- NRC-JNES bilateral meeting on Material Related Research -
May 20-21, 2009

Summary of the PRE-project of JNES

Prediction of Radiation Embrittlment
for highly irradiated Reactor vessel steels

Hiroshi Matsuzawa

Japan Nuclear Enargy Safty Organization (JNES)
3-17-1, Toranomon, Minato-ku, Tokyo, Japan
E-mail;matsuzawa-hiroshi@jnes.go.jp

Abstract

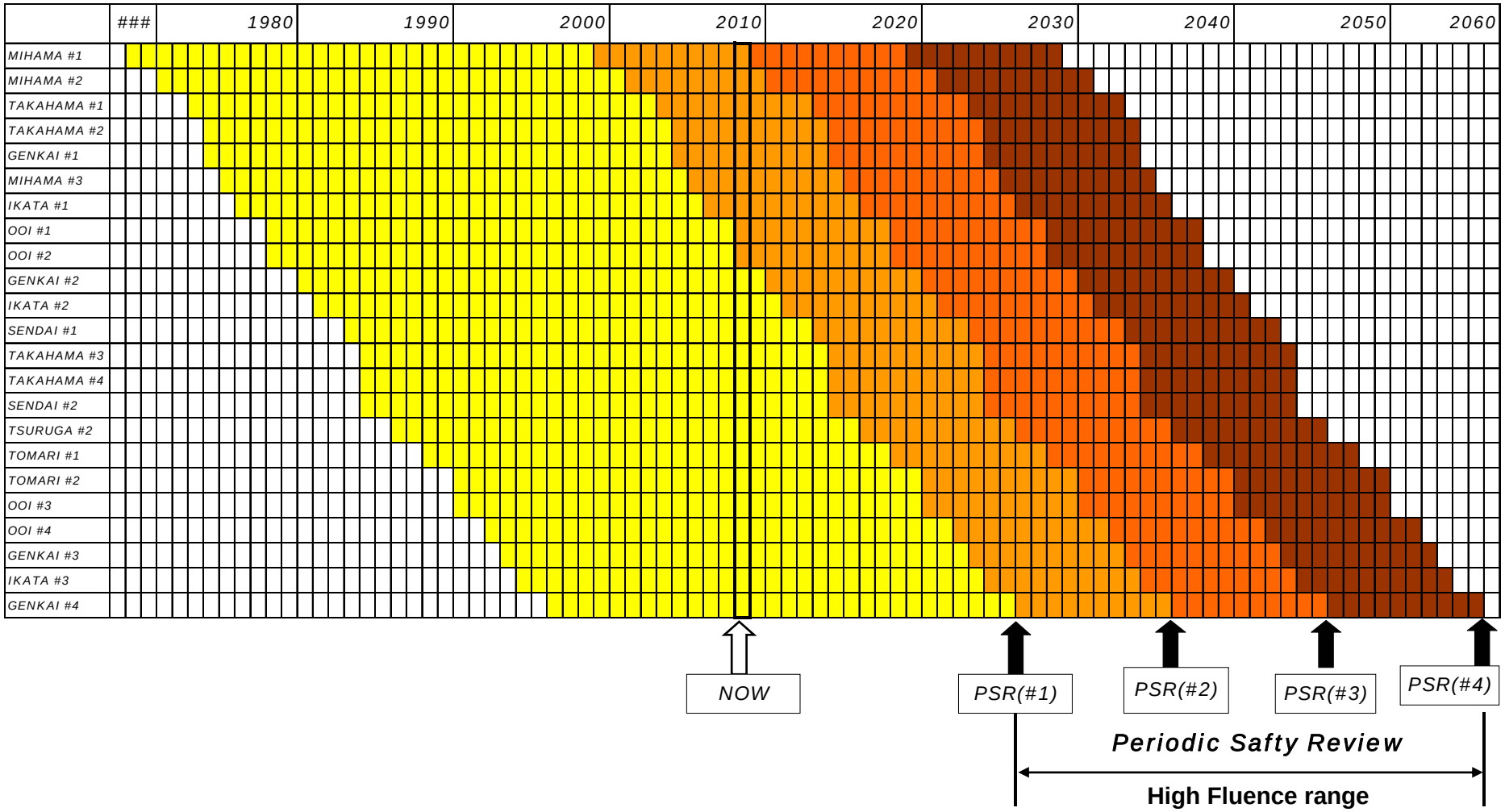
-There are 55 nuclear power plants (23 PWR and 32 BWR type plants) are now under operating in Japan.

-In 2010, 20 NPPs will have been operating for 30 years and 2 NPPs for 40 years since starting the commercial operation.

-Some of them will be operated until 60 years for operation periods, and will be verified the integrity for assessment of NPPs for every ten years in Japan.

PWR Reactor Pressure Vessel

Estimated Operartion Periods for Japanese PWR Plants



There are 23 PWR plants existing in Japan, and the RPVs of these plants will be operated over 30 years. In Japan, the Periodic Safety Review is required every 10 years in the case of over 30 years operation.

Abstract

-Reactor Pressure Vessels (RPVs) are required to be evaluated the reduction of Fracture Toughness and the Reference Temperature in the transition region.

-As the operating period will be longer, the prediction for these parameters will be more important.

-Recently the domestic prediction formula of embrittlement is revised based on the database of domestic NPP surveillance test results for about 30 year olds (JEAC4201).

-The adequacy for this prediction formula, whether is it valid for 60 year periods or not, should be verified by use of the accelerated irradiation test data.

PWR Reactor Pressure Vessel

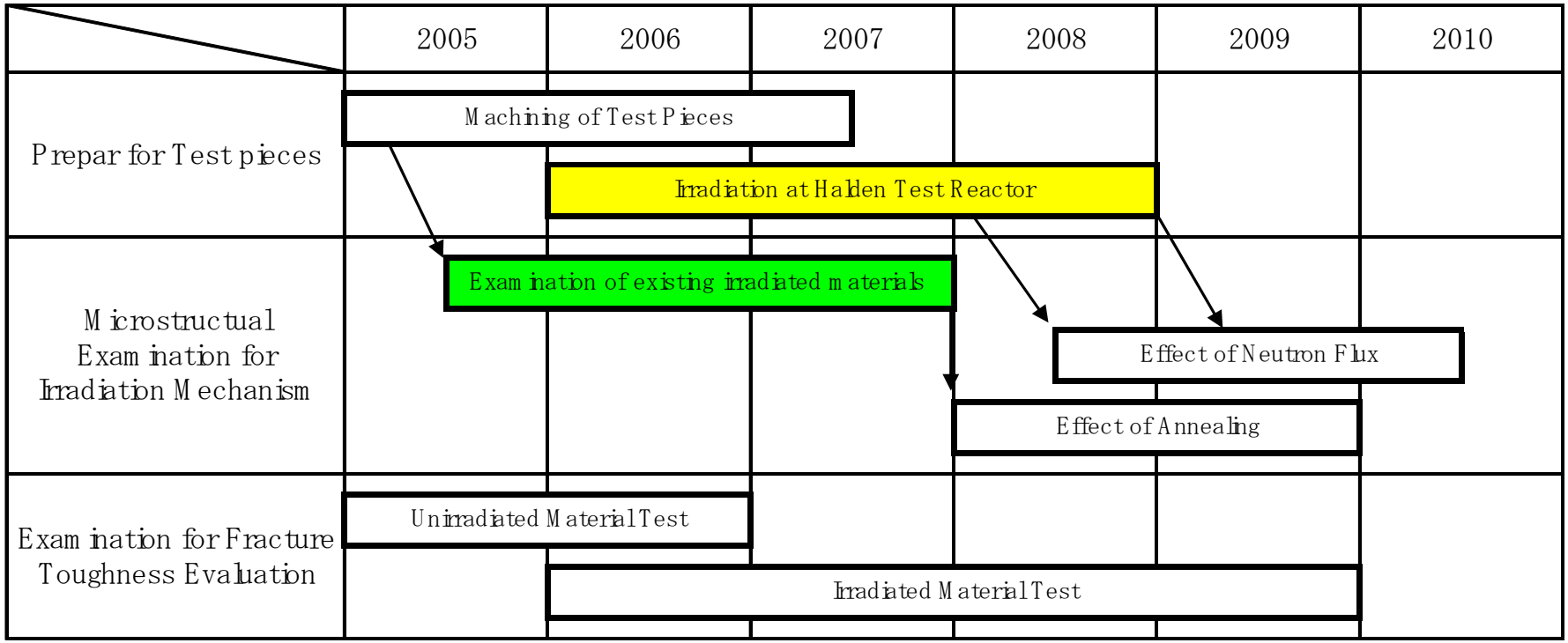
Object

- The target of the “PRE” project is to make clear the mechanism of the radiation embrittlement, especially the effects of the neutron flux, for the RPVs and to verify the prediction formula of the radiation embrittlement for highly irradiated RPV steels in order to apply the evaluation for the Periodic Safety Review in Japan.

- To achieve the above targets, microstructural examination for the several irradiated test materials modified for the RPV steels has been performed.

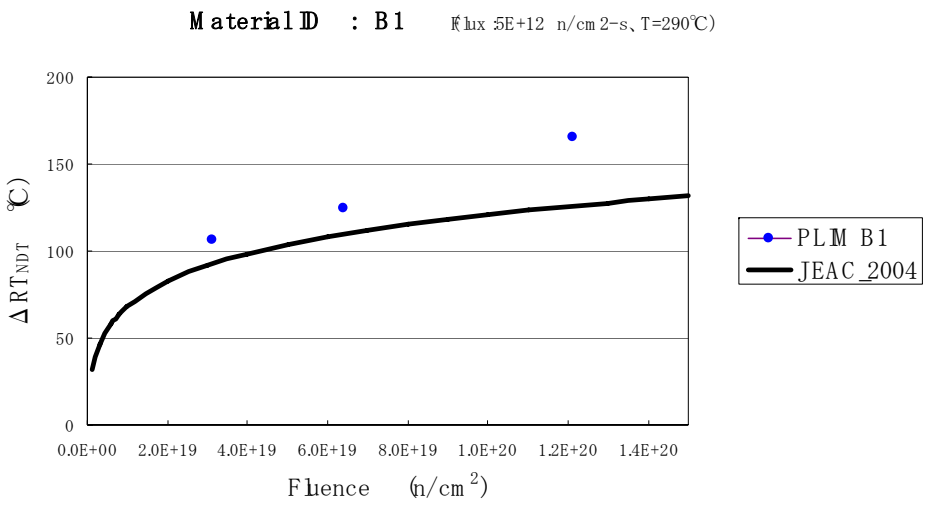
- Furthermore, PWR standard servarance test materials that had been irradiated in the actual plant in Japan have been reirradiated at the OECD/Halden reactor from 2006 to 2008 in order to investigate the effect of the neutron Flux.

