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POSSIBILITIES OF SURFACE TREATMENT OF PLASTERS BASED ON CLAY

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Abstract

One of the key challenges associated with traditional buildings constructed from vernacular materials is their response to climatic agents and microorganisms. These present a significant challenge also for the surface of constructions based on unbaked clay elements. This paper presents the possibility of creating plasters based on clay, lime and other additives, their behaviour in response to mould growth, and the potential for coating treatments to enhance their resistance to the harmful action of microorganisms. The antifungal treatment was conducted with the objective of exploring the potential for recycling expired non-food household and medical products. The experimental results indicated the initial development of colonies of Penicillium notatum and Aspergillus niger and subsequently demonstrated the potential for enhancing resistance to mould action through pellicular treatment and/or surface impregnation. It can therefore be concluded that not only is it possible to create clay-based plasters with enhanced performance, but also that this represents an original contribution to the implementation of the concept of the circular economy.

***Key words**: antifungal treatment, circular economy, clay soil, plaster mortar, vernacular constructions.*

INTRODUCTION

Vernacular architecture is a foundational model for contemporary energy-efficient construction, given its capacity to adapt to local microclimates and environmental conditions. Recent studies have demonstrated its exceptional energy conservation performance, often exceeding initial scholarly assumptions (Abdelrazek & Yılmaz, 2020; Martinovic et al., 2023). In Romania, earth-based constructions, primarily adobe and wattle-and-daub, account for approximately 32.9% of buildings nationwide, predominantly as single-story structures built before 1992, with a higher prevalence in lowland counties where timber resources are less abundant compared to hilly and mountainous areas (INCD URBAN-INCERC). Structures built with unfired loamy soil represent some of the earliest forms of building technology. A variety of construction techniques have been utilised across the globe over time, including the construction of rammed earth walls, cob walls, and walls constructed from naturally dried unfired clay bricks. These techniques have been preserved and continue to be employed in various regions, exhibiting regional variations influenced by local resources and traditional building practices. These constructions present numerous advantages: they contribute to a healthy indoor environment by maintaining thermal stability and consistent humidity levels, while emitting no toxic substances - a critical factor in reducing the risk of respiratory diseases and allergies, they are cost-effective to construct, require minimal specialist labour, have a low environmental impact due to the limited use of raw materials, integrate harmoniously with local architectural traditions, and have a minimal impact on the surrounding landscape. These specific elements have the capacity to exert a considerable influence on the enhancement of hygiene and health standards for occupants, whilst concurrently effecting the mitigation of energy consumption and pollutant emissions. This, in turn, results in a contribution to environmental sustainability (Androutsopoulos et al., 2020; Măgurean & Petran, 2023). There are many studies in the literature that demonstrate the benefits of building with clay using indigenous techniques in different geographical and climatic conditions (Minke, 2006; Bui, 2009a Ciurileanu & Bucur, 2011). However, the main challenge with these constructions is the high risk of cracking during the drying process and their low resistance to water and weathering, which often requires frequent surface repairs and maintenance (Vural et al., 2007; Bui et al., 2009a; Kiroff & Roedel, 2010;). The scientific literature indicates that clay soils used for earthworks must contain a minimum of 15-16% clay to ensure the plasticity and workability required for proper handling (Vural et al., 2007; Kiroff & Roedel, 2010). In addition, to effectively control the risk of cracking, the acceptable linear shrinkage should be in the range of 3-12% for soft mixes and 0.4-2% for drier mixes (Minke, 2006; Jayasinghe & Kamaladasa, 2007). A variety of organic or inorganic additives of plant, animal or industrial origin may be used to achieve these indicators (Bahobail, 2012). However, increasing the resistance of the exposed surface to climatic or biological factors remains a challenge, although these additives can significantly improve the bulk performance of the clay composite. Research aimed at identifying surface treatment solutions for clay surfaces has been reported in the literature, with some researchers analysing the possibility of developing sol-gel solutions based on TiO_2 (Calabria et al., 2010) while others have focused their attention on more traditional solutions, such as natural oil-based waterproofing agents of vegetable or animal origin, sodium silicate, silicon nanoparticles, titanium dioxide, silica nanoparticles, silane-siloxane, beeswax, ethyl silicate, organic polymer products, lime-based paints and others (Ferron, 2007; Li et al., 2009; Ferron & Matero, 2011; Pacheco et al., 2016; Stazi et al., 2016; Lanzón et al., 2017; Camerini et al., 2019; Elert et al., 2019; Rescic et al., 2023).

On the other hand, the cosmetics, pharmaceuticals and other non-food household products industries are experiencing rapid growth in correlation with the demands of modern society. The cosmetics market tends to grow year on year, making it the third fastest growing market, with the global beauty and self-care industry estimated to be worth USD 341 billion in 2020, USD 565 billion in 2022, USD 758 billion in 2025 and USD 480.4 billion in 2030 (Nciri et al., 2022; Morganti et al., 2023). It is therefore of interest to find solutions for the recovery and recycling of expired non-food household products, cosmetics, pharmaceuticals, various aromatic oils, etc., so that they no longer pose a pollution risk when disposed of in sewers or landfills (Juliano & Magrini, 2017; Cinelli et al., 2019; Bashir et al., 2021; Silletta et al., 2024). Research has shown that, in addition to specific functional ingredients, cosmetic products, for example, are rich in phenolic compounds, flavonoids and organic acids known for their antimicrobial properties, preservatives and bioactive molecules for the treatment of skin conditions (such as dermatitis, atopic eczema, atopic dermatitis or acne) (Varvaresou et al., 2009; Houston et al., 2017; Halla et al., 2018; Hamrita et al., 2022; Sandle, 2022).

A distinctive attribute of cosmetic products is the homogeneity of their component formulations, which is characterised by minimal variation in size, extending down to the nanometric level, even for mineral-derived products (Silletta et al., 2024).

The utilisation of non-food household products in the domain of construction has been documented in the extant literature. For instance, a group of researchers from the Department of Civil Engineering, PVPIT Budhgaon, Maharashtra, India (Patil et al., 2021) demonstrated the potential use of an aqueous soap solution to induce a high volume of pores in the cementitious matrix, enabling the production of concrete with a density lower than 1000 kg/m³, commonly referred to as "floating concrete". Moreover, other studies have emphasised the advantageous properties of soap, along with soap precursors such as oils or fats, in enhancing the performance of unfired clay bricks, particularly in terms of their resistance to water (Browne, 2009; Bahobail, 2012).

