MAR 11 1996

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Contract Program or Project Title:	Heavy-Section Steel Irradiation (HSSI) Program, Metals and Ceramics Division
Subject of this Document:	The Effect of Thermal Neutrons and Fast Flux on Tensile Properties of Ferritic Steels Irradiated at Low Temperature
Type of Document:	Letter Report
Authors:	R. E. Stoller, K. Farrell, and L. K. Mansur
Date of Document:	February 1996
Responsible NRC Individual and NRC Office or Division:	M. G. Vassilaros (301-415-6000) Division of Engineering U.S. Nuclear Regulatory Commission

Prepared for the
U.S. Nuclear Regulatory Commission
Washington, DC 20555
Under Interagency Agreement DOE 1886-8109-8L
NRC JCN No. L1098

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U.S. Department of Energy
under Contract No. DE-AC05-96OR22464

ORNL/NRC/LTR-96/3

THE EFFECT OF THERMAL NEUTRONS AND FAST FLUX ON TENSILE PROPERTIES OF FERRITIC STEELS IRRADIATED AT LOW TEMPERATURE

R. E. Stoller, K. Farrell, and L. K. Mansur

Manuscript Completed - December 1995
Date Published - February 1996

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Roger E. Stoller, Kenneth Farrell, and Louis K. Mansur

ABSTRACT

The effect of neutron energy spectrum and fast flux was investigated by irradiating four ferritic steels in several fission reactors. Tensile data were obtained following irradiation in the hydraulic tube of the High Flux Isotope Reactor (HFIR) at the Oak Ridge National Laboratory (ORNL), two locations in the High Flux Beam Reactor at the Brookhaven National Laboratory, and three locations in the Ford Nuclear Reactor at the University of Michigan. The materials included the A 212B and A 350LF3 steels used in the fabrication of the pressure vessel of the HFIR at the ORNL, an A 533B steel commonly employed as a correlation monitor in commercial reactor surveillance programs, and an A 36 structural steel. The materials were irradiated in the form of sheet tensile specimens at a temperature of ~50 to 60°C. The fast neutron flux (> 1.0 MeV) varied by over four orders of magnitude in these irradiations, from a low of 1.4×10^{10} to 4.0×10^{14} n/(cm^2·s). Use of tensile data from the HFIR surveillance program extends this comparison to a fast flux of only 1.8×10^{8} n/(cm^2·s) for the A 212B plate. The influence of thermal neutrons was explored for thermal-to-fast neutron flux ratios ranging from nearly zero to 370. Dose levels reached 1.5×10^{-3} dpa in the 370-ratio position and were higher in the other sites. Analysis of the results indicates that the radiation-induced yield strength changes at 50 to 60°C in these materials are quite insensitive to the fast flux. Comparison of very high and low thermal-to-fast flux ratios indicates a significant effect of thermal neutrons. The specimens that were shielded from thermal neutrons showed much less strengthening per unit of fast fluence than the unshielded specimens. However, the results indicate that the effects of thermal neutrons at this temperature can be well accounted for through the use of displacements per atom as a correlation parameter, i.e., the ability of atomic displacements from thermal neutrons to induce hardening appears to be nearly equivalent to those generated by fast neutrons.

INTRODUCTION

Radiation-induced changes in mechanical properties have traditionally been correlated using fast neutron exposure parameters such as the fluence above 0.1 or 1 MeV. The use of fast fluence reflects the implicit assumption that lower energy neutrons do not provide a significant contribution to the total damage generated. Although this is true for most cases of interest to commercial reactors, the general neglect of lower energy neutrons can confound the interpretation of data obtained from some materials test reactors. An absorbed dose parameter such as atomic displacements per atom (dpa) more adequately accounts for differences in neutron energy spectra since it incorporates the total energy absorbed by the material that contributes to atomic displacements.[1] Its superiority to fast fluence for correlating embrittlement data has been recognized and its use has been encouraged by many researchers.[2]

However, the possibility that the damage efficiency of all atomic displacement reactions may not be the same is a limitation of dpa. In particular, it has been proposed that low-energy displacements, such as those produced by thermal neutrons, may be more efficient (per unit dpa) at producing some property changes than are displacements from fast neutrons.[3] A second parameter that requires attention when correlating data from different environments is the neutron flux level or displacement rate. In some situations, a lower displacement rate could also be expected to lead to a greater damage efficiency per unit dose.[3,4]

Both thermal neutrons and a low displacement rate were initially suggested as possible causes of what appeared to be accelerated embrittlement in the surveillance program of the High Flux Isotope Reactor (HFIR).[3-6] Since the HFIR pressure vessel operates at about 50°C, the exposure conditions were considered relevant to commercial reactor support structures and the present study was undertaken to investigate the effect of thermal neutrons and fast flux by irradiating four typical ferritic steels in several fission reactors. Although subsequent work has implicated previously uncounted atomic displacements due to a very high flux of high-energy gamma rays as being responsible for the HFIR embrittlement,[7,8] the data reported here remain relevant to the broader issue of the influence of neutron flux and spectral effects on embrittlement.

