

## Half-life demonstrations

Radioactive decay and the half-life concept can be shown by three methods, each of which presents a negligible risk of harm: (1) by the evolution of Radon-220 from thoriated gas mantles; (2) by the elution of Barium-137m from Caesium-137; and (3) by the electrodeposition of daughter products of Radon-222 on a charged rubber balloon.

### Open source work

An *open source* is one where the radioactive material can be dispersed. A *sealed* or *closed* source is one where, by engineering design, the radioactive material is effectively enclosed and cannot contaminate the workplace.

On a point of principle, schools should never work with open radioactive materials unless there is a justifiable reason and the risk is trivial. As a general rule, radioactive materials should be in the form of sealed sources. An exception is the source used for showing radioactive decay and the half-life concept because this practice requires a short-lived radionuclide to be extracted from the parent nuclide and isolated for monitoring elsewhere.

Any open source work must comply with the *Ionising Radiations Regulations* (IRR). The conditions for working with open sources are stiff, putting almost any open-source operation outside the scope of what schools can do. However the new 1999 version of IRR allows for relaxations where the amount of open radioactive material is very low, applying the legal principle of *de minimis non curat lex*, which translates as 'the law does not concern itself with trifles'. In the three demonstration experiments written about here, the amount of material is very low and the risks are almost trifling<sup>1</sup>.

### Radioactive decay of <sup>220</sup>Rn with a Cooknell ionisation chamber

The original radon (or thoron) generator was banned in the 1980s because the source, a compound of thorium, was a fine dust that could be dispersed very easily causing widespread, random contamination. A new version has been devised by Ralph Whitcher<sup>2</sup> which uses four thoriated gas mantles as the source (Fig. 1). Radon gas <sup>220</sup>Rn (gas of this isotope is also known as *thoron*) emanating from the thorium is collected from the source and transferred to an ionisation chamber. The tiny current flowing between the electrodes in the chamber is indicative of the concentration of <sup>220</sup>Rn.

<sup>1</sup> In any work with radioactive material, whether the risks are trifling or not, the operator is morally and legally bound to protect him- or herself and any others who may be affected.

<sup>2</sup> Private correspondence.

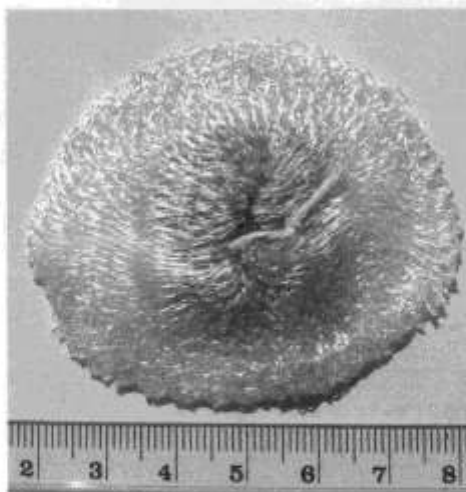


Figure 1 - Thoriated gas-lamp mantle.



Figure 2 - The Cooknell Ionisation Chamber. The bottle, chamber and balloon form a closed system; when the bottle is squeezed, air is displaced to inflate the balloon, enriching air in the ionisation chamber with thoron gas (<sup>220</sup>Rn).

As the radon decays, so does the current, which is amplified by an extra-high impedance amplifier and read on either a voltmeter or datalogger. The half-life of <sup>220</sup>Rn is 54 s.

The apparatus is made by Cooknell Electronics (Email: enquiries@cooknell-electronics.co.uk) and costs £200 including the source (Fig. 2). A sample has been tested for performance and assessed for safety. The source is easy to dispose of at the end of its working life by putting it out with normal refuse.

### Method

Connect either a digital voltmeter set to its 2 V DC range or a datalogger set to monitor voltage at 10 s intervals across the output electrical terminals of the ionization chamber. Open the Mhor clip and squeeze the bottle once, displacing radon-rich air from the bottle. This floods into the ionization chamber and temporarily inflates the balloon. When the hand squeezing the bottle is relaxed, the balloon deflates and air returns to the bottle. The end result is that the air in the ionization chamber is enriched with <sup>220</sup>Rn.

The apparatus is easy to operate and, with a computer and datalogger, will quickly provide a set of readings, six a minute, which, when graphed, shows the exponential decay of count-rate against time (Fig. 3). There is negligible background. The first three or so readings should be omitted from the analysis. The half-life value from the results at  $58 \pm 3$  s is a little above the accepted value (54 s).

