

Working Group "Hot Laboratories and Remote Handling"
Plenary Meeting, Studsvik, Sweden
5th and 6th of June '97

FORMATION OF ARTIFICIAL HYDRIDE-BLISTERS ON PRE-IRRADIATED FUEL CLADDING FOR IN-REACTOR DEGRADATION EXPERIMENTS

E.H.Toscano*, A.D. Stalios*, R.Schreiber*, P.Nascimento*,

H.Ruhmann^o, M.Körzendörfer^o

^o Siemens AG - KWU/BTW, Postfach 3220, 91050 Erlangen, Germany.

* Institute for Transuranium Elements, Postfach 2340,
76297-Karlsruhe, Germany.

ABSTRACT

1. INTRODUCTION

2. EXPERIMENTAL

2.1. Segment re-fabrication procedure

2.2. Surface preparation

2.2. Hydriding method

2.4. Experimental device

3. RESULTS

3.1. Scouting experiments

3.2. Experiments in the hot cell

CONCLUSIONS

ABSTRACT

Artificial hydride blisters were generated on the surface of two segments re-fabricated from two different BWR-fuel rods previously irradiated in BWR-reactors to burn-ups of 13 and 23 MWd/kgU. An experimental device, based on the local hydrogen diffusion under controlled temperature conditions (≤ 400 °C), was developed. Nickel metal was brought by mechanical alloying onto the previously machined surface of the segments to allow the ingress of hydrogen. Hydrogen gas was continuously supplied to the hydrating chamber at a pressure of about 2 bar. Temperature and pressure were maintained constant typically for about 20 hours. This treatment has successfully been performed in the hot cells and resulted in the formation of locally hydride areas (linear blisters).

1. INTRODUCTION

Failures in BWR barrier fuel rods is a serious problem due to the significant secondary degradation that occurs, causing high releases of activity to the coolant. The rod deterioration, after the occurrence of a primary defect, has been intensively studied by numerous authors on test rods containing artificial defects (see, e.g., refs. 1-2), as well on discharged failed commercial fuel rods (see, e.g., ref. 3).

In order to improve secondary defect behaviour of the Zr liner, Siemens has developed a Fe-enhanced Zr liner cladding⁴. An extended laboratory test program was initiated to check the performance of this type of liner and a product comparison on refabricated segments from pre-irradiated BWR-fuel rods between Zr-liner and a liner alloyed with Fe, was decided. The segments had to be locally charged with hydrogen since, after the occurrence of a primary defect, hydride blisters are considered to be the initiators for the development of long axial cracks typical for secondary degradation⁵.

This paper describes the development of the hydrogen charging procedure and the experimental device used. Periscope observations, as well as metallographic cross sections of the blisters obtained on irradiated material will be presented.

2. EXPERIMENTAL

Several methods are described in the literature (see, e.g., ref. 6) for the formation of a local hydrogen accumulation. Goal of the present work was the production of an artificial blister under the following conditions:

- the heating of the segments should be locally in order to avoid extended fission gas release
- the blister should affect only a part of the cladding wall to avoid problems during the shipment of the segments to the reactor for re-irradiation
- several blisters should be aligned in the longitudinal direction (linear blisters) to favour the development of long cracks.

2.1. Segment Re-fabrication Procedure

Segments, 500 mm in length, were cut from two BWR-fuel rods, irradiated up to 13 and 23 MWd/kgU. The re-fabrication technique is based on the removal of one or more pellets by drilling and the machining of the cladding, internally and externally, to prepare the surfaces for the insertion and subsequent welding of new end plugs. The determination of the fuel stack position was performed by depth measurements. The welding was performed by using an orbital tungsten inert gas (TIG) device from Arc Machines, Switzerland.

2.2. Surface Preparation

After re-fabrication a pre-selected region of the segment was machined in order to locally remove the oxide layer. This operation was performed using a small lathe, housed in an α -tight hot cell, combined with a rotating ceramic tool. In order to only affect part of the surface, the segment was mounted excentrically on the lathe. Afterwards, nickel was rubbed in onto the surface free from oxide, to perform the mechanical alloying.

