

Influence of the β energy decay spectrum and particle size on the self-charging rate of radioactive aerosol

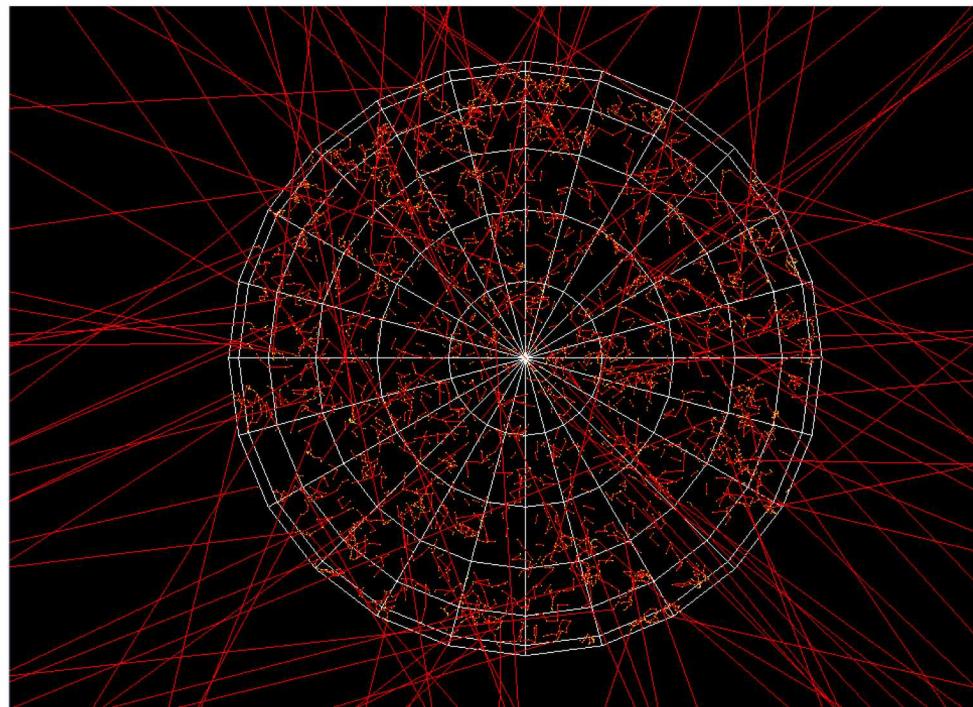
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What is self-charging of radioactive aerosol?

m : residual charge left on aerosol after 1 decay

$m = 1$ for β decay

- Reasonable assumption for classical radionuclides considered
- ^{198}Au (Yeh et al. 1976)
- ^{137}Cs (Gendsarmes et al. 2001)

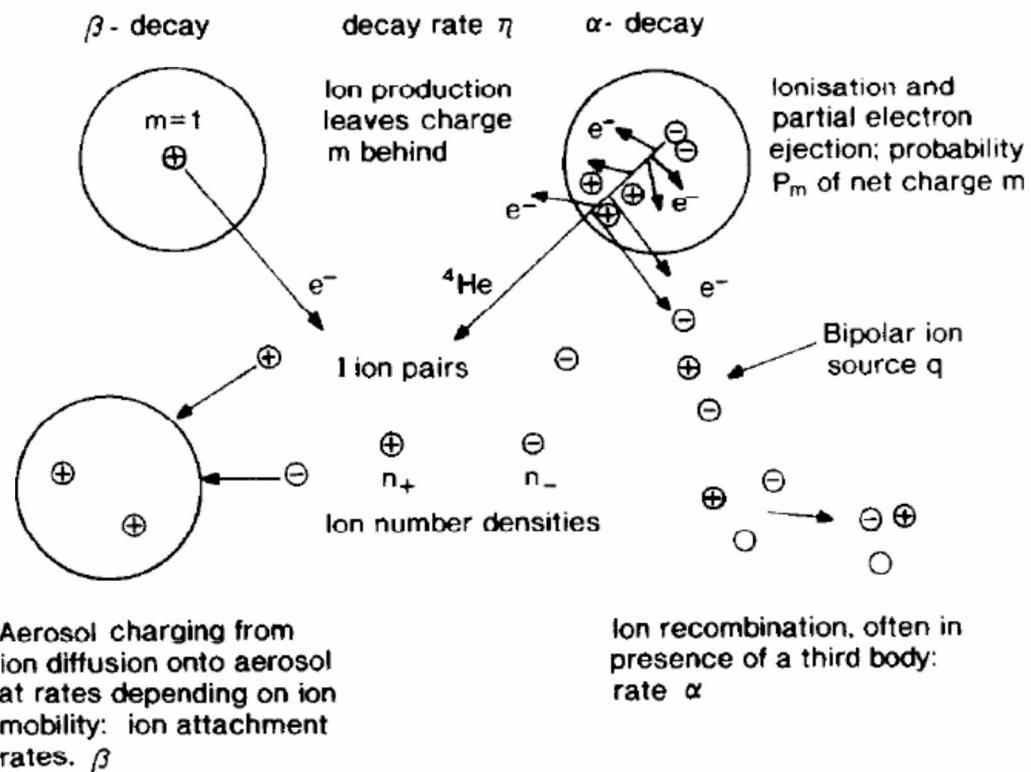


Fig. 1. Processes leading to the charging of radioactive aerosols.

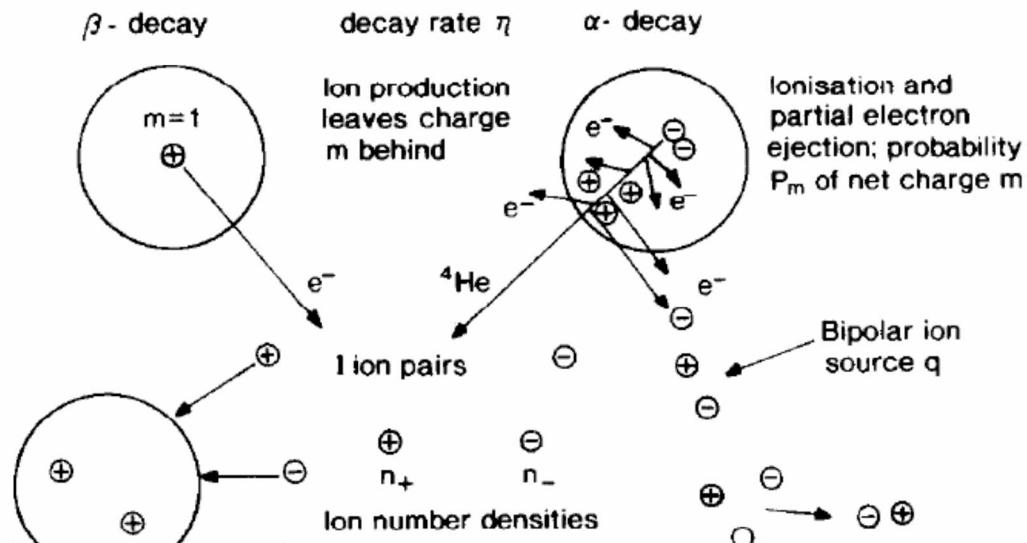
Clement and Harisson, JAS 1992

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- Renewed interest in electrostatic effects calculation related to radioactive aerosols, especially in the environment after the Fukushima accident (Dépée *et al.*, 2019)
- Lack of quantitative data on the mean value for m when the full energy spectrum of beta decay is considered !

