

CHAPTER 7.1

**Thermoluminescent Dating in Archaeology:
Introductory Review**

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The dating of ancient pottery by thermoluminescence measurements was suggested by Prof. Farrington Daniels some thirteen years ago². Encouraging initial trials of the technique were reported about six years ago^{3, 5}, and widespread interest was aroused among archaeologists; not only would it provide a check of radiocarbon results but it would have the strong advantage of being applicable to a material which was more plentiful than carbon and which was literally the fabric of ceramic typology studies. As yet there is no laboratory which has established the technique on a routine basis and it is still a matter of known-age test programmes. Any consideration of the solid-state physics involved leads one to question whether there can be real hope of success but, while test programmes based on comparatively naive techniques continue to show an approximate age correlation, one feels that a more sophisticated approach ought to lead eventually to a reliable system of absolute dating. The purpose of this paper is to review assumptions inherent

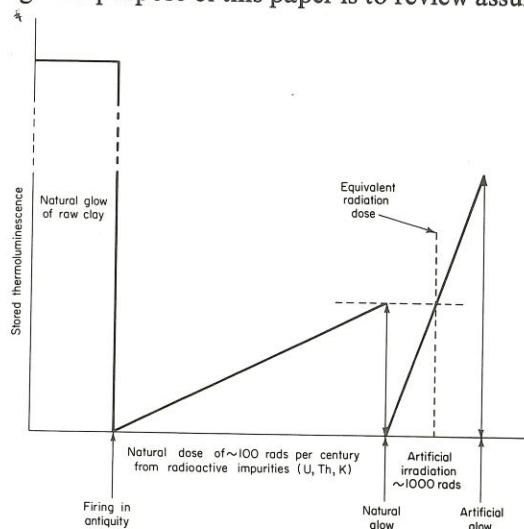


FIG. 7.1. Simplest basis for TL dating.

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Figure 7.1 illustrates the simplest basis for dating and it can be considered in three parts:

1. The acquisition of natural TL. It is assumed that the stored TL in the pot starts from zero at the time of firing from raw clay and that it then accumulates linearly with time.

2. Measurement of thermoluminescent sensitivity by exposure to an artificial source. It is convenient to express this as the radiation dose which induces TL of the same magnitude as the natural TL (the "equivalent radiation dose").

3. Assessment of the natural dose-rate to which the pot has been exposed. It is usually assumed that this is entirely due to the uranium, thorium and potassium contents of the pot itself and that the environmental contribution is negligible.

ACQUISITION OF NATURAL TL

Besides the TL induced by radiation most pottery carries an appreciable component of "spurious" TL. The origin of this is not well understood (see Paper 3-5) but it is of the utmost importance that the measurement technique used should eliminate this spurious component because it is not age dependent. In pottery the spurious TL is a surface effect which seems to be eliminated if the glow-curve is taken in an atmosphere of oxy-free nitrogen or oxy-free carbon dioxide. Unless this technique is used a sample of zero age is liable to exhibit non-zero TL.

The assumption that the acquisition of the true radiation-induced TL is linear with time presumes that the number of traps remains constant and that

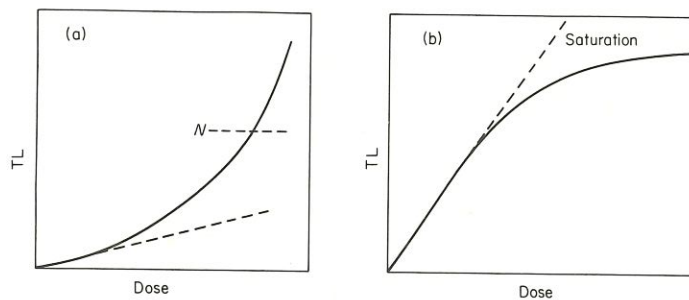


FIG. 7.2. Non-linearity of TL growth. (a) supralinear; (b) sublinear. *N* indicates typical level of natural TL with respect to supralinear portion of growth curve.

the population of the traps is well away from saturation. Figure 7.2 shows the "supralinearity" to be expected if new traps are created, and the exponential-type levelling off which will occur when saturation is approached. Some degree of supralinearity is often observed in pottery but it is attributed to subtle competition effects between different types of trap rather than to the creation of new traps (see Paper 7.3). Whatever its cause, supralinearity of acquisition is clearly a serious barrier.

