

1 An empirical failure model to predict biofouling growth on fired bricks due to 2 microalgae

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10 Highlights

- 11 - An empirical failure model about microalgae biofouling was developed
- 12 - The model takes into account the main substrate properties influencing the growth
- 13 - It also considers the environmental temperature and relative humidity
- 14 - The model was developed and validated on fired bricks

15 Abstract

16 The purpose of this study was to provide an empirical failure model predicting the microalgae growth on fired
17 bricks surfaces. It was developed through a numerical fitting of experimental data present in literature. It
18 considered the substrate properties related to biofouling (i.e. porosity and roughness) of different bricks under
19 several environmental conditions (i.e. relative humidity and temperature). Results shows that the model is able
20 to simulate the microalgae biofouling by explicitly taking into account such influencing factor. Finally, this
21 empirical failure model is validated on a different dataset from literature and applied to time varying temperature.

22
23 **Keywords:** *microalgae biofouling; fired bricks; empirical failure model; substrate properties; environmental*
24 *conditions;*

25 **1 Introduction**

26 When porous building materials are exposed to environmental weathering, their physicals and chemical
27 properties interact with biological factors, leading to changes in both its compositional and structural
28 characteristics [1–3]. The growth process and vegetative development of organisms have a direct consequence
29 on the material due to the metabolic activity connected with the growth of living organisms [4]. The living species
30 that commonly dwell on these materials are ranging from microscopical bacterial cells to higher plants and
31 animals [5].

32 Biofouling on porous building materials is a colonization process usually started by photoautotrophic
33 microorganisms since they only need light, water and some inorganic components to start growing [1,2]. Among
34 these, the most recurrent groups are green microalgae and cyanobacteria, shortly named as microalgae, and they
35 usually develop in combination, especially in the European context [6–8]. Frequent maintenance and repairing
36 interventions are then required in order to limit aesthetical, chemical and physical degradation they may produce:
37 both ways, either repairing or not, could ultimately cause serious losses (economical or even cultural, if cultural
38 heritage is involved) [9,10]. In order to describe and therefore limit microalgae biofouling risk on porous building
39 materials, in recent years researchers adopted two strategies [11]: (1) determining and thus limiting, when
40 possible, the influencing factors of biofouling growth; (2) providing models that can simulate and then forecast
41 the biofouling risk.

42 Regarding the first one, literature works mainly focused on the factors that influenced the water activity: that is
43 defined as the water available to microorganisms to growth [6,10,12–17]. They highlighted that a combination
44 of environmental conditions, substrate properties and intrinsic aspects of the plumbing system of buildings (i.e.
45 leaky parts and design defects of the construction) could ensure the growth and development of microalgae. For
46 what concerns the environmental conditions, it was demonstrated that microalgae growth occurred only at
47 saturation conditions [13,18], that is when water can be found at liquid state. From an engineering standpoint,
48 however, two assumption can be done: water activity can be approximated with the relative humidity RH , as
49 previously demonstrated in [19] and the saturation condition safety limit can be set for $RH \geq 98\%$ [18], even though

50 brick surface could not be wet, and water activity is only present by capillary condensation [20]. Moreover, it
51 was proved that temperature allows microalgae to develop between about 5°C and 40°C with an optimal growth
52 condition at about 27.5°C [18,21,22]. Out of this range, microalgae growth process is unable to start or, if already
53 started, it stops [1,2,18,23]. In this context, porosity and roughness were outlined as the main factors concerning
54 the substrate properties [13,24–27]. In fact, roughness promotes the adherence of microalgae to the substrate
55 while porosity is responsible for retaining water inside the material structure. Moreover, when the materials
56 structure and geometry, in terms of porosity and roughness, is able to afford enough water activity for microalgae
57 to start their growth, the chemical composition of the substrate may play only a secondary role, favouring or
58 limiting their development [28–32].

59 For what concern the second strategy, literature has provided a reliable model for describing microalgae growth
60 starting from experimental measures [33]. Based on the Avrami's theory, it connects the area covered by
61 biofouling over the time by denoting a first phase of latency (when microalgae stains are not still visible),
62 followed by a rapid growth, and finally a stagnation phase when the covered area reaches its maximum and
63 becomes constant over the time. Such approach was applied to several type of porous buildings materials (i.e.
64 mortars, fired bricks and stones), under different environmental conditions and biocides surface treatments,
65 confirming its capabilities [18,26,33–35]. However, it never explicitly and quantitatively accounts for the growth
66 influencing factors of the substrate and environmental conditions, as failure models usually do. This way, no
67 failure models still exist in literature for microalgae growth on porous building materials, to the authors'
68 knowledge, while numerous failure models have been presented for other type of biofouling (e. g. mould, fungi,
69 actinomycetes) [19,36–43].

70 Hence, the aim of this work is to propose a first empirical microalgae growth failure model, that is able to
71 explicitly take into account, for the first time, the main influencing factors of substrate parameters (such as
72 porosity and roughness) and environmental conditions (such as temperature and relative humidity), as well as
73 their variation over the time. Fired bricks are here considered due to the availability of a large set of experimental
74 data in literature, but the methodology can be extended to other porous building materials when enough

75 experimental data will be available. To this aim, this work is divided into three main phases. The first phase
76 involves the definition of: the model general requirements (Section 2.1); the variables domains (Section 2.2) and
77 the specific model equations (Section 2.3). The fitting method is then provided (Section 2.4). Lastly, resulting
78 equations are presented and validated and the application of the model under time varying temperature is reported
79 (Section 3).

80 2 Materials and methods

81 2.1 General requirements

82 As stated above, the starting point of this work is the modified Avrami's model [34,35] showed in (1):

$$X(t) = \frac{A_C}{A_T} \cdot (1 - \exp^{-K(t-t_1)^n}) \quad (1)$$

83 where the covered area by microalgae growth $X(t)$ [-] is given as a function of time t [day]. The final covered area
84 ratio is represented by the parameter A_C/A_T which expresses the percentage of the covered area at the end of the
85 process (A_C is the maximum covered area by biofouling on a specific sample, and A_T is the total area of the same
86 sample), K [day^{-4}] is a rate parameter determined by the least squares method using experimental measurements.
87 Lastly, the parameter t_1 represents the latency time [day] before a chromatic variation occurs on the material
88 surface and the coefficient n is the Avrami's coefficient which can be assumed equal to 4 [34].

89 The model variables are chosen according to literature findings. Porosity P [-] and roughness R [μm] are the main
90 factors characterising the substrate [6,25,26] and, their values are easily measurable and available in literature
91 (see Section 2.2). Temperature T [$^{\circ}\text{C}$] and relative humidity RH [%] are instead selected as the main factors
92 representing environmental conditions [13,14,18].

93 In this way:

- 94 - since literature showed that, regardless of the temperature, RH determines the actual possibility for
95 microalgae to growth, according to which growth happens only if $RH \geq 98\%$ [18], it can be considered as
96 an on/off factor;

97 - when $RH \geq 98\%$ and $5^\circ\text{C} \leq T \leq 40^\circ\text{C}$ growth can happen depending on T , P and R , otherwise it cannot
 98 [13,18];
 99 - thus, P , R and T directly affect the microalgae coverage X through its parameters A_C/A_T , K and t_l
 100 [18,26,34,35].

101 Inorganic compounds are here not directly considered due to their difficult measurement and their possible non-
 102 homogeneous distribution, but they are assumed as available. At the same time, an optimal day/night period is
 103 assumed to consider the influence of the light. These come from the choice of the experimental dataset (see
 104 Section 2.2). Both these assumptions can be considered as conservative standpoints.

105 Equation (2) shows the analytical translation of the previous requirements.

$$X(T, RH, P, R, t) = \Omega(RH) \cdot \frac{A_C}{A_T}(T, P, R) \cdot \left[1 - \exp^{-K(T, P, R) \cdot (t - t_l(T, P, R))^4} \right] \quad (2)$$

106 where the $\Omega(RH)$ is defined as in (3) and $5^\circ\text{C} \leq T \leq 40^\circ\text{C}$.

$$\Omega(RH) = \begin{cases} 0, & RH < 98\% \\ 1, & RH \geq 98\% \end{cases} \quad (3)$$

107 Besides, two analytical requirements over the time t can be stated, as reported in (4).

$$\begin{cases} X(t) = 0, \forall t < t_l(T, P, R) \\ X(t_i) \leq X(t_{i+1}), \forall t_i < t_{i+1} \end{cases} \quad (4)$$

108 The first one involves the latency time t_l and it tries to overcome an analytical inaccuracy. In fact, the use of t_l
 109 can lead to a miscalculation on the covered area: since the function is even, when the latency time is different
 110 from 0, the covered area at $t=0$ is higher than 0 with a decreasing trend between $t = 0$ and $t = t_l$. This means that
 111 the growth curve minimum is equal to 0 when $t = t_l$, too. This is not so correct from a physical description of
 112 microalgae growth, even if t_l is usually very short if compared to the total growth time and the predicted
 113 biofouling coverage in this interval is rather poor. Thus, microalgae growth is set to be 0 until the latency time is
 114 reached. The second condition states that the model has to be a monotonically not decreasing function, that is,
 115 the reached covered area can be constant or it can increase, according to the environmental conditions. In fact,

116 previous researches showed that, once settled, microalgae are able to retain water inside them and therefore
 117 survive i.e. dry periods [2,44].

118 2.2 The experimental dataset and domains determination

119 For the experimental dataset based on fired brick substrates, no novel experimental tests are performed, but a
 120 very large dataset coming from a previous work [18] is used. This dataset is in fact: (1) comparable to other ones
 121 of previous failure model used in literature [19,39,45,46] and (2) representative of the main influencing growing
 122 factors for microalgae, such as P and R . Moreover, it considers two of the most recurrent species of microalgae
 123 on building materials [6], namely *Chlorella mirabilis* (green microalga) and *Chroococcidiopsis fissurarum*
 124 (cyanobacterium), by accounting for them an optimal day/night period equal to 14/10 h respectively [18] and an
 125 adequate level of inorganic compound, according to ASTM D5589-09 standard method [47].

126 Table 1 summarizes the substrate properties (P and R) of the five fired bricks (SP₁, ..., SP₅), and the seven
 127 different environmental combinations of T and RH (EC₁, ..., EC₇).

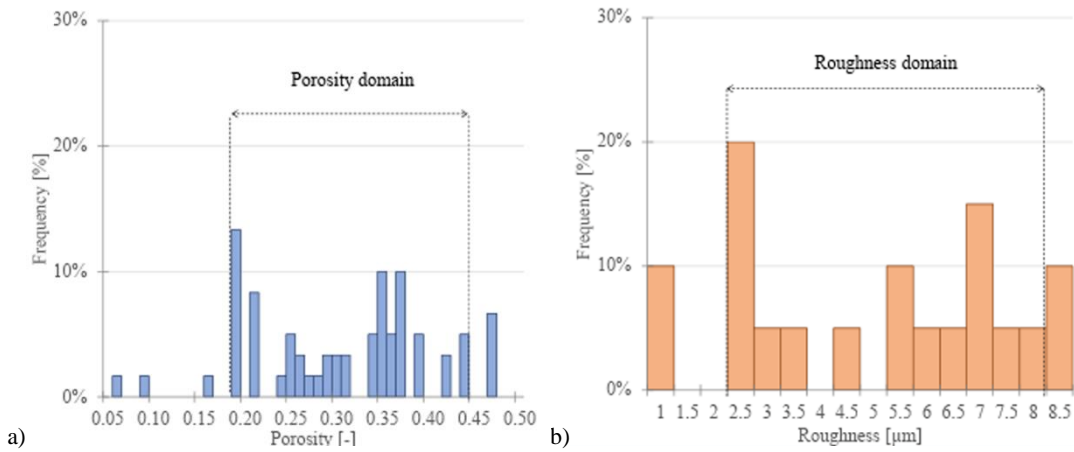
128 Table 1. Combination of the tested substrate properties (SP) and environmental conditions (EC) [18]. Three samples were tested for each substrate
 129 property (SP).

Substrate Properties			Environmental Conditions							
	Porosity [-]	Roughness [μm]	Temperature [$^{\circ}\text{C}$] – Relative humidity [%]							
			EC ₁	EC ₂	EC ₃	EC ₄	EC ₅	EC ₆	EC ₇	
SP ₁	0.19	4.50								
SP ₂	0.19	5.54								
SP ₃	0.25	2.95	T=27.5 RH=75	T=27.5 RH=87	T=27.5 RH=98	T=5 RH \approx 100	T=10 RH \approx 100	T=27.5 $^{\circ}\text{C}$ RH \approx 100	T=40 RH \approx 100	
SP ₄	0.44	6.60								
SP ₅	0.44	7.60								

130
 131 Materials SP₁, SP₃ and SP₅ are considered for the fitting process since they are comprehensive of the substrate
 132 domain, representing the minimum, maximum and intermediate values for both P and R . On the other hand, SP₂
 133 and SP₄ are used in the post fitting process in order to validate its results. All the tested environmental conditions
 134 are taken into account in the fitting process. Hence, the dataset for the experimental fitting process is composed
 135 by 63 experimental microalgae growth curves, referring to 3 samples for 3 substrates under 7 different
 136 environmental conditions and by 42 curves for the validation step.