How to relate Bq to charging rate ?

Assumption : Activity (Bq) = self-charging rate (s^{-1})[‡]

- PROS** Nuclide mean β energy → electron path length in matter **larger** than the particle diameter
- CONS** No study have ever taken in account the full β energy spectrum

Nuclide	Mean β energy [†] (keV)	Path length in SiO_2^* (μm)	
		with mean β	with full β spectrum
$^{137}_{55}\text{Cs}$	171	185	56
$^{132}_{53}\text{Te}$	80	52	26
$^{131}_{54}\text{I}$	202	241	54
^3_1H	6	< 1	< 1

[‡] Ivanov & Kirichenko, 1970 ; Yeh et al., 1976 ; Clement & Harisson, 1992, Gensdarmes et al., 2001

[†] Mean β energy = $E(\beta_{\max})/3$, $E(\beta_{\max})$ from LaraWeb. CEA/LNHB

^{*} Calculated from NIST/ESTAR, in CSDA approach. 10.18434/T4NC7P

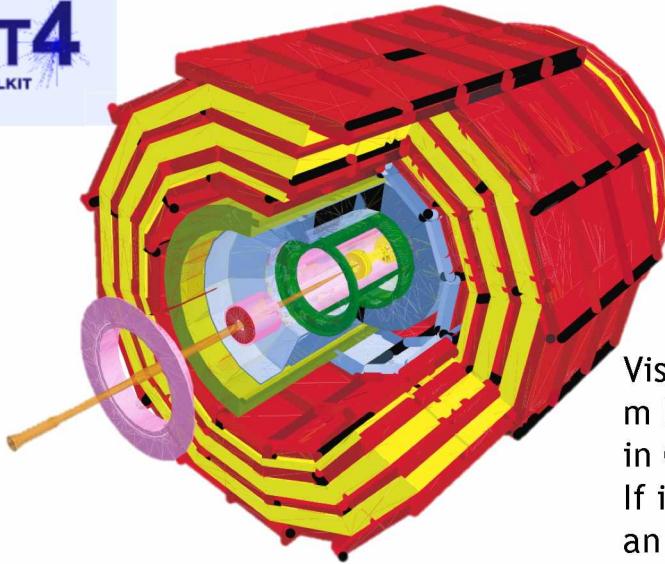
Electrons energies - Three cases

Previous and present ways to take account of β energies

1. Electron energy: single value = $\text{mean}(E(\beta)) \approx E(\beta_{\max})/3$
 - Kim et al. 2017
 - Without any information, **mean** is a pretty good assumption.
2. Electron energy: single value = $E(\beta_{\max})/4$
 - Gensdarmes et al. 2001
 - More representative has it takes into account the higher coefficients of linear energy transfert in matter for low energy electrons of the spectrum.
3. This study, electron energy: **full β energy spectrum**

How to use the full β spectrum ?

→ software for simulation of the passage of particles through matter !



Visualisation of the CMS detector. 15 m high, 21 m long, totally described in Geant4...
If it can handle this, we can describe an aerosol !

Geant4 is a toolkit for the simulation of the passage of particles through matter. Its areas of application include high energy, nuclear and accelerator physics, as well as studies in medical and space science. The three main reference papers for Geant4 are published in Nuclear Instruments and Methods in Physics Research A 506 (2003) 250-303, IEEE Transactions on Nuclear Science 53 No. 1 (2006) 270-278 and Nuclear Instruments and Methods in Physics Research A 835 (2016) 186-225.

Simulations variables for aerosols case study

Particle material	Tungsten, Iron, Beryllium
Particle density	19.3, 7.87, 1.85 g/cm ³
Particle diameter	20 nm → 200 µm
Electron energy	1 → 1200 keV
β spectrum †	³ H, ⁶⁰ Co, ¹³⁷ Cs, ¹³² Te

† ICRP, 2008. Nuclear Decay Data for Dosimetric Calculations. ICRP Publication 107. Ann. ICRP 38 (3)

Single sphere in vacuum

Material : Tungsten

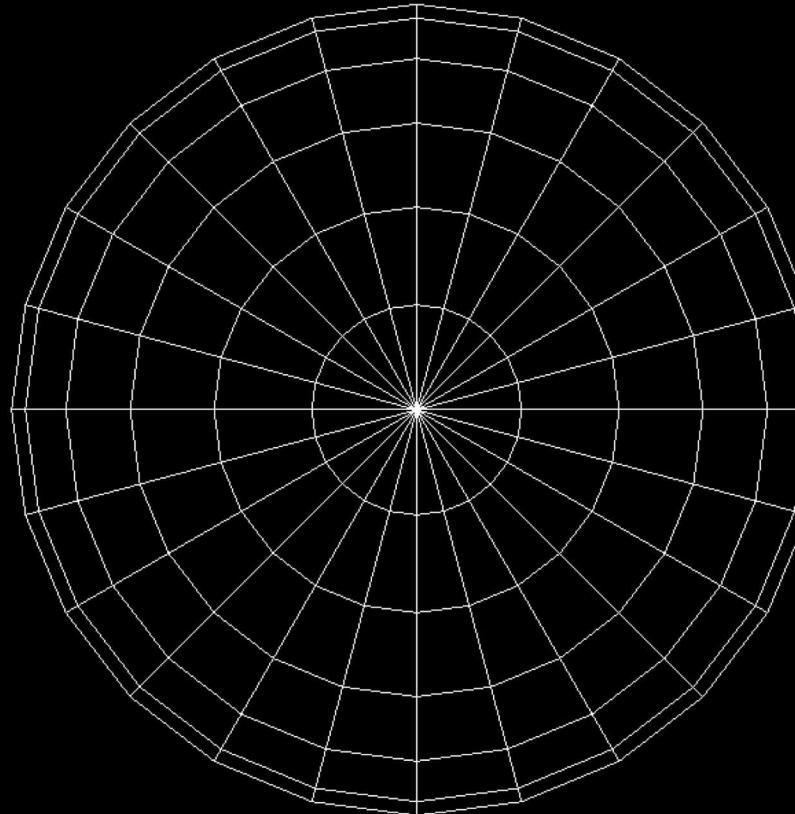
Diameter : 1 μm

Particle :

Energy :

e^- total :

e^- out :



