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A STUDY OF ACRYLIC-BASED MORTAR FOR STONE REPAIR

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Abstract

Repair of altered stone using mortar is an interesting approach, if it avoids a replacement and extends the lifetime of the original stone. In this paper we examine the properties of an acrylic dispersion-based mortar that had been developed in the late seventies for stone repair. The key features of this type of mortar are an easy adaptation to the stone color, a good workability and reversibility in an organic solvent. A mortar was prepared from stone powder and a dispersion of methacrylic ester-acrylic ester copolymer, and the flexural strength, capillary water absorption, hydric swelling and thermal expansion were measured and compared with the stone it aims to repair. The mortar has a low elastic modulus and a relatively low thermal expansion that avoid this repair material from damaging the original substrate. The high hydric swelling is compensated by the low elastic modulus in wet conditions.

Keywords: repair mortar, acrylic dispersion, reversibility

1. Introduction

In historical buildings, repair of altered stone using mortar is an interesting approach, if it avoids a replacement and extends the lifetime of the original material. In this paper we examine the properties of an acrylic-based mortar that had been developed in the late seventies in the Ecole Polytechnique Fédérale de Lausanne in Switzerland, and applied on the townhall. After more than thirty years, the durability of these mortars and their property of reversibility have relaunched the interest of stone carvers.

The repair mortars (thereafter also called artificial stone) are made by mixing a stone powder, in which the smaller grain fraction has been removed, with a dispersion of methacrylic ester-acrylic ester copolymer. The dispersion represents around 20% of the weight of the stone powder. The hardening of the mortar is due to the drying of the dispersion and the subsequent coalescence of the polymer particles. Due to the drying time, it is better indicated for repairs of degradations in the range of centimeters.

Alterations in this size range are commonly found in the weathering pattern of molasse sandstone, which was widely used in the construction of historical buildings in Switzerland. The molasse is a sedimentary stone mainly composed of quartz and felspaths cimented by calcite and clays (Kündig 1997), and is very sensitive to wetting and drying cycles. These cycles commonly lead to spalling of flakes of 0.5 to 3 cm (Furlan 1983 ; Jimenez-Gonzalez 2008). The stone is however in good conditions above this limit. A repair of these weathered stones with natural ones would imply a removal of potentially sound original material on at least 8 cm, to ensure a good placement, while the use of an artificial stone would save the original material. This approach is particularly relevant when applied to finely worked stone that can be repaired instead of being replaced by a new stonework. Examples of application are presented in **Fig. 1**.

Fig. 1 (a) Common dimension of spalling in a molasse sandstone - (b) and (c) Application of the artificial stone as a repair material.

An interesting feature of this mortar lies in its ability to be worked like a stone, and thus makes its integration possible by the normal tools of a stone carver. It also has the peculiarity of being solvable in an organic solvent (see **Fig. 2**). This makes the mortar easily removable without damaging the underlying original material and facilitates further treatments that could be applied to the stone for its conservation. It is interesting to point out that these features are still observable after thirty years of use in the townhall of Lausanne However, in some places the two materials show a differential erosion that could be attributed to a different durability or to a stress due to the mortar on the natural stone. This calls for an investigation of the underlying mechanisms controlling the compatibility of such mortars with the original stones they mimic. This article presents the first results made on this artificial stone, through the study of the elastic modulus, the capillary absorption, the hydric swelling and the thermal expansion of a natural stone and on the mortar made with it.

Fig. 2 The mortar is still reversible after thirty years.

2. Experimental

The experiments were aimed at examining the possibilities of repairing a molasse sandstone coming from the region of Bern, the blue Ostermundigen. We thus performed work both on this stone and on the artificial stone considered to be used as repair material. The later is made of powder obtained by grinding the natural stone. This powder is sieved and only the fraction higher than 0.125 mm is used. By doing this it is intended to obtain a similar granular skeleton to the stone but remove fines that would pack in between the larger grains and cause a much denser and smoother surface appearance than the molasses typically have.

The sieved powder is mixed with a commercially available aqueous dispersion of methacrylic ester-acrylic ester copolymer. The samples were then let to dry at room temperature during at least four weeks. This allows for film formation of the dispersion and causes the hardening of this type of artificial stone.

3.1 Color measurement

The color of the wet and dry stones are measured by a spectrophotometer Konica Minolta CM-700d with illuminant D65 and 10° angle, through an aperture of 8 mm, L*a*b* space is calculated. The samples were plates of natural and artificial stone of 5*20 cm^2. It was determined that a minimal number of six measurements should be taken to obtain a representative color of the samples (Giachetto 1994).