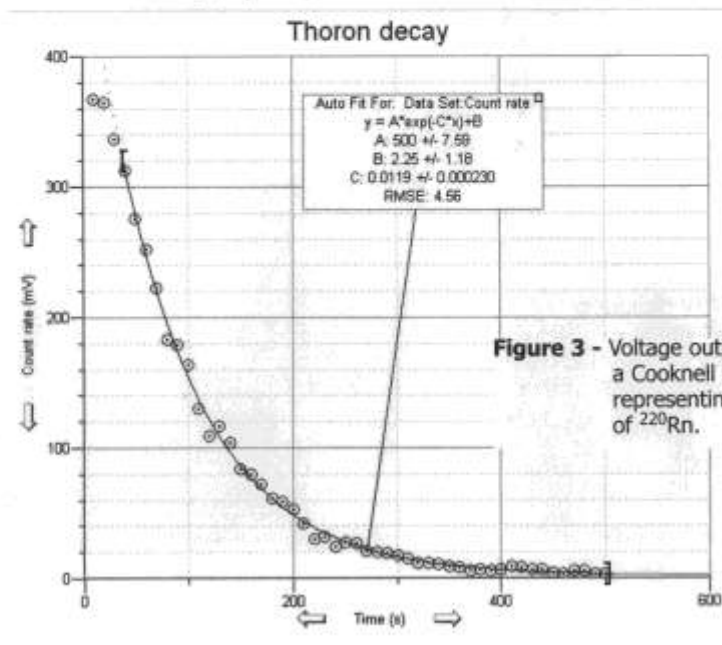


Figure 3 - Voltage output versus time from a Cooknell ionisation chamber representing the radioactive decay of <sup>220</sup>Rn.

**Thorium sources and gas mantles**

The mantle is woven with a cotton or silk thread impregnated with a thorium and cerium compound mixture (helping prevent a lit mantle clogging up with soot). (Mixture: 99% thorium(IV) oxide and 1% cerium oxide.) The mass of thorium is about 12.5% of the mantle's total mass. Some makes of gas mantle do not contain thorium.

The activity of a single mantle is roughly about 1 kBq. With a new mantle with freshly refined thorium, the activities of <sup>228</sup>Th and other progeny below this isotope are nearly equal to the parent of the series, <sup>232</sup>Th (Table 1). Thereafter the activity of the part of the series from <sup>228</sup>Th onwards drops to a minimum at 4 y then builds up to secular equilibrium at 40 y. The emanation of radon <sup>220</sup>Rn is a minimum at 4 y, being 40% of what it had been when the mantle was new, or what it will again become when the mantle is old.

About 25% of the radon <sup>220</sup>Rn generated by nuclear decay within the mantle is evolved as gas. An unused (in the sense of unburnt) thoriated mantle is unlikely to cause much contamination. However after use in a paraffin flame the mantle becomes very fragile and can disintegrate into a fine, thoriated ash. The main hazard is the inhalation of <sup>232</sup>Th and <sup>228</sup>Th dust.

**Radioactive decay of <sup>137m</sup>Ba from an AEA Isotope Generator**

Key words:

**Elution** - The process of removing an absorbed material from an absorbent by washing it in a liquid.

**Eluant or eluent** - The liquor used in the elution before the eluting begins.

**Eluate** - The solution consisting of the material washed out from the absorbent by the eluant.

**Elute, to elute** - Transitive verb describing the process.

**Eluting** - Adj. describing the process.

**How it works**

The Isotope Generator is an eluting source i.e. a small quantity of liquor (or eluant) is injected into the inlet at the top of the source holder, dribbles through the parent source (<sup>137</sup>Cs) and chemically extracts the short-lived daughter (<sup>137m</sup>Ba). The liquor (or eluate) drains from the outlet in the bottom of the source holder and is collected in a small glass vessel.

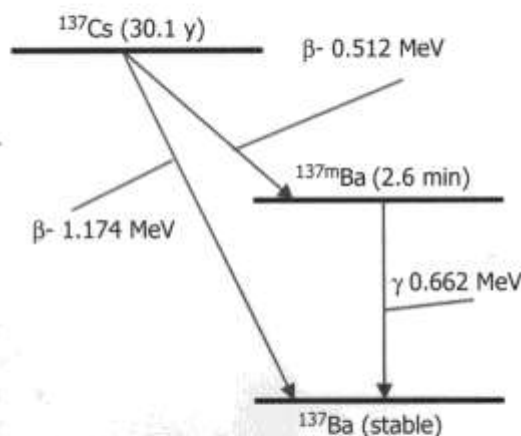
<sup>137</sup>Cs has a 94.6% probability of decaying by beta emission to <sup>137m</sup>Ba, an

