CRIEPI Research Activities on Neutron Irradiation Embrittlement of RPV and Core Internals

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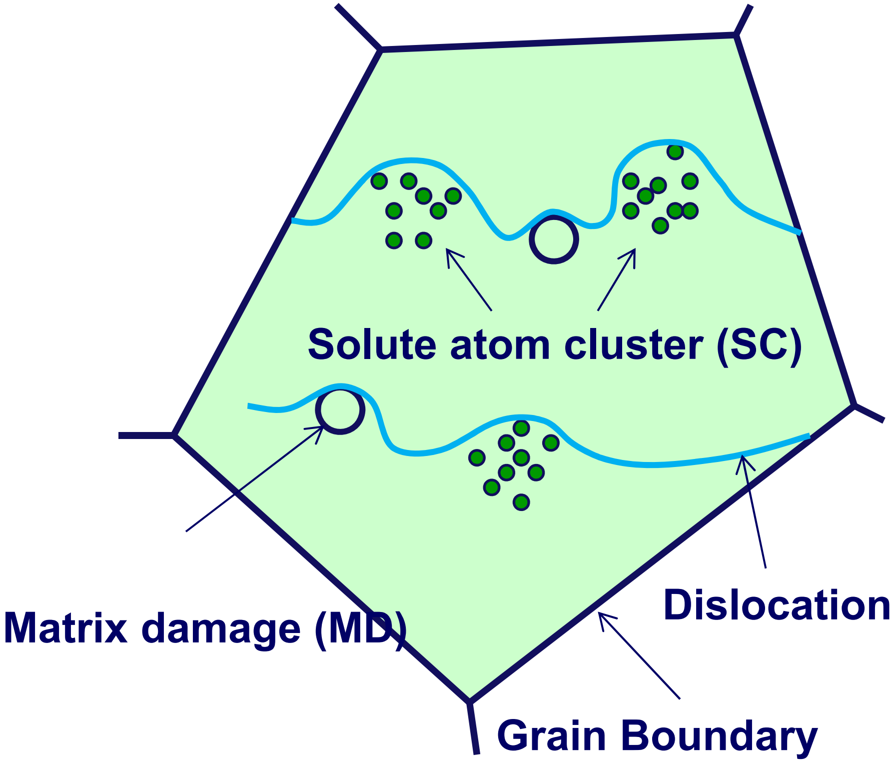
    - Elucidation of irradiation effect on stainless steels and development of correlation model between microstructure and mechanical properties

Development of new Japanese embrittlement trend curve(ETC)

- JEAC4201-2007 adopted the CRIEPI's model which is guided by the understanding on the mechanism of embrittlement
  - Atom Probe Tomography (APT) data were very limited
- JEAC4201-2013 is a revised ETC of JEAC4201-2007
  - Baseline model remained the same
  - Coefficients were re-calibrated using the high-fluence surveillance data
- Current situation
  - No new surveillance data have been obtained over the last couple of years.
  - A large mount of microstructural data, particularly the Atom Probe Tomography (APT) data, have been accumulated up to now.
  - CRIEPI started developing new ECT to revise the current JEAC4201-2013 ETC from 2016.

JEAC 4201, Method of Surveillance Tests for Structural Materials of Nuclear Reactors , Japan Electric Association,

Basic idea of microstructural changes in RPV steel



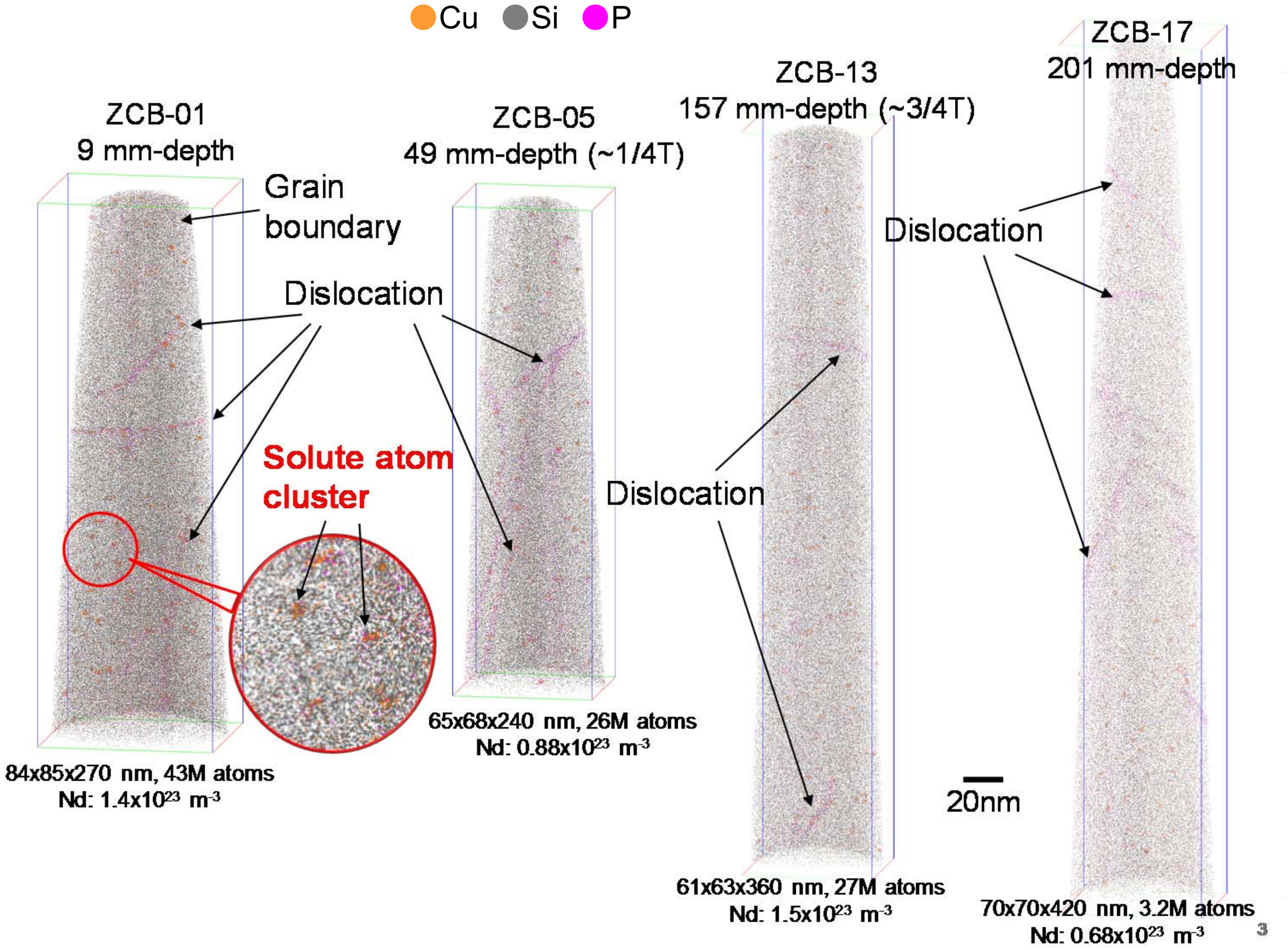
KEY PLAYERS

- **Solute atom cluster**
  - ➢ Irradiation enhanced cluster (homogeneous nucleation)
  - ➢ Irradiation induced cluster (heterogeneous nucleation)

→ detected by APT

- **Matrix damage**
  - ➢ Naked dislocation loops?

Example of 3D atom maps (Zion unit 1 RPV)



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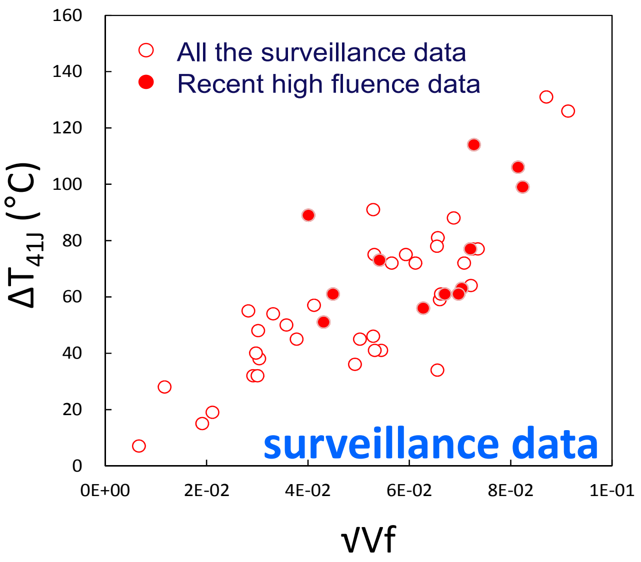
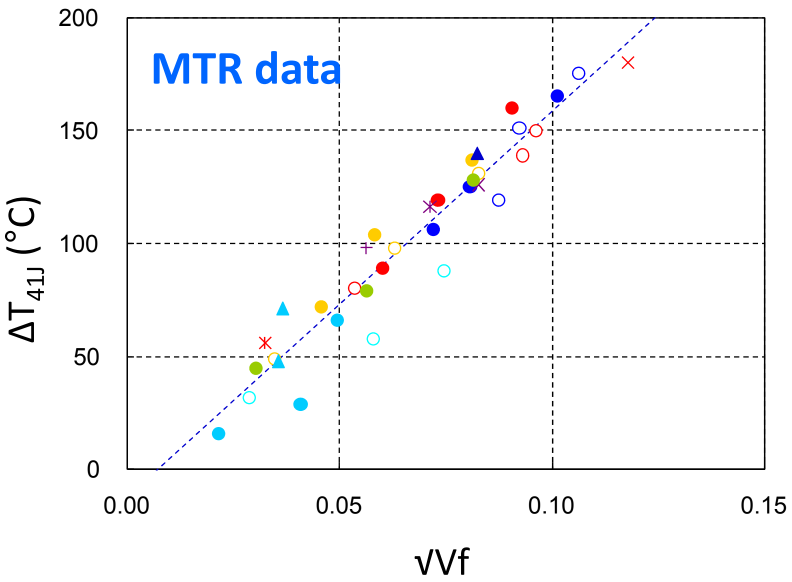
Basic idea on transition temperature sift (TTS) calculation





