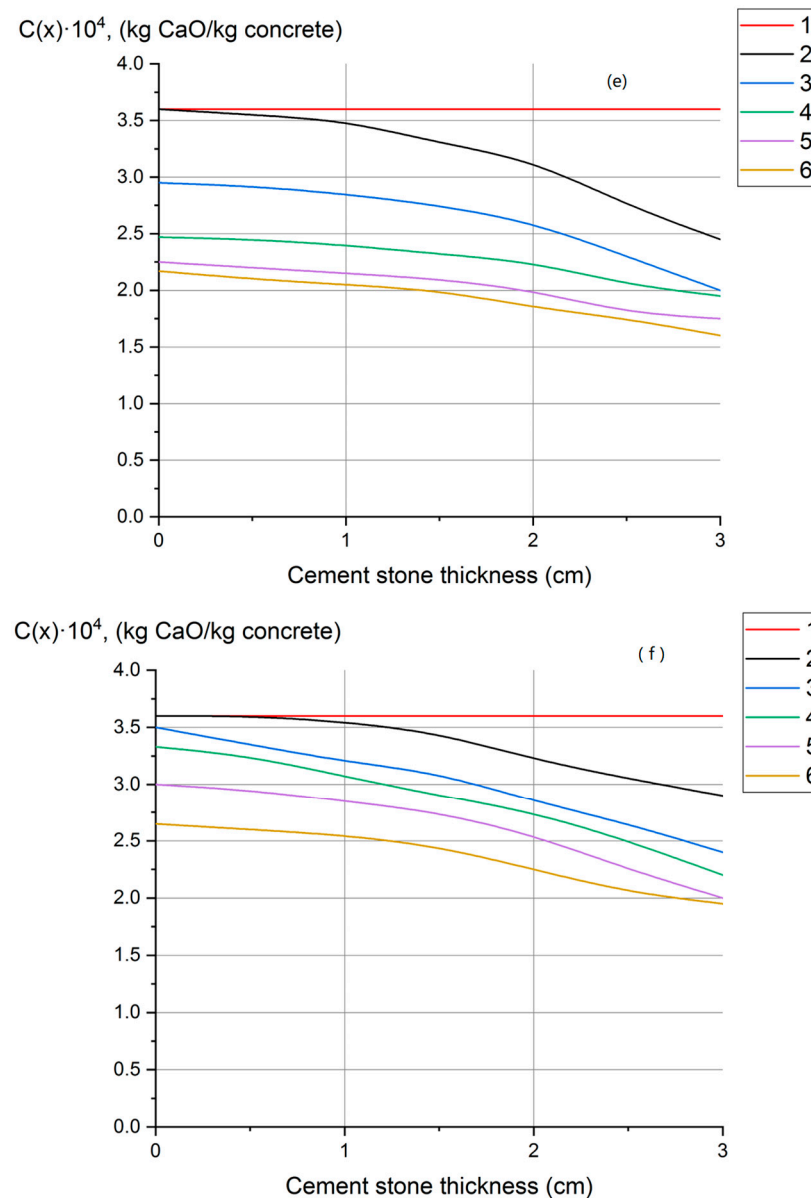


Figure 4. Profiles of Ca(OH)_2 concentrations within the thickness of cement stone samples at various time points: 1–0 d.; 2–14 d.; 3–28 d.; 4–42 d.; 5–56 d.; 6–70 d. (a) cement stone without additives infected with *Aspergillus niger* fungi is exposed to the air; (b) cement stone with 0.5 wt.% of calcium stearate infected with *Aspergillus niger* fungi is exposed to the air; (c) cement stone with 1 wt.% of calcium stearate infected with *Aspergillus niger* fungi is exposed to the air; (d) cement stone without additives infected with *Aspergillus niger* fungi is exposed to the water; (e) cement stone with 0.5 wt.% of calcium stearate infected with *Aspergillus niger* fungi is exposed to the water; (f) cement stone with 1 wt.% of calcium stearate infected with *Aspergillus niger* fungi is exposed to the water.

Figure 4a illustrates the change in calcium content across the thickness of the cement stone due to the action of waste products from *Aspergillus niger* fungi. After the samples were biofouled for 28 days, the initial concentration of calcium (line 1) in the surface layer decreased. Over time, the curves in Figure 4a become less steep, indicating a reduction in the rate of destruction of calcium-containing phases. Curves 5 and 6 converge at the same point on the outer surface of the cement stone, which signifies the cessation of chemical interactions between the calcium-containing phases in the cement stone and the fungal substances in the surface layer of the samples.

