DRICHTER

Ancient TL

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St. Louis, MO. 63130 U. S. A.

No. 3 Spring, 1978

"But about a little after sunset, whilst the twilight yet lasted, nay, this morning a pretty while after sun rising (but before I had been abroad in the more freely enlightened air of the chamber) I could upon a light affriction easily perceive the stone to shine." Sir Robert Boyle, in "Observations made this 27th of October, 1663 about Mr. Clayton's Diamond".

IN THIS ISSUE . . .

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The preparation of this document was supported in part by National Science Foundation Grant BNS 76-82645 and in part by subscription fees.

AMERICIUM-241 FOR ALPHA-IRRADIATIONS

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I. INTRODUCTION

The motivation for the present investigation was provided by the practical difficulties encountered in using a Cm-242 source for alpha-irradiation in fine-grain dating. These are: (a) short half-life (163d), (b) subsequent build up of Pu-238, which has a lower alpha-energy (5.5 MeV instead of 6.1 MeV), (c) source only available with a discreet and fragile window and (d) increasing difficulties in its commercial availability. It was therefore imperative to reconsider the available alpha sources, with the objective of finding a viable alternative. The obvious criterion for a suitable alpha-source is that it has a high alpha-energy, a long half-life and that, it does not give a significant dose rate from any other decay modes.

We shall first outline the difficulties (and possible complications) arising from the use of a Cm-242 source and then describe a feasibility study on the use of Am-241 (type AMM3, commercially available from R.C.C., Amersham, Bucks. England HP7 9LL). This is a robust rolled strip in which Am-241 is incorporated in a gold matrix ($\sim$ 1 μm thick,) backed with silver. The active surface has a protective coating of a gold-palladium alloy ($\sim$ 2 μm thick).

II. EXPERIMENTAL

A. Curium-242

Firstly, the short half-life of the source, necessitates a frequent recalibration of its strength. This is because the growth of Pu-238 does not seem to be accurately predictable theoretically (possibly due to contamination by Am-241). The construction of the curium sources is such that there is a protective 0.7 mg/cm^2 nickel foil across the active area; there is a further nickel window (0.1 mg/cm^2), separated by 1 mm of air. The lower alpha energy of the Pu-238 gives rise to serious complications and as will be shown the advantages gained by the higher alpha energy of Cm-242 (i.e. making alpha-irradiations in air possible) are thwarted by the Pu-238 build up. This was investigated in detail by looking at the alpha-spectrum under the actual experimental conditions (i.e. irradiations in air) used in fine-grain alpha irradiations. A surface-barrier detection system coupled to a 400 channel analyzer was utilized for the measurements. For most of the measurements reported here, the sources used were 10 microcuries or less. The fine-grain samples were simulated using aluminium foils of varying thickness (1-3.5 mg/cm^2). Figure 1 gives alpha-spectra (in air and source-detector distance $\sim$ 1 cm) from two Cm-242 sources, (a) with only small Pu-238 build up, (b) with 30% Pu-238 build up. Figure 2 gives a typical alpha-spectrum recorded from these sources with a 3.5 mg/cm^2

aluminium foil interposed. Figure 3 gives the variation in the normalized transmitted intensity with the thickness of the aluminium foil, for the Cm-242 source (b). This clearly indicates that for sample thicknesses $\gtrsim 1$ mg/cm^2, a finite fraction of the alpha-flux is stopped within the sample (and it is obviously due to Pu-238). Now this has a serious consequence in the dating application, because unless the light collection depth is less than the alpha track length (and this cannot be assumed to be so), the amount of alpha irradiated sample will be less than the amount of sample from which the natural TL is received and thus an erroneously low value for the alpha-sensitivity will be obtained, thus giving too old an age. Thus, the use of Cm-242 in air implies that Pu-238 build up may introduce errors in estimating the true TL alpha-sensitivity and hence the true archaeological dose. Although, in principle, this could be avoided by carrying out irradiation in vacuum (thus avoiding the energy loss in air), the presence of a discreet window eliminates this possibility in practice. The alternative of reducing the source-sample distance is not acceptable, since this results in an increased obliquity of the alpha-particles.

B. Americium-241

(i) Contaminating Radiation

Earlier experiments by Bowman (S.G.E. Bowman, Thesis, Oxford University 1976), resulted in anomalous discrepancies while using Am-241. This was attributed to the presence of some contaminating radiation, which could be stopped by using a 19 μm melinex foil. The source used by her was a weak reference source (type AMR 4, from R.C.C.) without a protective coating. It was thus expected that the usual Am-241 foil sources (type AMM 3) with an integral gold-palladium protective coating would be free of any contaminating radiation. This proved to be so; the TL per alpha measured using first an Am-241 source and then a recent Cm-242 source (negligible Pu-238) agreed to within 2%. Both the irradiations (Am and Cm) were carried out in vacuum (the nickel window of Cm-242 being deliberately punctured to allow this) and the alpha-flux was determined using the surface barrier system.

(ii) Gamma-contribution

We then considered the extent of a possible interference from the 60 keV gamma emission in Am-241 decay. This was investigated experimentally, using $CaSO_4$: Dy, by measuring the TL induced by the source (i.e. a combined dose from alphas and gammas as it is and by measuring the TL induced under exactly similar/with alpha-flux stopped with suitable absorbers ($\sim$ 30 μm aluminium foil). The typical gamma dose was found to be $\sim$ 0.04 rad/min as compared to equivalent beta dose of $\sim$ 130 rad/min from the alpha particles. Thus the gamma-dose works out to $\sim$ 0.03% of the effective alpha-dose and hence is unimportant in the present case. Theoretically also, the dose-rates were calculated using standard stopping powers and attenuation factors, and the dose rate ratio is calculated to be $\sim$0.03%. This is true for $CaSO_4$:Dy, with its alpha-efficiency a = 0.3. This indicates that even under the most unfavourable circumstances, a = 0.01 (the lowest value expected in routine dating), the dose-rate will be only $\sim$ 1%. An indirect proof of the negligible gamma-contribution is also obtained by the similarity in TL/alpha values mentioned earlier in B (i).

