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Disposal of Radioactive Wastes into the Underground in the Federal Republic of Germany - a Survey on Practical Experience and R+D Work -

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DISPOSAL OF RADIOACTIVE WASTES INTO THE UNDERGROUND IN THE FEDERAL REPUBLIC OF GERMANY

- A Survey on Practical Experience and R + D Work -

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KURZFASSUNG

In der Bundesrepublik Deutschland werden feste und verfestigte radioaktive Abfälle im Salzbergwerk Asse endgelagert. In der vorliegenden Arbeit wird ein kurzer Überblick über die damit im Zusammenhang stehenden, umfangreichen Forschungs- und Entwicklungsarbeiten gegeben; desgleichen werden die praktischen Erfahrungen aus dem Routinebetrieb des Endlagers diskutiert.

Das in Karlsruhe ausgearbeitete Versuchsprogramm für die Versenkung tritiumhaltiger Abwässer in isolierte, erschöpfte Öl-Linsen des tiefen Untergrunds wird beschrieben; mittels dieses Verfahrens kann Tritium ohne jede Belastung der Biosphäre beseitigt werden.

ABSTRACT

In the Federal Republic of Germany solid and solidified radioactive wastes are disposed of in the salt mine Asse. A short review is given of the extensive R + D programmes already executed and still under way, practical experience gained during routine disposal operations is described.

A survey is given of the programme on deep-well injection of tritium-containing effluents into isolated depleted oil horizons of the deep underground that is being studied at Karlsruhe as an approach of tritium disposal which does not lead to a pollution of the biosphere.

1. INTRODUCTION

One of the main problems our technical society is faced with is the problem of waste disposal. It is only since a couple of years of growing "bad experience" that the need for a revision of our waste disposal practice has been generally recognized and efforts are under way for the development and definition of new treatment and disposal procedures that do not add to the pollution of the biosphere.

An example for a waste management practice that from the very first moment has been focused on safety aspects, as far as the protection of man and his environment is concerned, is the management of radioactive wastes. Extensive efforts have been and continue to be undertaken for the development of safe treatment and disposal procedures that do exclude a contamination of the biosphere.

In this paper a review is given on the practical experience acquired and the R + D-work performed in the Federal Republic of Germany on the disposal of radioactive waste in the underground.

2. DISPOSAL OF SOLIDIFIED RADIOACTIVE WASTE IN A SALT DEPOSIT

In the Federal Republic of Germany, the disposal of solidified radioactive wastes in rock salt formations is considered to be one of the most promising and safest methods: Due to their geological history salt deposits offer the best containement, i.e. the most effective isolation from the environment, for the large periods of time needed until decay of the radionuclides to values that are no longer dangerous has been accomplished. This is why the abandoned salt mine Asse [1] situated some 20 km south-east of Brunswick in the Federal State of Lower Saxony was acquired by Gesellschaft für Strahlen- und Umweltforschung (GSF), München as the central disposal site for all radioactive wastes generated in the FRG up till the year 2,000. An extensive R + D-programme aiming at the definition of optimum operation-, disposal- and surveillance-conditions is executed since 1967 in close cooperation with the Decontamination Department of the Nuclear Research Center Karlsruhe. Routine disposal is performed since 1971.

2.1 Description of the Salt Mine

The Asse is a chain of hills nearly 8 km long, which is situated in the northern foreland of the Harz Mountains and which runs from west-north-west to east-south-east. In contrast to most of the other northern German salt domes the Asse is a relatively simple geological structure [2]. It is formed by an asymmetrical anticline of the Triassic age. The southern flank is nearly vertical, whereas the strata of the northern flank at first dip steeply and then flatten out to the north. The northern flank had been raised by 400 to 500 m in relation to the southern flank so the older strata crop out on the northern flank. Beneath a sedimentary mantle of Buntsandstein and Muschelkalk, some hundred metres thick, lies the salt anticline proper which is formed by strata of the German Zechstein series of the Permian age (fig. 1).

