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THE PROTECTIVE EFFECT OF GREASE STAINS CAUSED BY HANDS TOUCHING STONE RELICS

Yi-Min MA[1], Hui ZHANG[2], Bing-Jian ZHANG[1,2*], Mei-Feng SHI[3], Run-Ping ZHANG[3]

[1] Department of Chemistry, Zhejiang University, Hangzhou 310027, P.R. China.
[2] Department of Cultural Heritage and Museology, Zhejiang University, Hangzhou 310028, P.R. China
[3] Yungang Grottoes Cultural Research Institute, Datong 037007, P.R. China

Abstract

Many exposed surfaces of stone relics were covered by grease stains by tourists' touching or leaning. The grease stains have changed the appearance of stone relics seriously, so they are always supposed to be removed. However, the stones coated with grease always keep a better look than bare ones. In this work, a series of experiments were designed to explore whether grease can protect the stones. The results of SO_2 corrosion experiment showed that SO_2 had a corrosion effect on sandstones in the presence of water. Whereas the samples coated with grease almost remained unchanged after the corrosion test. From the salt destruction test, it was observed that grease stone samples were also less destroyed than blank ones. It means that grease stain can prevent the attack of SO_2 corrosion and salt destruction, which are the major factors of damage in Yungang Grottoes, China.

Keywords: *Grease; Stone relics; SO_2 corrosion; Salt destruction; Water absorption*

Introduction

There are many invaluable stone relics and monuments left behind by our ancients all over the world. However, a large amount of stone relics, which are large, immovable and unprotected, are exposed in the natural environment. As a result, these stone relics have been damaged to a varying extent and received increasing interest in recent years [1-4]. For example, Yungang Grottoes in Datong, China, listed by UNESCO as World Heritage Site, consist of 252 caves and more than 51000 sandstone statues. These stone relics represent the outstanding Buddhist grotto art of the 5th to the 6th century in China. Our investigation and analysis in Yungang Grottoes showed that many stone statues were covered with grease stains. These grease layers, which were caused by tourists' touching and leaning for a long time, were polished, black and shiny. The proportion of grease stain caused by touching in Yungang Grottoes is about 0.33% in the total contaminated surface, with an area of about 33m^2 [5].

Figure 1 shows a statue on the east wall of No. 40 cave in Yungang Grottoes. The statue has been weathered seriously, especially the lower part covered with dust. The lines of clothing became blurred and even invisible with large quantities of salt efflorescence on the surface.

* Corresponding author: zhangbiji@zju.edu.cn

However, it is interesting that both sides of the lower statue (Fig. 1B and 1C), which are coated with a thin layer of dark grease, seem to keep a better look. In view of this, it is supposed that grease caused by longtime touching may have a protect effect on these statues.