A group of researchers from Korea University of Technology & Education (Nciri et al., 2022) demonstrated that cosmetic waste can be integrated into the creation of green, sustainable road pavements. Specifically, the incorporation of cosmetic waste, such as lipstick (completely soluble at temperatures above 50ºC) or other cosmetic products containing oils, fats, emulsifiers, wax, fillers, mica, silicon, titanium oxide, or zinc oxide, into asphalt mixtures can act as additive materials that improve rheological properties, viscosity, and elasticity. This contributes to reducing fatigue-induced cracking, enhancing aging resistance, and improving durability, including through the influence of crystallization/hardening mechanisms.

This exploratory study aims to assess the potential reuse of expired cosmetic-pharmaceutical products for protecting clay surfaces against mould growth, which can act as a trigger for allergies. While the permeability of these compounds through clay materials represents an important consideration that may affect treatment effectiveness and durability, this initial investigation focuses on surface application effects. The method employed for repurposing these expired products involved their integration as functional additive raw materials in formulations designed for surface application, resulting in a protective coating with increased resistance to microbial activity. It is important to note that the scope of this research does not extend to evaluating the potential health implications associated with the utilisation of these products for construction purposes beyond the specified scope, nor does it include in-depth analysis of treatment permeability through the clay matrix - aspects that deserve further investigation in future studies. Therefore, the aim of the present study is to analyse the feasibility of recycling non-food household waste (namely cosmetic-pharmaceutical products) through the development of a protective and finishing coating aimed at enhancing the resistance of unfired clay construction surfaces to biological agents.

MATERIALS AND METHODS

The raw materials utilised in the fabrication of clay-based plaster mortar composites for the protection and finishing of vernacular construction surfaces comprised the following: clay soil (named "clay") extracted from Valea Drăganului, Cluj-Napoca, Romania, hydraulic lime with the commercial designation RÖFIX NHL5, with an apparent density of 520 kg/m³ and a minimum compressive strength of 5 N/mm², river sand with a maximum grain size of 4 mm, and water.

To characterise the clay that was utilised in this study, an XRF analysis was conducted (Table 1).

Table 1. The oxide composition of the clay, determined through XRF analysis

Oxides	SiO_2	Al_2O_3	Fe_2O_3	CaO
Content [%]	74.17	12.74	4.38	0.7
Oxides	MgO	K_2O	Na_2O	TiO_2
Content [%]	1.0	1.43	0.73	0.05

This non-destructive analytical technique utilises the measurement of characteristic secondary X-rays to determine the elemental composition and concentration of materials, through the exposure of a sample to high-energy X-rays or gamma rays, which results in the emission of secondary X-rays due to the excitation of inner-shell electrons. The subsequent analysis of the resultant data enabled the determination of the clay's oxide composition and enabled prediction of its classification according to the standard mineralogical nomenclature. The analysis revealed that the clay in question belonged to the clay loam domain (Figure 1), as indicated by its specific mineralogical composition (Figure 2).

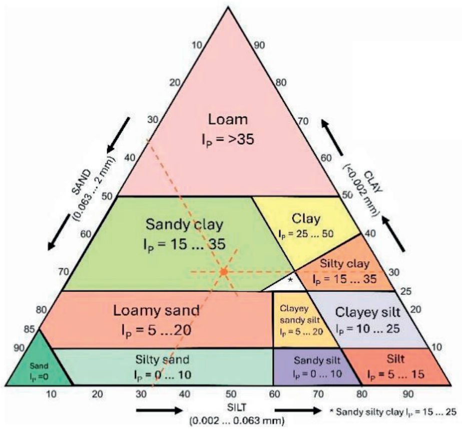


Figure 1. Characterization of the clay as a raw material – classification within the ternary diagram

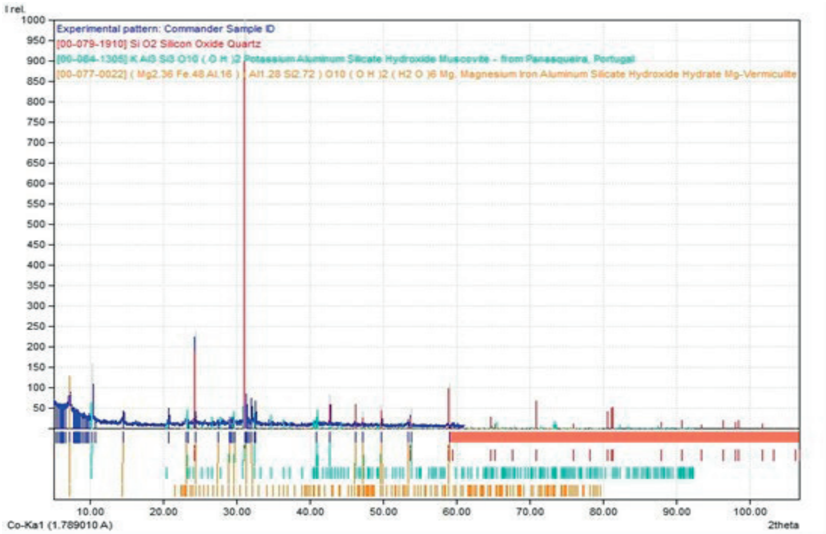


Figure 2. Mineralogical analysis of the clay used as a raw material

From a mineralogical perspective (Figure 2), experimental tests revealed the presence of predominant minerals, including quartz with approximately two-thirds of the composition, muscovite accounting for nearly one-third, and a minor fraction of vermiculite, along with montmorillonite, illite and kaolinite. (Minke, 2006; Niroumand et al., 2013; Dormohamadi & Rahimnia, 2020; Muñoz et al., 2020; Ige & Danso, 2022). In accordance with existing literature on the subject, these characteristics support the assessment that this raw material is suitable for use in the construction sector.

The selection of the hydraulic binder (hydraulic lime) was based on preliminary experimental tests and existing literature highlighting its beneficial effects (Minke, 2006; Pacheco-Torgal, 2015; Ige & Danso, 2022; Rescic et al., 2023) particularly the additional contribution of calcium oxide in mitigating drying shrinkage and reducing the risk of cracking. The water used in the preparation process was carefully dosed to maintain a consistent fresh mix consistency within a controlled range of 95 ± 5 mm.

In order to evaluate and analyse the performance of clay-based compositions intended for plastering applications, a series of mixtures were formulated and prepared under controlled laboratory conditions.

