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A b s t r a c t

The German Carbide Program: Performance, Experimental Findings, and Evaluation of Irradiation Results

In this report a synopsis of the German carbide program is presented. The program comprises the irradiation of about 100 carbide pins equipped with pelletted fuel. Most of these fuel pins were He-bonded, the sodium bonding concept taken as a back-up solution.

The main design parameters such as smear and pellet density, gap size, pin diameter and wall thickness as well as the irradiation conditions were varied mostly within wide ranges.

Based on a compilation of relevant pin parameters, irradiation conditions, and the results of various irradiation experiments conclusions on the optimum ranges of the main design parameters are drawn. Furthermore, some important aspects of fuel pin behaviour are discussed based on quantitative results from post irradiation examinations.

Z u s a m m e n f a s s u n g

Das deutsche Karbid-Programm: Durchführung, experimentelle Ergebnisse und Auswertung der Bestrahlungsergebnisse

Im vorliegenden Bericht wird eine zusammenfassende Darstellung des deutschen Karbidbrennstoff-Bestrahlungsprogramms gegeben. Das Programm beinhaltet die Bestrahlung von etwa 100 Karbidbrennstäben mit Tablettenbrennstoff. Neben der Darstellung des Verhaltens von Stäben des Helium-Bindungskonzepts werden auch Stäbe mit der sogenannten Natriumbindung untersucht.

Bei den Bestrahlungsexperimenten wurden die Hauptauslegungsparameter wie Schmier- und Tablettendichte, Spaltweite, Stabdurchmesser und Hüllwandstärke sowie die Bestrahlungsbedingungen innerhalb weiter Bereiche variiert.

Unter Zugrundelegung der wesentlichen Stabparameter, der Bestrahlungsbedingungen sowie der Ergebnisse der Bestrahlungsexperimente werden Folgerungen hinsichtlich der optimalen Auslegungsparameter gezogen. Basierend auf quantitativen Ergebnissen der Nachuntersuchung werden schließlich einige wesentliche Aspekte des Brennstabverhaltens diskutiert.

C o n t e n t s :

INTRODUCTION

Initiated in 1968 at Karlsruhe, FRG, the German carbide program aimed at a widespread investigation on the qualification of (U,Pu)C as Fast Breeder Reactor fuel. The results of this research and development program should lead to an optimized carbide fuel pin concept being qualified to undergo the German licensing procedure[1,2].

In the first seven years each of the two basic pin concepts with gas and sodium bonding has been investigated in parallel. In spite of the rather good performance in fabrication and irradiation of sodium bonded fuel pins this alternative has been deferred in favor of the technically easier gas bonding type.

In the following the carbide program as well as experimental results and their theoretical interpretations are presented.

THE GERMAN CARBIDE IRRADIATION PROGRAM

Apart from in-pile and out-of-pile tests with small fuel samples for the investigation of special material phenomena under irradiation like fuel creep and swelling the whole program comprises the irradiation of 86 carbide fuel pins. 61 of these pins were gas-bonded (He,Ar), 25 pins Na-bonded. A compilation of all these experiments together with the most important design parameters and irradiation conditions is given in Table I. All the experiments were carried out with pellet-type fuel.

The design parameters being considered as decisive for pin performance have been varied partially within wide ranges: The variations of greatest importance concern the smear density (72 - 91 % TD.), the pellet density (83 - 96 % TD.), the diametral gap size (50 - 900 μm), the pin diameter (5.6 - 10.0 mm), and the wall thickness (0.3 - 0.55 mm).

The fuel fabricated by carbothermic reduction of the oxides UO_2/PuO_2 [3] is a mixed (U,Pu) carbide with Pu-contents between 15 and 40 % Pu. During the carbide development period emphasis was layed on fuel quality improvement, especially on the reduction of N and O contents and hyperstoichiometric phases, leading to fuels with equivalent carbon contents around 5.00 w/o.

All the single pin irradiations have been carried out in the FR 2, BR2, and HFR reactors under thermal and epithermal conditions, while three 7 pin bundle tests were conducted under relevant fast breeder conditions in the DFR reactor. At maximum linear rod powers between 700 and 1400 W/cm the whole burnup range up to 100 MWd/kg metal was covered. At present time a total of 13 fuel pin failures was detected which can be attributed to irradiation capsule malfunctioning, fuel-clad mechanical interaction and clad carburization.

The entirety of experimental results and theoretical interpretation led to a He-bonded reference concept which shall be tested in the KNK-II reactor at Karlsruhe (Table II).

PERFORMANCE OF CARBIDE FUEL PINS AND DESIGN CONSIDERATIONS

For a distinct assessment of the performance of the fuel pin concepts tested the burnup values reached in the various experiments together with the defect rates which are identified as real pin failures, compiled in Table I, can be used. In view of the range of investigated design parameters, the limited number of irradiated pins, and the rather different irradiation conditions a statistical evaluation of the irradiation outcome is fairly inadequate. Therefore, conclusions on the optimum ranges of decisive design parameters are drawn from direct comparison of experimental results taking into account the different neutron flux conditions. One of the main differences between thermal and fast flux irradiations is the steep flux gradient under thermal neutrons leading to a high fission rate and burnup in the outer fuel zones which results in a more severe fuel and pin load. In the following the two Na and gas bonded pin types, by reason of different irradiation behaviour, will be discussed separately.

GAS-BONDED CARBIDE FUEL PINS

Gap heat transfer and mechanical interaction between fuel and cladding as two critical aspects of pin behaviour lead to the investigation of smear and pellet density, gap size and wall thickness.

As is suggested from Fig. 1 reachable burnups without pin failure seem to increase strongly with decreasing smear density: Burn up values of 7 at-% and more have been safely reached with smear densities of $\leq$ 80 % TD., fuel pins with smear densities $\geq$ 82 % TD. failed. The results in Fig. 1 furthermore suggest a failure threshold at smear densities of 81 - 82 % which is confirmed by an evaluation of carbide irradiation experiments in EBR II[4], showing a sharp increase of defect rates at smear densities of about 82 % TD. However, some of the failures occurred under thermal flux where the effective burnup in the interacting fuel rim is higher than indicated in Fig. 1, thus shifting the threshold line towards higher smear densities, resp. higher burnups. Some of the reasons for a density threshold can be seen in enhanced creep rates and accommodation of fuel swelling in the void volume at lower densities and a sharp increase of clad strain at higher densities.

Combinations of pellet densities above 90 % TD. and smear densities above 80 % TD. as realized in the experiments Vg. 6d and Mol-11/K1 and /K4 did not succeed in reaching high burnups at rod powers above 800 W/cm (s. Table I). The results of American[4] and Russian[5] irradiations suggest, however, that these pin concepts are feasible at lower linear rod powers ($\simeq$ 600 W/cm).

Intermediate pin concepts with higher pellet ($\simeq$ 90 % TD.) and low smear densities (< 75 % TD.) seem to meet the requirements at rod powers above 1000 W/cm (DFR-330/2), the disadvantage here being very large as-fabricated gap sizes above 600 μm and thus raising some problems with low gap heat transfer resp. very high fuel temperatures, especially during start-up. High temperatures at that moment, however, allow the fuel to exceed the transition temperature brittle-to-ductile ($\simeq$ 1200 °C) in a great part before reaching linear rod powers necessary for significant fuel cracking (550 - 600 W/cm). This obviously results in a considerable reduction of fuel cracking as is demonstrated by fuel aspects of destructive PIE in the case of the experiment DFR-330/2.

The above results show that the as-fabricated gap size can be varied within wide ranges and is no further a primary design parameter if the correlation between smear and pellet density is correctly adjusted in view of irradiation conditions and burnup. High pellet with high smear densities as well as high pellet with low smear densities should be avoided in the case of He-bonded fuel pins.

SODIUM BONDED CARBIDE FUEL PINS

The results of the Na-bonded pin irradiations were quite satisfying. Only one genuine pin failure was ascertained in this case, which could be related to excessive clad carburization in connection with locally enhanced mechanical interaction between fuel pellet and cladding. In this case, however, exceeding oxygen and carbon contents of the as-fabricated fuel are viewed to have played the dominant role.

The two other failures in the Mol-11/K3 series have been explained by gas blanketing in the NaK gap of the irradiation capsule leading to an excessive rise of the cladding temperature and subsequent clad damage.

