NOTCH DUCTILITY DEGRADATION OF LOW ALLOY STEELS WITH LOW-TO-INTERMEDIATE NEUTRON FLUENCE EXPOSURES

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INTRODUCTION

The embrittlement of reactor structural steels by neutron fluence typically is assessed from changes in Charpy-V (C_v) notch ductility. The progressive increase in embrittlement with neutron exposure is known to be non-linear with fluence (n/cm^2 > 1 MeV) and, in addition, has been shown to be dependent on the content of copper and phosphorus impurities in the steel. Through C_v data compilations and analyses, radiation embrittlement trends have been evolved for application in reactor vessel design and for guidance of fracture safe vessel operation. One example of the development of embrittlement versus fluence curves is the U.S.Nuclear Regulatory Commission Guide 1.99. While significant progress has been made, trend development efforts have been greatly impeded by a lack of radiation data for the extremes of the fluence range of interest. Projected end-of-life (EOL) fluences for many currently operating reactor vessels are on the order of 3 to 5 x 10^{19} n/cm^2. A large volume of data exists for 2 to 4 x 10^{19} n/cm^2 fluences; however, data for exposures below this fluence interval are scarce.

This study was undertaken with the objective of improving knowledge of both the extent and trend of C_v notch ductility changes in reactor vessel steels (plate and weld metals) at low-to-intermediate fluences, that is, between 0.1 and 1 x 10^{19} n/cm^2. A second objective was to obtain an experimental assessment of the level of conservatism in current embrittlement projection methods. A complete report on the study is given in reference (1)

MATERIALS

Several commercially produced plates and weld deposits were selected for the investigation. The materials are identified by NRL code number and composition in Table 1 and are fully representative of reactor vessel materials now in service. The A302-B plate is also the ASTM reference correlation-monitor steel used extensively in reactor surveillance programs.

Material irradiations (Table 2) were conducted at a nominal 288°C (550°F) at the State University of New York at Buffalo using its 2 MW pool reactor (UBR). Irradiation temperatures were monitored by multiple thermocouples in each specimen array. The ambient neutron flux was on the order of 7 X 10^{12} n/cm^2 - sec.

RESULTS

Experimental results from the study are summarized in Tables 3 and 4. Selected examples of individual material assessments are given in Figures 1 through

3. Figure 4 shows a limited comparison made of radiation effects to C_v notch ductility and dynamic fracture toughness (K_J) obtained by the fatigue precracked C_v test method.

Trend of Upper Shelf Reduction at Low-to-Intermediate Fluence

Analysis of the data relating percentage upper shelf reduction and fluence in the low-to-intermediate fluence interval suggests a relationship of the form:

$$\text{Upper Shelf Reduction (\%)} = A\,(\Phi^{CS})^n$$

where A and n are constants for a material. The value of A varies with material and is an indicator of material radiation sensitivity. The exponent, n, on the other hand, appears relatively material independent and has a value of about 2.3. Welds NRL 2 and NRL 8 which exhibitied the largest upper shelf reduction of the materials at 4×10^{18} n/cm^2 depart from the primary data pattern. In these two cases, lower values of n are required to fit the data. Investigations to further explore the trend relationship are in progress.

Data Comparison Against Regulatory Guide 1.99 Projections

Graphs given by NRC Regulatory Guide 1.99 for the projection of C_v notch ductility changes with fluence are reproduced in Figs. 5 and 6. Both figures take into account the role of impurity copper and phosphorus contents in radiation sensitivity development. One objective of the present study was to test the conservatism of Guide projections in the low-to-intermediate fluence range, recognizing that only sparse data were previously available for defining property-change limits in this interval. Entry of the new data on the graphs indicates that the Guide may be overly conservative in projecting upper shelf reductions at low fluences and at intermediate fluences. A lesser degree of overconservatism is noted for its transition temperature projections.

In Fig. 5, the data suggest that upper shelf change might be more accurately described by a set of bilinear curves such as used to project transition temperature behavior. (Fig. 6) Similar evidence from surveillance programs, however, would be required before new curves could be promulgated. Unfortunately, surveillance data appear to be more scattered in this format and are illustrative of a broad range of performance at low fluences.

In Fig. 7, measured and projected transition temperature increases are compared. Here, a broad scatter is evident, particularly at the higher embrittlement levels. The source of the scatter has not been established but may be due partially to the lack of a term for nickel content in the projection formula. That is, data are available which indicate that nickel content can cause a reinforcement of the primary copper content effect on radiation sensivity.

In summary, the new data suggest that current penalties for low fluence service could be reduced provided that reasons for inconsistencies in surveillance data become understood. The possibility for a reduced penalty appears to be greatest for upper shelf energy properties.

CONCLUSIONS

1. At low fluences of 1 to 2 x 10^{18} n/cm^2, high radiation sensitive steels begin to exhibit changes in notch ductility. Transition temperature elevations in excess of 11°C (20°F) but not upper shelf reductions were observed at this fluence level.

2. Fluences of 3 to 4.5 x 10^{18} n/cm^2 produced transition temperature elevations of 39 to 100° C (70 to 180° F) and upper shelf reductions of 0 to 15%.

3. At intermediate fluences of 6.5 to 9.5 x 10^{18} n/cm^2, large transition temperature elevations of 69 to 133° C (125 to 240° F) and large reductions in upper shelf energies of 15 to 44% were observed. One weld exhibited 49 J maximum after 7.8 x 10^{18} n/cm^2.

4. For low-to-intermediate fluences, a relationship of upper shelf reduction to fluence of the form, Reduction (%) = $A\ (\Phi^{cs})^n$, is suggested by the data for most of the materials. The value of A varies with material radiation sensitivity; the value of n is relatively material independent.

5. NRC Regulatory Guide 1.99 may be overly conservative in projecting upper shelf reduction at fluences less than 5 x 10^{18} n/cm^2. A lesser degree of overconservatism was indicated by the data for transition temperature projection.

6. The trend of log percent upper shelf reduction with log neutron fluence appears best described by a set of bilinear curves.

7. A correlation between transition temperature elevations measured by dynamic PCC$_v$ and C$_v$ test methods appears possible from initial experimental comparisons.

REFERENCES

[1] Notch Ductility Degradation of Low Alloy Steels With Low-To-Intermediate Neutron Fluence Exposures, NUREG,/CR 1053, NRL Report 8357, (in publication).

