



LAWRENCE
LIVERMORE
NATIONAL
LABORATORY

Electron Emission From Slightly Oxidized Delta-stabilized Plutonium Generated by its Radioactivity, and Radiation Induced Ionization and Dissociation of Hydrogen at its Surface

W. J. Siekhaus, A. J. Nelson

October 28, 2011

Disclaimer

This document was prepared as an account of work sponsored by an agency of the United States government. Neither the United States government nor Lawrence Livermore National Security, LLC, nor any of their employees makes any warranty, expressed or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States government or Lawrence Livermore National Security, LLC. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States government or Lawrence Livermore National Security, LLC, and shall not be used for advertising or product endorsement purposes.

This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

Electron emission from slightly oxidized δ -stabilized Plutonium
generated by its radioactivity,
and radiation induced ionization
and dissociation of hydrogen at its surface.

Wigbert J. Siekhaus, Art J. Nelson
Lawrence Livermore National Laboratory

Abstract

Energy dependent electron emission between zero and 1.4 keV generated by the natural reactivity of plutonium was measured by an electrostatic spectrometer with known acceptance angle and acceptance area. The electron spectral intensity decreases continuously except for a distinctive feature of unknown origin at approximately 180eV. The spectrum was converted to energy dependent electron flux ($e^-/\text{cm}^2 \text{ s}$) using the assumption that the emission has a cosine angular distribution. The energy dependent electron mean free path in gases and literature cross sections for electron induced reactions were used to determine the number of ionization and dissociation reactions per cm^2 second, found to be about $8 \cdot 10^8/\text{cm}^2\text{s}$ and $1.5 \cdot 10^8/\text{cm}^2\text{s}$, respectively, for hydrogen.

These results are to be used with caution until complementary measurements can be made, e.g. independent measurement of the total emitted electron current, since the results here are based on the assumption that the electron emission has a cosine angular distribution. That is unlikely to be correct.

This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344

1) Experimental Results.

Electron emission from a slightly oxidized ('golden') surface of plutonium of "typical composition", see Wolfer¹, into vacuum was measured using the hemispherical analyzer² of a X-ray Photoelectron Spectroscopy System (XPS system) while its X-ray source was turned off. The spectrometer's input axis was normal to the sample surface, and the distance between the spectrometer and the sample was one centimeter. The largest aperture was used (3mmX10mm) and the pass energy was set to the maximum, 178.95eV. The spectrometer has an acceptance angle of $\pm 4^\circ$. The ionization gauge was "on" during the run (43hrs) and during a background counting run with a non-radioactive sample in front of the analyzer and the plutonium sample on the sample holder, but removed from the acceptance area of the of the spectrometer. The results are shown in figure 1.

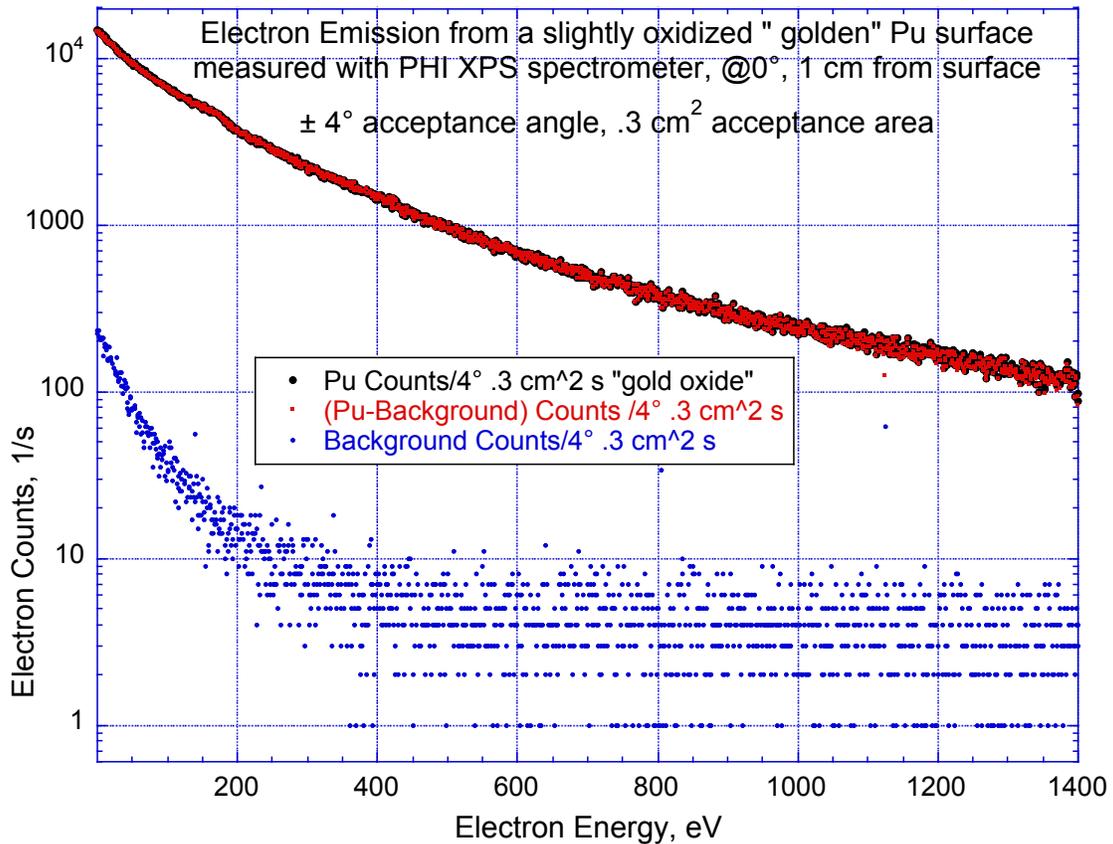


Figure 1. Electron count rate (counts per second) from a slightly oxidized ('golden') Plutonium surface. Signal with background counts are shown as black squares, background counts as blue circles, and (signal-background) counts as red open triangles.

The background counts per second are always at least a factor 10 below the signal counts per second. There is a noticeable "bump" at about 180eV in the measured

¹ Wolfer, W. G., A. Kubota, et al. (2007). "Density changes in Ga-stabilized delta-Pu, and what they mean." *Journal of Alloys and Compounds* **444**: 72-79..

² Physical Electronics PHI 5400

electron flux. In figure 2 the “smoothed” (using a span of 20 data points for smoothing) data of figure1 are plotted in black, and the derivative of the smoothed data in blue. It is evident in the derivative data that there is a distinctive – but so far undefined -process at about 180 eV and a less distinctive one at ~ 400eV. Electrons at these energies have a mean free path of only a few mono-layers (see figure 13). Hence they originate from the Pu-oxide layers. But there are no obvious Auger or photoelectric processes at those energies.

