

Highlights

- Biodeterioration of synthetic polymers in conservation is a concern
- Synthetic polymer-based composites are susceptible to microbial attack
- Selected microorganisms are used to remove undesired synthetic polymers
- Current research gaps and priorities are defined

1 **Interactions of microorganisms and synthetic polymers in cultural heritage**
2 **conservation**

3
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10

11 **Abstract** Since the 1960s, synthetic polymers (at the time assumed to be resistant to
12 microbial colonization) have been employed to mitigate the ongoing deterioration of
13 cultural heritage. Since then, the use of synthetic materials in heritage conservation
14 attracted much interest. This paper reviews the last two decades of advances in the
15 relationship microorganisms - synthetic polymers in the cultural heritage conservation
16 field. Three topics are considered: (1) biodeterioration of traditional synthetic polymers,
17 (2) biosusceptibility of novel composites based on synthetic polymers, and (3) biocleaning
18 to remove undesired synthetic polymers. It is know that, if the undesired polymer
19 chemical structures are not fully known, they are particularly difficult to remove.
20 Therefore, the future employment of synthetic polymers definitely requires a more
21 critical holistic conservation assessment.

22

23 **Keywords:** biosusceptibility; biodeterioration; biocleaning; acrylics; conservation;
24 microorganisms.

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- 29 - Synthetic polymer-based composites are susceptible to microbial attack
- 30 - Selected microorganisms are used to remove undesired synthetic polymers
- 31 - Current research gaps and priorities are defined
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44 **1. Introduction**

45 The synthesis of fully synthetic polymers dates back to the 19th century. Early
46 synthetic polymers were polystyrene, polyvinyl chloride, polyisobutylene,
47 polyisoprene and polyaniline (Rasmussen, 2018). After the Second World War, new
48 classes of synthetic resins were introduced, and later in the 1950s and 1960s, as a
49 result of economic growth, polymeric materials became commonly used materials.
50 During the same period, in the sixties, it became popular to employ synthetic
51 polymers to slow down the ongoing deterioration of cultural heritage assets, since
52 it was assumed that they were more resistant than natural materials to microbial
53 colonization. Over the next forty years, according to Doehne and Price (2010), epoxy
54 resins, acrylic polymers, organic silicones and fluoropolymers were the compounds
55 most commonly used for stone conservation. Some acrylics, polyvinyl acetates and
56 alkyds were likewise employed to produce and treat other artworks such as
57 paintings (Cappitelli and Sorlini, 2008). Though the application of synthetic
58 polymers has become a routine practice in consolidation and protection of cultural

59 heritage buildings and objects, their resistance to microbial colonization is
60 questionable from past experience (Gu, 2003).

61 Many phenomena, processes and/or activities by microorganisms in the synthetic
62 polymers are embraced by the terms biodeterioration, biodegradation and
63 biosusceptibility or bioreceptivity. The most accepted definition of biodeterioration
64 is that offered by Hueck in 1965: 'any undesirable change in the properties of a
65 material caused by the vital activities of organisms'. A term very close to it, also
66 including positive connotations, is biodegradation (including biocleaning
67 approaches) of which there are several definitions. While no formal definition has
68 been consolidated, the most widely used in the building stone decay and
69 conservation area is that of Allsopp et al. (2004): 'the harnessing, by man, of the
70 decay abilities of organisms to render a waste material more useful or acceptable'.
71 Eggins and Oxley (2001), in a work originally written in 1980, discussed these two
72 terms and showed that both are identical in principle, and their distinction is only
73 made on the basis of human needs. Biodeterioration decreases the value of a material
74 of relatively high value, while biodegradation occurs in materials of little or no
75 value, e.g. for the elimination of waste (Kurowski et al., 2017). In connection with
76 this, the term biodeterioration is closely related to inorganic building materials,
77 mainly stone and masonry from heritage, while biodegradation is often associated
78 with plastic and other synthetic polymeric materials. The terms biosusceptibility or
79 bioreceptivity (the latter defined by Guillitte in 1995 as 'the ability of a material to
80 be colonised by living organisms') and biodeterioration are confused in some cases.
81 In addition to the always negative connotations of the biodeterioration concept, the
82 target objectives are the effects of colonisation on the material. In contrast, in the case
83 of bioreceptivity, the focus is on the effects of the material on colonisation.

