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EFFECT OF THERMAL NEUTRON IRRADIATION ON MECHANICAL PROPERTIES OF ALLOYS FOR HTR CORE APPLICATIONS

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An industrial heat of Hastelloy-X containing 2.3 ppm boron was creep-tested at 900 °C after irradiating thermal neutrons by 6.6×10^{20} n/cm ^{2} at temperatures 670 to 880 °C in JMTR. Significant reduction in rupture life and ductility was observed, and large shift of accelerated deformation stage to short time side was also apparent at comparatively high stresses. Below about 2.2 kg/mm ^{2}, apparent relief from the degradation was seen. The elongation, however, was found to be due to the formation of numerous intergranular cracks in the premature stage of deformation. Based on the post irradiation tensile properties of several industrial alloys the degree of the ductility loss was found to be nearly dependent on the boron content of the alloys. The post irradiation tensile tests for a special low boron grade heat revealed the means of protecting materials from the effect to be feasible.

Keywords: Neutron Irradiation, Very High Temperature, Hastelloy-X, Creep Rupture Life, Creep Curve, Ductility Loss, Helium Embrittlement, Boron Content,

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+Division of Research Reactor Operation, Hot Lab.

HTR 炉心用耐熱合金の機械的性質におよぼす熱中性子照射の影響

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市販のハステロイ－XをJMTRで高温照射した後，900℃の大気中クリープ試験に供した。素材はCo＝0.62％，B＝2.3 ppm，結晶粒度；ASTM № 4のものである。照射条件は，温度；670～880℃，積算時間：1040 hr，照射量；6.6×10^{20} n／cm^2（thermal），1.1×10^{20}(fast)，であった。照射温度はキャプセル内の位置により異なり，その変動巾はピークの880℃で±5℃，その他は±30℃であった。照射後のクリープ試験条件は，応力；1.5～5.0 kg／mm^2，最長時間；12,000 hrとした。その結果，照射後のクリープ挙動は，応力が約2.2 kg／mm^{2}を境として，2つに区分できることが判明した。これより高応力側では照射脆化は著しく，延性と破断寿命は約1桁低下した。一方これ以下の応力では，低応力になるほど延性および破断寿命は非照射材の値に近くなる傾向が認められた。すなわち低応力の場合には，一見照射脆化がないようにみうけられる。しかし金相試験の結果，低応力のクリープ試験材では試料の平行部全面にわたって多数の粒界クラックの発生が認められたので，この全面クラックによって低応力のクリープ試験中にみかけの延性と破断寿命が保たれていると結論された。筆者らが従来行なってきた一連の高温照射後引張試験の結果から，耐熱合金中のホウ素含有量が照射後延性の支配因子であることが確認されているので，ホウ素量を1 ppmと低下したハステロイ－XRについて予備的な試験を行ない，照射後の延性低下が改善できる見通しが得られた。

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1 INTRODUCTION

Most iron- and nickel-base alloys loose ductility at elevated temperatures by thermal neutron irradiation. This embrittlement is due to intergranular cracking and is generally attributed to the helium formed primarily by the n·α type nuclear transmutation of boron (^{10}B) existing as impurity in the alloys. The reduction in ductility becomes significant as the temperatures of both irradiation and deformation at testing increase. The trend is seen to be related with the enhancement of intergranular deformation and fracture with increasing temperature.

In the high temperature reactors such as HTR and VHTR, heat resistant alloys are expected to be employed in thermal barriers as well as in the control rod cladding, where temperatures, neutron flux and stresses are in the range of concern.

2 PREVIOUS WORKS

The authors have examined post-irradiation tensile ductility of several commercial alloys including Hastelloy-X Incoloy-800, -807, Inconel-600, -617, -625 and -X-750 by tensile test with low strain rate at temperatures 800 to 1000 °C.

The main conclusions drawn are as follows; (1) ductility loss is intensified as both the temperature of irradiation and of tensile test are increased, (2) the boron content of the alloys was the primary variable in determining the ductility loss, (3) metallurgical modifications such as thermomechanical treatment and addition of carbide stabilizer etc. are effective only in limited temperature range.

The present work involves the results of creep rupture test on a commercial heat of Hastelloy-X irradiated at elevated temperatures before the creep rupture test.

Some exploratory results on the effect of reducing boron content in the alloy on the post-irradiation tensile ductility at elevated temperatures are also discussed in connection with the means of combatting the problem.

