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Effectiveness and Durability of Biocides in Building Coatings – Biological Aspects

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ABSTRACT

In 2004 an interdisciplinary publicly funded project on the effectiveness and durability of biocides in building coatings was started at the Fraunhofer-Institute for Building Physics in Holzkirchen, Germany. Material components, building coatings and biocide combinations were chosen and samples for outdoor weathering experiments were also designed, all in cooperation with partners from the industry. About 350 biocide-equipped samples together with control specimens without biocide were exposed to real climatic conditions at two locations in Germany. Emerging microbiological growth on the samples was assessed, according to a self-developed special scale. The periodic assessment took place in monthly/two monthly (first location) and half-year cycles (second location), respectively. For different samples of the same type the measurements were averaged and graphically evaluated in the course of the biological development on the surfaces. On most control specimens without biocide a wave like increase of the biological colonization could be detected. Whereas the conditions in Germany for growth on outdoor surfaces are best in the autumn, they stagnate or diminish in the dry seasons, winter and summer. All biocide equipped coatings showed, as expected, a retarded growth. However, some characteristic differences could be recognized which could be assigned to different features of the coatings and to different combinations of biocides.

KEYWORDS

Biocides, Biological colonisation, Retarded growth.

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1 INTRODUCTION

Unwanted emergence of microorganisms on building surfaces, especially on the surface of external thermal insulation composite systems (ETICS), is a growing problem. According to current literature and public understanding, the influencing variables can be classified in the following groups: Global climate change, air pollution, geographical location, exposition of a building, construction details and the choice of materials. Not all of these factors can be influenced by building techniques. Present state of the art does not allow guaranteeing the exemption of unwanted biological growth on building coatings under all circumstances, without the use of biocides [Lindner 2008]. In order to be effective against the target organisms, an active compound has to be water soluble. Otherwise it could not be incorporated in the living cells of the target organisms and would be ineffective. This water solubility does, however, limit the durability of biocides for the film conservation in coatings for façades that are exposed to rain or condensate. In order to achieve an appropriate and ecological use of biocides in building coatings, knowledge about their durability and effectiveness in combination with different matrices (binders) is of importance.

In Germany the warranty for facades being without complaints is at least five years. In Europe the Biocidal Product Directive (BPD) insists on minimising (as far as possible) the amount of biocides which are allowed to enter the environment. Until now there were only limited data available on the long-term field performance of biocidal coatings. Therefore, the industry showed great interest in having products tested with regard to performance in a long term investigation.

An interdisciplinary publicly funded project on effectiveness and durability of biocides in building coatings was therefore started in 2004 at the Fraunhofer-Institute for Building Physics. In cooperation with partners from industry, material components, building coatings and biocide combinations were chosen, whilst samples for outdoor weathering experiments were also designed.

2 MATERIAL AND METHODS

The investigations presented are part of a four year outdoor study. Special specimens with the structure of an external thermal insulation composite system (ETICS), finished with different coatings (plaster or plaster covered with paint) were designed [see also Hofbauer et al. 2006]. Plaster finish was carried out as a structured render with structure grain at a size of 2 mm. Therefore the surfaces were not smooth but somewhat rough. The investigated surfaces of the specimens were of the dimension 30x30 cm² and the polystyrene insulation in each case was 5cm thick. Five different kinds of plaster and three different types of paint were used, as shown in Table 1.

Table 1. Tested coatings.

Plasters		*Paints*
With inorganic binder	Lime cement plaster	Dispersion type silicate paint
	Silicate plaster	Styrene acrylate based emulsion paint
With organic binder	Styrene acrylate based	Silicon resin based paint
	Terpolymer based	
	Silicone resin based	

The final rendering thickness was about 0.5 cm. Four different biocide combinations were added to the surface coatings. Every biocidal mixture consisted of three active ingredients. Two ingredients were always consistent in all combinations, a third ingredient was added from a selection of four (see Table 2). The biocide combinations were added in concentrations of 0.5 mass-% to the plasters and 1.0 mass-% to the paints.

Approximately 350 test specimens based on ETICS structure were exposed to real climatic conditions at two locations in Germany, most of them in Holzkirchen (Fig. 1) in Upper Bavaria, north of the Alps. The climate is characterized by three main factors: heavy driving rain, temperatures below zero for long periods during winter and high daily differences in temperature, caused by radiation and occasional warm winds from the south (Foehn).

Table 2. Tested biocide mixtures.

Obligate compounds	*Variable compounds*
Terbutryne	Carbendazim
	Dichloro-Octylisothiazolinone (DCOIT)
Octylisothiazolinone (OIT)	Iodopropynylbutylcarbamate (IPBC)
	Zinc-pyrithione

Figure 1. Outdoor exposition of the specimen in Holzkirchen.

