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IRRADIATION OF PLUTONIUN FUELLED COATED PARTICLES IN FRJ 2

by

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(*) Kernforschungsanlage Jülich GmbH
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(****) EURATOM, delegated to KFA Jülich

1974

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Work sponsored by the Joint Plutonium Group formed by BelgoNucléaire S.A., Bruxelles - Belgium Centre d'Etude de l'Energie Nucléaire (S.C.K./C.E.N.), Bruxelles - Belgium and the Association Euratom/BBK/KFA

Association No 003-63-1 RGAD

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ABSTRACT

Two types of plutonium oxide fuelled coated particles with carbon diluted 550 μm-kernels and dense 250 μm-kernels were irradiated for 45.4 full power days in FRJ 2 (Jülich) at about 1 100° C. A burn-up of about 18% FIMA and a fast dose of less than 10^{20}n/cm^{-2}DNE were accumulated. Postirradiation fission gas measurements indicated that no particle was broken. The metallographic examinations show different behaviour of the inner pyrocarbon layers due to different plutonium migration into buffer layers during the coating process. Only small plutonium migration during irradiation could be found which has been stopped by the SiC layer.

KEYWORDS

COATED FUEL PARTICLES
PLUTONIUM OXIDES
COATINGS
SILICON CARBIDES
PYROLYTIC CARBON
SAMPLE PREPARATION
IRRADIATION
BURNUP
RADIATION DOSES
VERY HIGH TEMPERATURE
RADIATION EFFECTS
FISSION PRODUCTS
GASES
METALLOGRAPHS
PLUTONUIM
DIFFUSION

IRRADIATION OF PLUTONIUM FUELLED COATED PARTICLES IN FRJ-2

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BN	BELGONUCLEAIRE S.A. - Mol and Brussels.
C E N / S C K	Centre d'Etude de l'Energie Nucléaire Studie Centrum voor Kernenergie - Mol.
C N E N	Comitato Nazionale per l'Energia Nucleare - Rome.
K F A	Kernforschungsanlage Jülich GmbH - Jülich.
TU	C C E - Inst. für Transurane - C C R Karlsruhe.
*	Seconded by EURATOM to K F A Jülich.

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1.- INTRODUCTION.

The prospective plutonium utilisations were summarized in a recent contribution to the Working Group on Plutonium Recycle in Thermal Reactors [1].

Basically, the plutonium finds its best utilisation in the HTR's operating on the thorium cycle. In that cycle, the feed plutonium fuel will be subjected to a high burn-up and therefore will not be worth recovering. Since the aqueous reprocessing by the known techniques is complicated when plutonium is present, it is advantageous to separate the thorium breed particles from the plutonium feed particles so that the head-end of the reprocessing operation can mechanically screen the plutonium fuel to be discarded from the Th - U 233 fuel to be recuperated.

In the HTR's working on the low enriched uranium cycle, it is also advantageous to segregate the plutonium particles from the uranium ones, in order to reduce the economic penalty inherent to handling plutonium. A further but minor advantage is to enable to recuperate only the fresh plutonium bred in the uranium particles from the depleted plutonium having essentially no commercial value due to its high burn-up and its isotopic degradation. The inconvenience in that cycle is to insure a proper distribution of the plutonium particles in order to reduce the technological peaking factor to acceptable levels. It is therefore not certain that in a low enriched uranium cycle the segregated fuel will be the best solution.

* * *

2.- PREVIOUS BACKGROUND.

Feed plutonium particles have been developed at Mol, partially under the sponsorship of the DRAGON Project, since 1964. The necessary voidage for the fission gases and the carbon monoxide expansion was provided mainly by the porosity of the kernel. Due to the high burn-up envisaged (600,000 MWd/te.Pu corresponding to 65 % FIMA), it was indeed necessary to dilute the fissile material in a support enabling to accumulate the voidage.

As in the driver fuel utilized in the DRAGON reactor, carbon was chosen as diluent. However, the attempts to manufacture a kernel consisting of a dispersion of a plutonium carbide in a carbon matrix, according to the concept retained by DRAGON at that time, had to be abandoned due to the high losses of fissile material experienced in the carbochemic reduction process [2].

The development was then focused towards a dispersion of oxide within the carbon matrix, based on the equilibrium of the two materials under a certain CO pressure, as had been developed in Mol earlier [3]. This development lead to acceptable fuel [4][5] which was irradiated in the R2 reactor at Studsvik - Sweden, to extreme burn-ups of 200,000 MWd/t and 360,000 MWd/t at temperatures of 1850°C and 1200°C respectively [6].

Another test was carried out in the Second Charge of the DRAGON Reactor Experiment during the period, January 1967 to March 1968, when the central rods of two DRAGON fuel elements were constituted of plutonium fuel [7]. The plutonium coated particle fuel was placed loose inside a "telephone dial" arrangement of holes in graphite boxes. The number and size of holes were adjusted to give a constant fissile plutonium loading in each box using three batches of particles to deliberately vary the composition (Table I). The average burn-up at the end of the irradiation (equivalent to 224 days at full power) was estimated to be 600,000 MWd/t M and the fast fluence 1×10^{21} n/cm^2 at 700 - 1100°C. The value of R/B for Xe^{133}, Kr^{85m} and Xe^{135} as measured via the helium purge for the two fuel elements, remained between 10^{-6} and 10^{-5} ; these values indicated that there was no damage to the particles. Visual and ceramographic examinations of the particles after irradiation confirmed this.

3.- PURPOSE OF THE FRJ2 EXPERIMENT.

The good irradiation behaviour of the diluted kernels lead to the desire of improving the utilization of the plutonium fuel in a thorium cycle. In order to minimize the age factor, the plutonium should be concentrated. Therefore, it was decided to mount an irradiation to compare the behaviour of the following fuels :

- "diluted fuel" of the type tested earlier but with a kernel diameter of 550 microns (instead of 300) and the same carbon dilution. This already gives a higher plutonium self-shielding than the previous generation of particles ;
- "undiluted fuel" consisting of particles containing the same amount of plutonium into the same shell but with all the fissile material concentrated in a small kernel of pure plutonium oxide. The voidage and the difference in dimension of the kernel are compensated by a thicker buffer layer to produce approximately the same internal voidage expressed as total void per gram of fissile plutonium.

This report describes in detail a comparative irradiation of those two types of particles.

The diluted fuel has been manufactured completely by BN - C E N ; the undiluted kernels were manufactured according to a Sol-Gel technique by C C R - Karlsruhe and to the S N A M process by the C N E N (La Casaccia) ; BELGONUCLEAIRE seeked the collaboration of those two organizations in order to perform the experiment on a product that would be representative of potential industrial processes ; both organizations kindly accepted to collaborate to the experiment by manufacturing the necessary kernels. All the coating work was performed by BN - S C K / C E N.

For sake of simplicity, the terminology utilized throughout the present report will be :

"D" fuel, for the diluted fuel ;

"T" fuel, for the batch of Sol-Gel fuel ;

"C" fuel, for the batch of S N A M fuel.

Table I gives the main characteristics of the plutonium particles irradiated up to now and of the irradiations in which they have been tested.

* * *

4.- PREPARATION OF THE PLUTONIUM OXIDE KERNELS.

The C and T kernels were made by Sol-Gel process, while the D kernels were elaborated by the powder agglomeration process. Both techniques are able to produce both types of kernels [8] ; the choice of techniques to be applied for each fuel specification was based on the following reasons :

- the small concentrated kernels of pure plutonium oxide (300 μ) are difficult to manufacture to a good shape specification by the powder agglomeration technique ;
- the diluted kernels are more fragile when fabricated by a Sol-Gel technique than by a powder agglomeration technique.

The isotopic composition of the plutonium batches utilized is given in Table II.

The preparation of the different kernels can be summarized as follows :

4.1. C C R - KARLSRUHE SOL-GEL PREPARATION (T FUEL).

The process, which is described in detail in several reports [9][10], consists of the following steps : precipitation of $Pu(OH)_4$ from a $Pu(NO_3)_4$ solution, by adding this solution to an ammonia solution ; peptisation of the washed $Pu(OH)_4$ with HNO_3 at 85 °C to form a plutonium sol approx. 1.5 M. These operations must be carried out under severe control of pH. The sol is converted into droplets which are gelled by dehydration in 2 ethylhexanol at room temperature. This method for the production of microspheres is based on that developed at Oak Ridge National Laboratory which involves the water extraction of a sol low in nitrate with a long chain alcohol. The gelled kernels are washed in a suitable organic medium, dried slowly, calcined 3 hours at 950°C, and finally reduced 4 hours in hydrogen at temperature between 1600 - 1720°C to get dense plutonium oxide kernels with an $O/Pu < 2$ (mixture of PuO_2 and Pu_2O_3). One hundred grams of microspheres resulting from several batches have been produced for coating by this method. Under metallographic examination the kernels show cracks (Fig. 1). The heating schedule of green particles and the properties of sintered kernels are given respectively in Tables III and IV and the crystallographic density of the substoechiometric oxide was reported by C C R - Karlsruhe as 10.9 g/cm^3.

4.2. C N E N / S N A M SOL-GEL PREPARATION (C FUEL).

The 71 g PuO_2 particles were prepared at the Plutonium Laboratories of C N E N by a Sol-Gel process, which can be summarized by the following steps :

- Preparation of an oxide hydrosol solution :

by amine (Primene JMT) solvent extraction of nitric acid on freshly prepared Pu(IV) nitrate solution. 0.2 M solutions of $Pu(NO_3)_4$ are solvent extracted to get a NO_3^- / Pu ratio $\simeq$ 1. The diluted sol is finally concentrated to 2 M by evaporation. This oxide hydrosol is mixed with tetrahydrofurfuryl-alcohol and an organic polymetric thickner.

- Conversion of the colloidal hydrosol into gel spherical particles :

gelation is achieved by dripping the colloidal hydrosol into an ammonium hydrate solution and gelified spherical particles are washed with ammonium hydrate and deionized water. Green particles are then dried by azotropic distillation.

Organic material introduced in previous preparation steps is eliminated at this stage by firing at 450 - 500°C in air.

- High temperature treatment :

Firing of the gel product to the final high density oxide is carried out in furnace under an appropriate atmosphere (usually 4 v/o hydrogen in argon) according to a standard temperature cycle up to 1200°C.

Higher temperature and hydrogen concentrations can be used if an hypostoichiometric oxide is desired [11] [12].

As the first coating was done at 1450°C, these microspheres would not have withstood the dropping in the coating furnace at 1450°C. Therefore a further heat treatment has been made up to 1450°C in carbon monoxide atmosphere at the plutonium facilities of the joint C E N - BN Plutonium Group in Mol (see table III). The heating rate was 200°C per hour while the cooling down rate was 150°C per hour.

4.3. DILUTED KERNEL PREPARATION (D FUEL).

The procedure which has been described earlier [6] can be summarized as follows : the plutonium oxide powder is ground in a planetary mill in an agate pot, sieved (44 µm sieve), mixed with "United MT" carbon black ground again in a planetary mill and sieved (210 µm sieve). The mixture PuO_2 / C is blended with furfurly alcohol [10 cm^3 / 100 g (PuO_2 + C)] in a hand mortar and then crumbed by passing it through a 297 µm aperture sieve. The PuO_2 / C green crumbs are spheroidized in an aluminium pot rotated on a planetary mill and sieved (- 350 µm + 297 µm) and finally hardened by exposing them to formic acid vapour for 24 hours. The heat treatment is given in Table III. A typical cross-section of the fuel is given in Fig. 1.

* * *

5.- COATING OF THE KERNEL.

The coating of the three batches is based on a simplified "triso" concept PyC - SiC - PyC consisting of a porous pyrocarbon ("PyC") buffer layer, a dense inner PyC layer, a SiC layer and a dense isotropic PyC. Only the thickness of the layers varied (Table X) corresponding to the two different particle designs.

The coating furnace is a 40 mm diameter graphite resistance coater, incorporated in the facility described earlier [4].

All the details of produeing this layers are given in Table V.

5.1. POROUS PyC LAYER.

The porosity should provide the voidage necessary for the fission products accommodation ; the thickness chosen was determined by the rule "the free volume (voidage) should be 2 - 3 % of the fuel kernel volume for each atomic percent burn-up" [13]. This coating should also prevent mechanical stress resulting from the interaction between kernel and coating during irradiation. Finally, to prevent damage of the structural layer by the fission product recoil, a buffer carbon layer of 2.82 mg/cm^2 is required. Fig. 2 gives, as a function of the buffer density, the minimum buffer thickness to fulfil this requirement. A 150 % of this minimum thickness is used to avoid, for a non perfectly spherical kernel, a too thin buffer in some direction and to compensate for the thickness dispersion within a batch ; this type of coating is indeed produced within a few minutes in the coating apparatus.

In order to get the same voidage in the coated particles both for the diluted and undiluted fuels it was necessary to raise the buffer thickness from 30 mierons for the diluted to 160 mierons for the undiluted fuel.

5.2. DENSE INNER PyC LAYER (SEALING LAYER).

This coating is a vessel for the CO gas in equilibrium with the system oxide - carbon and a barrier for the diffusion of HCl gas produced during the pyrolysis of trimethylchlorosilane ($CH_3 - Si - Cl_3$) to form the SiC (next coating).

5.3. SiC LAYER.

This coating is the diffusion barrier for the metallic fission products and is partially the pressure vessel.

In order to avoid excessive stresses due to differential thermal expansion during fabrication and initial irradiation, the SiC layer has further been surrounded by two flash layers (of a few μm) of porous PyC to compensate differential thermal expansion and prevent crack propagation.

5.4. HDI (HIGH DENSITY ISOTROPIC) PyC LAYER.

This last layer is also part of the vessel of the fuel particle ; it compresses the SiC layer during irradiation, by its fast dose induced shrinkage.

* * *

6.- CHARACTERIZATION OF THE COATED PARTICLES.

Due to the small amount of material fabricated, a large scatter has been observed in the analytical results of the particles. They are deemed in most cases to be representative of sampling effects. In some circumstances however the difference between the results of the different samples cannot be explained by a scatter in the properties of the product. For instance, the sintered "T" kernels were analysed both at Karlsruhe and Mol. The results are presented in Table IV. It can be seen that the analytical data are consistently different. It might however indicate in this case an oxidation reaction of the kernels during the transport. Another example of the scatter in the data is given by the plutonium contents in Table VI.

The O/M ratio deduced from the relative intensity of two diffraction X-ray is characteristic respectively of the sesquioxide and the dioxide. The evolution apparent in Table VIII is probably realistic as a tendency. The scatter around this general tendency of each successive step is not representative of the average of the batch but due to the small sample size involved in the analytical technique.

The densities were measured both as bulk density and immersion density (Table VII). Here again a general tendency can be observed but the scatter of the individual values is not realistic. The layer density deduced from the particle density is obtained by difference and therefore not accurate. For what the density of the SiC is concerned, it includes the two flash layers deposited in order to protect the coating from differential thermal expansion effects as mentioned in paragraph 5.3. The contamination of the particle is very low (Table IX) and tends to decrease as the coating proceeds.

The coating thicknesses given in Table X are the results of cross-check between dimensional measurements and metallographic observations. Finally, Table XI summarizes the accuracy claimed for in various analytical results.

Fig. 3, 4 and 5 show respectively metallographic sections of particles D, T and C after each coating step. For C type, a bulk plutonium diffusion has appeared after PyC deposition ; this is also confirmed by the alpha autoradiography and polished section presented at Fig. 6. In the T type of fuel, the plutonium migration was limited to plutonium concentration spots in the buffer layer ; this was visible on the metallographic sections. Table XII indicates the extent of the plutonium displacements, measured by X-radiography (Cf. paragraph 8.6.) on 15 particles of each batch ; by comparison with Table X, it can be seen that the plutonium migration proceeds to the dense inner PyC.

* * *

7.- IRRADIATION.

7.1. IRRADIATION CAPSULE.

The samples were irradiated as loose coated particles in 8 small graphite (material : Ringsdorf graphite type Kr 66) containers of 12 mm diameter which were deposited in a telephone dial type graphite (material : Ringsdorf graphite type Kr 60) cartridge of 74 mm diameter (Fig. 7). The graphite cartridge was inserted into a sealed stainless steel capsule which was enclosed in the irradiation rig JK 4 (Fig. 8). The rig was of LV-4 type normally used for irradiation of five spherical fuel elements of 6 cm diameter. In this case, the plutonium particle capsule was inserted instead of the lower fuel ball.

Three Ni / CrNi thermocouples had been put into small holes at the outer circumference of the cartridge. Another thermocouple was located at the outer stainless steel wall.

7.2. CAPSULE LOADING.

Each of the 8 containers was filled with one of the three particle types. Additionally, five individual radiographed particles of each batch had been filled into the centre hole of container 1. The particle quantities filled into the containers are given in Table XIII and Fig. 7.

7.3. IRRADIATION CONDITIONS.

The rig JK 4 has been irradiated in FRJ2 (DIDO) Jülich in position D 1 for three reactor periods from January 27, 1970 till April 1st, 1970. The total irradiation time was 45.4 full power days at 15 W.

(1) Temperatures.

The designed temperature profile is shown in Fig. 9. However, the experimental temperatures remained about 150°C below the designed ones. Fig. 10 shows the temperature history during the three reactor periods. The graphite temperatures were measured by three thermocouples ; from this, the central temperature was calculated using the design temperature profile.

(2) Burn-up.

The axial profile of the perturbated thermal neutron flux in position D1 is given in Fig. 11. The mean thermal flux in the plutonium particle - capsule was :

0.92×10^{14} $cm^{-2}sec^{-1}$ during the first period
0.875×10^{14} $cm^{-2}sec^{-1}$ during the second period
0.76×10^{14} $cm^{-2}sec^{-1}$ during the third period [12]

Using these flux values the accumulated burn-up has been calculated (Fig. 12). At the end of irradiation, the following burn-up values have been achieved :

Particle type	Burn-up (% FIMA)
D Fuel	17.0
T Fuel	18.7
C Fuel	18.3

(3) Particle rating.

The initial rating per coated particle has been calculated using the mean particle data given in Table XIII and the plutonium composition given in Table II.

Particle type	Initial rating (W)
D Fuel	0.54
T Fuel	0.36
C Fuel	0.32

(4) Fast neutron dose.

The fast neutron flux ($E > 1$ MeV) monitored by nickel-wires in position D1, - 30 cm from core centre, is about 1.35×10^{13} $cm^{-2}sec^{-1}$. From that, the accumulated fast neutron dose has been calculated to be about 5.3×10^{19} cm^{-2} (DIDO Nickel Dose).

* * *

8.- POSTIRRADIATION EVALUATIONS.

8.1. FISSION GAS RELEASE (K F A Jülich).

After extraction from the irradiation rig, the plutonium particle capsule was punctured in a vacuum chamber. A 100 cm^3 aliquot of the sucked gas was analysed by means of a germanium - detector. As a result of this, only Xe 133 was measurable. Calculated on the last irradiation day it gives a Xe 133 quantity of 8.4×10^{11} atoms. Assuming that at the end of the last irradiation cycle an equilibrium was reached between the release and the decay of Xe atoms, we can calculate a R/B value for the last irradiation stage of :

$$R/B = 4.2 \times 10^{-7} \quad [14]$$

8.2. DISMANTLING (K F A Jülich).

After the puncture test the metallic capsule was cut at the base for cartridge extraction. Fig. 13 shows the different parts after this operation. One can notice black and white deposits on the inside of the metallic part and on the graphite.

8.3. VISUAL EXAMINATION.

Some difficulties have arisen with the unloading of the particles because they were stuck to each other and also on the wall of the graphite container. This can be seen on Fig. 14. Apparently, this was due to a white coloured product deposited on the surface of the coated particles. Examination of the particles of the complete contents of capsules 1, 3, 4 using the hot - cell periscope, showed no cracks or other major structural damage.

8.4. ANNEALING TEST (in hot cell K F A Jülich).

Each type of particles has been heated up to 1350°C (containers 2, 5 and 8) ; a check has been made up to 1500°C (container 2). As a function of the long half-lifes and the good condition of the particles, the measured release of Xe 133 was very small and, in the case of container 5, practically not detectable [14] . As no release of Kr 85 was measurable, it can be assumed that the particles were not damaged.

The following table gives the release rate of Xe 133 in atoms/sec, after steady release had been reached. The figures refer to the date of the end of irradiation.

Container nr	Number of particles	Particle type	Temperature (°C)	R (Xe 133) (atom/sec)	Fraction release R/B
2	930	C	1350	3.0×10^{7}	8×10^{-5}
			1500	1.1×10^{7}	2.9×10^{-5}
5	540	D	1350	less than background	-
8	800	T	1350	2.2×10^{7}	6×10^{-5}

These results confirm that no particle was broken during irradiation.

8.5. METALLOGRAPHY.

Microsections of each kind of irradiated particles were prepared first for one sample of each type of particles (containers 1, 3, 4) and then for the particles having been subjected to annealing test (2, 5 and 8). The results can be summarized as follows :

(1) Container 1 (D particles) - Fig. 15 and 16.

Apparently no change in the coatings could be detected in spite of the high anisotropy of the sealing layer (see Fig. 3). Even part of the sooty layers was still visible. Inside the kernel many cracks could be seen.

(2) Container 5 (D particles annealed at 1550°C) - Fig. 16.

The same remarks as for container 1 are also valid as no major difference could be detected.

(3) Container 3 (T particles) - Fig. 17 and 18.

From the microsections, it appears that the kernel did not diffuse through the sooty layer to a markedly larger extent than what was due to fabrication (Fig. 16). Quite an important delamination between the SiC and inner layers can be observed. The buffer layer is cracked over the outer half of its thickness in approximately half of the particles. The segregation of metallic parts into the kernel is important (Fig. 18).

(4) Container 8 (T particles after annealing at 1350°C) - Fig. 19.

The same general results as for container 3 are noticeable. Some particles show cracks which have started from the kernel but were stopped at the SiC layer.

(5) Containers 2, 4 and 7 (C particles) - Fig. 20.

The diffusion of the kernel into the porous layer resulting from the fabrication did not extend throughout the full layer. "Spearhead" attack is visible in the inner pyrocarbon coating whenever the plutonium is spread throughout all the buffer layer ; it is not visible when the plutonium diffusion is limited to part of the buffer layer. The SiC and outer PyC layers remained in good condition.

(6) Container 2 (C particles after successive annealing at 1350°C and 1500°C) - Fig. 21.

No much difference could be found by comparison with container 4.

8.6. INVESTIGATIONS ON MARKED PARTICLES.

Marked particles which were introduced in the central hole of the sample container have been examined after fabrication and after irradiation.

The characterization after fabrication consists in a X-radiography to indicate the plutonium location within the particle (Cf. item (6)).

The postirradiation examination consists in two different tests ; an alpha-autoradiography of the coated particles which indicates the evolution of the plutonium migration to be compared with the X-ray results and a polished section of these coated particles. These photographs are presented in Fig. 22 to 30. An estimate of the plutonium migration, given by a microdensimetry of the α-autoradiography of some of these particles, is presented at Fig. 31.