Saturation effects are not of importance in pottery dating because of the low dose levels involved. However a levelling-off in the TL growth curve can also occur if the rate of escape of electrons from traps becomes significant

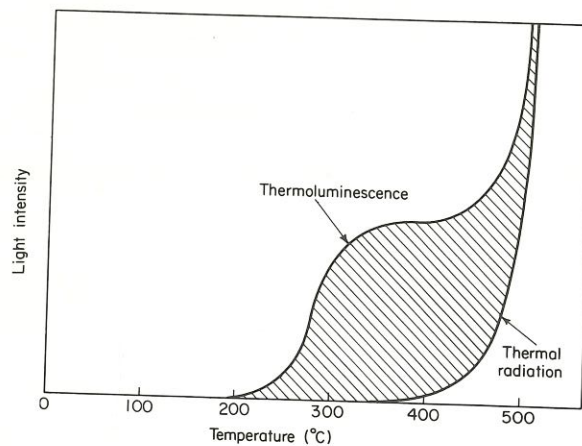


FIG. 7.3. Typical glow-curve from ancient pottery.

when compared with the rate of filling. The stability of the trapped electrons over thousands of years is then of vital importance. Figure 7.3 shows a typical natural glow-curve from pottery; the absence of appreciable TL below 250°C is taken to indicate that such decay effects are dominant for electrons from traps which are emptied in the early part of the glow curve. However, it is assumed that above about 350°C decay effects are negligible. Since this latter assumption is so vital it is worthwhile noting that there is some theoretical justification for it. To account for the absence of peaks in the glow-curve it is usually assumed that the traps have a continuum of depths such as is depicted in Fig. 7.4. The probability of escape from a trap of depth E at temperature $T^{\circ}\text{K}$ is given by $p = s \exp(-E/kT)$ where the frequency factor $s \sim 10^9 \text{ sec}^{-1}$,

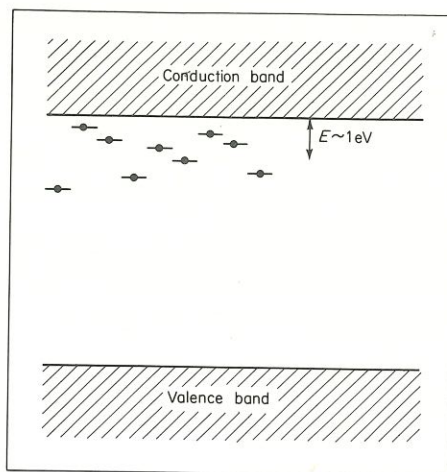


FIG. 7.4. Continuum of trap depths.

and Boltzmann's constant k equals $8.6 \times 10^{-5} \text{eV}/^\circ\text{K}$. The lifetime, τ , of an electron in such a trap is

$$\tau = \frac{1}{p} = \frac{1}{S} \exp(E/kT) \quad (1)$$

An approximation to the trap depth corresponding to a glow-curve temperature T_g is obtained by setting $\tau \sim 1$ sec, giving:

$$E \sim \frac{T_g}{500} \text{eV} \quad (2)$$

Substituting into (1) the lifetime during storage at 17°C of an electron in a trap which gives out its light at T_g may be calculated as

$$\tau \sim 10^{\frac{T_g}{29} - 17} \text{ years}$$

So for $T_g = (273 + 250)^\circ\text{K}$, $\tau \sim 10$ years, whereas for $T_g = (273 + 350)^\circ\text{K}$, $\tau \sim 30,000$ years.