Apart from chemical and physical measurements, the development of biological growth on biocidal surfaces in comparison to non biocidal surfaces of the same kind, was assessed by different methods. The composition and development of the biological processes were recorded regularly. A visual characterisation of the pattern of the infection with microorganisms and succession was performed. The assessment took place in a monthly/two monthly (first location) and half-year cycle (second location), respectively, under the use of a semi quantitative assessment scale according to Hofbauer et al. [2003], see Table 3. The lower categories of this rating scale provide a minute resolution of the first colonization, whereas the upper categories follow the percentage of coverage by growth. A rating between 0 and 3 can hardly be recognised by an untrained person, because there is very little growth. A rating from 6 to 10 is also recognizable as disfigurement by laymen and would cause complaints and rating 4 and 5 form a transition position where complaints might occur.

Additional developing growth was further recorded by a specially developed visual-optical raster scan, and also documented by photographs. The growth recorded was also categorised if consisting of fungi, algae or both.

In addition to the methodology specified, further measurements were made, the results of which are presented elsewhere. Biological measurements for instance, were completed by physiological investigations (efficacy-test) and a complex quantitative and taxonomical analysis of the microorganisms colonizing the different coating-systems. Furthermore, an ecological analysis and a resistance-test also accomplished the results of the visual observations.

Table 3. Rating scale for the assessment of surface growth on the specimen according to Hofbauer et al. [2003], simplified. White table lines indicate minor growth intensity that would not lead to complaints; bright green is used in a transition area where complaints may start. The intense green colour marks growth intensities that would most certainly provoke complaints.

Rating	*Microbiological growth covering examined area on an average of*
10	87,5% - 100%
9	75% - 87%
8	62,5% - 75%
7	50% - 62,5%
6	37,5% - 50%
5	25% - 37,5%
4	5% - 25%
3	less than 5 %
2	few single spots (less than 5 %)
1	1-3 single spots
0	0 % - no spots

3 RESULTS

As shown in previous studies [Hofbauer et al. 2006]; [Hofbauer 2007], development of growth followed cyclic fluctuations (Fig. 3). During humid periods of the year (autumn, in Central Europe for example) a more intense development of growth is recognized whereas during dry and warm or dry and cold periods (in Central Europe during mid summer and late winter), a stagnation or even reduction of growth occurs. Once growth is present, it rarely disappears completely again. During the next convenient climate phase the development will recommence at a higher level. Figure 3 shows a scheme of a typical cyclic fluctuation of growth development with a wavelike increase.

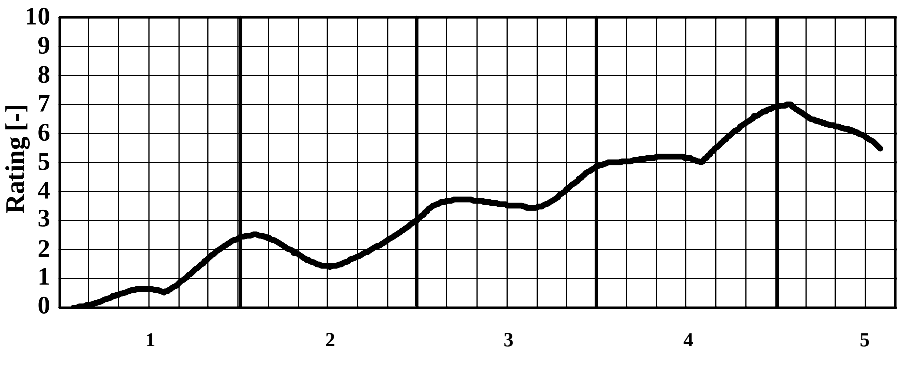

Figure 3. Typical growth development in phases as measured on a non biocidal surface during the project.

In general the efficiency of the biocide-mixtures used is confirmed, the growth development on all of the biocidal equipped coating systems is clearly retarded. Even after four years of exposition, the growth of some biocidal variants has to be rated as inconsiderable or stagnating. Some variants, on which only fungal growth occurred until the end of the project, are optically conspicuous, due to the presence of demaciaceous fungi (mould fungi with dark pigmentation).

A correlation between the effectiveness of biocides and the type of binding agent used can be observed. In the course of time, surface growth may occur earlier and more intensely on specimens with a high total measurable biocide content, in comparison to variants with a lower content over the same duration. This effect could be a result of a different withholding capacity for biocides or a different release, respectively.

Specimens with silicon resin based variants turned out to be quite durable against microbiological growth, as shown in Figure 4. By observation completion, growth intensity on samples of all biocide combinations had to be rated as inconsiderable, they were rated below 3. Even on the specimen without biocide, the critical rating of 3 was not reached until after two years of exposition.

The ratings outlined in figure 4 comprise of approximate-curves based on geometric mean values from the measurements. The rating of the growth intensity is shown through the ordinate, the numbers on the abscissa show the number of months elapsed.