For the T particles (Fig. 22 to 24), the plutonium migration to a few spots within the buffer layer during fabrication is emphasized by the X-ray examination. The alpha-autoradiography indicates that this preferential plutonium location has not changed during irradiation. However a densitometric analysis of the alpha-radiographies suggest a tendency to plutonium migration away from the spots. Outside the kernel, the α concentration for these particles varies between 5 and 15 % (particles 3 - 4 - 7)(Fig. 31). All these coated particles have an intact inner PyC layer and a partial discoupling of this layer from the SiC layer.

For the C particles, an important plutonium diffusion is noticed in the buffer after fabrication. The alpha-autoradiography obtained for the C particle nr 6 shows a high plutonium penetration generalized in the buffer PyC coating. Its α-microdensimetry leads also to the same conclusion (Fig. 31).

The destruction of the inner PyC coating is also noticed for particles nr 5, 9 and 13 (Fig. 27 to 29). This damage of the inner PyC would be induced by the combined effect of shrinkage induced by fast neutron dose and spearhead attack induced by fission product recoil of the migrating plutonium. For particle nr 8 (Fig. 25), the buffer layer is completely discoupled from the inner PyC layer, which remained intact. Nevertheless, the plutonium migrated through the total thickness of the buffer.

For the D marked coated particle which was examined, one notices quite the same behaviour as for the D particles from container 1. The inner PyC layer remained intact and the dispersed kernel in a carbon matrix shrinked avoiding any mechanical interaction with the pressure vessel. Unfortunately, no X-ray has been done after fabrication on this type of fuel, but no plutonium migration has been noticed in the inner PyC layer by metallography and alpha-autoradiography.

8.7. LATTICE PARAMETERS.

The lattice parameters were measured by X-ray diffraction at C C R - Karlsruhe. Table XIV gives the data obtained after irradiation. The lattice parameters of unirradiated PuO_2 and Pu(O,C) have been added for reference.

Within the sensitivity of the technique, no other structure than the fluorite one corresponding to PuO_2 was visible after irradiation. No attempt was made to correct for the presence of fission products dissolved in the lattice.

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9.- INTERPRETATION OF THE IRRADIATION RESULTS.

9.1. STRESS ANALYSIS.

Considering the microphotographs of the concerned coated particles, it is possible to distinguish three different behaviours which are :

- discoupling of the buffer layer from the inner PyC layer resulting in a pressure vessel constituted by the PyC/SiC/PyC layers ;
- discoupling of the inner PyC layer from the SiC layer resulting in a pressure vessel constituted by the inner PyC layer ;
- destruction of the inner PyC layer leaving a pressure vessel constituted by the SiC and the outer PyC layers.

These coated particle behaviours are differently distributed in the three batches. Table XV based on the microphotographs gives for each of the three batches the number and the percentage of particles in regard of the above described behaviour type. For each behaviour type, a stress - strain analysis can be performed, giving the gas pressure evolution and the stress history in the layers constituting the pressure vessel.

The mathematical model called COCONUT* [13] and developed by BN considers the irradiation effect on the mechanical properties of the coating, the PyC dimensional change induced by irradiation, the fuel swelling, the fission gas build-up and the CO produced. The calculations are performed using a time step procedure required by the use of an irradiation creep law for the PyC coating.

The CO pressure estimate versus temperature and stoechiometry is extrapolated for several plutonium oxides (Fig. 32) from free enthalpies data published by T.L. Markin and E.J. Metver (UKAEA - Harwell) [15] and L.M. Atlas and G.J. Sekelman (ANL) [16].

The variation of the fission gas release from the kernel has been considered as linear versus the burn-up. This release reaches 90 % for 18.0 % FIMA at 1200°C.

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*COating COmputation for NUclear Technology.

Since, some metallographies show adherence between the inner PyC and the SiC (Fig. 30), the disconnecting effect of the PyC flash layers deposited between the inner PyC and the SiC and between the SiC and the outer PyC has not been considered in the mathematical analyses.

The gas pressure evolution, for the three coated particle behaviour types, is given in Fig. 33 for a constant temperature of 1200°C. In the case where the pressure vessel is constituted by the inner PyC layer, the gas pressure is higher than in the other case where the void volume offered for the gas expansion is larger. For both cases, the gas pressure evolution is quite similar from one behaviour (PyC/SiC/PyC pressure vessel) to the other one (SiC/outer PyC pressure vessel), with a lower pressure for the last case according to the higher void volume induced by the inner PyC breakage.

For the neutron dose reached in this experiment, one remains in the region of volume shrinkage of the PyC (Fig. 34). This shrinkage is anisotropic and occurs mainly following the "C axis" of the crystal (radial direction of the layer (Fig. 35) [17].

For the plain PyC pressure vessel, a compression of this layer is induced (at start of life) by its dimensional change under irradiation (Fig. 36), this layer remaining attached to the buffer layer. At 0.3×10^{20} cm^{-2}, the fuel swelling influences the stress level in this layer and thus the hoop stresses increase, being moderated by the PyC irradiation creep.

The hoop stress evolutions in both other cases (SiC/outer PyC and PyC/SiC/PyC pressure vessel) are quite similar for the SiC and outer PyC layers (Fig. 37 and 38). On one hand, a decompression of the SiC layer results from the gas pressure build-up and the outer PyC dimensional change under irradiation ; on the other hand, for the fast neutron dose reached, the outer PyC dimensional change under irradiation induced a reduction of the hoop stresses in the outer PyC layer which remains in contact with the SiC layer.

In the case where the inner PyC is the inner part of the pressure vessel, the irradiation dimensional change and the gas pressure induce an increase of the hoop stresses. The stresses in all the PyC layers are reduced by the PyC irradiation creep.

9.2. CO PRESSURE EVOLUTION.

The CO pressure evolution has been calculated for the particles having the highest O/Pu ratio at start of life, and burn-up e.g. for the particles type T.

The input data are given below.

% FIMA	Irradiation temperature	O/Pu ratio*
0	1200	1.9
4	1140	1.922
8	1080	1.945
12	1050	1.967
16	1010	1.99
19	1000	2.007

*The O/Pu evolution as a function of burn-up is taken from [18].

The CO pressure evolution as a function of burn-up is given in Fig. 39.

9.3. PyC RESISTANCE TO FISSION PRODUCT BOMBARDMENT.

The damage to the PyC layers is not visible in the D particles and evident in the C particles.

On the basis of the evaluated plutonium concentration profiles, the damage to the non-fuelled inner coating can be calculated and related to an equivalent fast neutron dose [19]. Table XVI gives the results of these calculations. It can be seen that the porous PyC is resistant to irradiation damage while the anisotropic inner PyC shows spearhead attack at a threshold somewhat lower than reported in the literature ; mention is indeed made that the phenomenon starts between 2×10^{16} and 2×10^{17} FP/cm^2 [20].

* * *

10.- CONCLUSIONS.

For the considered coated particle design and manufacture, the following remarks can be pointed out :

10.1. PLUTONIUM DISPLACEMENT.

a. Pu displacement before irradiation.

An important plutonium migration occurred during the first two stages of coating for the dense kernels which led in one batch to a inner kernel relatively similar to the diluted kernel : i.e. the large buffer layer was completely impregnated with plutonium.

b. Pu displacement after irradiation.

The migration of plutonium during irradiation continues but is rather small compared with the diffusion during coating : i.e. a maximum migration value of 20 µm for the T type has been found from X-rays and alpha-autoradiography micrographies of the irradiated kernels. The plutonium migration has been stopped by the SiC layer ; this conclusion has to take account of the rather short irradiation time.

10.2. IRRADIATION DAMAGE.

In the limit of the relatively low value of burn-up, no broken or cracked particle has been detected in the three batches. The extremely good behaviour of D fuel has already been proved in earlier irradiations and in this case even the anisotropic inner dense pyrolytic carbon layer (sealing layer) remains intact.

* * *

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* * *

- T A B L E I -

TYPES OF PLUTONIUM PARTICLES

Type of fuel	Diluted						Undiluted	
Batch	P 352	P 353	P 372	P 373	P 382	P 383 "D"	THP 311 "T"	THP 321 "C"
Coated particle data :								
Diameter (μm)	700	600	600	700	800	890	790	850
Maximum HML (g Pu/cm^3)	0.04	0.07	0.10	0.09	0.26	0.22	0.20	0.14
g Pu/cm^3 CP	0.06	0.11	0.15	0.13	0.40	0.33	0,30	0.21
g Pu/cm^3 kernel x μm diameter (self-shielding factor)	250	250	340	340	700	800	2300* (1400)	2200* (410)
Peak irradiation data :								
Temperature (°C)	1450 1850		1200			1300		
Burn-up (GWd/t)	360 200		600			200		
R/B for Xe 133	5.10^{-6} 10^{-2}		7.10^{-6}			4.10^{-7}		
Sponsored by :	BELGONUCLEAIRE C E N / S C K DRAGON Project						BELGONUCLEAIRE C E N / S C K KFA Jülich	

*Theoretical values (actual values resulting from Pu diffusion).

- T A B L E II -

ISOTOPIC ANALYSIS AND ACTIVITIES OF Pu USED FOR PREPARING THE COATED PARTICLE FUELS

Specimen	Batch nr	Isotopes of Pu, in w/o				Activity curies/g
		239	240	241	242	
D	P 3/8/1 (Pu 019)	79.4	17.1	3.05	0.4	3.6
T	THP 3/1/1 D	91.44	7.9	0.67	0.030	1.9
C	THP 3/2/1 D (Pu 020)	88.3	10.3	1.3	0.09	1.55

- T A B L E III -

HEAT TREATMENT OF THE GREEN KERNELS

Batch nr	Temperature interval °C	Heating rate °C/h	Atmosphere
D (P 2/8/-)	20 - 200 200 - 1000 1000 - 1400 1400	200 533 200 0 (2 h)*	CO CO CO CO
T (THP 2/1)	950 1600 - 1700	0 (3 h)* 0 (4 h)*	Air H_2
C (THP 2/2)	20 - 20 - 900 900 - 1450 1450 - 1000	 200 200 -150	Air dried A - H_2 CO CO

* ( ) : time at temperature.

- T A B L E IV -

PROPERTIES OF SINTERED KERNELS

Batch nr	Hg density g/cm³	Porosity %	Pu/C ratio	O/M			Size (µm)		w/o Pu
				Chem.	X - ray		Sieve range	Average	
					$\frac{I\ Pu_2O_3(222)}{I\ PuO_2(111)}$	O/M			
D (P 2/8) (b)	2.75	31	1 : 19.5	n.d.	0.23	1.9	450 - 572	500	47.6
T (THP 2/1) (a)	10.5	3.7	-	1.77	n.r.	1.75	227 - 331	280	89.5
T (THP 2/1) (b)	10.2	8	-	1.90	0.25	1.9	254 - 312	270	88.73
C (THP 2/2)(b)	9.8	10	-	1.745	1.0	1.7	216 - 292	260	89.54

(a) Data received from C C R - Karlsruhe
(b) Analytical data obtained at Mol.

- T A B L E V -

COATING CONDITIONS

Fuel		Coating			Gas flows							
Type	Identification	Temperature °C	Layer type	Time (min)	Total (l/min)	C_2H_2 %	CH_4 %	H_2 %	CO total	A %	% H_2 via Silane	Silane %
D	P 3/8/1	1450	Soft PyC	8	4.325	23.1			76.9			
		1550	D PyC	80	4.125		17	32.7	50.3			
		1450	Flash	2	4.325	23.1				76.9		
		1450	SiC	62	5.646			35.4		21.7	35.4	7.5*
	P 3/8/2	1450	Flash	2	4.600	21.8				78.2		
		1800 2000	PyC HDI	8	4.100		12			88		
		2000	HD PyC	90	4.100		12			88		
	P 3/8/3	2000	HD PyC	120	4.100		12			88		

*Evaluation

TABLE V (continued)

Fuel		Coating			Gasflows							
Type	Identification	Temperature °C	Layer type	Time (min)	Total (l/min)	C_2H_2 %	CH_4 %	H_2 %	CO %	A %	% H_2 via Silane	Silane %
T	THP3/1/1A	1480	Porous	40	4620	50	-	-	50	-	-	
	THP3/1/1B	1550	D	80	4125	-	17	32.7	50.3	-	-	
	THP3/1/1C	1450	Flash	2	4625	28.1	-	-	-	71.9	-	
		1450	SiC	62	5699	-	-	35.1	-	21.5	5.1	8.5*
	THP3/1/1D	1450	Flash	2	4.9	26.5	-	-	-	73.5	-	
		1800-2000	HDI	8	4.1	-	12.2	-	-	87.8	-	
		2000	HDI	210	4.1	-	12.2	-	-	87.8	-	

*Evaluation

TABLE V (continued)

Fuel		Coating			Gas flows							
Type	Identification	Temperature °C	Layer type	Time (min)	Total (l/min)	C_2H_2 %	CH_4 %	H_2 %	CO %	A %	% H_2 via Silane	Silane %
C	THP3/2/1A	1530	Porous	34	4.620	50.0	-	-	50.0	-	-	
	THP3/2/1B	1550	D	80	4.125	-	17	32.7	50.3	-	-	
	THP3/2/1C	1450	Flash	2	4.625	28.1	-	-	-	71.9	-	
		1450	SiC	62	5.759	-	-	35.3	-	21.7	35.3	7.7*
	THP3/2/1D	1450	Flash	2	4.9	25.5	-	-	-	73.5	-	
		1800-2000	HDI	8	4.1	-	12.2	-	-	87.8	-	
		2000	HDI	210	4.1	-	12.2	-	-	87.8	-	

*Evaluation

- T A B L E VI -

PLUTONIUM CONTENTS

Fuel		Plutonium calculated from the chemical analysis of the sintered particles				Pu content (w/o Pu) by analysis after coating		
							Gamma-spectrometry	
Type	Identification	In	Out	Plutonium content in g	% Pu calculated	Chemical	Average	Standard deviation % relative
D	P 2/8/-	50	-	23.8	-	47.6	-	
	P 3/8/1	50	116.17	23.8	20.48	-	-	
	P 3/8/2	116	138.8	23.76	17.12	17.74	-	
	P 3/8/3	121.4	144.5	20.78	14.38	14.97	-	
T	THP2/1	50	-	44.365	-	88.73		
	THP3/1/1A	50	102.09	44.365	43.45		42.0	4.0
	THP3/1/1B	57.007	72.67	24.78	34.10		36.0	10.5
	THP3/1/1C	68.082	106.047	23.30	21.97		20.7	10.5
	THP3/1/1D	101.242	148.16	22.24	15.01	13.83	13.8	10.4
C	THP2/2	50	-	44.77	-	89.54		
	THP3/2/1A	50	127.233	44.77	35.18		30.2	4.2
	THP3/2/1B	57.052	70.273	20.07	28.16		-	-
	THP3/2/1C	66.927	108.09	19.11	17.68		14.3	11.9
	THP3/2/1D	100.7	148.68	17.80	11.97	9.9	9.94	11.6

- T A B L E VII -

DENSITIES OF SINTERED AND COATED PARTICLES

Type	Identification	Bulk density		Particles density	Layer density
		g/cm^3	% part. dens.	g/cm^3	
D	P 2/8/*	1.7	63	2.75	-
	P 3/8/2	1.5	66	2.3	n.d.
	P 3/8/3	1.3	59	2.2	2.0
T	THP2/1*	6	61	10.2	-
	THP3/1/1A	1.3	57	2.2	~ 1 (estim.)
	THP3/1/1B	1.2	59	2.1	1.7
	THP3/1/1C	1.3	59	2.3	2.8 *
	THP3/1/1D	1.3	59	2.1	1.9
C	THP2/2*	7	71	9.8	-
	THP3/2/1A	1.1	61	1.85	~ 1 (estim.)
	THP3/2/1B	1.1	64	1.8	1.6
	THP3/2/1C	1.3	61	2.1	2.9 *
	THP3/2/1D	1.3	64	2.1	2.0

*Including the flash PyC layers.

- T A B L E VIII -

X - RAY ANALYSIS OF COATED PARTICLE FUELS

Type	Identification	Chemical form of plutonium		$\frac{I\ Pu_2O_3(222)}{I\ PuO_2(111)}$	O/M
		Mean	Secondary		
D	Before coating	PuO_2	$\alpha\ Pu_2O_3$	0.23	1.9
	P 3/8/3	$\alpha\ Pu_2O_3$	PuO_2	1.3	1.75
T	Before coating	PuO_2	$\alpha\ Pu_2O_3$	0.25	1.9
	THP3/1/1A	$\alpha\ Pu_2O_3$	PuO_2	2.3	1.6
	THP3/1/1B	$\alpha\ Pu_2O_3$	PuO_2	3.7	1.6
	THP3/1/1C	$\alpha\ Pu_2O_3$	PuO_2	2.4	1.6
	THP3/1/1D	$\alpha\ Pu_2O_3$	PuO_2	2.5	1.6
C	Before coating	PuO_2	$\alpha\ Pu_2O_3$	1.0	1.7
	THP3/2/1A	$\alpha\ Pu_2O_3$	PuO_2	2.0	1.6
	THP3/2/1B	PuO_2	$\alpha\ Pu_2O_3$	0.6	1.8
	THP3/2/1C	PuO_2	$\alpha\ Pu_2O_3$	0.7	1.8
	THP3/2/1D	PuO_2	$\alpha\ Pu_2O_3$	0.8	1.75

- T A B L E IX -

EXTERNAL ALPHA - CONTAMINATION OF COATED PARTICLE FUELS

Type	Identification	dis/s g *	FVC *
D	P 383	670	3.4×10^{-8}
T	THP3/1/1A	123 000	1.5×10^{-5}
	THP3/1/1B	27 500	3.3×10^{-6}
	THP3/1/1C	350	4×10^{-8}
	THP3/1/1D	4 230	5×10^{-7}
C	THP3/2/1A	39 700	7×10^{-6}
	THP3/2/1B	24 300	4×10^{-6}
	THP3/2/1C	1 200	2.0×10^{-7}
	THP3/2/1D	46 980	8×10^{-6}

*dis/s g = disintegrations per second and per gram of particles.

FVC = fraction of visible core = ratio of alpha-count of the particle to alpha-counts of the fissile material included in the kernel.

- T A B L E X -

KERNEL AND COATING DIMENSIONS

Type	Identification	Layer type	Kernel diameter µm	Layer thickness µm	Cumulative coated thickness µm	CP diameter µm
D	P 3/8/1	Porous PyC	550^{+22}_{-27}	21	21	(590)
		Dense PyC		45 ± 5	66	(680)
		SiC		41 ± 4	107	(760)
	P 3/8/2	HD PyC		28 ± 10	135	(800)
	P 3/8/3	HD PyC		34 ± 10	169	888
T	THP3/1/1A	Porous	267^{+48}_{-31}	145^{+20}_{-14}	145	(560)
	THP3/1/1B	Dense PyC		31 ± 1	176	(620)
	THP3/1/1C	SiC		32^{+2}_{-3}	208	(680)
	THP3/1/1D	HDI		52^{+4}_{-7}	260	787
C	THP3/2/1A	Porous	248^{+40}_{-23}	187^{+30}_{-20}	187	(620)
	THP3/2/1B	Dense PyC		29 ± 2	216	(680)
	THP3/2/1C	SiC		32 ± 2	248	(740)
	THP3/2/1D	HD		53 ± 5	301	850

- T A B L E XI -

ACCURACIES OF THE DIFFERENT ANALYSES.

Analysis	M e t h o d	Accuracy in % relative
Hg density	Hg/vacuum	± 0.5
Bulk density	Weight of particles in known volume	± 5
Layer density	Computed from weight and volume gains	High density ± 5 Low density ± 20
Pu/C	Chemical calcinations	± 0.5
O/M	Gravimetric	± 0.5
$\frac{(Pu_2O_3)}{(PuO_2)}$	Ratio of the intensity of the 2.2.2 Pu_2O_3 line and the intensity of the 1.1.1. PuO_2 line	Indicative values
Particles sizes	X-ray photograph of Ni coated particles	± 4
Pu	Chemical calcination - dissolution - titration gamma-scanning	± 1
Alpha counting		± 10

- T A B L E XII -

OUTER DIAMETER OF THE PLUTONIUM ZONE (μm)

Transfer container number	Particle		Kernel before coating	After coating		After irradiation	
	Number	Type		Kernel	Diffusion zone	Kernel	Diffusion zone
1	3	T	267 μ + 48 - 31	270	570	270	550
2	4			246	460	260	500
3	7			250	460	250	500
	8	C	248 μ + 40 - 23	600	600	n.d.	
4	6			550	550	550	550
5	5			630	630	n.d.	
6	9			540	540	n.d.	
7	13			500	500	n.d.	
8	14	D	550 μ + 22 - 27	n.d.			475
9				Wasted before examination			

- T A B L E XIII -

SPECIFICATIONS OF EACH BATCH FOR CAPSULE LOADING

	D	T	C
Middle diameter in μ	888	787	850
Density of ended coated particle	2.23	2.14	2.08
Bulk density	1.3	1.26	1.33
% plutonium content (chemical)	14.97	13.83	9.9
Total plutonium content in one hole (1350°)(calculated with the various isotopic composition)	0.06633	0.0600	0.062 - 0.063
Volume 1 particle : x 10^{-4} cm^3	3.69121	2.53284	3.21555
Weight of 1 particle : g x 10^{-3}	0.82314	0.54203	0.66883
Weight of plutonium in 1 particle : g x 10^{-3}	0.12322	0.074963	0.066214
Number of particles in one hole	538	800	936
Weight of coated particles in one hole in g	0.4430861	0.433839	0.6262626
Bulk volume of these particles in cm^3	0.576	0.547	0.833

- T A B L E XIV -

LATTICE PARAMETERS (Å)

Particle types	Unirradiated		After irradiation	
	Kernel	CP	Mean	Standard deviation
Analytical data				
D	-	-	5.394	0.018
T	5.400	-	5.442	0.006
C	5.396	5.398	5.381	0.100
Theoretical values				
PuO_2	5.396			
$PuC_{0.2}O_{0.8}$	4.96			

- T A B L E XV -

BEHAVIOUR DISTRIBUTION VERSUS DISCOUPLING MECHANISM

Type	Batch number	Pressure vessel type					
		Inner PyC		SiC / outer PyC		PyC / SiC / PyC	
		Number of particles	%	Number of particles	%	Number of particles	%
D	P 3/8/3	0	0	0	0	17	100
T	THP 3/1/1	15	33	24	52*	7	15
C	THP 3/2/1	5	17	23	76**	2	7

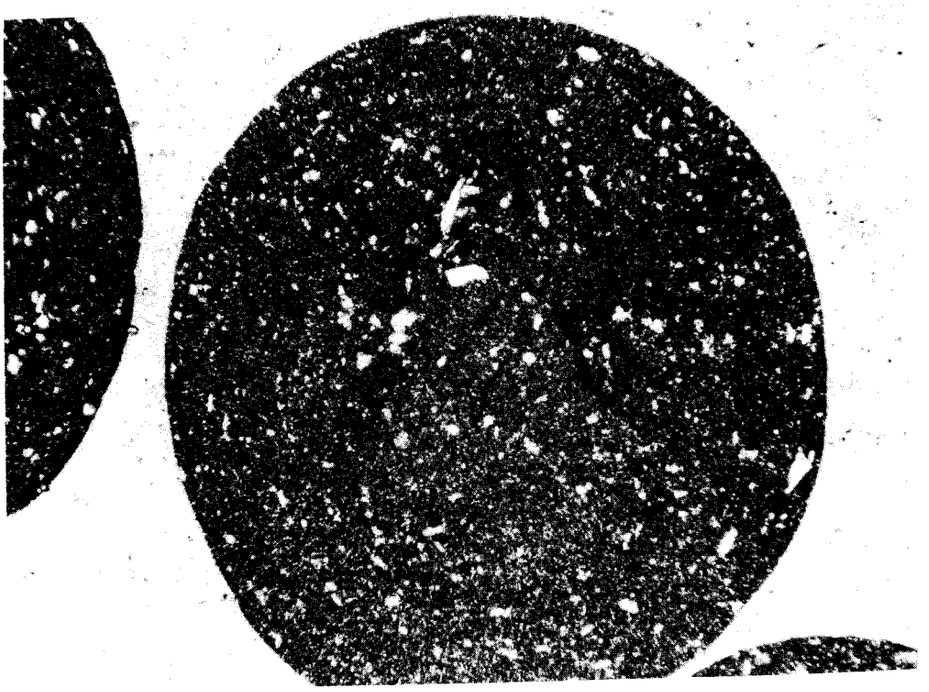
*Due to the breakage of the inner PyC.

