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Summary

The PIE techniques at the IFE nuclear fuel section are improved through continuous upgrading of equipment and method during many years. E.g., preliminary testing of a solid nuclear-track detector, designed for recording ionizing particles and for utilization in neutron radiography, gives promising results compared to the classic method or the standard dysprosium foil/x-ray film technique.

The track-etch recorder consists of a 100 μm thick film of lightly rose-tinted cellulose nitrate coated on both sides with an energy converter (n, α) material. This method gives sharp and detailed images. The use of the cellulose nitrate film not only eliminates the prolonged handling of activated screens required by classical methods, but also enables much higher resolving power to be achieved.

Both methods allow unfiltered neutron beams to be used, and enables neutron radiography of very active samples such as irradiated fuel rods.

Solid nuclear-track detectors have numerous applications [2], for example the dosimetry of neutrons and of the heavier ionizing particles such as alpha particles, protons, fisson fragments, spallation fragments, heavy ions and very heavy ions such as cosmic rays etc.. In every instance the thickness of the cellulose-nitrate used should be in accordance with the mass and energy of the type of particle to be detected in order to obtain the optimum level of energy transfer.

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"Neutron radiography of irradiated fuel rods – an approach to improve spatial resolution in neutron radiographs".

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Abstract

A dysprosium foil/X-ray film technique is normally applied under neutron radiography inspection of irradiated fuel rods consisting of fuel pellets and zircaloy cladding materials. The main objective here is to reveal fuel rod failures, i.e. intrusion of moisture and detection of major in-homogeneity.

The primary source of neutrons is obtained from one of the radial channel of the Jeep II reactor (2 MW, central thermal flux 3×10^{13} n/cm^{2}s) at the Institute for Energy Technology, Kjeller.

To improve the quality of the neutron radiographs in term of spatial resolution the normal technique was complemented with another method. The utilisation of the dysprosium foil/X-ray film technique is supplemented with a track-etch recorder consisting of a cellulose nitrate film. For further examination and handling the cellulose nitrate film can be digitised to allow electronic image treatment. Promising results were obtained with this technique on neutron radiographs of reactor fuel rods, namely higher spatial resolution compared to the normal technique, high contrast and sharp images. In the presentation the techniques and the film material shall be introduced and the obtained results will be illustrated with examples.

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1 Introduction

The PIE techniques at the IFE nuclear fuel section are improved through continuous upgrading of equipment and method during many years. E.g., preliminary testing of a solid nuclear-track detector, designed for recording ionizing particle tracks and for utilization in neutron radiography, gives promising results compared to the classic or standard dysprosium foil technique.

The track-etch recorder we used consists of a 100 μm thick film of lightly rose-tinted cellulose nitrate coated on both sides with an energy converter (n, α) material. This technique gives sharp and detailed images. The use of the film not only eliminates the prolonged handling of activated screens required by classical methods, but also enables much higher resolving power to be achieved.

Both methods allow unfiltered neutron beam to be, and enables neutron radiography of very active samples such as irradiated fuel rods. [2].

Solid nuclear-track detectors have numerous applications [2], for example the dosimetry of neutrons and of the heavier ionizing particles such as alpha particles, protons, fisson fragments, spallation fragments, heavy ions and very heavy ions such as cosmic rays etc. In every instance the thickness of the cellulose-nitrate substrate should be in accordance with the mass and energy of the type of particle to be detected in order to obtain the optimum level of energy transfer.

2 Neutron radiography of irradiated fuel rods - an approach to improve spatial resolution in neutron radiographs.

A dysprosium foil/x-ray film technique (activation transfer method) is normally applied under neutron radiography inspection (non destructive) of irradiated fuel rods consisting of UO_2 and PuO_2 pellets, zircaloy cladding and instrumentation devices such as fuel pressure transducer (PF), fuel thermocouple (TF) and cladding elongation transducer (EC). The main objective here is to reveal fuel rod degradation or failure, i.e. intrusion of moisture and detection of major fuel in-homogeneity. Preliminary testing of a cellulose nitrate film (Kodak CA 80-15 type B) coated with an energy converter (n, α) gives sharp and detailed images compared to the standard dysprosium foil/x-ray film technique [3, 4]. Illustrating examples of the various techniques are compared in the paper.

Device edges and other features between 50 – 100 μm are possible to detect if the quality of the neutron radiographs is satisfactory.

2.1 Experimental set-up in neutron radiography

Principle: The principle of image formation in neutron radiography is based on attenuation (i.e. the sum of absorption and incoherent scatter) of a primary beam of neutrons in an irradiated sample. The degree of attenuation is measured by detecting the intensity I of the neutron beam passing through the sample (fuel rod components) with unaltered direction and it is given by $I = I_0 \exp(-\Sigma_t z)$, where z is the sample thickness. I_0 is the intensity of the incoming beam and Σ_t is the total macroscopic cross section of the material, i.e. $\Sigma_t = \Sigma(n\sigma_a) / V + \Sigma(n\sigma_s) / V$. Σ is the sum of all the different atoms in the volume V, and n is the number of atoms of one kind with microscopic absorption cross section and microscopic scattering cross section given by σ_a and σ_s.

The primary source of neutrons was obtained from one of the radial channels of the JEEP II reactor (2 MW, central thermal flux 3×10^{13} n/cm^{2}s) at the Institute for Energy Technology, Kjeller. The samples (fuel rods) were irradiated by a collimated thermal neutron beam of height 220 mm and width 30 mm (figures 1 and 2).

2.2 Methods applied in neutron radiography

The disprosium foil/x-ray film technique: The method relies on the build-up of radioactivity in the foil produced by neutron absorption. In this way an activation image is formed in the foil. The neutrons captured by the dysprosium foil are generating radioactivity that subsequently decays with a convenient half-life [1],

$$^{164}_{66}Dy + ^{1}_{0}n \rightarrow ^{165}_{66}Dy \xrightarrow[2.3\ h]{\beta^-,\gamma} ^{165}_{67}Ho.$$

The patterns of radioactivity can be transferred to the x-ray film (i.e. radiograph) by simply placing the metal foil in close contact to the film after the irradiation. The x-ray film is irradiated for several hours and thereafter developed using a standard photographic technique.

