DOSE-RATE EFFECTS ON IRRADIATION EMBRITTLEMENT AND COMPOSITION AND TEMPERATURE EFFECTS ON ANNEALING/REIRRADIATION SENSITIVITY

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Abstract

Recent MEA investigations on the effect of neutron flux level on radiation-induced embrittlement accrual and the contributions of metallurgical variables to postirradiation annealing and reirradiation behavior are reviewed. Studies of dose-rate effects involved experiments in the UBR test reactor and separately, radiation sensitivity determinations for the decommissioned Gundremmingen (KRB-A) vessel material. Annealing-reirradiation studies employed 399°C and 454°C heat treatments.

Material composition is shown to play a major role in postirradiation annealing recovery. Results illustrate effects of variable copper and variable nickel contents on recovery for steel plate having low phosphorus levels. Composition effects on recovery were also observed for prototypic welds depicting high/low copper and high/low nickel contents and three flux types. The welds, in addition, indicate major differences in re-irradiation sensitivity.

The UBR investigations revealed a significant difference in flux level sensitivity between the ASTM A 302-B reference plate and a submerged-arc (S/A) Linde 80 weld, based on C_v tests and quasi-static fracture toughness (J-R curve) tests. Studies of the Gundremmingen reactor vessel, representing a joint USA-FRG-UK undertaking, revealed an anomaly in strong vs. weak test orientation radiation sensitivity.

1. INTRODUCTION

This report highlights certain MEA research accomplishments for the Nuclear Regulatory Commission (NRC) in calendar year 1986. The research findings build on previous MEA studies of variable radiation embrittlement sensitivity among reactor pressure vessel (RPV) steels

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and the qualification of in-situ annealing for controlling radiation embrittlement accrual in-service (Ref. 1). Investigations reported here focus on the potential effect on metal properties change of irradiation exposure rate or dose rate (neutron flux, n/cm^2-s^{-1}) and secondly, examinations of key variables influencing material behavior with irradiation-annealing (IA) and reirradiation (IAR) treatments.

Investigations made to qualify potential dose-rate effects depict two research thrusts. One involves the irradiation of reference plate and weld materials in a light water cooled and moderated test reactor (the UBR) at three different flux levels. The flux range brackets conditions incident to RPV walls and flux conditions typical of in-core irradiation facilities of many research reactors. The second thrust involves postservice properties and property changes of the pressure vessel of the decommissioned Gundremmingen BWR. This postmorten effort is under the auspices of a joint USA-FRG-UK program; MEA, StaatlicheMaterialPruefungsanstalt (MPA), and Harwell are the lead laboratories for the three respective countries.

MEA studies of the annealing method are qualifying the influence of material composition and heat treatment temperature on annealing recovery and equally important, the influence of both factors on reirradiation sensitivity and reembrittlement path after the anneal. The focus is on weld metal behavior, recognizing that weld deposits may well constitute the limiting material in older reactor vessels in PTS scenarios (Ref. 2).

2. DOSE-RATE EFFECT STUDIES -- UBR INVESTIGATIONS

2.1 Background

The MEA Dose-Rate Effects (DRE) experiment series is designed as a critical test of neutron flux level vs. radiation embrittlement resistance. The uncertainty over the effect of irradiation at high vs. low neutron flux frequently is placed in the framework of power reactor vs. test reactor environments. Flux levels at the inner walls of RPV's are on the order of 10^{10} n/cm^2-s^{-1} whereas levels of 10^{12} n/cm^2-s^{-1} or higher are typical of fuel lattice facilities in test reactors. Resolution of the flux-effect question is essential. Test reactor experiments offer the only feasible means of obtaining large irradiation volumes for systematically testing metallurgical variable or specimen-size effects and the flexibility required for annealing-reirradiation studies.

2.2 Irradiation Facilities

MEA irradiation facilities at the UBR are providing neutron flux levels of $\sim 8 \times 10^{10}$ (low), $5-6 \times 10^{11}$ (intermediate) and $8-9 \times 10^{12}$ (high) n/cm^2-s^{-1}. Facility locations relative to the fuel lattice are illustrated in Fig. 1. In each, specimen temperatures can be controlled, e.g., at 288°C, and are monitored continually by external instrumentation. The in-core (IC) and core-edge (CE) irradiation assemblies employ only gamma heating for temperature maintenance; the in-pool (IP) or reflector irradiation assemblies employ a combination of gamma heating and resistance heaters. Because of the primary dependence on gamma heating, specimen temperatures are automatically reduced at times of reactor outages to well below the nominal irradiation temperature. Thus, specimens are protected against inadvertent annealing of radiation effects between reactor runs.

2.3 Materials and Test Matrices

Materials selected for the DRE experiment series include two high Cu content (~ 0.20% Cu) reference plates, types A 302-B and A 533-B, and two high Cu content (~ 0.39% Cu) submerged-arc weld deposits. The welds were made commercially using the same lot of copper-coated filler wire; only the flux type differed (Linde 80 or Linde 0091). The A 302-B plate is widely known as the ASTM A 302-B reference plate. Compositions of two of the materials are given in Table 1.

Irradiation test matrices are summarized in Table 2. Specimens include Charpy-V (C_v), fatigue-precracked Charpy-V (PCC_v), tensile, and 0.5T compact tension (CT) types. For the A 302-B plate, specimens were oriented to represent the longitudinal (LT-strong) test direction. This direction was selected over the transverse (TL-weak) orientation because of the low preirradiation C_v upper shelf energy level [65 J (48 ft-lb)] of this material. The transverse orientation, on the other hand, was chosen for the higher toughness A 533-B plate, consistent with current surveillance program practices. Weld specimens spanned the deposit in the manner of ASTM Standard Practice E 185.

Table 2 indicates three target fluence levels for the two highest flux conditions. Irradiation to 0.5×10^{19} n/cm^2 at the lowest flux level (8×10^{10} n/cm^2-s^{-1}) represents more than a 3-year residence period in the UBR. Because of this time factor, higher fluence experiments in the low flux facility were not included in the matrix.

2.4 Progress

Table 3 indicates the number of irradiation exposures completed at the time of this report. Postirradiation testing has been completed for the intermediate flux exposures [Core-Edge (CE) experiments] of the A 302-B reference plate and the Linde 80 weld. Both of these materials have a "low" preirradiation C_v upper shelf level, unlike the A 533-B plate and the Linde 0091 weld. Accordingly, these received the greater program emphasis.

C_v data for the A 302-B plate and the Linde 80 weld are presented in Figs. 2 and 3, respectively. Companion fracture toughness determinations are given in Figs. 4 and 5. An immediate observation is that the 4X difference in fluence between experiments CE-1 and CE-3 produced only a small increase in C_v transition temperature elevation

(ΔT) in each case. For example, the ΔT at 41-J for the plate is only 20°C (35°F). That for the weld is only 25°C (45°F). Referring to the fracture toughness determinations, both materials show very little fluence sensitivity in terms of the ΔT at the 100 MPa$\sqrt{m}$ level. The difference in curve shape between CE-1 vs. CE-3 data sets, in part, is due to the computer curve-fitting of the data points. (The C_v curves are visual curve fits.)

Referring to the CE-1 data sets, relatively close agreement of the ΔT for C_v at 41-J and the ΔT for CT at 100 MPa$\sqrt{m}$ is observed for the weld but not the plate. In the case of the plate, the 41-J temperature elevation underpredicts the 100 MPa$\sqrt{m}$ temperature elevation. Application of the Irwin β_{Ic} adjustment (Ref. 3), as recommended by Merkle (Ref. 4), reduces the shift in the toughness transition. This brings the plate data sets into closer agreement. By the same token, the weld data sets diverge, with ΔT from CT exceeding the ΔT from C_v. Hiser has observed this pattern of C_v vs. CT data for many but not all plates and weld deposits in his development of a data bank on RPV materials (Ref. 5).

The results from in-core (high flux) DRE irradiations are not yet available; however, the core-edge data can be compared to data and data trends for the two materials generated by other MEA programs (Table 4). Specific data pairs denote a dose-rate effect which may extend to power reactor surveillance as well as encompass the test reactor case. For example, notice the intermediate flux irradiation of the weld to 6.4×10^{18} n/cm^2 produced an embrittlement level requiring 13.4×10^{18} n/cm^2 under high flux irradiation conditions. Overall, the intermediate flux level appears to be more damaging to the weld than the high flux level. This is not the case with the plate. The intermediate flux level produced significantly less embrittlement than the high flux level at the higher fluences of 12×10^{18} n/cm^2 and 24×10^{18} n/cm^2. (NOTE: Trend data indicate a 41-J increase of 92-97°C for the highest fluence, in-core.) A different sensitivity vs. dose-rate relationship is thus indicated for plate compared to weld deposit.

