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The Anti-Bacterial Lime Mortar Used in the Restoration of Ancient Limestone Buildings by Adding Heavy Metal Oxide

Dr. Mostafa Abd Elgaleel Hossin

Lecturer, Department of Architecture - OHI – Giza- Egypt

Dr. Taha M.Tiama

Department of Basic Sciences- OHI – Giza- Egypt

Eng. Amr Hussein A. Russlan

Assistant Lecturer, Department of Construction Engineering- OHI – Giza- Egypt

Abstract:

Recently, the awareness of the need for compatible materials to be used to preserve the architectural heritage has increased, which has led to an increase in the revival of lime-based mortar technology and its applications. However, it is known that the preparation process and procedures affecting the final quality of lime slurry are limited, and there is no doubt that there is still debate in the preservation community as to the most suitable material for preservation treatments (for example, slaked lime versus aged lime putty).

It is known to everyone interested in studying the restoration of ancient buildings that some problems are resulting from the growth of microorganisms, for example, bacteria and fungi.

Metal oxide nanoparticles (NPS), sometimes called engineered nanoparticles, have been used to protect building surfaces against biofilm formation for many years, but their lifespan in the cultural heritage world is short. Its first use was in 2010. Subsequently, it has had a wealth of reports in the literature, with Ti, Ag, and zinc oxides being the main heroes. In all surface treatments, NPs can leach into the surrounding environment, resulting in potential environmental toxicity to soil, water, and associated organisms. It is important to us that the testing of the biocidal products used for this is updated, as well as that of a standardized antimicrobial test. Highly efficient to determine their impact on the environmental processes of non-target surrounding organisms and their transformation.

Keywords: Antimicrobials, Heritage buildings, Metakaolin, kaolin, Nanoparticles, lime mortar, engineered nanoparticles, Heba.

1. Introduction:

There are other types of nanoparticles produced by man, known as engineering nanoparticles (ENPs), which are created using different materials such as metal oxides, metals, carbon, polymers, and fats [1]. Next to the potential of these nanoparticles (NPS) to preserve the building Protection has been made to standardize decomposing materials, producing decontamination surfaces, Self-cleaning, surface reinforcement, or lime mortar acting as a biocide for Reduce biological degradation [2-5]. Titanium dioxide (Anatase) has been employed for many years [6, 7] and sometimes it has become the gold standard for self-cleaning surfaces. TiO2 NPs have high stability and photo reactivity, they are also active against a wide spectrum of microorganisms, and most importantly they are cheap. Through the research higher recolonization occurred in the treated areas in areas of high humidity, near the ground. Similarly, spraying of TiO_2 NPs, with or without Ag and Cu NPs, was not able to increase the toughness of limestone (limestone) subjected to artificial accelerated aging [8]. Factors affecting the effectiveness of NPS on building materials include porosity and roughness; When these are high, NP treatment is less likely to be effective [9].

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1-1 Definition and history of structural bonding materials:

A binder is defined as a material which acts as a glue or cement and can be mixed with an aggregate (e.g., sand) and water to form a fresh plaster, render, mortar or concrete. Following setting and hardening, such composite materials play a structural and/or decorative role in building (Lea 1970). Since the advent of sedentism and building technology, which could be traced back to the Pre-Pottery Neolithic A in South-West Asia (e.g., Jordan river valley), ca.11,700 to 10,500 years ago (Finlayson et al. 2011), mankind has used many different binders.

Mud (clay-rich earth) was among the first binders used (Houben and Guillaud 1994) as exemplified by the adobe walls of Jericho (Palestine) dated to 8300 BC (Allen and Tallon 2011) or the earthen structures (rammed earth) in Çatalhöyük (Anatolia, Turkey) dated to 6000-7500 BC (Mellaart 1967). However, the use of this type of binder does not involve pyro technology (i.e., no heat treatment was required). As a result, earthen structures display a low strength and are prone to weathering and damage. With the advent of pyro technology ca. 12000 years ago (Kingery et al. 1988), a new class of binders emerged with superior properties in terms of strength and durability, as well as versatility and applicability. Among them, gypsum and lime have played an outstanding role in building history from the Neolithic until nowadays.

While a mortar is a material resulting of the intimate mixture of sand grains, a binder (lime, cement, etc.) and water, the properties and characteristic of the mortars mainly depend on the nature of the binder component.

With the purpose of modifying and/or improving some of the properties of the mortars, they have been mixed traditionally (together with the basic components), with some different products or additional constituents. These products have evolved along with the time. At the beginning the admixtures were composed of natural substances (blood, egg, fig juice, pig grease, manure, etc.).

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The current admixtures are generally industrial by-products, like fly ashes or blast furnace slags ..., or other more elaborated products, like organic polymers, acrylic resins, epoxy resins, etc.

Lime-Based Mortars for Construction &Architectural Conservation.

In architectural conservation, adherence to original materials in treatments and repair is highly advantageous for the sake of material compatibility as well as authenticity and aesthetics. However, the service life of repairs is also an undeniably critical consideration for practicality and sustainability. This is particularly relevant in the formulation of repair mortars for the bedding and pointing of masonry work.

A conflict exists between the idea of using lime mortars for repair and the insufficient long-term performance associated with these materials. As a result, lime mortars have frequently been gauged with, or entirely replaced with Portland cement in the repair of historic masonry structures.

1-2 The most important conventions and codes for dealing with ancient buildings and restoration methodologies:

The document will focus on modern diagnosis and repair techniques, and how to best apply advancing technology in each widely ranging case of lime deterioration. The document will track the development of lime as a building material, and the deterioration mechanisms that have been proven afflict structures with lime portions. Taking into account the developing theories on historic restoration, we will attempt to use what was learned from past blunders to suggest a common approach for investigation and remediation of structures in every case that will satisfy the standards and guidelines set forth by ICOMOS (International Council on Monuments and Sites.

From (INTERNATIONAL CHARTER FOR THE CONSERVATION AND RESTORATION OF MONUMENTS AND SITES) which announced (THE VENICE CHARTER 1964) (Content of 16 Article).

1-2-1 Article 12.

Replacements of missing parts must integrate harmoniously with the whole, but at the same time must be distinguishable from the original so that restoration does not falsify the artistic or historic evidence. (THE VENICE CHARTER 1964).

1-2-2 Article 13.

Additions cannot be allowed except in so far as they do not detract from the interesting parts of the building, its traditional setting, the balance of its composition and its relationship with its surroundings. (THE VENICE CHARTER 1964)

On the other hand, Records of the General Conference Seventeenth Session Paris, 17 October to 21 November 1972 (United Nations Educational, Scientific and Cultural Organization) & (INTERGOVERNMENTAL COMMITTEE FOR THE PROTECTION OF THE WORLD CULTURAL AND NATURAL HERITAGE).

