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Primary damage characteristics in different irradiation environments

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Outline

- Radiation damage
  - Damage correlation parameters
  - Displacement of atoms
  - NRT – model
  - PKA - spectrum
  - «Effective» dpa
- Primary damage characteristics in different reactor irradiation environments
  - WWER-1000
  - WWER-440
  - BN-600

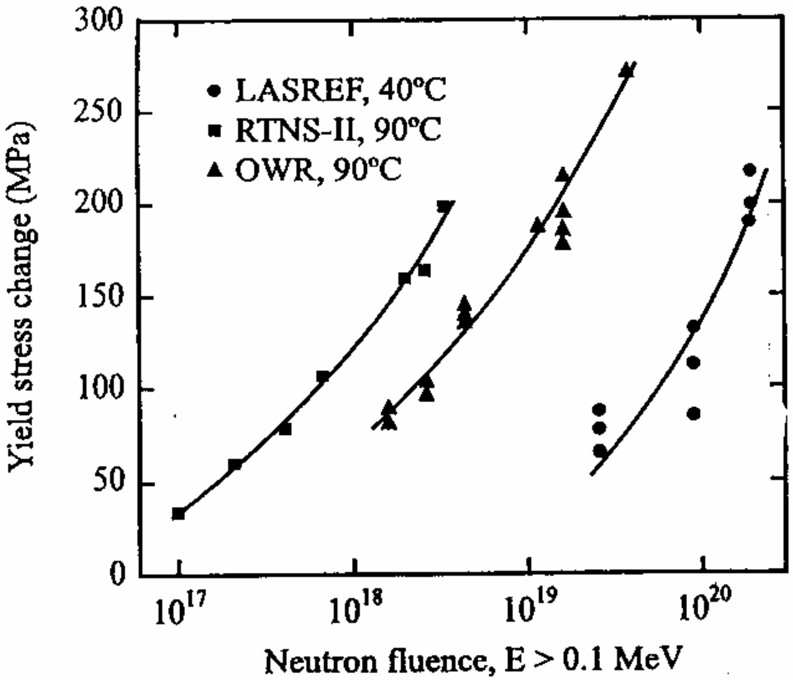
Introduction

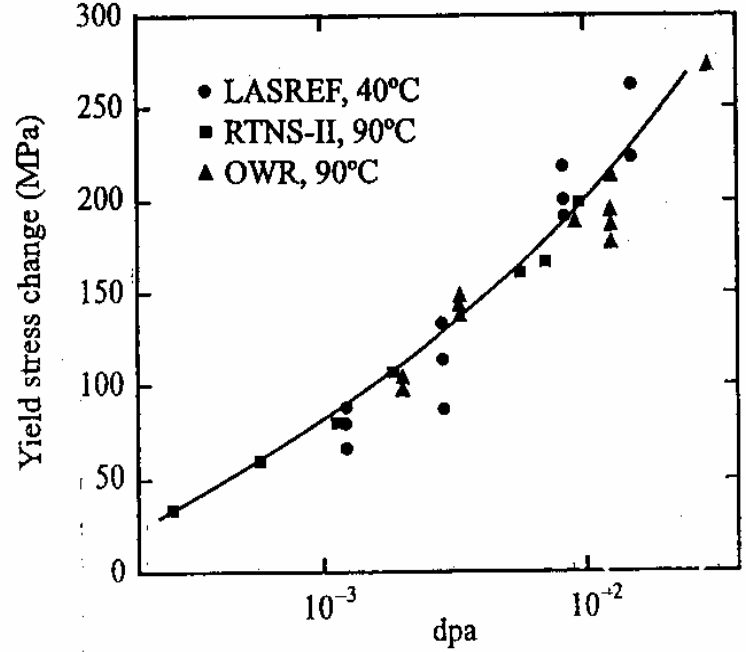
- For an analysis of radiation-induced phenomena in reactor structural materials the fast neutron fluence is usually used (for structural materials of the reactor core and internals the fluence of neutrons with energy >0.1 MeV, for RPV steels the fluence of neutrons with energy >0.5 MeV in Russia and East Europe, and with energy >1.0 MeV in USA and France).

- Displacements per atom (dpa) seem to be a more appropriate correlation parameter, especially, for structural materials of thermal reactors. The use of dpa allows to compare results of irradiation of materials in different neutron energy spectra or with different types of particles (neutrons, ions, fast electrons).

- Calculations of energy spectra of primary knocked atoms (PKA) and “effective” dpa, which are introduced to take into account the point defect recombination during the relaxation stage of a displacement cascade, are of both scientific and practical interests.

Correlation parameters





The increases in yield strength arising in single heat of 316 stainless steel during irradiation at relatively low temperatures in very diverse neutron spectra. OWR was a test reactor that had a typical LWR spectrum; RTNS-II produced a pure 14 MeV source and LASREF a very broad spectrum of high neutron energies. (after Grenwood, 1994, from Was, 2007)

While there is little correlation in terms of neutron fluence, the yield stress changes correlate well against damage dose in displacements per atom, dpa.

Dose rate

At an arbitrary location in the material the dose rate *K* (dpa/s) is given by the following expression :

$$K = \int_{E_{\min}}^{E_{\max}} \sigma_d(E)\varphi(E)\, dE \tag{1}$$

where $\varphi(E)$ is the incoming particles energy dependent flux, and $\sigma_d(E)$ is the displacement cross section

$$\sigma_d(E) = \int_{T_d}^{T_{\max}} \frac{d\sigma\ (E,T)}{dT}\ \nu(T)dT \tag{2}$$

where $\nu(T)$ is the number of displaced atoms per PKA of the energy T,

$d\sigma(E,T)/dT$ is the differential cross section for the transfer of energy T to the struck atom from the incoming particle of energy E

Radiation damage

The radiation damage event is defined as transfer of energy from an incident projectile to the solid and the resulting distribution of target atoms after completion of the event. The radiation damage event is actually composed of several distinct processes. These processes and their order of occurrence are as follows:

- **The interaction of an energetic incident particle with a lattice atom**
- **The transfer of kinetic energy to the lattice atom giving birth to a primary knock-on atom (PKA)**
- **The displacement of the atom from its lattice site**
- **The passage of the displaced atom through the lattice and the accompanying creation of additional knock-on atoms**
- **The production of a displacement cascade (collection of point defects created by the PKA)**
- **The termination of the PKA as an interstitial**

Production of defects

Time (s)	Event	Result
10^{-18}	Energy transfer from the incident particle	Creation of a primary knock-on atoms (PKA)
10^{-13}	Displacement of lattice atoms by the PKA	Displacement cascade
10^{-13}	Energy dissipation, spontaneous recombination and clustering	Stable Frenkel pairs (single interstitial atoms (SIA) and vacancies) and their clusters
$> 10^{-8}$	Defect reactions by thermal migration	SIA and vacancy recombination, clustering, trapping, defect emission

Approximate time-scale for the production of defects in irradiated metals (after Ullmaier, 1980 , from Was, 2007)

Neutron-Nucleus interactions

1. Elastic Scattering

When neutrons pass through a solid, there is a finite probability that they will collide with a lattice atom, imparting a recoil energy to the struck atom

2. Inelastic Scattering

Inelastic scattering is characterized by a reaction in which the emitted particle is experimentally the same as the captured particle, but there is a loss of kinetic energy in the system

3. (n, 2n) Reactions

Reactions such as the (n, 2n) reaction are important in radiation effects since they produce additional neutrons that can damage or transmutation reactions in components of interest

4. (n, γ) Reactions

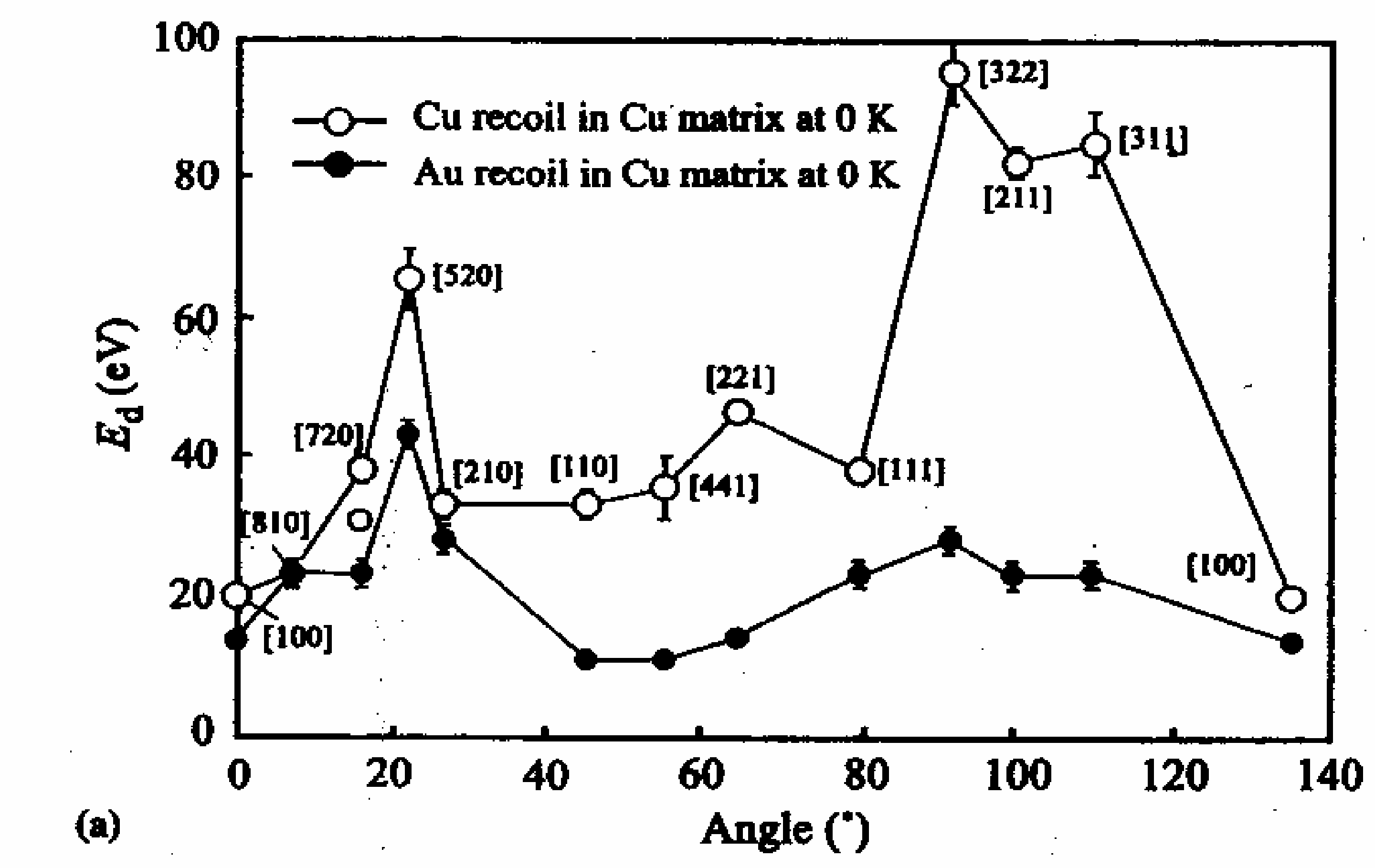
This reaction is important since the energy of the recoiling nucleus is sufficient to displace an atom

The details of energy transfer to lattice atoms and energy transfer cross sections for various types of neutron - nuclear reactions can be found e.g. in the book by G. Was, 2007