The initial calcium concentration in the samples with calcium stearate additives (Figure 4b,c) is uniform throughout the thickness. This indicates the absence of fungal

action on the cement stone prior to their accumulation in the biofilm on the surface. This initial concentration is particularly significant, as it correlates with the formation of numerous calcium-containing phases in the cement stone during hydration and hardening [56]. Over time, there is only a slight change in calcium concentration in the surface layer. The processes occurring on the surface cease after 28 days, as evidenced by lines 3–6 converging at the same point.

In samples immersed in water, calcium leaching occurs more intensively. The curves in Figure 4e,f, obtained for cement stone with calcium stearate additives, vary throughout the thickness, in contrast to those in Figure 4b,c, but exhibit a gentler appearance. After 28 days of exposure to the liquid medium, the lines converge at a point on the surface of the cement stone. This indicates the cessation of chemical interaction in this area of the samples. However, it does not completely end within the cement stone, as some fungal waste products have penetrated into the pore structure with the water.

The concentration gradients of the transferred component are determined from the obtained curves for calculating the mass transfer characteristics (Equations (2)–(4)). The results of these calculations are presented in Figures 5 and 6.

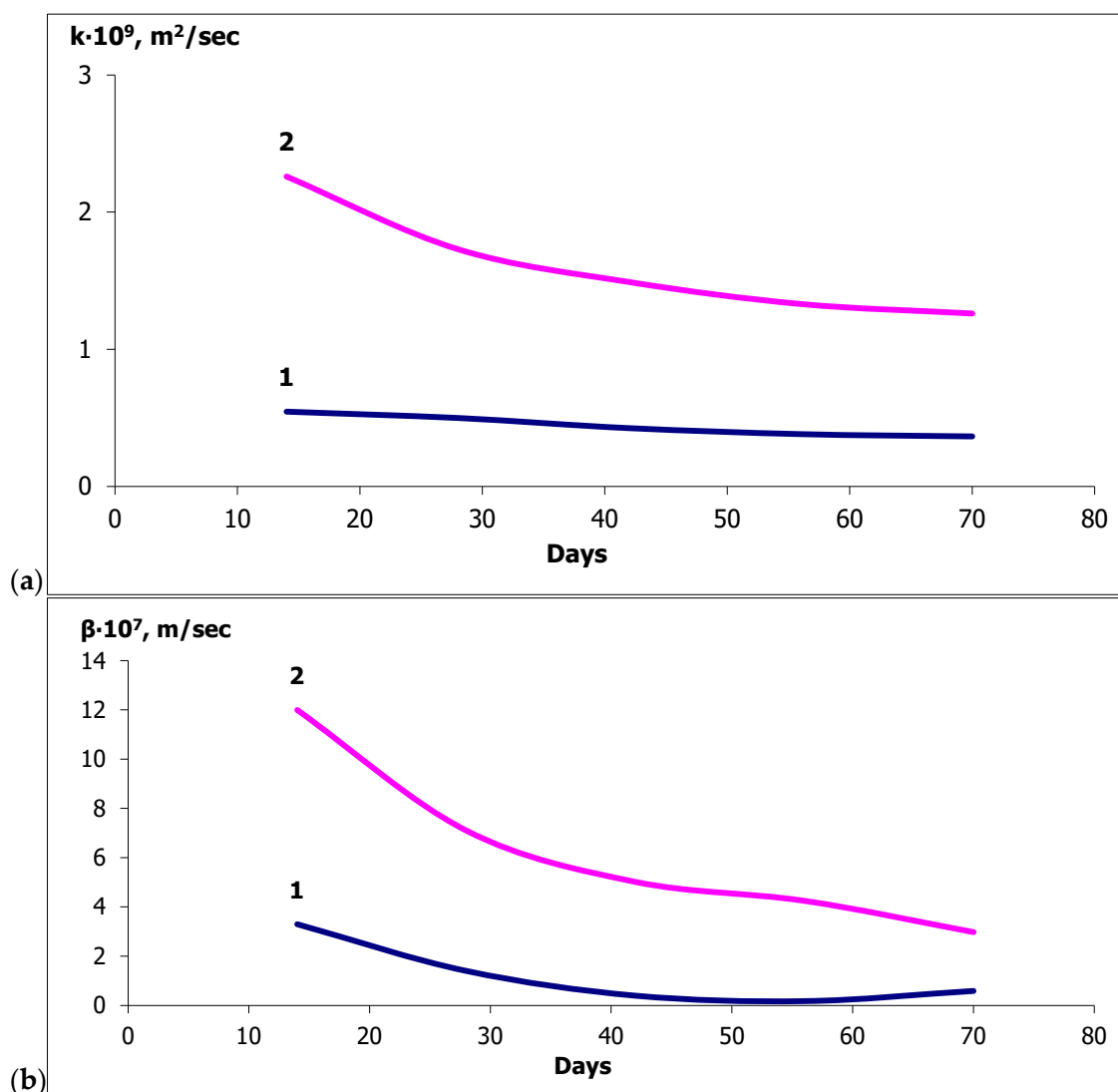


Figure 5. Changes in the values of the coefficients of mass conductivity (a) and mass transfer (b) during corrosion of cement stone samples without additives in water: (1) not infected; (2) infected with fungi.