The results show that a unique relationship between embrittlement and dose rate may not exist for all RPV steels and that earlier comparisons of high flux/low flux environment effects may not have placed sufficient emphasis on product form or composition influences. It is encouraging that the level of effects seen to date is not so large as to preclude the use of accelerated (test reactor) irradiation for screening metallurgical, irradiation, or postirradiation annealing variables.

3. DOSE-RATE EFFECTS STUDIES -- GUNDREMMINGEN INVESTIGATIONS

3.1 Background

The decommissioning of the KRB-A reactor in 1977 and the more recent locating of archive forging material from its vessel construction presents a unique opportunity for qualifying the effects of long-term irradiation on a prototypic steel. Recognizing this possibility, the NRC put in place a joint USA/FRG/UK program to investigate vessel properties in both the as-irradiated and postirradiation-annealed conditions. Among its objectives are verification of prediction methods for radiation effects and prediction methods for the attenuation of radiation effects with steel thickness. Through test method selection, the correlation of notch ductility vs. fracture toughness change with irradiation is also being evaluated experimentally. The vessel composition includes 0.15 Cu, 0.79% Ni, and 0.015% P.

3.2 MEA Program

MEA has developed through-thickness mechanical properties information for the archive material which is being used to index the irradiation effect to the vessel (Ref. 6). Additionally, MEA is performing irradiation and postirradiation annealing assessments of the archive material using C_v, tensile, and 0.5T-CT compact specimens irradiated at the nominal KRB-A vessel service temperature in the UBR.

Results will be compared against vessel properties being determined by MPA from a series of 100-mm diameter trepans removed from the beltline region. Harwell is performing irradiation assessments of the archive material using C_v specimens exposed in the Pluto heavy water test reactor.

3.3 Progress

Irradiation and postirradiation annealing data obtained through the UBR experiments are shown in Figs. 6 through 9. Fracture toughness data for 399°C and 454°C annealed conditions are currently under development.

Referring to Figs. 6 and 9, a comparable effect of irradiation to 41-J and 100 MPa$\sqrt{m}$ index temperatures is indicated (Δ44°C vs. Δ47°C). The accompanying drop in C_v upper shelf energy was small (about 10 J). The projection of 41-J transition temperature increase by NRC Regulatory Guide 1.99, draft Revision 2, for the fluence indicated is 59°C. Accordingly, the Guide is properly conservative in its embrittlement estimate.

Figures 7 and 8 show that a 454°C-168 h anneal but not a 399°C-168 h anneal produces essentially full transition temperature recovery. Full upper shelf recovery was obtained with both heat treatments. An explanation for the higher C_v upper shelf level after the anneal, compared to the preirradiation level, is not available. This may be simply an effect of the time-at-temperature during the irradiation. Thermal aging tests to evaluate this possibility are under consideration.

3.4 Comparison with FRG/UK Findings

The irradiation test results obtained by Harwell, depicting the effects of a heavy water environment, and the trepan test results obtained by MPA, depicting the effects of light water environment service, are very tentative. The preliminary results, however, are quite interesting.

In the case of the UBR vs. Pluto test reactor irradiations, the data show a general agreement in embrittlement sensitivity. On the other hand, the UBR test reactor data agree well in terms of damage magnitude, only with the trepan longitudinal orientation data. The UBR data considerably underpredict the trepan transverse orientation irradiation behavior.

A major difference in radiation sensitivity level between LT and TL orientations of the trepans has been found by MPA (Ref. 7) and currently is anomalous. If the LT vs. TL orientation difference is confirmed by the ongoing test programs, this observation will have a major impact on the application of Regulatory Guide 1.99 which has made extensive use of LT orientation data in its development, especially in the formulation of upper shelf behavior projections with fluence.

4. ANNEALING/REIRRADIATION STUDIES

4.1 Background

Material composition is expected to play a major role in postirradiation annealing recovery, both for welds and plate materials. The significance of this variable to annealing response is being probed by two MEA investigations. One set of investigations, coded Composition Effects on Annealing or CEA, is studying the annealing response of plate materials initially acquired for investigations of material sensitivity to 288°C radiation embrittlement and underlying damage mechanisms. The second group of investigations, identified as the High Temperature Annealing studies or HTA, is evaluating the annealing and reirradiation embrittlement behavior of weld deposits. Here, postirradiation annealing temperatures of 454°C vs. 399°C are being employed to gain insight into composition effects as well as annealing temperature effectiveness.

4.2 Progress by CEA Investigations

The CEA investigations center on 288°C irradiation and 399°C-168 h postirradiation annealing tests of two material groups identified in Table 5. Findings for group 1 were presented to the 1985 WRSM (Ref. 1); a primary determination was that residual embrittlement (after 399°C annealing) is a function of copper content but not phosphorus content. Determinations for material group 2 are summarized in Fig. 10.

Referring to the results for the plates from the high copper content melt no. 6, a detrimental effect of a high nickel content (0.69% Ni) on both radiation resistance and annealing recovery (residual embrittlement) indicated by cast B vs. cast C. Percentage recoveries for the two plates are about the same, however (56°F vs. 54%). Results for the plates from melt casts A and B do not describe a detrimental effect of nickel for the range of 0.05% to 0.27% Ni. This range is within the % Ni specification limits for A 302-B steel. Accordingly, nickel would appear critical to the performance of A 533-B steel (e.g., 0.69% Ni) and not A 302-B steel.

Results for plates from melt casts C and D of melt no. 5 show an effect of nickel content, but here the differences are less than those noted for melt no. 6 because of the lower copper content (0.16%). Consistent with the findings for materials group 1, a greater percentage recovery and smaller residual embrittlement is found for the lower of the two copper contents.

Summarizing CEA experiment observations in terms of residual embrittlement, copper and nickel but not phosphorus can exert a detrimental influence on notch ductility recovery of RPV steels.

4.3 Progress by HTA Investigations

Four prototypic submerged-arc (S/A) welds were obtained for the investigations. These represent the major weld types found in USA reactors and depict two copper levels (intermediate and high), two nickel content levels (low and high), and the use of three flux types. The materials are identified in Table 6. The irradiation matrix is illustrated in Table 7. Results are now available for all exposure conditions except that designated IAR_2.

Figure 11 summarizes the findings on 41-J transition temperature increase after irradiation and the residual 41-J transition temperature increase after annealing at 454°C or 399°C. The duration of the anneal, in each case, was 168 h. Samples of the four welds were commingled during the irradiation and again during postirradiation heat treatment. Figure 12 and 13 show the effects of reirradiation. The former shows the amount of the increase produced by the second cycle of irradiation <u>only</u>; the latter shows the total increase observed following the IAR treatment.

Several observations are permitted by the data. In general, the welds experienced different degrees of recovery, illustrative of a composition dependency of some type. Also, depending on the weld, the transition temperature recovery with 454°C annealing was or was not significantly greater than that by 399°C annealing. However, the extent of reembrittlement of the welds is consistently less following the 454°C anneal. In turn, the irradiation-anneal-reirradiation (IAR) benefit appears to be greatest with this anneal.

In Fig. 13, the difference in fluence between the first cycle irradiation data and the IAR_1 irradiation data must be kept in mind when judging the benefits of 399°C annealing. In this framework, a beneficial effect of IAR is described in each instance. Also, consistent with earlier experiment observations, the sensitivity to reirradiation embrittlement is high compared to material that received the same fluence but which has not been annealed.

Referring to Table 7, one objective of experiment IAR_2 is the definition of reembrittlement path with increasing fluence. The potential of the individual weld types for reirradiation embrittlement saturation will be tested by comparing this set of data against data for IAR_1.

4.4 Conclusions of CEA and HTA

The cited findings reinforce the original NRC concerns that composition effects and interactions must be understood to assess plant-specific annealing applications. Properly applied, IAR procedures may be a means of avoiding PTS limitations and a means for overall plant-life extension. A 454°C anneal, although more difficult to apply from an engineering and systems viewpoint, holds forth high promise for embrittlement mitigation and control.

