HANDLING AND TREATMENT OF URANIUM CONTAMINATED COMBUSTIBLE RADIOACTIVE LOW LEVEL WASTE (LLW)

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ABSTRACT

Studsvik RadWaste in Sweden has many years of experience in handling of low-level radioactive waste, such as burnable waste for incineration and scrap metal for melting. In Erwin, TN, in the USA, Studsvik Inc also operates a THOR (pyrolysis) facility for treatment of various kinds of ion-exchange resins. The advantage of incineration of combustible waste as well as of ion-exchange resins by pyrolysis, is the vast volume reduction which minimizes the cost for final storage and results in an inert end-product which is feasible for safe final disposal.

The amount of uranium in the incinerable waste has impact on the quality of the resulting ash. The quality improves with lower U-content. One way of reducing the U-content is leaching using a chemical process before and if necessary also after the incineration.

Ranstad Mineral AB has been established in the 1960s to support the Swedish national program for uranium mining in southern Sweden. Ranstad Mineral works among others with chemical processes to reduce uranium content by leaching. During 1998 – 2000 about 150 tons / year have been processed. The goal was to reach uranium residues of less than 0,02 % for disposal on the municipal waste disposal.

Studsvik RadWaste AB has treated about 60 – 80 ton/year U-contaminated incinerable waste from both Sweden and foreign countries between 1961 –1990, thereafter a smaller amount. Presently Studsvik has received new orders for treating U-contaminated burnable waste and a combination of the two methods as described above is advantageous for uranium content reduction in the final waste.

The experience from treatment of U-contaminated waste, advantages, restrictions, relations to Swedish authorities, and transport conditions are discussed in this paper.

BACKGROUND

Studsvik RadWaste AB

Studsvik RadWaste in Sweden has many years of experience in handling of low-level radioactive waste (LLW, sometimes also called intermediate low level waste ILLW), such as burnable waste for incineration and scrap metal for melting.
Fig 1 displays the various routes for LLW from both inside Sweden (hospitals, research institutions, nuclear power plants, etc) as well as from foreign NPPs and other nuclear LLW producers. This figure also shows the final exit route for the return of secondary waste to the customer or final storage in Sweden. In Erwin, TN, Studsvik Inc. operates a

pyrolysis facility based on the THOR-process for treatment of various kinds of ion-exchange resins from the American market. The advantage of incineration of combustible waste as well as of ion-exchange resins by pyrolysis, is the waste volume reduction which minimizes the cost for final storage and results in an inert end-product which is feasible for safe final repository.

Uranium contaminated waste comes mainly from nuclear fuel fabrication facilities and as concerns its treatment, the recycling of uranium is of course also attractive both commercially and environmentally.

Ranstad Mineral AB

At Ranstad Mineral AB in the south of Sweden a chemical process has been developed for uranium leaching of residues from uranium fuel fabrication. The base of the treatment is the process used for leaching of uranium during the mining of aluminum shales in Ranstad 1965 – 69. The process has been developed for leaching of low contents of uranium in the residues.

Different types of residues from uranium fuel fabrication can be processed such as air filters, ashes, ion exchanger resins and burnable material. The process starts with a pre-treatment as milling or disintegration. Leaching with sulfuric acid takes place either as agitation leaching or as percolation leaching. In liquid – liquid extraction equipment the uranium is separated and precipitated as ammonium di-uranate.

The process started in a small scale 1984. During 1998 - 2000 about 150 ton material was processed each year. The production of uranium each year was 700 – 800 kg. The leaching residues had uranium contents <0,02 %. With these low values combined with the strong leaching with sulfuric acid it has been possible to dispose these leaching residues at a municipal waste disposal facility.

Work has today started up to combine this leaching process with incineration steps to receive even smaller quantities of residues and even lower contents of uranium.

INCINERATION OF URANIUM CONTAMINATED BURNABLE WASTE

The handling and treatment of uranium contaminated burnable low-level-waste (LLW) has been conducted at Studsvik for many years with different modes of treatment with regard to pre- and post treatment.

The incinerator outline with primary and secondary chamber is given in Fig 2. The heat from the secondary chamber (about 900 ^{o}C) is reduced via a heat exchanger, which generates hot water for the site with a power of about 1 MW. The temperature is lowered to about 200 ^{o}C before the gases enter the two bag house filters and finally leave the system through a monitored chimney.

