Management of Disused Sealed Sources in Hungary – 13077

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ABSTRACT

Since 1976 the spent and disused radioactive sources arisen in Hungary are stored in a central storage facility called Radioactive Waste Treatment and Disposal Facility operated by Public Limited Company for Radioactive Waste Management. The Facility is responsible for the record keeping, the waste acceptance procedure, the shipment and the storage or disposal (whether a certain source meets the waste acceptance criteria for disposal or not) of sources. Based on the more than 35 year old operation of the facility many experiences have been gathered regarding the technology for long-term storage of sources, the attitude of the users of sources, the evolution of the legislation and the national record keeping system. Recently a new legislation for the security of radioactive materials (including sources) was introduced, first in Central-Europe. It requires special security arrangements from the facility for transport and for storage. Due to the ongoing retrieval of radioactive waste formerly disposed of, partly containing sealed sources, there is a new challenge in the physical inventory control of historical waste.

The paper would show the effect of the changes in the legislation system of record keeping or security on the users' attitude for discard of sources and on the management of the sources in the facility.

The facility has a unique storage technology (shallow boreholes) in the narrow region. The sealed sources are placed into vertical pipes sunk into the surface. In the beginning, each of the sources were dropped into the pipe directly, recently they are placed in a metal tube first ensuring the retrieval. The lessons learned will be presented.

There were several issues to introduce the new security arrangements (partly financially supported by US DOE) for storage and for transportation of sealed sources. These issues are addressed.

In the past part of the sealed sources were disposed together with solid radioactive waste packaged in plastic bags. A waste retrieval campaign was fulfilled in 2008 to retrieve the sealed sources. The paper demonstrates the conditions of sealed sources after twenty-year disposal period.

As a summary, the paper will share the main experiences of a 35-year old facility, managing radioactive sealed sources in Central Europe.

INTRODUCTION - HISTORY OF THE MANAGEMENT OF DISUSED SOURCES

The application of radioisotopes has begun in 1954 in Hungary. To serve the needs for the radioisotopes and for research activities a research reactor was commissioned in 1959. But most of the sealed sources were purchased abroad, mainly from SU. The radioactive isotopes and the sealed radioactive sources were used widely in research, education, industry, diagnosis and therapy, functional products and so on.

The license for the application of sources was issued by the National Health Service. The field of the license was the application and the storage at the owner's site. The license contained the physical protection rules prescribed by the police who was a subauthority in the licensing process. Separate license was needed for the transport of the sources.

The national register of the sources was operated by the „Isotop Institute" of the Hungarian Academy of Science. Unfortunately the content of licenses issued for the application and the physical inventory of registered sources was not closely linked, therefore there was some uncertainty in the registry.

Until 1990 the disused sources didn't become waste when the user had finished the use of it. First the national register looked over the possibility of the application of the source by another user. To give the source over the waste disposal facility the resignation statement of the national register was needed.

The first repository was operated by the National Health Service at Solymar village, in the neighborhood of the capital Budapest, in the period of 1960-1977. The solid waste packaged in plastic bags was stored in wells made of prefabricated concrete rings. The disused sealed sources were stored in shallow boreholes: large diameter steel pipes (wells) sunk horizontally into the ground. The sources was dropped into the pipes, the possibility of retrieval was not solved. The site proved unsuitable for final disposal or long term storage due to the level of groundwater table.

The new repository at Püspökszilagy village was commissioned in 1976, and the Solymar site was remediated and all the waste was shipped to the new site. Similarly to the Russian "radon" type facilities the solid waste packaged in plastic bags (later metal drums) was stored in concrete vaults, the disused sealed sources were stored in steel wells sunk vertically into the ground.

LEGAL FRAMEWORK AND RECORD KEEPING

The license for the applications and onsite storage of the sources is issued by the radiation health department of the regional governmental agencies (formerly National Health Service). The security measures regarding the application are licensed in separate procedure; the licensing authority is the Hungarian Atomic Energy Authority (HAEA), the relevant department of the Police is the sub authority. License is also needed to give up the application of the radioisotopes licensed before.

