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Title:	Validation of the single-event method and EPRDATA14 library for low-energy electron transport via stopping power calculations
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Validation of the single-event method and EPRDATA14 library for low-energy electron transport via stopping power calculations

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Executive summary

- The single-event method for electron transport is *complementary* to the traditional condensed history method, used for *low-energy* electron transport.
  - The SE method relies on the EPRDATA14 library (ACE conversion of EPICS2014).

- *General* validation of SE method at *low energies* (<100 keV) is needed.

- Performed this validation by computing electron stopping powers for:
  - 41 elemental solids, including 3 carbon allotropes
  - 14 compound solids: alloys, semiconductors, insulators, …
  - 5 rare gas solids

- Good agreement with experiment for most energies (to 1 keV or lower).

- Worse agreement for lower energies, particularly for compounds.
  - Much of this is due to limitations of the EPRDATA14 library for solid-state interactions.
  - Several limitations in our calculation procedure and assumptions.

- Verified MCNP implementation of SE method within these limits.

Introduction: Why single-event transport?

- Traditionally, electron transport is done by the condensed history method.
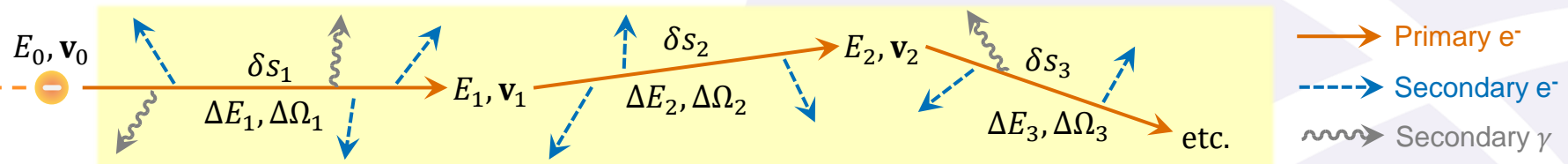
- This is accurate for most electron energies (e.g., β particles), but breaks down in the range from 1 to 10 keV, fails below 1 keV.
  - CH component theories reach limits here – Bethe stopping, multiple scattering, etc.
  - In MCNP, the CH method will not run below 1 keV.

- From MCNP 6.1 we have the optional single-event method to supplement the CH method at lower energies down to 10 eV.
  - <u>SE method does not replace CH method!!</u> It is complementary.
    - SE method is computationally very expensive at high energies – prefer CH for this regime.
  - Not enabled by default (default CH-to-SE switch and electron cutoff are both 1 keV).

Single-event electron transport
## (adapted from H.G. Hughes, LA-UR-12-23333)

- Start with cross sections, $\sigma_{i,j}$: $i :=$ material index, $j :=$ process label.
  - Processes: Elastic, atomic excitation, electro-ionization, bremsstrahlung

- Get total cross section and distance to collision:
  - $\Sigma_i = N_i\big(\sigma_{i,\mathrm{elast}} + \sigma_{i,\mathrm{excit}} + \sigma_{i,\mathrm{ioniz}} + \sigma_{i,\mathrm{brems}}\big)$
  - $D_\mathrm{c} = -\ln(\mathrm{rang})/(\Sigma_1 + \Sigma_2 + \cdots + \Sigma_\mathrm{m})$ for material with m components.

- Also get distance to cell interface, etc.
  - If distance to interface is less than distance to collision, transport to interface.
  - Otherwise transport to collision and select a process.

- If collision, select a process:
  - Select target atom (material):
    - If m = 1 this is trivial.
    - Else, choose randomly using $\{\Sigma_i\}$ as weights.
  - Select process:
    - Choose randomly using $\{\sigma_{i,j}\}$ as weights.

Material 1

D_c

D_i D_c D_i

Material 2

Select D_i   **Select D_c**
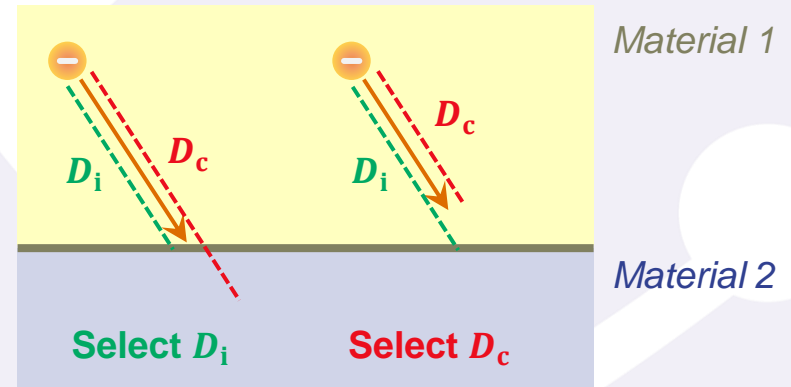
Collision processes

- Elastic scattering:
  - No energy loss.
  - No secondary particles.
  - Sample deflection μ from tabulated distribution $f_{\text{elast}}(E, \mu)$
  - EPRDATA14 enhanced forward elastic scattering vs. earlier versions.