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Table 1 Compositions of ASTM A 302-B Reference Plate and MEA Linde 80 Reference Weld

Material	MEA Code	Composition (wt-%)								
		Cu	Ni	P	C	Mn	Si	S	Cr	Mo
A 302-B Plate	23F	0.21	0.20	0.013	0.24	1.34	0.23	0.023	0.11	0.51
Linde 80 Weld	W8A	0.39	0.67	0.015	0.09	1.29	0.76	0.015	0.11	0.48

Table 2 Irradiation Matrix - UBR Series

Target Fluence[a] ($\times 10^{19}$)	High Flux (8×10^{12})[b]	Intermediate Flux (6×10^{11})	Low Flux (8×10^{10})
0.5	X	X	X
1.0	X	X	-
2.0	X	X	-
	(In-Core)	(Core-Edge)	(Reflector)

[a] n/cm^2, $E > 1$ MeV [b] $n/cm^2\text{-}s^{-1}$, $E > 1$ MeV

Table 3 Status of Irradiations[a] - UBR Series

Material	Target Fluence ($\times 10^{19}$)	Neutron Flux: High	Intermediate	Low
A 302-B	0.5	C	C	C
	1.0	C	C	-
	2.0	C	C	-
Linde 80	0.5	C	C	U
	1.0	C	C	-
	2.0	C	C	-
A 533-B	0.5	P	-[b]	C
Linde 0091	0.5	P	-[b]	U

[a] C = Complete U = Underway P = Planned [b] NRC Option

Table 4 Intermediate vs. High Flux Results

Fluence ($\times 10^{18}$)	Flux ($\times 10^{11}$)	ΔT (41-J)	Irradiation Facility
ASTM A 302-B Reference Plate (0.21% Cu)			
5.5	85	65°F	In-Core
6.5	6	85°F	Core-Edge
12.0	85	140°F	In-Core
12.8[a]	6	95°F	Core-Edge
Linde 80 Submerged-Arc Weld (0.39% Cu)			
6.4	6	225°F	Core-Edge
13.4	85	215°F	In-Core
12.8[a]	6	235°F	Core-Edge

[a] Preliminary estimate

Table 5 Materials - CEA Investigations

Irradiation Assembly Number	Melt	Plate	% Cu	% P	% Ni
1	1	67B	0.002	0.015	0.70
		67C	0.002	0.025	0.70
	2	68A	0.30	0.003	0.70
		68B	0.30	0.016	0.70
		68C	0.30	0.028	0.70
2	5	5C	0.16	0.002	0.27
		5D	0.16	0.002	0.68
	6	6A	0.28	0.002	0.05
		6B	0.28	0.002	0.27
		6C	0.28	0.002	0.69

Table 6 Weld Material Identification

Weld Code	Linde Welding Flux	% Cu	% Ni	% P
WW7	80	0.35	0.10	0.013
W8A	80	0.39	0.67	0.015
W9A	0091	0.41	0.70	0.010
WW4	124	0.16	0.65	0.013

Table 7 Irradiation Matrix

(I)	288°C Irradiation $(\sim 1.3 \times 10^{19})$
(IA)	Anneal 454°C-168 h (Group 1)
	Anneal 399°C-168 h (Group 2)
(IAR_1)	288°C Reirradiation $(\sim 0.3 \times 10^{19})$
(IAR_2)	288°C Reirradiation $(\sim 0.7 \times 10^{19})$

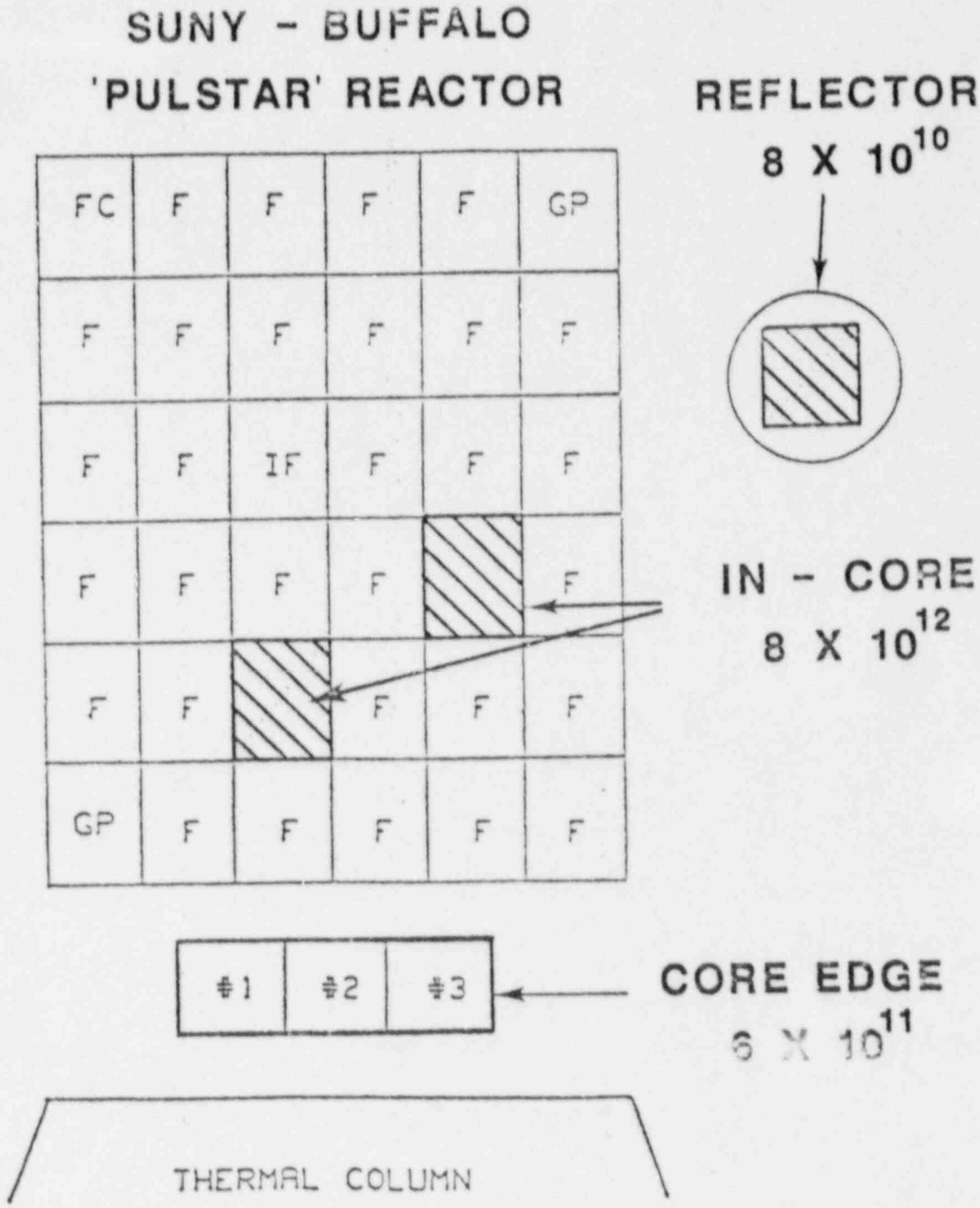

Fig. 1 MEA irradiation facility locations in UBR reactor.

