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in Thermoluminescence Dating

by V. Mejdahl

March 1972

DOSIMETRY TECHNIQUES IN THERMOLUMINESCENCE DATING

by

V. Mejdahl

Danish Atomic Energy Commission
Research Establishment Risø

Abstract

The principles employed at Risø for dating of ceramics and heated stones from archaeological excavation sites are outlined. The accumulatea exposure is evaluated from the thermoluminescence emitted by quartz and feldspar inclusions with a size of 0.3 - 0.5 mm.

The environmental radiation and the radiation from radioisotopes embedded in the ceramics or the stones are measured by means of the TL phosphors $CaSO_4$:Dy and $CaSO_4$:Mn respectively. The dosimetry techniques and the preparation and properties of the two phosphors are described.

The effect of a number of random and systematic errors associated with TL dating is discussed.

CONTENTS

1. INTRODUCTION

In recent years considerable progress has been made in the dating of ceramics by thermoluminescence, in particular through the work of M. J. Aitken and his associates (Aitken 1970). The fine-grain method (Zimmerman 1971) and the inclusion method (Fleming 1970) have been refined, and the two methods have been combined to give the subtraction method (Fleming and Stoneham 1971) in which the contribution from the environmental radiation is eliminated; unfortunately, the uncertainty of the subtraction method is rather high (more than 10%). Fleming and Stoneham (1971) have further demonstrated the usefulness of the predose effect in quartz for dating purposes.

The Oxford group has also achieved remarkable results in the application of the TL technique to authenticity tests, in particular of vessels and figurines in the Hacilar style (Aitken et al. 1971) and Etruscan wall-paintings on terracotta (Fleming et al. 1971).

The work at Risø has been concentrated on the application of the inclusion technique to the dating of ceramics. In a dating programme comprising sherds and heated stones from about twenty excavation sites of known age (Mejdahl 1972) a relative uncertainty of 3 - 5% was achieved; however, the accuracy of absolute dating was limited to the 5 - 10% level. Even with this uncertainty the method might be of value for specific dating problems, but it must be realized that an absolute uncertainty not exceeding 3 - 5% will be required if TL dating is to be of general use in contries such as Denmark where the chronological frame of prehistory is already quite well established.

Ultimately, systematic errors in the method may be a limiting factor, but at present it seems possible to improve the accuracy by refining the dosimetry technique and by careful consideration of known systematic errors. In this report the dosimetry technique employed in our dating method is described, and the effect of a number of systematic errors is discussed.

2. PRINCIPLE OF OUR DATING METHOD

A clay vessel that is buried in soil will be exposed to cosmic radiation and radiation from isotopes, in particular potassium-40 and the uranium and thorium decay series, distributed in the vessel itself and in the surrounding soil. With the assumption that the radiation intensity has been constant, the time elapsed since the firing of the vessel is given by the

following simple expression:

$$TL \text{ age} = \frac{\text{Accumulated radiation exposure}}{\text{Radiation intensity}}$$

For determination of the accumulated exposure we measured the TL of quartz and feldspar inclusions with a size of 0.3 - 0.5 mm and, consequently, neglected the contribution from alpha particles. The environmental radiation was measured with $CaSO_4$:Dy and the radiation from the pottery itself with $CaSO_4$:Mn.

For the two $CaSO_4$ phosphors the peak height of the glow curve was taken as a measure of the exposure. For quartz and feldspar we have a different technique which will be described later; it amounts to integration of part of the glow curve on the high-temperature side of the glow peak.

Calibration of the TL measurements was carried out with two Co-60 gamma-ray sources: a 10 Ci source for the accumulated exposure and a 1 Ci source for the radiation intensity measurements. One advantage of using the same radioisotope for the two calibrations is that only the ratio between the source strengths has to be determined accurately.

Our TL reader was constructed by Bøtter-Jensen and Beckmann (1968). The reader has an automatic sample changer which can accomodate twelve metal planchets and incorporates a unit for peak reading. Data-logging equipment permits transfer of readings to punched tape for computer processing.

3. PREPARATION AND PROPERTIES OF $CaSO_4$ ACTIVATED WITH Dy or Tm

Combining high sensitivity and relatively small fading, the phosphors $CaSO_4$:Dy and $CaSO_4$:Tm are well suited for measurement of small radiation doses. The phosphors were developed by Yamashita et al. (1968), and properties and applications have later been described by Nambi and Higashimura (1970), Mejdahl (1970), and Yamashita et al. (1971).

The two phosphors can be prepared by the same technique, and, except for differences in the TL spectra and in supralinearity, they seem to have identical properties. Our procedure for the preparation of $CaSO_4$:Dy was as follows: 0.690 g of Dy_2O_3 was mixed with 50 g of $CaSO_4$, $2H_2O$, and the mixture was dissolved in concentrated sulphuric acid which was kept boiling by means of an electric heating mantel. The solution was evaporated to

dryness, and during the recrystallization of $CaSO_4$ the activator is incorporated in the crystal lattice. The phosphor was then heated at $700^{\circ}C$ for two hours to remove all traces of H_2SO_4 and ground to a grain size of less than 200 μm. The standard deviation of the sensitivity of 50 g batches prepared in this way was generally less than 10%.

The Dy concentration was 1.5% by weight corresponding to a mole concentration of about 1.3%. The Japanese material (Yamashita et al. 1971) has a considerably lower activator content, about 0.1 mole per cent, but we found that a low activator content resulted in reduced efficiency.

In figure 1 the response of $CaSO_4$:Tm prepared in our laboratory (Tm 71) is compared with that of $CaSO_4$:Tm obtained in 1969 from Matsushita Electric Industrial Co. (Tm 69). The phosphors were exposed to 10 R of Co-60 gamma-radiation, and one set of samples (A) was measured immediately after the exposure, while another (B) was heated for one hour at $80^{\circ}C$ before measurement. The two phosphors have approximately the same sensitivity, but the main peak of the Japanese phosphor occurs at a higher temperature and, accordingly, it has a smaller fading: 1 - 2% per month at room temperature was reported by Yamashita et al. (1971). The fading of our material will be discussed later.

The effect of post-irradiation heating is further illustrated in figure 2 which shows glow curves for samples of $CaSO_4$:Dy that were heated for one hour at 25, 50, 80, and $110^{\circ}C$ before measurement. The glow peak was shifted from $174^{\circ}C$ to $213^{\circ}C$ which indicates that a low-temperature trap is being emptied, while the trap corresponding to the $213^{\circ}C$ peak is affected to a smaller extent.

In the measurement of environmental radiation we used the phosphor as a loose powder, but we found that hard and durable tablets can be prepared by adding $Li_2B_4O_7$ as a binder and sintering the tablets (made with a tabletting machine) at $700-800^{\circ}C$. A binder/phosphor ratio in the range 0.25-0.50 was found to be adequate. Addition of a small amount of fine-grained SiO_2 (5% of the phosphor content) gave the tablets a fine, smooth surface. The tablets can be measured with the hot-nitrogen reader developed by Bøtter-Jensen (1971).

Table 1 illustrates the accuracy that can be achieved with $CaSO_4$:Dy. The standard deviation of ten measurements is given as a function of the exposure in the range of 0.5 - 100 mR. Each of the ten values is the average of results for four 150 mg samples (a standard procedure). From 0.5 to 2 mR the standard deviation reaches 4%; above 2 mR it varies between 1 and 2%.

Table 1

Standard deviation of ten measurements with $CaSO_4$:Dy
as a function of exposure. Each measurement is
the average of the response of four 150 mg samples

Exposure (mR)	0.5	1	2	5	10	20	50	100
Standard deviation (%)	2.93	3.03	3.81	0.97	2.06	0.87	1.46	1.14

In figure 3 the response of $CaSO_4$:Dy is plotted as a function of Co-60 gamma-ray exposure in the range 0.5 - 100 mR. The response is linear down to 1 mR which was taken as the lower limit of application. Since exposure of the phosphor to light may influence the response, all measurements were carried out in yellow darkroom illumination.

As might be expected from figure 2, the fading of our $CaSO_4$:Dy is appreciable and must be taken into account. In figure 4 (B) the relative response is plotted as a function of storage time for samples that were stored at $25^{o}C$ after a Co-60 gamma-ray exposure of 1 R. During the first 40 days the response decreased by almost 30%, but during the following 160 days the decay was less than 10%. If the response on the fifth day is taken as 100%, the initial decrease is reduced to 17%. The fading for storage at 80 and $100^{o}C$ is illustrated in figure 5.

The fading of our $CaSO_4$:Dy is a regrettable disadvantage and necessitates a strict correction procedure which will be discussed in section 5.