137

138 Common P and R values of fired bricks were also investigated [25–27,29,35,48–54] to see how the experimental
 139 data set (Table 1) is representative. This way, an application range for the empirical failure model can be provided.
 140 The review described 60 different brick porosity and 20 roughness values, respectively (Figure 1). It is worth
 141 noting that the (open) porosity and the roughness, usually considered in literature [25,27,29,35,48,54,55] as
 142 microalgae growing factors, are those determined according to the ASTM D4404-10 standard [56] and UNI EN
 143 ISO 4287:2009 standard¹ [57], respectively. We will refer to such references in the following.



144
 145 Figure 1. Comparison between porosity and roughness values from literature [25–27,29,35,48–54] and the porosity and roughness domain from the
 146 selected database [18].

147 By comparing the literature review results and the experimental dataset reported in Table 1, the domain for P
 148 and R is set as reported in (5).

$$\begin{cases} 0.19 \leq P \leq 0.44 \\ 2.50 \mu m \leq R \leq 8.00 \mu m \end{cases} \quad (5)$$

149 In particular, it is worth noting that the porosity domain set for the model includes 87% samples' values, while
 150 the roughness domain covers 80% of samples provided by literature.

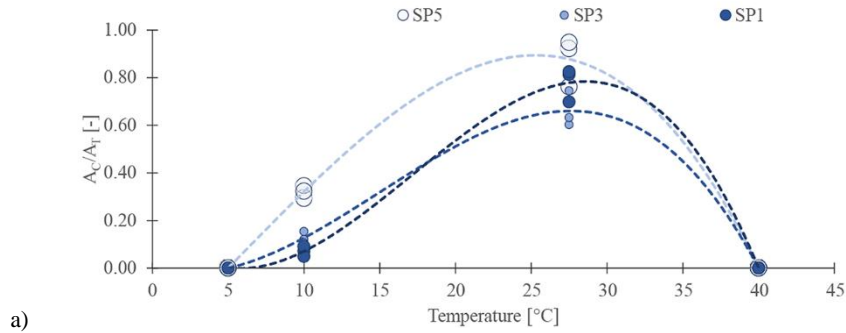
151 2.3 Experimental trend and analytical model definition

152 Figure 2 shows the experimental trend of the parameter A_C/A_T , K and t_I obtained from the used dataset: the
 153 parameter values refer to the specimens of the tested materials (SP₁, SP₃ and SP₅) under saturation condition (as
 154 reported in EC₄, ..., EC₇), for a total of 36 experimental data for each parameter. In the between of domain, T

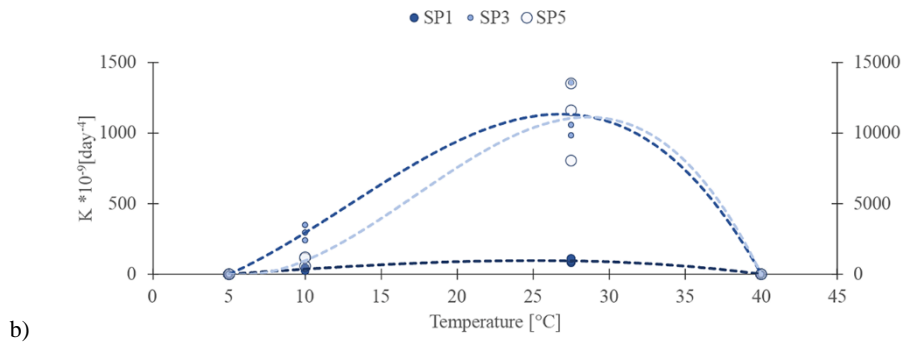
¹ Roughness values were determined according to R_a calculation.

155 predominantly influences A_C/A_T and K , determining an increasing trend from 5°C to about 27.5°C and a
 156 decreasing one from about 27.5°C to 40°C. These trends hold for each P and R . Lastly, in Figure 2 (c) the latency
 157 time t_l shows a constant trend not depending on temperature, conversely, it is influenced by P and R .

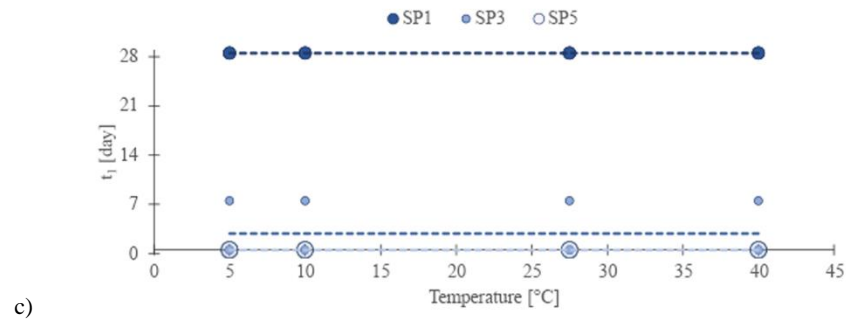
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161 Figure 2. Experimental trend of bricks parameters: (a) A_C/A_T parameter; (b) K parameter; (c) t_l parameter. The graphs are reported according to the
 162 temperature domain. The blue scale (dark-light) indicates the increasing porosity; the increasing dimension of the spot indicates the increasing
 163 roughness value. In (b) two y-axis were used since the K parameters are significantly different: SP_1 and SP_3 refer to the left y-axis, SP_5 refers to the
 164 right y-axis.

165 In this way, aiming at determining the simplest possible model, having only 4 different temperature values, a 3rd
 166 degree polynomial is set in order to describe A_C/A_T and K as functions of T , having its coefficients depending on
 167 P and R . For the t_l parameter, a constant coefficient depending only on P and R is set. The above observations
 168 are reported in (6):

$$\begin{cases} \frac{A_C}{A_T}(T, P, R) = c_{0,A}(P, R) + c_{1,A}(P, R) \cdot T + c_{2,A}(P, R) \cdot T^2 + c_{3,A}(P, R) \cdot T^3 \\ K(T, P, R) = c_{0,K}(P, R) + c_{1,K}(P, R) \cdot T + c_{2,K}(P, R) \cdot T^2 + c_{3,K}(P, R) \cdot T^3 \\ t_1(P, R) = c_{0,t}(P, R) \end{cases} \quad (6)$$

169 where the coefficients reported in (6) result from the fitting process (see Section 2.4).

170 Finally, the codomains of each involved parameter are reported in equation (7):

$$\begin{cases} 0 \leq \frac{A_C}{A_T}(T, P, R) \leq 1 \\ K(T, P, R) \geq 0 \quad , \quad \forall T \in [5; 40], \forall P \in [0.19; 0.44], \forall R \in [2.50; 8.00] \\ t_1(P, R) \geq 0 \end{cases} \quad (7)$$

171 2.4 The fitting process

172 From condition (6), a linear system of the temperature coefficient can be set as in (8).

$$\begin{cases} c_{n,A} = \alpha_{n,1,A}F_{1,A}(P, R) + \alpha_{n,2,A}F_{2,A}(P, R) + \alpha_{n,3,A}F_{3,A}(P, R) \\ c_{n,K} = \alpha_{n,1,K}F_{1,K}(P, R) + \alpha_{n,2,K}F_{2,K}(P, R) + \alpha_{n,3,K}F_{3,K}(P, R), \quad n = 0, \dots, 3 \\ c_{0,t} = \alpha_{0,1,t}F_{1,t}(P, R) + \alpha_{0,2,t}F_{2,t}(P, R) + \alpha_{0,3,t}F_{3,t}(P, R) \end{cases} \quad (8)$$

173 The F_1, \dots, F_3 considered the effect of P and R values and they are defined in (9)

$$\begin{cases} F_1(P, R) \in \{P^{j_1}, R^{k_1}, 0\} \\ F_2(P, R) \in \{P^{j_2}, R^{k_2}, 0\}, \quad j_1, \dots, j_3, k_1, \dots, k_3 \in \mathbb{Z} \\ F_3(P, R) \in \{P^{j_3}, R^{k_3}, 0\} \end{cases} \quad (9)$$

174 This means that the combination of F_1, \dots, F_3 can consider only P element, conversely only R elements, it may be

175 resumed in a constant value (e.g. for j and k exponent resulting equal to 0) or even it can result equal to 0. This is

176 due because the trends for both P and R were not clearly obtainable from the experimental evidences [18], and

177 therefore determined in advanced as for temperature.

178 Hence, once set conditions (8)-(9), the fitting process is iteratively run by determining the α values and the right
 179 combination of F_1, \dots, F_3 for the three parameters, following what is reported in (10) where F_1, \dots, F_3 have the
 180 same degree for each surface properties SP.

$$\begin{cases} \begin{matrix} \alpha_{0,1} & \alpha_{0,2} & \alpha_{0,3} \\ \alpha_{1,1} & \alpha_{1,2} & \alpha_{1,3} \\ \alpha_{2,1} & \alpha_{2,2} & \alpha_{2,3} \\ \alpha_{3,1} & \alpha_{3,2} & \alpha_{3,3} \end{matrix} \begin{matrix} F_1(P, R) \\ F_2(P, R) \\ F_3(P, R) \end{matrix} \Big|_{SP1} = \begin{matrix} c_0 \\ c_1 \\ c_2 \\ c_3 \end{matrix} \Big|_{SP1} \\ \begin{matrix} \alpha_{0,1} & \alpha_{0,2} & \alpha_{0,3} \\ \alpha_{1,1} & \alpha_{1,2} & \alpha_{1,3} \\ \alpha_{2,1} & \alpha_{2,2} & \alpha_{2,3} \\ \alpha_{3,1} & \alpha_{3,2} & \alpha_{3,3} \end{matrix} \begin{matrix} F_1(P, R) \\ F_2(P, R) \\ F_3(P, R) \end{matrix} \Big|_{SP3} = \begin{matrix} c_0 \\ c_1 \\ c_2 \\ c_3 \end{matrix} \Big|_{SP3} \\ \begin{matrix} \alpha_{0,1} & \alpha_{0,2} & \alpha_{0,3} \\ \alpha_{1,1} & \alpha_{1,2} & \alpha_{1,3} \\ \alpha_{2,1} & \alpha_{2,2} & \alpha_{2,3} \\ \alpha_{3,1} & \alpha_{3,2} & \alpha_{3,3} \end{matrix} \begin{matrix} F_1(P, R) \\ F_2(P, R) \\ F_3(P, R) \end{matrix} \Big|_{SP5} = \begin{matrix} c_0 \\ c_1 \\ c_2 \\ c_3 \end{matrix} \Big|_{SP5} \end{cases} \quad (10)$$

181 The process stops when the following two requirements are achieved. The first requirement verifies the analytical
 182 correctness of the model by determining the adjusted coefficient of determination R^2_{adj} since a multiple variable
 183 regression is considered. The calculation of R^2_{adj} is shown in equation (11) and all the R^2_{adj} have to be higher than
 184 0.85. **Such limit was previously adopted in other models to consider the accuracy of the fitting good [58–60].**

$$R^2_{adj} = 1 - \frac{RSS}{TSS} \cdot \frac{n-1}{n-p-1} \geq 0.85 \quad (11)$$

185 The RSS is the residual sum of squares between the experimental and the fitted data, TSS is the total sum of
 186 squares of the differences between the experimental data and its mean, n is the number of observation and p is
 187 the total number of explanatory variables in the model [61].

188 Subsequently, the second requirement involves the experimental correctness of the model. It is satisfied when
 189 conditions reported in (12) are fulfilled.

$$\left\{ \begin{array}{l} \min \left[\frac{A_C}{A_{T \text{ exp}}} \right] \leq \frac{A_C}{A_T}(T, P, R) \leq \max \left[\frac{A_C}{A_{T \text{ exp}}} \right] \\ \min \left[K_{\text{exp}} \right] \leq K(T, P, R) \leq \max \left[K_{\text{exp}} \right] \\ \min \left[t_{1,\text{exp}} \right] \leq t_1(P, R) \leq \max \left[t_{1,\text{exp}} \right] \end{array} \right. \quad (12)$$

190 After having defined all the parameters with their coefficients, the failure model is tested on SP₂ and SP₄ so as to
 191 have a first validation, following the same criteria: R^2_{adj} still has to be higher than 0.85, as analytical requirement
 192 and the experimental correctness must be verified by conditions reported in (12).

