

A CONTAINERLESS WASTE DISPOSAL CONCEPT
BY IN-SITU SOLIDIFICATION OF MLW/LLW
IN SALT CAVERNS

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INTRODUCTION

The paper describes an alternative concept for the treatment and disposal of MLW and LLW arisings generated mainly at the projected german reprocessing plant and other nuclear facilities (LWR, fuel fabrication, R and D establishments).

The main feature of this concept is an in-situ solidification of preconditioned waste granules in big salt caverns positioned in the deep geological underground thus avoiding any inactive ballast in form of lost concrete shielding and containermaterial.

The necessary R and D work is sponsored by the Federal Ministry for Research and Development and performed in three phases which may be characterized as follows

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|----------------------|------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| phase 1: 1977 - 1978 | Feasibility study and definition of a reference system /1/ |
| phase 2: 1978 - 1981 | Experimental demonstration of relevant components in a semi industrial inactive scale |
| phase 3: 1981 - 1984 | Performance of a large scale test which provides a direct filling of 10^3 m ³ reference waste into the 10^4 m ³ prototyp cavern located in 10^3 m below ground in the ASSE saltmine. |

We have just finished successfully phase 2 which gave rise to the sponsors for further support of this project in the last phase.

DESCRIPTION OF THE IN-SITU CONCEPT

The in-situ concept as presently developed covers the following process steps:

- taking over the waste concentrates
- preconditioning of waste by granulation at the waste producer site
- transport of granules to the final disposal site
- mixing of granules with grout above ground
- direct vertical transport of the reference mixture by gravity via pipeline into the cavern
- spread out of the reference mixture at the floor of the cavern
- in-situ solidification of the waste product in layers each representing one filling campaign
- after a 100 % filling of the available cavern space the borehole position will be decommissioned by filling and sealing with proper means.

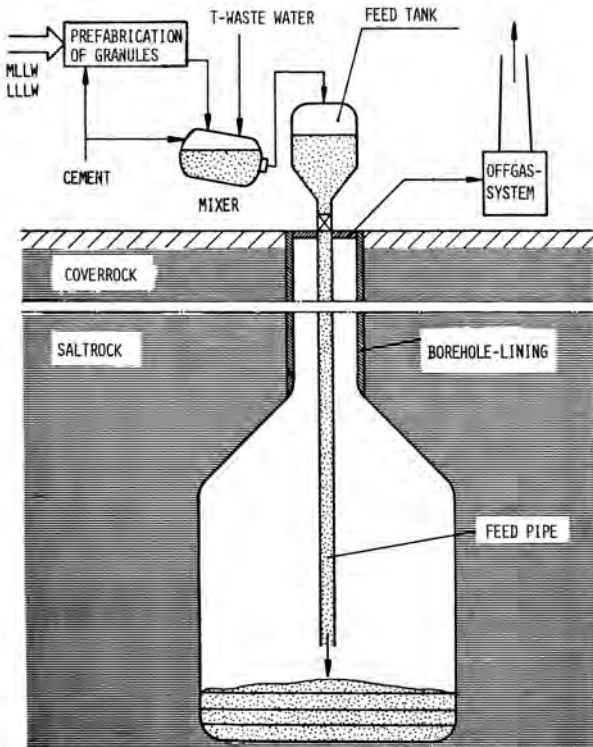


Fig. 1: Scheme of the in-situ concept

The preconditioning of the waste into granules became a necessary step due to the hydration heat of the cementitious waste-product in order to avoid undue temperature rise in the waste-product underground.

Detailed analysis of the product temperature characteristics during and after setting in a big underground cavern has shown that about 60 vol.% in the final product must be replaced by waste granules, which have already released their hydration heat above ground, in order to hold the product temperature underground significantly below the boiling point of the mixture.

On the other hand this pretreatment of the waste provides a number of advantages such as:

- to avoid transports of liquid radioactive wastes in case the waste producer site is in distance from the final disposal site
- rework of batches is possible in case unknown chemicals in the waste solutions prohibit hardening of the cement granules
- quality control of the waste containing solids (the granules) is fairly easy in contrast to a grout system made up by MLW-solution and cement, which sets only underground.