84 The presence of microorganisms on synthetic polymers may result in biofouling, the
85 degradation of the polymer and additives/residual monomers and penetration of

86 biological structures or of metabolites like pigments into the synthetic matrix
87 (Flemming, 2010). Indeed, some lipophilic pigments are capable of diffusing into the
88 polymer matrix, leading to ineffective cleaning of the polymer surface (Flemming,
89 2010). For some of these synthetic polymers, microorganisms and enzymes involved
90 in their degradation have been described (Kurowski et al., 2017; Sabev et al., 2006;
91 Purohit et al., 2020). Gaytán et al. (2021) suggested that acrylics undergo microbial
92 degradation in two steps: the first phase is an enzymatic elimination of side groups
93 and the second step is the cleavage of the C-C backbone. Biochemical processes,
94 mechanisms and initial pioneering species involved in biodeterioration of other
95 synthetic polymers are reported in Mohanan et al. (2020). Several techniques are
96 now available that can detect the presence of microbial growth on polymeric
97 substrates, for example, infrared spectroscopy (Cappitelli et al., 2005) and laser-
98 induced fluorescence scanning (Spizzichino et al., 2015).

99 This paper presents a concise review on microorganisms - synthetic polymers
100 interactions in cultural heritage conservation related to the last two decades. The
101 three issues examined include: (1) biodeterioration of traditional synthetic polymers,
102 (2) biosusceptibility of composites based on synthetic polymers, and (3) biocleaning
103 to remove undesired synthetic polymers.

104

105 **2. Synthetic polymers as a habitat for biological colonization**

106 *2.1 A growing concern*

107 Despite the fact that biocides are typical additives in artists' acrylic emulsion paints
108 (Learner, 2007), according to the Smithsonian Museum Conservation Institute,
109 fungal development on acrylic paintings is a rising concern among artists and
110 collectors

111 (https://www.si.edu/mci/english/learn_more/taking_care/acrylic_paintings.html#:~:text=Mold%20growth%20has%20been%20noted,when%20humidity%20and%20te

113 temperature%20rise). The American institute stated that no ideal conservation
114 treatment exists and intervention always causes some degree of damage to the
115 original paint.

116 This is not surprising as, in some instances, synthetic polymers in paints in cans have
117 been found to be already contaminated by microorganisms, especially fungi, mainly
118 *Rhizopus* and *Aspergillus* spp., before application (Okunye et al., 2013). In particular,
119 water-borne coating paints are potentially prone to in-can attacks as a consequence
120 of the use of contaminated recycled water, leading for example to loss of viscosity,
121 pH changes, sedimentation and separation into phases, discoloration, and potential
122 health hazards (Okunye et al., 2013). Similarly, in the interior of spray paint cans in
123 black, red and white colours, *Pantoea* sp., *Bacillus megaterium* and *Pseudomonas*
124 *mendocina* were found, all of them with potential biocleaning capacity against spray
125 alkyd and polyester resins paint graffiti (Sanmartín et al., 2015).

126 Primal AC33 and Plexisol P550 40TB 1997 (both purchased in 1997), Primal AC33
127 (purchased at the beginning of the 1990s), and Paraloid B72 (purchased in 1966) kept
128 in can for decades were tested according to the American Society for Testing and
129 Materials “Standard Practice for Determining Resistance of Synthetic Polymeric
130 Materials to Fungi” (ASTM G21-96(2002), growth rate 0-4, 0=no growth; 4=massive
131 growth) (Cappitelli et al., 2008). In contrast to the results obtained with the same
132 synthetic resins recently acquired, the polymers naturally aged in cans were more
133 susceptible to fungal attack. Specifically, Primal AC33 showed a level of growth 1;
134 Plexisol P550 40TB, growth rate 3; and Paraloid B72, growth rate 4.

135 It has been stated several times that synthetic polymers used for conservation
136 treatment can support the growth of microbes and thus be a possible habitat where
137 the conditions that define the niche of the taxon are met (Runeberg, 2008). Indeed,
138 early experiments showing evidence that some fungi were able to grow on a

139 synthetic adhesive, Vinavil Emulsion (polyvinylacetate), were conducted as early as
140 the 1950s at the Istituto Centrale del Restauro in Rome, Italy (Giacobini, 1957; 1958).

141

142 *2.2. Synthetic polymers change microbial ecology*

143 When biological growth occurs in the added exogenous material, such as the
144 synthetic polymers, a case of extrinsic bioreceptivity according to Guillitte (1995)
145 and quaternary bioreceptivity according to Sanmartín et al. (2021a) is taking place.
146 According to Guillitte (1995) when particles or substances that are not part of the
147 material are deposited and accumulated on the material and the colonisation
148 developed from there no longer relates directly to the properties of the starting
149 material, the bioreceptivity is extrinsic. According to Sanmartín et al. (2021a), when
150 new materials, such as any coating or chemical product leaving residues, are added
151 to the material, its bioreceptivity becomes quaternary. In this sense, the addition of
152 polymers onto a surface may change the physical properties like the wetting-drying
153 kinetics, for example, it may increase the water (and nutrient) retention inside the
154 fissures and voids in the polymeric coating. These cracks also enhance the anchorage
155 of colonizing cells (**Figure 1**), which added to the previous aspect, leads to biofilm
156 formation and, as a consequence, its bioreceptivity increasing (Quagliarini et al.,
157 2018; Sanmartín et al., 2021b).