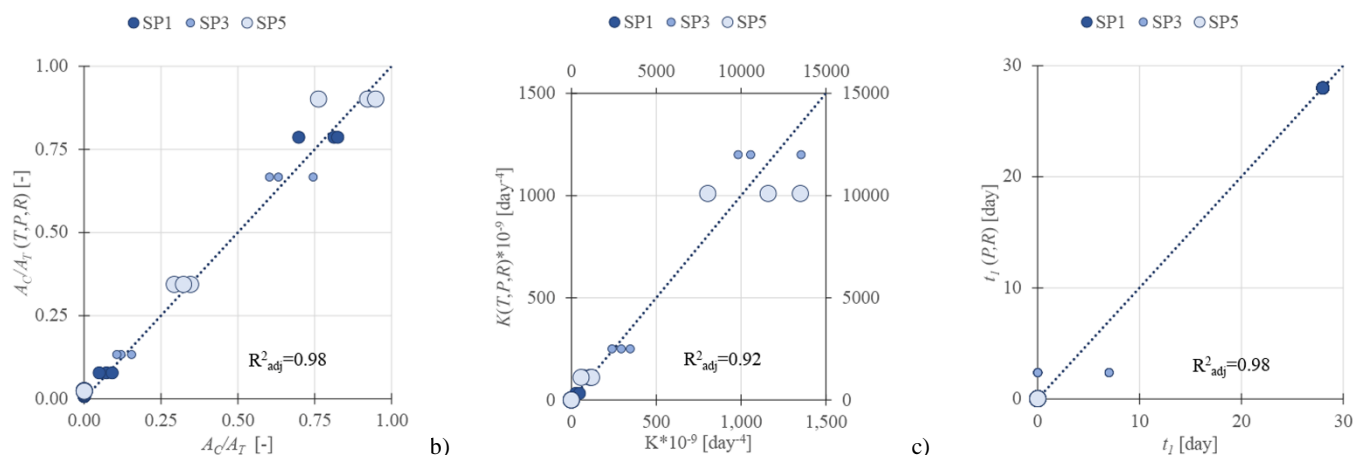
193 3 Results

194 3.1 The failure model for bricks

195 Condition (13) summarise the resulting α values and the combinations of F_1, \dots, F_3 of each parameter, by also
 196 resuming condition (6) for the temperature equations.

$$\left. \begin{aligned}
& \frac{A_C}{A_T}(T, P, R) = \begin{vmatrix} -3.419[-] & 9.2 \cdot 10^{-2} \left[\frac{1}{\mu m} \right] & -5.7 \cdot 10^{-3} \left[\frac{1}{\mu m^2} \right] \\ 8.798 \cdot 10^{-1} \left[\frac{1}{^\circ C} \right] & -3.1032 \cdot 10^{-2} \left[\frac{1}{\mu m \cdot ^\circ C} \right] & 2.16 \cdot 10^{-3} \left[\frac{1}{\mu m^2 \cdot ^\circ C} \right] \\ -3.98 \cdot 10^{-2} \left[\frac{1}{^\circ C^2} \right] & 2.8023 \cdot 10^{-3} \left[\frac{1}{\mu m \cdot ^\circ C^2} \right] & -2.184 \cdot 10^{-4} \left[\frac{1}{\mu m^2 \cdot ^\circ C^2} \right] \\ 5 \cdot 10^{-4} \left[\frac{1}{^\circ C^3} \right] & -5.21 \cdot 10^{-5} \left[\frac{1}{\mu m \cdot ^\circ C^3} \right] & 4.198 \cdot 10^{-6} \left[\frac{1}{\mu m^2 \cdot ^\circ C^3} \right] \end{vmatrix} \begin{vmatrix} P^2 \\ R \\ R^2 \end{vmatrix} \begin{vmatrix} T^0 \\ T^1 \\ T^2 \\ T^3 \end{vmatrix} \\
& K(T, P, R) = \begin{vmatrix} -1.018137 \cdot 10^{-6} [day^{-4}] & 2.638435 \cdot 10^{-5} [day^{-4}] & -1.109344 \cdot 10^{-3} [\mu m^8 \cdot day^{-4}] \\ 3.832828 \cdot 10^{-7} \left[\frac{day^{-4}}{^\circ C} \right] & -1.056712 \cdot 10^{-5} \left[\frac{day^{-4}}{^\circ C} \right] & 2.729806 \cdot 10^{-4} \left[\frac{\mu m^8 \cdot day^{-4}}{^\circ C} \right] \\ -3.978387 \cdot 10^{-8} \left[\frac{day^{-4}}{^\circ C^2} \right] & 1.173803 \cdot 10^{-6} \left[\frac{day^{-4}}{^\circ C^2} \right] & -1.081282 \cdot 10^{-5} \left[\frac{\mu m^8 \cdot day^{-4}}{^\circ C^2} \right] \\ 7.709831 \cdot 10^{-10} \left[\frac{day^{-4}}{^\circ C^3} \right] & -2.315303 \cdot 10^{-8} \left[\frac{day^{-4}}{^\circ C^3} \right] & 1.169671 \cdot 10^{-7} \left[\frac{\mu m^8 \cdot day^{-4}}{^\circ C^3} \right] \end{vmatrix} \begin{vmatrix} P \\ P^3 \\ R^{-8} \end{vmatrix} \begin{vmatrix} T^0 \\ T^1 \\ T^2 \\ T^3 \end{vmatrix} \\
& t_1(P, R) = \begin{vmatrix} 4.73 \cdot 10^{-5} [day] & -2.88 \cdot 10^{-4} \left[\frac{day}{\mu m} \right] & -2.66 \cdot 10^{-4} \left[\frac{day}{\mu m^2} \right] \end{vmatrix} \begin{vmatrix} P^{-8} \\ R \\ R^2 \end{vmatrix}
\end{aligned} \right\} \quad (13)$$

197 Figure 3 shows that all the R^2_{adj} are higher than 0.85 for A_C/A_T , K and t_I .



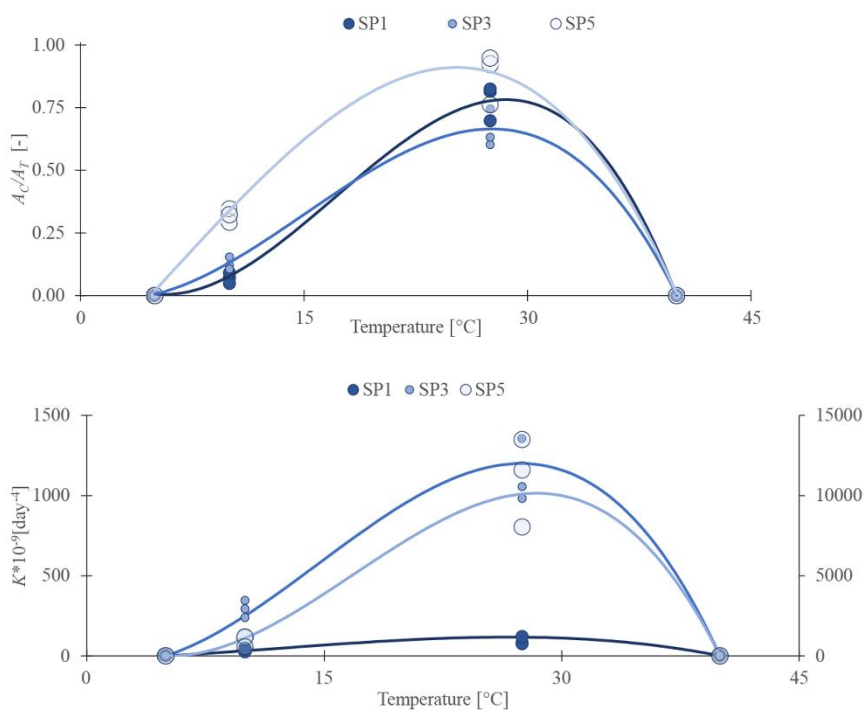
198 a)

b)

c)

199 Figure 3. Coefficient of determination R^2 of the parameter. (a) A_C/A_T parameters; (b) K parameter; (c) t_I parameter. The blue scale (dark-light)
 200 indicates the increasing porosity the increasing dimension of the spot indicates the increasing roughness value. In (b) two y-axis were used since the
 201 K parameters are significantly different: SP1 and SP3 refer to the left y-axis, SP5 refers to the right y-axis.

202 Lastly, both the experimental values and the fitted curves are reported in Figure 4. These last ones falls within the
 203 experimental values, verifying the condition proposed in (12).

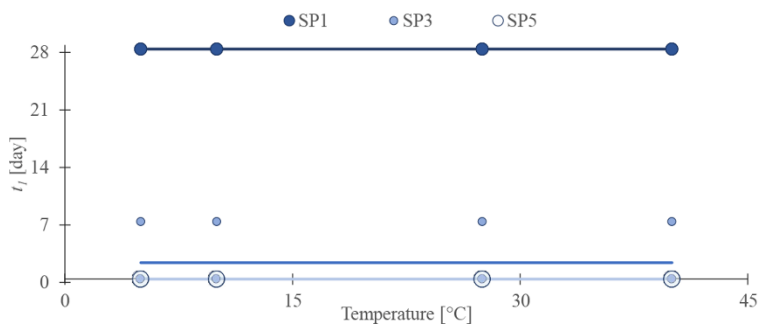


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a)

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b)



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c)

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Figure 4. Comparison between the the experimental values and the fitted curves for the surfaces properties SP₁ SP₃ and SP₅: (a) A_c/A_T (b) K and (c) t_f . The blue scale (dark-light) indicates the increasing porosity; the increasing dimension of the spot indicates the increasing roughness value. In (b) two y-axis were used since the K parameters are significantly different: SP₁ SP₃ refer to the left y-axis, SP₅ refers to the right y-axis.

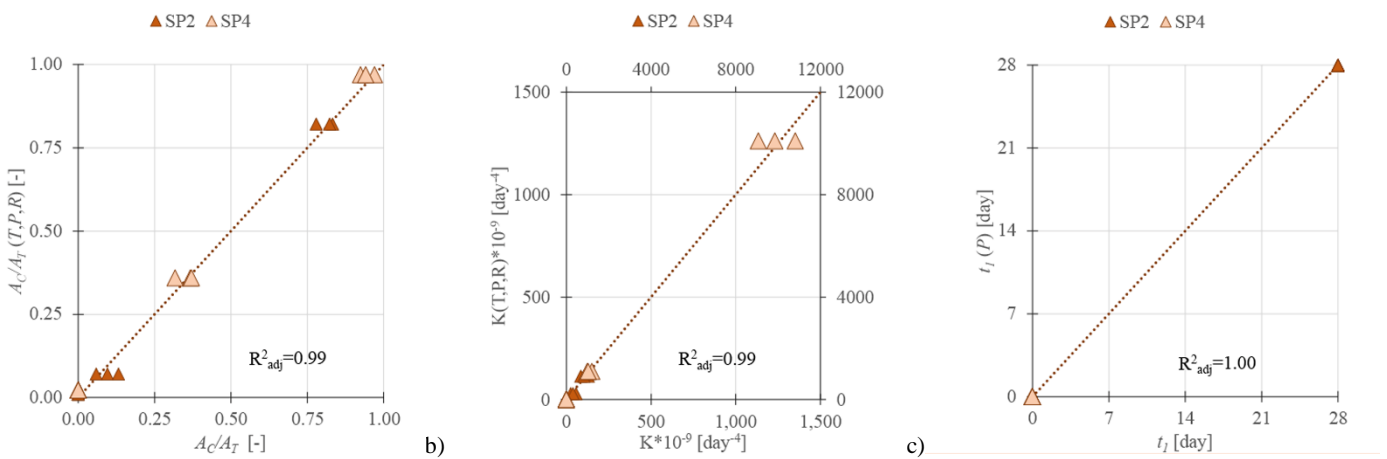
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3.2 Failure model validation

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The failure model is then applied on the second dataset. As shown in Figure 5, the three R^2_{adj} were always higher than 0.85.



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a)

b)

c)

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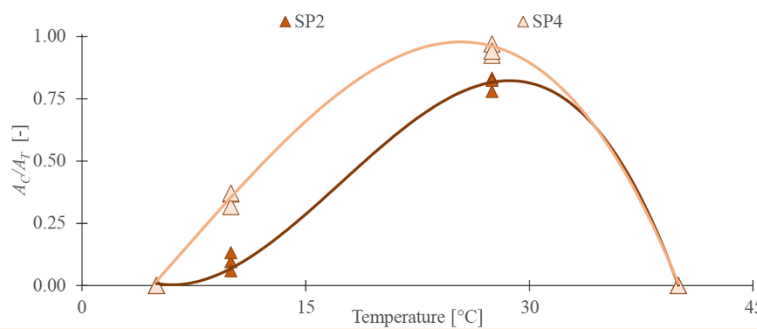
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Figure 5. Coefficient of determination R^2 of the parameters. (a) A_c/A_T parameters; (b) K parameter; (c) t_f parameter. The red scale (dark-light) indicates the increasing porosity; the increasing dimension of the spot indicates the increasing roughness value. In (b) two y-axis were used since the K parameters are significantly different: SP₂ refer to the left y-axis, SP₄ refers to the right y-axis.

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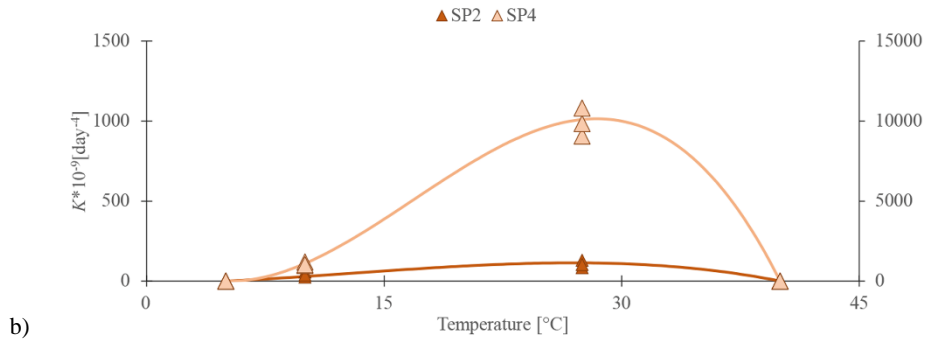
Figure 6, instead, shows that the curves determined by the failure model fell within the range of their respective experimental data.



