It seems our source has run out of alphas! The odd behaviour of some americium-241 cup sources

Ralph Whitcher, Robert D. Page and Peter R. Cole

ABSTRACT The alpha emission rate from some older americium-241 school cup sources, detected by a GM tube, appears much lower than when the sources were new. Strangely, the sources perform as expected when used with spark counters and ionisation chambers. This apparent reduction of emission rate adversely affects the demonstration of blocking alpha radiation with paper. The research jointly by the University of Liverpool and CLEAPSS revealed an increase in alpha energy straggling caused by a net migration of americium deeper into the source foil. The article gives advice to schools that have such a source behaving oddly.

A useful and popular physics demonstration is the blocking of alpha radiation using a sheet of paper, an easy practical to carry out that shows the low penetration of alpha particles. The alpha source is placed close to a detector and, when a sheet of paper is placed between them, the detection rate drops greatly. Figure 1 shows a set-up with a source and Geiger–Müller (GM) detector (also called a GM tube).

In the past few years, CLEAPSS and SSERC have received reports of older 185 kBq americium-241 sources that are behaving oddly in this paper-block demonstration. With the source positioned so that there is no more than a 10 mm gap between the front of the cup source and the metal end of the GM detector, the count rate appears much lower compared with when the source was new. Placing a sheet of paper between the source and GM detector no longer produces the considerable fall in count rate. The emissions seem to be predominantly gamma radiation. As one school put it, '*It seems our source has run out of alphas!*' The schools that reported this were confident that the sources, when new, had behaved as expected. Bizarrely, the sources were still behaving as expected when used with a detector such as a spark counter or an open-window ionisation chamber, demonstrating the blocking of alpha clearly. The cause of this odd behaviour was not apparent; the obvious suspects of incorrect apparatus or set-up, foil surface contamination, or faulty equipment were quickly eliminated by investigations.

The University of Liverpool and CLEAPSS collaborated to research the cause of this

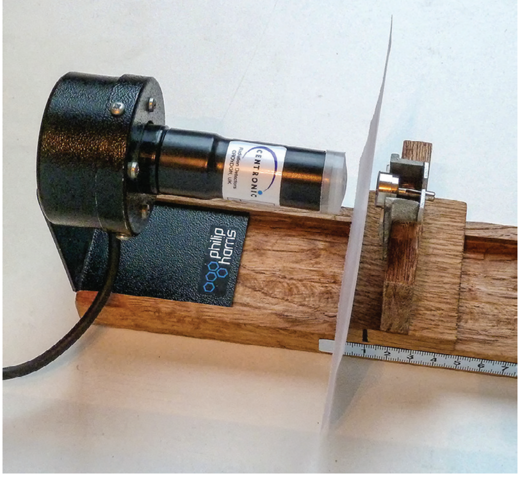

Figure1 Typical apparatus set-up for the alpha particle paper-block demonstration

CLEAPSS is an advisory service providing support in science and technology for a consortium of local authorities and their schools for England, Wales and Northern Ireland. SSERC is a local authority shared service providing support across all Scottish education authorities. Both organisations also support independent schools and FE colleges.

performance degradation. The aim was to determine the causes and assess whether this affected the useful and safe service life of the sources. The full technical report was published in the *Journal of Radiological Protection* (Whitcher, Page and Cole, 2014). The following gives a summary of the research and is aimed at helping school teachers and technicians who have come across this problem by providing practical advice on what can be done about it.

School alpha sealed-source design

The most common design of radioactive sealed source in UK school science is the cup style. It has changed little since it first appeared in the 1960s. The design follows a specification approved by the Department of Education and Science (DES) at that time. The radioactive material is sealed in a metal foil disc, about 10 mm diameter and 0.2 mm thick, mounted inside a plated brass cup and held in place with a circlip or adhesive. The foil is protected by a wire grill on the front of the cup, and the rear of the cup has a 4 mm diameter spigot allowing it to be handled with a tool such as long forceps. The cup-style sources are housed in a lead-lined wooden box (Figure 2). The sources are typically 185 kBq (5 μCi).

