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Verstärkung des Porenschwellens durch einen Temperaturgradienten

Zusammenfassung

Der Temperaturgradient in der Hüllwand eines Brennstabs von Schnellen Brutreaktoren führt zu einer Vergrößerung der Hüllaufweitung verglichen mit Materialbestrahlungen.

Untersuchungen an einem dafür speziell gestalteten Brennstab führten zu dem Schluß, daß die Ursache hierzu verstärktes Porenschwellen ist. Dieses wird durch Heliumbläschen herbei geführt, welche im Temperaturgradienten wandern und durch Koaleszenz wachsen können. Somit wird der kritische Porenkeimradius schneller erreicht, als ohne Gradienten. Folglich ergibt sich hier eine größere Hüllaufweitung als man aus Materialbestrahlungen erwarten konnte, beim vorliegenden Material (DIN 1.4981 lg) um etwa 50%.

Compact

The temperature gradient in the cladding of a Fast Reactor fuel pin leads to increased dilatation compared to material irradiations. Investigations of a specially designed fuel pin reached the conclusion that the cause is enhanced volume swelling. It is induced by He-bubbles, which migrate upwards the temperature gradient and coalesce. The critical size of nuclei for void swelling is thus reached much faster. Consequently, the p in deformation is larger than expected from materials irradiations, in the present case (DIN 1.4981 sa) by about 50%.

INTRODUCTION

In the early stages of investigating the deformation of Fast Reactor fuel pins it was already recognized that volume swelling and in-pile creep alone could not completely explain the observed radial dilatations. Further mechanisms, like thermal ratcheting and mechanical interactions between fuel and the cladding tube, had to be invoked. This aspect was discussed f.e. in /1/; the authors proposed a further mechanism, namely material transport induced by the considerable thermal gradient in the cladding. This mechanism was rejected by other authors who contended that the effect would be too small to have any macroscopic consequences /2/. An experiment was, therefore, devised to look into the effects which a large radial temperature gradient has upon the cladding dilation in a Fast Reactor.

EXPERIMENTAL

A specifically designed double-walled pin (KNK 1.32) was manufactured to replace the center pin of a standard-II fuel element of the Karlsruhe reactor KNK. A particular construction served to exclude all extraneous agents from the experimental cladding tube save, off course, the neutrons and the thermal gradient, and to establish axial positions in close neighborhood with, resp. without a thermal gradient /3/. To this end the fuel of an original KNK I pin (6.0 mmø, DIN1.4981 sa) was, over some axial positions, substituted by breeding pellets; since these segments would not produce fission energy no temperature gradient would evolve along these axial positions. Thus cladding sections with and without a temperature gradient would alternate in close proximity. The most important particular, however, was the use of a second outer cladding tube of original KNK II dimensions and material (7.6mmø, DIN1.4970 cw + a) as the actual experiment. The gap between the inner and the outer cladding, filled with reactor sodium, separated the latter from any mechanical influences by the fuel and/or the inner gas pressure. Indentations in the outer tube, 3 each on six levels and 150mm apart, kept the inner pin aligned. A half-schematic view of this double-walled pin is shown in Fig. 1 (modified from /4/). The pin was incorporated into the center position of a standard KNK II/2 element (KNK-BE NZ 301) and irradiated for 574EFPD's, equivalent to a maximum dose of 35 dpa[NRT]. The axial dose distribution and the temperature of both the inner and the outer cladding, taken from the technical report /4/, are shown in Fig.2.

After taking down the fuel element the diameter of both the inner and the outer cladding were determined. The results, measured and averaged over 12 angular positions, are shown in Fig. 3. A few features of this figure have to be explained in order to stress the essential results. Large wear marks are visible on the outer clad at the position of the spacer grids (lower curve); they are caused by vibrations in the core. Also, the indentations on the outer cladding, which served as spacers for the inner pin, are apparent; they in turn effected wear marks on the inner cladding. Consequently, these marks were utilized to check the actual axial alignment between outer clad and the inner pin during the irradiation. If these experimental artefacts are ignored then a complex picture transpires. The outer clad, fabricated from the low swelling DIN 1.4970 cw + a, shows only a small diameter increase, starting at the low temperature end. This observation is easily understood since this material has its swelling maximum in the temperature region from 400 to 450 °C /5/,/6/. So despite the fact that the dose increases to its maximum going down in axial position to 600 mm the diameter increase diminishes because of this temperature dependence. No other processes have to be invoked to explain the experimentally observed curve; particularly, there is no indication that the temperature gradient had a measurable effect. This finding contrasts with the results for the inner pin. The dilatometric curve shows definite reductions at exactly the axial positions where no temperature gradient evolves; in other words, the temperature gradient increases the dilatation of the clad.