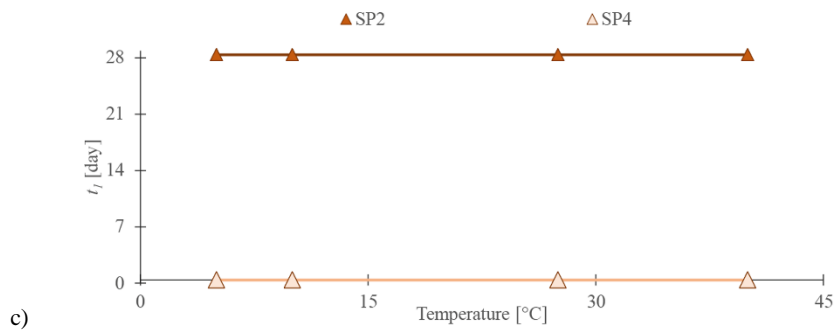
219

a)

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Figure 6. Comparison between the the experimental values and the fitted curves for the suces properties SP₂ and SP₄: (a) A_C/A_T (b) K and (c) t_f . The dotted red lines indicate the minimum and maximum curve for P and R values. The red scale (dark-light) indicates the increasing porosity; the dimension of the spot (6-7) indicates the roughness value. In (b) two y-axis were used since the K parameters are significantly different: SP₂ refers to the left y-axis, SP₄ refers to the right y-axis.

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As a last qualitative validation step, according to other previous model validation [39,46], the curves describing microalgae growth $X(t, T, RH, P, R)$ are determined for the validation substrates SP₂ and SP₄ under the tested environmental condition EC₅ and EC₆ (Table 1) and overlapped to the experimental data obtained in [18]. All the curves well fit the experimental values (Figure 7).

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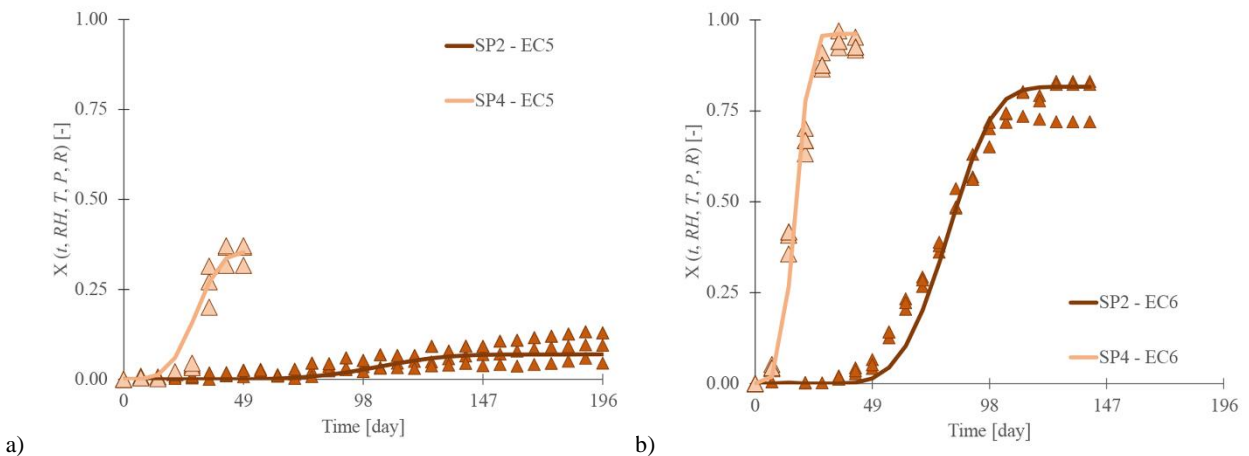


Figure 7. Comparison of the covered area $X(t, T, RH, P, R)$ obtained with the failure model and experimental data for SP₂, and SP₄ [18]:a) when exposed to EC₅;b) when exposed to EC₆. Lines indicate the failure model curves; points indicate the experimental data.

233 3.3 Failure model application over time-variable environmental conditions

234 Lastly, this paragraph shows the model application for a representative brick substrate exposed to time-variable
 235 environmental conditions (Table 2).

236 Table 2. Specification for the model application.

Material			Condition (C)	
Porosity [-]	Roughness [μm]	n°	Temperature [$^\circ\text{C}$]	Exposure Time [day]
0.19	2.75 μm	1	14	50
		2	7.5	50
		3	20	50
		4	27.5	50

237

238 The brick properties are chosen in order to describe the most recurrent ones according to Figure 1. The assumed
 239 four conditions do not simulate a real dataset, but they allow improving the readability of the combination process.
 240 In particular, the gradient between the four temperatures values allows having four distinctive branches that
 241 significantly differ from each other and the exposure time of 50 days led the growth process for each condition
 242 be clearly recognizable. Lastly, time and the time dependent variable (T and RH) are daily discretized and kept
 243 constant during the day for sake of simplicity.

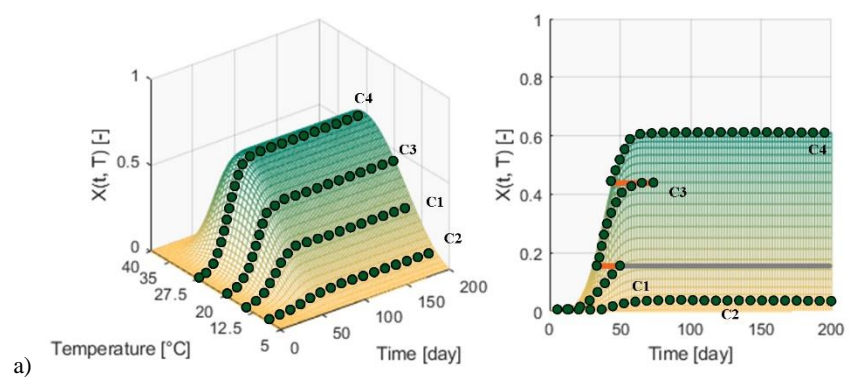
244 The combination of the branches over the time is made by following these 2 assumptions: (1) P and R are given
 245 and they both cannot change; (2) the coverage cannot go back and decrease, as already stated in equation (4).
 246 Hence, each involved n -branch of each T -dependent curve is joined to another $n+1$ -curve by simply determining
 247 the time shift $t_{s,n}$ following simple steps from equation as reported in equation (14):

$$t_{s,n} = +4 \sqrt{-\left(\frac{1}{K(T_{n+1})}\right) \cdot \ln \left(1 - \frac{X_n}{\frac{A_C}{A_T}(T_{n+1})}\right)} - t + t_1 \quad (14)$$

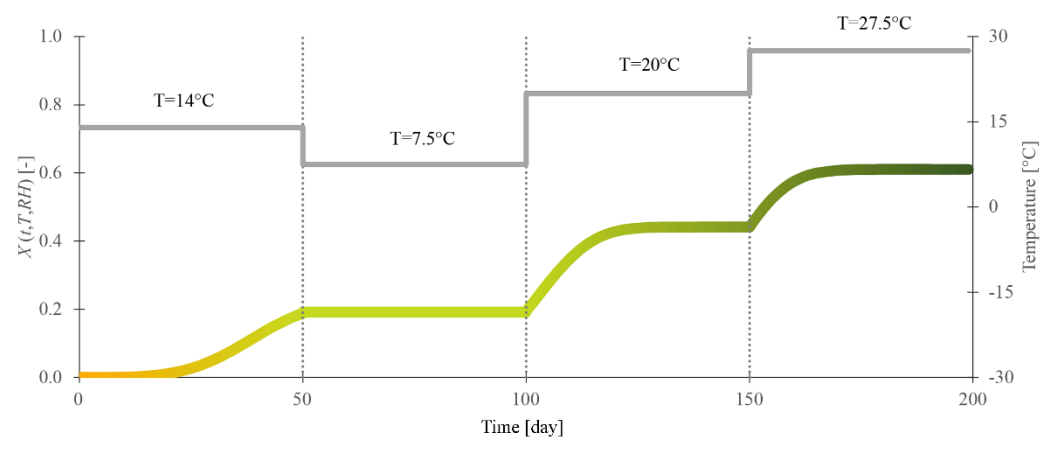
248 The logarithmic calculation is possible only if the reached covered area X_n is lower than the $A_C/A_T(T_{n+1})$,
 249 otherwise the covered area is kept constant over the time.

250 Figure 8. shows the graphical combination of such branches and the resulting curve under time variable
 251 environmental conditions. In Figure 8. (a), the T -curves are determined and each branch for the corresponding
 252 exposure time is selected. Since the covered area of C_1 ($X_{n=1}$) is higher than the A_C/A_T ($T_{n=2}$), the latency time is
 253 not determined, and hence, for all the C_2 exposure time the covered area is maintained constant. Figure 8. (b)
 254 shows the resulting combination of the branches for C_1 , C_3 and C_4 and the constant growth curve for $T=7.5^\circ\text{C}$.

255



256



257 Figure 8. Application of the model to time changing environmental conditions: a) combination methods; b) resulting curve for temperature variation
 258 over time. In (a) the covered area is following the colour scale green-yellow resembling microalgae biofouling effect; dots represent the selected
 259 curve; lines orange indicate the combination effect of the time shift; line grey indicates that it was not possible to determine the time shift; in (b) the
 260 colour scale yellow green is maintained for the total microalgae curve.

261 **4 Conclusion**

262 Failure models for biofouling on building materials are becoming a more and more unavoidable need: by making
 263 quantitative predictions, they can assist professionals and researchers in developing guidelines for interventions
 264 leading to a decrease in maintenance costs. Literature have already provided such models for mould and fungi
 265 growth, but it is still limited for microalgae growth. This work tries to fill this gap by presenting a novel empirical

266 failure model for fired bricks by taking into account the main substrate and environmental parameters influencing
267 such growth, which are porosity and roughness, as well as temperature and relative humidity. It starts from the
268 modified Avrami's model, by determining its three main parameters from experimental results, by also improving
269 it about some miscalculations in the range of the latency time. From the obtained results, it is evident that such
270 model is analytically intuitive and easy to implement. From an engineering standpoint, the novel empirical model
271 seems to be generally applicable since the tested domain of porosity and roughness covers more than 80% of the
272 fired bricks reported in literature. Finally, the application of such model considering time variable environmental
273 conditions is proposed too.

274 Future works should consider more brick type and different environmental conditions as soon as experimental
275 data will be available. Once confirmed its correctness for bricks, the model application could be extended to other
276 porous building materials (such as i.e. stones, plasters and mortars) prone to microalgae growth. Moreover, thanks
277 to its ability of considering time varying environmental conditions the model could be implemented on heat and
278 moisture simulation software, as it is already happening for other biofouling models. This will lead to the
279 application of the failure model to a real weather dataset, even considering the water content of the substrate
280 instead of solely relative humidity.

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283 sectors.

284

285 **Nomenclature**

286 *Experimental parameters by literature researches (based on modified Avrami's Theory)*

287 X covered area by microalgae biofouling [-]

288 A_C/A_T parameter of final covered area ratio [-]

289 K parameter of growth rate [day^{-4}]

290 t_l parameter of latency time [day]

291 *Fitted parameters (failure model)*

292 $X(t, T, RH, P, R)$ covered area by microalgae biofouling [-]

293 $A_C/A_T(T, P, R)$ parameter of final covered area ratio [-]

294 $K(T, P, R)$ parameter of growth rate [day^{-4}]

295 $t_l(P, R)$ parameter of latency time [day]

296 $\Omega(RH)$ on off parameter for relative humidity [-]

297 *Variables*

298 t time [day]

299 T temperature [$^{\circ}\text{C}$]

300 RH relative humidity [%]

301 P total porosity [%]

302 R roughness [μm]

303 *Coefficients*

304 c temperature equation coefficient

305 α coefficient for material properties

306 F element describing the effect of P or R

307 *Subscript and Superscript*

308 n number of temperature coefficients, from 0 to 3

309 $_{1,2,3}$ number of material coefficient for both α and F values

310	j	number of coefficient/exponents for porosity
311	k	number of coefficient/exponents for roughness
312		

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- 468

Highlights

- An empirical failure model about microalgae biofouling was developed
- The model takes into account the main substrate properties influencing the growth
- It also considers the environmental temperature and relative humidity
- The model was developed and validated on fired bricks

1 1 **An empirical failure model to predict biofouling growth on fired bricks due to** 2 3 4 5 **microalgae**

6
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15 D'Orazio).
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17 **Highlights**

- 18 - An empirical failure model about microalgae biofouling was developed
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20 - The model takes into account the main substrate properties influencing the growth
- 21
22 - It also considers the environmental temperature and relative humidity
- 23
24 - The model was developed and validated on fired bricks
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31 **Abstract**

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33 The purpose of this study was to provide an empirical failure model predicting the microalgae growth on fired
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35 bricks surfaces. It was developed through a numerical fitting of experimental data present in literature. It
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37 considered the substrate properties related to biofouling (i.e. porosity and roughness) of different bricks under
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39 several environmental conditions (i.e. relative humidity and temperature). Results shows that the model is able
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41 to simulate the microalgae biofouling by explicitly taking into account such influencing factor. Finally, this
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43 empirical failure model is validated on a different dataset from literature and applied to time varying temperature.
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50 **Keywords:** *microalgae biofouling; fired bricks; empirical failure model; substrate properties; environmental*
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53 *conditions;*
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25 **1 Introduction**

26 ³When porous building materials are exposed to environmental weathering, their physicals and chemicals
27 ⁵properties interact with biological factors, leading to changes in both its compositional and structural
28 ⁸characteristics [1–3]. The growth process and vegetative development of organisms have a direct consequence
29 ¹⁰on the material due to the metabolic activity connected with the growth of living organisms [4]. The living species
30 ¹³that commonly dwell on these materials are ranging from microscopical bacterial cells to higher plants and
31 ¹⁵animals [5].

