UNCEMENTED SUPERCOMPACTED ASH - THE PREFERENTIAL FINAL WASTE FORM FOR LOW LEVEL RADIOACTIVE WASTE

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ABSTRACT

Cementation of low level radioactive waste has been replaced by supercompaction. Low level aqueous waste as well as combustible solid waste are turned into supercompacted incinerator ash. The final waste package is a special container loaded with the pellets the voids between them backfilled with light weight concrete. This measure reduces the final volume of the waste by a factor of about 4. Consequently the costs for packaging and disposal fee are drastically reduced. Long term behaviour of the product is being investigated.

INTRODUCTION

Demands on the safety of waste products as well as on the safety of final repositories are continuously growing. This is a result of increasing knowledge about long term stability of the products and the mechanisms of the radioactivity release to the environment on the one side and the increasing public opposition to nuclear business on the other side.

In some cases this public pressure may result in unreasonably high product quality, stringent regulations and control measures. But at least it pushes ahead the introduction of necessary improvements which otherwise would not be put into practice for reasons of profitability.

However profitability is not evil in itself and the costs of waste management and treatment should be minimized whenever possible.

In the Research Center of Juelich the conception of waste conditioning has been changed in order to achieve a considerable cost reduction. The largest part of the liquid and solid wastes is turned into supercompacted incinerator ash instead of being cemented.

The product is promising because it is regarded to meet the requirements set for the German repository KONRAD and because repository fees can be saved in consequence of the volume minimization.

FACILITIES

In the Research Center of Juelich the Decontamination Department is in charge of the waste treatment. Its staff collects the waste, contaminated materials and equipment arising in the center and in the neighbouring firms, operates the facilities for decontamination, for waste conditioning and for waste storage.

The largest part of the waste both liquid and solid is active in the order of 5 E 4 Bq/kg. It is only slightly alpha contaminated in the order of 5 E 1 Bq/kg. Therefore neither remote handling nor alpha tight containments are necessary for routine operations.

The facilities used for volume reduction and solidification are

- two evaporators
- two drum driers
- one incinerator for solid waste
- one incinerator for liquid waste
- one mixing station for the cementation of concentrates
- one mixing station for the cementation of ashes
- one mobile super compactor, which is owned and operated by the Gesellschaft für Nuklear-Service mbH.

SOLIDIFICATION BY CEMENTATION

In the Juelich Research Center cemented waste in a 200 litre drum was the standard waste form for more than 20 years. This was one of the final waste packages accepted for disposal in the ASSE salt mine. Though the licence of this repository run out in 1978 this product has been produced throughout the following years for lack of another repository and of new acceptance criteria.

The process for the solidification of aqueous low level liquid waste (LLLW) includes evaporation followed by drying of the concentrates and in drum cementation of the dry product.

About 3500 m^3 LLLW per year has been treated in this way producing about 150 200 litre drums containing the cement product.

Combustible solid waste passes through incineration. The ash is fixed by in drum cementation.

About 100 Mg raw waste per year has been treated ending up with approximately 270 200 litre drums containing the cemented ashes.

A mechanical mixer is used for the cementation of the dry concentrates and a tumbling mixer for the cementation of the ashes. Due to both technics a certain void volume remains in the drums. This volume will be filled up with contaminated rubbish and scrap. The total weight of the drums ranges between 350 kg and 450 kg exeptionally it reaches 500 kg.

NEW REQUIREMENTS FOR FINAL WASTE PRODUCTS

The Physikalisch Technische Bundesanstalt (PTB) has issued acceptance criteria for the KONRAD site, the expected German repository for non heat generating radioactive wastes (1). The part of PTB which had been concerned with nuclear matters has recently been incorporated of the newly founded Bundesamt fü r Strahlenschutz (BfS).

Different waste products are listed with specified upper limits for radioisotop inventory. Supercompacted waste is among them. Drums are not accepted as final waste packages but a number of containers with defined dimensions and cast iron vessels. The total weight of each waste package must not exceed 20 Mg. It is expected that the voids in a loaded container must be filled up with concrete.

There are some major consequences on the future waste management. All the waste which has been solidified in drums must be put into containers. This will raise the volume by 90 % if optimum load can be achieved. The costs including repository fee and the price of the container will increase by about 110 %. The weight of approximately 60 % of the drums exceeds the weight limit for optimum load. The increase in costs for their disposal will be 150 %.