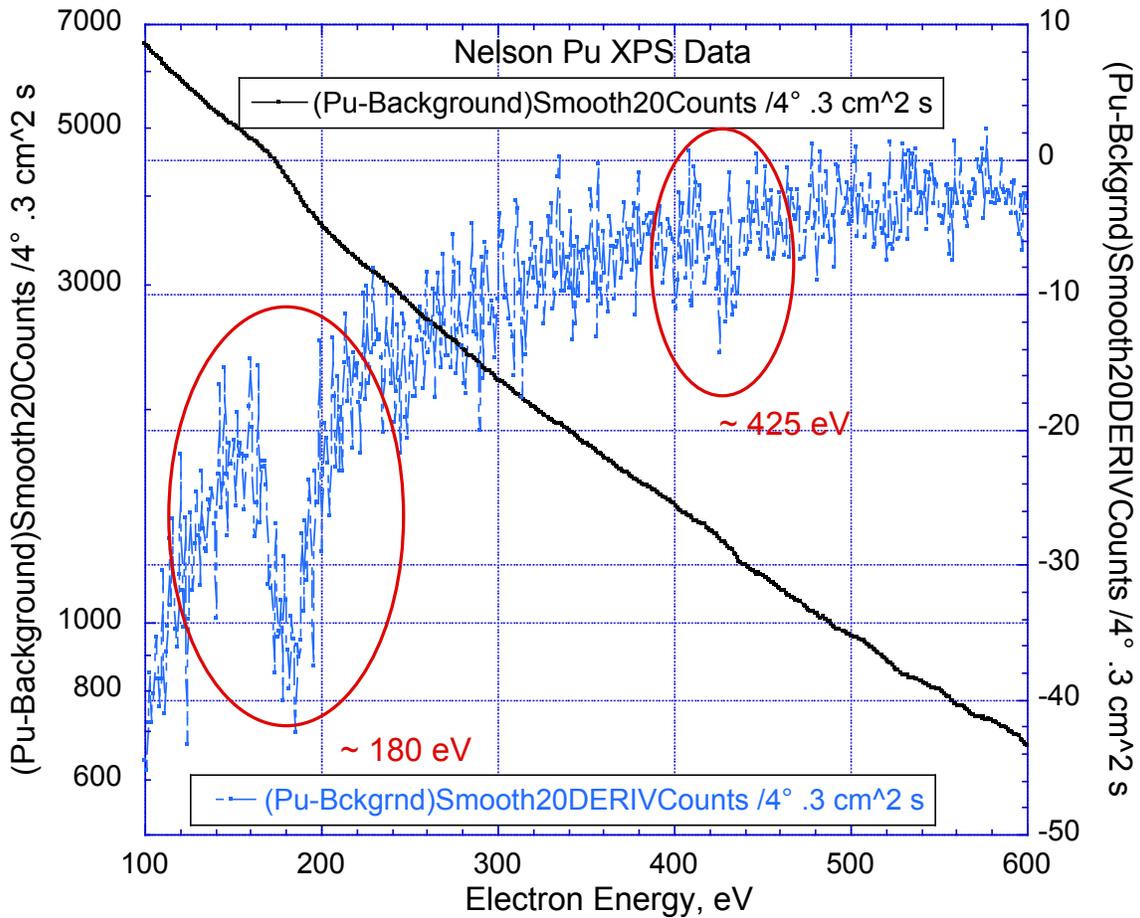
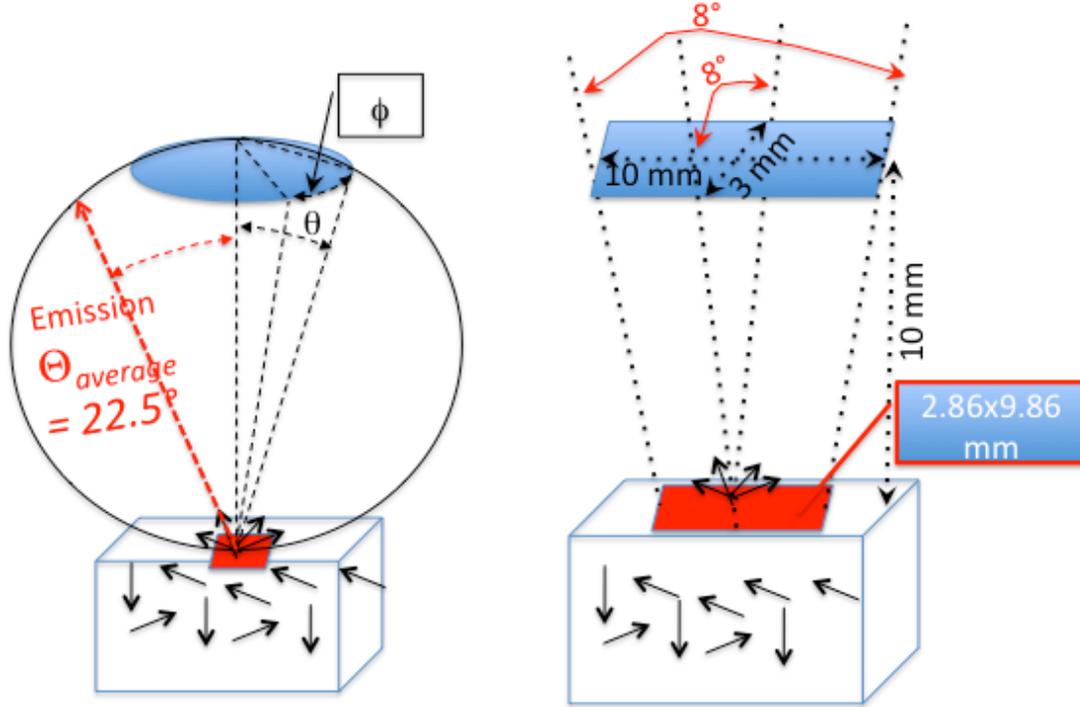


Figure 2. “Smoothed” electron count rate (counts per second) from a slightly oxidized (‘golden’) Plutonium surface shown in “black”, together with its derivative in “blue”. The derivative shows a distinctive feature of unknown origin at about 180 eV and a much less distinctive feature at ~ 425eV..

II) Conversion of experimental results (counts/s) to electron flux (e-/cm² s)

The data of figure 1 (counts/s) must be converted to “electrons emitted/cm²s” to calculate electron-induced chemistry, as illustrated in figure 3 below.



Flux at angle Θ : $flux(\Theta, All\Phi) = N_0 \cos \Theta$

Flux in cone of angle $\Theta = 4^\circ$

$$\int_{\vartheta=0}^{\Theta=4^\circ} \int_0^{2\pi} N_0 \cos \vartheta \sin \vartheta d\Phi d\vartheta = 2\pi N_0 \int_0^{\Theta=4^\circ} \cos \vartheta \sin \vartheta d\vartheta$$

$$= 2\pi N_0 * .002433 = .004866\pi N_0$$

for $\Theta = \pi/2$ the integral is $= 2\pi N_0 * .5 = \pi N_0$

The angle Θ into which 'on average' electrons are emitted

$$\Theta_{\text{average}} = \frac{\int_{\vartheta=0}^{\pi/2} \int_{\phi=0}^{2\pi} N_0 \vartheta \cos \vartheta \sin \vartheta d\Phi d\vartheta}{\int_{\vartheta=0}^{\pi/2} \int_{\phi=0}^{2\pi} N_0 \cos \vartheta \sin \vartheta d\Phi d\vartheta} = .392699 \rightarrow 22.5^\circ$$

Figure 3. The electron emission from a substrate in which the electron flux inside the substrate is N_0 electrons/cm²s for a cm² area of ANY orientation. With that assumption the flux from the surface has a “cosine distribution” with an ‘average’

emission angle of 22.5° , and the fraction of the total flux sampled by the spectrometer with an acceptance angle of 4° is .004866 of N_0 .

The analysis in figure 3 assumes that electrons inside the sample travel in all directions with equal probability. Then a spectrometer with an acceptance angle of $\pm 4^\circ$ around the normal to the surface samples only .004866 of the total flux emanating from its acceptance area. *The entrance aperture of the PHI 5400 analyzer is 3mmx10mm, and it has a narrow acceptance angle of 4° , resulting in an acceptance area of slightly less than $\sim 1\text{mmx}3\text{mm}$, $\sim .3\text{ cm}^2$.* Hence the counts per second of figure 1 must be divided by (.004866 * .3, actually .004866*.2819) to yield electrons/cm²s. The results of this division are plotted in figure 4. There is not unanimous opinion by representatives of the spectrometer's manufacturer whether the acceptance angle is 4° or 2° for this particular version of the spectrometer. If the angle is 2° , the fraction of emitted electrons measured would decrease by a factor 4, and the hence the calculated flux would increase by a factor 4.

Hence the counts per second of figure 1 must be divided by (.004866 * .3, actually .004866*.2819) to yield electrons/cm²s. The results of this division are plotted in figure 4. There is not unanimous opinion by representatives of the spectrometer's manufacturer whether the acceptance angle is 4° or 2° for this particular version of the spectrometer. If the angle is 2° , the fraction of emitted electrons measured by the spectrometer would decrease by a factor 4, and the hence the calculated flux would increase by a factor 4.

