FISPACT-II

Neutron-induced damage simulations

Beyond Defect Production Cross-section, Displacement per Atom and Iron-based Metrics

Applications: material sciences

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Background

- Nuclear interactions can be the source of atomic displacement, embrittlement and post-short-term cascade annealing defects in irradiated structural materials
- Such metrics are derived from, or can be correlated to, nuclear kinematic simulations of primary atomic energy distributions spectra and the quantification of the numbers of secondary defects produced per primary as a function of the available recoils, residual and emitted, energies and spectra.
- Recoils kinematics of neutral, residual, charged now including multi-particle emissions occurring at high MeV range energy are more rigorously treated when based on modern, complete and enhanced nuclear data

Background

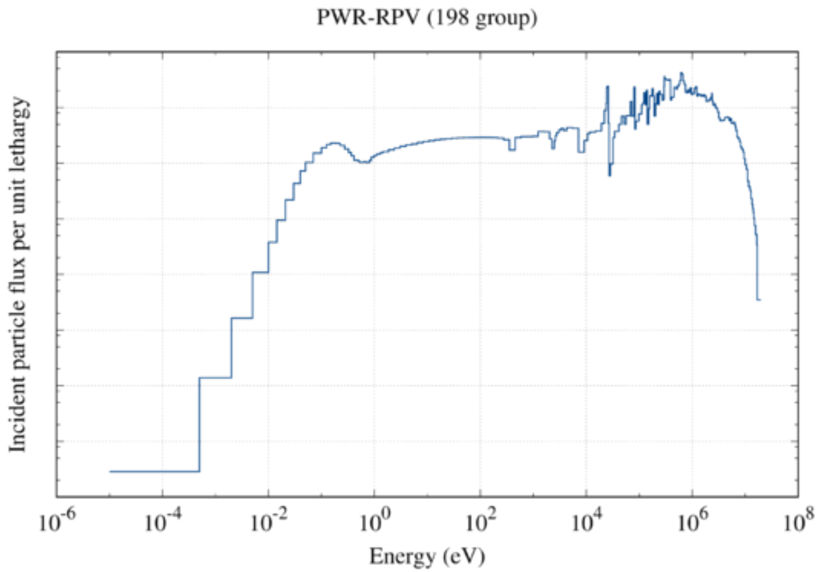
- A starting point in the complex problem of correlating and predicting the irradiation behavior of materials is calculation of Defect Production Cross Sections (DPCS) as a function of neutron/particle energy

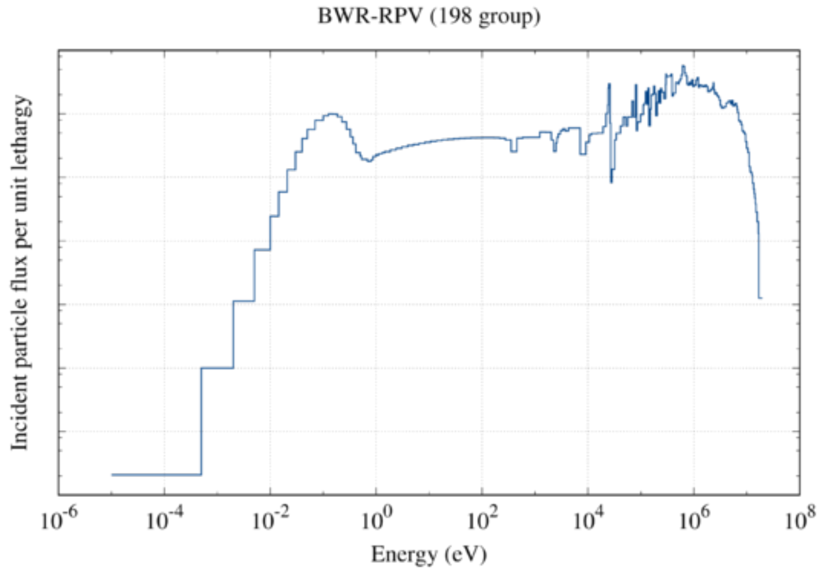
- The most common DPCS is for displacement of atoms. Displacement-Per-Atom irradiation exposure units are widely used for correlating neutron data and as partial basis for neutron-charge-particle inter-comparison.

- Although not an actual defect, the total kinetic energy, T_{dam} imparted to recoils atoms as a function of PKA energy is used as a damage exposure index. The remaining PKA's is dissipated to electrons by excitation and ionization

- Norgett-Robinson-Torrens NRT-dpa assumes a threshold energy E_d, with a probability = 0 below E_d and 1 above

The beginning: background ¼ RPV reference





Assuming a 40-year plant operation and a load factor of 0.8, giving an effective lifetime of 32 years, FISPACT-**II results**

PWR 1.3E-3 x 32 = **0.0416** dpa, total fluence 1.136E+20, > 1 MeV 2.387E+19 n/cm^2
BWR 2.3E-4 x 32 = 0.0073 dpa, total fluence 1.798E+19, > 1 MeV 4.469E+18 n/cm^2

An end-of-life fluence value of 3.0E+**19** n/cm^2, is quoted to produces about 0.045+/-0.05 dpa in *G. R. Odette and G. E Lucas, Embrittlement of Nuclear Reactor Pressure Vessels, JOM, 53 (7) (2001), pp 18-22*

FISPACT-II & Kerma/DPA responses: https://fispact.ukaea.uk/

UK Atomic Energy Authority

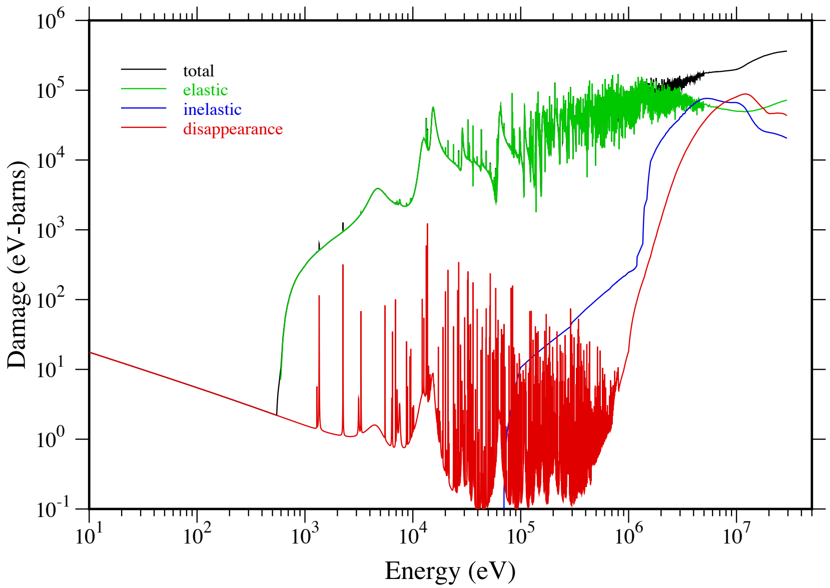
Processing protocols

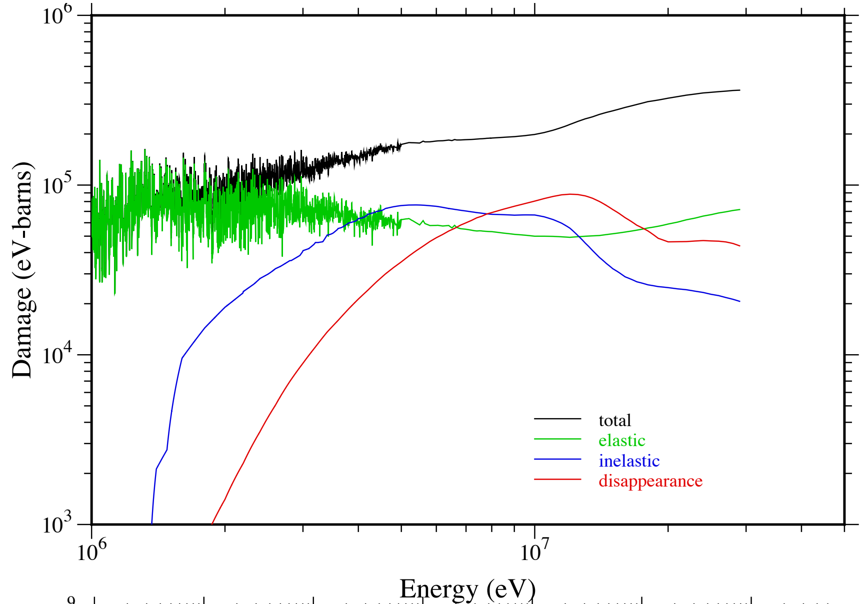
- Using the HEATR, GASPR and GROUPR module protocols of the most recent NJOY2016 release https://www.njoy21.io/
- Novel data forms for the 83 naturally occurring elements (assembled from their isotopic parts that in fine will compose the alloy) that include
  - total and partial neutron induced defect production
  - gas production cross section
  - kerma factors
- Detailed residual nucleus (A > 4) and emitted particle matrices: energy-angle distribution
- Have been systematically and uniformly derived from the latest ENDF/B-VIII.0, JENDL-4.0 and TENDL-2019

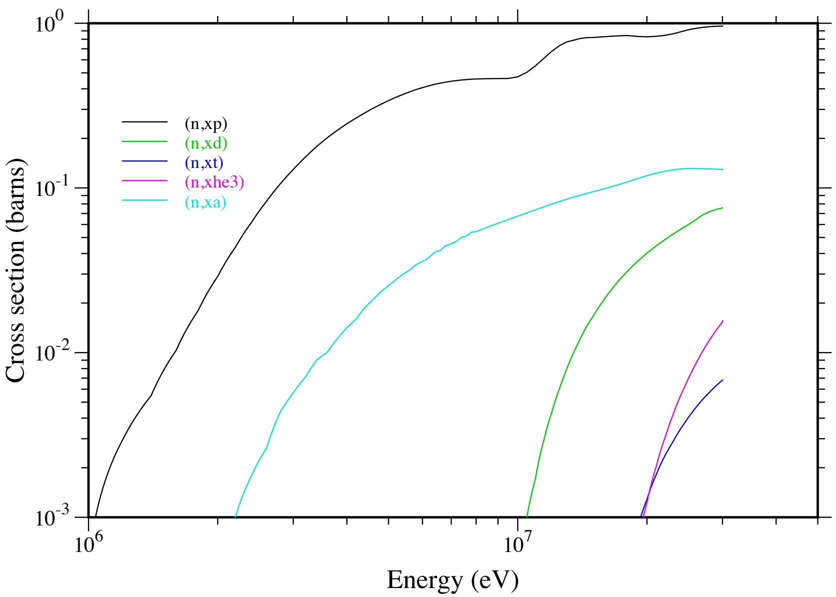
“Neutron-induced damage simulations: Beyond defect production cross-section, displacement per atom and iron-based metrics” Review in Eur. Phys. J. Plus (2019) 134: 350

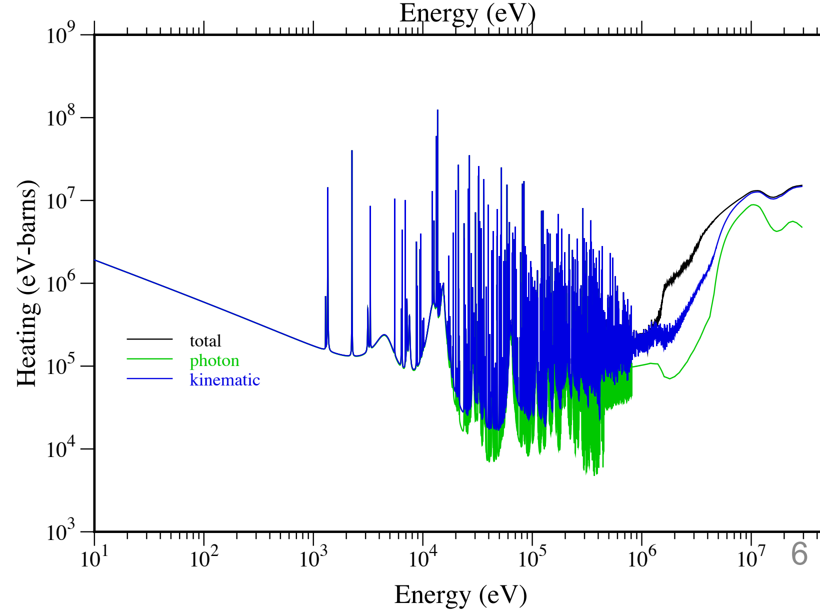
Processing protocols

- Kerma, Damage Energy, Gas Production (Ni)









Recap: Applications forms

- Pointwise forms, Temperature dependent
- 1102 Groupwise, T and sigma zeroes dependent, PT's SSF
- Variance and covariance (on cross section)
- MF-2 resonance widths for shielding
- ….
- **Matrices: n-n, n-g, n-prod and recoil**
- **All partials and total Kerma (7),**
- **Dpa (4) and gas production (5)**

MT	Description
201	(z,Xn) Total neutron production
202	(z,Xγ) Total gamma production
203	(z,Xp) Total proton production
204	(z,Xd) Total deuteron production
205	(z,Xt) Total triton production
206	(z,Xh) Total helion (^{3}He) production
207	(z,Xα) Total alpha particle production
301	Kerma total (eV-barns)
302	Kerma elastic
303	Kerma non-elastic (all but MT=2)
304	Kerma inelastic (MT={51-91})
318	Kerma fission (MT=18 or MT= {19, 20, 21, 38})
401	Kerma disappearance (MT={102-120})
402	Kerma for radiative capture
403	Kerma for proton emission
407	Kerma for alpha emission
442	Total photon (eV-barns)
443	Total kinematic kerma (high limit)
444	Dpa total (eV-barns)
445	Dpa elastic (MT=2)
446	Dpa inelastic (MT={51-91})
447	Dpa disappearance (MT={102-120})

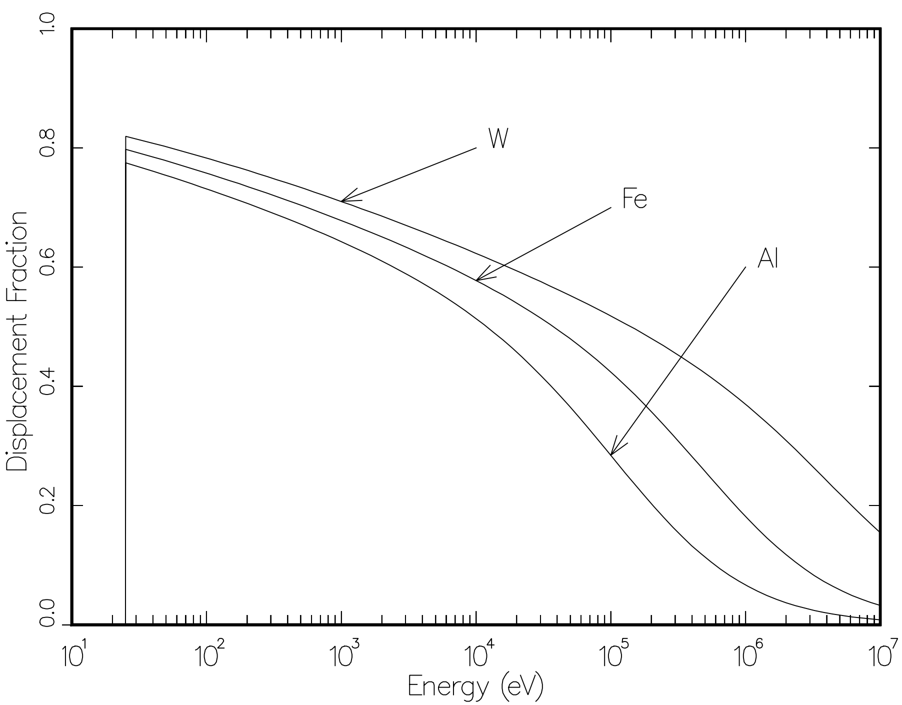
- For FISPACT-II & SPECTRA-PKA nuclear data forms, groupwise with probability tables, uncertainty, n-prod/recoil matrices, responses