Wireframe visualisation from Geant4

Single sphere in vacuum

Material : Tungsten

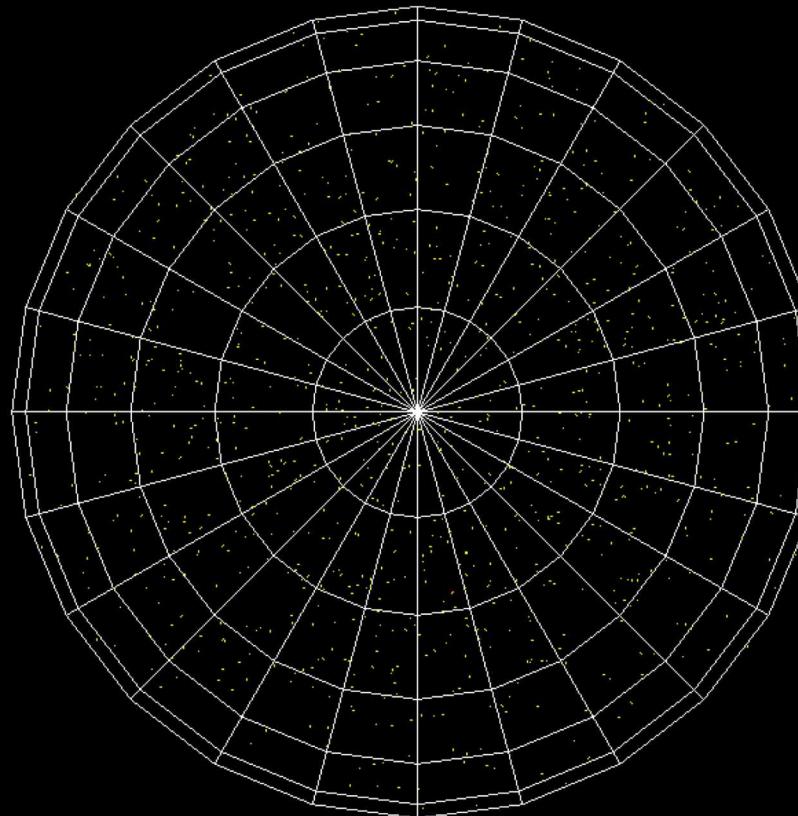
Diameter : 1 μm

Particle : e^-

Energy : 1 keV

e^- total : 1000

e^- out : 0



Wireframe visualisation from Geant4

Single sphere in vacuum

Material : Tungsten

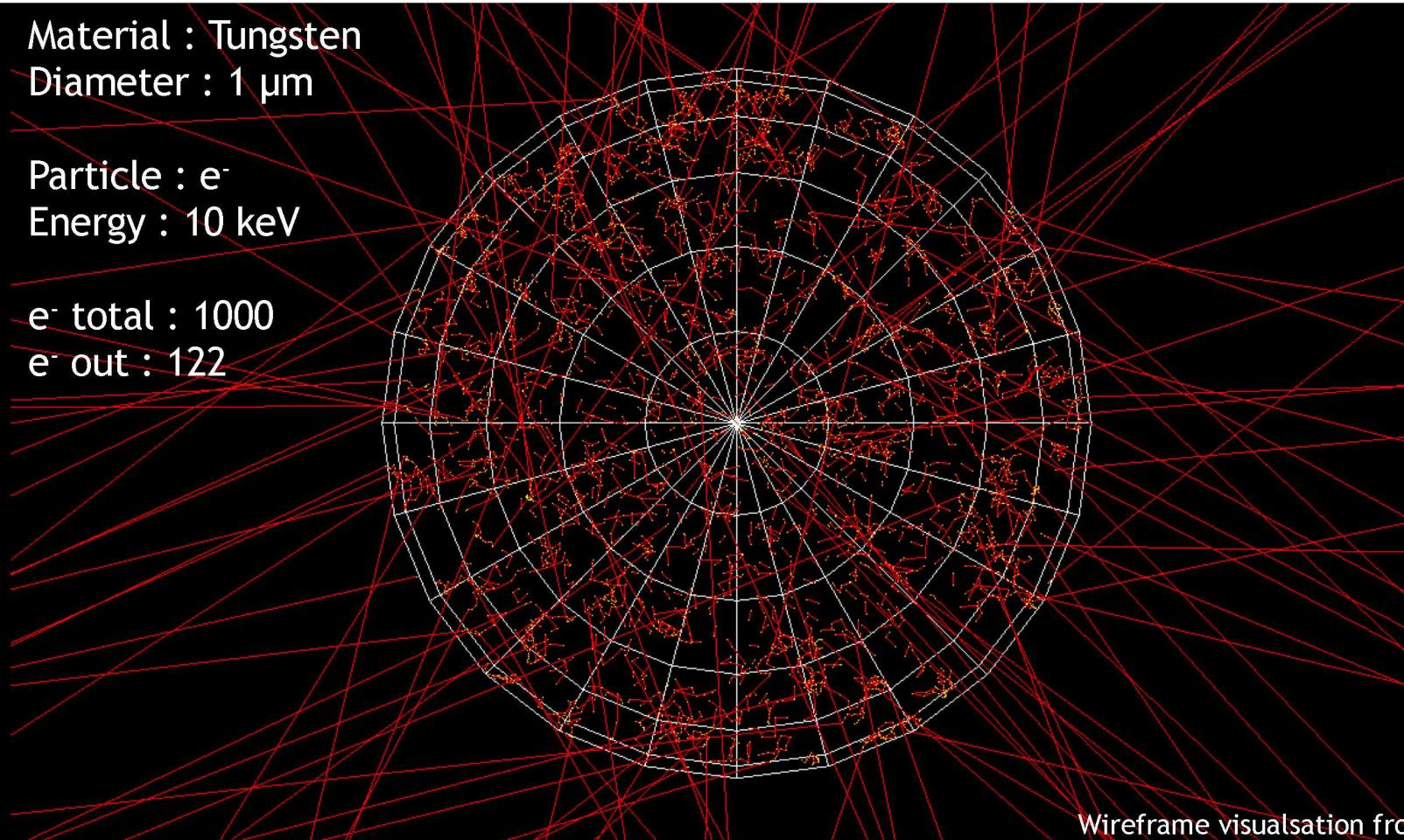
Diameter : 1 μm