Hence on the simple model used the above assumption of stability in the TL emitted at 350°C is justified. This is not necessarily the case for other models nor must the possibility of a "tunnelling" process, independent of

thermal excitation, be completely forgotten. Even on the simple model some variation in the transition temperature from unstable to stable TL is likely from material to material because of differences in the frequency factor, s , from the value assumed in deriving (2). A partial check of this for each individual pot is provided by the "ordinate ratio test". Here the ratio of the natural TL intensity to the TL intensity observed after artificial irradiation is plotted for each glow-curve temperature.

Effectively this is what is done in Fig. 7.5 by plotting the dose equivalent to the natural thermoluminescence as a function of glow-curve temperature.

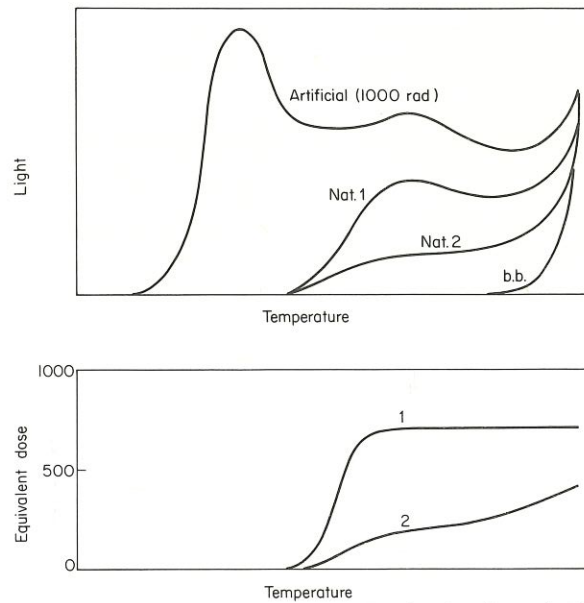


FIG. 7.5. The ordinate ratio test. (a) Glow-curves; (b) ratio of ordinates in (a) for case 1—good plateau, and case 2—no plateau.

Where this levels off to a plateau above a certain temperature—as in case 1—one is much more confident that decay effects are absent (above that certain temperature) than in case 2 where there is no plateau. Failure to obtain a plateau may also be due to the presence of spurious TL in the natural glow-curve because the glow-curve shape corresponding to spurious TL is different from that of radiation-induced TL. Whatever the cause, absence of a plateau is good reason for rejection of the pot from a dating programme.

The intensity of the TL from pot is often very weak and a highly sensitive detection system is necessary. While photomultiplier noise can be reduced by cooling the photocathode, the only way of reducing the signal due to thermal

radiation from the sample is to interpose blue filters; this also cuts down the TL and there is a limit to the beneficial effect of filters. The ratio of TL intensity to thermal radiation intensity can also be increased by heating-up at a faster rate; the former is proportional to the heating-rate but the latter is independent of it. The limitation here is due to difficulties arising from thermal lags in the sample: if these are excessive one part of the sample may be emitting TL appropriate to say 400°C while another part is still at 300°C so that the light received by the photomultiplier is a mixture of "stable" and "unstable" TL. In practice heating-rates of up to 20°C/sec with a 15 mg sample spread over one square centimetre can be used without such effects becoming serious. It is not necessary for the heating to be linear but it is essential that it is reproducible—otherwise the ordinate-ratio test will never yield a plateau.

MEASUREMENT OF TL SENSITIVITY

In the simple scheme of Fig. 7.1 it is implied that the artificial dose is applied to the same portion of ground pot as was used for measuring the natural TL. This procedure has the advantage of avoiding possible lack of homogeneity between two different portions, but it has the disadvantage that when heated for measurement of the natural thermoluminescence, the sample may have undergone mineralogical changes. If so, the sensitivity measurement is not necessarily relevant to the state of the material when it acquired its natural TL. The alternative procedure is to apply the artificial dose to a second, undrained, portion of the sample and obtain the magnitude of the artificial TL by subtracting the natural TL measured on the first portion. This procedure is satisfactory, apart from possible inhomogeneity effects, as long as the TL growth is linear. It is not at all clear what can be done to correct for non-linearity.

