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DEEP-WELL DISPOSAL OF TRITIUM CONTAINING LIQUID EFFLUENTS

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Kurzfassung

Radioaktive Abwässer können über Schluckbrunnen in Aquiferspeicher oder ähnliche Strukturen des tiefen Untergrunds eingebracht werden. Das Verfahren vermeidet jede Belastung der Biosphäre. Im Kernforschungszentrum Karlsruhe wird die Möglichkeit der Versenkung tritiumhaltiger Abwässer in isolierte, erschöpfte Öl-Linsen untersucht. Die vorliegende Arbeit stellt das Versuchsprogramm vor und gibt einen Überblick über die geologischen Gegebenheiten des in unmittelbarer Nähe des Zentrums gelegenen Ölfeldes, die technischen Einzelheiten des Schluckbrunnens und seiner Übertageanlagen, den vorgesehenen Betriebsablauf sowie die Überwachungsmaßnahmen und die Sicherheitsaspekte.

Abstract

At the Karlsruhe Nuclear Research Center deep-well injection of tritium-containing effluents into isolated depleted oil horizons of the deep underground is being studied as an approach of tritium disposal, which does not lead to a pollution of the biosphere. The geology of the oil field in the vicinity of the Center, the technical underground and surface adaptations of the selected well, the control and surveillance provisions are described together with the essential safety considerations and the anticipated operation conditions.

1. INTRODUCTION

1.1 The Tritium Problem

Decontamination techniques as already developed for and actually being applied in the treatment of radioactive wastes from nuclear installations are, generally, so effective that only negligible amounts of radionuclides are discharged into the environment. In addition, extensive worldwide R + D effort continues to provide further improvements to today's liquid waste management. This is why any hazard to the population by radioactive effluents can be excluded even in case of the expected tremendous expansion of nuclear energy.

Tritium, however, is an exception, as it cannot be separated by any of the liquid waste treatment procedures known and actually applied in routine operation. Due to its relatively low radiotoxicity,this radionuclide, after appropriate dilution, is actually being discharged to the environment by all nuclear installations without representing any particular hazard.

In addition to the natural T-formation by cosmic rays, nuclear energy produces this radionuclide both by various activation processes with light nuclei and by ternary fission. Taking into account the worldwide expansion of nuclear energy, there is a general impact towards revision of the actual release practice for tritium to be produced in the nuclear power reactors and reprocessing stations of the future. Consequently, numerous theoretical studies were performed on an international level in the attempt to assess the local and worldwide radiological hazards in connection with this particular problem.

Recently, BOEHNERT and BONKA (1) have shown that the maintenance of this release practice up till the year 2,ooo will not lead to a general T-problem, but most probably create local problems: Assuming worldwide homogeneous dilution by blending these authors were able to show that the resulting T-concentrations are by 5 orders of magnitude lower than actual MPC-values (Fig. 1); considering, however, the Federal Republic of Germany as an isolated and closed space, homogeneous dilution and blending of the tritium production expected up till the year 2,000 in the Federal Republic of Germany would lead almost to MPC-values (Fig. 2).

As the major part of tritium is released in the form of tritiated water (HTO, T_2O), the Karlsruhe Nuclear Research Center is studying other possibilities of establishing an alternative disposal procedure for T-containing effluents, that does not lead to a pollution of the biosphere.

1.2 Deep-Well Injection

Injection of the T-containing effluents into isolated aquifers or depleted oil horizons of the deep underground offers an interesting alternative approach to disposal. 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.

Although deep-well injection had been used for a long time by petroleum companies for rejection of oil-field brines brought to the surface in crude oil production, it is only since about 1960 that the use of this technique for disposing of industrial wastes has been increasingly used.

In the United States, for instance, some hundred injection wells were drilled and are actually in operation in depths between 60 m and 3,600 m (2, 3, 4, 5).

