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Original research

Enhancing the Preservation of Submerged Marble Artifacts with Cod Liver Oil in Alexandria, Egypt

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Abstract:

The versatility of marble has made it a favored material in construction, architecture, and the creation of statues, utensils, and antiques. Its use spans various civilizations, with many marble artifacts discovered in ancient sunken cities. To address the deterioration of these sunken artifacts caused by seawater salinity, pollution, and microbiological factors, research is focusing on their preservation in underwater museums, enhancing their display's appeal and artistic realism. This study replicates similar conditions by submerging marble samples in a marine environment, comparing untreated samples to those coated with cod liver oil. The oil is chosen for its hydrophobic and antimicrobial properties, making it an environmentally friendly preservative. The results indicate that samples coated with cod liver oil demonstrated higher fracture loads and enhanced structural integrity compared to uncoated samples. The uniaxial compression test showed a significant difference, with coated samples having a fracture load of 146.5 KN compared to 117.5 KN for uncoated samples. Scanning Electron Microscopy revealed fewer fractures, smaller pore spaces, and less failure in coated samples. Additionally, X-ray diffraction showed the presence of halite in uncoated samples, which was absent in coated samples, underscoring the protective effect of cod liver oil. Conversely, the coated samples demonstrated effective water resistance and protection from biological and chemical degradation, validating the hypothesis that cod liver oil is an effective preservative. The study strongly recommends using cod liver oil to coat submerged marble artifacts, ensuring their protection from underwater deterioration and aiding in their long-term conservation.

Keywords: Marble, Cod liver oil, Preserving, Sunken Antiquities, Deterioration Factors

1. Introduction:

1.1. Background:

Marble has been used in ancient buildings since the early New Kingdom and continued through the Dynastic and Ptolemaic eras. There are many ancient quarries from which stones were brought for construction, architecture, decorative and artistic purposes. Many of them have been studied, such as granite and granodiorite quarries, Nubian sandstone quarries in the Silsila and Cairo areas, basalt quarries in Fayoum, and gabbro quarries in the Toshka and Abu Simbel areas.

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The quarries were not only sources of stone but also rich archaeological sites, offering insights into the methods and tools used by ancient Egyptians for stone extraction. Marble was brought from the areas north of the Red Sea Mountains. More studies have addressed the study of marble monuments, including (the Society for the Study of Marble and Other Stones in Antiquity), which has promoted close cooperation and integration between human and scientific disciplines, which is a comprehensive approach to studying the problems of ancient marble. In recent years, the results of research conducted on sites (quarries, monuments, and archaeological areas) and the support of geologists in the field and laboratories have led to the identification of an integrated scientific approach to treating the damage to marble monuments (Rohleder, 2001; Harrell, 2005; Harrell, & Storemyr, 2009). Manifestations and factors of damage to marble resulting from immersion in seawater include mechanical, biological, and chemical processes. Prolonged exposure to seawater can lead to physical weathering, where the marble's surface becomes rough and uneven due to the abrasive action of water and suspended particles. Additionally, chemical weathering occurs as the minerals in marble, such as calcite, react with salts and ions in seawater, leading to the dissolution and weakening of the rock structure. Studies have shown that immersion duration significantly affects the marble's shear behavior, with longer exposure resulting in reduced shear strength and stiffness. The presence of water also induces thermal stress and oxidation, further contributing to the degradation of marble's mechanical properties (Long, 2012; Wei, et al., 2022). Many marble artifacts are located in the Underwater Antiquities Museum in the eastern port area of Aboukir Bay, about 26 kilometers east of the eastern port in Alexandria Governorate, where the ruins of sunken cities were discovered, such as the city of Heraklion (Othman, 2019; Dwidar, & Abdelsattar, 2019), which is located northeast of Aboukir Beach, and the ruins of the city of Canopus, including a temple and parts of statues of Egyptian gods from the Ptolemaic and Roman eras. The two cities were exposed to natural disasters and sank in the depths of the Mediterranean Sea more than a thousand years ago. Heraklion is located northeast of Aboukir Beach, while Canopus' remains include a temple and parts of statues of Egyptian gods from the Ptolemaic and Roman eras. Both cities were exposed to natural disasters and sank into the depths of the Mediterranean Sea over a thousand years ago (Radwan, 2021; Aref, 2023). Marble is preferred in construction, architecture and the manufacture of antiques because the advantages of marble include its resistance to heat up to 350 degrees Fahrenheit, and the ability to shape marble stone and the beauty of its colors, which led to its widespread use in all successive civilizations as an environmentally available material. Marble is mainly used in buildings, monuments, architecture, interior decorations, statues, tabletops and antiques. In addition to other advantages, marble has been the source of many amazing sculptures over the centuries, as there are many ancient marble monuments, sculptures and statues all over the world (Chabas, & Jeannette, 2001; Calcinai, et al., 2019; Giustetto, et al., 2020; Issa, 2024). As a natural stone, marble can last for hundreds or thousands of years with limited environmental impact. The most common reason why white marble turns yellow is iron, which can be found in many natural stones. When exposed to water, acids, or bleach, the iron in the stone begins to oxidize and turn yellow. Marble statues corrode because marble reacts with acidic compounds, such as sulfuric acid, carbonic acid, nitric acid, etc. (Chabas, & Jeannette, 2001; Calcinai, et al., 2019; Giustetto, et al., 2020). Marble artifacts are susceptible to water damage because marble is naturally porous, so water can seep in and cause damage and discoloration. Water can also weaken the natural components of marble, causing it to crumble, break, and crack easily. Marble statues are susceptible to fractures that can reach dangerous levels, due to the stresses they are subjected to due to their weight and shape (Cámara, et al., 2017), due to surface cracks that lead to fractures, such as microscopic cracks, joint cracks, and cracks. Marble, like all calcite stones, the main causes of marble decomposition in the aquatic environment are direct surface erosion by water current, crystallization of dissolved salts, etching by microorganisms and acids in the environment, and damage caused by frost. acidic solutions from water pollution cause calcite, the main component of marble monuments, to dissolve, resulting in a loss of sculptural details (Ilievska, et al., 2016; Kamally, 2021; Lee, et al., 2022; Brodziak-Dopierała et al., 2023; Pycia, et al., 2024).