The proportional composition of the raw materials, together with the corresponding sample codes, is shown in Table 2.

Table 2. The ratio of raw materials in composites intended for use as plaster mortar

Sample code	Clay (%*)	Hydraulic Lime (%*)	Sand (%*)
P1	60	0	40
P2	57	3	40
P3	55	5	40
P4	50	10	40
P5	45	15	40
P6	40	20	40
P7	35	25	40
P8	30	30	40
P9	25	35	40
P10	20	40	40
P11	10	50	40
P12	0	60	40

*Note: It should be noted that the percentage indicated is that by weight of the total dry material.

For each clay-based composite, a number of indicators were evaluated to characterise the material and confirm its suitability for use as a rendering mortar, in particular:

- apparent density in the hardened state, according to the standardised method EN 1015-10;
- axial shrinkage, according to the standardised method STAS 2634;
- presence of drying cracks, assessed on visual inspection;
- flexural tensile strength and compressive strength, according to the standardised method EN 1015-11;

- adhesion to the substrate, tested on ceramic bricks and unfired clay masonry units, according to the standard method EN 1015-12.

The experimental evaluation of the physical and mechanical performance was conducted under laboratory conditions using prismatic specimens of 40×40×160 mm, formed in metal moulds and demoulded 72 hours after preparation. To assess substrate adhesion, flat specimens were prepared by applying a 3-5 mm thick layer of the clay-based composite to the selected substrates. All samples were stored for 40 days until a constant mass was reached, which was taken as an indicator of full maturation and drying of the material. The samples were stored under controlled laboratory conditions at 23±2°C and 65±1% relative humidity, and then subjected to experimental testing. In order to ensure the repeatability of the experimental determinations, a minimum of three samples were prepared for each case and the values reported represent the arithmetic mean of the individual measurements.

Subsequently, to analyse the resistance to microbiological attack, a test scheme was designed for four clay-based composites selected on the basis of their physical and mechanical performance. The test procedure consisted of three stages:

- evaluation of the behaviour of plastering mortars in an environment contaminated with mould spores;
- design, preparation and application of a protective product on the clay surface;
- validation of the antifungal performance by evaluating the behaviour in an environment contaminated with mould spores.

Samples were selected on the basis of minimum thresholds for four indicators identified in correlation with references from the literature: (Minke, 2006; Pacheco-Torgal, 2015; Muñoz et al., 2020; Eslami et al., 2022; Azalam et al., 2024):

- absence of drying cracks;
- adhesion to substrate: minimum 0.5 N/mm²;
- compressive strength: at least 2.5 N/mm²;
- bending tensile strength: minimum 1.5 N/mm².

In addition, a batch of clay-based composites without hydraulic lime content (P1) was subjected to a comparative analysis.

The analysis of the resistance of the clay-based composites to microbiological attack was carried out by exposing the samples to a highly contaminated cross-infection environment with *Penicillium notatum* (PN) and *Aspergillus niger* (AN) spores. For this purpose, individual culture systems were prepared for each sample type. A disc-shaped specimen, 15 mm in diameter and 3 mm thick, prepared in the same way as the prismatic specimens used for the determination of physical and mechanical tests, was placed in a Petri dish on a potato dextrose agar (PDA) nutrient substrate (prepared by dissolving 39 g/L of granular substance in warm water). The test sample and the culture medium were sprayed vertically from a distance of 100 mm with a solution containing *Penicillium notatum* (PN) and *Aspergillus niger* (AN) spores (10 µl PN + 10 µl AN/20 ml distilled water). The entire system was then sealed and incubated at a constant temperature (23±1°C) in a BIOBASE BOV-D30 incubator. At predetermined time intervals, the systems were evaluated by observing the appearance and development of mould and the degree of surface coverage on the material sample. The evaluation was performed by microscopic examination using a LEICA SAPO microscope. The results were quantified using two indicators: one to assess fungal growth (Table 3) and the other to estimate product performance (Table 4).

Table 3. Fungal growth assessment

Class	Fungal Growth Evaluation
Class 0	No fungal growth is visible upon microscopic examination.
Class 1	The growth is invisible to the naked eye but is clearly visible under a microscope.
Class 2	Visible growth to the naked eye, covering up to 25% of the surface area that was tested.
Class 3	Visible growth to the naked eye, covering up to 50% of the surface area that was tested.
Class 4	Visible growth to the naked eye, with a surface coverage of more than 50%.
Class 5	Heavy growth, with complete coverage of the tested surface.

Table 4. Product performance estimation

Category	Product Performance Estimation
0	The material is characterised as being inert or fungistatic, thereby serving as an unsuitable nutrient medium for microorganisms.
1	The material contains minimal nutrients or is minimally contaminated, resulting in restricted microbial growth.
2-3	The material demonstrates susceptibility to microbial attack and contains nutrients that facilitate microorganism growth.

The surface treatment solutions were prepared using expired non-food household products (cosmetic-pharmaceutical products) together with dispersion matrices. The raw materials used were:

- dispersion matrices:
  - commercially available liquid wax, natural beeswax dispersed in water, manufactured by Borma Wachs, Italy;
  - rabbit skin glue solution, made from rabbit skin glue granules (manufacturer: Divolo, Italy, commercially available) dissolved in warm water at a ratio of 1:10 by mass (one part glue to ten parts water).
- cosmetic-pharmaceutical products:
  - liquid pharmaceutical product for the treatment and prevention of fungal infections of the mucous membranes, particularly in the oral cavity, based on glycerine. It is an over-the-counter solution available in pharmacies that combines the properties of glycerine, borax (B) (sodium tetraborate, a substance with antiseptic and antifungal properties) and nystatin (NT) (a broad-spectrum antifungal agent that inhibits the growth and spread of fungi by disrupting their cell membrane);
  - liquid antifungal pharmaceutical product for the treatment of fungal infections of the skin and nails. It is available over the counter and contains the active ingredient naftifine (NF), which belongs to the class of allylamines with antifungal and antimicrobial properties. Naftifine inhibits enzymes involved in the formation of fungal cell membranes, thereby affecting their growth and survival. It is effective against several types of fungi, including yeasts and moulds;
  - hand cream for dry skin, made in Romania (contains 15% glycerine);
  - lipstick, produced in Romania (contains 50% glycerine);
  - lavender oil, produced by Ecoland Production SRL, Romania.