(iii) Minimum Source-sample Distance

The next step was to determine the best irradiation geometry, i.e. a compromise between the maximum possible flux and the minimum number of oblique alphas. For this alpha-spectra were recorded with and without a 3.5 mg/cm^2 aluminium foil (to simulate fine-grain sample) as a function of source-foil distance. A typical set of alpha-spectra obtained with and without the foil at a source-foil distance of 10 mm, through a 10 mm diameter window (diameter of a normal fine grain disc) are given in Fig. 4. Fig. 5 shows the total flux at various source-sample distances. It therefore appears that the 10 mm spacer is the optimum choice. This is the closest distance at which the flux, with and without the foil are the same, indicating that there is no attenuation of alpha-flux in the foil. At lower source-sample distances the fall off in the alpha-flux is due to the absorption of oblique alphas within the foil.

(iv) a-value results

For the basic calibration of sources, the procedure originated by Zimmerman (Radiation Effects 14, 81, 1972) and later refined by Aitken and Bowman (Archaeometry 17, 132, 1975) was used. For convenience, particularly if a suitable detection system is not available, a-values were definitively determined for three phosphors (viz: CaF_2: nat; CaF_2: Dy and $CaSO_4$: Dy). Thus, now, given a calibrated beta-source, the calibration of an alpha-source can be easily obtained. These a-values were obtained using both a weak Am-241 source and a weak Cm-242 source, and the alpha-irradiations were carried out in vacuum. The source strengths were measured using the alpha-detector and the a-values were determined by comparing the TL response for alpha-particles to that with beta-particles using a calibrated ^{90}Sr source and then using eqn. 10 of Aitken and Bowman. These values are tabulated in Table 1. These values do indeed indicate good agreement between Cm and Am.

Table 1

S.No.	Phosphor	a-values Am	a-values Cm	Recommended values
1.	CaF_2: nat	.092	.096	.092
2.	$CaSO_4$: Dy	.292	.293	.292
3.	CaF_2: Dy	.505	.491	.500

Note:

(1) Typical errors in these values are: $\pm$ 3%.

(2) Both $CaSO_4$: Dy and CaF_2: Dy (TLD 200) were from Harshaw and CaF_2: nat, was from MBLE, type Super S.

(3) We suspect that there can be some batch to batch variation in a-values in CaF_2:Dy and in CaF_2:nat.

(v) The six-seater irradiator

A disadvantage in using Am-241 is that the commercially available source strengths are quite low and hence longer irradiation times are needed. With this in mind a six seater automatic alpha-irradiator was designed, Fig. 6. Here six alpha-foil sources (type AMM3 with an active area of 12.5 mm x 12.5 mm, but restricted by a circular aperture of diameter 12 mm) are used. By using different source-sample distances six discs can be irradiated simultaneously to different dose levels if required; for a source-sample distance of 10 mm the strength is about 0.33 micron^{-2} minute^{-1}, and for 40 mm it is lower by a factor of 14. Cross-talk from alpha particles is avoided by means of a plastic tube around each sample mount. Cross-talk from gamma radiation amounts to 0.004% of the alpha contribution for a = 0.1 and a source-sample distance of 40 mm; it is less for closer positions. In order to avoid a significant gamma dose-rate with the shutter closed, lead discs of thickness 1.6 mm were fitted into the shutter so as to be under the sources when the shutter is in the closed position; as a result the shutter-closed TL is only 0.001% of the shutter-open alpha TL for a = 0.1 and a source-sample distance of 10 mm; for 40 mm it is 0.004%. Without the lead discs the shutter-closed dose-rate was nearly an order of magnitude higher; this would have entailed the inconvenience of not being able to leave samples in position overnight after a short alpha irradiation. The other features of this irradiator are detailed in Fig. 6. It is designed so that it can be fitted on the top of a regular glow oven thus obviating the need for a separate vacuum facility.

III. A PRACTICAL ALPHA-SPECTRUM CHECK

On the basis of the simplifying assumptions that the TL per unit track length is independent of alpha energy one would expect that interposition of a thin aluminium foil between source and sample would cause no attenuation of the TL unless the thickness of the sample was sufficient to prevent complete penetration through the sample. This can be used to test whether or not a given source has a good alpha spectrum. For a Cm-242 source for which the Pu-238 contribution was about 7%, the TL observed with a 3.5 mg/cm^2 foil of Al interposed was 0.85 times the TL with no foil. This was using a sparsely-covered disc of ultrafine grains of CaF_2: Dy (corresponding to a settling time in acetone in excess of 20 minutes). On the other hand the corresponding value for a Cm-242 source, with a similar amount of Pu-238 build-up, but with a badly corroded window was only 0.4. Both these observations were in air and are for a source-sample distance of 9 mm. For the Am-241 source currently under discussion the value was 0.80, in vacuum and for a source-sample distance of 40 mm.

Fig. 7 shows the effect of different foil thicknesses using an ultrafine disc of $CaSO_4$: Dy for source-sample distances of 10 mm and 40 mm. It will be seen that the attenuation for the 3.5 mg/cm^2 foil is substantially more severe in the case of $CaSO_4$:Dy than in the case of CaF_2:Dy. It is presumed that this reflects that the simplifying assumption of a constant TL per unit track length, with alpha energy is not rigorously valid. Bowman (Ph.D. Thesis, Oxford University 1976) has in fact reported in detail the variation in TL per unit track length with alpha energy for $CaSO_4$:Dy and in the light of her results, the observed fall off is not altogether unexpected.