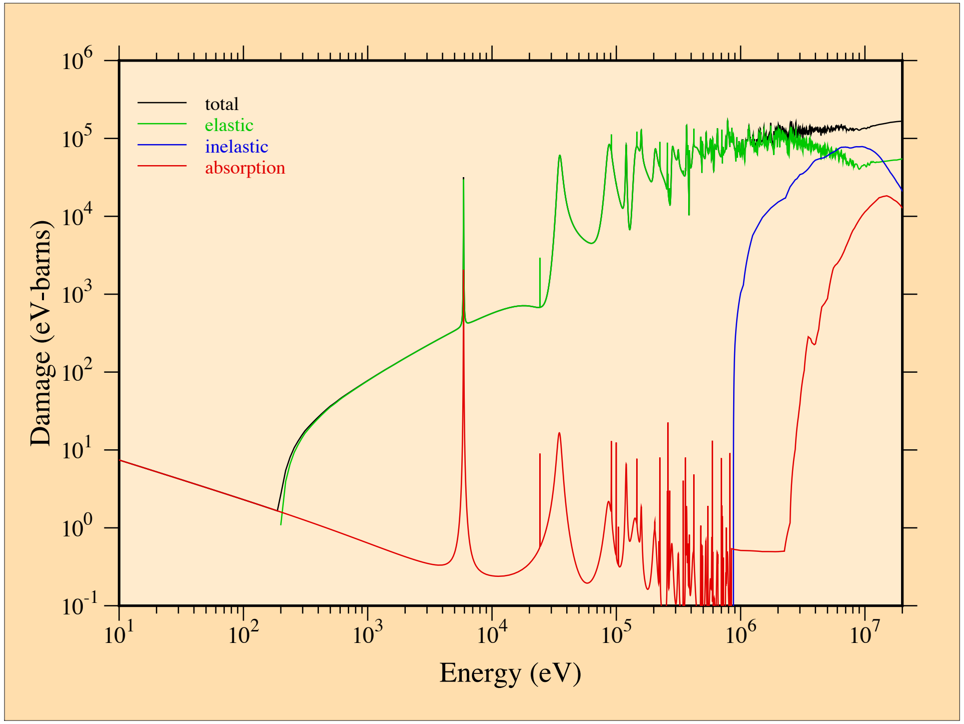
NJOY DPCS and NRT-dpa Ed

Element	E_d (eV)	Element	E_d (eV)
Be	31	Co	40
C	31	Ni	40
Mg	25	Cu	40
Al	27	Zr	40
Si	25	Nb	40
Ca	40	Mo	60
Ti	40	Ag	60
V	40	Ta	90
Cr	40	W	55
Mn	40	Au	30
Fe	40	Pb	25

The mean atomic displacement energy $\bar{E}_d$ is given by

$$\bar{E}_d = \sum_{i=1}^{N_n} N_i E_d(Z_i) / \sum_{i=1}^{N_n} N_i$$





No DPCS, DPA =0 below Ed

NJOY DPCS and NRT-dpa Ed

The number of displaced atoms depends on the total available energy E_a and the energy required to displace an atom from its lattice position E_d. Since the available energy is used up by producing pairs,

$$\mathrm{DPA} = \frac{E_a}{2E_d} \,. \qquad (165)$$

NJOY-2012 manual

- The damage output from HEATR is the Damage energy Production Cross Section (eV-barns), multiplying by the density and flux gives eV/s. Dividing by $2E_d$ gives displacements/s. This result is often reduced by an efficiency factor (say 80%) to improve the fit to the empirical correlations.
- the displacement rate may be estimated using the mean of the displacement rates of the constituents

$$D_{tot} = e_d \phi \sum_{i=1}^{N_n} N_i \bar{d}_i / 2E_d(Z_i) \,.$$

- The displacements per atom rate is given by dividing D_{tot} by the total number of atoms

