FRM II: REPLACEMENT OF THIMBLES MADE FROM ALMG3 (EN AW-5754) FORMING A BARRIER BETWEEN HEAVY AND LIGHT WATER AFTER DETECTION OF CORROSION

A. PICHLMAIER, H. GERSTENBERG, A. KASTENMÜLLER, C. KROKOWSKI, U. LICHNOVSKY, R. SCHÄTZLEIN, M. SCHMIDT

Technische Universität München, Forschungsneutronenquelle Heinz Maier-Leibnitz (FRM II)

Lichtenbergstr. 1, D-85748 Garching, Germany

ABSTRACT

The FRM II is a tank in pool type heavy water moderated multipurpose reactor with 20 MW thermal power. Its 12 beam tubes are mainly used for neutron scattering experiments. However, it also operates a dedicated neutron activation analysis instrument, a tomography facility and a positron source. One beamline is used mainly for medical applications. Furthermore, isotope production and Silicon doping are important activities at the FRM II. The FRM II became critical for the first time on March 2nd, 2004. Since the beginning of routine operation in 2005 it has now completed more than 40 reactor cycles in almost twelve years of service. In 2015/16, an extended maintenance break to accommodate novel scientific needs was held.

In late 2015, during this maintenance period with shut down reactor and low pool-water-level, above background Tritium (^3H) concentrations were discovered in the air of the reactor hall. Thorough investigation revealed that the source of this ^3H could only be the heavy water of the moderator tank. Further tests confirmed the initial assumption that one of the thimbles reaching from the light water of the pool into the heavy water of the moderator had a microleak: heavy water could trickle into the light water at a rate of less than 100 cc/day. We did an extensive analysis of the cause of this leak and finally decided to exchange not only the affected thimble but as a precaution also three identical ones.

In this paper, we report on the analysis of the cause of the leak and present its results. We will also describe the process of replacing the thimbles and the precautions taken to avoid similar impairment in the future.

1. Introduction

The FRM II is a tank in pool reactor with 20 MW thermal power. A single fuel element, containing 113 fuel plates with highly enriched Uranium, is cooled by light water and placed in a moderator tank filled with heavy water. This setup yields an unperturbed thermal equivalent flux of 8×10^{14} n/cm²/s over a cycle of 60 days. Generally, the reactor is run for up to four cycles per year. The FRM II has reached criticality for the first time on Mach 2nd, 2004.

The main purpose of the FRM II is scientific research in beam tube experiments. Nonetheless, it also is used for radioisotope production; it operates a Silicon doping facility and an installation for medical treatment. Details can be found e. g. in [1].

Several thimbles made of the Aluminium alloy AlMg3 (EN AW-5754) reach into the moderator tank from the top. Among these are four identical ones: three contain thermocouples for the determination of the temperature of the heavy water, one houses the start-up sources. The position and approximate size of one of the thimbles is indicated in Fig. *1*. The Fig. 2 shows a drawing to scale of the thimble. Its overall length is 6430 mm, the inner diameter 30 mm and the outer 38 mm. It is made of two pieces: a top tube-like part of 4440 mm length with flange and a bottom thimble-like part (1990 mm), both machined from one piece of solid material, welded together only once and in a position where low neutron flux is expected during reactor operation.

