



AKADÉMIAI KIADÓ

# Investigation of façade coatings containing algae-prone fillers

Holger Heinrich<sup>1\*</sup> , Adél Len<sup>2,3</sup> and Helmuth Venzmer<sup>4</sup>

Pollack Periodica •  
An International Journal  
for Engineering and  
Information Sciences

18 (2023) 1, 55–59

DOI:

10.1556/606.2022.00592

© 2022 The Author(s)

<sup>1</sup> Marcel Breuer Doctoral School, Faculty of Engineering and Information Technology, University of Pécs, Pécs, Hungary

<sup>2</sup> Department of Civil Engineering, Institute of Smart Technology and Engineering, Faculty of Engineering and Information Technology, University of Pécs, Pécs, Hungary

<sup>3</sup> Department of Neutron Spectroscopy, Center for Energy Research, Budapest, Hungary

<sup>4</sup> Department of Civil Engineering, University of Applied Sciences, Technology, Business and Design in Wismar, Wismar, Germany

Received: January 7, 2022 • Revised manuscript received: May 25, 2022 • Accepted: June 3, 2022

Published online: August 17, 2022

## ORIGINAL RESEARCH PAPER



### ABSTRACT

Algae are an evolutionary model of success and colonize all suitable ecological niches including building material surfaces that have favorable characteristics. In the last 25 years, building physics measures were developed to reduce water availability, especially on external thermal insulation composite systems.

Investigations into the influence of coating formulations have so far primarily focused on binder systems, biocides and hygrothermal properties. Research on the algal susceptibility due to the fillers is not to be found, but these regularly constitute a large proportion of final coatings. The present work investigates the influence of magnesium-containing fillers in the process of algal colonization of free-weathered façade coatings and a defense-strategy by water-activated pigment composition.

### KEYWORDS

algal colonization, BenthosTorch<sup>®</sup> fluorometer, magnesium-containing fillers

## 1. INTRODUCTION

Wherever the availability of water, light, mineral nutrients and carbon dioxide is guaranteed, algae can be found. Even to strong fluctuations of these growth factors, the autotrophic microorganisms are adapted [1]. Building façades offer a large contact surface for the ubiquitous algae species and in the case of External Thermal Insulation Composite Systems (ETICS) the thermal decoupled façade enables an increased presence of moisture films [2, 3]. Natural carbonate rock is also colonized by numerous microorganisms and thus contributes to its weathering and dissolution [4]. In addition to fungi and bacteria, even algae show great interest in the chemical components of carbonate rock [4]. It can be assumed that the algal colonization of façades is not just an aesthetic problem. Coatings, that are applied to protect façades, become substrates due to an unfavorable combination of ingredients. Airborne algae mainly get onto vertical façade surfaces by wind and wet deposition [5]. Once there, the production of biomass requires sufficient essential mineral substances.

It is assumed that these nutrients reach the façade through atmospheric dust in the same way as the algae [6]. An evaluation of the air quality annual reports of the state of Mecklenburg-Western Pomerania, Germany shows a declining development of the mineral nutrient content of the air in the last decade [7]. The present investigation was carried out in the geographical area of the air quality reports. In the search for alternative sources of nutrients within the final coatings - the focus is on fillers. Mineral fillers fulfill numbers of important technological functions and reduce manufacturing costs [8]. When looking through various recipes of façade paints, regularly magnesium-containing fillers like dolomite

\*Corresponding author.

E-mail: [goldabee@web.de](mailto:goldabee@web.de)



and talc will be found [8]. Calcium carbonate is the most important filler and, depending on its geological origin and quality, contains significant amounts of magnesium-based accessory minerals [9]. The chemical composition of the fillers ideally meets the requirements of phototrophic microorganisms as they are obtained from natural mining resources that are the result of primeval marine organism activities [10]. This common past of algae and fillers will meet again on modern façades today and prompts the author to investigate further, because there is no photosynthesis without the magnesium core-ion of chlorophyll-a.

