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피복관재료의 노내 시험

Inpile (in PWR) Testing of Cladding Materials

한 국 원 자 력 연 구 소

30-48

제 출 문

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본 보고서를 “피복관재료의 노내 시험” 관련 기술현황 분석보고서로 제출합니다.

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요 약 문

Ⅰ. 제 목 : 피복관 재료의 노내 시험

Ⅱ. 연구내용

먼저 노내 시험이 필요한 이유를 설명하였다. 고연소도 피복관으로 사용하기 위한 새로운 합금 개발에 필수적인 노내 시험과정이 전반적으로 설명되었고, sample geometry와 노내 부식 시험의 측정 기술을 상세하게 기술하였다. 피복관의 크립과 길이 변화 거동 시험이 간략하게 논의되었다.

최종적으로 향후 시험에 대한 문헌 조사 경향을 보고하였다.

SUMMARY

As an introduction, the reasons to perform inpile tests are depicted. An overview over general inpile test procedure is given, and test details which are necessary for the development of new alloys for high burnup claddings, like sample geometries and measuring techniques for inpile corrosion testing, are described in detail. Tests for the creep and length change behavior of cladding tubes are described briefly.

Finally, conclusions are drawn and literature citations for further test details are given.

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1. Introduction : Why inpile Testing

By irradiation, material properties such as point defect density, precipitate size and composition, or oxide microstructure are changed. That's why irradiated material or material under irradiation has properties different to unirradiated materials: hardness, ductility, creep resistance, or corrosion resistance is different. The differences in corrosion behavior are schematically shown in Fig. 1.
Some other material behavior is peculiar to irradiation, like growth, for example, and its characteristics can be determined only when the material has been irradiated.
To get an impression of the inpile behavior, and to get data which are necessary for an assessment of the behavior of nuclear materials, inpile tests have to be performed.
For some properties of some alloys for cladding material, there exists a clear and in the meantime well-established correlation between inpile and outpile behavior. For Zry and similar alloys, for example, the inpile corrosion rate can be predicted if the 350℃ outpile corrosion rate is known, so that for these alloys inpile testing is no longer mandatory. But when one leaves this group of alloys, one leaves these correlations too, as shown in Fig. 2 and 3, and with outpile test results only, no prediction on their inpile behavior is possible. For example, at present no generally accepted outpile test procedure is available which predicts the inpile corrosion rate of ZIRLO-type alloys.

Inpile measurements can be made in test reactors -they are built for this purpose- or in commercial (power) reactors. Test reactors have the advantage of offering clear and well-defined test conditions, and that's why they are advantageous for basic research. But for gaining a more technical or application-oriented view -which should prevail in alloy development programs for high corrosion resistant materials- power reactors have many advantages over test reactors. These advantages are summarized in Tab. 1.

2. General Test Procedure

Generally, inpile test procedure consists of the following four steps:

- Pre-characterization of the test samples
- Intermediate non-destructive inspection of the samples
- Destructive or non-destructive post-irradiation examination (PIE)
- Determination of the irradiation test conditions

This report relates to inpile test methods which have to be applied when one develops new cladding materials. There are some other inpile test procedures which are usually performed, like determination of spacer spring relaxation or of spacer grid dimensions. They will not be described here, since they are beyond the scope of this report.

In the first step, (parallel) outpile testing and characterization of material preferably from the same lot and within the same temperature (range) as in reactor should be done in order to evaluate irradiation effects and to separate them from temperature effects. It is also a means to assure that the material to be tested inpile is actually the material in question.
Since it is not economic to determine too many properties, enough archive material should be kept on store to reevaluate specific properties after the inpile test results are known, and from where perhaps new questions arose.
Since inpile testing is expensive and time consuming, clear and unambiguous markings of the samples, descriptions and documentation should be made to avoid misinterpretations, confusion, or even wrong conclusions and decisions later on.
The second and third steps concern the inpile testing and measuring procedures themselfs.
In the second step, intermediate inspections and measurements are performed. Since they are intermediate measurements, they have to be non-destructive. They may comprise visual inspections of the integrity of the sample (fuel rod)

or the appearance of the oxide scale. Due to the radiation level, they have to be done remotely with a TV camera, as sketched in Fig. 4, and should be recorded for later evaluation, since during reactor shut-down, time is often too short to look at all details. Intermediate inspection normally also comprises defect testing and oxide thickness determination via Eddy current (EC), dimensional measurements as the diameter changes with "Linear Variable Differential Transformers" (LVDT) to determine the inpile creep behavior, and length change measurements with a "Vernier Calliper" for growth characterization.

If the samples are in fuel rod positions, integrated into a fuel assembly, the above mentioned inspections can be made on the outside rods only.

Measurements in steps two and three, as well as sample types and the accuracy of the measurements, are described in more detail in chapter 3.

Second step-measurements can be made several times, at each reactor shut-down. Typical fuel inspection activities are listed in Tab. 2

The third step is called post irradiation examination (PIE) and consists of non-destructive testing (NDT), and of destructive measurements with their inherently higher accuracy. The samples are transferred from the power plant to hot cells. There, metallographic determination of oxide scale and hydride distribution may be made, as well as characterization of precipitates with TEM/SEM, or hydrogen content can be determined with standard methods like hot extraction. Post pile properties like irradiation or hydrogen embrittlement can be determined by standard mechanical test procedures.

Step four serves to determine the inpile test conditions. Test temperature is determined via the known power history of the reactor and by taking into account the geometrical position of the samples within the core of the reactor. Typical axial temperature and neutron flux profiles for PWR and BWR are shown in Fig. 5. From them, the fluence which the samples have experienced during their residence time can be calculated. The latter can be determined in a more direct way with so-called measuring lances, or with 'fluence monitors" positioned near the samples, or by gamma scanning of the samples themselves. Gamma scanning is performed with Germanium detectors.

The measurement of short-lived isotopes provides information about power distribution at the end of irradiation, whereas measurements of long-lived isotopes are used to determine the relative burnup along the axial position (burnup profile), Fig. 6.
Agreed-upon, recommended (e.g. by IAEA) and standardized (e.g. by ASTM) test procedures exist. They are beyond the scope of this report.

3. Procedures for Corrosion Testing

3.1 Sample Geometry

Corrosion tests could be performed as coupon tests where the samples are of sheet material, or as material test rods (MTR) with non-fueled tubes, or as pathfinder rods /pathfinder fuel assemblies where the tubes are fueled.

Coupon tests have the advantage that a great variety of different alloys can be tested, since the samples are small. The sheet samples are comparably cheap since they can be made from small (button) melts, which are deformed by simple rolls. But since it is not possible to apply the realistic thermo-mechanical treatment (i.e. the treatment necessary for tube production) to sheet material, coupon tests can be used as screening tests only. Especially, it is not possible to impose a representative state and degree of stress/strain to sheet material. Secondly, there is a comparably high proportion of "edge and corner corrosion", and thirdly, there is no heat flux through the samples to detect the effect of corrosion rate acceleration due to the temperature increase caused by the growth of oxide scale.

Instead of sheet material, 120-deg. tube segments can be tested as well. They have the advantage of having experienced proper thermo-mechanical treatment.

In MTRs, there is also no heat flux, but their thermo-mechanical treatment is representative. If they are filled with swelling mandrels of different chemical compositions, the effect of strain rate on time- or strain-to-failure can be studied. MTRs are also used when there are doubts or objections regarding reactor safety, which prohibits any testing as fueled rods.

Pathfinder rods and pathfinder fuel assemblies are the last test sample geometry before announcing the new alloy or design as "commercial".

3.1.1 Coupons (Sheet Material) and Tube Segments

Typical sample sizes are 8 mm * 20 mm. In order not to loose them by forces imposed by the streaming coolant water, they have to be "imprisoned" in tubes[1]. For size reasons it is advisable to put them into empty control rod

[1] That has to be taken very seriously. Even those small samples would cause severe "debris fretting", the

guide tubes which normally have larger inner diameters than fuel rods (~ 12 mm). For free exchange of the corrosion medium (=reactor water), holes should be drilled through the tube walls. Within the tubes, the sheet samples are fixed together to bundles of 5 to 10 sheets, the bundles themselves stacked one over the other. To avoid contact corrosion effects, they can be separated from each other by small ceramic "spacer" rings, as shown in Fig. 7. For more convenience during intermediate inspections, they can be clamped together. For the same reasons, it may be reasonable not to use full length guide tubes, but tubes which are axially segmented, with the segments screwed together. The screw should be secured by spot welds.

3.1.2 Material Test Rods (MTR)

Material test rods are positioned in empty control rod guide tube positions, as in Fig. 8. Since they have no UO_2 pellets inside, they are light. In order to prevent them from swimming up by the coolant water stream, they have to be axially secured by special hold down devices as shown in Fig. 9.