Table 1

TEST MATERIALS[a]

Material	NRL Code	Cu	P	C	Mn	Ni	Mo
A302-B Plate	F26	.20	.011	.24	1.34	.18	.51
A533-B Plate	N27	.13	.008	.17	1.21	.56	.50
	EBB	.10	.009	.19	1.28	.61	.55
	EDB	.14	.009	.20	1.31	.62	.59
	3MU	.12	.011	.20	1.26	.56	.45
A533-B S/A Weld	MY	.36	.015	.14	1.38	.78	.55
	W1	.35	.020	.09	1.45	.57	.39
	NRL 1	.19	b	-	1.43	.56	.36
	NRL 2	.29	-	-	1.56	.65	.39
	NRL 3	.30	-	-	1.53	.68	.40
	NRL 4	.16	-	-	1.51	.58	.37
	NRL 5	.39	-	-	1.60	.58	.39
	NRL 6	.16	-	-	1.49	.58	.39
	NRL 7	.27	-	-	1.42	.56	.36
	NRL 8	.32	-	-	1.51	.68	.39
	W[c]	.29	.020	.09	1.50	.62	.37
	62N(1)[c]	.18	.019	.08	1.55	.55	.38
	62N(2)[c]	.24	.013	.08	1.42	.50	.37
	63N[c]	.31	.017	.10	1.62	.69	.42

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[a] See reference for additional information

[b] Not determined

[c] Approximate composition

TABLE 2

IRRADIATION EXPERIMENTS

EXP. NO.	TARGET FLUENCE ($\times 10^{18}$ n/cm^2)	MATERIALS
1	1	PLATES A,B, WELD 1
2	5	PLATE B, WELD 1
3	7	WELDS 2,3,4
4	10	WELD 3
5	1	PLATES C,D, WELD 5
6	3	WELDS 6 TO 12
7	6	WELDS 6 TO 12
8	11	WELD 13
9 (Cv,PCCv)	18	PLATE E

Table 3

Summary of Charpy-V Notch Ductility Changes With 288°C (550°F) Irradiation

Material/Code	Fluence	Transition Temperature												Upper Shelf				
	10^{18} n/cm^2	Unirradiated				Irradiated								Unirradiated		Irradiated		
	(>1 MeV)	C_v 41J		C_v 68J		C_v 41J				C_v 68J				J	ft-lb	J	ft-lb	ΔJ
	(ϕ ^{cm})	°C	°F	°C	°F	°C	°F	Δ°C	Δ°F	°C	°F	Δ°C	Δ°F					
A302-B Plate																		
F26	1.2	-18	0	10	50	4	40	22	40	32	~90	22	40	117	86	117	86	0
A533-B Plate																		
N27	1.2	-62	-80	-43	-45	-62	-80	0	0	-40	-40	0	0	187	138	187	138	0
	6.6	-62	-80	-43	-45	-34	-30	28	50	-9	15	33	60	187	138	171	126	16
	21.0[a]	-62	-80	-43	-45	16	60	78	140	46	115	89	160	187	138	~134	~99	53
EBB	1.3	-12	10	13	55	≤2	≤35	≤14	≤25	29	85	17	30	137	101	137	101	0
EDB	1.3	1	30	21	70	18	65	19	35	41	105	19	35	160	118	160	118	0
3MU	17.0	-1	30	29	85	43	110	44	80	77	170	47	85	138	102	136	100	~0
S/A Weld																		
MY	1.2	-34	-30	-15	5	-4	25	31	55	21	70	36	65	145	107	142	105	~0
	6.6	-34	-30	-15	5	99	210	133	240	124	255	139	250	145	107	103	76	42
	26.0[a]	-34	-30	-15	5	141	285	175	315	174	345	189	340	145	107	76	56	69
W1	1.4	-29	-20	2	35	2	35	31	55	-	-	-	-	94	69	94	69	0
NRL 6	16.0	-23	-10	7	45	86	190	111	200	107	225	100	180	107	79	77	57	30
NRL 1[b]	4.2	-15	5	4	40	27	80	42	75	82	180	78	140	107	79	103	76	4
	9.5	-15	5	4	40	66	150	81	145	104	220	89	160	107	79	90	66	17
NRL 2	4.2	-34	30	41	105	57	135	58	105	96	205	56	100	104	77	91	67	13
	9.5	-34	30	41	105	93	200	94	170	129	265	89	160	104	77	81	60	23
NRL 3	3.9	-9	15	27	80	57	135	67	120	93	200	67	120	100	74	96	71	4
	8.5	-9	15	27	80	85	185	94	170	121	250	94	170	100	74	81	60	19
NRL 4[b]	3.6	-18	0	18	65	27	80	44	80	46	115	28	50	98	72	90	66	8
	7.6	-18	0	18	65	52	125	69	125	77	170	58	105	98	72	76	56	22
NRL 5	3.9	16	60	52	125	-1	30	53	95	113	235	61	110	83	61	83	61	0
	9.0	16	60	52	125	88	190	72	130	127	260	75	135	83	61	79	58	4
NRL 7	3.2	-26	-15	7	45	13	55	39	70	49	120	42	75	111	82	106	78	5
	6.7	-26	-15	7	45	43	110	69	125	79	175	72	130	111	82	90	66	21
NRL 8[b]	3.9	-9	15	24	75	63	145	72	130	93	200	69	125	106	78	90	66	16
	9.0	-9	15	24	75	91	195	100	180	116	240	92	165	106	78	79	58	27
W	8.9	-29	-20	-1	30	60	140	89	160	-	-	-	-	104	77	65	48	39
62N(1)	8.9	-29	-20	4	40	60	140	89	160	102	215	97	175	103	76	79	58	24
	12.1	-29	-20	4	40	71	160	100	180	-	-	-	-	103	75	68	50	35
63N	7.8	4	40	~38	~100	135	275	131	235	-	-	-	-	87	64	49	36	36

[a] Prior data.

[b] Upper shelf values taken at 160°C (320°F).