Since the atomic weight of Ca is relatively high, the energy dependence of the response per roentgen is significant for $CaSO_4$:Dy. Previously reported energy dependence studies (Mejdahl 1970) have shown that $CaSO_4$:Dy exposed in polyethylene bottles had a maximum response at 40 keV which was nine times that for Co-60 gamma radiation. When the polyethylene bottles were inserted in steel rods with a wall thickness of 1.5 mm, the maximum response occurred at 100 keV and was 2.5 times that for a Co-60 gamma-ray exposure. For a typical distribution of radioisotopes in soil the exposure would be overestimated by about 7%. However, the response approximated that of quartz and feldspar grains embedded in pottery; we therefore made no energy dependence corrections in the dating experiments.

4. PREPARATION AND PROPERTIES OF $CaSO_4$:Mn

The phosphor $CaSO_4$:Mn has been known since the end of last century and has been described by many authors. A recent survey of its properties has been given by Bjärngard (1963). In spite of the rapid fading of stored energy at room temperature (40% during the first 24 hours), $CaSO_4$:Mn has found application in the measurements of small doses (Bjärngard 1967). The main advantages of the phosphor are:

(i) High sensitivity to radiation
(ii) Negligible sensitivity to light
(iii) No annealing procedure is required; the heating during the read-out gives sufficient annealing.

$CaSO_4$:Mn can be prepared in the same way as $CaSO_4$:Dy, but larger batches can be made more easily by heating a mixture of the components at 800 - $900^{o}C$. Our method of preparation, adopted from Bjärngard (1963), was as follows: $CaSO_4$, $2H_2O$ and one mole per cent $MnSO_4$, H_2O were mixed with diluted H_2SO_4 (25%), and the mixture was left for twenty hours at 30 - $40^{o}C$. Then the water and the sulphuric acid were driven off by heating in a heating mantle, and finally the mixture was heated in air at $800^{o}C$ for 30 minutes.

Figure 6 shows a comparison of glow curves of $CaSO_4$:Mn, $CaSO_4$:Dy prepared in our laboratory, and a new Japanese phosphor, Mg_2SiO_4:Tb, which seems to have good properties (Hashizume et al. 1971). The exposure was 10 R of Co-60 gamma radiation. Before the measurement $CaSO_4$:Mn was kept for one hour at room temperature, while $CaSO_4$:Dy was heated at $80^{o}C$ for one hour.

We have used $CaSO_4$:Mn extensively for the measurement of beta-ray intensity at the surface of potsherds. Our dosimeter was a thin metal plate (kanthal) 0.35 mm x 40 mm x 60 mm which also served as heating element. 400 mg of the phosphor was fastened with silicone resin (MS 805) in a circular depression, 1 mm deep and with a diameter of 28 mm. The silicone resin was diluted with a thinner in the ratio 1:3, and 0.4 ml of the solution was added to the powder in each dosimeter. The dosimeters were then dried at room temperature for 20 hours and finally heated for 6 hours at $200^{o}C$. This treatment produced a hard layer of phosphor (65.0 mg/cm^2) with good mechanical stability. During the exposure, which lasted one week, the sherds and the dosimeters were kept in a freezer at $-25^{o}C$, and before each measurement the dosimeters were kept for one hour at room

temperature.

In the following some properties of the $CaSO_4$:Mn dosimeters relevant to our dating experiments will be discussed.

The dosimeters allow very accurate measurements; for exposures above 1 mR the standard deviation of measurements with 10 selected dosimeters was less than 1.5%. In our experiments 1 mR was taken as the lower limit of exposure, but as illustrated in figure 7 exposures as low as 0.1 mR could easily be measured.

Since we used Co-60 gamma radiation for calibration of dosimeters exposed to beta radiation, it was of interest to compare the response as a function of layer thickness for the two radiation types. In figure 8 the response to beta radiation from natural uranium (E_{max} = 2.32 MeV) and to Co-60 gamma radiation is plotted as a function of the thickness of the phosphor layer. The response was normalized to be equal for thin layers. With increasing thickness an increasing deviation from linearity occurs, but since the deviation is the same for the two radiation types, it is apparently caused by absorption of light and not by attenuation of beta radiation. Since the radiation from natural uranium probably rather well represents the radiation from pottery, the calibration should be independent of the thickness of the phosphor.

As mentioned above our normal procedure was to keep the exposed dosimeters for one hour at room temperature before the measurement; therefore, it was important to investigate the fading of exposure for periods around one hour. Figure 9 shows the response as a function of storage time for dosimeters exposed to 15 mR of Co-60 gamma radiation (exposure time 3 minutes) and kept at room temperature for periods ranging from 12 to 90 minutes. The fading rate around 60 minutes is about 0.14% per minute or 1.4% in 10 minutes.

We further investigated the fading as a function of temperature in the interval 20.0 - 28.8^{o}C for an exposure of 15 mR and a storage time of one hour (figure 10). The fading rate at 25°C is approximately 0.3% per degree C.

Even when the dosimeters were kept at -25°C, the fading was not negligible. We found a fading of nearly 4% during one week for dosimeters exposed to 15 mR of Co-60 gamma radiation.

5. MEASUREMENT OF ENVIRONMENTAL RADIATION

The intensity of the environmental radiation at archaeological excavation

sites was measured with $CaSO_4$:Dy prepared at our laboratory. The phosphor was contained as a loose powder in polyethylene bottles, 40 mm in length and with an outer diameter of 9 mm. The bottles were placed at 10 cm intervals in steel tubes, 1.5 - 2 m in length and with an inner diameter of 10 mm and a wall thickness of 1.5 mm. At each site three to five tubes were hammered into the ground and left for a period of one to three months. Included in each tube were a few bottles containing phosphor that had received a known large exposure, generally 3 R; this enabled us to evaluate the effect of fading during the exposure.

Because of the fading effect a strict calibration and correction procedure had to be followed. Phosphor samples exposed to calibration doses, and samples (A) exposed to a large dose equal to the dose given to the control bottles (B) in the steel tubes were kept for three days at room temperature before measurement. By comparison with (A) the fading of (B) could be evaluated. The fading of the dosimeters in the steel tubes was taken to be half that of (B), i.e. a linear fading was assumed, and their response was corrected accordingly.

For a three-month period (summer) the correction factor was around 1.05. For such short periods in which the temperature variation is small, the procedure described will be sufficiently accurate, but measurements over a whole year could not be carried out in this way. The fading problem might be eliminated by using the Japanese phosphors, $CaSO_4$:Dy or Mg_2SiO_4:Tb, but the high cost of these phosphors ($5-8 per g) does not allow this solution. However, a satisfactory compromise might be to include in each steel tube a few samples of a Japanese phosphor and then normalize the response of our $CaSO_4$:Dy to that of these samples.

Results of measurements at about thirty sites have been presented in a previous article (Mejdahl 1970) which included a discussion of various sources of error, in particular the energy dependence of the response of $CaSO_4$:Dy.

The environmental radiation originates from cosmic radiation and from radioactive isotopes, K-40 and the U-238 and Th-232 series, distributed in the soil. While the intensity of the cosmic radiation (about 30 mR/year at ground level) may be regarded as constant throughout the small area of Denmark, the radiation from the soil varies great with soil type. The lowest levels, 25 - 50 mR/year, were found in the melt-water deposits in West Jutland, whereas considerably higher values, up to 120 mR/year, were found in the moraine deposits in East Jutland and the Eastern Islands. Typical intensity values for different soil types are indicated in table 2.

Table 2

Intensity of environmental gamma radiation from different
soil types measured at a depth of 50 cm. The contribution
from cosmic radiation (about 20 mR/year) has been subtracted

Soil type	Radiation intensity (mR/year)
Moraine and diluvial clay from the last glacial period	60-120
Moraine and diluvial sand from the last glacial period	50- 70
Moraine and diluvial sand from an earlier glacial period	30- 40
Glacial and late glacial heath sand	25- 45
Post-glacial shifting sand	30- 50

As discussed by Adams et al. (1959) the variation of radioactivity with
soil type is caused primarily by the tendency of uranium and thorium min-
erals to adsorb to clay particles during weathering of the rocks and sub-
sequent leaching; clay-rich soils will, therefore, have a higher content of
radioactive minerals than sandy soils.

In homogeneous soil the standard deviation of measurements taken down
through the soil profile was about 2%, but in general fluctuations in soil
composition and the presence of stones with a higher content of radioactive
minerals than the soil caused a considerably larger variation. Since in
prehistoric times stones were used extensively - in particular for con-
struction of tombs - potsherds will often be deposited in a very inhomogen-
ous radiation field. In such cases pottery to be dated must be taken from
the immediate surroundings of a steel probe.