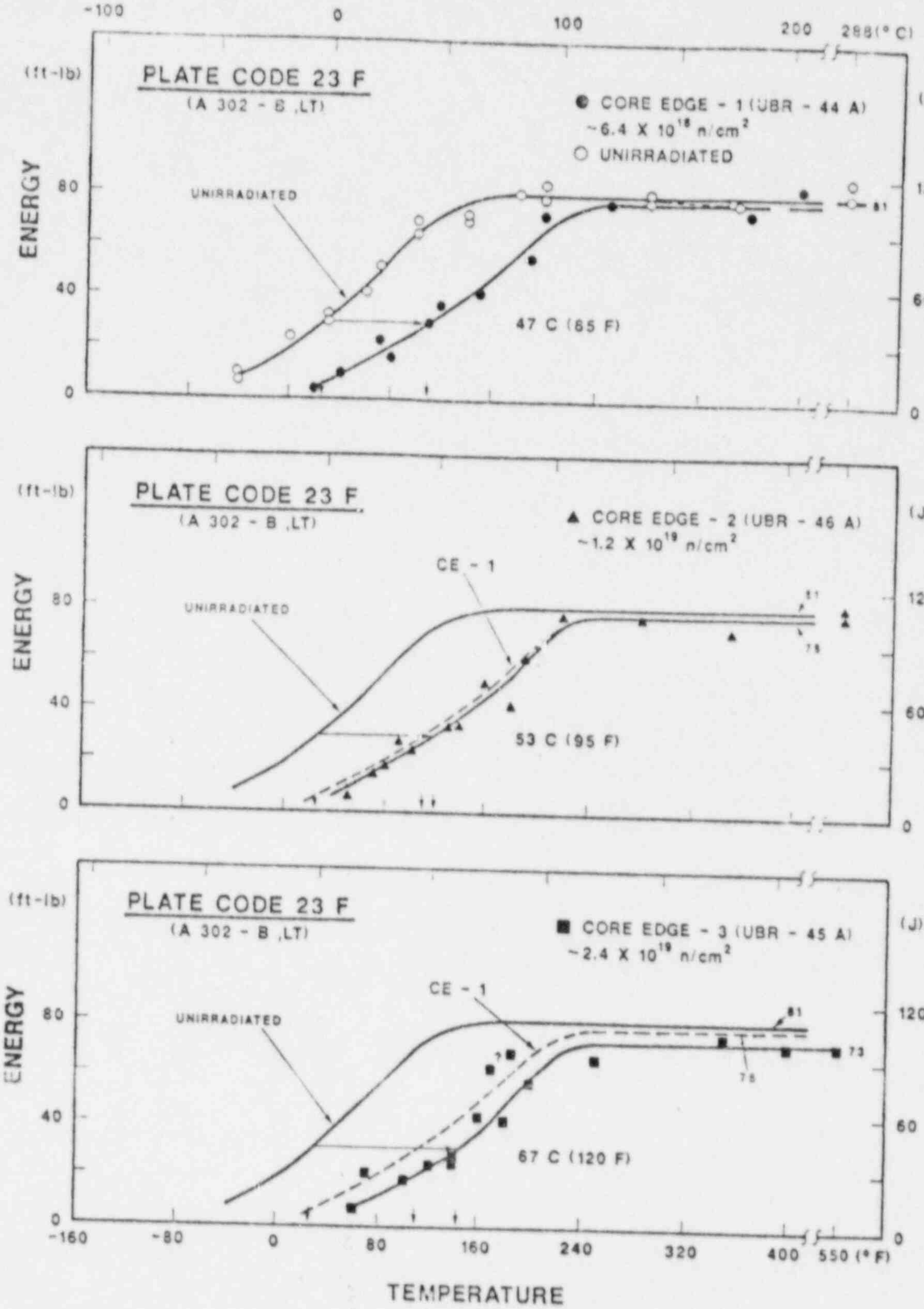

Fig. 2 C_v notch ductility of ASTM A 302-B reference plate after 288°C irradiation at intermediate flux level (Core-Edge Assemblies no. 1, 2, and 3).

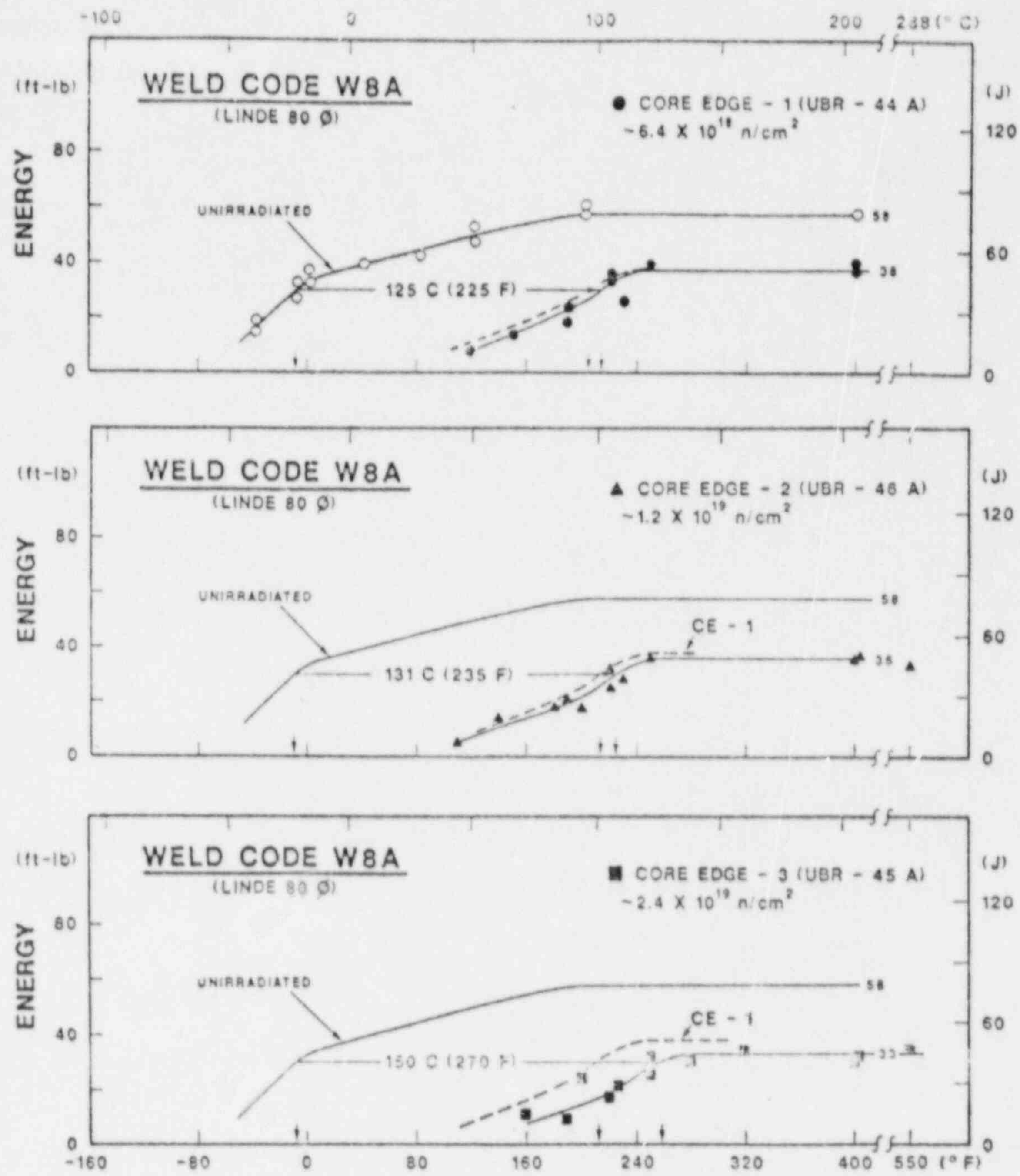

Fig. 3 C_v notch ductility of MEA reference weld (Linde 80 flux) after 288°C irradiation at intermediate flux level (Core-Edge Assemblies no. 1, 2, and 3).

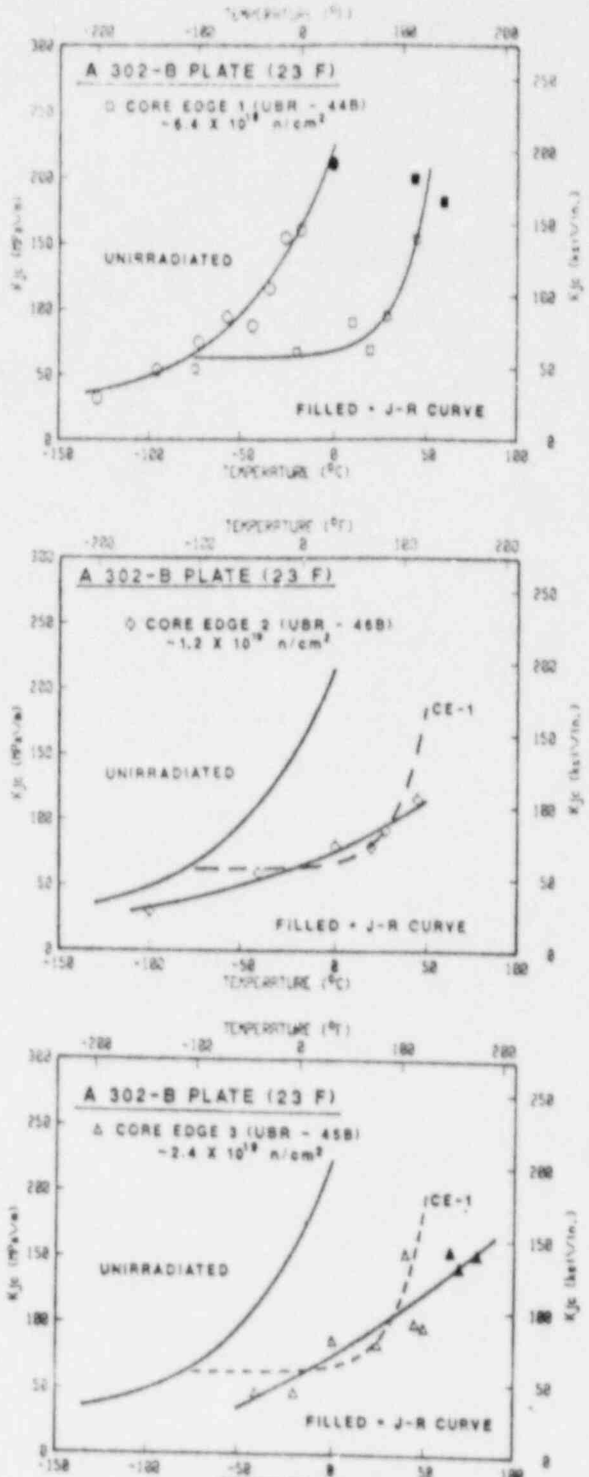

Fig. 4 Fracture toughness of A 302-B reference plate after 288°C irradiation at intermediate flux level (Core-Edge Assemblies no. 1, 2, and 3). **PRELIMINARY DATA.**