The chemistry of the gaseous waste stream is controlled by adding lime and charcoal in order to adjust the pH-value and to suppress dioxin generation. Measured emission values are of the order of 1 – 10% of the limit values stipulated by the authorities (see table below).

Incineration at Studsvik, in general, implies separated incineration campaigns for each customer. In addition to the normal cleaning of the incinerator in between the incineration campaigns a special cleaning is conducted before changing from Co-60 dominating to uranium-contaminated waste and vice versa in order to minimize the risk for cross contamination of the customers' nuclide vector.

Depending on the enrichment in fissile nuclides the incineration of such waste may result in a relatively high uranium concentration in the residual waste ash, since the volume reduction is of the order of 95 – 98 %. Thus restrictions have to be observed particularly on the total amount of uranium present in the incinerator per batch and on regulations for re-transport. Restrictions – mainly due to the risk for criticality - are related to licenses from the authorities, a limited amount of U-235 in the incinerator and the safety during handling of drums or waste packages by the personnel. Also the rules concerning allowed nuclide spectra or "nuclide vectors" for final storage of the processed in the country of the origin have to be taken into consideration before treatment. It is always the country of origin of the waste to which the ashes, secondary waste and outsorted, i.e. not incinerable, material has to be returned to after the treatment at Studsvik.

Our experience is that the lower the temperature during incineration and the lower the amount of uranium in the burnable waste, the better is the quality of the ash, i.e. finer ash with homogeneous distribution of the fissile component. At higher temperature and more uranium aggregation of uranium and silicon may produce lava-like stones of different sizes and the quality of the ash is deteriorated – as concerns homogeneity. Thus incineration above 750 ^{0}C deteriorates the ash quality with respect to later leaching.

Lower temperature in the incinerator can be achieved by spray cooling on the burning waste, but this is not allowed for U-contaminated material due to risk for leakage and consequential external contamination. Alternatively, temperature can to be controlled by feeding less amount of material and by reduction of air flow to the primary chamber.

The amount of uranium can also be reduced by chemical leaching – as mentioned earlier.

As for the incinerator, not more than 900 g U-235 is allowed to be in the incinerator in one batch. A batch comprises a load of up to 5 tons – through continuous feeding - of burnable waste and will burn through three days including during the nights. Thereafter the incinerator is emptied ("ashed out") and can be reloaded with the next batch of max. 5 tons. After the customer's campaign is finished the incinerator is cleaned – normally by a so-called "cleaning incineration" and ready to accept the next customers campaign.

The amount of uranium in the burnable waste can be reduced before incineration by leaching. In a first run about 95% of the uranium can be leached out from shredded material. Leaching of the burnable LLW either before or after incineration can also be applied in order to regain the uranium for recycling. In a second step the incinerated material can be leached again to obtain most of the remaining 5% uranium. Optional leaching is possible in Sweden via Ranstad Mineral AB, as described above, and can be integrated in the overall treatment package.

Measurements

At the entrance of the process stream packages of about 25 - 30 kg are lifted into transport boxes of a conveyor system (see Fig 3). These boxes are automatically weighed at the starting point of the conveyor, and then the waste is conveyed to the incinerator opening along a rail under the roof. The facility is equipped with fixed radiation monitors and the personnel with personal monitors for radiation control.

After the treatment standard measurements on the ashes and dust are necessary whereas additional special measurements maybe requested by the authorities on a case-by-case basis.

Standard measurement - like for Co-60 contaminated waste in a 100-liter-drum - takes about 5 min on a drum rotation table using a GeLi-detector for gamma spectroscopy. The results are stored in the measurement computer's database. Simultaneously dose rate at 1m distance (D_1) is determined with another detector system. The measurement geometry layout and the detectors efficiency is given in Fig 4.

For uranium contaminated material the ash and dust drums are normally measured for about 10 min. For such type of waste the authorities may even request special measurements as concerns uranium isotopes and Plutonium. Relevant analyses can thus be conducted at Studsvik by our accredited laboratory.

Measurement on ashes in 200-l drums when treated with 5 cm layer of cement shielding is not possible due to absorption-shielding of the low energy gamma spectrum.

