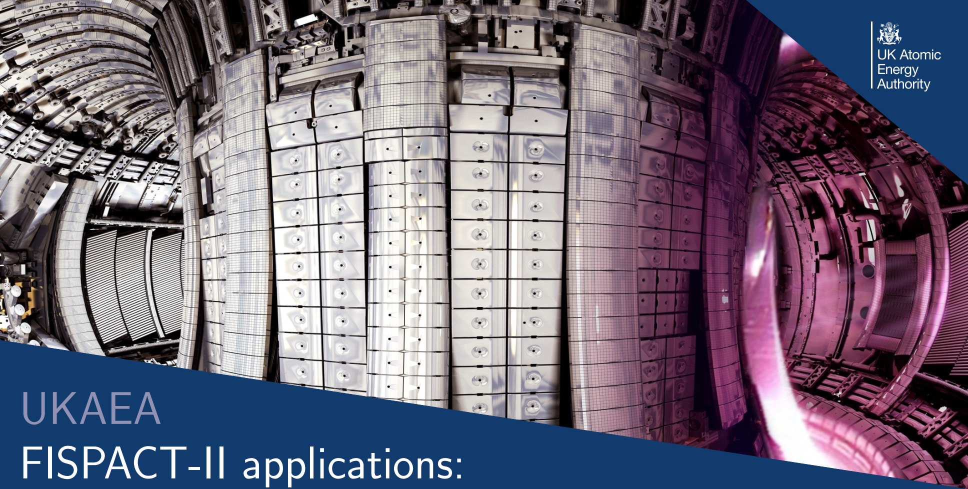

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FISPACT-II applications: materials modelling, scoping and damage metrics

Mark Gilbert
United Kingdom Atomic Energy Authority

FISPACT-II workshop

October 23-25, 2019, Manchester

This work was funded by the RCUK Energy Programme
[Grant number EP/P012450/1]

Energy For a Low Carbon Future

CCFE CULHAM CENTRE FOR FUSION ENERGY

Applications

- Integrated assessment of helium embrittlement
- Material response database
- Primary damage spectra (& emitted particle spectra)

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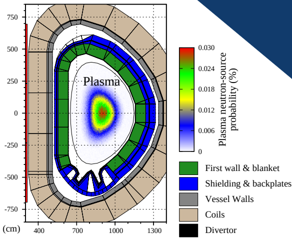
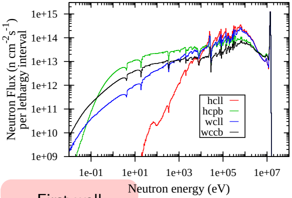
Integrated assessment: Outline

- Neutron transport modelling results for DEMO (MCNP)
- Neutron-induced transmutation (FISPACT-II)
  - helium production
- Helium embrittlement model and critical lifetimes



DEMO model with defined 3D plasma source

Gilbert, Dudarev, Nguyen-Manh, Zheng, Packer, Sublet
J. Nucl. Mater. **442** (2013) S755
Gilbert, Dudarev, Zheng, Packer, Sublet
Nucl. Fus. **52** (2012) 083019



First wall neutron spectra as a function of DEMO concept

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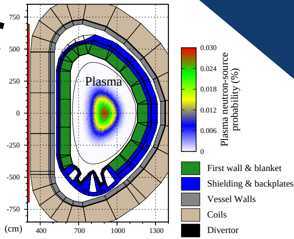

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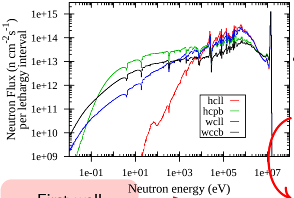

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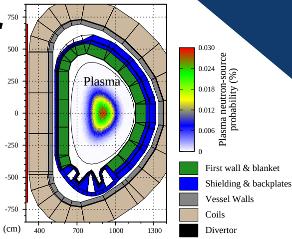

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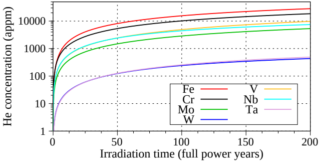
He production rates in materials



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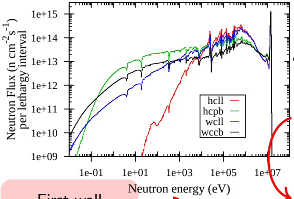

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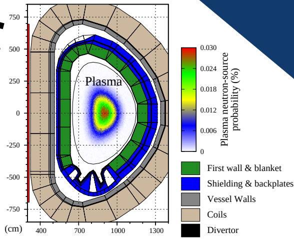

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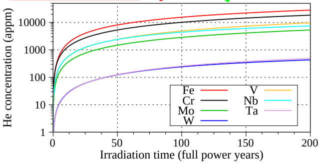

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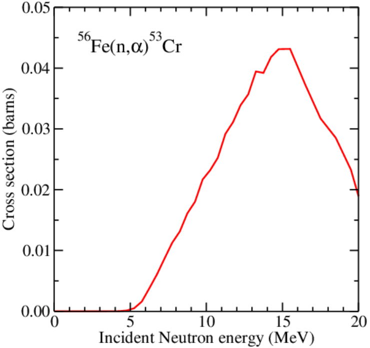
Helium production

- Helium production is a particular problem for fusion because of the generally higher neutron energies – many of the He producing reactions have thresholds
- Material comparison under identical DEMO hcpb conditions
- 3 fpy under outboard equatorial FW armour irradiation:



- FISPACT-II inventory calculations:
- Helium production highest in Be ($\sim$ 710 appm/fpy)
- More than an order of magnitude lower in Fe ($\sim$ 120 appm/fpy)
- Only $\sim$ 2 appm/fpy in W

appm=atomic parts per million; fpy - full power years

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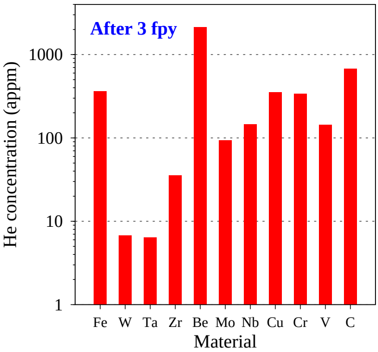

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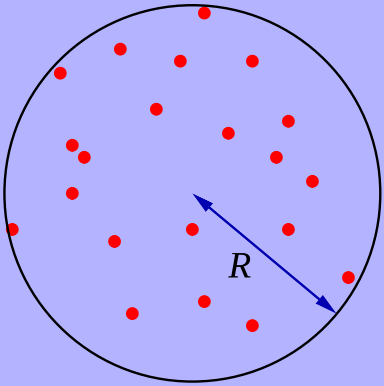
Simple modelling of grain boundary (GB) failure

1. Number of He atoms in spherical grain:

$$\boxed{N_{\mathrm{He}} \approx \frac{4}{3}\pi R^3 n G_{\mathrm{He}}}$$



Assumptions:

- All helium atoms produced migrate to grain boundary
  - traps neglected
  - most valid for small grains

G_{He} = bulk concentration (time evolution from inventory calcs.)

n = atom density

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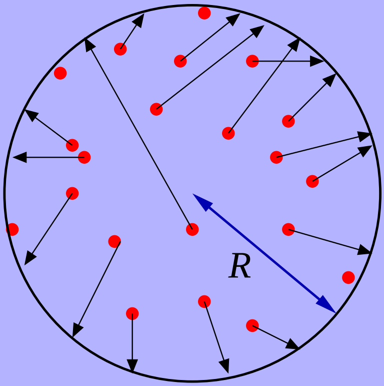
Simple modelling of grain boundary (GB) failure

2. All He atoms move to GB: – $\therefore$ surface total $\equiv$ bulk total

$$\boxed{4\pi R^2 \nu_{\mathrm{He}} = \frac{4}{3}\pi R^3 n G_{\mathrm{He}}} \quad \Rightarrow \quad \boxed{\nu_{\mathrm{He}} = \frac{R}{3} n G_{\mathrm{He}}}$$



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G_{He} = bulk concentration (time evolution from inventory calcs.)

