WERE CASTLE WALLS IN PORTUGAL COVERED WITH RENDERS? THE CASE OF ARRAIOLOS CASTLE

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Abstract

Arraiolos Castle is one of the medieval fortresses that still stands in Southern Portugal with few modifications of restoration and preservation interventions. Particularly relevant is the fact that large areas of the castle walls are covered with mortars and some have gothic inscriptions that point out to their production in the second half of the 14th century. This work was carried out in order to verify if the mortars present were the same with each other and with the age of the mortar inscriptions. The characterization methodology involved a multidisciplinary set of chemical and microanalytical techniques, as well as radiocarbon dating. The results showed that all render mortars have similar composition, with aerial calcium magnesium lime as binder and aggregates with siliceous nature and similar in composition to the rock of the region around Arraiolos. The radiocarbon blind analysis corroborated the date discovered in one of the gothic inscriptions found in the renders of the castle walls.

Keywords: Castle walls; Historical mortars; Radiocarbon dating

Introduction

It is common the idea that medieval castles in Portugal always fully revealed the ashlars or more crudely stacked stone and that these fortifications had the same appearance today as they had since its foundation. This belief is fuelled either by the romantic view of the castles as symbol of nationality, in vogue since the nineteenth century, or by the incorrect interpretation of these defensive buildings, muddying the reading of military heritage. However, disentangling ourselves from preconceived ideas and based on careful analysis of the most important medieval documents, in-situ observation of what still remains of the original ancient fortifications and laboratory analysis of samples of mortars present there - it becomes clear that castles, for the most part, were covered with renders. Furthermore, maintaining the current model of interpretation for these buildings constitutes a threat for their preservation [1].

This article relates to the analysis of a case study, in particular – Arraiolos Castle - whose constitution of stone masonry anticipates the likelihood of being fully rendered.

This fortification arrived fairly ruined to the twentieth century (see Fig. 1a). Although between 1961 and 1963 the South Tower has been rebuilt, the castle itself was generally spared

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to major restoration interventions, for example, significant cleaning of the coatings was not made, as happened in many other castles in Portugal.
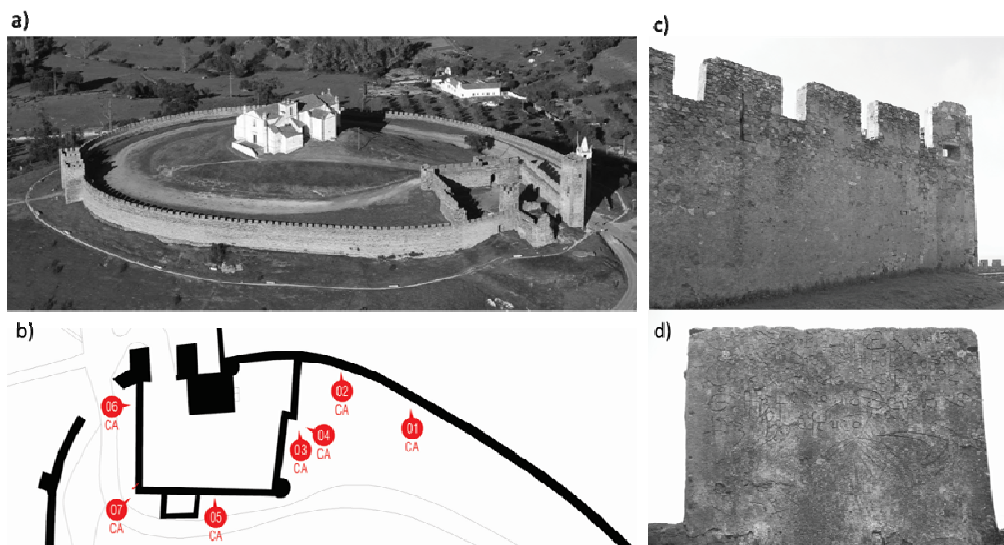

Fig. 1. Arraiolos Castle: a) general view, b) mortar sampling location,
c) area of castle wall with mortar renders, d) gothic inscription in a mortar

The fortress preserves large areas of renders in all the walls. These renders are possibly, according to their appearance, to belong to the same historical period. Moreover various gothic inscriptions (see Fig. 1b) are found especially on the north walls, where the coatings are best preserved. Although compelling, these would be similar to the findings in other castles, was not the existence of a particular inscription discovered by Bilou and Branco in 2010 [2] that could shed some light about the origin of these renders. This inscription, in excellent state of conservation, registers the making, in 1385, of "twelve niches", a "small restoration in the walled village" - and reads:

"*Era de Mil E IIII(c) E XXIIIº a (-) nos XXIIIIº dias d´aabryl forom fectas Estas XII ameas E Castelonas G(onçalo) Fernandez pedreyro d´evora Vasco Pirez ho Escriveo*"

According to the authors, the modern transcription of the inscription follows:
"*Age of thousand and four hundred twenty-three years (year of Christ 1385) twenty-four days of April, these twelve niches and 'castelonas' were made (by) Gonzalo Fernandez, mason of Evora; Vasco Pirez wrote.*"
The above mentioned study focused only on the transcription of this inscription and others also found in the castle walls and it became clear that all inscriptions were made in the same period. However, the authors did not extrapolate directly or indirectly, any reflection on the construction system, nor was the hypothesis of these structures been rendered entirely and not just in some parts put forward.
In order to prove, definitively, the reliability of the historical descriptions made and to understand whether all render remains are similar, a multi-analytical study was envisaged, that included the radiocarbon dating of the mortars from Arraiolos Castle, together with the chemical and mineralogical composition of a set of samples from different areas of the Castle.

Experimental

The sampling of mortars is a crucial step that can influence the success of the characterisation methodology. The selection of the sampling sites was made according to an architectural systematization study. The location of the samples was mapped onto the architectural plans and made its photographic record. The size of each sample was the minimum that could guarantee the success of the analysis. Seven mortars were sampled from different sites, as shown in Fig. 2 and described in Table 1. All samples correspond to mortar renders with exception to mortar CA4 which corresponds to a filling mortar.
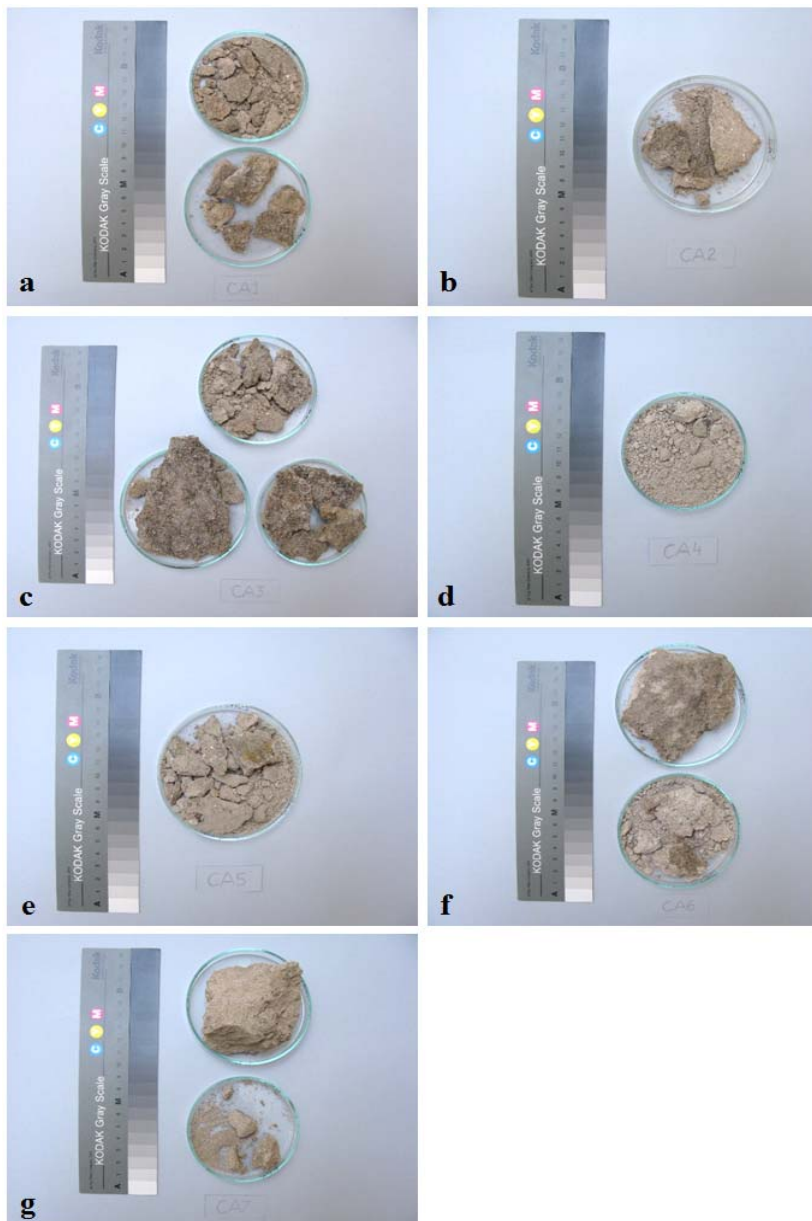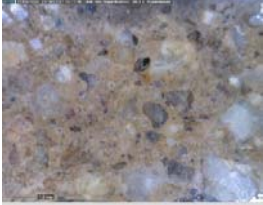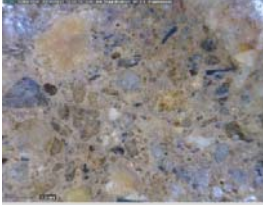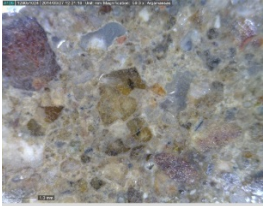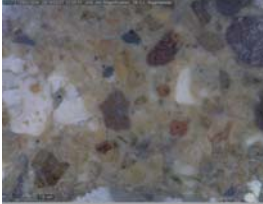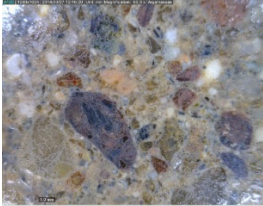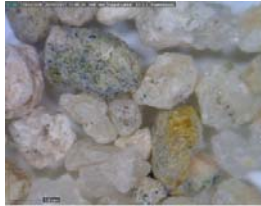
Fig. 2. Mortar samples collected: a) CA1, b) CA2, c) CA3, d) CA4, e) CA5, f) CA6, g) CA7

