USE OF URANIUM AND THORIUM IN TEACHING AND RESEARCH EXPERIMENTS

(AURPO Guidance Note No: 2)

1. Introduction

The use of uranium and thorium reagents in teaching experiments is carefully controlled because of the chemical toxicity of both elements. Consideration must however also be given to the potential radiotoxicity of these elements and the radiation fields which may be associated with them as, on occasions, these may be more limiting than the chemical toxicity. Not only must the handling of these substances be considered carefully but also the amounts that are held and the eventual disposal of any waste material and redundant reagent.

Some physical science departments may have thoron generators (colloquially known as "puffers") used in half life and cloud chamber demonstrations and experiments.

Of further important consideration is the status of geological collections which hold samples of uranium and thorium ores, some of which are from old collections and may include samples of high activity ores from such countries as the Belgian Congo.

The uranium and thorium in the reagents are generally chemically isolated before being converted to the salt form. The source of the uranium will alter the radiological risk. With natural uranium, it is likely that the U-234 will be present in equilibrium with the parent U-238 as any chemical extraction will extract both equally well from an ore source. The thorium and protactinium in the decay series between them will also rapidly grow into equilibrium. Also present will be the naturally occurring U-235.

If the reagent is made from depleted uranium then the U-235 and U-234 will be reduced by at least 40% but possibly up to 60%. The 1620 year half life of the Ra-226 effectively stops the decay series and ensures that once the uranium is isolated the subsequent daughters are not going to be present in laboratory reagents. The situation with thorium reagents is rather different and the quantity of any daughter products present will depend on the age of the reagent. Even on time scales of only a few years they could build up to significant levels.

The situation with the geological specimens is totally different. In the geological specimens all the daughters in both decay series could potentially be present. The levels of radon measured in rooms containing these specimens would indicate that there must be some disequilibrium in some samples because of the escape of gaseous radon daughters.

2. Specific activities of uranium and thorium

The data that follow have been selected for experiments using reagents:-

(a) Natural uranium of composition U-238 (99.3% natural abundance) in secular equilibrium with its daughter isotope U-234 (0.005%) and U-235 (0.7%). The calculated specific activity of this material is 25Bq mg^{-1}

NB If it is known that depleted uranium has been used then the activity can be assumed to be approximately 16 Bq mg^{-1}. Allowance may be made to the data listed in the following sections if depleted rather than natural uranium is to be used in experimental work.

(b) Natural thorium, of composition Th-232 (100%) in secular equilibrium with its daughter isotope Th-228 and having a specific activity of 8Bq mg^{-1}.

Throughout the rest of the note it is assumed that the uranium and thorium have been initially chemically purified to remove activities due to the daughter radionuclides in their decay series.

3. Legislative background

These materials are subject to the legislation controlling the holding and the use of radioactive materials. The three main pieces of legislation we are concerned with here are: The Radioactive Substances Act 1993 (RSA93); The Ionising Radiations Regulations 1999 (IRR99) and the Nuclear Safeguards Act 2000. Because of these legislative requirements, people working with U and Th should be included in the systems used to control and supervise work with any radioactive substance in the university (or research institution.). The nature of these substances and the departments in which they are used can sometimes make ensuring this happens not easy to implement, as often they may be the only source of radioactive material present in the department and there may not be a departmental structure for dealing with them in place.

3.1 Radioactive Substances Act 1993

Both reagents and ores are subject to exemption orders under the Act. At the time of writing this note these are under review and it is likely that the detail below will change when the new format for exemption orders is introduced. It is imperative that any new legislation is consulted in order to confirm the numerical values of any amounts quoted below. It is not anticipated that the exemption orders will be totally revoked but will be replaced by a simplified set of equivalent arrangements.

It is felt unlikely that a teaching laboratory will need to use or hold quantities of uranium or thorium in any form that will exceed that permitted by the relevant exemption order. Where this is not the case the situation is outwith the guidance given here and must be considered on its own merit and will need to be covered by a registration from the Environment Agency and be subject to a prior risk assessment under IRR99. Also note that once more than trivial amounts of uranium are purchased international regulations must also be considered and inspection by the HSE Safeguards Office on behalf of Euratom is carried out on the Universities which have such larger amounts.