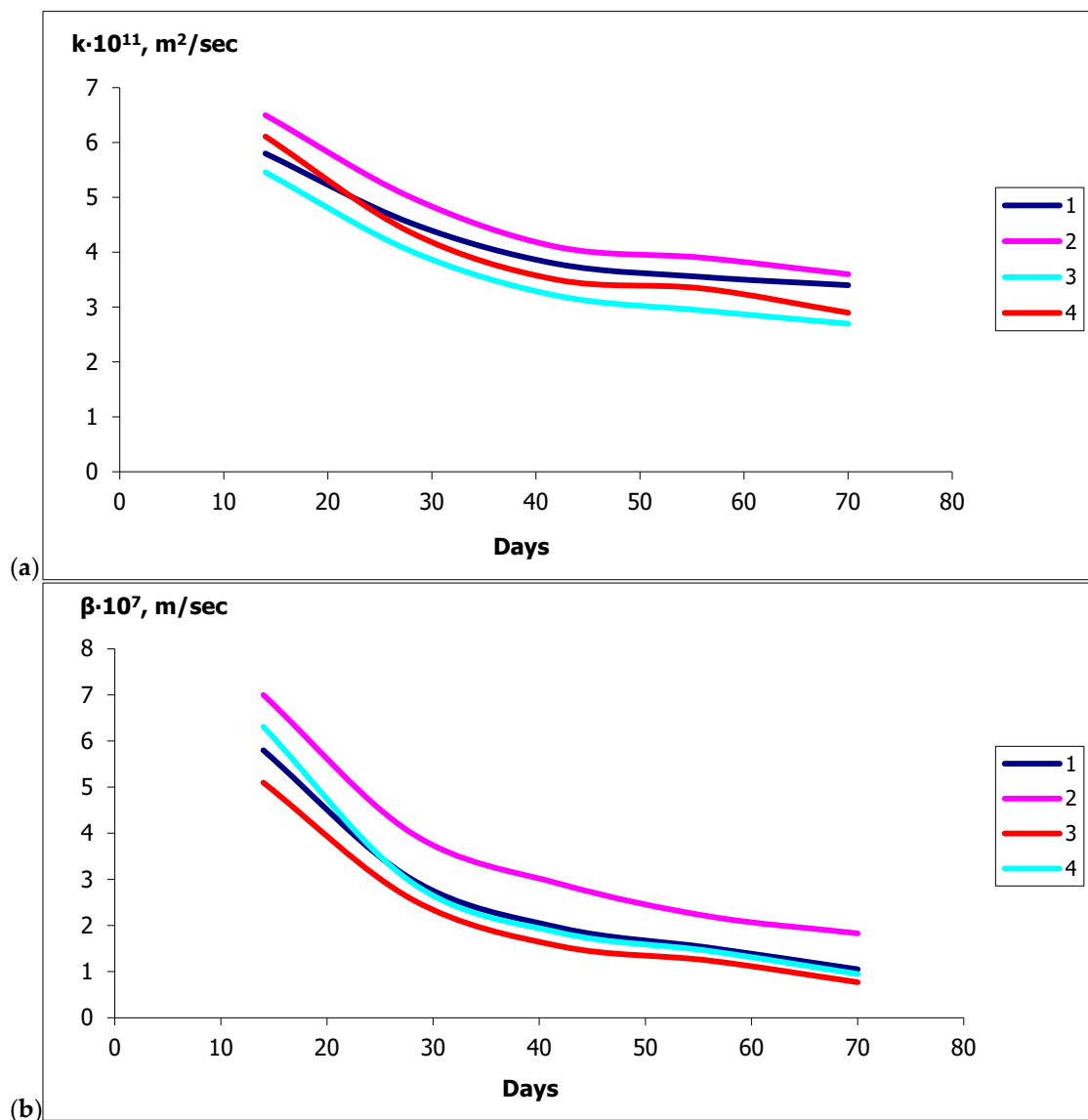


Figure 6. Changes in the values of the coefficients of mass conductivity (a) and mass transfer (b) during corrosion of hydrophobized cement stone samples: (1) with 0.5% calcium stearate, not infected with fungi; (2) with 0.5% calcium stearate, infected with fungi; (3) with 1% calcium stearate, not infected with fungi; (4) with 1% calcium stearate, infected with fungi.

Biogenic acids released by *Aspergillus niger* fungi enhance the leaching of calcium hydroxide from the cement stone structure. This is evidenced by an exponential change in the values of the mass conductivity coefficient and a steeper slope of the curve compared to the uninfected sample (Figure 5a). The mass transfer coefficient under fungal action on cement stone also exhibits a more pronounced change (Figure 5b).

