Review of Radioactive Waste in Ireland

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Introduction

Radioactive sources have been used in Ireland for the benefit of society since the early nineteen hundreds. In 1914, The Radium Institute was established in Dublin [1] and used radon gas collected in thin glass tubes to treat patients suffering from cancers. Until the mid 1970s the use of sealed radioactive sources in industrial applications in Ireland was on a very small scale. During the late 1970s and early 1980s their usage increased significantly as the level of industrialization in the country increased. Similarly in the 1980s there was a marked increase in the use radioactive sources for medical purposes, especially unsealed radioactive sources as more and more hospitals established their own nuclear medicine departments.

Radioactive waste in Ireland, originates mainly from the use of radioactive sources in industries, hospitals, educational establishments and research organisations. It consists of disused sealed and unsealed sources, disused equipment and consumer products containing radioactive materials such as smoke detectors, a small amount of contaminated materials and the uranium fuel slugs from a dismantled sub-critical assembly for teaching purposes.

No nuclear industry exists in Ireland and the manufacture of radioactive material is limited to the production of fluorine-18 for use in nuclear medicine diagnostic examinations. However, while the amount of radioactive waste in Ireland may be small compared with nuclear countries, the quantities involved still present a potential radiation hazard and must be maintained under secure regulatory control to prevent their unlawful disposal and to protect workers and members of the public against radiation exposure. In the case of long-lived radionuclides, this can place a long term burden on the users/owners, the regulatory bodies and possibly the State itself.

This paper presents, primarily, a summary of how Ireland deals with the radioactive wastes arising from the use of radioactive sources which are held under regulatory control, and possible future developments which could reduce the amount of this waste which is stored presently at locations throughout the country.

A comprehensive RPII report on these issues, entitled “Towards a Radioactive Waste Management Policy in Ireland”, was compiled and presented to the Irish Government in 2006. This report refers to all the legislation applying to the control of radioactive waste and radioactive materials in Ireland and discusses in greater detail all the issues mentioned in this paper.

Regulatory Control

The Radiological Protection Institute of Ireland (RPII) is the national competent authority which regulates the use, custody and disposal of radioactive materials and ionising apparatus. It was established under the Radiological Protection Act of 1991[2] which lays down its functions, responsibilities and powers.

Regulatory control of radioactive sources and materials is maintained through a system of licensing and inspection. The practices of custody, use and disposal of such sources or materials can only be carried out under a licence issued by the RPII. Prior to acquiring any such item, an application must be made to the RPII requesting a licence for the practice(s) required. A radiation risk assessment, information on how and where the source(s) will be used, the experience of the users and the radiation safety procedures to be implemented must be supplied with each application.

Each licence has a set of legally binding conditions attached. These include conditions relating to the licensed items both during their useful lifespan and to their storage when no longer of use i.e. when the source(s) has become "disused".

The main principles applied to control the build-up of radioactive waste in Ireland are:

- The requirement on licensees to have a take-back arrangement with the suppliers of sealed sources.
- The justification for the use of the radioactive source especially where a possible non radioactive alternative may exist. For example, this has been applied in the case of lightning preventors containing radium-226 and americium-241 foils the importation of which is prohibited.
- The licensee is responsible for the safe use, storage and disposal of each licensed source.

Main Legislation

Besides The Radiological Protection Act 1991, the main pieces of legislation relating to sources of radioactivity are:

- Statutory Instrument No.125 of 2000 [3] which substantially implements the EU Basic Safety Standard of 1996, EURATOM Council Directive 96/29/EURATOM
- Statutory Instrument No 875 of 2005 [4] which transposes the EURATOM High Activity Sealed Sources (HASS) Directive 2003/122/EURATOM into law.

Ireland is also a signatory to the IAEA Joint Convention on the Safety of Spent Fuel Management and the Safety of Radioactive Waste Management.

