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Experts report on the bituminisation of operational radioactive waste of the Atucha II Nuclear Power Plant

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Abstract

The Argentinian Commission for Nuclear Energy, CNEA (Bueones Aires), asked for advice on the planned bituminisation of low level and medium level liquid and solid radioactive waste from the operation of the nuclear power plants Atucha I and Atucha II. This expert's report gives some findings from the visit to CNEA and the Atucha II nuclear power plant, and summarises the most important aspects regarding the bituminisation of this radioactive waste (type of bitumen, characterisation of primary waste streams and bituminised waste product, bituminisation of solid waste).

1 Introduction

The Argentinian government decided to resume the construction of the Atucha II Nuclear power plant (CNA-II) and the auxiliary installations, after its construction had been stopped in the second half of the nineties. CNA-II is a pressurised heavy water reactor (PHWR, ~700 MWe) designed by Siemens – KWU. The design dates from the seventies. The heavy water of the coolant and moderator system is first mechanically filtered and subsequently radiologically purified over a mixed bed filter (cationic and anionic ion exchange resins). Also the water from the spent fuel pool is purified over a (second) mixed bed filter. The spent ion exchange resins (at least those of the purification of the coolant and moderator system) are first washed with light water before being sent to the storage container, to recuperate the heavy water.

The liquid and solid operational waste streams can be divided as follows (Table 1; adapted from [1,2]):

Table 1 Liquid and solid waste streams from CNA-II (and partly CNA-I)

Liquid waste		
Group L1	Low activity without tritium	bituminisation of concentrate
Group L2	Medium activity without tritium	bituminisation of concentrate
Group L3	Very low activity with tritium	discharged (Parana river)
Group L4	Medium activity of CNA-I	?
Solid waste		
Group S1-1	(Liquid) filter cartridges	bituminisation
Group S1-2	Ion exchange resins*	bituminisation
Group S2	Ventilation filters	compaction
Group S3	Lab materials, consumables, clothes	compaction
Group S4	Solid waste from CNA-I	compaction

* Ion exchange resins from coolant and moderator purification system loop and from spent fuel storage pool.

A number of these waste streams are foreseen to be bituminised (Table 1). To this purpose, the NPP is equipped with a bituminisation installation using a four-screw extruder with three evaporation domes and with automatic feed of concentrates and spent ion exchanges resins, *i.e.* allowing the continuous processing of the waste in full operation conditions. The bituminisation of resins and concentrates results in the production of so-called 'homogeneous' wastes. The envisaged encapsulation of filters with bitumen would give rise to so-called 'heterogeneous' waste. Details of the bituminisation process can be found in [1,2] and were given during my visit on July 5th, 2006.

2 Impressions after the visit of CNA-II on July 5th, 2006.

The resumption of the further construction of CNA-II and the bituminisation plant started only recently, and it is estimated that it will take at least five years before it will be commissioned. Consequently, only a few staff members are involved in the bituminisation issue. At this stage, their knowledge about the bituminisation process is relatively limited, but they are clearly willing to acquire the relevant knowledge and experience as to assure a smooth operation of the bituminisation installation and to produce bituminised radioactive waste of good quality. The technical staff as well as the direction seems to be to some degree aware of the possibilities and the risks that are related to the bituminisation of low and intermediate level radioactive waste. Minimisation of the amount of final waste products and related costs seems to be a very decisive factor.

3 Some general recommendations

3.1 Radiochemical and chemical content of the primary waste streams

The radiochemical inventory of the primary waste streams is usually reasonably well-known for normal operation conditions. However, failure of fuel assemblies, occasional contaminations *etc.* may result in waste streams with activities and compositions that differ considerably from the average normal operation values. Combination of calculations of the evolution of the fuel element radiochemical composition and regular sampling by the waste producer, backed-up by regular control of the calculations and the analytical results provided by the waste management organisation, will allow to keep track of the evolution of the radiochemical content of the primary waste streams, and to anticipate to the occasional production of waste products with activities that exceed the specifications. A similar remark can be made for the chemical analysis of the primary waste streams.

Further sampling and analysis (radiochemical, physico-chemical), as well as verification by the waste management organisation, is possible before the immobilisation in bitumen or cement, and after production of the final waste form.

3.2 Financial aspects of radioactive waste management

It is equally important that it is from the beginning clear to all involved parties how the costs for the overall waste management and disposal programme are calculated and charged to the waste producers. Optimally, a distinction is made between fixed costs (that are to be made in any case, independent of the number of waste packages) and the variable and marginal costs (the cost for disposal of one more drum, the cost for the geological disposal of a waste drum that was destined for surface disposal,…), and this for the three different disposal options considered in the republic of Argentina (surface disposal of LLW, surface disposal of short-lived MLW, and geological disposal). Logically, the cost for geological disposal of a waste package will be much higher than the cost for surface disposal, and this should be made very clear to the waste producer.