The exemption orders are summarised below:

3.1.1 The Radioactive Substances (Prepared Uranium and Thorium Compounds) Exemption Order SI 1962 No.2711

This Exemption Order includes both solid and liquid prepared compounds of both elements as defined in section 2. The limitations of the exemption are covered by this extract from the order -
The exemption order is granted to allow the following purposes:
"demonstrating, or testing , measuring or otherwise investigating (otherwise than by a process of isotopic separation) the characteristics of, any compound or substances falling within that Article (*ie .included in the definition of acceptable compounds)* or some other material which is not a radioactive material"
Subject to the condition "that the weight of all the uranium and thorium contained in all the compounds or substances …….. present on the premises at any one time does not exceed two kilogrammes"

The exemption order allows solid or liquid waste produced as a consequence of carrying out one of the above purposes to be disposed of subject to the following limitations. For solids only 100 grammes of uranium and thorium are disposed of in a day and this must be as refuse to a disposal contractor or deposition by someone else (other than a disposal contractor) on a tip or dump which is essentially used for non radioactive waste. Liquid waste is disposed of by the normal method of liquid waste disposal i.e. down the sink. Gaseous waste is subject to a limit in the aggregate of one gramme weight of the uranium and thorium added together and the discharge must be to the atmosphere.
(The practical application of this is discussed later in the note)

3.1.2 The Radioactive Substances (Uranium and Thorium) Exemption Order. SI 1962 No.2710

This exempts from registration and authorisation limited quantities of uranium and thorium metals and several items which contain natural thorium and uranium as defined above in section 2. These include thoriated tungsten, containing less than four per cent by weight of thorium, incandescent mantles and thoria ware. 2kg by weight of U and Th can be held on the premises under this exemption order and a 100g/ day of solid wastes can be disposed of from the premises each day as described in 3.1.1. In addition up to 25kg of thoria ware can be disposed of from the premises each month.

3.1.3 The Radioactive Substances (Geological Specimens) Exemption Order SI 1962 No.2712

This applies to natural rock or minerals where the activity of decay products of the parent uranium 238, thorium 232 and uranium 235 do not exceed the activity of their respective parent. Exemption from registration is granted for the purposes of displaying, demonstrating or investigating its characteristics or for sale purposes. It is further subject to the condition that the total weight of uranium and thorium present on the premises at any time does not exceed 100 kilogrammes. (This is more than sufficient for all but the very largest of geological collections)
Exemption from authorisation is subject to the materials being substantially insoluble in water, material which has been contaminated by an exempted specimen and solid or liquid substances arising from the use of exempted material. The limits on the amount that can be discharged are the same as for the Uranium and Thorium compounds and are subject to the same procedure as for these compounds to be described later in the note.

3.2 Ionising Radiations Regulations 1999

Although these materials are exempt from the RSA they are not exempt from the requirements of the ionising radiations regulations. It is not thought likely that students may be present who are under the age of 18. (The use of other radioactive trace experiments will almost certainly take place later in their training when they are over 18 years old)
To set this hazard in context, data from Schedule 8 of the Ionising Radiations Regulations illustrates that the radio toxicity of natural thorium and uranium is comparable to or greater than that of plutonium -239 depending on the state of equilibrium with their respective daughters.

These notes contain suggestions for radiological protection and recommend the maximum weights of uranium and thorium that should normally be used in experiments. The basic philosophy adopted is that conditions should never be created in a teaching laboratory that would require the creation of a controlled area or to require students to be designated as classified persons. It is suggested that these conditions will also normally form the basis of research experiments.

The Ionising Radiations Regulations limit not only the activity of materials that may be used in an experiment without detailed attention to radiological considerations, but also place other obligations on the designers and teachers of student experiments:

1) Students, aged 18 years and over, are regarded as employees of the host institutions for the purposes of the Regulations.