Tensile data were obtained following irradiation in the hydraulic tube of the HFIR at the Oak Ridge National Laboratory (ORNL), two locations in the High Flux Beam Reactor (HFBR) at the Brookhaven National Laboratory, and three locations in the Ford Nuclear Reactor (FNR) at the University of Michigan. The irradiations in the FNR were carried out by the University of California, Santa Barbara, as part of an informal collaborative agreement. The influence of thermal neutrons was explored for thermal-to-fast neutron flux ratios ranging from nearly zero to 370 and the displacement rate was varied from about 2×10^{-11} to 7×10^{-7} dpa/s. Dose levels reached 5×10^{-3} dpa in the site with a thermal-to-fast ratio of 370 and were higher in the other sites.

MATERIALS AND IRRADIATION CONDITIONS

The materials included the A 212 grade B (A212B) and A 350 grade LF3 (A350LF3) steels used in the fabrication of the pressure vessel of the HFIR, an A 533 grade B class 1 (A533B1) steel commonly employed as a correlation monitor in commercial reactor surveillance programs, and an A 36 structural steel. The A 36 steel is similar to that commonly employed in pressure vessel support structures and the A 533B material was Heavy-Section Steel Technology (HSST) Plate 02. The primary alloying constituents of all four materials are listed in Table 1.

All materials were irradiated in the form of ORNL SS3-type sheet tensile specimens and all tensile tests were conducted at ORNL. The SS3 specimen has a gage width and length of 1.52 and 7.62 mm, respectively. The specimen thickness is 0.762 mm and the overall specimen length is 25.4 mm. Tensile tests were performed in air at room temperature and a strain rate of 1.1×10^{-3}/s in a screw-driven machine; a crosshead speed of 0.51 mm/min was used. Two identical samples were tested and the results averaged in most cases.

Table 1. Major alloy components

Alloy	Composition (wt %)								
	Mn	Mo	Ni	C	Si	Cu	S	P	V
A212B	0.85	0.02	0.09	0.26	0.29	0.15	0.04	0.006	0.0005
A350LF3	0.55	0.03	3.3	0.18	0.29	0.11	0.02	0.01	0.001
A533B	1.55	0.53	0.67	0.23	0.20	0.14	0.014	0.009	0.003
A36	1.10	0.03	0.07	0.21	0.03	0.05	0.026	0.001	0.001

Irradiations were conducted at a temperature of ~ 50 to 60°C in the reactor sites listed in Table 2. As shown in this table, the fast neutron flux (> 1 MeV) varied by over four orders of magnitude in these irradiations, from a low of 1.4×10^{10} to 4.0×10^{14} n/(cm^2·s). The irradiations at the Oak Ridge Research Reactor (ORR) were conducted as part of the earlier evaluation of the HFIR vessel.[5] Use of tensile data from the HFIR surveillance program extends this comparison to a fast flux of only 1.8×10^8 n/(cm^2·s) for the A 212B plate. Since all materials could not be included in all of the irradiations, a summary of the materials irradiated in each site is shown in Table 3.

The highest thermal-to-fast neutron flux ratios were obtained in the HFBR in which heavy water is used as the moderator. The V14 site is located near the core edge, while the V10 site is located about 0.55 m from the core in the heavy water tank. Otherwise identical capsules were irradiated in the V10 site, with and without 1 mm Cd present as a thermal neutron absorber, to obtain a direct evaluation of the maximum effect of thermal neutrons. The cadmium-shielded V10 site is referred to as V10-Cd in the tables and figures included here. All of the capsules irradiated in the FNR were also partially shielded with 0.76 mm of cadmium to reduce the thermal neutron flux.

The neutron displacement rates in Table 2 are based on spectrally averaged displacement cross sections of 1500 b for the HFIR surveillance data, 1600 b for the HFIR HT, and 1500 b for the ORR irradiations.[7] Values for gamma ray-induced displacements in the HFIR surveillance data are taken from Reference 8. The values tabulated for the FNR and HFBR are based on the current estimated fluxes using displacement cross sections of 1500 and 10 b for the fast and thermal fluxes, respectively. More detailed dosimetry of the HFBR sites is in progress to obtain spectrally averaged displacement cross sections.

EXPERIMENTAL RESULTS AND DISCUSSION

The results of the tensile tests are summarized in Figures 1 through 6. In each case, the yield strength change is plotted as a function of fast fluence in part (a) and as a function of dpa in part (b). Figures 1 and 2 compare only the three different HFBR irradiation conditions for the HFIR A 212B and the HSST Plate 02, respectively. The other figures include data from all the available irradiations. Data from the HFIR surveillance program are included in Figure 3.