**Due to the inner PyC damage by spearhead attack.

- T A B L E XVI -

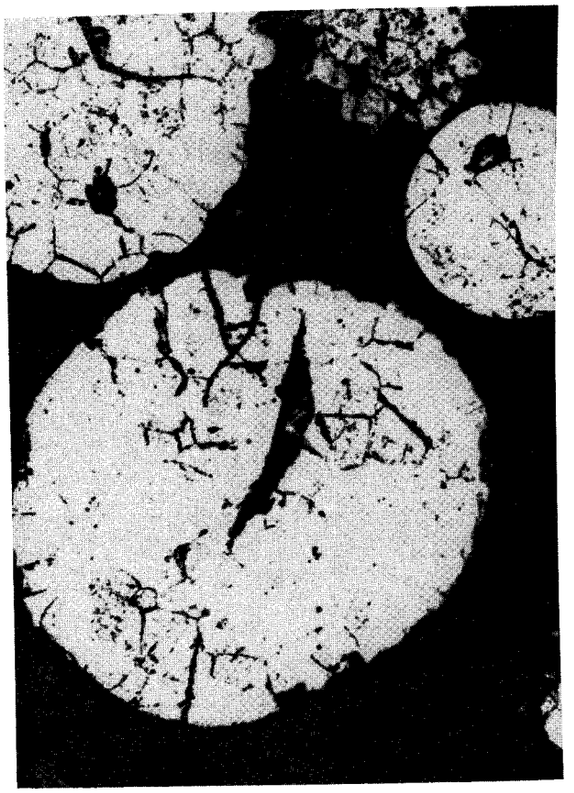
FISSION PRODUCT BOMBARDMENT OF THE COATING LAYERS

Particle type	D	T	C
Bombarded unfuelled coating	Porous PyC	Porous PyC	Inner PyC
FP/cm^2	3.8×10^{16}	1.8×10^{17}	1.6×10^{16}
$n.cm^{-2}$ DNE	1.52×10^{21}	7.2×10^{21}	0.96×10^{21}



(x 200)

D Fuel

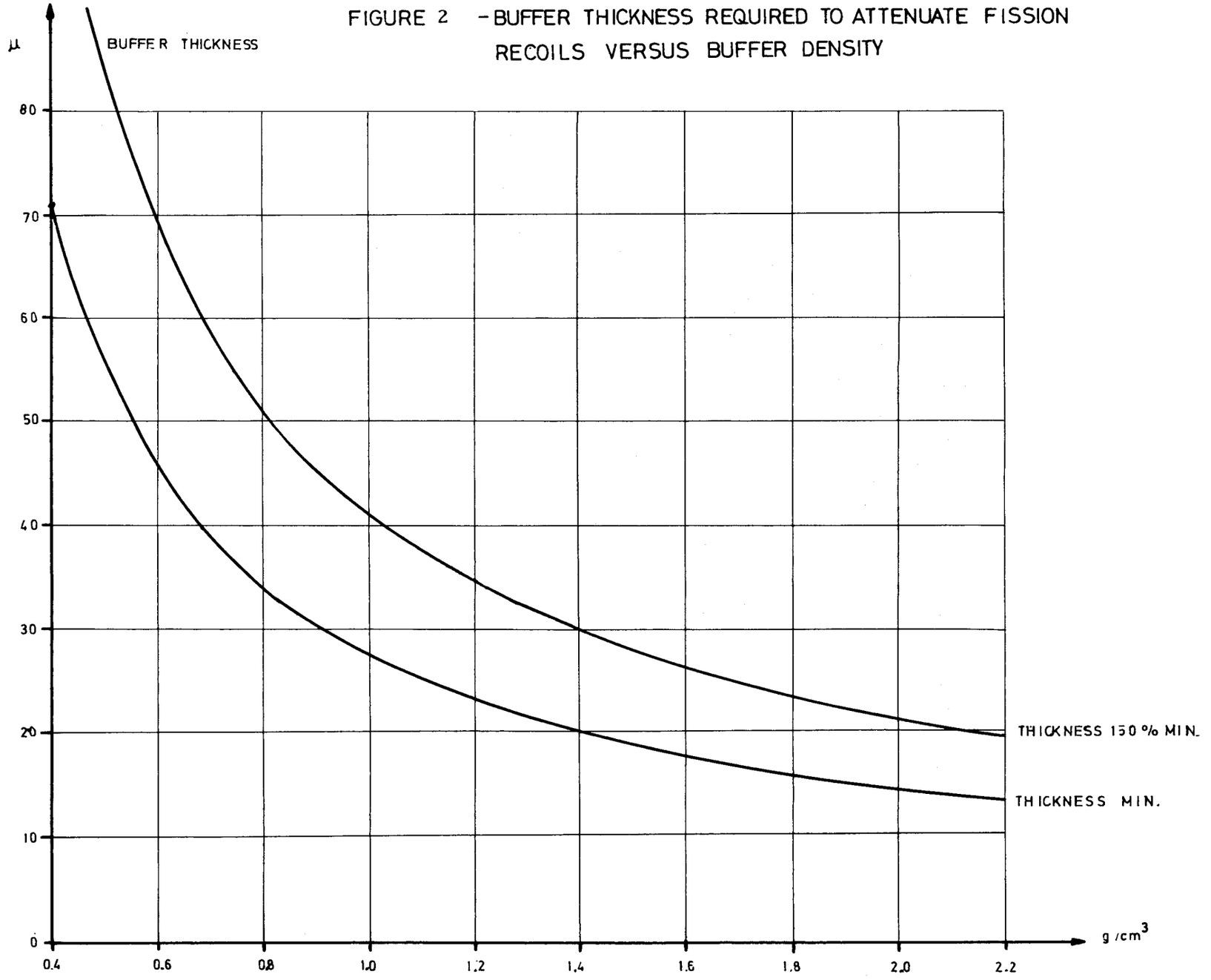


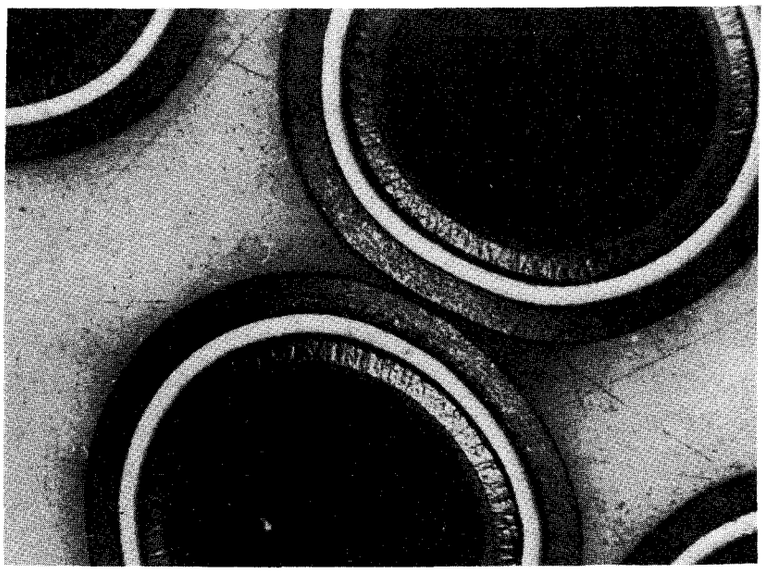
(x 200)

T Fuel

Figure 1 : Metallographic sections of undiluted and diluted kernels

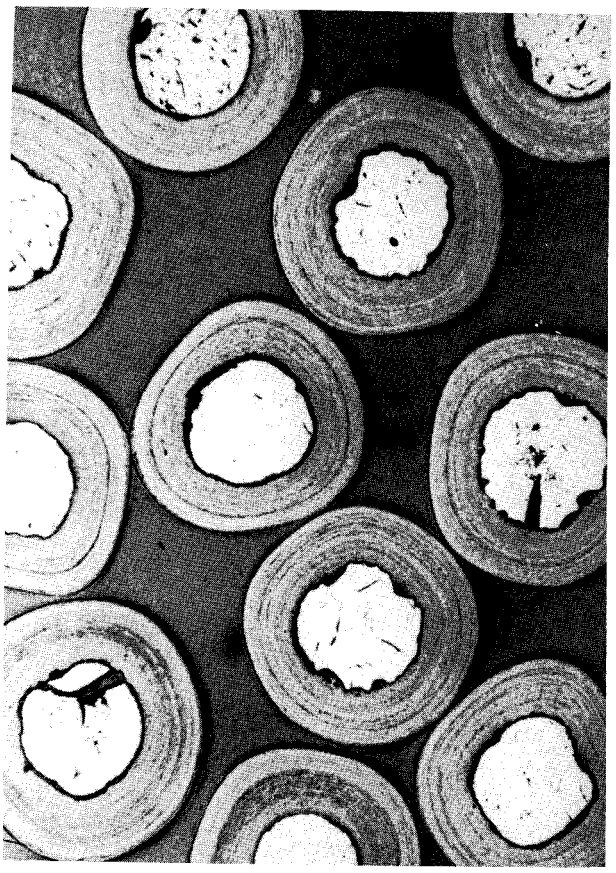
FIGURE 2 - BUFFER THICKNESS REQUIRED TO ATTENUATE FISSION RECOILS VERSUS BUFFER DENSITY



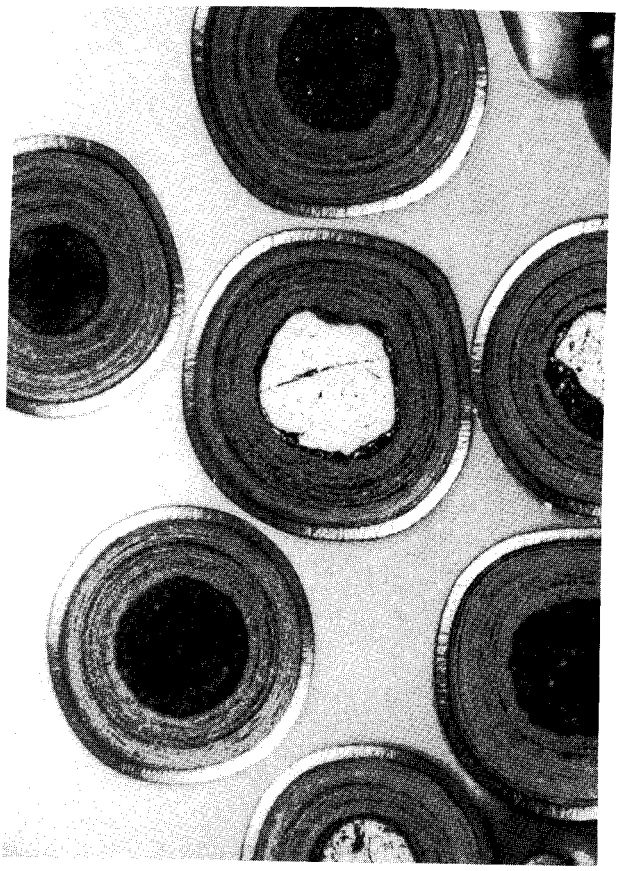


(x 80)

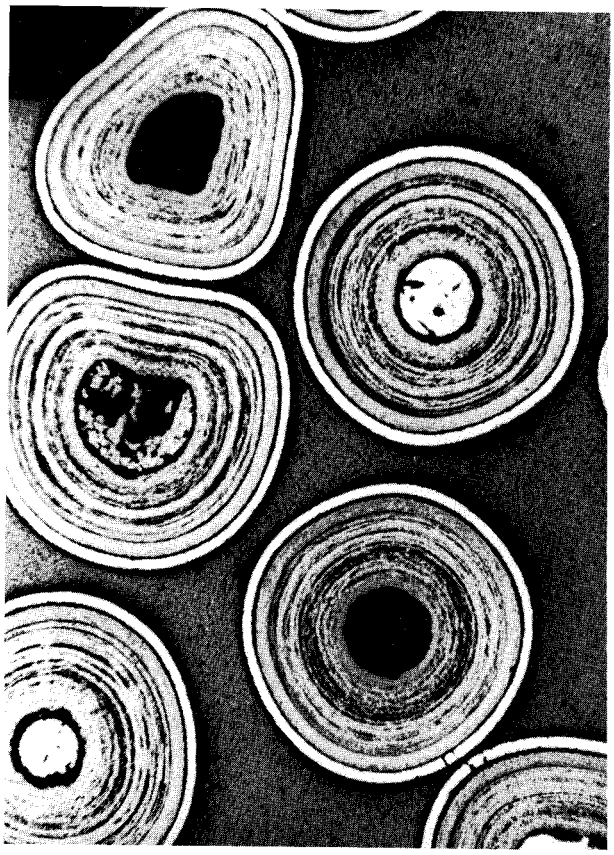
Figure 3 : Cross section of the D particles



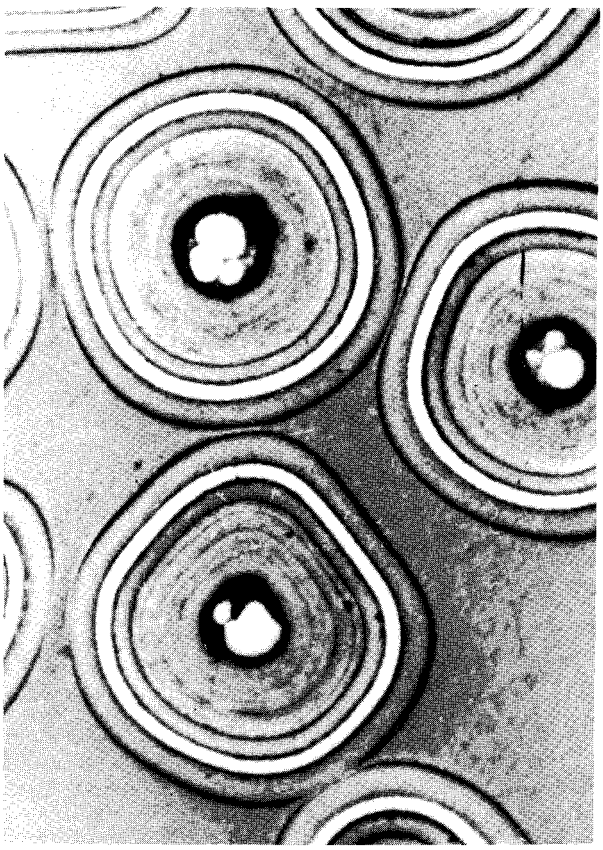
THP 311 A (x 60)



THP 311 B (x 60)

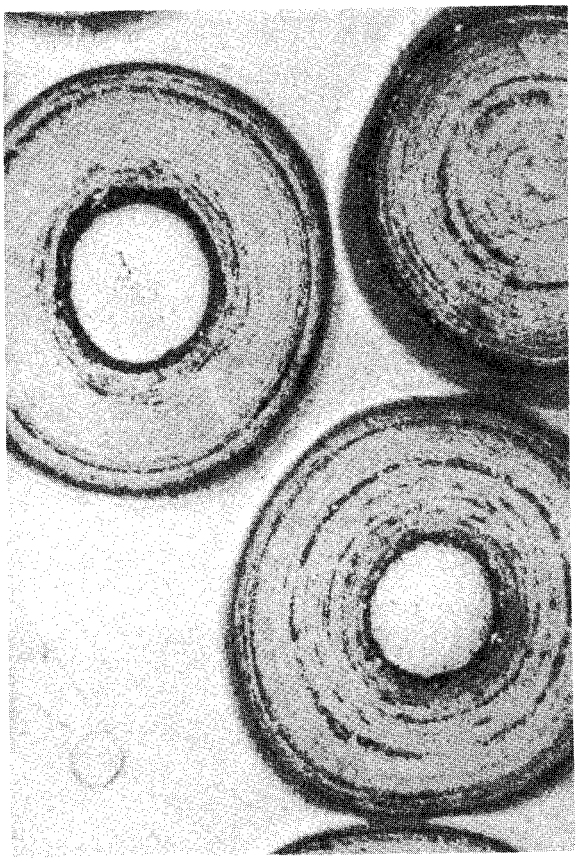


THP 311 C (x 60)

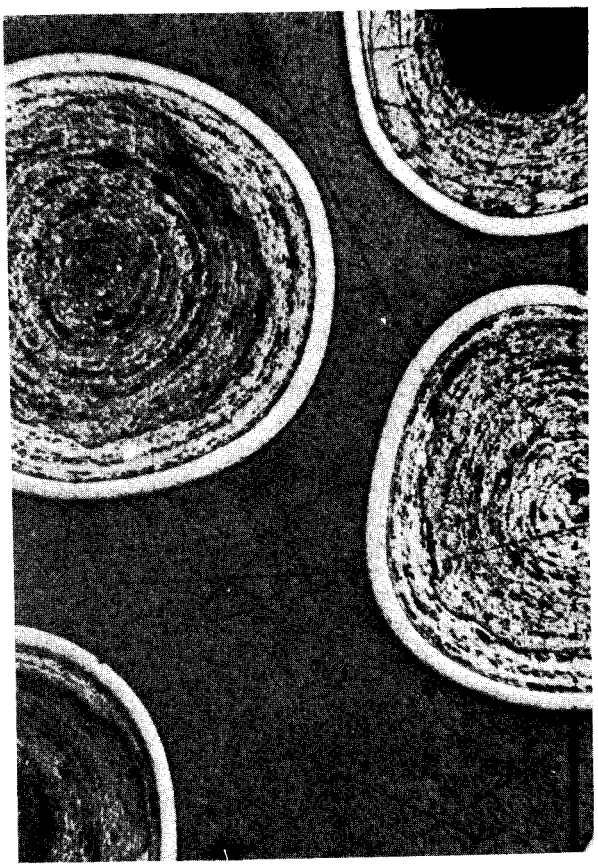


THP 311 D (x 60)

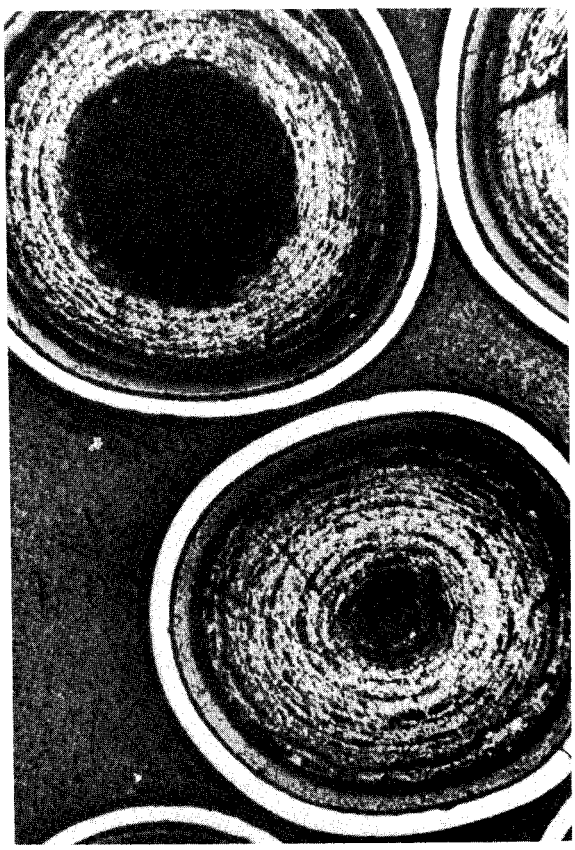
Figure 4 : Cross section of T particles



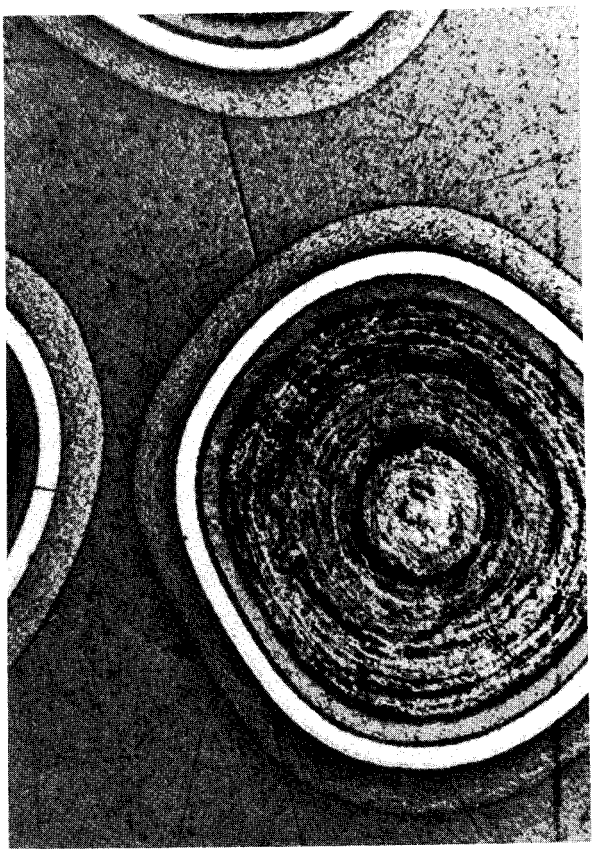
THP 321 A (x 90)



THP 321 B (x 90)

