IAEA Technical Meeting – Primary Radiation Damage
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Calculation of gamma displacement cross sections / Generation of recoil spectra from ENDF/B-VII

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Outline

- Introduction
  - General aspects of radiation damage
- Gamma displacement damage
- Recoil atom spectra
  - Neutron damage parameters
- Activities of damage simulation study
- Proposed work for CRP

Introduction

- Radiation damage in materials depends on many variables including,
  - Incident particle type, intensity and energy
  - Property of target materials
  - Environments (temperature, pressure...)
  - Irradiation time

- Damage to reactor structure by gamma-ray
  - Displacement damage by gamma, potentially significant depending on reactor designs
  - (Ex.) Displacement damage at 1/4 thickness of ABWR RPV

Damage source	Damage rate (dpa/s)
Gamma ray	1.0×10^{-13}
Neutron	2.0×10^{-13}

← D.E. Alexander, JNM 240 (1997) 196

Introduction

- Characteristics of neutron and gamma (KE = 1 MeV)

Characteristic	Gamma (γ)	Neutron (n)
Charge	neutral	neutral
Mass (amu)	-	1.008665
Velocity (cm/s)	c (= 2.998 x 10^{10})	1.38 x 10^{9}
Speed of light	100%	4.6%
Range in air (cm)	82,000	39,250

~ Gamma predominantly interacts with atomic electrons, while neutrons interact with nucleus.

Introduction

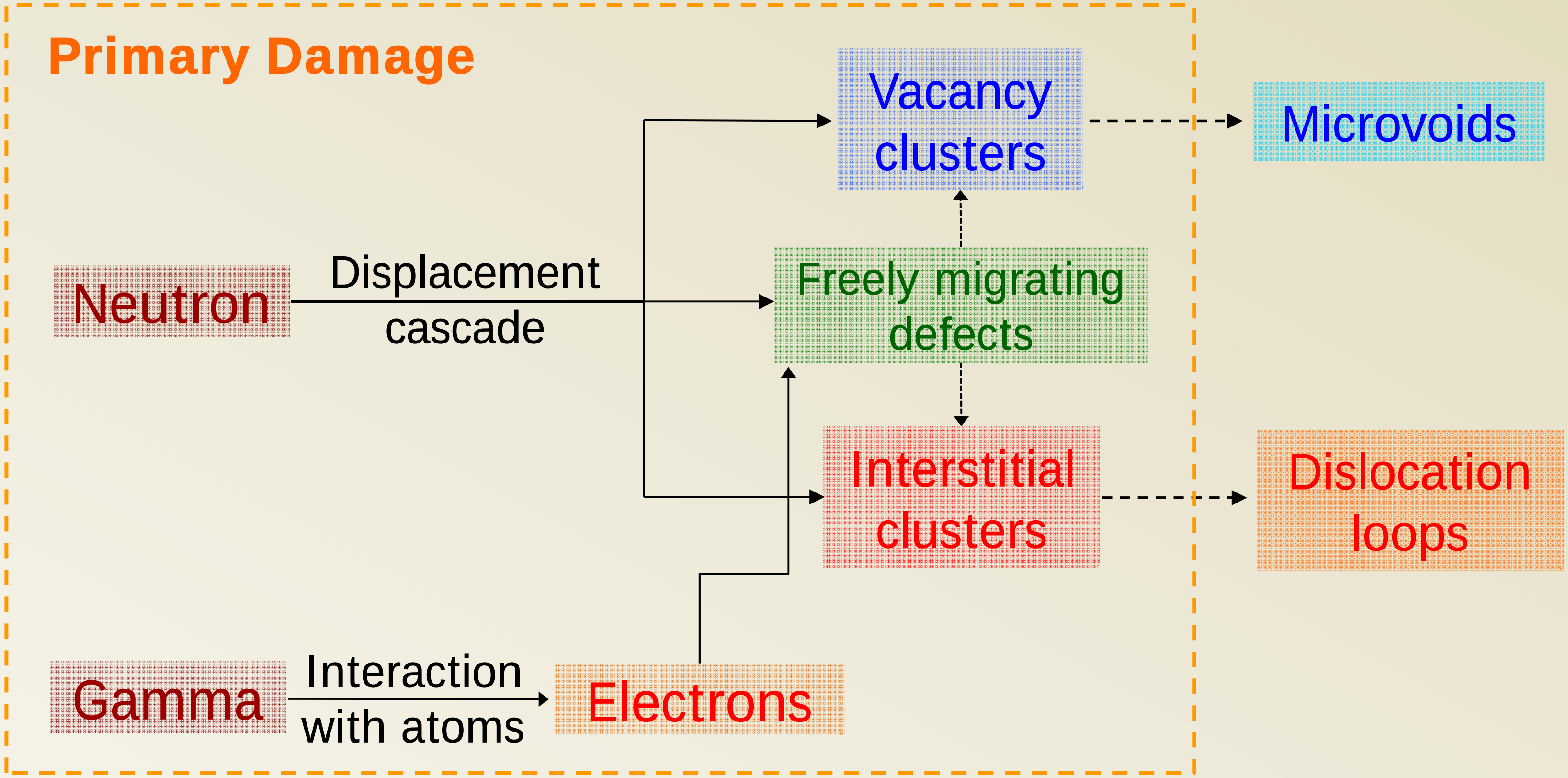
- Types of radiation effects on materials

Radiation / Effects	Neutron (n)	Gamma (γ)
Impurity production	Directly by absorption reactions	n/a
Ionization	Indirectly	Indirectly
Atomic displacement	Directly, multiple displacement via scattering reaction; cause displacement of recoil atoms	Rare displacement (via Compton effect)*

~ Interaction of gamma with atom → Production of energetic electrons → Collision reactions between electrons and lattice atoms may lead to atomic displacement

Introduction

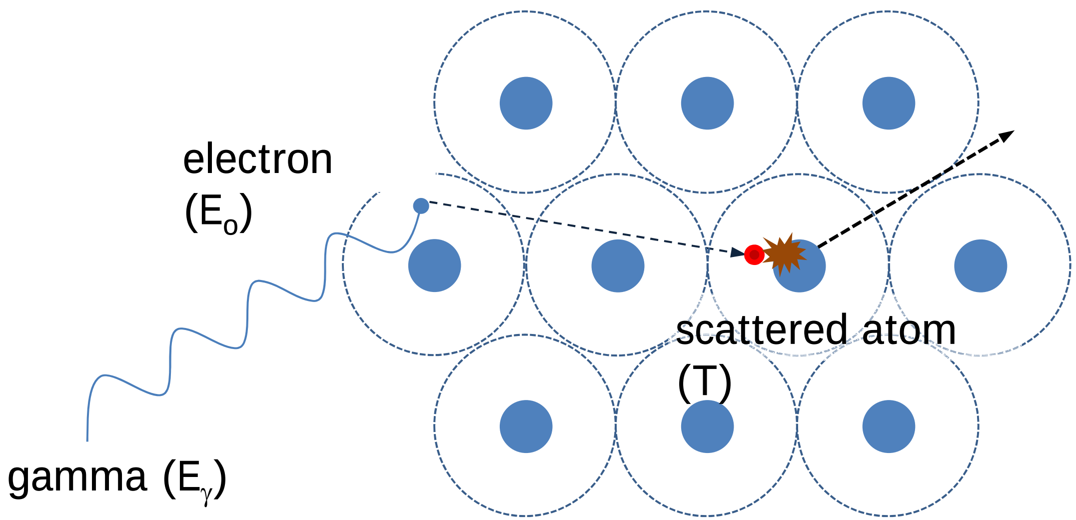
- Evolution of primary damage



- Lack of experimental information on gamma displacement damage
  → Calculation of gamma displacement cross sections
- Need to update neutron displacement damage parameters
  → Use of recent ENDF/B library

Gamma displacement damage

- Atomic displacement by gamma ray
  1. Production of energetic electrons by interaction with target atoms
  2. Displacement reaction by scattering collision between electrons and atoms

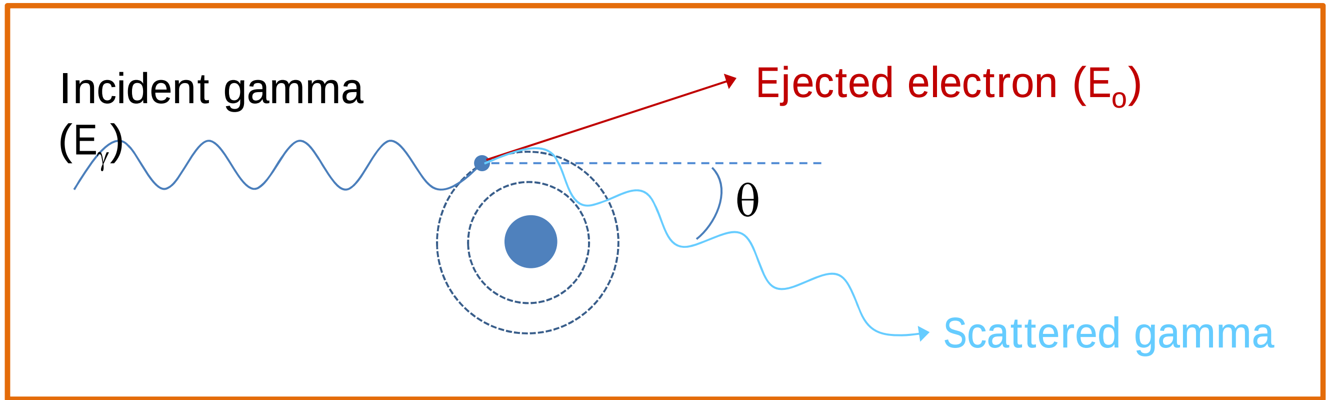


~Three major reactions to generate electrons
- Photoelectric effect (PE)
- Compton scattering (CS)
- Pair production (PP)

~ Scattered atom does not have enough energy (T) to induce displacement cascades.

