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The ancient mortars of Serravalle fortification (Bosa, Italy): a case study

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Abstract

The case study is the pentagonal tower from Serravalle Castle fortification, located near to the river Temo (west-cost of Sardinia), above the medieval Bosa village (XII-XIV cent.). The castle belongs to the medieval *Giudicato di Torres* (X cent.) and then to Malaspina from Lunigiana (XII cent.), Turrritani, Arborea, Aragonese, until to the Catalans (from Alghero) that led it to decline (XVI cent.).
The aim of this proposal is to analyze from mineralogical-petrographic point of view the mortars used in the construction of the tower. Samples were taken between the volcanic ashlars at different heights with respect to the ground level, to assess any compositional variations during the construction of the building. Through microscopic analysis, the compositional aspects concerning the nature and petro-mineralogical features of the raw materials used for the aggregate were defined. By image analysis (on meso-microscopic photographs) the vol.% of binder/aggregate ratios and their grain-size characteristics were determined and compared with the wt% data obtained from disaggregation method of mortars and acid attack of carbonate binder, in order to understand if the mixing ratios used by constructors have respected the production standards of those historic period. By X-ray diffraction (XRD) and termo-gravimetric/differential analysis (TGA-DSC) of enriched-binder samples, the presence of C-S-H phases and the hydraulic degree of mortars have been studied, to understand the possible use of pozzolanic material. Finally, to define the physical-mechanical properties of mortars were also determined: porosity, real and bulk density, PLT punching-index, theoretical values of compression and traction strengths.

Keywords: Mortar aggregate, Binder, XRD analysis, Mineralogical composition, Physical properties

1. Historical aspects and aims of work

The Castle of Serravalle is one of the most famous medieval fortifications in Sardinia (Fig. 1). It is located on the top of the homonymous hill, within the Bosa village. According to the historian Fara, the oldest nucleus of ancient construction is attributable to Malaspina, which arrived in Sardinia in 1112 AD. Vittorio Angius (1831) observed that the original fortification was much smaller than the present one but was later expanded in Aragonese period (Soddu 2005; Spanu 1981). Fara (1892) highlights that the Marquis Malaspina had founded a new settlement center on the slopes of the Serravalle hill on which they built a fortress (Scano 1936). Then, from the Aragonese period, the castle was expanded, strengthened and further protected to emphasize the defensive role played by the structure in the following centuries (Guagnini 1973).
The complex was built in three phases starting in 12th century:
1) realisation of four cantonal towers (with height of about 10 meters) with inner interposed wall (Figs. 2, 3); 2) construction of the main

tower of the doorman, the overhanging of the walls and the creation of a second wall structure forming a trapezium; 3) construction of embankments and the St. Giovanni church.
The castle then declines in 1571. In 1575 the problem was discussed in Spanish parliament where citizens urged urgent interventions to restore the castle's masonry.
In the last century, the city walls were demolished and began, according to the indications of the current urban planning tools, the development towards the sea. The castle affected by two restoration works by Filippo Vivanet and Dionigi Scano in 1893, which predominantly interested the main tower.
In the present century, the castle was subjected to numerous restoration works carried out by the Sassari Superintendence with the aim of replacing some ashlars of the pentagonal tower irremediably altered (Columbu & Meloni 2015; Fig. 4). The last restoration projects (1999-2005) and were joined by archaeological excavations inside the walls that allowed new further discoveries.

Fig. 1- Serravalle Castle and down the Bosa village (central-west Sardinia)

The research addressed to study the bedding mortars between the ashlars used to construct the Castle, taken as case study the pentagonal tower. In detail, the work has following objects: i) define the mineralogical composition of aggregate of mortars; ii) determine the binder / aggregate ratio (wt% and vol%); iii) grain size characteristics of aggregate; iv) identify the presence of any pozzolanic component used as hydraulic material in the mortars.

In this article we show the preliminary results of analysis on the mortars.

