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Effects of moisture on historical buildings TL ages

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This contribution is part of a multidisciplinary study on the *Indirizzo* Thermal Baths in Catania aimed to deepen the historical and architectural knowledge and the chronological framework. The application of ThermoLuminescence (TL) dating on the samples collected from the site, aimed to establish the chronology of the fabric, offers a starting point for a discriminating evaluation about the moisture effect on the age results. The water content is one of the most important values needed for age estimation and it must be taken into account for the dose rate calculation. The main goal of the present study is putting in evidence the importance of the wetness in the age calculation. To this purpose, some brick samples collected at specific points of the *Indirizzo* thermal baths were analysed assuming different water content values. The influence on age results was then discussed.

1. Introduction

The ThermoLuminescence (TL) age is determined from the ratio of two quantities: the Equivalent Dose (ED) and the Dose Rate (DR). Considering an historical building, the ED is the total dose accumulated from the last firing of *terracotta* samples, measured using luminescence signals while the DR is the quantity of the dose absorbed in one year.

About ED evaluation, different methodologies are routinely performed [1–5]. The DR evaluation is based on the measurement of the dose absorbed by luminescence crystals, derived from the emitters of natural decay series of ^{235}U, ^{238}U, ^{232}Th, ^{87}Rb and of the ^{40}K radioactive isotope. Each DR value was obtained considering different contributions. In the component of D_α and D_β, DR is calculated through the measurement of the aforementioned radioelements concentration and so the application of specific conversion factors [6]. The D_γ component, instead, is determined *in situ* using ThermoLuminescent Dosimeters (TLDs) [7,8] and counting devices (gamma spectrometer) [9,10].

The accuracy of luminescence ages depends on a large number of parameters. In particular, the uncertainties in the Dose Rate measurement are often dominated by issues of water content. In age calculation, an average value for moisture over the whole burial time is usually assumed [11–14], but the lack of information concerning the extent of moisture variation throughout the burial period means that this is often one of the source of uncertainty in a luminescence age. Calculations can compensate for this effect, but they require an estimate of the water content throughout the burial period. This is a difficult parameter to estimate, but some constraints can be put upon the value. Several ways exist by which water content and its fluctuations through time may be established. The first approach is the direct physical measurement shortly after sampling, as this will provide a solid estimation of present water content [1]. The second approach regards especially the sediment and is to establish water content of dried-up samples by approximating the water uptake capability experimentally [15].

The relative uncertainty may be larger for older samples. It is more difficult, in fact, establish potential effects of climatic changes over long time spans. For some type of samples, the uncertainties associated with changes in water content and other complicating factors connected to the environmental dose rate can be eliminated [16–19].

The DR calculation is based on the assumption of infinite matrix concept typical of radiation physics field elaborated by Mejdahl [20]. This assumption is based on conservation of energy such that all the energy that is emitted in an infinite matrix, in terms of dimensions greater than the range of the considered particles, is also absorbed in this matrix.

However, the different content of water in the "infinite matrix" influences the attenuation of the absorbed energy by the crystals respect to the radioelements emission and this determines a DR

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decrease. The presence of water, in fact, in the sample and in the soil, reduces the radioactivity per unit mass compared to the dry situation, acting as an inert absorber of the radiation that could otherwise be involved in the production of TL within the mineral grains. The larger the amount of water, the less radiation is absorbed by the minerals. Furthermore, water has different absorption coefficients for alpha, beta and gamma radiation, in comparison to a ceramic matrix. Since the infinite matrix dose rates are usually calculated for the "dry" situation these factors have to be taken into consideration.

Zimmerman [1] estimated the effect of water content, in conjunction with the relative stopping powers or absorption coefficients for water and the matrix, to allow the formulation of correction factors relevant to alpha, beta and gamma radiation. This attenuation factors are crucial for a more representative DR estimation [1]. Following this approach, in order to take in to account water content of a sample, two parameters are experimentally obtained [21]:

$$W = \frac{saturation\ wet\ weight - dry\ weight}{dry\ weight} \quad (1)$$

and

$$F = \frac{average\ water\ content\ in\ the\ site}{saturation\ water\ content}. \quad (2)$$

The parameter W, that indicates saturation content, is obtained using a fraction of the sample dried (*dry weight*) and saturated of water (*saturation wet weight*).