Table 2. Irradiation conditions for low-temperature irradiations

Reactor/site	Neutron flux (cm^{-2} s^{-1})		Displacement rate (dpa/s)	Thermal-to-fast flux ratio	Peak dose (dpa)
	> 1 MeV	Thermal[a]			
HFIR/K7-P5	1.75×10^{8}	3.64×10^{8}	1.71×10^{-12} [b]	2.1	1.21×10^{-3}
FNR/60D	1.4×10^{10}	$< 2.9 \times 10^{7}$	2.1×10^{-11}	< 0.003	2.94×10^{-4}
FNR/60B	1.8×10^{11}	$< 6.4 \times 10^{8}$	2.7×10^{-10}	< 0.003	4.44×10^{-3}
HFBR/V10-Cd	3.8×10^{11}	~ 0.	5.7×10^{-10}	~0.	1.52×10^{-3}
HFBR/V10	3.8×10^{11}	1.4×10^{14}	2.0×10^{-9}	368.	1.15×10^{-3}
FNR/60A	7.3×10^{11}	$< 2.2 \times 10^{9}$	1.1×10^{-9}	< 0.003	4.49×10^{-3}
ORR/P8	1.26×10^{13}	9.77×10^{13}	1.89×10^{-8}	7.7	2.55×10^{-2}
HFBR/V14	4.5×10^{13}	4.2×10^{14}	7.2×10^{-8}	9.3	3.10×10^{-2}
HFIR/HT-7	4.0×10^{14}	2.1×10^{15}	7.0×10^{-7}	5.3	5.60×10^{-2}

[a]Thermal flux estimate in FNR based on factor of 1000 reduction due to cadmium shield and essentially complete absorption by cadmium in HFBR V10-Cd.
[b]Neutrons contribute 2.78×10^{-13} dpa/s in this site and gamma rays add 1.43×10^{-12} dpa/s. Gamma ray contribution is negligible in other sites.

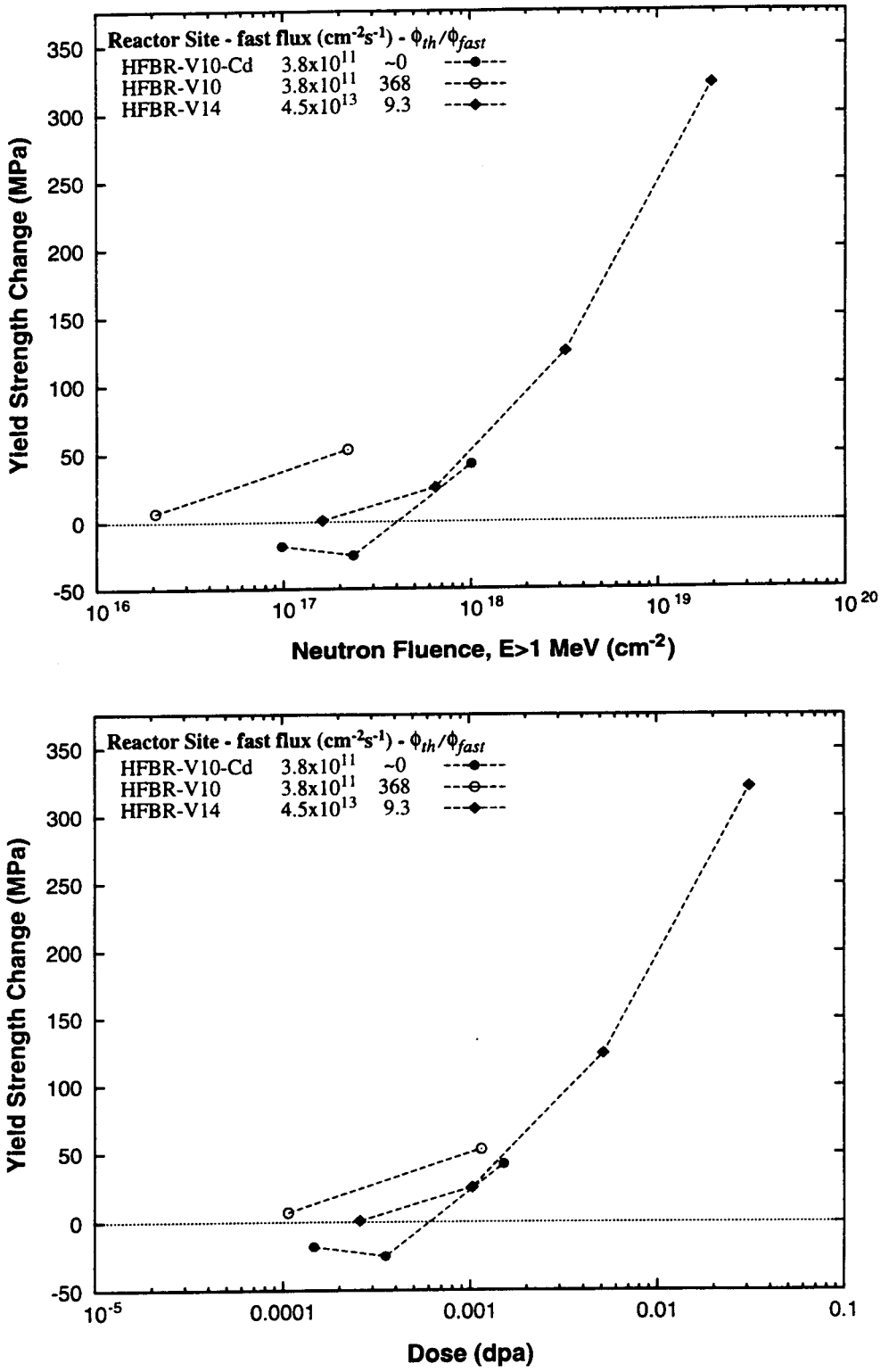

Figure 1. Yield strength change in HFIR A 212B material irradiated in the HFBR V10 and V14 sites; V10 irradiations were conducted both with and without cadmium shielding. (a) Neutron fluence dependence. (b) Dose (dpa) dependence.