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In the Asse salt mine halite was exploited from 1916 to 1964. During this period 145 rooms were constructed in the older and younger Halite (Na 2 and Na 3) on 15 levels between 490 and 800 m deep. These rooms have a total volume of $3.5 \times 10^6 \text{ m}^3$. Only part of them can be used directly for the disposal of low- and intermediate-level radioactive wastes. A normal room in the younger Halite is 60 m long, 40 m wide and 15 m high. So its volume is about 36,000 m³.

2.2 Evaluation of Disposal Procedures

As on one hand no experience was existing on the disposal of radioactive wastes in salt deposits and on the other hand the salt mine Asse was intended for the central storage of all radioactive waste generated in the FRG, the working programme was from the very first moment defined in close cooperation with the Mining Authority acting as the principal licensing authority in accordance with the Ministry of Economics of the federal state of Lower Saxony. A step by step experimental programme was defined and executed after completion of the first safety analysis and necessary repair and adaptation work on above ground and underground installations.

As far as the definition of optimum working procedures for the disposal of radioactive waste is concerned, the work was devided according to the following three waste categories:

 Low Level Waste (LLW), i.e. radioactive waste that does not require additional shielding, surface dose rate < 200 mR/h.

- Intermediate Level Waste (LLW) that requires shielding during transport and handling, surface dose rate > 1 R/h, the activity content of which does, however, not lead to noticeable selfheating during storage.
- High Level Waste (HLW)that is selfheating and requires heavy shielding during transport and handling.

The experimental storage operations were mainly aiming at the investigation of technical feasible and safe waste handling techniques both above and under ground, the elimination of irradiation risks for the operating staff, the integrity of the stored waste in the storage rooms, the definition of waste conditioning standards complying both with the safety postulations for transport and for the storage site and the evaluation of the corresponding acceptance conditions, development of appropriate shielded transport containers, etc.

Apart from the experimental work devoted mainly to the storage operation research work is carried out on the rock mechanics of the salt mine and on additional geological and petrographical data. Furthermore an hydrogeological research programme and an extensive surveillance and monitoring programme are under way. Safety evaluation continues on the basis of the results obtained and the experience collected.

2.2.1 Disposal of Low Level Waste

Starting in 1967 a total of roughly 10,000 containers (mainly 200 l steel drums) containing solid and solidified LLW were stored in two existing mined out rooms at the 750 m-level during four experimental storage phases. Drums containing the waste were first stacked by 4 standing on top of each other and later piled lying horizontally, up to 10 on top of each other. During this period radiation exposure monitoring of personnel showed no excess of the maximum permissible doses. No serious **con**taminations took place, air monitoring showed always values lower than maximum permissible concentrations [4].

Based on the experience of the experimental storage-phases, acceptance conditions for LLW were worked out in 1971 [3]. Only solid or solidified wastes are accepted. An attempt was made to classify the activity levels contained in the different categories of waste in accordance with the quality of the waste and its container. Table 1 lists the permissible β , γ -activities for the individual categories of LLW. The acceptance conditions are part of the authorisation granted in 1971 by the Mining Authority for long term storage of LLW, that started on a **rout**ine basis **in** November 1971.

Since now a total of about 25,000 drums with LLW representing roughly 5,000 m^3 and a total activity of almost 1,000 Ci have been stored. Two rooms of the 750 m-level are filled and were closed by brickwork.

2.2.2 Disposal of Intermediate Level Waste

The category of intermediate-level waste comprises all those wastes whose external radiation exceeds 1 R/h. The upper limit of this category is reached when the waste becomes selfheating.