The F value is related to the average water content of the sample during the period corresponding to the absorbing of the dose evaluated as ED. The determination of F has to made considering indications about climate conditions, cardinal directions, soil characteristics, contamination of the water capillarity from the ground [21].

We considered the environmental contribution D_{env}, coming from gamma radiation and cosmic rays, including attenuation due to water content because this component to DR was measured *in situ* [21]. While the D_α and the $D_\beta$contributions were corrected multiplying by the following h_α and h_β coefficients (from [21]):

$$h_\alpha = \frac{1}{1 + 1.5W \cdot F} \quad (3)$$

$$h_\beta = \frac{1}{1 + 1.25W \cdot F}. \quad (4)$$

Actually, the determination of an average humidity level, representative of the "life" of the sample, is still an open question.

Another important parameter is the particle size sample. About this argument, Clark carried out investigation of the effect of matrix water content variations by calculated effective Dose Rates for three different values of water content for isochron systems [22]. The finer grains are affected to a greater extent than the coarse grains this is due to the fact that the fine grains receive a greater proportion of their dose from alphas which are attenuated by water to a greater extent than betas or gammas. The rate of change in effective dose rate also appears to be greater at the finer end of the grain size scale. Banerjee showed that, in the case of sediments, the role of attenuation of external radiation by water may be even more pronounced because alpha radiation, which contributes as much as 40% to the total Dose Rate, is even more effectively attenuated by water [23].

Similarly, for the alpha efficiency, very important implications there are if one considers a case where the matrix water content changes during the burial time. If a sample becomes significantly wetter, or drier at some stage of the burial period then the stored dose may be composed of two (or more) periods of different effective dose rates [22].

The precise impact of water on Dose Rate, and hence the age, will vary from sample to sample [24]. For sediments, the estimation of water content since deposition presents a major source of uncertainty in the luminescence dating [25]. For caves, dunes and lakes, the uncertainty is likely to be fairly small, while for alluvial deposits and lacustrine environments it may be larger [24,26]. In this case, the water that attenuates external radioactivity can causes a lowering of the Dose Rate and could lead to shifts in calculated ages of >50% [27].

In the case of historical materials, the moisture content of the sample is often used as a guideline for age calculation but it is crucial that potential changes in the hydrological conditions are carefully considered during sampling. This is very important for TL dating historical buildings for which several studies put in evidence other difficulties related dating [11,12,28–30]. At first, the possibility, in the past, of materials reuse, leading to overestimated ages. Furthermore, the architectural destructions and reconstructions that can alter the Dose Rate and subsequent the calculated age.

Especially for archaeological site as thermal baths, all these considerations are very significant. In fact, for this type of building, more than for others, the evaluation of humidity level in function of the time is more difficult.

The aim of the paper is to give a contribution in a problem involving all researchers working in the historical buildings dating, above all, when fine grain fractions were used. Frequently, in fact, considering the difficult to evaluate the water content of the sample during its "life", the influence of moisture in the age calculation is not examined in depth [7,8,13,31] and the TL ages were calculated without moisture correction or, as already mentioned, with average or arbitrary values [11,12,14,32,33]. Only few studies take into account the possible variations of water content in bricks but in most cases for quartz grain fractions [34,35].

2. The *Indirizzo* thermal baths

The site object of the research represents one of the best-preserved thermal baths in the Roman Empire (Fig. 1). Wilson [36] has attributed the monument to the IV-V cent., while Branciforti [37] to the III-IV cent.

The monument is located in *Piazza Currò*, in the historic centre of the city, and were fed by river *Amenano*. The name is due to the adjacent church of *Santa Maria dell'Indirizzo* and to the near Carmelite convent, now used as a school. The church and the monastery, founded in the seventeenth century, were rebuilt after

Fig. 1. Southeast front of the *Indirizzo* Baths.