In principle two remedial measures can be taken for a future cost reduction. Firstly the in-drum-cementation could be changed into in-container-cementation. As a consequence the costs would be reduced by 45 % but a new process must be realized and a new equipment must be installed. This would be a lengthy and costly procedure. Secondly the final volume of the waste product could be reduced by changing the product itself.

Supercompacted incinerator ash is the product with the smallest possible volume which can be made using the existing equipment in the Juelich Research Center. Compacted ash falls in one of the possible product group according to the new acceptance criteria. Therefore it has been chosen as the preferable future waste form. The final waste package for the KONRAD repository will be a container loaded with supercompacted ash pellets the voids between them filled up with light weight concrete.

INCINERATION

Beside volume reduction there are other advantages of incineration.

The waste will largely be turned into an inert substance. The organic compunds which are partly hazardous will be decomposed. Biological and medical waste will be disinfected. the two-stage Juelich Incinerator with separate gasification chamber and separate burner (2) has the qualification for an effective combustion.

The most simple way to carry out the new process is the incineration of a mixture of combustible solid waste and of dry concentrates followed by the supercompaction of the ash. The behaviour of the dry concentrate during incineration has been tested in a full scale experiment with the Juelich Incinerator. The feed consisted of 2 Mg dry concentrates and 11 Mg combustible solid waste. Approximately 2.5 Mg ashes have been collected. The weight loss of the concentrate has experimentally been determined. It ranges between 30 % and 38 %. The weight reduction factor of the combustible part was much higher than 5 to 6 as usual because the portion of non burnables was very small in this campaign. About 150 kg dust has been discharged into the off gas system. This amount is 50 % higher as found during normal operation. The reason for this increase is the temperature of 850°C the materials are exposed to in the gasification chamber of the incinerator.

Laboratory investigations have shown that the dry concentrates already start to decompose slightly above 200°C with an exothermic reaction. In the second campaign therefore 3.7 Mg dry concentrates have been heat treated separately at only 450 - 550°C. The amount of the discharged dust was 20 kg, about 20 % less than usual. Consequently it has been decided to heat treat the concentrates in separate campaigns in order to investigate if the observed tendency can be proven. During 9 campaigns in 1989 14 Mg concentrates have successfully been heat treated. Consequently the separate heat treatment will be a standard conditioning step for this kind of waste in the future.

SOLIDIFICATION BY SUPERCOMPACTION

The ash is collected into barrels at the bottom end of the incinerator. Each barrel contains about 95 kg ash. The barrels are flanged and sent to the supercompactor. In the Juelich Research Center the mobile Supercompactor FAKIR (3) is used. It is owned by the Company Gesellschaft f r Nuklear-Service mbH and operated by their staff. The working pressure in 30 MPa as required for compacted waste forms.

Calculations based on last year's experience with incineration and on full scale experiments with the supercompaction of ashes show that the treatment of 3500 m^3 LLLW per year and 100 Mg solid waste per year will result in 29 Mg compacted ash pellets. Loaded into containers this waste will need 38 m^3 storage volume. The corresponding volume needed for the 420 drums with cemented products loaded into containers would be 180 m^3.

The saving in costs resulting from the volume reduction amounts to DM 800,000.- per year the price of the containers and the storage fee taken into account.

CONFORMITY WITH THE ACCEPTANCE CRITERIA

The pellets are produced with 30 MPa pressure as required to ensure the dimensional stability of the product.

Table I shows that the nuclide inventory in the container is below the limits set by the acceptance criteria.

The values for the container have been calculated on the basis of a special campaign with extremely high Sr-90 content. Nevertheless the inventory remains below the required limits.

Additionally limits are set for other nuclides which have not been found in the product. Low limits are set for Tritium, C-14, I-129 and Ra-226. Depending on the physical and chemical conditions these limits are up to 6 orders of magnitude lower than those in Table I. Radium however was not found in the ash and Tritium, C-14 and I-129 are not expected because of the heat treatment.

LABORATORY EXPERIMENTS

As the products passed through a high temperature treatment there should be no doubt about their thermal stability.

Concerning chemical stability the reaction of the ashes in air, with water, sodium hydroxide and backfill cement has been investigated. Gas development has been found to be the essential reaction. It occured in all cases except keeping the ash in dry air more or less vigorously.

The experimental conditions and targets were as follows.