In the tested range of smear density (72 - 82 % TD.) no clear evidence of the influence of this parameter on pin performance could be derived as is shown in Fig. 2. The competitive role of the Na-bonded pin concept against the He-bonded type can be seen in the fact that three fuel pins have safely attained a burnup of about 11 at-% at maximum linear rod powers between 800 and 1100 W/cm.

Considering the intrinsic problems of this pin concept - partial loss of the Na bond by fission gas accumulation, local mechanical interaction and enhanced clad carburization - operation at lower linear rod powers seems to have some decisive advantage. A compilation of best Na-bonded pin performances against linear rod power in Fig. 3 indicates an increase of attainable burnups with decreasing rod power.

Despite the few irradiation experiments interpretation of the results and derived consequences for concept modifications as e.g. fuel column liners with possible gettering effects show the high potential of this fuel pin concept, probably much higher than that of the He-bonded fuel pins.

SOME ASPECTS OF FUEL PIN BEHAVIOUR UNDER IRRADIATION

For the confirmation of safe operation of carbide fuel pins heat transfer and mechanical interaction between fuel and cladding as the main aspects concerning fuel pin performance have to be investigated thoroughly.

The heat transfer limiting gap conductance between fuel and cladding in pins with medium and large as-fabricated gaps remains - due to the high fuel thermal conductivity - the dominant factor for potential fuel melting, especially during start-up. As gap conductance improves with increasing gas temperature and decreasing gap width a distinct enhancement of heat transfer is realized with increasing linear rod power (Fig. 4): Experimental heat conductance values at start-up of fuel pins with diametral gaps of 400 μm and 650 μm are plotted versus linear rod power. After start-up the heat transfer improves rather fast as is shown in Fig. 5, where measured fuel centerline temperatures and derived gap conductance values for a 400 μm gap from the CATRI experiment in the HFR Petten for the first 20 days of irradiation are shown. After a 10 days irradiation the gap conductance already reaches values of 1 W/cm^2K, indicating a fast decrease of the gap width. The influence of fission gas on the gap conductance at begin-of-life can be neglected because of very low release rates up to 2 % burnup with the exception for very large gaps around 900 μm.

Thus, the improvement of heat transfer with increasing linear rod power extends the power design limit considerably: Incipient fuel melting at 1500 W/cm for as-fabricated diametral gap sizes of about 400 μm is reported in [6].

The understanding of the mechanical interaction and resulting specifications for design and operation is of an utmost importance for the viability of a pin concept. As has been derived from experimental findings, the reduction of smear density is most effective concerning the extension of burnup limits. As most of the fuel pins failed because of high clad strain due to excessive fuel swelling the mechanical interaction has to be limited to a defined value at the end of life. In Fig. 6 the mechanical clad strain of gas-bonded fuel pins for various smear densities as function of burnup is shown: smear density clearly separates the experimental data points, the strain rate sharply decreases with decreasing smear density.

Besides the consideration of inherent fuel and fuel pin characteristics, a thorough analysis of reactor operation has been carried out with the aim to look at additional pin load by severe cyclic reactor conditions: In spite of an excessive 30 - 40 shutdown and scram scenario of the BR 2 high burnup Mol-11 fuel pins show that clad distensions remain low for smear densities below 80 % TD. Furthermore, those smear densities seem to favour the accommodation of fuel swelling in the available porosity (Fig. 7) explainable by the clearly reduced restraint swelling rates of the fuel. All the results show that high pellet density (> 90 % TD.) combined with high smear density

(> 80 % TD.) and high rod power (≃ 1000 W/cm) is disastrous for He-bonded pin integrity, since more than three of these pins are believed to have failed at rather low burnup (5 - 6 %) in spite of a large 500 μm initial gap.

The analysis of these results show that the He-bonded pin concept has a limited potential in comparison with Na-bonded pin design, which does not show any dependance of smear density.

For the interpretation of fuel pin performance under irradiation the fission gas behaviour plays an important role with respect to swelling and bonding implications. Being quite different with respect to oxide fuel carbides are able to store gases: Significant gas release starts at burnups of about 2 at-%. Fig. 8 shows typical fission gas release rates over burnup, as well for He-bonded as for Na-bonded fuel pins: Na-bonded fuel releases fission gases very late due to its low average temperature. Besides temperature independant saturation effects, the initial fuel density seems to be a decisive retention resp. release rate factor: In high dense fuel pellets the overwhelming part of the retained fission gas is deposited in the matrix and small fission gas bubbles (Fig. 9). Fig. 9 shows the difference between fission gas retained in the matrix and small (≤ 1 μm) and large (> 1 μm) bubbles. Whereas the matrix stored gases seem to saturate over burnup, pore retained fission gas increases. Comparing the evaluation of restraint fuel swelling data with fission gas retention as function of smeared density, the coincidence of an increase of gas release with a decrease of restraint swelling rates with decreasing smear density seems to be functional.

One of the interesting phenomena in irradiated fuels is the redistribution of Pu and fission products. Carbide fuels show some redistribution effects which are helpful for modeling applications, especially the fission product agglomerations observable in nearly all carbide fuels. Thermodynamic considerations attach temperature levels to the deposition of complex f.p.C.[7] compounds: ≤ 1200 °C region allows the formation and thus concentration of these complex carbides, consequently, acting as temperature indicator.

Pu redistributions are mostly observable in crack regions at the outer surface sometimes impaired by Am-presence.

The results of PIE findings of Pu and F.P redistributions do apparently not affect the performance of fuel pin behaviour under steady state operation. The influences of e.g. Pu enrichment in the outer zones of He-bonded pins during transient overpower conditions, however, should be investigated.

INTERPRETATION OF PIE RESULTS

The overall performance of the fuel pins is of course governed by the main design parameters. Detailed post irradiation examination, however, is necessary for the understanding of the various mechanisms that influence the pin behaviour.

In the He-bonded concept cracking of the fuel pellets has been of much concern. Experiments show that although chips may break off from the pellet periphery they must not necessarily impair pin performance. As shown in Fig. 10 chips are attached to the main pellet matrix by a crack healing process which is certainly connected with the high temperature level that exists in He-bonded pins. By this way the symmetrical temperature distribution in the pellet is maintained as can be seen by the dark ring in the etched specimen.

For the large as fabricated gap in He-bonded pins a phenomenon has been observed that is able to improve the heat transfer situation substantially. As can be seen in Fig. 11 the fuel pellet is kept in its centrical position by a grid-like structure which connects the periphery of the pellet with the cladding. The material for this structure is vapor deposited fuel of different composition. Since this zone still maintains a rather high porosity also the hazard of fuel cladding interaction is reduced.

In contrast complete cracking of the original pellet seems to be typical for Na-bonded pins (Fig. 12). Although substantial restructuring has occured in the fuel matrix the pieces have been left separate, presumably due to the lower temperature level and the sodium acting as barrier.

As far as fuel cladding interaction is concerned, the broken pellets still behave like solid cylinders. Similar to light water reactor fuel pins local strain peaks are produced in the cladding at the position of pellet/pellet interfaces (Fig. 13). This effect, however, can be suppressed by a proper selection of the canning material.

CONCLUSIONS

The German carbide fuel irradiation program, covering a large field of parameter variations, resulted in a valuation of possible pin concepts. Taking into account available international experience the results of irradiation experiments, modeling, and PIE findings, lead to a recommendation of a feasible carbide fuel pin concept up to representative burnups. The analysis of decisive parameters on safe pin behaviour as heat transfer, mechanical interaction between fuel and cladding combined with fuel fabrication experience, lay out parameters as fuel- and smear density, and operation conditions, basing on quantitative PIE results and calculations, gives a clear picture of feasible concepts: He-bonded carbide fuel pins although well performanced have a limited potential in what concerns allowable smear density and rod power for a reasonable burnup. Na-bonded pins, in spite of less irradiation experience seem to be a highly valuable concept in view of pin performance and very high burnups. Failed fuel pin behaviour seems to be less severe than in the oxide case.

