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# ATTENUATION FACTORS FOR THE ABSORBED RADIATION DOSE IN QUARTZ INCLUSIONS FOR THERMOLUMINESCENCE DATING

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## ABSTRACT

The dose attenuation factors for alpha and beta radiation passing through a 100 micron quartz grain are presented here and the dose dilution due to isotropic etching of the grains in hydrofluoric is also described.

## ABSORBED DOSE FROM THE NATURAL RADIOACTIVE SERIES

The dose-rate conversion factors given previously in Bell (1977 and 1979) do not describe fully the actual radiation dose which effectively gives rise to TL from quartz grains embedded in a radioactive matrix. Two other factors which must be taken into consideration are (i) the relative inefficiency of the alpha particles compared to beta and gamma radiation at inducing TL, and (ii) the absorbed dose attenuation for both alpha and beta particles passing through inclusions of diameter comparable to or greater than the particle range in the absorber.

The relative inefficiency of alpha particles at inducing TL (compared to an equivalent dose of beta or gamma radiation) has been described in detail elsewhere by Zimmerman (1971 and 1972) and by Aitken and Bowman (1975) and the discussion shall not be repeated here. However, these efficiency factors must always be taken into account when determining the archaeological dose to a quartz grain.

The second factor, the grain-size dose attenuation, depends on both the type of radiation and on the size of the grains. In the 'fine-grain technique' of Zimmerman (1971) only grains in the size range 1-8 µm are used and this is much less than the range of alpha particles (typically of the order of 25 µm ) and more than two orders of magnitude less than the range of beta particles (typically a few millimetres) in TL minerals. It is consequently reasonable to assume that grain-size attenuation factors are negligible for beta radiation in the 'fine grain technique' and are likely to be very small for alpha radiation (see Bell, 1979a).

In contrast the 'quartz inclusion technique' of Fleming (1970) utilises quartz grains of average diameter approximately 100 μm and for these grains the alpha dose and to a lesser extent the beta dose will suffer significant attenuation. This dating technique also requires the grains to be etched in hydrofluoric acid and this treatment will cause a further reduction in the alpha and beta doses. The above-mentioned factors have all been described before in various reference sources, but the purpose of this paper is to bring all of the information together in an easily usable form and to update previous calculations where necessary.

## ALPHA DOSE ATTENUATION

Spiers (1953) first studied the problem of the energy deposited in one medium due to particles generated in another and Kononenko (1957) studied the absorbed dose in tissue cavities due to alpha-particle emitters in surrounding bone. Both workers made the assumption that the linear energy transfer (LET), that is the energy deposited at any point along the track of a single charged particle, is independent of the particle energy. Charlton and Cormack (1962) derived expressions for the absorbed dose in tissue cavities in bone taking account of the dependence of LET on alpha energy and Howarth (1965) developed methods for calculating these expressions and gave tabulated values.

Put simply, the absorbed dose, $D_\alpha(x)$, at a point distant x from the interface between the emitting and absorbing media is given by

$$D_\alpha(x) = \frac{n_o E_o}{{}_mS} G_\alpha(x/R_o) \qquad (1)$$

where $n_o$ is the number of alpha particles, of initial energy $E_o$ and range $R_o$ in the absorber, emitted per unit mass of the emitter and ${}_mS$ is the ratio of the mass stopping powers of the emitter and absorber. $G_\alpha(x/R_o)$ is a geometrical function which depends directly on the shape of the interface. The form of $G_\alpha(x/R_o)$ for a spherical interface is described fully by Howarth (1965) and this form, $S_\alpha(x/R_o, D/R_o)$, where D is the diameter of the spherical interface, was used in the present calculations. The absorber is taken to be the quartz grain immersed in a clay matrix with a uniform distribution of alpha sources which is taken to be the emitter. The absorber and emitter are assumed to have very similar mass stopping powers which implies ${}_mS$ is unity.

