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Effects of Thermal Neutron Irradiation on Ductility of Austenitic Heat Resisting Alloys for HTR Application

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Loss of high temperature ductility due to thermal neutron irradiation was examined by slow strain rate test in vaccum up to 1000°C. The results on two heats of Hastelloy alloy X with different boron contents were analysed with respect to the influence of the temperatures of irradiation and tensile tests, neutron fluence and the associated helium production due to nuclear transmutation reaction. The loss of ductility was enhanced by increasing either temperature or neutron fluence. Simple extrapolations yielded the estimated threshold fluence and the end-of-life ductility values at 900 and 1000°C in case where the materials were used in near-core regions of VHTR. The observed relationship between Ni content and the ductility loss has suggested a potential utilization of Fe-based alloys for seathing of the neutron absorber materials. Decreasing the impurity boron content is also suggested to be important in increasing the threshold fluence for embrittlement.

Keywords: High Temperature Ducility, Thermal Neutron Irradiation, Hastelloy Alloy X, Boron, Helium, Nuclear Transmutation Reaction, Threshold Fluence, VHTR, Ni Content, Ductility Loss, Fe-Based Alloys

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高温原子炉用耐熱合金の延性に及ぼす熱中性子照射の効果

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（1983年7月6日受理）

硼素含有量の異なる二種のハステロイーXの高温照射脆化に及ぼす照射温度，試験温度，熱中性子照射量および核変換反応にもとづくヘリウム生成の影響について検討を加えた。この場合，（n，α）反応によるヘリウム生成に関しては，^{10}B（n，α）^{7}Li反応に加えて，比較的近年になって発見された^{58}Ni（n，α）^{59}Ni（n，α）^{56}Fe 2段反応にも着目して脆化との関係を考察した。また，ニッケル基合金の他に鉄基合金も加えた両合金群のヘリウム脆化に対する感受性についても比較検討を行った。

これらの結果から，延性の低下は照射温度，試験温度および熱中性子照射量の増加により促進されることが明らかとなった。また，破断伸びに対する熱中性子照射量依存性の外挿結果から900°Cおよび1000°Cにおけるしきい照射量および高照射量域における延性値を推定した。一方，材料中のニッケル含有量と照射脆化との関連性から高温ガス炉の制御棒用部材には鉄基合金の方が照射環境に対する適応性が高いことが判った。

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1. INTRODUCTION

Neutron irradiation effects have not been paid much attention in the technology of metals for the conventional HTGR. In general, rather marked loss of ductility due to integranular cracking has been known to occur when austenitic alloys are irradiated with neutrons, and deformed at temperatures above the so-called equicohesion temperature of the alloys.(1)(2)

The cause of such ductility loss in the thermal neutron irradiation has been attributed to the transmutation of boron in the alloys by the reaction, $^{10}B(n{\cdot}\alpha)^{7}Li$ [3838 barn] to form helium.(3) More recently the two step reaction of thermal neutrons with nickel, $^{58}Ni(n{\cdot}\gamma)^{59}Ni$[4.4 barn] → $^{59}Ni(n{\cdot}\alpha)^{56}Fe$[13 barn], was recognized as the additional contributor to helium production.(4)∿(6) The extent to the loss of ductility is generally expected to be enhanced by increasing temperature and decreasing deformation rate. In HTGR of comparatively high operating temperatures, components such as the control rod seathing and thermal barrier liner may bare critical evaluation on their integrity during the projected life.

The present work aims at some clearer location of the problem area by making semi-quantitative analysis of the post irradiation ductility of some of the candidate alloys after giving various fluence up to 2×10^{21} n/cm^2.

2. PREVIOUS WORK(7)

Post irradiation creep tests of a commercial heat Hastelloy alloy X(boron 2.3 ppm) after irradiating thermal neturon to 6.6×10^{20} n/cm^2 at 670 and 880°C showed significant reduction in the rupture life and ductility at 900°C. The material degradation was seen to occur in the formation of premature integranular cracking resulting in earlier onset of the accelerated creep stage, which shortened essential life by approximately one order of magnitude. Based on the preliminary results of the post irradiation tensile tests on materials with different boron contents, the authors suggested possible means of combatting the problem by employing low boron grade materials.

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3. EXPERIMENTAL METHODS

3.1 Materials

Majority of the test results discussed in the present work were obtained from hot forged and solution annealed Hastelloy alloy X with 3.8 ppm boron and the grain size ASTM No. 4. Several other alloys were also tested for examining the effect either of boron or nickel contents. The chemical compositions are shown in Table 1. All the specimens used were the miniature size tensile bars of 3 mm dia. × 30 mmℓ at gage section.

3.2 Reactor irradiation

The materials were irradiated by either JRR-2 (CP-5 type) and JMTR using 3 types of capsules depending on the expected fluence and irradiation temperature. Thermal neutron fluences given were ranged between 2.7×10^{17} and 2.0×10^{21} n/cm^2. The fast neutron fluence given together was about one order of magnitude lower than that of thermal neutrons, and hence, the contribution to the loss of ductility is little in the present case.

The irradiation temperatures were controlled at isothermal levels which were devided in two groups. i.e. LT. (< 300°C) and HT. (820, 900 and 1000°C).

3.3 Post irradiation tests

Tensile tests were made at 700, 800, 900 and 1000°C in $(1 \sim 4) \times 10^{-6}$ torr vacuum. The strain rate employed ranged between 0.17 to 33 %/min. The preliminary results indicated that the obtained ductility descreased monotonically with decreasing the rate. Majority of the results were obtained with the rate of 0.67 %/min for convenience.