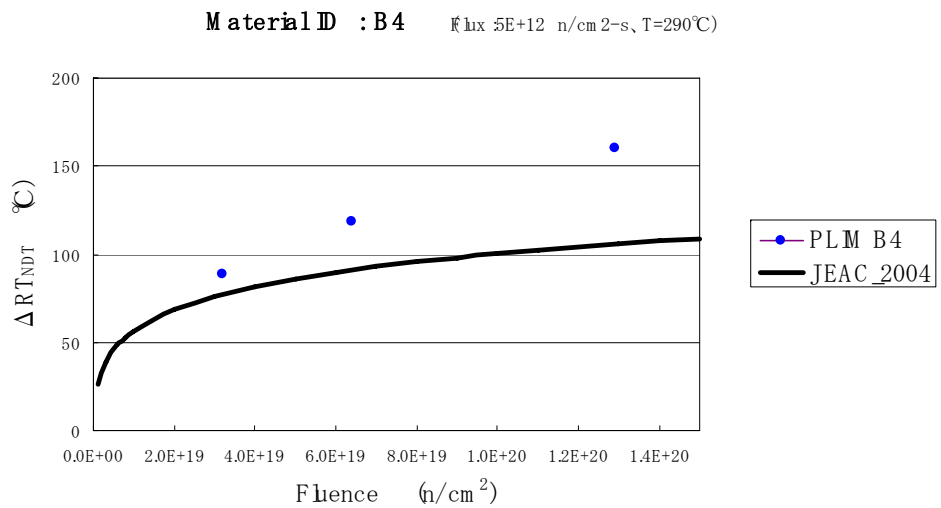
Schedule of "PRE" project

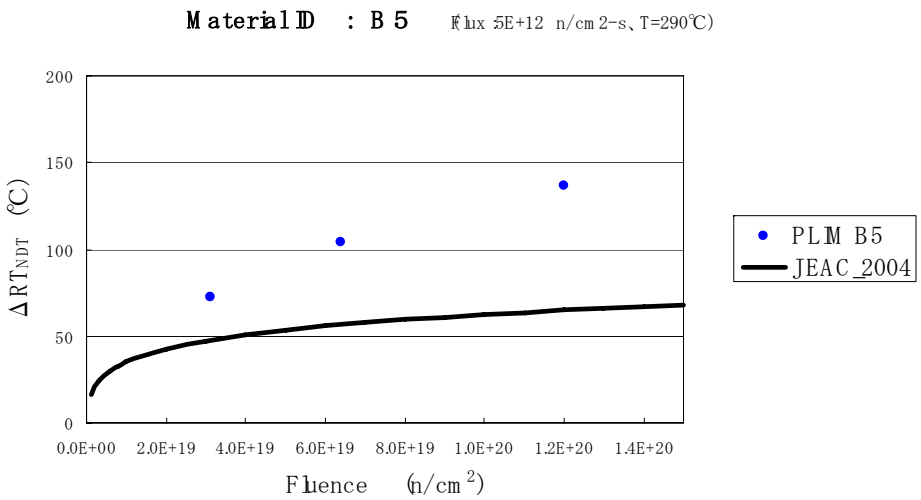


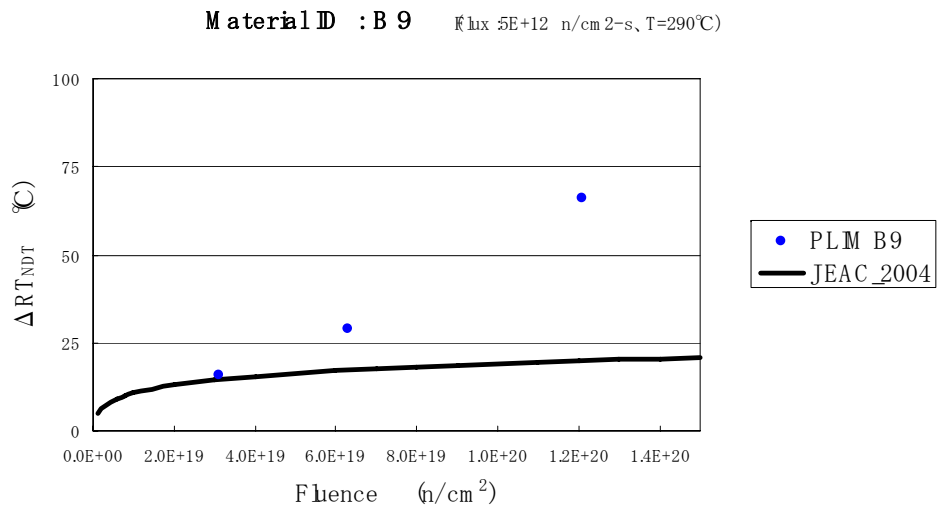
Material List for observation

Test materials		Chemical Composition (w.t.%)				Characteristic	Material I.D.	Fluence ($\times 10^{19}$n/cm^2)	Microstructual Obsevation	
		Cu	Ni	P	Si				3DAP(LEAP)	TEM
Base Metal	B1	0.21	0.63	0.009	0.17	High Cu material	B1-1	3.1	○	○
							B1-2	6.4	○	○
							B1-3	12.1	○	○
	B4	0.17	0.62	0.009	0.15	Medium Cu material	B4-1	3.2	○	
							B4-2	6.4	○	
							B4-3	12.9	○	
	B5	0.10	0.59	0.009	0.14	Low Cu material	B5-1	3.1	○	
							B5-2	6.4	○	
							B5-3	12.0	○	
	B6	0.10	0.92	0.008	0.13	High Ni material	B6-1	8.6	○	
	B7	0.10	0.63	0.017	0.14	High P material	B7-1	8.6	○	
	B8	0.10	0.63	0.009	0.32	High Si material	B8-1	8.7	○	
	B9	0.04	0.62	<0.005	0.22	Extra low Cu material	B9-1	3.1	○	○
							B9-2	6.3	○	○
							B9-3	12.1	○	○
	S1	0.09	0.62	0.007	0.26	PWR Standard surverance test material	S1-1	3.2	○	○
							S1-2	5.9	○	○
							S1-3	11.7	○	○
Weld Metal	W1	0.20	0.88	0.016	0.30	High Cu material	W1-1	3.3	○	
							W1-2	8.6	○	
							W1-3	11.8	○	
	W2	0.13	0.86	0.016	0.30	Medium Cu material	W2-1	3.2	○	
							W2-2	8.9	○	
							W2-3	11.2	○	
	W3	0.10	0.88	0.016	0.27	Low Cu material	W3-1	3.1	○	
							W3-2	8.3	○	
							W3-3	11.3	○	
	W4	0.02	0.84	0.008	0.28	Extra low Cu material	W4-1	3.1	○	○
							W4-2	9.0	○	○
							W4-3	13.0	○	○