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Table I: Irradiation experiments with carbide fuel pins

experiment	number of p.	clad dim. (mm x mm)	dia. gap size (μm)	Bond.	sm. dens. (% th.D.)	pell. dens. (% th.D.)	lin. rod. pow. (W/cm)	Burnup (MWd/kgM)	failed pins
FR2-Loop 4a	8	10x0.55	60/80/250/600	He/Ar	76/82/86	87	800-1100	<0,2	—
FR2-Loop 5k	8	8x0.45	30	He	82/88/91	85/91/93	800-1000	7-17	—
FR2-Vg6a	7	10x0.55	50	He	82/83/86	83/84/87	800-1200	1/15/25/50	1
6c	6	10x0.55	90	Ar	83/85/86	85/87/88	800-1100	21/27/33/52/68	3
6d	3	10x0.55	500	He	82	92	700-1050	74/77/81	3
6e	3	10x0.55	500	Na	82	92	650-1050	67/87/99	?
Mol-15	4	5.6x0.3	380/530	Na	75/80	92-94	1000-1150	75/82/87	—
Mol-11/K1	1	8.0x0.45	100	He	91	93	1150-1350	38	1
/K2	3	8.0x0.45	140/360	He	75/80	83	800-1150	41/68/74	—
/K3	4	8.0x0.45	750	Na	72	90	700-1000	<10/100	2
/K4	2	8.5x0.55	600	He	81	97	700- 800	40/66	1
/K5	4	8.5x0.55	400	He	75	84	700- 800	20	—
CATRI	6	8.5x0.55	200/400/650	He	70/75/80	84	900-1000	66/83/87	—
CAREL	6	8.5x0.55	400	He	75	84	800	70	—
DFR-330/1	7	9.5x0.5	800	Na	78	95	1150-1200	53	1
/2	7	9.5x0.5	580/900	He	72	83/90	1000-1100	63	—
/3	7	9.5x0.35	900/930	Na	72	90/93	1000-1150	75	—

Table II: Main data of the reference carbide pin

Fuel:	(U, Pu)C
Pellet density:	84% th. D.
Smear Density	75% th. D.
Bonding:	Helium
Cladding:	1.4970 cold worked SS
Pin diameter:	8.5 mm
Clad wall thickness:	0.55 mm
Max. nom. heat rating:	800 W/cm
Max, nom. burnup:	70.000 MWd/tM
Max. nom. clad temp.:	620°C