- Atomic excitation:
  - No angular deflection.
  - No secondary particles.
  - Energy loss is unique, single-valued function of energy $f_{\text{excit}}(E)$ - no sampling!
  - EPRDATA14 evaluation only considers outer subshell interactions.
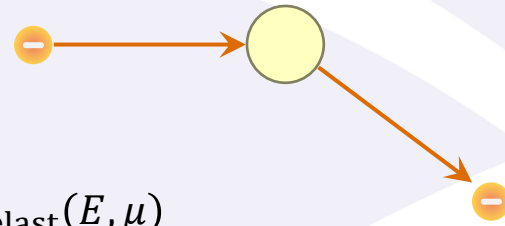
Collision processes

- Electro-ionization:
  - Sample cross sections for individual subshells.
  - Sample knock-on energy E_k from tabulated distribution $f_{\text{knock}}(E, E_k)$
    - Generate knock-on if $E_k > E_{\text{cut}}$
  - Reduce incident energy by $\Delta E = E_k + E_{b,s}$ where $E_{b,s}$ is subshell binding energy.
  - Get incident and knock-on directions from conservation of energy and momentum.
  - Fill subshell vacancy using atomic relaxation data.
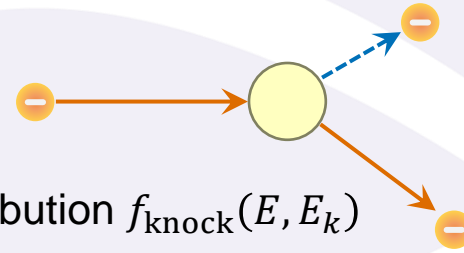
- Bremsstrahlung:
  - No angular deflection.
  - Sample photon energy from tabulated distribution $f_{\text{brems}}(E, E_\gamma)$
    - Generate photon if $E_\gamma > E_{\text{cut},\gamma}$
  - Reduce incident energy by E_γ
  - Sample photon direction from tabular distribution ($1\ \text{keV} \leq E_\gamma \leq 1\ \text{GeV}$) or from simple analytic distribution $P(\mu) = 0.5(1 - \beta^2)/(1 - \beta\mu)^2$ (other E_γ).
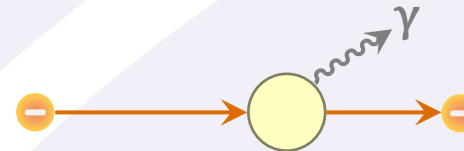
Validating the single-event method

- It is nice to have an algorithm that works.

- It is even nicer to know how well that algorithm works.

- MCNP team has validated SE electron transport above 50 keV against the Lockwood energy deposition experiments.
  - D.A. Dixon: LA-UR-21-25586 and LA-UR-21-25629.
  - No known *internal* validation below 50 keV.

- Broader literature uses SE electron transport extensively, but…
  - …mostly only in water (DNA modeling for radiology applications).
  - Sometimes nanoparticles in water – Au, ferrites, …
  - Studies on a wide range of general materials are very limited
    - Two papers by A. Poškus (2016) – backscattering and x-ray emission only.

- MCNP users need to know how well single-event electron transport works!
  - Is it accurate?
  - Uncertainty analysis is critical! What simulation conditions affect accuracy?

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Computing electron stopping power with MCNP

- Stopping power, $S(E) := \frac{dE}{dx}$, is the fundamental energy transport quantity.
  - In other words, we compute $S(E)$ because it says a lot about how well our transport works for many applications—not because $S(E)$ itself is an important application.
- Two ways to compute $S(E)$:
  - Directly from MCNP simulations – see next slides.
  - Integrate over the EPRDATA14 differential energy transfer cross sections:

$$S(E) = N \int_0^{T_{max}} T \frac{\partial \sigma(E,T)}{\partial T} dT$$

where $\int_0^{T_m} \frac{\partial \sigma(E,T)}{\partial T} dT = \sigma_{\text{inel}} = \sigma_{\text{excit}} + \sigma_{\text{ioniz}} + \sigma_{\text{brems}}$

- We want to use both methods:
  - <u>Verify</u> single-event method by comparing simulated and integrated stopping powers.
  - <u>Validate</u> single-event method by comparing both to experimental data.

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Computing stopping power from single-event electron transport simulations

- Based on <u>continuous slowing down approximation (CSDA)</u> and calculus:

$$R(E) \cong \int_{E_0}^{E_{\text{cut}}} -\frac{1}{S(E')} dE' \quad \therefore \quad S(E) \cong \left(\frac{dR}{dE}\right)^{-1}$$

- Given set of points $\{E_i, R_i\}$, approximate $R(E)$ as a quadratic near E_i,
  $R(E - E_i) \approx a_i + b_i(E - E_i) + c_i(E - E_i)^2$
  - Fit exactly to the set of points $\{(E_{i-1}, R_{i-1}), (E_i, R_i), (E_{i+1}, R_{i+1})\}$.
  - N.B. this is not a Taylor series… just an approximation!

- Stopping power is then

$$S(E_i) \approx \frac{1}{b_i} = \frac{E_{i+1} - E_{i-1}}{\frac{E_{i+1} - E_i}{E_{i-1} - E_i}(R_{i-1} - R_i) - \frac{E_{i-1} - E_i}{E_{i+1} - E_i}(R_{i+1} - R_i)}$$

- Thus, "all" we need to do is calculate $R(E)$…

Computing electron range from single-event electron transport simulations

- In principle this is simple: simulate an electron history with initial energy E_0 then add up all displacements $\{D_{\Omega,i}\}$ between change-of-direction events.

  - N.B. source-to-terminate displacement is less than range due to multiple scattering.

$$R = D_{\Omega,1} + D_{\Omega,2} + D_{\Omega,3} + D_{\Omega,4}$$

- In practice the trick is to obtain these change-of-direction events.
  - In MCNP we can use the PTRAC output file.
  - However, SE method in MCNP is not fully integrated – collisions are not tracked!
    - This may have been intentional to keep the PTRAC file size manageable.
  - Other events are tracked – source, bank, termination – so we can work around this.
    - Turn off elastic scattering – no energy loss thus no impact on stopping power
      - This is an undocumented option, but it does exist in the source code.
    - Excitation and bremsstrahlung collisions – no change in direction, so not a problem.
    - Ionization – change in direction, but we can locate these from the bank events for knock-ons.
      - Some error arises when $E_k \leq E_{cut}$ and no bank event is recorded. Mainly for very low energies, $E_0 \leq 100$ eV.

