USE OF OILS FOR THE PROTECTION OF CLAY MORTARS

Aspasia Karozou*
Laboratory of Building Materials, School of Civil Engineering
Aristotle University of Thessaloniki, Thessaloniki, Greece

Maria Stefanidou
Laboratory of Building Materials, School of Civil Engineering
Aristotle University of Thessaloniki, Thessaloniki, Greece

Keywords: clay mortars, oils, surface protection

1. Introduction

Nowadays, promoting sustainable construction materials such as earthen mortars is a significant matter in terms of resource scarcity, energy efficiency, pollution and economy. Since the building sector accounts for around 40% of the world's energy use, according to the World Business Council for Sustainable Development (WBCSD), the benefits offered by the use of earth in construction are significant. A material of low carbon footprint, earth is recyclable, inexpensive and easy to use; it has, moreover, been used for generations and has led to remarkable results. Today, it is once more in the spotlight. However, despite the beneficial aspects, earthen materials present several weaknesses that need to be eliminated in order to become commercial. The lack of specific regulations in most parts of the world, the low response to seismic reaction and its great vulnerability to water intake are the main reasons that earthen construction is still one step behind the more popular industrial materials such as cement [1]. Hence the need to find new ways of protecting earthen structures from damaging factors, so as to promote their use in modern construction. Furthermore, as far as cultural heritage is concerned, the majority of monumental structures worldwide consist of earthen materials that need to be protected and restored. Innovative techniques that are in agreement with the principles of restoration, while providing low environmental impact are considered a necessity.

Earth construction is the oldest technique known to man. More than 9,000 years ago adobes were used to create magnificent structures around the Mediterranean basin, Asia and the ancient known world. Various examples of building structures based on earthen materials can be found even in Central America. The city of Chanchán in Peru (around 850 AD) and the village of Taos in New Mexico (1000–1500 AD) are two examples that indicate the knowledge of earthen construction techniques. The most common techniques known for earthen construction are wattle and daub, cob, rammed earth and adobes. In the wattle and daub technique a woven lattice of wooden strips called wattle is daubed with a viscous material usually made of some combination of

* Corresponding author: akarozou@gmail.com

wet soil, clay, sand, animal dung and straw. It was used for almost 6000 years [2]. Rammed earth is a method of creating a compacted wall from the compression of a soil mixture, while in the cob technique masses of clay mixed with sand and straw are formed manually and are placed one above the other, thus forming monolithic structures with high stability. Lastly, adobes refer to the well-known sun-dried types of brick [3].

Numerous methods exist for the enhancement and protection of clay-based materials. Mankind has for centuries taken into consideration the protection of earthen structures by adopting different strategies according to their surroundings. Such strategies involved orientation of the building in accordance with rain direction and the use of protective coatings and additives [4]. The use of additives can modify certain aspects so as to manage shrinkage, mechanical characteristics and to obtain waterproofing capabilities. There are various categories of additives that can be used, such as mineral additives (sand, pozzolan and ashes), synthetic additives (Portland cement, hydraulic lime, hydrated lime, gypsum, soap bitumen), vegetable additives (fibers, oils, fats, tannins, latexes and molasses) and animal additives (fibers, excrements, urine, blood, casein, animal glues and oil-fats) [5]. The use of coatings for surface protection is also common, in order to offer permeability, durability and water resistance to sensitive earthen materials. Water resistance is achieved through the use of polyvinyl alcohol, ethyl silicates, epoxy and acrylic resins [6]. Although alkoxysilanes and alkoxypolysiloxanes were commonly used as treatment agents [7-10], recent studies have tried to exploit natural resins such as linseed oil in order to be more compatible with earthen materials [11-13]. Moreover, recent studies refer to increasing hydrophobicity through the use of vegetable oils and spent cooking oils inside the mass of concrete and lime respectively [14,15]. Nevertheless, most of these projects deal with cement or lime-based mortars and not clay-based ones.