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3 EXPERIMENTAL METHODS

a) Material and specimen

A commercial heat of forged Hastelloy-X with chemical composition shown in Table 1 was employed in solution annealed condition. The heat is characterized by the comparatively low Co and B. The grain size was of approximately ASTM No.4. The geometry of the specimen machined is given in Fig. 1.

b) Reactor irradiation

The specimens were irradiated in a capsule with controlled environment and temperature by JMTR for 43 days to accumulate the total thermal neutron fluence of 6.6×10^{20} n/cm^2, and 1.1×10^{20} n/cm^2 for fast (> 1 MeV) neutrons. The temperatures during the irradiation were different depending on the position of the specimens within the capsule and ranged from 670 to 880 °C. The control was made within ±5 °C at the peak position while in other positions the fluctuation maxima were ±30 °C. Special design was needed for specimens and capsules to make the specimens free from self welding and bending due to the high temperature and the protective helium atmosphere.

c) Post irradiation tests

The creep-rupture tests were made in air environment primarily at 900 °C. The initial stresses given were in the range of 1.5 to 5.0 kg/mm^2.

The creep strain during the test was monitored by two dial gauges per each run to observe the relative movement of the specimen grips. The slow speed tensile tests run for evaluation of boron effects were made with an instron machine with vacuum chamber. The strain rate and the vacuum pressure were 0.67 %/min. and 5×10^{-6} torr. respectively.

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4 RESULTS

a) Creep deformation

i) Behavior under high stresses

Under comparatively high stresses the irradiated material showed extremely low ductility. Figure 2 illustrates a typical contrast between the behaviours of the irradiated and unirradiated material, which were obtained with the tests with the initial loading of 5 kg/mm^2 (49 MPa). In the irradiated material the fracture was found to have occured at grain boundaries, and the initiation of the cracks were estimated to have commenced at a very premature stage of deformation. The degree of ductility loss was nearly one order of magnitude in the irradiated material.

Figure 3 compares unusual relation of creep strain rates among the curves obtained from the tests with stresses at around 4 and 5 kg/mm^2 where lower stress cases showed higher creep rates. It should be noted that none of the cases showed the final accelerated creep stage and came into final fracture directly from the secondary creep stage.

In the irradiated material, shortening of the secondary creep was apparent, and this caused a difficulty in measuring the strain rate acculately. On the other hand no significant acceleration of the minimum creep rate was observed when compared with the unirradiated material. The trend observed as in Fig. 2 and 3 was common in all the results with stresses down to about 2.2 kg/mm^2 (24.5 MPa).

ii) Behavior under lower stresses

The deformation and fracture behavior under the stresses below about 2.2 kg/mm^2 was somewhat different from the higher stress cases.

Figure 4 shows some of the typical results of low stress creep deformation for irradiated materials. A common feature was observed, which was the very early transition to a sort of accelerated stage followed by a kind of recovery to a steady linear creep although the latters were slightly steep as compared to the secondary creep of the irradiated materials. The recovery of this final linear deformations was seen to have a strain levels at around 10~20 %. For the convenience the authors may nominate this stage as the "fourth one."

Under stresses around 2 to 2.2 kg/mm^2 (19.6~21.6 MPa) the tertiary creep began at about 1/3 of rupture life and attained the final linear deformation, ie. the fourth stage, at approximately 2/3 of the total rupture life.

In order to see the cause of the observed irregularity, one of the tests, ie. G-3, was interrupted at a point in the fourth stage and metallographic examination was performed.

b) Rupture life

The rupture time obtained for the irradiated and unirradiated materials are shown in Fig. 5. For unirradiated material, two series of plots are given. In these the data with 6 mm dia. specimens are taken from other series of study in which the same heat of material was tested and analysed properly for its general creep characteristics and are based on a fair data basis. Another series with 3 mm dia. are to indicate the thermal control data for direct comparison with the irradiated material. In the comparison, it is clear that the time to rupture is reduced by approximately one order of magnitude by the irradiation when the applied stress is higher than about 2.2 kg/mm^2. On the other hand the plot for the results with lower stresses looks to become close to the line of unirradiated material as the stress level is lowered.(1),(2)

The similar apparent recovery of the property with lowering the stress was also observed in the elongation at fracture. Figure 6 shows the relationship between the final extention and the time to rupture.

c) Transition to the tertiary stage

Essential degradation of a structural component is regarded as to occur when the metal deformed or cracked severely even if it is not ruptured. In this respect, the initiation of the tertiary creep is of significance as one of the measures. Figure 7 is the plot of the transition time as a function of the given stress. The parallelity between the irradiated and unirradiated materials indicates that the degradation of the material due to the irradiation can not be recovered even in the low stress cases. The cause of the apparent increase in the elongation at fracture must be viewed more critically. The consistency of this behavior with the metallographic features described below explains the phenomena.

d) Metallographic observation

The examination of the microstructure near fracture surfaces and the uniformly deformed regions revealed that the final failure occured intergranularly in all cases. In the high stress cases, where significant reduction in the fracture elongation was observed, very low degree of grain deformation all over the specimen with rather descrete cracking localized only at the fracture surface was seen.