Compton scattering displacement σ

- Compton scattering
  - Scattering between incident gamma ray and orbital electrons in material
  - Dominant at intermediate energy range (0.1 to 10 MeV)
  - Recoil electron energy depends on θ (scattering angle)



$$E_o = \frac{E_\gamma(1-\cos\theta)}{(E_e/E_\gamma)+(1-\cos\theta)}$$

E_e = electron rest mass energy

Compton scattering displacement σ

$$\sigma_\gamma^{CS}(E_\gamma) = \int_0^{E_o^{max}} \frac{d\sigma^c(E_\gamma, E_o)}{dE_o} \cdot \overline{n}(E_o)\, dE_o$$

$$\frac{d\sigma^c(E_\gamma, E_o)}{dE_o} = \frac{\pi e^4 Z}{E_e(E_\gamma - E_o)^2} \cdot \left\{ \left[\frac{E_e E_o}{E_\gamma^2}\right]^2 + 2\left[\frac{E_\gamma - E_o}{E_\gamma}\right]^2 + \frac{E_\gamma - E_o}{E_\gamma^3}\left[(E_o - E_e)^2 - E_e^2\right] \right\}$$

: differential scattering cross section for gamma interaction with electrons (Klein-Nishina formula)

$$\overline{n}(E_o) = N_a \int_0^{E_o} \frac{\sigma_d^e(E)}{(-dE/dx)} dE$$

: total number of displaced atoms over the range of recoil electron of E_o

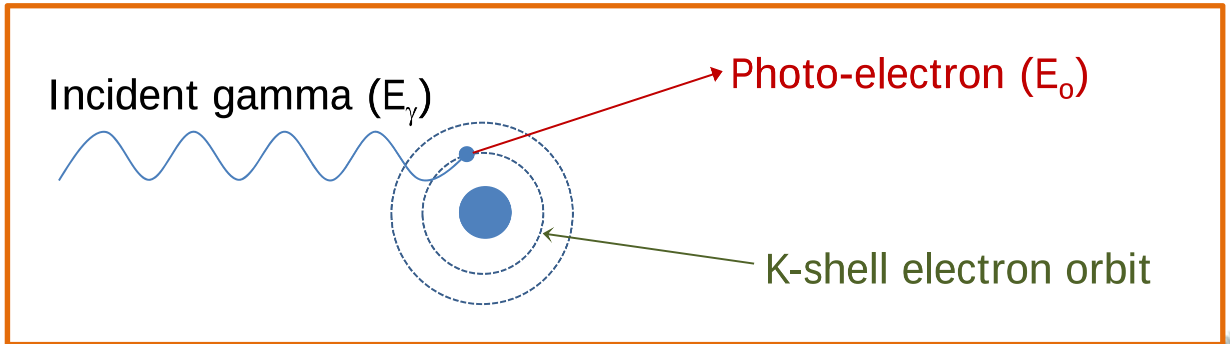
$\sigma_d^e(E)$: electron displacement cross sections from McKinley-Fesbach (Oen's table)

$(-dE/dx)$: electronic stopping power (eV/cm)

→ Numerical integration over $(0, E_o^{max})$ gives σ_γ^{CS}.

Photoelectric effect displacement σ

- Photoelectric effect
  - Gamma is absorbed by atom...photoelectron is emitted from one of its bound shells
  - Dominant at gamma energy below 0.1 MeV
  - Assume that all electrons have the same initial energy for E_γ resulting from K-shell ionization



$$E_o = E_\gamma - (BE)$$

BE = binding energy of electron to atom (~7 keV for Fe)

Photoelectric effect displacement σ

$$\sigma_{\gamma}^{PE}(E_{\gamma}) = \sigma^{PE}(E_{\gamma},E_{o}) \cdot \overline{n}(E_{o})$$

$\sigma^{PE}(E_{\gamma},E_{o})$: photoelectric effect cross section

(a) E_{γ} < 0.35 MeV (Sauter's Eq.)

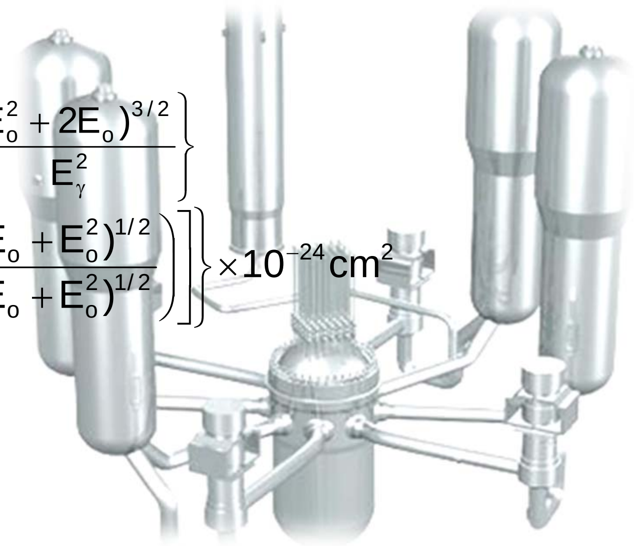
$$\sigma^{PE}(E_{\gamma},E_{0}) = \frac{3}{2}\,\varphi_{o}\,\frac{Z^{5}}{137^{4}}\,\xi^{5}\,(\Lambda^{2}-1)^{3/2}\left[\frac{4}{3}+\frac{\Lambda(\Lambda-2)}{\Lambda+1}\left(1-\frac{1}{2\Lambda(\Lambda^{2}-1)^{1/2}}\cdot\ln\frac{\Lambda+(\Lambda^{2}-1)^{1/2}}{\Lambda-(\Lambda^{2}-1)^{1/2}}\right)\right]$$

$$\Lambda = \frac{E_{\gamma}-BE-E_{e}}{E_{e}},\quad \xi = Ee/Eg,\quad \varphi_{o} = \text{Thompson cross section (0.6653 b)}$$

(b) E_{γ} > 2 MeV (Hall's Eq.)

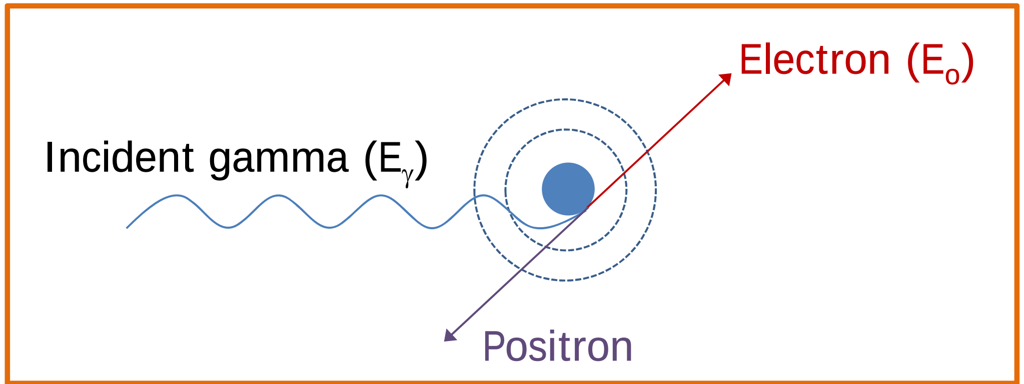
$$\sigma^{PE}(E_{\gamma},E_{o}) = \frac{5}{4}\frac{Z^{5}}{137^{4}}\frac{1}{E_{\gamma}}\exp\left\{-\frac{\pi Z}{137}+2\left(\frac{Z}{137}\right)^{2}\left(1-\ln\frac{Z}{137}\right)\right\}\left\{\frac{(E_{o}^{2}+2E_{o})^{3/2}}{E_{\gamma}^{2}}\right\}$$
$$\left\{\frac{4}{3}+\frac{E_{o}^{2}-1}{E_{o}+2}\left[1-\frac{1}{2(E_{o}+1)(E_{o}^{2}+2E_{o})^{1/2}}\cdot\ln\left(\frac{E_{o}+1+(2E_{o}+E_{o}^{2})^{1/2}}{E_{o}+1-(2E_{o}+E_{o}^{2})^{1/2}}\right)\right]\right\}\times 10^{-24}\,cm^{2}$$