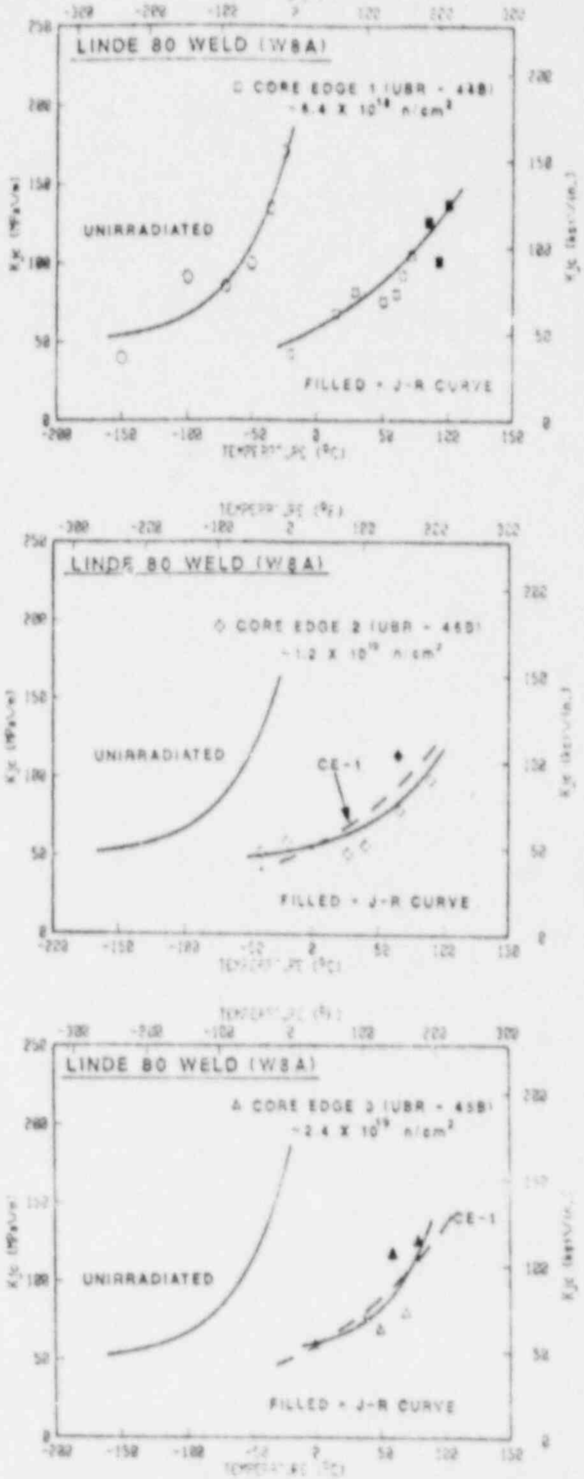

Fig. 5 Fracture toughness of reference weld (Linde 80 flux) after 288°C irradiation at intermediate flux level (Core-Edge Assemblies no. 1, 2, and 3). PRELIMINARY DATA.

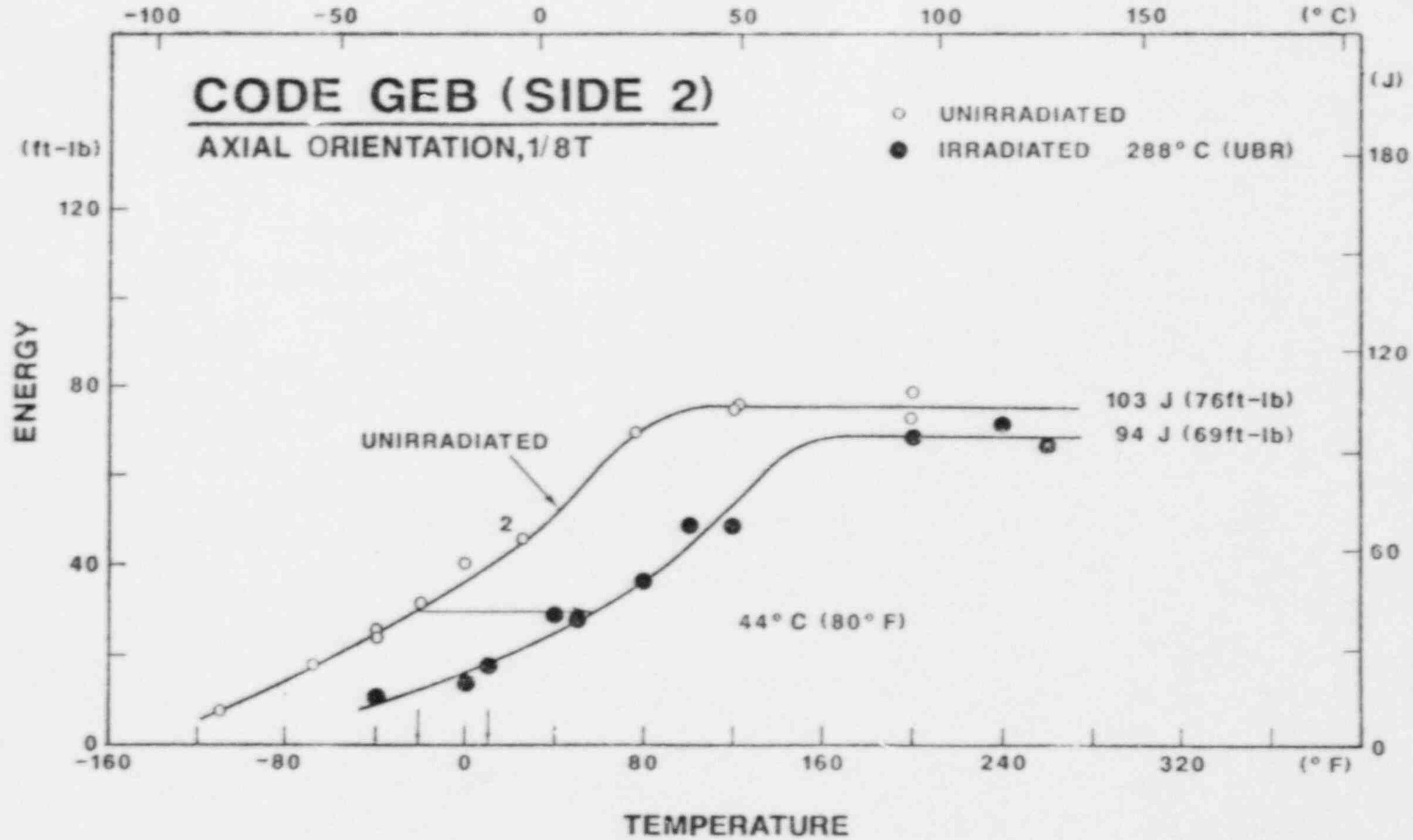

Fig. 6 C_v notch ductility of archive ring forging, GEB, before and after 288°C irradiation to 8.8×10^{18} n/cm^2, E > 1 MeV.