TABLE 4

RESULTS

1 TO 2 $\times 10^{18}$

- ΔTT 14 TO 31C (25 TO 55F)
- ΔUSE NONE APPARENT
- FUNCTION OF COPPER CONTENT

3 TO 4.5 $\times 10^{18}$

- ΔTT 39 TO 100C (70 TO 180F)
- ΔUSE 0% TO 15%
- USE BELOW 68J (2 WELDS)
- NONCONFORMANCE OF ONE WELD

6.5 TO 9.5 $\times 10^{18}$

- ΔTT 69 TO 133C (125 TO 240F)
- ΔUSE 15% TO 44%
- USE REDUCED TO 68J (1 WELD)

> 10 $\times 10^{18}$

- INCREASING EMBRITTLEMENT WITH INCREASING FLUENCE

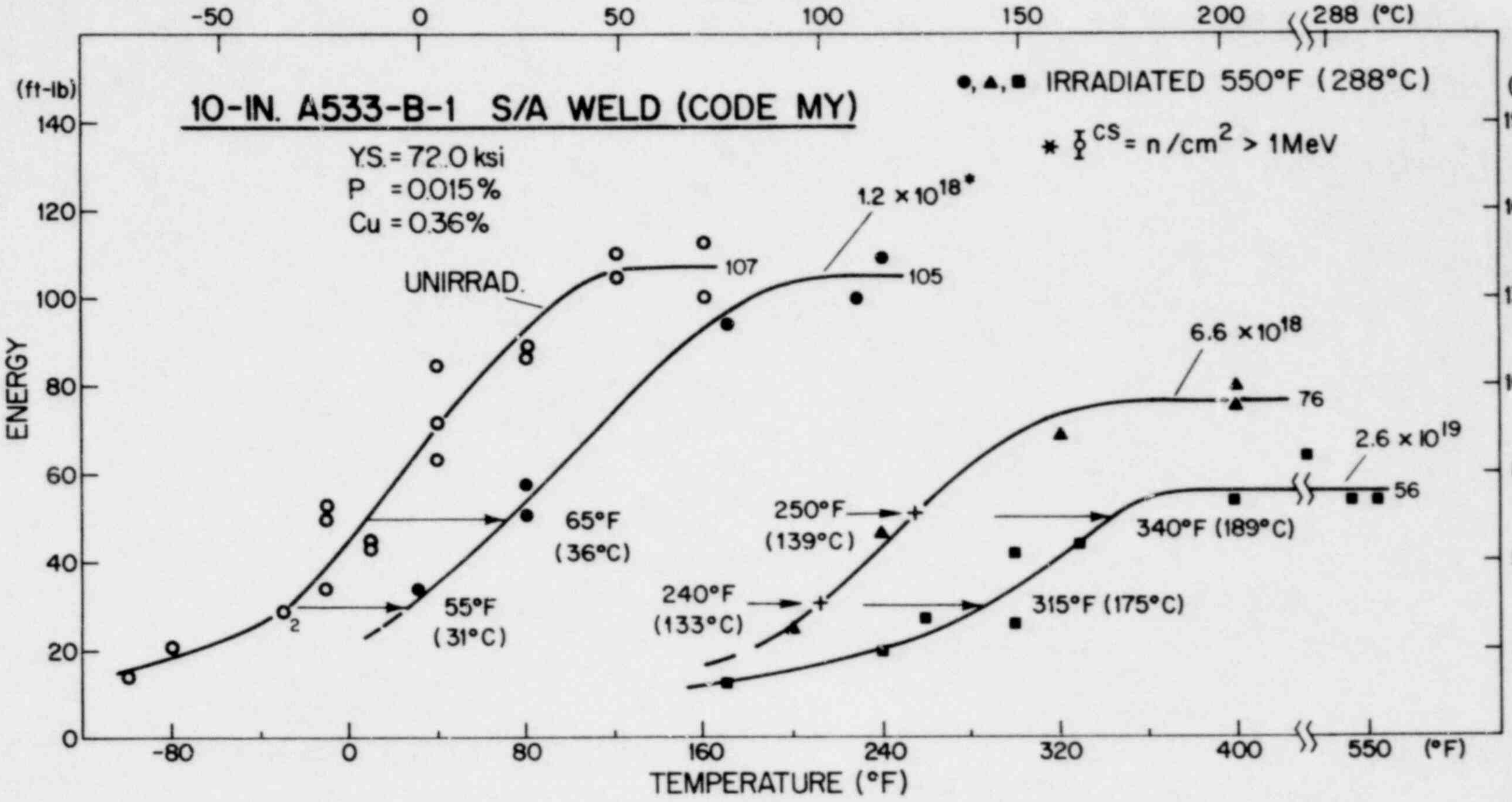

10-IN. A533-B-1 S/A WELD (CODE MY)
Y.S. = 72.0 ksi
P = 0.015%
Cu = 0.36%
●, ▲, ■ IRRADIATED 550°F (288°C)
* Φ CS = n/cm² > 1MeV
UNIRRAD.
107
1.2 × 10^18*
105
6.6 × 10^18
76
2.6 × 10^19
56
65°F (36°C)
55°F (31°C)
250°F (139°C)
240°F (133°C)
340°F (189°C)
315°F (175°C)
2
ENERGY
(ft-lb)
0
20
40
60
80
100
120
140
(J)
0
27
54
81
108
136
163
190
TEMPERATURE (°F)
-80
0
80
160
240
320
400
550
(°F)
-50
0
50
100
150
200
288
(°C)