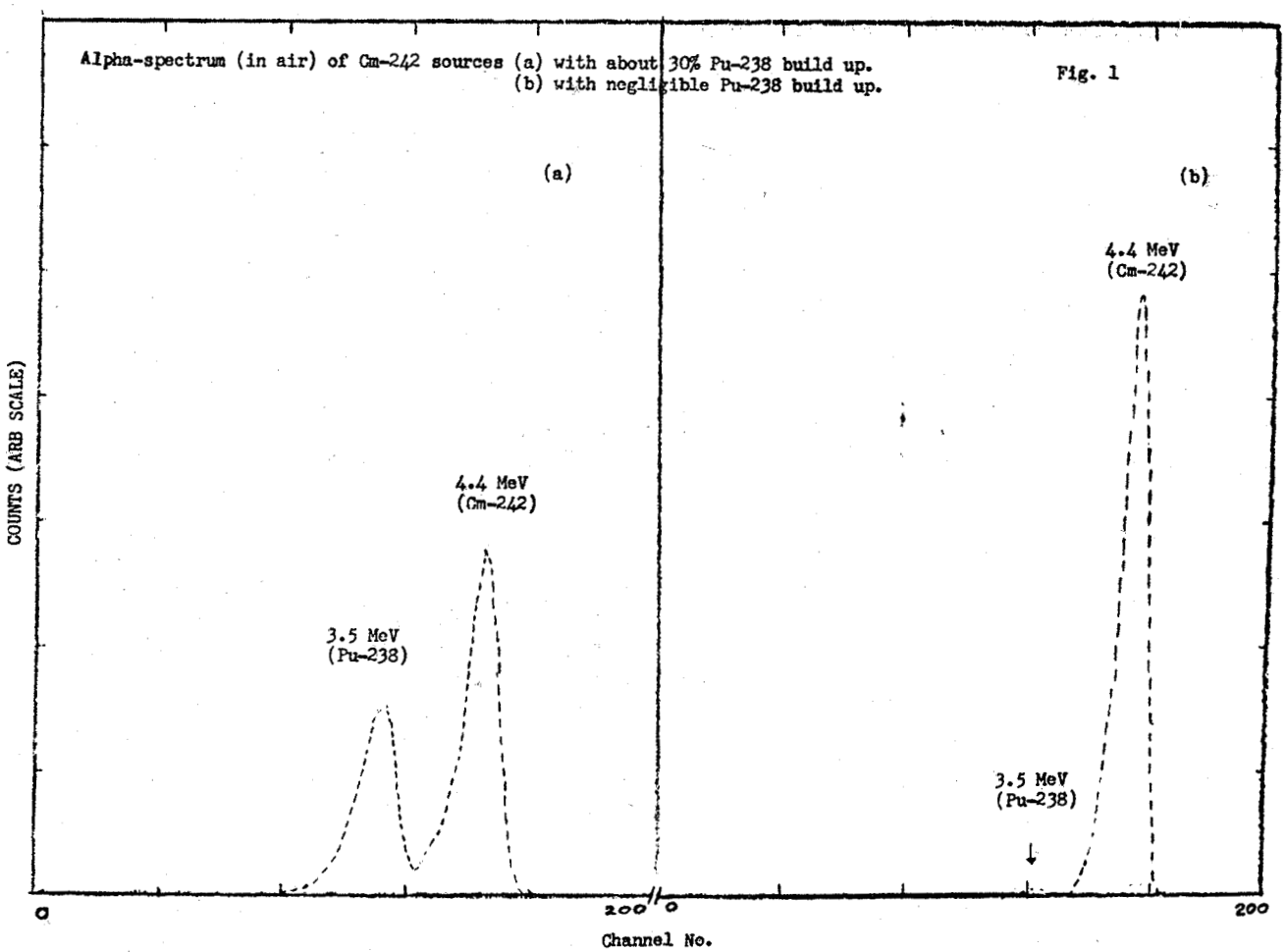
IV. DISCUSSION

It thus appears that Am-241 when used in vacuum provides a viable alternative to Cm-242. Its longer half-life is economically advantageous as also is its lower initial cost (currently 6 sources can be purchased for £75). Its robustness is in strong contrast to the fragility of the nickel window incorporated in the Cm-242 source and so the changeover eliminates a substantial hazard.

It has been suggested (D. W. Zimmerman, private communication) that the penetration test using a 3.5 mg/cm^2 aluminium foil is unnecessarily rigorous. This is on the basis that effective thickness of a fine-grain sample is never more than the diameter of an 8-micron grain, viz 2.2 mg/cm^2. Nevertheless it seems desirable to demand full penetration of a 3.5 mg/cm^2 foil; this means that for a thickness of 2.2 mg/cm^2 the particles will emerge with an energy of around 1.5 MeV and thereby one avoids using them in the lower energy region where we are most doubtful about the constancy of TL per unit track length.

Acknowledgements

We thank Mr. D. Seeley for all the fabrication work, Mrs J. Huxtable for sample preparation and A.K.S. gratefully acknowledges Ford Foundation (New Delhi, India) for a fellowship.



Alpha spectrum (in air) of Cm-242 sources with a 3.5 mg/cm^2 foil interposed
(1) for source (a)
(2) for source (b)

