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A new benchmark database based on ASP foil irradiation campaigns?

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Outline

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- Background to experiments at ASP
- Early processing and analysis
- Detailed analysis for Mo
- Discussion

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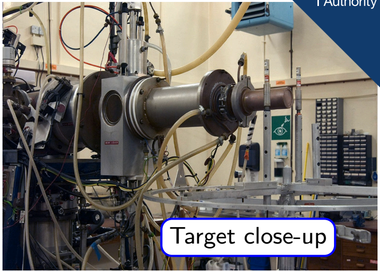
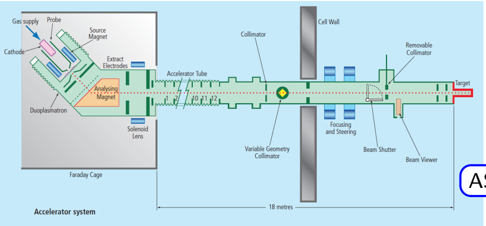
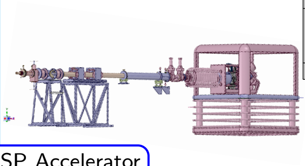
Nuclear data experiments at ASP

- Experiments in 2011-2015 by CCFE at ASP accelerator, AWE, Aldermaston
- Deuteron beam (up to 15 mA) onto a tritiated target producing 14 MeV DT neutrons at rate of up to 2.5×10^{11} n s^{-1}
- Variety of metal foils (< 1 g) irradiated in 300+ experiments
  - irradiation times from 1 min. to 1 hour
  - transfer to HPGe detector for γ-spectroscopy
  - count times from 5 min. to 1 day

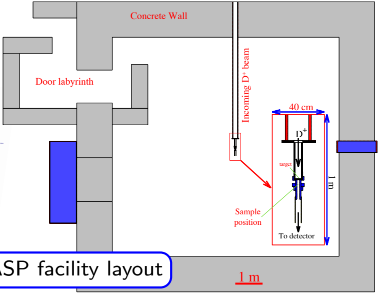


Target close-up





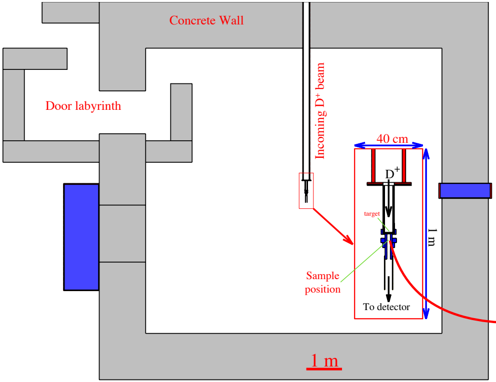
ASP Accelerator

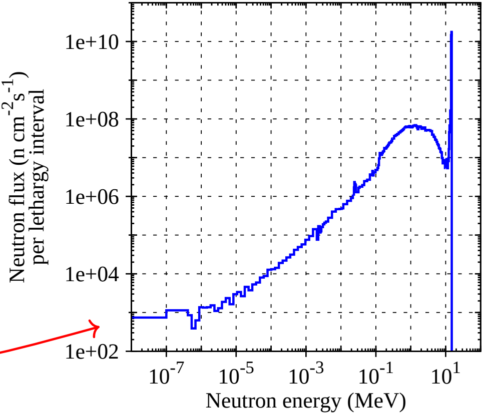


ASP facility layout

Neutron spectrum

- Characterisation of neutron spectrum at irradiation position using MCNP
  - Required for flux estimation using reference foils (see later)