FIGURE 1

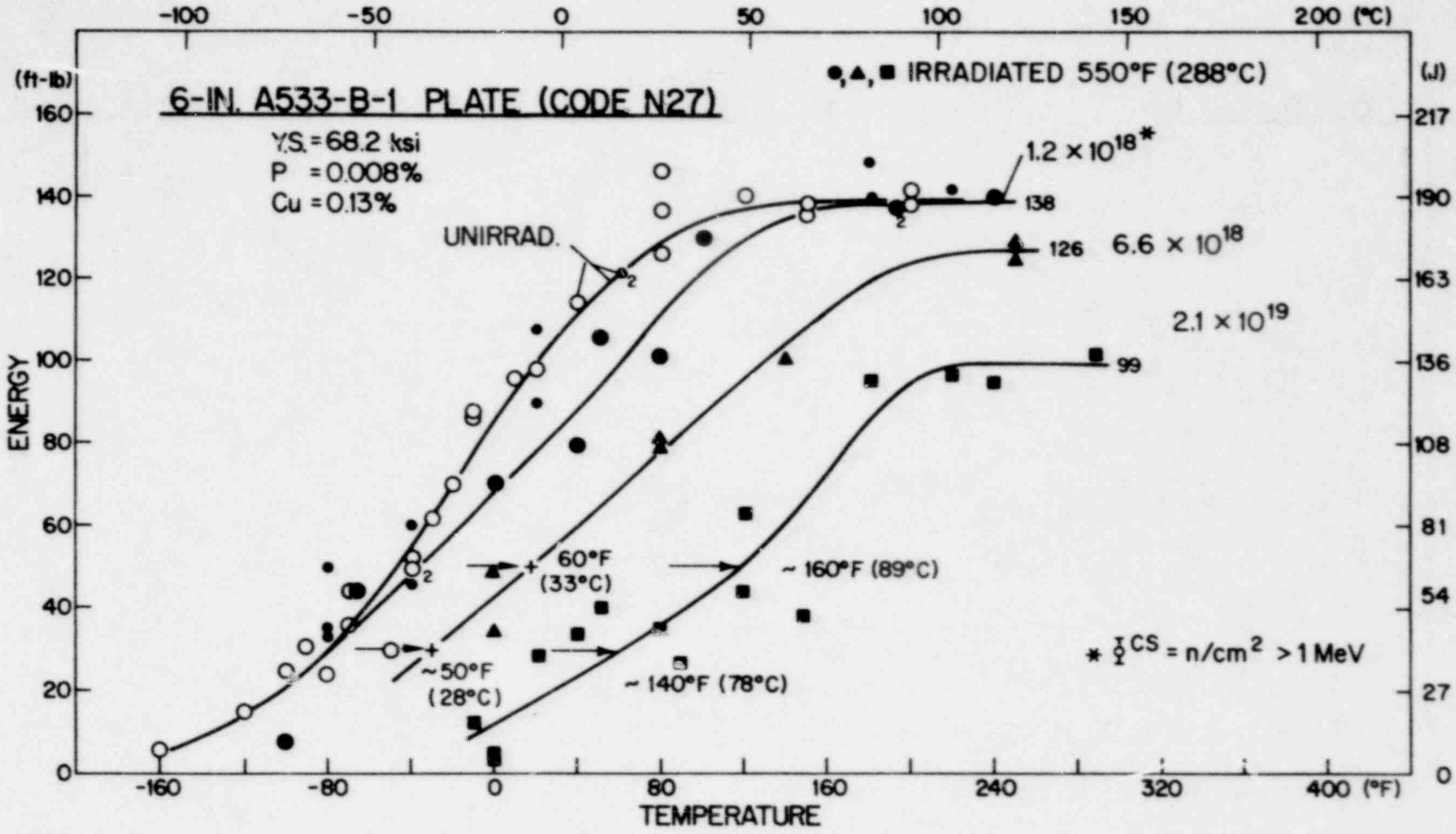

6-IN. A533-B-1 PLATE (CODE N27)
Y.S. = 68.2 ksi
P = 0.008%
Cu = 0.13%
●, ▲, ■ IRRADIATED 550°F (288°C)
UNIRRAD.
2
1.2 × 10^18*
138
6.6 × 10^18
126
2.1 × 10^19
99
60°F
(33°C)
~ 160°F (89°C)
~50°F
(28°C)
~ 140°F (78°C)
* ϕt^CS = n/cm² > 1 MeV
(ft-lb)
ENERGY
(J)
TEMPERATURE
-160
-80
0
80
160
240
320
400 (°F)
-100
-50
0
50
100
150
200 (°C)
0
20
40
60
80
100
120
140
160
0
27
54
81
108
136
163
190
217