The aim of this study is to find new environmental friendly techniques in order to preserve and protect earthen materials from deterioration and thus enhance water resistance. For this purpose, various oils, both natural and recycled, were applied on the surface of sample mortars, so as to increase the hydrophobicity of the specific clay produced in the laboratory that was used. The oils applied for the surface protection of the mortars were hemp oil, borage oil (natural) and recycled spent cooking oil, elin (provided by the company Elin and used as a code name for the oil). Besides water repellent capabilities, breathability is also an important aspect that enables the water vapor to be transported from the inside to the outside of the material's mass.

2. Materials and techniques

For the purposes of this study a specific clay mortar was manufactured in the laboratory and the various oils were applied as agents for surface treatment. The specimens created were of a rectangular shape measuring 4 × 4 × 5 cm and tiles with a dimension of 20 × 20 × 3 cm. The clay used comes from the area of Crete and is rich in lime and in addition to aluminum compounds contains quartz, calcite ($CaCO_3$) and gypsum ($CaSO_4$) in small percentages. The clay was sieved to obtain a grain diameter of less than 0.5 mm. The sand used for the mortars was river sand of silica composition, similar in color to that of the clay and with a grain size between 0-4mm. The mixture proportions of the mortar by mass were 1 (clay):2.5 (sand ratio) while the water: binder ratio was equal to 0.6 in order to achieve the desired workability of 15±1cm (tested by flow table). Surface treatment was conducted by brush and the main goal was to form a waterproof surface that allowed the materials to breathe. The procedure for application of the oil for all specimens was as follows:

- First application by brush, once in each direction (right to left and left to right).
- After two hours, the remaining proportion of oil that was not absorbed was wiped off and left in laboratory conditions of 25±1°C
- After 24 hours a second layer of oil was applied twice in each direction
- The specimens were left to dry in laboratory conditions for 15 days after the application of the oils.

All tests were conducted after the period of 15 days. The oils used were the following:

Natural oils:

- Borage oil (indicated as B)
- Hemp oil (indicated as H)

Recycled cooking oil:

- Elin oil - Fatty Acid Methyl Esters (indicated as E)

Hemp oil was chosen as a treatment agent since it is widely used for the protection of wooden structures, while offering waterproof capabilities and weathering resistance; it contains, moreover very low levels of toxic VOCs (volatile organic compounds), making it an excellent alternative to synthetic and petroleum-based polymer coatings [16]. Borage oil, on the other hand, is widely used in medicine due to the many favorable properties it possesses, mostly to treat skin disorders and rheumatoid arthritis [17,18]. Since it is native to the Mediterranean region, it was decided that an experimental study on its properties in construction as a coating would be of interest. Furthermore, in order to exploit industrial waste oils, recycled cooking oil (fatty acid methyl esters) was also chosen as a treatment agent and was provided by a Greek company (Elin). The main purpose of this study was to determine ways of protecting against water penetration. For this reason, the following tests were performed:

- Capillary absorption according to EN1015-18 & drying test
- Porosity according to RILEM CPC11.3
- Absorption of water drop test (heuristic test)
- Karsten tube penetration test (CSTLI7500-TQC)
- Surface roughness measurements using Mitutoyo profilometer and recording the roughness parameter (Ra) values - which are used widely as a one-dimensional roughness parameter and concerns the arithmetic average of the absolute values
- Water vapor permeability test (EN 1015-19:1999)
- Optical observation of color change using Munsell charts
- Stereoscopic observation using microscope LEICA WILD M10
- Microscopic examination by SEM (JEOL840A JSM)

For the purpose of comparison reference composition samples were left untreated (recorded as A).

3. Experimental results

In order to study the behavior of the samples for water penetration, various tests were performed, such as a capillary absorption test, a porosity test and the heuristic test of water droplet absorption test. The heuristic test measures the time it takes for a single droplet to be absorbed by the surface of the specimens. These experiments record the behavior of the samples when they come into contact with water. Moreover, roughness measurements were recorded for all the surfaces of each sample so as to calculate the average Ra value. Ra is a roughness parameter deriving from the arithmetic average value of the filtered roughness profile determined from deviations about the center line within the evaluation length. Table 1 reports the results of the physical properties of the samples and those of the tests mentioned previously.