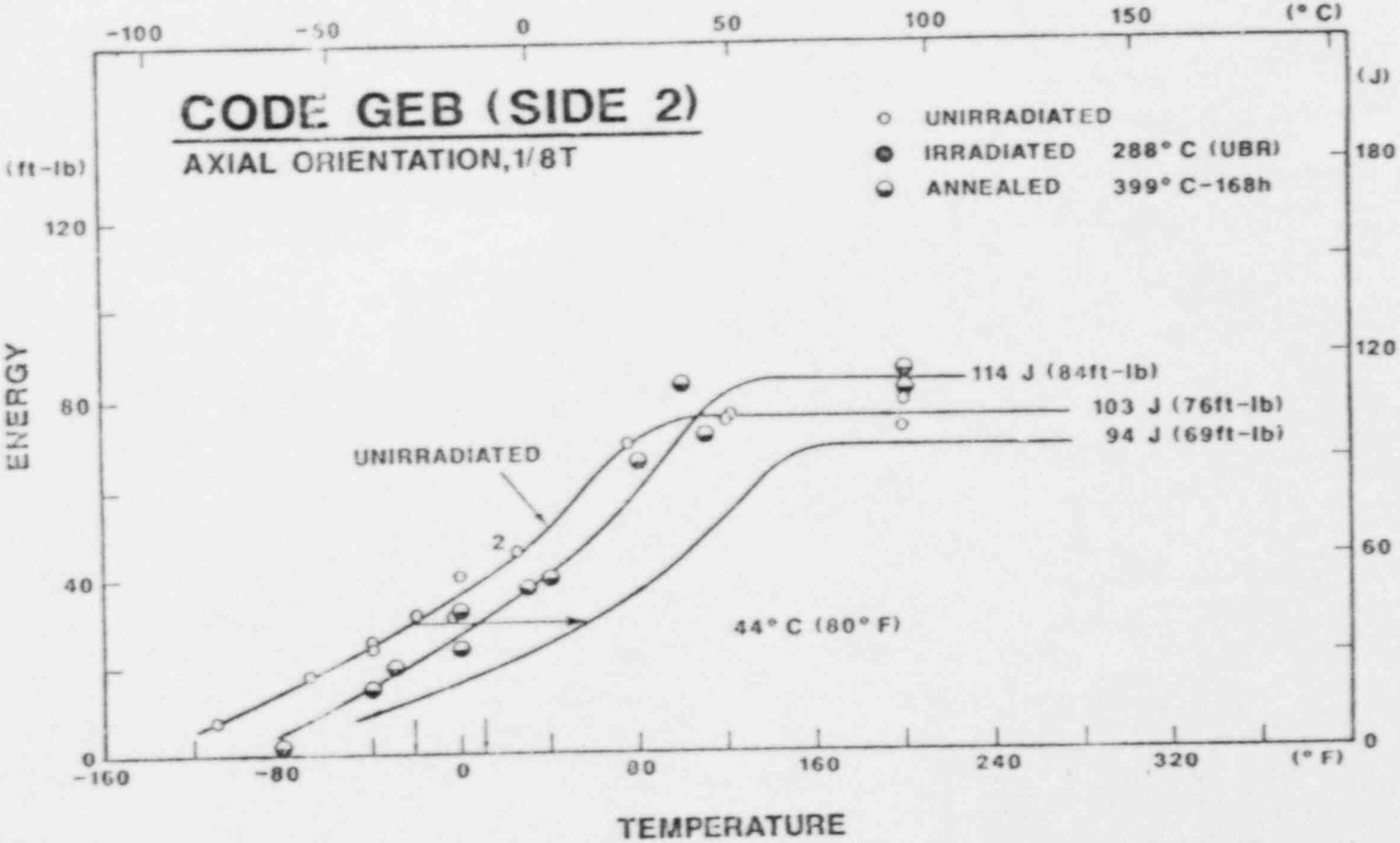

Fig. 7 C_v notch ductility of archive ring forging, GEB, after 288°C irradiation and after postirradiation annealing at 399°C for 168 h.

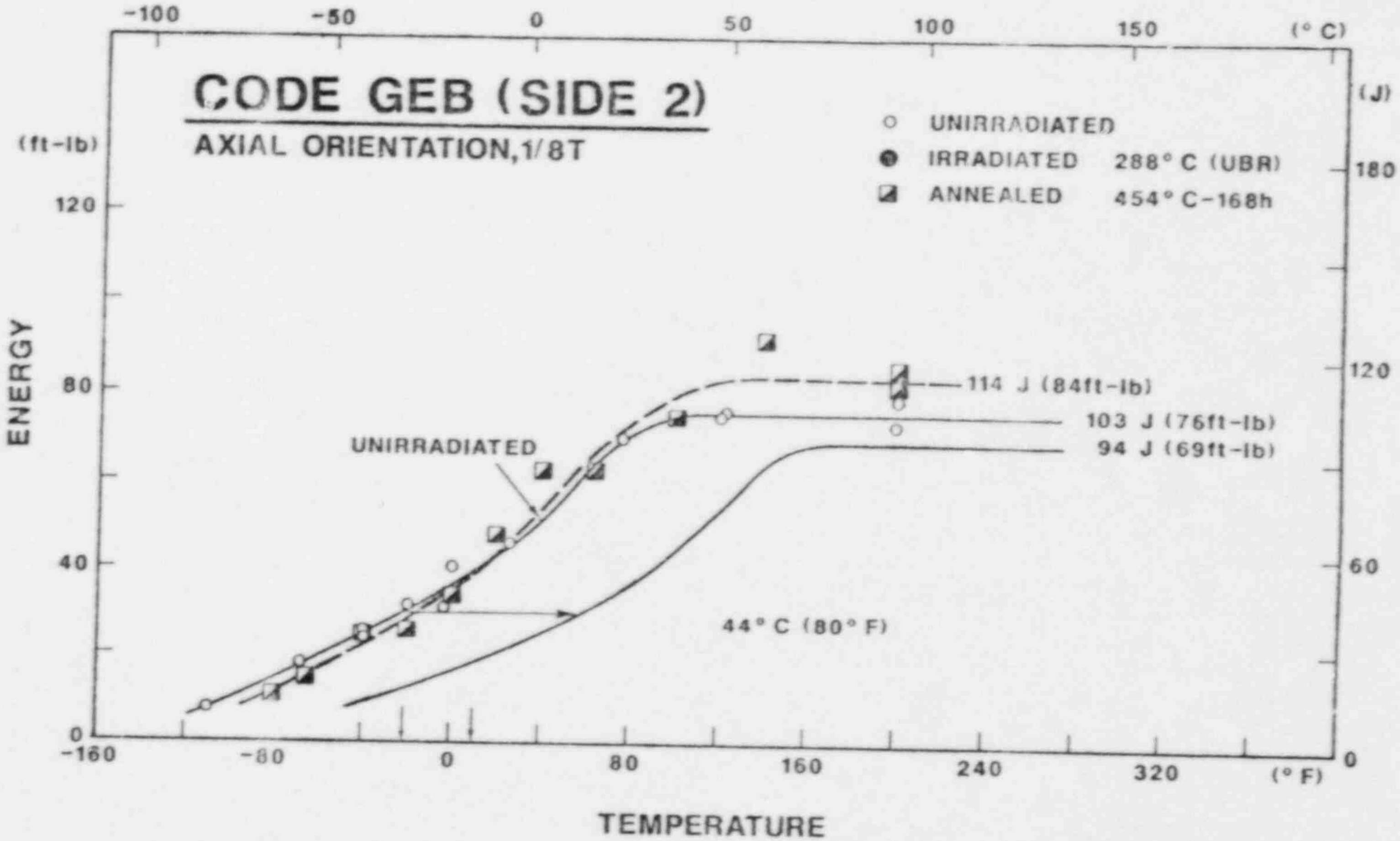

Fig. 8 C_v notch ductility of archive ring forging, GEB, after 288°C irradiation and after postirradiation annealing at 454°C for 168 h.

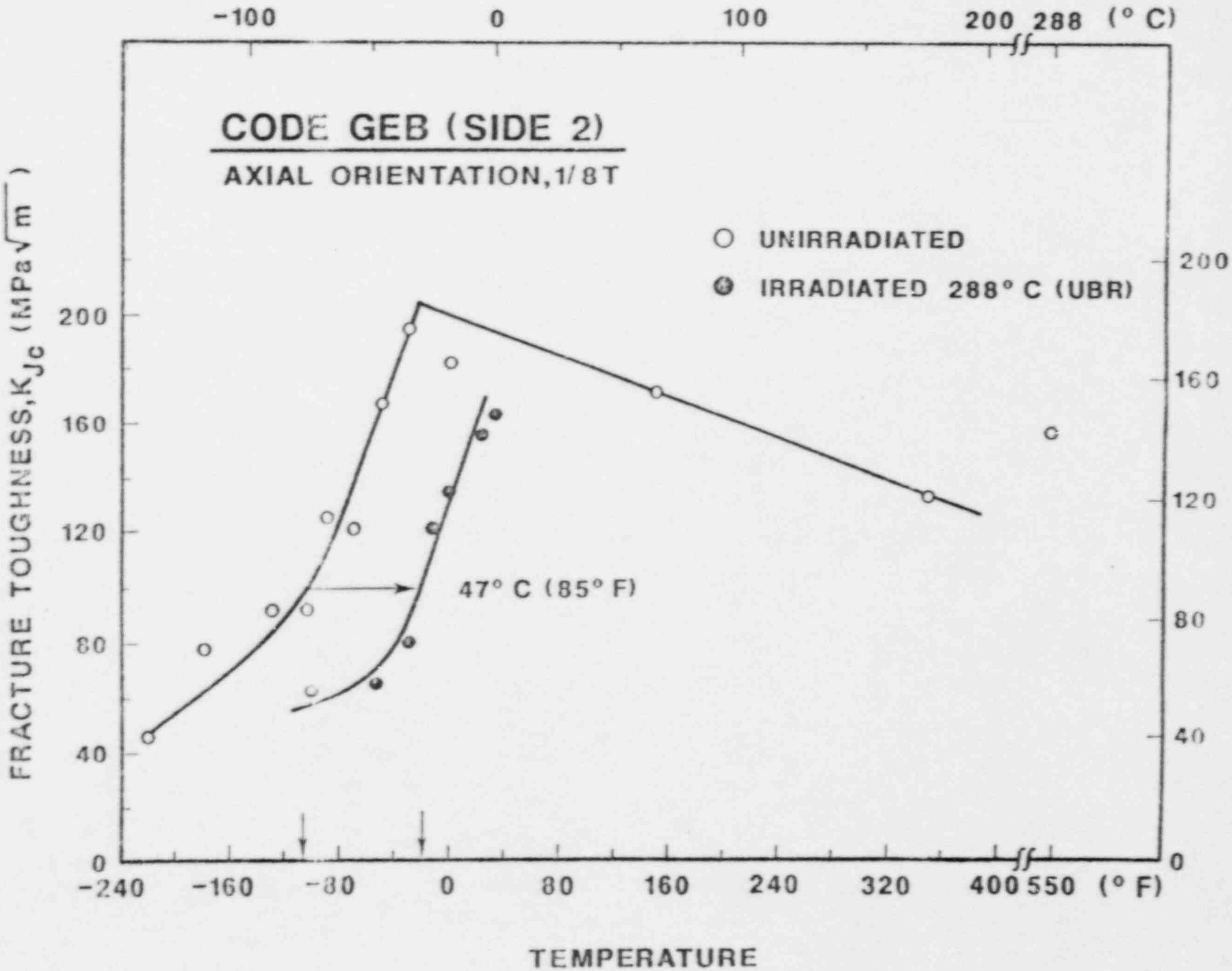

Fig. 9 Fracture toughness of archive ring forging, GEB, before and after 288°C irradiation to