$$DPA\ rate = D_{tot} / \sum_{i=1}^{N_n} N_i \,.$$

Transmutation: Gas production

- Gas production is treated specially in the output for each time interval in FISPACT-II
  - Important for predicting and modelling material embrittlement
- Estimate of damage rate – the “displacements per atom” (dpa) measure also included

```
0  Total Displacement Rate (n,Ddiss)= 1.19503E+13 Disp./sec = 3.64814E-12 Disp.Per Atom/sec = 1.15127E-04 DPA/year
   Total Displacement Rate (n,Dinel)= 9.08836E+16 Disp./sec = 2.77446E-08 Disp.Per Atom/sec = 8.75554E-01 DPA/year
   Total Displacement Rate (n,Del  )= 7.08317E+16 Disp./sec = 2.16233E-08 Disp.Per Atom/sec = 6.82378E-01 DPA/year
   Total Displacement Rate (n,Dtot )= 1.62935E+17 Disp./sec = 4.97403E-08 Disp.Per Atom/sec = 1.56968E+00 DPA/year
0  KERMA RATE (n,Kktot) =  1.45096E+23 eV/sec = 2.32469E+01 kW/kg = 4.48666E-01 kW/cm^3
   KERMA RATE (n,Kphot) =  1.45067E+23 eV/sec = 2.32423E+01 kW/kg = 4.48576E-01 kW/cm^3
   KERMA RATE (n,Kfiss) =  0.00000E+00 eV/sec = 0.00000E+00 kW/kg = 0.00000E+00 kW/cm^3
   KERMA RATE (n,Kinel) =  1.25683E+21 eV/sec = 2.01366E-01 kW/kg = 3.88637E-03 kW/cm^3
   KERMA RATE (n,Knone) =  1.45530E+23 eV/sec = 2.33166E+01 kW/kg = 4.50009E-01 kW/cm^3
   KERMA RATE (n,Kel  ) =  1.21903E+19 eV/sec = 1.95310E-03 kW/kg = 3.76949E-05 kW/cm^3
   KERMA RATE (n,Ktot ) =  1.45543E+23 eV/sec = 2.33185E+01 kW/kg = 4.50047E-01 kW/cm^3
0  GAS   RATE (n,Xa    ) =  3.34046E+09 atoms per sec =  1.01976E-09 appm/sec
   GAS   RATE (n,Xh    ) =  0.00000E+00 atoms per sec =  0.00000E+00 appm/sec
   GAS   RATE (n,Xt    ) =  8.73363E+06 atoms per sec =  2.66617E-12 appm/sec
   GAS   RATE (n,Xd    ) =  6.28774E+07 atoms per sec =  1.91950E-11 appm/sec
   GAS   RATE (n,Xp    ) =  8.51293E+08 atoms per sec =  2.59880E-10 appm/sec
0  GAS   RATE (a decay) =  1.66048E-01 atoms per sec =  5.06906E-20 appm/sec
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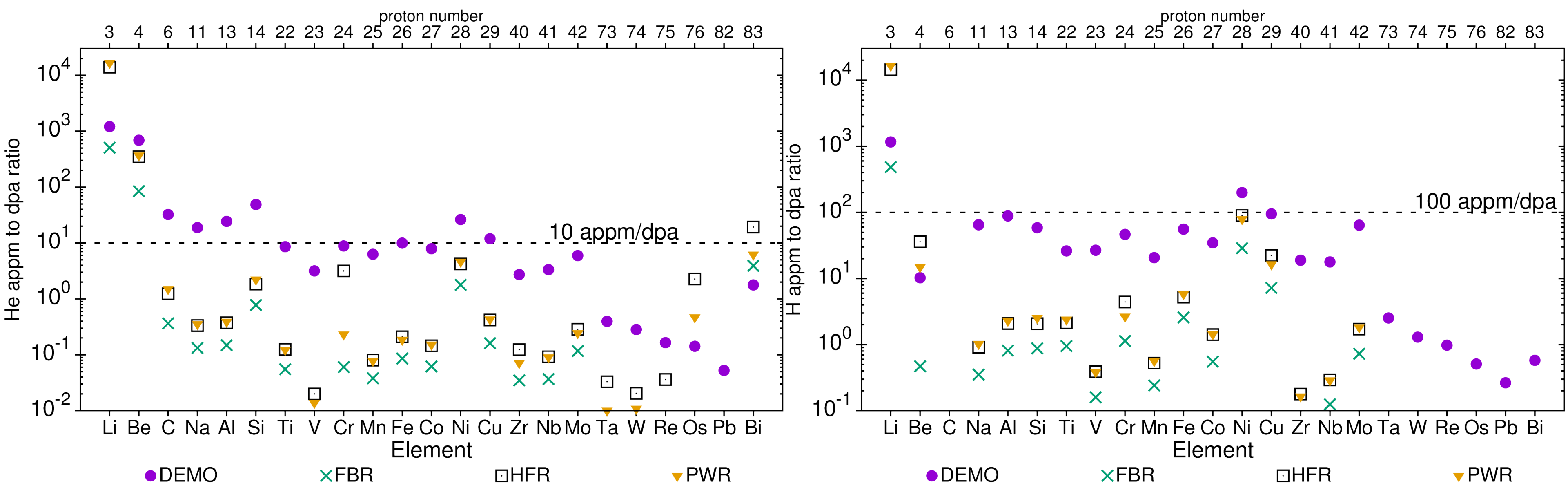
- The combined gas/dpa ratio is a useful measure
  - Particularly when designing experiments to mimic (fusion) reactor conditions

Transmutation: gas/dpa ratios



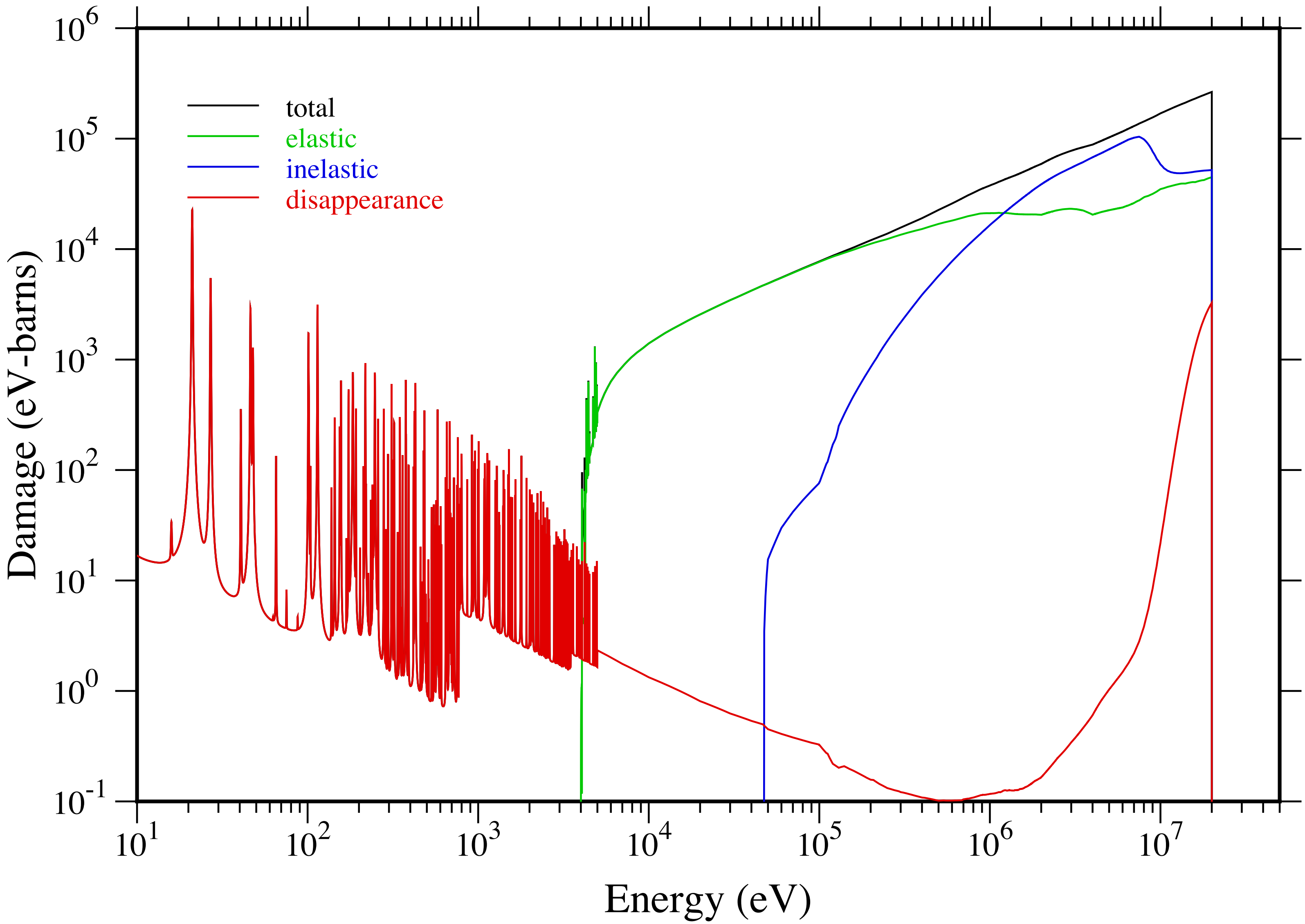