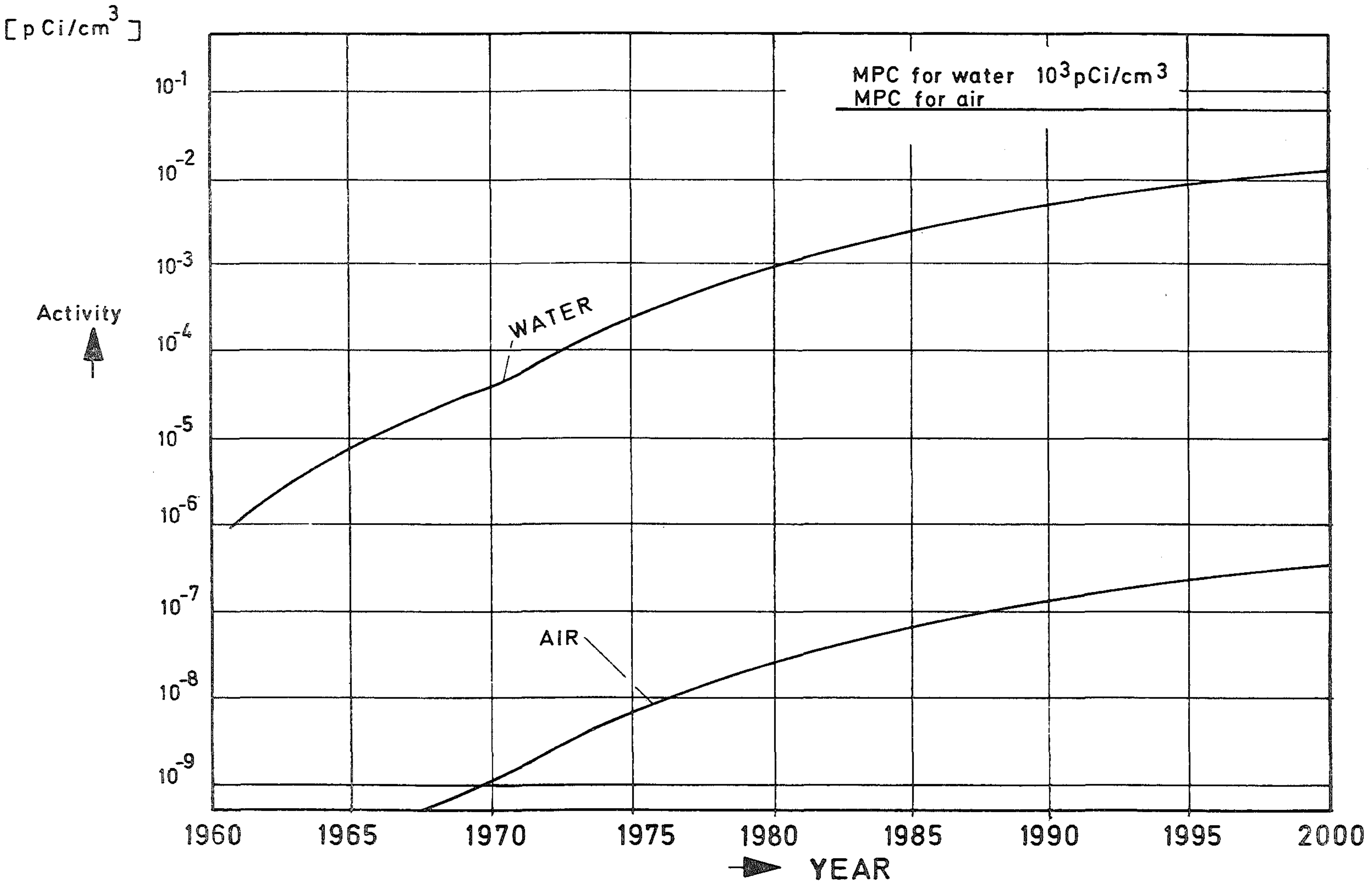


FIGURE 1 T-ACTIVITY IN WATER AND AIR FOR COMPLETE T-RELEASE FROM WESTERN NUCLER ENERGY FOR HOMOGENEOUS WORLDWIDE DISTRIBUTION

(From Boehnert R., Bonka H., Jül-763-RG (1971)

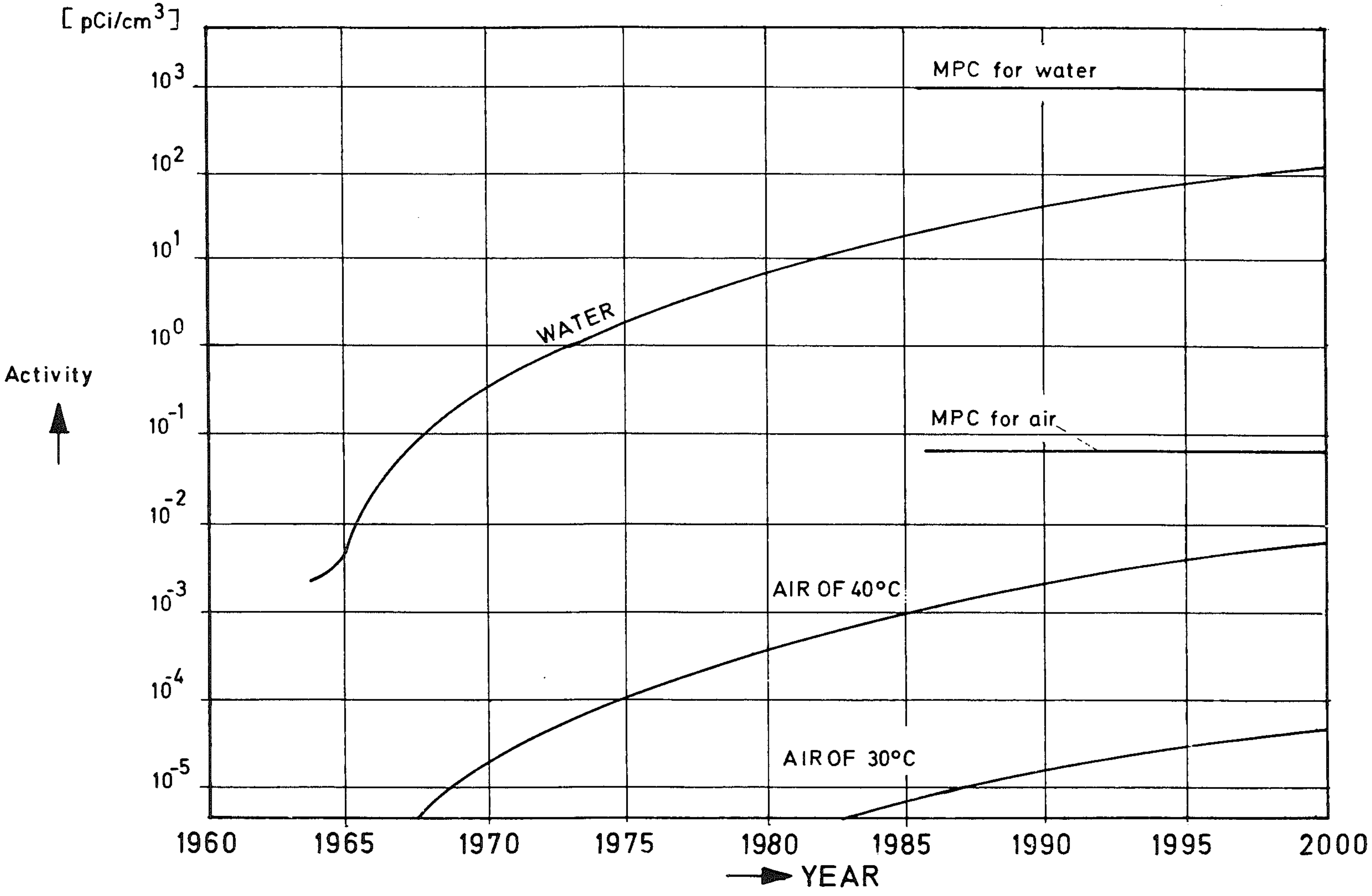


FIGURE 2 T-ACTIVITY IN WATER AND AIR FOR COMPLETE T-RELEASE FROM GERMAN NUCLEAR ENERGY FOR HOMOGENEOUS DISTRIBUTION IN THE FEDERAL REPUBLIC OF GERMANY (From Boehnert R., Bonka H., Jül-763-RG (1971)

Apart from oil-field brine rejection, more than 5oo million m^3 of brine from potash works have been disposed of successfully into some 3o deep wells of 2oo to 7oo m depth in the Werra potash district of Germany; present injection rates average roughly 1 m^3/sec (6). A deep well for injection of effluents from the chemical industry is in operation in Moosburg (Bavaria).

