

## Alpha dose rate calculations in speleothem calcite: values of $\eta$ and $k_{\text{eff}}/k_{\text{ref}}$

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The dose rate calculations for the age equation in ESR and TL dating must allow for the dependence of signal intensity produced by alpha radiation on alpha track length rather than on total energy deposited (Zimmerman, 1971; Aitken and Bowman, 1975; Lyons, 1988). Correction must also be made for any departure from linearity in the signal intensity vs track length response curve. In the  $k$  system of Zimmerman (1971) this is achieved by the use of  $k_{\text{eff}}$  instead of  $k$ , assuming the radionuclide chain is in equilibrium. In the  $a$  or  $b$  system (Aitken and Bowman, 1975; Bowman and Huntley, 1984) it is allowed for, firstly by using track length as the independent variable and, secondly, by applying the correction factor  $\eta$ .

At the 2nd International Symposium in ESR Dating and Dosimetry (held in München, October 1988), we presented the results of a study on alpha effectiveness including values for  $k_{\text{eff}}/k_{\text{ref}}$  and  $\eta$  in ESR dating for speleothem calcite which are up to 7% lower than the best estimates previously available (from the average of estimates for other materials). We believe these estimates to be the first direct calibrations for calcite. The magnitude of the difference that these correction factors will make to age estimates will depend, of course, on the relative contribution that the alpha dose makes to the total environmental dose-rate. Details of the methodology and details of calculation will be published in the Munich conference proceedings. However, because the error introduced by using the average estimate for other materials is systematic, we feel it is important that these figures be made available as soon as possible.

We used a small research nuclear accelerator as a source of alpha particles of various energies, as described in Lyons (1985, 1988), to determine the ESR response to alpha particles of different energies. Because of the nature of ESR measurement we were able to use alpha-thick instead of alpha-thin targets; the irradiating alpha particles were totally absorbed in the target, thus simulating the effect of internally produced alpha particles in a sample. This gave us the ESR response for alpha particles of the selected energies directly, without the need for further assumptions or calculations. In contrast, when thin targets are used,  $\delta$  signal/  $\delta$  range is measured and integrated to obtain the signal/range curve, and therefore will incorporate any uncertainty in

the signal response for low energies, the most difficult values to obtain experimentally (Bowman, 1976; Aitken and Bowman, 1975). A preliminary data set was presented in Lyons (1987, 1988). Here we present the full data set and summarize briefly the implications for alpha dose-rate calculations.

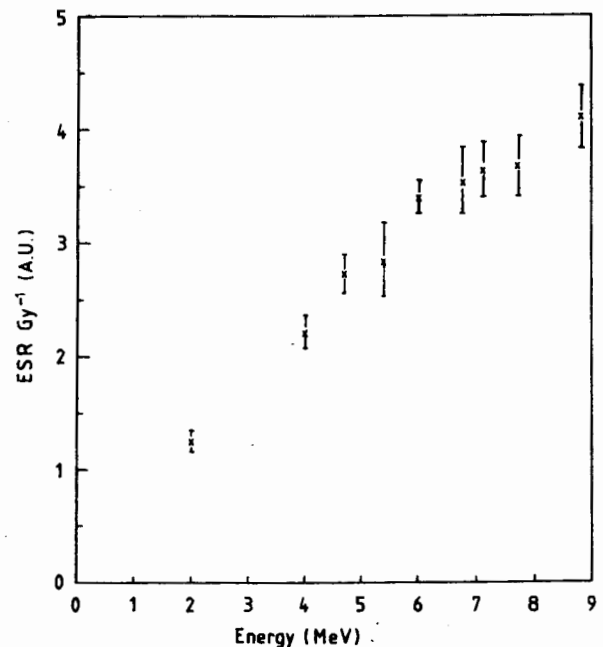


Figure 1. ESR per Gy versus energy of the incident alpha particles for a calcite speleothem sample.

Figure 1 shows that the ESR response per Gy of deposited energy depends on the energy of the irradiating alpha particle; Figure 2 shows that the total ESR response for an alpha particle which is completely absorbed in the sample, is an approximately linear function of the range of the alpha particle. Values for the range of alpha particles in calcium carbonate were calculated from data for stopping powers given in Zeigler (1977). Whereas the preliminary data set published in Lyons (1987) suggested that the response curve could be represented by a straight line passing through the origin, the full data set is best represented, for the range of energies occurring in the natural environment (i.e. for energies above 3 MeV), by a straight line with a non-zero intercept.