Before possible mechanisms for this behavior of the inner cladding tube are discussed other, not material related causes, have to be ruled out. The most obvious explanation, which could be brought forward to account for the asymmetric deformation of the inner pin, is an interaction between the fuel/breeder pellets and the clad. However, from numerous investigations of KNK pins and from a close inspection of this particular one /7/ it could safely be concluded that a mechanical interaction between the fuel/breeder pellets and the cladding was highly unlikely in this case; f.e. such deformations are more localized than the "valleys" found here and no strong ovalisation was observed. Furthermore, the ceramographic examinations of selected sections gave no indication of an interaction either /7/. Therefore, it has to be concluded that interactions between cladding and fuel/breeder pellets play no role in this irradiation. The same applies to a possible influence of the inner gas pressure which, off course, would not be affected by a temperature gradient. Therefore, the cause for the observed response of the cladding has to be found within the material itself.

Six sections, each 25 mm long, were cut for immersion density measurements on selected positions; in addition, two of these were further analyzed for volume swelling and microstructure by transmission electron microscopy (TEM). These density values were plotted vs. the axial position in Fig. 4 together with the results from the diameter measurements; the latter were calculated under the assumption that the increase in pin diameter is entirely due to volume swelling ($3 \times \Delta D/D_0 = \Delta V/V_0$). It can easily be seen that the results obtained by the various methods are in good agreement. This observation leads to the conclusion that the deformations measured on the inner pin can safely be interpreted as arising from volume swelling. A material transport, brought about by the large thermal gradient, does not contribute in a noticeable manner to the pin dilatations, contrary to the assumptions in /1/. Consequently, other processes in the material have to be established to explain the effect which is indeed linked to the temperature gradient.

The axial positions of the two cladding sections used for TEM investigations were selected such that one (Sample 1) was irradiated without, the other one (Sample 2) with a gradient at otherwise very similar temperatures and neutron flux conditions. In addition, one TEM disk of Sample 2 was prepared in a way that the investigated volume was close to the outside of the pin (Sample 2a) while the other one represented the inner part (Sample 2b). The void size distribution was determined for all three specimens and is displayed in Figs. 5 - 7; to judge the quality of statistics the total number of voids counted, the center of distribution, the volume increase and the void concentrations are specified in each figure.

The volume swelling of the three specimens differs quite appreciably in spite of virtually identical irradiation conditions; this can safely be stated regardless of the scatter in the data. The void size distribution in the specimen without a temperature gradient is bell-shaped with the center located at about 64 nm. This contrasts with a noticeable tailing toward larger void sizes for the specimens with a temperature gradient. The main differences between the two sections, however, are a decrease in the mean void diameter and a noticeable increase in the void concentration for the gradient affected specimens, which in combination results in an amount of void swelling larger by about 50% compared to Sample 1.

A strong clue as to the cause for the observed increase in volume swelling through a temperature gradient can be found in the difference between the two affected samples (Specimen 2a, resp. Specimen 2b): The void concentration increases by going up the temperature gradient while the mean void size decreases from about 59 to 49 nm. The TEM investigations revealed another important aspect: While He-bubbles are practically absent in the "normal" sample numerous ones were found in Sample 2b, the inner part of the cladding with a gradient. Their diameter is less than 5 nm; these bubbles are mainly concentrated in the grain boundaries but are also found on dislocations inside the grains. No He-bubbles could be observed in the outer slice of this section (Sample 2a). The other findings as f.e. to dislocations, precipitates and denuded zones do not deviate in any way from former observations on neutron irradiated material samples /8/.

DISCUSSION

Volume swelling under neutron irradiation occurs when nuclei collect enough He to reach a critical size and then grow by further accumulation of vacancies. If either or both of the two processes - nucleation or growth - is speed up, f.e. by a temperature or a stress gradient, enhanced volume swelling results. The observation of a large number of He-bubbles in Sample 2a leads to the conclusion that an accelerated nucleation is the cause for the enhanced swelling. It has been extensively discussed in the literature (see f.e. /9/ and /10/) that He-bubbles migrate up-wards a temperature gradient. This has the consequence that in a pin irradiation He-bubbles, which naturally are very small in the beginning, migrate under the influence of the thermal gradient towards the inner side. During their migration they have a chance to grow more rapidly by coalescence /10/. Consequently, they will arrive at a critical size much earlier than under "normal" conditions. This deduction is born out by the void concentrations cited on the figures. They increase, going from the outer to the inner slice, by roughly one third and between the normal specimen and the inner slice by as much as a factor of two. The mean void diameter decreases at the same time by approx. 15 nm. The latter effect is understandable since too many nuclei compete about the available vacancies. Still, despite the opposite trends of void size and void concentration the net effect is an increase of volume swelling by a thermal gradient.