Particle : e^-

Energy : 10 keV

e^- total : 1000

e^- out : 122



Wireframe visualisation from Geant4

Single sphere in vacuum

Material : Tungsten

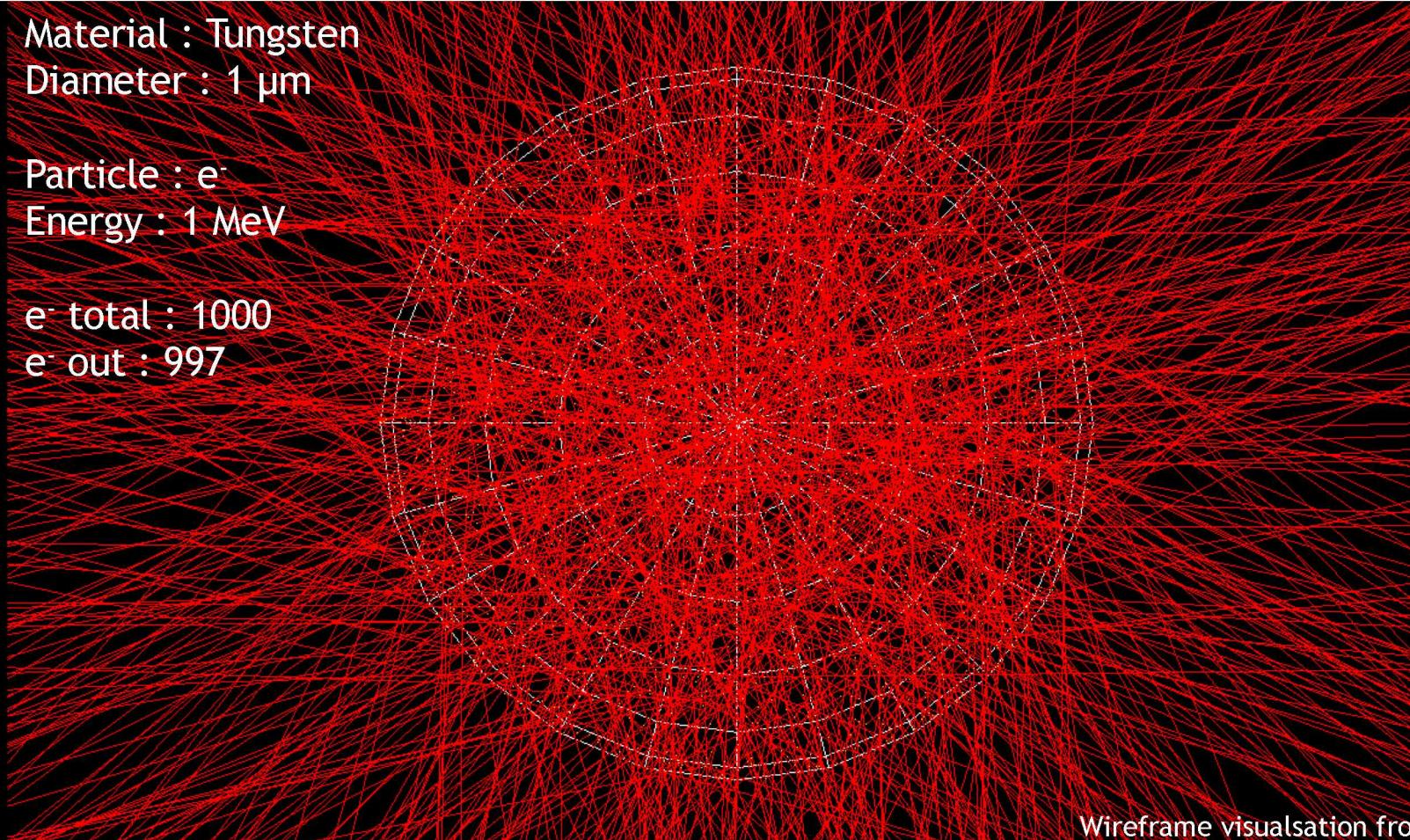
Diameter : 1 μm

Particle : e^-

Energy : 1 MeV

e^- total : 1000

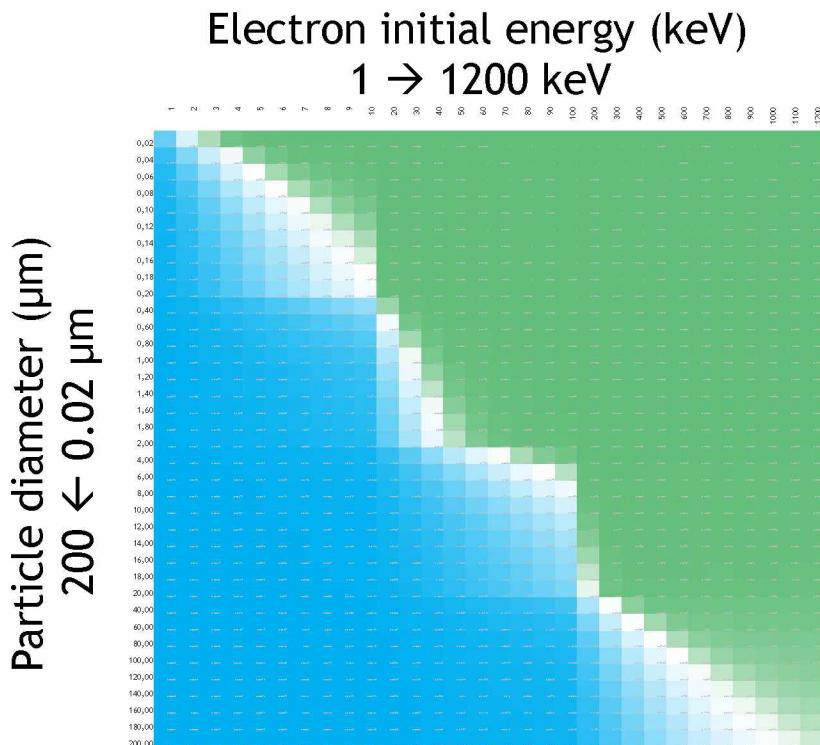
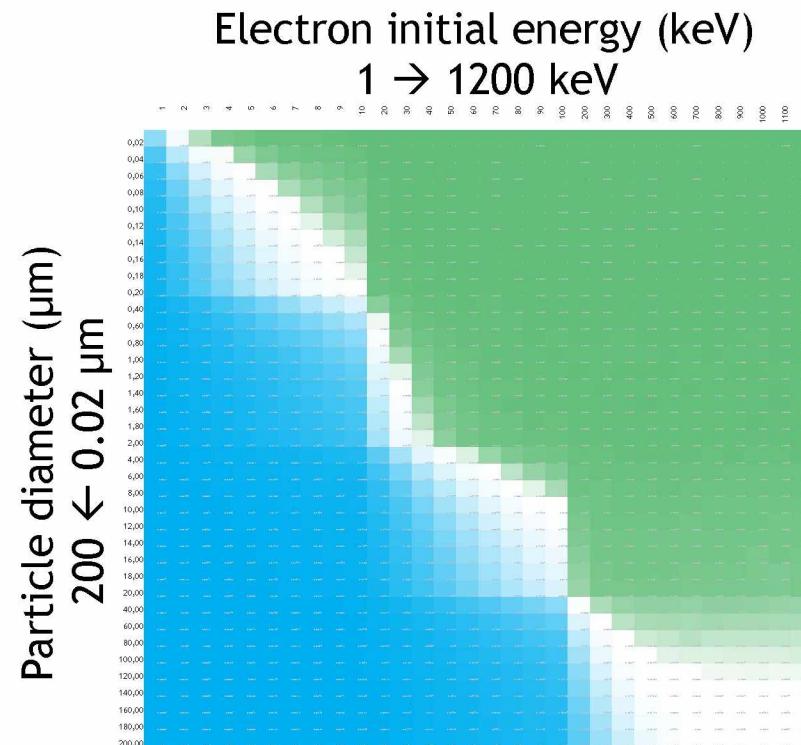
e^- out : 997



