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Protection of construction materials based on acrylates from biodeterioration

Защита строительных материалов на основе акрилатов от биоповреждений

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Key words: construction materials based on acrylates; funginertness; microfungi; enzyme activity; biodeterioration of buildings and construction; fungicidal additives; protection from biodeterioration

Ключевые слова: строительные материалы на основе акрилатов; грибостойкость; микроскопические грибы; активность ферментов; биоповреждение зданий и сооружений; фунгицидные присадки; защита от биодеградации

Abstract. The article is dedicated to the funginertness of different construction materials based on acrylates: emulsions Lakroten E-21, Lakroten E-31, Latacryl ZM-1, and Latacryl AF, as well as metacrylate, metacrylate copolymer with metacrylic acid, and n-chlorophenylmetacrylate acrylic glasses. All studied materials, except the Latacryl ZM-1 and copolymer acrylic glasses, demonstrated susceptibility to biodeterioration by microfungi. Multi-compound acrylic compositions appeared to exhibit the emergence, i.e. their funginertness cannot be predicted based on stability of their individual components. Fungal exo-oxidoreductases (catalase, peroxidase) were defined to contribute into the biodegradation processes in construction materials based on acrylates by micromycetes. The biocides Nuosept 78 and Rosima 243 demonstrated the ability to suppress exo-catalase and exo-peroxidase activity (exo-catalase and exo-peroxidase participate in the biodeterioration of the studied materials) in the fungus Aspergillus terreus; thus, they can be recommended for use as means of bioprotection. The non-fungi-resistant acrylic materials were protected from biodeterioration in a targeted and scientifically-grounded way by the introduction of the abovementioned biocidal additives into their compositions. The bioprotection is based on biochemical aspects of biodestructive microfungal activity.

Аннотация. Исследована грибостойкость ряда рецептур строительных материалов на основе акрилатов: эмульсий Лакротэн Э-21, Лакротэн Э-31, Латакрил ЗМ-1, Латакрил АФ и органических стекол на основе метилметакрилата и сополимеров метилметакрилата с метакриловой кислотой и n-хлорфенилметакрилатом. Показано, что все исследованы материалы, за исключением акриловой эмульсии Латакрил ЗМ-1 и сополимерных органических стекол, способны подвергаться биодеструкции микроскопическими грибами. Выявлено, что многокомпонентные акриловые композиции обладают свойством эмерджентности, и их грибостойкость не может быть спрогнозирована, исходя из устойчивости отдельных компонентов рецептур. Установлено участие грибных экзооксидоредуктаз (каталазы, пероксидазы) в процессах биодеградации микромицетами строительных композиций на основе акрилатов. Показано, что биоциды Nuosept 78 и Rosima 243 способны подавлять у гриба Aspergillus terreus активность экзокаталазы и экзопероксидазы, участвующих в деструкционном процессе изучаемых материалов, и, следовательно, могут быть рекомендованы в качестве средств защиты от биоповреждений. Осуществлена научно обоснованная и целенаправленная защита негрибостойких материалов на основе акрилатов от биоповреждений путем введения вышеуказанных биоцидных добавок в состав композиций, основанная на учете биохимических аспектов жизнедеятельности микроскопических грибов-биодеструкторов.

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1. Introduction

Construction materials based on acrylates (acrylicglasses, paints and lacquers) with increased thermostability, high water and atmospheric resistance are broadly used in various industrial fields and in construction [1–5]. Particularly, acrylic glasses are used as various elements of interior of buildings and structures, advertising materials, illuminating devices, vehicle glazing, instrumentation; acrylic emulsions are used for concrete water-proofing, drenching of porous structural materials, as the basis for paints and lacquers for interior and exterior works, for preparation of water putties, glues, fillers [6–8].

In the course of transportation, storage and use these materials are exposed to the destructive effects of microorganisms, which use them as a nutrient source [9–13]. Microfungi are the most active destructors of different synthetic materials, as they have powerful and labil enzyme systems (oxidoreductases, liases, hydrolases, etc.) enabling to metabolise different polymers, including acrylic-based polymers [14–18]. Biodeterioration of materials is accompanied by change of their decorative, physical and chemical properties, including cracking and layering of coatings, cavitation, foreign odour, deliquation, settling, jellification, viscosity loss, and colour change, which negatively affects appearance and functioning of building constructions [19–22].