In Melekess (USSR) approximately 1.2 million m^3 of radioactive wastes with a total ß-activity of roughly 5o million curies were disposed of between 1963 and 197o into 5 injection wells of about 4oo m depth at an average daily injection rate of 35o to 4oo m^3 (7).

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. 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:

The annual production from the various nuclear test reactors amounts to roughly 1,ooo m^3 with an average specific T-activity of 1 Ci/m^3; since the startup of the first German reprocessing plant WAK in late 1971 another 5oo m^3/year of roughly 2o Ci T/m^3 are scheduled to be produced in the HLW evaporation- and acid recovery step of this plant. The demonstration injections will be performed with the latter waste stream.

Apart from the demonstration of a new pollution-free disposal technique, the experimental injection programme has the direct practical advantage of considerably reducing the T-releases of the Center into the main canal.

2. THE OIL FIELD CHOSEN FOR THE INJECTION EXPERIMENTS

2.1 General Description

Figure 3 gives a general outline of the oil field Leopoldshafen in the vicinity of the Nuclear Research Center Karlsruhe. As indicated in this figure, the oil field extends in north-south direction and has an extension of approx. 3,ooo m x 6oo m. The oil has been found in the tertiary formation, the structure is an elongated anticline cut off in the west by an antithetic fault.

The stratigraphic column of the oil field may be characterized by the following sequence of well No. 1 (Figure 4):

- 19o m: Quarternary and pliocene strata (ground water bearing), divided in an upper gravel layer ($\approx$ 3o m), an intermediate sand layer ($\approx$ 3o m) and a lower sand layer with marl intercallations ($\approx$ 13o m).

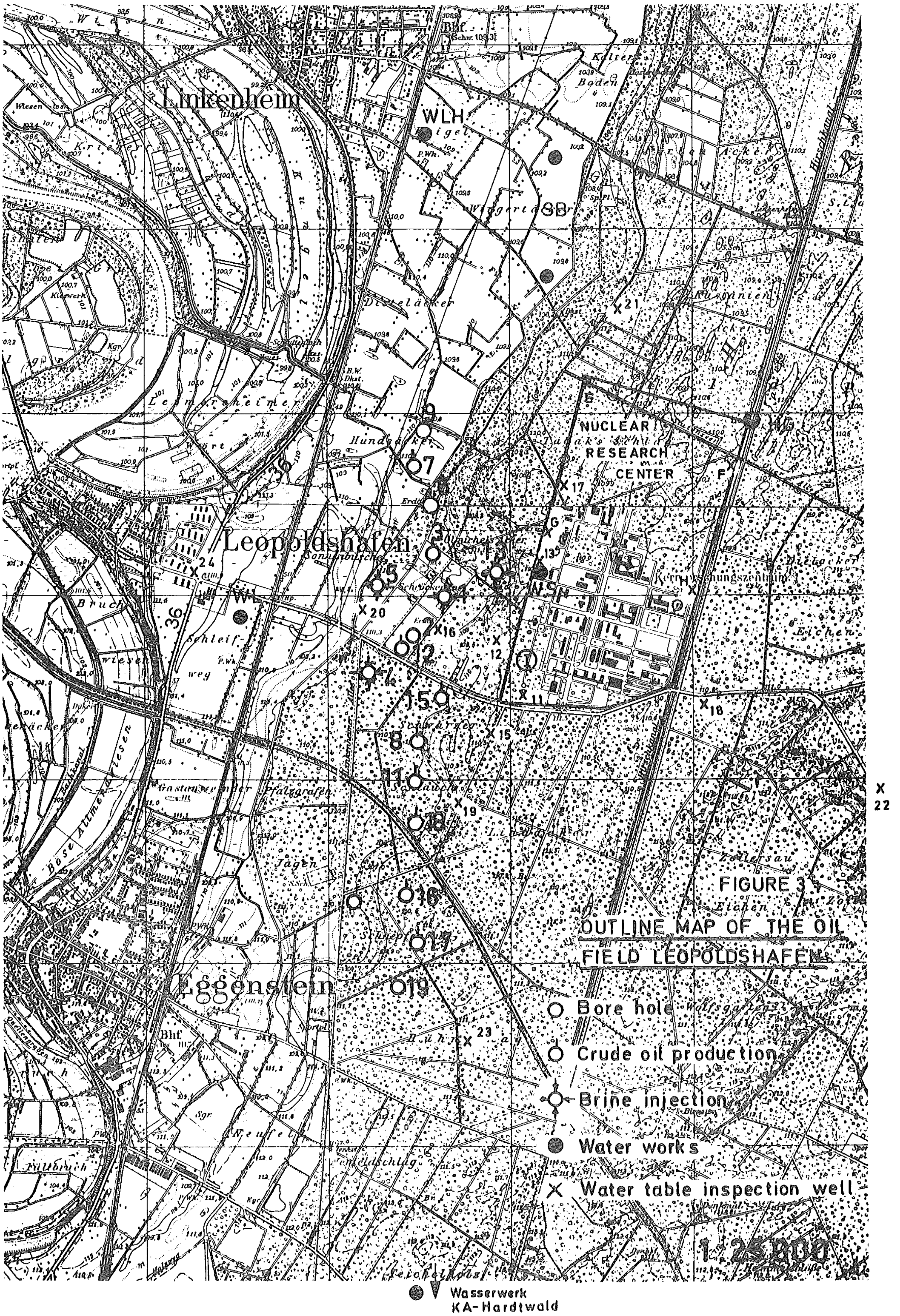
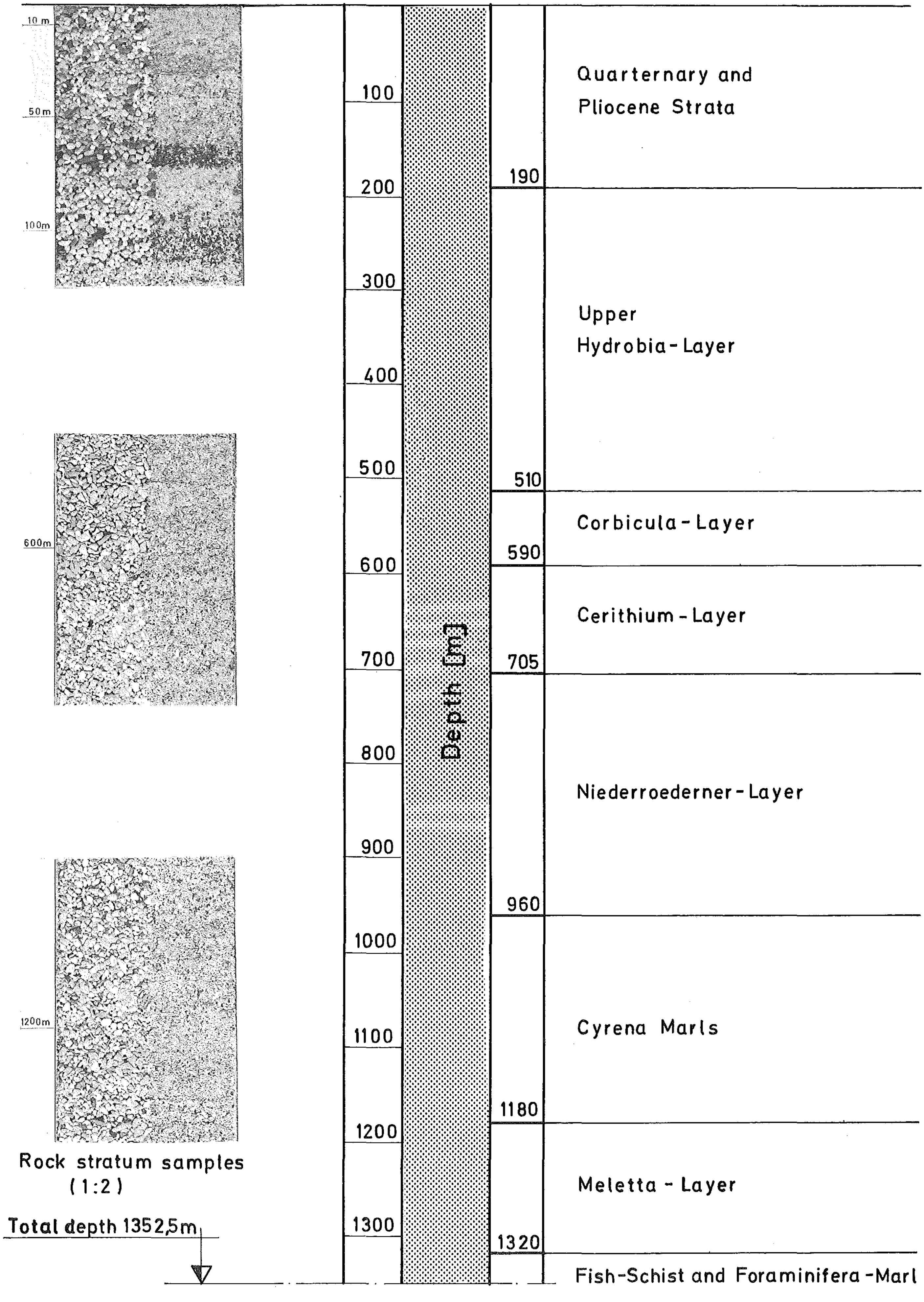