Energy transfer and energy transfer cross sections for various types of neutron - nuclear collisions (after Was, 2007)

Type of collision	Energy transfer and energy transfer cross section	Equation in text
Elastic scattering	$T = \frac{\gamma}{2} E_i (1 - \cos\phi)$	(1.13)
	$\sigma_s(E_i, T) = \frac{\sigma_s(E_i)}{\gamma E_i}$	(1.21)
Inelastic scattering	$T(E_i, Q_j, \phi) = \frac{\gamma}{2} E_i - \frac{\gamma}{2} \left[E_i \left(E_i + Q_i \frac{A+1}{A} \right) \right]^{1/2} \cos\phi + \frac{Q_j}{A+1}$	(1.27)
	resonance region	
	$\sigma_{s,j}(E_i, Q_j, T) = \frac{\sigma_{s,j}(E_i, Q_j)}{\gamma E_i \left(1 + \frac{Q_j}{E_i} \frac{l+A}{A} \right)^{1/2}}$	(1.30)
	unresolved resonance region	
	$\sigma_{is}(E_i, T) = \sigma_{is}(E_i) \int_0^{E_m'^{max}} \frac{f(E_i, E_m')}{4 \frac{1}{A+1} (E_i E_m')^{1/2}} dE_m'$	(1.31)
(n, 2n)	$T = \frac{A}{A-1} \frac{\eta_1}{\eta_2} E_m'' + \frac{A-1}{A} \bar{T}_\ell - 2 \left(\frac{\eta_1}{\eta_2} \right)^{1/2} (\bar{T}_\ell E_m'')^{1/2} \cos\phi$	(1.39)
	$\sigma_{n,2n}(E_i, T) = \int_0^{E_i - U} \frac{E_m'}{I(E_i)} e^{-E_m'/E_D} \times \int_0^{E_i - U - E_m'} \frac{E_m''}{I(E_i, E_m')} e^{-E_m''/E_D} dE_m' dE_m''$	(1.40)
(n, γ)	$\bar{T} \cong \frac{E_\gamma^2}{4(M+m)c^2}$	(1.42)
	$\sigma_{n,\gamma}(E_i) = \sigma_0 \sqrt{\frac{E_0}{E_i}} \left\{ \frac{1}{[(E_i - E_0)/(\Gamma/2)]^2 + 1} \right\}$	(1.44)

Displacement energy



Displacement energy E_d as a function of recoil direction in (a) fcc Cu and Au crystal (after Bacon, 1993, from Was, 2007)

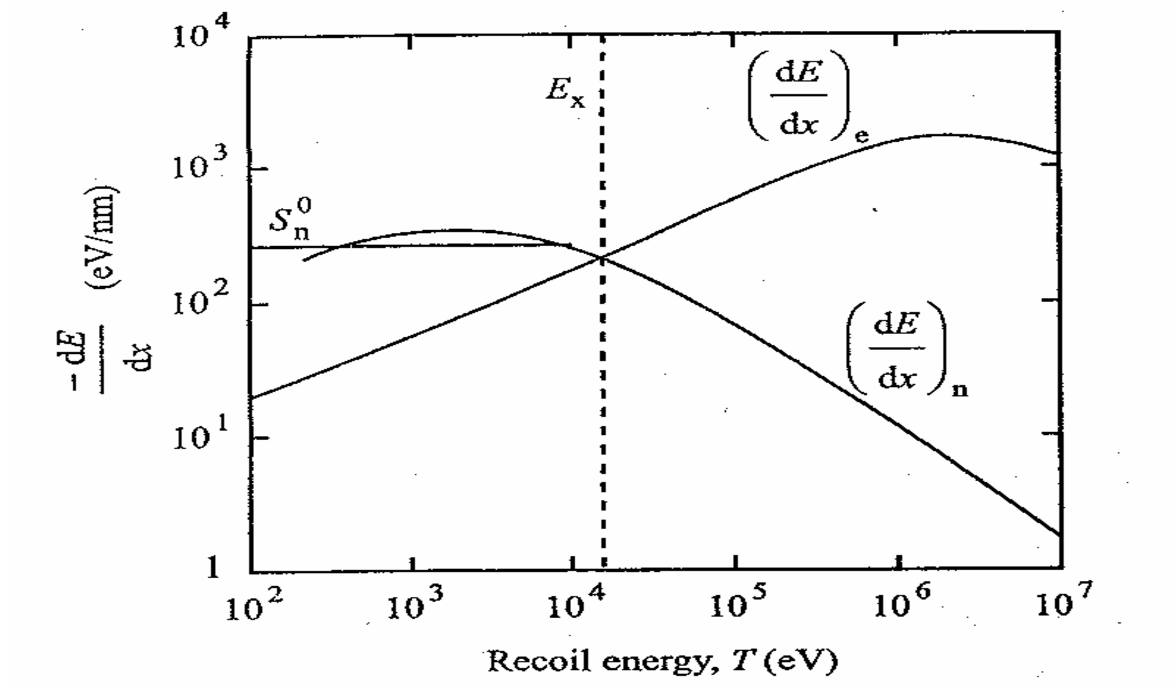
A lattice atom must receive a minimum amount of energy E_d in the collision to be displaced from its lattice site. Some averaged effective energy for each metal is used usually in displacement calculations.

Effective displacement energies

Metal	Lattice (c/a)	$E_{d,min}$ (eV)	E_d (eV)
Al	fcc	16	25
Ti	hcp (1.59)	19	30
V	bcc	–	40
Cr	bcc	28	40
Mn	bcc	–	40
Fe	bcc	20	40
Co	fcc	22	40
Ni	fcc	23	40
Cu	fcc	19	30
Zr	hcp	21	40
Nb	bcc	36	60
Mo	bcc	33	60
Ta	bcc	34	90
W	bcc	40	90
Pb	fcc	14	25
Stainless steel	fcc		40

Recommended values of the effective displacement energies for use in displacement calculations (from ASTM E521 (1996) Standard Practice)

Energy loss contributions



When an ion or atom (e.g. PKA) is traveling through a lattice, the total energy loss per unit length – dE/dx can be approximated by a sum of the components:

$$\left(-\frac{dE}{dx}\right)_{total} = \left(-\frac{dE}{dx}\right)_n + \left(-\frac{dE}{dx}\right)_e + \left(-\frac{dE}{dx}\right)_r, \qquad \textbf{(3)}$$

Where the subscripts are defined as follows:

n = elastic, e = electronic, r = radiation

For most of the applications energy loss by radiation is small.

In the simple Kinchin-Pease (1955) displacement model (hard-sphere elastic collisions) only elastic energy losses are accounted at $T<E_x$ and only electronic ones at $T>E_x$ for PKA

Displacement of atoms

Now the *NRT* – model (Norgett, Robinson and Torrens, 1975) is widely used for dpa calculations)) with more accurate Lindhard's partitioning between elastic and electronic energy losses:

$$\nu_{NRT} = \frac{0,8}{2T_d} T \frac{1}{1 + k_0 g(\varepsilon)} \qquad (4)$$

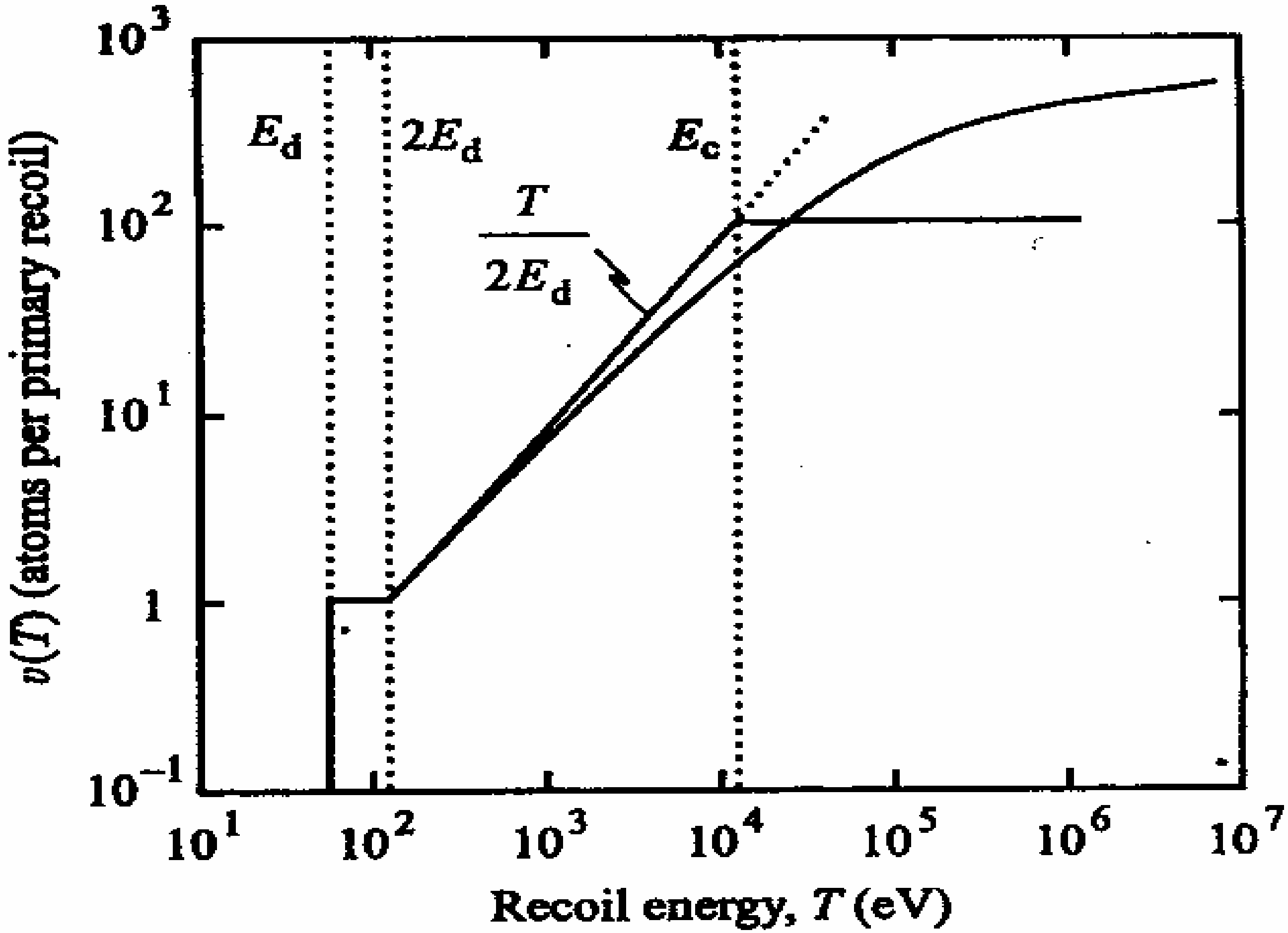
where $k_0 = 0,1337\ Z^{2/3} A^{-1/2}$; ($k_0 = 0,157$ for Fe);

$$g(\varepsilon) = \varepsilon + 0,4024\varepsilon^{3/4} + 3,401\varepsilon^{1/6} \quad ; \quad \varepsilon = T / 0,086937\ Z^{7/3}$$

($\varepsilon = T / 174,104$ for Fe)

Z – the atomic number, *A* – the atomic mass.