The cellulose nitrate film technique (track-etch recorder): The film consists of a 100 μm thick layer (substrate) of lightly rose-tinted cellulose nitrate. The film is primarily intended for recording the emission from α-particle sources (below 4 MeV) and the film intended for neutron radiography is therefor coated with an energy converter (n, α) material. The record thus obtained is a neutron radiographic image formed by means of the (n, α) energy converter, i.e. the neutrons impinge the vacuum deposited lithium tetraborate layer or converter screen that promptly emits ionizing α-particles intruding and generating damage pits in the cellulose nitrate matrix. The α-particles takes short and straight paths through the material and so give good resolution.

After irradiation, the converter layer is removed by washing off in water and the cellulose nitrate film is thereafter etched in a 10 % solution of caustic soda (NaOH) at 60 °C for approximately 10 minutes. *The damage produced along the particle tracks alters the chemical properties of the cellulose nitrate material in this region in such a way that the damaged area is removed by the etching process [2].*

Electronic data treatments (digitising): After developing, the neutron radiographs (cellulose nitrate- and x-ray film) are digitised in transparent mode with a "state of the art" photo-scanner controlled from a power Macintosh computer. The main purpose here is to reproduce neutron radiographs for reports etc. with a highest achievable quality in order to visualise for instance retrofitted TF tip position and other fuel rod details (e.g. pellet crack pattern and device dimensions). By electronic data treatment, the neutron radiographs can be enlarged, spliced and combined with other PIE data of interests (e.g. fuel rod macro photos).

2.3 Qualification tests performed on neutron radiographs from irradiated fuel rod

Quality inspection of neutron radiographs: The cellulose nitrate film and dysprosium foil techniques were both applied in neutron radiography of an irradiated fuel rod from a fuel assembly irradiated in the Halden Boiling Water Reactor (HBWR). For assessment of the neutron radiography quality, it is necessary to have certain indicators that expose the resolution and sharpness details of the neutron radiographs. A bellows pressure transducer (PF) is used as a quality indicator in the following example. Fig. 3 shows several radiographs (1:1) of both methods from the upper end of the fuel rod. Important details are marked on the figure where the plenum spring, the blinded PF and the upper end plug (EC support) are seen. The neutron radiographs were digitised with a relatively high resolution equal to 600 dpi, i.e. approximately pixel size of 45 μm. From a first glance at Fig. 3, it can be seen that the images digitised from the cellulose nitrate film exhibit higher quality in terms of sharpness. To really check the quality of the images, it is necessary to enlarge them. The PF in Fig. 4 is enlarged three times in example B. It is easy to see the bellows and other details on the copies from the cellulose nitrate film. On the copies taken from the x-ray film the details are almost lost. This fact is even better illustrated at still higher magnifications. From Fig. 4 (example C) it can be seen that the x-ray film contains no important information at all. The situation is quite different for the cellulose nitrate film. The enlargements show sharp details around 100 μm size. For instance, the PF's bellows (steel) is easy to see and dimensional measurements can also be done. Two other enlargements are given in examples D and E. It is again a striking feature that the x-ray film neutron radiographs contain more noise than the cellulose nitrate films. The reason for the quality difference is mainly related to the spreading of the gamma rays generated in the dysprosium foil material and the spreading of the light beam in the x-ray film under digitising. With a thinner Dy-foil the gamma radiation will spread less and thereby produce sharper film details. However, the α-particles generated in the energy converter (deposited on the cellulose film) during irradiation produce sharp details in the cellulose nitrate film.

Instruments readings from neutron radiographs: An example from a *sphere pac* fuel rod is used to illustrate how the *end of life pressure* can be estimated from the total displacement of the core holder or PF's bellows squeezing after irradiation. The PF bellows is shown in fig. 5 section A. The PF bellows and fuel rod are pre-pressurised to 21.5 and 22.5 bar respectively at room temperature. The sensitivity of the bellows is given to 11.14 bar pr. mm and an initial core holder position of 0.1 mm from the bottom or fundament (see figure) is assumed. The total displacement of the core holder is measured to be 1.26 mm. The initial filler gas amount is now at elevated temperature because of decay heat from the fuel. The pressure should therefor be slightly higher than 22.5 bar concerning only the fuel rod filler gas. If we assume a fission gas temperature of 30 °C the associated pressure is 23.6 bar, i.e. the PF bellows is squeezed due to fission gas release and also because the filler gas now is at higher temperature. The bellow displacement is approximately 0.1 mm due to the filler gas temperature increase. This gives a bellow squeezing of 1.16 mm. The fission gas pressure is therefor 12.7 bar at the actual temperature and 12.1 bar at room temperature, i.e. a final pressure of 34.6 bar for fuel rod gas composition.

The fuel rod pressure was measured in the HBWR to 34 bars at room temperature and the two results are in good agreement. The small uncertainty of the *end of life pressure* estimation given above is related to where the core holder initially was located at room temperature. An acquisition of neutron radiographs acquired before irradiation of the fuel rod would remove this uncertainty.

Inspection of fuel and canning details: An example of fuel inspection from neutron radiographs acquired with the two techniques is given below. Figure 6 shows part of the fuel column from a rod utilised in a dry-out fuel behaviour experiment. The region exposed is characterised with a relatively high degree of fuel cracking and it is away from the dry-out zone, i.e. lower end of fuel rod. A 1:1 image is first given (A) and we will again try to compare the quality of the cellulose nitrate and x-ray films. The images in B and C are enlarged 2 and 5 times respectively and again it is obvious that the neutron radiographs acquired with cellulose nitrate film are much more detailed than the x-ray film examples.

Figure 5 example B shows an image acquired in the dry-out region of another fuel rod. It is possible to see that the canning is squeezed in between the pellet interface in a permanent plastic deformation. The neutron radiograph is acquired with the cellulose nitrate film technique.

Examples from a *fuel degradation test* are given in the next figures, i.e. Nos. 7, 8, 9 and 10. The objective of the *fuel degradation test* was to study phenomenon and mechanisms of severe degradation of the fuel cladding caused by a small primary defect away from the location of secondary failure. For simulation of the primary defect, the interior of the rods was exposed to the coolant water by opening an in-core valve.

