Paul Scherrer Institut

Wir schaffen Wissen – heute für morgen

Activation and Radiation Damage of Components in the Environment of Proton Accelerators

Daniela Kiselev

Contents

1. Activation of material

- Mechanism for activating material
- Time evolution of the activity
- Calculation methods
- Examples of Activation
- Composition of radioactive waste at accelerators

2. Radiation damage of components

- Influence on physical and mechanical properties
- Mechanism for producing radiation defects
- Definition of DPA, Displacement cross section
- Defect effiency
- Calculation methods
- Practical example: - target hull of ESS (European Spallation Source)
  - Cu-collimator at PSI

3. Summary

When is a material radioactive?

1. Activity: decays/sec, unit: Bq

$$\sum_i \frac{A_i}{R_i} > 1$$

A_i: specific activity [Bq/g]
R_i: exemption limit
given in the radioprotection regulation

OR

2. Dose rate D: ~ absorbed energy/kg x biological factor, unit: [Sv/h]
measure for the damage to human tissue

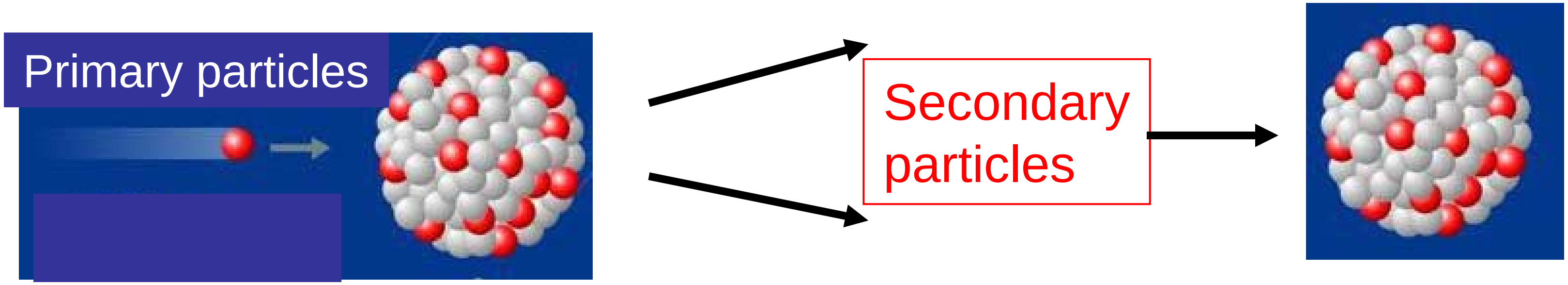
$D > 0.1\ \mu$Sv/h

OR

3. Surface contamination:
   - > 1 Bq/cm^2 in case of unidentified β- and γ-emitters
   - > 0.1 Bq/cm^2 in case of unidentified α-emitters
   - > CS-value (given in regulation) for specific isotope

How does the material get activated?

Reaction of beam with atomic nuclei of the component:



Primary particles

Secondary particles

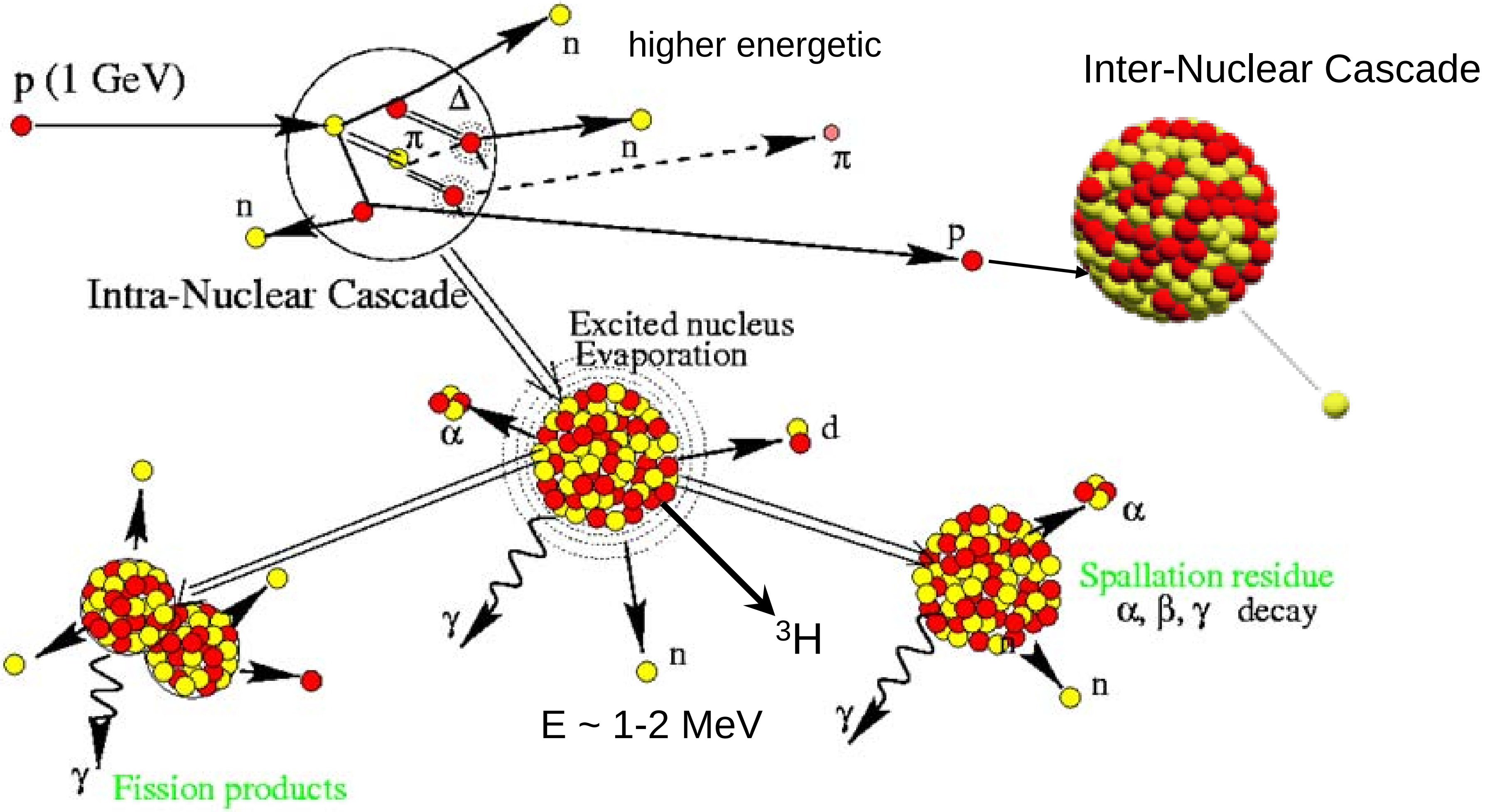
→ Nuclear reactions: Change of the number of protons and neutrons
→ Transmutation into other isotopes, often radioactive

Production rate (Isotope/s):

$$P_Y = N_A n_x \int \frac{d\phi_x(E)}{dE} \cdot \sigma_{xA \to Y}(E) \cdot dE$$

Dependent on
- production cross section σ for xA → Y + secondaries
- energy distribution of the projectile per area (fluence): Φ [1/cm^2]
- number of atomic nuclei : N_A
- number of particles per sec.: n_x

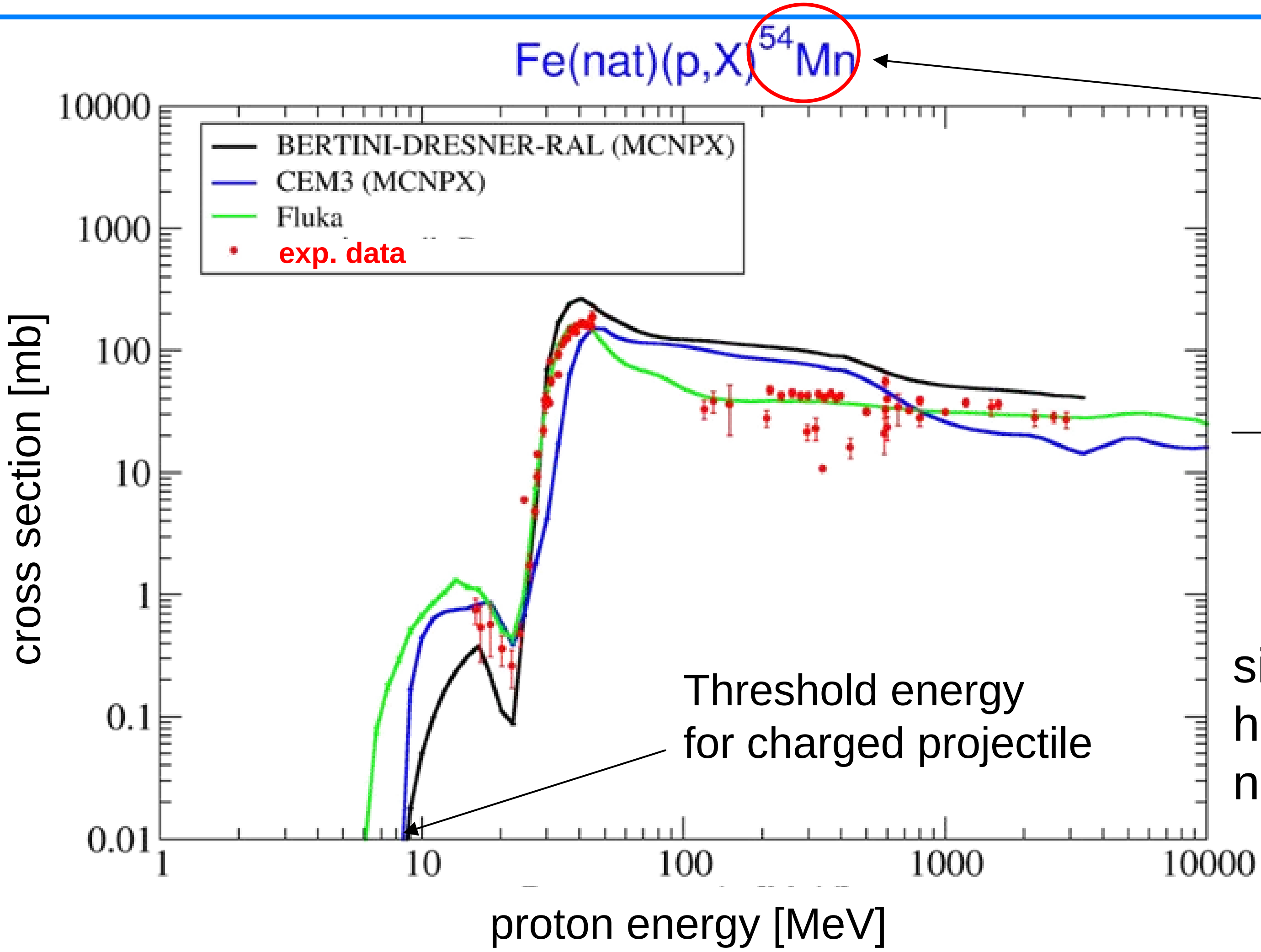
Nuclear processes for direct irradiation (e.g. in target)



Example:
Pb-Target: ~ 20 n/p,

complicated coupled processes
→ Monte Carlo Simulation (MCNPX)