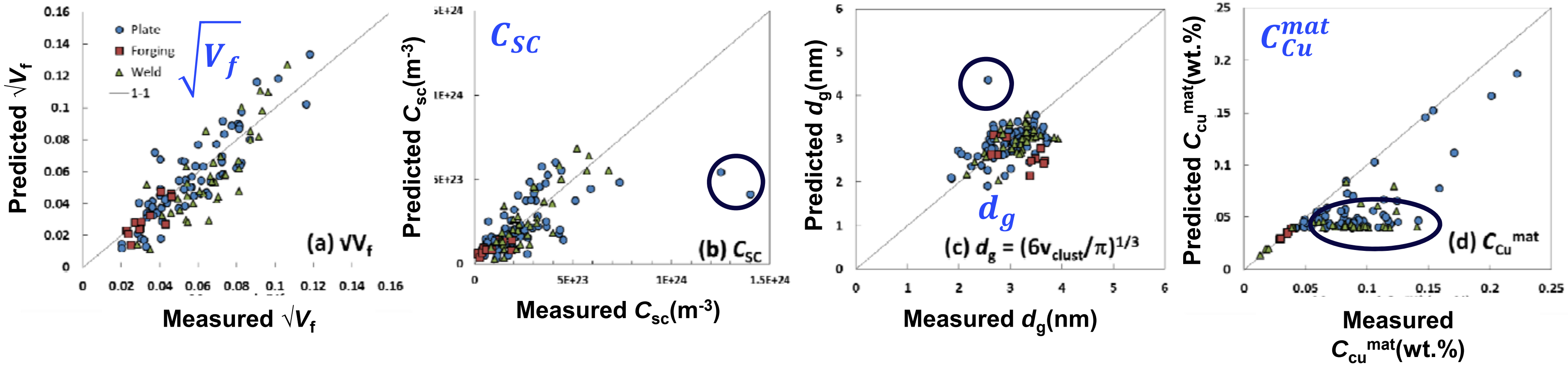
$$TTS = \sqrt{\left(a\sqrt{V_f}\right)^2 + \left(b\sqrt{C_{MD}}\right)^2}$$

Prediction of V_f is the key.
We revisited the prediction model of microstructure change.

Y. Hashimoto et.al,, fontevraud 9, 2018.

V_f: volume fraction of solute atom cluster, C_{MD}: number density of matrix damage

JEAC4201-2013 prediction of microstructure change



The V_f prediction is in good agreement with the APT data.
The trends of predictions of C_{sc}, d_g and matrix Cu are roughly similar to the APT data.
There is room for improvement of prediction performance

Generalized form, Consideration of other elements, Fitting to the APT data and No use of differential equations

V_f: volume fraction of solute atom cluster, C_{SC}: number density of solute atom cluster,
d_g: bulk Cu content, C_{Cu}^{mat}: Cu content in matrix

Y. Hashimoto et.al,, fontevraud 9, 2018.

Analytical solution of revised model

Once the chemical composition, temperature and flux are given, the variables are functions of time as follows:

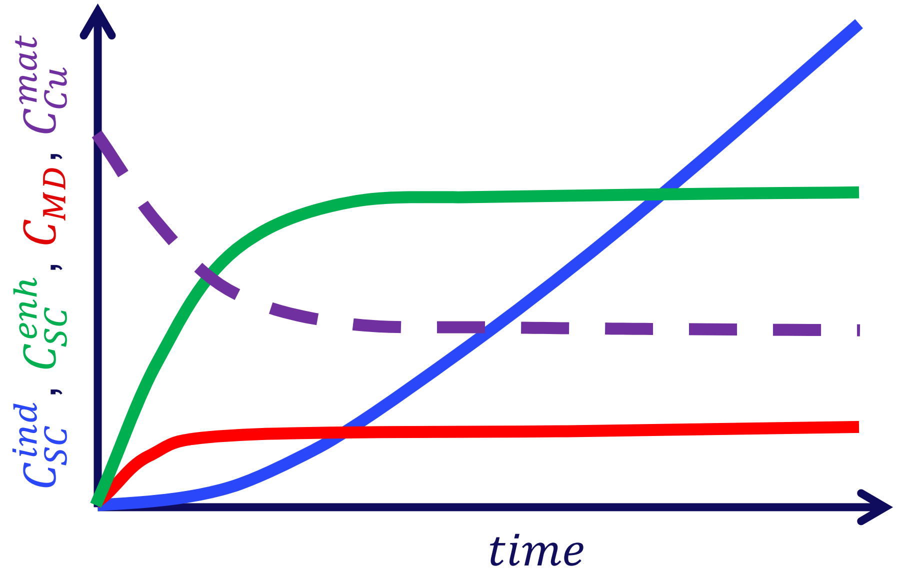
$$C_{SC}^{ind} = \gamma_1 t - \frac{\gamma_1}{\alpha_1}(1 - \exp(-\alpha_1 t))$$

$$C_{SC}^{enh} = \beta_4 \left(1 - \frac{1}{\beta_3 \beta_4 t + 1}\right)$$

$$C_{MD} = \frac{\gamma_1}{\alpha_1}(1 - \exp(-\alpha_1 t))$$

$$C_{Cu}^{mat} = C_{Cu}^{0} - \delta_1 C_{SC}^{enh}$$

$$V_f = 1.1 v_{clust}(C_{SC}^{ind} + C_{SC}^{enh})$$



C_{MD}: number density of matrix damage
C_{SC}^{ind}: number density of irradiation induced solute atom cluster
C_{SC}^{enh}: number density of irradiation enhanced solute atom cluster
C_{Cu}^{mat}: Cu content in matrix
v_{Clust}: average cluster volume

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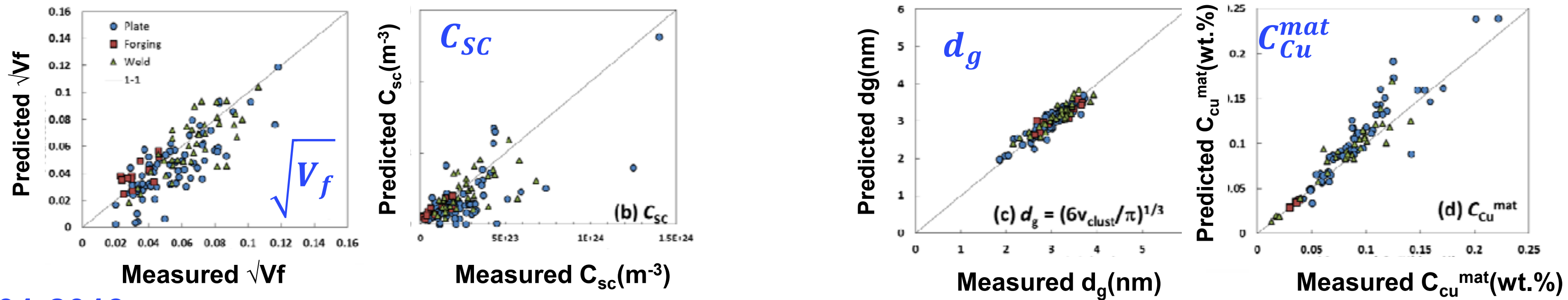
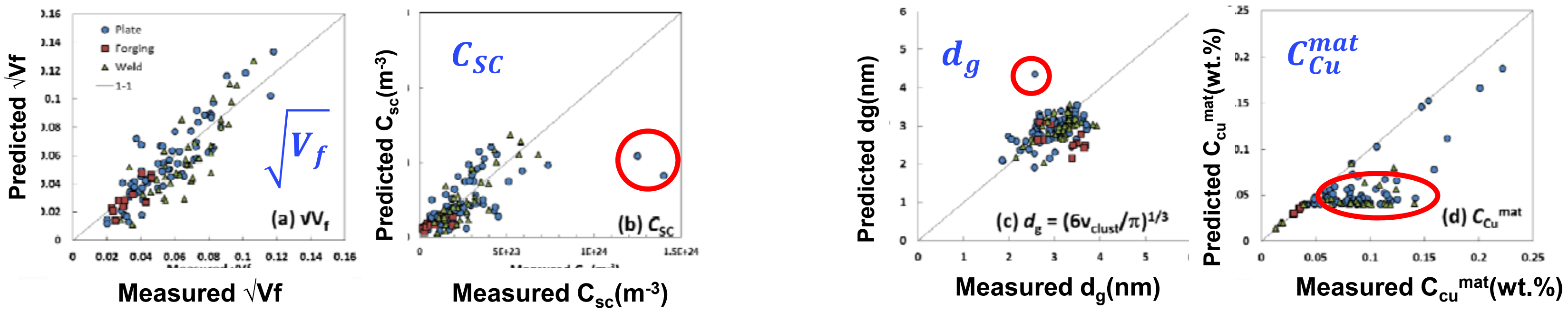
Prediction of microstructure changes