From the present data it cannot be decided whether or not the influence of a thermal gradient affects the alloy DIN 1.4970 in a similar fashion. One reason for the absence of a noticeable effect in this experiment might be the high swell-resistance in comparision to DIN 1.4981 /5/,/6/ which is demonstrated even in this experiment. However, information from both our own studies as well as from the literature suggest that the drift of the small He-bubbles might be suppressed in DIN 1.4970. Small He-bubbles have been observed to be trapped on the dislocations which are stabilized by the numerous TiC-particles; an example is shown in Fig. 8 /11/,/12/. Other authors have reported similar results (See f.e. /13/ for further references). Under this circumstances there would be no drift of He-bubbles and, consequently, no accelerated coalescence into nuclei for void formation. On the other side, a temperature gradient effect has been observed after a much higher dose in DIN1.4970 /14/ similarly to results on AISI 316Ti /15/; however, no post-irradiation investigations were performed. One could speculate that the trapping of the He-bubbles is overcome after a mucher larger dose. This question has to be left open for the present.

CONCLUSIONS

The temperature gradient in the cladding of a fuel pin fabricated from DIN1.4981 results in additional volume swelling because He-bubbles migrate under the influence of this gradient and have, therefore, a greater chance to combine into larger ones. Thus they reach the critical size to commence void swelling much earlier than without a thermal gradient. The net effect is an increased swelling compared to a material irradiation with otherwise identical irradiation parameters. This effect could in this project only be proven for the material DIN1.4981. It is expected that this mechanism operates in other steels as well unless, off course, the He-bubbles are trapped as might be the case for DIN 1.4970. This finding then has the consequence that materials irradiations might give too low swelling values for design purposes even if all other experimental parameters are the same.

ACKNOWLEDGEMENTS

This project was initiated and designed by our former collegue Dr. H. Venker, who finished the construction before his untimely death.

Many people from various institutions helped to get this experiment under way through construction, irradiation in KNK and afterwards; the authors appreciate their support and wish to thank them without naming them individually.

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FIGURE CAPTIONS

Fig. 1. Half-schematic cross section of the double-walled fuel pin KNK 1.32.

Fig. 2. Axial distribution of temperature and dose for KNK 1.32.

Fig. 3. Results of dilatrometric metrology for the inner and the outer pin.

Fig. 4. Dependence of volume swelling for the inner pin upon axial position, obtained from dilatrometry (closed curve), immersion density measurements and TEM

Fig. 5. Void size distribution for Sample 1, gradient free (35 dpa, appr. 480 °C).

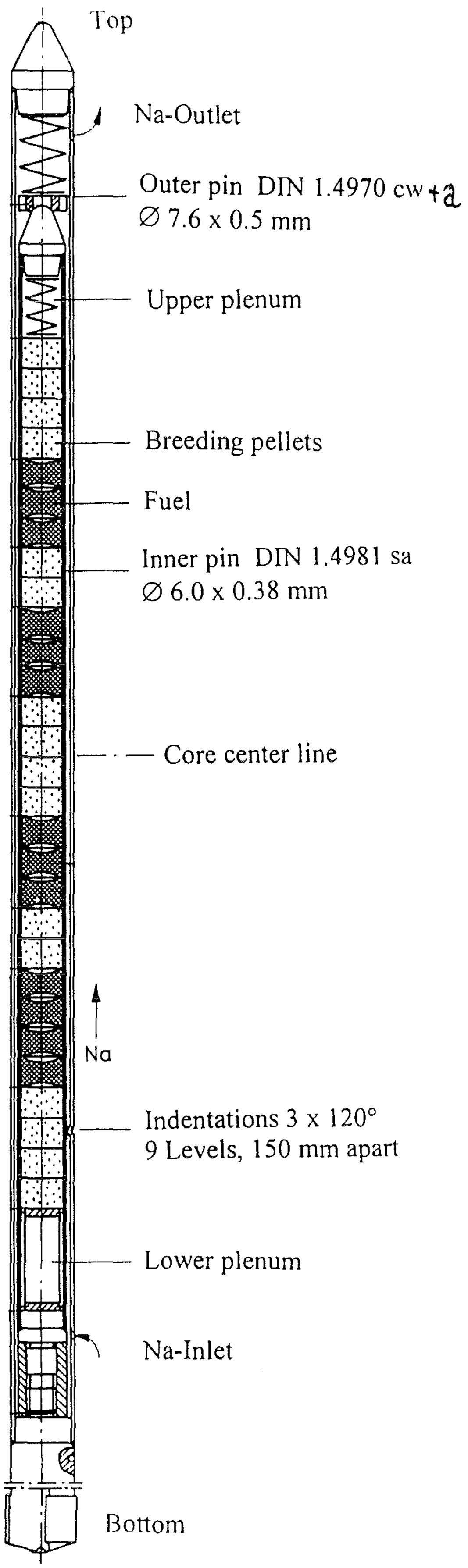
Fig. 6. Void size distribution for Sample 2a, outer section (35 dpa, appr. 480 °C).