It is known from the industrial harvesting process of green algae cultures that they can be precipitated by calcium- and magnesium-compounds at certain pH-values [11]. Flocculation of algae is driven by a complex bonding interaction of cationic metal-ions with the cell-wall-located functional groups [12]. Especially magnesium-compounds have shown high flocculation efficiency over a wide pH-range for *Chlorella sp.* [13]. Transferring the principle of algae harvesting to building material surfaces might deliver an explanation about the initial contact and the following colonization process. Instead of elaborately avoiding condensation, this should become the subject of a water-activated control strategy. Manipulating the pH-level at the coating surface by in-situ generated protons via zinc molybdate is known as active technology against bacteria and viruses [14, 15]. Research reports on the use of zinc molybdate against algae on weathered façades are not to be found and are to be examined by the present work. Overall, the composition of the façade determines a subsequent possibility of recycling and goes beyond the carbon footprint [16].

## 2. MATERIAL AND METHODS

### 2.1. Test location and conditions

The aim of the weathering test was to compare the algal susceptibility of acrylic-dispersion paints containing different types and amounts of magnesium-containing fillers. The outdoor exposure lasted from December 2019 until December 2021 and included 7 painted panels. The panels were mounted vertically on the north gable of a brick house at an angle of 5°. The test site is situated in a rural area with dense vegetation and agricultural use (Fig. 1). The direct distance to the Baltic Sea is 4.5 km in a North-Western direction. The A24 motorway runs 2 km southern of the site. Air temperature ranged from -11.5 to 36.6 °C with a mean of 10.6 °C and relative humidity ranged from 36.9 to 100% with a mean of 81.3% ( $n = 36,372$ ).

### 2.2. Set of materials

The materials to be tested were seven laboratory-made façade paintings containing different amounts and types of magnesium-containing fillers (Table 1). The pigment volume concentration kept at the same level but intentionally set to be more critical to simulate an accelerated aging



Fig. 1. Weathering location rural area near Wismar, Germany (Google Earth for Chrome, Goldebee 53°89'44"N 11°60'08"E, © GeoBasis-DE/BKG 2009, URL: <http://google.com/earth>)

process and having the filler particles more present to the surface. Sample PK06 contains 5% (calculated on non-volatiles) zinc molybdate (Carl Roth GmbH & Co KG, Karlsruhe, Article No. 0874.4) to evaluate an approach as defense-strategy against algal growth [14, 15]. Close to the test area, samples of airborne algae were collected over a period of 14 days and a microscopic evaluation of main algae species revealed *Scenedesmus spp.*, *Kirchneriella spp.* and *Chlorella spp.* Direct mounting to the building simulates a future energetic renovation using different paint coatings. The weather data were recorded via data logger for air temperature, relative humidity, air pressure and dew point temperature in a 30-min-interval. This configuration resulted in a similar specific heat capacity of the surfaces, avoided different condensation loads and possible problems of comparability [3].

The coatings were applied on expanded-polystyrene-based, lightweight Ultrament® building boards (Ultrament GmbH & Co. KG, Bottrop, Germany) with dimensions of 20 × 600 × 600 mm each. The concrete-slurry-coated surface, reinforced by a 10 × 6 mm plastic mesh, delivered a well-defined structure for every sample surface. The amount of roller-applied wet paint was 250 g m<sup>-2</sup> for each panel. Pigment volume concentration was set by varying the amount of acrylate binder. Additional biocides were not used. The non-ionic pure acrylate dispersion K498 (Kremer Pigments, Aichstetten, Germany) contained an in-can antibacterial preservative due to the manufacturing process.

### 2.3. Detection of algal biomass

The algal biomass was quantified using the BenthosTorch® fluorometer (from German company bbe moldaenke GmbH, 24,222 Schwentinental [17], referred to as BTo for short in the following). The mobile device enables non-destructive, area-related detection of three algae groups (green algae, cyanobacteria, diatoms) within a detection range of 0–15 µg cm<sup>-2</sup> chlorophyll-a. While sampling was not necessary the destruction of microorganisms and components could be avoided. The panels were humidified in a controlled manner by using a water spray bottle 15 min before measurement. Algae discrimination and quantification is based on specific wavelength absorption of photopigments at 470, 525 and 610 nm. The sum of biomass results from the chlorophyll-a