2.3. Hydriding Method

The method is based on the locally heating in a hydrogen atmosphere of a oxide free surface of a segment, previously treated by nickel mechanical alloying in the way described in § 2.2. Nickel acts as the pathway for hydrogen into the Zircaloy. Depending on the temperature, the hydrogen pick-up takes place slowly and the hydrides precipitates in the zones presenting a temperature gradient.

2.4. Experimental Device

In **Fig. 1**, a schematic diagram of the equipment is shown. The hydriding was performed in a chamber, in which the portion of the segment where the blister was to be developed, was introduced through quick connectors providing the gas sealing. Perpendicular to the segment axis, a brass heating socle was pressed onto the nickel-alloyed surface of the segment by an Inconel spring. A glass window allowed the observation of the correct positioning of the heating socle on the segment surface.

Two thermocouples were brought at the heating socle, one to measure and the other to control the temperature near to the contact surface. The temperature

regulation system allows the target temperature to be obtained with a precision of $\pm 1^{\circ}\text{C}$.

Hydrogen (99.9999 % purity) was continuously supplied through flexible pipes into the hydriding chamber at a pressure of about 2 bars. The hydrogen was introduced passing a heated chamber (about 80°C), containing Zr-chips as getter in order to eliminate any oxygen present in the gas. The purity of the gas was of paramount importance for the successfully production of blisters, H₂ having a certified purity of 99.9999%, was used in all the experiment performed.

3. RESULTS

3.1. Scouting experiments

Several scouting experiments were performed on non-active materials to determine the optimum conditions (time, temperature, gas pressure, etc.) for the formation of the desired blister configuration. In **Fig. 2.** a blister developed on a similar non-irradiated material, is shown. It can be seen that the blister developed up to about one half of the wall thickness. This was the desired configuration and of paramount importance to preserve the mechanical integrity during the transport, since the segments had to be shipped to the reactor for the irradiation and a full hydride wall could had been dangerous for this operation.

3.2. Experiments in the Hot Cell

In **Fig. 3** a photo of the experimental device, before introduction in the hot cell, is shown.

Fig. 4 shows the typical appearance of the machined region, where the Ni was brought and the blister formed, as seen through the periscope in the hot cell. The presence of several blisters randomly distributed on the heat affected zone, marked by the halo, is clearly seen.

In **Fig. 5** the metallographic pictures taken on similar irradiated material is shown. As expected, several blisters with the typical "sunburst" shape were formed. Again, as in the cold tests, the hydrogen accumulation affected only about one-half of the wall thickness. Moreover, from **Fig. 5** can be seen that several blisters are aligned in the axial position (linear blisters) to favour the formation of longitudinal secondary degradation cracks during irradiation.

After the hydriding operation, a hole 1 mm in diameter, was drilled about 100 mm from the hydrided zone to simulate the primary defect.

CONCLUSIONS

A method to produce artificial hydride accumulation (blister), able to operate under remote handling conditions, was developed. Two pre-irradiated segments were locally charged with hydrogen and transported to the Halden reactor for irradiation to perform a product comparison.