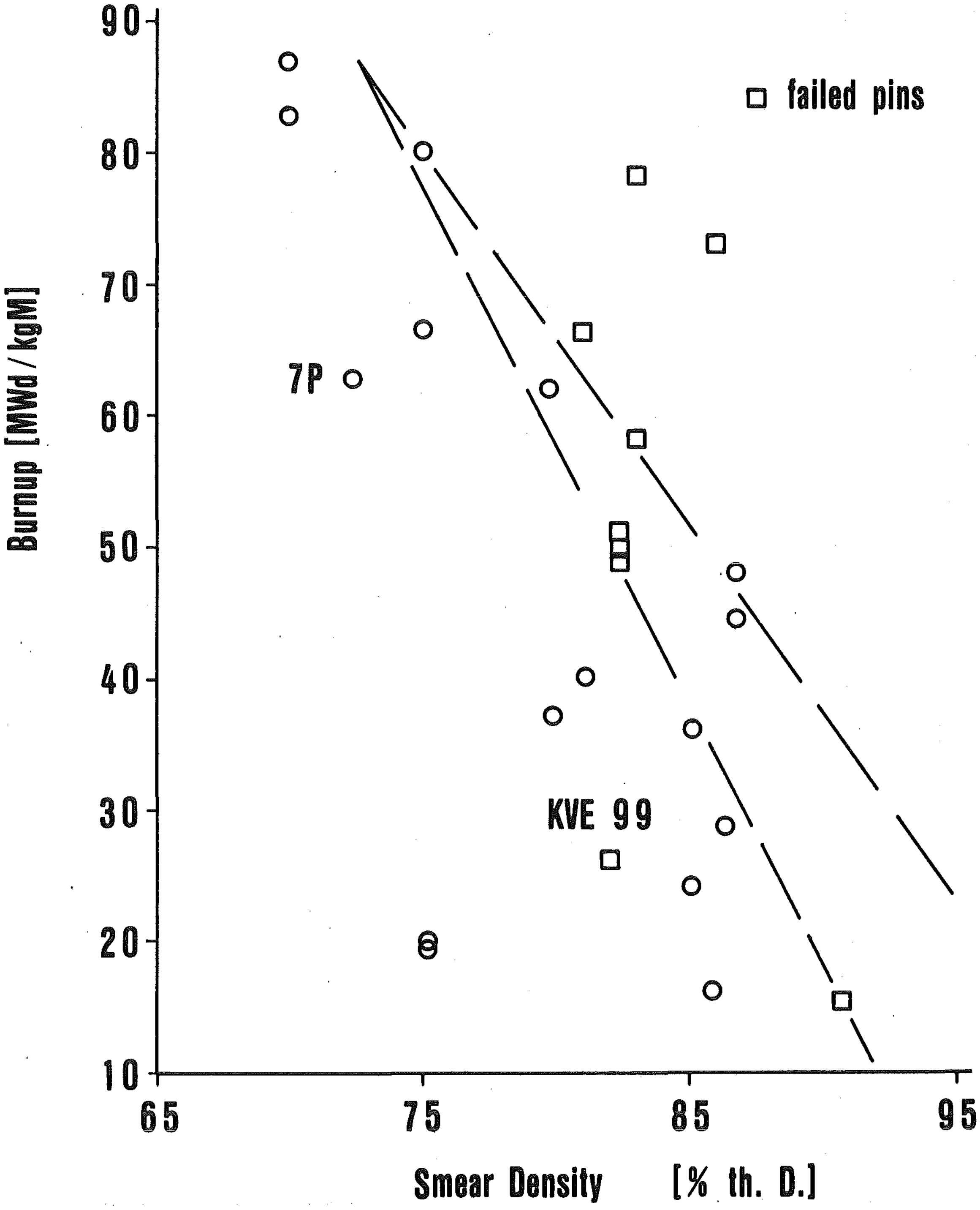


Fig. 1 Burnup of He-bonded pins versus smear density

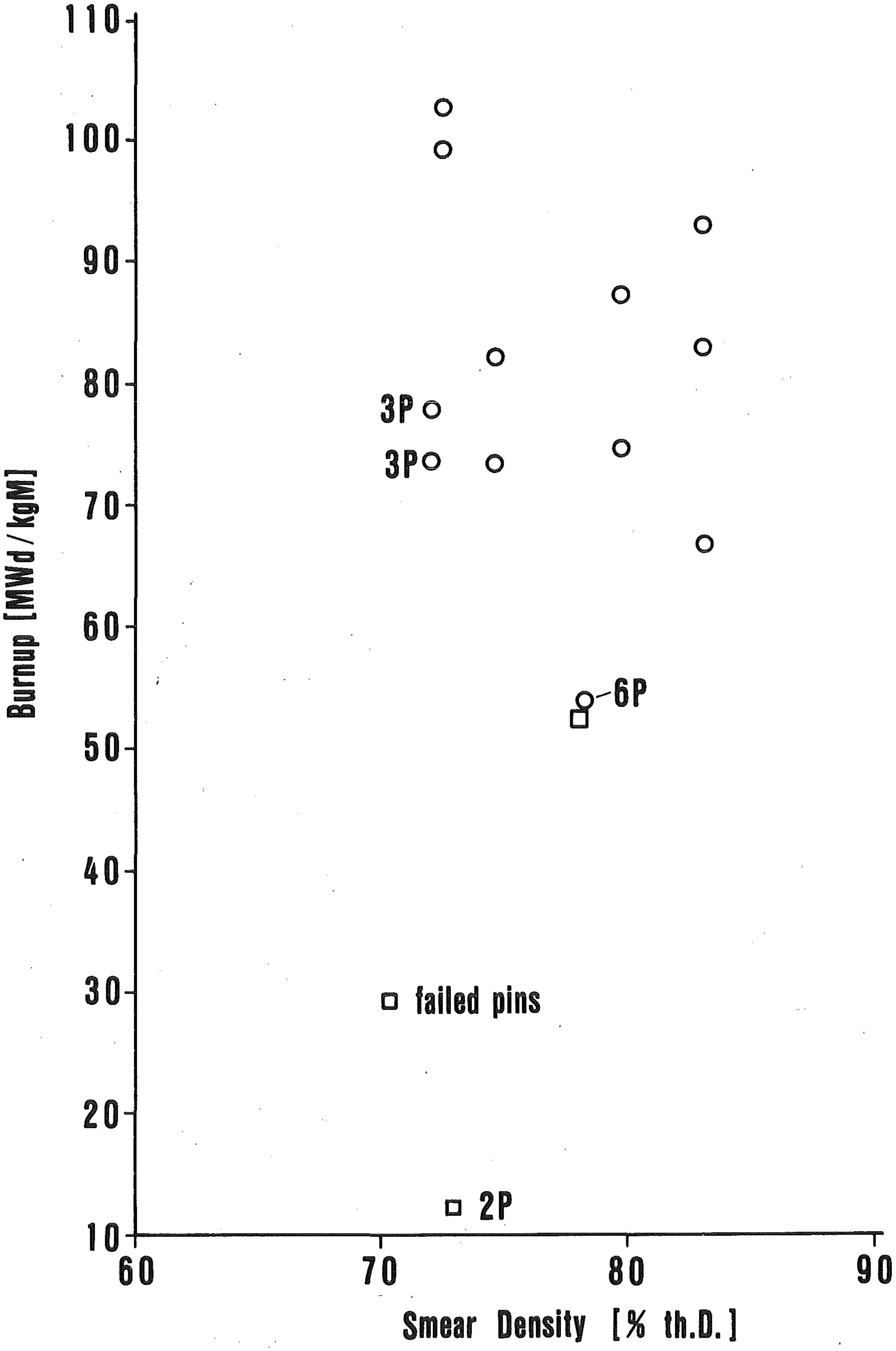


Fig. 2 Burnup of Na-bonded pins versus smear density

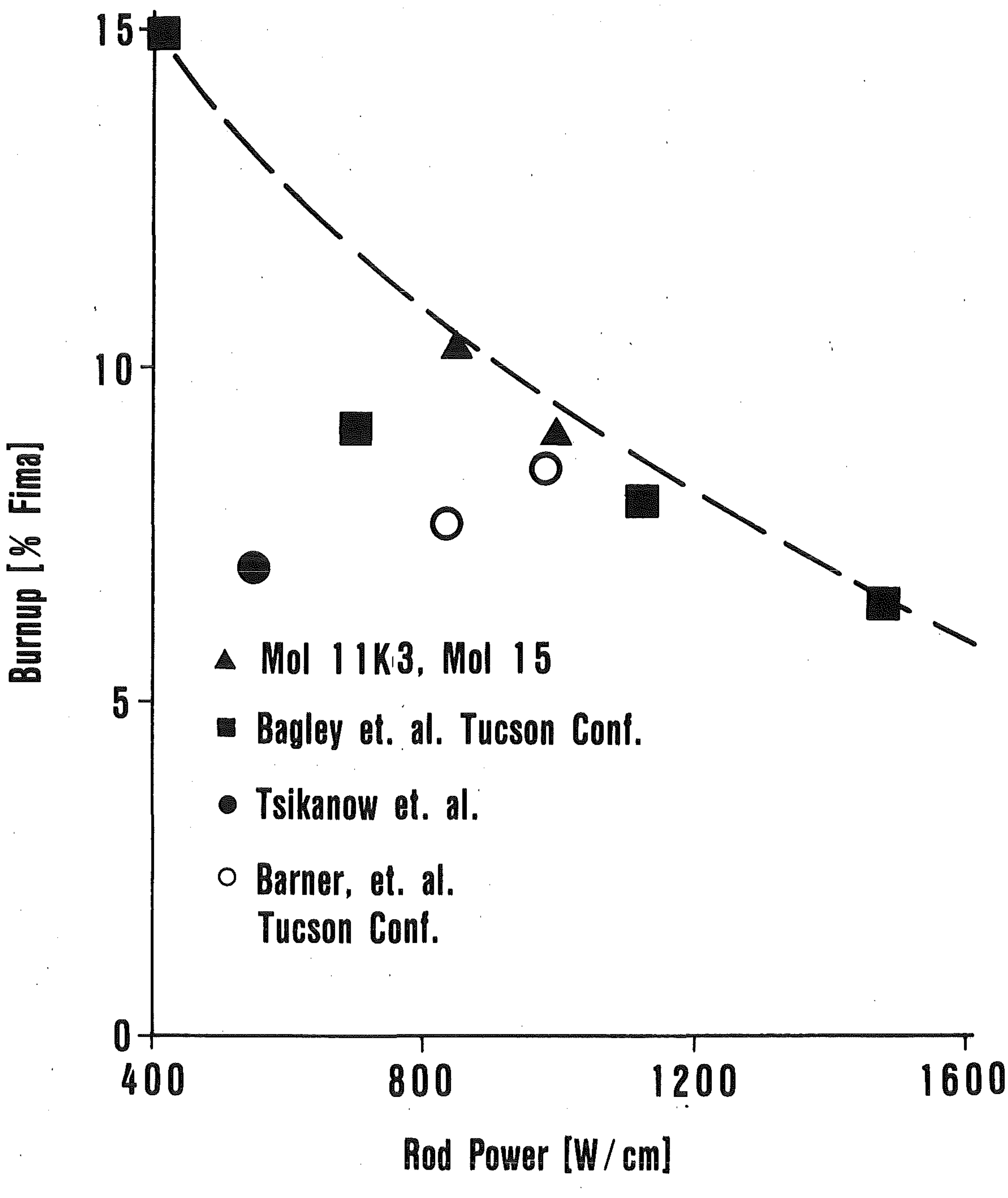


Fig. 3 Burnup of Na-bonded fuel pins versus rod power