Nuclide	Half-life	Alpha	Beta	Gamma
			energy (MeV)	
Thorium-232	1.41 x 10 <sup>10</sup> years	4.0	-	0.06
Radium-228	6.7 years	-	0.04	-
Actinium-228	6.1 h	-	2.1	1.6
Thorium-228	1.9 years	5.4	-	0.08
Radium-224	3.6 days	5.7	-	0.24
Radon-220	54 s	6.3	-	-
Polonium-216	0.158 s	6.8	-	-
Lead-212	10.6 h	-	0.57	0.3
Bismuth-212	60.5 min	6.1	2.3	1.6
Polonium-212	3 x 10 <sup>-7</sup> s	8.8	-	-
Thallium-208	3.1 min	-	1.8	2.6
Lead-208	Stable	-	-	-

**Table 1** - Thorium-232 decay series.

isomeric nuclear state of barium (Fig. 4). The half-life of this decay is 30.1 y. The barium isomer, <sup>137m</sup>Ba, makes an isomeric transition to a lower energy state accompanied by the emission of gamma rays. The half-life of this decay is 2.6 min. The thick-walled plastic enclosure of the source stops beta radiation, but not gamma. The source is therefore effectively a gamma emitter and can be used in demonstration experiments to show the properties of gamma radiation.

Because the half-life of <sup>137m</sup>Ba is very much shorter than the half-life of the parent, <sup>137</sup>Cs, parent and daughter will be in stable equilibrium except for a period of about 30 minutes after an elution. If the activity of <sup>137</sup>Cs is 33 kBq and the eluant is completely efficient at washing <sup>137m</sup>Ba out of the Generator, then the activity of <sup>137m</sup>Ba in the eluate immediately after elution can be presumed to be around 33 kBq.



**Figure 4** - Energy state diagram of <sup>137</sup>Cs and daughters showing the radioactive emissions associated with changes of state. There is a 94.6% probability of the decay transitions going via the <sup>137m</sup>Ba isomer.

**The source**

During manufacture, the radioactive material <sup>137</sup>Cs is incorporated within ion-exchange beads placed within a glued cylindrical plastic enclosure (diameter about 40 mm, height about 30 mm) between two membrane filters with a sub-micron pore size. A pre-stage filter has been added. The glued plastic enclosure has two openings through one of which <sup>137m</sup>Ba, the short-lived radioactive daughter product of <sup>137</sup>Cs, can be eluted by using 0.9% NaCl solution in 0.04 M HCl. When not in use, the openings on the Isotope Generator are sealed by caps (Fig. 5).



**Figure 5** - Isotope Generator in storage box with syringe and tubing. The eluant is in the bottle alongside.

There may be a small amount of bleedthrough of <sup>137</sup>Cs with the eluate. AEA have reported that the <sup>137</sup>Cs content of the eluate was assayed 45 minutes after elution with 2 ml of solution in a batch of 14 generators. One generator showed 200 Bq of <sup>137</sup>Cs in the eluate; all the other generators had less than 50 Bq. The

measurements were repeated over a period of 3 months and the bleedthrough did not deteriorate.

