Semi-long term storage of uranium containing waste from molybdenum production

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Abstract

In Petten, molybdenum is produced in the High Flux Reactor (HFR) by irradiating high enriched uranium for about one week. By a wet chemical process, the fission products are freed from the uranium, and the molybdenum is extracted. The uranium containing waste is collected on stainless steel filters. For mostly political reasons, the uranium had to be transported to the HABOG facility of COVRA, where it will be stored for at least 100 years.

***Keywords**: Waste storage; waste transport package; uranium waste; molybdenum; intermediate storage*

1. Uranium containing waste from molybdenum production

The uranium target used for the production of molybdenum in Petten consists of a thin foil made of high purity uranium-235 enclosed by an aluminium envelope. After irradiation, the target, containing all of the fission products including the molybdenum, is dissolved in NaOH. The uranium does not dissolve in NaOH and remains as a solid in a uranium filter. The nuclear license sets a maximum to the amount of uranium that can be stored in one filter. When the uranium filter is filled with the maximum amount of uranium, the filter is disconnected from the production process and stored in the pool of the Hot Cell Laboratories (HCL) to cool down. After a cooling period of 2 years, the filters can be stored in a dry storage facility, the Waste Storage Facility (WSF).

2. History

Originally, the fission molybdenum manufacturing process was set up (uranium-wise) as a closed cycle. After the production process, the uranium was solidified in the form of sodium-di-uranate and stored until enough filters had accumulated to be transported to UKAEA Dounreay. At Dounreay, the uranium containing waste was reprocessed and with the uranium-235, which was won from the reprocessing, new targets were being made for the fission molybdenum production.

Because of the unforeseen closure of the Dounreay reprocessing plant, a new strategy had to be developed in Petten. A water pool was built designed to store a large amount of irradiated fissile material, enough for quite some years of molybdenum production.

The possibility of building a small-scale reprocessing unit at Petten was investigated, but proved to be unfeasible, not because of economical or technical reasons, but because lack of political and public support at that time.

It was decided to put the uranium containing waste in HABOG, a facility for intermediate term storage, which was being built in the South of the Netherlands. Thus an OTTO cycle (Once Through, Then Out) was adopted for the uranium. Before this facility would open its doors, however, the storage pool would be filled up, endangering the continuity of the molybdenum production. An alternative storage facility in Petten, the (dry pit) Waste Storage Facility, proved to be suitable for storage of the excess uranium, provided it had cooled down in the wet storage for at least 2 years.

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3. HABOG facility

In the 1980's, the Dutch government had decided that all nuclear waste in the Netherlands would have to be stored in one place, at COVRA, Borssele, the Netherlands. In 2003, the facility for intermediate storage (100 years) of High Level Waste and Fissile Waste became operational; this facility is called the HABOG facility. It can handle 2 types of transport containers: the TN-28 and the CASTOR ® MTR 2. Since the uranium filters contain fissile material, these filters have to be stored in the HABOG facility. To store and transport these filters to the HABOG, a method had to be developed that involved using one of the two admitted containers. Since the TN-28 is a very large container, about 100 tonnes, the MTR 2 was used as transport container for the uranium filters. Once this had been decided, a packaging method had to be developed to transport the uranium filters as economically as possible, using this MTR 2 container. The container is designed for the transport of material test reactor fuel elements. So, the task was to find a way to store the filters in 'fuel element like' units. It was decided to pack 3 uranium filters in 1 aluminium filter bin, in which the filters are put at fixed distances from each other with spacers. The dimensions of the aluminium bin are much like those of a MTR fuel element with the heads and tails removed. See figure 1.
The HABOG facility was designed to handle containers with a maximum weight of 1000 kilogrammes. Since the container with the uranium filters would weigh about 1250 kilogrammes, a statement of the builder of the HABOG (Areva) was needed that the heavier containers could be handled and that the heavier canisters with waste could be stored safely at the HABOG facility.