The selection of these raw materials was driven by the following criteria:

- representation of waste materials for which potential reuse solutions are being explored;
- affordability, accessibility, and absence of health risks to users;
- capability to serve as natural treatment solutions suitable for vernacular construction applications;
- liquid wax has been demonstrated to reduce the water sensitivity of clay surfaces by filling pores and forming a water-repellent sealing film, thereby enhancing resistance to water exposure and climatic agents. When utilised in an efficient manner during the mixing process, it functions as an effective dispersion medium for fine powdered substances and lavender oil;
- rabbit skin glue solution is recognised as an effective surface treatment material, frequently used in furniture and painting restoration, as well as in traditional and eco-friendly construction works. The formation of a varnish-like film is the result of the interaction of the glue with the substrate, and this results in strong adhesion. It is expected that this will enhance surface resistance to water exposure and climatic agents. The solution's application as an aqueous medium facilitates the dissolution of other raw materials (e.g., expired pharmaceutical products) while ensuring the effective dispersion of fine powdered substances;
- lavender oil is renowned for its antiseptic, antifungal, antimycotic, and antibacterial properties, as well as its effectiveness as a natural insect repellent. Its efficacy in treating contamination caused by *E. coli*, *P. aeruginosa*, *S. aureus*, *P. vulgaris*, *E.s faecalis*, *L. monocytogenes*, *B. subtilis*, *A. niger*, *P. chrysogenum*, and *C. albicans* has been well-documented (de Elguea-Culebras et al., 2016; Vasileva et al., 2018; Silletta et al., 2024);
- the improper disposal of expired cosmetic pharmaceutical products in household settings represents a significant environmental concern, as these materials have the potential to contaminate water sources and soil. The selected products are intended for external use, are available without medical restrictions, and do not present risks associated with use by children, pregnant women, or breastfeeding individuals. Furthermore, the glycerine content present in these products has been demonstrated to

enhance the flexibility of the protective film (for rabbit skin glue film, the required glycerine content is 5%). Finally, while some of these materials function exclusively as treatment agents to enhance resistance to mould growth, others can readily serve as pigments, thereby enabling a broad spectrum of colours and shades in the protective coating.

The evaluation of coating coverage uniformity was conducted by applying the treatment solutions to a white cementitious substrate characterised by a smooth surface (specimens fabricated from white cement paste, aged two years post-casting). Preliminary tests demonstrated that, under constant exposure and lighting conditions, the coatings were more visible on the white cementitious substrate compared to the clay substrate. The coding and composition of the treatment solutions are presented in Table 5.

Following the preparation stage, the treatment solutions were applied by brushing onto the surface of the selected clay-based composite specimens in two layers, with a 24-hour interval between applications.

Table 5. Ratio of raw materials utilised in the formulation of clay surface treatment solutions.

Sample code	Liquid wax (ml)	Rabbit skin glue solution (ml)	B+NT+NF Total content (%)	Cosmetics (g hand cream/g lipstick)	Lavender oil (ml)
S1	100	-	0.0017	10/7	1
S2	50	50			
S3	-	100			

Following the application of the second layer, the specimens were left to dry for a period of seven days. Thereafter, the contamination exposure procedure described earlier was employed to test the specimens. Figure 3 provides a schematic representation of the experimental research methodology.

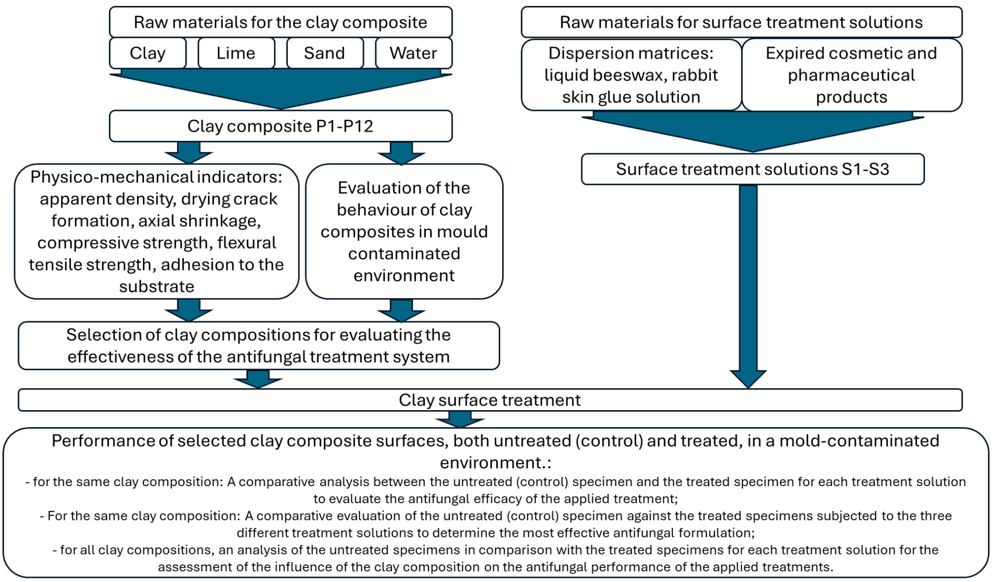


Figure 3. Schematic representation of the experimental research methodology

RESULTS AND DISCUSSIONS

The experimental results obtained in this study demonstrate that, in relation to physical and mechanical indicators, an increase in the hydraulic lime content in the composition results in a decrease in apparent density (Figure 4) and a reduction in axial shrinkage (Figure 5). These outcomes are particularly favourable for such materials, as they are accompanied by the absence of cracks.

However, it is crucial to acknowledge the unfavourable consequence of elevated hydraulic lime content, namely the decrease in mechanical strength, both in compression and flexural tensile resistance, which underscores the necessity for the identification of the optimal hydraulic lime content in each instance where the composition of the clay material is subject to alteration (Figure 6).