Americium-241 is the radionuclide used in the foil of these alpha-emitting cup-style sources (plutonium-239 and radium-226 have been used in the past). Americium-241 emits alpha particles with high-enough energy to be detected by thin-window GM detectors such as the ZP1481 used in schools. It has a useful long half-life of 432.6 years, so the percentage loss of americium-241 through radioactive decay will be negligible even for relatively aged sources. The decay product, neptunium-237, has a half-life of 2.1×10^6 years so the decay chain products in the source during its service life are also negligible. But, in another aspect, americium-241 is not ideal for demonstrating the characteristics of alpha radiation: it also emits significant gamma and X-ray radiation. Consequently, when a paper sheet is placed between the source and the GM detector, the count rate does not reduce to background, and this needs to be explained in the demonstration. Nonetheless, end-window GM detectors generally have a low gamma and X-ray detection efficiency, about 1%, so the count rate does fall enough to show convincingly that paper blocks alpha radiation.

Originally, the foils were manufactured by the Radiochemical Centre, Amersham, but the manufacturing has changed hands many times and is now part of the Eckert and Ziegler Group. The basic structure of the metal foil remains unchanged since it was first introduced (Figure 3). It comprises several layers, with the active layer being a mix of gold and a small quantity of americium oxide sintered into a metal 'matrix'. This active layer is sandwiched between a gold or gold-palladium alloy face and a thicker silver substrate. From information supplied by the UK distributor, High Technology Sources Ltd (HTSL), the active layer is ~1 μm thick and the face thickness is ~2 μm. The thin face layer allows the penetration by alpha radiation while retaining the active layer beneath.

Figure 2 A cup-style americium-241 source and its storage box

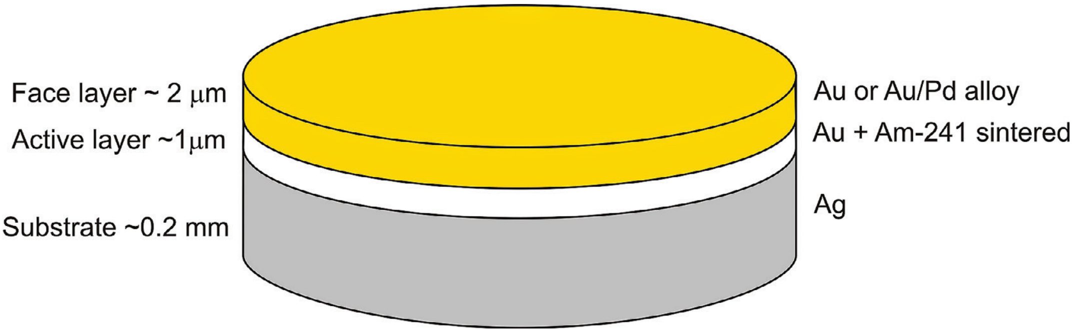

Figure 3 The foil layers in an americium-241 alpha cup-style source; not to scale – the layer thicknesses have been exaggerated to show the construction

The discs are stamped from a foil strip – the stamping effectively cold-welds the edge of the disc, completing the sealing of the americium-241. The manufacturer's data state the activity of the disc as 185 kBq ± 20% but investigations by Lucas (1966) with radium-226 foil sources suggested that in earlier decades foils showed a 50% variation from the nominal activity of 185 kBq. The layer thicknesses may also have varied more in earlier decades: a National Radiological Protection Board (NRPB) report (Williams, 1974) gave the thicknesses differently, with the gold-americium layer being between 1 and 2 µm thick and the source face layer being up to 3 µm thick. There is evidence of differences in the construction of the foils: the Amersham packing note (1991) shows a diagram where there is a gold backing layer between the silver back and the active gold matrix, but the NRPB report (Williams, 1974) does not mention this.

The alpha radiation will lose energy in penetrating the foil layers and there will be a spread in the energy of the alpha particles as they emerge from the face. This is termed energy straggling and a measure of straggle is the full width at half maximum (FWHM), which is the energy difference between either side of the peak at half the spectral peak value (Figure 4). As shown by Comfort *et al*. (1966) and others, the spectral peak alpha energy reduces and the energy straggling increases as the thickness of the material through which the alpha particles travel increases.