32 ¹⁸Biofouling on porous building materials is a colonization process usually started by photoautotrophic
33 ²⁰microorganisms since they only need light, water and some inorganic components to start growing [1,2]. Among
34 ²²these, the most recurrent groups are green microalgae and cyanobacteria, shortly named as microalgae, and they
35 ²⁵usually develop in combination, especially in the European context [6–8]. Frequent maintenance and repairing
36 ²⁷interventions are then required in order to limit aesthetical, chemical and physical degradation they may produce:
37 ³⁰both ways, either repairing or not, could ultimately cause serious losses (economical or even cultural, if cultural
38 ³²heritage is involved) [9,10]. In order to describe and therefore limit microalgae biofouling risk on porous building
39 ³⁵materials, in recent years researchers adopted two strategies [11]: (1) determining and thus limiting, when
40 ³⁷possible, the influencing factors of biofouling growth; (2) providing models that can simulate and then forecast
41 ⁴⁰the biofouling risk.

42 ⁴²Regarding the first one, literature works mainly focused on the factors that influenced the water activity: that is
43 ⁴⁴defined as the water available to microorganisms to growth [6,10,12–17]. They highlighted that a combination
44 ⁴⁷of environmental conditions, substrate properties and intrinsic aspects of the plumbing system of buildings (i.e.
45 ⁴⁹leaky parts and design defects of the construction) could ensure the growth and development of microalgae. For
46 ⁵²what concerns the environmental conditions, it was demonstrated that microalgae growth occurred only at
47 ⁵⁴saturation conditions [13,18], that is when water can be found at liquid state. From an engineering standpoint,
48 ⁵⁷however, two assumption can be done: water activity can be approximated with the relative humidity RH , as
49 ⁶⁰previously demonstrated in [19] and the saturation condition safety limit can be set for $RH \geq 98\%$ [18], even though

50 ¹brick surface could not be wet, and water activity is only present by capillary condensation [20]. Moreover, it
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51 ³was proved that temperature allows microalgae to develop between about 5°C and 40°C with an optimal growth
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52 ⁵condition at about 27.5°C [18,21,22]. Out of this range, microalgae growth process is unable to start or, if already
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53 ⁸started, it stops [1,2,18,23]. In this context, porosity and roughness were outlined as the main factors concerning
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54 ¹⁰the substrate properties [13,24–27]. In fact, roughness promotes the adherence of microalgae to the substrate
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55 ¹³while porosity is responsible for retaining water inside the material structure. Moreover, when the materials
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56 ¹⁵structure and geometry, in terms of porosity and roughness, is able to afford enough water activity for microalgae
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57 ¹⁸to start their growth, the chemical composition of the substrate may play only a secondary role, favouring or
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58 ²⁰limiting their development [28–32].
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59 ²²For what concern the second strategy, literature has provided a reliable model for describing microalgae growth
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60 ²⁵starting from experimental measures [33]. Based on the Avrami's theory, it connects the area covered by
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61 ²⁷biofouling over the time by denoting a first phase of latency (when microalgae stains are not still visible),
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62 ²⁹followed by a rapid growth, and finally a stagnation phase when the covered area reaches its maximum and
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63 ³²becomes constant over the time. Such approach was applied to several type of porous buildings materials (i.e.
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64 ³⁵mortars, fired bricks and stones), under different environmental conditions and biocides surface treatments,
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65 ³⁷confirming its capabilities [18,26,33–35]. However, it never explicitly and quantitatively accounts for the growth
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66 ³⁹influencing factors of the substrate and environmental conditions, as failure models usually do. This way, no
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67 ⁴²failure models still exist in literature for microalgae growth on porous building materials, to the authors'
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68 ⁴⁴knowledge, while numerous failure models have been presented for other type of biofouling (e. g. mould, fungi,
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69 ⁴⁷actinomycetes) [19,36–43].
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70 ⁴⁹Hence, the aim of this work is to propose a first empirical microalgae growth failure model, that is able to
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71 ⁵²explicitly take into account, for the first time, the main influencing factors of substrate parameters (such as
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72 ⁵⁴porosity and roughness) and environmental conditions (such as temperature and relative humidity), as well as
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73 ⁵⁷their variation over the time. Fired bricks are here considered due to the availability of a large set of experimental
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74 ⁵⁹data in literature, but the methodology can be extended to other porous building materials when enough
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75 experimental data will be available. To this aim, this work is divided into three main phases. The first phase
 76 involves the definition of: the model general requirements (Section 2.1); the variables domains (Section 2.2) and
 77 the specific model equations (Section 2.3). The fitting method is then provided (Section 2.4). Lastly, resulting
 78 equations are presented and validated and the application of the model under time varying temperature is reported
 79 (Section 3).

80 **2 Materials and methods**

81 **2.1 General requirements**

82 As stated above, the starting point of this work is the modified Avrami's model [34,35] showed in (1):

$$X(t) = \frac{A_C}{A_T} \cdot (1 - \exp^{-K(t-t_1)^n}) \tag{1}$$

83 where the covered area by microalgae growth $X(t)$ [-] is given as a function of time t [day]. The final covered area
 84 ratio is represented by the parameter A_C/A_T which expresses the percentage of the covered area at the end of the
 85 process (A_C is the maximum covered area by biofouling on a specific sample, and A_T is the total area of the same
 86 sample), K [day^{-4}] is a rate parameter determined by the least squares method using experimental measurements.
 87 Lastly, the parameter t_1 represents the latency time [day] before a chromatic variation occurs on the material
 88 surface and the coefficient n is the Avrami's coefficient which can be assumed equal to 4 [34].

89 The model variables are chosen according to literature findings. Porosity P [-] and roughness R [μm] are the main
 90 factors characterising the substrate [6,25,26] and, their values are easily measurable and available in literature
 91 (see Section 2.2). Temperature T [$^{\circ}\text{C}$] and relative humidity RH [%] are instead selected as the main factors
 92 representing environmental conditions [13,14,18].

93 In this way:

- 94 - since literature showed that, regardless of the temperature, RH determines the actual possibility for
 95 microalgae to growth, according to which growth happens only if $RH \geq 98\%$ [18], it can be considered as
 96 an on/off factor;

97 1 - when $RH \geq 98\%$ and $5^\circ\text{C} \leq T \leq 40^\circ\text{C}$ growth can happen depending on T , P and R , otherwise it cannot
 98 3 [13,18];

99 5 - thus, P , R and T directly affect the microalgae coverage X through its parameters A_C/A_T , K and t_l
 100 8 [18,26,34,35].

101 10 Inorganic compounds are here not directly considered due to their difficult measurement and their possible non-
 102 12 homogeneous distribution, but they are assumed as available. At the same time, an optimal day/night period is
 103 15 assumed to consider the influence of the light. These come from the choice of the experimental dataset (see
 104 18 Section 2.2). Both these assumptions can be considered as conservative standpoints.

105 20 Equation (2) shows the analytical translation of the previous requirements.

$$X(T, RH, P, R, t) = \Omega(RH) \cdot \frac{A_C}{A_T}(T, P, R) \cdot \left[1 - \exp^{-K(T, P, R) \cdot (t - t_l(T, P, R))^4} \right] \quad (2)$$

106 26 where the $\Omega(RH)$ is defined as in (3) and $5^\circ\text{C} \leq T \leq 40^\circ\text{C}$.

$$\Omega(RH) = \begin{cases} 0, & RH < 98\% \\ 1, & RH \geq 98\% \end{cases} \quad (3)$$

107 32 Besides, two analytical requirements over the time t can be stated, as reported in (4).

$$\begin{cases} X(t) = 0, \forall t < t_l(T, P, R) \\ X(t_i) \leq X(t_{i+1}), \forall t_i < t_{i+1} \end{cases} \quad (4)$$

108 38 The first one involves the latency time t_l and it tries to overcome an analytical inaccuracy. In fact, the use of t_l
 109 40 can lead to a miscalculation on the covered area: since the function is even, when the latency time is different

110 43 from 0, the covered area at $t=0$ is higher than 0 with a decreasing trend between $t = 0$ and $t = t_l$. This means that
 111 45 the growth curve minimum is equal to 0 when $t = t_l$, too. This is not so correct from a physical description of

112 48 microalgae growth, even if t_l is usually very short if compared to the total growth time and the predicted
 113 50 biofouling coverage in this interval is rather poor. Thus, microalgae growth is set to be 0 until the latency time is

114 52 reached. The second condition states that the model has to be a monotonically not decreasing function, that is,
 115 55 the reached covered area can be constant or it can increase, according to the environmental conditions. In fact,

116 previous researches showed that, once settled, microalgae are able to retain water inside them and therefore
 117 survive i.e. dry periods [2,44].

118 2.2 The experimental dataset and domains determination

119 For the experimental dataset based on fired brick substrates, no novel experimental tests are performed, but a
 120 very large dataset coming from a previous work [18] is used. This dataset is in fact: (1) comparable to other ones
 121 of previous failure model used in literature [19,39,45,46] and (2) representative of the main influencing growing
 122 factors for microalgae, such as P and R . Moreover, it considers two of the most recurrent species of microalgae
 123 on building materials [6], namely *Chlorella mirabilis* (green microalga) and *Chroococcidiopsis fissurarum*
 124 (cyanobacterium), by accounting for them an optimal day/night period equal to 14/10 h respectively [18] and an
 125 adequate level of inorganic compound, according to ASTM D5589-09 standard method [47].

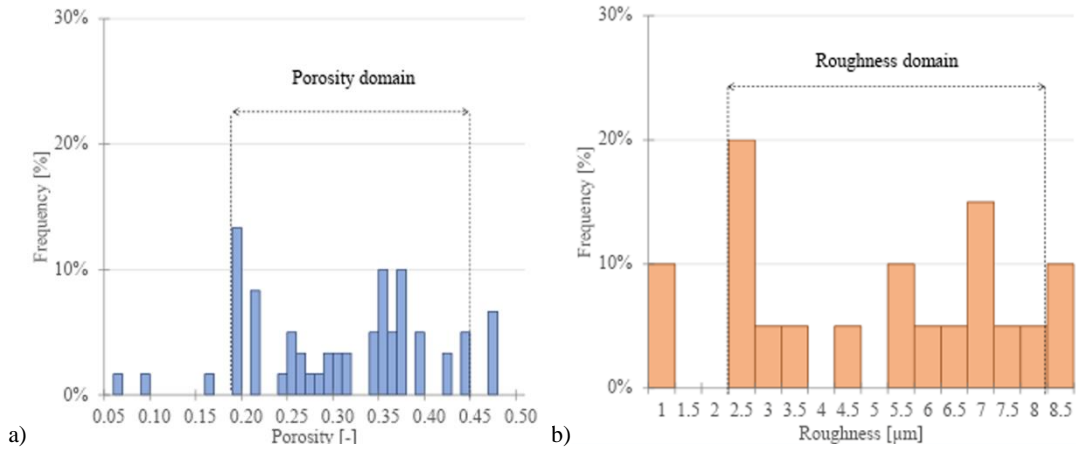
126 Table 1 summarizes the substrate properties (P and R) of the five fired bricks (SP₁, ..., SP₅), and the seven
 127 different environmental combinations of T and RH (EC₁, ..., EC₇).

128 Table 1. Combination of the tested substrate properties (SP) and environmental conditions (EC) [18]. Three samples were tested for each substrate
 129 property (SP).

	Substrate Properties		Environmental Conditions						
	Porosity [-]	Roughness [μm]	Temperature [$^{\circ}\text{C}$] – Relative humidity [%]						
			EC ₁	EC ₂	EC ₃	EC ₄	EC ₅	EC ₆	EC ₇
SP ₁	0.19	4.50							
SP ₂	0.19	5.54							
SP ₃	0.25	2.95	T=27.5 RH=75	T=27.5 RH=87	T=27.5 RH=98	T=5 RH \approx 100	T=10 RH \approx 100	T=27.5 $^{\circ}\text{C}$ RH \approx 100	T=40 RH \approx 100
SP ₄	0.44	6.60							
SP ₅	0.44	7.60							

130 Materials SP₁, SP₃ and SP₅ are considered for the fitting process since they are comprehensive of the substrate
 131 domain, representing the minimum, maximum and intermediate values for both P and R . On the other hand, SP₂
 132 and SP₄ are used in the post fitting process in order to validate its results. All the tested environmental conditions
 133 are taken into account in the fitting process. Hence, the dataset for the experimental fitting process is composed
 134 by 63 experimental microalgae growth curves, referring to 3 samples for 3 substrates under 7 different
 135 environmental conditions and by 42 curves for the validation step.

138 Common P and R values of fired bricks were also investigated [25–27,29,35,48–54] to see how the experimental
 139 data set (Table 1) is representative. This way, an application range for the empirical failure model can be provided.
 140 The review described 60 different brick porosity and 20 roughness values, respectively (Figure 1). It is worth
 141 noting that the (open) porosity and the roughness, usually considered in literature [25,27,29,35,48,54,55] as
 142 microalgae growing factors, are those determined according to the ASTM D4404-10 standard [56] and UNI EN
 143 ISO 4287:2009 standard¹ [57], respectively. We will refer to such references in the following.



145¹ Figure 1. Comparison between porosity and roughness values from literature [25–27,29,35,48–54] and the porosity and roughness domain from the
 146² selected database [18].