Production cross section



residual nuclei

~ constant

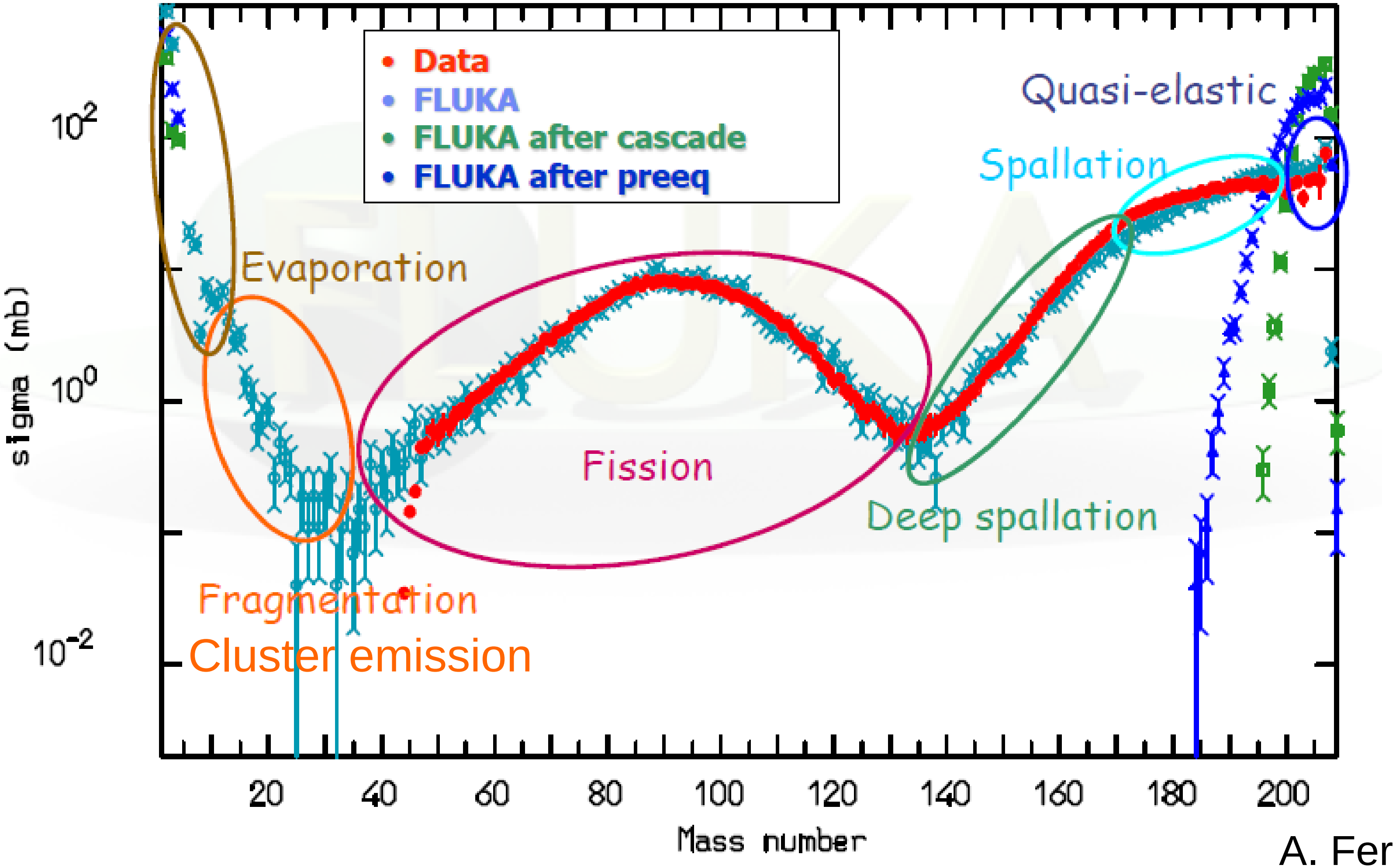
Threshold energy
for charged projectile

similar for
high energetic
neutrons (> 20 MeV)

Models are needed to describe all reaction channels, including the production of secondaries and their reactions

Production cross section for residual nuclei

1 A GeV ^{208}Pb + p reactions Nucl. Phys. A 686 (2001) 481-524



A. Ferrari, CERN

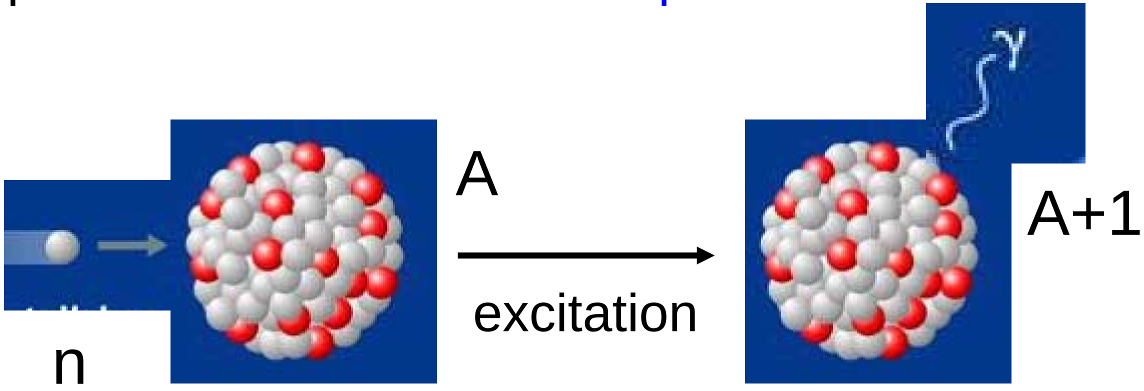
Nuclear reactions in the environment (e.g. shielding)

- mainly secondary particles
- charged particles already slowed down or absorbed
- neutral particles are left: e.g. neutrons

→ Energy loss due to many collision (particularly with light nuclei)
= Moderation, thermalization

→ the shape of the neutron spectrum does not vary much with thickness

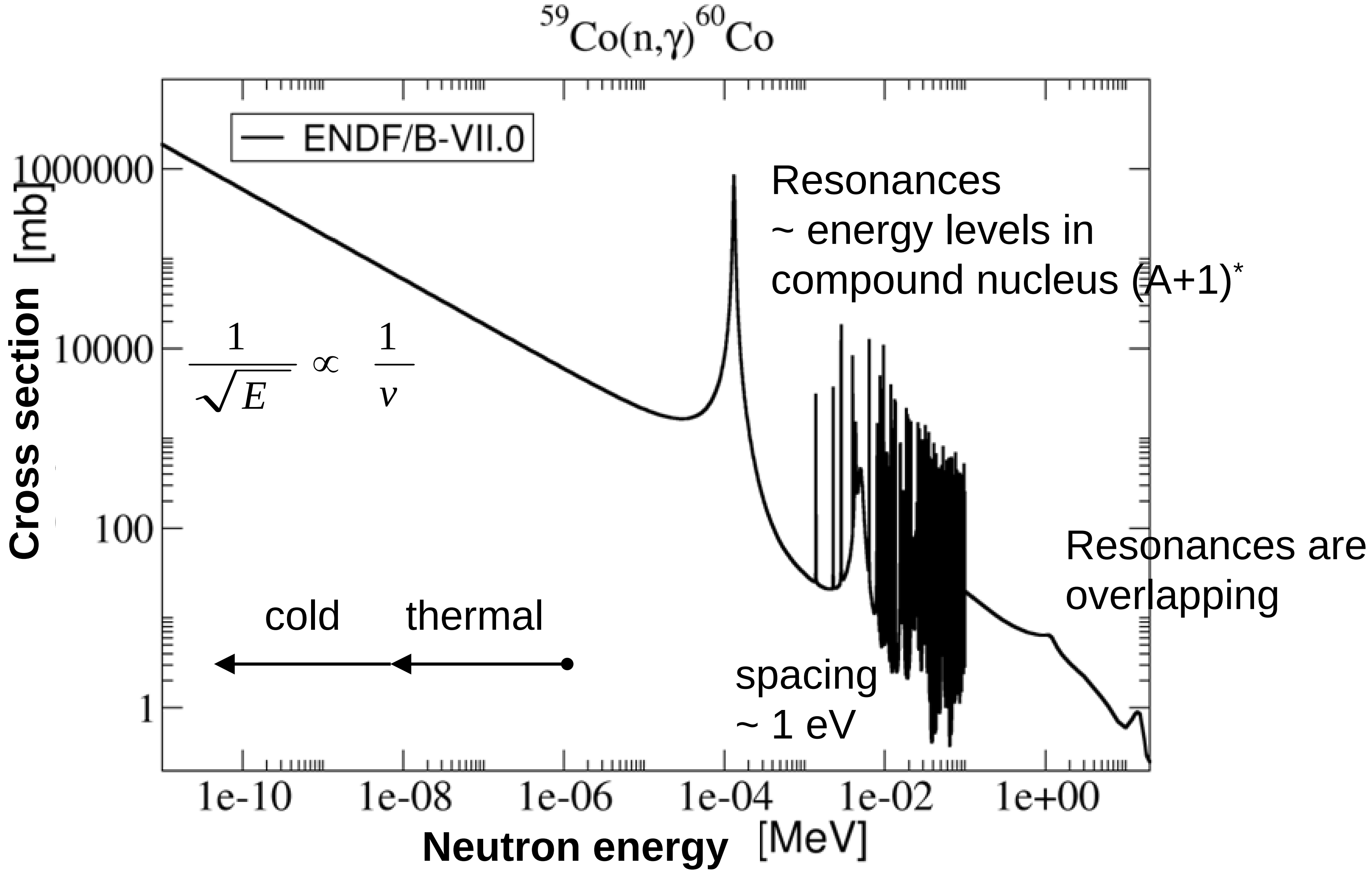
Important reaction: neutron capture



e.g. $^{59}\mathrm{Co}(n,\gamma)^{60}\mathrm{Co}$, $^{107}\mathrm{Ag}(n,\gamma)^{108m}\mathrm{Ag}$

Production cross section using neutrons < 20 MeV

→ large cross sections at thermal energies



$$\frac{1}{\sqrt{E}} \propto \frac{1}{v}$$

Resonances
~ energy levels in
compound nucleus $(A+1)^*$

spacing
~ 1 eV

Resonances are
overlapping

Time evolution: Build-up and Decay of isotopes

$$A(t) = -\frac{dN(t)}{dt} \underset{\uparrow}{=} \lambda N(t) = \lambda \, \mathrm{N}_{\mathrm{o}} e^{-\lambda t}$$