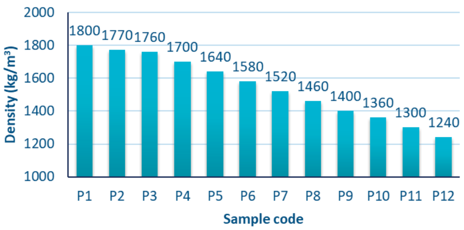


Figure 4. Bulk density of clay composites in their hardened state

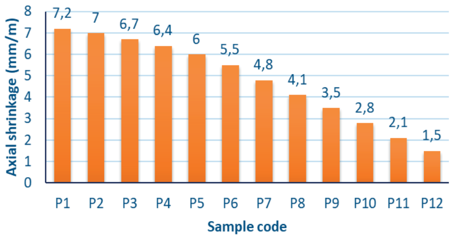


Figure 5. Axial shrinkage of clay composites

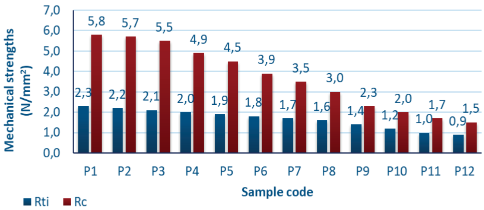


Figure 6. Mechanical strengths of clay composites

Concerning the adhesion of clay-based composites to the substrate, a Gaussian trend was observed (see Figure 7).

In particular, an increase in hydraulic lime content, coupled with a reduction in clay content (while maintaining a constant sand proportion in the total dry mix), resulted in enhanced adhesion to both adobe brick and fired ceramic brick surfaces. This adhesion exhibited a peak at composition P8 (30% clay, 30% hydraulic lime, 40% sand). Conversely, compositions where the lime content exceeds the clay content (P9-P12) exhibited a decline in adhesion to the substrate. It is therefore essential to establish the optimal hydraulic lime content to achieve a sustainable balance between its benefits and potential drawbacks.

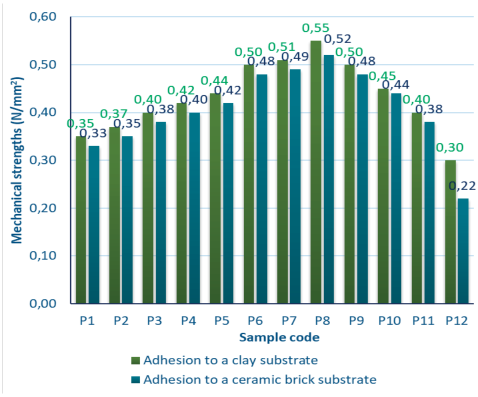


Figure 7. Adherence of clay composites to the substrate

The experimental results regarding the evaluation of the resistance of clay-based compositions under cross-contamination conditions with *Penicillium notatum* and *Aspergillus niger* revealed the following:

- All clay-based compositions invariably allowed the growth of one or both types of mould on their surface, although at different rates. The initial indications of mould growth (3 days after contamination) manifested in compositions with a reduced lime content, thereby validating the advantageous and sanitizing effect of lime.
- Following a period of seven days of exposure to the contaminated environment, all compositions exhibited indications of fungal growth, which was classified as Class 1 according to the classification system (i.e. invisible to the naked eye but clearly visible under a microscope). According to the performance estimation indicator (Table 4), all compositions were categorised as Category 1 (i.e. the material contains very few nutrients or is minimally contaminated, allowing only very limited fungal growth).
- During the exposure period of 7 to 33 days in the contaminated environment, visual and microscopic evaluations of the clay surfaces indicated varied behaviours. Some compositions exhibited a more intense development of PN (*Penicillium notatum*) colonies, while others showed a more pronounced growth of AN (*Aspergillus niger*) colonies. Additionally, the extent of

surface coverage by the microbiological film varied depending on the composition. Consequently, the analysis of the results has not permitted the establishment of a clear correlation between the clay composition and the predominant type of mould growth.

- The final evaluation, conducted 33 days after exposure to the contaminated environment (see Figure 8), revealed the following classification based on fungal growth: Class 1 (invisible to the naked eye but clearly visible under a microscope) for compositions P8-P12 (minimum 30% lime content), Class 2 (visible growth covering up to 25% of the tested surface) for compositions P3-P7 (minimum 5% lime content), and Class 3 (visible growth covering up to 50% of the tested surface) for compositions P1 and P2 (0% or 3% lime content in the dried material). In terms of product performance estimation, the P3-P12 compositions were classified as Category 1. The rationale behind this categorisation is that these materials contain negligible nutrients or minimal contaminants, which precludes substantial fungal growth. Conversely, the compositions with minimal or no lime content (P2 - 3% lime and P1 - 0% lime) were classified as Category 2. This classification is attributed to the fact that these materials lack the resilience to microbial attacks and contain nutrients that facilitate microorganism growth. One potential explanation for these results is the presence of residual organic content in the clay soil, despite the processing of the soil through sorting and sieving to remove vegetative materials prior to use. Consequently, in order to enhance the durability and hygienic properties of clay surfaces, the implementation of an appropriate surface treatment is considered beneficial.

The selection criteria for the clay-based compositions (absence of drying cracks; adhesion to the substrate of at least 0.5 N/mm²; compressive strength of at least 2.5 N/mm²; flexural tensile strength of at least 1.5 N/mm²) as identified in Figure 9 were applied, and three compositions that met all these requirements were selected for validation testing of the antifungal treatment through surface coating. These selected compositions are: P6 (40% clay, 20% lime, 40% sand), P7 (35% clay, 25% lime, 40% sand) and P8 (30% clay, 30% lime, 40% sand).

The protective and surface treatment solutions for clay-based surfaces, as prepared (Figure 10), were found to be homogeneous, easy to apply by brushing, and demonstrated good surface coverage. The most homogeneous solution was the one prepared with liquid wax (S1). However, with the addition of rabbit skin glue, the dispersion of raw materials derived from the recycling of expired cosmetic products became more challenging.

Despite this, the resulting coatings were found to be smooth, continuous, and uniform, ensuring adequate surface coverage. However, it was observed that the presence of rabbit skin glue in the treatment solution had a slightly negative effect on coating coverage capacity. This effect was particularly noticeable on the white cementitious substrate, which remained slightly visible beneath the treatment film, especially in the case of solution S3, where rabbit skin glue was used exclusively as the dispersion matrix (Figure 10d).

In relation to the investigation of the resistance exhibited by clay-based composites following treatment with a liquid beeswax-based solution, observations made after a seven-day exposure period (see Figure 11) indicate that the surface remains impermeable when exposed to a mould-contaminated environment. The moisture generated by the mould's metabolic processes manifests as water droplets, thereby supporting the hypothesis of the surface's hydrophobic nature. No signs of mould growth were present on the surface of the specimens. Furthermore, the testing systems exhibited a slight inhibition halo around the clay-based specimens, with the halo size ranging from 1 to 5 mm from the edge of the specimen, forming a circular pattern around it, as exemplified in Figure 11b-c. In accordance with the fungal growth evaluation system and product performance estimation criteria (Tables 3 and 4), it can be concluded that treating clay surfaces with solution S1 resulted in their classification under: Class 0 (No signs of fungal growth upon microscopic examination),

Category 0 (The material does not serve as a nutrient medium for microorganisms – it is inert or fungistatic).