1.2. Topic Importance:

Preserving the sunken antiquities and displaying them in an underwater museum, the museum is the first in Egypt and Africa and will contribute to relieving pressure on existing diving sites and preserving the “natural treasures of the region,” and the visitors will enjoy the spirit of excitement and adventure. Imagine transitioning into a marvellous underwater world where sunken antiquities are showcased in the first underwater museum in Egypt and Africa. This pioneering move not only alleviates pressure on existing diving sites but also preserves the region's "natural treasures". Visitors will feel a sense of excitement and adventure as they immerse themselves in a journey through history beneath the waves. Additionally, the museum offers a unique opportunity to study the preservation of sunken antiquities, contributing to advancing global underwater heritage conservation techniques. This incredible experience will inspire future generations to appreciate and protect the region's history.

1.3. Study Hypotheses:

There is a group of materials that have the ability to repel water, known as hydrophobic materials, including oils, which isolate marble from the harmful aqueous environment. Some oils have other properties that can be used for research, such as resistance to fungi that damage marble, such as cod liver oil. The study assumes that the hydrophobic properties of cod liver oil repel seawater from submerged marble, thus protecting the marble from various deteriorating factors, in addition to previous studies that addressed the resistance of cod liver oil to fungi that cause damage to marble.

1.4. Study Objectives:

The study aims to address two pivotal questions concerning the protective capabilities of cod liver oil when applied to marble submerged in seawater. Firstly, the research seeks to evaluate the effectiveness of cod liver oil in repelling seawater by examining the oil's ability to create a hydrophobic barrier on the marble's surface. This would significantly reduce water absorption and protect the marble from detrimental effects, such as salt crystallization, which can lead to physical stress, cracking, and chemical reactions that deteriorate the stone. Secondly, the study investigates the antimicrobial properties of cod liver oil, particularly its ability to resist fungi that contribute to the degradation of marble. This aspect focuses on assessing how well cod liver oil can inhibit fungal growth, thus preventing biological processes that weaken marble structures and cause surface damage. The primary objective is to employ an integrated scientific approach to experiment with materials that can effectively protect the marble from seawater, reduce its damaging salty effects, and prevent internal damage. The study also seeks to identify materials that can resist harmful fungi, meeting criteria such as being natural, non-destructive, environmentally friendly, and harmless to both the marble and the surrounding marine ecosystem. By addressing these questions and objectives, the study aims to demonstrate the dual functionality of cod liver oil in maintaining marble’s structural integrity in harsh marine environments. The goal is to offer a sustainable and environmentally friendly solution for conserving submerged marble artifacts, enhancing their longevity, and preserving cultural heritage. The findings could pave the way for broader applications in underwater conservation efforts and the establishment of underwater museums, which would serve as educational and touristic attractions while promoting the importance of preserving historical artifacts in their natural environments.