Currently, there are no biological resistance (fungal resistance) polymeric acrylate-base compositions [23]. The assortment of acrylic materials is being constantly updated. The new formulations will have different resistance to activity of microorganisms, in particular, microfungi, depending on their chemical composition [24, 25]. The non-fungi-resistant compositions need to be protected from biodeterioration caused by microfungi. There are various methods of material/product bioprotection (mechanical, physical, chemical), with the chemical methods being the most wide-spread. The chemical method features an introduction of special biocidal additives into the material/product composition [26, 27]. Unfortunately, now such additives are selected by trial-and-error method, based only on the information about their fungicidal properties. However, task-orientated and systematic additives selection based on studying the mechanisms of the suppression of the activity of fungal aggressive metabolites, in particular, enzymes taking part in construction materials biodeterioration is required.

In this respect, the purpose of this study is to evaluate funginertness of different construction materials based on acrylates and to develop a targeted and systematic approach to protection of non-fungi-resistant materials from biodeterioration based on biochemical aspects of microorganisms' activity. For this purpose the following objectives have been set:

– Evaluation of the degree of degradation of constructional materials based on acrylates by microscopic fungi

– Revealing the effect of components of acrylic compositions on the degree of their funginertness

– Identification of the most active mycodestructor of the research materials

– Protection of the test materials from microbiological deterioration based on used fungicides ability to suppress the activity of aggressive metabolites taking part in the biodeterioration of construction materials based on acrylates.

2. Methods

The following construction materials based on acrylates were used as the subject of this study: acrylic glasses based on metacrylate copolymer, metacrylicacid, and n-chlorophenylmetacrylate; acrylic emulsions used as film-forming basis for paints for interior and exterior works: Lakroten E-21 (butylacrylate-styrene-acrylicacid copolymer), Lakroten E-31 (butylacrylate-methylmetacrylate-metacrylicacid copolymer), Latacryl ZM-1 (butylacrylate-metacrylicacid-methylmetacrylate copolymer), and Latacryl AF (butylacrylate-methylmetacrylate-styrene-metacrylicacid copolymer).

The preparations Nuosept 78 and Rosima 243 were used as biocides. The active substances of the preparations Nuosept 78 and Rosima 243 are triazine and thyasolinone, respectively.

Funginertness of lacquers and paints, their components, and acrylic glasses was defined using the method described in the literature [28, 29]. This method allows estimating the natural resistance of the materials to microfungal activity, i.e. the possibility to use them by micromycetes as a nutrient source. The biodeterioration level of the polymer materials was studied both in respect to separate microfungal species, and to the mixed culture (mixture of fungi). Samples purified from external contaminations were infected with an aqueous suspension of fungal spores and allowed to stay for 28 days under conditions optimum for their growth. Tests of paints and their components were performed with the use of the following kinds of fungi: Aspergillus terreus, A. niger, A. ustus, Alternaria alternata, Fusarium moniliforme, Penicillium ochrochloron, P. chrysogenum, P. funiculosum, P. martensii, P. brevicompactum, Trichoderma viride, Gliocladium virens; acrylic glasses: Aspergillus niger, A. oryzae, A. terreus, Chaetomium globosum, Penicillium funiculosum, P. chrysogenum, P.cyclopium, Trichoderma viride, Paecilomyces variotii. These species of fungi are active biodegradants of different construction materials, including based on acrylates [30]. Given fungi are mandatory testing cultures, which are used in national and international standard test methods for the degree of polymers and paint-and-lacquer materials biodeterioration. Fungal resistance was estimated from the intensity of fungal development (0-6 points). A material was considered to be fungi-resistant, if it acquired 0–2 points.