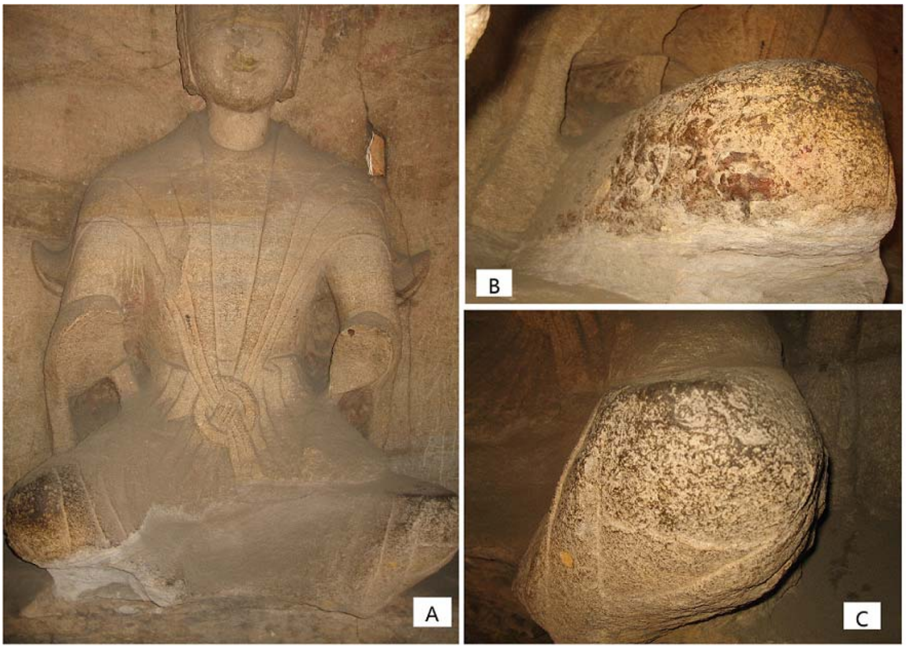

Fig. 1. Image of the statue: A - the statue on east wall of No. 40 cave in Yungang Grottoes, B and C - the left and right sides of the statue.

People love to touch the religious statues to show their respect, and believe that touching has brought them great fortune. Therefore, a lot of Buddha statues in Yungang Grottoes have been touched. Those parts, which are relatively prominent and easy to touch, were always covered with grease stain. One example is the Yuan Dynasty statues outside the Lingyin Temple (Fig. 2 and 3). Lingyin Temple, one of the famous Chinese Buddhist monasteries, is located in the northwest of Hangzhou, China. The Flying Peak Hill outside the Lingyin Temple is famous for numerous stone sculptures and inscriptions. The statue in figure 2 is at the foot of the Flying Peak Hill. Because of visitors' touching, the statue appears to be smooth, oily and shining. The facial features, crown and clothing lines are clearly visible (Fig. 2B). However, most of the Yuan Dynasty statues are located in top high caves, out of reach, weathered seriously and thus became blurred. These stone statues degraded seriously because of acid rain corrosion, salt efflorescence and environmental changes. Figure 3 shows the sitting statues in Qinglin Cave on the top hill, which were severely corroded.

Whether these contaminants are harmful to the stone relics and how to remove them, if they do harm, are fundamental scientific concerns for conservators. These grease stains changed the artwork's original appearance and reduced the artistic value of cultural relics. Therefore, according to the traditional intervention criteria for cultural heritage, they should be removed [6-8]. However, the grease stains are also the historical evidence of human activities and part of the relic's original conditions. If these stains are proved to be unharmful, they should not be removed actively according to the minimal intervention principle. Moreover, oil and waxes had been used traditionally to protect stone surfaces, according to the record by Herodotus in 4th century BC [9, 10]. In modern times, people also tried to use oil and waxes as coatings to protect stone surface against salt corrosion [11, 12]. There are also many examples showing that oil and waxes have been used in ancient paper, pottery and teeth protection [13-17].

Fig. 2. The Yuan dynasty Buddhism statue in the foothills of the Flying Peak Hill

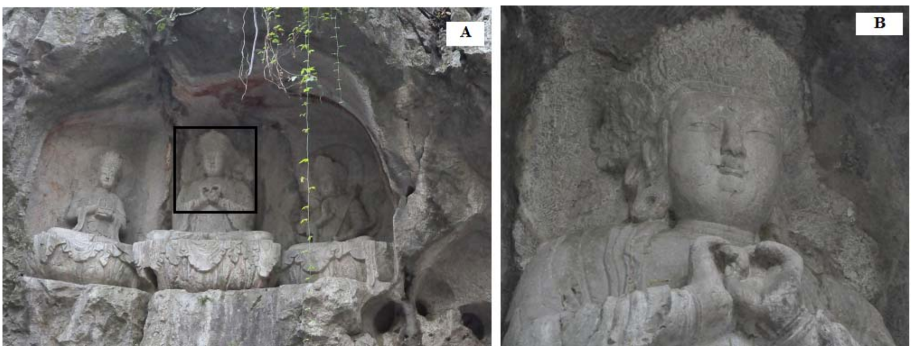

Fig. 3. The three saints of Huayan's sitting statues above the hole of Qinglin Cave.