The Institute of Isotopes maintains a centralized registry of radioactive materials at the Hungarian level. This task has been assigned to the Institute by the HAEA. The HAEA revised the whole inventory (2003-2005), most of the local inventories were inspected and compared to the content of the licenses, and only a very small uncertainty remained in the national register. 5745 sources were recorded in the national inventory at the end of 2012.

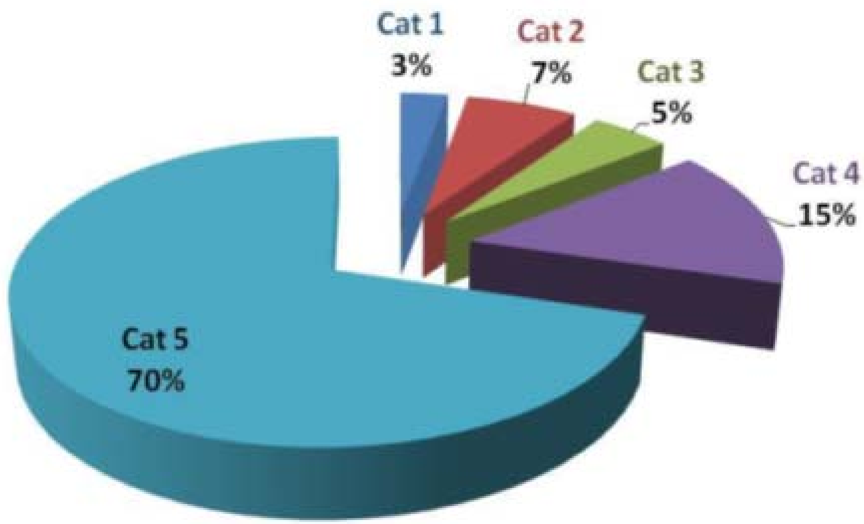
There is no more resignation statement, the reuse of the sources does not happen frequently. The local and the centralized record keeping are based on the software provided by the HAEA for the users. Only those sources have to be registered of which original activity is above the exemption level. If a source is declared to be a disused source, the owner has to take over it to the repository and the HAEA's certificate has to be sent to the HAEA. In the centralized inventory the repository is a special owner of the sources. Every change in the owner's/user's local inventory of sealed sources has to be notified in 15 days.

The local and national inventory of the sources containing the following data [1]:

- the unique number of the certificate issued by the HAEA of the source
- the number of the test certificate issued by the producer/distributor
- owner's data
- user's data if different from the owner
- total licensed activity by isotopes
- number of the license issued by the radiation health agency
- radionuclide
- activity and date of the activity
- function
- serial number
- physical and chemical form
- photo on the high activity source and on its holder
- the goal, the time and the place of application
- the name of the person responsible for the radiation protection measures

In 2012 a new decree on security of radioactive materials (has been come into force, which is also relevant to the sealed sources. The sources are categorized into 5 categories (Diagram 1) based on the ratio of the total activities used/stored/transported and the D-value of the radioisotopes. For each category different security levels (A, B, C and D) has to be applied. The security measures are different for transport and storage at each level. The main philosophy of the measures is the gradual, multilevel defense and the 4 security processes: the prevention/deterrence, the detection, the retention and the response. The decree initiated an avalanche of delivering of radioactive materials to the repository: the yearly amount of the shipments made by the repository was doubled. The issue is that the measures presented in the decree seem very rigorous. Although the decree allows differing from the prescribed measures by the Police's approval, some requirements make the users uncertain how to fulfill them.

Diagram 1 – Distribution of sources by the security categories [2]



Challenges concerning to fulfill the security measures:

- More severe requirements comparing to the prescriptions of HASS directive [3]
- Equipment of the transport vehicle with unique locks, detection devices
- Equipment of the storage with unique locks, detection devices, additional barriers
- Physical training of the guards

INVENTORY OF SEALED SOURCES IN HUNGARY

Diagram 2 and 3 show the distribution of the number and activity of sealed sources by function and by radionuclides [4]