Fig. 7. Void size distribution for Sample 2b, inner section (35 dpa, appr. 480 °C).

Fig. 8. He-bubbles appearing in the inner part of sample 2 (sample 2b)

Fig. 9. No He-bubbles in the outer part of sample 2 (sample 2b)

Fig. 10. He-bubbles trapped by dislocations in the vicinity of a TiC-particle in DIN 1.4970, irradiated at 600°C to about 30 dpa /11/.



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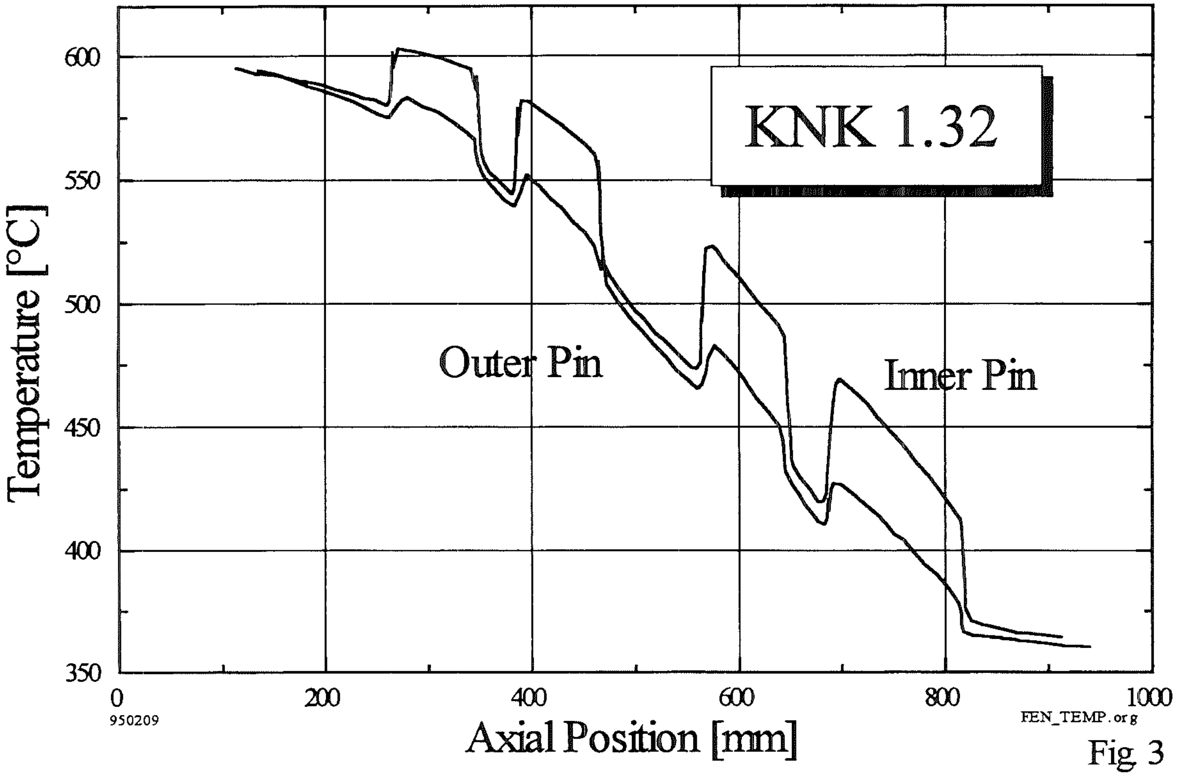