Table 1. Results of the physical properties and tests.

	Water absorption (%)	**Porosity (%)**	**Capillary Coefficient ($g/cm^2 *min^{1/2}$)**	**T (min)**	**Ra (mm)**	**Water penetration value (ml/min)**
A	7.04	13.71	0.43	0.58	0.111599	0.241
B	4.34	8.87	0.28	19.0	0.233832	0.128
E	4.21	8.46	0.29	2.50	0.227467	0.100
H	5.20	10.55	0.32	2.00	0.108695	0.155

It is clear that the values of the treated samples B, E and H, in the case of water absorption, porosity and capillary coefficient differ in comparison with the untreated samples. The protective role of the oils applied is obvious since water absorption and porosity are lower in each case. It can be noted that the samples treated with hemp oil present values close to the reference samples A. Moreover, roughness values are in agreement with the time measured for the absorption of one water droplet, indicating the hydrophobic nature of the samples treated with borage oil. However, the results obtained for time absorption indicate the low hydrophobic nature of both the E and H specimens. The capillary absorption test results are recorded in Figure1.

The surface of reference sample A shortly after contact with the water was destroyed. The capillary coefficient calculated after conducting this test, indicates the low water uptake for all three treatments, while for the reference sample A, the capillary coefficient was calculated using the last measurement taken. Besides damage of the surface of sample A, the drying procedure was conducted as a reverse capillary test for all samples, naturally without the presence of water (Figure 2). Duration of the test was until stabilization of the samples' mass. Both A and H samples possess the longest drying period, with A sample presenting a massive weight loss. This fact is explained by the large water uptake and the unaffected surface that allowed the water to move more easily from the inside out. The samples treated with borage and Elin oil present a slower rate of water evaporation, butt indicate good drying behavior by allowing the sample to breathe. The samples treated with hemp oil show the highest water intake, yet the drying procedure indicates a slower rate of water loss compared to the other treated samples.

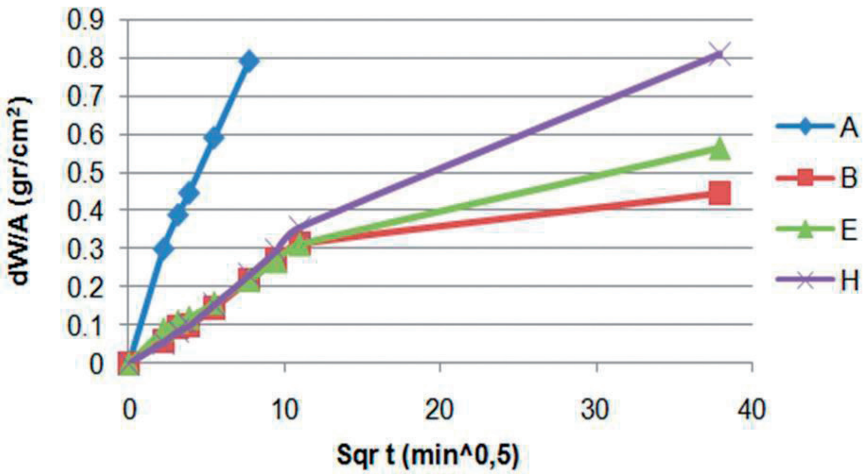

Figure 1. Capillary absorption of the specimens.