Diagram 2 – Distribution of sources by functions

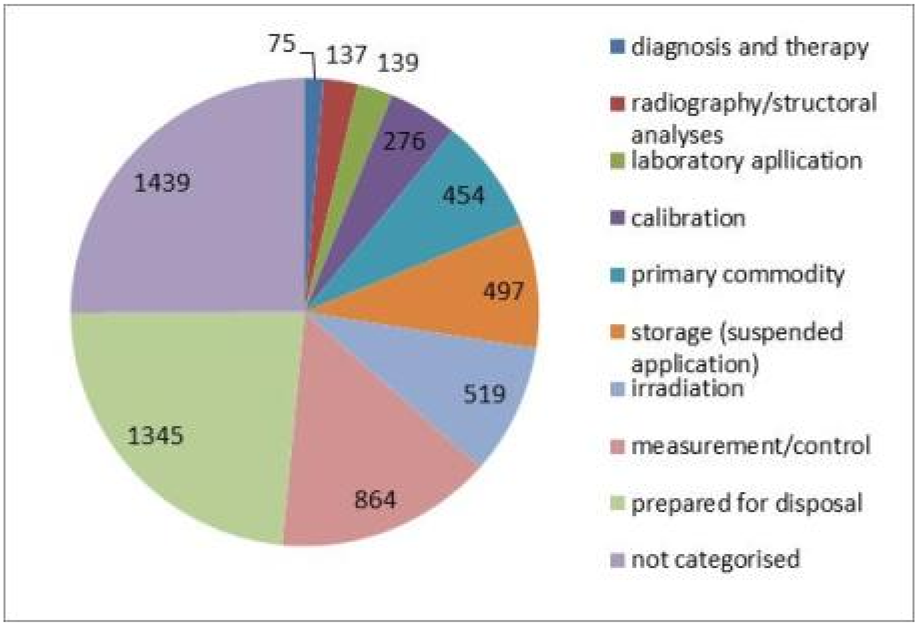
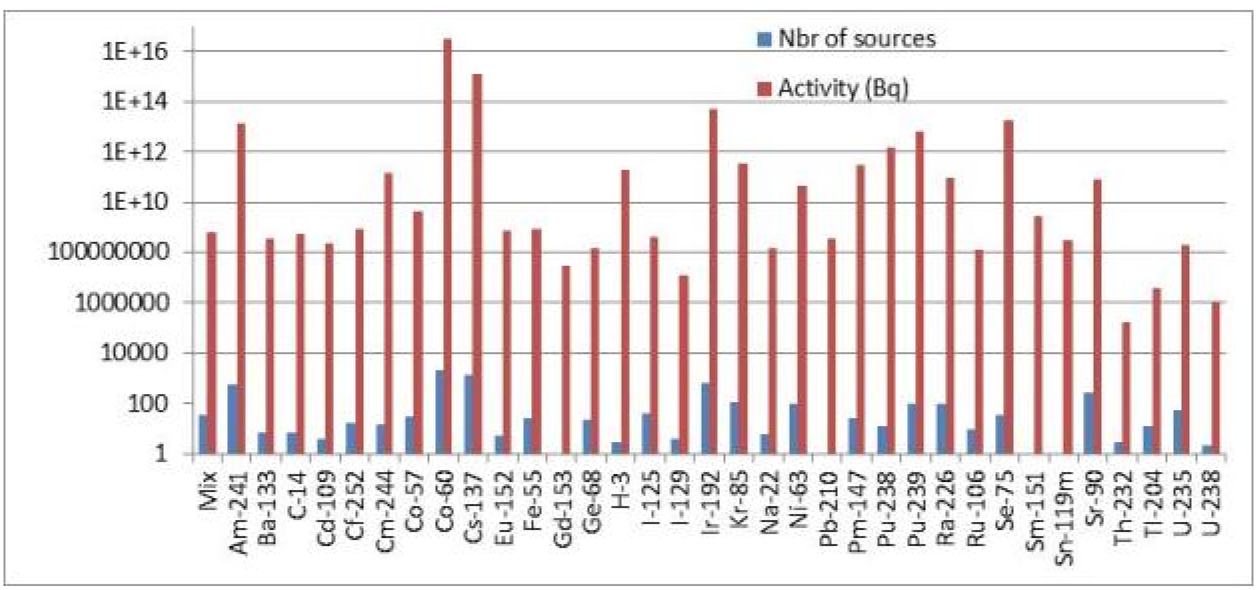


