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Project Fuel Development

Annual Report 1980

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PROJECT FUEL DEVELOPMENT

ANNUAL REPORT 1980

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1. INTRODUCTION

1.1 Project Aims and Scope 1980

The overall project aims were set out in the previous year's report[1]. Basically these have not changed. The activities continue on lab-scale production of uranium-plutonium carbide fuel for the fast reactor using gelation methods, irradiation testing and performance evaluation. Whereas in earlier years a balance was maintained between research and development or with emphasis on research in 1978/79, 1980 was marked by a concentrated equipment development effort for an increased throughput with improved process control and product reproduceability and installation of new equipment for large pin fabrication.

1.2 International Developments

In the year immediately following the publication of the INFCE reports it was perhaps too much to expect new and more rapid progress in the direction of the deployment of the Fast Breeder Reactor. INFCE confirmed the correctness of many countries' decision to develop breeder technology (with reprocessing) but in the Western world (with the possible exception of France) hesitation and caution prevailed. In spite of (or because of) a development vacuum sphere pac fuel produced by gelation methods still appears an attractive candidate for both LWR and FBR fuel with attention being given by several groups to the hybrid

concept of pellets produced from gel derived particles[2,3,4]. Studies being carried out in the USA, partly with EIR sponsorship are looking at the safeguardability of the wet-route fuel fabrication concept compared to a pellet route[5]. The interest in extending the burn-up of Light Water Reactor (LWR) fuels is also leading to the demonstration of sphere pac oxide fuels where it is possible that an improved fuel clad interaction may give sphere pac fuels an advantage.

During 1980 EIR was exposed directly to the effects arising out of the fear of nuclear weapons proliferation. In spite of general agreement on the value of the proposed joint EIR-US irradiation of advanced fuel in the US Fast Flux Test Facility, and good progress at the technical level, political clouds in Washington affected US-Swiss collaboration in several ways. One was to delay the signing of the EIR-US-DOE FFTF agreement until early 1981. Preparations however continued at EIR.

1.3 Project programme 1980

A Summary of the project programme as given in the project request for 1980 can be seen from the following headings.

1.3.1 "Fabrication of FFTF fuel and supporting development"

Large efforts were devoted to commissioning the new gelation box. Far more time was needed to get the individual process stages funtioning correctly and reliably than was forseen. Only by the end of the

year, when the equipment was performing without trouble could the process (using UC as the dummy material) be demonstrated satisfactorily. Improvements were made throughout the fabrication line including the drying, calcining and sintering stages and in the treatment of α-active ammonia wastes (volume reduction by distillation). Some progress was made in systematic documentation of the development results and establishing a standard fabrication flow sheet.

In parallel,new analytical equipment was installed or completed commissioning - Leco oxygen analyser, air densitometer, BET surface analyser and the X-ray diffractrometer attachment to the X-ray fluoroscen-spectrometer.

Fabrication of FFTF fuel was naturally pushed back due to the extended commissioning period and due to the delay in signing the FFTF agreement.

1.3.2 "Preparation of a pin fabrication line and start of manufacture" (FFTF)

As with the fuel line heavy investment of time and materials was made in 1980 on major modifications to the pin fabrication boxes. The vibro-filling box was completely rebuilt (following the developments reported in 1979). A new welding chamber was installed, the weld box modified to take the revised pin transfer and sealing system and transfer jigs and tools were manufactured and tested. The decontamination box was completed and will be later fitted out following the development of new pin decontamination procedures. The basic pin design, modified for the sphere pac fuel and for EIR's handling

restrictions was completed. The γ-scan (axial densitrometry) equipment was modified and prepared for 2½ m pins, including the data handling programme.

1.3.3 "Post irradiation examination and analysis of irradiation performance"

Examination of the final set of four pins irradiated in the DIDO reactor (UK) proceeded only slowly during the year. This was partly due to operational problems in the Hot Labor. Nevertheless it was determined that unlike six of the eight earlier DIDO pins, pins 9-12 were all intact. Axial γ-scanning, profilometry gas,analysis,sample cutting and burn-up analysis were performed.

A post irradiation examination report on the clad carburization test Filos 07[6] *was published as was the report on the performance of the EIR pin irradiated in DFR*[7]*. Both experiments provided valuable information and extended the data base on sphere-pac fuel. The 1979 presentation (Hamburg, German Atom Forum) on the comparison between pellet - sphere-pac carbide behaviour was published in extended form*[8]*.*

Work continued on evaluating fuel performance with the strong support of Oregon State University (OSU) whose contract was extended. The thermal modelling of sphere pac fuel in the SPECKLE code was sufficiently developed to carry out calculations on the possible start up behaviour (center fuel temperatures, power-to-melt) of sphere pac carbides in the FFTF[9]*. Towards the end of the year attention turned more towards methods of modelling fuel-clad mechanical*

interaction, supported by simple lab experiments at OSU. In addition the whole SPECKLE code is to be revised to improve its flexibility and operational efficiency as well as correcting modelling deficiencies discovered during the 1980 studies.

1.3.4 "Follow and contribute to FBR carbide fuel development together with other groups"

With the completion of the joint report "Pellet and sphere pac (UPu)C fuel comparative irradiation tests"[8] a decade of close and successful collaboration on carbide fuels with the Belgian research Institute SCK/CEN, came effectively to an end. The collaboration with the Fast Breeder Project (PSB) of the Kernforschungszentrum Karlsruhe (KfK) continued. The EIR pin Mol-11/K5 produced for KfK for irradiation in the Belgian reactor BR-2 was mounted in its irradiation capsule ready for testing at the end of the year. The collaboration with the United Kingdom Atomic Energy Authority (UKAEA) was extended by one year (again in 1981).

In the USA several meetings and visits took place between EIR and the US participants in the joint FFTF test - Westinghouse Advanced Reactors Division (WARD), the FFTF teams in Richland and the US-DOE. Oregon State University was represented at some of the meetings concerning performance analysis. Technical discussion also continued with General Atomic Co, General Electric, and Oak Ridge National Laboratory. As well as modelling studies Oregon State Uni-

versity began an investigation of the diversion resistance of sphere-pac fuel in a safeguarded fuel cycle with EIR and the Electric Power Research Institute as co-sponsors. EIR also received visits from sphere pac fuel experts from Italy and Japan.

1.3.5 "Prepare post FFTF programme"

Several possibilities for the continuation of the fuel development programme after 1982 were under discussion during the year. It is too early to say which of several factors will influence the direction of the programme most: successful completion of the FFTF pins leading to further large scale tests; continuing good irradiation test results bringing an extension of testing under off-normal conditions; Collection of performance data for model input; plant developments in direction of LWR oxide particle fuels; use of sol-gel for waste conditioning; the choice will depend on both internal and external factors, not the least the atmosphere in nuclear fuel cycle R&D "post - Carter"!

In the meantime, during 1980 and continuing into 1981, the EIR management have reduced the effort available by about 40% to a stable level designed to bridge the programme review period. The Advisory Commission of EIR has set up an ad hoc working group to review these options taking into account Swiss needs. The pilot plant concept study for carbide fuels has been reduced to near zero effort but with a study planned in 1981 to have further plant development include improvements in the FBR and LWR fuel cycle.

References to ch 1

1. Plutonium Brennstoffprogramm. Jahresbericht 1979
EIR Report No. 395

2. SCK-CEN Annual Scientific Report 1978.
"New Fabrication methods", page 1-4/1-6

3. Nuclear Fuel Pellets Fabricated from Gel-Derived Microspheres
R.B. Mathews, P.E. Hart
Journ.Nucl.Mat. 92 (1980)207-216

4. Report on a paper by JFW Bishop to the British Nuclear Energy Society. October 1980
J. Daglish, p.14 Atom 291 January 1981.

5. Diversion Resistant Fuel Fabrication. Report on work carried out by Oregon State University 1980.
B. Spinrad et al
(Sponsor EPRI-EIR) OSU-NE-8103

6. FILOS 07. The post irradiation examination of a corrosion test pin clad in M 316 material and fuelled with (UPu)C microspheres.
L. Smith, G. Bart, B. Burgisser, R. Hofer, E.Keller, J. Mitar, D. Orciuolo, F. Petrik, J. Reindl.
EIR Report No. 403 July 1980.

7. Performance of a mixed carbide sphere-pac pin irradiated in the Dounreay Fast Reactor (DFR 527/1 Experiment)
K. Bischoff, L. Smith, R. Stratton
EIR Report 415 October 1980

8. Pellet and Sphere-pac (UPu)C fuel comparative irrstiation tests.
A.Delbrassine, L. Smith, Nucl. Tech 49 (June 80) pp 129-135

9. Calculations on the thermal behaviour of sphere-pac carbide fuel pins under start up conditions in the US Fast Flux Test Facility using the code SPECKLE-1
K.L. Peddicord, S.D. Montgomery, R.W. Stratton
EIR Report No. 396 / OSU-NE-8002 May 1980.

2. FUEL FABRICATION AND DEVELOPMENT

2.1. Introduction

The work of the group Plutonium Chemistry (Vorhaben Fuel Fabrication and Development) concentrated on the commissioning of the new equipment for producing (dried) microspheres using the internal gelation method. A series of runs were made using plutonium free materials in order to allow direct adjustments to be made to the equipment in the box.

As far as time and effort allowed, further development work was also carried out on the internal gelation process itself. Other experiments covered the recycling of uranium-xerogel scrap and porosity measurements on uranium-xerogel microspheres.

2.2. Commissioning the gelation equipment

At the beginning of the year the new gelation box together with the ammonia distillation box (wash fluid recycling) was virtually complete. In January the final outstanding item - the rotating drum dryer was supplied by the Matter Co. of Buchs and installed in the gelation box.

During the first part of the year, relatively small batches of fuel (a few hundred grams of mixed feed) were produced to run-in the various items of equipment. The resulting and expected technical problems which then appeared took considerable time to resolve. Among others

the following defects had to be dealt with!

- A leak in a double walled and cooled mixed feed delivery pipe due to a fracture at a weld to the metal bellows (inner tube).
- Belt-filter: The belts had to be fitted with steel wire along each side to prevent "wandering". The form of the end rollers had to be adjusted to suit and the motor drive strengthened.
- The drive to the rotating drum dryer had to be modified due to sticking caused by overheating.
- Repairs to glass containers for ammonia: glass tube failures due to thermal stresses.
- Repair of the heating element on the CCl_4 distiller (suppliers guarantee).
- Mounting a conductivity meter on the NH_3 distillation equipment.
- Replacement of the lines to the CCl_4 pump with flexible metal tubes. The rigid tubes had caused the pump bearings to fail.

By the middle of the year runs were under way with full capacity ie for the large fraction with 1.8 kg and fine fraction 1.5 kg of mixed feed. These have been choosen so that each pin can be filled with two batches of large and one batch (or ½2 batch) of fine material[1]. All the process parameters were optimized satisfactorily[2][3] with the exception of the carbon stoichiometry which will be referred to later.

The drop generation, being the start of the continuous part of the process sets the production throughput. All the subsequent stages are laid out to allow continuous production of a batch without hold up. The current limiting factors are the regulations for Pu (criticality)

and the sinter oven capacity.

The process parameters and control values for intermediate - end and by - products were collected and set down to act as the process specification of the fuel production with allowance for modifications due to the influence of plutonium.

The end products of these development trials indicated a depletion in carbon the reason for which was found to lie partly in drying with air instead of inert gas (nitrogen)[4] and secondly in the method of dispersing the carbon in the mixed feed. Experience showed that the qualitative dispersion of carbon for the quantities of mixed feed over 1 kg was very unreliable using the method used to date whereby the dry carbon is added directly to the Hexa-solution. In the meantime a paste method has been tried which allows the addition of highly concentrated but wetted carbon to the solution.

The amount of uranium carbide produced in these development trials is about equivalent to that required for the FFTF campaign ((UPu)C).

Internal Reports (confidential)

TM-42-80- 7
TM-42-80-14
TM-42-80-15
TM-42-80-23

Persons contributing

W. Hausmann
Ch. Jungo
V. Birchmeier
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J. Hörler
M. Gehringer
E. Keller
D. Peter
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G. Bart
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Mrs. M. Krois
Miss E. Dickenmann

2.3. Process development

The internal gelation requiring only a simple feed preparation (also with several metals) needs for the gelation itself a heat transfer medium, which must be removed before washing out the reaction products. At the moment EIR uses silicone oil with a special viscosity but this has the difficulty of needing trichlorethane to remove it. For producing oxides Oak Ridge National Laboratory has simplified the flow sheet so that a Trichloretheylene is already used as the heat transfer medium which can be removed by simple filtration before the excess nitrates and the waste reaction products from hexamethylentetramine are washed out. It appears to us however that this is insufficient to produce a workable particle of $UPuO_2$ + C at a gel temperature of 60-80°C.

The development work for 1980 concentrated on the simplification of the gelation. That is the heating up of the spheres and the transfer of the particles to the vibro-bed (wash) column to provide a controlled and continuous washing stage. The first tests have shown that the silicone oil can be replaced and the mechanical filtration is not required[/5//6/]. In this way the flow sheet for the internal gelation is identical to that of the external gelation in which the particles are brought directly into the ammonia solution, are washed and then dried. In this way the advantages of a simple feed preparation are combined with a simplified gelation-washing stage. The production of very large spheres (2 mm dia, dried) is also possible.

At present a small line for the production of ∿ 100 g

uranium material/hour is being constructed to test out the components and to assess the requirements for a larger throughput.

The transfer of the washed microspheres from the vibro-bed column to the drying stage is by filter vessels moved by hand. Only a free flowing pre-dried product can be loaded into the rotating drum dryer. In testing the MATTER belt-filter and dryer difficulties arose with the fine fractions connected with feeding the wet spheres and removing the dried spheres from the belt. The equipment has been modified but at the same time a drum filter is being examined.

Internal reports (confidential)	Persons contributing
TM-42-80-19	Ch. Jungo
TM-42-80-26	G. Ledergerber.

2.4 Laser light scattering

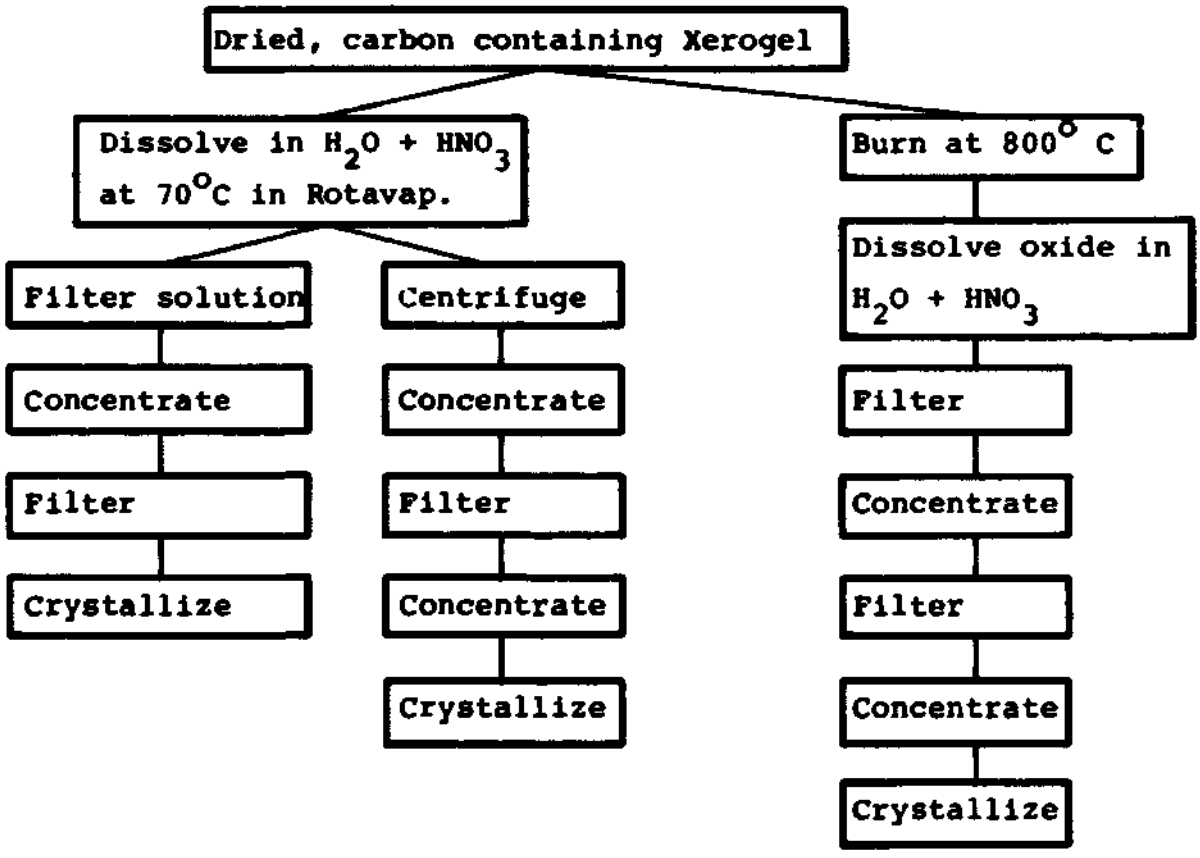
The work on laser-light scattering on suspensions could not be continued due to pressure on time and personnel. The preliminary tests with simple apparatus showed that a large effort would be required to produce worthwhile results. Nevertheless the theoretical basis for such experiments was established so that practical tests could be restarted at any time.

Persons contributing

Ch. Jungo.

2.5. Recycling of carbon containing Xerogels

During the commissioning of the new gelation equipment several kilograms of dried, carbon containing uranium-xerogels were produced which were not required for further processing (scrap). Three methods were tested for extracting the uranium and converting directly to the nitrate as shown in the diagram.