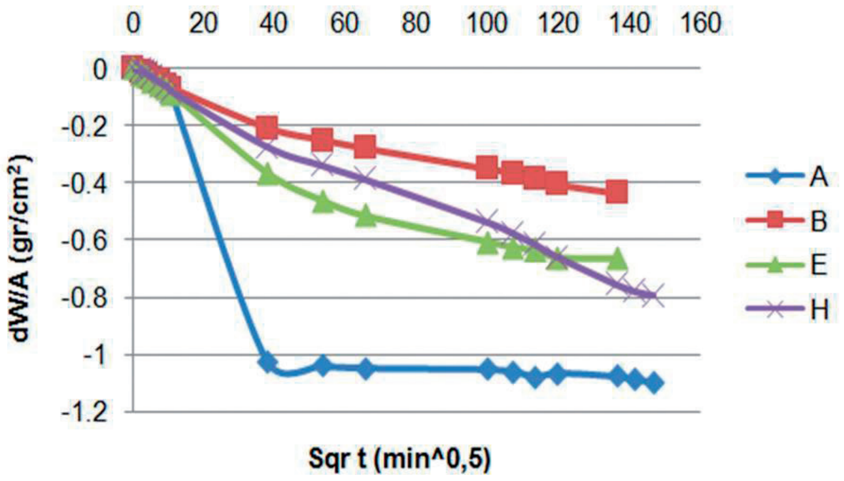

Figure 2. Drying test of the specimens.

Another way to test water absorption on a horizontal surface is through the Karsten tube penetration test. Karsten tubes were applied horizontally onto the surface of the samples using plasticine and the tubes were filled with 2 ml water. The test lasted ten (10) minutes in total for each sample, while every 15 seconds the water absorbed was recorded. The results are presented in Figure 3, while the water penetration value for each sample is indicated in Table 1. Again, the results show the hydrophilic nature of the untreated samples and the protective nature of the treated ones. The hemp samples once more present a similar behavior to the untreated specimens compared to the other samples B and E. However, B and E samples show a slower rate of uptake of 0.128 and 0.1 ml/min respectively.

The breathability of the samples is of the utmost importance since moisture entrapment is a catastrophic factor for porous materials. If appropriately treated water can be prevented from reaching the upper surface and evaporating [19]. For this reason, the water vapor test was conducted on both treated and untreated tiles. Each tile was applied on top of a container half filled with water using silicon and was then preserved in a chamber with steady conditions (20 °C, 60%RH). The weight loss of the tile-container system indicates the passage of water vapor through the tile and into the atmosphere. The system was measured for four months and the results are presented in Figure 4. As expected, the weight loss of sample A was the highest compared to the treated samples at a value of 4.64%. Despite the previous results that indicate the similar behavior of hemp treated samples to the reference ones, it is noted that the H specimens show the lowest water loss of the tile-container system with a value of 2.10%. The values of both samples treated with borage and Elin are very close with 2.65% and 2.68% weight loss respectively.

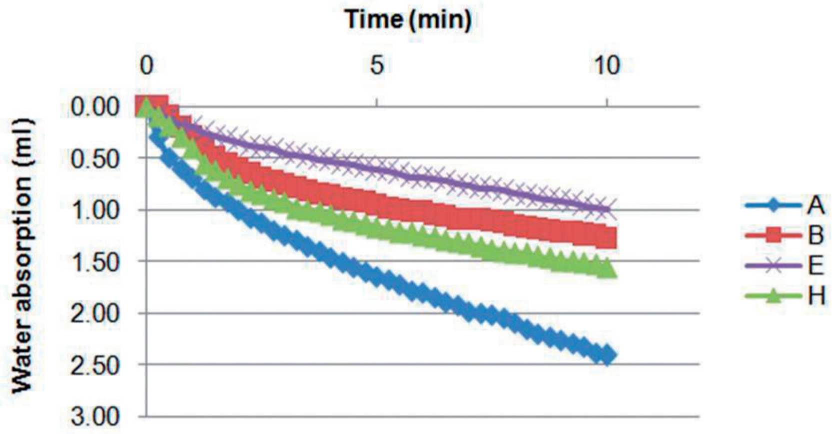

Figure 3. Karsten tube test results.