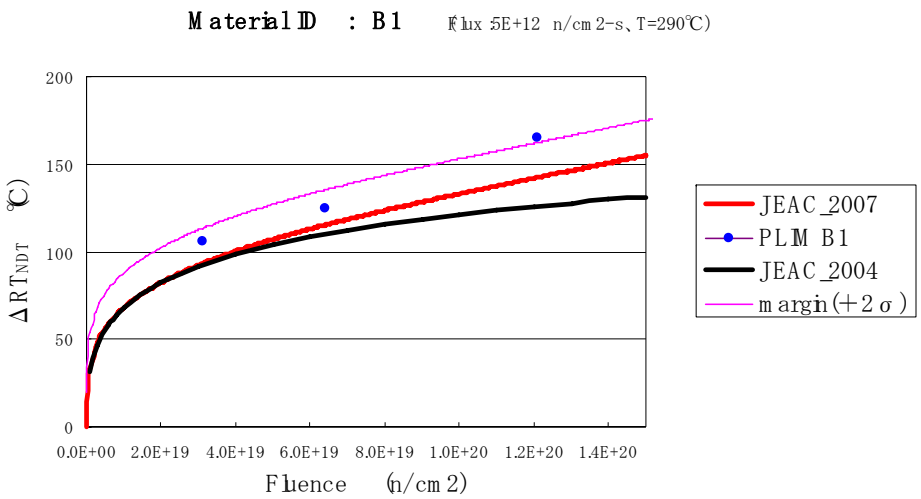


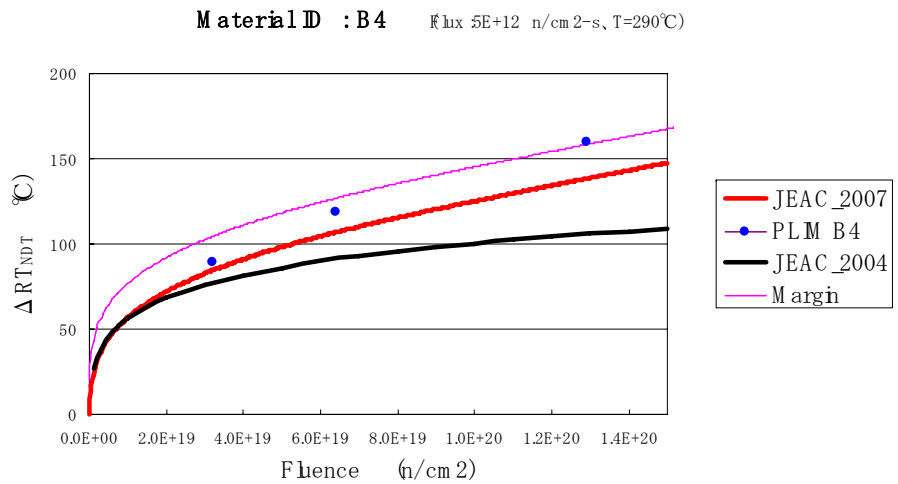


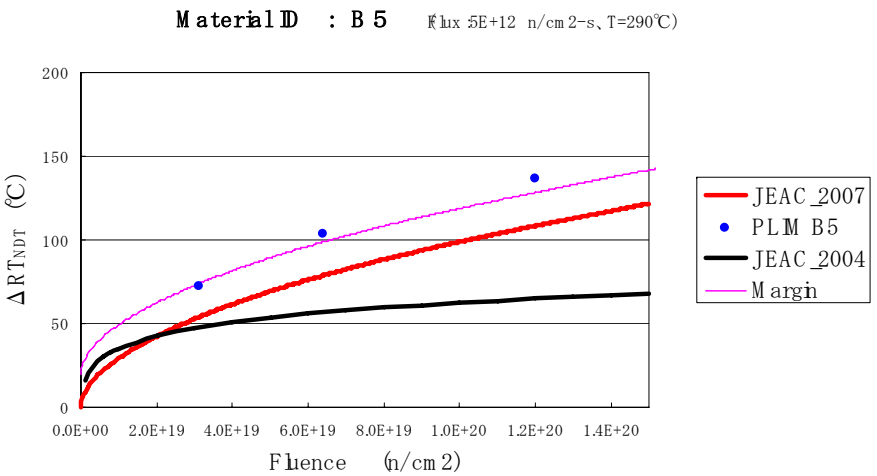


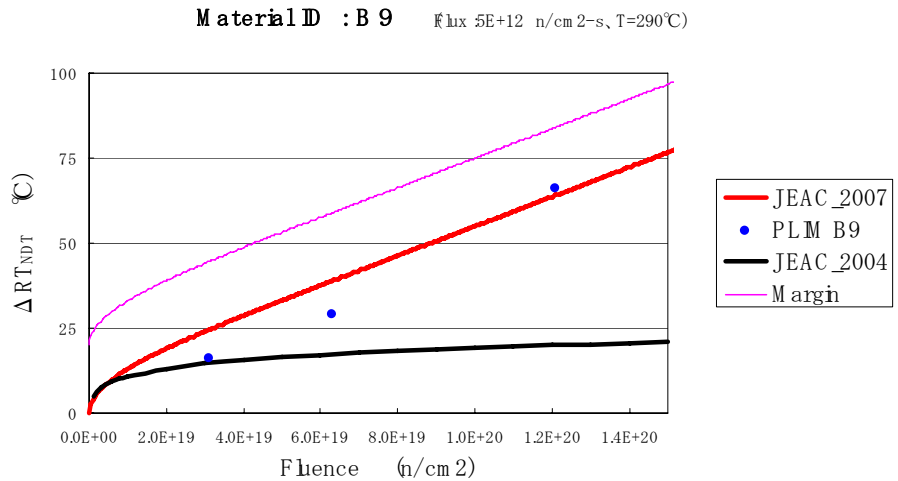


Trend Curve of JEAC4201-2004 vs. PLIM MTR data

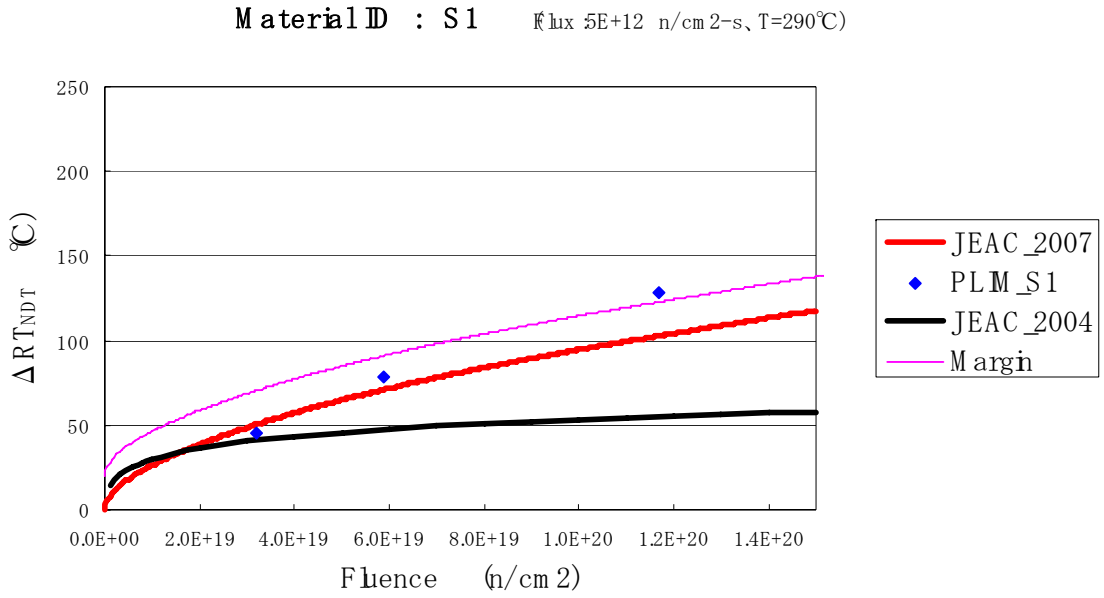


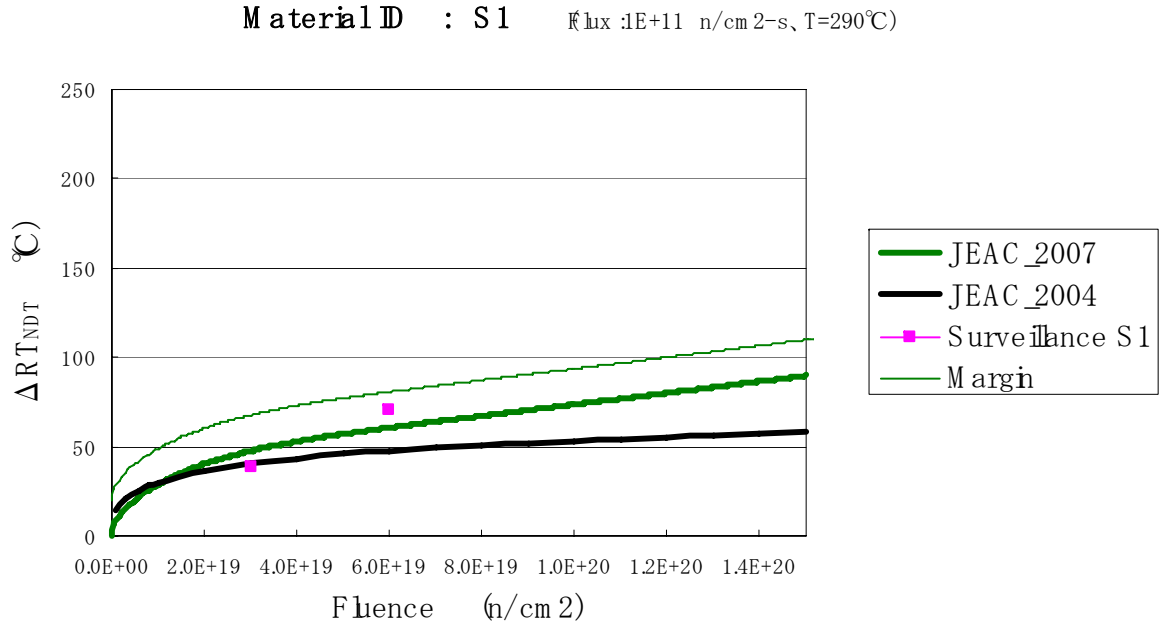




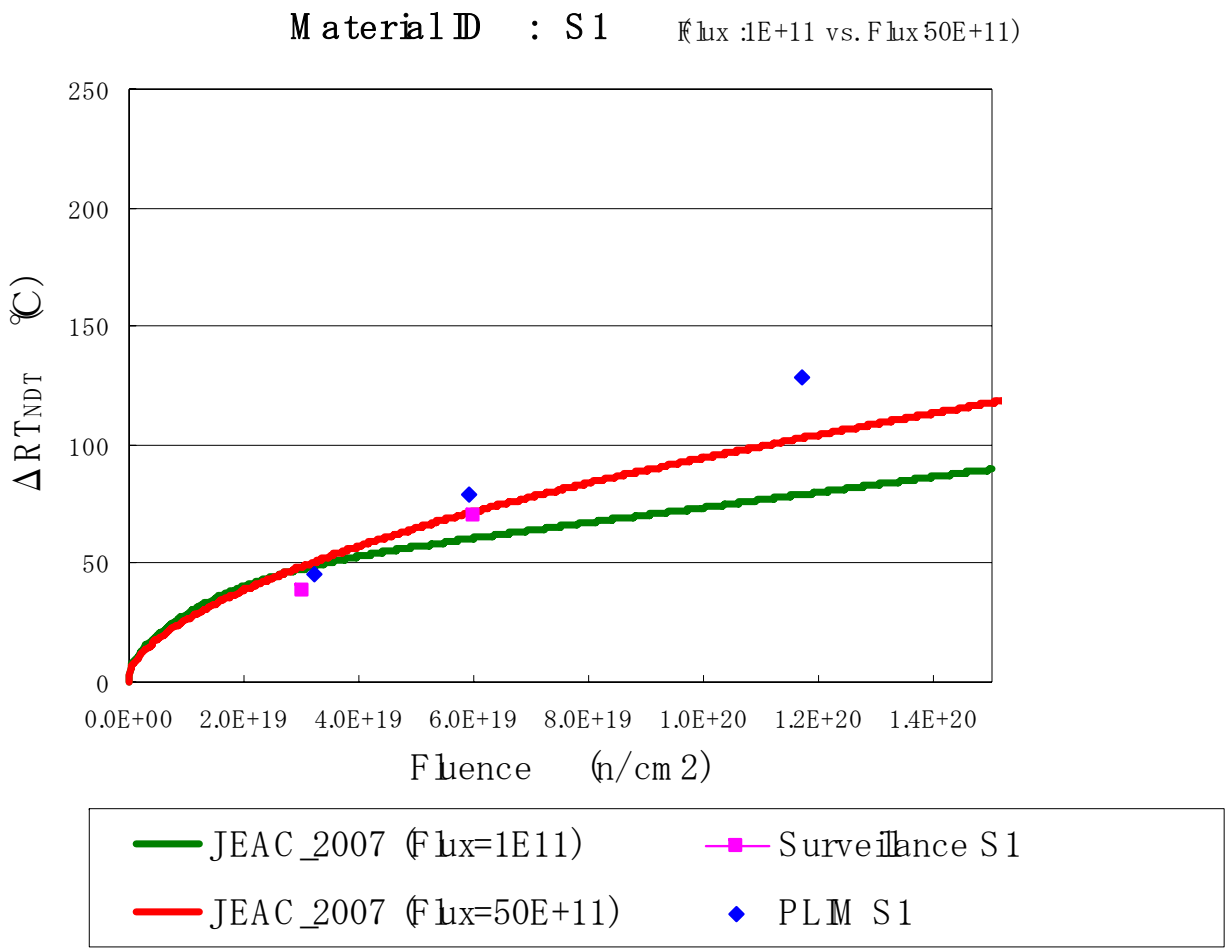


Trend Curve of JEAC4201-2007 vs. PLIM MTR data



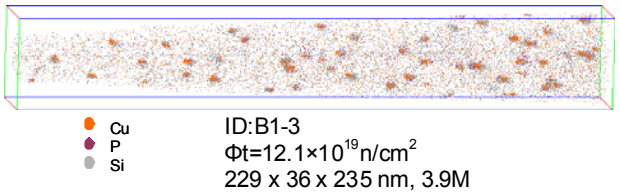


Trend Curve of JEAC4201-2007 vs. PWR Standard test data

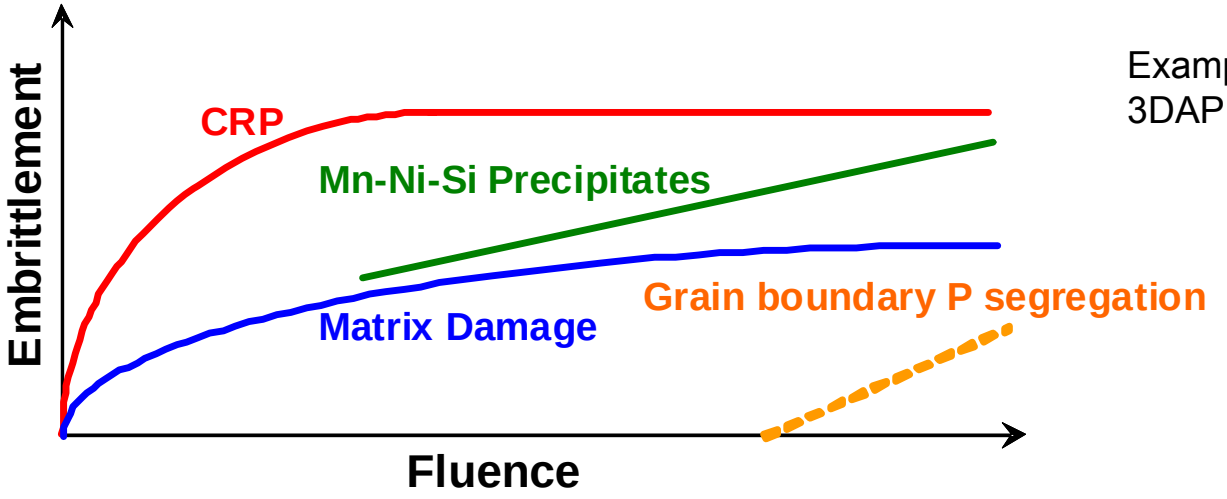


Comparison between Flux 1E+11 vs. 50E+11(n/cm²/s)

- Nutron Irradiation Embrittlement
  - Embrittlement caued by Hardening
    - Solute atom Clusters (SC)
      - Cu Rich Precipitates (CRP)
      - Mn-Ni-Si Precipitates
    - Matrix Damage
  - Embrittlement caued by Non-hardening
    - Grain boundary P segregation
    - Other segregation