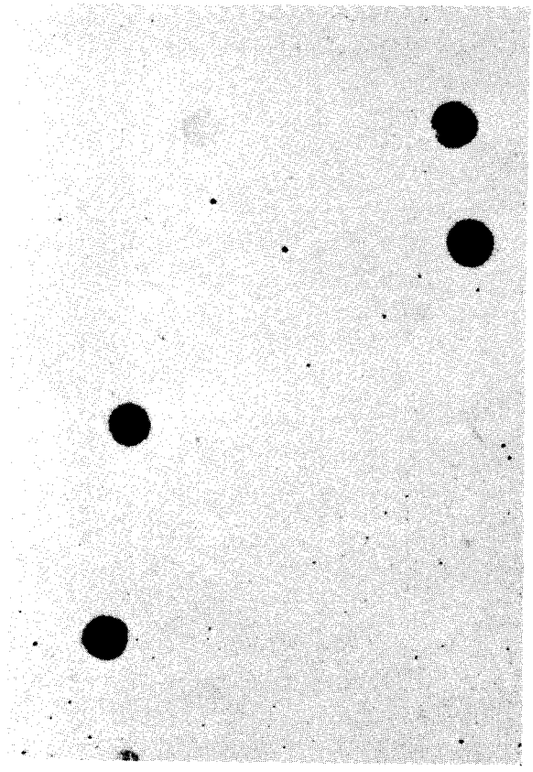


THP 321 C (x 90)

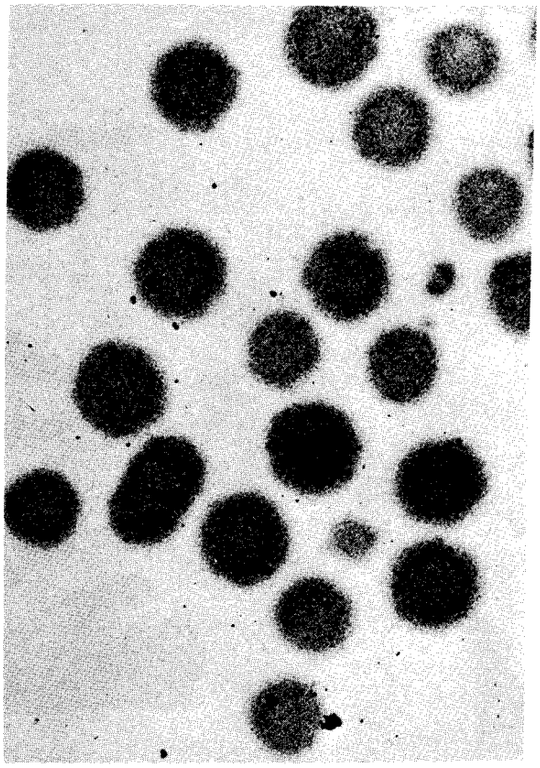


THP 321 D (x 90)

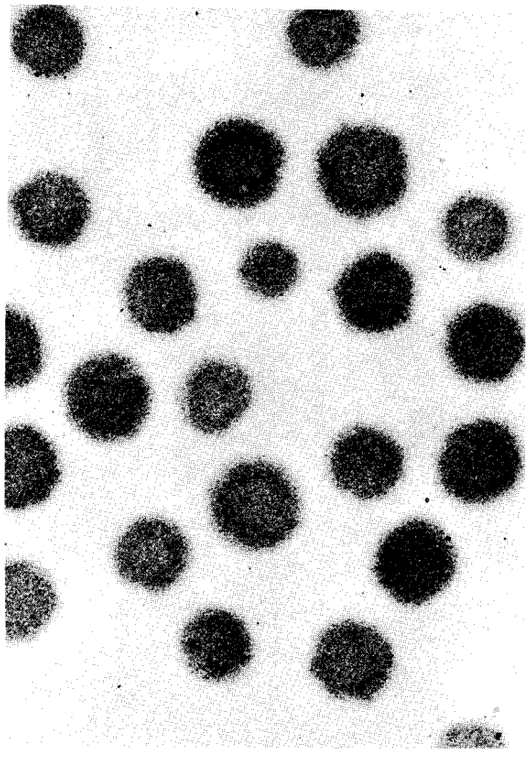
Figure 5 : Cross section of C particles



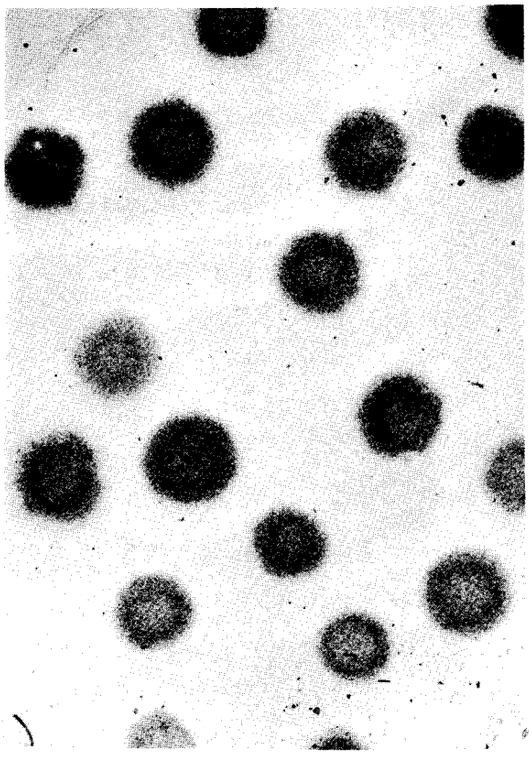
THP 3/2/1/A



THP 3/2/1/B



THP 3/2/1/C



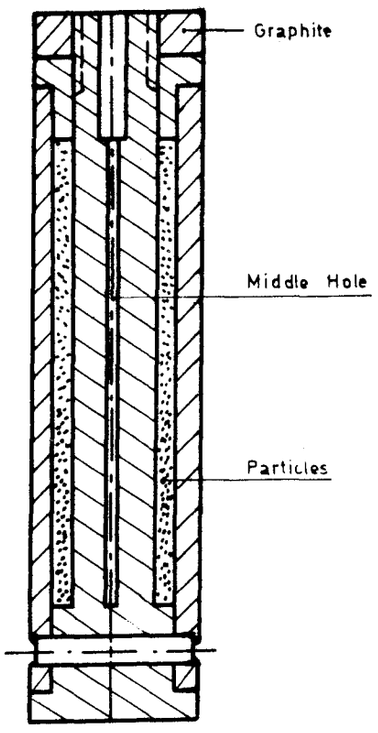
THP 3/2/1/D

Figure 6 : γ Autoradiograph of C kernel polished section after each carbon coating step

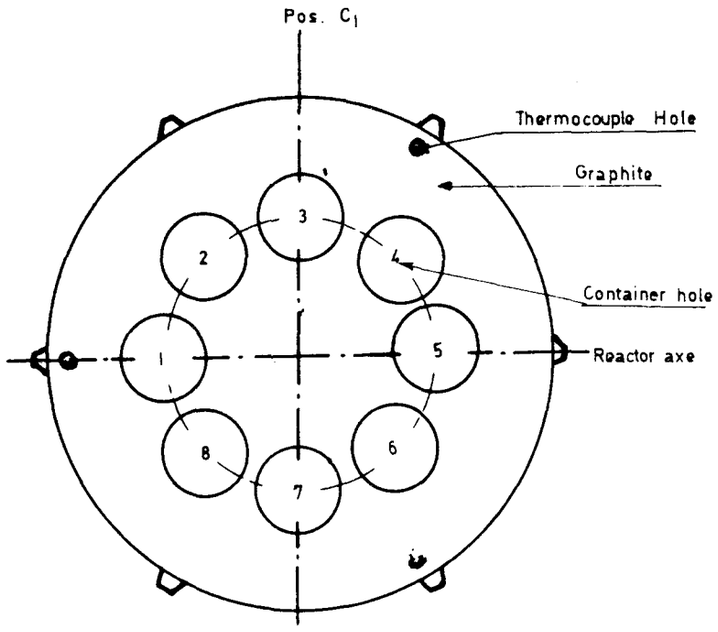
LOADING of the Pu COATED PARTICLES

for the JK4 IRRADIATION EXPERIMENT

CONTAINER. Nr.	TYPE OF PARTICLE.	PARTICLE-DIAMETER. (mm)	WEIGHT OF PARTICLES (g)	Pu - WEIGHT. (g)
1	D	-	0,4330	0,06482
2	C	0,699	0,6260	0,061974
3	T	0,835	0,4330	0,05988
4	C		0,6262	0,061993
5	D	-	0,4330	0,06482
6	T	0,699	0,4330	0,05989
7	C	0,835	0,6261	0,061983
8	T		0,4330	0,05988
CONTAINER Nr.1 (MIDDLE HOLE)		LOADING.		
1a 1b 1c	DRAGON CNEN KARLSRUHE	5 Particles of each type carefully checked before irradiation.		



LOOSE PARTICLES CONTAINER
PERPENDICULAR CUT
ENLARGEMENT 2:1



TELEPHONE DIAL TYPE GRAPHITE CARTRIDGE
UPPER VIEUW
ENLARGEMENT 1:1

Figure 7

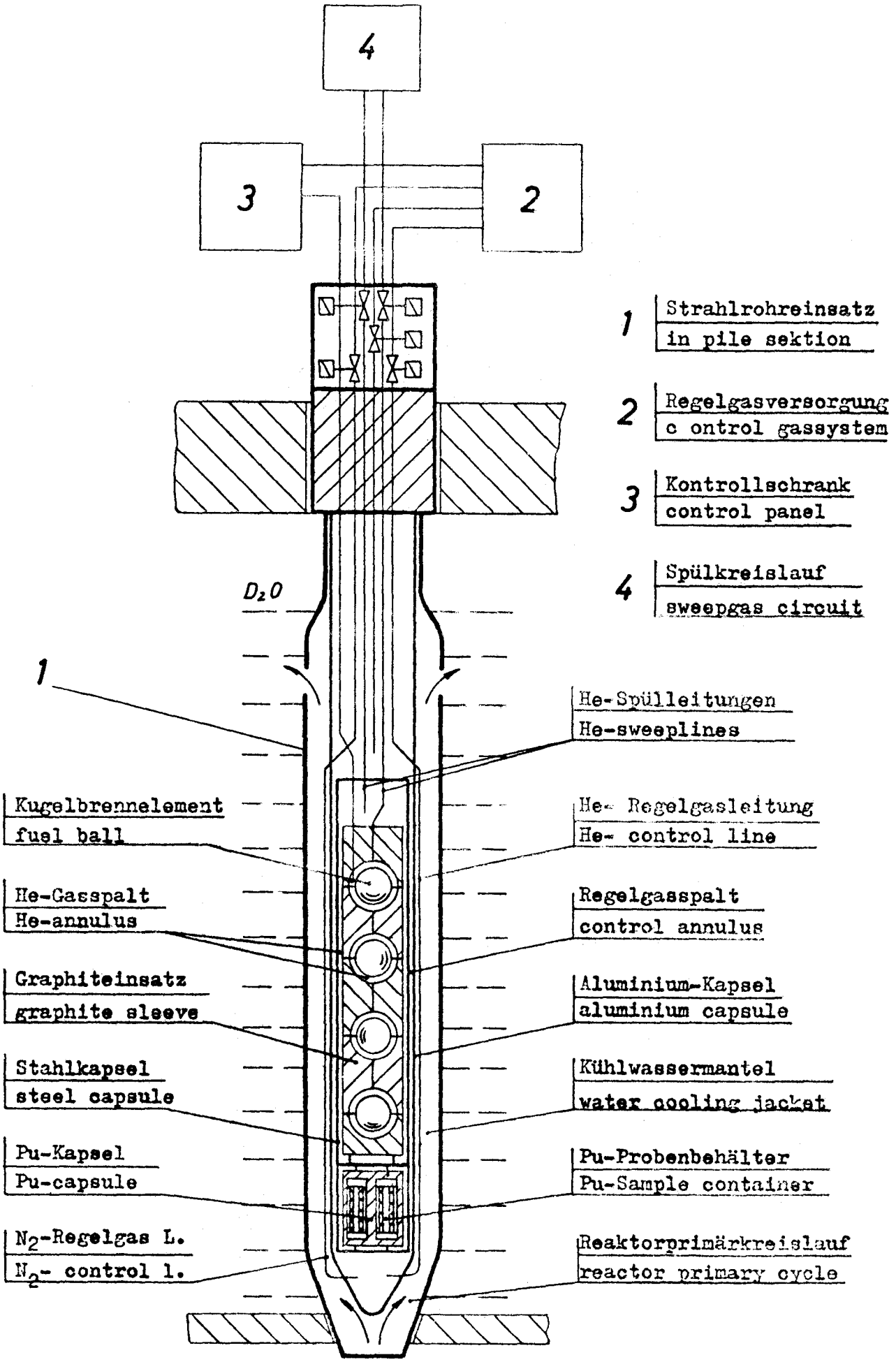


Figure 8 : Irradiation Rig JK 4

RADIAL TEMPERATURE PROFILE OF THE Pu CAPSULE

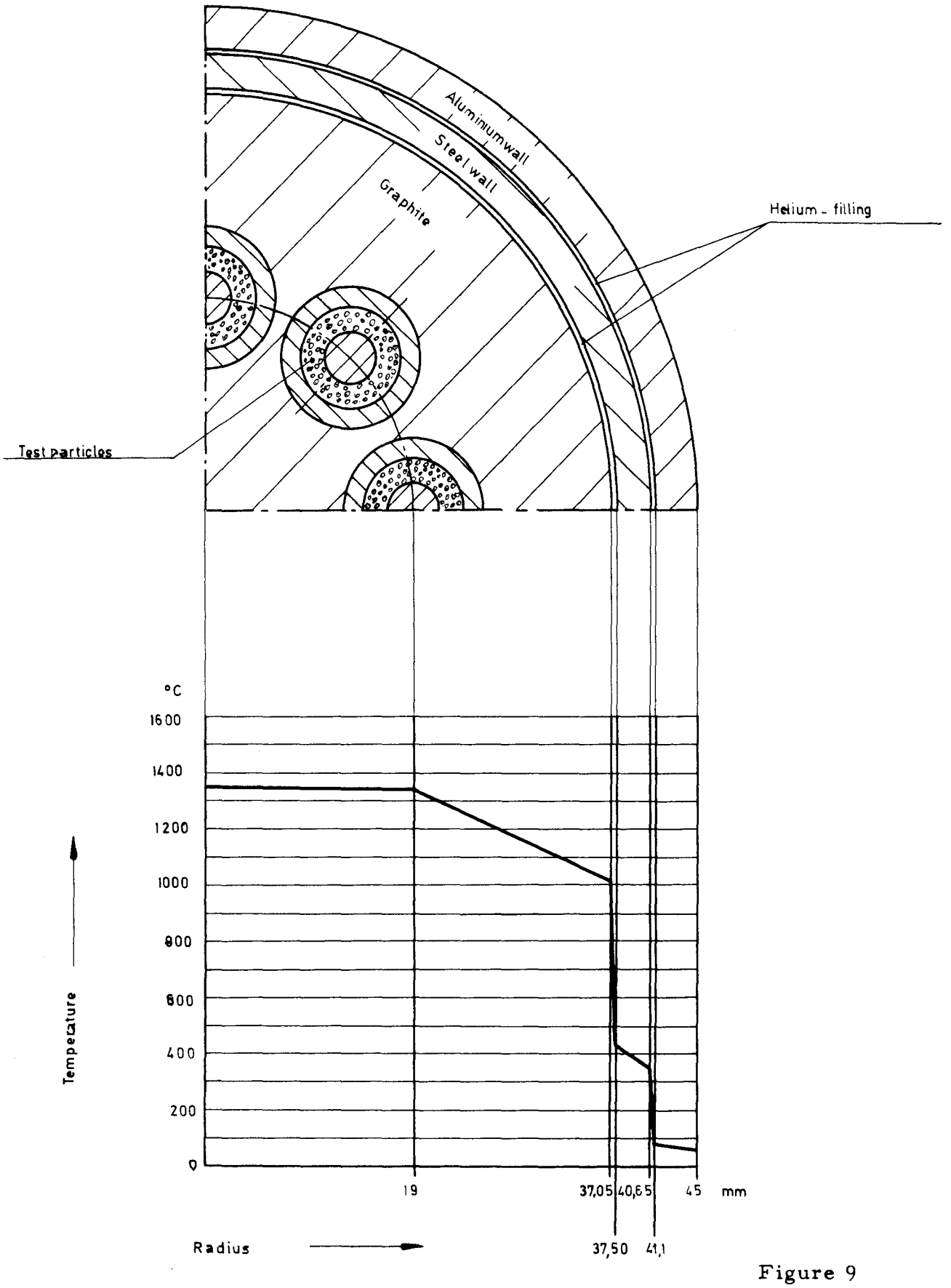


Figure 9

Figure 10 : **TEMPERATURE** HISTORY **OF LV4/JK4** Pu
KERNEL TEMPERATURE (calculated value) ± 50° C

