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Comparative analysis of dose rates in bricks determined by neutron activation analysis, alpha counting and X-ray fluorescence analysis for the thermoluminescence fine grain dating method

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HIGHLIGHTS

- Dose rates from natural U, Th and K in bricks were determined for the purpose of TL dating.
- Results from neutron activation analysis, alpha counting and X-ray fluorescence analysis were compared.
- Good match was observed for K determination.
- Systematically lower dose rates from U and Th were obtained from alpha counting.
- Further work will be pursued for determination by X-ray fluorescence analysis.

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ABSTRACT

In order to evaluate the age from the equivalent dose and to obtain an optimized and efficient procedure for thermoluminescence (TL) dating, it is necessary to obtain the values of both the internal and the external dose rates from dated samples and from their environment. The measurements described and compared in this paper refer to bricks from historic buildings and a fine-grain dating method. The external doses are therefore negligible, if the samples are taken from a sufficient depth in the wall. However, both the alpha dose rate and the beta and gamma dose rates must be taken into account in the internal dose. The internal dose rate to fine-grain samples is caused by the concentrations of natural radionuclides ^{238}U, ^{235}U, ^{232}Th and members of their decay chains, and by ^{40}K concentrations. Various methods can be used for determining trace concentrations of these natural radionuclides and their contributions to the dose rate. The dose rate fraction from ^{238}U and ^{232}Th can be calculated, e.g., from the alpha count rate, or from the concentrations of ^{238}U and ^{232}Th, measured by neutron activation analysis (NAA). The dose rate fraction from ^{40}K can be calculated from the concentration of potassium measured, e.g., by X-ray fluorescence analysis (XRF) or by NAA. Alpha counting and XRF are relatively simple and are accessible for an ordinary laboratory. NAA can be considered as a more accurate method, but it is more demanding regarding time and costs, since it needs a nuclear reactor as a neutron source. A comparison of these methods allows us to decide whether the time- and cost-saving simpler techniques introduce uncertainty that is still acceptable.

1. Introduction

Evaluating age from the equivalent dose and achieving an optimized and efficient procedure for dating archaeological materials generally requires an analysis of the internal and external dose rates from samples of dated materials and from their environment. The measurements described and compared in this paper refer to bricks from historic buildings and a fine-grain dating method. It is therefore not necessary to measure external doses from the environment (e.g., from the soil, as when dating archaeological finds of ceramics). The dose from the environment is negligible, if samples for dating are taken from a sufficient depth in the wall. The internal dose rate depends on the concentrations of natural radionuclides ^{238}U, ^{235}U, ^{232}Th, ^{40}K and on the water content in the dated material (Aitken, 1976, 1985; Goedicke et al., 1985). The alpha dose rate and also the beta and gamma dose rates must be taken into account in

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determining the internal dose (the procedure for fine grain dating of brick buildings is described, e.g., in Goedicke et al., 1985). The alpha dose rate is given by the natural concentrations of U and Th and their daughters, while the K concentration, together with U and Th and some contribution from cosmic rays, provides the beta and gamma dose rates. In the age equation algorithm, as described by Aitken (1985), the dose rate from the natural decay series can be calculated directly from the alpha count rate, without knowledge of the exact concentrations, but a fixed Th:U ratio must be assumed. A comparison of the dose rates from U and Th calculated from alpha counting and from their concentrations measured by NAA can provide information about the degree of approximation introduced by the use of alpha counting. In addition, we compared the concentrations of K measured by NAA and XRF, from which the dose rate from ^{40}K can be determined, and the reliability of XRF can be compared with the reliability of NAA.

Alpha counting with the Daybreak Model 583 Intelligent Alpha Counter and X-ray fluorescence analysis (XRF) were used in the laboratory at the Faculty of Nuclear Sciences and Physical Engineering (FNSPE). NAA can be considered to be more accurate, but it is more demanding regarding time and costs, since it needs a nuclear reactor as a neutron source. NAA was carried out in the laboratory of the Nuclear Physics Institute, which uses the LVR-15 research reactor. By comparing the dose rates determined on the basis of different methods, and using NAA as a "reference method", since NAA has recently been recognized as a primary ratio measurement method (Greenberg et al. 2011), we can decide how serious the uncertainty introduced by procedures accessible for an ordinary laboratory is, i.e., we can evaluate the suitability of the simpler methods for archaeological samples.

2. Experimental

Historical brick buildings are the dated objects, therefore the age equation algorithm, as mentioned in the preceding paragraph and displayed in Table 1, enables us to determine the dose rate directly from the alpha count rate. It is not necessary to compute the exact Th and U concentrations, but the fixed "most usual" Th:U ratio needs to be introduced into the calculation as an approximation of the real ratio. Since data from NAA was acquired in parallel, this approach can be replaced by the result calculated using the Th:U ratio obtained by NAA. We can therefore compare the dose rates from U and Th determined from alpha counting and NAA without using this approximation. In addition, K concentrations, which have been measured by NAA and XRF, contribute to the dose rate. The dose rates determined from the K concentrations obtained by these two methods were compared. In this case, the dose rate is calculated from the concentration using the same formula. The dose rate conversion factors (Adamiec and Aitken, 1998; Guérin et al., 2011), as shown in Table 2, were used in the age equation algorithm (Aitken, 1985). The question of disequilibrium in the uranium and thorium series due to radon escape during the dated period does not play a significant role, as the samples were taken from a thick compact wall. Factors for 0% Rn loss were therefore used.