This waste category has to be transported and handled in shielded containers. Consequently prior to starting expe-

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Table 1: Permissible activity per 200 litres of waste (Ci) (LLW Acceptance conditions)

Waste category A:

Solid, dry wastes with activity loosely adhering or not bound (e.g. air filters, paper, textiles, ashes, dried evaporator concentrates, dried ion exchangers)

Group	Container: Treatment	Sheet-metal drum	Sheet-metal drum lined with con- crete (d \geq 5 cm) reinforced drum
1	Without treatment	0	0,2
2	Wastes fixed in concrete or equivalent	0,2	1

Waste category B:

Solid, dry wastes with activity firmly bound (e.g. activated metals, contaminated parts with firmly bound activity, solid sources in unbreakable containers)

Group	Container: Treatment	Sheet-metal drum	Sheet-metal drum lined with con- crete (d \geq 5 cm) reinforced drum
1	Without treatment	0	1
2	Wastes fixed in concrete or equivalent	1	5

Waste category C:

Wastes solidified in binding agents (e.g. sludges, evaporator concentrates, ion exchangers fixed in cement, bitumen or similar)

Group	Container: Treatment	Sheet-metal drum	Sheet-metal drum lined with con- crete (d \geq 5 cm) reinforced drum
1	Fixation in materials so- luble in wa- ter and brine	0	1
2	Fixation in materials in- soluble in water and brine	5	5

rimental disposal operations, shielded containers and appropriate manipulation techniques had to be developed.

Small quantities of intermediate-level wastes are transported to the mine in single-shielded containers that can receive one 200-1 drum. A shielded container (130 mm cast iron, weight 5.5 tonnes) is shown in Fig. 2.

For larger amounts the transport in single containers is uneconomical owing to the relatively large distances between most nuclear facilities and the Asse salt mine and to the unfavourable weight ratio between shielding and waste. A bulk transport container of cast steel with a variable shielding of eihter 150, 200, or 245 mm has been developed for this case. The total weight including the waste drums is about 25, 30, and 35 t, respectively. Increased shielding can be obtained by inserting the base container into one or two additional shieldings (Fig. 3). The shielding of the bulk transport container is sufficient for all wastes with a dose-rate up to some 10⁴ R/h, depending upon the energy of radiation. Waste with higher external radiation should be transported in a single container with 140-mmthick lead shielding.

The single shielded container is introduced in the mine directly through the shaft. Because of the limited capacity of the hoisting cage, the contents of the bulk transport containers must be reloaded into single containers above ground.

For storage, the single-shielded containers are first lowered to the 490 m level. Here, a crane removes them from the hoisting cage and transfers them on a drift car for transport to the loading room (Figs. 4 and 5). A 10 t crane lifts the shielded container from the drift car and places it on the radiation protection cover of the loading bore hole. Afterwards, the coupling of a 1 t crane is connected with the head of the drum grab; the bottom plate of the shielded container is opened simultaneously with the cover of the loading bore hole and the waste drum is lowered into the storage room.

After arrival at the top of the pile the drum grab is disengaged by means of a magnetic coupling and returned into the shielded container. Afterwards, the bore hole and the bottom plate of the shielded container are closed and the empty container is taken away. The whole process of storage can be followed by television. The first experimental intermediate-level wastes dumping by this technique started in September 1972. About 100 drums ($\sim 20 \text{ m}^3$) with a total activity of some 10³ Ci were successfully disposed of up till now.

Preliminary acceptance conditions for MLW have been evaluated which according to further experience gained in the experimental phase are due to revision prior to start up of routine disposal operation.

2.2.3 Disposal of High Level Wastes

No actual disposal of high-level wastes has been carried out up to now but a broad R + D-programme is carried on mainly at the Karlsruhe Nuclear Research Center, the Technical University Aachen, and at the Asse Salt Mine. High level waste, i.e. essentially the highly active fission product solutions resulting from reprocessing of irradiated fuel elements will be solidified by incorporation into

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glass [5]. Cylindrical glassblocks of roughly 25 1, sealed in stainless steel canisters (\emptyset :20 cm, height: 100 cm), the production of which is scheduled to start in 1976 will contain fission products in the order of 10⁵ to 10⁶ Ci with a decay heat production of up to 1 KW.