Exo-enzyme activity was studied by cultivation of the fungus Aspergillus terreus, one of the most aggressive biodestructor of acrylate-based materials, on carbon-depleted liquid Czapek-Dox medium with the following composition (g/L): $NaNO_3$– 2.0, KH_2PO_4– 0.7, K_2HPO_4– 0.3, KCl – 0.5, $MgSO_4$*7H_2O – 0.5, $FeSO_4$*7H_2O – 0.01, sucrose – 1.5. The modified Czapek-Dox medium which additionally contained the acrylic emulsion Lakroten E-21 as a nutrient source was used in some experiments. The biocidal preparations were introduced to the media on the fourth day of the fungus cultivation.

The enzymatic activity was defined by spectrophotometric method (UV-mini 1240, Shimadzy, Australia): catalase activity – by consumption of H_2O_2 (λ=240 nm) [31], peroxidase activity – by oxidation of n–phenylenediamine (λ=535 nm) in presence of H_2O_2 [32]. The enzymatic activity was expressed in conventional units (c.u.). Change of optical density of the reaction mixture taken per 1 mg of protein in 1 minute was considered to be an activity unit for each of the abovementioned enzymes. The protein content in my celium and culture fluid were defined by Lowry – Folin method [33].

The statistical processing of results and the reliability assessment of differences in mean values were carried out according to the criteria of a student's test for the probability level of no less than 95% with the use of Microsoft Excel 2007 and Statistica 10.0 software.

3. Results and Discussion

The information on evaluation of funginertness of acrylic polymers (both to mixture of different cultures and to individual cultures) is represented in Table 1.

The obtained results demonstrated, that in the case influence of a mixture of fungal cultures, Latacryl ZM-1 revealed fungi-resistant characteristics; other tested acrylates did not demonstrate funginertness to micromycetic activity and can be consumed by them as a nutrient source. Comparison of destructive abilities of fungus strains posed an interesting task. In this regard, we evaluated funginertness of compositions to activity of various fungus cultures. The tested materials demonstrated different levels of resistance to particular fungus species. The fungi Aspergillus terreus, Fusarium moniliforme, Penicillium funiculosum appeared to be the most active biodegradants of acrylic emulsions.

Table 1. Evaluation of growth of individual fungus species and their mixture on the tested materials

	Growth level, points			
Material / Fungus species	Lakroten E-21	Lakroten E-31	Latacryl AF	Latacryl ZM-1
Mixture of fungus cultures	3	3	3	2
Aspergillus terreus	3	3	3	3
A. niger	2	2	2	2
A. ustus	1	2	2	2
Alternaria alternate	1	2	3	3
Fusarium moniliforme	2	3	2	3
Penicillium ochrochloron	1	1	2	2
P. chrysogenum	2	1	3	2
P. funiculosum	3	3	3	3
P. martensii	2	2	3	2
P. brevicompactum	2	2	2	3
Trichoderma viride	1	1	2	2
Gliocladium virens	1	1	2	2

The funginertness of acrylic glasses of various acrylate-based compositions was studied in the same way (Table 2).

Table 2. Evaluation of growth of individual fungus species and their mixture on the acrylic glasses

	Growth level, points		
Material / Fungus species	Polymethylmetacrylate	Polymethylmetacrylate – metacrylic acid – n-chlorophenyl-metacrylate copolymer	Polymethylmetacrylate – metacrylic acid copolymer
Mixture of fungus culture	4	2	2
Chaetomium globosum	2	3	4
Penicillium funiculosum	2	3	4
Aspergillus terreus	4	3	3
P. chrysogenum	3	3	4
P. cyclopium	3	3	4
Trichoderma viride	2	3	3
A. oryzae	4	3	3
A. niger	1	1	1
Paecilomyces variotii	4	4	3

Among acrylic glasses, copolymer formulations of acrylic compositions appeared to be resistant to activity of mixture of fungus cultures. Tests for individual fungus species proved that all studied compositions of acrylic glasses appeared to be non-fungi-resistant, i.e. they are susceptible to biodeterioration by microfungi (except the variants with Aspergillus niger). The most intensive growth on these materials was expressed by the fungi Paecilomyces variotii, P. chrysogenum.

Notably, the biodeterioration process is more intensive for acrylic glasses in comparison with emulsions, as in this case the observed growth of fungi on the materials was evaluated as 4 points (the fungus growth covers less than 25% of the surface and can be observed with the naked eye). Various levels of biodeterioration of polymer acrylate materials, in our opinion, may be caused either by difference in their compositions, or by physiological and biochemical characteristics of the biodestroying fungi.