3.3 Acceptance criteria

It is imperative to define relevant acceptance criteria for the different waste forms. Ideally, these acceptance criteria are defined before production of the waste form, and based on a well-defined and well-understood concept for interim storage and final (surface or geological) disposal. If the final disposal concept is not (completely) known, a preliminary list of waste acceptance criteria can be drawn up (such as maximum activities for different alpha and beta/gamma radionuclides, maximum leach rates, solid waste with homogeneous distribution of the radionuclides within the waste product, absence of water, flammable products, heavy metals, toxic organic compounds, *etc.*).

4 Bituminisation of low and medium level radioactive waste

During my visit and the related discussions, the important aspects to be taken into account were discussed in detail. Below, I summarise these important points.

4.1 The bituminisation process

By means of an extruder, the primary waste stream is intimately mixed with pre-heated bitumen and the remaining water is removed by evaporation (final water content typically < 1 weight%). The bituminised waste product (BWP) is finally poured in three steps into a steel container that is positioned on a turn-table with three drums. The filling in three steps aims at maintaining a sufficiently low temperature, in order to reduce the fire risk, and to increase the filling degree of the drum (due to shrinkage of the cooling BWP). It is common practice to limit the filling degree of a drum to 70 – 90 %, depending on the radioactivity of the BWP and the type of bitumen used, allowing for swelling due to radiolytic gas production.

4.2 Choice of type of bitumen

The choice of the type of bitumen strongly depends on the type of the waste (low or medium active, long-lived or short-lived, presence of dehydrated salts, presence of reagents which, when mixed and heated, result in exothermic reactions), and on the final destination of the waste form (surface disposal versus geological disposal).

Basically, two types of bitumen are used for bituminisation of radioactive waste: soft (distilled, straight-run) and hard (blown, oxidised) bitumen. The soft bitumen is 'slowly-flowing' at a lower temperature than the hard bitumen. Use of soft bitumen enables to work at a lower operating temperature (with less risk of fire, and with less risk of clogging of the extruder) than the hard bitumen, but may result in more important swelling of the bituminised waste product (BWP) due to radiolytic gas production (this is essentially the case for the MLW). Use of a hard(er) bitumen requires a higher operating temperature (up to ~190°C), and this gives more risk to the occurrence of fire and explosion (see Section 3).

The choice of the bitumen can be inspired by the average and, if possible to be known, the extreme chemical composition of the waste stream. If these compositions are not likely to give problems with exothermic reactions (see Section 3), a hard bitumen may be preferred for the immobilisation of low and medium-level waste (for low-level waste, also a soft bitumen can be applied). Radiolytic gas generation in a homogeneous BWP based on hard bitumen does not lead to important swelling. This way, the BWP remains a full solid product. A medium-level BWP based on a soft bitumen might become a compressible product containing small hydrogen-filled pores. Additionally, due to the smaller swelling, the use of a hard bitumen can allow to reduce the void in each drum to about 10 volume% (for a BWP based on soft bitumen, a void of 20 to 30 % is preferable).

4.3 Characterisation of the primary waste streams and bituminised waste products

To avoid spontaneous exothermic reactions occurring in the extruder and/or during or just after the filling of the waste drum, it is of utmost importance that the wastes to be bituminised are characterised in a very detailed way. There are some very important aspects related to this issue.

First, before sampling, it has to be assured that the primary 'liquid' waste stream is very well homogenised. One should always check whether one or more segregated layers remain stuck on the bottom of the storage container. The segregated layer(s) contain(s) compounds that are not present in the overlying liquid phase: at the conditioning temperature and in contact with bitumen, these compounds may give rise to spontaneous exothermic reactions to occur, finally resulting in fire accidents.

Second, besides the trivial analyses such as pH, ionic composition (ICP-AES, ICP-MS, IC, ISE,…), organic composition (TOC, presence of oils, detergents,…), TGA (thermo-gravimetric analysis, DTA (differential thermal analysis), and DSC (differential scanning calorimetry) of the primary waste streams and of the bituminised waste product are absolutely necessary, to assure that no exothermic reactions will occur when mixing bitumen with the waste stream, at the temperature of the bituminisation process, and/or during and after (*i.e.* during the cooling phase) the filling of the waste containers.