$D_\alpha(x)$ must be integrated over the volume of a grain in order to evaluate the mean absorbed alpha dose in the grain, $\overline{D_\alpha(Abs)}$. This integration has been performed numerically for a 100 μm quartz grain (full details of this and the other calculations found later in this paper are given in Bell, 1978) and the results for the two alpha emitting series are:

$$\text{Uranium series,} \quad \overline{D_\alpha(Abs)_U} = 0.215 \times \frac{n_o E_o}{m^S} \tag{2}$$

$$\text{Thorium series,} \quad \overline{D_\alpha(Abs)_{Th}} = 0.248 \times \frac{n_o E_o}{m^S} \tag{3}$$

This means that a 100 μm quartz grain will absorb only 21.5% of the alpha dose that would be absorbed from the uranium series by an infinitesimally small grain and 24.8% of the dose from the thorium series. Fleming (1969, 1970) has also calculated the mean absorbed alpha dose for the uranium and thorium series using a similar method to that described above. He approximated the integration of $D_\alpha(x)$ over the volume of a grain by the use of summation techniques rather than the more accurate numerical integration used here. Nevertheless, his results were 21% and 25% for the uranium and thorium series respectively which are in very good agreement with equations 2 and 3 above.

## ALPHA DOSE REDUCTION BY HYDROFLUORIC ACID ETCHING

Fleming (1970) has recommended that for the quartz inclusion technique the grains should be etched in concentrated hydrofluoric acid (HF) for approximately 40 min. at room temperature in order to remove any discoloured surface resulting from the diffusion of impurities into the quartz during firing and also to dissolve most non-quartz grains. He suggested that this etching process would reduce to negligible importance the alpha dosage to the quartz grains since the alpha particles penetrate only the outer grain layers. Bell and Zimmerman (1978) have shown that this may not be strictly correct due to the wide variations in the effect of HF etching on different quartz grains and that some error may be introduced by assuming the residual alpha dose is negligible. However, it is still important to estimate by what factor the alpha dose is reduced by HF etching even if it is assuming only isotropic removal of material.

To evaluate the mean absorbed alpha dose in a spherical quartz grain after etching, $\overline{D_\alpha(Abs)}^*$, the numerical integration of $D_\alpha(x)$ described above, must be performed over a grain volume with a layer of thickness d removed isotropically from the surface. The resultant absorbed dose after etching as a fraction of the dose before etching is given in Figure 1 for both the uranium and thorium series as a function of the etching depth d. The results of Fleming (1970) are also shown in Figure 1 and the difference is seen to be significant probably because the earlier curves were evaluated for calcium fluoride and then assumed to be the same for quartz to a "first order approximation" (Fleming, 1969 and 1977). The results given here indicate that the alpha dose to a 100 µm quartz grain will be reduced by a factor of 0.5 for an isotropic etching depth of 6.4 µm for the uranium series and 7.9 µm for the thorium series whereas Fleming (1970) quotes 4.2 µm and 5.4 µm for the uranium and thorium series respectively.

To assess the length of time required for the etching of the quartz grains a relationship between etching depth and etching time is necessary. Such a relationship will vary between samples, however, because of the variations in etching rates likely to be found between the quartz from different parts of the world. The assessment of the etching depth for individual samples is also quite complex because the isotropic etching depth is calculated from an experimental determination of the weight loss with etching time and this weight loss could include small quantities of feldspars or other non-quartz grains which might be present in the sample and then dissolved by the HF.

It is useful, therefore, to consider the results of etching experiments done by Fleming (1969, 1970) on 100 µm grains of Norwegian alpha quartz. He gives figures of 4.2, 5.4, and 6.0 µm as being the isotropic etching depths corresponding to 20, 30 and 40 minutes of acid treatment respectively. These figures are for concentrated HF at room temperature. It is apparent that if the alpha dosage to 100 µm quartz grains is to be reduced by at least a factor of 0.5 then etching times of one hour must be considered as a minimum. The isotropic etching depth for Norwegian alpha quartz after one hour of acid treatment is 9 µm (Fleming, 1969).

The alpha contribution to the overall radiation dose will depend on several parameters (i) the relative amounts of the natural radioactive decay series present in the surrounding clay matrix (this is because $^{40}K$ emits only beta and gamma radiation whereas the uranium and thorium series also emit alpha radiation), (ii) the relative efficiency (or k value) of the alpha particles at inducing TL in the quartz grains, (iii) the attenuation of the alpha dose (and to a much lesser extent the beta dose as described in the next section) due to the size of the quartz grains, and (iv) the length of time the grains are immersed in HF acid. Table 1 shows how each of the above parameters affects the alpha contribution to the overall dose for 100 µm quartz grains embedded in three clays of different radioactivities. Alpha efficiency factors (k values) of 0.1 and 0.05 and the effect of etching the grains for one hour in concentrated HF at room temperature are each considered in Table 1. The data presented in Table 1 uses the dose-rates given in Bell (1979) and the dose attenuation factors for alpha radiation calculated in this section and for beta radiation calculated in the next section.

