Treatment of Radioactive Sludge From Waste Water Treatment for Weight and Volume Reduction with FreezeTec-Technique – 17010

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ABSTRACT

At the Studsvik site in Sweden, Studsvik Nuclear AB operates a water treatment facility that treat all the waste water from the nuclear facilities at the site. The facility contains several pools for waste water treatment. The standard procedure for treating the water consist of adding $FeCl_3$ to precipitate any particulates. Historically this waste has been mixed with concrete and grouted into barrels which will increase the volume needed for disposal in final repository. The sludge that is generated from the water treatment facility has been subject to treatment with the Freeze-Tec method where the sludge is freeze dewatered and after thawing generating a solid phase with high dry substance and a clear water phase.

The radioactive particles/nuclides tend to stay in the solid phase while the water phase has very low concentration of radioactive particles.

99.9% of the Co-60 and Cs-137 activity end up in the solids and a dry solid with smaller volume and weight is obtained after treatment.

INTRODUCTION

At the Studsvik site (Sweden), Studsvik Nuclear AB operates a water treatment facility that treat all the waste water from the nuclear facilities at the site. The treatment facility is called B4 and contains several pools for waste water treatment. Usually the water is treated by adding $FeCl_3$ to precipitate any particulates creating a sedimentary phase.

Sludge/sediments generated over a three year period were pumped to, and stored in a separate pool in B4. This sludge has previously been mixed with concrete and grouted resulting in very large waste volumes (volume increase by a factor of 5:2). The sludge was stored for many years in the pool while alternatives to this grouting-process were investigated. Sludge has also previously been incinerated. The incinerating process is a very energy consuming process due to the large water content, the sludge has a dry substance content (% ds) of approx. 17 % by weight. Due to the high water content of the sludge, additives were needed in order to achieve a higher %ds for the process. Unfortunately that also increased the incinerating volume greatly.

For disposal in the existing and planned repositories for final disposal of radioactive waste in Sweden, the volume of the waste is a key parameter and proportional to the disposal cost. Therefore there is a strong incentive for volume reduction of the waste aimed for final disposal.

It is however possible to treat the sludge in a more energy efficient way by using the Studsvik Freeze-Tec method. By freeze treating the sludge with this method it is possible to separate the solid content in the sludge from the four different kinds of water that are present. These four different kinds of water are:

- Free water
- Bound water
- Interstitial water
- Surface water

See Figure 1 for schematic overview of the different types of water.

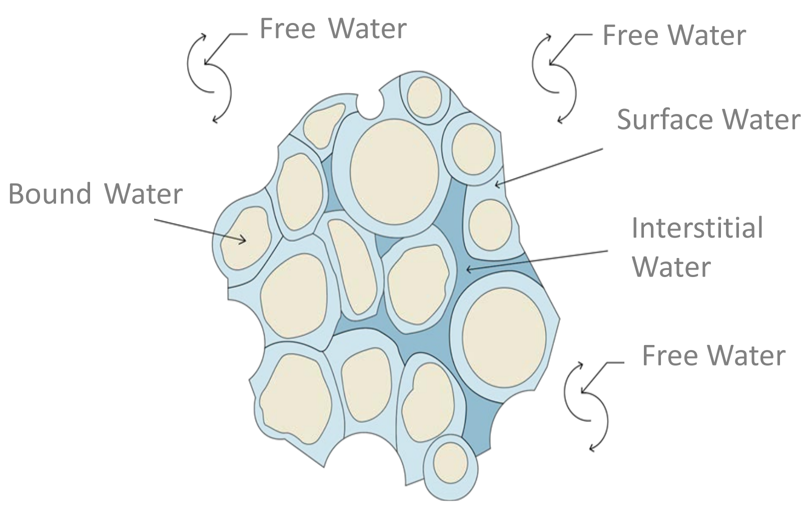

Fig. 1. Different types of water in the sludge.