One of the basic assumptions in TL dating is that the radiation field in
the soil has been constant during the whole period for which a sherd has
been buried. Hansen and Huntington (1969) have shown that in podsolized
soils thorium formed complex organic compounds which leached down
through the soil and were reprecipitated at a lower level. The process was
similar to hardpan formation, but thorium was precipitated in a broader
zone not coincident with the hardpan. We have found indications of leaching
of radioactive minerals in medium podsolized soils, but more detailed in-

vestigations will be required to establish whether significant leaching has
taken place.

In clay-rich soils a downwash of clay may occur which results in forma-
tion of horizons with a high concentration of clay (luvisol formation). Since
uranium and thorium minerals adher to clay particles, this process will
inevitably cause a movement of those minerals as well. In one profile of
this type we have found a close correlation of thorium concentration with
clay content, which indicates that a movement of thorium has taken place
(Kristoffersen et al. 1972).

As discussed by Zimmerman and Huxtable (1969) seasonal fluctuations
in radiation intensity occur in all soil types. The fluctuations are caused
by the seasonal variation in humidity which has two opposite effects:

(1) An increase of the water content will raise the average density of
the soil; because of the increased absorption of radiation this will result in
a decrease of radiation intensity.

(2) When the pores of the soil are filled with water, the emanation of
radioactive gases - in particular Rn-222 - will be suppressed; this will
cause an increase of the radiation intensity in the soil.

We measured the combined effect of these variations over a period of
more than two years. Dosimeters from a probe placed at a locality in
Roskilde were changed and measured each month. Intensity values averaged
over the depths 30 - 70 cm are presented in figure 11 (with the contribution
from cosmic radiation included) together with average values of the monthly
precipitation. The intensity shows no pronounced seasonal periodicity, but
certain trends can be distinguished: During January, February, and March
there seems to be an increase in intensity - perhaps caused by the snow
cover which prevents the emanation of radon. During April and May a de-
crease occurs, a minimum being reached in May. During summer and
autumn the intensity in 1970 is rather close to the annual average, whereas
a certain fluctuation occurs in 1971. No obvious correlation with monthly
precipitation can be seen; this is perhaps not surprising since the emanation
of radon depends in a complicated way on several meteorological factors
(see for instance Hosler 1968 and Mattsson 1970). Average intensity values
and standard deviations for the total period and for the years 1970 and 1971
are given in table 3. It is clear from figure 11 that in general the intensity
value for a single month will not be sufficiently accurate; it is necessary to
average over several months, preferably a whole year. Table 3 shows that
the average intensity value for 1971 is significantly higher (1.6%) than the

value for 1970. This brings in the important question of long-term variations of average annual values which can be answered only by extending the measurements over a period sufficiently long to include extremes of climatic conditions.

Table 3

Average values and standard deviations of monthly measurements of gamma-ray intensity in soil. Depth range 30 - 70 cm. Cosmic radiation is included.

	Average intensity (mR/year)	Standard deviation (%)
Sep. 1969 - Dec. 1971	107.7	3.86
Jan. 1970 - Dec. 1970	107.3	3.47
Jan. 1971 - Dec. 1971	109.0	2.95

The question remains whether the intensity of the cosmic radiation at ground level had been constant during the periods of interest. The discrepancies between C-14 chronology and tree-ring chronology (Suess 1970) have revealed that the production of carbon-14 in the atmosphere has not been constant. Two types of variations have been recognized: a small fluctuation around the mean value, probably caused by fluctuations in the solar wind, and a larger deviation from the present-day value which has been ascribed to a long-periodic variation of the magnetic field of the earth which would change the magnetic shielding and thereby the intensity of the cosmic-ray flux at the upper part of the atmosphere. Measurements by Bucha (1970) show that during the past 8000 years the magnetic field of the earth may have varied by as much as $\pm$ 50% and suggest a periodic variation with a period of about 8000 years.

Also for TL dating a significant change in the magnetic shielding of the cosmic-ray flux would be perceptible. However, at the surface of the earth the effect is much less pronounced because the shielding provided by the magnetic field is small in comparison with that of the atmosphere. An estimation can be based on the so-called latitude effect, the systematic variation of cosmic-ray intensity with latitude, which is caused by the fact that the magnetic shielding is strongest around the (magnetic) equator and practically zero in regions with a latitude of more than about 50° where the

cosmic particles move parallel to the magnetic field lines. Since the average value of the latitude effect at sea level is about 10% (UNSCEAR 1962, p. 209), and the major component of the primary cosmic radiation is isotropically distributed, one can conclude that a 100% variation in the magnetic field would cause a 10% variation in cosmic-ray intensity at sea level in the equatorial regions and no essential change beyond a latitude of 50°. Since cosmic radiation contributes only about 10% of the dose absorbed by the larger minerals in ceramics, the effect on TL dates should be less than 1% even in the equatorial regions.

6. RADIATION FROM RADIOISOTOPES EMBEDDED IN THE CERAMICS

More than two thirds of the radiation dose received by the mineral grains in a sherd is contributed by radiation from radioactive elements in the sherd itself. Our procedure for evaluation of the intensity of this radiation, mainly beta radiation, can be described as follows: The sherds were ground in a mortar and dried in air; then the clay was placed in a polyethylene bag and left for at least a week to allow build-up of the radon equilibrium. The measurement was carried out with the $CaSO_4$:Mn dosimeters described in section 4. A dosimeter was attached to either side of the bag and between the phosphor and the bag were inserted several layers of plastic foil (total thickness 30 mg/cm^2) to absorb the alpha radiation from the clay (figure 12). During the irradiation, which lasted one week, the bag and its dosimeters were kept in a freezer at -25°C. The exposure was evaluated from calibration with a Co-60 gamma-ray source, and the radiation in the interior of the bag was taken to be twice the surface intensity. The standard deviation of a single measurement was about 3%. In general at least three measurements were carried out for each sherd, and therefore the standard deviation of the mean value will be less than 2%.

The technique described has one important limitation: The plastic foil absorbs not only the alpha radiation, but also part of the beta radiation as illustrated by figure 13; the thickness of the layer therefore enters as a parameter to be fixed. The thickness of 30 mg/cm^2 was adjusted so that the TL dates of a selection of accurately dated sherds from the site Dankirke agreed with the archaeological estimation. The sherds were dated to late Roman iron age, chiefly by the occurrence of fibulae in the occupation layers.

The procedure described is not regarded as a strict calibration of our technique, but as a provisional measure which allows testing of the method

by ensuring proportionality (in the absence of systematic errors) between our TL dates and absolute dates. One inherent assumption is that the energy spectrum of the beta radiation from the sherds is the same for all samples. This question has not been throughly investigated, but radioactivity measurements on a number of sherds indicated that the ratio of the concentrations of radioactive elements in sherds from Danish sites does not vary much.

As discussed by Aitken and Alldred (1972) TL dating is in principle an absolute method, not dependent on comparison with other methods, and our "semi-absolute" technique might, therefore, seem to be less satisfactory. It has the advantage, however, that one can utilize the high precision of TL dosimetry without becoming too deeply involved in the complex problems of absolute dosimetry.

Figure 14 shows the distribution of beta-ray intensity values for a total of 296 samples including sherds, bricks, clay from kilns, and heated stones. The value for sherds ranged from 90 to 220 mR/year (equivalent Co-60 gamma-ray exposure) with a mean value of 157 mR/year. The value for bricks extended over approximately the same range, whereas clay from kilns gave only about 100 mR/year. As might be expected the value for heated stones showed a much wider distribution: from 130 to more than 500 mR/year.

For a sherd that is buried in soil, fluctuation in beta-ray intensity similar to that observed for gamma radiation in soil (figure 11) might be expected. However, for the wet climate of northwestern Europe the experience expressed by Aitken (1970 p. 85) is that sherds in situ are permanently saturated with water. Also Fleming (1970 p. 140) in his work on Romano-British sherds assumed complete saturation, and Zimmerman (1971 p. 49) found that the in situ water content of a number of sherds from a British site was within a few per cent of saturation. For countries with a warmer and drier climate permanent saturation of sherds cannot be expected. For sherds from two sites in Greece Zimmerman (1971 p. 49) assumed a degree of saturation of 25 and 50% respectively.

Since the climate of Denmark is similar to that of Britain, sherds from Danish sites should also be nearly saturated with water. Preliminary investigations in the summer of 1971 (during a rather wet period) of forty-one iron-age sherds from Fredbjerg and eight neolithic sherds from Myrhøj showed that their water content in nearly all cases was within 1% of saturation.