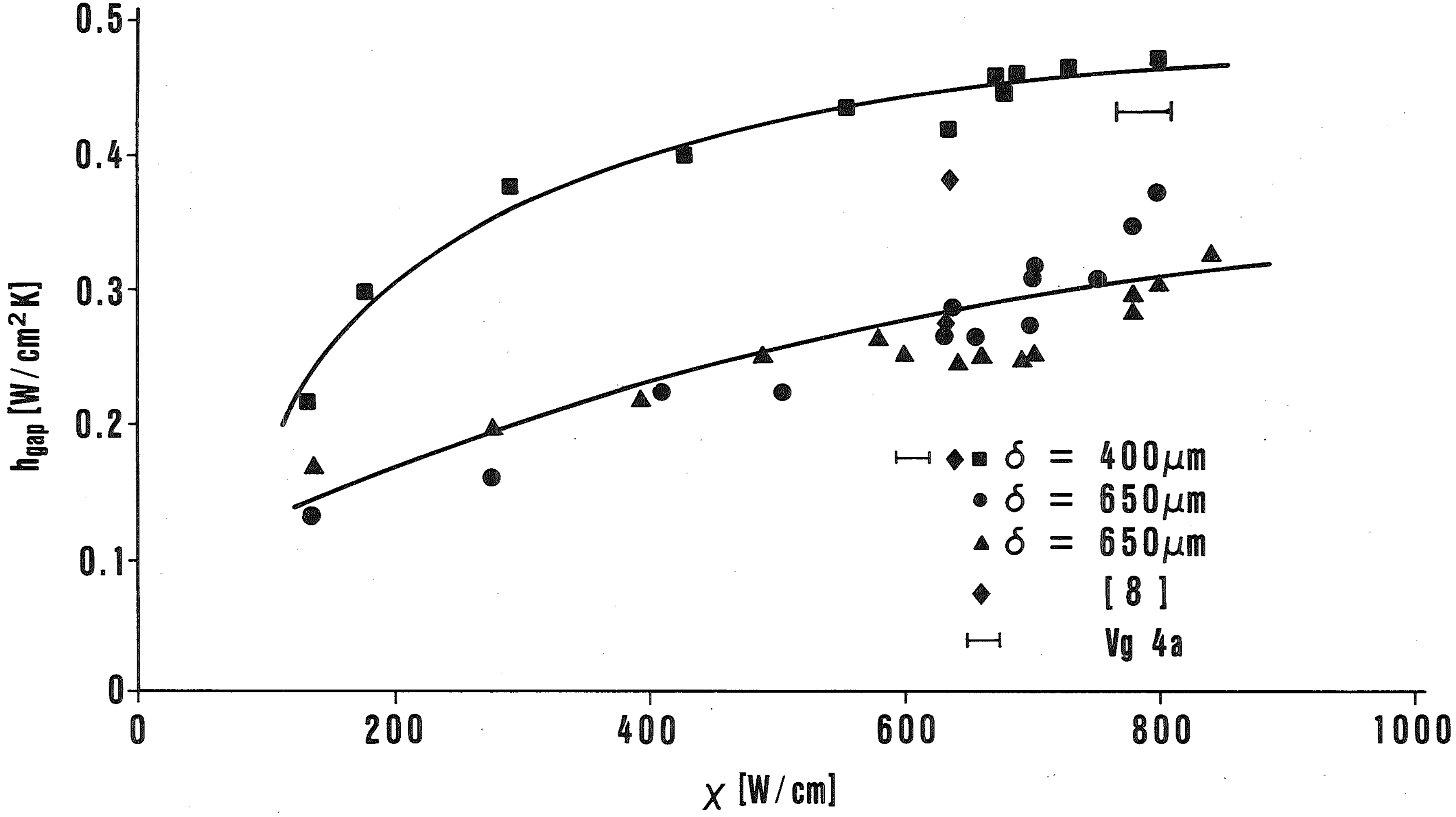


Fig. 4 Experimental heat transfer coefficients of He-bonded pins at BOL

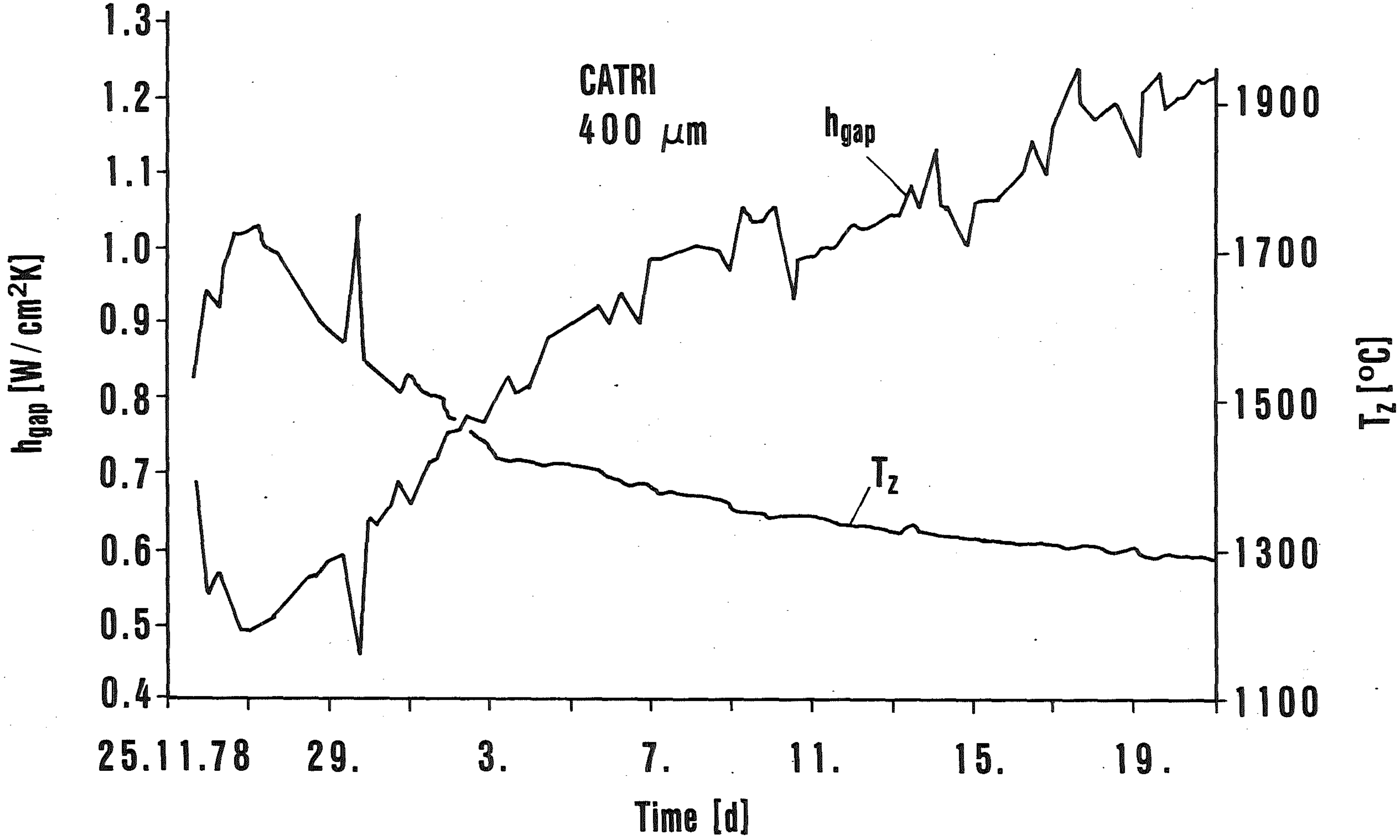


Fig. 5 Fuel centerline temperatures and gap coefficients at BOL of CATRI

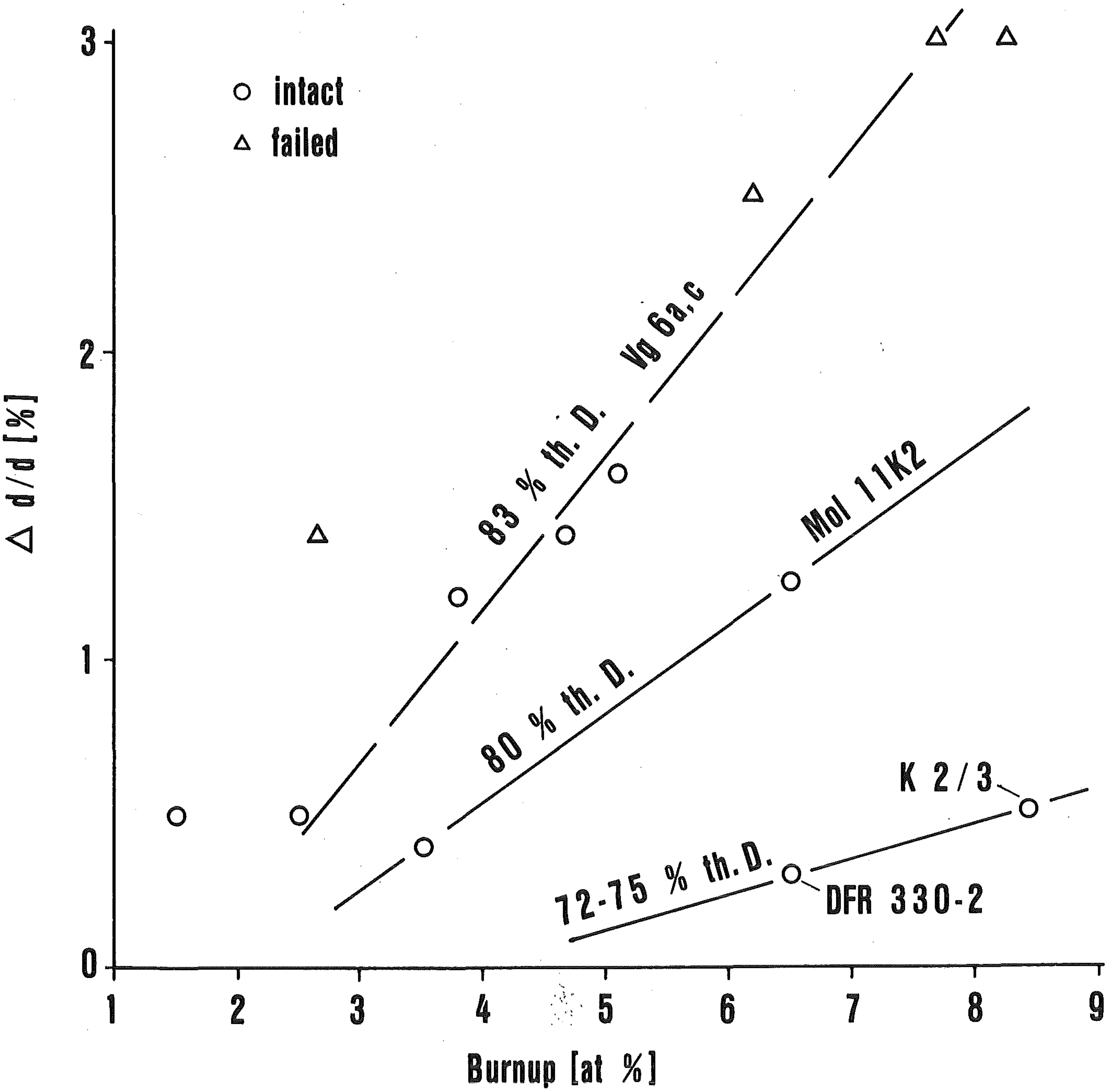