Table 1. Optical microscopy observations.

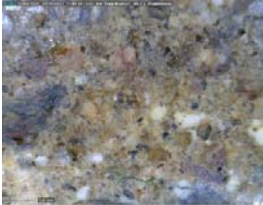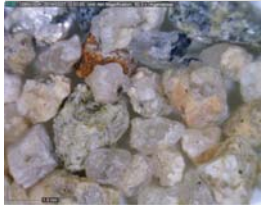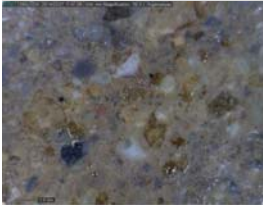| Sample	Polished surface	Insoluble residue	Observations
CA1			Binder colour: light cream colour Aggregates morphology: sub-angular and angular Insoluble residue %: 60 Main features: quartz (hyaline), feldspars, amphiboles, ceramics
CA2			Binder colour: light cream colour Aggregates morphology: sub-angular and angular Insoluble residue %: 65 Main features: quartz, feldspars, amphiboles, ceramics
CA3			Binder colour: light cream colour Aggregates morphology: sub-angular and angular Insoluble residue %: 73 Main features: : quartz, feldspars, amphiboles
CA4			Binder colour: light cream colour Aggregates morphology: sub-angular and angular Main features: : quartz, feldspars, amphiboles, ceramics
CA5			Binder colour: light cream colour Aggregates morphology: sub-angular and angular Insoluble residue %: 74 Main features: : quartz, feldspars, amphiboles, ceramics
CA6			Binder colour: light cream colour Aggregates morphology: sub-angular and angular Insoluble residue %: 71 Main features: : quartz, feldspars, amphiboles
CA7			Binder colour: light cream colour Aggregates morphology: sub-angular and angular Insoluble residue %: 61 Main features: : quartz, feldspars, amphiboles

The mortars were thoroughly examined in the laboratory using a stereo-zoom microscope and carefully disaggregated to avoid breaking the existing aggregates.

Optical microscopy observations and the corresponding photographic documentation were obtained by a dark field microscope Leica DM2500 (x100 and x200 magnifications) equipped with a Leica DFC 290HD camera.