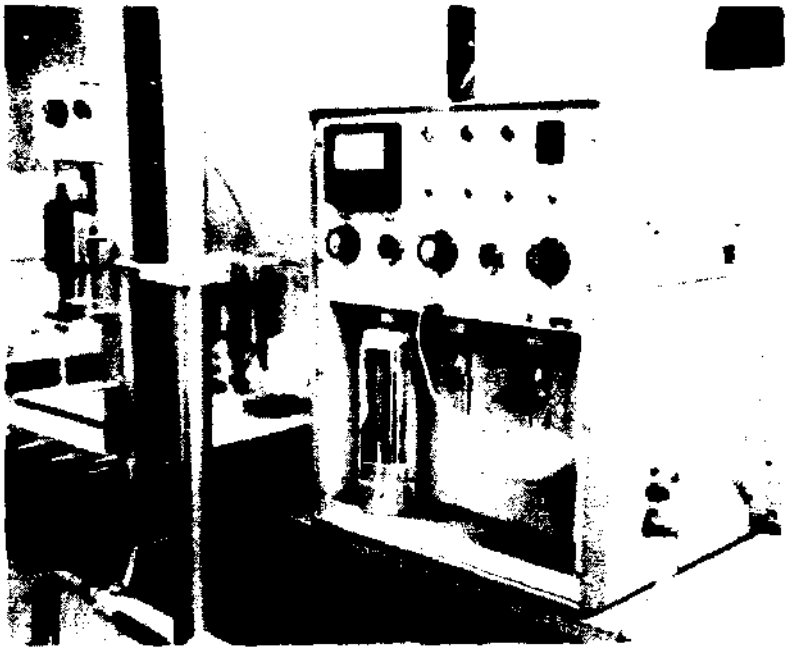


Fig. 2.1 a) Apparatus for determining the BET surface area

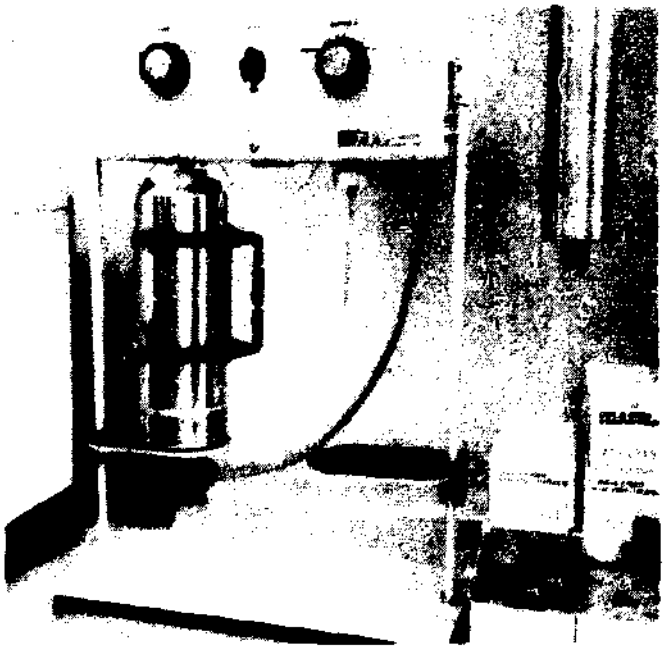


Fig. 2.1 b) Glove box unit for the BET apparatus.

The results of the laboratory trials were as follows

Method I : Unsuitable since the carbon-containing solution is difficult to filter.

Method II : Good, since the solution can be readily centrifuged.

Method III: A good method but unsuitable because of the effort needed since only a small muffle-oven was available requiring a large number of small runs.

The recycling therefore followed method II. In total 8.4 kg of scrap was re-worked from which 7.4 kg of re-crystallized Uranylnitrate-hexahydrate was obtained.

Internal Report	Persons contributing
TM-42-80-29	Miss M. Schärli W. Hausmann

2.6. Pore-structure analysis

The pore-structure analysis begun in 1979 on uranium-xerogels measuring the specific surface area via BET was continued.

The new mercury pressure porosimeter (CARLO ERBA Model 200) provides pore-volume and pore size distribution so that the following characteristics (at present on inactive samples) can be determined:

- Specific surface area - BET in the range 0.1 to 1000 m^2/g (fig. 2.1)
- Pore volume (total and partial) by Hg penetration (fig. 2.2)

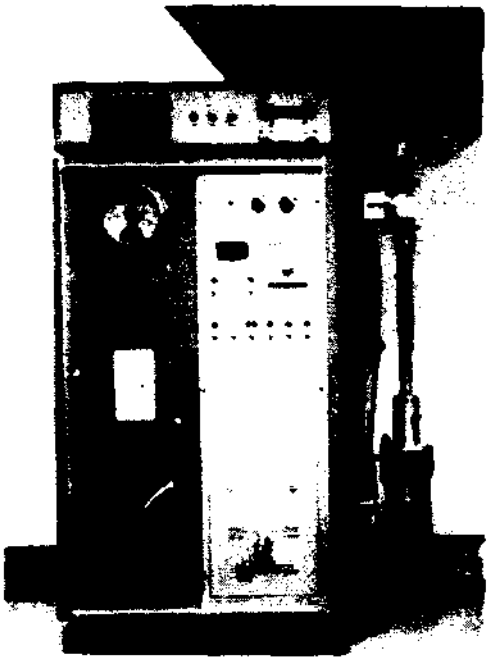


Fig. 2.2 a) Mercury-pressure porosimeter for determining the pore volume and pore size distribution.

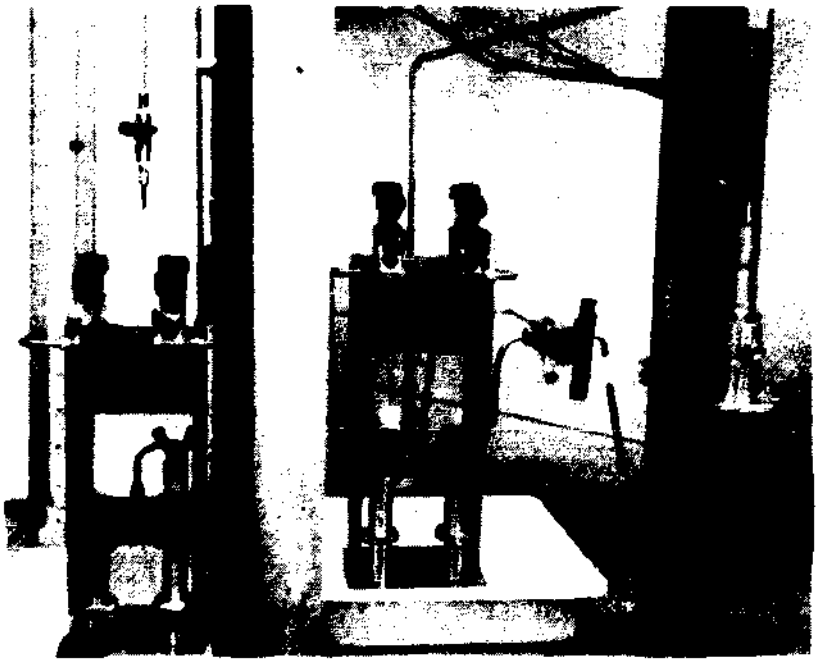


Fig. 2.2 b) Filling station and glove box unit for the Mercury pressure porosimeter with the glove box still open.

- Pore size distribution in the range 37 to 75'000 Å radius
- Estimation of the pore-shape (incl. bottles) from the decompression behaviour (re-emergence of the mercury).

In support of this the Analytical Group are able to determine the true density (BECKMAN Air pyrometer) and the apparent density (Hg-pyknometer). Together with the scanning electron microscope the Hot Lab therefore possesses a complete capability for characterizing porous materials.

From a large series of tests on dried Uranium-xerogels the influence of the production parameters on the microstructure will be examined.

Persons contributing

W. Hausmann
J. Hörler

References to ch 2

/1/ Planung der Mengenverhältnisse zur Herstellung des Brennstoffs for das Bestrahlungsexperiment AC-3 im FFTF.
M. Hausmann vertraulich:TM-42-80-7

/2/ Druckregulierung zur Kontrolle des Durchsatzes und der Grössenverteilung beim Kreuzstrahl-Tropfengenerator
Ch.Jungo, G.Ledergerber, D.Peter vertr. TM-42-80-15

/3/ Die neue Gelierbox in Labor 214.
G. Ledergerber vertr. TM-42-80-23

/4/ Der Drehrohrofen zum Trocknen von Uran-Plutonium Xerogel-mikrokugeln
M. Gehringer, W. Hausmann vertr. TM-42-80-14

/5/ TM-42-80-19 Ch. Jungo confidential

/6/ TM-42-80-26 Ch. Jungo confidential

3. FUEL ANALYSIS

During the year the work of the analytical group (principally analysis on non irradiated fuel) and the radio-chemistry (PIE) groups of the Hot Labor were combined. The work covered also overseeing of the post irradiation examination of the DIDO-III experiment (with the Hot Cell group).

3.1. Thermogravimetric Analysis

The thermogravimetric balance was installed earlier to provide (among others) a better understanding of the reactions occurring during the thermal treatment stages of the mixed carbide microsphere production. Work in 1979 was concerned with modifying the installation to accept α-active materials. This was completed in 1980. Fig. 3.1 shows a view of the modified equipment in the α-box.

To reduce the maintainance requirements the electrical control panel and the rotary pump were mounted outside the box. The changeover from the heavy high-temperature oven to the super-high temperature oven is now made pneumatically. A cooler was installed to remove the heat from the ovens, this too is positioned pneumatically above the oven. A special balance-repair box with associated transfer system was built.

The thermal balance was used inactively during the difficult period when these modifications were under way. The main activity for the project concerned the study of the calcination stage of the fuel fabrication. The indivi-

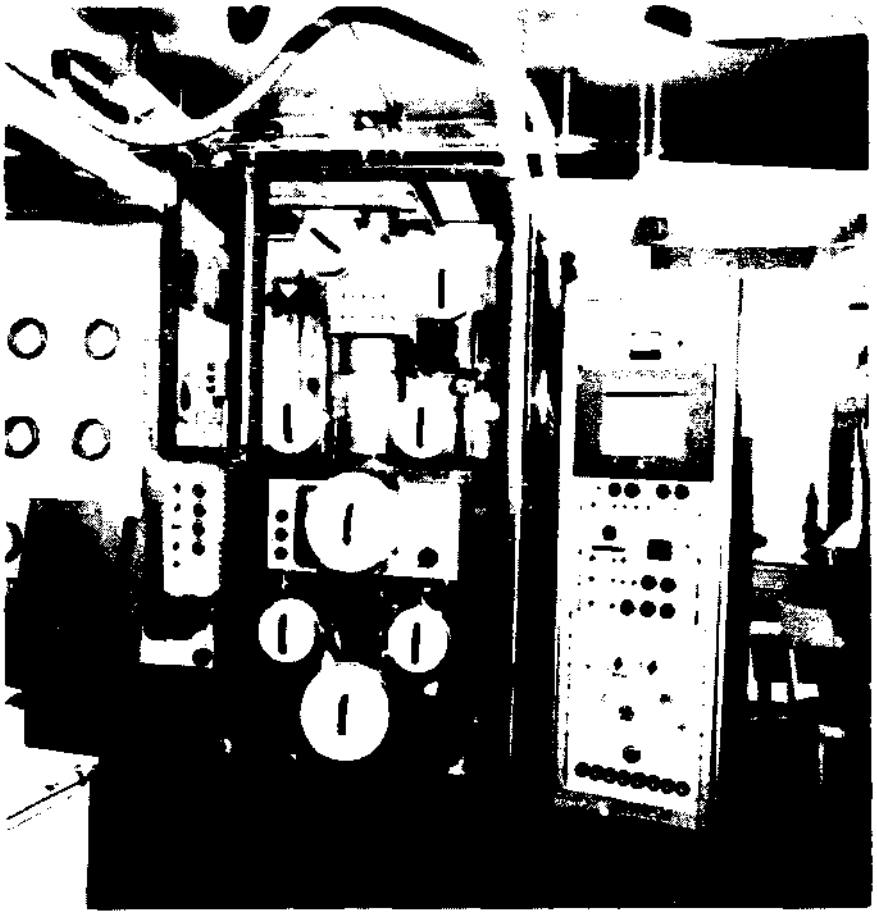


Fig.3.1 Front view of the thermo-gravimetric balance, rebuilt to accept α-and weak βγ-active samples.

dual weight changes during the process were better characterized thanks to the use of the Quadropole retained gas analyser. An extended study of the fuel sinter programme was started.

Thermogravimetric oxidation trials were carried out on UC samples investigating among other things the behaviour and result under O_2 and CO_2. The aim was to produce an easily soluble UO_2 or U_3O_8 for waste handling, storage or recycling. In the test with O_2 three stages were observed. The main second stage suggests a reaction

$$UC_{1.43} \xrightarrow{+O_2} UO_2 \text{ (or } U_3O_8) + CO_2 \text{ (or CO)}$$

the final phase being the oxidation of the remaining UO_2 to U_3O_8. In a first phase however lasting from 100°C the 350° level a reaction forming the oxycarbide

$$UC_{1.43} + 0.2O_2 = UC_xO_{0.4} + (1.43-x)C$$

appeared.

For the trials under CO_2 it was found that at lower temperatures up to 750°C a mixture of UO_2+C is obtained depending on the temperature and reaction time. Above 850°C the reaction goes more quickly to completion producing UO_2 alone. Addition of oxygen causes the U_3O_8 to be formed. A detailed study of the intermediate products and processes was possible. The use of these results and the examination of the solubility of the product can be found in ref /1/.

During optimization of the UC microsphere production, the question was raised whether additives used during fabrication could be contributing to the carbon source in the sintering stage (carbothermic reduction). A series of tests with the TG balance followed the weight loss by

evacuation and heating to 1450° C.

Internal Reports	Persons contributing
TM-42-80-6	F. Petrik
TM-42-80-28	G. Bart
AN-42-80-2	S. Huwyler
	K. Bischoff
	G. Ledergerber

External Report

IAEA-SM-246/12

3.2. Work with the X-ray fluoroscopy apparatus (XRF)

Earlier the XRF had been installed for making analysis of the chemical content of samples for the HL, in particular the analysis of U and Pu and later for trace-elements in materials. A special packing machine was developed for handling α-active materials. At the beginning of 1980 a Diffractometer attachment was installed to carry out a quantitative analysis on the sequicarbide and dicarbide content of the mixed carbide fuel, and first tests carried out.

Samples were obtained consisting of reasonably pure UC and UC_2 from the metallurgy department and U_2C_3 from the fuel fabrication. The carbon contents ranged from 4.79 (UC) to 9.18 (UC_2).

Seven standards were produced with the following (corrected) compositions

1.	UC	91.4%	4.	UC	78.3%
	U_2C_3	4.7%		U_2C_3	17.7%
	UC_2	3.9%		UC_2	4. %

2.	UC	87.2%
	U_2C_3	8.9%
	UC_2	3.9%
3.	UC	83. %
	U_2C_3	13.1%
	UC_2	3.9%
5.	UC	91. %
	U_2C_3	0.5%
	UC_2	8.5%
6.	UC	86.1%
	U_2C_3	0.3%
	UC_2	13.6%
7.	UC	73.9%
	U_2C_3	17.4%
	UC_2	8.7%

It was found that it was not possible to produce a UC_2 free sample since the "pure UC and U_2C_3" material were slightly over-stoichiometric.

The results of the measurement with the XRF are within a few percent (absolute) of the experimentally determined values using a method developed by Ganguly and Vollath (ref 2) which means that after further verification in 1981 the procedure can be used for a semi quantitative check on the fuel production.

Internal Reports	Persons contributing
TM-42-80-25	S. Huwyler
	J.L. Delplancke

3.3. Gammaspectroscopy

The programme library of the Gamma-measuring center TN-4000 was extended. The equipment can measure now simultaneously axial scans on fuel pins and radial surface scans across on fuel sections. A plotting

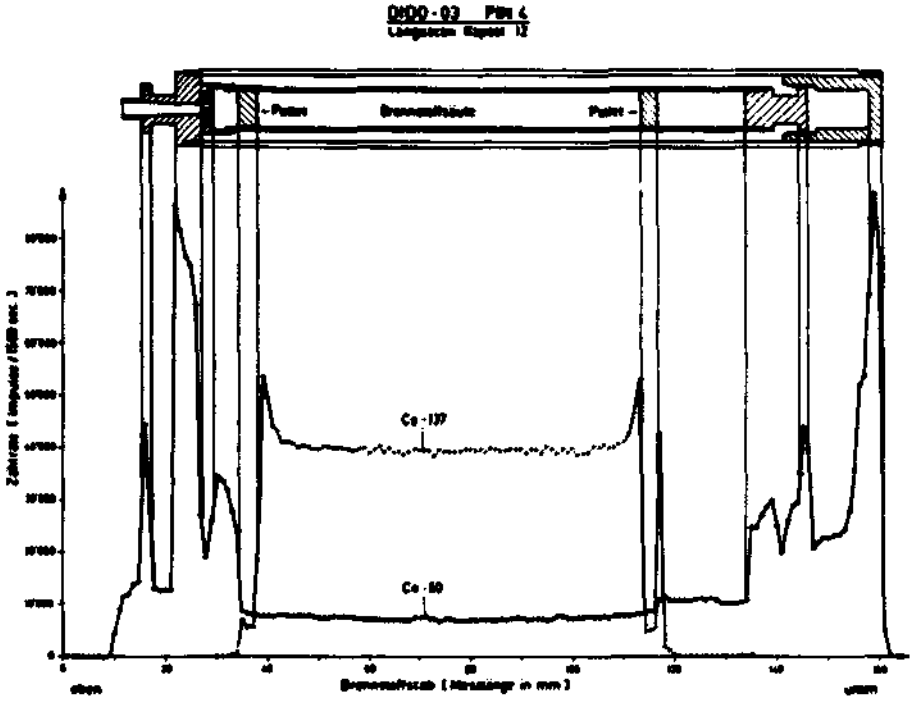


Fig. 3.2 Gamma scan axial profile of DIDO pin showing the build-up of Caesium 137 at the outer face of the end-pellet.

programme was written to permit several nuclides from an axial scan to be plotted together on the same diagram.