- Fusion trend is lower gas production and hence lower ratios as element mass increases
- The difference between fusion and fission gas-to-dpa ratios (NRT* dpa) is high
- Makes it difficult to realise the correct rates for fusion using fission conditions

TENDL results

*Norgett, Robinson, Torrens, *Nucl. Eng. Des.* **33** (1975) 50-54

Tungsten from JENDL-4

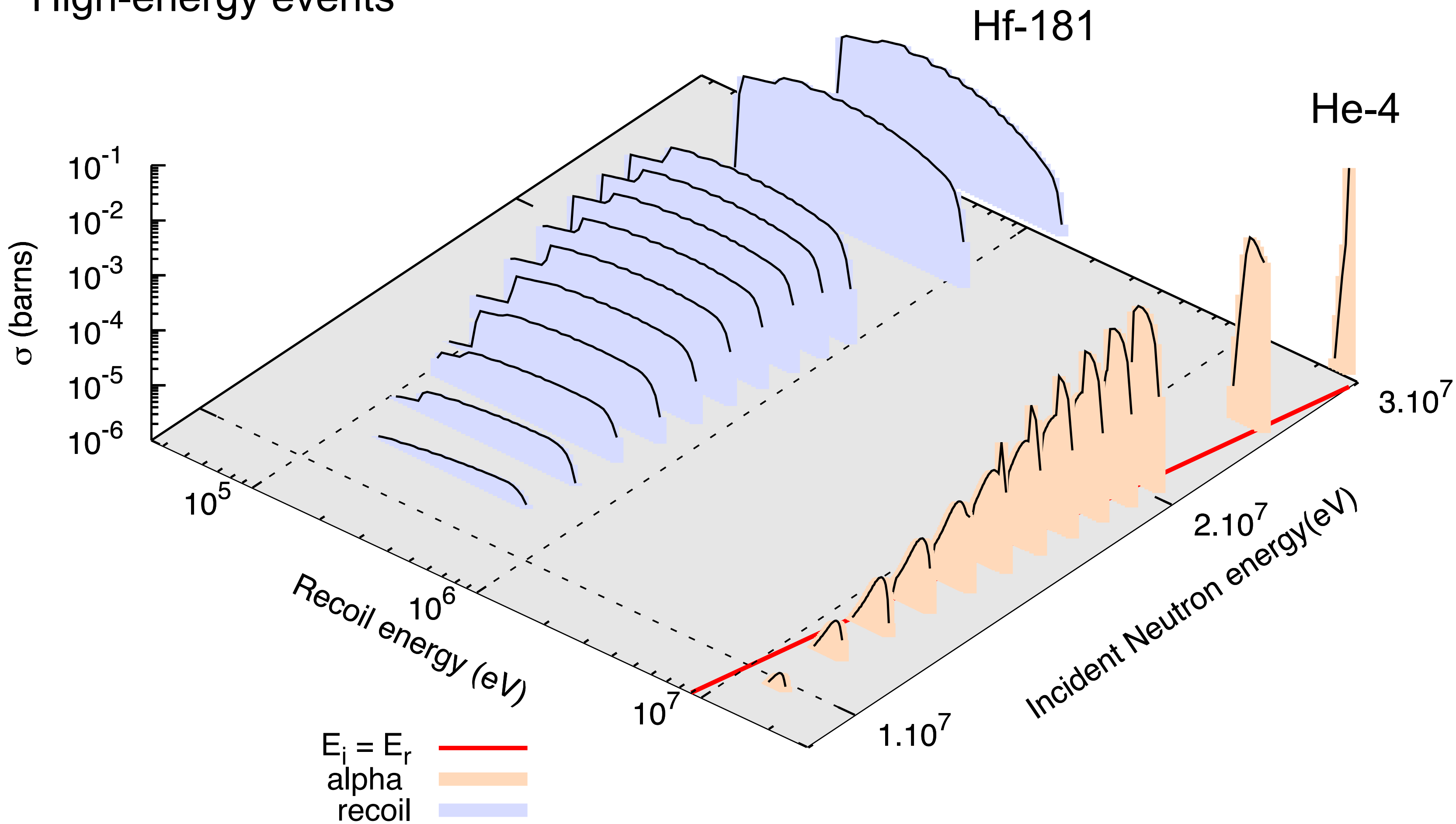
- In red capture elemental (n,γ) channel



W-184 example of matrices

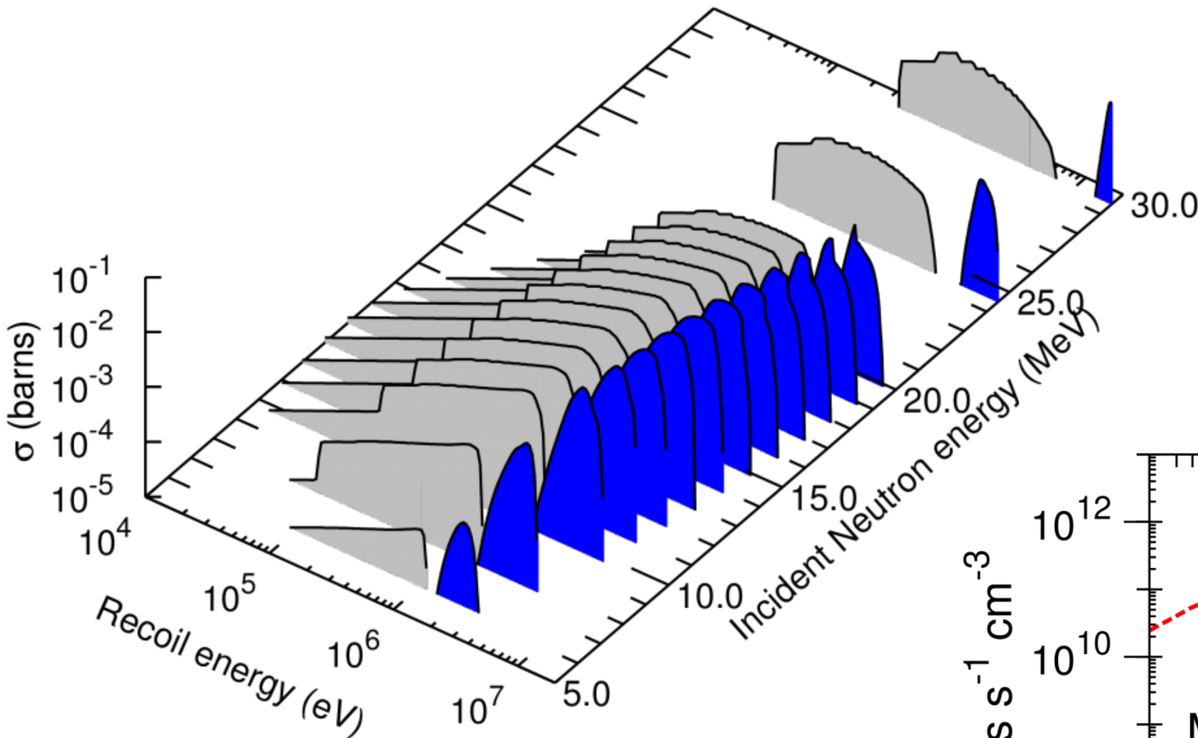
Residual Hf-181 ; $T_{1/2}$=42 days, Beta- to Ta-181 (stable)

High-energy events



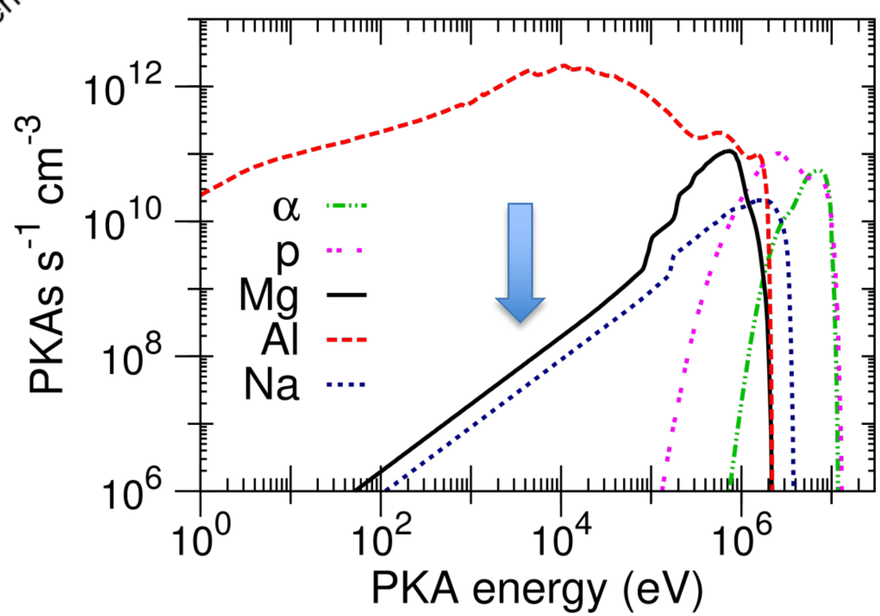
Q positive (7.3 MeV) means that the alpha energy can be much higher than the energy of the n-incident !!! At 22.7 MeV and above the secondary energy grid is truncated !!!

NJOY processing ismooth = 1



Tails are important

Transmuted residuals also: Mg, Na



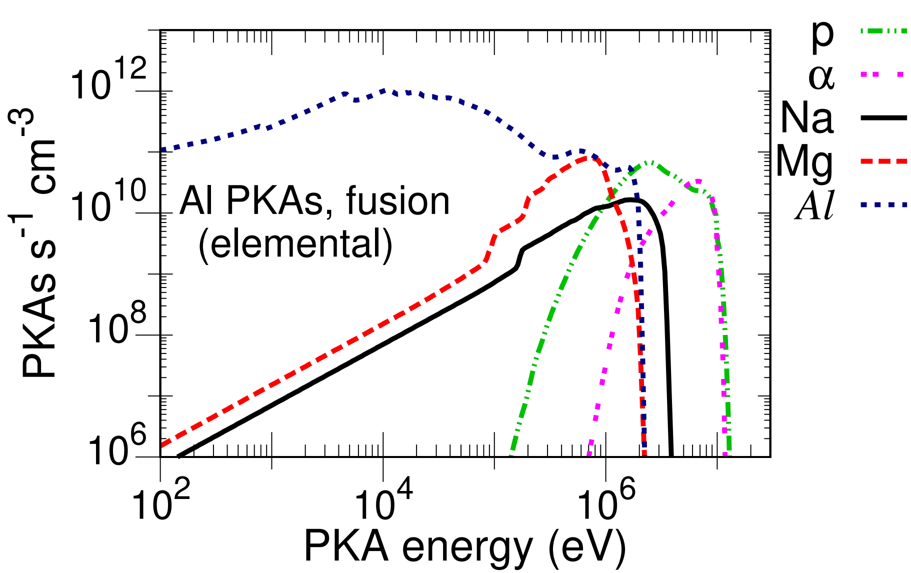
Q negative this time, but NJOY ismooth =1 ($\sqrt{E}$ shape) for when the evaluator decided to cut short the secondary energy grid of the recoil!

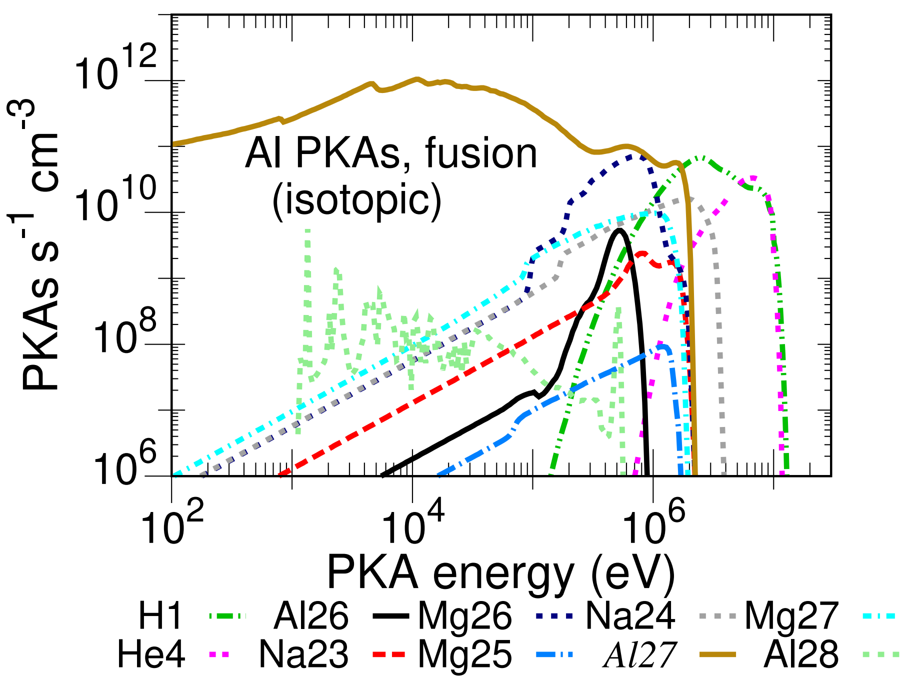
Damage metrics

- SPECTRA-PKA code reads-in the aforementioned recoil matrices and combines these with an incident neutron energy spectrum to define PKA event and energy distributions
- The code has the advantage of being fully compatible with the latest modern nuclear data libraries, for both neutron and charge particles, and can handle fine group structures
- The code can also consider any complex material composition containing an arbitrary distribution of target nuclide species.
- Even more significantly, it treats every nuclear reaction channel (on every target nuclide considered), and its associated recoil matrix, separately, which allows a deeper interrogation of the underlying nuclear data

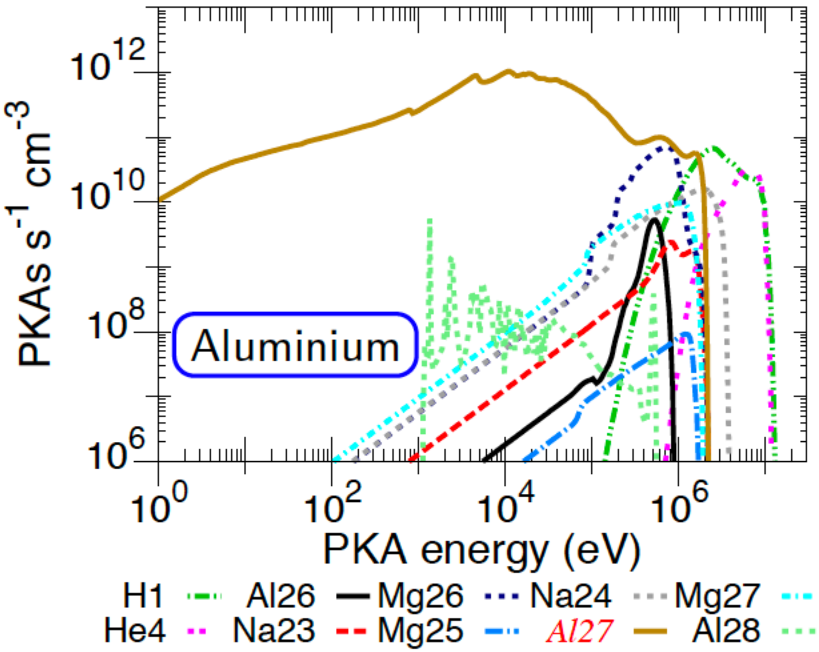
Detailed metrics

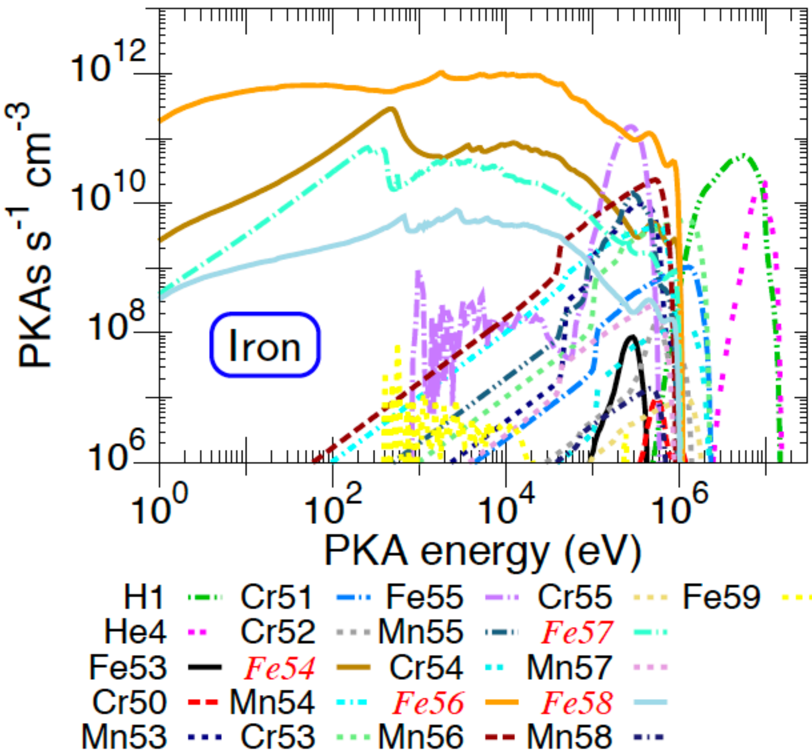
- Pure aluminum (100% ^{27}Al) transmuted residual elements and emitted particle PKA distributions under fusion neutron conditions, right elemental, left isotopic





Underlying complexity shown with SPECTRA-PKA





- very complex results with numerous recoils species (isotopes & elements, many radioactive)