After drying, the difference between the artificial and the natural stone is hardly distinguishable by naked eye. However, a slight darkening of the artificial stone can still be perceived in the dry state. This can be explained by the fact that the finer fraction of the grains, which is removed, is lighter than the higher fraction, and thus induces a darkening of the mortar. This difference is attenuated when the stones are wet, suggesting it is an issue of scattering. This tendency is shown in **Table 1**; a just noticeable color difference is defined as a difference of 2.3.

Table 1. Color difference ΔE* (CIE1976) between the natural and the artificial stone.

dry	5.6
wet	2.8

3.2 Capillary water absorption

The tests were performed using at least three cylindrical samples of 4.5 cm diameter and 2.5 cm height for each material, dried to constant mass at 105°C beforehand. The samples were hung from electronic balances having a resolution of 0.01 g and placed in contact with water in a closed container to avoid the evaporation of water. The water uptake was monitored continuously by a computer and the capillary absorption coefficient (in $mg.cm^{-2}.min^{-0.5}$) was calculated from the slope of this initial water uptake (see **Fig. 3**) and are reported in **Table 2**.

Table 2. Sorptivity coefficients of the natural and artificial stones. For the natural stone the orientation of the bedding with respect to the direction of rise is given.

Sample	**Sorptivity** $mg.cm^{-2}.min^{-0.5}$
Natural ⊥	73.7
Natural //	82
Artificial	9.86

The two materials have very different absorption kinetics. The natural stone is fully impregnated in the range of minutes while the artificial stone needs days.

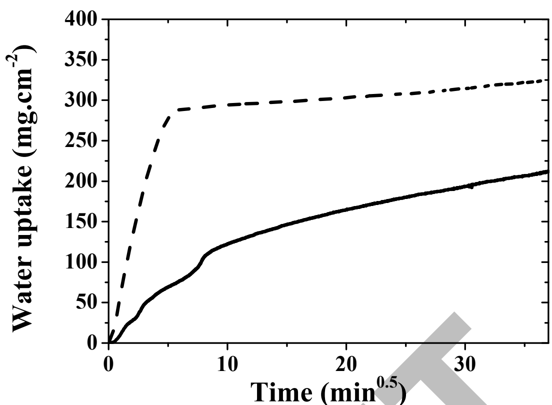

Fig. 3 Capillary sorption of the artificial stone (full line) and the natural stone (dash line).

3.3 Hydric expansion

The linear hydric expansion was measured by a LVDT displacement sensor from TESA with a resolution of 0.1 µm, placed on aluminium tips on both ends of the sample to avoid any movement of the sensor. Three samples for each material of dimension of approximately 6 cm were prepared for the test. They were immersed in water and their expansion continuously recorded.

Both materials swell when they are in contact with water. The swelling of the stone is attributed to the presence of swelling clays (Steiger 2011) while the swelling of the mortar is related to the swelling of the polymer. These processes probably have different time frames, but the difference in sorptivity certainly plays a key role in the difference of swelling kinetics. It can be noticed that the artificial stone swells more than the natural stone and the consequences of this are examined further in the paper.

First of all there is a big difference in the swelling kinetics (see **Fig. 4**). Only a few minutes are needed for the stone to swell up to 1.5 mm/m, a few days are needed for the mortar to reach a value of 2.1 mm/m. The values are reported in **Table 3**. This value is due to the spontaneous water absorption, but when water is forced to enter (by vacuum impregnation) the swelling can go up to 5 $mm.m^{-1}$ (not shown here).

Sample	**Hygric swelling** ($mm.m^{-1}$)
Natural ⊥	1.46
Natural //	0.69
Artificial	2.1

Table 3 Values of free swelling strain.

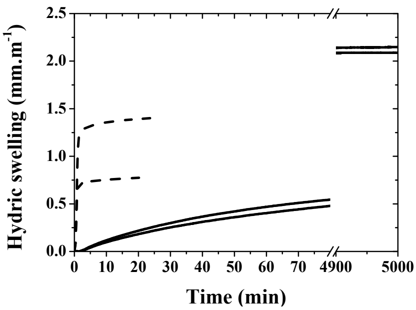

Fig. 4 Hydric swelling of the natural stone (dash line) and the artificial stone (full line).

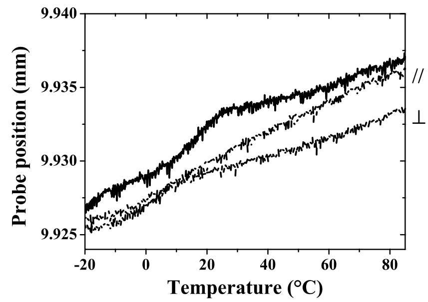

Fig. 5 Thermal expansion of the artificial stone (full line) and the natural stone (dash line). The dependency of the thermal expansion with the bedding for the natural stone is reported.