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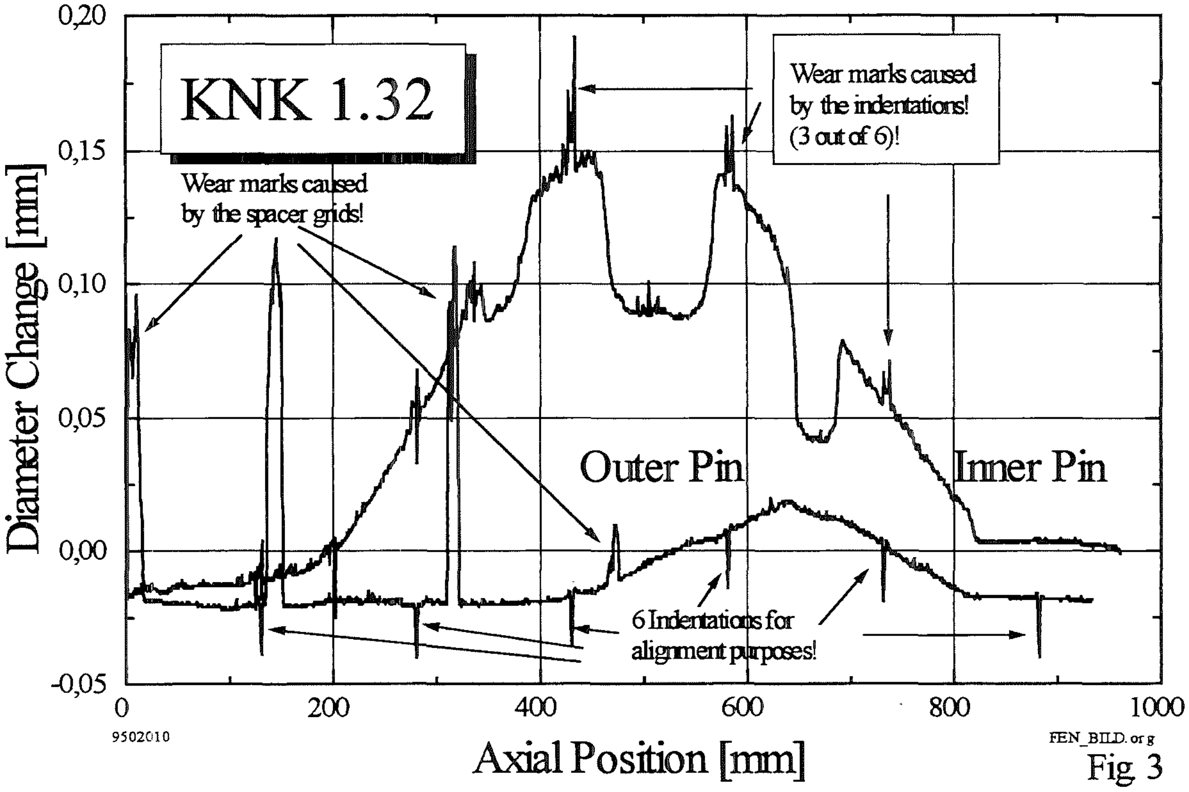

Fig. 3. Results of dilatrometric metrology for the inner and the outer pin.