Now the question is, do the grease stains, caused by touching, really have an effect to resist atmosphere pollutants' corrosion and reduce natural weathering's threat? In this paper, a series of experiments, taking sandstone as example, were carried out in the laboratory to verify whether grease has a protective effect on stone relics. In addition, the major destructive factors were investigated to explain the protective mechanism.

Experimental

Samples and Instruments

On-site sampling and observation

Real samples coated with grease were taken from Yungang Grottoes. These samples were observed using 3D digital microscope (Keyence VHX-1000), SEM (SIRION-100, FEI America) and polarizing microscope (Nikon Eclipse E600 POL).

Model samples preparation

Lanolin was selected for the imitation of grease stain caused by touching. Lanolin is a natural substance secreted by the sebaceous glands of sheep. Compared to body oil, it has analogical structure and properties [18].

Sandstones from Yungang area were used for the model sample preparation. Yungang sandstone is composed of quartz, orthoclase and phyllosilicates, with quite different percentages of SiO_2 (ca. 70%) and CaO (ca. 0.55%) [19]. Mineralogical composition is rather heterogeneous, due to the presence of various amounts of lithic clasts: Fe_2O_3 (4.66%), Al_2O_3 (14.95%), K_2O (2.54%), TiO_2 (0.32%). Carbonate cement is universally more than 10% in sandstones with pack-pore cementation [20, 21].

Stone samples were cut into rectangular blocks of 5×5×3cm. Each specimen was washed with deionized water, and then kept at 60±2 ºC until a stable weight was recorded. The fresh samples obtained by this method were also used as the blank samples for comparison. To prepare grease samples, lanolin was heated in water-bath and smeared evenly on the stone surface.

SO_2 corrosion test

According to the environmental conditions of Yungang area, a SO_2 corrosion test chamber (Fig. 4) was designed for our study [22].

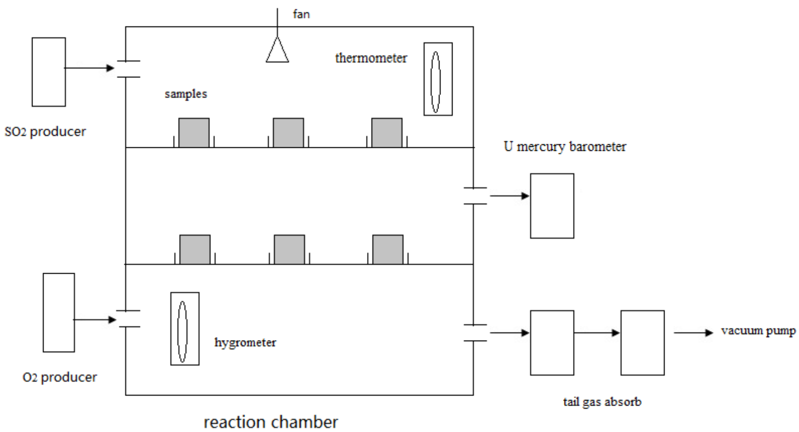

Fig. 4. Diagram representing SO_2 corrosion test chamber

The main component of reaction chamber was made of 8mm-thick organic glass, which was divided into several parts to place stone samples. In the chamber, a thermometer and a hygrometer were placed to monitor the reaction conditions. A fan was equipped to distribute the gas evenly. Two pipes were installed to connect SO_2 and O_2 producers. On the other side one pipe was installed to connect a vessel containing 15% solution of alkali (e.g. sodium hydroxide), which was connected with a vacuum pump. A U-shape mercury barometer was also used to keep a negative pressure between inside and outside.