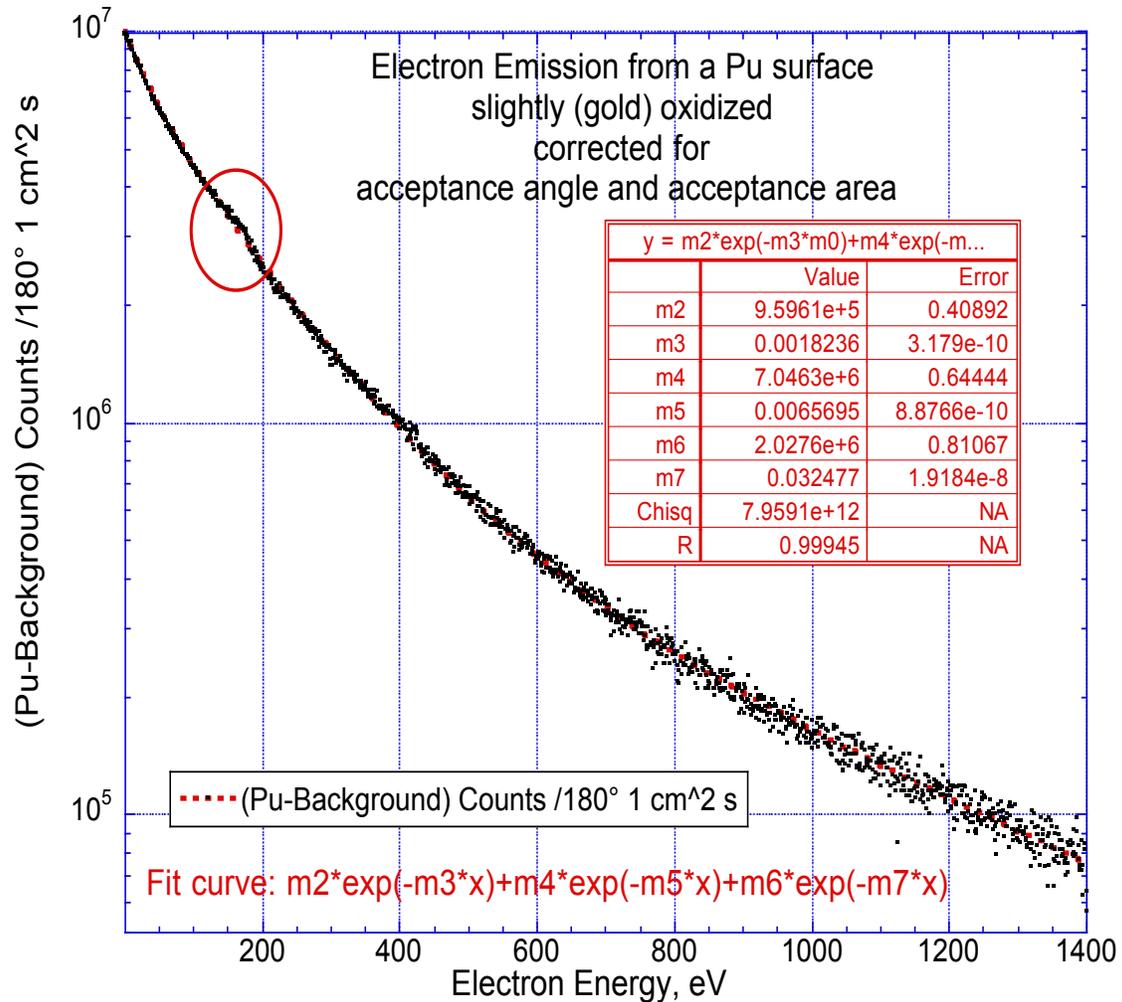


Figure 4. The electron flux/cm²s derived from “counts per second” of the spectrometer, together with a fit to the data. The fit is excellent, “R=.99945”.

The fit to the data is of high quality (R=.99945) and allows extrapolation to higher electron energies, as shown in figure 5. Furthermore, having an analytical function describing the energy flux facilitates subsequent calculations that determine radiation induced chemical reaction rates. Using the fit, the total electron current is the integral over the fit curve in figure 4, $2.8 \cdot 10^{-10}$ A. The total energy flux is the integral of the product $\{\text{fit}(E) \cdot E\}$ which yields $\rightarrow 7 \cdot 10^{-8}$ watt/cm². The specific activity of Pu 239 is $6.4 \cdot 10^{11}$ Becquerel/g, alpha emission with 5.5 MeV energy, resulting in an energy generation of .56 watt/g. The electron energy flux is clearly a small part of the total energy flux from the surface of plutonium.

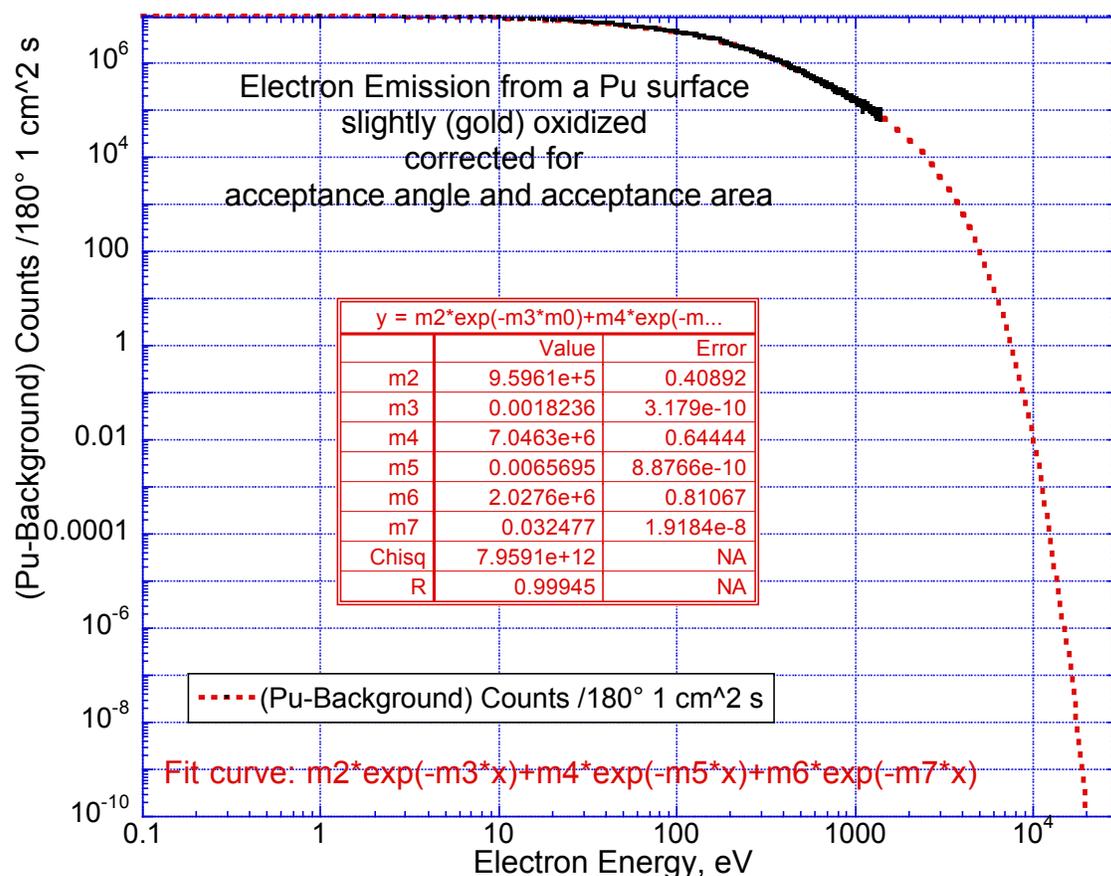


Figure 5. Extrapolation to higher electron energy of the exponential fit to the data measured below 1400eV predicts a very rapid decrease in electron flux with increasing electron energy.

The rate of chemical reactions, R_{ctn} (reactions/cm²s) is determined by equation (1) below;

$$R_{ctn}(E) (1/cm^2s) = \text{electron flux}(E)(e-/cm^2s) * \sigma(E) = \text{reaction cross section (cm}^2)$$

$$*M(E) = \text{number of accessible reactants (\#/cm}^2) \quad [1]$$

$M(E)$ = mean free path of electrons at energy E , (g/cm²)

* $\cos\Theta_{average}$, the cosine of the angle at which electrons are emitted from the surface 'on average' (see figure 1)

* {[Number of reactants/mole]/[Molecular weight, (g)]}

The cosine of the average angle of electron emission in equation (1) is calculated in figure 1.