The DIDO-III pins under examination in the Hot Cells following their return from Harwell in 1979 underwent axial gamma-scanning. By measuring in very narrow steps (0.5 mm) a very detailed profile of the distribution of fission products and activated elements along the pin could be obtained. This not only revealed the underlying structure of the pin but showed that the volatile fission product Cs-137 had moved and collected partly on the outside face of the upper (plenum side) end pellet (Fig. 3.2). This is a significant finding concerning the movement of caesium in carbide fuels.

Persons contributing

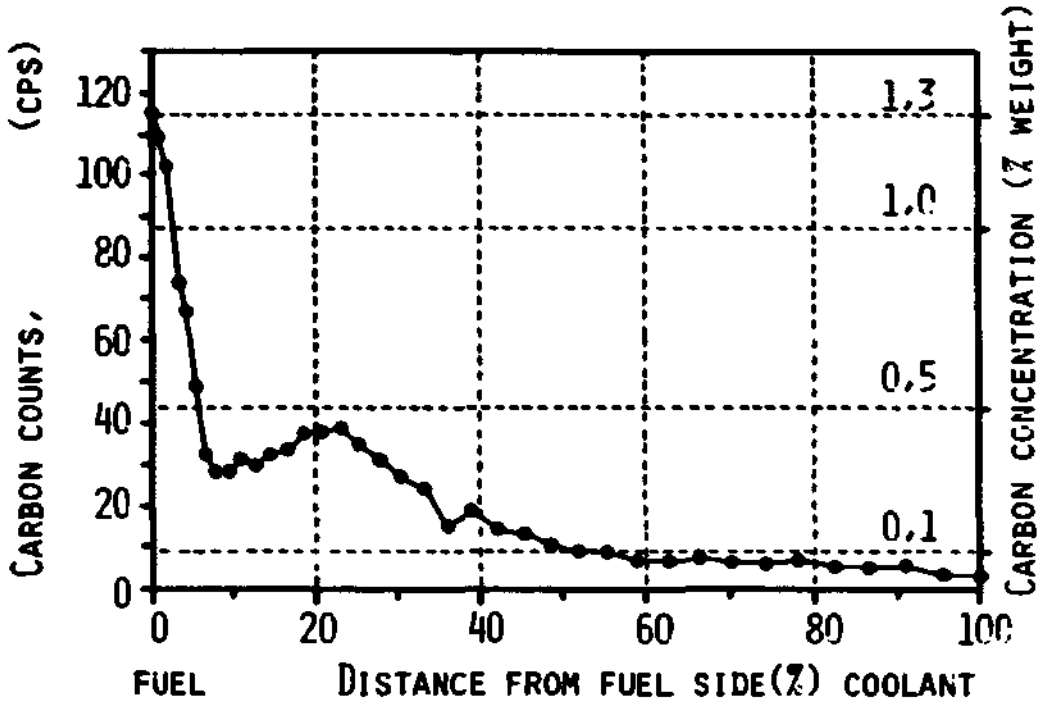
G. Bart
T. Aerne

3.4 SIMS Development

The secondary ion mass spectrometer was purchased and installed a few years ago when the machine itself was a rarity in research labs. Seeing the additional advantage for examination of radioactive materials, efforts were immediately begun on the complex task of building the spectrometer into a shielded facility. Shielding was also required inside the instrument. In 1980 the spectrometer was first brought into uee for active materials.

The first examination was to produce carbon profiles for cladding material from the FILOS 07 experiment[3].

PROFILE 1



SAMPLE PREPARATION

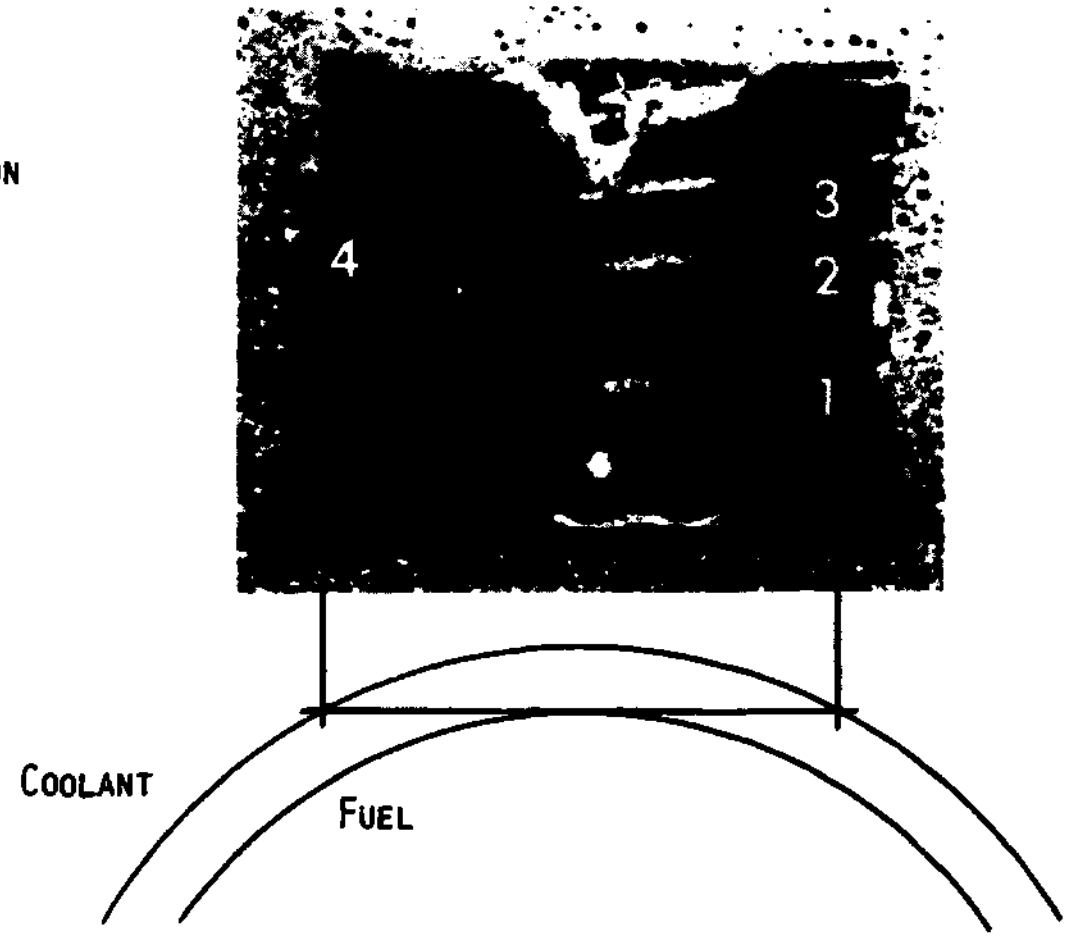


Fig. 3.3 Clad carbon profile No.1 of Filos 07 sample P-2. The cross section shows the eroded pathways of the SIMS beam 1-4. The diagram shows the method of cutting the cladding sample.

As a sample a specially prepared angled cross section of clad was used. Fig. 3.3 shows the carbon profile No.1 of the sample FILOS 07. P-2. The Hot Lab SIMS analysis is qualitatively in good agreement with the profile obtained from the Harwell analysis (op cit ref 3).Fig. 3.3 also shows the angled sample where the traces of the SIMS beam erosion can be seen as well as the irregular impression of the large fuel spheres in the clad wall.

The problem of the sample observation and positioning the beam was elegantly solved by installing an alignment telescope. The rebuilding of the equipment for beta-gamma active operations is described in[4]. In order to reduce the background in the Channeltron zero-count-rate in the gamma field of active samples a shield was first modelled in brass and installed in the receiving chamber between the sample and the Quadropole. This required strengthening of the Quadropole support structure. Tests with the model shielding were positive and a contract for manufacture of the special shield in tungsten was placed at the end of the year.

Because of the requirement for highly active samples uncontaminated with carbon a system for embedding the material in Bi/Sn alloy was built following French plans. Inactive tests have already been carried out. The first carbon free active clad samples were prepared by this method in 1981.

External Report	Persons contributing
EIR-390	G. Bart
	T. Aerne
	U. Flückiger
	E. Sprunger.

3.5 Fuel Analysis

A knowledge of progress in and control over the fuel fabrication process requires detailed analyses of feed, intermediate and end product materials. The heavy effort on process development for the FFTF campaign described in chapter 2 caused a constant demand for routine quality control measurements. New equipment and revised procedures were also introduced.

The oven of the Leco-oxygen analyser was rebuilt for installation in a glove box. From the second quarter the carbide O_2 analyses were carried out with this new apparatus. Due to significant discrepancies between the determination of the oxygen content in UC from the new equipment compared to the older Leybold-Heraus, attempts are being made to obtain well characterized UC samples from outside sources.

The introduction of a titroprocessor for poteniometric uranium and plutonium concentration determination in the feed solutions took place. Comparison between these measurements on uranium concentrations and analyses made in the Chemistry division (R.Keil) were in excellent agreement. (Table 3.1).

The time-taking mercury density determination was replaced with the much simpler density measurement using an Air pyknometer. The new method gives the density of the fine fraction material which the mercury could not wet. (Table 3.2)

Persons contributing

Mrs. I. Adeler
Mrs. J. Mitar
Mrs. M. Krois
Miss E. Dickenmann
G. Bart

Table 3.1 Uranium determination

(sample U-007-030)

	Potentiograph	Titroprocessor
	U. mg/g	U. mg/g
	351.82	350.52
	S = ±0.03	S = ±0.06
	V = ±0.05	V = ±0.05
Metallurgy Dept (Keil)	Photom. 353.61	Titrat. 350.61

Table 3.2 Comparison of Mercury Immersion and Air pyknometer measurements of fuel density (UC)

Sample No	Hg-min. g/cm^3	Air-Pyk. g/cm^3
K-176	12.91	13.03
K-177	12.85	13.06
K-178	12.81	12.95
(Granite	2.67	2.695)

References to ch 3.

1) S. Huwyler, K. Bischoff
"Recycling of (UPu)C scrap by incineration in a controlled atmosphere".
Int. Symposium on the management of Alpha contaminated wastes.
IAEA-SM-246/12 Vienna 2-6 June 1980.

2) C. Ganguly, D. Vollath,
Quantitative phase analysis in the U-Pu-C system by X-ray diffraction.
KfK 2049 (Sept 1974).

3) L. Smith, G. Bart, B. Bürgisser, R. Hofer, E. Keller, J. Mitar, D. Orciuolo, P. Putrik, J. Reindl.
"Pilos 07, The post Irradiation Examination of a corrosion test pin clad in M 316 material and fuelled with (UPu)C microspheres".
EIR-Report No. 403.

4) G. Bart, T. Aerne, U. Flückiger, E. Sprunger.
"Modification of a sencondary ion mass spectrometer to allow the examination of highly radioactive specimens".
EIR Report 390.

4. IRRADIATION AND DEVELOPMENT

During the period under review the activities of the irradiation group have been focussed mainly on the developments for the proposed FPTF/AC-3 experiment. No irradiation testing was carried out by EIR during the year.

A new fuel pin fabrication line has now been installed in the Hot Laboratory and successful precommissioning took place with inactive (dummy) material on the vibro-filling box. Ancillary equipment such as a gas system for controlling the atmosphere of the weld chamber was also designed and tested. Weld trials with both US and EIR designed end-caps were carried out and a weld test programme is now being established to meet the required standards. The whole box line is based on the use of a custom-built pin-to-box coupling and transfer system designed to prevent the pick-up and spread of contamination during fabrication.

The Mol-11/K5 experiment was put back due to the delayed restart of the BR-2 reactor. Irradiation began early in 1981.
Post Irradiation examination of the four DIDO-III pins was under-way in the Hot Cells.
Results of the DFR/527-1 experiment performance were reported (see ch 5).

The studies in support of a conceptual study for an improved (UPu)C fuel line were heavily cut back during the year. Preliminary engineering drawings for the thermal treatment stages were prepared supported

by computer calculation of the thermal losses and temperature distributions. The results are currently being evaluated.

4.1 Preparations for an experiment in the US Fast Flux Test Facility (FFTF)

Since the beginning of 1979 EIR has been working on concrete proposals to test some of its fuel in the FFTF. This is the culmination of many years development as described in this series of annual reports.

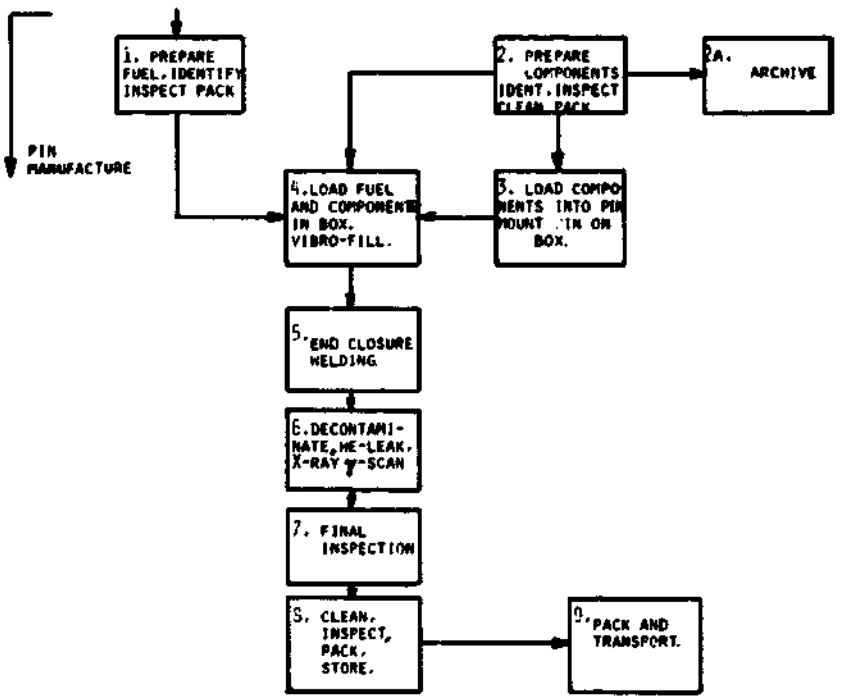
As stated earlier, the task would be to deliver up to 20 (UPu)C sphere pac pins to the USA during 1982 for irradiation up to 8% fima, later to 12% fima with a comparison with the US pellet fuel.

The fabrication of these 20 pins represents for EIR a further step from laboratory scale production of a single short pin to a larger, almost pilot scale fabrication of full sized rods. This drastic programme change was not without consequences for the earlier Pu line. It was decided in 1979 to rebuild the line to accept pins of 2.4 m and the first developments were reported last year[1].

Although having a long experience regarding the vibro-filling of rods extensive testing of the new concept was required before a final decision could be taken regarding the pin loading and vibro-filling techniques. The concept of a new docking and sealing port for the pin to the filling, welding and decontamination box was described last year. This concept was built, tested and refined during 1980 and finally confirmed in its operations.

4.1.1 The Fuel Pin Fabrication line

The development of the line was governed as much by the available space and positioning problems as by technical requirements due for example to the stringent Quality Assurance demands. All of the laboratory space for the pin fabrication and controls has now been designated and most of the equipment is now in place. The concept for material flow, especially the transfer stages has been influenced by the location of labs and buildings. The following steps are forseen.