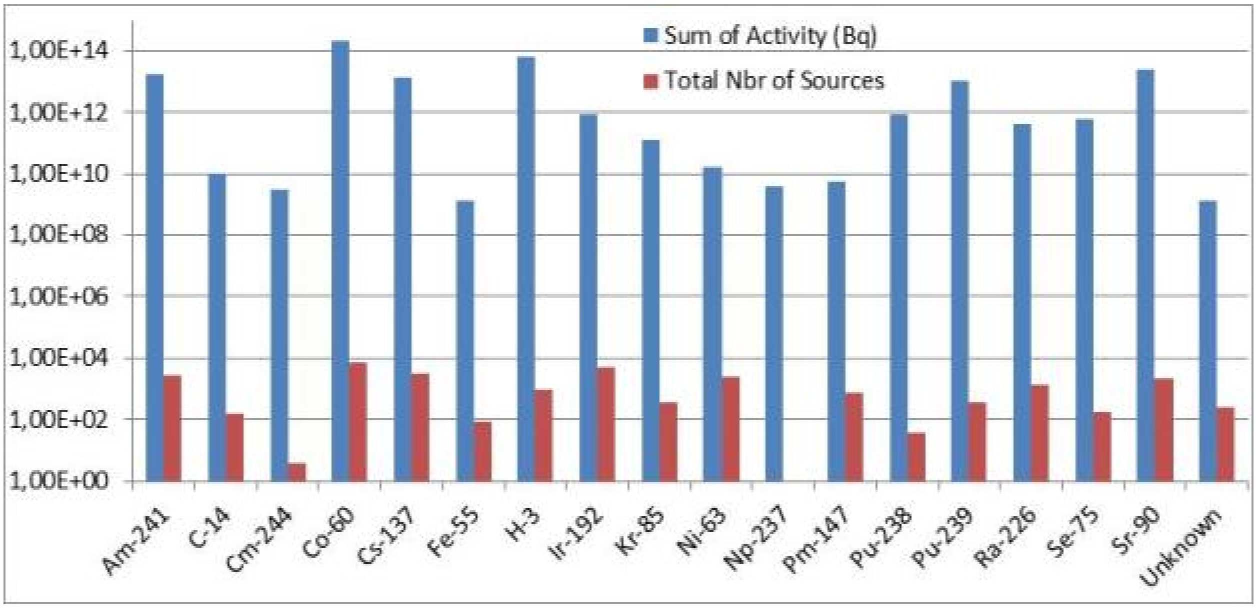
Diagram 2 – Distribution of sources by radionuclides



Remarks: Radionuclides in case of the total activity of the sources exceeds 1 MBq are shown. The neutron emitter sources are not presented separately.

The inventory of disused sealed sources stored in the RWTDF is shown in Diagram 4.

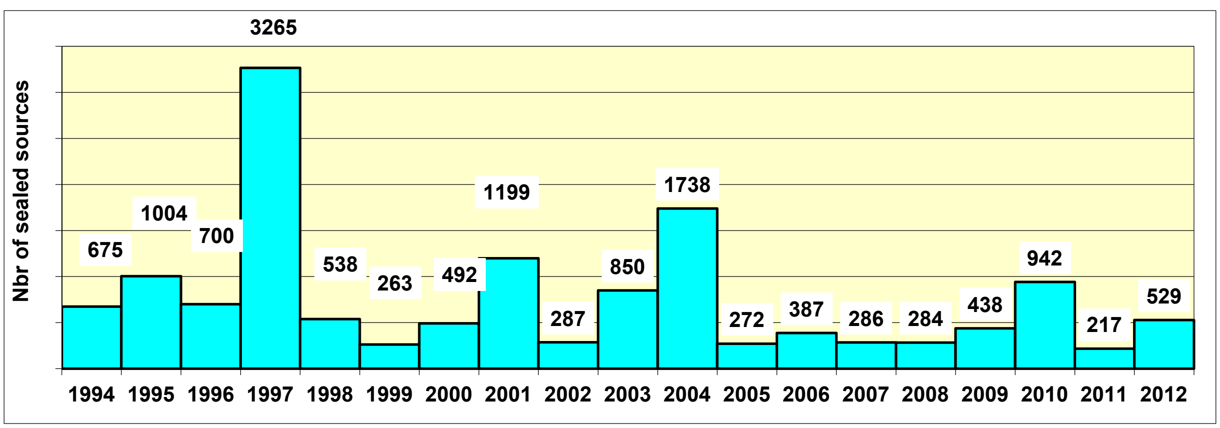
Diagram 4 - Inventory of disused sealed sources stored in the RWTDF in 2012