Fig.2

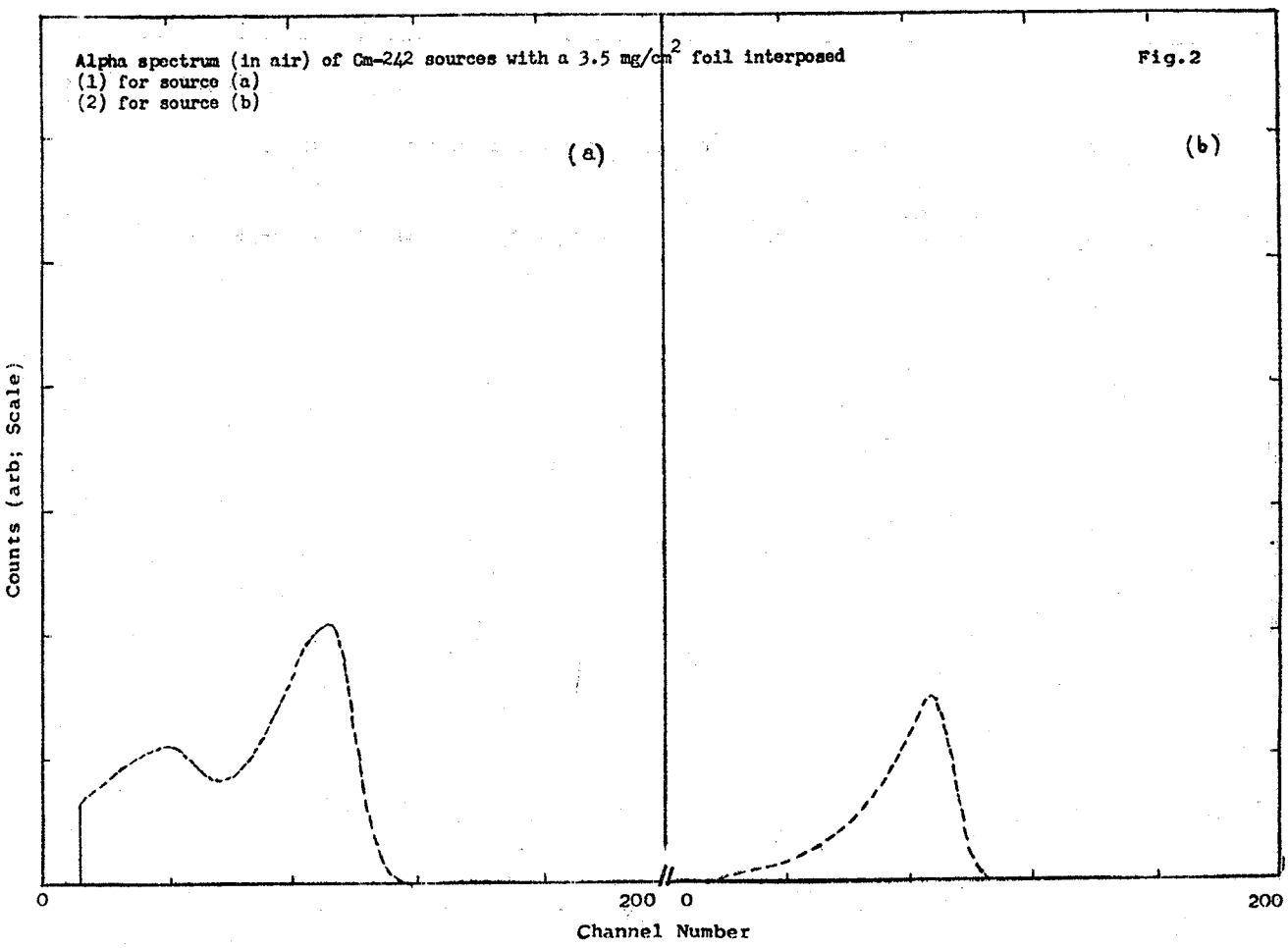


Fig.3

Variation in the transmitted alpha flux with the absorber thickness (source a):
(1) Source-absorber separation of 7 mm (air).
(2) Source-absorber separation of 11 mm (air).

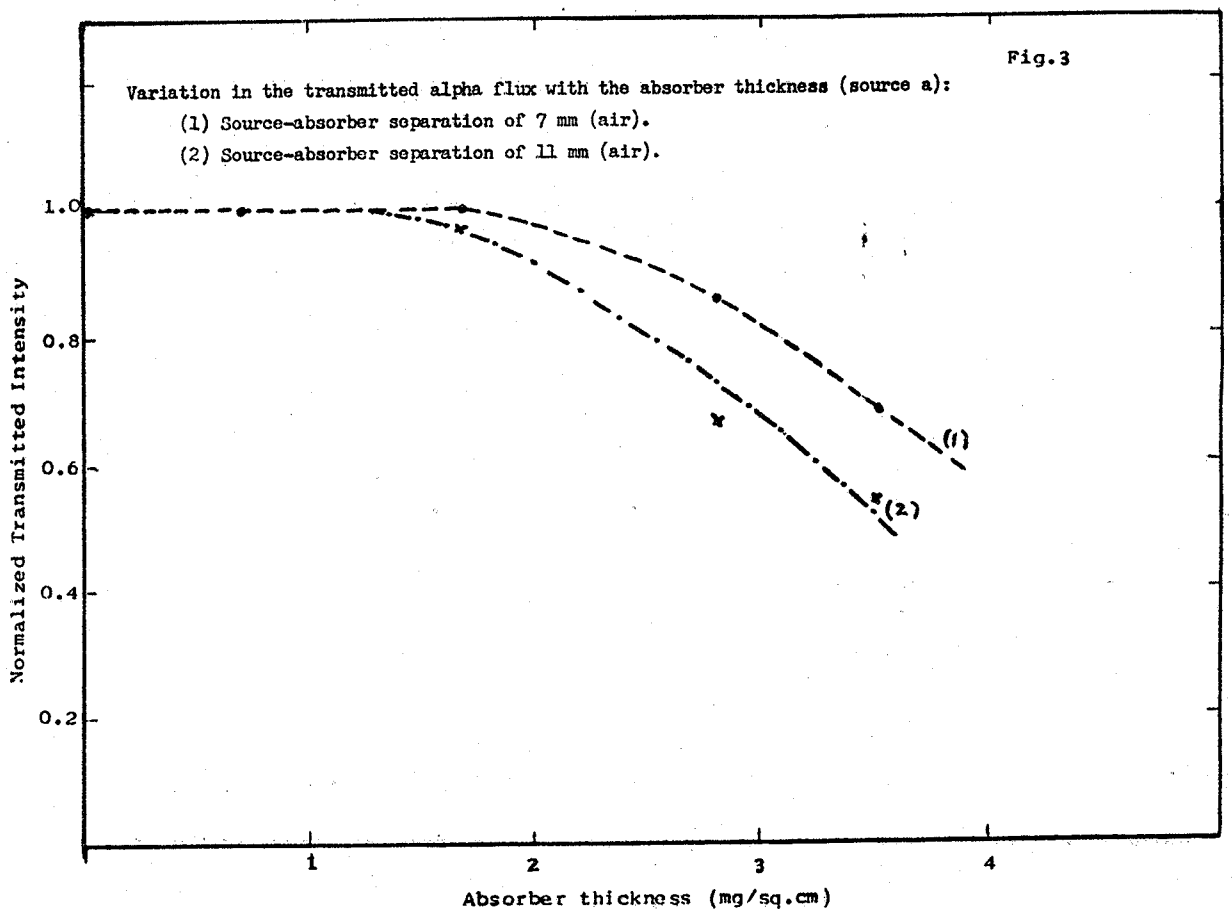


Fig.4

Alpha-spectrum of Americium-241 in vacuum at a source foil distance of 10 mm.

(1) without any foil
(2) with a 3.5 mg/cm^2 Al foil.