23. Any work done on the cultural heritage should aim at preserving its traditional appearance, and protecting it from any new construction or remodeling which might impair the relations of mass or color between it and its surroundings (Records of the General Conference Seventeenth Session Paris, 17 October to 21 November 1972).

1-3 History of the use of lime as a binder:
Historical Use of Lime as Binding Material

Archaeological evidence shows that lime was used in the construction of some of the floors and paving of the ruins excavated in Çatalhüyück (Mellaart 1967), dated between 10000 and 5000 BC (Von Landsberg 1992; Kingery et al. 1998). These archaeological findings, along with the remains of 4500 years old lime kilns found in Khafaje, Mesopotamia, confirm that lime was a common building material in the Levant during the Neolithic (Davey 1961).

The Egyptians also used lime as a binder. Some coatings of lime in different pyramids have been dated ca. 4000 BC (Boynton 1980).

However, this is challenged by Lucas and Harris (1962) and Ghorab et al. (1986) who indicate that the Egyptians did not use lime in construction (they used gypsum) until Roman times.

Other ancient civilizations, like India, China and the different cultures of pre-Columbian America (e.g., Mayans and Aztecs) systematically used lime as a building material (Gárate Rojas1994).

The Greek and Roman civilizations discovered that calcination of marly limestones, i.e., with a concentration of alumina silicates (clays) > 10 wt. %, yielded a binding material that hardened underwater (hydraulic setting) and had improved mechanical properties (Malinowski 1981).

Upon calcination of impure limestones, clays dehydroxylate at 400 to 600°C, the resulting silica and alumina combine with CaO formed after the decomposition of CaCO3at 950 to 1250º C, to produce calcium aluminates and silicates (Callebaut et al. 2001). These limes are called natural hydraulic lime. Dicalcium silicate (C2S) is the main phase that reacts with water causing their hydraulic setting, unlike in the case of cement where tricalcium silicate (C3S) is the main hydraulic phase (Callebout etal., 2001). In addition to natural hydraulic lime, the socalled artificial hydraulic limes have been also used. They were discovered by the Phoenicians and perfected by the Greeks and the Romans.

Artificial hydraulic lines were obtained by mixing lime with a pozzolanic material, name taken from the town of Puzzoli, located nearby the Vesuvius in Italy, where a tuff with high hydraulic capacity was extracted. A pozzolanic material contains highly reactive silica and alumina. When combined with Ca (OH)2 in the presence of water generates new products, mainly hydrated calcium silicates and aluminates, with superior binding or cementing properties (Mertens et al. 2009).

The Romans used lime in construction since the last two centuries of the Republic (200-100 BC). In addition to air lime, they routinely used lime mixed with either natural (pozzolana) or artificial (brick powder) pozzolanic materials, thus obtaining the well-known opus cementitious and the cocciopesto described by Vitruvius (30 BC).

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After the fall of the Roman Empire, natural and artificial hydraulic limes, including Roman cement, together with traditional air lime were the most common binders in construction since Byzantine time, though the Middle Ages, Renaissance and Baroque, until the discovery of Portland cement in theearly19th century by Aspdin (Lea 1970).
However, since mid-18thcentury, traditional limes began to be replaced by high performance artificial hydraulic limes, made by researchers such as Smeaton (1791) or Vicat (1837). In 1824 Aspdin patented a process for obtaining a cement which hardened under water (hydraulic setting) and acquired the strength of Portland limestone, one of the most resistant stoned used in British architecture, hence the name "Portland cement" Haswell(1865).
The homogeneity and uniform properties, easy of application and high mechanical strength after a rapid setting explains the immediate success and massive use of Portland cement until present day. However, in recent decades, lime has reemerged as the optimal material for the conservation of historic structures and buildings (Teutonico et al. 1994), replacing cement in such interventions. There are several reasons for the revival of lime (Elert et al. 2002). Compared with cement, lime is more compatible from a mechanical, physical and chemical point of view when applied in historical structures (brickworks and/or stone) (Elert etal. 2002; Lanas and Alvarez 2003; Hansen et al. 2008). For instance, the absence of alkalis and sulfates in lime prevents salt weathering due to the formation of deleterious carbonates and sulfates of Na/Ca or Mg, as can be seen after the application of Portland cement (i.e., Rodríguez-Navarro et al. 1998; Maravelaki-Kalitzaki et al. 2003; Hansen et al. 2008). Crystallization of such salts causes extensive damage in both new construction and historic structures Figure (1)

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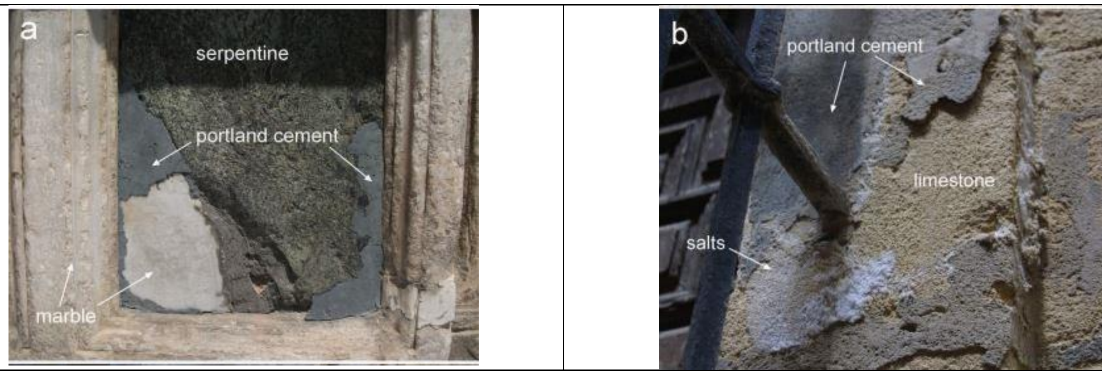

Figure (1). Examples of the high incompatibility between Portland cement and historic structures: a) Portland cement applied on serpentine and marble at the Chancilleria (Granada, Spain); b) salt efflorescence and detachments associated to Portland cement applied on limestone ashlars (Granada Cathedral, Spain).
(Rodríguez-Navarro, 2012)

In the Fig. (2) Use of binders during history (Furlan, V., Bissegger, P. 1975).