Remarks: Radionuclides in case of the total activity of the sources exceeds 1 GBq are shown. The neutron emitter sources are not presented separately. The ^{3}H sources are tritium-targets, and its activity (over)estimated.

The tendency in the yearly amount of disused sealed sources disposed is shown in Diagram 5.

Diagram 5 - Yearly amount of disused sealed sources in the past



Evaluating the Diagram 5 it can be shown the changes in the users attitude using and discarding the sources:

- In the second half of 1990s many enterprise gave up its activity, after that time the volume of the application of the sources and the delivery of the disused sources to the repository decreased.

- when the ministerial decree on local and national record keeping of radioactive materials came into force in 2004 many users delivered their unused open and sealed sources to the repository
- when the update of ministerial decree on local and national record keeping of radioactive materials came into force in 2010 many users delivered their unused open and sealed sources to the repository
- when the ministerial decree on security of radioactive materials came into force in 2012 many users delivered their unused open and sealed sources to the repository

RADIOACTIVE WASTE TREATMENT AND DISPOPSAL FACILITY

Overall description of the facility

The spent and disused radioactive sources arisen in Hungary are stored in a central storage and disposal facility called Radioactive Waste Treatment and Disposal Facility (Image 1) and commissioned in 1976. In the past the site was operated by the licensing body (National Health Service). Since 1998 the site has been operated by the Public Limited Company for Radioactive Waste Management, which is a governmental agency. The Facility is responsible for the record keeping, the waste acceptance procedure, the shipment and the storage or disposal (whether a certain waste meets the waste acceptance criteria for disposal or not) of all radioactive waste except nuclear power plant. The total volume of short-lived LIL solid waste disposed of in concrete vaults is around 5 000 m^3.

The facility includes near-surface disposal vaults, and shallow bore-holes for interim storage.

The original layout of the facility showed 4 units (Image 2):

- “A” vaults: 70 m^3 vaults for final disposal of solid waste
- “B” wells: small diameter shallow boreholes for storage of DSS
- “C” vaults: 1,5 m^3 vaults for storage of solidified organic solvents packaged in gas cans
- “D” wells: large diameter shallow boreholes for storage of DSS

The long lived LILW and the sealed sources taken over after 2005 are stored in the interim storage located in the cellar of waste processing building (Image 3).