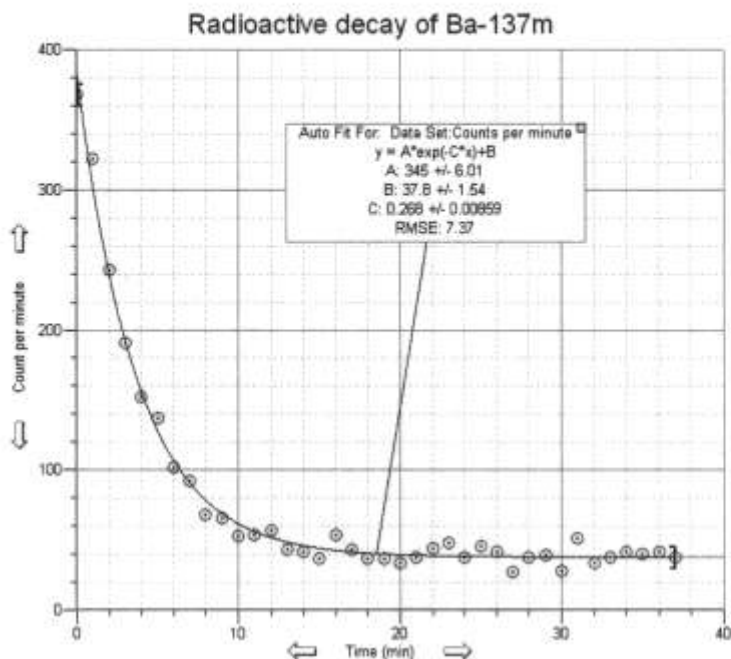
## Procedure

The demonstration is carried out over two drip trays one metre apart. The elution is done over the first tray and the counting over the second one. Beginning with the elution, a receiving bottle with a capacity of 10 ml is stood in a flat glass dish on one of the trays, lined with absorbent paper. The end caps of the source are removed. 2 ml of eluant is drawn into the syringe through plastic tubing dipped in the eluant stock bottle. The tubing is removed from the syringe and the syringe is screwed into the top aperture of the Isotope Generator, which is held in one hand over the receiving bottle while the other hand depresses the plunger transferring the eluant into the source (Fig. 6). Almost immediately liquor (now called the elu-



**Figure 6** - Isotope Generator during an elution. (The source should be gripped in the operator's other hand for greater stability.)

ate) drips from the outlet in the base of the source into the receiving bottle. The whole elution is over in a time of about 15 s. The receiving bottle must then be transferred immediately without delay to the other tray and placed in front of the GM tube already set up waiting (Fig. 7). Counting from the source is then begun. The recommended counting period is 60 s and counting should continue for at least 15 min, but preferably 30, so that the decay is seen to have run its course (Fig. 8). The half-life value derived from



(17.6224, 177.2)

**Figure 8** - Typical set of results from an Isotope Generator. The count was detected with a small-window GM tube.

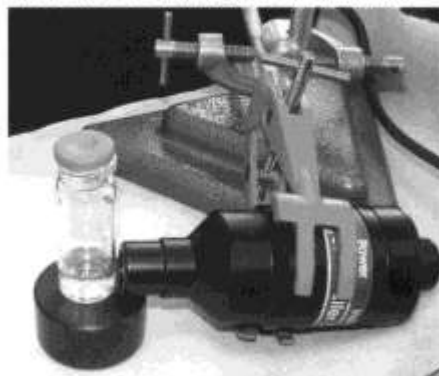
the results worked out at  $2.6 \pm 0.2$  min, matching the accepted value. The counting of background should have begun before the elution and if using a computer datalogger you should time the beginning of the elution so that it ends for the elute to be in position just before a fresh minute's count is about to begin.

## Risk assessment

The Isotope Generator is an open source and during an elution radioactive material is extracted from the source in liquid form and collected in a beaker. The amount of radioactive material is tiny and is such that from any likely accident the risk of harm would be negligible. Comparing the radioactivity of a freshly prepared eluate with a charged rubber balloon (next demonstration), the balloon can be much more radioactive. A risk assessment can be downloaded from the SSERC website.

## What can go wrong?

It is foreseeable that the eluate might spill (but having done about fifty elutions without spillage, this is unlikely).



**Figure 7** - Monitoring count rate from eluate.

Because the volume is limited to 2 ml, the contaminated area will be small. Any spilt  $^{137m}\text{Ba}$  will be self-cleaning. After a period of 45 min, it will have almost completely run-out, its activity being effectively zero. Nevertheless there is a possibility that a small amount of  $^{137}\text{Cs}$  will also have bled through with the eluate. The amount is unlikely to exceed 50 Bq. Therefore you would have to treat a spillage as though you were cleaning up a spill of 50 Bq of  $^{137}\text{Cs}$ . This is done by placing absorbent paper towels on the spilt liquor, which will gather up virtually all the spillage, and putting the wet paper directly on a drip-tray where it should be left for 45 min letting the  $^{137m}\text{Ba}$  decay away and the paper dry. The paper can then be disposed of in a polythene bag with normal refuse.

## Disposal and acquisition

After an elution, the eluate should be left for half an hour for most of its radioactivity to decay. Then it should be poured into a laboratory drain and the drain flushed with water for several minutes. Schools are legally permitted to make this sort of disposal for the small amount of radioactive material being handled. They are also allowed to dispose of an open  $^{137}\text{Cs}$  source with normal refuse provided that the activity does not exceed 40 kBq.

There are two types of Isotope Generator. There is one of low activity at 33 kBq and another of high activity at 370 kBq. Only the low-activity generator should be obtained because it can be disposed of easily at no cost at the end of its working life. The high-activity one must not

## Physics

eluate with  $^{137m}\text{Ba}$ . The count rate  $N$  at time  $t$  for a radionuclide with a half-life  $\tau$  and decay constant  $\lambda$  is given by :-

$$N = N_0 \exp(-\lambda t) + B$$

where  $N_0$  is the initial count rate from the source and  $B$  is the count rate from background. The half-life and decay constant are related by

$$\tau = (\ln 2)/\lambda = 0.693/\lambda$$

The total count from a short-lived radioactive material can be found by integrating the function  $N = N_0 \exp(-\lambda t) + B$  with respect to time for values between zero and infinity. This yields  $N_0/\lambda$  omitting background.

