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Antibacterial Properties of AquaSun Sol–Gel Coating

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The antimicrobial activity, surface features, wettability, and adhesion of photocatalytic xerogel AquaSun deposited on shipbuilding steel in comparison to those of a commercial antifouling paint are investigated. The antimicrobial activity of AquaSun deposited on a steel substrate pretreated with primer and tiecoat paint exposed to a microbial consortium composed of *Bacillus aquimaris* and *Pseudomonas poae* is significant and of broad scope. Taking into account the low cost, high stability, and lack of ecotoxicity of this hybrid sol–gel coating, these results are promising en route to developing novel antibacterial coatings of broad applicability.

1. Introduction

Consisting of organically modified silica (ORMOSIL) doped with flower-like microparticles of the visible light photocatalyst bismuth tungstate, AquaSun is a xerogel coating showing promising antifouling (AF) activity both in laboratory[1] and in real-life comparative tests carried out in highly polluted seaport waters.[2]

Developed with the aim to merge the solar-driven photocatalytic generation of powerful oxidizing species such as electron holes (h^+) and reactive oxygen species (ROS) including H_2O_2, hydroxyl (OH·) and oxygen ($\cdot O_2^-$) radicals,[3] and the foul release properties of biocide-free ORMOSIL xerogels,[4] AquaSun consists of a C18 1%/C8 49%/TEOS 50% silane (wherein C18 stands for *n*-octadecyltrimethoxysilane, C8 for *n*-octyltriethoxysilane, and TEOS for tetraethylorthosilicate) solution in 2-propanol containing suspended Bi_2WO_6 microparticles prehydrolyzed under acidic conditions.[5] The waterborne coating is readily deposited on the surface of shipbuilding steel by simple brushing, followed by curing at room temperature, eventually affording a ≈200 μm-thick film.[5] In light of forthcoming practical applications, it is also remarkable that the AquaSun glassy coating is a less rigid material when compared to state-of-the-art commercial AF topcoat, but highly adherent to the shipbuilding steel substrate, thanks to the formation of a strongly cohesive thin film.[6]

Biofouling is a complex phenomenon starting with bacteria colonizing the immersed surface to growth of a biofilm due to bacterial cell multiplication and extracellular polymeric substance synthesis using soluble organic substances in the aqueous environment.[7] In general, due to their ease of production, low cost, lack of toxicity, and high mechanical and chemical stability, ORMOSIL coatings have a large application potential in preventing biofouling even in outdoor environments.[4,8] One noticeable example is buildings whose construction porous ceramic materials are susceptible to water accumulation, microbial colonization and subsequent related structural and biological damages including formation of toxic mold and mycotoxins.[9] The sol–gel entrapment of Bi_2WO_6 in transparent ORMOSIL glasses not only ensures the photocatalytic generation of highly reactive holes (h^+) and radical species (OH· and $\cdot O_2^-$)[3] that promote cell membrane destruction, leading to cell lysis and death,[10] but also promotes effective electron–hole effective separation at the surface of the semiconductor (delayed recombination), eventually improving the photocatalytic activity.[11] Now, to investigate the antibacterial properties of AquaSun, we conducted a number of comparative biological in vitro tests using a microbial consortium composed of the Gram-negative strain *Pseudomonas poae* and the Gram-positive strain *Bacillus aquimaris*. The coating was applied to shipbuilding steel and comparison was made with a state-of-the-art commercial AF paint consisting of a silyl acrylate "self-polishing" topcoat (SeaQuantum Ultra S)

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DOI: 10.1002/adem.202300626

containing high amounts of cuprous oxide (785 g L^{-1}) and copper pyrithione (66 g L^{-1})[12] purchased from Jotun (Sandefjord, Norway). Furthermore, we assessed the surface features, wettability, and adhesion of the coatings after several months of immersion in seawater, which are crucially important parameters to assess the real performance of marine coatings from the practical application standpoint.[13]

2. Results and Discussion

To evaluate the antimicrobial activities of the coatings, we analyzed the ability of a Gram-positive strain (*Bacillus aquimaris* DSM 16205^T) and a Gram-negative strain (*Pseudomonas poae* DSM-14936^T) to grow and form colonies as indicators for bacterial viability.