The dysprosium foil/x-ray film technique was applied for neutron radiography on *the fuel degradation test* rods in two angular orientations. These radiographs were used for an overview inspection of the fuel rods. From the overviews (figures 7 and 9) it was easy to sort out interesting fuel rod regions for further examinations. These specific regions were again neutron radiographed, but now the cellulose nitrate film technique was applied. Several conclusions were drawn from these pictures, e.g. some pellets located near position 795 mm have centre voids and there is also a crack (defect) in the canning at the same location (see the 4X magnification in figure 8) that is generated due to pellet cladding mechanical interaction (PCMI).

At position 735 mm in figure 10 (another fuel rod in the test) we can again see voids in the pellet centre and in addition there is an axial crack in the canning that is generated from a PCMI mechanism.

3 Concluding remarks

Some examples given in the paper show that it is possible to enlarge neutron radiographs acquired with the cellulose nitrate film technique. The fuel rod details are still quite sharp after 10 times magnification so dimensional measurements of fuel rod details are possible with great precision (50 μm) in contrast to the dysprosium foil/x-ray film technique. Fuel rod instrument devices and other details could therefore be detected with high reliability and the results are used in for instance estimation of *fuel rod end of life pressure.*

It is also useful to combine the dysprosium foil/x-ray film and the cellulose nitrate techniques in evaluation of fuel and canning conditions.

The last three years upgrading and use of advanced electronic data treatments in post irradiation examination (PIE) have improved the quality of all PIE, e.g. neutron radiography.

4 Acknowledgements

I wish to thank Mr. Per Arnesen, Mr. Knut Bryhn-Ingebrigtsen and Dr. Barbara C. Oberländer for interesting discussions and suggestions during the neutron radiography project.

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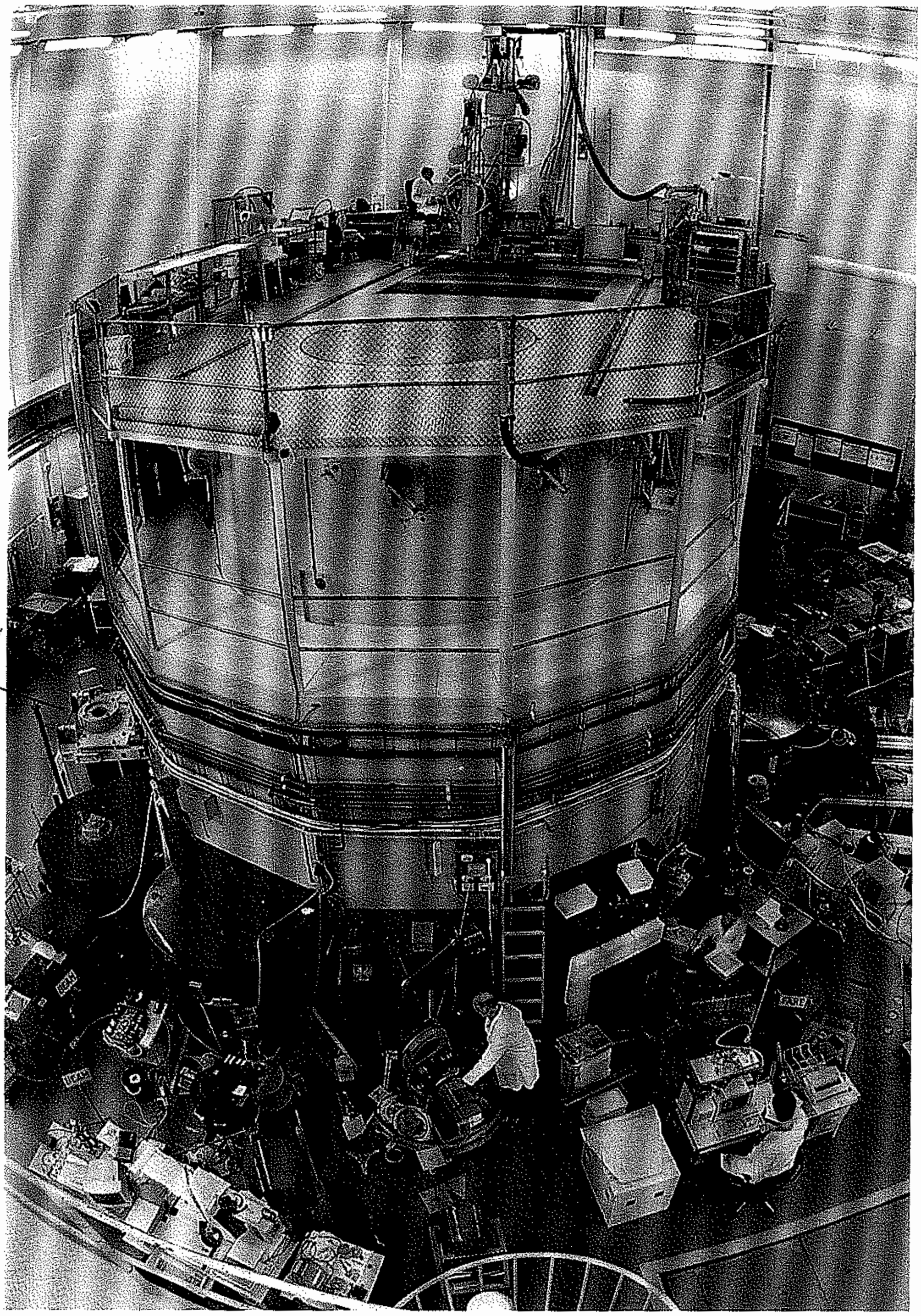