147³ By comparing the literature review results and the experimental dataset reported in Table 1, the domain for P
 148⁴ and R is set as reported in (5).

$$\begin{cases} 0.19 \leq P \leq 0.44 \\ 2.50 \mu m \leq R \leq 8.00 \mu m \end{cases} \quad (5)$$

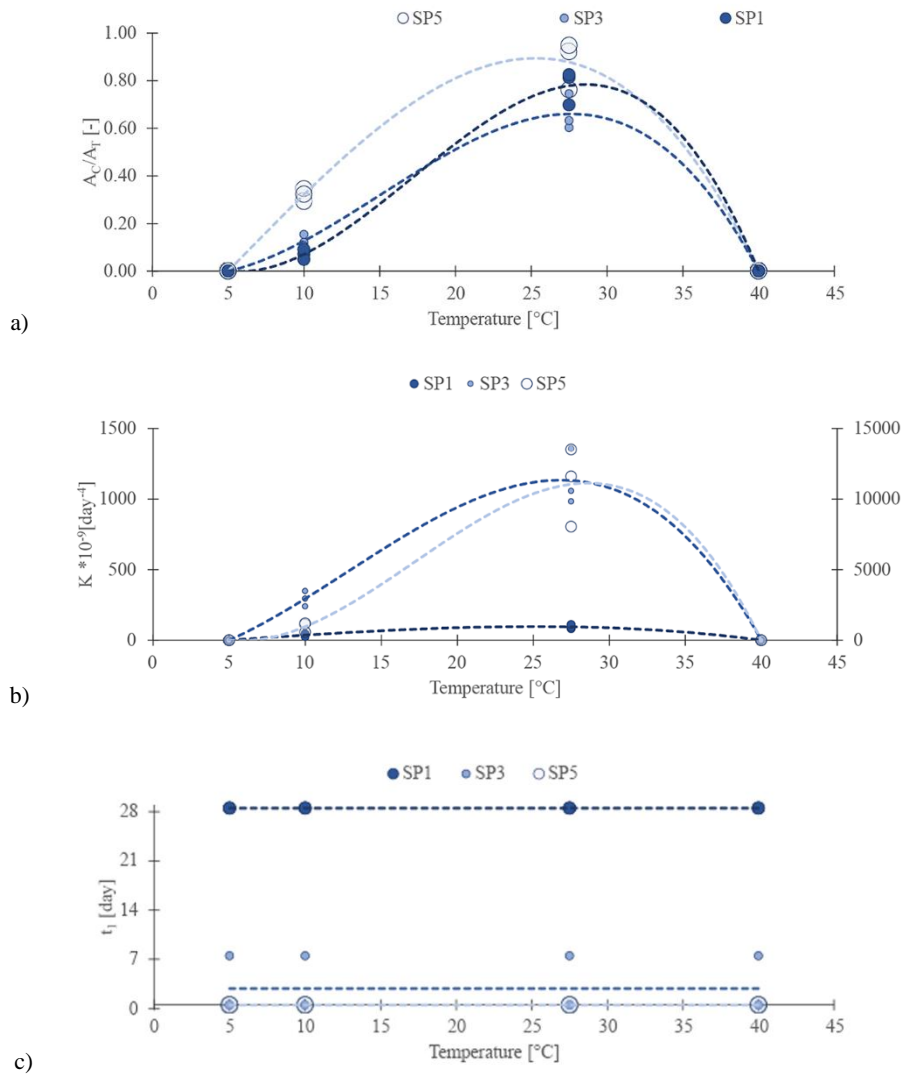
149⁵ In particular, it is worth noting that the porosity domain set for the model includes 87% samples' values, while
 150⁶ the roughness domain covers 80% of samples provided by literature.

151⁷ 2.3 Experimental trend and analytical model definition

152⁸ Figure 2 shows the experimental trend of the parameter A_C/A_T , K and t_I obtained from the used dataset: the
 153⁹ parameter values refer to the specimens of the tested materials (SP₁, SP₃ and SP₅) under saturation condition (as
 154¹⁰ reported in EC₄, ..., EC₇), for a total of 36 experimental data for each parameter. In the between of domain, T

159¹¹ ¹Roughness values were determined according to R_a calculation.

155 predominantly influences A_C/A_T and K , determining an increasing trend from 5°C to about 27.5°C and a
 2
 156 decreasing one from about 27.5°C to 40°C. These trends hold for each P and R . Lastly, in Figure 2 (c) the latency
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 157 time t_l shows a constant trend not depending on temperature, conversely, it is influenced by P and R .



161 Figure 2. Experimental trend of bricks parameters: (a) A_C/A_T parameter; (b) K parameter; (c) t_l parameter. The graphs are reported according to the
 162 temperature domain. The blue scale (dark-light) indicates the increasing porosity; the increasing dimension of the spot indicates the increasing
 163 roughness value. In (b) two y-axis were used since the K parameters are significantly different: SP₁ and SP₃ refer to the left y-axis, SP₅ refers to the
 164 right y-axis.

165 In this way, aiming at determining the simplest possible model, having only 4 different temperature values, a 3rd
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 166 degree polynomial is set in order to describe A_C/A_T and K as functions of T , having its coefficients depending on
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 167 P and R . For the t_l parameter, a constant coefficient depending only on P and R is set. The above observations
 55
 168 are reported in (6):

$$\begin{cases}
\frac{A_C}{A_T}(T, P, R) = c_{0,A}(P, R) + c_{1,A}(P, R) \cdot T + c_{2,A}(P, R) \cdot T^2 + c_{3,A}(P, R) \cdot T^3 \\
K(T, P, R) = c_{0,K}(P, R) + c_{1,K}(P, R) \cdot T + c_{2,K}(P, R) \cdot T^2 + c_{3,K}(P, R) \cdot T^3 \\
t_1(P, R) = c_{0,t}(P, R)
\end{cases} \quad (6)$$

where the coefficients reported in (6) result from the fitting process (see Section 2.4).

Finally, the codomains of each involved parameter are reported in equation (7):

$$\begin{cases}
0 \leq \frac{A_C}{A_T}(T, P, R) \leq 1 \\
K(T, P, R) \geq 0 \quad , \quad \forall T \in [5; 40], \forall P \in [0.19; 0.44], \forall R \in [2.50; 8.00] \\
t_1(P, R) \geq 0
\end{cases} \quad (7)$$

2.4 The fitting process

From condition (6), a linear system of the temperature coefficient can be set as in (8).

$$\begin{cases}
c_{n,A} = \alpha_{n,1,A}F_{1,A}(P, R) + \alpha_{n,2,A}F_{2,A}(P, R) + \alpha_{n,3,A}F_{3,A}(P, R) \\
c_{n,K} = \alpha_{n,1,K}F_{1,K}(P, R) + \alpha_{n,2,K}F_{2,K}(P, R) + \alpha_{n,3,K}F_{3,K}(P, R), \quad n = 0, \dots, 3 \\
c_{0,t} = \alpha_{0,1,t}F_{1,t}(P, R) + \alpha_{0,2,t}F_{2,t}(P, R) + \alpha_{0,3,t}F_{3,t}(P, R)
\end{cases} \quad (8)$$

The F_1, \dots, F_3 considered the effect of P and R values and they are defined in (9)

$$\begin{cases}
F_1(P, R) \in \{P^{j_1}, R^{k_1}, 0\} \\
F_2(P, R) \in \{P^{j_2}, R^{k_2}, 0\}, \quad j_1, \dots, j_3, k_1, \dots, k_3 \in \mathbb{Z} \\
F_3(P, R) \in \{P^{j_3}, R^{k_3}, 0\}
\end{cases} \quad (9)$$

This means that the combination of F_1, \dots, F_3 can consider only P element, conversely only R elements, it may be

resumed in a constant value (e.g. for j and k exponent resulting equal to 0) or even it can result equal to 0. This is

due because the trends for both P and R were not clearly obtainable from the experimental evidences [18], and

therefore determined in advanced as for temperature.

178 Hence, once set conditions (8)-(9), the fitting process is iteratively run by determining the α values and the right
 179 combination of F_1, \dots, F_3 for the three parameters, following what is reported in (10) where F_1, \dots, F_3 have the
 180 same degree for each surface properties SP.

$$\begin{cases}
 \begin{pmatrix} \alpha_{0,1} & \alpha_{0,2} & \alpha_{0,3} \\ \alpha_{1,1} & \alpha_{1,2} & \alpha_{1,3} \\ \alpha_{2,1} & \alpha_{2,2} & \alpha_{2,3} \\ \alpha_{3,1} & \alpha_{3,2} & \alpha_{3,3} \end{pmatrix} \begin{vmatrix} F_1(P, R) \\ F_2(P, R) \\ F_3(P, R) \end{vmatrix}_{SP1} = \begin{vmatrix} c_0 \\ c_1 \\ c_2 \\ c_3 \end{vmatrix}_{SP1} \\
 \begin{pmatrix} \alpha_{0,1} & \alpha_{0,2} & \alpha_{0,3} \\ \alpha_{1,1} & \alpha_{1,2} & \alpha_{1,3} \\ \alpha_{2,1} & \alpha_{2,2} & \alpha_{2,3} \\ \alpha_{3,1} & \alpha_{3,2} & \alpha_{3,3} \end{pmatrix} \begin{vmatrix} F_1(P, R) \\ F_2(P, R) \\ F_3(P, R) \end{vmatrix}_{SP3} = \begin{vmatrix} c_0 \\ c_1 \\ c_2 \\ c_3 \end{vmatrix}_{SP3} \\
 \begin{pmatrix} \alpha_{0,1} & \alpha_{0,2} & \alpha_{0,3} \\ \alpha_{1,1} & \alpha_{1,2} & \alpha_{1,3} \\ \alpha_{2,1} & \alpha_{2,2} & \alpha_{2,3} \\ \alpha_{3,1} & \alpha_{3,2} & \alpha_{3,3} \end{pmatrix} \begin{vmatrix} F_1(P, R) \\ F_2(P, R) \\ F_3(P, R) \end{vmatrix}_{SP5} = \begin{vmatrix} c_0 \\ c_1 \\ c_2 \\ c_3 \end{vmatrix}_{SP5}
 \end{cases} \quad (10)$$

181 The process stops when the following two requirements are achieved. The first requirement verifies the analytical
 182 correctness of the model by determining the adjusted coefficient of determination R^2_{adj} since a multiple variable
 183 regression is considered. The calculation of R^2_{adj} is shown in equation (11) and all the R^2_{adj} have to be higher than
 184 0.85. Such limit was previously adopted in other models to consider the accuracy of the fitting good [58–60].

$$R^2_{adj} = 1 - \frac{RSS}{TSS} \cdot \frac{n-1}{n-p-1} \geq 0.85 \quad (11)$$

185 The RSS is the residual sum of squares between the experimental and the fitted data, TSS is the total sum of
 186 squares of the differences between the experimental data and its mean, n is the number of observation and p is
 187 the total number of explanatory variables in the model [61].

188 Subsequently, the second requirement involves the experimental correctness of the model. It is satisfied when
 189 conditions reported in (12) are fulfilled.

$$\left\{ \begin{array}{l} \min \left[\frac{A_C}{A_{T \text{ exp}}} \right] \leq \frac{A_C}{A_T}(T, P, R) \leq \max \left[\frac{A_C}{A_{T \text{ exp}}} \right] \\ \min \left[K_{\text{exp}} \right] \leq K(T, P, R) \leq \max \left[K_{\text{exp}} \right] \\ \min \left[t_{1,\text{exp}} \right] \leq t_1(P, R) \leq \max \left[t_{1,\text{exp}} \right] \end{array} \right. \quad (12)$$

After having defined all the parameters with their coefficients, the failure model is tested on SP₂ and SP₄ so as to have a first validation, following the same criteria: R²_{adj} still has to be higher than 0.85, as analytical requirement and the experimental correctness must be verified by conditions reported in (12).

Results

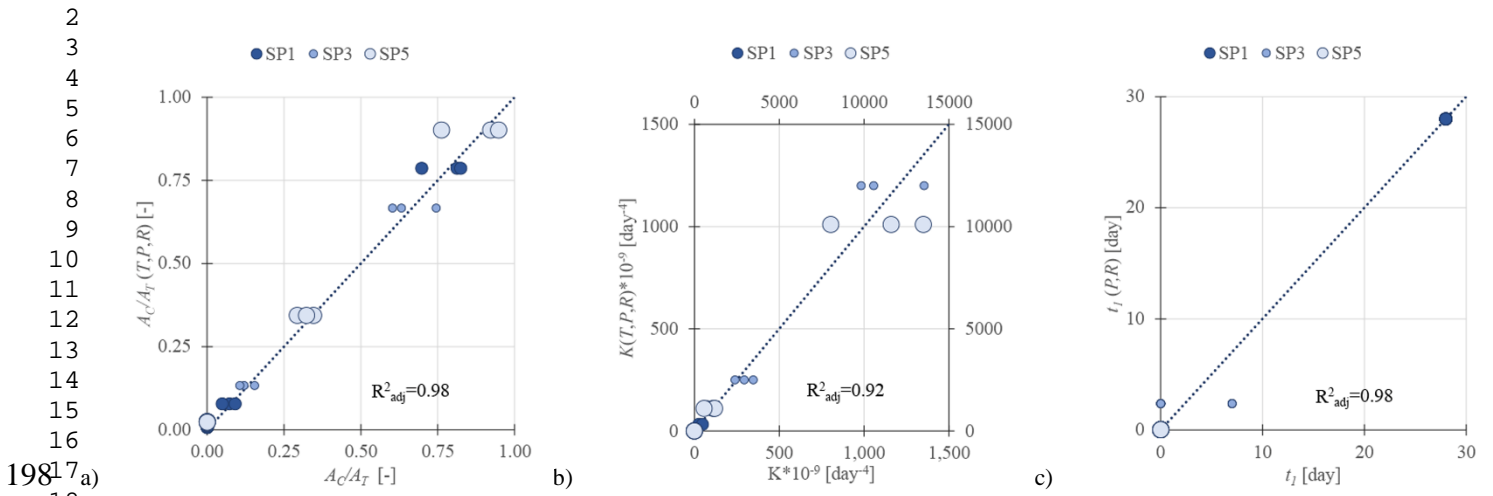
3.1 The failure model for bricks

Condition (13) summarise the resulting α values and the combinations of F₁, ..., F₃ of each parameter, by also resuming condition (6) for the temperature equations.

