Appl. Phys. A (2014) 114:647–653
DOI 10.1007/s00339-014-8272-z

Applied Physics A
Materials Science & Processing

INVITED PAPER

XRF investigation on skeletal remains from King Peter III of Aragon (1239–1285 A.D.) and Queen Blanche of Anjou (1280–1310 A.D.)

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Received: 17 January 2014 / Accepted: 21 January 2014 / Published online: 4 February 2014
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Abstract We conducted an X-Ray Fluorescence investigation on bone fragments belonging to King Peter III of Aragon and Queen Blanche of Anjou. The spectroscopic analysis was carried out in selected points of the bone fragments. Several transitional elements normally unexpected in the bone composition have been found at varying level of concentration. The presence of these elements was interpreted in relation to chemical treatments for mummification of bodies as well as to dietary habits, including tools used for cooking and for the consumption of food.

1 Introduction

Bones and teeth are often the only direct remains of animals and humans and hence represent valuable archives for archeology and forensic sciences. To characterize and quantify the structural and chemical integrity of bones, different physico-chemical and spectroscopic techniques can be applied [1]. X-ray fluorescence spectrometry (XRF) is a technique that has been recently employed in both archeological and forensic contexts. In archeology, it is useful to study elemental concentrations in bones of different age and conditions [2–9] and to evaluate diagenesis or fossilization processes [10–13] even though this technique is barely used for this scope. It has been introduced into the forensic domain in the search for trace evidence at crime scenes [14] including gunshot residue and bodily fluids [15] and used to differentiate human from non-human material in cases of fragmented degraded samples*** [16]. XRF analyses can be applied even in the case of bodies with a precise archeological and burying context, especially in cases of important historical characters because known information allows to verify the hypothesis. This is the specific case that we are going to present hereafter.

In March 2010 in the Royal Monastery of Santes Creus (Aiguamúrcia, Catalonia, Spain), the graves of King Peter III, James II and Queen Blanche of Anjou were exhumed because of the restoration and research project of the Royal Tombs of Santes Creus [17]. The bones were analyzed in the laboratories of *Unitat d'Antropologia Biològica* (Universitat Autònoma de Barcelona, Spain) for anthropological and forensic investigations. Some chemical and physical analyses were carried out in the University of Sassari (Italy).

The main aim of this work is to present a multi-elemental analysis carried out with a laboratory XRF spectrometer and with a portable energy-dispersive XRF instrument on bone fragments belonging to Queen *Blanche of Anjou* and King Peter of Aragon, in order to evaluate the bone elemental composition and to reconstruct possible *in-vitam* or *post-mortem* contaminations.

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2 Brief historical and anthropological informations

2.1 (a) King Peter of Aragon

Peter "*the Great*" (Naples, 1239 A.D.—Vilafranca del Penedès, 11th November 1285 A.D.) was the King of Aragon (as Peter III) of Valencia (as Peter I), and Count of Barcelona (as Peter II) from 1276 A.D. to his death. He conquered Sicily and became its king in 1282 A.D. He was one of the greatest medieval Aragonese monarchs.

The historiography relates that King Peter of Aragon died at the Royal Palace of Vilafranca del Penedés on 11th November 1285 A.D. He was buried in front of the altar of the Royal Monastery of Santa Maria de Santes Creus. We must note that the burial was not immediate after death. Furthermore, on 2nd or 3rd December 1302 A.D., the body of the King was placed in his definitive grave, a porphyry basin, in a monumental tomb. The bones were in a state of excellent preservation and conservation. The skeleton was partially covered by white linen fabric and several layers of dried organic material (see Fig. 1).

Although it is a secondary deposition, the body has maintained a perfect skeletal articulation, except the feet bones that have been intentionally placed between the legs with the aim of matching the body to the porphyry basin. The manipulation of the feet bones is an anthropological evidence of the tomb exchange.

2.2 (b) Queen Blanche of Anjou

Blanche of Anjou (Naples, 1280 A.D.—Barcelona, 14th October 1310 A.D.) was the second Queen consort of James II of Aragon, the second son of Peter III of Aragon and Constance of Sicily. She was a member of the Capetian House of Anjou; she is also known as *Blanche of Naples*.

The grave of James II and Blanche of Anjou was desecrated in 1836, after the confiscation of Mendizabal [18]. It contained human remains belonged to a minimum of three adult individuals. The skeleton of Blanche of Anjou recomposed in laboratory (see Fig. 2) corresponds to a young adult woman of about 150 cm [17].

