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PROGRESS AND PROBLEMS WITH AUTOMATED TL DATING

Hugh McKerrell and Vagn Mejdahl

Abstract. A number of basic problems connected with the measurement of beta and gamma dose-rates are discussed, and the possibility of using low-temperature peaks in quartz and feldspar for dose-rate measurements is examined. Preliminary results of TL measurements on individual grains of quartz and feldspar are presented. A TL dating method based on the difference in the archaeological dose received by potassium feldspar and quartz grains is proposed.

INIS descriptors: AGE ESTIMATION, ARCHAEOLOGICAL SPECIMENS, BETA DOSIMETRY, DOSE RATES, FELDSPARS, GAMMA DOSIMETRY, QUARTZ, THERMOLUMINESCENCE, THERMOLUMINESCENT DOSIMETRY.

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FOREWORD, JANUARY 1981

The present paper was prepared originally for the Proceedings of the 16th International Symposium on Archaeometry and Archaeological Prospection, Edinburgh 1976, but these have not yet been published*. An effort was made about a year ago to publish revised versions of the papers submitted, but we felt that our paper would require too much revision for inclusion in this publication. We have found, however, that reference is frequently being made to some of the ideas put forward in the paper and have decided, therefore, to make it available as a Risø Memo. The paper is published here with minor changes as explained by notes inserted in the text.

* Publ. Feb 1981

INTRODUCTION

The first successful approaches to dating of ceramics by thermoluminescence (TL) were made nearly ten years ago with the introduction of the inclusion technique (Fleming 1966) and the fine-grain technique (Zimmerman 1967). Extensive test programmes carried out in connection with further developments of these methods have demonstrated that freshly excavated material can be dated with an accuracy of 5-10% (Fleming 1970; Zimmerman 1971; Mejdahl 1972a, b). The different techniques and the problems encountered with them have been discussed in detail by Aitken and Fleming (1972). A briefer survey has been given by Seeley (1975). In recent years, the Oxford laboratory has applied TL dating to archaeological material of less well known or unknown age (Whittle 1975; Whittle and Arnaud 1975; Huxtable et al. 1976). With the exception of one date (7.3%), the stated uncertainty (one standard deviation) of the twenty three dates given in the three publications is within 5-6%.

A fair assessment of the present situation seems then to be that TL dating of well behaved material can be carried out with an accuracy better than 7%. While still not quite satisfactory this accuracy will allow TL dating to be usefully applied to a wide range of chronological problems which cannot be approached by other methods. It seems desirable, therefore, to take steps towards the establishment of a dating service on a routine basis similar to radiocarbon dating. Clearly, at the same time extensive research work must be aimed at improving the accuracy of the method.

Because of the formidable number of measurements required in TL dating application of the method on a larger scale must be based on automised and computerised equipment. In this paper we describe preliminary work in this direction comprising:

1) Construction and testing of measuring equipment based on heating of samples with hot nitrogen. Description of a registration system suitable for computer processing of data.

2) Discussion of calibration problems encountered in the dosimetry of beta and gamma radiation. Examination of the possibility of using individual grains of quartz and feldspar for dose-rate measurements.

3) Application of individual quartz and feldspar grains for determination of archaeological dose. Discussion of dating systems in which the whole dosimetry is based solely on quartz or feldspar grains. Such systems would avoid a number of fundamental calibration problems.

4) Discussion of a subtraction technique based on differences in archaeological dose between large grains of potassium feldspar and large grains of quartz. In this technique the age determination can be based solely on the dose contributed by inherent potassium in the feldspar grains. All problems associated with the measurement of external beta and gamma dose-rates will therefore be eliminated. There can be no doubt that this would lead to a significant improvement in accuracy as well as a greater flexibility since the method would no longer be restricted to fresh excavations.

Note 1, 1981

Further work on heating systems has shown that heating by means of an electrically heated metal strip is superior to hot-nitrogen heating for quartz and feldspar samples, one reason being that the heating rate can be more easily controlled. The section on the hot-nitrogen reader has, therefore, been omitted. The metal-strip heating has been incorporated in an automated read-out system (Bøtter-Jensen and Mejdahl, 1980).

MEASUREMENT OF BETA DOSE-RATE BY MEANS OF TL PHOSPHORS

Application of sensitive TL phosphors for determination of the radioactivity of ceramics and other material used for TL dating was first proposed by Aitken (1968). Further studies by Fleming (1968), Aitken and Fleming (1972), Bailiff et al. (1974), and Mejdahl (1969, 1972a) have demonstrated the advantages of this technique, in particular for determination of beta dose-rate in inclusion dating.

A version of the technique suitable for practical applications has been designed at Edinburgh. The dosimeter unit is a small polypropylene wafer, diameter 12 mm, thickness 0.8 mm, with a depression which can accommodate a disc of $CaSO_4$:Dy-teflon, 5 mm in diameter and 0.4 mm thick (fig. 1A). A lid sealed on to the wafer prevents radon and thoron from diffusing to the surface of the phosphor. The beta 'window' on both sides of the teflon disc is 40 mg/cm^2. This thickness of shielding effectively stops the alpha particles and transmits about 55% of the beta radiation.

For irradiation of the dosimeters, the ground and dried pottery is placed in a polypropylene capsule (diameter 1.5 cm, height 5 cm) together with the dosimeters as shown in fig. 1B. Three dosimeters can be accommodated in one capsule in a geometry approaching 4π. The amount of sample required to fill the capsule is about 8 g. However, using only one dosimeter it is possible to obtain a measurement from as little as 4 g. The capsule, sealed with a lid in order to prevent escape of radon and thoron, is stored for a minimum period of four weeks. During this period doses of 10 mrad or more will be accumulated; these can be measured with good accuracy with the $CaSO_4$:Dy dosimeters.

Calibration of the reading of the dosimeters after storage is done by comparison with dosimeters stored in capsules containing standard material prepared with known concentrations of uranium, thorium and potassium. The calibration is thus based on calculations of beta-ray intensities from known amounts of radioisotopes; and the resulting dose-rate value will refer to a

point in an infinite medium of the material. The calculations will be based on the conversion values given by Bell (1976).

