SECURE LONG TERM STORAGE OF WASTE PRODUCTS AT THE KARLSRUHE RESEARCH CENTER

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ABSTRACT

Due to the political situation in Germany there will presumably be no final disposal for radioactive waste in the next 30 years. This means, that the operators of nuclear facilities have to ensure a secure long term intermediate storage for radioactive waste products.

The Karlsruhe Research Center operated and cooperated with different nuclear research facilities and laboratories with hot cells, which are now dismantled.

During operation and decommissioning of the nuclear facilities radioactive waste was produced. The Central Decontamination Department (HDB) of the Research Center Karlsruhe has been conditioning radioactive waste into waste products for final disposal. Until the opening of a final disposal the resulting waste products will have to be stored at the HDB. To ensure secure long term storage, quality preserving measures will have to be taken. For example, the corrosion-preventing coating of the containers has to be kept intact. In case of damage the container has to be repaired or should be replaced. Another long term protection measure is the casting of drums in concrete inside the containers. This provides an additional barrier layer in case of drum corrosion.

At HDB 46,500 m³ of radioactive waste products are in intermediate storage. The main project of the next few years will be the realization of the quality measures. In this paper the different methods are described in detail.

INTRODUCTION

For almost 40 years, the Central Decontamination Department (HDB) of the Karlsruhe Research Center is conditioning radioactive waste into waste products suitable for final disposal. Customers of the HDB are:

- Research institutes of the Research Center
- Shut-down nuclear reactor prototypes, such as the Niederaichbach Nuclear Power Plant (KKN), the Karlstein Superheated Steam Reactor (HDR), the Research Reactor (FR2), the Multi-Purpose Research Reactor (MZFR) and the Compact Sodium Cooled Nuclear Reactor (KNK)
- Karlsruhe Reprocessing Plant (WAK) and the European Institute for Transuranium (both on the premises of the Research Center)
- Baden-Wuerttemberg state collection facility
- External customers

Before radioactive waste can be delivered to HDB, it has to be declared by the customer according to the acceptance criteria regarding its material characteristics, origin, mass, volume, dose rate and nuclide inventory as well as the total activity. Data on the radioactive waste, such as pre-treatments, the conditioning and the waste products themselves are stored in the Karlsruhe Data Bank for Radioactive Waste (KADABRA).

HDB operates several facilities for the conditioning of radioactive waste:

- Solid and liquid burnable waste is incinerated and the resulting ashes are super-compacted, resulting in a waste product suitable for final disposal.
- Solid non-burnable waste is super-compacted or fixed in cement.
- Liquid non-burnable waste is, if necessary, either evaporated and solidified or dried.

The resulting waste products are either returned to the external customers or stored in the intermediate storage facility of the HDB until the hand-over to a final disposal. At the moment, about 46,500 m³ of radioactive waste products are stored there. Currently a minimum storage time of 30 years is considered as realistic and the conditioning and declaration of the waste products is done according to the preliminary acceptance criteria of the final disposal project KONRAD. Due to this facts, steps have to be taken to ensure a secure long-time storage of the waste products, as well as to bring the declaration in line with the acceptance criteria of KONRAD.

RE-DECLARATION

In the course of the changing repository situation in Germany from the research repository named ASSE up to the repository project called KONRAD, the requirements for the declaration of waste packages has changed completely. While a minimal declaration of only the nuclides and the total activity was sufficient for the delivery to the ASSE repository, it is necessary to declare a large number of nuclides for the KONRAD final disposal. According to the increasing standards for declaration the HDB has adjusted their acceptance criteria in 1995. Therefore, the older radioactive waste as well as the resulting waste products have to be re-declared. A procedure was developed to re-declare first the waste and afterwards the waste products.