If they are filled with swelling mandrels for determining their strain to failure behavior, (see chapter 4), this device is possibly not necessary.

To have space for more test species, the MTRs can be segmented as described above, with each segment consisting of different alloys or made by different manufacturing routines.

3.1.3 Pathfinder Rods

They are inserted into normal fuel rod positions. For inspection reasons, they should be put into the outside positions of the fuel assembly. The pellets must have an enrichment, porosity, grain size and geometry which is representative of high burnup fuel, to assure reasonable heat flow, shrinkage and swelling, fission gas release and ridging.

To get an impression of the safety margins of the pathfinder tubes, some tubes should be fueled with "higher demanding" pellets as well, imposing, for example, higher heat fluxes to the tubes than the heat fluxes envisaged for

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main cause for core damage within the last few years.

"high burnup fuel claddings". For safety reasons, these higher demanding conditions should be imposed one cycle later than the "normal" high burnup conditions.

3.2 Measuring Procedures

3.2.1 Intermediate Inspections

The visual appearance of the oxide, such as color and homogeneity, (spalling offs, flaws, scratches, crud deposits), is assessed from pictures which are taken with a remote TV camera. The pictures have to be recorded together with the position they are taken from.

Since visual examination is subjective, visual (color) standards are helpful for the interpretation of the pictures.

Visual inspections should be made by experienced inspection engineers.

For both sheet and tube materials, integrity and oxide thickness are determined by Eddy current (EC) methods, which are based on the differences in the (complex) electric conductivity of oxide and metal. (For integrity determination, different frequencies have to be applied than those used for oxide thickness determination). A sketch of an EC measuring device is shown in Fig. 10. Instead of EC, interference/diffraction methods using infrared light are available for oxide thickness determinations today too.

As for all non-destructive test procedures, the EC method has to be calibrated by standards. For oxide thickness measurement, Zry tubes or Zry sheet material of the same geometry (diameter, wall thickness) with well-known layer thickness are used. For integrity determination, tubes with well-defined failures (depths of notches) are used as standards.

An example of an EC trace from defective tubes is given in Fig. 11.

Since the EC data are dependent on the distance between coil and oxide surface as well as on the angle between coil axis and sheet normal, the results depend on the skill and experience of the staff. It should be trained at dummy fuel assemblies outside the reactor.

"EC traces" can be made "linear" along sample length, or spiral shaped. For the latter, the tubes have to be rotated with respect to the measuring coil.

Typical traces are shown in Fig. 10.

For hydrogen determination, to my knowledge no reliable non-destructive test method exists up to now, even though much research was done, eg for determination of H content by use of ultrasonic methods.

3.2.2 Final (Post Irradiation) Examinations (PIE)

PIE may consist of nondestructive testing as described in chapter 3.2.1. PIE are made in the spent fuel pool. They are the basis for the selection for the more detailed destructive measurements like metallography and chemical analysis, which have normally to be performed in hot cells and which therefore are more expensive and time consuming, but they are more exact. Fig. 13 shows a comparison between NDT and metallografic results. One can see, that by NDT, an accuracy of =/- 5㎛ can be achieved.

Very small specimens, like those who are necessary for TEM/SEM inspections for oxide structure or precipitate characterization, can be examined outside hot cells if their radiation level is low enough. It is low enough for sample weights of several milligrams. If larger samples have to be examined by TEM/SEM, sometimes replica techniques can be applied.

To reduce the level of radiation of pathfinder tubes, it is advisable to remove all fuel from the cladding inside after arrival in hot cells. That means that the rods have to be cut by a saw inside the cell, the fuel has to be forced out the tube segment mechanically, and the fuel particles sticking to the inner surface of the tube have to be wiped out.

4. Procedures for Creep Testing

To determine inpile creep behavior, the rod diameter has to be measured. For intermediate as well as post irradiation examinations, this is done by a "Linear Variable Differential/Displacement Transducer (LVDT), Fig.14. The LCDT has to be is calibrated with standards. These standards consist of a set of tubes with different outside diameters.

The stress to cause creep is imposed by internally pressurizing the tube segments. A drawing of a creep sample is given in Fig. 15.

To determine strain-to-failure of different cladding materials, certain defined strain rates can be imposed to the cladding by the use of swelling mandrels. A swelling mandrel is a pellet made from materials which swell under the action of neutrons. Typical materials are mixtures of sintered Al_2O_3+B_4C powders, Fig. 16. With higher B_4C content, higher swelling rates under neutron irradiation can be achieved. Other swelling material (with low swelling rates) is the PWR control rod material AgInCd.

5. Procedures for Growth and Length Change Testing

Length change of fuel rods consist of two parts: of irradiation induced growth and of creep (for anisotropic structures)

In order to determine the pure irradiation induced growth component itself, material test rods (without fuel, and which are open to assure that no stress acts onto the tube wall) are positioned adjacent to standard fuel rods. Special hold down device has to be applied to prevent the tubes from swimming up due to water coolant.

Length of the rods is measured with a vernier calliper-type device.

For the determination of the length change due to anisotropic creep, which is normally 10% of the total length change, the material test rods are prepressurized with different pressures. Subtracting the known irradiation growth, the creep portion of the total length change at the given pressure/stress can be calculated.

To determine the total length change of fuel rods, the same vernier calliper is used. Since the irradiated fuel rods are at higher temperatures even after a long time after their removal from reactor, the length change due to thermal expansion has to be subtracted by calculation.

6. Conclusions

Inpile testing has to be performed, especially for alloys far away from Zry ASTM range, for fore them no generally accepted correlations between inpile and outpile behavior exist. Also from safety and licensing aspects these tests are mandatory, before an alloy can become a "commercial alloy".
Since they are time consuming, they should be started as early as possible.
Since they are expensive, great care has to be taken to test only "reasonable" alloys. That is in conflict/contrast to the precedent sentence. A compromise between both aspects of inpile testing has to be found.

7. Literature

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Tab. 1 Advantage of In-PWR Measurements over Test Reactor Measurements

1. Realistic operational conditions: actual n-flux, n-spectrum, coolant flow, water chemistry, temperature, pressure, long term (>1*E22 cm^{-2}, several years test duration)

2. Large number of specimens

3. (in Germany normally lower costs per sample, depending on agreement with plant operator)

Measurements

1. Precharacterization of the Test Samples: Parallel outpile testing of the same properties at the same temperature (-range) to evaluate/separate the irradiation effects. (Leave enough material from the same lot in archive for later-on examinations. Make clear, unambiguous markings, descriptions and documentation of the test samples)

2. Intermediate non-destructive Inspection in Spent Fuel Pool: Visual inspection with TV camera, EC-measurements of oxide thickness and of integrity, dimensional measurements of diameter (LVDT) and length (vernier calliper).(Those measurements have to be planned very precisely, with back-up solutions to be considered in advance, since sometimes the refueling shut-downs are short, and other, more important things have to be done with priority)

3. Destructive Post-Irradiation Examination (PIE) in Hot Cells: Characterization of the "post-pile" material properties, like oxide thickness via metallography; H-content/ hydrogen distribution via hot extraction/ metallography; precipitate characterization via TEM/SEM

4. Determination of the Irradiation Conditions: Temperature–(history), n-flux/fluence with measuring lances in the core, or calculated based on power history of the reactor, with additional verification (e.g. by evaluating "fluence monitors" positioned in or near test samples, or by gamma scanning of the test samples)

Tab. 2 Typical Intermediated Pool Inspections and Standard Pool Examination Technology for Surveillance of Fuel Assemblies

type of examinations	*typical extent*	*objectives*
sipping test	100 % (or all assemblies to be reloaded)	identification of leaking ass'ies (only if indicated by coolant activity)
visual inspection	10 – 30 %	mechanical integrity, wear, crud deposits rod–skeleton interaction (rod growth and bow)
special examinations, i. e. ● fuel rod oxide thickness ● length of ass'ies and rods ● bow of ass'ies and rods ● hold down spring force	2 – 5 assemblies ≤ 10 assemblies	provide performance data for design: ● fuel rod water side corrosion ● dimensional behaviour of fuel rods and skeleton ● spring relaxation

Continued.