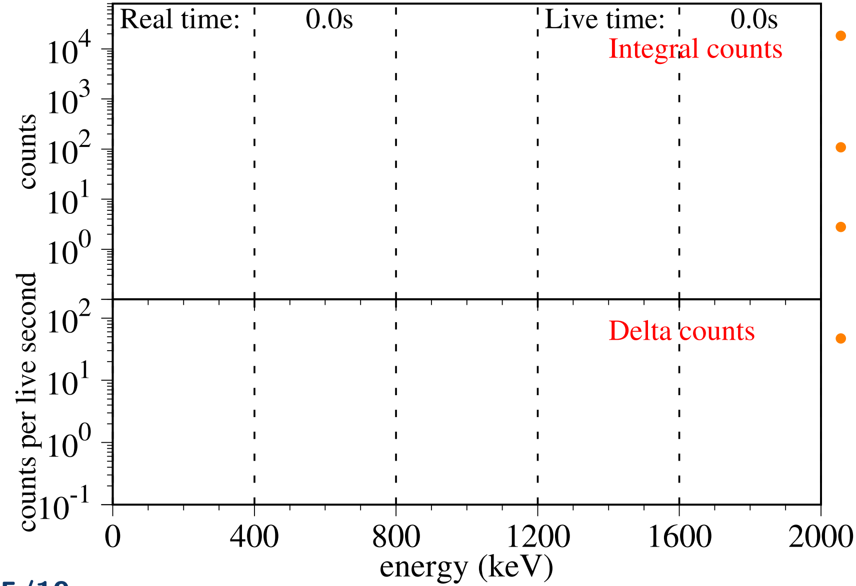




Lilley *et al.*, *Fus. Eng. Des.* **88** (2013) 2627

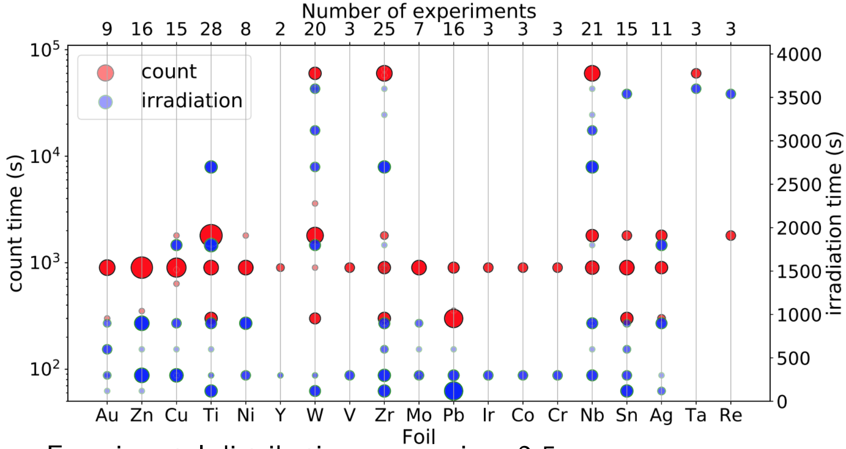
- Mainly 14 MeV, but with a distribution of moderated neutrons

Typical results: foil stack of Mo+Fe+Al



- Peaks correspond to γ-emissions of decaying radionuclides
- background associated with Compton scattering
- Time-sequence of γ-spec recordings captures nuclide decay
- can be used to calculate $t = 0$ activity more accurately compared to single snapshot in time (see later)

Size of experimental database



Experimental distribution - campaigns 2-5

Stainer *et al.*, *EPJ Web Conf.* **247** (2021) 09010

- 330+ experiments in 8 campaigns
- $\sim$20000 individual γ-spec
- samples foils covering wide number of metals of (fusion) interest, and also some alloys

(campaign 1 likely unusable due to absence of flux measurement foils)

Development of automated processing

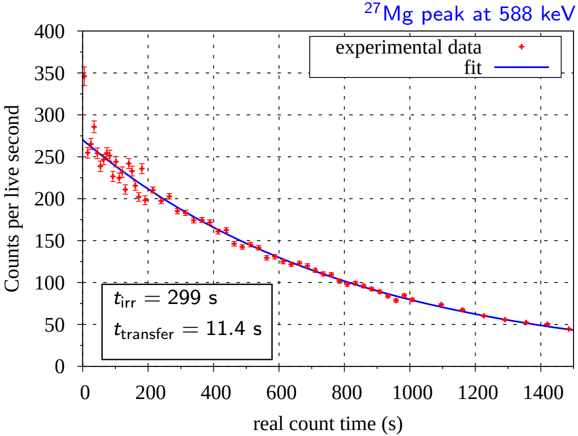
- Automated processing required to handle such a large amount of data with as little user-input as possible

- Features:
  - Handling of the complete set of γ-spectra from one experiment together
    $\longrightarrow$ takes advantage of the extra time-dependent data available from acquisitions
  - User-defined set of emission peaks identified and counts traced in time
    - including background and Compton corrections and handling of overlapping peaks
  - Least squares fitting of data to decay functions to obtain decay-corrected activities (directly comparable with inventory simulations)
  - Flux ϕ estimation from peak counts of well-characterised reactions (^{56}Fe(n,p)^{56}Mn, ^{27}Al(n,p)^{27}Mg, and ^{27}Al(n,α)^{24}Na)
  - Integral cross section σ calculation
  - Automatic plotting of data and decay fit
  - easily scriptable to process full experimental set (or subset) using identical inputs