2) Students between the ages of 16 and 17 years are limited to a maximum annual radiation dose of 6mSv. Students aged 18 years and over are limited to maximum annual radiation dose of 20mSv. The following notes assume that students in universities and polytechnics will fall into this latter category, (i.e. aged 18 years and over).

3) Although it is expected that students will encounter very small doses in the course of teaching experiments, they must be fully briefed that any exposure to ionising radiations may involve a health risk.

4) Full responsibility rests on the host institution to provide adequate training in the hazards of ionising radiations and the specific manipulations required in the course of the experiment. This responsibility extends to ensuring that adequate protective clothing is provided.

5) Section 28 of the regulations requires records to be kept of the quantity, location and disposal of compounds of uranium and thorium. Copies of the records must be kept for at least 2 years (5 years is recommended to tie in with minimum SEPA requirements for work with radioactive materials). It is suggested that for most purposes it would be more appropriate to describe the 'amount' of these substances in terms of mass rather than activity.

4. Experiments involving the use of CLOSED SOURCES

In these notes, closed sources of uranium/thorium are regarded as sources that would not be expected to give rise to any contamination in the course of normal manipulation. If there is a significant risk that the source might be dropped, containment (for example of uranium/thorium reagent powders) in simple glass bottles with a screw top should not be regarded as providing adequate security against the risk of contamination.

In, assessing what might be regarded as an acceptable dose to which a student might be exposed in the course of a teaching experiment, many institutions will already have their own agreed limits. However, the following considerations should be noted:

1) the Regulations require that all necessary steps are taken to restrict so far as is reasonably practicable the extent to which employees and other persons are exposed to ionising radiation.

2) where exposure of the hands is concerned, the Regulations require that the time averaged (over 8 hours) dose rate should not exceed 75 μSvh^{-1} outside a "controlled area".

It is proposed, therefore, that the maximum weight of material that should be available to students as a closed source be such that the dose rate measured 'at a few centimetres' from the source does not exceed 75 μGyh^{-1}. In the absence of precise data, the approximate weights of metal that correspond to this limitation are:

250 g uranium 100 g thorium

In deriving these weights, it is assumed that

1) The dose rates arise essentially from gamma emissions alone, beta and alpha particles being absorbed within the source containment material.
2) The source will always be handled with tongs or remote handling tools, never picked up directly by hand.

If the source containment is insufficiently absorbing to shield the beta radiations, the mass of materials available to students must be more restrictive to compensate for the increased radiation dose. Rudimentary measurements suggest that under these circumstances, the masses of uranium and thorium listed above should be further reduced by a factor of ten.

5. Experiments involving the use of OPEN SOURCES

If experiments make use of open sources of uranium or thorium (for example solutions, on uncontained powdered reagents) it is important to minimise the risk of contamination by careful experimental design. The effectiveness of these measures (in minimising contamination) must be proven by regular use of a suitable contamination monitor. In some circumstances the most appropriate instrument will be an alpha monitor, since the background reading of these devices is essentially zero. However, users must be made aware that alpha particles have a low penetrating power and that anomalously low readings will be obtained if, for example, a spill of uranium/thorium solution is absorbed within a protective bench coating material.

If experiments make use of powdered reagents or coloured solutions, a simple expedient that may be used in conjunction with instrumental contamination monitoring is to carry out such work on a white experimental tray and/or bench coating material. Some uranium reagents are strongly coloured and even small spillages of powder or solution may be visually detected against a suitable background surface more sensitively than with simple monitors.

If there is likely to be any uncertainty in the interpretation of contamination monitor readings <u>or</u> if airborne contamination is possible but cannot be monitored satisfactorily, it will be necessary for the amounts of material used to be kept strictly within the numeric limits specified below to avoid the necessity of designating the laboratory as a controlled area.

5.1 Uranium

The amount of uranium reagent that complies with contamination criteria depends on the chemical form (the 'lung class'); the least soluble reagents suffering the greatest restriction since they are retained in the lungs for the longest period.

