

Treatment of Corroded Metallic Uranium Fuel- 9213

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ABSTRACT

This paper describes the extensive planning and treatment of corroded metallic uranium fuel performed in Studsvik in 2007. This included conversion of possible pyrophoric uranium hydride to uranium oxide and separation of intact parts of fuel rods from corrosion products.

The first nuclear reactor in Sweden was the R1 reactor in Stockholm, with fuel of natural metallic uranium. After shut-down of the reactor, part of the fuel had been placed in waterproof canisters in an interim storage pool at Studsvik. In 1988, corrosion of the fuel had been discovered in one of the canisters due to water leakage. Exposure of metallic uranium to water causes pyrophoric uranium hydride to be formed, which poses a severe risk of fire or explosion if subjected to oxygen in air. The canister with corroded fuel had then been placed in an outer container to which inert gas with low oxygen concentration was supplied, in order to slowly oxidize hydride.

A project to take care of the corroded fuel was set up in 2006, comprising several preparatory steps: research, equipment design, pre-treatment and safety-case demonstrations. Treatment comprised two stages, pre-treatment at the storage location, and final treatment in a hot-cell. The pre-treatment stage was performed in order to study the reaction inside the container with a controlled oxygen supply and regularly control of atmosphere inside the container. In addition, equipment was developed for controlled movement of the container. The results from the pre-treatment stage were used for the planning and acceptance of the transport to and handling in the hot-cell facility in Studsvik.

The work performed in the hot-cell consisted of opening the container, sequentially cutting the container and canister and separation of intact parts of fuel rods from the corrosion products and cladding, top and bottoms of aluminium. No evidence of uranium hydride was found throughout the operation. The corrosion products was expected to be in form of powder, but showed to be pressed together as a cake.

The project was carried out safely and in accordance with the time schedule. An important lesson learned was the advantage of a thoroughly analyzed pre-treatment work as the basis for the planning and acceptance of the final treatment. Another principle finding was the positive experience of a small, non-hierarchical project organization involving designers and cell operators at an early stage in the planning. This improved the possibilities of achieving a fast, effective and creative project. The project was finalised in the summer of 2007.

INTRODUCTION

The first reactor in Sweden was the R1 reactor located in Stockholm, which was in operation between 1954 and 1970. Fuel was metallic uranium with no enrichment. A batch of spent fuel was in 1966 transported to Studsvik for storage. It was placed in waterproof canisters in a pool of de-mineralised water in a storage building for spent fuel. The remaining fuel from R1 was also taken to Studsvik later but was placed in dry storage. It was found in 1988 leakage from one of the canisters in the pool as gas bubbles, indicating that corrosion occurred in the canisters. Analyses of the gases samples show that it was mainly hydrogen. Since 1988 it has been discussed how this canister should be treated, and in 2007 it was finally done.

Responsible for the R1 fuel is AB SVAFO a company established for taking care of decommissioning of former sites and installations used for basic research and testing when nuclear power was developed.

A project organization was set up in 2006 to prepare the corroded fuel for final storage. The project included personnel from AB SVAFO and Studsvik Nuclear AB, providing expertise in the fields of chemistry, physics, engineering design, radiation protection, hot-cell operation, and authority contacts. Throughout the project, regular project meetings were held where different problems were discussed in an open atmosphere. Special expertise from Nexia Solution, Great Britain on treatment of metallic uranium fuel was consulted on a regular basis, and meetings with authorities were organized. This paper presents the project with special focus on the treatment of the canister containing corroded fuel. The intact fuel was taken care of in another project and is not considered.

THE FUEL RODS

The design of the fuel rods were basically the following: a bottom and top of aluminium, for most of the rods, the top is about half a meter and the bottoms were all the same about 5 cm. Between the top and the bottom was the metallic uranium in a cladding of aluminium.

The canister contained seven fuel rods of totally 161 kg metallic uranium. In table 1 below the characteristics of the seven rods are given

Table 1: Data for the fuel rods in the canister with corroded fuel.