In Figure 4, source B is designed for alpha spectroscopy; the americium-241 is a very thin layer on the surface (and consequently easily damaged). The three principal alpha energies

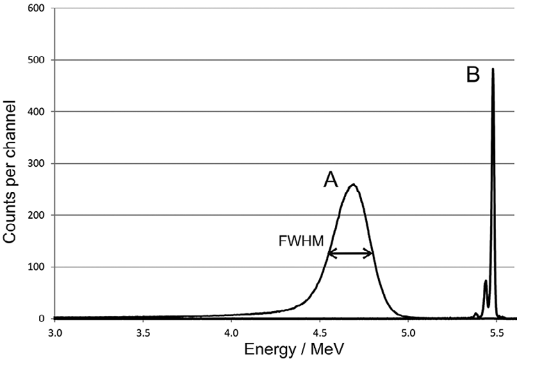

Figure 4 A graph comparing the energy spectrum of alpha radiations from two americium-241 sources, showing alpha energy straggle; FWHM = full width at half maximum

of americium-241 can be observed: 5.486 MeV, 5.442 MeV and 5.388 MeV. Source A is a foil from a new americium-241 cup source, with the active layer being beneath a gold-palladium layer about 2 µm thick. The peak alpha energy is reduced and there is a larger spread of energy. From information supplied by HTSL, the alpha foils for the cup sources are designed to give a spectral peak alpha emission of 4.5 MeV within a tolerance of 10%, and with energy straggling FWHM not exceeding 0.5 MeV. HTSL also believed this was the specification of americium-241 foils made in previous decades.

The sources used in the investigation

Schools were contacted to obtain sources that had been reported showing the odd behaviour, where the school was confident that the source had performed satisfactorily in the past. A school was also contacted that had relatively

Table1 Americium-241 185 kBq cup sources used in the initial study

Source identifier	Date purchased	Front face (from surface colour)	Reported showing reduced count rate	Original supplier
S1	1990	Gold	Yes	Philip Harris
S2	~1980	Gold-palladium	Yes	Griffin & George
S3	1987	Gold-palladium	No	Griffin & George
S4	~1980	Gold-palladium	No	Philip Harris
S5	2006	Gold-palladium	No	Philip Harris
S6	2006	Gold-palladium	No	Philip Harris
S7	2013	Gold-palladium	No	HTSL

new americium-241 cup sources. CLEAPSS holds cup sources for training purposes and one americium-241 source was found to show a similar low alpha count rate using a GM detector. From these, seven sources were selected (Table 1). S1 and S2 were reported showing the reduced alpha detection rate while S3, S4, S5 and S6 were not reported as showing it. S5 and S6 were newer, having been purchased in 2006. S7 was a new source supplied by HTSL. All were 185 kBq cup-style sources.

The sources were leak-tested by a dry-wipe test following the procedure in L93 (science.cleapss.org.uk/Resource-Info/L093-Managing-Ionising-Radiations-and-Radioactive-Substances-in-Schools-and-Colleges.aspx). No contamination was detected. The faces of the foils were remotely examined using a 12 megapixel digital camera with a macro lens. The faces were highly reflective with no visual evidence of surface degradation or any coating (Figure 5).

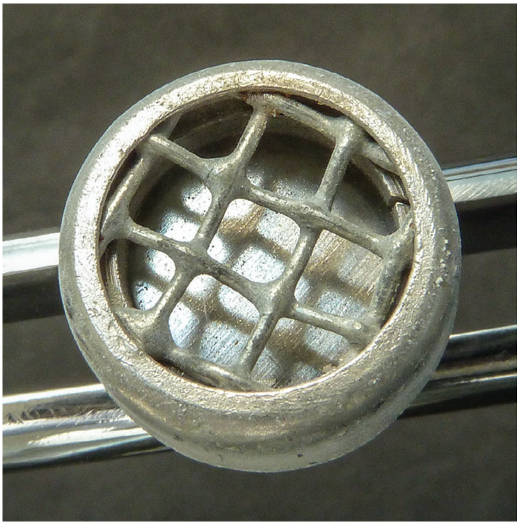

Figure 5 The front of source S3 (which is being held by forceps); the foil is held in place by a circlip; on some sources there are flat marks on the cup edge where the cap was pressed on in manufacture

The foil face on each test source appeared to be in good condition with no observable surface defects. In source S1, the foil was not quite concentric with the brass cup and a foil edge could just be observed on one side.