Fig. 6 Mechanical strain of gas-bonded carbide fuel pins versus burnup

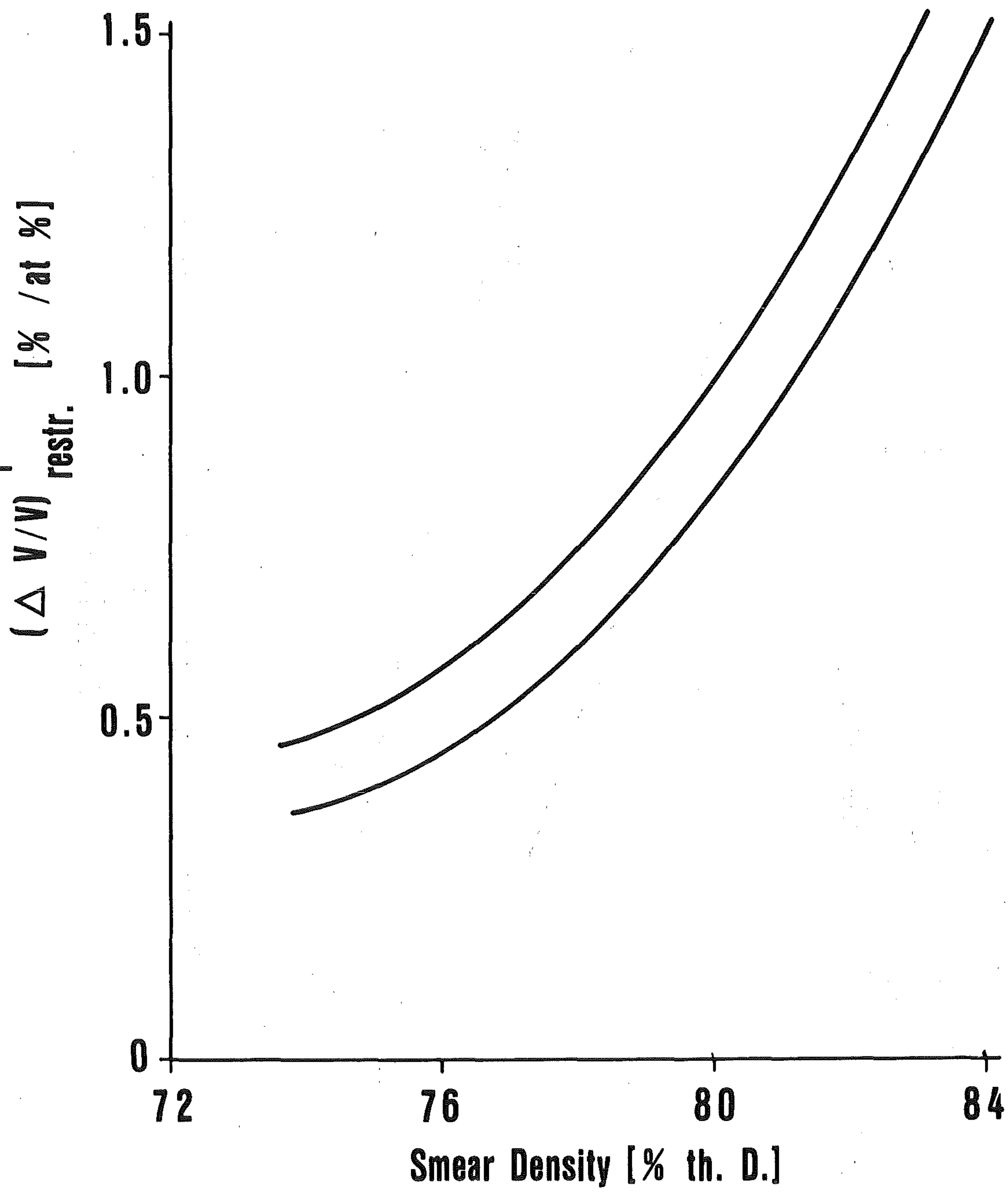


Fig. 7   Restraint carbide swelling rates versus smear density

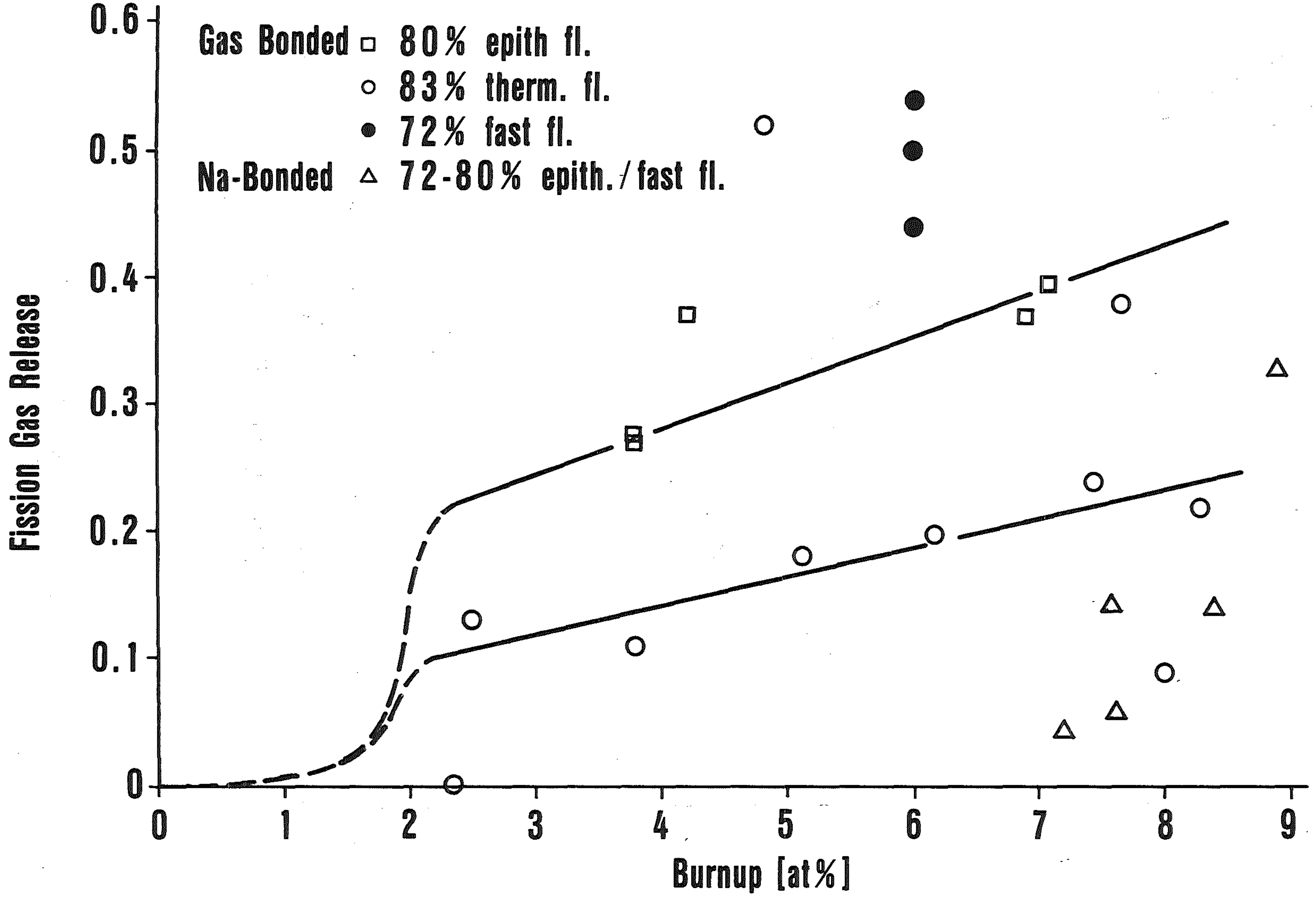


Fig. 8 Fission gas release in carbide fuel pins versus burnup

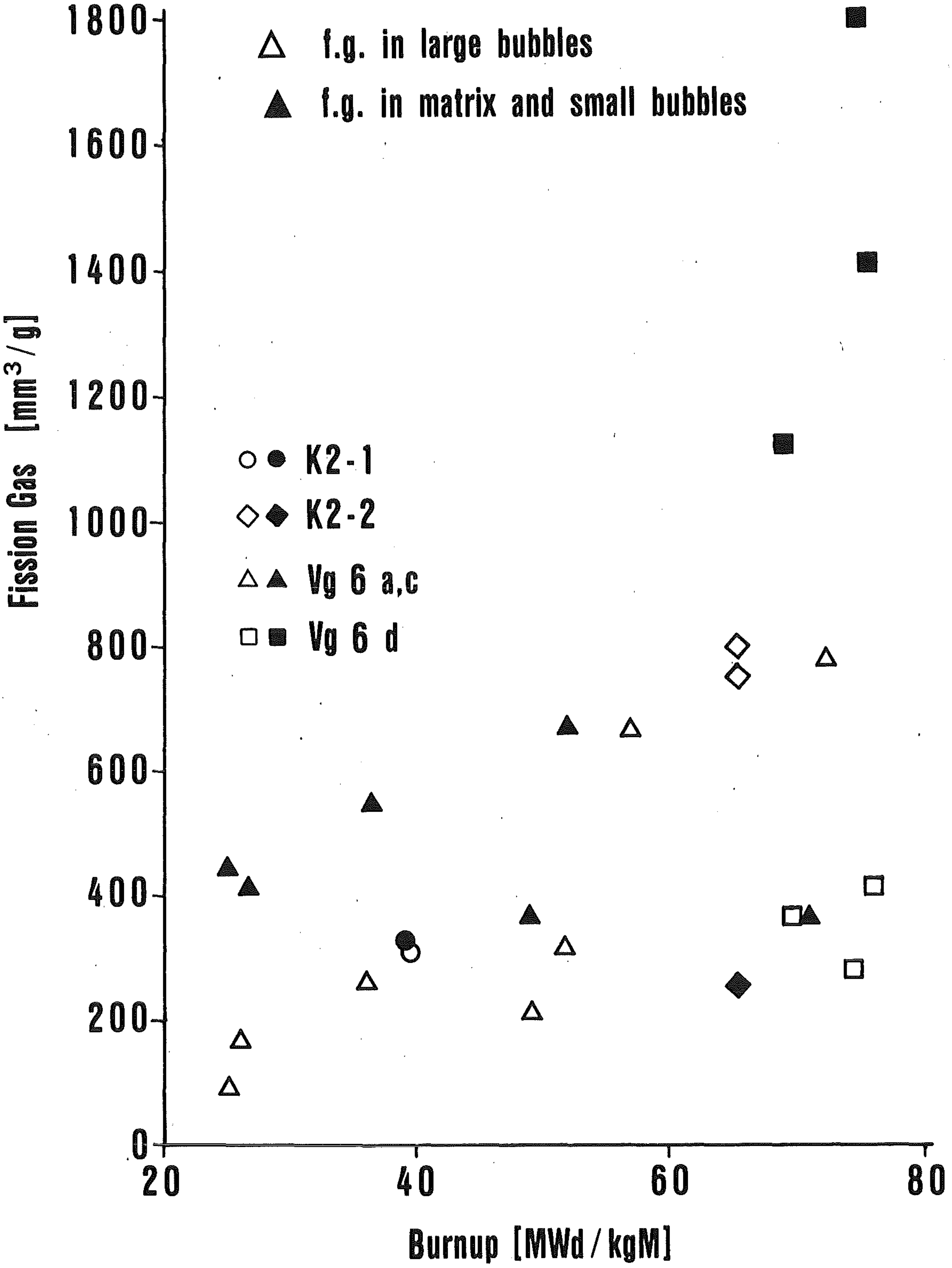
