TRANSAT

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Interim report containing research for disposal, reuse, neutralisation and partners, tritium inventory measurement

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Summary

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Approval

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Abbreviations

BP	Belgo Process
BR2	Belgian Reactor 2
DCP	Decontamination and Clean-up Processes
DoA	Description of Action
EC DG RTD	European Commission – Directorate General for Research and Innovation
ECCP	Electronic Collaborative Content Platform
ExCom	Executive Committee
G1L	Getter 1 long
G1S	Getter 1 short (where short stands for the size of the container)
G2L	Getter 2 long
G2S	Getter 2 short
G3L	Getter 3 long
GB	Governing Board
NaK	Liquid metal mixture or sodium and potassium (Natrium and Kalium in Dutch)
PMO	Project Management Office
PQP	Project Quality Plan
PR	Periodic report
QA	Quality assurance
VNS	Variable Neutron Shield
WP	Work package
WPL	Work package leader

Summary

SCK•CEN holds 5 TI-getters and one NaK getter. The tritium content of each of the getters was unknown at the start of this project. To complicate things NaK is a very reactive liquid metal. In this document, as foreseen in deliverable 4.5 of the horizon 2020 TRANSAT project, there will be reported on: the research for disposal or possible reuse options for the getters, the investigation for possible neutralization of the NaK getter, partners used in this project and the tritium inventory measurements.

1 Introduction

The irradiation device VNS (Variable Neutron Shield) provided the possibility to vary the fission power produced in a fuel rod test in the BR2 reactor at SCK•CEN. It consists of a fuel rod which was surrounded by an annular gas cap filled with He-3 gas (99% pure), also called the He-3 shield. He-3 is a strong neutron absorber and absorption of a neutron by the He-3 atom results in one hydrogen and one tritium atom. The produced hydrogen and tritium were removed from the in pile section by sending the gas flow through titanium retention traps located in the out-of-pile equipment mounted in a glovebox in the reactor building.

