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corrosion, corrosion types, operating experience, nuclear power plants

**Abstract**

It is well known that operational conditions in light water reactors strongly influence the corrosion processes. This paper gives an overview which types of corrosion are identified in operating practice based on the evaluation of events which are reported to the authorities in line with the German reporting criteria. It has been found that the main contributor is the stress corrosion cracking. Several examples of different corrosion mechanisms and their consequences are provided for PWR although a high standard of quality of structures, systems and components has been achieved. Recommendations have been given to check the plant specifications concerning the use of auxiliary materials or fluids during maintenance as well as to examine visually the outer surfaces of austenitic piping with regard to residua of adhesive or adhesive tapes within the framework of in-service inspections. However, events in the last two years shows that such problems cannot be totally avoided.

**1. Introduction**

In light-water reactor (LWR) plants corrosion processes are strongly affected by operational measured variables such as environment medium, construction, material and/or mechanical load. The substantial material strains by the operating pressure, the mass flow, the temperature of the cooling water, the special requirements of water chemistry (conductivity) represent a special hazard regarding corrosive material changes in this range. It requires complex testing facilities and measures (recurrent in-service inspections), in order to exclude to a large extent an occurring of disturbances and/or to prevent and/or limit, if necessary, effects of an event (e.g. by loss of coolant). Thus the safety-relevant components and systems are supervised in determined periods by recurrent in-service inspections regarding aging phenomena and aspects of their behaviour. In case of irregularities this leads to repairs or in individual cases to the change of the components concerned. Events which occurred in a plant are reported to other plants, so that necessary precaution measures are performed also in these plants. The evaluation of the result of the event analyses demonstrate the current safety status of the plant. The operational experiences are efficiently recorded by the

application of national and international data bases such as those implemented by the Electric Power Research Institute in the US.

**2. Operational factors influencing corrosion processes**

Important influence factors which can favour corrosion processes at safety-relevant components are the operating conditions existing in LWR plants such as water chemistry, assigned materials, mechanical and thermal loads, neutron irradiation, operational state (full power or outage) and geometrical factors. In particular in the first years of nuclear energy production, corrosion damages in the nuclear power plants led to undesired consequences. However, these undesired consequences could be reduced due to further developments and realizations in the condensate or feed-water treatment as by the implementation of more highly alloyed steel (thermal treatment, material status) as well optimization of manufacturing and construction of endangered components.

## 2.1. Assigned materials

Beside the austenitic CrNi steel which is used primarily world-wide also nickel base alloys are used as construction materials in LWR plants. In particular the nickel base alloy such as Inconel-600 (NiCr15Fe), which have been implemented in pressurized water reactors (PWR) of American, French and Japanese design particularly for steam generator tubes have shown a larger number of cases of damage due to cracking corrosion. Corrosion-supported cracking appeared at steam generator heat tubes in the tube plate area and also at control rod execution connecting pieces of reactor pressure vessel head and - internals. In German LWR, nickel base alloys are used to a substantially smaller extent. E.g. the tubes of steam generators consist of Incoloy-800 which has a less corrodibility. In comparison to Inconel-600 this alloy has a substantially higher chrome content. However, also in Germany for closure head penetrations nickel base alloys are used of the type Inconel-600. But this material has so far shown not any corrodibility.

Due to the fact that more corrosion-resistant materials are selected for the primary circuit of a PWR plant, that the material status has been approved by thermal treatment and reduction of mechanical tensions as well as that adherence to the required quality criteria of cooling agent chemistry (pH value, electrical conductivity, concentration of damaging ions) is achieved, damage cases due to corrosion cracking are limitable regarding the state of the art of science and technology.

## 2.2. Water chemistry

The most important operating medium in the primary circuit of the pressurized water reactor is water. For the minimization of corrosion processes and undesired lining formation on the hot water-affected metallic material surfaces the characteristics of the operating medium are influenced by chemical-technological measures. Aided substances are added to the operating medium, which positively affect the corrosion procedures apart from the technological processes. From the requirements of technical rules such as German utility specific (VGB) guides, ISO standards or TRD sheets (special technical rules for steam boilers) and the American Water Chemistry Guidelines for PWR and BWR (boiling water reactors), the relevance of water chemistry in the power plant operation is evident. The first VGB guides were provided since 1925 and are regularly revised with regard to the current state-of-the-art.