158 Paraloid B72-treated and untreated painted specimens from the chapel of the Holy
159 Nail in what is now Santa Maria della Scala Museum, Siena (Italy) showed potential
160 deteriogen taxa in a quiescent state in both, while bacteria were mainly retrieved on
161 untreated samples (Milanesi et al., 2009). This difference in the microbial community
162 demonstrates that bioreceptivity is affected by the presence of the acrylic polymer.
163 The same was true for unpainted and concrete sites with acrylic paint coatings in
164 Georgia, USA, showing that the presence of polymers can affect the microbial
165 ecology (Giannantonio et al., 2009). While studying the Klippe statues, a group of

166 ancient Chinese stone Buddhas in Hangzhou, China, and the feasibility of using
167 synthetic polymers for their conservation, Li et al. (2017) tested lipolytic and ester-
168 hydrolytic activity of the microorganisms they isolated to study potential
169 biodeterioration mechanisms of synthetic materials. Production of esterases was
170 evaluated on mineral medium with added Tween 20®, while Tween 80® and
171 tributyrin were employed as substrates for lipases. Enzymatic tests showed that the
172 majority of bacterial isolates produce lipases while esterases were produced by a
173 few microorganisms.

174 Unlike in buried environments where wooden materials are barely biodeteriorated
175 because few microorganisms are able to degrade lignin anaerobically (Caneva et al.,
176 2008; Billings et al., 2015), wooden surfaces are easily biodeteriorated in subaerial
177 environments and therefore have been sometimes protected from biological attack
178 with a synthetic polymer (Czajnik, 1968). In research by De Souza and Gaylarde
179 (2002), pinewood was brushed with a transparent alkyd varnish (Tintas Renner,
180 Gravatai, Brazil) and inoculated with fungi, bacteria and algae above or below the
181 coating. The latter case simulated the polymer application over contaminated
182 surfaces and the researchers observed that varying initial contamination could lead
183 to different deterioration results.

184 Finally, the presence of synthetic polymers can influence how deeply endolithically
185 the microbial community can grow. This is the case of the Grande Albergo Ausonia
186 & Hungaria (Venice Lido, Italy), which has an Art Nouveau tiles on its façade, which
187 underwent restoration in 2007 including treatment with Paraloid B72. On studying
188 the microbial structure of the community, it was observed that when Paraloid B72
189 was detected, the biofilm was growing in a deeper position than in uncoated
190 specimens (Giacomucci et al., 2011), as also previously reported (Ariño and Saiz-
191 Jimenez, 1996).

192

193 2.3. *Chemical structure, ageing and application method affect biosusceptibility*

194 In the review by Cappitelli and Sorlini (2008) it was reported that when some
195 polymers were synthesized in the laboratory (with the advantage of having only the
196 polymer to deal with and not the presence of additives), the general order of
197 susceptibility to fungi according to the ASTM G21-96(2002) was alkyds > polyvinyl
198 acetates > acrylics. Investigating the biodegradation of alkyd resins used in
199 household paints and artists' paints, Doménech-Carbó et al. (2008) showed that
200 microorganisms led to the hydrolysis of the polymeric resins with the formation of
201 short-chain fatty acids. In a work by the same team (Doménech-Carbó et al., 2009)
202 microbial biodeterioration of PVA emulsions coincided with the decrease in the
203 relative content of external plasticiser of the phthalate type employed. In addition, a
204 different behaviour was observed, depending on the plasticiser used in each PVA
205 emulsion product investigated.

206 The paint of a cracked pineapple in the artwork 'Teca con Frutta' by Massimo
207 Zuppelli (1967) was found to be made of cellulose nitrate and alkyd resin (Macro et
208 al., 2020). The paint was deteriorated by fungi, as proven by infrared spectroscopy
209 and scanning electron microscopy. Consequently, conservators applied an
210 antifungal biocide as part of the cleaning treatment. When specific attention was
211 paid to the chemical composition, it was noted that the combined use of acrylic and
212 fluorinated monomers promoted the fungal deterioration of the polymeric material
213 (Sabatini et al., 2018).