SO_2 and O_2 were prepared by the following reactions:

$2NaHSO_3 + H_2SO_4$ (concentrated) $\rightarrow Na_2SO_4 + 2SO_2\uparrow + 2H_2O$

$2H_2O_2 \rightarrow 2H_2O + O_2\uparrow$ (MnO_2 as the catalyst)

There were three steps in SO_2 corrosion test: wetting, corrosion and drying in order to mimic the SO_2 reaction process with stones in the caves. SO_2 can react with stones at the presence of water and O_2 to generate acid. When water evaporates, sulfate will be produced on the stone surface. In the wetting step, model samples were immersed in a water container for half an hour. Water level was kept at about 3mm above the sample bottom. Water was adsorbed and transported in the stone samples and made the sample wet. In the SO_2 corrosion step, samples were placed in the corrosion chamber for 4h. 2/5 of the fresh sample was covered with plastic wrap to avoid any other reaction occurring on this part. In this study, 20,000 times higher concentration (1.86×104mg/m^3) than the daily average SO_2 concentration per year in Yungang grottoes' air (0.093mg/m^3) was used to effectively accelerate the process of SO_2 corrosion. A small quantity of atmospheric air was pumped into the test chamber to keep the air flowing and maintain a 5mm negative pressure in the chamber. The reaction conditions of the chamber were shown in Table 1. After the corrosion step, the samples were then dried for 10h at 50ºC.

Table 1. The reaction conditions of the SO_2 corrosion test chamber

Environmental characteristics into the test chamber	Value
Temperature (ºC)	25
Relative Humidity (%}	>20
Pressure (mmHg)	-30
Concentration of SO_2 (mg/m^3)	1.86×10^4

Several groups of model samples were investigated in different tests to understand the effect of destructive factors. In each group, three blank samples and one grease sample were numbered A, B, C and D, respectively. Samples A and D experienced all the steps; while sample B experienced only SO_2 corrosion and drying steps, and sample C experienced only wetting and drying steps. The whole experiment was designed to repeat 45 times and the weights of these samples were recorded at each time.

Salt destruction test

Yungang area is under the continental monsoon half dry climate with the diurnal range of temperature up to 20ºC and annual average precipitation amount of 330 ml. In the whole year, the main rainfall period is between July and September and the humidity is relatively low in other seasons [23]. The accumulation of salt on the surface of stones is closely related to the annual seasonal change. During the summer time, the temperature is lower inside the caves with higher humidity. As a result, condensed water will form on the cooler stone surface and adsorb acid gases (SO_2, etc.) in the air to generate dilute acid solution. During winter, the temperature is higher inside the caves, therefore, salt solution can be transported toward the outside surface and crystallization will occur at lower temperature [24]. According to our on-site investigation, the area of salt damage in Yungang Grottoes is about 2236m^2, which is 31% of the total disease area.

According to these features, salt destruction experiments on five groups of both blank and grease sandstone samples were performed, using the orthogonal experimental design, to verify the effects of grease on salt damage. In this test, 1/3 of the four sides of sample were covered with plastic wrap, and lanolin was spread evenly onto the front surface (5×5cm^2). In the orthogonal experiment, two samples from both blank and grease samples, which were numbered as B4, B5 for the blank stones and 2d, 2e for the grease stones, were placed on metal net which was entirely immersed in sodium sulfate solution for 4.5h and then kept in freezer at -40ºC for 2.5h. Then the samples were placed in vacuum drier at 60ºC for 2h. After that, the samples were sprayed with 5% sulfuric acid (mimic the acid rain and fog); 20min later, the samples were placed in vacuum drier at 40ºC for the rest hours in a 24h cycle, and the weights were recorded. This cycle continued until the surfaces of the stone samples were destroyed or the obvious weight loss was observed. Another three samples from both blank and grease stones, which were numbered as B1, B2, B3 for the blank stones and 2a, 2b, 2c for the grease stones, were undergone the cycle of destruction experiments except one factor, which was, for B1 and

2a, the salt solution soak step was neglected, for B2 and 2b, the freeze-thaw step was neglected, and acid solution spraying was neglected for B3 and 3c.