$N_{clust}^{meas} \geqq 50$

Analytical solution



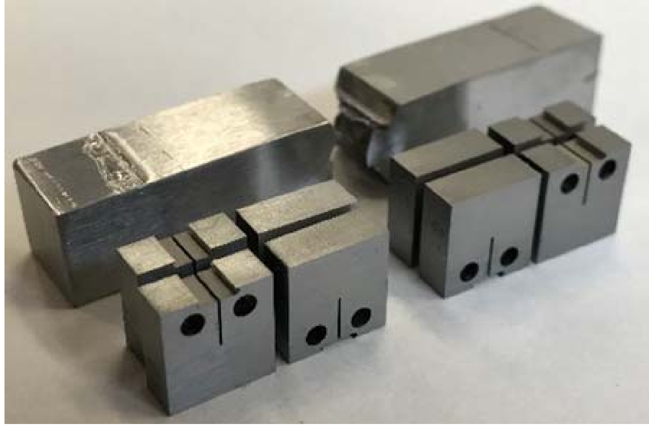
JEAC4201-2013



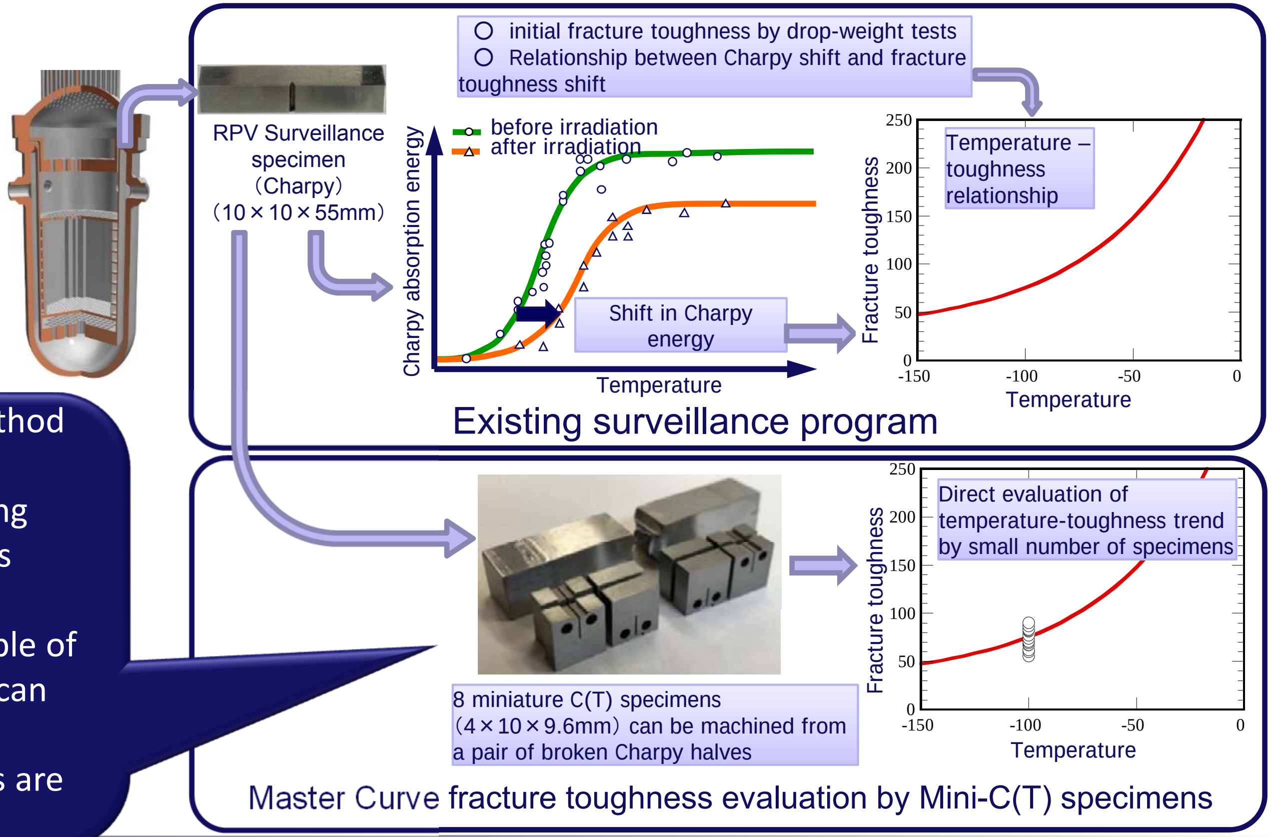
- The draft revised ETC model have been reviewed by experts of academic society in Japan.
- We would like to propose new ETC model to Japan Electric Association as soon as the review is over.
- We expect that new ETC can be implemented into JEAC 4201 in 2019 or 2020 JPY.

Y. Hashimoto et.al,, fontevraud 9, 2018.

Development of miniature C(T) master curve method



The Master Curve + Miniature C(T) specimens



RPV Surveillance specimen (Charpy) (10×10×55mm)

○ initial fracture toughness by drop-weight tests
○ Relationship between Charpy shift and fracture toughness shift

Shift in Charpy energy

Temperature – toughness relationship

Existing surveillance program

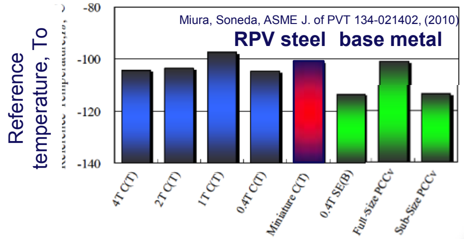
8 miniature C(T) specimens (4×10×9.6mm) can be machined from a pair of broken Charpy halves

Direct evaluation of temperature-toughness trend by small number of specimens

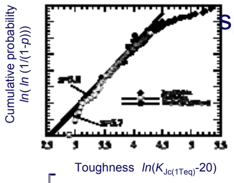
Master Curve fracture toughness evaluation by Mini-C(T) specimens

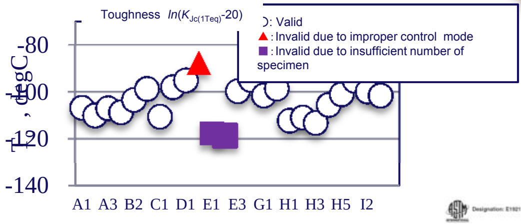
The **Master Curve** method is a method to **directly determine fracture toughness** by statistically processing data from small number specimens based on Weibull distribution.
The **Master Curve** method is capable of the **specimen size correction**. We can use very small specimens if the appropriateness of test techniques are confirmed

Development of Mini-C(T) technique

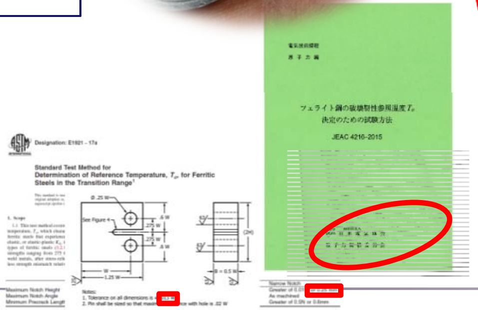

Miura, Soneda, ASME J. of PVT 134-021402, (2010)

Consistency of toughness between Mini-C(T) and larger specimens





Round robin test for confirmation of reproducibility


Designation: E1921 – 17a

Standard Test Method for Determination of Reference Temperature, T_o, for Ferritic Steels in the Transition Range[1]

フェライト鋼の破壊靭性参照温度T_o
決定のための試験方法

JEAC 4216-2015

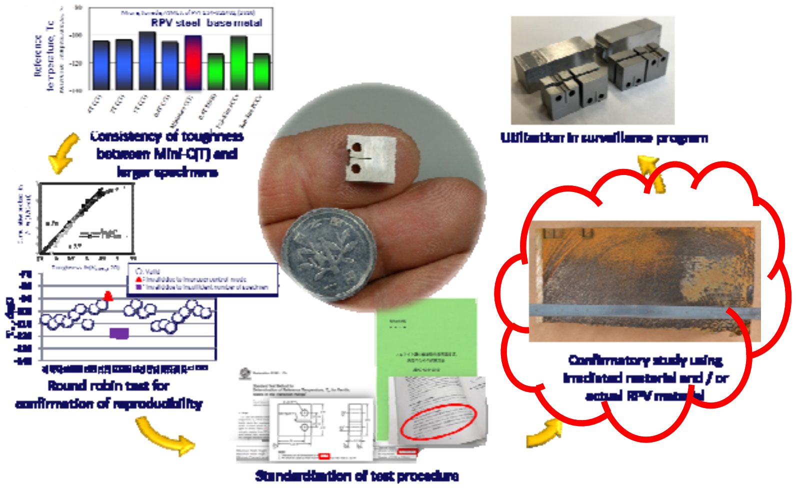
Standardization of test procedure

Confirmatory study using irradiated material and / or actual RPV material

Utilization in surveillance program

Step by step results

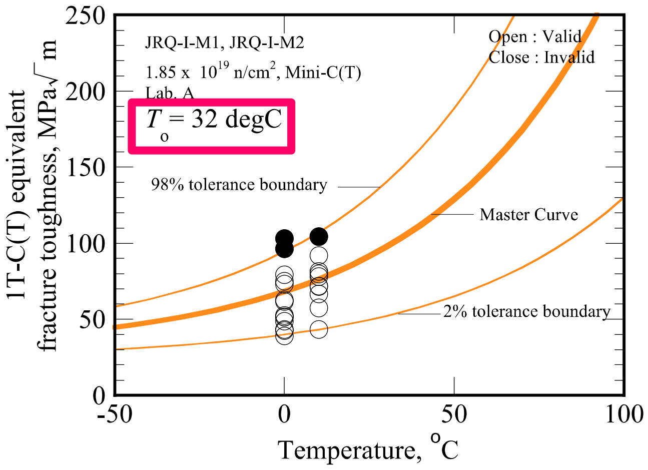
ONGOING CONFIRMATORY STUDY USING IRRADIATED MATERIALS



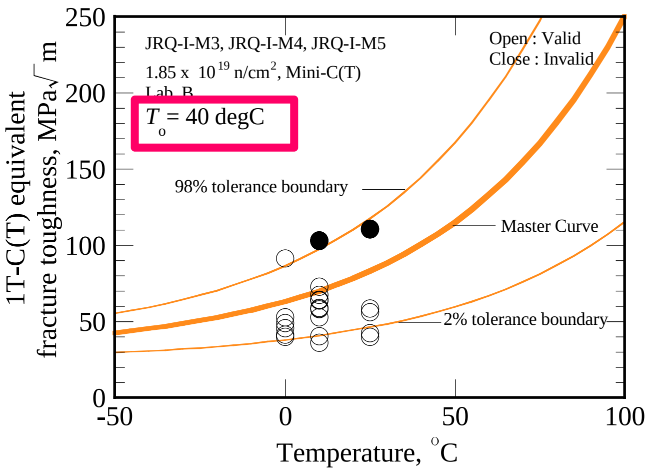
Irradiated material

- ✓ IAEA JRQ plate
- ✓ Linde 80 weld metal
- ✓ Zion Unit RPV base metal

Mini-C(T) application on IAEA irradiated JRQ



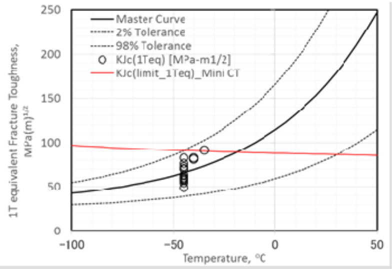
Irradiated Mini-C(T) by Lab. A



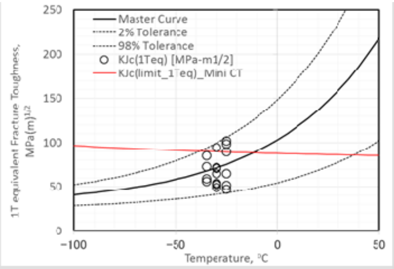
Irradiated Mini-C(T) by Lab. B

- ➢ T_o could be evaluated in both of Laboratories A and B on irradiated JRQ.
- ✓ T_o from Labs. A and B (32°C, 40°C) is consistent with each other.
- ✓ 5 of 25 (20%) Mini-C(T) specimens from Lab. A are out of 2 and 98 % tolerance boundary.
- ✓ 3 of 24 (13%) Mini-C(T) specimens from Lab. B are out of 2 and 98 % tolerance boundary.