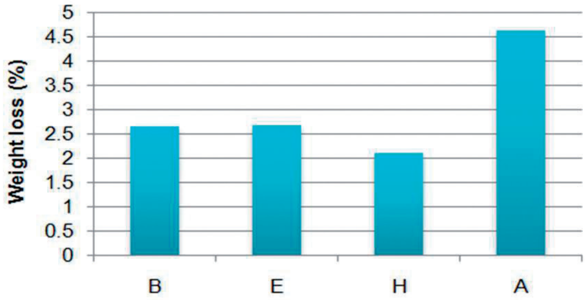

Figure 4. Weight loss in the tile-container system.

The water vapor permeance Λ was calculated from the data collected so far. Again, it can be concluded that the untreated samples present the highest water vapor flux in comparison to the treated samples, according to the high value of the Λ factor. From Table 2 it can also be noted that H samples prevent the passage of water vapor compared to samples B and E that again present similar behavior.

Table 2. Results of water vapor permeability test.

Water vapor permeance Λ (kg/m^2 *s*Pa)			
A	B	E	H
1.00118E-11	5.1293E-12	5.89365E-12	4.49E-12

All samples were examined to detect alterations on their surfaces, in terms of color change, cracking and roughness. In order to determine alterations in color after treatment, Munsell charts were used. The results are listed in Table 3 below. Both B and H samples differ from the color of the untreated samples A, in terms of value and chroma, and developed a darker surface. On the contrary, the E samples showed small alterations compared to the reference samples, with a slight differentiation in hue.

Alterations in the color of the samples were also detected through stereoscopic observation (Figure 5). Moreover, shrinkage cracks were evident on the surface of all specimens, with H samples showing a smoother surface. This element is in line with the results of the roughness measurements, since H samples presented the lowest Ra value of 0.108 mm. Samples treated with elin oil presented many shrinkage cracks and a rougher surface. The rougher surface spotted through microscopic observation comes in agreement with the Ra value of 0.227 mm, indicating the roughness of samples treated with elin oil. Despite the highest roughness value in the samples treated with borage oil (Ra=0.234), the microscopic observation reveals a relatively smooth surface in accordance with E and A samples. Macroscopic observation showed that in all cases the oils were able to penetrate up to a depth of 2.0 mm from the surface, with a maximum penetration depth of 5.32 mm in the samples treated with borage oil (Figure 6).

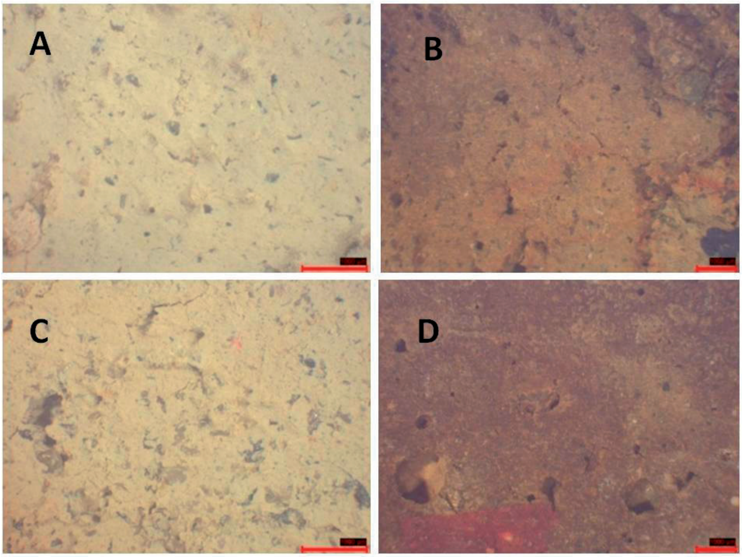

Figure 5. Stereoscopic images of the various specimens: A) Untreated sample; B) sample treated with borage oil; C) sample treated with Elin oil; D) sample treated with hemp oil (scale-1000μm).

Figure 6. Area of penetration of sample treated with hemp oil.

Table 3. Optical observation of color changes using Munsell charts.