2.1. Colony Assay

The colony-forming unit (CFU) count was used to assess the concentration of living cells upon contact with differently coated substrates. This implies that, during the incubation in the liquid medium, the cells may have had time to recover from the oxidative stress induced by native H_2O_2, which may lead to underestimating the real inhibitory effect of the tested material. The surviving bacterial cells after the exposure to experimental coatings under an aquarium LED light were quantified and outcomes are plotted in **Figure 1**.

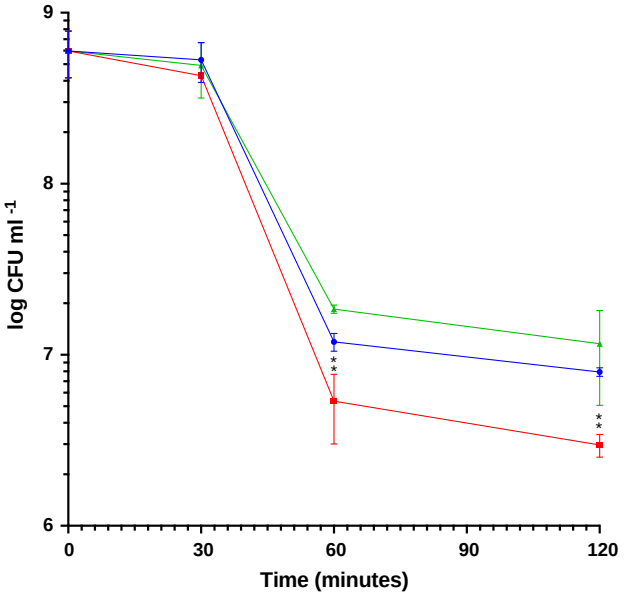


Figure 1. Surviving bacterial colony numbers in microbial consortium composed of *Pseudomonas poae* (DSM 14 936) and *Bacillus aquimaris* (DSM 16 205) on different substrates: shipbuilding steel coated with commercial primer + commercial tie-coat, both purchased from Jotun (Sandefjord, Norway): P + T (blue line); shipbuilding steel coated with commercial primer Jotun + commercial tie-coat + AquaSun sol–gel coating: P + T + THA8 (red line); shipbuilding steel coated with commercial primer + commercial tie-coat + AquaSun blank sol–gel coating: P + T + THA8B (green line), (*$P \leq 0.05$; **$P < 0.005$; ***$P < 0.0005$).

In detail, at 30 min incubation time, the CFU count did not show significant differences among all the substrates tested ($P > 0.05$). After 60 min, the bacterial counts on P + T + THA8 in comparison to control intermediate tie-coat paint (P + T in the graph) showed favorable antimicrobial activities, with a log reduction of 0.34. The log reduction remained constant (0.39) until 120 min. At the end of experiments (120 min), the number of surviving bacteria on P + T + THA8 was significantly lower than those on intermediate tie-coat paint and on the blank coating (P + T + THA8B). No CFU were counted after the exposition on the commercial coating used as positive control (P + T + COMM) at the dilution tested.

Under natural light irradiation, the quantification of the surviving bacterial cells on experimental coatings did not reveal any statistically significant differences with indoor experiments after 30 and 60 min time exposition (data not shown). Unfortunately, in the outdoor trials, at 120 min, the bacterial suspension on the four substrates was nearly entirely evaporated, making not possible the direct comparison of indoor and outdoor experiments. We did not measure the performance of the AquaSun sol–gel coating without irradiation, because it is known since the early days on the AquaSun coating performance that irradiation with visible light is key to ensure lack of microbial colonization and subsequent biofouling.[1,3]

2.2. Bacterial Cell Viability Assay

Adenosine triphosphate (ATP)-dependent enzyme that gives rise to luminescence was used to determine bacterial ATP levels. The assay (the BacTiter-Glo reagent is added causing cell lysis and the production of a luminescent signal measured with a luminometer) determines the number of viable microbial cells by monitoring the presence of ATP, an indicator of the presence of metabolically active cells.[14] In order to obtain insight on the cell viability/vitality, and thus to evaluate the bacteriostatic/bactericidal effect of hydrogen peroxide[15,16] generated in situ by the sol–gel-entrapped photocatalyst under visible light irradiation, a preliminary test of bacterial cell viability was conducted by exposing the microbial consortium to increasing concentration of H_2O_2. The overall time of exposure of the cells to aqueous H_2O_2 was 3 h.

