POST-IRRADIACION EXAMINATION OF U_3SI_X-AL FUEL ELEMENT MANUFACTURED AND IRRADIATED IN ARGENTINA

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Abstract

As a part of CNEA´s qualification program as a supplier of low enriched Al-U_3Si_2 dispersion fuel elements for research reactors, a post irradiation examination (PIE) of the first prototype of this kind, called P-04, manufactured and irradiated in Argentina, was carried out. The main purpose of this work was to set up various standard PIE techniques in the hot cell, looking forward to the next steps of the qualification program, as well as to acquire experience on the behaviour of this nuclear material and on the control of the manufacturing process. After an appropriate cooling period, on May 2000 the P-04 was transported to the hot cell in Ezeiza Atomic Centre. Non destructive and destructive tests were performed following the PIE procedures developed in Argonne National Laboratory (ANL), this mainly included dimensional measurement, microstructural observations and chemical burn-up analyses. The methodology and results of which are outlined in this report. The results obtained show a behaviour consistent with that of other fuel elements of the same kind, tested previously.

On the other hand the results of this PIE, specially those concerning burn-up analysis and stability and corrosion behaviour of the fuel plates, will be of use for the IAEA Regional Program on the characterization of MTR spent fuel.

1. INTRODUCTION

Fuel element P-04, first full scale prototype of U_3Si_x-Al plate type dispersion fuel manufactured in Argentina, is considered as the precursor of a new series of this type of Si-based LEU fuel elements, in the framework of CNEA´s activities regarding the development and qualification of technology for the manufacturing of this kind of fuel elements. [1]

Test P-04´main characteristics of the fuel matrix meat are Al - 39% $U_3Si_{1.5}$ volumen (active compound is 5.5 % wt in Si) with about 15 % of porosity and the resulting uranium meat density was 5 gU/cm3. The nominal thickness of the meat is 0.51mm , clad with 6061 aluminium alloy and the plates thickness are 1,3 mm and 1.6 mm for the inner and outer, respectively. All the as-fabricated specifications and attributes were based upon ORNL specifications. The assembly finally reconstructed was of a standard RA-3 box-type with 19 flat plates.

Test P-04 was irradiated in the RA-3 reactor during 300 effective day of operation of 5 Mw, exposed to a maximum thermal flux of 3.6×10^{13} $n.seg^{-1}.cm^{-2}$. The fuel element was removed from the core on October 27, 1997, when it had reached an average burnup (^{235}U depletion) of 23.5 % according with the reactor fuel management calculation.

2. POSTIRRADIATION EXAMINATION OF THE FUEL ELEMENT

On May 18, 2000, the P-04 was transported to the hot cell at Ezeiza Atomic Centre (Photograph 1) and the PIE´plan discribed in [1] was started. That included the main non-destructive and destructive postirradiation standard tests and metallographic examinations[2].

After visual examination and dimensional inspection of the entire element, P04 was dismantled and all the plates were carefully inspected. Some plates were selected for thickness measurement, determination of the gamma scanning profile and volume measurement by the immersion method. One inner and an outer plate were also tested for blister threshold temperature and behaviour of the meat-clad bonding. The one other outer and inner plates that had been gamma scanned were selected for destructive examination, to obtain samples for microstructural observation and chemical burnup analyses.

2.1 Visual Examination of the entire fuel element

The elements were examined visually through the cell window with the help of a magnification glass and a telescope. The outer appearance of P-04 was as in the as-fabricated condition, with the exception of the oxide film and several handling marks. No abnormal features were observed. Photographs 2 shows the external state of P04.

2.2. Dimensional inspection

The estimation of dimensional changes and the measurement of the more relevant outer dimensions of the fuel element were made by positioning the element on a gauging table with x-y movements and a micrometer at the desired point of measurement, and comparing the readings to reference blocks. No significant bow, twist or swelling was observed on any face of the fuel element. The width between side plates outer surfaces and the height between outer fuel plates outer surfaces did not show changes from the as-fabricated specifications.

2.3. Coolant channel gap estimation

To allow the inspection of all the fuel plates and the corresponding coolant channels, it was necessary to remove the handling bar. A machine that had been specially designed for the dismantling of the fuel element was used, which disengages both structural side plates by rolling them. A low stress was applied with this machine to begin the rolling operation, which was enough to break the handling bar weld.

After this operation, the coolant channel gap thickness was estimated using backlighting illumination. The constancy of the coolant channel gap was visually verified all along the plate length, by varying the focus of the telescope. In addition a plastic strip gauge, 2.5 mm thick, was easily slid along several channels to verity that the gap width exceeded this value. The perfect flatness of the plates could also be observed.

Photographs 3 shows the view through coolant channels after the removal of the handling bar, focussing on the upper part of the assembly. The bottom plate of the stack corresponds to the plate located at slot number 1. The plate at the 6^{th} slot shows a small dent in its upper edge, caused by an impact made during the fuel element handling at the pool. The plate located on slot number 10, which is situated just below the handling bar, showed a leaking blister, convex on both sides of the plate, of approximately 3 cm in diameter. This defect was responsible of the fission product release detected in the reactor pool, that led to the decision of ending the P-04 irradiation. This failure was clearly identified as caused by damage done on this plate during the assembling of the element. The analysis of this failure will not be discussed in the present report, as it does not relate to the behaviour of the fuel element, however it will be the subject of another report regarding the degradation of defective Si–based dispersion fuel.

2.4. Dismantling of the fuel element

Photograph 4 shows the rolling operation with the machine mentioned above. In this method the upper ends of the both side plates are clamped to two lateral pulleys, and by rolled slowly over them, the fuel plates were disengaged from the side plate**.**. Photograph 5 shows P04 during the beginning of the side plate rolling operation. No shavings of the side plates are formed and the fuel plates were pulled off without any damage.

3. EXAMINATION OF FUEL PLATES

3.1. Visual inspection of the plates

After the removal of the fuel plates a complete visual examination was done, as well as the verification of the identification number of each plate with the help of a magnification glass. They were immediately

numbered with a marking pen, according to the position of the corresponding slot, so as to be easily identified inside the hot cell.