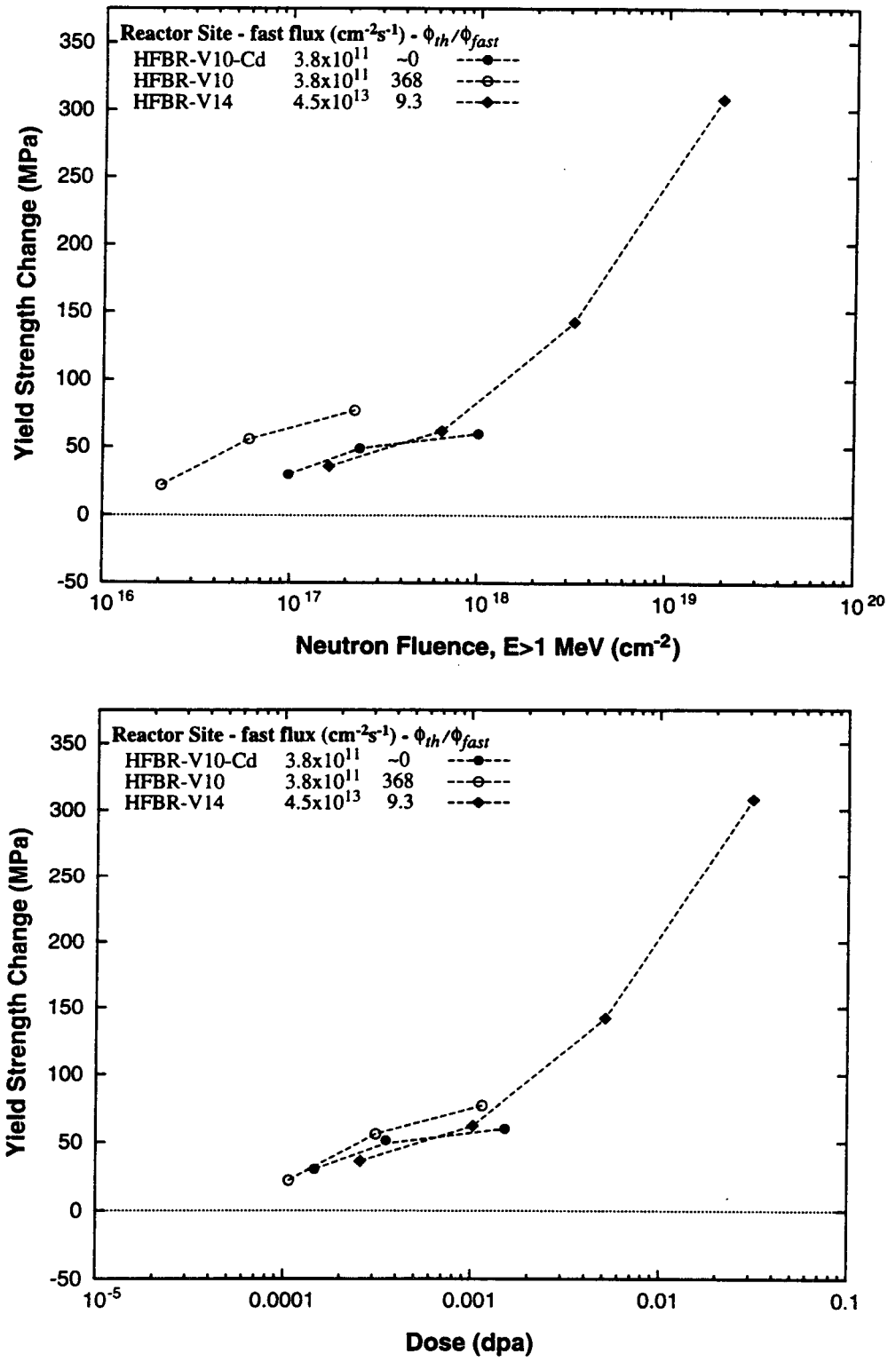

Figure 2. Yield strength change in A 533B correlation monitor irradiated in the HFBR V10 and V14 sites; V10 irradiations were conducted both with and without cadmium shielding. (a) Neutron fluence dependence. (b) Dose (dpa) dependence.