## 2.3. Mechanical loads by temperature influence

Components such as tubes and containers, which contain steam or water with changing temperature, can be destroyed because of the periodic expansion and contraction of the material (thermal periods) and a corrosive medium by corrosion fatigue. The complicated interfaces between material, mechanical loads and medium is shown in *Figure 1*. The load cases, which can provide information about corrosion fatigue, are usually very plant specific. From the behaviour of other identically constructed nuclear installations operating experience for the own plant cannot be concluded in a straight forward manner. These plant-specific experiences have to be determined specifically and evaluated by fatigue monitoring which is performed parallel to operation.

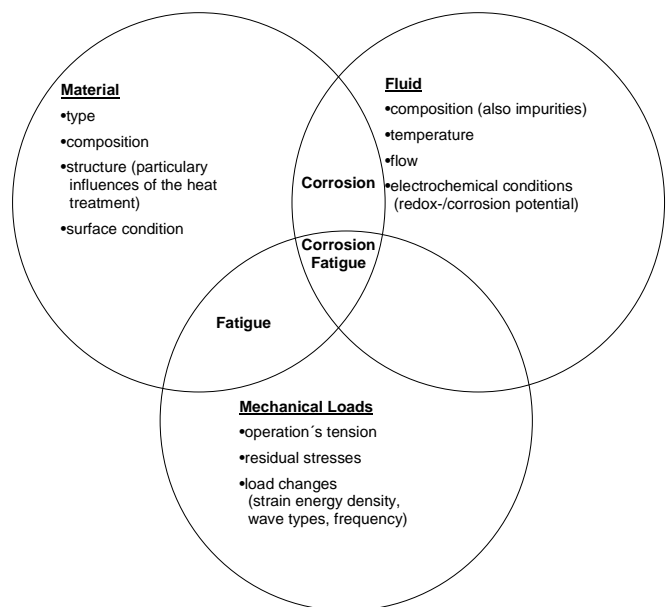


Figure 1. Factors influencing corrosion fatigue

The investigation regarding fatigue is performed plant specific and is done in Germany following „general analysis of the mechanical behaviour" or as “component specific check” described in the relevant German Nuclear Safety Standard KTA 3201.2 [7].

Avoiding of cracking due to corrosion fatigue is increased due to strict application of existing guidelines, monitoring of safety-relevant components by non-destructive testing methods, intensified observation of endangered components and avoidance of notches (increased local expansions and plasticizing), high sulfur content in the material, modifications in the tube diameters, water circulation with high oxygen content and low flow rate.

Mechanical and thermal loads on components can be reduced by careful plant operation.

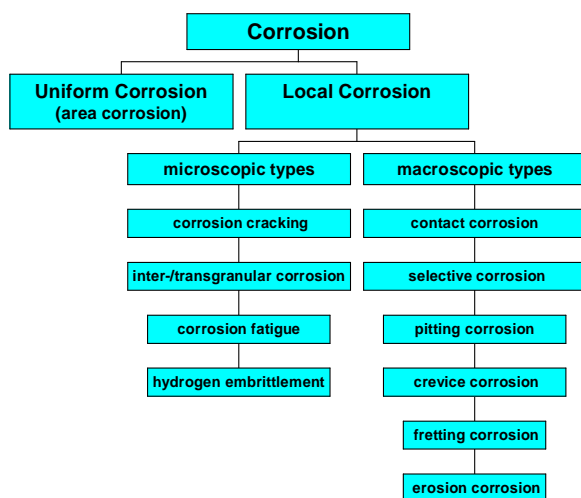
## 2.4. Neutron irradiation

It is to be assumed that with increasing operation times of nuclear installations and the associated rising of neutron fluences the importance of material modifications, caused by radiation, increases. In this case also processes in the material can be effective, which appear in case of very high neutron fluences and may lead to material damages. The corrosion behaviour of metallic materials can be influenced by radiation in two different ways:

- Irradiation-induced modifications of the microstructure (radiation-induced grain boundary segregations and concentration modifications at the grain boundaries e.g. by radiation-induced chrome depletion). Radiation conditioned material modifications regarding radiation-induced stress corrosion can not easily be differentiated from the inter granular stress corrosion.
- Irradiation-induced modification of the water-chemical operating conditions. Also under reducing water-chemical conditions damage cases have been detected (cracking at austenitic screws within the core area of a PWR). These damages happen due to radiolysis and formation of oxygen and  $H_2O_2$ .