However, even for completely saturated sherds the actual water content may vary considerably from one sherd to the other because of differences in pore volume; consequently, the fraction of beta energy absorbed in water will not be the same for all sherds. Figure 15 A shows the distribution of water absorption measurements for 96 iron-age sherds from Hurup of different sizes and from a variety of vessels (kindly supplied by K. Salewicz). The water absorption of a sherd is expressed as the difference in weight of the sherd in saturated and dry condition relative to the weight in dry condition. The water absorption ranged from 5 to 22% with a mean value of 10.7%.

The water absorption of 81 samples of sherds, bricks, tiles, and heated stones from different periods is illustrated in figure 15 B. The range for sherds was from 2 to 20% with a mean value of 12.6%. The absorption of eight samples of bricks and tiles ranged from 10 to 20%, whereas the absorption by heated stones was very small, 0 - 4%. The results indicate that bricks and tiles which have been fired at a high temperature tend to have a high porosity, but for sherds no dependence on degree of firing or age was evident (table 4). The results in table 4 are given merely as an illustration; clearly, a larger number of samples would be required for a complete investigation on the factors determining porosity.

Table 4

Water absorption by sherds (S), stones (St), bricks (B), and tiles (T) from different periods; % of weight in dry condition

Site	Material	Period	No. of samples	Absorption (%)
Farum	S	Middle Age	2	8.9 - 10.1
Farum	B	Middle Age	4	13.0 - 15.0
Rosenhof	S	Slavic	4	7.4 - 9.7
Lindholm Høje	S	Viking	6	8.3 - 15.0
Dankirke	S	Rom. Iron Age	6	7.4 - 10.8
Årupgård	S	Early Iron Age	7	15.3 - 18.9
Italy	T	Etruscan	3	11.7 - 17.5
Jyderup Skov	St	Late Bronze	3	0.4 - 1.5
Knudshoved	St	Late Bronze	6	0.8 - 2.1
Knossos	S	Minoan	2	17.8 - 19.4
Stengade	S	Early Neolithic	14	13.0 - 16.2
Ring Kloster	S	Ertebølle	4	13.5 - 16.5
Brovst	S	Ertebølle	3	9.5 - 10.5

When samples are ground to powder and dried before measurement of beta-ray intensity, a correction for variation in pore volume is required. At present we measure water absorption of all sherds accepted for dating with a view to correction for the pore size effect, but it should be noticed that the dating results presented by Mejdahl (1972) have not been corrected for this effect.

If the concentration of radioisotopes in a sherd differs from that of the surrounding soil, the outer layer of the sherd will be subject to a beta-ray intensity which is different from that in the interior of the sherd. We measured the beta-ray intensity at the surface of a layer of clay from a sherd as a function of the thickness of the layer and obtained the build-up curve shown in figure 16. The curve indicates that the thickness of the transition layer (equal to the range of the beta particles) is 1 - 1.5 mm. Since it may not always be desirable or possible to remove the outer layer of a sherd, an evaluation of the error that would arise from this surface effect might be of interest. In figure 17 the calculated error in beta-ray intensity is plotted as a function of the ratio of soil activity to the activity of the sherd for different thicknesses of sherds; a linear build-up of intensity in the transition layer (taken to be 1 mm thick) was assumed. The figure indicates that the error may be appreciable; for a 5 mm thick sherd and a ratio of soil activity to sherd activity of 50% it reaches 10%.

Table 5 presents beta-ray intensity values obtained with $CaSO_4$:Mn dosimeters for samples from typical Danish soils. The values are mean values for the depth range 30 - 100 cm. Also included is the ratio of the beta-ray intensity of soil samples to the mean value (equivalent to 157 mR/year) of that of sherds. Even for moraine clay the beta-ray intensity is less than that of pottery (ratio 85%), and for heath sand, which has the lowest content of radioactivity, the ratio is only 31%.

It is evident from figure 17 and table 5 that the error arising from the reduced beta-ray intensity in the surface layer of the sherds must be eliminated. In our present dating programme we therefore remove the outer layer (about 1 mm) of all sherds before grinding, by means of a dentist's drill equipped with a diamond bit. It should be noticed, however, that this has not been done for sherds dated earlier (Mejdahl 1972).

Table 5

Beta-ray intensity of samples from different Danish soils.
Mean value for the depth range 30 - 100 cm. Ratio of beta-ray intensity to the mean value (157 mR/year) of that of pottery

Site	Soil type	Beta-ray intensity (eq. mR/year)	$\frac{\text{Int. in soil}}{157}$ (%)
Birkendegård	Moraine clay	134	85
Skamlebæk	Moraine sand	79	50
Lånum	Diluvial sand	68	43
Engedal	Diluvial sand	54	34
Gørding Hede	Heath sand	49	31

7. ACCUMULATED EXPOSURE

The total radiation exposure incurred by a sherd since the firing of the vessel from which it originated was determined by means of quartz and feldspar grains with a size of 0.3 - 0.5 mm which were separated from the clay by sifting. The grains were washed with diluted HCl and HF, and magnetic grains were removed. If necessary the samples were treated with a mixture of bromoform and acetone adjusted so that grains with a lower density than quartz and feldspar could be removed.

Figure 18 shows the concentration (per cent by weight) of quartz and feldspar grains with a size of 0.3 - 0.5 mm for 238 sherds. The mean of the distribution is 3.7%; a sample weighing 50 g thus contained about 2 g of tempering material in the range used. However, it should be noted that since the sherds were ground in a mortar, the concentrations given in figure 18 will probably be somewhat less than the actual concentrations.

Since the beta radiation will be attenuated when traversing a large grain, the intensity at the centre of a grain will be less than the average intensity in a sherd. One might expect, therefore, that the value obtained for the accumulated exposure would depend on the size of the grains used for the determination. However, results of a preliminary investigation of this effect (figure 19) indicated that the variation of accumulated exposure with grain size was not significant. It should be noted that the sample used had a rather high content of potassium feldspar (about 22%) which will be irra-

diated uniformly by the K-40 atoms in the crystal lattice; the attenuation effect might, therefore, be less pronounced for this sample. Another factor could be attenuation of light from the centre of a large grain, which would also reduce the effect of attenuation of beta radiation.

For the TL measurement, 200 mg samples (if possible twelve for each sherd) were placed on metal planchets and heated in the reader for about 30 seconds. The initial heating rate was about 18°C per second, but for continued heating an increasing deviation from linearity occurred. The non-linear heating did not cause a reduction in accuracy - the heating cycle could be reproduced with a standard deviation of 1% - but identification of glow peaks and comparison with results obtained by linear heating is difficult. The read-out equipment (figure 20) included a data-logging unit that could register the TL signal at selected time intervals, for instance each second. A read-out cycle consisted of preheating for ten seconds followed by registration of twenty TL-intensity values which were automatically transferred to punched tape for computer processing.

When the naturally acquired TL (NTL) had been measured, the samples were annealed at 350°C for 30 minutes and exposed to Co-60 gamma radiation for calibration; normally three different exposures were applied successively to the same sample. The high-temperature parts of the glow curves for NTL and a calibration exposure of 300 R are shown in figure 21 for samples from a typical sherd. As indicated in the figure the twenty TL readings were divided into five groups and for each group the accumulated exposure was evaluated. In this way five exposure values: D1, D2, D3, D4, and D5 corresponding to different parts of the glow curve were obtained for each sherd. The D1 region normally started a little before the main peak of the NTL curve which occurred at a temperature of $220 - 230^{\circ}$C, but direct comparison of the regions with particular peaks in the glow curves of quartz and feldspar was not possible because of the non-linear heating. With linear heating the temperature regions corresponding to D1, D2, and D3 would be roughly as follows:

D1: $210 - 250^{\circ}$C
D2: $250 - 290^{\circ}$C
D3: $290 - 330^{\circ}$C

Good agreement was nearly always found between D2 and D3 (figure 22A) which indicated that fading was negligible for traps corresponding to this part of the glow curve. D1 was normally about 20% lower than D2 and D3; fading therefore seems to have occurred for the traps responsible for D1.

The accumulated exposure was taken as the mean value of D2 and D3 and for sherds older than 1500 years D4 was included, although D4 on the average was somewhat lower than D3 (figure 22B); D5 was not used.

The standard deviation of the measurement of a single 200 mg sample of quartz and feldspar grains for exposures ranging from 1 R to 1000 R is shown in table 6. For exposures above 10 R the standard deviation was 7% or less. The standard deviation of the mean value for four samples (the smallest number used) will, therefore, be about 3.5%, and when twelve samples are used, the standard deviation of the mean value will be about 2%.

Table 6

Standard deviation of a single measurement calculated
from the measurement of ten 200 mg samples of quartz
and feldspar grains. Content of feldspar about 13%

Exposure (R)	1	3	10	30	100	300	1000
Standard deviation (%)	8.0	11.1	5.9	7.0	6.8	5.9	5.1

Figure 23 shows sections of response curves corresponding to D2 for a number of samples and for exposures ranging from 100 to 1800 R. Most of the curves show a small degree of supralinearity, in particular for exposures exceeding 800 R.