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Scanning electron microscopy coupled with energy dispersive X-ray spectrometry (SEM-EDS) was carried out using a Hitachi S-3700N variable pressure scanning electron microscope coupled with a Bruker XFlash 5010 SDD Detector. The observations were done under a pressure of 40 mPa and the current used was 20 kV. For SEM-EDS and optical microscopy the micro-samples were prepared as polished-sections, mounted in epoxy polymeric resin and polished with silicon carbide with the granulometry of 1200 µm in order to distinguish between the different layers and to easily identify the composition of particles by backscattering electron imaging and also to obtain good quality X-ray spectra and 2D compositional maps.

In order to confirm the mineralogical phases of the mortars, micro-X-Ray diffraction (µ-XRD) was performed using a BRUKER AXS D8 Discovery diffractometer with a Cu Kα radiation source and a BRUKER LynxEye energy-dispersive one-dimensional detector. The diffractogram was obtained in the interval 3-74º 2θ, step of 0.05º, time per step of 1s. The identification of the phases was performed with the Bruker EVA software package (Version 3.0) using the PDF-ICDD Powder Diffraction Database (International Centre for Diffraction Data). EVA software enabled also semi-quantitative analysis based on the reference-intensity-ratio (RIR) method by scaling the maximum intensities of the ICDD PDF patterns to the observed peaks in the powder pattern.

The mortar's binder composition and CO_2 content was assessed by thermogravimetry (TGA) and differential thermal analysis (DTA) performed on a Netzsch STA 449F3 Jupiter analyser, under nitrogen atmosphere, with heating rate of 10ºC/min, from room temperature to 1000ºC. TGA measures the weight (mass) change of a sample as a function of temperature while the DTA measures the energy changes, represented by endothermic or exothermic peaks in the DTA curve.

Quantitative analysis is based on the TGA curves (thermograms) while DTA provides information for the qualitative identification of the components that undergo weight losses [3, 4]. The weight loss of the decomposed main phases was determined by Proteus© software for the selected temperature ranges.

A fraction of the disaggregated sample was afterwards digested in warm dilute hydrochloric acid (1:3) to separate the siliceous aggregates from the lime paste. The insoluble residue was weighed and sieved to determine the particle size distribution of the aggregate fraction i.e. the siliceous sand.

Taking into account the unique opportunity to study the Arraiolos Castle plasters with incised Gothic calligraphy, it was considered relevant to proceed with the dating of two samples

using the technique of Accelerator Mass Spectrometry Radiocarbon Dating. The analyses were performed as outsourcing at the Department of Physics and Astronomy, University of Aarhus in Finland led by Alf Lindroos and Jan Heinemeier, global experts and forerunners of this technique for dating mortars [5, 6].

Results and Discussion

Observation by optical microscopy

The first approach to assess the composition of the mortars was the observation with naked eye of samples and under optical microscope of the polished surfaces and the insoluble residue obtained after acid attack. Table 1 depicts the observations for each mortar. All samples have the same type of mortar with a heterogeneous composition and light cream colour that disaggregates easily, revealing low cohesion. Mortar CA4 exhibits a lighter whitish colour in respect to the others. Regarding the aggregates, quartz is the dominant mineral. It is also possible to identify abundant presence of feldspars and plagioclases and some amphiboles. The composition of the aggregates is characteristic of the rocks of Arraiolos region where metamorphic rocks of intermediate to high degree abound, like amphibolites, and minor occurrences of igneous rocks, such as granites. The aggregates have predominantly sub-angular morphology, being less frequent angular which is characteristic of sands with little transport. It was also possible to observe the presence of some ceramic fragments.