We tested individual components of acrylic emulsions for funginertness (Table 3).

Table 3. Funginertness of components of acrylic emulsions

Components of acrylic emulsions	Growth level, points	Evaluation of funginertness
Methylmetacrylate	3	Non-fungi-resistant
Ethylacrylate	0	Fungi-resistant
Methacrylic acid	0	Fungi-resistant
Butylacrylate	0	Fungi-resistant
Butylmetacrylate	0	Fungi-resistant
Styrene	3	Non-fungi-resistant
Vinylidene dichloride – vinyl chloride copolymer latex (VDVC-65 latex)	1	Fungi-resistant
Emulsifier S-10	5	Non-fungi-resistant
Neonol (emulsifier)	0	Fungi-resistant
Sulphonol (emulsifier)	4	Non-fungi-resistant

The experiment results represented in Table 3 demonstrated that ethylacrylate, butylmetacrylate, VDVC-65 latex, neonol, metacrylic acid, and butylacrylate have fungi-resistance characteristics. Meanwhile, such compounds as methylmetacrylate, emulsifier S-10, styrene, and sulphonol appeared to be non-fungi-resistant (the observed fungus growth was evaluated as 5 and 4 points, respectively).

According to the data presented in literature, biodeterioration of polymers is mainly an enzymatic process [8, 19]. Many acrylate-based materials are suspected to micromycetic destruction by activity of exo-oxidoreductases (catalase and peroxidase). In this regard, on the next stage of the study peroxidase and catalase activity of A. terreus, one of the most active biodestructors of the tested materials, was analysed during the cultivation in the medium with acrylate (Figure 1).

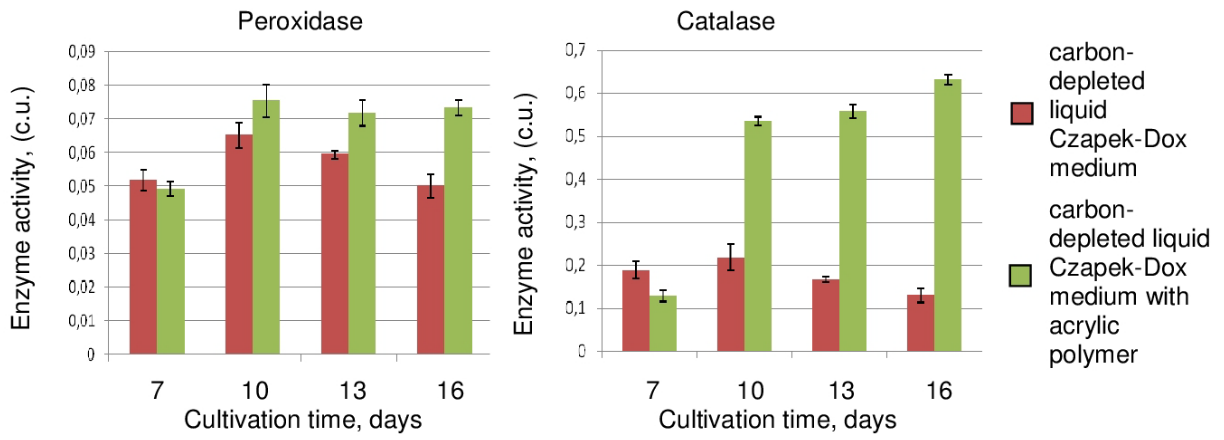

Figure 1. Peroxidase and catalase activity of A. terreus in the course of cultivation on media with different composition

We discovered peroxidase and catalase activity in A. terreus, where the latter exceeded the peroxidase activity significantly. It was demonstrated, that adding acrylic materials to the nutrient medium causes the increase in exo-catalase and exo-peroxidase activity compared with the control experiment, which shows the possibility of participation of these exo-oxidoreductases in the deterioration of the examined acrylic polymer.

On the next stage of this study, the influence of Nuosept 78 and Rosima 243, biocides widely used presently for bioprotection of various construction and industrial materials from mycodestructors, on exo-catalase and exo-peroxidase activity of A. terreus was defined on carbon-depleted liquid Czapek-Dox medium enriched with an acrylic polymer (Figure 2).