When three calibration points A, B, C are given (with A corresponding to the smallest exposure), the deviation from linearity can be expressed by the ratio of the slopes of the lines BC and AB. This ratio was calculated for a total of 173 samples, and the distribution of the values for different exposure regions is shown in figure 24. For exposures below 800 R the mean value of the ratio determining the linearity is close to 1 (or 100%), but for 800 R and 1200 - 1500 R the mean value of the ratio is significantly higher than 1 and reflects the increasing degree of supralinearity of the response curves.

In figure 25 the response per roentgen is plotted as a function of exposure in the exposure range $1 - 10^{6}$R for two samples A and B (No. 701338 and No. 700252) with a feldspar content of 9 and 22% respectively. The response was normalized to 1 at 100 R, and in the range 5 - 1000 R the deviation from 1 is small. The slight increase at 1 R is probably caused by the effect of light and is of no importance, but beyond 1000 R a steep

increase occurs, in particular for curve A which reaches a peak of nearly 3.8 at 10^5R. At 10^6R curve A has decreased to 1.2 and B to 0.4.

Supralinearity is exhibited by many TL phosphors and is generally only a minor inconvenience. In some cases, however, it may be accompanied by (or caused by) changes in sensitivity resulting from previous exposures. This predose effect has been studied extensively, in particular for LiF (Cameron et al. 1968); and Fleming (1970) has shown that it occurs for some of the glow peaks of quartz and has proposed to utilize the predose effect of the 100^oC peak in dosimetry (Fleming and Thompson 1970) and in TL dating (Fleming and Stoneham 1971). Furthermore, Fleming (1970, p. 129) reported that heating alone (to 500^oC) could modify the trapping process. A detailed study of the radiation-induced increase in the sensitivity of the 100^oC peak in quartz has been carried out by J. Zimmerman (1971).

We did not study these effects systematically, but occasionally exposed the samples again to the lowest calibration value after completion of the calibration. Normally the response obtained was within a few per cent of that obtained in the first exposure, but for some samples it was evident that a significant change in sensitivity had occurred. Therefore we now intend to carry out this test routinely for all samples. However, by the procedure described one would not be able to detect a sensitivity change caused by the annealing following the read-out of NTL.

Since our samples were mixtures of quartz and feldspar, their behaviour might be related to the relative concentration of the two components. The feldspar grains could be distinguished from quartz grains by a staining technique which we adopted from Reeder and McAllister (1957). The minerals were treated with 40% HF for three minutes and then immersed in a concentrated sodium cobaltinitrite solution which stained the potash feldspars yellow, and this treatment was followed by immersion in a buffered hematein solution which stained the plagioclase feldspars purple. Quartz was not affected by the treatment. The concentrations of quartz and feldspar could then be determined by counting under the microscope. Practically all feldspars were stained yellow and were thus potash feldspars. The feldspar concentration in per cent (number of grains) was determined for a selection of samples, and the distribution of the values is shown in figure 26. For 48 samples the range extended from 0 to 40% with a mean value of 17%.

The feldspars can be removed by treatment of the samples with HF. We found that treatment with 5% HF for 20 hours made the feldspar grains so brittle than one could grind them to powder in a mortar without affecting the quartz grains and remove them by sifting.

Figure 27(A-F) shows NTL glow curves for typical samples containing quartz and feldspar grains; curve 1 gives the temperature of the heating planchet, curve 2 is the glow curve, and curve 3 is the thermal radiation from the planchet. The thermal radiation was suppressed by means of a blue-filter. The feldspar content of the samples is indicated in table 7.

Table 7

Feldspar content of the samples used to obtain the glow curves in figure 27

Figure	Sample No.	Feldspar content (%)
A	700617	18
B	701327	4
C	701501	38
D	702215	1
E	702707	1
F	702803	36

The main glow peak occurred for all samples at 220-230^oC, and a second peak can be distinguished at 310 - 320^oC except for C and E; A has perhaps a third peak at 350^oC.

For comparison, figure 28 (A, B) shows NTL glow curves for two types of potassium feldspars used for manufacture of porcelain (A is from Finland, B is from Forshammer, Sweden). The curves are similar to those in figure 27, but the main peak occurs below 200^oC, probably because the high internal radiation in the potassium feldspar allows the equilibrium between trapping and release of electrons to be extended to shallower traps.

Figure 29 shows glow curves of samples that received an exposure of 400 R and a post-irradiation heat treatment of one hour at 100^oC. A is for a sample containing 22% of feldspar, and B is for the same sample after complete removal of the feldspar grains, i.e. it is for pure quartz. Three peaks at 80, 200, and 340^oC are prominent in B. The acid treatment caused a reduction in sensitivity by a factor of four, but one cannot tell without further investigation whether this was a consequence of the removal of the feldspar grains or was due to a reduction in sensitivity of the quartz grains caused by the acid treatment. It is probable, however, that feldspar grains are considerably more sensitive than quartz grains so that the response of

the samples might be determined to a large extent by their content of feld-spar. In order to elucidate these problems we are planning a more system-atic investigation of the response of quartz and feldspar grains.

The luminescence of feldspar has been studied by Haberlandt and Köhler (1940) who found that europium and cerium were possible activators. Recent investigations on the TL of feldspars with a view to their application in dosimetry have been reported by Ichikawa et al. (1970).

In connection with the response of quartz and feldspar it is of interest to notice the large variation in sensitivity exhibited by samples from dif-ferent sherds. Figure 30 shows the distribution of response of 112 samples for an exposure of 500 R. Values for samples from the site Dankirke are confined to the region 10 - 45 on a relative scale, whereas values for samples from another site, Fredbjerg, extend through the entire range 10 - 185. It is important to note that the large variation observed is not caused by differences in feldspar content, but must be related to sensitivity differences. A study of the impurities in quartz and feldspar grains which differ widely in sensitivity might, therefore, be of value for the under-standing of the response of the grains.

8. CONCLUSION

Judging from advances made in TL dating in recent years there seems to be a possibility that the accuracy can be improved to such a degree that the method can be used for practical dating purposes.

The survey of the dosimetry technique given in this report allows an evaluation of the uncertainty resulting from random errors in the dose measurements. As mentioned previously the TL age of a sherd is given by the following expression:

$$TL\ age = \frac{D}{b+d}$$

where .

D = accumulated exposure

b = intensity of beta radiation in the sherd

b = intensity of environmental radiation.

The standard deviations associated with the three measurements can be estimated as

D: 4%

b: 3%

d: 6%

The standard deviation associated with the intercalibration of the two Co-60 sources used for the calibration yielding D and those yielding b and d is about 2%. With the assumption that b = 2/3 (b+d) and d = 1/3 (b+d) the resulting standard deviation (SD) of the TL age will be given by

$$(SD)^2 = 4^2 + (3 \times 2/3)^2 + (6 \times 1/3)^2 + 2^2 = 28$$

or

$$SD = 5.3\%$$

If no correction is made for variation in pore volume of the sherds, this factor will enter as a random error of about 4% (figure 15 A) in the measurement of sherds from the same context. When this error is in-cluded, the resulting standard deviation for a single TL age determination will be

$$SD = \sqrt{28 + 5^2} = 6.6\%$$

This value would be the expected standard deviation in a comparison of TL datings of sherds from the same context. We normally use at least four sherds from each context; the expected standard error of the mean value would thus be about 3.3%. Standard errors obtained for a number of contexts in a previously described dating programme (Mejdahl 1972) agree very well with the figure derived above.

In addition to the random errors various systematic errors have been discussed, but an estimation of the combined effect of these errors will not be attempted here since some of them may be eliminated, and others are not yet known with sufficient accuracy. Perhaps the most difficult to deal with are those associated with the response of quartz and feldspar grains to radiation. Since these materials cannot be regarded as homogen-ous TL phosphors, results obtained for a limited number of samples will not necessarily have general validity.

Information on systematic errors can be gained by employing the TL method on material from sites which have been well dated by other methods,

in particular the carbon-14 method combined with dendrochronology. The technique described in this report is now being used in a new programme comprising about twenty carefully selected sites.