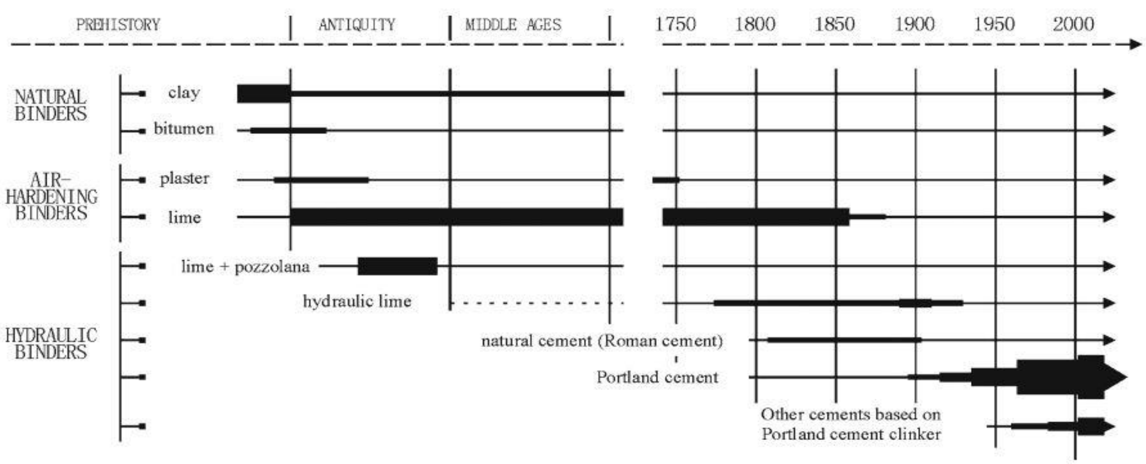

Figure (2): Use of binders during history (Furlan, V., Bissegger, P. 1975)

1-4 Types of archaeological facilities in Egypt in the ancient age:

Homes and other buildings continued to be made from mud brick during the Middle Kingdom; stone was only used for temples and monuments and this was usually limestone, sandstone or, in some cases, granite which required the greatest skill to work in. A little known masterpiece of the Middle Kingdom, long ago lost, was the pyramid complex of Amenemhat III (c. 1860 - 1815 BCE) at the city of Hawara.

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This complex was enormous, featuring twelve great separate courts which faced one another across an expanse of columned halls and interior hallways so intricate that it was called "the labyrinth" by Herodotus. The courts and hallways were further connected by corridors and colonnades and shafts so that a visitor might walk down a familiar hall but take an unfamiliar turn and wind up in a completely different area of the complex than the one they had intended.

Criss-crossing alleys and false doors sealed by stone plugs served to confuse and disorient a visitor to protect the central burial chamber of the pyramid of the king. This chamber is said to have been cut from a single block of granite and to have weighed 110 tons. Herodotus claimed it was more impressive than any of the wonders he had ever seen

Egypt is a country rich in stone and was sometimes even referred to as the "state of stone". Egypt has a great quantity of limestone formation, which the Egyptians called "white stone", because during the Cretaceous period Egypt was covered with seawater. The country is also rich in sandstone, but it was never really used much until the New Kingdom.

Limestone seems to have first been employed in Saqqara, where it is of poor quality but layered in regular, strong formations as much as half a meter thick. This limestone is coarse grained with yellow to greenish gray shading. The layers are separated from each other by thin layers of clay and the coloration may vary according to layer. It could often be quarried very near the building sites, and quarries have been found at Saqqara, Giza, Dahshur and other locations.

In order to quarry this stone, the blocks were marked out with just enough space in between each to allow for a small passageway for the workers to cut the blocks. The workmen would use a number of different tools to cut the blocks, including copper pickaxes and chisels, granite hammers, dolerite and other hard stone tools.

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The finer, white limestone employed in the pyramids and mortuary temples was not as easy to quarry and had to be found further from the building site. One of the man sources for this limestone was the Muqattam hills on the west bank of the Nile near modern Tura and Maasara. This stone laid buried further from the surface, so tunnels had to be dug in order to reach the actual stone quarry. Sometimes these deposits were as deep as fifty meters, and huge caverns had to be built to reach the quarry. Generally, large chunks of stone were removed, and then finely cut into blocks. The blocks were then moved to the building site on large wooden sledges pulled by oxen. The path they took would be prepared with a mud layer from the Nile in order to facilitate the moving.

Pink granite, basalt and alabaster were used much more sparingly. Most of this material was moved from various locations in southern Egypt by barges on the Nile. Pink granite probably most often came from the quarries around Aswan. **Table (1):**

Title	Author	Date	Publisher	Reference Number
Atlas of Ancient Egypt	Baines, John; Malek, Jaromir	1980	Les Livres De France	None Stated
Oxford History of Ancient Egypt.	Shaw, Ian	2000	Oxford University Press	ISBN 0-19-815034-2
The Pyramids, The Mystery, Culture, and Science of Egypt's Great Monuments	Verner, Miroslav	1997	Grove Press	ISBN 0-8021-1703-1
Life of the Ancient Egyptians	Strouhal, Eugen	1992	University of Oklahoma Press	ISBN 0-8061-2475-x

1-5 Ancient Egyptian Building Materials:

A- Wall Plaster:

The painted wall plaster in the photograph below came from the walls of a tomb. We can't be sure what was actually painted on it because it is a very small piece.

The richer and more important the person had been during their life, the larger and more decorative the paintings would be. Sometimes they were used to tell of the person's achievements in their life or offer prayers to the gods.

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B- Mud and Straw Brick:

In ancient Egypt people used mud and straw to make bricks for their houses. The straw was added to the mud to make the brick stronger. They were placed in a mold to give them an oblong shape, which is easier to build with. This material helps to keep the houses cool when it is hot and warm when it is cool.

In some parts of the world people still build their houses with these materials today because they are cheap and easy to find.

C- **Air lime:**

The used hydrated (slacked) air lime (Ca (OH2)) powder was a commercial product supplied by Hamaco is of class EN 459-1 CL70-S satisfying BS EN 459-1. Air lime was used to produce the control mortar and modified mixtures. Two different types of Kaolinite and Metakaolin were used for the proposed modification in addition to Hib (Heeba in Arabic), Burned Hib, and fine crushed clay bricks (Homra in Arabic). Metakaolin and Burned Heeba were prepared by heating Kaolinite and Heeba on the oven to reach 850°C, and beyond 900°C

In the following paragraphs, sources of the lime modification materials will be introduced.

D- **Kaolinite:**

Kaolinite [Al2Si2O5 (OH) 4] is a naturally formed clay that is widely used in ceramic industries. Upon sintering, kaolinite undergoes a phase transformation process over a relatively broad temperature range and this thermal behavior has been extensively studied. Kaolinite, with the 1:1-type layered structure, has high crystallinity and unique structure: one side of the interlayer space is covered with hydroxyl groups of the Al2 (OH)4 octahedral sheets, and the other side is covered by oxygens of the SiO4 tetrahedron.