$$N(t) = N_o e^{-\lambda t}$$

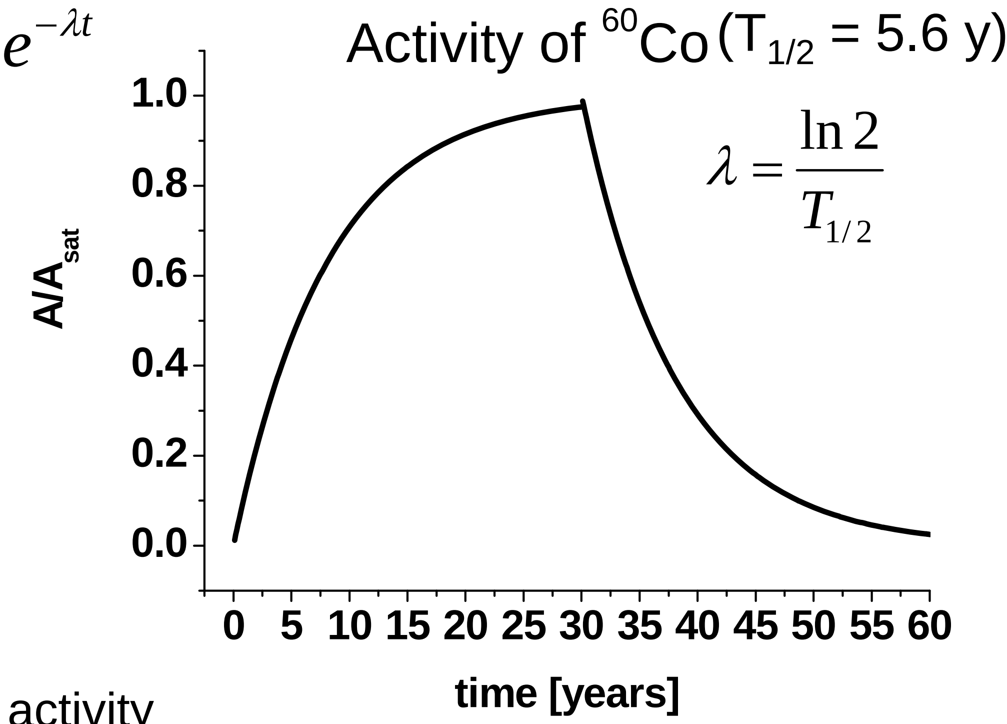
The simple case: 1 radioisotope

$$\frac{dN(t)}{dt} = P - \lambda N(t)$$

$$A(t) = P\left(1 - e^{-\lambda t}\right)$$

$$t \rightarrow \infty : A \rightarrow P = A_{sat}$$

P: production rate A_{sat}: saturation activity


Activity of ^{60}Co $(T_{1/2} = 5.6\ \mathrm{y})$

$$\lambda = \frac{\ln 2}{T_{1/2}}$$

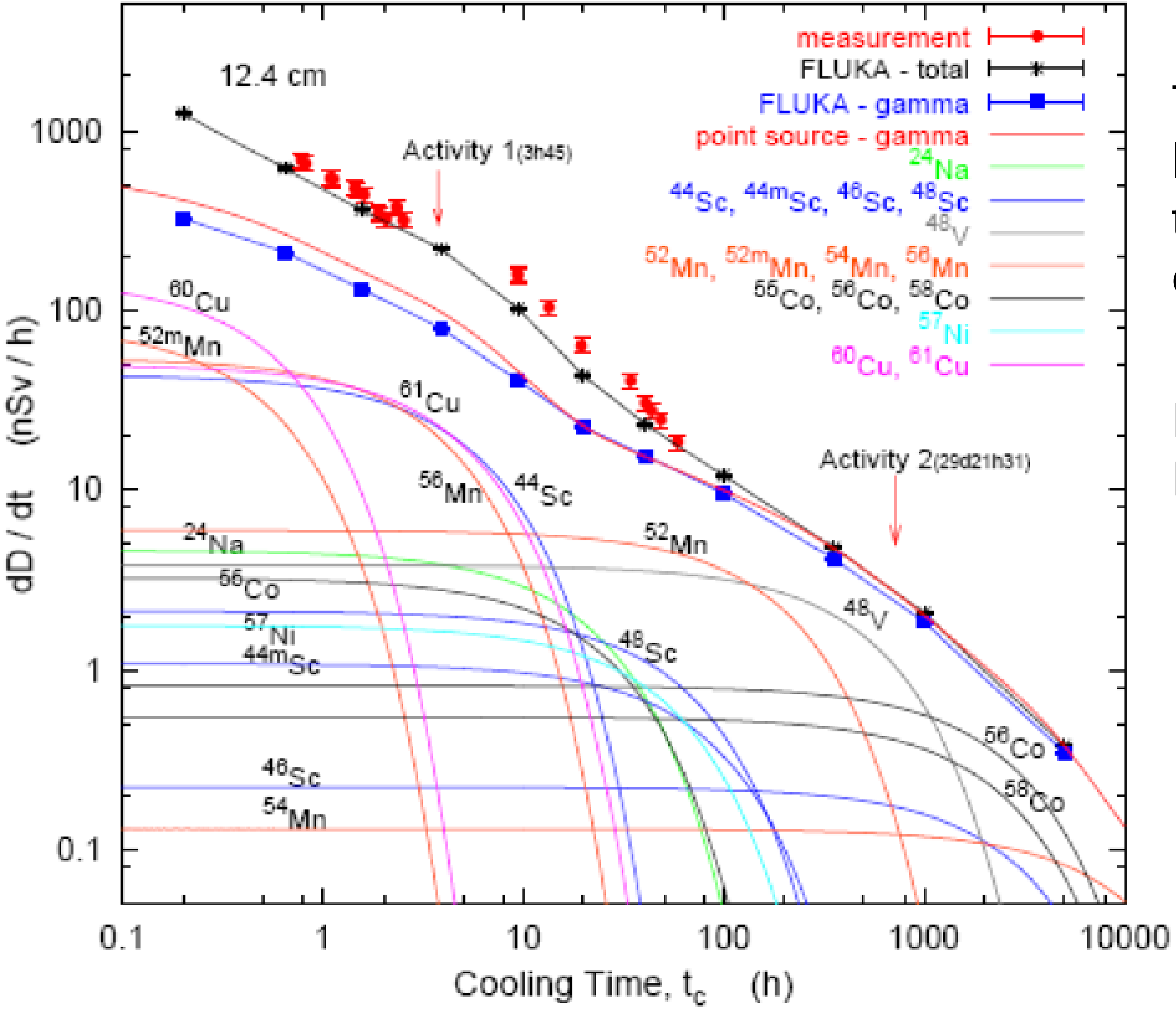
For many isotopes contribution via decay chain: Bateman equation

$$\frac{dN_m(t)}{dt} = \underbrace{\sum_{k \neq m} N_k(t) \varphi_x \sigma_{k \rightarrow m}}_{\text{production}} - \underbrace{N_m(t)\left(\lambda_m + \varphi_x \sigma_{m+x}^{abs}\right)}_{\text{loss due to decay + absorption}}$$

φ_x : flux [cm^{-2}s^{-1}]

Relevant isotopes

Proton irradiation of copper



The isotope with the largest contribution to the γ-dose rate changes with time.

Nuclide inventory Depends also on the Irradiation time

talk from L. Ulrici, CERN

The need for calculations

- for disposal as radioactive waste
  - Nuclide inventory needed before disposal (required by authorities)
  - periodical validations are required: comparison to experimental data

- for future (planned) installations/facilities:
  - estimation of the amount of total waste after operation
  - dose rates needed for construction of shielding

- for repair/dismantling → planning of work procedures
  dose rate estimation needed

Measurements are needed for checks:

- γ's in Ge-detector
- chemical + quantitative Analysis
  + β measurement in Liquid Scintillator
- Accelerator mass spectrometry (AMS) for isotopes with long life time
- α: dissolution out of the solid, surface detectors
  → time elaborate, not all isotopes can be measured

Calculation of the Activation: Method

case 1: due to direct irradiation

Monte Carlo particle transport program: n,p,γ,α,π,d,^{3}H....

Input:

- dedicated geometry
- material compositions
- cross sections: for n < 20 MeV (e.g. ENDF-B-VI.6)
- models for all other reactions

Output: dependent on the code

MCNPX, PHITS

- n-fluxes (E<20 MeV)
- residual nuclei production rates

FLUKA, MARS

built-in buildup & decay codes

coupling to
buildup & decay codes

Cinder'90

DCHAIN

SP-Fispact

Orihet3

Input:

- irradiation and cooling history
- n-cross sections built-in or external library
- decay properties of isotopes

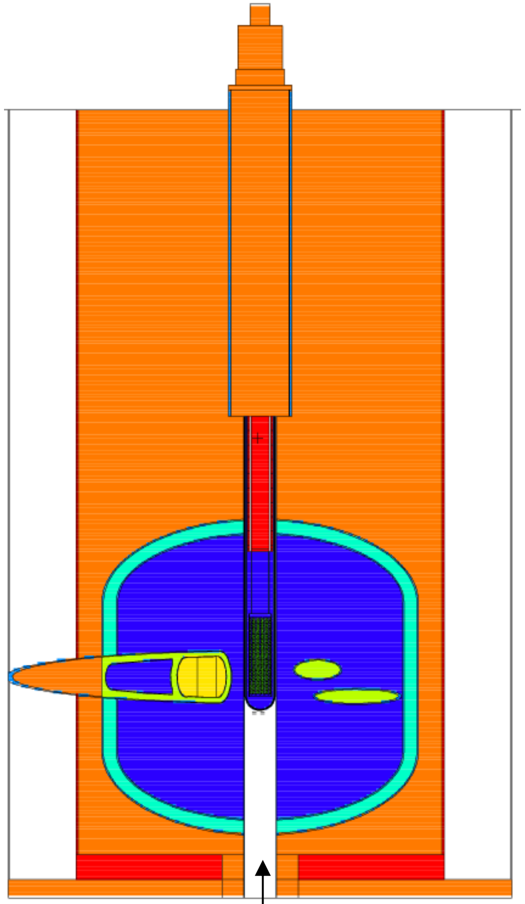
Output::

- nuclide inventory
- residual (or remanent) dose rate

Example: Activation at the SINQ-Target 3 (PSI)

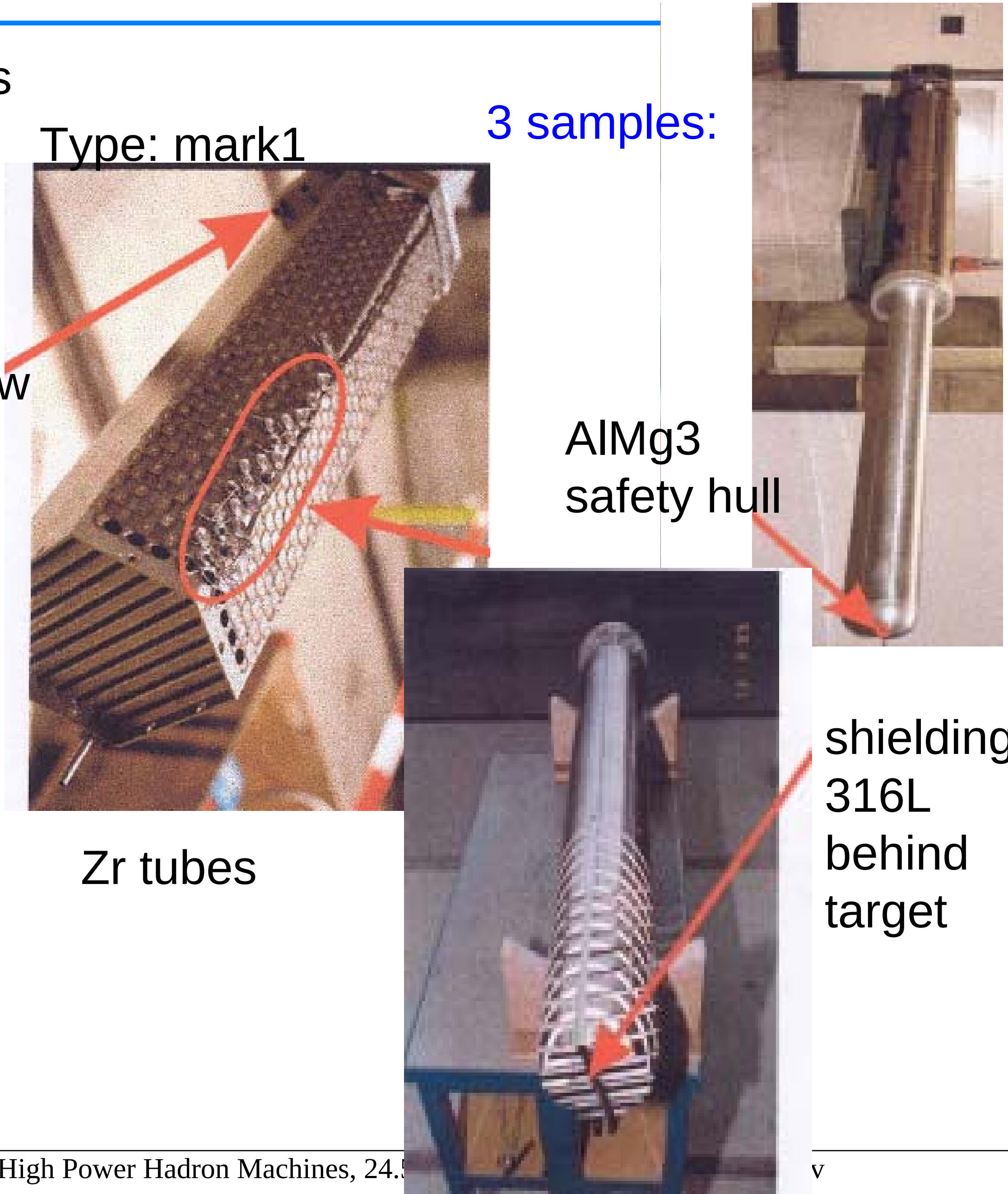
irradiation: 6.77 Ah for 2 years

geom. model in MCNPX:



p-beam

3 samples:



Type: mark1

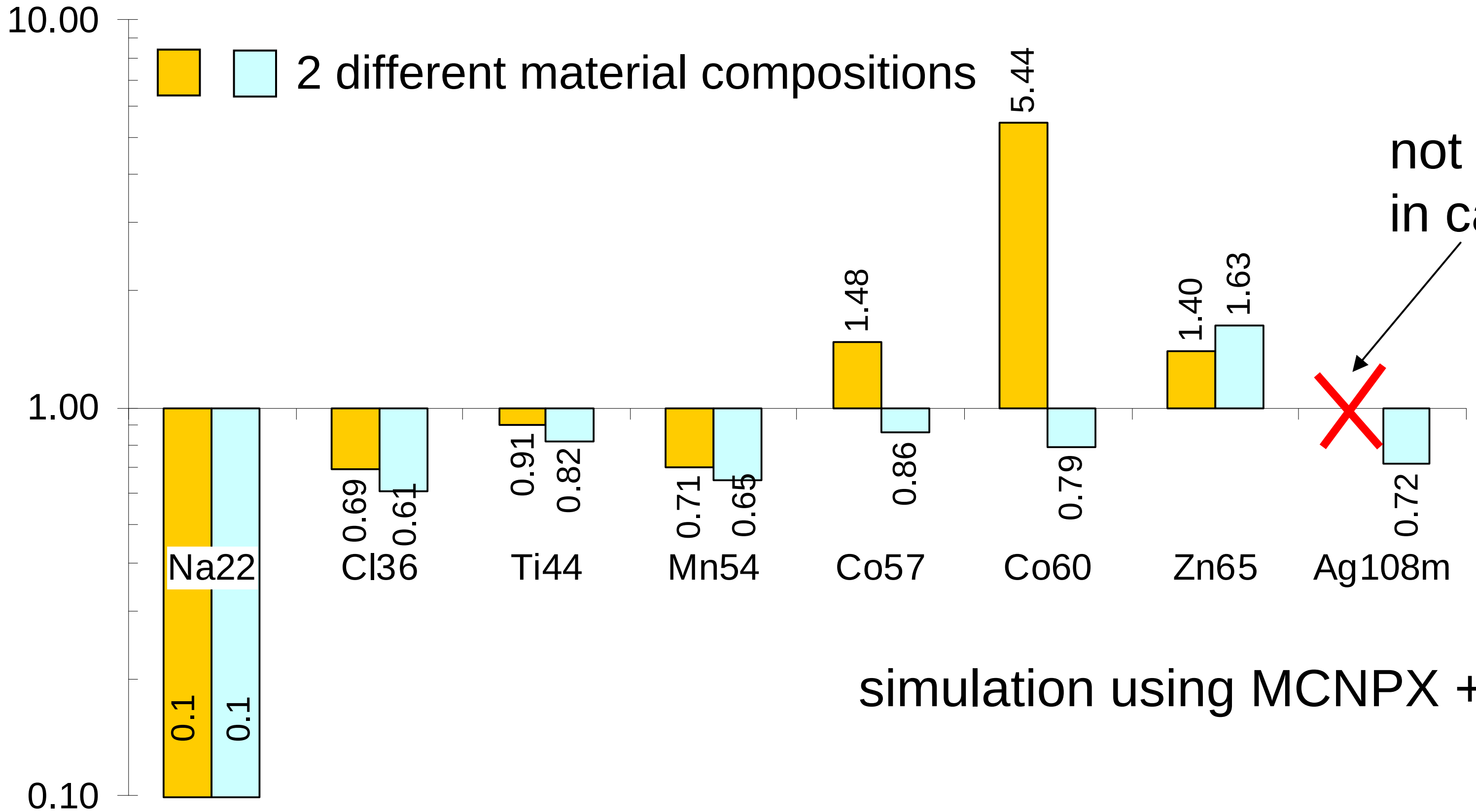
Zr screw

Zr tubes

AlMg3 safety hull

shielding 316L behind target

Results for $AlMg_3$ safety hull: exp. / calc. activity



not obtained in calculation

simulation using MCNPX + Cinder'90

remarks:

- overall good agreement with MCNPX+Cinder'90
- ^{22}Na: Data/SP-Fispact = 0.72
- the right material composition (including impurities) is important!