Apart from the above considerations it should not be forgotten that the artificial TL is obtained from a dose-rate which is often 10^8 times that of the natural dose-rate. However, in the field of medical dosimetry where this question has received attention, no evidence of dose-rate dependence has yet been found.

Another difference between the natural dose and the artificial dose is that the former is carried predominantly by alpha particles whereas the latter is more conveniently applied in the form of beta, gamma or X-radiation. Artificial dosage with alpha particles is difficult because of their very short range, but there is growing evidence that in most substances alpha particles are between one-fifth and one-tenth as effective for inducing TL, when compared per rad of absorbed dose, with beta and gamma radiation^{1, 4}. In addition to this the effective alpha particle dose may be further reduced if, as

is often the case, the particle size of the thermoluminescent material is larger than the ranges of the alpha particles. A later paper in these proceedings (Paper 7.8) describes circumstances in which the alpha-dose contribution to the TL is negligible.

THE NATURAL RADIATION DOSE

The essential features of the decay schemes of the relevant radio-isotopes are shown in Table 7.I; in addition to those shown there is also a small amount of actinium associated with the uranium. It is usual to determine the uranium and thorium content of pottery by alpha counting and the potassium by chemical analysis. The conversion factors for obtaining the dose-rate in rads/yr are given in Table 7.II. Because the alpha count that corresponds to a total dose-rate of 1 rad/yr is nearly the same for uranium and thorium, a knowledge

TABLE 7.I
Decay schemes for ²³⁸U, ²³²Th and ⁴⁰K.

²³⁸ U	4.56 × 10 ⁹ yr	²³² Th	1.42 × 10 ¹⁰ yr	
	1α, 2β			
²³⁴ U	2.67 × 10 ⁵ yr		2α, 2β	
	2α			
²²⁶ Ra	1590 yr	ThX(²²⁴ Ra)	3.64 days	
	1α		1α	
²²² Rn	3.8 days	²²⁰ Tn	54.5 sec	
			1α	
		ThA (²¹⁶ Po)	0.16 sec	
			2α, 2β	
²⁰⁶ Pb		²⁰⁸ Pb		
Q = 46.7 MeV		Q = 39.8 MeV		
8α, 6β		6α, 4β		

The diagram shows the decay of ⁴⁰K. It can decay via Electron Capture (EC) to ⁴⁰Ar (11%) or via beta decay to ⁴⁰Ca (89%). The beta decay has a maximum energy of 1.36 MeV. The ⁴⁰Ar product is in an excited state and decays to its ground state by emitting a gamma ray of 1.46 MeV.

TABLE 7.II

Dose-rates and alpha counts corresponding to given concentration of uranium, thorium and potassium.

Concentration	Uranium	Thorium	Potassium
Alpha count /1000 sec, thick sample, 15.2 cm ² , eff = 0.85	3.2 ppm	12.7 ppm	8.85%
	6.8	7.15	—
Total Dose	1 r/yr	1 r/yr	1 r/yr
	α / β / γ ↓ ↓ ↓ 0.92 0.042 0.038	α / β / γ ↓ ↓ ↓ 0.905 0.028 0.067	β / γ ↓ ↓ 0.765 0.235
For $(\beta + \gamma) = 0.1$ r/yr	4 ppm	13.4 ppm	0.885%

of the relative concentrations only becomes necessary when the individual contributions of beta and gamma radiation are needed. A rough value for the uranium/thorium ratio can be obtained by the "pairs" technique⁶ in which use is made of the fact that the 0.16 sec half-life of thorium A is much shorter than any other in either chain.

Alpha counting is preferable to chemical determination of uranium and thorium because of the gross errors which would be introduced by the latter procedure if the decay chains were not in equilibrium. Because of the 267,000 year half-life of ²³⁴U this may be the case for some glacial and post-glacial clays. Another cause for disequilibrium in the uranium series is the escape of radon (²²²Rn) from the pottery so that subsequent daughters are deposited elsewhere. Measurements by Waine⁷ indicate that for typical pottery only about 10% of the radon escapes; also the percentage escaping is the same whether the pot is in solid form or whether it is ground for counting. Since the alpha count corresponding to 1 rad/yr does not differ markedly between the two halves of the chain (pre-radon and post-radon) the error introduced is small.

