Development of Multi-Walled Cold Drawing Process for Improved Accident Tolerant Fuel Cladding in Light Water Reactors

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1. Introduction

The nuclear fuel claddings used in light water reactors (LWRs) consist of zirconium and a small number of metals. However, at the time of the accident in Fukusima, Japan, zirconium alloy (Zr alloy) cladding reacted with high-temperature steam in loss of coolant accident (LOCA) and a large amount of hydrogen gas was generated [1]. In order to solve these problems, various studies are underway to development of the accident-tolerant fuel (ATF) cladding.

In this research, a drawing process was used to improve mechanical strength under high temperature conditions by using stainless steel 316(SUS-316) tube inside and outside of the existing ZIRLO cladding. Because of good plastic workability compared to other ferritic/martensitic stainless steels, 316 austenitic stainless steel was chosen for drawing process materials. But the thermal conductivity is low and thermal neutron absorption cross section is more than 10 times larger than Zr alloy [2]. Therefore, using as thin as possible SUS tube is very important factor of drawing process. The development of Multi-Walled Drawing process shown in Fig. 1 which is very simple and performs at room temperature can be mass-produced using existing Zr-alloy cladding. High temperature exposures up to 1200°C in air were performed to demonstrate the oxidation resistance of Multi-Walled Drawing Cladding (MWDC) and evaluate each layer interface between SUS and ZIRLO [3].



Fig. 1. Images of the drawing process (a) schematic diagram of the drawing and (b) a photograph of a sample placed into the machine, and (c) a photograph during drawing.

2. Methods and Results

In this section some of the techniques using swaging and drawing process are described. Swaging is a process that produces an outer diameter of the desired size by applying force towards a tube having a large outer diameter [4]. Drawing is a metalworking process which uses tensile forces to stretch metal, glass, or plastic [5].

2.1 Swaging and drawing process

The Multi-Walled Drawing Cladding (MWDC) consists of three-layer tube. Drawing process is carried out inside and outside the ZIRLO tube (outer diameter: 9.50mm, inner diameter 8.23mm, length 0.5m). The inner tube was SUS-316 (outer diameter: 9.93mm, inner diameter: 9.80mm, length: 0.5m) and the outer tube was SUS-316 (outer diameter: 8.15mm, inner diameter: 7.70mm, length: 0.5m). Fig. 2a shows picture of each tube diameter and size comparison. The MWDC was prepared by inserting inner and outer SUS-316 tube in the ZIRLO shown in Fig. 2b. When drawing, adjust the size of the dies and draw to the desired outer diameter. Swaging process should be preceded, about 15cm in length of the multi-walled tube to hold the front part when drawing. The first one was drawn twice with 9.6Φ , 9.5Φ die and the other was drawn with 9.6Φ , 9.5Φ and 9.4Φ die three times. In order to secure the inner diameter, put a metal filler of desired size inside the tube shown in Fig. 1a. The 7.5Φ diameter cylindrical metal filler was used to secure the inner diameter. Fig. 2c shows photographs of the MWDC after multi-walled tube drawing. It is confirmed that the length of the external SUS-316 tube is about 1~2mm became longer than the length of the ZIRLO tube.

2.2 Heat treatment test

ZIRLO, 9.4Φ MWDC and 9.5Φ MWDC cut into 1.5cm were prepared and a furnace was used for heat treatment. The three specimens were exposed to 600°C, 900°C and 1200°C respectively in air condition. Each condition was increased at a rate of 300°C/h, at room temperature. Each specimen was maintained at 600°C, 900°C and 1200°C for 10 min and decreased temperature naturally to room temperature.



Fig. 2. Sample preparation process pictures: (a) Comparison picture of each tube diameter; (b) Tube assembly before drawing; (c) Photo of Multi-Walled Drawing Cladding (MWDC); (d) Samples cut for examination and heat treatment testing.