The energies shown are for a 10 microcurie source (AMM 7). For AMM 3 the energy in case (1) is 4.2 MeV due to a thicker protective coating.

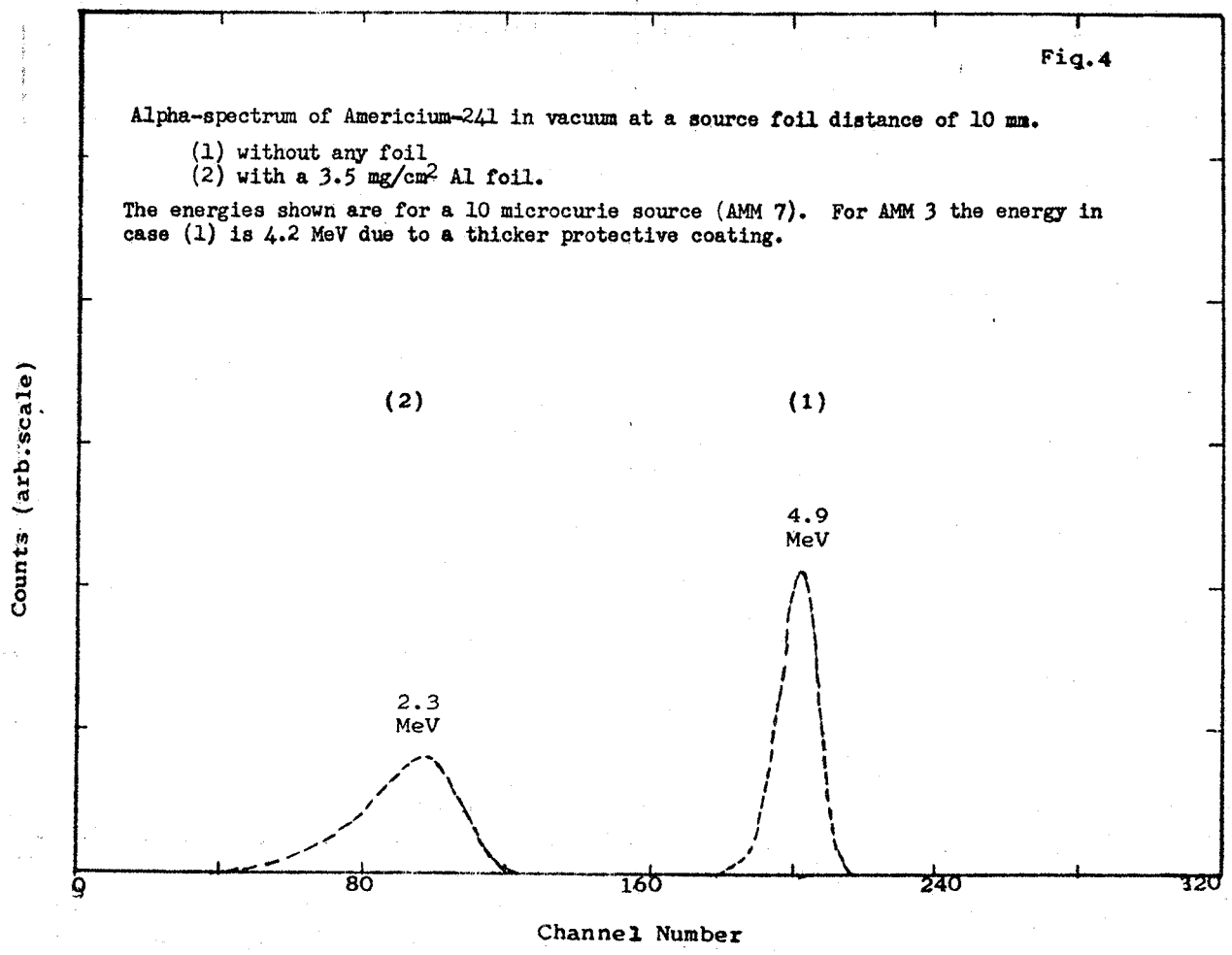


Fig.5

Variation in the alpha-flux from Am-241 source in vacuum with different source-detector distances

(a) without foil
(b) with a 3.5 mg/cm^2 Al foil.