(c) 0.35 < E_{γ} < 2 MeV ~ extrapolation

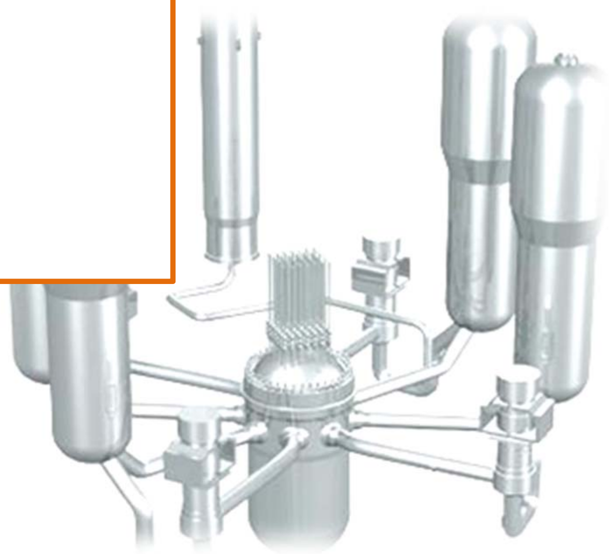


Pair production displacement σ

- Pair production
  - Gamma is completely absorbed in the field of nucleus... electron-positron pair is created
  - E_γ > 1.02 MeV (= $2 \cdot E_e$)
  - Effect of positron on atomic displacement is not included because of its annihilation with electrons (e^+-e^-)



$$E_o = \frac{1}{2}\left(E_\gamma - 2 \cdot E_e\right)$$



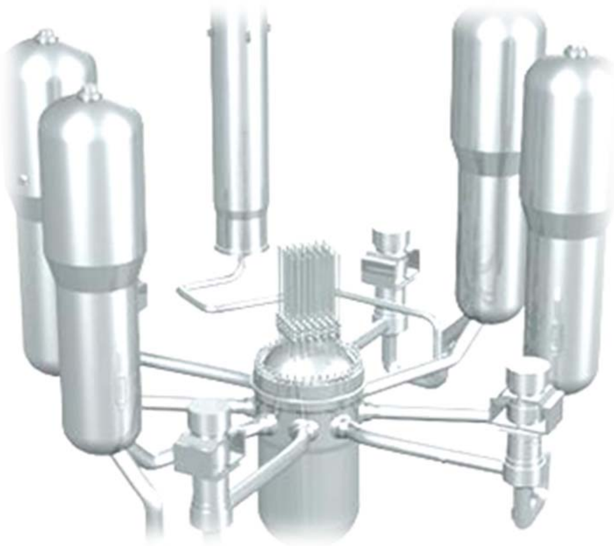
Pair production displacement σ

$$\sigma_{\gamma}^{PP}(E_{\gamma}) = \sigma^{PP}(E_{\gamma},E_{o}) \cdot \overline{n}(E_{o})$$

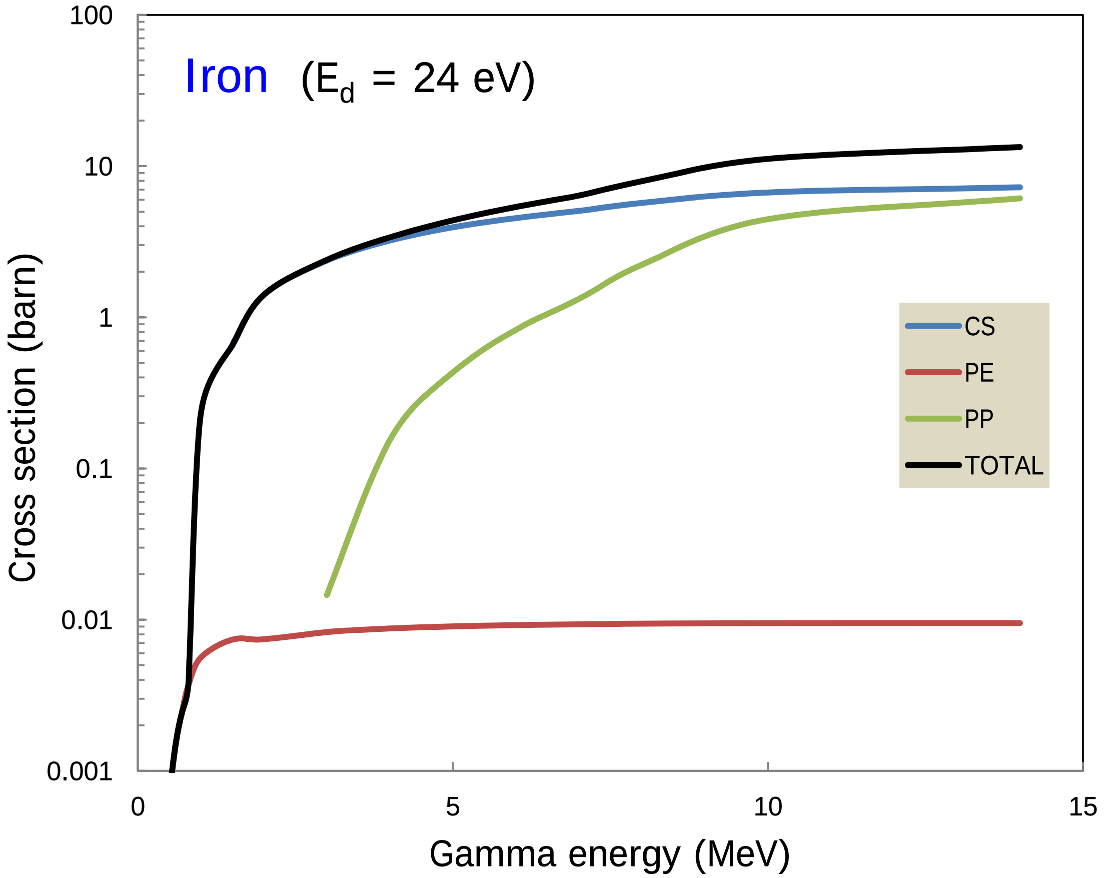
$\sigma^{PP}(E_{\gamma},E_{o})$: pair production cross section

$$\sigma^{PP}(E_{\gamma},E_{o}) = \sigma_{co} \cdot Z^{2}\left\{\frac{28}{9}\ \ln\left(\frac{2E_{\gamma}}{E_{e}}\right) - \frac{218}{27}\right\}$$