First described in Gilbert *et al.*, *Nucl. Data Sheets* **119** (2014) 401 (but continuing to evolve)

Flux calculation

- ASP flux measurements typically performed using fission counters
- not reliable due to uncertainties in geometry correction for difference between foil and counter locations – hence Fe+Al foils from campaign 2 onwards



- Zr-Fe-Al foil stack; fit to ^{27}Mg

$$C(t) = C_0 \exp\left(-\frac{\ln 2}{T_{1/2}}\left[t + t_{\mathrm{transfer}}\right]\right)$$

$$C_0 = 273.5,\ T_{1/2}(^{27}\mathrm{Mg}) = 567.5 \text{ s}$$

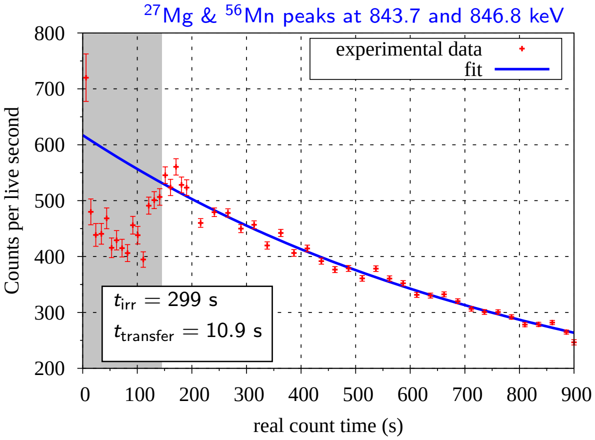
- $A_0 = 16418.8$ Bq, $\sigma = 0.057$ barn
- Flux estimated from:

$$A_0 = N_A \sigma \phi \left[1 - \exp\left(-\frac{\ln 2}{T_{1/2}} t_{\mathrm{irr}}\right)\right]$$

$$\phi_{\mathrm{est}} = 1.13 \times 10^9 \text{ n cm}^{-2} \text{ s}^{-1}$$

Flux calculation (2)

- Automated processing also accounts for overlapping peaks



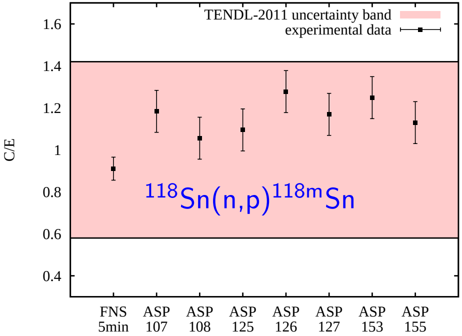
- Cr-Fe-Al foil stack - double decay fit to ^{27}Mg & ^{56}Mn $\left(T_{1/2}(^{56}\mathrm{Mn}) = 9296.6\ \mathrm{s}\right)$
- Dead-time issues due to activity in Cr
- But can be excluded from fit in processing tool
- from ^{27}Mg: $A_0 = 10875.8$ Bq, $\phi_{est} = 7.32 \times 10^8$ n cm^{-2} s^{-1}

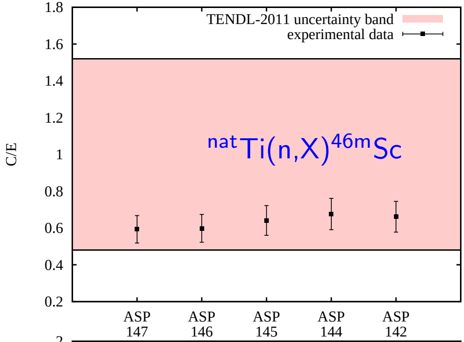
  from ^{56}Mn: $A_0 = 1452.7$ Bq, $\phi_{est} = 6.94 \times 10^8$ n cm^{-2} s^{-1}