1.5. Literature Review:

Many studies have addressed the types of damage to which marble artifacts are exposed. Examination of marble damage in Alexandria using scanning electron microscope of the marble surface areas revealed many cracks and micro-holes, especially black spots due to real and molecular change (Ezz Eldin, & Magdy, 2024; Ugolotti, et al., 2021; Wang, et al., 2022). Microbes were also examined, including fungi such as Cladosporium and Aspergillus 1, which cause black spots on the marble surface. In addition to carbon, the solution reacts with organic materials such as humus, acrylic resin, decomposing plants, and microorganisms, dissolves inside the stone, migrates, and succeeds on the surface and base of the marble statues, forming complete pieces and structures. Many research attempts have been made to treat and protect sunken marble artifacts. One notable method involves coating these artifacts with nano-titanium, creating a self-cleaning surface that repels water and contaminants. This technology helps prevent the formation of biofilms, which are harmful colonies of bacteria and microorganisms that produce acidic byproducts. These acids can degrade marble over time, so reducing biofilm growth is crucial. Other innovative materials, like silicon-based coatings, have also been employed to protect marble from acidic attacks. Beyond these treatments, preservation strategies are implemented, such as maintaining controlled environmental conditions, monitoring regularly, and performing routine maintenance (Gelany, et al., 2024). These combined efforts are essential for safeguarding the integrity and longevity of these priceless cultural artifacts. In addition to these innovative coatings, other approaches have been explored to protect submerged marble artifacts. Researchers have developed techniques to infuse marble with materials that enhance its structural integrity. This involves using compounds that can penetrate the porous structure of marble, providing added resistance to environmental stresses (Mohsen, et al., 2024). Moreover, some studies focus on applying protective films that can adapt to the marble's natural expansion and contraction due to temperature changes, thus preventing cracks. Advanced laser cleaning methods have also been employed to remove biofilms and other deposits without causing mechanical damage to the marble surface. Combined with preservation strategies like monitoring underwater conditions and adjusting conservation techniques as needed, these measures ensure that marble artifacts remain preserved for future generations to appreciate (Naidu, et al., 2016; Salam, et al., 2024; Ashraf, et al., 2024; Nader, et al., 2024; Elsayed, et al., 2024; Moustafa, et al., 2024; Ebrahem, et al., 2024).

2. Material and Methods:

2.1. Sampling and Treatment Processes:

Samples were prepared that matched the white marble samples that make up the sunken marble monuments. Then, the samples were coated with cod liver oil and placed in water in 30 days from the Aboukir area. Other samples were placed in the water from the archaeological Aboukir area to compare the samples coated with and uncoated with cod liver oil. Figure 1.

Shows the preparation of marble samples, coating them with cod liver oil, and placing them in a similar seawater medium.

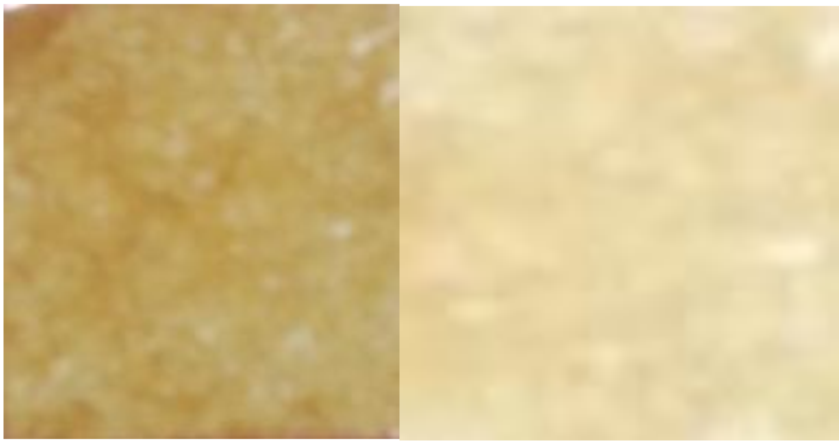


Figure 1. Samples of marble coated by cod liver oil

For X-ray investigation, two Marble samples were prepared for this study. The first sample was subjected to sea water exposure to simulate natural weathering conditions, while the second sample was treated with a protective oil coating. Both samples were obtained from the same source material to ensure compositional consistency for comparative analysis.