Figure 2 shows the luminescence values measured after 40 min in vitro exposure of microbial consortium to increasing concentration of aqueous H_2O_2, from 3.7×10^{-4} to 1.8 per cent). Results show a significant inhibition of cellular activity till 0.037% H_2O_2 concentration only. Lower concentrations of H_2O_2 did not affect the emitted luminescence, which was comparable to that of the control. Remarkably, after 40 min the measured luminescence values remained constant.

The microbial consortium viabilities using the ATP assay were calculated in indoor and outdoor experiments. As control, luminescence counts of samples exposed to P + T under identical conditions were set as 100% bacterial viability registered at the same timelapse. Data normalization was due to the significant differences of cells concentration on P + T substrate for experiments duration (**Figure 3**B).

In the indoor experiments (Figure 3A), after 30 min contact time with P + T + COMM, the vitality of microbial consortium was almost completely compromised, while exposure to

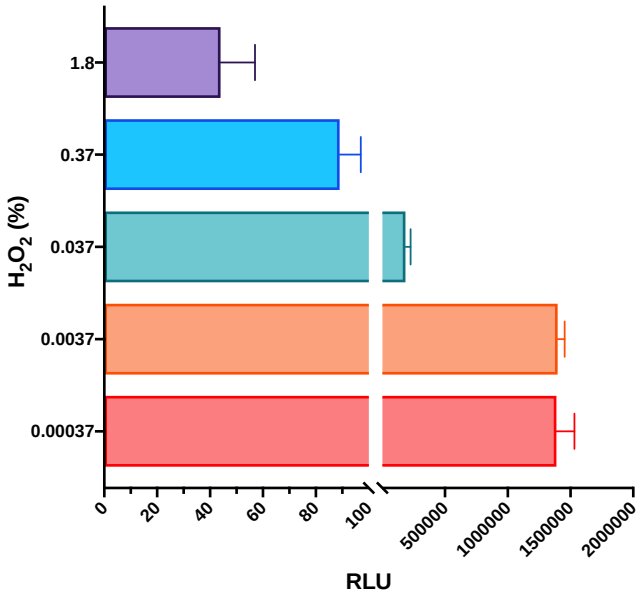


Figure 2. Bacterial cell viability in microbial consortium composed of *P. poae* (DSM 14 936) and *B. aquimaris* (DSM 16 205), measured by the presence of ATP luminescence, after 40 min exposure to increasing concentrations of aqueous H_2O_2.

P + T + THA8 and P + T + THA8B coatings reduced bacterial ATP levels respectively by 15% and by less than 5%.

After 60 min exposure to P + T + THA8 and P + T + THA8B, the ATP levels decreased by ≈30% and by less than 20% respectively. Finally, after 120 min contact time with P + T + THA8, the ATP levels were reduced by ≈40%, while P + T + THA8B was less active inducing a reduction of ATP levels of ≈25% only.

During the outdoor experiments (**Figure 4**A), the maximum inhibitory effect of P + T + THA8 was observed after 60 min exposure of the cells, when the ATP levels were reduced by 30%. Instead, on the substrates coated with blank AquaSun (P + T + THAB) and commercial AF coating (P + T + COMM), a stronger decrease of bacterial viability was detected just at 30 min, when ATP levels were reduced by ≈35% and ≈55% respectively. After 60 min, the percent killing, normalized against control, returned for both substrates to a value comparable with that given by P + T + THA8, with ≈70% of bacterial

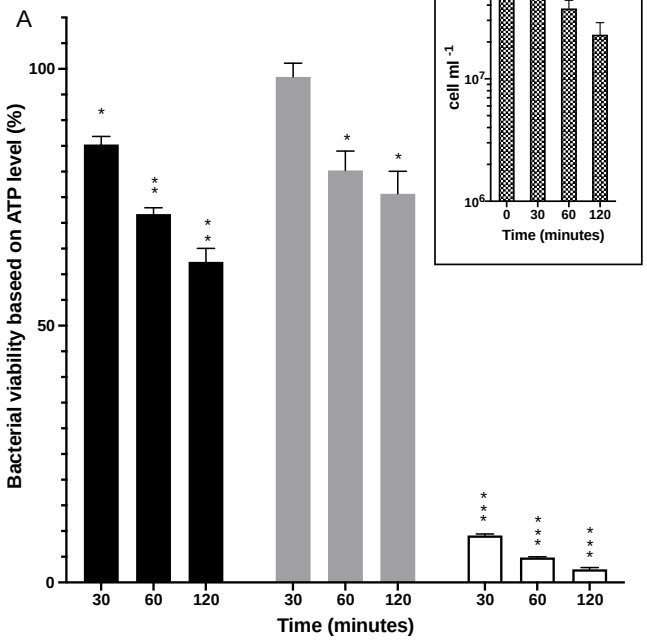


Figure 3. A) Microbial consortium (*P. poae* and *B. aquimaris*) viability, measured by the presence of ATP, during exposure to P + T + THA8 (black histograms), P + T + THA8B (gray histograms), and P + T + COMM (white histograms) under artificial light (*$P \leq 0.05$; **$P < 0.005$; ***$P < 0.0005$). B) Cell concentration of microbial consortium exposed to P + T under identical conditions.