None of the plates did show any warping or bowing, appearing on the contrary perfectly flat. With the exception of the defective plate, no evidence of any kind of blisters was found on any other plate. The oxide layer seemed to be uniform, not showing any pitting or other kind of localised corrosion. In all cases the oxide layer show a white and reflective colour on the meat zone, and a little darker on the frame. On plate No. 11, at the face opposite to the leaking hole of the blister of the defective plate, a stain left by the release of active material was observed.

Only five plates were chosen for the first stage of the postirradiation standard tests, the rest being stored temporarily in the storage channels attached to the hot cell. The selected plates were the two outer plates (No. 1 and No. 19), and three inner plates (Nos. 8, 9 and 11). Also the defective plate (No.10) was kept to perform additional examinations.

3.2. Plate thickness measurements

Plate thickness measurements were made with a micrometer with a ¼ " diameter spherical tip at the desired points of measurement. The points were aligned along two tracks, each one at 2 cm from each side edge of the plate. Two points outside the meat, at the upper and bottom part of the plate, and five points equally spaced over the meat zone, were taken on each track. All the selected plates were measured in this way. The following Table 1 shows the obtained thickness values, indicating for each case the corresponding burnup range, according to the gamma scanning activity profile (to be detailed later).

Table 1: Plate thickness (average values in mm)

	As-fabricated (mean value)	Unfuelled Region	Low Burnup	Medium Burnup	Peak Burnup
Plate 1	1.643	1.62	1.66	1.67	1.65
Plate 9	1.298	1.30	1.32	1.34	1.31
Plate 11	1.298	1.30	1.36	1.38	1.38
Plate 19	1.640	1.67	1.67	1.66	1.65

Data in Table 1 shows there appears no indication of swelling in the peak burnup zone. Some thickness values would indicate a small volume increase, however the volume change calculated by the immersion method was practically nil or slightly negative. Variations in thickness can be solely attributed to characteristics of oxide formation and/or dirt particles on the surface of the plate.

3.3. Volume measurements by the immersion method

Before performing the measurement of the plate volume by the immersion method, the oxide layer was removed with 20% v/v phosphoric acid solution. One inner plate (Nª 9) and one outer plate (Nª 19) were selected for this test. The inner plate was attacked with the solution at about 70ºC and the outer plate at 30ºC. In both cases, on the periphery of the plate the layer was removed much faster than on the central zone of plate. This indicates very different thickness and nature of the oxide. In the first case, with the hot treatment, the periphery of the plate was quickly attacked while on the central zone the oxide film dissolved slowly; at the end of the process the weight loss was close to 10 g, indicating a high degree of metal dissolution. For the second (outer) plate, at a lower solution temperature, the rate of dissolution was slower and a more controlled oxide removal was obtained; the weight loss was less than 4 g.

The volume measurement by the immersion method gave a decrease in the volume of the plate between 0.1 and 0.2% Taking into account that this value is very close to the accuracy of the method, and assuming that this variation is solely attributable to the meat, this would give as a maximum, a variation in the meat volume of -0.6 % for the inner and of –0.4 % for the outer plate. This indicates a very light meat contraction (shrinkage), which is consistent with the known behaviour of redensification of the meat material at a low burnup level.

3.4. Blister threshold test

A split furnace made *ad-hoc* for the dimensions of the P-04 plates, with a temperature controller, was used to perform this test. The electrical heater was carefully designed, in order to guarantee a flat temperature profile along the total length of the plate. A extractable built-in holder allows the removal of the plate from the furnace, to cool it and examine it. Then the test can be continued, going on to the next step at a higher temperature, without turning off the furnace.

Due to the low burnup reached by the P-04 and the meat contraction (shrinkage) detected, no blister formation was expected; however this test is also useful to verify the behaviour of the meat-clad bonding and evaluate the quality of the lamination process performed during the manufacturing.

The outer plate Nº 1 and the inner plate Nª 11, were tested. The plates were held for intervals of 30 to 60 minutes at temperatures from 350 ºC to 500 ºC, increasing in 25 ºC steps. In both cases no blistering was observed, confirming the previous hypothesis. The surface of the plate remained intact; only a loose oxide layer (built up in the air of the hot cells) was observed. Only at 500 ºC the plates began to show a slight warping.

In plate Nº 1, the test was continued up to 575 ºC. This plate still remained free of blistering but the warping was very extensive in the meat zone. Photograph 6 shows expanded meat zone while the frame remained flat (metallographic observations will be done to study the transformation occurred in the meat at this temperature). Even in this condition of high stress and strain the meat-clad bonding resisted perfectly and no cracks or fissures were observed.

3.5. Plate Gamma Scanning

The same plates were used for the volume measurements and for gamma scanning, the inner plate Nº 9 and the outer one Nº 19.

The plates were moved on a track in front of a collimator. This consists of a hole of 2 mm diameter and 150 mm long, made in a Pb block. The high resolution HP Ge detector was installed at a distance of 3 meters from the collimator to avoid the incidence of scattered rays. Gamma energy spectra were obtained at intervals of 1 cm , along the axial and transverse center lines of the plates. The main peaks found, in decreasing order of intensity, are Cs137, Ce144/ Pr 144 and Cs134.

For any desired peak both profiles, axial and transverse, can be obtained from the net area under the corresponding peak in each measurement. Figure Nº 1 shows, for plate Nº19, the gamma activity axial profile normalised with respect to the maximum value for the three nuclides mentioned above. The square root of the Cs134 activity accounts for the fact that a double neutron capture is needed for its formation. This figure also includes the normalised values of the axial thermal neutron fluxes measured during irradiation of P04, obtained by the foil activation method [3]. A good agreement between the profile shape and the foil activation data is observed for all the nuclides.

The form-factor Count max./Count average of center line axial profile, with the average value calculated from the integral of the values in each measurement interval, is 1.30 to 1.33 for both plates Nº 9 and Nº 19 and for all the nuclides considered. However, for the following analysis the results for Cs137 are used, as this nuclide is recognised to be the one which best reflects the U235 depletion profile in the fuel plate.