$\sigma_{co} = 2.8 \times 10^{-4}$ barn

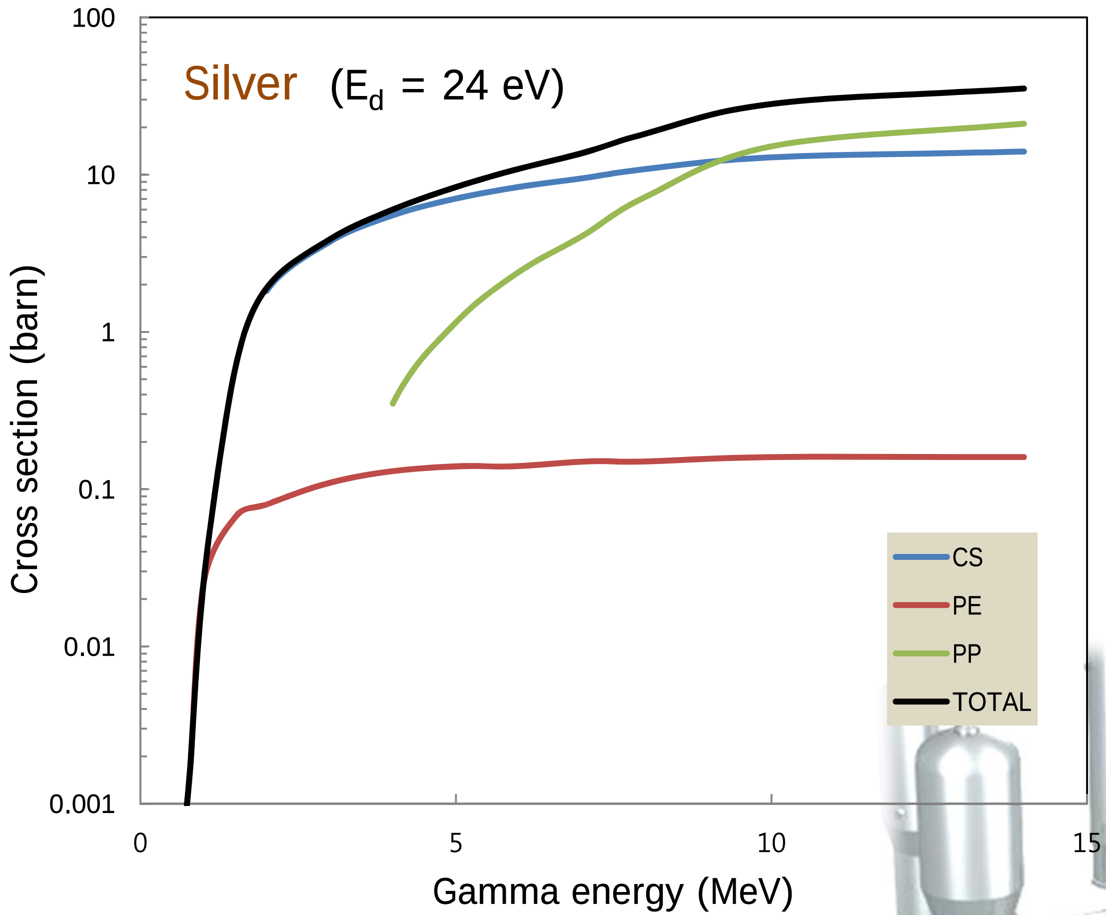


Total gamma displacement cross section



- Displacement due to Compton scattering is dominant for elements (Z < 30)
- Effect of pair production on displacement becomes significant at higher gamma energy.

Total gamma displacement cross section



- Displacement due to pair production is increasingly dominant at higher gamma energy for heavier elements

Neutron displacement damage

- Damage starts from PKA (Primary Knock-on Atom) production
  - Interaction of neutron with lattice atom
  - → Transfer of neutron energy to lattice atom
  - → Displacement of struck atom (PKA) from lattice sites
  - → Create additional displacements (displacement cascade)
  - → Production of point defects (interstitials and vacancies)

- Damage parameters - dpa
  - displacements per atom: calculated number of recoil atoms that are displaced from their lattice sites as a result of neutron bombardment
  - total initial energy available to produce damage
    ~ a measure of the maximum damage possible
  - lack of information on net damage

Neutron displacement damage

- Calculation of dpa

$$\text{dpa rate (dpa/s)} = \sum \phi(E) \cdot \sigma_d(E)$$

$$\sigma_d(E) = \sum_i \sigma_i(E) \int_{T_{min}}^{T_{max}} f_i(E,T) \cdot \nu_{NRT}(T)\ dT$$

where, $\phi(E)$ = neutron spectra of energy E

σ_d = neutron displacement cross section

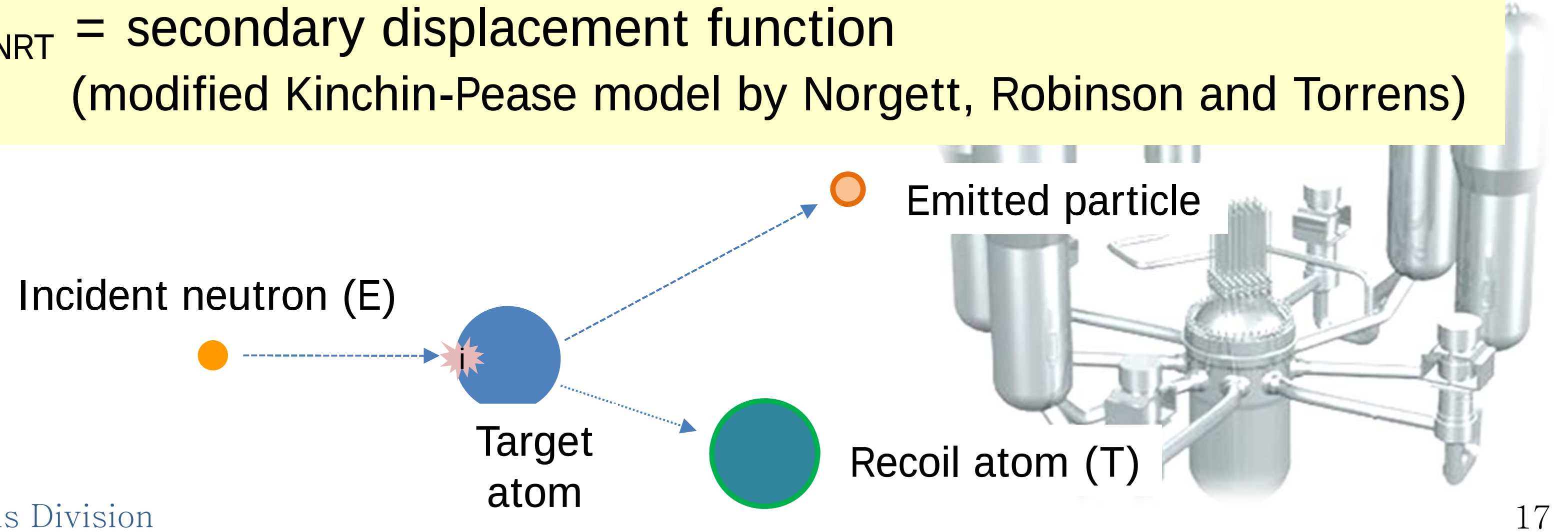
σ_i = nuclear cross section for channel i (reaction type)

$f_i(E,T)$ = neutron-atom energy transfer kernel for channel i

T = recoil atom energy

ν_{NRT} = secondary displacement function

(modified Kinchin-Pease model by Norgett, Robinson and Torrens)



Displacement cross section by neutrons

- Determining factor for affecting neutron damage

$$\sigma_d(E) = \sum_i \sigma_i(E) \int_{T_{min}}^{T_{max}} f_i(E,T) \cdot \nu_{NRT}(T)\ dT$$

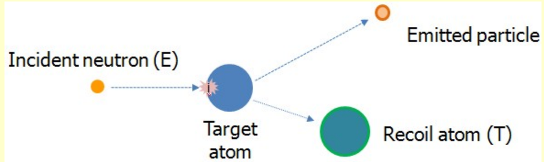
~ Recoil atom energy (T) is a basic estimator for evaluation
~ Number of surviving defects (residual defects) is strongly dependent on recoil energy, T

- Estimation of recoil atom spectra
  - Use of SPECTER computer code
    ~ Convenient code to calculate damage parameters due to neutron irradiation [dpa, σ_d, recoil spectra, gas production]
    ~ Old ENDF library (ENDF/B-V)
  - Derive recoil atom spectra R(T) using latest ENDF library

Recoil atom spectra

- Definition
  ~ Probability that a recoil atom has its kinetic energy (T, T+dT) for a given neutron spectrum ϕ(E)

$$R(T)dT = \sum_i \int_0^{E_{\max}} \phi(E) \cdot \sigma_i(E) \cdot f_i(E,T)\, dE\, dT$$



- Variables for R(T) calculation
  1. Neutron spectra, ϕ(E)
     ~ depends on neutronic environments (reactor type, operation condition etc.)
     ~ output from commercial codes (MCNP, DORT...)
  2. Microscopic cross section, $\sigma_i(E)$
     ~ Available from *ENDF/B library* | Need data processing
  3. Neutron-atom energy transfer kernel, $f_i(E,T)$
     ~ Available from *ENDF/B library* | Need data processing & mathematical derivation

ENDF/B library format

- Target material [mat]

  Ex) Fe-26-55 → mat=2628 (Iron, Z=26, A=55)

- Data type [mf]

  - mf=3 Microscopic reaction cross section data
  - mf=4 Angular distributions for an emitted particle (n)
  - mf=5 Energy distributions for an emitted particle (n)
  - mf=6 Energy-angle distributions for all emitted particles (recoils, n...)