FIGURE 3

OUTLINE MAP OF THE OIL FIELD LEOPOLDSHAFEN

Figure 4 Bore hole Nr 1 (1:5000)



- 51o m: Upper Hydrobia layer, mainly consisting of grey marly clay with many small limestone and dolomite seams, and some sandstone intercallations.
- 59o m: Corbicula layer, of dark grey laminated marls, partly bituminous with some anhydrite.
- 7o5 m: Cerithium layer of greenish clay and marl.
- 96o m: Coloured Niederröderner layer consisting of coloured marls and clays with dolomite.
- 118o m: Cyrena marls, of grey marls with limestone and sandstone.
- 132o m: Meletta layer, of similar composition as the Cyrena marls.

Below these layers follow the fish-schist and Foraminifera-marl (≈ 2o m), the Pechelbronner layers of clay with lens-shaped bodies of sandstone (≈ 2oo m) and the Lymnaea marls, a marly clay layer extending to approximately 2,ooo m of depth.

The oil field is 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 the startup of the field operation in 1957 a total of roughly 135,ooo t of crude oil has been pumped out from the sandstone parts of these three strata. At the same time, approximately 24o,ooo m^3 of oil-field brines brought to the surface during crude oil production were reinjected into the coloured Niederröderner layer and the Meletta layer through the wells 1, 4, 5 and 13 (cf. Fig. 3).

As indicated in Figure 3, crude oil production is still continuing through 1o wells. Yearly production rates are, however, steadily decreasing, averaging for the time being roughly 6,ooo t of crude oil/year. The complete depletion of the oil field is expected within a few years.

2.2 Selection of a Depleted Oil Reservoir

As already indicated in 2.1, the oil field is characterized by a lenticular structure in the three main oil bearing strata. This particular situation 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 considerably decrease the cost, as new wells have not to be drilled.

Therefore, the decision was in favour of a depleted oil stratum, preferably an isolated oil lense. After thorough investigation of the problem performed in close cooperation with geologists from the oil producing firms, the University of Heidelberg, and the Geological Survey of Baden-Württemberg ,the final choice was taken for well No. 2 (Lh 2) of the oil field (see Fig. 3).