FIGURE 2

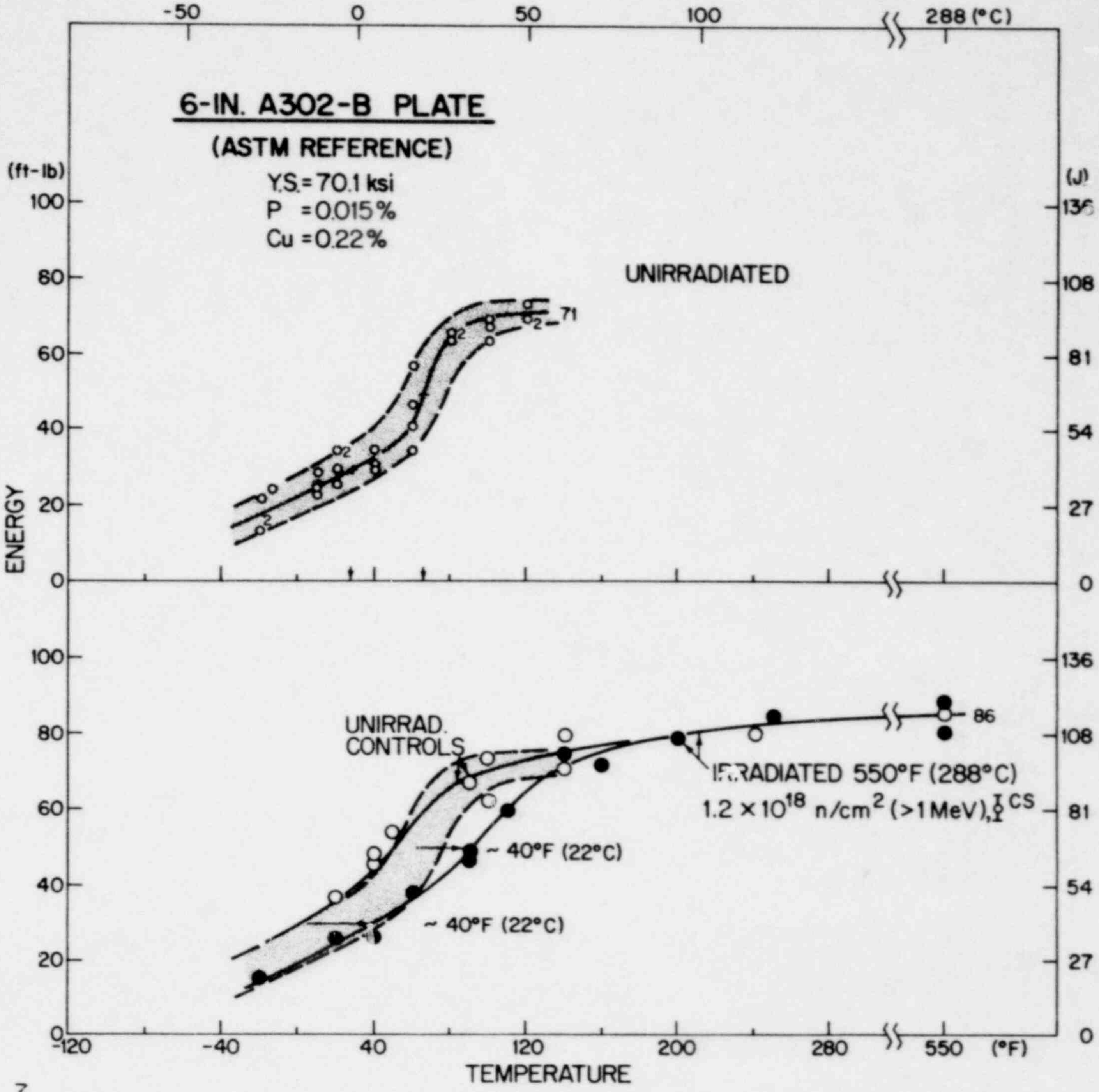
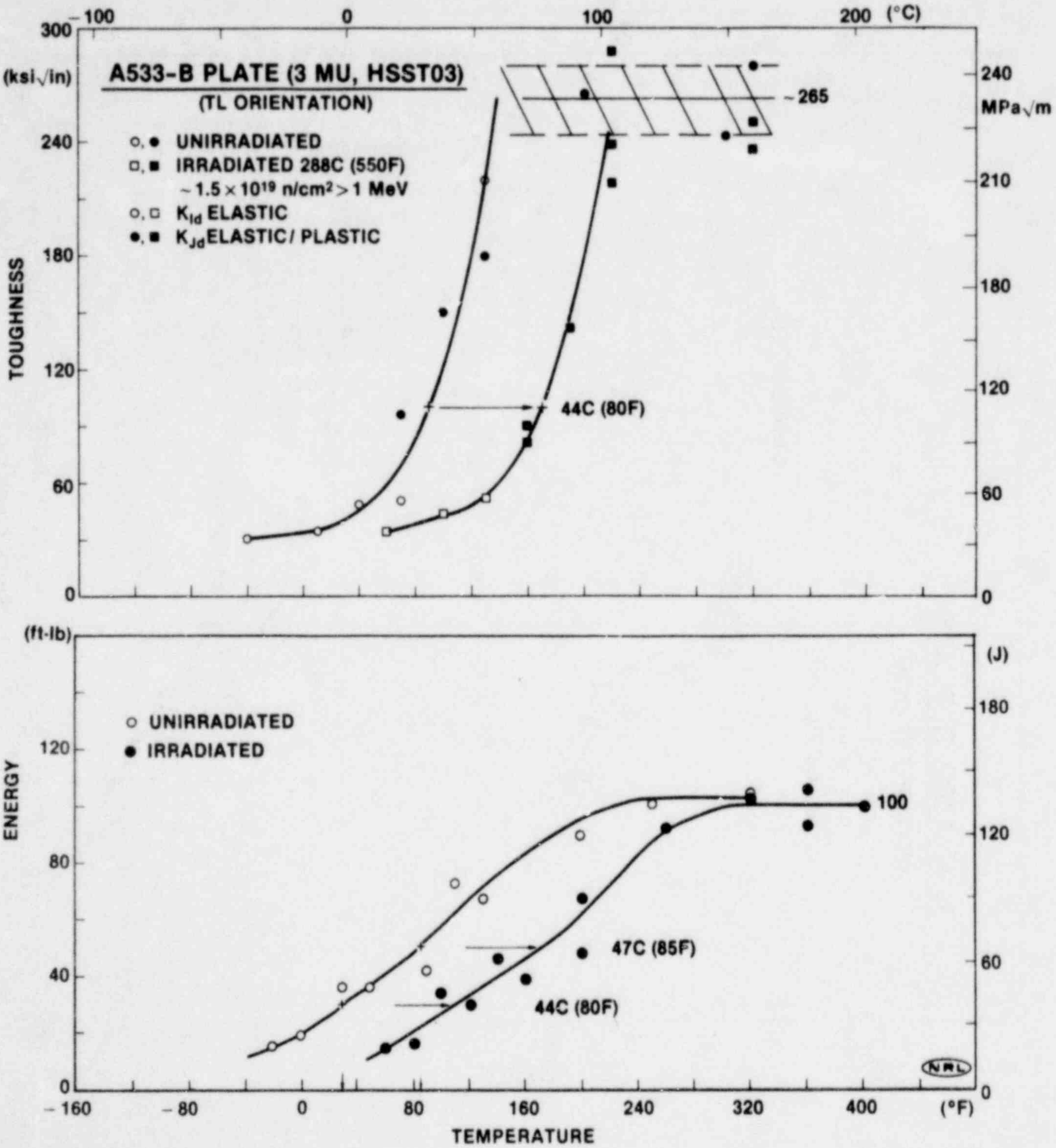


FIGURE 3

1603 049



A533-B PLATE (3 MU, HSST03)
(TL ORIENTATION)
○, ● UNIRRADIATED
□, ■ IRRADIATED 288C (550F)
~1.5 × 10¹⁹ n/cm² > 1 MeV
○, □ K_Id ELASTIC
●, ■ K_Jd ELASTIC/PLASTIC
TOUGHNESS
(ksi√in)
300
240
180
120
60
0
−100
0
100
200
(°C)
MPa√m
240
210
180
120
60
0
~265
44C (80F)
ENERGY
(ft-lb)
120
80
40
0
(J)
180
120
60
0
○ UNIRRADIATED
● IRRADIATED
100
47C (85F)
44C (80F)
NRL
−160
−80
0
80
160
240
320
400
(°F)
TEMPERATURE