ID Nr	Length total (mm)	Length uranium (mm)	Weight uranium (g)	Burn up (MWd/tU)	Date into the core	Date out of the core
455	2360	1840	23140	229	1954-07-12	1963-08-07
C36	2360	1840	23260	229	1954-07-12	1963-08-06
50	2440	1930	24550	298	1954-07-12	1963-08-08
448	2360	1840	23090	229	1954-07-12	1963-08-06
409	2445	1930	24330	209	1954-07-12	1964-01-09
317	2360	1840	23170	160	1954-07-12	1964-01-07
69	2350	1550	19610	1	1954-07-12	1956-09-21
	Total:		161150			

EARLY TREATMENT 1988-2006

Leakage of gas was observed for the first time in the end of the 70'ies. In 1988 the leaking canister was identified and examinations were initiated. Gas was collected and analyzed and showed to consist of 77 % hydrogen. The canister was lifted out of the pool and several holes were drilled near the bottom of it with the purpose of removing water. However, no water came out, but black slurry could be washed out. Analyses showed that it was uranium dioxide, and no other chemical form could be identified. A radiograph was made which showed severe corrosion of several of the rods. The other canisters in the pool were also X-rayed, but no water or corrosion could be seen.

The canister with the corroded rods was placed in an outer container with inert gas (Ar) and reinsert in the pool. The container was equipped with connection for exchange of gas. Since 1990 a mixture of about 2% of oxygen in helium has been supplied to the container, in order to achieve a slow reaction from uranium. Gas analyses have been made in order to monitor consumption of O₂ and the release of H₂. In the mid of 90's, water in the canister was evacuated using vacuum pumping and freeze dehydration. The total amount of water removed during the years was about 9 kg.

The exchange of gases was most intensive in the beginning, with a more or less daily exchange of gases. Since June 2003, the procedure was applied less frequently, the gas exchange was made once a month until the mid of 2006.

THE PROBLEM

The Swedish concept for final storage of nuclear fuel is a direct geological disposal of oxide fuel, the so called SKB method. The safety case does not take in consideration any other chemical form of the fuel than uranium oxide. Therefore it would be necessary to transform the metallic fuel to oxide. For the remaining intact metallic uranium a project for transform it to oxide form was ongoing. A project for taking care of the majority of the R1 fuel had already started and it was decided to include the metallic fraction from the canister with corroded fuel together with major part. This imposed a tight schedule.

The main problem was that the corrosion products in the canister were expected to contain uranium oxide and uranium hydride, all in the form of powder. Uranium hydride is very reactive with both water and oxygen. If there is a large quantity of hydride present when the canister was opened, oxygen in the air would cause a violent reaction, which could lead to severe problems of contamination, fires and other damages. Even though the container has been treated for about 15 years it was not possible to exclude that uranium hydride still is present. It was therefore necessary to transform any uranium hydride to oxide before opening the canister, or do the opening and sorting within an inert atmosphere.

INITIATING INVESTIGATION

The project to finally take care of the corroded fuel was started in 2006. Until then several studies had been performed to find a suitable solution. Many different options and ideas had been discussed how the container and its content should be taken care of.

The new project also started with discussions of different solutions and final treatment was an open matter. For example one option put forward was to transport it to take the canister as it was to an external facility with suitable equipment. This option was discarded since getting a licence for such a transport was recognized to be very difficult and this option was therefore excluded. Several methods for treatment at Studsvik were discussed in which special equipment was proposed and evaluated. A valuable partner in discussions and evaluation of different options was Nexia Solution, which held an advisory position through out the project.

The purpose of the initial investigations was to as far as possible settle the condition of the uranium before opening the container and canister. And then if uranium hydride still was present in the material matrix transform it to uranium oxide smoothly without any severe contaminations.

The gas exchange was intensified and the methods for analysing the gas mixture was validated and improved, and analysis of water vapour was included. Gas exchange was made by vacuum pumping to ensure that the gas for analyse came from inside the canister. The equipment for gas exchange and sampling was upgraded with new valves and tubes when some leakage in the system was found. Normally the pressure within the container was kept at normal atmospheric pressure.

The concentration of oxygen was kept low to avoid any violent reaction of uranium hydride. Oxygen concentration in the gas had been 2% but was changed to 4% in order to increase the reaction rate but without the risk for violent reactions. The increase was thoroughly evaluated by calculations and judgement of experts with years of experience.

