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Green antifungal waterborne coating based on essential oil microcapsules

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A R T I C L E I N F O

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A B S T R A C T

There is a great concern about the indoor microbial colonization especially in places that should have high standards of environmental hygiene. Besides, due it shortened the useful life of the coating by discoloration and degradation. Currently this is a problem that must be solved in an innovative and eco-friendly way. In this sense, the aim of the work was to develop a green waterborne paint formulated with microcapsules containing essentials oil as biocide agent. This novel hygienic coating would be applied as to protect indoor surfaces from fungi. The microcapsules were synthesized by interfacial polymerization. Melamine-formaldehyde (MF) resin was used for the microcapsule shell wall, and Lavandin and Tea Tree essential oils (EOs) as core materials. The synthesized microcapsules were characterized by scanning electron microscopy, Fourier transform infrared spectroscopy and particle size analysis. Preparation of acrylic waterborne paint was performed and the microcapsules MF-EOs were dispersed into the original paint just before application on commercial gypsum boards. The stability of the MF-EOs in the paint was successfully achieved. Two control paints were used, one containing EOs in free form and other without biocide.

The effectiveness of the microcapsules into paint film against spore suspension of *Aspergillus fumigatus* was evaluated by seeding the fungus on the painted surface. The fungal growth was evaluated according to ASTM D5590 standard specification. The score obtained from MF-Lavandin paint indicates just a trace growth onto the painted surface (<10 %). On the control and MF-Tea tree paints the growth was 70 %. The MF-Lavandin showed a high inhibition activity against fungal in the dry paint film compared with the one containing free form EOs paint.

1. Introduction

Microbial colonization of painted surfaces is a major concern because it shortened the useful life of the coating by discoloration and degradation. Besides indoor microbial colonization especially in places that should have high standards of environmental hygiene, as in the food industry and those related to human healthcare, is a great concern [1]. Among the most deleterious organisms are fungi, a very large and diverse group which is found in practically every ecological niche. The genera most frequently isolated inside buildings are *Penicillium, Aspergillus, Cladosporium* and *Alternaria* [2–5].

The antimicrobial coatings can supply an extra line of defense for maintaining hygiene standards. The most important property of hygienic coatings is its antimicrobial activity (against bacteria, and fungi) but they also need to be non-toxic and non-allergic to humans while, at same time, they must be non-odoros, durable and cost effective [6,7]. Water-borne paints arecomplex mixtures of polymers, pigments and functional additives. In these paints the vehicleisan emulsion of resin in water and have been created as alternative to solventborne paint because of the volatile organic compound (VOC) content in these paints is significantly lower, thereby reducing VOC emissions. They play an important role in cultural heritage conservation as consolidants and/or protectives for wall paintings, statues, stones and porous materials [8]. Among various coating constituents, binder represents the matrix structure and its amount relative to the amounts of pigments and fillers can significantly affect the structure, hence the barrier property of the coating. Acrylic polymers and copolymers are widely used as a binder in paint formulations due to their good adhesion and film forming properties [8]. Wall waterborne paints, commonly acrylic-based, are a target to microorganisms because they contain cellulosic compounds as

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thickeners. These compounds can be used by the microorganisms as carbon source and cause biodeterioration of the paint [9–11].

Conventional additives used as biocides in paints and coatings are often toxic, causing environmental pollution and health problems [12–14]. An eco-friendly alternative for replacement could be the use of natural products, such as essential oils.

Essential oils (EOs) are known for their antiseptic, i.e. bactericidal, and fungicidal, and medicinal properties ; they are commonly used in pharmaceutical, cosmetic and food industries [15]. Generally, EOs are volatile substances sensitive to oxygen, light, moisture, and heat, characterized by a strong odor. They are formed by aromatic plants as secondary metabolites [16,17]. These reported special characteristics could impair their applicability.

The coating technology based on the incorporation of microcapsules has emerged recently as a strategy in the advance of protective and functional materials, promising an environmentally friendly approach. The challenges for developing new materials are to accomplish more functionality with less material due to the increasing efficiencies of the smart approaches.

Microencapsulation provides many benefits for coatings containing biocides. The microcapsule protects the biocide from degradation and allows the proper control of the release process, prolonging thereby the duration of the biocidal effect and reducing the waste of biocides [18,19]. In this sense, essential oils had been reported more effective when they are microencapsulated, thus improving their long-term function and durability [20]. There are several chemical methods for the microencapsulation of essential oils such as in situ polymerization, coacervation, and interfacial polymerization [18,21]. The in-situ polymerization relies on prepolymers formed in a continuous phase. It involves monomer intercalation followed by polymerization, which can facilitate extraordinarily high coverage and shell strength owing to the chemical polymerization occurred on the surface of core material [22]. Melamine-formaldehyde resins (MF resins) are derived from the polycondensation of melamine and formaldehyde molecules, which represent a widely-used coating shell for the preparation of core-shell microcapsules. The characteristics of microcapsules, such as morphology and particle size distribution, rely on the preparation conditions including the rate of shear, the shearing period, the kind of emulsifier used, and the viscosity of the core material [23]. The aim of this work is to develop a green waterborne paint formulated with melamine-formaldehyde (MF) microcapsules containing essentials oil as biocide agent and to evaluate the antifungal activity. The innovation consists on apply microcapsules loaded with bioactive in hygienic coatings that make it a potential protector for indoor surface against fungal growth.