With the insoluble residue it was also possible to determine the particle size distribution which is presented in figure 3. The granulometric curves and the insoluble residue amount are similar to all samples which tends to indicate that all mortars were prepared with the same type of raw materials (aggregates and binders) and mortar preparation procedures.
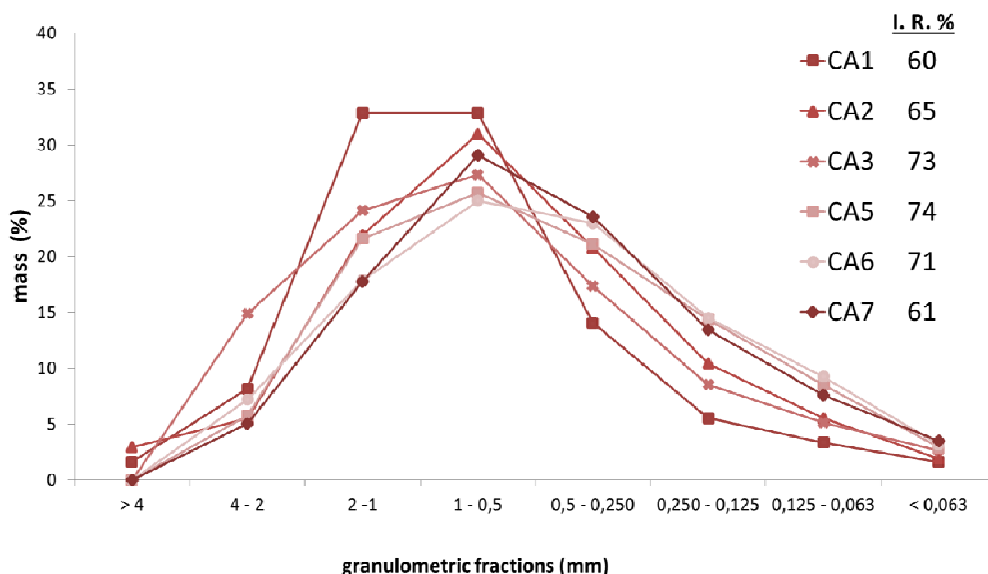

Fig. 3. Particle size distributions of Arraiolos Castle mortars (I.R% - insoluble residuum percentage).

X-ray diffraction

X-ray diffraction patterns are similar for all the render mortars studied. The results corroborate that quartz is the main mineral present. These mortars are still characterized by abundance of feldspars from the group of potassium feldspars and plagioclases and the presence of considerable amounts of amphibole. Table 2 presents the XRD phase's semi-quantification by RIR method.
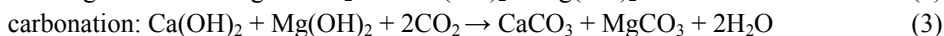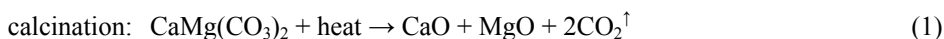
Table 2. XRD phase quantification.

	calcite	quartz	feldspar	amphiboles	muscovite
CA1	12.3	42.0	23.5	6.2	16.0
CA2	8.6	48.2	21.0	6.2	16.0
CA3	5.2	40.1	33.6	2.6	18.5
CA4	8.1	33.7	32.3	5.4	20.5
CA5	4.9	54.6	17.0	8.5	15.0
CA6	3.6	57.8	18.1	6.0	14.5
CA7	7.8	51.0	17.0	5.2	19.0

The results show that the amount of calcite is very low which is consistent with the low mechanical properties displayed by these mortars. Furthermore, the mortars composition variability is very low which is consistent with the previous observations that these mortars were most certainly produced at the same time.

Scanning electron microscopy

Scanning electron microscopy observations allowed a deeper insight on the microstructure and composition of the mortars. The results show that, for all the render mortars, the binder morphology is typical of aerial lime mortars (Fig. 4a) and its composition is enriched in calcium and magnesium (see Fig. 4 b, c). This result is typical of aerial lime mortars of dolomitic origin. In most cases, calcium and magnesium come apart, indicating the presence of calcite ($CaCO_3$) and magnesium carbonate compounds formed during the carbonation of a lime produced from a dolomite [$CaMg(CO_3)_2$)] or a limestone with magnesium, according to the following chemical reactions:

calcination: $CaMg(CO_3)_2 + heat \rightarrow CaO + MgO + 2CO_2\uparrow$ (1)
slacking: $CaO + MgO + 2H_2O \rightarrow Ca(OH)_2 + Mg(OH)_2 + heat$ (2)
carbonation: $Ca(OH)_2 + Mg(OH)_2 + 2CO_2 \rightarrow CaCO_3 + MgCO_3 + 2H_2O$ (3)