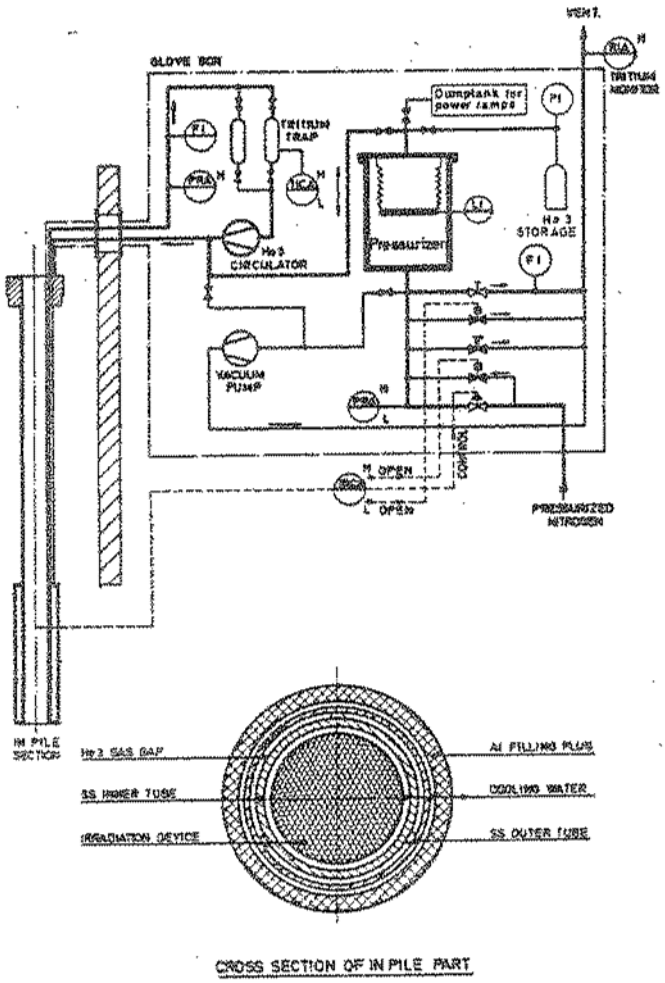

Figure 1: Original PID-scheme of the VNS-1 installation

The maximum tritium retention capacity of the original VNS-1 installation was about 370 TBq (10000 Ci) of tritium. Currently the VNS-1 installation is de-coupled from the in-pile-sections from the BR2 reactor. The VNS-2 installation has the same basic structure as the VNS-1 installation. It is no longer operational as power cycling device. Part of the installation is still pressurized with inert gas and regularly vented for controlled tritium release. The exact tritium inventory of each getter bed is unknown. Calculated estimates show that 3 half lives after the operation the theoretical maximum tritium content of a Ti-getter could contain up to 46 TBq of tritium. At the start of this project 2 Ti-getters (G1S and G2S) were stored in the VNS 1 glovebox, 3 Ti-getters (G1L, G2L and G3L) were located in the VNS 2-installation.

To make things more complicated, a NaK getter is stored in the VNS-1 glovebox. This is an experimental liquid NaK getter in a stainless steel housing, with a micro filter to optimize tritium trapping while passing through the liquid. NaK reacts vigorously with water and oxygen. This getter must be handled carefully before and during dismantling. Besides the chemical risks, the tritium content of this getter was also unknown at the start of this project. In early technical notes [1] the maximal capacity was written to be 17.3 TBq/g NaK, but that number is derived from a theoretical approach in which was assumed that every mole of NaK would bind a mole of HT. The cold storage of tritium on NaK is also described in [2].

So in order to find a disposal route for the getters, the tritium inventory of each getter must be determined and possibly a treatment of the NaK getter to limit its reactivity.

2 Search for partners in this project

2.1 Internal SCK•CEN partners

This project is led by members of the Decontamination and Clean-up Processes group at SCK•CEN (DCP). Most of the work will be executed by them. People of various other departments will be involved during the process. But their contribution is isolated to certain tasks.

2.2 External partners

2.2.1 KEP nuclear – Calorimetry

The determination of the tritium content in getters was assigned to KEP nuclear through a tender [3]. The offer of KEP nuclear met all selection and exclusion criteria listed in the call and came out as best candidate according to the set evaluation criteria.

2.2.2 Other external help

There are other external institutes whose equipment we could use on the project or whose expertise would help us to gain a better understanding of the safety risks. Although necessary to complete this project and while we appreciated their specific contribution, it does not qualify them for being a formal partner in this project.

Examples of such support are:

- BelgoProcess provided their fork lift truck and driver for loading and unloading the calorimeter from a transport truck.
- François Beauchamps from CEA helped with the analysis of a radiography of the NaK getter in order to confirm the absence of NaK oxides.

3 Tritium inventory measurements

Tritium inventory measurements are being done with a KEP µLVC calorimeter. The average measured tritium content is listed in Table 1 below.

In summary it can be said that tritium is present in measurable amounts on each Ti-getter. 2 Ti getters contain 20 – 30 TBq of tritium, which is close to what could be expected current date if they were loaded to the maximum allowed capacity (370 TBq or 10000 Ci) several years ago. The 3 other Ti getters contain an order of magnitude less tritium. The tritium amount of the NaK getter was below the minimal measurable amount that could be detected with the current setup.

Table 1: Measured tritium inventory

Getter id	Average measured tritium content (TBq)
G1S	2.7
G2S	23
G1L	29
G2L	1.7
G3L	4.0
NaK	<0.5[#]
Total	**60,4**

[#] Based on reproducibility experiment

4 Disposal / recycling options

In this chapter the various options investigated for the getters will be discussed. The options listed in this interim report should not be considered as limiting. Others might be triggered during the progress of this project. Also, the full analyses of each option might not be complete at the moment of writing of this interim report.

Based on the measurements reported in Table 1 we will consider that a total amount of 60 TBq tritium is stored on the getters.