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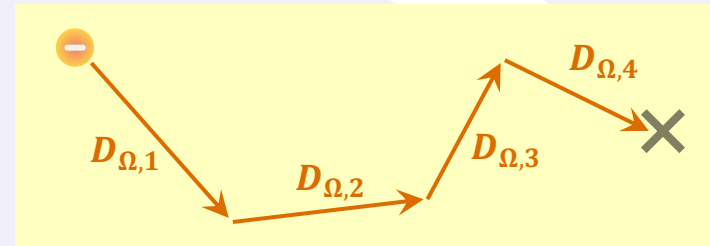
MCNP simulation setup

- Geometry: homogeneous slab, infinite in x and y, $-1 \leq z \leq 1$ cm.
  - Effectively an infinite medium since MCNP doesn't like actual infinite cells.

- Use EPRDATA14 library with EL03 for bremsstrahlung angular distributions.

- Generate electrons at (0,0,0) with energies randomly selected from the range $\{E_{\min}, 50 \text{ eV}, 63 \text{ eV}, \dots, 30 \text{ keV}, 37.5 \text{ keV}\}$ where $E_{\min} > E_{\text{cut}}$, usually 12 eV.
  - Endpoints are used only to fit $R(E)$, no stopping power is calculated for these.
  - Total of 63,488 histories yields ~2,048 per energy point → uncertainty $U[R(E)] \leq 1\%$.

- Simulated 60 materials:
  - 41 elemental solids
  - 14 compounds solids
  - 5 rare gas solids

- Compared with experimental measurements via electron energy loss functions (ELFs) and universal empirical fit
  - A. Jablonski et al (2008).

| Material type	Materials simulated	Expt. data sources
S-block elements	Li, Be, Na, Mg, K, Cs	Shinotsuka et al. (2012)
P-block elements	Al, Si, Ge, In, Sn, Bi	
D-block elements	Sc, Ti, V, Cr, Fe, Co, Ni, Cu, Y, Nb, Mo, Ru, Rh, Pd, Ag, Hf, Ta, W, Re, Os, Ir, Pt, Au	
F-block elements	Gd, Tb, Dy	
Carbon allotropes	Graphite, diamond, glassy carbon	
Compounds	$H_2O(s)$	Luo et al. (1991), LaVerne and Mozumder (1985)
	GaSb, guanine, InSb, MgO, MoS_2	Luo (1994)
	Al_2O_3, SiO_2	Luo (1994); Joy et al. (1996)
	CuAu, GaAs, SiC, ZnS, ZnSe, ZnTe	Joy et al. (1996)
Rare gas solids	He, Ne, Ar, Kr, Xe	LaVerne and Mozumder (1985)

Results and discussion:
Elemental solids, s-block

- These generally show the best agreement for any periodic table block simulated.

- Plateau and rise at extremely low energies in experimental data is an electronic structure phenomenon.
  - Specifically due to highest core subshell contributions.
  - EPRDATA14 atomic excitation neglects this as only valence subshells are considered.