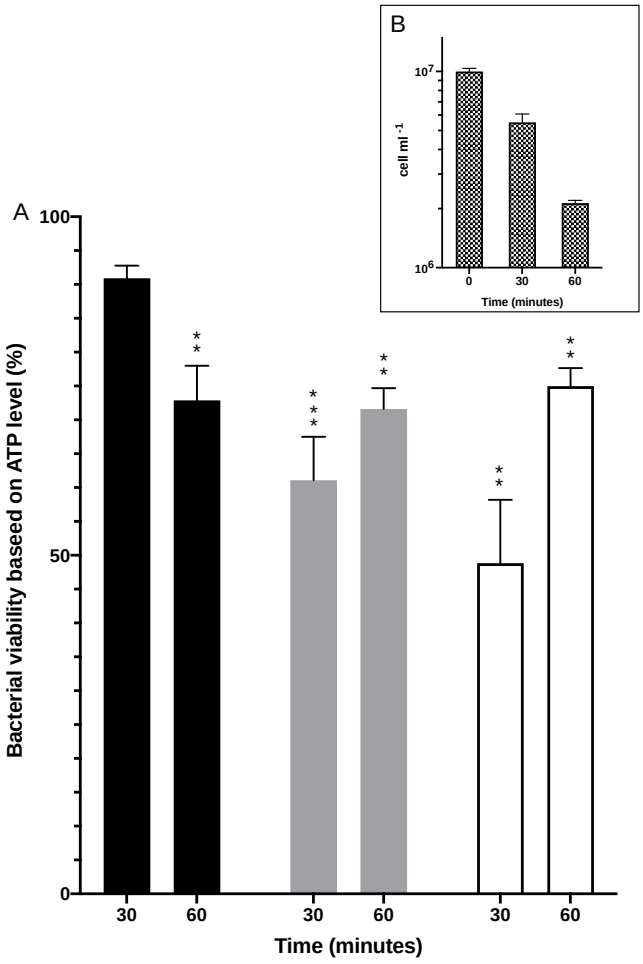
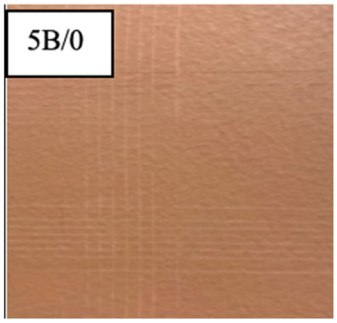
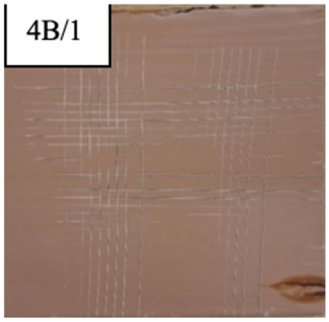
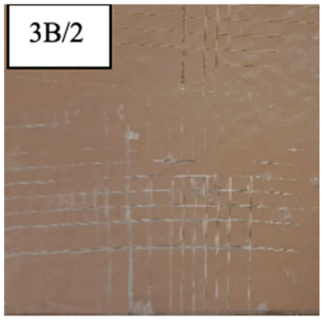
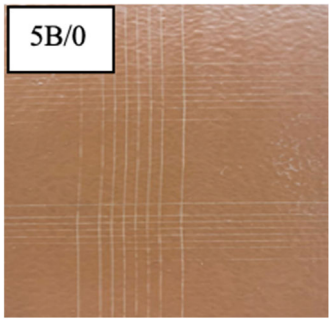
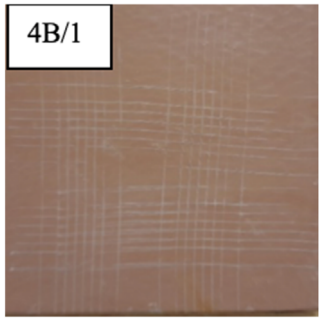
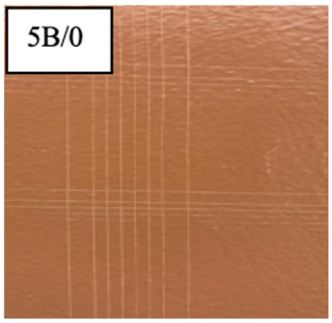
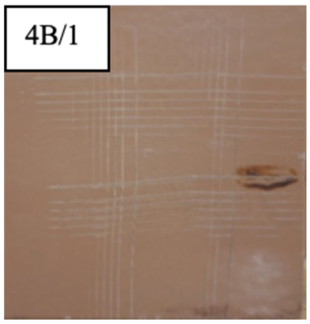
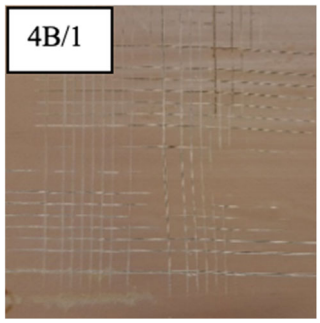