- Reaction type [mt]

  - mt=1 (n,total)
  - mt=2 (n,n) → Elastic scattering
  - mt=3 (n,n') → Inelastic scattering
  - mt=51 (n,n') → Inelastic scattering with 1st excited state of target atom

Ex) mat=2628, mf=3, mt=2 → Microscopic cross section for the elastic scattering reaction between a neutron and Fe-26-55 atom

Energy transfer kernel

- Energy transfer kernel for recoil atoms is NOT usually available directly from ENDF/B library
  ~ Most data in ENDF are related to neutron's behavior
  ~ Require a conversion from neutron reaction data to recoil data to obtain $f_i(E,T)$; $R(T)$

- Data [$f_i(E,T)$] available from ENDF/B
  ~ Angular distribution of emitted neutron (mf=4)
  ~ Energy distribution of emitted neutron (mf=5)
  ~ Energy-angle distribution of all emitted particles (mf=6)

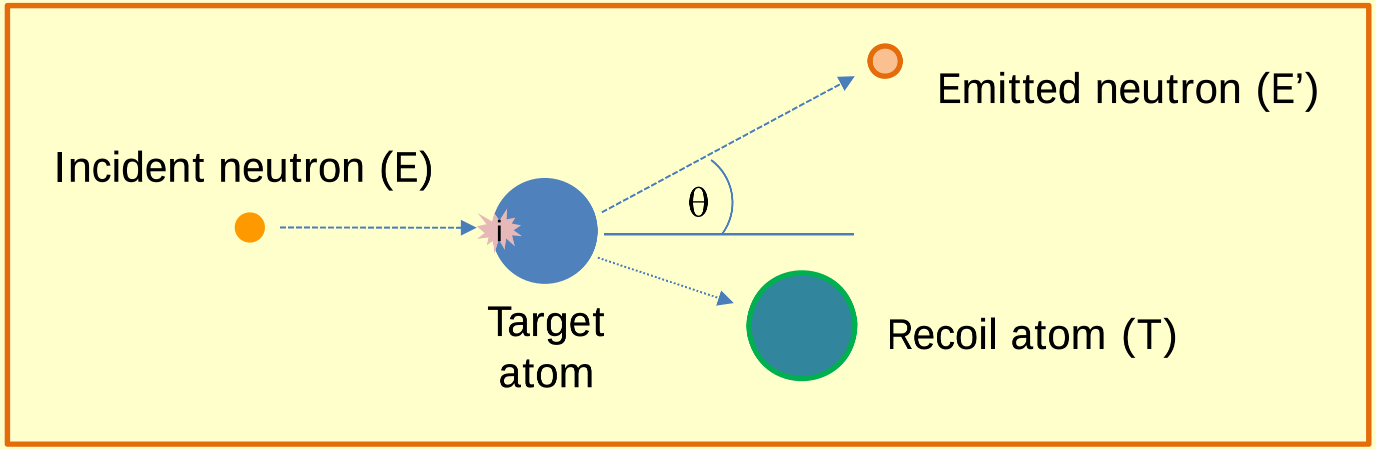
  ~ Special treatment for radiative capture (n,γ)
  ~ Special treatment in the absence of energy transfer kernel

Energy transfer kernel (mf=4)

- Angular distribution of emitted neutron (mf=4)
  - ~ Elastic scattering and inelastic scattering with discrete γ
  - ~ Probability that a neutron of energy E will be scattered into the interval $(\mu, \mu+d\mu)$ is given by:

$$f(E,\mu)\, d\mu$$

where, μ is the cosine of the scattering angle (θ)



Energy transfer kernel (mf=4)

- Energy transfer kernel f(E,T) for mf=4
  ~ Conversion of f(E,μ) into f(E,T)

$$f(E,\mu)\,d\mu = f(E,T)\,dT \rightarrow f(E,T) = f(E,\mu)\left|\frac{d\mu}{dT}\right|$$

  ~ Use of momentum and energy conservation law in 2-body collision → Derive relationship of E, T and μ

– **In Center of Mass system (CMS)**

$$T = \mu_1\mu_4 E + \mu_3(\mu_2 E - Q) - 2\sqrt{\mu_1\mu_3\mu_4 E(\mu_2 E - Q)}\,\mu_c$$

– **In Laboratory system (LS)**

$$T = \mu_1 E + \mu_3 E - \mu_3 Q - 2\sqrt{\mu_1\mu_3 E E'}\,\mu_l$$

where $\mu_1 = 1/(1+A)$, $\mu_2 = A/(1+A)$, $\mu_3 = 1/(1+A')$, $\mu_4 = A'/(1+A')$ and μ_c is the scattering cosine in CMS, μ_l is the scattering cosine in LS.

→ $\left|\frac{d\mu}{dT}\right|$

where, A & A' = mass of target atoms before & after collision, respectively
Q = de-excitation energy (for elastic scattering Q=0)

Energy transfer kernel (mf=4)

- Energy transfer kernel f(E,T) for mf=4
  ~ In ENDF/B library, angular distribution of scattered neutron f(E,μ) is given in terms of Legendre polynomial series,

$$f(E,\mu) = \sum_{\ell=0}^{L} \frac{2\ell+1}{2} a_\ell(E) P_\ell(\mu)$$

where, P_ℓ = ℓ^{th} Legendre polynomial & a_ℓ = its coefficient (ENDF/B)

→ Plugging f(E,μ) & |dμ/dT| gives f(E,T) for mf=4

– **In CMS**

$$f(E,T) = \sum_{l=0}^{L} \frac{2l+1}{4\sqrt{\mu_1\mu_3\mu_4 E(\mu_2 E - Q)}} a_l(E) P_l\left(\frac{\mu_1\mu_4 E + \mu_3(\mu_2 E - Q) - T}{2\sqrt{\mu_1\mu_3\mu_4 E(\mu_2 E - Q)}}\right)$$

– **In LS**

$$f(E,T) = \sum_{l=0}^{L} \frac{2l+1}{2} \frac{2\sqrt{\mu_1\mu_3 E E'}}{\sqrt{\mu_1\mu_3}\sqrt{\frac{E}{E'}} - 1} a_l(E) P_l\left(\frac{\mu_1 E + \mu_3 E - \mu_3 Q - T}{2\sqrt{\mu_1\mu_3 E E'}}\right)$$

Energy transfer kernel (mf=6)

- Energy-angle distribution of all emitted particles (mf=6)
  - ~ Inelastic scattering with continuous γ and (n,p) & (n,α) reactions
  - ~ Energy-angular distribution of emitted particles is given by normalized probability distribution function such as:

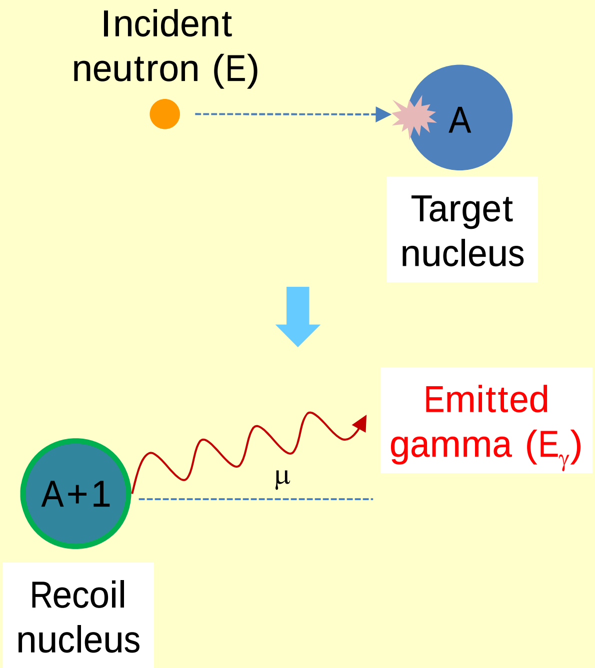
$$f(E,\mu,E')d\mu dE' \text{ or } f(E,\mu,T)d\mu dT$$

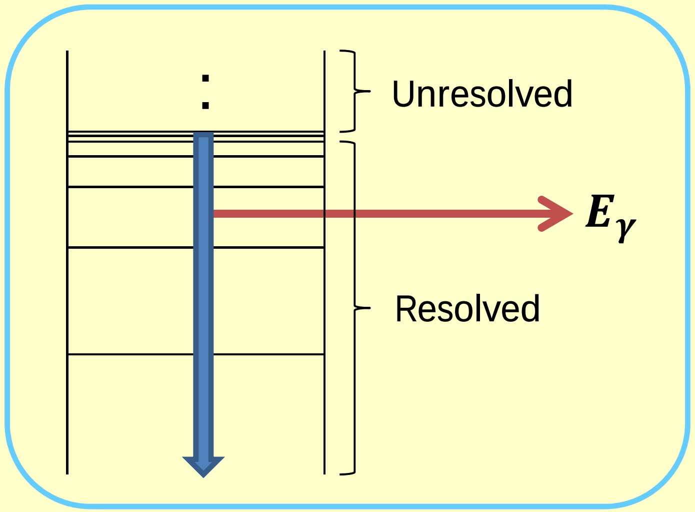
  - ~ Energy transfer kernel f(E,T) is readily obtained

$$f(E,T)dT = \int_{-1}^{1} f(E,\mu,T)\,d\mu dT$$

Energy transfer kernel (radiative capture)

