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Active coating including microorganism for indoor formaldehyde degradation

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Abstract. Indoor environmental contamination of residential units and workplaces is a major problem that needs efficient, durable and environmentally friendly solutions. The development of active coatings capable of entrapping and degrading selectively pollutants such as formaldehyde is herein proposed. Freeze-dried *Pseudomonas putida* sp. cells were introduced in sol-gel coatings. Precursor ratio, thickness and water content of the coatings were the parameters used to optimize their effectiveness on formaldehyde degradation. Although with different abilities, bacterial cells immobilized into the distinct sol-gel formulations were able to degrade formaldehyde. The presence of 3-glycidoxypolytrimethoxysilane and polyethylene glycol in the sol-gel promoted the enzymatic degradation of formaldehyde as they provide the stability of the humidity of the coatings and so, the well-being of the cells. Storage temperature and time proved to be important variables for keeping the enzymatic activity of degrading-microorganisms incorporated in the sol-gel coatings. Coating formulations promoting the higher abilities to degrade formaldehyde provided also good mechanical properties.

1. Introduction

Indoor air quality of residential units and workplaces has attracted attention in recent years due to the frequent association of indoor air pollution with various health problems. Volatile organic compounds (VOCs), in particular formaldehyde, are widely used in construction materials, wood processing, furniture, textiles, carpeting commonly found in indoor spaces [1]. The adverse effects in health caused by this toxic pollutant have promoted the development of distinct physical, chemical and biological strategies for removing it from the indoor air [2]. Among the existing commercial solutions, the enzymatic degradation of formaldehyde is being consistently explored as a flexible, low-cost and efficient strategy, respecting health and environmental procedure. The development of active coatings capable of entrapping and degrading selectively pollutants is of great interest. In this goal, the encapsulation of a formaldehyde-degrading microorganism into sol-gel coatings has been studied. Many microorganisms that degrade formaldehyde, namely bacteria, yeasts, fungi and marine algae, have been isolated and characterized [3]. Anabolic or catabolic pathways contained in the microorganism can be used for metabolizing the pollutant to other less toxic compounds or cell components [4]. *Pseudomonas putida* is one of the microorganisms commonly used for the natural biodegradation of formaldehyde from waste-water or soil [5]. The use of microorganism whole cell, compared to extracted/purified enzymes, allows conserving the enzyme expressed in an optimized environment, particularly with other enzymes and cofactors. It should also allow a better durability. The immobilization of bioactive particles for coating formation has been deeply studied [6-8]. One way of obtention of stable and active coating is to use sol-gel method. Using Si based coatings allows to tune water content, mechanical constraint and particles accessibility. The major drawbacks being the gestion of acidic pH during hydrolysis and condensation phases and the alcohol produced at these steps, with potentials of reducing enzymatic



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activity. Different strategies of partial hydrolysis, alcohol removal and buffering have been used successfully for bacteria, enzymes or yeasts [9-10]. In the present work, freeze-dried *Pseudomonas putida* ATCC 47054 was incorporated in sol-gel coatings used for formaldehyde degradation. Coatings resistance and thickness characterisation tests were also performed. Enzymatic accessibility, formaldehyde diffusion and water availability will be key parameter for optimised depollution coating.

2. Materials and methods

2.1. Coatings preparation

Freeze dried *Pseudomonas putida* sp. cells were introduced in hydrophilic sol-gel coatings composed by a mixture 3-glycidoxypolypropyltrimethoxysilane (GPTMS) and tetraethoxysilane (TEOS). A part of PVOH was also added to allows room temperature reticulation. After 24h of hydrolysis and condensation through intense stirring, alcohol removal and buffering were performed before microorganism addition.

Different formulation parameters, namely precursor ratio, polyethylene glycol (PEG) concentration and microorganism content, were used to optimize coatings effectiveness on formaldehyde degradation. Eight coatings formulations were prepared (Table 1). Coatings were applied by spraying to a surface of a thin-layer chromatography (TLC) paper and stainless steel blades in six layers.