In contrast, the irradiated specimens tested under stresses below about 2.2 kg/mm^2, where the apparent elongation at fracture was seen to be approaching to the unirradiated material, were thought to be deformed quite differently. Figure 8 shows the longitudinal cross section of the specimen G-3 (See Fig. 4) which was tested with stress of 1.5 kg/mm^2, and the test was terminated for the examination right after the creep process come into the fourth stage. An extensive occurence of intergranular premature cracks are seen all over the uniformly elongated zone. The observed effect is interpreted as the evidence of highly enhanced ductility loss accompanied by the lowering of the threshold condition for the intergranular cracking. The reappearence of the linear time dependence at the fourth stage is hypothesized as the influence of strengthening by either oxidation or nitriding at the internals of the fine intergranular cracks, which could arrest the crack growing under low stresses. The apparent elongation, therefore, should not be interpreted as essential ductility recovery under low stress conditions.

e) Role of boron in alloys

A comparative examination of several commercial alloys was made by performing vacuum tensile test with strain rate 0.67 %/min. Figure 9 compares the fracture strain after irradiation. The trend is clear that the amount of B contained in the alloys have substantial influence on the observed ductility loss. It is also noteworthy that the nickel rich alloys are generally more sensitive to the irradiation effects, while Incoloy 800, which happens to be an iron-base alloy with low boron content has shown good ductility.

As an exploratory trial, a heat of Hastelloy-X, which has a chemical composition modified for nuclear application and is named as alloy X-R in Japan, with low B content was compared with a commercial heat by a post irradiation tensile test. The results are summarized in Fig. 10. Some substantial improvement is evident for the given test conditions.

The test conditions given, however, is rather moderate, in terms of the irradiation temperature and thermal neutron flux, is 50 °C and 2.4×10^{19} n/cm^2 respectively. Further critical works are required to establish a proper means of combatting the problem.

The source of helium is not limited in $B(n,\alpha)$ reaction, and with the fluence over 10^{21} n/cm^2, the formation will be dominated by the reaction involved in $^{58}Ni(n,\gamma)\,^{59}Ni(n,\alpha)^{56}Fe$.

5 CONCLUSIONS

1. The creep-rupture properties at 900 °C of a commercial heat of Hastelloy-X with comparatively low boron content, 2.3 ppm, was influenced strongly by neutron irradiation.
2. The ductility loss observed was due to an extensive intergranular cracking and the reduction of the effective life by about one order of magnitude throughout the range of stresses applied in the tests.
3. Reduction of boron in the alloy or employment of appropreate iron-base alloys was suggested as possible means to improve the situation.

REFERENCE

1. F.C. Robertshaw et al; STP 341 (1963) P.372.
2. GEAP-177A (1963) P.75.

Table 1 Chemical composition of specimen
wt %, (B; ppm)

