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INFLUENCE OF STORAGE CONDITIONS OF ALABASTER MONUMENTS ON THE NATURE OF SURFACE CONTAMINATION

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Abstract

Studies of the deterioration of alabaster products have been conducted in many countries; however, this topic is still an inexhaustible source for scientists. Alabaster from different regions has its own distinctive morphological peculiarities. Climatic conditions, air pollution, storage and operation conditions affect the conservation state of the stone to a certain extent. Without sufficient information about the causes of stone degradation, it is impossible to estimate the risks of its destruction under the influence of external factors and to provide appropriate recommendations on the storage conditions of monuments. In order to determine the causes of destruction of the alabaster monuments and to study the nature of the stone surface contamination, a number of fields, microscopic and comprehensive thermal studies were conducted. Based on the study results, the nature of alabaster destruction was outlined, the danger of atmospheric and anthropogenic factors was estimated, and the comparison of the change in material composition of the crystalline mass of the alabaster surface layer was carried out. Study results are presented in the form of photography and microphotography, summary tables of thermal analysis, comparison of thermogravimetric curves of the alabaster fragment and encrustation with a near-surface layer of alabaster.

***Keywords**: Alabaster, Monument, Deterioration, Causes of destruction, Thermal analysis*

Introduction

Alabaster is a specific type of gypsum, characterized by greater hardness (Mohs hardness up to 3.0 and higher) and a fine-grained or a cryptocrystalline structure of grains aggregates. The name ‘alabaster’ is considered to come from the Greek ‘ἀλαβαστρίτης’, which presumably comes from the name of the Greek town *Alabastron*. Alabaster is easily worked and extensively used to make a wide range of products for secular and ecclesiastical purposes, and interior or exterior decoration. The appearance of this stone bears a resemblance to marble. The colour scheme varies from snow-white to honey-like and dark grey; the texture of the stone may be veined, spotted, banded or pure white. In Ukraine, alabaster was the most common stone in the Lviv region. Alabaster products were one of the main products sold by the 19th century guild artisans in Zhydachiv, Rozdil, Zhuriv, Lukovytsia, Zhuravno and Novoshyno 0.

In Ukraine, there are marvellous examples of alabaster decoration in architecture and art, interior and exterior design. These include Adam Kysil’s headstone (the Dormition Church, Nyzkynychi village); Sieniawski family’s headstones in the Olesko Castle, Olesko town;

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entrance portal of the St. Bartholomew Church in Drohobych; window trimming of the Arian Tower in Belz; window trimming in Bandinelli Palace's interior; etc.

Depending on the storage conditions, alabaster monuments are affected by various external factors that cause the appearance of a certain type of contamination and gradual destruction of the monuments.

The contamination that gets on the stone surface from the atmosphere can be natural and anthropogenic. Natural dirt that gets on the monument's surface from the atmosphere includes dust that is blown away by the wind from the soil surface, and that occurs as a result of fires in forests and steppes. Dust contains easily soluble salts that have risen through the capillary tubes in soil during dry seasons, and salts contained in precipitation. Dust consists of spores of different fungi and microorganisms that live in the organic component of dust 0. Dust particles have a clay constituent that can keep moisture. They penetrate into the smallest cracks and cavities and have a negative effect on alabaster. Dust is also a substrate for the development of living organisms: mosses, fungi, lichens, vascular plants or bacteria [4, 5].

Anthropogenic pollution is a result of burning fossil fuels. Pollutants include SO_2 (sulfur dioxide), CO (carbon monoxide) and NO_x (nitrogen oxide). They are contained in emissions from thermal power stations and vehicles [6]. Contaminants resulting from human activities affect not only human health but also the conservation state of alabaster monuments. Nitrogen oxide and sulfur dioxide have high toxicity and can cause changes in the atmosphere, such as acid rain and smog 0.