## 3. Corrosion types

The differentiation of occurring corrosion types can be done according to the visible picture of the corrosive attack. An outline of the schematic partitioning is shown in *Figure 2*.



*Figure 2.* Outline of corrosion types

In practice, special attention is dedicated to the local corrosion, because this type of corrosion can lead to unexpected damage [4]. A further distinction of the local corrosion into a macroscopic and microscopic type of attack appears appropriate [10]. In contrast to the macroscopic type of attack, practically no "

visible " corrosion product occurs in the microscopic type. These corrosion types are mostly caused by unexpected material failure, without any preceding considerable material losses.

## 4. Inspections of corrosion findings

Recurrent in-service inspections with suitable procedures as well as the execution of operating supervisions (e.g. leakage monitoring, oscillation monitoring etc..) represent the most important measures in order to determine material damages and thus also corrosion damages. As an additional measure for extending the level of knowledge on existing operating-conditioned damage mechanisms, supplementing tests are performed e.g. more detailed non destructive tests as well as destructive investigations at representative places of structures, systems and components, which were removed in the framework of exchange measures.

An overview of applied test kinds, and test techniques contains *Table 1* (cf. [8]). All specified testing methods only respond when separations in the material are present. Crack growth investigations on several years or fracture-mechanics crack growth computations give information over the further course of a crack.

In the following in the testing methods which are usually applied in nuclear power plants are presented briefly:

With the help of visual examinations, which are performed by application of suitable video cameras, leakages and breaks by piping systems are predominantly determined.

The ultrasonic testing (US) represents a frequently applied non-destructive inspection method in the context of the monitoring measures in nuclear installations. It enables the check of errors by the echolot principle. Short ultrasonic impulses with high pulse rate are led into the material, which are reflected at available material defects and represented at the testing set, according to the run time of the ultrasonic impulse.

In the nuclear technology the US check serves, in particular, for identification of the location of crack at surfaces of welding seams. The US testing method enables a good controllability of thick-walled components and of cracks as well as the determination of crack depths. However, practical experiences show that cracks often transmit only weak US signals. Additionally, echoes of cracks in geometrically more complicated places of clutters, as they frequently occur for example at welding seam roots, are often very difficult to differentiate from background echoes.

Table 1. Inspection types, procedures and techniques

Type of Test	Test Procedure	Test Technique
Examination with regard to cracks in the surface or in near-surface regions	Magnetic particle flaw detection	Magnetic particle examination, magnaflux examination
	Liquid penetrant examination	e.g. dye penetrant examination
	Ultrasonic examination procedure	e.g. surface waves, mode conversion, dual search units with longitudinal waves, electromagnetic ultrasonic waves
	Eddy-current examination procedure	Single frequency, multiple frequency
	Radiographic examination procedure	X-ray Radioisotope
Volumetric examination	Selective visual examination	With or without optical means
	Ultrasonic examination procedure	e.g. single probe technique with straight (ES) or angle beam scanning, tandem (angled pitch-catch) technique, mode conversion
	Radiographic examination procedure	X-ray Radioisotope
	Eddy-current examination procedure for thin walls	Single frequency Multiple frequency
Integral examination	Integral visual examination	—
	Pressure test	—
	Functional test	—

The eddy current examination is an important testing method in the nuclear technology that is based on the principle of the electromagnetic induction or eddy currents. In an electrically leading material with a field coil or a measuring probe, by which an alternating current flows, a magnetic field is produced that induces an electrical eddy current. This produces again a further magnetic field, which influenced the exciting magnetic field and thus causes a deviation of the impedance. The modification of the impedance of the receipt coil indicates cracks in the component. However, also local deviations of the electrical conductivity, the magnetic permeability and component geometry can lead to fault signals.