FIGURE 4

PREDICTED DECREASE IN UPPER SHELF ENERGY

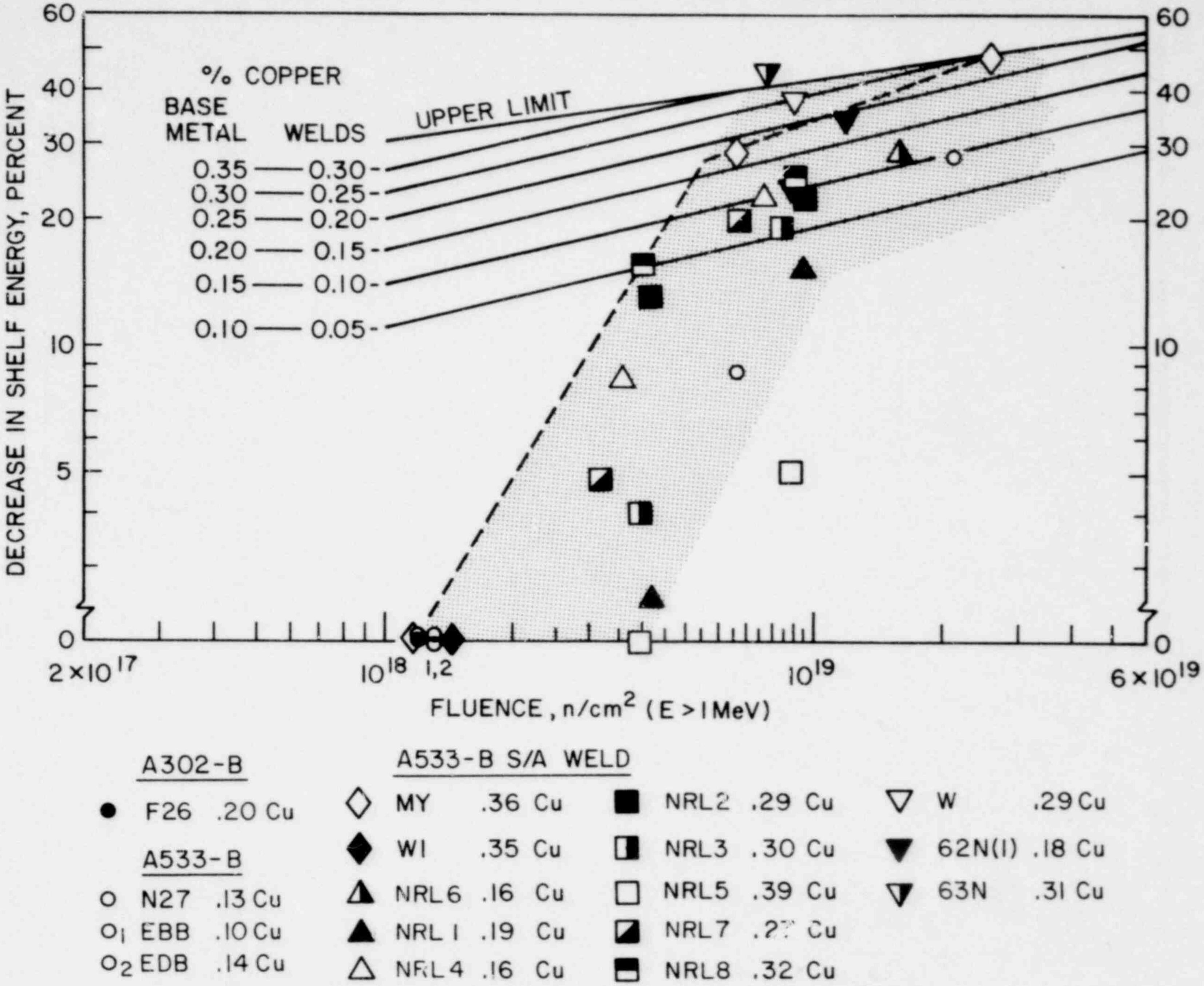
