



THICK SOURCE ALPHA PARTICLE SPECTROSCOPY: POSSIBILITIES AND PROSPECTS

C.T. Michael¹, N. Zacharias^{2,1} & A. Hein¹

*¹Laboratory of Archaeometry, Institute of Materials Science, NCSR Demokritos,
153 10 Ag, Paraskevi, Attiki, Greece*

*²Laboratory of Archaeometry, Department of History, Archaeology and Cultural Resources Management,
University of Peloponnese, Old Camp, 24 100 Kalamata, Greece*

Corresponding author: ctmichael@ims.demokritos.gr

ABSTRACT

The new technique for the calculation of U and Th concentration which is based on the alpha particle spectrum taken from a thick sample by using a silicon detector is briefly described. Within the present study two major advantages of the technique will also be presented: the potentiality for detecting and providing an estimation of disequilibrium in the U and Th series -when present- for especially young sediments, and the potential use of the technique as a new method for isotopic dating of speleothems and other materials. Also the validity of the basic equation and the accuracy of this technique is tested.

KEYWORDS: alpha counting, PIPS, secular equilibrium

INTRODUCTION

Alpha counting techniques are widely used for dose rate determination in Luminescence and ESR dating applications (Soumana et al., 1997; Sjostrand and Prescott, 2002; Turner et al., 1958.). Since in many cases the major part of the natural radiation dosage absorbed by the crystals of the samples to be dated is due to the U and Th series, an increased accuracy is required. It is also known that the dose rates calculated by alpha counting are more accurate even in the case of an unknown disequilibrium. Nevertheless, an approximate knowledge of disequilibrium increases the accuracy of the calculated dose rates. The presented technique is an improvement and a further development of a previously described new technique (Michael and Zacharias, 2000) for the calculation of U and Th concentrations based on the alpha-particle spectrum taken from a thick sample by using a silicon detector which gives also the possibility for the estimation of disequilibrium in our samples (Michael et al., 2008). This possibility can be used for U-series disequilibrium dating methods (Liritzis, 2006) for speleothems and other materials which will be the subject of a future paper. The sample preparation, the instrumentation and calculations are explicitly described in the first two cited papers.

CONCENTRATION AND DISEQUILIBRIUM ESTIMATION

The basic equation of this technique is the following

$$(1) \quad N_{mi} = \frac{4n_{Int}}{R_1 - R_2}$$

where N_{mi} (Bq/g) gives the number of alpha particles emitted by the radionuclides of the U and Th series (activity concentration, a.c.) per unit mass per unit time (s) with energies higher than a certain value E (MeV); n_{Int} is the integral of counts per unit surface (cm^2) per unit time (Ks) in the energy interval between E_1 and E_2 and R_1, R_2 (mg/cm^2) are the ranges in the sample corresponding to E_1 and E_2 , respectively. This interval can be anywhere between E and the next lower energy of the emitted alpha-particles by the two series taken from spectra as in Fig. 1, which corresponds to the alpha particles emitted from a modern thick sample of water salts. It is evident that this interval must not include an energy which corresponds to alpha-particles emitted by either of the two series. Since the alpha-particles emitted by the two series have 15 different energies (8 from U series and 7 from Th series) the number of intervals is 14. The values N_{mi} for all these intervals plus one more interval between 0 and the lowest alpha-particle energy can be calculated from Eq. (1), where i takes values from 1 to 15.

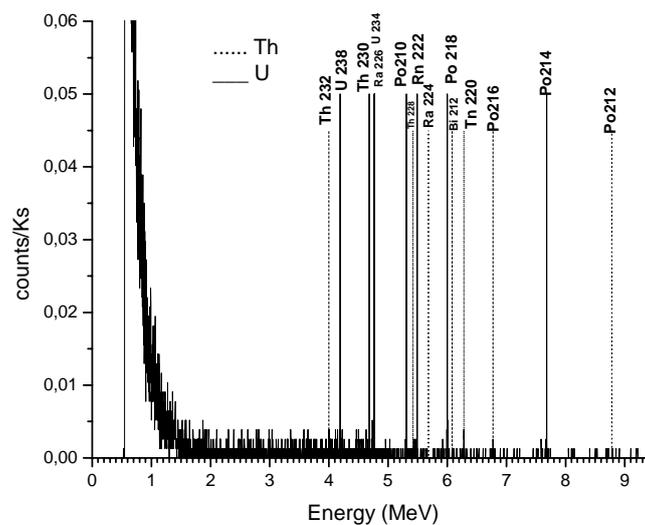


Fig. 1. The alpha particle spectrum taken from a thick sample of water salts. The solid lines correspond to the energies of the emitted α -particles of the U series and the dotted of the Th series. The rapid increase of counts below 1.8 MeV is due to β -particles and electronic noise.