4. RESULTS

4.1 Influence of test temperature on post irradiation tensile properties

In the results of Hastelloy alloy X after irradiating 6×10^{17} n/cm^2 neutrons at 940°C, the ductility loss with increasing the test temperature was almost monotonic as seen in Fig. 1. The apparent strength change, in contract, was negligible (Fig. 2). The microstructures near fracture surface and uniform elongation region are shown in Fig. 3. Sharp intergranular cracking with little plastic deformation in each grain suggests the occurence of early premature cracking during the test.

4.2 Effect of thermal neutron fluence on total elongation

The effect of the neutron fluence on ductility was examined at 900 and 1000°C after irradiating at 60°C. The results were plotted in the double logarithmic graphs as shown in Fig. 4, where a set of straight lines were obtained. Extrapolation to the lower fluence levels yields estimated threshold fluences of 6×10^{16} and 2×10^{16} n/cm^2 at 900 and 1000°C, respectively.

4.3 Influence of irradiation temperature on total elongation

Figure 5 compares the plots of total elongation versus neutron fluence for two groups of specimens irradiated at different temperatures. Irradiation at 900 to 1000°C range has enhanced the ductility loss significantly as compared with the low temperature case, and the difference is seen to become more pronounced as fluence is increased, for which the materials spend more time at the elevated temperature.

4.4 Influence of boron content in alloy on post irradiation ductility

Up to some 10^{19} n/cm^2 levels, the low boron heat of Hastelloy alloy X showed higher post irradiation ductility at all the test temperatures examined. With the fluence above some 10^{20} n/cm^2, however, little difference was seen between the two heats as typically shown in Fig. 6.

4.5 Relation between irradiation embrittlement and nickel content in alloys

Five iron-based austenitic alloys including experimental heats were irradiated together with the two heats of Hastelloy alloy X to 2.7×10^{20} n/cm^2 at 60°C.

Post irradiation tensile ductility obtained from the tests at 900°C are summarized in Fig. 7 and Fig. 8. Despite considerable variability in the composition of those alloys the degree of ductility loss, as determined by the fraction of the net loss to the total elongation of the unirradiated specimens, was seen to have consistent dependence to the nickel content of the materials.

Fig. 9 shows the microstructures of fractured and uniformly elongated regions taken after tensile testing. The fracture mode was intergranular with little or no plastic deformation occurring within the grain internals. The difference between Fe-based and Ni-based alloys, however, was not clearly recognized in a glance as long as the feature of the fracture mode was concerned.

5. DISCUSSION

Hastelloy alloy X shows minima in the tensile ductility at around 700°C. The observed loss of ductility is the phenomenon specifically occurring at temperatures above such minima. In the present work it was confirmed that the trend persists with temperature increase up to 1000°C in spite of the considerable softening of the grain at such high temperature.

Contribution of boron has been known to be dependent on its concentration and distribution(9)(10) in the metal. Because of its high transmutation cross section and limited quantity in the alloy, helium formation is expected to level off with the thermal neutron fluence of some 10^{20} n/cm^2 range, while the two step reaction of Ni and thermal neutrons is expected to become dominant above such level.(4) The amounts of helium produced by those reactions are calculated on the present two heats of Hastelloy alloy X, and the results are plotted in Fig. 9. As seen in the plot, the difference between the two materials disappears essentially at the fluence level between 10^{20} and 10^{21} n/cm^2.

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The plot of total elongation and fraction of ductility loss versus helium concentration is shown in Fig. 11 in connection with Fig. 6. The result indicates that total elongation decreases with increasing the fluence, i.e. amount of transmuted helium, and then the trend of leveling off was noted at higher fluence region. As for the fraction of ductility loss, the threshold value of helium concentration for irradiation embrittlement to occur is dependent upon boron content of the alloys and tends to increase with helium concentration increase. Concerning the ductility versus fluence relations obtained by the extrapolation of the trend shown in Fig. 4 and Fig. 5 the following facts are of particular significance:

i) Ductility loss at fluence of 10^{21} to 10^{22} n/cm^2 is significantly great to cause practically a state of nil ductility, where the projected design life in terms of neutron fluence has been set for the seathing of the neutron absorber materials.

ii) The estimated threshold fluences for the occurence of appreciable ductility loss were as low as 10^{16} n/cm^2.

In the design of VHTR, the projected neutron fluence on the thermal barrier liner may go at least to about 1×10^{18} n/cm^2. In view of the comparatively low boron contents of the materials used in the present study, there can be a possibility of having much lower threshold values for general commercial heats manufactured without boron control, as they often contain boron over some 10 ppm. Service performance prediction in such case should take the neutron irradiation into account as one of the major environmental degradation factors.

The calculated helium concentrations corresponding to the estimated thresholds for 900 and 1000°C are 9.4×10^{-10} and 3.2×10^{-10} atomic fraction respectively, which are considered to be within resonable order of magnitude referring to the values reported by Stiegler et al.(3) on type 304 stainless steel at 700°C. In the material selection for seathing of B_4C neutron absorbers, the use of iron-based alloys may be preferred since the expected life time is much more than other structural materials and the life is determined rather by the amount of neutrons absorbed.

6. SUMMARY

Ductility loss of neutron-irradiated Hastelloy alloy X and several other Fe based austenitic alloys at elevated temperatures has been examined by post irradiation tensile test. The contribution of helium production due to the $^{58}Ni(n,\gamma)^{59}Ni(n,\alpha)^{56}Fe$ reaction in addition to the $^{10}B(n,\alpha)^{7}Li$ reaction was considered.

The results obtained are summarized as follows:

(1) The post irradiation ductility in low strain rate (0.67 %/min.) tensile test decreases with increasing test temperature up to 1000°C.