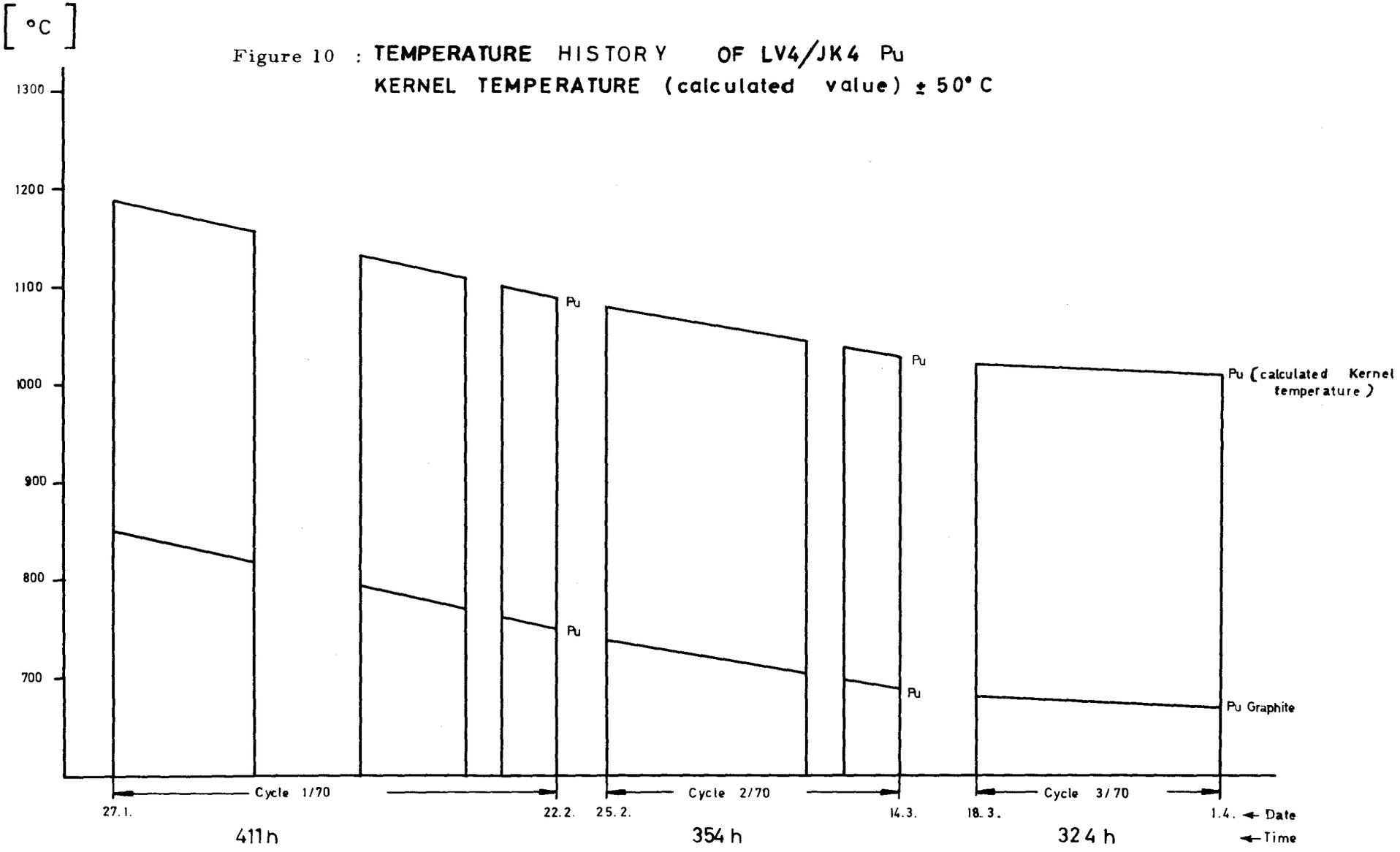


Figure 11 : FLUX DISTRIBUTION IN EXPERIMENT
LV4 / JK4Pu

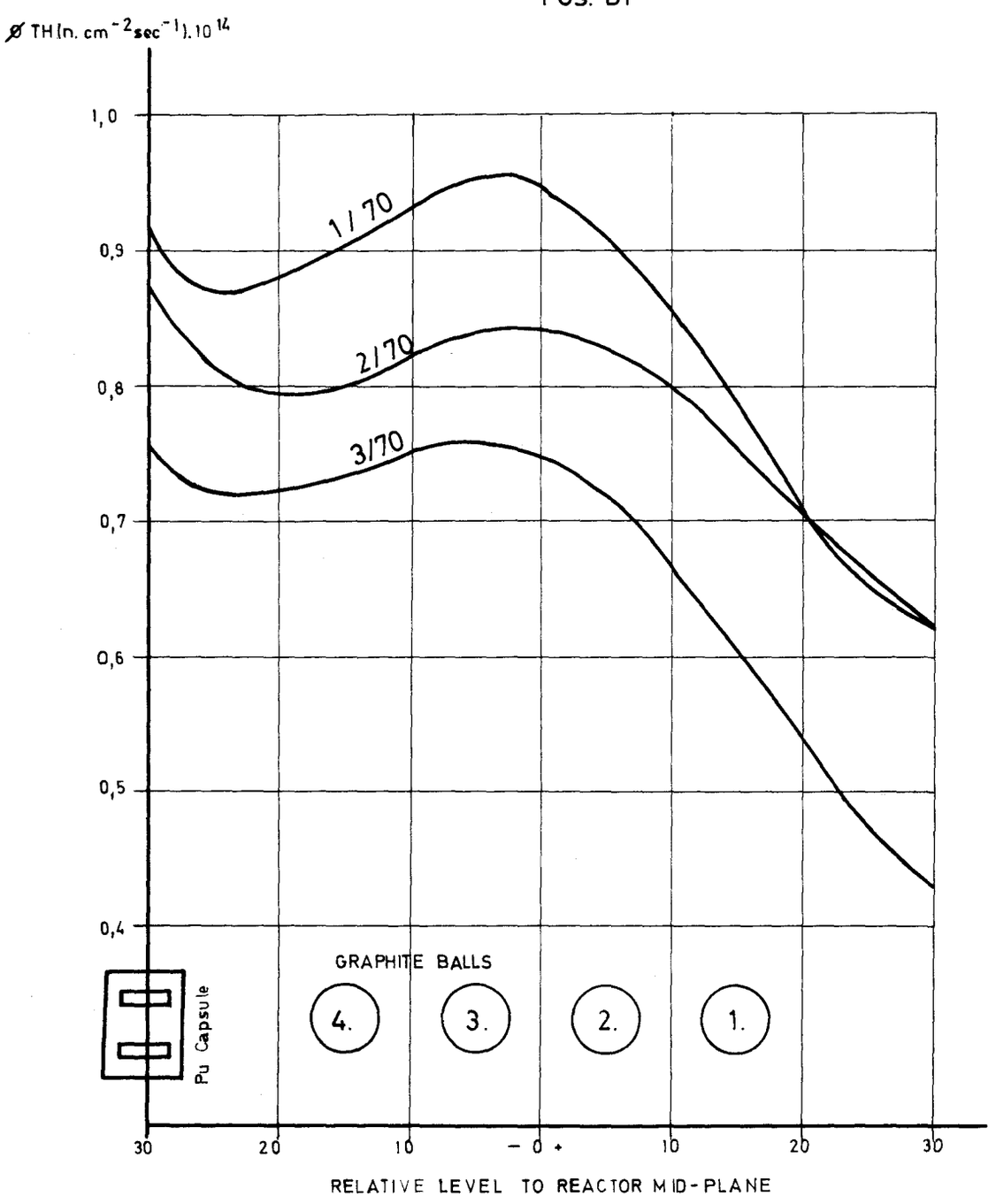


Figure 12 : BURN-UP DIAGRAM OF A JK 4 Pu

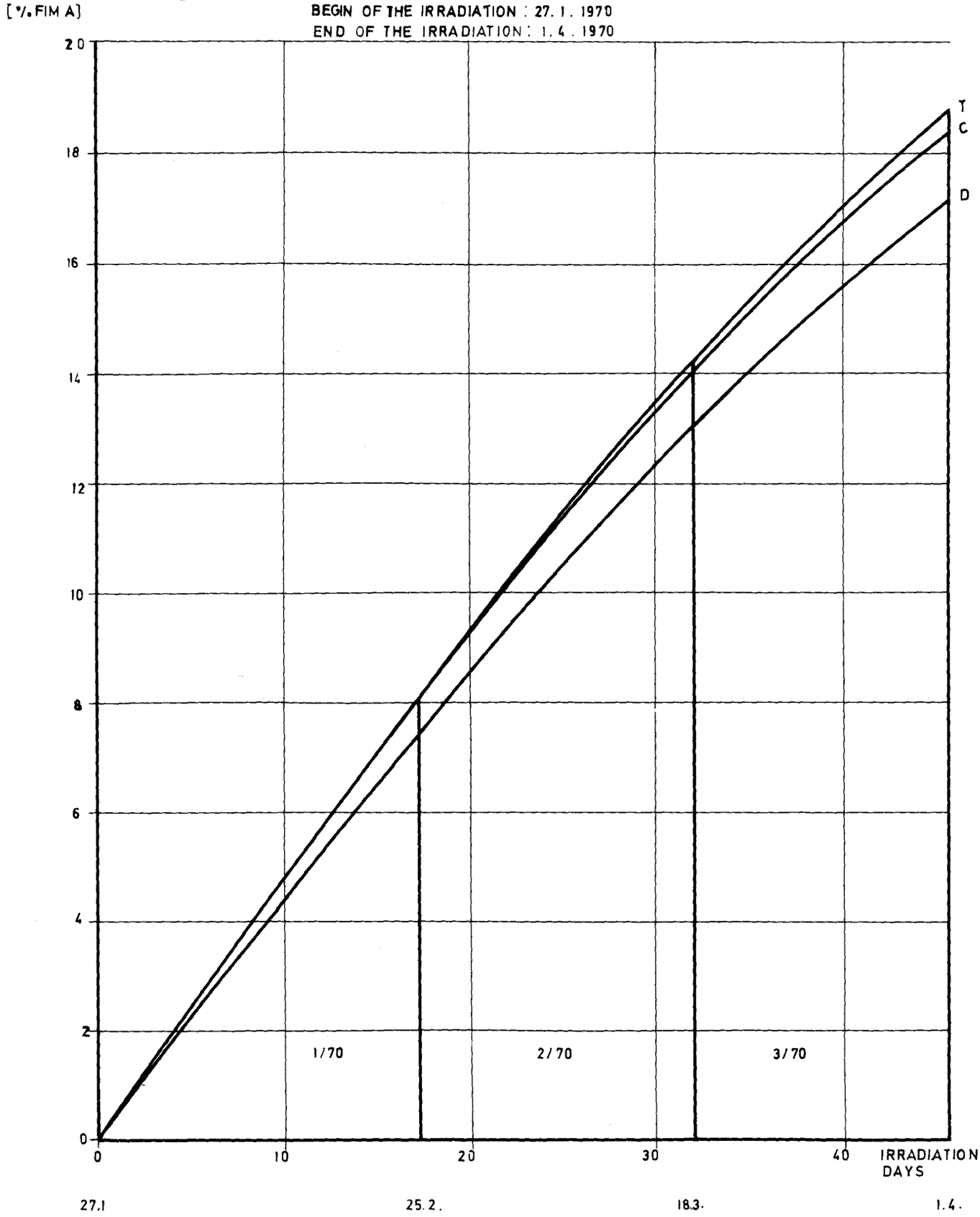


Figure 13 : Cut aluminium capsule after irradiation

Figure 14 : Irradiated coated particles in container 3

Figure 15 : Container 1 (D particle)

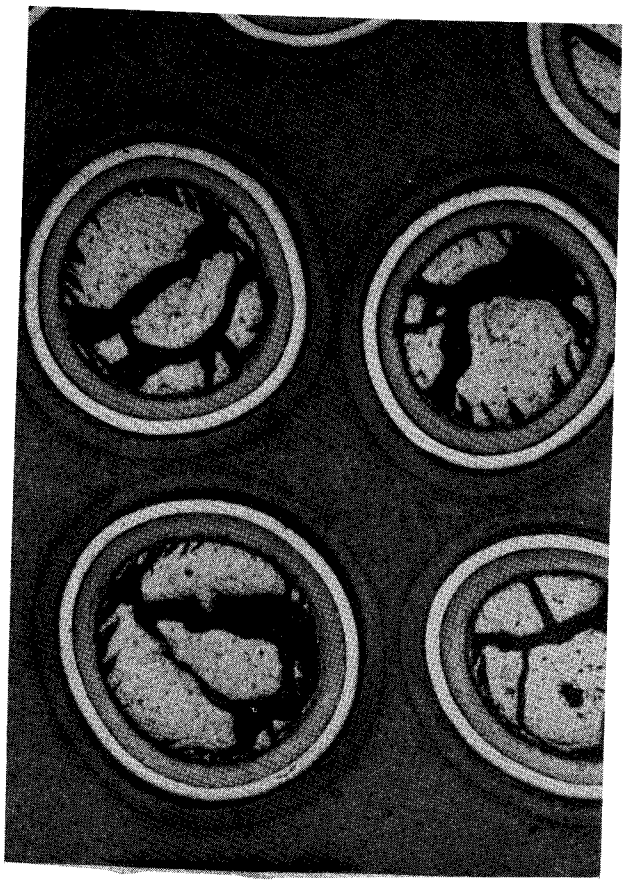


Abb. 1
(2817)

50 : 1

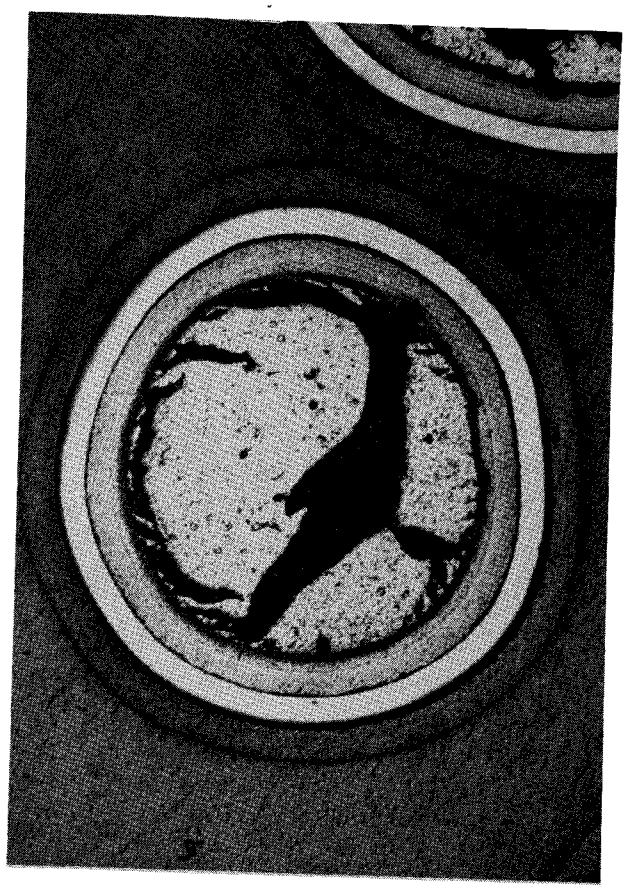


Abb. 2
(2818)

80 : 1

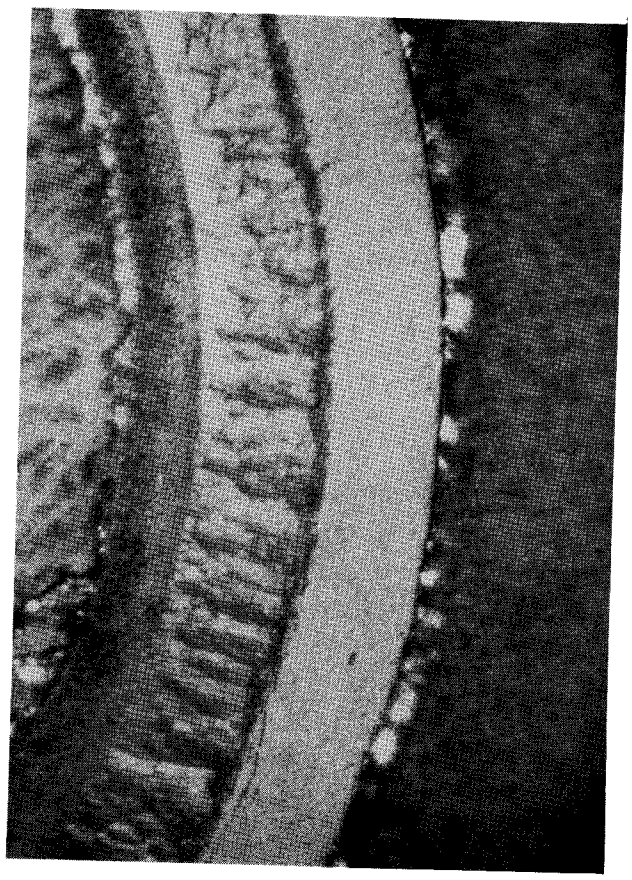


Abb. 3
(2819)

500 : 1
Pol

Figure 16 : Container 5 (particle after annealing at 1350° C)

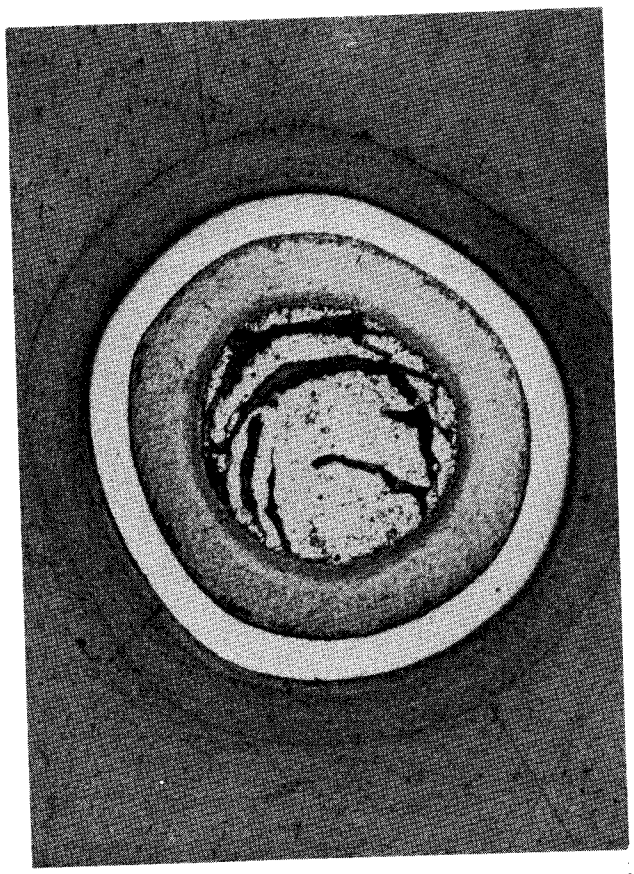


Abb. 17
(2748)

100 : 1

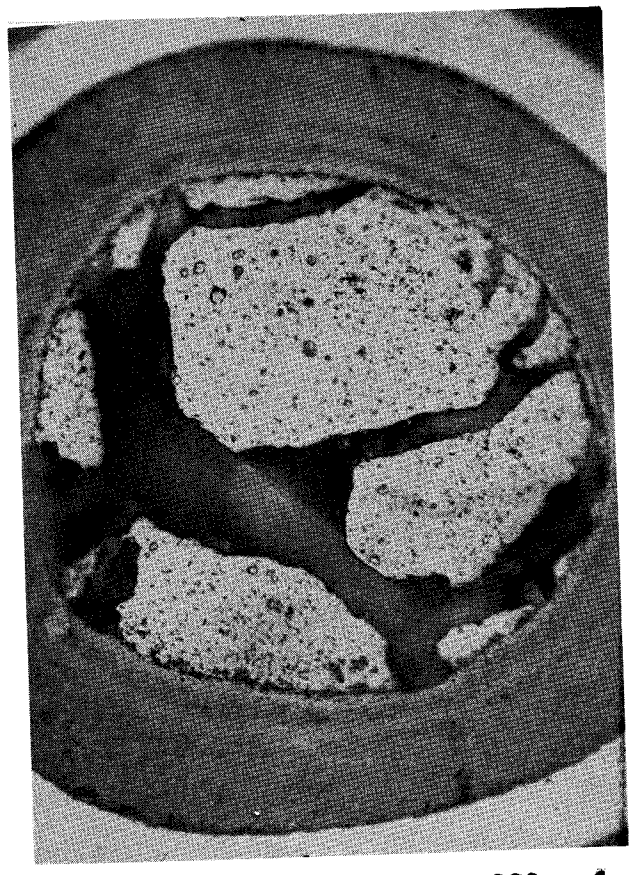


Abb. 18
(2750)

200 : 1

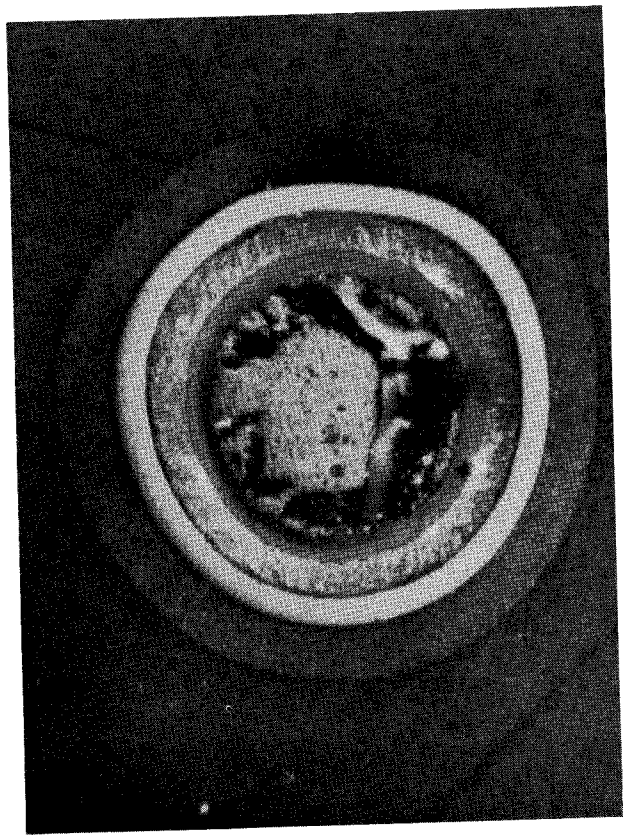


Abb. 19
(2752)

100 : 1

Abb. 20
(2751)

500 : 1

Figure 17 : Container 3 (T particle)

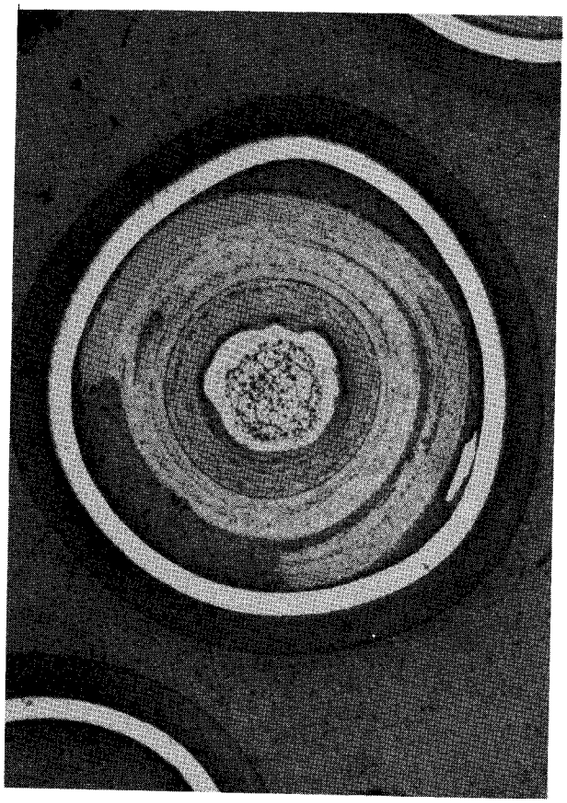


Abb. 8
(2820)

100 : 1

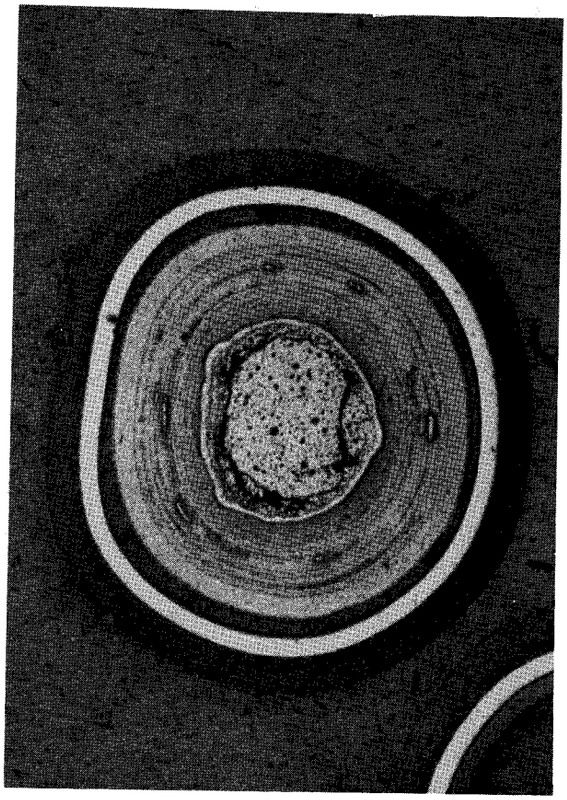


Abb. 9
(2824)

100 : 1

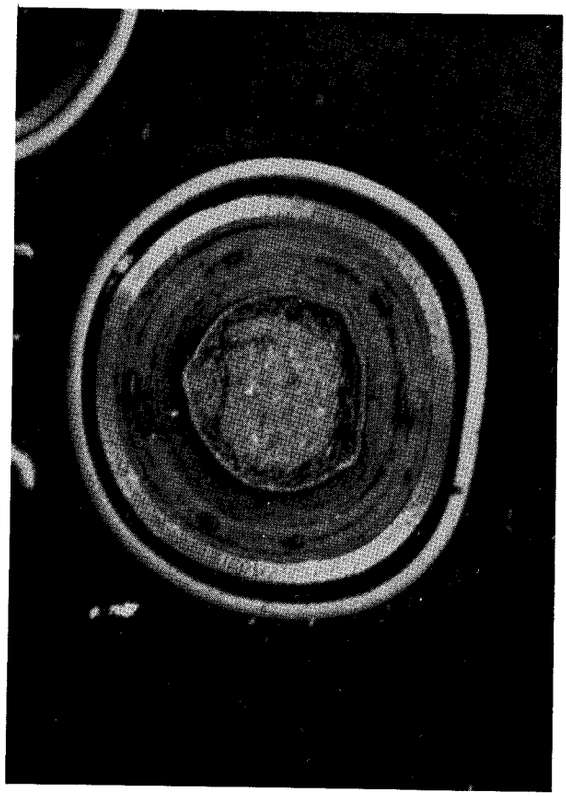


Abb. 10
(2825)

100 : 1

Figure 18 : Container 3 (T particle)

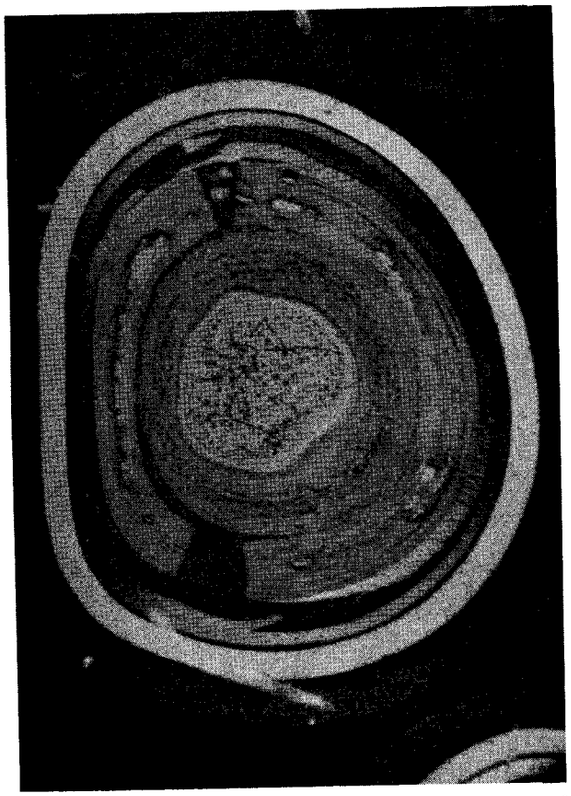


Abb. 11
(2822)

