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論文内容要旨

For safety operation of nuclear power plants, structural material of components has to have high reliability. Stress corrosion cracking (SCC) is very important to consider the plant aging, and in this thesis, SCC in simulated BWR environment that is high temperature water and high purity water was studied. The transition conditions to steady propagation are important to prevent SCC, and then the conditions in various structural materials were discussed.

In chapter 2, SCC initiation behavior in non-sensitized stainless steels was discussed based on the observation of creviced bent beam (CBB) specimens. It is well known that cold work prior to SCC test accelerates SCC initiation, but the mechanism was unclear. It was made clear that cold work promotes trans-granular SCC (TGSCC) initiation and coalescence of infrequent inter-granular SCC (IGSCC) by TGSCC in this study. Crack depth in the non-sensitized stainless steels was fitted well above 50 μm in the normal distribution plot. It indicates that 50 μm of crack depth is criterion of transition to steady propagation. All the crack above 50 μm is recognized IGSCC by optical micrograph (OM) and electron back scatter diffraction (EBSD) observation, and then it was thought that the IGSCC susceptibility is needed to steady propagation. From the view point of slip/dissolution model, IGSCC susceptibility is related to local plastic strain rate when the micro-crack tip encounters the first grain boundary or triple junction. Evaluation of localized plastic strain was performed by Kernel average misorientation (KAM) in EBSD analysis using interrupted tensile test specimens. KAM value is a function of plastic strain, grain size and measured pitch in EBSD measurement. Type 316NG with deeper cracks by CBB test shows higher KAM values at triple junctions, and type 304NG with only micro-cracks shows lower. Therefore, high local plastic strain rate was expected when micro-cracks reaches the first grain boundary or triple junction in type 316NG stainless steels.

In chapter 3, the effect of cold work on SCC initiation behavior for non-sensitized nickel base alloy (alloy 82) was studied. Cold rolled and subsequently ground CBB specimens were prepared to investigate the SCC resistance. Because of surface hardened layer, crack depth doesn't show the linearity above 50 μm . This result is different from the results of non-sensitized

stainless steels. Compared the total number of cracks on cross section surfaces between nickel based weld metal and non-sensitized stainless steels, the number of cracks in nickel based weld metal is much larger than the number in non-sensitized stainless steels. It suggests that the SCC initiation behavior is different from each other because the specimens of nickel base weld metal have hardened layer by grinder. Micro hardness distribution shows approximately 200 μm of hardened layer in case of material ground, and 200 μm is defined as criterion of the transition conditions. Higher rolling reduction, alloy82 has larger number of cracks above 200 μm . Location parameter (that is representative value) in Gumbel distribution for Maximum crack depth exceeds 200 μm when materials were given more than 15% rolling reduction. Therefore it was found that 15% of rolling reduction is transition condition to steady propagation for alloy82. Alloy52 which has higher chromium content and might be alternative to alloy82 shows excellent SCC resistance, although they had been given 15% or 30% cold rolling and subsequent ground surface treatment prior to CBB test. It is reported that oxidization rate coefficient, k_p decreases with chromium content increasing. It is also reported that creep rate decreases with chromium content. Lower k_p and lower creep rate decreases crack propagation rate, and then alloy52 with high chromium shows excellent resistance to SCC. It suggests that the influence of cold rolling and grinder on oxidization rate is not significant for alloy52 in high oxygenated environment.

In chapter 4, the criteria of propagation into low alloy steel (LAS) was investigated on the assumption that stress corrosion crack initiates and propagates in nickel base alloy (alloy182) and reaches the interface between the nickel base alloy and LAS. Modified large scale CBB test method was developed, and cracks on surface distribute uniformly by proper crevice thickness and load at set up. The criterion of SCC propagation into LAS side was investigated using the CBB specimen with the interface in thickness direction. When crack tip reaches the interface between alloy182 and LAS, spherical oxidation and crack in LAS side were observed after modified large scale CBB test. Interfacial cracking was also observed. To quantify the mechanical criterion to SCC propagation into LAS, Elasto-plastic parameter, K_J which is derived from J value, was calculated for each crack by FEM analysis. Sustained crack propagation in LAS side was observed when K_J was more than 75 $\text{MPa}\sqrt{\text{m}}$ in Na_2SO_4 injected environment. The criterion of SCC propagation into LAS was interpreted by competition between crack propagation rate and oxidization rate. From the results of all depo materials, SCC incubation time and propagation rate in alloy182 were estimated. Oxidization rate was evaluated from the estimation and the results of observation on cross section surfaces of modified large scale CBB specimen. Maximum oxidization rate was 3.5×10^{-11} m/s. K dependency of crack propagation rate of LAS itself were reported by many researchers. Crack propagation rate in Na_2SO_4 injected high temperature water is constant below 80 $\text{MPa}\sqrt{\text{m}}$, and it increases significantly above 80 $\text{MPa}\sqrt{\text{m}}$. It is also reported that K_J is almost same as K because of the low work hardening coefficient in case of LAS. Therefore, it was concluded that the conditions where crack propagation rate overcomes oxidization rate is 80 MPa of K_J . This criterion is coincident with our data.

Criteria of the transition to steady crack propagation were described for non-sensitized stainless steels, non-sensitized nickel

base alloys and LAS. Throughout this study, SCC propagation is interpreted by the competition between crack propagation rate and oxidization rate. In chapter 2, it was suggested that local plastic strain rate might increase IGSCC crack propagation rate in non-sensitized stainless steels. Cold work is thought to increase the crack propagation rate for micro cracks. For it helps TGSCC initiation and infrequent IGSCC coalescence by TGSCC and K value in depth direction increases due to low aspect ratio. In chapter 3, crack propagation rate increases by yield strength effect. Yield strength increases by cold rolling. Therefore, SCC in alloy82 given more than 15% cold rolling has high crack propagation rate when crack propagates over surface hardened layer. Below 15% cold rolling, micro cracks would be re-passivated because crack propagation rate could not overcome the re-passivation rate. Alloy52 shows excellent SCC resistance. The excellent resistance is interpreted by high re-passivation rate, and even micro crack didn't initiate. In chapter 4, high K_I increases crack propagation rate in LAS side. When K_I is more than $75\text{MP}\sqrt{\text{m}}$, crack propagation rate could overcome oxidization rate in LAS. Na_2SO_4 injection slightly increases crack propagation rate.

論文審査結果の要旨

原子炉発電設備の運営においては、構造機器の高い信頼性が不可欠である。材料経年劣化事象の一つとして応力腐食割れ (SCC) があり、SCC のリスクを回避する上では、持続的に進展するき裂へと遷移する条件を把握することが重要である。本論文では、沸騰水型原子炉発電設備 (BWR) 構成材料の SCC における進展性き裂への遷移条件について論じており、全 5 章より成る。BWR 構成材料としては、非鋭敏化ステンレス鋼、非鋭敏化 Ni 基合金、低合金鋼 (LAS) を取り扱っている。

第 1 章は序論であり、本研究の背景ならびに以降の章における進展性き裂の定義を述べている。

第 2 章は非鋭敏化ステンレス鋼における SCC について論じている。まず 2 章の前半では、非鋭敏化ステンレス鋼の SCC における塑性予ひずみの重要性について指摘しており、その役割は、粒内型 SCC (TGSCC) の発生促進とそれによる粒界型 SCC (IGSCC) の合体助長であることを初めて明らかにした。また、2 章の後半では、微小き裂が進展性き裂に遷移する条件として、粒界 3 重点での局所塑性ひずみ速度が重要であることを初めて指摘した。鋼種間の局所塑性ひずみ分布の違いを後方散乱電子回折法 (EBSD) による方位差測定結果から定量的に表現する手法を提案し、粒界 3 重点近傍の測定点にて抽出した Kernel Average Misorientation (KAM) 値と進展性き裂への遷移の傾向に相関性があることを見いだしている。

第 3 章は非鋭敏化 Ni 基合金溶接金属の SCC について論じている。ここでは、プラントに適用されている Alloy82 の SCC における塑性予ひずみの影響を調査している。種々の圧下率にて冷間圧延を加え、さらにグラインダ加工を施した試料に対してすきま付定ひずみ曲げ (CBB) 試験を行った結果、表層硬化層を越えて持続的に進展するには、15%以上の塑性予ひずみが必要であると結論付けており、工学的に有用な知見を得ている。また、Alloy82 の対策材として Alloy52 を提案し、塑性予ひずみを付与された条件においても極めて高い耐 SCC 性を有することを明らかにしている。

第 4 章では、圧力容器本体を成す LAS とその上に溶接された Ni 基合金溶接金属との界面近傍での SCC の進展あるいは停留挙動について論じている。ここでは従来の CBB 試験法を応用した中型 CBB 試験法を開発し、効率の高い試験を実現している。板厚内に Ni 基合金溶接金属と LAS 界面を含むよう試験片を準備し、Ni 基合金溶接金属側から発生した SCC の界面近傍でのき裂先端の状況を詳細に観察しており、LAS 側にき裂状に進展するケースと、球状腐食を形成して停留するケースを確認している。また、各々のき裂に対して、与えた曲げひずみとそのき裂形状から FEM 解析を用いてき裂先端の KJ 値を算出し、持続的に LAS 側でき裂が進展する条件は、500ppb の硫酸ナトリウム添加環境かつ KJ 値が $75\text{MPa}\sqrt{\text{m}}$ 以上であること定量的に示した。この条件は、LAS のき裂進展速度と LAS の酸化速度の競合により決まるとの仮説を立て、LAS 単体のき裂進展線図から予想されるき裂進展速度と中型 CBB 試験片観察結果から見積もった酸化速度を比較している。き裂進展速度が酸化速度を上回る条件はおおよそ $80\text{MPa}\sqrt{\text{m}}$ 以上であり、上述の仮説を検証している。

第 5 章は結論であり、種々の材料において SCC が進展性に遷移する際の主要パラメーターの寄与を総括して述べている。

以上、本論文は BWR の主要な機器を構成する材料における SCC において、割れが進展性に遷移するためのキーパラメーターを明らかにしたものである。本論文で得られた知見は、BWR プラントの健全性維持の高度化に有益な指針を与え、材料工学ならびに量子エネルギー工学の発展に寄与するところが少なくない。

よって本論文は博士 (工学) の学位論文として合格と認める。