Authorities

In Sweden both SKI (the Swedish NRC) and SSI (Swedish Radiation Protection Institute) are the relevant authorities to stipulate the rules and grant permissions for the import and export of such material as well as the treatment at Studsvik's facilities. Transport and treatment is regulated by the EU-Directive 92/3 for the European countries. Studsvik can and has treated material from countries outside the European Community.

Licenses and limit values

Studsvik RadWaste is certified for ISO 9001, 14001.
The licenses from the Swedish authorities include permission for treatment of foreign material, both burnable and meltable waste incl. decontamination and segmentation.

The licenses from Swedish authority SKI maximize the enrichment of U-235 to 5%.

The emission values are limited as given below, with normal production values in parenthesis:

DUST:	20 mg/Nm^3	mean value per month	(normally 1 mg/ Nm^3)
DIOXINE:	0,1 ng/ Nm^3		(normally 0,003 ng/ Nm^3)
CO:	100 mg/ Nm^3	max value per 15 min	(normally 10 mg/ Nm^3)
Hg:	30 µg/ Nm^3	mean value per month	(normally < 3 µg/ Nm^3)
HCl:	100 mg/ Nm^3	mean value per month	(normally < 30 mg/ Nm^3)

Post treatment

Post–treatment, like conditioning of the ash drums with an inside layer of 5 – 7 cm cement according to authorities' license requests and as agreed with the customer can be conducted at Studsvik. This is done in order to avoid risk for critical concentrations and effects of mechanical impacts or by water from outside during temporary storage and transportation. To use a mixture of ash and cement is not recommended due to the risk for producing gases which can deform or even destroy the drum in due time.

The return drums are normally color coded by Studsvik to indicate the content being: ash or dust or mixed ash and dust. Outsorted material – which is not incinerable – is returned in separate drums.

The treatment has to be finalized within 2 years from arrival at Studsvik to the return of secondary waste to the customer. Studsvik never takes over the ownership of the waste. And Studsvik always has to return the secondary waste.

Transport conditions

Transportation is normally conducted by train, truck or ship. Deliveries on railway are re-loaded for a couple of miles of truck transport to the Studsvik site.

As concerns Co-60 dominating burnable material transport conditions are related to surface dose rates alone.

As for ashes and dust from uranium contaminated material the re-transport is limited by

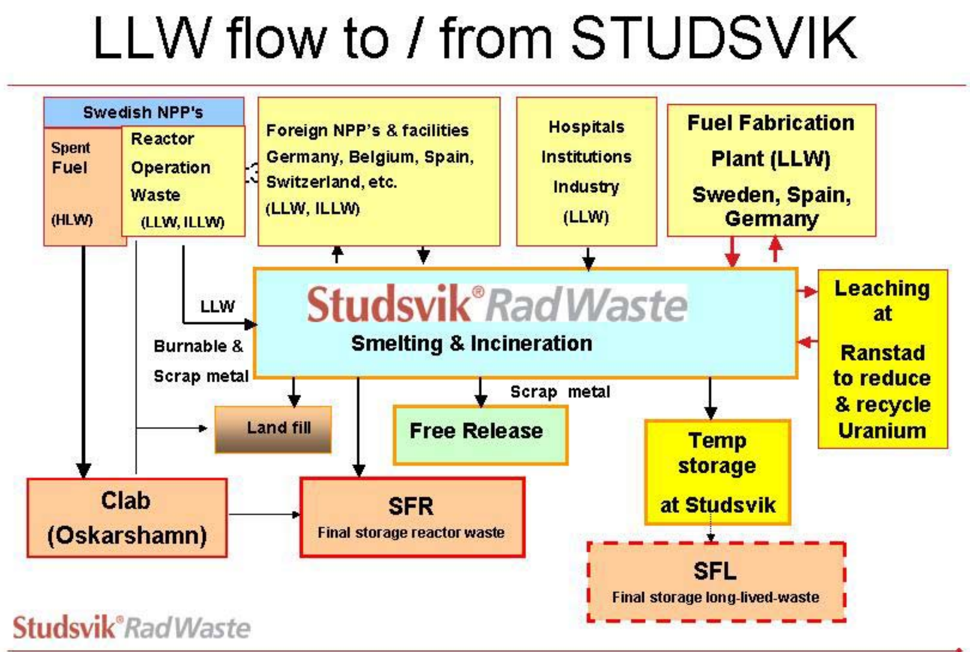
- 5 g U-235 per 10 liter (homogenized ash in drums)
  or
- 15 g per container (where the "container" normally is a 100-liter-drum, in certain cases the 220-l volume can be used for normalization)