N_{mi} are plotted versus energy as steps in Fig. 2 (thick line), where N_{m1} corresponds to the a.c. of ^{212}Po , N_{m2} to the a.c. of $^{212}\text{Po} + ^{214}\text{Po}$ and so on. Finally, N_{m15} corresponds to the sum of a.c. of all the daughter nuclides of the two series.

In case of equilibrium the a.c. N_U and N_{Th} of ^{238}U and ^{232}Th , respectively, are given by

$$N_U = (N_{m15} - 9.36N_{m1})/8 \quad (2)$$

and

$$N_{Th} = N_{m1} / 0.641 \quad (3)$$

from which the corresponding concentrations in ppm

$$C_U = N_U / 0.0124 \quad (4)$$

and

$$C_{Th} = N_{Th} / 0.00406 \quad (5)$$

are given.

In the case of disequilibrium in U series (for Th series usually disequilibrium can be excluded due to the short half-lives of all daughter nuclides with respect to parent) the value of N_U is given by

$$N_U = \frac{N_{m15} - 9.36 N_{m1}}{2 + t + r + 4rn} \quad (6)$$

where t and r are the disequilibrium fractions of ^{230}Th and ^{226}Ra , respectively, with respect to the series parent ^{238}U , so that the corresponding a.c. of these two nuclides are $N_{230\text{Th}} = t N_U$ (7) and $N_{226\text{Ra}} = r N_U$ (8), respectively, and n is the disequilibrium fraction of ^{222}Rn with respect to its parent nuclide ^{226}Ra , so that its a.c. is $N_{222\text{Rn}} = r n N_U$ (9). In case of equilibrium $t = r = n$

$= 1$ and thus Eq.6 becomes the same with Eq.2. The theoretical N_{mi} diagrams presented in Fig. 2 allow for an approximate quantitative estimation of disequilibrium in our samples by comparing them with the experimental N_{mi} (thick line) diagram resulting from Eq. 1. For this comparison the theoretical N_{mi} diagrams are calculated by giving different values t , r and n , which correspond to the different most usual cases of disequilibrium, and calculating the successive a.c. N_{m1} , N_{m2} , ... N_{m15} from Eq. 3, 6, 7, 8, 9. This can be easily done using a software program until for some values of t , r and n we achieve the best possible fit of the theoretical with the experimental diagram. This procedure is analytically described in Michael et al., 2008. In Fig. 2 the thin line (Equilibrium) corresponds to the theoretical N_{mi} diagram for the case of equilibrium and the dotted line (Disequilibrium) corresponds to the theoretical N_{mi} diagram for the case of 100% ^{230}Th and 100% ^{226}Ra deficiency. In the latter case, for which $t = r = 0$ and n any value, there is also 100% deficiency for the ^{222}Rn and for all its successive daughter nuclides, because these follow after ^{226}Ra and have short half-lives. In steps 15th and 1st the experimental and theoretical diagrams coincide always because the calculations of N_{mi} in all theoretical cases are based in the experimental values of N_{mi} in these two steps (Eq. 2, 3, 6).

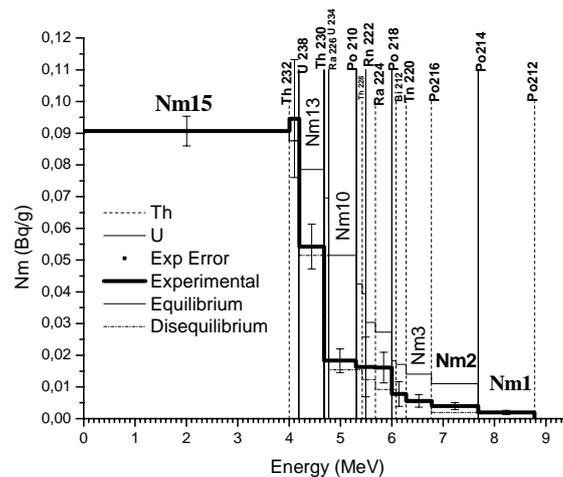


Fig.2. Diagram of experimental N_{mi} resulting from the spectrum of Fig. 1 (thick line) and the corresponding theoretical diagrams for the cases of equilibrium (thin line) and for 100% ^{230}Th and 100% ^{226}Ra deficiency disequilibrium (dotted line).