Figure 4. A) Microbial consortium (*P. poae* and *B. aquimaris*) viability, measured by the presence of ATP, during exposure to P + T + THA8 (black histograms), P + T + THA8B (gray histograms), and P + T + COMM (white histograms) under solar irradiation (*$P \leq 0.05$; **$P < 0.005$; ***$P < 0.0005$). B) Cell concentration of microbial consortium exposed to P + T under identical conditions.

Table 1. Crosscut test result of the P + T + COMM, P + T + THA8, P + T + THA8B coatings after 0, 1, and 4 months of immersion in seawater.

Immersion time	0 M	1 M	4 M
Sample's code			
P + T + COMM			
P + T + THA8			
P + T + THA8B			

viability. Compared to indoor trials, outdoor experiments presented several technical complications due to fluctuating temperature, light, and wind conditions that can partially explain these differences.

2.3. Coating Adhesion and Wettability

The results of crosscut test summarized in **Table 1** indicate that the adhesion power of the P + T + COMM, P + T + THA8, P + T + THA8B coatings progressively decreases during the immersion time.

After 1 month immersion time, the adhesion of all coatings decreased from 5B/0 to 4B/1, whereas after 4 months, the degree of adhesion of the coatings decreased to 3B/2 for both the commercial and the AquaSun coatings. Only for the undoped AquaSun coating THA8B, the degree of adhesion decreases to just 4B/1, as a result of the strong adhesion power of dopant-free ORMOSIL coatings.[17]

3. Conclusion

In summary, the results of the first investigation of the antimicrobial properties of the photocatalytic AquaSun xerogel coating show evidence that both under artificial and natural (sunlight) solar irradiation, AquaSun exhibits powerful and broad-scope antibacterial activity, similar to what is observed for free bismuth tungstate coatings under visible light irradiation.[18] Observed bacterial mortality is more evident under monitored conditions (indoor experiments) rather than in outdoor experiments where the fluctuation of temperature, light, and wind frequently occurred. The outcomes of the ATP assay first reported in this study using a bacterial consortium of Gram-positive and Gram-negative bacteria clearly indicate that the AquaSun coating exerts significant antimicrobial activity of broad scope. Other important features for practically useful protective coatings are their mechanical and physical properties.[13] AquaSun exhibited good adhesion properties compared to

the commercial AF paint at long immersion time, retaining the hydrophobic character (and thus the low surface energy). Finally, likewise other waterborne photocatalytic sol–gel coatings,[19] the AquaSun paint can be easily applied on surfaces of widely different compositions (construction materials, metal, wood, glass, and polymer) via professional cost-effective spraying.

Antimicrobial coatings devoid of human and animal toxicity are in great demand. Especially, photocatalytic coatings such as bismuth oxides and bismuth tungstate are promising due to the fact that the $\cdot O_2^-$, OH·, and h^+ generated by these coatings under visible light are able to degrade antibiotic-resistant pathogens and antibiotic-resistant genes.[20] Being easily made at low cost, highly stable, and devoid of ecotoxicity,[3] in conclusion, AquaSun is a truly sustainable functional coating[21] whose applications may soon extend beyond protection from biofouling of immersed surfaces. The results reported in this study suggest that this new coating might impart antimicrobial properties to widely different surfaces with numerous potential health, environmental, and economic benefits. The outcomes of new investigations will be reported in the near future.