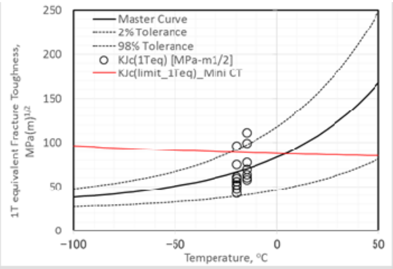
Mini-C(T) application on Zion unit 1 (decommissioned PWR) RPV plate material



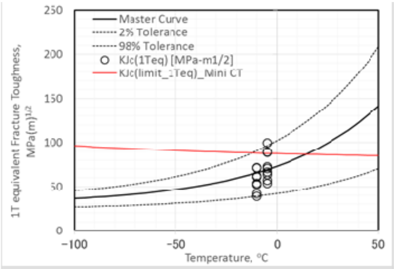
Layer 1, T-L, T_0= -9.7°C



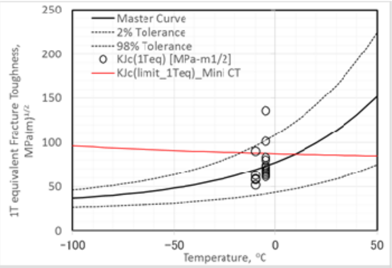
Layer 2, T-L, T_0= -2.2°C



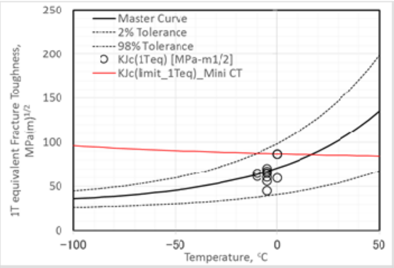
Layer 3, T-L, T_0=14.5°C



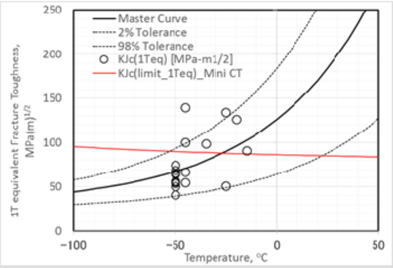
Layer 4, T-L, T_0=25.5°C



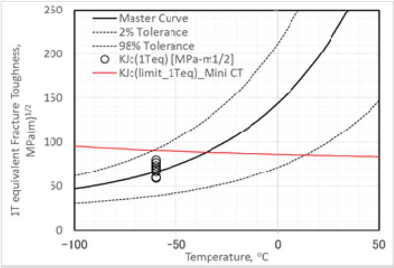
Layer 5, T-L, T_0=20.7°C



Layer 7, T-L, T_0=28.8°C

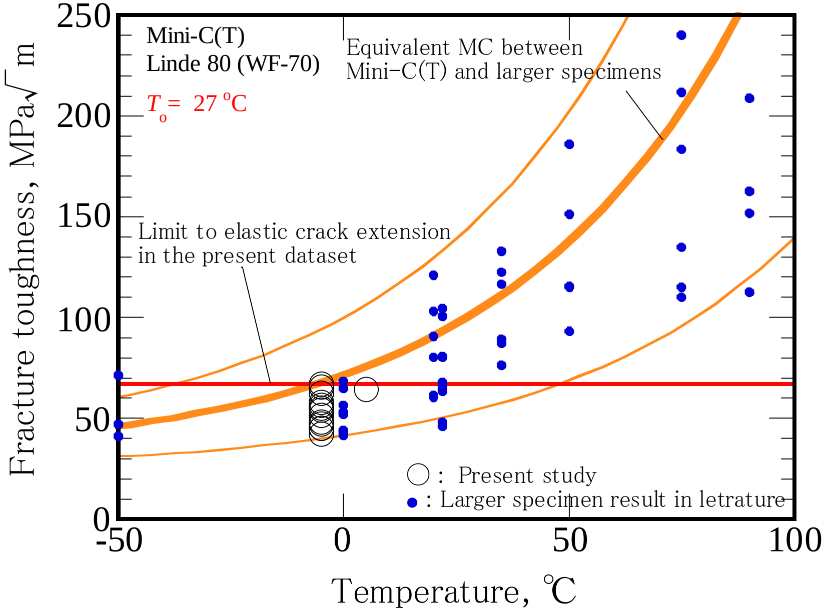


Layer 16, T-L, T_0= -16.6°C



Layer 17, T-L, T_0= -25.7°C

Mini-C(T) application on low upper shelf weld metal, Linde 80, in irradiated state



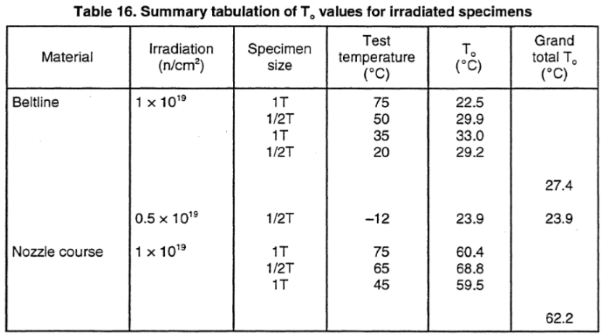


Table 16. Summary tabulation of T_o values for irradiated specimens

Material	Irradiation (n/cm^2)	Specimen size	Test temperature (°C)	T_o (°C)	Grand total T_o (°C)
Beltline	1 × 10^{19}	1T	75	22.5	
		1/2T	50	29.9	
		1T	35	33.0	
		1/2T	20	29.2	
					27.4
	0.5 × 10^{19}	1/2T	−12	23.9	23.9
Nozzle course	1 × 10^{19}	1T	75	60.4	
		1/2T	65	68.8	
		1T	45	59.5	
					62.2

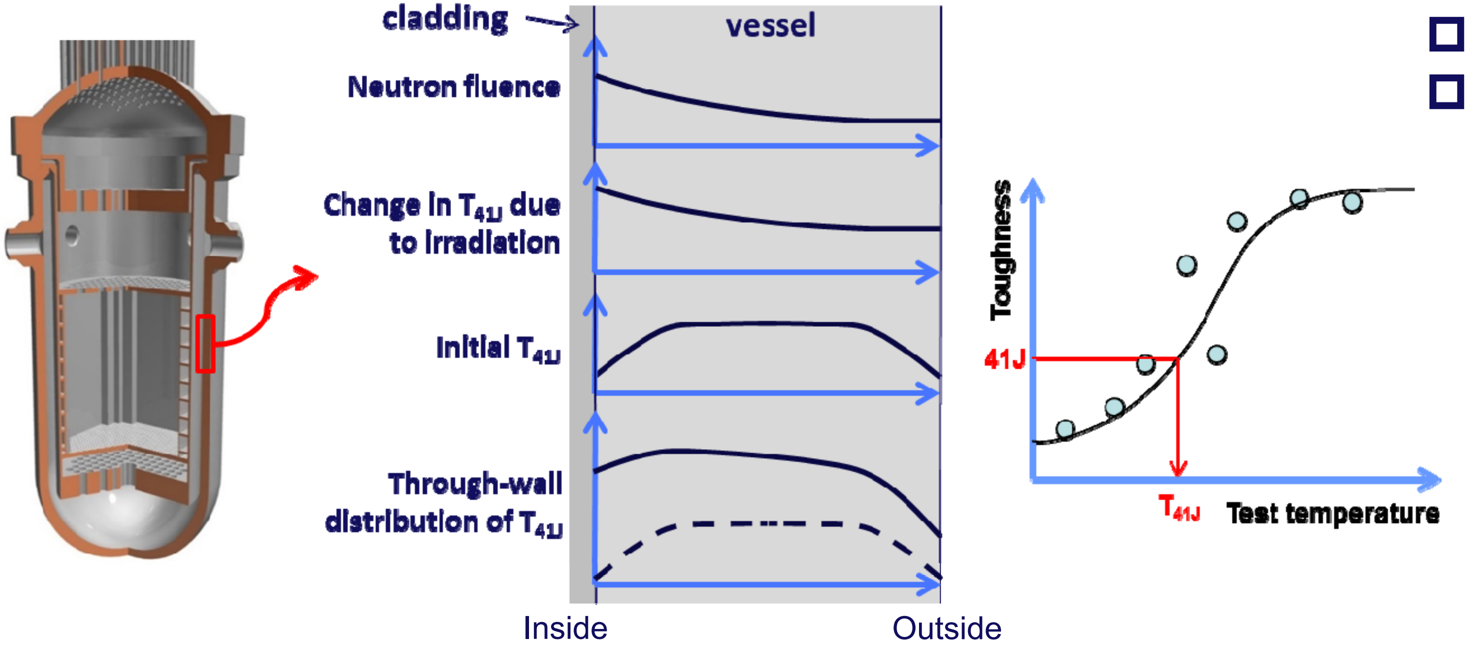
NUREG/CR-5736

- Linde 80 weld metal retrieved from unused Midland NPP RPV beltline weld
- Fluence (Irradiated in HSST project): 1 × 10^{19} n/cm^2 (>1MeV), 288℃
- Excellent agreement between Mini-C(T) and larger specimens

M. Yamamoto, ASME PVP2017-84906

Evaluation of Through-wall attenuation of irradiation embrittlement

- **Investigate combined effect of initial toughness distribution and fluence attenuation in a RPV steel to identify the conservatism in integrity assessment.**