When account is taken of the reduced effectiveness of the alpha dose a more

Careful scrutiny of the above procedure is necessary, and certainly when circumstances are such that the effect of the alpha dose is negligible it is far from satisfactory. If the alpha dose does not have its full value, the beta and gamma doses become important, and because of their greater range this

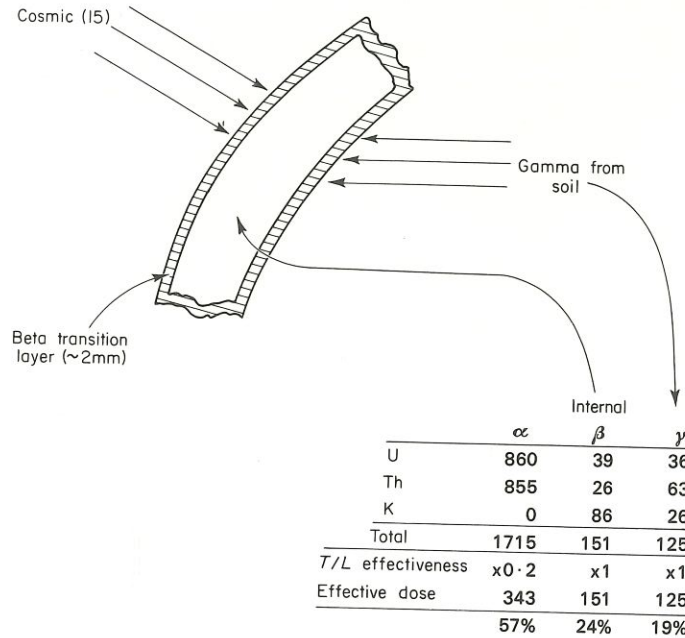


FIG. 7.6. Typical annual radiation dose for fragment of pot buried in soil, both having U 3 ppm; Th 12 ppm; K 1%.

means that the dose received by the pot is determined not only by its own radioactive contents but also by environmental radiation. The situation for typical pot is shown in Fig. 7.6. The major environmental contribution is the gamma dose from the burial soil but even for the beta dose there is a transition layer at the surface of the pot in which the dose is partially determined by the radioactive content of the pot and partially by that of the soil. The cosmic-ray contribution is estimated for burial 5 ft below the surface.

Because of the uncertainties inherent in converting alpha counts to effective dose it is desirable to use a more direct technique for evaluating the latter—such as dosimetry by thermoluminescence itself as described in a later paper in these proceedings (Paper 8.3).

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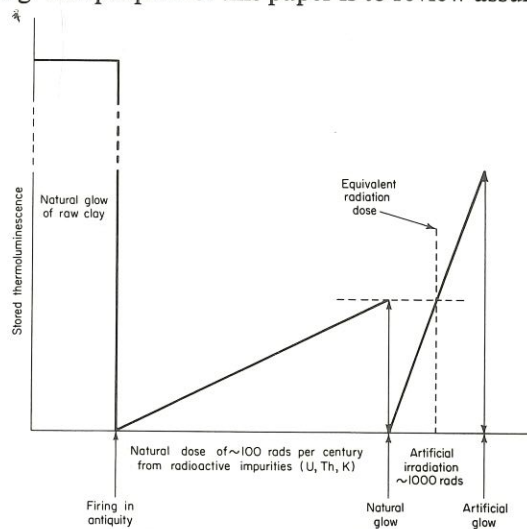


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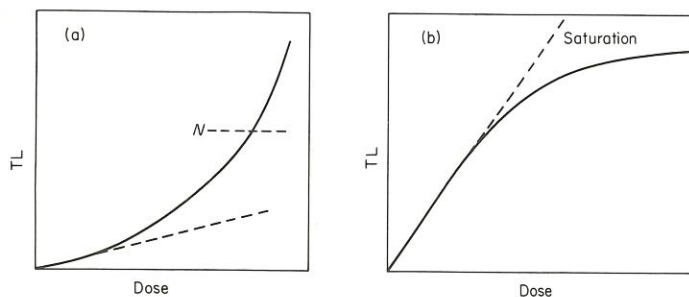