Fig. 2- South-west view of pentagonal castle

Fig. 3- East facade of pentagonal tower (view from inside of the castle

Fig. 4- Altered ashlars of inner vault of pentagonal tower (east facade) and the bedding mortars

2. Material and analytical methods

Sampling (according to the NORMAL Recommendations 3/80) was carried out taking into account the need to not disfigure the monument in any way. In addition, sampling was performed taking into account the most representative and/or predominant mortars in the entire architectural building.

2.1 Materials

42 samples of mortars were collected from the tower and analysed. The specimens are representative of bedding mortars used between the ashlars (see a part of sample in Table 1).
To define any compositional variation or different mixture ratios of aggregate and binder, the mortars were sampled according to different heights in the structure and/or in diverse environments.
Ten lime lumps of mortars were also sampled and analysed to understand their composition and modality of formation.
Samples of mortar were taken from the limited portions of monument material, compatibly with the limits imposed by the local Superintendence of Cultural Heritage.
However, the size of the material taken from the tower is representative and suitable to determine the compositional and physical characteristics of the mortars studied.

2.2 Analytical methods

Petrographic determinations of mineralogical composition and the modal analysis of mortars were carried out by optical polarised microscopy on polished thin sections on 42 samples.
The binder/aggregate ratio (B/A) of mortars was calculated in two different ways: i) through image analysis (by ImageJ 1.47v) on thin section photographs detected with the flatbed scanner; ii) using weight data from acid dissolution of mortar binder to determine the particle size of aggregate (see text and figure captions of manuscript).
XRD analysis were performed by X-ray diffractometry using a Philips PW 1050/37 with X'Pert PRO Philips data acquisition system, operating at 40 kV-20 mA, with Cu-anode, graphite monochromator, interval 2θ 5 -70 °, detection limit of 4%.
Regarding the thermo-gravimetric analysis, two grams of each mortar (without the coarse aggregate) were ground by Giuliani IG colloidal mill. W2/E/S. To enrich the sample in the binder fraction, the powder was treated with Frantz magnetic separator for the removal of the iron-magnetic mineral fraction belonging mainly to the volcanic aggregate.
Thermo-gravimetric analysis (TGA) measurements were carried out at atmospheric pressure using a Perkin Elmer instrument model TGA7. The measurements were performed under Ar flow (60 mL min^{-1}). Samples of 10 mg were placed in platinum crucibles and scanned in the temperature range of 30–900 °C with a heating rate of 10 °C min^{-1}. TGA7 instrument was calibrated with Curie points of Alumel, Nickel, Perkalloy and Iron standard samples and the temperature was obtained with an accuracy of ± 2 °C. Differential scanning calorimetry (DSC) measurements were carried out at atmospheric pressure using a Perkin Elmer instrument model DSC7. The measurements were performed

under Ar flow (60 mL min^{-1}). Samples of 5 mg were placed in platinum crucibles and scanned in the temperature range of 30–650 °C with a heating rate of 10 °C min^{-1}. DSC7 instrument was calibrated by measuring the melting temperature of metallic Indium and Zinc (99.999 mass% purity) and the temperature was obtained with an accuracy of ± 0.5 °C.

Physical tests were determined on 42 cubic specimens (with an average size of 15•15•15 mm) extracted from unaltered portion of samples after removing the exterior part of mortar. Specimens were dried at 105 ± 5 °C and then the dry solid mass (m_D) was determined. The solid phases volume (V_S) of powdered specimens (on 5-8 g and with particle size less than 0.063 mm) and the real volume (with $V_R = V_S + V_C$, where V_C is the volume of pores closed to helium) of the specimens were determined by helium Ultrapycnometer 1000 (Quantachrome Instruments).

Then, the wet solid mass (m_W) of the samples was determined after water absorption by immersion for ten days. Through a hydrostatic analytical balance, the bulk volume V_B (with $V_B = V_S + V_O + V_C$, where $V_O = (V_B-V_R)$ is the volume of open pores to helium) is calculated as: $V_B = [(m_W-m_{HY})/\rho_w T_{25°C}]•100$

where m_{HY} is the hydrostatic mass of the wet specimen and $\rho_W T_{25°C}$ is the water density at a temperature of 25 °C.