A significant slowdown in mass transfer within cement stone containing a hydrophobic additive is indicated by the gentler appearance of the curves and the low values of the coefficients of mass conductivity and mass transfer (Figure 6). Additionally, the level of corrosion processes is markedly reduced.

4. Discussion

The presence of fungi on the concrete surface facilitates the leaching of calcium from the cement stone structure (Figure 3). This acceleration of the process is attributed to the organic acids released by fungi, which readily engage in chemical reactions with the

calcium-containing components of the cement stone. This leads to their destruction and the extraction of calcium from the structure. This mechanism of concrete destruction by biogenic organic acids is described in the works [57–59].

It is known that calcium stearate gives the surface and porous structure of concrete hydrophobicity, which is reflected in a decrease in water absorption and an increase in the water contact angle, as established in [60]. Calcium stearate also fills pores and capillaries during the formation of the structure, as demonstrated in studies [41,54]. This property explains the prevention of liquid medium penetration deep into the cement stone. Consequently, equilibrium in the system is established more quickly with a reduced amount of reacted calcium, as illustrated in Figure 3. It can therefore be concluded that, in this case, calcium cations are leached by water solely from the surface of the cement stone.

Biogenic acids released by fungal microorganisms infiltrate the cement stone with water. They chemically interact with free calcium hydroxide in the pore fluid and with calcium-containing components of the cement stone [56]. As a result, the calcium content in the cement stone decreases (Figure 4).

Figure 3 shows that calcium stearate additives facilitate the establishment of equilibrium in the cement stone structure and reduce the leaching of calcium hydroxide from the structure by microorganisms. In cement stone containing the hydrophobic additive, only a slight change in calcium content is observed, and this occurs solely in the surface layer.

Recommendations have been developed for hydrophobizing concrete using calcium stearate additives in the context of studying concrete degradation in chloride media of varying degrees of aggressiveness. It was shown in [61,62] that the introduction of 0.3–0.7 wt.% of calcium stearate in the cement mixture effectively prevents the effects of a chloride medium of low and medium aggressiveness (using 0.6–2% MgCl_2 solutions as an example). It is well-established that this hydrophobic additive enhances the strength properties of concrete and mitigates the corrosive effects of chlorides [39,40,54,55,60]. Previous studies on the degree of damage to concrete by microorganisms indicate the importance of implementing measures to protect concrete from biofouling [63,64]. It was established in [63] that irreversible processes of corrosion destruction of reinforced concrete in conditions of fungal corrosion will begin in 2.5 years, in conditions of bacterial corrosion—in 5.5 years. In the study [64], the authors studied the condition of a reinforced concrete wall that has been biodegradable for 20 years. It was found that during this time, the substances released by microorganisms reached the surface of the steel reinforcement through a 5 cm thick concrete coating and caused corrosion of the steel.

Therefore, calcium stearate was selected to prevent the biodegradation of concrete. It was found that the addition of calcium stearate reduces the extent of fungal damage to concrete by threefold, judging by the change in water absorption, porosity, and compressive strength [59,65,66]. These results are consistent with the patterns of mass transfer in cement stone observed during this study. These findings are consistent with the patterns of mass transfer in cement stone observed in this study.

It was shown in [25,65,67] that in concrete samples containing a hydrophobic additive, a more highly crystalline structure with a greater abundance of calcium hydrosilicates is formed during hydration and hardening. The profiles of calcium concentrations in the cement stone shown in Figure 3 indicate increased calcium content within the structure.

Concentration profiles are used to visually represent the rate of change in the calcium content in cement stone over time [68,69], as well as to quantify the intensity of mass transfer of gases [70] and liquids [71] at various points in time. The concentration gradients obtained from these profiles are used to calculate the service life of concrete. They are involved in mathematical models describing heat and mass transfer during concrete hardening [72], simultaneous transfer of chlorides and moisture in concrete [73], chloride corrosion under

mechanical stress [74], corrosion in seawater [75,76], chloride corrosion under the action of deicing salts [77]. Such models are based on the diffusion equations of aggressive substances [78–80], such as chlorides [81,82] and carbon dioxide [83,84], in concrete. They are used to estimate the time frame for achieving maximum concentrations of aggressive components in concrete. This approach is essential for assessing the periods leading to corrosion damage of steel reinforcement in concrete during the accumulation of a threshold value of chlorides [85], as a result of cracking of the concrete coating [86], under the influence of corrosion reactions on the properties of concrete [87].