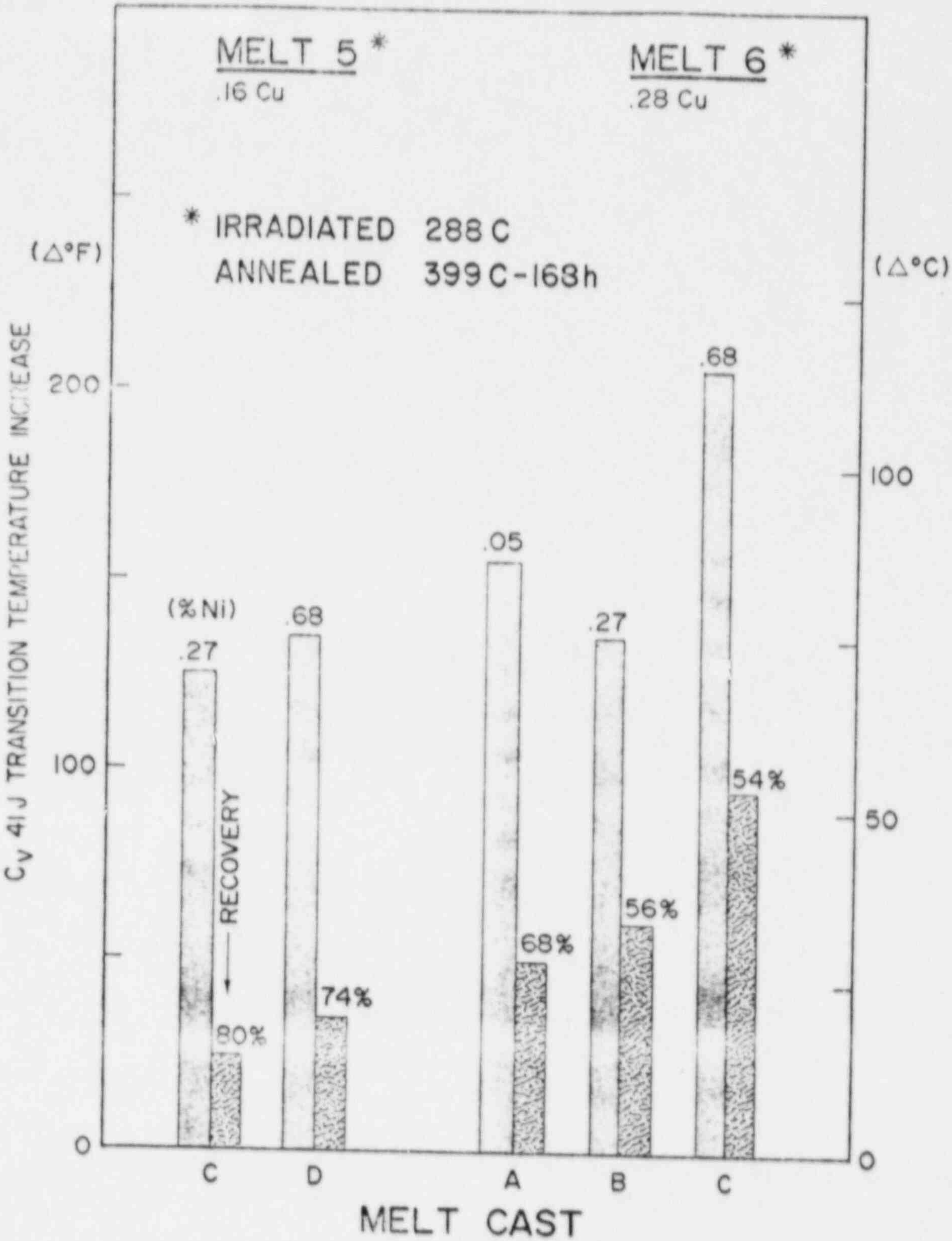

Fig. 10 C_v 41-J transition temperature changes for A 302-B plates (casts A and B) and A 533-B plates (cast C) with 288°C irradiation (left hand bars) and with 399°C-168 h postirradiation annealing (right hand bars).

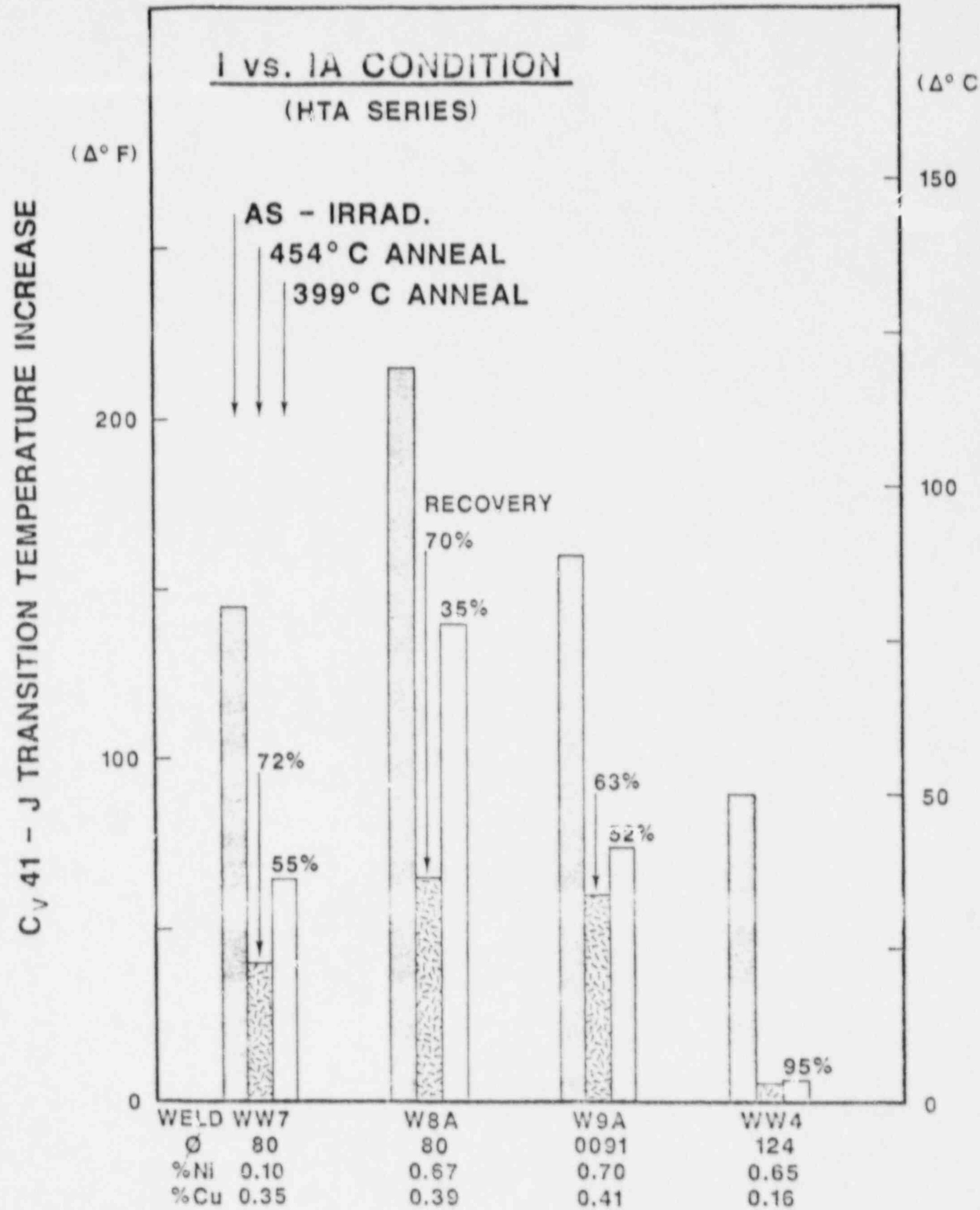

Fig. 11 C_v 41-J temperature changes for S/A welds after 288°C irradiation (left hand bars) and recovery by postirradiation annealing at 454°C for 168 h (middle bars) or 399°C-168 h (right hand bars).