214 Many studies show that these synthetic materials are more bioreceptive (more
215 suitable for biological colonization and its development) when the synthetic
216 polymer is physically and chemically degraded (Cappitelli and Sorlini, 2008). For
217 example, in the acrylic resins of the marble-built Tempio Malatestiano in Rimini,
218 Italy (Pinna and Salvadori, 1999) and the Milan cathedral, Italy (Cappitelli et al.,
219 2007), fungi colonized the synthetic material, rather than lithic material, causing its

220 physical disruption with biopitting and the formation of cracks and fissures (**Figure**
221 **2**). Moreover, black fungi were isolated from the Milan cathedral stone and the fungi
222 of the standard ASTM G21-96(2002) method were used to investigate whether the
223 detected poly-isobutylmethacrylate and epoxy resin could be used as the sole
224 sources of carbon and energy (Cappitelli et al., 2007). The freshly dried acrylic resin
225 was susceptible to an isolated *Cladosporium* sp. (Cappitelli et al., 2007). Wall
226 paintings of UNESCO Wu-Kui Tomb in China were treated with acrylic varnish B01-
227 6 applied by spraying to protect them (Sun et al., 2019). On the B01-6 coatings area
228 on the murals seasonally 'white moldy spots' were observed and some
229 microorganisms were identified as novel, e.g. *Leptobacillium muralicola* (Sun et al.,
230 2019). In the dark-gray stone of the Pasargadae World Heritage Site in Iran, lichens
231 were only detected on the areas of the stone where polyester and epoxy resin were
232 employed for conservation treatments (Shekofteh et al., 2018). In the former
233 Czechoslovakia, epoxy resins were frequently employed in sculpture, from the late
234 1950s for the creation of copies of existing statues, like the Cyril and Methodius
235 installed outside a church in Bratislava in 1989. In research by Pangallo et al. (2015)
236 on these figures, it was seen that the epoxy resins were heavily colonized by fungi,
237 algae and cyanobacteria. Suits from the Apollo lunar missions stored at the
238 Smithsonian Institution's National Air and Space Museum are composed of multiple
239 layers, many of which have begun to degrade. *Paecilomyces* and *Cladosporium* fungi
240 were isolated from two synthetic polymers and were capable of degrading the
241 synthetic polymers themselves, confirming that they were a deterioration factor to
242 consider in the conservation of the spacesuits (Breuker et al., 2003). Danko et al.
243 (2020) reported that in 48 samples from spacesuits subjected to shotgun metagenome
244 sequencing, a diverse microbial population included *Curtobacterium* and
245 *Methylobacterium*.

246 In addition to ageing, the susceptibility to microbial attack also depends on the way
247 the synthetic polymers have been applied (Borgia et al., 2003; Cappitelli et al., 2006;
248 Cappitelli and Sorlini 2008), the environment, and the amount of product applied
249 (Ferreira Pinto and Delgado Rodrigues, 2014). Four architectural acrylic paints were
250 exposed to outdoor conditions for seven years in both the urban site of São Paulo
251 and the coastal site of Ubatuba in Brazil (Shirakawa et al., 2011). Staining and
252 detachment of coatings were evaluated and the microbial community identified. The
253 painted surfaces were much more discolored in the marine environment, where the
254 principal taxa were the cyanobacteria *Scytonema* and *Gloeocapsa*. These results were
255 related to the higher rainfall in Ubatuba as well as the presence of trees in the marine
256 environment increasing the airborne microbiota. At the Maiji Grottoes on the ancient
257 “Silk Road”, China, where PVA emulsion has been applied to protect mural
258 paintings, the enclosed environment was seen as another factor affecting fungal
259 growth (Meng et al., 2020). Indeed, in two grottos that were always kept closed,
260 murals were heavily attacked by fungi. Interestingly, fungal growth was found on
261 the coating and not on the murals, indicating that the synthetic resin was the energy
262 source for microorganisms.

263 Microbial inhibition conservation strategies are often based on the employment of
264 water repellents to decrease water absorption, hence bioreceptivity, and on the
265 application of biocides (Moreau et al., 2008). One of the few in-situ studies
266 (archaeological stones in Fiesole, Italy), led by Pinna et al. (2018), regarding water-
267 repellents and consolidants (tetraethylorthosilicate, methylethoxy polysiloxane,
268 Paraloid B72 and tetraethylorthosilicate) showed that even when in presence of
269 biocides (tributyltin oxide + dibutyltin dilaurate) and copper nanoparticles, the
270 coatings had no crucial role in preventing microbial development.

271

272 **3. Biosusceptibility of composites based on synthetic polymers**

273 Polymer composite materials (or polymer composites) have aroused interest for the
274 conservation of cultural heritage and artistic objects. Polymer composites are
275 composed of different unique materials with at least one polymer, tailored for a
276 particular application. They consist of a minimum of two components, the matrix
277 and the reinforcement. The polymer composites show improved physical properties
278 as compared to the individual materials. In this paper, we will refer only to
279 composite materials with synthetic polymers as matrix.

