



Erosion Processes due to energetic particle- surface interaction

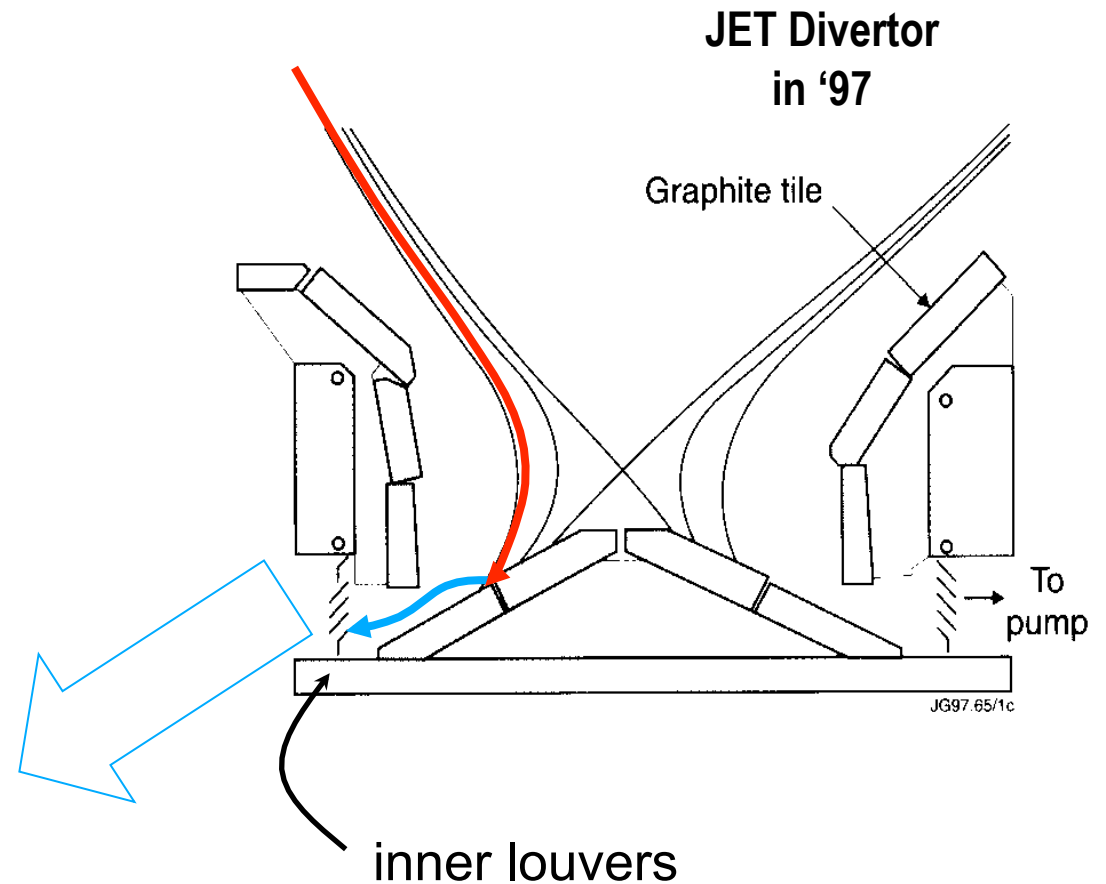
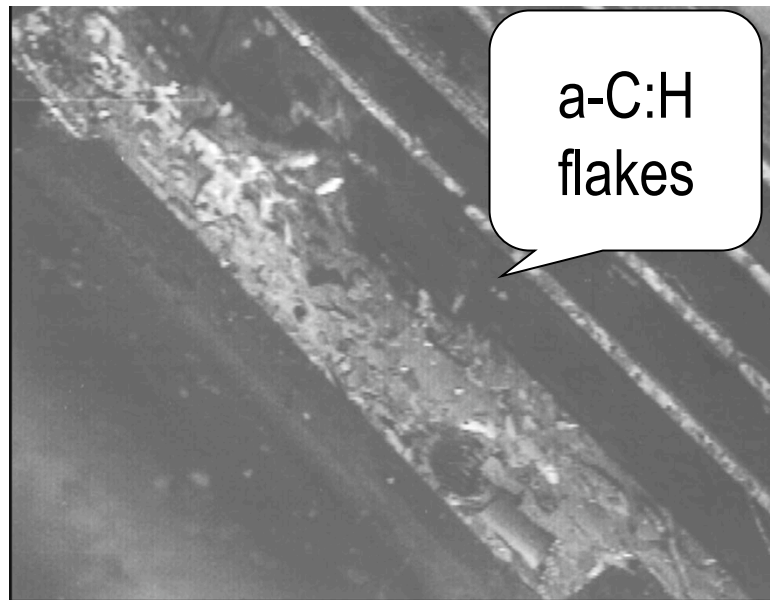
Klaus Schmid

- **Introduction**
- **Physical sputtering**
- **Chemical erosion**
- **Chemical sputtering**
- **Radiation enhanced sublimation**
- **Impact of impurities on fusion plasmas**

Introduction

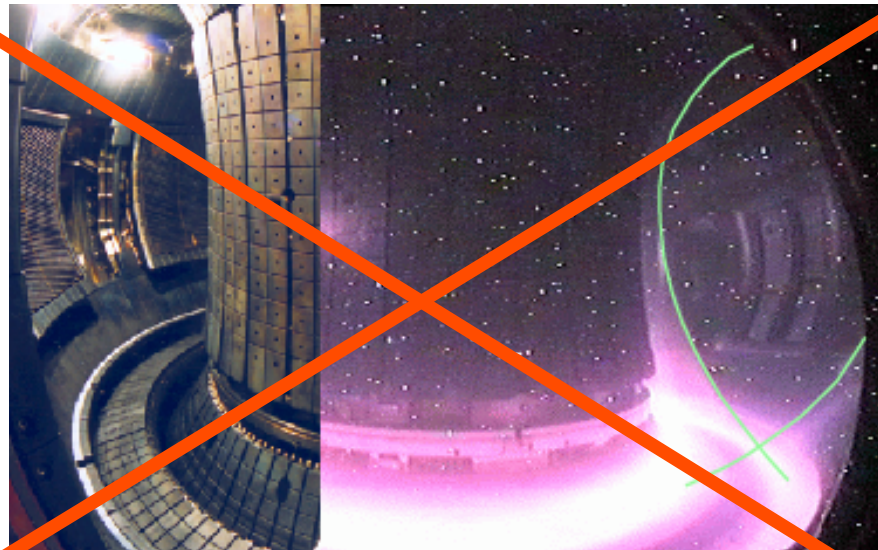
why a whole lesson on erosion?

- lifetime considerations
- source of codeposition
- Plasma cooling & dilution



Introduction

experimental approach

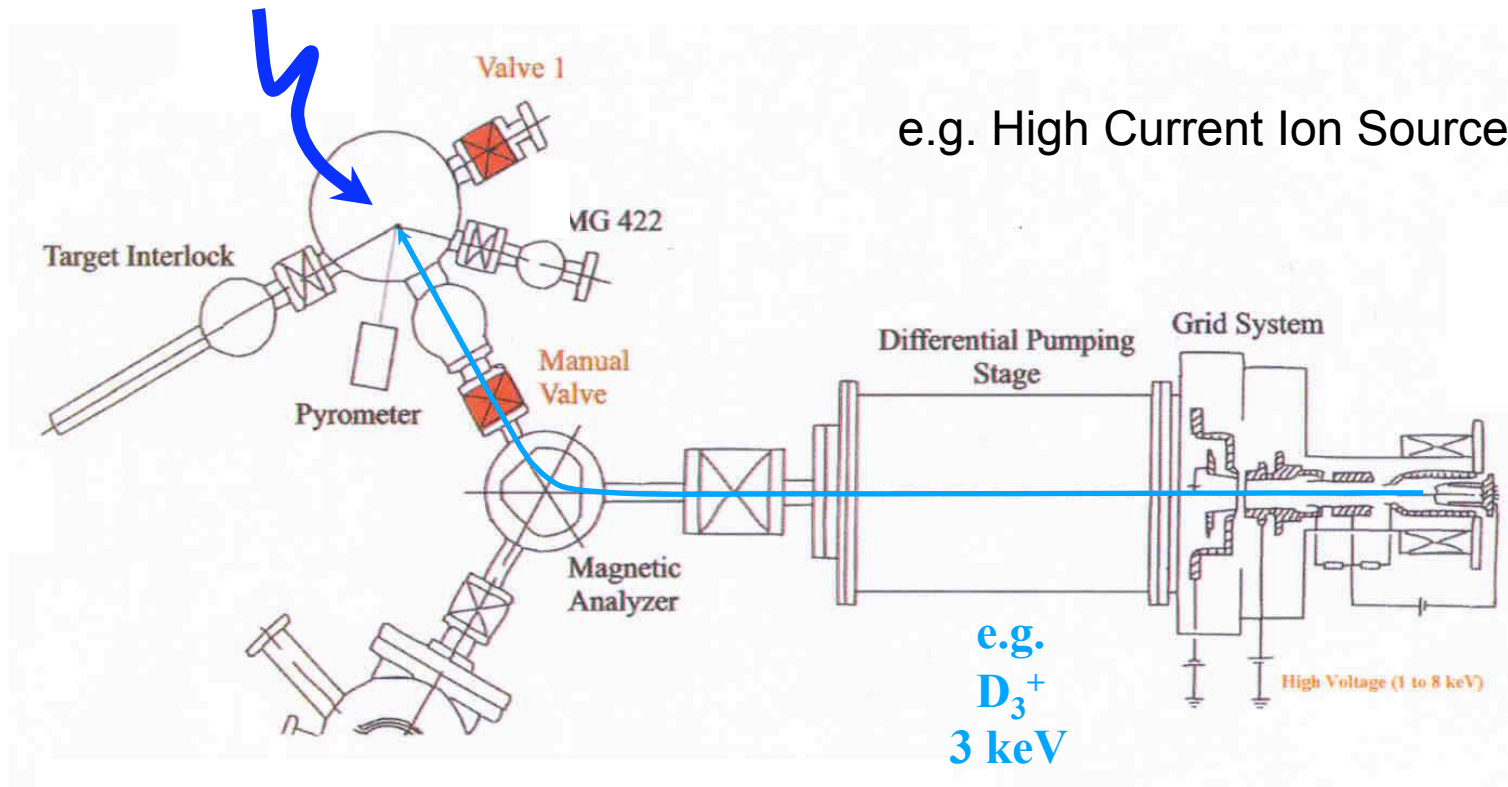


- ?Surface temperature
- ?Incident particle flux
- ?Incident particle energy
- ?Incident flux compositions

Need control over these parameters

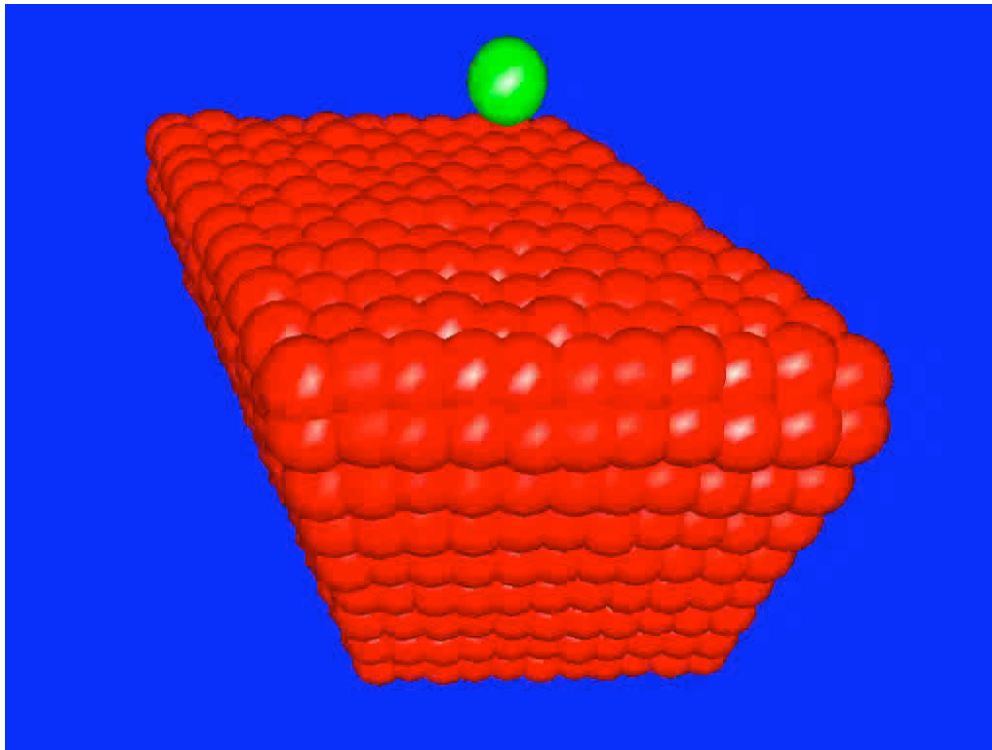
Introduction

erosion of target materials with $j < 10^{19} \text{ m}^{-2}\text{s}^{-1}$, $100 \text{ eV} < E < \text{keV}$