Figure Nº 2 shows the Cs137 activity axial profile for both plates, normalised to the maximum value. The meat position along the length of the plate can be clearly seen, as well as a very significant effect of active material accumulation done during the manufacturing of the plate, “dog boning effect” at the both plate ends, which is shown by large activity peaks. It is consistent with the maximum area loading of about 28 % estimated for the manufacturing process of these plates. In other hand, the fact that this effect could be detected in a very narrow zone, indicates the effectiveness of the collimating arrangement.

The Cs137 activity profile are the main guide for the selection of the places where the puncturing will be performed, to obtain samples for chemical burnup analysis, and SEM and metallographic observations.

Figure Nº 3 shows the Cs137 activity transverse profile at maximum axial activity for both the inner and the outer plates, in comparative counts for the same counting conditions. The end peak effect caused by the different neutron thermal flux into the assembly box, from the edge to the centre of each plate and from the outer to the inner plate, is clearly noticed.

4. DESTRUCTIVE ANALYSIS

4.1. Sampling procedure

Appropriate side of samples for metallographic and SEM observations and chemical burnup analysis were obtained with four puncturing dies driven by a manual hydraulic pump. Samples of 2 mm diameter are used for SEM observations, 8x15 mm rectangular samples are used for metallographic examination by optical microscopy and 4x4 mm samples are bound for chemical burnup analysis.

4.2. Metallographic examination

Samples were obtained from the different part of the inner plate No 8, in order to examine the fuel microstructure and cladding oxide thickness. The samples were mounted in low viscosity epoxy resin in ad-hoc aluminium test tube to allows the handling operation and positioning. Semi-automatic commercial polishing machine were adapt to operate through the telemanipulator of the cells. The samples were ground 1.5 mm deep and polished with up to 0.25 μm diamond paste. Micrographs were taken of the as-polished surface.

4.2.1. Cladding oxide thickness:

Samples from the midcenter of the plate and midside of the plate (edges of the meat zone) as wells as in the bottom frame part of the plate were examined for the cladding oxide layer growing.

The table 3 summarizes the results for the inner plate No 8, which was irradiated during 300 effective power day. The quality of the primary system water was maintained into the required range of operation, it was conductivity less than 1 μS/cm and pH 5,8 to 6,5.

	Table 2: Oxide layer thickness (Average to maximum values in μm)
midcenter of the plate	15 to 20
midside of the plate	10 to 15
bottom frame	5 to 10

Clearly at mid center of the plate the oxide layer was the thickest where plate reach the maximum power density of about 30 Watt/cm^2 and the maximum cladding surface temperature aprox. 100 °C. Figure No 4 shows cross-sectional views of the oxide layer observed at midcenter of the plate and in the bottom part of the unfuelled frame.

Non-destructive measurements of the thickness of the oxide layer in other plate were also done by using Eddy Current coil probe with a commercial specific equipment for the measurement of non-conductive coating over non-ferrous metal substrate. Primary the calibration curve was obtained using calibration foils over Aluminum fine plate and then it was corroborated with the results from the micrographs of plate 8. Several plate tested for this methods show the same range of values, 10 to 20 μm in the meat zone, no different develop the oxide layer thickness was observed between the inner and outer plates. This non-destructive methods is useful to follow the oxide layer growing during irradiation of the fuel element by underwater measurement at reactor pool.

4.2.2. Fuel microstructure.

The main objective was develop the methodology for the polishing of the sample and measurement of the interaction layer between the aluminium matrix and fuel particle. At the maximum burnup location, the typical thickness of interaction layer was 1.5 μm. Taken in account low Bu and thermal density of power, the temperature gradient in the cross section of the plate is rather small, and thus, the interaction zone is rather uniform in any position of the meat and primarily only a function of burn up. See Figure No 5 shows micrographs from different part of the inner plate 8.

The fuel particule swelling was determined by measuring the residual, as fabricated porosity on the micrographs, and from the immersion volume values mentioned in Section 3.3. A value of approximately 15% $\Delta V^F/V_o^F$ at the peak burnup location of plate No 8 corresponding fission density (FD) on fuel of 1.5 x 10^{27} fis.m^{-3}, was thus found and is compared with previous experimental swelling data on U_3Si_X [4] and [5].

4.3. SEM observation

Samples for examination of fuel fracture surfaces by Scanning Electron Microscopy were obtained from the outer plate No 19 at the mid-center corresponding the maximum Bu and power of the element and in the bottom fuelled part of the plate away from the dog bone area. The entire sample was split in two halves by a micro nippers, obtaining a clean fracture surface of the fuel particles. The sample at midcenter where the local Bu was 26 % U235 depleted (FD: 1.7 x 10^{27} fis.m^{-3}) shows small fission gas bubbles in the fuel particles. Typically the bubbles are 0.1 to 0.3 μm in diameter and the number density of bubbles in most of the fuel particles was low to negligible in other particles. This particle to particle variability in visible gas bubbles is likely due to the presence of two different phases (U_3Si and U_3Si_2) in this P04 fuel compound. This is further shown in Figure 5, where the bubbles appear to be confined to the lighter (more dense) phase in the particles, this being U_3Si.

No observable gas bubbles in the bottom sample of the plate where the local Bu was 15.6% (FD: 1.1 x 10^{27} fis.m^{-3}).

Figure No 6 shows the views obtained by SEM.

4.4. Chemical Burnup determination

Seven equi-spaced 4x4 mm samples are bound for chemical burnup (Bu) analysis along the axial center line and close the meat edge of both the inner plate No 9 and the outer plate No 19.
The purpose is to evaluate the Bu distribution along the entire fuel element, and to have the best estimation of the average to compare which the Bu value calculated with the fuel management code used in the RA-3 reactor.