The results also show in some cases an enrichment of magnesium in fractures and at the binder-aggregates interface (see Fig. 4d). The aggregates are of siliceous origin with prevalence of quartz and the presence of other aluminossilicates (see Fig. 4e, f). It was also possible to detect the presence of ceramic fragments.
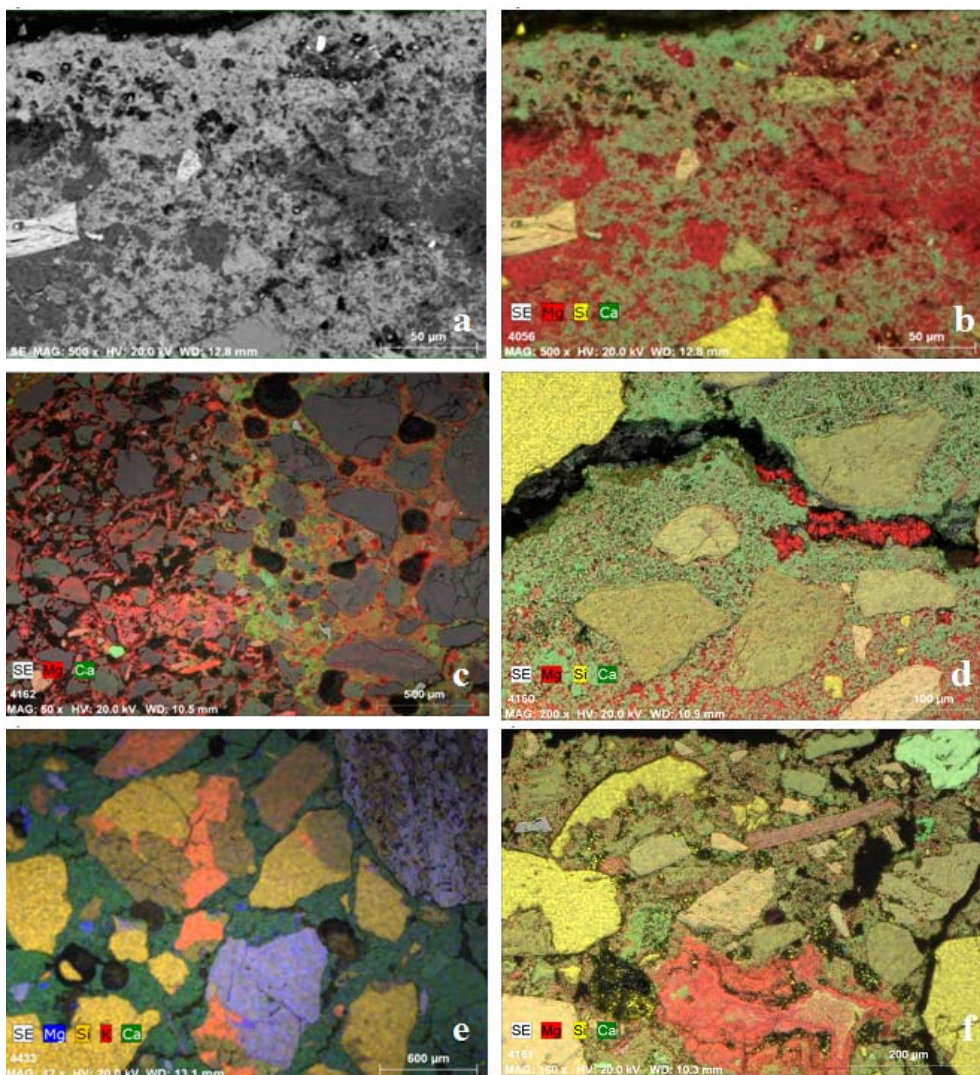
Fig. 4. SEM-EDS microanalysis: a) lime paste microstructure morphology- sample CA3; EDS 2D elemental distribution b) sample CA3 – lime paste, c) sample CA5 – lime paste, d) sample CA2 - magnesium enrichment in a fracture , e) sample CA4 – aggregates composition and calcitic lime paste, f) sample CA5 – aggregates composition

Thermogravimetric analysis

The studied samples display similar thermograms for all the samples besides CA4 (Fig. 5a) mortar for which can be defined similar temperature ranges (Table 3); TG, DTG, DTA results of Arraiolos castle render mortars (Fig. 5b) display a continuous and smooth weight loss until 200°C which can be attributed to removal of adsorbed water, followed by the dehydration of hydromagnesite ($3MgCO_3.Mg(OH)_2.3H_2O$) between 200-350ºC marked by peaks around 250ºC. A mass loss at around 396-429ºC is attributed to the dehydroxilation of hydromagnesite. Calcium carbonate is marked by a pronounced weight decay between 711 to 773 ºC. The drops between 450-600ºC for CA5, CA6 and CA7 samples can be attributed to decarbonation of