☐ IAEA JRQ

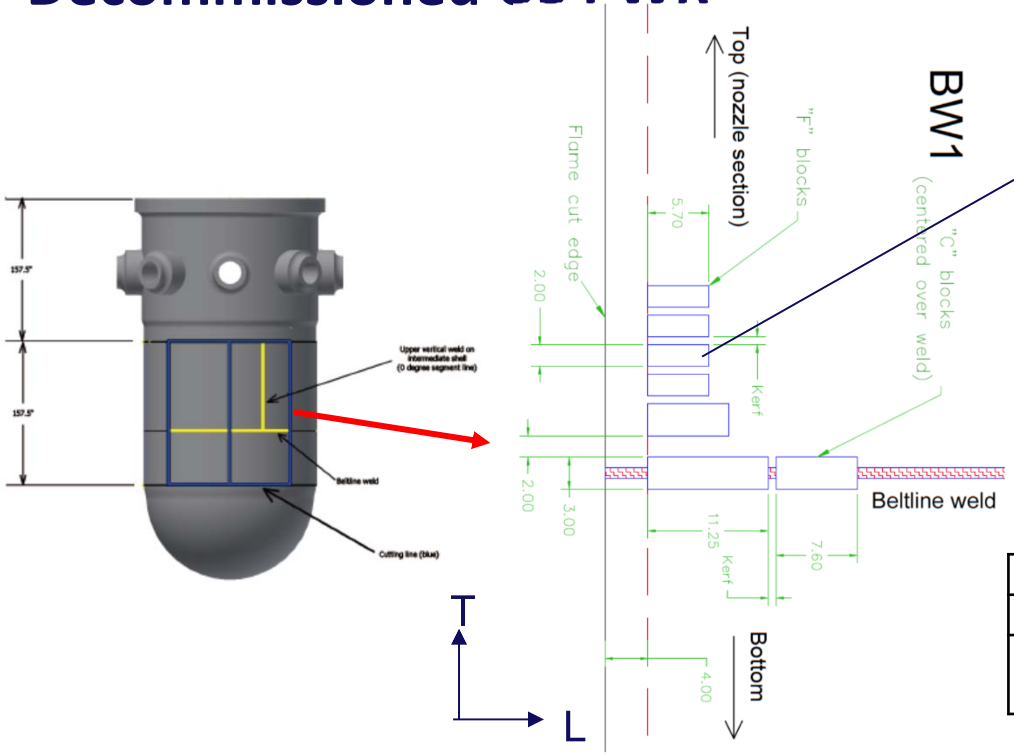
☐ Zion unit 1 RPV

↑

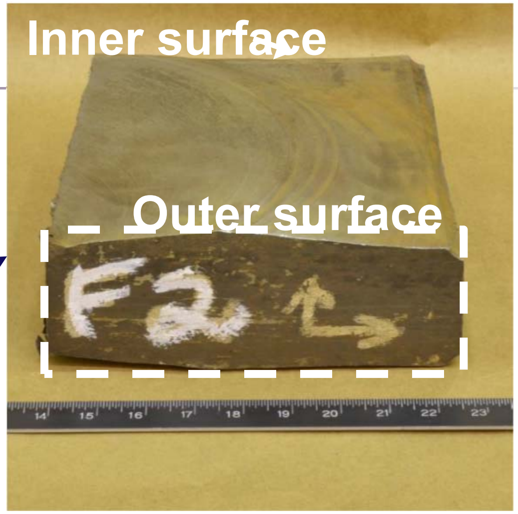
Collaboration with LWRS by ORNL

Zion unit 1 martial

Decommissioned US PWR



View from outside of vessel



- Zion Unit 1 block F02 (L216xT73xS216 mm)
- A533B plate base metal

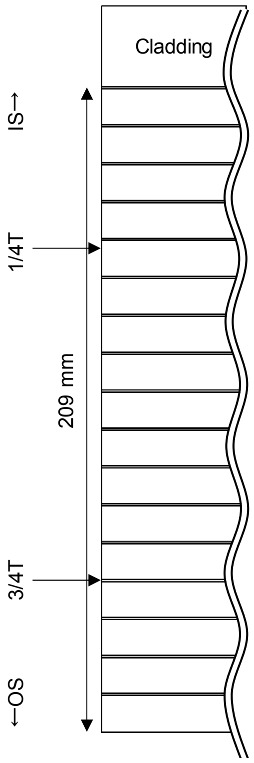
Chemical composition of Zion RPV base metal (wt%)

	Cu	Ni	Mn	Si	P
Nominal	0.11	0.49	1.30	-	0.001
C2 block[1)]	0.11–0.15	0.44–0.58	1.26–1.52	0.20–0.30	0.006–0.016

- Fluence at clad – base metal interface (CBMI) : 7×10^{18} n/cm^2(E > 1MeV)

[1)] ORNL/TM-2018/861

Investigation on Zion Unit 1 material



Layer No.	ID	Sampling depth (mm)	Fluence ($\times 10^{18}$ n/cm^2)	Flux ($\times 10^{10}$ n/cm^2·s)	APT	Vickers hardness	Metallographic	MC(T)_T-L	MC(T)_L-S	EPMA
1	ZCB-01	9	6.4	1.4	✔	✔	✔	✔	✔	✔
2	ZCB-02	19	5.8	1.2	✔	✔	✔	✔	✔	✔
3	ZCB-03	29	5.3	1.1	✔	✔	✔	✔	✔	
4	ZCB-04	39	4.8	1.0	✔	✔	✔	✔	✔	
5	ZCB-05	49	4.4	0.93	✔	✔	✔	✔	✔	✔
6	ZCB-06	59	4.0	0.85	–	–	–	–	–	–
7	ZCB-07	70	3.6	0.77	✔	✔	✔	✔	✔	✔
8	ZCB-08	93	2.9	0.61	–	–	–	–	–	–
9	ZCB-09	117	2.3	0.49	–	–	–	✔	–	–
10	ZCB-10	127	2.1	0.44	–	–	–	–	–	–
11	ZCB-11	138	1.9	0.40	–	–	–	✔	–	–
12	ZCB-12	147	1.7	0.37	–	–	–	–	–	–
13	ZCB-13	157	1.6	0.34	✔	✔	✔	✔	–	✔
14	ZCB-14	169	1.4	0.30	–	✔	–	✔	✔	–
15	ZCB-15	180	1.3	0.27	–	✔	–	✔	✔	–
16	ZCB-16	192	1.1	0.24	–	✔	–	✔	✔	–
17	ZCB-17	201	1.0	0.22	✔	✔	✔	✔	✔	✔

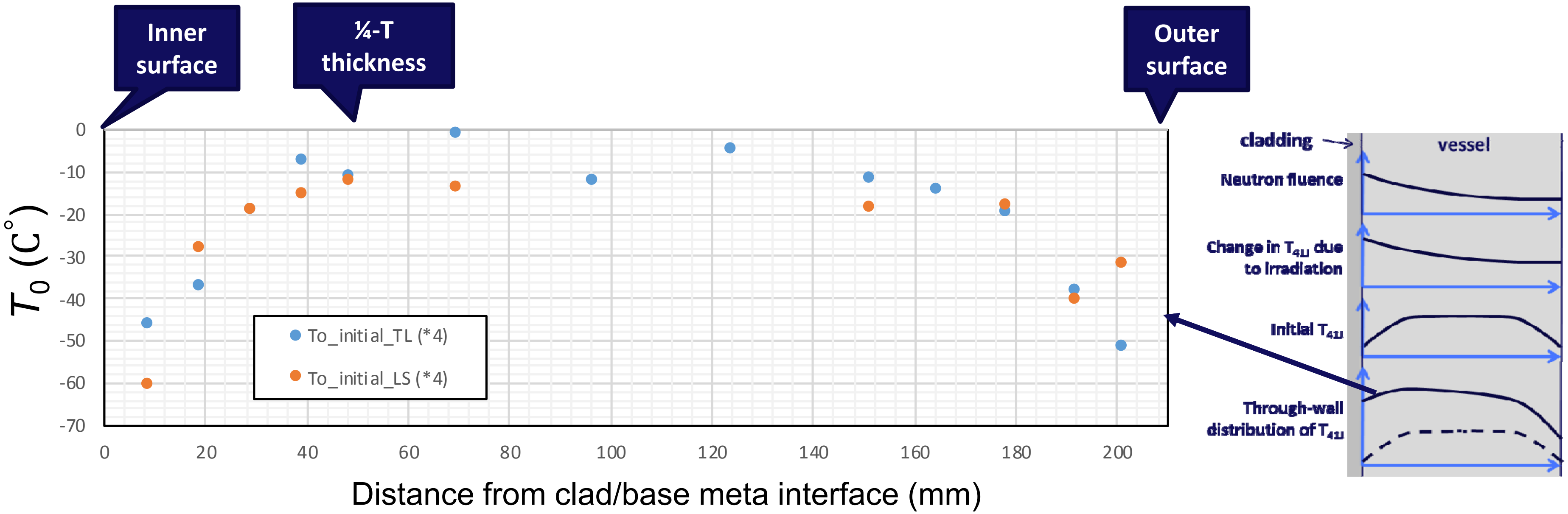
- Fluence is calculated using eq.(1)

$$F = F_0 \cdot \exp(-0.24/25.4 \cdot a) \qquad (1)$$

F_0: fluence at inner surface of RPV
a: sampling depth (mm)

- Flux is calculated using fluence and 15 EFPY

Through-wall Fracture Toughness (T_o) distribution of Zion Unit 1 RPV



- Mini-C(T) technology confirmed the very high initial fracture toughness (low T_o) in inner and outer surface region than ¼-T location, where the surveillance specimens are machined.

Irradiation effect on stainless steels

Back ground

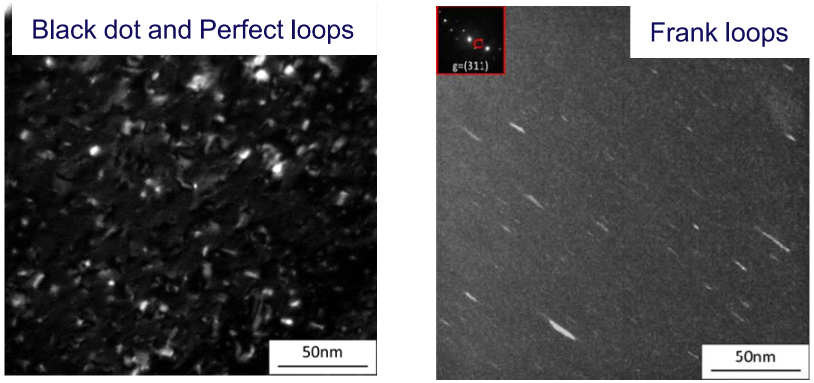
- The microstructural and mechanical properties of the internal components in a light water reactor core change under different operating conditions.
- Conventionally, microstructural evolution has been performed using TEM.
- Recently, trace element segregations at the grain boundaries and solute-enriched clusters in the matrices have been observed by atom probe tomography (APT).
- However, quantitative data supporting these findings are limited.