2. Materials and methods

2.1. Chemicals

Poly (ethylene-alt-maleic anhydride) (Poly (E-MA)), formic acid were purchased from Sigma Aldrich (Argentina). The commercial melamine-formaldehyde (MF) prepolymer Beetle® PT312 (73 % solids content and 0.2 % to 0.3 % free formaldehyde) was purchased from BIP Limited (Oldbury, UK). Lavandin (*Lavandula hybrid* var. *Abrialis)* oil was purchased from Indukern (Brazil)while Tea Tree (*Melaleuca alternifolia*) oil was purchased from Sigma Aldrich (Argentina). Agar-agar was supplied by Parafarm (Argentina) and proteose peptone was purchased from Oxoid (Tecnolab, Argentina). All materials were used as received without further purification.

2.2. Preparation of the MF microcapsules by in situ polymerization

The MF microcapsules were prepared using a modified in situ polymerization procedure [24]. MF resin was used for the microcapsule shell wall and two essential oils (EOs) as core materials. The biocide agents studied were Lavandin (LVO) and Tea Tree (TTO) essential oils. Experimentally, 2.55 g of MF resin was first dissolved (in a 250 mL beaker) in 30 g of water containing the surfactant Poly (E-MA) (3.3 % w/w) and sodium hydroxide (1.4 % w/w). Then, 17 g of essential oil was added slowly to the aqueous continuous phase under stirring to form the emulsion. The resulting mixture was stirred at 1000 rpm for 1 h. After this time, the pH of the reaction mixture was adjusted to 5 using formic acid, and the temperature of the reaction was raised up to 70 °C and maintained for 3 h under stirring. After 3 h, the synthesized MF microcapsules were cooled down to room temperature, cleaned with methanol/water. The storage stability of the methanol / water solution microcapsules is approximately by 1 week at 8 °C. At the time of use, the microcapsules were suspended in water. Once incorporated in the paint, the useful life depends on the paint.

2.3. Characterization of the MF-Essential oil microcapsules

2.3.1. Scanning electron microscope (SEM)

Field emission scanning electron microscopy (FE-SEM) was used to characterize the structure of the MF-EOs microcapsules, in terms of shape, size, thickness of the shell wall and morphological surface of the manufactured microcapsules. The micrographs were performed by a FEI Quanta 650 microscope (FEI, USA) at high vacuum and operated at a voltage of 20 kV with 4.5 spot. Before imaging, the samples were dried at room temperature, supported on aluminum tape and then coated with Au-Pd.

2.3.2. Particle size distribution

Particle diameter of the MF-Essential oils microcapsule suspension and its size distributions were determined by light diffraction (LD) on a Mastersizer 2000 (Malvern Instruments, UK). Microcapsules were dispersed in water in a sonicator before testing. The refractive and absorption indexes used to determine the particle size distribution were 1.33 and 0.1, respectively. The particle size was expressed as the equivalent volume diameter and three replicates were performed for each batch of microcapsules, to reduce error, an average curve and standard deviations were calculated.

2.3.3. Fourier transform infrared (FT-IR) spectroscopy

The infrared spectra of the MF-Essential oils microcapsules and EOs (Lavandin and Tea Tree), as KBr pellets, were obtained by a FT-IR (Spectrum, Perkin Elmer, USA) spectrometer according to diamond ATR method, in order to identify their chemical structure. The FT-IR spectra were recorded in the 4000 to 400 cm^{-1} range and scanned with background correction at 60 scans with 4 cm^{-1} resolution.

2.4. Preparation and characterization of antifungal waterborne coating

2.4.1. Paint formulation

Acrylic waterborne paint was formulated without any kind of biocide; the composition is shown in Table 1. Paint preparation was performed in a high-speed disperser. In the first step water was mixed with dispersing, antifoaming and thickener additives. Then, the

Table 1
Paint composition.

Component	% (w/w)
Water	25.20
Antifoaming	0.30
Cellulose Thickener	0.50
Dispersing agent	0.45
Wetting agent	0.15
Pigments	64.3
Resin	7.20
Mineral spirit	1.30
Butyl glycol	0.60

pigments (titanium dioxide, calcium carbonate) were added and finally the resin (an acrylic styrene) together with the co-solvents. After preparation, the paint was filtered and kept in a closed jar under laboratory conditions until use.

Two samples of 8 g with waterborne paint were fractionated just before applied. To each sample, a total of 2% (w/w) essential oil was added, either in free [25,26] or encapsulated form. The concentration of MF microcapsule suspension added into a wet paint correspond to a 6% (w/w) by weight of the total paint composition. As negative control a paint without biocide was used. After, the formulation only requires a simple, but thorough, mechanical stirring of the paint system that ensures a homogeneous dispersion of the added substances.

Commercial gypsum boards, cut into test pieces (2.5cm × 2.5cm) and sterilized at 121 °C for 20 min, were used as substrate. The panels were painted with two coats of the formulated paints. Then the panels were kept under laboratory conditions for 15 days to cure the paint.

2.4.2. Characterization of control paint

The hiding power of control paint was evaluated by ASTM D2805 Standard Method [27]. A paint film was extending uniformly with an applicator (75 μm gap), onto a combination black-and-white board. If the black-and-white squares are still seen after drying, another application is done, until complete hiding. Results are given by the wet thickness of the films applied to hide the board.

