Feedback of the Dutch Historical Waste Management Program-17085

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ABSTRACT

This paper provides a feedback on the Dutch historical waste management program. Over the past decades, the High Flux Reactor located in Petten in the province of North Holland has tested numerous types of materials and fuels irradiation, for both medical and nuclear energy applications. A number of waste containers was accumulated on site, where the storage facility was initially built. Following Dutch political decisions in the 1980's, the contents of the waste storage facility in Petten must be transferred to another location. The consequences of this decision included the transportation of waste containers from Petten to the new location, which is located in the South-West of the Netherlands.

This project was initiated and is now running. It includes a complex structure and can be described as follows: each historical-waste container will be opened, its contents segregated in three categories depending on the dose rate emission, characterized and repacked in new containers suitable for transportation and storage at the new site. The approach chosen is based on the final dose rate emitted by the repacked containers, which are sub-divided into3 categories.. The lowest dose-emitting containers are transported directly to the new storage facility. The 2 other categories are first transported to a third party to be super compacted and subsequently transported to the new storage facility.
So far, the segregation method is based on Co-60 gamma emission and nuclide vectors associated to the different materials. A pre-sorting has been established to fit as much as possible IAEA recommendations. Historical waste has then to be classified in families sharing common history. Thus, a set of canisters containing the pieces of the old reactor vessel, which was replaced in 1984 was the first challenge, followed by a set of canisters containing residues of instruments only and pieces coming from the top of the control rods of the reactor which were not directly irradiated in the reactor.
So far, the two first sets of canisters are being treated and characterized as well as possible. Reporting and official documents have been filled up to comply to Dutch transport regulations and to define waste acceptance criteria for the storage facility. The first transports of containers with Low Level Waste occurred in October 2015, and another transport took place at the end of 2016.

INTRODUCTION

Since the commissioning over fifty-five years ago of the High Flux Reactor, used for isotope production for medical purposes as well as material research, in Petten, Netherlands, radioactive waste has been produced. That waste has been stored for

many years in the dedicated surface storage facility at ECN/NRG's research site: the Waste Storage Facility (WSF).

In its 'Memorandum on Radioactive Waste', the Dutch government stipulated that from 1984 all the waste produced in the Netherlands must be collected, processed and stored by a central organization. The Central Organization for Radioactive Waste (COVRA) was established for that purpose. Until the early 1990s, COVRA was based in Petten, with the WSF serving as 'national storage facility'. Thereafter COVRA moved to Nieuwdorp (Zeeland).

Although already hundreds of canisters containing historical waste of low and medium activity were transferred from the WSF to the COVRA facilities, the WSF still stores low and intermediate level waste, fissile material containing waste generated during experiments, as well as operational waste that must be disposed of once it is sufficiently decayed. The intention was to remove these wastes from the WSF at Petten once COVRA had a special facility ready for it. This was the case in September 2003 when the High Activity Treatment and Storage Building (HABOG) was opened at COVRA. Thereafter, NRG started the Radioactive Waste Project (RAP).

THE RAP PROJECT

Quickly after the start of the RAP project, it was recognized that the storage containers holding waste in the WSF did not comply with the packaging requirements for transport to and storage at COVRA. Therefore, a consensus between COVRA and NRG was established around a treatment implying a segregation of the contents of the historic waste drums per activity and content.

In practice, RAP now consists of the following elements:

- Developing equipment for sorting, separation, characterizing and packaging of the historic waste to facilitate transportation and guarantee waste acceptance at COVRA;
- Adapting existing laboratories at Petten to implement the project safely and responsibly
- Arranging safe transport in containers specially developed and certified for the purpose. The 'Intermediate Level' waste is cemented and compacted in Belgium to make it suitable for storage in the HABOG facility at COVRA.

The general approach of NRG concerning waste treatment and waste management can be summarized as outlined in Figure 1. This approach is, when dealing with historical waste, difficult to implement in retrospect. However, because of economic, environmental and societal reasons, the minimisation of the quantity of high intermediate level waste (ILW-H) and low intermediate level waste (ILW-L) by sorting and extracting low level waste, is one of the highest priorities of NRG.