Calculation Methods

Case2: further away from loss points, e.g. behind shielding

remember:

- neutral particles (especially neutrons) are most relevant
- the shape of the neutron spectrum is almost constant (amplitudes varies)

→ simplification:

- n-spectra have to be determined once for a larger region (via measurement or MC-simulation) → Φ
- include cross section library → σ
- defining material compositions → N
- collecting operational data (irradiation periods, currents)

➔ $P \hat{=} \phi \cdot \sigma \cdot N$ + coupling to buildup and decay code

➔ nuclide inventory, normalized to measured surface dose rate

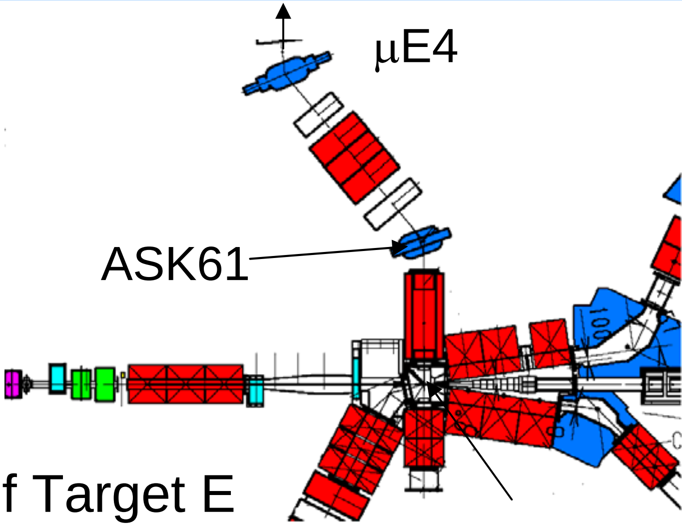
Code at PSI: PWWMBS,

at CERN: Jeremy, using spectra for neutrons, protons, pions and photons

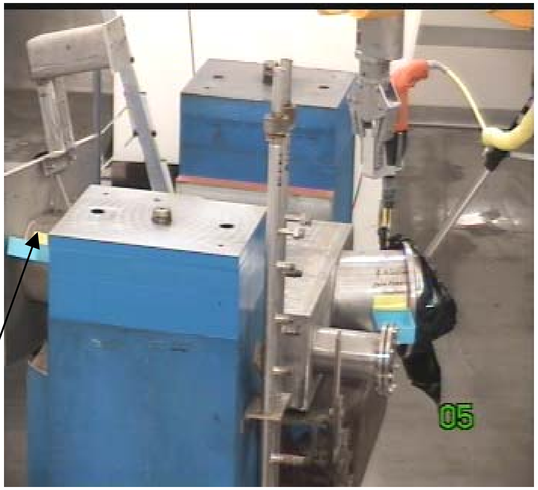
Example: Activation at the μE4 beam line (PSI)

2004: Several samples were taken from
- bending magnet ASK61
- shutter behind ASK61
- shielding around shutter

→ components are not directly irradiated
→ calculation of the activity with PWWMBS
use representative n-flux spectrum in vicinity of Target E



bending magnet ASK61 (stainless steel)

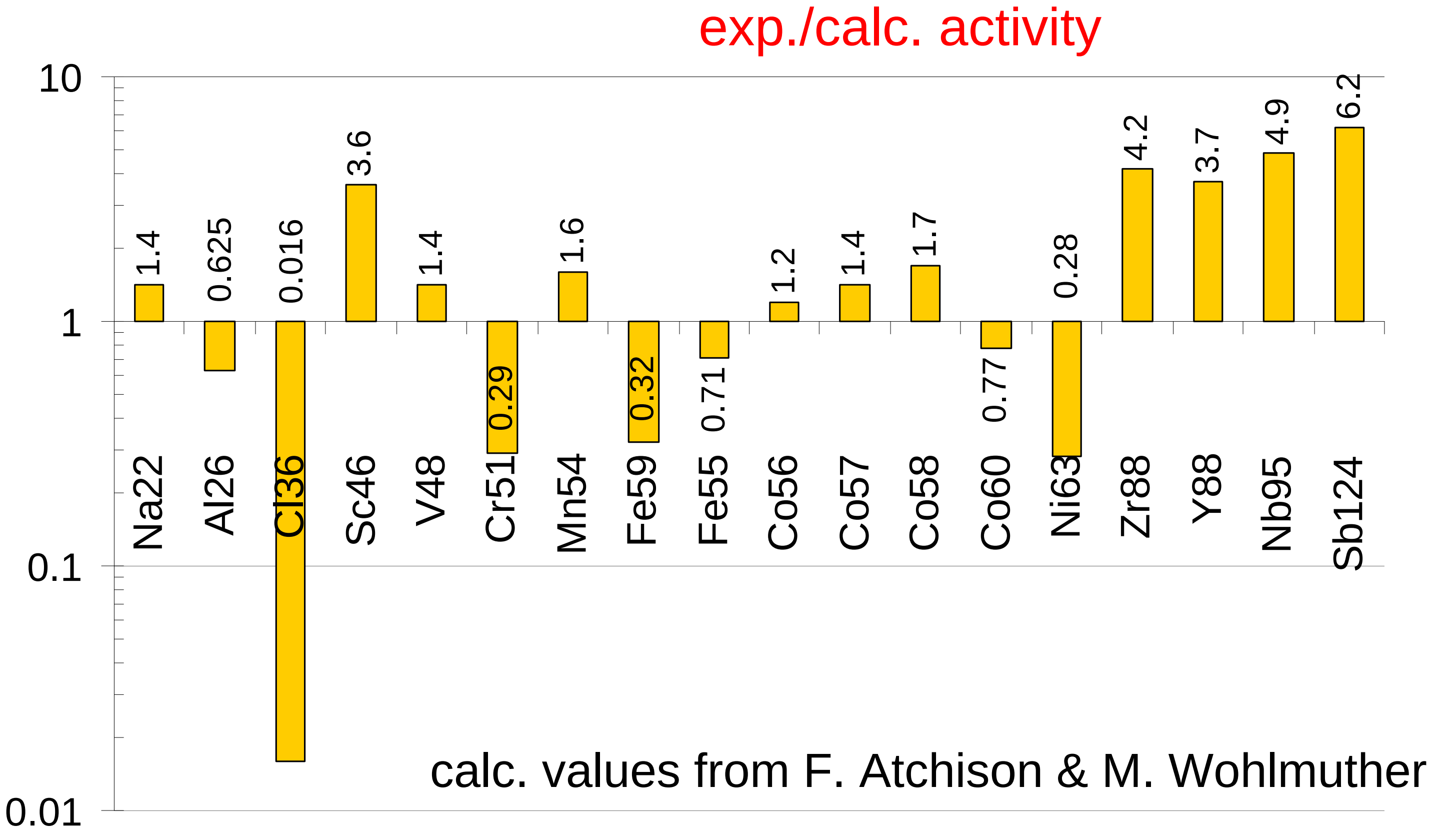


beam entry

irradiated for 13 years

sample from beam tube

Results for beam entry at ASK61



Calculation with PWWMBS + typical stainless steel composition

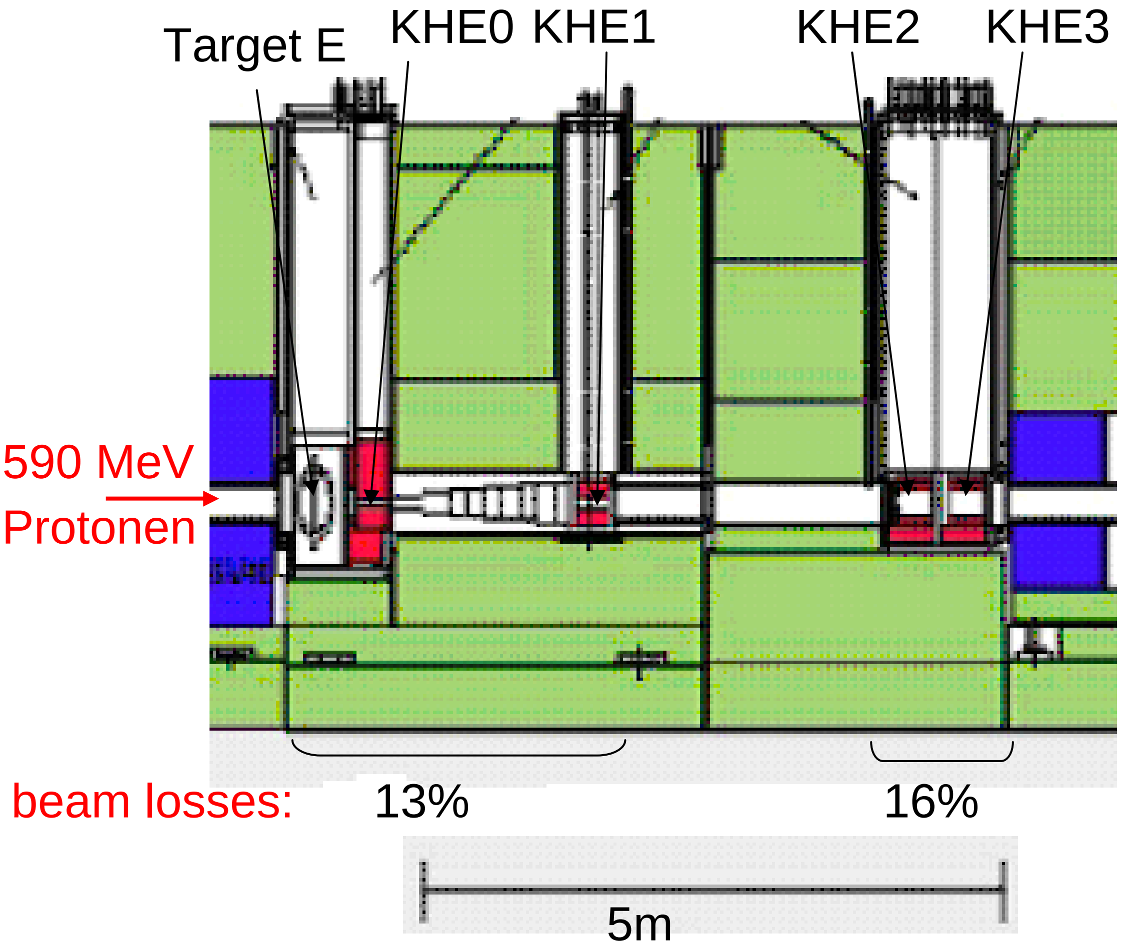
Satisfying agreement: < factor 10

exception: ^{36}Cl

reason: Cl content too large in material composition

→ large sensitivity on material definition

Proton beam line from Target E to KHE2&3



Target E:
purpose: meson production

- 4 cm graphite wheel
- rotating with 1 Hz
- additional beam spread:
  ~6 mrad

Collimator system: OFHC-Cu

- protection of the beam line
- reduction of beam losses
  (activation)