In case of radiographic examination procedure the material errors can be seen as shadows on a radiographic film or a picture-giving electronic system. In the framework of the recurrent in-service inspections, the radiographic examination procedure is not often used, which is justified by the relatively difficult searching of cracks. Inspection of thin-walled, austenitic welding seams show, however, that in case of good instrumentation prerequisites and using improved inspection techniques the finding of cracks leads to a satisfying inspection result.

### 5. Selected corrosion findings at pressure conducting components in PWR-systems

The substantial material strains due to operating pressure, mass flow, and temperature of the cooling water, the special request of water chemistry (conductivity), radiation influences represent a higher hazard regarding corrosive material modifications in the primary circle of a nuclear power plant. This hazard requires complex testing facilities and measures in order to prevent or limit if necessary effects in case of occurring disturbances or

of an incident (e.g. by loss of coolant). Thus the piping systems and pressure conducting components in LWR plants are of special interest in safety engineering. Apart from operating pressure and temperature, to the characteristics of the used materials as well as the sizing and constructional execution of piping systems in LWR plants, also the degree of purity of the cooling water plays an important role regarding corrosion processes.

In Figure 3 the number of reportable events whose causes were attributed to corrosion is presented for the period 1989 to 2001 for all nuclear power plants operated at present in Germany. In this time period 136 events were reported in thirteen PWR plants and 86 events in five BWR plants.

Figure 4 lists the more recent number of reportable events whose causes were attributed to corrosion is presented for the period 2002 to 2008 for all nuclear power plants operated at present in Germany. In this time period 37 events were reported in twelve PWR plants and 41 events in five BWR plants, i.e. one PWR is out of operation in the meantime.

It is to be considered that the oldest PWR was already in operation since 1968 (the first one out of operation) and the youngest PWR since the end of 1988. The oldest BWR is since the year 1976 at the grit, the youngest since 1984.

Regarding the operational aging phenomena a rise of the events would be to be expected with increase of the operational years. However, an easy minimization of the occurrences is to be determined. Evaluation of reportable events allows to introduce preventive measures against certain corrosion phenomena in order to avoid corrosion damage. Due to the operational experience existing safety concepts could be further developed and effectively used in the context of nuclear safety research.

Root cause analyses of corrosion-afflicted safety-relevant components resulted in different corrosion types such as pitting corrosion, surface (uniform) corrosion, stress corrosion, corrosion fatigue, erosion corrosion, strain induced cracking corrosion as well as gap, contact, idle, and cavitation corrosion as well as hydrogen induced corrosion.

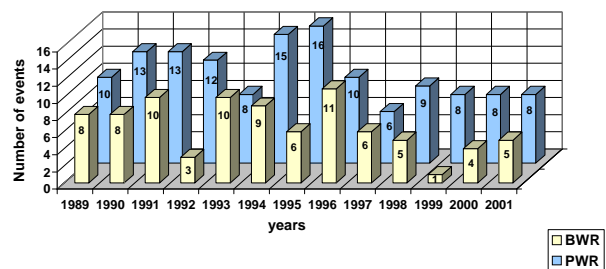


Figure 3. Reportable events regarding corrosion in German nuclear power plants (13 PWR, 5 BWR)

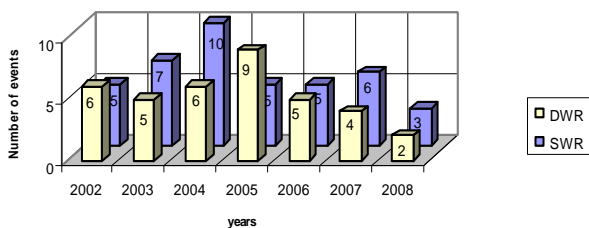


Figure 4. Reportable events regarding corrosion in German nuclear power plants (12 PWR, 5 BWR)

Figure 5 contains the distribution of the different corrosion types for all reportable events of all PWR and five BWR which are in operation in the Federal Republic of Germany in the two time schedules shown in Figure 3 and Figure 4. As a result one can see that stress corrosion was most frequently identified both in PWR and in BWR plants. Pitting corrosion occurs in the PWR plants with 19% whereas corrosion fatigue in the BWR systems occurs with 17% fatigue.