Western Ukraine is in a temperate continental climate zone. In winter, the average daily temperature reaches below zero, and the relative humidity exceeds 80%. In summer, the temperature is above 15°C, humidity varies from 60 to 70%. Temperature fluctuations in different seasons and the effect of anthropogenic factors significantly influence the conservation state of architectural monuments. Therefore, the aim of the work is to study the influence of various factors on the conservation state of alabaster monuments using physicochemical methods of analysis, and to identify the nature of contamination on the stone surface.

Experimental part

Materials

In order to study the influence of the storage conditions, four monuments were chosen: a headstone from Lychakiv Cemetery (20th century), a baptismal font from the Pidhirtsi Castle (19th century), a baptismal font from the courtyard of the Dormition Church (18th century). For a long time, they were kept in various conditions.

Methods

The study used general scientific methods of field studies in lateral and frontal lighting. Electron light microscope with reflected light (Digital Microscope, H800/U500X ST2) connected to the personal computer was used to record the relief, structure and texture of the stone, to carry out a detailed analysis of the state of alabaster crystalline mass, to study the structure and type of surface contamination.

The study of the objects in thin section was done with the use of a petrographic microscope МИН-8 (MIN-8).

The nature of impurities was discovered using comprehensive thermal analysis. The thermal analysis was carried out using Q-1500D derivatograph (system F. Paulik, J. Paulik and L. Erdey) connected to the personal computer, in a temperature range 20-1000°C. The samples were analysed dynamically at a heating rate of 5°C per minute. The mass of the samples was 270-500mg; the reference substance was aluminum oxide [10].

Thermal analysis is one of the important physical and chemical methods for studying properties of a substance. Based on thermal analysis data, thermal stability of substances is studied, and the nature of the processes occurring during the heating of substances is researched. In addition, the method of thermal analysis can determine the mineral composition of the rock. Thermal analysis results can give a clear understanding of the nature of bound water, and by the nature of the mass lost during the heating of a sample, it is possible to study the influence of various factors on its structure and chemical composition **Error! Reference source not found.**.

Results and discussion

Headstone from the Lychakiv Cemetery

The headstone is made of white alabaster with light-grey and light-brown veins. The alabaster has a fine-crystalline structure. The headstone was made in the mid-20th century, and it was polished.

The headstone is located in the park area of Lychakiv Cemetery, in the open, without protection from climatic factors, and in a high humidity area (Fig. 1).

Fig. 1. Overall view of the alabaster headstone from the Lychakiv Cemetery

Water caused dissolution, corrosion, cracking of crystalline mass of the stone (Fig. 2).

Because of water pressing on the headstone, thin furrows were formed in the direction of the water flow. The consequence of high humidity is the dissolution of crystalline mass and excessive moistening of clay impurities (Fig. 3). Minerals that make up clay can absorb moisture, increase in volume, and release moisture at high temperatures. Sudden temperature change causes the formation of new cracks and increases the size of the existing cracks in the stone. Ceaseless process of the cleavage of alabaster can be seen on the headstone. In fact, cracks form in the lines of clay veins (Fig. 4).

Dust, clay, and sand brought by wind to the surface of the stone act as a substrate for the development of the organic matter, and high humidity creates favourable conditions for its growth. Cavities and cracks are filled with dirt and become inhabited by small insects and microorganisms. The organic components, such as moss, lichens and vascular plants, are evident (Figs. 5, 6 and 7).

Fig. 2. Deep cracking of the stone

Fig. 3. Dissolution and corrosion of crystalline mass, volume increase of clay component

Fig. 4. Evolution of organic matter on the stone surface

Fig. 5. Microphotograph of the crystalline structure and evolution of organic matter

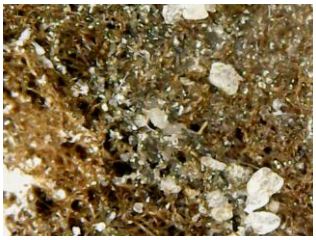

Fig. 6. Microphotograph. Dead organic matter with torn alabaster crystals

Fig. 7. Microphotograph. Cleavage. Organic component between the layers of crystalline mass

Their roots grew deep between the alabaster crystals that led to deep cleavage and breaking of crystalline mass of the stone. Small insects, cobwebs and larvae inhabit cavities, cracks and hollows.