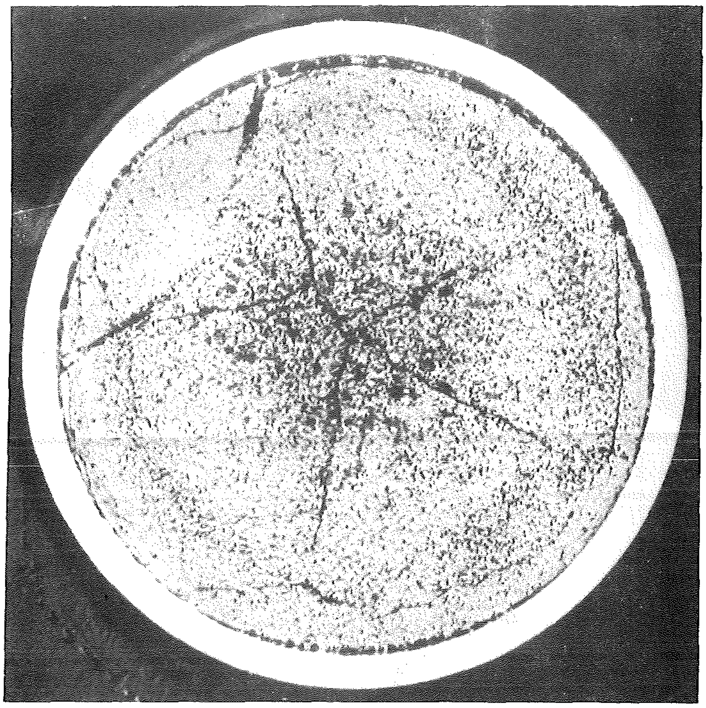


Fig. 9 Fission gas stored in fuel versus burnup



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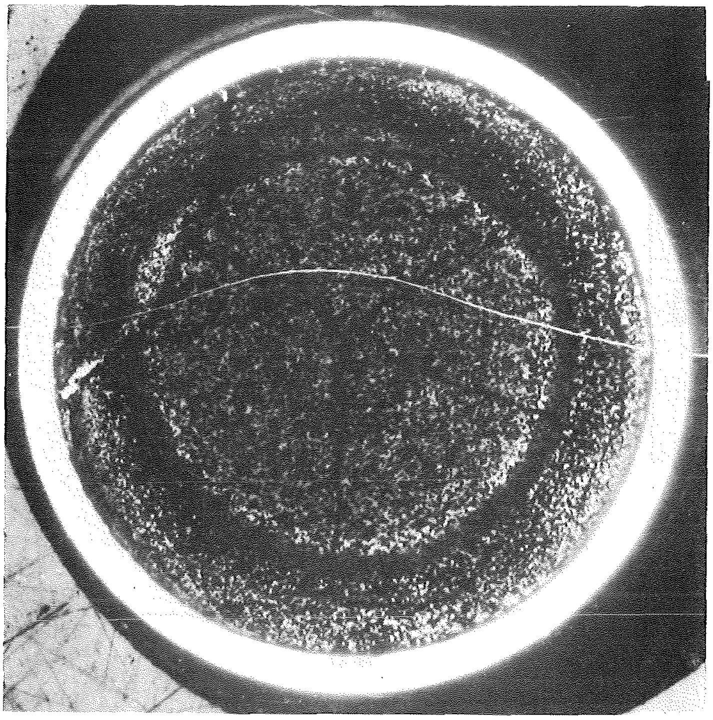

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Fig. 10 Crack healing in a He-bonded pin
(Exp. DFR-330/2, Pin V1)