The drying time at room temperature was evaluated by ASTM D1640 Standard Method [27]. The set-to-touch time is determined lightly touching the film after application with the tip of a clean finger. Immediately, the fingertip is placed against a piece of clean, clear glass to determine whether the paint does not adhere to the finger or transfer to the glass. Is considered dry-through time the time elapsed until the paint is not distorted or detached when the thumb is applied to it and rotated through a 90° angle.

The washability parameter of wall paints, also referred to as 'resistance to scrubbing' or 'resistance to wet abrasion', was determined by ASTM D2486 Standard Method [27]. In this case, the coating was applied to a black plastic, dried and scrub with a nylon brush. The number of back-and-forth strokes (cycles) required to remove the film is determined.

The color and gloss measurements were performed by a ByK Gardner gloss-meter.

2.4.3. Fungal strain and culture

Aspergillus fumigatus, isolated from contaminated interior paints, was used in the study.The main reasons for choosing *Aspergillus* was that this fungus is a primary colonizer of building materials because its low water activity request, fast growth, ease viewing and major resistance to biocide [2,28].

The fungus was cultured in agarized media (AM): 1.5 g agar-agar, 1.0 g dextrose, 0.5 g protease peptone, 0.1 g KH_2PO_4, 0.05 g $MgSO_4$ and distilled water up to 100 mL and incubated at 28 °C for 7 days. The spore suspension was prepared from an AM culture and incubated in the conditions before mentioned. The spores were removed from the plate and placed in a test-tube with 5 mL of NaCl 0.85 % (w/v) and 0.005 % (w/v) of Tween-20. The spore concentration was adjusted to 10^6 spores/mL employing a Neubauer chamber.

2.4.4. Antifungal assay on paint film

Once the paint curing time is over, the panels were irradiated with U. V-light for 20 min, for providing a superficial sterilization and, then, placed into Petri dishes containing filter paper moistened with 1 mL of sterile water. Each panel was inoculated with 100 μL of the spore suspension of *A. fumigatus* and kept in a culture chamber at 86 % RH and 28 °C for 4 weeks. Four replicates were used for each paint. The fungal growth was estimated as a percentage of surface coverage. The panels were scored according to ASTM D5590 standard specification (Table 2) [29]. Additionally, observations by scanning electron microscopy (SEM) were made. The selected samples for further studies were the control and those that presented satisfactory results. The panels were prepared as follows: small sections of the panel's surfaces were cut and fixed in 2.5 % v/v gluteraldehyde (24 h) and dehydrated in graded series of ethanol solution from 20 % v/v at 100 % v/v. Finally, they were dried with the critical point and coated with gold. The coated samples were examined by a FEI Quanta 650 microscope (FEI, USA), at high vacuum. Besides, the internal structure of the microcapsules into the dry paint film was examined by cross-sectioning of the microcapsules.

Table 2
Fungal growth qualification over the surface (ASTM D 5590).

Growth observation	Qualification
None	0
Trace of growth (<10 %)	1
Light growth (10–30%)	2
Moderate growth (30–70%)	3
Heavy growth (>70 %)	4

3. Results and discussion

3.1. Characterization of the MF-Essential oil microcapsules

SEM photographs of the structure and morphology of microcapsules prepared by the *in-situ* polymerization method, containing the essential oils, are shown in Fig. 1 A and B. As can be seen, microcapsules have spherical shape and a smooth surface.

The graphical distribution of mean diameters for the different microcapsules is also indicated in Fig. 1 A and B. Both microcapsules present bimodal distribution in which the volume fraction of the first peaks is ∼ 2%. This bimodal shape distribution is consistent with the SEM images, where two sizes particles were observed. The mean particle sizes showed changes depending on the essential oil employed. The average particle size diameter was 15 ± 3.0 μm in the case of Lavandin microcapsules while those Tea Tree size microcapsules reached 22 ± 7.0 μm. Thus, the prepared microcapsules in this study seemed to be adequate for incorporation into a paint, due to their mostly monodisperse size distribution. In addition, the greatest mean particle sizes of MF- Tea tree can be attributed to the viscosity of the core material used for microencapsulation, which is an important factor affecting the particle size of the MF microcapsules [21].

FT-IR studies were conducted to confirm whether the MF-Essential oil microcapsules were prepared successfully or not. Fig. 2 A and B depict FTIR spectra of microcapsules containing LVO or TTO as core material and melamine-formaldehyde as the shell wall, respectively.

First, melamine-formaldehyde resin can be assumed to constitute in the capsule shell walls because the secondary amine group N—H stretching vibration was observed at 3393 $cm^{-1}$and 3349 cm^{-1} in the respective spectrum (Fig.2. A (b) y B (b)). This result is supported by a previous investigation [30] of melamine-formaldehyde prepolymer prior to encapsulate core material which showed N—H vibration at 3300 cm^{-1}. The C—N stretching vibration was shown at 1339–1336 cm^{-1}. These findings partially matched those reported in a previous study [30,31] dealing with melamine formaldehyde as shell wall material of microcapsule. In addition, strong adsorption peaks located at 2967–2963 cm^{-1} are associated with the C–H stretching vibration.