A bulb on the inner canister was seen on a radiograph, and could not be explained from what it was in the canister. It was discussed if this could be due to a violent reaction/explosion of uranium hydride. Later when the canister was opened a reasonable explanation could be given. The bulb is shown on figure 1 as well as the upper part of the fuel elements.

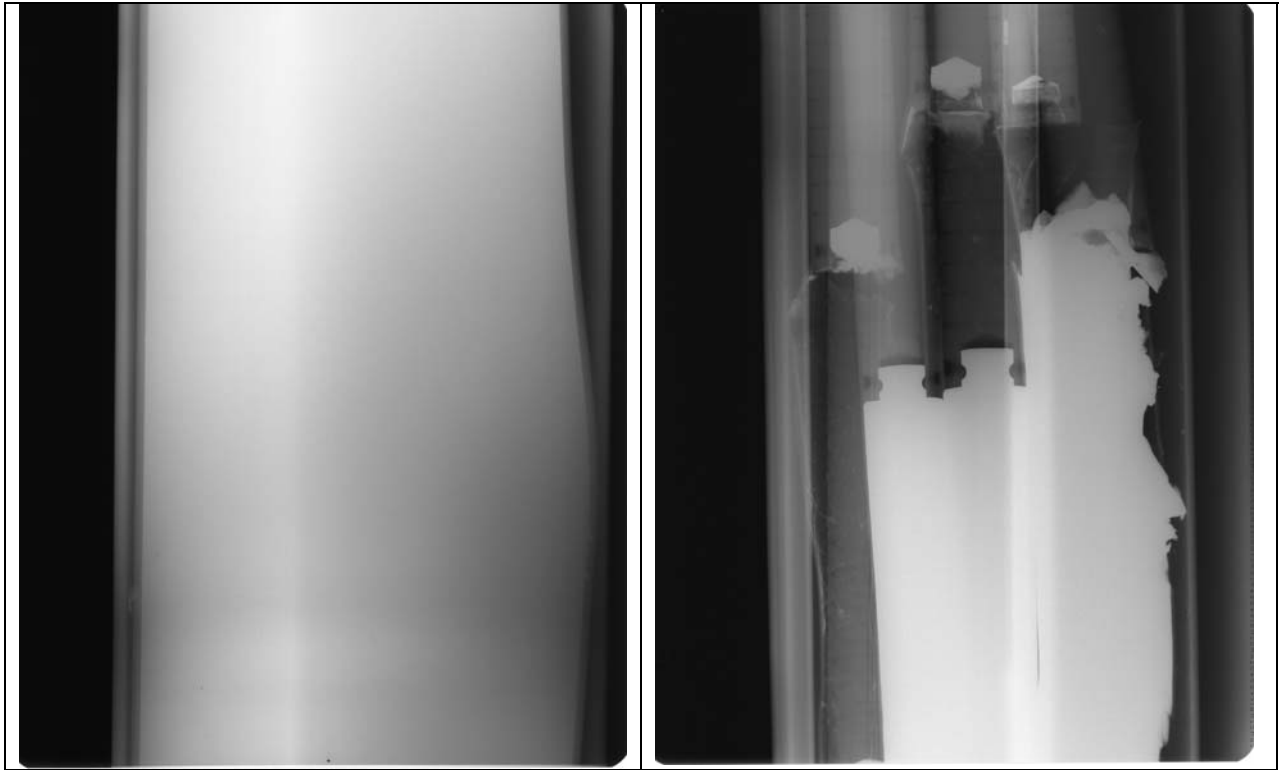


Figure 1 Radiograph of the canister. Left: the bulb on the canister. Right: upper part of fuel rods.

PRE-TREATMENT

The result of the initial investigations could not be excluded that there still was uranium hydride within the canister since the oxygen consumption and hydrogen production still occurred. The purpose of the pre-treatment was to expose and oxidize as much uranium hydride as possible in a slow and controlled manner, thus minimize the risk at transporting and emptying the canister.