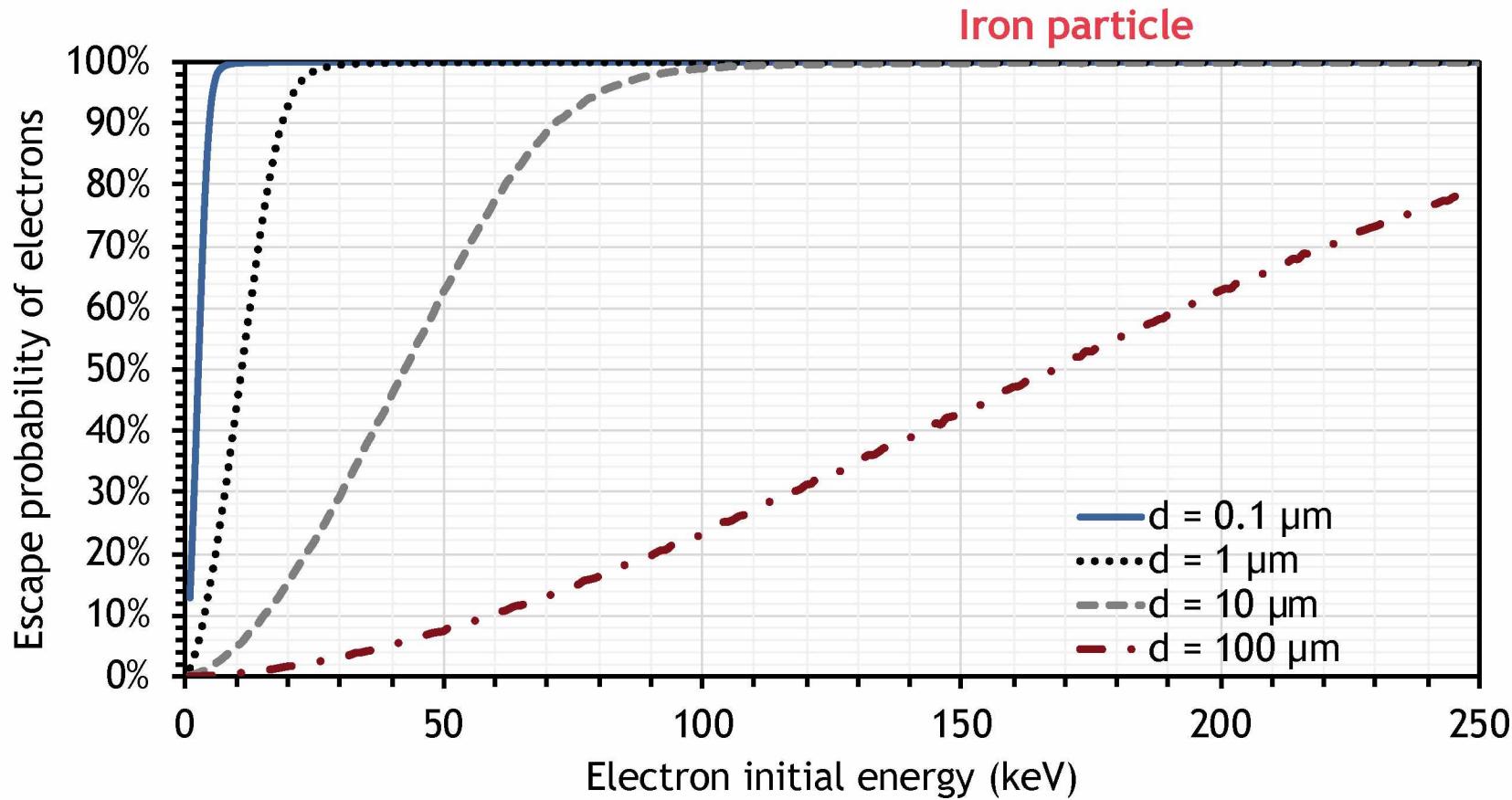
Wireframe visualisation from Geant4

Escape probability of electrons

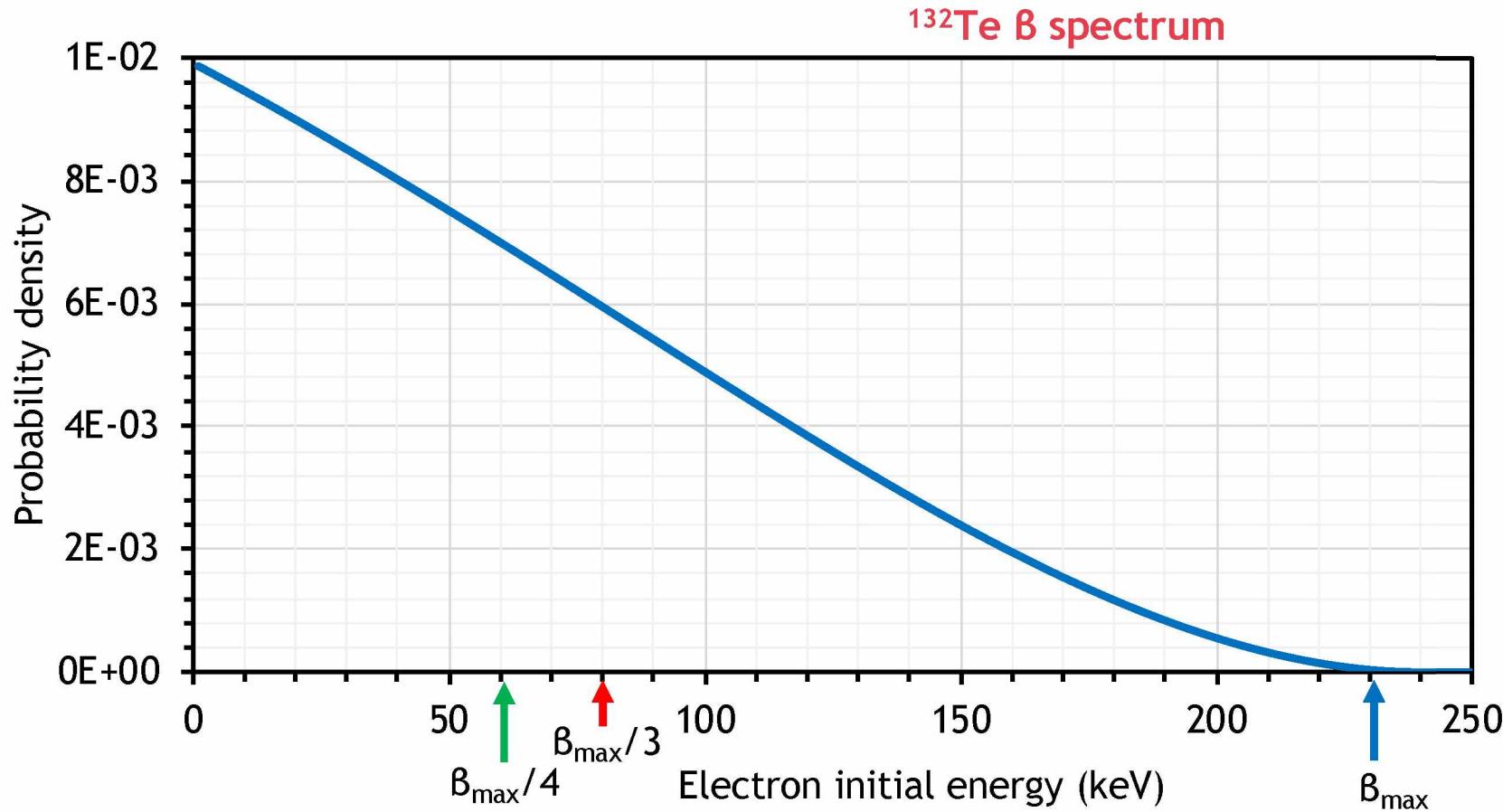
(~log scale)