The area chosen for the disposal of high-level glass blocks is situated in the very depth of the mine. In this part of more than 775 m depth no old mine workings are existing. So the dimensions for the charging rooms can be properly designed. They are mainly governed by rock mechanical aspects and by the dimensions of equipment. The large mass of compact rock salt available to the depth offers to use boreholes several ten meters deep and to stack several blocks of solidified high-level waste on top of each other in a borehole. Positioning of these boreholes, i.e. the storage geometry is determined by the need of providing for appropriate heat dissipation.

Optimal storage geometry is actually being investigated by field experiments and computer calculations [6]. At the same time development of the transport and handling system is carried out.

First experimental storage of HLW is scheduled to start in 1976/1977, when approximately 50 to 100 glass blocks per year will arise from the solidification of the high level wastes from the reprocessing plant WAK in Karlsruhe. During the experimental phase sufficient experience will be gained to define an appropriate transport and handling system as well as the storage technology needed for the disposal of the HLW of a large commercial reprocessing plant that is scheduled to deliver HLW glass blocks during the third quarter of the eighties.

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2.3 Outlook

The execution of an extensive programme on the disposal of radioactive wastes in rock salt formations has up till now successfully demonstrated that this storage method ensures safe disposal of radioactive wastes eliminating any risk, of pollution of the biosphere. There is confidence that in continuing these efforts, the suitability of this technique for safe perpetual disposal will be verified further.

In view of the development of nuclear energy estimated for the Federal Republic of Germany, the storage capacity of the Asse salt mine will be sufficient to hold the radioactive waste generated up to the year 2,000. However, from time to time the technical installations of the mine will have to be adapted to the requirements of the increasing waste arisings. It is thus that the Asse salt mine provides all conditions for perpetual storage of all types of radioactive wastes in the future.

It is interesting to note in this connection, that recently activities started for the disposal of solid and solidified toxic conventional wastes from chemical industries in exhausted pottash mines, too [7].

3. DEEP-WELL DISPOSAL OF TRITIUM CONTAINING LIQUID EFFLUENTS

During the last years, deep-well injection has been applied on a steadily increasing scale for the disposal of heavily polluted and poisonous effluents originating for instance, from mineral oil production and chemical industry.

Apart from oil-field brine rejection, more than 500 million m^3 of brine from potash works have been disposed of successfully into some 30 deep wells of 200 to 700 m depth in the Werra potash district of Germany; present injection rates average roughly 1 m^3 /sec (8). A deep well for injection of effluents from fuller earth production is in operation in Moosburg, Bavaria (9).

In an attempt to establish a disposal procedure for tritium-containing liquid effluents, that does not lead to a pollution of the biosphere, it was decided at the Karlsruhe Nuclear Research Center to study the possibility of disposing of this type of effluents by deep-well injection (10). In an R + D programme sponsored by the Federal Ministry of Education and Science this disposal technique will be tested for the first time in the Federal Republic of Germany by experimental injections of real T-containing effluents. This programme aims at the establishment and demonstration of operation criteria for the safe and large-scale application of this disposal technique.

The Karlsruhe Nuclear Research Center is best suited for these demonstration experiments, since favourable geological conditions allow the preparation of an experimental deep well in the immediate vicinity of the site, utilizing the still existing borehole of a depleted oil reservoir. Furthermore - for the time being - the largest amounts of T-containing effluents of the Federal Republic of Germany are produced in Karlsruhe, it is thus that apart from the demonstration of a new pollution-free disposal technique, the experimental injection programme has the direct practical advantage of considerable reducing the T-releases of the Center into the main canal.

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3.1 Description of the Oil Field and the Well selected

The oil field in the vicinity of the Nuclear Research Center Karlsruhe extends in north-south direction and has an extension of approx. 3,000 m x 600 m. The stratigraphic column may be characterized by the sequence of well No. 1 shown in figure 6.