The samples were processed by the acid dissolution and ion-chromatographic separation [6]. The isotopic analysis was performed on an appropriate aliquote of the solution . The termo-ionization mass spectrometric were used [7], calibrated with U235 15% enriched standard (U-150 NIST). Isotopic composition of U was determined to evaluate the local depletion of U235 as Bu indicator.
Figure No 7 shows the Bu distribution in both plates. A differences of 22% in U235 local depletion was found between the midcenter samples of the middle inner and outer plates. A similar relative difference was observed with the edge of the meat. These variation show the influence of the neutron thermalization distribution in the RA-3 reactor fuel.

For same of the sample the Nd148, total U and Pu concentration were also analysed, so as to obtain to exact evaluation of the percentage of the fissioned heavy metal (HM) respect to the total preirradiated HM.

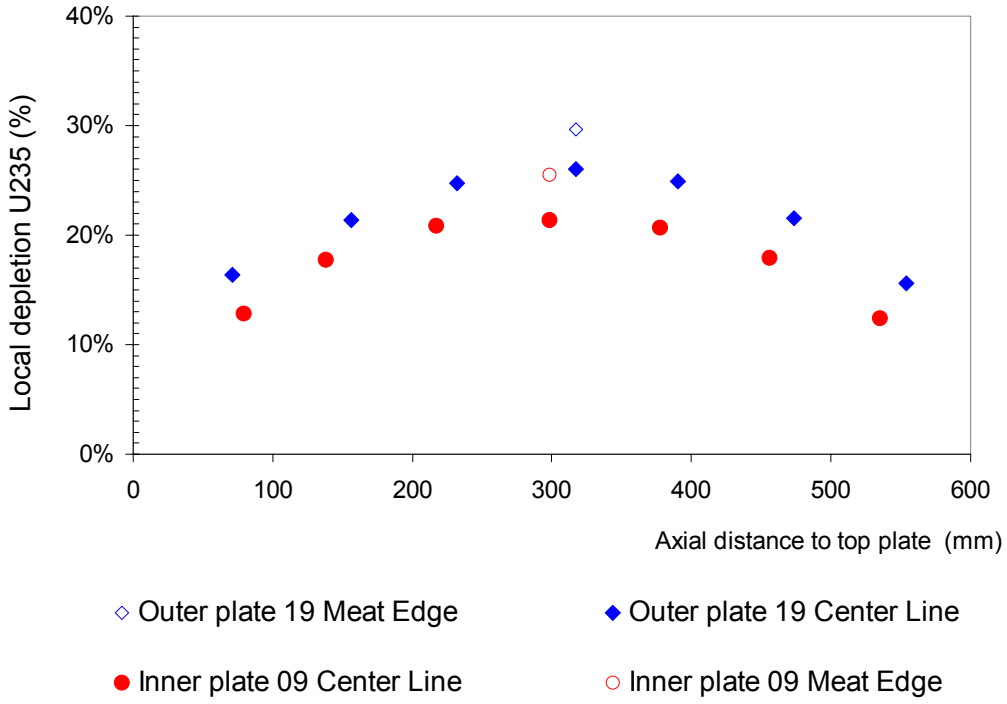

Figure No 7 Bu distribution

5. FINAL CONSIDERATION

A complete set of standard PIE techniques for MTR fuel plate were set up in ours hot cells following the methodology performed in Argonne National Laboratory. The implementation and achievement of this PIE plan was performed in the framework of the Implementing Arrangement for Technical Exchange and Cooperation in the Area of Peaceful Uses of Nuclear Energy between the Department of Energy of the United States of America and the National Atomic Energy Commission of the Argentine Republic, through a joint project with Argonne National Laboratory.

Non destructive and destructive tests included microstructural observations and chemical burn-up analyses were performed on the first full scale prototype P-04 of Si-based LEU, manufactured and irradiated in Argentina. The results obtained show a behaviour consistent with that of other fuel elements of the same kind, tested previously [2].

Much experience at CNEA was acquired on the behaviour of this fuel material and inthe PIE techniques. In this particular case data of fuel material contributed to the overall database of Si-based LEU fuel at low fission rate.

This work contributed to CNEA´s qualification program as a supplier of low enriched Al-U_3Si_2 dispersion fuel elements for research reactors

This work also contributed to IAEA Regional Program on the characterization of MTR spent fuel especially in Burn-up analysis and corrosion behaviour of the fuel plates.

6. REFERENCES

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[7] Análisis de Espectrometría de Masas muestras P0-4. Informe interno, octubre 2002 UA Química; Alfredo Lobo, Ricardo Garavaglia, Eduardo Gautier.