Equation (1) has three factors:
the electron flux,
the reaction cross sections,

and the number of reactants accessible to an electron of energy E .

The flux is described above. The cross sections are available in the literature (e.g. Yoon³ for hydrogen). The reactive path length of electrons, $L_{e\text{-reactive}}$, in equation [1] is the amount of matter that electrons travel through with energy sufficient to cause a particular reaction (e. g. to cause dissociation if the matter is gaseous,). This length in equation [1] determines the total number of atoms or molecules that the electrons encounter as they traverse the material, and hence the total number of reaction products they can generate per unit area and unit time. A discussion of the minimum and maximum reaction path length follows. A typical path of a charged particle through matter is shown in figure 6 which illustrates the decrease in energy the charged particle experiences as it travels to its maximum path length, its “range”.

Figure 6 illustrates that the charged particle continues to travel beyond the first energy-losing encounter, the inelastic mean free path (IMFP), and has typically the majority of its collisions towards the end of its travel, called “range”, because at higher energy the collision cross sections increase with decreasing energy, see Yoon’s graph (figure 11) later in this paper.

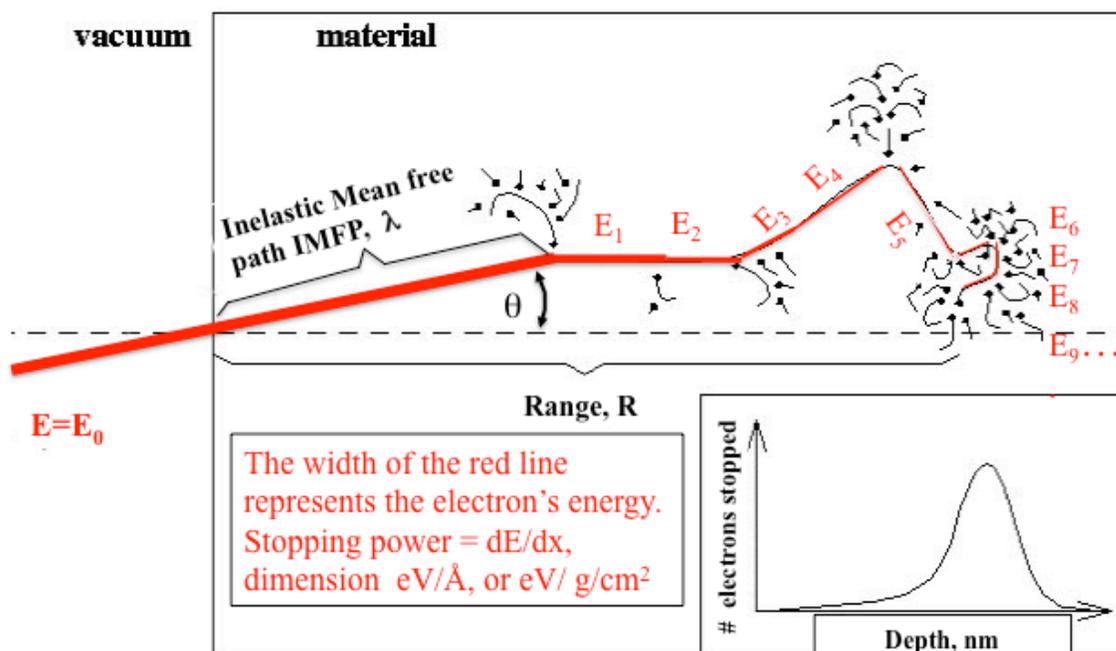


Figure 6. Schematic representation of a charged particle’s trajectory and associated energy loss along the electron’s path, illustrating the concept of “inelastic mean free path ‘IMFP’ ”, “range”, and “stopping power”. The electron’s energy is represented by the width of the path, decreasing with each inelastic reactive encounter with matter.

³ Yoon, J. S., M. Y. Song, et al. (2008). "Cross sections for electron collisions with hydrogen molecules." *Journal of Physical and Chemical Reference Data* 37(2): 913-931.

A Monte Carlo calculation by Postek⁴ following an electron of 1keV energy into a silicon substrate is shown in figure 7 depicting the trajectories actually followed, albeit only in two, rather than 3 dimensions. It is impossible to determine from figure 7 what the “IMFP” is, though one can conclude that the “range” is about 30 nm. As will be shown below, calculations predict an IMFP of about 2 nm, and a range of 30 nm. The difference between IMFP and range increases with increasing energy. For example: In hydrogen gas the dissociation energy is 4.5 eV, the ionization potential is 15.4 eV. Hence an electron with a kinetic energy of 20eV may loose most of its energy in a single interaction, while an electron with keV energy can sustain many such interactions before it is at the end of its range.

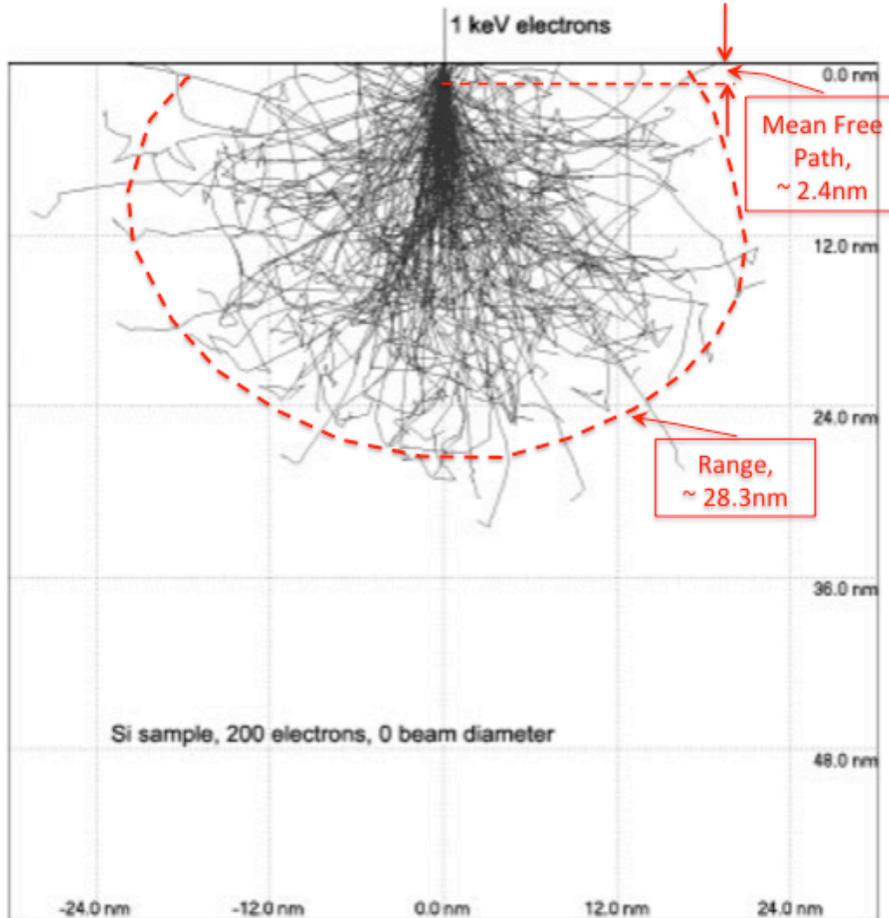


Figure 7. Schematic representation of the trajectory of an electron with 1 keV energy traversing silicon. After Postek⁵.

At minimum $L_{e\text{-reactive}}$ is the “inelastic electron mean free path [IMFP]”. This “IMFP” is the path-length, expressed either as matter mass traversed (e.g. [g/cm²], number of reactants passed by [#reactants/cm²], or simply - for a material of

⁴ Postek, M. T. and A. E. Vladar (2011). "Modeling for Accurate Dimensional Scanning Electron Microscope Metrology: Then and Now." *Scanning* 33(3): 111-125.