4. Experimental Section

Coating Deposition: The preparation and application of the AquaSun coating, taking place via a three-step process, has been described in detail elsewhere.[5] The commercial topcoat was applied to shipbuilding steel donated by Fincantieri SpA (Trieste, Italy) by a three-step coating process consisting of sanding the steel substrate followed by spraying the 1) primer; 2) tie-coat; and 3) AF topcoat followed by curing at room temperature.

Bacterial Strains, Culture Conditions, and Microbial Consortium Preparation: *Pseudomonas poae* (DSM 14 936)[22] and *Bacillus aquimaris* (DSM 16 205)[23] were used in this study. The bacterial cultures were grown overnight in Bacto marine broth 2216 at 24 °C. At the point when the cell concentration reached values of 10^8 cells mL^{-1}, corresponding to midlog phase of growth (measured by DAPI counterstain), the biomass was pelleted (9000xg, 10 min) and washed twice with 1x phosphate-buffered saline (PBS; 140 mM NaCl, 2.7 mM KCl, 4.3 mM $Na_2HPO_4 \times 7H_2O$, and 1.5 mM KH_2PO_4). The cells were then resuspended in 1x PBS and mixed to a final concentration of 10^8 cell mL^{-1} for each strain.

Direct Cell Counting via DAPI Staining: The bacterial abundance was measured by direct counting using a epifluorescence microscope following the methodology reported by Porter and Feig.[24] In detail, subsamples were collected and fixed with formaldehyde (2% final concentration). Cell counts were performed by cell staining using 4′,6-diamidino-2-phenylindole (DAPI, Sigma-Aldrich, Milan, Italy).

Samples were prepared using a mixture consisting of glycerol-PBS AF1 (Molecular Probes, Eugene, OR, USA), Vectashield mounting medium (Vector Laboratories, Burlingame, CA, USA), and DAPI (final concentration 1 mg mL^{-1}). The slides were examined by epifluorescence with Axioplan 2 Imaging microscope (Carl Zeiss, Thornwood, NY, USA). All results were expressed as the number of cells mL^{-1}.

Experimental Design: Photocatalytic antimicrobial effect of THA8, AquaSun xerogel incorporating 12 wt% of Bi_2WO_6,[2] against microbial consortium, was evaluated exposing 1 mL of culture in triplicate on the surface of steel plate (7 × 15 cm) pretreated with primer and tie coat paint and further coated with a thin layer of sol–gel coating deposited by simple brushing with a paintbrush. The antimicrobial activity of four different substrates was assessed: shipbuilding steel coated with commercial primer + commercial tie coat, both purchased from Jotun (Sandefjord, Norway): P + T; shipbuilding steel coated with commercial primer Jotun + commercial tie coat + AquaSun sol–gel coating: P + T + THA8; shipbuilding steel coated with commercial primer + commercial tie coat + AquaSun blank sol–gel coating: P + T + THA8B; and shipbuilding steel coated with commercial primer + commercial tie coat + commercial AF paint (top coat): P + T + COMM. The experiments were conducted under the Aquarium light-emitting diode (LED) DSunY PopBloom 60 W system (purchased from Shenzhen DSunY Electronic Science and Technology, China) suspended 30 cm above the steel plates in controlled room temperature (24 ± 2 °C). The LED lighting system was used at 100% of its maximum power (60 W). The system consisted of 72 LEDs: 7 LEDs 20,000 K (18 W), 28 LEDs 420–450 nm, d 7 LEDs 520 nm (3 W), 14 LEDs 470 nm (36 W), 12 LEDs 4500 K, and 4 LEDs 700 nm (3 W). The distribution of the LEDs inside the lamp was homogeneous and did not show significant variations in wavelength and intensity amid the four steel plates (**Figure 5**a). The light intensity, measured at sample levels as photosynthetically active radiation (PAR), was 1759 ± 250 μE m^{-2} s^{-1} and was comparable to a Summer sunny day.[25]