	Hue	Value	Chroma	Name
A	2.5Y	8	2	pale yellow
B	2.5Y	6	3	dark yellowish brown
E	5Y	8	2	pale yellow
H	2.5Y	7	3	pale yellow

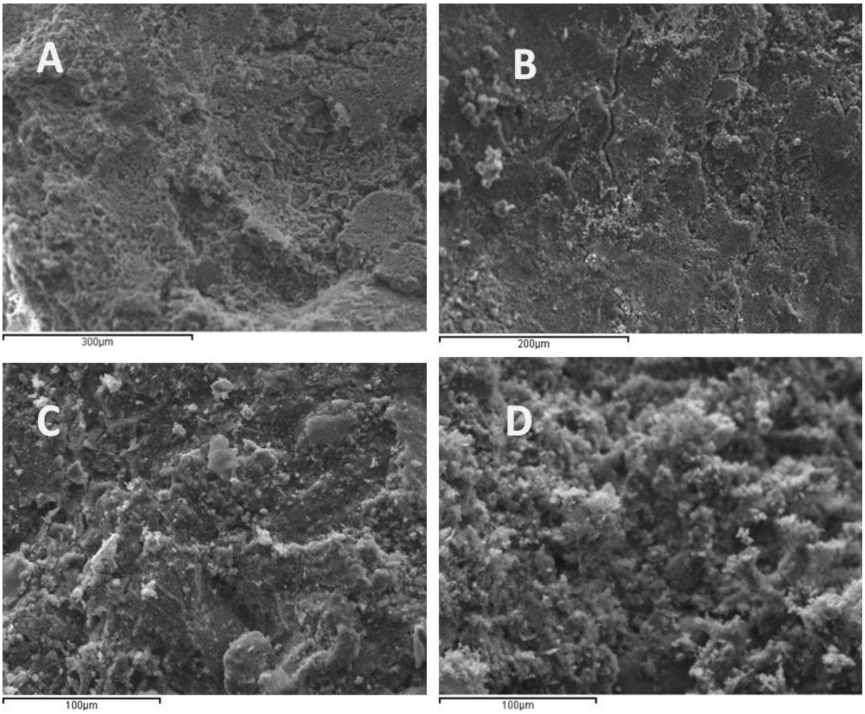

Figure 7. SEM images of the various specimens A) Untreated sample, B) sample treated with borage oil, C) sample treated with Elin oil, D) sample treated with hemp oil (scale-300μm for A, scale-200μm for B, scale-100μm for C and D).

Similar to the stereoscopic observation are the results conducted using a scanning electron microscope (SEM). The surface of the untreated sample A is smoother than the treated sample's surface, while the borage treated specimens present a much more stable and consistent surface (Figure 7). Micro roughness is observed on B, H and E samples which can justify the hydrophobic properties in accordance with the relevant literature [9].

4. Conclusions

Using oils for the protection of clay-based materials has given some promising results. The clay structure becomes more resistant to water penetration in each test conducted using the various oils. The decreased tendency of the treated specimens to absorb water and the good drying behavior of all the samples make the oil coatings used suitable for surface protection. After concluding the tests, as expected, the untreated samples presented the highest values in water absorption and porosity, while the time of absorption for the water droplet was high. The high values of capillary coefficient and water penetration compared to the treated samples indicate the protective role of the coatings used.

It was noted that the B and E samples presented similar behavior in almost every test conducted, while the H samples were closer to the behavior of the untreated samples. The high values of Ra and time of absorption of the water droplet indicate the hydrophobic nature of borage oil, while the alteration in color is considered to be a negative aspect. Despite the high level of water absorption and porosity of the hemp treated samples, they had the lowest weight loss during the water vapor permeability test, a fact that indicates prevention of the water vapor flux. The smooth compact surface of the hemp samples spotted through microscopic observation, are in line with the low Ra value measured. In comparison to the B and E samples, the H specimens show a good drying behavior, while a negative aspect is the significant color alteration of the surface.