EQUIPMENT	SCOPE
Sipping test equipment	- Leak-tightness of fuel rods
Underwater TV-camera with manipulator	- General appearance, integrity, wear, surface deposits (CRUD), debris, welded and screwed connections
Underwater TV-camera with accessories:	
- Ruler scale	- Axial clearance rod/skeleton, assembly bow (PWR)
	- Gap rod shoulder/upper tie-plate, distance channel/edge of foot piece (BWR)
- Tape measure	- Length of assembly and channel (BWR)
- Feeler gauge device	- Rod to rod distance
- Spacer measuring gauge	- Outer grid dimensions (PWR)
- Oxide thickness probe	- Oxide layer thickness of peripheral rods, channel, spacer grids *)
	- Wall thickness of spacer grids *) (PWR)
Guide thimble test device (PWR)	
- with depth gauge	- Length of assembly
- with eddy-current probe	- Guide tube wear
Force measuring device	- Hold-down spring characteristic (PWR)
Channel measuring device	- Profile of deflection and bow, twist, length (BWR)
Multi-pin gauge	- Rod length in the assembly (BWR)
Fuel rod exchange device with friction mandrel	- Spacer spring relaxation
Multiple measuring device for single fuel rods	
- LVDT-gauge	- Outer diameter profilometry
- Oxide thickness probe	- Oxide layer profilometry
- Eddy-current encircling coil	- Material inhomogeneities, metal-loss*)

*) under development

In-PWR(BWR) Examinations

Advantage of PWR Measurements over Test Reactor Measurements

1. Realistic operational conditions: actual n-flux, n-spectrum, coolant flow, water chemistry, temperature, pressure, long term (>1E22 cm-2)

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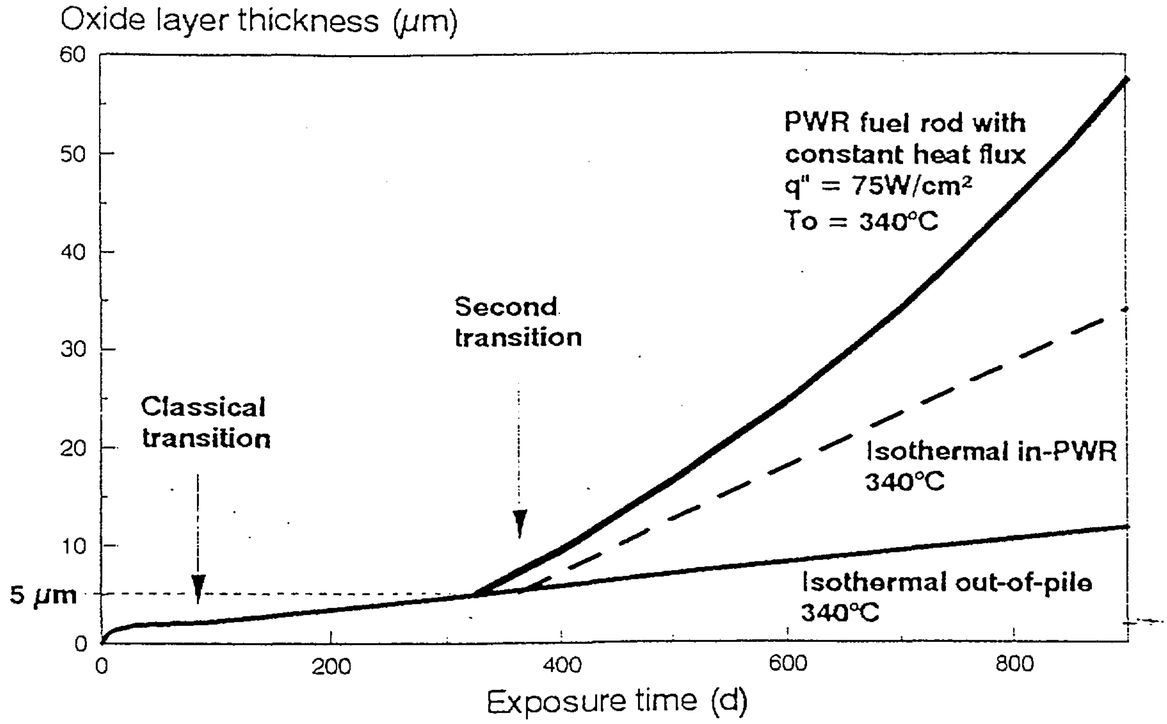


Fig. 1 Inpile and outpile Oxide Scale Growth

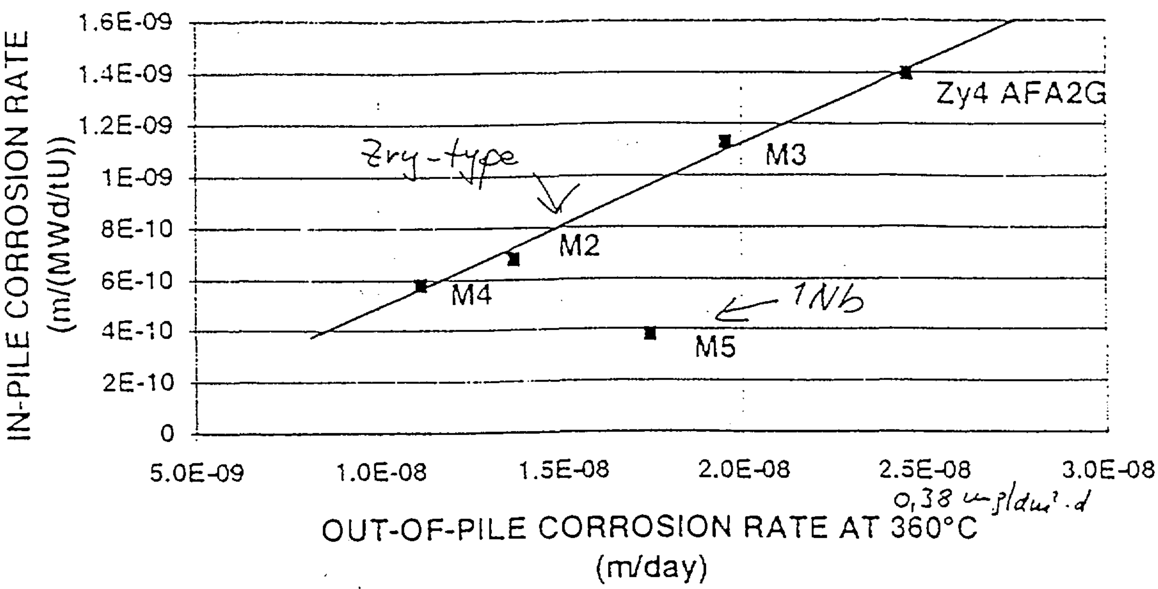
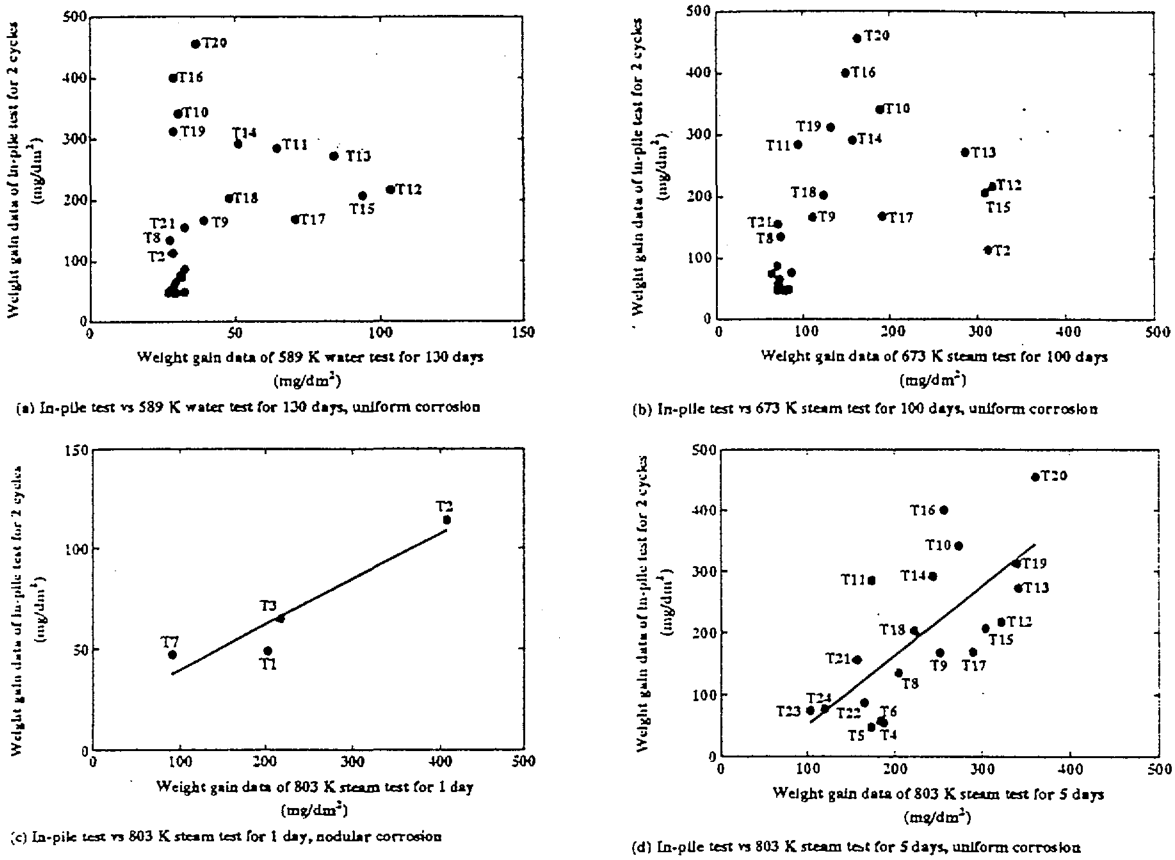