280 Examples of synthetic polymer composites are fiber-reinforced polymer composites,
281 where carbon fibers are the reinforcements and acrylic polymers, epoxy, vinyl ester,
282 and polyesters are the matrix. These novel polymer composites show interesting
283 properties for the conservation of objects of art, e.g. remarkable physical strength,
284 desired hydrophobicity and optical features, large specific surface area for the
285 absorption of other nanomaterials and appreciable biocompatibility (David et al.,
286 2020).

287 However, these materials do not escape from being colonized by microorganisms.
288 Recently, Breister et al. (2020) studied the mechanisms underpinning the
289 biodeterioration of polymer composites used in construction and conservation. The
290 researchers combined material investigations with multi-omics analyses to assess
291 the role and mechanisms of biodeterioration of vinyl ester-based polymer
292 composites over time. Six metabolisms that potentially affected the polymer
293 composite structure were identified and associated to taxa belonging to
294 CPR/Patescibacteria, Deltaproteobacteria, Chlorobi, and Chloroflexi, which possess
295 pathways for breaking down the main component of the material such as acrylate,
296 esters, and bisphenol. The degradation of the binding matrix (acrylate, bisphenol A,
297 esters) through chain scission of chemical bonds and the physical damage induced
298 by gas production (H_2S , H_2) contributed to the biodeterioration of the material.

299 The results presented by van der Werf et al. (2015) showed that only the samples
300 treated with Estel1100 (a silicon-based product employed as consolidant/water-
301 repellent for many lithotypes) and not Estel1100/ZnO nanocomposites, allowed for
302 the growth of the fungus *A. niger*, a ubiquitous fungus on stone substrates, more or
303 less in the same amount as on the bare calcareous stones from the south of Italy.
304 In order to obtain new stone coatings, two oxides were dispersed in Paraloid B72
305 and the fluorinated polymer Akeogard CO (Ruffolo et al., 2010). When the two
306 polymer matrices alone were tested for their biocidal properties (5% concentration)
307 on *A. niger* in a few days they were easily attacked. In contrast, in the presence of
308 ZnTiO₃ a complete inhibition was observed after a month. *A. niger* was inoculated
309 on treated stone specimens by La Russa et al. (2012) and the microbial development
310 interfered with the structural fabric of the acrylic resin, decreasing its
311 hydrophobicity.

312 Traditional conservation treatments typically rely on the employment of water
313 repellents to reduce water absorption, and hence bioreceptivity, and on the
314 application of biocides to reduce the colonization process (Moreau et al., 2008). In
315 this respect, polymer composites made with nanoparticles (NPs) and synthetic
316 polymers opened up new opportunities for designing and improving conservation
317 materials of enhanced hydrophobic character (Reyes-Estebanez et al., 2018;
318 Aslanidou et al., 2018) and self-cleaning properties (Crupi et al., 2018; Kapridaki et
319 al., 2018; Ruffolo and La Russa, 2019). An example of polymer nanocomposites is the
320 highly hydrophobic Paraloid B72 coatings obtained by including silica nanoparticles
321 into the acrylic matrix (Ntelia et al., 2020). Polymer nanocomposites offer the
322 advantage of the NPs immobilization within the polymeric matrix, limiting the
323 release of NPs to humans and the environment (Baalousha et al., 2016). Colangiuli
324 et al. (2019) added titanium nanoparticles to a fluorinated polymer for assessing the
325 self-cleaning and photocatalytic features of such a coating over time in a real urban

326 environment. After natural ageing, the coated stone was still protected. However,
327 the photocatalytic efficiency progressively is reduced as result of the loss of the
328 photocatalyst from the coating surface due to a polymer modification by
329 weathering. For this reason, the use of organic binder should be replaced, or at least
330 combined with the inorganic ones.

331 Recently Aldosari and colleagues (2019) dispersed photocatalyst zinc oxide
332 nanoparticles in the acrylic Paraloid B44 to produce novel polymer composites with
333 effective biocidal, self-protection, and hydrophobic properties, to be used in the
334 protection of deteriorated marble heritage surfaces. Fungal inhibition and self-
335 protection properties were confirmed without any surface colour change.
336 Nanoparticles suspended in Primal AC33 and a silicon polymer applied onto stone
337 reduced by between 61% and 68% the presence of *Escherichia coli*, *Streptomyces*
338 *parvulus* and *B. subtilis* (Franco-Castillo et al., 2021). Helmi et al. (2015) recommended
339 using Ag nanoparticles for heritage surfaces instead of Cu nanoparticles, due to the
340 stronger colour variation of the latter, but also reported promising antimicrobial
341 activity of Cu nanoparticles, comparable with those of its Ag counterparts, against
342 strains of bacteria and fungi isolated from funeral masks from Saqqara necropolis
343 (Egypt). Similarly, Essa and Khallaf (2014) combined Ag nanoparticles with a silicon
344 polymer, highly reducing bacterial and fungal growth on the coated stones (*S.*
345 *parvulus* development by 98.6% and the growth of *A. niger* was almost nonexistent).
346 Copper has strong biocidal activity, which is observable when adding an element of
347 this material in a building or structure (see e.g., Sanmartín et al., 2021a). These same
348 authors, Essa and Khallaf (2016) through a novel bioprocess which was inexpensive
349 and eco-friendly, where the embedding of Cu nanoparticles into a polymer matrix
350 led to nanocopper composites with remarkable antimicrobial properties, improved
351 the physical features of the treated stones and also inhibited microbial development
352 on the historical surfaces.