For the determination of the new waste data, the delivering institutions are divided into three groups:

Group 1: Customers for which a nuclide composition can be calculated supported by radiochemical analysis (research reactors)

Group 2: Customers which handle only single nuclides (institutes of the Karlsruhe Research Center)

Group 3: HDB as a producer of secondary waste, which occurs in the process of the waste treatment

About 26,500 resulting waste products have to be re-declared. Table 1 shows the original waste, which has to be re-declared in the first step.

Table I: Overview of the radioactive waste in need of re-declaration

	Number of waste units
Group 1	24,723
Group 2	39,288
Group 3	44,029

Preparation of the Original Data

Before 1990, HDB's acceptance criteria did not require the declaration of all separate nuclides, if the total alpha activity and the total beta activity were below 1.0 E +07 Bq and 1.0 E +08 Bq, respectively. If the total activity was below the above mentioned values and a declaration of separate nuclides existed, it was not entered into the KADABRA data base. To facilitate a re-declaration based on all separate nuclides, about 180,000 covering letters originating between 1977 and 1990 were re-entered manually. These data were checked for typing errors and consistency of customer, accounting, nuclide, activity, measurement unit information and balance between mother and daughter nuclides. Since the nuclide compositions of group 2 customers are based on the date 31/12/1994, the corresponding decay calculations were done for this reference date.

Determination of Re-Declaration Nuclide Compositions

Determination of Re-Declaration Nuclide Compositions for Research Reactors (group 1):

Conservatively estimated nuclide compositions were determined by burn-up calculations and analytic methods for different time spans and reactors parts of the research reactors. Those compositions were assigned to the respective covering letters for the re-declaration.

Determination of Re-Declaration Nuclide Compositions for Institutes of the Karlsruhe Research Center (group 2):

The following data is collected and evaluated to determine the nuclide compositions for institutes which handle single nuclides :

- Monthly information about changes in the inventory of radioactive materials (required by law) between 1964 and 1994
- Inventory declaration of the institutes on 31/12/1994
- The accounting documents of the facility which centrally documents all in- and outgoing radioactive isotopes of the research center institutes

Based on this data, an upper limit for each institute's handled activity is estimated and used for the determination of the following conservative nuclide compositions:

- General nuclide composition without nuclear fuels
- Plutonium nuclide composition
- Uranium nuclide composition
- Thorium nuclide composition

Determination of Re-Declaration Nuclide Compositions for Secondary Waste of the HDB (group 3):

The majority of all activity treated at HDB originates from the Karlsruhe reprocessing plant. Therefore, the determination of the nuclide composition is based on the activation and the fission products of the burn-up calculation. It is corrected by analyses of the effluents of each treatment facility.

Application of the Re-Declaration Nuclide Compositions

Application of the Re-Declaration Nuclide Compositions for the Waste of the Research Reactors (group 1) and the Secondary Waste of HDB (group 3):

If Cs-137 or Co-60 are declared in the covering letters, this nuclide is used as the key nuclide; the declared total activity is maintained and the other nuclides are calculated by their relation to the key nuclide in the nuclide composition. If no single nuclides are declared in the covering letters, the total activity is maintained and the nuclides are calculated by their percentage in the re-declaration nuclide composition.

Application of the Re-Declaration Nuclide Compositions for the Waste of the Institutes of the Research Center (group 2):

The re-declaration of group 2 waste results from the correlation of the total activity with the nuclide composition. Three different cases are distinguished:

Case 1: No declaration of separate nuclides

The activity of the separate nuclides is calculated by using the total activity and the general nuclide composition.

Case 2: Declaration of separate nuclides, but no declaration of fuel elements

The activity of all separate nuclides of the nuclide composition is calculated using the total activity. Missing nuclides are supplemented. The declaration of separate nuclides is utilized if the declared activity is greater or equal to the calculated activity. If the calculated activity is greater than the declared activity, the calculated activity is used. In the end, the new total activity is calculated by adding the single nuclides' activity.