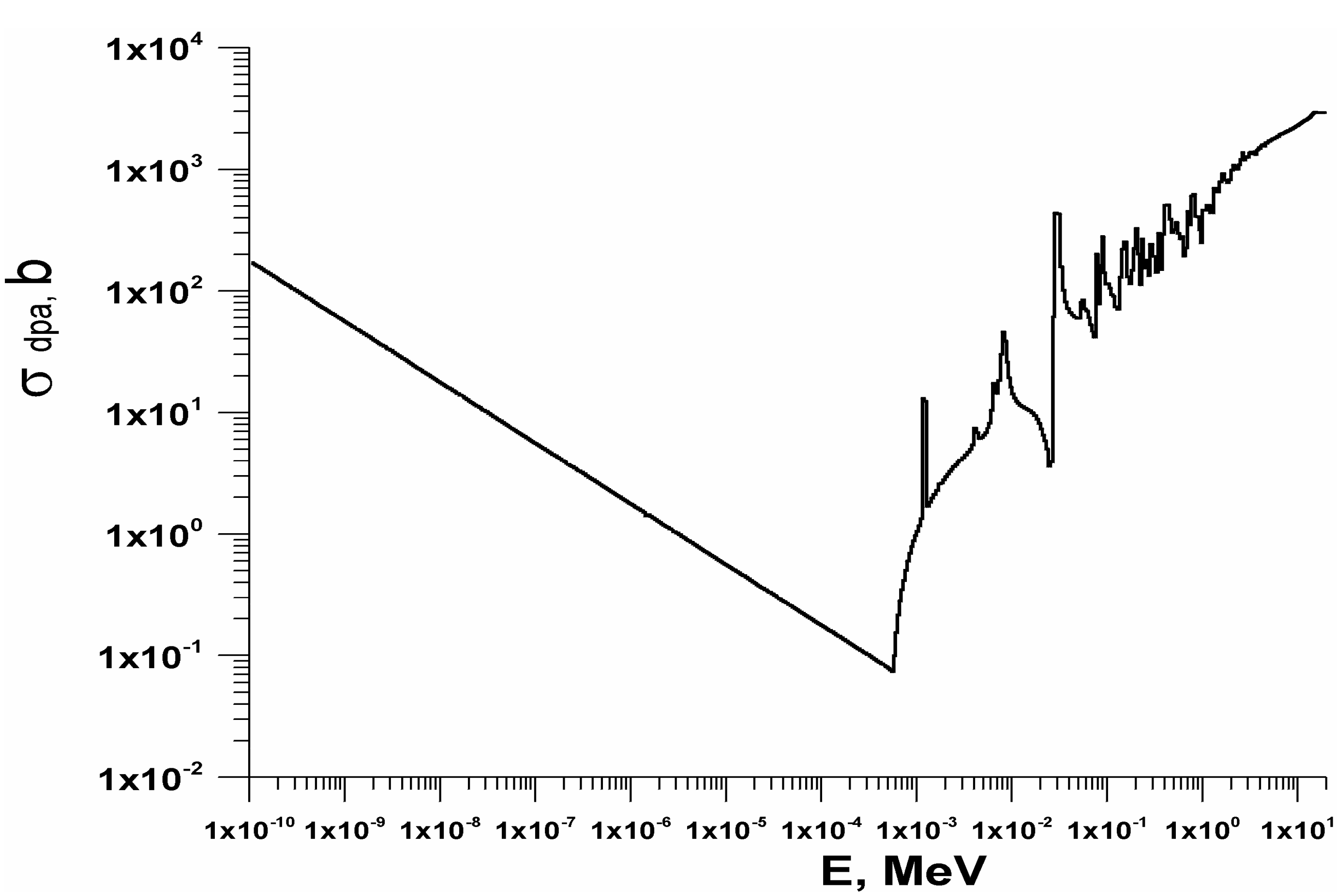
Displacement of atoms



Number of displaced atoms per primary recoil (PKA) in graphite using Lindhard's model compared to simple Kinchin-Pease of $T/2E_d$ (after Robinson, 1969, from Was, 2007)

E_c is the energy dissipated by PKA in elastic collisions

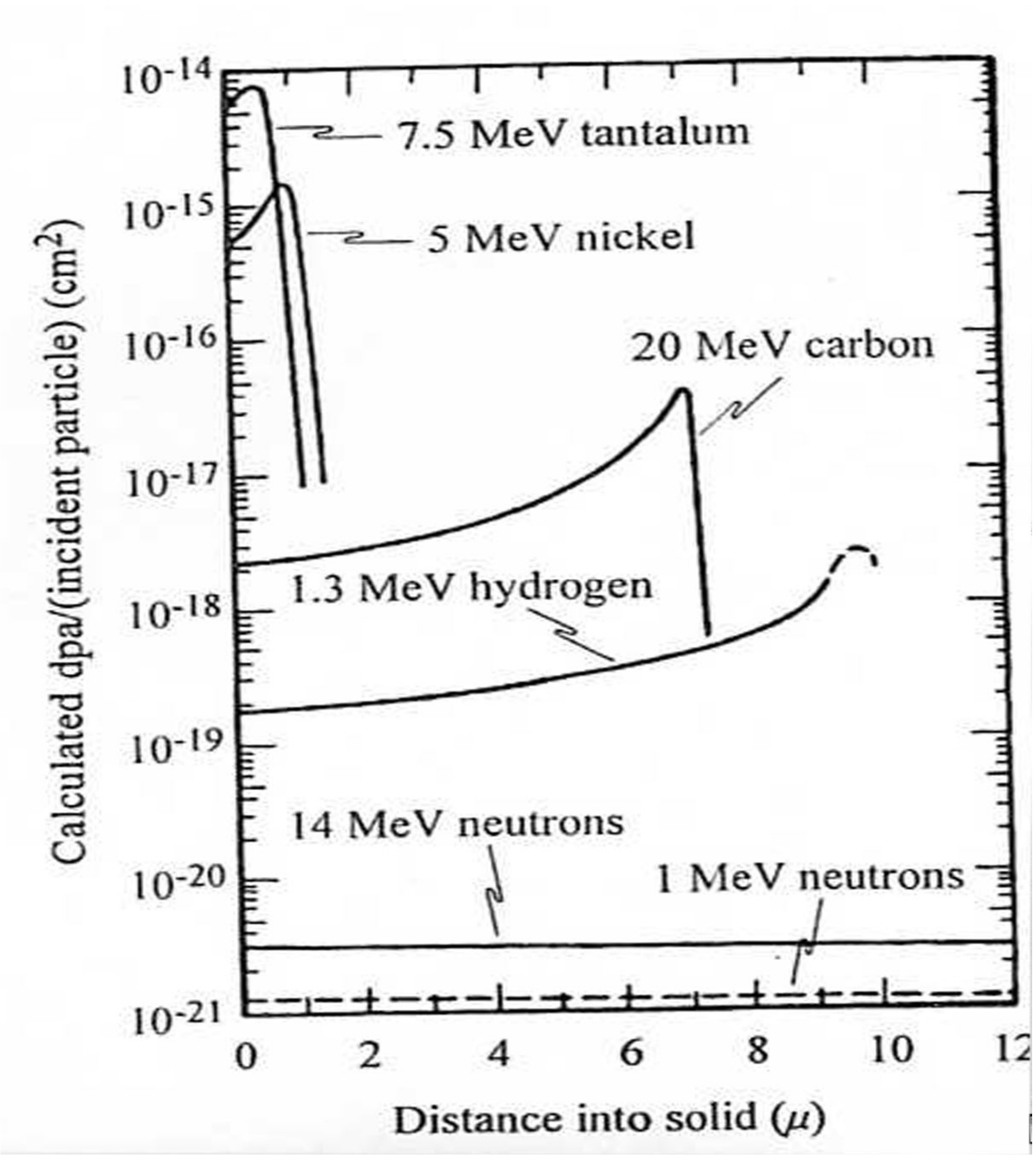
Displacement cross section



The displacement cross section (in barns, 10^{-24} cm^2) in dependence on neutron energy (in MeV) for Fe based on a Lindhard model and ENDF/B cross sections

$\sigma_{DPA}^{Fe}(E)$ **is large for neutrons with very low and very high energies**

Damage effectiveness



Displacement-damage effectiveness for various energetic particles in Ni (after Kulcinski, 1972)

Ions of heavier mass have a shorter penetration distance and higher damage rates

Charge particle irradiation

Electrons

Advantages
Relatively "simple" source – TEM
Uses standard TEM sample
High dose rate – short irradiation times

Disadvantages
Energy limited to ~1 MeV
no cascades
Very high beam current (high dpa rate) requires high temperature
Poor control of sample temperature
Strong "Gaussian" shape (nonuniform intensity profile) to beam
No transmutation

Heavy Ions

Advantages
High dose rate – short irradiation times
High T_{avg}
Cascade production

Disadvantages
Very limited depth of penetration
Strongly peaked damage profile
Very high beam current (high dpa rate) requires high temperature
No transmutation
Potential for composition changes at high dose via implanted ion

Protons

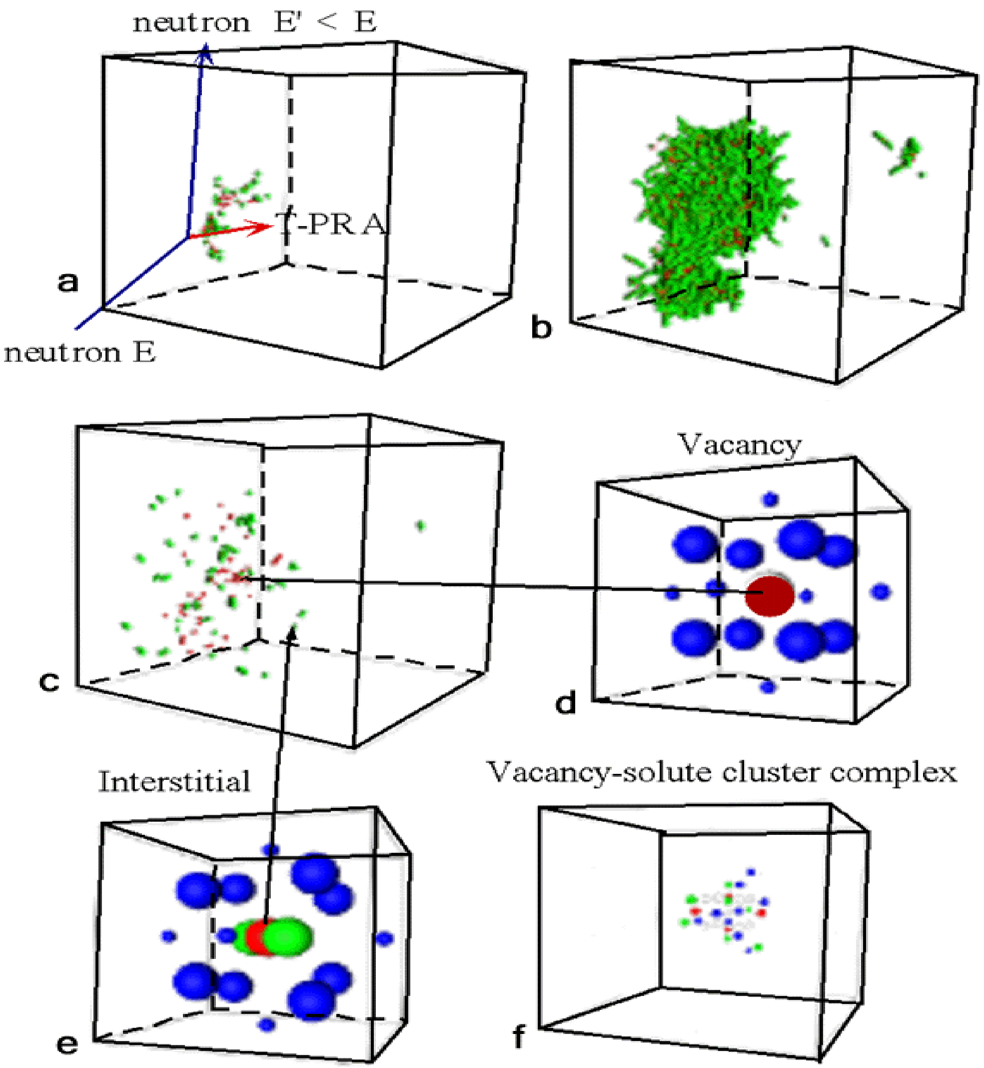
Advantages
Accelerated dose rate – moderate irradiation times
Modest ΔT required
Good depth of penetration
Flat damage profile over tens of μm