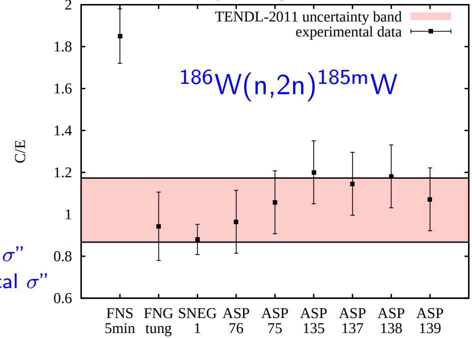
$$C(t) = C_0^1 \exp\left(-\frac{\ln 2}{T_{1/2}^1}\left[t + t_{transfer}\right]\right) + C_0^2 \exp\left(-\frac{\ln 2}{T_{1/2}^2}\left[t + t_{transfer}\right]\right)$$

Early integral σ measurements

- After first campaigns the processing system was used to calculate integral cross sections for several peaks for comparison to TENDL-2011
- Reasonably successful, but not taken further at the time







Packer *et al.*, *Nucl. Data. Sheets* **119** (2014) 173

C= "Calculated σ"
E= "Experimental σ"

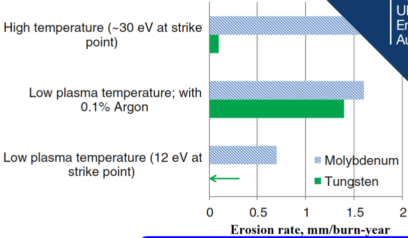
FNS – Japan; SNEG – St. Petersburg, Russia

Mo analysis

Gilbert *et al.*, *Nucl. Fusion* **60** (2020) 106022

Interest in Mo

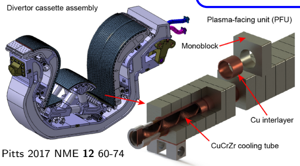
- Mo is a potential alternative to W in high neutron flux and high thermal load regions
  - the plasma facing components
- acceptable sputtering/erosion (Brooks et al. 2015)
- good thermal properties
  - Thermal conductivity $k \approx 100$ W m^{-1} K^{-1} (≈ 110 for W) at 1200K, $t_m = 2622°$C
- but has long-term activation issues that need validation and confirmation