Fig. 1 JeepII reactor with experimental beam channels

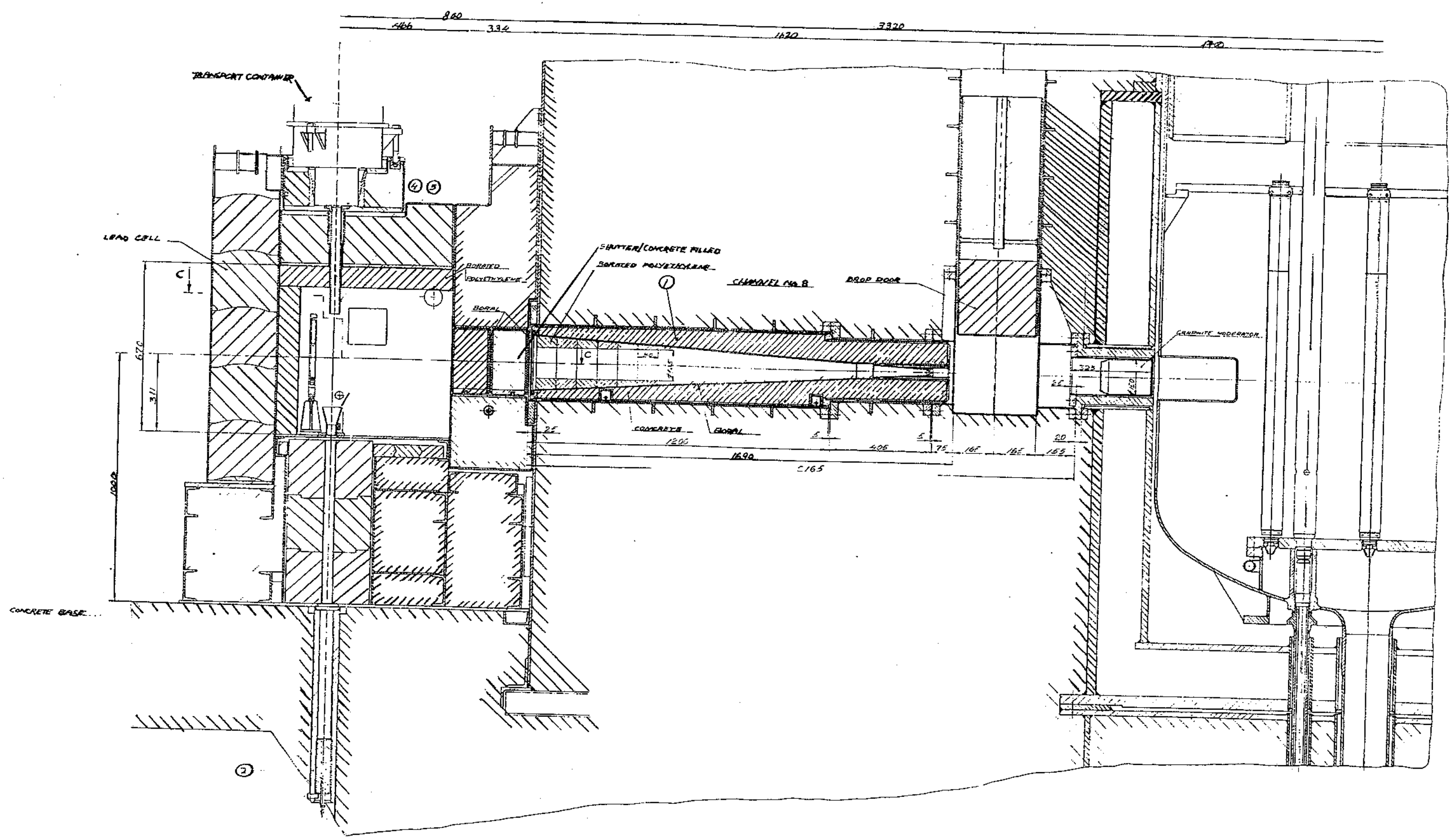

Fig. 2 Schematic view of the neutron radiography facility.