353

354 **4. Biocleaning to remove synthetic polymers**

355 Besides coming from non-renewable sources, poor compatibility, and appearance
356 changes, synthetic polymers are not readily biodegradable, being difficult to remove
357 and often not re-treatable (Yang and Liu 2014; Mistretta et al., 2019). Traditional
358 conservation techniques for the removal of synthetic resins may lead to more
359 damage compared to uncoated surfaces (Ocak et al., 2015).

360 As relatively few studies have addressed the biodegradation and biocleaning of
361 synthetic polymers, few microorganisms with the potential to removal those target
362 materials have been identified (**Table 1**). Thus, focusing on the selection of suitable
363 microorganisms is an important area of study. Candidate microorganisms can be
364 found in international microbial collections (DSMZ, ATCC, CECT, etc.) or in natural
365 environments where synthetic polymers are present. Some Vicenza white limestone
366 and veined Carrara marble specimens treated with the consolidant EP2101
367 (cycloaliphatic epoxy resin) were exposed outdoors for one month and then kept for
368 three months in a box at 24–26°C with 95% relative humidity (Tesser et al., 2018).
369 The dominant fungi that grew on the treated and untreated specimens were
370 identified as *Alternaria alternata*, those developed only on treated stones were
371 *Cladosporium oxysporum* and *Chaetomium* spp. Interestingly, the researchers
372 proposed the use of the fungi identified on the treated specimens for biocleaning.

373 Fifty-four strains were retrieved from environments where recent and old graffiti
374 were present and their potential ability to attack Montana gold acrylic professional
375 spray paints (shock black, shock red, shock white, ultramarine and R-9011, Montana
376 Colors) was assessed (Sanmartín et al., 2015). Bacteria able to grow on acrylic paints
377 belonged to *Arthrobacter*, *Bacillus*, *Gordonia*, *Microbacterium*, *Pantoea* and *Pseudomonas*
378 and fungi to *Alternaria*. To the best of our knowledge, there is only one other study
379 involving the microbial ecology of graffiti paintwork (Bosch-Roig et al., 2021), where

380 from 31 putative bacteria from naturally aged graffiti, 13 were tested by screening,
381 and three were of interest as they used the graffiti paint as the sole carbon and
382 energy source. Among these, two were identified as the same taxon, the yeast
383 *Candida parapsilosis*, and the other one as the actinobacterium *Rhodococcus*
384 *erythropolis*. In the study by Cattò and Sanmartín et al. (2021), among the 8 bacteria
385 investigated, *Enterobacter aerogenes* ATCC 13048, *Comamonas* sp. ATCC 700440 and a
386 mixture of *Bacillus* sp., *Delftia lacustris*, *Sphingobacterium caeni*, and *Ochrobactrum*
387 *anthropi* ATCC 53922 had the highest potential for bioremoval of a black non-
388 metallic spray paint (R-9011, Montana Colors) and a silver metallic spray paint
389 (Silver Chrome, Montana Colors), characterized as alkyd and polyester-based resin,
390 and polyethylene-based resin, respectively.

391 In the study by Germinario et al. (2017) the removal activity of lipases for acrylic
392 marker pen inks was evaluated. Better performances were shown with the non-aged
393 specimens, suggesting the urgency to perform biocleaning before ageing.

394 To the best of our knowledge, no bacteria capable of attacking Paraloid B-72 or
395 Paraloid B-44 (by using it as their sole carbon and energy source) have yet been
396 found (Nugari and Priori, 1985; Li, 2006; Troiano et al., 2014). Only the successful
397 use of a *Candida cylindracea* lipase to remove aged Paraloid B-72 from a couple of
398 paintings is reported (Bellucci et al., 1999), but *Candida* strains are not considered
399 safe microorganisms for operators.