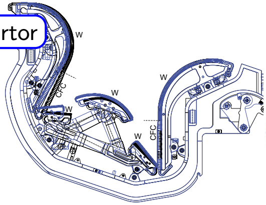
Plasma-facing erosion rate

Brooks et al. 2015 Nucl. Fus. **55** 043002

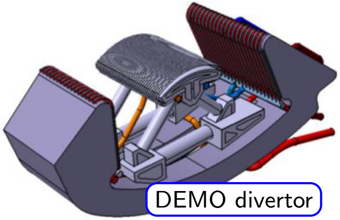
ITER divertor



Pitts 2017 NME **12** 60-74

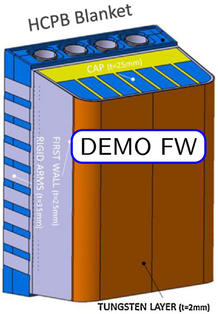


Pitts 2009 Phys. Scr. **T138** 014001



DEMO divertor

You 2017 Fus. Eng. Des. **124** 364-370

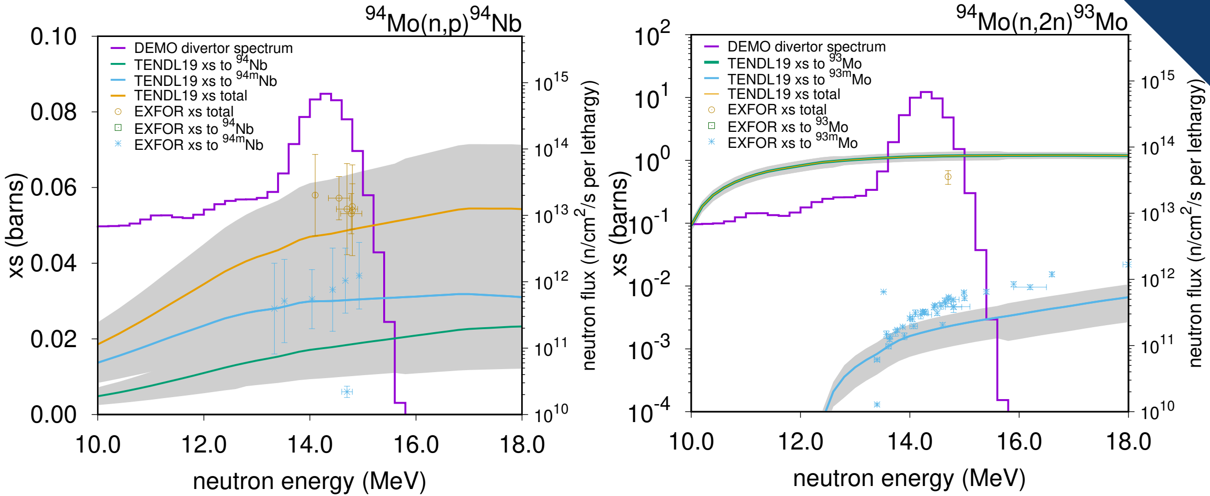


DEMO FW

Barrett 2016 Fus. Eng. Des. **109-111** 917-924

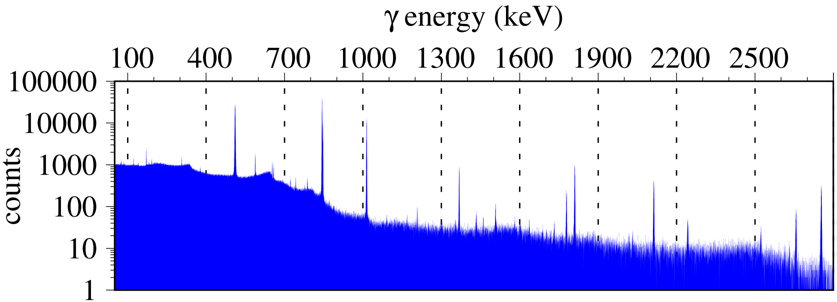
Examples of data deficiencies on Mo

- Not all reactions are well measured (particularly at 14 MeV)



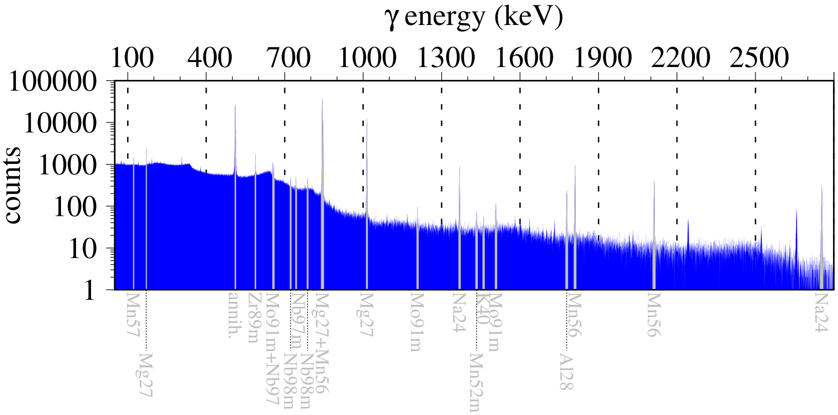
- Initial question: do ASP data cover these?

Mo experiments: peak identification



- 8 experiments included Mo (and necessary Fe+Al foils)
- all foils of stack in an experiment measured together
  - not ideal, but necessary for these short irradiation, short measurement & single detector experiments
- γ spectra contained several identifiable peaks

Mo experiments: peak identification



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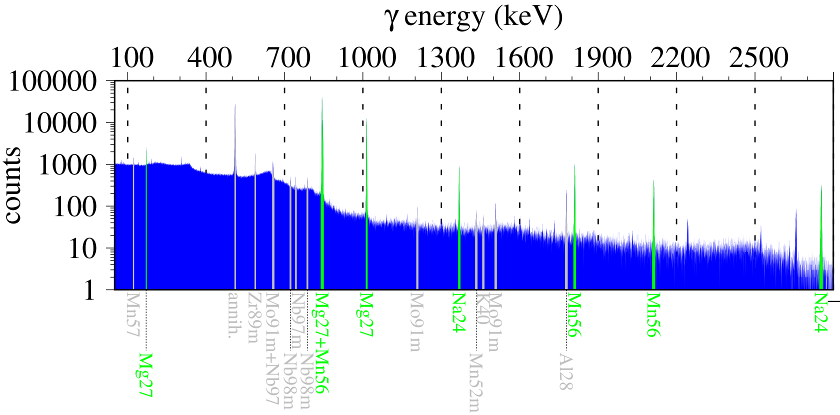
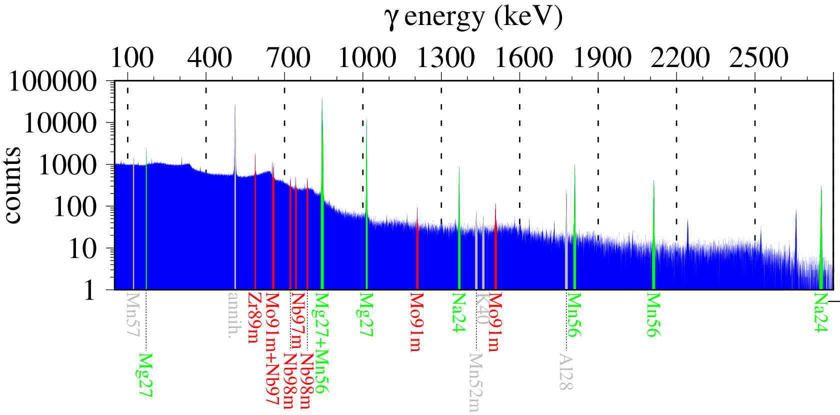
Mo experiments: peak identification