Fig. 2 A (a) shows FTIR spectra corresponding to Lavandin oil. The basic components of the LVO are linalool, linalyl acetate, lavandulol, and citronellol [32]. These components have characteristic molecule groups such as −COOR and C═O. In Fig. 2A the strong absorption peak of the LVO due to carbonyl was found at 1741 cm^{-1}. Besides, other notable peaks appeared at 2970 cm^{-1}, 1460 cm^{-1}, 1371 cm^{-1}, and 1000 cm^{-1}, which are due to C—H stretching vibration, CH_2 asymmetric deformation, CH_2 symmetric deformation [33].

Fig. 2B (a) shows FTIR spectra corresponding to Tea Tree oil. The

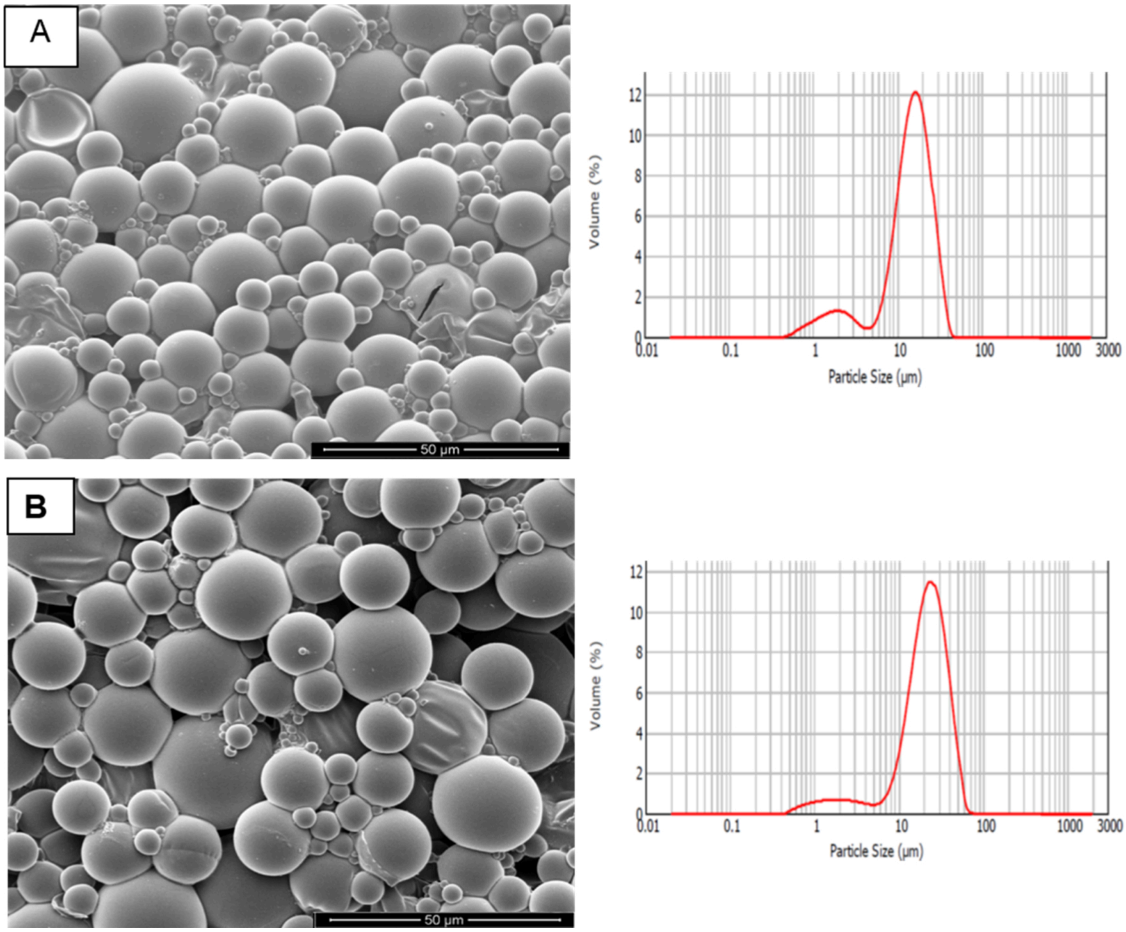

Fig. 1. SEM micrographs at 2400x magnifications and Particle-size distribution measured by laser diffraction for MF microcapsules containing: A) Lavandin oil and B) Tea Tree oil.

