Influence of the $\beta$ 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 $\beta$ decay

→ Reasonable assumption for classical radionuclides considered

- $^{198}$Au (Yeh et al. 1976)
- $^{137}$Cs (Gensdarmes et al. 2001)

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

Clement and Harisson, JAS 1992
What is self-charging of radioactive aerosol?

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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

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Mean β energy (keV)</th>
<th>Path length in SiO₂* (μm) with mean β</th>
<th>Path length in SiO₂* (μm) with full β spectrum</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{137}_{55}$Cs</td>
<td>171</td>
<td>185</td>
<td>56</td>
</tr>
<tr>
<td>$^{132}_{52}$Te</td>
<td>80</td>
<td>52</td>
<td>26</td>
</tr>
<tr>
<td>$^{131}_{54}$I</td>
<td>202</td>
<td>241</td>
<td>54</td>
</tr>
<tr>
<td>$^1$H</td>
<td>6</td>
<td>&lt; 1</td>
<td>&lt; 1</td>
</tr>
</tbody>
</table>

† Ivanov & Kirichenko, 1970; Yeh et al., 1976; Clement & Harisson, 1992, Gensdarmes et al., 2001
‡ Mean β energy = $E(B_{\text{max}})/3$, $E(B_{\text{max}})$ 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 $B$ energies

1. Electron energy: single value = mean($E(B)$) ≈ $E(B_{\text{max}})/3$
   - Kim et al. 2017
   - Without any information, mean is a pretty good assumption.

2. Electron energy: single value = $E(B_{\text{max}})/4$
   - Gensdarmes et al. 2001
   - More representative has it takes into account the higher coefficients of linear energy transfert in matter for low energy electron of the spectrum.

3. This study, electron energy: full $B$ energy spectrum
How to use the full $\beta$ spectrum?

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

### Simulations variables for aerosols case study

<table>
<thead>
<tr>
<th>Variable</th>
<th>Values</th>
</tr>
</thead>
<tbody>
<tr>
<td>Particle material</td>
<td>Tungsten, Iron, Beryllium</td>
</tr>
<tr>
<td>Particle density</td>
<td>19.3, 7.87, 1.85 g/cm³</td>
</tr>
<tr>
<td>Particle diameter</td>
<td>20 nm → 200 µm</td>
</tr>
<tr>
<td>Electron energy</td>
<td>1 → 1200 keV</td>
</tr>
<tr>
<td>β spectrum†</td>
<td>$^3$H, $^{60}$Co, $^{137}$Cs, $^{132}$Te</td>
</tr>
</tbody>
</table>

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
Single sphere in vacuum

Material: Tungsten
Diameter: 1 \(\mu\)m

Particle: e\(^-\)
Energy: 10 keV

e\(^-\) total: 1000
e\(^-\) out: 122
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

Tungsten
Electron initial energy (keV)
1 → 1200 keV

Iron
Electron initial energy (keV)
1 → 1200 keV

Electron escape probability (%)
Escape probability of electrons

Iron particle

Electron initial energy (keV)

Escape probability of electrons

- $d = 0.1 \, \mu m$
- $d = 1 \, \mu m$
- $d = 10 \, \mu m$
- $d = 100 \, \mu m$
Electrons energies - Three cases

\[ \text{Probability density} \]

\[ \begin{align*}
\text{Electron initial energy (keV)} &
\end{align*} \]

\[ \begin{align*}
0 & \quad 2 \quad 4 \quad 6 \quad 8 \quad 10 \quad 12 \quad 14 \quad 16 \quad 18 \quad 20 \quad 22 \quad 24 \quad 26 \\
\text{Probability density} & \quad 10^{-2} \quad 10^{-3} \quad 10^{-4} \quad 10^{-5} \quad 10^{-6} \\
\end{align*} \]

\[ \begin{align*}
\beta_{\text{max}}/4 & \quad \beta_{\text{max}}/3 & \quad \beta_{\text{max}} \\
\end{align*} \]

\[ \begin{align*}
0 & \quad 50 & \quad 100 & \quad 150 & \quad 200 & \quad 250 \\
\end{align*} \]

\[ {^{132}\text{Te}} \beta \text{ spectrum} \]
Escape probability of electrons

\[ \text{Escape probability of electrons} \]

\[ \text{Particle diameter (µm)} \]