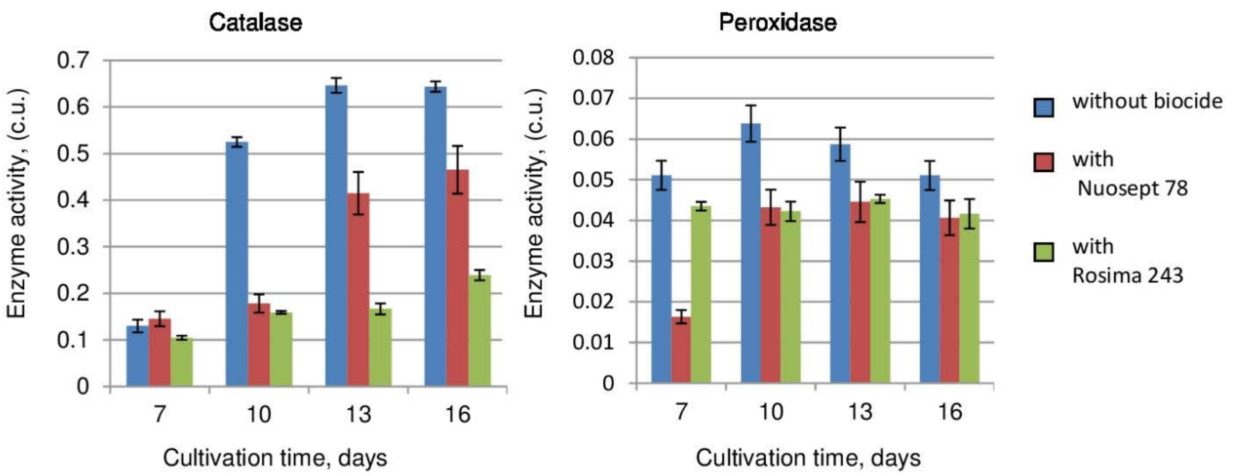

Figure 2. Catalase activity during cultivation of A. terreus on carbon-depleted liquid medium with added acrylic polymer

The experiment results showed that adding biocides to the acrylate-rich cultivation medium causes inhibition of catalase and peroxidase activity. The maximum inhibiting effect of the Rosima 243 in respect of catalase and peroxidase developed on the 13-th – 16-th day of cultivation, and of the Nuosept 78 – on the 7-th – 10-th day of exposure.

Due to the fact that the examined compounds inhibited the activity of fungal exo-oxidoreductases involved into the deterioration of acrylic polymer compositions, it was interesting to assess the possibility to use Rosima 243 and Nuosept 78 as the biodeterioration protective means for non-fungi-resistant acrylic polymer compositions. These biocides were added to the acrylic compositions. The effective biocide concentrations providing resistance of the examined materials to fungal activity were selected by experiment. The results of these experiments are given in Table 4.

Table 4. Funginertness of acrylic polymer compositions with added biocidal additives

Biocide / Material	Growth level, points	
	Rosima 243 (0.5 % w/w)	Nuosept 78 (0.5 % w/w)
Lakroten E-21	0	0
Lakroten E-31	0	1
Latacryl AF	1	1
Latacryl ZM-1	1	0
Polymethylmethacrylate	2	2
Polymethylmethacrylate copolymer - metacrylic acid - n-chlorophenylmethacrylate	2	2
Polymethylmethacrylate copolymer - metacrylicacid	2	2

The results given in the table showed that adding Nuosept 78 and Rosima 243 biocides to the non-fungi-resistant polymer compositions (in the concentration not more than 0.5 % w/w) gave them resistance to microfungal activity. The achieved results justify to the full extent our approach to targeted and systematic protection of specific materials (in this case acrylic polymers) and building construction s on their basis from microbiological deterioration based on used protection means (fungicides) ability to suppress the activity of aggressive metabolites (exoenzymes) taking part in the biodeterioration of construction materials based on acrylates.