The corrosion products in the canister were expected to be in form of powder, either as uranium hydride or uranium oxide or both. There is also a possibility that there are pockets of uranium hydride covered by layers of uranium oxide, preventing oxygen to react with the hydride. These pockets may be disturbed when moving the canister, exposing fresh surfaces to sufficient amounts of water or oxygen. To increase the oxidation rate and make sure that most material would have been exposed to oxygen the idea was to shake the container. The shaking was done by tilting, rotating and finally vibrating the container.

Equipment for tilting and moving the container was constructed by Studsvik Nuclear AB. It made it possible to tilt the container stepwise and rotate it, all controlled remotely. The rotation could only be done 45 degrees to each side, otherwise the connections for gas exchange would have been damaged. When the equipment was in place, a test was done with a dummy container (see Figure 2). The equipment worked exceptionally well after a few small corrections. At the same time as the construction was done a safety analysis was prepared and could be presented to the authorities shortly after the construction was settled.

The tilting procedure started with steps of 15 degrees with rotation at each step. The pressure in the container was monitored so the procedure could be stopped if pressure rise. The entire tilting procedure up to 90 degrees continued without any increase of the pressure in the container.

When the container was horizontal equipment was applied for vibrating it. Then the oxygen content of the supplied gas was increased stepwise, and the container was rotated and vibrated at each step. The final concentration of oxygen was 20% corresponding to normal air concentration.

Nothing exceptional, as pressure increase or changes in the gas composition, was observed throughout the treatment. Additional radiographs were taken, and did not show any changes in the location of the rods in the canister. At that time, the hypothesis was that there was still a lot of moist present in the corrosion products and therefore no movements in canister occur, even though vacuum and freezing procedures had been carried out. When the container was opened later the explanation was given.

The purpose of the pre-treatment was to expose and oxidise as much uranium hydride as possible in a slow and controlled manner, with the objective to minimize the risk at transporting and emptying the canister.



Figure 2 The tilting equipment with the dummy container. The container with corroded fuel is seen at the far side of the pool.

OPENING AND SORTING

As the tilting and vibration did not give any noticeable effect, the container was transported from the storage facility to the hot-cell facility without any special arrangement than normal safety arrangement for internal transports of material of high activity. The only extra safety action was that during the transport the container was filled with inert gas.

It could not be excluded that there were still uranium hydride within the canister, though. There might be pockets of uranium hydride that was protected by layers of uranium oxide. When the canister was opened and its content disturbed, the hydride could be exposed to air and give an intensive reaction.

For practical reasons, it was impossible to have inert gas atmosphere within the hot-cell while opening and handling the canister. Therefore, the procedure for opening the canister and remove the content was analyzed thoroughly. Protective measures were taken, firstly, to avoid any violent reaction, and, secondly, to douse possible fires.

A safety analysis was sent to the authorities for approval. It focused on the possibility of violent reaction of uranium hydride. Special equipment was designed or adaptation of existing tools was made for the hot-cell work. Detailed work instructions were produced, and training sessions for critical moments were carried out with inactive material. Salt bags of different shapes were manufactured, and were used in fire fighting exercises with non-active material with similar chemical reactions as uranium hydride.

The container was too large to fit in the hot cell, and therefore a docking procedure was employed, see Figure 3. The transport flask was considered as an extension of the cell.