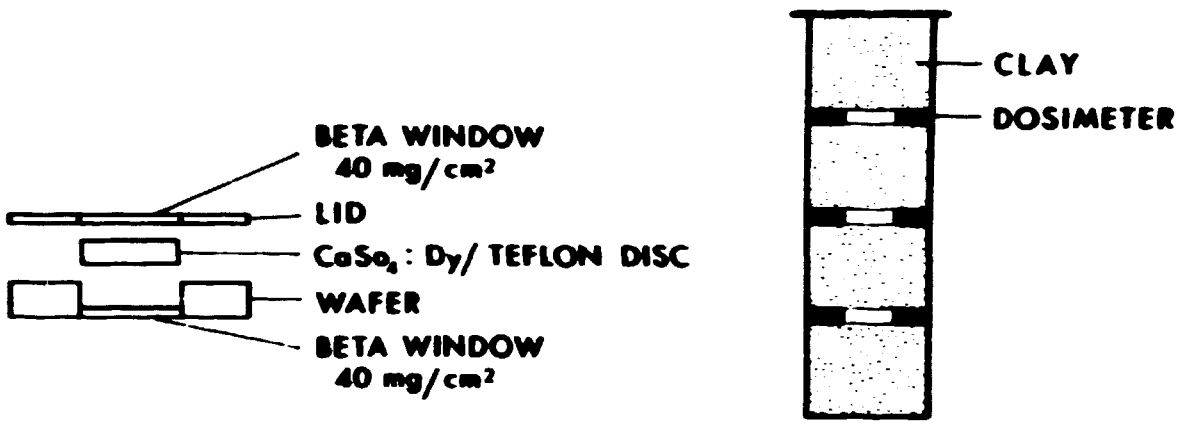

Fig. 1. Dosimeter and storage capsule for measurement of beta dose rate.

A. Dosimeter: $CaSO_4$: Dy embedded in teflon discs, 5 mm diameter x 0.4 mm thick. To be inserted in recess in polypropylene wafer, 12 mm diameter x 0.8 mm thick, and sealed with a lid.

B. Polypropylene storage capsule containing ground-up pottery and wafers with $CaSO_4$: Dy/teflon discs. Dimensions of capsule: outer diameter 15 cm, height 5 cm, wall thickness 0.1 cm.

The method outlined above is very convenient, and for homogenous material the accuracy can be extremely good - mean error for three dosimeters about 1.5%. There are, however, a number of perturbing factors which will affect the overall accuracy of the dose-rate value; some of these will be discussed briefly.

Note 2, 1981

The method has been used for determination of the beta dose-rate in TL dating of ceramics from Glozel (Francois et al. 1977; Mejdahl 1980). Further development of the method has led to a more convenient technique in which the material is sealed in a polyethylene bag and the dosimeters attached to the surface of the bag (Mejdahl 1978a).

(i) Variation in relative abundance of radioisotopes. The relative abundance of the three radiation sources potassium and the uranium and thorium series can vary markedly from sample to sample. However, provided that the attenuation of the radiation by the shield covering the phosphor is the same for the three contributors the response of the dosimeter should be independent of the relative abundance of the radioisotopes. An additional advantage is that no assumption of secular equilibrium in the decay series is involved (apart from the radon problem which will be discussed later). A preliminary calculation of the attenuation by a 30 mg/cm^2 shield based on the Loevinger formalism (Fitzgerald et al. 1967) indicated that the attenuation was in fact the same (40%) within 2% for the three sources. Extrapolation to 40 mg/cm^2 gave an attenuation value of 45%, i.e. a transmission of 55% of the beta radiation. However, it is not necessary to know the transmission with any accuracy because it will be the same for the unknown sample and the standard calibration sample if equal relative attenuation of the radiation from the three sources can be assumed. Since this is an important point it must be investigated in detail by exposing the dosimeters to radiation from material having a wide range of known relative abundances of potassium, uranium and thorium.

(ii) Attenuation of beta radiation by the quartz and feldspar inclusions. As mentioned above the dose-rate value obtained from the measurement refers to a point in an infinite matrix of homogeneous material. However, the value of interest in inclusion dating is the average dose to quartz and feldspar grains of a finite size ranging from 0.05 to 2 mm. Therefore, corrections must be made for attenuation of the beta radiation by the grains. Fig. 2 shows an attenuation curve giving relative attenuation of beta dose as a function of grain diameter for the following concentrations of beta emitters:

Thorium	12 ppm
Uranium	3 ppm
K_2O	1 %

Since the individual attenuation curves for the three beta contributors differ somewhat, the resulting attenuation curve will depend on the relative abundance. The variation in abundance normally encountered will not cause any significant deviation from the curve in fig. 2; but in extreme cases it will be necessary to recalculate the attenuation from the individual attenuation values.

Note 3, 1981

Figure 2 has been recalculated on the basis of the attenuation values listed by Mejdahl (1979).