2.2. Examination processes:

The experimental process included several stages: (1) preparing marble samples similar to the sunken marble artifacts, (2) coating the samples with cod liver oil, (3) comparing the coated and uncoated samples, (4) comparing the breaking strength of coated and uncoated samples, comparing X-ray diffraction of coated and uncoated samples, and (5) comparing coated and uncoated samples using a scanning electron microscope under different magnifications.

2.3. Testing of mechanical processes (Uniaxial Compression Test):

To simulate the vulnerability of marble when exposed to different damage factors under sea water, it was compared with a similar marble coated with cod liver oil that was exposed to the same damage factors and under the same conditions. The fracture load was tested by a uniaxial compression tester, at the Soil and Foundation Testing Unit, Faculty of Engineering, Alexandria University. and the Specifications of the tested samples were as shown in Table 1.

Table 1. Specifications of the tested samples

Type of samples	Dimensions (cm)			Size ($cm^{3)}$	Weight(gm)
	Length	Width	Height		
Uncoated Marble	5.6	5.1	2.8	79.968	199.5
Coated Marble	5.6	5.1	2.8	79.968	198.41

2.4. Xray Differaction:

X-ray diffraction is a technique that can compare samples that have been deteriorated by salts with those that have not been deteriorated by salts. Samples coated and uncoated with cod liver oil were analyzed at the laboratories of the Faculty of Science, Cairo University. The mineralogical composition of both samples was determined using X-ray diffraction (XRD) analysis. The measurements were performed under the following conditions:

- Instrument: Bruker D8 Advance X-ray Diffractometer
- Radiation Source: Cu-Kα radiation (λ = 1.5406 Å)
- Operating Conditions: Voltage: 40 kV - Current: 40 mA
- Scanning Parameters: 2θ Range: 14° to 79° - Step Size: 0.02° - Scan Speed: 2°/min
- Detector: Lynx Eye XE detector -Sample Mounting: Zero-background silicon holder
- Analysis Software: DIFFRAC.EVA software for peak identification and phase analysis

2.5. Scanning Electron Microscope:

Scanning electron microscopy at different magnifications is a technique that can show the deterioration of marble crystals and grains, as well as microcracks and fragmentation of rock grains. The scanning electron microscopy was carried out in the laboratories of the Faculty of Science, Cairo University.

3. Results:

3.1 Result of Uniaxial Compression Test:

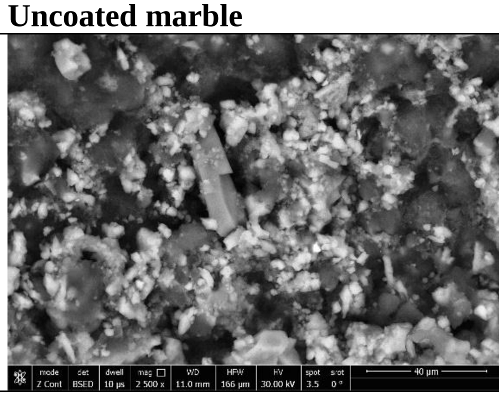
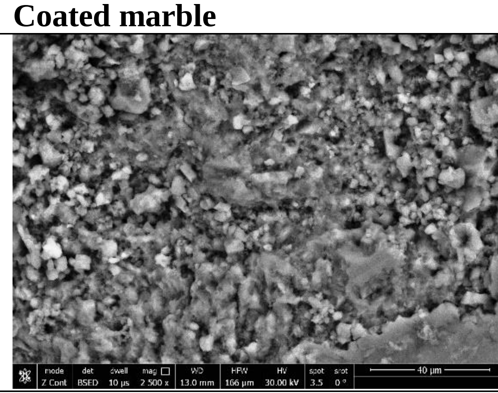
The results of the fracture load test showed a large variation between the coated and uncoated samples. The fracture load of the samples coated with cod liver oil is 146.5 KN, equal to 14938.6 Kg, while the fracture load of the uncoated with cod liver oil is 117.5 KN, equal to 11981.5 Kg. That means the fracture load of the samples coated with cod liver oil is greater than the fracture load of the samples uncoated with cod liver oil, as shown in Table 2.

