Stress Corrosion Cracking Induced by Heavily-rolled Lamellar Microstructure in Non-sensitized Austenitic Stainless Steels in PWRPrimary Water

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学　位　論　文　題　目	Stress Corrosion Cracking Induced by Heavily-rolled Lamellar Microstructure in Non-sensitized Austenitic Stainless Steels in PWR Primary Water (加圧水型原子炉内一次冷却水中における強圧延層状微細組織に起因した非鋭敏化ステンレス鋼の応力腐食割れ現象の解明)
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論文内容要約

Chapter 1: Introduction

After the catastrophic Great East Japan Earthquake on March 11, 2011, operation of all Nuclear Power Plants (NPPs) in Japan has been suspended. Even though no NPP is in operation at present, the basic energy plan decided by the Japan cabinet council has addressed that nuclear power generation should be base load power supplies and NPPs are planned to restart after their safety is confirmed by nuclear regulation authority. Thus, it is indispensable to assure and improve the safety and reliability of NPPs for future practical use of nuclear power generation in Japan. In order to achieve the highly reliable integrity of NPPs, it is very important to eliminate any potential severe accidents and to establish the countermeasures against existing and potential aging degradations of materials used in NPPs.

Throughout the field experiences in NPPs so far, many components made of austenitic materials were suffered from stress corrosion cracking (SCC) in both boiling and pressurised water reactors (BWRs and PWRs), which indicated that SCC is one of the critical mechanisms of aging degradation of the materials.

Most of SCC failures of austenitic stainless steels were initially observed in sensitized stainless steels in BWRs. While L grade stainless steels (low carbon content) were developed and applied to the components in BWRs as metallurgical countermeasures to suppress the sensitization of the materials, SCC failures associated with cold work were subsequently recurred even in the non-sensitized austenitic stainless steels.

On the contrary, it has been generally accepted, for a long time, that neither sensitized nor non-sensitized austenitic stainless steels were susceptible to SCC in PWR primary water because of its deaerated water chemistry. However, it has been recently reported that a failure caused by SCC associated with cold work also appeared in the component exposed to strictly controlled PWR primary water. Therefore, this type of SCC has become the common concern not only for BWRs but also for PWRs.

The main issue investigated in this study is a consequential intergranular stress corrosion cracking (IGSCC) associated with cold-work-induced damage of the materials which is generated during manufacturing processes of the components in NPPs. Since there was great difficulty in reproducing IGSCC by SCC tests using plane-type specimens made of non-sensitized austenitic stainless steels in simulated PWR primary water, crack propagation tests using compact tension (CT) specimens with a fatigue pre-crack have been mainly conducted in PWR environments.

With regard to the crack propagation behaviour of the cold-rolled austenitic stainless steels, two specific phenomena have been notably reported: The first one was that crack growth rates (CGRs) changed drastically depending on the specimen's orientation relative to the rolling direction/plane. The second one was that the off-plane cracking was occasionally observed out of the fatigue pre-crack plane in the specimens with the limited orientations. Since the amplitude of the applied stress on the plane of off-plane cracking in CT specimens under mode I loading was much lower than that on the fatigue pre-crack plane, it is difficult to explain the off-plane cracking by conventional fracture mechanics. It is considered that the off-plane cracking behaviour implies that some unique metallurgical damages due to cold work caused IGSCC phenomenon of non-sensitized austenitic stainless steels in PWR primary water. The reason why these phenomena appeared and their influence on the structural integrity of actual components in PWRs have been still unclear, because these phenomena have not been received but scant attention and studied systematically, so far.

Therefore, it is important to clarify the mechanism of these phenomena associated with cold work and the dominant metallurgical and mechanical factors for assuring and improving the safety and reliability of actual components in PWRs from engineering point of view.

Focusing on the off-plane IGSCC, it was considered that the anisotropic metallurgical damages caused by cold work increased the IGSCC susceptibility of non-sensitized austenitic stainless steels. On the basis of this hypothesis, experimental studies were systematically conducted to validate the hypothesis and clarify the mechanism of these phenomena in this study.