⁵ Ibid.

known density - length [cm,nm,Å]) at which “in the mean” electrons experience the first energy-losing interaction, discussed in III. 1) and III.2) below. The energy loss may be small or large. At maximum, $L_{e\text{-reactive}}$ is the mass “range” at which electrons are reduced to thermal energy, discussed in section V) below. It is apparent from both figure 5 and 6 that there can be a large difference between IMFP and range.

IV) The minimum $L_{e\text{-reactive}}$ is the “inelastic electron mean free path, ‘IMFP’”, λ (g/cm², #/cm², number of monolayers, number of accessible molecules or atoms, or Å)

IV. 1) The mean free path derived from total inelastic cross section data.

If all cross sections for inelastic electron-matter collisions are known, the inelastic mean free path (in “units” of ‘number of accesible molecules or atoms) can be derived from equation (1). :

$R_{ctn}(E)$ =the number of reactions/cm²s is given by

$$R_{ctn}(E)(1/cm^2s) = \text{electron flux}(E) (1/cm^2s) * \sigma_{TOTAL\ INELASTIC}(E)(cm^2) * M(E)(1/cm^2)$$

$$M(E) = \text{number of accessible reactants/cm}^2$$

$$= \text{density}(g/cm^3) * (\text{Avogadro's number}/\text{Molecular weigth}(g)) * \text{IMFP} (cm)$$

By definition the inelastic mean free path IMFP has been reached when one half of the electron flux has experienced an energy-losing ‘inelastic’ collision, i.e. when

$$R_{ctn}(E)(1/cm^2s) = (1/2)\text{electron flux}(E) (1/cm^2s). \text{ Therefore when:}$$

$$\frac{1}{2}\text{electron flux}(E) = \text{electron flux}(E) * \sigma_{TOTAL\ INELASTIC} * M(E)=\text{IMFP}. \text{ Hence}$$

$$M(E)=\text{IMFP} = 1/(2*\sigma_{TOTAL\ INELASTIC}) = \lambda(\text{reactants/cm}^2) \quad [2]$$

Equation [2] is only valid if only *inelastic* cross sections are included in the total cross section.

IV. 2) The inelastic mean free path derived from optical data.

The inelastic mean free path of electrons is of interest in all electron spectroscopy techniques {e.g. Auger Spectroscopy (AES), Photo Electron Spectroscopy (XPS)} since it determines the thickness of the surface layer sampled by the technique. Many sophisticated calculations of the IMFP for materials based on optical data have been done since Seah and Dench⁶ derived IMFP’s by fitting

⁶ Seah, M. P. Dench W. (1979). "Quantitative electron spectroscopy of surfaces: a standard data base for electron inelastic mean free paths in solids"

curves to experimental data. {see Powell et al.⁷ (2011) for IMFP calculations of 41 elements and references therein}. Figure 8 shows Powell's IMFP curves (λ in Å) for 41 elements. (from Lithium to Bismuth). Alkali metals produce curves that deviate most from the "typical" pattern. Despite these differences, the IMFP's do NOT differ dramatically, as figure 8 below shows, keeping in mind, however, that figure 8 is a double logarithmic plot. The spread in the curves would be much smaller if the range were expressed in g/cm² rather than Å.

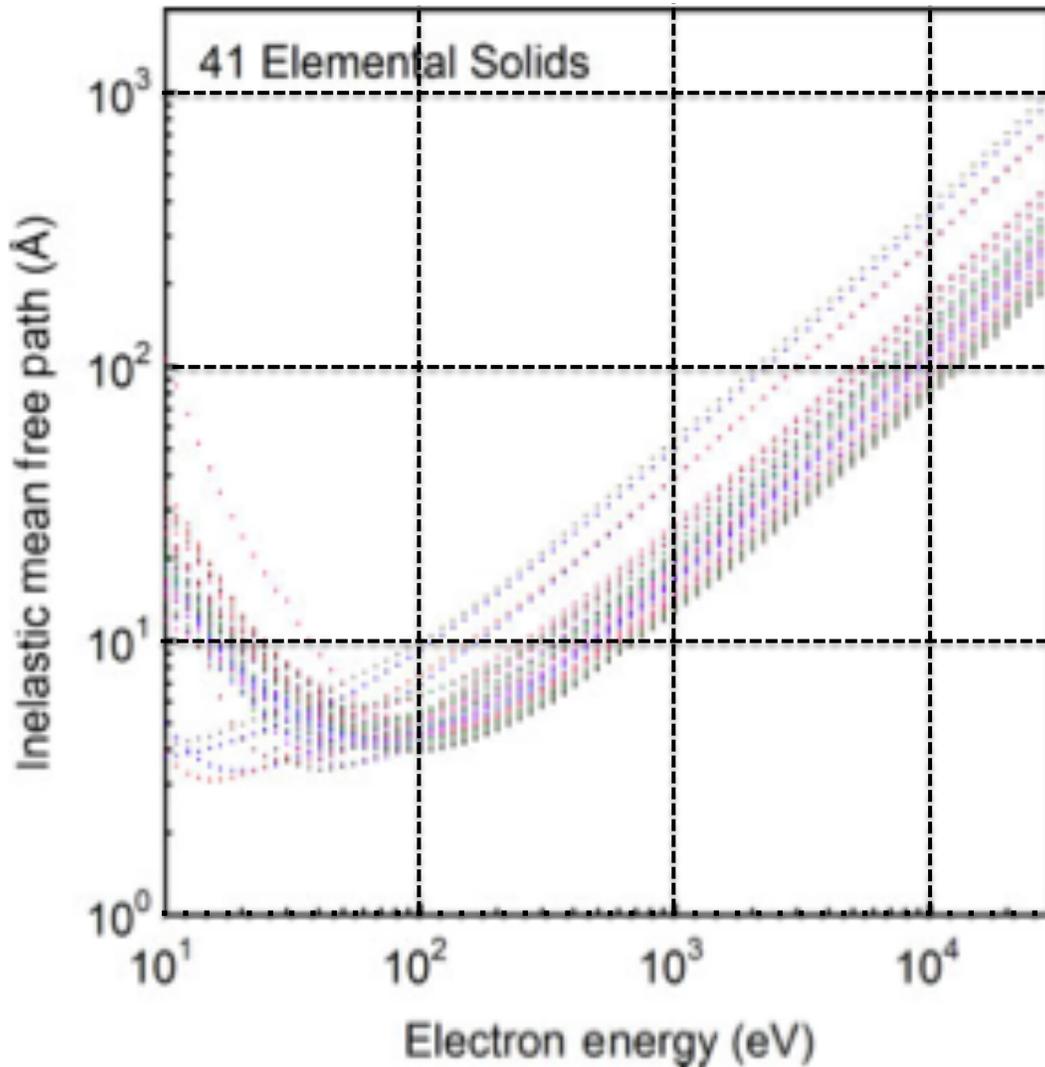


Figure 8. IMFP for 41 elements, after Powell et al.

Surface and interface analysis [0142-2421] **vol.1**, (no.1): pg:2 -11.

⁷ Powell, C. J., S. Tanuma, et al. (2011). "Calculations of electron inelastic mean free paths. IX. Data for 41 elemental solids over the 50 eV to 30 keV range." *Surface and Interface Analysis* **43**(3): 689-713.

V) The mean free path of molecules in the gas phase.

It is sometimes of interest to determine the number of reaction products within the reactant's mean free path, rather than the number of reactants in the electron's mean free path, since one half of the reaction products will strike the substrate without any collision with other molecules, and initiate a reaction on the surface. . The mean free path of *molecules* in a gas $\lambda_{\text{molecule}} = 1/(2.5\pi*d_0^2*n)$, where n is the number density and d_0 is the molecular diameter. {e.g. $d_0 = (\sim 2 \rightarrow 6 * 10^{-8} \text{ cm}, \sim 2.93 * 10^{-8} \text{ cm}$ for hydrogen. The diameter of the hydrogen atom is about twice the Bohr diameter, i.e. 1.1 \AA). Hence the number of molecules within a distance $\lambda_{\text{molecule}}$ from a surface of area 1 cm^2 is $= \lambda_{\text{molecule}}*n = 1/(2.5\pi*d_0^2) = 5.63 * 10^{14}/\text{cm}^2$ for hydrogen, $= \sim <$ the equivalent of one monolayer adsorbed onto a surface. This number is independent of pressure. Only one half of the reaction products travel in a direction towards the surface, assuming that the reaction with electrons that produces the reaction product did NOT generate a preferred direction for the reaction products.