Figure 6 shows an overview of the root causes of different corrosion mechanisms, the developed corrosion types and those preventive measures implemented in nuclear power plants.

In the following some practical examples of different corrosion types are explained, identified on the basis of root cause analyses by the operators of nuclear power plants with PWR.

### 5.1 Stress corrosion

During the spent fuel exchange, a fuel element (three service lives) was inspected which was detected by a Sipping test as a faulty element. The investigation showed reduced frictional forces of fuel rods and a cladding damage by fretting in the area of the first measuring rod. During the endoscopy of the rod meshes one found a broken and a marked out feather/spring. The findings were detected by visual examination and eddy current examination of fuel rods as well as frictional force measurement and endoscopy investigations.

As a root cause, inter granular stress corrosion of the Inconel feathers/springs was determined due to insufficient thermal treatment. The recovery of the damage took place by removing the faulty fuel rod from the fuel element. As a precautionary measure the fuel elements designated for the reapplication were enhanced by using an Inconel rod instead of zircaloy. The fuel elements designated for the new application were already designed using zircaloy.

### 5.2 Pitting and transgranular stress corrosion

During a compression test sample of the leakage detection line of the reactor pressure vessel (RVP) flange gasket at the pipe line section, a leakage occurred within the area of the RVP isolation.

A material investigation of the faulty tube part resulted in trans granular stress corrosion and pitting corrosion as a cause starting from a limited area with chloride deposits inside the tube. Those limited deposit and damage areas were determined by the temperature distribution (reactor pressure vessel outside temperature to ambient temperature) in the line.

For the recovery of the damage, the concerned and a comparable piece of piping was exchanged by two new tube parts with more corrosion-resistant material. A check of the comparable piece of pipe by a surface crack check of the half shells after isolating did not result in any findings.

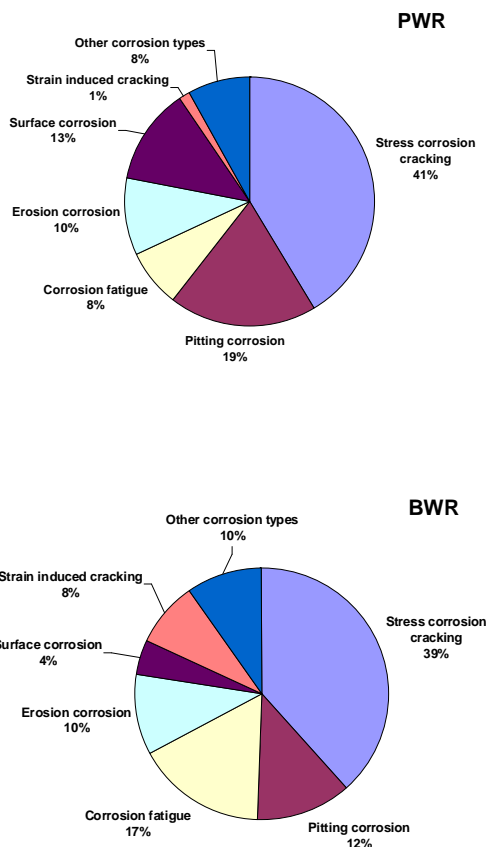


Figure 5. Distribution of corrosion types in PWR and BWR plants in Germany (Evaluation of reportable events 1968 – 2001).