Disadvantages
Minor sample activation
Smaller, widely separated cascades
No transmutation

Advantages and disadvantages of various particles types in simulating neutron irradiation

(from Was, 2007)

Stages of cascade development



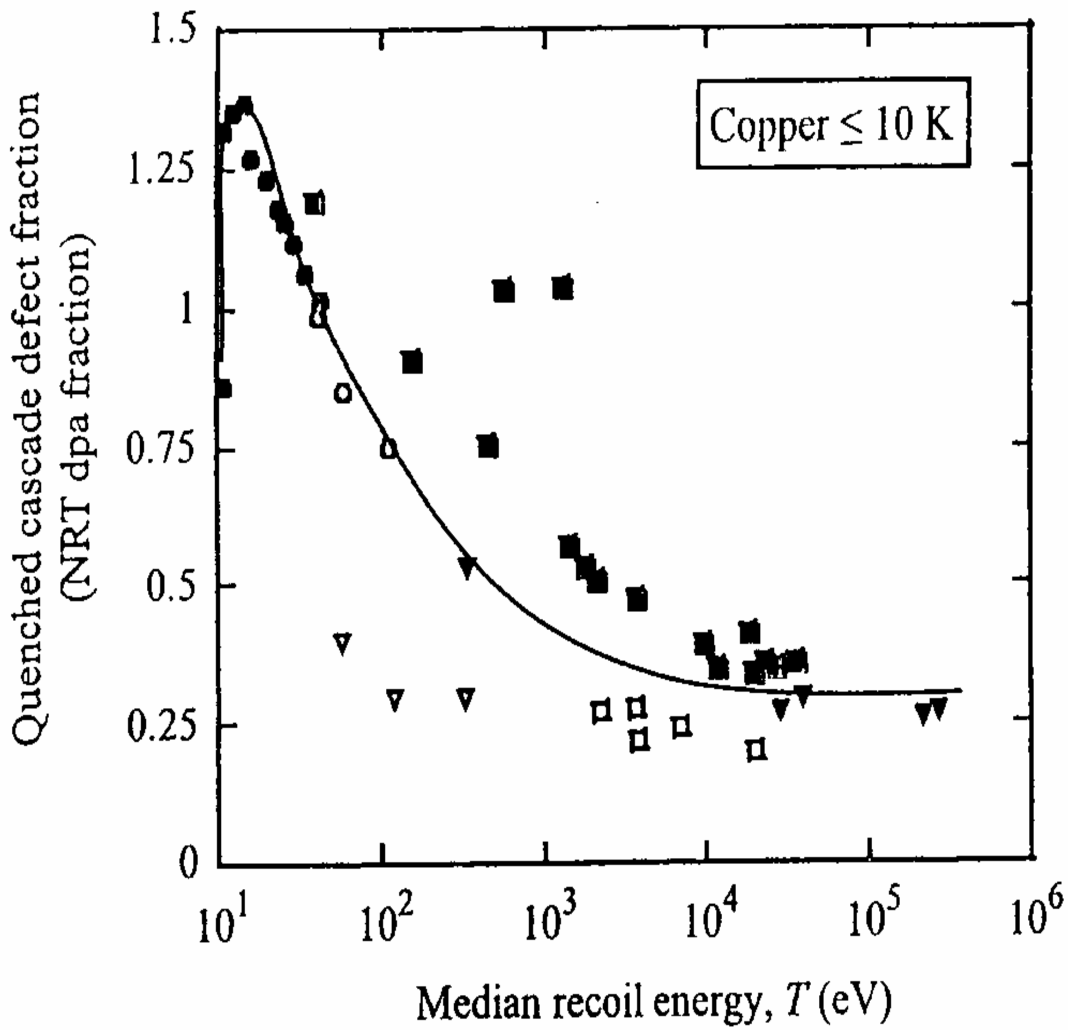
An illustration of cascade primary-damage production (iron atoms not shown in a–c and f): (a–c) MD simulation snapshots of initial, intermediate and final dynamic stage of a displacement cascade; (d–e) vacancy and self interstitial defects; (f) vacancy-solute cluster complex formed after long-term cascade aging (from Odette, 2001)

Stages of cascade development

Cascades evolve in stages and time-scale given as follows:

- *Collisional (< 0.1 ps).* A cascade of displacive collisions continues until no atom have enough energy to create further displacements
- *Thermal spike ($\sim$ 0.1 ps).* The spike occupies a region in which the energy is high enough so that the atoms resemble molten material
- *Quenching ($\sim$ 10 ps).* Molten zone returned to be condensed and thermodynamic equilibrium is established. The total number of point and clustered defects is much less than the number of atoms displaced in the collisional stage
- *Annealing ($>$ 1 ns).* This stage lasts until all mobile defects escape the cascade region

Cascade efficiency



Dependence of the cascade efficiency

$\eta=\nu(T)/\nu_{NRT}(T)$

on median PKA in copper.

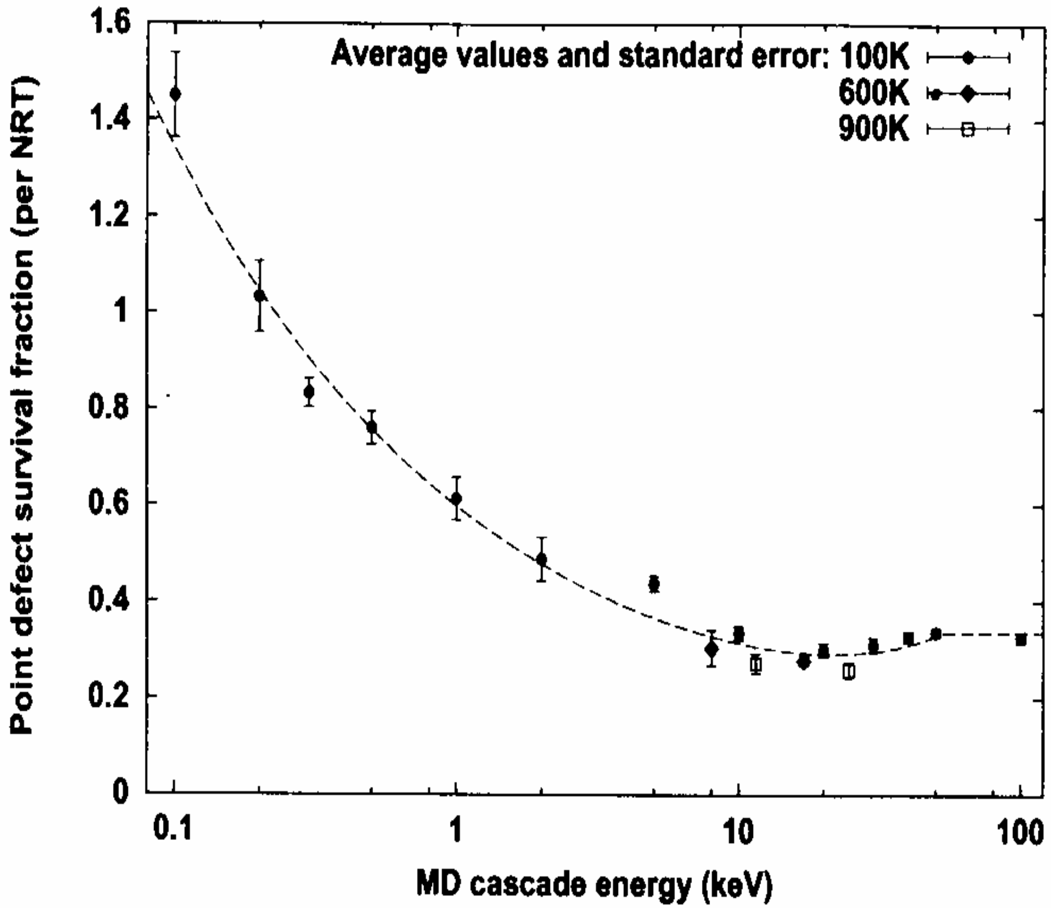
Open symbols

refer to MD calculations and *filled symbols* refer to experimental measurements as the result of electron (●), ion (■), fission fragment (♦) and neutron (▼) irradiation at low temperatures

(after Zinkle, 1993 , from Was, 2007)

«Effective» dpa

Number of displacements per atom calculated with account of only defects surviving after in-cascade recombination will be called as «effective» dpa.



Energy dependence of total point defect survival η_{MD} in Fe obtained by R. Stoller from MD cascade simulations. MD defect survival divided by NRT displacements.

The ratio $\eta_{MD}=\nu(T)/\nu_{NRT}(T)$ was approximated as follows:

$$\boldsymbol{\eta_{MD}=0.5608T^{-0.3029}+3.227 \times 10^{-3}\, T}$$

PKA - spectrum

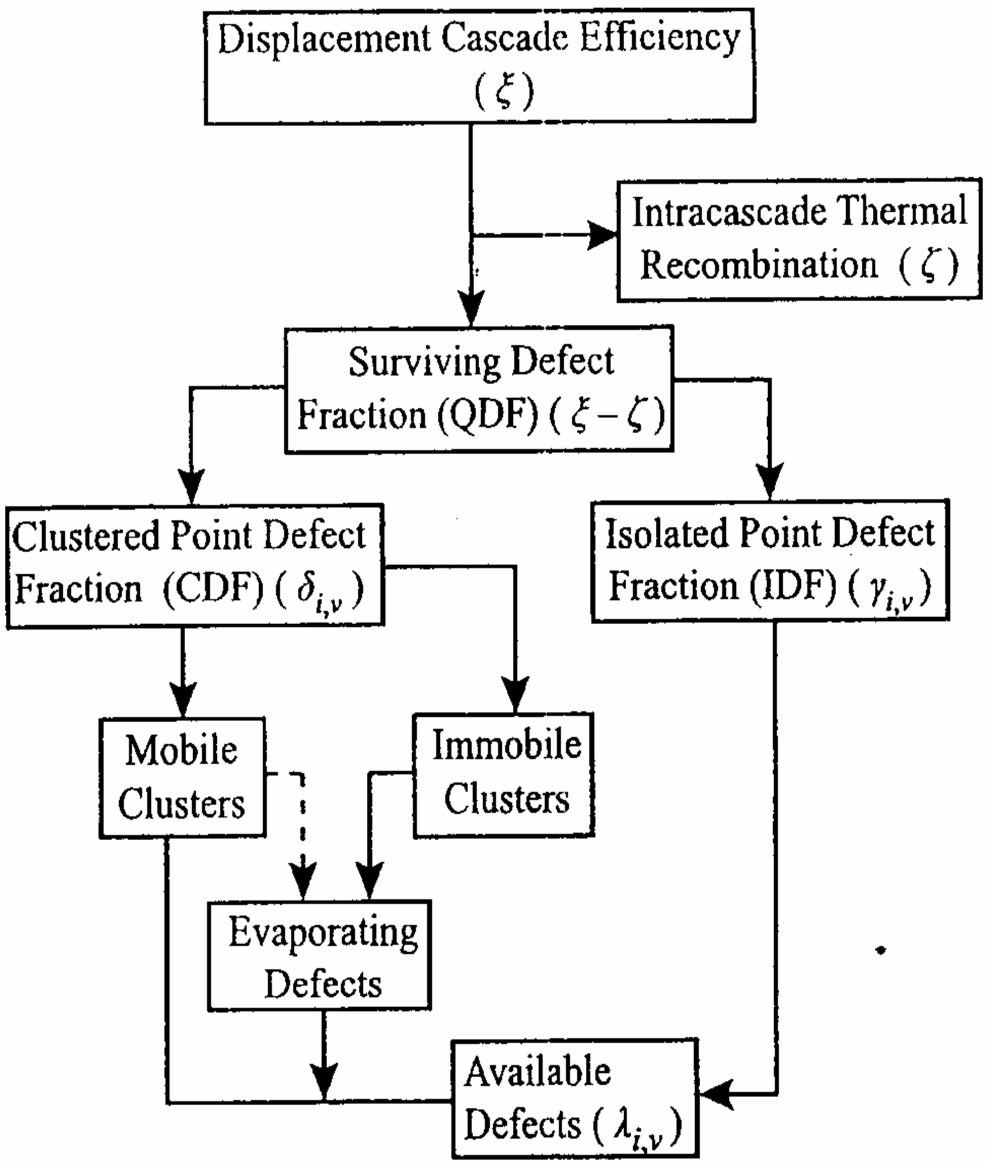
Inserting eqn. (2) in eqn. (1) and changing the order of integration, one can find