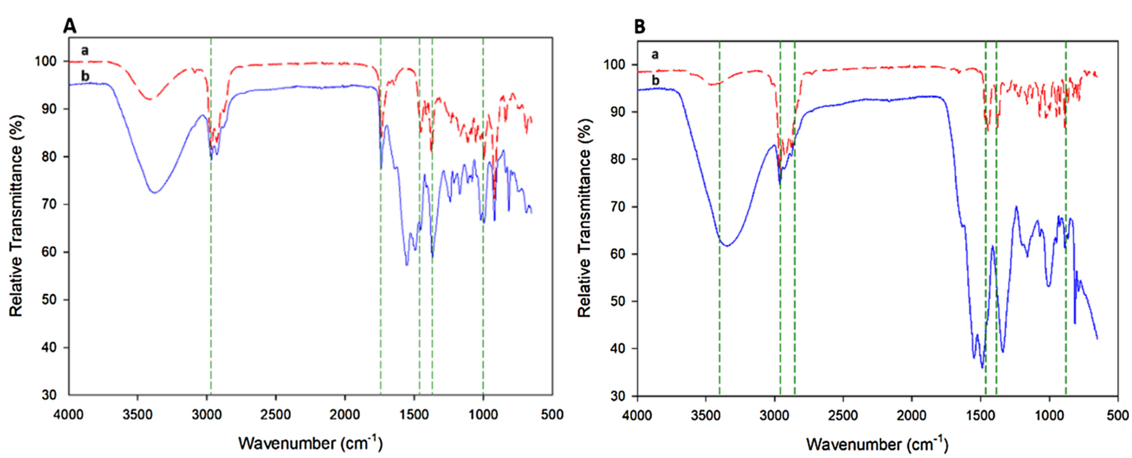

Fig. 2. FT-IR spectra: (A) a) Lavandin oil, b) MF-Lavandin microcapsules. (B) a) Tea tree oil, b) MF-Tea tree microcapsules.

TTO spectrum shows characteristic bands corresponding to its major components such as 1-terpinen-4-ol, γ-terpinene, 1,8-cineole and α-terpinene, among others, in published reports [34]. It shows characteristic peaks corresponding to the vibrations OH around 3400 cm^{-1}, alkyl C–H stretching vibration between 2960 and 2854 cm^{-1}. Between 1464 and 1385 cm^{-1} the C—H bending vibration was observed. Characteristic absorption band of vibration C—O—C aromatic rings appeared at 881 cm^{-1}.

In summary, all the characteristic peaks for LVO and TTO can be clearly distinguished in the spectra of the FT-IR spectrum of the microcapsule samples, which verifies that essential oils have been successfully encapsulated by melamine–formaldehyde resin as shell wall [35]).

3.2. Characterization and performance of antifungal waterborne coating

3.2.1. Characterization of control paint

The control paint evaluation parameters can be seen in Table 3. It is

Table 3
Control paint: evaluation parameters.

	Hiding power	Drying time		Washability	Color			Gloss
		set-to-touch	dry-through		L*	a*	b*	
Control paint	75 μm	90 min	142 min	1000 cycles	96.2	−0.43	1.55	2.2

shown that hiding power, drying times and washability were acceptable in this type of paints while its color is almost white, with very low gloss.

3.2.2. Observations of cross section of dry paint films

The incorporation of microcapsules into wet paint as an additive can be performed at any time prior to the application of the coating and not major workload and equipment requirements.

Two factors must be considered when preparing the microcapsules addition: the water content and the concentration of the microcapsule suspension [20]. The water content of the microcapsule suspension needs to be significantly decreased, using for instance centrifugation before the addition of the microcapsules to the wet paint. The reason is that a large amount of the continuous solvent phase can cause undesirable effects in the paint. The maximum concentration of a biocide dispersed in the coating is limited, as it often alters the imperative mechanical properties of the coating and sometimes also the aesthetic appearance [36].Therefore, a higher biocide concentration can be achieved in encapsulated form, prolonging its protection and without affecting the coating.

To confirm the feasibility of this method in producing added microcapsules into the waterborne coating, SEM characterization on dry paint film was conducted. The micrographs of the area inside dry paint film were revealed through cross sections and two different magnifications can be observed to 800x and 2000x, Fig. 3 A and B. It is clear a completely homogeneous dispersion of the microcapsules into the wet paint system compared to the control paint system. The stability of the microcapsules was observed and their geometry was unaffected by the surrounding chemicals. The robustness of the MF microcapsule was clearly perceived. In addition, Fig. 3.B (c) can be seen the oily content spilled from the microcapsule when it suffers a cut.

The core – shell structure of the microcapsules and thickness shell wall were confirmed by SEM. The cross-sectional view shown a microcapsule consisting of a hollow-core, Fig. 4 (a), and a dense shell wall MF with an approximately thickness of 0.94 ± 0.06 μm, Fig. 4 (b). Both attributes, particle thickness and robustness of the microcapsules show that the experimental conditions used in the *in situ* polymerization procedure such as, rpm and stirring time, temperatures and pHs and adjusting the amounts of material have been the appropriate.

All results indicate a successful production of microcapsules incorporated into the waterborne coating after the mechanical effort in manipulation process and curing process.