4.1 Disposal via release the stack of SCK•CEN's tritium laboratory

4.1.1 The basic idea

SCK•CEN has a tritium laboratory with an inventory license of 25 TBq [4]. The stack limits of the tritium laboratory are 69400 kBq/m³ for HT and 20 kBq/m³ for HTO. Taking into account the ventilation flow of 9500 m³/h these rates convert into 659 GBq/h for HT and 0,19 GBq/h for HTO.

By heating a getter and flowing an inert gas through it tritium can be released through the stack.

The ideal situation would be the following:

- The tritium release from a getter can be precisely controlled by heating the getter.

- By introducing any one of the getters into the tritium laboratory inventory limit will not be exceeded
- HT from the getter will not be oxidised to HTO between its release from the getter and its detection in the stack release analyser.

In such a situation the 60 TBq could be released in a minimal timeframe of 91 hours ; or when 8 working hours are considered in 12 working days.

But if we assume that between the release from the getter and the detection in the stack release analyser 1% of all HT would be converted in to HTO [5] then the HT release from the getters should be limited in such a way that the release to the stack does not surpass 2000 kBq/m³. In practice this means a release of maximal 19 GBq/h. To release 60 TBq to the stack without surpassing any limits the operation would take minimal 3158 hours or nearly 400 working days with 8 hours of operation.

4.1.2 Advantages

- Heating elements for the Ti getters are still available.
- The principle of heating and flushing is simple.
- Space can be made available to build an installation for this purpose in the tritium laboratory.

4.1.3 Timings

Excluding development and testing time in an ideal situation the operation on its own would only take 12 working days.

However when as a safety margin a 1% conversion of tritiated hydrogen gas in to tritiated water is taken into account the release will need minimally 400 working days. This estimation is just for the release and does not take into account development and testing and approval of a release installation.

4.1.4 Tritium release

The full content of the getters, currently estimated at a total maximum of 60 TBq will be released to the environment.

4.1.5 Secondary or remaining waste

After the treatment the Ti-getters and the NaK will remain as waste. At this moment we do not know how much tritium will remain present in these getters after heating and flushing. Therefore we also

do not know whether they need to be disposed off as regular or radioactive waste. Heating and flushing with inert gas will not change the reactivity of the NaK.

4.1.6 Disadvantages / threats

The tritium contents of G2S and G1L are to high to transfer them to the tritium laboratory for the current exploitation license. A new license would be necessary.

Although probably the release can be organised that no stack limits will be surpassed a large amount of tritium (60 TBq) will be released to the environment; internal and external approval for this release will also be necessary. The tritium in the getters is not inherent due to operations that were foreseen to execute in the tritium laboratory, this might obstruct the approval for the proposed treatment / release.

Chemical neutralisation of the NaK will remain necessary after this treatment.

4.1.7 Safety risks

The main safety risks that we can foresee this time are the following:

- Uncontrolled tritium releases
- Reactivity of NaK

In theory also the explosive character of tritiated hydrogen gas should be taken into account. Enough flushing gas should be used to ensure that the hydrogen content of the gas leaving the getter is less than 4%. This is not expected to be a limiting factor.

4.1.8 Overall evaluation

The fact that the tritium content of some single getters surpasses the inventory license makes this certainly not a viable solution for all getters. It might be an option for the getters containing up to a few TBq of tritium. For the latter further investigation is necessary if releasing to the stack can be approved.

4.2 Disposal via the stack of the BR2 reactor

4.2.1 The basic idea

The BR2 reactor has a monthly release limit of 43,6 TBq for total tritium for its stack. Where total tritium is the sum of the tritiated water vapour and tritiated hydrogen gas.

By heating a getter and flowing an inert gas through it tritium can be released through the stack.

In such a situation the 60 TBq could be released in a minimal timeframe of approximately 1.5 months. However the tritium release of the reactor in operation should also be taken into account in order not to surpass the limit.