Total porosity (Φ_T), open porosity to water and helium ($\Phi_O H_2O$; $\Phi_O He$, respectively), closed porosity to water and helium ($\Phi_C H_2O$; $\Phi_C He$), bulk density (ρ_B), real density (ρ_R), solid density (ρ_S) were computed as:

$\Phi_T = [(V_B-V_S)/V_B]•100$

$\Phi_O H_2O = \{[(m_W-m_D)/ \rho_W T_X]/V_B\}•100$

$\Phi_O He = [(V_B-V_R)/V_B]•100$

$\Phi_C H_2O = \Phi_T - \Phi_O H_2O$

$\Phi_C He = \Phi_T - \Phi_O He$

$\rho_S = m_D/V_S$; $\rho_R = m_D/V_R$; $\rho_B = m_D/V_B$

The weight imbibition coefficient (IC_W) and the saturation index (SI) were computed as:

$IC_W = [(m_W-m_D)/m_D]•100$

$SI = (\Phi_O H_2O/\Phi_O He) = \{[(m_W-m_D)/ \rho_W T_X]/V_O\}•100$

The punching strength index was determined with a Point Load Tester (mod. D550 Controls Instrument) according to ISRM (1972, 1985) on the same pseudo-cubic rock specimens used for other physical properties. The force was exerted via the application of a concentrated load with two opposing conical punches.

The resistance to puncturing (I_S) was calculated as P/D_e^2, where P is the breaking load and D_e is the "equivalent diameter of the carrot" (ISRM, 1985), with $D_e = 4A/\pi$ and $A = W•D$, where W and 2L are the width perpendicular to the direction of the load and the length of the specimen, respectively. The index value is referred to a standard cylindrical specimen with diameter D = 50 mm for which I_S has been corrected with a shape coefficient (F) and calculated as:

$I_{S(50)} = I_S•F = I_S•(D_e/50)^{0.45}$

The simple compression resistance (R_C) and the traction resistance (R_T) of the mortar were indirectly calculated (according to ISRM 1985) using the value of normalized punching resistance, each of them as:

$R_C = K• I_{S(50)}$ $R_T = I_{S(50)}/0,8$

where K (multiplication coefficient) = 14 (Palmström 1995).

To proceed with the particle-size analysis, the mortars were first disaggregated with the use of a mortar and pestle, dried at 105 ± 5 ° C, weighed to measure the dry mass (m_{dM}), and then attached with acid solution (HNO_3, 13% vol.) for a period of immersion of 48 hours, so as to eliminate the carbonate binder matrix of the mortar. The samples were then filtered with Whatmann 41 paper, washed in distilled water, placed in an oven at 105 ± 5 ° C to determine the dry mass of the residual aggregate (m_{dR}) and, indirectly, the bulk mass of the binder (as: $m_{dB} = m_{dM}-m_{dR}$). Then, the particle-size distribution was performed using sieves with mesh opening of 6300, 4000, 2000, 500, 250, 125, 63 mm with sifter Giuliani IG3.

3. Results and discussion

3.1 Mineralogical and petrographic characterization of mortars

Overall, the results of macroscopic and microscopic analysis highlight a compositional variability of the mortar samples, related to a different mixture employed in their production (Table 1).