After an extensive series of trials with a box mock-up the actual vibro-filling box was mounted in place (Fig. 4.1) where commissioning trials are virtually complete,aiming for trouble free filling, maintaining

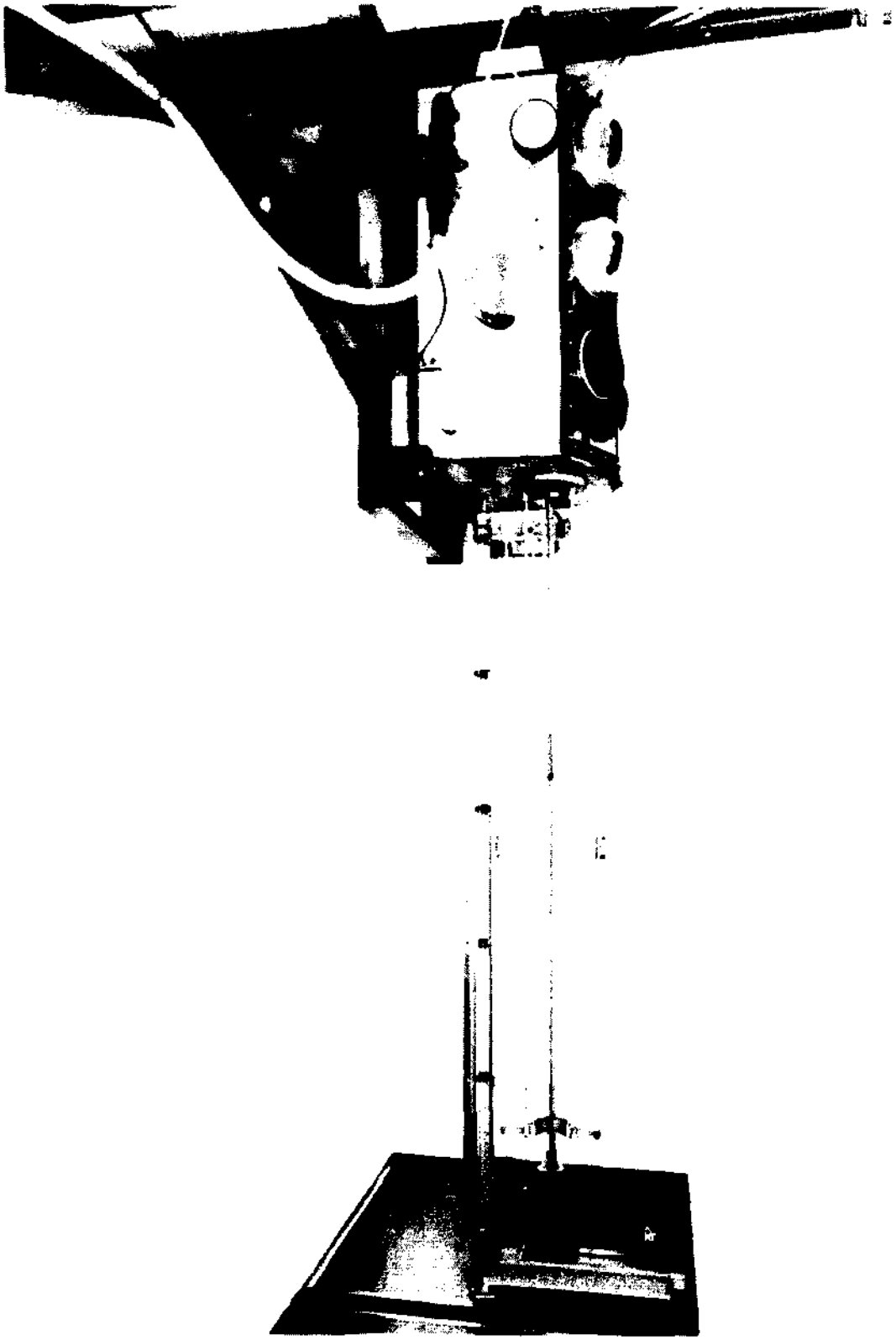


Fig. 4.1 View of the vibro-filling box mounted in place with vibrator, protection tube and pin to box docking device.

a sealed box at all times and avoiding the spread of contamination from box to box. (An example of the packing quality can be seen in fig. 4.5)

It was decided to keep and use the original welding box replacing the pin docking and sealing port and fitting the new welding chamber whose design was reported last year.

The welding chamber and its new gas system (see below) plus newly fitted controls and recorders was duplicated for the inactive workshop where welding trials have been under way.

The final box in the chain - the decontamination box has also been built and is mounted in position. Its fitting out is delayed until the exact and permitted decontamination procedures can be determined in trials for which a test programme has been prepared.

Other EIR groups have been approached to advise on component cleaning prior to use and attempts have been made to establish the maximum surface contamination permitted for the pins.

For the pin inspection,equipment has been provided or is in manufacture to cover:

- Radiography - a jig to support the pins for the full length and weld region radiography together with the establishment of procedures to comply with US standards.
- Helium leak test container and provision of a He leak detector

Reports	Persons contributing	
TM-45-80-52	M. Nicolet	and
TM-45-80-47	F. Botta	Metallurgy Group.
	H. Leute	Technical Lab.
	L. Wiezel	Health Physics.
	E. Klarer	
	H. Lehner	
	J. Sidler	
	P. Wyss	

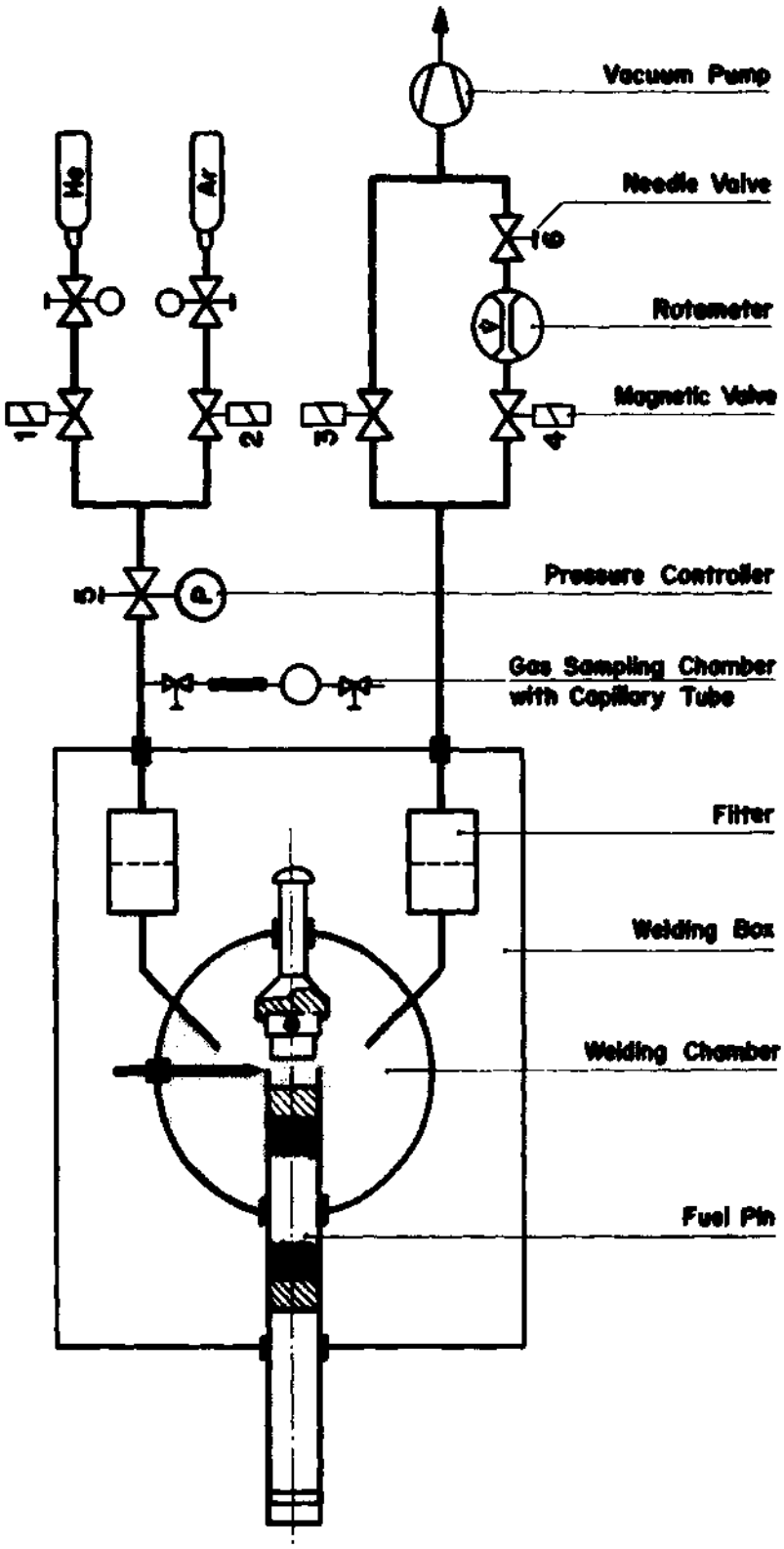


Fig. 4.2 Diagram of gas filling and evacuation system

4.1.2 Gas system for end-cap welding

As described last year[1] the weld chamber in the nitrogen filled welding box functions also as a gas filling and evacuation station for the pin. Before welding the pin, which comes from the nitrogen (N_2) filled vibro-filling box, must be filled with helium (He) and is then welded under argon (Ar). The N_2-He exchange in the pin is achieved by repeated evacuation and He filling. The change-over from He to argon in the weld chamber can however only occur when the end-cap is firmly in place and immediately before welding. To minimize the risk of Ar contamination of the pin helium the change-over must occur quickly and at as near constant pressure as possible.

A gas system meeting these requirements with semi-automatic controls was developed, built and mounted first on the inactive welding test-bed during 1980. Fig. 4.2 shows the simplified diagram of the system and an overview of the complete set-up is shown in fig. 4.3.

Evacuation and filling the pin occurs by alternately opening and closing valves 3 (V3) and 1 (V1) either manually or automatically. The automatic operation can be set by choosing the time step and number of cycles. Following this V1 and V4 are set for helium purging until - just before welding begins - V1 is closed and V2 simultaneously opened to purge the weld chamber with argon.

A gas sampling bottle is mounted in parallel with the weld chamber and is connected via a capillary tube. The capillary is choosen to represent the gas flow resistance of the fuel column and the sample volume is equivalent

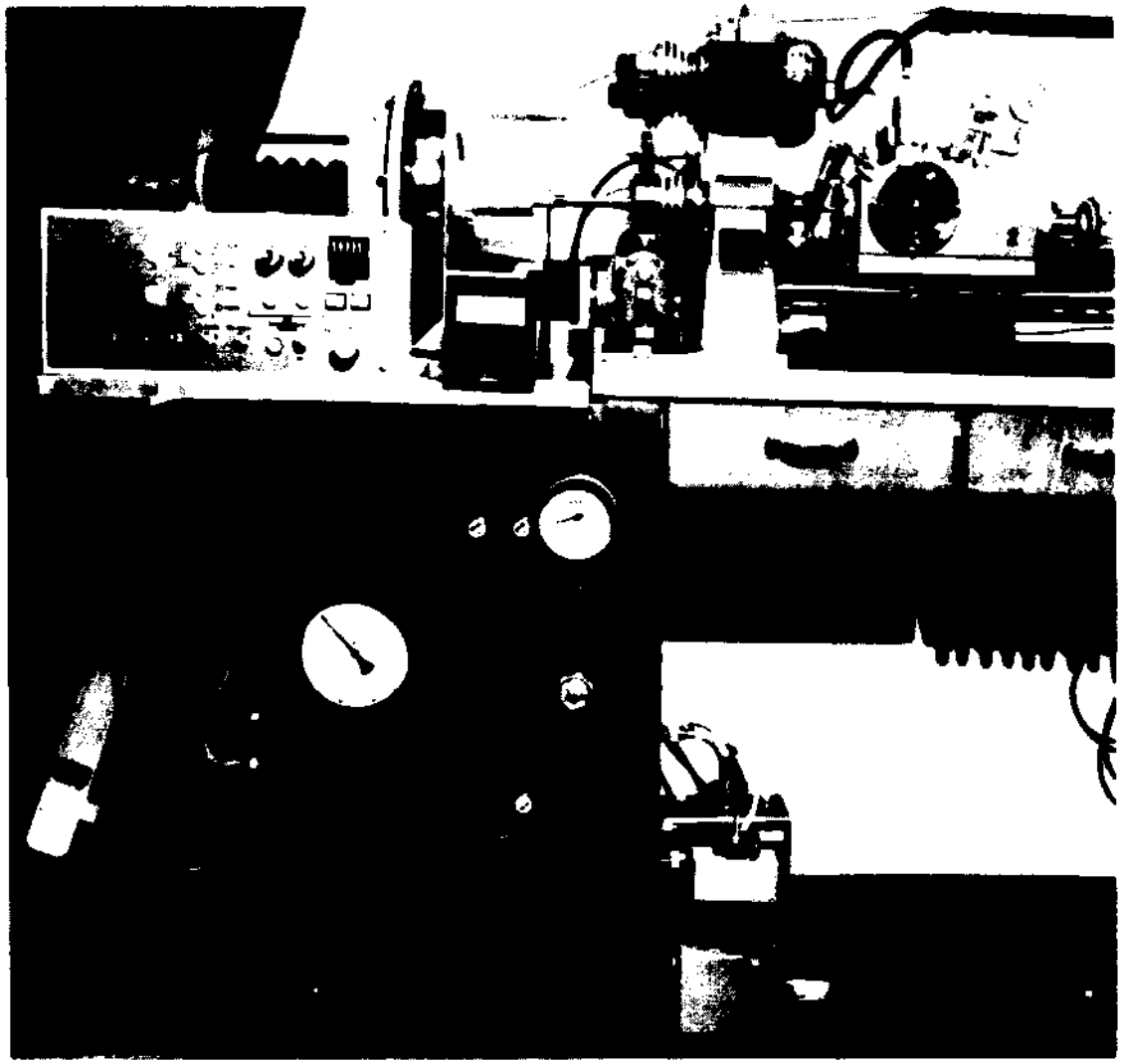


Fig. 4.3 Gas system and control panel attached to welding chamber for inactive weld trials.

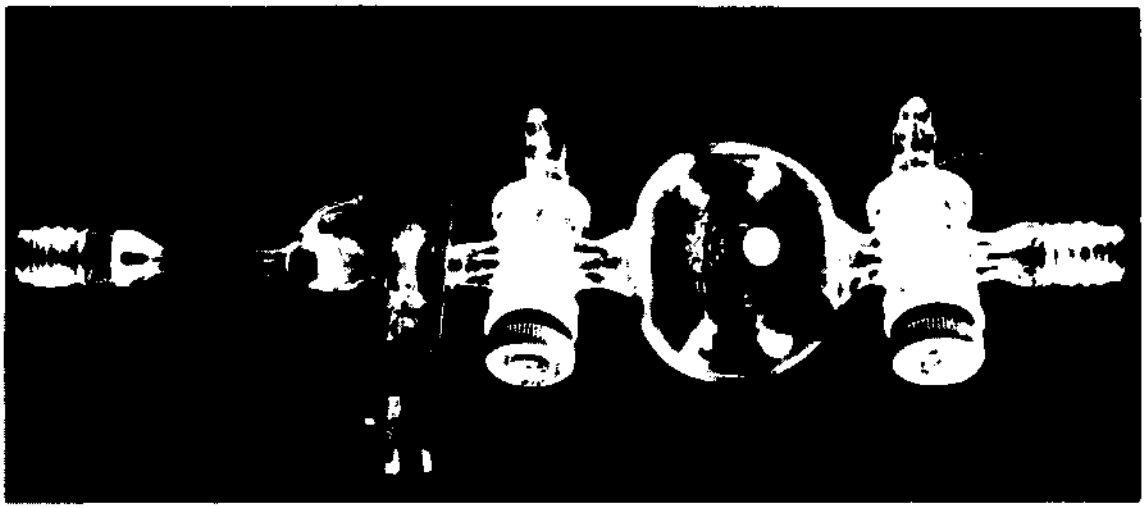


Fig. 4.4 Gas sampling chamber with capillary tube.

to that of the pin plenum. The content of the sample bottle which is filled with N_2 at the start of the evacuation fill routine is therefore a reference for the gas to be found in the pin after welding and can be analysed as a check on the filling routine.
Fig. 4.4 shows the sample chamber and the capillary which is a 1 mm dia x 30 mm tube filled with 70 µm microspheres.

Persons contributing

P. Botta
L. Wiezel
J. Sidler
H. Lehner.

4.1.3 Pin axial density distribution by gamma scanning

Following the rebuild of the γ-scan equipment in 1979, a data handling routine was written in 1980 with the help of the EIR computer center. Taking into account the actual materials and pin internal geometries and the specific γ-absorbtion values the code provides a graphic print out of the smeared density axial profile over the fuel column and adjacent components (fig 4.5).

Reports

TM-15-81-3
TM-45-81-3

Persons contributing

F. Botta
J. Patry
M. Nicolet
H. Leute
P. Wyss

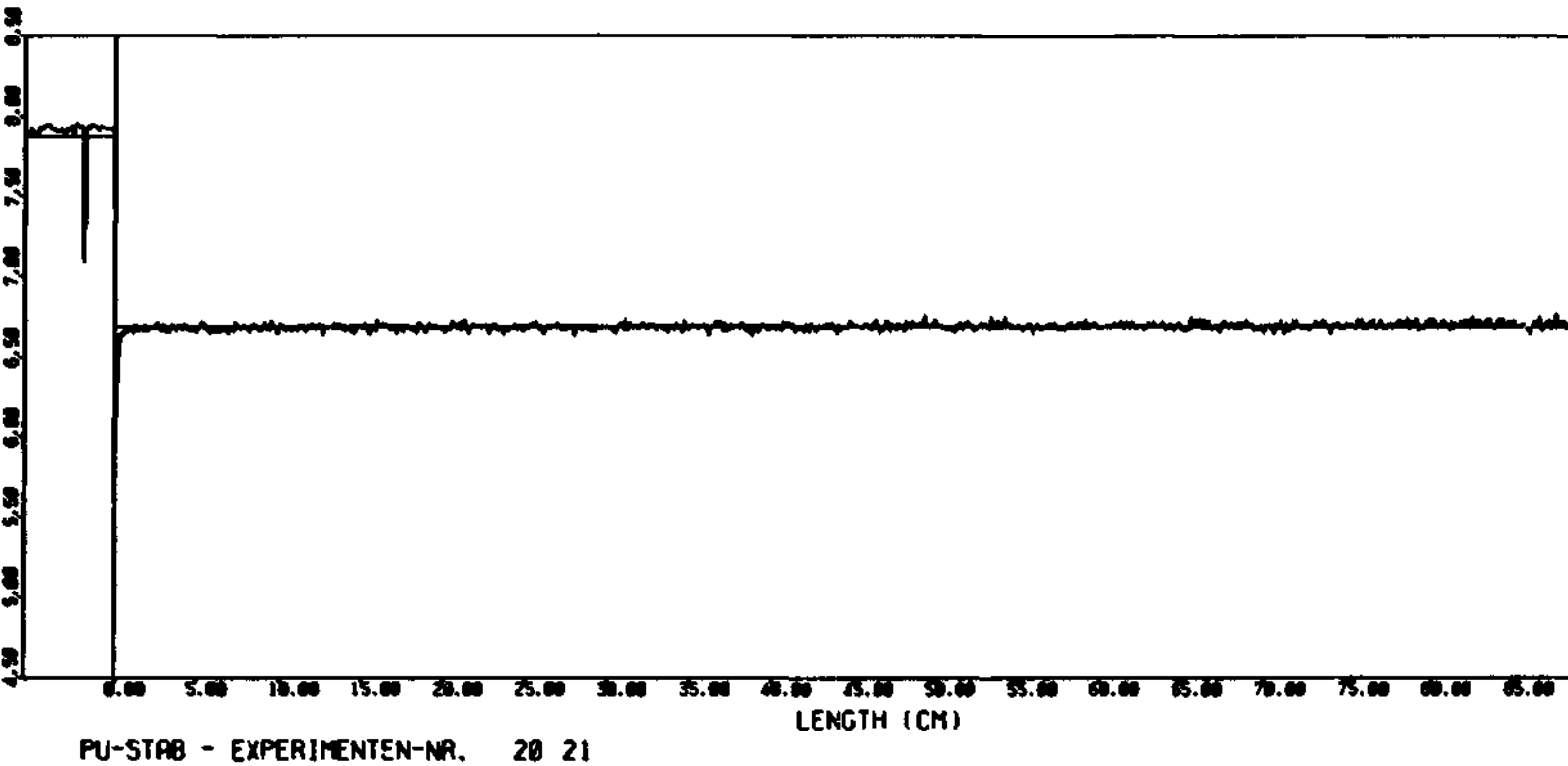


Fig. 4.5 Computer plot of axial density distribution measured by γ-scanning, showing the dummy fuel and end cap pellets of a dummy AC-3 pin.