The retention times in the pulmonary region of the lung are defined as follows:

	Retention half time
lung class D	less than 10 days
lung class W	10 to 100 days
lung class Y	more than 100 days

In a teaching experiment involving, for example, the manipulation of powdered reagents, the <u>entire</u> class should not normally have access to more than the following amounts in any laboratory:

lung class D 19.1 g of soluble uranium reagent (e.g. UF_6, UO_2F_2, $UO_2(NO_3)_2$)
lung class W 11.4 g of less soluble uranium reagent (e.g. UF4, UC14, U03)
lung class Y 0.38 g of highly insoluble oxide (e.g. UO_2, U_3O_8)

As an alternative, the experiment must be so designed that there is no reasonable possibility of uranium contamination levels exceeding 2mgcm^{-2} averaged over 300 cm^2 (i.e. 600 mg over 300 cm^2).

5.2 Thorium

The maximum weight of thorium reagent to which the class has access should be limited to 0.086 g. If the experiment demands the use of greater weights of reagent, experimental procedures must be designed such that there is no reasonable possibility of thorium contamination levels exceeding 0.84 mgcm^{-2} averaged over 300cm^2 (i.e. 252 mg over 300 cm^2).

In deriving these data, it has been assumed that reagents are sufficiently coarsely ground and techniques used are such as to prevent airborne contamination.

If there is a risk of airborne contamination, then adequate facilities (e.g. a glove box) must be provided. Such considerations are particularly important in the case of thorium since as little as one microgram of finely divided thorium per cubic meter of air requires the designation of a controlled area.

It is expected that experiments involving the use of uranium/thorium in solution form will conform to the limits outlined above provided concentrations are restricted to 1 mg ml^{-1} and normal solution handling techniques are employed. In such work MOUTH PIPETTES MUST NEVER BE MADE AVAILABLE to students.

In deriving these limits it is assumed that students will be required to wear a lab coat and disposable gloves whenever handling uranium/thorium reagents of any mass and that the limitations on the mass or uranium and thorium available apply to the entire laboratory in which experiments are carried out, not an individual bench.

Although the above limits are considered to be appropriate for first-degree course students in universities and polytechnics, it should be noted that the International Commission on Radiological Protection recommends that for students under the age of 18 years, no more than 50 Bq (5 mg) of natural uranium and 5Bq (1mg) of natural thorium should be used for separate teaching exercises.

6. Geological samples and Minerals

Collections of geological specimens containing uranium and thorium minerals can easily produce dose rates and radon levels which could involve the creation of controlled areas. Historically geologists have not considered specimens like this to be a problem and this attitude still pertains. These collections are currently rarely used and often are just sitting, almost forgotten, in the store room containing the geological specimens. Occasionally the more demonstrative departments could have some of them on display. A case can be made for disposing of them and some EA/SEPA inspectors are known to support this option. The department holding them may agree and if this is the case they should be disposed of under the exemption order immediately so that the problem is removed. Many departments will however take the view that although they are not currently being used it is highly likely that in the not too distant future interest in these minerals will be revived and the collections will again be used. Resurrecting such collections will not be easy if at all possible. It is felt that keeping these collections should be supported and that means should be developed to ensure compliance with the regulations. One unusual aspect of uranium/thorium geochemistry is that these elements can concentrate in minor phase mineral grains in some rock types (e.g. granites). If work involves separating these minerals, especially zircon and rare earth element grains, from matrix phases, enhanced activities of uranium, thorium and daughter isotopes will be encountered compared with the bulk specimens. In these circumstances, the legal obligations of the Ionising Radiations Regulations may apply even though the major sample specific activities fall below the threshold of 100 Bq g^{-1} on which legal definition of a radioactive substance is normally based.