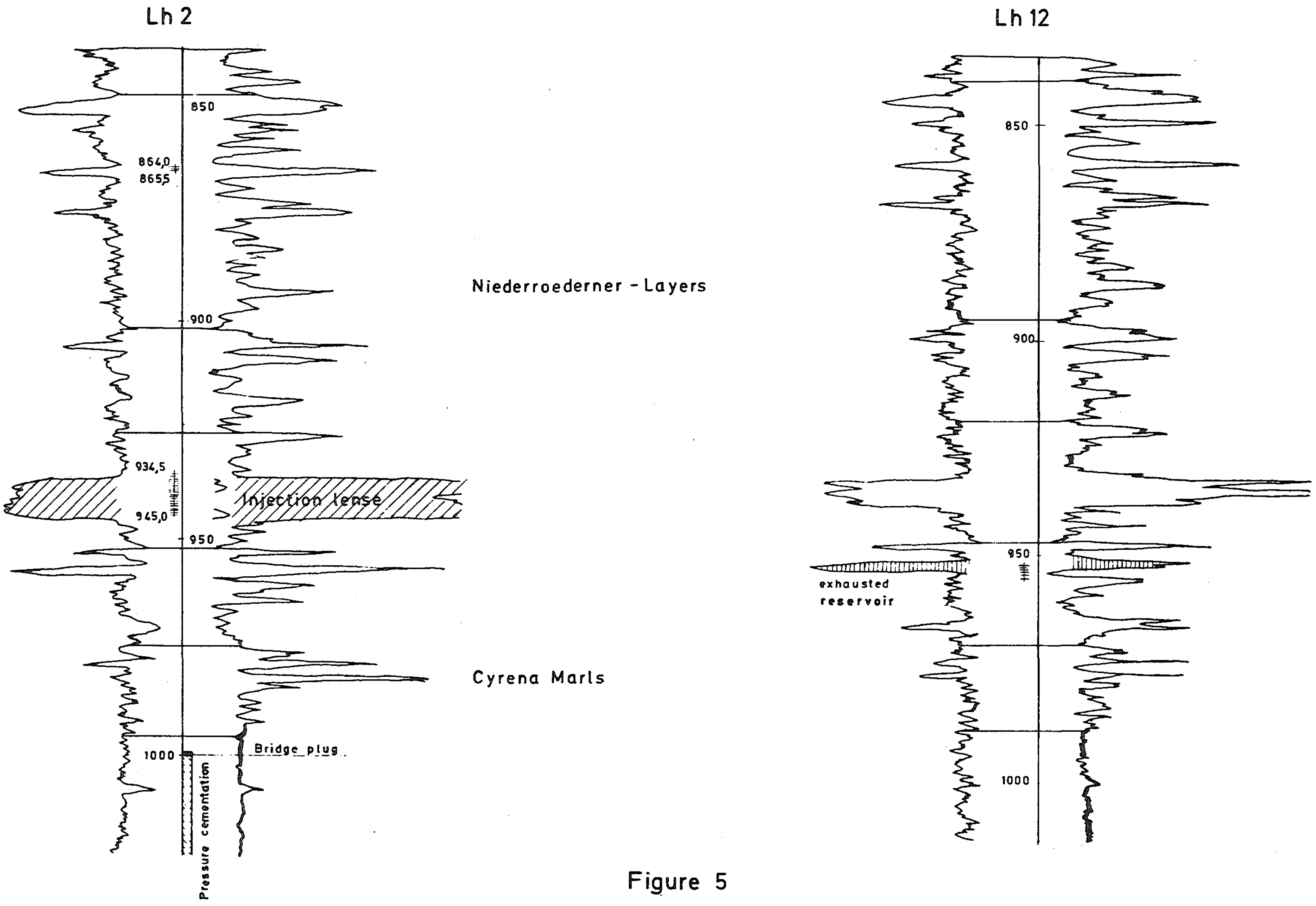


Figure 5
"ES" diagrams of the bore holes (1:1000)

The sandstone provided for the injection is 11 m thick (-934.5 m to -945.5 m) in the upper region of the Cyrena-marls at the Lh 2. As can be deduced from the electric spontaneous potential log (E.S.) diagrams of the wells in Figure 5, this sandstone is developed as a lense that can be found again only in the well No. 12 (Lh 12, see Fig. 3) situated some 35 m south of Lh 2, where this same sandstone is only 7.5 m thick and shows increased intercallation of clay-layers.

The sandstone lense 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: Whereas initially roughly 2o m^3 of liquid (crude oil and brine) were pumped out per day at a liquid level of approximately -1oo m in the well, the pumping rate decreased continually to about 5 m^3/day and the liquid level in the well went down to -89o m, i.e. roughly 5o m above the top of the reservoir. According to this liquid level and the casing pressure (41 atm), the bottom hole pressure in the sandstone lense was estimated to range between 4o and 5o 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,ooo m^3 crude oil and brine had been withdrawn from the oil lense. Assuming that no secondary infiltration occured in the meantime, it was estimated that the same volume could be safely replaced by reinjecting the T-containing effluents.