4.2.2 Advantages

- Heating elements for the Ti-getters are still available.
- The principle of heating and flushing is simple.
- Space for a heating and release installation could be made in an obsolete tritium glovebox in the reactor building of BR2.

4.2.3 Timings

The BR2 is a research reactor that works in cycles. In 2019 5 cycles are foreseen, 4 cycles last 5 weeks and there is one cycle lasting 3 weeks foreseen. The time between the cycles can vary between 3 weeks and 2 months.

Taking into account the release limits for the BR2 reactor and the scheduled cycles of the reactor the emptying of the getters will always be interrupted by at least one reactor cycle. Minimal time is currently therefore estimated to 3 months. This estimation is just for the release and does not take into account development and testing and approval of a release installation.

4.2.4 Tritium release

The full content of the getters, currently estimated at a total maximum of 60 TBq will be released to the environment.

4.2.5 Secondary or remaining waste

After the treatment the Ti-getters and the NaK will remain as waste. At this moment we do not know how much tritium will remain present in these getters after heating and flushing. Therefore we also do not know whether they need to be disposed off as regular or radioactive waste. Heating and flushing with inert gas will not change the reactivity of the NaK.

4.2.6 Disadvantages / threats

Although probably the release can be organised that no stack limits will be surpassed a large amount of tritium (60 TBq) will be released to the environment; internal and external approval for this release will be necessary. The operation of the BR2 reactor needs to be taken into account.

Chemical neutralisation of the NaK will remain necessary after this treatment.

4.2.7 Safety risks

The main safety risks that we can foresee at this time are the following:

- Uncontrolled tritium releases
- Reactivity of NaK

4.2.8 Overall evaluation

At this moment release from the BR2 stack seems a more viable option than release from the tritium laboratory's stack. There are two reasons for this:

- There are no problems with the tritium inventory limits.
- The tritium in the getters was generated by operation of the reactor. Therefore – at least in theory – the tritium can be released via the BR2 stack.

Further investigation is necessary to determine whether release to the stack can be approved.

4.3 Disposal as nuclear waste in Belgium

4.3.1 The basic idea

After an internal review with management of waste and liabilities specialists [6] it seems that the Ti-getters might be disposed off as nuclear waste when they are considered as a radioactive source.

BelgoProcess is the processing plant for nuclear waste in Belgium. A radioactive NaK – neutralisation installation is under development at BelgoProcess it is expected to be operational by 2021, but its start has been delayed several times in the past. Theoretically our NaK can be processed there as well.

4.3.2 Advantages

An approval for waste management via this route could establish a route in Belgium for tritiated waste disposal.

Limited tritium release to the open environment.

4.3.3 Timings and cost

Both timings and cost for both the Ti and NaK getters are subject to large uncertainties. The Ti-getters' dimensions are (L) 236 and (D) 28 mm, that is without the closing valves. The storage part of a storage vessel for radioactive sources is a cylinder shaped container with dimensions of (L) 500 mm and (D) 100 mm.