$$K = \int_{T_d}^{T_{\max}} dT \cdot \nu(T) \cdot \int_{E_T}^{20\,MeV} \varphi(E) \cdot \frac{d\sigma^{Fe}(E,T)}{dT} dE \qquad (5)$$

where the integral over E is the spectrum of energies transferred to lattice atoms (if $T>T_d$, then the spectrum of primary knocked atoms, PKA-spectrum).

Substitution of $\nu_{NRT}(T)$ or $\nu_{MD}(T) = \eta(T)\ \nu_{NRT}(T)$ in (5) allows to calculate dpa (NRT) or "effective" dpa respectively

Freely migrating defects



Interdependence of isolated point defects, mobile defect clusters, and thermally evaporating defect clusters that contribute to the fraction of surviving defects that are “available” for radiation effects (from Was, 2007)

Primary damage characteristics in different reactor irradiation environments

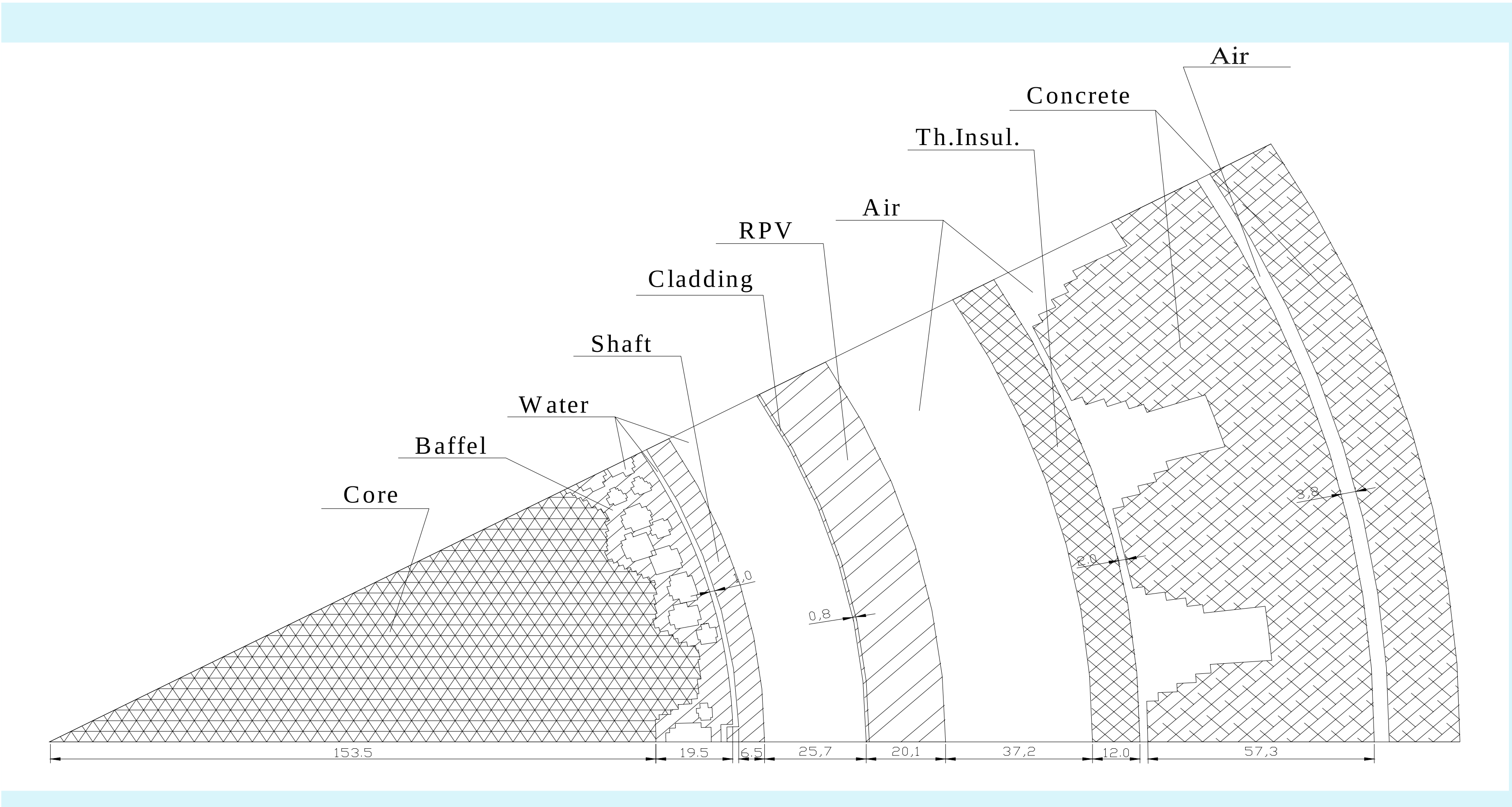
Some damage characteristics : dpa (NRT), “effective” dpa, PKA-spectrum are considered in different locations of Russian power reactors WWER-1000 and WWER-440 (both thermal reactors) and BN-600 (fast reactor). Most calculations are performed for Fe.

Besides, the radial function $K(r)$ that defines the ratio between the damage exposure in DPA units and in fast neutron fluence (FNF) units are calculated

$$K(r) = \int_{E_T}^{20MeV} \varphi(r,E)\sigma_{DPA}^{Fe}(E)dE \Bigg/ \int_{E_{fast}}^{20MeV} \varphi(r,E)dE$$

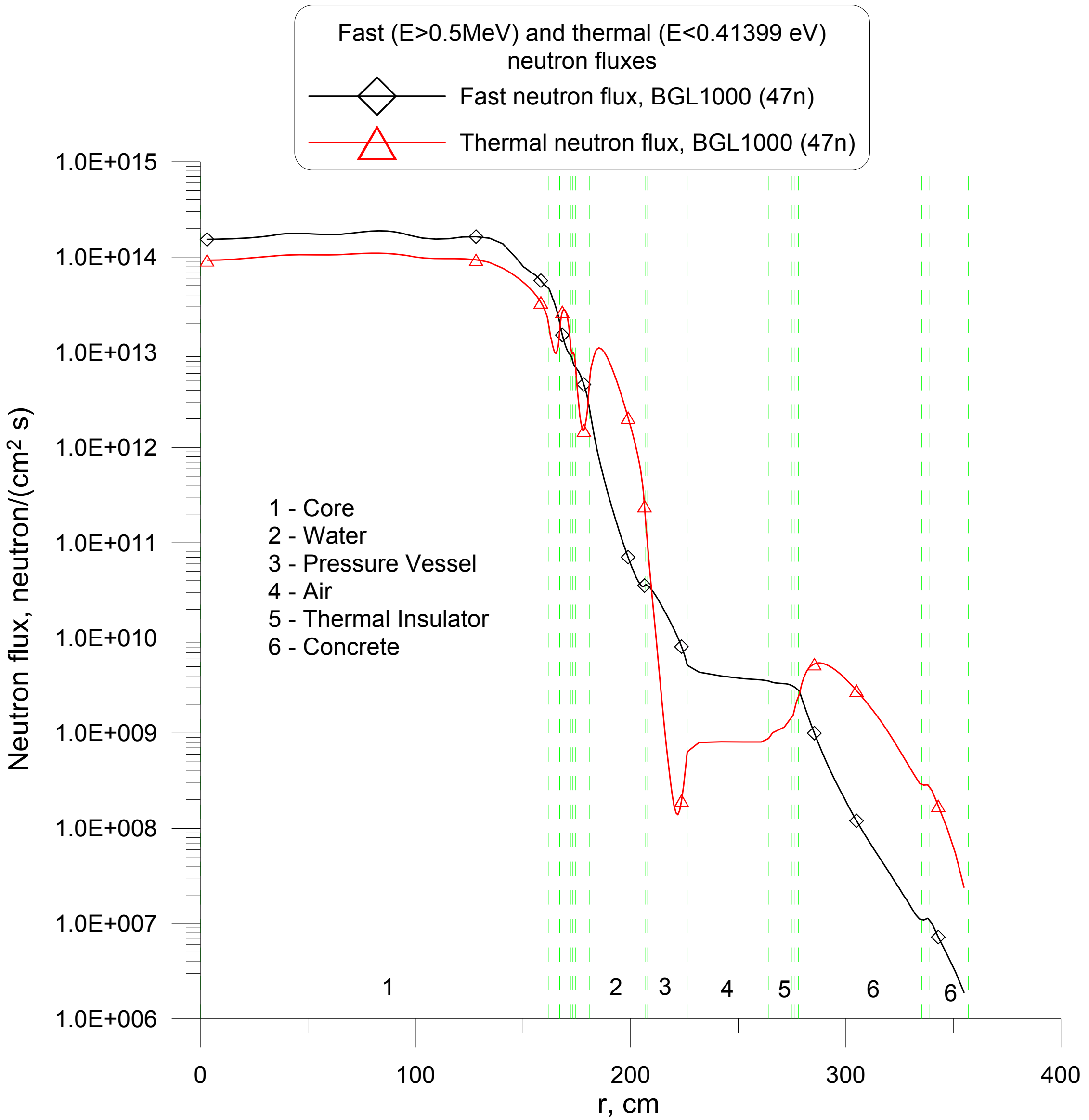
where $\sigma_{DPA}^{Fe}(E)$ is the DPA (NRT) cross-section for iron, E_{fast} is the lower energy boundary of the fast neutrons region (*0.1, 0.5 and 1.0 MeV.In Russian normative documents 0.5 MeV for RPV*) and E_T is the lower energy boundary of the cross-section library used.