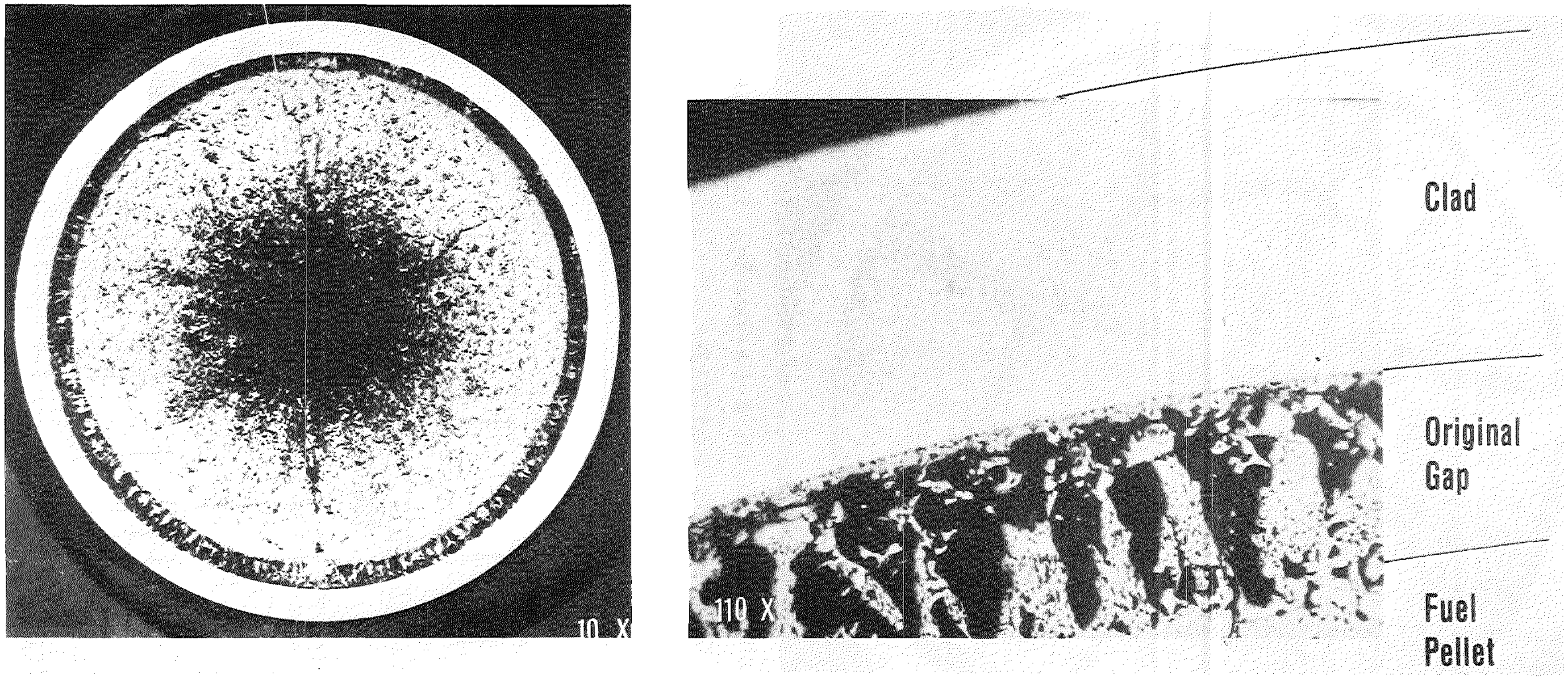

Fig. 11 Formation of a grid-like structure in a large fuel/clad gap (Exp. DFR-330/2, Pin U4)

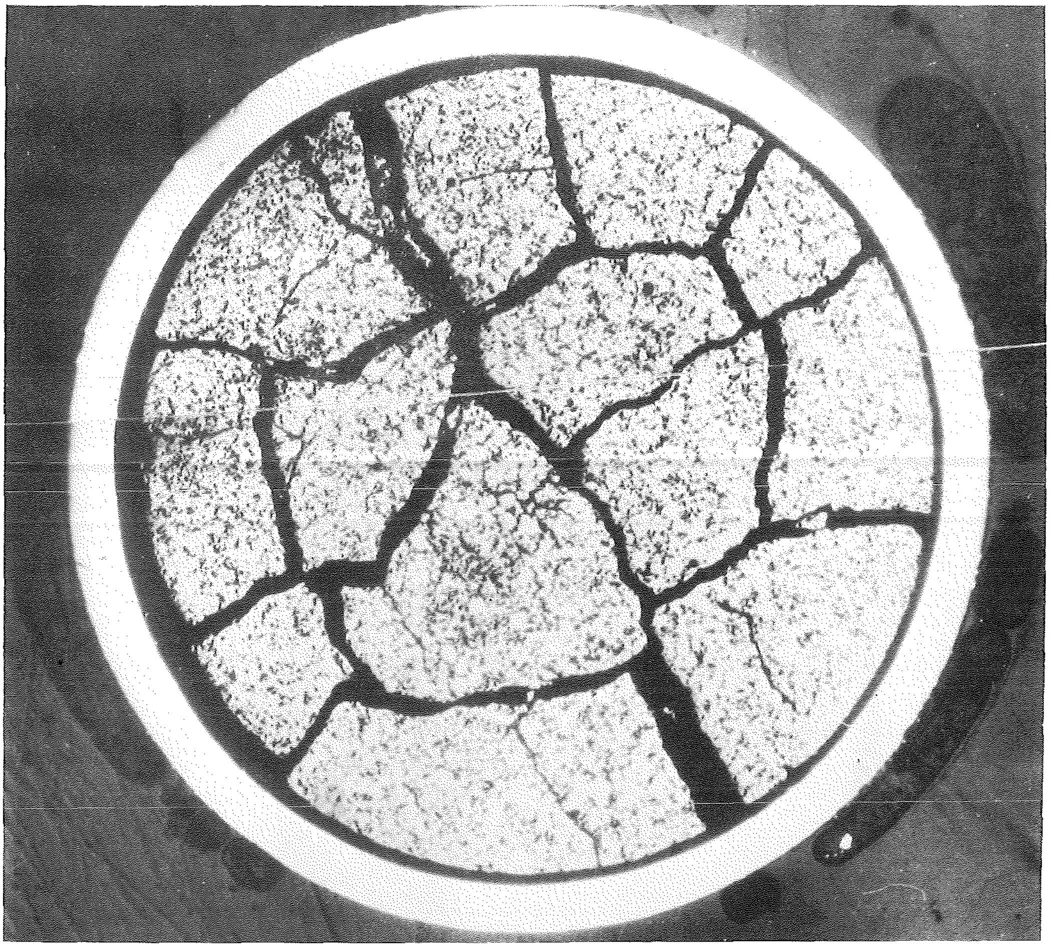

Fig. 12 Cracking pattern in a Na-bonded fuel pin (Exp. DFR-330/3, Pin Y1)

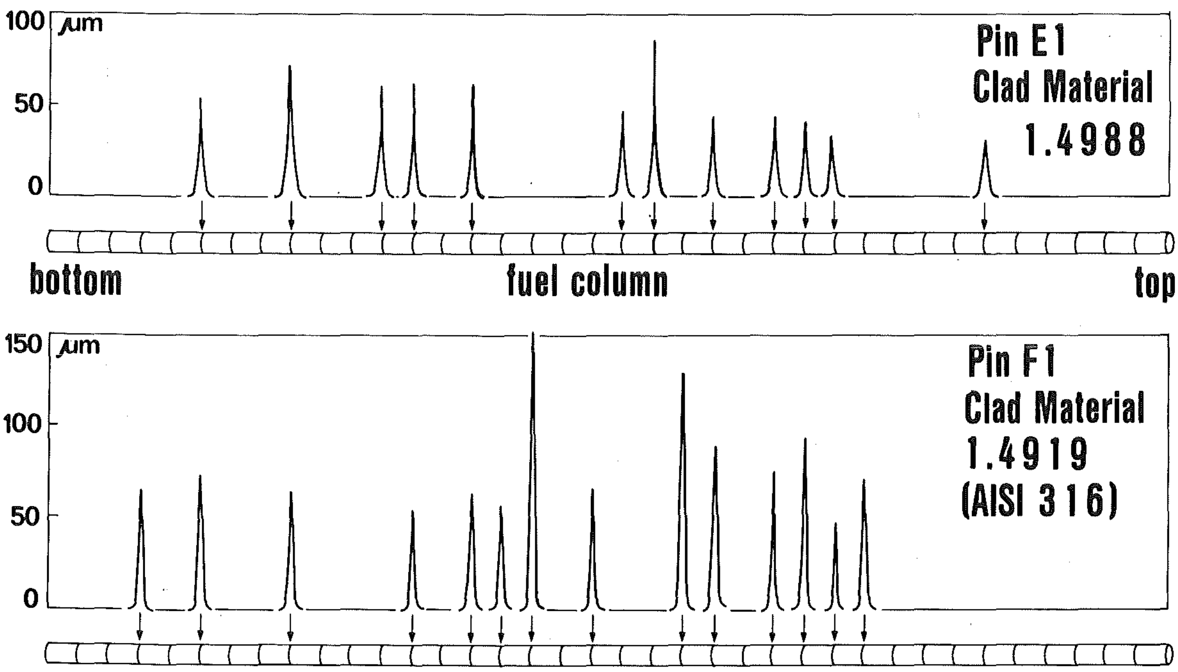

Fig. 13 Local strain peaks in Na-bonded fuel pins (Exp. DFR-330/1, Pin E1 and F1)