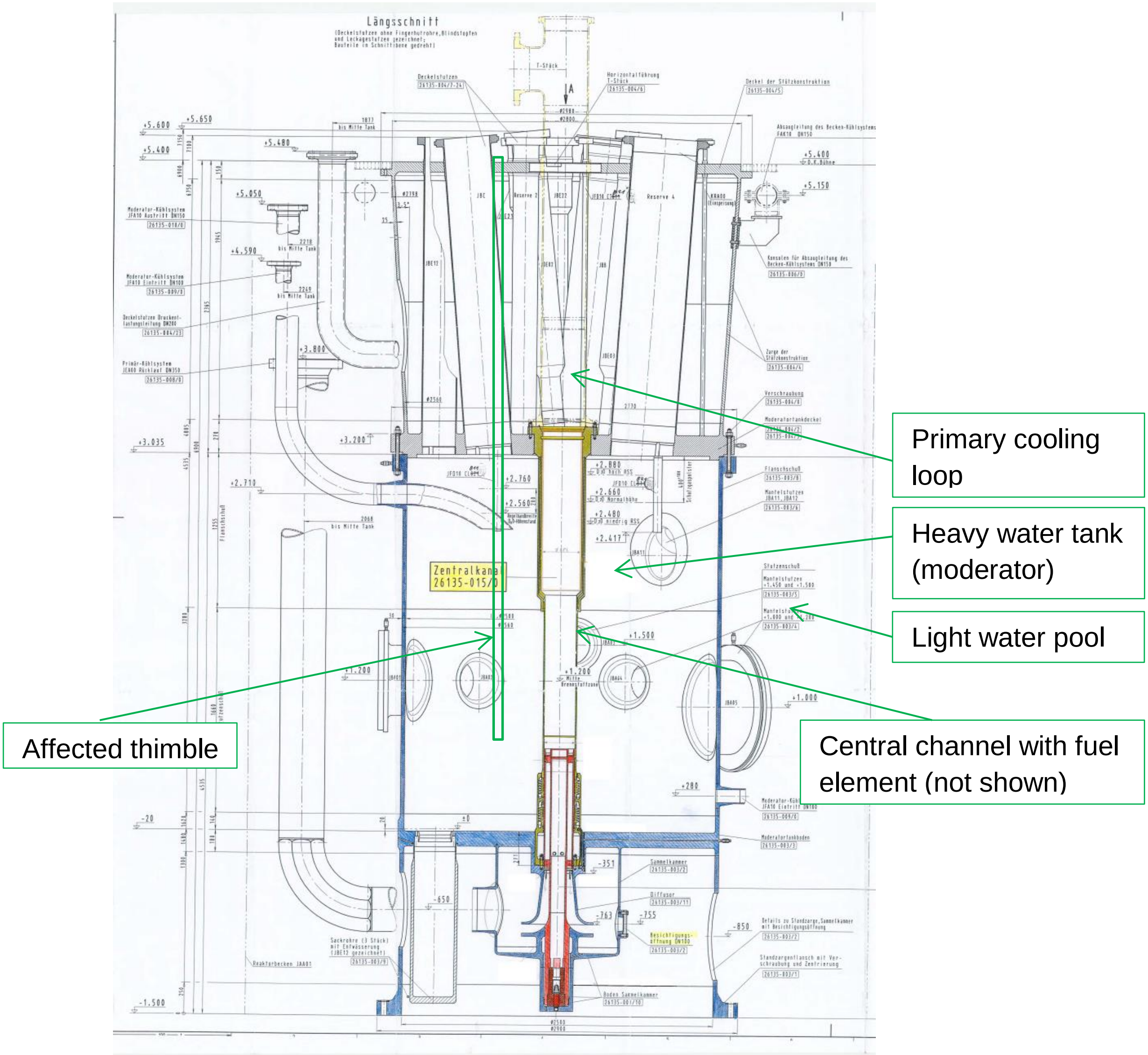

Fig. 1: The FRM II Moderator tank, surrounded by light water. The affected thimble is indicated in its approximate position and scale.

2. Discovery of a leak in one thimble and consequences

During a maintenance break with low level of water in the reactor pool a slight increase of the ^{3}H concentration in the reactor hall, far below any release limits, was detected. Soon it became clear that it had to be related to the heavy water (D_2O), the only significant ^{3}H source available. Since no immediate detailed cause could be determined the water level in the pool was raised which stopped the ^{3}H emission. Further investigation soon revealed a small concentration of ^{3}H also in the pool water. It was estimated from the ^{3}H concentration that a total amount of some hundred cc of D_2O had contaminated the H_2O. Looking at the phenomenon in even more detail a thimble housing a thermocouple was identified as the main ^{3}H source. An endoscopic inspection of the thimble was carried out. Several spots of a few square millimetres in size that resembled ironoxide in colour were discovered, a finding not to be explained straight forwardly in a component made from an Aluminium alloy (Fig. 3).

Final proof of a leak was achieved through an easy but effective test: the thimble was removed from the moderator tank and, still under water, a few mbar of air overpressure were applied. A string of bubbles (Fig. 4) lead to the faulty position which later could be linked to one of the affected spots identified earlier by endoscopic inspection. This test, although in general quite simple, required substantial and time consuming preparations: because the thimble had to be removed from the moderator tank, the heavy water had to be drained and subsequently the tank to be dried in order to avoid any further release of ^{3}H. This operation, although almost routinely done at the FRM II, takes several weeks to accomplish. A picture taken by the endoscope and the bubbles emanating from the tube are shown in Fig. 3 and Fig. 4 respectively.