3.2.3. Antifungal assay

Fig. 5 shows in detail the results obtained in the antifungal assay on a

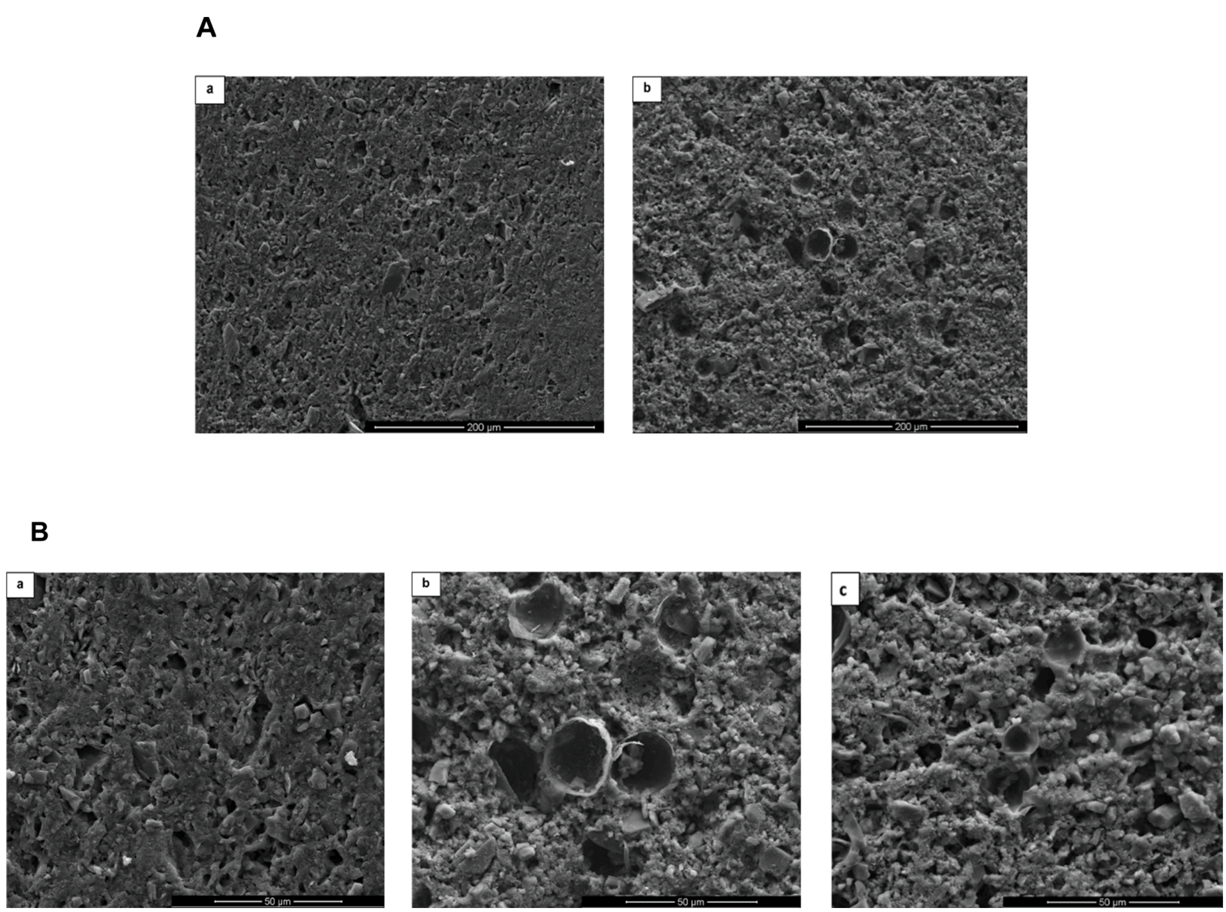

Fig. 3. SEM micrographs of cross section paint film. A) at 800x magnifications: a) control paint and b) MF-EO into the paint. B) at 2000x magnifications: a) control paint and b y c) MF-EO into the paint.

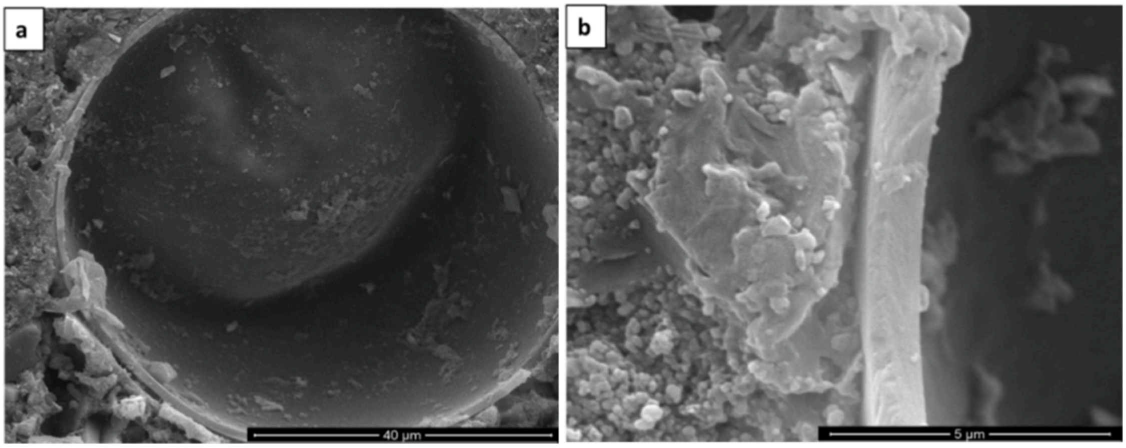

Fig. 4. SEM micrographs of cross section microcapsule. a) at 4000 x magnifications. b) view of shell wall thickness, at 30,000 x magnifications.