Table 2. Results of the fracture load test for the samples:

Type of samples	Fracture load (KN)	Fracture load (Kg)
Coated Marble	146.5	14938.6
Uncoated Marble	117.5	11981.5

3.2 Result of scanning Electron Microscope:

Figure 2. shows images of Scanning Electron Microscopy; samples coated with cod liver oil and others not coated with cod liver oil were examined. At 40 µm, fractures in the calcite crystals of the uncoated sample were evident. At 50 µm, a greater variation in the width of the pores between the grains of the uncoated marble sample was observed compared to that of the cod liver oil-coated sample. At 100 µm, the failure of the uncoated limestone grains was evident to a greater extent than the deterioration of the grains of the coated samples. At 400 µm, more salt crystals appeared in the uncoated sample than in the cod liver oil-coated sample.

Samples	**Uncoated marble**	**Coated marble**
Fracture Calcite crystal (Scale 40µm)		

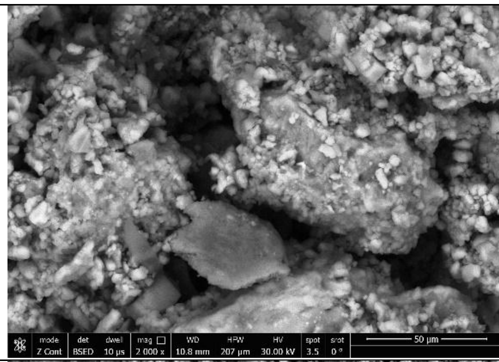
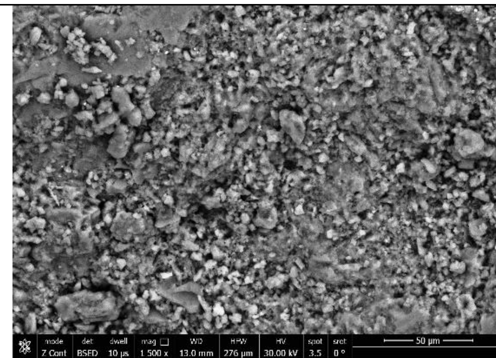
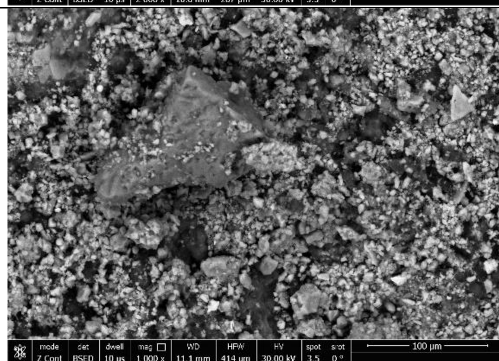
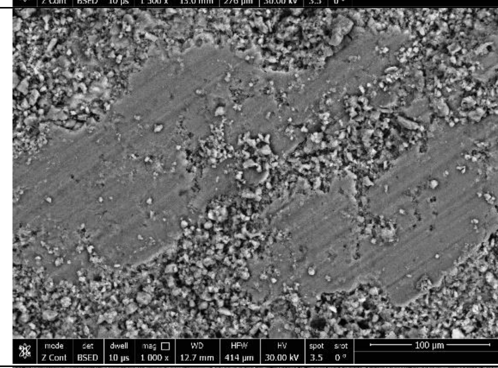
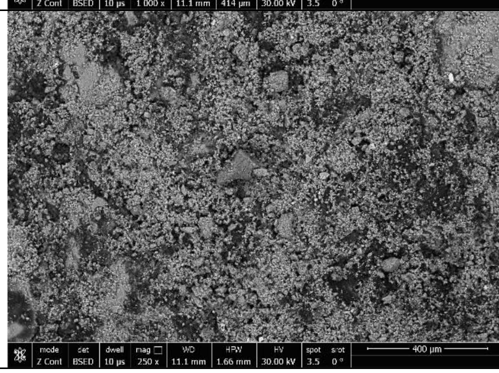
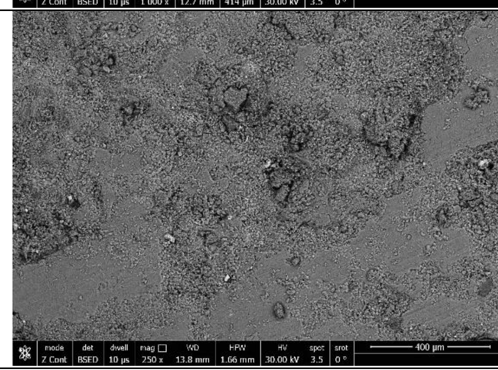
Whole Pore space (Scale 50µm)		
Failure in particle (Scale 100µm)		
Appearance of salts (Scale 400µm)		

Figure 2. Comparison between coated and uncoated samples under various scales.