Coatings were selected to be able to increase enzymatic activity for formaldehyde degradation (considering pH, reticulation, solvent). After 24 h of application, the paper was stored at room temperature (about $18 \pm 2^\circ\text{C}$) and 37°C to evaluate formaldehyde efficiency throughout storage time.

Table 1. Sol-gel coatings formulation

| | GPTMS ratio | PEG (g) | Cells concentration (% w.w ⁻¹) |
|---------|----------------|---------|--|
| Serie 1 | + | 8 | 40 |
| Serie 2 | + | 0 | 40 |
| Serie 3 | - | 8 | 30 |
| Serie 4 | - | 0 | 30 |
| Serie 5 | + | 0 | 30 |
| Serie 6 | - | 0 | 40 |
| Serie 7 | + | 8 | 30 |
| Serie 8 | - | 8 | 40 |

2.2. Formaldehyde quantification

Formaldehyde degradation by bacterial cells incorporated in sol-gel coatings stored at two different temperatures was determined every week, during 5 weeks. The coated paper was put in contact with the reactional mixture in aqueous solution containing formaldehyde (1 mM) and a co-factor NAD⁺ (1 mM). Formaldehyde degradation was quantified by a colorimetric method using the NASH reagent during 48 h [11].

2.3. Mechanical tests

Mechanical characterisation of coatings was also performed. Coatings thickness deposited on the surface of stainless steel was measured using an Elcometer 456 Thickness Gauge, 24 h after spraying. Three measures were taken in different positions of the coating surface.

Coating adhesion of under external solicitations were estimated by scratch test. An indenter was used to generate a scratch on the coated surface. Surface scratch load and length were user-dependent and were performed by three different technicians. Adhesion was evaluated according to ISO 2409:2013.

3. Results and discussion

3.1. Formaldehyde degradation by immobilized bacteria

Bacterial cells incorporated in the different sol-gel coating, prepared using different formulations, were able to degrade formaldehyde (Figure 1a). Series 1, 2, 7 and 8 obtained the best degradation yields (85, 80, 78 and 85%, respectively) after 24 h of deposition, compared to the others (30%, in average). After 5 weeks of storage at room temperature, formaldehyde degradation yields for the same series were 94, 53, 75 and 45% and the remaining series obtained 13.5% in average. A small decrease was generally verified, except for serie 1 that kept the highest degradation yield. The presence of PEG in serie 1 allowed the retention of large contents of water that improved the activity of formaldehyde-degrading enzymes, which were present in high concentration (40% (w.w⁻¹) of cells). On the other hand, after 5 weeks of storage at 37°C, formaldehyde degradation yield was about 10% in average for all the series (Figure 1b). Storage temperature decreased the enzymatic activity needed for the transformation of formaldehyde.