	C	Mn	Si	P	S	Cr	Co
Hastelloy-X	0.07	0.59	0.24	0.012	0.005	21.59	0.62

Mo	W	Fe	N	B	Ni
8.98	0.46	17.45	0.029	2.3	bal

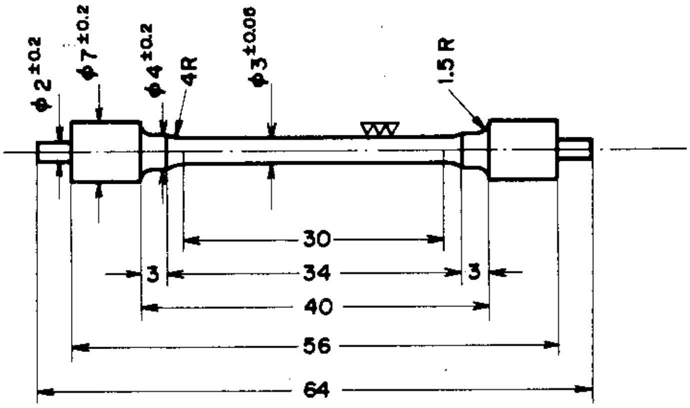

Fig.1 Geometry of specimen for post-irradiation creep test

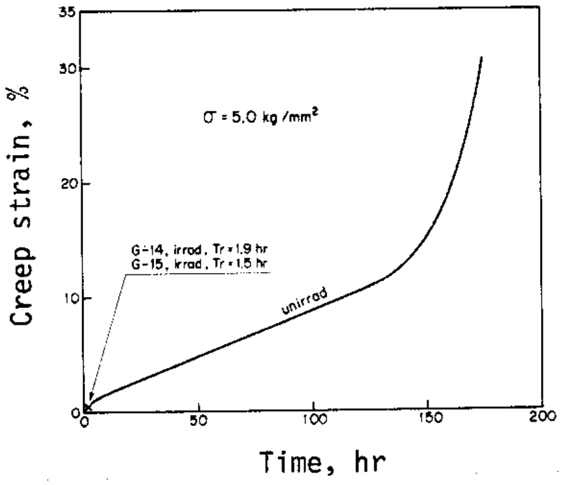

Fig.2 Creep curve of pre- and post-irradiated Hastelloy-X tested at 900°C under comparatively high stress
Tr; Time to rupture

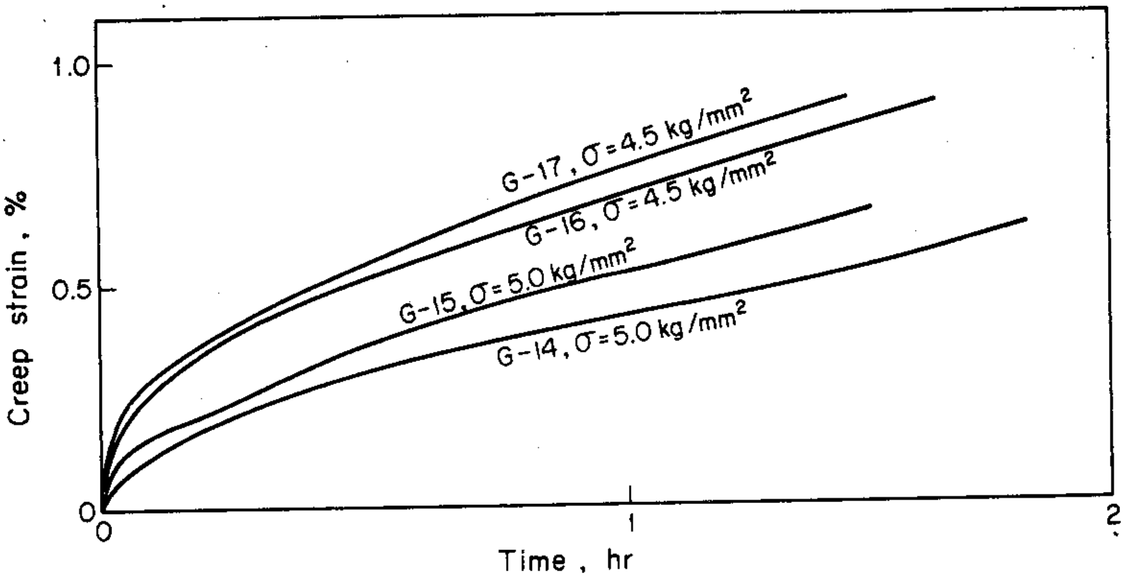

Fig.3 Creep curve of irradiated Hastelloy-X tested at 900°C under relatively high stresses
Irrad.temp; 742～790°C