100 : 1
Pol

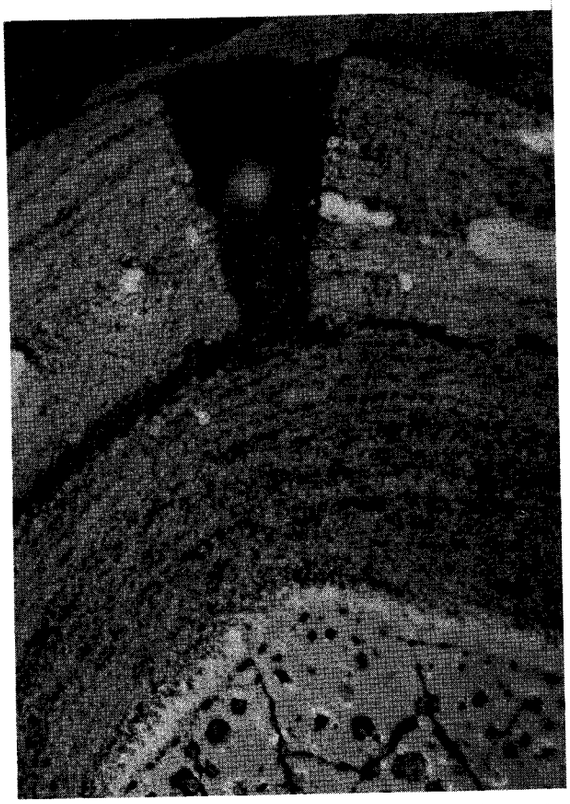


Abb. 12
(2823)

500 : 1

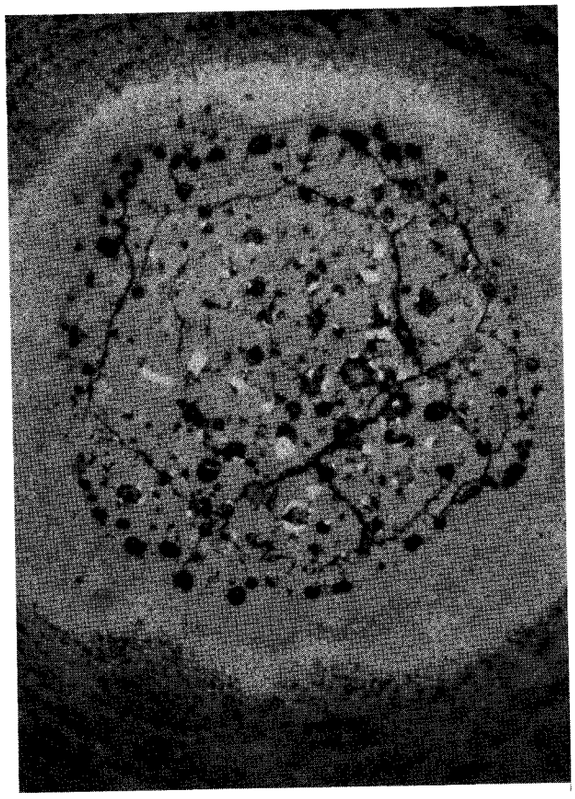


Abb. 13
(2824)

500 : 1

Figure 19 : Container 8 (T particles after annealing at 1350° C)

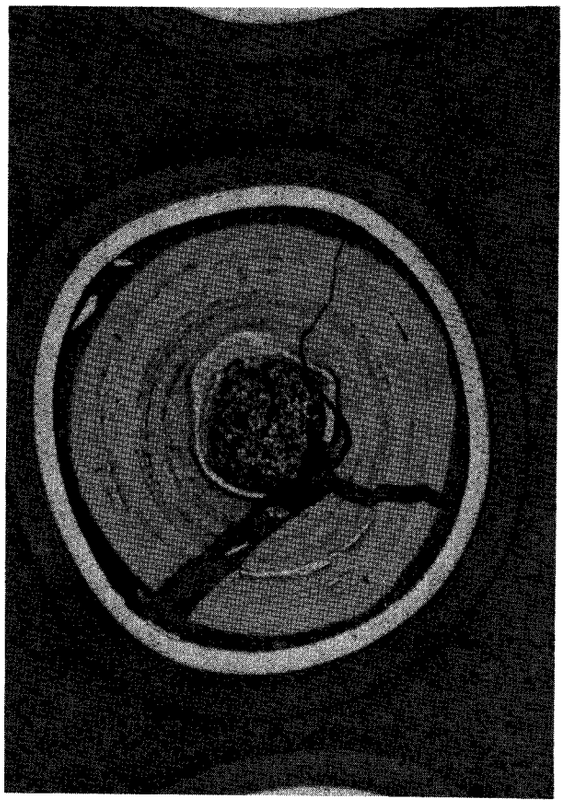


Abb. 21 (2754) 100 : 1

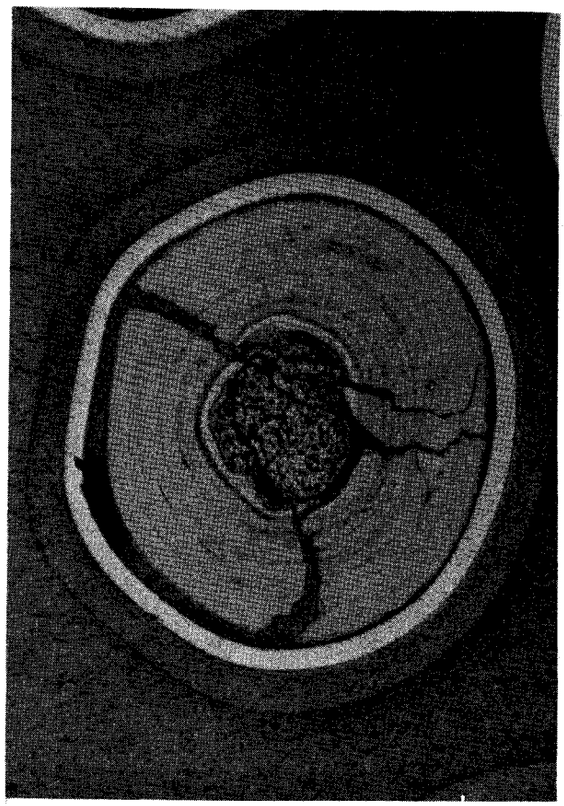


Abb. 22 (2755) 100 : 1

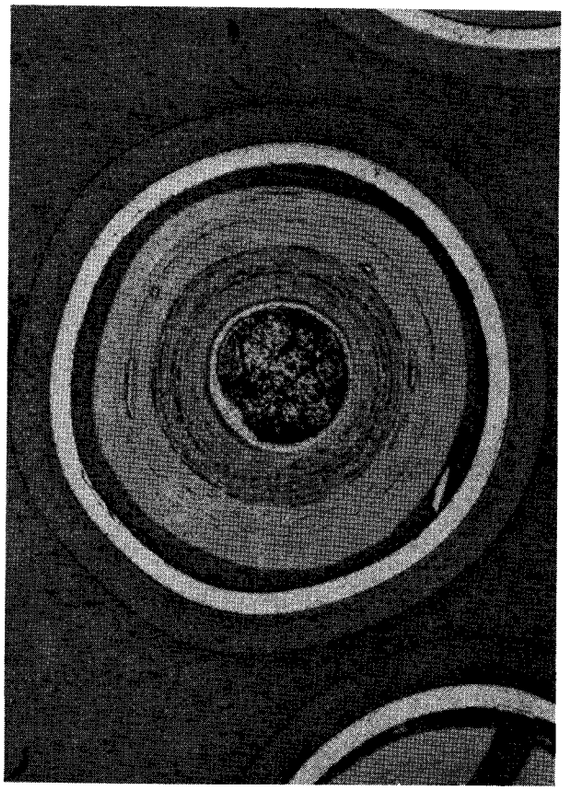


Abb. 23 (2756) 100 : 1

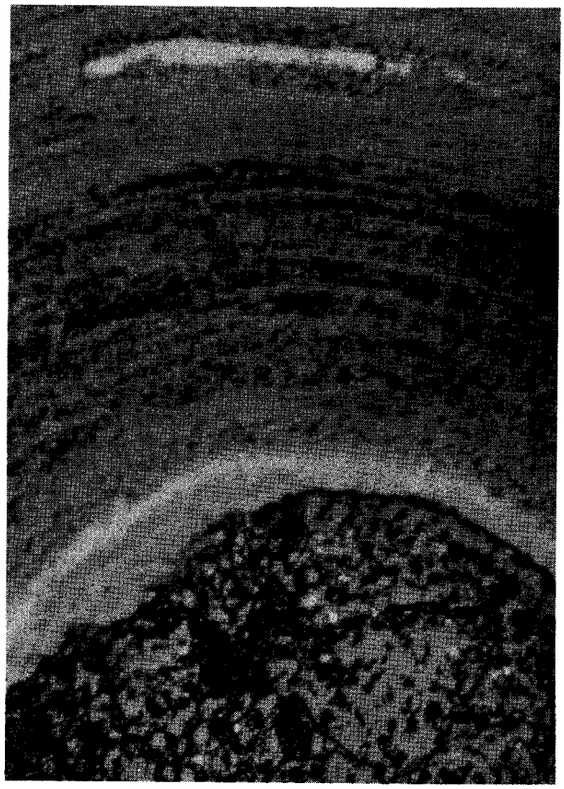


Abb. 24 (2758) 500 : 1

Figure 20 : Containers 4 and 7 (C particles)

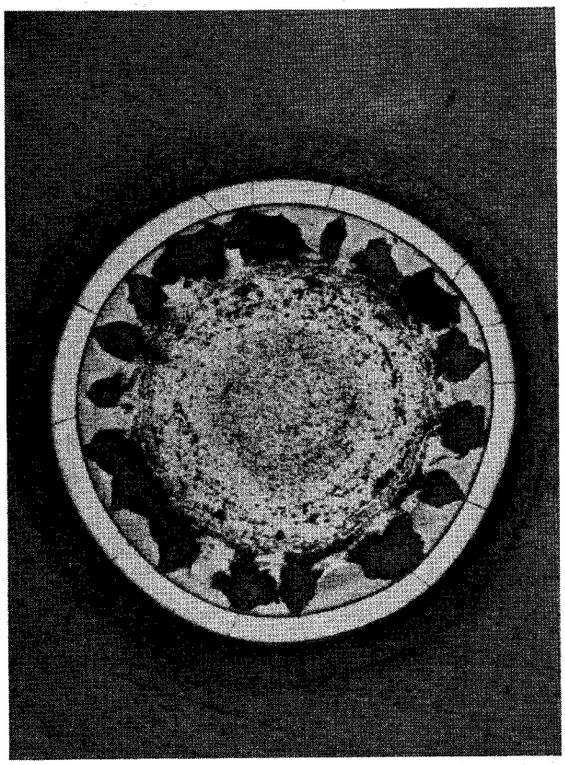


Abb. 14
(2759)

100 : 1

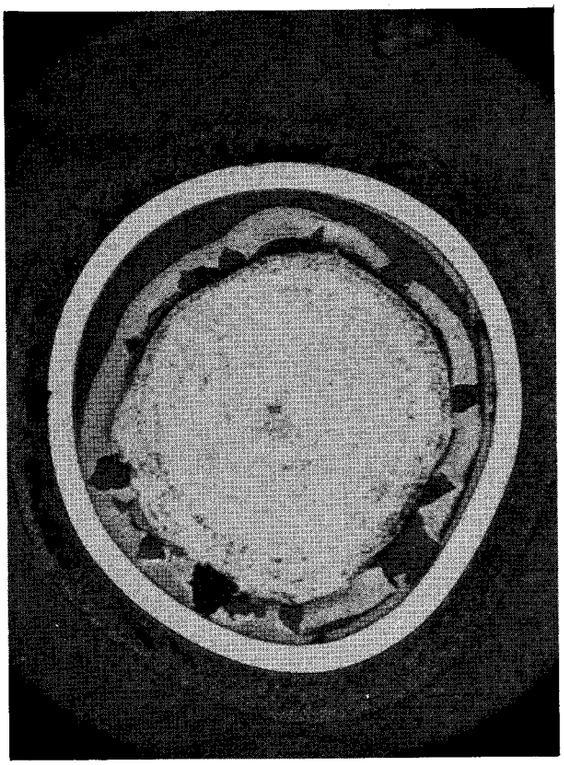
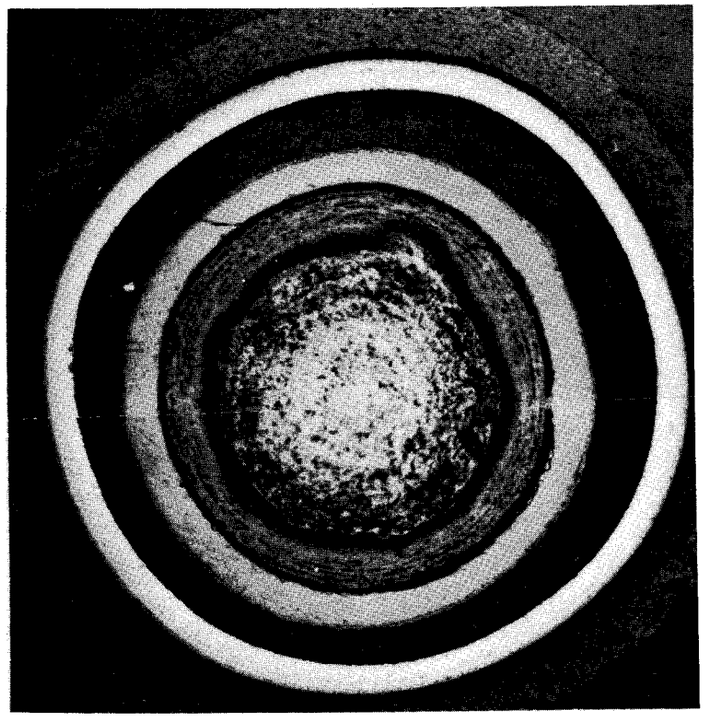


Abb. 15
(2760)

100 : 1

Container 4



Container 7

Figure 21 : Container 2 (particle after annealing at 1350° C and at 1500° C successively)

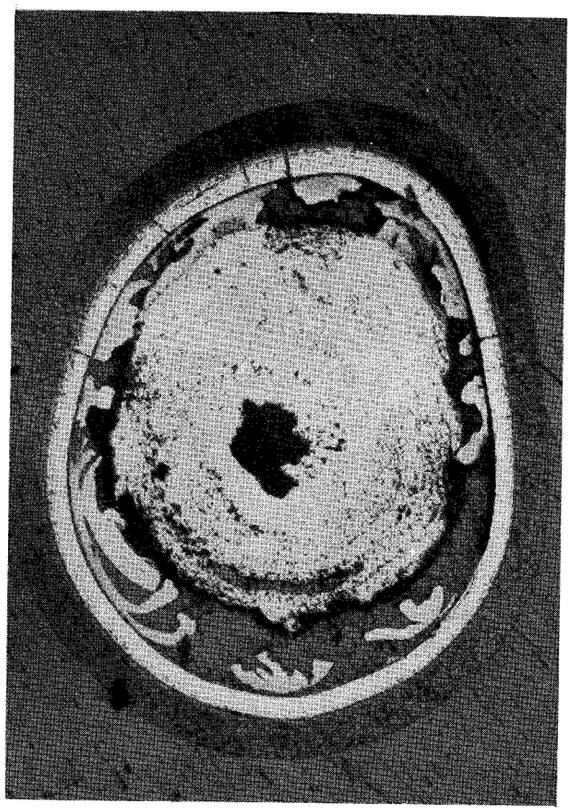


Abb. 4
(2826)

100 : 1

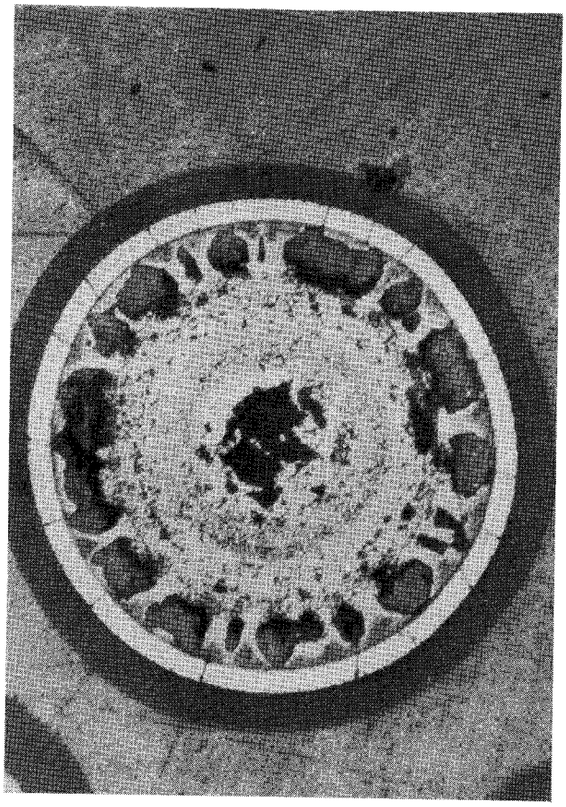


Abb. 5
(2827)

100 : 1

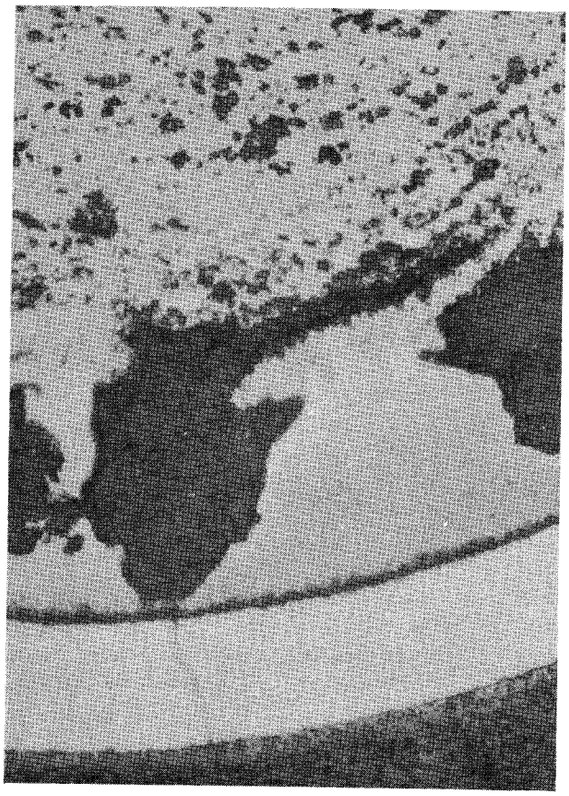


Abb. 6
(2829)

500 : 1

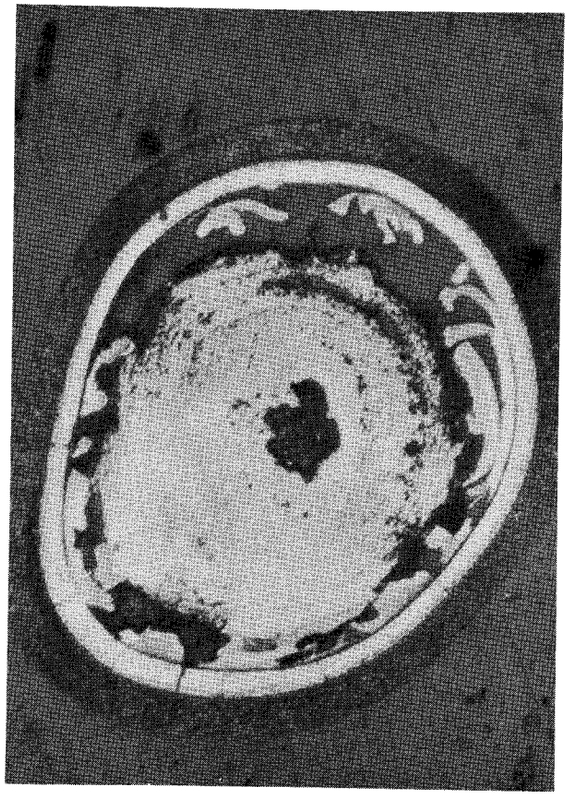
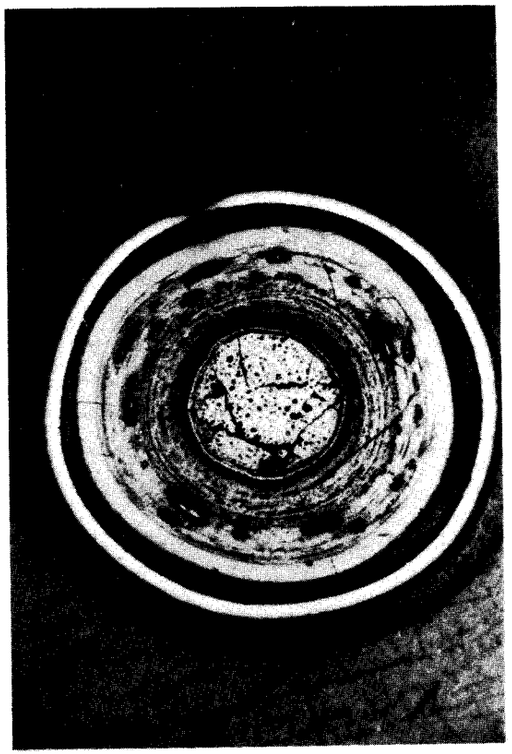


Abb. 7
(2828)

100 : 1

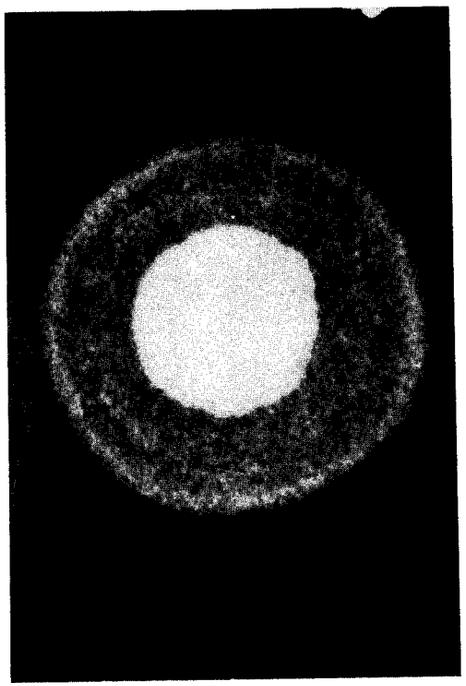
Figure 22 : Container 1 - Particle 3 T

12295



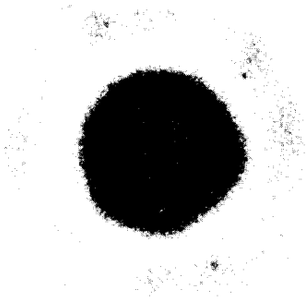
(x 95)

polished section



(x 95)