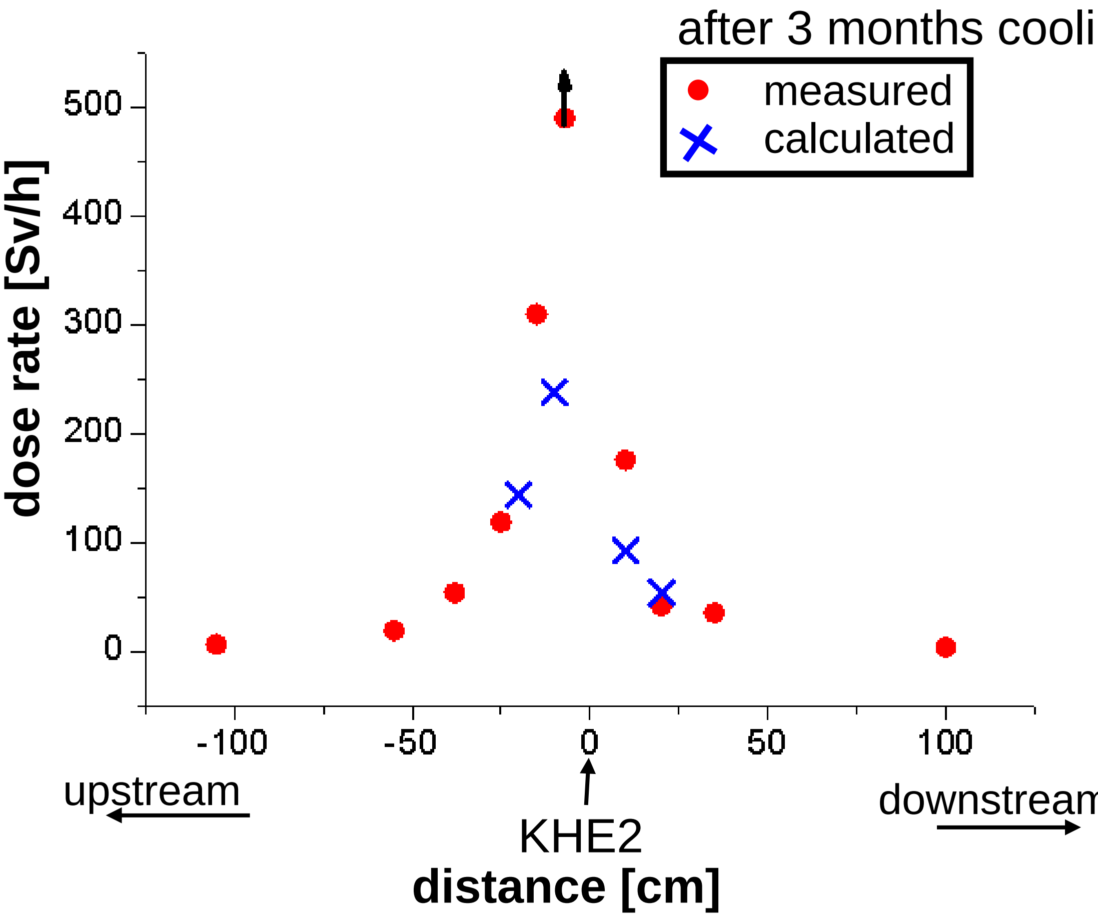
→large power deposition:
150 kW in KHE2

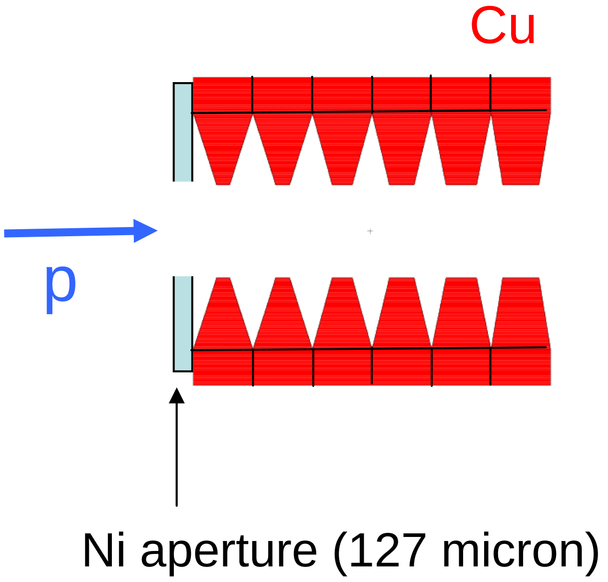
Dose rates at KHE2

- purpose of the calculation before inspection:

- planning of the shielding needed for camera in inspection tool
- shielding during transport sufficient

- calculated with MCNPX + Cinder'90, later measured in the hotcell (ATEC)





highest dose ever measured at PSI,
limit of the dose meter: 500 Sv/h

Radioactive waste

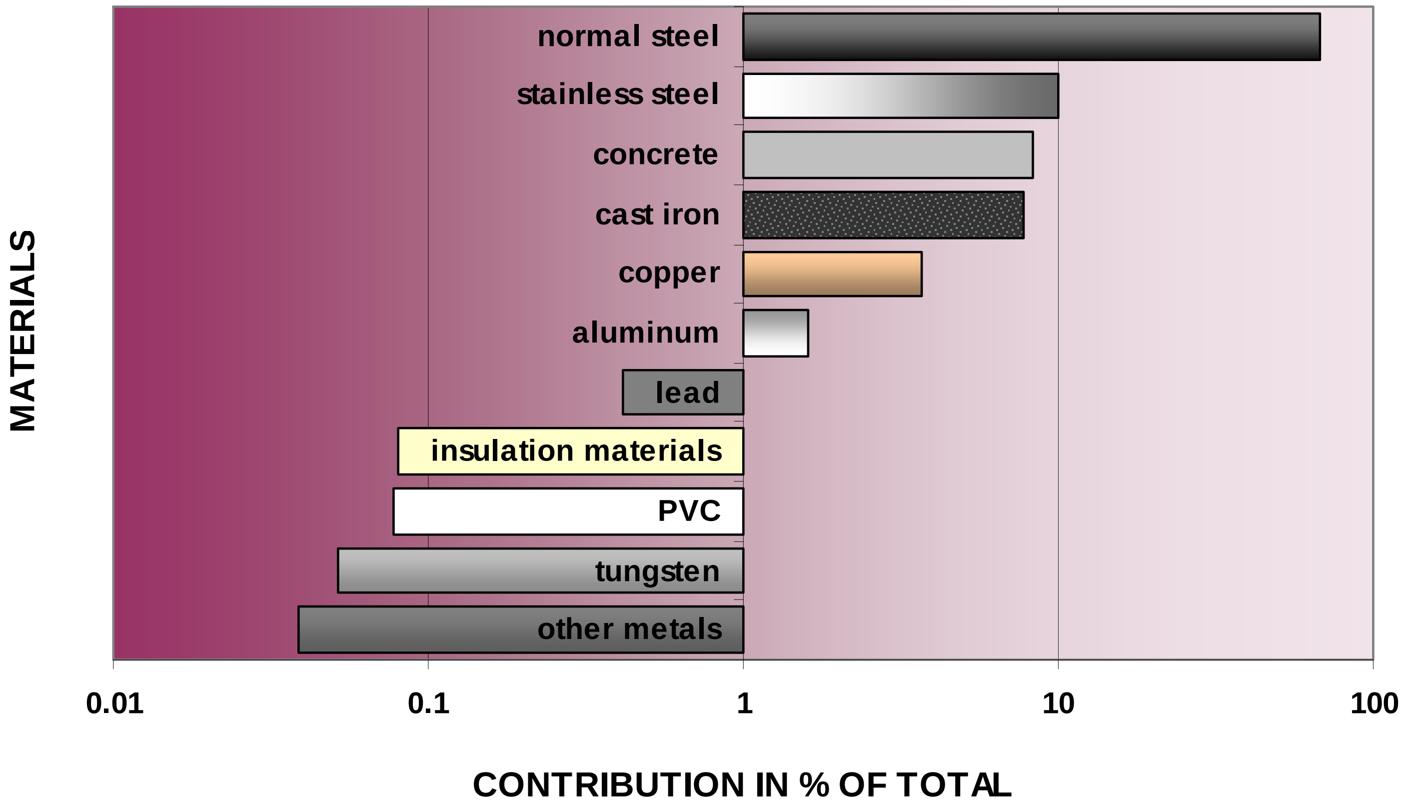
mainly low level waste: $1 < \sum_i \frac{A_i}{R_i} < 1000$

For final disposal:

- filled into concrete containers or steel drums
- components fixed with concrete (conditioning)

Accelerator waste at PSI:

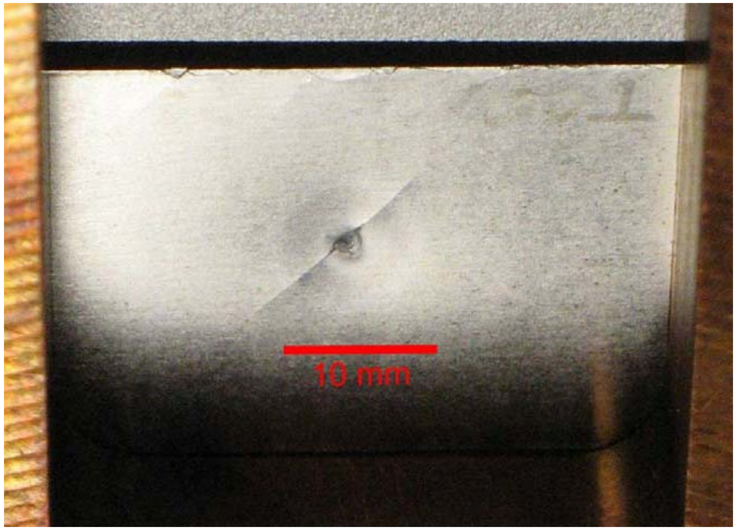
activity per container: 10^{10} – 10^{12} Bq (4.5 t of waste)



Radiation damage on materials

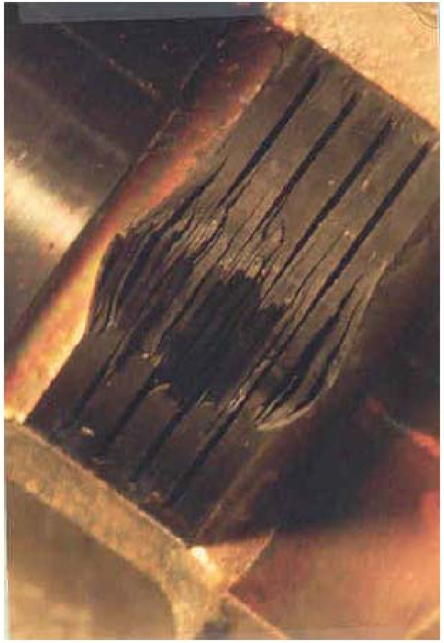
NSCL,
MSU

crack



Tungsten target 580 mg/cm2 (0.03 cm)
^{76}Ge^{30+} at 130 MeV/nucleon (5.77 x 10^{16})

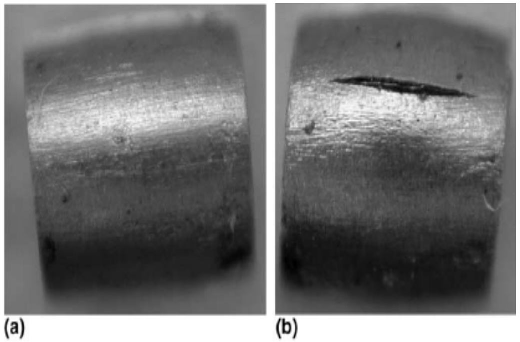
swelling + deformation



500 MeV
protons

Water-cooled/Edge-cooled graphite target at TRIUMF

before after irradiation



800 MeV
protons, LANL

Change of mechanical properties

Tungsten after compression test

important for high-power beams
on targets, collimators, beam dumps

Macroscopic Effects on Structural Materials

Degradation of mechanical properties:

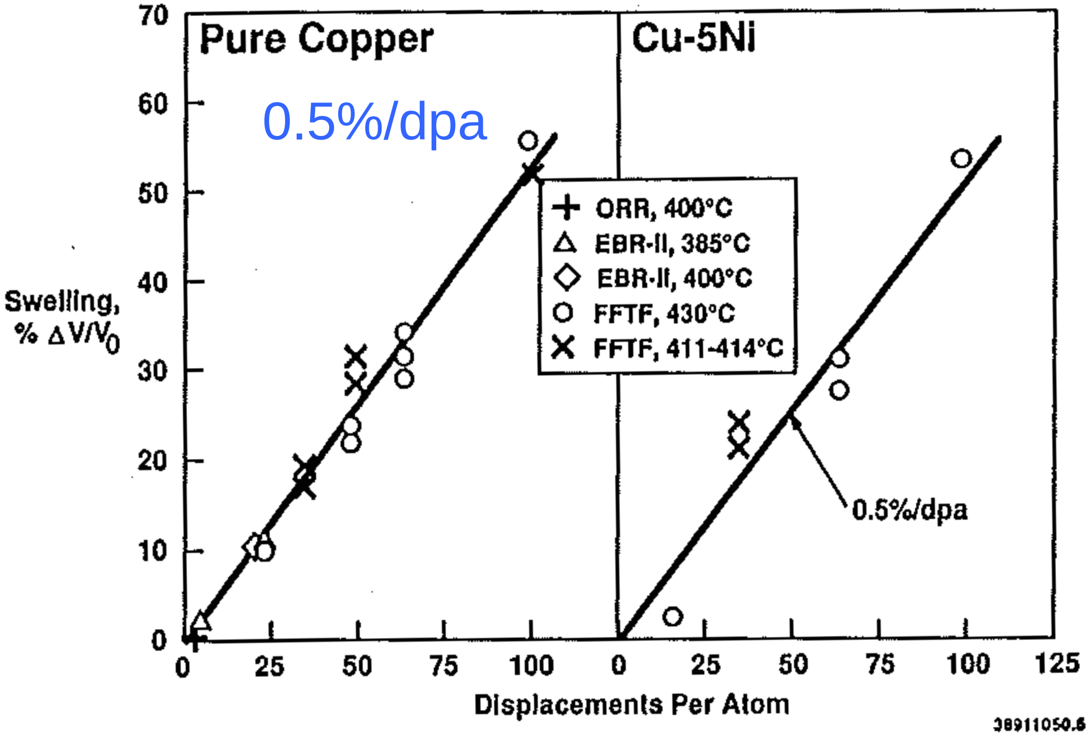
- Hardening (ductility, tensile strength)
- Irradiation embrittlement → cracks
- Radiation induced growth and swelling
- Irradiation creep
- Phase transformation
- Segregation of alloying elements

Affects life time of the component

Change of physical properties:

- Thermal conductivity ←→ electrical resistivity
- Thermal expansion
- Thermoelectric voltage

Swelling after irradiation with neutrons



DPA:
Displacement Per Atom,
a measure for radiation damage

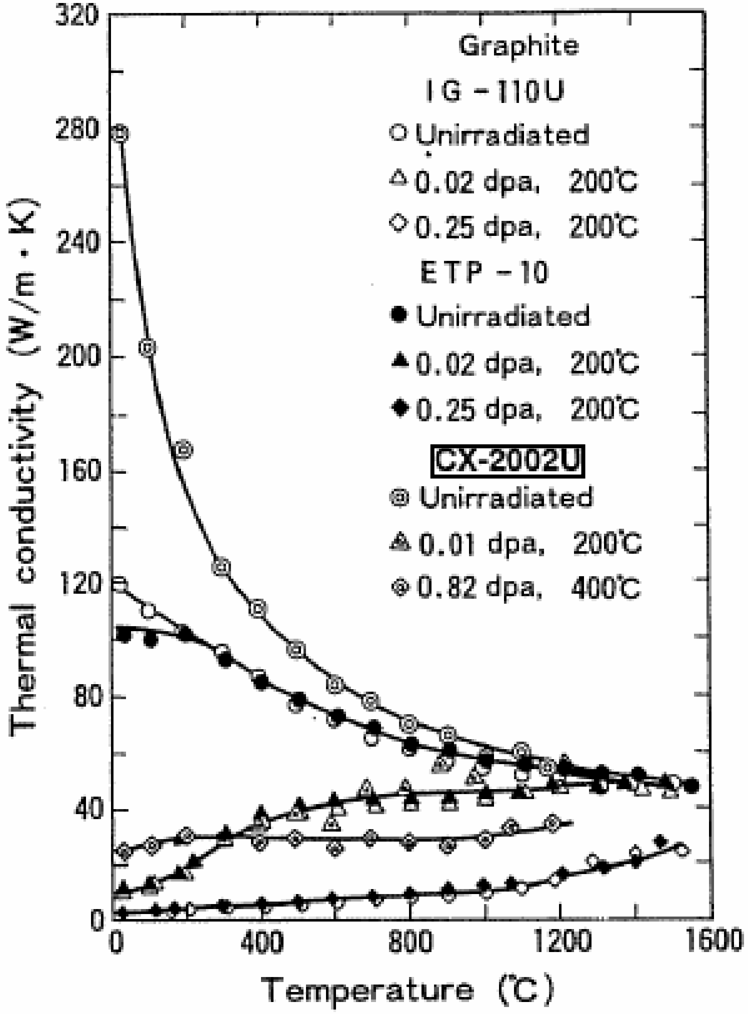
S. Zinkle, Effects of Radiation
on Material, 15th Symposium 1992,
p. 813-834

- no saturation up to 100 dpa
- data are for neutrons only (thermal and fast reactors)

→ Not much is known for high energetic protons

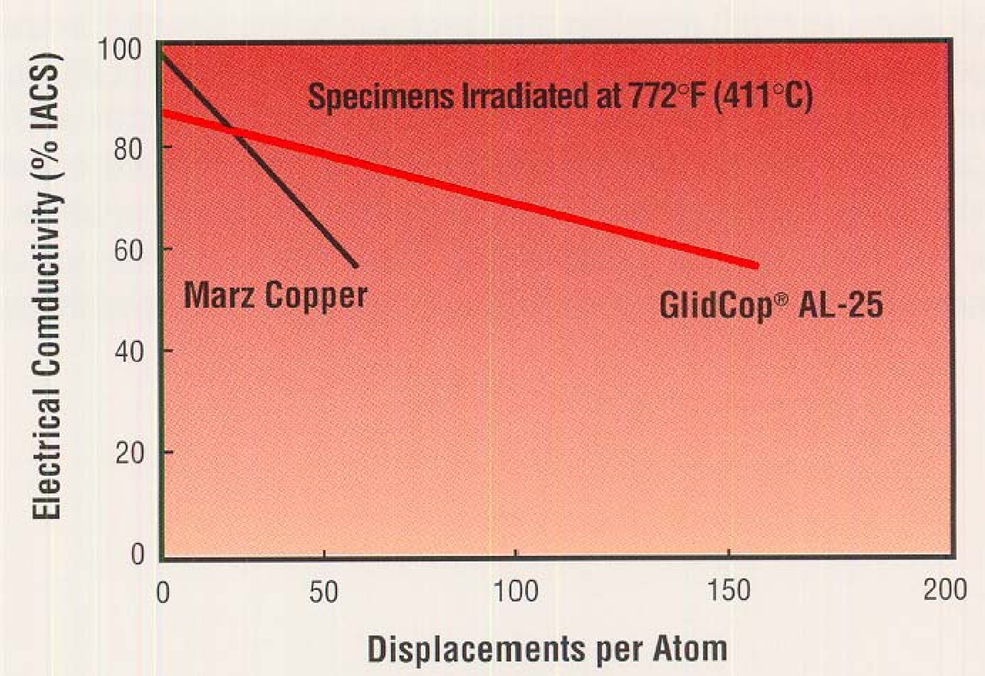
Thermal and electrical conductivity

Graphite



Thermal conductivity of neutron-irradiated graphites.

Copper + Glidcop



SCM Metal Products

Search for new materials,
which are especially radiation hard

T. Maruyama, M. Harayama
J. Nucl. Mat.195 (1992), 44-50

Energy loss of particles in material

- Electronic excitations (ionization): for charged particles

dissipated as heat,no damage (except temperature gets too high)

- Elastic interaction → transferring of recoil energy to a lattice atom

recoil nucleus loses energy due to

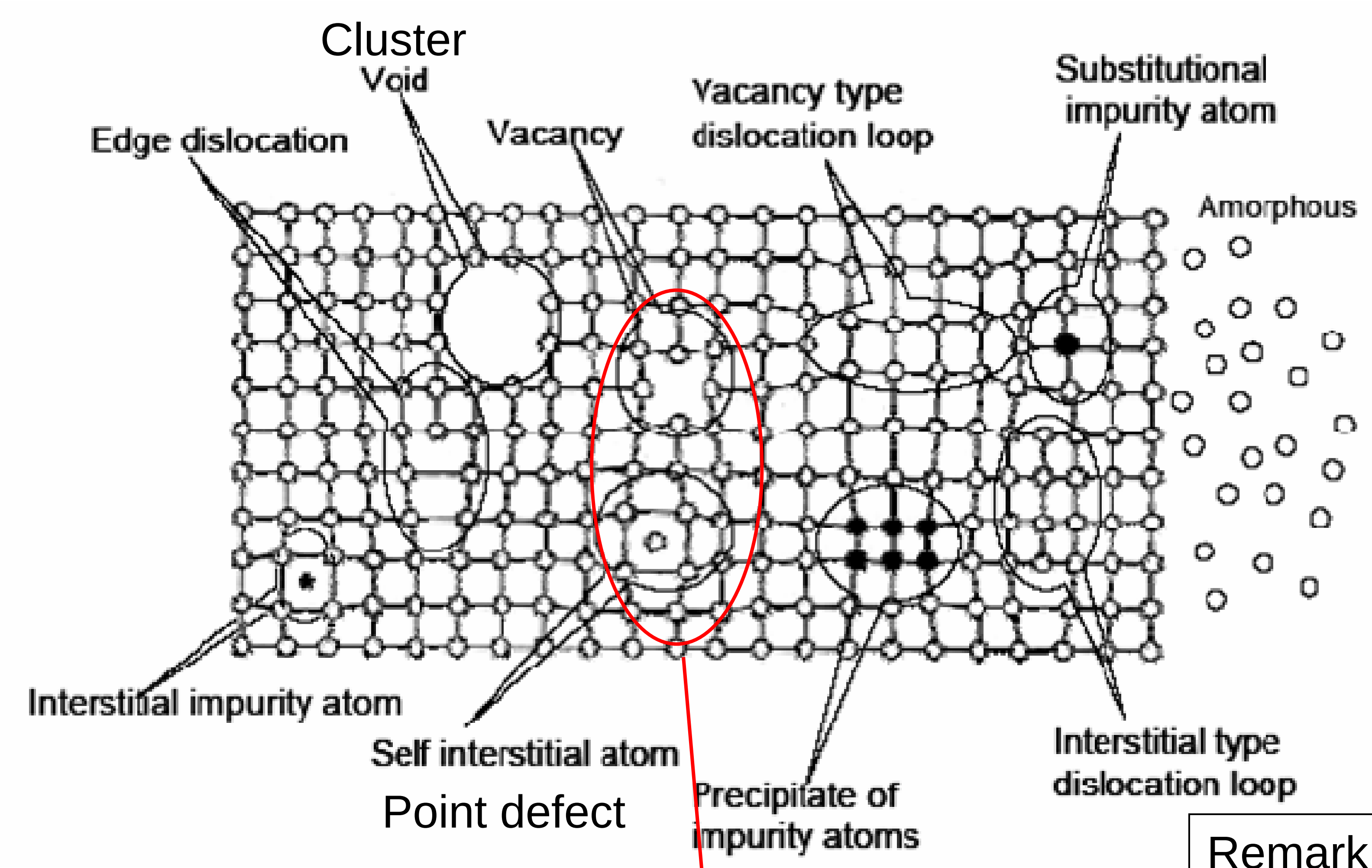
- ionization/excitation → dissipated as heat
- nuclear reactions → cascade of collisions (displacement cascade)
  → production of vacancies, interstitials

→ damage of material structure

→ change of mechanical and physical properties

- Inelastic interaction → transmutation of nuclei
  → activation
  → dose rate
  → impurities → effect on thermal conductivity
  → change of material properties (H, He production)
  → energy to recoil nucleus (s. above)

Microscopic and sub-microscopic effect on structural materials



vacancy + self-interstitial atom = Frenkel pair.