Capillary water absorption

Capillary water adsorption test was performed to quantitatively investigate the hydromechanics change of stones coated with grease. The amount of water absorbed by capillarity Q (g) is defined as: $Q = CAt^{0.5}$, where $C(g/(m^2s^{0.5}))$ is the water absorption coefficient of the sample, A (m^2) is the water contact area, and t (s) is the water adsorption time. The European Standard EVS-EN 1925(2001) [25] was applied to determine sandstone capillary water absorption. All the samples were dried until constant weight (Q_0) at 50ºC and then immersed in the container with the water level of about 3mm above the bottom. At increasing time intervals (1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 15, 20, 25, 30, 40, 60, 80, 100, 120, 140, 160, 180, 240, 300min), the samples weights were recorded and subtracted Q_0 to get the mass of adsorbed water Q. By fitting the Q～t curve, water absorption coefficient C can be obtained.

Results

Real sample analysis

Real sample from Yungang Grottoes had shiny and smooth surface, which was caused by black-brown grease coating. Figure 5-1 shows the 3D digital microscopic picture of real sample. Yellow part on the left side of the figure was sandstone bulk grey part on the other side was grease layer. It suggested obviously that grease had already penetrated into sandstone surface layer binding tightly to the stone.

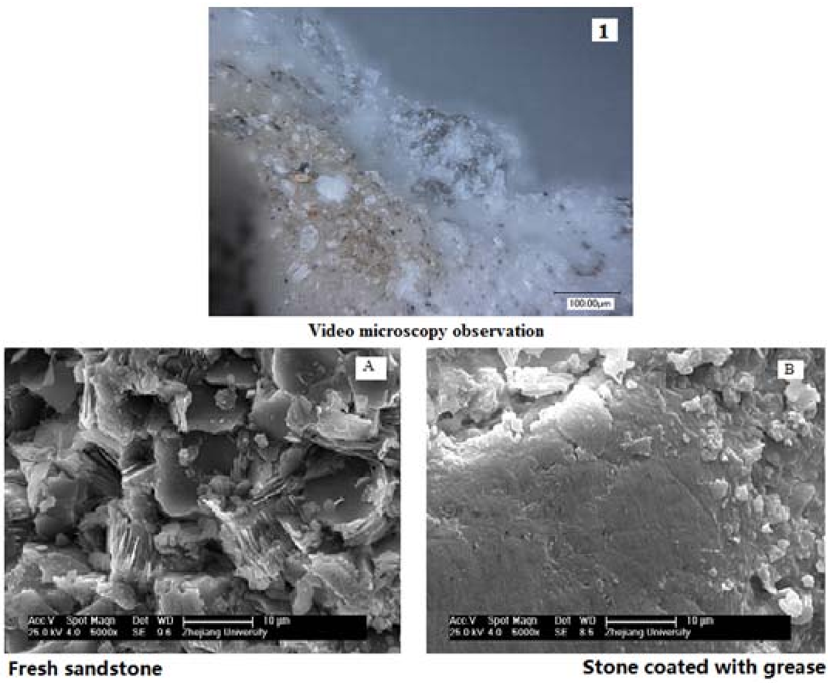

Fig. 5. Microscopic observation and analysis of real sandstone samples from Yungang Grottoes

Figure 5-A and 5-B show the SEM pictures (5000X) of the blank sandstone and real samples from Yungang Grottoes respectively. The lamellar structure of sandstone crystal can be

seen in the blank sample. But the surface of real sample was covered with a compact film, filling the voids among crystals to form almost smooth surface.

It can be seen from the polarized microsectile (cross-section) pictures (Fig. 6) that the grease stain generated by touching was brown continuous film evenly covering the stone surface.

For unweathered rock particles, such as quartz, the thickness of grease film was less than 10μm (Fig. 6-1a and 6-1b). In soft part of rock, grease could penetrate several hundred micrometers into micro pores (Fig. 6-2a and 6-2b). Grease film was very thin in the bulges of rock, while in the rock hollows the grease film was thick, filling the voids and forming a flat surface (Fig. 6-3a and 6-3b). The microstructures of the model samples were quite similar to the real samples with grease stains.