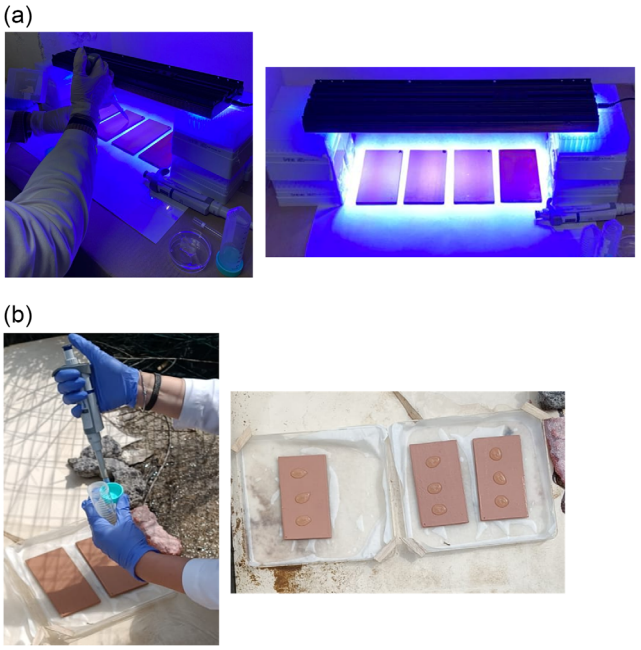


Figure 5. a) Indoor and b) outdoor microbiology experiments with shipbuilding steel substrates coated with different coatings.

Three different measures were carried out among the four plates to assess the uniformity of light flux on each plate tested (data not shown). Aliquots of cultures (100 μL) for colony and bacterial cell viability assays were collected at 0, 30, 60, and 120 min. Moreover, the photocatalytic antimicrobial effect of THA8 was evaluated by exposing the microbial consortium to natural light irradiation. All trials were conducted outdoors of Messina's laboratories of the Institute of Polar Sciences of Italy's Research Council in July 2022 in the hour time interval between 11:00 and 14:00 of maximum daily solar irradiation (Figure 5b). To mitigate the temperature rise effect, the four different metallic substrates were placed in four squared sterile Petri dishes filled with moist tissue paper and triple distilled water. The dishes in their turn were placed in a bath under constant flow of marine water at room temperature (24 ± 2 °C), thereby ensuring an average constant temperature between 22 and 28 °C, well within the limits of tolerability of the bacterial strains used for the tests. Aliquots of cultures (100 μL) for colony and bacterial cell viability assays were collected at 0, 30, 60, and 120 min.

Colony Assay: Aliquots of 100 μL of culture, collected directly from the surface of steel plates at established time and 10-fold serially diluted, were spread on sterile Bacto marine agar and incubated at 25 °C for 48 h. Only plates showing CFU between 30 and 300 colonies were counted. For consistency, we performed each colony assay test three times.

Bacterial Cell Viability Assay: The bacterial cell viability was determined using the BacTiter-Glo microbial cell viability assay kit (Promega, Madison, WI, USA) at various treatment times. Briefly, BacTiter-Glo reagent was prepared by combining lyophilized BacTiter-Glo enzyme/substrate mixture with the buffer at room temperature. 100 µL sample of bacterial culture was transferred to an opaque-walled 96-well plate and combined with 100 µL of BacTiter-Glo reagent. After mixing the contents in the dark for 5 min, the luminescence intensity was measured using a Promega GloMax luminometer. Percent killing was calculated from Equation (1)

$$\%\,\text{Reduction} = 1 - (\text{luminescence of experimental top-coat} / \text{luminescence of commercial tie-coat}) \times 100 \quad (1)$$

The resultant graphs showed mean ($n = 3$) and standard deviation (SD) for each data point, prepared using Prism 7 biostatistcal software (GraphPad Software, San Diego, CA, USA).

Statistical Analysis: The statistical analyses were conducted using the PAST software (version 4.03) for data analysis.[26] The Shapiro–Wilk test[27] was used for assessing normality of datasets, whereas we used the one-way ANOVA analysis of variance to determine the significance of the treatments.[28] Relative importance of each treatment group was investigated by pairwise multiple comparisons procedures using Tukey's honestly significant difference (HSD) test in case of normally distributed data and Dunn's posthoc in case of not normally distributed data. Differences between treatments were considered significant at $P < 0.05$.

Coating Resistance in Seawater: The adhesion resistance of all coatings was tested in seawater according to the ASTM D870-97 standard. DH36-Shipbuilding steel plate (150 × 75 mm, thickness 5 mm), commonly used in offshore and marine construction, was coated with P + T + THA8, P + T + THA8B, and P + T + COMM, as previously described. The samples were placed in aquarium tank and immersed in seawater to three-fourths of their height (**Figure 6**).