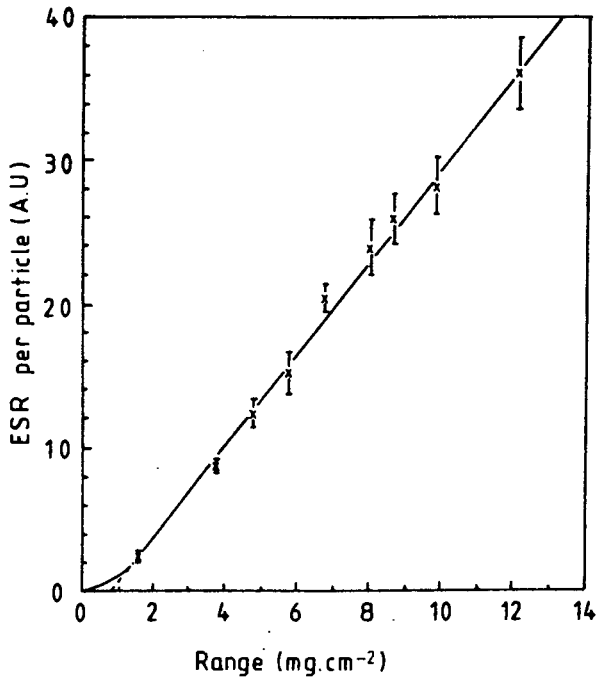


Figure 2:  
ESR per particle versus range of the incident alpha particle for a calcite speleothem sample.

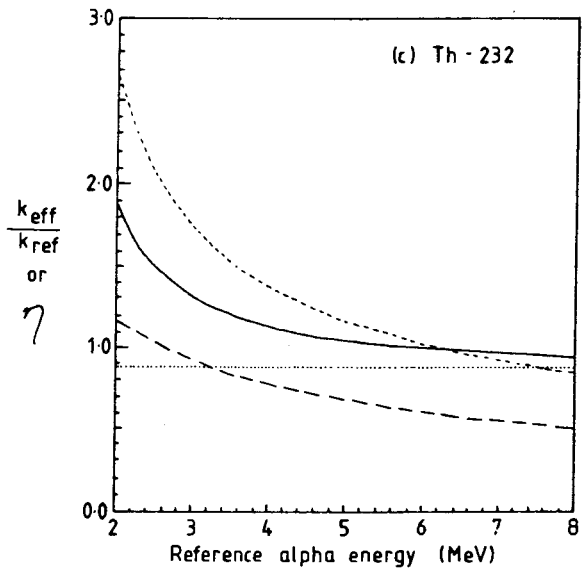
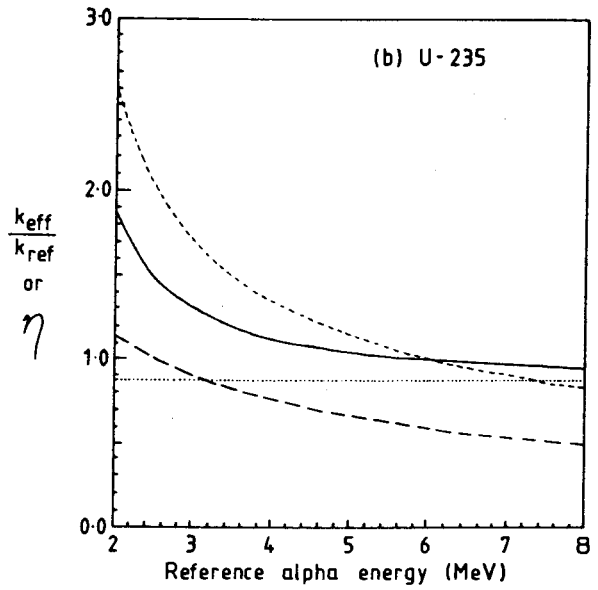
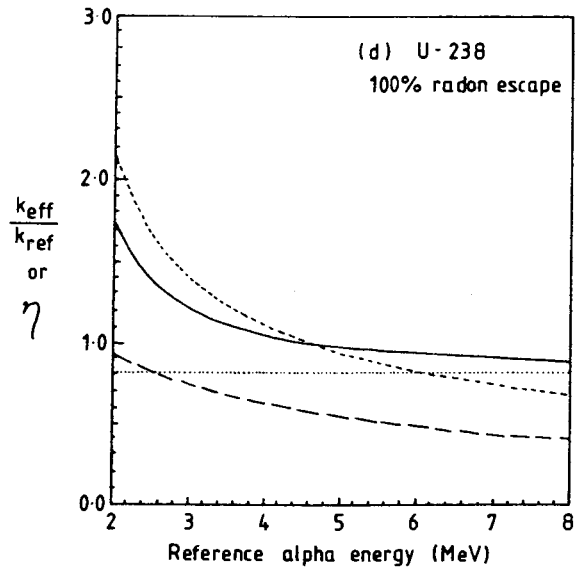
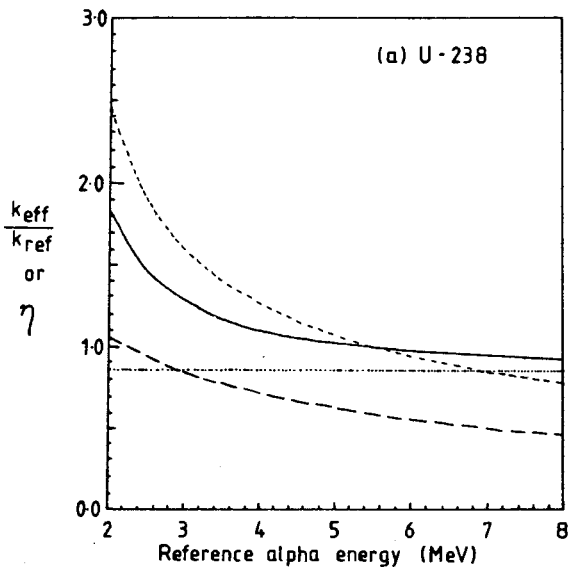