Table 1. Basic paint components of sample set (quantities in grams)

Components [g]	PK00	PK01	PK02	PK03	PK04	PK05	PK06
Titanium dioxide	7.5	7.5	7.5	7.5	7.5	7.5	7.5
Calcium carbonate	112.5	112.5	112.5	112.5	112.5	112.5	112.5
Quartz powder	4.5	4.5	4.5	4.5	4.5	4.5	4.5
Dolomite	–	21.0	45.0	77.0	76.0	75.0	–
Talcum	–	–	–	–	26.0	–	–
Mg-Hydroxycarbonate	–	–	–	–	–	70.0	–
Zinc molybdate	–	–	–	–	–	0	9.4
Pure Acrylic Disp. 50%	105.0	125.0	140.0	170.0	190.0	250.0	110.0
Water	60.0	70.0	80.0	95.0	110.0	125.0	65.0
Total [g]	289.5	340.5	389.5	466.5	526.5	644.5	308.8
Pigment vol. conc. [%]	50.0	50.0	50.0	50.0	50.0	50.0	50.0
Non volatiles [g]	177.0	208.0	240.0	287.0	322.0	395.0	189.0
Solid content [%]	61.0	61.0	61.0	61.0	61.0	61.0	61.0

content, which is common to all three groups of algae and has a fluorescent signal at 690 nm. On-board calibrations deliver area-related cell numbers [ $\text{cells mm}^{-2}$ ] and chlorophyll-a [ $\mu\text{g cm}^{-2}$ ]. Using a 13-dot template, each test panel was measured 13 times between December 2019 and December 2021 ( $n = 169$ ). The single-mode measuring process took 20 s (including 10 s diode initialization) and covered an area of  $1 \text{ cm}^2$ . Previous dark adaptation [18] was omitted and limited to the initialization phase of the diodes before the measurement process started. The visual surface disfigurement was also evaluated according to the requirements of German version DIN EN 16492: 2014 [19] and a digital 8 bit greyscale picture analysis with the open-source software ImageJ [20].

### 3. RESULTS AND DISCUSSION

#### 3.1. Development of algal biomass – BenthosTorch<sup>®</sup> fluorometer

During weathering, the coatings showed a strong fluctuating stock of algal biomass. The first signals of green algae were detected after 44 (PK04) and 89 days (PK03) but disappeared again in the further course and were below the visual threshold. Above exposure time of 327 days permanent signals of green algae appeared (PK03, PK04, PK05). Sample PK05 was continuously and visibly colonized from day 327. Figure 2 depicts the mean value of the total cell numbers of all algae species after 730 days. The corresponding chlorophyll-a contents were between 0.01 and  $0.60 \mu\text{g cm}^{-2}$ . The mean value of fluorescence detected

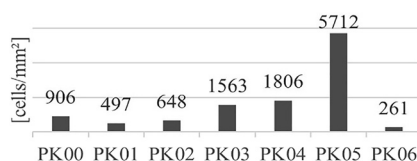


Fig. 2. Mean values over all species after 730 days outdoor exposure ( $n = 169$ , 13-dot template)

algal biomass was composed of 92.3% green algae, 6.3% cyanobacteria, and 1.4% diatoms.

#### 3.2. Evaluation of algal biomass according EN 16492:2014

According to the German standard DIN EN 16492:2014 [19] the evaluation of the surface disfigurement caused by fungi and algae on coatings was conducted visually. Evaluation carried out according to the criteria of intensity, quantity and area proportion, see Figs 3 and 4.

#### 3.3. Evaluation of algal biomass via ImageJ area percentage method

In addition to the first two evaluation methods, each test specimen was recorded photographically. After conversion into 8-bit grayscale images, the analysis was carried out using ImageJ processing and analysis software [20]. The optical influence of the mounting brackets was eliminated by using a central square section of 95% image area fraction.