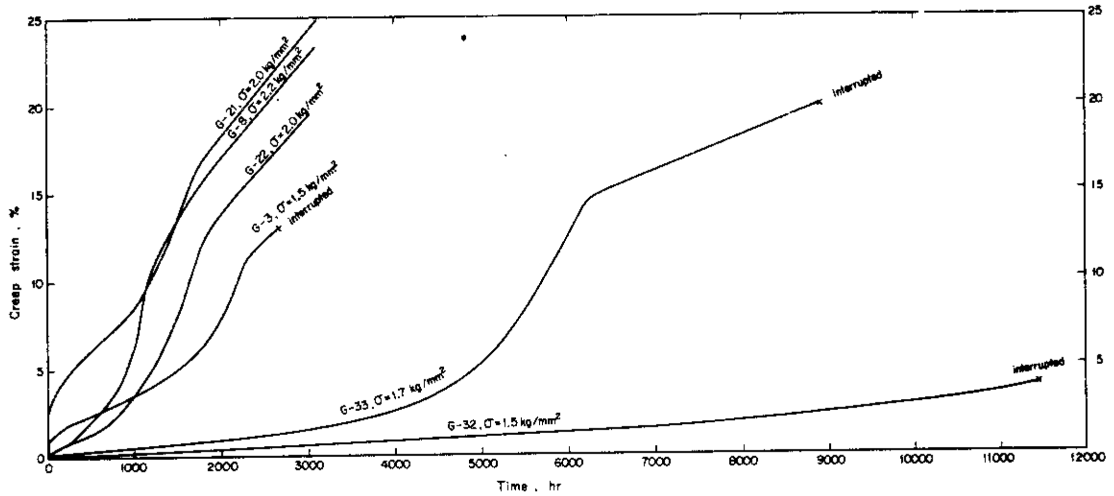

Fig.4 Creep curve of irradiated Hastelloy-X tested at 900°C under relatively low stresses
Irrad.temp; G-21,22;815～840°C, G-8;620～635°C,
G-32,33; 655～675°C.

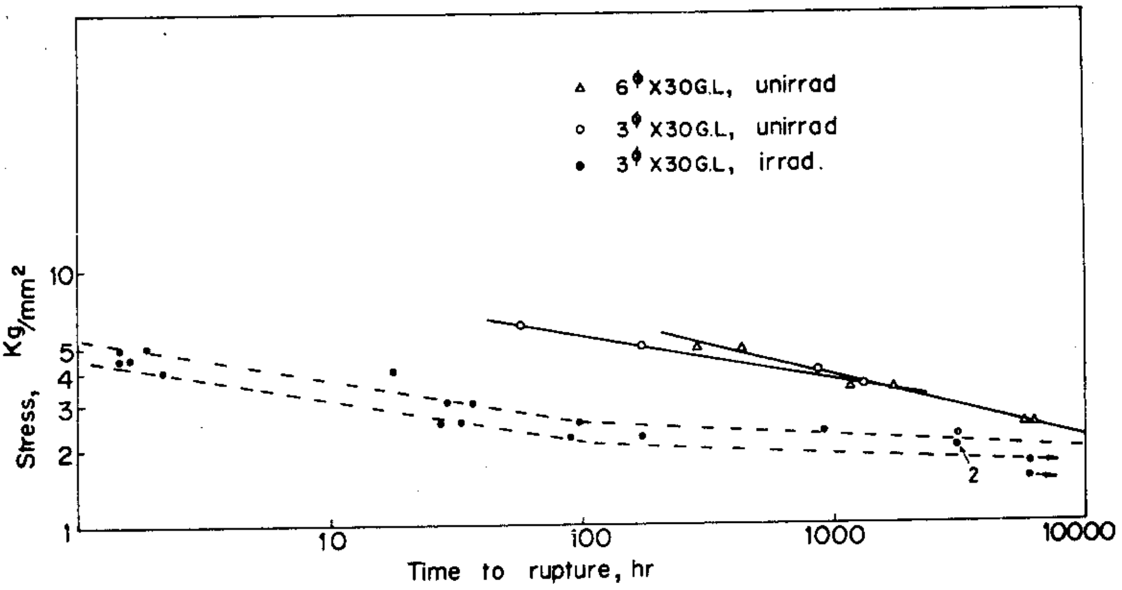

Fig.5 Stress versus rupture life of pre- and post-irradiated Hastelloy-X tested at 900°C