3.3 Result of Xray diffraction:

Figure 3. illustrates the result of X-ray diffraction of the uncoated sample and coated sample; Halite appears in uncoated samples while it disappears in coated samples.

4. Discussion:

The weight of the samples varied despite the homogeneity of the size of the coated and uncoated samples, which means that the marble coated with cod liver oil as a hydrophobic substance prevents water from entering the pores of the stone to some extent. The variation in the fracture load test result also explains the weakness, failure and fragmentation resulting from the interaction of seawater waves and salts in the uncoated sample. X-ray diffraction analyses confirmed the appearance of salts in the uncoated samples and the absence of salts in the coated samples due to the lack of infiltration of salty seawater into the marble sample. Mineralogical Alterations: The XRD patterns reveal significant mineralogical changes in the deteriorated Marble compared to the oil-treated sample. Oil creates a hydrophobic barrier, reduces water penetration, and maintains structural integrity, thus preventing mineral dissolution and stress-induced microcracking. This comparative analysis highlights the destructive effects of seawater on granite and the protective benefits of oil treatment, providing valuable insights for conservation practices. Scanning electron microscope examinations showed clear signs of damage in the uncoated samples, such as fracture of calcite crystals, erosion of marble grains, widening of pores between marble grains, and the appearance of destructive salts, which was not evident when compared to the examination of samples coated with cod liver oil, at the same SEM scale.