- but already hiding some of the per-channel information that is available from the output

https://github.com/fispact/SPECTRA-PKA

PKA spectra

- The term primary knock-on atoms (PKA) refers to the initial recoil atom(s) created by neutron (or proton, deuteron etc.) interactions with the constituent atoms of a material
- These PKAs can initiate cascades of displacements, leading to the accumulation of structural damage
  - Life-limiting consequences for materials
- A full description of the PKAs generated under neutron irradiation is a necessary source term input to modelling and simulation of the creation & evolution of radiation damage
  - Can also be used to design experiments to mimic neutron damage

PKA spectra

- Complete description of PKAs requires all possible nuclear reaction channels to be considered
  - More than scattering
  - non-elastic reactions
  - & including all recoiling species of a reaction (secondary emitted, light particles)

- The resulting energy versus flux spectra of PKAs provides a much richer data-set
  - More detail than simpler damage measures (e.g. DPA)
  - uses more of the information available from particle transport and inventory simulations

Computational methodology:

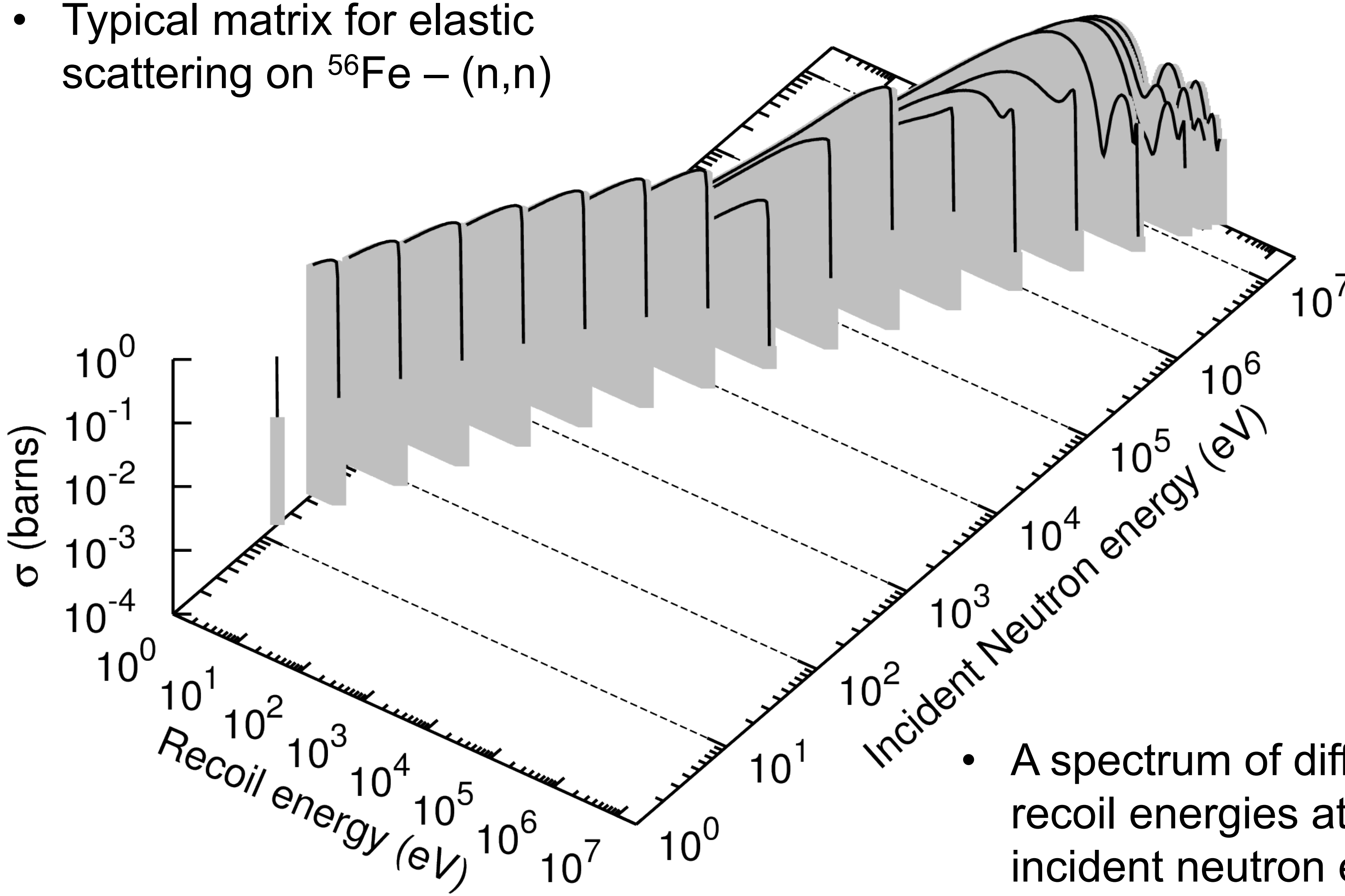
- NJOY processes the raw point-wise nuclear data and calculates neutron-interaction recoil matrices:

$$M^{x\longrightarrow y} \equiv \left\{m_{ij}^{x\longrightarrow y}\right\}$$

  - $m_{ij}^{x\longrightarrow y}$ 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
  - Derived from reaction cross sections and angular distributions
  - For each parent target isotope (e.g. ^{56}Fe) there will be a set of $M^{x\longrightarrow y}$
    - & perhaps more than one matrix for a given nuclear reaction e.g. (n,α) will produce a heavy recoil and a ^{4}He

PKA spectra – recoil matrices

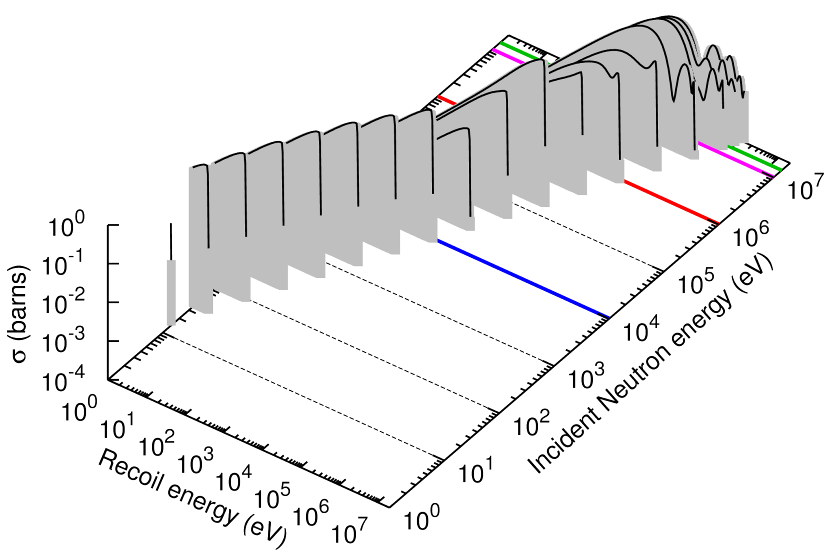
- Typical matrix for elastic scattering on ^{56}Fe – (n,n)

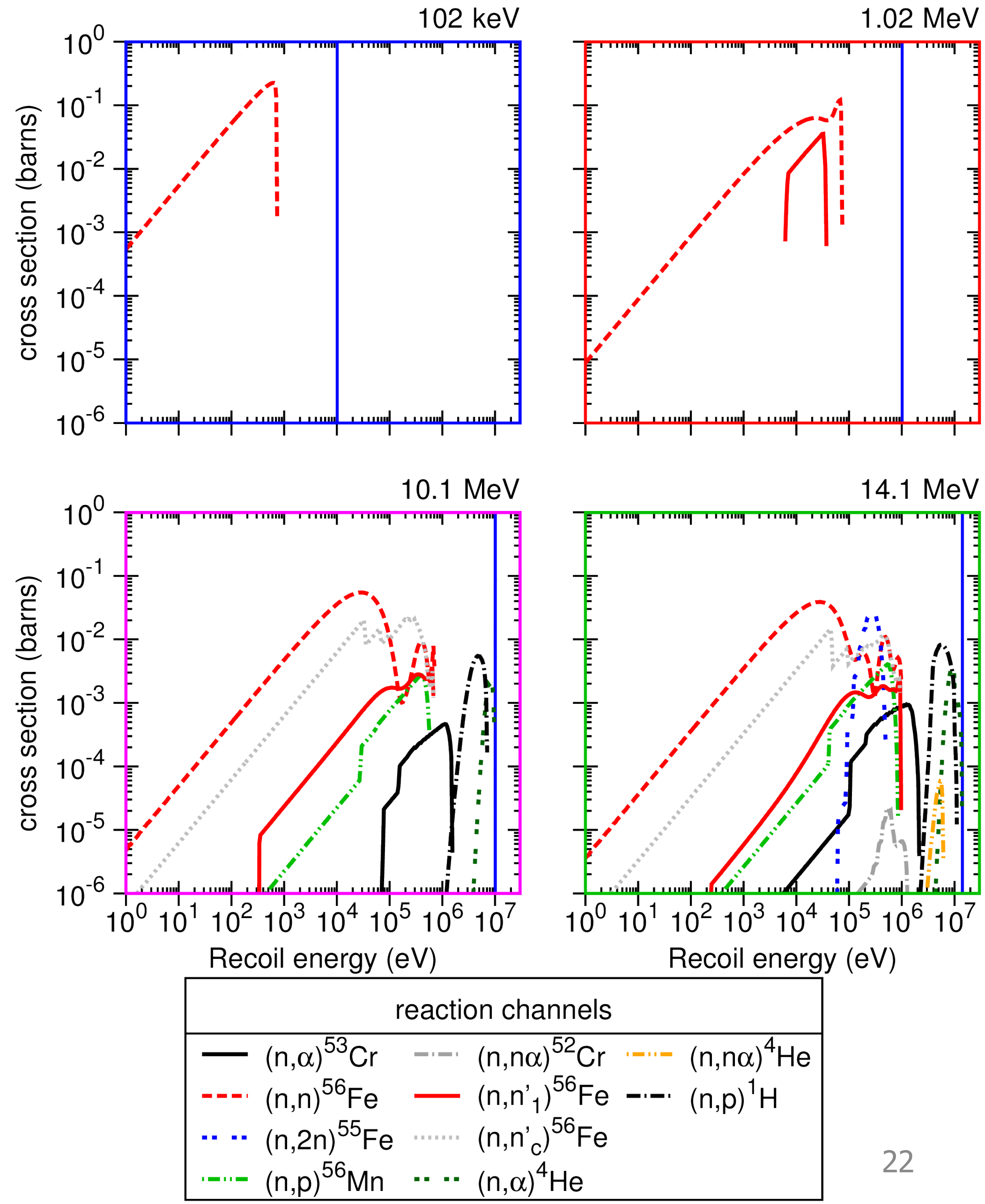


- A spectrum of different recoil energies at each incident neutron energy

TENDL results

PKA spectra – recoil matrices & vectors



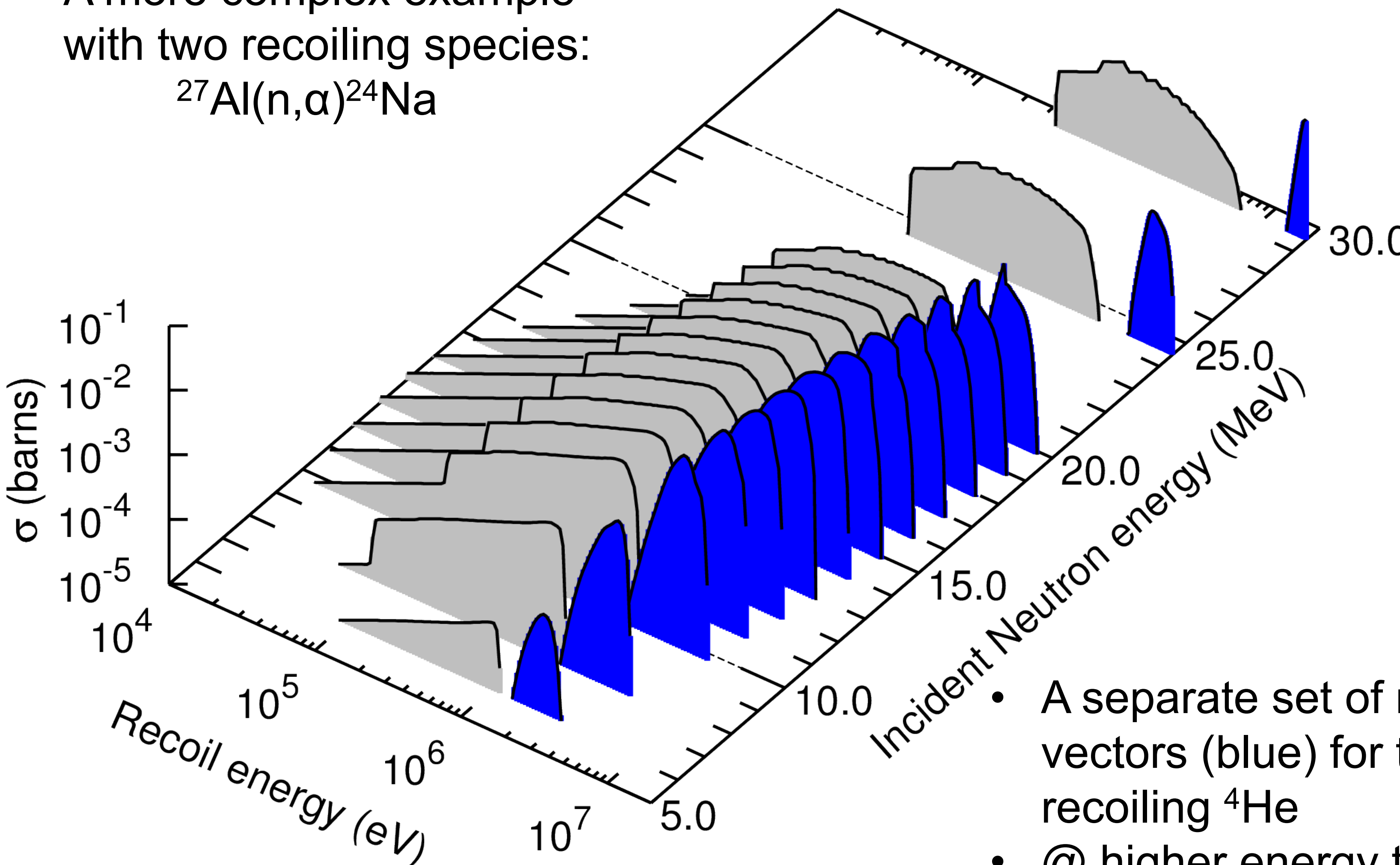