Example of Atom map obtained by 3DAP



Mechanism of Embrittlement

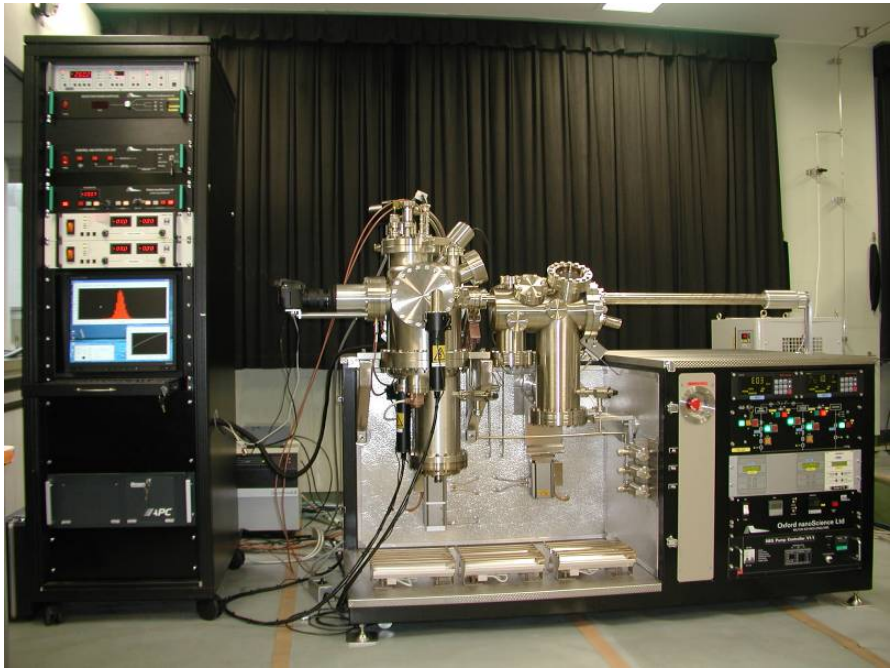
3D Atom Probe Observation

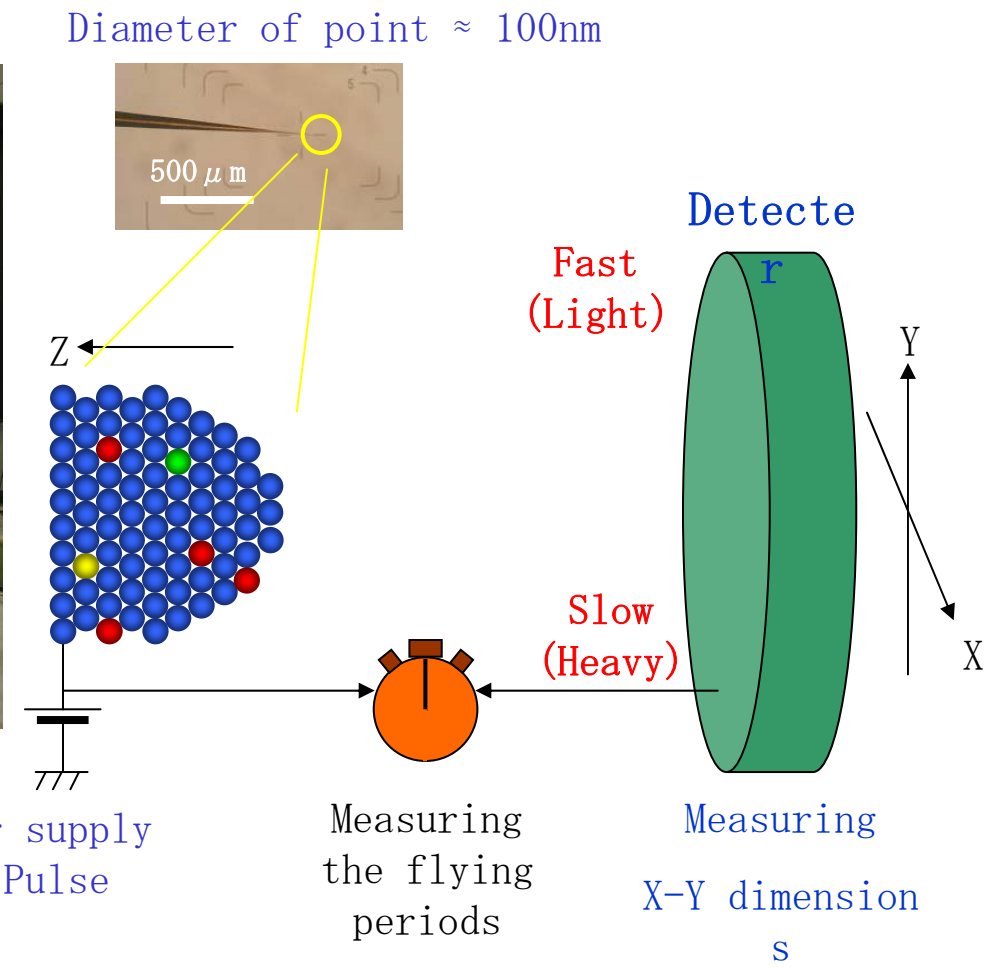
- The 3D-Atom Probe(3DAP) experiments were performed in order to investigate the microstructual change effected by irradiation.
- The typical results “B-1(high Cupper case)” and “B-9(low Cupper case)” are shown.
- Both materials could be observed clusters that are considered the main cause of the irradiation embrittlement.
- Although clusters in manganese, nickel, silicon, and phosphorus could be found for both materials, but copper clusters could not be found for material “B9”.

Microstructual examination results (sample)

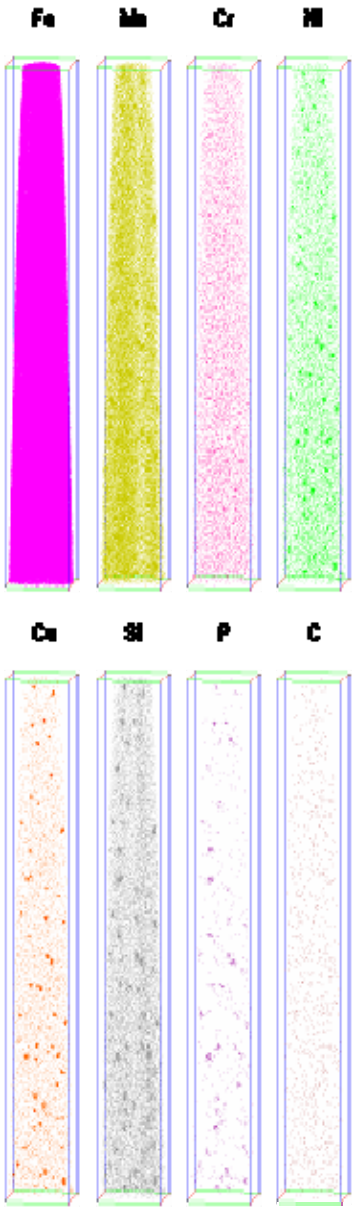
- Material type
  - Base metal (SA533B equivalent)
  - Material ID: **B1**
- Chemical composition:
  - Cu: 0.21wt.%, Ni: 0.63wt.%, P: 0.009 wt.%, Si: 0.17wt.%
- Irradiation condition:
  - Flux:
    - 5×10^{12} n/cm^2, E>1MeV
      (about 50 times higher to the actual RPV)
  - Fluence:
    - B1-1: 3.1×10^{19} n/cm^2, E>1MeV
    - B1-2: 6.4×10^{19} n/cm^2, E>1MeV
    - B1-3: 12.1×10^{19} n/cm^2, E>1MeV
  - Irradiation temperature:
    - 290 +/- 10 ^{o}C

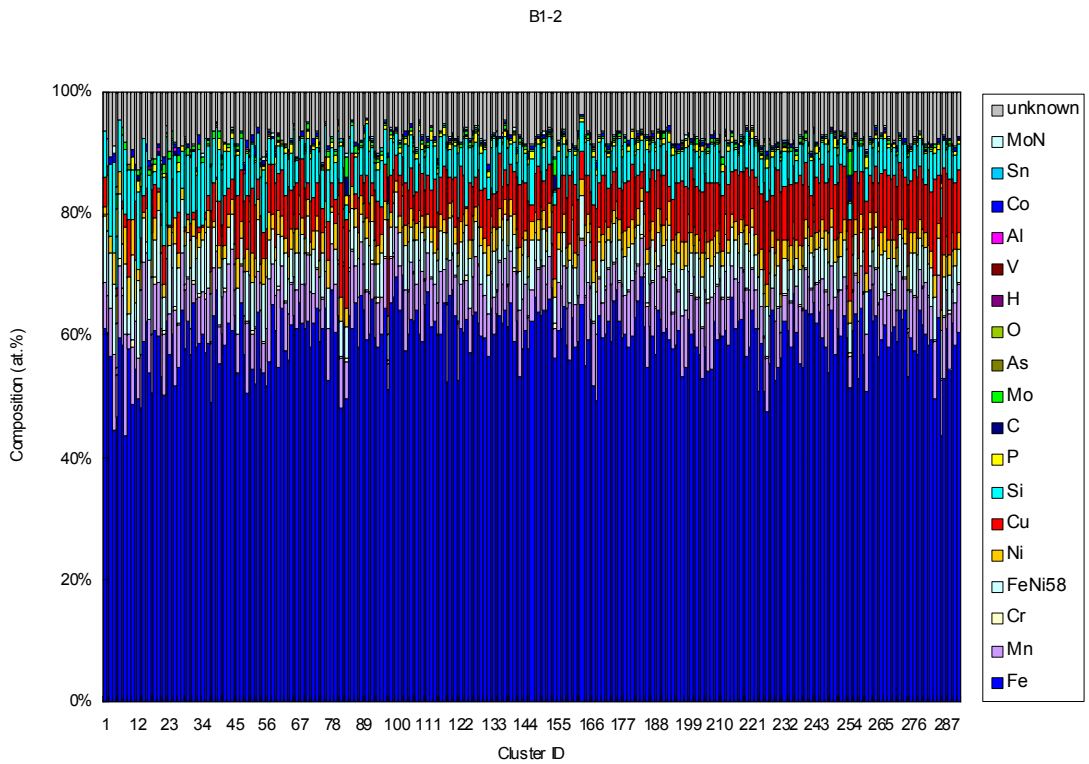
Principle of the 3D Atom Probe





Microstructual examination results

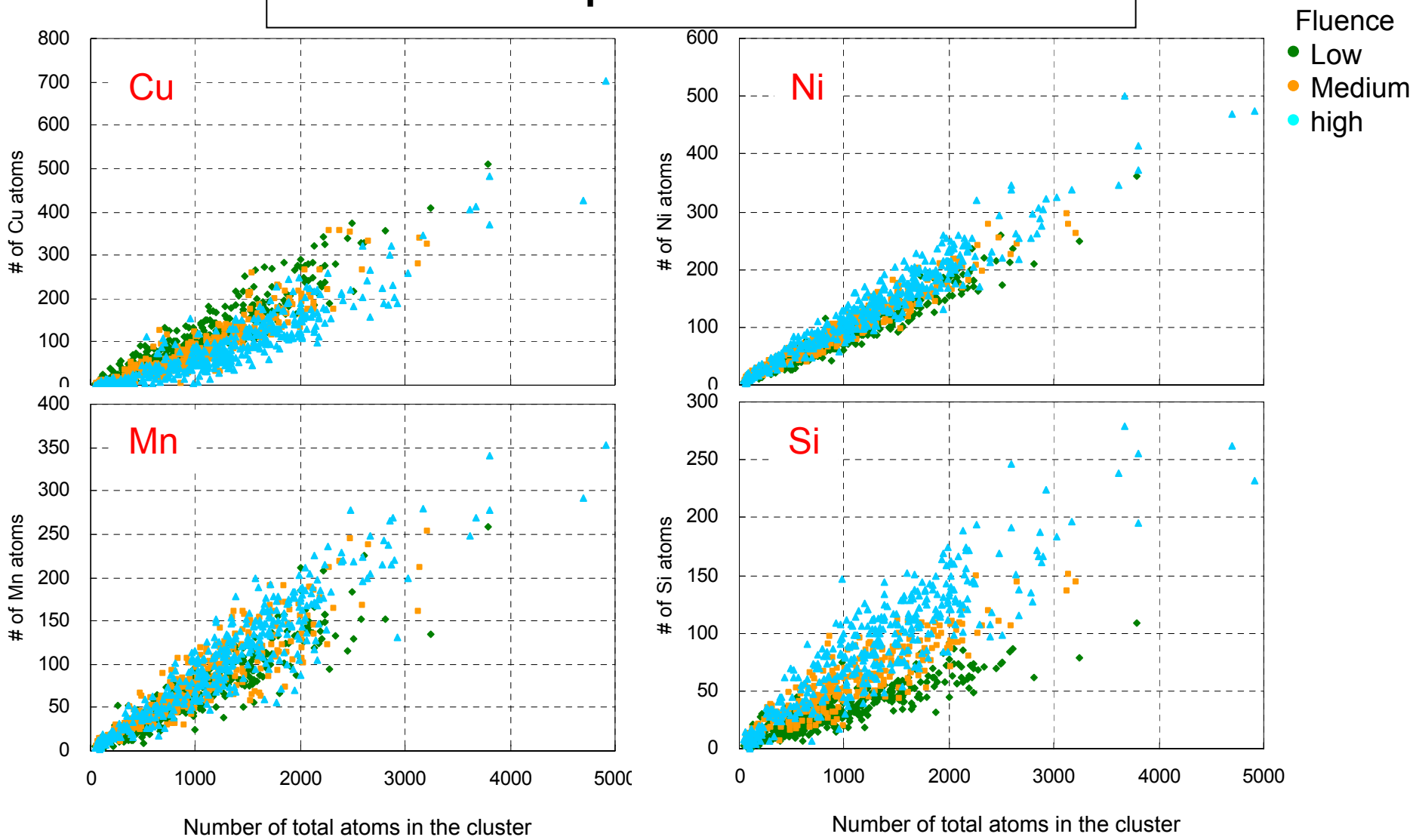




<u>Irradiated Material: B1-2</u>

(Box size= 32.8 x 40.3 x 274 nm^3)