ν_{He} = surface density

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Simple modelling of grain boundary (GB) failure

3. GB destabilize when E of inserted He equals E of surfaces:

$$\boxed{E_{\mathrm{He}}^{\mathrm{insrt}} \nu_{\mathrm{He}}^{c} \approx 2\varepsilon_{\mathrm{surf}}}$$

Material	He insert. energy (eV)† – $E_{\mathrm{He}}^{\mathrm{insrt}}$	Surf. energy $(\mathrm{Jm}^{-2})^{*}$ – $\varepsilon_{\mathrm{surf}}$	Critical He conc. at GBs (cm^{-2}) – ν_{He}^{c}
Fe	2.77	2.4	1.08×10^{15}
Cr	2.68	2.3	1.07×10^{15}
Mo	1.91	3.0	1.96×10^{15}
Nb	1.60	2.7	2.11×10^{15}
W	1.61	3.5	2.71×10^{15}
V	2.30	2.6	1.41×10^{15}

* Averages of experimental data reported in: L. Vitos *et al.*, 1998, *Surf. Sci.*, **411** 186–202

† DFT values – Gilbert *et al. J. Nucl. Mater.* **442** (2013) S755

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Modelling: He embrittlement of GBs

Simple modelling of grain boundary (GB) failure

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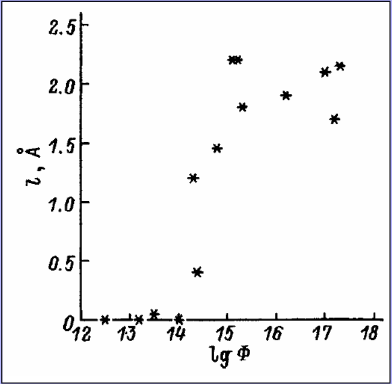


- Experimental confirmation:
  - Helium irradiated W bicrystals
  - Expansion of grain boundaries at He fluence of 10^{14}–10^{15} ions cm^{-2}
  - our ν_{He}^{c} value: 2.71×10^{15}

Gerasimenko, Mikhaĭlovskiĭ, Neklyudov, Parkhomenko, and Velikodnaya
Tech. Phys. **43** (1998) 803

Simple modelling of grain boundary (GB) failure

4. Critical bulk He concentration:

$$\boxed{G^c_{\mathrm{He}} = \frac{3}{Rn}\nu^c_{\mathrm{He}}}$$

Material	ν^c_{He} (cm^{-2})	n (cm^{-3})	G^c_{He} (appm)
Fe	1.08×10^{15}	8.5×10^{22}	764.6
Cr	1.07×10^{15}	6.4×10^{22}	771.9
Mo	1.96×10^{15}	5.5×10^{22}	1833.8
Nb	2.11×10^{15}	6.3×10^{22}	2275.2
W	2.71×10^{15}	1.2×10^{23}	2582.1
V	1.41×10^{15}	4.7×10^{22}	1172.2

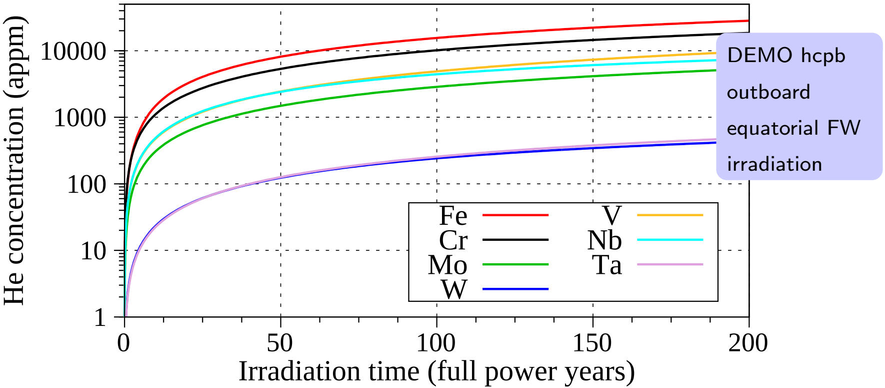
- Assumed Grain size of $R = 0.5\mu m$
- G^c_{He} varies with $1/R$

appm – atomic parts per million

FISPACT-II results: helium production rates

- standard output during irradiation in FISPACT-II simulation

- Estimated "time to destabilize" t^c_{He} based on inventory calculations



appm = atomic parts per million

He embrittlement: Critical times

- t^c_{He} for irradiations in outboard equatorial DEMO FW armour position assuming linear grain size of 0.5 μm

Element	ν^c_{He} (cm^{-2})	G^c_{He} (appm)	t^c_{He} (fpy)			
			hcll	hcpb	wcll	wccb
Fe	1.08×10^{15}	764.6	9	6	8	7
Cr	1.07×10^{15}	771.9	7	7	7	7
Mo	1.96×10^{15}	1833.8	58	63	59	62
Nb	2.11×10^{15}	2275.2	45	47	46	46
W	2.71×10^{15}	2582.1	700+	597	700+	700+
V	1.41×10^{15}	1172.2	25	25	25	25

- For some elements, such as W, the He production rates are so low, and/or the ν^c_{He} is so great that the critical lifetimes are many hundreds of years
- only for $t^c_{\mathrm{He}} \lessapprox 10$ is this failure mechanism likely to be of concern

appm = atomic parts per million, fpy = full power year

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Applications

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- Primary damage spectra (& emitted particle spectra)

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Application: material response database

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CCFE-R(16)36
August 2016

Mark R. Gilbert
Jean-Christophe Sublet

Handbook of activation, transmutation, and radiation damage properties of the elements simulated using FISPACT-II & TENDL-2015; Magnetic Fusion Plants

785 pages
∼60000 calculations

www.ccfe.ac.uk

- Reference document of typical activity and burn-up responses of the elements to neutron irradiation is a useful tool during material selection exercises for nuclear components
- Results for all naturally occurring elements from H to Bi, with separate reports for:
  - predicted DEMO and ITER conditions
  - fission conditions (PWR, FBR, HFR)
- Available to download from http://fispact.ukaea.uk

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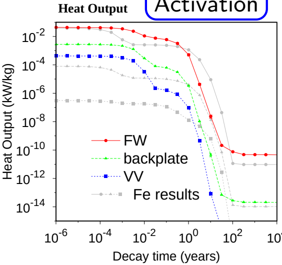
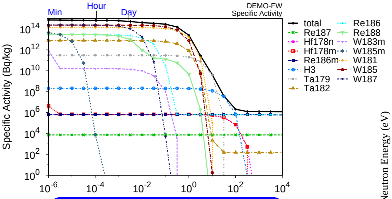
Scoping calculations for each element:

1. Tabulated activation response
   - % contributions of important radionuclides to various radiological quantities as a function of cooling time after irradiation
2. Graphs of activation response after irradiation
   - decay evolution of total activity, heat, and γ-dose under three different spectra & compared to Fe + indicative dominant nuclide contributions
   - nuclide contributions as a function of time
3. Importance diagrams
   - spectrum independent mapping of important radionuclides in the neutron-energy vs. decay time phase-space
4. Transmutation response
   - time evolution under irradiation of initially-pure elemental composition
   - nuclide concentration maps
5. Primary knock-on atom (PKA) distributions
   - $t_{irr} = 0$ spectra plotted as both elemental and isotopic sums
6. Reaction pathways
   - major production pathways for important radionuclides at four characteristic neutron-energy ranges

Example outputs: W

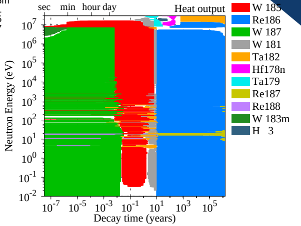
Activation



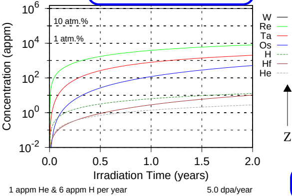


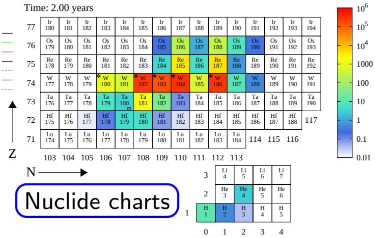
Nuclide contributions

Importance diagrams



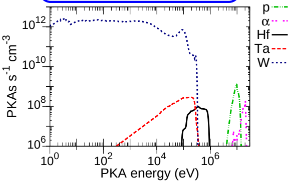
Transmutation





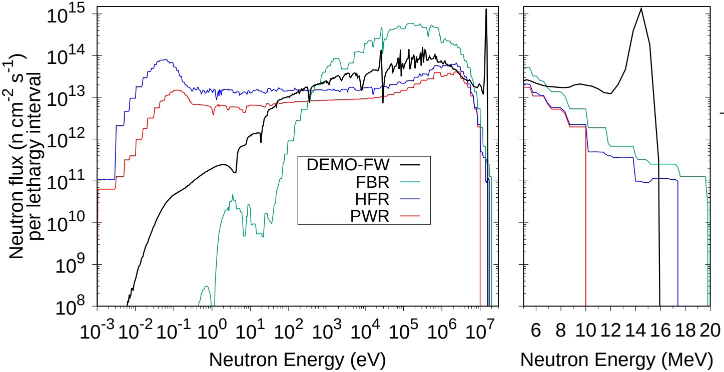
Nuclide charts

PKA distributions



Neutron irradiation fields

- Monte Carlo simulations of a fusion DEMOnstration power plant & typical fission reactors



total fluxes ϕ:
($\times 10^{14}$ n cm^{-2} s^{-1})

DEMO	6
FBR	24
HFR	5
PWR	3

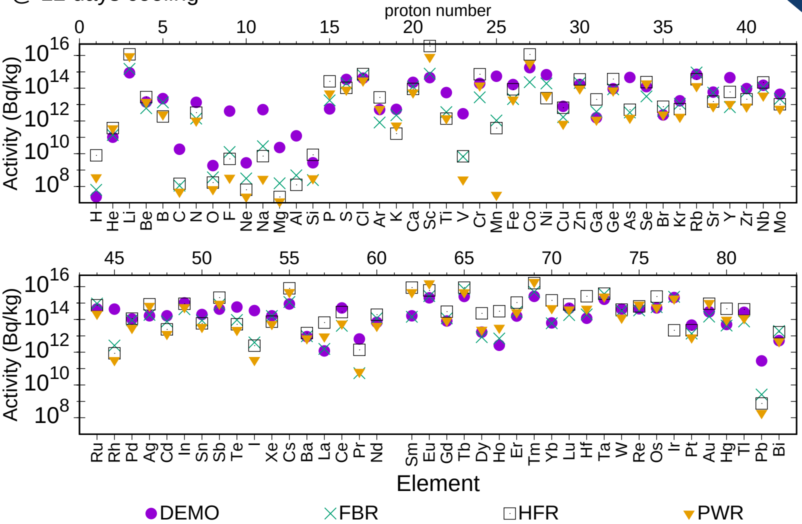
- fusion spectrum in first wall (FW) dominated by 14 MeV peak
- well-moderated (averaged) fission spectra don't have such dominant peaks but can have tails that explore the 14 MeV region of fusion

FBR – superphenix Fast Breeder Reactor
HFR – High Flux Reactor, Petten
PWR – Pressurized Water-cooled Reactor

Activation – fusion vs. fission

- time snapshot of activity during decay cooling§:

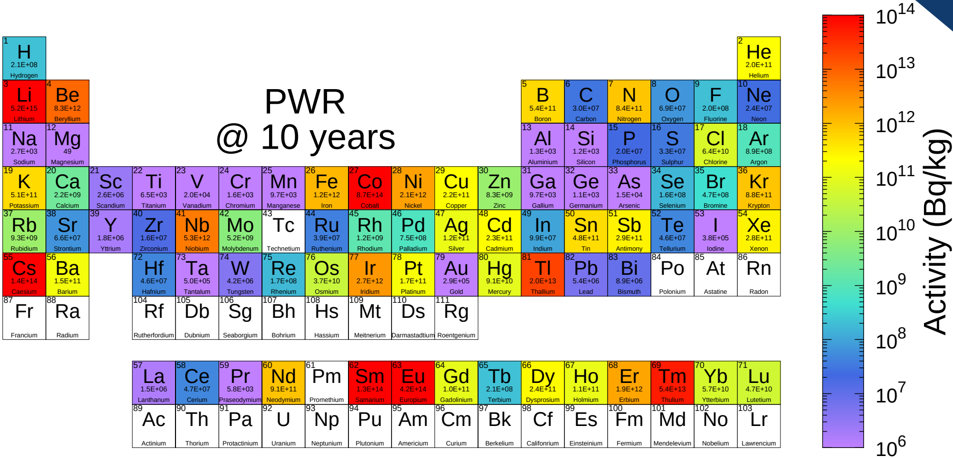
@ 12 days cooling



§after 2 years of continuous irradiation

Activation summary – periodic table plot

- total becquerel/kg activity from each element at 10-years following a 2-year PWR irradiation:



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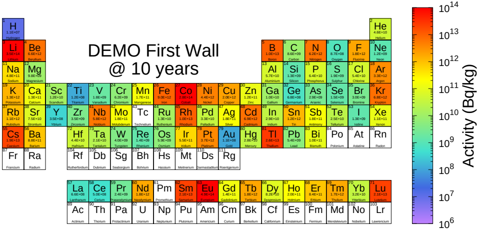
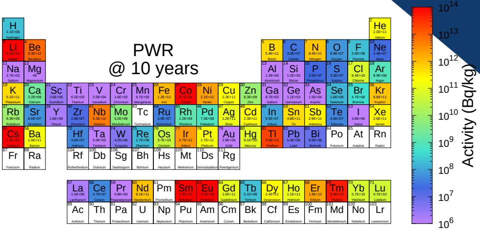
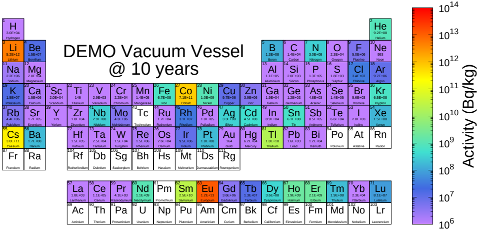
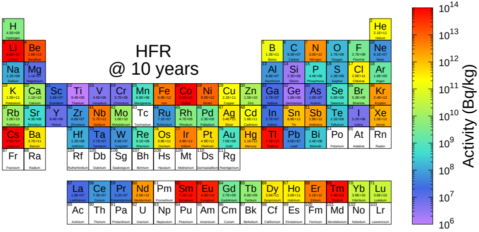


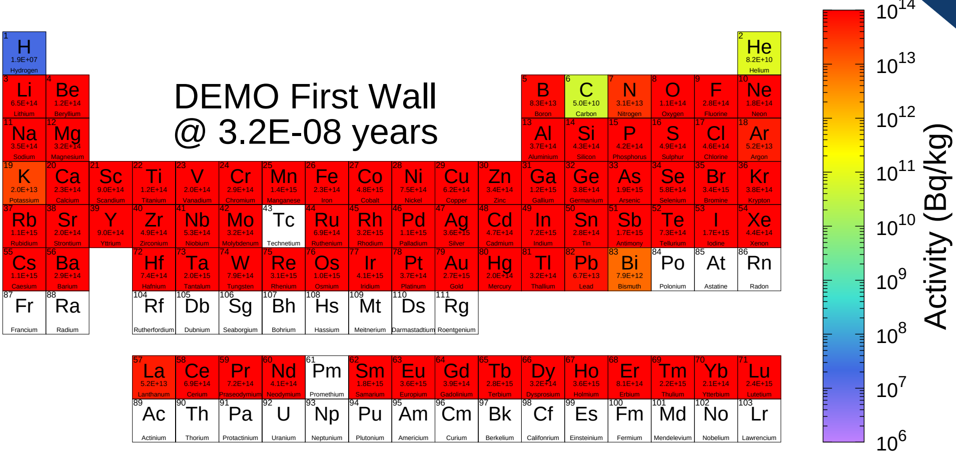






Activation summary – animated

- total becquerel/kg activity from each element as a function of time following 2-year irradiation



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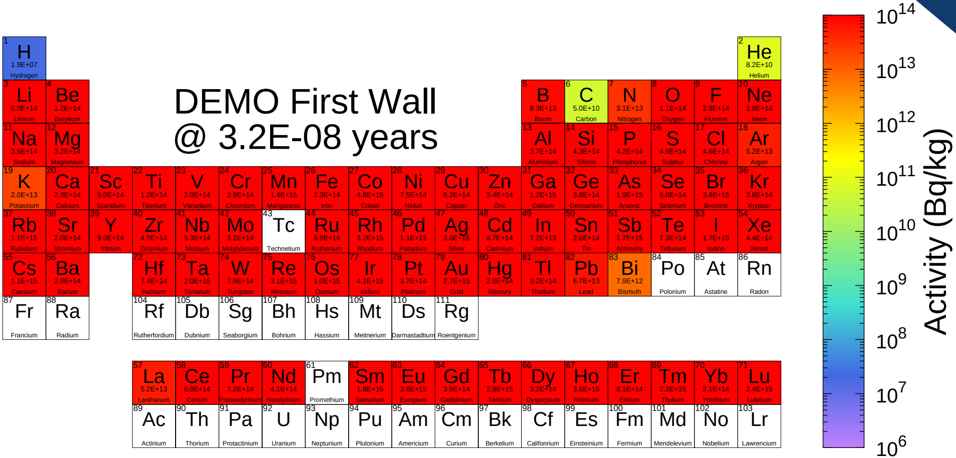
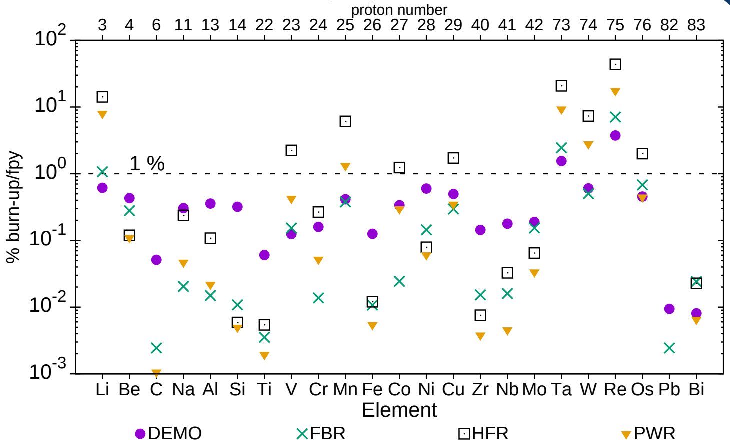


Transmutation – fusion vs. fission

- % burn-up per full-power year (fpy) for selected elements:



- Fission spectra are more-moderated ("softer") than fusion & combined with high-flux in HFR produces greater burn-up rates in many elements[§] ⇒ spectrum modification (via shielding) required to match fusion in experimental campaigns

[§] due to higher neutron capture rates

Gas production – fusion vs. fission

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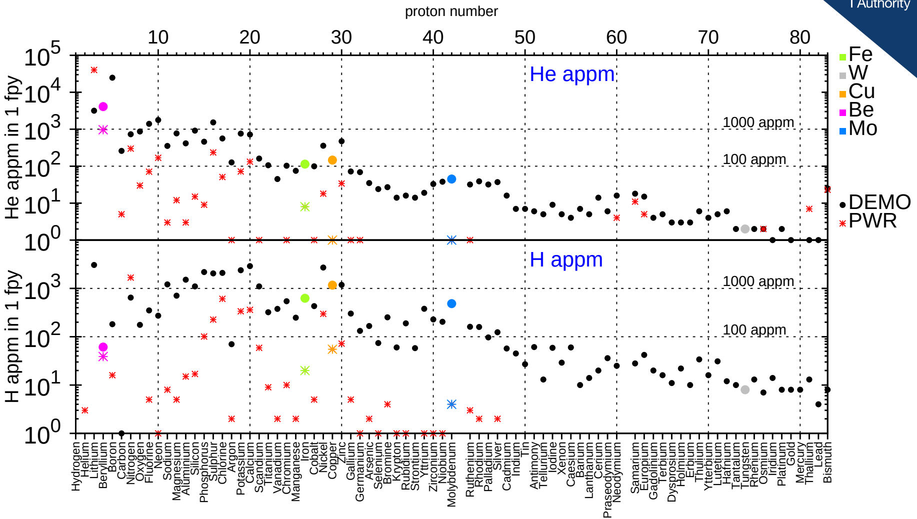
- Gas appm after 1 fpy in DEMO first wall and PWR:



- gas production is significantly higher under fusion neutrons
  - ▶

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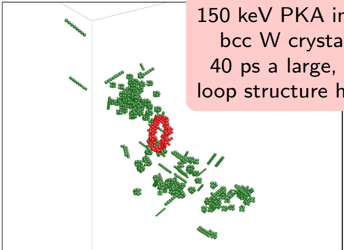
Applications

- Integrated assessment of helium embrittlement
- Material response database
- Primary damage spectra (& emitted particle spectra)

Application: PKA distributions

- Motivation: a full description of the initial damage events (the primary knock-on atoms or PKAs) under irradiation is a necessary input to modelling and simulation of radiation damage
  - PKA energy.vs.flux distributions provide more information than simpler damage-dose measures (e.g. dpa)

- Complete evaluation requires all possible nuclear reaction channels to be considered:
  - elastic & inelastic scattering, and non-elastic reactions ($(n,p),(n,2n)$, etc.)
  - including secondary emitted (light) gas particles - α (^{4}He), protons (^{1}H), etc, as PKAs



(A.E. Sand, Helsinki – 2012)

150 keV PKA in a perfect bcc W crystal - after 40 ps a large, coherent loop structure has formed

PKA – primary knock-on atom

Additional need

- emitted particle spectra for α, protons, neutrons, γ, etc.
- could be used in FISPACT-II to compute transmutation from secondary particles

PKA – primary knock-on atom

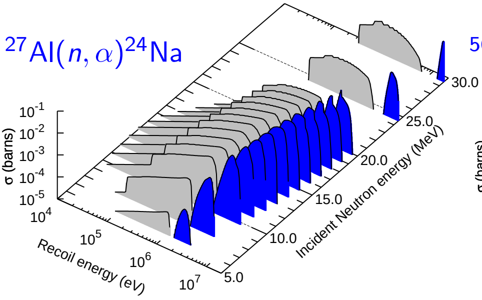
PKA distributions

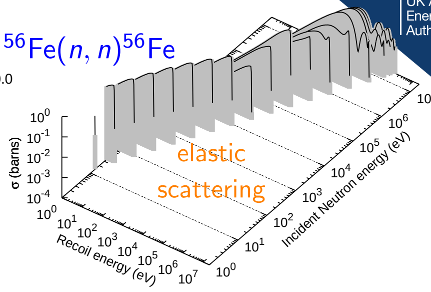
Calculation method (1)

- From raw pointwise nuclear data (TENDL)
- Neutron interaction recoil matrices $M^{x\to y} \equiv \{m_{ij}^{x\to y}\}$ calculated using NJOY[§] (via GROUPR)
  - $m_{ij}^{x\to y}$ is the recoil cross section (in barns) for a recoil energy E_i of daughter y resulting from an incident neutron energy E_j on parent x
- for each target (parent) isotope x there will be a set of $M^{x\to y}$
  - (at least) one for each reaction channel
- & each reaction may have more than one associated matrix
  - for example, (n,α) will produce a heavy recoil and an α-particle (^{4}He nucleus)
- recoil matrices can also be produced for charged particle interactions

[§] the nuclear data processing system developed at LANL

$M^{x \to y}$ matrices & vectors

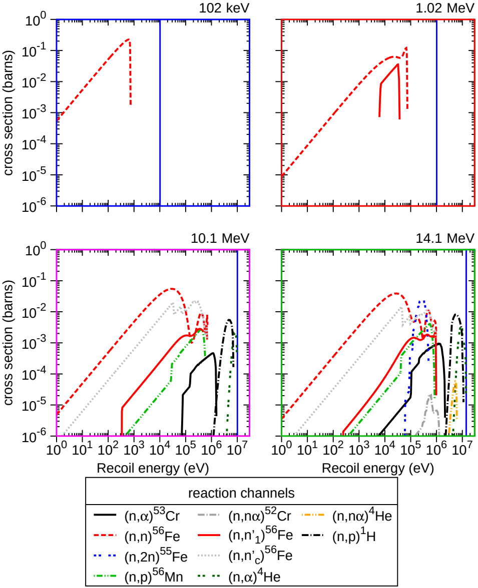


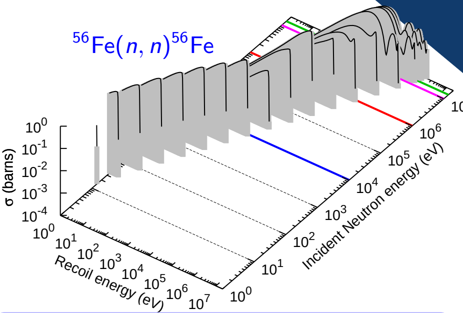


- Non-elastic nuclear reactions can produce two recoiling species
- The main (heavy) residual and a higher energy light (gas) particle
- On ^{27}Al the threshold (n,α) reaction produces ^{24}Na (grey) recoils and ^{4}He particles (blue)

- The primary reaction channel is normally simple elastic scattering
- Produces recoils for all incident neutron energies
- Other channels only become important at higher energy...

$M^{x \to y}$ matrices & vectors – ^{56}Fe





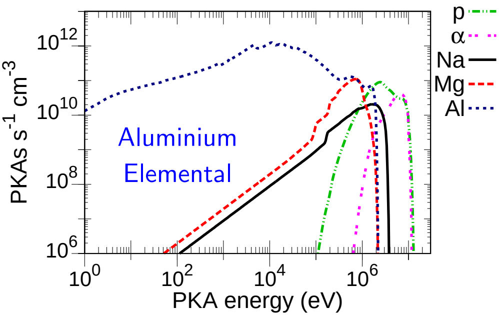
- In ^{56}Fe, at low incident energy only elastic and inelastic channels are open
- Number of contributing channels increases at higher energies (MeV range)

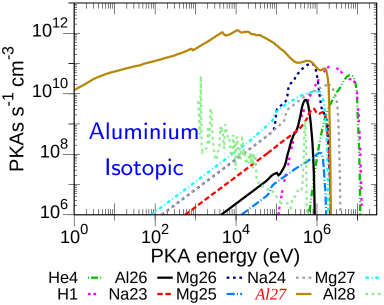
Calculation method (2)

- Collapsing each recoil matrix with a neutron irradiation spectrum $\{\phi_j\}$ gives the recoil-energy spectrum $R^{x\to y}(E)$:

“PKA-spectrum” under neutron irradiation $\equiv R^{x\to y}(E) \equiv \{r_i^{x\to y}\} = \left\{ \sum_j m_{ij}^{x\to y}\phi_j \right\}$