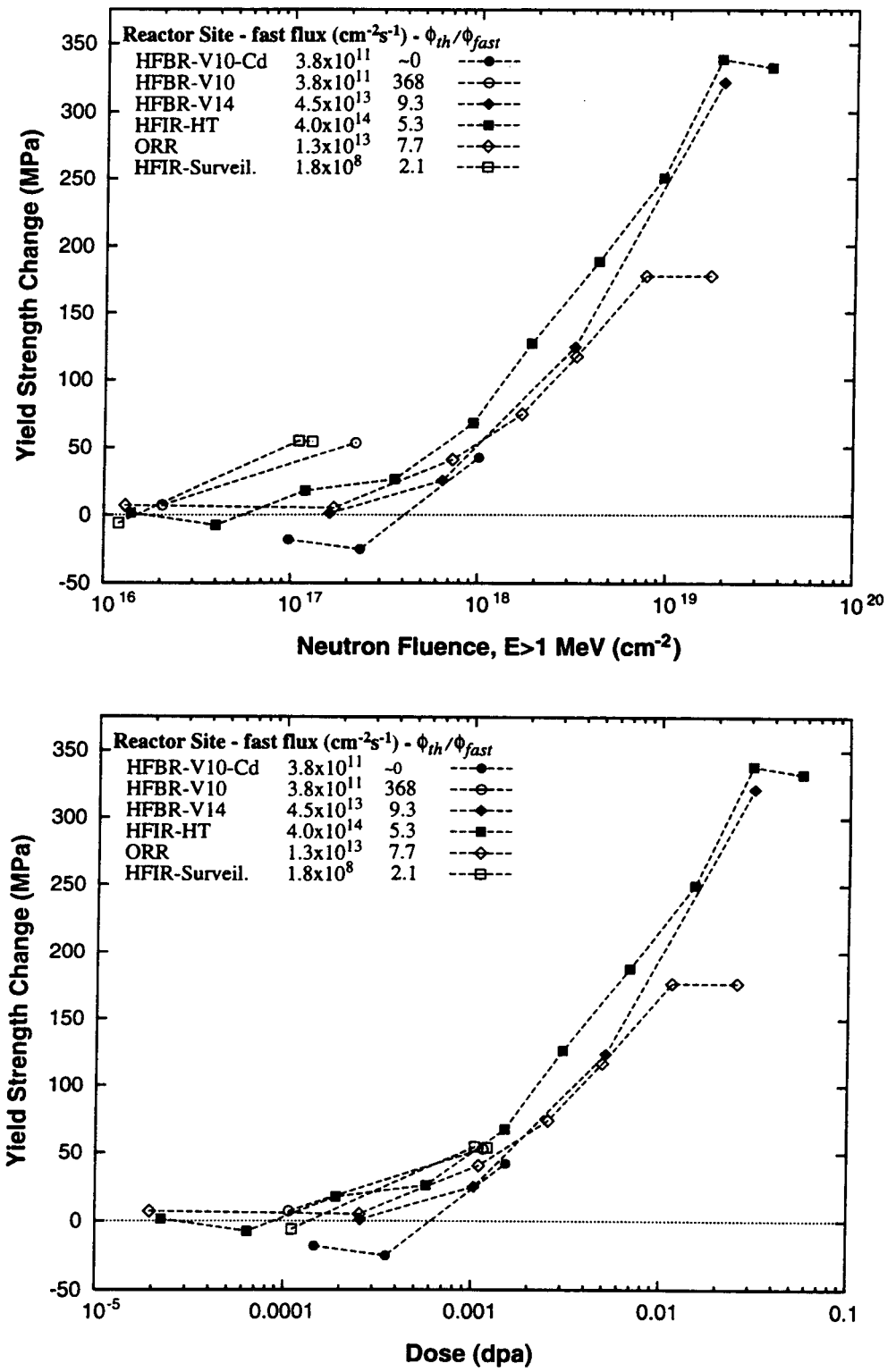

Figure 3. Yield strength change in HFIR A 212B material irradiated with a range of fast fluxes and thermal-to-fast flux ratios. (a) Neutron fluence dependence. (b) Dose (dpa) dependence.

Table 3. Sites (denoted by √) from which tensile data are available for each alloy

Reactor/site	A 212B	A 533B	A 350	A 36
HFIR/K7-P5	√			
FNR/60D	√	√		
FNR/60B	√	√		
HFBR/V10-Cd	√	√	√	√
HFBR/V10	√	√	√	√
FNR/60A	√	√		
ORR/P8	√		√	
HFBR/V14	√	√	√	√
HFIR/HT-7	√	√	√	√

A clear influence of thermal neutrons can be seen in Figures 1(a) and 2(a). If the data are compared on the basis of fast fluence only, the data from the highly thermalized V10 site appear to show a shorter threshold dose for hardening than either the V14 or the V10-Cd. Comparing the V10-Cd data with the V14 data indicates that increasing the fast flux by a factor of 120 had no significant effect on the radiation-induced yield strength change. When the data are compared on the basis of dpa in Figures 1(b) and 2(b), the correlation is much improved. In particular, the displacements from thermal neutrons seem to be nearly equivalent to those from fast neutrons. The small residual differences between the A 212 data from the V10 site and the other two data sets are most likely due to the low total dose obtained; note that the change in yield strength is small and can be either positive or negative at doses up to about 5×10^{-4} dpa. Further irradiations are under way to extend the V10 data to higher doses. In addition, the HFBR dosimetry work mentioned above may alter the total number of dpa when a more complete description of the neutron spectrum, including epithermal neutrons, is available.