(2) As for the helium embrittlement attributed to (n,α) reaction, the $^{10}B(n,\alpha)^{7}Li$ reaction is dominant at lower fluences, and Ni-two step reaction becomes predominant with fluence increase to above 10^{21} n/cm^2.

(3) In the low temperature irradiation results, extrapolation gave the ductility values of about 3.5 % at 900°C and about 1.5 % at 1000°C for the thermal neutron fluence of 10^{22} n/cm^2.

(4) The threshold fluences for embrittlement were also estimated as 6×10^{16} n/cm^2 at 900°C and 2×10^{16} n/cm^2 at 1000°C, which corresponded to the calculated atomic helium fractions of 9.4×10^{-10} and 3.2×10^{-10}, respectively.

(5) Increase of irradiation temperature to 900 ∿ 1000°C range intensified the ductility loss at high fluence range.

(6) The degree of ductility loss was dependent roughly on the nickel contents of alloys and possible preferred use of iron-based alloys for in-core components was suggested.

REFERENCES

1) R.S. Barns: Nature, 206 (1965), p. 2307
2) D.R. Harriers: J. Bri. Nucl. Energy Soc., 5 (1956), p. 74
3) J.O. Stiegler and J.R. Weir, Jr.: Ductility (1967), p. 311 [ASM]
4) J. Weitman, N. Daverhög, and S. Farvolden: Trans. ANS, 13 (1970), p. 557
5) L.R. Birss and W.E. Ellis: BNES European Conf. on Voids formed by Irradiation of Reactor Materials, (1971), p. 339
6) A.A. Bauer and M. Kangilaski: J. Nucl. Mater., 42 (1972), 91
7) Y. Ogawa, et al.: The Second US-Japan HTGR Safety Tech. Seminar. (1978)
8) K. Watanabe, et al.: Tetsu-To-Hagane (J. Iron Steel Ints. Japan), 68, (1982) 682
9) W.R. Martin and J.R. Weir: ASTM STP 426, (1967), p. 440
10) D.R. Harris, A.C. Roberts, G.T. Rogers, J.D.H. Hughes, and M.A.P. Dewey: Radiation Damage in Reactor Materials, 2 (1969), p. 357 [IAEA]

Table I Chemical Compositions of Specimens

Chemical Compositions of Specimens (wt.%) B ; ppm

	C	Si	Mn	P	S	Cr	Co	Mo	W	Nb	Fe	Ni	Aℓ	Ti	N	B
Hastelloy -X	0.06	0.43	0.60	0.007	0.005	21.49	0.98	8.82	0.53	—	18.03	Bal.	0.41	0.03	0.027	3.8
Hastelloy-XLB	0.08	0.03	0.65	<0.005	<0.005	21.98	0.05	8.81	0.54	—	18.35	Bal.	<0.02	0.02	0.005	1.1
Fe-30Ni-20Cr	0.21	0.12	0.23	0.007	0.004	19.67	0.02	3.07	1.02	1.07	Bal.	30.12	—	—	0.012	5.0
Fe-14Ni-16Cr (I)	0.07	0.89	1.73	0.006	0.007	15.99	0.02	2.52	<0.01	—	Bal.	13.93	0.006	0.11	0.005	38.0
Fe-18Ni-14Cr	0.07	0.33	1.73	0.004	0.011	14.04	0.03	4.47	<0.01	—	Bal.	18.01	0.004	0.24	0.004	36.0
Fe-16Ni-15Cr	0.06	0.32	1.73	0.004	0.009	15.03	0.02	3.49	<0.01	—	Bal.	16.05	0.004	0.24	0.003	36.0
Fe-14Ni-16Cr (II)	0.07	0.32	1.74	0.005	0.009	15.99	0.02	2.48	<0.01	—	Bal.	13.89	0.004	0.24	0.003	36.0