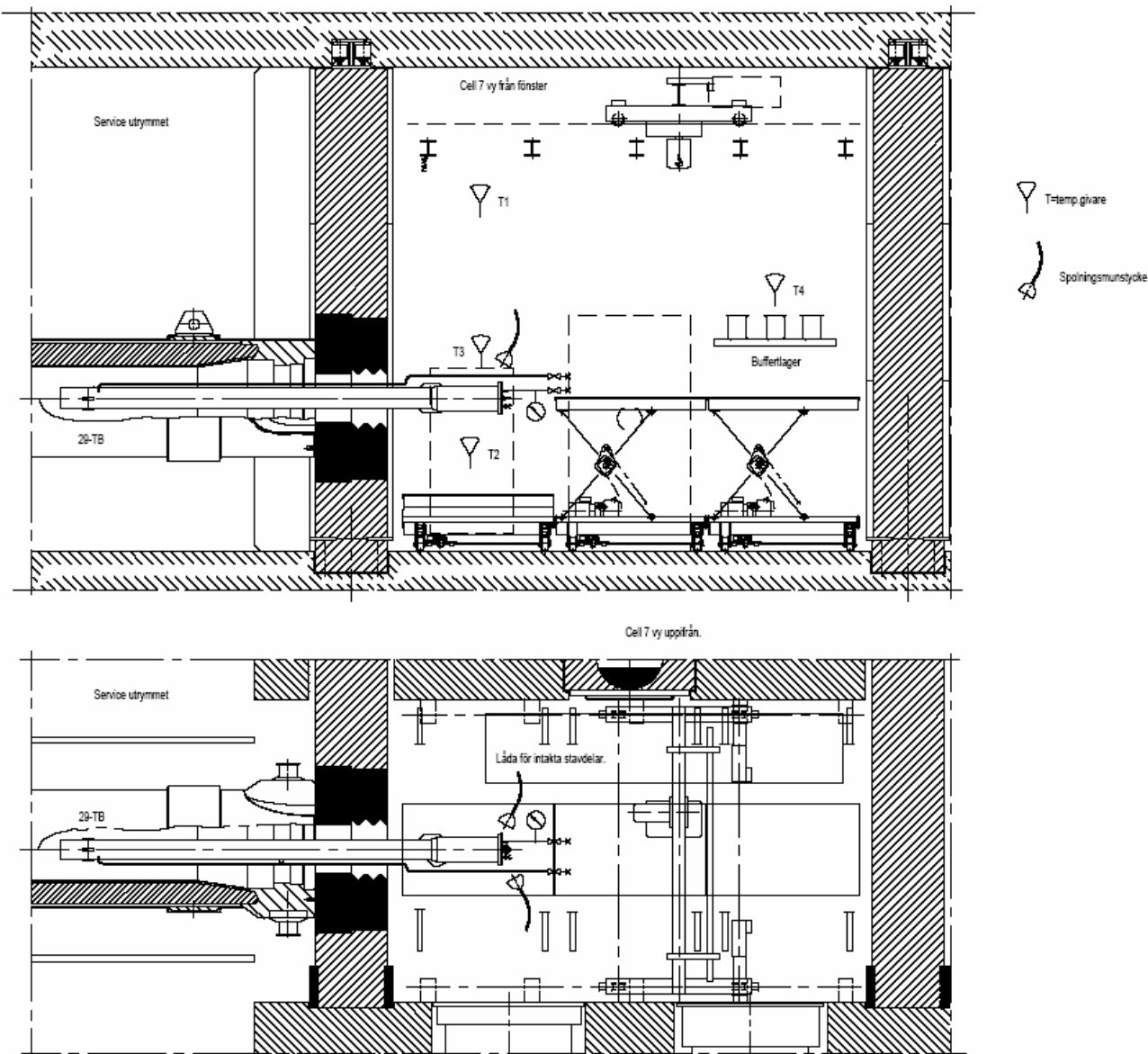


Figure 3: Schematic of the hot-cell and the container. Top: side view, Bottom: top view. The transport flask was docked to the cell and seen partly on the left side

The treatment started with the removal of the lid of the outer container which was done without any problem. As a mitigation action inert gas was flushed through the container via connection in the bottom previously used for the gas exchange. Inert gas was also flushed around the opening of the container through special nozzle. The outer container was cut just behind the top of the inner canister, see Figure 4. The cutting was performed with a commercial circular saw equipped with special designed stand.

The lid of the inner canister was then removed and the top of the fuel rods were exposed for the first time since 1966. The top pieces of four of the rods could be removed with ease, while the other three was stuck. This was surprisingly as it was assumed that the corrosion products should be more or less as powder, and furthermore all rods were expected to be corroded.

Further cutting of the container and canister was made and more of the corrosion products were visible and possible to remove. When the canister was cut back it became obvious that the corrosion products were not at all in the form of a powder but more or less a solid cake but in the shape of rods.