It should be stressed again that these results are assuming isotropic etching of all the grains which is certainly not the case as illustrated by Bell and Zimmerman (1978) who have shown that certain grains lose a substantial portion of their inner volume as etching tunnels are eaten through the grains. For clay matrices of a type similar to clay I in Table 1 it is obvious that large errors are possible because of this phenomenon particularly if the quartz has a high alpha efficiency factor (k value). For the majority of samples with a reasonable amount of potassium present ($\doteqdot$ 1% $K_2O$) and with a k value of the order of 0.05, then after etching the grains for about one hour in concentrated HF the error involved in neglecting the alpha particle contribution to the overall dose is likely to be much less than 5%.

## BETA DOSE ATTENUATION

Spencer (1959) has calculated the average energy dissipated near a point isotropic source of monoenergetic electrons by a moments method of solution of the electron transport equation. He presented numerical values of a "de-dimensionalised energy dissipation distribution" J(z) evaluated for electrons in various media (z is the distance from the source of electrons divided by their range $r_o$). This treatment has been extensively tested by Cross (1967, 1968) and it was found that over a wide range of spectral shapes there is excellent agreement between theory and experiment for distances from the source within which 95% of the energy is deposited. Charlton (1970) has incorporated the theory of Spencer (1959) into a convenient formalism which may be extended to the case of quartz inclusions embedded in a clay matrix in order to evaluate the beta dose attenuation factors for the natural radioactive series.

In similar fashion to equation (1), the beta dose absorbed at a point within a sphere distant x from the interface with a medium having a uniform distribution of beta sources is given by

$$D_\beta(x) = \frac{n_o E_o}{{}_m S} \; S_\beta(x/r_o, D/r_o) \qquad (4)$$

where $n_o$ is the number of electrons of initial energy $E_o$ and range $r_o$ originating per unit mass of the active medium, ${}_m S$ is the ratio of the mass stopping powers of the surrounding medium and the sphere, D is the diameter of the sphere, and $S_\beta(x/r_o, D/r_o)$ is the geometrical dose distribution function for beta particles given by Charlton (1970).

The values of $S_\beta(x/r_o, D/r_o)$ for quartz have been interpolated from the values for carbon and aluminium given by Charlton (1970) using the interpolation method recommended by Spencer (1959) and then $D_\beta(x)$ was integrated over the volume of a quartz grain of diameter 100 μm (Bell, 1978). The resultant mean absorbed dose for beta particles, $\overline{D_\beta(\mathrm{Abs})}$, from each series is then given by:

Uranium series,

$$\overline{D_\beta(\mathrm{Abs})_U} = 0.938 \times \frac{n_o E_o}{{}_m S} \qquad (5)$$

Thorium series,

$$\overline{D_\beta(\mathrm{Abs})_{Th}} = 0.910 \times \frac{n_o E_o}{{}_m S} \qquad (6)$$

Potassium-40,

$$\overline{D_\beta(\mathrm{Abs})_K} = 0.966 \times \frac{n_o E_o}{{}_m S} \qquad (7)$$

Rubidium-87,

$$\overline{D_\beta(\mathrm{Abs})_{Rb}} = 0.426 \times \frac{n_o E_o}{{}_m S} \qquad (8)$$

It is appropriate, however, to include internal conversion (IC) electrons with the beta attenuation factors and when this is done the mean absorbed beta doses for the uranium and thorium series become

$$\overline{D_\beta(\mathrm{Abs})_U} = 0.899 \times \frac{n_o E_o}{{}_m S} \qquad (9)$$

$$\overline{D_\beta(\mathrm{Abs})_{Th}} = 0.845 \times \frac{n_o E_o}{{}_m S} \qquad (10)$$

Therefore, a 100 μm quartz grain will absorb 89.9% of the beta dose that would be absorbed from the uranium series by an infinitesimally small grain, 84.5% of the dose from the thorium series, 96.6% of the dose from potassium-40 and 42.6% of the dose from rubidium-87. These values are in excellent agreement with the figures of 90.5%, 85.5%, and 96.5% given by Mejdahl (1979) for the uranium and thorium series and potassium-40 respectively.