For a typical elution of  $^{137m}\text{Ba}$ ,  $N_0 = 300 (1 \text{ min})^{-1}$  and  $\lambda = 0.267$ , giving a total count of about 1,000.

For a typical charged balloon,  $N_0 = 90 (1 \text{ min})^{-1}$  and  $\lambda = 0.0139$  (approx.), giving a total count of about 6,500.

Knowing forbye that the energy spectrum from the balloon's emissions is

higher than that from the eluate's, which of these two sources presents the greater radiological risk? The main point is that the risk from both sources is trivial.

### Conclusion

Following the withdrawal of the protactinium generator because of the old age of that source there has been no way of showing radioactive decay and obtaining the half-life of a source. This has been a serious omission in the set of practical experiments that underpin the science curriculum.

Here then are three ways of meeting this task. Of the two radiation generators, the revised thoron generator with Cooknell ionisation chamber is the easier to use and costs rather much less than the Isotope Generator. A clear winner? Well no. The Isotope Generator scores tops on the educational ground that the short-lived daughter is plainly seen to be extracted from the parent and taken off elsewhere to be monitored with a GM detector whereas it is less clear what is

happening with the squeeze bottle and ionisation chamber. Also an elution is an interesting bit of theatre. Radioactive decay is an important demonstration. Both of these radiation generators are worth getting. Ask your employer for funding to get both and pay for balloons from your own budget.

### Acknowledgements

I am grateful to Ralph Witcher, RPA for West Sussex Council and Chairman of the ASE Safeguards Committee, for the prior risk assessment and discussions on the new thoron generator, and Cooknell Electronics for the loan of apparatus for testing; to Steven Judge, then of AEA Technology, for technical information on the Isotope Generator, arranging for assays of  $^{137}\text{Cs}$  bleedthrough and the loan of apparatus; and to Marion Milton (NRPB, now HPA) for suggestions on the elution risk assessment, and to specialist radiation inspectors of the HSE, Jo Nettleton and Lindsey Cairns, for their checks and comments. The balloon experiment has been written about in several articles, the first I saw being that by Walkiewicz (The Physics Teacher, Vol. 33, Sept. 1995, 344-5). And my thanks go to Ciara Walsh of Nuclear Technologies for solving the electrodeposition puzzle.

## Annual Conference & AGM of SSERC Limited

Friday 8th of December 2006 at BLCC, Dunfermline

### Draft Programme & Application Form

9:15-10:00	Registration, exhibitions open and coffee.
10:00-10:05	Welcome and introduction:
10:05-10:30	Keynote Address: To be confirmed
10.30-11:00	Supporting Science Education through CPD Project Overview by Kath Crawford, Depute Chief Executive Officer, Projects Primary CPD by Aileen Gray, Development & Educational Support Officer
11:00-11:30	Coffee and exhibitions
11:30-11:50	CPD & related activities for School Science Support Staff Training Packages under Development – by the Project Team
11:50-12:00	SafetyNet CD News by Ian Birrell, Network Designer, SSERC
12:00-12:20	Radioactive Sources in Schools by Jim Jamieson, Radiation Protection Adviser, SSERC
12:20-13:00	Health & Safety – HSE Inspector - School visits and what they throw up!
Closing remarks	– Bristow Muldoon MSP
13:00-14:00	Lunch and Exhibitions
14:00 ->	Annual Report and General Meeting Board Meeting of SSERC Ltd. (Directors & Officers of the Company)
14:00-14:30	Exhibitions
15:00-16:30	Open afternoon in SSERC facilities at Pitreavie, Dunfermline

I wish to reserve a place at the 41st Annual SSERC Science, Technology & Safety Conference:

Name : \_\_\_\_\_ Position \_\_\_\_\_ Date \_\_\_\_\_

Address \_\_\_\_\_

I shall / shall not attend the SSERC Open Afternoon at the SSERC facilities in Pitreavie, Dunfermline in the afternoon. I enclose my cheque/official order\* in payment of the delegate fee(s) of £60 + VAT (£70-50) for members\*/£80 + VAT (£94) for non members\* [delete if inapplicable]. I would like\*/do not require\* a receipt.

Return to: Catherine Russell, 2 Pitreavie Court, South Pitreavie Business Park, Dunfermline, KY11 8UB