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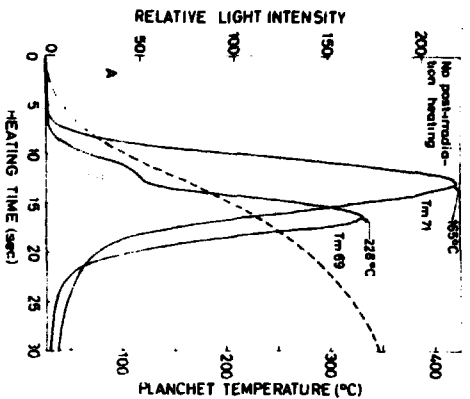
RELATIVE LIGHT INTENSITY

50 100 150 200

0

5

10

HEATING TIME (sec)

15

20

25

174°C

172°C

184°C

215°C

Post-irradiation heating

1 hour at 25°C

1 hour at 50°C

1 hour at 60°C

1 hour at 110°C

100 200 300 400

PLANCHET TEMPERATURE (°C)

Fig. 2. Glow curves of CaSO₄:Dy samples kept for one hour at different temperatures before the read-out.

PEAK-HEIGHT RESPONSE (relative units)

0.5 1 10 100 1000

EXPOSURE (mR)

Fig. 3. Response of CaSO₄:Dy as a function of Co-60 gamma-ray exposure.

RELATIVE LIGHT INTENSITY

50 100 150 200

0

5

10

HEATING TIME (sec)

15

20

25

30

A

No post-irradiation heating

Tm 71

Tm 69

165°C

228°C

-100 -200 -300 -400

PLANCHET TEMPERATURE (°C)

RELATIVE LIGHT INTENSITY

50 100 150 200

0

5

10

HEATING TIME (sec)

15

20

25

30

B

Post-irradiation heating one hour at 60°C

Tm 71

Tm 69

188°C

224°C

100 200 300 400

PLANCHET TEMPERATURE (°C)

Fig. 1. Glow curves of CaSO₄:Tm without (A) and with (B) post-irradiation heating. Tm 69 was obtained from Matsushita Electric Industrial Co., Japan. Tm 71 was prepared in our laboratory. Exposure 10 R of Co-60 gamma radiation. The dashed curve indicates the temperature of the heating planchet.

Fig. 4. Fading of CaSO$_4$:Dy at a storage temperature of 25°C. Exposure 1 R of Co-60 gamma radiation.

A: With the response on the fifth day taken as 100%.

B: With the response immediately after the exposure taken as 100%.
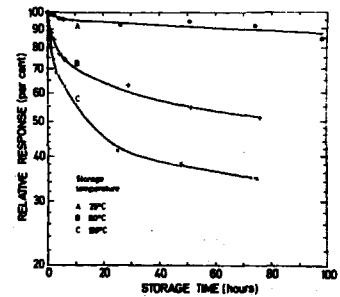
Fig. 5. Fading of CaSO$_4$:Dy at different storage temperatures. Exposure 1 R of Co-60 gamma radiation.
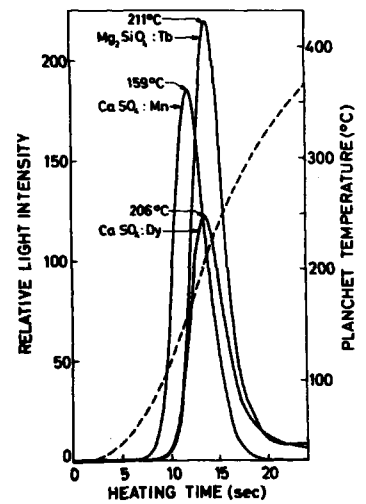
Fig. 6. Glow curves of CaSO$_4$:Mn, CaSO$_4$:Dy, and Mg$_2$SiO$_4$:Tb at a Co-60 gamma-ray exposure of 10 R. The dashed curve indicates the temperature of the heating planchet.
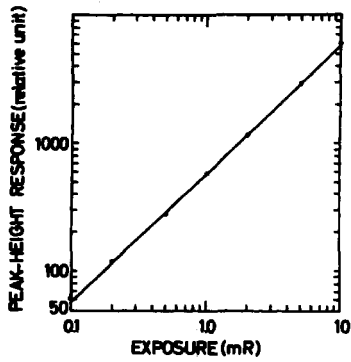
Fig. 7. Response of CaSO₄:Mn as a function of Co-60 gamma-ray exposure.
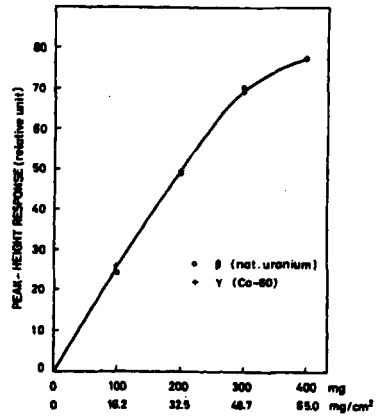
Fig. 8. Response of a CaSO₄:Mn dosimeter as a function of the thickness of the phosphor layer.
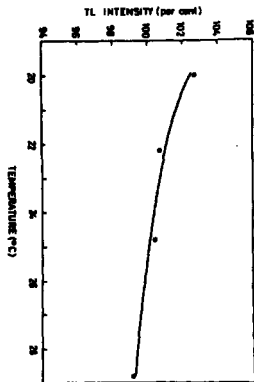
Fig. 10. Fading of CaSO₄:Mn as a function of storage temperature. Storage time 60 minutes. Exposure 15 mR of Co-60 gamma radiation.
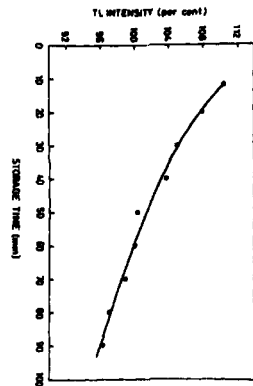
Fig. 9. Fading of CaSO₄:Mn as a function of storage time. Storage temperature 25°C. Exposure 15 mR of Co-60 gamma radiation.
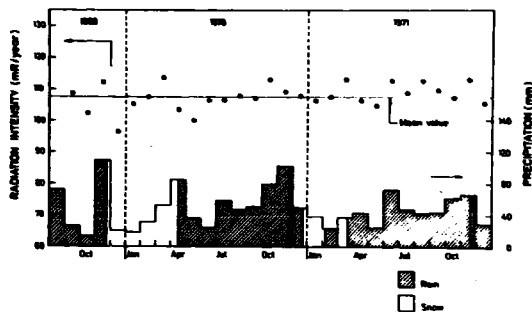
Fig. 11. Monthly variation of radiation intensity in soil. Average values for the depth range 30 - 70 cm. Monthly average values of precipitation.

Fig. 13. Response of CaSO₄:Mn dosimeters to beta radiation from clay as a function of the thickness of the absorbing layer of plastic foil between the clay and the dosimeters.

CaSO₄ : Mn

Metal planchet (kanthal)
Plastic foil (30 mg/cm²)

Plastic bag with clay

Plastic foil (30 mg/cm²)
Metal planchet (kanthal)

CaSO₄ : Mn

Fig. 12. Measurement of beta radiation from clay with CaSO₄:Mn dosimeters.
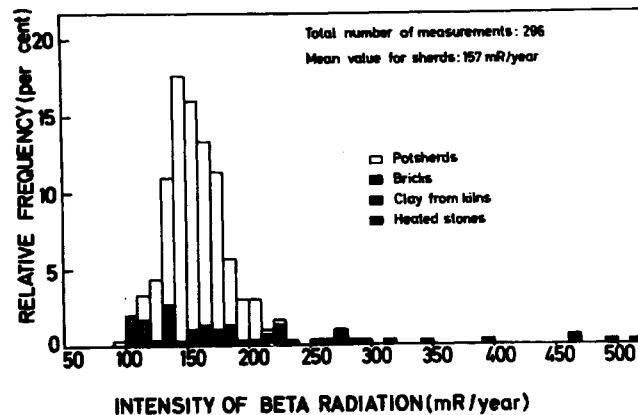
Fig. 14. Distribution of intensity values of beta radiation in clay from pottery, bricks and kilns, and in heated stones.
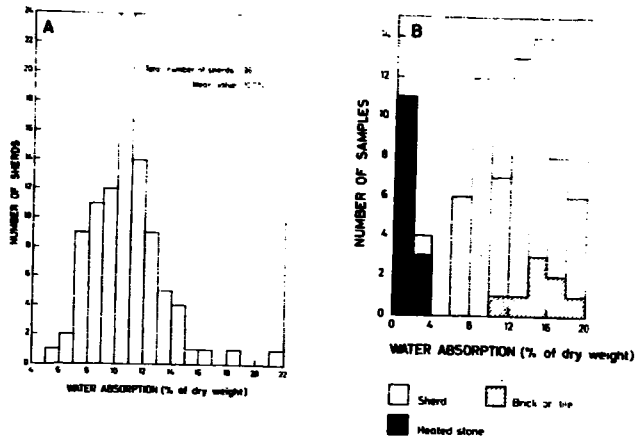
Fig. 15. Distribution of values of water absorption (% of weight in dry condition) for