2.1. Samples

Samples were collected at two historic sites—Hradec Králové (HK1–HK3) and Nymburk (NB1–NB5); in addition, samples NS1 and NS2 were prepared from modern brick and tile. The samples from Hradec Králové were taken from the wall of the gothic basilica, and the samples from Nymburk were taken from the remains of the town ramparts. All samples were crushed with a jaw crusher and a cup mill, and then sieved through a 100 μm sieve. Then homogenised samples with grain size <100 μm were analysed by alpha counting, NAA and XRF methods.

2.2. Alpha counting

Alpha counting was performed using the Daybreak Model 583 Intelligent Alpha Counter with two photomultipliers and scintillation ZnS:Ag screens (1.75″=4.445 cm in diameter, ZnS:Ag coated mylar screen, EJ-440 Eljen Technology). The phosphor layer is smooth and moderately robust. The wavelength of maximal

Table 2
Dose rate conversion factors.

Concentration[a]	α (Gy ka^{-1})	β (Gy ka^{-1})	γ (Gy ka^{-1})
1 mg kg^{-1} Th			
0% Rn loss	0.7375	0.0277	0.0479
100% Rn loss	0.3093	0.0094	0.0180
1 mg kg^{-1} U			
0% Rn loss	2.795	0.1457	0.1116
100% Rn loss	1.267	0.0603	0.0042
1% K		0.7982	0.2491
Alpha count rate (mm^{-2} Ms^{-1})[b]		β (Gy ka^{-1})	γ (Gy ka^{-1})
Th 0% Rn loss		0.0783	0.1366
Th 100% Rn loss		0.0732	0.1427
U 0% Rn loss		0.1210	0.0936
U 100% Rn loss		0.1241	0.0087

ka—Kiloannum, thousand years. Ms—megasecond, 10^6 s. It is assumed that the efficiency of the scintillator is 100%, *i.e.*, that every particle rises to an output pulse from the photomultiplier; however studies by Woithe and Prescott (1995) indicate that the efficiency of a ZnS screen may be only about 90%. The assumed electronic threshold setting is 85% for a sample containing only ^{232}Th, 82% for ^{238}U and 85% for ^{235}U. For 100% Rn loss, the values are 80% and 78%, respectively.

[a] Guérin et al. (2011).
[b] Adamiec and Aitken (1998).

Table 1
Dose rate equation algorithm summary.

Dose rate from U and Th		Dose rate from K
$D_{UTh}=D_{\alpha UTh}+D_{\beta UTh}+D_{\gamma UTh}$		$D_K=D_{\beta K}+D_{\gamma K}$
From alpha count rate	**From concentration**	
$D_{\alpha UTh}=1.78\times a\times\alpha_B$	$D_{\alpha UTh}=a\times(f_{\alpha U}\times U+f_{\alpha Th}\times Th)$	$D_{\beta K}=f_{\beta K}\times 0.997\times m$
$D_{\beta UTh}=k_\beta\times 0.997\times\alpha_B$	$D_{\beta UTh}=0.997\times(f_{\beta Uc}\times U+f_{\beta Thc}\times Th)$	$D_{\gamma K}=f_{\gamma K}\times m$
$D_{\gamma UTh}=k_\gamma\times 0.9\times\alpha_B$	$D_{\gamma UTh}=f_{\gamma Uc}\times U+f_{\gamma Thc}\times Th$	

Aitken 1985, D_{UTh}, D_K—dose rate from uranium, thorium and potassium [Gy a^{-1}],
a—annum, year. $D_{\alpha UTh}$, $D_{\beta UTh}$, $D_{\gamma UTh}$, $D_{\beta K}$, $D_{\gamma K}$—alpha, beta, gamma dose rate from uranium, thorium and potassium [Gy a^{-1}].
a—*a*-value. α_B—alpha count rate [ks^{-1} mm^{-2}]. k_β, k_γ—beta, gamma value for Th:U ratio.
$f_{\alpha U}$, $f_{\alpha Th}$, $f_{\beta Uc}$, $f_{\beta Thc}$, $f_{\gamma Uc}$, $f_{\gamma Thc}$, $f_{\beta K}$, $f_{\gamma K}$—conversion factors (Table 2) for alpha, beta, gamma radiation, uranium and thorium alpha count rates, and for uranium, thorium and potassium concentrations. U, Th—uranium and thorium concentration [mg kg^{-1}]. *m*—potassium content [w%].