To simulate the movement of seawater, a submerged aquarium pump equipped with a filter was used to maintain a constant recirculation of water on the surfaces. The water temperature was kept at 24 °C, whereas the average outside temperature was 23 °C. The seawater was changed every two weeks. The samples were then analyzed for possible bubble formation, softening, and loss of adhesion, using the crosshatch test (Cross Hatch Adhesion Tester by Sama Tools, SADT502-5) before and after immersion in seawater after 1 month and after 4 months. If the edges of the cuts are completely flat, and none of the grid squares are detached, the adhesion degree is in accordance with the maximum adhesion value, that is, 5B of the reference standard ASTM D 3359-09e2 or value 0 for the ISO 2409:2007 standard (Figure 3A). If small flakes of paint detach at the intersection of the cuts, the value 4B of the reference standard ASTM D 3359-09e2 or the value 1 for ISO 2409:2007 (Figure 3B) is assigned. Finally, if the coating is partially or totally detached along the edges of the cuts, the 3B ASTM value or ISO2 value is assigned to the coating (**Figure 7**c).

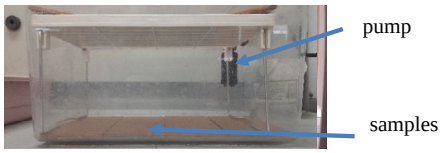


Figure 6. Shipbuilding steel probes coated with different coatings into an aquarium tank with a recirculating pump.

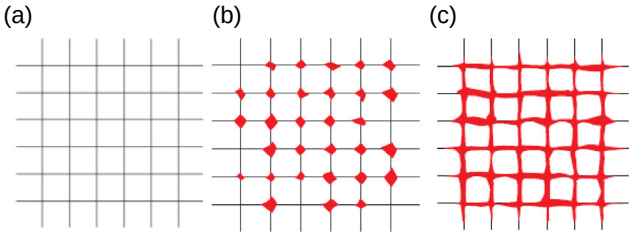


Figure 7. Coating detachment types according to ASTM/ISO standard: a) 5B/0, b) 4B/1, and c) 3B/2.

Surface Features of the Coatings: Obtained using a Mitutoyo Surftest SJ-210, series n. 178 portable surface roughness instrument (Kawasaki, Japan), the surface roughness (R_a) was calculated as the arithmetic mean of the absolute values of the evaluation profile deviations (Y_i) with respect to the mean line (Equation (2)).

$$R_a = \frac{1}{N}\sum_{i=1}^{n} |Yi| \quad (2)$$

The contact angles θ of Wenzel (θ_w, dependent on the roughness) and of Young (θ_Y, not dependent on the roughness) were measured by means of a prototype instrument (Department of Engineering, University of Messina), which measured the contact angle of 1 µL drop of deionized water of on the horizontal surface of the film, according to Equation (3) and (4)

$$\theta_w = 2arctg\left(\frac{2h}{d}\right) \quad (3)$$

$$\theta_Y = arcos\left(\frac{\cos\theta_w}{r}\right) \quad (4)$$

wherein d is the drop diameter, h its (mm), and r is the surface roughness.

Acknowledgements

This study is dedicated to Professor Clément Sanchez, CNRS and Collège de France, Paris, for all he has done to advance the science and technology of sol–gel materials. The authors thank NAVTEC (Consorzio di Ricerca per l'Innovazione tecnologica, Sicilia, Trasporti navali, commerciali e da diporto s.c.a.r.l) for research funding, grant number ARS01_00293 PON2014-2020 of research project "Thalassa" (TecHnology And materials for safe low consumption and low lifecycle cost veSSels And crafts). Thanks to Giuseppe Napoli and Nunzio Gallì, Istituto per lo Studio dei Materiali Nanostrutturati, CNR, for administrative and technical assistance, and to Franco Decembrini, Istituto di Scienze Polari, CNR, for technical assistance. Work of the researchers of at the Institute of Polar Sciences of Italy's Research Council has been supported by the project 'Emergence of virulence and antibiotic-resistance vectors in coastal and deep sea marine environments and analysis of the mechanisms and conditions underlying their spread and evolution', Progetti di Ricerca di Rilevante Interesse Nazionale (PRIN), Bando 2017 - grant 201728ZA49_001, of Italy's Ministry of Research and Education.

Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

antibacterial, antifouling, AquaSun, hybrid materials, sol–gels

Received: May 2, 2023
Revised: July 26, 2023
Published online: September 8, 2023

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