The oil field ic characterized by a lenticular structure in the following main, oil- bearing strata: Coloured Niederröderner layer, Cyrena marls and Meletta layer from which crude oil is produced by the oil industry (C. Deilmann AG, Wintershall AG).

Since startup of the field operation in 1957 a total of roughly 135,000 t of crude oil has been pumped out from the sandstone parts of these three strata. At the same time, approximately 240,000 m³ of oil-field brines brought to the surface during **crude** oil production were reinjected into the coloured Niederröderner layer and the Meletta layer. Crude oil production is still continuing through 10 wells. Yearly production rates are, however, steadily decreasing, averaging for the time being roughly 6,000 t of crude oil/year. Complete depletion is expected within a few years.

The lenticular structure has been considered to offer versatile advantages for the performance of the anticipated deep-well injection tests with T-containing effluents:

- Safe isolation from the main aquifer is guaranteed.
- Injection of effluents up to the amount of crude oil withdrawn from the different carriers would not change the original geological conditions.

- Injection into an exhausted isolated stratum with the crude oil production continued would allow the observation of any migration eventually occurring in the deep underground between the different oil-bearing strata still in operation.
- Utilization of an existing well would considerable decrease the cost, as new wells have not to be drilled.

Therefore, the decision was in favour of the depleted isolated oil lense of well Lh 2, the sandstone of which (-934.5 m to -945.5 m) was originally filled with crude oil and brine at an initial pressure of 99.5 atm. relative to the top of the reservoir. The production characteristics of the Lh 2 confirmed the lense-like structure of the sandstone formation, as the pumping rate decreased continually and at the same time the liquid level in the well went down to roughly 50 m above the top of the reservoir. The bottom hole pressure in the sandstone lense was estimated to range between 40 and 50 atm. when oil production stopped and the well was closed by a bridge plug at -912.5 m in 1963. At that date a total of 23,000 m³ crude oil and brine had been withdrawn from the oil lense.

3.2 Technical Adaptation of the Well

Conception of the measures needed for the adaptation of the Lh 2 well was performed on the basis of providing the utmost precautions to safely exclude any T-contamination of the biosphere during the injection tests. Prior to starting the technical realization an application for the approval of an operation plan for the project containing the description of the project, the operating conditions, the conception of the adaptation measures and the conception of the control and monitoring provisions was placed with the Mining Authority in June 1971, and a safety analysis was sent to the Ministry of Economics in December 1971.

According to legislation, the Mining Authority of Baden Württemberg is acting as the principal licensing authority granting the authorization for the project in close cooperation with the competent Ministries of the state of Baden Württemberg, i.e. the Ministry of Agriculture and Environmental Protection, the Ministry of Economics, and the Ministry of Labour and Social Affairs.

After several lenghty discussions with the authorities concerned, the Mining survey granted a partial authorization for the technical adaptation of the well at the end of May 1972.

Underground adaptation operation according to the conception shown in figure 7 was started early in June 1972. First cement bond log measurements, squeeze cementations of old perforations, and the elimination of the old cement plug and the bridge plug were performed.

In order to increase the absorption capacity of the injection strata, an additional perforation was executed between -934.5 m and -945.5 m. Afterwards the 2 7/8 in injection tubing was mounted, each tubing connection being tested for leak tightness at 280 atm. After filling the annulus (6 5/8" - 27/8") with some 17 m³ of corrosion inhibitor the Baker-lock-set-plug was placed together with the sealing connector. To clean out the injection strata in the immediate vivinity of the perforation, some 50 m³ of liquid were withdrawn from the sandstone lense by swabbing. Measurement of the formation pressure in the lense revealed roughly 77 atm. which indicates that the effective volume of effluents that can be safely injected into the lense without surpassing the original formation pressure, amounts to approximately 11,000 m³. First brine-injection tests showed injection rates of 8.5 m³/h for pumping pressures between 60 and 80 atm.