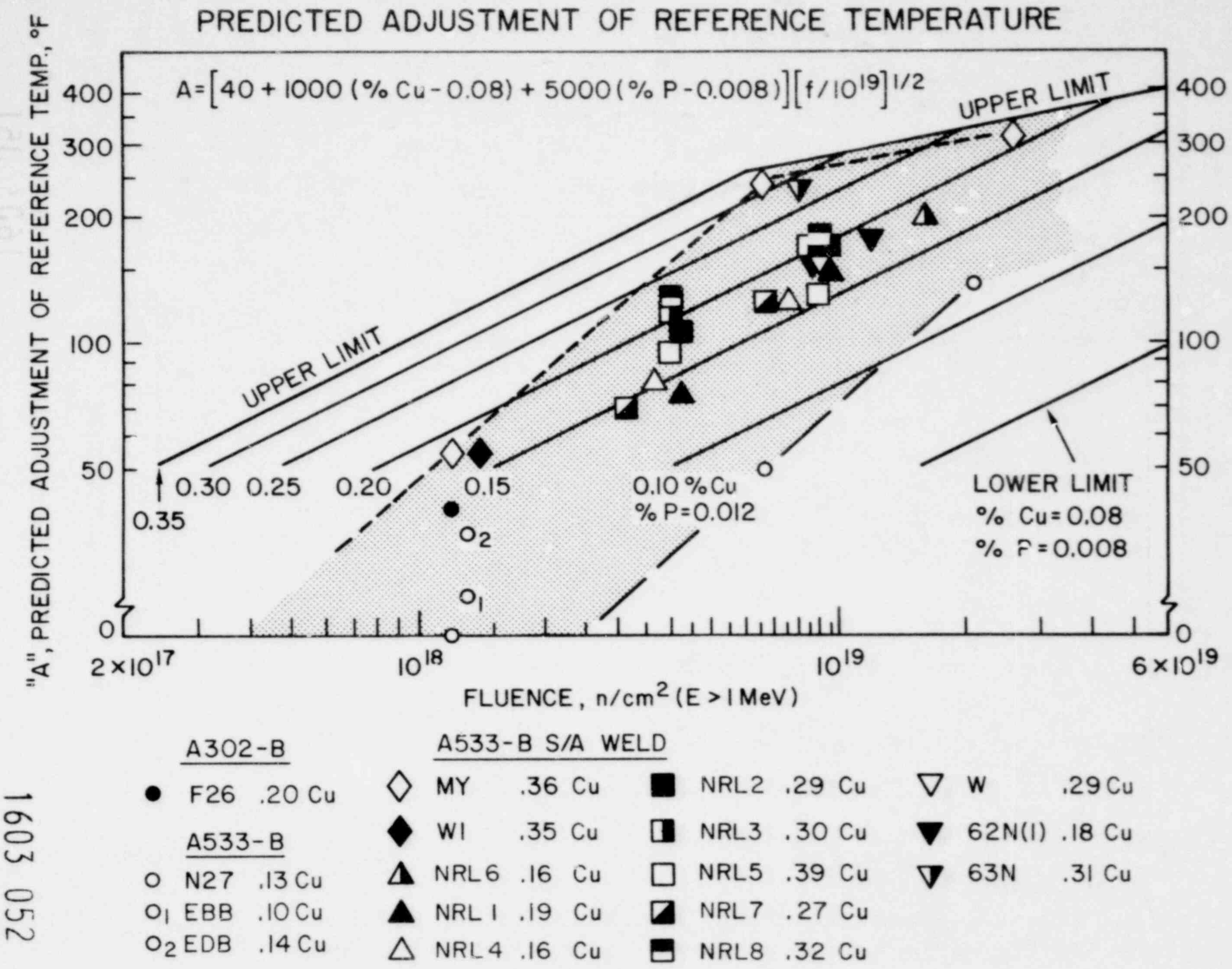
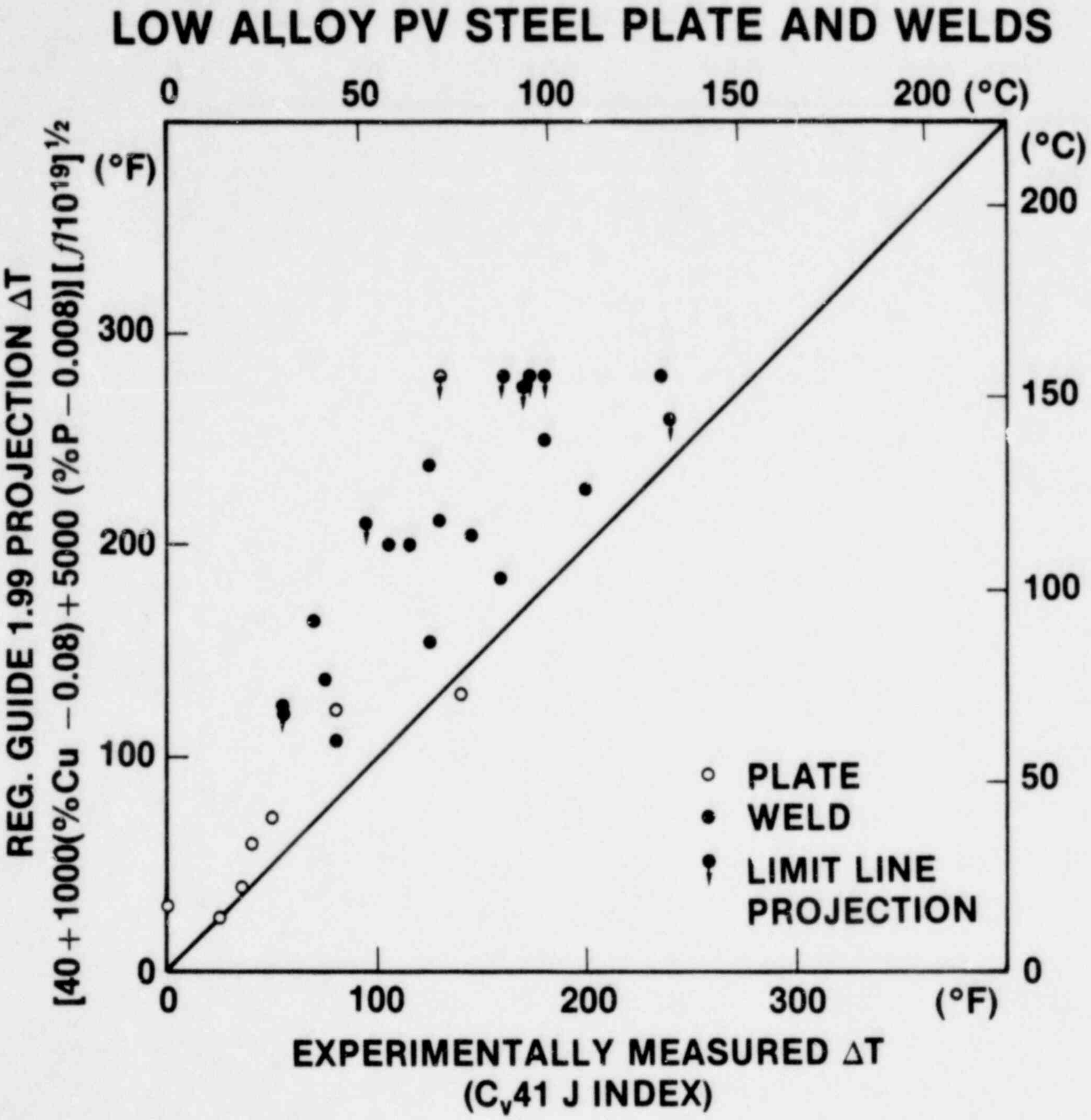