Typical activities reported by Dixon (NRPB-R143) found in these minerals are as follows:

Mineral	Chemical formula	Principle source of activity	Typical Specific activity Bq g^{-1}
Bastnaesite	$CaFCO_3$	Th	5
Baddeleyite	ZrO_2	U	18
Ilmenite	$FeTiO_3$	Th	1
Phosphate rock (apatite)	$Ca_5(PO_4)_3(OH,F,Cl)$	U	3
Pyrochlore	$NaCaNb_2O_6F$	Th	70
Zircon	$ZrSiO_4$	Th	8

6.1 Storage of samples

6.1.1 Doserates

The gamma photons emitted from some of the daughters in the U- 238 and Th-232 decay series are very energetic. (The lead half value layer for Ra-226 in equilibrium with its daughters is nearly 17mm). There is a popular misconception amongst Geologists that putting them in a box lined with lead flashing 2mm thick will provide adequate shielding. This is not the case. Even small collections of these rocks will give doserates in excess of 7.5 µS/h a metre away with large collections giving substantial doserates. It is generally not easy to store them along with the other geological specimens to which access is regularly required by departmental staff and students. Normally the easiest way is to isolate them in some way and create a controlled area to which access can be restricted to a few designated persons entering under a working procedure to ensure they do not require to be classified.
The doserates close to geological specimen displays should be checked to ensure that a controlled area does not exist. If it does the easiest way to deal with it is to remove the offending specimen(s).

6.1.2 Radon Levels

Geological stores are frequently closed rooms with little ventilation. Even if the specimens have been isolated into a secure area the radon levels in these rooms can easily rise to above $1000Bqm^{-3}$ again resulting in the creation of a controlled area. Radon measurements must be made in all areas storing geological specimens to ensure that the $400\ Bqm^{-3}$ controlled area limit is not exceeded. If it is, the level must be reduced or entrance to the area must be restricted using working procedures to ensure that anyone entering is not required to be a classified worker. Experience has shown that the only practical way of reducing these levels is generally to introduce a system of forced ventilation. The creation of a gas tight enclosure for these samples is very difficult.

6.2 Work with geological specimens

The National Radiological Protection Board (NRPB-R131 : 1983) deals with the possible radiation hazards to collectors of geological specimens containing natural radioactivity. However sample collections should be regarded as stand alone collections and each one should be treated on its own characteristic dose rates and activity.

It is worth highlighting again that unlike laboratory reagents that have undergone chemical purification, these specimens will also contain activities of all daughter isotopes and these are likely to be in secular equilibrium with parent U-238, U-235, or Th-232. These daughters will therefore contribute substantial additional activity to the sample.

The author of the NRPB report assumed that a typical collection of minerals includes about 1% of samples that contain significant amounts of radioactivity. Of these, a proportion may be 'high activity' specimens that have been obtained from “high grade” mineral areas. It was assumed that if 100 hours per year are spent handling the collection and the average annual handling time of radioactive specimens could be one hour.
Persons working on the high activity specimens could however have a significantly longer period of exposure and each case must be considered on an individual basis and a proper risk assessment carried out.

However, if realistic assumptions are made about collecting and handling habits (as listed above) annual doses arising from collecting are likely to amount to only 10-15 % of the level appropriate for limiting radiation exposure to members of the general public. A suitable working procedure can therefore ensure that classification of workers is not required.
The dose for person actively working with them however could be much higher and although classification is still not required it will be useful to issue a personnal dosimeter to demonstrate that this is the case.

Sensible precautions applicable to the collecting, handling and viewing of uranium/thorium minerals include the following:

1) Wear disposable gloves, wash hands after handling samples.
2) Do not store samples in an occupied area.
3) Extreme caution must be taken should it be necessary to grind such samples to a fine powder prior to chemical analysis. The worker and laboratory should be treated in exactly the same way as any other laboratory handling radioactive materials.
4) If possible, samples should be kept wet during the grinding process. If dry grinding is necessary, grinding apparatus should be fitted with an effective air/dust extract system. It is recommended that the extraction efficiency of such equipment should be tested by direct measurement of airborne contamination levels.

7. Use of Thoron generators

The release of Rn-220 ("thoron") gas from thorium compounds is still used in some teaching laboratories to provide a convenient source of short-lived activity.