\[ 10^2 \rightarrow 10^3 \]

\[ \text{Full } \beta \text{ spectrum} \]

\[ \text{Single } \beta = \beta_{\text{max}}/3 \]

\[ \text{Single } \beta = \beta_{\text{max}}/4 \]

\[ \text{132} \text{Te in iron particle} \]

m = 1
## Escape probability of electrons

### $^{132}\text{Te}$ in iron particle

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

<table>
<thead>
<tr>
<th>Case</th>
<th>Particle Diameter $\mu$m</th>
<th>$\beta$ energy ($^{132}\text{Te}$)</th>
<th>Activity Bq ($730$ MBq/g)</th>
<th>Charging rate $s^{-1}$</th>
<th>Case source</th>
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<tr>
<td>1</td>
<td></td>
<td>$\beta_{\text{max}}/3$</td>
<td></td>
<td>3.0</td>
<td>Kim et al.</td>
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<td>2</td>
<td>1</td>
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<td>Gensdarmes et al.</td>
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<tr>
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<td></td>
<td>Full $\beta$ spectrum</td>
<td></td>
<td>2.7</td>
<td>This study</td>
</tr>
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<td></td>
<td>2857</td>
<td>Kim et al.</td>
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<tr>
<td>2</td>
<td>10</td>
<td>$\beta_{\text{max}}/4$</td>
<td>3 kBq</td>
<td>2336</td>
<td>Gensdarmes et al.</td>
</tr>
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<td>3</td>
<td></td>
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<td>1994</td>
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## Escape probability of electrons

**$^{132}$Te in iron particle**

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<td>2 336</td>
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<tr>
<td>3</td>
<td>Full $\beta$ spectrum</td>
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<td></td>
<td>1 994</td>
<td>This study</td>
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Escaping $e^-$ average energy

$^{132}$Te in iron particle

Out average energy (keV)

Particle diameter (μm)

Full $β$ spectrum

Mean ($d=0.02 \rightarrow 10 \text{ μm}$) = 60 keV

Kim et al.

Gensdarmes et al.
How to validate model & simulation?

**Geant4 toolkit is many times validated**
- From nano scale to huge scale (CMS)
- For e, α, γ, π, H, ...

**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 vacuum
  - diffusion-charging mechanism;
  - ion-particle attachment;
  - ion-ion recombination...

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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 B 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

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

\[ d_{50} = 1 \, \mu m \]
\[ C = 1000 \, \# /cm^3 \]
\[ \sim 10^{10} \text{ molecules/particle} \]
\[ 7 \text{ days in atmosphere} \]

Natural
\[ ^{210}Po \]
\[ A = 100 \, \text{mBq/m}^3 \]
\[ T_{1/2} = 22.23 \, \text{years} \]
\[ 0.1 \text{ atom/particle} \]
\[ 6.10^{-5} \, e^-/\text{particle} \]

Anthropogenic
\[ ^{137}Cs \]
\[ A = 10 \, \text{Bq/m}^3 \]
\[ T_{1/2} = 30.05 \, \text{years} \]
\[ 14 \text{ atom/particle} \]
\[ 6.10^{-3} \, e^-/\text{particle} \]

Radioactive aerosol self-charging

**Radioactive Aerosol Self-Charging**

**Aerosol**

- diameter $d_{50} = 1 \mu m$
- concentration $C = 1000 \#/cm^3$
- ~$10^{10}$ molecules/particle
- 7 days in atmosphere

**Tokamak**

- $^{3}H$ nucleus
- activity $A = 10 \text{ GBq/g}$
- $T_{1/2} = 12.6$ years
- 6.10⁷ atom/particle
- 6.10⁴ e⁻/particle

**ICARE**

- $^{137}Cs$
- activity $A = 100 \text{ Bq/m^3}$
- $T_{1/2} = 30.05$ years
- 140 atom/particle
- 6.10⁻² e⁻/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
Escaping $e^-$ average energy

$^{132}$Te in iron particle

- Kim et al.
- Gensdarmes et al.

Graph showing the relationship between particle diameter (μm) and out average energy (keV) for different spectrum models.
Geant4 physical model impact

Physical models: Electromagnetic standard 4, Penelope & Livermore

$^{198}$Au in iron particle

![Graph showing escape probability of electrons as a function of particle diameter (µm) for different materials and physical models.](image-url)