- Ashes have been contacted with humid air, water and solution of caustic soda in order to investigate the rate of gas production, the total volume produced and the gas composition.
- Ashes have been supercompacted without encapsulation as well as coated with sheet iron, then embedded into concrete in order to investigate the gas production and behaviour of the package.
- Ashes have been pretreated with solution of caustic soda or with steam in order to complete gas production prior to compaction.
- Coated pellets have been covered with bitumen prior to embedding in order to prevent contact with concrete.
- Ashes have been impregnated with oil prior to compaction in order to prevent contact with reacting agents.

EXPERIMENTAL RESULTS

The gas released from the ashes consists of hydrogen and methane. Nearly all samples contain 98 % - 99 % H_2 and 1 % - 2 % CH_4. Four samples out of ten contained 90 % H_2 and 10 % CH_4 only one sample was 100 % H_2.

The analysis of the ash showed that there is about 40 mg Al and 20 mg Fe per gram ash.

Hydrogen is produced by the corrosion of metals methane is supposed to be produced by the reaction of water with aluminum carbide. A part of the metals in the raw waste passes through the incinerator without being oxidized completely. The material passes first through the gasification chamber where the atmosphere is reducing due

TABLE I

Nuclide Inventory in a Final Storage Container Compared with Required Limits

Nuclide in container	Activity in Container	Required Limit
Sr-90	5.77 E 10	2.1 E 12
Sr-89	2.08 E 9	9.4 E 14
Co-60	1.48 E 9	8.7 E 12
Co-58	1.66 E 7	5.9 E 14
Cs-137	1.3 E 9	9.4 E 12
Cs-134	7.47 E 7	3.4 E 13
Eu-155	6.64 E 7	3.7 E 14
Eu-154	1.49 E 8	1.1 E 13
Zn-65	7.47 E 7	5.2 E 13
Mn-54	1.66 E 7	1.6 E 14
Na-22	1.66 E 7	5.2 E 12
β/γ-activity	1.21 E 11	4.2 E 14
α-activity	1.04 E 8	2.1 E 13

to the adjusted oxigen defficiency. Carbides can be formed in the gasification chamber. Though the temperature in this chamber is limited to 850°C there are hot local temperature peaks far above that value.

Bulk ash kept in humid air releases almost constantly about 0.035 cm^3 gas per gram ash and per day.

The gas production rate rises to about 0.3 cm^3 per gram and day if the bulk ahs is contacted with 0.5 g water per gram ash. Also in this case it is constant for weeks.

The pH value of the water filtrated from ash suspension ranges between 9 and 10.5.

Gas production becomes vigorous if the bulk ash is contacted with a solution of caustic soda.

Typically about 55 cm^3 gas/g ash to 60 cm^3 gas/g ash will be produced if more than 0.35 cm3 NaOH solution (45 %) per gram ash is added. This is well in accordance with the values calculated on the basis of the metals in the sample. The gas production rate is very high in the first 30 minutes. During this time up to 99 % of the total amount of gas which can be produced in the sample will be released. The highest production rate can be observed after the first 10 minutes with 12 cm^3 gas/g . d min. It slows down after the first 15 minutes.

More than 90 % of the gas produced is due to metal corrosion in the ash. Theoretically the sorting out of metals prior to combustion could be a remedial measure but practically it would be too costly and would result in elevated dose to man. Therefore other alternatives have been investigated to prevent or reduce gas production.

The ash always stays in an alcaline medium either by contact with water or with backfill cement if no remedial measures are taken. Because of this condition also the compacted ash will release gases.

Dry ash compacted in a sheet iron cover and embedded in concrete releases about 2 cm^3 gas per gram during the first 10 days. The highest production rate is 0.55 cm^3/g . d during the second day and drops to 0.033 cm^3/g . d after 10 days. A low but almost constant rate is expected for months. After ten days several cracks appeared in the concrete block which contained the pellets.

The gas development can drastically be reduced by covering the pellets with bitumen. This prevents penetration of alcaline solution into the pellets. Only 0.15 cm^3 gas per gram could be measured during the first day then gas development stopped. Also in this case several cracks appeared in the concrete block.

Ashes impregnated with silicone oil prior to compaction develope very little gas. The production rate could not be measured accurately.

Additional substances specially organics should not be given to the waste whenever possible because they reduce the thermal stability. A pretreatment which leaves the ash without residual substances would be the best solution of the problem.

Experiments with steam have been started which indicate that the total amount of gas expected could be driven out in 30 minutes. This is a promising effect but other problems remain to be solved for example the discharge of activity.

FUTURE WORK

Future work will be focused on the investigation of the long term behavior of the compacted ashes in full scale waste packages.

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