Apparatus suppliers offer(ed) flexible plastic squeeze-bottles which contain approximately 25g thorium hydroxide. By squeezing the container, a quantity of air/gas mixture can be ejected from the space above the solid. A simple cloth filter is incorporated to discourage the release of powder. (Old devices do not incorporate this rudimentary safeguard and some may still be held in long established departments.)

Typically a thoron generator of the kind described above will, in the absence of significant leakage, contain about 100 kBq of Rn-220 in equilibrium with the thorium hydroxide. If part of this Rn-220 is puffed out into the room air, it will decay with a half-life of 52s into the daughters which are also radioactive and have somewhat longer half-lives. The ICRP recommendation for the Annual Limit on Intake for Rn-220 daughters, in terms of equalibrium Radon activity, is 800 kBq.

Making reasonable (but pessimistic) assumptions, it is unlikely that the intake resulting from a single "puff" of the thoron generator in a badly ventilated small laboratory, would be greater than about 1 kBq. Thus occasional use of these devices is unlikely to be a significant radiological risk. A greater risk might be caused by the contamination which could result from ineffective filtration of finely divided thorium hydroxide powder, and this should be taken into account in making these devices available to students.

8. Disposal of Uranium & Thorium Ores, Compounds, Items and Liquids.

The situation with regard to the disposal of these materials has been clarified by the issue of a guidance note by the Environment Agency. This note issued by the Environment Agency's Surplus Source Disposal Programme Team (now disbanded) in June 2006 was primarily aimed at helping schools but the practical techniques for dealing with these materials apply equally well to Universities/Research Institutions and it is recommended that they be adopted. The Scottish Environmental Protection Agency have a slightly more relaxed approach to the interpretation of the exemption orders but adopting the EA recommendations are considered to be good practice for institutions in Scotland.

8.1 Compounds (Soluble and Insoluble) and Liquids

The normal refuse from your establishment must be removed by your Local Authority's internal waste disposal service or by an external company who operates a contract on their behalf or alternatively by a contractor under direct contract to your establishment who then disposes of it to a landfill site. If either of these criteria are satisfied then the material must be split into aliquots each with no more than 100 grammes of uranium and thorium (together). If already in a smaller amount than this all labels etc must be removed from the bottle. These should be placed into a small "rugged" container such as a plastic screw top pot and the pot filled with "polyfilla" or similar propriety grouting compound. One of these can be disposed of in your rubbish per day. The wording of the order is 'disposed of in any day on or from the premises'. This implies that although you can dispose of one per day, this is only the case if the rubbish is removed every day, so it is essentially 100 grammes per rubbish removal. The waste contractor does not need to be told you are doing this.
Compounds that are soluble in water or dilute acid and can not be disposed of as above for some reason by the preferred method above can be disposed of down the sink subject to the requirement that the discharge goes to a public sewer. Again there is a limit of 100 grammes per day of uranium and thorium together. You do not need to inform the sewage undertaker that these disposals are taking place.

8.2 Items

If exempt these can be put out as normal refuse. Again no limit appears to be in place regarding the number of items but it would appear prudent to limit them to one item per dustbin. There is the same weight restriction of 100g of total uranium and thorium per waste disposal collection unless it is thoria ware when the limit is 25 kilogrammes per month.

8.3 Uranium and Thorium Ores

The normal refuse from your establishment must be removed by your Local Authority's internal waste disposal service or by an external company who operates a contract on their behalf or alternatively by a contractor under direct contract to your establishment who then disposes of it to a landfill site. If either of these criteria are satisfied then the material must be split into aliquots each with no more than 100 grammes of uranium and thorium (together). These can then be put out into the normal waste. The amount of uranium and thorium in rock samples is normally quite small and the Environment Agency suggest that as a rule of thumb a 5:1 ratio of rock sample weight to uranium/thorium weight would given a margin of safety in creating suitable aliquots.

8.4 Non Exempt material and Items

If the routes of disposal identified above are not available or the waste is outwith the exemption orders then a specialist waste contractor must be used.

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