As mentioned above, treated E samples shared similar characteristics with B samples and the results indicate the hydrophobic nature of the coating applied. However, despite having the lowest water penetration value and relatively low values of porosity, water absorption and capillary coefficient, the fast absorption of the water droplet and the cracks developed on the surface diminish its protective role. Nevertheless, the positive aspects of the Elin oil application were that it was fast drying, had good water vapor permeability behavior and produced an insignificant color alteration of the samples. Also, the low water penetration value presents resistance against horizontal water penetration.

In conclusion, there are many positive aspects in each of the coatings applied. However, further investigation and perhaps modification of the oils used is proposed. The durability of the treated samples proved greater compared to the untreated ones, making the protective role of the agents obvious. The use of such materials in construction is promising in terms of sustainability and their use is thus to be encouraged.

References

[1] Sameh, Sherin H. (2014) Promoting earth architecture as a sustainable construction technique in Egypt, Journal of Cleaner Production 65, pp. 362-373

[2] Pacheco-Torgal, F., Jalali, S. (2012) Earth construction: Lessons from the past for future eco-efficient construction, Construction and Building Materials 29, pp. 512–519

[3] Morton, T. (2008) Earth Masonry – Design and Construction Guidelines, IHS BRE Press, Watford

[4] Aubert, J.E., Fabbri, A., Morel, J.C., Maillard, P. (2013) An earth block with a

compressive strength higher than 45 MPa!, Construction and Building Materials 47, pp. 366–369
[5] Bahobail, M. A. (January 2012) The Mud Additives and Their Effect on Thermal Conductivity of Adobe Bricks, Journal of Engineering Sciences, Assiut University, Vol. 40, No 1, pp. 21-34
[6] Elert, K., Sebastián, E., Valverde, I., Rodriguez-Navarro, C. (2008) Alkaline treatment of clay minerals from the Alhambra Formation: Implications for the conservation of earthen architecture, Applied Clay Science 39, pp. 122–132
[7] Stefanidou, M., Karozou, A. (2016) Testing the effectiveness of protective coatings on traditional bricks, Construction and Building Materials 111, pp. 482–487
[8] Dhakal, H.N., Bennett, N. (2012) Titanium dioxide and zinc oxide nano-particulate enhanced oil-in-water (O/W) facade emulsions for improved masonry thermal insulation and protection, Energy Build. 52, pp. 86–92
[9] Manoudis, P., Karapanagiotis, I., Tsakalof, A., Zuburtikudis, I., Matziaris, K., Panayiotou, C. (2008) Surface properties of superhydrophobic coatings for stone protection, Proceedings of the International Symposium, Stone Consolidation in Cultural Heritage, Research and Practice, Laboratorio Nacional de Engenharia Civil, Lisbon, 6–7 May, pp. 193–201
[10] Falchi, L., Müller, U., Fontana, P., Izzo, F.C., Zendri, E. (2013) Influence and effectiveness of water-repellent admixtures on pozzolana–lime mortars for restoration application (Construction and Building Materials 49, pp. 272–280
[11] Nunes, C., Slížková, Z. (2014) Hydrophobic lime based mortars with linseed oil: Characterization anddurability assessment, Cement and Concrete Research 61–62, pp. 28–39
[12] Stefanidou, M., Karozou, A., Kapsimani, M. (2016) Protecting adobes by increasing their hydrophobicity using alternative methods, 5th International Conference YOCOCU, 21st–23rd September, Madrid, Spain
[13] Nunes, C., Slízková, Z. (2016) Freezing and thawing resistance of aerial lime mortar with metakaolin and a traditional water-repellent admixture, Construction and Building Materials 114, pp. 896–905
[14] Justnes, H., Østnor, T.A, Barnils Vila, N. (2014) Vegetable oils as water repellents for mortars, Available at: https://www.researchgate.net/publication/264849005
[15] Pahlavan, P., Manzia, St., Rodriguez-Estrada, M.T., Bignozzi, M.C. (2017) Valorization of spent cooking oils in hydrophobic waste-based lime mortars for restorative rendering applications, Construction and Building Materials 146, pp. 199–209
[16] Hemp Industries Association https://www.thehia.org/Building-Materials
[17] Takwale, A., Tan, E., Agarwal, S., Barclay, G., Ahmed, I., Hotchkiss, K., Thompson, J. R., Chapman, T., Berth-Jones, J. (2003), Efficacy and tolerability of borage oil in adults and children with atopic eczema: randomised, double blind, placebo controlled, parallel group trial, BMJ 327:1385
[18] Belch, J. JF, Hill, A. (2000), Evening primrose oil and borage oil in rheumatologic conditions, The American Journal of Clinical Nutrition, 71:1, pp. 352s–356s, https://doi.org/10.1093/ajcn/71.1.352s
[19] Cultrone, G., Madkour, F. (2013) Evaluation of the effectiveness of treatment products in improving the quality of ceramics used in new and historical buildings, Journal of Cultural Heritage 14(4), pp. 304–310.