VI.1) The range of electrons in hydrogen, NIST data.

Figure 9 shows the range of electrons and the stopping power for electrons in hydrogen (NIST data).

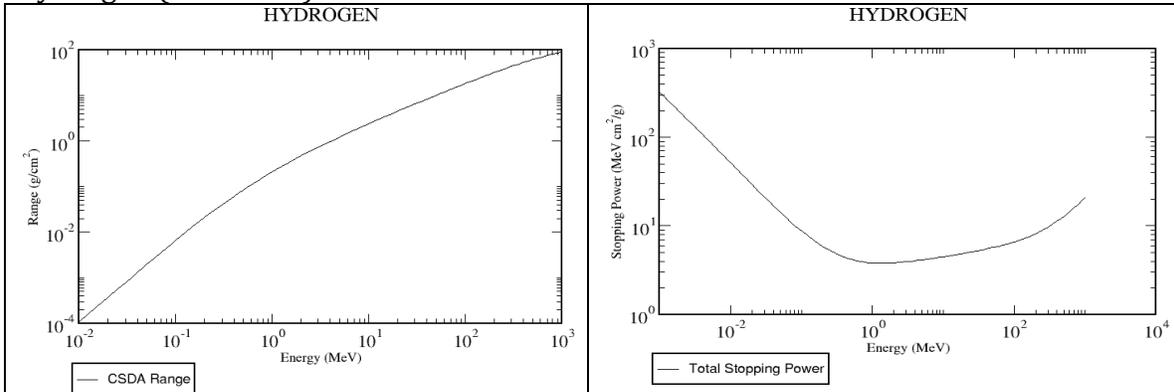


Figure 9. Left image: “Continuous Slowing Down” range, g/cm² ,(CSDA=Continuous Slowing Down Analysis) of electrons in hydrogen, with electron energies from .01 to 1000 MeV. Right image: “Total Stopping Power” , MeV cm²/g, for hydrogen. The site provides stopping power for energies higher than 1 keV, but range only for energies above 10 keV.

From www.nist.gov/PhysRefData/Star/Text/ESTAR.html

As is discussed in detail at NIST's website, range calculations become increasingly more unreliable with decreasing energy “due to the lack of shell corrections which are required when the velocity of the incident electron is no longer large compared to the velocities of the atomic electrons, especially those in the inner shells.”

Therefore NIST stops the CSDA curves at 10keV. Nevertheless, the same NIST site shows stopping power curves down to 1keV, see figure 9.

VI.2) Comparison of 1keV electron range in silicon (NIST data) with Monte Carlo Calculations, and with the calculated inelastic mean free path.

To make a visual comparison of the calculated IMFP's of Powell, and NIST's range, we extend NIST's CSDA range curve for silicon to energies below 10keV and plot the IMFP and CSDA for silicon in figure 9. We use silicon for comparison since a Monte Carlo calculation for silicon is available at 1keV (see figure 6). Comparison between the IMFP and range at 1 keV in figure 9 provides a quantitative measure of the difference between mean free path and range, complementing the graphic depiction in figure 5. In addition, the agreement between the range observed for silicon in figure 6 and the range in figure 9 derived from NIST's data suggests that at least down to 1 keV the extrapolation of NIST's range data is reasonable.

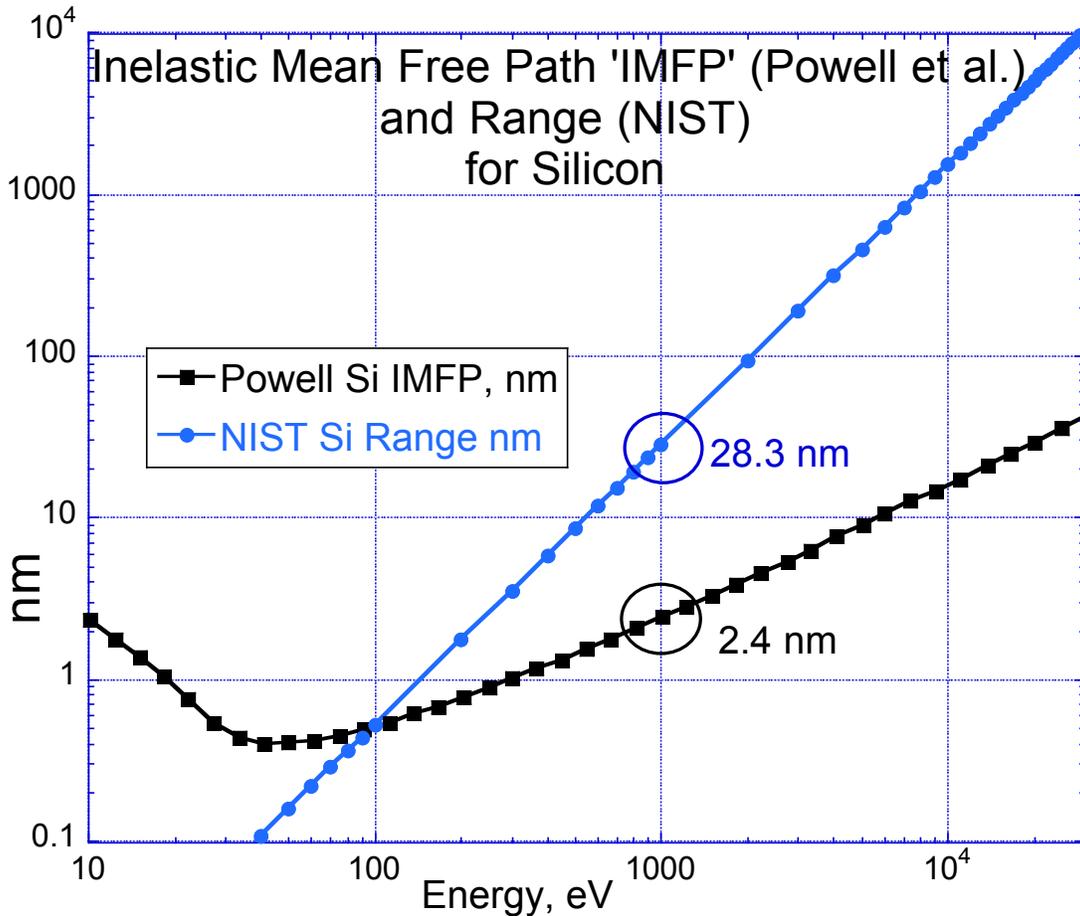


Figure 10. Comparison of the inelastic mean free path and the range of electrons in silicon.

Clearly it is NOT reasonable to extrapolate the range to energies to 100eV, since the range must be at least twice the mean free path because at the mean free path only half the electrons have undergone an “energy-losing” collision. The “energy-losing” collision does NOT in many cases reduce the electron’s energy to thermal energy. For instance, an electron impact ionization of hydrogen requires ~ 13 eV, hence a 100 eV electron can induce at least 6 six ionizations before it is reduced to thermal energy. Since both NIST’s data and Powell’s calculations are consistent with experimental data and with Monte Carlo calculations for silicon, one can conclude from figure 9 that at 1 keV the an electron has more than 12 (28.3 nm divided by 2.4nm) collisions before it is thermalized. Many more than 12, because with each collision its energy decreases by ΔE , and its inelastic mean free path at $E-\Delta E$ is shorter than at $E=1\text{keV}$, until its energy has decrease to below 100eV, where Powell’s IMFP has a minimum.

VII) Reaction of electrons emanating from a plutonium surface with hydrogen molecules.