Figures 3 and 4 extend the A 212B and A 533B plate data to the full range of irradiation conditions used. In the case of the A 212B plate, the V10 irradiations and HFIR surveillance data in Figure 3(a) show similar early increases in strengthening when compared to the other data on the basis of fast fluence. The neglect of displacements caused by thermal neutrons in the V10 has a similar effect to that of neglecting those from gamma rays in the HFIR data. When correlated on the basis of dpa in Figure 3(b), the data show a striking insensitivity to fast flux over the range of 1.75×10^{8} to 4.0×10^{14} n/(cm^2·s), and the V10 and HFIR surveillance data are also well correlated. Similar correlation is observed for the A 533B HSST Plate 02 plate in Figure 4.

Data on the A 350LF3 forging using in the HFIR pressure vessel are shown in Figure 5 and the A 36 structural steel in Figure 6 for a more limited set of irradiation conditions. The yield strength changes in these two alloys are similarly well correlated on the basis of dpa, independent of the fast flux or thermal-to-fast flux ratio.

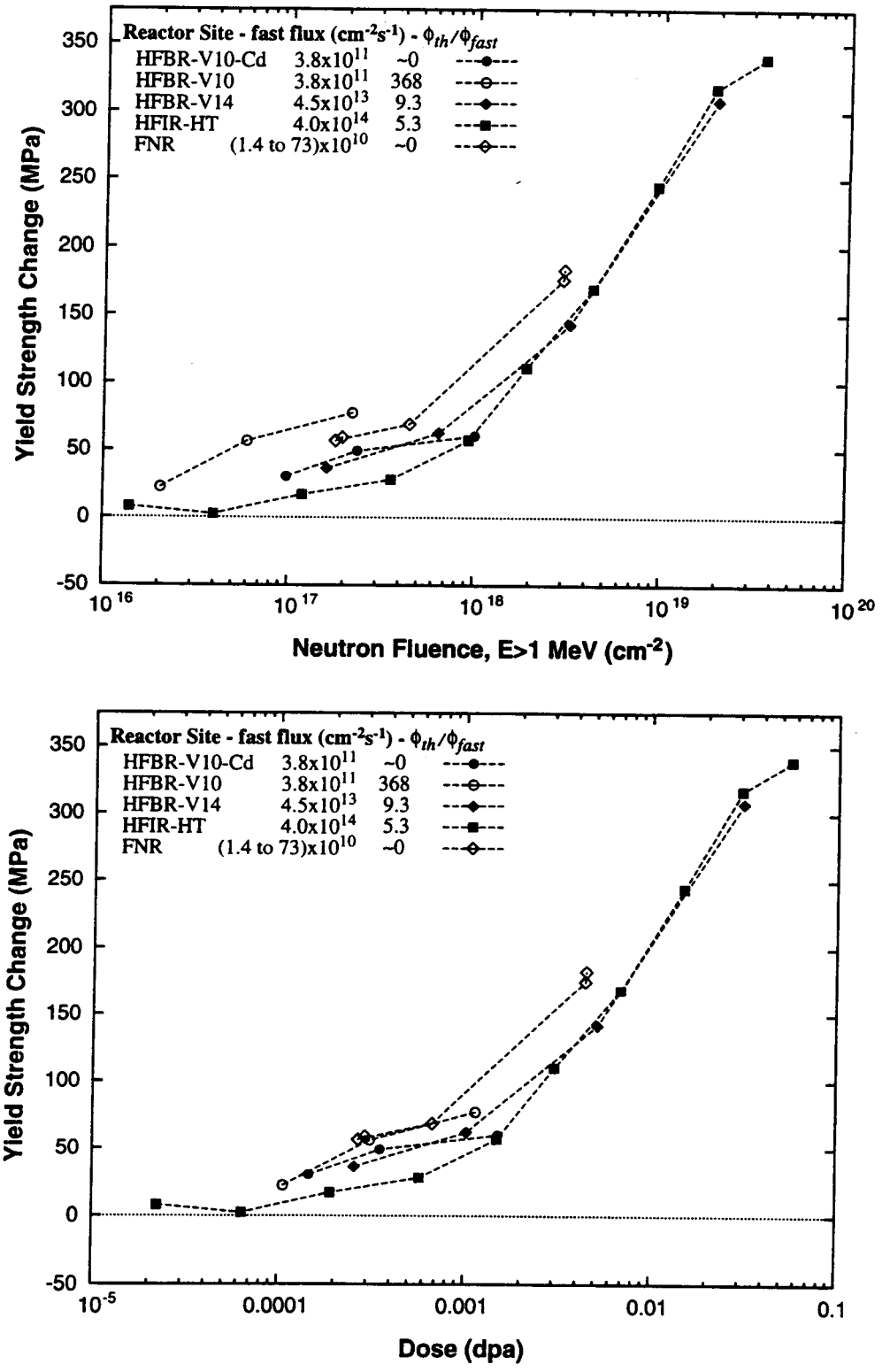

Figure 4. Yield strength change in A 533B correlation monitor irradiated with a range of fast fluxes and thermal-to-fast flux ratios. (a) Neutron fluence dependence. (b) Dose (dpa) dependence.