E- **Metakaolin:**

The less than 2µm kaolinite fraction was calcined in a programmable muffle furnace at 900 C for 2 h, and the final product is calcined kaolinite (metakaolinite, Cal K).

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F- Heeba (Hib):

(Heeba in Arabic) is usually naturally formed clay containing calcitic soil deposits. Hib has historically been used in plasters in the Theban area, and while precise source locations for the row materials of ancient plasters are difficult to determine after the passage of 3000 or more years. Hib is usually provided by local contractors who use to work in Pharaonic tombs conservation.

Hib has historically been used in plasters in the Theban area, and while precise source locations for the row materials of ancient plasters is difficult to determine after the passage of 3000 or more years (Wong, L. 2012), (Lucas, A.1906).

G- **Zinc oxide:**

Is an intriguing material since it has a wide range of applications, including optical, piezoelectric, magnetic, and gas sensing capabilities. Zinc oxide is an intriguing material since it has a wide range of applications, including optical, piezoelectric, magnetic, and gas sensing capabilities. Aside from these characteristics, ZnO nanostructures have a high surface area. high catalytic efficiency and adsorption capacity Recently, biosensing has become the most researched use of ZnO biocompatibility, and a high isoelectric point (9.5).

1-6 The main properties and characteristic of lime mortars are the following:

1-Low mechanical strengths, due to the low affinity of the calcite and quartz crystal, as well as to the weak linkage among the calcite particles.

2-Easy workability, due to the slow process of setting (carbonation) that depends on the environmental conditions.

3-High capacity of deformation (low module of elasticity). It allows the material to absorb small movements of the adjacent materials.

4-High permeability to water and water vapor.

5- Low resistance to the freeze-thaw cycles.

No presence of soluble salts, what avoids the processes of dissolution-crystallization of the salts, and therefore the appearance of Efflorescence and Sub florescence.

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These properties can be relatively modified by altering the process of production of the mortar, the type of aggregate, the aggregate/binder and water/binder ratios, etc.

1-6-1 The main properties of lime plus pozzolan mortars are the following:

1-Relatively low mechanical strength, although superiors to those in lime mortars.
2-Certain capacity of deformation (low module of elasticity).
3-Low resistance to the adverse climatic conditions.
4-Scarce presence of soluble salts.
5-Lower permeability to water than lime mortars.

1-6-2 Repairing mortars:

Materials used in the repair of buildings (especially those belonging to the Cultural Heritage) should have one essential requirement: not to accelerate deterioration of the adjacent materials. The Symposium held at Rome in 1981 recommended that restoration mortars should be prepared taking into account the characteristics of the materials on which to be applied or to replace. (Ariño X, Saiz-Jiménez C. 1996).
Numerous studies have been carried having the objective of determining the way at which the problem of repair should be approached; the errors made in the past (causing very serious damages in the Cultural Heritage) have stimulated these kinds of investigations. In the previously mentioned Symposium of Rome the following main aspects were pointed out to keep in mind in the mortars used for restoration purposes:
Research should be carried out in parallel on both new and ancient mortars.Characterization of mortars from different points of view: physical, chemical, mineralogical, etc.
Methods for measuring the fundamental parameters should be standardized. Clifton J.A. (1980).

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In 1981, for the first time, a tentative list of the characteristics of an ideal mortar for restoration was presented.

These characteristics were:

1-Easy workability.

2-Rapid and reliable setting in both dry and wet environments

3-Slow shrinking during setting

4-Mechanical and thermal characteristics similar to those of the mansory components (natural stones, bricks, etc.).

5-Soluble salts content as low as possible.

(Rossi,D.1981) indicated that repair mortars should be specifically designed for each particular case, and that some previous studies allowing to determine the mortar strength, its innocuous character, and the compatibility with the basic material should be carried out.

An important characteristic of the repair mortars is determined by the color. The white color is usually, for aesthetics, very appreciated for this type of materials. Unfortunately, the color of the white portland cement is due to the almost total absence of ferrites in the composition. It forces to white cement producers to increase the aluminates content, and it makes white cement to be especially sensitive to the attack of the sulfates. Blanco et al have developed a white portland cement resistant to the sulphates and the sea water, eliminating (almost totally) the aluminate phases of this cement and substituting them for phases that incorporate CaF_2.(Holmström, I.1981).

1-7 General requirements for lime used in the restoration of archaeological and historical buildings:

1- It is necessary to remove the lime from the product of fresh free lime.

The lime should be removed as soon as it reaches the work site.

2- It should not be used before the passage of one week. Calcium-magnesium-coated lime shall not be less than 80%

3- Fatty lime used in whiteness should not be less than 95% calcium oxide, of which 20% magnesium oxide.

4- Lime the sultan used for brushing and the paste of the mace should be of the former white high-grade burning with quiet fire.

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5- It is necessary to use water lime to make it into calcium hydroxide, and the required quantity is one third of the weight of the living lime.
6- This is accompanied by heat and an increase in volume from 2 to 3 times.
7- Add more water than usual to the regular loaf of lime with flipping to be a spicy dough. To obtain a cubic meter of lime paste, 400 grams of lime are required.
8- The lime used in the building works shall be suitable in terms of the ability to carry sand and the amount of mortar given by the force and acceptable for the rate of reaction with water, smoothness, color, hardness and time of doubt and in accordance with the Egyptian general specifications.
9- Lime is often used in construction works, where it is mixed with sand to be the mortar used to reduce shrinkage and costs. Mixing one lime to every 2-5 tons of sand to be mortar Easy to operate and hard enough to withstand the weight of the bricks.
10- The lime is sold on a lump or soft container filled with shaker. It is necessary to wash it before use in the workplace. It is fresh and must be used to remove the foreign material. If the amount of water is increased and the lime paste is formed, leave it for about two weeks before use to get suitable plasticity.

Mortar resistance to moisture:

Mortar is the weakness of the wall due to limestone hardness and moisture resistance, so moisture is leaked through it.
Moisture weakens the mortar after dehydration and leads to its solving and falling.

1-8 Apparent stone shape and color connotations:

1- Grey color indicates the ratio of carbon presence by stone.
2- Red color indicates the proportion of iron oxide in stone.
3- White color indicates the incidence of stone calcium and its purity from other substances.

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1-9 Technical problems and limestone defects:

1- Moisture effect on limestone. (Moisture retention appears on a limestone slurry and leads to mud and mold on the stone) and leads to mortar and stone falls.

2-The effect of plant growth on limestone& Sandstone. (Plants grow in the weaker part of the stone which is mortar and have their roots pressed against the stone leading to the fall of the stone).