hydromagnesite. These results corroborate the data prior observed by SEM-EDS that showed the presence of magnesium and calcium in the binder.
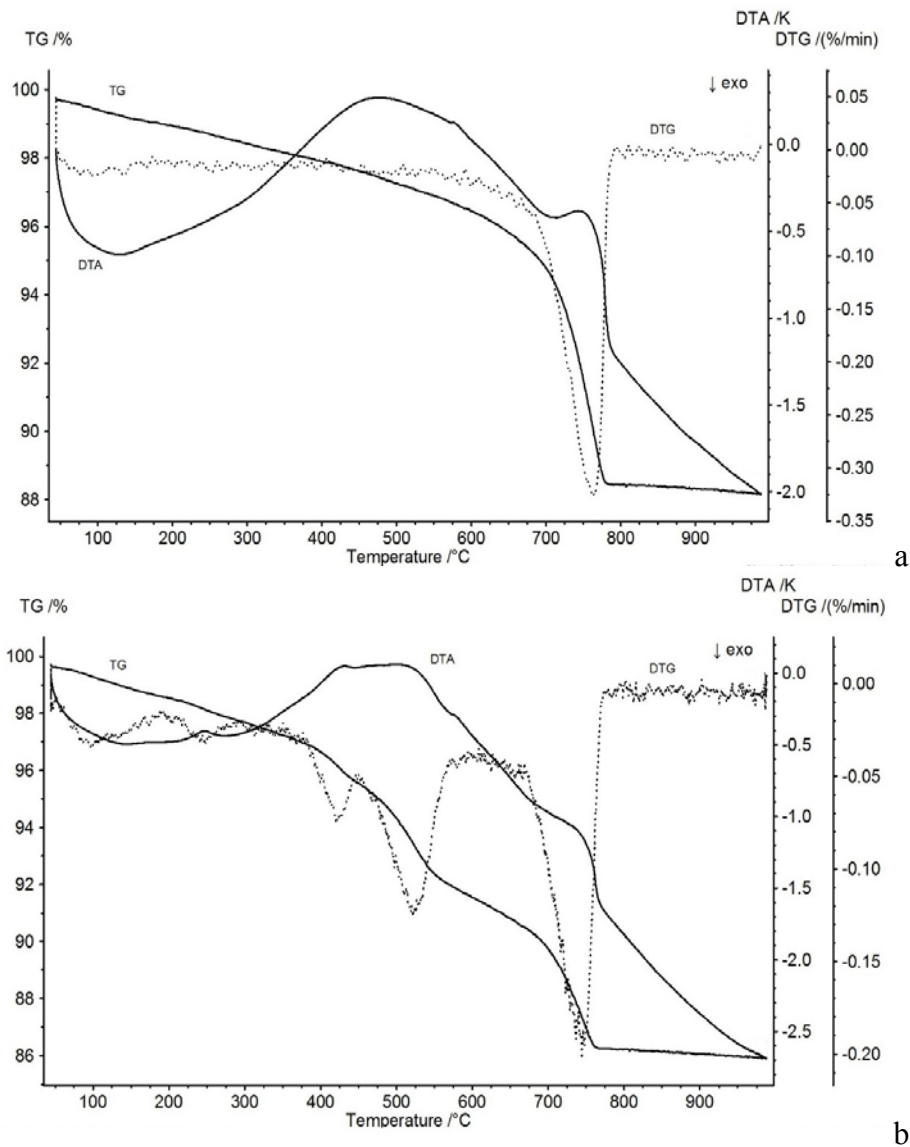

Fig. 5. Thermograms (TG, DTG and DTA curves) from Arraiolos Castle mortars; a) calcitic filling mortar (sample CA4); b) representative thermogram from magnesium render mortars (CA6 sample).

Contrary to the other thermograms CA4 doesn't show mass losses that can be attributed to the presence of magnesium carbonate. The only significant drop is attributed to decarbonation of calcite with a peak at 763ºC (Table 3). This is consistent with the fact that CA4 is a filling mortar while the others correspond to render mortars.