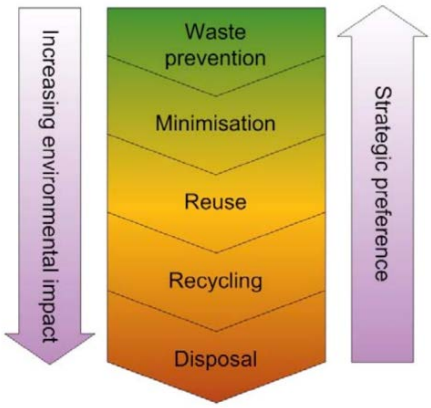

Figure 1: NRG waste management approach

To build the safety case required by the Dutch regulator and COVRA, information has been retrieved from the archives and coupled with newly calculated nuclide vectors and high resolution gamma measurements. As the waste streams considered consist of solid material, homogeneity and representability of measurement/sampling could be challenged. The current strategy includes the sorting of the historical waste canister from the WSF at the Petten site into 3 categories of waste, primarily according to their respective activities: Low level and two intermediate levels activity ("Intermediate low" and "intermediate high"). Each category will follow specific routes (direct storage or 'supercompaction', cementation and storage). The resulting challenge for the Petten site lies in the process steps that consists of segregating, sorting, characterizing and packaging each vessel.

Following IAEA recommendations [IAEA-TECDOC-1537] smaller waste streams were distinguished in what will be later called later "waste families": A waste family is represented by a particular waste stream with similar characteristics, e.g. identified materials with a known irradiation history/cooling time and contamination. The simplified processed is presented in the figure below.

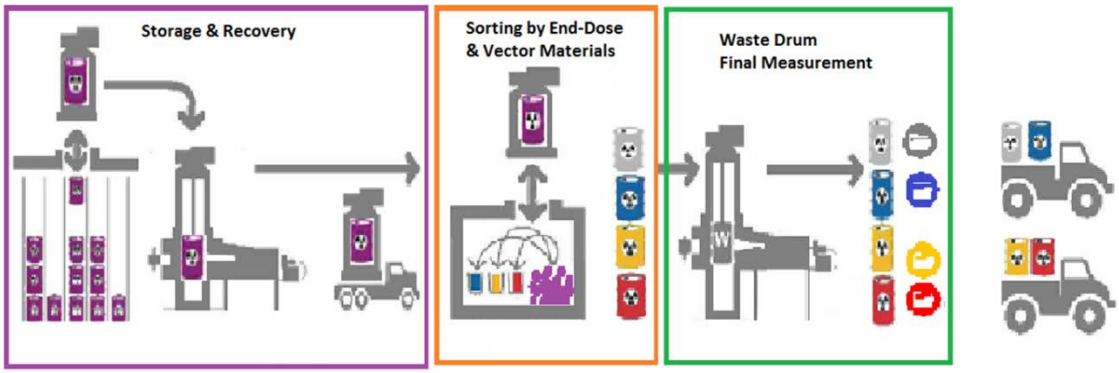

Figure 2: Simplified representation of the process

Figure 2 shows a simplified version of the a more complex process as it only clarifies the concept of sorting and the general path.
A comparison of the waste stored in the old WSF and the new situation is shown in Figure 3.

Figure 3: Waste storage facility, under construction (left) and nowadays (right)

The logistic around the recovering of the old canisters and the subsequent storage of the newly produced waste canister waiting for transportation and storage represents a complex issue and will not be discussed in this article. Still the full representation of the process is depicted in figure below.