Case 3: Declaration of separate nuclides and fuel elements

In case of the re-declaration of non fuel element nuclides, the practice is the same as described in case 2. A re-declaration of the fuel element nuclides is done only if the fuel elements are not under EURATOM accounting supervision. The re-declaration of missing fuel element nuclides is done on the basis of a special fuel element nuclide composition. This special nuclide composition is created from the isotope ratio of the elements (U, Pu, Th) in the general nuclide composition.

According to the acceptance criteria of HDB it is always mandatory to declare fuel elements in the covering letters. Otherwise, no re-declaration is necessary.

Re-Declaration of the Waste Products

On the basis of the re-declared waste data the activity during all treatment steps and of the resulting waste products is recalculated. The calculation of the new values is done by the accounting system KADABRA, and a total computing time of about one year is currently estimated.

REQUIREMENTS AND MEASURES FOR SECURE LONG TERM INTERMEDIATE STORAGE

The HDB Intermediate Storage for Non Heat Generating Waste

On the premises of the Karlsruhe Research Center HDB operates an intermediate storage for non heat generating waste.

Part I of the storage (building 519) serves as a intermediate storage for drums, concrete-shielded casks and cast iron casks. It is divided into several fork-lift operated areas. The different cask types are stored separately. Drums are stacked in 4 layers and shielding casks in 3 layers.

Areas A – C of **part II of the storage (building 526)** serve as a storage for steel and concrete containers. The containers are a special development in accordance with the acceptance criteria of the KONRAD repository and have a storage volume of 8 m³. The filled containers are brought into the loading zone by a fork-lift. From the loading zone they are lifted into the storage areas by a 250 kN crane. Each area accommodates 7×46 containers horizontally and 8 containers vertically. This provides 2,576 containers per area and a total number of 7,728 containers in all three areas.

Preparation for the secure long term storage and quality insurance will be done in the **handling area D**, after it is finished in June 2002.

The packing and unpacking of the drum-filled containers takes place in the **packing area E.** Measurements of the dose rate and activity as well as gas analyses are taken there.