Fig. 2 Comparison of inpile and outpile Test Results



(a) In-pile test vs 589 K water test for 130 days, uniform corrosion

(b) In-pile test vs 673 K steam test for 100 days, uniform corrosion

(c) In-pile test vs 803 K steam test for 1 day, nodular corrosion

(d) In-pile test vs 803 K steam test for 5 days, uniform corrosion

Fig. 3 Comparison of inpile and outpile Test Results

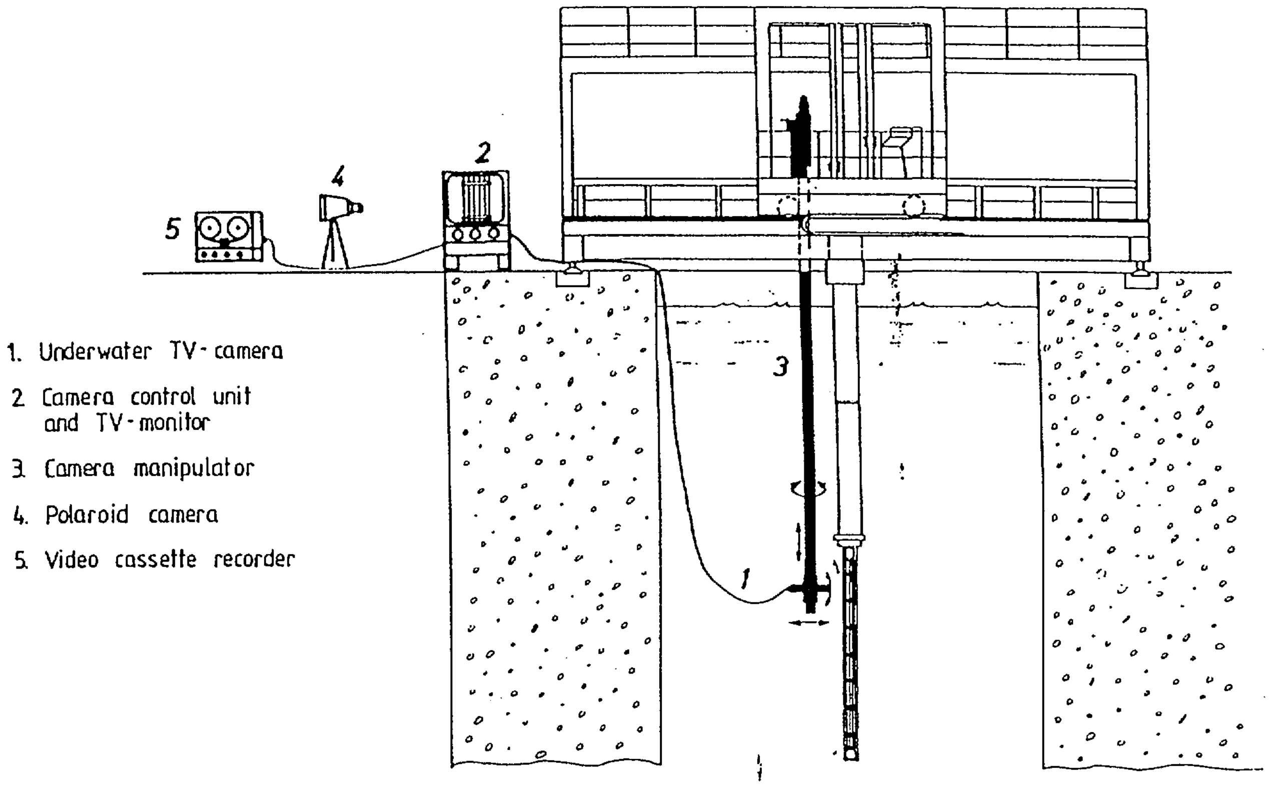


Fig. 4 Visual Inspection of Fuel Assemblies in a PWR Power Plant

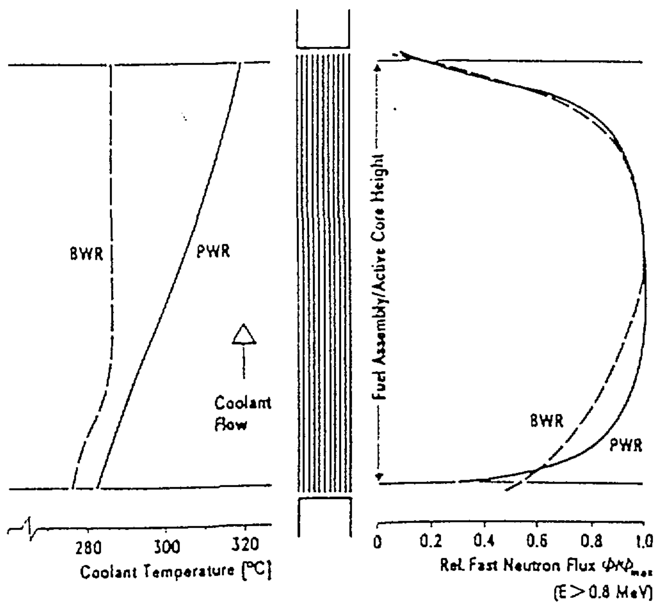


Fig. 5 Axial Temperature and n-Flux Profiles

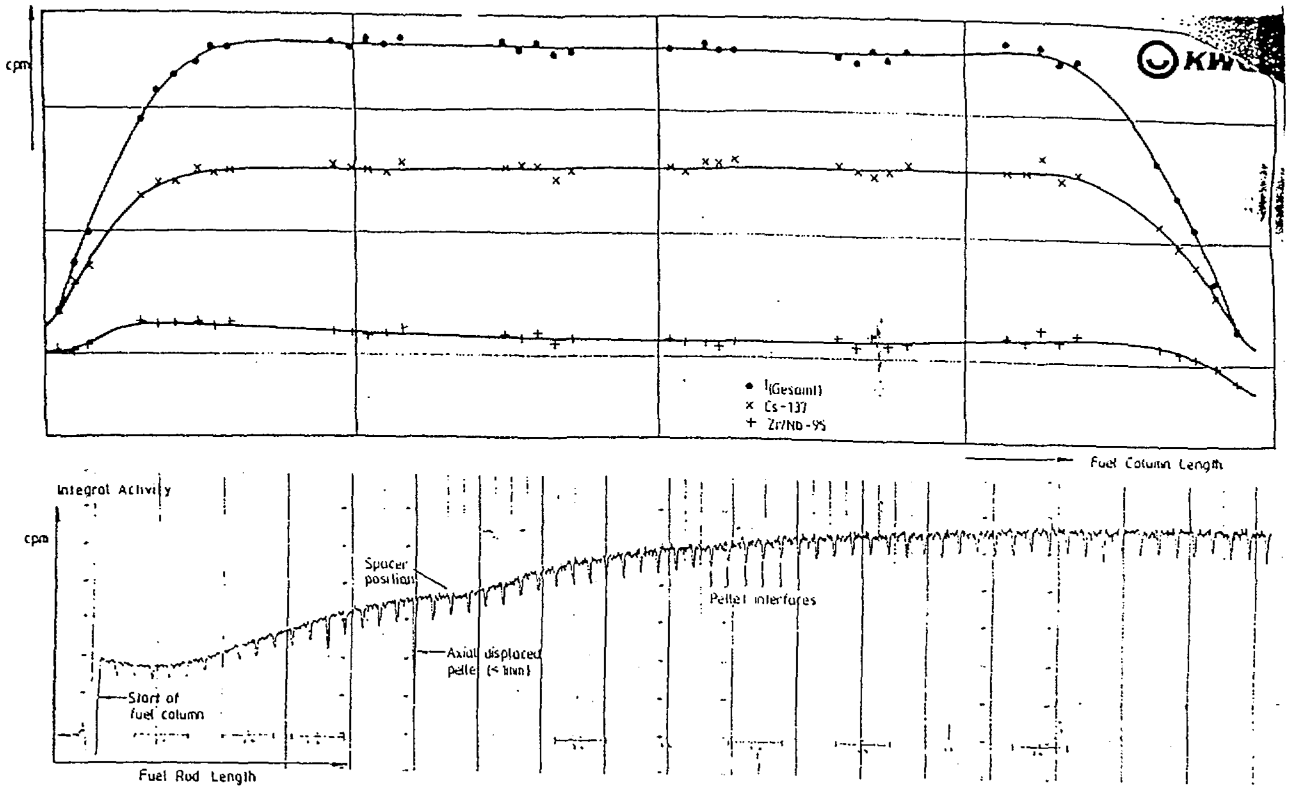


Fig. 6 Example of γ - Scan Data

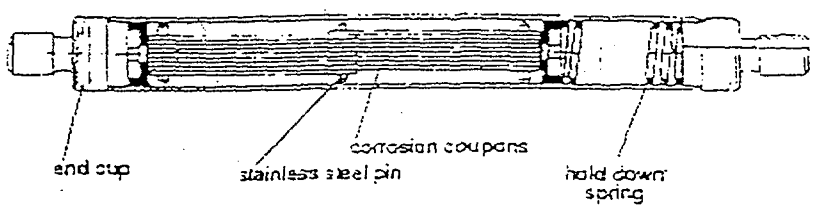
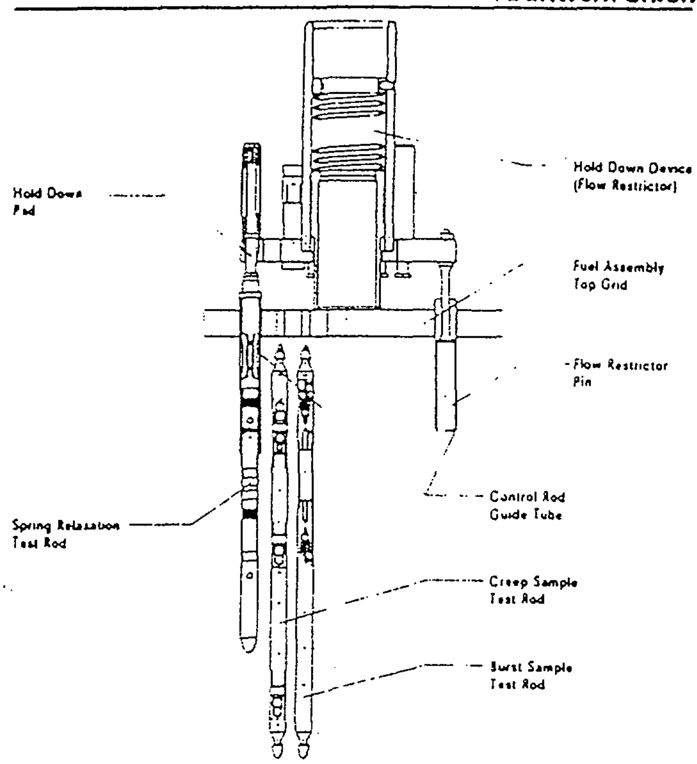


Fig. 7 Coupon Test Assembly in BWR Water Channel. The same Design is used for Control Rod Guide Tubes for in-PWR Testing