	$T_{1/2}$	Pathways
^{56}Mn	2.58 h	^{56}Fe(n,p)^{56}Mn
^{27}Mg	9.46 m	^{27}Al(n,p)^{27}Mg
^{24}Na	14.96 h	^{27}Al(n,α)^{24}Na

- γ spectra contained several identifiable peaks
- 9 associated with well-known reactions in Fe & Al

Mo experiments: peak identification

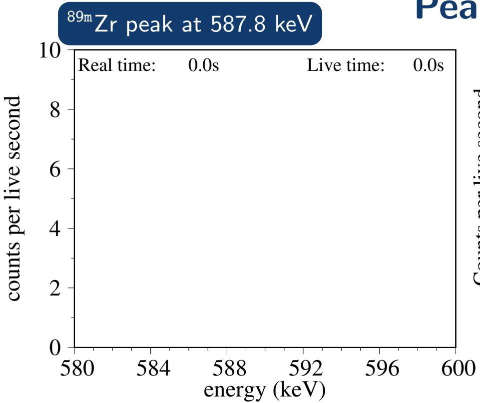


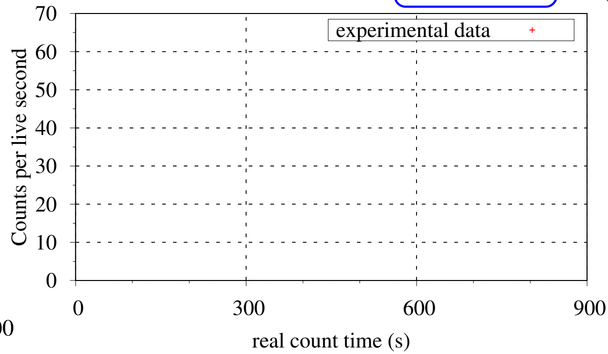
	$T_{1/2}$	Pathways
^{97m}Nb	53.0 s	^{97}Mo(n,p)^{97m}Nb
^{91m}Mo	1.08 m	^{92}Mo(n,2n)^{91m}Mo
^{89m}Zr	4.13 m	^{92}Mo(n,α)^{89m}Zr
^{98m}Nb	51.30 m	^{98}Mo(n,p)^{98m}Nb
^{97}Nb	1.23 h	^{97}Mo(n,p)^{97}Nb

- γ spectra contained several identifiable peaks
- 9 associated with well-known reactions in Fe & Al
- & 8 peaks corresponding to γ-emissions from radionuclides generated in Mo

Peak count tracking

experiment 82





- (Compton) background subtracted from the peak Δ-counts in each time-step
- Remaining peak area provides counts per live second (i.e. accounting for detector dead-time)

~9000 total background-corrected counts during 15-minute acquisition

Peak count tracking + fitting

^{89m}Zr peak at 587.8 keV

experiment 82

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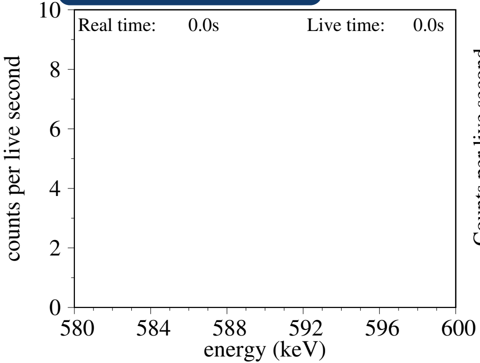