Fig. 1 The skeleton of Peter III of Aragon, partially covered with white linen fabric and layers of dried organic material (photo courtesy of C. Aymerich and R. Maroto MHC-CRBMC)

Fig. 2 The mummified body of Queen Blanche of Anjou after the recomposition in the laboratory (photo courtesy of C. Aymerich and R. Maroto MHC-CRBMC)

3 Materials and methods

3.1 XRF analysis

The bone material analyzed consisted of rib and femur fragments belonging to King Peter III of Aragon and metacarpal, rib and femur fragments belonging to Queen Blanche of Anjou.

The bone samples were analyzed as received with a Bruker M4 Tornado μ-XRF spectrometer using a Rh X-ray source model MCBM 50–0.6 B working at 50 kV and 600 μA under vacuum (20 mbar) and using a Al filter 12.5 μm thick. To check the macroscopic chemical homogeneity, a series of 20 spectra were collected for each bone specimen. Each spectrum was accumulated for 600 s. The bone samples were analyzed also with a portable XRF equipment, composed of an Ag-anode X-Ray tube by Amptek (model Mini-X), operating at 40 kV and an SDD detector. The limit of detection of the portable system is about 10 ppm.

An innovative fast XRF Monte Carlo quantification approach has been used [19]. This approach allows one to significantly improve the sensitivity of the spectrometers and also to take into account the roughness of the sample surface analyzed here. A model of the surface as a binary file can be loaded in the Monte Carlo at run time.

This code is based on *XRaylib,* a constantly updated X-Ray dataset [20, 21]. The Monte Carlo code has been tested on a variety of bones samples and cultural heritage items [9, 22, 23].

3.2 XRD analysis

The XRD patterns were recorded overnight with a Bruker diffractometer model D8 in the Bragg–Brentano geometry with CuKα radiation ($\lambda = 1.54178$ Å). The X-ray generator worked at a power of 40 kV and 30 mA and the resolution of the instrument (divergent and anti-scatter slits of ca. 0.5°) was determined using α-SiO_2 and α-Al_2O_3 standards free from the effect of reduced crystallite size and lattice defects [24]. The goniometer was equipped with a graphite monochromator in the diffracted beam and the

patterns were collected with 0.05° of step size which turned out to be adequate for the range of crystallite size in apatite phases here investigated. The powder patterns were collected in the angular range 10°–140° in 2θ, with counting time of 40 s per point. This strategy is suitable also for other phases, such as quartz, which has crystallites normally extended in size more than 2,000 Å, i.e., above the upper resolution limit of the instrument. Digitized diagrams were analyzed according to the Rietveld method [25], using the programme MAUD [26].

3.3 FT-IR analysis

FT-IR spectra were collected with a Bruker Vertex 70 V interferometer in terms of absorbance vs wavenumber ν in the range 400–4,500 cm^{-1}, with a resolution of 4 cm^{-1}. About 3 mg bone was hand-grinded and mixed with KBr to a weight ratio 1:100, respectively, to make pellets suitable for beam irradiation. Each spectrum was obtained by averaging 250 interferograms.

4 Results and discussion

4.1 (a) preservation state of Queen Blanche of Anjou and King Peter of Aragon bones using X-Ray Diffraction and FT-IR spectroscopy

The X-Ray Diffraction (XRD) and Fourier transform infrared spectroscopy (FT-IR) give a useful hints on the state of preservation of the bones through the observation of mineralogical phases and vibrational properties from specific molecular groups [1, 11, 12].

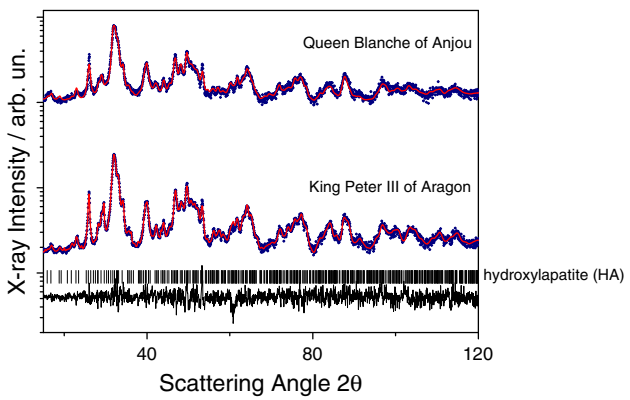

Fig. 3 The XRD powder patterns of femur samples belonging to Blanche of Anjou and King Peter. Experiment are data points, *full line* is the Rietveld refinement on the basis of Hydroxylapatite structure factors. The *line at the bottom* refers to the residuals, i.e., the difference between calculated and experimental square root intensities, which is indicative of the agreement obtained

In Fig. 3, we report a pattern of X-ray diffraction intensity from King Peter and Queen Blanche of Anjou femur samples, here studied as a function of the scattering angle 2θ.