The density of uranium metal is about 19 kg/dm^3 and uranium oxide and hydride about 11 kg/dm^3 . This means that the corrosion products need more space than uranium metal. It was assumed that the space in the canister has been limited for the corrosion products and therefore been compressed. The bulb seen in the X-ray pictures, which was speculated due to some kind of explosion, could get a plausible explanation by the fact that the corrosion products needed more space and give rise to the bulb via its expansion.

The cutting and removing of corrosion products continued but the three intact rods were impossible to move and therefore could not be removed from the solids in the canister. In order to be able to continue to pull the container into the cell those intact rods must be removed. The intact rods were split by cracking. Same method had been used on the intact rods. The three rods were more or less intact to the very bottom of the canister.

During the planning it was assumed that the corroded material should be more or less as powder and therefore was the prepared tools adopted for this. As the corrosion products were not in a powdery form it was therefore necessary to modify the procedure as well as the tools. The creative and prestige free atmosphere in the project contributed to a successful solution of all such technical problems without compromising the safety case.

During the work in the hot-cell it was never any problem with reaction of uranium hydride since, probably, almost all the corrosion products in the canister had already been oxidised to uranium oxide. The container have been treated with more or less frequent exchange of gas with low oxygen content for more than 15 years, the importance of the activities, i.e. shaking, tilting and vibration, during last half year can not be specified but probably not of any major importance. However, since the state of the material in the canister could not be analysed or verified in any other way, it was necessary to perform that shaking procedure to be on the safe side.

Of the total inventory of uranium which was just above 161 kg almost 90 kg of metallic fuel could be recovered, the main part was from the three intact rods. This was converted with the rest of the intact fuel from the R1-reactor into uranium oxide. The remaining part just less than 72 kg of uranium as oxide will be taken care in a separate process for conditioning and packing to fulfil the final storage acceptance criteria for to the Swedish concept.

Even though the corrosion products were handled freely in the hot-cell the contamination of the cell was very low. This was determined by reference areas, where smear test were taken before, during and after the treatment in the cell and these showed very low levels of contamination. Before the work started the risk of contamination of the cell was a major technical risk. This was however based on the assumption that the corrosion products should be a fine powder which easily could spread on the cell but instead was more solid and fell apart in larger pieces.

The work in the cell did not turn out as it was planned, and it should be said that this was beneficial to the project. There was no uranium hydride and therefore no violent reactions which of course was a relief. The fine powder which was expected was instead more like a solid ceramic. The methods and tools had

therefore to be developed to get the material out of the canister and separate the metallic fraction from the oxide fraction. Due to an efficient organisation for decision and which also included the ability to perform the necessary tasks these chances could be taken care of relatively smoothly, which was due to the positive approach among the staff involved in the project.



Figure 4: Left: Top of inner canister when the container is removed. Right: Top of the fuel element in the canister when the aluminum top of the 4 corroded rods is removed

CONCLUSION

The solution of the corroded metallic uranium has been a problem since 1988 when it first was examined. The fear of a violent reaction of uranium hydride has been the main factor considered since then. However, no reaction between hydride and oxygen could be seen when the container was opened. Neither the conditions in the canister were such that it led to production of uranium hydride, nor the long pre-treatment with gas of low oxygen concentration had any effect. Four of the rods showed to have corroded more or less completely while three were more or less intact. The uranium oxide was not a powder but more like a solid cake. The most likely explanation is that there was little free space in the canister, and the corrosion product was squeezed to solid cake when the corrosion process proceeded. This is probably also the explanation to the bulb on the canister. The tilting procedure probably did not have any effect at all.

Emptying the canister was more complicated than expected, and gave rise to other problems than foreseen. The planning was focused on mitigation of possible consequences of reaction between uranium hydride and air, which could have caused fire and contamination within the hot cell. The main problem instead showed to be to remove the material stuck in the canister. The top pieces of the rods could be removed easily, while the intact rods were not possible to pull apart from the corroded material.

The project was carried out safely and in accordance with the time schedule. An important lesson learned was the advantage of a thoroughly analyzed pre-treatment work as the basis for planning and acceptance of the final treatment. The use of mock-ups for training showed to be of great benefit. Another principal finding was the positive experience of a small, non-hierarchic project organization and involving all competences at an early stage in the planning. This improved the possibility of achieving a fast, effective and creative project.

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