detection of erosion products by mass spectrometry or weight loss measurements

Molecular Dynamics simulation of 50eV He \rightarrow Be

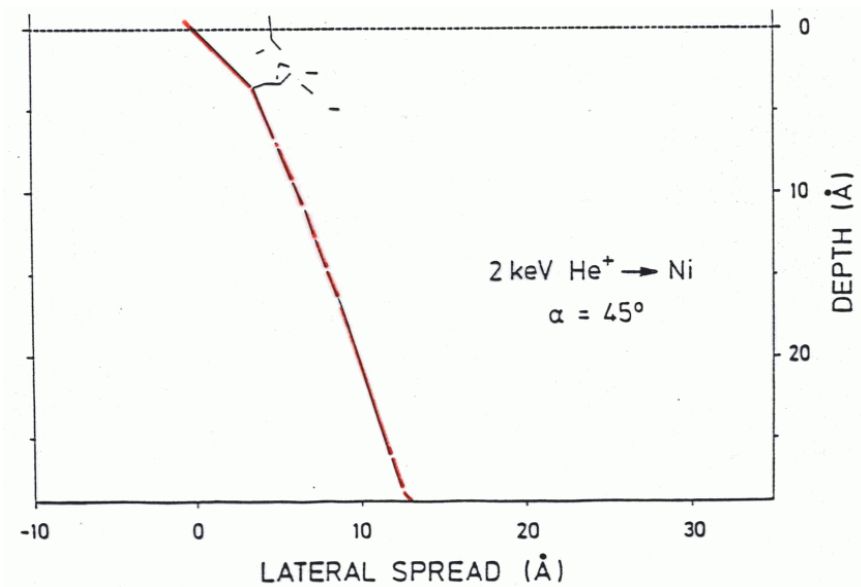
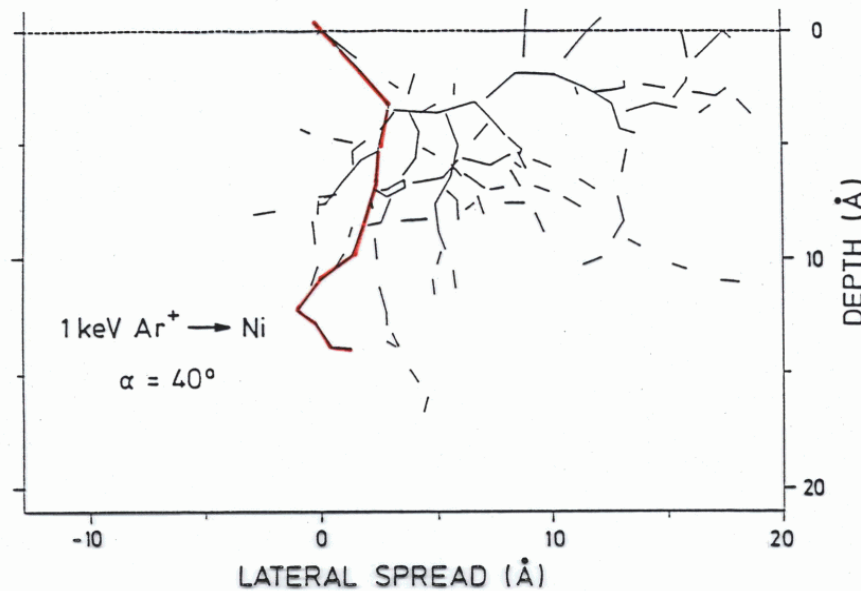


❖ Energetic particle impact involves a complex collision cascade during which:

- The projectile may be reflected back out of the surface
- The projectile may remain in the surface (=implantation)
- Surface atoms may be ejected out from the surface (= physical sputtering)
- The surface may be left with crystal damage.

Energetic particle impact is a stochastic process and is therefore described by giving average yields for the different processes

- Physical sputtering is the kinetic ejection of surface atoms by incident energetic ions or atoms due to collision processes.
- As surface atoms can escape only if it receives an energy larger than the surface binding energy, a threshold energy for the incident particles is required.
- In fusion application physical sputtering by hydrogen ions and atoms is important, but also the self-sputtering due to returning impurity atoms.



- TRIM Monte-Carlo Code simulation

Heavy ions:

- large collision cascade
- isotropic velocity distribution
- yield proportional to energy deposited in first two layers

Light ions:

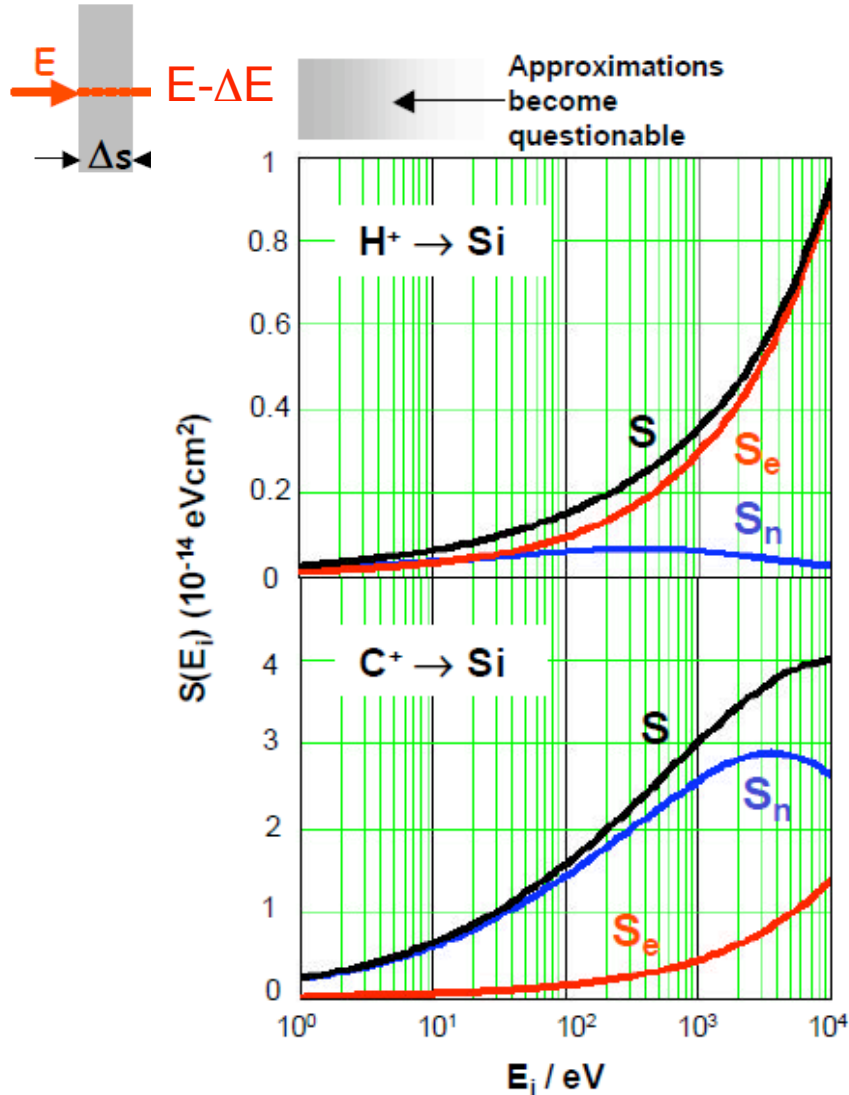
- few collisions
- energy transfer in single collision
- Momentum inversion in two or more collisions
- Sputtering occurs mostly by reflected particles

- $$T = E_0 \frac{M_1 M_2}{(M_1 + M_2)^2} \cos^2 \delta$$

Physical sputtering



stopping power



Stopping Power $B = - \frac{dE}{ds}$ E Energy
s Pathlength

Stopping Cross Section $S = - \frac{1}{n} \frac{dE}{ds}$ $n = \frac{\rho L}{m_2}$
Atomic Density

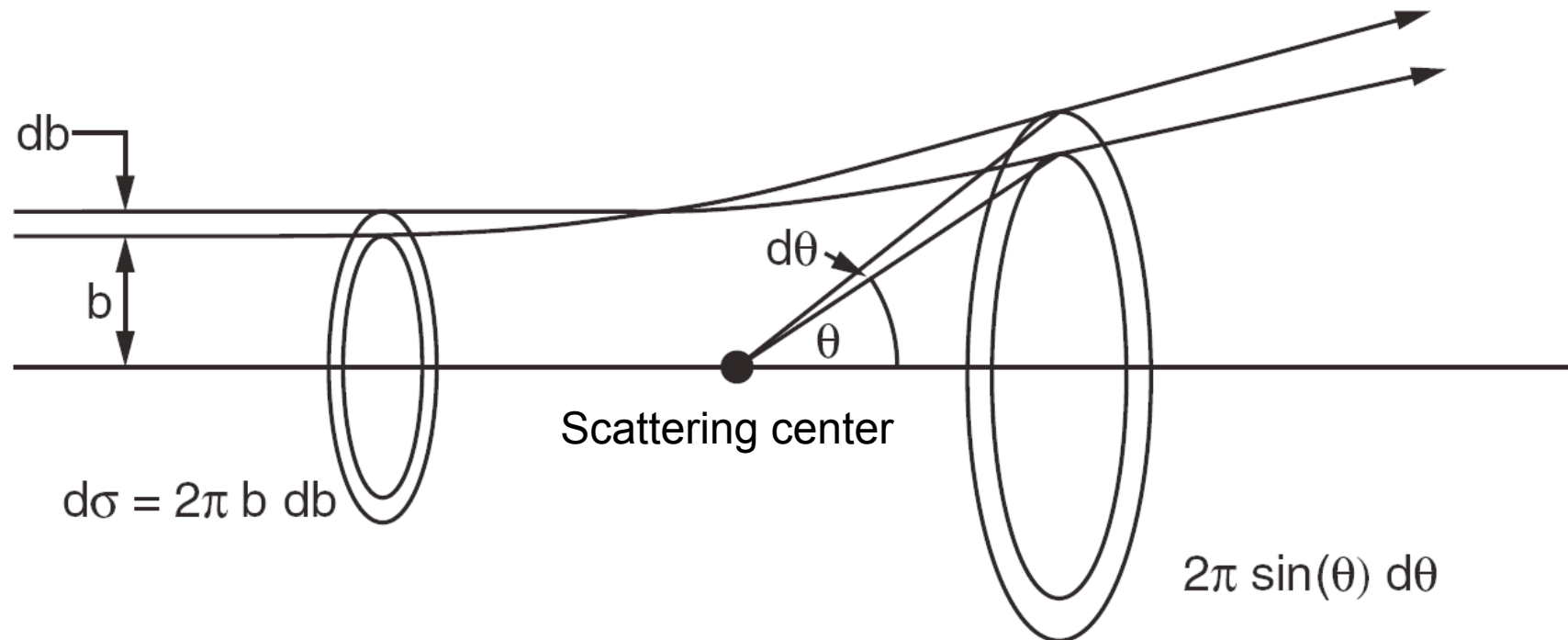
Interaction with Target Atoms ("Nuclei") and Electrons: $S = S_n + S_e$

❖ Stopping power is velocity dependent

➤ Fast particles stopping dominated by S_e

➤ Slow particles stopping dominated by S_n

Scattering cross section

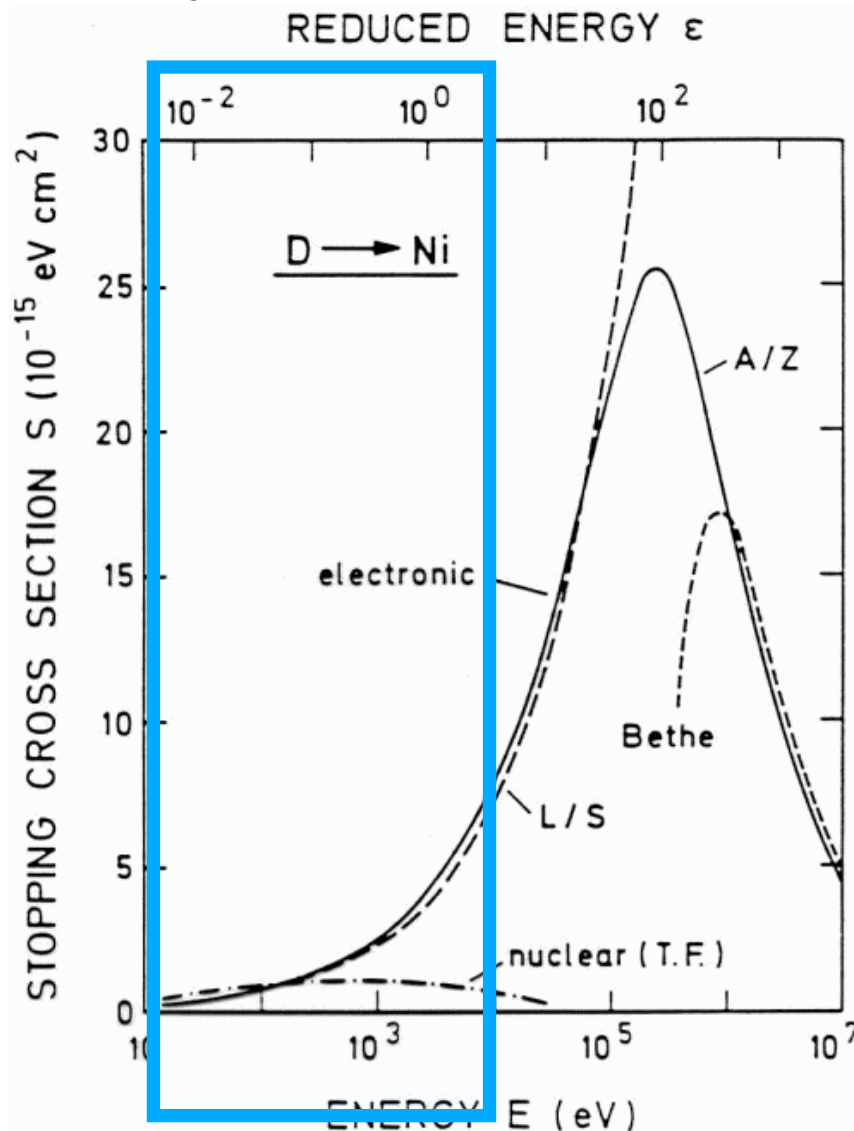


Probability dw for a scattering event with a scattering angle $\theta \pm d\theta$

$$dw = N_A d\sigma(\theta) = N_A 2\pi b db$$

N_A areal density of scattering centers (i.e. atoms) $d\sigma/d\Omega(\theta) \propto Z_1 Z_2 / E^2 \sin^4(\theta)$
 $d\sigma(\theta(b)) =$ Scattering cross section