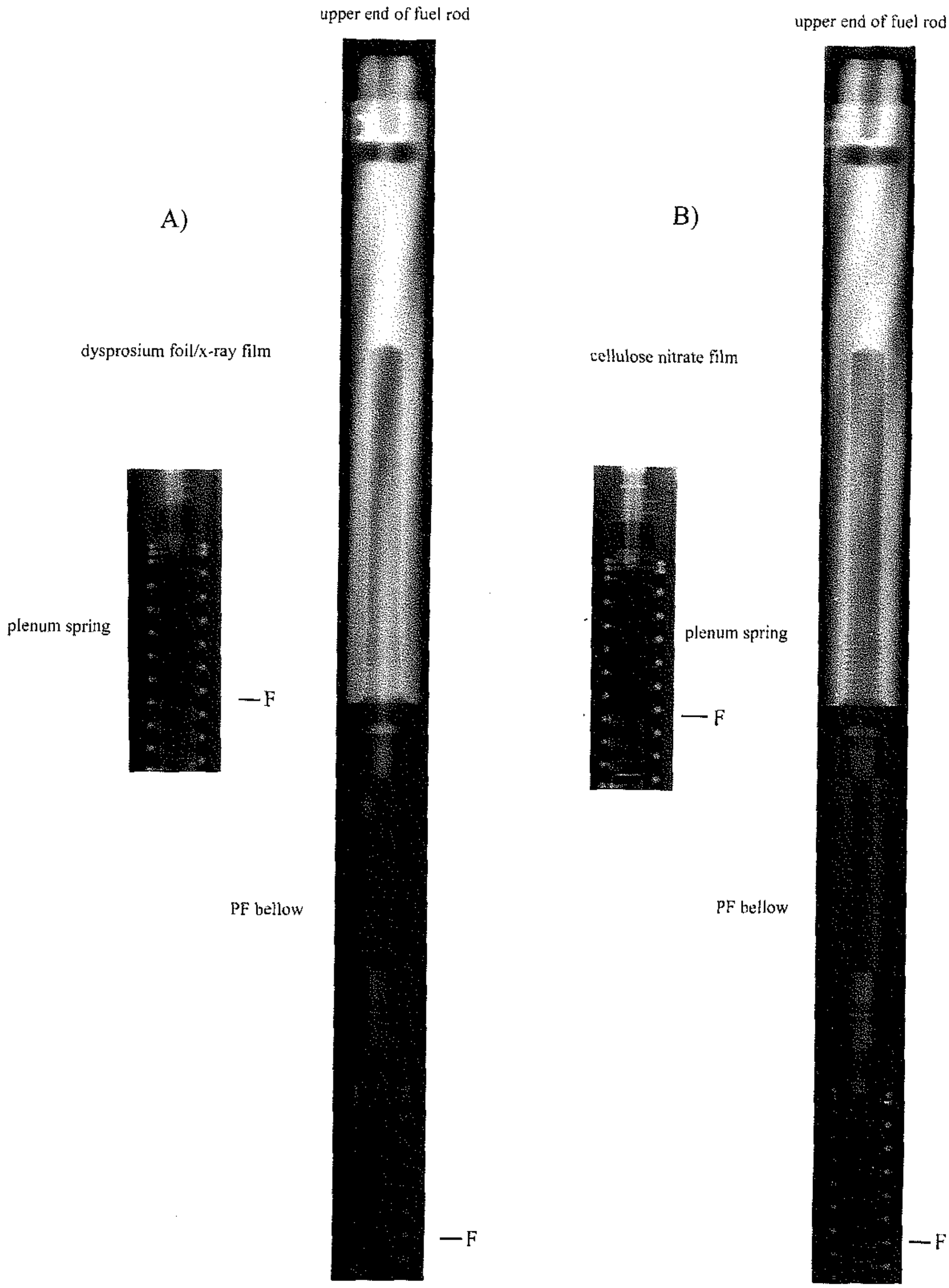

Fig. 3 Neutron radiography applied with the dysprosium foil (A) and cellulose nitrate (B) techniques.

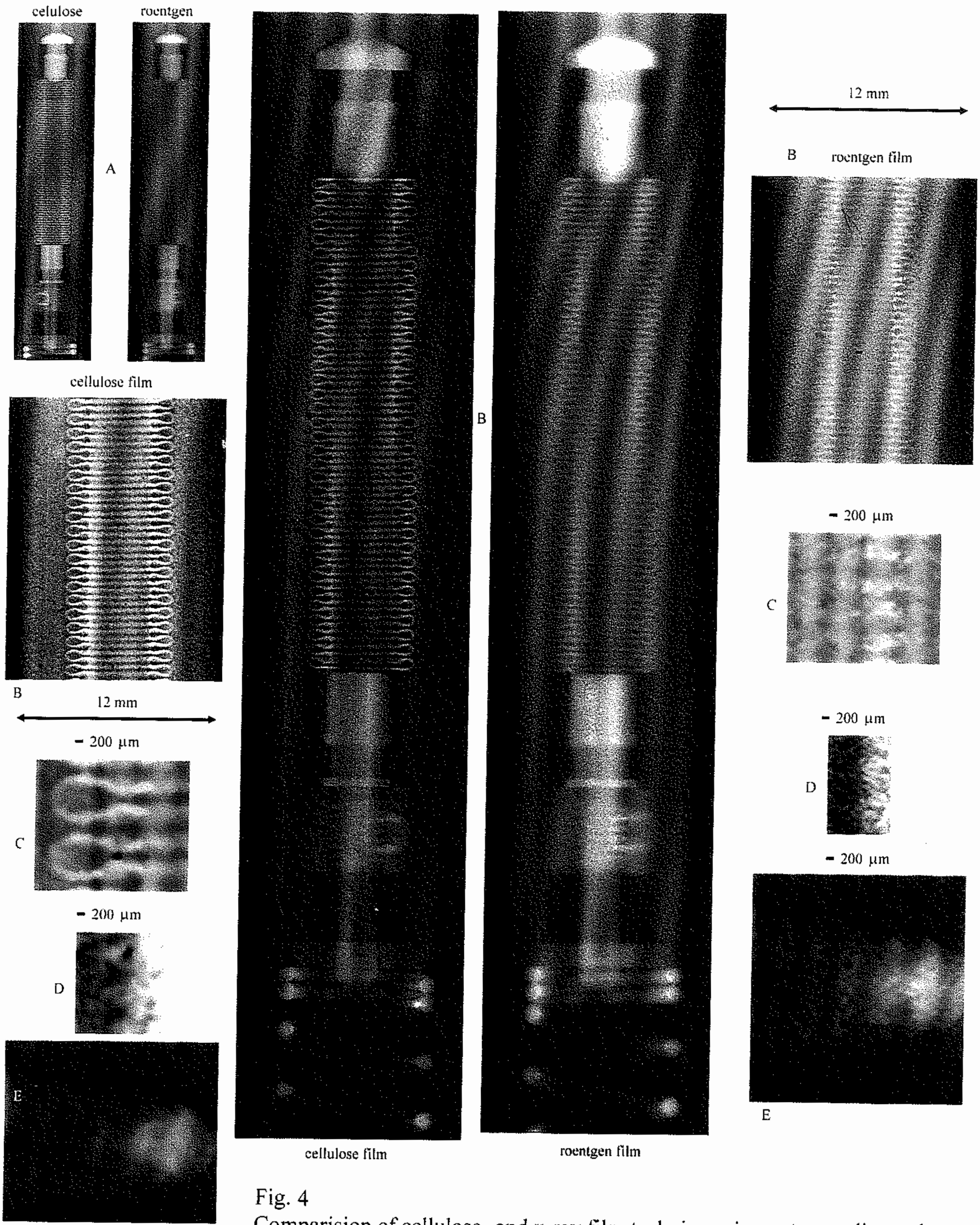

Fig. 4
Comparision of cellulose- and x-ray film techniques in neutron radiography.