X-ray after fabrication

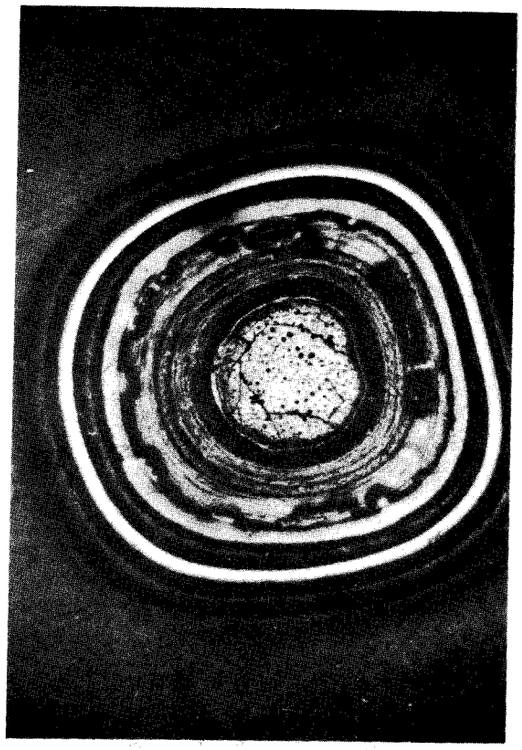


(x 95)

γ - autoradiography after irradiation

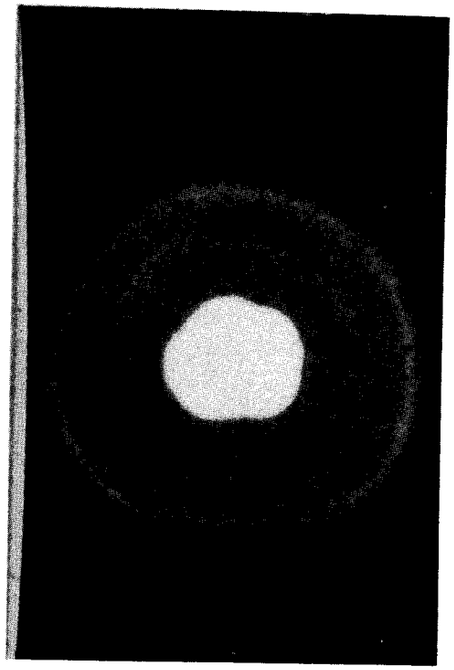
Figure 23 : Container 2 - Particle 4 T

12296



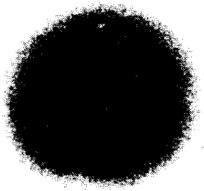
(x 95)

polished section



(x 95)

X-ray after fabrication

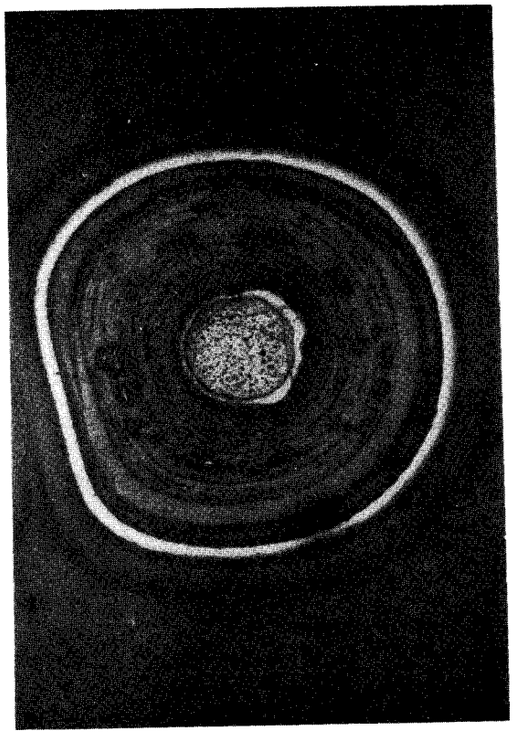


(x 95)

γ - autoradiography after irradiation

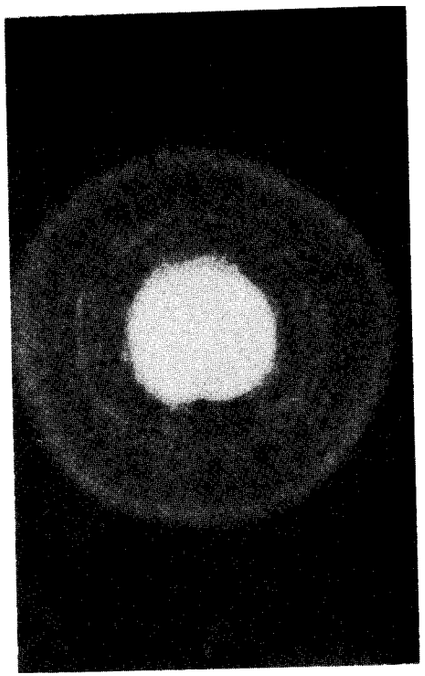
Figure 24 : Container 3 - Particle 7 T

12299



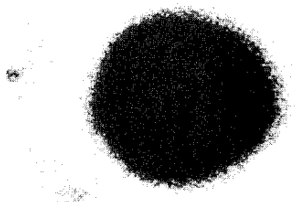
(x 95)

polished section



(x 95)

X-ray after fabrication

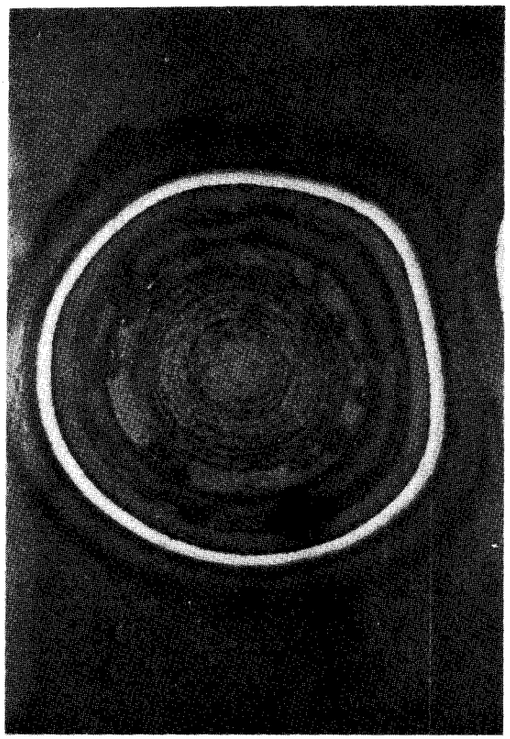


(x 95)

γ - autoradiography after irradiation

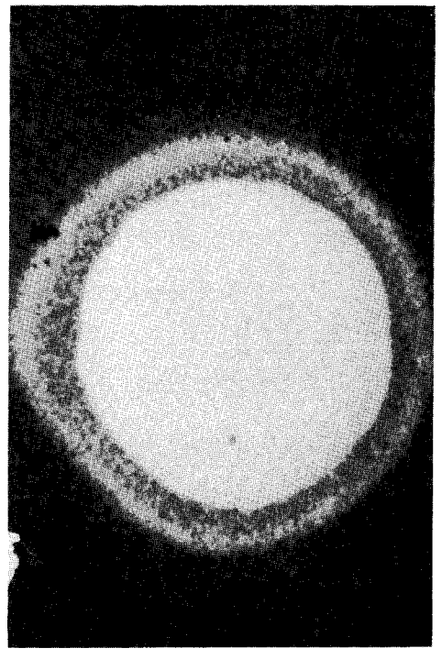
Figure 25 : Container 3 - Particle 8 C

12656



(x 95)

polished section



(x 95)

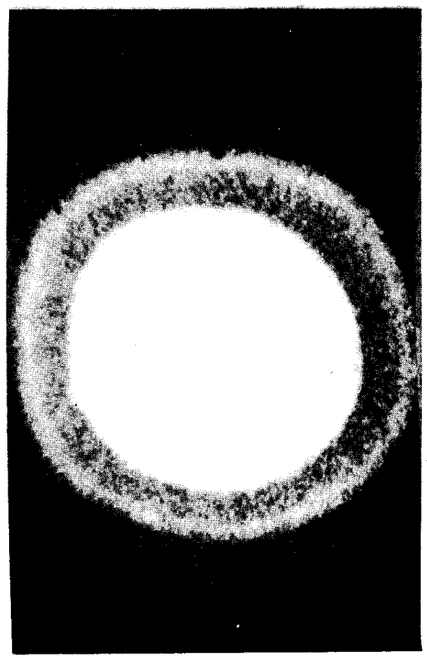
X-ray after fabrication

Figure 26 : Container 4 - Particle 6 C

12654

(x 95)

polished section



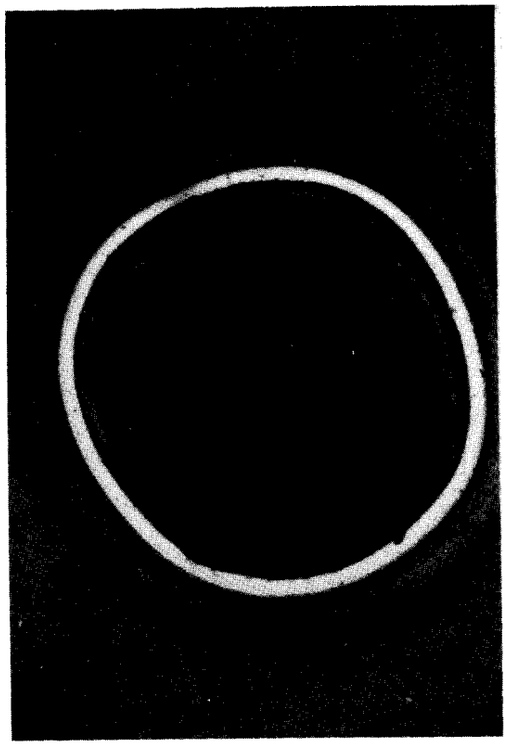
(x 95)

X-ray after fabrication

(x 95)

γ - autoradiography after irradiation

Figure 27 : Container 5 - Particle 5 C



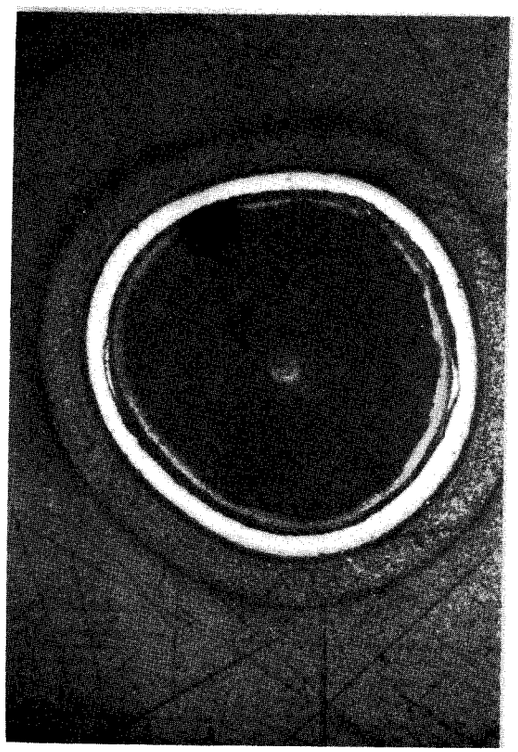
(x 95)

polished section

(x 95)

X-ray after
fabrication

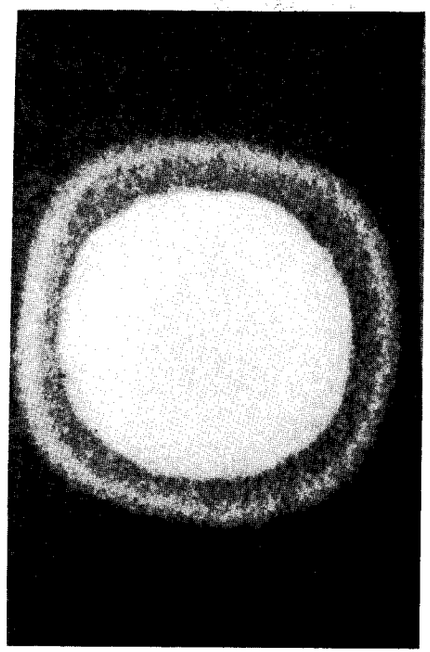
Figure 28 : Container 6 - Particle 9 C



(x 95)

polished section

12567

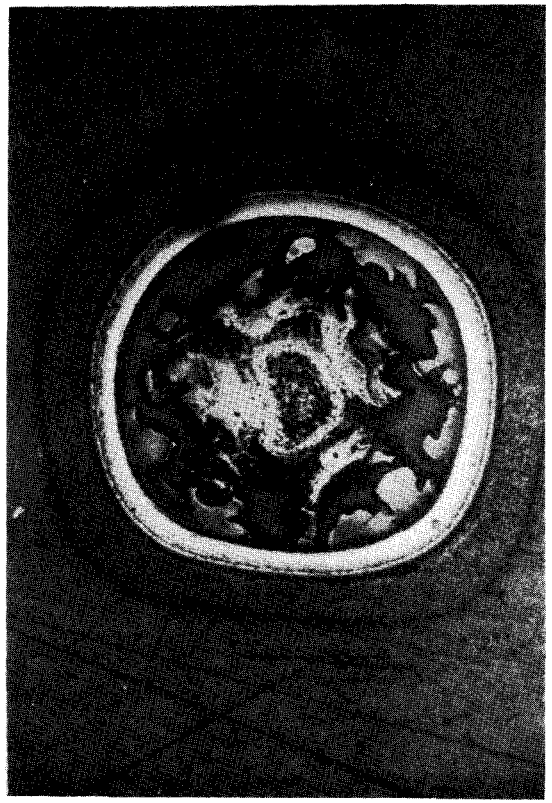


(x 95)

X-ray after
fabrication

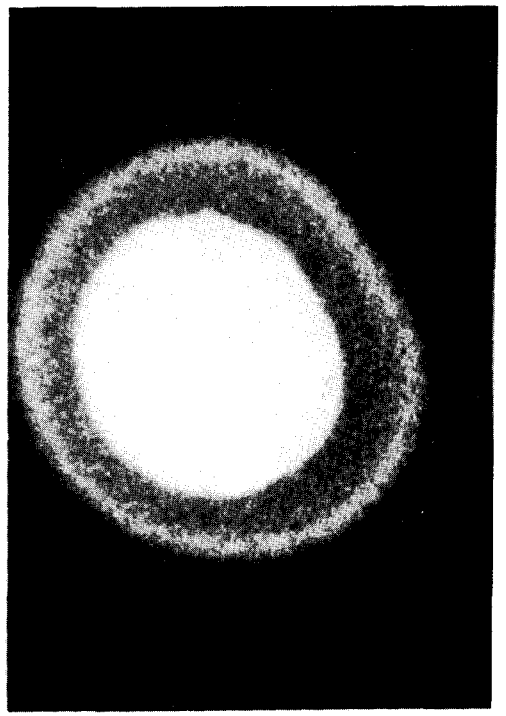
Figure 29 : Container 7 - Particle 13 C

12661



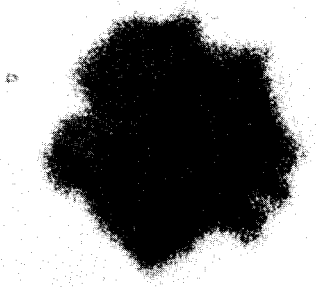
(x 95)

polished section



(x 95)

X-ray after fabrication

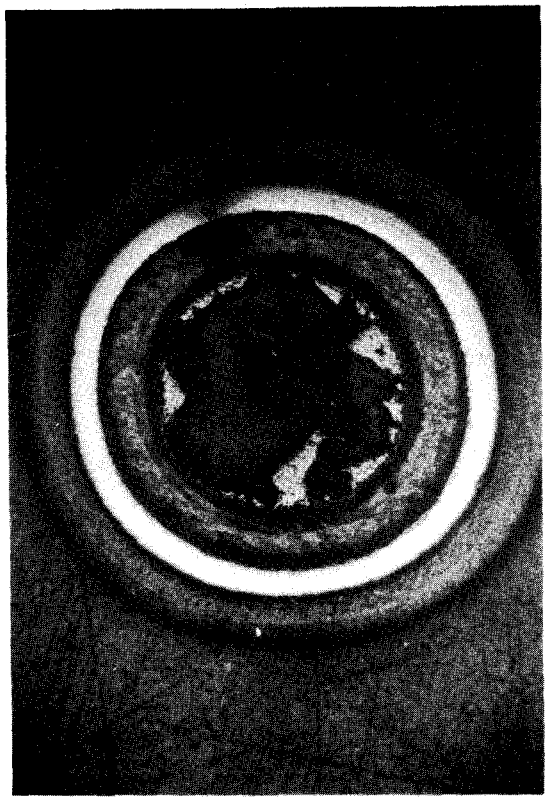


(x 95)

γ - autoradiography after irradiation

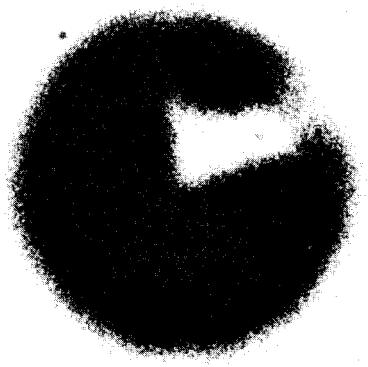
Figure 30 : Container 8 - Particle 14 D

12653



(x 95)

polished section



(x 95)

γ - autoradiography
after irradiation

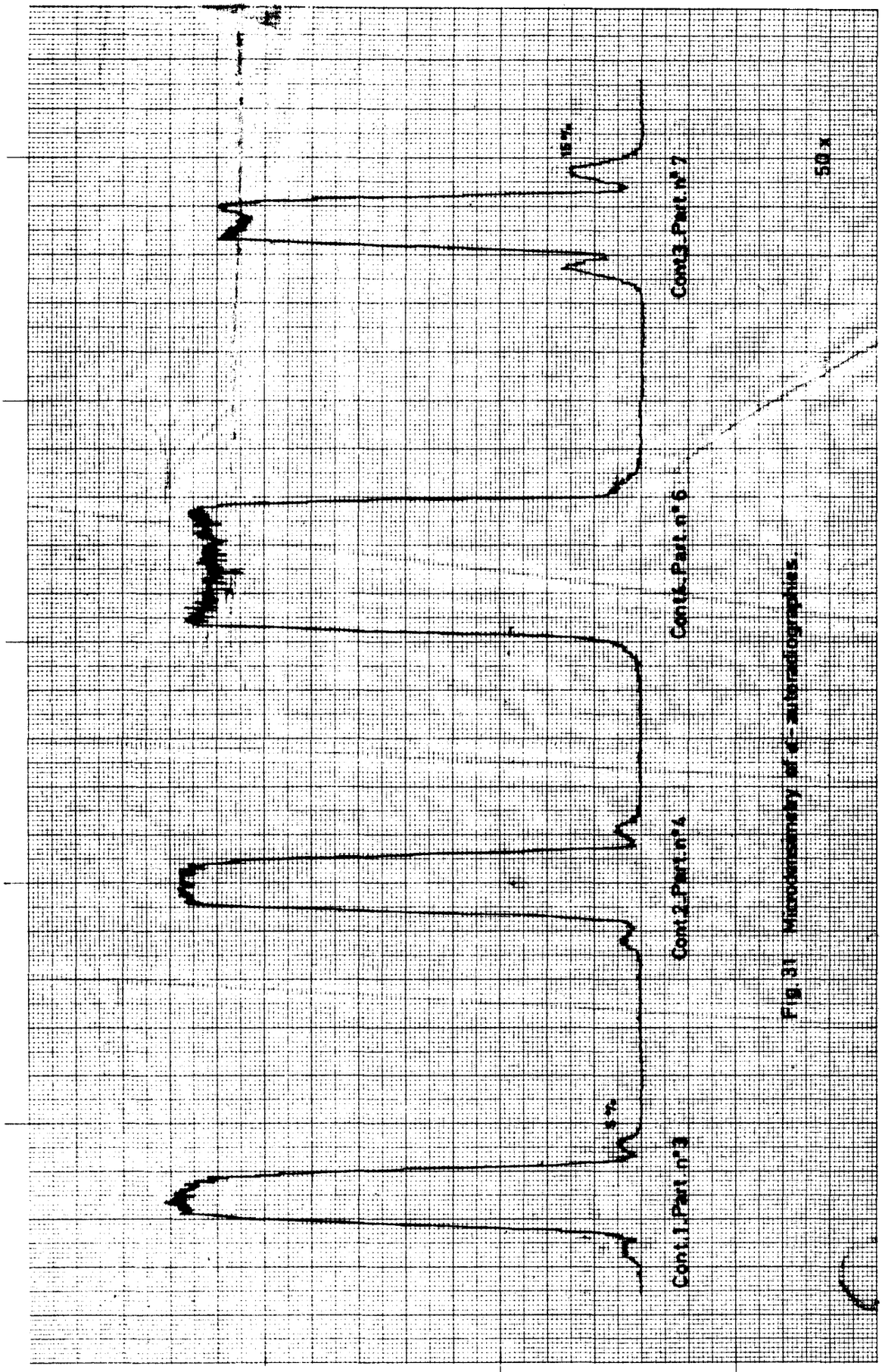


Fig 31 Microddensimetry of α-autoradiographies.