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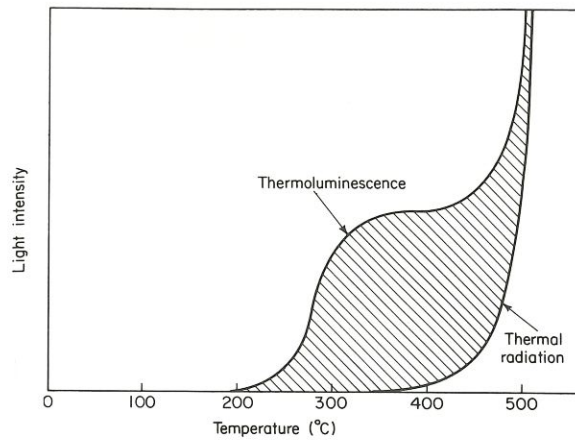


FIG. 7.3. Typical glow-curve from ancient pottery.

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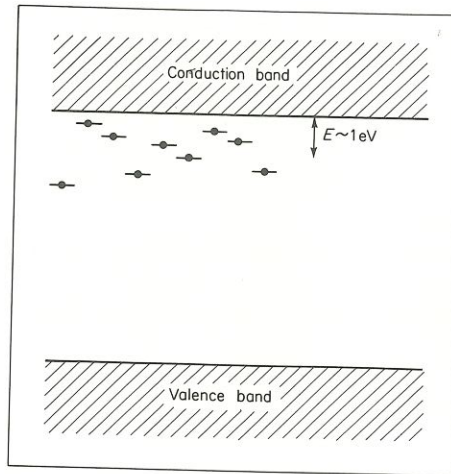


FIG. 7.4. Continuum of trap depths.

and Boltzmann's constant k equals $8.6 \times 10^{-5} \text{ eV}/^\circ\text{K}$. The lifetime, τ , of an electron in such a trap is

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So for $T_g = (273 + 250)^\circ\text{K}$, $\tau \sim 10$ years, whereas for $T_g = (273 + 350)^\circ\text{K}$, $\tau \sim 30,000$ years.

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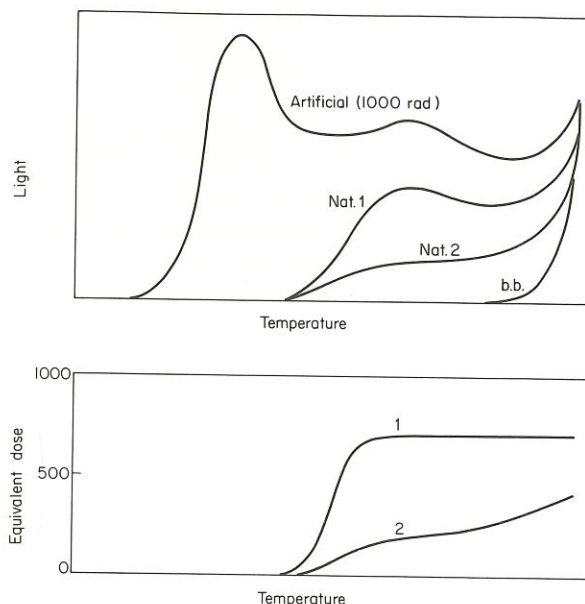


FIG. 7.5. The ordinate ratio test. (a) Glow-curves; (b) ratio of ordinates in (a) for case 1—good plateau, and case 2—no plateau.

Where this levels off to a plateau above a certain temperature—as in case 1—one is much more confident that decay effects are absent (above that certain temperature) than in case 2 where there is no plateau. Failure to obtain a plateau may also be due to the presence of spurious TL in the natural glow-curve because the glow-curve shape corresponding to spurious TL is different from that of radiation-induced TL. Whatever the cause, absence of a plateau is good reason for rejection of the pot from a dating programme.