The cross sections for $e^- \rightarrow \text{H}_2$ reactions are given in Yoon⁸ and shown in graphical from in figure 10. In figure 10 the total cross section, labeled “tot” includes both elastic (labeled “elas”) and inelastic collisions (all other collisions). The energy loss in momentum transfer collisions (labeled “mom transf”) is very small, since the mass of the electron is about two thousand times smaller than the mass of a hydrogen molecule and will NOT be included as an inelastic collision. Figure 11 indicates that reactions of practical interest, e.g. ionization, dissociation, have significant cross sections only above about 9 eV electron energy. In section III.1) it was pointed out that only the total inelastic cross section is relevant for determining the inelastic mean free path. Figure 12 shows the *inverse* of the total *inelastic* cross section for hydrogen, derived by subtracting the *elastic* cross section in figure 10 from the *total* cross section.

⁸ *ibid.*

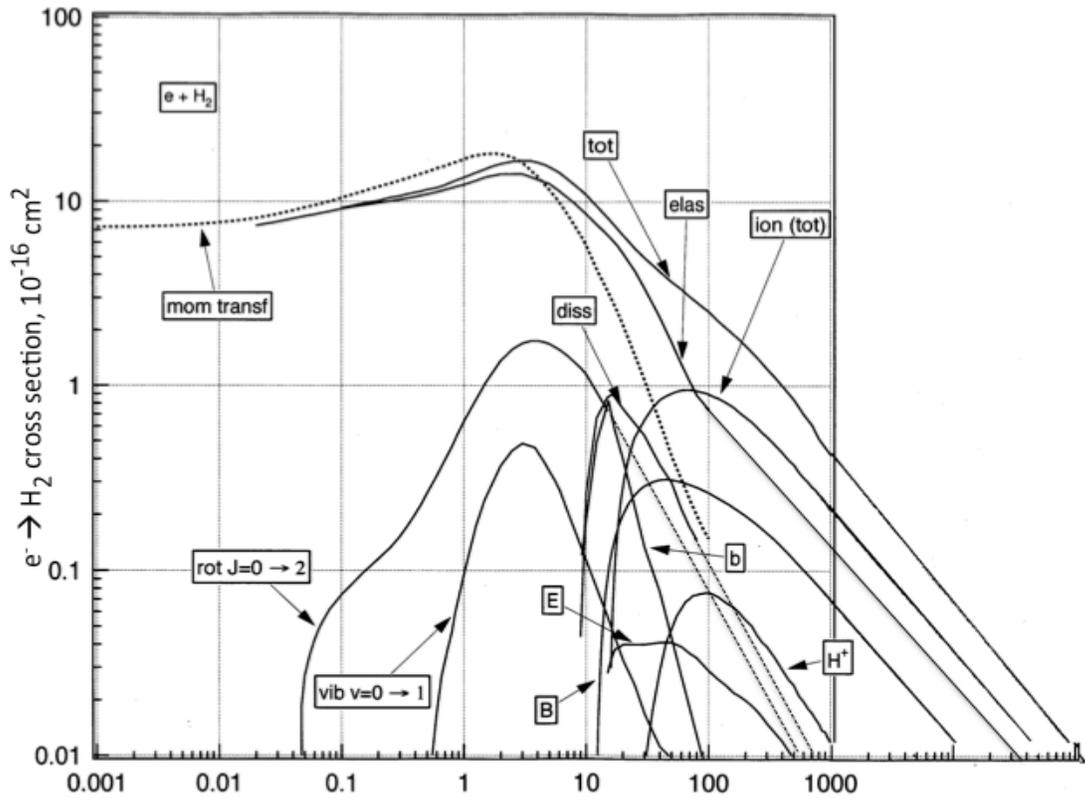


Figure 11. Reaction cross sections of electrons with hydrogen molecules as a function of electron energy. Some of the cross-sections are extended beyond their original range or beyond their maximum energy (1 keV) of the original graph using a continuation of their slope within the graph.

(Molecules/cm²)*10⁻¹⁶

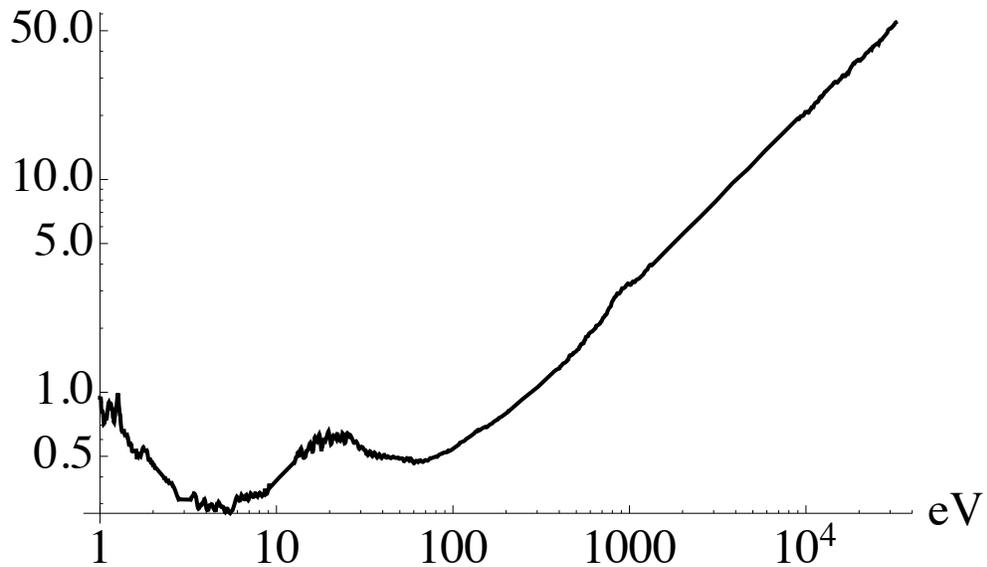


Figure 12. The inverse of the total *inelastic* cross section of hydrogen, expressed as molecules/cm². After Yoon et al.

VII.1) The “minimum number of reactions”, i. e the reactions that occur within the inelastic mean free path of electrons.

In figure 13 the inelastic mean free path of electrons in hydrogen is plotted using the inverse of (2* total inelastic cross section shown in figure 12). Also shown is the electron range in hydrogen derived from NIST data by converting NIST's range given in g/cm² to 'number of hydrogen molecules/cm²' by assuming hydrogen to be in solid form, with a density of .088 g/cm³. Furthermore Powell's IMFP data for Lithium are plotted also for comparison after converting Å to 'number of hydrogen molecules/cm²' by assuming that solid hydrogen at .088g/cm³. Lithium is used for comparison, since all elements in the first column of the periodic table (the Alkali metals) show very similar behavior, and since Lithium is closest in density to solid hydrogen.

Figure 13 demonstrates that the concept of deriving the IMFP from measured total inelastic cross sections is consistent in magnitude and energy dependence with the IMFP derived from Powell's calculations. The NIST range is included to illustrate that electrons will undergo many collisions – and create many more reaction products than occur in the IMFP - before they are thermalized.