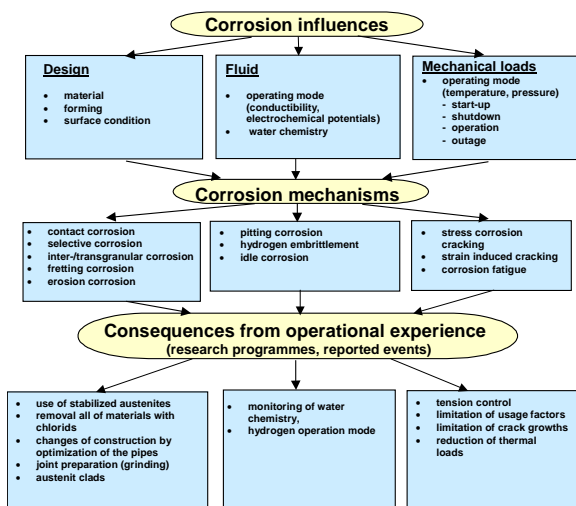


Figure 6. Root causes of different corrosion mechanisms and the operational implementation of protection goals in nuclear power plants

### 5.3 Chloride-induced stress corrosion

In the context of the annual complete overhaul in a plant by a compression test of the leakage exhausting line, tear findings at small lines of the control valves of the pressurizer equipment station were determined. Due to these findings extensive additional examinations of the small lines of the pressure owner armature station took place. Tear findings resulted in the case of two further leakage exhausting lines as well as at the stuffing box return pipe of the pressurizer relief / isolating valve.

All other inspections of the small lines showed no findings. The tear findings did not have influence on the function behaviour of the pressure water relief valves. As a cause, a chloride-induced trans granular stress corrosion starting from the tubing inside was determined. The places corroded were in the transition from the thermally isolated to the not-isolated part of the piping within the areas where steam condenses.

A concentration of minimal chloride quantities which can not be excluded could not be avoided in this area. The damage was recovered by the exchange of the affected piping. To clarify the root cause further investigations of the concerned piping are performed. As a precautionary measure, an improved monitoring of the plugging book and leakage exhausting lines was realized by annual compression tests and non-destructive tests every four years.

Also in case of stabilized austenitic steel stress corrosion can occur under unfavourable conditions. Thus e.g. in earlier years the phenomenon of the "trans granular stress corrosion" was determined in particular within the area of austenitic piping and identified by extensive investigations as "chloride-induced stress corrosion". Due to the fact that the

cooling agent does not contains chloride, no findings in respective through piping were observed. A "chloride-induced stress corrosion" can develop only if chloride containing substances together with water (sometimes only humidity) react due to concentration or deposits.

More general one can say that chloride-induced trans granular stress corrosion cracking (TGSCC) has occurred in German plants on fasteners as well as at inner and outer surfaces of piping all made of stabilised stainless steel due to contact with chloride-containing lubricants, sealing and auxiliary materials or fluids. The fasteners affected were located in the connections of core barrel/core baffle and in the reactor pressure vessel internals.

To prevent future damage, a chloride-free lubricant is to be used and changes are made in the design of the bolts to reduce notch stresses, TGSCC at inner surfaces has mainly been observed in small diameter pipes due to chloride-containing seals, which were replaced with chloride-free ones to prevent future damage. However, in some small pipes in which moistening and drying alternate for technological reasons, chloride concentrations may reach a critical level due to fortification even without any outer chloride source.

In the 90s some crack incidents from outer surface occurred even at piping of larger nominal bore. They have had no direct impact on plant safety. None of the events with leakage, which occurred at operating systems, would have led to an actuation of the safety systems, even in the case of pipe rupture. The availability of the safety systems concerned was given because of their redundancy. However, there was a loss of reliability in operating and safety systems. Recommendations have been given to check the plant specifications concerning the use of auxiliary materials or fluids during maintenance as well as to examine visually the outer surfaces of austenitic piping with regard to residua of adhesive or adhesive tapes within the framework of in-service inspections [9], [11].

In the revision of the year 2007 cracks were found in austenitic armatures of a nuclear power plant with BWR (cf. Figure 7). 23 of 34 analyzed armatures are showing typical intra granular corrosion cracking.

This phenomenon typically occurs in the temperature region between 60°C und 90°C under stagnating medium conditions with a concentration of chlorine ions. Obviously it's most important to avoid chloride concentrations as far as possible in the future.