Image 1 - The RWTDF site

Image 2 –The disposal units and storage wells

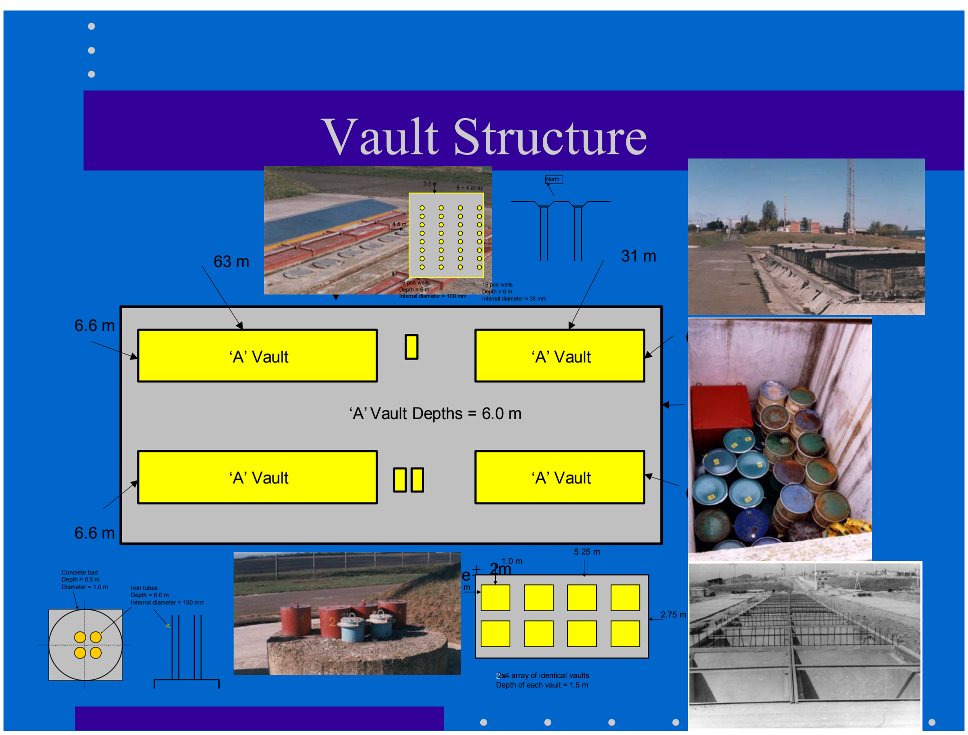


Image 3 – The interim storage rooms and storage wells

The waste acceptance process of disused sealed sources

- The user fills out the data sheet
- The repository officer checks the compulsory data: serial number, number of certificate, etc.
- In case of incorrect data: cross check with the user and/or the national register
- Managing the shipment (by the repository staff or by the user, mainly exempted, A or B type packages)
- User: inform the national registry in 15 days
- Repository: inform the national registry in 15 days
- In case of Plutonium and Uranium sources: safeguard processes are followed as well

Conditioning of disused sealed sources

First the sealed sources are collected in a temporarily storage room, sorted by type and radionuclide. In a hot cell (Image 4) the sources are encapsulated into a metal tube (diameter 40, 100 or 200 mm) sealed at the bottom. After filling the tube, the top of it is sealed by welding along the periphery of the tube (Image 5). In case of neutron emitters additional neutron shielding is placed inside.

If the sealed source could not be placed into the tube due to its dimension or to the non-demountable device size, then the source is embedded in cement matrix in the center line of a 200 l drum.

Most of the cases there have not been any contamination of sealed sources even if their service period has expired.

Image 4 – Hot cell

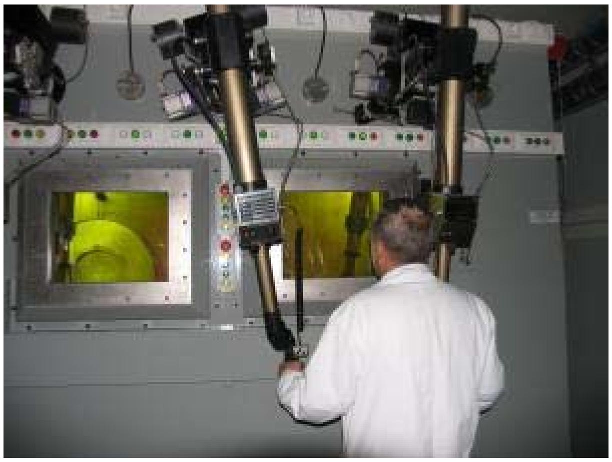


Image 5 - Encapsulation

Storage of disused sealed sources

The disused sealed sources were designed to place in the "B" and "D" wells. The steel pipes sunk horizontally into the ground. The depth of the wells is 6 m. For the storage of short lived nuclide the "B" type wells made of stainless steel consist of 16 pcs wells with diameter of 40 mm and 16 pcs wells with diameter of 100 mm each of 6 n deep. For the storage of long lived nuclides the "D" type wells made of carbon steel consist of 4 wells with diameter of 200 mm. The sources was generally dropped into the pipes without the possibility of the retrieval separately but the "B" type wells accommodate the byproducts of ^{60}Co-sources and high activity ^{192}Ir-sources and the "D" type wells accommodate the ^{226}Ra-needles sealed into retrievable metal tubes. The "D" type wells are filled completely and the "B" type wells are half empty.