Licence Conditions

Conditions are attached to each licence to ensure that good radiation safety principles are applied by each licensee in their work practices. The main conditions applying to radioactive waste can be summarised as follows:

- The disposal of any radioactive source is subject to prior approval by the RPII unless the practice of disposal is specified on the licence.
- Licensees must notify the RPII of any proposed change in the licensed inventory of sources.
- Licensees are required to ensure the presence of all sources on a monthly basis and maintain a record of these checks.
- Disused sources are to be securely stored and clearly labelled.
- A source take-back agreement with the supplier is required prior to licensing of sealed sources.
- Licensees are encouraged to return disused sources which are covered by a take-back agreement.
- Transportation of sources is carried out in accordance with IAEA Transportation Regulations.
- The licensee is required to maintain proper records, sources and containers are to be clearly labelled, adequate training and radiation safety procedures are to be provided to staff. The licensee is also required to appoint a Radiation Protection Adviser and /or Radiation Protection Officer, depending on the facility.

The RPII carry out inspections to ensure compliance with these and other conditions of the licence. In addition, the RPII has commenced inspecting radioactive source storage facilities while accompanied by an officer from the National Crime Protection Office of An Garda Síochána (the Irish Police Force). This has proved a very useful initiative and has added to the security of radioactive sources at the premises visited.

Radioactive Wastes from Regulated Practices

As of January 2008, there are approximately 475 licensees in Ireland who are licensed for the custody and use of radioactive sources, and of these 84 have custody of disused sources. A description of the various wastes arising from these licensees is provided in the sections below.

Disused Sealed Sources

Sources that are no longer needed but are still held in custody by the licensee are held under licence for "custody only". An inventory of the disused sealed sources at the end of 2007 is provided in Tables 1. The original use of these sources range from industrial gauges containing high activity sources to low levels of activity in sources used for teaching purposes. The radionuclides are grouped into 4 categories, those with a half-life greater than 10 years and those with a half-life of 5 to 10 years, 1 to 5 years and less than 1year. Within each category they are listed in the order of their total activity. There are 1982 disused sources with a total stated activity of 1.73TBq. The radionuclide contributing the most activity to the inventory is tritium with an activity of 598GBq from 36 sources, with the next largest contribution of 225GBq coming from 15 americium-241/beryllium neutron sources. Thirty-three of the tritium sources are emergency exit signs held by one licensee and have a total activity of 555GBq.

The activities of the radionuclides in Table 1 are based on the activity of each source as provided by the licensee. Many of the activities refer to the date of manufacture and have not been corrected for radioactive decay. As some of these sources date back twenty years or more, the activities shown are an overestimate of the true activity – especially in the case of the shorter half-life sources.

There are 1830 sealed sources with half-life greater than 10 years. Of these, 1400 are uranium-238 fuel slugs from a dismantled sub-critical assembly held at a university. In the remaining sources there are 7 which fall under the HASS Directive and these are listed in Table 2. Five of the HASS sources are held by 3 universities and the remaining two by an industrial company.

There are 152 sealed sources with a half-life less than 10 years. These are held by 37 licensees. Twenty-five of these licensees also hold disused sources of half-life greater than 10 years. Due to the age of many of these sources, their total activity may be considerably less than the 184GBq recorded.

Sealed sources held in temporary storage while awaiting their return to the supplier or export from the country are omitted from Table 1, as are numerous miscellaneous disused sealed sources, of low or unknown activity, which are held in storage by a small number of licensees

A pie chart of the distribution of disused sealed sources with a half-life greater than 5 years, by user category, is shown in Figure 1. Due to the 1400 fuel slugs from the dismantled sub-critical assembly, the 1563 disused sources held by the educational institutes far outnumber those held by the other user categories. However the total activity of these sources, 566GBq, is less than the 615GBq activity for the 144 disused sources held by the state agencies (This category includes both State and Semi-State Bodies). These activities are dominated by the presence of 33 emergency exit lights containing a total of 555 GBq tritium in the inventory for state agencies and in the case of the educational institutes by 2 sources, a strontium-90 and an americium-241, that contribute 325GBq between them. The industrial and medical categories hold 116 and 59 disused sealed sources respectively with corresponding total activities of 326GBq and 96GBq. The numbers of licensees holding these sealed sources in each user category are: State Agencies 10; Medical 9; Industry 34 and Educational Institutes 14.