- In ^{56}Fe: at low incident energy only elastic and inelastic channels are open
- Number of contributing channels increases dramatically at higher energies (MeV range)

TENDL results

PKA spectra – recoil matrices

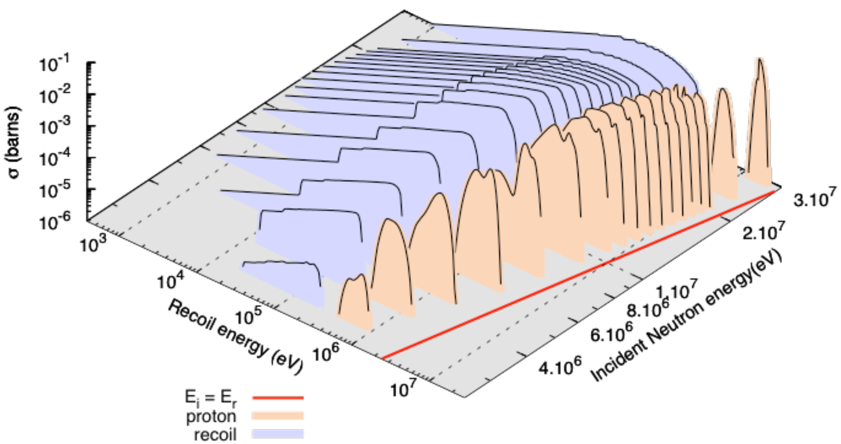
- A more complex example with two recoiling species:
  $^{27}Al(n,\alpha)^{24}Na$



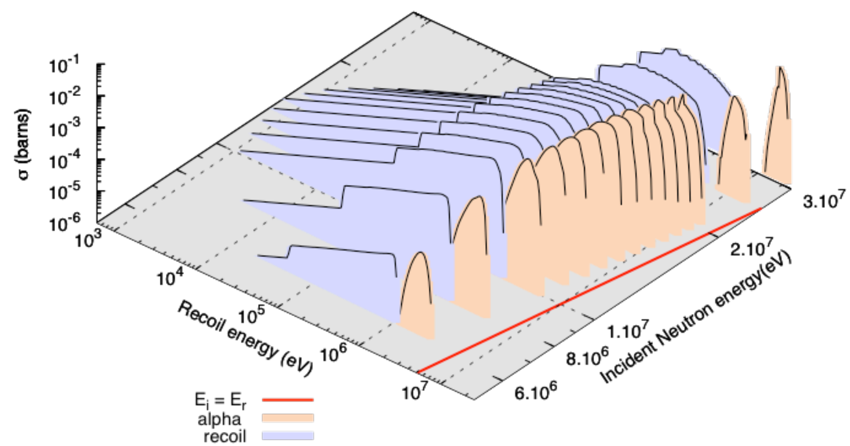
- A separate set of recoil vectors (blue) for the recoiling ^{4}He
- @ higher energy than heavy ^{24}Na recoil (mass partition)

TENDL Al-27 and Fe-56

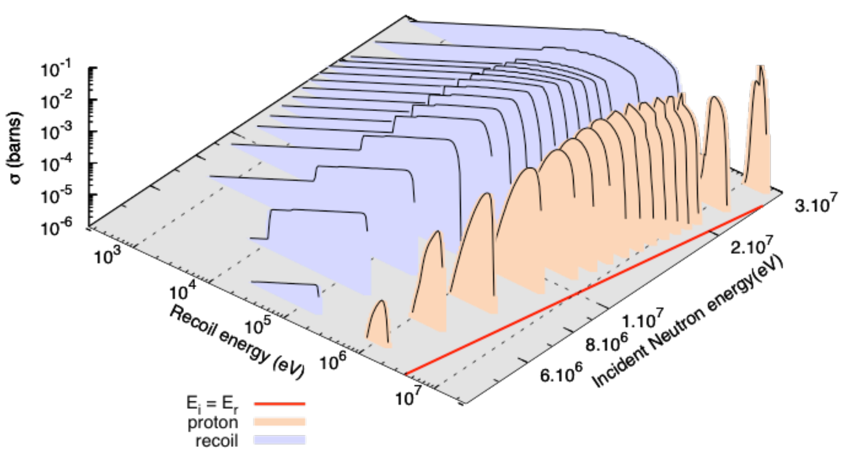
Al027 (n,p)



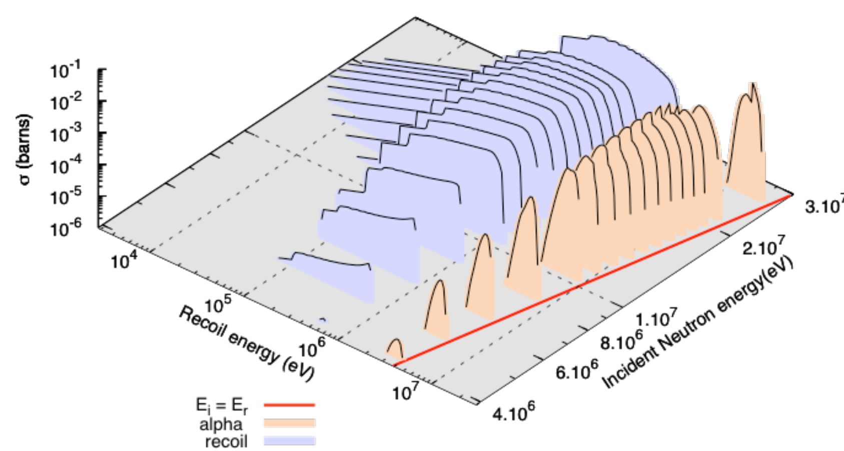
Al027 (n,a)



Fe056 (n,p)



Fe056 (n,a)



PKA spectra

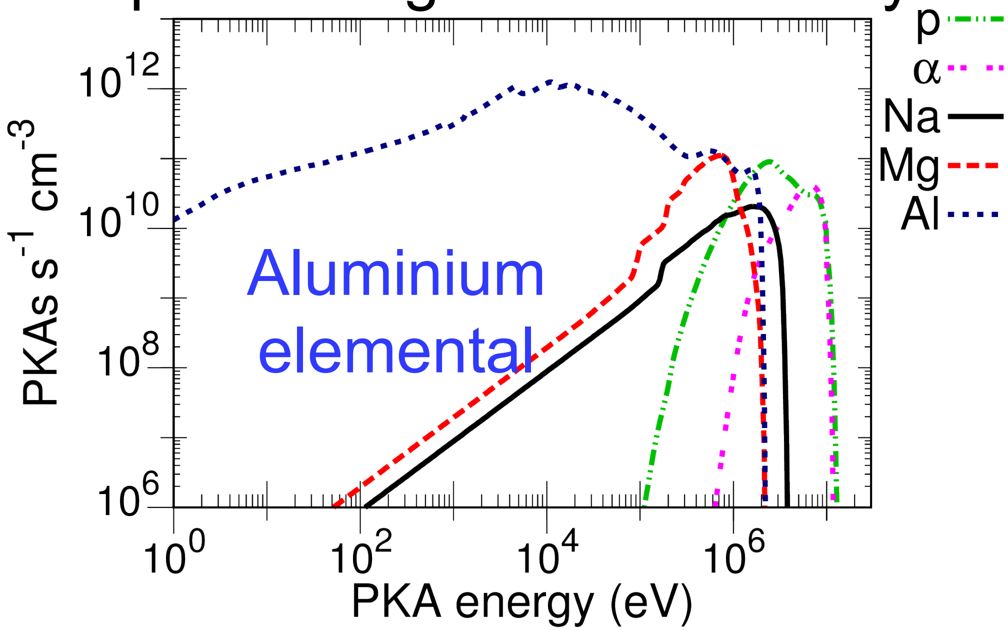
Computational methodology (b): *Gilbert et al., *JNM* **467** (2015) 121-134

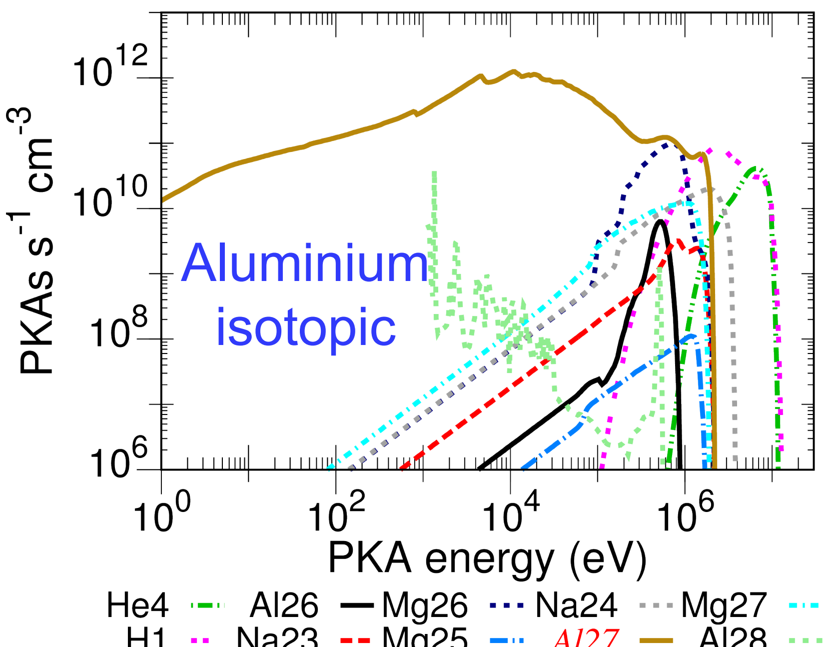
- Collapsing each matrix with a neutron flux spectrum gives the recoil (PKA) spectrum $R^{x\longrightarrow y}(E)$:

$$R^{x\longrightarrow y}(E) \equiv \left\{r_i^{x\longrightarrow y}\right\} = \left\{\sum_j m_{ij}^{x\longrightarrow y} \phi_j\right\},$$

where ϕ_j is the neutron flux at energy E_j for a given neutron field

- processing done with newly written SPECTRA-PKA*

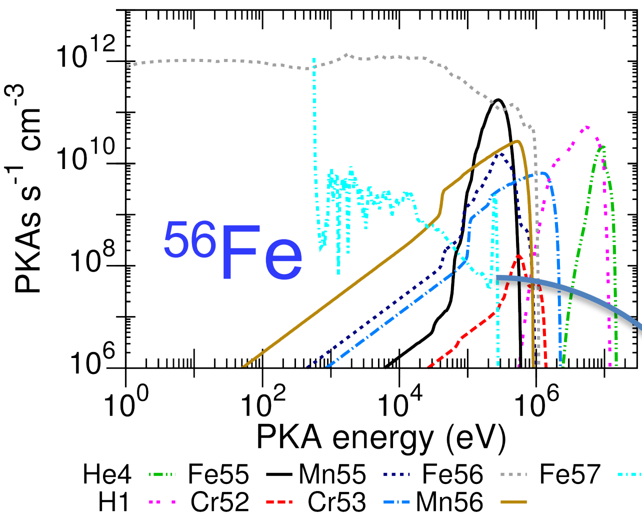


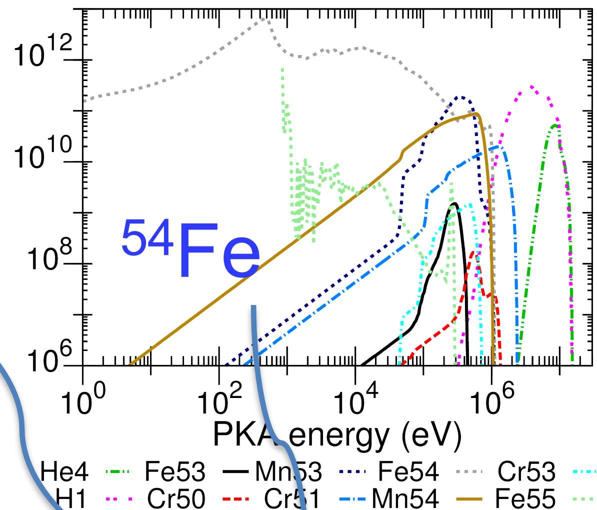


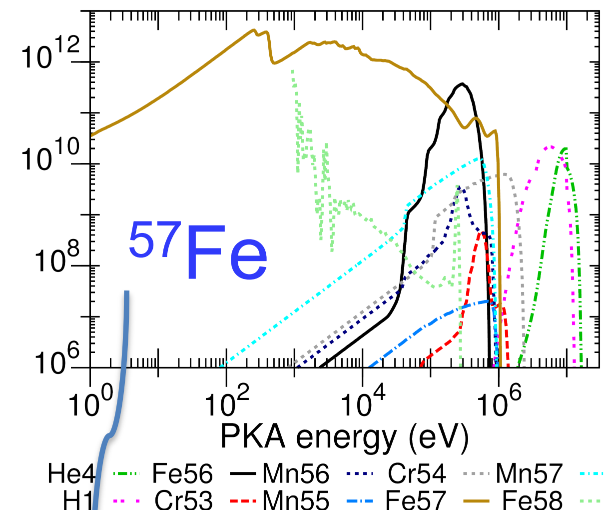
- Al under DEMO first wall conditions (TENDL results)
- Even with only one target (^{27}Al), there are many different recoiling species

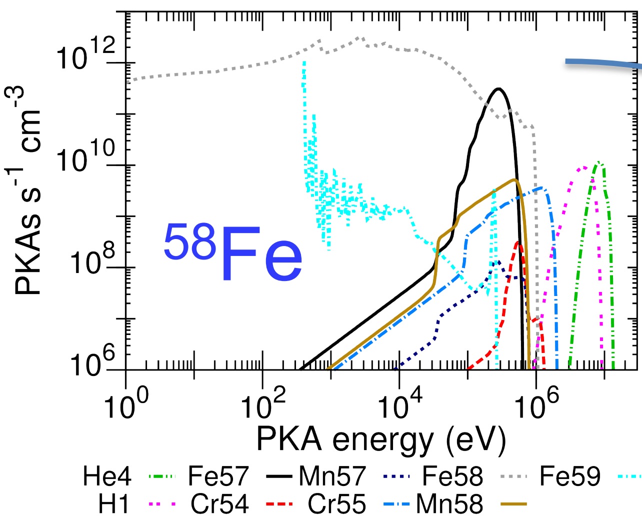
PKA spectra – a multi-target (isotope) material

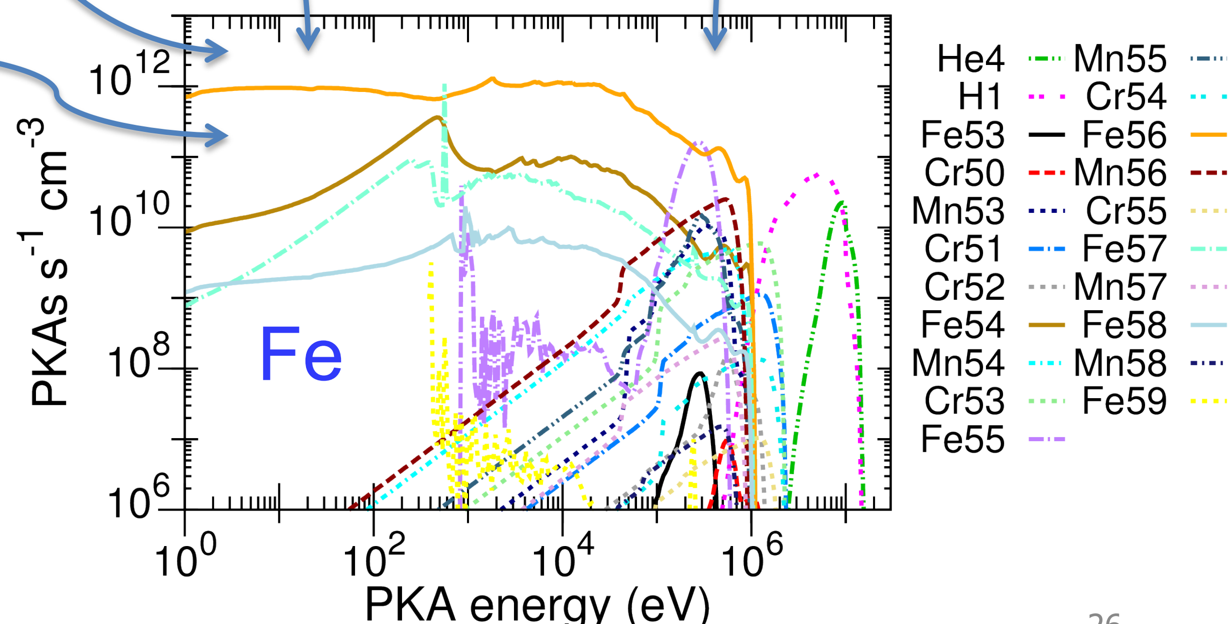
- In most cases, results must be weighted and merged for multiple targets











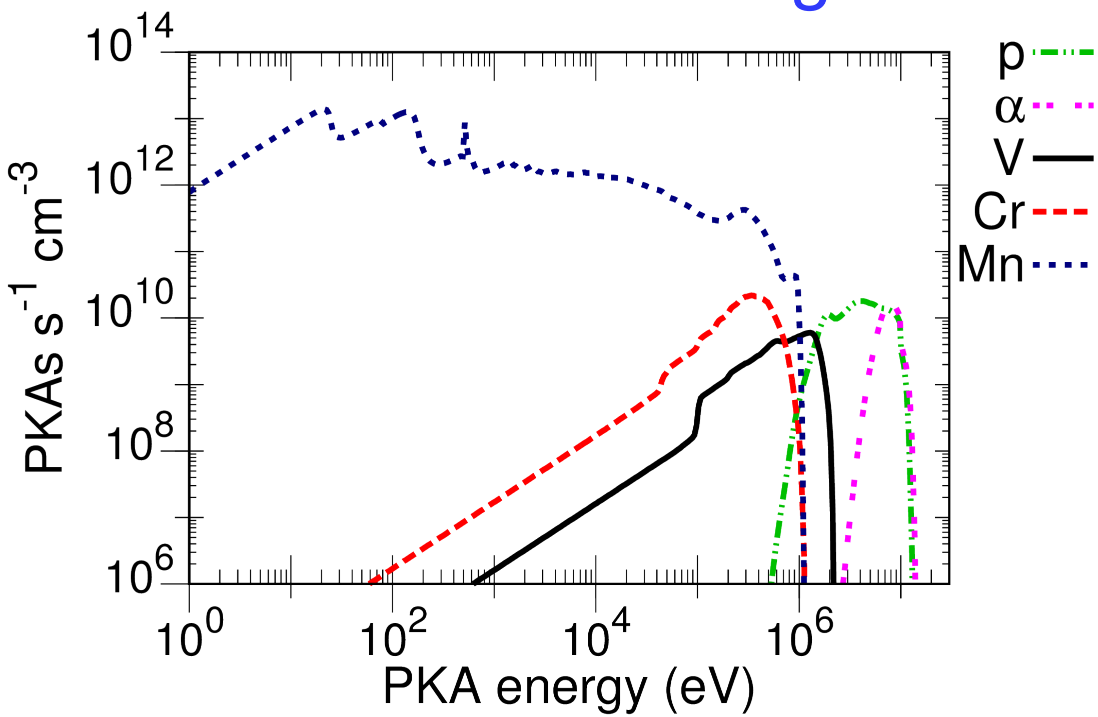
Fe - 5.845% ^{54}Fe, 91.754% ^{56}Fe, 2.119% ^{57}Fe, 0.282% ^{58}Fe

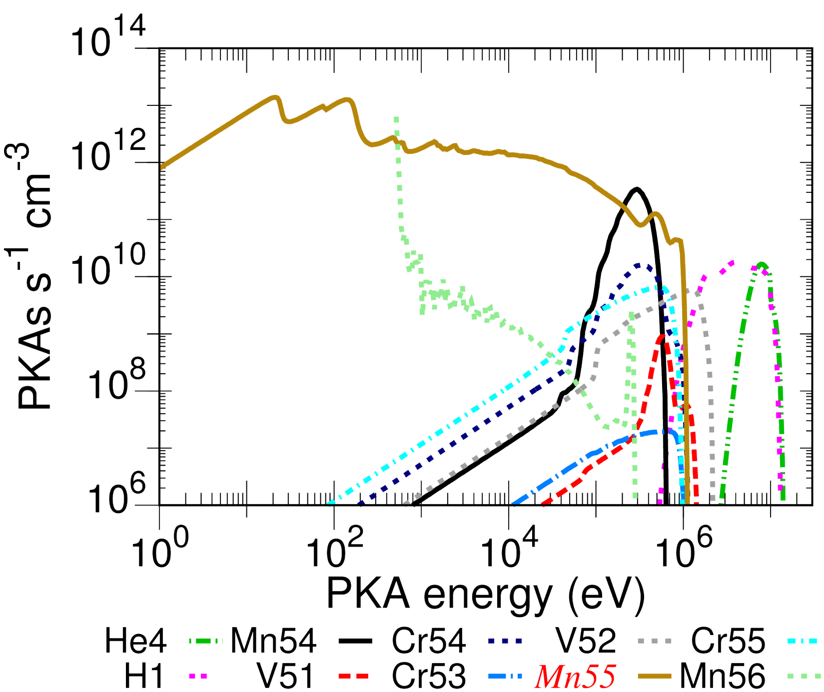
- SPECTRA-PKA module handles all the necessary processing

PKA spectra

- Handbooks present elemental and isotopic PKA spectra for each element under relevant neutron irradiation
  - A t=0 snapshot (before any transmutation)

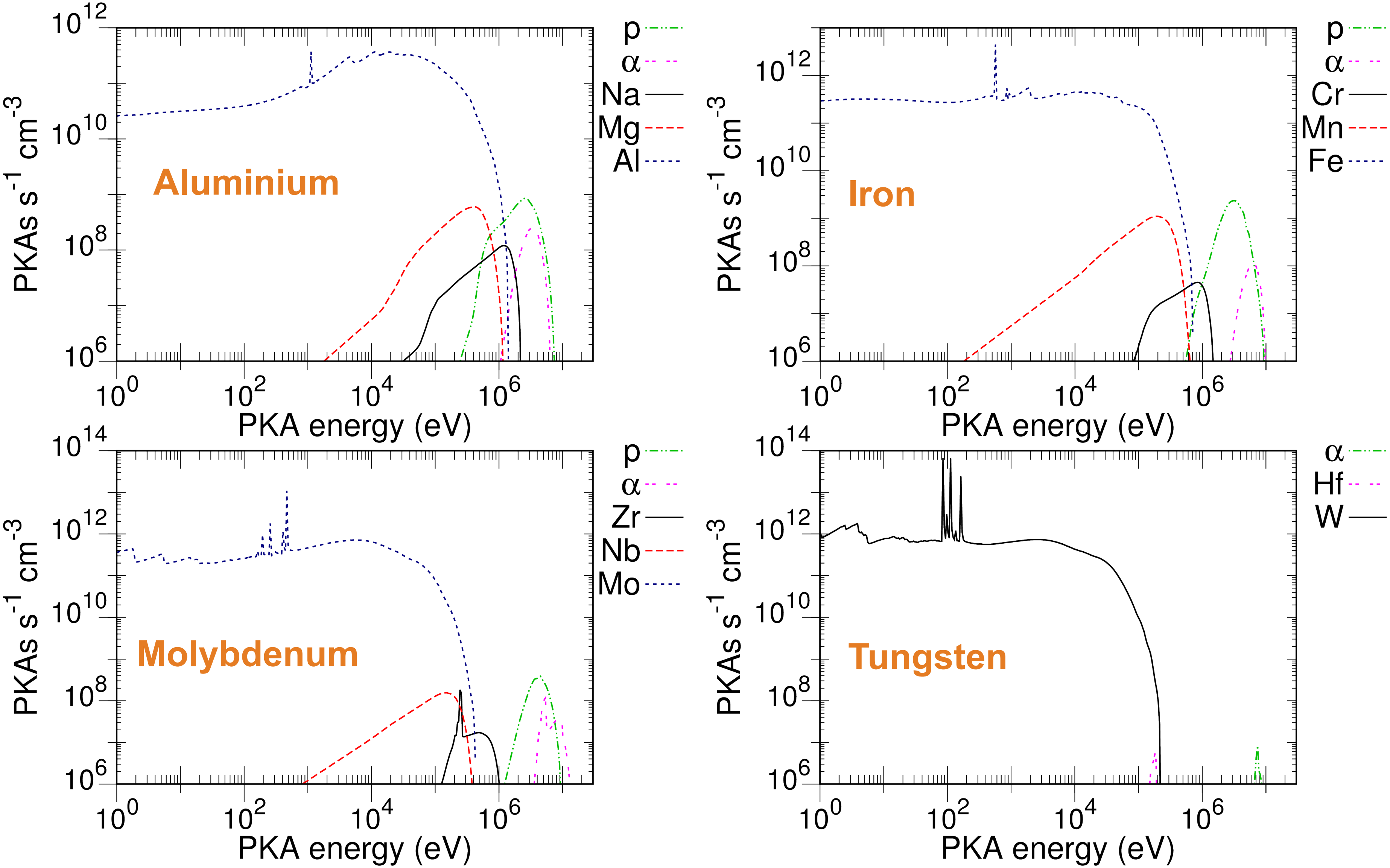
Manganese





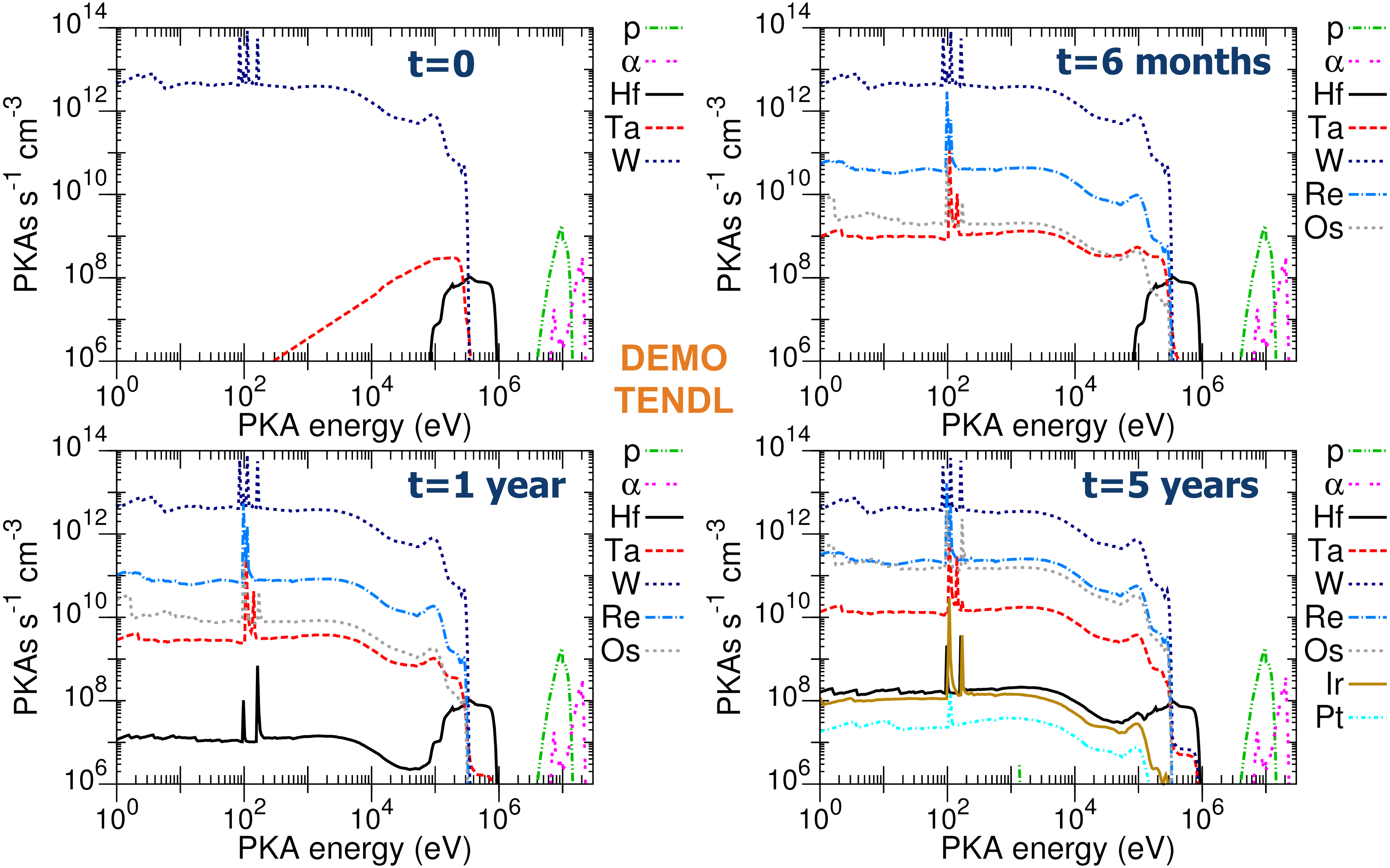
PKA spectra

- PKAs in **different elements** under PWR conditions:



PKA spectra – time evolution

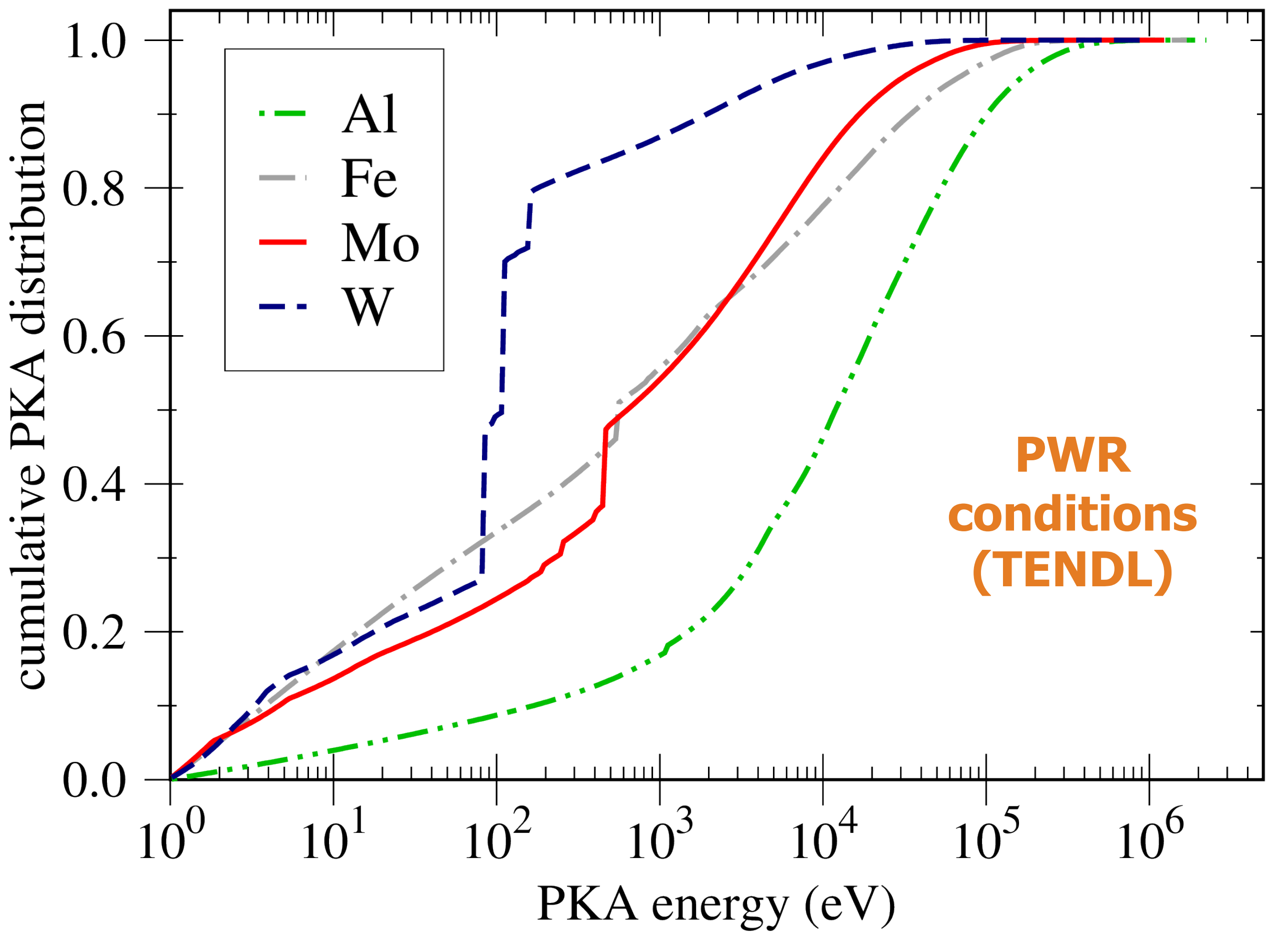
- FISPACT-II & SPECTRA-PKA can show time evolution due to transmutation



- PKAs from collisions with Re, Os, Ta, etc., appear with time

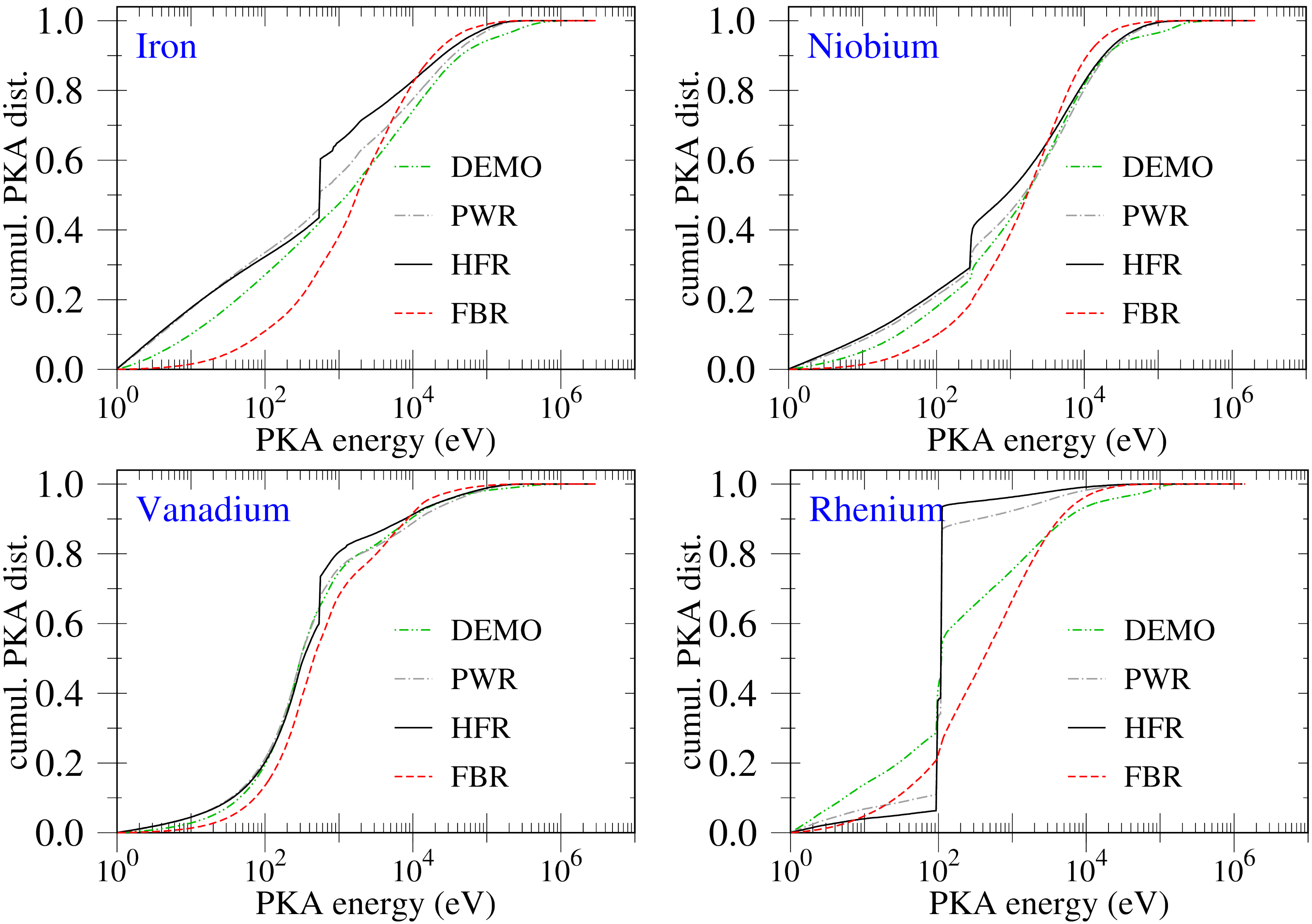
Cumulative PKA distributions (CPDs)

- Summing over all heavy recoils (gases excluded) reveals fundamental differences between materials
- **CPDs can be used as a sampling distribution for damage cascade modelling**



- Distributions of lighter elements shifted to higher energies

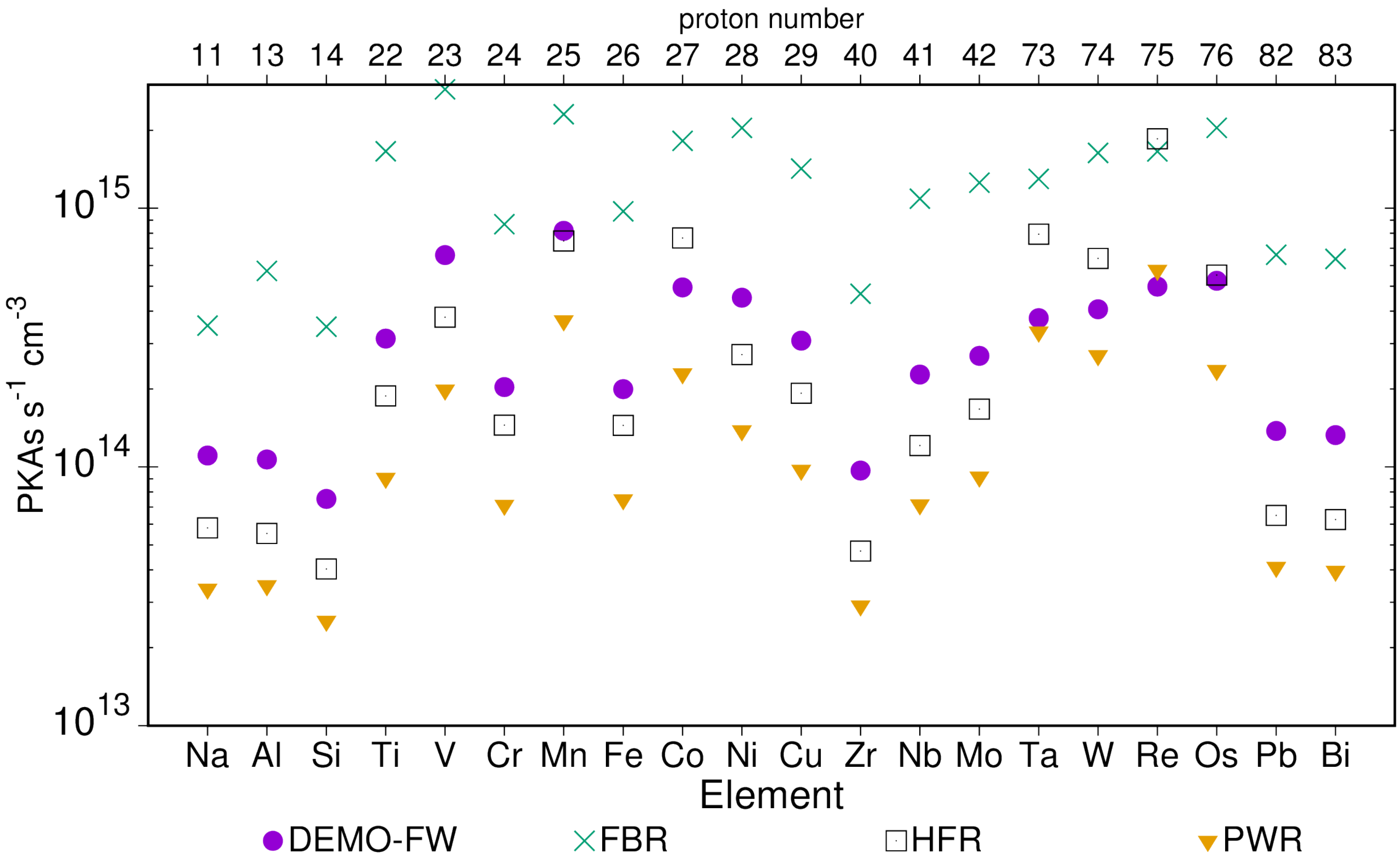
(5) CPDs: fusion vs. fission



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

(5) Total PKAs: fusion vs. fission

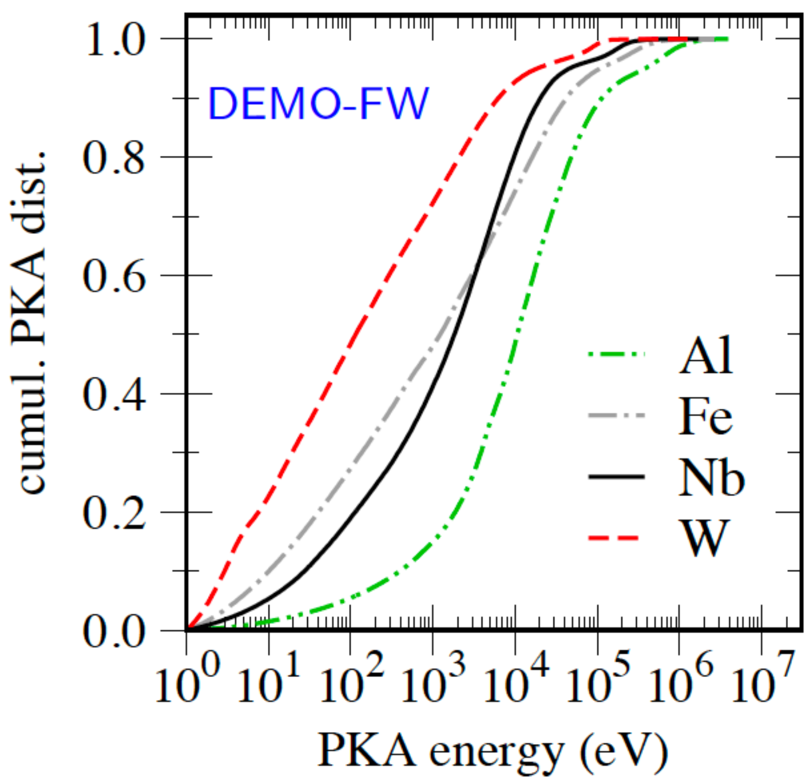
- Total PKAs for selected materials:

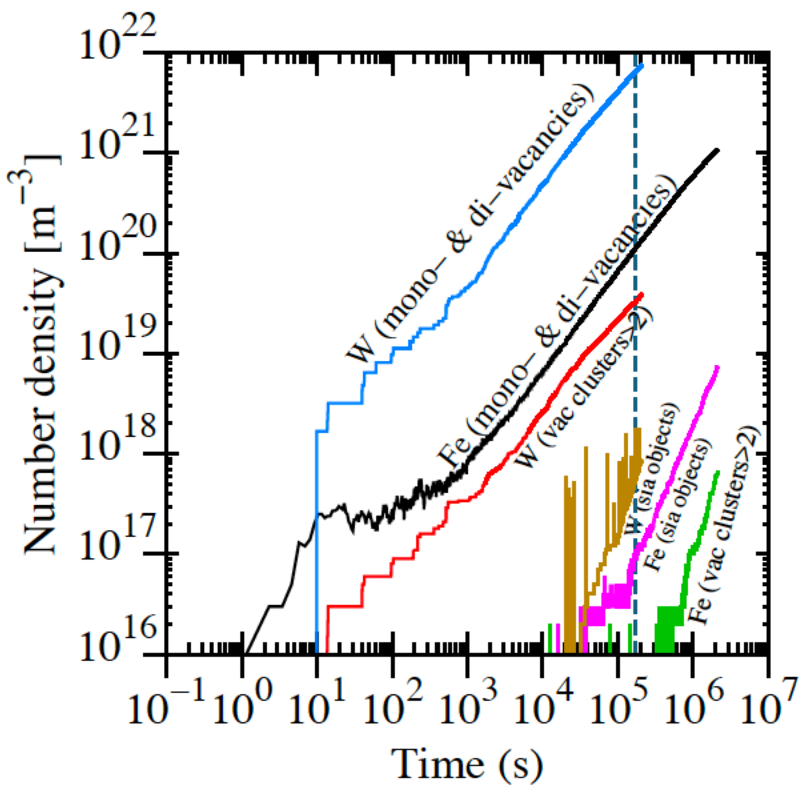


- FBR spectrum produces highest rates due to high, fast flux (compare with HFR where high, soft flux instead produced high transmutation)
  - "existing" fission (PWR) the lowest
- More problems matching fusion in fission systems

Cumulative PKA distributions CPDs

- From a material modelling perspective a more important metric than the raw PKA flux-energy distributions are the cumulative probability distributions of PKAs
- CPDs show the relative energy distribution of PKAs
- can be applied as sampling distributions for defect formation & evolution simulations

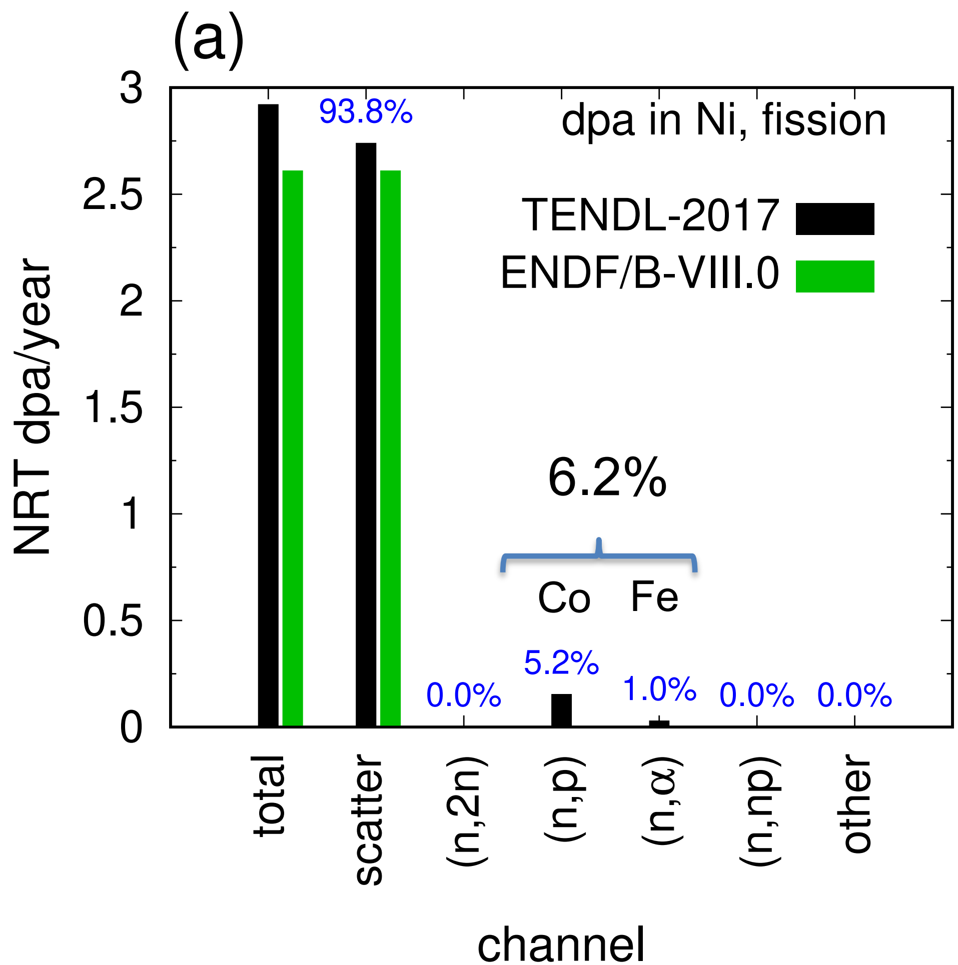


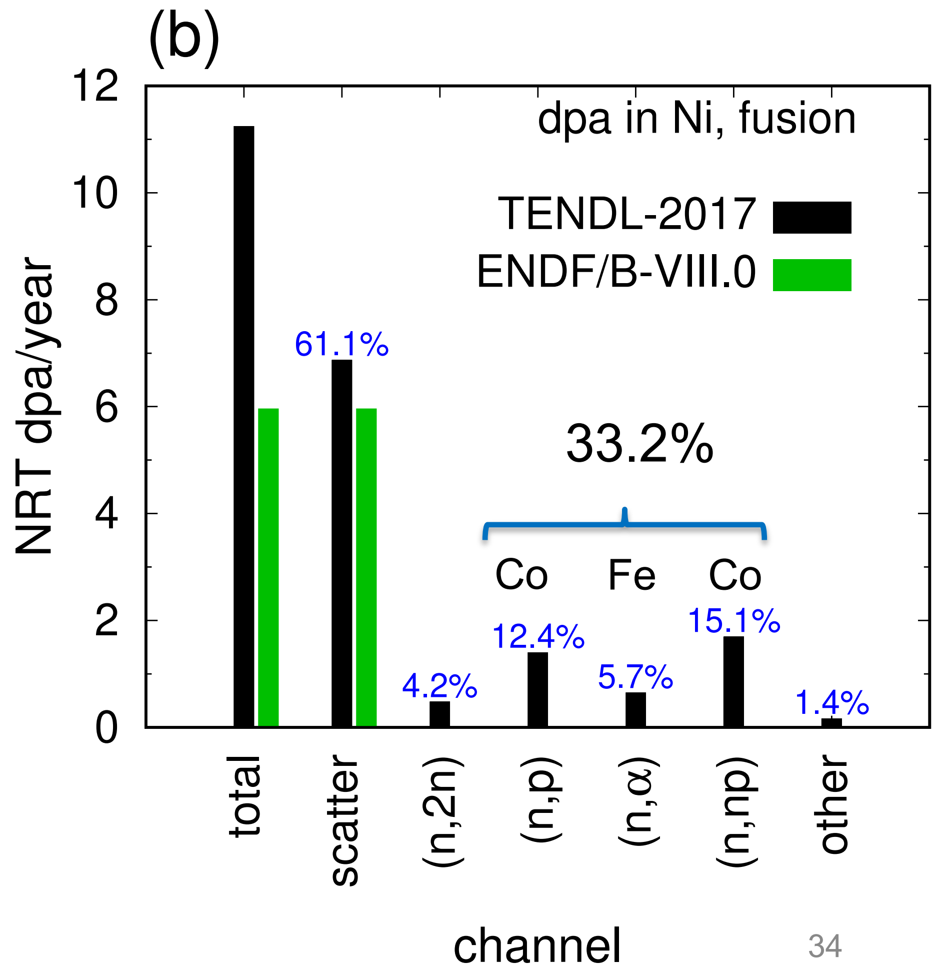


Stochastic cluster dynamics

Novel metrics

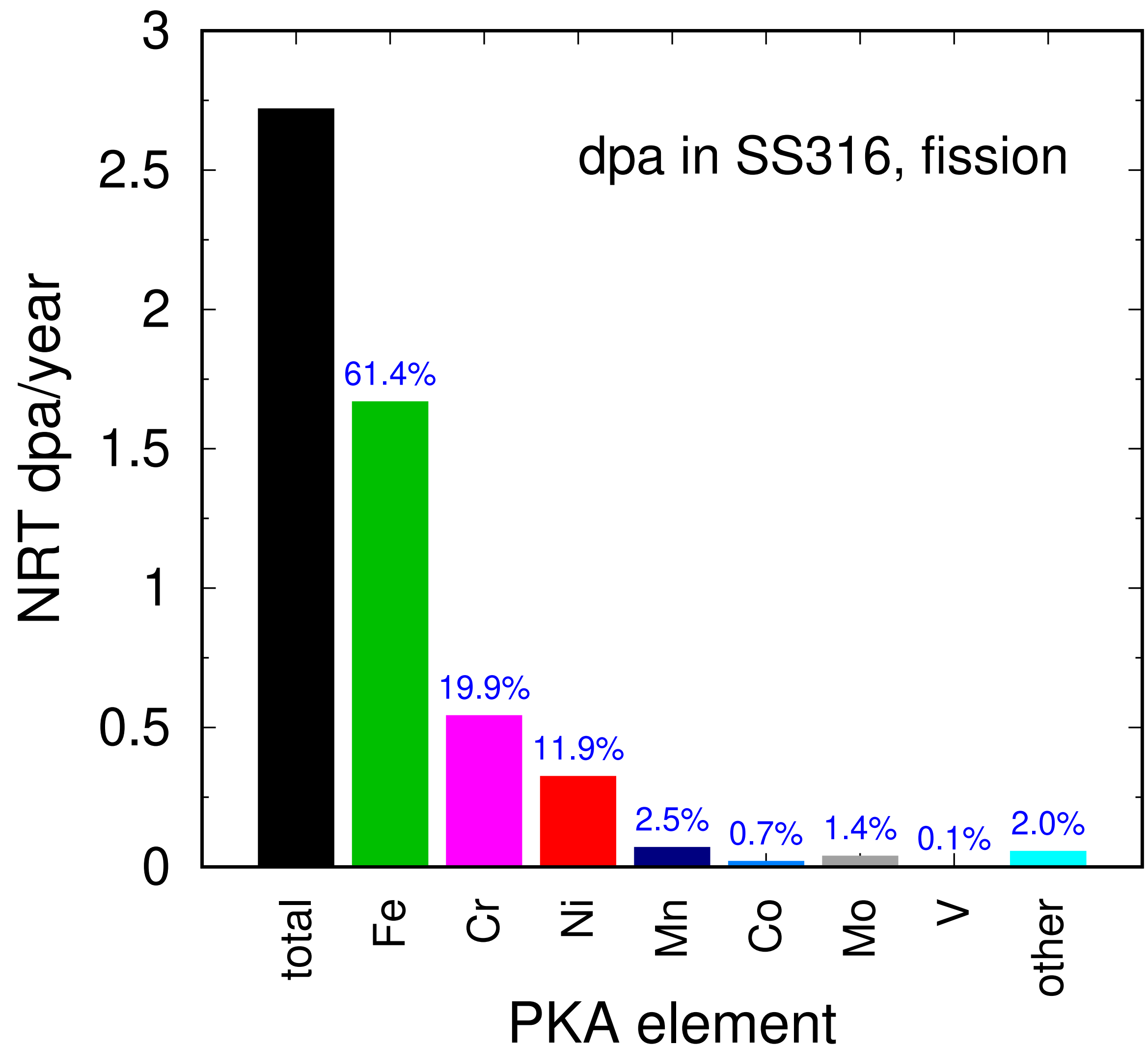
- New capabilities of SPECTRA-PKA have been exploited to analyze the relative significance of different nuclide channels to dpa damage production rates: (a) PWR - (b) Fusion FW





Novel metrics

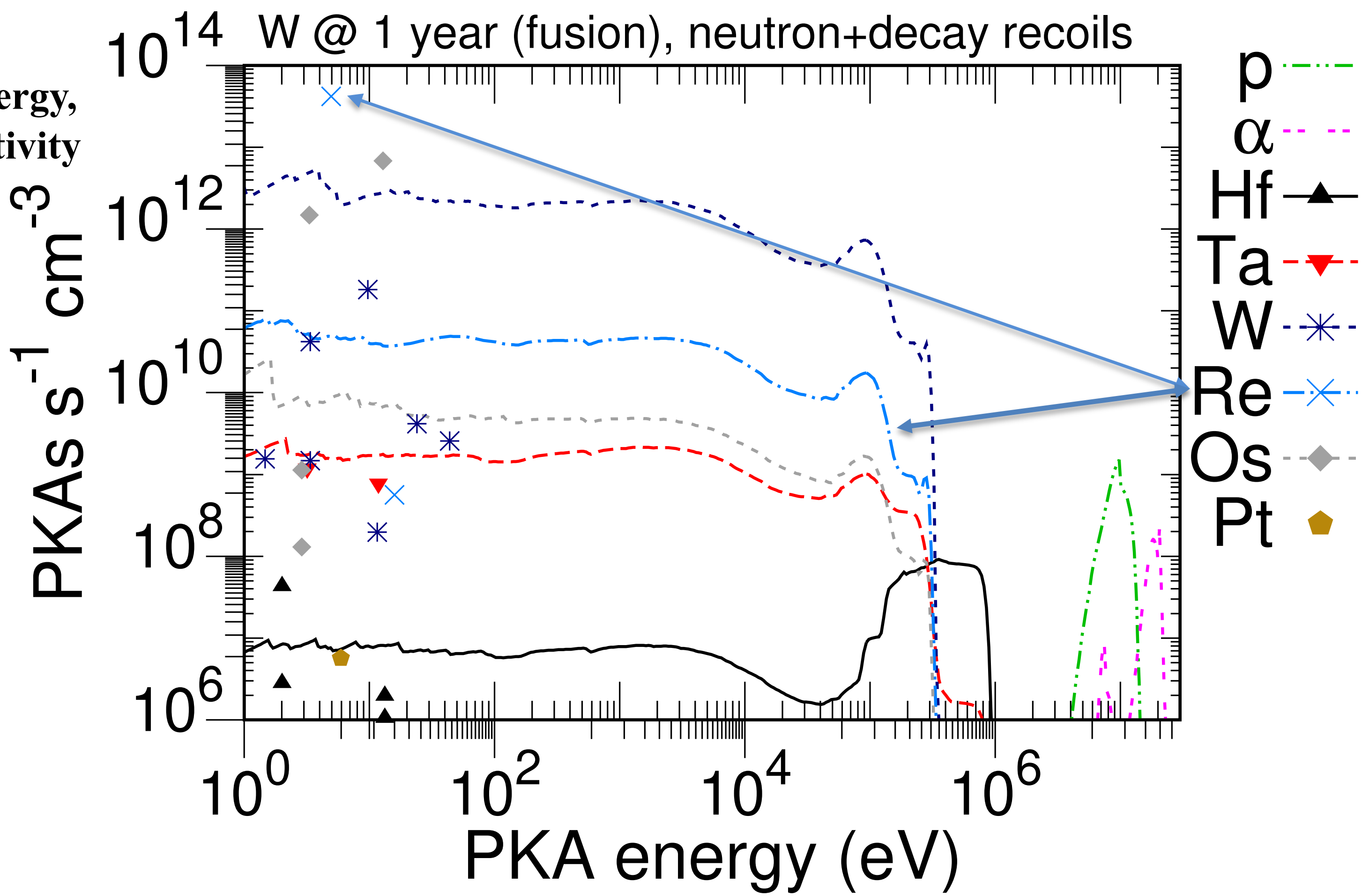
- Dpa contributions to the total damage rate in SS316 steel under PWR conditions



Novel metrics

- PKA contributions from both transmutant/descendant elements (curves) and decaying species (points) to the PKA distributions in pure tungsten after a 1-year irradiation in a typical fusion neutron field

Low energy, high activity



Progress @ nuclear scale