The investigation methods and findings

The first step was to reproduce the results from the sources reported as showing a reduced alpha count rate, including the low reduction in count rate using a paper block. For quantitative comparison of the paper-block demonstration results, a set of equipment was selected that was representative of what might be found in schools. The equipment chosen was a Philip Harris Digicounter, a Philip Harris GM detector holder and a Centronic ZP1481 GM detector manufactured in 2007 that was in good condition.

There are shortcomings of equipment to keep in mind. When a GM tube detects a radioactive emission, for a short time after it cannot detect any further emissions – this is termed dead time. It is about 120 μs for a ZP1481. What is not so well known is that the detector holder and counter circuit can also add considerably to the dead time by the way in which the pulse from the GM detector is processed, and this can worsen the underdetection of emissions in high radiation fields. The combined dead time of the ZP1481, holder and Philip Harris Digicounter that were used in this investigation was roughly 300 μs, although it varied with detection rate, becoming longer at higher count rates.

The sources were each set coaxially to the ZP1481 end-window GM detector (which was connected to the Philip Harris Digicounter) using a radioactivity bench, and the count recorded for a 100 s period at various distances of the detector window to the foil front face, first with paper between the source and detector and then without. The GM detector was operated at 450 V; its protective plastic grill cap was removed for maximum efficiency. (Note that in schools it is common, and sensible, to leave the cap on to protect the GM detector.) The count rate was corrected for background radiation but not corrected for detector/counter dead time, which reflects how the paper-block demonstration is usually done.

Figure 6 shows the data that confirmed the problem. At a 10 mm distance between the foil and the GM detector window (which equates to about 5 mm separation between the edge of the cup and the metal end of the GM detector), the relatively new source S6 gave a 32-fold reduction in count rate without and with the paper block. But with source S2, it was 11.8 and S1 was even worse at 3.8.

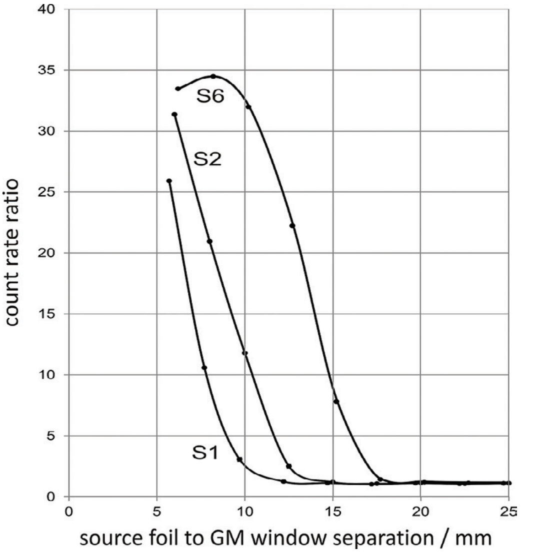

Figure 6 Each graph (for sources S1, S2 and the newer S6) shows how the ratio of count rate with and without paper varies as the source is moved further from the detector; the paper thickness was 0.1 mm

Gamma spectrometry

A gamma spectral analysis was taken of the sources to look for any anomalies in radionuclide composition of the sources with the odd behaviour. The energy spectrum was measured with a calibrated high-purity germanium gamma detector. No anomalies were found.

Alpha spectrometry

A possible cause of the sources S1 and S2 producing the lower alpha count rates is energy attenuation of the alpha emissions. To investigate this, the alpha energy spectrum of the sources was analysed using a calibrated high-resolution alpha spectrometer. The energy spectra from the cup sources are shown in Figure 7.