Sigle	Tower facade	Exposition	H (m)	Macroscopic description	XRD analysis				
					Cc	Qz	Pl	K-Fds	Other phases
CB3	N	Faced to the Mistral wind (outside the castle)	0.06	Coherent mortar with withish binder, fine grey-black aggregate, Qz-Fds sands, on average from <1 to 1.5 mm	xxx	x	x	-	Ms (tr), Gy (tr)
CB6	N	Faced to the Mistral wind (outside the castle)	2.02	Not consistent mortar with greyish binder, fine grey-black and Qz-Fds aggregate on average from <1 to 4 mm	x	x	xx	-	Mo (XX), Cp (tr), Ms (tr)
CB27	NW	Faced to the sea (outside the castle)	4.57	Medium-coherent mortar with withish binder, medium grey-black aggregate, Qz-Fds sands, pyroclastic fragments, from <1-4 mm	xxx	x	x	-	Mo (tr?)
CB22	E	Vault of first floor within the room of scale	5.08	Low-coherent mortar with: beige binder, low and fine grey-black aggregate, Qz-Fds sands, pyroclastic fragments, on average from <1 to 5 mm	xxx	x	x	-	-
CB24	SW	Faced to the sea (outside the castle)	5.92	Coherent mortar with: withish binder, medium grey-black aggregate, Qz-Fds sands, on average from <1 to 6 mm	xxx	x	xx	-	-
CB36	NW	Faced to the sea (outside the castle)	6.02	Coherent mortar with: greyish binder, fine grey-black aggregate, Qz-Fds sands, on average from <1 to 4 mm and with wide rock fragments	xxx	x	x	-	Ms (tr)
CB21	N	Faced to the Mistral wind (outside the castle)	6.28	Coherent mortar with: beige binder, coarse-medium grey-black aggregate, Qz-Fds sands, on average from <1 to 4 mm and with wide rock fragments (some times flattened fragments)	xxx	x	x	-	-
CB33	S	Faced to the Temo river (outside the castle)	6.79	Coherent mortar with withish-beige binder, fine-medium grey-black aggregate, Qz-Fds sands, on average from <1 to 3 mm	xxx	x	x	Sd (X)	-
CB35	SW	Faced to the sea (outside the castle)	7.93	Coherent with beige binder, fine-medium grey-black aggregate, Qz-Fds sands, on average from <1 to 4 mm	xxx	x	x	Sd (X)	Ka (tr)
CB54	E	North-side within the room of scale	8.21	Medium-coherent mortar with beige binder,very fine grey-black and Qz-Fds aggregate, pyroclastic fragments (2-6 mm), on average from <1 to 2 mm (excluding the pyroclastites)	xxx	x	x	-	-
CB41	SW	Faced to the sea (outside the castle)	8.96	Coherent mortar with withish-beige binder, fine-medium grey-black aggregate, Qz-Fds sands, on average from <1 to 4 mm	xxx	x	x	-	Ms (tr)
CB42	S	Faced to the Temo river (outside the castle)	9.51	Coherent mortar with withish binder and grey-black aggregate (Qz-Fds sands, rock fragments) on average from <1 to 2 mm	xxx	x	x	-	Gy (tr)
CB40	NW	Faced to the sea (outside the castle)	10.1	Medium-coherent mortar with withish-beige binder, medium grey-black aggregate, Qz-Fds sands, on average from <1 to 4 mm	xxx	x	x	-	-
CB61	SW	Faced to the sea (outside the castle)	11.3	Medium-coherent mortar with beige binder, medium grey-black aggregate, Qz-Fds sands, on average from <1 to 4 mm	xxx	x	x	-	-
CB57	E	Located within the room of scale	12.4	Medium-coherent and heterogeneous mortar with: withish-beige binder, medium grey-black aggregate, Qz-Fds sands, various rocks on average from <1 to 8 mm, high pores (1-4 mm)	xxx	x	x	-	-

Table 1- Macroscopic analysis and XRD results of selected mortar samples taken from the pentagonal tower of Serravalle Castle. Data ordered according to the height from the ground of external wall. Abbreviations: H = height on the ground; Cc = calcite; Qz = quartz; Pl = plagioclase; K-fds = K-feldspar; Ms = muscovite; Sd = sanidine; Ka = kaolinite; Mo = mordenite; Cp = clinoptilolite; Gy = gypsum; xxx = high amount; xx = medium amount; x = low amount; tr = trace; - = absent

At the macroscopic observation, the binder matrix of samples shows a colour from greyish to whitish as function of decay degree on the surface portion of material. The samples exposed directly to the weathering on the monument show a colour from ochre to grey more intense due to the incipient chemical alteration: dissolution and / or sulfation processes (as highlighted by the presence of gypsum, see XRD results in Table 1).