    A: Potsherds from a variety of vessels from one site
      (Hurup, Iron Age, A.D. 50).
    B: Sherds, bricks, tiles, and heated stones from sites ranging
      from the Viking to the Ertebølle period (see table 4).
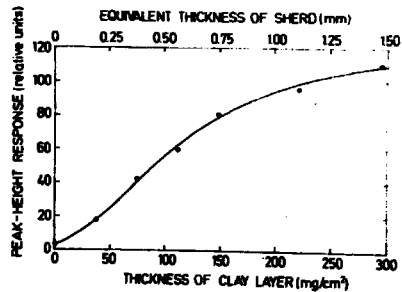
Fig. 16. Response of a $CaSO_4$:Mn dosimeter to beta radiation from clay as a function of the thickness of the clay layer. Plastic absorber 30 mg/cm^2.
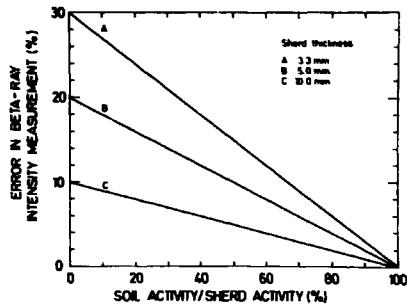
Fig. 17. Calculated error in the measurement of beta-ray intensity in pottery resulting from a difference in the concentration of radioactive elements in a sherd and the surrounding soil.
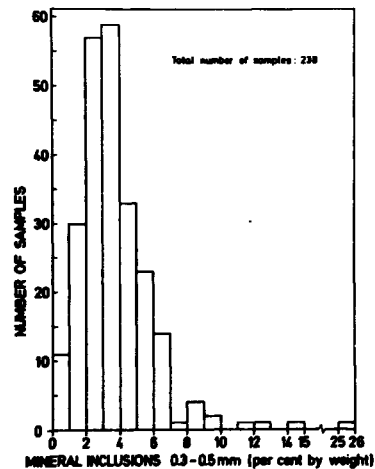
Fig. 18. Distribution of the values of the content of tempering material with a size of 0.3 - 0.5 mm.
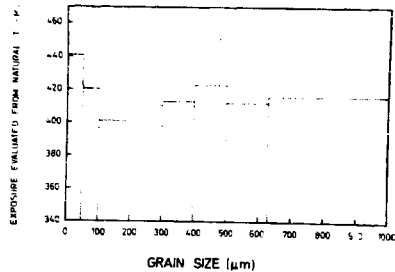
Fig. 19. Accumulated exposure of a sherd evaluated from mineral inclusions of different sizes. Feldspar content 22%.
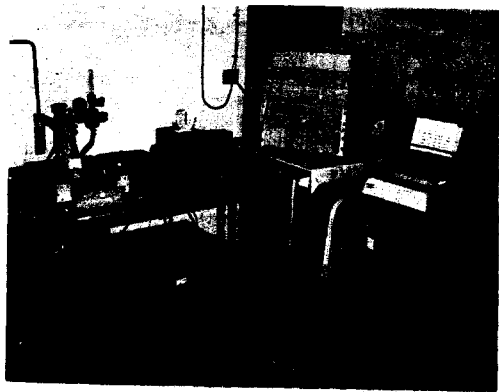
Fig. 20. Equipment for measurement of TL. Left: Reader incorporating an automatic sample changer with a capacity of twelve samples. Centre: Data-logging unit. Right: "Teletype" printer and puncher.
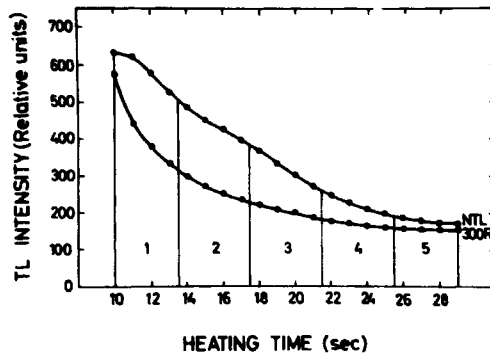
Fig. 21. TL intensity as a function of heating time for quartz and feldspar grains extracted from a sherd. NTL is the TL released during the first heating of the sample; "300 R" is the TL induced in the same sample by a Co-60 gamma-ray exposure of 300 R. The five regions indicated yield five exposure values: D1, D2, D3, D4, and D5. Initial heating rate about $18^{\circ}C/s$.
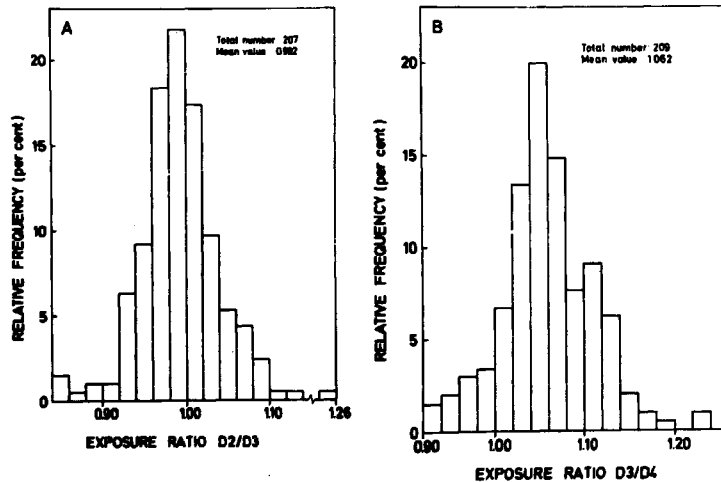
Fig. 22. Distribution of values of
A: The exposure ratio D2/D3.
B: The exposure ratio D3/D4.
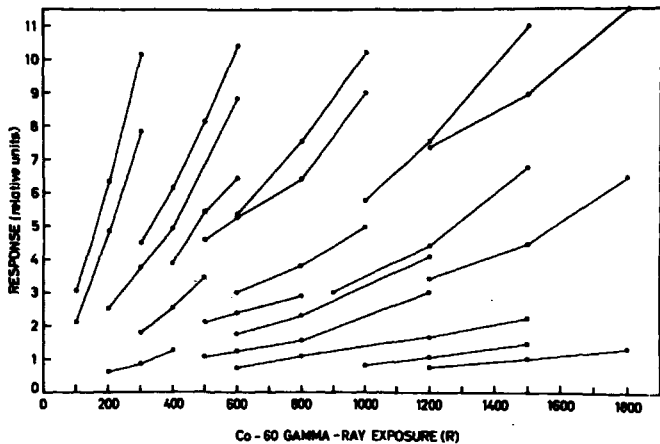
Fig. 23. Calibration curves for quartz and feldspar grains from pottery and heated stones. Exposure Co-60 gamma radiation.
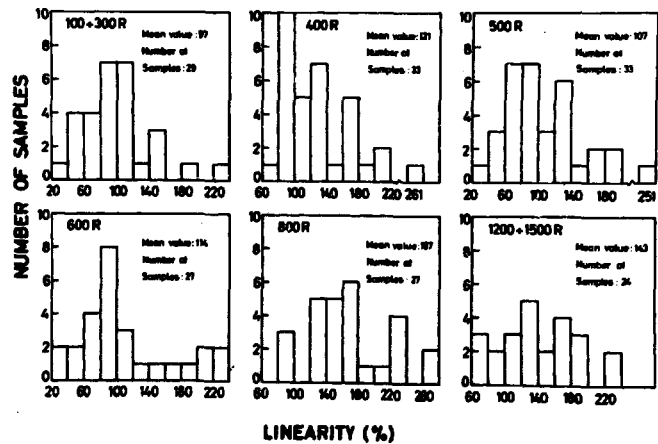
LINEARITY (%)

Fig. 24. Distribution of linearity values for calibration curves for quartz and feldspar grains. Linearity of a section containing three points A, B, C is defined by the ratio; Slope of BC/slope of AB.
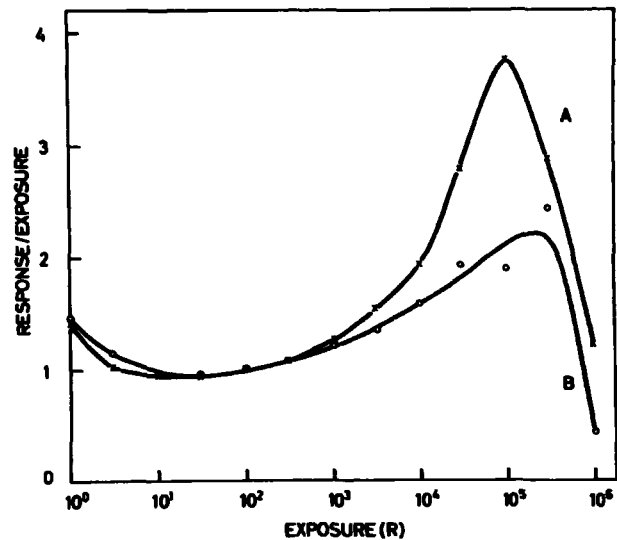