Natural external factors – high humidity, wind, rain, snow, frost, and high air temperature – affected the aesthetic appearance of the monument: destroyed the polished surface; caused the loss of characteristic transparency of the stone; moss, lichens and dust changed the colour of the stone.

The roundness of alabaster crystals can be clearly seen under the microscope.

Table 1. Results of thermal analysis of the alabaster fragment and encrustation of the headstone from Lychakiv Cemetery

Sample	Stage	Temperature interval, °C	Mass loss, %
Alabaster fragment of the headstone	I	20 - 103	0,12
	II	103 - 298	22,56
	III	298 - 417	-
Encrustation with a near-surface layer of alabaster of the headstone	I	20 - 103	0,75
	II	103 - 268	24,65
	III	268 - 428	2,45
	IV	428 - 625	0,99

According to the results of thermal analysis, the negligible mass loss of the alabaster fragment (0,13%), in a temperature interval 20-103°C, at the first stage of thermal decomposition, corresponds to the release of moisture adsorbed by the sample.

An intense mass loss (22.56%) of the alabaster fragment sample in a temperature interval of 103-298°C, at the second stage of thermal decomposition, which takes place with the appearance of a double endothermic effect, corresponds to the loss of crystalline water bound with gypsum. The first endothermic effect with a maximum at 199°C corresponds to the loss of 1,5 water molecules. The next endothermic effect with a maximum at 224°C corresponds to the loss of 0,5 water molecules **Error! Reference source not found.**. In the same temperature range, the residue of physically bound water by the sample is lost.

The appearance of negligible exothermic effect on a differential thermal analysis (DTA) curve of a sample 1 in a temperature range 300 – 417°C, which is not accompanied by the mass loss of the sample, may be due to the processes of polymorphic conversion of anhydrite.

The different shape of the TG and DTG curves of the alabaster fragment and the encrustation with a near-surface layer of alabaster indicates the presence in the sample of encrustation of a certain type of dirt, which was not identified by microphotography.

The appearance of significant exothermic effect on a DTA curve of the sample of encrustation with a near-surface layer of alabaster, in the temperature range 268 – 428°C, accompanied by the mass loss, corresponds to the combustion of the sample's organic component.

A more significant mass loss of the encrustation sample (24.65%) in the temperature range 103 – 268°C, in comparison with the alabaster fragment, may be due to the release of interlayer water in a clay component of the sample.

The presence of clay contaminants is confirmed by the mass loss of the encrustation sample in the temperature range 428 – 625°C, at the fourth stage of thermal decomposition. In this temperature range, there is a gradual destruction of the structure of clay minerals, which is accompanied by the loss of constitutional water 0.

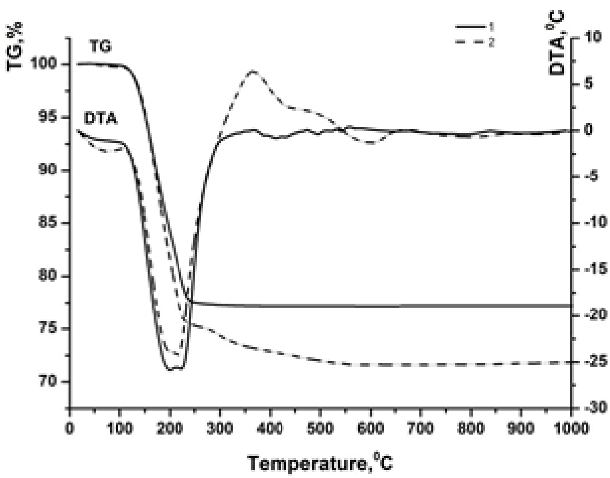

Fig. 8. TG DTA curves of the Lychakiv headstone samples:
1 - alabaster fragment, 2 - encrustation with a near-surface layer of alabaster