- processing (& summing) done with `SPECTRA-PKA`[§] utility





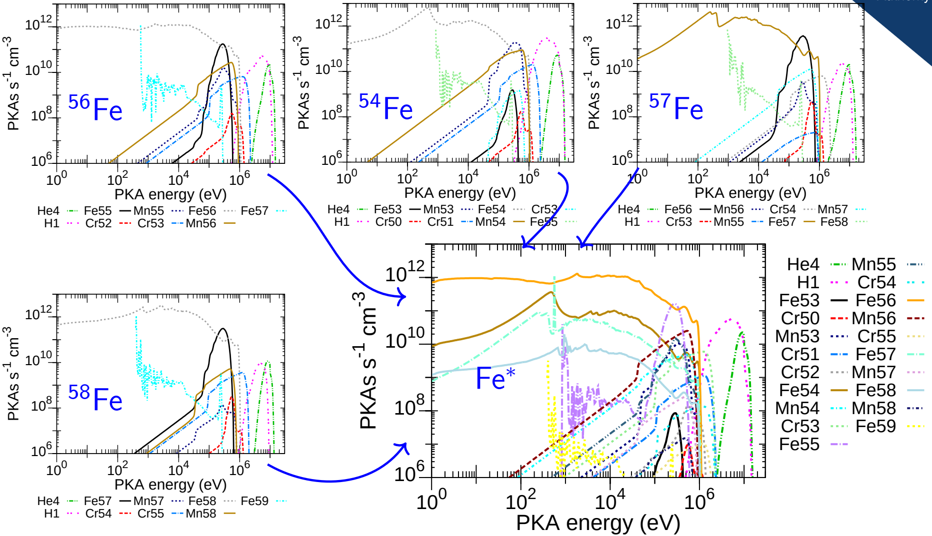
- PKA spectra of pure Al under DEMO conditions
- many different recoil species (even with just Al27 target)

§ Gilbert *et al.*, *JNM* **467** (2015) 121-134

PKA results – e.g. Fe

UK Atomic Energy Authority

- Isotopic results for each nuclide in the material must be merged:

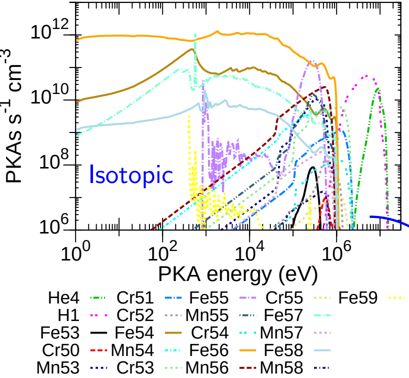


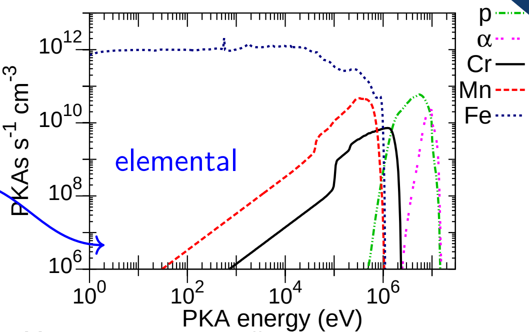
*5.845% ^{54}Fe, 91.754% ^{56}Fe, 2.119% ^{57}Fe, 0.282% ^{58}Fe (atomic percentages)

CCFE CULHAM CENTRE FOR FUSION ENERGY

PKA results – e.g. Fe

- For modelling it is more useful to sum the isotopic spectra:

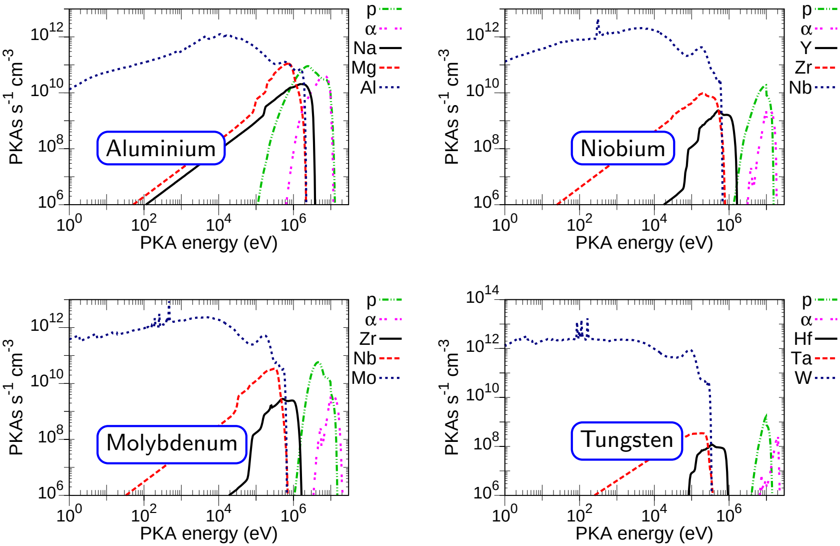




- Main constituent usually dominates

- Total PKAs (s^{-1}cm^{-3}) in material can be calculated by summing (& integrating over) all elemental distributions
- here there are 4.33E+14 PKAs s^{-1}cm^{-3} above 1eV (light particles excluded)
- the average PKA energy above 10 eV for the Fe+Mn+Cr spectrum is 18.8 keV