From the three diagrams for the water salts of Fig. 2 it is evident that the experimental N_{mi} diagram is much closer and within the calculated error to the disequilibrium diagram than that of equilibrium and thus we can conclude that for these water salts there is deficiency of about 100% ^{230}Th and 100% ^{226}Ra in the U series. This result is expected because our sample of water salt was modern and taken from a modern water heater. The calcite crystals in our sample, as like the calcite crystals of speleothems, are formed from calcium carbonate in water in which there is usually U present but no Th, due to the lower solubility of Th. Some of the U is incorporated in the calcite crystals and through radioactive decay there is gradual accumulation of ^{230}Th . Thus by estimating the ratio of $^{230}\text{Th} / ^{234}\text{U}$ using this technique, it is possible to have an estimate of the time that has elapsed since crystal formation.

ACCURACY TESTING

In a recent paper (Michael et. al., 2010) it was estimated that by this new technique it is possible to measure U concentration of some few decimals of ppm, as the calculated systematic errors are lower than 0.16 ppm and for Th concentration the calculated systematic error is 0.35 ppm. These estimations were based on a perspex cover of the Si detector with activity concentration between 0 and 0.015 Bq/g which was estimated using a suitable technique described in the above paper. The systematic errors could be reduced in case we find a material with lower activity than the real activity of the perspex or if we can know its real activity with a relatively good accuracy. Recently, iron disc newly constructed was used as a cover with an activity concentration which was estimated less than 0.005 Bq / g, that is 3 times lower than that of perspex, resulting in a respective decrease of the calculated systematic errors. This means that the calculated systematic errors have restricted to about 0.05 ppm for U and 0.12 ppm for Th. These systematic errors could be calculated even less if we find an other material with lower activity concentration than the iron disc used.

As mentioned in Michael et al (2010), this new method has the advantage of being self-tested for the accuracy of the calculated concentrations for each sample. The main criterion is based on whether the estimated a.c. N_{mi} for the same sample remain equal, within the calculated errors, if the calculations are made in various parts of the alpha particles spectrum (regions of interest, ΔE) and within the same of the 15 energy intervals mentioned above. It is obvious that for our measurements being correct the calculated N_{mi} should be stable, i.e. there is a plateau region in our results, as these are referring to the same amount. This criterion should be tested mainly for the intervals 0-4.01 MeV and 7.683-8.78 MeV in which the values of N_{m15} and N_{m1} , respectively, are calculated, as these values are the only needed for the U and Th concentration calculations in case of equilibrium.

In case of disequilibrium this criterium should be tested also for other intervals. In Fig. 3. the a.c. for the in-house standard (Pikermi clay, Hein et al., 2002.), used for inter-laboratory calibration, is presented, calculated in the different regions of interest ΔE 0.5-1, 1-1.5, 1.5-2, 2-2.5, 2.5-3, 3-3.5, 3.5-4, versus the mean value for each of these energy regions. We notice that there is a plateau region from 1.5-4 MeV. For ΔE in lower energies the calculated a.c. is higher due to β -particles and electronic noise.

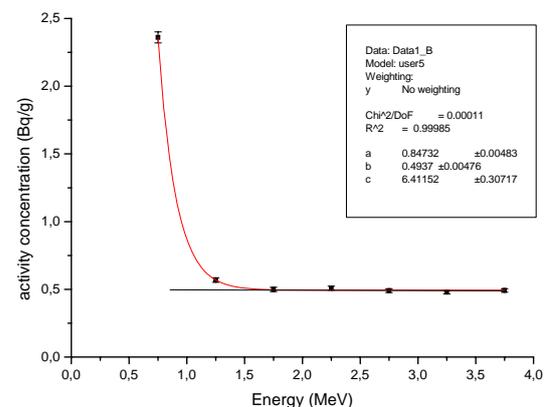


Fig.3. The activity concentration for the sample Pikermi calculated in the different regions of interest 0.5-1, 1-1.5, 1.5-2, 2-2.5, 2.5-3, 3-3.5, 3.5-4, versus the mean value for each of these energy regions.

In Fig. 4 similar results are found for the case of water salts mentioned above, but for a shorter region (2.5-4 MeV).