Cluster Composition : B1 .21Cu/.63Ni

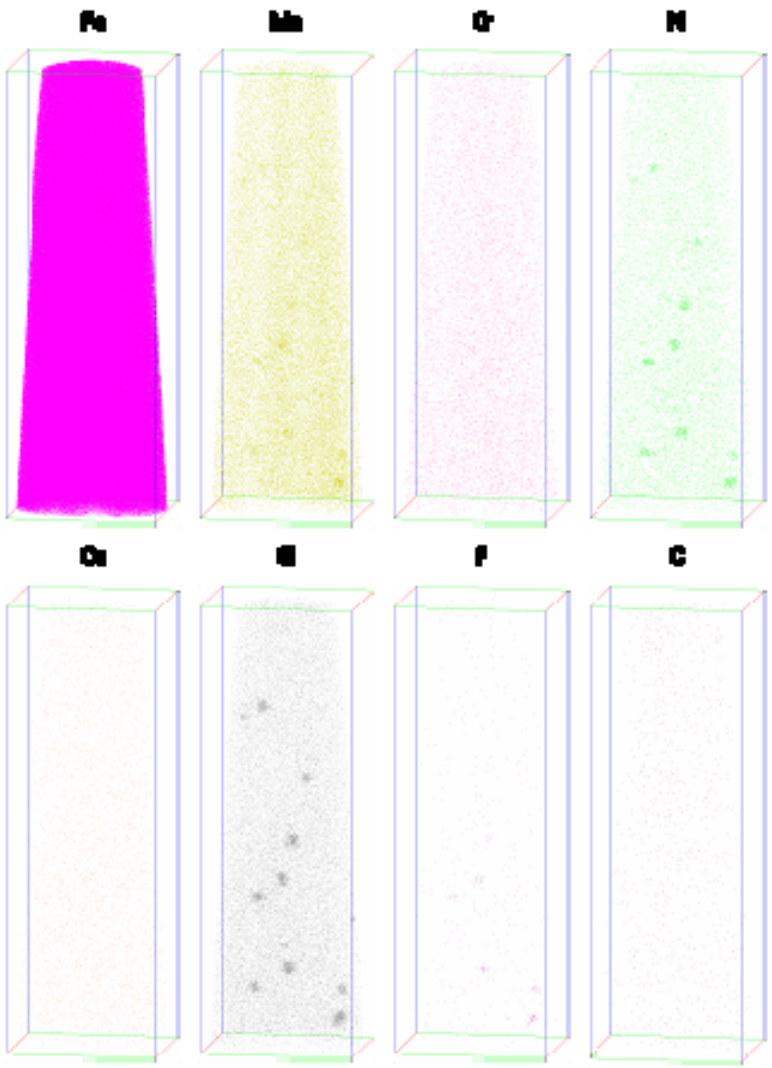


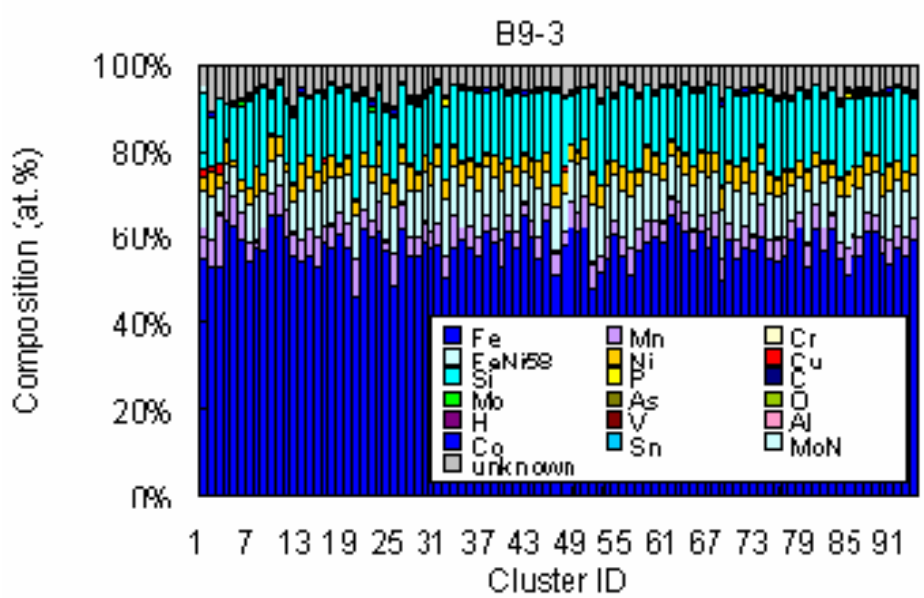
- The Ni and Si contents in clusters become larger at higher fluences.

Microstructual examination results (sample)

- Material type
  - Base metal
  - Material ID: **B9**
- Chemical composition:
  - Cu: 0.04wt.%, Ni: 0.62wt.%, P: <0.005 wt.%, Si: 0.22wt.%
- Irradiation condition:
  - Flux:
    - 5×10^{12} n/cm^2, E>1MeV
      (about 50 times higher to the actual RPV)
  - Fluence:
    - B9-1: 3.1×10^{19} n/cm^2, E>1MeV
    - B9-2: 6.3×10^{19} n/cm^2, E>1MeV
    - B9-3: 12.1×10^{19} n/cm^2, E>1MeV
  - Irradiation temperature:
    - 290 +/- 10 ^{o}C

Microstructual examination results

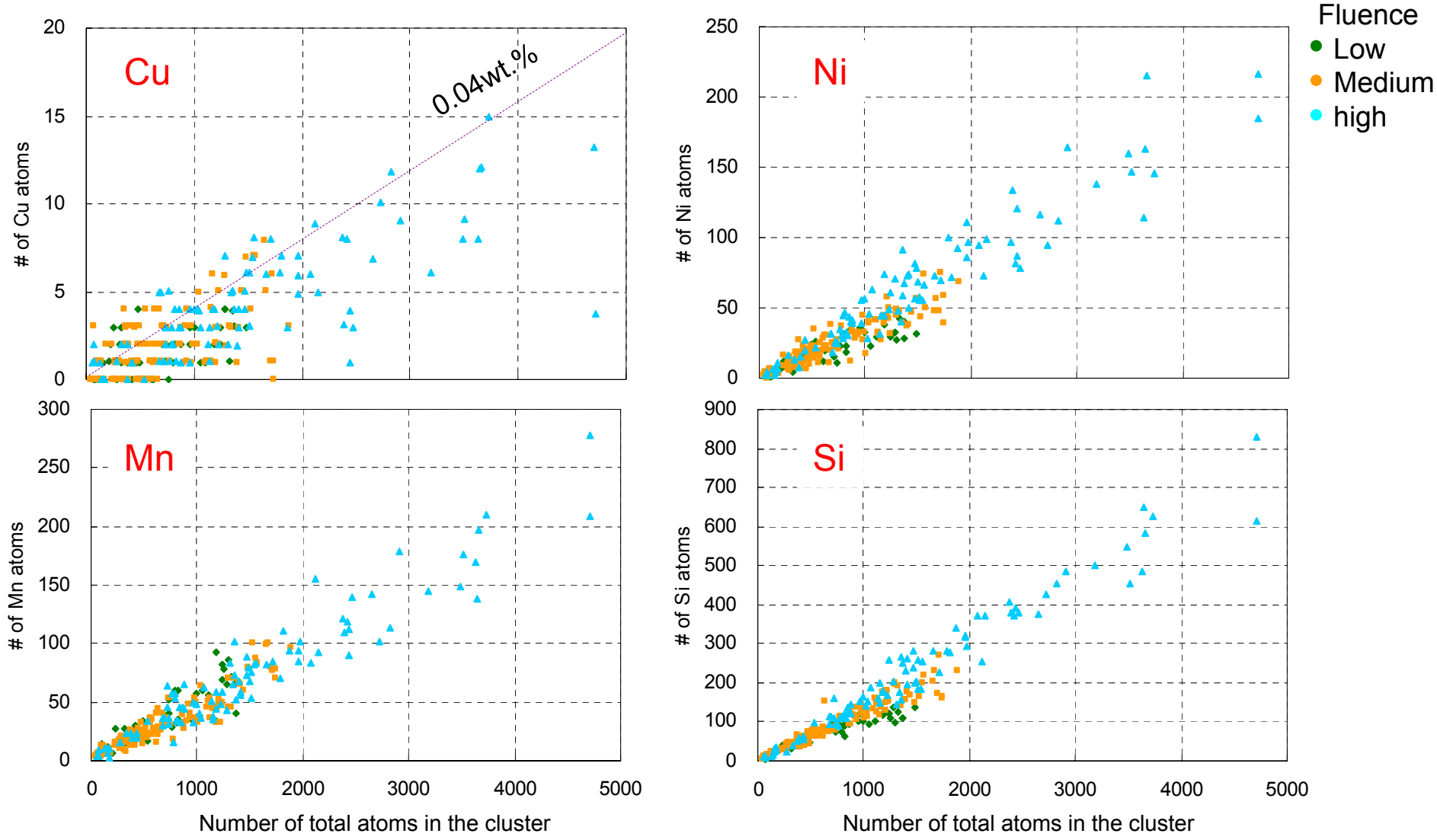




<u>Irradiated Material: B9-3</u>

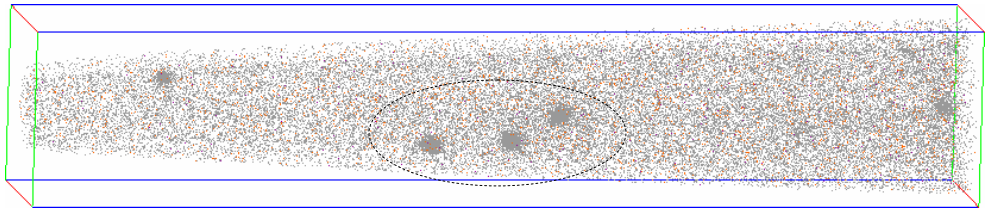
(Box size = 42.7 x 50.7 x 148.9 nm^3)

Cluster Composition : B9 .04Cu/.62Ni



- The Ni and Si contents in clusters becomes slightly larger at higher fluences.