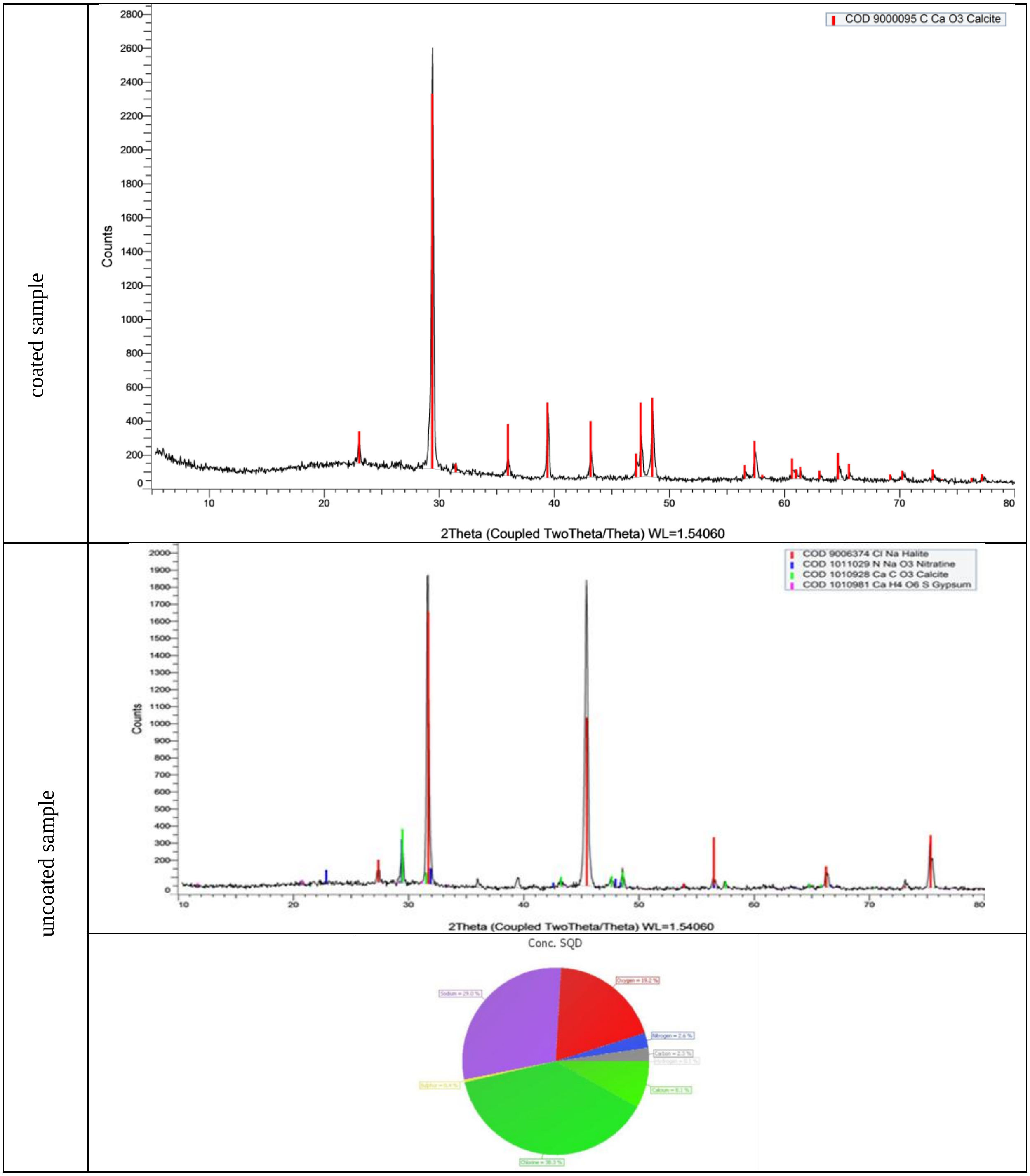


Figure 3. X-ray diffraction of uncoated and coated marble.

5. Conclusion:

The research hypothesis successfully demonstrated the effectiveness of using cod liver oil to protect marble submerged in seawater. Previous studies have established that cod liver oil acts as a water-repellent and antimicrobial agent for marble, making it an ideal candidate for this application. The successful protection of marble in seawater environments, similar to that of submerged marble monuments, is a compelling motivator for further studies. These studies could pave the way for applying such techniques to protect submerged marble monuments, thereby fostering the establishment of museums dedicated to these underwater artifacts. In this research, X-ray diffraction (XRD) patterns were utilized to reveal the mineralogical changes between deteriorated marble and the cod liver oil-treated samples. The application of cod liver oil forms a hydrophobic barrier on the marble's surface, significantly reducing water penetration. This barrier helps maintain the marble's structural integrity by preventing mineral dissolution and stress-induced microcracking, which are common issues uncoated marble faces in marine environments. The destructive effects of seawater on marble are well-documented; exposure leads to salt crystallization, biological growth, and chemical reactions that degrade the stone. Using cod liver oil as a protective coating mitigates these effects by creating a protective layer that repels water and inhibits microbial activity. This is particularly important for submerged marble monuments, which are constantly exposed to harsh marine conditions. Furthermore, the success of this method underscores the potential of using natural oils to conserve historical artifacts. Natural oils, such as cod liver oil, offer an environmentally friendly alternative to synthetic sealants and coatings, which can harm the artifacts and the surrounding ecosystem. The findings of this study suggest that cod liver oil could be a sustainable solution for the long-term preservation of submerged marble monuments, promoting both cultural heritage preservation and environmental protection. This research highlights the importance of exploring natural, effective methods for protecting and preserving historical artifacts submerged in seawater. The use of cod liver oil not only enhances the durability of marble but also supports sustainability goals by reducing the need for synthetic materials and preserving the natural marine environment. This innovative approach aligns with the broader goals of cultural heritage preservation and sustainability, providing a strong foundation for future research and application in the field of underwater conservation.

6. Recommendation:

To ensure the preservation of sunken marble artifacts, it is highly recommended to apply a coating of cod liver oil, a natural substance that poses no harm to marine life, and has been proven effective in protecting underwater marble. Acting as both a water-repellent and antimicrobial barrier, cod liver oil significantly reduces water penetration and inhibits microbial growth, thereby maintaining the structural integrity of the marble. This innovative technique has demonstrated its potential in safeguarding underwater marble monuments from deterioration caused by environmental factors, making it an excellent solution for long-term conservation. Additionally, establishing underwater museums to display these protected artifacts can create unique and engaging attractions, promoting cultural heritage conservation and sustainable tourism. By presenting preserved historical artifacts in their natural submerged state, these museums offer an immersive experience that educates the public on the importance of marine conservation and historical preservation. Continued research and application of cod liver oil coating technology will further enhance its efficacy and contribute to broader efforts in preserving our cultural and historical legacy while supporting environmental sustainability.

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