3- The impact of insects on the limestone wall. (Insects from stone mortar take places to coexist and breed as they begin to drill tunnels and corridors leading to the abrasion of the stone).

4- Mortar carving effect used for limestone construction. (Impact of weather and rain factors on corrosion of stone and mortality)

5-The effect of overloading on the limestone wall.

Figure (3): The effect of plant growth on limestone & sandstone

1-10 Traditional methods of treating historic buildings of water:

1- Groundwater: Soil dysfunction and ventilation to prevent groundwater movement due to poetic property.

2- Rain moisture insulation: Rainwater-prone surfaces are insulated using bitumen and making trails to drain rain.

Figure (4): Soil dysfunction and ventilation to prevent groundwater movement due to poetic property.

Figure (5): Rainwater-prone surfaces are insulated using bitumen and making trails to drain rain.

2. Materials and methods:

2.1. Mortar mix proportions

A total number of 4 exploratory mortar mixtures were studied. Each mix had water/binder (W/b) weight ratio and binder to sand weight ratio of 1/3 to achieve constant flow around 160–170 mm. The various mixtures were prepared with 33,50, % (by weight) replacement of CL70-S by PK,DK,PMK,DMK and BHE respectively and 33,50, 67 % (by weight) replacement of CL70-S by HE. The mixtures proportions of mortar are shown in Table (2).

Procedure of mixing mortar was performed according to EN 196-1. The flow test was performed according to EN 196-1(BS EN 459-2:2010 - 6.8).

Table (2): Mix proportions of mortar mixtures (on weight basis)

Mix ID	Hydrated Lime	Pale Kaoline	Dark Kaoline	Pale Meta Kaoline	Dark Meta Kaoline	Homra	Heeba	Burned Heeba	Sand	W/B
CNTRL	1	-	-	-	-	-	-	-	3	0.93
LDK33	1	-	0.5	-	-	-	-	-	3	0.855
LPMK50	1	-	-	1					3	0.86
LBHE33	1	-	-	-	-	-	-	0.5	3	0.81

- **CNTRL** = lime control mix,
- **LDK33** = lime mix had 33.33% Dark Kaolinite add of 76.66% Hydrated Lime ,
- **LPMK50** = lime mix had 50% Pale Meta Kaolinite add of 50 % Hydrated Lime ,
- **LBHE33** = lime mix had 33.33% Burned Hib add of 76.66 % Hydrated Lime ,
- **Binder** is the mixture of Air lime in addition to clay materials (i.e. LPK and LHO)

2.2. Fabrication of zinc oxide/graphene oxide nanoparticles (ZnO/GO - NPs).

ZnO/GO nanocomposites were prepared using the co-precipitation method. In a typical method; a mixture of 1 M Zinc (II) acetate dehydrate in deionized water (100 ml) and 100 mg of GO were heated to 70°C followed by dropwise addition of 2 M NaOH with stirring for 1 h. The precipitate was collected by centrifugation at 8000 rpm, then washed several times with DI water, then dried in an oven overnight at 80 °C, and finally calcined at 300 °C for 4 h.

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2.3 .Chemecal & Physical and Mechanical properties studies:

Table (3): The chemical composition of lime and mineral additives used

The test was carried out according to ASTM C114-2013

Component	%			
%	* Lime	Dark Kaolinite	Kaolinite	Burned Hib
(SiO_2)	0.7003	64.3729	70.7901	43.0601
(Al_2O_3)	0.1213	17.8449	24.521	48.7578
(Fe_2O_3)	0.0062	3.0557	0.9951	4.436
(CaO)	71.8745	1.1008	1.061	1.0053
(MgO)	0.5170	0.5964	0.271	0.2806
Cl^-	0.0103	0.0282	0.091	0.0924
(P_2O_5)	0.0084	0.0113	Nil	Nil
(k_2O)	0.1672	0.1275	0.282	0.2059
(Na_2O)	0.3599	0.5890	1.2201	1.234
(TiO_2)	Nil	Nil	Nil	Nil
(SO_3)	0.0399	0.0070	0.2203	0.2514
(L.O.I)	26.1254	12.1263	0.2101	0.5766
Total	99.9311	99.9211	99.944	99.9001
$Ca(OH)_2$	96.8745	Nil	Nil	Nil
Active – SiO2 %	Nil	40.005	49.989	15.6223

Table (4): Grading of the used sand

Sieve size mm	9.51	4.75	2.83	1.41	0.707	0.354	0.177	0.075
% Passing	100	100	100	94.33	71.48	15.7	3.75	0
% Passing ES 1109/71	100	90-100	85-100	75-100	60-79	12-40	0-10	0

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2.3. Antibacterial activity of the ZnO/GO-NPs:

Growth inhibition tests against bacteria were used to investigate the bactericidal activity of the lime mortar (control) (S_1), (ZnO/GO-NPs) + Metakaolin (S_2), (ZnO/GO-NPs) + kaolin (S_3) and (ZnO/GO-NPs) + Heba(S_4). shows in table (5)

The antibacterial properties of the samples (S_1, S_2, S_3, and S_4) in comparison to the unmodified (ZnO/GO-NPs) were examined by means of agar diffusion method on Muller Hinton agar. The wells (8 mm diameter) were cut using a sterile cork borer on Muller Hinton agar (MHA, India). 24 h young culture of using pathogenic bacteria including bacteria that are Gram-positive and Gram-negative. All bacterial strain used in this article are (ATCC Strain) with the following numbers Staphylococcus aureus 6538, Escherichia coli 8739. All of the tensions, Faculty of Science Culture Centre of Microorganisms Azhar University sold them to us. The bacterial strains used in the antibacterial assay were initially, the bacteria were cultured on a solid nutrition agar medium, and then they were transferred to the agar media. Fresh colonies were injected into 100 cc of water on plates medium for nutrition broth.