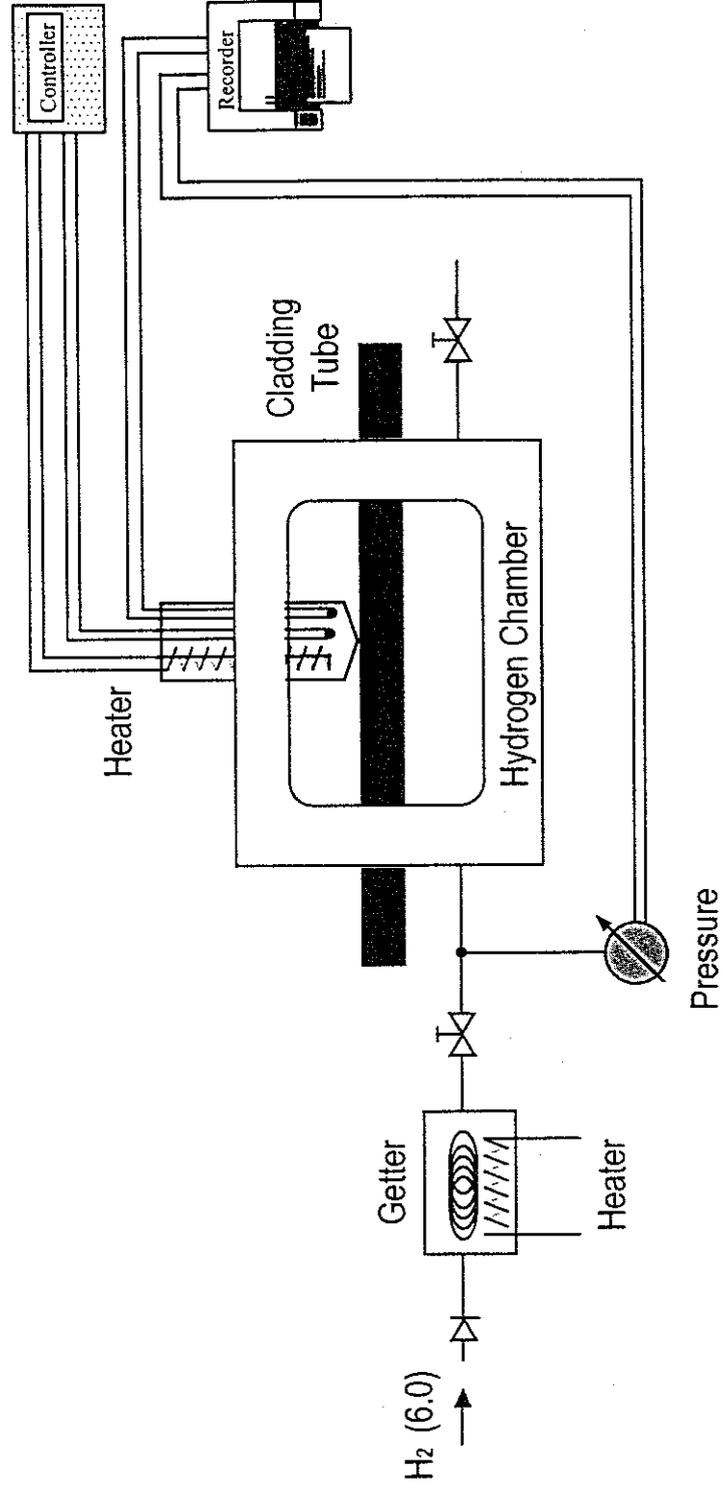
References

1. C.Gräslund et al, "Studies of the secondary hydriding process in fresh test fuel with simulated primary defects", Proceedings of the ANS, Portland, Oregon, March 2-6, 1997, pag 342.
2. SK.Yagnik et al, "Assessment of BWR fuel degradation by post-irradiation examinations and modelling in the defect code", Proceedings of the ANS, Portland, Oregon, March 2-6, 1997, pag 329
3. A.Hüttmann et al, Post-irradiation examination of failed KKK-barrier fuel rods", Proceedings of the ANS, Portland, Oregon, March 2-6, 1997, pag 350.
4. A.Seibold et al, "Fe-enhanced Zr liner cladding", Proceedings of the ANS, West Palm Beach, Florida, April 17-21, 1994, pag 337.
5. J.S.Armijo, "Performance of failed BWR fuel, Proceedings of the ANS, Portland, Oregon, March 2-6, 1997, pag 337.
6. G.Domizzi et al, "Blister growth in zirconium alloys: experimentation and modelling", J.Nucl. Mater. 229 (1996) 36-47.

FIGURE CAPTIONS

1. Scheme of the experimental set-up.
2. Artificial blister on a non-irradiated Fe-liner cladding.
3. Experimental set-up before introduction into the hot cell.
4. Typical appearance of the segment surface after blister formation (Periscope picture, 10x).
5. Metallography of a blister on irradiated Fe-liner material.