400

401 **5. Final Remarks**

402 One of the most important aspects in the use of synthetic polymers in conservation
403 applications would be the definition of a protocol to test the biodeterioration and
404 the biodegradation (in case of biocleaning procedure) of the materials. So far, most
405 of the studies have been based on testing the impacts and/or performance of single
406 microorganisms. However, these researches do not fully describe the impacts of a

407 complex microbial community on polymeric materials, thus the lack of information
408 on the polymers' biocleaning mechanisms and degradation pathways is not
409 surprising. Moreover, many of the employed microorganisms in prior studies are
410 scarce in molecular surveys carried out in the field and are thus unlikely to be the
411 key drivers of deterioration of synthetic polymers. Therefore, despite the decades of
412 use of synthetic polymers in conservation, little is still known about the diversity
413 and impacts of microorganisms dwelling on synthetic polymers in their heritage
414 context. Test methods should be created based on the practical development of a
415 standard procedure for the qualitative and quantitative evaluation of the durability
416 and removability of the synthetic polymer, to understand under which condition its
417 application can be considered safe for the heritage surface.

418 The conservation of cultural heritage demands complex interdisciplinary research
419 for developing suitable materials for the protection of heritage surfaces. In the
420 context of synthetic polymers, compatibility with the original material, durability,
421 resistance to adverse environmental conditions, transparency and
422 reversibility/retractability of the treatment are mandatory prerequisites that still
423 represent challenges in conservation practices. For traditional synthetic polymers,
424 such as acrylic and methacrylic polymers, durability and removability is affected
425 mainly by photo-oxidation phenomena (Favaro et al., 2006; Artesani et al., 2020).
426 Thus, the future use of synthetic resins should be seen in a wider and more complex
427 scenario than only the microbiological point of view to assess if, overall, their
428 benefits outweigh the risks and the damage that these materials can pose.

429 As a consequence of all the above shortcomings of synthetic resins and composites,
430 researchers are now reconsidering the use of natural polymers rather than synthetic
431 polymers to protect heritage from damage and weathering (Ocak et al., 2015;
432 Mistretta et al., 2019). The green chemical approach is driving attention towards
433 biopolymers, especially those from renewable resources. One example is polylactic

434 acid (PLA), which can be produced from lactic acid-derived biodegradable and
435 renewable material (Ocak et al., 2015). Although novel biopolymers are among the
436 most interesting materials for cultural heritage conservation nowadays, their
437 chemical properties and stability over time need to be thoroughly examined. Most
438 of these biopolymer-based solutions are represent only by a single trial, undertaken
439 at the very beginning of the study, but they are nonetheless considered relevant for
440 future investigation.

441

442

443 **Declaration of interests**

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

447

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- 702

703 **Figure 1.** Image under stereomicroscope showing the cracks in the coating made of
 704 nano-sized silica consolidant containing TiO₂ that served as an ecological habitat for
 705 phototrophic organisms. The image was taken from a study conducted by Sanmartín
 706 et al. (2021b) and is reproduced by courtesy of the authors.

707

708 **Figure 2.** A view (X40 magnification) of sample 14F035 taken from the Milan
 709 cathedral, Italy, in an area where poly-isobutylmethacrylate has been used to protect
 710 the marble. Meristematic and sometimes yeast-like growth with budding cells was
 711 abundant on this sample (see Cappitelli et al., 2007).