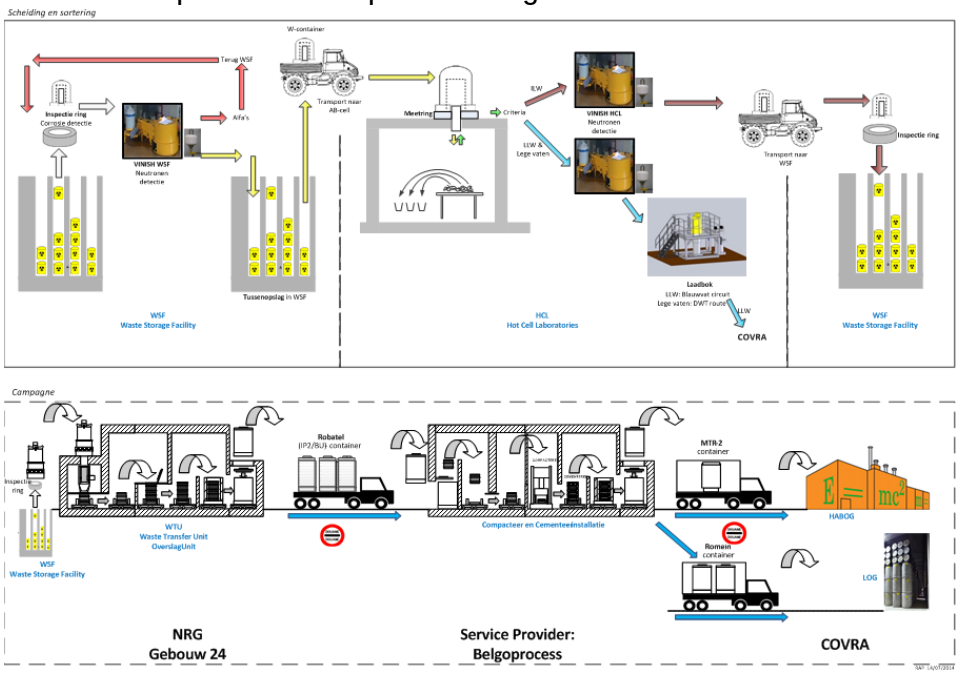

Figure 4: Overall representation of the RAP process

Over the past months the waste sorting of the first waste family was performed, representing canisters containing the old reactor vessel which was dismantled and replaced in 1984. The foreseen development of the project is to proceed from simpler waste families to more complex and contaminated waste families, which may contain leftovers of fuel or materials which have not been documented in the past. This way NRG will gain knowledge about the containers' sometimes complex contents, manage the further breakdown of waste streams, and further reduce the footprint of the historic radioactive waste.

TOOLS DEVELOPED FOR THE SEGREGATION AND THE CHARACTERIZATION

- **VINISH[2]**

VINISH stands for Visual Inspection and Nuclide Identification System for High level radioactive waste, which is essentially a nuclide specific gamma scanning device. Two of these devices, shown in Figure 5, have been built.

During the scanning procedure, the historic waste canister is vertically rotated to average out the asymmetrical packing of the waste in the drum. Subsequently, "slices" of the canister (around 25% of the height) are measured by moving the vessels vertically, while the detector position being fixed. Gamma-ray energies in the range 100keV to 2 MeV can be measured by the high purity germanium detector.
Even though issues have been encountered with high dose vessels, VINISH has been built to handle activities up to 10 TBq Co-60. This characteristic is not met yet and some adjustments are still necessary.
It is also used after the segregated historic waste has been packed into the new vessels which are ready for transport, for drawing up the list of nuclides and their activities that are (legally) required for transport and storage.

Figure 5: VINISH: High resolution gamma spectrometer

- **HIRARCHI[3]**

HIRARCHI stands for High Radioactive Raw waste Characterization and Identification. It is a sorting system consisting of a gamma detector with a movable camera in the horizontal plane and a vertically movable platform. The system is emplaced in one of NRG's hot cell facilities. The historic waste from the containers is spread over the platform, inside a shedding ring: an oval ring of metallic sheet that keeps small waste parts from rolling and sliding away. This area is fully scanned by the movable detector, Co-60 response of the waste is then translated into "isoplots" and colored depending on the waste category.