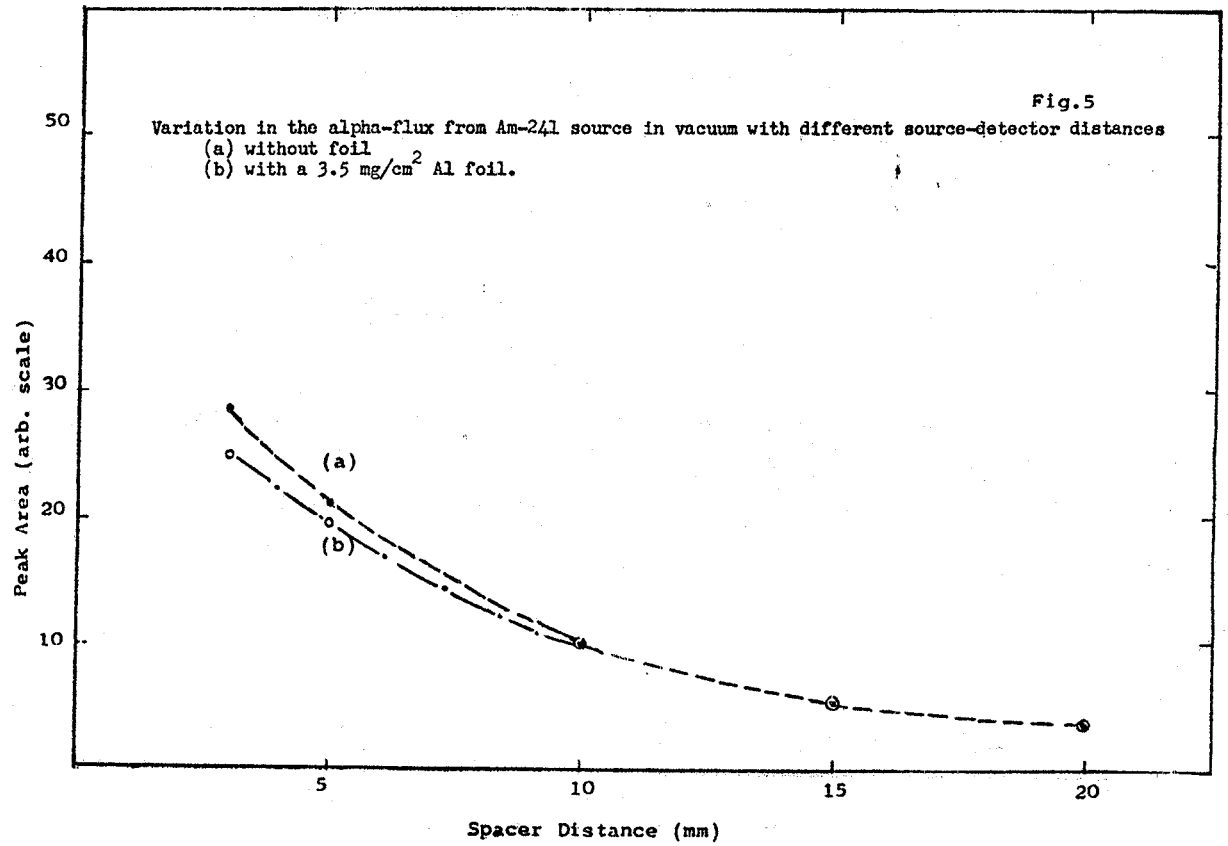
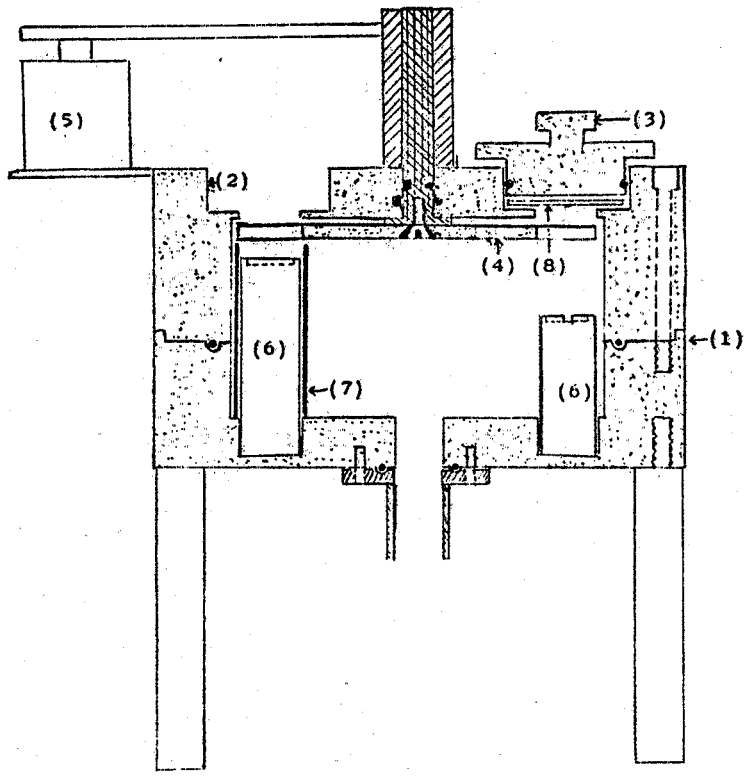


Fig. 6. The automatic six seater irradiator.



(1) Aluminium housing. Has six seats (2) for housing six foil sources.

(2) Housing for the sources. The source (8) is kept with its active surface facing downwards. A rubber bungy kept between the source and the holder stub (3).

(3) Aluminium holder with o-rings for the vacuum.

(4) Aluminium disc with six holes appropriate to the source size. Operated by the magnetic latch (5).

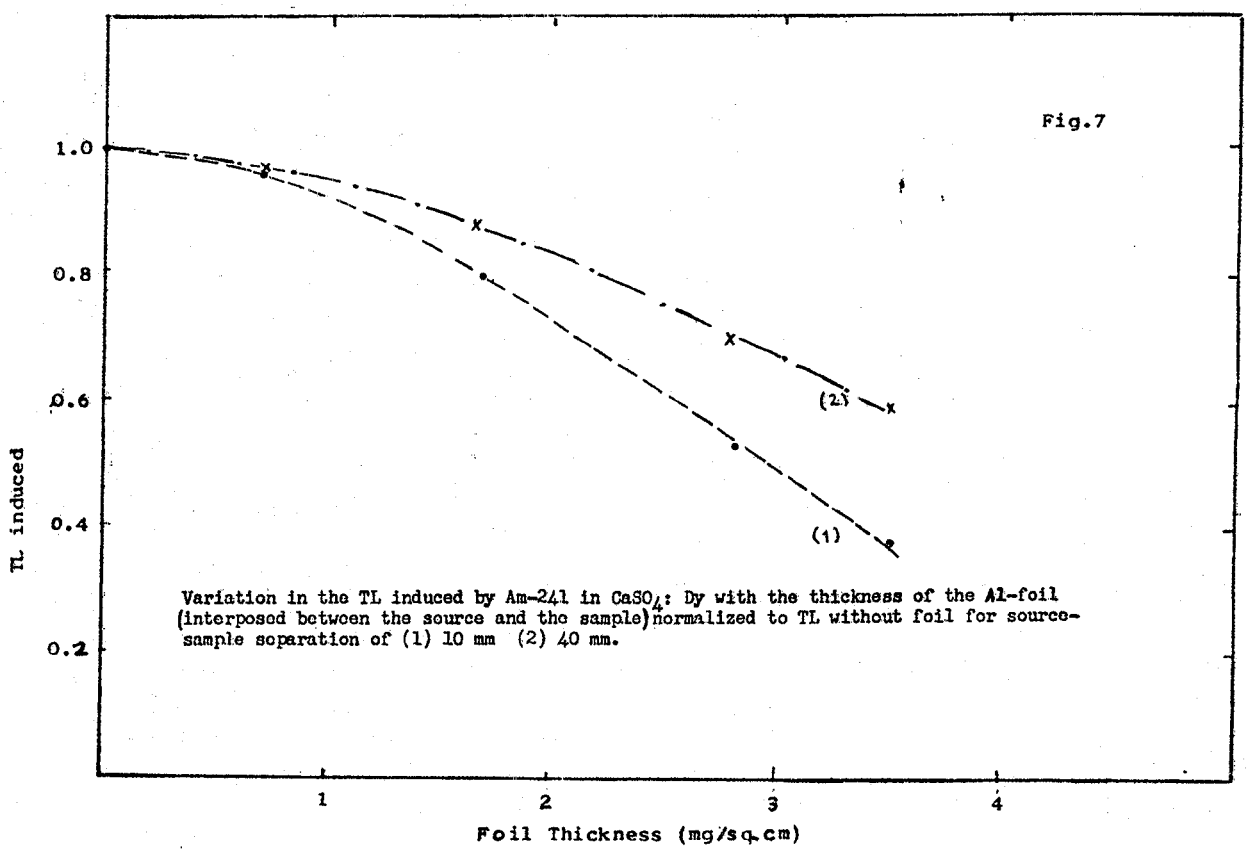
(5) An electromagnetic switch operated with a timer arrangement. The brass rod is latched on to it and this starts the irradiation by bringing holes in (4) in front of sources.