As an additional safety measure a cement bond log and a squeeze cementation was performed at a borehole situated roughly 35 m south of the injection well that continues oil production from the Meletta sandstone stratum at -1,225 m.

The underground adaptation was completed successfully in mid-August 72.

Surface adaptation of the well comprises mainly leaktight plastic lining of the cellar, construction of a concrete trough with leak-tight plastic lining, installation and connection of an Oilwell-Triplex pump, construction of a mobile light-metal shed equipped with a ventilator (air renewal 10 times per hour) and construction of a fence of 2 m height. Realization has been delayed due to a still pending water-law authorization.

3.3 Anticipated Operation Procedure

The deep-well injection tests will be performed with the condensates from the high-level waste- and acid recovery evaporators of the reprocessing plant WAK arising at roughly 500 m³/y with an average T-activity of 20 Ci/m³.

Other radionuclides are only tolerated up to MPC-values eventually necessitating further evaporation in the waste treatment station of the Center. Batchwise injection of the T-containing condensates will be performed directly from the tank truck which transports 10 m³ of charges to the deep well. Injection pressures will be limited to \leq 90 atm. in order not to surpass the fracturing pressure of the sandstone lense. After each injection the liquid level in the well will be lowered by injecting an appropriate amount of brine from oil production, in order to avoid any T-atmosphere in the upper part of the bore hole. Injections will be stopped as soon as the original pressure of the oil lense has been reestablished.

During injection operation, the site of well Lh 2 is temporarily declared a controlled area. Health and safety specialists ensure surveillance on the well site during operation by measurement of the T-concentration of air and eventual contaminations. Staff participating in the operations are controlled for T-ingestion on a routine basis.

In order to examine whether T-migration is occurring from the isolated sandstone lense of Lh 2 into other oil-bearing horizons, the oil production of the 10 boreholes still in operation will be checked for T-contamination on a routine basis.

In addition to an extensive activity surveillance programme performed by the Nuclear Research Center on a routine basis, the water of two water table inspection wells in the immediate vicinity of the injection well Lh 2 will be subjected to routine tritium checking.

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3.4 <u>Safety considerations</u>

Contaminations of the ground water during injection is excluded due to the fact that at least triple barriers are provided towards the water table. Any effluent leak would during pumping collect in the sump of the bore cellar from where it would be taken up again by a sump pump. Leaks occurring in the injection and casing tubings would be detected immediately by the recording pressure control instruments of the injection tubing and the anti-corrosion liquid in the annulus. As a result of the reduced pressure in the sandstone lense, leaks occurring in the injection tubing in periods of non-operation could only lead to a migration of the anticorrosion liquid from the annulus into the injection tubing.

Due to the fact, that the amount of T-containing effluents to be injected under the selected injection conditions is limited to the volume for which the original pressure conditions in the horizon will be reestablished, a disturbance of the **natural** geological isolation of the horizon can be excluded with certainty.

Due to the favourable geological conditions and the technical provisions taken, it can be concluded that a contamination of the oil field and the water table is to be excluded. Even for the case that a contamination of the water table is still feared, this could only occur by diffusion through fissures and crevices from the 940 m deep horizon into the water table situated roughly 700 m higher. Diffusion over such a distance would, however, require a period of time which is more than 3 decimal powers longer than the half-life of T; thus, this nuclide would have decayed long before reaching the water table.

3.5 Status of the Project

As already mentioned in section 3.3, underground adaptation has been terminated successfully. Provided that the still pending authorizations will be granted within the following quarter, startup of the injection experiments is scheduled for mid 1973.

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Shielded container for medium level wastes

Fig. 2



Multiple transport container for medium level wastes with Fig. 3 transfer container on the top



storage of medium level radioactive wastes generale view

Fig. 4





Fig. 5





figure 7: Conception of underground adaptation for Lh 2

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