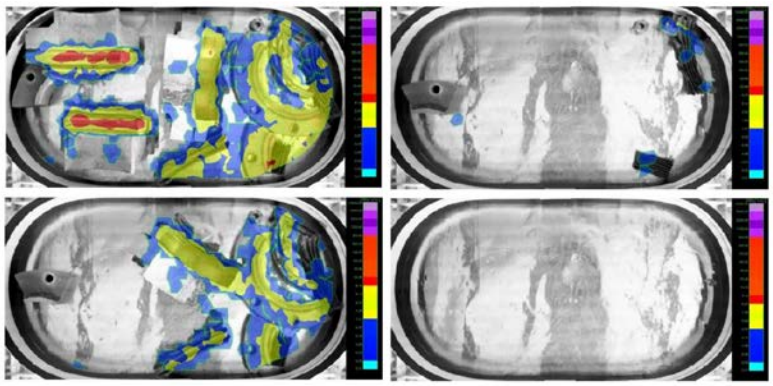

Figure 6: Example of isoplots obtain by HIRARCHI sorting system

- **COMPUTATIONAL NUCLIDE VECTORS**

A number of pure beta or pure alpha emitters, such as Ni-63 and Fe-55, are not integrated in the nuclide content coming from the VINISH. For the first two waste families, a satisfying approach was chosen using computational nuclide vectors. Based on original material chemical compositions, material irradiation conditions and cooling times, and using the FISPACT activation code, computational evaluations of nuclide contents can be obtained. Some results are given in the following sections.
As mentioned above, Co-60 is the key nuclide to evaluate the nuclide content of the historic waste. Results are then given and applied as a ratio compared to Co-60 content.
As any calculation, input data are of prime importance and specifically the original compositions of the historic waste. For instance, the presence of traces of cobalt as an impurity will result in significant ratio differences with other isotopes and then ultimately lead to different nuclide contents and important volatility in the reported values for those nuclides.

So far, the calculation carried are performed with the maximum constraint regards to their output, giving conservative estimation.

THE FIRST SET OF CANISTERS: FAMILY ONE, OLD REACTOR VESSEL

After careful archive investigation, it was concluded that two materials only were present inside the canisters containing the bits and pieces of the old reactor vessel. This vessel has been replaced in 1984. The old vessel was constructed of aluminum and the bolts and screws and thermal shield outside the core box some parts were made of stainless steel. The composition of the stainless steel and of the aluminum were found in the archives which allowed an accurate calculation of the nuclide vectors.
The main issue when facing the old part of the reactor vessel is the dependence on its position with respect to neutron beams in the HFR. To assess this a conservative approach was taken based on the sorting process, i.e. the Co-60 emission. The results of this approach are provided in the table below, showing scaling activity ratio to Co-60, due to activations.

Aluminium

Nuclide	Ratio to Co-60
Co-60	1
Fe-55	248
Ni-63	232
H-3	35
Al-26	3.7 E-02
Mn-53	1.7 E-05
Na-22	1.9 E-05
Co-60m	7.4 E-06
Fe-60	7.4 E-06
P-32	4.0 E-07
Si-32	4.0 E-07
Mn-54	3.6 E-08
C-14	5.6 E-08
Ni-59	6.0 E-10
Zn-65	1.1 E-11
Cl-36	7.1 E-13
Se-79	8.4 E-15
V-50	2.9 E-23

Stainless Steel

Nuclide	Ratio to Co-60
Co-60	1
Ni-59	139
Fe-55	132
Ni-63	1.3 E+04
H-3	2.8 E-03
C-14	4.2 E-04
Mn-53	1.2 E-05
Cl-36	2.3 E-06
Co-60m	5.0 E-06
Fe-60	5.0 E-06
Mn-54	5.6 E-08
P-32	8.6 E-08
Si-32	8.6 E-08
Ar-39	2.5 E-09
Al-26	6.2 E-10
K-42	1.1 E-11
Ar-42	1.1 E-11
V-50	4.2 E-14
V-49	5.5 E-14
Ca-41	1.6 E-15
K-40	9.0 E-20

Mix stainless steel and aluminium:

In case of the presence of a mixed material or non-identified material, the `worst case scenario` is applied by calculating the highest activation of both materials. For each nuclides the highest ratio to Co-60 is taken creating a hybrid vector. This nuclide vector does not represent a specific material then but the conservatism applied allow a safe assumption of the nuclide content.