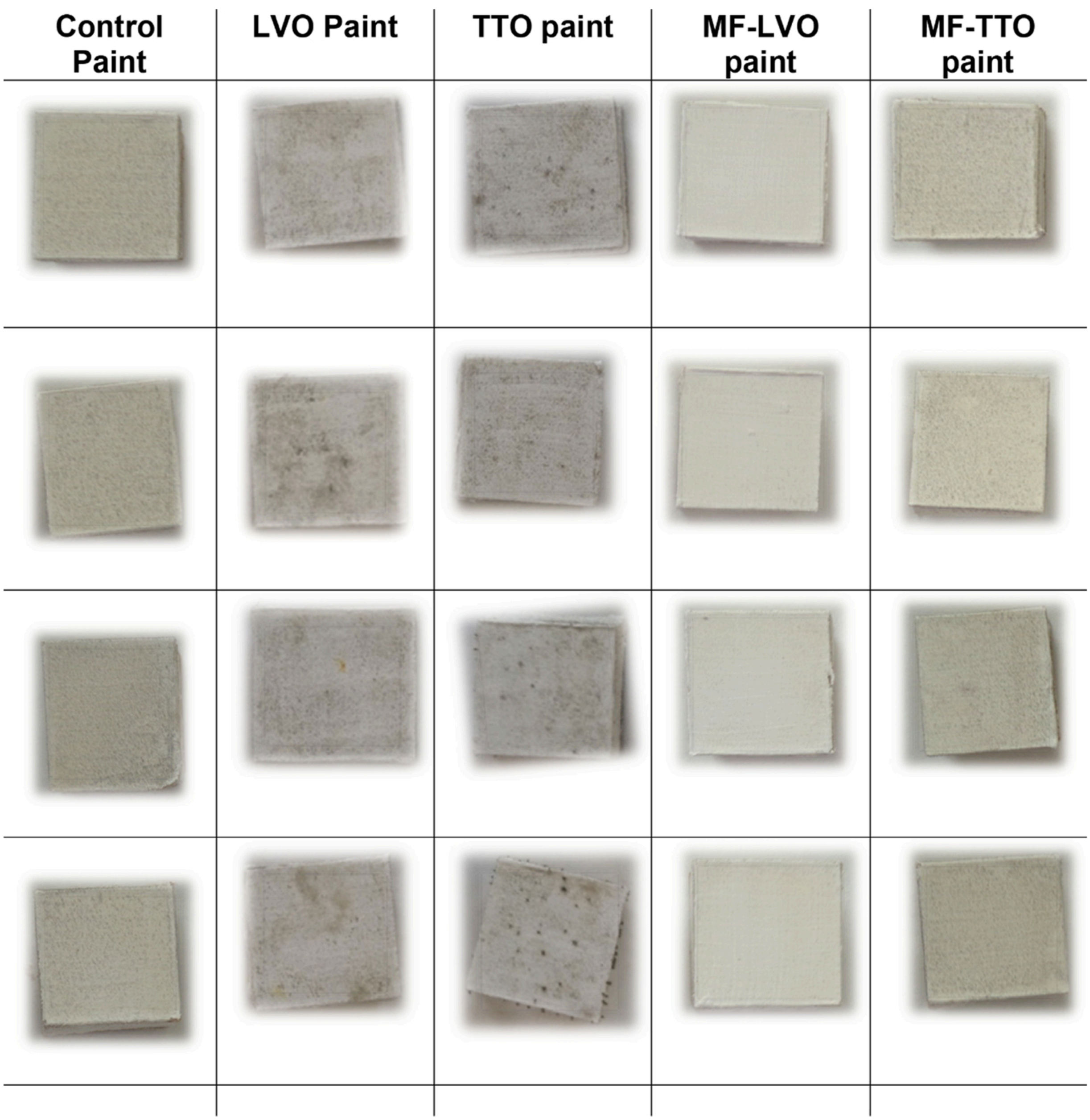

Fig. 5. Antifungal assay on paint film with EOs in free form and contained in MF microcapsules, exposed to *A. fumigatus* after 4 weeks and 28 °C (quadruplicate).

paint film exposed to *A. fumigatus* after 4 weeks and 28 °C. The test is done by quadruplicate. Paints films were compared; control paint (without EO addition), with the EO in free form and contained in the MF microcapsules.

Evaluation of the fungi growth degree on paints by ASTM D5590 (4th week) is shown in Table 4.

According to standard specification, the score obtained by MF-lavandin paint was 1, which indicates just a trace growth onto the painted surface (<10 %). On the other hand, the control, both essential oils in free form and MF-Tea Tree paints obtained the same score: 4 points, the highest qualification. This score corresponds to degree of coverage by the fungal growth higher than 70 %. Comparing the results is observed that *A. fumigatus* growth reduction was significant when MF-Lavandin paint was employed, suggesting that these microcapsules has fungal growth inhibition activity on the dry film whereas MF-Tea Tree did not affect the growth.

In the case of Lavandin oil, there is a clear demonstration of encapsulation effect, where the biocide is protected from the surrounding substances, compared to paint containing oil in free form.

3.2.4. Observations of antifungal behavior

Macroscopic observations were confirmed by SEM. Fig. 6 shows the micrographs of the fungal film of the control paint and paint with MF-Lavandin microcapsules.

As shown in Fig. 6 (a), the fungal colonization is abundant, with a wide mycelial development covering all control paint surface together with many spores and conidiophores. The spores are dark pigmented and lead to an aesthetic damage in the paint. Moreover, it is well documented that spores are the principal source of fungal bioaerosol that cause irritative disorders (i.e. allergy and asthma) [5,37]. On the other hand, in Fig. 6 (b) it can be seen a poor mycelial development and a few spores and conidiophores. In addition, Fig. 6 (c), was observed only trace vegetative mycelium of *A. fumigatus* confirming, the antifungal activity of MF-LVO paint. Furthermore, Fig. 6 (c) again confirms the stability of the microcapsules by distinguishing them immersed into the paint film.

Therefore, the waterborne coating containing MF-LVO microcapsules presents an optimal antifungal performance against *A. fumigatus* due to the addition of the prepared microcapsules.