Chapter 2: Effect of Experimental Variables on IGSCC of Non-sensitized Austenitic Stainless Steels in Oxygenated and Hydrogenated High Temperature Water

To reproduce off-plane IGSCC out of a fatigue pre-crack plane and to investigate the effects of materials, stress and environment factors on its susceptibility of non-sensitized austenitic stainless steels, constant displacement SCC tests were conducted using T-L oriented Double Cantilever Beam (DCB) specimens in oxygenated and hydrogenated high temperature water. Total numbers of 85 specimens were examined under the various conditions with different experimental variables considering the cold-rolling reduction ratios of the test materials, stress intensity factors and water chemistries.

Two types of SCC were observed in the specimens. The first one was called Type I IGSCC as main cracking observed on the fatigue pre-crack plane, and the second one was called Type II IGSCC as one of the off-plane cracks observed out of the fatigue pre-crack plane. The relationship between Type II IGSCC susceptibility and the combination of test parameters such as cold-rolling reduction ratios, stress intensity factors and water chemistry were clarified.

The main features of Type II IGSCC are as follows: Type II IGSCC appeared only in the specimens made of heavily cold-rolled stainless steels; Type II IGSCC was reproduced on the S plane in T-L oriented DCB specimen under mode I loading; the mechanical driving force for IGSCC initiation and propagation on the S plane was significantly lower than that on the T plane that was the preferable crack propagation plane in T-L oriented specimen under mode I loading of an isotropic materials.

It is deduced that Type II IGSCC on the S plane is strongly associated with metallurgical factors due to cold rolling and the S plane parallel to the rolling plane is the most susceptible to IGSCC in non-sensitized, heavily cold-rolled, austenitic stainless steels.

Chapter 3: Characterisation of Type II IGSCC Initiation and Propagation Behaviour of Non-sensitized Austenitic Stainless Steels in Simulated PWR Primary Water

To characterise the features and conditions of the initiation and propagation of Type II IGSCC, constant load SCC tests were conducted using T-L oriented CT specimens designated in ASTM E399 in simulated PWR primary water. Crack propagation tests using CT specimens have been widely conducted in hydrogenated high temperature water and/or simulated PWR primary water in the world, and several researchers reported that a specific type of off-plane cracking, called finger and/or secondary cracking, resembling Type II IGSCC in this study was occasionally observed on the plane perpendicular to the fatigue pre-crack plane in T-L oriented CT specimens. The experimental results and test conditions in this chapter are quantitatively compared with these literatures.

Type II IGSCC was also reproduced on the S plane in T-L oriented CT specimens made of heavily cold-rolled stainless steels in simulated PWR primary water, as well as constant displacement SCC tests using DCB specimens. With the increase of the cold-rolling reduction ratio, both the numbers and the depths of Type II IGSCC clearly increased. This result supported that Type II IGSCC was strongly associated with metallurgical factors due to cold rolling, and the S plane parallel to the rolling plane seemed to be the most susceptible to IGSCC in non-sensitized, heavily cold-rolled austenitic stainless steels. This deduction was also supported by the experimental results that S-L oriented CT specimens where the fatigue pre-crack plane was equivalent to the S plane showed CGRs higher than those obtained from other oriented specimens.

Based on the data obtained from CT specimens in this study and literatures reporting the finger and/or secondary cracking, the initiation and propagation behaviour of Type II IGSCC were summarised as follows: Type II IGSCC initiated at the cold-rolling reduction ratios larger than approximately 15% to 20%; in addition, the existence of main cracking, i.e. Type I IGSCC or transgranular stress corrosion cracking (TGSCC), on the T plane was one of the indispensable conditions for the appearance of Type II IGSCC; Type II IGSCC didn’t seem to progress significantly.

Chapter 4: Mechanistic Analysis of Type II IGSCC in Non-sensitized Austenitic Stainless Steels in Simulated PWR Primary Water

To clarify the mechanism of Type II IGSCC in PWR primary water, three experiments were conducted from metallurgical and mechanical points of view, in this chapter.

First, electron backscatter diffraction (EBSD) analyses revealed that heavy cold rolling induced significant variations in metallurgical properties such as the size and shape of grains, crystallographic orientation, distribution of dislocations, and so on. In particular, the distribution of the localised dislocations varied drastically depending on the orientation of planes between the S and T planes in the cold-rolled materials. On the S plane, the areas of the highly localised dislocations remained around grain boundaries and formed into the lamellar microstructures, parallel to the rolling plane.