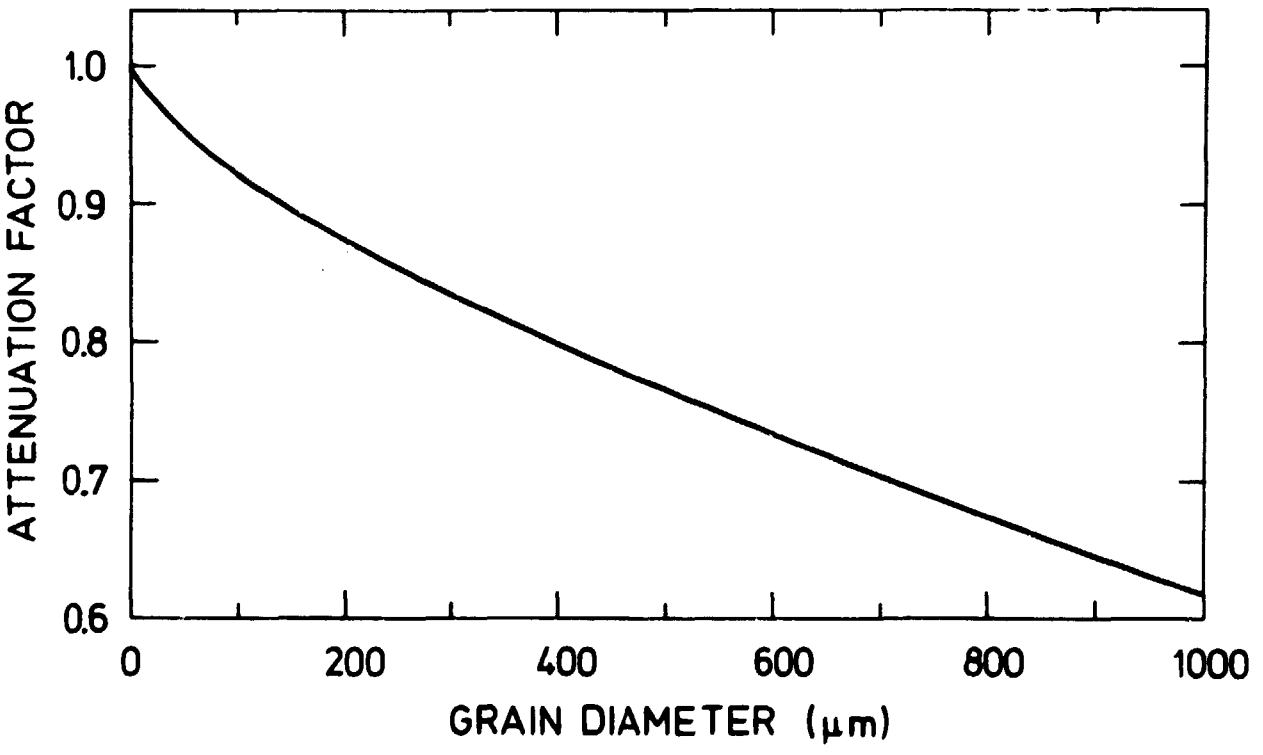

Fig. 2. Attenuation of beta radiation by quartz or feldspar grains. The graph is based on attenuation values listed by Mejdahl (1979). Assumed concentrations of radioisotopes: 12 ppm Th, 3 ppm U, and 1% K_2O.

(iii) The radon problem. This problem arises from the fact that radon being a gas with a relatively long half-life, 3.8 days, tends to diffuse out of pottery samples. Since radon occurs about half way down the uranium series, total loss of radon will

cause a decrease of the uranium beta activity by about 60% according to the calculations by Bell (1976).

The first consideration concerns the build up of radon to equilibrium in a sample which has been sealed off in the capsule for measurement of beta dose-rate. Assuming that all radon was lost from the sample at the moment of sealing, the activity will grow to about 80% of its equilibrium value in a period of two weeks. Calculations based on the concentrations of radioisotopes listed above show that the difference in dose accumulated during four weeks in the two extreme cases: build up from total disequilibrium (i.e. no radon present initially) and full equilibrium during the whole period is about 3%. In practice, total initial loss of radon would be unlikely; and in any case the problem could be eliminated by extending the storage period. It is possible to study the build up of radon - and thereby the relative amount of radon present initially - by replacing the $CaSO_4$:Dy phosphor with $CaSO_4$:Mn (also available in teflon). This phosphor can measure the radiation accumulated during a few days, but because of the rapid fading of the peak at room temperature, the capsule would have to be stored in a freezer during the irradiation period.

Assuming that the measurement period has been of a sufficient length for build up effects to be negligible, the beta dose-rate value obtained will refer to complete retention of radon. However, since the pottery could have been losing radon (and possibly thoron) during burial in the ground, the true dose rate during burial might have been lower than that derived from the laboratory measurement. The magnitude of this effect can be illustrated by the following calculations of the difference in annual dose in the two extreme cases: no loss of radon and total loss of radon during burial. The concentrations assumed for thorium and uranium are those given above (Th 12 ppm and U 3 ppm). Three K_2O values, 1%, 2% and 4%, and two values of the annual gamma dose, 100 mrad/year and 200 mrad/year are considered. Results are given in Table 1.

Table 1. Effect of radon escape from pottery during burial. Assumed concentrations of thorium and uranium: 12 ppm Th and 3 ppm U. Ratio of beta + gamma assuming total loss of radon to beta + gamma assuming no loss of radon. Recalculation 1981 using conversion values given by Bell (1979).

K_2O	γ 100 mrad/yr	200 mrad/yr
1%	0.895	0.925
2%	0.919	0.938
4%	0.944	0.954

Clearly, if all radon escaped from the pottery during burial, the resulting error would be quite substantial. In practice, however, a loss of 10% would be more likely; and in this case the error would be negligible. The relative amount of radon present in freshly excavated sherds could be measured by means of $CaSO_4$:Mn dosimeters attached to the surface of the sherds. Sherds and dosimeters would have to be cooled down quickly in a freezer. The radon would thereby be sealed in, and the build up would be evaluated. Studies of this sort on a variety of sherds would provide an extremely useful basis for the estimation of the error that could result from radon escape.

(iv) Water content of pottery. At northern latitudes sherds buried in soil will normally be almost saturated with water all the year round. Since the water causes attenuation of the beta radiation the true dose rate will be lower than the value derived from laboratory measurements on dry pottery clay. This error is common for all methods of beta activity measurement and can be corrected by measuring the water uptake of the sherds allowing for the higher stopping power of water (1.25 times that of clay). When the beta dose-rate is measured by means of TL phosphors the correction can be incorporated in the measurement by adding the calculated amount of water to the dry clay in the capsules. It would also be feasible to attach dosimeters to the

surface of sherds saturated with water and freeze the assembly. Thereby water (and radon) would be contained. By these techniques the effect of water attenuation could be studied.

By the dosimetry technique outlined above the present-day beta activity of a sherd can be determined with good accuracy. However, this value could differ from the activity during burial if substantial leaching or accumulation of radioactivity had taken place in the past. For a sherd buried in a clay-rich soil the radioactivity content is not likely to vary because of the restricted water circulation in clay; but significant leaching could occur from sherds buried in sandy soil with a humus-rich top-soil (podsol).

The principal drawback of the technique is the necessity to base the calibration on dose-rate values calculated from given activity concentrations in the standard samples. These calculations will depend on values of half-lives and beta-ray energies which are liable to change as a result of improved methods of measurement. The deviation from earlier values of the dose-rate conversion values obtained by Bell (1976) demonstrates that such changes can be quite dramatic. The problem is of importance because once TL dating is established as a routine method general changes in reference values leading perhaps to a correction of published dates would tend to discredit the method.

We have investigated the possibility of avoiding this problem by using the quartz and feldspar inclusions themselves as built-in dosimeters for measurement of the beta dose-rate. The two phosphors have low-temperature peaks with a sensitivity sufficient to measure doses in the mrad region. In quartz the 110°C peak can be activated to a high sensitivity by irradiation of the sample with a large dose (some kilorads) and subsequent heating to 500°C. Feldspar (fig. 4) has a sensitive peak at about 140°C which requires no activation.