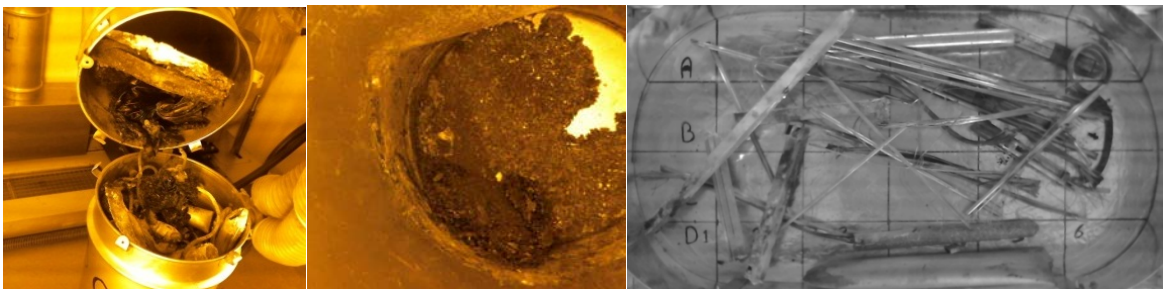

Figure 7: Content of a historical waste vessel from the first set of canisters disposed on the segregation table inside a hot cell

THE SECOND SET OF CANISTERS: THERMOCOUPLES AND RESIDUES OF CONTROL RODS

A similar approach was chosen to characterize the second set of canisters containing not two but four different materials clearly identified. An archive research revealed the composition of the different materials, which is given on Figure 8.

Wastes were produced at different point in time and were irradiated from different positions in the reactor and not from the same period of time. The choice was then made to run several calculation to observed those effects on the production of some nuclides. From those calculations, the worst case scenario for each nuclide was extracted cumulating the effects that were the less favorable, giving ultimately the highest ratio to Co-60.

Cadmium

Nuclide	Ratio to Co-60
Co 60	1
Ag108m	2490
Ni 63	363
H 3	24.5
Fe 55	4.09
Ni 59	3.11
Sn121m	0.70
Sn121	0.54
Sb125	0.50
Te125m	0.12
Pd107	1.77E-02
Se 79	2.62E-06
Mn 53	3.34E-07
Sn 126	2.04E-07
Tc 99	2.71E-08

Thermocouples

Nuclide	Ratio to Co-60
Co 60	1
Ni 63	10100
Ni 59	98.3
Fe 55	3.44E-01
H 3	2.30E-04
Al 26	2.34E-08
Si 32	2.96E-08
P 32	2.96E-08
Mn 54	1.01E-11
Na 22	6.57E-12
Mn 53	2.37E-14
V 50	6.20E-16

Stainless steel (based on Alloy 316 Ti)

Nuclide	Ratio to Co-60
Co 60	1
Ni 63	1545
Fe 55	42.46
Ni 59	14.60
C 14	2.90
Nb 93m	0.22
Tc 99	5.07E-02
H 3	6.90E-02
Nb 94	3.99E-05
Si 32	8.89E-06
P 32	8.89E-06
Mn 53	1.75E-06
Cl 36	3.02E-07
Zr 93	2.63E-07
Mn 54	1.33E-07
Sr 90	3.09E-11
Al 26	6.17E-11
Ca 41	3.53E-15
V 50	5.40E-15