A full curve using the Rietveld fit has been calculated according to the 100 wt% contribution of the structure factors of Hydroxylapatite (HA) (monoclinic, space group $P2_1/c$, refined lattice parameters $a = 9.440$, $b = 18.898$, $c = 6.896$ Å and $\beta = 120.67°$). The quantitative Rietveld analysis points to the fact that hydroxylapatite is the main phase for both sample bones.

As shown in curves Fig. 4a and b, the FT-IR spectra of Blanche of Anjou and Peter III of Aragon bones have a relatively poor signal-to-background ratio, showing the ν_4 and ν_3 phosphates bands (occurring in the frequency ranges of 500–650 and 1,000–1,200 cm^{-1}, respectively) that overlap with other bands, probably due to the presence of collagen and/or organic matter. Also the band ν_3 of carbonates (1,400–1,600 cm^{-1}) is quite broad, as can be seen clearly from 1,300 cm^{-1} onwards, similarly to that found recently by Lebon et al. [27].

Such transmittance profiles are different if compared with the analogous spectrum of a historic bone fragment (belonging to the Spanish Necropolis of *S'Illot de Porros*)

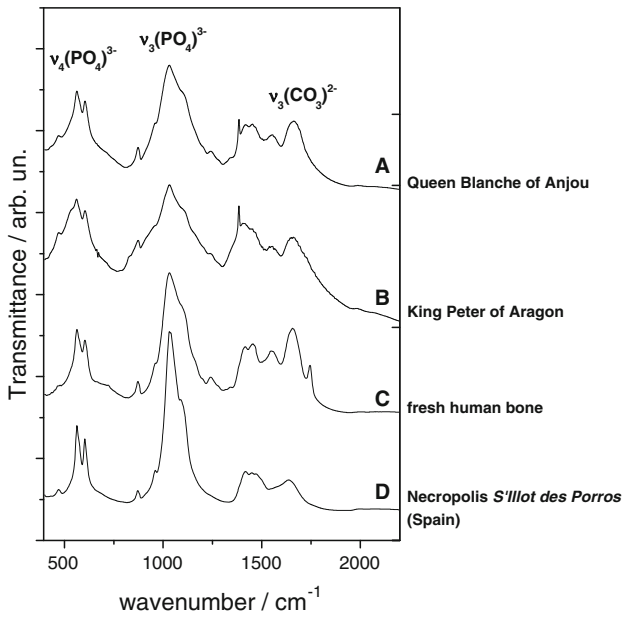

Fig. 4 *a* the FT-IR patterns of Queen Blanche of Anjou and King Peter III of Aragon compared with: a fresh human bone and a bone fragment, belonging to the Necropolis of *S'Illot des Porros* (Mallorca, Spain). It is possible to recognize three main groups of band in the range 500–700, 1,000–1,200 and 1,400–1,600 cm^{-1}, which for ancient bones are generally assigned to the energy modes of phosphate groups ν_4 and ν_3 and to the ν_3 of carbonate groups, respectively. Each curve highlights the differences in the shape of the three main groups of bands due to the presence of collagen in the cases *a* and *b*