Since the peaks are not stable at room temperature, the pottery samples must be stored at a low temperature during the irradiation period; and the subsequent preparation for measurement

(cleaning, etching etc) must be timed accurately. Results of calculations of the stability of the 110°C quartz peak at various temperatures are summarized in Table 2. The calculations were based on first order kinetics; and the 'stability period' at room temperature was taken to be one minute. The fading in this period is less than 2%. The fading over stability periods calculated for other temperatures will then also be less than 2%. A trap depth of 0.99 eV was assumed (Aitken and Fleming 1972).

Table 2. Stability of the 110°C peak in quartz at different temperatures. Assumptions: First order kinetics; trap depth: 0.99 eV; 'stability period' at room temperature: one minute.

Temperature (°C)	Stability period (fading less than 2%)
0	20 min
-25	22 hours
-50	174 days
-70	80 years

Since storage periods of months would be required, it is evident that only temperatures below -50°C would ensure sufficient stability. Storage at the temperature of solid CO_2 (-78°C) would be adequate; but in practice it would probably be simpler to use liquid nitrogen. The life-time at 0°C (20 min) is sufficient for cleaning and etching of grain samples at that temperature after storage.

We have carried out preliminary investigations of the response of the low-temperature peaks in quartz and feldspar using large individual grains (weight 1-3 mg). The grains were heated on a nichrome heating strip, and the TL signal was measured by photon counting. The signal was integrated in temperature intervals of 50° or 100° around the peaks. Using a commercial light guide we registered as much as 27 000 counts over a temperature interval

of 50° for a 3 mg feldspar grain exposed to a dose of one rad. This level is very typical for feldspar; and activated quartz grains are about four times as sensitive. The response is strictly linear over the dose range of interest. Results of the sensitivity measurements are summarized in Table 3.

Table 3. Sensitivity of the low-temperature peaks in quartz and feldspar. No blue filter. Commercial light guide.

	Peak temperature (°C)	Temperature interval (°C)	Counts/ rad/mg	Min. dose (10 mg sample)	Storage period for 3 mrad/week
Quartz	110	85-135	36 000	12 mrad	4 weeks
Feldspar	140	120-170	9 000	50 mrad	17 weeks
	140	110-210	13 500	30 mrad	11 weeks

The minimum dose for a 10 mg sample corresponds to 4500 counts, i.e. a standard deviation of 1.5%. The assumed dose rate, 3 mrad/week, is the average beta dose to small grains. For larger grains, around 1 mm diameter, the dose to quartz would decrease by about a factor of two because of attenuation; the dose to feldspar would increase by more than a factor of two because of the increased contribution from inherent potassium 40 in the grains.

These preliminary results indicate that determination of the beta dose-rate by means of quartz and feldspar grains would be perfectly feasible. In practice each sample to be dated could be divided in two, one half for the dose-rate measurement and the other for the measurement of the archaeological dose. The two measurements could then be carried out on identical grains, same grain size, etched to the same extent etc. For feldspar grains there is the alternative possibility of measuring the dose-rate and the archaeological dose on exactly the same grains, the dose-rate being determined before the measurement of the archaeo-

logical dose. The measurement of the dose stored in the low-temperature trap does not affect the archaeological dose stored in the high-temperature trap. This possibility is of particular importance for feldspar dating of inhomogeneous samples such as burned stones.

The problems associated with water uptake and radon escape from pottery are not avoided by the proposed technique. If the sherds are saturated with water during the irradiation period the radon will be sealed in; and considerations of the errors involved will be similar to those pertaining to measurements with $CaSO_4$:Dy dosimeters.

The method of measuring beta dose-rates by means of quartz and feldspar grains seems well worth developing in spite of the technical difficulties involved with sortage of samples at a low temperature. The method has a number of advantages which can be summarized as follows:

1) Exact absolute calibration of dose-rate values is not required provided that the same source, or two accurately intercalibrated sources, are used for calibration of beta and gamma dose-rate measurements and the measurement of archaeological dose. The resulting date will then be derived from a consistent dosimetry system, independent of any tabulated figures and of absolute calibration of radiation sources.

2) Since the beta dose-rate measurement and the measurement of archaeological dose are carried out on identical samples no correction is needed for attenuation of the beta radiation by the grains. In fact, all grain sizes present in the material can be utilized.

3) Whole sherds saturated with water can be used for the dose-rate measurement. The attenuation of the radiation by the water can then be included directly in the measurement so that no knowledge of the stopping power of water relative to that of clay is required.

4) In addition to the beta dose-rate the measured value will include a small fraction of the gamma dose-rate, the so-called gamma self-dose originating from the radioactivity in the pottery. Since this part is particularly difficult to evaluate with the dosimeters for gamma radiation described in the next section, the calibration of the gamma dose-rate values will be facilitated considerably.

MEASUREMENT OF ENVIRONMENTAL RADIATION

Our procedure for measurement of environmental gamma radiation is essentially as described by Mejdahl (1972a), except that $CaSO_4$:Dy embedded in teflon is used rather than loose powder.

Discs of $CaSO_4$:Dy-teflon, 5 mm diameter x 0.5 mm thick, are contained in polyethylene capsules with an outer diameter of 9 mm and a wall thickness of 1.5 mm. The capsules are placed inside a steel tube at intervals varying from 4 to 20 cm according to the homogeneity of the soil at the site where the probe is to be placed. The inner diameter of the steel tube is 10 mm and the wall thickness is 1.5 mm. The tubes vary in length from 0.75 to 2 m, and if necessary, several tubes can be joined together. Normally, three to five tubes are placed at each site in order to obtain a good estimate of the spatial variation of the gamma radiation at the site.

If the soil at the site is very inhomogeneous, sampling of sherds for dating is restricted to the immediate environment of a probe, and care is taken to ensure that the gamma dose-rate for a particular sherd is evaluated from the capsules closest to the sherd.

Fading is assumed to be negligible for the dosimeters, at least for periods up to one year. Poul Christensen, Risø, (personal communication) has found that the fading of $CaSO_4$:Dy at room temperature over a period of two years was less than 5%.

Calibration of the exposures will be carried out with radium, Co-60 or Cs-137 gamma sources in a scatter-free geometry. For these sources the energy is so high that the energy dependence of the response of the $CaSO_4$:Dy dosimeters will be negligible.