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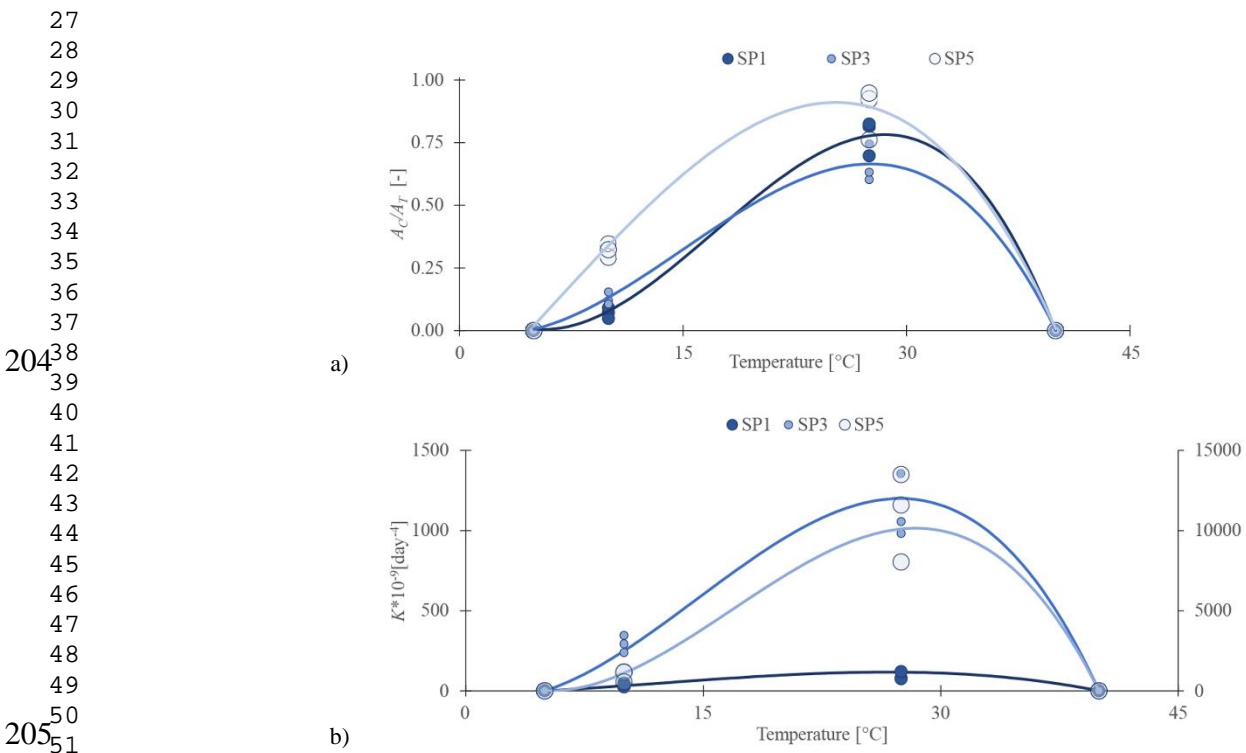
$$\left. \begin{aligned}
 & \frac{A_C}{A_T}(T, P, R) = \begin{vmatrix} -3.419[-] & 9.2 \cdot 10^{-2} \left[\frac{1}{\mu m} \right] & -5.7 \cdot 10^{-3} \left[\frac{1}{\mu m^2} \right] \\ 8.798 \cdot 10^{-1} \left[\frac{1}{^\circ C} \right] & -3.1032 \cdot 10^{-2} \left[\frac{1}{\mu m \cdot ^\circ C} \right] & 2.16 \cdot 10^{-3} \left[\frac{1}{\mu m^2 \cdot ^\circ C} \right] \\ -3.98 \cdot 10^{-2} \left[\frac{1}{^\circ C^2} \right] & 2.8023 \cdot 10^{-3} \left[\frac{1}{\mu m \cdot ^\circ C^2} \right] & -2.184 \cdot 10^{-4} \left[\frac{1}{\mu m^2 \cdot ^\circ C^2} \right] \\ 5 \cdot 10^{-4} \left[\frac{1}{^\circ C^3} \right] & -5.21 \cdot 10^{-5} \left[\frac{1}{\mu m \cdot ^\circ C^3} \right] & 4.198 \cdot 10^{-6} \left[\frac{1}{\mu m^2 \cdot ^\circ C^3} \right] \end{vmatrix} \begin{vmatrix} P^2 \\ R \\ R^2 \end{vmatrix} \cdot \begin{vmatrix} T^0 \\ T^1 \\ T^2 \\ T^3 \end{vmatrix} \\
 & K(T, P, R) = \begin{vmatrix} -1.018137 \cdot 10^{-6} [day^{-4}] & 2.638435 \cdot 10^{-5} [day^{-4}] & -1.109344 \cdot 10^{-3} [\mu m^8 \cdot day^{-4}] \\ 3.832828 \cdot 10^{-7} \left[\frac{day^{-4}}{^\circ C} \right] & -1.056712 \cdot 10^{-5} \left[\frac{day^{-4}}{^\circ C} \right] & 2.729806 \cdot 10^{-4} \left[\frac{\mu m^8 \cdot day^{-4}}{^\circ C} \right] \\ -3.978387 \cdot 10^{-8} \left[\frac{day^{-4}}{^\circ C^2} \right] & 1.173803 \cdot 10^{-6} \left[\frac{day^{-4}}{^\circ C^2} \right] & -1.081282 \cdot 10^{-5} \left[\frac{\mu m^8 \cdot day^{-4}}{^\circ C^2} \right] \\ 7.709831 \cdot 10^{-10} \left[\frac{day^{-4}}{^\circ C^3} \right] & -2.315303 \cdot 10^{-8} \left[\frac{day^{-4}}{^\circ C^3} \right] & 1.169671 \cdot 10^{-7} \left[\frac{\mu m^8 \cdot day^{-4}}{^\circ C^3} \right] \end{vmatrix} \begin{vmatrix} P \\ P^3 \\ R^{-8} \end{vmatrix} \cdot \begin{vmatrix} T^0 \\ T^1 \\ T^2 \\ T^3 \end{vmatrix} \\
 & t_1(P, R) = \begin{vmatrix} 4.73 \cdot 10^{-5} [day] & -2.88 \cdot 10^{-4} \left[\frac{day}{\mu m} \right] & -2.66 \cdot 10^{-4} \left[\frac{day}{\mu m^2} \right] \end{vmatrix} \begin{vmatrix} P^{-8} \\ R \\ R^2 \end{vmatrix}
 \end{aligned} \right\} \quad (13)$$

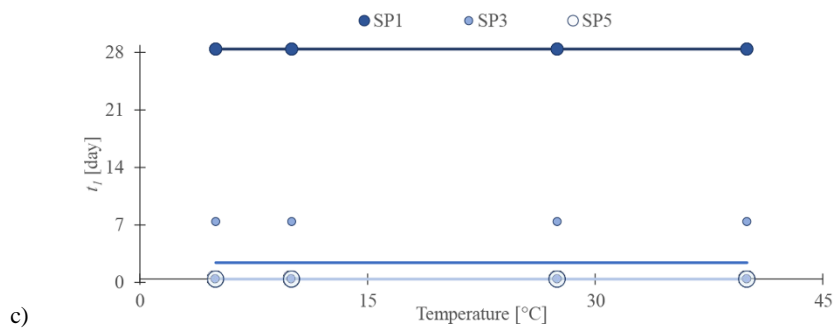
197 Figure 3 shows that all the R^2_{adj} are higher than 0.85 for A_C/A_T , K and t_I .



199 Figure 3. Coefficient of determination R^2 of the parameter. (a) A_C/A_T parameters; (b) K parameter; (c) t_I parameter. The blue scale (dark-light) indicates the increasing porosity the increasing dimension of the spot indicates the increasing roughness value. In (b) two y-axis were used since the K parameters are significantly different: SP1 and SP3 refer to the left y-axis, SP5 refers to the right y-axis.

202 Lastly, both the experimental values and the fitted curves are reported in Figure 4. These last ones falls within the experimental values, verifying the condition proposed in (12).

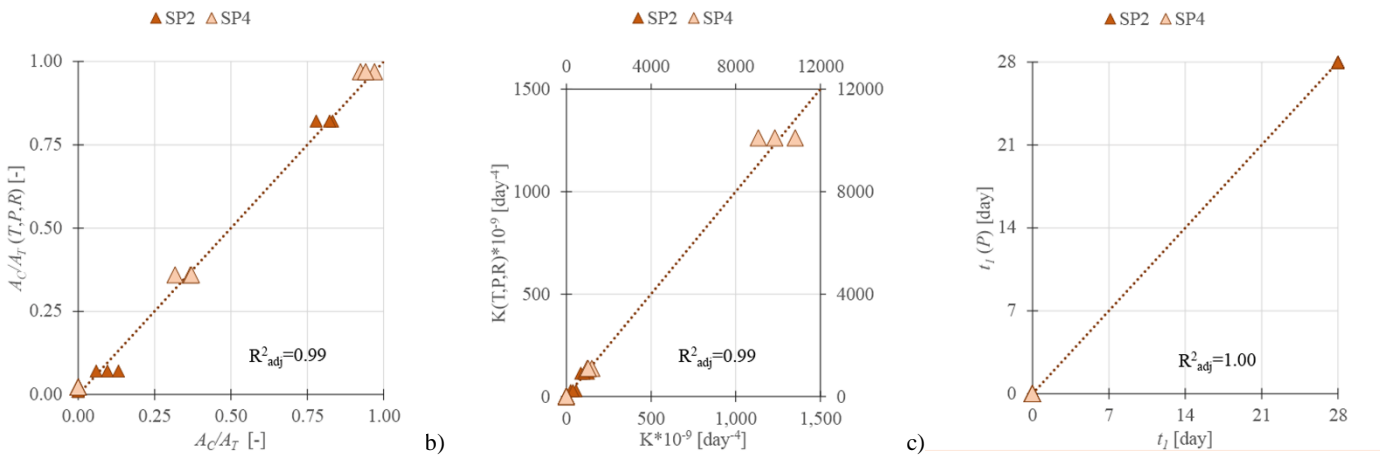




2071 3 Figure 4. Comparison between the the experimental values and the fitted curves for the surfaces properties SP₁ SP₃ and SP₅: (a) A_c/A_T (b) K and (c) t_f .
 2081 4 The blue scale (dark-light) indicates the increasing porosity; the increasing dimension of the spot indicates the increasing roughness value. In (b) two
 2091 5 y-axis were used since the K parameters are significantly different: SP₁ SP₃ refer to the left y-axis, SP₅ refers to the right y-axis.

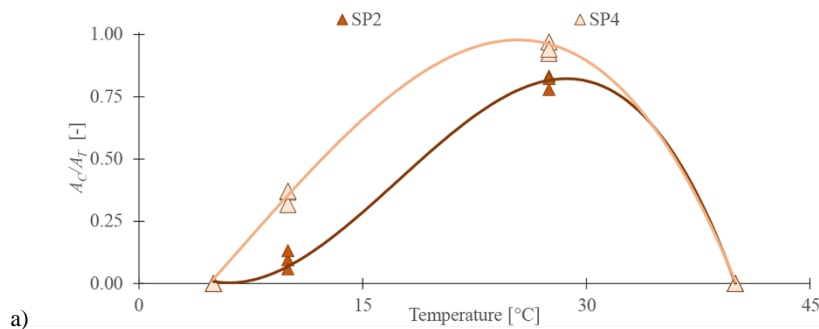
2101 7 3.2 Failure model validation

2111 9 The failure model is then applied on the second dataset. As shown in Figure 5, the three R^2_{adj} were always higher
 2122 2 than 0.85.



2144 0 Figure 5. Coefficient of determination R^2 of the parameters. (a) A_c/A_T parameters; (b) K parameter; (c) t_f parameter. The red scale (dark-light)
 2154 1 indicates the increasing porosity; the increasing dimension of the spot indicates the increasing roughness value. In (b) two y-axis were used since the
 2164 2 K parameters are significantly different: SP₂ refer to the left y-axis, SP₄ refers to the right y-axis.

2174 4 Figure 6, instead, shows that the curves determined by the failure model fell within the range of their respective
 2184 7 experimental data.