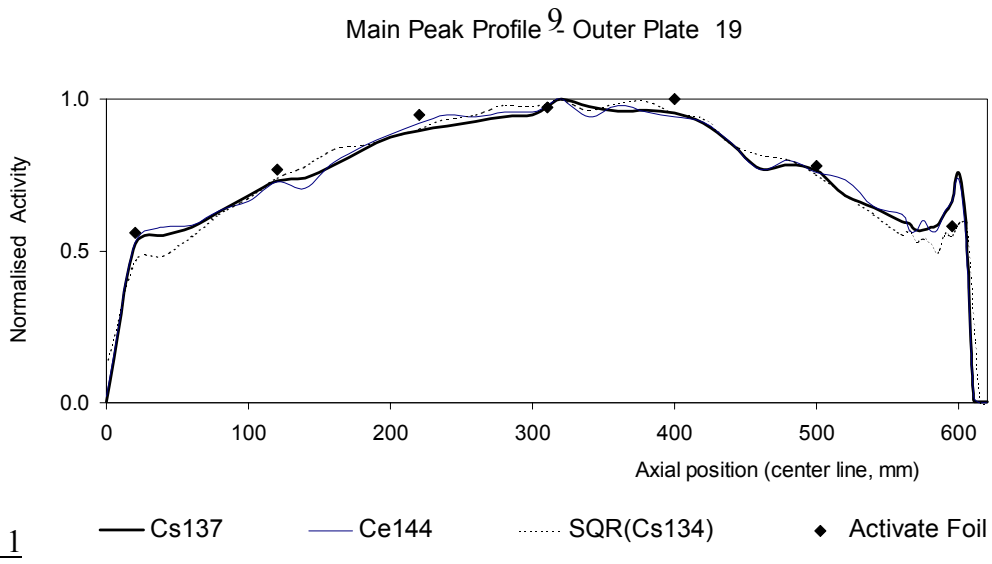


Figure 1

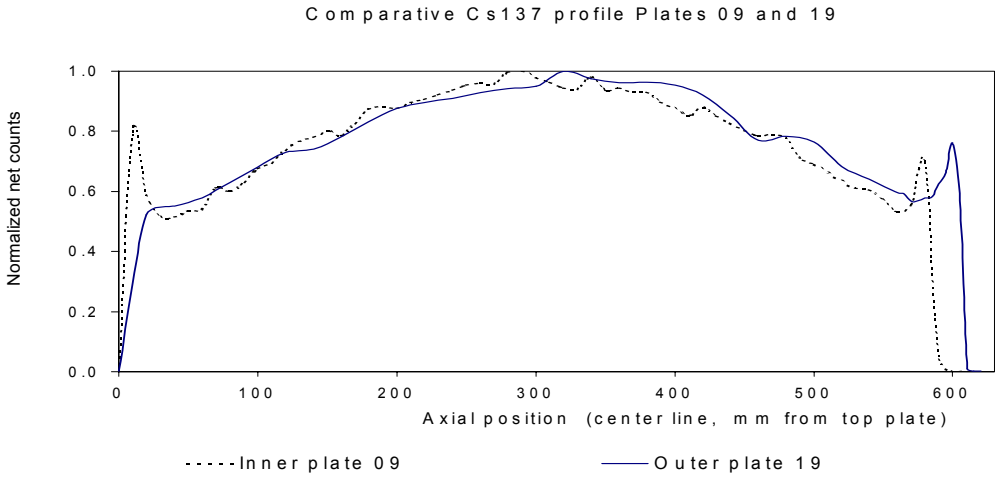


Figure 2

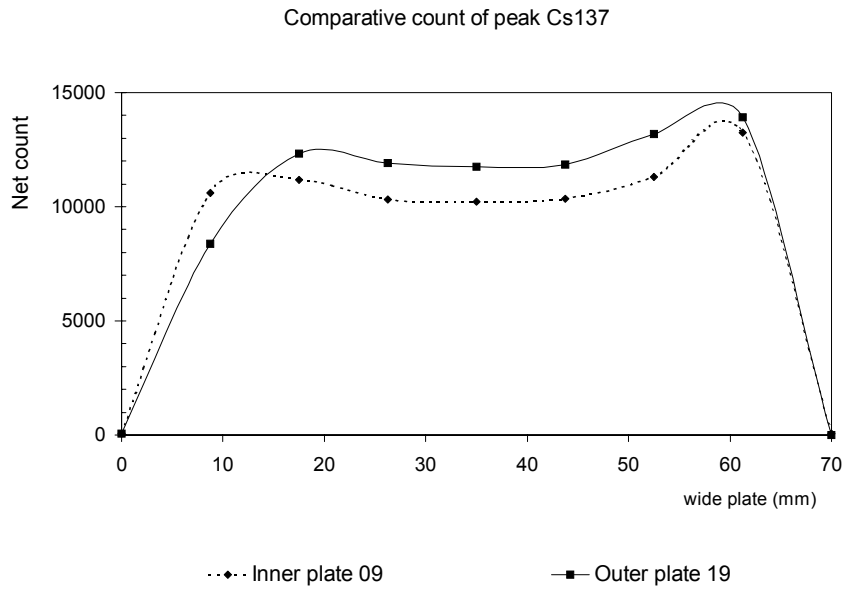
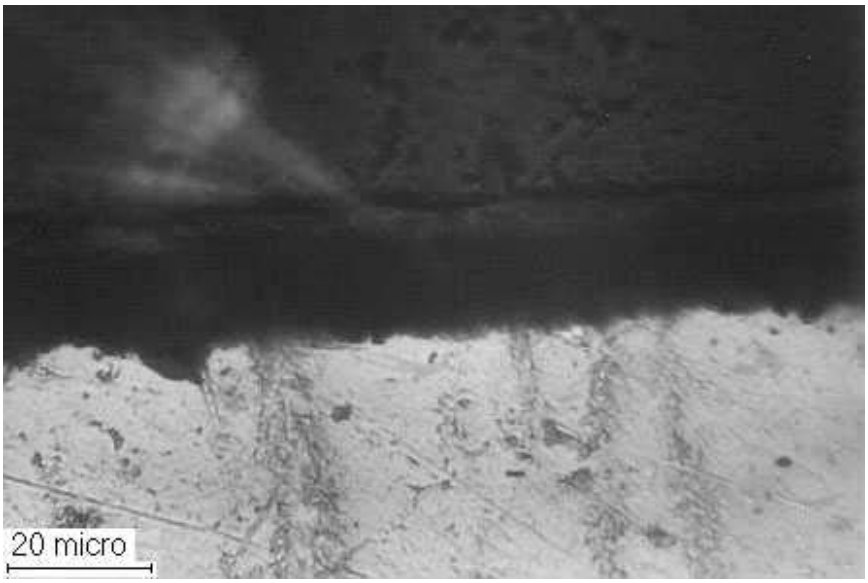
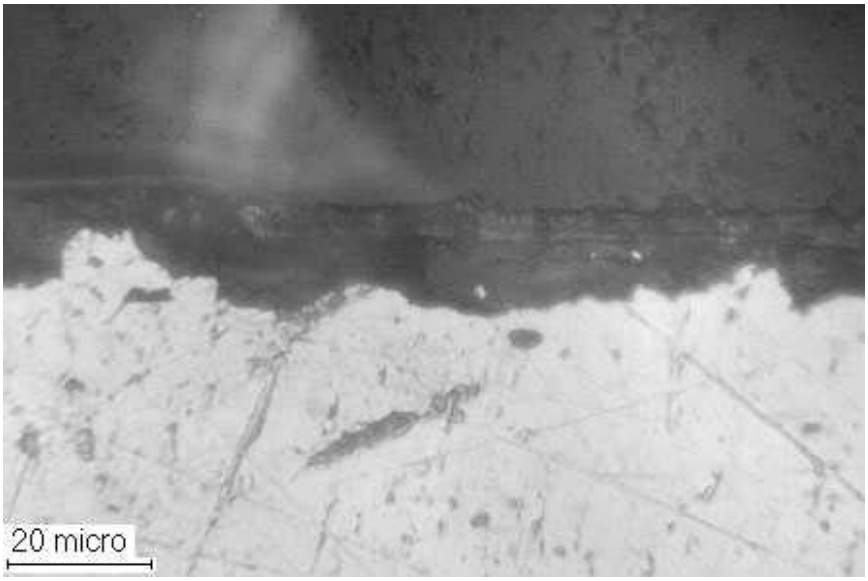