Table 1 - Free capacity and Activity limits of the wells

Unit	***Activity* (Bq)***	***Remark***
Storage wells inside the interim storage building	1,30e+13	16 wells under filling 34 wells empty
D-type storage wells (shallow boreholes)	5,15e+12	4 wells completely filled
B-type storage wells shallow boreholes)	2,28e+14	5 wells under filling 12 wells completely filled 15 wells empty
Nuclear storage	9,34e+12	plutonium sources in B(U) type packages
Hot cell	1,15e+12	sources waiting for encapsulation

*12 Dec 2012

At the beginning the function of the wells was similar than the Russian “radon” type shallow bore-hole system for spent sources [5], but the concept was more simple. Hence the first safety assessment made in 2000 stated it is not suitable for disposal only for storage.

In 2004 the treatment building was completely renovated. A hot cell was installed to enclose the sealed sources into an metal tube and at the basement of the building an interim storage area was evolved. The interim storage composed 4 units: two room for storage drummed solid waste, one storage room for nuclear materials and a well-type storage for disused sources (Image 3). The Interim Storage Wells system composed of 10 pcs of 40 mm, 20 pcs of 100 mm and 20 pcs of 200 mm stainless steel pipes, each of 4 m deep (Image 6).

Image 6 – Presentation of emplacement of sources

In the last 7 years mainly the Interim Storage Wells are used for emplacement, the “B” type wells are reserved for byproducts of ^{60}Co-sources, high activity ^{192}Ir and other short lived sources.

The ^{239}Pu and ^{239}Pu-Be sources are stored in special containers in a separate room.

The license for storage of sealed sources and the waste acceptance criteria determine the total activities and the maximum activity per source in the different well-type units.

Lessons Learned

The environmental monitoring has not shown any leakage form the wells until now (36 year period)

The ambient dose rate measured above the storage wells is low, below 20 μSv/h.

The Interim Storage Wells were designed on the basis of pipe-in-pipe concept: the cylinder shaped retrievable metal tube containing the sealed sources is placed the cylinder shaped storage pipe with larger diameter. The pipes used for the wells and for the tube have standard diameters. Regarding the nominal 200 mm well, the diameter of the storage well is 206 mm, the diameter of the tube would be 198 mm. The problem is that the pipes with diameter of 198 mm are very rarely and only in large quantity produced by the metal works, the repository requires only 1 m long pipes per year. The first attempt to solve the issue was to create a 198 mm pipe from the 206 mm pipe cutting out a stripe from the cylinder and curve the cylinder to the preferable shape. Unfortunately the shape could not be perfectly symmetric circle, and the semi-automatic welding device could not weld properly the lid of the tube. The second attempt was to use 168 mm standard pipes instead of 198 mm pipes. In that case spacer wings have to be used ensuring that the metal tube do not stand diagonally in the larger storage well. At present we apply the second method.

The hermetical tightness is not required until now but in case of tritium and other gas form radionuclides.

The free space is limited in the hot cell, so the hermetic test is hardly feasible for each tube. On the other hand it is questioned, what to do with the welded tube if the hermetic test failed. That is why we investigate the technical solution to apply a screw thread releasable connection instead of welding of the lid of the tube.

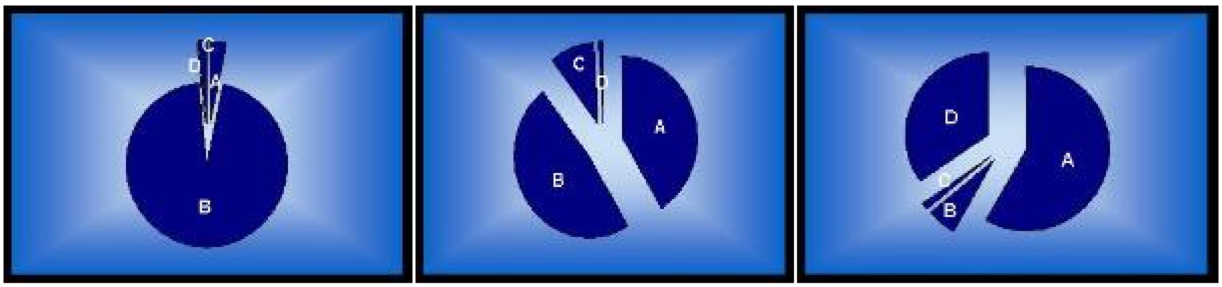
Although the hot cell serves only to enclose the sources into a tube, there were suggestions by Belgian experts to enhance the tightness and the design of the hot cell.

