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Fabrication of Fast Reactor Fuel Pins for Test Irradiations

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FABRICATION OF FAST REACTOR FUEL PINS FOR TEST IRRADIATIONS +)

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+) Work performed within the association in the field of fast reactors between the European Atomic Energy Community and Gesellschaft für Kernforschung mbH., Karlsruhe

SUMMARY

The fabrication of vibratory-compacted UO_2-PuO_2 fuel bearing pins is described under the aspect of test irradiations under the development program of a Fast Breeder Reactor. Such a fabrication always will start with a design which is based on theoretical studies and calculation of parameters needed. Thus a start for a fuel theory had to be done, the first results of it are in use already. The procedure is such as to put in the as calculated parameters of specific power, diameter, fuel and cladding properties in formulae and thus find the maximum available burn-up and the density of the fuel pin. After that the final design is made. Then fuel and cladding selection is done and the materials are tested. Special items of the fuel control are homogeneity of the mechanically mixed UO_2-PuO_2, the Pu-analysis and the density control. Great efforts have been done on the tubing control. Here a long list of points has to be checked in a quality test. Because of its special problems the vibratory compaction method is being described though both it and the pellet line are generally followed in the development program. These special problems are the fuel density, contamination and welding. Here the development is proceeding.

THE THEORETICAL DESIGN BASIS

The target of the design is to reach a maximum burnup of about 100 000 MWd/t or in other words an operation time of 20 000 hours maximum. So it is important to know the behaviour of the fuel pin over that time. For the analysis a simple model is used: Two limitations are given for the fuel and the cladding. First the fuel-cladding contacts should be reduced to a minimum. Then the fuel operation time will come to an end after the most critical fuel cross section has been completely filled by fission-induced fuel swelling. Second the maximum inside pressure-induced plastic diametral creep deformation must not exceed 1%.

According to the temperature distribution the fuel cross-section is divided into a plastic zone V_{pl}, the columnar grain region, with

temperatures above 1700°C [1], a creep zone V_{cr} at 1300-1700°C, where creep and diffusion are of remarkable velocity, and a low temperature zone V_{lt} below 1300°C, where creep and diffusion seem to be very slow [2,3,4]. Now two data have to be known, namely the swelling rate and the rate of availability of the as-fabricated porous volume for swelling. The swelling rate caused by solid fission products at 100% dense oxide material and 10 000 MWd/t burnup seems to be 1,6% volume change, as an analytical compilation has shown [5,6,7,8,9]. The contribution of fission gas on swelling seems to be remarkable only in the creep region from 1300-1700°C. Above 1700°C almost complete immediate gas release seems to happen, whereas below 1300°C the gas remains trapped [3] with practical no effect on swelling [9]. According to different publications [3,4,10] the swelling rate due to fission gas is taken as 0,4% volume change per 10 000 MWd/t in the fuel zone V_{cr} between 1300-1700°C.

As to the availability of the porous volume to swelling there is clear indication from all irradiation experiments known that is is not complete. The evaluations are not numerous, but show that the porosity should be available to any expansion about 80% above 1700°C [11], 50% at 1300-1700°C [12,13,14] and according to a numerical evaluation not more than 30% below 1300°C. These rates might rise a bit during long operation time as the fuel becomes more plastic at low temperatures [5,15].

A maximum burnup b, after which a unit of fuel volume V_f plus the volume V_g of an as-fabricated hot gap between fuel and cladding and the volume V_d of dishing have been filled by swelling now is defined in the following way:

$$b = \frac{1}{1,6} \frac{\varrho_{th} \cdot 10^6}{\varrho \; V_f} \left[p(0,8\, V_{pl} + 0,5\, V_{cr} + 0,3\, V_{lt}) + V_g + V_d - \Delta V_m - 0,04\, V_{cr} \right]$$

b	=	burnup (MWd/t)
ϱ_{th}	=	theoretical density
ϱ	=	as-fabricated density
V_f	=	as-fabricated fuel volume
p	=	porosity = $1-\frac{\varrho}{\varrho_{th}}$

V_{pl} = plastic volume above 1700°C

V_{cr} = creep volume, 1300-1700°C

V_{lt} = low temperature volume, below 1300°C

V_g = gap volume

V_d = dishing volume

ΔV_m = volume increase by melting = $(\frac{\rho}{\rho_{th}} - 0{,}91)V_{molten}$

$\frac{0{,}04 \cdot V_{cr}}{V_f}$ = total amount of creep due to fission gas after b = 100 000 MWd/t

The creep due to fission gas is introduced here as a sum for the total burnup time; this will work somewhat pessimistic if low burnups are considered. A plot in Fig.1 teaches that e.g. for vibrated fuel, that is without gap and dishing, about 80 000 MWd/t burnup can be reached at densities of 80-82% of theoretical as described in this paper.