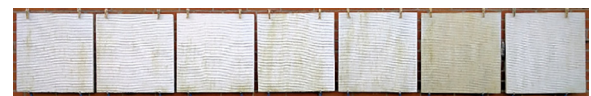


Fig. 3. Visual impression after 730 days PK00 to PK06 (from left to right)

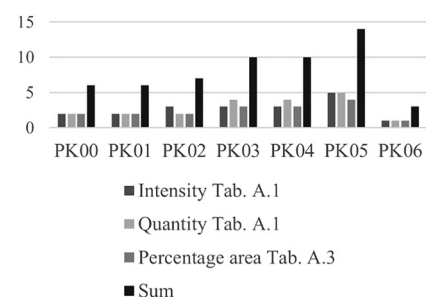
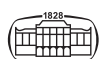


Fig. 4. Results of visual evaluation after 730 days weathering according EN16492:2014, annex A



Subtracting a fix offset for the structural-borne shadows, the percentage area of the algal biomass was calculated with histogram threshold between 100 and 150.

The coefficients of determination between results of evaluation methods (Table 2) were BTo [cells/mm<sup>2</sup>] vs. DIN EN 16492 sum ( $R^2 = 0.95$ ) and BTo [cells mm<sup>-2</sup>] vs. ImageJ area [%] ( $R^2 = 0.97$ ). After comparing the test methods, the correlation of the determined algal biomass to the magnesium content of the coatings was performed. Figures 5 and 6 are revealing the relationship between algal biomass and magnesium content of painted samples (Table 2).

### 3.4. Statistical data set analysis – post-hoc Tukey HSD test

Statistical evaluation was carried out according to the Tukey Honestly Significant Difference (HSD) test [21]. This multiple comparison test followed the one-way analysis of variance to determine significant differences between group means. The data set analyzed based on total cell numbers measured with BenthosTorch<sup>®</sup> fluorometer. The Tukey test

Table 2. Data sets for correlation of results between all methods

Sample	ImageJ area [%]	BTo [cells mm <sup>-2</sup> ]	EN16492 sum	Mg [%]
PK00	7.52	906	6	0.76
PK01	13.75	497	6	1.96
PK02	16.26	648	7	3.00
PK03	23.98	1,563	10	3.96
PK04	22.19	1,806	10	5.00
PK05	38.46	5,712	14	7.35
PK06	4.95	261	3	0.71

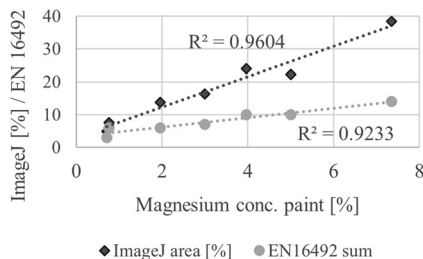


Fig. 5. Correlation of visual methods vs. Mg-content of weathered samples

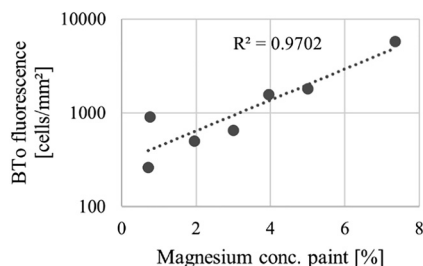


Fig. 6. Correlation of total cell numbers all algae species vs. Mg-content weathered samples

identified 7 out of 21 treatment pairs which are significantly different from each other. The pairs were PK00/PK05, PK01/PK05, PK02/PK05, PK03/PK05, PK04/PK05, PK06/PK05, PK04/PK06.

### 3.5. Discussion

Algal biomass measured during outdoor weathering could not be distinguished according to whether it was physical adsorbed or biological reproduced. It can be assumed that initial signals were dominated by adsorption (harvesting step) and successive growth processes emerged later (growth process). At this point, the dual function of magnesium-containing fillers might become apparent. In the short-term they promote the accumulation of airborne algal cells and in the long-term being supplier of essential minerals. This could be explained by the large fluctuations in cell numbers during the start-up phase until the algal biomass is established.