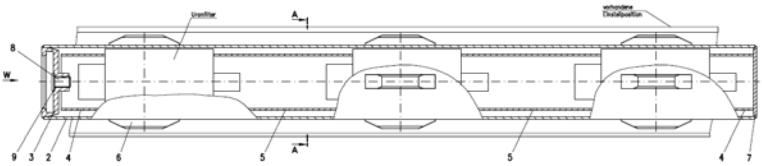

figure 1: Aluminium filter bin for 3 uranium filters.

4. Licence

The MTR 2 container is a German container, manufactured by GNS. In order to get a licence for transporting uranium filters with this container, a German licence is needed, which then must be validated by the Dutch government. The producer of the container has to apply for the licence at his competent authority, in this case the German authorities. The license application consists of a lot of paper work, in which one has to prove that the uranium filters can be transported safely and fulfils all regulations related to such a transport. For example, one needs a mechanical report about the strength of the packaging, and a criticality report about the spatial distribution of the fissile material in the container. When the pile of paper is complete it is sent to the BAM (Bundesanstalt für Materialforschung) and after approval, it is sent on to the BfS (Bundesamt für Strahlenschutz) and this department will hand out the German licence for transporting uranium filters with a CASTOR MTR 2

container. This licence is sent to the Dutch government and on basis of the German licence, a Dutch licence is received by NRG.
So to summarize: even though the material will never be in Germany, a German licence is needed just because the transport container is a German container. And next to this licence, a Dutch licence is needed for transporting the material in The Netherlands.
During the whole licensing process, the question was raised whether the aluminium filter bins should legally be considered to be radioactive waste or packaging. When the bin is part of the packaging, the material requirements for the filter bin are much higher than when the bin is considered to be waste. It was decided by the German authorities that the filter bin is part of the packaging, thus has to comply with strict requirements, regarding mechanical properties and material properties. The spacers between the uranium filters had the highest material requirements imposed on them, since they can have an influence on the criticality properties of the ensemble.

5. Technical

The filters had to be stored in a standard MTR 2/33 basket, so the packaging had to fit into the empty fuel element positions within this basket. It was decided to make aluminium bins that can contain 3 steel filters. Amongst the aluminium alloys readily available on the market, the one least sensitive to corrosion and crack sensitivity during welding was chosen: AlMg4.5 (ENAW 5083). The packaging had to be lightweight to avoid damage of the inside of the container in case of any accident during transport. On the other hand, the bins had to be filled with helium to create an inert atmosphere around the steel filters in order to avoid corrosion. Since the required maximum allowed helium leak rate from the bins was 10^{-5} mbar·l/sec (10^{-6} Pa·m^3/s), the bins had to be welded in a hot-cell. For this, a standard TIG (Tungsten Inert Gas) machine was adapted for use in a hot-cell, see figure 2. Most of the adaptations had to do with the fact that all actions had to be done with removal of radiation sensitive materials and the fact that the whole welding process was done by remote handling. The welding was done without the application of excess metal, since this process is difficult to control in a remote handling environment. In case of a welding failure, it is not possible to enter the hot cell because of the radioactive material present there.
To determine the welding parameters, an array of experiments was set up, with varying welding current and rotation speed. After that, the ‘nominal’ parameters were defined and a validation of the welding process was performed by executing 8 welding experiments at the nominal settings, 5 welding experiments with 5% more heat input, and 5 welding experiments with 5% less heat input compared to the nominal parameters. Cross sections of all these welds were examined by microscopy to define the welding A-size, the minimal welding ‘thickness’, see figure 3A. It appeared that all welds comply with the welding requirements. The aluminium welding process is very sensitive to pollutants, these might cause porosity and/or cracks in the weld, resulting in high leak rates. To reduce the presence of pollutants, the welded parts were cleaned very carefully before welding.

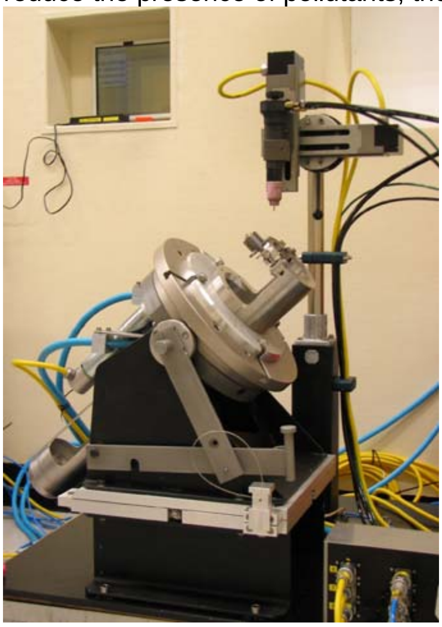

figure 2: Welding set-up (outside Hot-Cell).