For exposure in soil, shielding of the dosimeters by metal (for instance by insertion in a steel tube) is preferable in order to reduce the energy dependence of the response. An investigation carried out some years ago (Mejdahl 1970) gave the following results: Without shielding, the response per rad reached a maximum at 40 keV which was nine times the response to Co-60 gamma radiation. Shielding by 1.5 mm steel shifted the maximum to 100 keV and lowered it to 2.5 times the response to Co-60 gamma radiation. Below the maximum the response decreased rapidly because of the cut-off of the softer radiation; at 40 keV it was one, and at 20 keV practically zero. An attempt to calculate the response of $CaSO_4$:Dy inside a steel tube to a typical soil spectrum gave the result that the dosimeter overestimated the dose by 6.6%. However, it was not possible to obtain a completely accurate estimate in this way because the spectrum of the radiation below 100 keV was not known.

Note 4, 1981

A short paragraph on comparison of $CaSO_4$:Dy with LiF has been omitted. As the quantity required in inclusion dating is the dose to quartz or feldspar grains, it is preferable to use a dosimeter that has a quartz-equivalent response. Later experiments have shown that the response of $CaSO_4$:Dy contained in steel tubes is practically quartz equivalent (Mejdahl 1978b).

Properly calibrated, a site measurement will give the gamma dose-rate at a point in the soil. What is required, however, is the dose rate experienced by a quartz or feldspar grain embedded in a sherd buried in the soil. The gamma dose-rate inside a sherd differs from the value in the soil for two reasons:

1) The gamma radiation from the soil will be attenuated by tne sherd.

2) The sherd will absorb a certain fraction, the gamma self-dose, of the radiation emitted by its own radioisotopes.

If the sherd and the soil have the same concentrations of radioisotopes (and the same water content) the two effects will cancel completely, and the soil value will be equal to the sherd value. This means that the absorbed fraction of gamma radiation in the sherd must equal exactly the relative attenuation of the radiation from the soil. If on the other hand the concentrations of radioactivity in the sherd and in the soil differ markedly, a correction may be necessary.

An attempt has been made to evaluate this correction from tabulated values of absorbed fractions obtained by Monte Carlo calculations (Brownell, et al. 1968; Ellett and Humes 1971). Their calculations were made for a unit-density soft-tissue medium but by a straightforward conversion the values can be used for other densities and other low-Z materials. For the calculations the entry m = 300 g in Brownell et al (1968), Table 11, has been used with backscatter values from Table 12 included. Table 11 gives absorbed fractions for ellipsoids with principal axes in the ratio 1/0•5/2. The entry will correspond to a sherd weighing 61 g and having approximate dimensions 3 x 1•3 x 6 cm. The calculations were applied to the soil spectrum given by Mejdahl (1970, fig. 8) for which an average absorbed fraction of 0•083 was obtained. The relative attenuation of the soil radiation will then also be 0•083; that is the correction factor for attenuation will be 0•917. It must be noted that the calculations could not be extended to energy values lower than 100 keV. Assuming that the sherd contains 12 ppm of thorium, 3 ppm of uranium, and 1% of K_2O, the infinite-medium gamma dose-rate (Bell, 1979) will be 116•8 mrad/yr; with an absorbed fraction of 0•083 the self-dose to the sherd will be 9•7 mrad/yr.

Assuming that the gamma radiation from the soil is attenuated by the sherd by the factor 0•917, and that the contribution c from cosmic radiation is not attenuated, we find that the desired gamma dose-rate (d_{sherd}) inside the sherd, including cosmic radiation, is related to the measured gamma dose-rate (d_{soil}) in the

soil, including cosmic radiation, by the following expression:

$$d_{sherd} = (d_{soil} - c) \times 0{\cdot}917 + c + 9{\cdot}7 \text{ mrad/yr}$$

Table 4 shows the ratio of d_{soil} to d_{sherd} for various soil types and for the sherd referred to in the calculations.

Table 4. Ratio of gamma dose-rate (d_{soil}) in soil to gamma dose-rate (d_{sherd}) inside a sherd buried in the soil. Cosmic radiation included in both values. Assumptions for the sherd: weight 61 g; infinite-medium gamma dose-rate 116·8 mrad/yr; absorbed fraction of gamma dose 0·083. Assumed contribution from cosmic radiation 15 mrad/yr.

Soil type	d_{soil} (mrad/yr)	d_{sherd} (mrad/yr)	d_{soil}/d_{sherd}
Bog, peat	25	33.9	0.74
Diluvial sand	65	70.6	0.92
Moraine sand	95	98.1	0.97
Moraine clay	135	134.7	1.00
Granite	165	162.3	1.02
Granite	215	208.1	1.03

The values in Table 4 indicate that except for clay-rich soils and granites the correction is not negligible; and for sherds buried in peat it becomes substantial. Since the gamma-ray contribution from peat is very low, the correction of the total dose-rate value would be considerably less than the indicated 24% but it could well be 10 mrad in 150 mrad, i.e. 7%.

The absorbed fraction will increase with increasing sherd size. Table 5 lists approximate values of the absorbed fraction as a function of weight of sherds or burned stones of ellipsoidal or spherical form. An average gamma-ray energy of 1 MeV - indicated by the average absorbed fraction 0·083 derived above - was assumed for these calculations. The backscattered radiation has been included.

Table 5. Absorbed fraction of gamma radiation as a function of weight of sherds or stones of ellipsoidal or spherical form. Back-scattered radiation included. Assumed average gamma-ray energy 1 MeV. Brownell et al. (1968), tables 10, 11 and 12. Assumed density of stone 2·7 g/cm^3.

Weight of sherd or stone (g)	Absorbed fraction		Diameter of sphere (cm)
	Flat ellipsoid	Sphere	
61	0·083	0·097	3·5
122	0·110	0·122	4·4
408	0·151	0·169	6·6
1020	0·195	0·225	9·0

Note 5, 1981

A few paragraphs describing calibration problems have been omitted because these problems do not arise when a quartz-equivalent dosimeter is used for the measurement of environmental radiation.

MEASUREMENT OF ARCHAEOLOGICAL DOSE

Note 6, 1981

Two introductory paragraphs have been omitted.

In order to investigate the feasibility of using large individual grains for determination of the archaeological dose we have begun a study of quartz and feldspar grains in the 1-2 mm diameter range. Selection of grains can be done visually and the measurement is quite straightforward. Since each sample consists of a pure mineral this technique provides an excellent opportunity of studying the TL properties of the quartz and feldspar grains occurring in pottery. As an example of the kind of infor-

mation that can be obtained, some very preliminary results for quartz grains from a Danish Iron Age sherd (Fredbjerg, lab. no. 731307) are given below:

1) Most grains had a well defined peak at 360°C (fig. 3). A marked variation in glow-curve shape occurred from grain to grain, in particular regarding the 110° peak and less pronounced peaks in the region 120 - 250°. There was a considerable variation in sensitivity to radiation. The distribution of the response (integrated counts per mg in a 50°C interval centered at the 360° peak) of 30 grains exposed to a dose of 600 rad is given in Table 6.