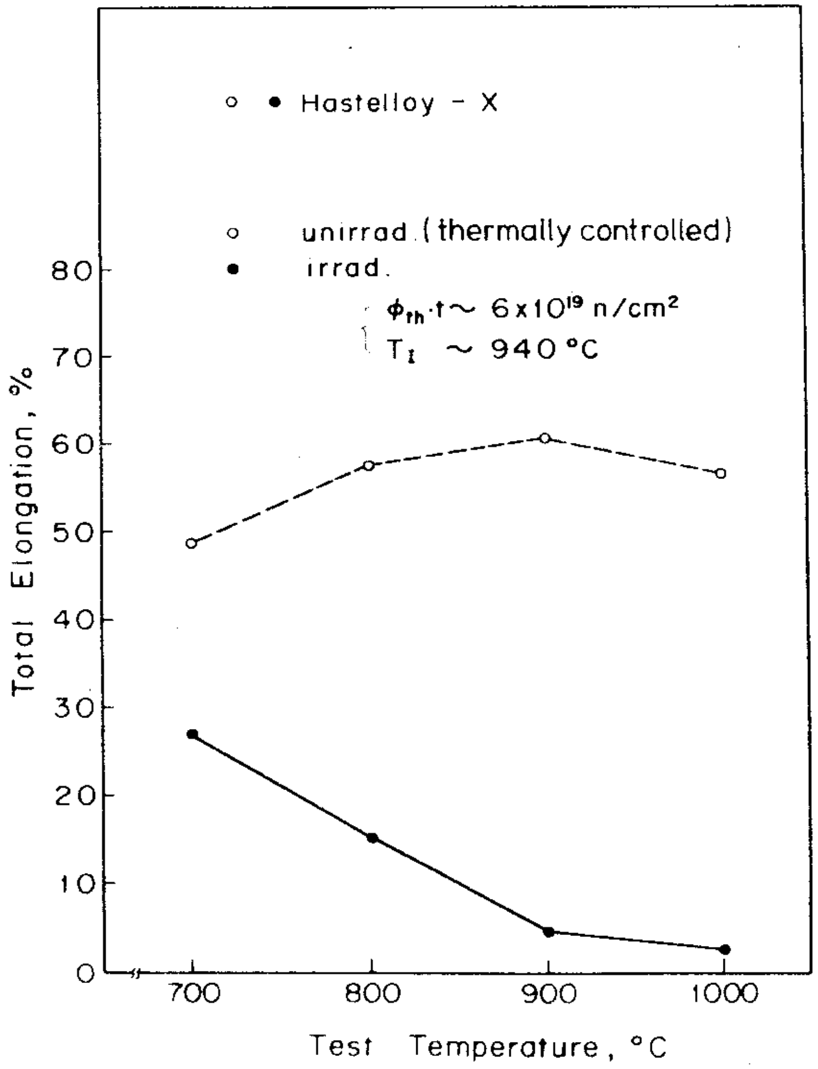

Fig. 1 Total elongation versus test temperature for unirradiated and irradiated Hastelloy-X

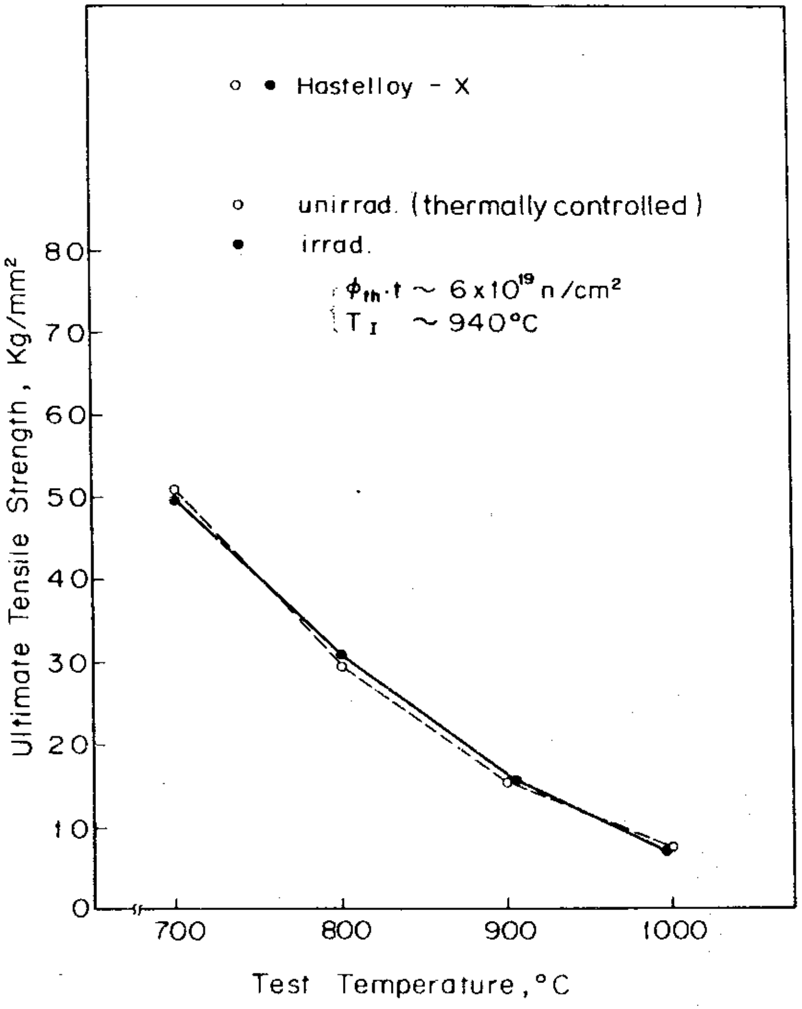

Fig. 2 Ultimate tensile strength versus test temperature for unirradiated and irradiated Hastelloy-X