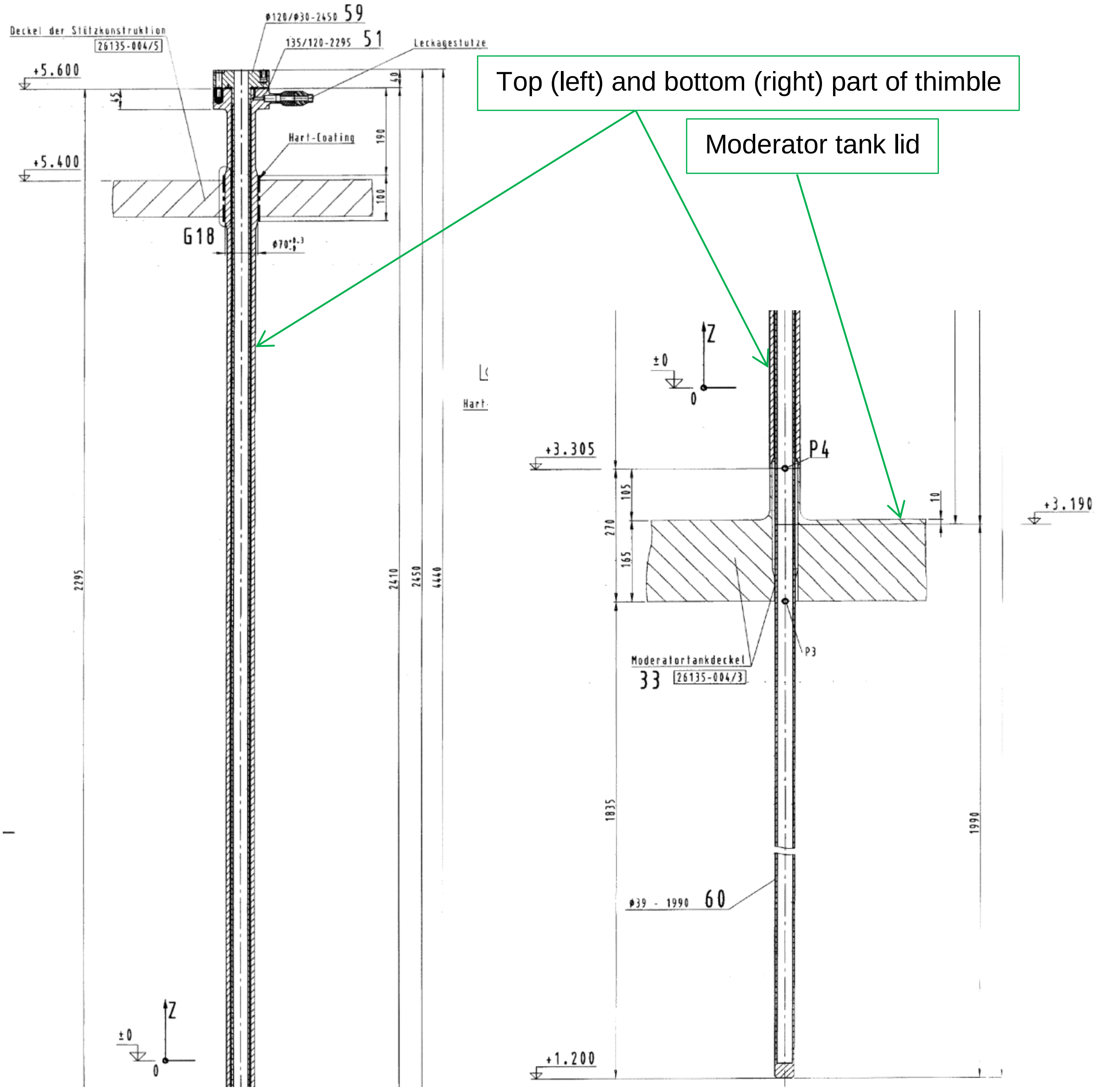

Fig. 2: Drawing of the thimble (left: top, right: bottom)

All four identical thimbles installed at the FRM II were inspected. Even though the three thimbles housing the thermocouples exhibited several of the same conspicuous spots as the one leaking no further leak could be found. In the thimble containing the start-up sources only very few of these spots could be found. As a precaution, it was decided to have new thimbles made to replace all four of the old ones.