Table 6. Distribution of response per mg of individual quartz grains exposed to a dose of 600 rad. Counts in a 50° interval centered at the 360° peak.

Counts	No. of grains	%
0 - 500	2	6•7
500 - 1000	9	30•0
1000 - 1500	11	36•7
1500 - 2000	4	13•3
2000 - 2500	2	6•7
2500 - 3000	1	3•3
4500 - 5000	1	3•3
Total	30	100

2) For about 20% of the grains a shift in peak position from the first to the second heating occurred. Such grains would be unsuitable for dating and have not been included in the statistics above. The shift is apparently caused by a growth in the sensitivity of the 325° peak. Fleming (1970) has discussed this phenomenon in more detail and has shown that the 325° peak is

not well suited for dating. The shift is not related to any obvious features in the natural glow curve and the grains for which it occurs cannot be distinguished microscopically from the 'good' grains. With the "single grain" technique, grains showing peak shift can easily be detected and discarded. Detection of such grains in a composite sample is more difficult but their presence should be revealed by the so-called plateau test i.e. a comparison of the ordinates in the natural and the second glow curve over the relevant temperature region 300 - 400°C (Aitken and Fleming 1972).

3) Direct investigation of the sensitivity change by adding a large beta dose (3000 rad) to the archaeological dose (500 rad) and comparing the response with the response to a subsequently applied laboratory dose of 3500 rad. For 14 grains the ratio of the second to the first response ranged from 0·46 to 0·68 with a mean value of 0·57, a standard deviation of 10·3% and a corresponding mean error of 2·7%. In view of the large variation in absolute response of the grains the variation in sensitivity change is remarkably small. The results indicate that it might in fact be feasible to determine the archaeological dose for a single quartz grain by comparison with a second build up curve corrected for sensitivity change using an average correction factor. The building up of a calibration curve for a grain would require several laboratory doses and therefore more than one heating of the grain; however, subsequent brief heatings to 500°C generally cause no further sensitivity change.

4) The correction procedure outlined above has been applied to 18 grains from the same sherd. The natural TL signal was compared with the signal induced by a laboratory dose of 600 rad beta radiation using the average correction factor 0·57 derived above. Since the response of the grains from this sherd was approximately linear up to 1000 rad only one calibration dose was required. The variation of doses obtained in this way was from 318 rad to 770 rad with a mean value of 546 rad, a standard deviation of 23·4% and a mean error of 5·5%. The dose values obtained need a further correction because of the attenuation by the grains of the radiation from the Sr 90 calibration source,

but this will not affect the variation. Clearly, the variation cannot be explained by the uncertainty in the sensitivity change; there does seem to be a real dose variation from grain to grain in the sherd. In view of the very inhomogeneous fabric of a sherd such a variation might not be unlikely. The results indicate that in order to reduce the mean error to 3% it would be necessary to use an average result for 60 grains, equivalent to about 100 mg.

The results outlined above indicate that determination of archaeological dose and thereby dating of a sherd could be based on individual grains of quartz provided that a sufficient number of grains was used and that determination of dose rate was done with grains of the same size. The main advantage would be that unsuitable grains could be discarded.

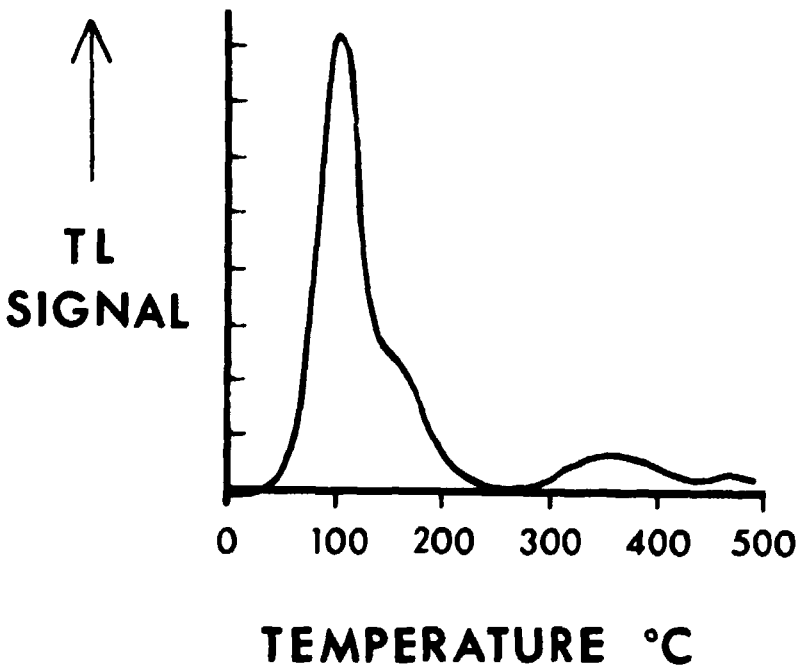

Fig. 3. Glow curve from a single quartz grain (about 1 mm in diameter) extracted from a sherd (Fredbjerg, Denmark. Iron Age. Lab. No. 731307). Dose: archaeological dose plus 2000 rad Sr-90 beta radiation.

A few preliminary results have been obtained for individual feldspar grains. Typical glow curves are shown in fig. 4. General features of the curves are: a low-temperature peak around 140°C,