Kraftwerk Union

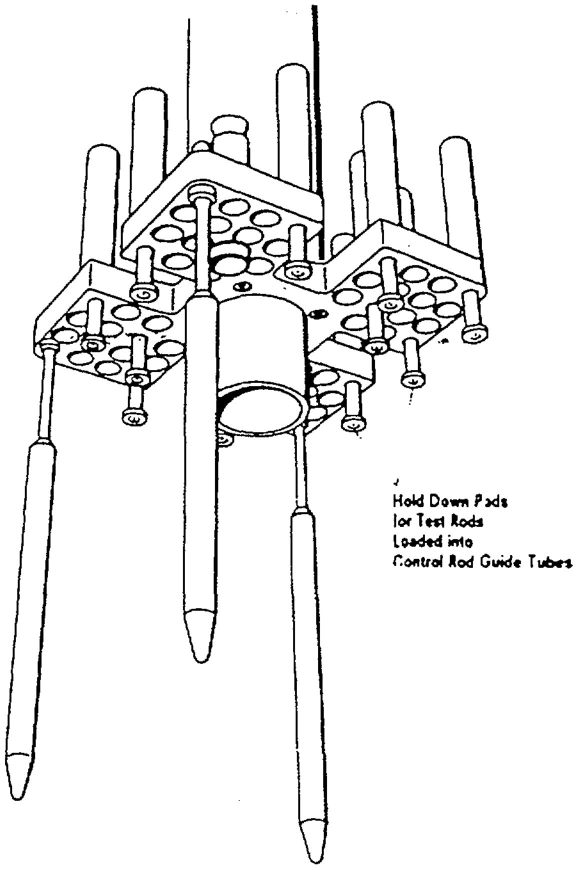


Schematic Diagrame of Material Test Rods
in a PWR Fuel Assembly

Fig. 5

Fig. 8 Material Test Rod (MTR) in PWR Fuel Assembly

Kraftwerk Union



Hold Down Device for Test Rods