- Definitely a step forward in the proper understanding of materials defect metrics induced by radiations
  - much better nuclear data (with uncertainty)
  - more complete data forms
  - Transmutation, decaying effects
  - non-elastic events
  - Incident particle energy dependence
- A much better coverage of the high energy range
- Novel event per event, channel metrics: "Differential dpa calculations with SPECTRA-PKA" Journal of Nuclear Materials 504 (2018) 101-108
- Uncertainty quantification and propagation UQP

==> to better serve multi-scale, -physics simulations software

Litterature

https://www-nds.iaea.org/CRPdpa/

https://github.com/fispact/SPECTRA-PKA

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Nuclear Data Sheets
Volume 139, January 2017, Pages 77-137

FISPACT-II: An Advanced Simulation System for Activation, Transmutation and Material Modelling

J.-Ch. Sublet [a], J.W. Eastwood [b], J.G. Morgan [b], M.R. Gilbert [a], M. Fleming [a], W. Arter [a]

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Neutron-induced damage simulations: Beyond defect production cross-section, displacement per atom and iron-based metrics

J. -Ch. Sublet, I. P. Bondarenko, G. Bonny, J. L. Conlin, M. R. Gilbert, L. R. Greenwood, P. J. Griffin, P. Helgesson, Y. Iwamoto, V. A. Khryachkov, T. A. Khromyleva, A. Yu. Konobeyev, N. Lazarev, L. Luneville, F. Mota, C. J. Ortiz, D. Rochman, S. P. Simakov, D. Simeone, H. Sjostrand, D. Terentyev & R. Vila

The European Physical Journal Plus **134**, Article number: 350 (2019) | Cite this article

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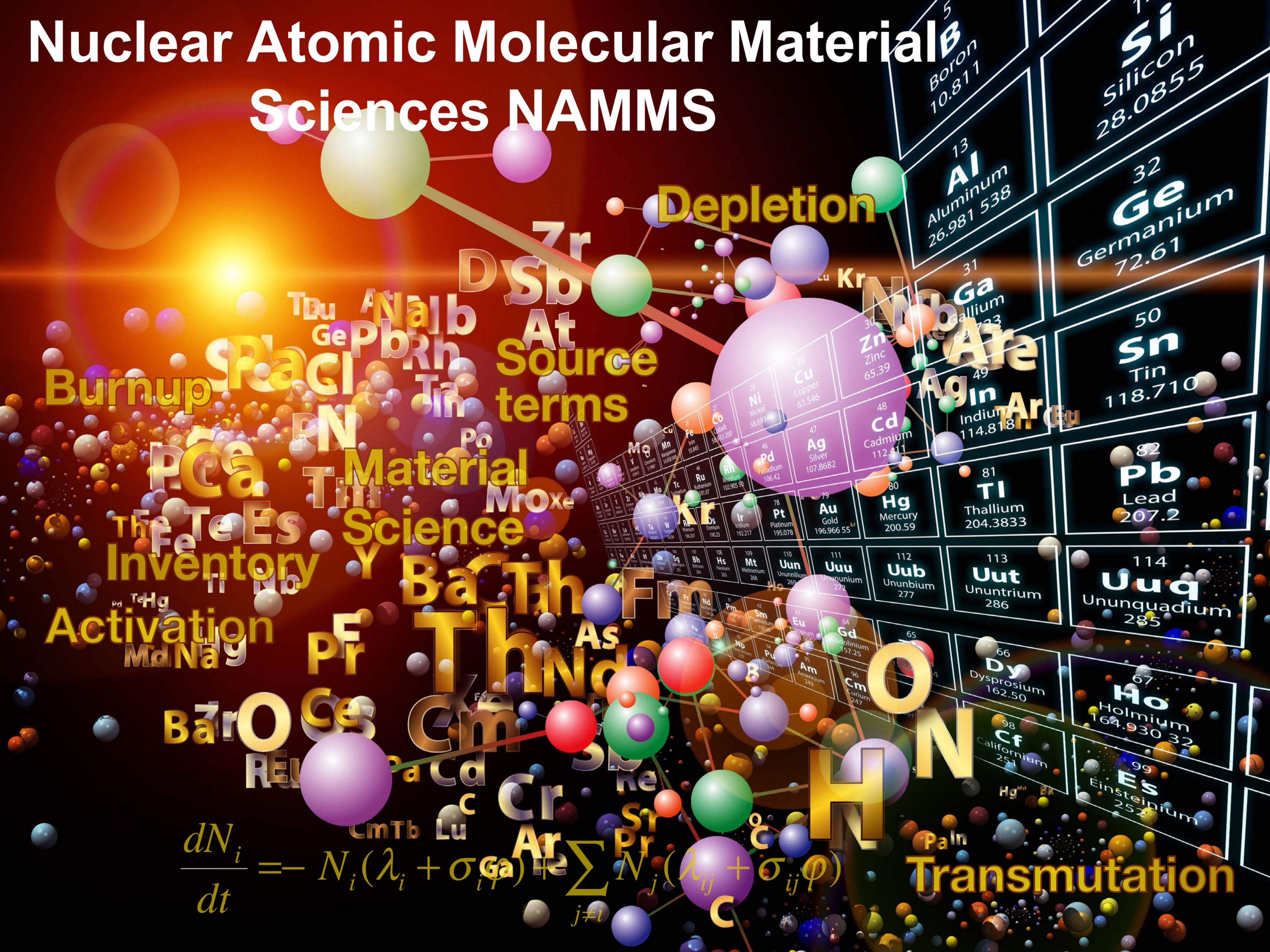
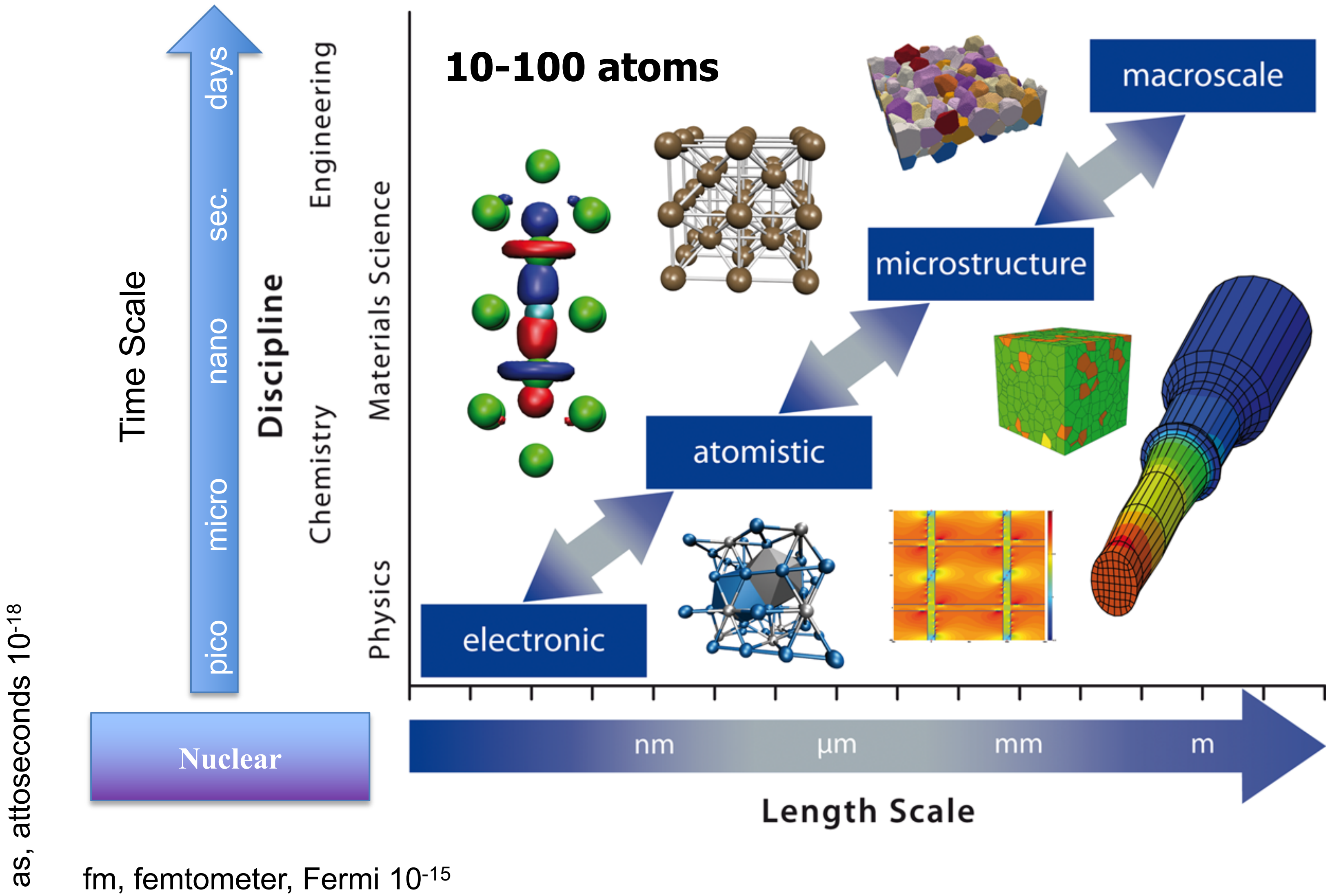
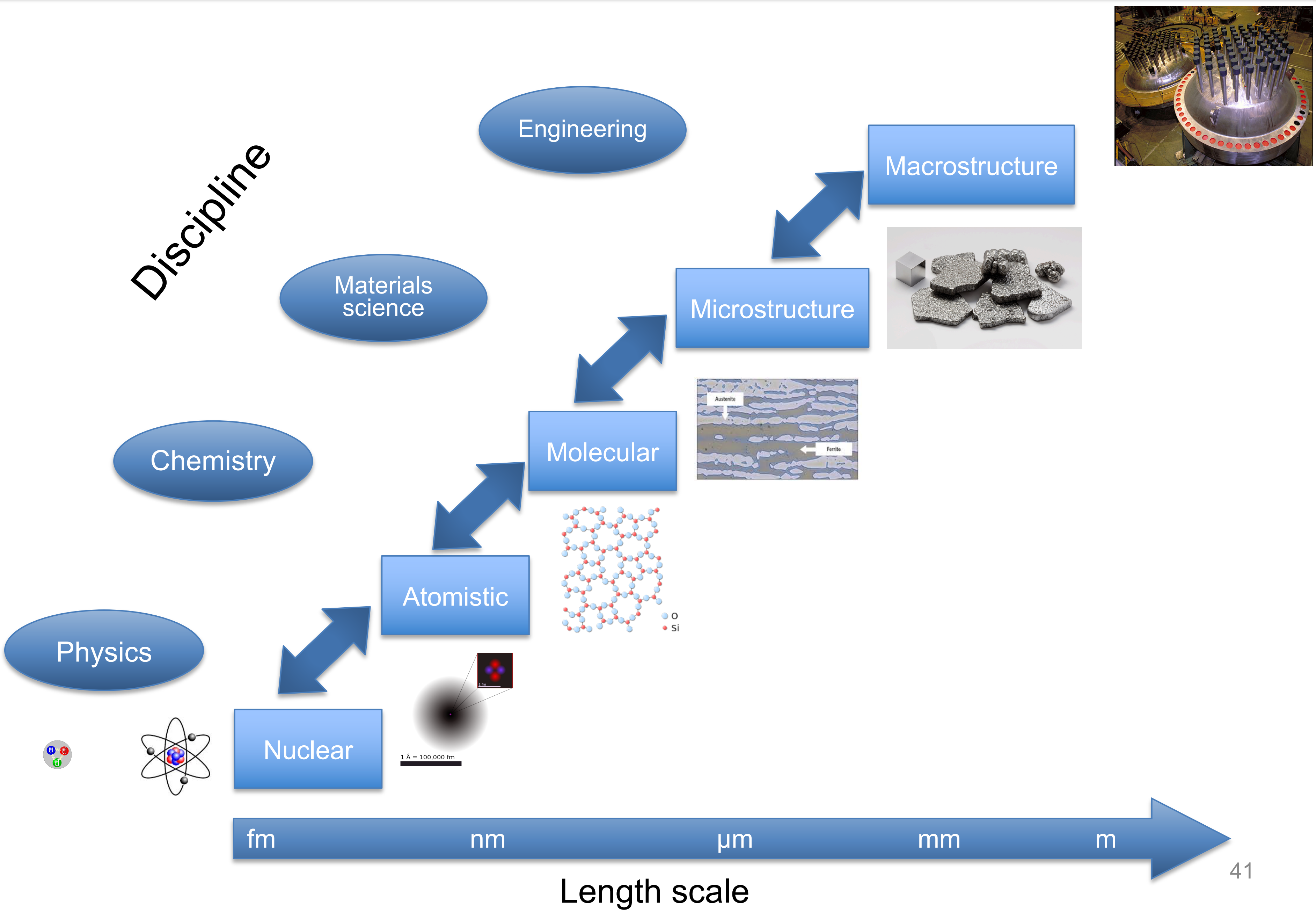


Multi-scales modelling

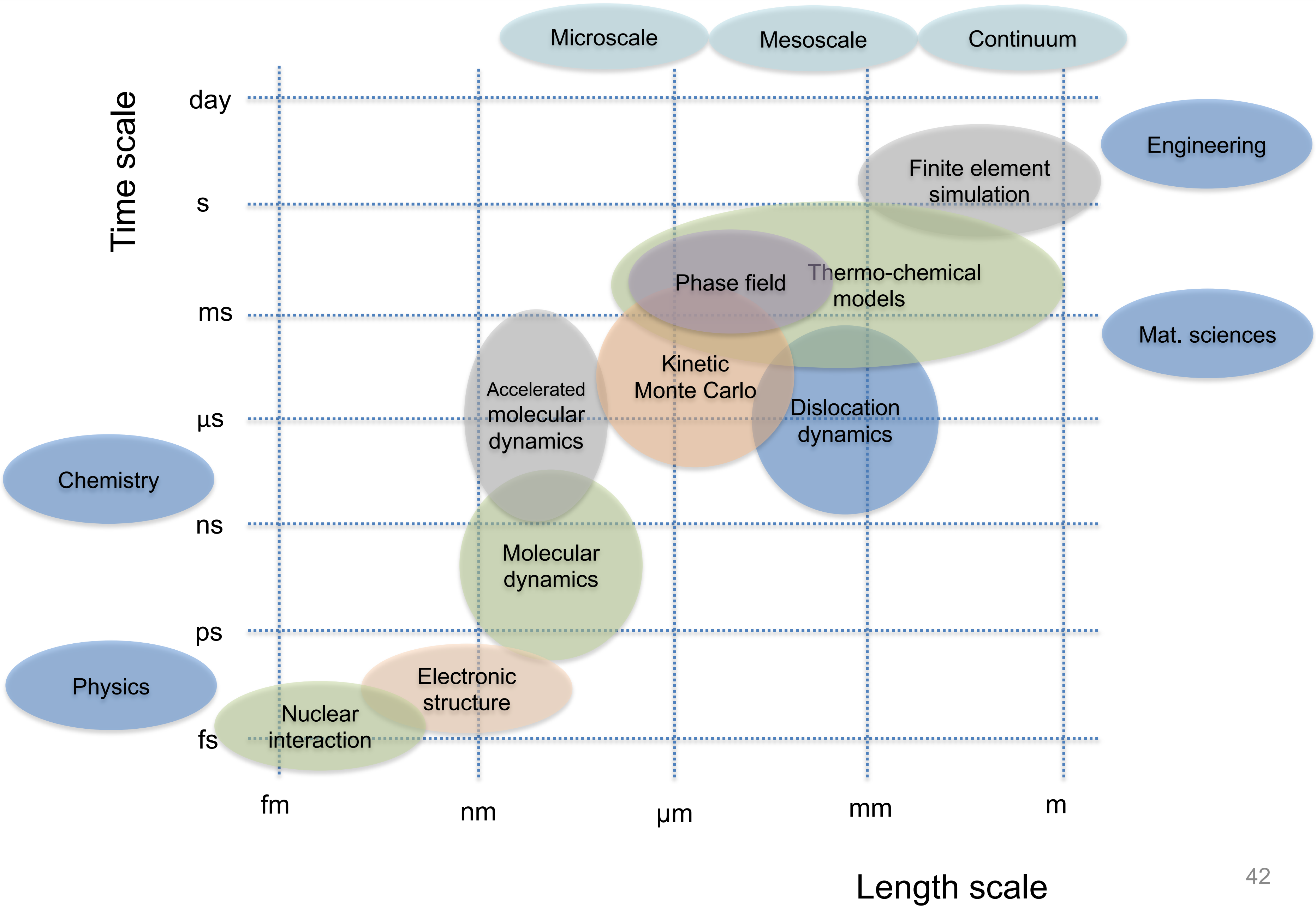


Traditionally different disciplines focus on different length scales. Multiscale modelling of materials across the length scales requires overcoming the borders between the disciplines for a seamless integration of the models on different length scales into one coherent multi-scale modelling framework (After D.G. Pettifor, 1991).

Multi-scales modelling



Multi-scales modelling



Nuclear inputs to multi-scales modelling