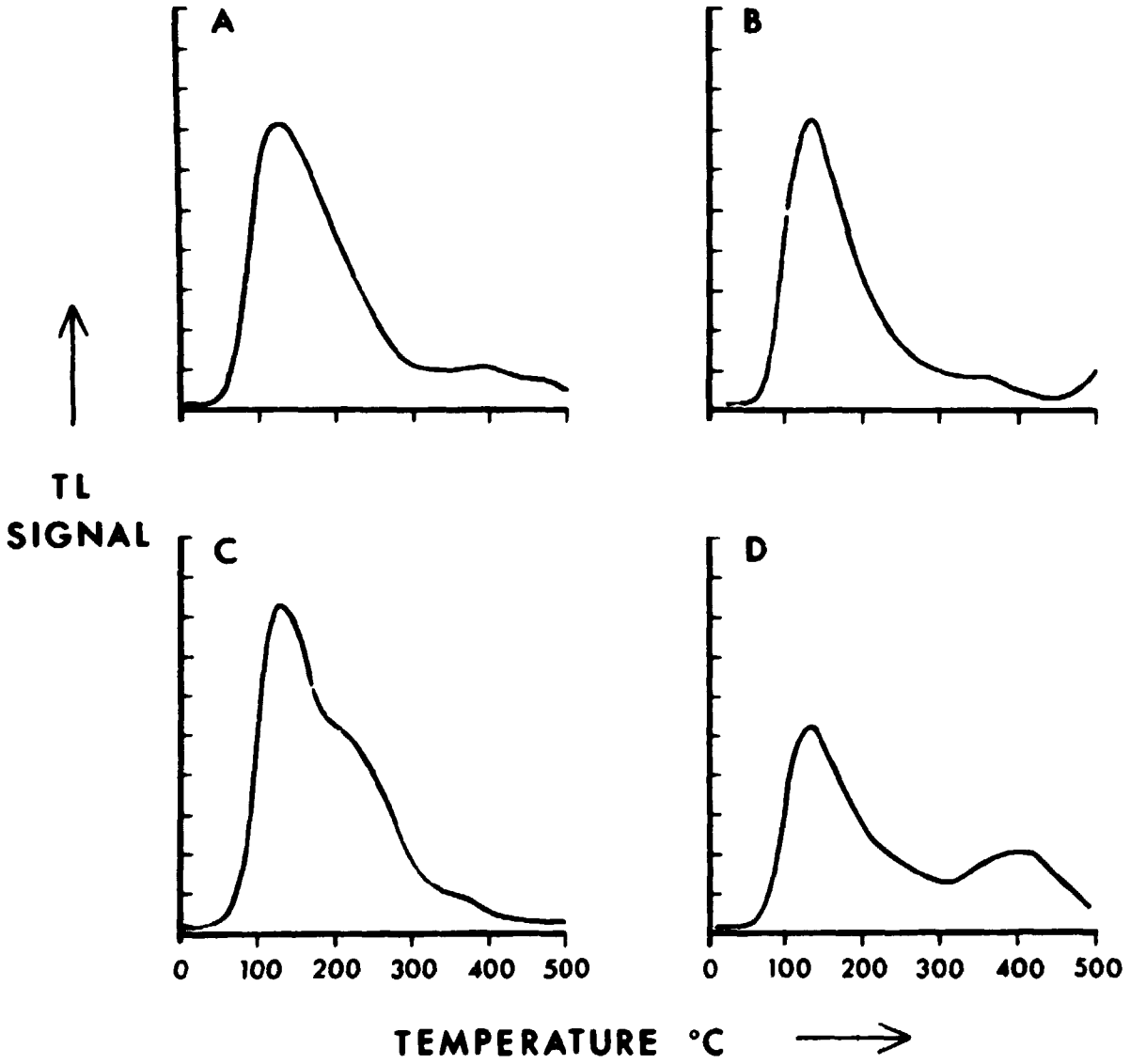

Fig. 4. Glow curves for single grains of potassium feldspar (about 1 mm in diameter) extracted from pottery or burned stones. All grains have been exposed to Sr-90 beta radiation. Only the shapes of the curves cah be compared, not the TL intensities.

A. Sherd from Brovst, Denmark. Ertebølle period. Lab. No. 721001. Dose: 150 rad beta radiation.

B. Inscribed tablet from Glozel, France. Lab. No. 744110. Dose: 350 rad beta radiation..

C. Sherd from Sarup, Denmark. Neolithic, MN IV. Lab. No. 741309. Dose: 700 rad beta radiation.

D. Burnt stone from Trabjerg, Denmark. Viking period. Lab. No. 755004. Dose: 600 rad beta radiation.

a composite peak at 200°C tailing off towards 350°C, and a high-temperature peak in the 350-400°C region which can be more or less developed.

Previously reported dating results based on feldspar inclusions (Mejdahl 1972) were obtained mainly from peaks in the region 250° - 330°C. It is obvious, however, that the high-temperature peak would be better suited for dating; and it would be advisable to restrict feldspar dating to material which has a clearly distinguishable high-temperature peak.

A dramatic fading termed anomalous fading (as much as 40% after storage for three days at room temperature) has been reported by Wintle (1973) for various feldspar types extracted from lava. Anomalous fading has also been observed for fine grains from pottery originating from vulcanic regions in Portugal (Whittle and Arnaud 1975). For the granitic feldspars - mainly potassium feldspar - from Scandinavian material a fading of this nature has not been found. A normal temperature-dependent fading was noticed for the 250-330° region amounting to 10-15% over a few months. A test carried out for a few samples over a period of two years showed a fading of 15% during the first six months and no further fading in the remaining period. Fading of the high-temperature peak has been tested for a few grains for a period of one month, and no fading was found for this period. Tests over longer periods are in progress, but it seems unlikely that a peak at this temperature should have any appreciable fading.

Since TL properties of feldspar are closely related to the type and concentration of trace elements, with europium and cerium as possible activators (Haberlandt and Köhler 1940), it is not surprising to find a marked variation with geochemical conditions at the time of formation. In fact, it is well known from investigations on preparation of artificial TL phosphors that the properties depend drastically on the activators and a number of other factors such as flux, firing temperature and cooling rate.

As for quartz, a major problem in the determination of the archaeological dose is the change in sensitivity that occurs

from the first to the second measurement. The change is not so pronounced for feldspar as for quartz; but a decrease to 70% has been observed for individual grains from different samples. Oddly enough, if a grain is left overnight after the first measurement it seems to recover its original sensitivity. An example of this recovery effect is given in Table 7 for a grain from a burnt stone (Lab. no. 755004, Viking Age, Trabjerg, Denmark). The response is expressed as integrated counts over the temperature intervals stated. The archaeological dose to the grain was approximately 800 rad. Similar results were obtained for grains from two Danish sherds (Lab. no. 720227, Iron Age, Dankirke, and lab. no. 741309, Neolithic, MNIV, Sarup).

Table 7. Response of a feldspar grain showing loss of sensitivity and overnight recovery. Archaeological dose 800 rad.