Initial algae appearance took place randomly spot-wise and could be underestimated while using a fix template pattern as the other applied methods included the entire sample area. The measuring spot of the BenthosTorch<sup>®</sup> fluorometer is 1 cm<sup>2</sup> and was collected via 13-dot template (13 × 1 cm<sup>2</sup>). This represented 0.36% of the total weathered sample area, but the supposed disadvantage could not be confirmed. BenthosTorch<sup>®</sup> fluorometer was able to detect algal cells, especially in the early phase of settlement while visual assessment was not possible. The BTo enables an enhanced evaluation for inexperienced user as there is no need for microscopes or having a high microbiological expertise. The necessary differentiation between fungi and algae according EN 16492:2014 [19] was automatically preceded by the measuring system. An uncomplicated handling of the mobile device avoided invasive sampling and data sets were directly stored. All measurements were performed in the field without time-consuming sample preparation. This was an advantage compared to pulse-amplitude-modulated fluorometry, which required a lot of lab-depending instrumental effort due to other target values [18]. Three different evaluation methods confirmed the experimental observations. After a lead time of around 300 days, the presence of algae increased sharply. Considering the critical formulation of the paints, this colonizing course corresponded to the results of earlier outdoor weathering tests [22, 23] and the desired accelerated effect of sample preparation.

## 4. CONCLUSIONS

Two years of weathering revealed a significant difference in the algal susceptibility between the identical constructed samples. The fluorescence-measured sum of algal biomass [cells/mm<sup>2</sup>] is positive related ( $R^2 = 0.97$ ) to the magnesium content of applied paint coatings. The cross-check according to the standard DIN EN 16492:2014 ( $R^2 = 0.92$ ) and the digital image area analysis ( $R^2 = 0.96$ ) ensured the results.



The accelerated coating modifications with higher magnesium contents caused by fillers triggered the highest value of algal biomass. The water-activated defense-strategy was significantly able to limit the biomass within the experimental conditions and algal species involved. Construction planner and designer are calculating with a wide variety of load cases. In terms of future façade-projects the assessment of final coatings can be improved based on these results. The simultaneous combination of environmentally hazardous biocides with algae-prone fillers in façade paints is not advisable. Especially since the organic biocides have only a temporary effect and the fillers reach the surface during the aging process to develop the effects revealed in this research work. Understanding these interactions provides important information and methods on the reduction of organic biocides in product development. These research results contribute to more sustainable designed true green façades without killing future algal populations.