Table 3. Weight losses (%) as a function of temperature ranges (ºC) for render mortars.

weight losses (%) as a function of temperature ranges (ºC)					
ref.	**40-200**	**200-350**	**350-600**		**600-1000**
CA1	1,30	1,87	5,63		10,78
CA2	1,23	1,67	6,00		7,39
CA3	1,02	1,15	3,30		6,55
CA4	1,82				9,73
	40-200	**200-350**	**350-455(50)**	**455(50)-600**	**600-1000**
CA5	1,17	1,46	2,21	2,68	5,50
CA6	1,16	1,30	1,68	4,02	5,63
CA7	1,38	1,18	3,63	4,05	4,98

Radiocarbon dating

One has to take into consideration that the dating of mortars by the radiocarbon method is very recent and requires special conditions such as the absence of calcite aggregates, as these necessarily lead to obtaining a production date prior to the date of the mortar, and the absence of dissolution processes and re-carbonation because this results in the incorporation of carbon dioxide in the structure and consequently at a later date to the mortar. The composition of the Arraiolos mortars which had no evidence of calcite aggregates or dissolution processes and re-carbonation, and the fact that it is a technique used successfully in several case studies prompted the team to consider its application and therefore, three samples were selected for radiocarbon dating using AMS. The dating was only possible in two samples.

Radiocarbon ages obtained are reported in conventional radiocarbon years BP (before present = 1950) according to the international convention [7]. Thus, all calculated radiocarbon ages were corrected. Ages in calibrated calendar years were obtained from the calibration curves of Reimer et al. [8], using Oxcal [9] v4.1 program calibration curve using the terrestrial calibration IntCal09. The probability method was used to calculate the calibrated age ranges corresponding to 68.2% probability (1x standard deviation) and 95.4% probability (2x standard deviation). The radiocarbon dating curve is presented in figure 6 and the results in table 4.

Table 4. Radiocarbon calibrated dates.

sample	callibration curve and correction	Calibrated data 68.2% probability	Calibrated data 95.4% probability
1	IntCal09 (Atmospheric)	1324-1346 (31.4%)	1308-1362 (47.9%)
		1393-1416 (36.8%)	1386-1430 (47.5%)
2	IntCal09 (Atmospheric)	1423-1445 (68.2%)	1408-1456 (95.4%)

The calibrated dates obtained by the radiocarbon method demonstrate that the existing mortar coating on the renders of the Arraiolos Castle walls are from the late fourteenth / early fifteenth century. These results are consistent with the gothic inscription reporting the date of 1385, and a clear evidence of the use of mortar coating in the castle walls in this period. Furthermore, and taking into consideration the inscription date, it is also a significant result on the usefulness of radiocarbon dating of mortars.
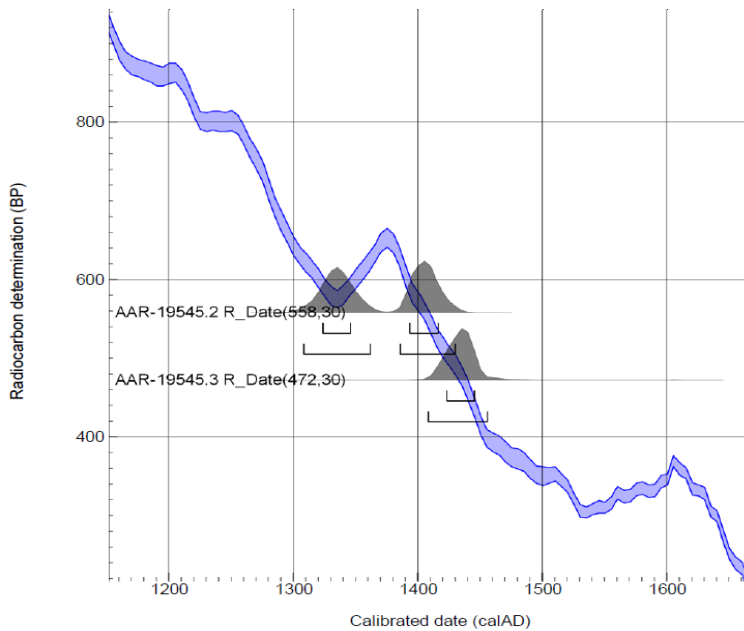
Fig. 6. Radiocarbon determination as a function of calibrated data

Conclusions

The multianalytical methodology applied showed that Arraiolos Castle mortars have similar compositions with magnesium-calcitic binder, and siliceous sands with compositions similar to the rocks from Arraiolos surroundings. It was finally possible to understand that renders in areas located far from each other are similar and originate from the same period - which would lead to conclude that most probably, the whole castle walls had renders that covered the whole surface. Particularly relevant was the fact that radiocarbon dating is consistent with the date discovered in an incisive inscription found in one of the mortars of the castle which points out its production in the second half of the fourteenth century beginning of the fifteenth century.

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