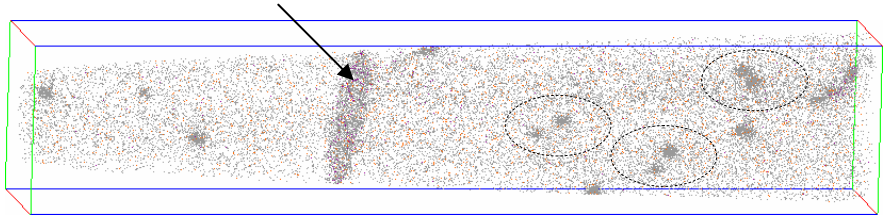
Non-uniform cluster formation in B9-3



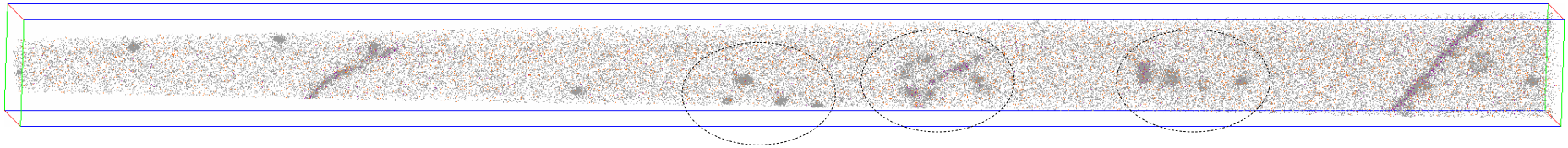
●Cu, ●Si, ●P

B9-3: R7_02605, 39.0x44.5x241nm, 8.97M

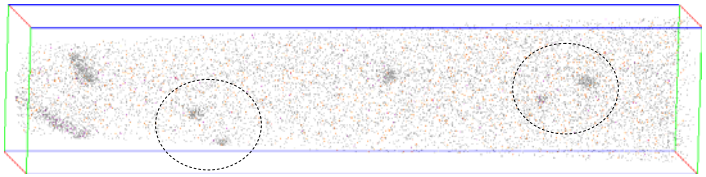
Grain boundary



B9-3: R7_02592, 32.8x37.9x191nm, 5.53M



B9-3: R7_02283, 33.5x38.0x552nm, 14.3M



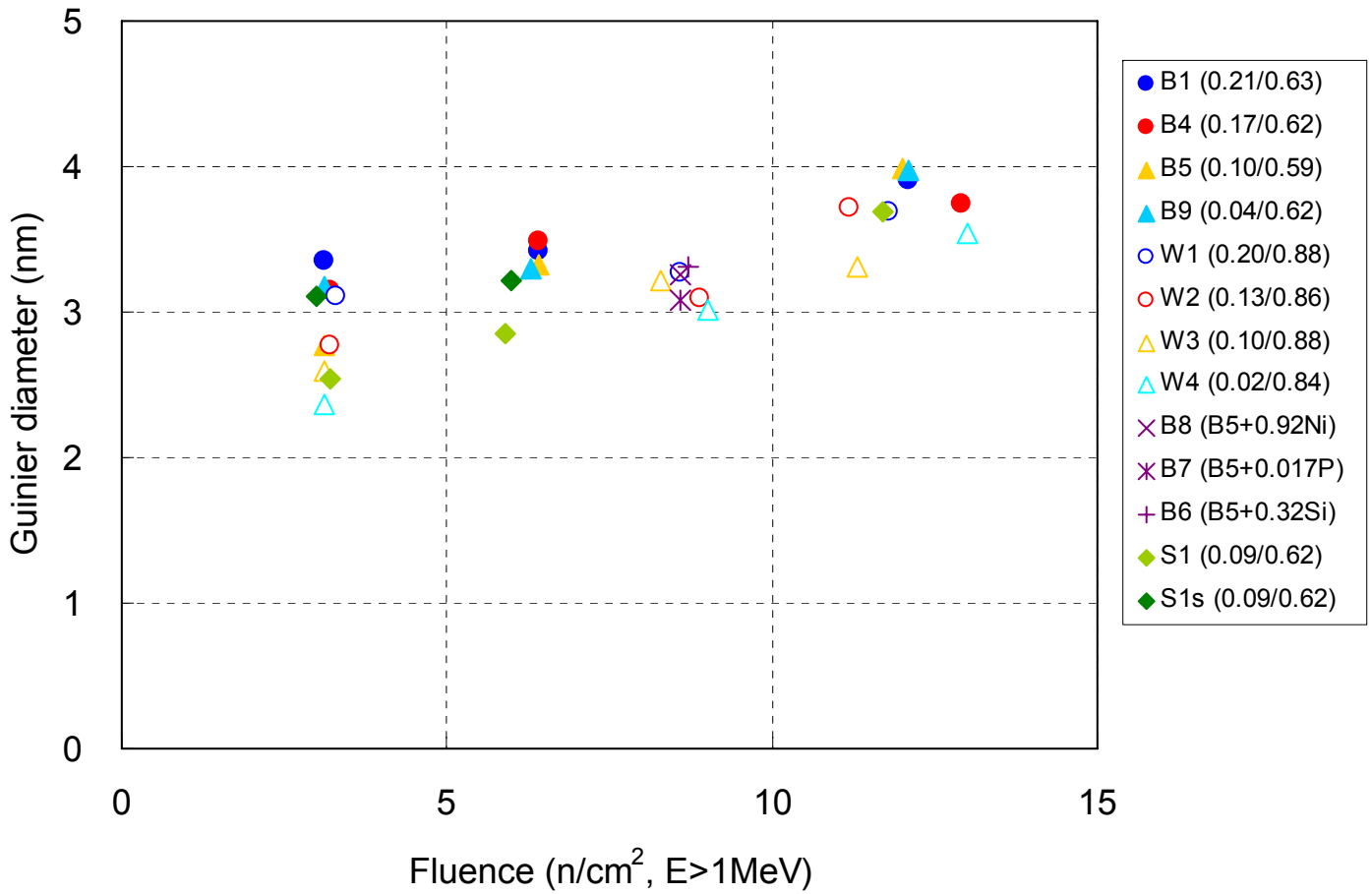
B9-3: R7_02448, 28.7x33.0x150nm, 3.30M

- Some solute atom clusters are located close to each other.

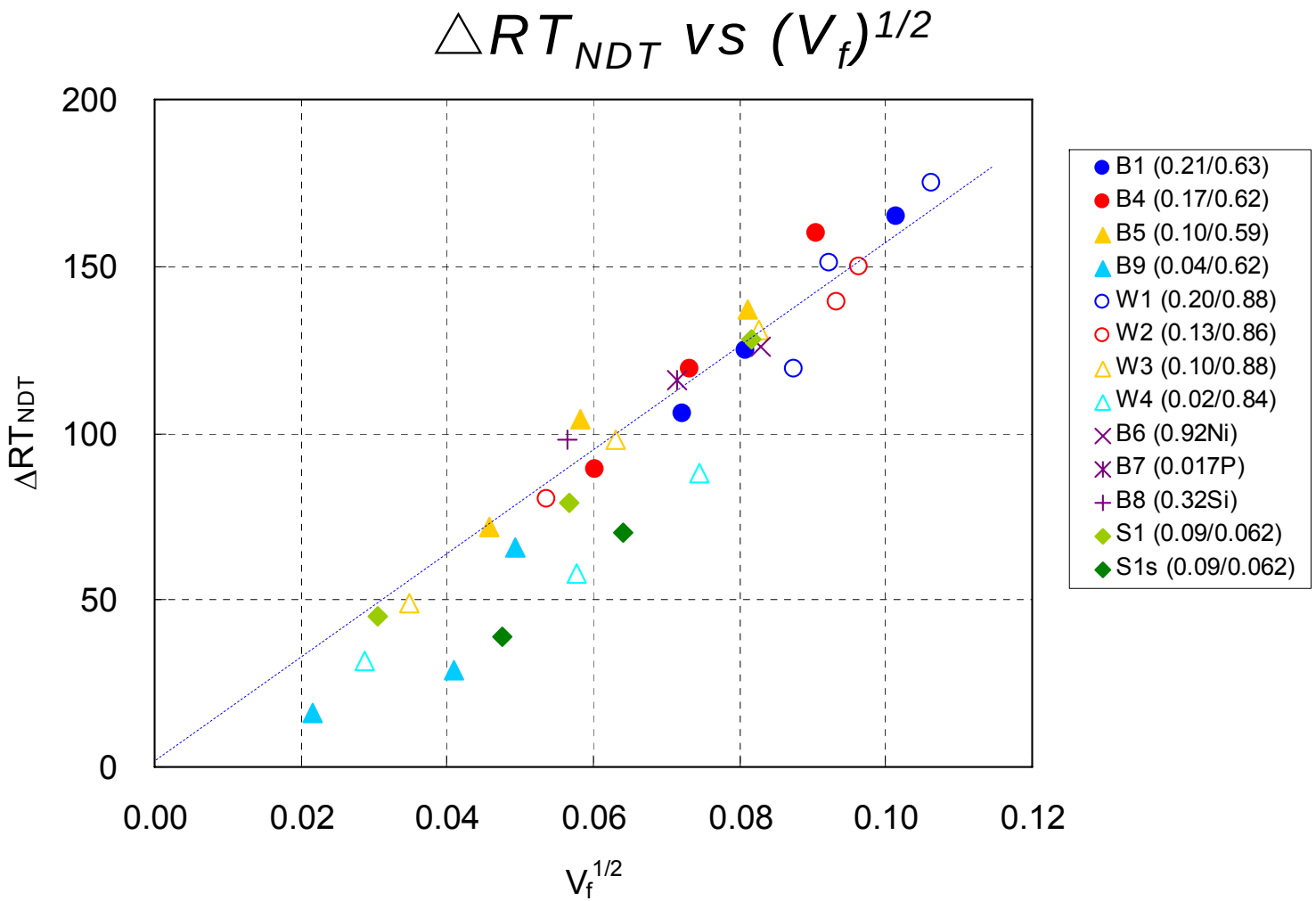
Results of the 3D Atom Probe

ID	Chemical Composition (wt.%)				Fluence	ΔRT_{NDT}	Total # of atoms	Nd	Vf	Gunier diameter	Cluster Chemical Composition (at.%)							
	Cu	Ni	P	Si	($x10^{19}$ n/cm^2)	(^oC)	($x10^7$)	($x10^{23}$ m^{-3})		(nm)	Fe	Mn	Cr	Ni	Cu	Si	P	C
B1-1					3.1	106	5.52	3.3	0.0052	3.35	61.3	6.6	0.2	8.5	10.5	3.5	0.5	0.1
B1-2	0.21	0.63	0.009	0.17	6.4	125	3.38	3.7	0.0065	3.42	59.8	7.6	0.2	9.4	8.3	5.3	0.6	0.1
B1-3					12.1	165	4.28	4.1	0.0103	3.91	58.1	8.1	0.2	10.5	7.2	6.8	0.5	0.1
B4-1					3.2	89	1.31	1.8	0.0031	3.43	62.1	5.6	0.1	8.2	10.9	3.4	0.7	0.0
B4-2	0.17	0.62	0.009	0.15	6.4	119	1.71	2.4	0.0056	3.71	61.3	7.2	0.3	9.4	6.1	5.1	0.6	0.1
B4-3					12.9	160	3.22	3.4	0.0082	3.75	60.8	6.5	0.2	11.4	4.8	7.3	0.6	0.1
B5-1					3.1	72	1.02	2.1	0.0020	2.82	61.0	7.7	0.2	9.8	5.3	4.4	1.3	0.1
B5-2	0.1	0.59	0.009	0.14	6.4	104	1.40	2.0	0.0034	3.32	63.0	7.1	0.3	8.8	4.9	5.0	1.0	0.0
B5-3					12.0	137	2.14	2.4	0.0066	3.98	60.0	7.7	0.2	11.6	3.1	7.9	0.7	0.1
B6-1	0.1	0.92	0.008	0.13	8.6	126	0.50	4.7	0.0069	3.26	60.5	6.6	0.1	12.9	2.8	6.3	1.1	0.0
B7-1	0.1	0.63	0.017	0.14	8.6	116	0.70	3.9	0.0051	3.08	61.6	6.3	0.1	10.3	4.0	6.7	1.1	0.1
B8-1	0.1	0.63	0.009	0.32	8.7	98	0.59	2.2	0.0032	3.31	63.1	6.1	0.2	8.8	2.2	9.8	1.5	0.0
B9-1					3.1	16	6.08	0.3	0.0006	3.18	66.0	5.4	0.2	10.1	0.2	10.4	1.7	0.1
B9-2	0.04	0.62	<0.005	0.22	6.3	29	5.19	1.2	0.0017	3.30	62.3	4.7	0.1	12.0	0.3	12.6	0.6	0.1
B9-3					12.1	66	3.04	1.0	0.0024	3.97	57.8	4.7	0.1	14.7	0.3	15.8	0.1	0.0
W1-1					3.3	119	2.39	5.7	0.0076	3.11	61.0	6.8	0.1	10.1	7.3	5.2	0.7	0.1
W1-2	0.2	0.88	0.016	0.3	8.6	151	1.05	5.5	0.0085	3.27	61.4	6.5	0.1	10.4	5.1	7.6	0.7	0.1
W1-3					11.8	175	1.18	5.2	0.0113	3.69	58.5	6.6	0.1	11.9	5.1	9.0	0.7	0.1
W2-1					3.2	80	1.22	3.1	0.0029	2.77	62.5	6.5	0.1	9.8	4.7	6.1	1.0	0.2
W2-2	0.13	0.86	0.016	0.3	8.9	139	0.61	6.8	0.0087	3.09	61.4	7.0	0.1	11.6	2.8	8.0	0.7	0.1
W2-3					11.2	150	0.62	4.3	0.0093	3.71	58.7	5.2	0.1	12.9	3.2	11.0	0.7	0.0
S1-1					3.2	45	0.51	1.8	0.0015	2.61	56.8	5.7	0.4	10.6	3.3	9.4	1.2	0.0
S1-2	0.09	0.62	0.007	0.26	5.9	79	1.00	3.2	0.0032	2.85	57.9	7.3	0.3	10.9	3.3	8.6	0.3	0.1
S1-3					11.7	128	1.31	5.4	0.0071	3.08	61.4	6.5	0.3	10.6	2.3	8.8	0.2	0.1