(6) Sample mounts and seat.

(7) Plastic tubes.

The other features of the system are self-explanatory.

Fig.7



Variation in the TL induced by Am-241 in $CaSO_4$: Dy with the thickness of the Al-foil (interposed between the source and the sample) normalized to TL without foil for source-sample soperation of (1) 10 mm (2) 40 mm.

ATTEMPTS TO CIRCUMVENT ANOMALOUS FADING

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Anomalous fading (the instability of the stored TL at high glow-curve temperature) prevents the reliable dating of most minerals other than quartz. We have attempted to circumvent anomalous fading using three different approaches; spectral resolution, thermal annealing, and slow glow-curve heating rate. These experiments were based on the hypothesis that the stored high-temperature TL immediately after irradiation consists of two parts; an "unstable" part which will anomalously fade, and a "stable" part which is the same part as in the natural TL. Unfortunately, all three approaches were unsuccessful.

In the first experiment the emission spectra of various minerals exhibiting anomalous fading were measured immediately after beta irradiation and, using a second aliquot, one day later. If the "stable" and "unstable" TL were emitted at different wavelengths, anomalous fading could be avoided, or at least reduced, by doing the TL measurements using a filter that would transmit only the wavelengths that faded the least. The measurements were made with an optical multichannel analyzer (OMA) attached to the TL oven instead of the regular photomultiplier tube. The OMA consists of a polychromator and a 500 element vidicon tube with an image intensifier. The polychromator disperses the incident light across the face of the vidicon tube so that each element receives a different range of wavelengths between 400 and 600 nm. The signal into each element is integrated over a present length of time (> 30 msec), then read out and stored in a computer memory. In this experiment, four materials were measured (zircon, apatite, orthoclase, and pumice), the TL spectra being integrated between 300 and 450°C in the glow curve. The anomalous fading was easily measurable in all four materials, being from 35 to 50% in one day, but was found to occur to the same degree at all wavelengths in all four materials. Fig. 1 shows the results for the feldspar sample.

In the second experiment, we tried to accelerate the fading of the "unstable" TL in the pumice by holding samples at elevated temperatures after irradiation in hopes of having rapid fading to a stable level. Fig. 2 shows the isothermal decay curves. Although the fading rate is increased at high storage temperatures, at 120° a stable level has still not been reached after 80 days. At 160°C it looks as if a stable level might be reached after about 30 days. Unfortunately, the natural TL at 450°C in the glow curve decreased by 20% after one week at 160°C. Thus, this type of thermal treatment seems to be unable to separate "unstable" and "stable" components.

The third experiment determined the fading using a much slower heating rate on the TL oven (0.18°C/sec instead of our usual 7°C/sec). The motivation for this approach was two-fold: 1) a reference (Shulman, 1967) to unpublished data by Ginther that "slow-glows" eliminated anomalous fading in the dosimetry phosphor CaF_2:Mn (in recent conversation, Ginther could not confirm this observation); and 2) the "slow-glow" will better separate individual glow peaks, and in this respect make the annealing characteristics simpler. We found that the "slow-glow" nicely separates the peaks for LiF (Fig. 3), but produces essentially no separation for CaF_2:Mn (Fig.4), apatite, zircon, albite, orthoclase, and pumice. We compared the fading of the pumice and apatite high-temperature TL using the normal and slow heating rates and the results are shown in Table 1. The "slow-glows" produced no substantial reduction in the fading of these samples.

The failure of all three experiments contradicts the hypothesis that the stored high temperature TL consists of separate "stable" and "unstable" components.

REFERENCE

Schulman, J. H., 1967, Survey of Luminescence Dosimetry, in Luminescence Dosimetry, ed. F. H. Attix, CONF - 650637, NBS, Springfield, Va., 28.