Remark:
Liquids do not suffer radiation damage

Displacements of atoms (DPA) in cascades

Mechanism:

1) p/n interacts with nucleus ➔recoil energy E_R to nucleus

2) recoil nucleus looses energy due to

- ionization/excitation → E_e dissipated as heat

→ energy left for nuclear reactions: $T_{dam} = E_R - E_e = \xi(E_R)\,E_R$

damage energy

partition function, damage efficiency

To displace an atom: bonds need to be broken

→Threshold energy called displacement energy E_D

range: 10 - 60 eV (~ twice the sublimation energy)

Cu: 30 eV, Fe, Ni,Co: 40 eV

a) $E_D < E_R < 2\,E_D$

1 atom is displaced to an interstitial site

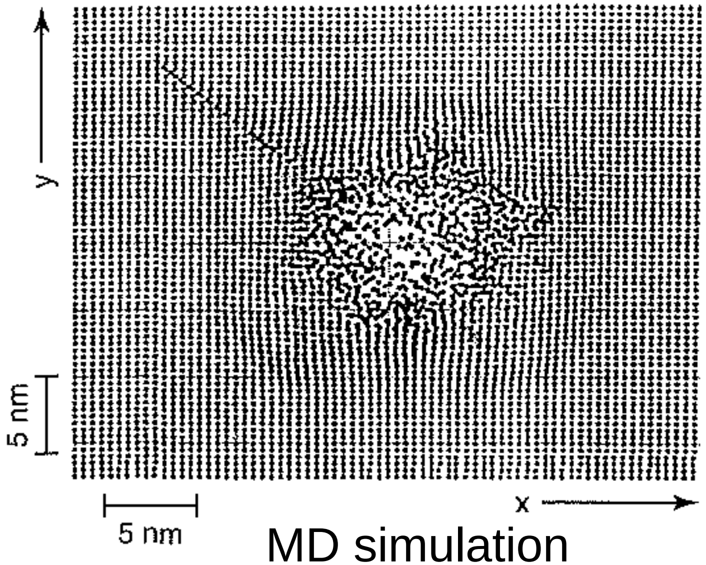
→ a vacant lattice site is created

b) $E_R > 2E_D$

cascade of collisions within a small range

→ displacement spike

(∅ ~ 10 nm after 1ps for E_R = 10 keV (PKA)



MD simulation

Calculation methods

Atomic nature of radiation damage (e.g. distribution, no. of vacancies):

- Molecular dynamics (MD): needs a lot of computer power
  - recombination of defects are taken into account
  - solves the equation of motion for all atoms at the same time
  - computing time ~ E_R^2

- Binary collision approximation (BCA): faster
  - discrete collisions in a lattice, other atoms are treated as spectators
  - particles are followed via trajectories
  - works well at higher energy

Estimation of DPA:

- Phenomenological approach (NRT = Norgett, Robinson, Torrens: 1975)
  - use of particle transport codes (e.g. MARS, FLUKA, MCNPX),
  - particles are followed via trajectories
  - no properties of the solid (e.g. lattice structure, atomic bound energy)
  - codes have models for all nuclear reaction cross sections
  - folding neutron/proton fluxes with displacement cross sections (dcs)

Displacement cross section (dcs)

$$\sigma_{dis}(E) = \int_{E_D}^{E\max} \frac{d\sigma_{dam}(E, E_R)}{dE_R}\, \nu(E_R)\ \mathrm{d}E_R$$

particle energy

damage cross section:

$$= \frac{w(E_R)}{xN_V}$$

damage function (no. of displaced atoms):

$$= \frac{\kappa T_{dam}}{2E_D}$$

modified Kinchin-Pease m.
= NRT model:
Nucl.Eng.Des. 33 (1975) 50

$w(E_R)$: recoil spectrum
needs nuclear reaction models

x: thickness of the sample (thin)

N_V: atomic density (atoms/cm^3)

$T_{dam}(E_R)$: damage energy

displacement efficiency $\kappa = 0.8$

some remarks on uncertainties:

- E_D e.g. in Cu set to 30 eV but varies 18 – 43 eV
- $\kappa = 0.8$: compensates for forward scattering,
  derived from BCA simulation of Robinson, Torrens 1972

Displacements per atom

Displacements Per Atom (DPA):

- how often an atom is displaced during the irradiation period

$$DPA = \int \sigma_{disp}(E)\ \frac{d\phi(E)}{dE}\ \mathrm{dE}$$

$\phi(E)$: fluence (particles/cm^2)

σ_{disp}: displacement cross section

Related to the number of Frenkel pairs N_F:

$$DPA = \sum_i N_i N_F^i$$

i: number of reaction channels

N_i = number of particles

DPA is used to quantify the radiation damage

Problem:

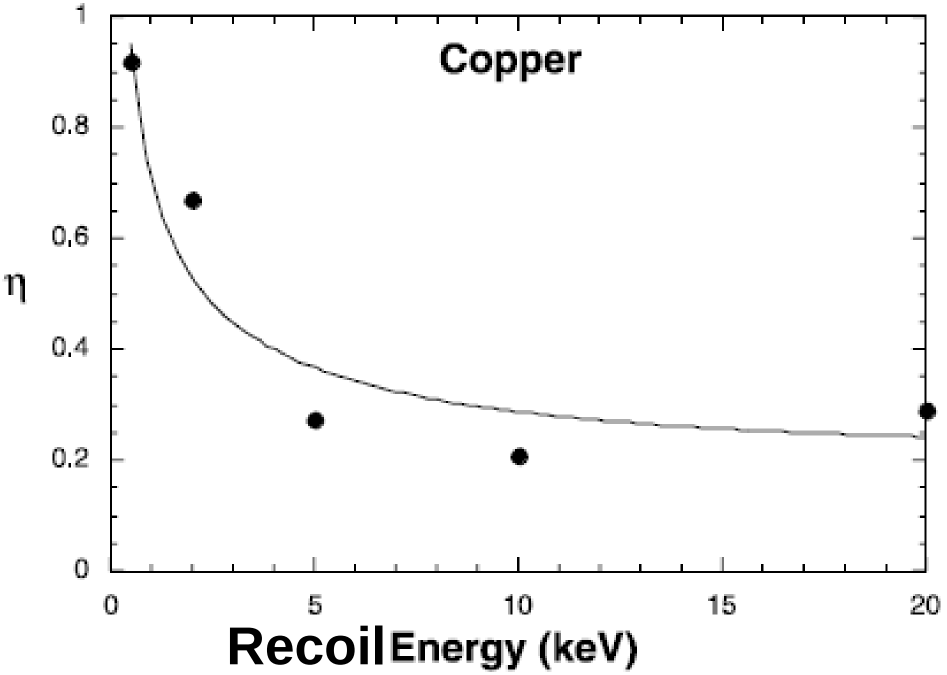
It cannot be measured!

Reason:

only a small fraction of the displaced atoms

leads to permanent lattice defects

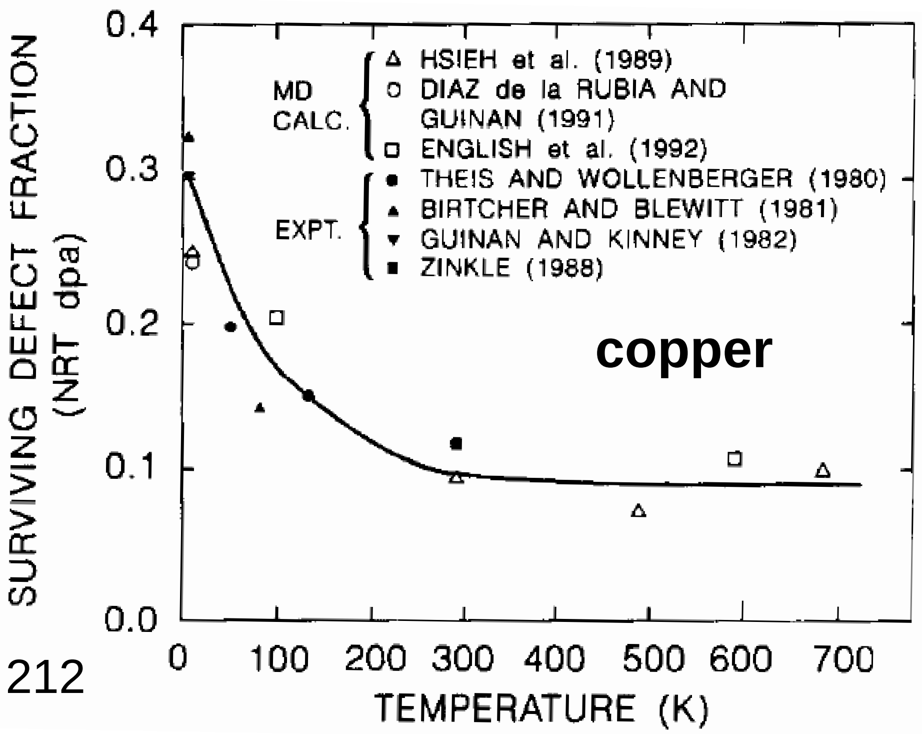
Defect production efficiency η



Cartula et al.,
J. Nucl. Mat. 296 (2001) 90

$$\eta = \frac{\text{stable defects}}{\text{displacements}}$$

$$\nu(E_R) = \eta \frac{\kappa T_{dam}}{2E_D}$$

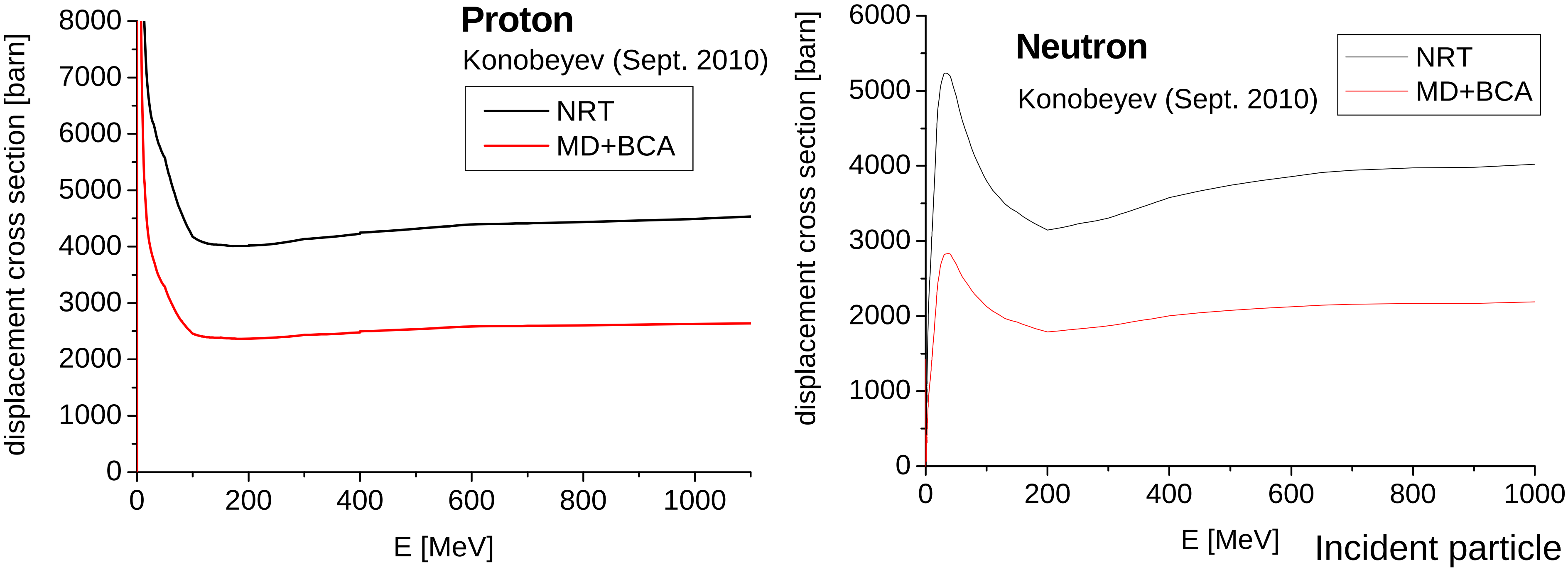


Singh, Zinkle et al.,
J. Nucl. Mat. 206 (1993) 212

Displacement cross sections (A. Konobeyev, KIT)

on copper

E < 28 keV: MD, E > 28 keV: BCA



Incident particle energy

$$DPA = \int \sigma_{disp}(E)\ \frac{d\phi(E)}{dE}\ \mathrm{dE}$$

$\phi(E)$: fluence (particles/cm^2)

σ_{disp}: displacement cross section

Main difference between proton and neutron dcs:

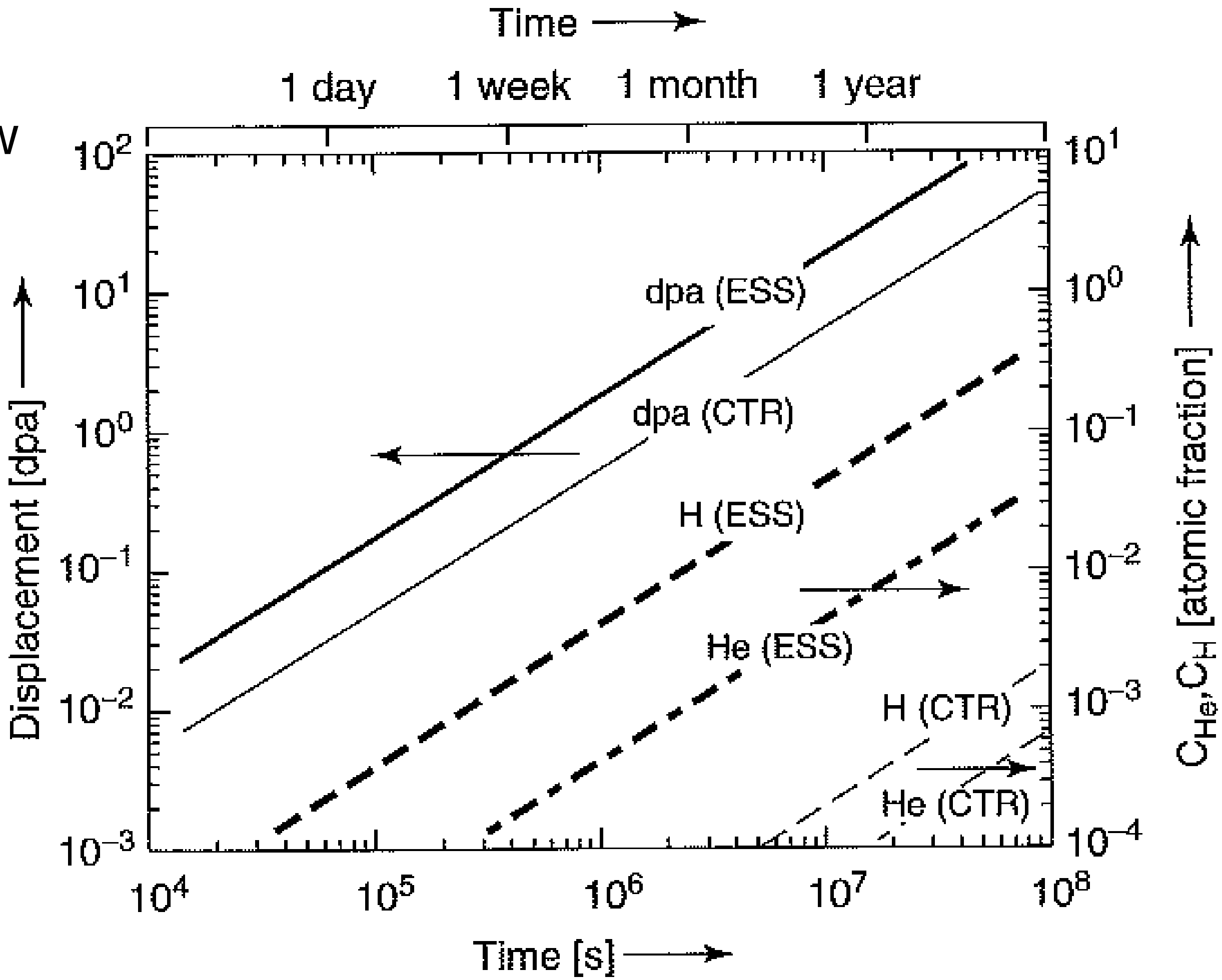
Coulomb interaction, important at low energy

Practical example: target window of ESS

5 MW proton beam, 1.3 GeV → $\phi = 2\ 10^{22}\ /\mathrm{cm}^2$ protons in 1 year
dcs for iron: 3000 barn at 1 GeV p
→ DPA = 3000 barn * $2\ 10^{22}\ /\mathrm{cm}^2 = 60$

Life time for T91 window for MEGAPIE at PSI: ~ 10 DPA
(Y. Dai et al., J. Nucl. Mat. 356 (2006) 308)

Caution: life time predictions depend on many parameters!

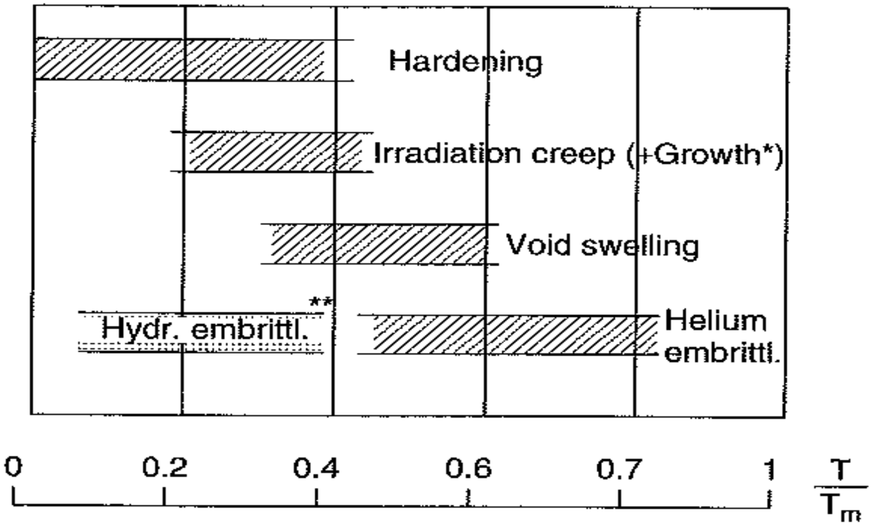


Filges, Goldenbaum, Handbook of Spallation Sources

Effect on material properties

Very difficult to predict,
dependent on

- temperature
  (healing!, defects get mobile)
- impurities (partly produced)
- grain size
- rate of irradiation: [dpa/s]
- kind of particle irradiation, its energy



T_m: melting temperature

+ lots of data for thermal neutrons

- not many data available for high-energy particles

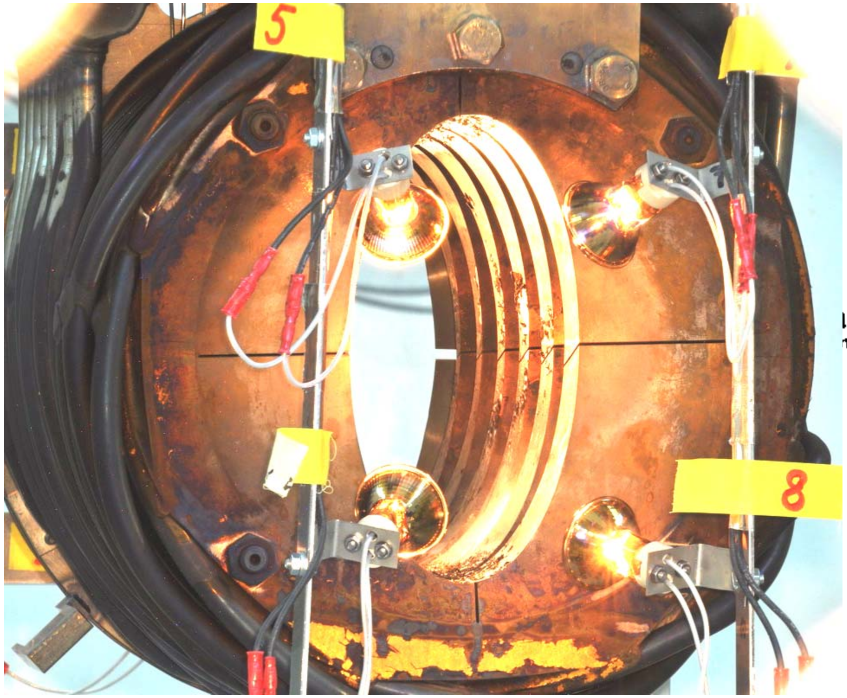
How to transfer mechanical/physical property changes measured on thermal/fission reactor neutrons (a lot!) to high-energy particle beams?

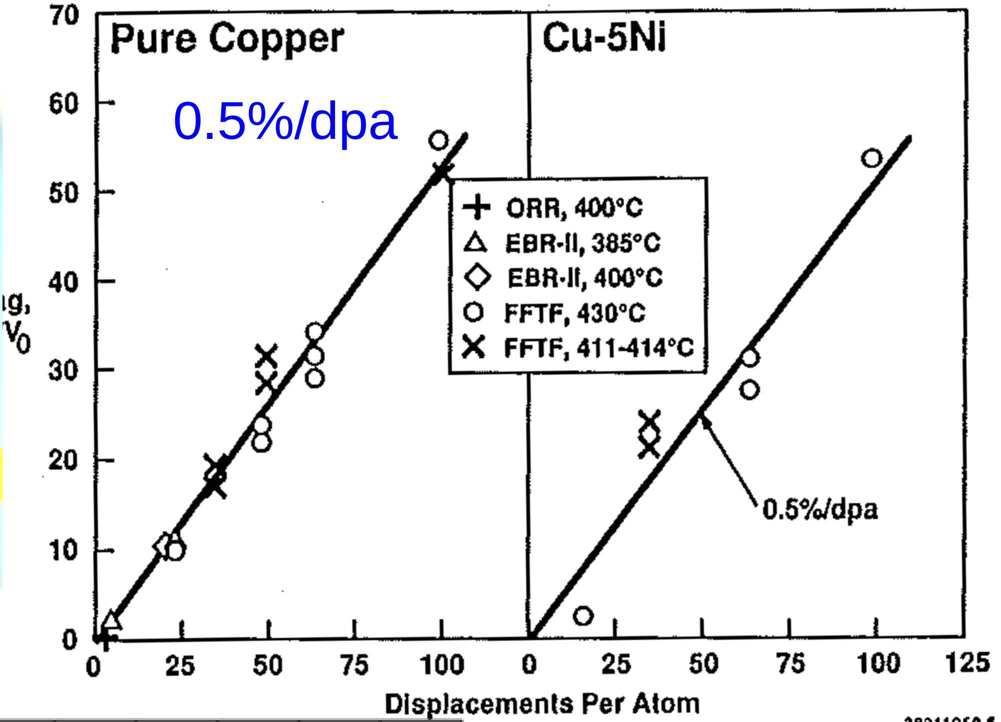
→damage correlation

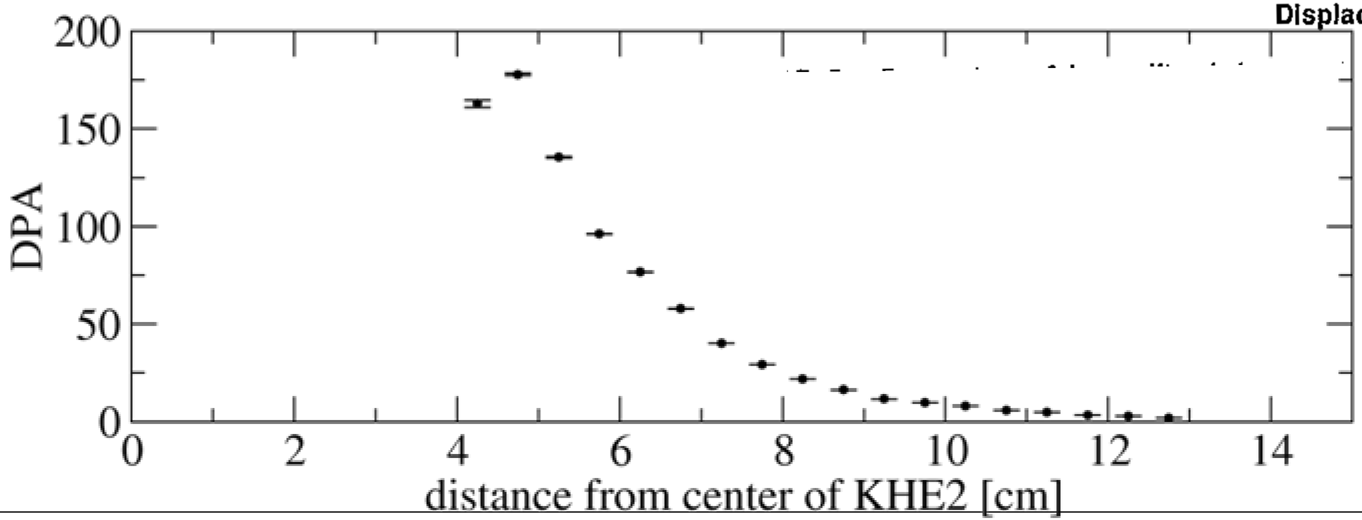
→very complex problem

irradiation test experiments are needed!

Cu-collimator at PSI: 20 years in use, ~ 15 Ah







Radial dependence of DPA on 1. tooth

very high DPA at the inner side

Summary

Activation:

- important reaction mechanisms:
  - spallation → excitation → emission of p,π, evaporation of n, light ions
  - neutron capture

Calculation:

1) direct irradiation:

- particle transport codes: MCNPX, FLUKA, MARS, PHITS ….
  - physical reactions models
  - evaluated cross section for n < 20 (150) MeV

2) in the environment of loss points:

- simplification due to almost constant neutron fields possible

Radiation damage:

- defects in the lattice structure due to recoil nuclei
- change of material properties: mechanical, physical
- calculation: MC, BCA, displacement cross sections
- to quantify radiation damage: DPA
- due to recombination (defect efficiency) DPA cannot be measured
- radiation damage depends on many parameters
  → very difficult to predict