Figure 3:  
Correction factors for calcite, for thick and thin calibration targets, a or b, and k systems as a function of reference energy: (a) U-238 chain in equilibrium; (b) U-235 chain in equilibrium, (c) Th-232 in equilibrium, and (d) U-238, assuming 100% radon escape.

- $k_{eff}/k_{ref}$ , thick target calibration
- $k_{eff}/k_{ref}$ , thin target calibration
- ....  $\eta$ , thick target calibration
- .....  $\eta$ , thin target calibration



The implications for dose rate calculations are:

- (i) confirmation that track length, not energy, should be the fundamental variable in dose rate calculations for ESR as well as TL. The direct application of dose rate tables expressed in terms of energy, such as given in Bell (1979) or Nambi and Aitken (1986) is incorrect.
- (ii) a correction term should be subtracted from the track length for each alpha energy present, to allow for the non-zero intercept and thus give the 'effective track length' for use in the dose rate calculation. In the case of speleothem calcite, we estimate the subtraction term to be  $0.875 \text{ mg.cm}^{-2}$ . The uncertainty in this correction term is no more than 0.06 which corresponds to an uncertainty of approximately 1% in the effective track length for a typical alpha energy of 5 - 6 MeV. The effect of the alpha component on the dose rate is given in Table 1.

If the relative concentrations of the radionuclides present is known, an overall correction factor may be calculated. This correction factor will depend on the system used (*a* or *b*, or *k*), on the energy of the alpha particles used to determine the alpha effectiveness, and on whether alpha-thick or alpha-thin targets are used for calibration. Figure 3 is based on the data of figure 2. It presents the factors  $\eta$  and  $k_{\text{eff}}/k_{\text{ref}}$  as previously defined by Aitken and Bowman (1975) and Zimmerman (1971), respectively, as a function of reference energy for thick and thin target calibrations, for the U-238, U-235 and Th-232 chains in equilibrium and for the U-238 chain with 100% radon escape. It should be noted that, although  $\eta$  for a thin-target calibration appears to be independent of the calibration energy used, in practice experimental difficulties, particularly in ensuring a uniformly alpha-thin target for low energy calibration

alpha particles (less than 4 MeV), may lead to increased uncertainties and a serious underestimate of alpha effectiveness.

In Table 1, values for  $\eta$  and  $k_{\text{eff}}/k_{\text{ref}}$  for calcite from this study are compared with commonly quoted values for other materials in the literature. Using the *a* or *b* systems, the new correction factors give an estimate of effective alpha dose rate for the complete U-238 chain in calcite that is 6% lower than the best previously available estimate, and for the radon escape case, one that is 5% lower. For the *k* system the revised estimate is 7% lower. The effect on age estimates will depend, however, on the proportion of the environmental dose rate due to alpha irradiation. Note that the values from this study for calcite are not inconsistent with previous estimates for other materials but are more precise, with an uncertainty of less than 2%, as the use of thick rather than thin targets eliminates the uncertainty in all data points due to inadequate knowledge of the slope of the signal vs range curve for low energies. Bowman (1976) found values for different materials ranging from 0.84 - 0.95 and Aitken (1985) proposes an average value of  $0.90 \pm 0.05$  for use with all archaeological dating materials; the error bar partly reflects the variation between materials and partly the difficulty of using thin targets for calibration.