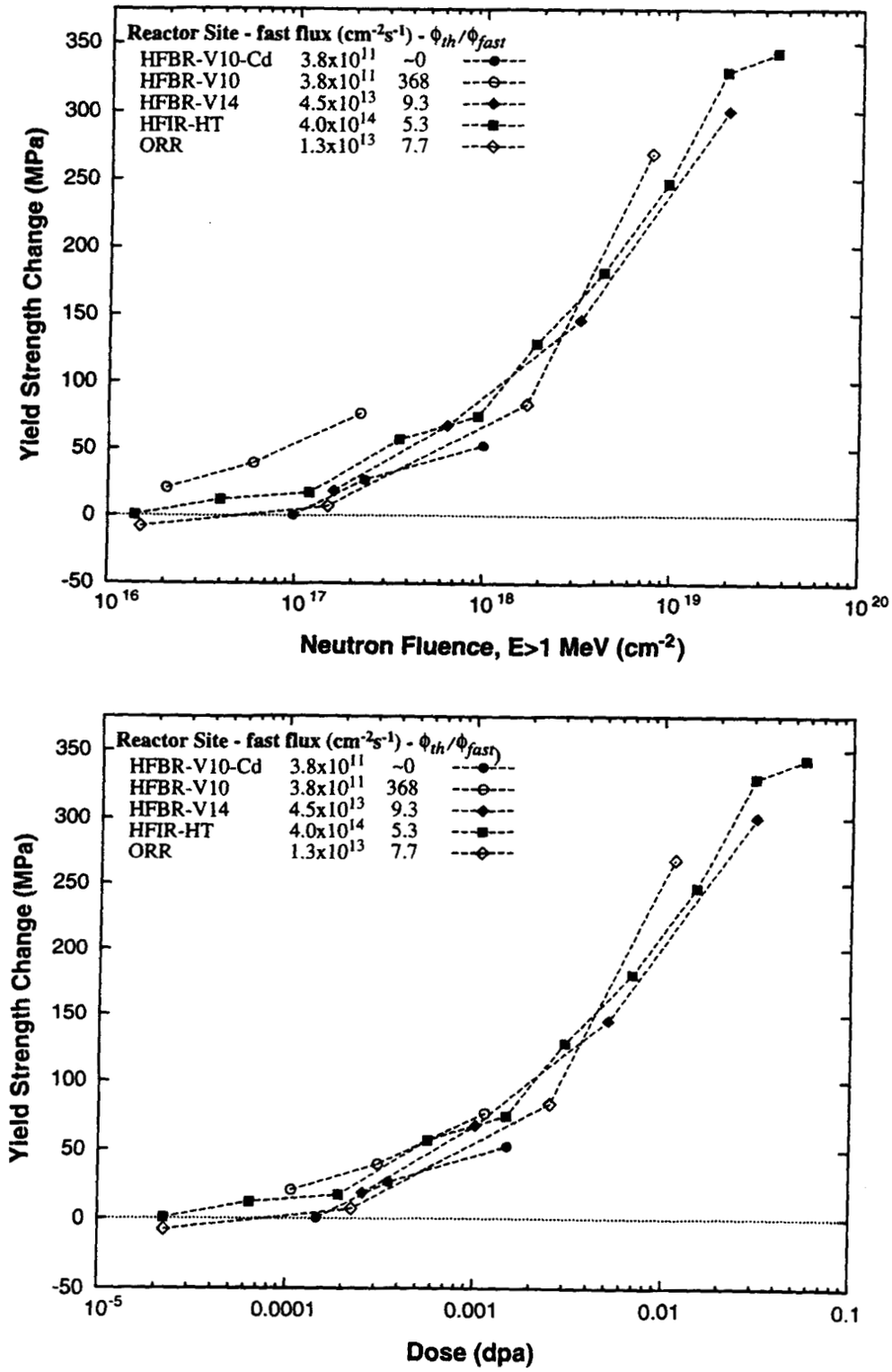

Figure 5. Yield strength change in HFIR A 350LF3 material irradiated with a range of fast fluxes and thermal-to-fast flux ratios. (a) Neutron fluence dependence. (b) Dose (dpa) dependence.