Stopping power

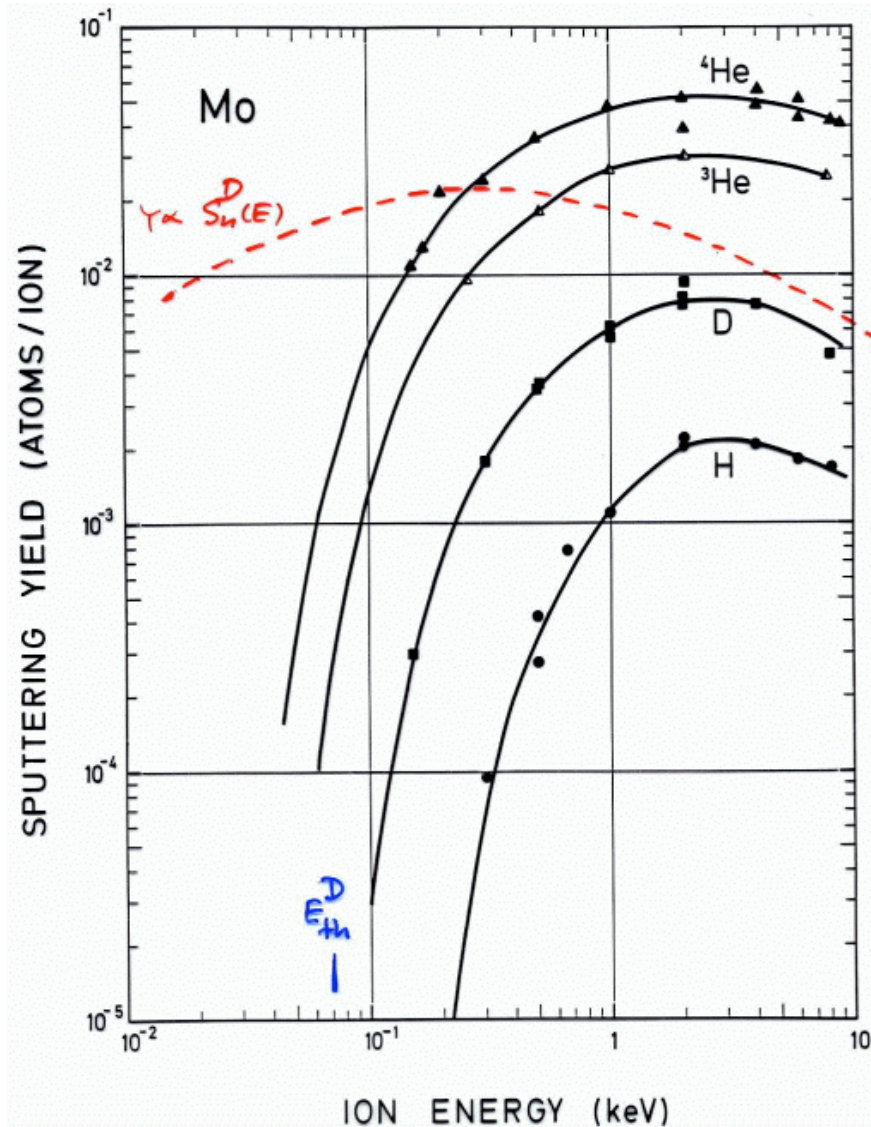


- Projectiles and recoil atoms lose energy in elastic collisions (nuclear stopping) and collisions with electrons (inelastic stopping)
- Nuclear stopping decreases for high energies. (E^{-2} dependence of scattering cross section)
- Nuclear stopping increases for low energies. (Overcomes screening of coulomb potential)
- Nuclear stopping can be calculated for a given interaction potential (screened coulomb):

$$S_n(\varepsilon) = \frac{0.5 \ln(1 + 1.2288\varepsilon)}{\varepsilon + 0.1728\sqrt{\varepsilon} + 0.008\varepsilon^{0.1504}}$$

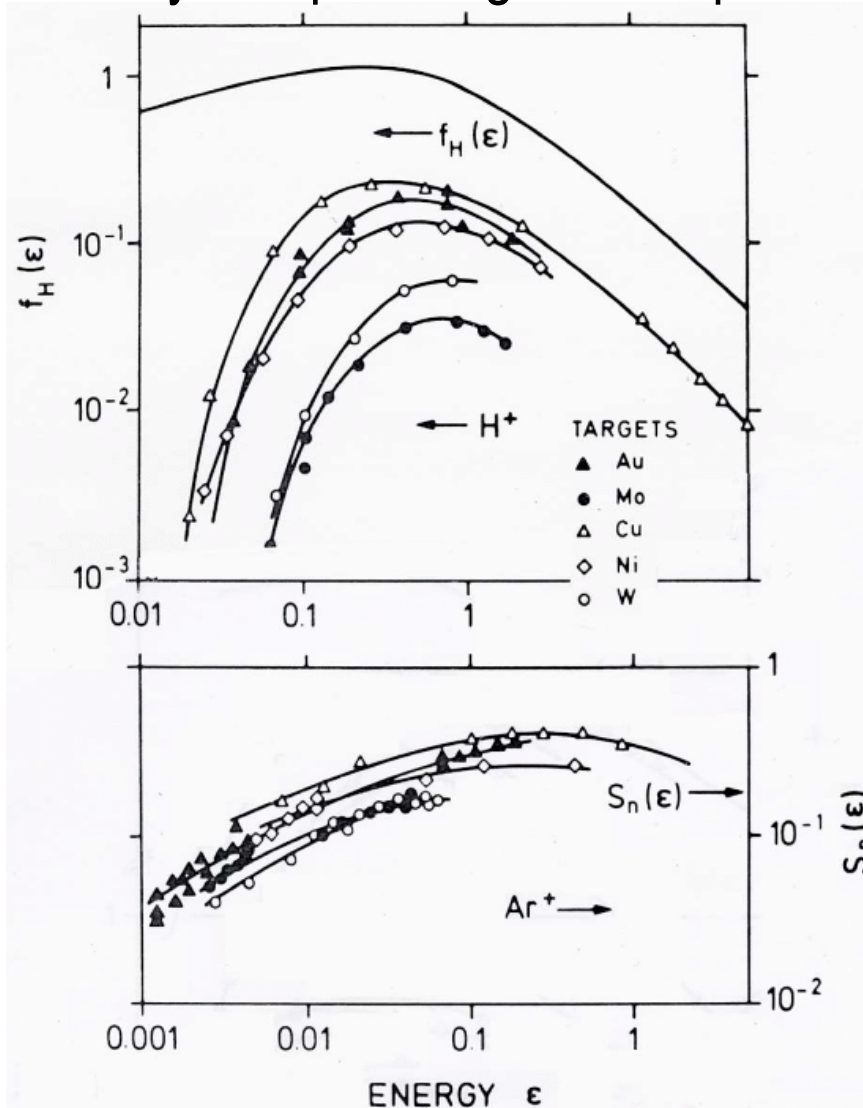
$$\varepsilon = E_0 \frac{M_2}{M_1 + M_2} \frac{a_{Bohr}}{Z_1 Z_2 e^2} = \frac{E_0}{E_{TF}}$$

Threshold energy



- Light ion sputtering in fusion application is dominated by threshold effects
- Energy loss by nuclear stopping alone does not explain physical sputtering.

Theory for sputtering in isotropic collision cascades



Ansatz:

(P. Sigmund (1969))

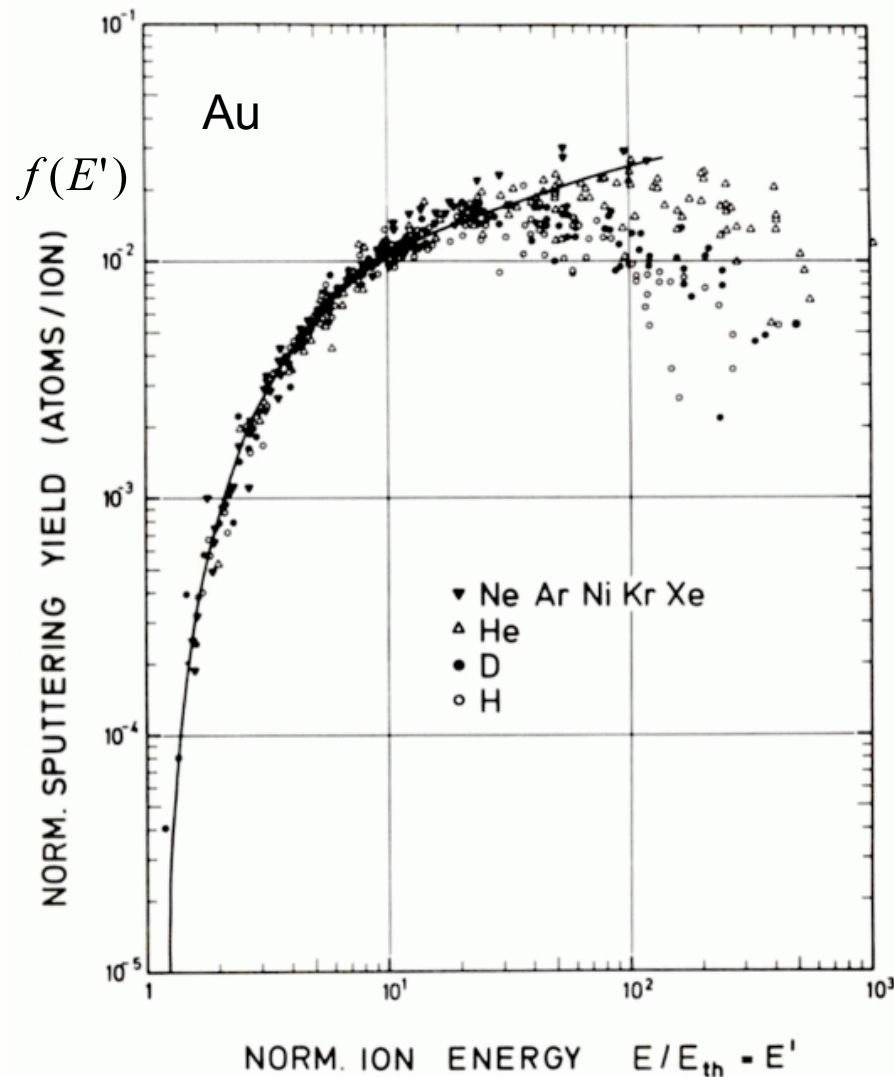
Sputtering yield proportional to the energy deposited into collisions near the surface

$$Y(\epsilon) \propto S_n(\epsilon)_{x=0}/E_s$$

$$Y(\epsilon) = Q(M_1, M_2, E_s) * f_H(\epsilon)$$

E_s = Surface binding energy $\cong \Delta H$
heat of sublimation

Threshold function

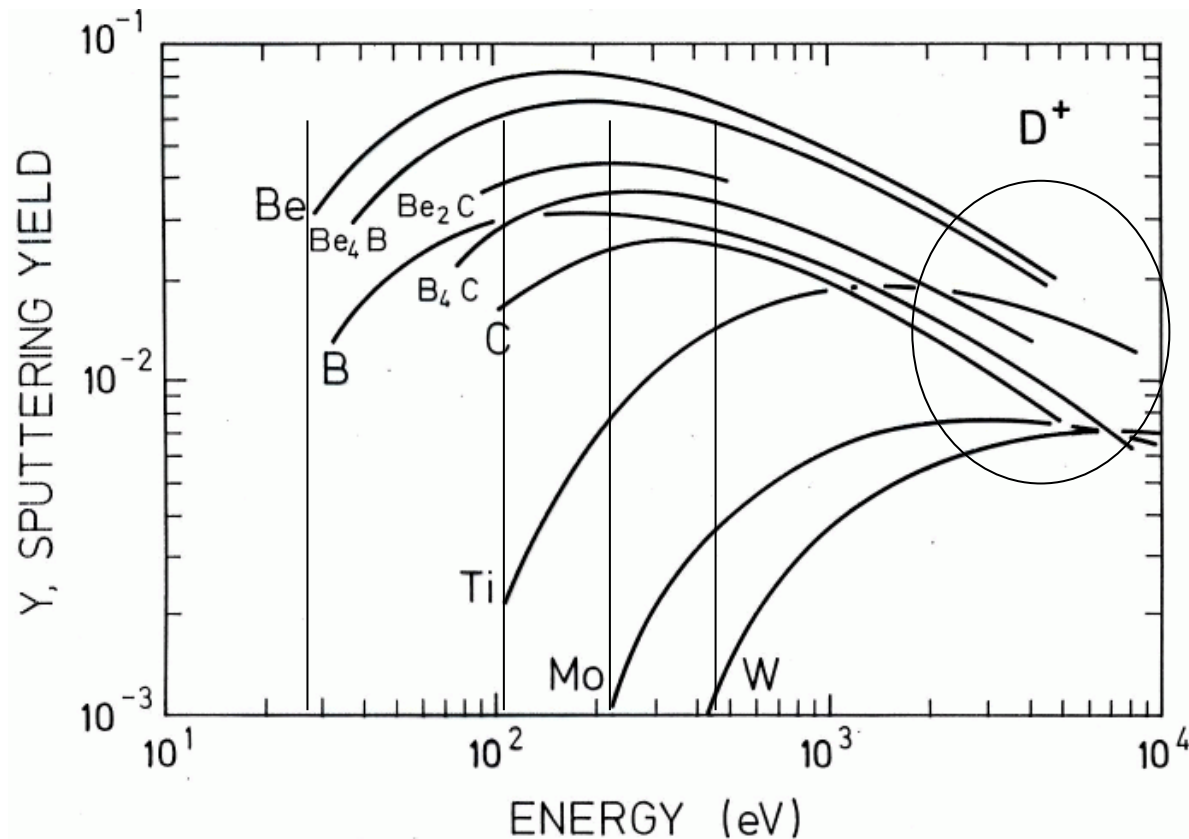


- In the threshold regime all experimental data show a similar energy dependence
- normalized energy scale
 $E' = E/E_{th}$
- Good fit to universal function with