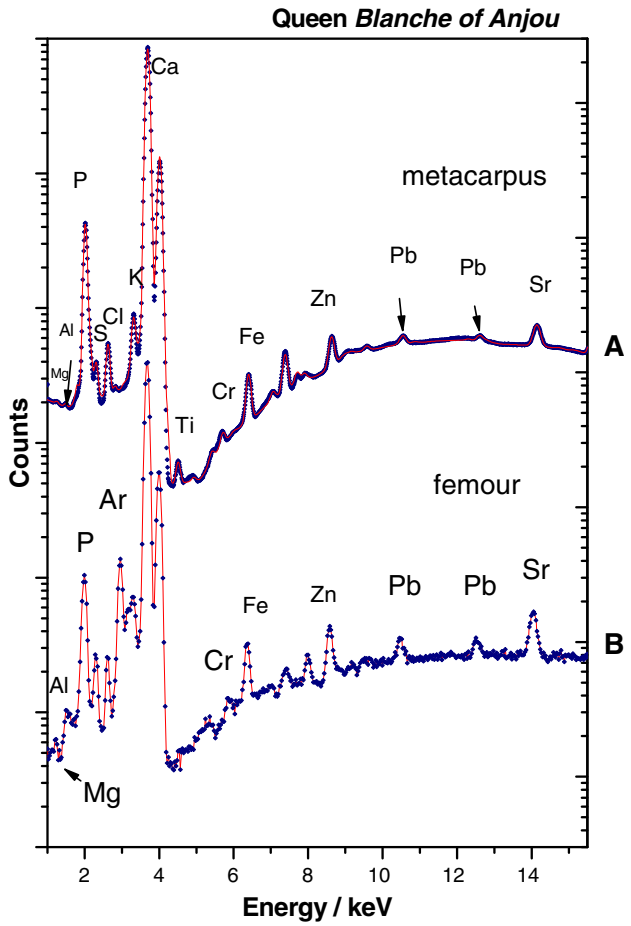

Fig. 5 X-ray fluorescence spectra taken at various points in the metacarpus and femur samples of Queen Blanche of Anjou with a laboratory XRF spectrometer (curve *a*) and with a portable XRF instrument (curve *b*)

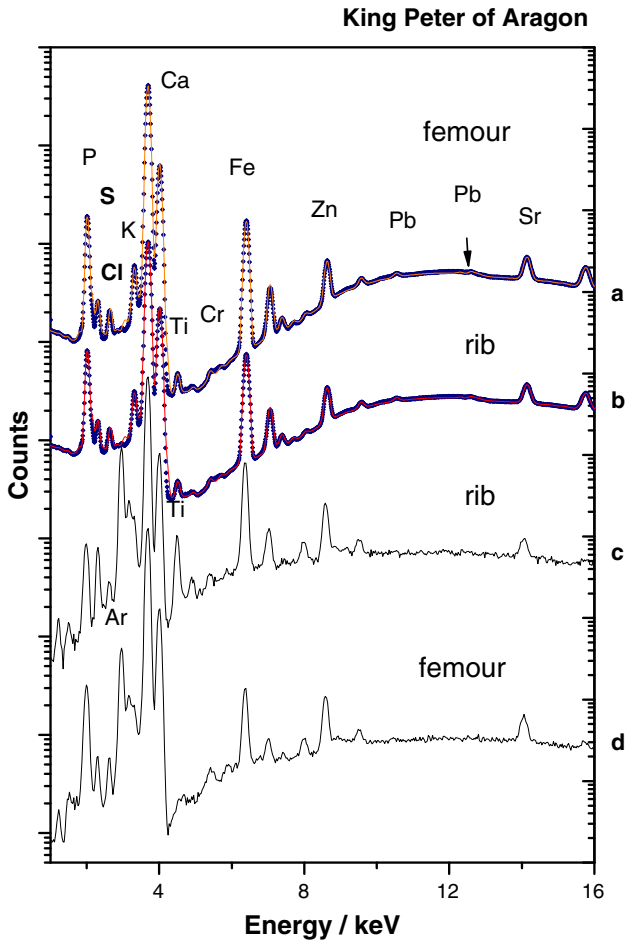

Fig. 6 X-ray fluorescence spectra taken at various points in the femur and rib samples of King Peter with a laboratory XRF spectrometer (curves *a* and *b*) and with a portable XRF instrument (curves *c* and *d*)

that has undergone a diagenetic process with the consequent loss of collagen (see curve Fig. 4d).

Rather, the profile of the two spectra of the Spanish Royals in the frequency range 1,300–1,700 cm^{-1} has some similarities with curve Fig. 4c belonging to a fresh human bone, which is known to have not undergone any significant diagenetic process This may also suggest that the organic part of the bones of the Royal bodies remained relatively well preserved into their coffins over the centuries.

4.2 (b) XRF analysis on Queen Blanche of Anjou and King Peter of Aragon fragments bones

Figure 5, top curve displays the spectrum obtained from a metacarpal bone belonging to Queen Blanche of Anjou, and the curve at the bottom shows a spectrum obtained by the portable equipment on femur bone sample.