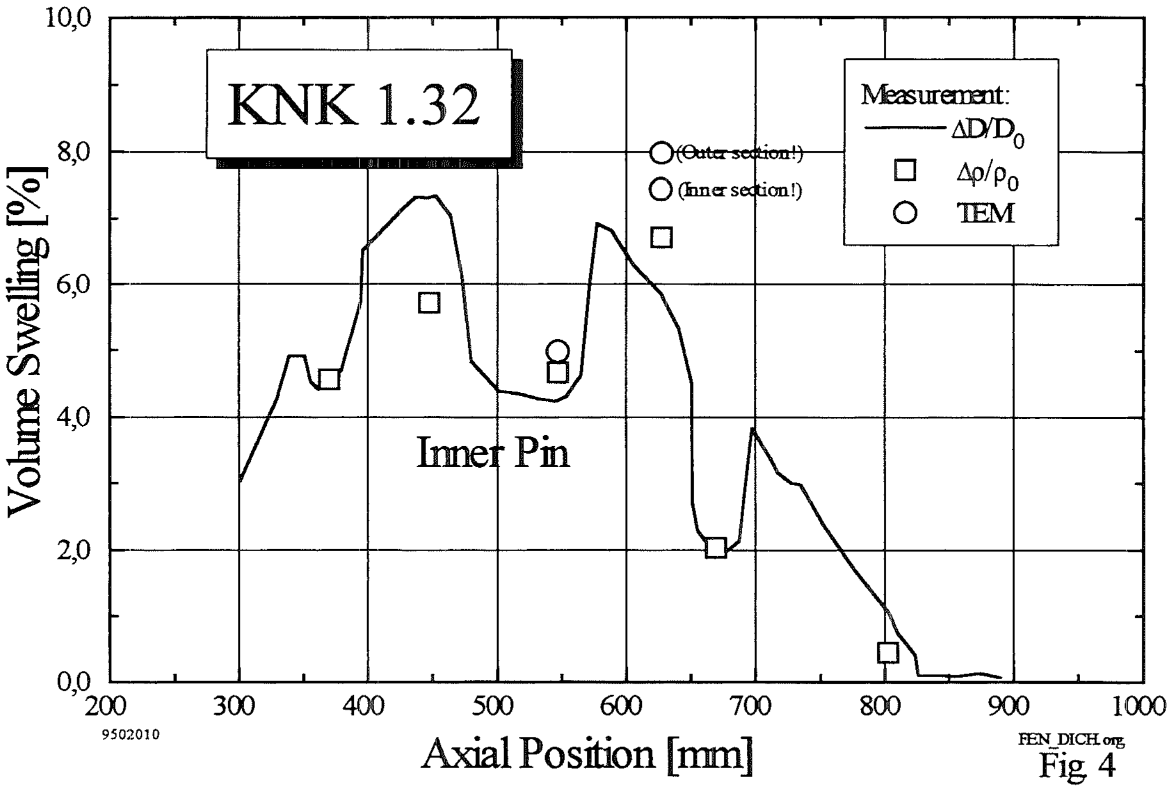

Fig. 4. Dependence of volume swelling for the inner pin upon axial position, obtained from dilatrometry (closed curve), immersion density measurements and TEM

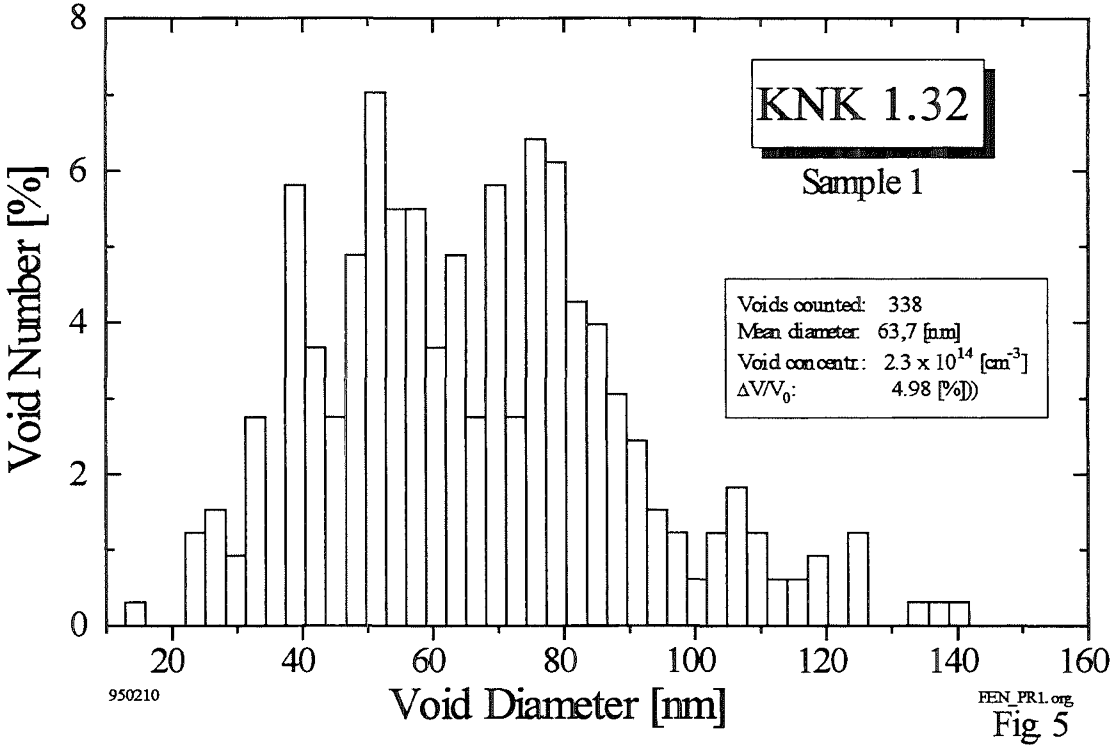

Fig. 5. Void size distribution for Sample 1, gradient free (35 dpa, appr. 480 °C).