4.1.4 Design Layout of the AC-3 sphere-pac pin

The AC-3 experiment has the requirement that the EIR pin design should be as nearly possible identical to the pellet filled pins. The following special needs of the sphere pac pin have to be taken into account however:

- *Retention of the fuel particles in the fuel zone, at the same time permitting free gas movement in the pin.*
- *Support for the fuel column so that it is fixed during filling, handling and transport and yet free to expand towards the gas plenum during irradiation.*

These aspects plus some further restrictions arising from the available filling equipment, materials and EIR filling practices have led to a pin design as shown in fig. 4.6 where the sphere pac pin is compared to a pellet design. The special components were developed manufactured and tested during 1980 as described below.

Fuel seal disc

The sealing disc has to meet several requirements. It must retain particles > 40 µm dia in place at the same time providing a permeable barrier for gas flow. The fuel must be held during vibro-filling and transport and at least until the fuel starts to sinter in reactor. A high permeability is required at room temperature for evacuation - helium filling and sufficient during operation to allow pressure equalization. It has to be compatible with the fuel, easily loaded into the pin and made of a material available and suitable for small batch production.

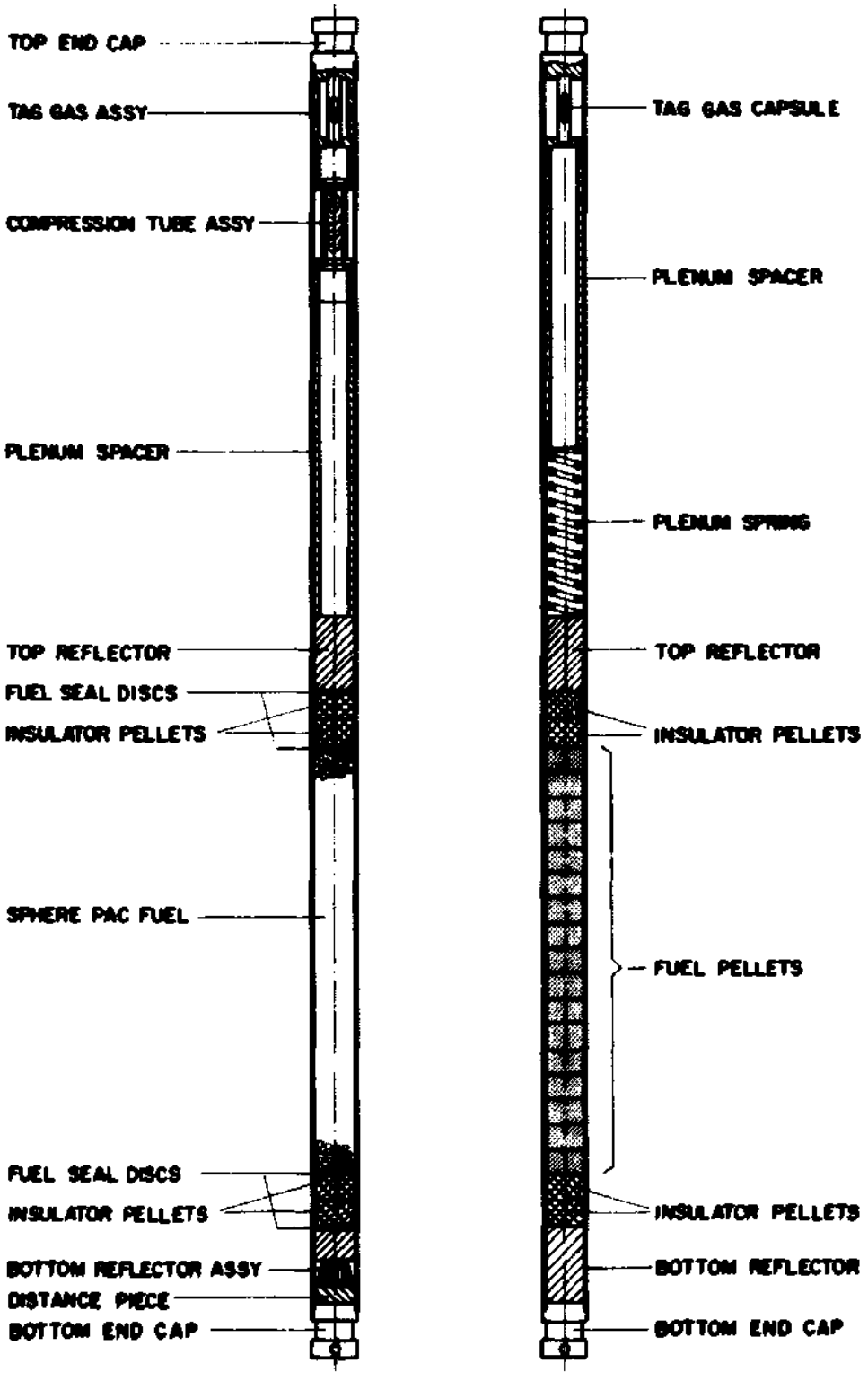


Fig. 4.6 Comparison of typical sphere pac and pellet test pin designs.

The solution was a molybdenum disc ∿ 0.1 mm thick. Taking into account possible tube ovalities and eccentric positioning a mean radial gap of 10-12 µm could be allowed. This can only be achieved by matching the discs to the tubes in question. Special attention was given to a means of determining non-destructively the dimensions of the disc-clad combination. A simple but very sensitive method was found by calculating the mean radial gap from measurements of the laminar gas-flow resistance of the disc in the tube.

Compression tube assembly

This component replaces the helical spring used in the pellet pin. The spring restores a pellet fuel column to its original position if shaken out of place. In operation the spring compensates for differential axial expansions in the pin components and limits the axial loads developed.

In the case of a sphere-pac pin any expansion of the column is virtually irreversible (except possibly by re-vibrating) and therefore has to be prevented. A compressive component was developed which is almost rigid in the elastic range but which deforms plastically in a controlled way once a given load has been exceeded. For the specified load and collapse length stress and buckling analyses were done. For the promising geometries component tests were done to check the reproduceability of collapse and the response to manufacturing stresses caused by vibro-filling and tag gas capsule puncturing. By the end of the year the design was confirmed with only a few final tests outstanding.

This side blank

Other components

Two other pin internals were modified firstly to by-pass the loads on the gas tag capsule by addition of a surrounding support tube, secondly to hold the fuel column in place when the pin is being moved to the weld box and horizontally welded by adapting the end of the lower reflector to become an expanding collet which can be locked before transfer. A distance piece matched to the actual pin component lengths is loaded into the pin just prior to welding.

Persons contributing

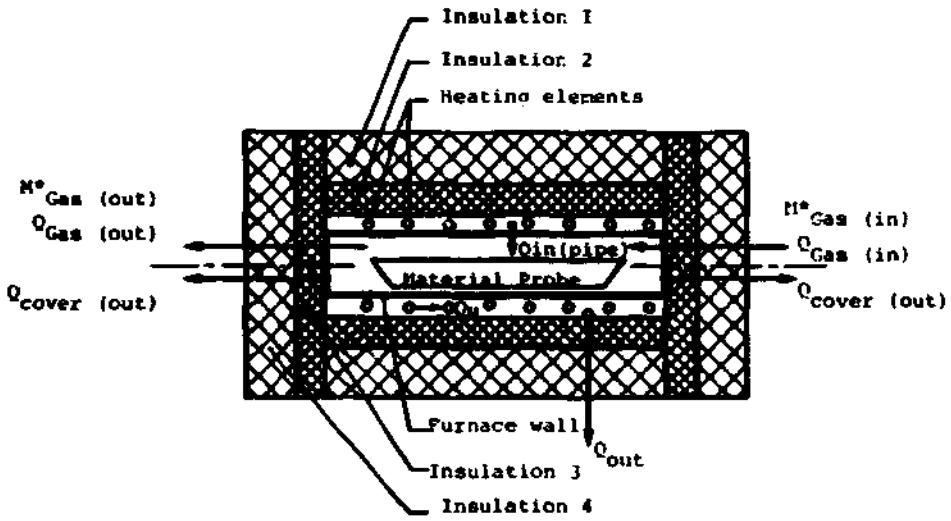
F. Botta
L. Wiezel
E. Klarer
H. Lehner.

4.2 Pilot Plant Engineering and Development

In support of the pilot plant conceptual design study for an improved (U,Pu)C fuel fabrication line, design work was done by the Matter Co. for EIR.

In 1979 a study was made comparing the feasibility of continuous production compared to batch production. The low throughput and difficulty of designing a continuous sintering furnace caused the batch concept to be choosen.

Fig. 4.7 General model used for simulating the heat transfer and temperature distribution of a furnace



The general heat balance is:

$$\Delta Q_{In} = \Delta Q_{In\ (pipe)} - 2\,\Delta Q_{cover\ (out)} - \Delta Q_{Gas\ (in-out)}$$

with $\Delta Q_{In\ (pipe)} = Q^*_{Heater} \cdot \Delta t - Q^*_{out} \cdot \Delta t$

where: ΔQ_{In} = Net radiant heat flux inside the furnace

$\Delta Q_{In\ (pipe)}$ = heat flux through the furnace wall

$\Delta Q_{cover\ (out)}$ = heat loss through the end cover door

ΔQ_{Gas} = heat loss due to the innert gas.

The computer programme includes a table of the thermal conductivity data of different insulation materials as well as of some innert or reducing gases. It requires as input:

- Total heating time and heating power
- number of insulating layers and type of insulation
- furnace dimensions
- mass and physical properties of the material probe.

In order to be able to design the furnaces properly a more detailed study of the dynamic behaviour of such furnace during the heating up and cooling down period was required. The objective was to evaluate the commercially available refractory lining (type and thickness) required for preventing heat losses during the high temperature processes. In the temperature range at or above 1800°C the engineering problems are mainly related to the mechanical and chemical behaviour of both fuel and engineering materials used (refractory lining, crucible material, etc.). The heat transfer problems and the temperature distribution through the furnace and the refractory lining were simulated with a computer programm especially suited for a dynamic response. Based on the results of this study an optimization of the furnace design was carried out, for which basic engineering drawings were prepared. Finally a proposal regarding manpower and development costs was compiled.

4.2.1 Simulation of the thermal behaviour of a batch type furnace with indirect heating

The design and optimization of high temperature furnaces suitable for the production of nuclear fuel containing plutonium as well as other actinides is a difficult task since besides the criticality problems which restrict furnace capacity there are mechanical design and material problems to be solved.

In view of the rather high temperature required and of the long sintering time involved (ca. 20 hours), the heating and cooling period must be kept to a minimum.

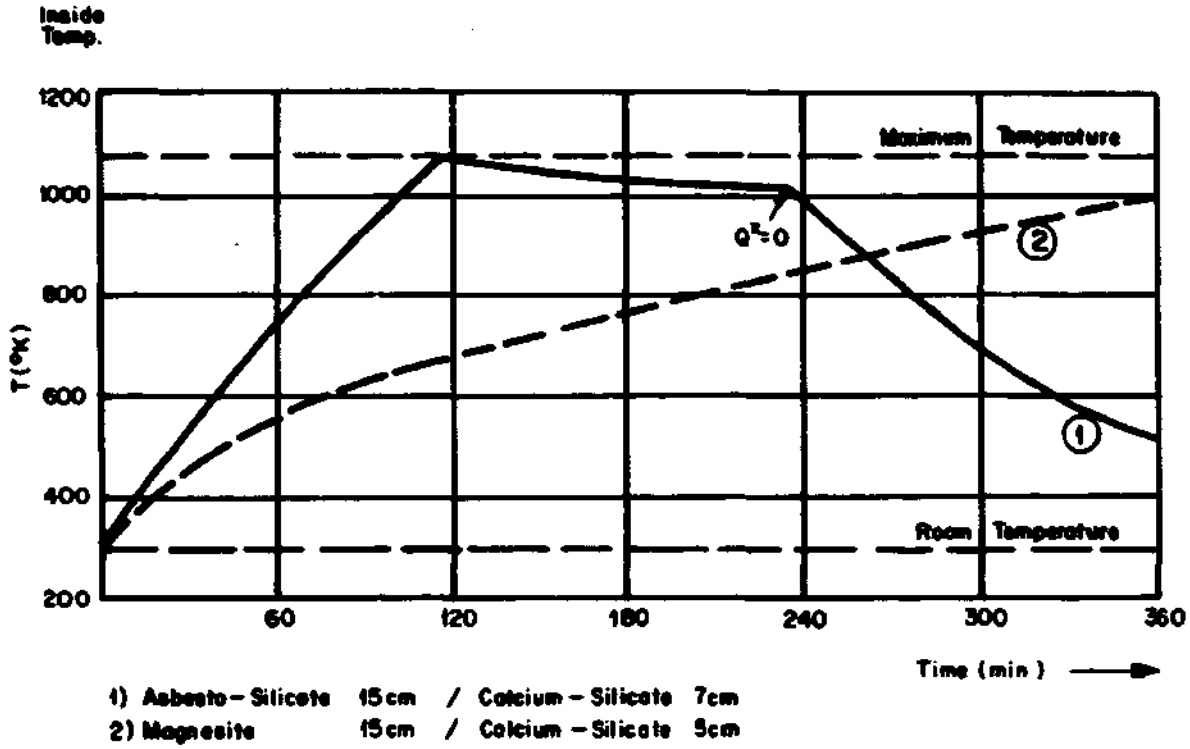


Fig. 4.8 Change of temperature with time inside the calciner furnace pipe (heating power 5 kw)

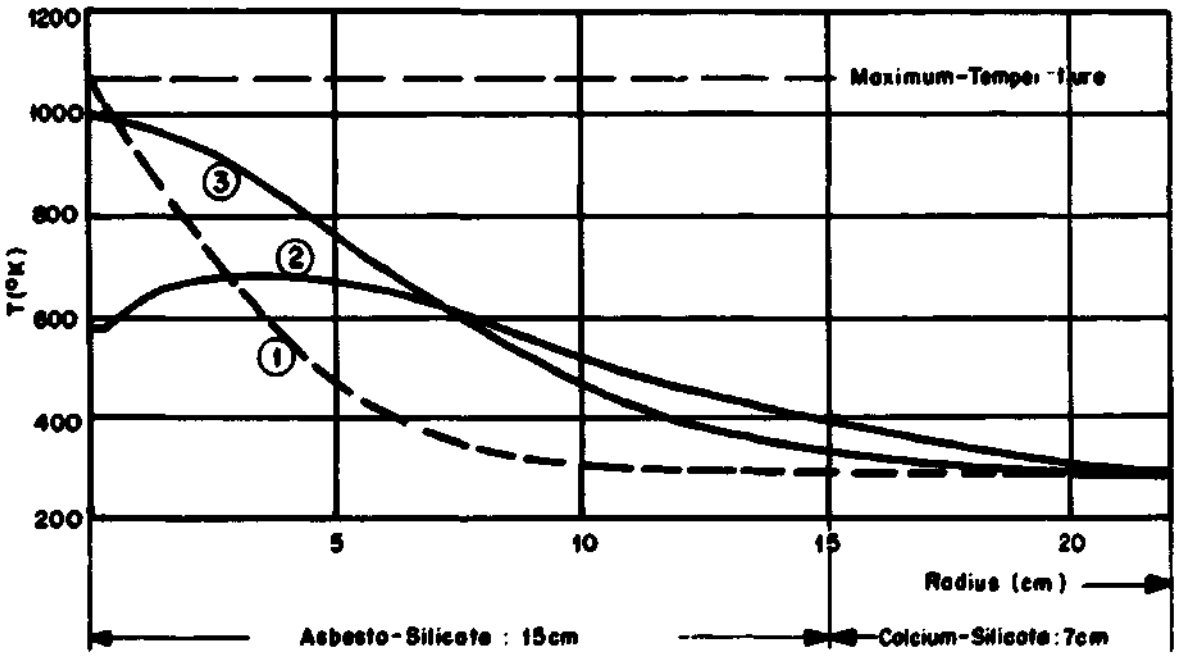


Fig. 4.9 Temperature profile through the insulation in function of the radial distance.