- Special treatment for (n,γ)
  - ~ Recoil energy (T) is determined by emitted gamma energy (E_γ)
  - ~ Consider two different cases:
    1) Resolved E_γ
    2) Unresolved E_γ





Energy level of compound nucleus (A+1)

Energy transfer kernel (radiative capture)

- Radiative capture with resolved E_γ
  - ~ Possible to derive relationship among E, T, E_γ and μ through energy and momentum conservation laws

$$T = \frac{E_\gamma^2}{2(A+1)m_o c^2} + \frac{E}{A+1} + \frac{E_\gamma}{(A+1)m_o c}\sqrt{2E}\,\mu \qquad m_o = 1 \text{ amu}$$

  - ~ T and μ has one-to-one correspondence since E_γ is resolved and $f(E,\mu)$ is available from ENDF/B library

$$f(E,T;E_\gamma)\,dT = f(E,\mu)\,d\mu$$

$$f(E,T;E_\gamma) = f(E,\mu)\frac{(A+1)m_o c}{E_\gamma\sqrt{2E}}$$

Energy transfer kernel (radiative capture)

- Radiative capture with unresolved E_γ
  - ~ Continuous energy distribution of secondary photon is available from ENDF/B library in the form of normalized probability such as $f(E,E_\gamma)\ dE_\gamma$
  - ~ Assume isotropic emission of secondary photons

$$f(E,T)dT = \int_0^\infty f(E,E_\gamma) \times \frac{1}{T_{max}(E_\gamma) - T_{min}(E_\gamma)} dE_\gamma dT$$

- $f(E,E_\gamma)$: Probability of γ-ray emission at E_γ (ENDF/B)
- $\frac{1}{T_{max}(E_\gamma) - T_{min}(E_\gamma)}$: Probability that this emission transfers energy T to target nucleus (isotropic emission)

$$f(E,T)\, dT = \frac{(A+1)m_o c^2}{2\sqrt{2m_o c^2 E}} \times \int_0^\infty \frac{f(E,E_\gamma)}{E_\gamma} dE_\gamma\ dT$$

Energy transfer kernel (charged particle emission)

- No information on f(E,T) in ENDF/B
  - ~ Nuclear reactions involved with charged particle emission except (n,p) and (n,α)
  - ~ Use of theoretical models for deriving f(E,T)
    1) Evaporation model
    2) Two-body kinematics
  - ~ Recoil energy (T) is determined through energy and momentum conservation laws in LS such as:

$$T = \frac{1}{A+1+a}\left(aE_a - 2\sqrt{aE_aE}\mu_\ell + E\right)$$

where E_a = KE of emitted particle, a = mass ratio of emitted particle to neutron, μ_ℓ = scattering angle in LS ($-1 < \mu_\ell < 1$)

  - ~ Assume isotropic emission of recoil atoms, f(E,T) is:

$$f(E,T) = \frac{1}{T_{max}(E) - T_{min}(E)}$$

Energy transfer kernel

- Energy transfer kernel for Fe
  ~ Following information is included in ENDF/B-VII

Reaction type	mt	Energy transfer kernal	mf
(n,n)	2	derive from f(E,μ)	4
(n,2n)	16	direct f(E,T)	6
(n,n') with discrete γ	51-75	derive from f(E,μ)	4
(n,n') with continuous γ	91	direct f(E,T)	6
(n,γ)	102	derive from f(E,μ) & f(E,E_γ) with y(E)	12
(n,p)	103	direct f(E,T)	6
(n,d)	104	derive f(E,T) from model	-
(n,t)	105	derive f(E,T) from model	-
(n,^{3}He)	106	derive f(E,T) from model	-
(n,α)	107	direct f(E,T)	6

Recoil atom spectrum generator (RASG)

- Summary of RASG code (underway)

Module 1	Module 2	Module 3
• Generation of microscopic nuclear cross sections from ENDF/B-VII • Use of NJOY code (Nuclear Data Processing Code)	• Calculation of energy transfer kernel for each nuclear reaction	• Calculation of neutron flux • Use of neutron transport code (MCNP...)

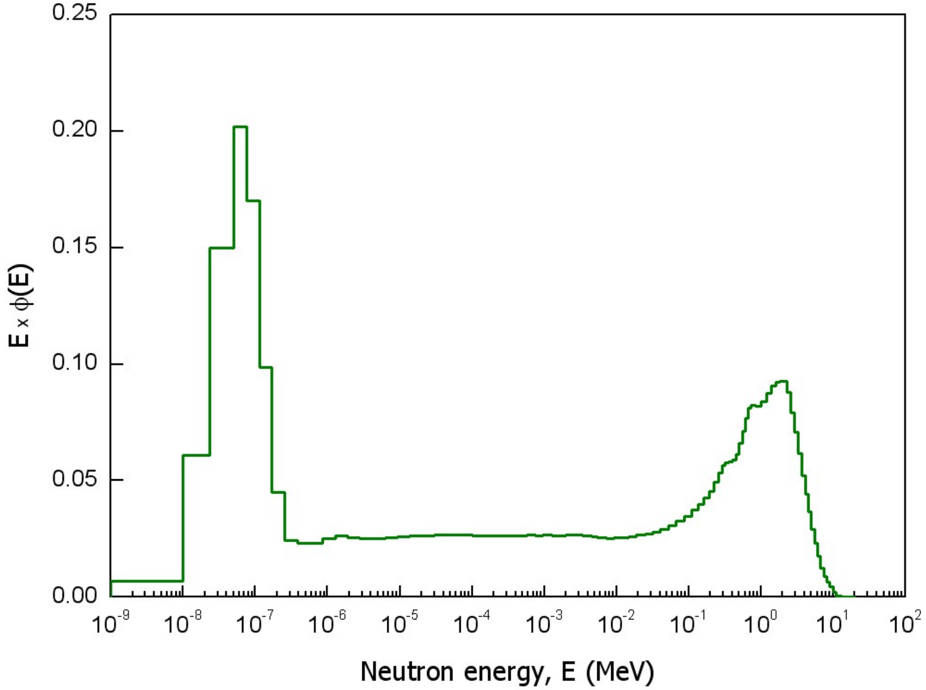
$$R(T)dT = \sum_i \int_0^{E_{max}} \phi(E) \cdot \sigma_i(E) \cdot f_i(E,T)dE\, dT$$

RASG code

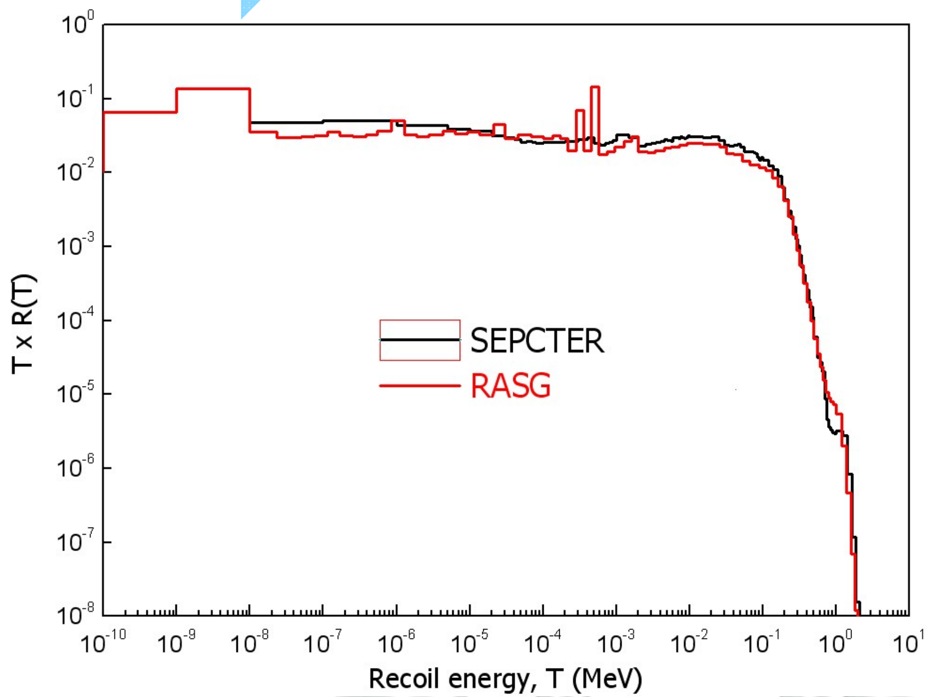
→ Recoil atom spectra

Recoil atom spectrum (RASG vs. SPECTER)