This revealed why sources S1 and S2 were exhibiting the odd behaviour. The larger energy straggling and lower spectral peak energy could clearly be seen for sources S1 and S2. (Just discernible was a small peak at the higher end of S4 graph, which indicated trace levels of americium-241 at or very near the foil surface.) Figure 8 shows the spectral peak energy and associated FWHM of each source in relation to the indicative source value of

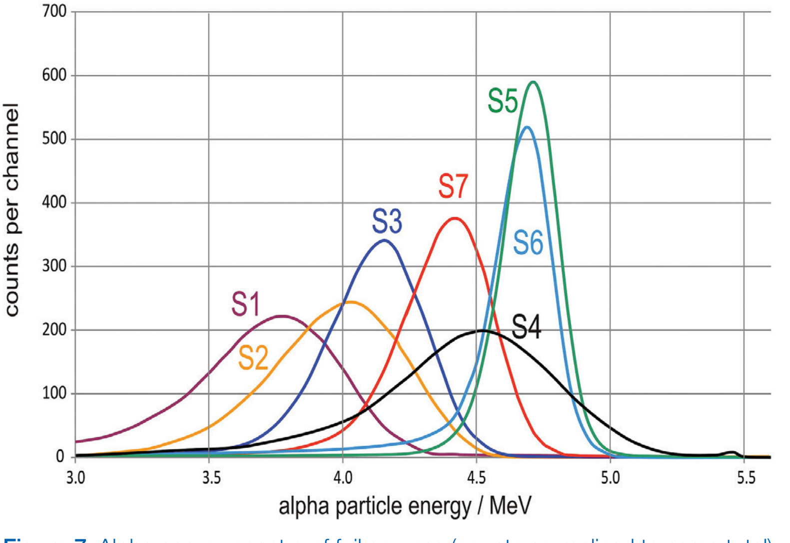

Figure 7 Alpha energy spectra of foil sources (counts normalised to same total)

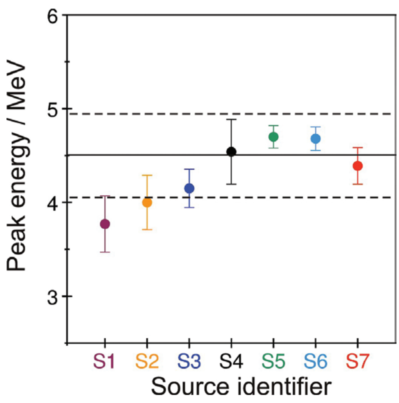

Figure 8 Spectral peak alpha energy and associated FWHM of the sources in this study

4.5 MeV and the 10% tolerance limits (shown in dotted lines).

The spectral peak energies of S1 and S2 were outside of the 10% tolerance of 4.5 MeV. S1, S2 and S4 had an FWHM greater than 0.5 MeV. Using ion transport simulation software (Ziegler, Ziegler and Biersack, 2010), the alpha energy spectra of the sources investigated were found to be consistent with a normal (Gaussian) distribution of americium within the active layer, and the decrease in peak alpha energy of S1 and S2 was consistent with migration of the americium away from the face layer, effectively widening the active layer about 1 μm in a direction away from the foil front surface.

Discussion

The reduction in spectral peak energy is the reason for this odd behaviour of americium-241 sources – much of the emitted alpha radiation will not penetrate the GM detector window. The ZP1481 GM detector has a mica window areal density in the range 2.5–3.0 mg cm^{-2}. The alpha energy needs to be at least 3.0 MeV to penetrate a 2.5 mg cm^{-2} window, and 3.4 MeV to penetrate a 3.0 mg cm^{-2} window. The recess design of the cup sources means there will be at least a 4 mm air gap between the foil and window. So, for alpha radiation travelling the minimum distance from the source to the detector window, the practical minimum alpha energy for detection is going to be in the range 3.6–3.9 MeV, depending on detector window thickness. Compounding the problem, in a typical demonstration set-up in schools, the protective GM detector cap is normally kept in place, which is sensible but reduces the alpha detection efficiency considerably.