2.3 Results

After drawing, the thickness change of each tube and MWDC presented in Table. 1. Comparing the outer inner diameter sizes of ZIRLO with 9.4 Φ MWDC and 9.5 Φ MWDC, the outer diameter was almost similar, with a difference of 0.06mm and 0.03mm respectively, and the inner diameter difference was the same as 0.63mm.

In the optical microscopy (OM) picture, Fig. 3, it was confirmed that a slight gap occurred between the inner tube and the ZIRLO, and a gap of about $5\mu m$ for 9.4Φ MWDC and about $20\mu m$ for 9.5Φ MWDC was observed, and there was no gap between the ZIRLO and the outer tube interface.

In Table. 2 shown the weight change for each specimen due to the oxidation in the air at 600°C, 900°C, 1200°C. The weight change after heat treatment of each specimen was significantly different at high temperature. At 1200°C, the mass increase of the 9.4 Φ MWCD specimen and the mass increase of ZIRLO differed by more than about two times. The mass of the 9.5 Φ MWDC specimen was found to have a weight increase about 5% compared to the 9.4 Φ MWDC specimen. Specimens heat-treated at 1200°C were examined by scanning electron spectroscopy (SEM) in conjunction with energy dispersive spectroscopy (EDS) for microstructure, oxidation effects and interdiffusion between ZIRLO and inner outer tube (Figure 3). As a

result of EDS analysis, the O/Zr atomic ratio of 9.4 Φ MWDC was the lowest at 0.75 and the ZIRLO atomic ratio was the most oxidized at 1.14 and the atomic ratio of 9.5 Φ MWDC was 1.

Table. 1 Changes in the size of the outer diameter (O.D) and inner diameter (I.D) before and after the drawing process.

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	Outer SUS 316 tube	ZIRLO	Inner SUS 316 tube	9.5Ф MWDC	9.4Ф MWDC
O.D	9.95 mm	9.50 mm	8.15 mm	9.53 mm	9.44mm
I.D	9.80 mm	8.23 mm	7.70 mm	7.60 mm	7.60mm
Thickness change after drawing (%)	6.93% decrease	1.09% decrease	6.66% decrease	1.07% decrease	2.62% decrease

Table. 2. Measurement of weight change of oxidation degree after high temperature heat treatment.

	ZIRLO	9.5Φ MWDC	9.4Φ MWDC
Weight change at 600°C(mg)	1662→1665 (0.18% increase)	1980→1982 (0.10%increase)	2257→2258 (0.04%increase)
Weight change at 900°C(mg)	1510→1551 (2.72% increase)	2577→2612 (1.36% increase)	2344→2373 (1.24% increase)
Weight change at 1200°C(mg)	1687→2275 (34.85%increase)	2071→2509 (21.15%increase)	2141→2504 (16.95%increase)

3. Conclusions

A ZIRLO based MWDC was successfully fabricated using a thin thickness SUS-316 tube swaging and drawing process. To confirm the oxidation resistance and high temperature stability of MWDC, heat treatment was performed at 600°C, 900°C and 1200°C and the weight change was measured. Compared to other specimens, 9.4Φ MWDC specimen was the most effective, and it was confirmed that the integrity of the interface after drawing influences the degree of oxidation. As a result of EDS analysis, it was also confirmed that the oxidation degree of the 9.4Φ MWDC specimen was the least. In order to analyze the phase transformation of the MWDC and its oxide more accurately, it is planned to proceed further with an XRD pattern. In conclusion, it is expected that better accident-tolerant fuel (ATF) can be derived if the interface problem between tubes is solved in Multi-Walled Drawing Cladding (MWDC) production.



Fig. 3. Cross-sectional OM images (a) to (c) for 9.4Φ MWDC, and (d) to (f) for 9.5Φ MWDC, to analysis each specimen interface.



Fig. 4. SEM cross-sectional images of after 1200°C heat treatment: (a) ZIRLO; (b) 9.5Φ MWDC; (c) 9.4Φ MWDC

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