### 5.4 Flow-accelerated corrosion

Flow-accelerated corrosion (FAC) is a world-wide problem in carbon or low-alloy steel piping of water/steam circuits of power plants. The experience

with FAC on carbon steel piping in plants with PWR is summarised in Figure 8 (see [11]). To avoid FAC, in the 80s the German utilities replaced their condenser tubes made of copper alloys with new ones made of stainless steel or titanium. This replacement action creates suitable conditions for changing the water chemistry to „High“-All Volatile Treatment (HAVT, pH level >9.8).

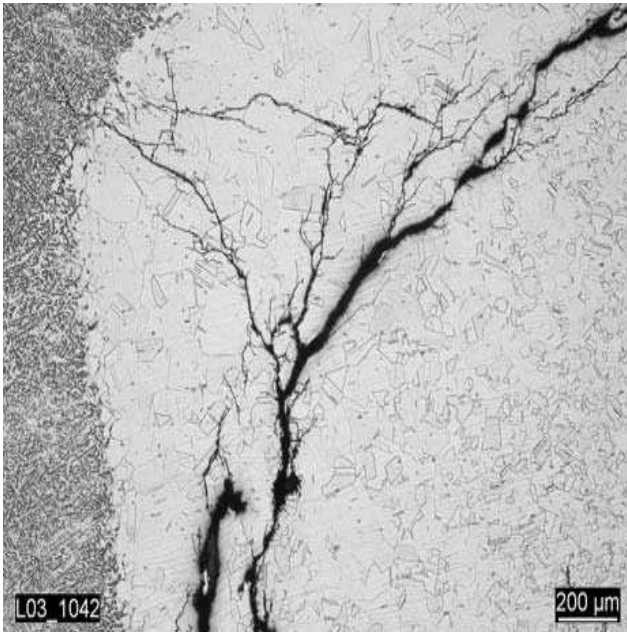


Figure 7. A typical example of a chloride-induced stress corrosion

Furthermore, the implementation of the basic safety concept led to improved flow conditions. In consequence, no damage with safety relevance has occurred in German NPPs due to FAC.

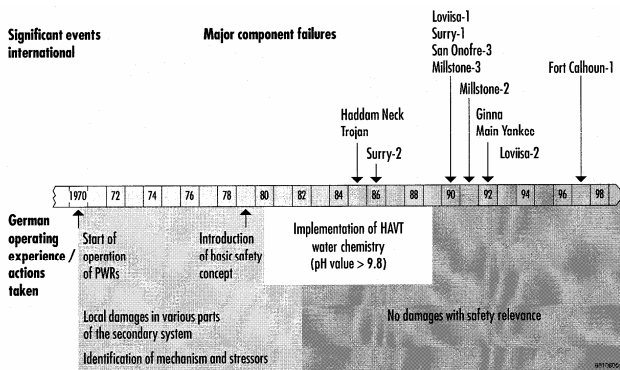


Figure 8. Thinning in PWR carbon steel piping due to flow-accelerated corrosion

Boric acid corrosion of plants with PWRs may cause boric acid corrosion damage to low-alloy or carbon steel base material. Corresponding incidents occurred e.g. in the 80s in some US plants in areas on the reactor vessel head. In Germany, it is good practice

not to operate with primary coolant leakage. In so far, boric acid corrosion is not an issue in Germany.

### 3.5 Idle corrosion

In the reactor cooling system of a PWR plant findings were determined in the area of the ground hand plating of the main cooling agent line. The findings were analyzed in the context of the recurrent in-service inspection (visual examination of the interior surface of the component) in the current revision by means of submarine. In relation to preceding checks the interior surfaces were investigated with an improved video system. First findings were identified in the area of the ground hand plating of the cold TH feeding connecting piece in one loop. After extension of the examination to all loops several displays were determined. The displays were situated all in the area of the ground hand plating. The integrity of the pressure boundaries was not impaired. As a root cause production related local and slag inclusions in ground position transitions of the hand plating were identified. The evaluation is not yet completed; however, operators and experts determined idle corrosion as a root cause. Whereas ferritic and austenitic steel is steady to a corrosion attack during the operation phase due to adjusted water chemistry, an idle corrosion is possible in case of a longer system status at lower temperature and sufficiently loosened oxygen in the medium. The following assumptions are currently under discussion:

1) During operation the corrosion potentials are alike with contacting ferritic and austenitic material areas (e.g. ferritic basic material with austenitic plating). By the use of an anti-corrosive protective layer consisting of magnetite of both materials the same corrosion potentials are assumed. Because the temperature was below the operating temperature sturdier magnetite is converted into the fewer sturdy trivalent ferric oxide/hydroxide. Breaking the ferric oxide/hydroxide layer and the medium enriched with oxygen cause a corrosion attack of the unprotected ferrite.