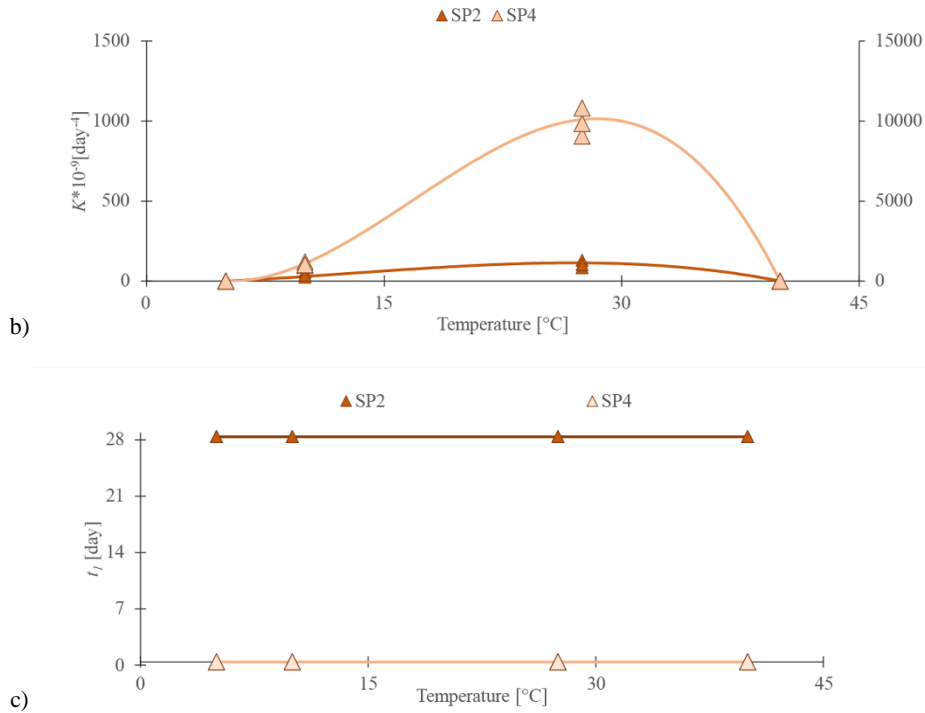


Figure 6. Comparison between the the experimental values and the fitted curves for the surfaces properties SP₂ and SP₄: (a) A_C/A_T (b) K and (c) t_l . The dotted red lines indicate the minimum and maximum curve for P and R values. The red scale (dark-light) indicates the increasing porosity; the dimension of the spot (6-7) indicates the roughness value. In (b) two y-axis were used since the K parameters are significantly different: SP₂ refers to the left y-axis, SP₄ refers to the right y-axis.

As a last qualitative validation step, according to other previous model validation [39,46], the curves describing microalgae growth $X(t, T, RH, P, R)$ are determined for the validation substrates SP₂ and SP₄ under the tested environmental condition EC₅ and EC₆ (Table 1) and overlapped to the experimental data obtained in [18]. All the curves well fit the experimental values (Figure 7).

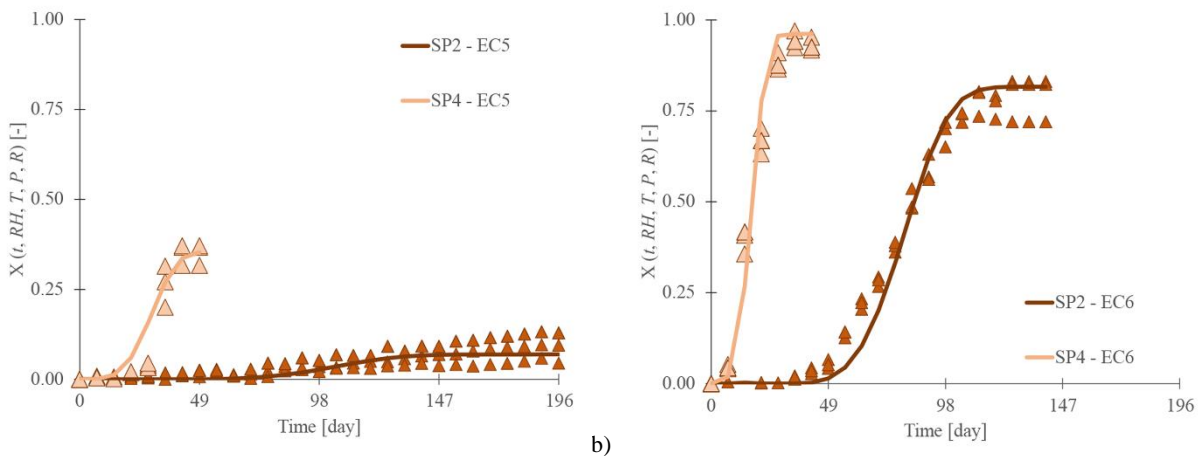


Figure 7. Comparison of the covered area $X(t, T, RH, P, R)$ obtained with the failure model and experimental data for SP₂, and SP₄ [18]:a) when exposed to EC₅;b) when exposed to EC₆. Lines indicate the failure model curves; points indicate the experimental data.

233 1.3.3 Failure model application over time-variable environmental conditions

234 Lastly, this paragraph shows the model application for a representative brick substrate exposed to time-variable
 235 environmental conditions (Table 2).

236 Table 2. Specification for the model application.

Material			Condition (C)		
Porosity [-]	Roughness [μm]	n°	Temperature [$^\circ\text{C}$]	Exposure Time [day]	
0.19	2.75 μm	1	14	50	
		2	7.5	50	
		3	20	50	
		4	27.5	50	

237 The brick properties are chosen in order to describe the most recurrent ones according to Figure 1. The assumed
 238 four conditions do not simulate a real dataset, but they allow improving the readability of the combination process.

239 In particular, the gradient between the four temperatures values allows having four distinctive branches that
 240 significantly differ from each other and the exposure time of 50 days led the growth process for each condition
 241 to be clearly recognizable. Lastly, time and the time dependent variable (T and RH) are daily discretized and kept
 242 constant during the day for sake of simplicity.

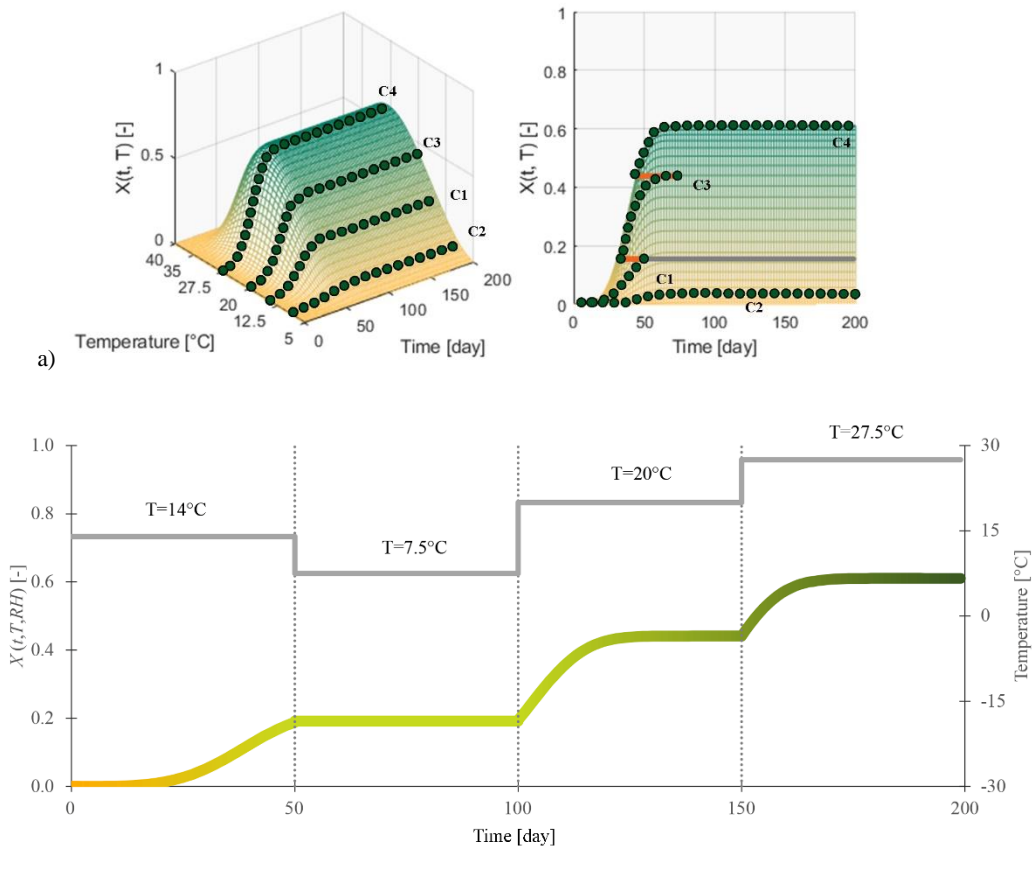
243 The combination of the branches over the time is made by following these 2 assumptions: (1) P and R are given
 244 and they both cannot change; (2) the coverage cannot go back and decrease, as already stated in equation (4).

245 Hence, each involved n -branch of each T -dependent curve is joined to another $n+1$ -curve by simply determining
 246 the time shift $t_{s,n}$ following simple steps from equation as reported in equation (14):

$$t_{s,n} = +4 \sqrt{-\left(\frac{1}{K(T_{n+1})}\right) \cdot \ln \left(1 - \frac{X_n}{\frac{A_C}{A_T}(T_{n+1})}\right)} - t + t_1 \quad (14)$$

247 The logarithmic calculation is possible only if the reached covered area X_n is lower than the $A_C/A_T(T_{n+1})$,
 248 otherwise the covered area is kept constant over the time.

250 Figure 8. shows the graphical combination of such branches and the resulting curve under time variable
 251 environmental conditions. In Figure 8. (a), the T -curves are determined and each branch for the corresponding
 252 exposure time is selected. Since the covered area of C_1 ($X_{n=1}$) is higher than the A_C/A_T ($T_{n=2}$), the latency time is
 253 not determined, and hence, for all the C_2 exposure time the covered area is maintained constant. Figure 8. (b)
 254 shows the resulting combination of the branches for C_1 , C_3 and C_4 and the constant growth curve for $T=7.5^\circ\text{C}$.



257 Figure 8. Application of the model to time changing environmental conditions: a) combination methods; b) resulting curve for temperature variation
 258 over time. In (a) the covered area is following the colour scale green-yellow resembling microalgae biofouling effect; dots represent the selected
 259 curve; lines orange indicate the combination effect of the time shift; line grey indicates that it was not possible to determine the time shift; in (b) the
 260 colour scale yellow green is maintained for the total microalgae curve.

261 Conclusion

262 Failure models for biofouling on building materials are becoming a more and more unavoidable need: by making
 263 quantitative predictions, they can assist professionals and researchers in developing guidelines for interventions
 264 leading to a decrease in maintenance costs. Literature have already provided such models for mould and fungi
 265 growth, but it is still limited for microalgae growth. This work tries to fill this gap by presenting a novel empirical

266₁ failure model for fired bricks by taking into account the main substrate and environmental parameters influencing
267₃ such growth, which are porosity and roughness, as well as temperature and relative humidity. It starts from the
268₅ modified Avrami's model, by determining its three main parameters from experimental results, by also improving
269₈ it about some miscalculations in the range of the latency time. From the obtained results, it is evident that such
270₁₀ model is analytically intuitive and easy to implement. From an engineering standpoint, the novel empirical model
271₁₃ seems to be generally applicable since the tested domain of porosity and roughness covers more than 80% of the
272₁₅ fired bricks reported in literature. Finally, the application of such model considering time variable environmental
273₁₈ conditions is proposed too.

274₂₀ Future works should consider more brick type and different environmental conditions as soon as experimental
275₂₃ data will be available. Once confirmed its correctness for bricks, the model application could be extended to other
276₂₆ porous building materials (such as i.e. stones, plasters and mortars) prone to microalgae growth. Moreover, thanks
277₂₉ to its ability of considering time varying environmental conditions the model could be implemented on heat and
278₃₁ moisture simulation software, as it is already happening for other biofouling models. This will lead to the
279₃₄ application of the failure model to a real weather dataset, even considering the water content of the substrate
280₃₇ instead of solely relative humidity.

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285 1 **Nomenclature**

286 3 *Experimental parameters by literature researches (based on modified Avrami's Theory)*

287 6 X covered area by microalgae biofouling [-]

288 8 A_C/A_T parameter of final covered area ratio [-]

289 11 K parameter of growth rate [day^{-4}]

290 14 t_l parameter of latency time [day]

291 15 *Fitted parameters (failure model)*

292 18 $X(t, T, RH, P, R)$ covered area by microalgae biofouling [-]

293 21 $A_C/A_T(T, P, R)$ parameter of final covered area ratio [-]

294 24 $K(T, P, R)$ parameter of growth rate [day^{-4}]

295 27 $t_l(P, R)$ parameter of latency time [day]

296 30 $\Omega(RH)$ on off parameter for relative humidity [-]

297 31 *Variables*

298 34 t time [day]

299 37 T temperature [$^{\circ}\text{C}$]

300 40 RH relative humidity [%]

301 43 P total porosity [%]

302 46 R roughness [μm]

303 49 *Coefficients*

304 52 c temperature equation coefficient

305 55 α coefficient for material properties

306 58 F element describing the effect of P or R

307 61 *Subscript and Superscript*

308 64 n number of temperature coefficients, from 0 to 3

309 67 $1,2,3$ number of material coefficient for both α and F values

310	1 _y	number of coefficient/exponents for porosity
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311	3 _k	number of coefficient/exponents for roughness
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Declaration of interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

Authors Statement

Quagliarini: Supervision, Validation, Writing - Review & Editing.

Gregorini: Methodology, Formal Analysis, Writing – Original Draft.

D’Orazio: Supervision, Conceptualization.