## REFERENCES

- [1] A. A. Gorbushina, "Life on the rocks," *Environ. Microbiol.*, vol. 9, no. 7, pp. 1613–1631, 2007.
- [2] M. Krus, K. Sedlbauer, and K. Lenz, "Influence of different measures on the undershooting of the dew point on exterior surfaces" (in German), Special Edition 4. *Dahlberg-Kolloquium*, Wismar, Germany, H. Venzmer, Ed., Berlin: Huss-Medien GmbH, Verlag Bauwesen, 2003, pp. 83–94.
- [3] T. Steffgen, "Experimental studies – Building physics, investigation in condensation water on plaster surfaces," *Pollack Period.*, vol. 14, no. 1, pp. 167–175, 2019.
- [4] B. Lian, Y. Chen, L. Zhu, and R. Yang "Effect of microbial weathering on carbonate rocks," *Earth Sci. Front.*, vol. 15, no. 6, pp. 90–99, 2008.
- [5] C. Rüger, "Dust binding – Wet adhesion" (in German), in *The Ways of Dust – In Human Environment* (in German). Berlin Heidelberg: Springer Verlag GmbH, 2016.
- [6] K. Freystein and W. Reisser, "Algal colonization of facades - Influence of fungi on colonization, resistance and control of algal coatings" (in German), in *Façade Renovation* (in German), H. Venzmer, Ed., Beuth-Verlag Berlin, 2011.
- [7] Annual air quality reports, State Office of Environment, Nature Conservation and Geology, Mecklenburg-Western Pomerania, Güstrow, 2011–2020 (in German). [Online]. Available: [www.lung.mv-regierung.de/umwelt/luft/lume.htm](http://www.lung.mv-regierung.de/umwelt/luft/lume.htm). Accessed: Dec. 16, 2021.
- [8] R. Baumstark and M. Schwartz, *Dispersions for Architectural Coatings – Acrylate Systems Sin Theory and Practice* (in German). Hannover: Vincentz Verlag Hannover, 2001.
- [9] O. Lückert, *Pigment + Filler Tables* (in German), 6th ed. Vincentz Verlag Hannover, 2002.
- [10] F. Ahnert, *Introduction to Geomorphology* (in German). Verlag Eugen Ulmer Stuttgart, 1996.
- [11] Z. Wu, Y. Zhu, W. Huang, C. Zhang, T. Li, Y. Zhang, and A. Li, "Evaluation of flocculation induced by pH increase for harvesting microalgae and reuse of flocculated medium," *Bioresour. Technol.* vol. 110, pp. 496–502, 2012.
- [12] M. Vogel, "For the uptake and binding of uranium (VI) by the green alga *Chlorella vulgaris*" (in German), Doctoral Dissertation, Technische Universität Dresden, 2011.
- [13] D. Vandamme, I. Foubert, I. Fraeye, B. Meeschaert, and K. Muylaert, "Flocculation of *Chlorella vulgaris* induced by high pH: Role of magnesium and calcium and practical implications," *Bioresour. Technol.*, vol. 105, pp. 114–119, 2012.
- [14] S. Guggenbichler, T. Fey, and J. P. Guggenbichler, "Hospital acquired infections with multiresistant microorganisms: UN Inter-agency Coordination Group on antimicrobial resistance demands immediate, ambitious and innovative action," *Integrated Biomed. Sci.*, vol. 6, no. 1, pp. 84–104, 2020.
- [15] "Zinc molybdate with triclinic crystal structure as antimicrobial agent", EP 3643177A1, Amistec GmbH & Co KG, Kössen/Austria, 2020.
- [16] M. Porhinčák, A. Eštoková, and S. Vilčeková, "Comparison of environmental impact of building materials of three residential buildings," *Pollack Period.*, vol. 6, no. 3, pp. 53–62, 2011.
- [17] bbe moldaenke GmbH, Schwentinental/Germany, Measurement of Phytobenthos Fluorescence (in German), 2022. [Online]. Available: <https://www.bbe-moldaenke.de/en/products/chlorophyll/details/benthotorch.html>. Accessed: Jan. 2, 2022.
- [18] J. Von Werder and H. Venzmer, "The potential of pulse amplitude modulation fluorometry for evaluating the resistance of building materials to algal growth," *Int. Biodeterior. Biodegrad.*, vol. 84, pp. 227–235, 2012.
- [19] *DIN EN 16492:2014*, Paints and varnishes – Evaluation of the surface disfigurement caused by fungi and algae on coatings, German version (in German), DIN Deutsches Institut für Normung e.V., Beuth Verlag GmbH, Berlin, 2014.
- [20] ImageJ, Image processing and analysis in Java, 2018. [Online]. Available: <https://imagej.nih.gov/ij/index.html>. Accessed: Dec. 27, 2021.
- [21] N. Vasavada, One-way ANOVA (ANALYSIS OF VARIANCE) with post-hoc Tukey HSD (Honestly Significant Difference) test calculator for comparing multiple treatments, 2016. [Online]. Available: [https://astatsa.com/OneWay\\_Anova\\_with\\_TukeyHSD/](https://astatsa.com/OneWay_Anova_with_TukeyHSD/). Accessed: Dec. 27, 2021.
- [22] J. von Werder, H. Venzmer, and R. Cerny, "Application of fluorometric and numerical analysis for assessing the algal resistance of external thermal insulation composite systems," *J. Build. Phys.*, vol. 38, no. 4, pp. 290–316, 2015.
- [23] R. Schwerdt, *Durability of Biocidal Agents in Architectural Coatings in a Multi-year Field Trial* (in German). vol. 8, Research Results from Building Physics. Stuttgart: Fraunhofer Verlag, 2011.

**Open Access.** This is an open-access article distributed under the terms of the Creative Commons Attribution 4.0 International License (<https://creativecommons.org/licenses/by/4.0/>), which permits unrestricted use, distribution, and reproduction in any medium, provided the original author and source are credited, a link to the CC License is provided, and changes – if any – are indicated. (SID\_1)