Tungsten**Iron**Electron escape
probability (%)

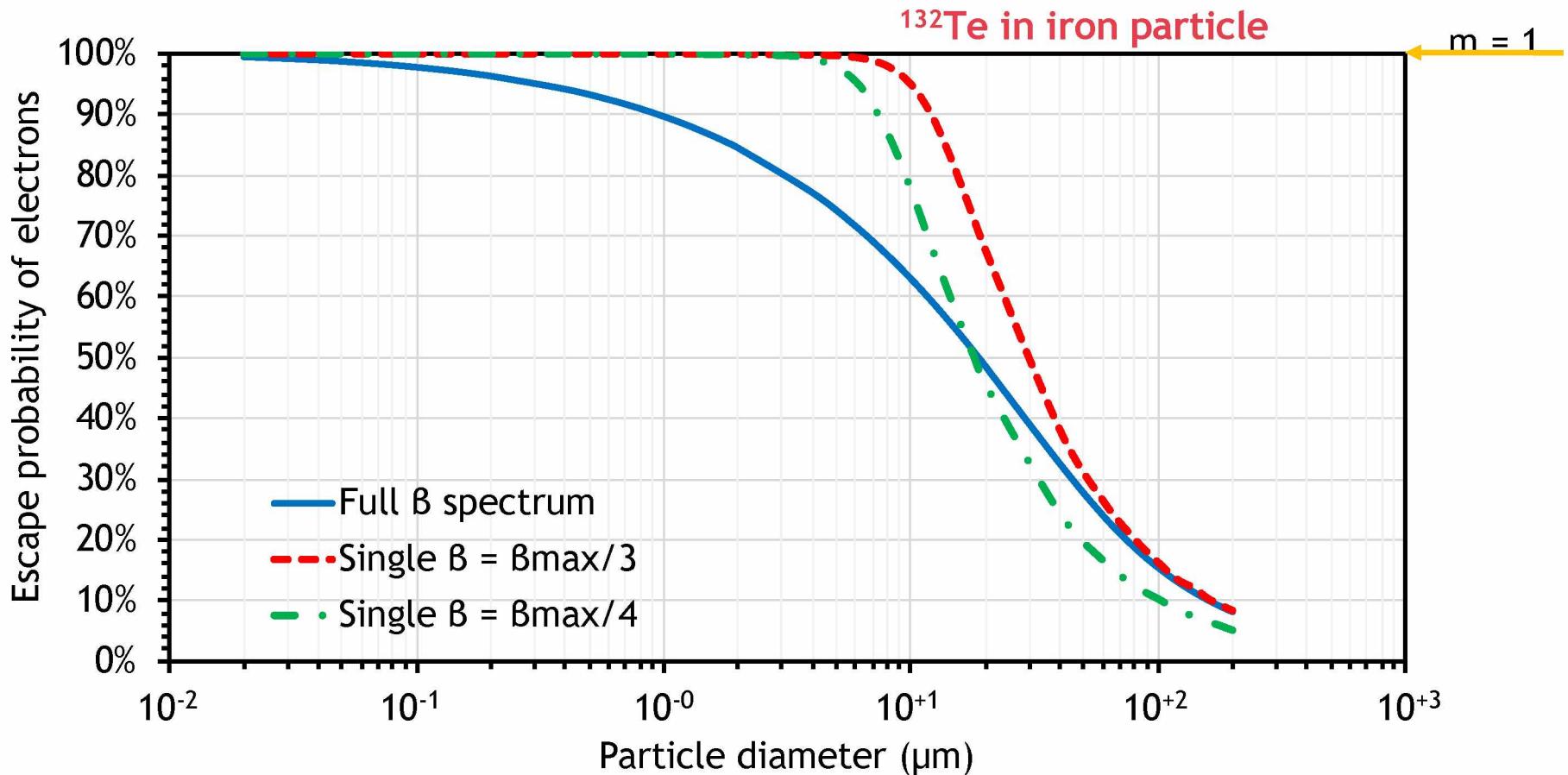
Escape probability of electrons



Electrons energies - Three cases



Escape probability of electrons



Escape probability of electrons

^{132}Te in iron particle

Quantitative assessment of the gap for self charging rate calculation according to the different approaches

Case	Particle Diameter μm	β energy (^{132}Te)	Activity Bq (730 MBq/g)	Charging rate s^{-1}	Case source
1		$\beta_{\max}/3$		3.0	Kim et al.
2	1	$\beta_{\max}/4$	3 Bq	3.0	Gendsarmes et al.
3		Full β spectrum		2.7	This study
1		$\beta_{\max}/3$		2 857	Kim et al.
2	10	$\beta_{\max}/4$	3 kBq	2 336	Gendsarmes et al.
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Escape probability of electrons