The most evident peaks were obviously those from Ca and P, but we can appreciate at a variable extent degree the presence of S, Cl, K, Ti, Cr, Fe, Zn, and Sr. The attribution of lead may be controversial from curve A, because it stands on the credibility of the simultaneous presence of Pb Lα, β lines weak lines at 10.55 and 12.61 keV, respectively. In any case, the same couple of lines emerges rather clearly form spectrum B. Moreover, Al, Mg and Mn may also be envisaged at trace level. With regard to the elements that emerge from the bone samples of King Peter (see Fig. 6), we can note that the emission of Fe is relatively more intense compared to the Blanche of Anjou spectra in Fig. 5. This seems also to hold, but to a lesser extent, for the Zn lines, while the Pb emission is significantly less intense.

The bone samples of Blanche of Anjou and King Peter did not offer other significant differences in terms of major and minor elements.

Table 1 shows the quantitative elements obtained with Monte Carlo simulations. The presence at the surface of the

Table 1 Chemical content in bone samples belonging to King Peter of Aragon and Queen Blanche of Anjou (concentration in Wt%)

Element	*Queen Blanche of Anjou* (Metacarpus) Wt%	*Queen Blanche of Anjou* (Femur) Wt%	*Queen Blanche of Anjou* (Rib) Wt%	*King Peter* (Rib) Wt%	*King Peter* (Rib) Wt%	*King Peter* (Femur) Wt%
P	14.54	14.56	15.32	14.95	16.17	19,09
Mg	0.45	–	0.15	0.30	0.50	–
Al	0.20	0.22	–	0.20	0.25	0,31
S	0.72	0.48	0.37	1.53	1.02	1.00
Cl	0.72	0.70	0.75	–	–	0.50
K	0.51	0.51	0.73	0.92	0.92	0.74
Ti	0.03	0.03	0.08	0.02	0.05	0.02
Cr	0.01	–	–	0.01	0.01	0.01
Mn	0.01	–	–	–	–	–
Cu	0.01	–	–	0.01	0.01	0.004
Fe	0.09	0.10	0.20	1.17	1.44	0.76
Co	–	–	–	–	–	0.0005
Ni	–	–	–	–	–	0.005
Zn	0.08	0.07	0.06	0.15	0.26	0.15
Sr	0.18	0.18	0.15	0.22	0.45	0.11
Pb	0.12	0.13	–	0.05	0.02	0.01

Monte Carlo error is about 5 % of the value for concentration $>10^{-1}$; 10 % for concentration between 10^{-2} and 10^{-1}; 20 % for concentration between 10^{-3} and 10^{-2} and about 50 % for the concentration less than 10^{-3}

bones of several transitional and/or heavy metal elements in the case of unearthing is generally attributed to contamination from the soil or fossilization processes [10, 12, 28, 29].

In our case, due to the relative isolation of the coffins, also warranting a relative constant temperature and humidity, the conditions of bodies may be regarded virtually unchanged during the centuries, without significant incidence of external factors to a hypothetical process of bone contamination.

To understand the presence of observed elements, we can consider two hypotheses of contamination processes before burying that may have occurred *intra-vitam* or *post-mortem*. Actually forensic studies conducted at the Universitat Autonoma of Barcelona prove the existence of a body treatment and embalming practices consisting in a procedure without evisceration combined with applications of chemicals and plant to ensure the preservation of the bodies [17].

The presence of chlorine ions appears a bit puzzling but it can be reconducted to the presence of NaCl salt probably used in the bodies mummification processing deposited on the bone surface after the fleshy part consumption. A similar explanation can be advanced to explain the presence of K, Al and S. In fact, according to one of various formulae used in the Middle Ages for the mummification of bodies, at the end of the mummification process were closing all the bodily orifices with aloe, myrrh, alum, amber, musk, potassium nitrate and salt in equal parts [30].

The potassium alum compound is the hydrated potassium aluminum $KAl(SO_4)_2 12H_2O$ while the potassium nitrate is an ionic salt of potassium ions K^+ and nitrate ions NO_3^-.

Moreover, as it concerns the sulfur presence, we remind that a XRD analysis conducted on the linen fabric that covered entirely the body of King Peter of Aragon has shown a considerable amount of gypsum ($CaSO_4$-$2H_2O$) (see Fig. 7a, b), which explains coherently the elevated % of S that we found, especially in the external part of the King Peter's rib (1.53 %).

The observed moderate degree of contamination of the bodies was probably due to the change of burial in the case of King Peter and the desecration of the James II and Blanche of Anjou's coffin [18] involving unknown manipulations.