Biographical notes

Stefanidou Maria, Associate Professor is the director of the Laboratory of Building Materials in the Civil Engineering department of the Aristotle University of Thessaloniki. Her interests are the study of traditional mortars and the composition of compatible ones as well as the improvement of their properties by sustainable methods and by using nanotechnology. Her research interests include producing and testing artificial stones, bricks, mortars and grouts for restoration purposes as well as experimenting with innovative techniques for building materials such as the use of phase change materials, self-healing renders and nano-modified coatings.

Karozou Aspasia, M.Sc. is currently working as a PhD student at the laboratory of building materials in the civil engineering department of the Aristotle University of Thessaloniki. She holds a Diploma in civil engineering and has a Master's degree in conservation and restoration of monuments. Her main research interest focuses on finding innovative ways to protect and enhance clay for use in restoration projects.

Summary

In this study an effort has been made to find innovative and sustainable ways of increasing the hydrophobicity of clay-based mortars used in restoration by applying natural oils on the surface of selected samples. The concept was to find a new way of protecting the surface of these materials, but also to promote the idea of sustainability by using natural and recycled cooking oils. Thus, the oils applied for the surface protection of the mortars were hemp oil, borage oil (natural) and recycled spent cooking oil. The tests conducted on the treated specimens were, a capillary absorption test, drying test, porosity, Karsten tube test, stereoscopic observation, color alteration using Munsell charts, water vapor test and absorption of water droplet test. The results indicate the protective role of all the oils, since they reduce porosity and water uptake. Borage and hemp oil tend to alter the color of the specimens, but offer increased hydrophobicity, while the latter allowed the faster drying of the samples. The water vapor test indicates again the low level of water loss from the surface of the treated specimens, while for the specific test a sample with an admixture of recycled cooking oil inside the clay mass was created.

Riassunto

Lo studio riguarda modi innovativi e sostenibili per aumentare l'idrofobicità delle malte a base di argilla utilizzate nel restauro, applicando oli naturali sulla superficie di campioni selezionati. Il fine è trovare un nuovo modo per proteggere la superficie di questi materiali, ma anche di promuovere l'idea di sostenibilità utilizzando oli da cucina naturali e riciclati. Pertanto, gli oli applicati per la protezione superficiale delle malte erano olio di canapa, olio di borragine (naturale) e olio di cottura esausto riciclato. I test condotti sui campioni trattati sono stati: test di assorbimento capillare, test di essiccazione, porosità, test del tubo di Karsten, osservazione stereoscopica, alterazione del colore con cartografia Munsell, test del vapore acqueo e assorbimento del test delle gocce d'acqua. I risultati indicano il ruolo protettivo di tutti gli oli poiché riducono la porosità e l'assorbimento di acqua. La borragine e l'olio di canapa tendono ad alterare il colore dei campioni ma offrono una maggiore idrofobicità. L'olio di canapa ha permesso l'essiccazione più rapida dei campioni. Il test del vapore acqueo indica nuovamente

il basso livello di perdita d'acqua dalla superficie dei campioni trattati, mentre per il test specifico è stato creato un campione con una miscela di olio di cottura riciclato all'interno della massa argillosa.