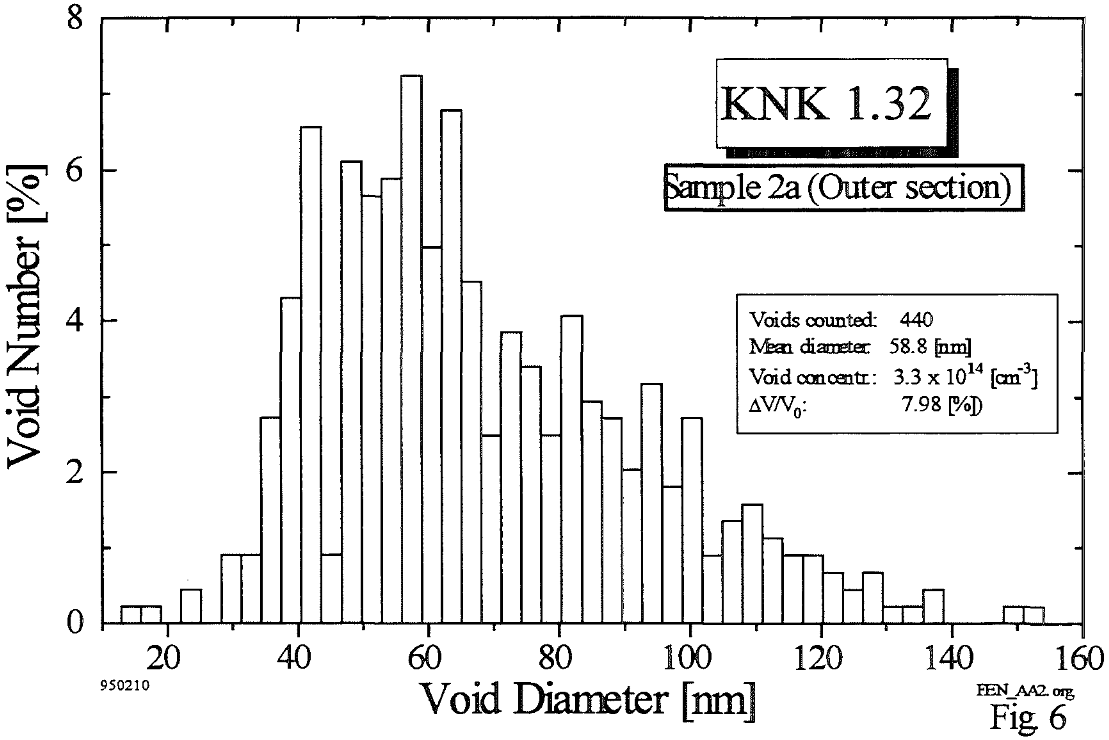

Fig. 6. Void size distribution for Sample 2a, outer section (35 dpa, appr. 480 °C).

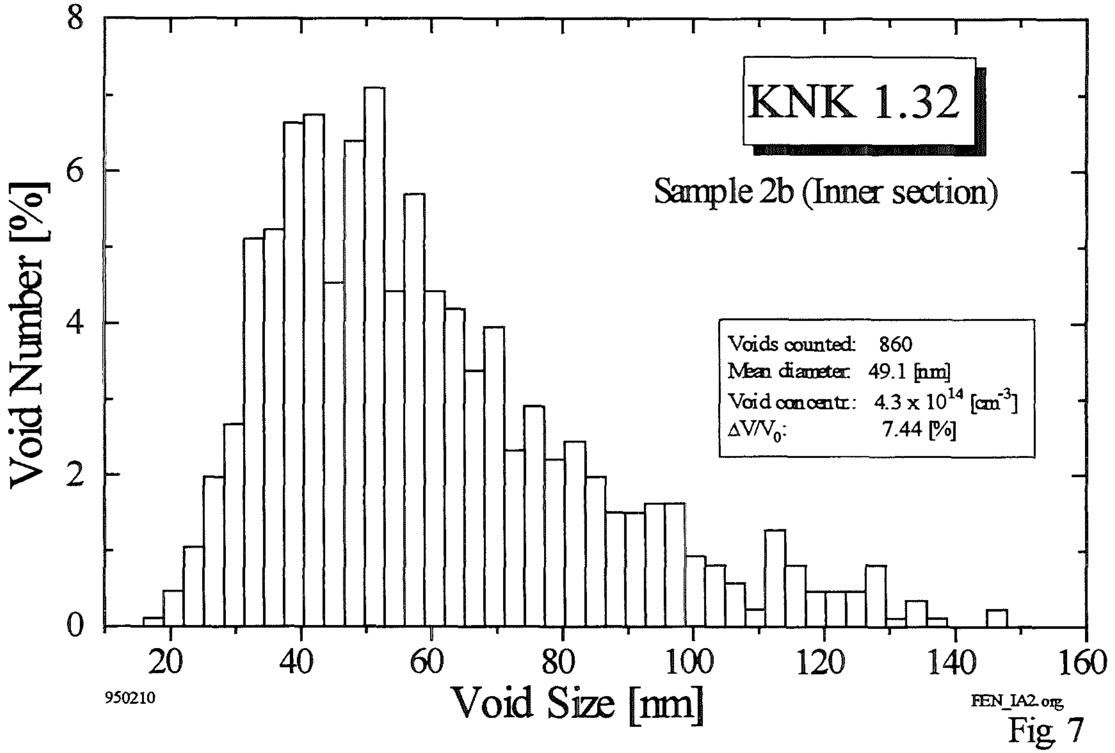

Fig. 7. Void size distribution for Sample 2b, inner section (35 dpa, appr. 480 °C).