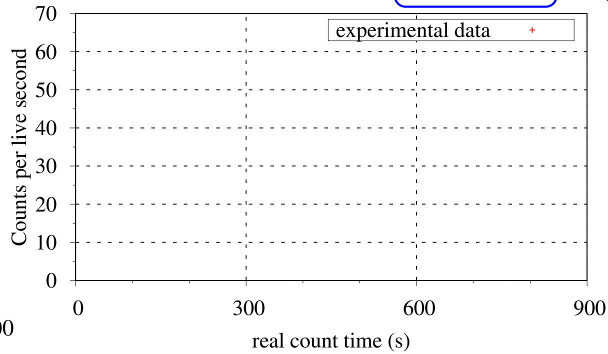

- Data fit to give C_0 count rate at end of irradiation
- $C(t) = C_0 \exp\left(-\frac{\ln 2}{T_{1/2}}\left[t + t_{\text{transfer}}\right]\right)$
- t is real time, C is counts per second
- t_{transfer} is transfer time between end of irradiation and start of acquisition (typically ~ 10 s via pneumatic rabbit tube)

$C_0 = 47\ \text{s}^{-1}$

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Experiment vs. simulation

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- Experimental activity at end of irradiation:

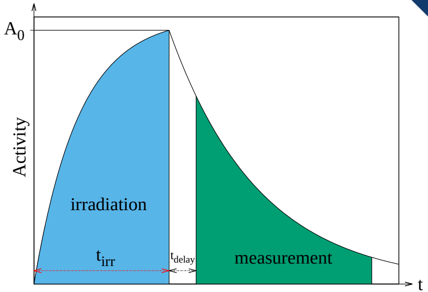
$$A_0 = \frac{C_0}{D^{\mathrm{eff}}(E_p^\gamma) I_p}$$

D^{eff} – calibrated HPGe detector efficiency

E_p^γ – γ energy of peak

I_p – peak intensity based on decay branching ratios

- total flux ϕ averaged from ^{27}Mg, ^{56}Mn, and ^{24}Na peaks



- Calculated A_0 activity for $^{91\mathrm{m}}$Mo, $^{98\mathrm{m}}$Nb, $^{97\mathrm{m}}$Nb, $^{89\mathrm{m}}$Zr, ^{97}Nb
  - obtained from FISPACT-II simulations using TENDL-2019 nuclear cross sections and averaged ϕ value

$$A_0 = N\sigma\phi(1 - \exp^{-t_{\mathrm{irr}}\lambda})$$

N – number of parent atoms (e.g. ^{56}Fe)

λ – radionuclide decay constant

t_{irr} – irradiation time

- ratio between calculated and experimental activities ("C/E") – measure of simulation quality

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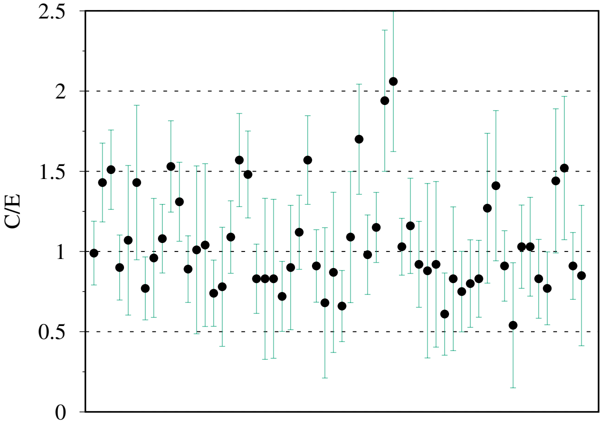
C/E results



- Experiments were not optimised for these measurements and there are high uncertainties due to high background (leading to poor statistics in some cases)
- But C/E values are reasonable
- calculated values generally within a factor of 2 of experiment (good for γ-spec)

All data provided in paper:
Gilbert *et al.*, *Nucl. Fusion* **60** (2020) 106022