- Divergence between integrated and simulated $S(E)$ at very low energies ($E_0 \leq 300$ eV).
  - Breakdown of CSDA assumption due to large per-collision energy losses.
  - This is a general feature in simulated $S(E)$, so I won't remark on it every time it shows up.
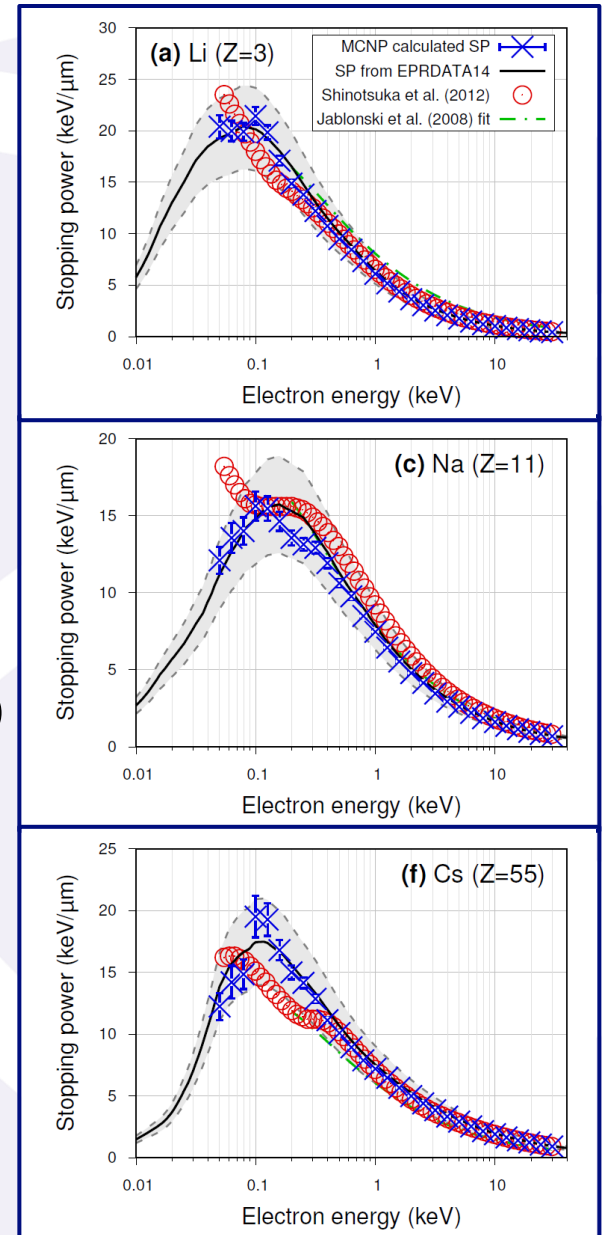
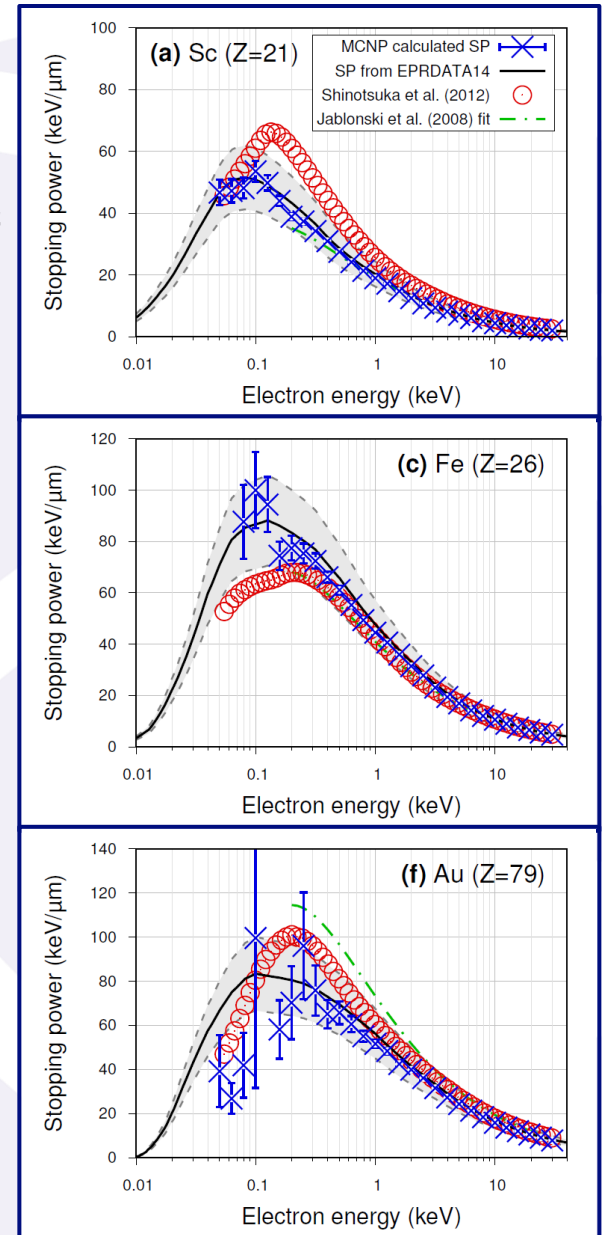# Results and discussion: Elemental solids, p-block

- Things start to get a little more complicated.

- Generally underestimated experimental measurements by -10% for $E \geq 1$ keV.

- In some cases, the same electronic structure effects are visible as for s-block elements.

- Typically, big disagreements between simulations and experiments about the peak of $S(E)$.
  - Peak position/energy differs by factor of ~2.
  - Usually, good agreement about S_{max}.
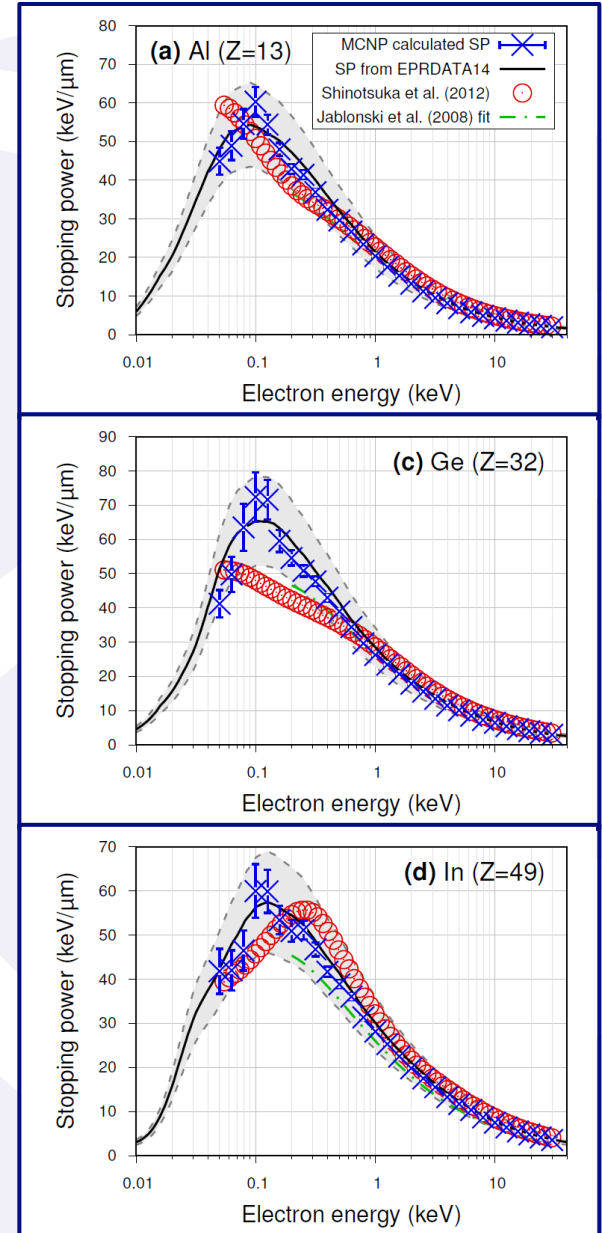
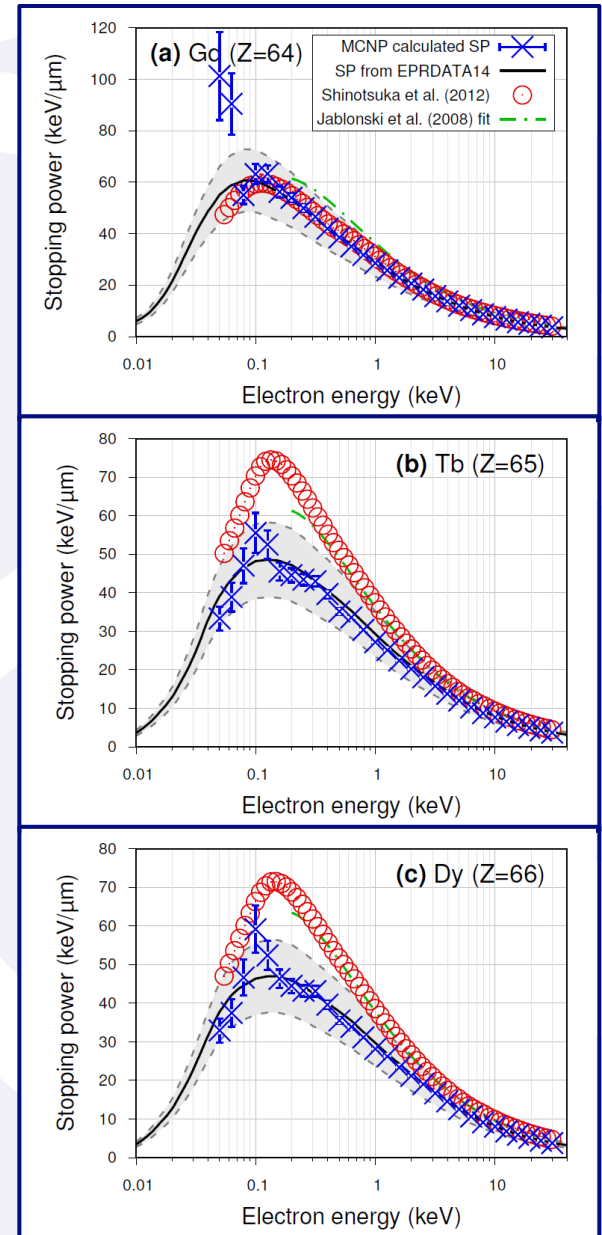# Results and discussion: Elemental solids, d-block