When the material (sludge) is put in a freezer under controlled conditions, the material temperature start to drop. When the water in the material starts to freeze, i.e. there is a phase transition, the temperature drop decreases. The temperature will, during the phase transition, be stable at the phase transition temperature which for water is 0 ºC. In Figure 2 this is described in a simple phase transition diagram. From left to right, the water is cooled to 0 ºC (red line), after this the temperature is stable at 0 ºC due to crystallization of the water (green line,). When the phase transition is completed the temperature drop continues (blue line).

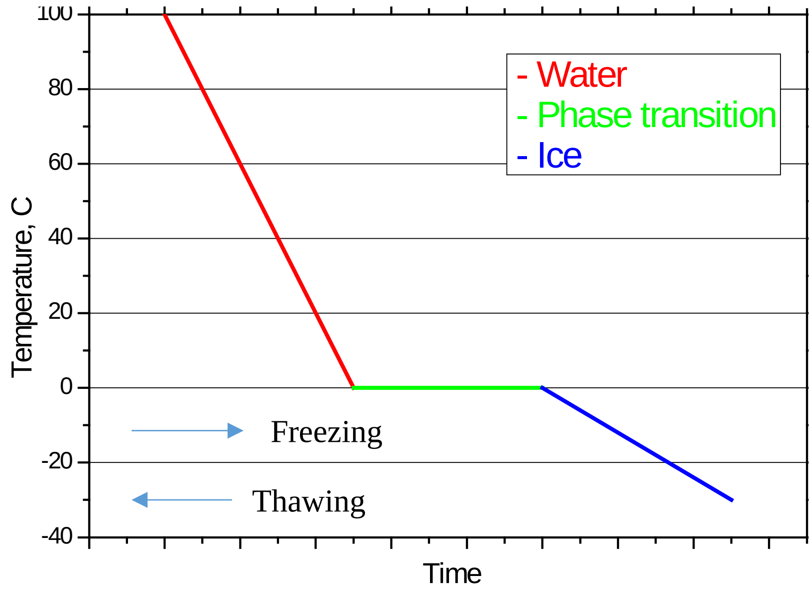

Fig. 2. Simple phase transition diagram for water

In the thawing phase the process is reversed (from right to left in Figure 2). The temperature will rise from – XX ºC to 0 ºC (blue line) where it will stabilize. During the phase transition (from ice to water) the gained heat is spent on breaking molecular bonds, i.e. the temperature is constant until the phase transition is complete. After this the water temperature will continue to increase.

When a sludge is frozen and the phase transition temperature is reached, bound water is released from the solid. Concurrently, the free water migrates towards the cold surface generating a separation between the solid and the liquid phase.

When frozen sludge is left on a mesh to thaw, the different phases (water and solid phase) easily separate by allowing the water to drain through the mesh.

Freeze-dewatering of sludges allows for energy efficient drying of the sludge, in-house know how show that only a fraction of the energy needed to dry sludge with heat is required with Freeze-Tec. The process also results in a good volume reduction since the vast majority of the water in the sludge can be separated from the solid phase.

The process will create a clear liquid phase, with no or very small amounts of radioactive nuclides since they tend to stay in the solid phase. The solid phase that remains will have a high % ds that is more suitable for further treatment such as incineration or direct grouting or just disposal as is. Since the water phase will have very low activity content it is possible to free release or treat as a lower risk waste water.

EXPERIMENTAL

Dry substance content

The first thing to evaluate for the sludge is the initial dry substance content (%ds). This is performed by taking a known amount of sludge and placing it into a heating cabinet at 40 ºC for 24 h. By comparing the weight before and after the drying the %ds is obtained.

Radiological characterization of the sludge

The sludge was characterized by gamma-spectrometric measurement as well as total alpha and beta measurements. The sludge is very homogenous and the sample can be taken in a very representative manner which means that the result from the analysis is seen as representative for the entire batch. The purpose of the radiological characterisation is to construct an activity balance and to visualise where the different radioactive nuclides ends up. The main objective is of course that all radioactive nuclides should remain in the solid phase which would facilitate the release of the clean water phase. For this purpose some assumptions have been made. The activity is assumed not to evaporate which means that a mass balance can be set up even if analysis on all three fractions has not performed. The activity that is not found in the first two fractions is assumed to be in the third.