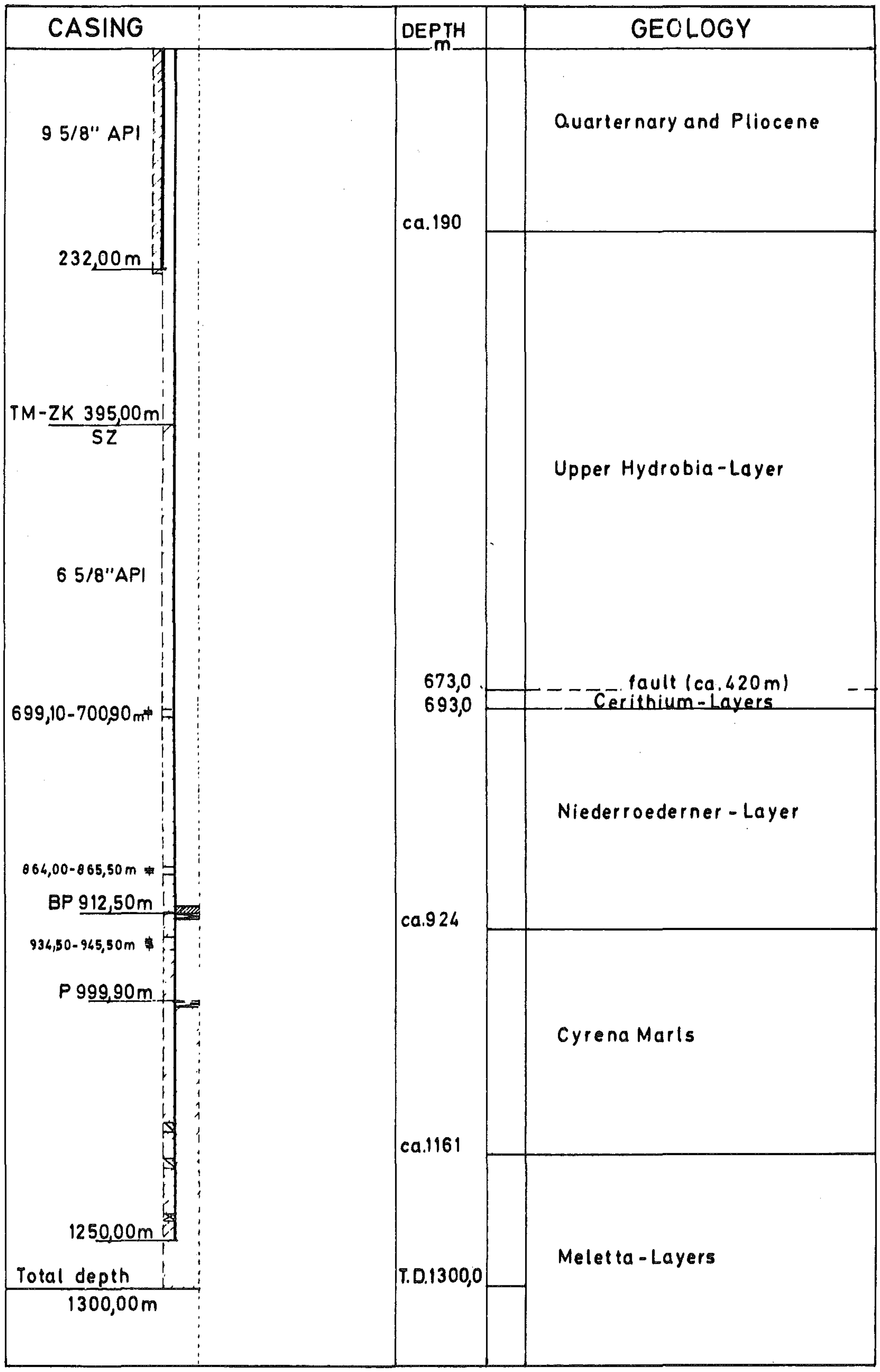


Figure 6: Status of Lh2 bore hole after oil production

3. TECHNICAL ADAPTATION OF THE WELL FOR DEEP-WELL INJECTIONS

3.1 Conception of Adaptation Measures

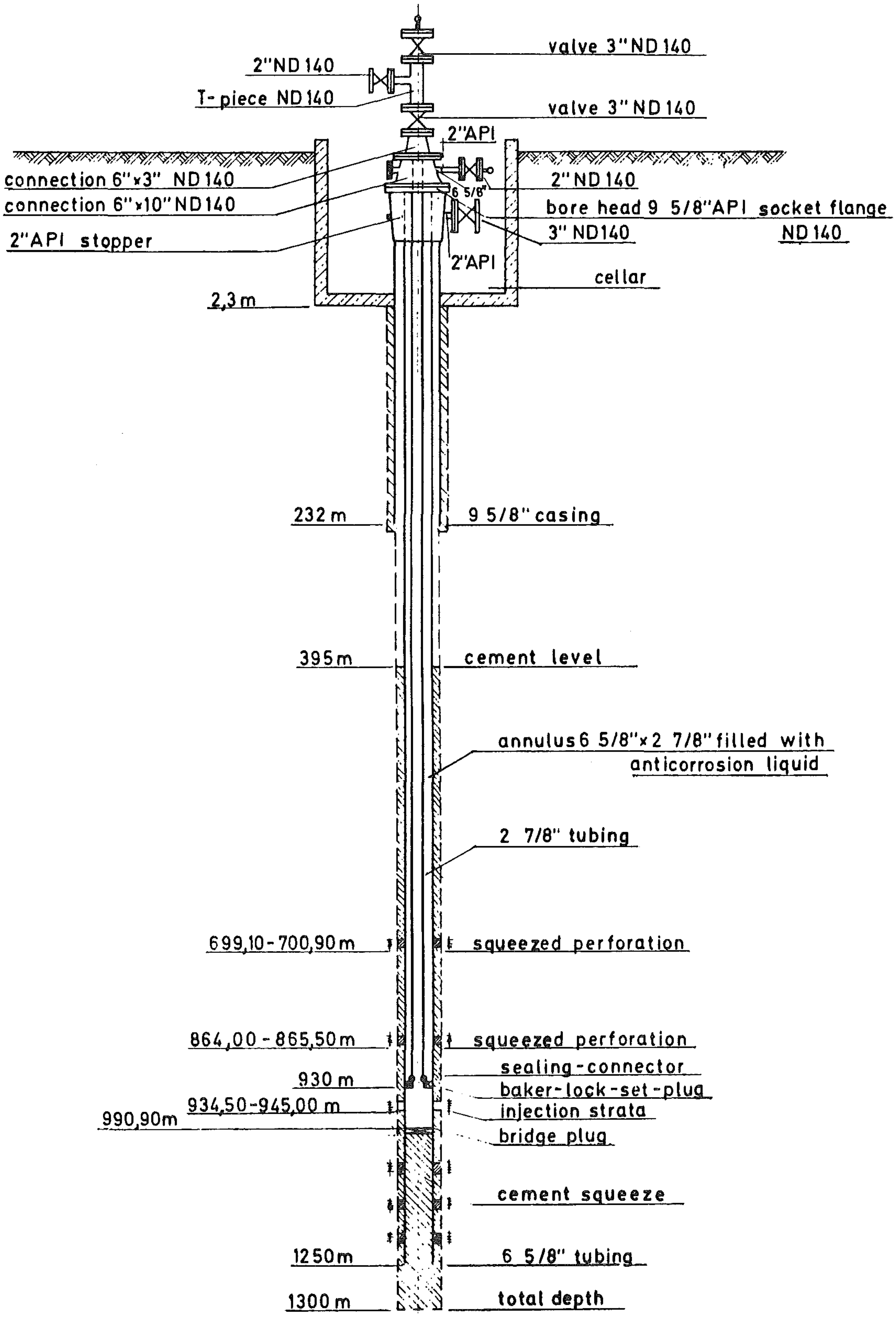
Conception of the measures needed for the adaptation of the Lh 2 well was performed on the basis of providing the utmost precautions to safely exclude any T-contamination of the biosphere during the injection tests.

3.1.1 Anticipated Operation Procedure

As already mentioned, the deep-well injection tests will be performed with the condensates from the high-level waste- and acid recovery evaporators of WAK arising at roughly 5oo m^3/a with an average T-activity of 2o Ci/m^3. 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 1o m^3 of charges to the deep well. Injection pressures will be limited to $\leq$ 9o 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.