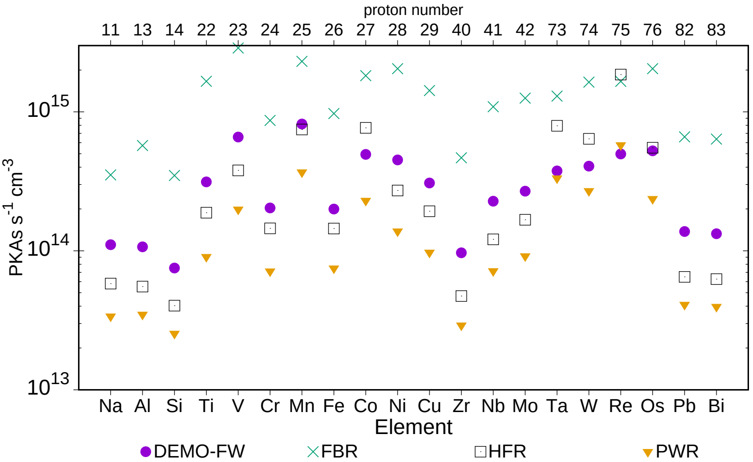
PKA distributions – elemental



- Under DEMO FW conditions

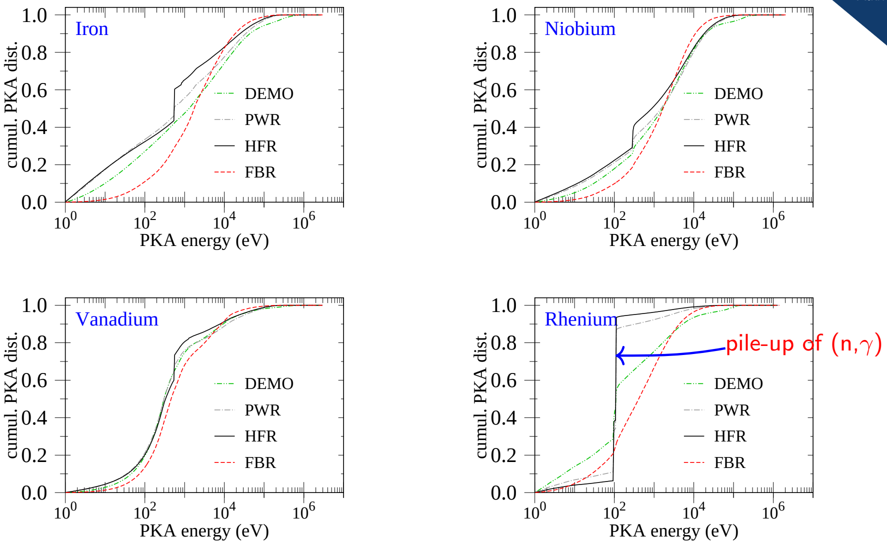
Total PKA rates – fusion vs. fission

- total heavy PKAs (H/He excluded) for selected materials



- FBR spectrum produces highest rates due to high, fast flux
- PWR the lowest

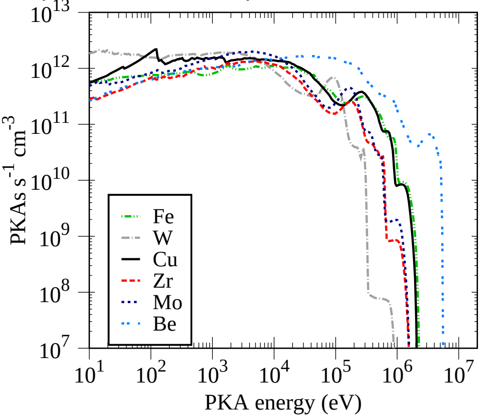
Cumulative (PKA) probability distributions



- FPR missing low and high energy parts due to peaked spectrum

(mis)interpretation?

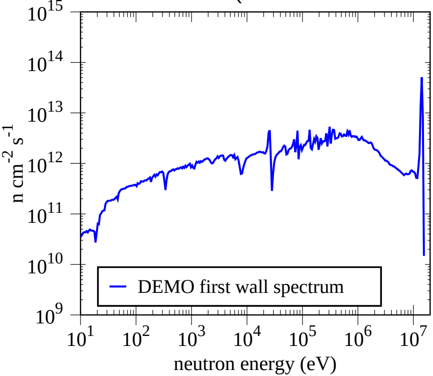
- Typical results from SPECTRA-PKA
- total PKAs in pure elements exposed to DEMO first wall neutrons

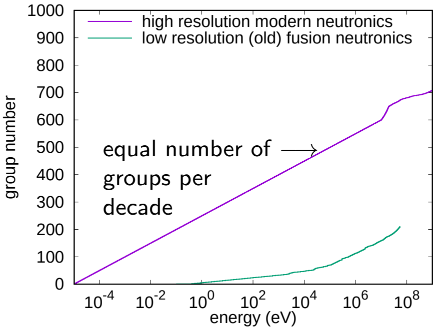


- Perhaps gives the impression that there are a relatively constant (flat) PKA fluxes at the majority of PKA-energies in most materials

Scope for misinterpretation

- <u>But</u>: these PKA flux spectra are mapped onto the same energy grid as that used for the neutron transport calculations (and hence nuclear data libraries)

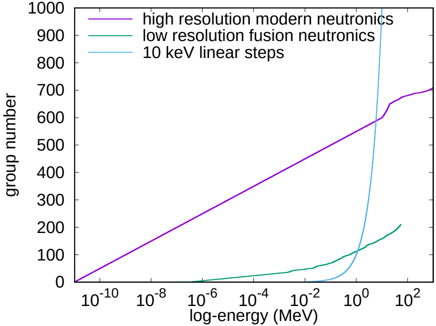


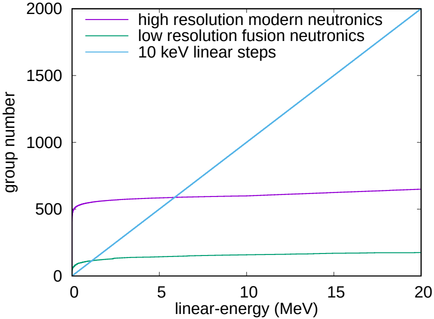


- Such grids are designed to give equal representation to low (thermal∼eV) and high (fast∼>MeV) neutron energies
- ⇒ not appropriate for representation of a structural damage metric like PKA spectra

Alternative

- For this reason part of the recent developments in SPECTRA-PKA have included the ability to (user-)define an output energy grid more relevant to damage modelling

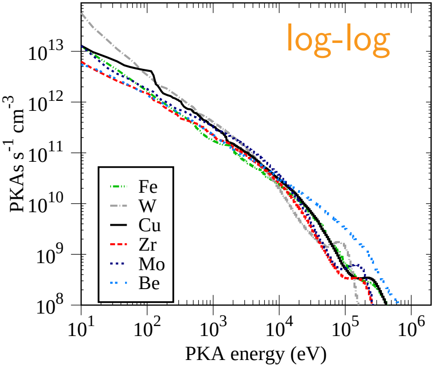


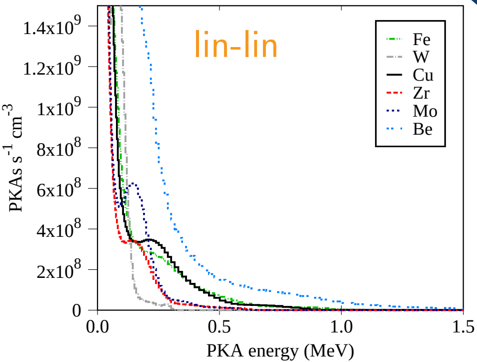


- E.g. an energy grid with equal bins (groups) of 10 keV in width
  - this is linear on a linear energy axis (and exponential on a logarithmic one)

Implications for interpretation

- Returning to the first wall DEMO example:
  - and using a very fine linear grid spacing of 10 eV

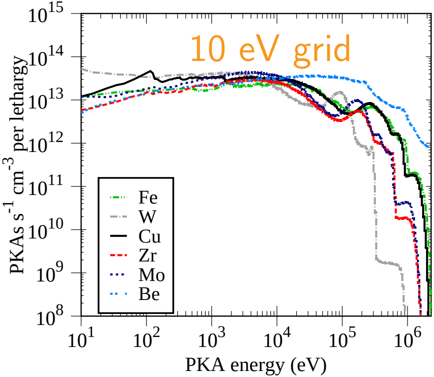


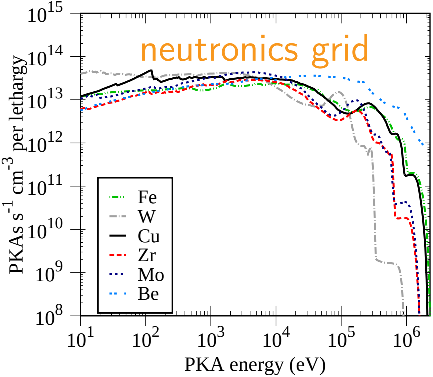


- on a log-log scale the profiles have a power-law appearance
  - the profiles are similar regardless of target mass
- & in linear energy versus PKA-flux there is an exponential decay

Cross-check

- No fundamental change in results – just re-binning
- Only the visualization is different
- To check use the standard (neutronics) approach of dividing by lethargy to smooth variations in grid