Alabaster baptismal font from the Pidhirtsi Castle

The alabaster baptismal font is kept in one of the halls of Pidhirtsi Castle. It dates back to the 19th century. The alabaster baptismal font consists of two parts: a basin and a pedestal (Figs. 9-11). The font is 110cm long. It is kept at the entrance to the hall of Pidhirtsi Castle (Pidhirtsi Castle is located in the village of Pidhirtsi in Lviv Region. The castle is a late

Renaissance and baroque monument. It was built in 1635-1940 under the charge of Andrea del Aqua by order of Grand Crown Hetman Stanisław Koniecpolski). Relative humidity of the room is 60%, the temperature fluctuates from +10 to +15C°C. Boris Voznytskyi, the former director of the Lviv Art Gallery, brought the baptismal font there.

Fig. 9. Overall view of the alabaster baptismal font

Fig. 10. Cleavage, cracks and dirt on the pedestal surface of the alabaster baptismal font

Fig. 11. The flowing water effect. Dissolution of alabaster crystalline mass in the direction of water flow.

The baptismal font pedestal has deep cracks and stone cleavage, as well as a thick incrustation of black patina and dust contamination. Protruding formations on the surface of the stone, confined to the cracks, are documented (Fig. 10). The cracks and cleavage appeared because of sudden temperature change and frequent soaking of the stone.

Microscopic study of the alabaster fragment of the baptismal font in thin section found that the alabaster aggregates have a fine-grained structure, and among the gypsum crystals there are areas of fine-grained carbonates clusters with the clay minerals impurities. They form grey areas and give alabaster a heterogeneous colour.

The alabaster's clay component can facilitate the formation of cracks on the surface of the baptismal font. It is known that clay minerals with a swellable component can actively absorb moisture 0. It is known that when water freezes (in cold seasons) it can increase in volume. It leads to the appearance of internal pressure in crystals, under the influence of which the existing cracks increase and new cracks form on the alabaster surface. Rise in air temperature and exposure to direct sunlight lead the clay minerals to the loss of surface water. This causes dehydration and breaking of clay inclusions from the crystalline mass, and formation of microcracks.

Microphotographs of cracking of alabaster crystalline mass and cracks filled with surface contamination are presented in Figures 12-14.

Fig. 12. Microphotograph of the cracking of crystalline mass, and

Fig. 13. Microphotograph of cleavage process of alabaster.

Fig. 14. Clusters of dirt. Surface formations confined to cracks

filling of the cracks with dirt Surface contamination

The influence of atmospheric pollution and climatic conditions caused the change of aesthetic appearance of the monument, namely: colour change of the stone (it can be seen that under the layer of dirt the alabaster is white, veined and has a cream tinge); polished surface of the stone is lost, the luminescence and characteristic transparency are missing.

The results of microscopic studies are confirmed by the data of thermal analysis, which was presented in Figure 15 and Table 2.

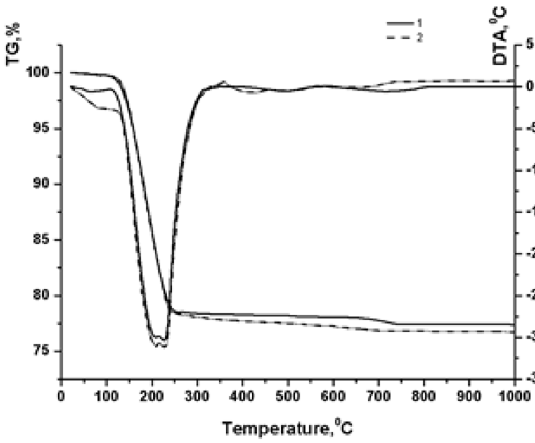

Fig. 15. TG and DTG curves of baptismal font samples from the Pidhirtsi Castle:
1- alabaster fragment, 2 - encrustation with a near-surface layer of alabaster