in PWR Fuel Assembly Guide Tubes

Fig. 9 Hold down Device for MTR

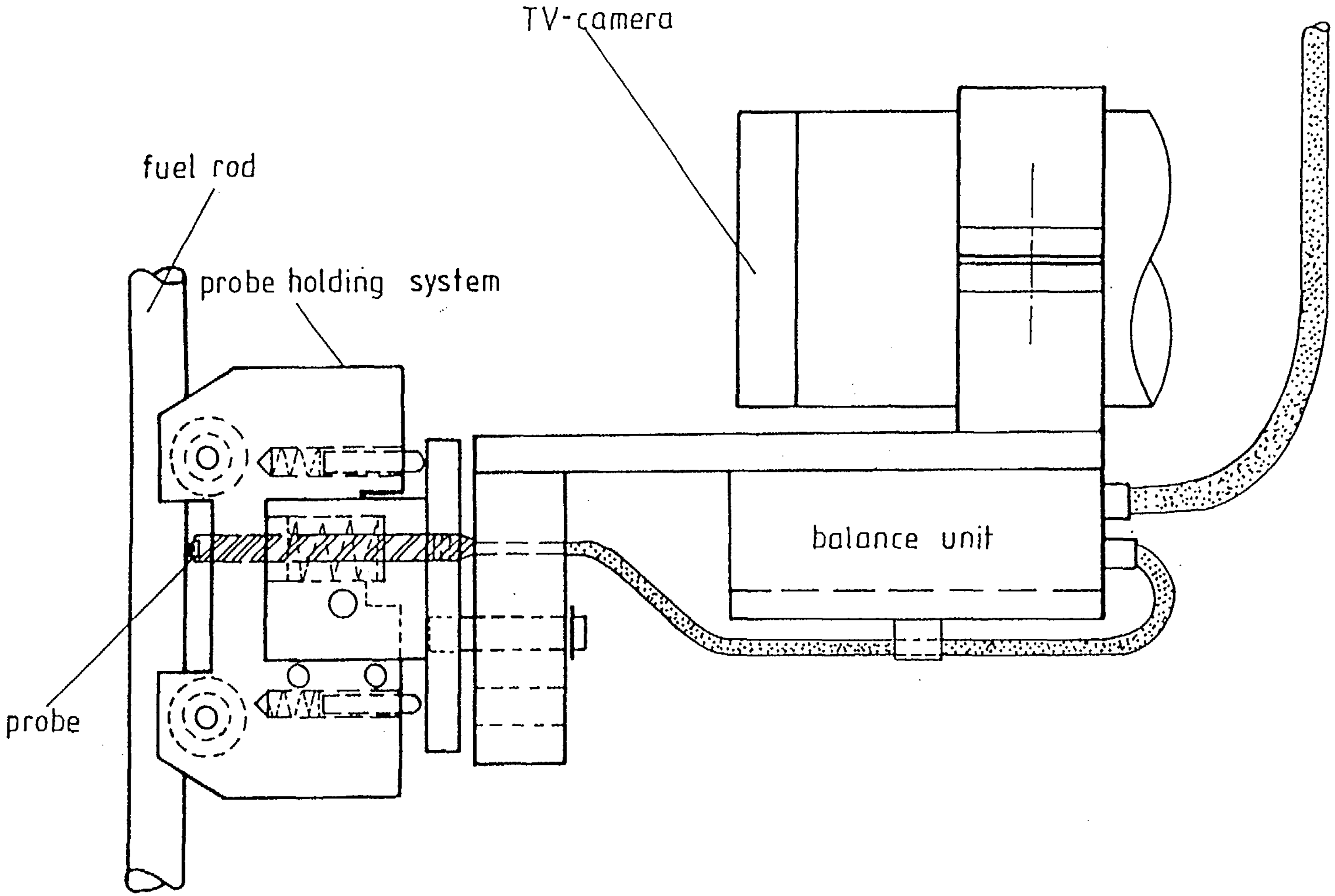


Fig. 10 EC Device

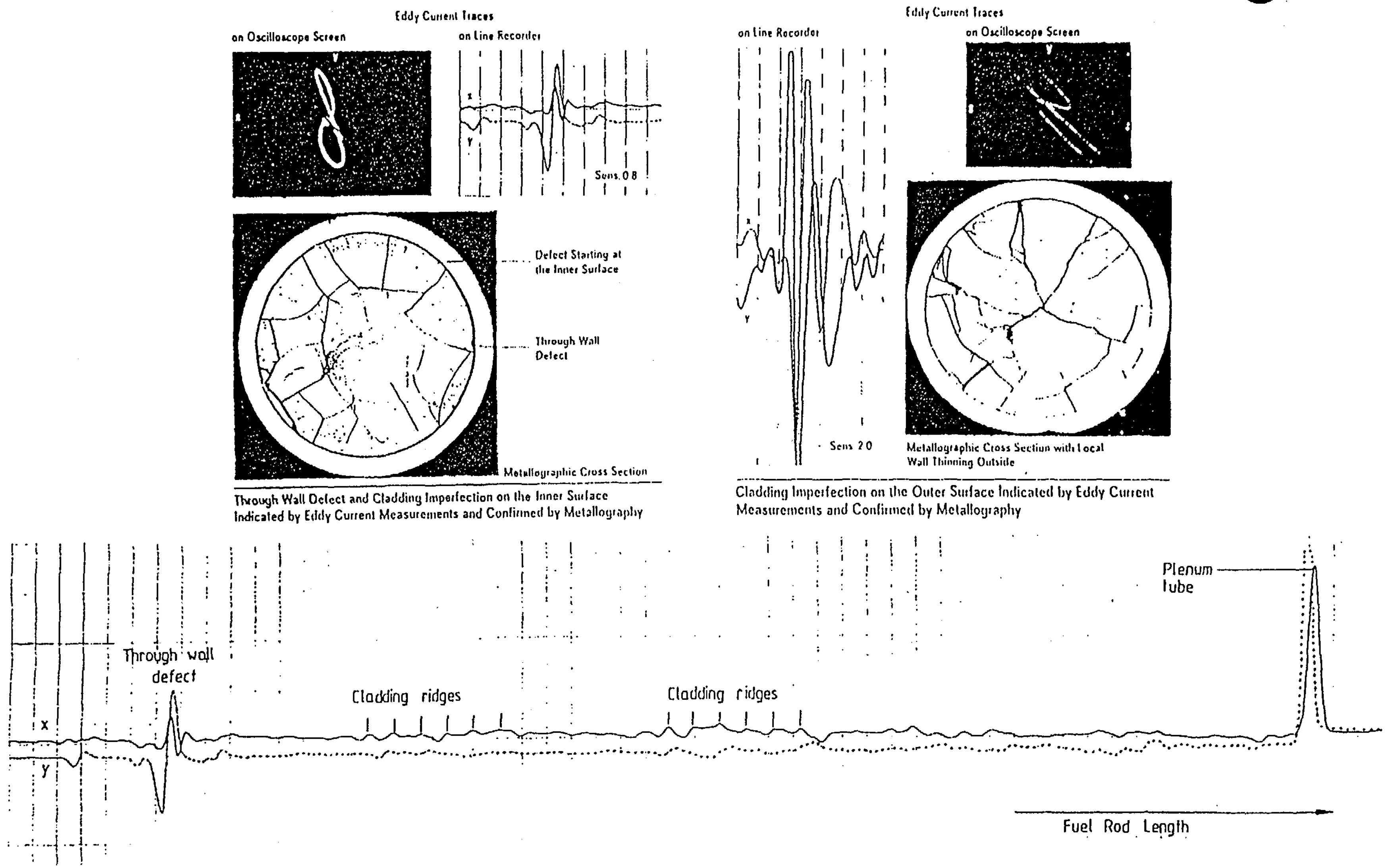
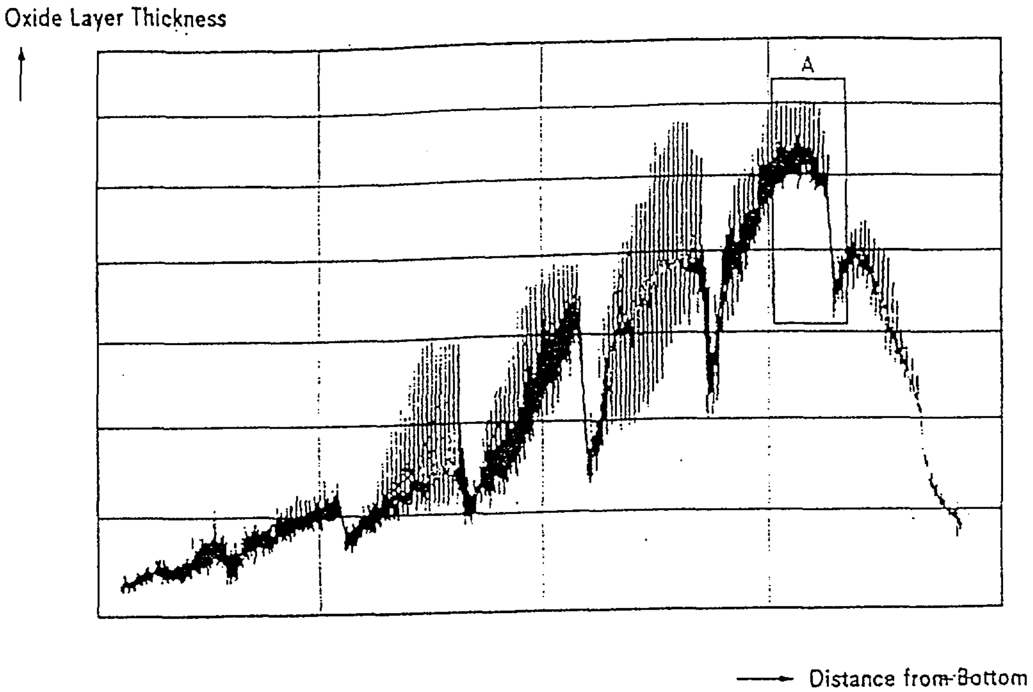
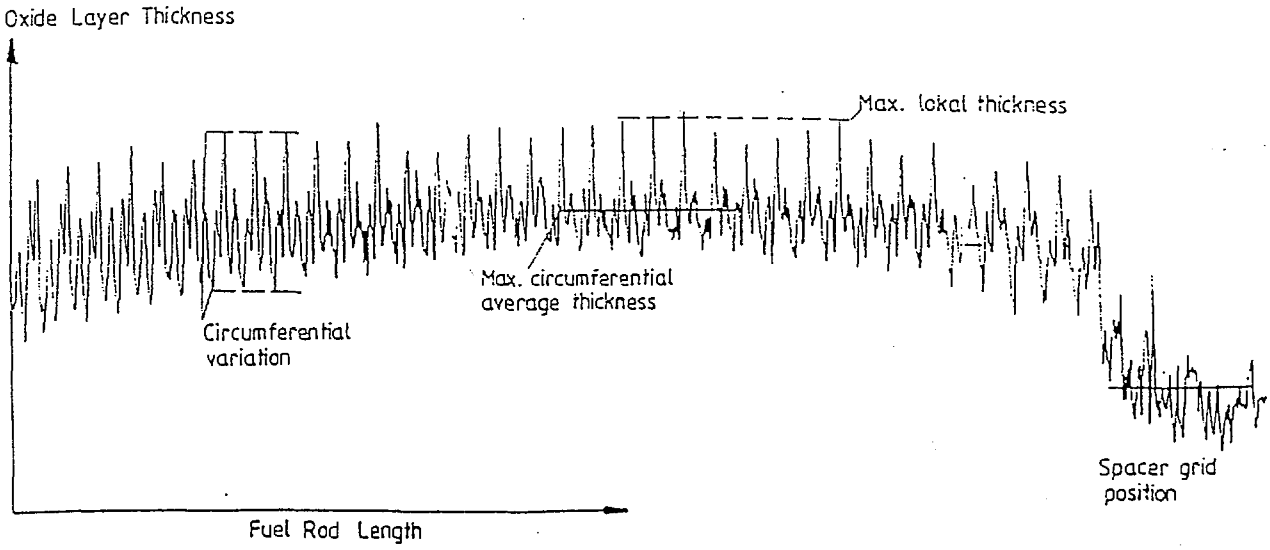