1) Time 120 min.: at end of heating period
2) Time 330 min.: cooling period with internal gas cooling
3) Time 240 min.: five minutes after heater off.

Heat losses to the environment must also be considered seriously otherwise the glove box where the furnace is supposed to be installed will heat up gradually with time and exceed the allowable limits (60-70^{o}C).

A general furnace model for dynamic computer simulation has been developed which allows the estimation of heat transfer rates through the furnace walls and insulation layer and calculates also the temperature distribution in function of the heating or cooling time. The reference furnace is of a batch type with indirect heating and consists of a cylindrical pipe which holds the crucible with the material sample. The electrical resistance is located between the furnace wall and the different insulation layers (two or more). Both end cover doors may also be insulated. The simulation model allows for cooling of the lateral wall and cover doors with water or inert gas. In sore cases it may be necessary to flush the material sample undergoing thermal treatment with an inert or reducing gas. Provision was made also to simulate this forced convection effect. The model and related programme is very flexible and allows a general calculation and layout of any type of furnace with indirect heating.

Reports

Ingenieur Bureau F. Matter AG: Simulation eines Rohrofens mit beliebiger Kombination von Isolations- und Kühlschichten.

Persons contributing

Matter Co. (Buchs AG)
M. Nicolet
W. Hausmann.

References to ch 4.

/1/ Pu-Programme Jahresbericht EIR 395

5 FUEL PERFORMANCE

The Vorhaben "Fuel Behaviour" set up in 1980 has the task of developing an understanding of LWR fuel behaviour as well as that of Fast Reactor fuels. It is part of the project "LWR core behaviour". That part of the work concerned with the modelling of FBR carbide fuels is reported here. Only a small amount of work could be accomplished in 1980 at EIR, and then only with difficulty due to an understaffed Vorhaben. Thanks to a strong effort at Oregon State University (OSU) USA (financed by EIR) the development of the SPECKLE-1 code for sphere-pac carbide fuel behaviour was able to continue.

5.1 Modelling sphere-pac carbide fuel behaviour

The code SPECKLE-1[1] is being developed at Oregon State University, Corvallis, USA financed by EIR and with EIR's technical support. The code is in an early stage of development. A package of sub-routines covering the thermal behaviour of the fuel is in place, gas release and restructuring has been modelled and sub-routines for radial heat source distribution are being incorporated. Local fuel swelling is as yet only crudely modelled and effort is just beginning on the modelling of the fuel-clad mechanical interaction. This last is perhaps the most important part of the code - all other sub-routines lead

finally to the mechanical load on the clad which together with the chemical attack and the changes in materials properties determines if and when the clad may fail.

During the year, after discussion with EIR, OSU began a series of lab experiments aimed at investigating the special characteristics of sphere pac loading on a cylindrical clad. A large scale model was used consisting of a perspex tube filled with steel spheres representing up to 3 size fractions to scale. Fig. 5.1 shows the partly filled tube and the hydraulic loading device to produce a radial load on the clad. The tube is partly silvered to reflect light. One of the aims is to attempt to use photo-elastic stress analysis to examine the stresses on and around the contact points. Strain gauges have also been placed on the tube for additional information.

SPECKLE-1 was used in its preliminary form in 1979/80 to calculate possible center fuel temperatures in EIR's sphere-pac carbide fuel pin in the Fast Flux Test Facility under start-up conditions[/2/]. This work was also reported in the previous Annual Report.

Later in the year, use was made of the observed structural changes in the DFR experiment to use as temperature markers with which to compare SPECKLE-1 calculations. Use of the Blank et al[/3/] designation of structural zones in advanced fuels provided estimates of the temperature at the zone boundaries. Comparison of these temperatures with the radial temperature profile calculated from SPECKLE-1 gave good agreement for cross sections having a well developed restructuring. In a section at lower temperature and power the agreement with SPECKLE was less good. Similar agreement was found when comparing

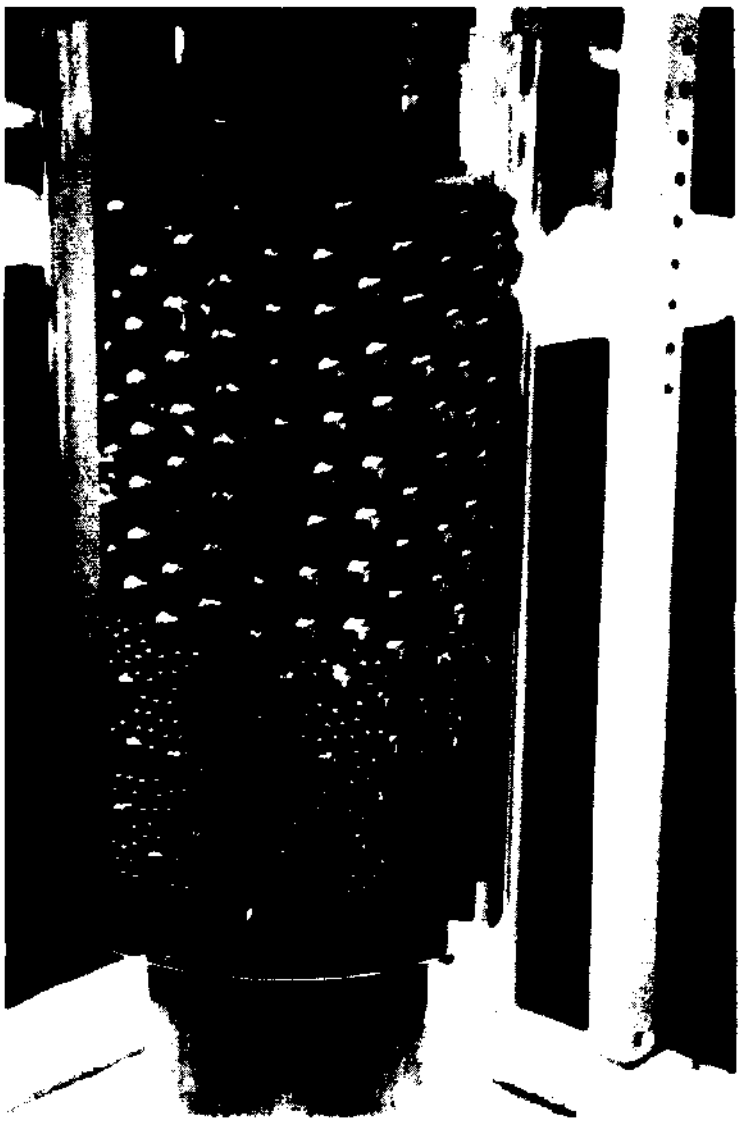


Fig. 5.1 Rig for simulating radial clad loading of sphere-pac fuels. Note the reflective part of the plexiglass clad for photo-elastic stress analysis. (Oregon State University).

SPECKLE-1 prediction of the radius of the central sintered zone[4]. Fig. 5.2 shows the comparison for the sections M and S.

It is important to re-state that SPECKLE-1 is not complete. It still contains oversimplifications in attempting to model the complex events in a sphere pac pin under irradiation. Cross checking with the experimental observations as described above gives a useful measure of progress but good agreement cannot be taken to mean that all mechanisms are correctly modelled. Use of the code has shown several areas where the format and subroutines need improving. Together with EIR OSU will undertake a revision to improve the code efficiency and flexibility as well as making its operation more easily understandable[5].

At the end of the year, use of the code at EIR (comparing with earlier experimental results) had thrown up a number of anomalies which OSU began tracking down. More experiments (lab or in-reactor) will be needed to improve the data base.

Also during the year, OSU completed a study of the effect of the early gas release from the sphere pac fuel on thermal conductivity[6]. Experimental evidence tends to show however that no significant release occurs until a burn up of ∿ 3% fima has been reached (according to results of separate experiments).

Reports		Contributions
EIR Report 396	EIR	R.W. Stratton
EIR Report 415		M. Nicolet
OSU-EIR-49		J. Reindl
OSU-EIR-47	OSU	K.L. Peddicord
OSU-EIR-48		S. Montgomery
		C. Robinson
		C. Bennet
		T. George
		R. Henke
		M. Hoy
		A. Vincent.

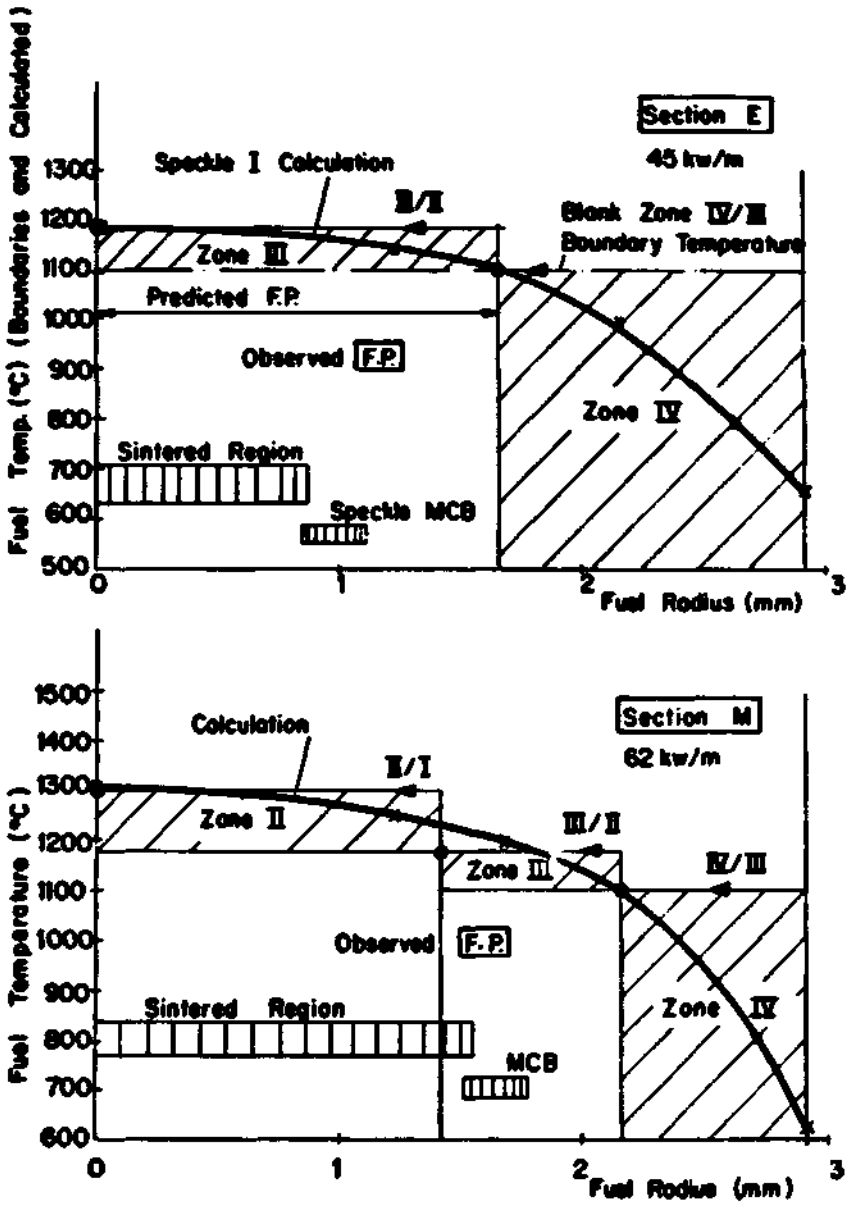


Fig. 5.2 Diagram of observed structural zones, temperature boundaries and calculated radial temperature profiles for a (UPu)C sphere-pac pin irradiated to 7% fima in DFR[4].

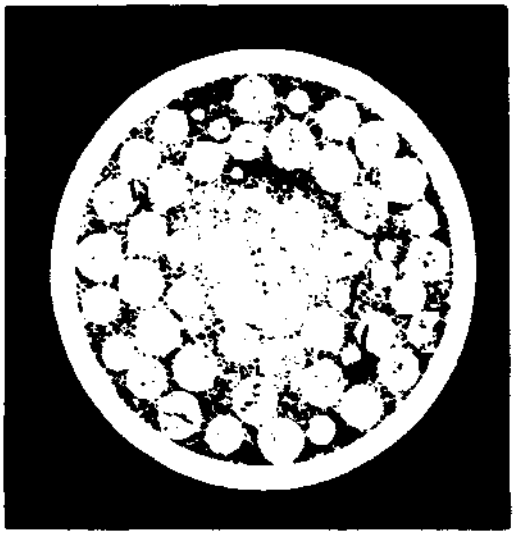
5.2 Irradiation performance of sphere-pac fuel

Reports on comparative irradiation tests of pellet and sphere-pac carbides[7] and the results of the Filos 07 experiment[8], both of which were described in the previous annual report, were published in 1980. Also published at the end of the year was the report on the performance of the EIR sphere-pac fuel pin irradiated in the Dounreay Fast Reactor (DFR) Scotland[9]. This, together with the continued examinations of the DIDO-III experiment was the only post irradiation work done in 1980.

5.2.1 DFR Experiment 527/1

As has been reported earlier the fast flux test of EIR sphere pac fuel took place in the DFR between 1975 and 1977 (4 cycles; 270 full power days) and reached a burn-up ∿ 7.5% fima at a peak linear power of ∿ 70 kw/m. The pin was clad in 20% CW stainless steel 316 L. With a coolant inlet temperature of 230°C this was basically a low tempereature experiment. Peak clad temperature reached ∿ 620°C (EOL).

The pin was intact and the examination gave results which generally followed what had been found in previous experiments. Due to the modest power and temperature restructuring had not proceeded sufficiently for a porous central region to form although with the exception of the coldest fuel section a solid fuel matrix had formed in most of the pin center along the pin length (Pig 5.3). Due to the hyperstoichiometric fuel used (30-40% M_2C_3 in the fine fraction) extensive carburization of the clad occured above 450°C although this did not lead to pin

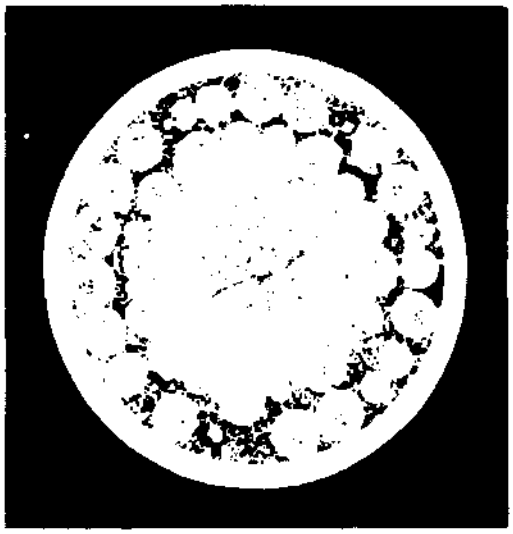


Section S

49 kw/m

5.7% fima

320°C.

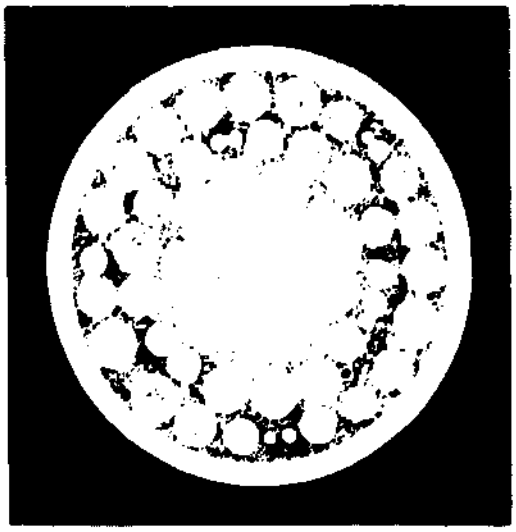


Section M

62 kw/m

7.3% fima

458°C.



Section E

45 kw/m

5.3% fima

610°C.

Fig. 5.3 Ceramographic cross sections of DFR Pin 527/1 showing linear power (EOL) Burn-up and mean clad temperature.

failure (max 0.75 ^{w}/o carbon). Even more marked however was the carbon uptake in the hottest clad region from the coolant side. This effect has been seen in other (oxide) pins irradiated in DFR. The carbon uptake (estimated 37 mg) may be sufficient to explain the slight weight gain of the pin measured after irradiation (+ 44 mg).

Although the sphere-pac pin showed, as in an earlier experiment[7] less overall clad diametral strain than a corresponding pellet pin and with a smoother profile, the axial diameter profile did exhibit an unexpected twin maxima at the ends of the pin ($\sim$ 1% $\frac{\Delta D}{D}$ at 350 and 610°C) compared to the pin center (0.5 - 0.7% $\frac{\Delta D}{D}$ at 400-580°C)(Fig 5.4). This pattern has been seen in certain other pins irradiated in DFR and the report discusses in detail some possible causes. Of the three mechanisms - axial variations in mechanical interaction and material properties, - double maxima in neutron induced void swelling rates for cw 316 steel, - chemical effects (caesium movement or carbon uptake) weight was given to the clad volume change due to clad carburization. Other workers have seen increased clad carburization on fuel and/or coolant side in the hottest clad section (> 600°C). The amount of carbon in the clad in the EIR pin would give a volume change (according to Bellamy 0.17 $\frac{\Delta V}{V}$ per 0.1% carbon increase) sufficient to account for the observed excess diametral strain. Too little is known about the mechanical behaviour of fuel and clad and the neutron dose received by the clad appears too low to account for the strains by neutron induced void swelling.