Table 2. The results of thermal analysis of the alabaster fragment and encrustation with a near-surface layer of alabaster of the baptismal font from the Pidhirtsi Castle

Sample	Stage	Temperature range, °C	Mass loss, %
Alabaster fragment of the baptismal font from the Pidhirtsi Castle	I	20 - 110	0.32
	II	110 - 310	21.29
	III	310 - 623	0.31
	IV	623 - 753	0.63
Encrustation with a near-surface layer of alabaster of the baptismal font from the Pidhirtsi Castle	I	20 - 110	0.32
	II	110 - 310	21.67
	III	310 - 434	0.37
	IV	434 - 623	0.51
	V	556 - 753	0.31
	VI	753 - 1000	0.07

The mass loss of the alabaster fragment sample of the baptismal font from the Pidhirtsi Castle (0.32%) at the first stage of thermal decomposition, in the temperature range 20 – 110°C, is conditioned by the release of adsorbed water.

At the second stage of thermal decomposition, in the temperature range 110 – 310°C, the sample loses its mass due to the release of water of crystallization bound by alabaster. In the same temperature range, the interlayer water of the clay minerals, present in the structure of alabaster, is lost.

At the fourth stage of thermal decomposition in the temperature range 623 – 753°C, the carbonate constituent of the alabaster fragment sample decomposes.

The water loss at different stages of thermal decomposition and the decomposition of carbonate constituents are accompanied by the appearance of endothermic effects on the DTA curve. The presence of clay and carbonate components in the alabaster structure is confirmed by the results of microscopic studies.

As opposed to the alabaster fragment sample, the appearance of exothermic effect can be seen on the thermogram of the encrustation with a near-surface layer of alabaster, in the temperature range 310 – 434°C, at the third stage of thermal decomposition. This reaction is accompanied by the mass loss (0.37%) of the sample and corresponds to the combustion of the sample's organic component, present in the form of impurities.

In contrast to the alabaster fragment sample, the sample of encrustation with a near-surface layer of alabaster is characterized by a higher content of clay component. This is evident in the higher mass loss at the second (21.67%) and fourth (0.51%) stages of thermal decomposition, which corresponds to the release of interlayer and constitutional water by the clay minerals.

The DTA curve of the sample of encrustation with a near-surface layer of alabaster shows a negligible mass loss in the high temperature area of 753 – 1000°C. This mass loss corresponds to the combustion of soot present on the surface of the sample in the form of thick grime **Error! Reference source not found.**. Microphotographs of the stone surface confirm the presence of black patina on the surface of the stone.

Alabaster baptismal font from the courtyard of the Dormition Church in Lviv

The alabaster baptismal font is kept in the courtyard of the Dormition Church in Lviv. It dates back to the 18th century. The baptismal font consists of two parts: a basin and a pedestal (Fig. 16). It is made of grey banded alabaster. The baptismal font is whitewashed in several layers with a lime mortar, which formed a solid carbonized encrustation on the surface of the stone (Fig. 17).

The baptismal font was kept disassembled in the courtyard of the Dormition Church (the pedestal was separated from the basin). The Dormition Church is a working church in Lviv, built in 1591-1629; made of brick and ashlar stone, situated in the centre of Lviv, where a large number of toxic gases, combustion gases and dust is emitted.

Fig. 16. The baptismal font pedestal

Fig. 17. A part of the baptismal font basin

For a long time, the baptismal font served as a flowerbed, and was placed under the gutter. It was in no way protected from the direct sunlight, rain or snow. Because of water pressing on the stone surface, in the direction of the water flow thin furrows were formed (about 2 to 4mm). The surface became ribbed and uneven. Small internal cracks, confined to the clay impurities area, are noticeable (Figs. 18-20). Such microcracks are caused by hydration-dehydration cycles and changes of atmospheric temperature.