Calculation of neutron and photon fluxes in WWER-1000



An approximation of the 30° symmetry sector of the WWER-1000 in geometry (r, ϑ)

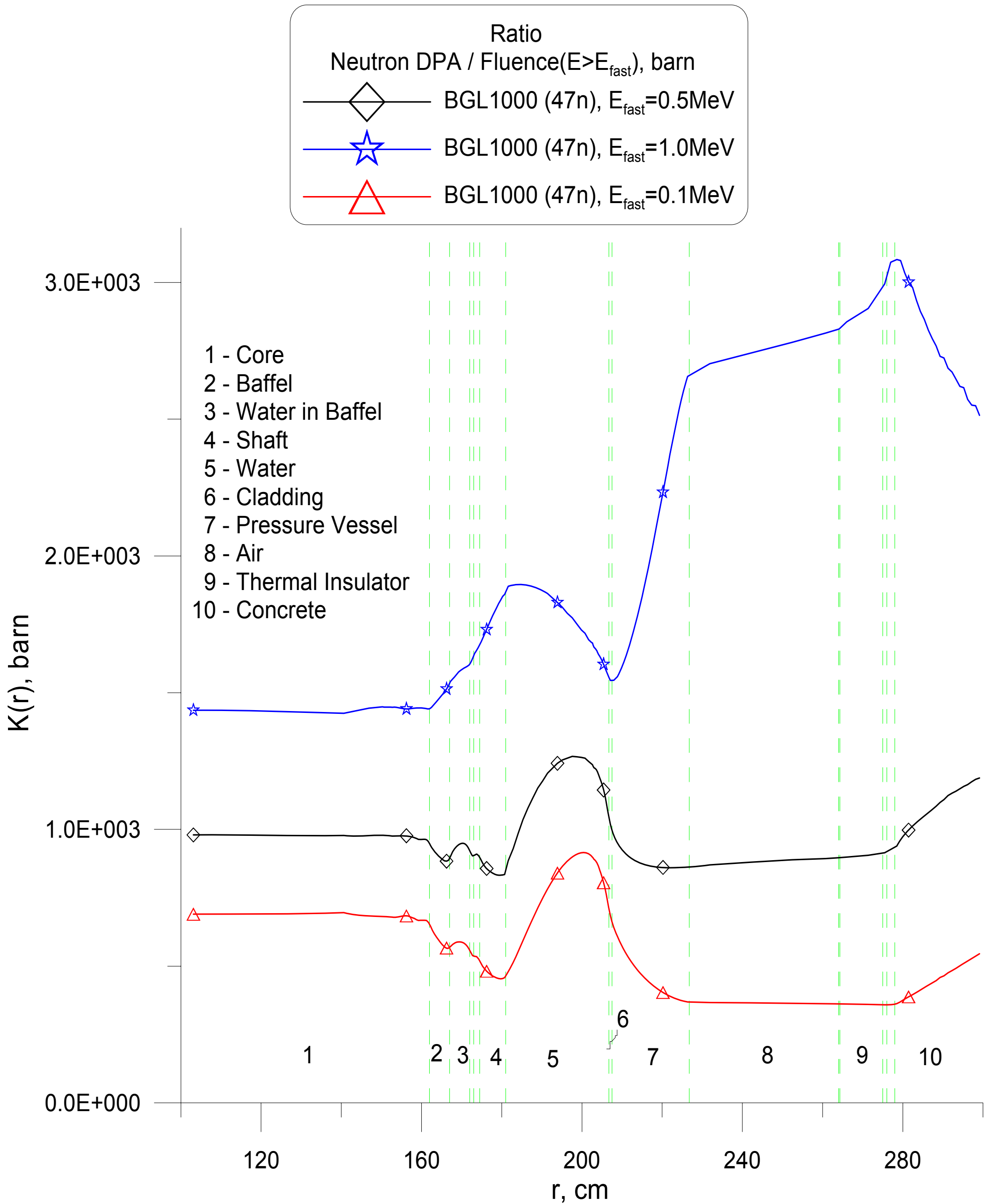
Fast and thermal neutron fluxes



Fast and thermal neutron flux radial distributions in the point $\vartheta = 10°$ $z = 211.8$ cm.

In the spatial zones, where the neutron spectrum becomes softer, the sufficiently large number of thermal neutrons can give an essential contribution in DPA that is comparable with the fast neutrons one.

Relation between DPA and FNF units in WWER-1000

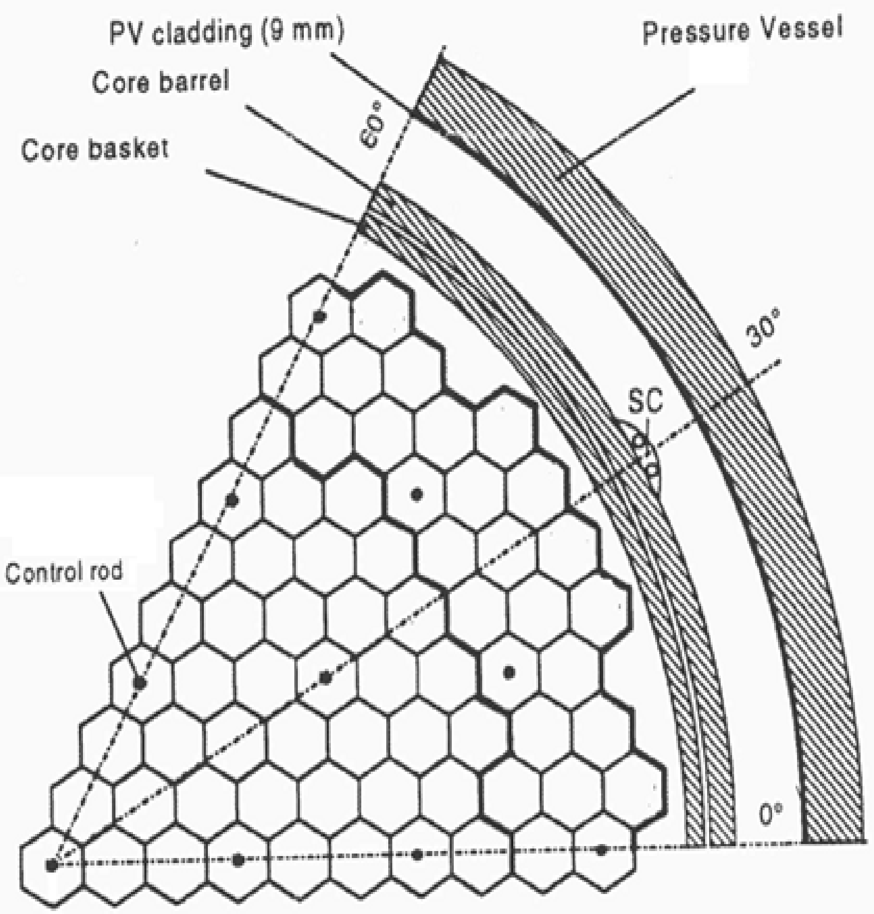


Ratio of the damage exposure in the DPA units to the one measured in the FNF units in the point $\vartheta = 10°$, $z = 211.8$ cm.

There is no linear dependence between damage exposure given in DPA and FNF units.

The FNF is not a universal quantity for definition of the damage exposure and the use of DPA for different elements of reactor construction seems more universal tool for this purpose, as it takes into account all available neutrons.

Calculation of damage characteristics in WWER-440



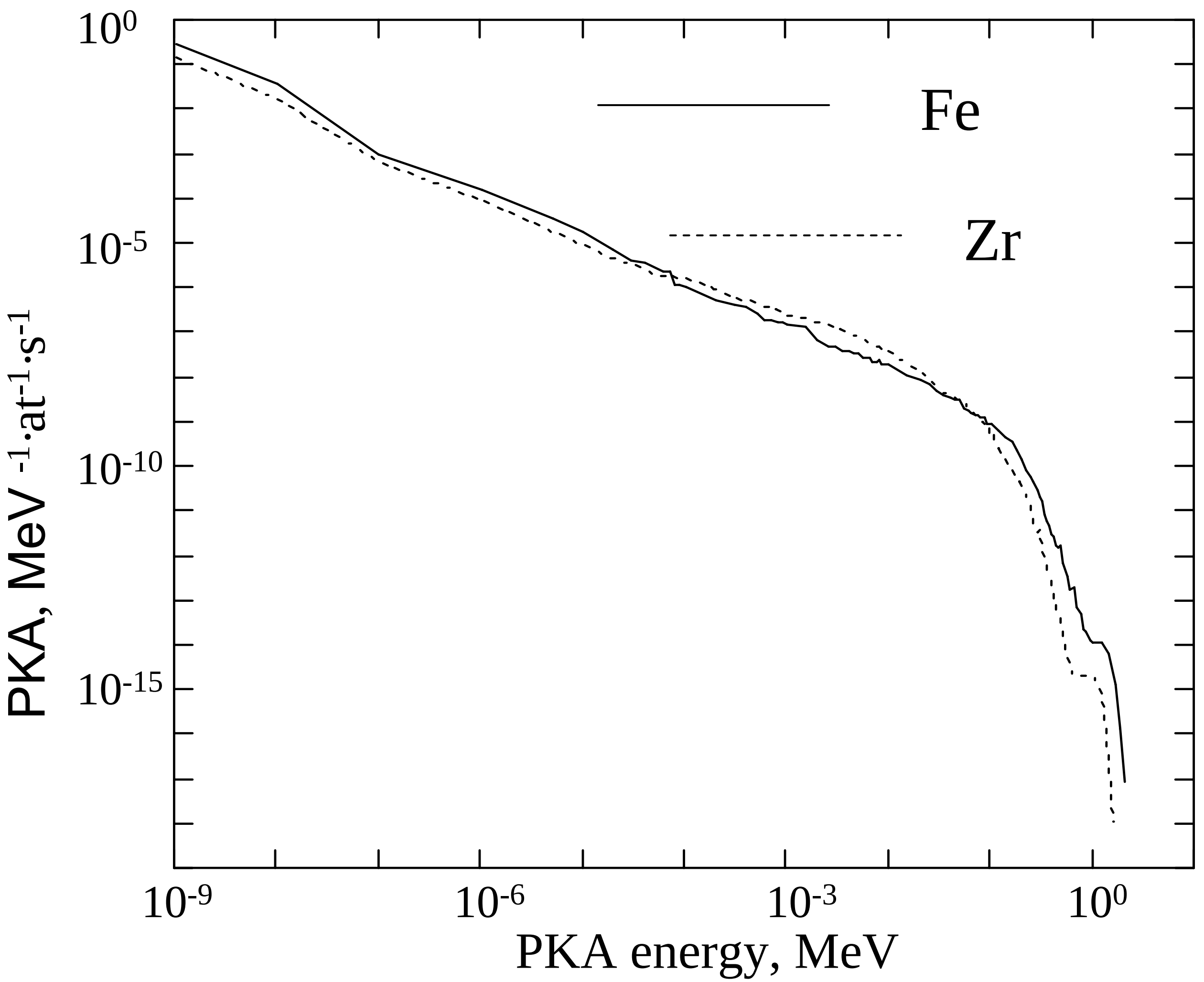
Location of surveillance chains (SC) in VVER-440/230 reactor.