Numerous experiments have shown that in a sodium cooled reactor the stainless steel X8CrNiMoVNb1613 probably will be used as a cladding (TABLE I). The theoretical deliberations for a calculation of the cladding behaviour under internal pressure of fission gas and fuel are based on a creep evaluation. Here the Norton law

$$\dot{\varepsilon} = k\,\sigma^n$$

is modified to three working formulae by integrating mathematical operations

a) $$\varepsilon_t = \frac{4.k}{3(n+1)}\left[\frac{p \cdot r}{s}\right]^n \cdot t$$

b) $$p_{max} = \frac{s}{r}\left[\frac{4 \cdot \varepsilon_t\,(n+1)}{3 \cdot k \cdot t_s}\right]^{\frac{1}{n}}$$

c) $$t_{max} = \left(\frac{s}{r.p}\right)^n \left[\frac{4 \cdot \varepsilon_t \cdot (n+1)}{3k}\right]$$

ε_t = amount of plastic diametral deformation after time t

k = creep constant

n = creep exponent

p = internal pressure
r = inner radius of the tubing
s = wall thickness
t = operation time

Under the assumption that t_{max} should be 20 000 hours and ε_t not more than 1%, p_{max} results to 110 kg/cm^2 with s = 0,4 mm and r = 2,775 mm, Fig.2. Thus two data which are standing in close correlation to each other, could be evaluated for this pin design, namely the specific ratio $\frac{s}{r}$ and the size of the gas plenum, Fig.3, TABLE II. The lay-out of the gas plenum was such as to meet 100% gas release and release of absorbed gases under fabrication, which is about 0,5 cm^3 (0°C, 1 atm) per 1 cm^3 fuel, so that the gas pressure at operation temperatures will be about 80 kg/cm^2. The rest to p_{max} of 20-30 kg/cm^2 probably will not be exceeded by fuel-clad contact pressure at fuel surface temperatures above 900-1000°C and central temperatures near the melting point before total consumption of available voids has happened as an evaluation shows which includes recently published mechanical oxide data [16,17].

FUEL PREPARATION, MATERIAL SELECTION AND CONTROL

UO_2-PuO_2 powder for vibro-compaction has to be supplied for 20 fuel pins. The target enrichment was 15 wt.% Pu, the uranium fully enriched. The blanket material should be natural UO_2. First 93% enriched UO_2 was prepared from UF_6 by a H_2/H_2O mixture at 700°C and then milled to grain sizes of about 1-2 microns average diameter. PuO_2 was received from Hanford, the natural UO_2 was delivered by industry. The characteristic quality data of the source materials are given in TABLE III. Now the following procedures of the fuel production will be described.

a) mixing
b) granulation
c) pellet pressing
d) breaking of pellets
e) rounding of particles
f) dewaxing and sintering.

Dry mixing of the oxides was performed in a special equipment (Lödige type mixer) for 8 hours. After that the mixture was prepared for granulation in the same equipment by addition of a 14% polivinyl-alcohol mixture and a 12% alcoholic solution of stearic acid up to contents of 1,2% polivinyl alcohol and 1,0% stearic acid. Green pellets of 10 mm diameter then were pressed at 5-6 t/cm^2 pressure to densities of about 6,7 g/cm^3. Breaking the pellets to 1,8 - 2,5 mm again supplied the material for the rounding process which was done in cylindrical rotating equipment with a coarse inner surface. After periods of 2-4 hours portions of target grain fractions were separated, the rest being further rounded. The resulting grain fractions were dewaxed for 2 hours at 400°C and 2 hours at 750°C unter CO_2-atmosphere. The sintering time was 3 hours at 1600°C under an Ar/6 vol.% N_2 gas mixture. The heating rate must not exceed 400°C/h. A total material balance of the fabrication procedure is given in TABLE IV.