FIG. 1

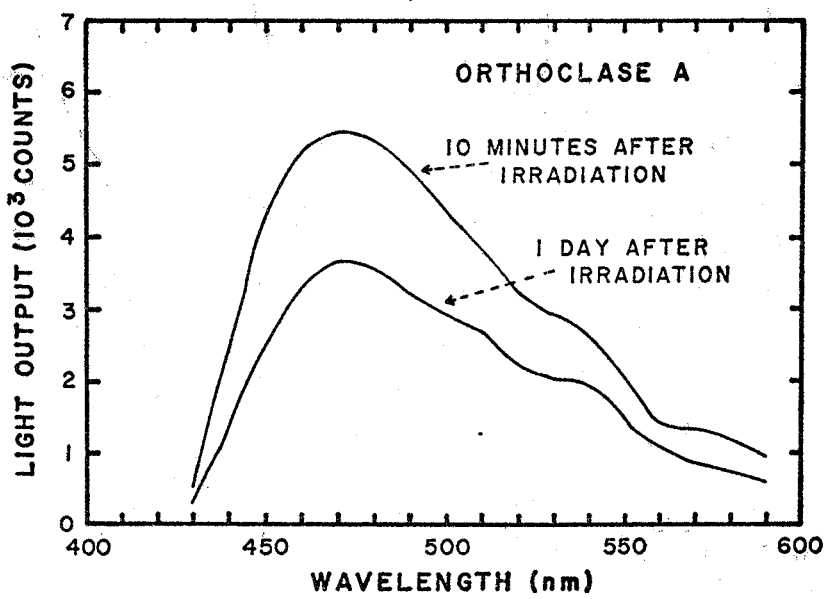

FIG. 2

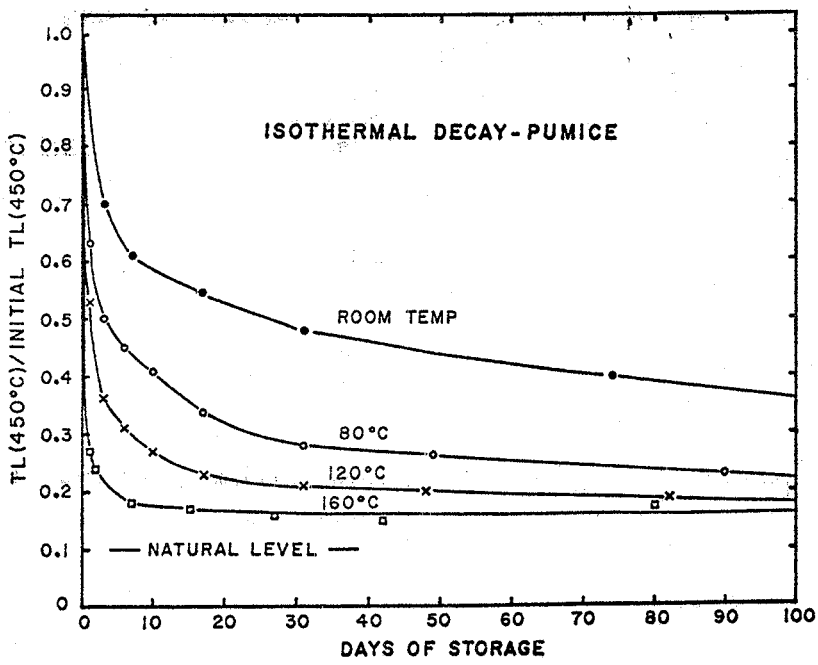

FIG. 3

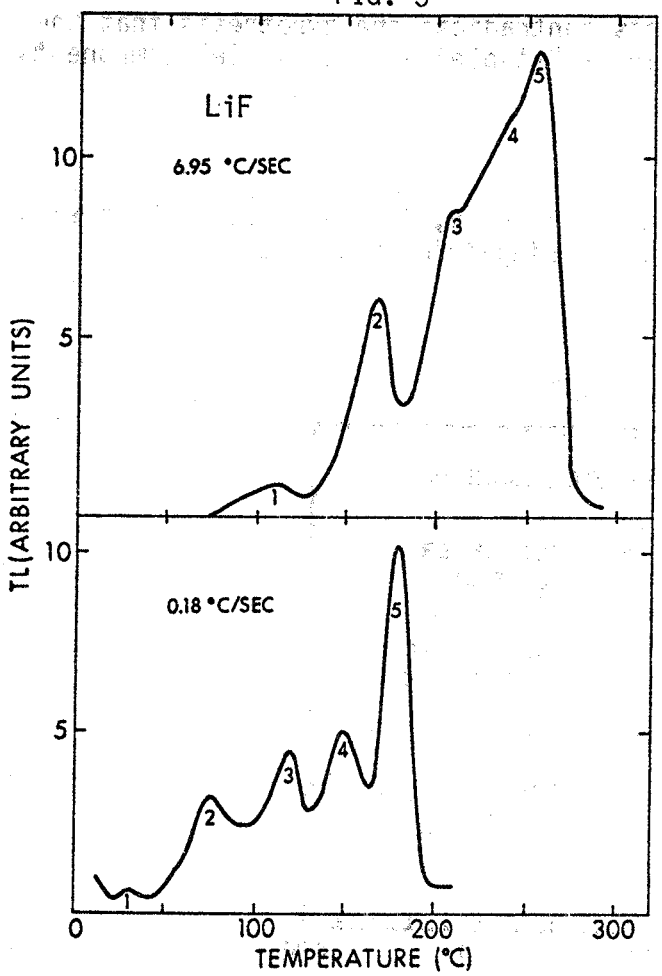

FIG. 4

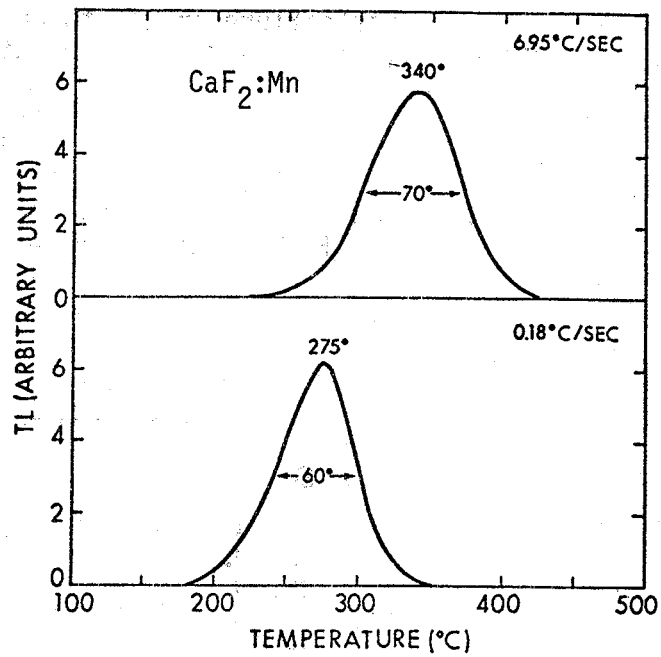

TABLE 1

EFFECT OF GLOW CURVE HEATING RATE ON ANOMALOUS FADING

Sample	Glow Curve Heating Rate (°C/sec)	TL Lost After 1 Day At Room Temperature* (%)
Apatite	7.35	41
	0.18	36
Pumice	7.35	24
	0.18	22

*Expressed as percent of the TL measured 30 minutes after irradiation.