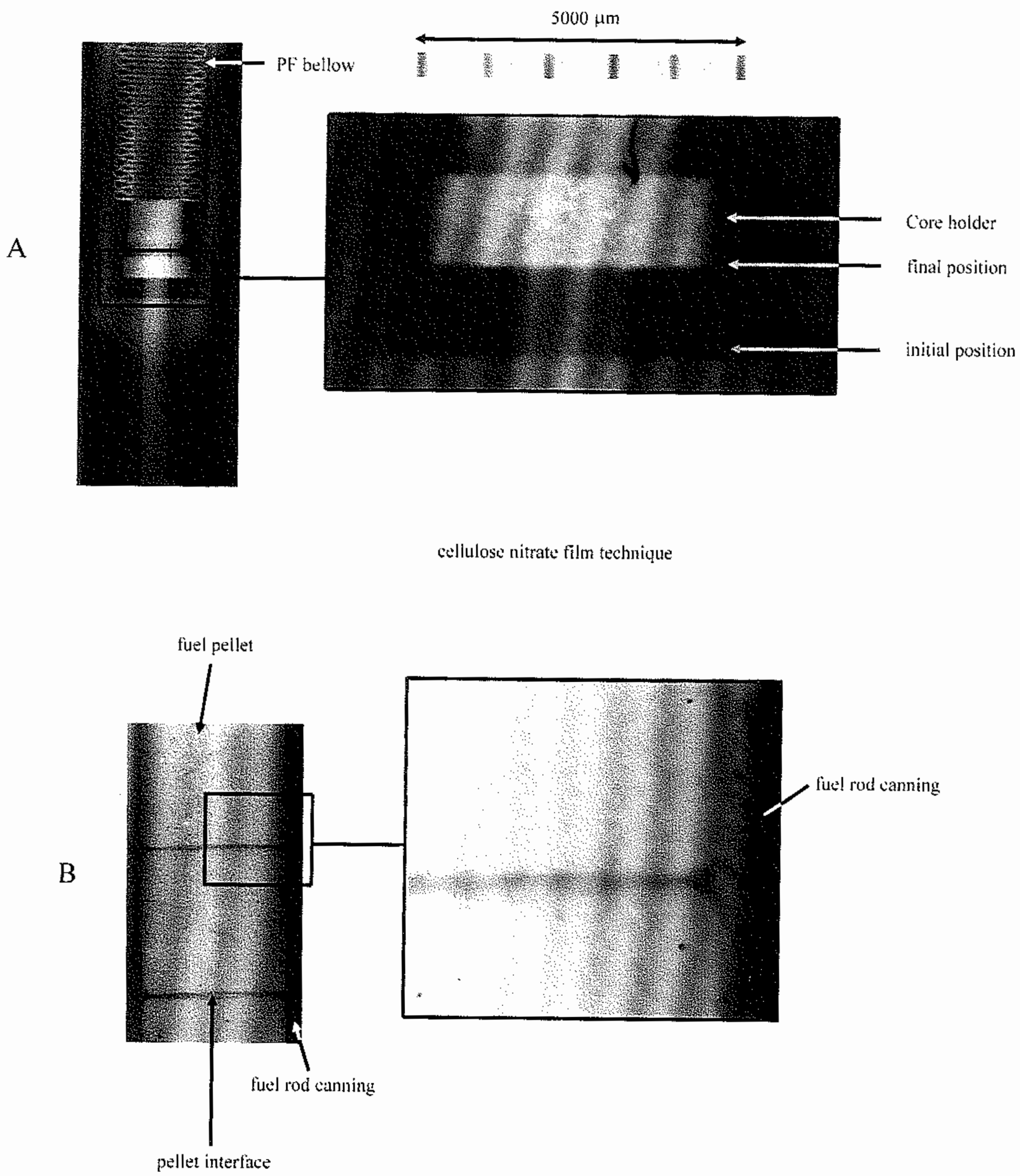

Fig. 5 Neutron radiograph of PF core holder (A) and canning (B).

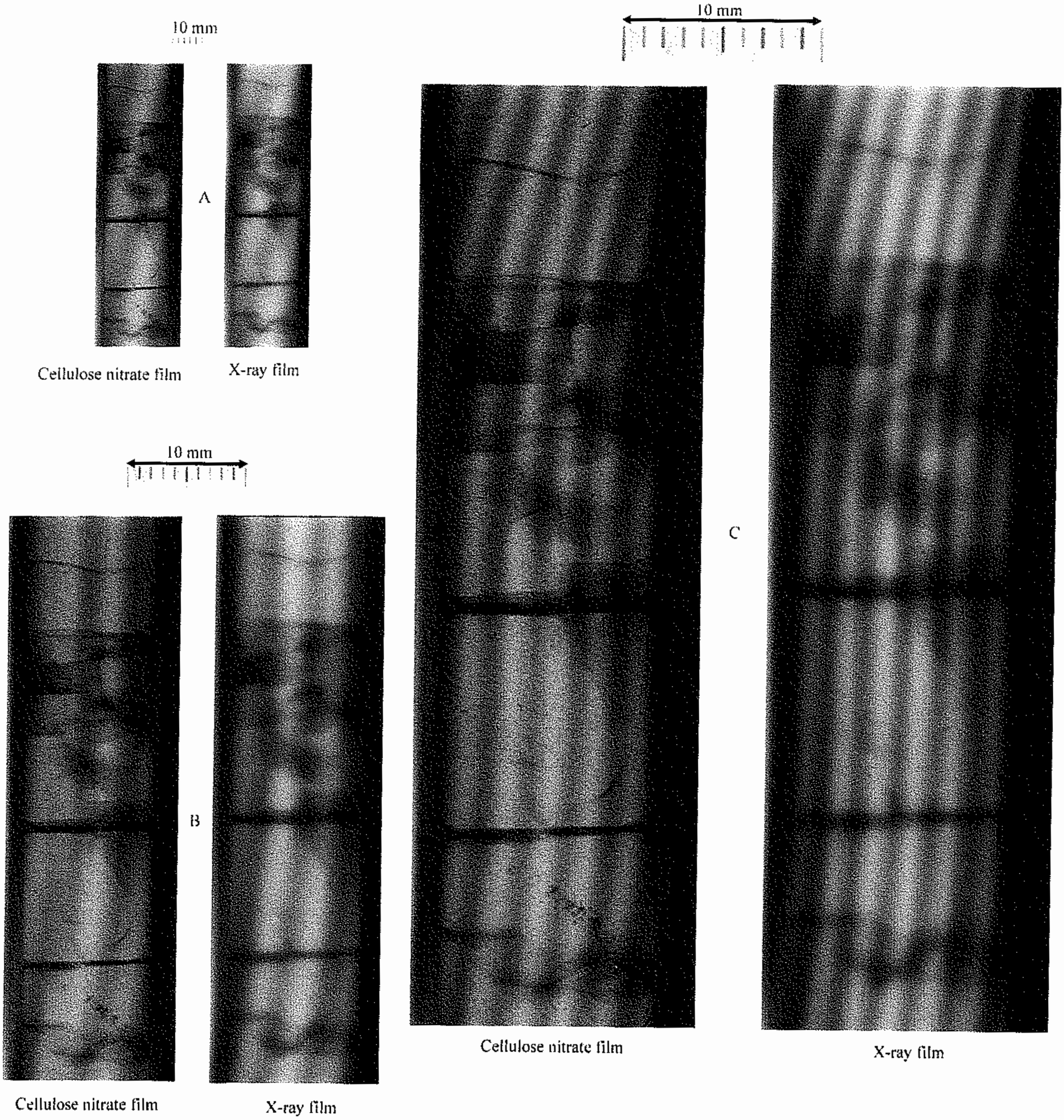

Fig. 6 Neutron radiographs of fuel column.