Cluster size



Cluster size changes with fluence in all materials.



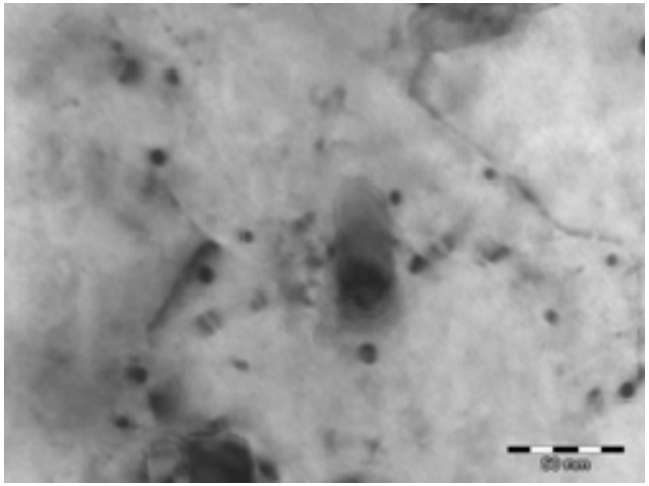
Transition temperature shifts have good correlation with the square root of the volume fraction of solute atom clusters.

TEM Observation

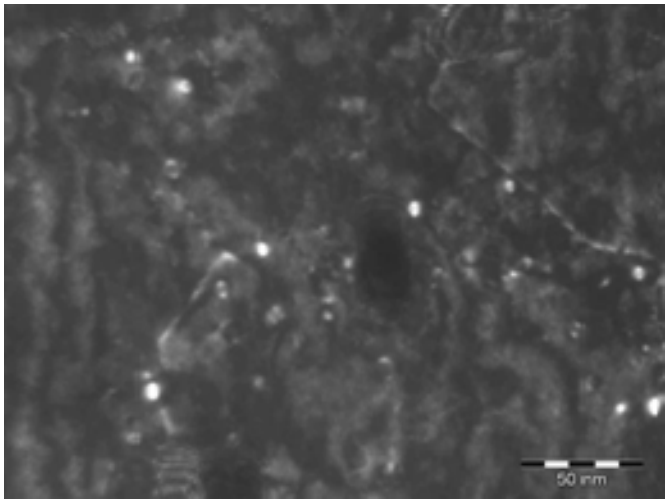
- The matrix damage is considered for another cause of irradiation embrittlement. So, the microstructual observation using TEM were performed for the materials.

- Several dislocation loops could be found in materials “B9-1”, “B9-2”and “B9-3”. It can be understand that as the fluence is increased, the density of the dislocation loops will be incresed.

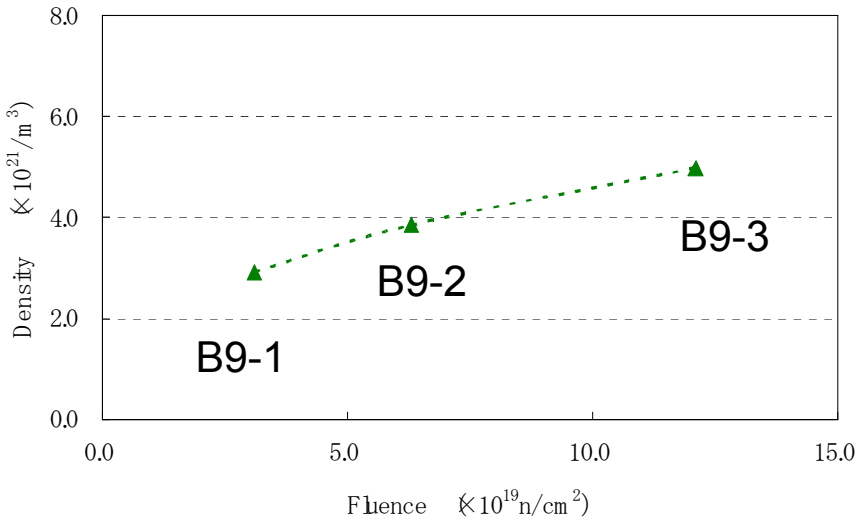
Results of TEM Observation (B9-3)



a) bright field image; B9-3



b) dark field image; B9-3



Irradiation Test at OECD/Halden reactor

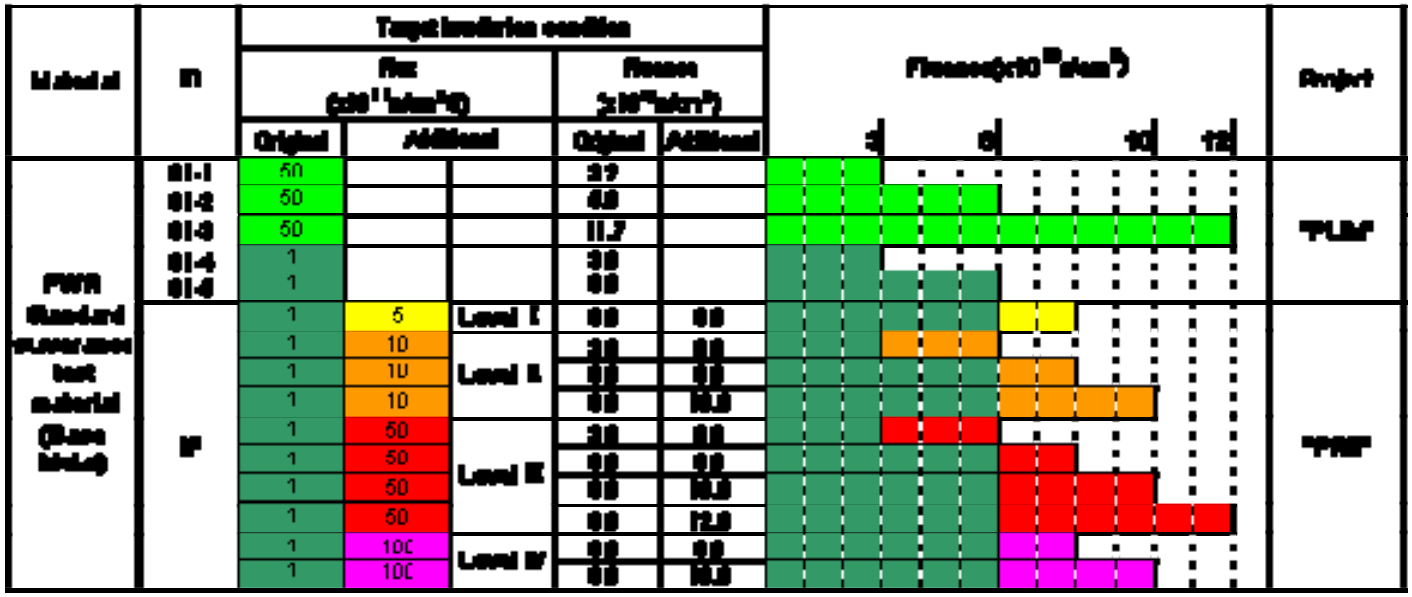
-The RPV materials irradiated in the actual PWR plant have been re-irradiated in the OECD/Halden reactor by several different flux rates.

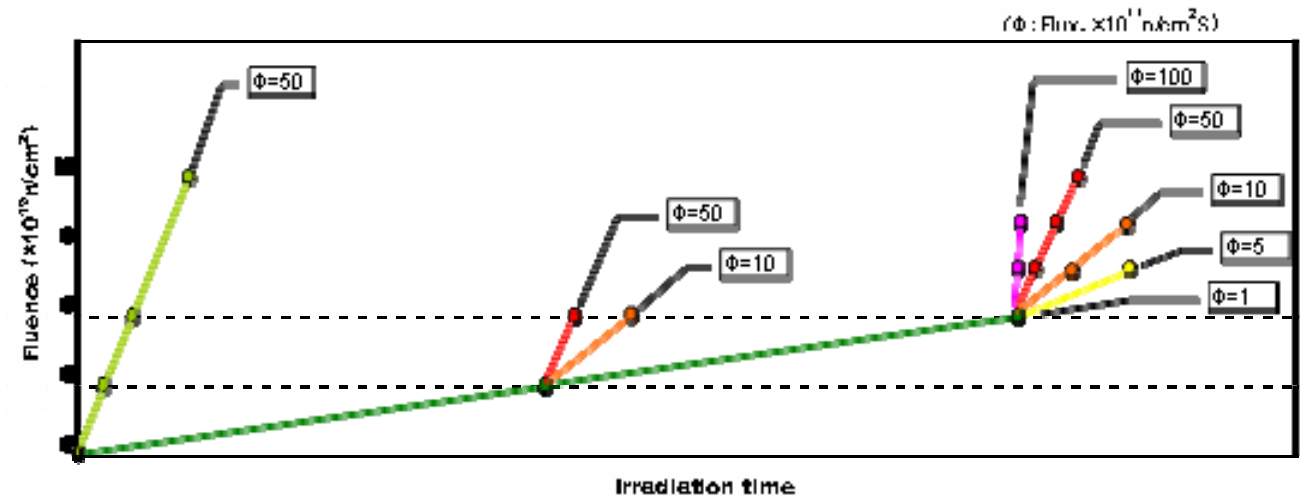
The purpose is to make clear how dose the flux rate effect to the embrittlement.

-Irradiation of the high flux such as Level-Ⅲ(flux;50×10^{11}n/cm^{2}s) and Level-Ⅳ(flux;100×10^{11}n/cm^{2}s) had been already completed and microstructual observation for these materials had been finished .

-Irradiation of the low flux test such as Level- Ⅰ (flux;5×10^{11}n/cm^{2}s) and Level-Ⅱ(flux;10×10^{11}n/cm^{2}s) had been completed. Observation for these materials will be performed this year.