The results of the 7-day exposure period in a highly contaminated environment demonstrated the treatment solution's high efficacy and a significant improvement in fungal resistance. The treatment of clay surfaces with solution S2, which uses a liquid beeswax and rabbit skin glue mixture (1:1 mass ratio) as the dispersion matrix, resulted in a loss of surface hydrophobicity compared to specimens treated with solution S1 after seven days of exposure to a contaminated environment (Figure 12).

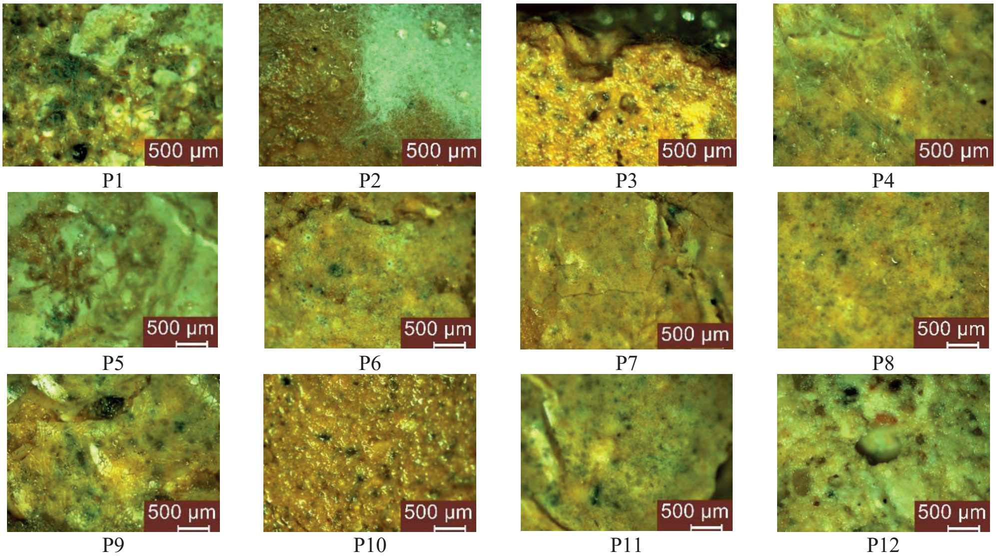


Figure 8. Optical microscopy images depicting the behaviour of clay composites in a mould-contaminated environment following 33 days of exposure.

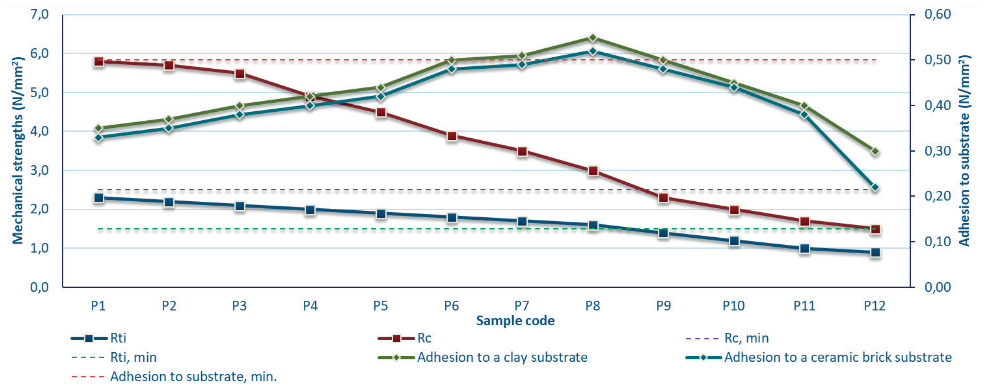
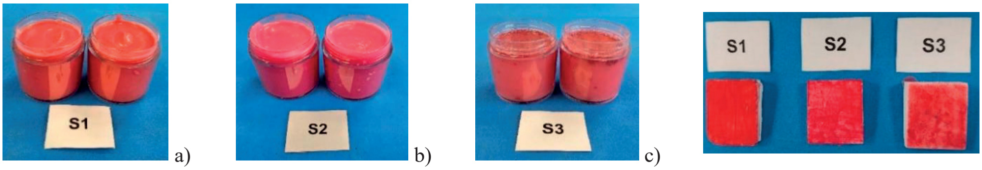


Figure 9. Selection of compositions for testing the efficacy of the antifungal treatment system.

It is noteworthy that no mould development was detected on the surfaces of any of the specimens. According to the fungal growth evaluation system and product performance criteria (Tables 3 and 4), treatment with solution S2 classified high-lime-content compositions (P7 and P8) in Class 0/Category 0 (no fungal growth detected, inert or fungistatic).

In contrast, the control (P1) and the 20% lime composition (P6) were placed in Class 1/Category 1 (microscopic fungal growth with minimal contamination).



a) b) c) d)

Figure 10. Representative images of protective and surface treatment solutions and their coating appearance on a white cementitious substrate

Therefore, the efficacy of solution S2 was clearly influenced by the nature of the clay substrate to which it was applied. The treatment of clay surfaces with solution S3, in which rabbit skin glue was used exclusively as the dispersion matrix, yielded the poorest results in terms of protection and coating durability after seven days of exposure in a contaminated environment (Figure 13). Exposure to moisture from mould growth compromised the integrity of the rabbit skin glue matrix, leading to the softening, dissolution, and absorption of the coating into the clay substrate. Consequently, only minimal traces of glycerine-based cosmetic products remained on the surface, appearing as dispersed lipstick residues. Additionally, a mucilaginous layer formed on the clay surface, with a more distinct and continuous film observed in compositions with higher lime content. This suggests that the mucilaginous film, likely containing traces of glycerine and rabbit skin glue, was less readily absorbed into the clay matrix as the lime content increased.