Many mathematical models predict the critical condition of concrete based on the concentration of calcium hydroxide. The amount of calcium hydroxide in the cement stone influences its strength and, consequently, the overall strength of the concrete [88]. It was shown in [89] that the additional introduction of calcium hydroxide into the cement mixture increases the compressive strength of super sulfated cement. In studies [90,91], it was found that the introduction of mineral additives into concrete increases the content of calcium hydroxide, which contributes to the Pozzolan reaction. As a result, the amount of C-S-H gel formed during hydration increases, which makes the structure more compact and increases strength.

Studying mass transfer in cement stone during the initial stage of exposure to aggressive environments is essential for determining the timing of equilibrium state establishment in the system. For this condition, equilibrium concentrations of calcium hydroxide and mass transfer parameters are calculated, which are then incorporated into equations for determining the calcium content in concrete at any given time. This approach allows us to determine the time periods during which the calcium concentration in concrete can decrease to levels corresponding to the degradation of key components responsible for maintaining strength [69,92]. In [93], this approach was used to assess the degree of neutralization of concrete based on the results of existing environmental impact tests and accelerated carbonation tests. In [94], the authors calculated the leaching of calcium from cement-asphalt mortar to establish the preservation of structural integrity under dynamic loads. In [95], numerical modeling shows that the dissolution of calcium hydroxide leads to a significant increase in the porosity of hydrated cement paste, which negatively affects the properties of the material.

The scientific school of the authors has developed mathematical models for describing and predicting the development of corrosion processes in concrete during leaching [69,96,97], as well as when exposed to salts and acids [92,98,99] and under microbiological action [100,101]. The work presented by the authors is a continuation of research on the kinetics and dynamics of corrosive mass transfer. The insights gained regarding the intensity of corrosion processes in cement stone are consistent with findings from other researchers and enhance the overall understanding of corrosion phenomena in concrete. The mass transfer indicators established in this study can be integrated into existing mathematical models to predict the service life of concrete or to develop and validate new equations that describe mass transfer processes.

For example, the durability of concrete exposed to *Aspergillus niger* fungi during humidification will be predicted using a mathematical model of microbiological corrosion developed by our scientific school (Equation (5)). This model is based on differential equations of mass conductivity and is formulated from the boundary value problem of mass transfer, as shown in [69,97,100]. The mathematical model is employed to predict the distribution of “free” calcium hydroxide throughout the thickness of the concrete under fungal action.

$$\begin{aligned}
Z(\bar{x}, Fo_m) = Z_\delta & \frac{Bi_m \bar{x} + 1}{Bi_m + 1} + \frac{Bi_m \bar{x} + 1}{Bi_m + 1} \int_0^1 Po_m^*(\xi)(1 - \xi) d\xi \\
& + 2Bi_m \sum_{m=1}^{\infty} \frac{Bi_m \sin(\mu_m \bar{x}) + \mu_m \cos(\mu_m \bar{x})}{(Bi_m^2 + Bi_m + \mu_m^2) \cos \mu_m} \exp(-\mu_m^2 Fo_m) \\
& \cdot \left[\frac{Z_\delta}{\mu_m} - \int_0^1 Z_0(\xi) \sin[\mu_m(1 - \xi)] d\xi \right] \\
& - 2Bi_m \sum_{m=1}^{\infty} \frac{Bi_m \sin(\mu_m \bar{x}) + \mu_m \cos(\mu_m \bar{x})}{\mu_m^2 (Bi_m^2 + Bi_m + \mu_m^2) \cos \mu_m} \exp(-\mu_m^2 Fo_m) \\
& \cdot \int_0^1 Po_m^*(\xi) \sin[\mu_m(1 - \xi)] d\xi
\end{aligned} \quad (5)$$

where $Z(\bar{x}, Fo_m)$ is the dimensionless concentration of calcium hydroxide in the pore structure of concrete; $\bar{x} = \frac{x}{\delta}$ is the dimensionless coordinate; x is coordinate, [m]; δ is concrete thickness, [m]; $Fo_m = \frac{k\tau}{\delta^2}$ is the Fourier mass transfer criterion; k is the mass conductivity coefficient of calcium hydroxide in concrete, [m²/s]; τ is time, [s]; $Bi_m = \frac{\beta\delta}{k}$ is the Bio mass transfer criterion; β is the mass transfer coefficient of calcium hydroxide in a liquid medium, [m/sec]; Z_δ is the dimensionless concentration of calcium hydroxide over the thickness of a micro-section of cement stone; ξ is the coordinate of integration in the range $0 \leq \xi \leq \bar{x}$; μ_m is the root of the characteristic equation $tg\mu_m = \frac{\mu_m \cdot Bi_m}{\mu_m^2 - Bi_m \cdot K_m}$; $Po_m^* = \frac{q_v \delta^2}{C_0 k \rho_{con}}$ is the modified Pomerantsev mass transfer criterion; q_v is the power of volumetric release (absorption) of calcium hydroxide due to phase and chemical transformations, [kg Ca(OH)₂/(m³·s)]; C_0 is the initial uniformly distributed value of the mass content of free calcium hydroxide in the pore structure of concrete by thickness, [kg Ca(OH)₂/kg concrete]; ρ_{con} is concrete density, [kg/m³].