Irradiation Test at OECD/Halden reactor



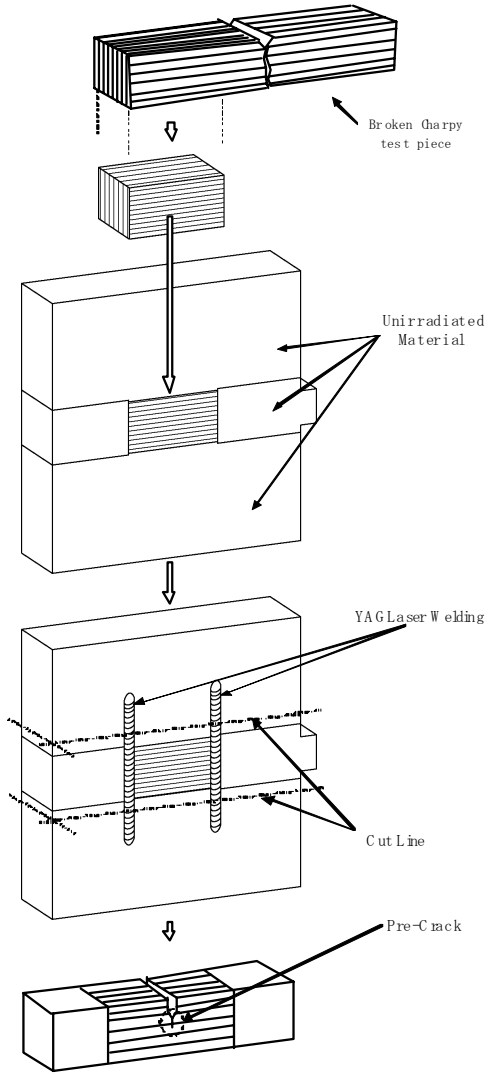


Verification for the Fracture Toughness Evaluation method

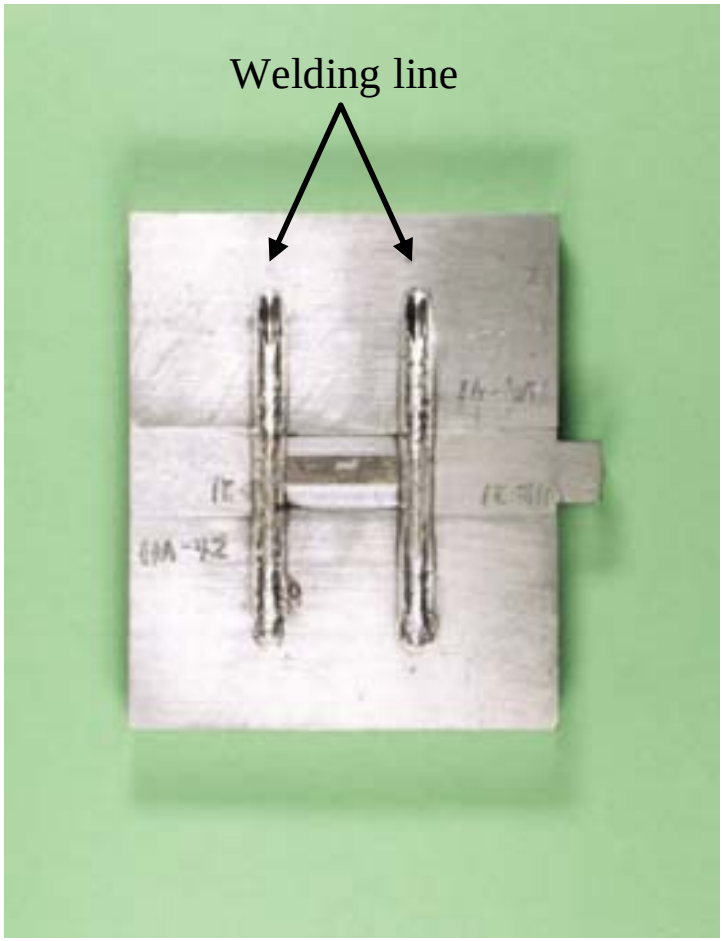
-In the “PRE” project, the verification for the method itself to obtained the reference temperature shift (ΔRT_{NDT}) has been performed.

-In Japanese code, ΔRT_{NDT} is defined as the shift of temperature corresponding to the 41J level of Charpy V notch absorbed energy. In this project, this value ΔT_{41J} is verified by the comparison with the value of ΔT_0 of the Master curve method shown in ASTM E1921-03.

-The value of ΔT_0 is obtained directly from the results of three point bending fracture toughness test. The irradiated 3-point bending test pieces are made by reconstitution from the broken halves of the Charpy impact test specimens.



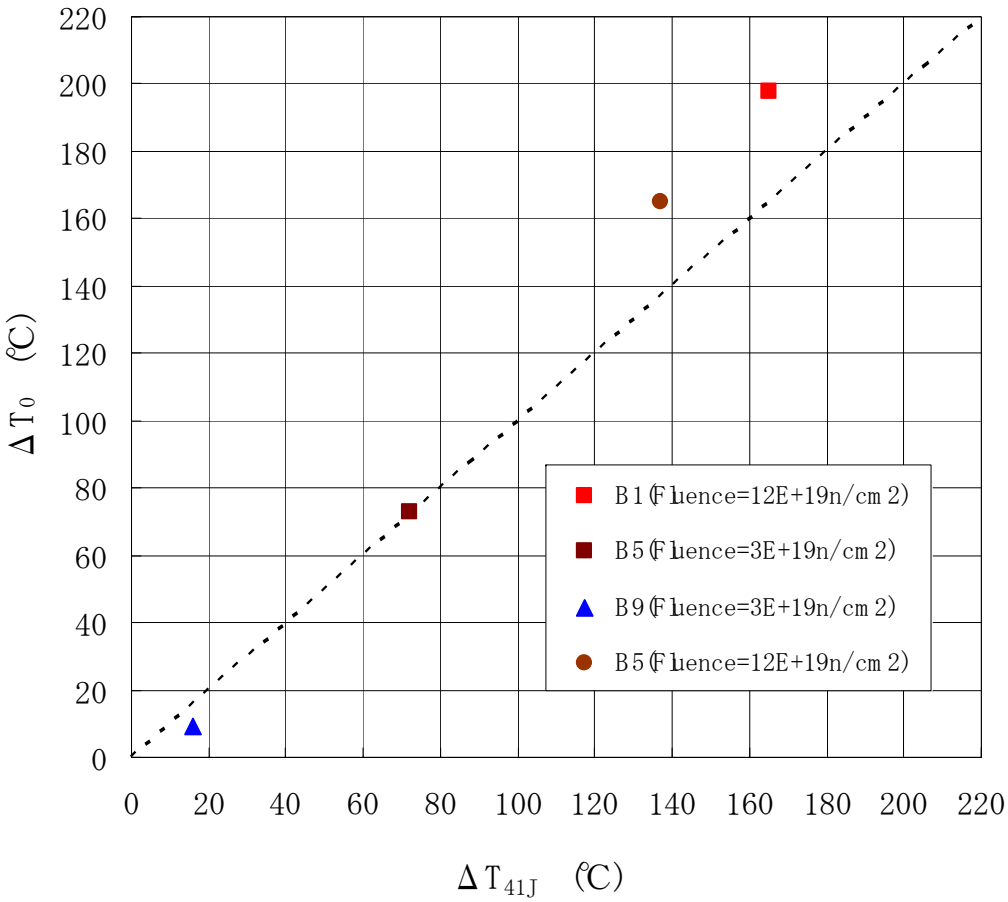
Procedure of re-built-up the 3 point bending specimen



reconstituted

after weld

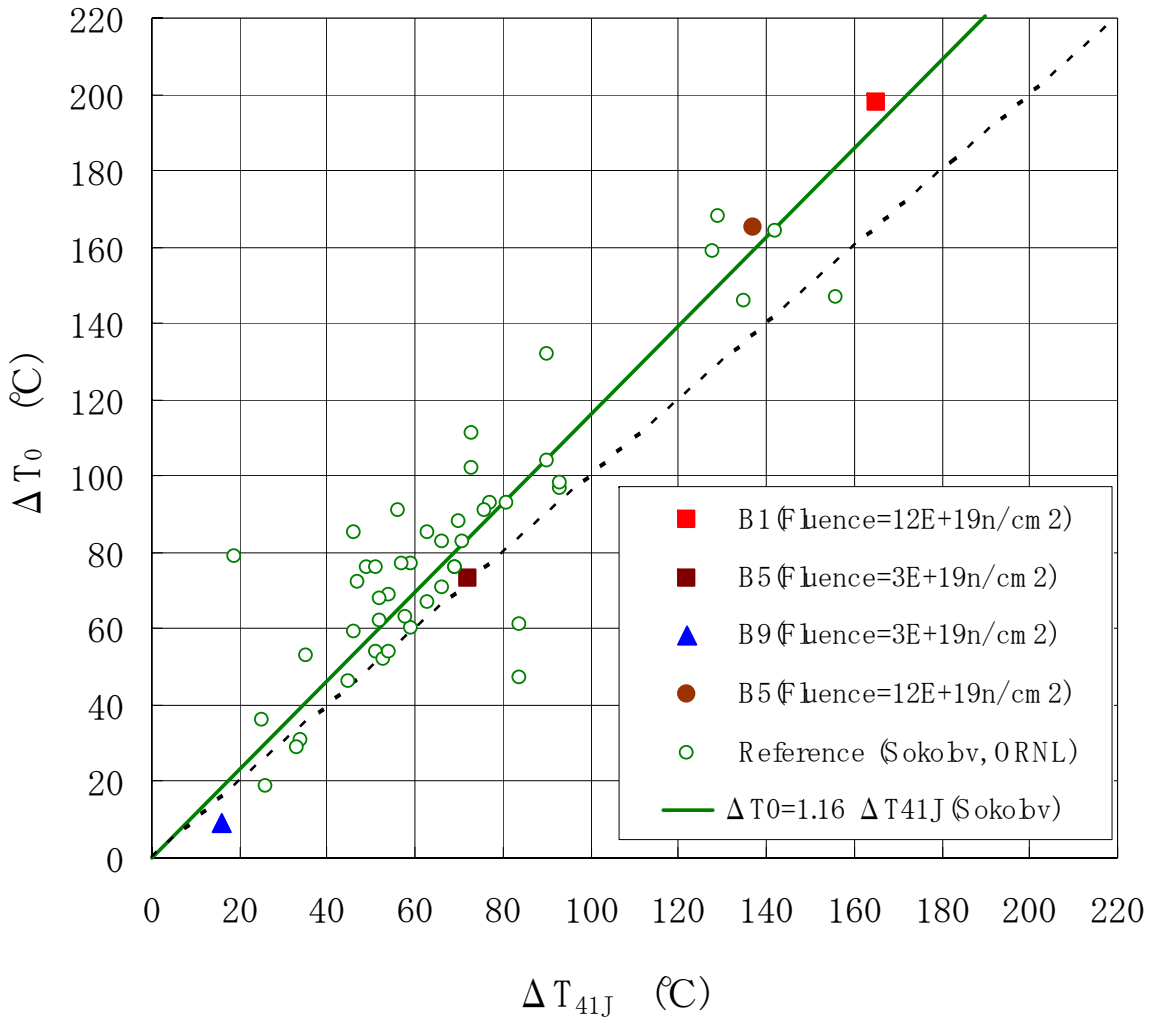
Photpgragh of the re-built-up 3 point bending specimen



Comparison between ΔT_0 and ΔT_{41J}

In the low fluence region (nearly 3×10^{19}n/cm^2), both values were well corresponded, but in the high fluence region (nearly 12×10^{19}n/cm^2), ΔT_0 were about 30℃ greater than ΔT_{41J}.

Comparison between ΔT_0 and ΔT_{41J} with reference data from Sokolov et al.



The same tendency was reported in the reference paper by Sokolov et al.[1] Figure shows that the data from Sokolov et al. were plotted on the test results obtained in this project. The differences between ΔT_0 and ΔT_{41J} will be evaluated furthermore in this project.

[1] Sokolov, M. A., Nanstad, R. K., "Comparison of Irradiation-Induced Shifts of KJc and Charpy Impact Toughness for Reactor Pressure Vessel Steels", NUREG/CR-6609, November 2000.

Thank you for attention!