Unsealed Sources

The use of radioactive solutions and salts results in unsealed radioactive waste, in liquid or solid form. Hospitals and university/research establishments are licensed to discharge water soluble radioactive waste directly to the sewage system. The licence specifies daily discharge limits which are in keeping

with the exemption levels contained in the SI 125 of 2000. However in keeping with ALARA, where possible, radioactive waste arising from radionuclides with a short half-life is segregated and stored securely until it has decayed to near background level. It can then be disposed of in water soluble form to the sewage system. An annual summary report of the total annual amounts of the commonly used nuclides discharged to the sewer is reported by licensees to the RPII.

An indication of the annual amounts of unsealed activity used in Ireland is provided by the annual discharge of radioactivity to the sewers from all licensees. For 2006 this figure was of the order of 15TBq. By far the largest contribution to this is from technetium-99m (14TBq) with tritium (1TBq) being the next largest contributor. The other commonly discharged radionuclides are iodine-131(0.42TBq), sulphur-35(0.66TBq), iodine-125(0.14TBq), Carbon-14(0.06TBq), phosphorous-32(0.04TBq), and chromium-51(0.001TBq).

Hospitals use unsealed radioactive sources for in-vivo and in vitro applications. Contaminated materials such as gloves, syringes, swabs and test tubes/containers arising from the use of the sources are stored in appropriately labelled and shielded containers, and allowed to decay to background level before being disposed of as clinical or bio-hazardous waste. Clinical waste is sterilised, shredded and disposed of to deep landfill. Bio-hazardous waste is exported as incineration is not permitted, at present

Thyroid ablation using iodine-131 must be carried out in a dedicated room designed for the purpose. At present, with the exception of one hospital, excreta from patients undergoing this treatment are discharged directly to the sewage scheme. The question of whether hospitals should fit iodine storage tanks is being addressed by the RPII. A review of the policies adopted in other countries has been undertaken. The findings and recommendations of the review will be considered in the coming months.

The most common radionuclide used in hospitals is technetium-99m. Hospitals are permitted to store an agreed number of spent technetium-99m generators at a secure location on their premises, while awaiting their return to the supplier. In addition it is estimated that there may be up to 7000 old generator cores dating back to the 1970-1980 period stored in hospitals in Ireland. Many of these cores have been removed from the lead generator housings which were melted down for use as shielding blocks and source containers.

A few hundred miscellaneous disused unsealed sources are held in storage by a small number of licensees. These vary in nature from contaminated materials and objects to varying amounts of radionuclides of low activities. These radionuclides include liquid sources of radium-226, caesium-137, carbon-14, and strontium-90 and various amounts of thorium and uranium compounds in solid and liquid forms. The sources are held by licensees such as universities, third level educational institutes and a small number of hospitals. One licensee has solutions of caesium-137(11MBq) and strontium-90(189MBq) salts which are for disposal. These activities are much higher than those for the solutions mentioned above.

One unusual source of radioactive waste comes from 877 unsealed radium-226 sources designed for use with illuminated level bubbles. These sources contain a total of 38.6MBq. The radium is absorbed on paper strips placed behind the levels.

Other Sources of Radioactive Waste

Disused Sources at Post Primary Schools

In late 2001 – early 2002 the quantity of radioactive materials held by post primary schools was assessed as part of a larger project carried out by the RPII in collaboration with the Department of Education and Science. This assessment was based on a survey by questionnaire which was sent to 753 schools. Completed questionnaires were received from 301 schools, 179 of which indicated that

they had radioactive sources. These schools are in possession of 856 sources, 378 of which are classified as disused. This number of sources represents 190 sealed sources and 188 unsealed sources.

Extrapolating the data received to the non-respondent schools, it has been estimated that there are approximately 475 disused sealed sources held by post primary schools. The sources involved include radium-226, strontium-90, americium-241, cobalt-60 and a small number of plutonium-239. Most of the sources have an activity of 185kBq or less with the maximum activity being 370kBq.

Similarly extrapolating the data for unsealed sources it is estimated that there are 470 disused unsealed sources held at post primary schools. These sources are compounds of uranium-238 and thorium-232. Applying a nominal weight of 20g to each of the unsealed sources, a maximum of 9 kg of these compounds is held in storage, mostly in the form of thorium-232 salts.

NORM Waste

The by-products of various industrial processes where NORM wastes could arise have been examined by the RPII. These studies mainly centred on the by-products of electrical power generation from peat and coal burning stations, the processing of bauxite, the off-shore gas industry and the use of thoriated welding rods.