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THE NATURAL RADIATION DOSE

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TABLE 7.I
Decay schemes for ^{238}U , ^{232}Th and ^{40}K .

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	$1\alpha, 2\beta$			
^{234}U	2.67×10^5 yr		$2\alpha, 2\beta$	
	2α			
^{226}Ra	1590 yr	$\text{ThX} (^{224}\text{Ra})$	3.64 days	
	1α		1α	
^{222}Rn	3.8 days	^{220}Tn	54.5 sec	
			1α	
		$\text{ThA} (^{216}\text{Po})$	0.16 sec	
			$2\alpha, 2\beta$	
^{206}Pb		^{208}Pb		
$Q = 46.7$ MeV		$Q = 39.8$ MeV		
$8\alpha, 6\beta$		$6\alpha, 4\beta$		

The diagram shows the decay of ^{40}K . It has two main decay paths: Electron Capture (EC) with a probability of 11% leading to ^{40}Ar , and Beta decay (β) with a probability of 89% leading to ^{40}Ca . A gamma ray (γ) with an energy of 1.46 MeV is emitted from the ^{40}Ar state.

TABLE 7.II

Dose-rates and alpha counts corresponding to given concentration of uranium, thorium and potassium.

Concentration	Uranium	Thorium	Potassium
	3.2 ppm	12.7 ppm	8.85%
Alpha count /1000 sec, thick sample, 15.2 cm ² , eff = 0.85	6.8	7.15	—
Total Dose	1 r/yr	1 r/yr	1 r/yr
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careful scrutiny of the above procedure is necessary, and certainly when circumstances are such that the effect of the alpha dose is negligible it is far from satisfactory. If the alpha dose does not have its full value, the beta and gamma doses become important, and because of their greater range this

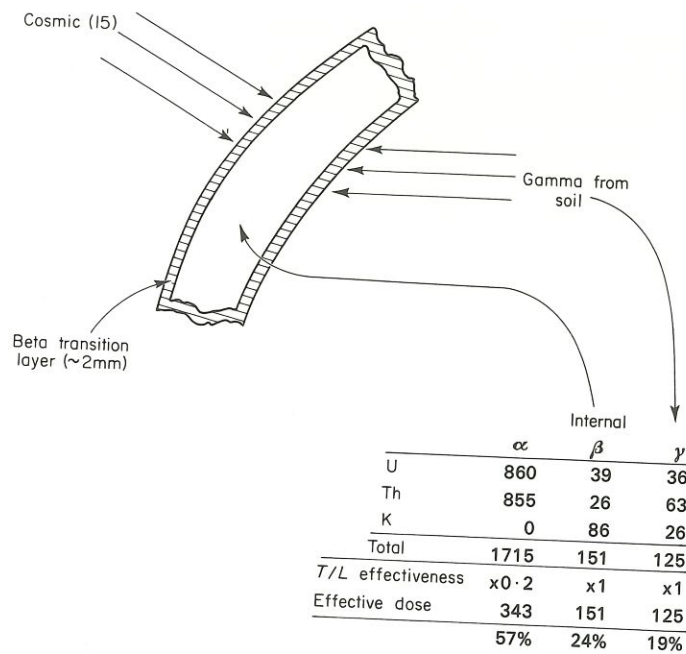


FIG. 7.6. Typical annual radiation dose for fragment of pot buried in soil, both having U 3 ppm; Th 12 ppm; K 1%.

means that the dose received by the pot is determined not only by its own radioactive contents but also by environmental radiation. The situation for typical pot is shown in Fig. 7.6. The major environmental contribution is the gamma dose from the burial soil but even for the beta dose there is a transition layer at the surface of the pot in which the dose is partially determined by the radioactive content of the pot and partially by that of the soil. The cosmic-ray contribution is estimated for burial 5 ft below the surface.

Because of the uncertainties inherent in converting alpha counts to effective dose it is desirable to use a more direct technique for evaluating the latter—such as dosimetry by thermoluminescence itself as described in a later paper in these proceedings (Paper 8.3).

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