The antifungal behavior of the coatings depends on the presence of the EO inside the microcapsules, so in order to avoid fungal growth. The release from the core oily material of the microcapsule diffuses thanks to the fact that it overcomes two types of barriers. One of them is the porosity of the shell wall microcapsule and the other is the internal channels of the paint matrix, to finally reach the outer surface of the coating [36].This means that the sustained release is controlled by the permeation through the microcapsule barrier (the shell) and the coating matrix [38]. Thereby, the purposes of encapsulating the biocides into micrometer-sized reservoirs are controlled release of the active preventing absorption of the external medium and the possibility to prolong the protection of coatings. It is best-known liquid core are oils and their entrapment inside a protective shell, it not only prevents the rapid and undesirable degradation but also light, air and heat [39]. On the other hand, as has been commented, a higher concentration of biocide when the encapsulation method is uses.

Table 4
Fungal growth qualification over the paint surface.

Paints containing	Code	Qualification
None microcapsules	Control	4
Lavandin oil in free form	LVO	4
Tea tree oil in free form	TTO	4
Lavandin oil microcapsules	MF- LVO	1
Tea tree oil microcapsules	MF-TTO	4

4. Conclusions

This work has proposed the use of melamine-formaldehyde resin microcapsules containing two different essential oils as a natural biocide, for the development of a green hygienic coating. The formation of microcapsules containing LVO and TTO obtained by polymerization *in situ* was achieved. The MF-EO microcapsules were evaluated regarding their morphologies, size distribution, and chemical structure. These has evidenced smooth surface morphologies and the mean particle sizes reached values around 15 and 22 μm. FT-IR studies have demonstrated that the essential oils were successfully encapsulated by melamine-formaldehyde as shell wall. These microcapsules were effectively incorporated in the paint in addition to achieving a correct homogeneous dispersion and adequate stability and robustness in the paint system

It is important to keep in mind that a paint is a complex system, and, at the same time, essential oil is a volatile compound. For this reason, it is important to achieve a good system of encapsulation to reduce the loss of essential oil and decrease the possible interaction with others paint components, thus, the oil will be available exclusively as biocide. In this sense, the results obtained in the antifungal test on paint film were promising. It can be seen that the growth reduction against *A. fumigatus* was satisfactory when MF-Lavandin were incorporated into the paint, evidencing the protective effect of the microcapsule in front of Lavandin oil in free form. This suggests that *Lavandula Abrialis* essential oil encapsulated in MF microcapsules has fungal growth inhibition activity in the dry paint film.

This innovative development of waterborne coating based on MF-microcapsules loaded with essential oil could be used as a potential green protector for indoor surface against fungal growth.

This kind of coatings has a good potential, given that the essential oil is a natural resource, renewable and eco-friendly. An inconvenience, in this time, is the cost of raw materials. When the process is considered for mass production, cost can be high, but may be acceptable if they are considered for special applications, those cases where conventional technology fails or when restrictions to traditional biocides would be implemented. Further experimental investigations are needed and work together with companies to make feasible the selection of ideas, technologies and eco-friendly compounds for the generation of commercial products.

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CRediT authorship contribution statement

M.V. Revuelta: Conceptualization, Visualization, Methodology, Investigation, Writing - original draft, Supervision. **S. Bogdan:** Visualization, Methodology, Investigation, Writing - original draft. **E. Gámez-Espinosa:** Methodology, Investigation. **M.C. Deyá:** Visualization, Methodology, Supervision, Project administration, Writing - review & editing, Funding acquisition. **R. Romagnoli:** Methodology, Resources, Project administration, Funding acquisition.

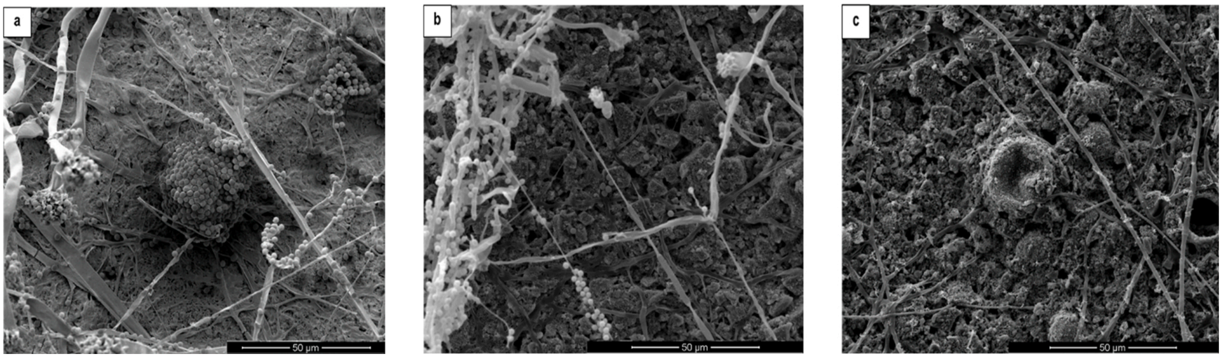

Fig. 6. SEM micrographs: a) control paint, b) LVO paint and c) MF-LVO paint, exposed to *A. fumigatus* after 4 weeks at 28 °C, at 2000x magnifications.

Declaration of Competing Interest

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.

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