FIGURE 5



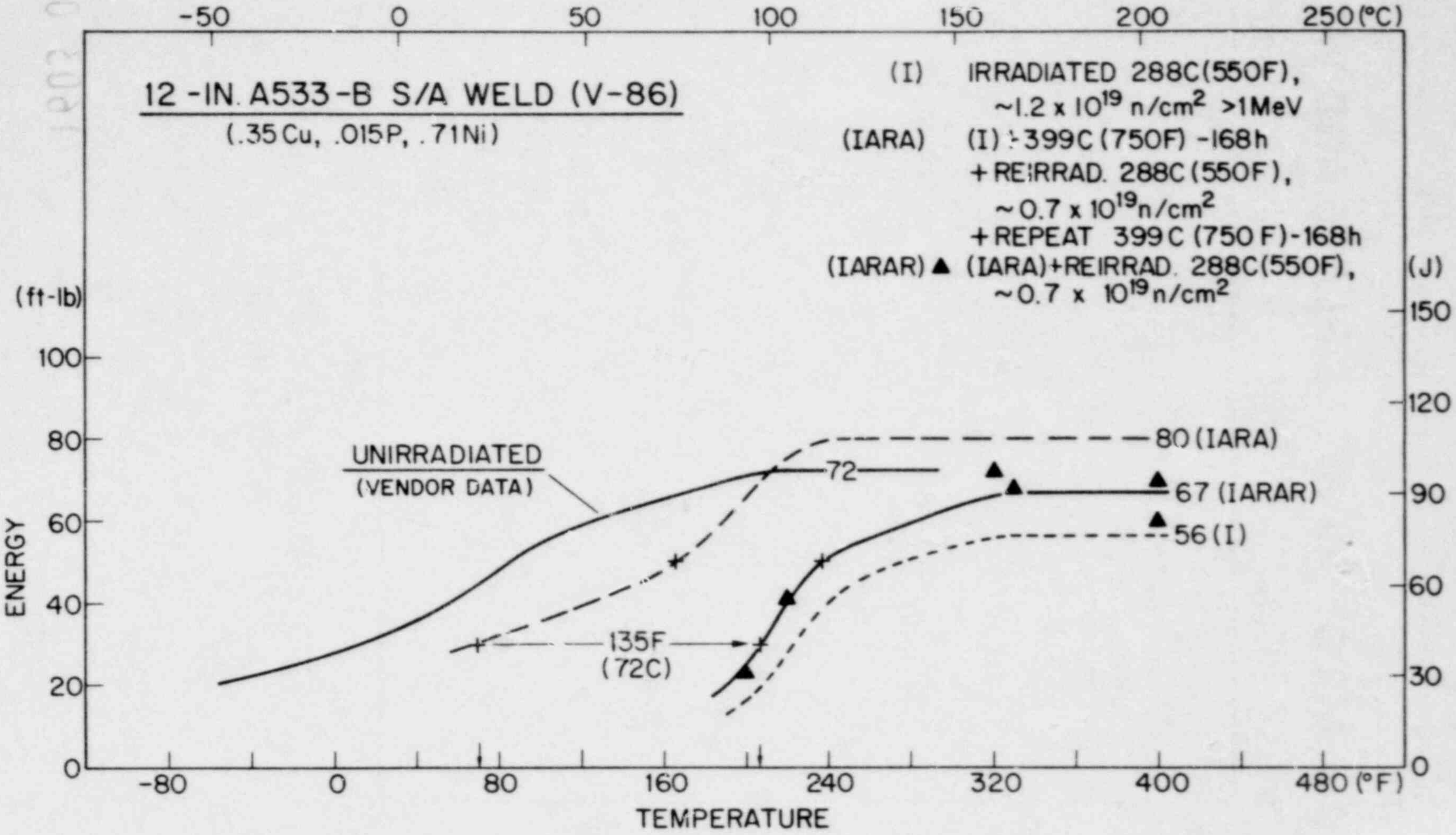
1603 052

FIGURE 6

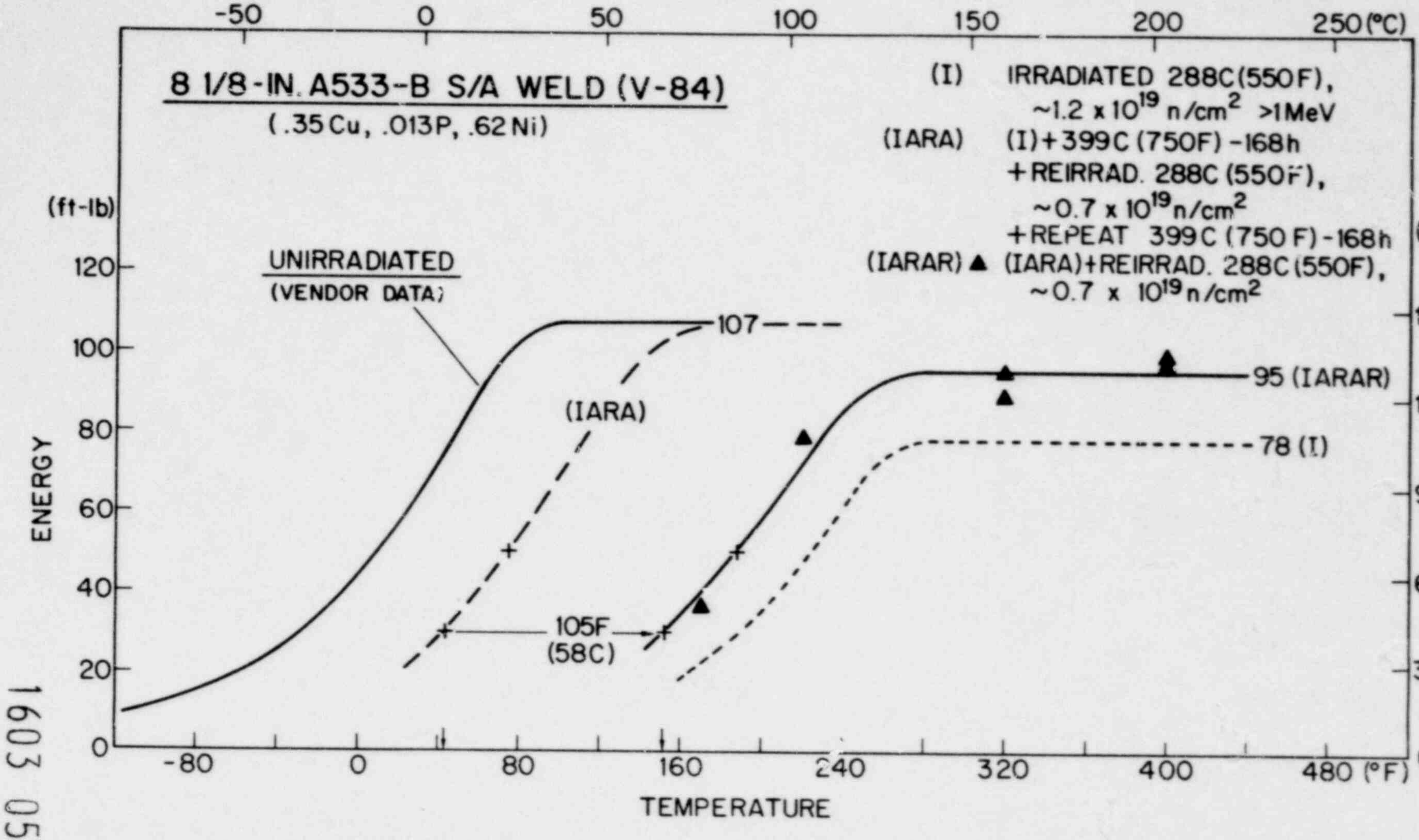


LOW ALLOY PV STEEL PLATE AND WELDS

FIGURE 7


12 -IN. A533-B S/A WELD (V-86)
(.35 Cu, .015P, .71Ni)
(I) IRRADIATED 288C(550F), ~1.2 x 10^{19} n/cm^2 >1MeV
(IARA) (I) + 399C (750F) -168h + REIRRAD. 288C (550F), ~0.7 x 10^{19} n/cm^2 + REPEAT 399C (750 F)-168h
(IARAR) ▲ (IARA)+REIRRAD. 288C(550F), ~0.7 x 10^{19} n/cm^2
UNIRRADIATED
(VENDOR DATA)
72
80 (IARA)
67 (IARAR)
56 (I)
135F
(72C)
ENERGY
(ft-lb)
0
20
40
60
80
100
(J)
0
30
60
90
120
150
TEMPERATURE
-80
0
80
160
240
320
400
480 (°F)
-50
0
50
100
150
200
250 (°C)

1603 054


8 1/8-IN. A533-B S/A WELD (V-84)
(.35 Cu, .013P, .62 Ni)
(I) IRRADIATED 288C(550F), ~1.2 x 10^{19} n/cm^2 >1MeV
(IARA) (I)+399C (750F) -168h +REIRRAD. 288C (550F), ~0.7 x 10^{19} n/cm^2 +REPEAT 399C (750 F)-168h
(IARAR) ▲ (IARA)+REIRRAD. 288C(550F), ~0.7 x 10^{19} n/cm^2
UNIRRADIATED
(VENDOR DATA)
(IARA)
107
95 (IARAR)
78 (I)
105F
(58C)
ENERGY
(ft-lb)
0
20
40
60
80
100
120
(J)
0
30
60
90
120
150
TEMPERATURE
-80
0
80
160
240
320
400
480 (°F)
-50
0
50
100
150
200
250 (°C)