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The Slow Strain Rate Dependence of Zircaloy-4 Cladding Tube in Iodine Atmosphere (I)

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요드분위기에서 지르칼로이 피복재의 저변형율속도 의존성(I)

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Abstract

The effects of temperature and strain rate on the I-SCC behaviors of Zircaloy-4 were investigated by constant load test at 300°C and constant elongation rate test at 300, 350 and 400°C in 3.34mgI_2/cm^3. The results showed that I-SCC susceptibility increased as the strain rate decreased or the temperature increased. The empirical relation between the stress and the time to failure at 300°C was given by $1/t_f \propto \exp(0.3\sigma/\sigma_{UTS}-31.5)$ When the I-SCC susceptibility was experessed by the ratio of fracture energy in iodine atmosphere to that in the inert atmosphere, severe I-SCC susceptibility was found near 7.6×10^{-6}/sec at 300°C and the maximum point of I-SCC susceptibility tended to shift to the higher strain rate with increasing the temperature. The quasi-cleavage fracture was observed in I-SCC fracture surface. From these results, it was certain that the film repture step was involved as an important process in the I-SCC mechanism of Zircaloy-4.

요 약

온도 및 연신율 변화가 Zircaloy-4의 요드 응력부식 거동에 미치는 영향을 300°C에서 일정 하중법과 300, 350, 400°C에서 일정 연신율법으로 (10^{-5}/sec~10^{-6}/sec) 3.34mg I_2/cm^3의 요드분위기에서 연구하였다. 요드 응력부식균열에 대한 저항성은 온도가 상승하거나 변형속도가 감소하면 감소했고 파손 시간과 응력과의 관계는 $1/t_f \propto \exp(0.3\sigma/\sigma_{UTS}-31.5)$로 표시할 수 있었다. 300°C에서 요드 응력부식 균열에 대한 저항성을 불활성 분위기에서의 파손에너지에 대한 요드분위기에서의 파손 에너지의 비율로 표시할 때 변형속도가 7.6×10^{-6}/sec 부근에서 저항성이 가장 낮았고 온도가 350°C, 400°C로 증가함에 따라 보다 높은 변형속도에서 최저 저항성을 나타내는 경향을 보였다.

요드 응력부식 균열의 파단면에서 준-벽계 파면(quasi-cleavage fracture)을 관찰했다. 전술한 결과에 의하면 Zircaloy-4의 요드 응력부식균열의 기구에 있어서 보호 피막파손단계(film rupture step)가 중요한 과정으로 추정된다.

Introduction

Zircaloy-4 has been employed as nuclear material for their excellent neutron economy and corrosion resistance. Zircaloy cladding material however, is subjected to stress corrosion cracking (SCC) induced by fission products such as iodine, cesium, etc.[1),2)]

Some phenomenological examinations indicated that the stress corrosion cracking behavior depended on strain rate[3),4)], temperature and corrosion agents.[5),6)]

Recently, many laboratory experiments have been undertaken on iodine-induced SCC(I-SCC) of Zircaloy. Their test methods can be grouped into five; mandrel[7),8)] indentor[9)], internal pressurization[10),11)], tensile loading and constant elongation rate method.[12),13)] Among these methods, the internal pressurization method has been wildely used as a standard for examining I-SCC behavior of Zircaloy tubing because it provides a similar stress condition of pellet-cladding-interaction (PCI) event. Several workers who used this method have reported that SCC susceptibility of Zircaloy-4 in the gaseous iodine environment varied with strain rate and particularly, suppressed at very low strain rate($\sim 10^{-7}$/sec).[14),15)] But the interal pressurization method has disadvantage that strain rate shoud be calculated by the total strain rate divided by the time to failure. On the other hand, constant elongation rate test (CERT) is more useful to record the stress level and strain rate at which severe I-SCC behavior is behavior is observed. It also is simpler than the interal pressurization method and has advantage of using a smaller specimen size.

Therefore the purpose of this work is to investigate the strain rate dependence of I-SCC of Zircaloy-4 at 300, 350 and 400°C with various strain rates (10^{-5}/sec$\sim 10^{-6}$/sec) using CERT and elucidate the I-SCC mechanism of Zircaloy-4.

Experiment

The material used in the present study is stress relieved Zircaloy-4 cladding tube (inner diameter 12.08mm, thickness 0.7mm), which was produced by Sandvik Special Metals Corp. The specimens of 5.0mm in length was used to eliminate the specimen size effect on the tensile properties of the ring specimens.[16)]

Table-1 lists the chemical compostion of the Zircaloy-4 used in this test.

Two kinds of test, CERT and costant load test, were conducted: CERT was carried out in iodine containing Ar atmosphere as well as in Ar gas atmosphere at 300, 350 and 400°C. Constant load test was also performed in iodine containing Ar at 300°C. The test system used in this work is shown in Figure 1. Cross head speeds of 0.1, 0.05, 0.01 and 0.005mm/min. were chosen for the CERT. Iodine content was 3.34mg/cm^3 for every test run. Load-elongation curves were recorded for the CERT, while the applied load and the time to failure were obtained for the constant load test.

Table 1. The Chemical Compositions of The Zircaloy-4 (ppm by weight unless otherwise stated)

Element	Composition	Element	Composition
Sn	1.46%	Cl	<20
Fe	0.21%	Pb	<20
Cr	0.10%	Co	< 5
Fe+Cr	0.31%	Cu	<10
0	1160	Hf	40
Al	<20	H	11
C	130	Mn	<20
Nb	<20	Mo	<10
Ta	<20	Ni	15
N	35	Zr	balance

Table 2. Circumferential Tensile Properties

Temperature (°C)	Strain rate ($sec.^{-1}$)	Environment					
		Ar			Ar+3.34mgI_2/cm^3		
		UTS (MPa)	0.2% Y.S (MPa)	Elongation (%)	UTS (MPa)	0.2% Y.S (MPa)	Elongation (%)
300	3.8×10^{-6}	406.1	348.4	15.9	399.0	344.4	14.0
	7.6×10^{-6}	414.0	350.0	16.9	403.7	331.4	6.4
	3.8×10^{-5}	417.2	355.9	15.6	406.0	352.9	12.2
	7.6×10^{-5}	431.7	379.2	15.5	417.7	354.0	16.0
350	3.8×10^{-6}	383.4	336.0	18.8	361.7	314.4	8.7
	7.6×10^{-6}	385.1	347.2	15.5	364.1	297.6	9.3
	3.8×10^{-5}	395.1	350.2	10.8	382.0	321.9	12.0
	7.6×10^{-5}	408.2	367.3	14.5	408.2	345.4	18.8
400	3.8×10^{-6}	355.6	261.5	12.1	350.7	247.4	6.4
	7.6×10^{-6}	365.3	270.8	11.1	344.9	244.2	7.5
	3.8×10^{-5}	374.7	277.7	17.5	371.1	274.7	11.0
	7.6×10^{-5}	403.7	332.5	20.6	402.4	308.1	16.0

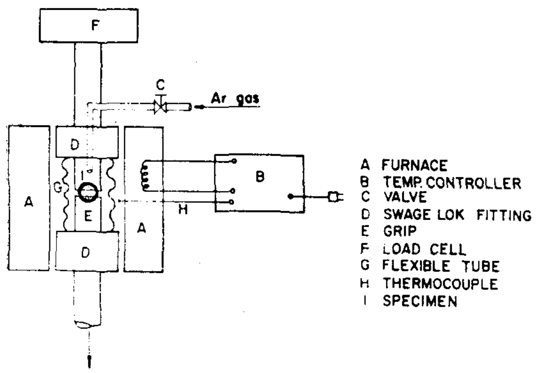


Fig. 1. Schematic Diagram of Test Appratus.

I-SCC susceptibilities were assesed by measuring ultimate tensile strength, 0.2% yield strength and relative fracture energy.[17] Fracture surfaces were examined using scanning electron microscope.

Results and Discussion

The effects of strain rate and temperature on tensile properties in Ar and iodine containing Ar are showed in Table 2. The gauge length was assumed to be 43% of the circumference based on the average distance over which plastic deformation had been detected. This assumption is well in accordance with the previous reports.[4),18)] These results show that the increase of initial strain rate led to the increase of the ultimate tensile strength and 0.2% yield strength. They also show that the increase of the test temperature by 50°C decreased the mechanical properties by about 21MPa. The serration phenomena could not be found except under the test condition of 400°C and 3.8×10^{-6}/sec.

Figure 2 shows the variation of time to faliure with applied stress at 300°C for constant load test. It was found that the time to failure increased with decreasing applied stress. I-SCC

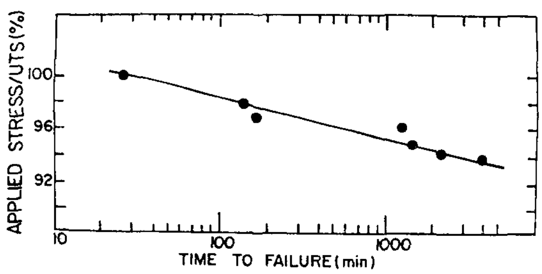


Fig. 2. Circumferential Stress Dependence of I-SCC at 300°C, 3.34mgI_2/cm^3.

is known for thermally activated process and its behavior is expressed in stress and temperature terms.[15),19)] The following relationship between time to failure and stress was obtained at 300°C;

$1/t_f \propto \exp(0.3\sigma/\sigma_{UTS} - 31.5)$

On the other hand, the mechanical properties in iodine atmosphere are lower than those in the inert atmosphere under the testing conditions, Thus it is concluded that maximum stress, which might contribute to crack initiation, was lower in iodine containing Ar atmosphere than in Ar gas.

Figures 3 and 4 show typical shapes of the failed specimen and fractographs. The specimens tested in Ar gas alone falied in cup-cone mode with dimple but the specimens in iodine atmosphere failed in the quasi-cleavage fracture mode.

From these results, it is apparent that gaseous iodine contributes to decreasing the stress required to cause failure at a given time.

It generally known that if SCC process involves the rupture of protective films, the minimum values of the average strain rate at which SCC is found should be observed and increase with increasing temperature to balance the

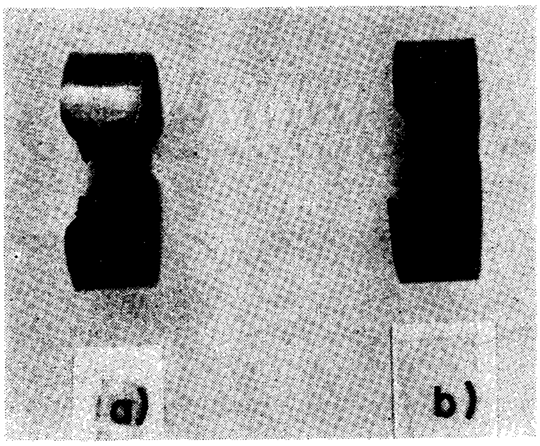
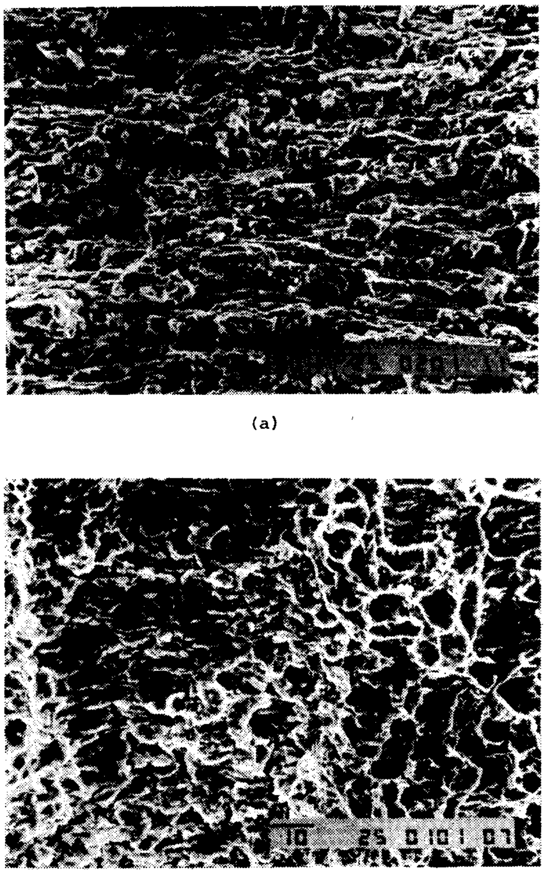


Fig. 3. Failure Types of Ring Tension Specimen by CERT at 300°C ($\dot{\varepsilon}=3.8\times10^{-6}$/sec).
a) Ar gas
b) Ar+3.34mgI_2/cm^3



(a)

(b)

Fig. 4. Scanning Electron Micrographs of Fracture Surface Tested by CERT at 300°C, 3.34mgI_2/cm^3.
a) $\dot{\varepsilon}=3.8\times30^{-6}$/sec.
b) $\dot{\varepsilon}=7.6\times10^{-6}$/sec.

increasing rate of the competing film formation process.[20),21)] It also was reported that the formation of solid iodide was favored and the small amount of iodine remaining in the vapour phase was entirely in the form of Z_rI_x at zirconium-iodine system.[22),23)] Figure 5 illustrates the relative fracture energy ratios of iodine containing Ar to Ar gas alone with temperature. It shows that the maximum susceptibilities of I-SCC occurred and tended to shift to the higher strain rate from about 7.6×10^{-6}/sec with temperature. Therefore, these results are qualitatively consistent with expectations based

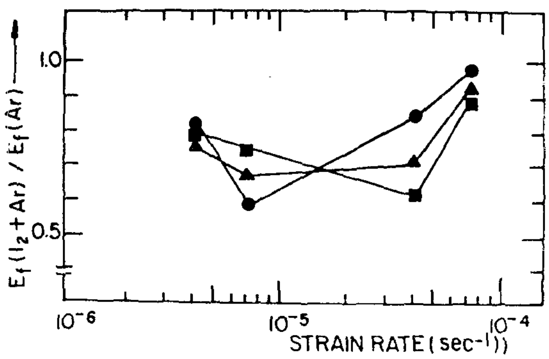


Fig. 5. I-SCC Susceptibility of Zircaloy-4 Expressed by Fracture Energy Ratio with Strain Rate at Various Temperature.
• 300°C ▴ 350°C ▪ 400°C

on SCC mechanism involving film rupture.

Conclusions

1. I-SCC susceptibilities of Zircaloy-4 were dependent upon strain rate and temperature.

2. The empirical relation between stress and time to failure of I-SCC at 300°C is: $1/t_f \propto \exp(0.3\ \sigma/\sigma_{UTS}-31.5)$.

3. The mechanical properties and fracture energies were substantially lowered by gaseous iodine attack, and severe I-SCC behaviors were observed near 7.6×10^{-6}/sec at 300, 350 and 400°C in 3.34mgI_2/cm^3.

4. The quasi-cleavage feature was observed in I-SCC fracture surfaces.

5. under the test condition, I-SCC behaviors of Zircaloy-4 indicated that the film rupture step was involved as an important process in the I-SCC mechanism of Zircaloy-4.

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