This location is very near to the core, so the damage rate here is an order of value higher then in RPV

Neutron and gamma spectra at different locations in WWER - 440 were obtained using the 3D transport code KATRIN developed by Voloschenko et al

PKA-spectra were calculated using the SPECTER code developed by Greenwood

PKA energy spectrum in Fe and Zr in the core center of WWER-440



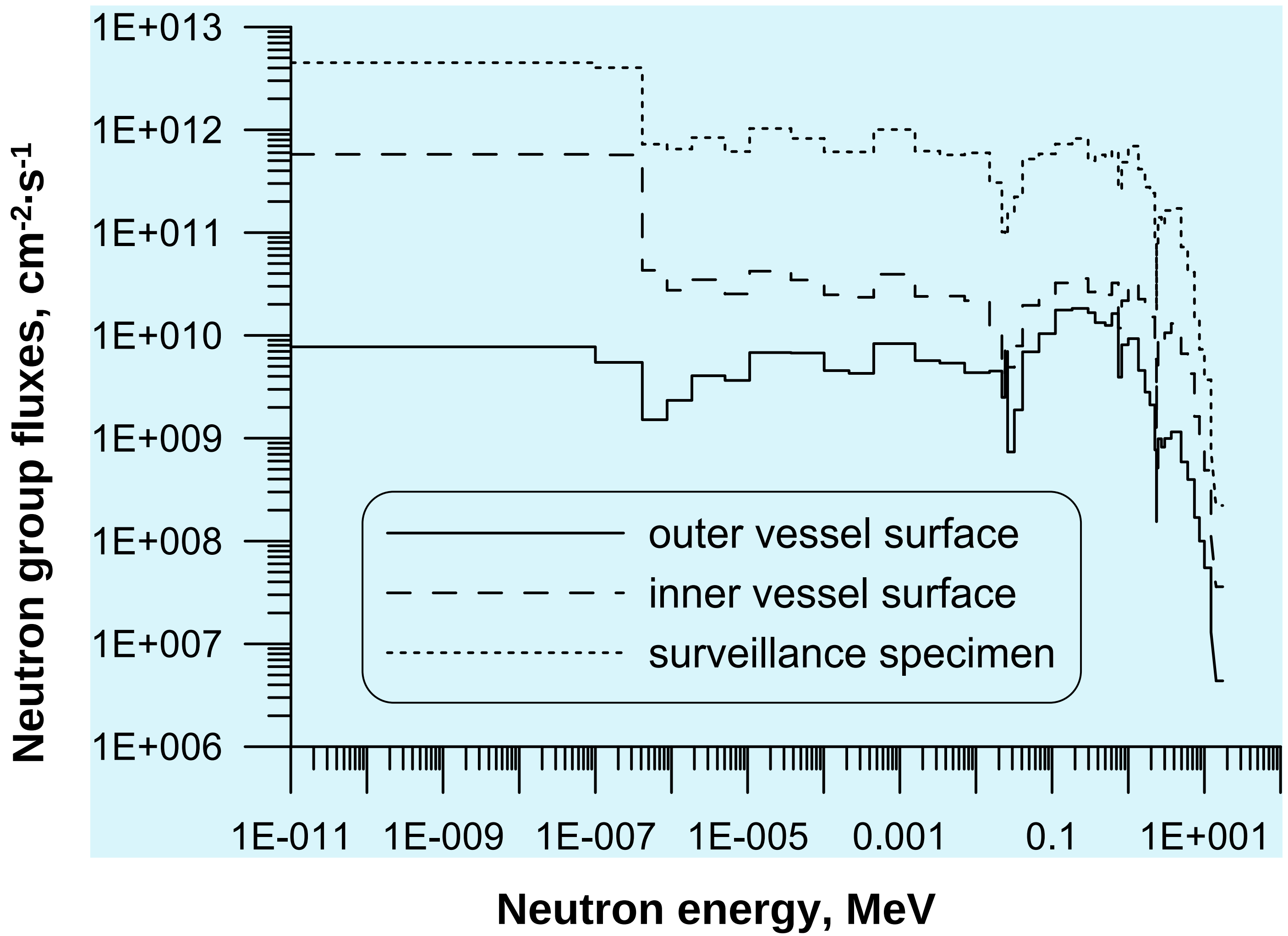
Generation rate of PKAs with the energies T from 40 eV (E_d for Fe and Zr) to 0.1 MeV is higher in Zr, but with higher T in Fe

Neutron fluxes, dose rates and mean PKA energies in the core of WWER-440

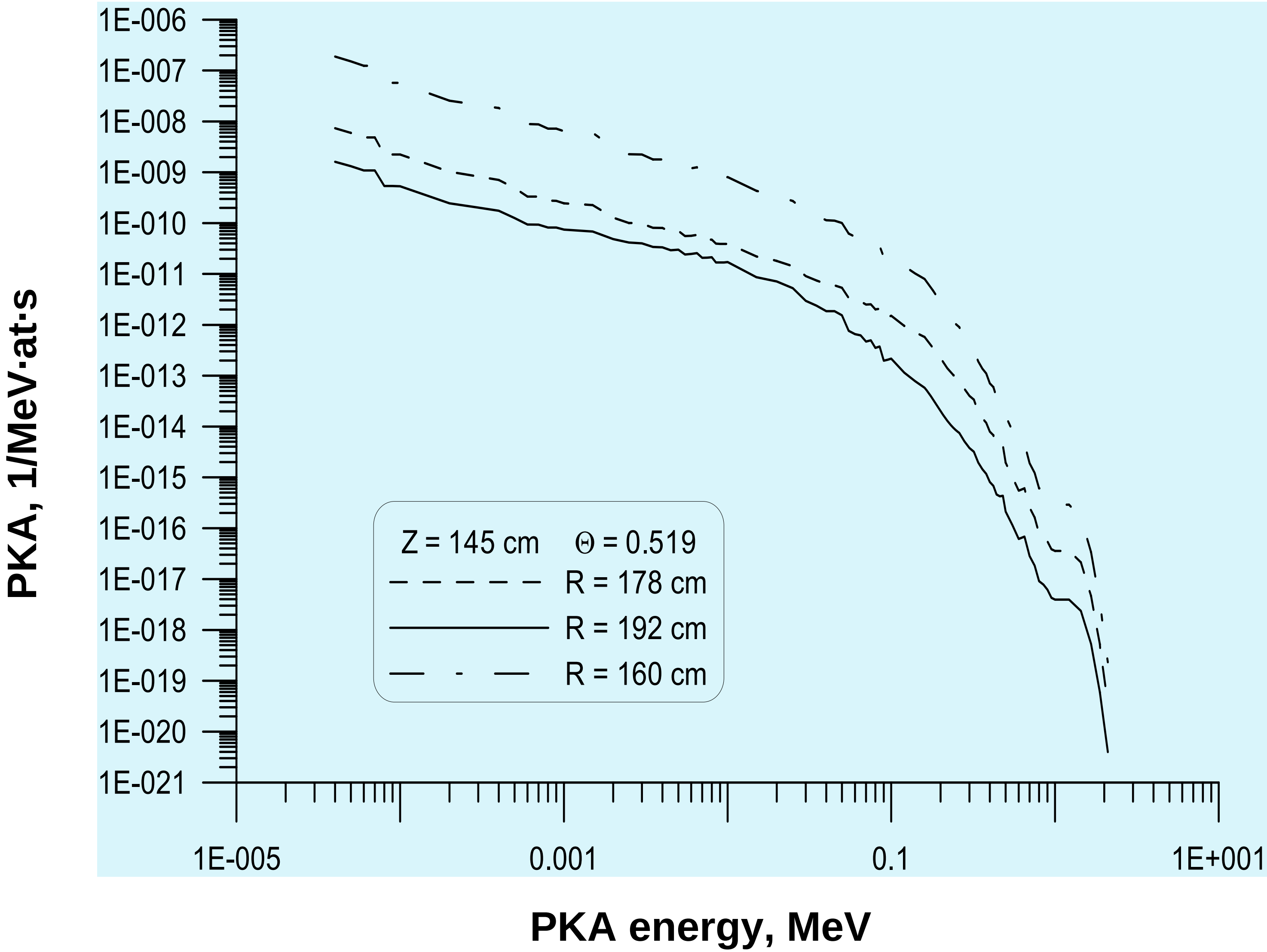
Fast neutron flux, 10^{14} n/(cm^2×s)			Dose rate (effective dpa) 10^{-7} dpa/s		Mean energy of PKAs, keV	
E > 0.1 MeV	E > 0.5 MeV	E >1 MeV	Fe	Zr	Fe	Zr
1.77	1.26	0.853	1.24 (0.396)	1.32 (0.409)	15.6	8.89

Mean energy of PKAs is higher in Fe, but dose rates are similar in Fe and Zr

Neutron spectra in surveillance and RPV locations



PKA – spectra in the same surveillance and RPV locations



Mean energies of neutrons and PKAs

WWER-440 locations,	Mean neutron energy, MeV	Mean energy of neutrons with energies above 454 eV, MeV	Mean energy of PKAs, keV
surveillance specimens	0.304	0.673	10.9
inner vessel surface	0.253	0.892	14.5
outer vessel surface	0.432	0.54	10.6

Mean energy of PKAs does not correlate with mean neutron energy and depends on neutron spectrum.

Neutron fluxes, dose rates and mean PKA energies in WWER-440 and WWER-1000

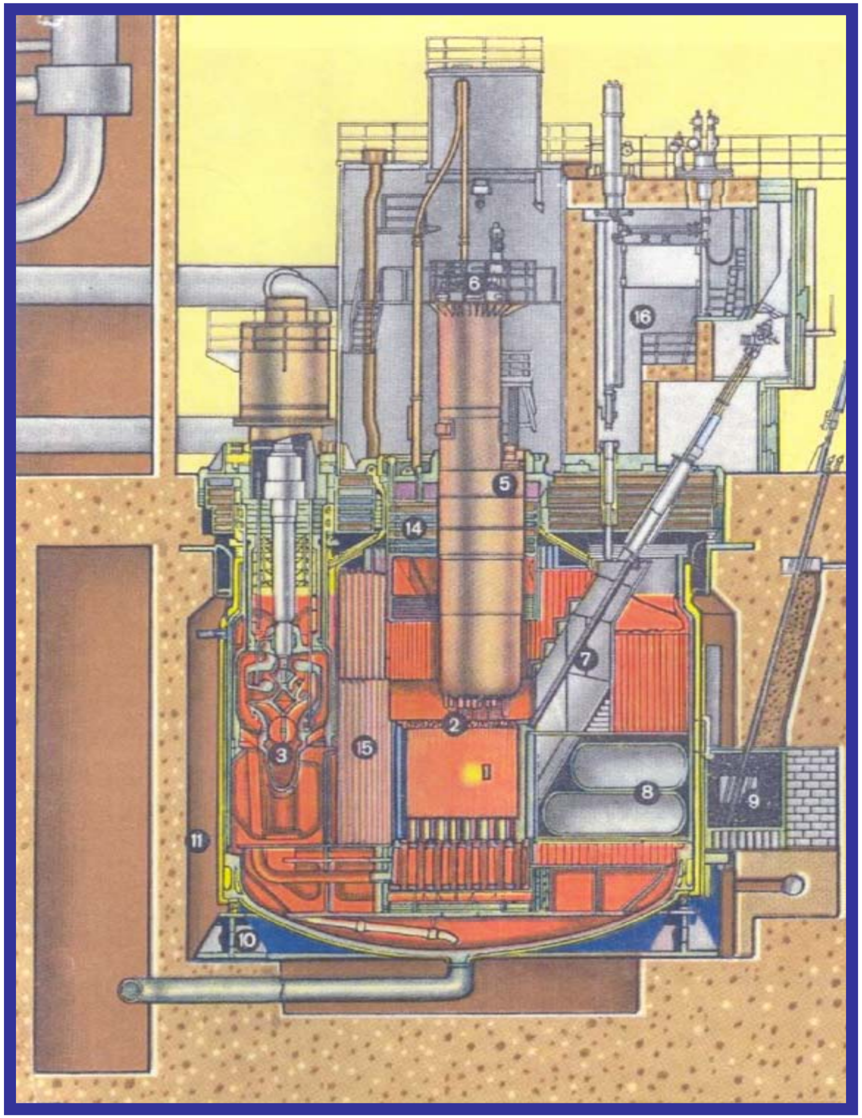
Type of reactor	Locations	Neutron flux, 10^9 n/cm^{2}s		Dose rate, 10^{-12} dpa$_{NRT}$/s ν_{NRT}	Dose rate calculated using σ_D, eq.(1) 10^{-12} dpa$_{NRT}$/s	Dose rate, 10^{-12} dpa$_{eff}$/s	Mean energy of PKA (keV)
		>0.5 MeV	>1 MeV				
WWER – 440	surveillance specimens	4450	2540	4130	4080	1330	10.9
WWER – 440	inner vessel surface	245	154	243	242	75.7	14.5
WWER – 440	outer vessel surface	66.5	25.5	57.7	55.6	18.8	10.6
WWER – 1000	inner vessel surface	35.4	22.3	35.0	34.9	11.0	16.6
WWER – 1000	outer vessel surface	5.23	1.67	4.77	4.55	1.57	8.82