Code of sample	Contents
S_1	lime mortar (control)
S_2	(ZnO/GO-NPs) + Metakaolin
S_3	(ZnO/GO-NPs) + kaolin
S_4	(ZnO/GO-NPs) + Heba

3. Results and discussion

3.1. Characterization studies

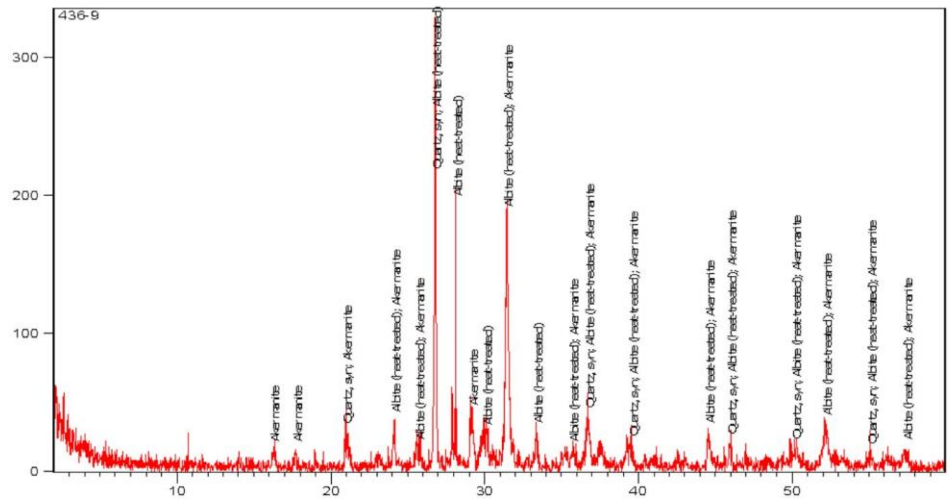

Figure (6): XRD image of Burned Hib

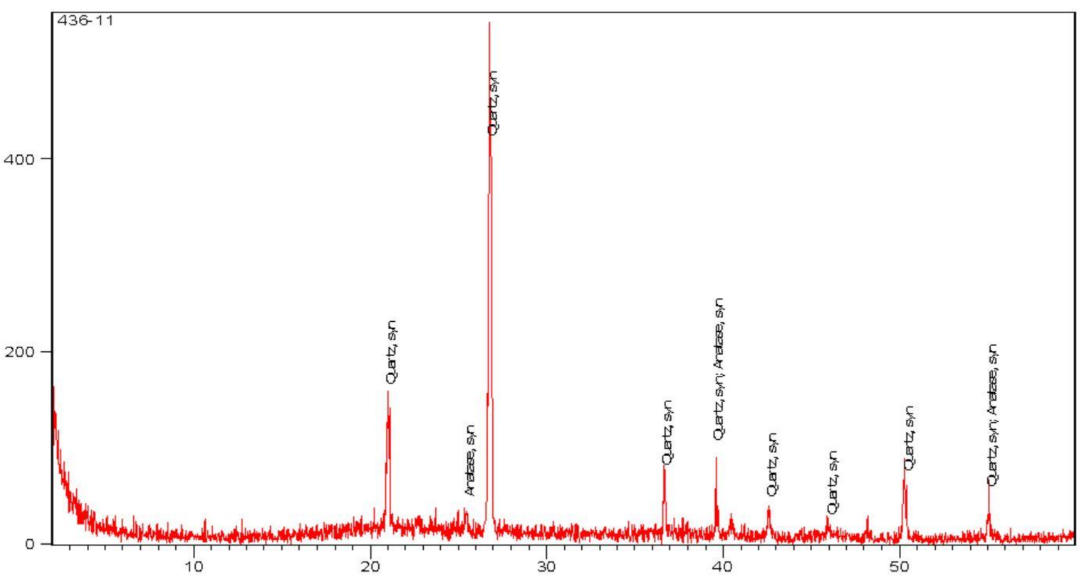

Figure (7): XRD image of Pale Meta Kaoline

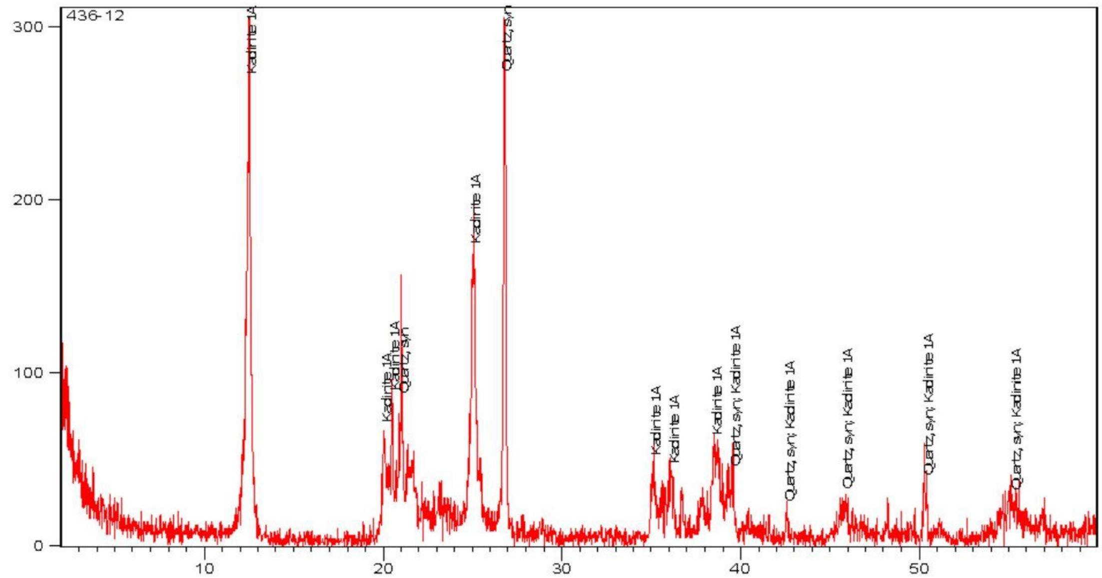

Figure (8): XRD image of Dark Kaoline

Table (5): The chemical composition of Air lime mortar mixes

The test was carried out according to ASTM C114-2013

Com.	%			
%	CNTRL	LDK 33	LPMK 50	LBHE 33
(SiO_2)	0.36	37.0	36.16	33.54
(Al_2O_3)	0.06	15.29	14.63	23.22
(Fe_2O_3)	0.004	1.58	0.09	1.29
(CaO)	60.09	32.06	39.03	31.85
(MgO)	0.27	0.59	0.28	0.22
Cl-	0.01	0.02	0.02	0.06
(P_2O_5)	0.005	0.01	0.02	0.03
(k_2O)	0.11	0.08	0.21	0.14
(Na_2O)	0.18	0.31	0.47	0.79
(TiO_2)	Nil	Nil	Nil	Nil
(SO_3)	0.02	0.02	0.13	0.15
(L.O.I)	38.01	13.02	8.94	8.42
Total	99.12	99.98	99.98	99.71
Active–SiO2%	Nil	25.02	30.24	17.73