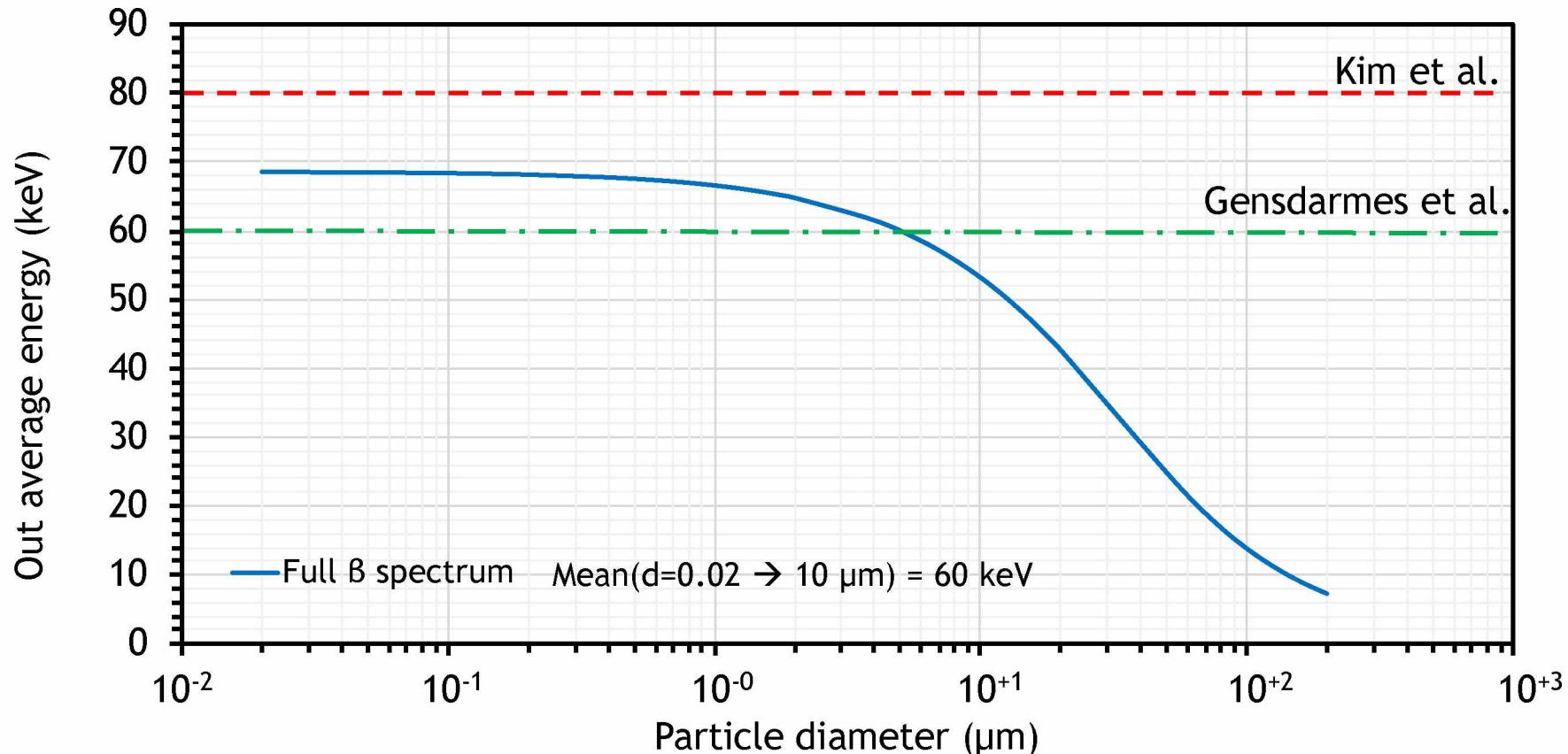
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Escaping e⁻ average energy

¹³²Te in iron particle



How to validate model & simulation ?

I Geant4 toolkit is many times validated

- From nano scale to huge scale (CMS)
- For e, α , γ , π , H, ...

I This model has to be improved to mimic real situations

- Secondary electrons from ionisation
- Electrostatic field in particle
- Infusion depth of nuclides in particle
- Particle in open air and not vaccum
 - diffusion-charging mechanism;
 - ion-particle attachment;
 - ion-ion recombination...

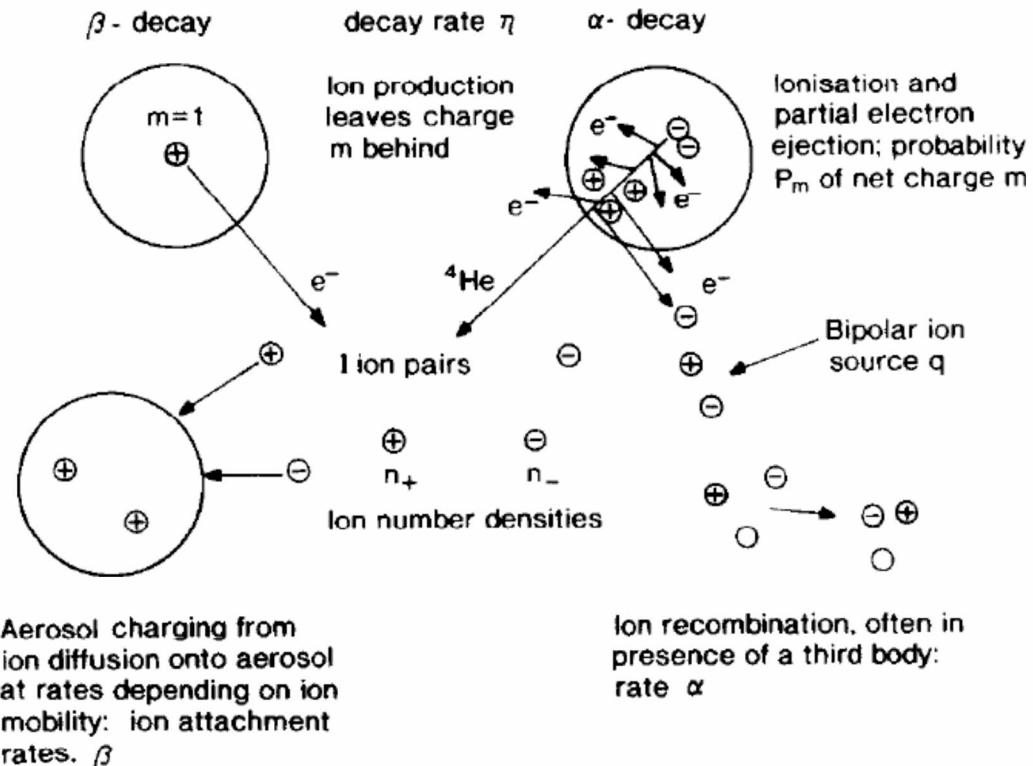


Fig. 1. Processes leading to the charging of radioactive aerosols.

Clement and Harisson, JAS 1992

To conclude

- Single particles in vacuum, uniformly filled with nuclides
 - Geant4, aka numerical simulations, is a great tool to calculate the escape probabilities of electrons
 - Significant impact on β distribution choice
 - single energy or full spectrum
 - $m \leq 1$
residual charge left after 1 decay
 - $m = f(\text{nuclide, particle diameter...})$
- Incorporate more phenomena to become experimentally validable
1. Nuclides infusion in particle
 2. Secondary electrons
 3. Electrostatic field
 4. Particle in air
& energy deposition for ionisation...