Currently, a number of various methods based on microbiological, physical, and chemical principles is used for biodeterioration evaluation for industrial and construction materials [34]. The funginertness evaluation method used by us is based on the visual evaluation of fungal growth on the materials. Although the long time required for this method (28–30 days) is its drawback, its advantage is evident, i.e. only this method allows concluding quite reliably on the possibility to use the material by fungi as a nutrient source. This principle is broadly used both in original and in conventional studies of fungal resistance [35–37].

The question of the possibility to predict funginertness of the whole formulation, if funginertness of separate components is known, needs to be answered. The obtained data show that it is not possible. All tested formulations of acrylic emulsions contain both fungi-resistant and non-fungi-resistant components. However, the fungus growth levels are different on these materials. Consequently, the biodegradation level of the final formulation may depend on the quantitative ratio of initial ingredients, i.e. in this case the phenomenon of emergence was observed. The same situation was observed by us before, when we studied funginertness of acrylic emulsions of the trademarks Acremos and MBM [38]. In case if a non-fungi-resistant polymeric composition needs to acquire fungi-resistant properties, an additive with fungicidal effect should be introduced into this composition.

As mentioned before, now construction and industrial materials are protected not by scientifically-grounded means, but bytrial-and-error method, i.e. a fungicidal additive is selected from the existing list of technical biocides used as the means of protection from biodeterioration. In this case not always an effective biocide can be selected immediately; this process can be time-sapping. Besides, this method is not ecologically safe, as the biocidal effect of the selected protective mean may have the “total” effect and induce extinction of fungi, which are not engaged into the biodestruction process. In our opinion, the physiological and biochemical approach based on the study of mechanisms of biocidal inhibiting effects on fungal metabolites (exoenzymes), involved in biodegradation of materials, provides the optimal and targeted protection from biodeterioration. Nowadays biochemical approach is present in studying the process of construction materials biodeterioration in particular Serova T.A. et al. propose a wood deterioration diagnostic technique based on the determination of its lignin and cellulose content [39].

Our approach was tested by us for protection of acrylates from biodeterioration, as an example. As noted above, the fungi Aspergillus terreus are the most aggressive among the destructors of the studied materials. These micromycetes are known to be good producers of such extracellular enzymes as catalase and peroxidase [29]. These enzymes are able to participate in biodestruction of such materials as acrylates, polystyrenes, epoxide resins [30, 11].The biocides Nuosept 78 and Rosima 243 are broadly used for protection of these materials. The investigation on the dependence of their high bioprotective effect and their ability to inhibit such enzymes as catalase and peroxidase was a matter of a particular interest.

Actually, the results of our experiments confirmed this assumption; and it allowed using the abovementioned biocides as the effective protective means for acrylates against fungal biodeterioration. The physiological and biochemical approach can be successfully extrapolated for the targeted protection of other construction materials, used in construction of various buildings and structures, to enhance its efficiency and reduce the biocidal additives' selection period.

4. Conclusions

In the work, it was revealed that the most of the studied compositions of acrylic emulsions and acrylic glasses widely used in construction industry for creation of various buildings and structures can be subjected to fungal biodeterioration i.e. be used as a nutrient source. Acrylic emulsion Latacryl 3M-1 and copolymer glasses (copolymer polymethylmethacrylate-methacrylic acid- n-chlorophenyl methacrylate and polymethyl methacrylate copolymer – methacrylic acid) have exhibited funginertness. The scientifically based and task-orientated protection of acrylic materials against biodeterioration by adding biocidal additives into the mixture was achieved with respect to physiology-biochemical aspects (the activity of exo-oxidoreductases participating in the biodeterioration) of fungi-decomposers. In the connection with these facts, Nuosept 78 and Roisma 243 were recommended for the protection of non-funginert acrylates against biodeteriorations. Given biocides are able to suppress oxidoreductase activity. This approach will allow considerably optimize and specify the search of biocidal agents for the protection of materials against fungal biodegradation. With the use of the new approach based on the consideration of a biocides ability to inhibit fungal aggressive metabolites in particular exoenzymes that are capable of participating in construction materials biodeterioration, effective biodeterioration protective means (Nuosept 78 and Roisma 243) were selected. The incorporation of given biocides into acrylate compositions provided their persistence to fungal biodeterioration.

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