3.4 Thermal expansion

The thermal expansion was measured for both the artificial and the natural stone by Dynamic Mechanical Analysis in a Perkin Elmer DMA 7e, on cubes of 1 cm^3. The thermal range was -30 to +90°C with a heating rate of 1°C.min^{-1} under nitrogen atmosphere. Dilation curves are reported in **Fig.** 5 and the average expansion coefficients are given in **Table 4**.

Table 4 Thermal expansion at 20°C.

Sample	**Thermal expansion** 10^{-6} K^{-1}	
Natural ⊥	12.9	
Natural //	8.9	
Artificial	-20 – 10°C	8.6
	10 – 25°C	19.5
	25 – 60°C	4.9

While acrylics are reported to have high thermal expansion values (Brady 2002), the expansion of the artificial stone is only slightly higher than the Ostermundigen stone. This is because most of the mass of this artificial stone is made of stone powder and not of polymer. However, due to the glass transition (29°C) the thermal expansion is not linear over the temperature range studied, and shows its higher value, 19.6 $°K^{-1}$ in the range of 10 to 25°C.

3.5 Flexural strength test

The flexural strength test provides the flexural moduli, the flexural strengths and the critical strains of our materials. For this we performed a three-point bending test on a Zwick 1454 10kN, with a span of 12 cm, for both dry and wet artificial and natural stone. The dimensions of the samples were 1*5*16 cm^3, giving a span to thickness ratio of 12:1. Due to the very different plasticity behavior of the materials, the load rate was 1 $mm.min^{-1}$ for the artificial stone and 0.5 $mm.min^{-1}$ for the natural stone. At least five samples for each material and conditions (dry, wet, perpendicular and parallel to the bedding for the natural stone) were used. The flexural strengths refer to the maximum strengths developed by the samples during loading. The elastic moduli are calculated using the slope of the initial elastic part of the stress-strain curves, using the following relation:

$$E = \frac{L^3 F}{4wh^3 d} \quad (1)$$

with *L* being the span, *F* the applied load, *w* and *h* respectively the width and the height and *d* the deflection at maximum load.

In the beam bending theory, the critical deflection, meaning the deflection reached at maximum load (not necessarily the deflection at break) can be calculated as following:

$$\varepsilon_c = \frac{600h}{L^2} \cdot d \quad (2)$$

The artificial stone shows a very high critical deformation compared to the natural stone and a low flexural modulus as can be seen in the **Fig. 6**, and of which the consequences are discussed in the next section.

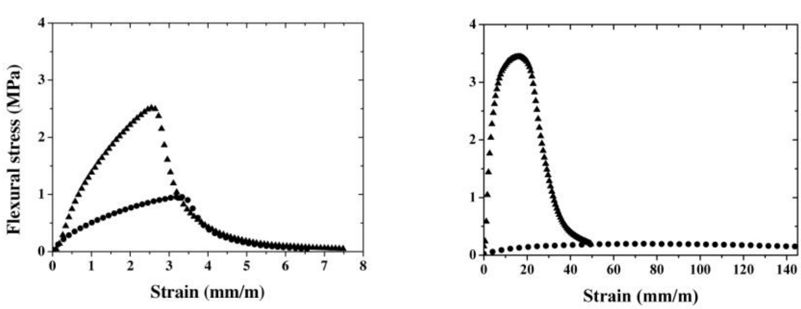

Fig. 6 Strain-stress curves of the natural stone (left) and the artificial stone (right). The triangles represent the dry state while the circles represent the wet state.

The results are presented in **Table 5**.

Table 5. Mechanical characteristics of the natural and artificial stone determined by three point bending.

	Ostermundigen blue			Artificial stone	
	Bedding	Conditions		Conditions	
Flexural modulus (GPa)	⊥	dry	1.87	dry	0.75
		wet	0.43		
	//	dry	1.37	wet	0.01
		wet	0.36		
	Bedding	Conditions		Conditions	
Flexural strength (MPa)	⊥	dry	2.44	dry	2.82
		wet	0.94		
	//	dry	1.87	wet	0.18
		wet	0.66		
	Bedding	Conditions		Conditions	
Critical deformation (%)	⊥	dry	0.28	dry	1.92
		wet	0.36		
	//	dry	0.30	wet	7.23
		wet	0.29		

3. Discussion

The present part focuses on the possible mechanical damage provoked by the differential response to a hydric or thermal stimulus. The question to be answered is whether the expansion of the artificial stone can damage the original material. We examine mainly this case as it would put the natural stone in tension, which is much more critical than putting it in compression.