	Low-temperature peak Temp. int. 125-175°C			High-temperature peak Temp. int. 325-375°C		
	Dose	Counts	Rel. resp.	Dose	Counts	Rel. resp.
First day	10 rad	12016	0.91	Arch. + 6000 rad	3938	1.00
	10 rad	11819	0.90	6800 rad	3597	0.91
Second day	10 rad	13168	1.00	6800 rad	3919	1.00

The results for the high-temperature peak indicate a decrease of sensitivity to 0.91 as a result of the first heating and a complete recovery overnight. A simple way of eliminating the problem of sensitivity change for feldspar might be, therefore, to postpone the calibration for one day after the measurement of the archaeological dose. For the low-temperature peak a similar recovery effect is indicated. This suggests that the sensitivity change could be measured directly for each grain by measuring the response of the low-temperature peak before and after the measurement of the archaeological dose. Since only a few rads would be

required, the resulting correction of the archaeological dose would be almost negligible. The absence of any pre-dose effect for the low-temperature peak, apparent in the results above, is a general feature of that peak.

Dating by means of individual feldspar grains and - for smaller grains - samples containing a number of grains would proceed exactly as described for quartz grains. All grain sizes ranging from 0.1 mm to several mm (in burned stones) can be used. Feldspar has two important advantages:

1) The dose-rate can be determined in advance for the very sample or grain used for determination of the archaeological dose. This is of particular importance for grains from burned stones where the environment of a grain will vary so much that an average dose-rate value will not be accurate enough.

2) The dose to large potassium feldspar grains will be much larger than the dose to quartz grains of the same size because of the contribution from inherent potassium in the feldspar. The doses can differ by a factor of two for grains from pottery and even more for grains from burned stones. As an example, for the grain referred to in table 7 (fraction of a big grain, several mm in diameter) an approximate estimation of the relative contributions would be:

Internal potassium	500 rad
External beta	150 "
External gamma	150 "
Total	800 rad

The fact that potassium feldspar grains have a much larger dose than quartz grains leads to the possibility of a new important differential technique which eliminates virtually all sources of error in the conventional techniques associated with dose-rate measurements. Dating with this technique using quartz and feldspar grains from pottery would proceed as follows:

Determination of the average feldspar dose D_F
Determination of the average quartz dose D_Q
Calculation of difference $D = D_F - D_Q$

When quartz and feldspar grains of the same size are used, the average contribution from external beta and gamma radiation will be exactly the same for the two grain types. Therefore, the dose difference D must be due solely to the inherent potassium in the feldspar. The dose rate from the potassium can be determined simply by storing the feldspar grains at a low temperature in a non-radioactive material with the same scattering properties as clay (SiO_2). The gamma contribution during the storage must be measured also and subtracted:

Storage of feldspar grains at $-70^{o}C$ for a few months
Storage of gamma dosimeters for a suitable period
Determination of net dose rate $\dot{d}$ (mrad/year) for the grains

$$\text{Age (years)} = \frac{D \text{ (rads)}}{\dot{d} \text{ (rads/year)}}$$

Since the age obtained in this way is based solely on inherent potassium in the feldspar grains, all errors previously discussed in connection with the determination of the beta and gamma dose rates are eliminated. It should be possible, therefore, to achieve a considerable improvement of the accuracy of TL dating. In addition, since no knowledge of the gamma radiation at the site is required, dating could be extended to inhomogeneous sites and to material already excavated and stored on the museum shelves. This would greatly increase the flexibility of the TL method.

CONCLUSION. SUMMARY OF DATING TECHNIQUES

Note 7, 1981

A paragraph on the hot-nitrogen system has been omitted.

Two methods for measurement of beta and gamma dose rates are considered. One is with TL dosimeters using teflon discs containing $CaSO_4$:Dy. This phosphor is well tested and well suited for measurement of small doses. The other method utilizes low-temperature peaks in the quartz and feldspar grains already present in the material for the dose-rate measurements. This is technically more complicated because of the necessity to keep the material at a low temperature ($-70^{o}C$) during the irradiation period. However, the system has a greater consistency and would eliminate some fundamental calibration problems. It would also permit the use of a wider range of grain sizes for dating.

Some preliminary investigations on the TL properties of large individual grains of quartz and feldspar related to the determination of archaeological dose are discussed, including measurements of the sensitivity change caused by the first heating to $500^{o}C$.

From our examination of the different techniques for measurement of dose rates and archaeological dose four basic principles of TL dating can be formulated:

1) The 'conventional' inclusion method. Absolute dose rates are determined by means of synthetic TL phosphors such as $CaSO_4$:Dy or by natural CaF_2; and the determination of the archaeological dose is based on quartz or feldspar inclusions about 0.100 mm in diameter.

2) Dose-rate determinations and determination of archaeological dose are based on identical samples of quartz or feldspar grains. The same radiation source, or accurately intercalibrated sources, are used for all calibrations. The system can then be completely independent of source calibration and of tabulated values of

isotope half-lives and radiation energies. A wide range of grain sizes can be used, from 0.050 mm to the largest grains found in pottery (1-3 mm).

3) Dating based on individual feldspar grains with a sensitivity high enough to allow determination of the beta dose-rate (from external isotopes and inherent potassium) for each grain prior to the measurement of the archaeological dose for that grain. This principle is particularly well suited for burned stones which often contain large feldspar grains but are so inhomogeneous that an average beta dose-rate cannot be used.

4) A quartz-feldspar subtraction method. The age determination is based on the difference in archaeological dose in potassium feldspar and in quartz. Grains of the same size will receive exactly the same dose from external gamma and beta radiation; the difference in archaeological dose will therefore be contributed solely by the inherent potassium in the feldspar grains. The age obtained will thus be independent of external beta and gamma radiation; and all difficulties associated with the measurements of these radiations will be eliminated. The beta dose-rate from the inherent potassium is determined after the measurement of the archaeological dose by storing the feldspar grains at a low temperature.

We are studying thoroughly the possibilities of developing the quartz-feldspar subtraction method which evidently has great potentialities. Our preliminary investigations indicate that the method is perfectly feasible: for large grains the doses can differ by a factor of two; and the measurement of the dose rate using the low-temperature peak is straightforward. Not all sherds are suitable for this technique; some do not have both minerals and others have no large grains; but for those that can be used it should be possible to achieve a substantial improvement in the accuracy of dating. In addition, the method would be much more flexible; it could be extended to complex sites with inhomogeneous gamma-ray fields, and it could be used for already excavated material stored in the museums.

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Progress and problems with automated TL dating

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Abstract

A number of basic problems connected with the measurement of beta and gamma dose-rates are discussed, and the possibility of using low-temp temperature peaks in quartz and feldspar for dose-rate measurements is examined. Preliminary results of TL measurements on individual grains of quartz and feldspar are presented. A TL dating method based on the difference in the archaeological dose received by potassium feldspar and quartz grains is proposed.

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