

The internal pickling of Zr-1%Nb cladding tubes to enhance the dimensions control and surface quality

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Abstract

The internal surface quality of cladding tubes and also, the proper gap between the inside-diameter of Zr-Nb 1% nuclear fuel cladding tubes and uranium dioxide pellet, outer-diameter are the important factors regarding the pellet-clad mechanical interaction, corrosion and heat transfer in the fuel rods. One approach to attain the proper gap is by obtaining the correct inside-diameter of the cladding tubes, which is close to the design nominal diameter. Hence, A novel pickling system based on the flowing the specified pickling solution in the cladding tube was considered. The results show the suitable dimensions control with small variation, and the enhancement of the surface quality. Also, the effect of the suggested pickling on the fluoride surface compared to the common immersion pickling, display the lower fluoride adsorption. So, the operation of the internal pickling system is promising for economical and large scale production.

Keywords: Cladding Tubes, Pickling, Zr-Nb 1% alloys, Inside-Diameter

Introduction

Generally in Zr-Nb alloys cladding tubes for boiling water reactors (BWR's), the inside surfaces are pickled with $\text{HF}+\text{HNO}_3+\text{H}_2\text{SO}_4+\text{H}_2\text{O}$ and neutralization is applied at the final stage of fabrication to ensure the surface cleanliness [1, 2].

The dipping or immersion method is usually used for this etching and rinsing process, in which tubes are dipped into vertical or horizontal tanks. In this system, an equal amount of pickling removal in the longitudinal direction cannot be easily obtained. So, it is very difficult to control dimension and to have correct surface quality. To resolve these problems, we developed a unique pickling system with inside surface pickling can be carried out. Though this system produced good dimension and surface quality, manual measurement of diameters by an air micrometer was needed just before etching each tube. This hand operation is not suitable for economical and large scale production.

To eliminate manual diameter measurement, we have developed a pickling system to control the inside diameter of the cladding tubes. Furthermore, the inside-diameter of all the tubes can be controlled more accurately by applying this system.

This paper presents an outline of the inside-diameter control system and quality data on tubes processed by the internal pickling system. The effects of parameters such as pickling solution temperature, velocity, HF concentration, pickling and rinsing time on the rate of internal surface corrosion and surface quality were considered.

Experimental

The internal pickling system

The new internal pickling system is performed in four main steps: 1) degreasing, 2) internal pickling, 3)

rinsing by Nitric acid (stop bath solution), and 4) rinsing by the cold and hot soft water.

A schematic of the inside surface pickling equipment is shown in Fig.1, which shows Zr-Nb 1% cladding tubes on the supporting rolls attached to a nozzle. The inside surface is etching by the pickling solution ($\text{HF}+\text{HNO}_3+\text{H}_2\text{SO}_4+\text{H}_2\text{O}$), which is pressurized by a pump and supplied into the tube from a service tank by opening valve. The pickling solution in a receiving tank and circulated back to the service tank.

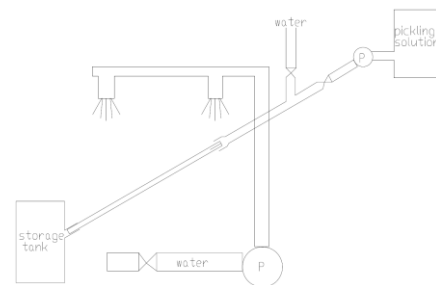


Fig. 1) Schematic of the internal pickling system

During the pickling process, water is sprayed on to the outlet surface of tube by the shower nozzles attached above the equipment. This keeps the tube temperature uniform throughout the whole length.

Characterization

The surface quality of the final pickled cladding tube, is characterized with SEM. Also, the manual measurement of inside-diameter is measured by a micrometer.

The fluoride ion selective electrode (F-ISE) for fluoride determination is used which is widely adopted as a routine method for fluoride determination in nuclear industry [3].

Results and discussion

According to the screening designs, the effective parameters such as pickling solution composition, temperature and velocity, HF concentration and pickling time were determined. The results showed that according to the limitations of the fluoride specification ($0.15 \mu\text{g}/\text{cm}^2$) and scale removal (about $10\text{-}15 \mu\text{m}$ in radial of tube), the pickling solution composition should be defined as Table 1.

Table 1. composition of pickling solution

Pickling solution composition	%
H ₂ SO ₄	30 ±5
HNO ₃	30 ±5
HF	1.5 ±0.5
H ₂ O	38

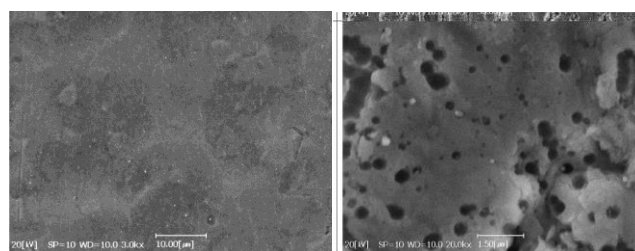
30 ±5 %, 30 ±5 % and 1.5±0.5 % HF. Also, the solution velocity was adjusted around 3 m/s.

According to the exothermic reaction, pickling solution temperature was kept around ambient temperature (25 °C) and in addition, the outer surface of tube should be sprayed by water shower nozzle to control the solution temperature. It is important to control the temperature because of the high temperature could be enhanced the fluoride adsorption and scale removal.

After pickling step is completed, immediately, the preliminary rinsing should be pumped in tubes. The preliminary rinsing of zirconium-niobium alloys must be in a 50 volume % of HNO₃ solution at room temperature as an aid in removal of a black surface residue (smut) which may develop during etching. Following the preliminary rinses with HNO₃, rinse the specimens in flowing water. The final rinse may be performed by hot soft water (80 °C). Then, pickling tubes may be air-dried, wiped dry with a clean, lint-free cloth, or blown free of moisture with dry air that is free of dust and acid fumes. Figure 1 shows the photograph of zirconium-niobium clad tubes with delay and no delay in rinsing. Also, Figure 2 shows the SEM of zirconium-niobium clad tubes with delay and no delay in rinsing. It is shown that delay in rinsing can impose the reaction products deposit on the surface.



Figure 1. The photograph images of zirconium-niobium cladding tube a) no delay, b) with delay in rinsing step



a b

Figure 2. The SEM images of zirconium-niobium cladding tube a) no delay, b) with delay in rinsing step

The results of fluoride determination were shown in Table 2. The results of the pickling process with optimum conditions of this research on fluoride adsorption on the surface tube showed that the reported data is under the specification limit ($0.15 \mu\text{gr}/\text{cm}^2$).

Table 2. The results of fluoride determination before and after the pickling process

Step	Before Pickling	After Pickling
Fluoride Test ($\mu\text{gr}/\text{cm}^2$)	≤0.1	0.12±0.03

Conclusions

The internal pickling of Zr-Nb 1% cladding tubes as the novel technique, provides suitable dimensions control with small variation, and enhances the surface quality. Also, the effect of the suggested pickling on the fluoride surface compared to the common immersion pickling, displays the lower fluoride adsorption.

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