The stone after repair can be seen as a juxtaposition of two layers that expand. In the simplest case, we consider that the stress is homogenous in each layer. In this simplified situation the equilibrium position in case of differential dilation is satisfied by the following equation:

$$\sigma_m h_m = \sigma_s h_s \tag{3}$$

where σ_s and σ_m are the stresses respectively in the natural and the artificial stone (mortar), h_s and h_m are the thicknesses of the corresponding layers. The situation is schematized in **Fig. 7** .

By using the Hooke's elastic law, we can write:

$$E_m(\varepsilon_m - \varepsilon)h_m = E_s \varepsilon h_s \tag{4}$$

with E_s and E_m respectively the flexural modulus of the stone and the mortar, ε_m the free swelling strain of the mortar and ε the final strain of the bilayer composite. It is also the strain felt by the stone layer.

This leads to:

$$\varepsilon = \frac{1}{\frac{E_s h_s}{E_m h_m} + 1} \varepsilon_m \tag{5}$$

The applications of artificial stone that we are considering concern 1-2 cm layers applied on top of natural stone of which the degraded outer surface was removed and for which an overlay is needed. This means that for most cases we can consider $h_s >> h_m$. Moreover, for all situations involving wetting the modulus of the artificial stone is very low. This means that the stress equation (5) reduces to:

$$\varepsilon \cong \frac{E_m h_m}{E_s h_s} \varepsilon_m \tag{6}$$

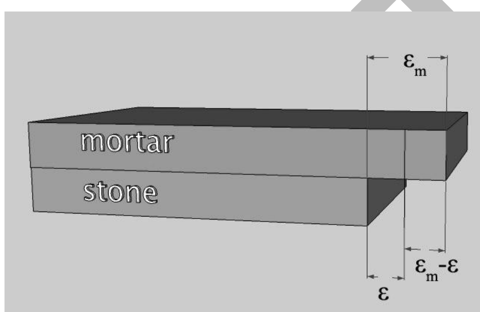

Fig. 7 The expansion of the mortar and the induced expansion ε of the underlying stone.

As mentioned above, we should have $h_s >> h_m$. However, even if $h_s = h_m$, we find that the strain resulting from (6) remains below the elastic limit of the stone. Therefore, we do not expect the wetting of the artificial stone to damage the underlying one.

If the underlying stone gets wet it would partially expand. This would reduce the value of ε_m to be used in (6). However, one would use the elastic modulus of the wet stone which is lower and counter balances this. The natural stone is then strained slightly more than previously. Even then, this remains well below the elastic limit (0.04 $mm.m^{-1}$ max versus 0.6 $mm.m^{-1}$).

The same calculations show that the layer of artificial stone would basically be compressed and almost fully prevented from expanding. This comes from its low elastic modulus in the wet state. Moreover, the deformation it is exposed to remains well below its own elastic limit, so that it would not get damage, despite its larger free swelling strain.

The case of thermal dilation is quite interesting. The artificial stone has a higher thermal expansion than the natural stone between 10°C and 25°C. However outside of this range it is low. This means that very large temperature differences between the artificial stone extending beyond this temperature range lead to a reduction in differential thermal dilation. The most critical situation is therefore if the underlying stone is at 10°C and the mortar overlay is heated to 25°C due to the sun appearing.

Owing to the low elastic modulus in the artificial stone when wet, the worse situation for the dry stone is if the artificial stone is dry. Whether the underlying stone is dry or wet, we once again find that the strains do not exceed the elastic limit (about 0.2 $mm.m^{-1}$versus 0.6 $mm.m^{-1}$). While the difference is significant it is less important than in the case of hydric swelling.

Concerning the artificial stone, the worst situation would be for it to be fully confined. Here we find that the temperature difference taken above would not lead to straining it beyond its elastic limit. Therefore we also expect the artificial stone not to be affected by differences in thermal dilation coefficients.

4. Conclusions

The low elastic modulus of the artificial stone, associated to a relatively small thermal expansion avoids this repair material from damaging the original substrate. Moreover, despite a large hydric swelling, its low elastic modulus especially in wet condition, once again avoids subjecting the original stone to critical stresses. It is also worth noting that the large elastic limit of the artificial stone together with its low modulus allow it to accommodate deformations of the stone without being damaged or damaging the natural stone.

These assumptions obviously do not consider the phenomenon of fatigue due to repetitive stress cycles that should be studied by experimental means. Another situation needing consideration is damage at the interface between both materials. While this may be an issue for the longevity of the repair, it is however not expected to be a cause that would drive substantial damage to the underlying original stone. These first results are encouraging parameters for the long-term behavior as a repair material. However, since laboratory tests are not able to completely reproduce the passage of time, a monitoring of the artificial stone in a newly-repaired building is also ongoing.

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