Freeze tests

For the freeze tests specially made steel troughs were constructed. The troughs had four slightly inclined walls for easier release of the freezed sludge prior to thawing. The volume of the trough was in total 2.7 liters and it was filled with 2 liters of sludge. The sludge has a density of approximately 1 100 kg/m^3.

The top of the trough walls was coated with styrofoam for insulation with the purpose of keeping the top surface warmer than the other surfaces. That will allow the water to migrate towards the cold surfaces of the container during the freezing process.

The trough was also provided with a lid made of the same type styrofoam. The purpose of the styrofoam is to prevent the top surface of the sludge from freezing and thus leaving one of the surfaces warmer than the others. Temperatures were measured at three positions in the sludge, at the centre and at the middle of the short and long side of the trough. When the thermocouples are mounted and the lid has been applied the package is ready for the freezing process. The temperature in the freezer was measured and monitored with a separate thermocouple.

The freezer used for the tests is a modified laboratory freezer that can produce temperatures down to -40 ºC. The sludge was left in the freezer for at least 48 hours, ensuring that the phase transition was complete.

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After the freezing procedure where the water is released from the solid on a microscopic scale the water is separated by letting the frozen sludge thaw on a fine mesh under which the clean water can be collected

The thawing step can be performed both passively and actively. Active thawing is obtained by applying a constant airflow over the thawing sludge with a heighten temperature. Alternatively, passive thawing can be achieved by allowing the sludge to thaw at room temperature with no special arrangements. The target air temperature for active thawing was set at 40 ºC as this reflects the temperature that can be obtained from the excess heat generated by the Freeze-Tec equipment. The excess heat is recycled and used to actively thaw the sludge. The thawing steps were performed in a fume cupboard with a heating fan as source for heat and airflow.

Two freeze tests were performed to get more statistics on volume and weight reduction.

Compaction Tests

Compaction tests were performed in order to investigate how much the remaining dry solid further could be minimized.

For this purpose a subset of the dry solid was transferred to an empty paint container, with a volume of 250 ml. On top of the solid a filter (Al_2O_3) was added and the lid was equipped with four small holes to facilitate the evacuation of the free air inside the container. See Figure 3 for (a) container filled with dry solid, (b) filter and (c) holes in the lid for evacuation of air. Density of the dry solid before compaction was 400 kg/m^3.

(a) *(b)* *(c)*

Fig. 3. (a) Container filled with dry solid, (b) filter and (c) holes in the lid for evacuation of air.

The container was compacted using a laboratory compactor with a maximum pressure of 30 metric tons. For this compaction test the pressure was limited to eight metric tons.

RESULTS

Dry Substance Content

The analysis for dry substance content showed a 17 % ds before freezing tests.

Radiological Characterization of the Sludge

Results for gamma emitting nuclides are presented in Table I and in Table II for total alpha and beta measurements

TABLE I. Radiological activity in the sludge prior to treatment, gamma emitting nuclides.

Nuclide	**Activity, Bq/g**
Co-60	6.30E1
Ag-108m	4.42E0
Cs-137	1.34E2
Eu-154	2.94E0
Am-241	4.20E0

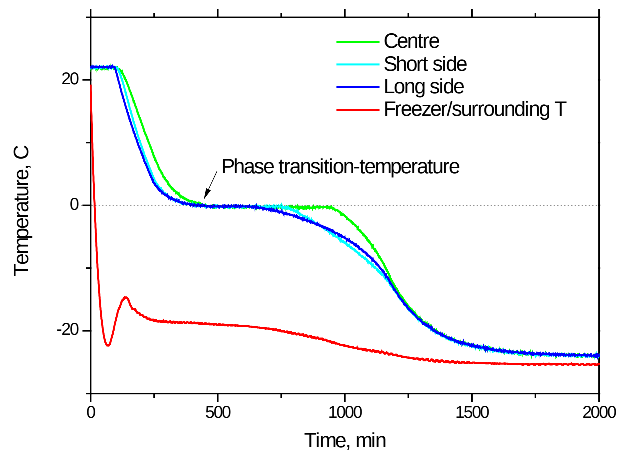
TABLE II. Radiological activity in the sludge prior to treatment, alpha and beta emitting nuclides.