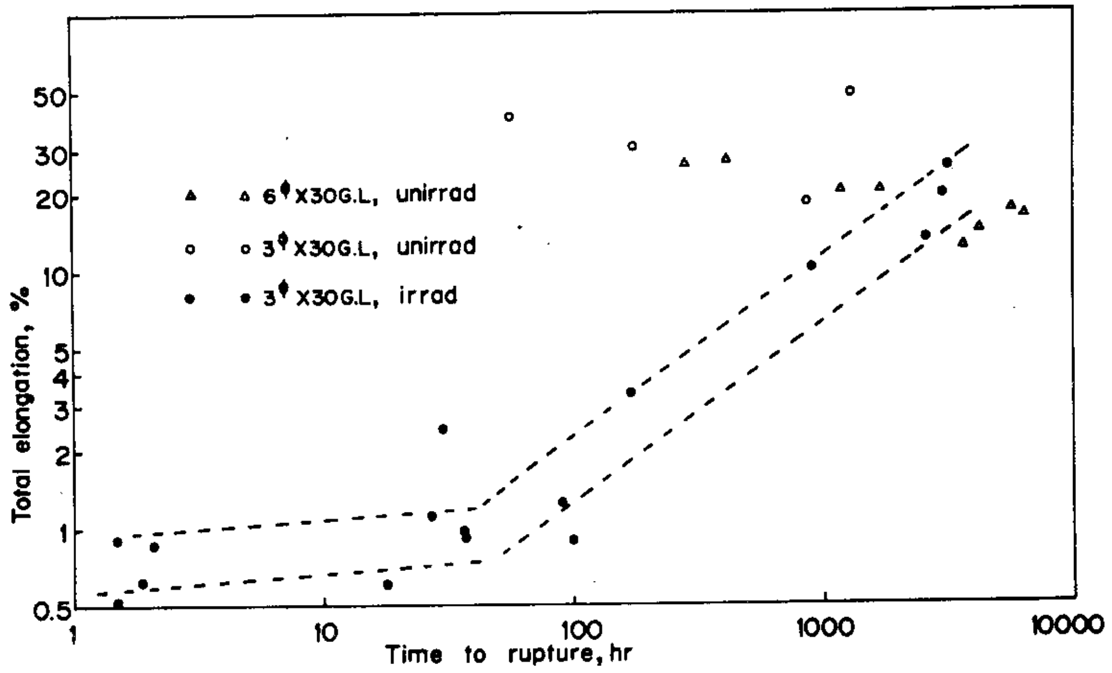

Fig.6 Relation between total elongation and time to rupture of pre- and post-irradiated Hastelloy-X creep-tested at 900°C