Artificial Blister

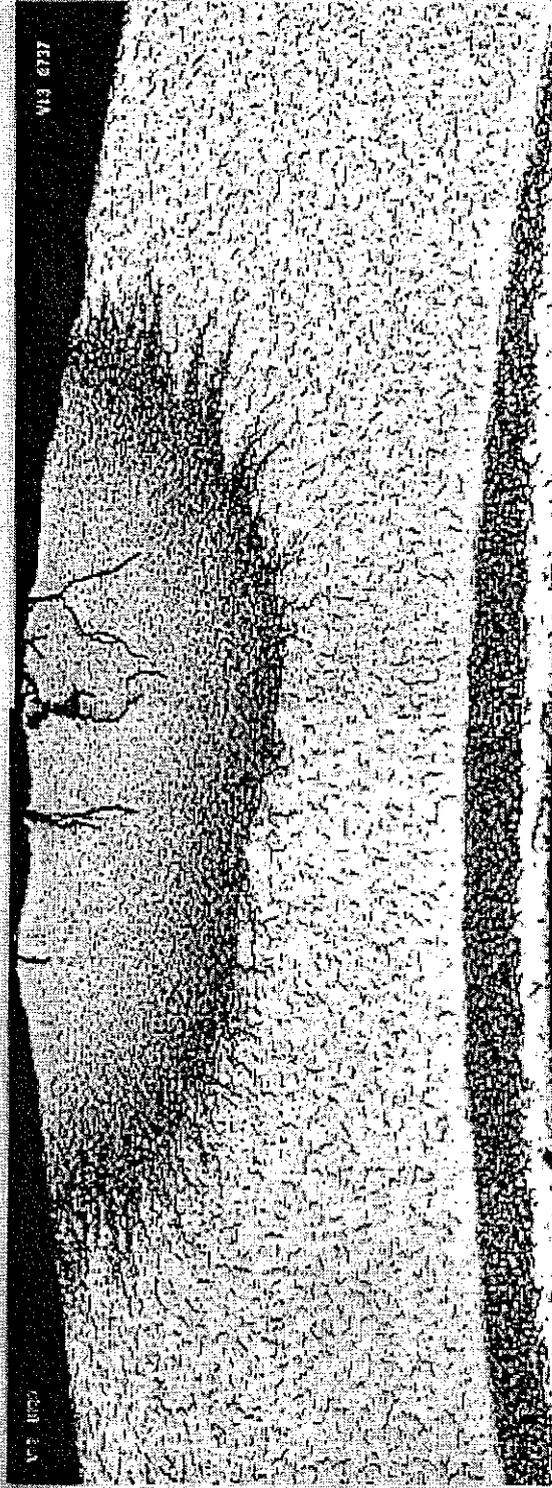


Experimental Set-Up

Figure 1



**Artificial blister
(Pretest for liner-cladding