Type of measurement	**Activity, Bq/g**
Total alpha	1.2E0
Total beta	4.2E2

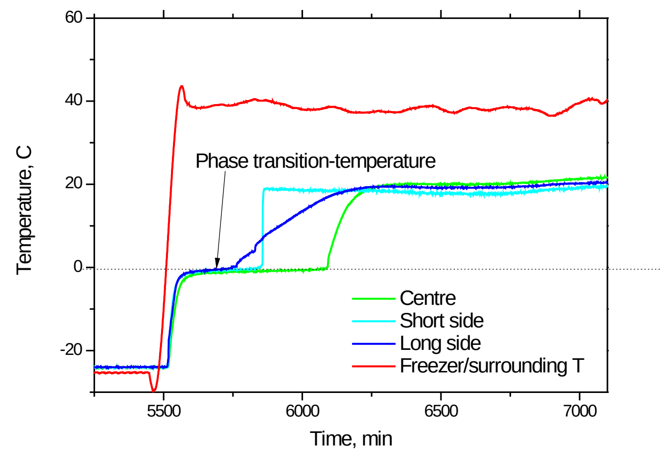
Freeze-dewatering Test

For the first freezing test a total of 2,203 g of sludge was treated. For temperatures during freezing phase, see Figure 4 (a). The temperature curves demonstrate how the sludge temperature stabilizes at 0 °C during the phase transition. The sludge was left in the freezer over a long period (>48 hours) to ensure that all material had reach the targeted temperature. For the thawing phase the material was taken out of the trough and placed on a mesh so that the clean water could be separated from the solids. The sludge was thawed actively by using a heating fan set at 40 ºC.

The temperature during thawing resembles the ones from the freezing in reverse, showing the same expected unchanged temperatures during the phase transition followed by an increase in temperature after completion of the phase transition, see Figure 4 (b).



(a)



(b)

Fig. 4. Freezing of sludge with the expected temperature curves (a), and temperatures during thawing (b)

The frozen sludge with visible thermocouples is seen in Figure 5.

Fig. 5. Frozen sludge prior to thawing, with visible thermocouples

The frozen sludge is placed on a mesh placed over a trough to collect thawed water, see Figure 6. Any agglomerated particles can easily be filtered.

Fig. 6. Thawing of frozen sludge

The sludge had a 17 %ds prior to freezing. After the freezing test the weight was reduced to 513 g and the dry substance content had increased to 73%ds. This means that more than 92 % of the available water was removed by freeze-dewatering, see Table III.

TABLE III. Weights and %ds before and after tests.

	Before freeze test	**Solid phase after freeze test**
ds, %	17	73
Weight, g	2 203	513
Of which:		
Weight of dry solid, g	375	375
Weight of water, g	1 828	138*

*i.e. > 92% of the available water is removed.

Figure 7 shows the dry solids after thawing phase.

Fig. 7. Dry solid after thawing

Radiological Content in the Derived Fractions

The majority of the alpha, beta and gamma activity stays in the solid phase and only traces of alpha, beta and gamma emitting nuclides is found in the water after de-watering step. See Table IV and V for gamma activity content in the different phases after treatment. See Figure 8 for compilation of the results in the different fractions.

TABLE IV. Activity content (gamma emitting nuclides) in the different fractions.

	Activities, Bq/g				
Sample	**Co-60**	**Ag-108m**	**Cs-137**	**Eu-154**	**Am-241**
Sludge, untreated	6.30E1	4.42E0	1.34E2	2.94E0	4.20E0
Dry solid	2.56E2	1.73E1	5.0E2	1.11E1	1.46E1
Water	6.7E-2	*6.2E-3*	8.7E-2	*9.6E-3*	*2.1E-2*

Note: Italic letters: below limits of detection; detection limit is shown.