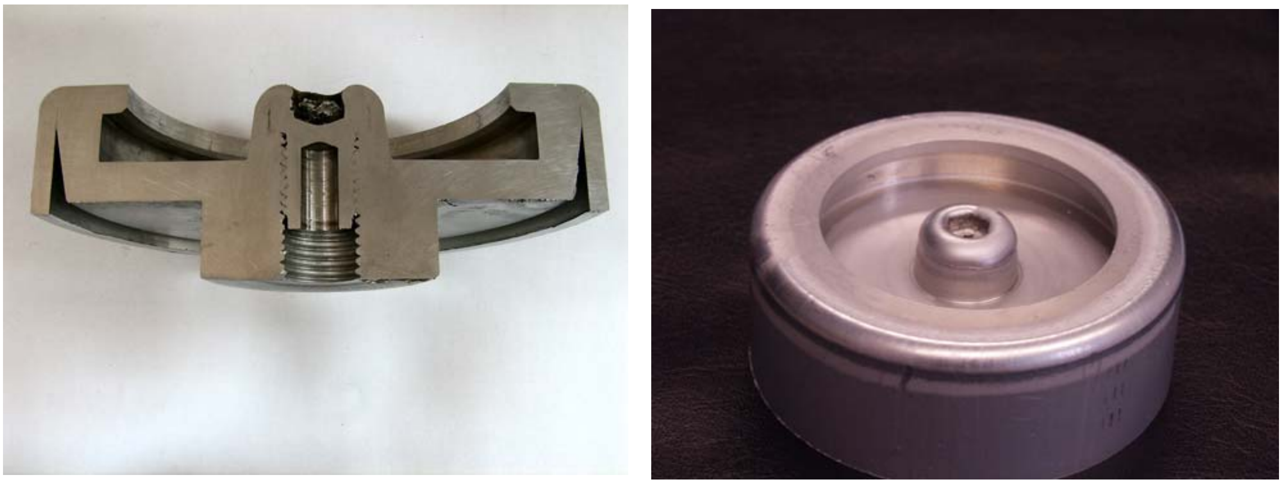

figure 3: **A)** Cross section of a welded lid **B)** Close-up of a welded lid of a bin.

On the other had, the bins also have an helium leak rate requirement of < 10^{-5} mbar·l/sec and not all bins welded with 5% less heat input did fulfil this requirement directly. They did after a second welding round at nominal settings.
For measuring the helium leak rate, a standard leak-testing device was bought and a 'vacuum chamber' was designed and developed in-house, see figure 4. After welding (and filling the bin with helium) the bin is placed in the vacuum chamber and the helium leak rate is measured.

figure 4: Vacuum chamber of leak-testing set-up.

6. Discussion and conclusion

In the whole process of the development of the welding set-up and the leak-testing set-up there were several parties involved who sometimes had conflicting interests. For example, the bins needs to fulfil the requirement for fitting in the transport container, the welding expert has requirements to have a robust welding process, the hot-cell operators have requirements to be able to perform the processes in a hot-cell with manipulators, and the receiver of the uranium filters has requirement for intermediate and final storage of the fissile waste. These, sometimes conflicting, interests resulted in a long development process which yielded an aluminium welding process that can be carried out in a hot-cell. Especially the requirement that we had to design a welding process for aluminium bins was a tough one to meet. But for the licence for the transport container we needed aluminium bins and the receiver of the waste demanded a gastight package that complies with a helium leak rate < 10^{-5}

mbar·l/sec. So, before one starts to develop such a set-up it is very useful to have well-defined requirements of all of the stakeholders in the project.