φ(E) → SPECTER / RASG → R(T)

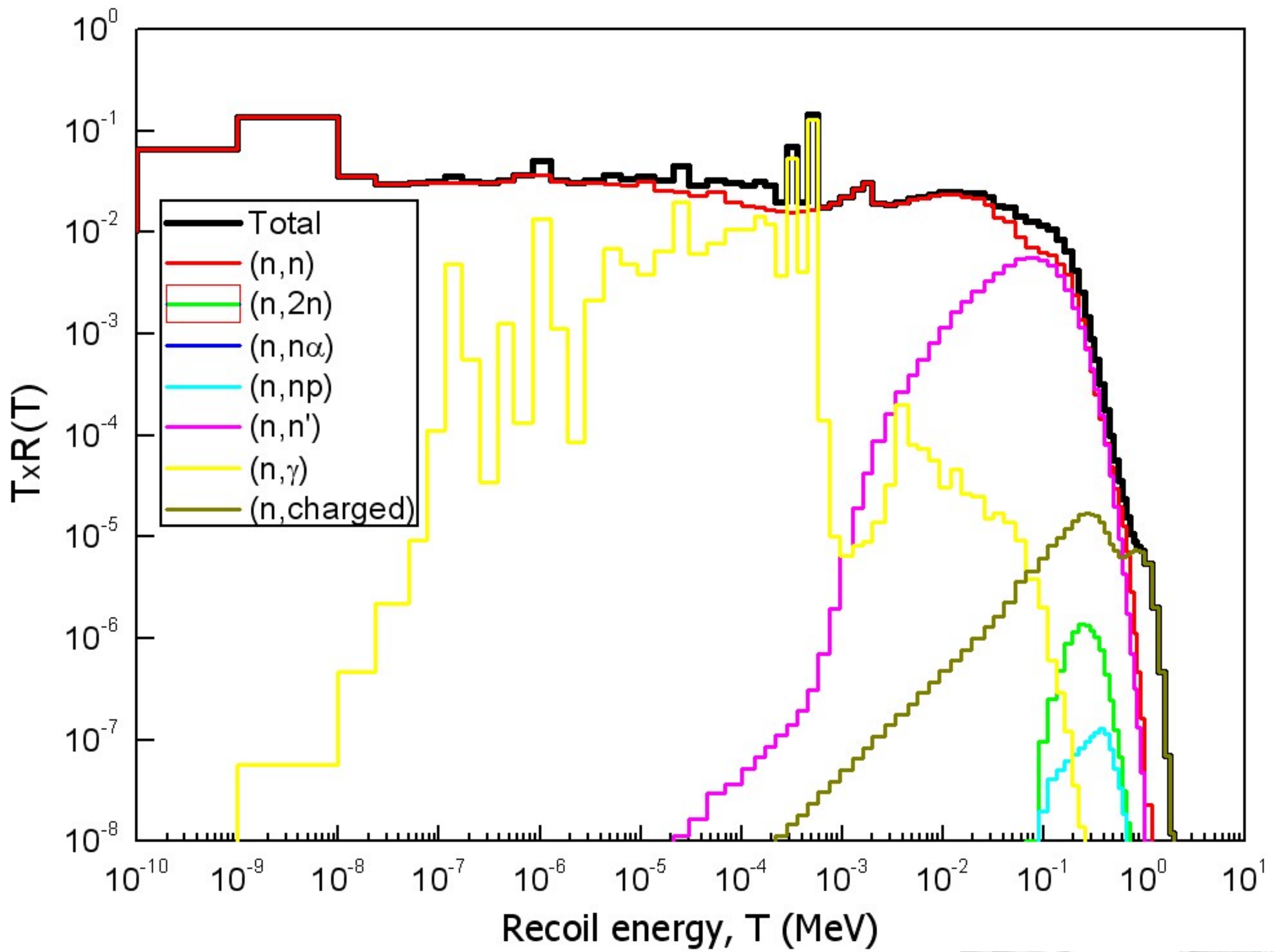


HFIR neutron spectrum
(from SPECTER)



Iron recoil atom spectra

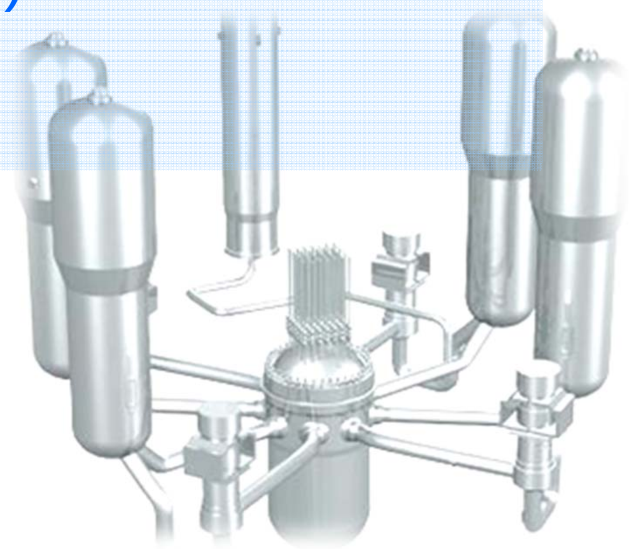
Recoil atom spectrum



Energy distribution of Fe-recoils in HFIR from RASG
(contribution of each reaction type to total spectrum)

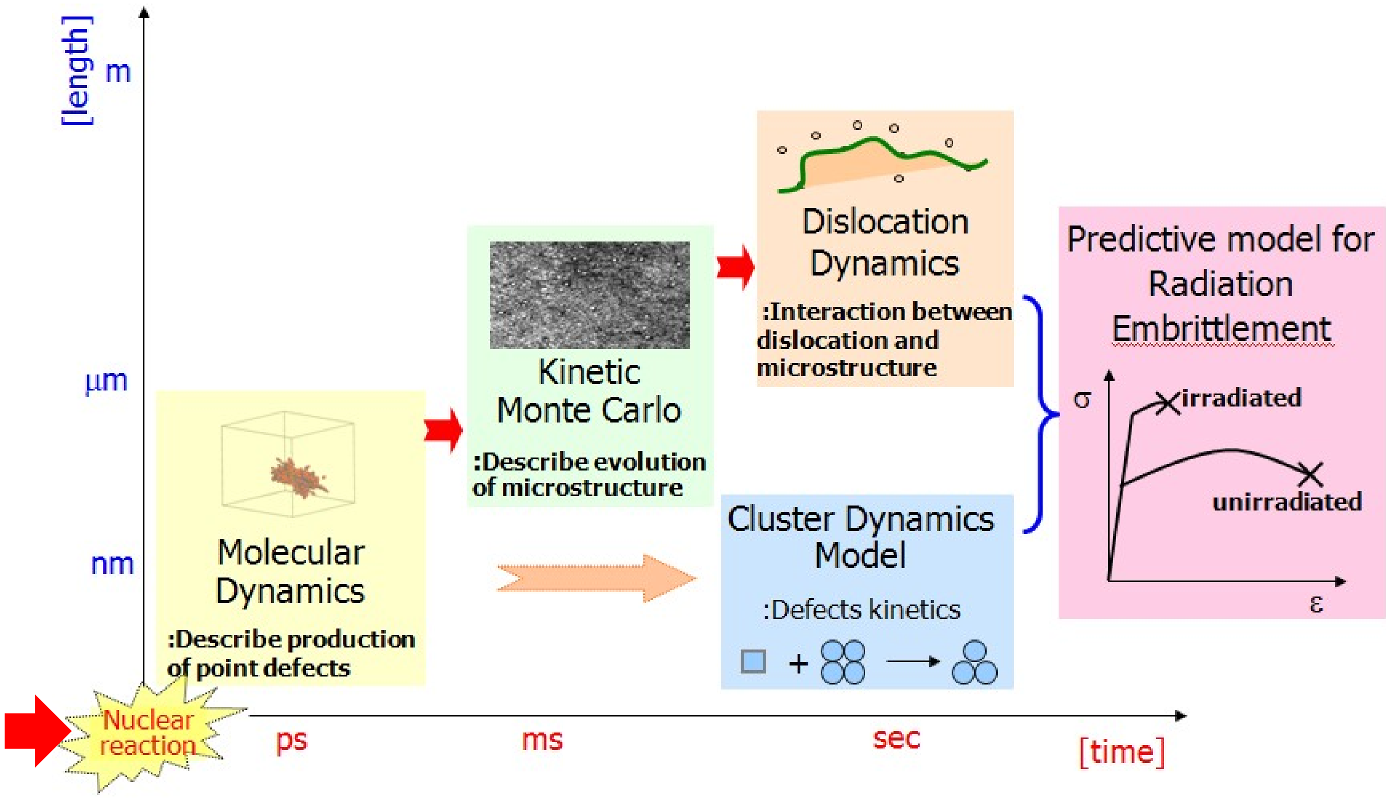
Summary

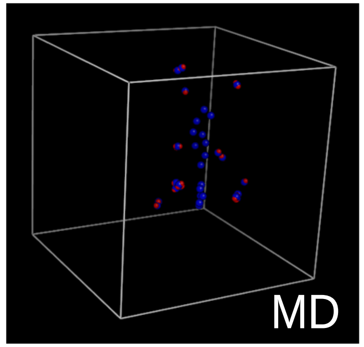
- Calculation of gamma displacement cross section
  - ~ Account for 3 major interactions including PE, CS and PP
  - ~ Not significant but efficient at producing freely-migrating defects
- Generation of recoil atom spectra using ENDF/B library
  - ~ Develop code by combining three modules
    - - Neutron flux, $\phi(E)$
    - - Microscopic nuclear cross section, $\sigma_i(E)$
    - - Energy transfer kernel, $f_i(E,T)$
  - ~ Use latest ENDF for calculation