- lethargy is natural log of ratio of upper to lower bin energy

Damage energy & dpa from SPECTRA-PKA

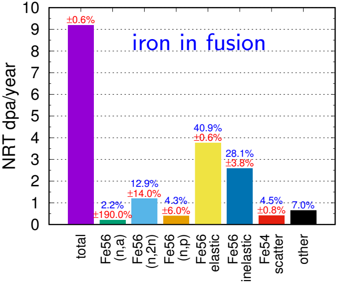
- The displacement energy in the NRT-dpa formula is normally calculated using the total damage kerma cross section
- However, with SPECTRA-PKA, it is possible to calculate the damage contribution as a function of reaction channel
  – a new & novel capability
- Standard LSS§ formula to account for electronic loss and convert PKA energy into damage energy – including correct treatment of parent and daughter mass
- displacement-energy rate accumulated and summed using PKA rate at each damage energy. NRT formula can be applied to the total
- For a given reaction channel, the total displacement energy is:

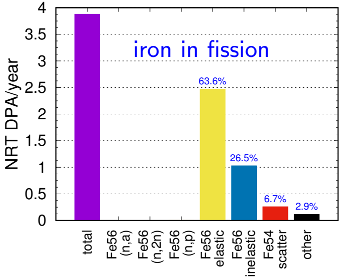
$$\sum_i T^{LSS}(E_i^{pka})R_i^{pka},$$

where $T^{LSS}(E^{pka})$ is the LSS equivalent energy for PKA energy E^{pka}, and R_i^{pka} is number of PKAs at energy E_i^{pka}

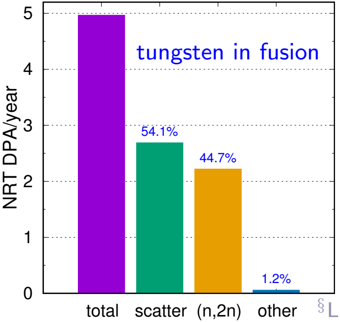
§Lindhard, Scharff, Schiøtt, *Mat. Fys. Medd. Dan. Vid. Selsk.* **33** (1963) 1–42

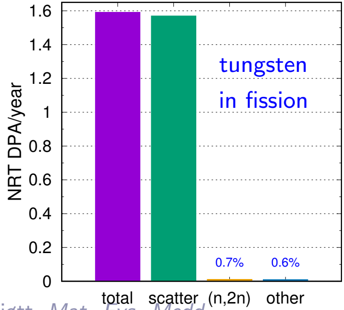
dpa evaluations with SPECTRA-PKA





The per-reaction-channel approach of SPECTRA-PKA can also be applied to displacements per atom (dpa) evaluations to produce new insight





§Lindhard, Scharff, Schiøtt, *Mat. Fys. Medd. Dan. Vid. Selsk.* 33 (1963) 1-42

J. Nucl. Mater. **504** (2018) 101-108

Running SPECTRA-PKA

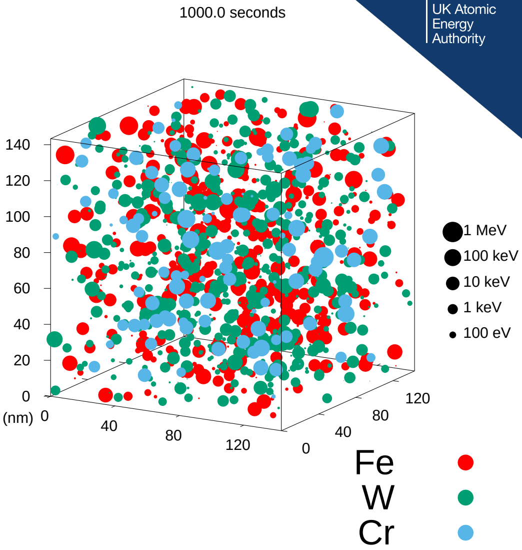
- SPECTRA-PKA is now open-source and downloadable from: https://github.com/fispact/SPECTRA-PKA
- code is command-line driven
- execution controlled by an input file of code words and values (similar to FISPACT-II, but order does not matter)

  `<location of SPECTRA-PKA>SPECTRA-PKA input.in`
- manual on github repository explains use of all code-words & there is an example provided...
  - including example plotting script to visualize results (with gnuplot)
- ...this is a good place to start
  - & example input can be easily modified to a specific material/spectrum of interest
  - if not already compiled on your system, the executable for SPECTRA-PKA can be easily created
    - navigate to fispact/source/spectra-pka and type `make`

Gilbert and Sublet, JNM **504** (2018) 101-108

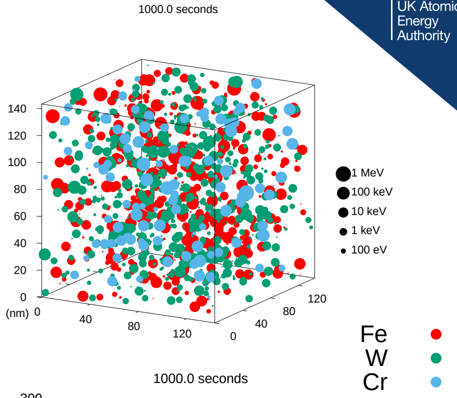
Further input to damage modelling

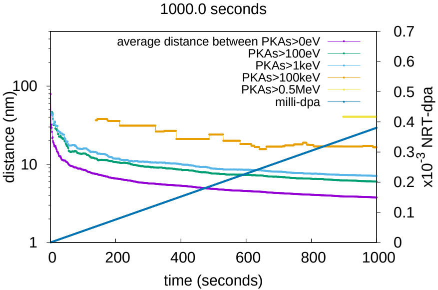
- Stochastic sampling of PKA distributions to define separation in time and space of PKA events in a given volume
  - prototyping for future `SPECTRA-PKA` development
- Example:
  - lattice of Fe-25%Cr-25%W (randomly distributed)
  - fusion first-wall neutron flux
  - PKAs introduced in 1 second timesteps in a square box of size ~140 nm^3 (equivalent to 250M atoms)

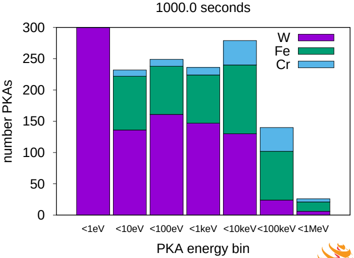


Statistics

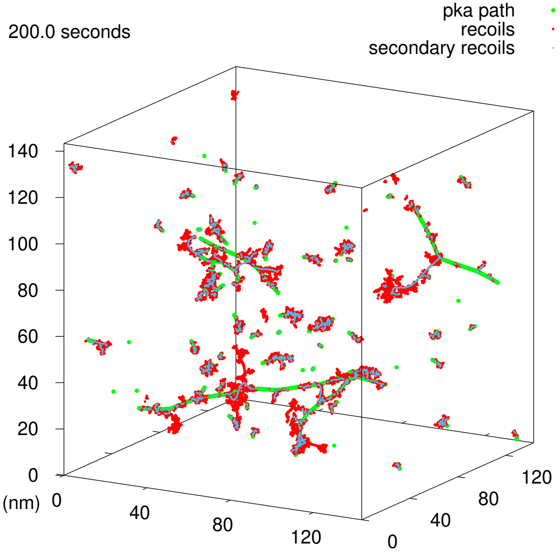
- Fe+Cr majority of higher energy PKAs, W predominant at energies below displacement thresholds
- Future: compare to cascade sizes to understand overlap thresholds, etc.
  - integrated modelling







Prototype integration



- e.g. using a binary collision approximation (BCA) code to define the PKA trajectories and recoils
  - same FeCrW system
  - each PKA given random initial direction