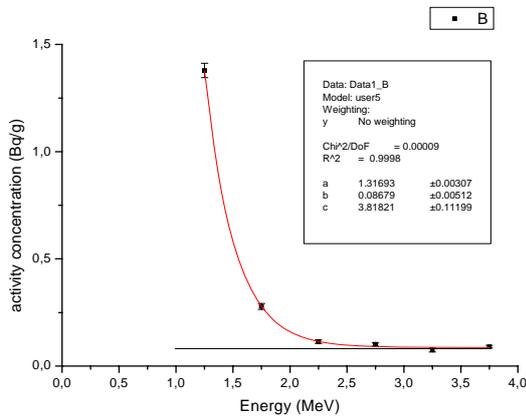


Fig. 4 The activity concentration for the sample of water salts of Fig. 1 and 2 calculated in the different regions of interest ΔE 1-1.5, 1.5-2, 2-2.5, 2.5-3, 3-3.5, 3.5-4, versus the mean value for each of these energy regions.

In Fig. 5 the calculated U concentrations for the sediments V4, Y and W1 are presented for different initial values E_1 of ΔE , while the last E_2 remains 4 MeV for all. It can be seen the calculated values are correct only for the E_1 values higher than 2 MeV, because of the stability of the results for these values, which is obviously due to the stability of N_{m15} in this region. For E_1 lower than 2 MeV the calculated concentrations are not correct because they increase as E_1 decreases due to beta-particles and electronic noise.

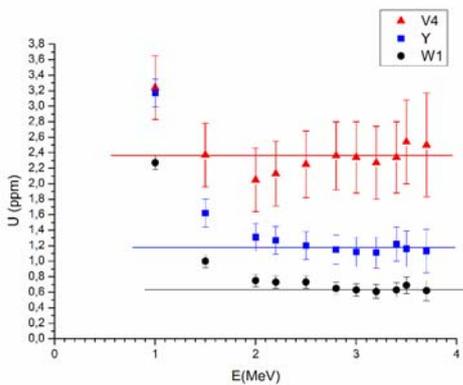


Fig. 5 U concentrations (ppm) calculated for different energy regions ΔE (MeV) of alpha particle spectrums of the sediments V-4, Y and W-1 versus initial value of ΔE while the last is 4 MeV for all .

In some cases samples usually of low concentrations are presenting insufficient stability (plateau) of the calculated results (Fig. 6, 7 and 8).

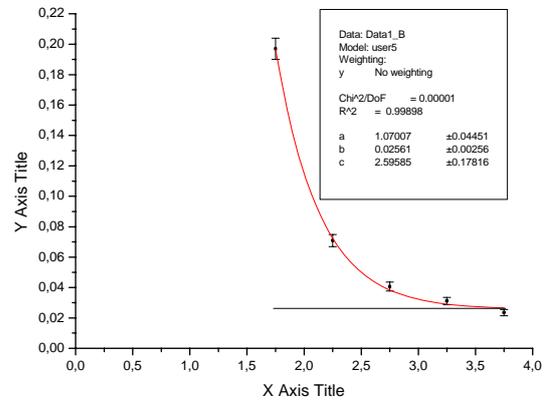


Fig. 6 The activity concentration for a sample of flowstone calculated in the different regions of interest ΔE 1.5-2, 2-2.5, 2.5-3, 3-3.5, 3.5-4, versus the mean value for each of these energy regions.

This was the subject of intense study from which it was concluded that this is probably due to fractionation during the preparation of the sample or for another reason, for example dust on the surface of the sample, causing a decrease or increase of a.c. in a very thin layer on the surface of the sample. Another reason could be radon (^{222}Rn and ^{220}Tn) escape from the surface of the sample causing also a decrease of a.c. in a very thin layer on the surface of the sample. In Fig.6 the calculated a.c. for a sample of flowstone, taken from the cave of Diros in Peloponnese, in the different regions of interest ΔE 1.5-2, 2-2.5, 2.5-3, 3-3.5, 3.5-4, versus the mean value for each of these energy regions, are presented, from which it is observed that there is no stability of the results and thus these results are not accurate. A possible reason could be a fractionation during the preparation of the sample or radon escape due to the porosity of this sample. The flowstone consists of different materials and mainly calcite, which includes soil grains with aluminum silicate compounds in which the concentration of U and Th is much higher than in calcite. In Fig. 7 there is a diagram similar to the diagrams of Fig. 3, 4 and 6. From this diagram it is indicated that when on the surface of the samples there is a substance of lower a.c.,

as in this diagram, in which the results for a sample of Pikermi, covered with a layer of flowstone of about 0.6 mg/cm^2 , are presented, the calculated a.c. decrease for ΔE of higher energies (the a.c. of flowstone is much lower than Pikermi). On the other hand in Fig. 8 it is indicated that when on the surface of the sample there is a substance of higher a.c., as in this Fig., in which the results for a sample of perspex covered with a thin layer of Pikermi are presented, the calculated a.c. increase for ΔE of higher energies (the a.c. of Pikermi is much higher than of that when using the perspex).