- Simulated 23 transition metals with a wide range of agreement between simulations and experiments.
  - Typically, agreement within ~5-10%.
  - Full range varies from 80% to 110% of experimental.

- Uncertainty at very low energies due to CSDA breakdown is much larger for many transition metals.
  - Transition metals have larger ionization stopping powers.
  - S- and p-block elements have larger atomic excitation stopping powers.
  - Therefore, average energy losses at very low energies are greater for transition metals → larger errors.

- Peak position *and* magnitude disagree significantly with experimental curves.

Results and discussion:
Elemental solids, f-block

- These data are difficult to assess.
  - Great agreement for Gd.
  - Terrible agreement for Tb, Dy, ~15-30% errors.

- Lanthanides have complex electronic structures.
  - 4f, 5d, 5p, 6s subshells all sit very close together.
  - Solid-state bonding modifies electronic structure in several subshells as a result.
  - EPRDATA14 cross sections will not capture this complexity.

- Would like to have more f-block data to assess these conclusions more rigorously!

Results and discussion: Elemental solids, carbon

- The picture is complex here.
  - Graphite: generally good agreement.
  - Diamond: good above 250 eV, huge overestimate for very low energies.
  - Glassy C: good above 1 keV, huge overestimate for lower energies.
- Carbon allotropy leads to diverse electronic structures for solids of the same element.
  - EPRDATA14 does not capture solid-state bonding effects.
  - Other than cross sections, only parameter in MCNP to treat different materials in a low-energy electron transport context is the density.
- But… single-event method does better/more robust than the "universal" empirical fit.
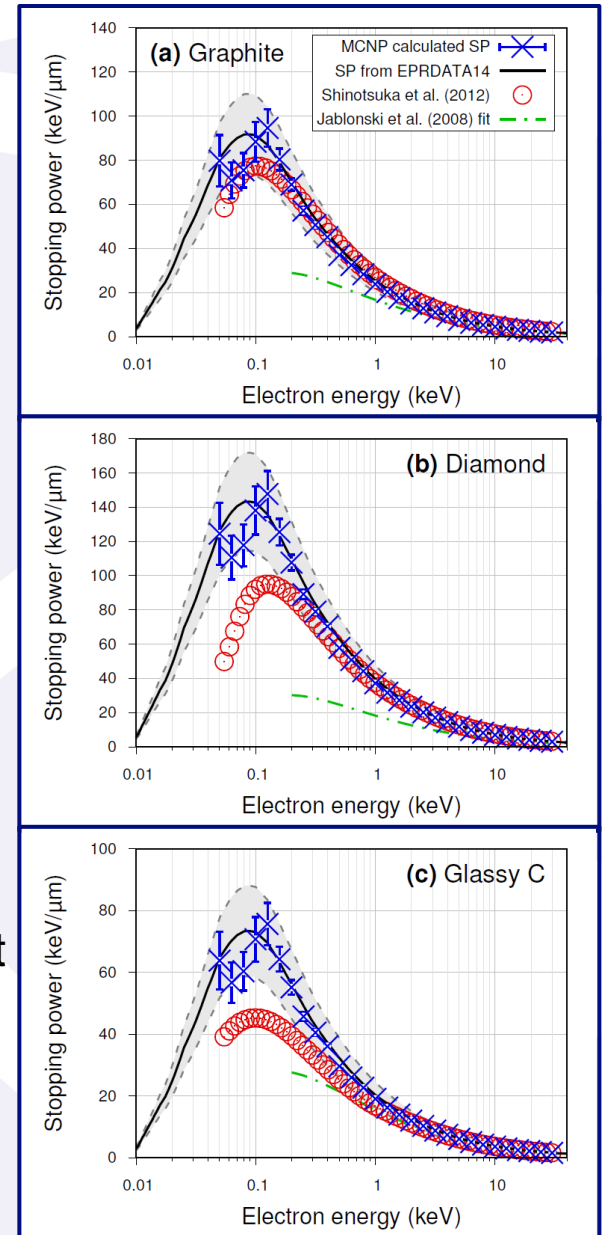
Summary: Elemental solids