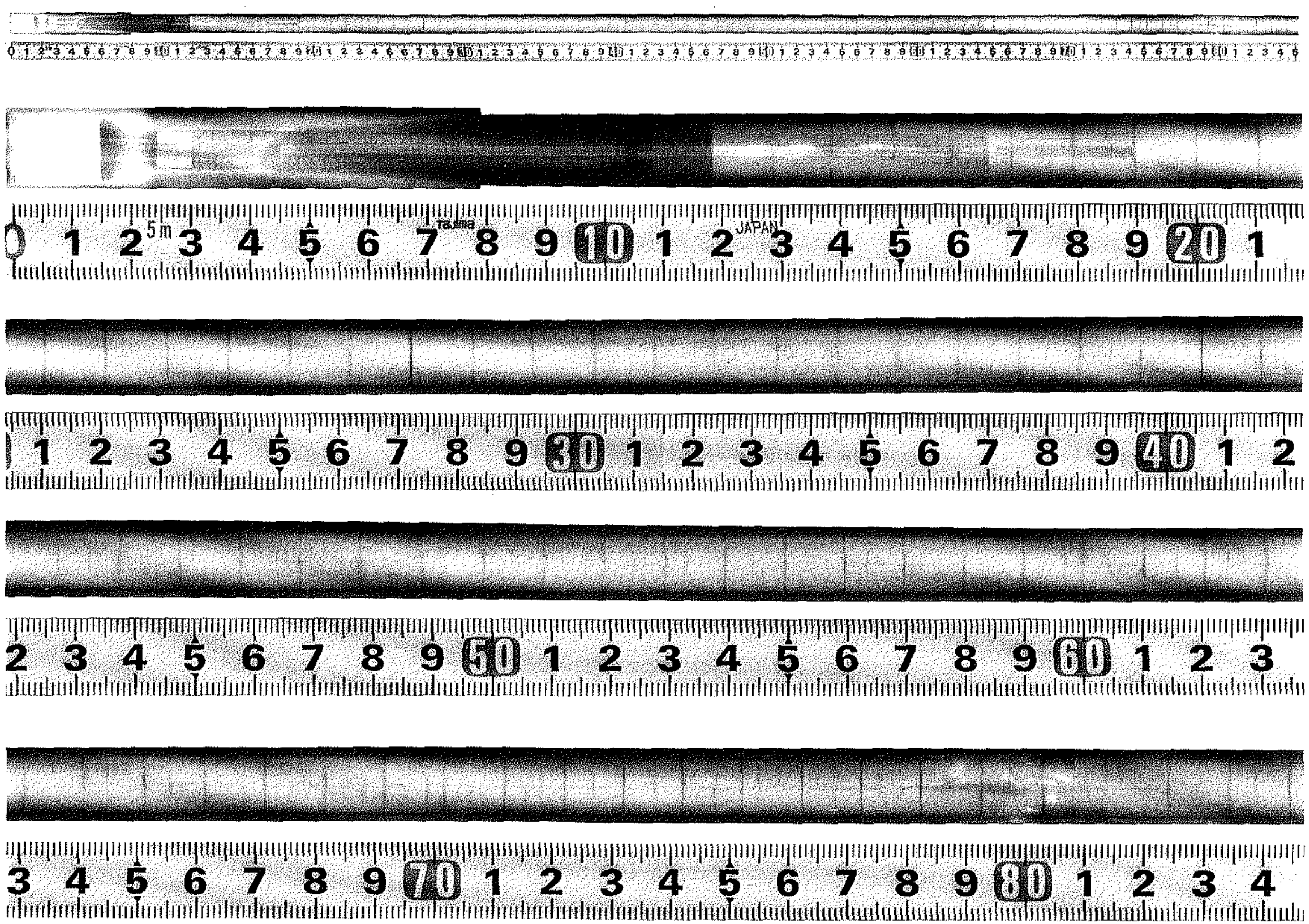

Fig. 7 Neutron radiographs of a fuel rod at 90 degrees orientation from the fuel degradation test.