Fig. 11 EC Trace of a defective Fuel Rod



Typical Oxide Layer Thickness Profile (Spiral Trace) of a PWR Fuel Rod



Detail A: Spiral Oxide Thickness Trace

Fig. 12 EC Trace of an Oxide Layer Thickness Profile

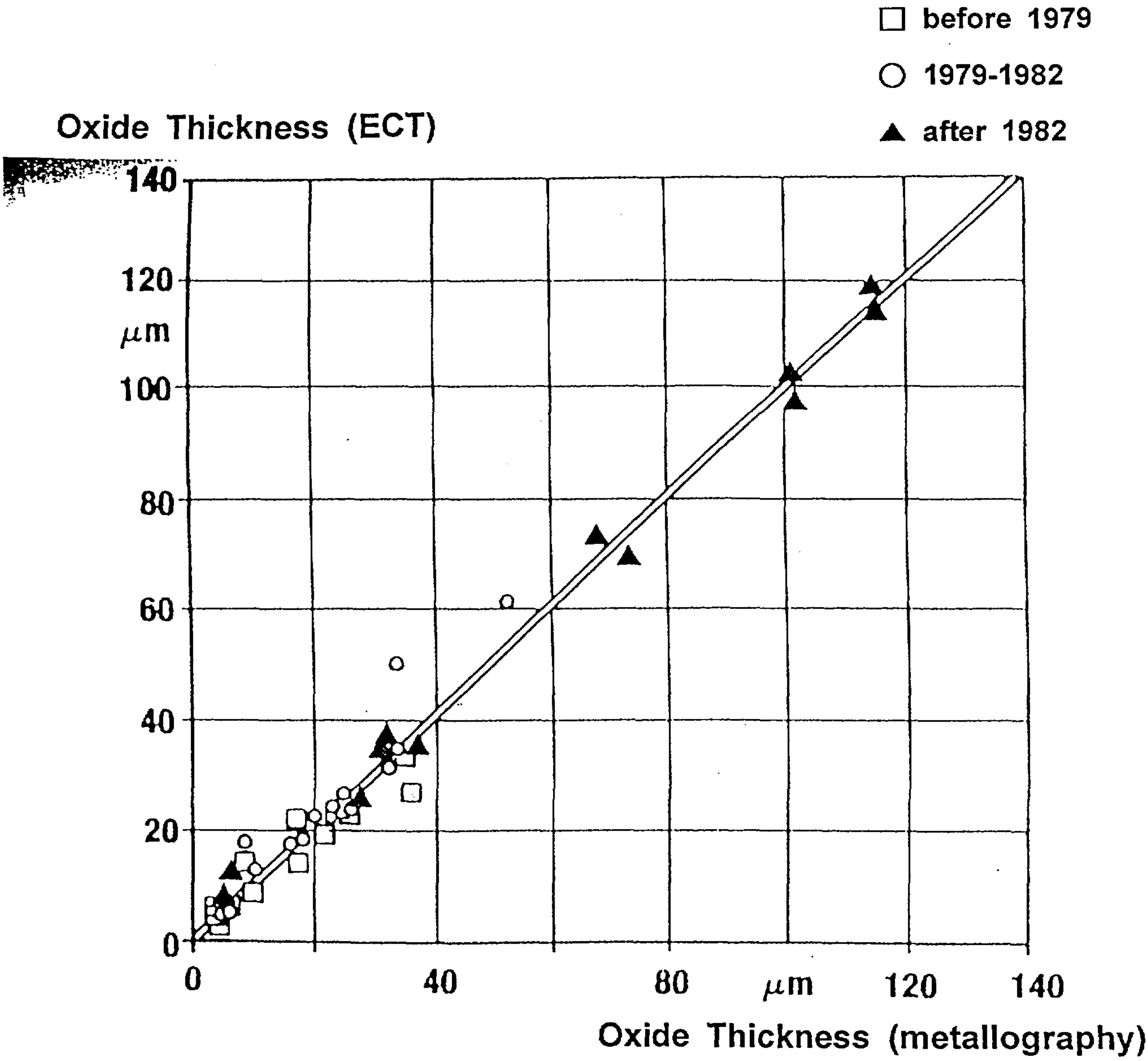


Fig. 13 Comparison of non-destructive testing and metallografic measuring Results

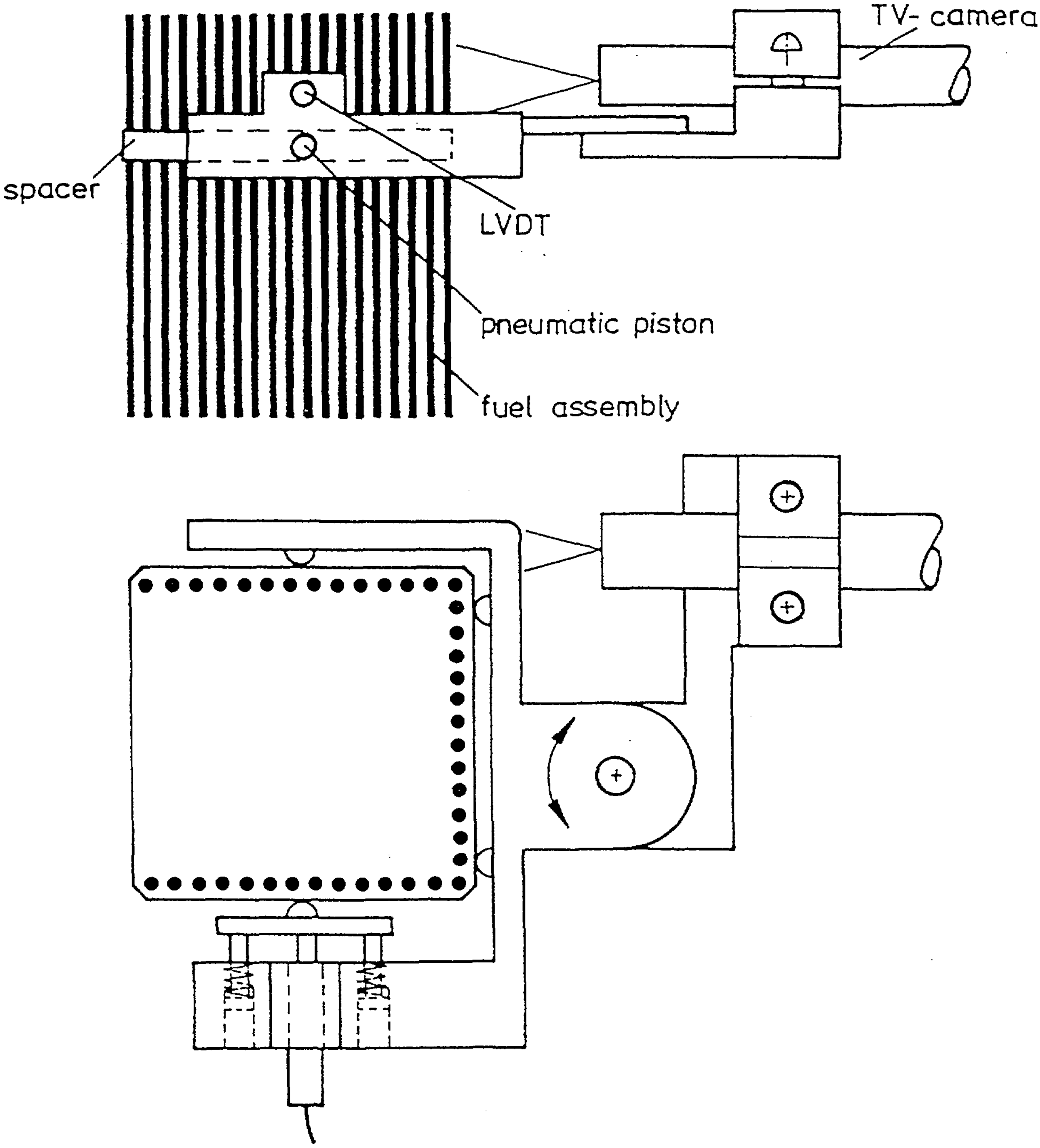
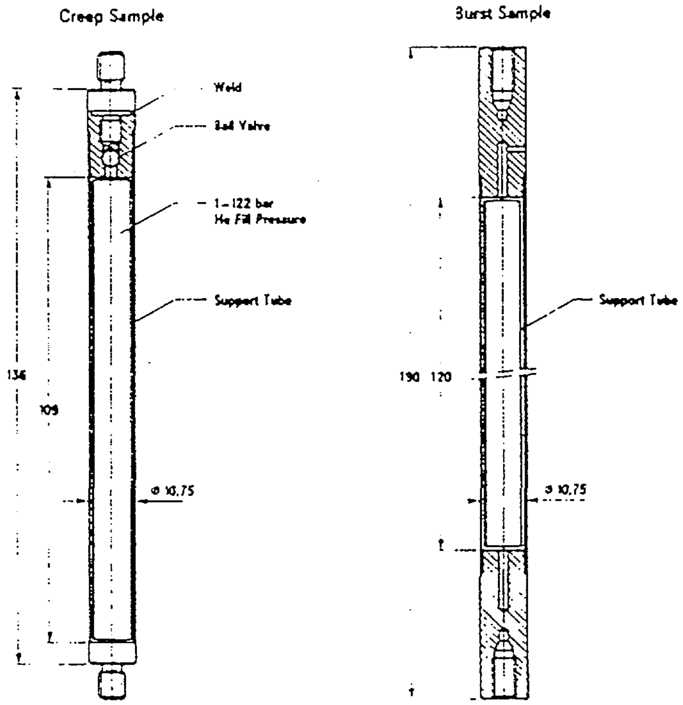