There have been enough reports from schools of americium-241 foil sources where the alpha count rate detected by a GM detector has dropped considerably compared with when the source was new to be confident that this is not caused by a batch of foils with reduced spectral peak energy at the time of manufacture. The condition of the sample sources and the leak tests were evidence the source foils were in good condition, so surface degradation is unlikely to be the cause. The reduced alpha count rate was observed both in gold and gold-palladium faces, so the source face composition does not seem to be a determining factor. The evidence strongly indicates that the reduced spectral peak energy with increased energy straggling of S1 and S2 is caused by a net migration of the americium-241 away from the foil face, effectively increasing the thickness of the active layer (Figure 9).

The ion transport modelling points to the active layer in sources S1 and S2 effectively increasing by about 1 μm in thickness in the direction of the substrate. From the gamma spectrometry, an analysis of the X-ray emission attenuation from S1 and S2 agrees well with this, and additionally rules out a low-density contamination on the foil front face (X-rays are emitted by a percentage of decay transitions from americium-241 to neptunium-237). This net migration does not raise a concern for the safe condition of the sources because the americium will remain within the foil but it does have a bearing on the source service life. If the spectral peak energy decreases too far, the source becomes ineffective for this demonstration with a GM tube.

For source S4, if originally the thickness of the foil layers layer met the manufacturer's indicative data, it would suggest some of the americium has migrated towards the face of the source. If this suggestion is correct, this would have a bearing on the safe condition of the foil. The small alpha energy peak of 5.48 MeV is from americium-241 near or at the surface, plausibly at the site of a microscopic pit in the gold-palladium layer. Since the original research, another source has been found exhibiting americium-241 at the foil surface to a greater extent. It reinforces the importance of periodically remotely examining the foil surface (e.g. by digital camera) to check that foil surfaces remain in good condition.

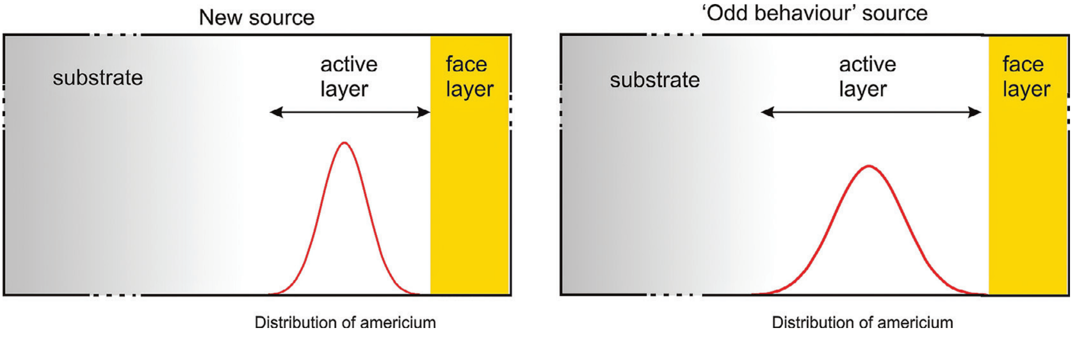

Figure 9 Schematic diagram illustrating the migration of americium, with the red line representing graphically the distribution of americium in the foil, from nothing in the face layer and opposite side of the foil, to a maximum concentration around the middle of the active layer; in the odd behaviour sources, the americium has migrated leftward away from the face layer and deeper into the foil, effectively widening the active layer

The question remains as to why this increased energy straggling over time appears in some americium-241 foil sources and less so in others. For example, in contrast to sources S1 and S2, S3 has a spectral peak energy and FWHM still within the manufacturer's indicative data. HTSL explained that the foil manufacturer at that time – Amersham – produced a variety of alpha foils that had a different ratio of materials in the surface layers, and consequently there is today some uncertainty about the composition of the foils used in school sources from that period. It is therefore a reasonable hypothesis that there are variations in foil design of which some are more predisposed to this change in energy straggling over time.

Following the original research, CLEAPSS asked schools to send in measurements of americium-241 count-rate ratios without and with paper, and more odd-behaving sources have been revealed – the odd behaviour is more common than we thought. However, more data would be very useful; see the 'Further research' section below.