Fig. 18. Limewash

Fig. 19. Internal microcracks

Fig. 20. Furrows in the direction of water flow, the result of crystalline mass dissolution

Storage conditions of the baptismal font and anthropogenic factors led to a change of the aesthetic appearance of the monument and a change of functional purpose. The once polished, transparent surface of the stone is lost.

Microscopic study shows the clusters of contaminants with inclusions of various kinds - fibres, sand, clay, living organisms, and products of metal corrosion (Figs. 21 and 22).

Fig. 21. Micrograph of the dirt on the basin of the alabaster baptismal font

Fig. 22. Micrograph of limewash on the basin of the alabaster baptismal font

The microscopic study of alabaster fragment of the baptismal font in thin section revealed that the alabaster has a fine-grained structure of high crystal density. There are clay minerals impurities in gypsum crystals. The alabaster is dark grey with a brown tinge.

The results of microscopic studies correspond with the data of thermal analysis (Fig. 23 and Table 3).

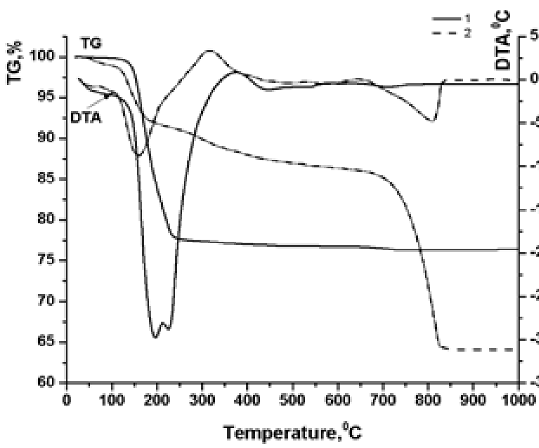

Fig. 23. TG and DTA curves of the baptismal font from the Dormition Church: 1 - alabaster fragment, 2 - encrustation with a near-surface layer of alabaster

Table 3. The results of thermal analysis of the alabaster fragment and encrustation with a near-surface layer of alabaster of the baptismal font from the Dormition Church

Sample	Stage	Temperature range, °C	Mass loss, %
Alabaster fragment of the baptismal font from the Dormition Church	I	20 - 110	0,23
	II	110 - 310	22,43
	III	310 - 408	0,35
	IV	408 - 622	0,21
	V	622 - 800	0,44
Encrustation with a near-surface layer of alabaster of the baptismal font from the Dormition Church	I	20 - 110	1,30
	II	110 - 246	7,59
	III	246 - 395	3,09
	IV	395 - 647	1,97
	V	647 - 860	22,05

Mass loss of the alabaster fragment (0.23%) in the temperature range 20 – 110°C, at the first stage of thermal decomposition, corresponds to the release of sample's adsorbed water. At the second stage of thermal decomposition, in the temperature range 110 – 310°C, the sample loses its mass (22.43%) due to the release of water of crystallization in alabaster. In this temperature range, interlayer water in clay minerals, present in the structure of alabaster, is lost.

The appearance of exothermic effect in the temperature range 310 - 408°C, at the third stage of thermal decomposition, which is accompanied by the mass loss (0.35%) of the sample, corresponds to the combustion of organic components of the sample.

The loss of water of constitution by clay minerals (0.21%) is observed at the fourth stage of thermal decomposition, in the temperature range 408 – 622°C. At the fifth stage of thermal decomposition, in the temperature range 622 – 800°C, the decomposition of the carbonate constituent of the sample takes place. Endothermic effects on the DTA curves accompany the mass loss processes at the different stages of thermal decomposition and the decomposition of the carbonate constituent [10, 19].