The method adopted by Mejdahl (1979) for calculating the beta dose attenuation factors was based on the scaled absorbed-dose distribution functions given by Berger (1971, 1973) which are directly analogous to the dose distribution functions given by Charlton (1970) used in this paper. Both methods use the work of Spencer (1959) as a base on which to develop their separate formalisms and it is not surprising, therefore, that the results for the beta dose attenuation factors given here are in such excellent agreement with those presented by Mejdahl (1979).

For a typical clay matrix having 3 ppm uranium, 12 ppm thorium and 1% $K_2O$ (clay II in Table 3) the overall beta attenuation factor for a 100 μm quartz grain calculated here is 0.918. Mejdahl (1979) quotes 0.921 for the same grain size and concentration of radioisotopes, but he has also extended his work to include grain sizes from 5 μm up to 10 mm. For example, he gives beta dose attenuation factors of 0.875 and 0.836 for 200 μm and 300 μm diameter quartz grains respectively in the same clay as above. For these larger grain sizes the residual alpha dose will be very small before etching and completely negligible if the grains are etched in HF. The use of grains larger than 100 μm may prove to be more propitious, therefore, for certain dating projects.

## BETA DOSE REDUCTION BY HF ETCHING

The mean absorbed beta dose in a quartz grain after etching, $\overline{D_\beta(Abs)}^*$, is evaluated by numerical integration of $D_\beta(x)$ over the grain volume with a layer d removed from the surface. The absorbed dose after etching is given as a fraction of the dose before etching for each member of the uranium series in Figure 2 and of the thorium series in Figure 3. The dose attenuation of Rb-87 is also shown in Figure 3 whereas the potassium-40 beta dose suffers negligible dose reduction even at extreme etching depths.

Etching the grains for one hour in concentrated HF at room temperature, corresponding to an etching depth of 9 μm, reduces the beta dose from the thorium series by a factor of 0.915, the beta dose from the uranium series by a factor of 0.940 and the beta dose from rubidium-87 by a factor of 0.755.

## CONCLUSIONS

The attenuation factors for alpha and beta radiation which must be used in the dose assessment for quartz inclusions of average diameter 100 μm have been evaluated and the reduction of the radiation dose effected by etching the grains in HF has been illustrated. No mention has been made of the gamma radiation dose. This is because gamma rays from the natural radioactive series have a range of up to 30 cm or more in clay material and hence they will suffer negligible attenuation when passing through a 100 μm quartz grain. Care must be exercised, however, when dealing with a surrounding clay matrix which has an atomic number very different from that of the TL grains. At low energies the absorbed gamma energy varies approximately as the fourth power of the atomic number of the absorber and thus significant differential energy absorption between the TL grains and the surrounding matrix could occur. This phenomenon has been described by Chan and Burlin (1970), but for most pottery samples the atomic number of the clay matrix is very similar to that of quartz inclusions and negligible error is introduced by ignoring the effect.

## FIGURE CAPTIONS

Figure 1: The mean absorbed alpha dose in a 100 μm quartz grain with a layer d removed by etching, $\overline{D_\alpha(\mathrm{Abs})}^{*}$, as a fraction of the dose before etching, $\overline{D_\alpha(\mathrm{Abs})}$, for the Uranium and Thorium series. The results of Fleming (1970) are shown in dotted lines.

Figure 2: The mean absorbed beta dose in a 100 μm quartz grain with a layer d removed by etching, $\overline{D_\beta(\mathrm{Abs})}^{*}$, as a fraction of the dose before etching, $\overline{D_\beta(\mathrm{Abs})}$, for the Uranium series.

Figure 3: The mean absorbed beta dose in a 100 μm quartz grain with a layer d removed by etching, $\overline{D_\beta(\mathrm{Abs})}^{*}$, as a fraction of the dose before etching, $\overline{D_\beta(\mathrm{Abs})}$, for the Thorium series and Rubidium-87.