TABLE V. Activity content in the different fractions, expressed as percentage of total activity in the sludge

	Percent of total				
Sample	**Co-60**	**Ag-108m**	**Cs-137**	**Eu-154**	**Am-241**
Sludge, untreated	100	100	100	100	100
Dry solid	95	91	87	88	81
Water	0.1	0.1	0	0.3	0.4

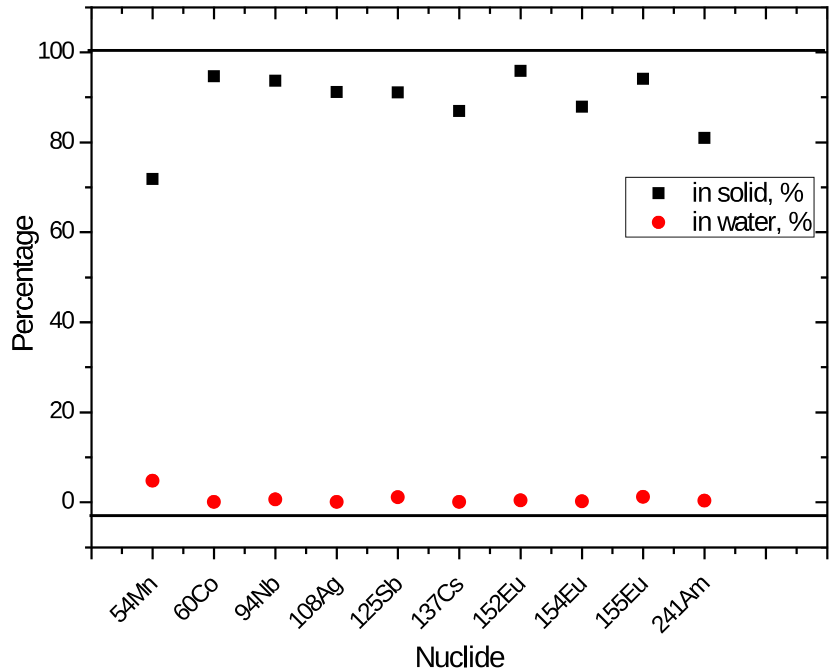


Fig. 8. Distribution of the gamma emitting nuclides in the solid and water phase

The radiological content in the different fractions is presented in Table VI-Table VII below. For alpha emitting nuclides in the water, see Table VI. The only beta emitting nuclides that can be found in the water phase is Sr-90 (and Y-90). 3 Bq/g of Sr-90 is found in the water see.

Table VI. Nuclide specific alpha analysis in the water.

Nuclide	**Activity, Bq/g**
Am-241	< 7.1 E-5
Cm-242	< 3.5 E-5
Cm-244	1.3 E-4
U-234	< 5.9 E-5
U-235	< 3.0 E-5
U-238	< 2.4 E-5
Pu-238	< 2.9 E-5
Pu-239	< 1.8 E-5

Table VII. Beta emitting nuclides in the water

Fraction / Nuclide	Dry solid (510 g), activity in Bq/g	Water (1710 g), activity in Bq/g	Total activity in dry solid (510 g), Bq	Total activity in water (1 710 g), Bq
Sr-90	3.7E2	3.0E0	1.9E5	5.1E3

CONCLUSION

The sludge initially had a 17 %ds prior to the freeze procedure. After the freeze dewatering test and thawing, the dry substance content increased to 73-83 %. This means that more than 92 % of the available water is removed in the freeze-dewatering step.

For Co-60 and Cs-137 approximately 99.9% of the activity after freeze-dewatering is found in the solid phase leaving the liquid phase almost free from radioactive nuclides.

Treatment of radioactive sludge with Studsvik FreezeTec method generates two well defined and separated fractions, solid and liquid phase. The activity is found in the solid phase and the water is almost free from radioactive nuclides. The solid phase is in the form of a dry powder which is easily handled and possible to treat further by for example incineration but with a positive heating value. The energy required for the freezing process is only a fraction of the energy consumption compared to ordinary heating/drying which, especially, for large volumes of sludge would lead to major cost savings.