$$Y(E') = Q(M1, M2, E_s) * f(E')$$

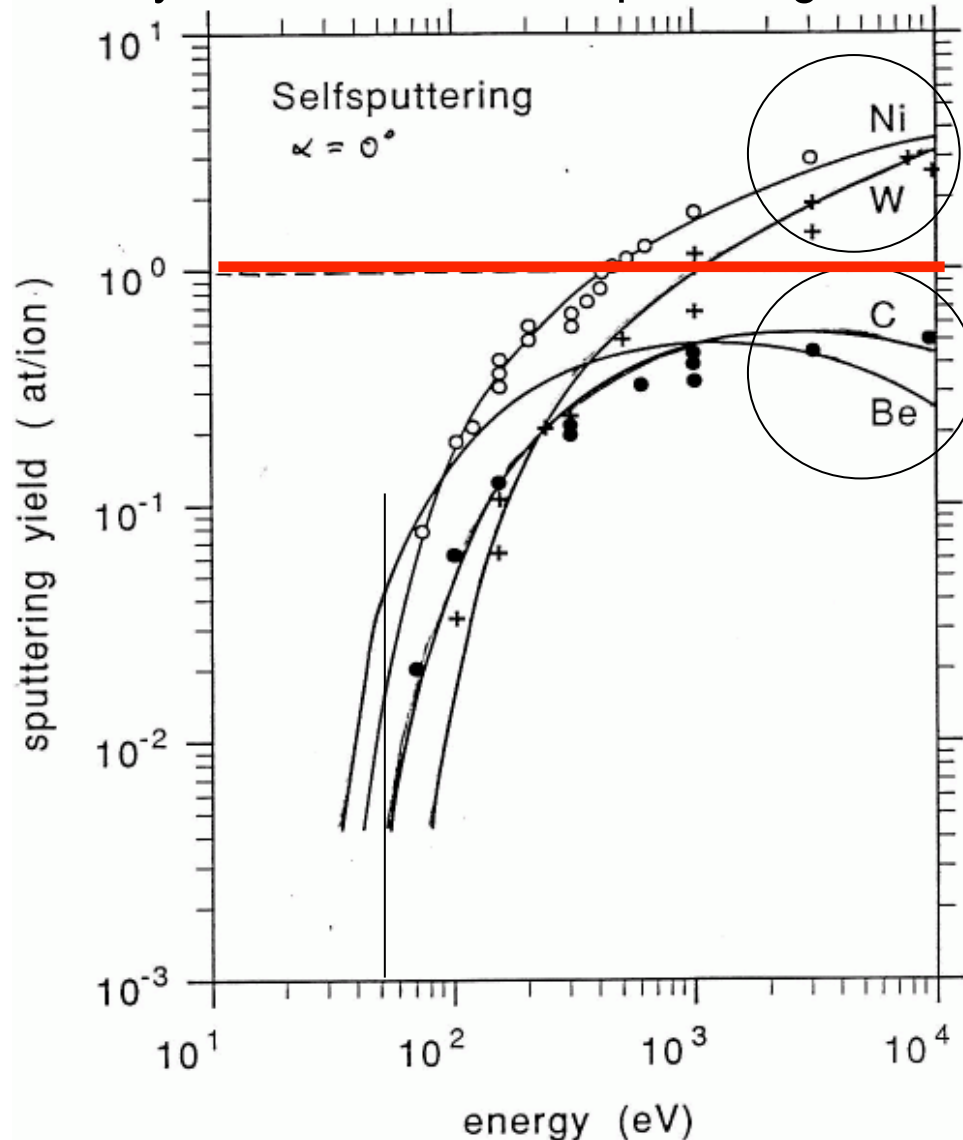
$$f(E') = \left(1 - \frac{1}{E'}\right)^{3.5}$$

Systematics for light ions



- Similar yields in isotropic cascade regime
- Strong influence of Z_2 on threshold energy E_{th}

Systematics for self-sputtering & heavy ions



- No dependence of E_{th} on target mass, but on surface binding energy E_s
- Strong dependence of yield on mass in isotropic cascade regime due to nuclear deposited energy.
- Most important is the yield range close to unity, as runaway impurity production may occur

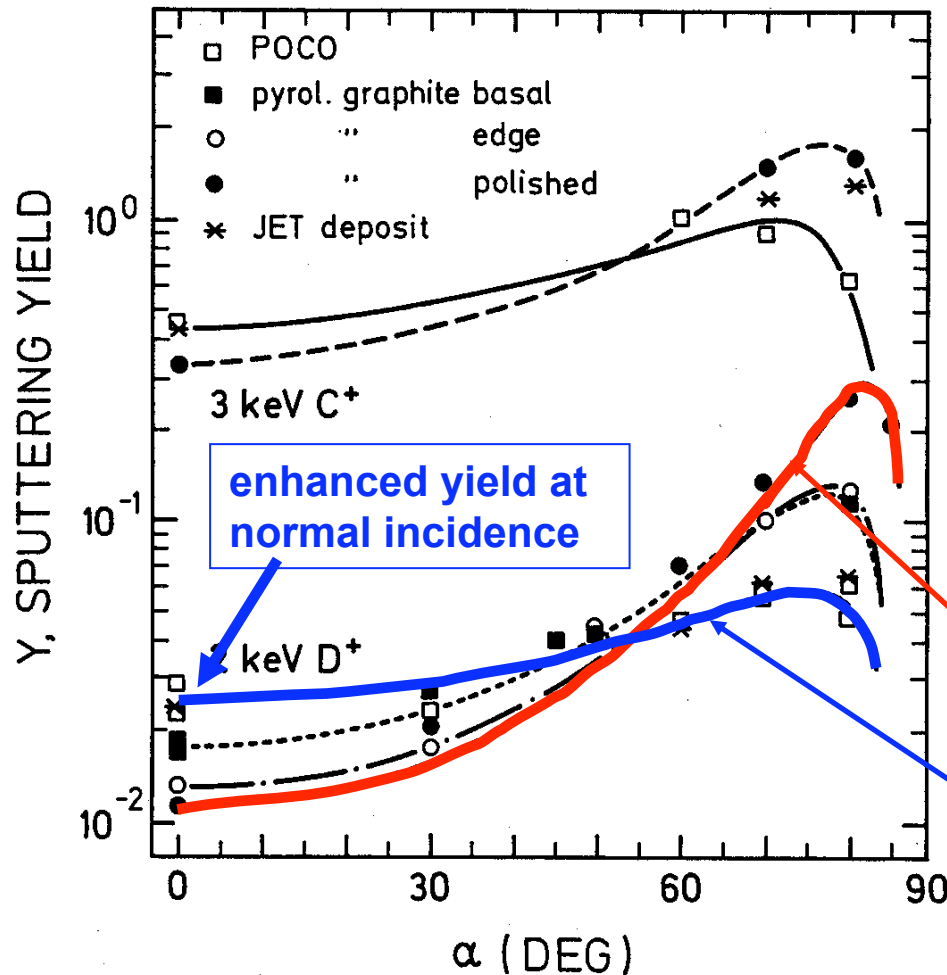
$$\Gamma_{Ero} = \Gamma_D Y_D + \Gamma_{Ero} Y_{self}$$

$$\Rightarrow \Gamma_{Ero} / \Gamma_D \equiv Y_{eff} = \frac{Y_D}{(1 - Y_{self})}$$

Physical sputtering



angular dependence



physical sputtering yield

depends on:

| angle of incidence

| target roughness

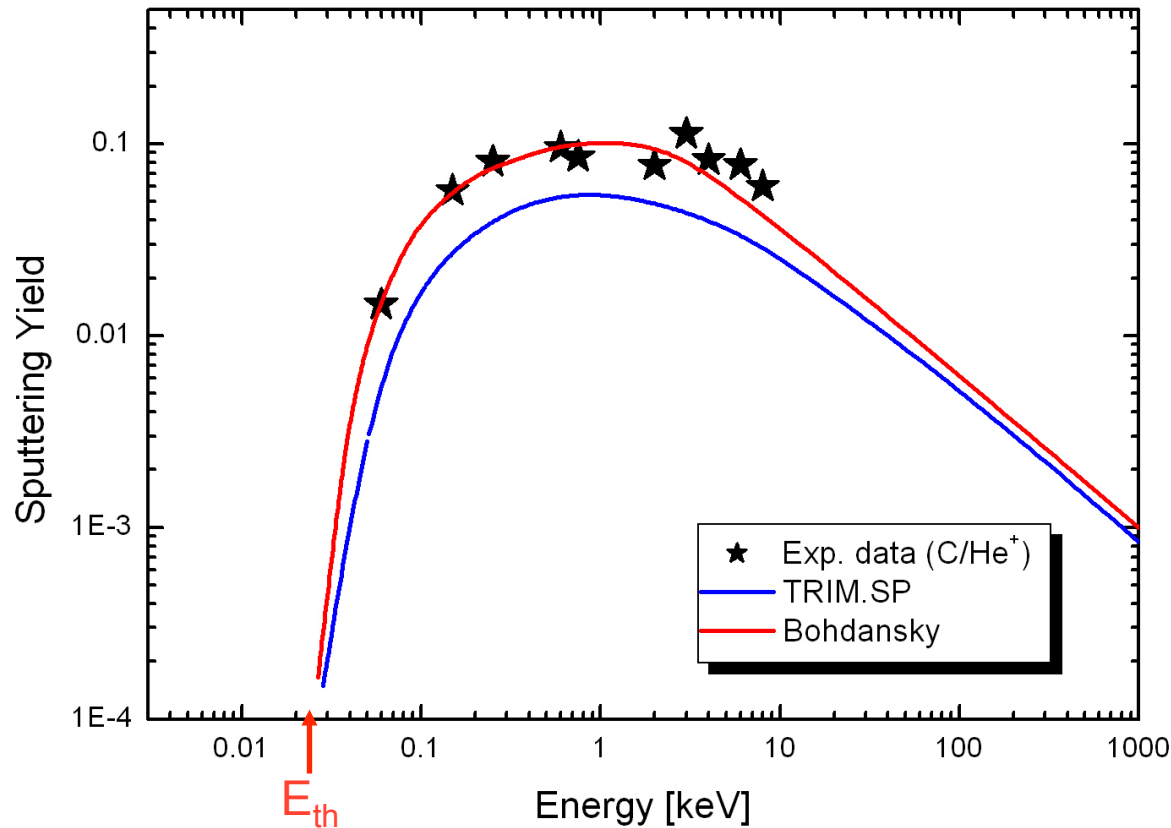
- Maximum at $\sim 70^\circ$
- For higher angles reflection increases \rightarrow sputtering decreases

highly-oriented, smooth graphite

rough graphite

J. Roth, W. Eckstein et al., J. Nucl. Mater. 179-181, 34 (1991)

quantitative description of physical sputtering



the experimental data is fitted with the **Bohdansky** formula:

$$Y = Q \cdot s_n^{TF} \cdot \left(1 - \left(\frac{E_{th}}{E_o} \right)^{2/3} \right) \cdot \left(1 - \frac{E_{th}}{E_o} \right)^2$$

$$s_n^{TF} = s_n^{TF}(\varepsilon)$$

$$\varepsilon = E_o \frac{M_{target}}{M_{ion} + M_{target}} \cdot \frac{a_L}{Z_{ion} \cdot Z_{target}}$$

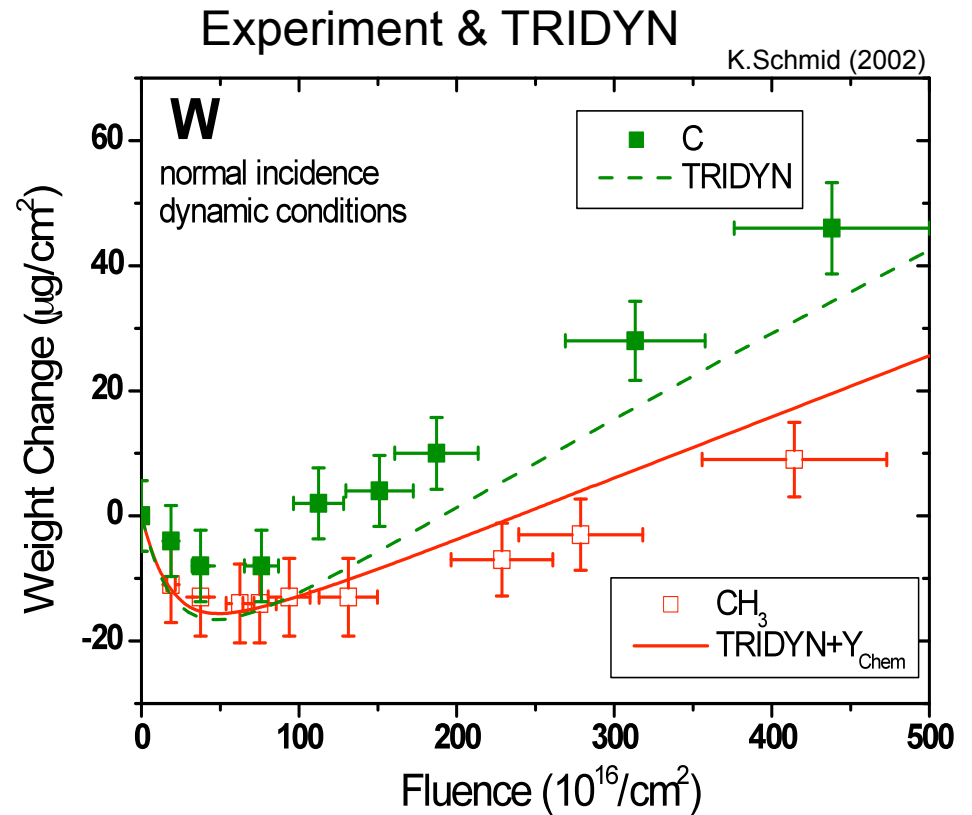
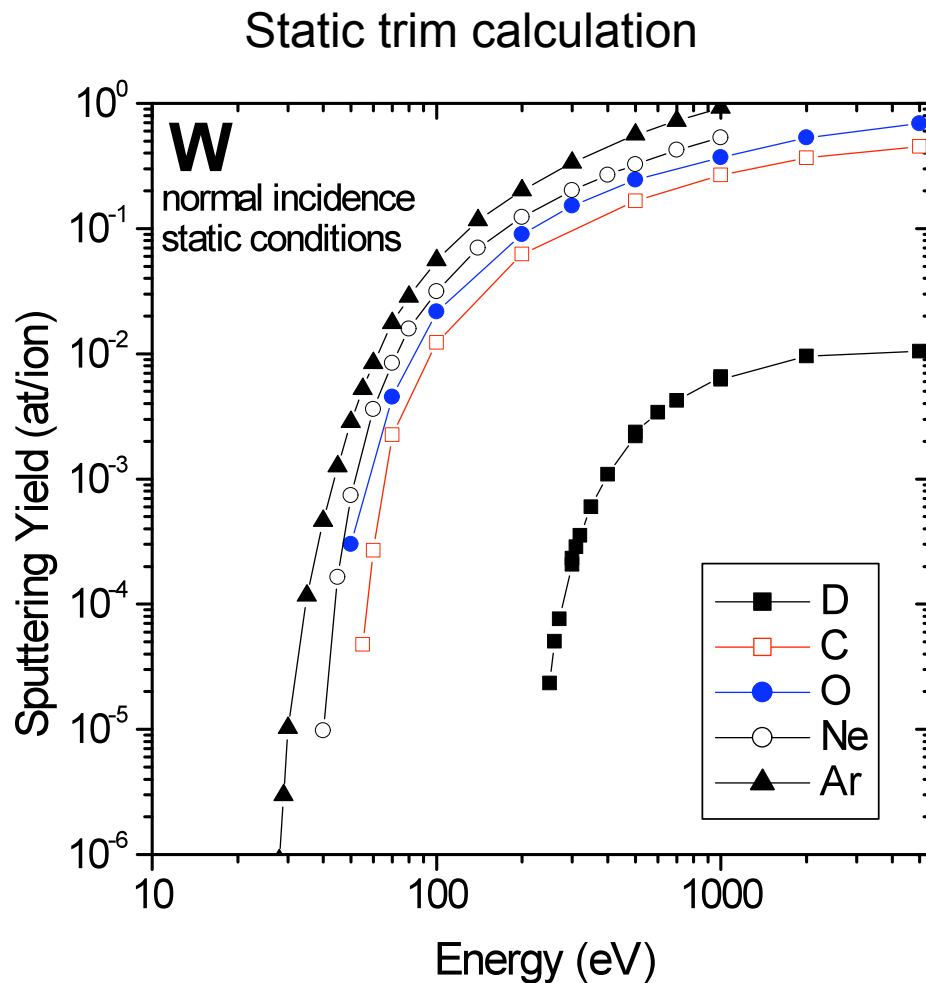
$$Q = 0,169 \text{ [atoms/ion]}$$

$$E_{th} = 25,4 \text{ [eV]}$$

J. Roth, E. Vietzke, A.A. Haasz; *Atomic and Plasma-Material Interaction Data for Fusion, Suppl. to Nuclear Fusion* **1** (1991) 63.