The quality control of the fuel consisted of Pu-analysis, sinter density, stochiometry and homogeneity, TABLE V. The Pu-analysis, for which the amperometric method, using AgO for oxidation of Pu, was used, showed, that the accuracy of Pu-U mixture was not better than 15 $\pm$ 0,2 wt.%, an experience which was stated very often before. The problem of mixing UO_2 and PuO_2 to a target mixture has lead to different studies on that item with the result that within one pellet the Pu-contents can be 15 $\pm$ 1 wt.% and 15 $\pm$ 0,2 wt.% from batch to batch. The density control by pycnometric analysis gave results of 91 $\pm$ 1%, with the coarse fractions. The determination of the O:M ratio by oxidation to $PuO_{2,00}$ and U_3O_8 still is somewhat unsatisfactory as it only gives the total result and does not allow a conclusion as to the state of single components. The homogeneity of the material analysed by autoradiography gave indication that the PuO_2 particle size would not exceed 0,1 mm.

The quality control of the tubing. Dimensions of the tubing, specified as given in TABLE VI were tested with mechanical, air gauge and ultrasonic-vidigauge methods. The specification did comprise the following items:

Surface conditions:
Surface cracks and seams are not permitted, tested with ultrasonic method.
Average surface roughness shall not exceed 2 µm, tested with profilograph.
Surface groves and defects shall not exceed 20 µm, tested by ultrasonic method.
Inside and outer surface must be free from discoloration, grease, dirt, metal particles and other foreign matter, tested with qualified optical method.

Material conditions:
As annealed, maximum grain size ASTM No. 5.
Inner defects and discontinuities shall not exceed 10% of the wall thickness, tested with ultrasonic method.
Inclusions and stringer: Samples were taken from five finished tubings of each alloy and inclusion contents determined on longitudinal sections were no worse than Background Classification "C" Plate 9 (ASTM E45, Specification 1965, Part 31).

Each length of tubing must be hydrostatically pressure tested at 20°C to 200 atmospheres.
Flattering Test according to DIN 50136 and ASTM A 450-65.
Flaring Test according to DIN 50135 and ASTM A 450-65.
Rockwell Hardness Test of the outside of the tube.

THE PIN FABRICATION

The parts of the cladding including the end caps were machined in GfK workshops, the welding was done with the electron beam method. The fuel, the grain fractions given in TABLE VII, was vibrated by an electrodynamic equipment in a range of 400-4000 Hz. This range was covered 30 times for getting the final density of 81 $\pm$ 3%. A special problem came up from the fine dust contaminating the end of the tubing when vibrating the fuel. Thorough mechanical decontamination is leading to tight adhesion of organic means which is the reason

for porosity on electron beam welding. The difficulties seem to be overcome by application of chemical decontamination means and argon arc welding. The quality control was performed by dimensional and weight checks, He-leak test, γ-scanning density control and X-ray testing of the welds. The X-ray method applied uses a special equipment and allows thorough control of the welds.

CONCLUSIONS

The beginning theoretical deliberations which have lead to some working formulae have been applied for the development of fuel pin design for a test irradiation covered by the development program of the Fast Breeder Reactor. With such a lay-out 20 pins with UO_2-PuO_2 fuel have been loaded with vibratory compacted powder to a density of 81 $\pm$ 3 %, which density might allow a burnup of 80 000 MWd/t. The difficulties during fabrication came from mixing UO_2 and PuO_2, decontamination and welding. Here good experiences were gathered. The development of the tube testing program has lead to a standard check list of thorough control performance. In conclusion it can be said that the production was a first successful step to pin fabrication experience.