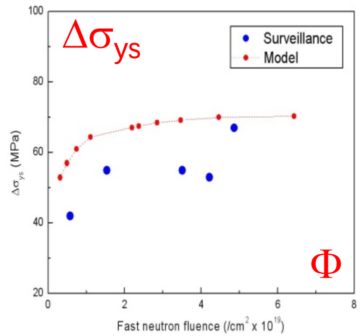


Activities of damage simulation (KAERI)

Multiscale Modeling for Radiation Effects







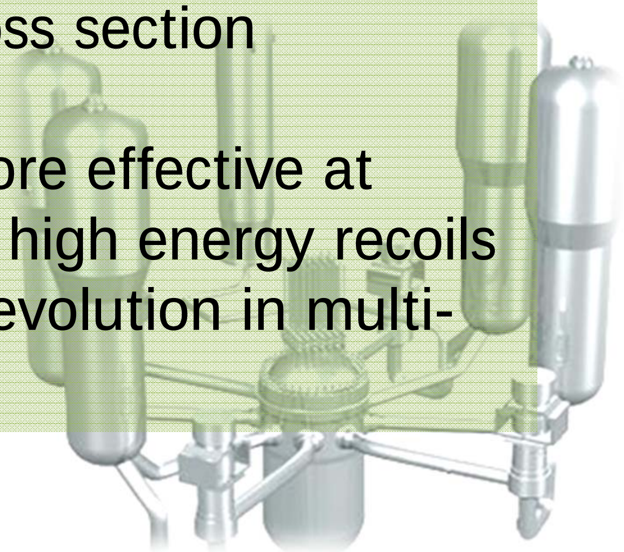
Korean NPP
RPV Steels (YG2)

- Developed integrated models for irradiation embrittlement of RPV steels
- Developing multiscale models for estimating radiation damage to SS (hardening, segregation, swelling etc.)

Proposed Work

- Pros and Cons of *dpa* parameter
  - ~ Successful as correlation parameter which is linked to materials property
  - ~ Universal parameter applicable to any types of radiation
  - - Indication of total energy available to induce damage to matrix (not permanent damage)
  - - Depending on used model for secondary displacement

- Development of modified displacement cross section
  - ~ close to real/net damage
  - ~ known fact: lower energy recoils are more effective at producing freely-migrating defects than high energy recoils
  - ~ Useful for prediction of microstructural evolution in multi-scale radiation damage study



Proposed Work

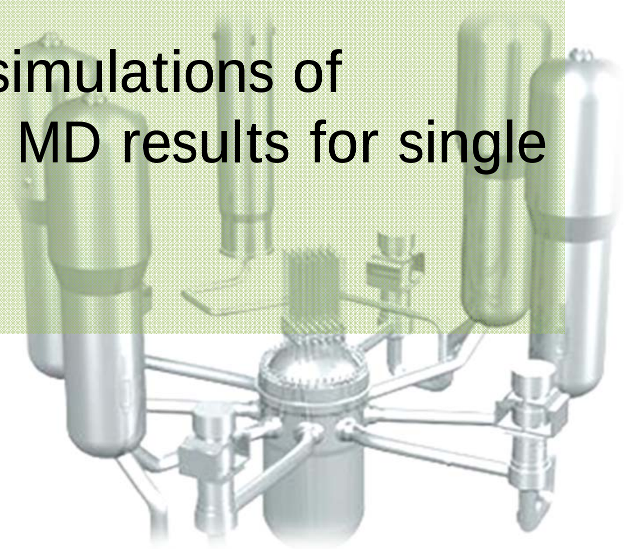
- Modified displacement cross section

$$\sigma_d^{mod}(E) = \sum_i \sigma_i(E) \int_{T_{min}}^{T_{max}} f_i(E,T) \cdot \nu_{NRT}(T) \cdot \eta\,(T)\, dT$$

~ σ_i & f_i : derive from recent ENDF/B library

~ η(T) : fraction of point defects (FMD+PDC) which escape from in-cascade recombination (cascade efficiency)

- depends on recoil atom energy (T)
- results from molecular dynamics (MD) simulations of displacement cascade (large amount of MD results for single elements)
- need experimental validation*



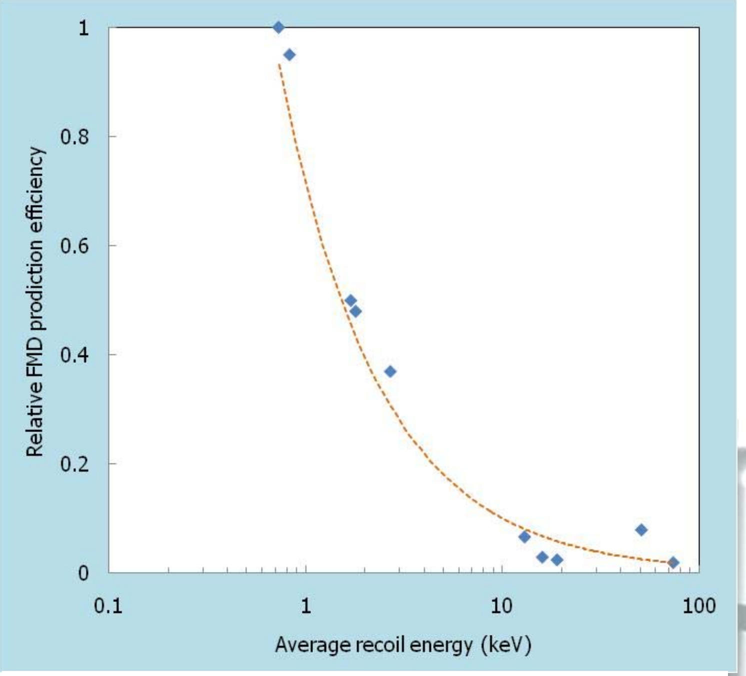
Proposed Work*

- Relative efficiency for FMD production
  ~ FMD production data obtained from RIS experiments
  ~ RIS is caused primarily by FMD migration

Irradiating particle	Target Material	Temperature	Average Recoil Energy (keV)	Relative Efficiency
1.0 MeV H^1	Ni-Si	350-650 °C	0.73	**1**
1.8 MeV H^1	Cu-Au	350 °C	0.83	0.95
1.8 MeV He^4	Cu-Au	350 °C	1.7	0.5
2.0 MeV He^4	Ni-Si	350-650 °C	1.8	0.48
2.0 MeV Li^7	Ni-Si	350-650 °C	2.7	0.37
1.8 MeV Ne^{20}	Cu-Au	350 °C	13	0.067
1.8 MeV Ne^{20}	Mo-7Re	1200 °C	16	0.03
1.8 MeV Ne^{20}	Mo-30Re	1200 °C	19	0.025
3 MeV Ni^{58}	Ni-Si	350-650 °C	51	0.08
3.25 MeV Kr^{84}	Ni-Si	350-650 °C	74	0.02

Reference

-T. Hashimoto, L.E. Rehn and P.R. Okamoto, Phys. Rev. B 38 (1988) 12868.
- R.A. Erck and L.E. Rehn, J. Nucl. Mater. 168 (1989) 208.
- L. E. Rehn, P. R. Okamoto and R. S. Averback, Phys. Rev. B30 (1984) 3073.



→ Normalized to 1 MeV proton irradiation having 100% efficiency with an average energy of 0.73 keV

→ Qualitatively compatible with MD simulation results

Thanks for your attention.

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