C. Garcia-Rosales, W. Eckstein, J. Roth; *J. Nucl. Mater.* **218** (1994) 8-17.

Sputtering by non-recycling ions



- For non recycling impurities deposition and layer growth lead to complex mixed material formation
- TRIM for static conditions
- TRIDYN for dynamic conditions

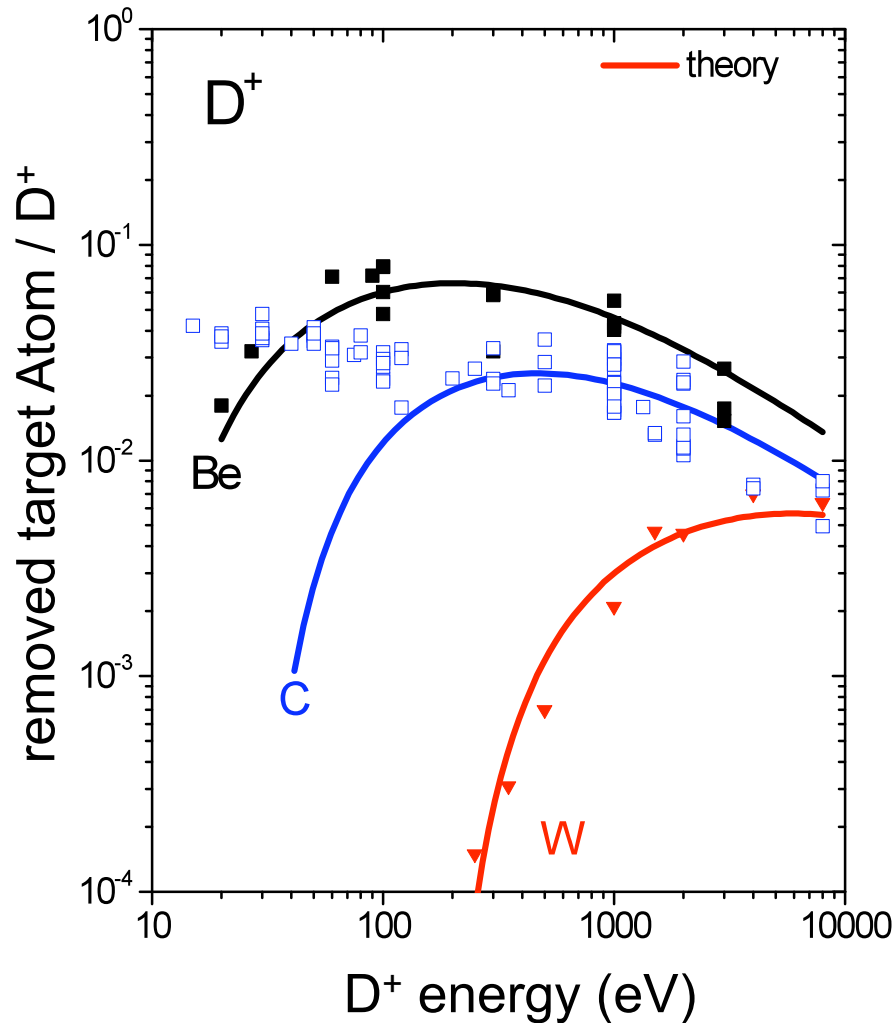
physical sputtering

- Physical sputtering is the ***kinetic ejection of surface atoms*** by incident energetic ions or atoms ***due to collision processes*** (playing pool with surface atoms).
- As surface atoms can escape only if it receives an energy larger than the ***surface binding energy***, a threshold energy for the incident particles is required.
- In fusion application sputtering by hydrogen and helium ions and atoms is important, but also the self-sputtering due to returning impurity atoms.

Chemical Erosion

- Chemical erosion originates from the formation and release of volatile molecules in the interaction of incident plasma particles and target atoms.
- In fusion application the formation of hydrocarbons in the interaction of hydrogen atoms with carbon surfaces is the dominant example of chemical erosion
- As chemical reactions are involved, chemical erosion shows a strong temperature dependence in contrast to physical sputtering.
- Chemical erosion is due to interaction of thermal atoms and does not require a threshold energy.

Chemical erosion at room temperature



- threshold behaviour for Be, W
- advantage for high-Z materials
- no threshold for C

⇒ what is different?

⇒ chemical reactions between D and C forming volatile hydrocarbons

chemical erosion

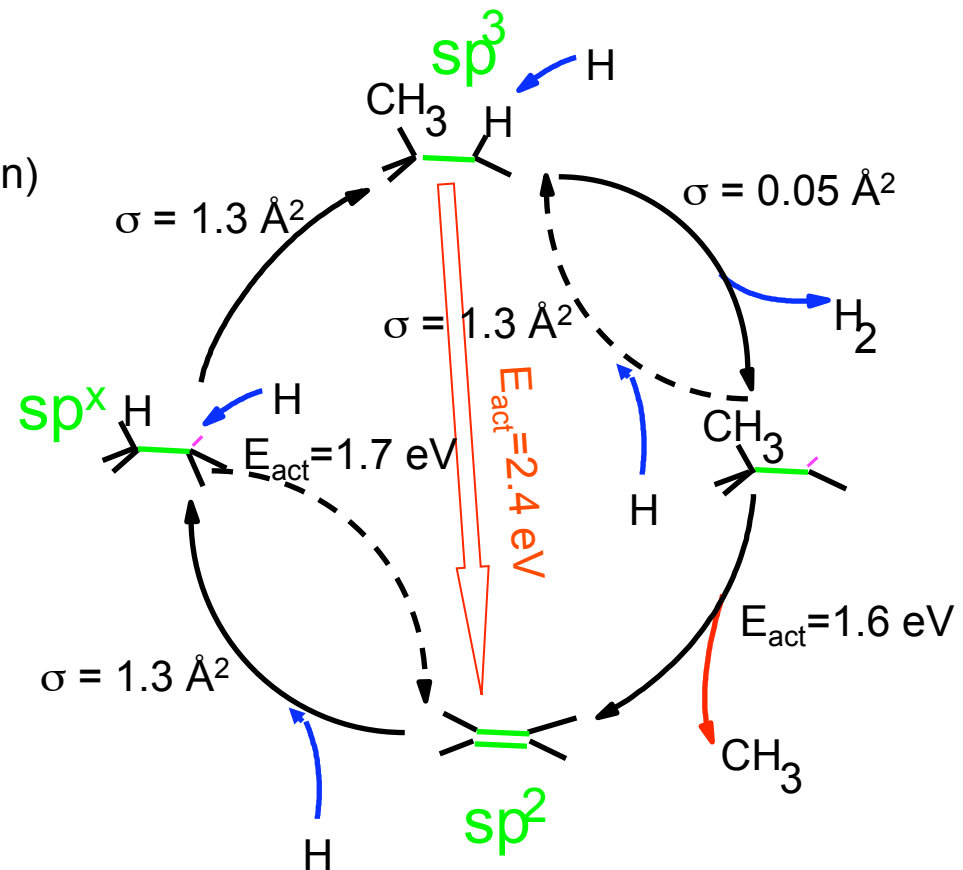
microscopic model for erosion of graphite by H_0

Hydration and erosion circle:

Horn et al., Chem. Phys. Lett. 231, 193 (1994)

Zecho et al. J. Phys. Chem. B 105 (2001).

- 1) chemisorption of H on sp^2 site
- 2) chemisorption of H on sp^x site (hydration)
- 3) abstraction of H to form H_2
- 4 a) thermal release of CH_3 radicals from activated sites above 400 K
- 4 b) chemisorption of H on sp^x site
- 5) relaxation back to sp^2 above 750 K
- 6) direct thermal decomposition to sp^2 above 900 K with $E_{act}=2.4$ eV



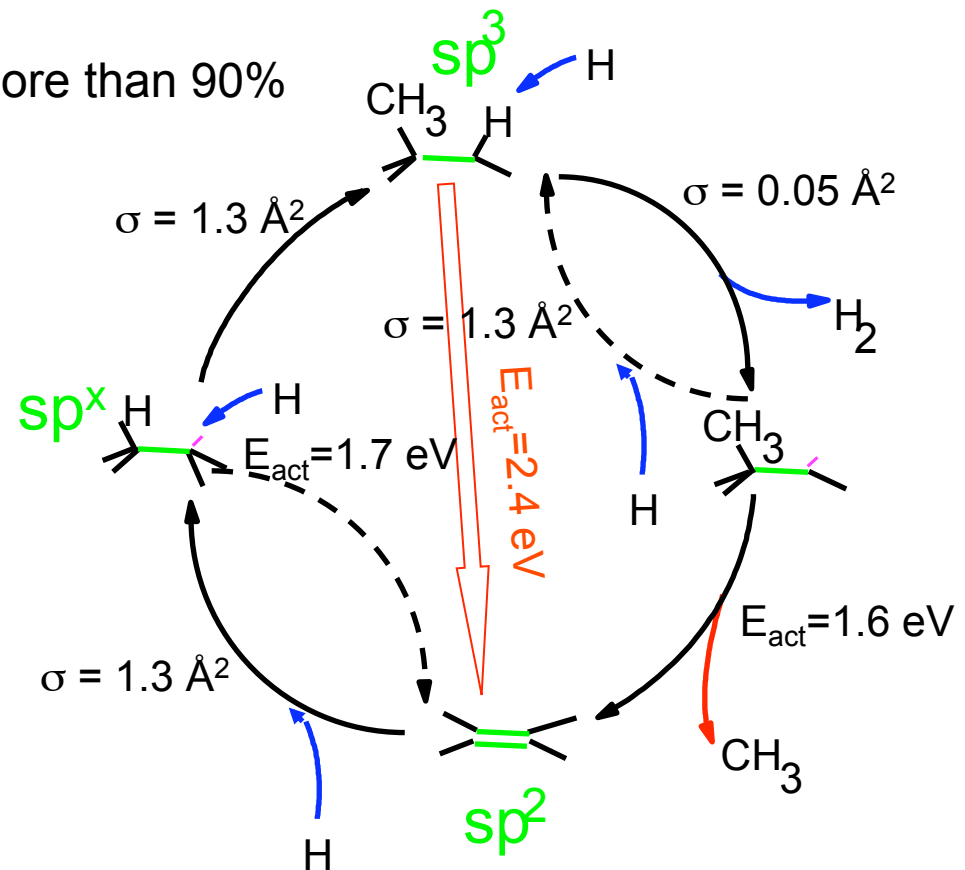
chemical erosion: microscopic model

Hydration and erosion circle:

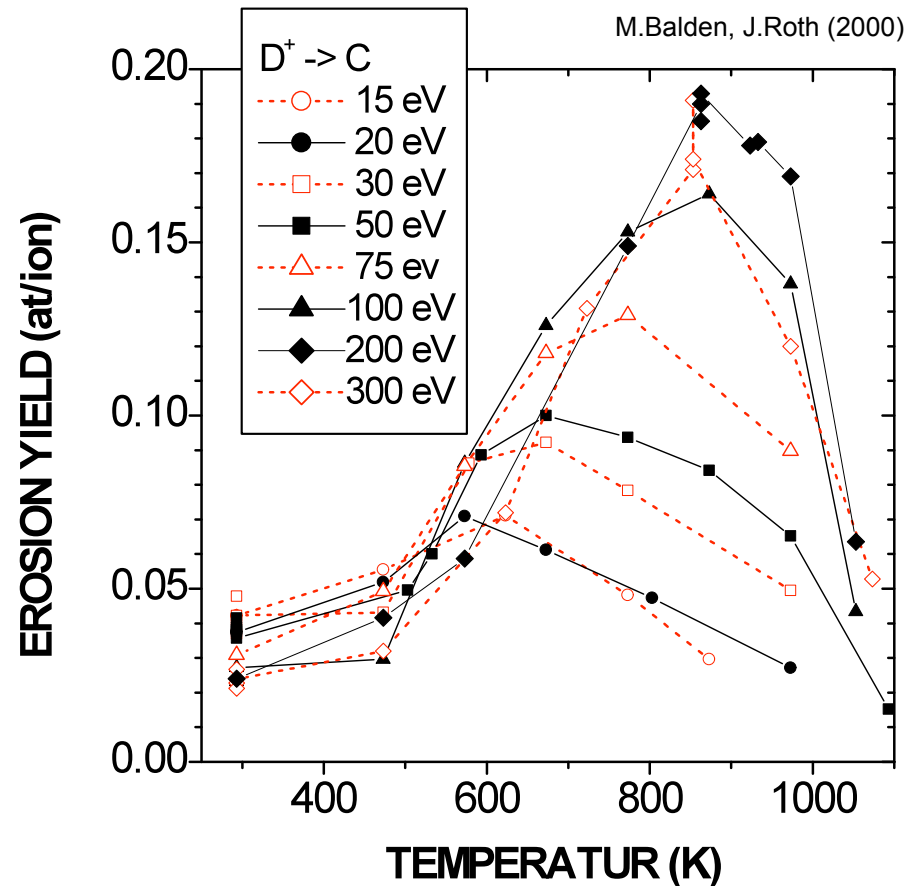
Horn et al., Chem. Phys. Lett. 231, 193 (1994)

Zecho et al. J. Phys. Chem. B 105 (2001).