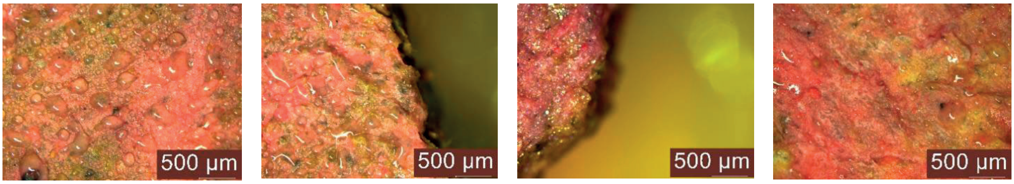
The analysis revealed the loss of inhibition halo formation, surface hydrophobicity, and visible moisture, suggesting its absorption into the system, which contributed to the dissolution of the rabbit skin glue coating and the formation of a mucilaginous film.

Clear signs of mould proliferation were observed, particularly in specimens with lower lime content, such as the control (P1) and sample P6. Quantitative evaluation based on fungal growth assessment criteria and product performance indicators confirmed the ineffectiveness of solution S3, with specimens classified as Class 1 (microscopic fungal growth) and Category 1 (minimal contamination allowing limited growth).

The final evaluation of the clay specimens treated with solutions S1-S3, conducted 33 days after exposure in a contaminated environment, demonstrated good resistance in the specimens treated with the beeswax-based solution (S1) (Figure 14). No fungal growth was observed on the specimens, and the coating maintained its hydrophobic properties. Although the inhibition halo previously observed at shorter exposure durations was no longer present, microscopic evaluation of the specimen surfaces confirmed their classification as: Class 0 (no signs of fungal growth under microscopic examination), Category 0 (the material does not serve as a nutrient medium for microorganisms – it is inert or fungistatic).

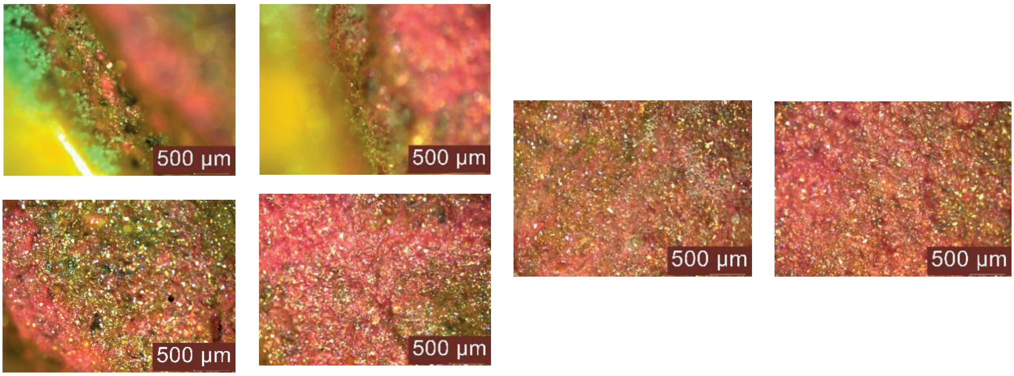
As rabbit skin glue was incorporated into the dispersion matrix (S2 and S3), the coatings on the clay specimens demonstrated diminished resistance to the contaminated environment (Figures 15 and 16). In the case of treatment solution S2, a decline in hydrophobicity was observed, accompanied by distinct indications of mould growth on the control specimen (P1) and the low-lime-content clay specimen (P6). Furthermore, the presence of fungal growth was detected in the cracks that formed on the surfaces of the high-lime-content specimens (P7 and P8). Utilising the product performance estimation criteria outlined in Tables 3 and 4, the treatment of clay surfaces with solution S2 led to the classification of the high-lime-content compositions (P7 and P8) as: Class 1 (Growth invisible to the naked eye but clearly visible under a microscope), Category 1 (The material contains very few nutrients or is minimally contaminated, allowing only very limited fungal growth). In contrast, the control composition (P1) and the composition with 20% lime content (P6) exhibited behaviour classified as Class 2 (visible growth covering up to 25% of the tested surface) and Category 2 (the material does not resist microbial attacks; it contains nutrients that support microorganism growth).

For treatment solution S3 (100% rabbit skin glue dispersion matrix), microscopic examination confirmed significant mould growth on all clay surfaces, resulting in a loss of colour and a transition to a mucilaginous state. The classification results are outlined below: P1: Class 3/Category 2 (Visible growth covering up to 50% of the tested surface; material supports microbial growth); P6-P7: Class 2/Category 2 (Visible growth covering up to 25% of the tested surface; material allows microorganism development); P8 (highest lime content): Class 1/Category 1 (Growth invisible to the naked eye but clearly visible under a microscope; very limited fungal development). As illustrated in Table 6, a comprehensive overview of the temporal progression of quantification indicators, contingent on the nature and implementation of treatment, was provided. It is evident that surface treatment resulted in an enhancement of resistance within the contaminated environment, with an increase ranging from 1 to 3 classes and 1 to 2 categories, depending on the treatment modality and the clay composition. This is particularly influenced by the presence and proportion of lime in the mixture.



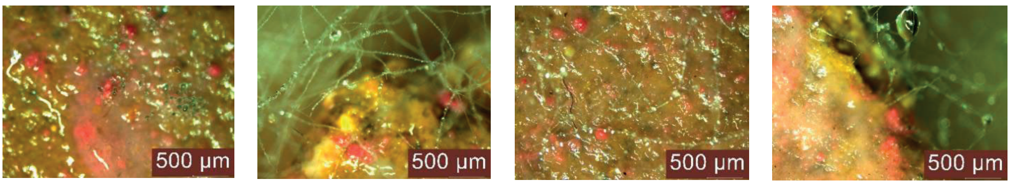
a) P1 b) P6 c) P7 d) P8

Figure 11. Optical microscopy images depicting the performance of clay composites treated with solution S1 in a mould-contaminated environment following 7 days of exposure



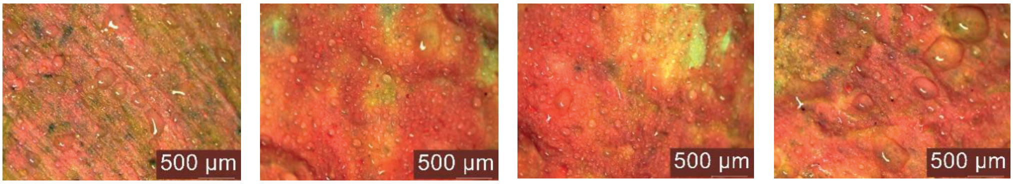
a) P1 b) P6 c) P7 d) P8

Figure 12. Optical microscopy images depicting the performance of clay composites treated with solution S2 in a mould-contaminated environment following 7 days of exposure