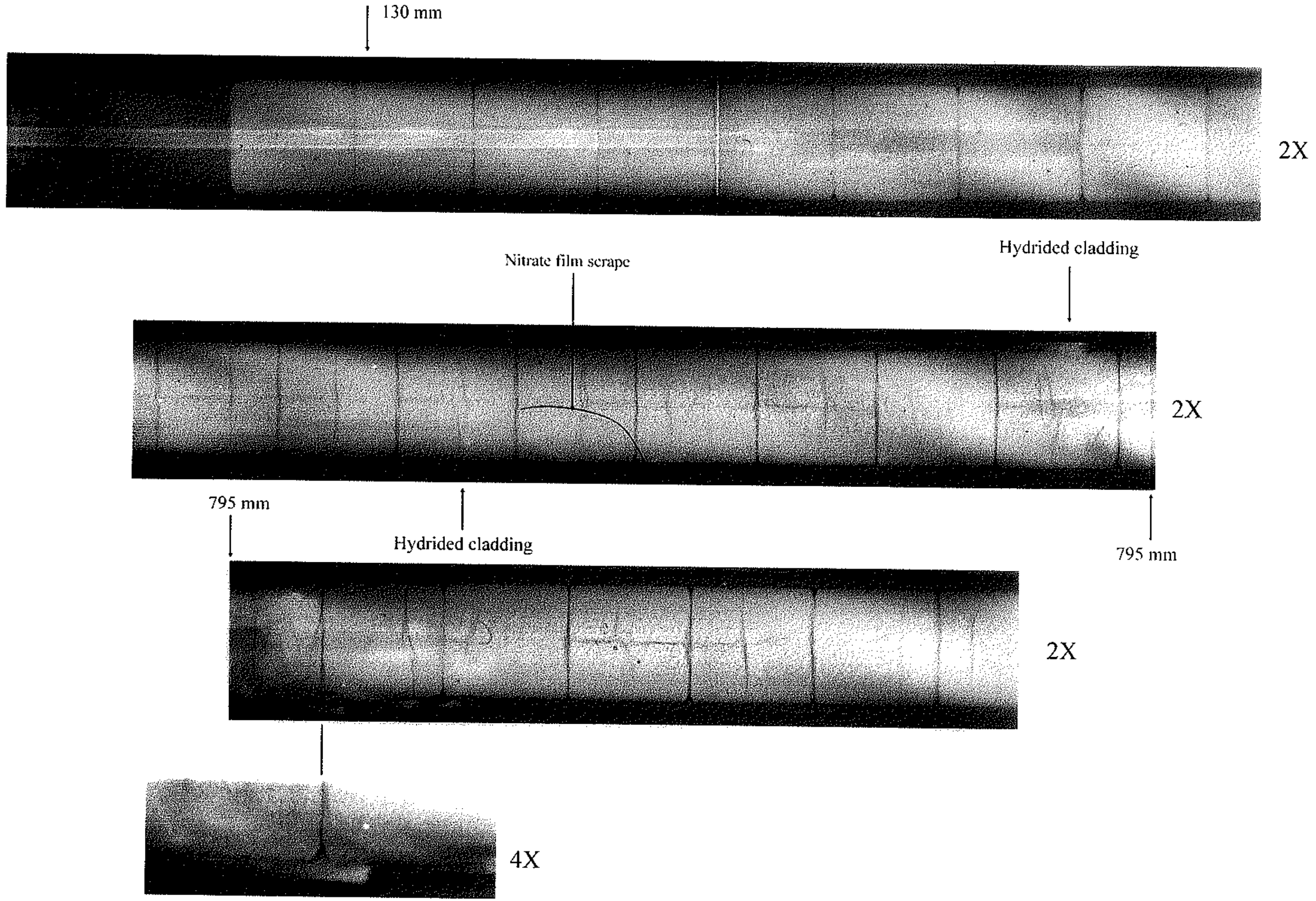

Fig. 8 Neutron radiographs(cellulose nitrate film) of a fuel rod at 90 degrees orientation from the fuel degradation test.

Fig.9 Neutron radiographs of a fuel rod at 90 degrees orientation from the fuel degradation test.

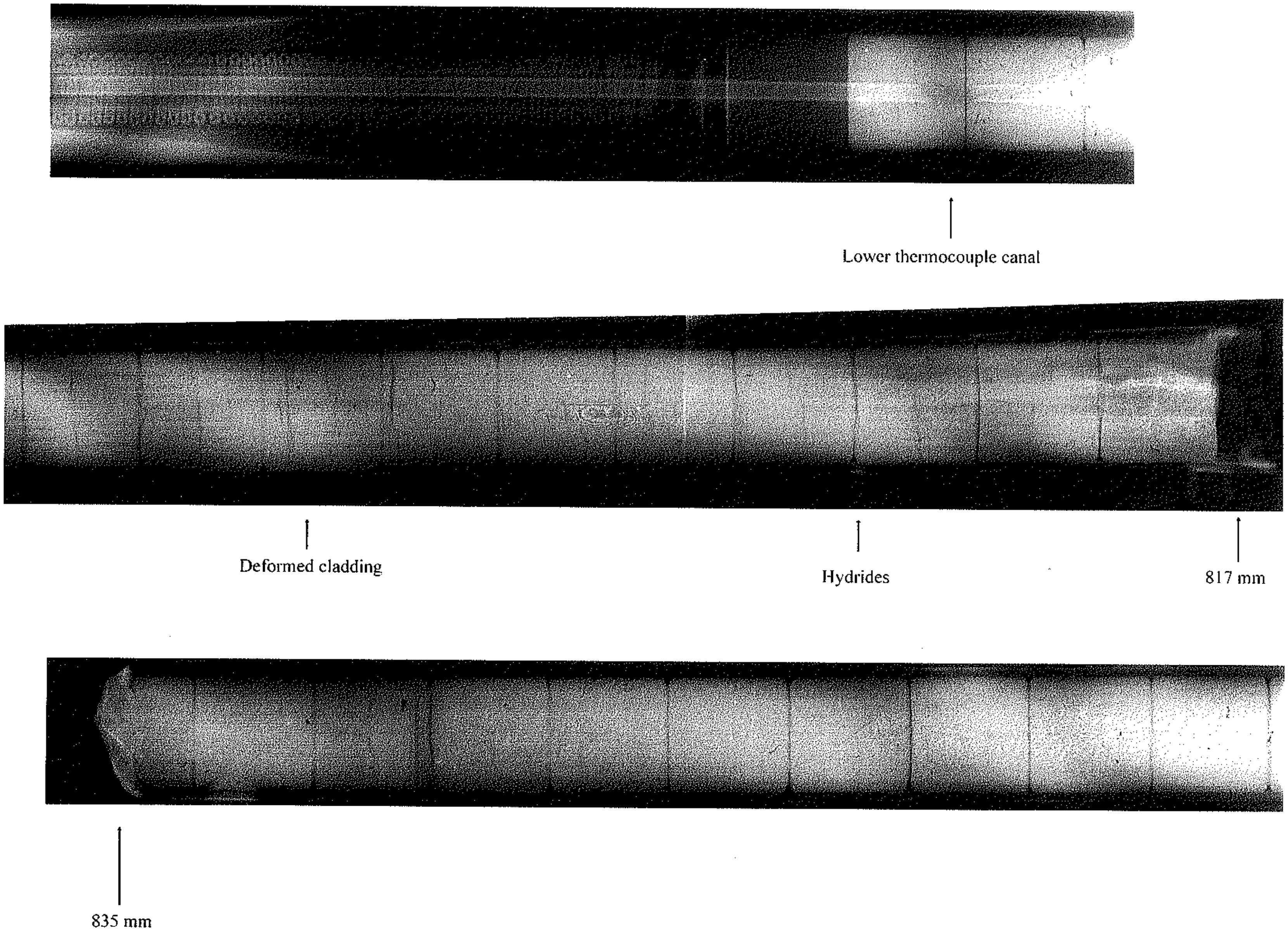

Fig.10 Neutron radiographs (cellulose nitrate film) of a fuel rod at 90 degrees orientation from the fuel degradation test.