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TABLE 1

The contribution from alpha particles as a percentage (%) of the overall radiation dose to 100 μm quartz grains embedded in three clays of different radioactivities. The contribution is calculated for alpha efficiency factors of 0.1 and 0.05, both before and after etching the grains in HF acid for one hour (assuming isotropic removal of material).

| | Alpha Dose before Etching (%) | | Alpha Dose after Etching (%) | |
|---|---|---|---|---|
| | k = 0.1 | k = 0.05 | k = 0.1 | k = 0.05 |
| Clay I | 20.2 | 11.2 | 9.0 | 4.7 |
| Clay II | 14.2 | 7.6 | 6.0 | 3.1 |
| Clay III | 10.9 | 5.8 | 4.5 | 2.3 |

The radioactive composition of the three clays is as follows:

| | |
|---|---|
| Clay I | 12 ppm Th, 3 ppm U |
| Clay II | 12 ppm Th, 3 ppm U, 1% $K_2O$ |
| Clay III | 12 ppm Th, 3 ppm U, 2% $K_2O$ |

FIGURE 1

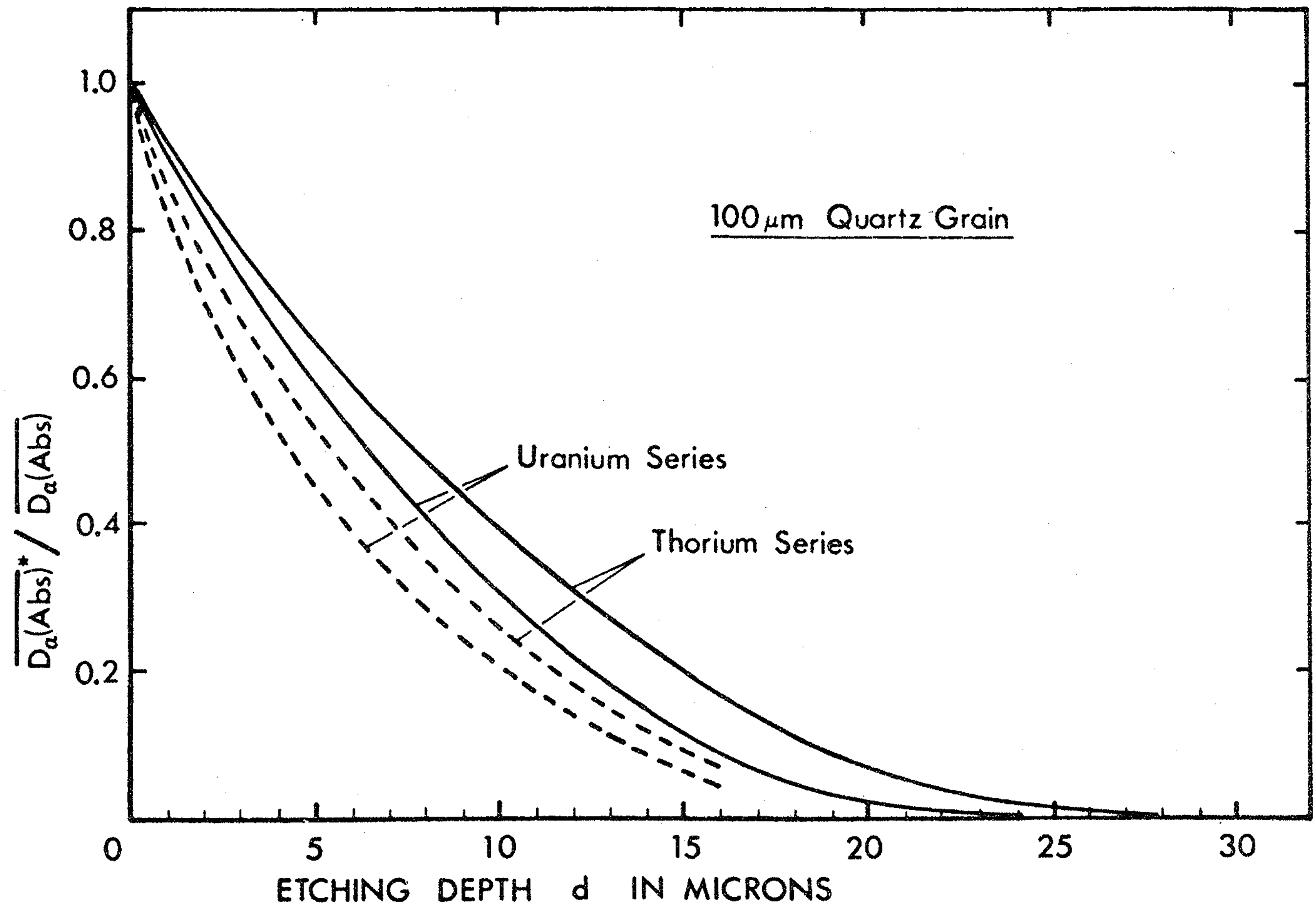



FIGURE 2

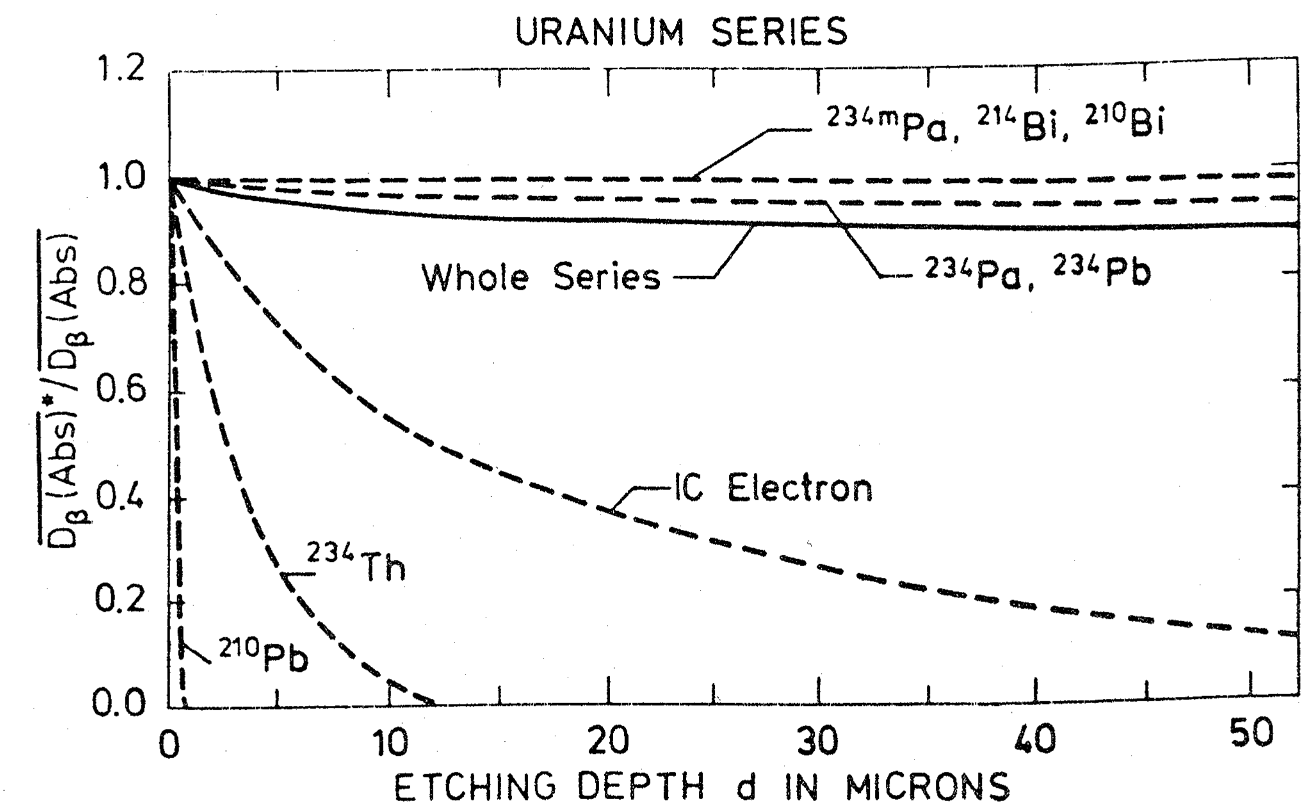
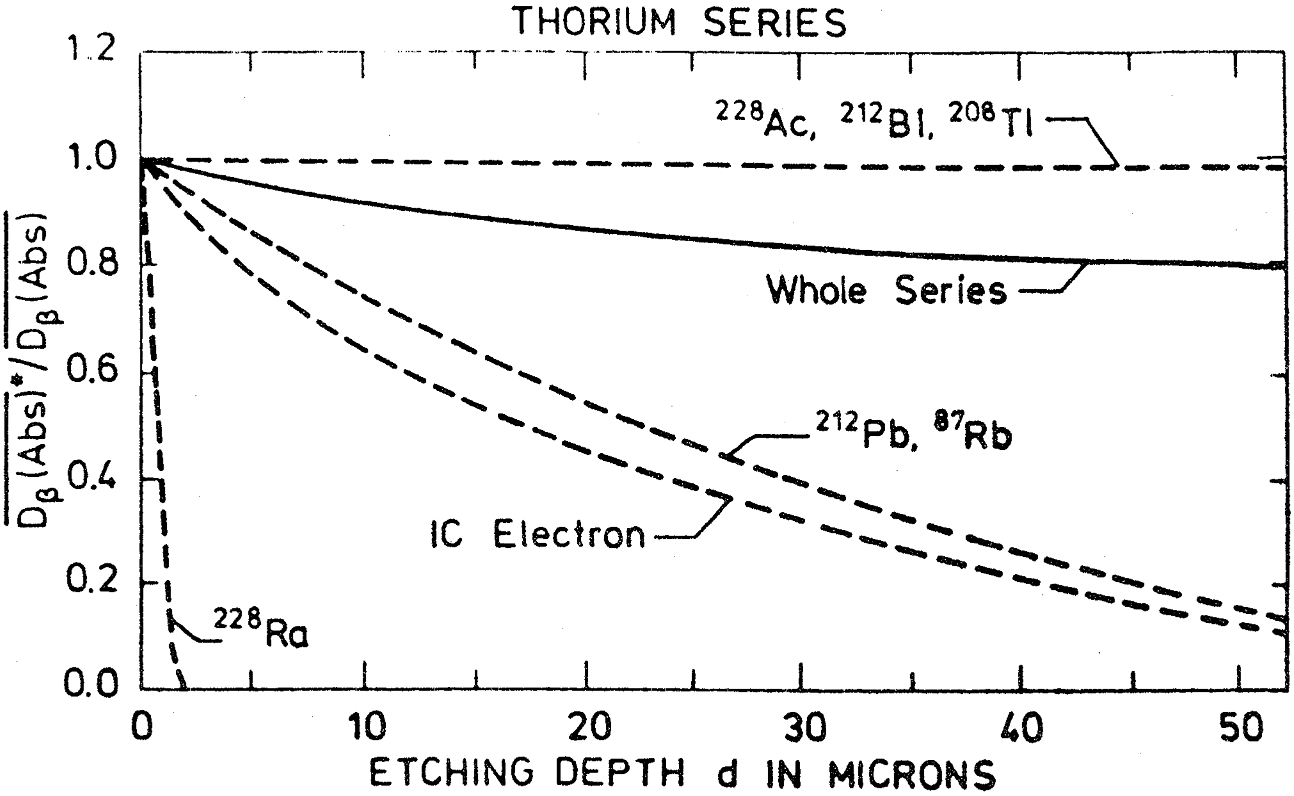


FIGURE 3




# A SWEDISH VITRIFIED FORT: DATING BY CONVENTIONAL TL

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## (1) Introduction.

This paper gives an account of measurements of conventional TL on samples discussed in a previous publication. (Wright 1979a). In that paper dating was carried out by the predose method on material from a Swedish vitrified fort. In the present work the samples and their preparation were as in the earlier paper. The standard procedure has been followed, including plateau test, correction for supralinearity, check of parallel slope criterion, and check for fading.

## (2) Results.

### (a) Samples treated in dilute HCl only. 4e, 8a, 8b, 12, 14.

This treatment removed the limestone but feldspar was retained with the quartz, (feldspar 17.6%, quartz 82.4%). The TL behaviour was normal, giving plots of which Fig. 1 is typical (sample 4e). The TL was large, and the tests were satisfactory; for sample 4 the fading in two months was about 16% consistent with feldspar being the main source of the TL (Fleming 1976). The archaeological dose for sample 8 (350 $\mu$) was however equivalent to a $\beta$-irradiation time of 220 min. compared with the predose result of 90 min.