712

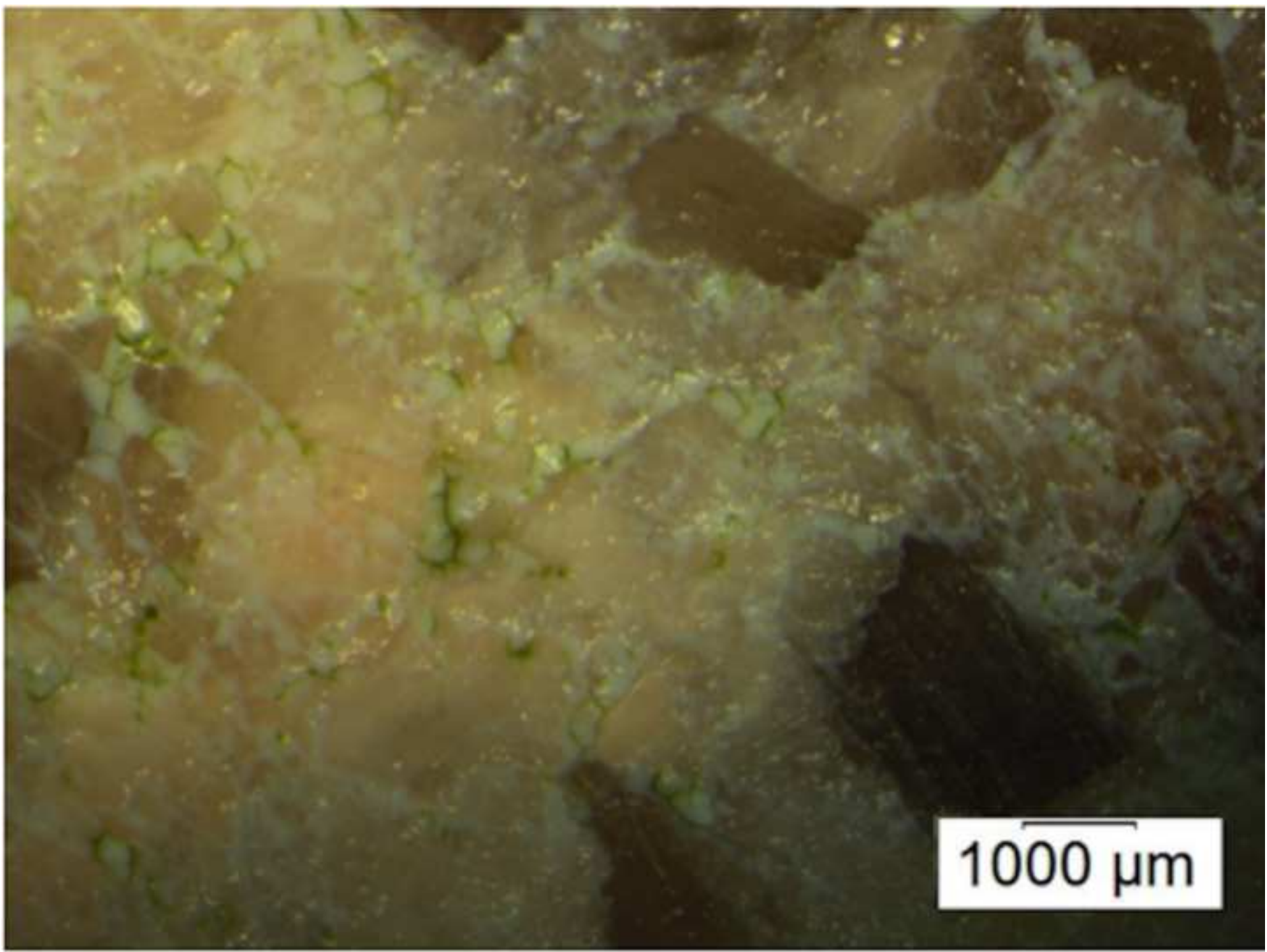
713

714

715 **Table 1.** List of articles ordered by year of publication, where microorganisms that
 716 can be used for synthetic material removal are indicated.

Microorganism	Classification	Role/application	Reference
<i>Candida cylindracea</i>	Yeast	Removal of Paraloid B72 from paintings through a lipase enzyme	Bellucci et al. (1999)
<i>Pseudomonas aeruginosa</i> (PA01), <i>Pseudomonas stutzeri</i> (ATCC 23856), <i>Pseudomonas putida</i> (DeFENS collection, isolated from wastewater treatment plant), <i>Escherichia coli</i> (ATCC 25404), <i>Bacillus licheniformis</i> (DeFENS	Bacteria	Four-year dried Paraloid B72, originally solubilized in 15% ethyl-acetate	Troiano et al. (2014)

collection, isolated from a biodeteriorated acrylic painting on canvas by a contemporary artist)			
<i>Arthrobacter oryzae</i> , <i>A. aurescens</i> , <i>Bacillus megaterium</i> , <i>B. aquimaris</i> , <i>Gordonia alkanivorans</i> , <i>Microbacterium oleivorans</i> , <i>Pantoea</i> sp. and <i>Pseudomonas mendocina</i> and <i>Alternaria alternata</i> .	Bacteria and fungi	Acrylic paints	Sanmartín et al. (2015)
<i>Cladosporium oxysporum</i> and <i>Chaetomium</i> spp..	Fungi	Cycloaliphatic epoxy resin	Tesser et al., 2018
<i>Enterobacter aerogenes</i> ATCC 13048, <i>Comamonas</i> sp. ATCC 700440 and a mixture of <i>Bacillus</i> sp., <i>Delftia</i> <i>lacustris</i> , <i>Sphingobacterium caeni</i> , and <i>Ochrobactrum anthropi</i> ATCC 53922	Bacteria	Alkyd and polyester-based resin and polyethylene-based resin	Cattò and Sanmartín et al. (2021)
<i>Candida parapsilosis</i> and <i>Rhodococcus</i> <i>erythropolis</i>	Bacteria	Alkyd and polyester-based resin	Bosch-Roig et al. (2021)



14F035_1_40X_f particolare.png



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: