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Beryllium Analysis on the Brazing Zone of Zircaloy-4 Cladding

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Zircaloy-4 피복관 부레이징 계면의 베릴륨 분석

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Abstract

The distribution behaviors of beryllium which may produce a deleterious damage in the zircaloy cladding have been investigated by the X-ray line scanning of EPMA. The results obtained are as follows;

1) The alloy phase formed by the brazing contains ~6.3 mass% of beryllium.
2) The beryllium diffusion in the base metal (cladding and bearing pad) is recognized only in the range of ~5 μm from the brazing interface.

요 약

Zircaloy-4 피복관의 조사손상에 영향을 줄 수 있는 부레이징 계면의 베릴륨 분포거동을 EPMA의 X-선 선분석으로 조사하여 다음과 같은 결과를 얻었다.

1) 부레이징에 의해 생성된 합금상의 베릴륨 함유량은 ~6.3mass%이었다.

2) 베릴륨의 기지금속(피복관 및 베어링 패드)내 확산은 부레이징 계면으로부터 ~5μm 범위 외에서는 무시될 수 있었다.

1. Introduction

The joining of bearing pad or spacer to the zircaloy cladding is one of the major process in the fuel rod manufacturing. The bearing pad or spacer is joined to the cladding by brazing in which a filler material adheres on base metal by diffusion and alloying.

Beryllium has particularly attractive properties for using in zircaloy brazing, because this metal has a low thermal neutron absorption cross section[1] and has a low eutectic melting point with zirconium[2]. However, this metal has a large absorption cross section[3] for (n, α) reaction as shown in Table 1, so a deleterious damage may be produced in the brazing zone of zircaloy cladding dependent on beryllium distribution. Therefore, it is considered that distribution behaviors of beryllium in the brazing zone may influence on the integrity of fuel rod, but there is no available paper on the distribution behaviors of beryllium in the brazing zone of zircaloy cladding.

The objective of the present study is to investigate the distribution behaviors of beryllium which induce the severe irradiation damage in the brazing zone of zircaloy cladding.

Table 1. Cross Section for (n,α) Reaction of Fission Neutron[3]

Elements	Cross Section (mbarns)	Elements	Cross Section (mbarns)
Be	125	Ni	0.60
B	99.5	Fe	0.44
Al	3.10	Cr	0.38
Mg	2.90	Zr	0.18
Mo	0.72	Nb	0.04

2. Experimental

2.1 Specimen Preparation

Commercially available zircaloy-4 cladding and plate were used for this investigation. The chemical composition of zircaloy-4 is shown in Table 2. The bearing pad was prepared from the plate by the press and followed cleaning to get a fine beryllium coating. Thin film coating of ~5 μm thickness of beryllium on the pad surface was carried out in non-oxidation atmosphere. Before brazing of the pad to cladding, the pad was tightly contacted to the cladding by the spot welding and was then brazed at 1273K for 20 sec using high frequency induction heating type furnace under a vacuum of 10^{-3} Pa.

Table 2. Chemical Composition of Zircaloy-4

Elements	Sn	Cr+Fe	Others	Zr
Mass %	~1.4	~0.35	~0.15	balance

2.2 EPMA analysis

The analysis of composition in the brazing zone was performed by using of Electron Probe Micro Analyzer(EPMA). The specimen for EPMA analysis was grinded with SiC emery paper and was then polished with diamond paste to obtain a fine flat surface. Zirconium analysis was performed by the ZrLα X-ray line scanning and tin analysis was also conducted by the SnLα X-ray line scanning. However, beryllium content in the brazing zone was estimated from the content of zirconium and tin obtained from ZrLα and SnLα X-ray line scanning, because beryllium, which is lighter than nitrogen, can not analyze by the X-ray line scanning of EPMA.

2.3 Hardness Measurement

Vickers micro hardness test was also used to investigate compostion difference in the brazing zone of zircaloy cladding, because a test of highly sophisticated microhardness can be applied to the assessment of composition difference in alloy phase. The microhardness readings in the brazing zone was conducted each with 50 and 100 gf indentation loading.

3. Results and Discussion

Zircaloy-4 seems to be inherently resistant to void formamtion and associated swelling in nuclear reactor[4-8]. However, zircaloy brazed with beryllium can produce void filled of helium gas, because beryllium easily transmutes to hellium by the (n, α) reaction in the nuclear reactor. Therefore, the distribution behaviors of beryllium in the brazing zone of zircaloy cladding is of special interest.

Figure 1 shows the results of X-ray line scanning of EPMA across the brazing zone which do not contains metallic islands. The content of zirconium and tin obtained form the results of ZrLα and SnLα X-ray line scanning in base is ~92 and ~1.2 mass%, respectively. With consideration of that zircaloy-4 contains 0.5 mass% of minor alloying elements such as chromium, iron and oxygen as shown in Table 2, the content of beryllium in the alloy formed by the brazing is estimated ~6.3 mass%. The diffusion layer thickness of beryllium in the base metal (cladding and bearing pad) is measured to be about 5 μm from ZrLα X-ray line scanning as shown in Figure 1(b).

X-ray line scanning of EPMA across the brazing zone containing metallic islands. The marked a, b and c in the microphotograph show the metallic islands which are known to be zirconium base metal from the analysis results of ZrLα and SnLα X-ray line scanning. The major composition

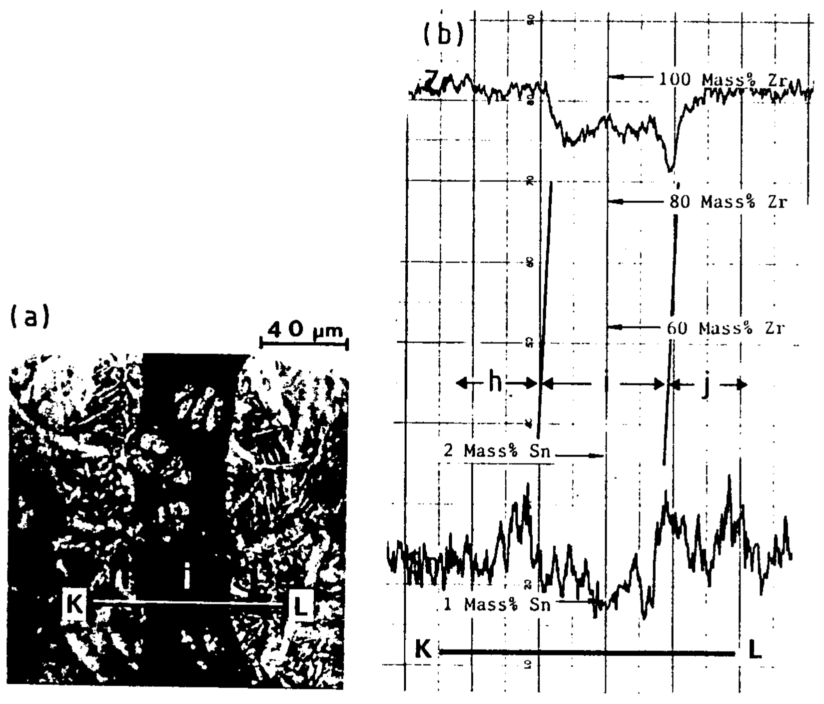


Fig. 1. Electron Probe Microanalysis of Alloy Formed During the Brazing; (a) Microphotograph, (b) ZrLα and SnLα Line Profiles Across the Line K-L in (a).

elements of this metallic islands are zirconium and tin of which the contents are ~97.5 and ~1.15 mass%, repectively. In view of minor alloying elements contained in zircaloy-4, the beryllium in the metallic islands was estimated to be ~1.5 mass%. Such amount of beryllium is recognized, because the diffusion thickness of beryllium in the zircaloy base metal is estimated to be only about 5 μm.

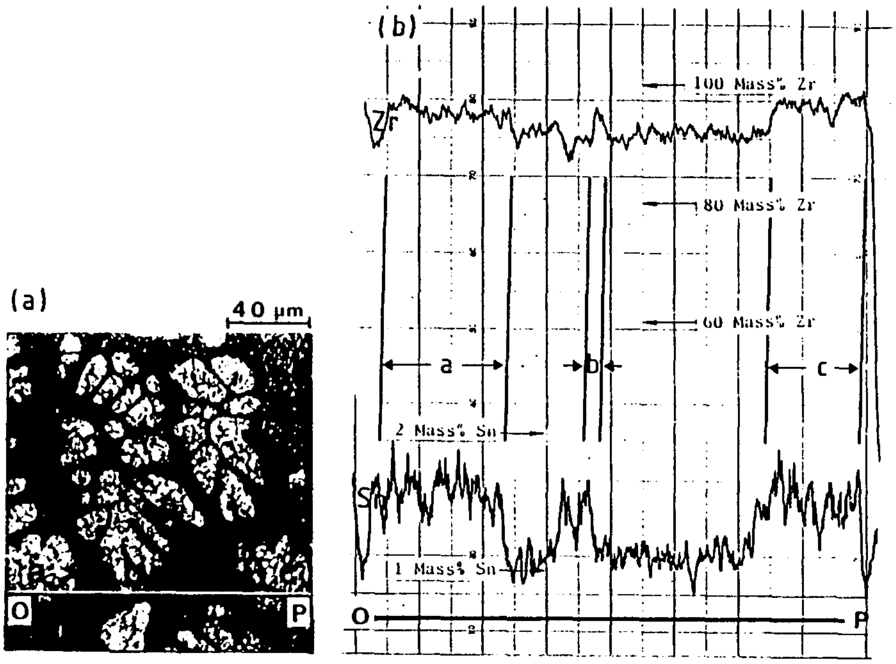
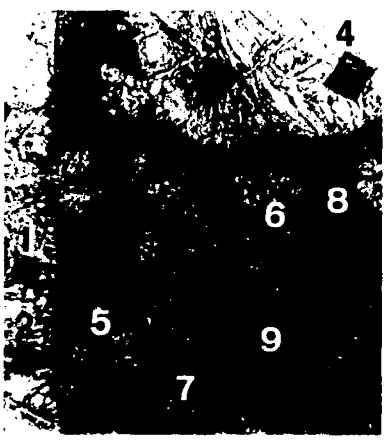


Fig. 2. Electron Probe Microanalysis of Brazing Zone with Metallic Island; (a) Microphotograph, (b) ZrLa and SnLα Line Profiles Across the Line O-P in (a).

Table 3. Vickers Hardness in the Brazing Zone



No	Load (gf)	Hardness (Hv)	Position
1	50	237	base metal
2	50	244	base metal
3	100	239	base metal
4	100	227	base metal
5	50	265	metallic island
6	50	281	metallic island
7	50	253	metallic island
8	100	315	alloy formed during brazing
9	100	306	alloy formed during brazing

The results of hardness measurement conducted in the brazed zone is shown in Table 3. The hardness of the metallic island is scattered in the range of 253-281 Hv. These values are higher than the hardness number of the base metal. Substitutional elements that are different size from the solvent atom induce solid solution hardening, because single atom or groups of atoms whose size differ from the solvent atom impose a system of internal stress dependent on the atomic size misfit ratio[9-11]. Since Goldschmidt atomic diameters of zirconium and beryllium are 3.19 nm and 2.25 nm[12] respectively, a large solid solution hardenings occurs in the zirconum-beryllium alloy system. Therefore, a lot of hardening in the metallic islands is another evidence of containing beryllium.

4. Conclusions

By the X-ray line scanning using of the EPMA, the distribution behaviors of beryllium in the brazing zone of zircaloy-4 cladding have been investigated. The results obtained are as follows;

1) The alloy phase formed by the brazing contains ~6.3 mass% of beryllium.
2) The amounts of beryllium diffused in base metal (cladding and bearing pad) is negligible except the region of ~5 μm from the brazing interface, thus the deleterious damage due to (n, α) reaction of beryllium diffused in cladding is not expected.

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