ACKNOWLEDGEMENT

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TABLE I - STAINLESS STEEL X8CrNiMoVNb1613 PROPERTIES

a) Analysis

Nominal analysis	wt.%
Ni	12,5 - 14,5
Cr	15,5 - 17,5
Mo	1,1 - 1,5
Nb	10 x C + 0,4 %
V	0,65 - 1,2
Mn	1,0 - 1,5
C	0,1 max
Si	0,3 - 0,6
B	20 ppm max
N	0,1

b) Mechanical Properties

Temperature °C	Yield Strength k/mm^2
650	42,2
700	36,7
750	31,2
800	27,3
850	22,0

TABLE II - PIN DESIGN

	Breakdown (mm)
Upper End Cap	16,1
Upper Gas Plenum	188,4
Blanket	150,0
Fuel Zone	700,0
Insulator Pellet	8,0
Lower Gas Plenum	195,0
Lower End Cap	28,8
Total Length	1286,3

TABLE III - SOURCE MATERIAL PROPERTIES

material	BET-Surface m^2/g	grain size µm
UO_2-nat.	3,6 - 3,8	< 125
UO_2-enriched	1,6 - 1,8	30 - 80
PuO_2	5,5 - 5,8	25 - 30

TABLE IV - MATERIAL BALANCE

Autoradiography	3,14 %
Loss on Dewaxing	2,21 %
Fuel	85,17 %
Rest for Refabrication	6,10 %
Waste	3,38 %

TABLE V - AS-FABRICATED FUEL QUALITY

Sinter Density	91 $\pm$ 1,0 wt.%
O:U Ratio	2,00 $\pm$ 0,015
Pu-Analysis	15 $\pm$ 0,2 wt.%
Loading Density	81 $\pm$ 3 %
PuO_2 Particle Size	$<$ 0,1 mm

TABLE VI - TUBING SPECIFICATIONS

Inner Diameter	5,55 $\pm$ 0,025 mm
Wall Thickness	0,4 $\pm$ 0,02 mm
Ovality	within max. and min. diameter
Straightness	1:1500 per 30 cm length
Tubing Length	1257,4 mm

TABLE VII - FUEL GRAIN FRACTIONS

grain size (mm)	percentage
1,5 - 1,25	40
1,0 - 0,8	20
0,2 - 0,1	25
$<$ 0,06	15

$$b = \frac{1}{1.6}\,\frac{\varrho_{th}\cdot 10^{6}}{\varrho\cdot V_f}\left[P(0.8\,V_{pl}+0.5\,V_{cr}+0.3\,V_{lt})+V_g+V_d-\Delta V_m-0.04\,V_{cr}\right]$$