2) The active corrosion in the ferritic material affected via a locally different concentration of oxygen in the medium with ventilated (cathode) and not ventilated areas (anode) at the metal surface. The difference between the oxygen concentration and an oxygen depletion within the hollow area causes a potential difference and produces a current flow, which leads to the local dissolution of metal.

Results of these discussions are expected up to the end of this year.

## 6. Final considerations

The corrosion protection in the nuclear power plant technology is primarily the result of the operational experience over many years. During the last thirty years the high quality standard was developed by construction, manufacturing and quality assurance. It corresponds to the guidelines of the German Reactor Safety Commission and the relevant safety rules of the Nuclear Safety Standards Committee for German nuclear power plants.

By the application of optimized manufacturing processes and inspection techniques, materials with high-quality properties, in particular the tenacity, conservative limitation of the voltages, minimization of voltage peaks by optimal construction (avoidance of notches, sharp edges etc..) as well as evaluation of occurred failures the important measured influence factors of the corrosion phenomena could be reduced. The design of safety-relevant components is executed in nuclear power material -, manufacturing and test-fairly manner, also concerning the recurrent in-service inspections.

Thus following the state of the art for example in case of the reactor pressure vessel, welding seam constructions are minimized and replaced by smooth forgings. The use of qualitatively perfect product forms as well as a qualified and controlled welding seam manufacturing, by which the growth of stress corrosion is reduced as a consequence of the elimination of chromium carbides at the grain boundaries, is at present standard.

Numerous research work in the field of corrosion in nuclear installations is in close connection with the study of the boundaries, where corrosion cracking can occur. Experiences on the measured variables, which for example, determine the effectiveness of the environment medium, the height of the voltages and the corrosion resistance of the materials, allow to improve the construction and production of component components, but also the operation of nuclear power plants in an optimal manner.

Operational modifications such as aging by increase of the corrodibility are pursued by recurrence in the course of the decades with the help of further developed measuring technique and better estimation of life times. Likewise it is possible by the modern measuring technique to identify findings which could now be made visible but not in earlier years.

There are, nevertheless, still questions which are not yet sufficiently clarified in the area of the corrosion research. For a further extension of the present level of knowledge, investigations would be desirable in particular to the following topics:

- investigations of individual alloying constituents of austenitic materials and nickel based alloys,

by which the radiation-induced stress corrosion can be influenced.

- investigations to radiation-influenced material modifications, radiation-induced stress corrosion and its distinction from inter granular stress corrosion.
- investigations of linings of intergranular cracking particularly in stabilized austenitic CrNi steel. Further autoclave experiments in sulfate and/or chloride-doped water.

One intergranular cracking phenomenon is the irradiation assisted stress corrosion cracking which is investigated experimentally in more detail in [3] providing further insights.

For predictions, a mechanistic understanding of key parameters has to be developed, reliable predictive models have to be formulated based on the mechanistic understanding and cost-effective mitigation technologies for stress corrosion cracking derived and are part of current comprehensive research [1].

In contrast to leaks and breaks of pipelines, quantitative predictions and reliability values for the different failure mechanisms are still not yet available. Therefore, large investigation programmes for different types of nuclear power plants have been performed on international level [5] or are explicitly planned for 2009 and the following years, e. g. by the Electric Power Research Institute [1], [2] within the primary systems corrosion research projects such as improved mechanistic understanding of crack initiation and early crack propagation processes that control stress corrosion cracking, improved predictive models and countermeasures for material corrosion in reactor internals as well as improved prediction and evaluation of environmentally assisted cracking in light water reactor structural materials.

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