Fig. 14 Linear Variable Differential Transformer (LVDT) Device for dimensional Measurements

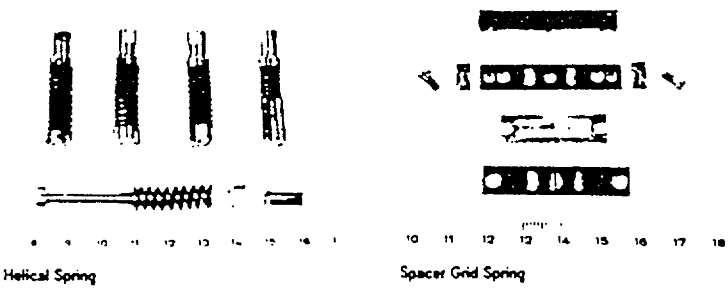
Kraftwerk Union



Creep and Burst Test Samples
of Zircaloy-4 Cladding Tubes in PWR

Fig. 7

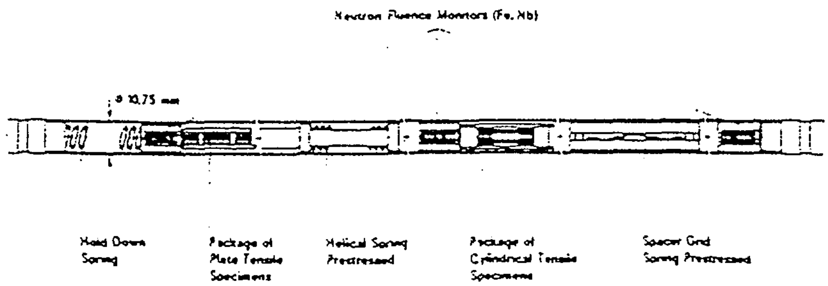
Kraftwerk Union



Spring Samples for Relaxation Testing

Fig. 15 Creep Sample

Kraftwerk Union



Material Specimens for Irradiation Testing in PWR

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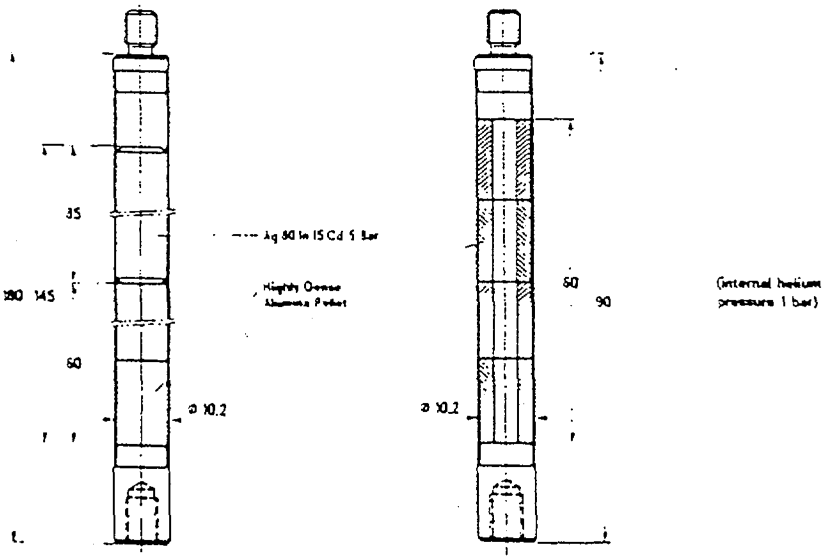

Irradiation Samples with Absorber Bar and Al_2O_3 Sintered Pellets

Fig. 16 Burst Sample to determine Time-to-Failure

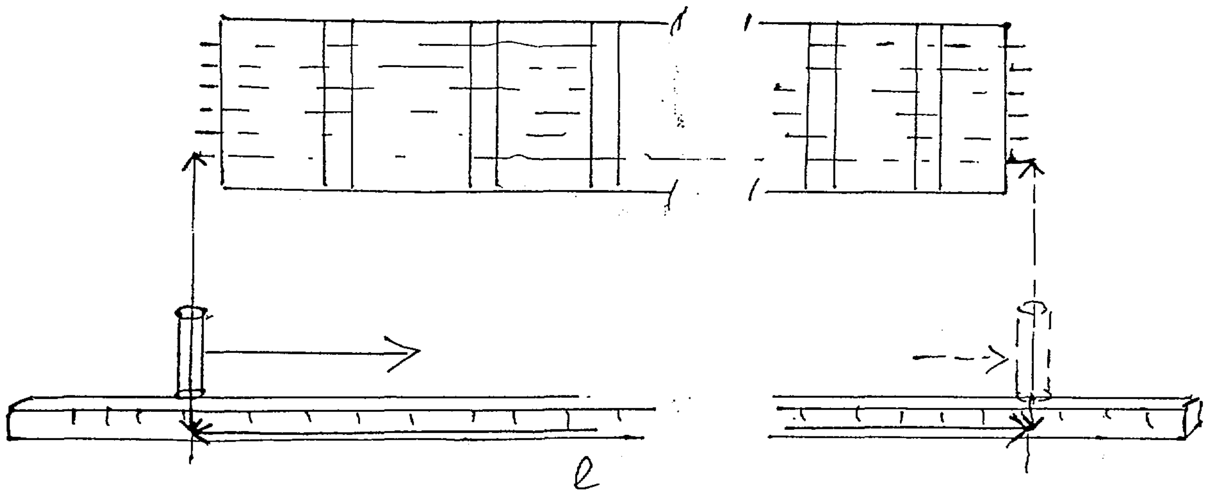

Fig. 17. Arrangement for the Length Change Determination of Fuel Assemblies

서 지 정 보 양 식

수행기관보고서번호	위탁기관보고서번호	표준보고서번호	INIS 주제코드
KAERI/AR-531/99			

제목 / 부제

피복관 재료의 노내 시험

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출 판 지		발행기관	한국 원자력 연구소	발행년	1999년
페 이 지	37 p.	도 표	있음(○), 없음( )	크 기	Cm.
참고사항					
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연구위탁기관			계약 번호		

초록 (15-20줄내외)

먼저 노내 시험의 이유를 설명하였다. 고연소도 피복관으로 사용하기 위한 새로운 합금 개발에 필수적인 노내 시험과정이 전반적으로 설명되었고, sample geometry와 노내 부식 시험의 측정 기술을 상세하게 기술하였다. 피복관의 크립과 길이 변화 거동 시험이 간략하게 논의되었다.

최종적으로 향후 시험에 대한 문헌 조사 경향을 보고하였다.

주제명키워드 (10단어내외)	Zr합금, 핵연료 피복관, 합금개발, 부식, 고연소도, 합금제조, 노내 시험, 크립

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Abstract (15-20 Lines)

As an introduction, the reasons to perform inpile tests are depicted. An overview over general inpile test procedure is given, and test details which are necessary for the development of new alloys for high burnup claddings, like sample geometries and measuring techniques for inpile corrosion testing, are described in detail. Tests for the creep and length change behavior of cladding tubes are described briefly.

Finally, conclusions are drawn and literature citations for further test details are given.

Subject Keywords (About 10 words)	Zr Alloy, Fuel Cladding, Alloy Development, Corrosion, High Burnup, Manufacturing, Inpile Test, Creep