WASTE SPECIFICATIONS

The waste arisings which can be disposed off with the in-situ concept cover a great variety with respect to their physical state, origin and activity concentration. The main criteria are as follows:

- i) the waste must be compatible with a cement matrix
- ii) the activity level is restricted due to the production of radiolytic gases
- iii) the waste must be able to be transformed in granules thus forming the grout a pouring medium through a relatively thin pipeline (50 - 80 mm diameter). The granule size is set to be between 0,3 mm and 5 mm.

The following waste categories generated mainly in a reprocessing plant and in nuclear powerstations have been investigated:

Medium and Low Level Aqueous Waste Concentrates

This category represents process and decontamination waste solutions with high salt content (350 g NaNO₂/l) and a specific activity level of about 300 - 700 β Ci/m³ which amount up to 5 m³ per tonne heavy metal reprocessed. The main R and D activities during phase 2 of the in-situ project were focussed on the granulation technique of these wastes.

Ion Exchange Resins (grain and powder)

This waste is mainly generated at nuclear power stations and represents already a waste form which can be mixed directly with grout and disposed off according to the in-situ concept.

Ashes from Waste Combustion

The ashes appear primarily as slags which can be shredded to proper dimensions of less than 5 mm grain size which are compatible with the in-situ concept.

Tritiated Waste Water

The in-situ concept represents an important alternative disposal concept for H₃ - waste water generated at the reprocessing plant in relatively large specific volumes of about 2 m³ per tonne heavy metal reprocessed. This waste water is used for the preparation of the filler suspension in the reference product. As the waste receiving cavern is not man operated, a contamination of the cavern atmosphere can be tolerated even at high activity levels.

Iodine Absorbermaterial

Loaded iodine absorbers in form of silver nitrate-impregnated silica grain as used in nuclear power station and reprocessing plants are also compatible with the in-situ concept and may be disposed off in the grout without pretreatment.

PRECONDITIONING OF WASTES BY GRANULATION

Two techniques to fabricate granules from aqueous waste solution i.e. rotating dish and a plough share mixer have been developed to an extent that a process comparison could be performed. The plough share mixer did show the better results with respect to dust production, grain size spectrum and the higher granule density which indicated less porosity.

The plough share mixer was then used to demonstrate the granulation process with the help of an inactive pilot plant which has a production capacity of 1,5 t per day. In total about 100 t of granules in specified form have been fabricated.

The main characteristics of the fabricated granules are summarized in Table I

Table I: Main Characteristics of the Granules

Cement = Portland 35 F	76 wt %
Bentonite	5 "
Waste Concentrate	19 "
Water/Cement Ratio	0,15
Grain Spectrum	0,3 - 5 ₃ mm
Density	2,6 tm ⁻³
Bulk Density	1,56 tm ⁻³
Porosity	18 vol. %
Strength	10 cm ² /J

The granulation process is operated batchwise with a residence time in the mixer of about 30 minutes. The green granules immediately after discharging show poor stability and must be stored in a relatively thin layer (less than 25 cm) for about 30 minutes presetting time which can be realized by discharging the granules on a conveyor belt. Afterwards they are stable enough for further procedures like sieving, transportation and storing in silos.

After 7-10 days setting time in special silos which guarantee to keep the initial grain spectrum the granules are loaded via pneumatically driven pipes into special transportcontainers with 5 m³ capacity.

MIXING AND VERTICAL TRANSPORT OF THE REFERENCE PRODUCT INTO THE UNDERGROUND CAVERN

The following paper from Homann et al. /2/ gives all details of this process step. In brief it is described by following features:

The granulated waste product which has passed a setting time of at least seven days to dissipate the heat of hydration is mixed with a binder suspension of cement without additional sand and either fresh water or waste water containing the tritium content of the aqueous reprocessing waste solutions. The content of granulated waste should be close to 60 % by volume not only because of temperature reasons but also in order to prohibit effectively any segregation of the granules which show a significantly higher density than the surrounding filler suspension. The high package density of the granules in the reference mixture does not allow any sedimentation as well during vertical transport as during spread out and setting time.