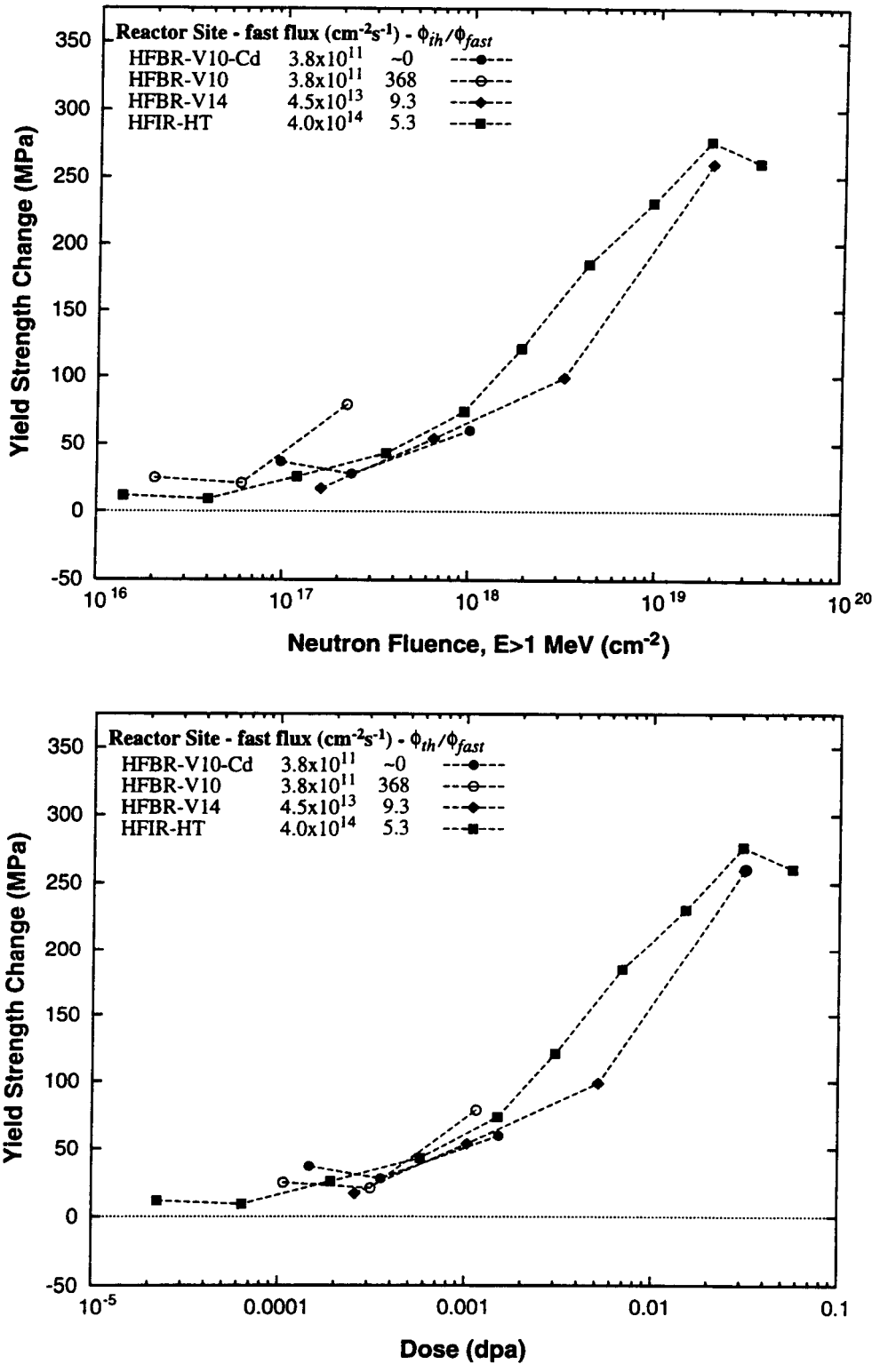

Figure 6. Yield strength change in A 36 structural steel irradiated with a range of fast fluxes and thermal-to-fast flux ratios. (a) Neutron fluence dependence. (b) Dose (dpa) dependence.

A comparison of the data for all four materials shown in Figures 1 to 6 indicates that dpa is an adequate correlation parameter for low-temperature tensile data. The small variations from material to material further support the conclusion that the minor differences between the V10 and either the V10-Cd or V14 data shown in Figure 1(b) for the A 212B are probably insignificant.

SUMMARY

Analysis of the tensile data indicates that radiation-induced yield strength changes at 50 to 60°C in typical pressure vessel and structural steels are essentially independent of the fast flux or displacement rate for the irradiation doses examined here. The fast fluxes in these irradiations ranged from 1.75×10^8 to 4.0×10^{14} n/(cm^2·s) and the peak fluence was 3.7×10^{19} n/cm^2 (0.056 dpa). The absense of a flux effect is consistent with data reported by Odette et al.* A comparison of very high and low thermal-to-fast flux ratios demonstrated the importance of accounting for atomic displacements generated by thermal neutrons. The specimens that were shielded from thermal neutrons showed much less strengthening per unit of fast fluence than the unshielded specimens. However, the results indicate that the effects of thermal neutrons at this temperature can be well accounted for through the use of displacements per atom as a correlation parameter, i.e., the embrittling efficiency of atomic displacements from thermal neutrons appears to be nearly the same as those induced by fast neutrons.

It should not be assumed that these conclusions would uniformly apply to higher temperature irradiation conditions; for example, a stronger dependence on fast flux is known to exist at 288°C for certain combinations of fast flux and fluence.[9] The impact of neutron energy spectrum could also be more important at this higher temperature where strengthening is believed to arise primarily from the formation of small precipitates. The expected higher point defect survival fraction from low-energy cascades could manifest itself by increasing radiation-enhanced solute diffusion. However, these results provide some support for the predictions that a region of relatively weak flux dependence should be observed at 288°C.[9-11]

ACKNOWLEDGMENTS

This work is supported in part by the Division of Materials Sciences, U.S. Department of Energy under contract DE-AC05-96OR22464 with Lockheed Martin Energy Research Corp. The authors would like to thank G. R. Odette and G. E. Lucas from the University of California, Santa Barbara, for including our specimens in their FNR irradiation experiment, which was funded by the Office of Energy Research, U.S. Department of Energy.

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