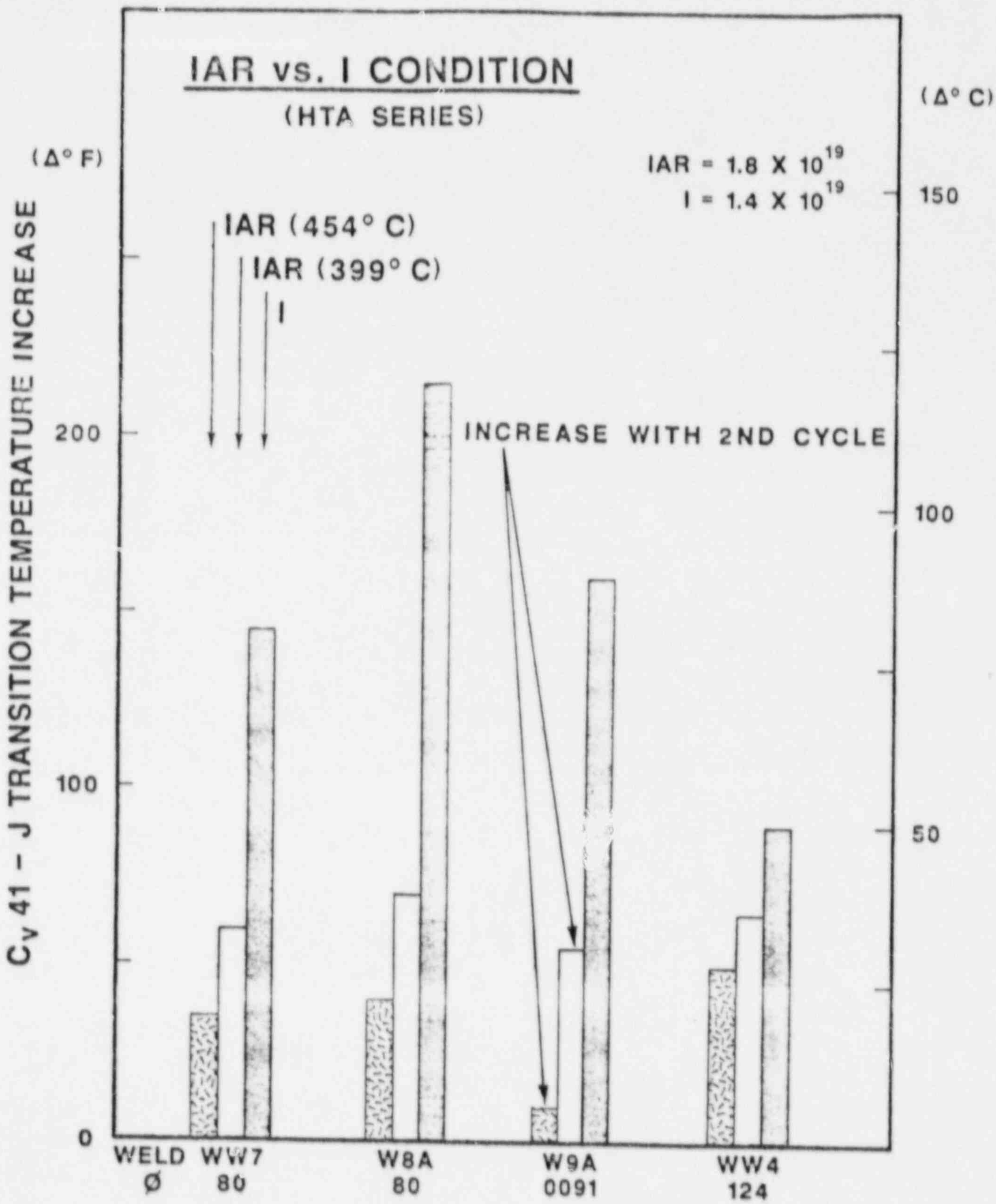

Fig. 12 C_v 41-J temperature changes for S/A welds produced (only) by 288°C reirradiation after a 454°C or 399°C anneal. The solid bars show the embrittlement of the welds before the anneal for reference.

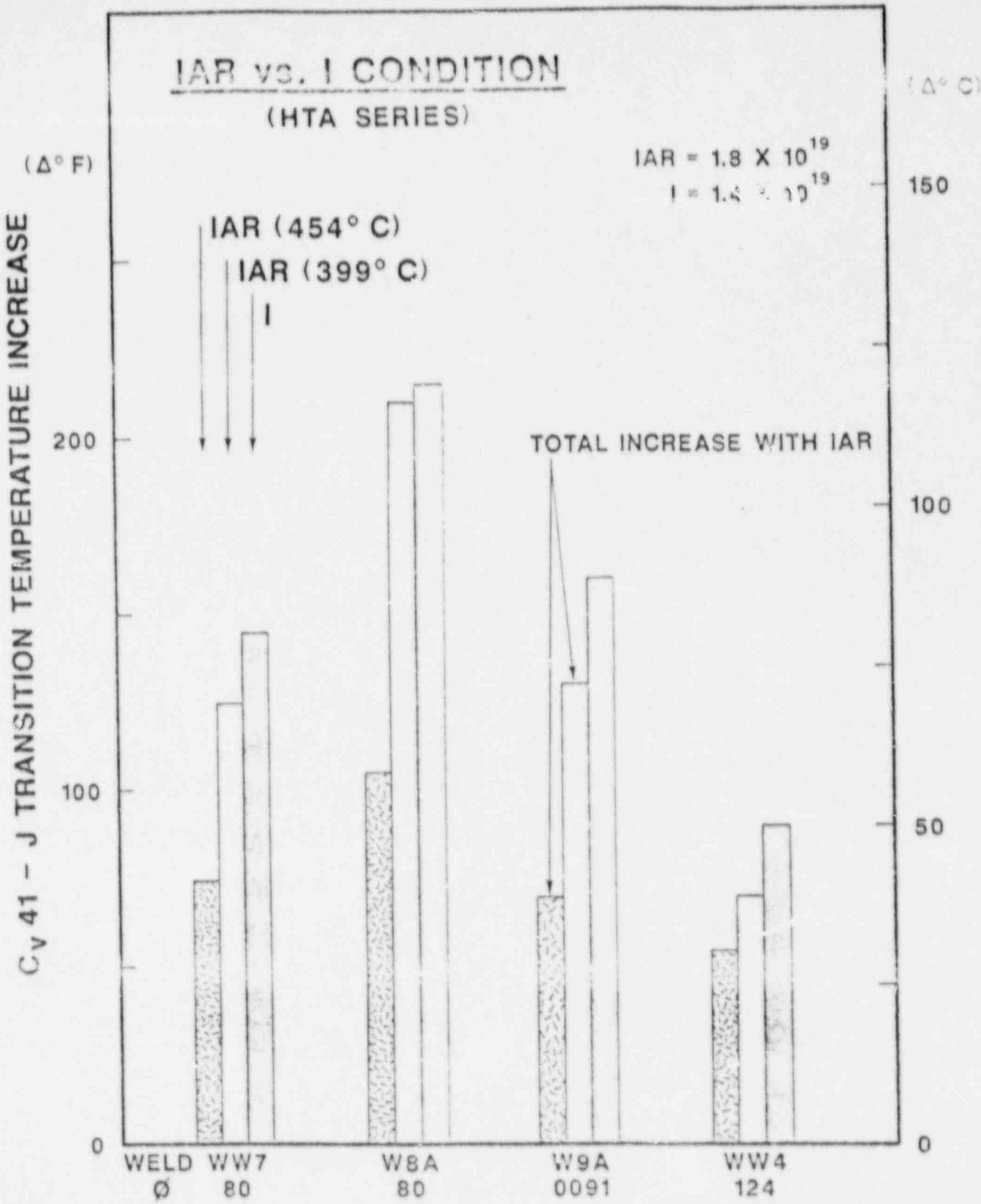

Fig. 13 Total C_v 41-J temperature ch ges for S/A welds produced by both cycles of irradiation with interm di , 454°C or 399°C annealing. The solid bars show the 41-J increase by first cycle of irradiation for reference.

NUREG/CP-0082
Vol. 2

Proceedings of the U.S. Nuclear Regulatory Commission

Fourteenth Water Reactor Safety Information Meeting

Volume 2
- Research in Nondestructive Evaluation
- Environmental Effects in Primary System Components
- Pressure Vessel Research
- Irradiation Effects on RPV Steels
- Degraded Piping Research

Held at
National Bureau of Standards
Gaithersburg, Maryland
October 27-31, 1986

U.S. Nuclear Regulatory Commission

Office of Nuclear Regulatory Research

Proceedings prepared by
Brookhaven National Laboratory