Effect of neutron spectra on dpa and fluences in WWER-440

Damage rates and neutron fluxes	R (cm)		
	160	178	192
10^{-11} $\frac{dpa}{s}$ for neutrons with E> 0.5 MeV (contribution to the total rate)	324 (78%)	19.7 (81%)	3.94 (68%)
10^{11} $\frac{fluence}{s}$ for neutrons with E> 0.5 MeV (contribution to the total flux)	44.5 (16.9%)	2.45 (12.4%)	0.665 (28.3%)
10^{-22} $\frac{dpa}{fluence}$ ratio of the total rate to the flux of neutrons with E> 0.5 MeV (with all energies)	9.28 (1.7)	9.92 (1.25)	8.88 (2.36)

BN-600 – the unique fast neutron breeder reactor

The only reactor of that type working as a commercial enterprise.



1, 2 – core, fuel assembly

3 – primary circuit circulating pump

4- heat exchanger of intermediate circuit

5 – central column

6 - scram system mechanism

7 – reloader

8 – neutron flux control channel

9 – neutron measuring chambers

10 – reactor support

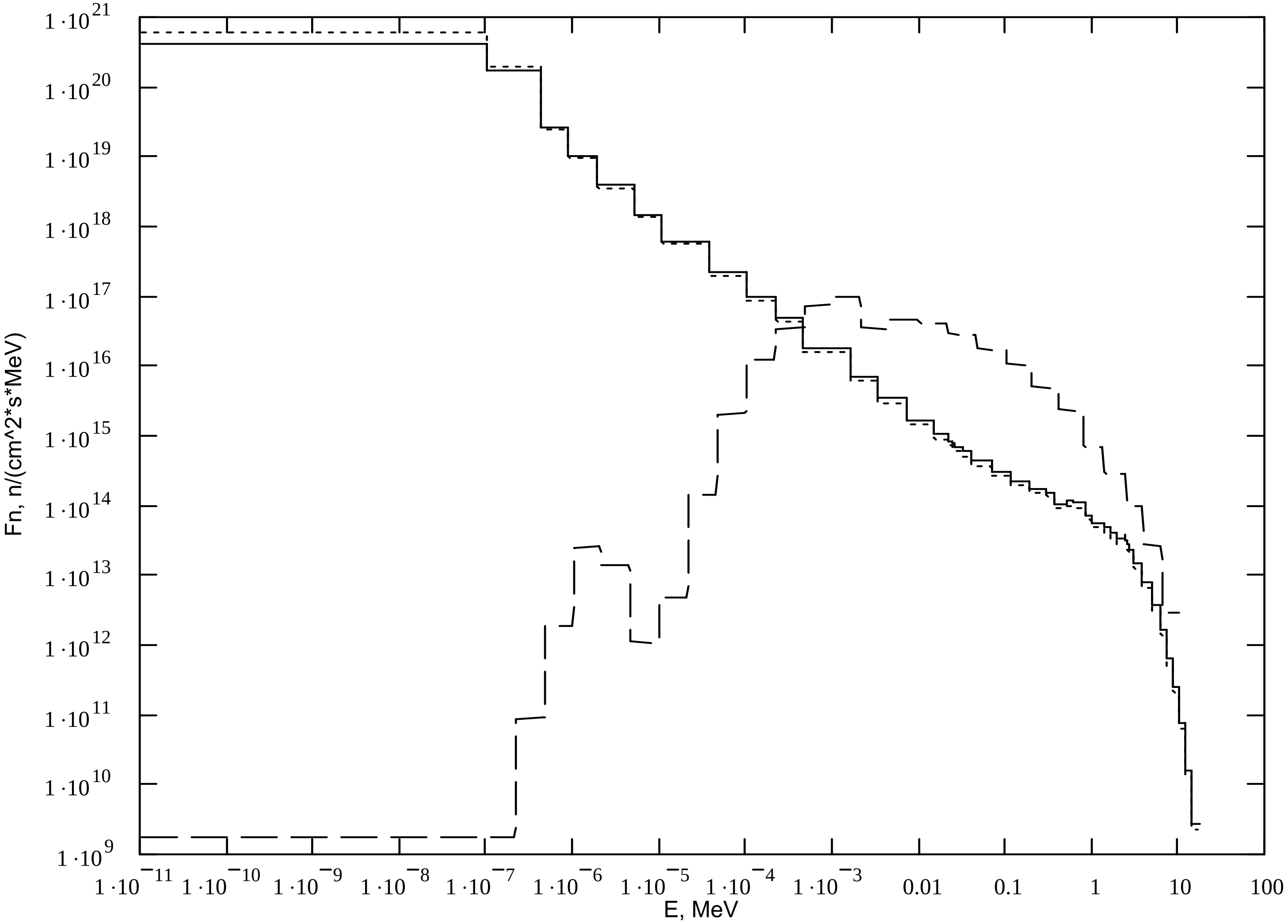
11- pit of reactor

14- rotating plug

15- neutron shield

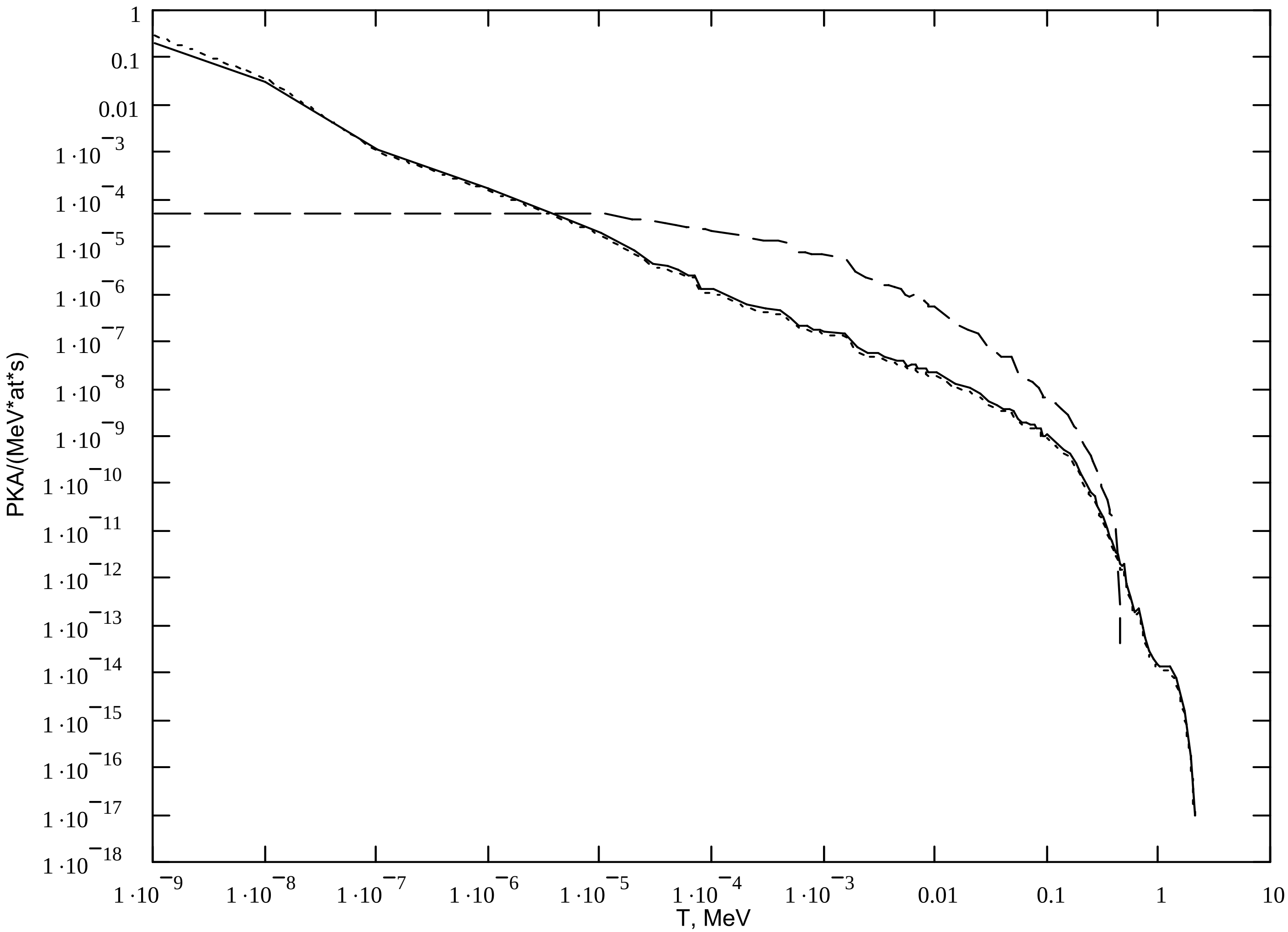
16- reloading cell

Neutron spectra



Neutron spectra in the cores of BN-600 (dashed line), VVER-440 (dotted line) and VVER-1000(solid line)

Recoil energy spectra



Recoil energy spectra in the cores of BN-600 (dashed line), VVER-440 (dotted line) and VVER-1000(solid line)

Dose rate in the core of BN-600

Metall	Dose rate, 10^{-8} dpa/s (dpa/year)			Mean energy of PKA (keV)
	v_{NRT} T_d = 40 eV (T_d^W = 90 eV)	v_{NRT} T_d = 30 eV (T_d^W = 90 eV)	v_{MD}	
Fe	176 (55.4)	234 (73.8)	59.2 (18.7)	7.23
Zr	204 (64.5)	273. (86.0)	66.6 (21.0)	4.44
Cu	178. (56.1)	237. (74.9)	57.6 (18.2)	5.22
Ti	189. (59.5)	251. (79.4)	88.7 (28.0)	3.70
W	44.9 (14.1)	44.9 (14.1)	19.7 (6.21)	2.47

Conclusions

- Fast neutron fluence is not a universal quantity for definition of the damage exposure in different reactor irradiation environments

- Displacements per atom (dpa) seems more universal tool for this purpose, as it takes into account all available neutrons and allows to compare results of irradiation with different types of particles (neutrons, ions, fast electrons)

- “Effective” dpa, which are introduced to take into account the point defect recombination during the relaxation stage of a displacement cascade, can be still better representation of the effect of irradiation on materials properties