We have also calculated the variation in  $\eta$  and  $k_{\text{eff}}/k_{\text{ref}}$  due to the accumulation of daughter products with time, assuming thorium and its daughters are not initially present, as is usual with speleothem calcite (figure 4). If a systematic error of this magnitude is unacceptable, then the age equation should include the concentrations of the daughter products together with their appropriate correction factors as outlined above.

Parameter	This paper (calcite)	Previous estimates (other materials)	Difference in effective alpha dose estimate
<b>Equilibrium Case</b>			
$\eta_{\text{thin}}$	0.85	0.90	- 6%
$(k_{\text{eff}}/k_{\text{ref}})_{\text{thin}}$	0.75	0.80	- 7%
$\eta_{\text{thick}}$	1.14		
$(k_{\text{eff}}/k_{\text{ref}})_{\text{thick}}$	1.34		
<b>100% Radon Escape</b>			
$\eta_{\text{thin}}$	0.81	0.86	- 5%
$(k_{\text{eff}}/k_{\text{ref}})_{\text{thin}}$	0.66		
$\eta_{\text{thick}}$	1.09		
$(k_{\text{eff}}/k_{\text{ref}})_{\text{thick}}$	1.18		

Table 1: Comparison of correction factors for calcite (this study) with previous estimates for other materials, for the U-238 chain.  $\eta$  values are average values for several different materials from Aitken and Bowman (1975). The *k* ratio, taken from Zimmerman (1971), is for Norwegian quartz, an energy of 3.7 MeV being used for  $k_{\text{eff}}$ .

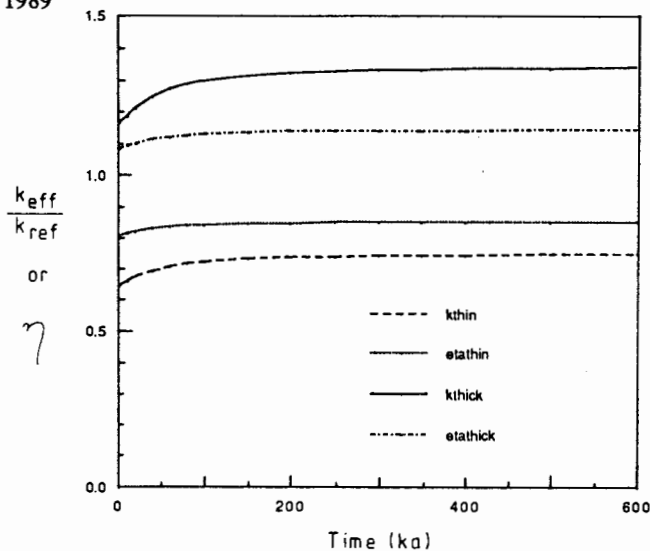


Figure 4: Variation of  $\eta$  with time for the U-238 chain in calcite, assuming initial concentrations of thorium and its daughters to be zero.

As alpha effectiveness is most probably dependent on the stopping power of the material for different energies (and thus on the form of the range vs energy curve) and on the saturation properties of the material, it should not be assumed that the above values are definitive for other dating materials. The experimental data from this study is specific to calcite and has smaller uncertainties than the average estimates previously available: we suggest that these values be used for calcite and aragonite (which have the same range vs energy curve as calcite), rather than average values.

#### Acknowledgements

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#### PR Reviewer's comments (M.J. Aitken)

Measurement of alpha effectiveness using thin layers of sample (as is necessary for TL) is made difficult by the tendency of fine grains to agglomerate on deposition, as the authors mention. It therefore reflects well on the experimental work of Zimmerman (1971) and Bowman (1976) that the values they obtained are close to the values now found by the authors using the thick sample technique (as is possible with ESR).

While the effect on dating is barely significant, this work is important not only in substantiating the values of Zimmerman and Bowman but also in giving further confirmation of the validity of the 'TL/ESR per unit track length' model. We look forward to learning the actual  $a$ -,  $b$ -, or  $k$ - values determined by the authors for ESR and a definitive comparison with TL values.