Advice for educational establishments

For schools and other educational establishments that have americium-241 foil sources exhibiting the greater energy straggling and reduced spectral peak energy, there is no evidence currently to suggest that the foils have become unsafe. However, they may be of limited use as alpha emitters when detected by an end-window GM detector such as the ZP1481. The sources can still be used with spark counters and ionisation chambers, including the demonstration of blocking alpha radiation by paper, because these detectors have no solid window. If the protective cap of the ZP1481 detector is removed and the gap between the source cup and GM detector window is kept as small as practicable, 2 mm or less, then the blocking of alpha radiation by paper demonstration using the GM detector may still be effective if the reduction in spectral peak energy has not become too great. Care needs to be taken with the source so close to the unprotected GM detector window; the window would be broken easily if the source were to be knocked into it, damaging the detector permanently. Some school dataloggers have radioactivity detector accessories that use the thinner window ZP1401 or LND712 GM tubes, which have window thickness in the range 1.5–2.0 mg cm^{-2}, but a smaller diameter window than the ZP1481. If the school has one of these thinner window GM detectors, it would be worth trying to see whether it produces a better drop in count rate with the paper block. If these workarounds fail, the source will need replacing for this application.

Although this study looked at just a small sample of sources, a sensible precautionary measure for this type of foil source would be to carry out leak tests at least yearly with remote inspection of the face to check that the surface remains in good condition, and with no discolouration that could be from gold or americium-241 that has migrated through the face layer. CLEAPSS document L93, or for Scotland, the advice from the SSERC website, gives details on how to do leak tests and inspections of cup sources.

Further research

The research into this phenomenon is continuing and we would like your help. This extends to any school in the UK. If you have an americium-241 185 kBq (5 μCi) cup-style source and ZP1481 (or equivalent) GM detector, you can help by taking measurements of the count rate without and with a paper block and sending the results to CLEAPSS. This has to be done in a specific way so that the data can be compared reliably with Figure 6. Details are on document E267 on the CLEAPSS website (science.cleapss.org.uk/Resource-Info/E267-Investigation-of-school-americium-sources.aspx; no password needed). Comparing the results with Figure 6 will indicate sources where there may be a net migration of americium-241, and the direction of migration – away or towards the foil front surface.

Acknowledgements

The authors would like to thank Oriel High School, Crawley, and St Benedict's Roman Catholic School, Bury St Edmunds, who loaned their americium sources for the original study, and High Technology Sources Ltd for the loan of a new source and the information supplied.

References

Amersham (1991) *Safety Instructions for Unpacking and Use of Alpha Foil Sources*. HI 041 Issue 1.

Comfort, J. R., Decker, J. F., Lynk, E. T., Scully, M. O. and Quinton, A. R. (1966) Energy loss and straggling of alpha particles in metal foil. *Physical Review*, **150**(1), 249–256.

Lucas, J. W. (1966) Precautions in the use of radioactive sealed sources in schools. *School Science Review*, **47**(164), 19–27.

Whitcher, R., Page, R. D., Cole, P. R. (2014) Evidence for age-related performance degradation of ^{241}Am foil sources commonly used in UK schools. *Journal of Radiological Protection*, **34**(2), 347–361.

Williams, T. G. (1974) *The Use of Plutonium-239 Sources in Schools and Other Educational Establishments*. NRPB-R28. Harwell: HMSO.

Ziegler, J. F., Ziegler, M. D. and Biersack, J. P. (2010) SRIM – the stopping and range of ions in matter. *Nuclear Instruments and Methods in Physics Research Section B*, **268**(11–12), 1818–1823.

Ralph Whitcher is a CLEAPSS radiation protection adviser, the chair of the ASE's Health and Safety in Science Expert Group and chair of the Society for Radiological Protection's Research and Teaching Sectorial Committee. Email: ralph.whitcher@cleapss.org.uk

Robert Page is a professor of experimental nuclear physics at the University of Liverpool and is leader of the Nuclear Physics Group; he has a broad portfolio of leadership roles in research projects at international accelerator laboratories.

Peter Cole is the University of Liverpool's Radiation Protection Adviser and Radioactive Waste Adviser; he is a professor of physical science at the University of Liverpool and is the president of the Society for Radiological Protection.