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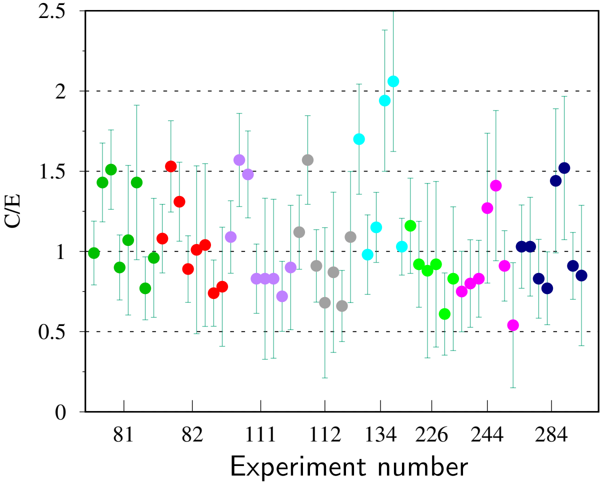
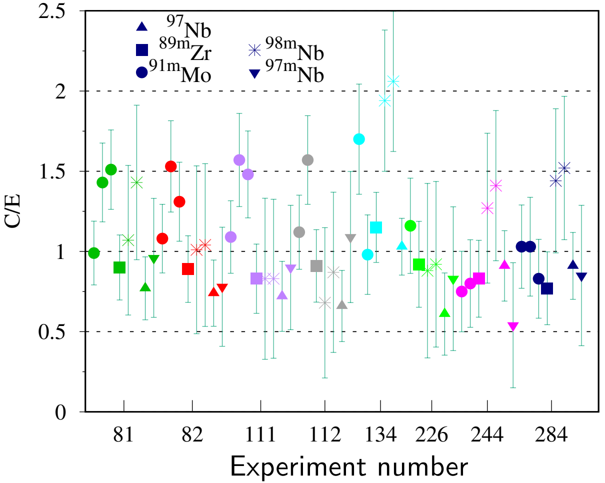
C/E results



- Some experiments appear to be better than others
  - Experiment 134 bad due to interference from Ti foils

All data provided in paper:
Gilbert *et al.*, *Nucl. Fusion* **60** (2020) 106022

C/E results



- Predictions for some radionuclides better than others (but not much in it)
- Open questions:
  - Statistics too poor for xs measurement?
  - But good enough for scoping benchmark?

All data provided in paper:
Gilbert *et al.*, *Nucl. Fusion* **60** (2020) 106022

coverage limitations

- Nuclides explored

	$T_{1/2}$	Pathways	
^{97m}Nb	53.0 s	^{97}Mo(n,p)^{97m}Nb	ASP
^{91m}Mo	1.08 m	^{92}Mo(n,2n)^{91m}Mo	
^{89m}Zr	4.13 m	^{92}Mo(n,α)^{89m}Zr	
^{98m}Nb	51.30 m	^{98}Mo(n,p)^{98m}Nb	
^{97}Nb	1.23 h	^{97}Mo(n,p)^{97}Nb	
^{91}Mo	15.49 m	^{92}Mo(n,2n)^{91}Mo	FNS, Japan
^{99}Mo	2.7 d	^{100}Mo(n,2n)^{99}Mo	
^{95}Nb	35 d	^{95}Mo(n,p)^{95}Nb	
^{91m}Nb	61 d	^{92}Mo(n,np)^{91m}Nb	
^{91}Nb	680 years	^{92}Mo(n,np)^{91}Nb	fusion reactor
		^{92}Mo(n,2n)^{91}Mo(β^+)^{91}Nb	
^{93}Mo	3500 years	^{92}Mo(n,γ)^{93}Mo	
		^{94}Mo(n,2n)^{93}Mo	
^{94}Nb	20000 years	^{94}Mo(n,p)^{94}Nb	
		^{95}Mo(n,np)^{94}Nb	
^{99}Tc	210000 years	^{98}Mo(n,γ)^{99}Mo(β^-)^{99}Tc	
		^{100}Mo(n,2n)^{99}Mo(β^-)^{99}Tc	

- Short irradiation, short measurement, small sample experiments
  - but showed that simulations with modern data do well at predicting short-lived activity and decay-heat on Mo
  - important for post-operation maintenance and remote handling in a fusion reactor
- no data on the production of important long-lived nuclides

Gilbert *et al.*, *Nucl. Fusion* **60** (2020) 106022

Summary, status and future

- Over several years, a large database of γ-spectroscopy measurements were taken for a variety of metal foils irradiated for short periods in the 14 MeV ASP accelerator at AWE
- some limitations, which might prevent and individual experiment being used for high-accuracy cross section measurements
- but as a whole, the data, if treated in a consistent way, could be utilised as a scoping benchmark data-set to test inventory simulations and nuclear libraries for several key elements
- will only test production of short half-life radionuclides

Future

- repeat Mo analysis for other elements to mature those data subsets into usable benchmarks (some work already begun for the 20+ experiments on W)
- eventual release of processed data with inventory simulation input parameters as a benchmark suite

Thank you