Objectives

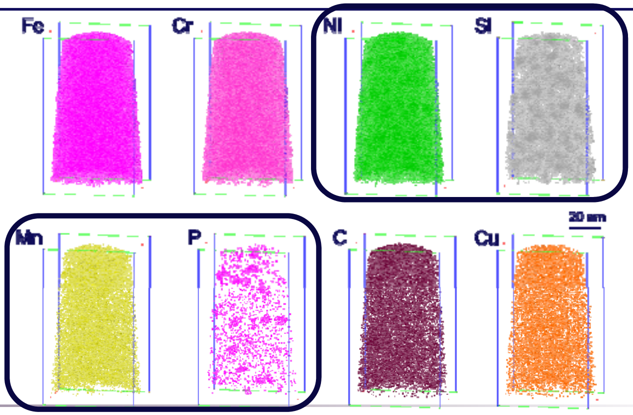
- Development of irradiated microstructure database, including APT as well as TEM.
- Clarify the correlation between microstructure and yield strength (hardness) to estimate the materials degradation(IASCC, fracture toughness).

Microstructural characterization by TEM and APT

Irradiation	**Material**	**Project**
Thermal reactor	316L (used in JNES IASCC project)	Japanese national project (sponsored by METI) “Improvement of Evaluation Methods of Irradiation Effects on Reactor Pressure Vessel and Core Internals” , collaborating with LWRS project
	304L (used in JNES IASCC project)	
Fast reactor	316 (used in CIR project)	
	304L (used in CIR project)	
PWR	347 (BFB from Westinghouse Two-loop down-flow type PWR)	
PWR (Chooz A)	304 (from decommissioned French PWR)	MAI-VIP

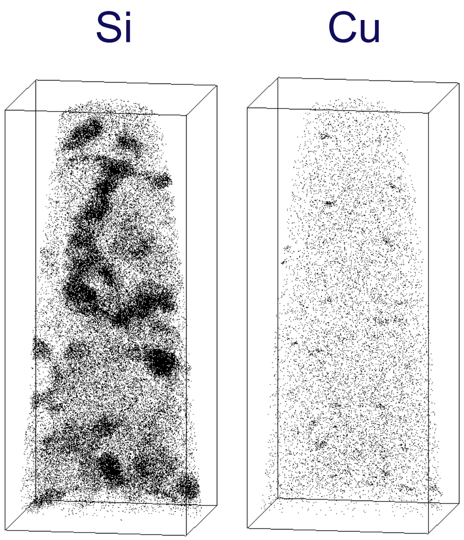


TEM (Black dot and Perfect loop, Frank loop)

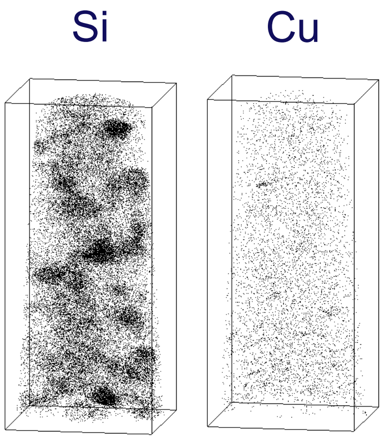


APT (Solute enriched cluster)

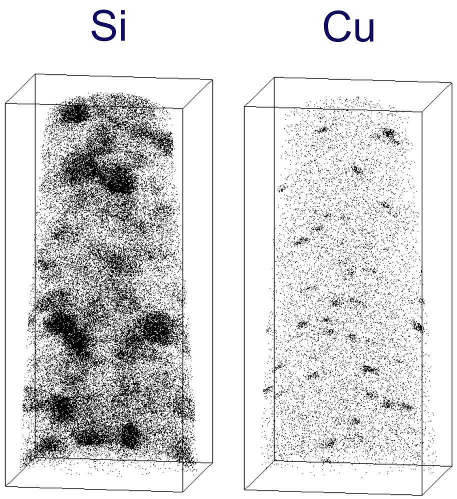
APT Maps: Type 347



14.9 dpa
52 × 54 × 125 nm,
6.0 M atoms

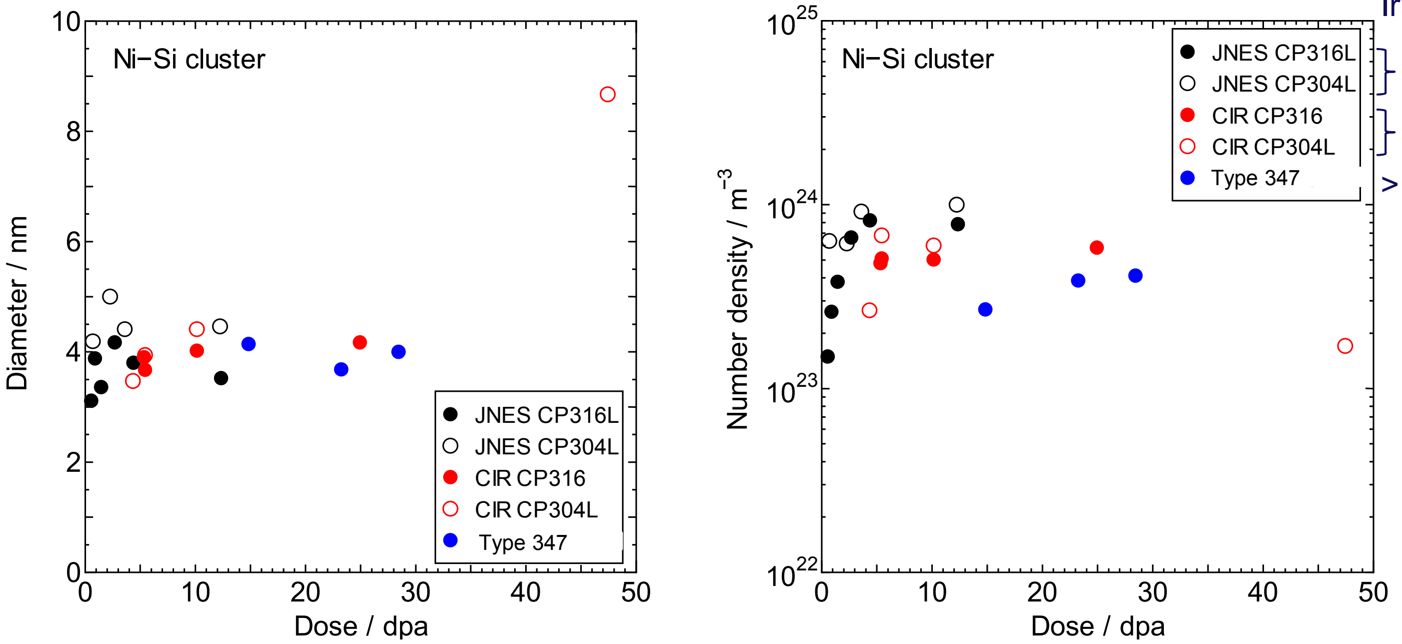


23.3 dpa
42 × 44 × 98 nm,
3.4 M atoms



28.5 dpa
50 × 53 × 110 nm,
5.8 M atoms

APT: Dose dependence of cluster size and number density

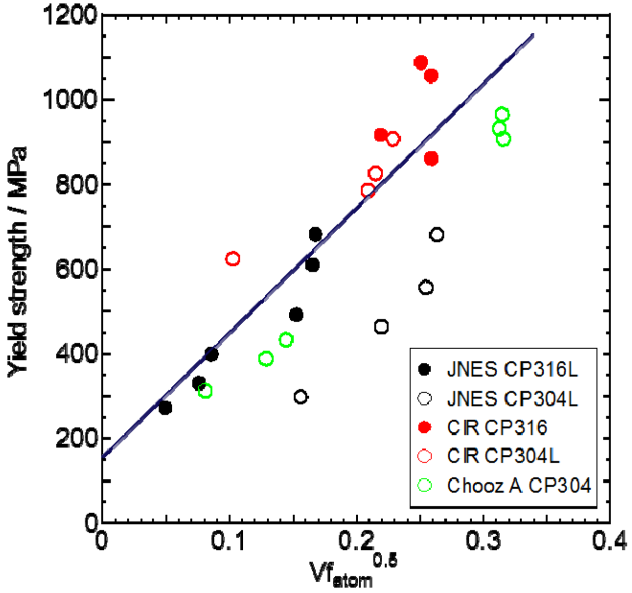


Irradiation temp.
} 288 ℃
} 330 ℃
> 330 ℃ ?

- ➢ Ni-Si cluster size rapidly reaches to 3～5 nm then do not change dose up to 25 dpa.
- ➢ Ni-Si cluster size increase to 8.5 nm at 47.5 dpa
- ➢ Number density of Ni-Si cluster rapidly increase less than 1 dpa then tend to reach saturation level at 5～10 dpa.
- ➢ Number density of Ni-Si cluster tends to be low when the irradiation temperature is high.

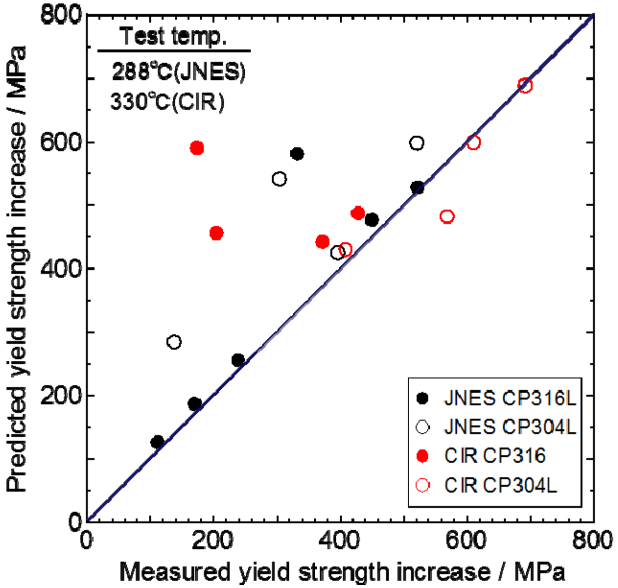
Microstructure and yield strength correlation

Empirical model (APT)



- The $Vf^{1/2}$ of clusters have a relatively good correlation with the yield strength.