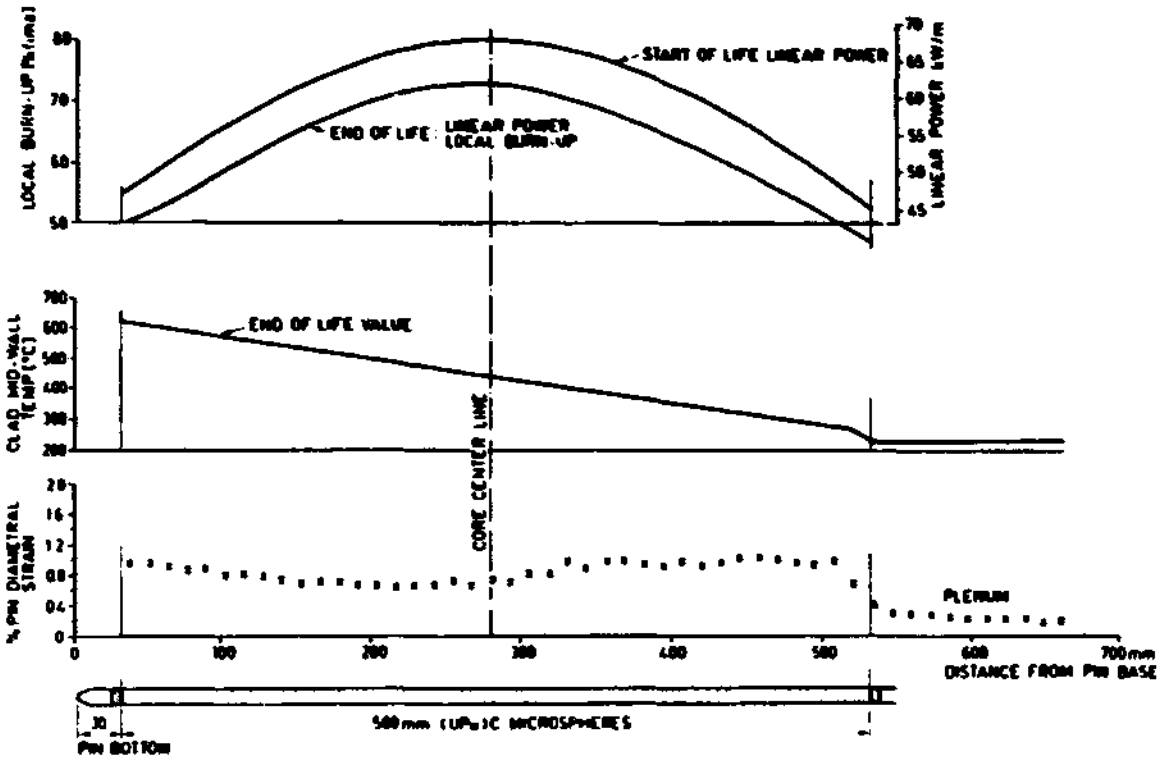


Fig. 5.4 DFR 527/1 Pin strain, Burn-up power and cladding temperature.

Reports	Contributions	
TM-42-80-1	K. Bischoff	J. Mitar
AN-42-80-20	L. Smith	G. Bart
	R. Stratton	M. Nicolet
TM-45-80-24	R. Hofer	J. Reindl
EIR Report 395	B. Bürgisser	K. Peddicord OSU
	A. Erne	S. Montgomery OSU
	F. Petrik	
	E. Keller	Fuel Group HL
	T. Aerne	Irradiation Group BS.

5.3 Supporting calculations and studies

In spite of a reduced manpower availability, studies and supporting calculations were carried out on the project's experiments. Use of the SPECKLE-1 code at EIR increased and showed up several anomalies in the code structure and mechanisms modelled. The work benefitted from a summer visit of OSU personnel.

A large number of SPECKLE calculations were carried out based on MFBS-7, DFR and Filos operating conditions. As well as providing data for the experiment evaluation (temperature profiles) these exercises gave an insight into the functioning of the SPECKLE code. A plotting programme was used to provide a visual output of SPECKLE results (Fig. 5.5).

Earlier versions of SPECKLE-1 (version SPJ) featured a immediate rise to full power on starting a calculation. In most cases this is unrealistic since (particularly for pellet fuel) the rate of initial rise to power can dominate subsequent pin behaviour and life. A version was then developed which incorporated a slower rise to power (version Ramp) enabling the PFTF start conditions to be modelled. Differences between "SPJ" and "Ramp"

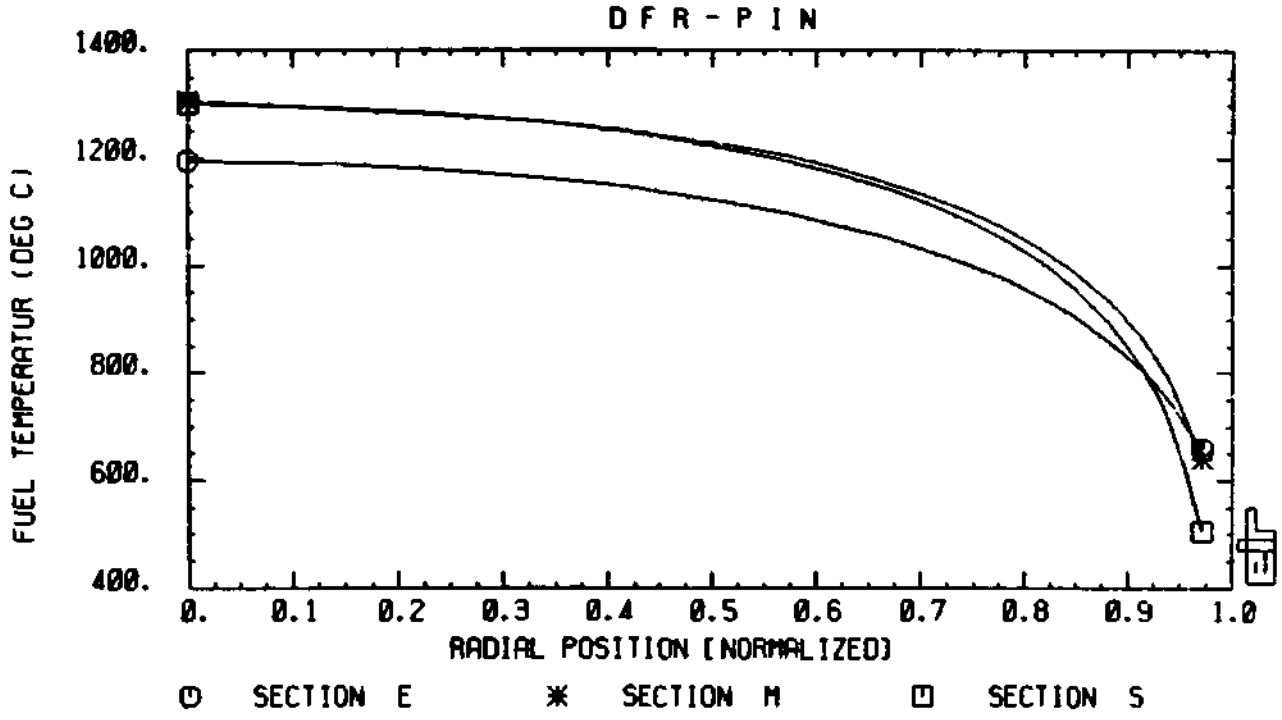


Fig. 5.5 Radial temperature distribution in sections E, M and S (DFR 527/1) as calculated by SPECKLE

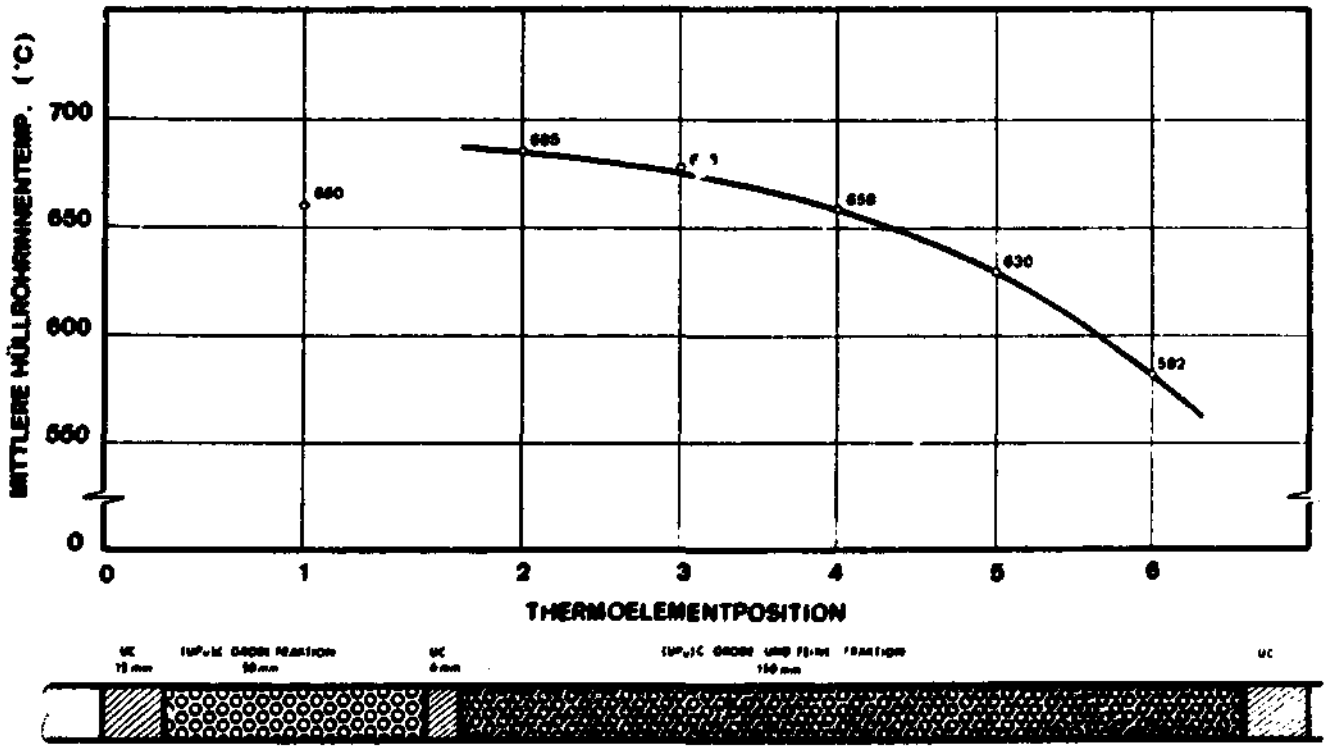


Fig. 5.6 Axial cladding temperature profile for the experiment FILOS 07 related to the sodium thermocuple positions.

were being investigated at the end of the year.

Calculations (not using SPECKLE) were made to fix the axial temperature profile in the cladding of the Filos 07 test[/8/]. Fig. 5.6 shows the results for the pin over the two fuel zones based on the measured sodium temperatures (six thermocuples). Studies also continued on ways of approaching the modelling of the sphere-pac mechanical interactions. At EIR the first approach was to use the existing models of pellet-clad interaction with a view to adaption. EIR looked at the SATURN code, OSU at GAPCON-THERMAL-3. It was generally found that existing codes and the use of finite element methods are less suitable as a starting base and the approach will be made to model sphere pac mechanical behaviour from first principles. EIR will continue to gain experience of existing pellet models for comparison purposes.

Report	Persons contributing
OSU-EIR-47	EIR J. Reindl
TM-45-80-24	M. Nicolet
	OSU K. Peddicord
	C. Robinson
	S. Montgomery
	R. Henke

References to chapter 5

/1/ Modelling of the Thermal Behaviour of Irradiated Sphere-pac Mixed Carbide Fuel. PhD Thesis,Dept of Nuclear Engineering, Oregon State University
M. Ades OSU-NE-7902 (1979)

/2/ Calculations of the Thermal Behaviour of Sphere-pac Carbide Fuel Pins under Start-up Conditions in the US Fast Flux Test Facility using the code SPECKLE-1,
K.L. Peddicord, S.D. Montgomery, R.W. Stratton
EIR No. 396 OSU-NE-8002
May 1980.

/3/ Swelling in Advanced Fuels up to High Heat Ratings.
H. Blank, M. Coquerette, Hj Matzke, C. Ronchi
Int Conf on FBR fuel performance,Monterey
March 5-8 1979.

/4/ Sphere-pac carbide fuel pin thermal calculations
compared with post irradiation examinations.
R. Stratton, M. Nicolet, J. Reindl, K. Peddicord.
Paper submitted to the ANS Summer meeting 1981.

/5/ Code modifications to SPECKLE-1
T.L. George, K.L. Peddicord OSU-EIR-49, Oct. 1980.

/6/ Early Gas Release in temperature calculations for
sphere-pac carbide fuel
C.W. Bennett, S.D. Montgomery, K.L. Peddicord
TANSAO 34, ANS Meeting June 1980.

/7/ Pellet and sphere-pac (UPu)C fuel comparative
Irradiation Tests.
A. Delbrassine, L. Smith Nucl.Tech. 49 June 1980

/8/ FILOS 07. The post irradiation examination of a
corrosion test pin clad in M-316 material and
fuelled with (UPu)C microspheres.
L. Smith, G. Bart, B. Bürgisser, R. Hofer, E. Keller
J. Mitar, D. Orciuolo, P. Petrik, J. Reindl.
EIR-403.

/9/ Performance of a sphere-pac mixed carbide fuel
pin irradiated in the Dounreay Fast Reactor
(DFR-527/1 Experiment)
K. Bischoff, L. Smith, R. Stratton. EIR-415.

6 SPHERE PAC FUEL IN A SAFEGUARDED FUEL-CYCLE

At the end of 1979 EIR joined with the Electric Power Research Institute (EPRI) of Palo Alto CA. to co-sponsor a research project at Oregon State University on a comparison of the diversion strengths of a sphere-pac fuel cycle (plant). For EPRI this was seen as a part of a much larger study on proliferation resistant (co-located) fuel recycling plants and for EIR it could provide additional justification for following the sphere pac development.

6.1 A principal concern regarding the nuclear fuel cycle is that of properly safeguarding the material being processed so as to assure that it is not diverted for use in weapons. Domestically, this concern centers on diversion by sub-national groups. Internationally, the central problem is to assure that countries acquiring nuclear power facilities do not use these facilities as a means of initiating or augmenting national nuclear weapons programs.

In spite of differences, the two problems are similar in many respects:

- The ultimate target is zero diversion,
- Even if this target is achieved, a "public" must be given reasonable assurance of this fact;
- If diversion does occur, it should be at as low a level as possible;
- If diversion does occur, its occurrence should be known as soon as feasible, so that counter-measures can be activated.

An additional aspect of safeguards, closely related to the requirement that the safeguards process provide assurance against diversion, is the desire to achieve a decoupling between nuclear power and nuclear weapons. If nuclear weapons do get manufactured, they should not be produced with material diverted from nuclear power.

The recently completed International Fuel Cycle Evaluation Program explored a variety of technical measures proposed as "fixes" for safeguarding problems throughout the fuel cycle. A general conclusion of this program was that technical fixes alone cannot provide assurance against diversion. The impression can be obtained that, indeed, the evaluation heightened appreciation of the fundamental importance of institutional safeguards. Nevertheless, in some comparisons, technical superiority of one process or system over another with regard to ease or assurance of safeguards could be concluded, even though such advantages could rarely be assigned definitive importance.

6.2 The Sphere-pac Process

A process that fits into the category of potentially advantageous technical approaches to safeguards is that of sphere-pac fuel, where the fuel is manufactured in the form of microspheres by wet chemistry methods. Fuel pin fabrication consists of loading and vibrating appropriate size fractions into clad tubing. The sphere-pac form offers the advantage of utilizing well-characterized fuel with remote pin fabrication.

As EIR has shown development of sphere-pac mixed carbide fuel for fast breeder reactors has shown promising results in comparison to more traditional fuel forms. Two size fractions of fuel are used. The fuel in many cases

outperforms pelleted fuel while retaining the advantage of carbides. A growing body of experimental evidence demonstrates a potentially reduced swelling behaviour and a more favorable overall response.

As part of the program to achieve improved performance and higher burnup, sphere-pac oxide fuel is being evaluated for light water reactors. In this application, three size fractions are used. Potential advantages in both fuel manufacture and pin fabrication have been identified.

A critical question that will mark the ultimate implementation of a development such as sphere-pac mixed carbide fuel concerns its adaptability to a safeguarded fuel cycle. In this respect also, the particulate fuels exhibit potentially significant advantages over pellet fuel forms. Specifically, the sphere-pac process is implemented primarily by wet chemical process steps that would seem to be readily adaptable to remote or automatic operation; it does not require mechanical operations that are sources of waste and contamination; and it can achieve high throughput rates in semicontinuous operation, which could be an advantage with regard to inventorying operations.

Possible use of recycled plutonium in LWR's raises many similar questions of fuel cycle safeguards. Furthermore, because of the adaptability of the sphere-pac concept to remote operation, the concept may be well suited for, other, alternate fuel cycles. In either FBR or LWR scenarios, the incorporation of sphere-pac mixed ceramic fuel into an integrated fuel recycle suggests potentially favorable impacts on the utilization of the fission energy option.

6.3 Goals of This Research

The research has been concerned with studying the sphere-pac fuel process with the aim of identifying its safeguards characteristics. This study has two objectives:

- First, to compare sphere-pac fuel cycles with the standard, pelletized fuel cycles so as to determine whether the postulated safeguarding advantages of sphere-pac are real and/or significant.
- Second, to utilize the experience of undertaking such a comparison to improve our methodologies for evaluating safguarding capabilities of processes.

Although these dual goals are separately, and indeed sequentially stated, it became apparent very early in the work that they are more closely interrelated than the statements imply. Specifically, the analysis of a fuel cycle implies the existence of methods for making the analysis, even while it is serving as a case study for further methodology development. Therefore, the two objectives have, throughout the study been pursued in parallel.