degradation experiment in Halden)**



Test No. 13, Test conditions: Ni scratched, 400 °C, 20 h,
cross section

Figure 2

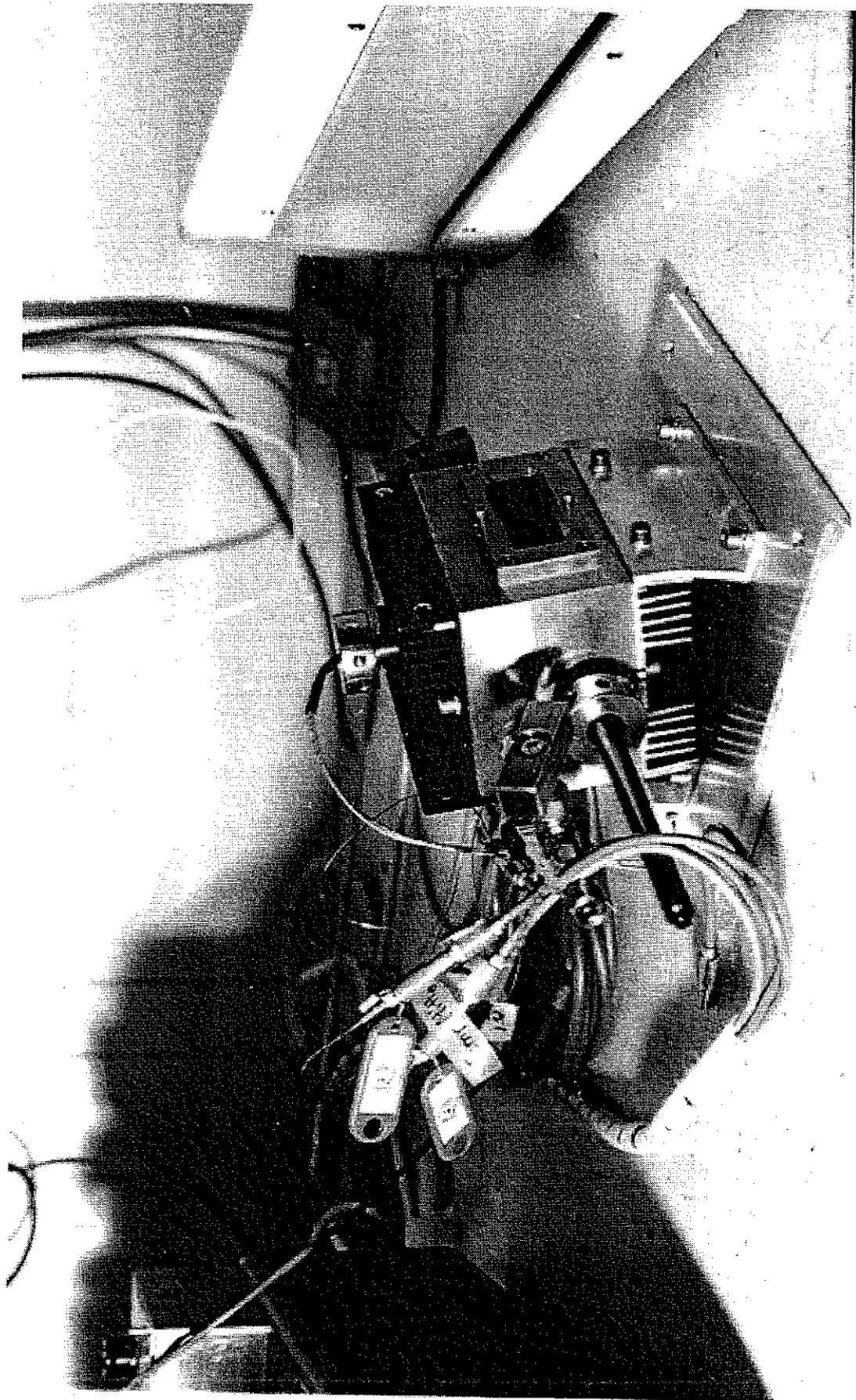


Figure 3

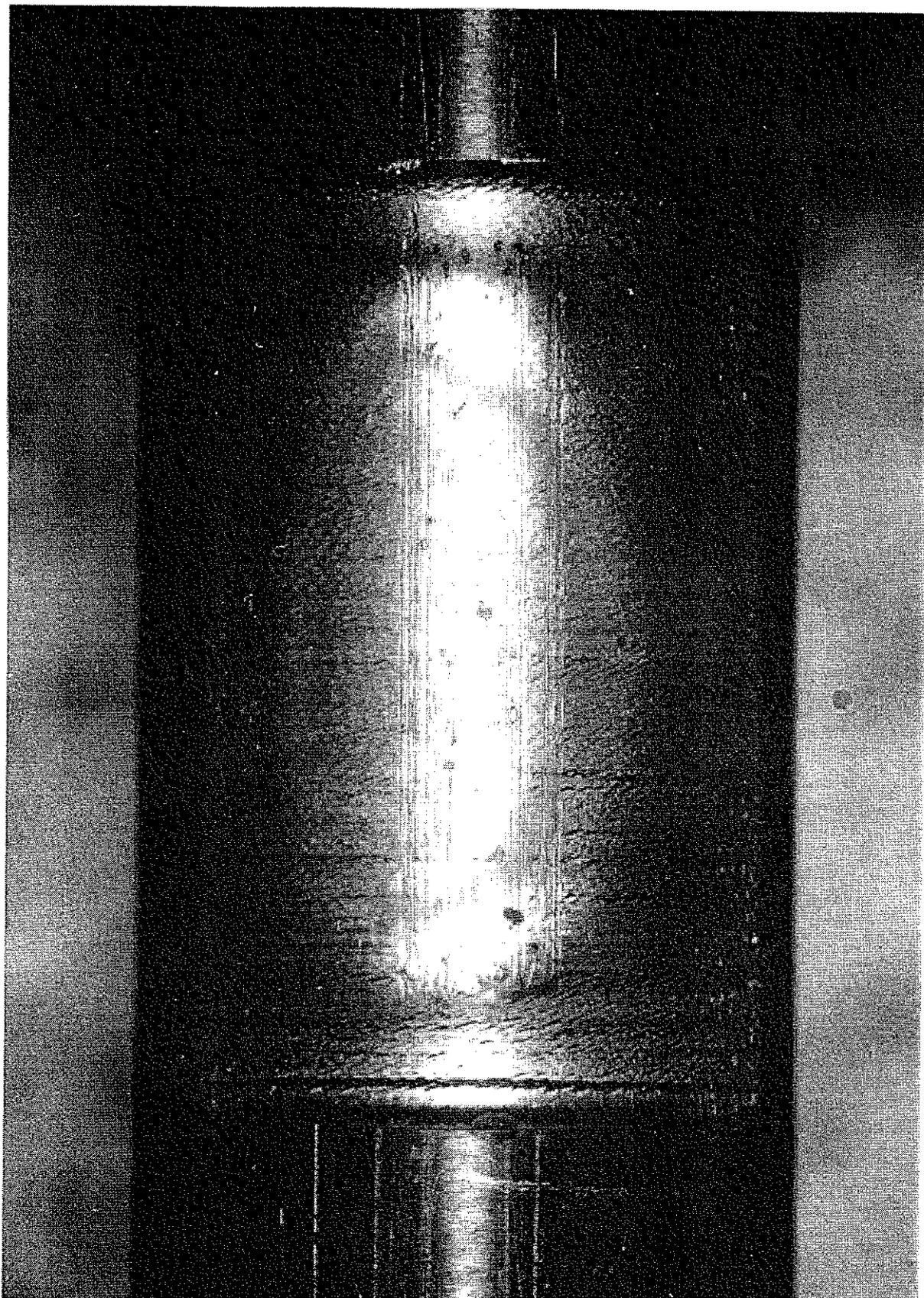


Figure 4

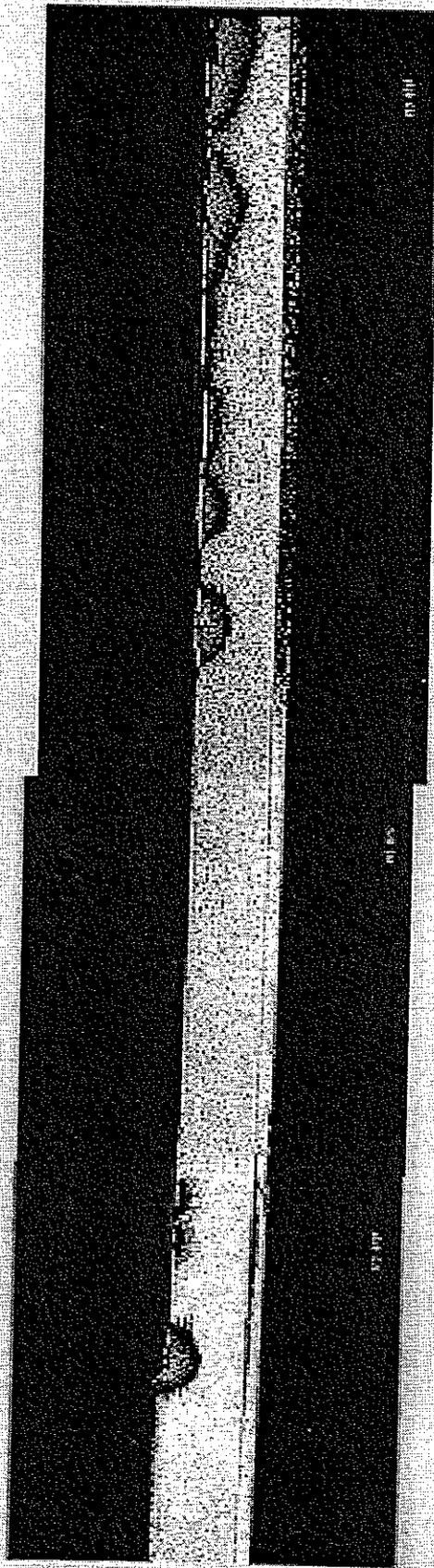


Figure 5