- General trends:
  - Good agreement vs. experiment down to energies of about 300 eV.
  - General tendency towards ~5-10% underestimation over this energy range.
  - Simulated stopping powers diverge/large error due to CSDA breakdown at lower energies.
  - Integrated stopping powers disagree markedly for lower energies, especially regarding peak $S(E)$ placement and magnitude.
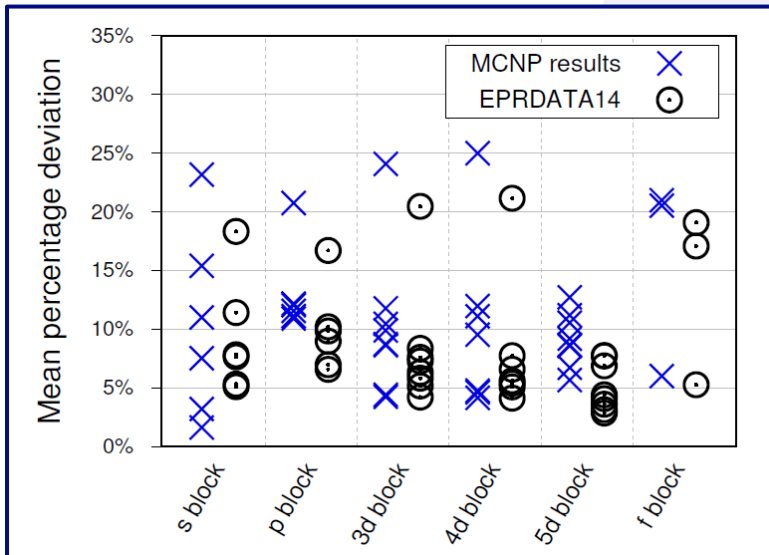
- Key sources of error/uncertainty:
  - Errors in cross section data
    - Quoted uncertainties are ±20%.
    - Data were evaluated on atomic basis; electronic structure of bonding is missing.
    - Simplistic model of atomic excitation.
  - Errors in calculation procedure:
    - Breakdown of CSDA at very low energies.
   - CSDA bias error due to ΔE distributions.

Results and discussion:
Compound solids, alloy

- Good agreement for 200 eV and higher.

- N.B. in MCNP, cross sections for compounds are the density-weighted average of the constituent element cross sections.
  - Good agreement here implies that weighted-average model works well for conductive alloys.
 - However, with only one data set this is not a firm conclusion.
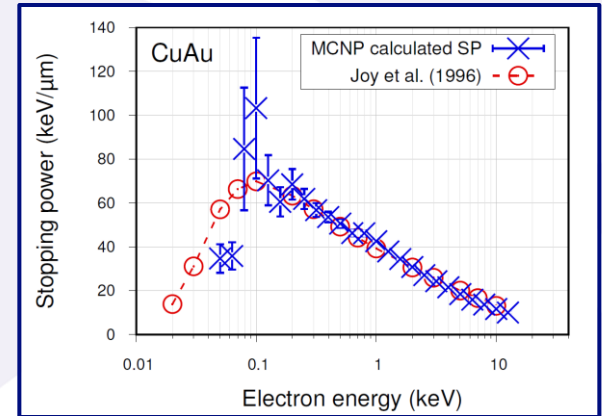
Results and discussion:
Compound solids, semiconductors

- Generally good agreement for 1 keV and above.

- Peak magnitude of $S(E)$ tends to be significantly overestimated by simulations.
  - Experimental curves tend to show broad distributions rather than sharp peaks.
  - Most likely due to complex electronic/band gap structures.