6.4 Chronicle of The OSU Study

The work documented in this report began at the beginning of academic year 1979-80, specifically on Sept. 15, 1979. The first phase of the work was a period of indoctrination and more detailed problem definition, which lasted up to the end of 1979. For these purposes, the project personnel were assigned the job of representing the flow of material throughout the nuclear fuel cycle by means of detailed flow diagrams. Emphasis was placed

on specifying points in these flow diagrams at which material is actually or potentially removed as waste, and (often, other) points within the flow at which assay or analysis is customary and/or desirable.

Two separate assignments were carried out in this way. The first consisted of overall flow charts describing the "front end" of the standard LWR fuel cycle from uranium exploration through enrichment. While not closely connected to the sphere-pac process, this assignment served to familiarize the personnel with flow chart notation, to illustrate the degree of detail that must be supplied, and to document these processes in case the work touched on them.

The second assignment was to make a first attempt diagramming a sphere-pac process, a sol-gel pelletizing process, and a blended powder pelletizing process. Because the literature on oxide fuel preparation is extensive, and because mixed oxides are at present the reference fuel material for fast breeder reactors, the study was limited to this fuel type.

In parallel a memorandum was written which touched on the interactive nature of physical security, material and process containment and surveillance and analysis. It also suggested that process yield, process envelope size (particularly in ratio to throughput rate, but also as an absolute variable), process cleanliness - which is different from yield in that a clean process of low intrinsic yield might still be a high-recycle, low-waste process, and process closure as measured by the number of envelope penetrations required, could all be considered significant variables in defining safeguardability.

Contact was established with the Exxon Nuclear Company's operation at Richland, Washington, which turned out to be

useful subsequently. These contacts were with both the fuel fabrication group, and the process development laboratory, which has been engaged in development of the sphere-pac process for slightly enriched LWR fuel, and is also studying the safeguarding of MOX sphere-pac processes.

During the summer and early fall of 1980, substantial progress was made on defining ideal processes and plants for the two fabrication processes selected. In particular, enough information was generated so that the first phases of the safeguarding comparison could begin. This comparison was carried to a semiquantitative finding in the late fall of 1980. It consists of two separate pairs of estimates. One is of analytical errors and how they contribute to uncertainties in throughput and inventory of the process: in older notation, to LEMUF (limits of errors on material unaccounted for). The other is of inspectorate forces and devices that are required. These two estimates bear, respectively, on the comparative inventorying and surveillance properties of the two processes.

In November 1980 EPRI and EIR agreed to continue supporting the project into 1981 whereby the qualitative appraisal of the comparative safeguardability of the processes would be presented as a 1980 Yearly Report and the work would move toward a quantitative appraisal using the developed methodology and considering the idealized plants.

Persons contributing

OSU B. Spinrad
K. Peddicord
M. Azarfar
A. Vincent
P. Paul
K. Green
S. Stroudt.

PART II

PROJECT STATISTICS AND PUBLICATIONS

II-1 Budget, Manpower and Costs

The project expenditure for 1980 totalled 2.54 MFr. which was 26% less than in 1979 (3.04 MFr.). The main items were (MFr.):

	1980	*(1979)*
Operations	*0.69*	*(0.68)*
Plant capital	*0.11*	*(0.46)*
Plant charges	*0.10*	*(0.04)*
Personnel	*1.60*	*(1.79)*

51247 Man hours were booked to the Project 7% less than budgeted at the start of 1980. This is equivalent to 29 Manyears (1979 = 30 Manyears).

Tables II-1 and II-2 give the breakdown of the man-power used and costs for each Vorhaben (VH). The under-booking in manpower in the VH Project Management can be attributed to little effort being available for plant engineering due to pressure of other work. The VH Post Irradiation and Analysis also heavily underbooked due to technical problems in the Hot Lab delaying completion of the DIDO-III PIE.

The underspending of the Operations Budget by 20% is largely because of the delay in the PFTF programme due to the delay in agreement signing.

Table II-2 Project Expenditures 1980 (KFr.)

Vorhaben	Operations and materials U+P	Capital plant.	Computer	Plant charges	Various	Personnel	Total	% of Pro
4.07.2 Project Management	31.35	-	-	-	-	113.48	144.83	6
4.07.4 Irradiations and Development	273.90	11.4	22.01	0.93	3.35	690.90	942.49	37
4.075 Post Irrad. Exam.and Analysis	64.76	28.97	-	98.55	5.71	165.06	363.05	14
4.079 Fuel Manuf. and Development	322.83	74.83	-	2.30	3.71	687.15	1090.82	43
Total	692.84	115.20	22.01	101.78	12.77	1596.6	2541.2	100
Budget 1980	867.00	125.0						
Total as % of Budget	80 % (without FFTF Budget - 100 %)	92 %						

Neglecting this item, the Operations expenditure reaches 100% of that budgeted.

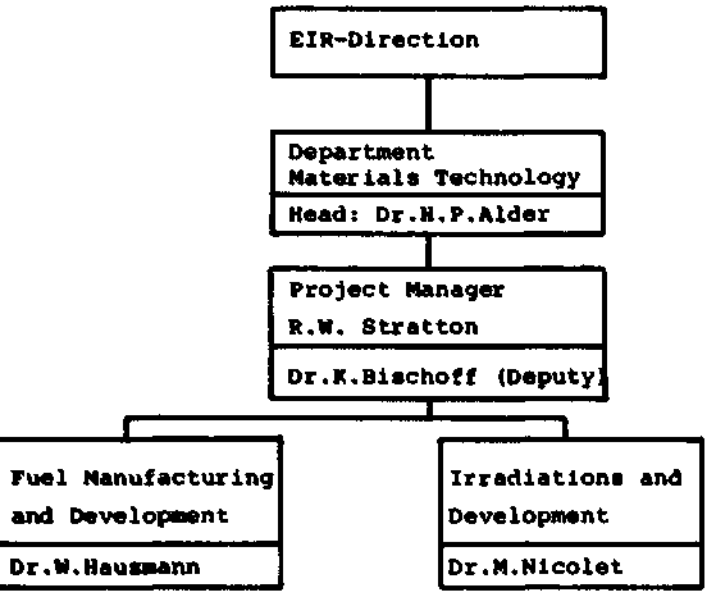
Table II-1 Personnel Expenditure

Man Hours

Vorhaben	Budget	Used	% Used
4.07.2. Project Management	2811	2097	75 %
4.07.4. Irradiation and Development	20733	20244	98 %
4.07.5. Post Irradiation Exam.and Analysis	9136	5463	60 %
4.07.9 Fuel Manufacture and Development	22665	23443	103 %
Total	55345	51247	93 %

II-2 Organization, Personnel

Changes in the Hot Labor meant that from early 1980 onwards the work of the Analytical Group was combined with the Radiochemistry group under Dr. Bart bringing together all fuel characterization and analyses both pre-and post irradiation. Dr. Huwyler, for many years the leader of the Analytical Group, moved to other duties outside the Project. We thank him for his services to the fuel development.



Dr.Ch.Jungo
G. Ledergerber (Deputy)
J. Horler
M. Gehringer
E. Keller
D. Peter
J. Wichser

F. Botta (Deputy)
H. Leute
L. Wiezel
H. Lehner
J. Sidler
F. Wyss*
vacant

Associated and supporting groups

Modelling Sphere Pac Fuel Behaviour	Fuel PIE and Analysis	Hot Cells
	Dr.G.Bart	R.Hofer

J. Reindl
vacant

T. Aerne (Dep.)
F. Petrik
I. Adeler
J. Mitar
M. Krois
E. Dickenmann
M. Mohos
Dr. Zwicky

A.Erne
A.Christen
H.Gyr
A.Bächli

Fig. II-1 Project: Fuel Development
Organization 1 Jan. 1981.

* on loan from TL

Work on fuel behaviour modelling continued in the Project "Fuel Cycle" although a large part of the effort concerned sphere-pac carbide fuel studies and is reported here (ch 5). This work suffered from loss of 2/3 of the staff in 1979/80 and during 1980 no replacements could be found. 1980 saw the departure of Dr. V. Ionescu who made worthwhile contributions in the area of thermal conductivity measurements.

One of the project's most loyal colleagues, Mr. V. Birchmeier, was promoted outside the project in 1980. Most ot the fuel produced by the project up to 1980 passed through his(gloved) hands. Joining the fuel group in 1980 were Mr. D.Peter and Mr. J.Wichser. Mr. E.Keller was transferred from the PIE and Analytical group to assist with the fuel fabrication. The small amount of activity covering plant engineering was done in 1980 under the heading of Project Management. Included in that Vorhaben was also the work contracted to Oregon State University on "Safeguarded Fuel Cycles". At the end of the year, Mr. E.Klarer was transferred to other work.

A diagram of the project organization is shown in fig. II-1.

II-3 Presentations at Conferences etc.

S. Huwyler:
K. Bischoff
(presented by
M.Güntensperger)

"Recycling of (UPu)C scrap by incineration in a controlled atmosphere"
Int. Symposium on the Management of alpha-contaminated wastes.
IAEA Vienna 2-6 June 1980.

Poster session:

"Modification of an A-DIDA secondary ion mass spectrameter to allow the examination of alpha and beta/gamma emitting specimens".
G. Bart, T. Aerne, U. Flückiger, E.Sprunger.
BNES Conference on Post Irradiation Examination, UK, May 1980.

Poster session:

A comparison between alpha-autoradiography and some measured alpha-emitting actinides in a (UPu)C sphere pac fuel.
L. Smith.
BNES Post Irradiation Examination Conference UK May 1980.

II-4 External Publications

U.Flückiger. "A Comparison between alpha-autoradiography and some measured alpha emitting actinides in a (UPu)C sphere-pac fuel".
EIR Report Nr. 389 April 1980.

G.Bart, T.Aerne, U.Flückiger, E.Sprunger

Modification of a secondary ion mass spectrometer to allow the examination of highly radioactive specimens
EIR Report Nr. 390 April 1980.

R. Stratton, K.Bischoff, F.Botta (Editors)

"Plutonium Brennstoffprogramm. Jahresbericht 1979"
EIR Report Nr. 395 May 1980.

K.Peddicord, S.Montgomery, R.Stratton

Calculations of the thermal behaviour of a sphere-pac carbide fuel pin under start-up conditions in the US Fast Flux Test Facility using the code SPECKLE-1
EIR Report Nr. 396 May 1980.

A.Delbrassine, L.Smith

Pellet and sphere-pac (UPu)C fuel comparative irradiation tests
Nuclear Technology Vol 49, pp 129-135
June 1980.

L.Smith, G.Bart, B.Bürgisser, R.Hofer, E.Keller, J.Mitar, D.Orciuolo, F.Petrik, J.Reindl.

FILOS 07. The post irradiation examination of a corrosion test pin clad in M 316 material and fuelled with (UPu)C microspheres
EIR Report Nr. 403 July 1980.

K.Bischoff, L.Smith, R.Stratton

Performance of a sphere-pac mixed carbide fuel pin irradiated in the Dounreay Fast Reactor (DFR 527/1 experiment)
EIR Report Nr. 415 October 1980.

G.Bart "Sekundaerionen-Massenspektrometer zur Analyse radioaktiver Proben"
Neue Zürcher Zeiting (Forschung und Technik)
216, 73 1980.

R.Casanova, W.Hausmann

"Wiederaufarbeitung schwach radioaktiver Lösungen"
Neue Zürcher Zeitung (Forschung und Technik)
240, 65 1980.

II-5 List of project internal publications

Note the Plutonium Programme Report reference system (PPR.Nos.) was replaced in 1980 by a 4.07 prefix with continuation of the serial numbers from the PPR prefix.
Due to the change-over several reports "fell through the gap".

TM-42-80-1 Nachbestrahlungsuntersuchungen der Hotzellen-
PPR-612 gruppe am Filos 07 Experiment"
R.Hofer, B.Bürgisser 11.1.1980.

AN-45-80-9 Qualitätssicherung bei der Brennstabherstellung
PRR-613 für das Bestrahlungsexperiment AC-3 in FTR
F.Botta 25.1.1980.

AN-45-80-12 Projektorganisation und Vorschläge für die
PPR-614 Arbeitsaufteilung im EIR
M.Nicolet, R.Stratton 11.2.1980.

TM-42-80-6 UC oxidation mit O_2 und CO_2. Thermogravimetri-
PPR-615 sche Studien
F.Petrik 21.2.1980.

TM-42-80-7 Planung der Mengenverhältnisse zur Herstellung
PPR 616 des Brennstoffes für das Bestrahlungsexperiment
AC-3 im FFTF
W.Hausmann 11.3.1980.

TM-42-80-14 Der Drehofen zum Trocknen von Uran-Plutonium-
4.07-618 Xerogel-Mikrokugeln
M.Gehringer, W.Hausmann 23.5.1980.

TM-42-80-15 Druckregulierung zur Kontrolle des Durch-
4.07-619 sätzes und der Grössenverteilung beim
Kreuzstrahl-Generator
Ch.Jungo, G.Ledergerber, D.Peter 30.6.1980.

AN-41-80-12 EIR-EPRI-OSU study on sphere-pac fuel cycle
4.07-620 safeguards
R.Stratton 24.6.1980.

AN-41-80-15 High Burn-up effects programme
4.07-621 (Note not carbide programme)
R.Stratton 24.6.1980.

AN-41-80-14 FFTF Irradiation AC-3. Report on a visit to
4.07-622 W-ARD Pittsburgh June 5/6 1980
R.Stratton 26.6.1980.

TM-42-80-23 Die neue Gelierbox im Labor 214
4.07-623 G.Ledergerber 25.7.1980.

TM-42-80-25 Quantitative phase analysis in the UC system
4.07-624 by X-ray diffraction
J.L.Delplancke, S.Huwyler 13.8.1980.

AN-42-80-20 EIR sphere-pac carbide pin. DFR irradiation
4.07-625 data for temperature profile calculations
using SPECKLE-1 code
K.Bischoff 4.9.1980.

TM-42-80-26
4.07-626 Ch.Jungo 3.9.1980.

TM-42-80-29 Aufarbeiten von C-haltigen Uranxerogel-
4.07-627 Mikrokugeln zu Uranylnitrat
M.Schaerli, W.Hausmann 9.10.80.

AN-41-80-32 FFTF Irradiation AC-3. Notes on a discussion
4.07-628 with W-ARD. Washington D.C. Nov. 1980.
R.Stratton 15.12.80.

II-6 Other internal publications

Materials Department 41

AN-41-80-5 Specifications, Rules for layout, distribution and revision
R.Stratton 12.2.80.

AN-41-80-10 Swiss interest in Pilot Plant
R.Stratton 27.5.80.

AN-41-80-11 The Fast Breeder Reactor in the EIR five year plan 1980-84
R.Stratton 19.6.80.

AN-41-80-17 Options for continuing with practical fuel activities in EIR. Note to the Arbeitsgruppe Karbidbrennstoffprogramm
R.Stratton 2.7.80.

AN-41-80-20 Summary of Nuclear Fuel Development studies in EIR
R.Stratton 19.8.80.

AN-41-80-26 Report on a visit to the Projekt Schneller Brüter (PSB) at the Kernforschungszentrum Karlsruhe (KfK), 11.11.80
R.Stratton 13.11.80.

AN-41-80-30 Notes on: Co-ordinating Commitee meeting General Atomic Co - EIR Collaboration Washington D.C. 20.11.80
R.Stratton 15.12.80.

AN-41-80-31 Discussion at the AI'S Washington meeting with General Electric, Nov.80
R.Stratton 15.12.80.

TM-41-80-1 Neptunium and its separation from spent nuclear fuel
R.Stratton 13.8.80.

TM-41-80-2 Fuel Fabrication development
R.Stratton 24.9.80.

Hot Labor Division 42

TM-42-80-28 Thermische Untersuchungen am Kohlenstoff Regal 660
F.Petrik 9.10.80.

TM-42-80-31 Survol des possibilités d'application du procédé sol-gel, en particulier aussi, à la solidification de déchets radioactifs.
J.Houriet 19.11.80.

Fuel Division 45

TM-45-80-24 Filos 07 Berechnung der Hüllrohrtemperaturen aus den gemessenen Natrium Temperaturen und der linearen Stableistung.
J.Reindl 24.6.80.

AN-45-80-26 Visit to W-ARD Pittsburgh and to HEDL. 7/8, 20/21 March 1980
M.Nicolet 22.5.80.

AN-45-80-30 Visit to GE Co. Vallecitos Nuclear Center, 27 March 1980
M.Nicolet 18.6.80.

AN-45-80-31 Visit to GAC. San Diego 26 March 1980
M.Nicolet 30.6.80.

TM-45-80-44 Mathematical Approach to the Analysis of the thermal behaviour of an Axial Heat Flow Apparatus
V.Ionescu 25.11.80.

TM-45-80-52 Entwicklung des Füllprozesses für die AC-3 Brennstäbe. (Stand und weiteres Vorgehen)
H.Leute 24.12.80.

AN-45-80-33 NFP-Pilotanlage Zusammenfassung der Koordinations-Besprechung bei Fa.Matter AG, Buchs am 26.6.80
M.Nicolet 2.7.80

AN-45-80-34 Visit to ORNL. Part I Status of
Sphere-pac fuel work
31 March/1 April 1980
M.Nicolet 17.7.80.

AN-45-80-35 Visit to ORNL. Part II Synroc
31 March/1 April 80
M.Nicolet 10.7.80.

AN-45-80-37 Comments on the "sphere-pac" situation
in the US
M.Nicolet 22.7.80

AN-45-80-47 Versuchsprogramm für die Dekontamination
von verstrahlten (sic) Brennstäben
M.Nicolet 6.11.80.