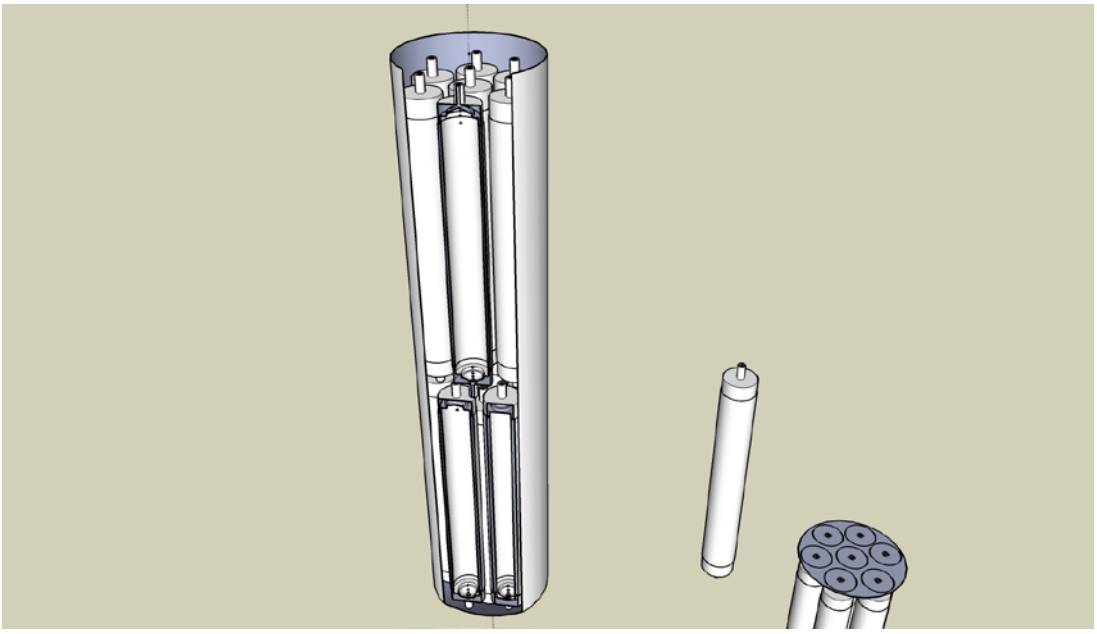

Figure 2: Dimensional analyses shows that up to 14 SCK-type Ti-getters could be stored in the storage part of a vessel for radioactive sources.

Dimensional analyses (Figure 2) shows that the 5 Ti-getters can be stored into one container/vessel. The combined cost of processing and conditioning, interim storage, final storage and other fixed costs are several times 100 kEUR per vessel.

Current estimates for the cost of NaK processing is 100 KEUR/L but with a high degree of uncertainty. This cost does not cover the processing and / or disposal of the resulting end product (K- and Na- salinated water, that might contain tritium as well).

4.3.4 Tritium release

There is not a massive tritium release on a relative short period to the environment as with other options listed in this document. However some release to the environment cannot be excluded. There is no recycling nor reuse.

4.3.5 Secondary or remaining waste

The storage of Ti-getters in a vessel should not lead to too much secondary waste. The end product of the NaK processing is K- and Na- salinized containing water which contains tritium. The amount of water that would be generated is unknown.

4.3.6 Threats

The cost of this operation will be high, but it uncertain how high at this moment.

Further on there are a lot of other uncertainties.

It is unsure whether the tritium containing Ti-getters will be accepted as “radioactive” sources. The primary use of the envisaged vessels is storage of obsolete Co-60 sources. There are many restrictions to the materials that can be stored in these vessels. Most of them don’t seem problematic, except for the restrictions on the redox potential. The redox potential of elements with a metallic form of which the redox potential is lower than the redox potential of H_2/OH^- (E0 = -0.84V, SHE) is not allowed, although It has been confirmed that NIRAS does not consider this as a problem for the Ti-getters because the oxide form of Ti is very stable [6].

The tritiated NaK from our VNS installation was not taken into consideration at the design stage of the NaK processing device. It is unsure whether our tritiated NaK will be accepted for treatment in this device. It is unsure if tritium contamination in the generated secondary waste will be problematic or not.

4.3.7 Safety risks

For the Ti-getters only tritium handling risks need to be taken into account. The NaK getter treatment should take into account the chemical reactivity of NaK. We assume that an installation for NaK neutralisation is designed to do so.

4.3.8 Overall evaluation

This is also not a recycling option, but tritium release to the environment is less than with a number of other options that have been investigated in this document. If it can be done however, an internal disposal route is established.

To conclude:

Despite that there are a lot of uncertainties in this route and that the costs will can be very high this option is worth further investigation.

4.4 Transform into tritiated water

4.4.1 The basic idea

Release the tritium from the getters via heating and flushing with an inert gas. Following that the gas stream can be sent through an oxidation installation where all HT will be oxidised to HTO. Downstream the tritiated water should be distilled out of the gas stream and collected. After that the last remaining tritiated water needs to be taken out from the gas stream by the use of molecular sieve beds. Then a solution needs to be found for the tritiated water. Following options could be considered in this analyses: release to the environment, storage or recycling.