SAFETY ENHANCEMENT OF THE REPOSITORY

In the past some of the disused sources packaged together with solid waste were placed into the vaults instead of the wells due to its size or conditioned form (Diagram 6). The wells of Solymar site containing sources were placed also there. The waste placed into the vaults contained long lived waste as well.

Diagram 6 - Distribution of the activity of sealed sources in the storage/disposal units

< 5 years **5-30 years** **> 30 years**



The 2002 safety assessment recommendations to enhance the safety of the site

- Long lived DSRS in the 'B' and 'D' wells should be removed before the closure of site.
- Large activity ^{137}Cs sources and long lived sources should be recovered from 'A' vaults
- All vaults should be backfilled to provide chemical conditioning
- The waste packaged in plastic bags should be repackaged and compacted into drums
- The inventory should be revised.
- Waste acceptance requirements in the future:
    - The disposal of long lived radionuclides is no permitted.
    - The long lived waste accepted should be accepted for temporary storage pending final disposal in geological repository.
    - The limits of concentration of long lived components are suggested by the SA.

Meanwhile the site had run out of the free capacity for disposal of solid waste. Therefore there was a strong economic pressure to open the vaults, to remove all the waste not backfilled, to separate the sources and the ling lived waste from the short lived waste and to place them into interim storage, and to compact the short lived waste before replacing. After removal the long lived waste and compaction the short lived waste, about 20 % of the volume of the vaults could become free.

In 2007-2009 a demonstration phases was carried out to prove we are able to fulfill the work and to achieve the aim of the project. 4 vaults (nominal volume is 280 m^3) were opened and 220 m^3 volume of waste was retrieved, segregated and repackaged.

In the demonstration phase all items which could have been or could have include a „sealed" sources was separated: capsules, tin boxes and cans, holders, symmetric metal scraps, items with significant surface dose rate. These items were clearly identifiable and separable (Image 7, 8, 9).

Image 7 – Retrieved holders of disused sealed sources

Image 8 – Retrieved source-holder packaged in plastic bag in the past

Image 9 – Segregated disused sources

The conditioned packages (cans backfilled with bitumen, drums backfilled with cement mortar) were not opened, and therefore the numbers of the sources contained are only estimated.

Regarding the content of one vault there were big differences in the number of the sources recorded and finally identified. But after summarizing the content of 4 vaults the difference between the record and the physical inventory decreased: we were looking for 482 pcs of sealed sources and finally found 498 pcs of sources (Table 2). The principal question is unanswered: what items were recorded as sealed sources in the past and what items were not? The categorization of the tritium targets as sealed sources and recording the irradiated activity of them must be reconsidered as well.

Table 2: The forecasted and the identified sources

Isotope	Activity of sources in the inventory (Bq)	Activity of sources found (Bq)	Number of sources in the inventory	Number of sources found
^{226}Ra	~2E+10	1,20E+10	14	11
^{60}Co	~3E+09	2,23E+07	53	158
^{137}Cs	~5E+09	7,20E+09	32	40
$^{90}Sr/^{90}Y$	~3E+09	2,26E+06	21	39
^{3}H	~3,5E+13	1,40E+12	362	227

Positive experience was that there was not fund any sources without packaging. All sources were placed in some can, box or other holder inside the plastic bags.

There was no measurable significant contamination of these holders.

In the inventory there were recorded other short lived nuclides like ^{51}Cr, ^{55}Fe, ^{59}Fe, ^{65}Zn, ^{109}Cd, ^{144}Ce, ^{185}W and ^{192}Ir. We suppose these sources were decayed and they possibly were in the drums backfilled with cement mortar.

The next phase of safety enhancement project is going to start 2014 after we has got all licenses and install a steel frame hall above 24 vaults to retrieve and repackage about 1400 m^3 waste.

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