Figure 3

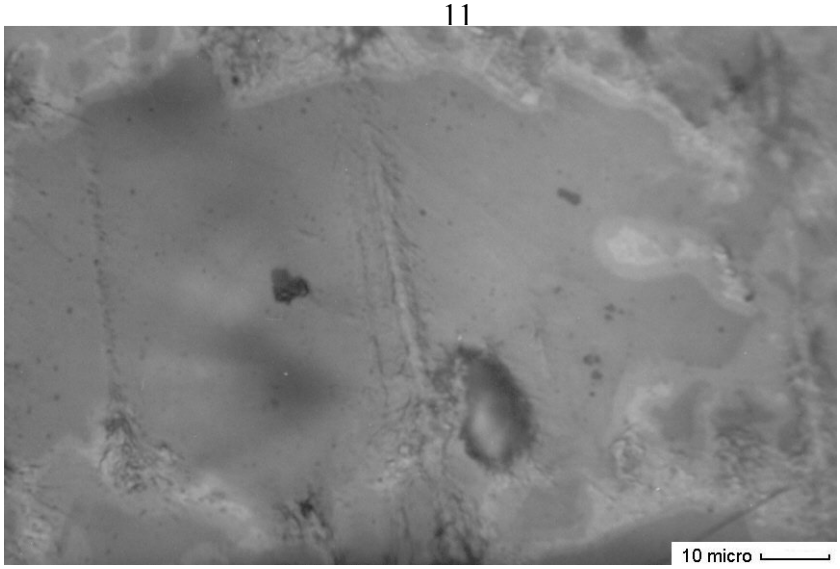


(a)

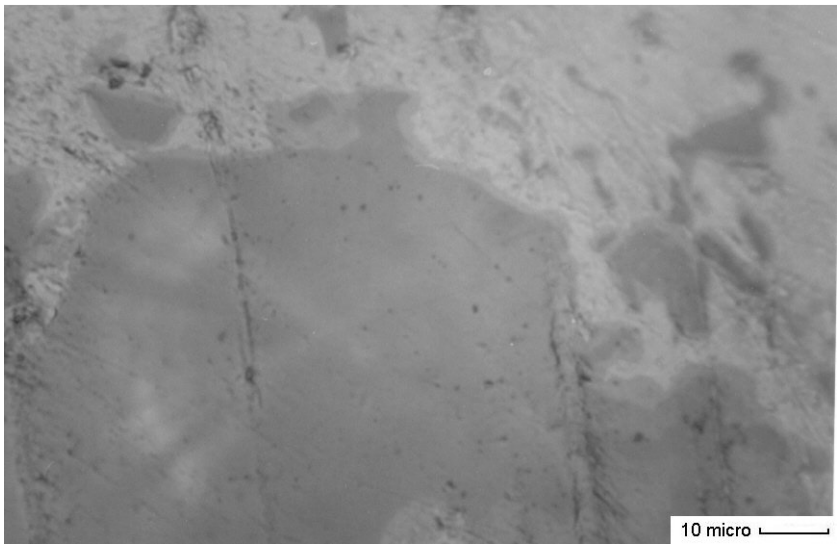


(b)

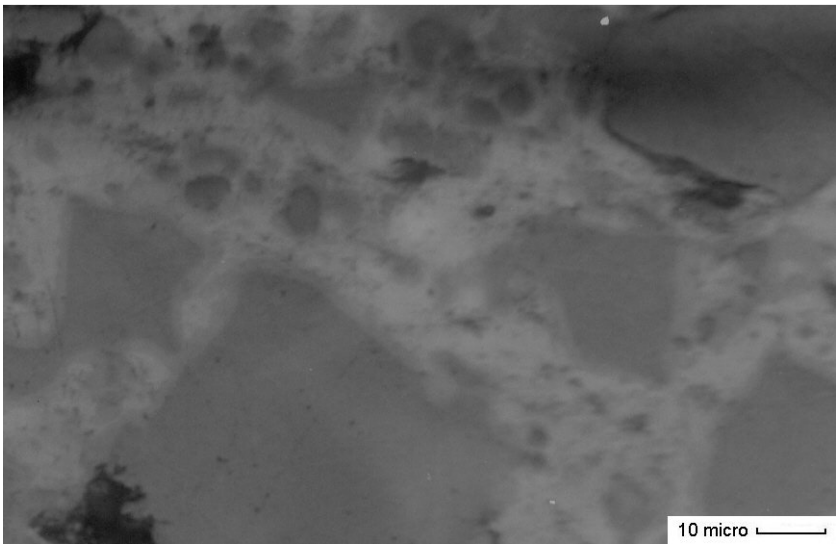
Figure No 4: cross-sectional views of the oxide layer observed at midcenter of the plate (a) and in the bottom part of the unfuelled frame (b).