By proper additives to the cement binder suspension, the grout can be held viscous for transport via a vertical pipe for about 15 - 20 hours. The dimension of the transport tubing governs the flowrate, because the mixture can be fed directly through a pipe of about 1000 meter length. The driving force of gravity is counterbalanced by the friction in the tubing which results in a relatively slow flowspeed of the reference product, e.g. a pipe

diameter of 64 mm restricts the flowspeed at 0,37 m/s or 4,3 m³/h flowrate.

The resulting pancake like product spreading in the cavern shows excellent flowing behaviour, no segregation of granules and filler suspension has been observed so far. The slope of the underground product surface is less than ten degrees which enables to fill underground caverns of proper design (height 80 m / diameter 40 m) to completeness.

The main characteristics of the in-situ reference product which is able to treat specified waste arisings from a reprocessing capacity of 1400 tU/yr are given in Table II.

Table II: Reference In-Situ-Wasteproduct

Reference in-situ-Product: 15000 m ³ /yr	Granules for MLW/LLW	Filler Suspension for H3-Waste Water
Volume Ratio	60	40
Netvolume	9000	6000
Density	2,6	1,6
Mass	23400	9600
Mass Ratio	71	29
Additives	1170 (Bentonite)	32 (Retarder)
Water/Cement Ratio	0,15	0,5
Wastes	4000	3200
Salt/Cement Ratio	6 - 9	-

With one vertical pipeline of 64 mm diameter one would need less than 4000 hours per year to dispose off the reference product specified in Table II. With 24 hrs/day operation time one would spent 160 net operationdays per year which will be performed in three filling campaigns.

IN-SITU-SOLIDIFICATION AND WASTE PRODUCT PROPERTIES

The in-situ concept applies a ventilation system for the cavernatmosphere which does not provide any air exchangerate but only a relatively small flow out rate which is equivalent to the input flow rate of the reference mixture. The in-situ solidification can be considered to take place in a quasi closed system having only very limited exchange with the open air above ground. This design keeps the release of tritium waste water during the setting time at a minimum. It is assumed that the humidity in the cavern air is close to 100 %.

The in-situ solidification process and the saltrock/waste interaction has been closely investigated. The radionuclides which are embedded in the granules may only interact with the saltrock and other leaching media if they are able to pass through the filler suspension during and after the setting time.

Tracer-active experiments have shown that 3 - 5 % of the Cs and Sr activity may be leached by the filler suspension during transport and setting of the product. Any diffusion into the saltrock could not be observed. At the boundary of waste and rocksalt one could identify a cement layer of about 5 mm which was enriched with chlorides.

In order to know the consequences of an accidental brine intrusion during operation time extensive leaching experiments of reference product have been performed using different kinds of leaching media i.e. saturated sodium solutions and quinary salt-solutions as well as demineralized water for reference reasons. Nuclid specific leachrates averaged over a leaching time of 200 days according to ISODRAFT-Specifications are summerized in Table III.

Table III: Mean Leachrates of the In-Situ Reference Product after 200 days Leaching Time.

Leaching media	Mean Leachrates (g cm ⁻² d ⁻¹)		
	Cs-137	Sr-85	H-3
Deminer. Water	3,2 E-5	1,1 E-4	2,4 E-3
Sat. Sodium Solution	4,8 E-5	1,1 E-4	-
Quinary Salt Solution	2,3 E-5	3,1 E-5	-

The results show significant better leachrates (factor 3-10) than homogeneous cement products. In addition on may account with a surface to volume ratio of the waste monolith which is smaller by a factor of 100 than of corresponding drum items.