Second, transmission electron microscopy, energy dispersive X-ray spectroscopy, electron diffraction, and EBSD analyses revealed the metallurgical characteristics around the crack tips of Type II IGSCC reproduced in CT specimens. The grain boundary oxidation was clearly detected not only in the crack of Type II IGSCC but also in its leading area beyond the crack tip. This result indicated the existence of leading edges that were still un-cracked but were already oxidised beyond the physically opened crack tip. The crack travelled in the area with the highly localised dislocations/strains.

Third, the accelerated SCC test in boiling magnesium chloride solution showed that the residual stress due to cold rolling should have played a mechanical role on the appearance of Type II IGSCC. Several cracks of TGSCC were observed on the side planes of the specimen, although no external stress was applied on the side planes. These cracks seemed to initiate and propagate selectively along L direction on the S plane, parallel to the rolling plane, as Type II IGSCC. The appearance of TGSCC suggested that the residual stresses existed in the cold-rolled stainless steel. In addition, it is considered that the volume expansion of the newly grown oxide should have caused the wedge effect of creating/increasing the tensile stress at the crack tip and enhanced the appearance of Type II IGSCC because the crack tip of Type II IGSCC including its leading edge was oxidised.

Based on the above-cited metallurgical and mechanical characteristics of cold-rolled stainless steels and those of the crack tip of Type II IGSCC, the "lamellar microstructure-induced oxidation model" was proposed, which can understand and explain how Type II IGSCC initiates and propagates in non-sensitized austenitic stainless steels in simulated PWR primary water. The main concept of this model is as follows:

(i) One-directional cold rolling induces heavily damaged lamellar microstructure near grain boundaries on the S plane.

(ii) The diffusivity of oxygen at grain boundaries near lamellar microstructure is enhanced due to the increased number of defects caused by cold rolling although grain boundaries originally have the diffusivity higher than that in intragranular.

(iii) The grain boundaries near lamellar microstructure are selectively oxidised due to the high diffusivity of oxygen along the grain boundaries in the S plane.

(iv) Because of the combination of the residual stresses on the S plane and the wedge effect caused by the volume expansion of the newly grown oxide, the selectively oxidised grain boundaries can be cracked easily. After the oxidation of grain boundaries, the strength of oxidised grain boundaries is deteriorated due to the embrittlement such as the concentration of voids at the metal/oxide interface.

The proposed mechanism is applicable to the clarification of the reasons for the appearance of off-plane cracking and the CGR dependence on the CT specimen orientations, in other words, the effect of cold work damage on IGSCC in non-sensitized austenitic stainless steels. These specific phenomena of cold-rolled stainless steels can be correctively understood and explained taking into account the combination of the configuration of the heavily elongated grains and anisotropic IGSCC susceptibility in cold-rolled materials.

Considering the features of the initiation and propagation behaviours of Type II IGSCC, it is possible to prevent the appearance of Type II IGSCC in actual components in PWRs by the following managements: It is important to minimise the amplitude of the localised strain because Type II IGSCC initiates and propagates in heavily cold-rolled stainless steels with the reduction ratios larger than approximately 15% to 20%. In addition, it is also important to prevent the appearance of Type I crack by controlling crack tip mechanics such as the amplitude of the applied stress during operation because Type I IGSCC or TGSCC is one of the indispensable conditions for Type II IGSCC.

Finally, it was revealed that there is low risk of the appearance of Type II IGSCC in the core barrels, which is one of the important components, exposed in PWR primary water environment and is made of non-sensitized austenitic stainless steels, even though some hardened layers are generated due to cold work during manufacturing process.

Chapter 5: Conclusion

In conclusion, it is deduced that Type II IGSCC appears due to the selective oxidation along grain boundaries with the heavily damaged lamellar microstructure, in particular, on the S plane parallel to the rolling plane in cold-rolled stainless steels. Therefore, the existence of the lamellar microstructure plays a key role on IGSCC in non-sensitized austenitic stainless steels in simulated PWR primary water. Throughout this study, un-notable phenomenon of off-plane cracking including Type II IGSCC in non-sensitized, cold-rolled austenitic stainless steels was clarified. In addition, it was revealed that Type II IGSCC isn’t critical concern about the structural integrity of actual components in PWRs. With regard to Type I IGSCC of non-sensitized austenitic stainless steels, proactive countermeasures already applied to BWRs are commonly effective for the components in PWRs. Therefore, this study contributes to the assurance and improvement of the reliability and structural integrity of actual components in PWRs against both Type I and II IGSCC of non-sensitized austenitic stainless steels in PWR primary water.