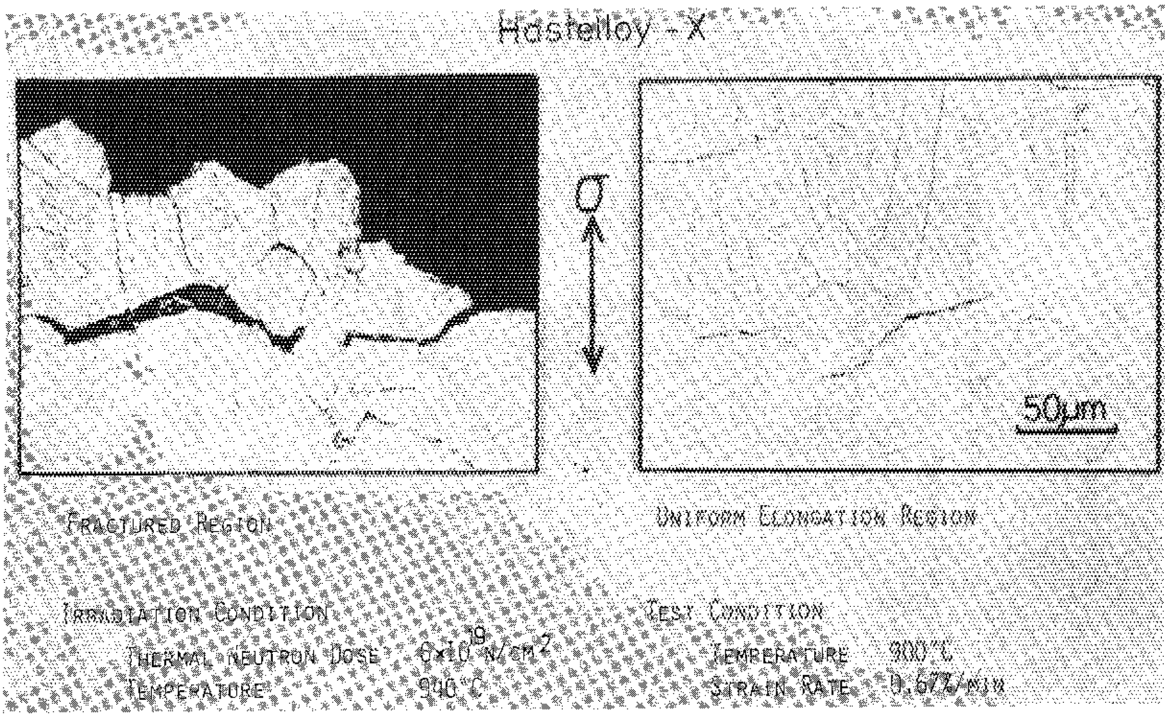

Fig. 3 Microstructures of irradiated Hastelloy-X after tensile testing

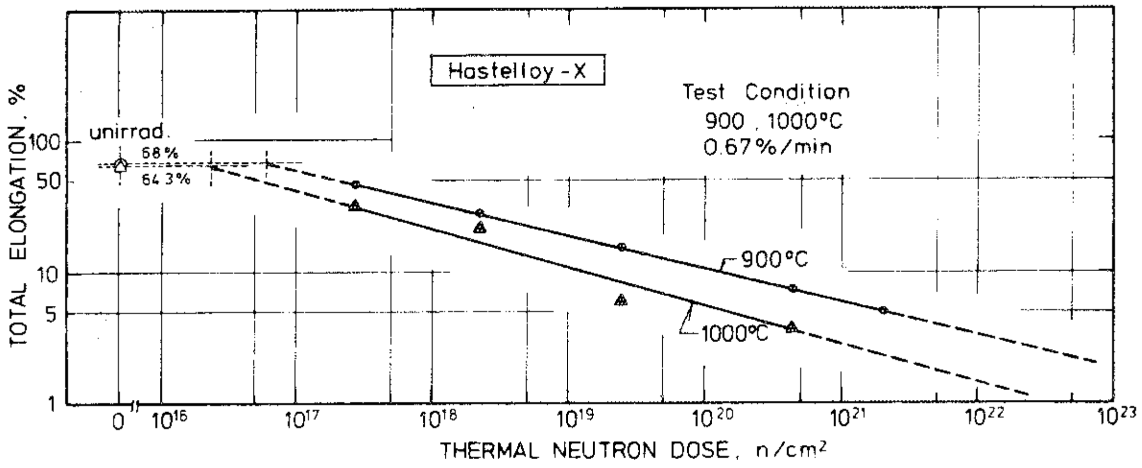

Fig. 4 Effect of thermal neutron dose on the total elongation of Hastelloy-X at test temperature of 900 and 1000°C

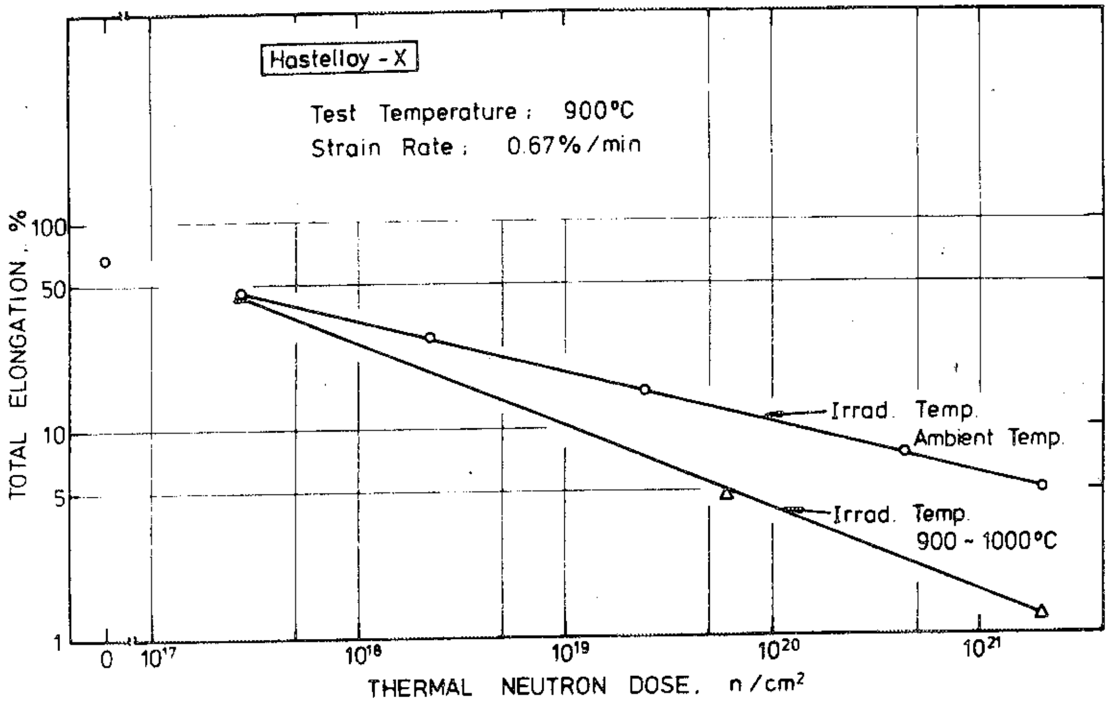

Fig. 5 Total elongation versus thermal neutron dose for Hastelloy-X irradiated at different temperatures