UKAEA is commissioning a water detritiation plant. This route has not been investigated yet. Exploratory discussions will start in march 2019.

4.4.2 Advantages

Conversion to tritiated water was long considered as a way to handle tritiated waste. SCK•CEN has operated in the past an installation to oxidize organic tritiated liquids to tritiated water [7].

4.4.3 Timings

To be investigated if this would be a viable option.

4.4.4 Tritium release

From the process itself this will depend whether there will be made use of molecular sieves as a final drying step or that after the distillation the gas stream will be released to the stack.

This would need to be investigated if this would be a viable option.

For the majority of the activity this would depend on the end goal of the resulting tritiated water.

4.4.5 Secondary or remaining waste

Tritiated water with a total activity of 60 TBq will be generated. If oxidation would be a viable option further investigation is necessary to determine the volume and concentration of this water.

4.4.6 Disadvantages / threats

Because HTO is more radiotoxic than HT by an order of magnitude of 4, the release to the environment via the stack after conversion to HTO would strongly enhance the disadvantages of releasing as HT discussed in sections 4.1.6 and 4.2.6.

About up to a decade ago SCK•CEN generated about 1000 L of tritiated water with a total tritium activity of 7 TBq. Finding a feasible disposal route (including storage and recycling in Canada) was very problematic [8]. The single solution that is currently still being pursued, chemical purification and release to the environment, will definitely not be feasible for 60 TBq of water. The tritium disposal limits of the envisaged sites are high enough to accept an additional 7 TBq of tritiated water on their normal operation, but not to accept 60 TBq of additional tritiated water.

4.4.7 Safety risks

To be investigated if this would be a viable option.

4.4.8 Overall evaluation

Seen the current problems SCK•CEN has with finding a destination for its current tritiated water stock this is considered as not a viable option.

4.5 Recycling of tritium at NSSI (US)

4.5.1 The basic idea

NSSI is an American company, which has shown interest is recycling the tritium on both our Ti- and our NaK getters. The tritium can be recycled for scientific / medicinal purposes. Following examples were given to us as possible reuse destinations of our tritium:

- fuel for laser-fusion projects,

- tracer for medicinal applications
- battery for pacemakers

4.5.2 Advantages

The advantage that this solution has over all the other solution so far is the re-use of tritium. Also the company seems very knowledgeable and offers neutralisations / passivation of both the Ti-getters as the NaK Getter.

4.5.3 Timings and cost

Still need to be investigated. Discussions with NSSI still need to be started.

4.5.4 Tritium release

Needs to be investigated.

4.5.5 Secondary or remaining waste

Needs to be investigated.

4.5.6 Threats

At this moment the cost is unknown, the possibility of transfer of tritium to another country needs to be investigated.

4.5.7 Safety risks

Needs to be investigated

4.5.8 Overall evaluation

There are number or uncertainties. Further discussions with NSSI will have to take place. Nevertheless, this is, until now the only effective recycling option for our tritium.

4.6 Re-use tritium for getter research

4.6.1 The basic idea

SCK•CEN has a collaboration agreement with ICSI (RO). Scientists at ICSI are investigating hydrogen isotope loading / unloading characteristics of new materials that can be used as tritium getters. However they do not have the capability to perform this research with tritium.

Experiments at SCK•CEN could be set-up to make use of the tritium in one or two getters that would contain only a few TBq of tritium. That way at SCK•CEN these experiments can be done in SCK•CEN's tritium laboratory without the risk of surpassing it's tritium content limit of 25 TBq.

A couple of TBq can be unloaded from a Ti-getter, possibly the NaK getter on to the new getter material. Unloading from the new getter material and release the tritium to the stack of SCK•CEN's tritium laboratory can then be an option.

4.6.2 Advantages

Some of SCK•CEN's tritium would be used for scientific research. During this research most of the tritium will be released from one of the old getters of SCK•CEN.