SAFETY ASPECTS OF THE IN-SITU CONCEPT COMPARED WITH DRUM DISPOSAL TECHNIQUE

The drum disposal concept, which is presently planned by the Physikalisch Technische Bundesanstalt (PTB) for the Gorleben site in Lower Saxony / FRG requires a complete mine with two large diameter shafts and many large underground disposal caverns. It requires millions of cubicmeters of open mine volume for a period of about fifty years. So the stability requirements for this large underground facility are much more severe than for an anticipated in-situ-cavernvolume of 75 000 cubicmeters, which will be back-filled after five years.

The borehole-diameter required for the anticipated solution mining technique and the feed operation will be 300 mm which represents a thin pipe compared with the shaft dimensions of a man operated mine.

Another favorable feature of the in-situ-concept is the complete backfilling of the cavity with a concrete monolith which restabilizes the geological strata and stops the movements of convergence. On the other hand, convergence is a very positive safety factor because it closes any gap between monolith and salt formation, so that no liquid can move at this boundary for eventual transport of activity. Thus a filled and sealed cavern needs no more surveillance.

Even during the time of operation any water intrusion does not pose mayor problems. Calculations on the activity release into groundwater tables with contaminated brine solutions from the flooded in-situ-cavern did not show groundwater contamination above MPC-levels beyond a distance of 300 m from the borehole.

As a further remedial action it is feasible to overcast the waste product in the cavern after water inrush with a layer of insoluble matter as e.g. bentonites, which will result in a very long term diffusion process for the underlying activity. This layer lowers further the groundwater contamination in case of this very improbable accident by several orders of magnitude.

A similar approach for a man operated disposal mine is at least much more complicated if not impossible. The application of the drum disposal technique leaves always open space between and above the drum layers, which cannot be completely backfilled. Therefore to arrive at a safety standard equivalent to the in-situ cavern, the underground disposal rooms must be sealed by dams of huge dimensions which is only possible in the post operational phase of the respective room.

The costanalysis of the in-situ-concept showed considerable reduction (factor 3 to 6) of specific costs compared with the drum disposal concept. One important cost factor of this comparison was identified to be the high specific underground space requirements for disposing a MLW-drum embedded in lost concrete shielding which amounts to a factor of 7 more than the equivalent space requirement of the in-situ-concept.

Further one can dispose off some nuclides with a potential to contaminate the mine ventilation as e.g. tritium and carbon-14 containing waste. A ventilation of the cavern during or after operation is not necessary and shall be avoided.

CONCLUSIONS

Starting with the idea on the in-situ-solidification in 1976, we soon discovered, that the hydration heat of the grout was a limiting factor. The solution for this problem was the granulation technique. By using a size distribution ranging from 0,3 - 5 mm grain size the granule filling volume of 66 % could be attained. Therefore an easily attainable lower limit of 60 % is achieved to guarantee a homogenous pancake like layer of grout in the cavern. The maximum grain size is limited by the minimum diameter of the transport pipe and should be well below 10 % of this diameter.

The concept enables further to dispose off possibly volatile radionuclides, which are strictly limited in a man operated disposal mine. All granular wastes like ion exchange resins, ashes of incineration processes, iodine containing filter materials can be handled together with the cementitious products of suitable grain size.

The vertical transportation is very simple and needs only tubing of 50 - 80 mm diameter. No pumps or other vulnerable equipment are necessary, because the friction in the tubing keeps the gravity flow to a low velocity. The safety of this concept is partly inherent due to the complete refilling of the underground cavern within a short time period. The easy sealing of a small diameter shaft makes further surveillance superfluous. In the very unlikely case of a water inrush during the operational phase, efficient remedial actions can be taken, so that in this case no surveillance of the sealed facility will be necessary later.

On top of all these favorable safety features of this new concept calculations show that economics are very attractive. Though a thorough comparison is not yet possible, space requirements are much lower for in-situ-solidification, operational and transport costs are also much less and a price reduction for disposal of factors rather than percentages can be predicted.

REFERENCES

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- 2 H.H. Homann, M. Weber, Y. Dedegil, "Transport of MAW/LAW Granule Suspension through Vertical Pipes into Caverns". Proceedings of this symposium.

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