Figure 7 illustrates the profiles of calcium hydroxide content in concrete exposed to fungal activity under humidification conditions for different predicted exposure periods.

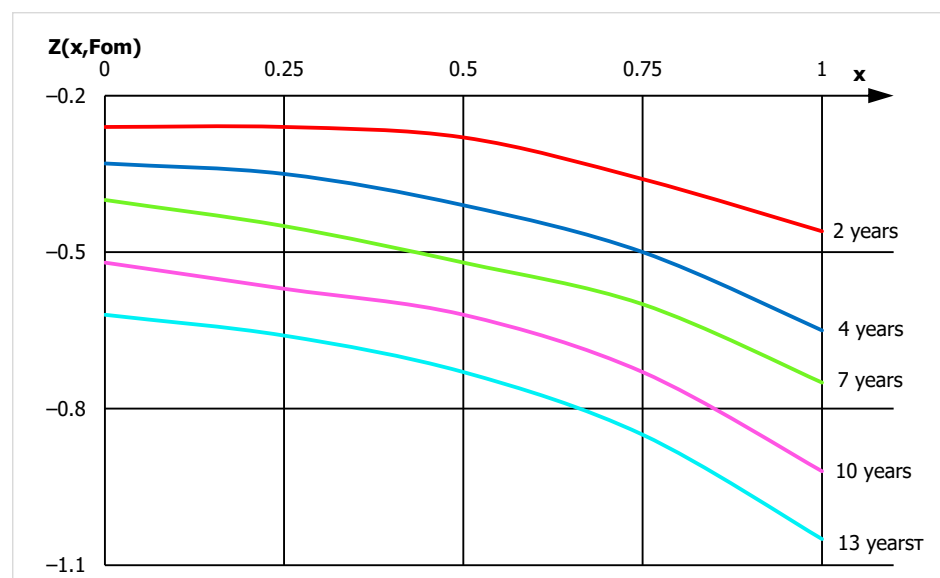
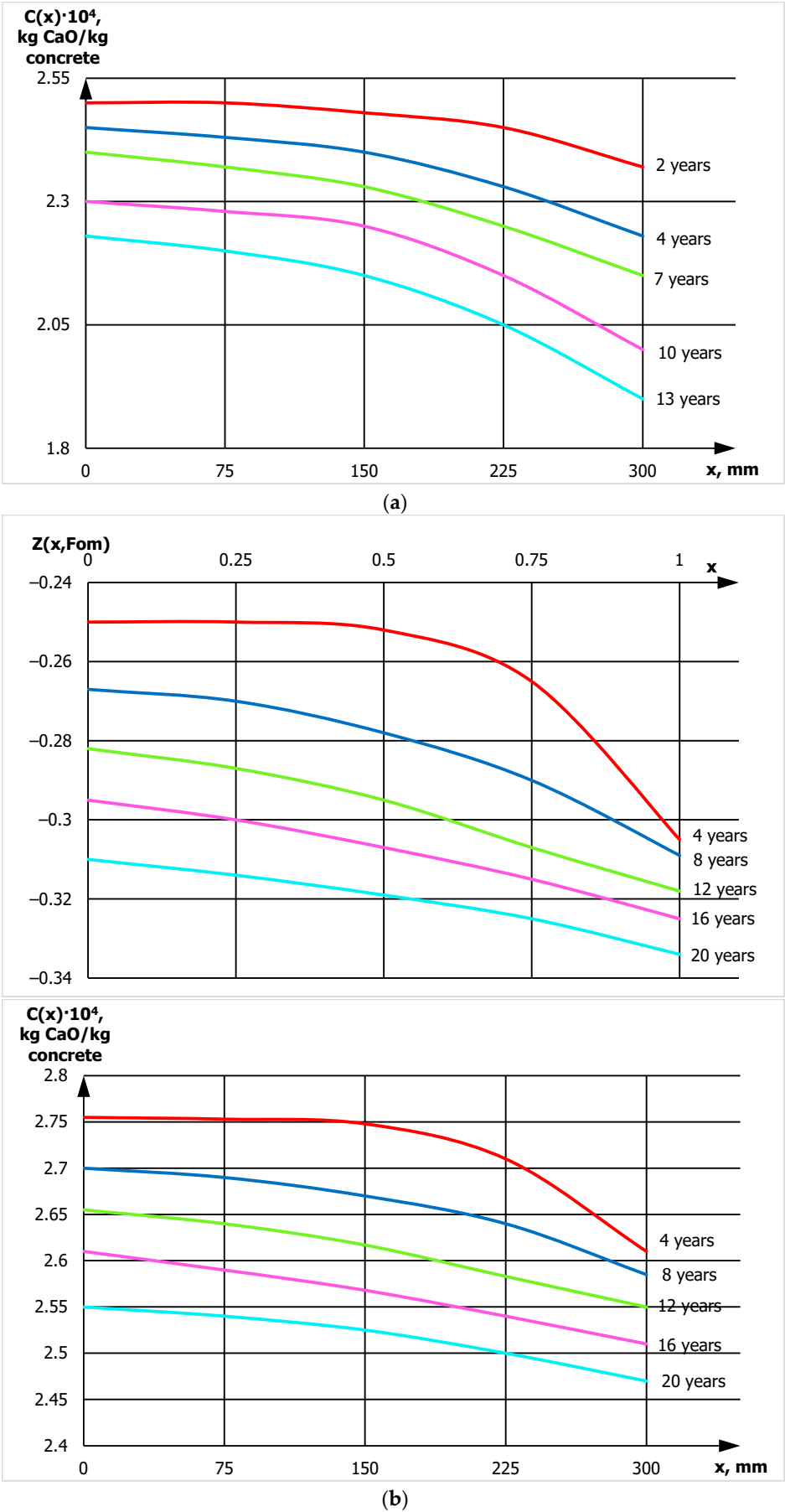