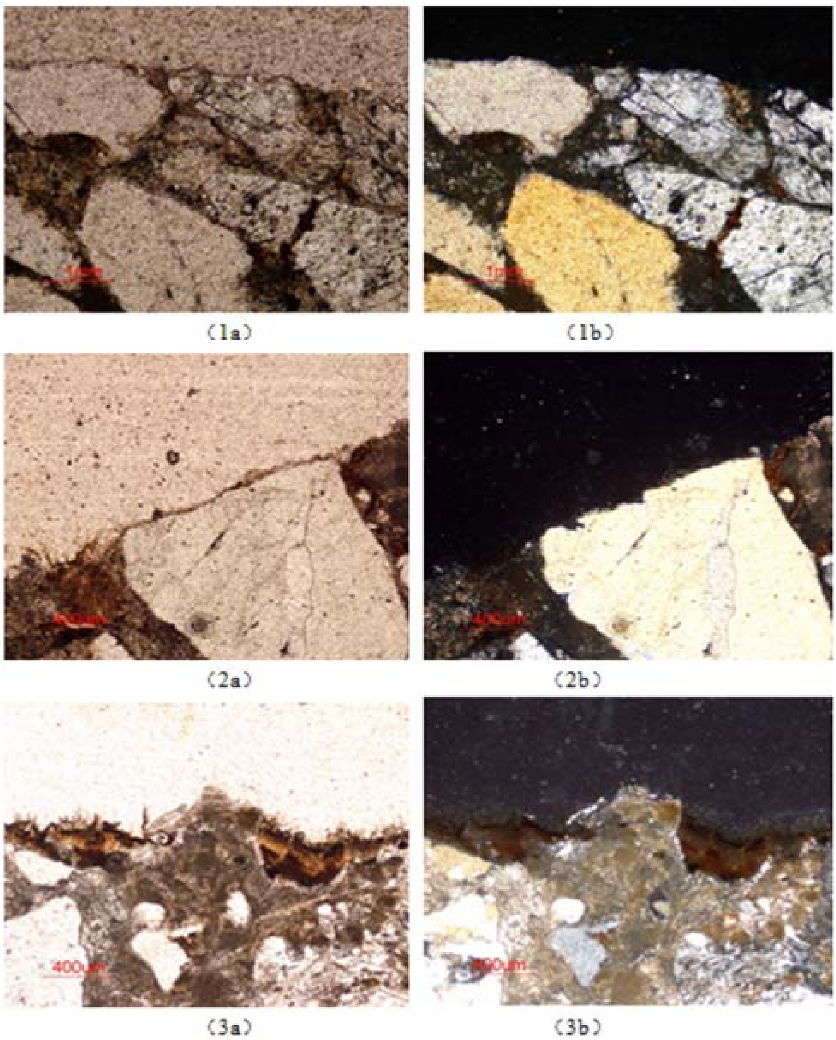

Fig. 6. Polarized cross-section images of the grease stains

SO_2 corrosion test

The results of SO_2 corrosion test are shown in figure 7 for three groups of blank samples, respectively. The mass of samples in group A, experiencing wetting, SO_2 corrosion and drying, rose gradually during the first 10 cycles and then dropped suddenly, which indicated obvious corrosion phenomenon during the latter part of test. During these cycles, SO_2 reacted with the stone material to produce sulfate, as a result, the mass of samples increased in the beginning. When there was enough sulfate under wet condition, salt crystallization occurred to cause severe damage. Then the stone samples broke and the mass dropped dramatically. The samples lost 25% of their mass at the 45th cycle meaning that they had degraded severely. The mass of samples in group B, experiencing only corrosion and drying, almost remained the same weight during the process. It suggested that SO_2 was not converted into sulfuric acid without the wetting step therefore, the corrosion didn't happen. The mass of samples in group C neglecting SO_2 corrosion, increased a little bit. So SO_2 plays an essential role in the sandstone corrosion and the control of SO_2 content in the atmosphere is important for the protection of grottoes.