The transport preparation of the uranium filters was performed in a campaign lasting 8 weeks. In the hot-cells of the Hot Cell Laboratories in Petten, the bins were filled with uranium filters, welded and leak tested. After all these steps were successfully performed, the bin was transported to the High Flux Reactor (HFR) and placed in a rack capable of holding 33 of these bins, situated in the storage pool. After having transported all of the 33 bins, an MTR 2 container was loaded in the pool of the HFR and filled with these bins under the supervision of Euratom, IAEA and the Dutch authorities. The IAEA also performed some measurements to verify that indeed uranium-235 was present in the bins.

At present, the Netherlands are the only country with a licensed packaging and disposal process and a waste repository for uranium containing waste from the fission molybdenum production process. In this year's campaign, we have demonstrated that the method works. In the end, we can say that the project was executed in time without serious problems. Sometimes, fast changes or interventions were needed to get everything done in time. But because of the fact that there were so many parties involved in the whole process, getting fast action was not always an option. If all requirements would have been known at the start of the project, it would have saved both time and money.

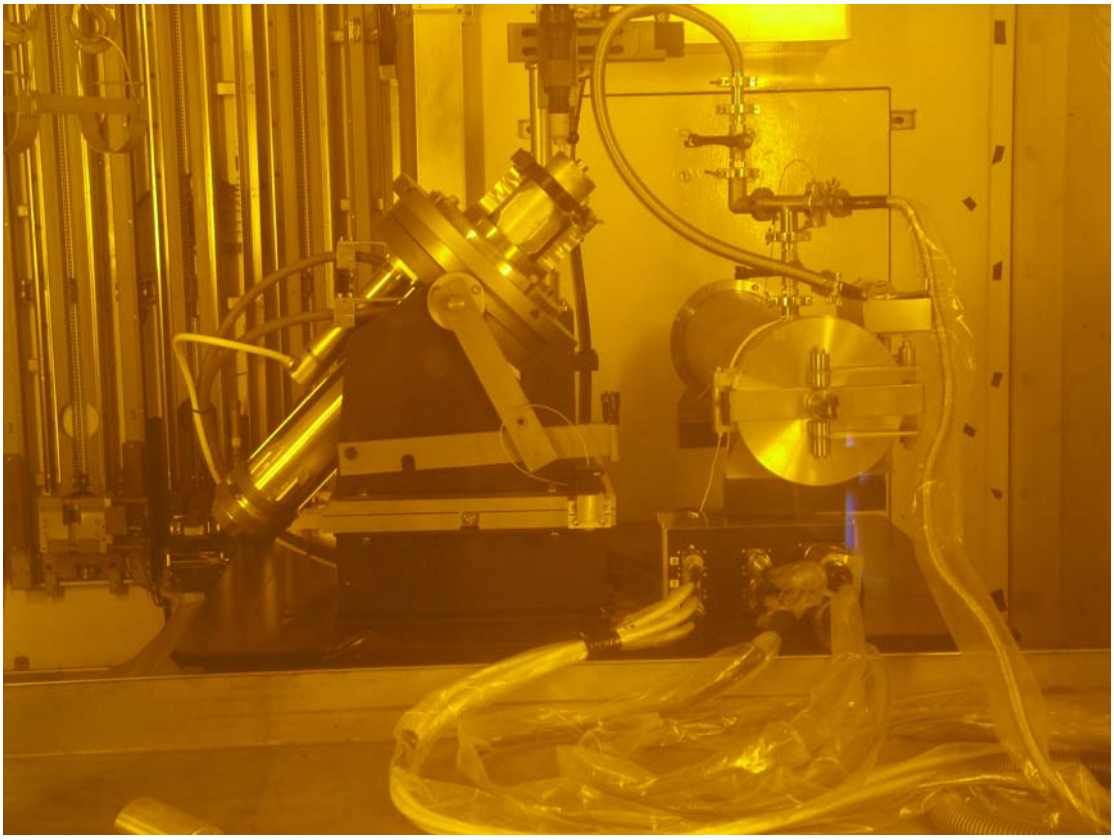

figure 5: the welding machine and leak-testing set-up in the hot cell in Petten.