As opposed to the alabaster fragment sample, the sample of encrustation with a near-surface layer of the alabaster baptismal font has a considerable content of organic matter (3.09%), which combusts in the temperature range 246 – 395°C, at the third stage of thermal decomposition. The presence of the considerable number of organic contaminants can be explained by the fact that the baptismal font has been used for growing flowers for a long time.

In contrast to the alabaster fragment sample, the sample of the encrustation with a near-surface layer of alabaster contains a significantly higher number of carbonates, which decompose in the temperature range 647 – 860°C. During their decomposition, the considerable mass loss (22.05%) and appearance of a deep endothermic effect on the DTA curve take place. The sample also contains a small amount of calcium hydroxide, which decomposes together with the clay components in the temperature range 395 – 647°C. It is evidenced by a significant amount of mass loss (1.97%) of the sample of encrustation with a near-surface level of alabaster, as compared to the alabaster fragment sample at the fourth stage of thermal decomposition.

The sample of the encrustation with a near-surface layer of alabaster contains a significant amount of $CaCO_3$ and $Ca(OH)_2$. According to the micrography, it is due to the limewash on the surface of the stone.

Conservation problems

According to the findings of field studies and laboratory research, the features and problems of conservation of the alabaster products described above have been determined.

Therefore, the main problem is water in any state of matter. Water causes dissolution, corrosion, cleavage and cracking. To preserve the alabaster tombstone, it is important to install horizontal and vertical waterproofing around the perimeter of the object to prevent water from entering the stone. To preserve the baptismal fonts from the courtyard of the Dormition Church and The Pidhirtsi Castle, it is important to ensure stable climatic conditions, to store the fonts in a dry place, where the humidity does not exceed 60%. The problem of structural strengthening is that the stone, namely clay minerals, are supersaturated with moisture, thereafter their volume is to some extent increased. The stone must be dry in order to be strengthened. Hence, there is a problem of stabilization of water content in the structure of the stone. Elimination of biological damage causes a number of problems: to stop the growth of the root systems of mosses and vascular plants; to remove the remnants of organic matter from the pores, cracks, hollows without destroying the stone; to carry out antiseptic procedures to disinfect the deposits of biological material. The problem of removing surface contamination is to choose effective and non-destructive cleaning methods and materials. Thus, water, strong acids, alkalis and mechanical cleaning lead to the destruction of the crystalline mass. In addition, it should be noted that the damaged surface of the stone is vulnerable to any external factors [15, 16].

Conclusions

The study found that the influence of storage conditions of the monuments has comprehensive destructive effects on alabaster, changes the aesthetic appearance of the monument, in particular loss of colour saturation, loss of the polished surface stone and its transparency, loss of the characteristic sculptural form of the objects. The degree and nature of destruction depends on the composition of alabaster and storage conditions. A common feature of the consequences of stone structure destruction, under the action of natural atmospheric factors, is dissolution and corrosion of crystals and clay impurities, as well as formation of cracks and cleavage. Comprehensive thermal analysis revealed the form of contamination that occurs on the stone surface under the influence of external factors. Soot, dust, fibres, sand and clay were found in the surface layer of the crystalline mass of alabaster and in the objects located in the places with high air pollution. Surface contamination of the objects, which were situated in the park area with high humidity, contained a large amount of organic matter.

Field study and microscopic analysis outlined the destruction process of the alabaster structure and described the form of this destruction. Sudden temperature change leads to cracking, reduction in stone hardness, and cleavage. Rainwater causes dissolution of alabaster crystals, which in turn causes the formation of furrows in the direction of water flow; dissolution of stone surface; dissolution of stone, which leads to the spalling of large gypsum crystals from the alabaster crystalline mass; cracking, cleavage and formation of protruding areas in the form of encrustation and dirt outgrowth.

High humidity facilitates the growth of organic matter, which in turn destroys the crystalline mass; mold and lichens change the colour of the stone and destroy it. Humidity, wind, and chemicals that settle on the stone cause chemical and physical weathering.

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