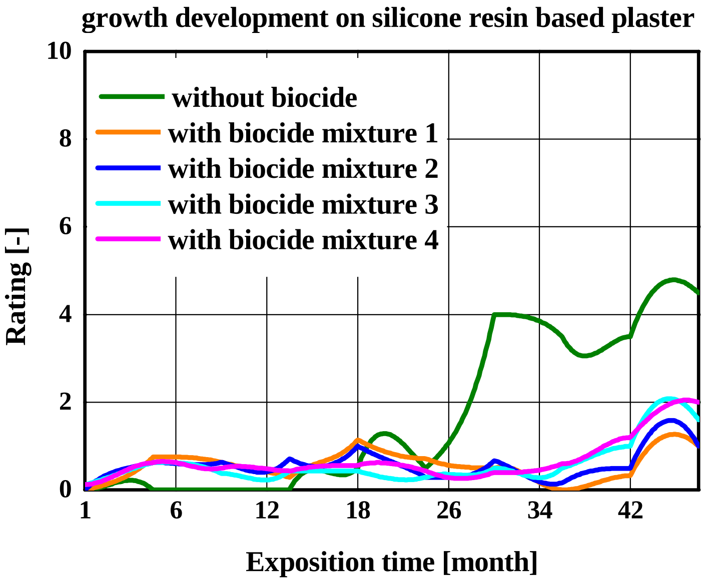

Figure 4. Observed growth on silicon resin variants.

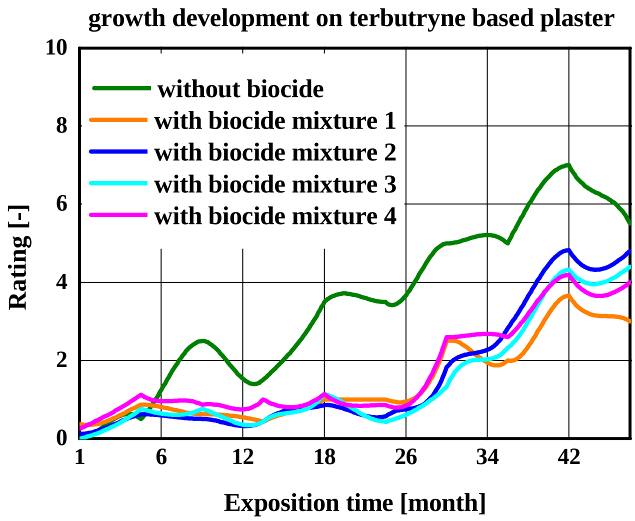

Figure 5. Observed growth on terpolymer variants.

Unlike the previous variants, the specimen with plaster coating based on terpolymer already showed significant growth earlier, on both biocidal equipped and non equipped variants, as shown in Figure 5. Within the measured period the "critical" rating of 3 was even reached by all biocidal equipped systems.

The lime cement plaster could not be rated conclusively, because the biocidal effect was overlaid by other effects, for instance by chalking and weathering because of heavy driving rain or hail.

While in some coating systems all active biocidal components show a similar effect, in some coating systems special biocidal combinations performed better than others. In general, systems with paint coating showed better results than systems without paint coating. The paint coatings were equipped with twice as much concentration of active compounds than the render. However, due to the substantially lower thickness of the paint coatings, the total content of active compounds is less than in renders.

4. DISCUSSION

The results show, that the concentrations of active compounds in a coating do not allow direct assessment of the effectiveness against biological growth. The interaction between the type of biocidal composition, the type of coating (binder) and the physical building conditions is of essential importance. Coatings upon which microorganism colonization is generally less because of their chemical constitution are protected by lower total amounts of active compounds. It is important to supply the immediate coating surface with effective concentrations of biocides. Therefore, coating systems with a pronounced withholding capacity for biocides may lead to an undersupply at the surface.

Depending on the chemical character of the biocides used (e.g. Terbutryne, IPBC), the formation of even more active transformation products from the biocidal agents, is possible. Transformation may be due to effects caused by temperature, oxidation, hydrolysation, sunlight, etc. These processes could cause an initial decrease in the concentration of the biocidal compounds primarily used, whilst, at the same time, the protection of the façade is still ensured through the potentially longer lasting biocidal effective transformation products.

The additional taxonomical and ecological analysis of the microorganism species found in the early stages of colonization on the surfaces confirmed that, according to the various biocidal compositions and coating systems, different ecological groups of organisms are present. Gaps in the efficiency spectrum of the other biocidal compounds became obvious, in the case of a decrease or loss of an active compound, by the appearance of typical species. For example; *Alternaria* spp., *Ulocladium* spp., are already known for an increased tolerance against Carbendazim (e.g. [Bollen et al. 1983], [Hassal 1990], [Deising et al. 2008]). Therefore, specimens equipped with a biocidal combination comprising of Terbutryne, OIT and Carbendazim were earlier colonised by the fungi listed, because OIT diminished faster than other biocides.

The results of the study presented show that biocidal equipped paints are very effective in the protection of building coatings against unwanted growth of microorganisms. If only the paint finish is equipped with biocides but not the underlying render, the total amount of biocides necessary may be reduced significantly.

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