Theoretical model (TEM and APT)



- Orowan based dispersed barrier model provide a good correlation with the yield strength increase.

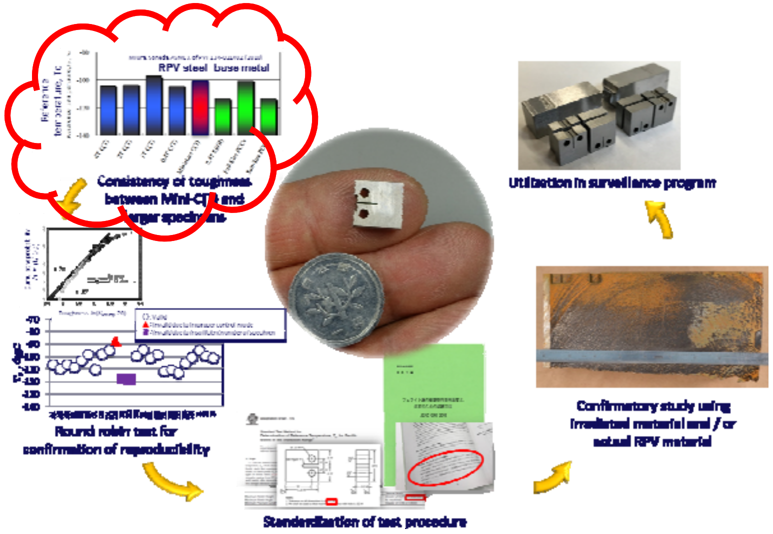
Summary

- Neutron Irradiation embrittlement of RPV
  - Based on the accumulated APT analysis data, the prediction accuracy of the microstructural change by irradiation was improved. We are examining a new ETC that incorporates this improvement.
  - After basic studies on the feasibility of the Mini-C(T) master curve technique, we are conducting research to confirm the applicability to irradiated materials.
  - We investigated through wall distribution of macro and micro-structure, hardness, toughness in Zion unit 1 PRV. We obtained data which shows combined effects of initial distribution and fluence attenuation in a RPV steel .
- Irradiation effects on Stainless steels
  - We conducted TEM and ATP analyses on irradiated stainless steels with various material and irradiation conditions and enhanced data base on microstructure of irradiated stainless steels.
  - We are developing the correlation model between microstructure and mechanical properties based on these findings.

Acknowledgment

- Following Research items of were conducted in the national project of “Improvement of Evaluation Methods of Irradiation Effects on Reactor Pressure Vessel and Core Internals” (2015 to 2018 JFY) sponsored by METI. The project was collaborated with LWRS project by ORNL.
  - Development of miniature CT master curve method (for Irradiated material)
  - Evaluation of Through-wall attenuation of irradiation embrittlement
  - Elucidation of irradiation effect on stainless steels and development of correlation model between microstructure and mechanical properties (except Chooz A material)

Supplemental slides

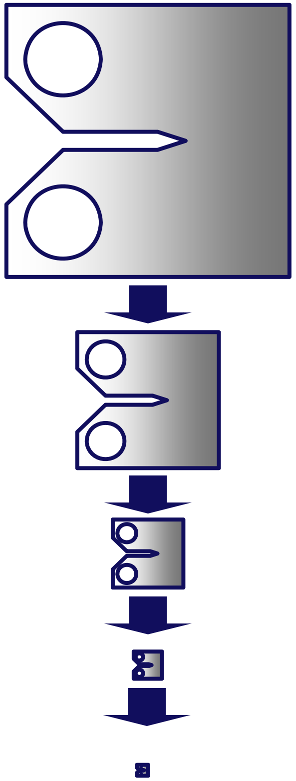


Step by step results

CONSISTENCY OF FRACTURE TOUGHNESS BETWEEN MINI-C(T) AND LARGER SPECIMENS

T_0 obtained using various size of specimens

- Appropriateness of use of Mini-C(T) specimens for the Master Curve evaluation was confirmed for 1 forging and 2 plate un-irradiated RPV base metal steels



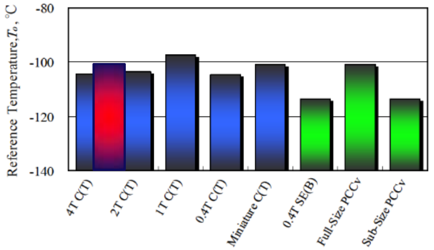
4T-C(T)

2T-C(T)

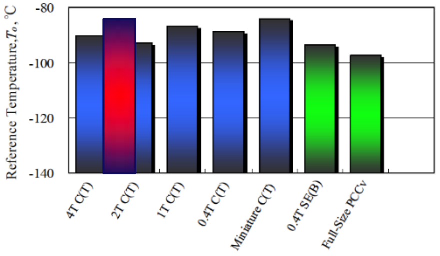
1T-C(T)

0.5T-C(T)

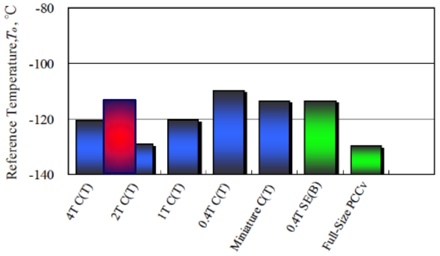
Mini-C(T)



(a) SFVQ1A forging



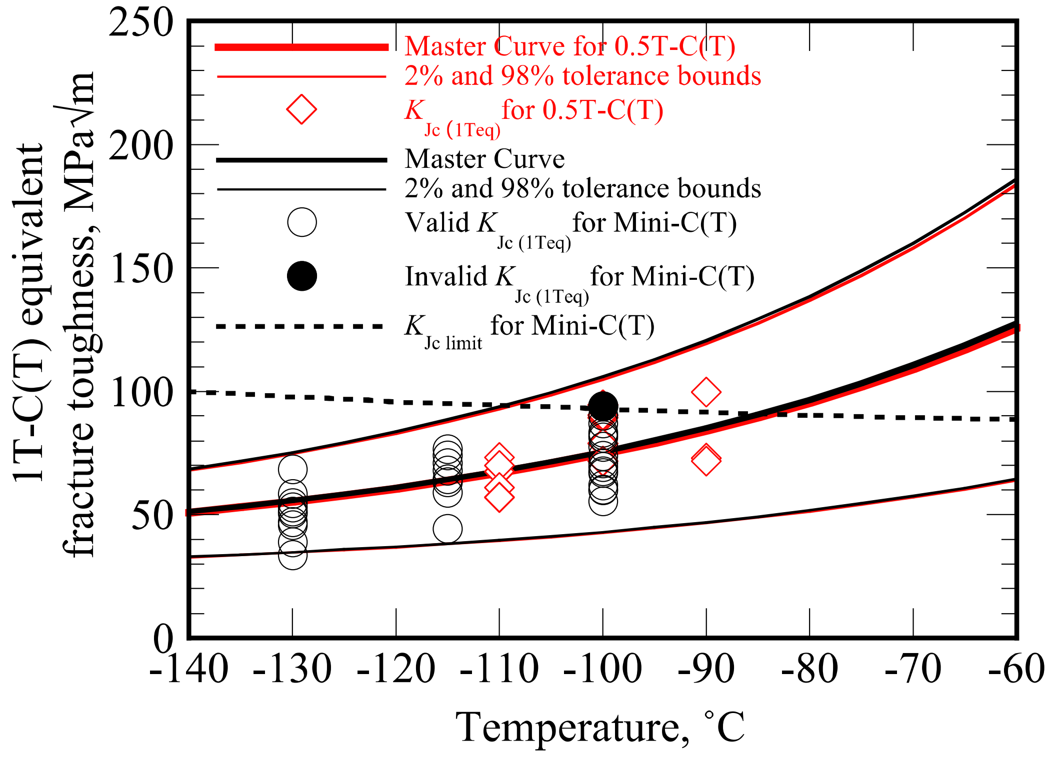
(b) SQV2A (Heat 1) plate



(c) SQV2A (Heat 2) plate

Miura et al., ASME Journal of PVT Vol.134-021402

Comparison of fracture toughness between 0.5T-C(T) and Mini-C(T) specimens of weld metal



T_o by 0.5T-C(T) : -77˚C

T_o by Mini-C(T) : -77˚C

Consistency of toughness between Mini-C(T) and larger specimens

Utilization in surveillance program

Round robin test for confirmation of reproducibility

Confirmatory study using irradiated material and / or actual RPV material

Standardization of test procedure

Step by step results

ROUND ROBIN TEST

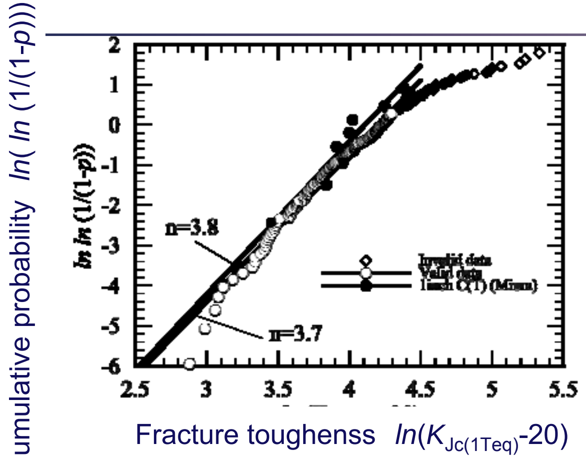
Mini-C(T) round robin test

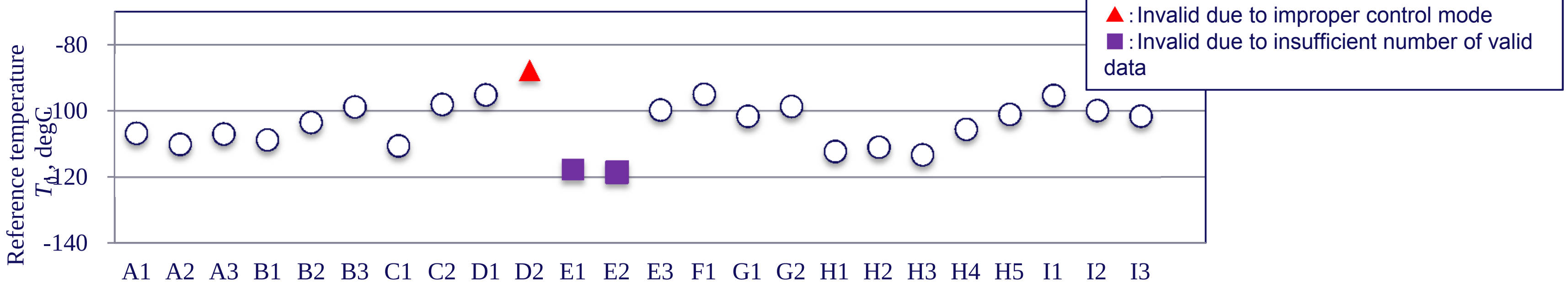
Purpose:

Ensure the reproducibility of T_o evaluation on Mini-C(T) specimens (RPV base metal unirradiated) machined and provided from CRIEPI

Participants:

Kyoto-Univ., JAEA, Toshiba/Hitachi GE/NFD, MHI/NDC, ORNL(USA), EPRI(USA), Westinghouse(USA), VTT(Finland), UJV(Czech), HZDR(Germany), SCK･CEN(Belgium), CRIEPI





- Equivalent To between 1T-C(T) and Mini-C(T) specimens.
- Consistent results among the participants.

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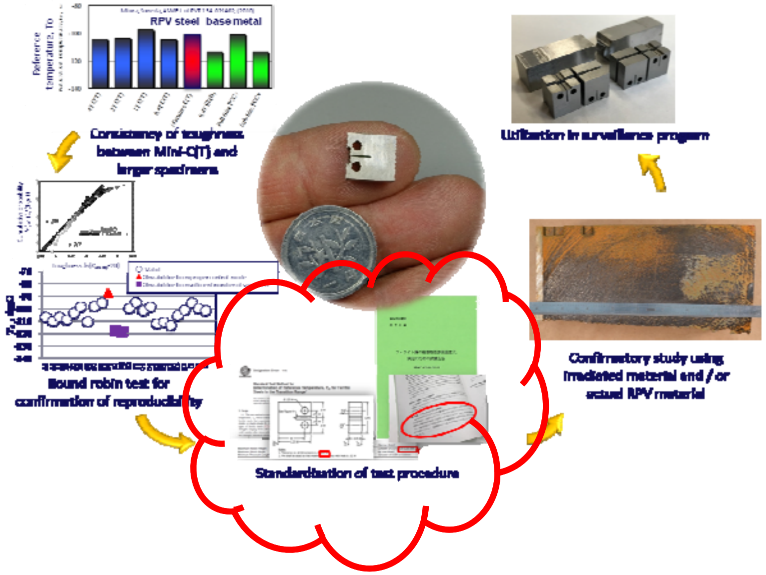

Consistency of toughness between Mini-C(T) and larger specimens

Utilization in surveillance program

Confirmatory study using irradiated material and / or actual RPV material

Standardization of test procedure

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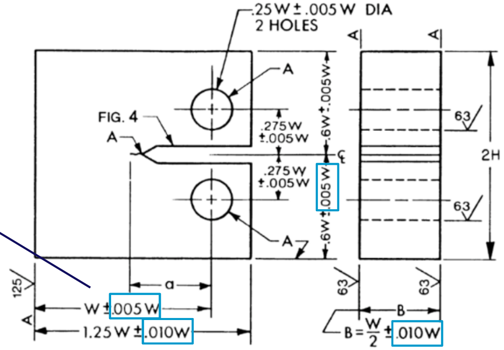
Step by step results

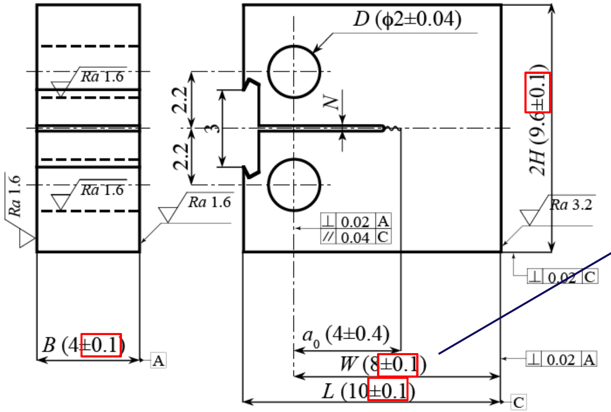
STANDARDIZATION OF TEST AND EVALUATION METHOD

Change in geometry and tolerance requirement

W ± 0.05W

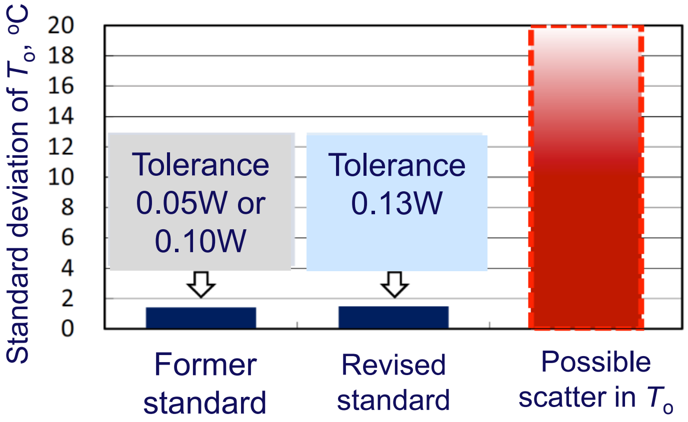


Tolerant requirement in former standard



W ± 0.1

Revised tolerant requirement
(0.1mm in Mini-C(T) = 0.13W)



- Change in notch height and tolerance requirements
- Notes and suggestions for usage of small specimens
- JEAC4216-2015
- ASTM E1921-2017a

M. Yamamoto, N. Miura et al., ASME PVP2015-45503

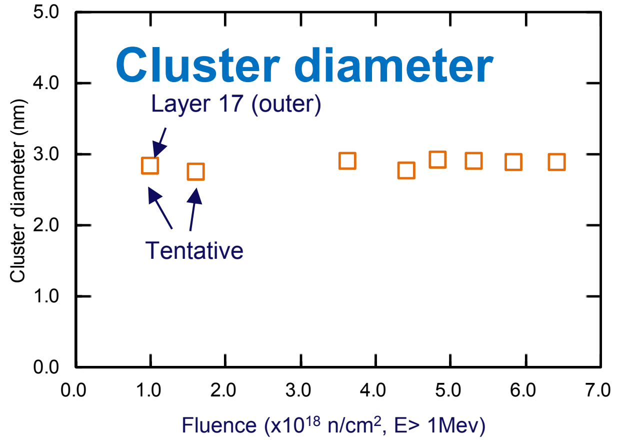
JRQ material

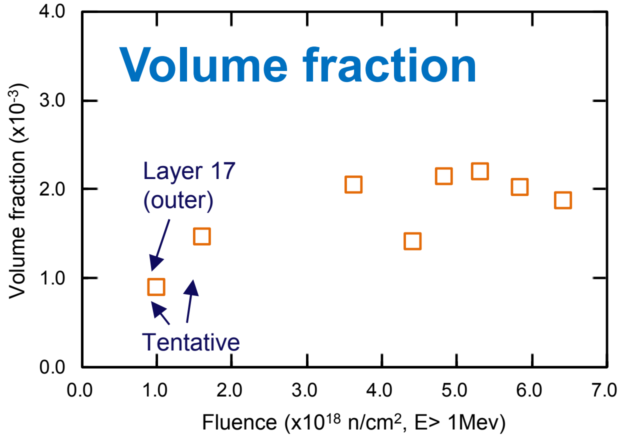
- Material : ASTM A533B Cl.1
- Cu : 0.14 wt.%
- Wall thickness : 225 mm (as fabricated), 190mm (for irradiation)
- Fluence range : 5.38 X 10^{19}(inside) to 0.31 X 10^{19} n/cm^2 (outside)(E>1 MeV)
- Flux : 7.07 X 10^{12} (inside) to 0.40 X 10^{12} n/cm^2・s(outside)(E>1 MeV)

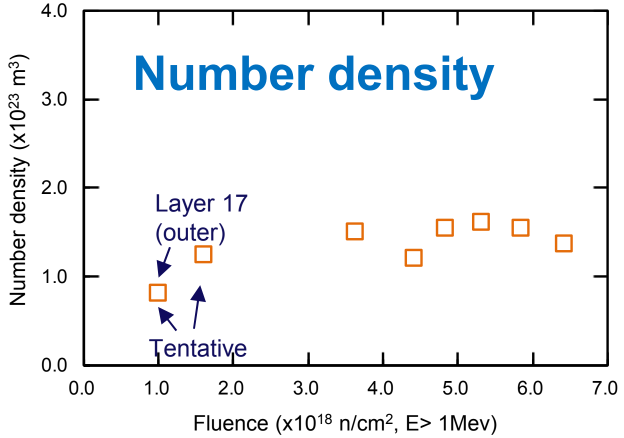
Zion base metal

- Material : ASTM A533B Cl.1
- Cu : 0.12 wt.%
- Wall thickness : 224 mm
- Fluence range : 0.64×10^{19}(inside) to 0.10×10^{19} n/cm^2 (outside)(E>1 MeV)
- Flux : 1.40×10^{10} (inside) to 0.22×10^{10} $n/cm^2 \cdot s$(outside)(E>1 MeV)

APT results of Zion materials







- ☐ Solute atom clusters formed in the inner to the outer layers of the Zion RPV.
- ☐ Cluster diameter is almost constant through the wall.
- ☐ Number density and volume fraction of cluster increased with increasing fluence.