Mid-center of the plate, center of the meat

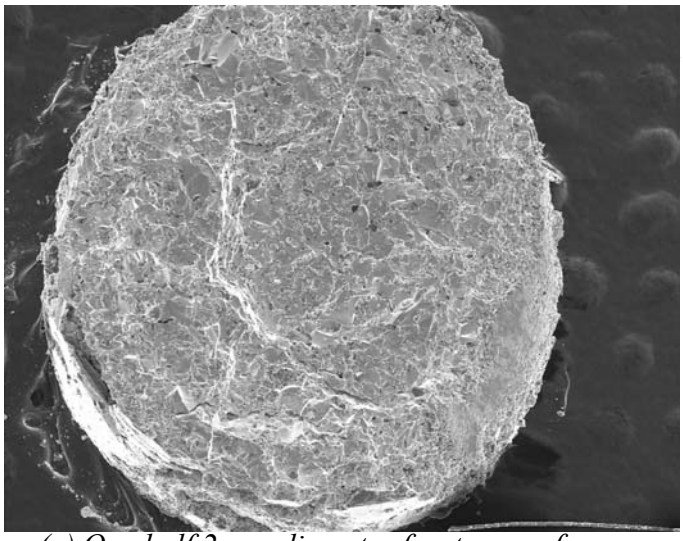


Mid-center of the plate, edge of the meat

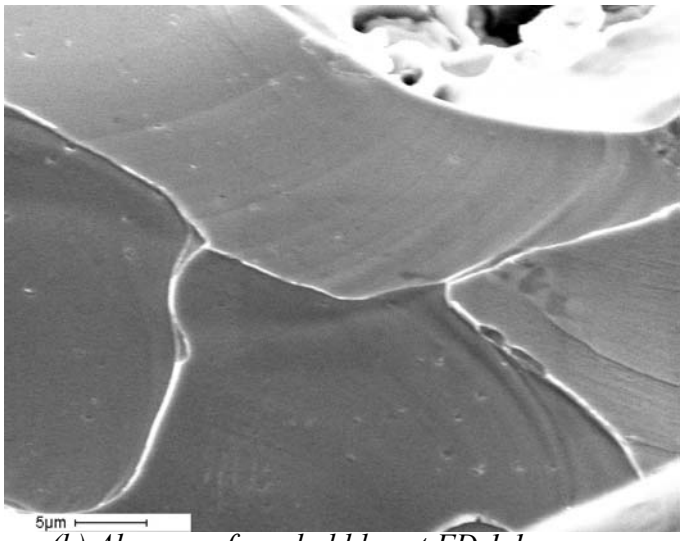


Mid-edge of the plate, center of the meat

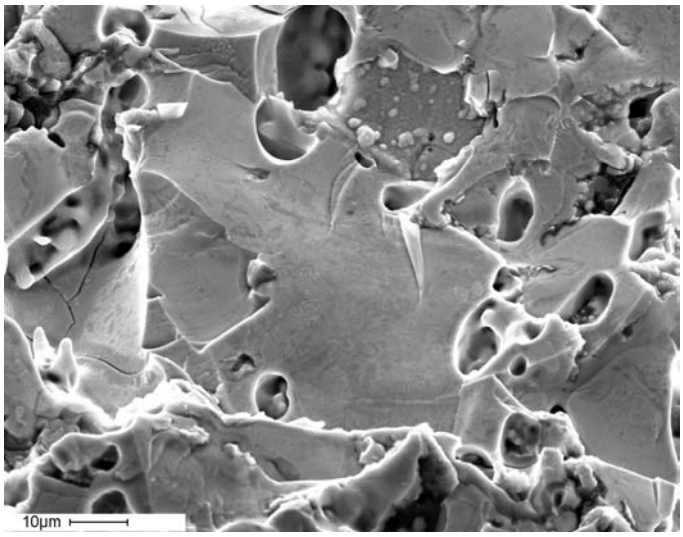
Figure N° 5 Micrographs of inner plate No 8.



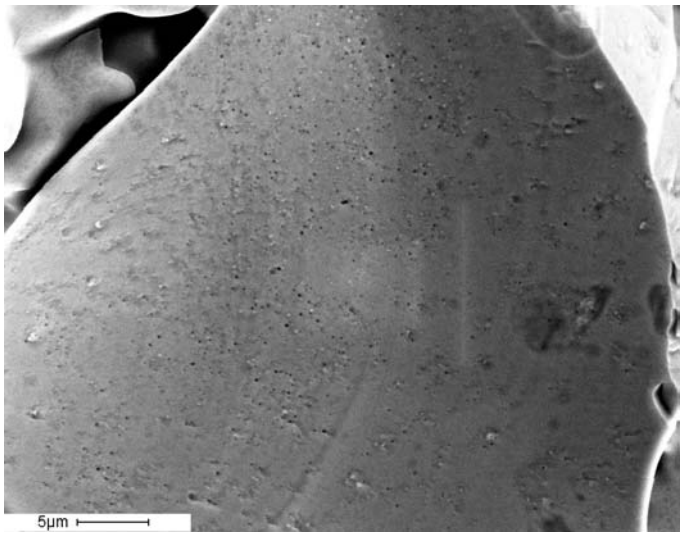
(a) One half 2 mm diameter fracture surface sample



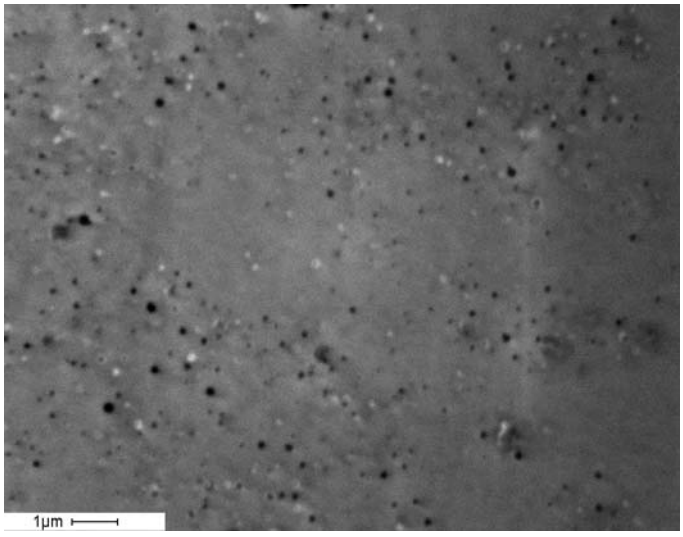
(b) Absence of gas bubbles at FD 1.1 x 10^{27} f.m^{-}