Fig. 25. Response per roentgen of quartz and feldspar mixtures for the exposure range $1 - 10^6$R. Feldspar content: A 9% and B 22%.
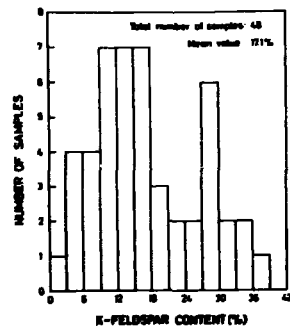

Fig. 26. Distribution of values of the potassium feldspar concentration in samples of tempering material with a size of 0.3 - 0.5 mm.
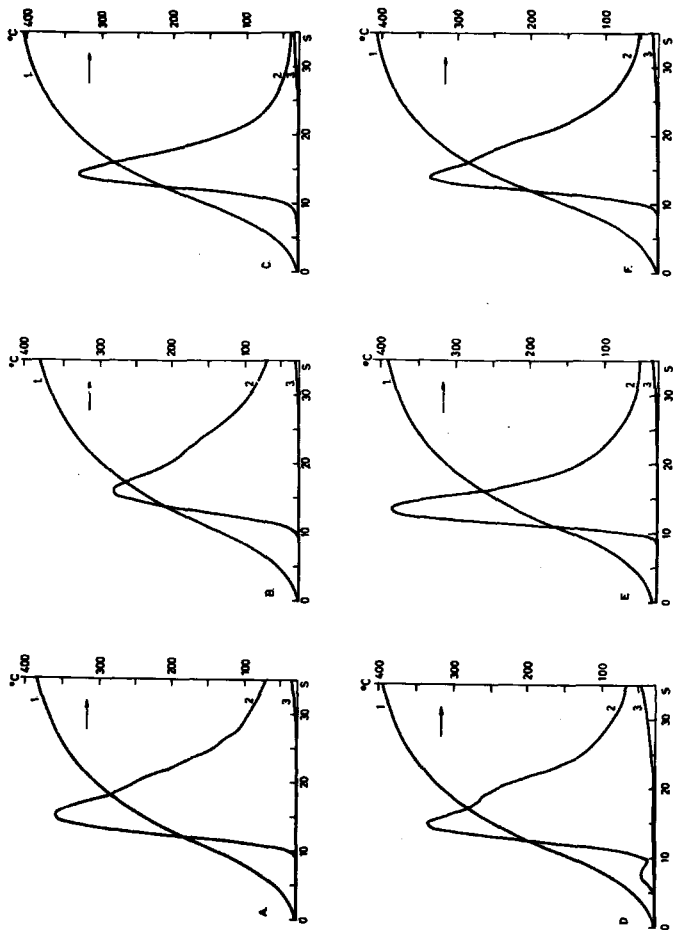
Fig. 27. Typical natural glow curves (NTL) of mineral inclusions in
pottery. Abscissa: Heating time.
Curve 1: Planchet temperature.
Curve 2: Natural glow curve.
Curve 3: Thermal glow from the planchet.
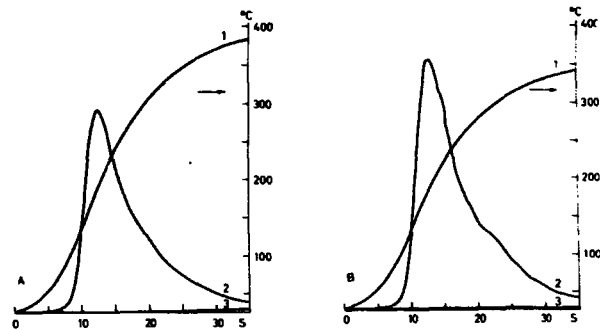Potassium feldspar concentrations: See table 7.

Fig. 28. Natural glow curves of feldspars used for manufacture of porcelain.
Abscissa: Heating time.
A: Flotation feldspar from Finland.
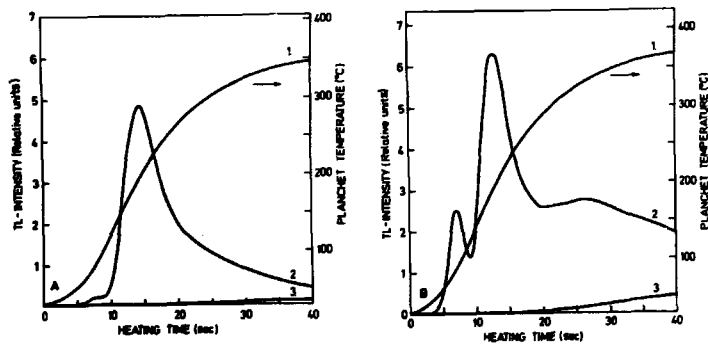B: Feldspar from Forshammer, Sweden.

Fig. 29. Glow curves of mineral inclusions at an exposure of 400 R of
Co-60 gamma radiation.
A: Sample containing 22% of potassium feldspar.
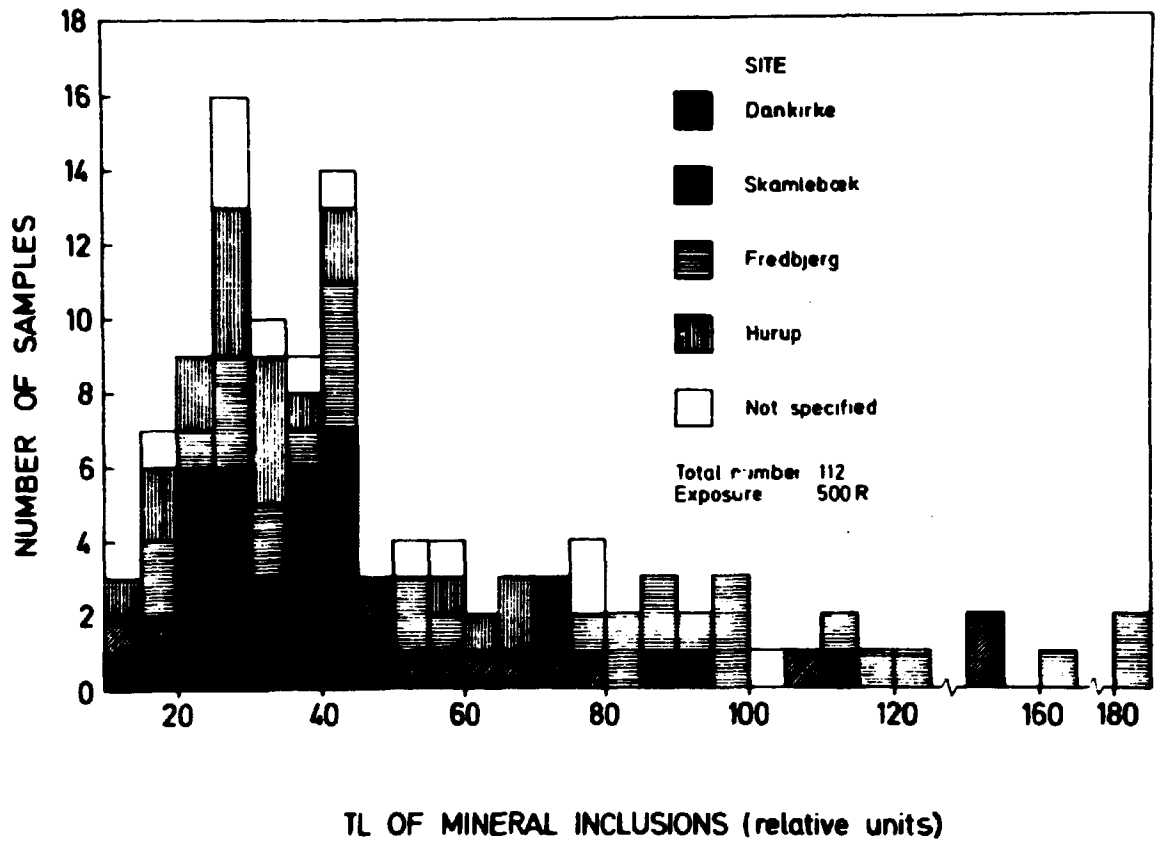B: The same sample after complete removal of the feldspar content.

Fig. 30. Distribution of response (ir the D2 region) of mineral inclusions at a Co-60 gamma-ray exposure of 500 R.