3.1.2 Status of the Well

Figure 6 shows the status of the well after stopping the oil production activities. From its end depth at -1,3oo m to roughly -1,ooo m the bore hole is closed by cementation and a bridge plug. At -912,5 m another bridge plug headed figure 7: Conception of underground adaptation for Lh 2



by a 4 m high cementation closes the perforation of the injection structure between -934.5 m and 945.5 m. A cemented casing of 9 5/8 in. diameter (7.9 mm wall thickness) separates the bore hole down to -232 m from the water table extending to approximately -18o m. In this casing a production string of 6 5/8 in. diameter (7.3 mm wall thickness) is inserted which is cemented between -395 m and-1,25o m. Two perforations are existing in the coloured Niederröderner layers ranging from -699.1 m to -7oo.9 m and from -864 m to -865.5 m.

3.1.3 Conception of Underground Adaptation

Taking into account the anticipated operation procedure and the status of the well, conception of the underground adaptation comprised the following measures (Fig. 7):

- Control of casing cementation by cement bond log measurement.
- Closing of the perforations at -864 m to -865.5 m and at -699.1 m to -7oo.9 m by squeeze cementation .
- Elimination of cement residues in bore hole by drilling, scraping and circulating.
- Elimination of cement plug and bridge plug at -912.5 m by drilling, scraping and circulating.
- Control of squeeze cementation and casing cementation between -912 m and -1,ooo m by another cement bond log.
- Eventually amelioration of insufficient casing cementation by additional squeeze cementations after appropriate perforation of casing.
- Reperforation of the space between -934.5 and -945.5 m.
- Providing an injection tubing of 2 7/8 in. diameter and 5.5 mm wall thickness. Each tubing connection is checked

for leak tightness through individual pressure tests.

- Filling the annulus between casing and injection tubing with an anti-corrosion liquid.
- Closing the annulus between casing and the injection tubing by setting a Baker-lok-set-plug with sealing connector at -930 m depth.
- Mounting of a X-mas tree 3" x 2" at the well head (pressure standard 140 atm.; 600 ASA).

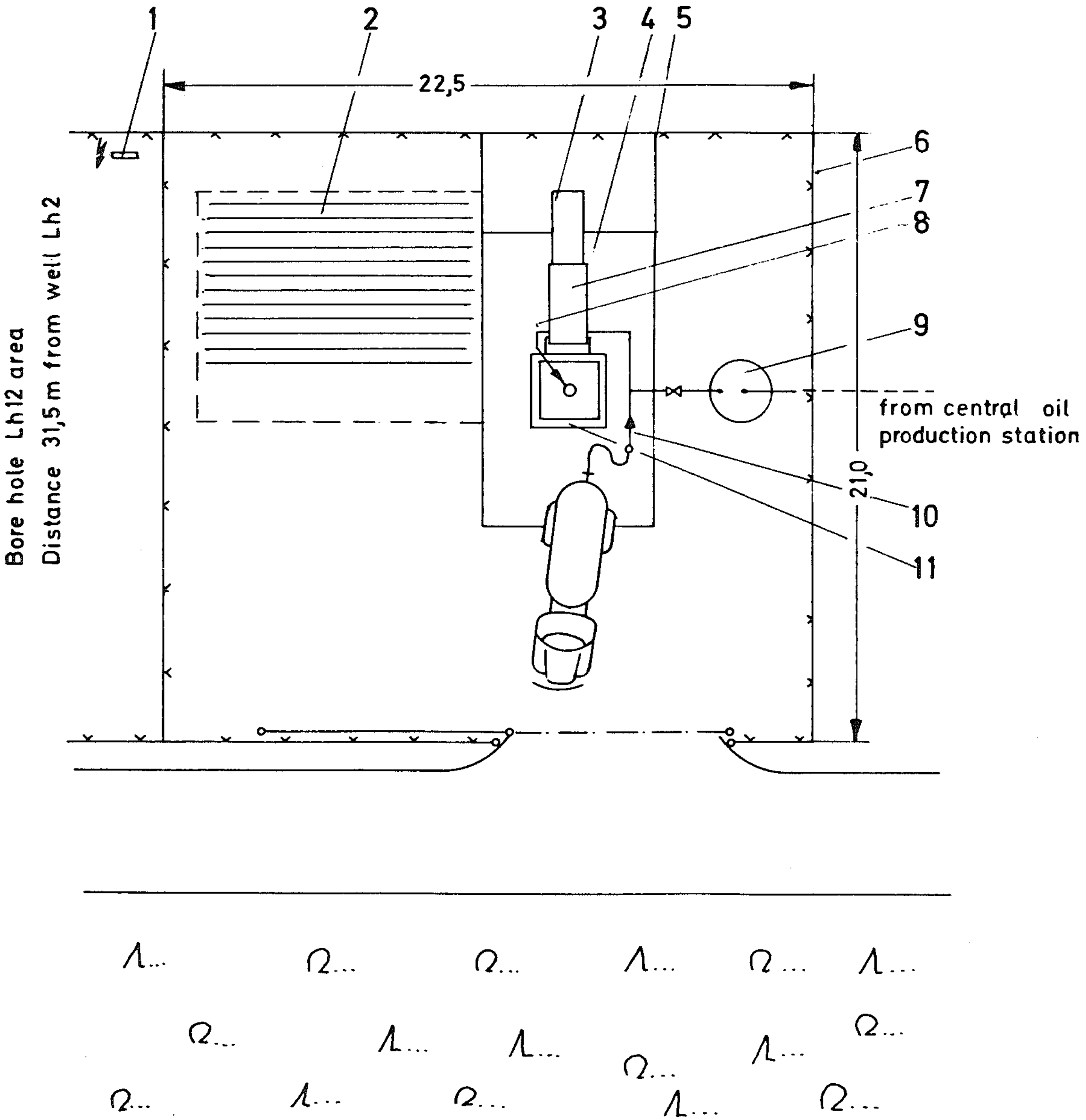
3.1.4 Conception of Surface Adaptation

As schematically indicated in Figure 8 the conception of the above ground adaptation measures comprised:

- Sealing of cellar by leak-tight plastic lining.
- Construction of a concrete trough with leak-tight plastic lining for tank truck positioning during injection.
- Installation of an Oilwell-Triplex-pump and appropriate tubing connection (pressure standard 140 atm).
- Installation and connection of an oil-field brine storage tank of approximately 3 m^3 of volume for the anticipated lowering of the effluent level in the injection tubing.
- Construction of a mobile light-metal shed equipped with a ventilator which allows to renew the air 10 times per hour. (The shed must be mobile in order to allow hoist operation for maintenance and inspection).
- Construction of a fence of 2 m height around the well Lh 2.