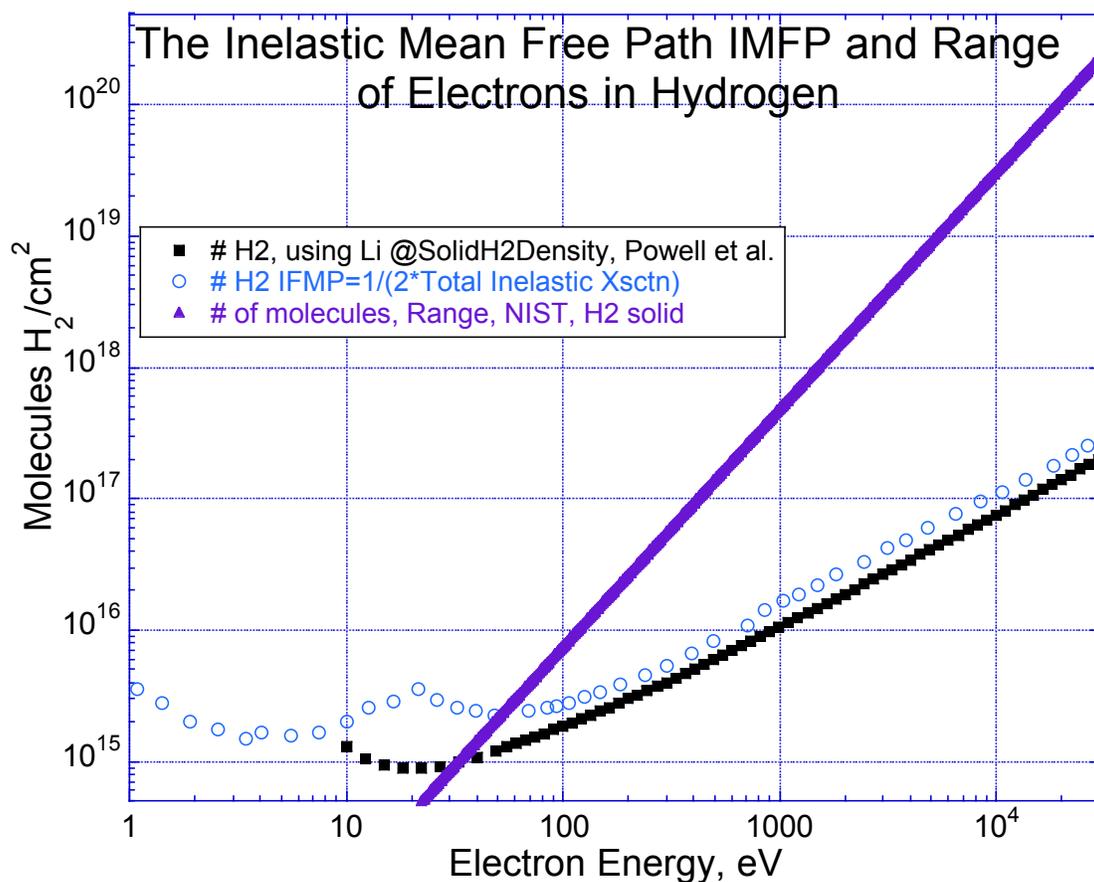


Figure 13. Comparison of the inelastic mean free path calculated from the total inelastic cross section derived from Yoon et al., and of that derived from Powell’s calculation for Lithium (at solid hydrogen density) with NIST’s range.

The number of reaction products $R_{ctn}(E)$ occurring within the energy dependent inelastic mean free path are now calculated at each energy E , using equation (1), employing the energy dependent electron flux, electron mean free path, and Yoon’s cross section $\sigma(E)$ of each type of reaction, and the energy independent correction factor for mass density. The total number “ R_{ctn} ”, e.g. for the dissociation reaction, is found by integrating $R_{ctn_{dissociation}}(E)$ over the energy range where dissociation cross sections exist, i.e. $R_{ctn_{dissociation}} = \int R_{ctn_{dissociation}}(E) dE$. The “ R_{ctn} ” values for a few electron induced reaction products are listed in Table 1 below. The program “Mathematica” is used for the numerical integration. Mathematica interpolates between energy values for each set of data, so that data sets with data at different energy-values can be used.

Cell Type: Pu surface facing H ₂ gas	Rctn _{All Ionization} H ₂ +e ⁻ → H ₂ ⁺ & H ⁺	Rctn _{H2->H+ only} H ₂ +e ⁻ → H ⁺	R _{Dissociation} H ₂ +e ⁻ → 2H
Rctn/cm ² s=# of H ₂ reacted within <i>electron</i> inelastic mean free path =1/2*(Total Inelastic Cross Section)	8.1*e+8	5.1*10 ⁷	1.5*10 ⁸
Rctn/cm ² s=# of H ₂ reacted within <i>electron</i> inelastic mean free path derived from Powell et al.	2.7*10 ⁸	1.8*10 ⁷	3.3*10 ⁷
Rctn/cm ² s=# of H ₂ reacted within mean free path of hydrogen molecules in hydrogen gas	5.5*10 ⁷	3.5*10 ⁶	1.4*10 ⁷
Rctn/cm ² s=# of H ₂ reacted within <i>electron</i> inelastic mean free path =1/2*(Total Inelastic Cross Section) for 7-100eV, plus within the NIST range for 100->3000eV	4.8*10 ⁹	2.8*10 ⁸	2.5*10 ⁸

Table 1. The reaction products produced by electrons emitted from a plutonium surface into hydrogen gas.

VII.2) The “maximum number of reactions”, i. e the reactions that occur within the “total energy range” (NOT “range=molecules/cm²s”) of electrons.

A more realistic *estimate* of the reactions occurring within the *energy* range of electrons incorporates the change in mean electron energy and the associated change in reaction cross section and in the inelastic mean free path (expressed in molecules/cm²) occurring with each energy-losing inelastic collision. Assume that the change in energy in each inelastic collision is ΔE. The number of collisions “n” that can occur for an electron of energy E is then n=E/ΔE, where n is the integer part of the fraction, and n=0 for E/ΔE<1. To visualize the process, see figure 12: An electron entering the gas with 1 keV energy has its first collision while traversing the IMFP(E) number of molecules shown in figure 11 and 12 at 1 keV with the reaction probability defined by Yoon’s crosssection at 1keV(see figure 10), loses on average ΔE eV, continues to traverse the next IMFP at (1keV- ΔE), reacts within the

IMFP(1keV-ΔE) with the probability defined by the cross section at (1keV-ΔE), loses on average ΔE, continues with energy (1keV-2ΔE) ... etc., until it has reached an energy so low that further reactions of interest are impossible. With that assumption, the number of reactions due to electrons of any energy E is given by equation [3] below

ΔE = mean energy loss per inelastic collision

IMFP = inelastic mean free path

$|n(E)| = |E / \Delta E|$, where $|n(E)|$ is an integer with minimum value = 0

$$R_{ctn}(E) = Eflux(E) * \int_{E_{min}}^{E_{max}} \sum_{n=0}^{|n(E)|} \sigma(E - n * \Delta E) * M(E - n * \Delta E) dE \quad [3]$$

$R_{ctn}(E)$ = number of reactions/cm²s

$Eflux(E)$ = electron flux at energy E, 1/cm²

$\sigma(E)$ = cross section of the reaction of interest, cm²

$M(E)$ = number of accessible molecules per IMFP, 1/cm²

E_{min} = minimum energy of electron flux

E_{max} = maximum energy of electron flux

Cell Type: Pu surface facing H ₂ gas	R _{ctn} All Ionization H ₂ +e ⁻ → H ₂ ⁺ & H ⁺	R _{ctn} H ₂ ->H ⁺ only H ₂ +e ⁻ → H ⁺	R _{Dissociation} H ₂ +e ⁻ → 2H
R _{ctn} /cm ² s=# of H ₂ reacted within <i>electron Energy range</i> using the inelastic mean free path at each energy derived from Powell et al.	4.4*10 ⁹	2.8*10 ⁸	6.7*10 ⁹

Table 2. The “maximum” number of reactions/cm² s based on the assumption that electrons lose an energy of ΔE=20eV in each mean free path and continue to react at successively lower energy until they are below the energy where inelastic collisions can occur. See figure 6 for illustration.

The program “Mathematica” is used to solve equation [3] for the numerical integration and summation, using a working precision of 60 digits.

The reaction rates “R_{ctn}” are nevertheless only estimates, since the energy loss ΔE=20eV in each mean free path is only a guess.

VIII. Summary

The spectroscopically measured emission of electrons from plutonium generated by its natural radioactivity is high, but the electron energy flux, $\int \text{flux}(E) \cdot E \cdot dE$ integrated from 0 to 1400 eV is nevertheless much smaller than the total energy generated by natural radioactivity of one gram of Pu239, hence does not violate conservation of energy.

The electron flux derived from the spectroscopy data together with known reaction cross sections for hydrogen leads to substantial dissociation and ionization reactions per cm^2 in the mean free path of those electrons.

There is, however, a likely source of error in the assumption that the electron emission has a cosine distribution.

Therefore the results must be used cautiously until they are confirmed by other measurements, such as, e.g. measurement of the angular distribution of electron emission and of the total electrical current emitted by the sample.