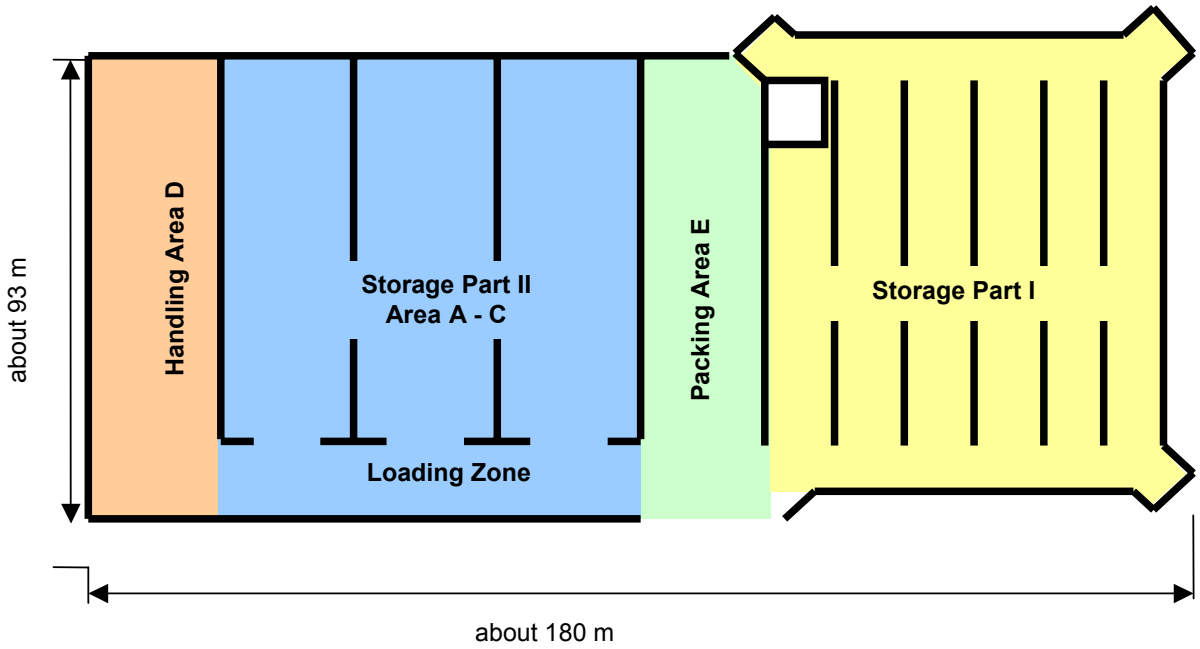

Fig. 1: Ground plan of the storage parts I and II

The storage part I consists of six storage areas with a total storage volume of 15,600 m³. The halls A – C of storage part II have a total storage volume of 61,824 m³. The HDB intermediate storage for non heat generating waste has a storage volume of 77,424 m³ altogether. On 01/06/2001 occupancy was as shown below:

Table II: Storage capacity and occupancy in the storage part I and II

	Container types	Number of containers	Occupancy [m³]	Capacity [m³]
Storage Part I	Waste products drums (150- up to 400-l-drums)	2,097	857	15,600
	Concrete cask type I	4,001	6,402	
	Heavy concrete cask type II	3,046	4,874	
	Cast iron cask type II	166	266	
	MOSAIK container	40	86	
	Sum	9,350	12,485	
Storage Part II (Areas A-C)	Steel container type IV (FSC)	2,301	18,408	61,824
	Concrete container type IV (NBC)	862	6,896	
	Heavy concrete container type IV (SBC)	1,143	9,144	
	Product steel container type IV (PSC)	745	5,960	
	Steel container type II (Version 1 – 4)	135	675	
	Sum	5,169	41,083	
Total			**53,568**	**77,424**

The accumulation of waste volume until 2080 results in a predicted total storage volume of 75,900 m³, without taking a possible reduction due to transports into an eventual final disposal into account.

Fig. 2: View of the partially filled storage area C of storage part II

Requirements for Long Term Secure Storage of Waste Packages

Formerly, it was expected that only a short term intermediate storage (between 5 and 10 years) of radioactive waste would be necessary before relocation to a final disposal. Changes in the political situation in Germany increased that time to a minimum of 30 years. A secure intermediate storage requires the inclusion of the activity during the whole storage time span. Any release of radioactive materials from the waste packages in liquid, aerosol or gaseous form must be excluded.

Accordingly, the following requirements for the waste products and containers are established:

- There must not be a build-up of sizeable amounts of gases in order to avoid a pressurization of the waste packages or the creation of flammable or explosive gas mixtures.

  Different mechanism may be responsible for the creation, release and destruction of gases inside waste packages. The main reasons for gas formation are:

  - Metallic corrosion
  - Bacterial decomposition of organic materials
  - Radiolytical decomposition

It is shown by experience and experimental results that packages filled either with loosely inserted or super-compacted mixed waste are especially prone to gas formation. The formation of H_2 is most important, while CO_2 and CH_4 can be neglected.

- The integrity of the container must not be diminished by mechanical damage or corrosion. Improper handling of the containers could damage the seal and the integrity could be no longer ensured.

  The corrosion protection could be damaged by careless filling, by the transportation conditions and by dangerous substances (e.g. solvents) in the waste. During long term storage, it is possible that moisture in the waste or humidity from the surroundings will condense on the container walls. As a result, the damaged parts of the corrosion protection will start to corrode.

  Metal cast in steel drums can develop contact corrosion, since the contact of different metals with each other will cause electrochemical reactions.

- The waste packages must remain transportable and must correspond to the acceptance criteria of the KONRAD repository.

Measurements for a Secure Long Term Intermediate Storage

Due to the long intermediate storage time of about 30 years, measurements have to be taken to ensure security during the whole time. Older waste products have to be controlled visually for changes caused by corrosion or gaseous build-ups during their previous storage time. To increase security and to avoid further corrosion, the drums are cast in containers with inactive concrete, according to the KONRAD acceptance criteria.