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Fig. 3: Affected inner surface of the thimble.

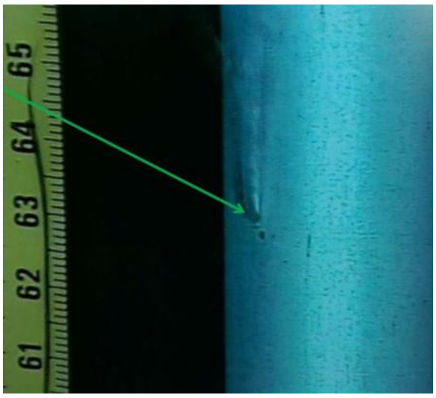

Fig. 4: Air bubbles emanating from the thimble during application of slight overpressure.

3. Detailed Analysis of the Phenomenon and Results

In order to find the reason for the leak a detailed investigation has been carried out. The affected thimble was cut into transportable segments and sent off for further investigation to the AREVA hot cells in Erlangen, Germany [2], and to the ZTWB Radiochemie München RCM, Garching, Germany [3], our neighbour institute at the Technical University of Munich.

Several tests were carried out by AREVA:

To verify that indeed the material AlMg3 (EN AW-5754) had been used for the bulk, stationary spark spectral analysis was performed. The composition of the material was clearly as expected. Also the overall granular structure met the expectations linked to AlMg3. At the affected positions, however, the situation was different: an EDX-analysis revealed clearly visible concentrations of Si, Cl, Cr, V, Mn and Fe (Fig. 5). It turned out that the black spots indeed were small cavernous holes with tungsten detectable on the surface. Outside the affected areas, unusual concentrations of materials foreign to the used alloy AlMg3 could be identified. Details of the analysis are shown in Tab 1.

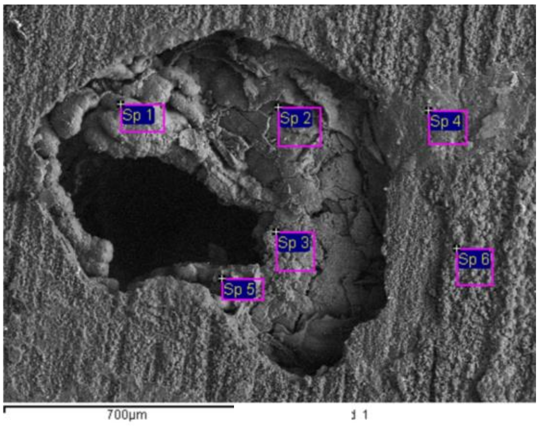

Fig. 5: REM-EDX-Analysis of the outer surface. At the positions Sp 1 … SP 6 the material composition was determined (cf. Tab 1) [2].

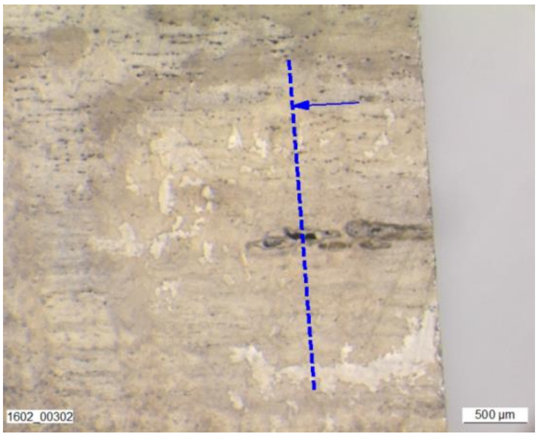

Fig. 6: Local indentations and indication of the position of a cut through the affected position [2].

Metallographic analysis of a cut through the affected sample is shown in Fig. 9 In this figure, the thimble has been cut through one of the prominent black spots that had not yet lead to a leak. The remaining wall thickness is about 2.8 mm.

Spectrum	O	Al	Si	Cl	V	Cr	Mn	Fe	Sum
1	64.1	34.2				0.7	1.0		100.0
2	60.1	37.0			0.5	0.5	1.2	0.7	100.0
3	60.0	37.9			0.6		0.9	0.6	100.0
4	60.4	38.5					1.1		100.0
5	58.3	36.5	1.4		0.7	0.8	2.3		100.0
6	62.4	35.6		0.4	0.4		1.2		100.0

Tab 1 : EDX-analysis of surface contamination, all numbers given in weight-%, cf. Fig. 5 [2].