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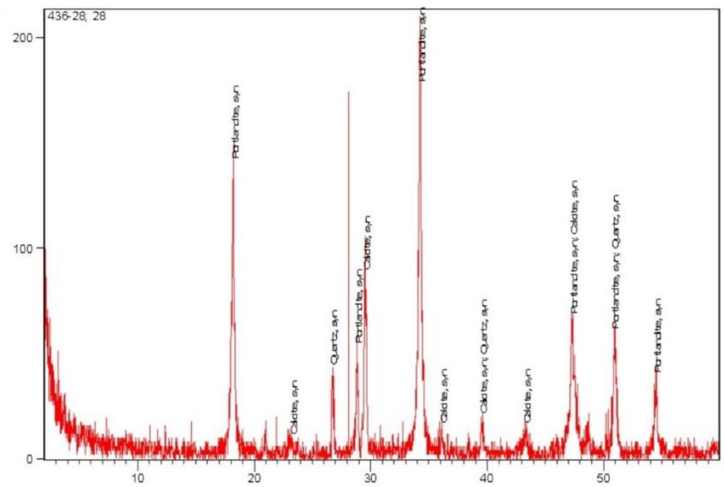


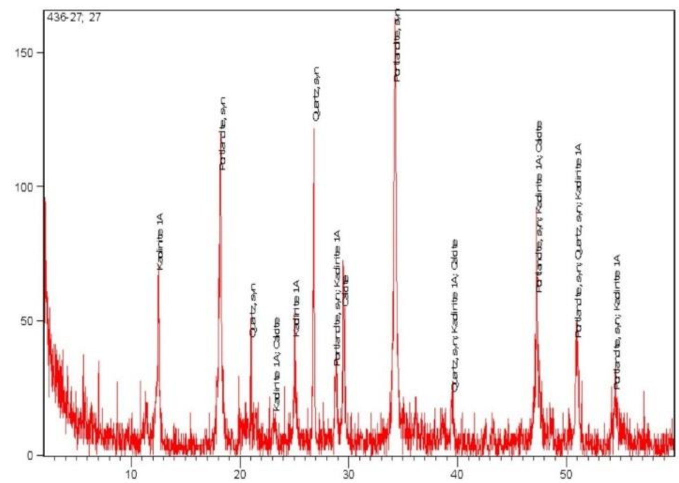
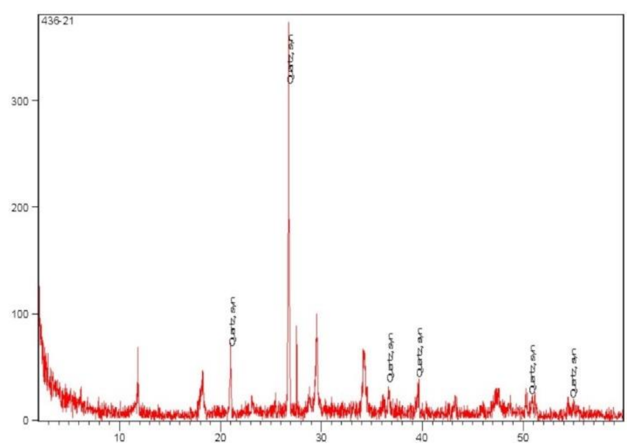


Figure (9): XRD image of base lime control mix	Figure (10): XRD image of base control lime with dark kaoline (LDK33)



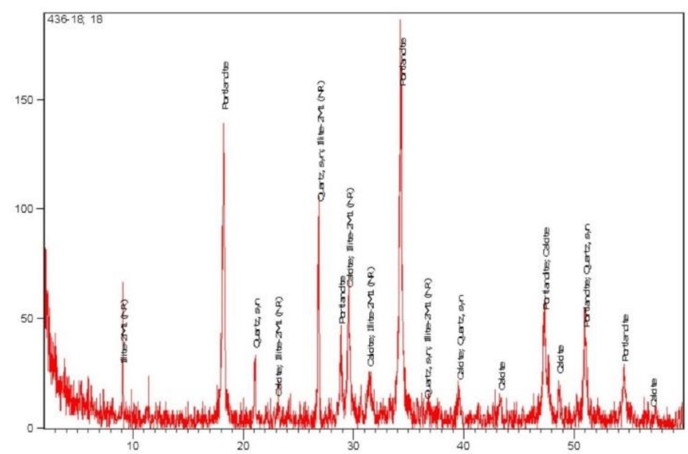


Figure (11): XRD image of base control lime with pale meta kaoline (LPMK50)	Figure (12): XRD image of base control lime with Burned Heeb (LBHE33)

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3.2. Mechanical properties studies

(a) (b)

Figure (13): Compressive strength test of mortar samples according to ASTM
;(a) Testing samples, and (b) Mode of failure

(a) (b)

Figure (14): Flexural strengths tests of mortar samples according to EN ; (a) Flexural strength test setup, and (b)) Flexural strength test failure

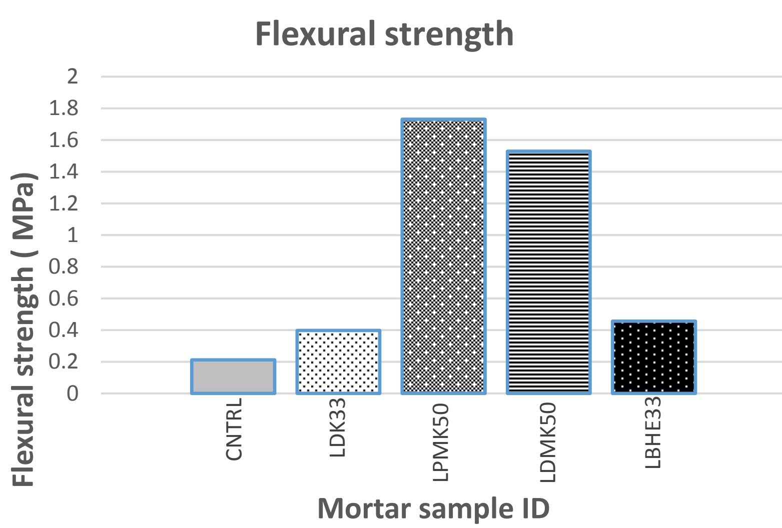

Figure (15): Flexural strength test result for 5 mixes

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Age (days)	CNTRL	LDK33	LPMK50	LBHE33
25			2.185	
27				
34				
36				
37				
39				
41				0.765
44				
51	0.325			
86				
91				
92				
93				
105		0.582		
106				
166			3.393	
168				
175				
177				
178				
180				
182				0.957
185				
192	0.540			
215			3.718	
217				
224				
226				
227				
229				
231				0.985
232				
233				
234				
241	0.577			
246		0.693		
247				
276				
281				
282				
283				
295		0.720		
296				

Table (6): Compressive strength of different mortar samples (MPa)

3.3. Antibacterial activity

The antibacterial properties of the samples were tested using Gram-positive S. aureus and Gram-negative Bacteria E. coli. The results of the bacterial colony counting experiment are shown in Figure 16. It was shown that ZnO/GO NPs can cling to the membrane of pathogenic bacteria via an electrostatic process that produces harmful ions and then forms reactive oxygen species (ROS), which prevents the bacteria from growing. [34.38]. Fig. 16 shows photographic images of the agar plats after 24 h.