Aluminum

Nuclide	Ratio to Co-60
Co 60	1
Fe 55	128
H 3	8.92
Al 26	6.01E-03
Ag108m	8.86E-05
Si 32	8.54E-06
P 32	8.54E-06
Mn 53	5.28E-06
Mn 54	5.20E-07
Ni 63	3.11E-07
Na 22	8.63E-08
C 14	8.15E-10
Pd107	7.66E-10
Ni 59	6.51E-12
V 50	2.80E-14

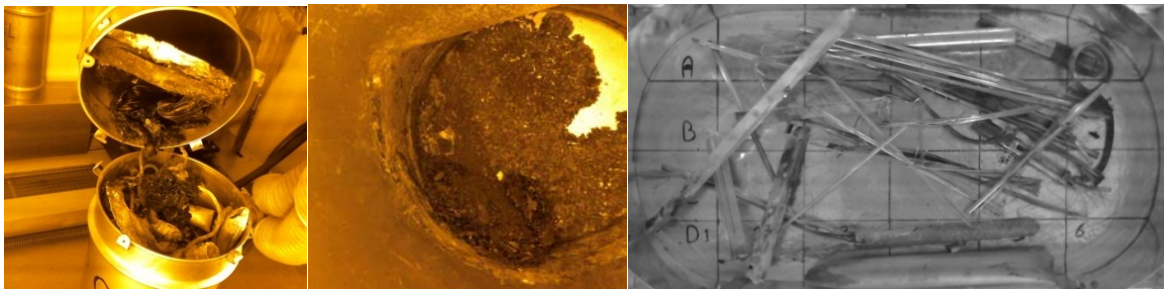

Figure 8: Content of a historical waste vessel from the second set of canisters disposed on the segregation table inside a hot cell

FURTHER DEVELOPMENT FOR MORE COMPLEX SETS OF WASTE

The next set of canisters that will be treated includes residues from experiments that were treated inside NRG's hot cells after irradiation. Thus due to cross contaminations by the processes in hot cells, we cannot rely on only vector-nuclides by activation.

- New instrumentation

The presence of fissile material inside these canisters is still uncertain and the archives do not provide sufficient information to safely open these canisters in the hot cells without contaminating them with alpha emitting isotopes.
The complexity for this new set of canisters lies on the increasing number of materials, the variety of the experiments executed in the past, non-uniformity of their irradiation, and on the relatively poor archives.
The approach is then to create alternative waste streams containing a selected number of materials for a range of parameters linked to the measured irradiation as narrow as possible to allow for the calculation of useful nuclide vectors.
The ambition is, in a parallel route, to acquire a neutron detection system to evaluate the amount of fissile material located in the various containers.

- Fingerprint for contamination evaluation

Contamination will be evaluated and conservatively assessed using swipe tests for detailed analysis. If needed (but this is not the preferred way) chemical destructive analysis will be carried out to evaluate isotopic contents of the materials inside the canisters.

- Toolbox of nuclide vectors

The goal of the toolbox is to obtain, based on the recovered archives and experiment histories, a maximum of "pure" nuclide vector corresponding to known materials with a known history.
When established with sufficient confidence, it is expected that the nuclide content can ultimately be retrieved.

- An inter-comparison between non-destructive analysis and destructive ones

An inter-comparison of different measurements and results is planned by practicing destructive analysis and measurements on certain pure beta-emitters present in the waste such as Fe-55 and Ni-63. The resulting comparison is aimed to indicate error margins of the respective nuclide vectors to assess their ranges.

The combination of all the instruments readouts even though fastidious will allow the formation of new containers of known nuclide content authorizing a good description of the historical waste and the definition of the correspond waste acceptance criteria developed together with the storage facility.

CONCLUSION

Historical nuclear waste characterization represents a complex challenge for the nuclear world in general. Inhomogeneity of the solid waste stream makes representative sampling inefficient and inaccurate to assess nuclide content. The combination of techniques deployed by NRG has so far been successful into determining nuclide content of simple solid waste streams.

For the following families, those techniques will require a development of new techniques as well as a close collaboration with the authorities and the storage facilities.

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