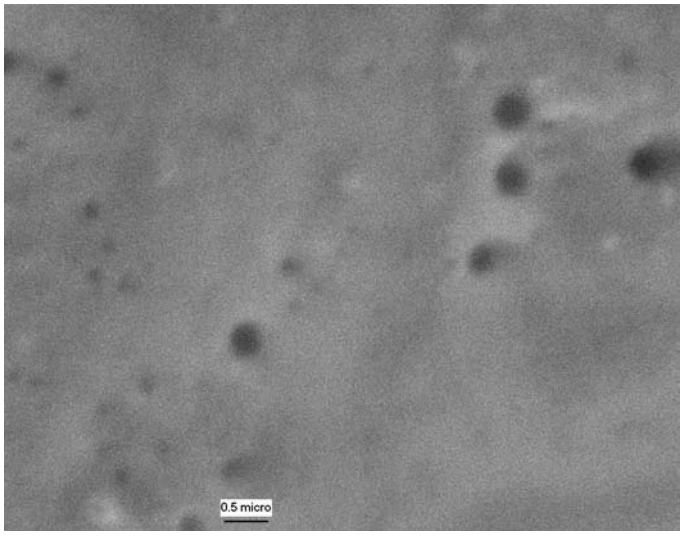


(c)Meat microstructure showing rounded as-fabricated porosity



(d)



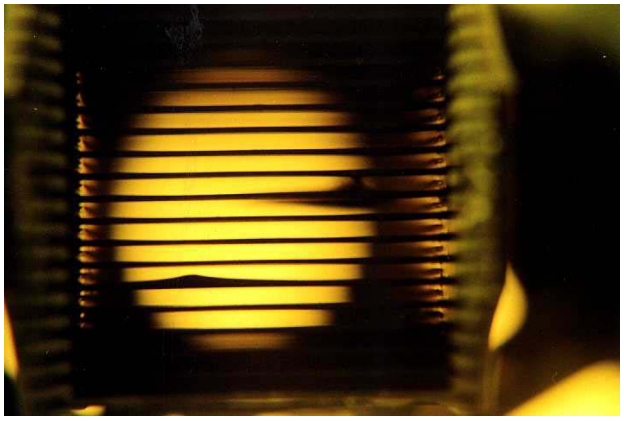


(d) (e) and (f) Bubbles morphology at FD 1.7 x 10^{27} f.m^{-3}.

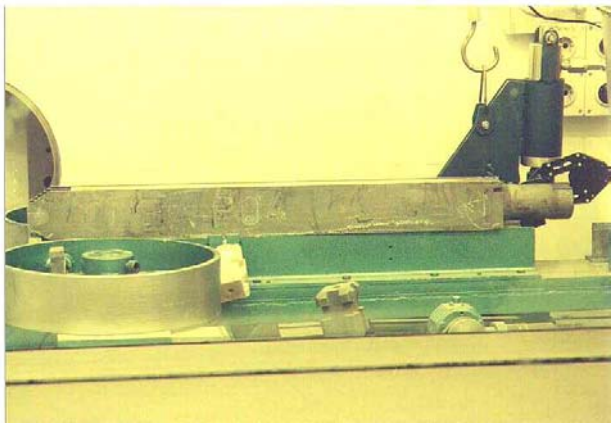
Figure No 6 SEM micrographs obtained at mid center and bottom position of the outer plate No 19

Photograph 1: during arriving of P-04 to the hot cell at Ezeiza Atomic Centre

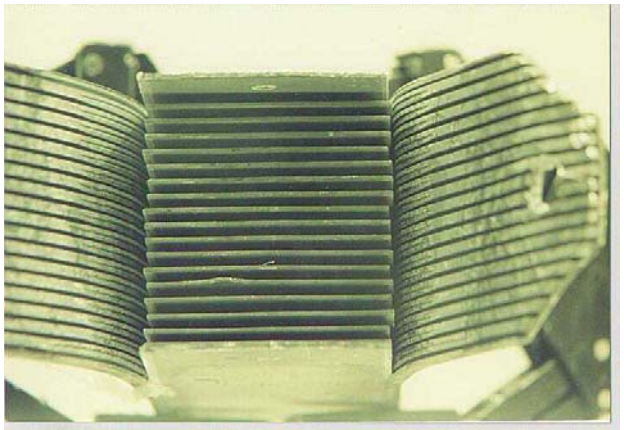
Photographs 2: external state of P04.



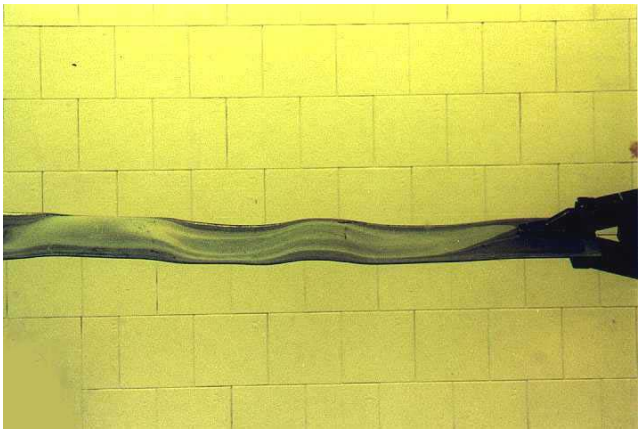
Photograph 3: view through the coolant channels alter the removal of the handling bar.
Defective blister in plate Nro 10.



Photograph 4: machine for dismantling of the fuel plates by rolling of side plates.



Photograph 5 : P04 during the beginning of the side plate rolling operation



Photograph 6: Outer plate after blister test up to 575 ºC
The meat zone suffered large deformation without blister formation.