4.6.3 Timings

To be investigated if this would be a viable option.

4.6.4 Tritium release

Possibly most of the tritium from one Ti-Getter (a couple of TBq) will eventually be released to the stack.

4.6.5 Secondary or remaining waste

Although tritium will be released from a Ti-getter, the used getter will still be considered as radioactive material or waste. Also the new getter material that would be tested will end up as radioactive material as well as some connection tubes and other materials necessary to perform these tests.

4.6.6 Disadvantages / threats

This research would be to be approved and funding needs to found for this research. Although a couple of TBq of tritium will be used for investigations, eventually it will be released to the stack. This

is only a solution for ONE Ti-getter with only a couple of TBq of tritium. Getters G1S, G2L or G3L could be an option. For the other getters a solution still needs to be found.

4.6.7 Safety risks

To be investigated if this would be a viable option.

4.6.8 Overall evaluation

If another solution has been found this path can be considered for as an alternative for a getter which only contains a few TBq of tritium.

4.7 Recycling of tritium at RCTritec

4.7.1 The basic idea

RCTritec is a Swiss company. It is a tritium supplier which also delivers decontamination and disposal services for tritiated materials or installations. The company was contacted[9] to verify it would be interested in recycling the tritium on the Ti-getters from SCK•CEN.

4.7.2 Threats

The company can only accept tritium on getters when the hydrogen isotope composition on the getter consists at least for 80% out of tritium (our maximum is 50%), but more importantly RCtritec is bound to legislation which prohibits them to accept radioactive material from abroad [9].

4.7.3 Overall evaluation

Recycling at RCTritec is not an option.

5 Neutralisation options for NaK

5.1 Condition of the NaK

NaK is a very reactive liquid metal. It can even react when it is mixed with its own oxides [10]. The state of the NaK in the getter was unknown. The getter was frozen to avoid mixing of the liquid with the oxides during transport. When it was back at room temperature, radiographs were taken.