- This highlights the impact of bonding effects on electron transport in compound solids.
  - Weighted average for cross sections does not work as well when bonding is more complicated.
  - Example: SiC – error below 300 eV is 50% to 100% greater than for elemental Si or graphite.
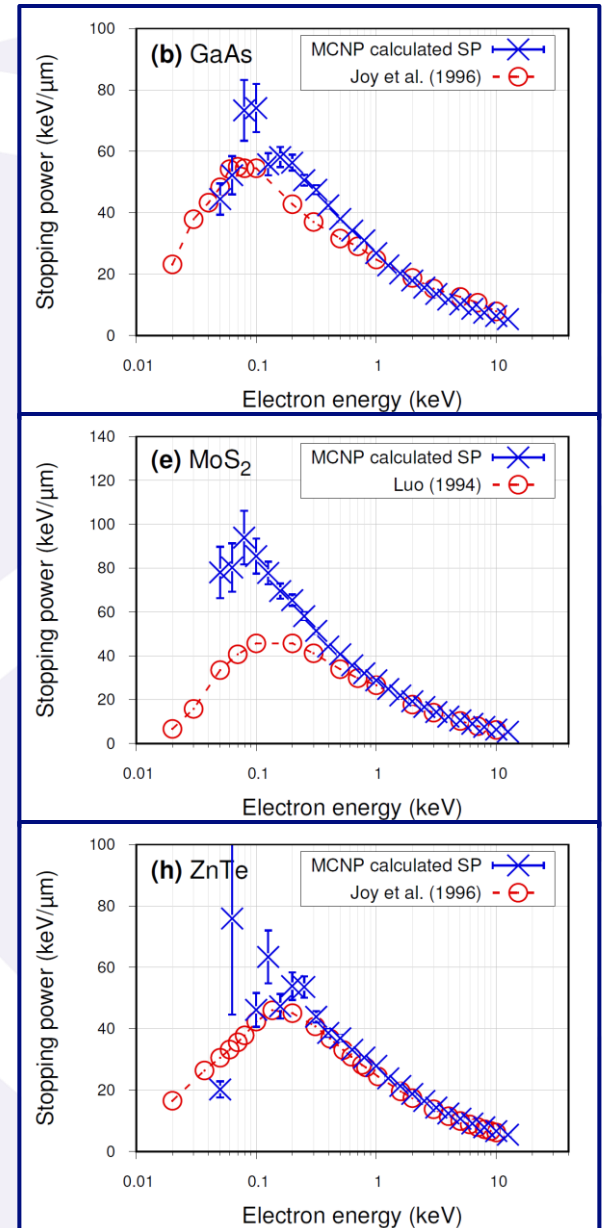
Results and discussion: Compound solids, insulators

- Broadly similar to semiconductor cases:
  - Good agreement above 1 keV, typically down to 500 eV or so.
  - Large overestimation of peak stopping power magnitude at lower energies, around 100 eV.
  - Again, bonding effects are the likely culprit.

- N.B. change in bonding vs. elemental solids should generally reduce the cross section and thus the stopping power.
  - This is because compound bonding results in a more stable configuration than in elemental solids. More stable electrons = harder to knock around.

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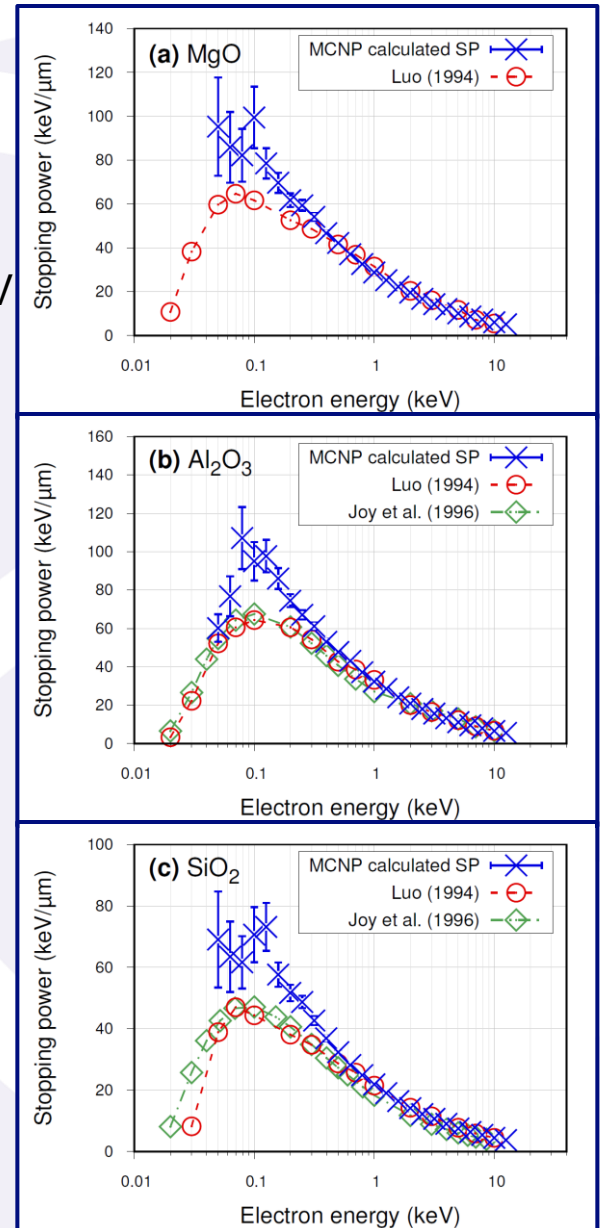
Results and discussion: Compound solids, molecular

- No clear, general trend here.

- Water: generally excellent agreement!
  - This is good news since water is the most commonly simulated material for low-energy electron transport…
  - Unclear why the agreement is so good – high probability that this is fortuitous, not physical.

- Guanine: not so good agreement…
  - Divergence from experiment even above 1 keV.
  - Very large deviations at very low energies.
    - Not shown: $S(E)$ values of 221 keV/um at 63 eV and 391 keV/um at 50 eV – very unphysical!
    - Likely cause: large ionization energies for elemental H, N, O combined with complex molecular structure.
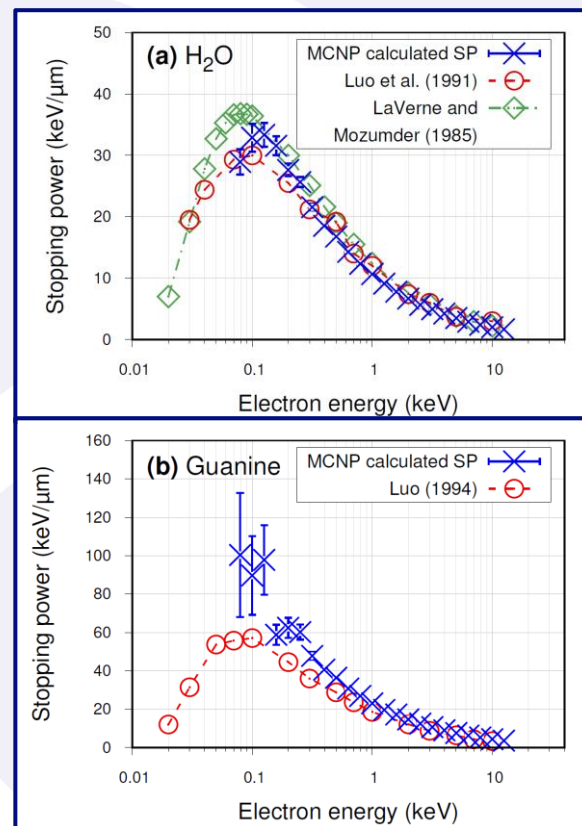
Summary: Compound solids