Additionally, thorough visual inspection revealed a quite large number of statistically distributed black spots. Analysis of the surface showed that they were mainly made up from grainy oxide layer, while small amounts of other material (e. g. W and Cr) could also be detected. This pattern appears to be quite similar to the one detected around the leak position which leads to the assumption that a similar mechanism was at work.

Finally a penetration test (PT) was performed. Other than a spiral-like rough area in the thimble that houses the start-up sources which could be linked to the manufacturing process, very few linear indications and none of any significant size were found.

The Fig. 7 and Fig. 8 show enlarged photos of a cut through and of the surface respectively of the affected position.

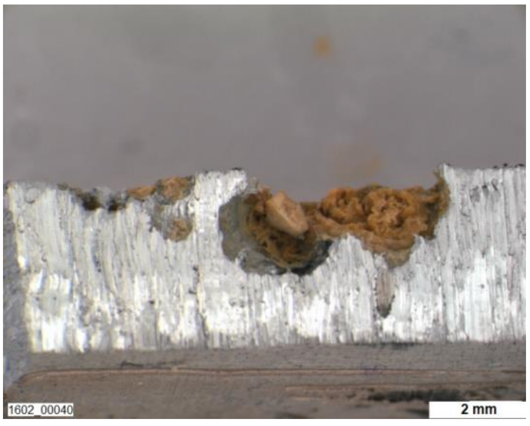

Fig. 7: Enlarged photo of a cut through a typical indication [2].

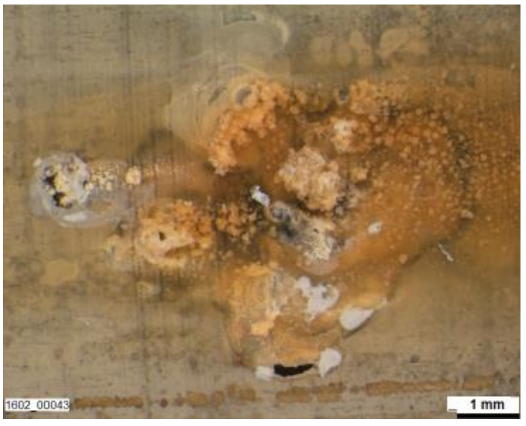

Fig. 8: Photograph of the inner surface in characteristic yellowish color [2].

A complementary analysis of the affected thimble using gamma spectroscopy and REM-EDX has been carried out by RCM [3]. Striking is the quite different texture of the outer surface of the thimble compared to its inside. Pronounced granular structures mainly of Aluminiumoxide were found on the sample surfaces (Fig. 10). These are not typical for AlMg3 and considered as a hint of unusual corrosion. In the thimble containing the startup sources the surface fraction affected by this corrosion appeared to be smaller.

Tungsten could only be detected in traces and only because it had been especially looked for. Other small contaminations detected include particles containing Na, Ca, Mg, Cr, Ni and Ti. These are attributed to the manufacturing process and to the procedure of sample taking.

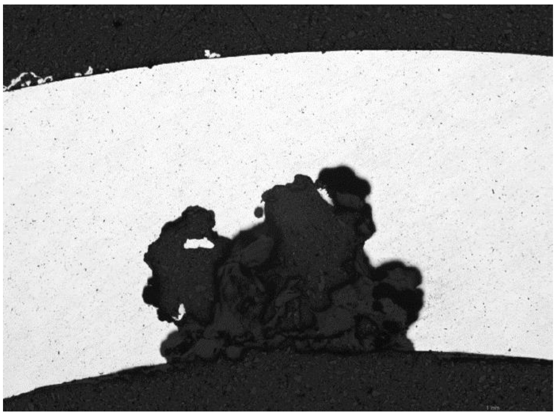

Fig. 9: Metallography, cut through a "black spot" where the corrosion has not yet penetrated through the thimble. [2].