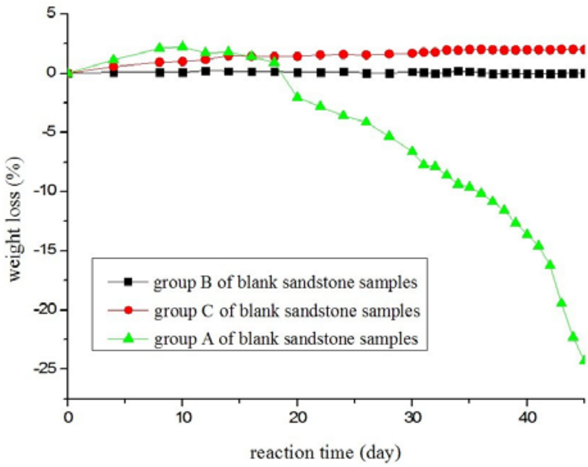

Fig. 7. Weight loss versus reaction time for three groups of blank sandstone samples in the SO_2 corrosion test.

The weight losses of blank samples (group A) and model samples coated with grease (group D) were plotted against the corresponding reaction time and shown in Fig. 8. The mass of samples in group D almost remained the same during the whole corrosion process, which indicated little corrosion.

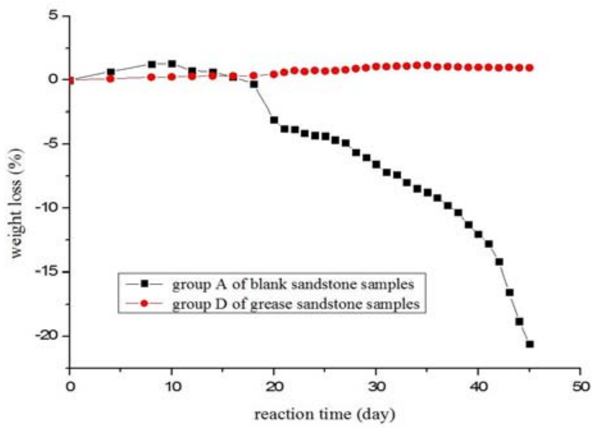

Fig. 8. Weight loss versus reaction time for blank and grease sandstone samples in the SO_2 corrosion test.

The pictures of blank and grease sandstone samples after SO_2 corrosion test are shown in figure 9 with fresh sandstone as a reference. The sandstone sample coated with grease almost remained unchanged, while the blank sandstone sample was broken down. The result suggested that grease might protect the sandstones from SO_2 corrosion.

Fig. 9. (Left) fresh sandstone; (Middle) blank sandstone sample taking part in wetting, SO_2 corrosion and drying tests; (Right) grease sandstone sample taking part in wetting, SO_2 corrosion and drying tests

Salt destruction test results

The cycle of destructive test lasted for 56 days. The phenomenon of salt crystallization was visually observed, as shown in figure 10. Surface of B1 and 2a sandstone samples, which didn’t take part in salt solution soaking, changed slightly. However there were large amounts of salt crystals on the surface of other blank samples, and further destruction was shown obviously at the end of the cycle. Sample B3 was the most severe with stone pieces peeling off the surface. There were also some fragments peeling off the surface of B2, B4, B5. On the contrary, sandstone samples coated with grease remained almost unchanged, while only a few salt crystals could be observed. Salt could destroy the structure of sandstone by transporting and crystallizing. However, grease stains may resist the salt damage according to our results.

Fig. 10. Picture of sandstone samples after salt destruction cycle

Capillary water absorption

The sandstone coated with grease showed very different pattern of water absorption compared to blank sandstone (Fig. 11). The water absorption coefficient of blank sample, calculated as $C = Q/(At^{0.5})$, was C_0=14.53 g/($m^2s^{0.5}$), while grease sandstone had a lower water absorption coefficient C_1 = 3.533 g/($m^2s^{0.5}$). The greased sandstone showed great reduction in both absorption speed and total water absorption capacity. So the presence of grease could reduce the water absorption coefficient of sandstone.