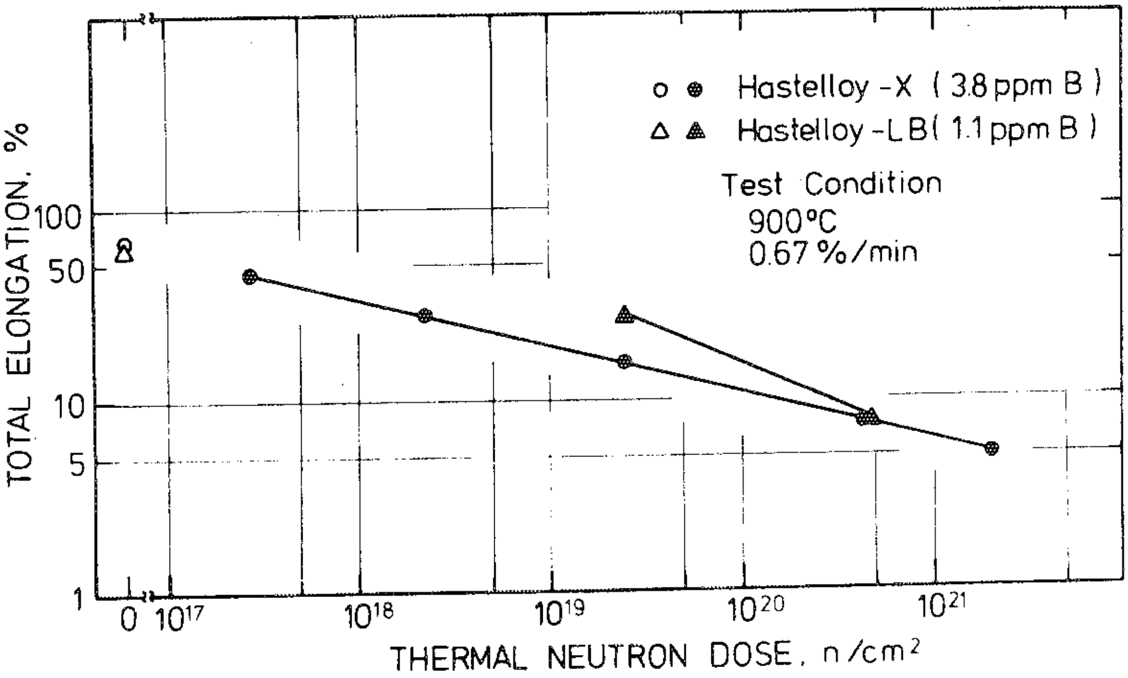

Fig. 6 Effect of thermal neutron dose on the total elongation of Hastelloy-X and Hastelloy-XLB

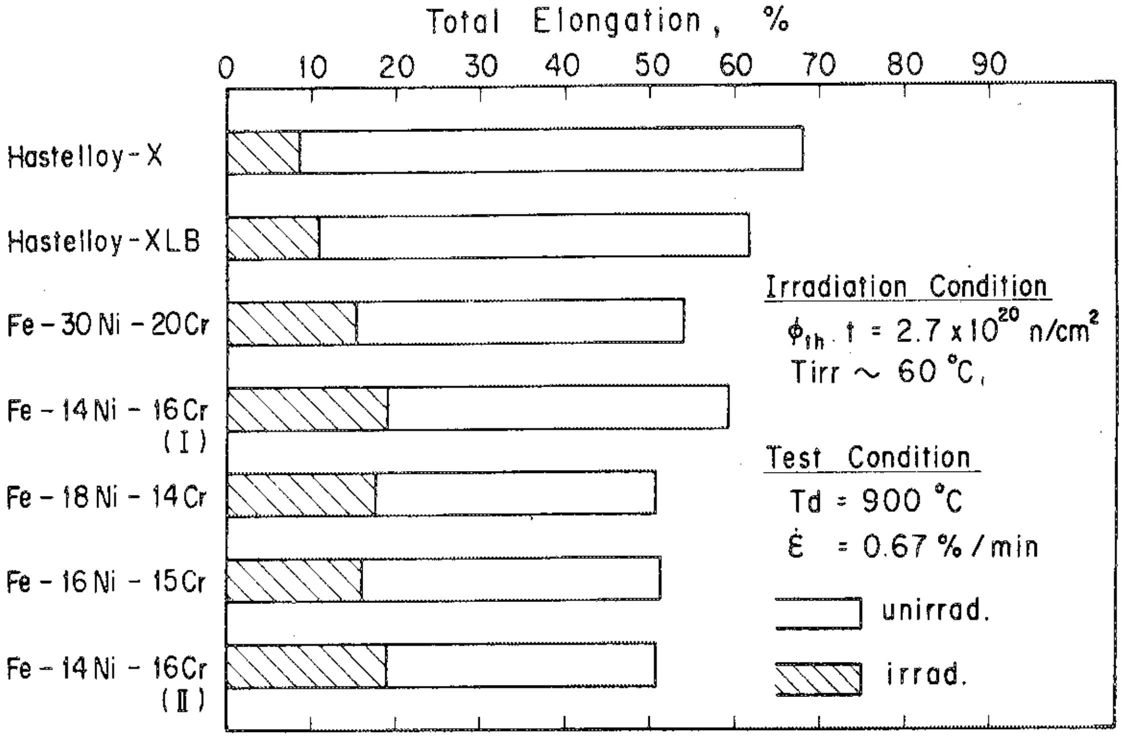

Fig. 7 High temperature ductility of various austenitic alloys with different Ni content after neutron irradiation