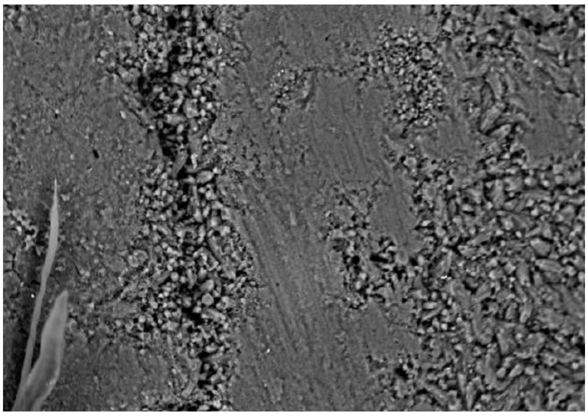

Fig. 10: REM-EDX image of the inner surface of the thimble (mag 928 x) [3].

4. Lessons learned

The alloy EN AW-5754 is generally considered resistant against corrosion and is even used for applications in the marine environment. Nevertheless, it is also well known that Aluminium alloys are generally prone to mechanisms of chloride induced corrosion, such as e. g. pitting corrosion. The initiation of pitting still requires the presence of an oxidant, which can be dissolved oxygen in water or other oxidizing chemical species. The presence of H_2O_2 is thus significantly enhancing the corrosion reaction. Temperature and pH are also important parameters which contribute when they lie in an unfavourable range. In general, the oxide layer on the Aluminium surface is sufficient protection against hole corrosion even in the presence of Chloride.

The thimble housing the startup sources was mainly unaffected by the above described effects. However, all three thimbles housing the thermocouples showed clear indications of corrosion. This corrosion is believed to have been induced by Chloride; it always started at the inner surface which is in contact with H_2O, grew and finally (in one case) penetrated completely through the wall; this opened up a channel into the D_2O-filled moderator tank. Chloride surface contamination in these channels, either grown fully through the wall or still under formation, was found. The peroxide H_2O_2, always present in water under irradiation, in combination with traces of Tungsten has most likely contributed to the initiation of this corrosion.

A combination of the above mentioned conditions may still trigger corrosion; it is concluded that several of these conditions combined were present in the FRM II thimbles. The process of corrosion, once started, was aggravated by the formation of potential differences that "pump" the negatively charged Chloride and Sulphate ions into the holes that start to form; this creates local environmental conditions even more unfavourable thus accelerating further corrosion.

All of the above factors were present.

A whole set of measures has been undertaken to avoid such pitting in the future:

- During the manufacturing of the new thimbles extreme care has been taken not to contaminate the surface with Tungsten or Chloride.
- Under a new procedure the water trapped in the thimbles is regularly exchanged to avoid uncontrollable and thus unknown water conditions.

- The thermocouples have been mounted in a new way so that they are insulated from the surrounding thimble. Additionally they have been equipped with a tube serving as a sacrificial anode should different chemical potentials still lead to electrical currents.
- A new repeated regular test has been introduced: the thimbles are inspected visually. Should suspicious surface structures appear these will be further investigated by eddy current and ultrasonic testing (ET and UT).

5. Conclusion

At the FRM II, a leak from the heavy to the light water system has occurred. Several hundred cm³ of D_2O have contaminated the H_2O (total amount of H_2O about 700 m³). A pinhole in a thimble made of AlMg3 (EN AW-5754) could be identified as the cause. This pinhole had been formed by Chloride induced corrosion. A small initial Chloride contamination had likely induced the corrosion, accelerated by an "ion pump" made up from the thermocouple in the thimble and the surrounding Aluminium in an environment favourable to corrosive effects. As a remedy, new thimbles have been made and installed, the mounting of the thermocouple has been improved and additional scheduled tests have been introduced to monitor the surface conditions of the thimble in order to be able to react quickly to deteriorating conditions.

All in all we are confident that due to the measures taken ideally this kind of corrosion will not happen again in the future; should it reappear regardless we will at least detect it early and take counter measures before a new leak can form.

6. Acknowledgement

The detailed investigations of the thimbles have been carried out by C. Müller, A. Rempel, A. Roth at al. in the AREVA hot cells in Erlangen, Germany. A complementary study was done by F. Kortmann, C. Lierse von Gostomski at al. at the Radiochemie München (RCM) of the Technical University of Munich. We greatly acknowledge the support of AREVA and RCM. The successful analysis of the event could not have been completed without it.

7. References

[1] FRM II description, http://www.frm2.tum.de/die-neutronenquelle/
[2] Reproduced with permission from the source: Technical Center PTCM-G, AREVA GmbH, Erlangen, Germany
[3] Reproduced with permission from the source: Technical University of Munich, ZTWB Radiochemie München RCM, Garching, Germany