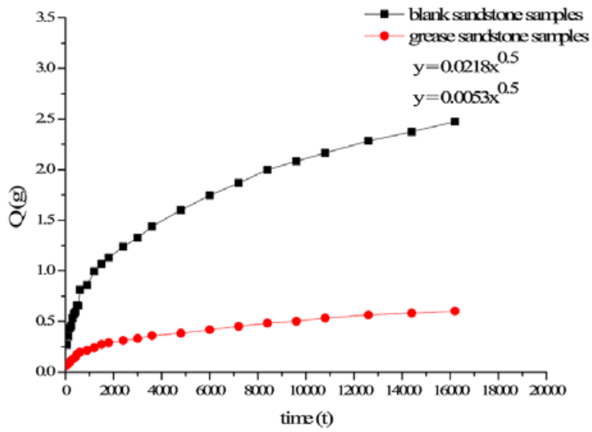

Fig. 11. Amount of water absorbed (g) by capillarity as a function of time (s) in blank and grease sandstone samples

Discussion

As can be seen in the real sample analysis, grease caused by touching formed a continuous and stratified film with a thickness from 10 to several hundred micrometers, which covered stone surface evenly and tightly. The physical properties of stone surface have changed due to the grease stain, such as the porosity, cementation, permeability and water-absorption. Our experiments focused on physical properties of the affected stone surface, so the possible chemical reaction between grease and stone was neglected. The chemical content and structure of lanolin used in the experiment were similar to but not strictly the same as real grease on stone relics.

It is verified that SO_2 could corrode stones. The mass of group C, experiencing wetting-drying cycles only, was almost unchanged. While the samples of group B, experiencing SO_2 corrosion only, showed slight increase in the mass. It is because that the carbonates in rocks could react with SO_2 to form sulfates, which is heavier than carbonates. Whereas comparing group A to the other two groups, SO_2 can corrode rock seriously with the presence of water. SO_2 could dissolve in water to form dilute acid solution which has stronger corrosion effect than SO_2 itself. In the corrosion process, both water and SO_2 are necessarily needed. Compared with blank samples, the grease samples remained better and the weight losses were less after the SO_2 corrosion test. In other words, grease could reduce SO_2 corrosion.

Sandstone deterioration results from continued precipitation of salt crystals, which is caused by evaporation of water from the sandstone surface. Crystallization pressure of salt is one of the major factors that may induce serious damage to stone relics [26, 27]. When grease penetrated into sandstone and filled the pores, the amount of salt crystals on the surface decreased so that their crystallization pressure was relieved. On the other hand, salt could precipitate on the grease surface and salt crystal fragments could be seen on the grease. As a result, the damage by salt crystallization pressure could be reduced.

Almost all the chemical and physical damages in the stone microstructure are related to water, since water is always the medium of various destruction factors. For example, water is indispensable for the conversion of calcite into gypsum during the SO_2 corrosion process. The hydration and expansion of gypsum are also related to water. From the results of water adsorption test on blank and grease stone samples, it can be seen that grease reduced the damage both by SO_2 corrosion and salt crystallization. It is because that grease changed the hydromechanics of stone surface. The water adsorption coefficient of grease samples declined about 3/4, and the water contact angle was increased from 15° up to 70°. As a result, SO_2 was not easy to be converted into sulfuric acid on the stone surface due to less amount of water. Grease could also reduce the moisture adsorption on the surface so that the adsorption of sulfuric acid was also reduced. Therefore, the acid corrosion and gypsum formation could be diminished. Moreover, thin film of grease was still a little bit hydrophilic, which could keep the access of salt transport and prevent the damage by hydrophobic/hydrophilic interface [28], but its mechanism is still unknown.

Conclusion

Grease stains caused by touching were found to have a protective role for stone relics. SO_2 corrosion and salt destruction experiments were carried out to prove and explain the mechanism of such protection. It was observed from corrosion test that grease coating could resist SO_2 corrosion. The salt destruction test showed that grease could also reduce the salt crystallization damage.

In the practice of stone relic conservation, grease stains caused by touching are usually considered to be harmful and dangerous because of their ugly appearance. Thus, grease stains are supposed to be removed. In this paper we showed that grease stains might be retained, because they could prevent the corrosion caused by acid and reduce the deterioration caused by salt. We suggest that some grease stains caused by touching stone relics should be preserved during the conservation procedures.

Acknowledgements

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