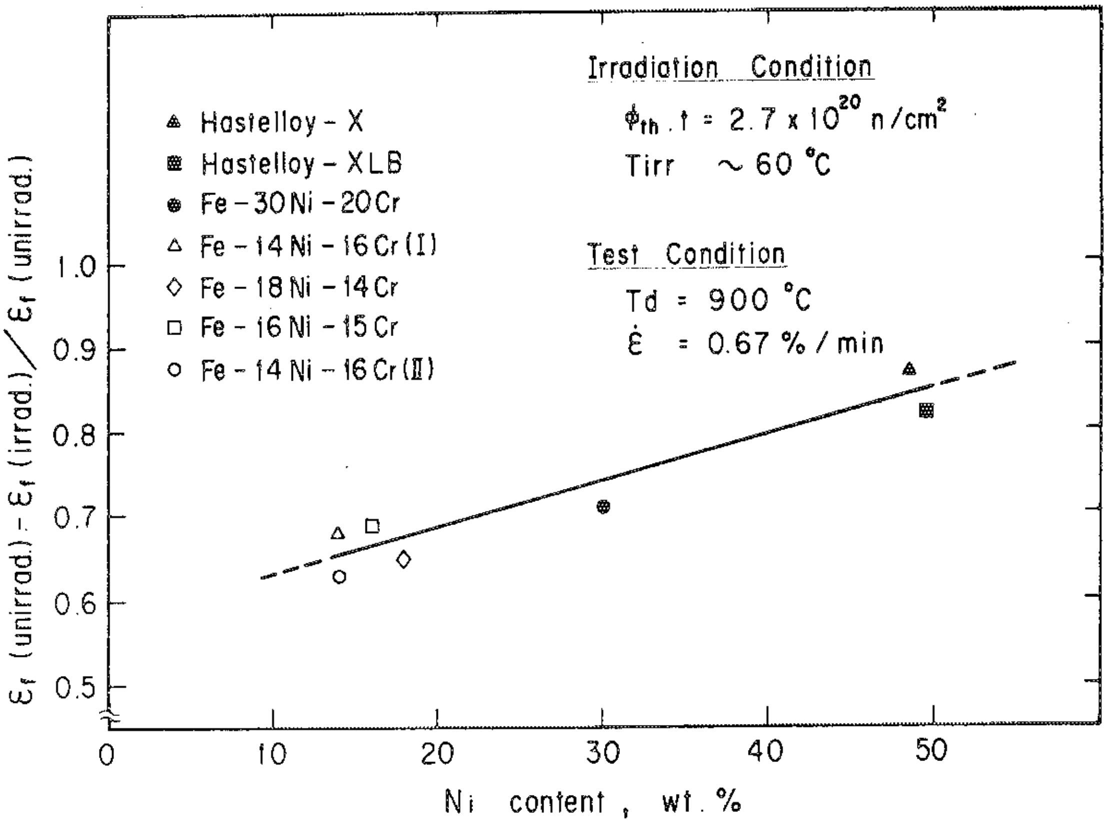

Fig. 8 The relation between Ni content and ductility loss in irradiated austenitic alloys