- hydration at room temperature of more than 90% of all possible adsorption sites
- erosion maximum as function of temperature



Chemical Erosion at elevated temperatures

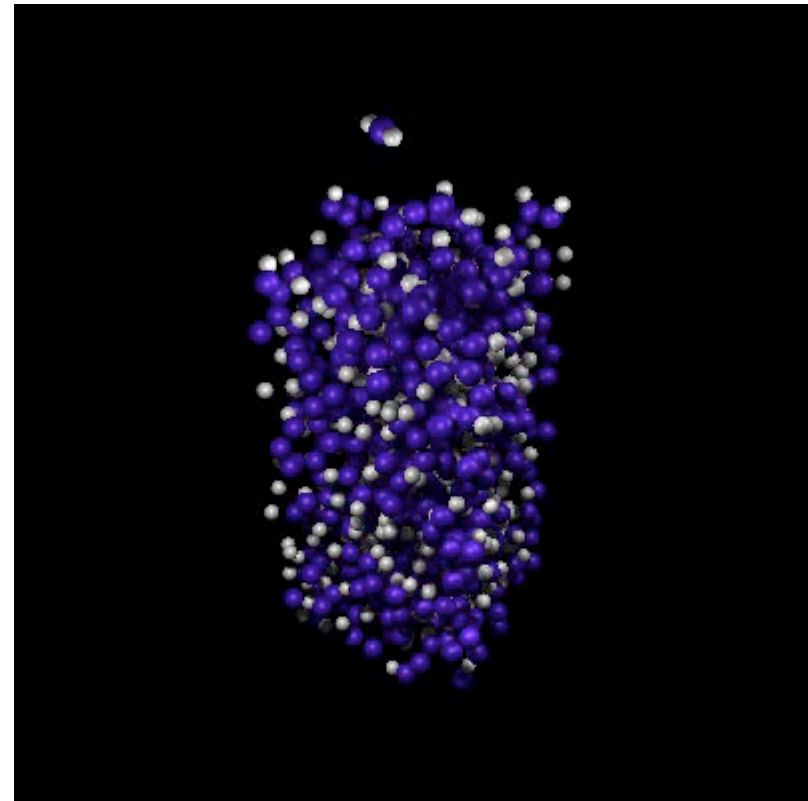


- „Basic“ understanding and modeling of chemical erosion
- First modeling attempts using MD codes
- ? Shift of erosion maximum with energy. (Different erosion products)
- ? Flux dependence of chemical erosion

chemical erosion

- chemical erosion originates from the formation and release of volatile molecules in the interaction of incident plasma particles and target atoms.
- in fusion application the formation of hydrocarbons in the interaction of hydrogen atoms with carbon surfaces is the dominant example of chemical erosion.
- as chemical reactions are involved, chemical erosion shows a strong temperature dependence in contrast to physical sputtering.
- chemical erosion can occur with low-energy ions or thermal atoms and does not require a threshold energy.
- erosion will only take place at the very surface (1.4 nm penetration depth) or at the end of range of energetic particles.

- **Chemical Sputtering** is a process whereby *ion bombardment causes or allows a chemical reaction to occur* which produces a particle that is weakly bound to the surface and hence easily desorbs in the gas phase.
 - Bombardment of carbon by hydrogen ions
 - Bombardment of carbon by noble gas ions in the presence of atomic hydrogen
 - Bombardment of carbon by noble gas ions in the presence of molecular oxygen (or water)
 - Etching of silicon by fluorine plasmas (this is by far the most important industrial plasma process)



it is not chemical erosion

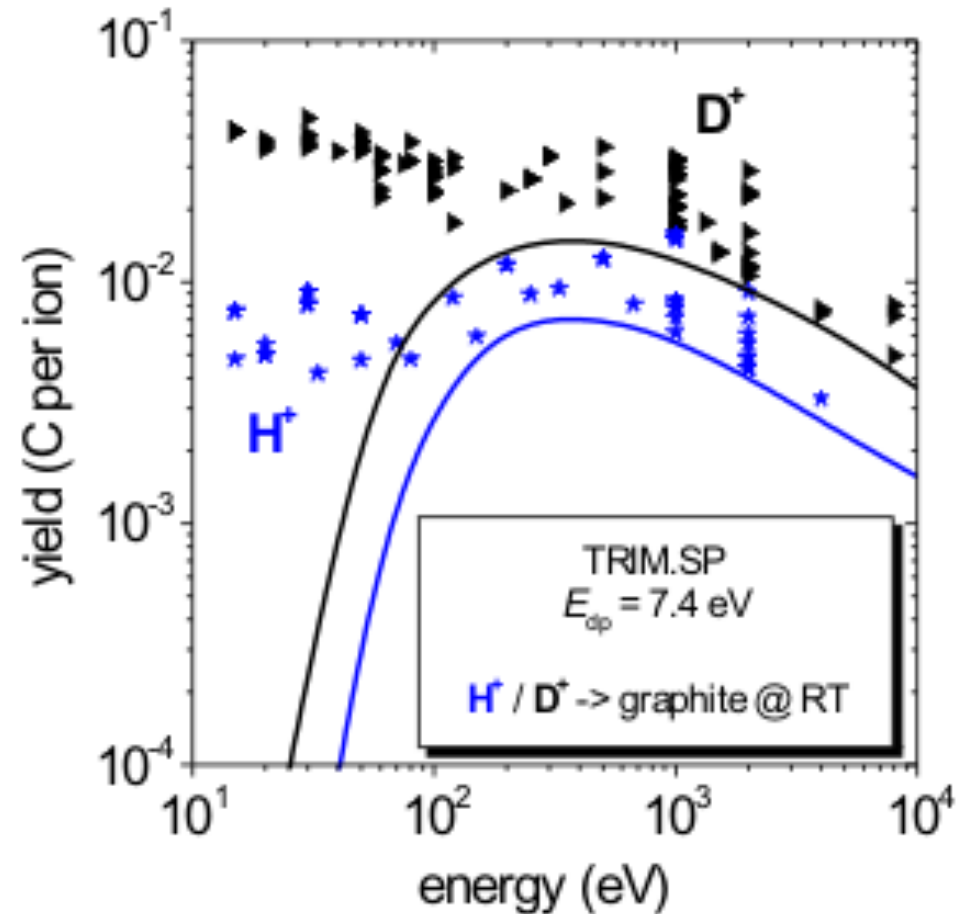
H^0 at $T > 400$ K with a max. at
 ≈ 650 K – 800 K

it is not physical sputtering

energetic ions $E > E_{th}$,
no chemistry

but its chemical sputtering

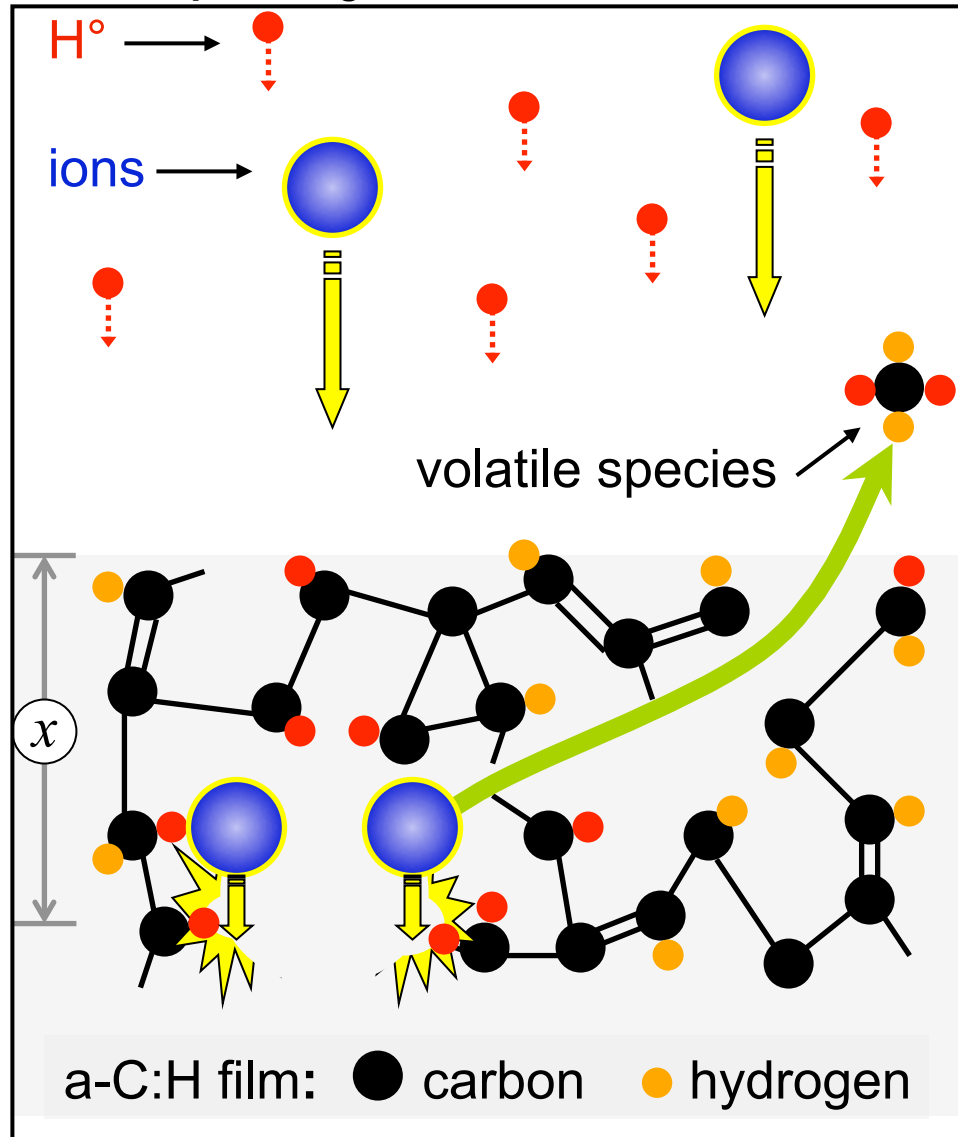
hydrogen ions, low-T, low-E



Data: M. Balden and J. Roth, J. Nucl. Mater. 280 (2000) 39–44

Chemical Sputtering

chemical sputtering mechanism



1. ions break C-C bonds

2. H° passivates broken bonds

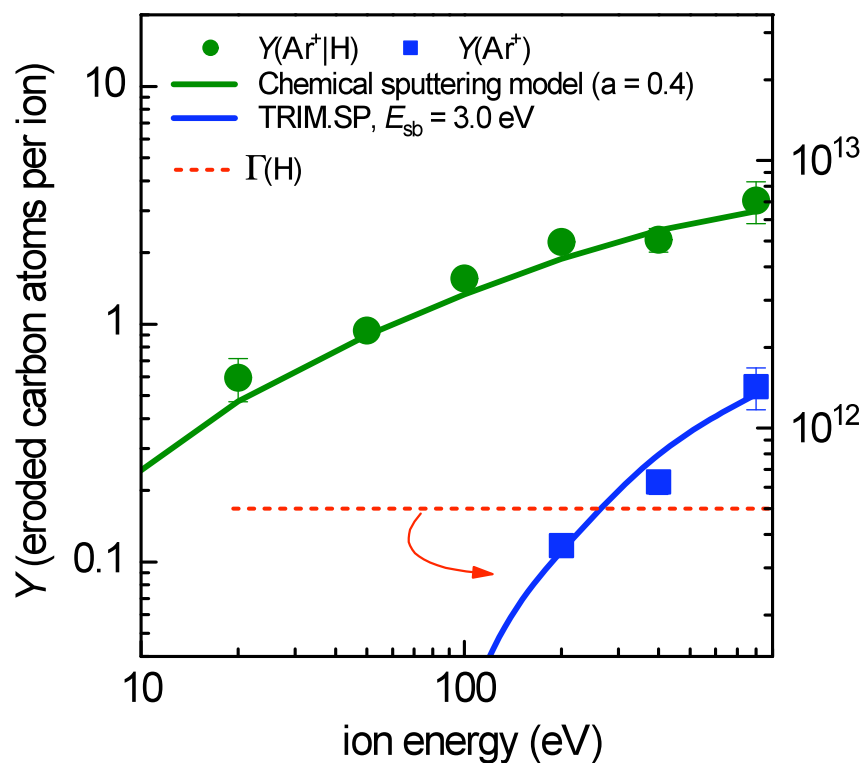
Repetition of 1 and 2

⊗

3. volatile hydrocarbons

diffusion to the surface

desorption



Erosion of a-C:H layers

Comparison of simple **physical sputtering** (blue symbols) due to Ar ions

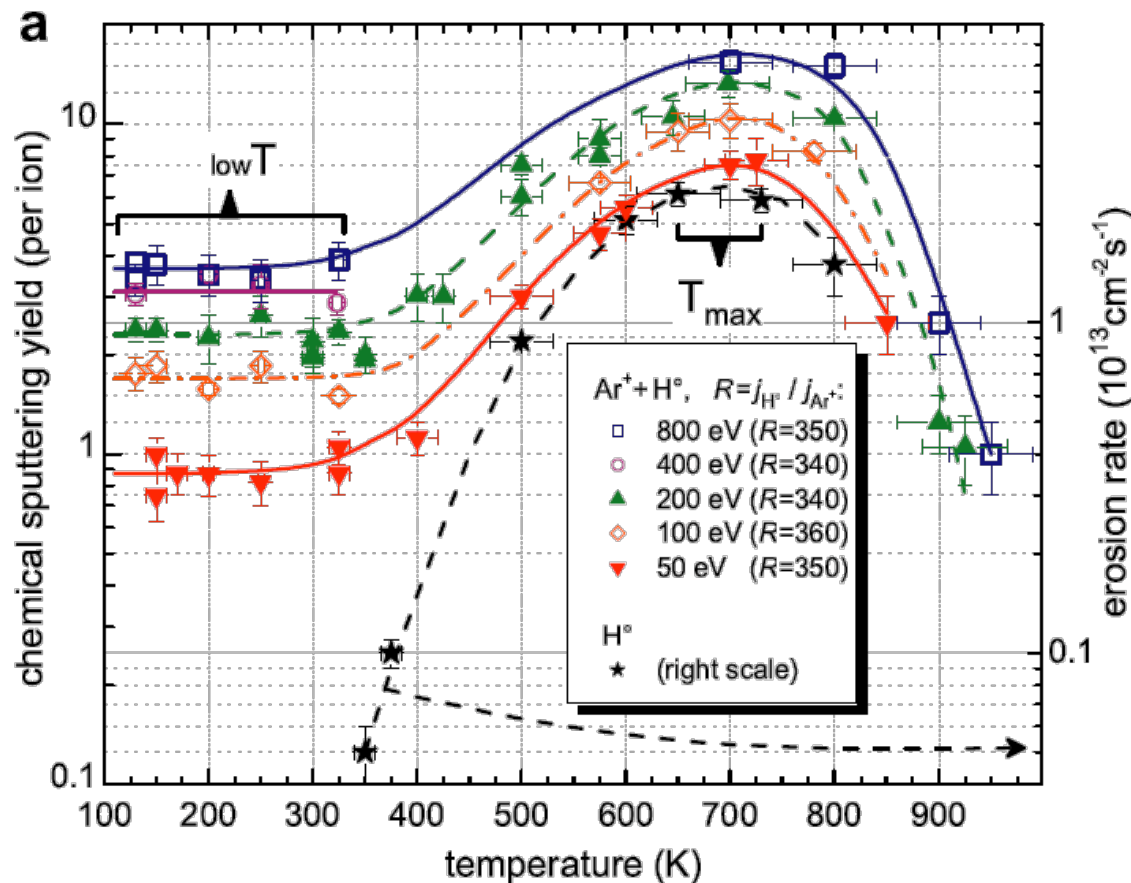
with

erosion due to simultaneous interaction of H and Ar⁺ (green symbols).