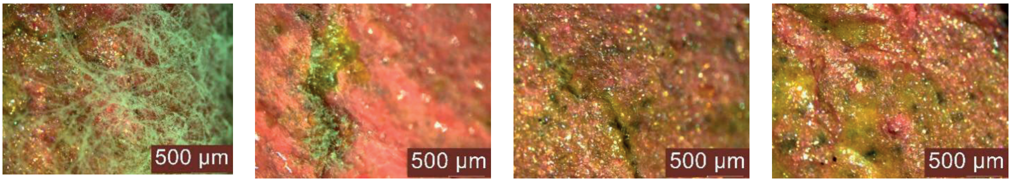
a) P1 b) P6 c) P7 d) P8

Figure 13. Optical microscopy images depicting the performance of clay composites treated with solution S3 in a mould-contaminated environment following 7 days of exposure



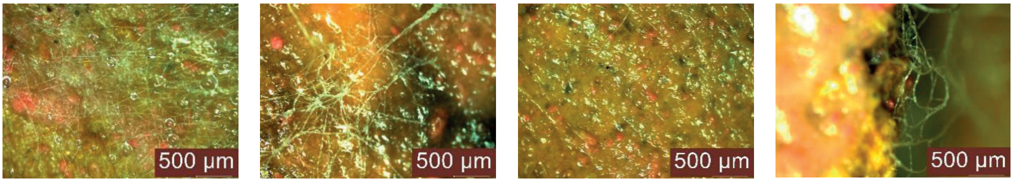
a) P1 b) P6 c) P7 d) P8

Figure 14. Optical microscopy images depicting the performance of clay composites treated with solution S1 in a mould-contaminated environment (33 days of exposure)



a) P1 b) P6 c) P7 d) P8

Figure 15. Optical microscopy images depicting the performance of clay composites treated with solution S2 in a mould-contaminated environment (33 days of exposure).



a) P1 b) P6 c) P7 d) P8

Figure 16. Optical microscopy images depicting the performance of clay composites treated with solution S3 in a mould-contaminated environment (33 days of exposure)

Table 6. The evolution of quantification indicators over time, with a focus on the impact of clay composition, the existence of treatment, and the nature of the treatment itself

Sample code - Treatment		P1				P6				P7				P8			
Exposure time	Quantification indicator	P1	P1-S1	P1-S2	P1-S3	P6	P6-S1	P6-S2	P6-S3	P7	P7-S1	P7-S2	P7-S3	P8	P8-S1	P8-S2	P8-S3
7 days	Class	1	0	1	1	1	0	1	1	1	0	0	1	1	0	0	1
	Category	1	0	1	1	1	0	1	1	1	0	0	1	1	0	0	1
33 days	Class	3	0	2	3	2	0	2	2	2	0	1	2	1	0	1	1
	Category	2	0	2	2	1	0	2	2	1	0	1	2	1	0	1	1

Although equal amounts of expired non-food household cosmetic-pharmaceutical products were used in all three prepared solutions - some selected for their fat content, others for their antifungal properties, and some only as colour pigments - only the beeswax-based solution proved fully effective. This solution imparted hydrophobic properties to the clay surface and utilised the antifungal effect of the non-food household additives, as evidenced by the formation of an inhibition halo around the specimen in the test system.

The beeswax-based surface treatment resulted in more favourable classifications in both fungal growth evaluation and product performance estimation compared to untreated surfaces. In contrast, the rabbit skin glue solution reduced the antifungal efficiency of the non-food household products.

The addition of rabbit skin glue to the mixture, even when combined with beeswax, resulted in only modest outcomes in terms of clay surface protection. The nature of the clay substrate also influenced the outcomes, with a higher lime content in the clay composition contributing to slightly improved outcomes.

CONCLUSIONS

Vernacular constructions, which frequently comprise unfired clay walls, offer a number of advantages, including enhanced indoor air quality, minimal environmental impact, and a beneficial role in preserving and promoting local identity. However, these structures are not without vulnerability; a primary concern pertains to their diminished resilience to Vernacular constructions, which frequently comprise unfired clay walls, offer a number of advantages, including enhanced indoor air quality, minimal environmental impact, and a beneficial role in preserving and promoting local identity. However, these structures are not without vulnerability; a primary concern pertains to their diminished resilience to biological and environmental factors. The objective of this experimental research was to investigate the feasibility of repurposing non-food household waste and minimally processed products through the development of surface treatment solutions aimed at enhancing the durability of unfired clay-based wall surfaces against mould growth.

The experimental research yielded the following conclusions:

- The composition and ratio of raw materials utilised in clay-based composites significantly impact the physical and mechanical performance, resistance, and durability. Among the 12 analysed clay compositions, three were identified as having the most suitable clay-to-lime ratio for the intended application: P6, P7, and P8, which are characterized by the following ranges of raw materials: 30-40% clay, 20-30% lime, and 40% sand.
- With regard to resistance to mould growth (*Penicillium notatum* and *Aspergillus niger*), it was found that all clay compositions ultimately permitted the development of one or both types of mould on their surface. The initial signs of mould appeared earliest (3 days after contamination) in compositions with lower lime content, thereby confirming the beneficial and sanitizing effect of lime.
- The efficacy of surface treatment in enhancing the resistance of clay surfaces to contamination is dependent on the nature of the treatment solution.
- The enhancement of clay surfaces through surface treatment has been shown to occur within a range of 1 to 3 classes and 1 to 2 categories. The most effective performance was observed in the clay composite with the highest lime content (P8), treated with the solution using a 100% liquid beeswax dispersion matrix (S1). The replacement of beeswax with rabbit skin glue (S2) led to a reduction in the antifungal efficacy of the surface treatment for all clay surfaces that were tested, including the composition with the highest lime content (P8). Furthermore, when beeswax was entirely substituted with rabbit skin glue (S3), this decline in antifungal protection persisted.
- Rabbit skin glue (S3) functions as a dispersion matrix, yet it does not yield a substantial beneficial effect in terms of enhancing mould resistance on clay surfaces. In comparison, liquid beeswax has been demonstrated to be a significantly more effective dispersion matrix.

In conclusion, based on the findings presented, new research hypotheses can be formulated, focusing on the use of liquid beeswax as a dispersion matrix while exploring and identifying new expired non-food household products that could be repurposed to develop surface treatment solutions for unfired clay surfaces in construction, with the aim of enhancing their resistance and durability.

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