Figure 7. Cont.



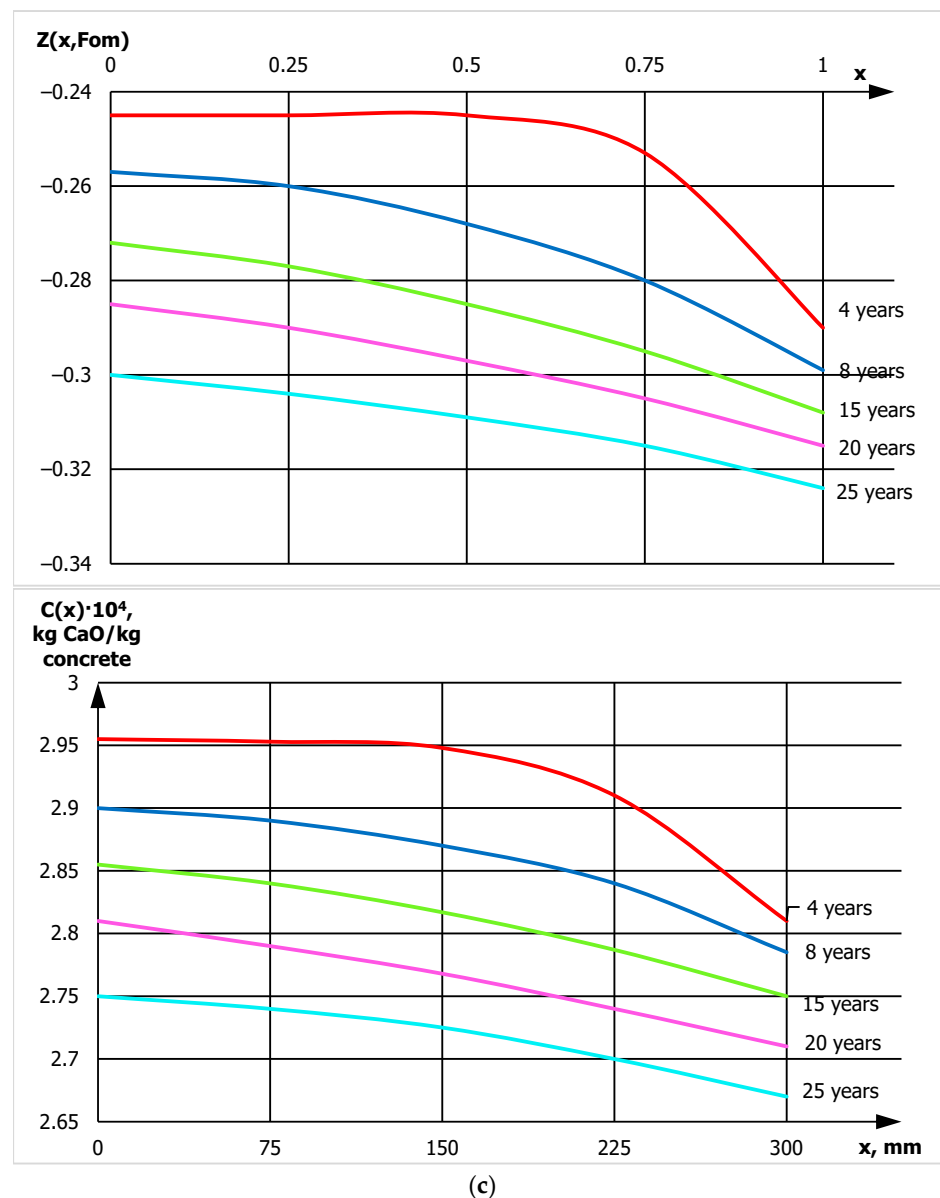


Figure 7. The profiles of the distribution of calcium hydroxide content over the thickness of concrete (a) without additives; (b) with the addition of calcium stearate 0.5 wt.%; (c) with the addition of calcium stearate 1 wt.% at different stages of operation during fungal destruction and humidification in dimensionless form (Z) and dimensional value (C).