Simultaneous interaction leads to:

- Erosion even below threshold for physical sputtering (threshold energy for physical sputtering ≈ 60 eV)
- Enhanced erosion above 200 eV
- Erosion at 20 eV \rightarrow pure chemical erosion \Rightarrow '**chemical sputtering**'

Temperature dependence



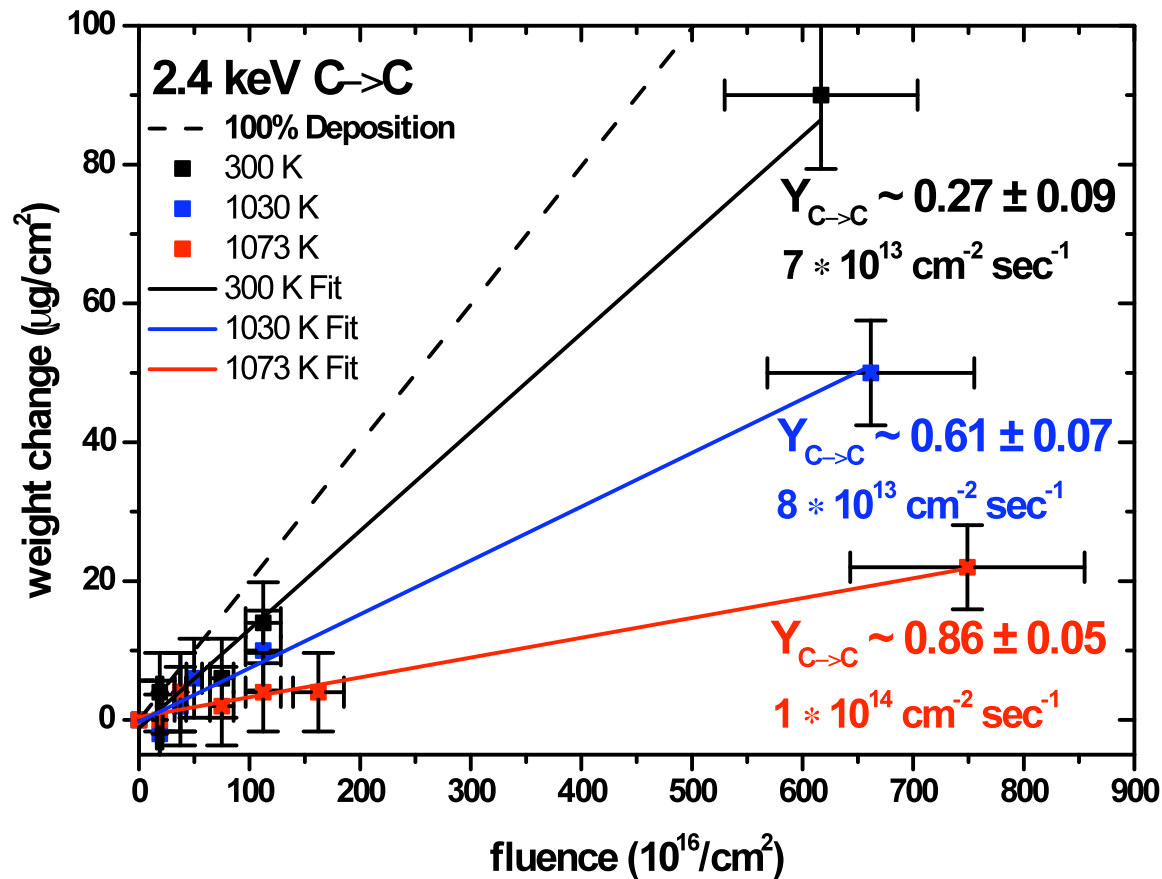
- Temperature dependence similar as for chemical erosion
- Radiation damage enhances chemical reactivity
- Value at T_{max} is larger than sum of chemical erosion and low temperature chemical sputtering

- Chemical Sputtering is a process whereby *ion bombardment causes or allows a chemical reaction to occur* which produces a particle that is weakly bound to the surface and hence easily desorbs in the gas phase.

- As for chemical erosion a basic model exists but there are still open questions:
 - temperature dependence of the erosion maximum not understood
 - decrease in erosion rate at high fluxes not understood

Radiation enhanced sublimation

- At high temperatures graphite exhibits an exponential increase in the erosion rate during energetic particle impact that can not be explained by sublimation



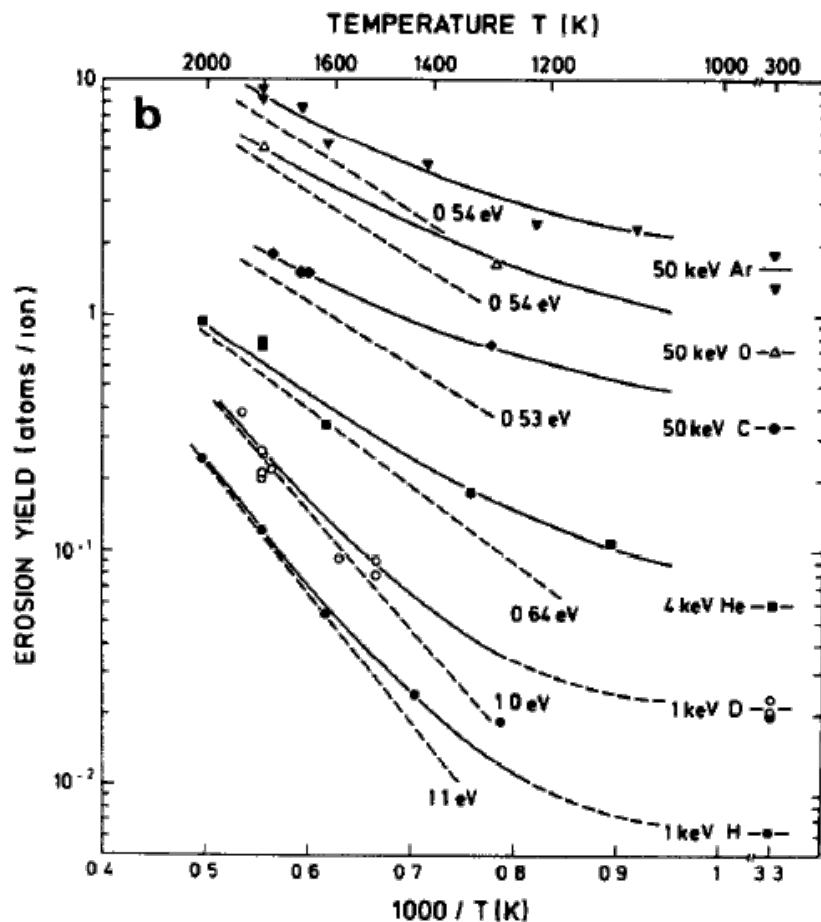
❖ carbon self sputtering

➤ around 1000 K onset of enhanced sputtering

Radiation enhanced sublimation



Exponential fits yield activation energies



- Arrhenius activation energies are well below the graphite sublimation energy.
- Process occurs for both inert and reactive sputtering species
- Experiments indicate that eroded species have thermal energies

RES mechanism

- Energetic ion bombardment generates vacancy/interstitial pairs (Frenkel pairs)
- vacancies & interstitials are highly mobile (diffusion) at elevated temperatures
 ← slower
- Interstitials annihilate with vacancies and at the surface where they can sublime
- Vacancies annihilate with Interstitials and at the surface



➤ Diffusion trapping model describes RES:

$$\frac{\partial C_i}{\partial t} = D_i \frac{\partial^2 C_i}{\partial x^2} - \underbrace{D_i a N C_i C_v}_{\text{Annihilation}} + \underbrace{\Phi_D}_{\text{Source}}$$

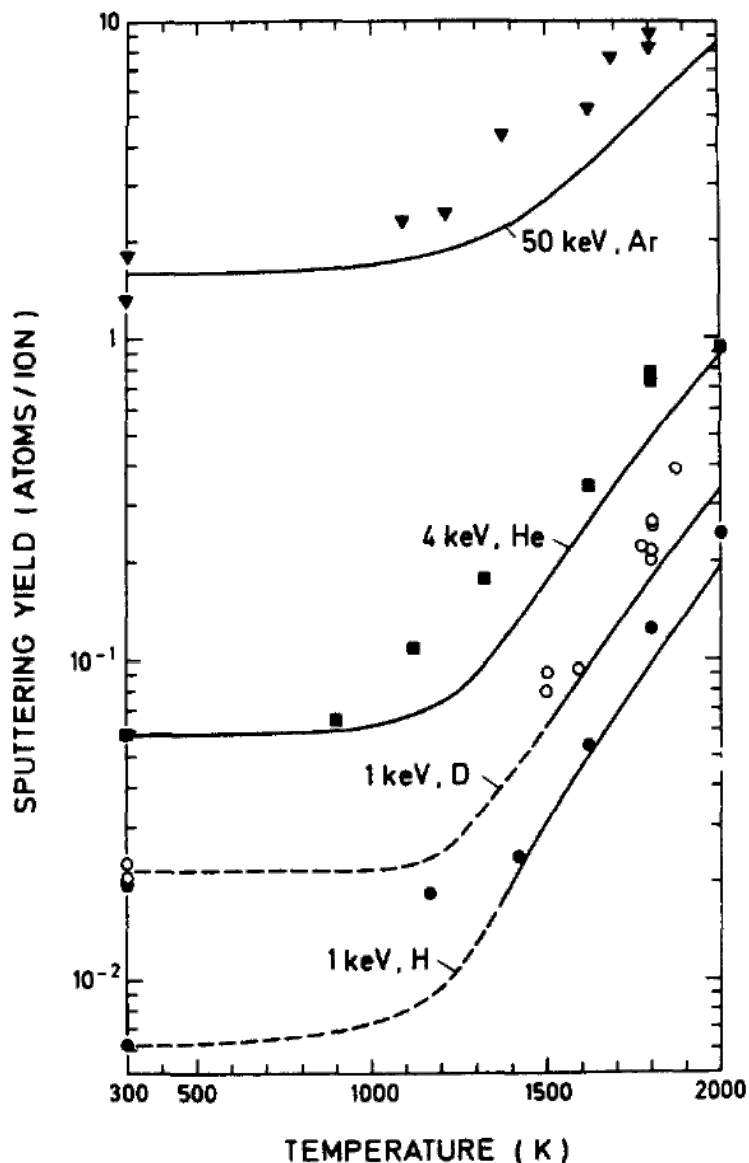
$$\frac{\partial C_v}{\partial t} = D_v \frac{\partial^2 C_v}{\partial x^2} - \underbrace{D_v a N C_v C_i - D_v a N C_v^2}_{\text{Annihilation}} + \underbrace{\Phi_D}_{\text{Source}}$$

➤ Net RES erosion flux is given by the diffusive flux of interstitials to the surface

$$\Gamma_{\text{Res}} = D_i \frac{\partial C_i}{\partial x}$$

K. Schmid 2009

Radiation enhanced sublimation



- Diffusion trapping model quantitatively describes RES
- Damage profile calculated by TRIM.
- Under fusion conditions the influence of RES is not very pronounced:
 - At high fluxes the vacancy concentration becomes very high leading to fast annihilation of the more mobile interstitials.
 - For low particle energies close to the damage threshold no Frenkel pairs are created.

- At high temperatures graphite exhibits an exponential increase in the erosion rate during energetic particle impact that can not be explained by sublimation
- A model using the sublimation of weakly bonded surface defects, quantitatively describe the process.
- Similar effects are also seen for metallic targets at very high fluxes and temperatures. ← more relevant for fusion.

- As atoms are eroded from the first wall they enter the plasma
- In the plasma they are ionized and transported throughout the machine.
- What how does that affect the plasma ?