The findings of these studies indicate that the present levels of radioactivity found do not require the implementation of regulatory control from a radiological protection point of view.

Proposals for Future Improvements

The present system of regulation provides for the tracking of sources from the time that they first enter the system to the time of their disposal. However there are a few areas where further improvements can be made to increase source security, reduce the amount of disused sources presently in existence and to minimise the number of such sources requiring long-term storage in the future.

Such areas include:

- The establishment of a national repository for radioactive waste or an agreement with a recognised installation abroad for the disposal of such waste.
- Improving take-back agreements with sealed source suppliers
- Agreeing with all stakeholders on a procedure for the disposal of thorium and uranium salts
- Adopting a procedure for the disposal of old sources which have decayed to below exemption levels

National Radioactive Waste Repository

At present there is no national repository for the storage of disused radioactive sources or orphan sources. For many years the RPII has called for the establishment of such a facility which is seen as a key element of the national infrastructure for the management of radioactive waste. While acknowledging that the issue is complex, the RPII are concerned over the slow progress being made on its establishment [5].

The absence of this facility has caused difficulties in the past. As the RPII has no storage facility for disused sources, it has had to, on occasions, rely on the support of certain licensees for the storage and management of sources which were falling out of regulatory control. This situation is far from ideal.

Many of the disused sealed sources in Table 1 were purchased prior to 1991, when the requirement for a take-back agreement between the source purchaser and the supplier came into effect. While these sources are held in secure storage by the licensees, there may be difficulties with their ultimate disposal. Should such a licensee go into liquidation, or for whatever reason is unable to maintain

security on the sources, the existence of a national repository would provide a temporary secure storage location for these sources, while arrangements are made for their final disposal. In the event that these arrangements cannot be made, the sources could be permanently stored at the repository.

Take-Back Agreements

The policy of requiring licence applicants to enter into a take-back agreement with the supplier of sealed radioactive sources has been operating satisfactorily since it's commencement in 1991. In these agreements, the supplier agrees, prior to the purchase of the source, to accept its return when the licensee no longer has need of it. However these agreements have, in general, only been used in cases where a replacement source is being purchased.

In cases where no replacement source is purchased, the supplier will honour the agreement to accept back the source but there is usually a large financial cost to the licensee. Many licensees may find this financial cost prohibitive, especially where several sources are involved and will therefore continue to store the source under licence from the RPII.

Another problem which could weaken or negate such agreements arises where the source may have been damaged. It is not clear if the supplier will always honour the agreement in such circumstances. In a recent case, the supplier of a level gauge containing a source of 1670MBq americium-214 refused to accept it back as the gauge had been damaged in a fire [6].

Agreements with all stakeholders on the disposal of thorium and uranium salts

The continued storage of these sources, especially at the post primary schools is a concern to the RPII and a method for their safe disposal is being progressed. A procedure for the disposal of these salts has been formulated and consultations are on-going between the RPII and the relevant stakeholders such as the Environmental Protection Agency with regard to the acceptance of the procedure.

A possible option is to dispose of the salts to the sewage system as water soluble solutions of low activity. This disposal would be carried out by competent persons under the approval of the RPII.

Procedure for the disposal of old sources which have decayed to below exemption levels

In Irish law, once a source has been licensed it can only be removed from regulatory control via a licence for its disposal. This has resulted in many disused sources remaining under licence although they have decayed below the exemption level for licensing. The establishment of procedures, and activity levels, for the clearance of these sources from regulatory control would facilitate the removal of these sources held by licensees and provide better source management.

The RPII are advancing proposals on the matter in consultation with the relevant stakeholders. The adoption of the present exemption levels in SI 125 of 2000, or some fraction of them, for the removal of sources from regulatory control is a likely solution, with ultimate disposal of the sources to landfill.

Conclusions

The establishment of a small national repository for sealed disused sources, and for any orphan sources that may be found, would improve the overall security and radiation safety associated with radioactive waste in Ireland. The long-term storage of disused sources by licensees is not satisfactory and the RPII have continued to advise the Government to establish a national repository. However it has proved difficult to resolve this situation.