Tack för din uppmärksamhet

Thank you for your attention

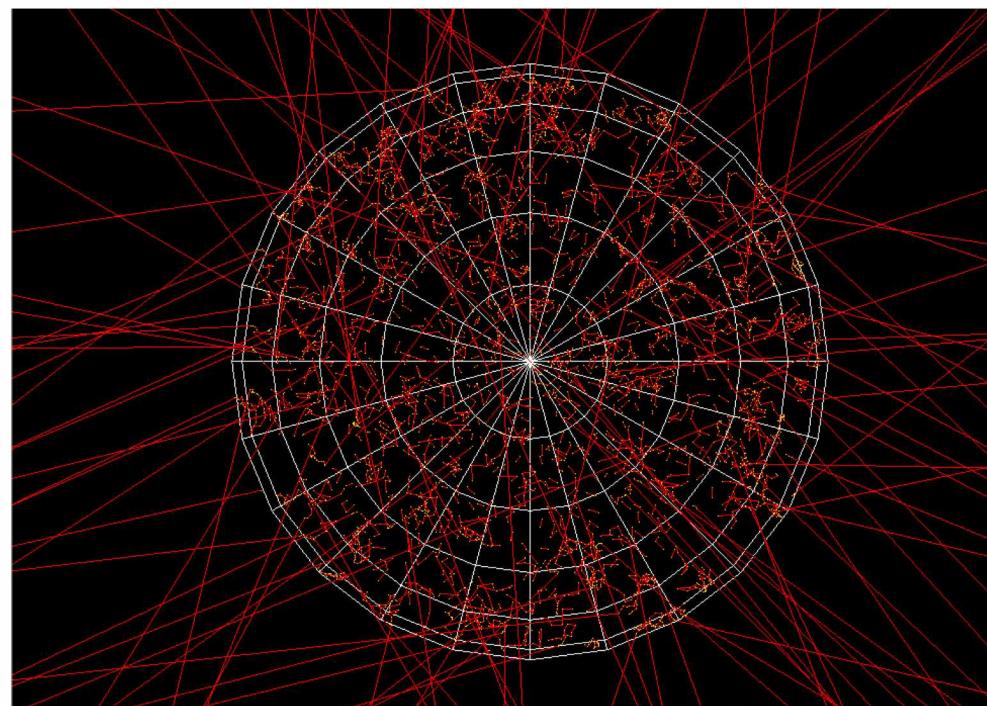
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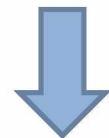


Radioactive aerosol self-charging



$d_{50} = 1 \mu\text{m}$

$C = 1000 \#/\text{cm}^3$



$\sim 10^{10}$ molecules/particle

7 days in atmosphere



Natural
 ^{210}Po

$A = 100 \text{ mBq}/\text{m}^3$ †

$T_{1/2} = 22.23 \text{ years}$



Anthropogenic
 ^{137}Cs

$A = 10 \text{ Bq}/\text{m}^3$ †

$T_{1/2} = 30.05 \text{ years}$



$0.1 \text{ atom/particle}$

$6 \cdot 10^{-5} \text{ e}^-/\text{particle}$

14 atom/particle

$6 \cdot 10^{-3} \text{ e}^-/\text{particle}$

Aerosols undergo a light electrical self-charging due the radioactivity they carry.

† IRSN (2009) Surveillance radiologique de l'environnement : la surveillance des aérosols

Radioactive aerosol self-charging



$d_{50} = 1 \mu\text{m}$

$C = 1000 \#/\text{cm}^3$



$\sim 10^{10}$ molecules/particle

7 days in atmosphere



6.10^7 atom/particle



$T_{1/2} = 12.6 \text{ years}$



ICARE
 ^{137}Cs

$A = 100 \text{ Bq/m}^3$

$T_{1/2} = 30.05 \text{ years}$



140 atom/particle

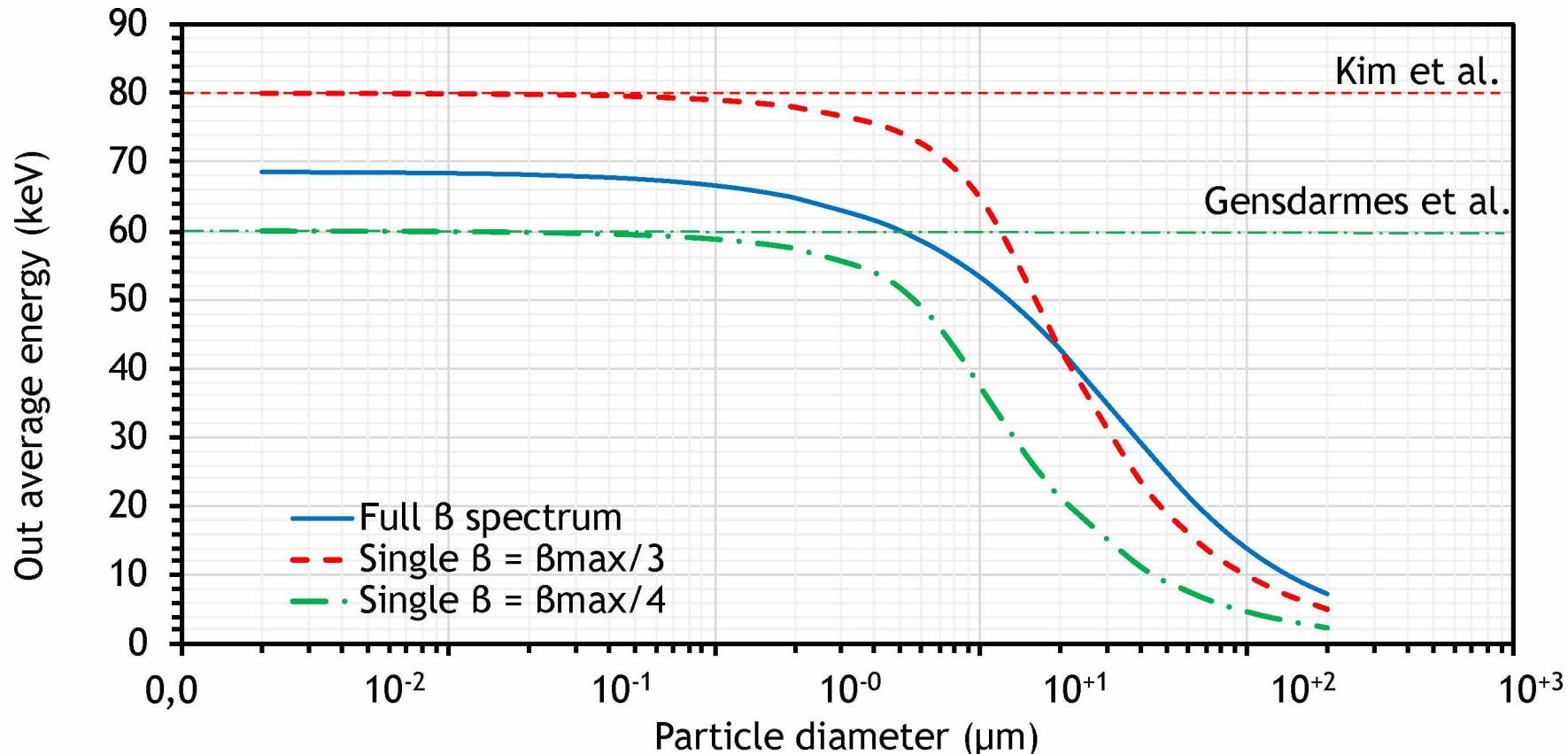
$6.10^{-2} \text{ e}^-/\text{particle}$

Aerosols undergo a significant electrical self-charging due the radioactivity they carry in nuclear specific facility.

[†] El-Kharbachi, A. et al., 2014, [10.1016/j.ijhydene.2014.05.023](https://doi.org/10.1016/j.ijhydene.2014.05.023)

Escaping e⁻ average energy

¹³²Te in iron particle



Geant4 physical model impact

Physical models: Electromagnetic standard 4, Penelope & Livermore
 ^{198}Au in iron particle