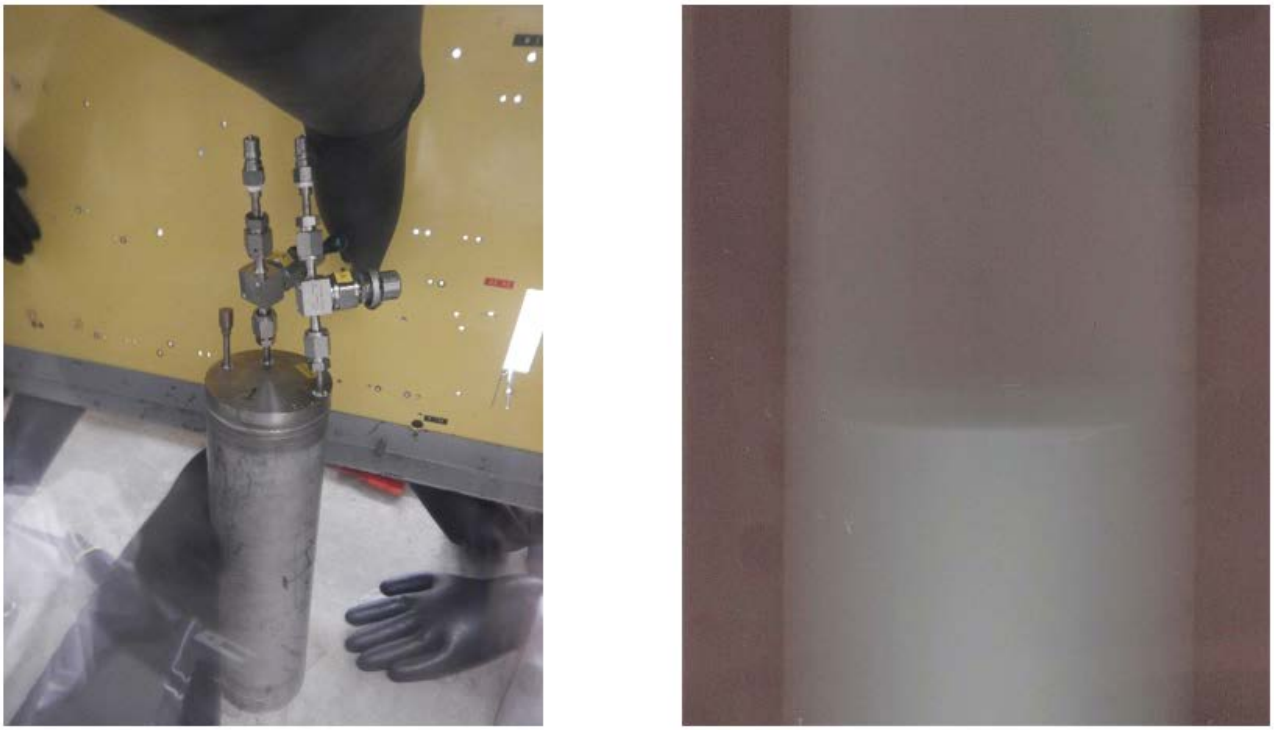

Figure 3: (left) NaK getter stored in the VNS-1 glovebox, (right) radiography of the NaK getter showing a sharp edge between the liquid and the gas indicating the absence of (NaK) oxides

These radiographs (see Figure 3) showed the border between Nak and was very clear and flat so that we can say that the Nak is clean without oxides. This was also confirmed by an evaluation from a CEA specialist [11]. Seen its reactivity handling of NaK is challenging, with oxides it would even be more challenging.

5.2 NaK neutralization process at SCK•CEN

More than 20 years ago SCK•CEN started to look for an efficient and safe technology for metallic sodium waste resulting in a waste form which is ready for further conditioning [12]. Direct controlled

oxidation with water or with an alcohol was considered to be inadequate for our purposes mostly for safety reasons and because of possible complications during the further immobilization of the produced waste. In the early 2000's SCK•CEN started the development of a simple and safe batch process to meet safety requirements and which would render the sodium into a non-reactive form, compatible with the further conditioning of the waste into an acceptable form.

This resulted in the development of a process consisting out of a three-step reaction in a batch reactor. The end product is sodium carbonate. The different reactions involved in the process for sodium are indicated below. The reactions for potassium are similar.

- Oxidation reactions:

  $4\ Na + O_2 \rightarrow 2\ Na_2O$
  $2\ Na + O_2 \rightarrow Na_2O_2$

- Carbonation reactions:

  $Na_2O + CO_2 \rightarrow Na_2CO_3$
  $2\ Na_2O_2 + 2\ CO_2 \rightarrow 2\ Na_2CO_3 + O_2$

After initial testing of the process on lab scale, a small pilot installation was constructed to demonstrate the process with non-active sodium in batches of about 1-5 kg sodium in 50 kg sodium carbonate.

Most of the project people are not longer employed at SCK•CEN, but a great deal of their knowledge has been documented. A new investigation can be started to see whether this process can also be applied for NaK waste. This option looks very promising, the further investigations however are outside the scope of this project.

5.3 NaK at Belgoprocess

Belgoprocess is working to have a NaK neutralisation process in place by 2021. This item has already been covered in section 4.3.

6 Conclusions

At the start of this project SCK•CEN had 6 getters of which the tritium content was unknown. At that point in time these getters were located in 2 different gloveboxes. At the time of writing of this

document the calorimetric measurements to determine the tritium content of each getter were just finished. For the calorimetric measurements SCK•CEN uses KEP as a partner.

Five getters are Ti getters the other one is a NaK getter. NaK is a very reactive liquid metal. Radiographic analyses revealed that in our getter no oxides were present on the liquid NaK. The presence of oxides would have made the handling and this getter even more challenging.

With the characterizations a number of disposal and/or recycling options have been addressed in this report as well as two options for the neutralization of the NaK. In the next report a better evaluation between these latter reports can be included.

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