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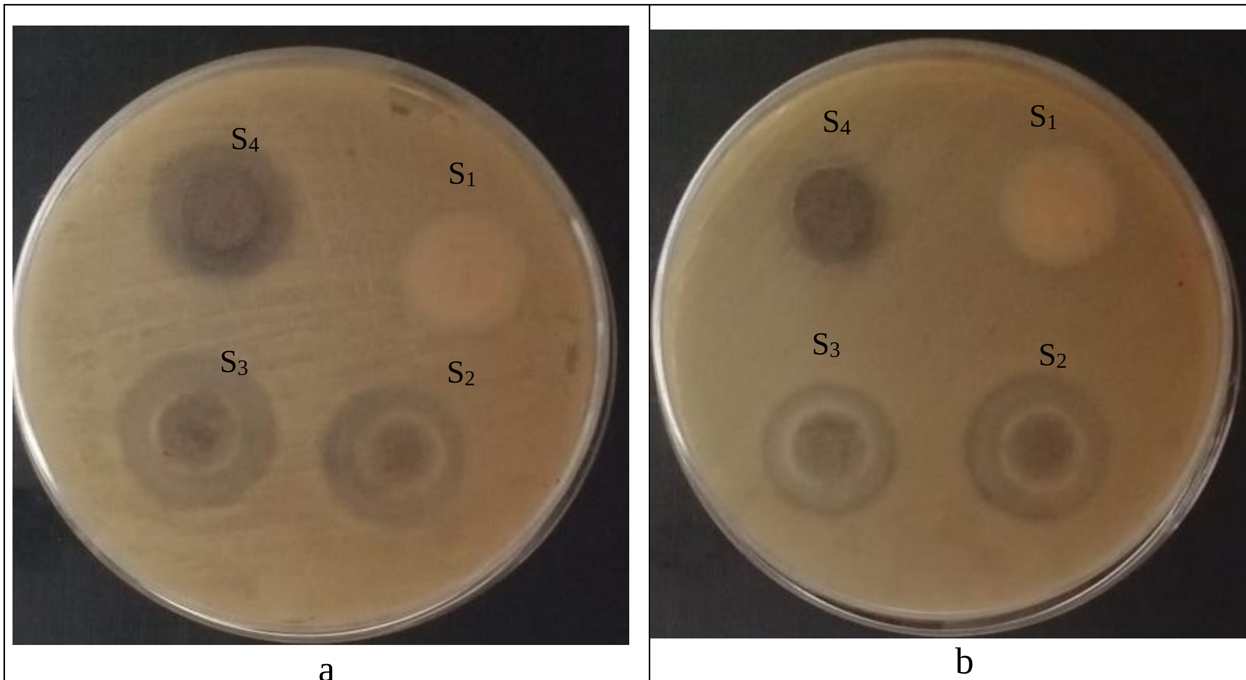

Fig. 16: Disk diffusion assay for analyzing the effect of lime mortar (control) (S1), (ZnO/GO-NPs) + Metakaolin (S2), (ZnO/GO-NPs) + kaolin (S3) and (ZnO/GO-NPs) + Heba(S4). against Gram negative (a) Escherichia coli and Gram positive (b) Staphylococcus aureus.

The results of antimicrobial tests indicated that the modified ZnO/GO NPs have higher activity than pure toward all the tested pathogenic bacteria. It was observed that the antibacterial ability increased with ZnO/GO NPs. Additionally, the inactivation of Metakaolin and kaolin was better than that of Heba and lime mortar (control) no antibacterial effect, as shown in Fig. 17

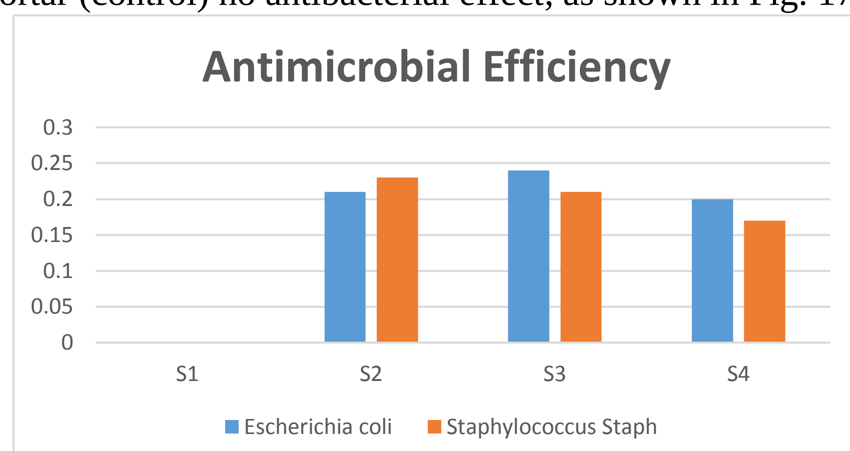

Fig. 17: Inhibition zone for lime mortar (control) (S1), (ZnO/GO-NPs) + Metakaolin (S2), (ZnO/GO-NPs) + kaolin (S3) and (ZnO/GO-NPs) + Heba(S4) against Gram negative (a) Escherichia coli and Gram positive Staphylococcus aureus.

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Recommendations for further research studies:

General recommendation

In general, there is a great demand for structural engineering research work in the field of restoration and maintenance of historical and ancient buildings. Through this study and in this regard, there is an ongoing need to develop antimicrobial solutions for heritage materials.

- Different mechanical and physical properties (e.g. strength and porosity) need to be considered well in producing conservation mortars in order to be compatible with the original materials under various exposure conditions. Such prospective complies with the general recommendation of the ICOMOS and THE VENICE CHARTER.
- Common ground between archeological, artistic and structural viewpoints needs to be linked to achieve the restoration and conservation goals. On the other hand, innovative testing programs need to be provided (e.g. nondestructive testing) to assure better routines for assessing and restoring the components of the historical building (e.g. mortars and building blocks).
- In addition, space's size between the historical building's blocks and any new blocks should consider the stability of the new mortar to assure better protection for the historical blocks.

- The new mortar color and shape is very important to achieve the compatibility of the historical mortars with modified ZnO/GO NPs have higher activity than pure toward all the tested were observed that the antibacterial ability increased with ZnO/GO NPs, In addition, space's size between the historical building's blocks and any new blocks should consider the stability of the new mortar to assure better protection for the historical blocks.

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