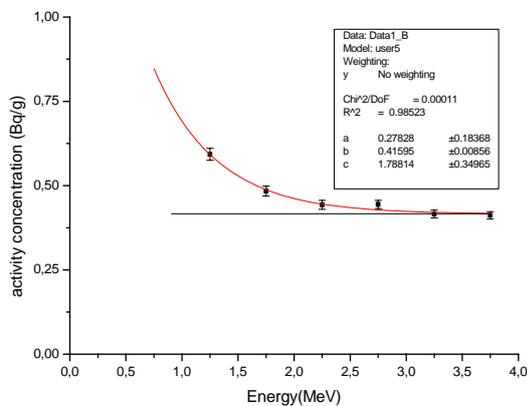


Fig.7. The activity concentration for the sample of Pikermi covered with flow stone for the values of ΔE in the different regions of interest 1-1.5, 1.5-2, 2-2.5, 2.5-3, 3-3.5, 3.5-4, versus the mean energy of these energy regions.

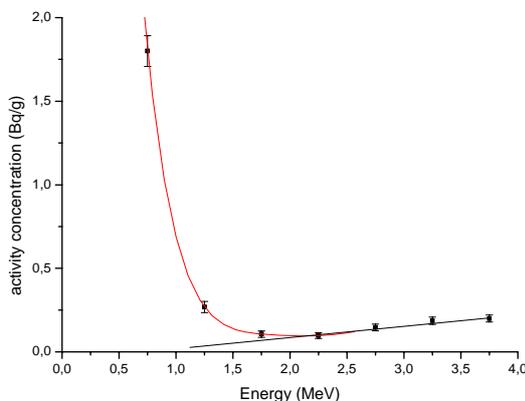


Fig.8. The activity concentration for the sample of perspex covered with a thin layer of Pikermi for the values of ΔE in the different regions of interest 1-1.5, 1.5-2, 2-2.5, 2.5-3, 3-3.5, 3.5-4, versus the mean energy of these energy regions.

DISCUSSION OF THE RESULTS

The present experimental results show that in order to use this technique for isotopic dating for speleothems and other materials much effort should be done for avoiding fractionation during the sample preparation procedure. These measurements also show that this technique has the additional advantage, in comparison with the alpha counting pairs technique, except those referred in the previous papers, of detecting any surface differentiation which could introduce inaccuracy in our measurements, even of a very thin layer of the order of some decimals of mg/cm^2 , undetectable by other means. This advantage could be useful when the U and Th should be estimated by a non distractive method for which grinding of the samples should be avoided. The problem of fractionation is more important when the measured sample incorporates inclusions of very different a.c. than the rest material.

For speleothems in order to have accurate results the percentage of soil should be very low. This is valid for two reasons, first for avoiding fractionation, and second for better accuracy in the comparison between the experimental diagram and the theoretical diagrams, corresponding to the different disequilibrium cases, in order to find the disequilibrium state in our sample. It should be taken into account that in nuclear counts the error is the square root of the counts, i.e. increases as the counts increase. Therefore, we have increased error when the soil content and thus the Th content is increased compared with that of U, which gives the important counts for our measurements, since in the soil the Th concentration is usually 3 times that of U. This happens because in the total a.c. of the sample the a.c. of the Th series is added. In this case the error of the experimental N_{mi} values is increased. This has as a result the comparison of the theoretical with the experimental N_{mi} diagrams to be not possible in case the difference between these two diagrams is small and within the errors.

For the water salts measured the calculated concentration of U is 1.76 ± 0.20 ppm and of Th is 0.86 ± 0.30 ppm. Also for the flowstone the calculated concentration of U is 0.13 ± 0.35 and of Th is 0.78 ± 0.24 . Thus for the water salts the

ratio of Th/U is about 0.49 while for the flowstone is about 6. The low ratio for water salt is due to the fact that there is no soil in this sample. Another advantage of this sample is that its a.c. is much higher than the systematic error of our detector. These two advantages are the reason why we have satisfactory and clear results for the disequilibrium state of this sample, from which we conclude that the ratio $^{230}\text{Th} / ^{234}\text{U}$ is

about 0 and therefore the sample is very new, as it is expected. On the other hand the results for the flowstone are not clear due to the much higher ratio of Th/U and also to the low a.c. which for the U falls within the error. For these cases of low U concentration a better detector with lower background would be necessary in order to have satisfactory results.

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