Impact of impurities on fusion plasmas



In 1957 Lawson introduced power balances:

Break-even: the fusion power equals the loss by radiation, and by transport (diffusion, convection, charge-exchange):

$$P_{\text{fus}} = n_D \cdot n_T \cdot \langle \sigma \cdot v \rangle \cdot E_{\text{fus}} \geq$$

$$P_{\text{bremsstrahlung}} = c_1 \cdot n_e^2 \cdot Z_{\text{eff}} \cdot (kT)^{1/2} + P_{\text{loss}} = 3 n kT / \tau_E$$

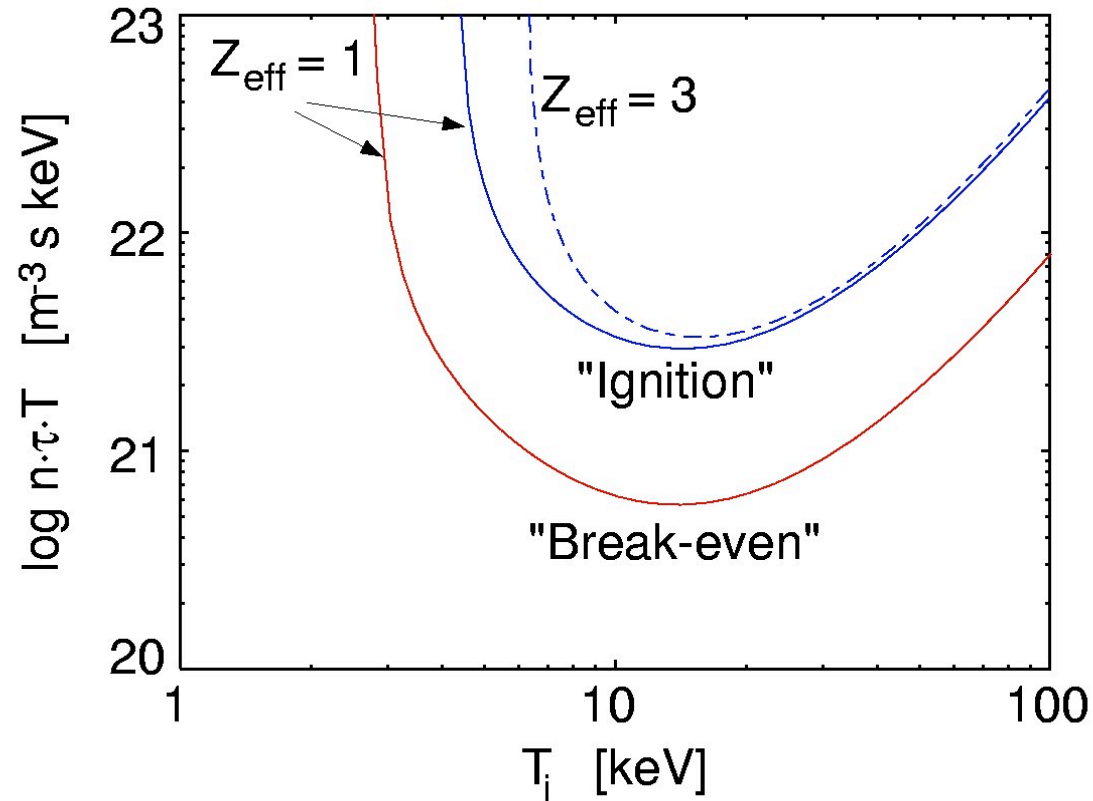
where E_{fus} is the α particle heating, $c_1 = 5.4 \cdot 10^{-37} \text{ Wm}^3 \text{keV}^{-1/2}$, and $Z_{\text{eff}} = \sum f_i Z_i^2$ is the effective plasma charge

with $n_D = n_T = n/2$, and $T_i = T_e = T$ we find a condition for the fusion product $n\tau T$:

$$n \tau T = \frac{12 (kT)^2}{\langle \sigma \cdot v \rangle \cdot E_{\text{fus}} - 4 c_1 Z_{\text{eff}} (kT)^{1/2}}$$

Ignition Criteria

- **Ignition:** The neutrons leave the plasma, the α -particles are confined and heat it. Only their energy should enter the balance! $E_{\text{fus}} \rightarrow E_{\alpha}$



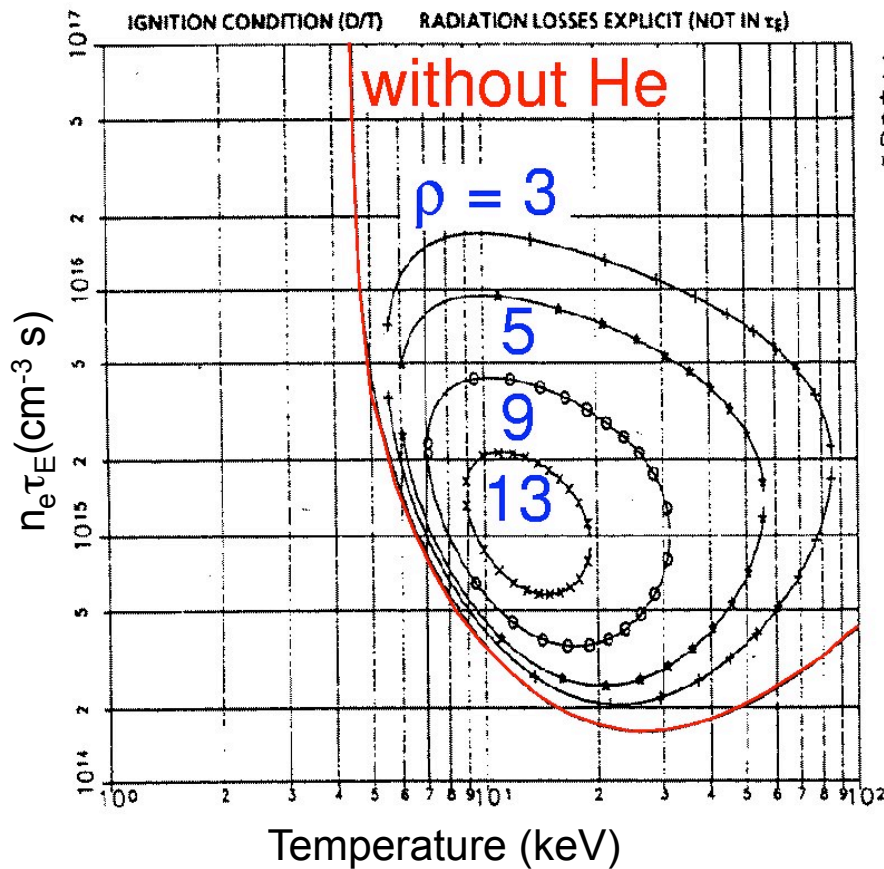
Impact of impurities on fusion plasmas



- The α -particles also dilute the plasma, as they are intrinsically coupled to fusion power ($3.53 \cdot 10^{11}$ atoms/s/W).

⇒ upper limit for particle confinement time

For steady state conditions, power and particle balances have to be solved simultaneously .
 → closed curves, parameterized by the He-confinement time



$$\rho_{\text{He}} \propto \tau_{\text{He}}^* / \tau_E$$

τ_{He}^* global particle confinement time

τ_E energy confinement time

here: $Z_{\text{eff}} = 1$ (no other impurities)

D. Reiter et al.
 Nuclear Fusion 30 (10), 2141 (1990).

Impact of impurities on fusion plasmas



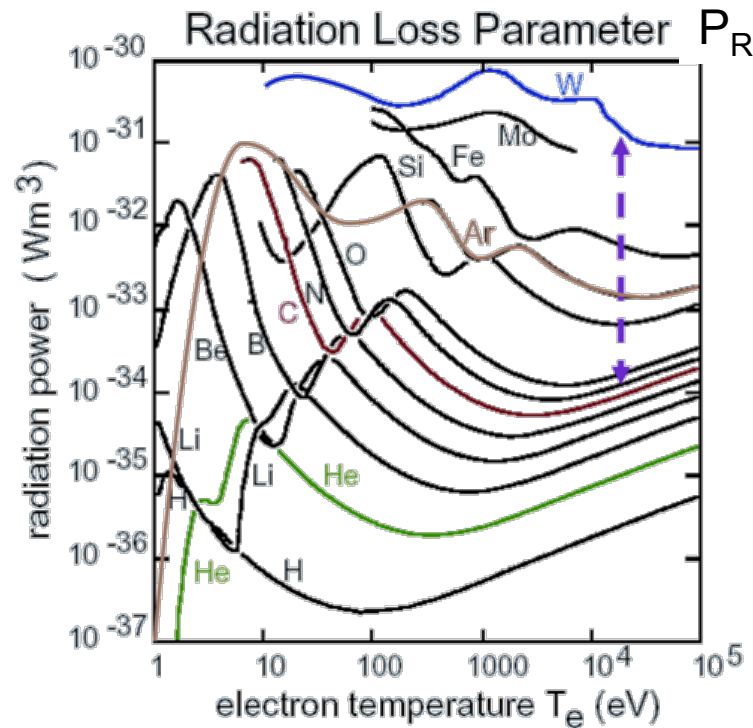
• Other impurities have a similar impact as He

- Dilution of fuel ions

$$n_i = n_e - Z n_z = n_e (1 - Z c_z)$$

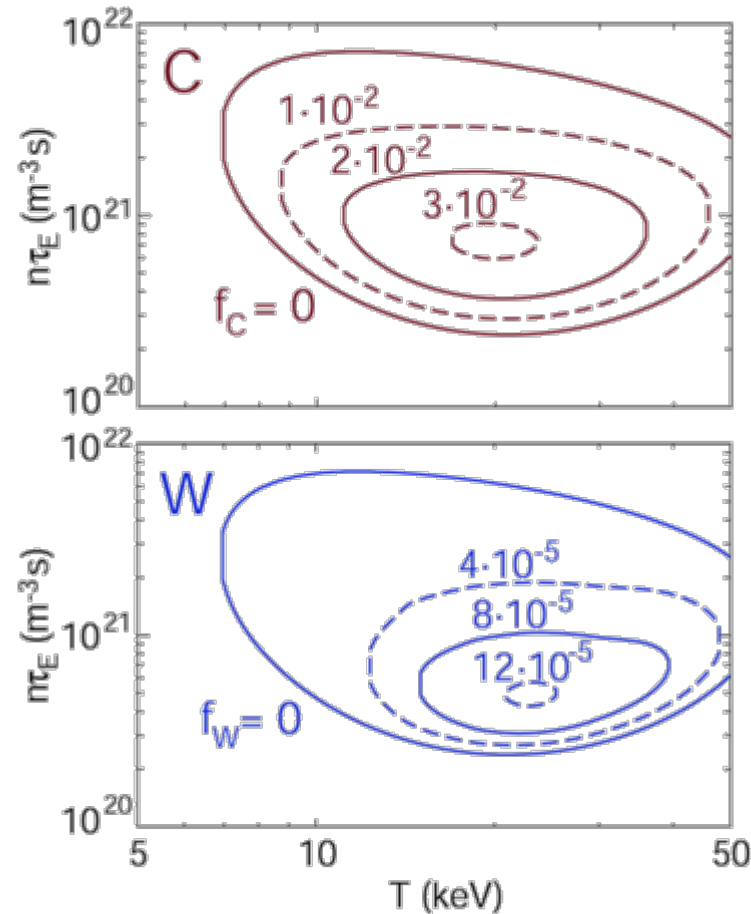
- Energy loss by line radiation

$$P_{\text{rad}} = n_e n_{\text{imp}} P_R$$



R. Neu, R. Parker, T. Pütterich

Ignition condition



R. Neu

➤ No ignition for core W conc. $> 10^{-4}$

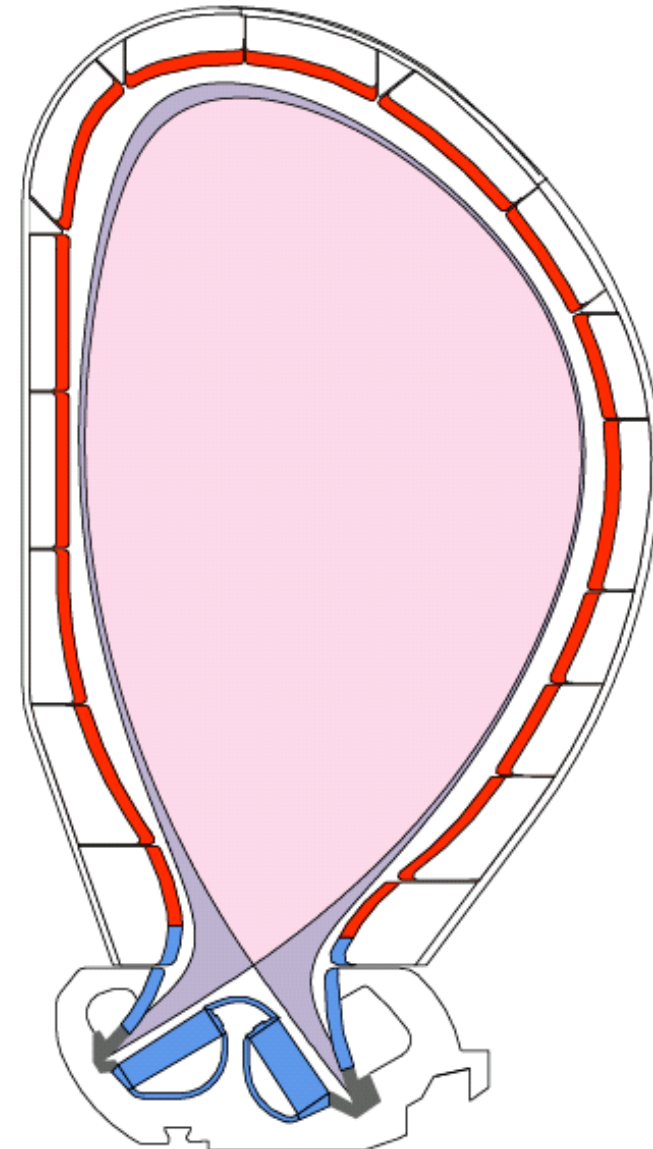
Material selection for ITER

- Main chamber first wall
 - Inevitably erosion by CX-neutral impact
 - Low particle flux and power load
 - Minimize radiation losses

⇒ Beryllium
- Divertor wall
 - Particle energy < 200 eV
 - Medium particle flux and power load
 - Use high sputtering threshold

⇒ Tungsten
- Target plates
 - Particle energy < 100 eV
 - High particle flux and power load
 - Extremely high transient power load
 - No surface melting

⇒ Carbon - CFC



- As atoms are eroded from the first wall they enter the plasma
- In the plasma they are ionized and transported throughout the machine.
- In the plasma they radiate energy through line radiation and Bremsstrahlung.
- They also dilute the plasma.
- The radiative loss of energy from the plasma and its dilution through these impurities has fundamental implications for the operation of a fusion reactor!
- The erosion of wall components poses a lifetime problem
- The co-deposition of impurities (mainly C) with fuel ions poses a radiation hazard