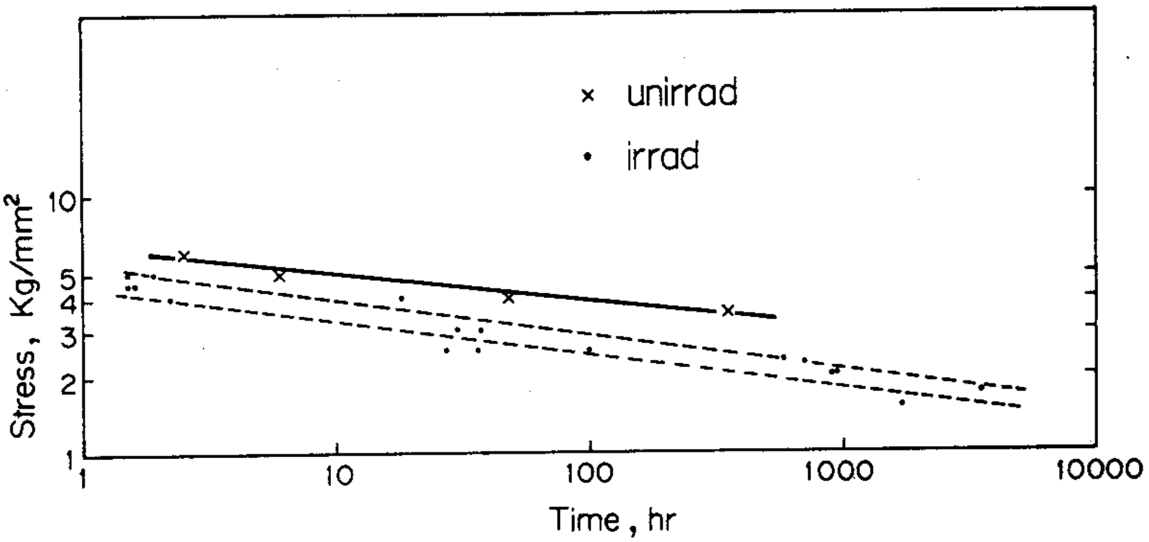

Fig.7 Stress versus time at end of secondary creep of irradiated Hastelloy-X

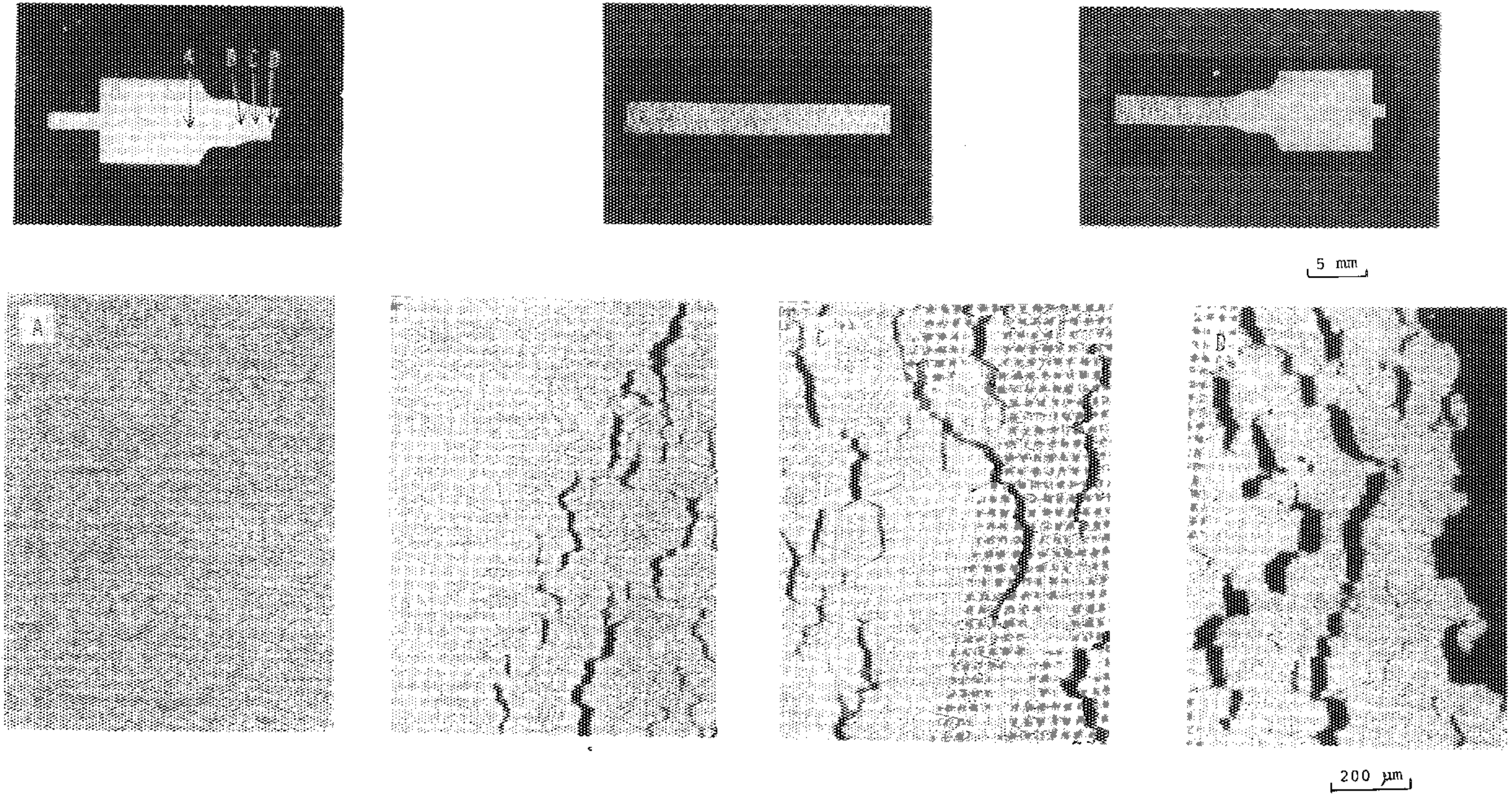

Fig.8 Macro- and micro-structure of irradiated Hastelloy-X creep-tested under comparatively low stress. The specimen was removed before final fracture
Stress; 1.5 kg mm^2, Time; 2667.5 hr, Strain; 12.9 %