- Usually, good agreement above 1 keV.

- Below 1 keV there is divergence from experimental measurements, usually in the form of large overestimations.
  - N.B. this can be separated from errors due to CSDA breakdown, which occur below 300 eV so there are different energy domains for each effect.

- We attribute this to changes in electronic structure due to bonding in the real systems, which are not modeled by EPRDATA14 cross sections.
  - More so than the difference between atomic physics to elemental solids, previously.
  - We would like to have more data to rigorously characterize these effects related to the type of bonding, e.g., for conducting alloys this may not be a problem.

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Results and discussion:
Rare gas solids

- Rare gas solids are an interesting case physically:
  - Almost no change in electronic structure due to dispersion force "bonding" only.
  - Very large ionization energies.

- Generally good agreement down to 400 eV.

- Overestimate experimental measurements at lower energies (see integrated $S(E)$ - black lines).
  - No significant bonding effects – must be due to uncertainty or inaccuracy of the cross sections!

- CSDA-related errors are also quite extreme (see simulated $S(E)$ - blue marks).
  - In some cases negative stopping powers result!
  - Likely due to very large ionization losses as well as discretization effects near the cutoff energy.
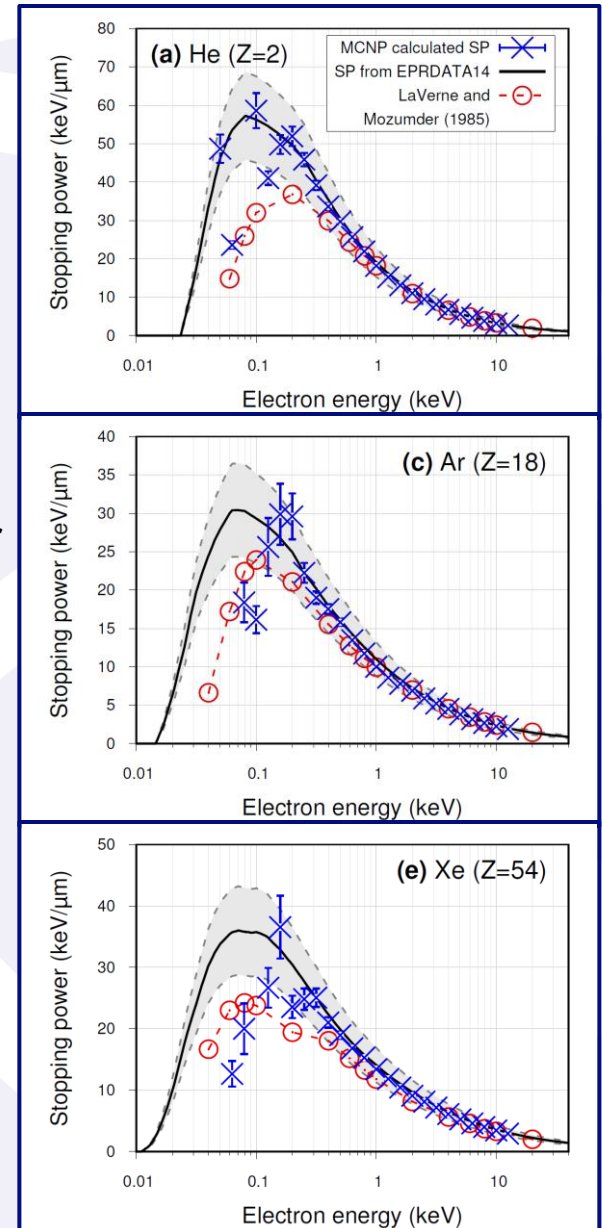
Discussion: limitations of the calculation procedure

- Our calculation procedure relies on the CSDA assumption, which imposes several limits:

    - Breakdown at low energies (below 300 eV) leads to large uncertainties, particularly for materials with large ionization losses.

    - Additionally, the CSDA implicitly assumes uniform energy loss $\Delta E = f(E_0)$. Statistical analysis shows that this leads to underestimating the stopping powers by 2-5% in simulations compared to the integrated stopping powers.

- Importantly, within these limits we find that MCNP reproduces the integrated $S(E)$ curves correctly.

- Therefore, this work **verifies that single-event electron transport in MCNP functions correctly** given the expectations set by the cross section library.

Conclusions

- Validated single-event electron transport at low energies (50 eV to 30 keV) by calculating stopping powers from simulations and cross section integration.

- Generally good agreement for most energies:
  - Elemental solids: above 300 eV
  - Compound solids: above 1 keV
  - Rare gas solids: above 400 eV

- Agreement is weaker for lower energies, principally due to data limitations:
  - Atomic physics evaluations for cross sections do not include solid state effects of bonding on electronic structure.
  - Simplistic model of atomic excitation may underestimate stopping contribution.

- Verified the single-event method within the limits of our calculation procedure.
  - MCNP works… not really a surprise.