Fig. 8. He- bubbles appearing in the inner part of Sample 2 (Sample 2b).

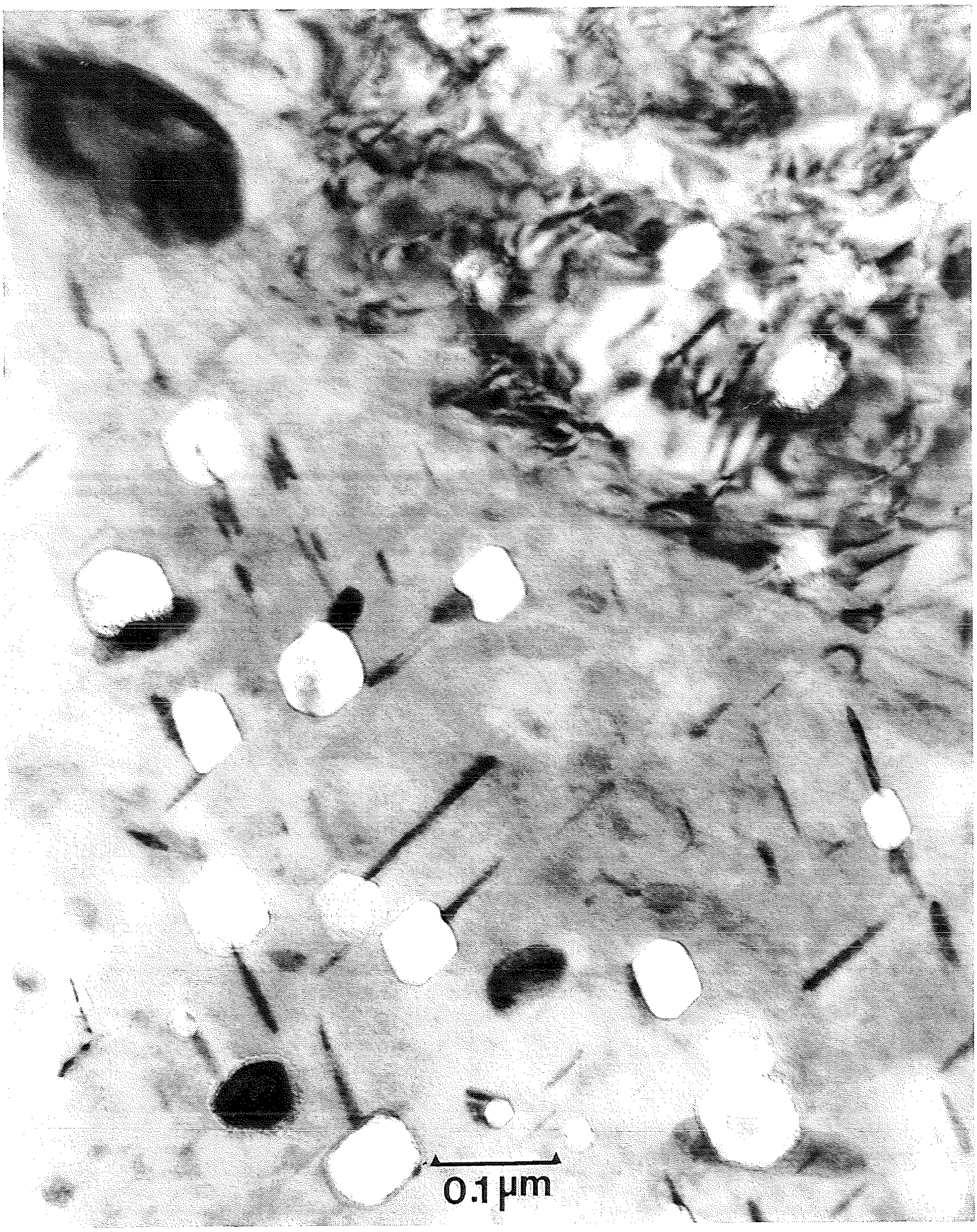

Fig. 9. No He-bubbles found in the outer part of Sample 2 (Sample 2a).

Fig. 10. He-bubbles trapped by dislocations in the vicinity of a TiC-particle in DIN 1.4970, irradiated at 600°C to about 30 dpa /11/.