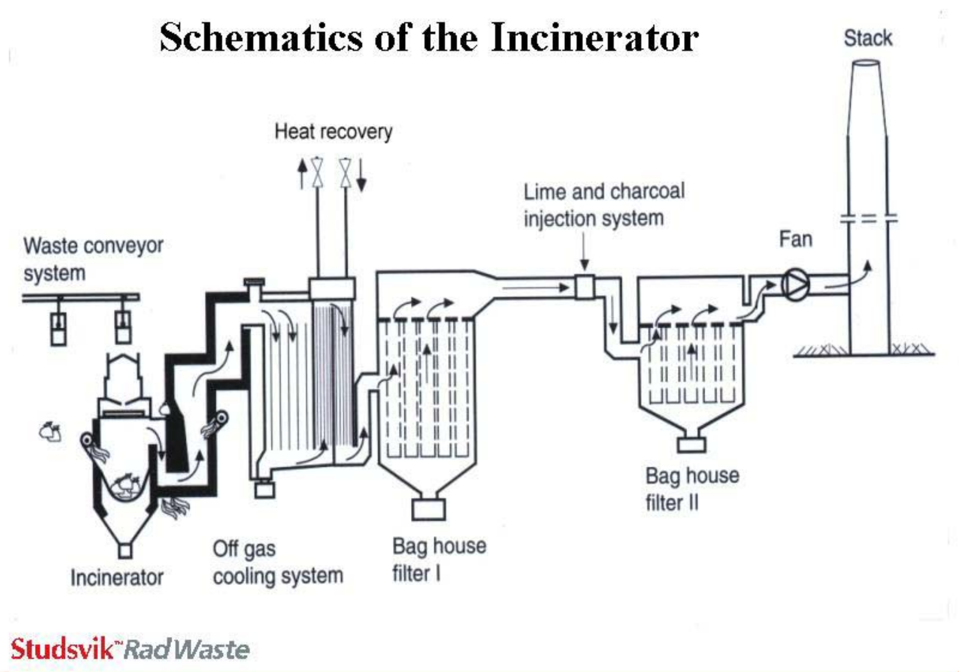
Conditions for final storage

Studsvik has to fulfil the conditions for Final Storage as relevant for, or as demanded by the customer, whoever is the owner resp receiver of the material and destination for return.

Alternatives are:

- delivery in 180-liter drums, e.g. to be supercompacted by the customer or his agent
- delivery in 100-liter drums , conditioned (cemented) inside 200-liter drums for direct final storage.
- different types of drum wall material ; (relevant for return of higher amount of residual $Bq_{(U\text{-}235)}/g$
- different types of lid (bolted, screwed, or otherwise designed)


LLW flow to / from STUDSVIK
Swedish NPP's
Spent Fuel
(HLW)
Reactor Operation Waste
(LLW, ILLW)
Foreign NPP's & facilities
Germany, Belgium, Spain, Switzerland, etc.
(LLW, ILLW)
Hospitals
Institutions
Industry
(LLW)
Fuel Fabrication Plant (LLW)
Sweden, Spain, Germany
Studsvik® RadWaste
Smelting & Incineration
Leaching at
Ranstad to reduce & recycle Uranium
LLW
Burnable &
Scrap metal
Land fill
Scrap metal
Free Release
Temp storage
at Studsvik
Clab
(Oskarshamn)
SFR
Final storage reactor waste
SFL
Final storage long-lived-waste
Studsvik® RadWaste


Schematics of the Incinerator
Stack
Heat recovery
Lime and charcoal
injection system
Fan
Waste conveyor
system
Bag house
filter II
Incinerator
Off gas
cooling system
Bag house
filter I
Studsvik RadWaste

Loading into the conveyor box

Studsvik® RadWaste

Gamma-measurement of drums at the incineration facility