In some zones of tower facades exposed to the north (then in absence of the sun radiation) and located in low-lying of the building, biological patinas (*e.g.*, molds, mosses, lichens) are present on the stone and mortars. In all samples of mortar there are often lime lumps with different dimensions (with frequently range from <1 to 15 mm), in some cases with radial fissuring or fractured. According also with XRD (Figs. 5, 6), the binder matrix is mainly constituted by microcrystalline calcite. At macroscopic scale, pores with size range between 1-3 mm is often observed while, at microscopic scale, the presence of micro-porosity (<100 μm) finely distributed in the binder-paste is observed.

Different gravel, sands and crushed rocks were used in the aggregate of mortars, mainly fragments of volcanics and other rocks, crystal-clasts (quartz, plagioclase, K-feldspar, sanidine, muscovite, kaolinite; Figs. 5, 6; Tab. 1).

At the moment chemical and petrographic analysis to study the provenance of raw materials are still in progress, but preliminary it's possible to assume that the volcanic fragments used as aggregate belong to local outcrops (near to the castle), belonging to the sardinian Oligo-Miocene volcanic cycle (32-11 Ma; Beccaluva et al., 1985; Columbu et al., 2011). These local pyroclastic rocks were used mainly as coarse aggregate in the bedding mortars analysed (with frequently size: 1-7 mm), showing porous and glassy appearance.

XRD analysis (Table 1) also shows the presence of zeolitic phases (mordenite, clinoptilolite) in the aggregate of mortars (CB6 sample, Fig. 6).

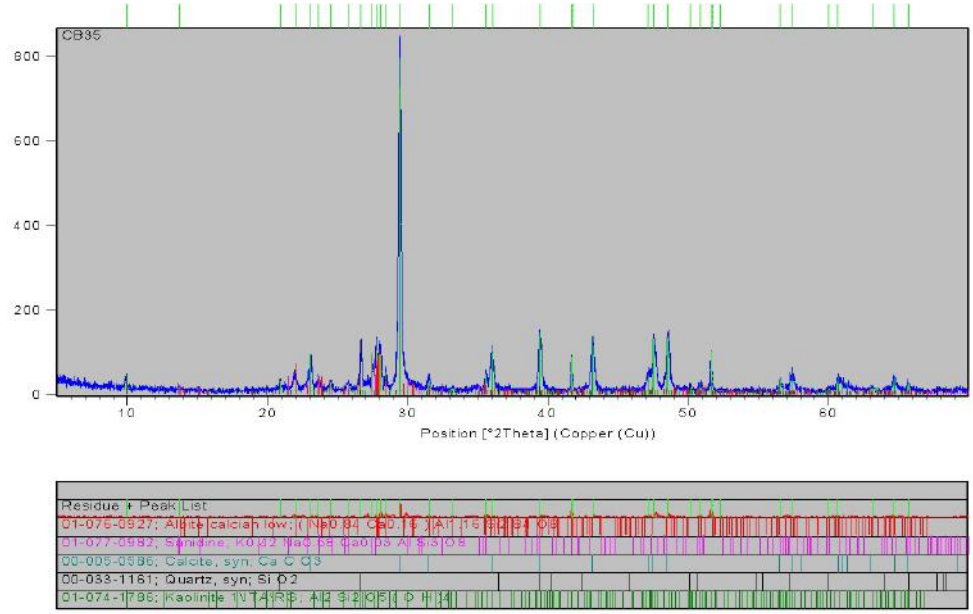

Fig. 5- XRD diffractogram on the enriched-binder specimen of CB35 mortar sample; there are following crystalline phases: calcite, quartz, plagioclase, sanidine, kaolinite