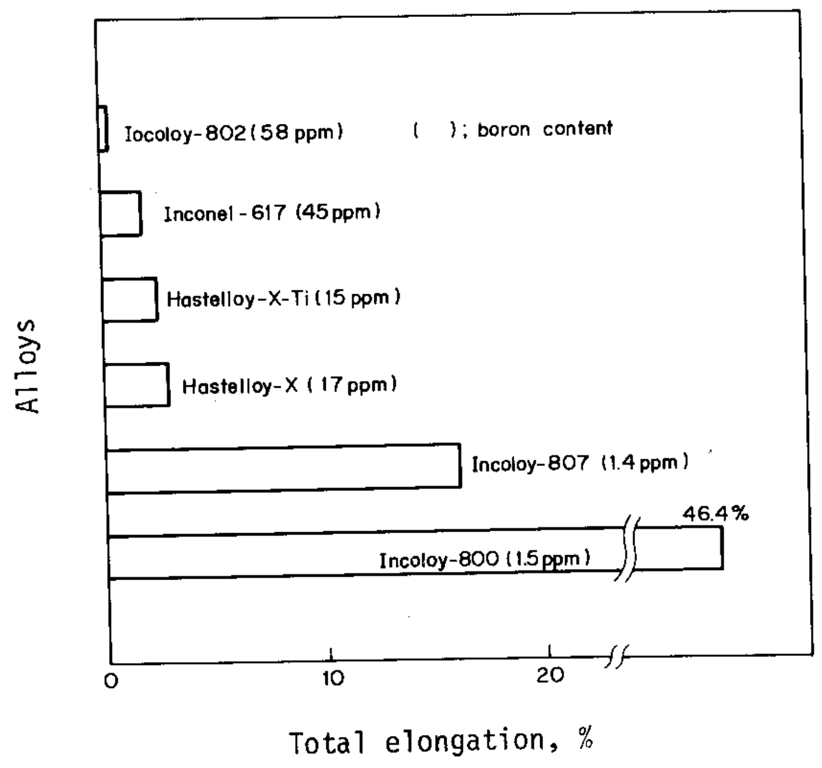

Fig.9 Effect of boron content on the tensile ductility of irradiated super alloys

Irrad; 2.1 x 10^{20} n/cm^2 (thermal)

Inc.; 640°C x 320 hr,

Hx; 910°C x 320 hr

Test; 900°C in vacuum, 0.67 %/min

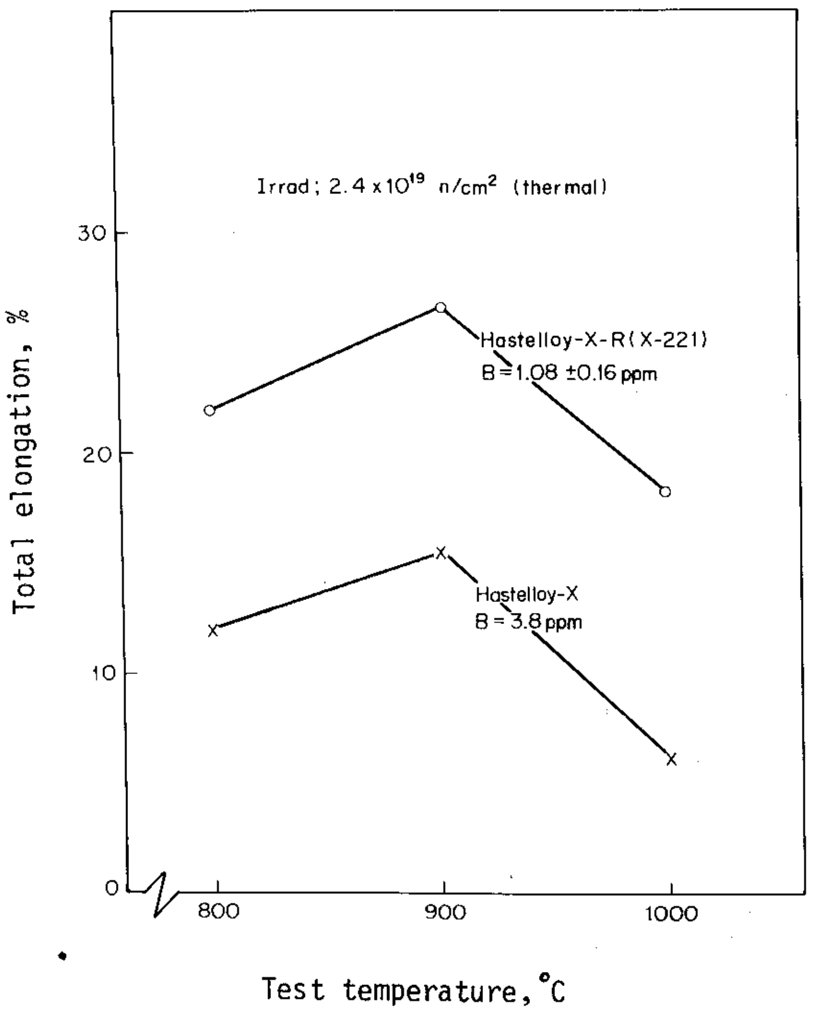

Fig.10 Effect of boron content on the tensile ductility of irradiated-Hastelloy-X

Irrad; 2.4 x 10^{19}n/cm^2(thermal)

50°C x 268 hr

Test; 900°C in vacuum, 0.67 %/min