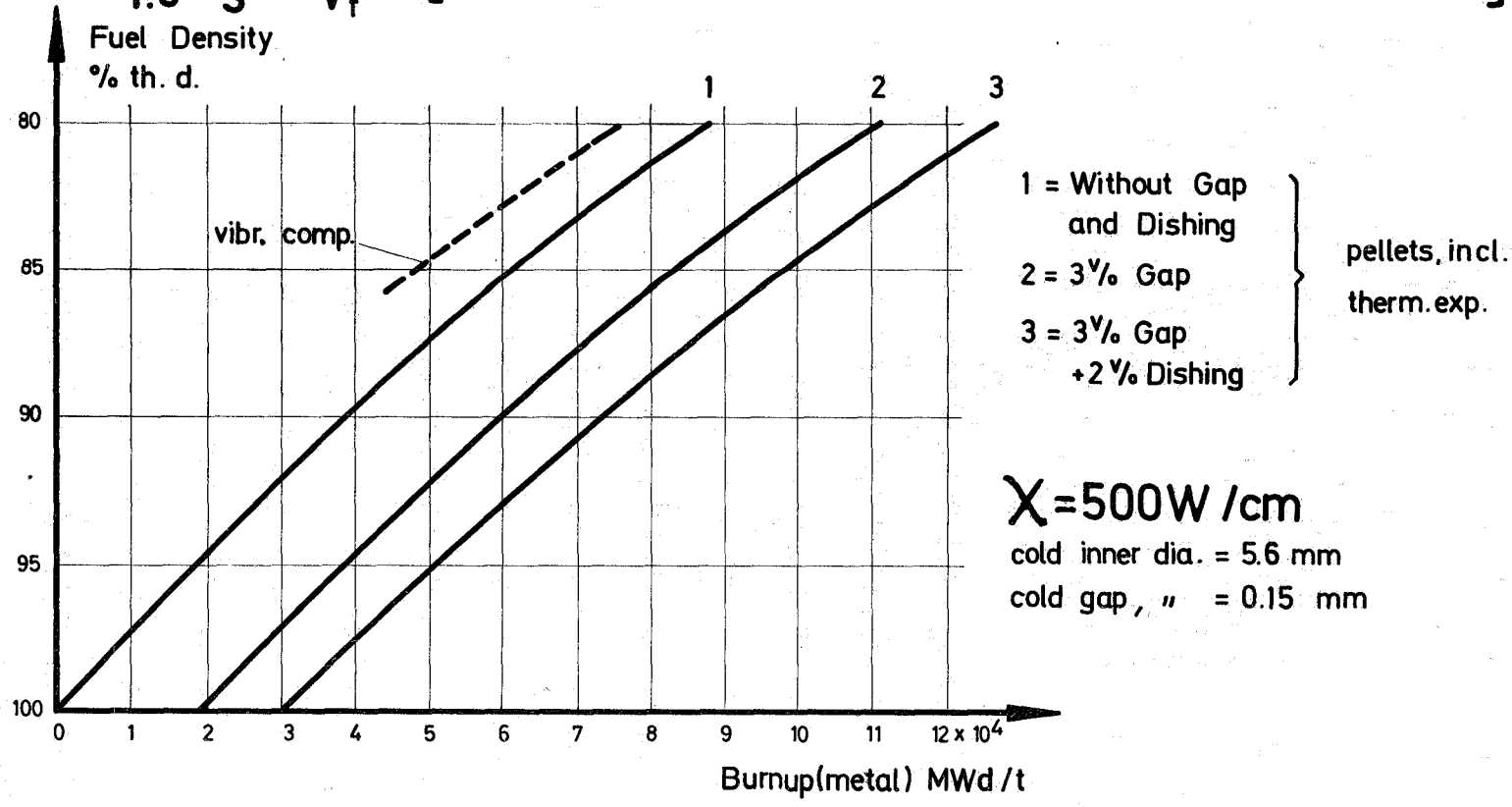

Fig. 1 Maximum Fuel Burnup Dependence on Density (cold)