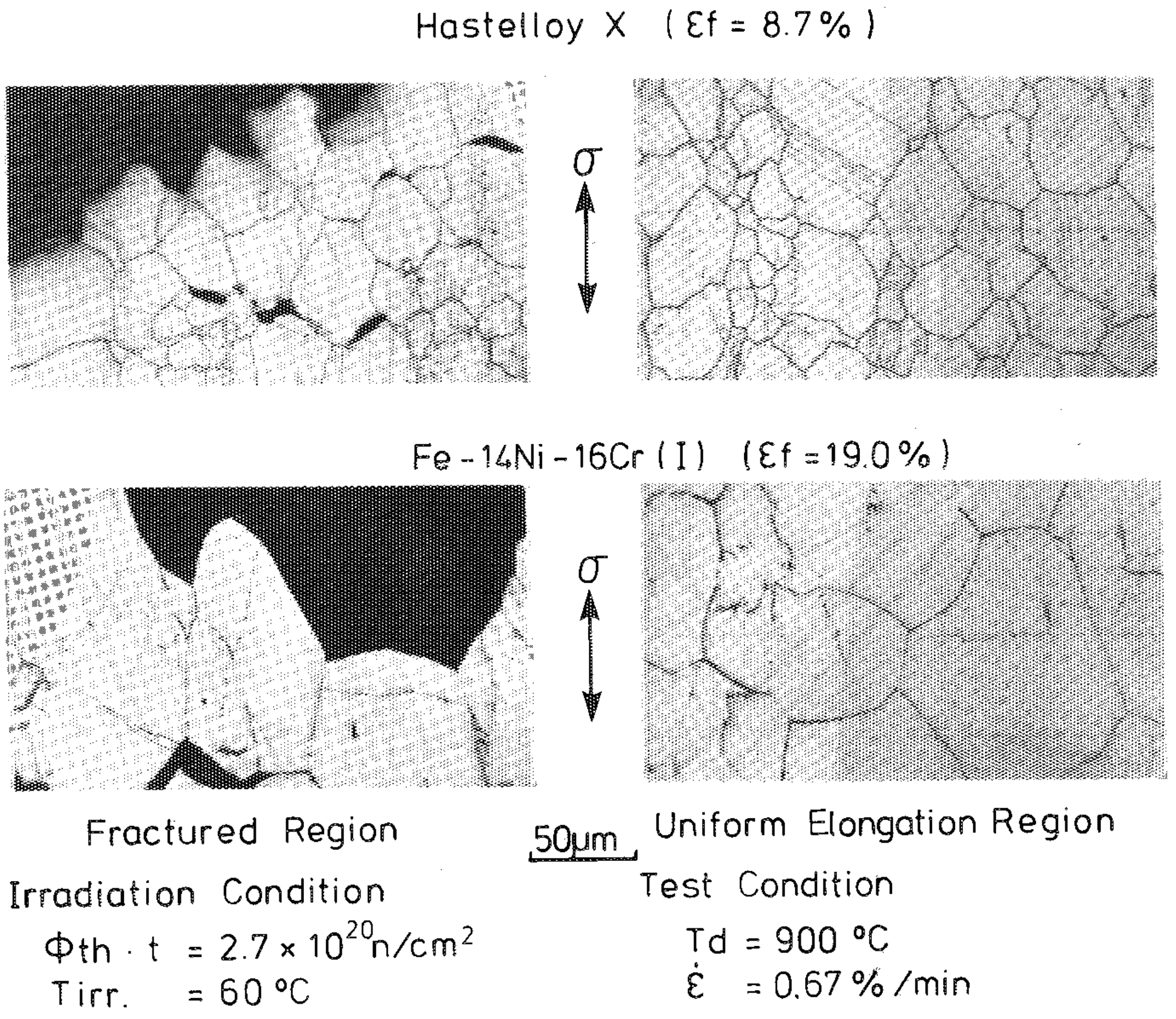

Fig. 9 Microstructures of irradiated Ni-based and Fe-based alloy after tensile testing

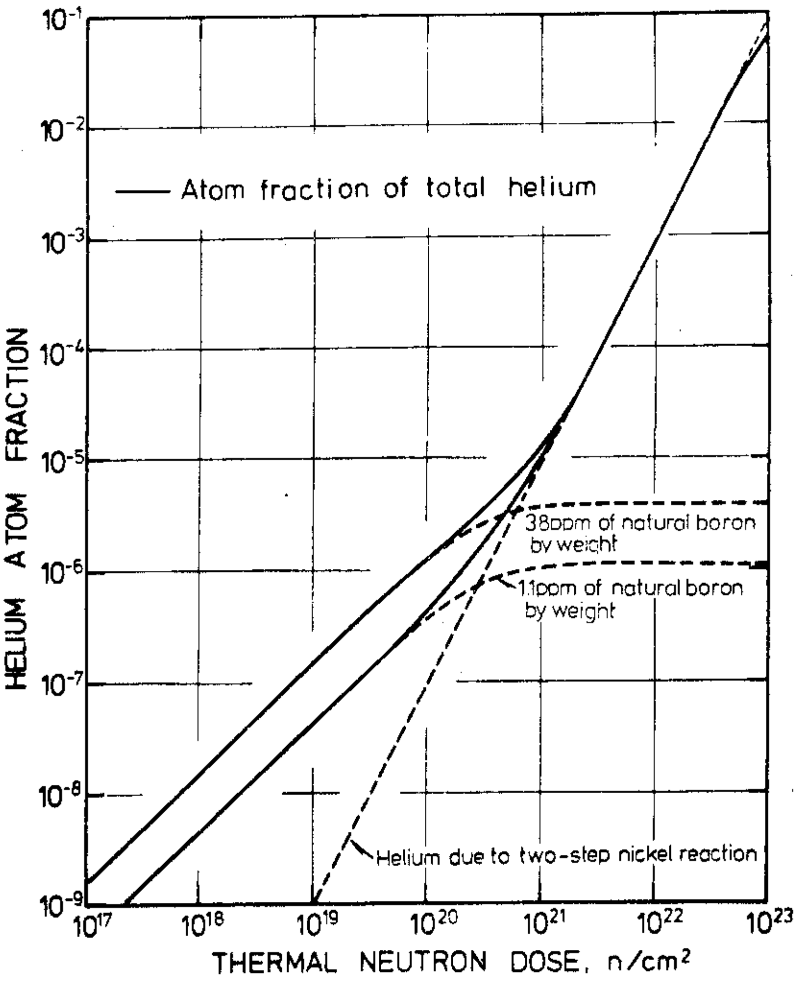

Fig. 10 Helium production resulting from transmutation reactions in Hastelloy-X and Hastelloy-XLB

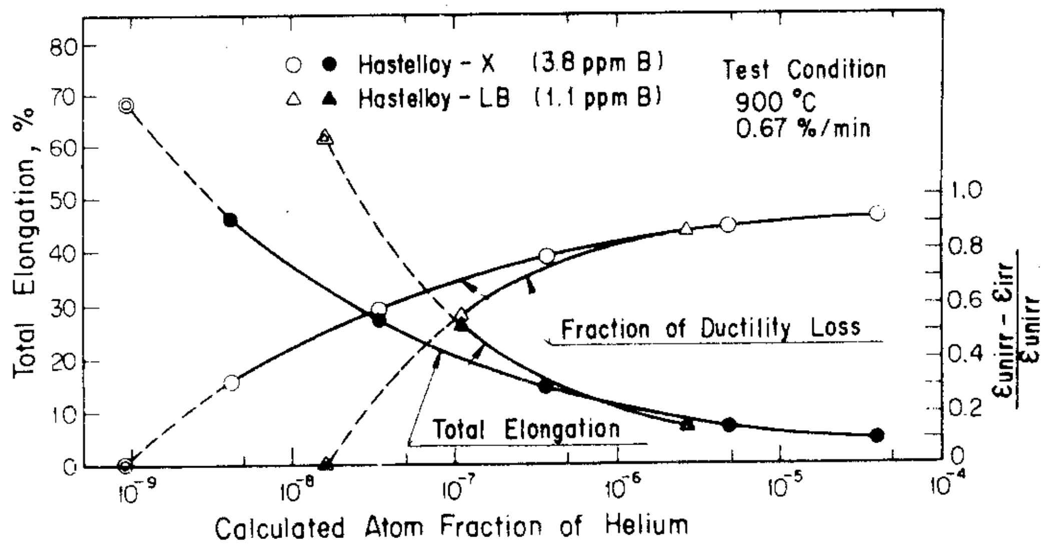

Fig. 11 Total elongation and fraction of ductility loss versus helium concentration