3.2 Licensing Procedure

According to legislation, the Mining Survey of Baden Württemberg is acting as the principal licensing authority gran-



1	Electrical power supply
2	Tubing rack
3	Pump foundation
4	Concrete trough+moveable shed
5	Rails for moveable shed
6	Fence
7	Pump
8	Pressure line
9	Brine storage tank
10	Suction line
11	Cellar

Figure 8
Schematic outline of above ground adaptation

ting the authorization for the project in close cooperation with the competent Ministries of the state of Baden Württemberg, i.e. the Ministry of the Interior, the Ministry of Economics, and the Ministry of Labour and Social Affairs.

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 Survey in June 1971. In addition, a safety analysis was sent to the Ministry of Economics in December 1971.

After several lengthy discussions with the authorities concerned, the Mining survey granted a partial authorization for the technical underground and surface adaptation of the well at the end of May 1972. In July 1972, a draft for the nuclear license was issued by the Ministry of Economics and distributed as an invitation to comments. Final authorization is exspected for the end of 1972.

3.3 Execution of Underground Adaptation

After receipt of the above-mentioned authorization for the technical adaptation of the Lh 2, underground adaptation operation was started early in June 1972. As expected, the first cement bond log measurement showed satisfactory casing cementation and the perforations at -864 m to -865.5 and at -699.1 m to -7oo.9 m were successfully closed by squeeze cementation with 4.8 m^3 and 6.5 m^3 cement slurry. Elimination of the cement residues did not raise any problems, whereas the elimination of the cement plug and the bridge plug at -912 m was rather difficult due to detriments of the perforating gun left in the borehole from former perforation operations. After a second cement bond log measurement an additional perforation and squeeze cementation was performed between -954 m and -957 m.

In order to increase the absorption capacity of the injection strata, an additional perforation by 136 shots 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" - 2 7/8") with some 17 m^3 of corrosion inhibitor the Baker-lock-set-plug was placed together with the sealing connector.

To clean out the injection strata in the immediate vicinity of the perforation, some 50 m^3 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 m^3. First brine-injection tests showed injection rates of 8.5 m^3/h for pumping pressures between 60 and 80 atm.

As an additional safety measure a cement bond log of the casing of the Lh 12 borehole situated roughly 35 m south of the Lh 2 was performed (see Fig. 3). Furthermore, the perforation at -952 m to -954.4 m of the upper Lh 12 oil lense (see Figure 5) from which 16,500 m^3 of crude oil and brine in total were withdrawn until depletion, was closed by squeeze cementation with 5.6 m^3 cement slurry. The Lh 12 is now continuing oil production from the Meletta sandstone stratum at -1,225 m.

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

4. CONTROL AND SURVEILLANCE PROVISIONS

4.1 Operation of the Deep-Well

The injection operations (see section 3.1.1) including truck transport, pump connection, injection, brine flushing etc., are performed by staff of the waste treatment department of the Nuclear Research Center. During injection operation, the site of the well Lh 2 is temporarily declared a control area, and the ventilator is operated. Health and safety specialists ensure surveillance on the well site during operation by measurement of the T-concentration of air and eventual contaminations. After completion of the injection operations the ventilator is stopped, the shed and the fence are locked and the control area is no longer maintained. Staff participating in the operations are controlled for T-ingestion on a routine basis.

4.2 Control of the Oil Field

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 1o boreholes still in operation (see Fig. 3) will be checked for T-contamination on a routine basis. The production of the Lh 12 in the immediate vicinity of the injection well Lh 2 will be checked monthly, the other 9 wells will be checked at 3 months intervals.

As mainly crude-oil/brine emulsions are produced, a special thermal treatment technique has been developed for separating the crude oil. The brine is redistilled and the condensate measured for T. Part of the oil phase is catalytically combusted in a special furnace and the resulting aqueous phase is measured, too. All operations, starting from field sampling up to T-measurement, are performed in an inert atmosphere to exclude T-contamination from the atmosphere.

First background control measurements showed that no tritium was present in the condensate from oil combustion, whereas the condensates from the brine revealed values twice as high as the detection limit (o.3 pCi/ml). The latter result is most probably due to a contamination effect resulting from brine injections during routine borehole inspection and maintenance operations.

4.3 Control of the Water Table

In addition to the extensive activity surveillance programme performed by the Nuclear Research Center on a routine basis, the water of the two water table inspection wells Nos. 16 and 2o (Fig. 3) in the immediate vicinity of the injection well Lh 2 will be subjected to routine tritium checking at 3 months interval.

5. SAFETY CONSIDERATIONS

Contamination of the ground water during injection is excluded due to the fact that at least triple barriers are provided towards the water table. Furthermore, the tank truck remains in the leak-tight trough during pumping and leak-proof couplings and joints are used. Any effluent leak would anyhow collect in the sump of the bore cellar from where it would be taken up again by a sump pump. Leaks occuring 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. Due to the fact that on the one hand the liquid in the annulus completely fills the void up till the bore head and that on the other hand the liquid level in the injection tubing will always be lower (actually at -2oo m) as a result of the reduced pressure in the sandstone lense, leaks occuring in the injection tubing in periods of non-operation could only lead to a migration from the annulus into the injection tubing.

As far as the injection horizon is concerned, both geological data and operation characteristics applicable during crude oil production demonstrate the complete isolation from the other oil-bearing strata. 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 94o m deep horizon into the water table situated roughly 7oo 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.

6. STATUS OF THE PROJECT

As already described in section 3.3, underground adaptation has been terminated successfully. Surface adaptation is actually under way. Provided that the official authorization will be granted at the end of this year, startup of the injection experiments is scheduled for early 1973.

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