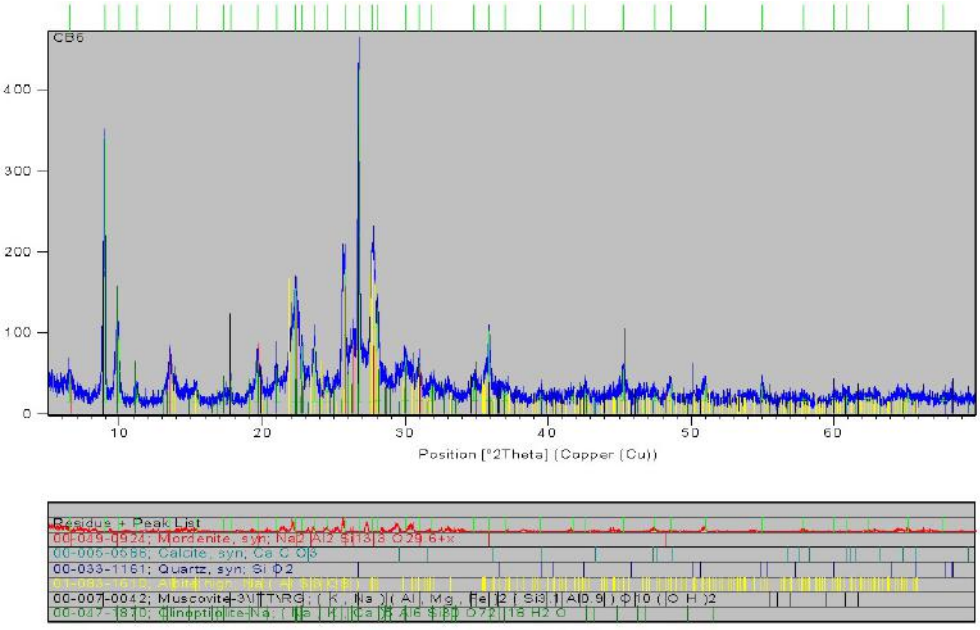

Fig. 6- XRD diffractogram on the enriched-binder specimen of CB6 mortar sample; there are following crystalline phases: calcite, quartz, plagioclase, muscovite, zeolite (i.e. mordenite, clinoptilolite)

The size of mortar aggregate is much variable: in general from submillimetric to subcentimetric. The shape varies from sub-spherical to angular.

3.2 Physical-mechanical characterisation

The physical data of the tower mortars show a value dispersion, due to the different binder / aggregate ratios of samples and to the relationships between the size of aggregate and dimensions of bulk mortar specimens. So, the great variability of open porosity and bulk density in the mortars is affected by also variable incidence of the binder and aggregate in the mixture. Moreover, in some cases the data variability is due to a different compaction and laying of mortars by workers at medieval time.
The porosity and bulk density, normally well correlated between them in inverse proportion, show no good correlation coefficients, and they show a low-medium compactness of mortars. This aspect is also highlighted by the physical mechanical characteristics of mortars, showing low values of punching strength index (frequently $Is_{50} < 1$ MPa).

4. Conclusions

The first results of this research allowed a preliminary mineralogical, petrographic and physical-mechanical characterisation of the mortars used in the construction of the pentagonal tower of the Serravalle Castle.
By XRD analysis, the binder mainly consists of calcite. As aggregate used sands (mainly represented by crystal-clasts of quartz, plagioclase, K-feldspar and subordinately, sanidine, muscovite) and rock fragments (mainly volcanics belong to local outcrops, together other rocks). The presence of volcanic aggregate (at the amorphous state and with acid composition) and zeolite (which may play a non-secondary role in the hydraulicisation process) suggest that such mortars could be hydraulic. However, at the moment preliminary XRD analysis of enriched-binder specimens of mortar samples not highlight any presence of hydraulic phases (i.e. C-S-H) deriving from chemical reaction between the fine volcanic aggregate and the Ca-carbonate binder. Moreover, considering their relatively low compactness and hardness, such mortars probably are basically aerial lime or only partially hydraulic. In any case, the results of TGA-DSC analysis (in progress) will highlight these technological aspects. The low punching strength (and its slightly correlation with the porosity) indicate that the resistance of mortars is affected by different factors: i) high and variable porosity of the bulk sample; ii) small dimensions of the specimens respect to aggregate size; iii) variable characteristics of the binder (*i.e.*, cohesion degree, microporosity); iv) sorting degree and particle size of the aggregate.
Subordinately, the mechanical resistance depends on the thickness of mortars at the moment of their laying.

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