If a hard bitumen would have been chozen for bituminisation of the radwaste, and if the results of the TGA, DTAanalyses suggests that there might be a risk of exothermal reactions occurring, decreasing the fraction of radioactive waste in the final BWP may help to reduce this risk (but this obviously increases the amount of waste produced). Also a better removal of the heat during and after the filling step – for instance by applying liquid nitrogen – can reduce this risk.

4.4 Bituminisation of filters

Bituminisation of the highly radioactive filters (encapsulation) gives rise to a so-called 'heterogeneous' waste product, and will definitely generate swelling of the bituminised waste product. This is even the case when hard bitumen is used, and it is due to the fact the radiolytically produced gas cannot be removed at a sufficiently high rate. Such swelling of drums, with overflowing of bitumen, has been observed in several countries (e.g. [3]).

4.5 Bituminisation of ion exchange resins

It is not common practice to bituminise spent ion exchange resins (IERs) through an extruder. In the existing bituminisation installations, spent IERs are mixed with bitumen either in a bath process, or, after grinding, in a thin film evaporator. It will therefore have to be verified if the mixing of IERs with bitumen in the extruder will not raise problems (blockage, local heating due to friction).

Grinding of the resin beads prior to feeding to the extruder can help to avoid these problems and can contribute to a good homogenisation of the waste in the bitumen. Grinding will also help to avoid possible problems of segregation. Use of a hard bitumen can also decrease the risk of segregation, and will help to allow the evacuation of radiolytically produced hydrogen (*i.e.* less swelling). At temperatures of about 150°C anionic ion exchange resins start to decompose, resulting in the release of amines. This requires a well-performing off-gas purification system and the treatment of the condensate containing high amine concentration.

During the bituminisation process, the IERs are dried. When later they come in contact with water or high humidity atmosphere, the dried resins will take up water, giving rise to the swelling of the BWP. Consequently, the filling degree (and related remaining void) should enable the swelling of the BWP without reaching unacceptably high swelling pressures. When the amount of IERs is limited to 40 weight%, the extent of swelling should be relatively small. Also allowing a higher residual water content in the resin beads helps to reduce the later swelling.

According to Mr. Guala from NA-SA, the tritium content in the ion exchange resins after heavy water removal would be 0.5 %. Although this is indeed a small fraction, it has to be taken into account. Evaporation of the residual water in the extruder might with time result in a wide-spread tritium contamination of (that part of) the bituminisation plant. If necessary, special measures might have to be taken to minimise this type of contamination.

For several reasons (homogeneity, segregation, swelling), the fraction of IERs in the BWP should be strictly limited to 40 weight%.

4.6 Testing

It is obvious that before starting the bituminisation of the different primary waste streams, tests will have to be performed. In the first place, non-radioactive and active testing on laboratory scale is needed to decide on the type of bitumen, the ratio bitumen / waste (which may be different for the different primary waste streams), *etc.* In a second phase, full-scale tests with realistic non-radioactive waste streams are to be performed (inactive start-up phase of the bituminisation plant). I advise to keep for each category a number of non-radioactive drums for later studies on the compatibility with the final disposal medium.

5 Conclusions

For the treatment of short-lived low level and medium level liquid and solid radioactive waste, stemming from the operation of the Atucha I and Atucha II nuclear power plants, a bituminisation installation is being constructed at the nuclear site of Atucha.

Bituminisation (encapsulation in a bitumen matrix) of this type of waste is a proven methodology that has been applied and that is still being applied in many countries all over the world. Crucial for the smooth processing of the radioactive waste streams is the choice of the type of bitumen, the well thought choice of the relevant parameters for the bituminisation process (temperature, ratio of bitumen to waste, …), and, most importantly, the very detailed characterisation of the primary waste streams and the final bituminised waste product. In this regard, TGA, DTA, and DSC are indispensable characterisation tools to prevent fire and explosion accidents. Encapsulation with bitumen of large pieces of solid waste is to be avoided because of the swelling risk.

6 References

[1] Presentation on the bituminisation of radwaste from C.N. Atucha II, July 5th, 2006.

[2] Technical specification document on the bituminisation installation.

[3] S. Caraguti, 'Experience with a batch process for incorporating low level evaporator concentrate in bitumen developed at Risö National Laboratory, Denmark', in Proceedings of the International Workshop on the Safety and Performance Evaluation of Bituminisation Processes for Radioactive Waste (Radwaste Bituminisation '99, Nuclear Research Institute Rez, Prague, Czech Republic), Ed. R. Vanbrabant (Belgoprocess, Mol, Belgium) and P. Selucky (NRI Rez, Prague, Czech Republic), 1999.