After casting the drums, further quality assurance or re-conditioning is no longer possible. Therefore, the necessary procedures have to be realized beforehand. To implement this, the documentation of the waste products has to be revised in order to correspond to the current standard. This documentation has to be checked by officials of the Federal Office for Radiation protection. Only then does the Federal Office for Radiation Protection allow the casting of the containers.

Before the release for casting, there are different activities necessary:

Unloading of the older drums to inspect them for corrosion

The containers are transported from storage area A – C to the handling area E before unpacking the drums from the containers. There, the containers are unloaded at ventilated working areas with security devices such as remote controlled handling. After unloading, the dose rate is measured on the surface and in 1 m distance from the drums, wipe tests are taken and a visual control is done. Damaged drums are taken away for later re-conditioning.

Fig. 3: Examples of corroded or damaged drums

Repair of steel containers with damaged corrosion protection

Steel containers with damaged corrosion protection or even actual corrosion damages are sorted out and repaired according to a special regulation.

Sampling of gases for analysis

The amount of sampled products is determined by the federal experts (normally 30% per treatment campaign). The gas samples give information about gas builds-ups due to chemical or biological reactions. To prepare for sampling, the lids of the drums are pierced under a nitrogen atmosphere in order to insert a special sampling device. The drums are locked airtight for at least 10 days before the sample is taken. The analysis of the samples gives information about the gas composition (H_2, N_2, O_2 and CH_4) as well as the pressure inside of the drums. If the results show a gas build-up (H_2-content greater than 4%), all resulting waste products of the same treatment campaign are either dried or silica gel is added to the drum contents.

Spectrometric Measurements of older waste products

Part of the waste quality control is the spectrometric measurement of the waste. Normally about 30% of all waste products are spectrometrically analyzed to verify the declared activity. This is done with a special spectrometer for 200-l and 400-l drums. 50 drums are measured automatically in one run to determine

- the weight,
- the medium and maximum dose rate on the surface and in 1 m distance,
- gamma nuclides (qualitatively and quantitatively),
- distribution of the activity.

Sorting of older drums and re-packing into containers

The drums are re-packed together in new or repaired containers grouped by customer in order to fulfill the regulations of final disposal and transportation. Before re-packing the drums, the waste package quality control such as gas analyses and spectrometry has to be

performed. In addition, the dose rate in different positions is measured and pictures are taken of each drum.

Fig. 4: Packing of 14 200-l-drums per steel container

Casting of the Containers

Waste products which pass all quality control steps and have a documentation accepted by the Federal Office for Radiation Protection as being in accordance with the KONRAD disposal acceptance criteria can be cast in containers. First, the containers are filled with inactive concrete. The casting is done in a campaign of 24 units daily. On the next day, the containers are transferred to a special storing area, where they remain for about 4 weeks until the concrete is completely set. Federal experts control the setting and give their permission to close the containers and to put them into storage.

SUMMARY

Due to the increased storage time a number of preventive measurements have to be taken to ensure a secure long term storage. The enhanced requirements for the declaration of waste products make a re-declaration of older products necessary. To minimize radiation exposure, the re-declaration is done only by readjusting the original calculations. Because of the formerly low declaration requirements, a complete reproduction of the activity is not possible, so it is necessary to construct conservative nuclide compositions for the declaration. The nuclide compositions are used in coordination with the Federal Office for Radiation Protection.

Besides the re-declaration of the waste products, a visual control for corrosion and, if necessary, a re-conditioning has to be done. To get the certification that the waste products correspond to the KONRAD acceptance criteria, a number of product quality controls such as gamma spectrometry and gas analyses have to be done.

The final step for an additional security barrier is the casting of the waste products to avoid corrosion processes and therefore, the release of radioactivity.