There are 84 licensees holding 1982 disused sealed sources throughout Ireland with a stated total activity of 1.73 TBq. The majority of these sources have a half-life of greater than 10 years. However, this activity is an overestimate as in many cases the activity provided for the sources relates to the date of manufacture. The sources originate mainly from sources used for industrial applications, emergency lights containing tritium, uranium fuel slugs from a dismantled sub-critical assembly used for teaching purposes and calibration and check sources. Many of these sources were brought into the country prior to the requirement for a take-back agreement with the source supplier. The disposal of these sources will place a large financial burden on the licensees.

In addition it is estimated that there are a further 475 disused sealed sources held by post primary schools.

While the total activity of the radioactive waste is relatively small, there are steps that can be taken to further reduce these levels. At present the RPII is working with the relevant stakeholders to get acceptance on a method for the disposal of thorium and uranium salts, and a proposal for the introduction of activity levels for the clearance of sources from regulatory control. Satisfactory outcomes on both these issues would greatly reduce the number of disused sources in existence.

References

[1] Murnaghan, D. "The Irish Radium Institute, Dublin", Some People and Places in Irish Science and Technology, Royal Irish Academy, 1985, pp.102-103.
[2] The Radiological Protection Act, 1991, Act No 9 of 1991
[3] The Radiological Protection Act, 1991, (Ionising Radiation) Order 2000, (S.I. No. 125 of 2000).
[4] The Radiological Protection Act, 1991, (Control of High Activity Sealed Sources) Order 2005, (S.I. No. 875 of 2005).
[5] Chairman's Statement, Annual Reports and Accounts, 2005 and 2006. Radiological Protection Institute of Ireland.
[6] Duffy, J. and Madden, J, "Analysis of a Radiological Incident (Case No. 22): retrieval of a Fire damaged Gauge Containing a Radioactive Source in Ireland" European ALARA Newsletter, Issue 21. October 2007.

Table 1 Disused Sealed Sources

Radionuclide	No. of Sources	Total Activity GBq	Half-life
Hydrogen-3	36	598.48	12.26 y
Americium-241/Be	15	225.00	432 y
Strontium-90	65	194.04	29 y
Krypton-85	9	126.75	10.7 y
Caesium-137	72	117.84	30 y
Americium-241	58	87.90	432 y
Plutonium-238	1	92.50	87.8 y
Uranium-238	1404	63.55	$4.47.\ 10^{10}$ y
Nickel-63	56	24.68	96 y
Curium-244	3	11.14	18.1 y
Samarium-151	3	7.77	90 y
Radium-226	41	1.46	1600 y
Carbon-14	67	0.16	5730 y
Totals	**1830**	**1551 GBq**	
Co-60	40	52.56	5.27 y
Ba-133	2	0.34	7.2 y
Pm-147	6	37.06	2.62 y
Fe-55	2	2.03	2.7 y
Tl-204	11	0.751	3.8 y
Cd-109	3	0.677	464 d
Gd-153	3	74	242 d
Co-57	77	14.53	271 d
Sn-119	6	0.8	245 d
Ge-68	2	0.4	288 d
Totals	**152**	**184 GBq**	

Table 2 High Activity Sealed Sources

Licensee Category	Radionuclide	Activity, GBq
Educational	Strontium-90	11.1
	Strontium-90	140.6
	Americium/Beryllium	185
	Caesium-137	50
	Cobalt-60	11.1
Industrial	Strontium-90	11.1
	Strontium-90	29.6

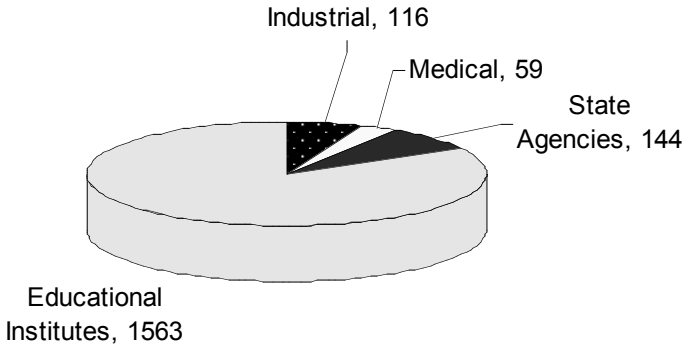

Figure 1

Number of disused sealed sources with a half-life of > 5 years by user category