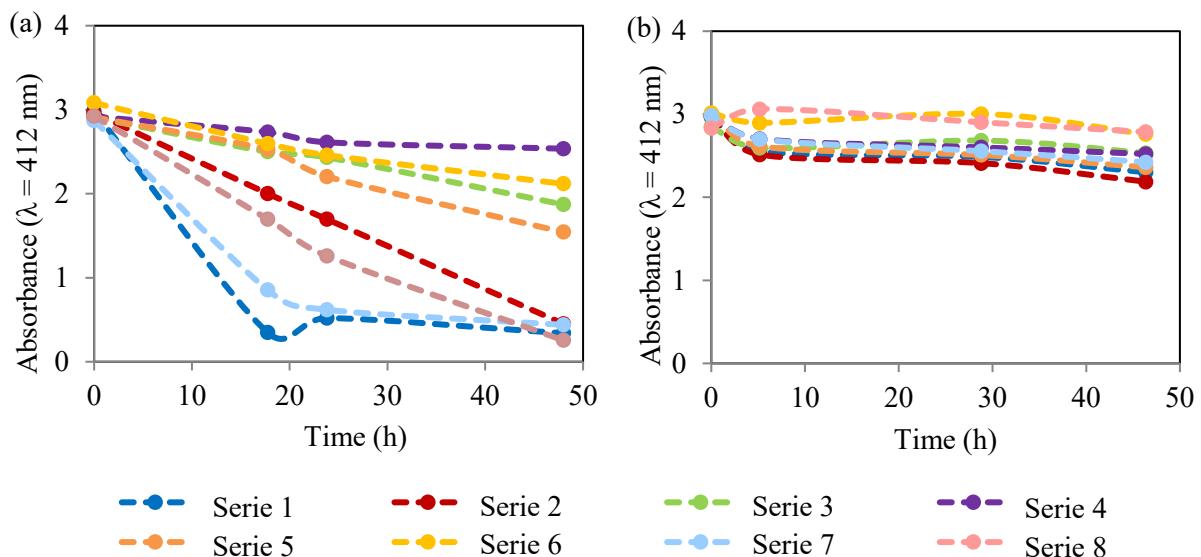


Figure 1. Enzymatic degradation of formaldehyde by immobilized *Pseudomonas putida* ATCC 47054 after 5 weeks storage at (a) Room temperature ($18 \pm 2^\circ\text{C}$); and (b) 37°C .

3.2. Mechanical tests

Thickness values determined for the stainless steel blades (Figure 2) are similar (average of 11.7 ± 2.3 μm) for the eight sol-gel coatings. Among the formulations prepared, the presence of PEG, which is added to retain the water in the sol-gel, seems to influence the thickness of the coating deposited on the surface of the substrate.

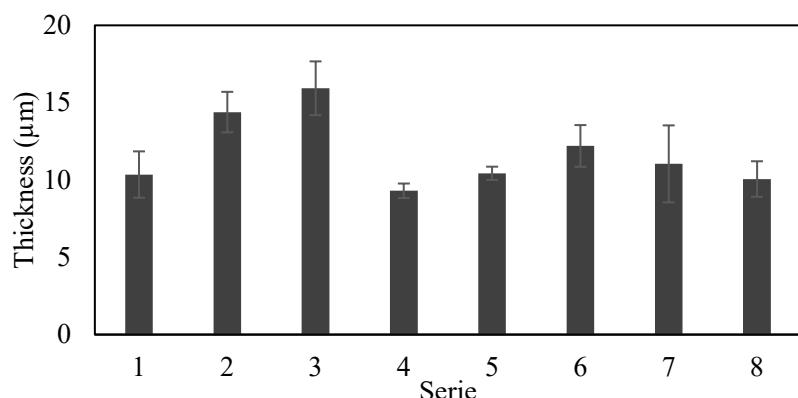


Figure 2. Thickness of stainless steel determined for the different sol-gel coatings.

Results from scratching tests performed for the sol-gel coatings prepared and deposited on the surface of the stainless steel blades are presented in Table 2. Sol-gel formulation composed by the lower concentrations of GPTMS, PEG and microorganism (serie 4) presented the higher abrasion resistance. This can be explained by the lower water-content of the mixture which allows higher adherence of the sol-gel to the substrate surface.

Table 2. Scratching tests evaluation. **0:** the edges of the incisions are perfectly smooth; none of the squares of the grid can be detached; **1:** detachment of small scales from the coating at the intersections of the incisions. Less than 5% of the grid area is affected; **2:** the coating has flaked along the edges and / or at the intersections of the incisions. The grid area is affected over 5% but less than 15%

| <i>ISO 2409:2013</i> | |
|----------------------|---|
| Serie 1 | 2 |
| Serie 2 | 1 |
| Serie 3 | 1 |
| Serie 4 | 0 |
| Serie 5 | 2 |
| Serie 6 | 2 |
| Serie 7 | 1 |
| Serie 8 | 1 |

In general, series 2, 7 and 8 showed a good adjustment between the mechanical properties, thickness and abrasion resistance, and the ability to degrade formaldehyde by enzymatic transformation.

4. Conclusions

Bioactive coatings containing freeze-dried bacterial cells were efficient in formaldehyde degradation in solution. The presence of PEG in the sol-gel improves the enzymatic degradation of the pollutant. Formaldehyde degradation efficiency can be kept by storing sol-gel coatings at room temperature. Further studies will be performed in order to evaluate the efficiency of the optimised coatings in gaseous formaldehyde decontamination. The industrial application of this technology for indoor air decontamination is very promising, although some optimisations must be performed regarding coatings performance, resistance, efficiency and durability, among others.

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