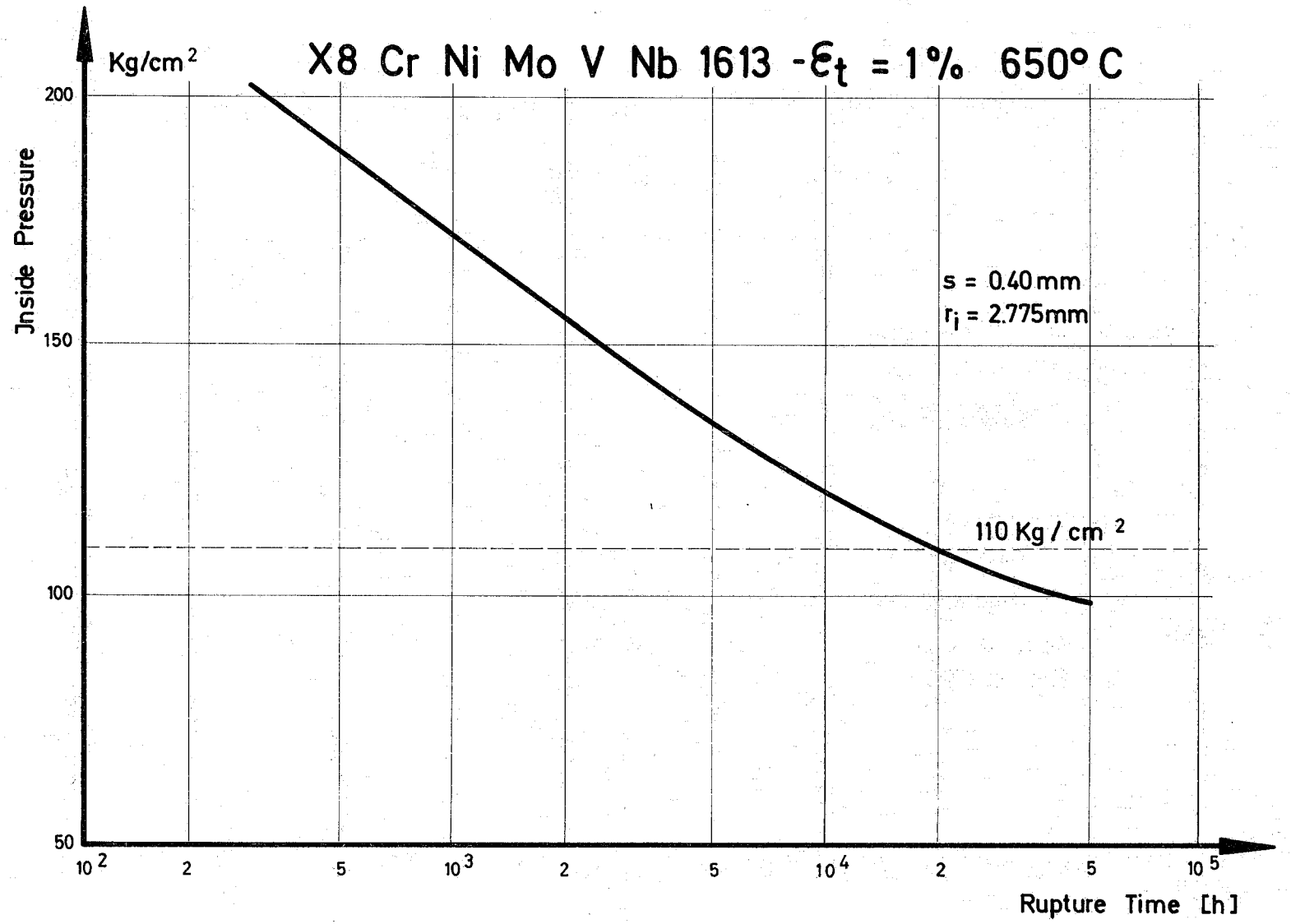

Fig. 2 Influence of Tubing Parameters on Pin-Layout

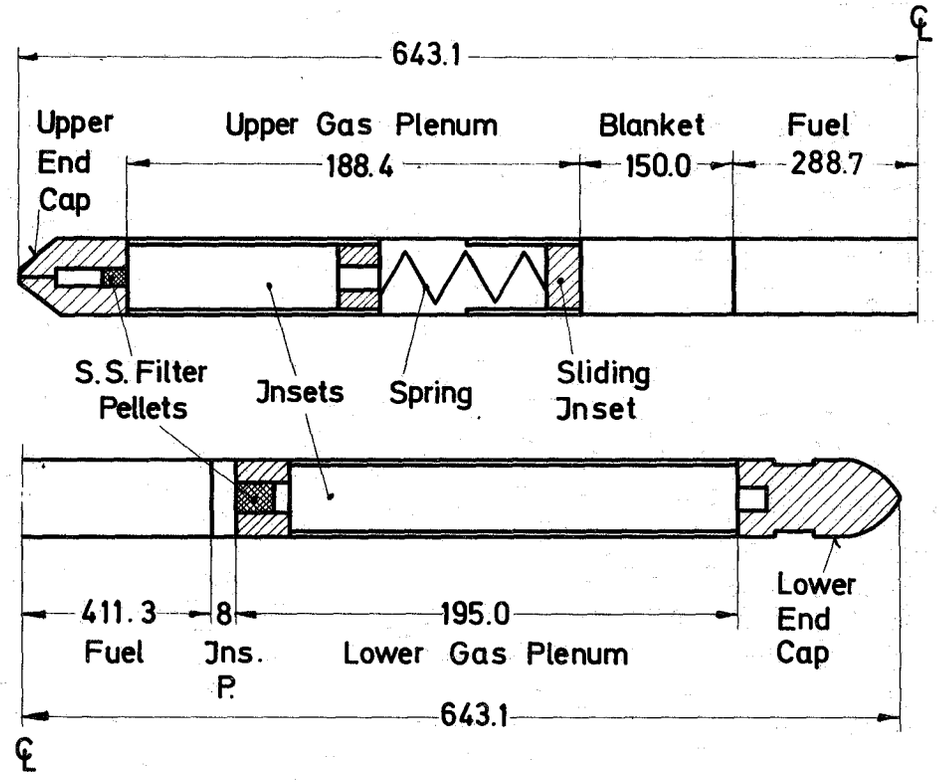

Fig. 3 The Pin Design (schematic)