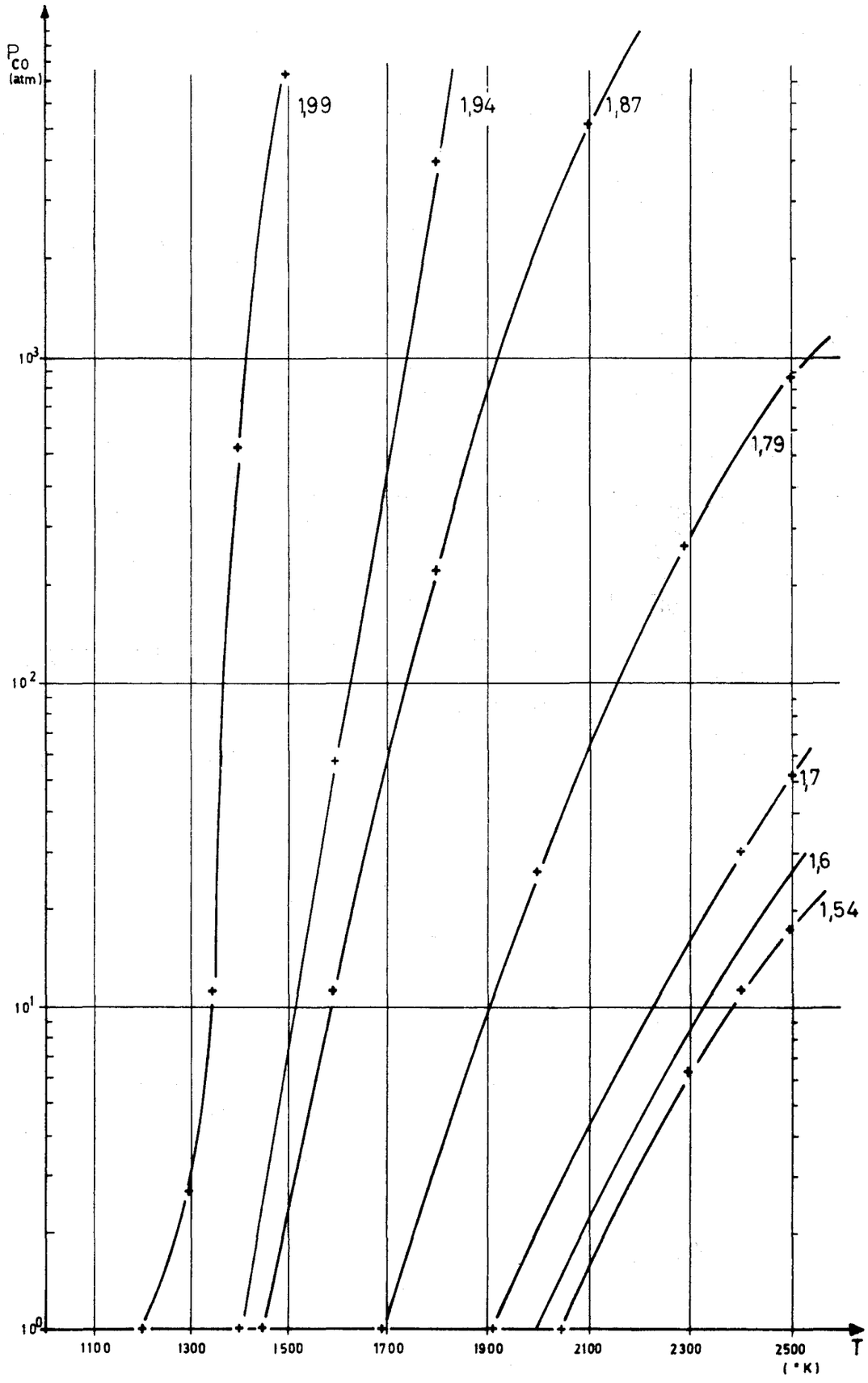


Figure 32 : CO equilibrium pressure for several PuO_2 stoechiometries

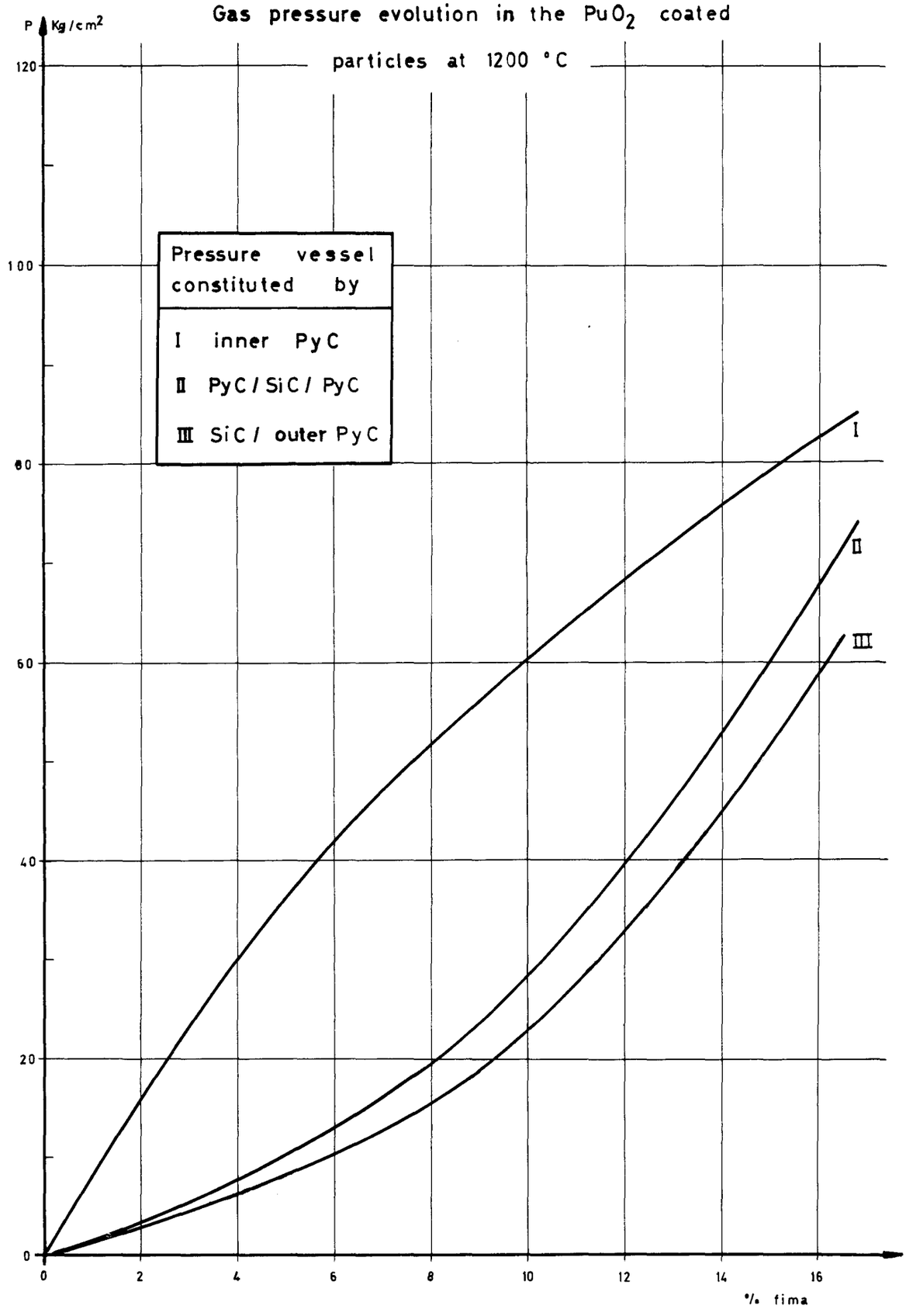

Gas pressure evolution in the PuO_2 coated particles at 1200 °C
P Kg/cm²
120
100
80
60
40
20
0
Pressure vessel constituted by
I inner PyC
II PyC / SiC / PyC
III SiC / outer PyC
I
II
III
0
2
4
6
8
10
12
14
16
% fima

Figure 33

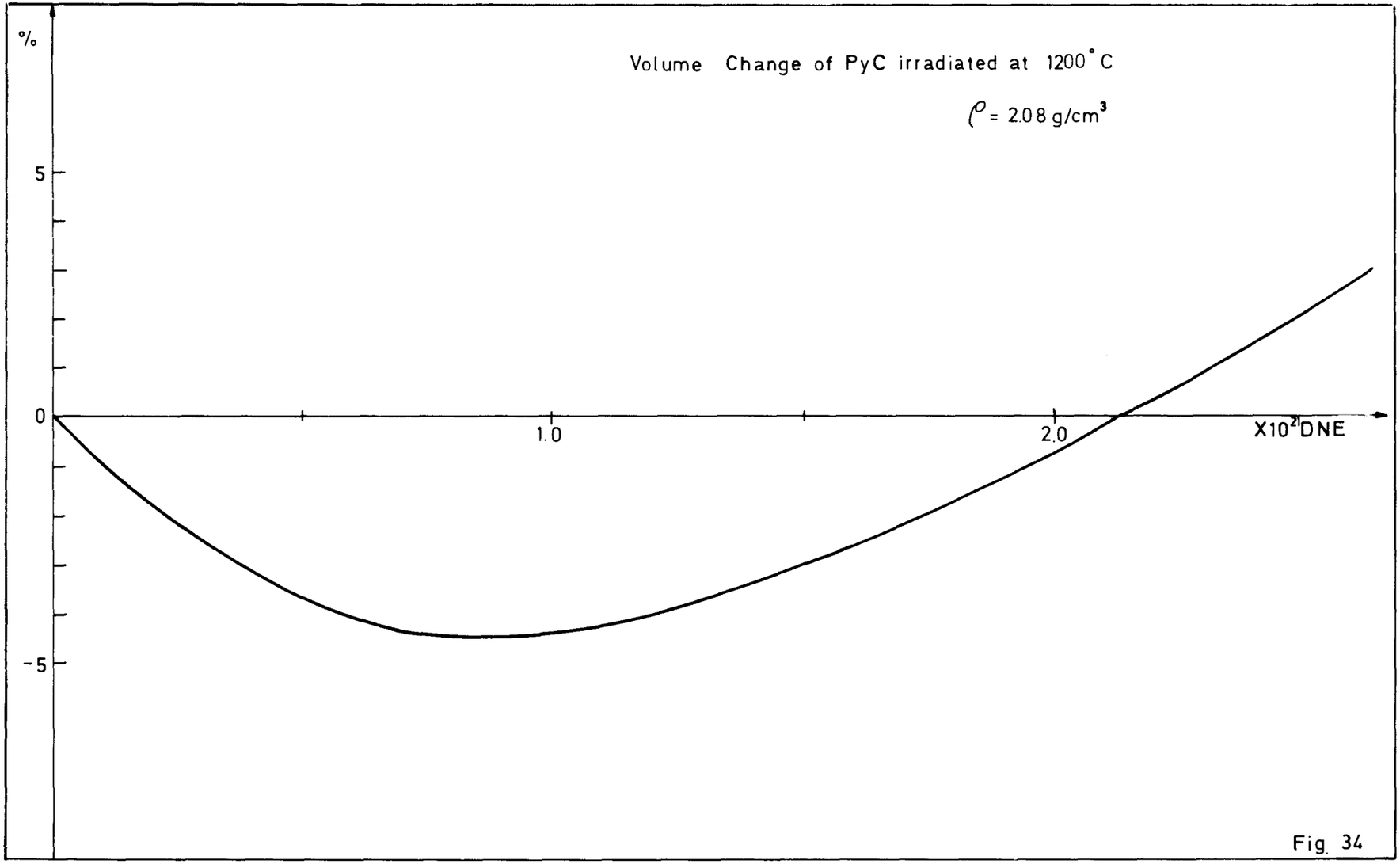

%
Volume Change of PyC irradiated at 1200° C
ρ = 2.08 g/cm³
5
0
-5
1.0
2.0
X10²¹DNE

Fig. 34

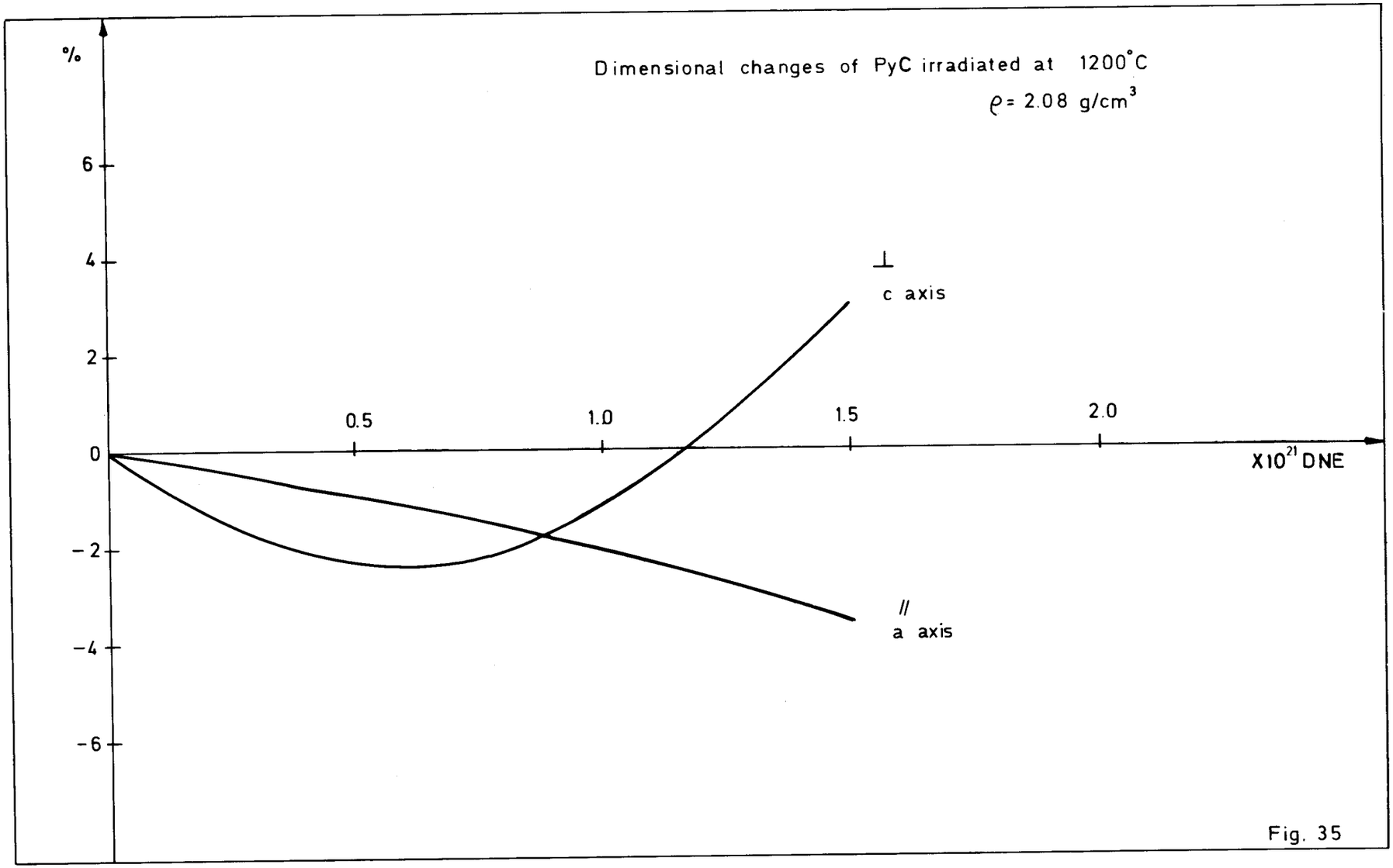

Dimensional changes of PyC irradiated at 1200°C
ρ = 2.08 g/cm³
%
6
4
2
0
-2
-4
-6
0.5
1.0
1.5
2.0
X10²¹ DNE
⊥
c axis
//
a axis

Fig. 35

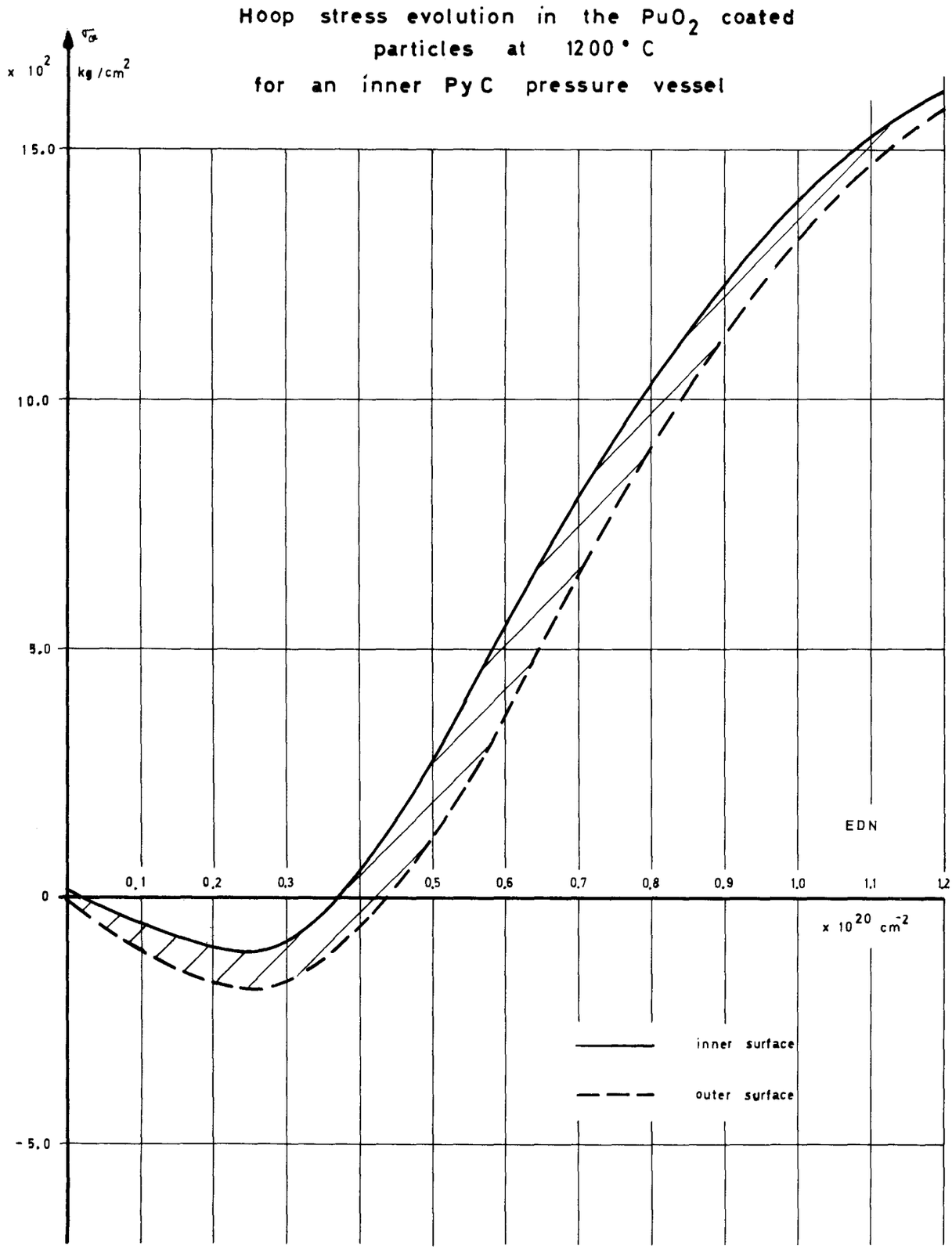

Hoop stress evolution in the PuO_2 coated particles at 1200 °C for an inner PyC pressure vessel
σ_θ
x 10^2 kg/cm^2
15.0
10.0
5.0
0
-5.0
0.1
0.2
0.3
0.5
0.6
0.7
0.8
0.9
1.0
1.1
1.2
EDN
x 10^{20} cm^{-2}
inner surface
outer surface

Figure 36

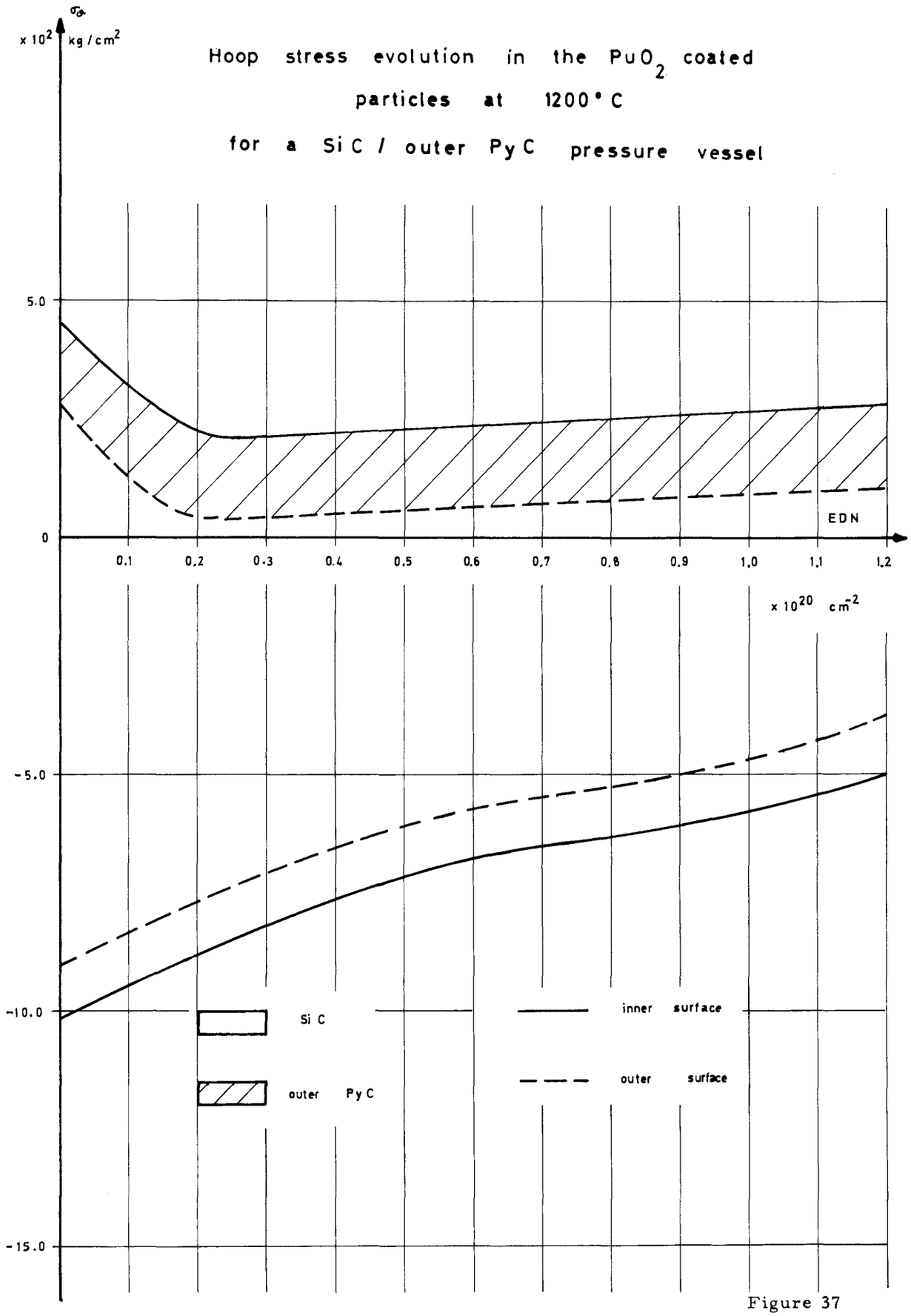

Hoop stress evolution in the PuO_2 coated particles at 1200°C
for a SiC / outer PyC pressure vessel
σ_θ
x 10^2 kg / cm^2
5.0
0
-5.0
-10.0
-15.0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1.0
1.1
1.2
EDN
x 10^{20} cm^{-2}
Si C
outer Py C
inner surface
outer surface

Figure 37

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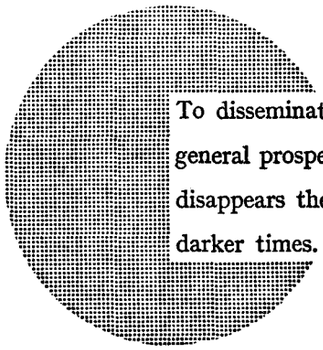

To disseminate knowledge is to disseminate prosperity — I mean general prosperity and not individual riches — and with prosperity disappears the greater part of the evil which is our heritage from darker times.

Alfred Nobel

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