The long-term forecast indicates that the primary damage from fungal exposure will be concentrated on the concrete surface, although destructive processes will also initiate along its thickness. In concrete with calcium stearate additives, the most intense leaching of calcium from the surface layer is expected to occur during the first four years of bio-damage, after which the processes will slow down, following a pattern similar to what was experimentally observed during the initial stages of corrosion (Figure 4).

In concrete without additives, the decomposition of calcium-containing phases in the surface layer will begin after 4 years of fungal degradation under moist conditions and will spread throughout the entire thickness by the 10 years of concrete operation under these conditions. Based on the profiles in Figure 7a, it can be anticipated that the integrity of the concrete structure will be compromised due to the destruction of bonds between components and the loss of strength approximately 15 years after biofouling.

The period for establishing the critical state of decomposition of the hydrosilicate phase in the structure of hydrophobic concrete is estimated to take 12 to 15 years (Figure 7b,c). It can be inferred that during this time, substances released by the fungi will penetrate through the pore structure of the concrete coating to reach the surface of the reinforcement. The development of destructive processes and a decline in concrete strength will continue after 20 to 25 years of operation, ultimately reaching critical levels within 30 to 35 years, at which point damage will extend throughout the entire volume of the concrete.

The proposed addition of calcium stearate in an amount of 0.5% by weight of cement effectively prevents biofouling of concrete and significantly slows down the mass transfer processes within the cement stone.

Thus, volumetric hydrophobization of concrete with calcium stearate enhances the corrosion resistance and extends service life of concrete products operated under conditions conducive to surface biofouling by fungal microorganisms.

5. Conclusions

The effect of the addition of calcium stearate on the kinetics of mass transfer in cement stone and concrete under the influence of *Aspergillus niger* fungi has been studied. The established changes in mass transfer parameters over time allow us to assess the degree of deceleration of fungal corrosion when calcium stearate is added to the cement mixture. Using a mathematical model of biocorrosion, the time frame for the decomposition of calcium-containing components of cement stone was established, indicating the critical values at which concrete destruction occurs.

The main conclusions of the study:

- In concrete of waterproofness classes W6–W10, the degree of decomposition of calcium-containing phases by fungal microorganisms and the removal of calcium from the structure is reduced by approximately 3 times compared to conventional concrete.
- Profiles of calcium hydroxide concentrations throughout the thickness of concrete have been obtained, indicating that fungal destruction of hydrophobic concrete primarily occurs at the surface. In ordinary cement concrete, the decomposition and leaching of calcium hydroxide occurs throughout the thickness.
- The values of the mass conductivity coefficients for fungal destruction of ordinary concrete during moistening and for concrete with calcium stearate additives differ by two orders of magnitude: 10^{-9} and 10^{-11} [m²/s], respectively, indicating a significant slowdown in mass transfer.
- Using a mathematical model of concrete biocorrosion, it is predicted that destructive processes in concrete without additives will lead to dangerous conditions after 15 years of fungal exposure, while for hydrophobic concrete, this will occur 25–30 years.

The results of the study will aid in developing comprehensive measures to prevent the corrosion and destruction of reinforced concrete products under multifactorial influences, including temperature fluctuations, exposure to chloride and sulfate media, and biofouling. It is advisable to continue researching the proposed recommendations for incorporating calcium stearate additives into concrete to enhance its resistance to biofouling and to assess the degree of damage caused by other organisms, such as bacteria, fungi of various species, algae, lichens, and mosses.

Author Contributions: Conceptualization, V.S.K. and K.B.S.; methodology, V.S.K., A.A.G. and D.G.N.; validation, V.S.K. and K.B.S.; formal analysis, V.S.K. and D.G.N.; investigation, V.S.K., A.A.G. and D.G.N.; resources, V.S.K.; data curation, V.S.K. and A.A.G.; writing—original draft preparation, V.S.K.; writing—review and editing, V.S.K.; visualization, V.S.K. and D.G.N.; supervision, V.S.K.; project administration, V.S.K. and K.B.S. All authors have read and agreed to the published version of the manuscript.

Funding: This research received no external funding.

Data Availability Statement: The original contributions presented in this study are included in the article. Further inquiries can be directed to the corresponding author.

Conflicts of Interest: The authors declare no conflicts of interest.

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