After these considerations, we believe that the significant presence of unattributed metals such as Fe, Zn and Pb to a very low extent may be related to the eating habits and use of Sn crockery metal, for example, which contains many of the transition metals. In particular, the presence of Pb traces is suggestive of true exposure at significant levels during the Royals lifetime.

The Middle Ages saw a marked increase in the use of Pb and Pb-containing products and there were occasional publications on the toxic dangers associated to the preparation and use of various metals [31]. Pewter, an alloy composed mainly of tin and, at that time, Ag and Pb, was commonly used in the Middle Ages to make tableware and kitchen utensils, for wealthy people in particular. The concentrations of Pb in bones from different historical periods have been reported by diverse authors [32–34] as

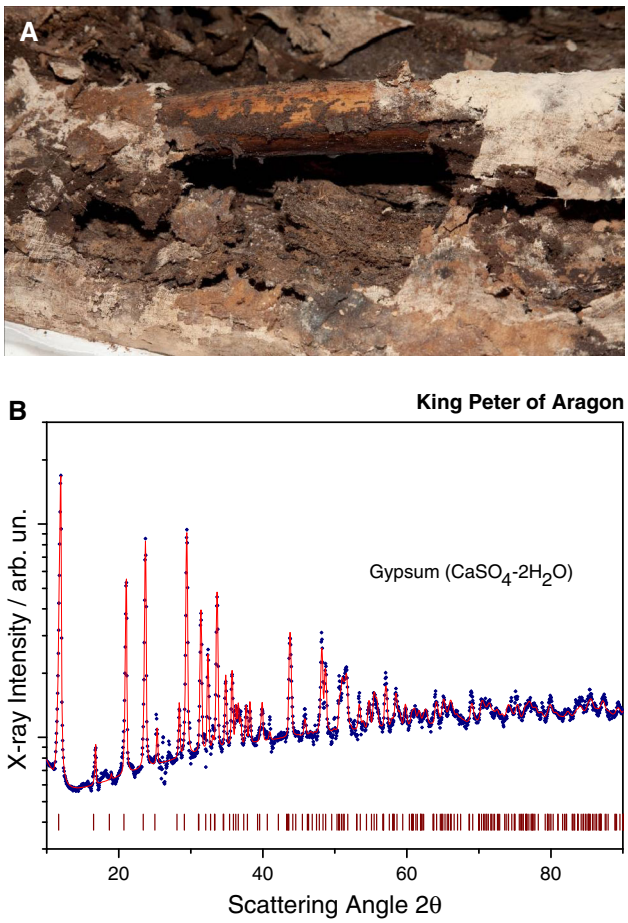

Fig. 7 **a** Particular of white linen fabric that covered the body of king Peter (photo courtesy of C. Aymerich and R. Maroto MHC-CRBMC). **b** The XRD patterns of gypsum found in the linen cloth that covered the body of King Peter of Aragon

evidence of metal intoxication but a comparison with our spectra is unfortunately unavailable.

Moreover, the slow cooking used in the Middle Ages, carried out in metal containers for foods and beverages with high acidic content, such as wine, could dissolve Pb and various metals (Ti, As, Cu, Fe, Pb, Zn), facilitating its release and ingestion. Pb-containing additives were also used to sweeten and preserve wine [31], and their consumption may ultimately have contributed to the Pb accumulation observed in the King's and Queen's bones.

5 Conclusions

In the present study, we have conducted a investigation through XRF spectroscopy on human bone samples belonging to Peter III of Aragon and Queen Blanche of Anjou that lived during the Middle Ages period. Although documentary historical sources and anthropological/forensic studies highlighted a number of changes of coffins and grave desecration, the bodies were found in very good condition. The XRF investigation showed the presence of several chemical elements despite the bodies have always remained isolated and well preserved inside their coffins.

The appreciable concentration of metals, transitional elements and elements normally unexpected in the bone composition can be related to the processes of bodies mummification and the dietary habits of the Royals, including tools used for cooking and for the consumption of food.

Acknowledgments The authors thank the Generalitat de Catalunya (2009, SGR 566) and the *Museu d'Historia de Catalunya* for supplying the osseous materials employed in this study. The authors also thank: Prof. Plinio Innocenzi and Dr. Luca Malfatti (Sciences and Nanotechnology Laboratory (LMNT), University of Sassari, Italy). This work is supported by Autonomous Region of Sardinia, with the project titled: *"Multispectral analysis of cultural heritage samples"*.

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