INVERSE MEAN FREE PATH, STOPPING POWER, CSDA RANGE, AND STRAGGLING IN POLYSTYRENE FOR ELECTRONS OF ENERGY ≤10 keV

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## Scientific Report

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Inverse Mean Free Path, Stopping Power, CSDA Range, and Straggling in Polystyrene for Electrons of Energy up to 10 keV

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The interaction of electrons with solid polystyrene, -(C₆H₅)-, is described based on a model insulator theory to account for the response of the valence electrons, and carbon K-shell ionization cross sections derived from atomic, generalized oscillator strengths. Contributions to the inverse mean free path and energy loss due to these two excitation processes are tabulated for incident electrons with energies from 5 eV to 10 keV. Electron ranges in the continuous-slowing-down approximation and straggling are tabulated for electrons with energies from 15 eV to 10 keV.
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I. INTRODUCTION

A quantitative description of the interaction of electrons with solids over a wide range of energies is a subject of importance in a wide variety of basic and applied physical problems. Theoretical calculations of energy loss and range of electrons in many materials have formed the basis of at least two extensive tabulations.\textsuperscript{1,2} Both of those tabulations are based on the Bethe theory of stopping power and are restricted to electron energies \( \geq 10 \text{ keV} \). To complement these results we have employed several theoretical models to provide calculations of inverse mean free path, energy loss, csda range, and straggling for electrons with energies \(< 10 \text{ keV}\). Tables of these quantities are now available for the solids Al and Al\(_2\)O\(_3\) (Reference 3); Si and SiO\(_2\) (Reference 4); Ni, Cu, Ag, and Au (Reference 5); and Ge and GaAs (Reference 6). These tables should provide useful guides for interpretation of experimental data as well as input for calculations in applied areas.

The work presented here for the organic insulator polystyrene, -(C\(_8\)H\(_8\))-\(_n\), employs a model insulator theory\textsuperscript{3,6,7} to describe the response of the valence band electrons. The states of the tightly bound K-shell electrons are assumed to retain a free-atom-like character so the excitation of these electrons to the continuum is described by cross sections derived from atomic, generalized oscillator strengths (GOS's).\textsuperscript{8} In the following sections we describe the calculation of differential inverse mean free paths (DIMFP's) for interaction of an electron with the valence band or carbon K-shell electrons in the solid polystyrene and the derivation of inverse mean free path (IMFP) and energy loss from these DIMFP's.
Results are presented graphically for the IMFP and energy loss (or stopping power of the polystyrene) and in tabular form for the IMFP, stopping power, csda range, and straggling for electrons of energy from a few electron volts through 10 keV.

II. GENERAL FORMULATIONS

A charged particle passing through a solid interacts with a large number of electrons simultaneously, and it is thus appropriate to speak of a mean free path of the charged particle for energy transfer to the solid. Assuming the effect of the charged particle on the medium may be described in first Born approximation, the inverse mean free path, differential in momentum transfer $\Pi k$, and energy transfer $\Pi \omega$, for a particle of velocity $\vec{v}$ is given by

$$\frac{d^2 \mu}{dk d\omega} = \frac{2e^2}{\pi \hbar v} \frac{1}{k} \operatorname{Im} \left[ \frac{-1}{\varepsilon(k,\omega)} \right]$$

where $\varepsilon(k,\omega)$ is the dielectric response function of the solid.\textsuperscript{9,10} We assume in this work that the solid is isotropic and homogeneous so that $\varepsilon$ is a scalar function of $k$ and $\omega$. 
For our calculations of inverse mean free path, stopping power, etc., it is sufficient to compute inverse mean free paths differential in energy transfer only. This differential inverse mean free path (DIMFP) for energy loss \( \eta \omega \) by an electron with energy \( E = mv^2/2 \) in the solid is given by

\[
\tau(E, \eta \omega) = \frac{d\mu}{d(\eta \omega)} = \frac{1}{\pi a_0 E} \int_{k}^{k+} \frac{dk}{k} \text{Im}\left[ \frac{-1}{\epsilon(k, \omega)} \right],
\]

where \( \epsilon_k = \sqrt{2m \left[ \sqrt{E + \sqrt{E - \eta \omega}} \right]} \) and \( a_0 = \hbar^2/m e^2 \). This expression assumes that the energy-momentum relation for a swift electron in the solid does not differ appreciably from that of a free electron in vacuum.

Given \( \epsilon(k, \omega) \) for the solid, the quantities of interest here follow directly from \( \tau(E, \eta \omega) \). The inverse mean free path of the electron, \( \mu \), is given by integrating over allowed energy transfers as

\[
\mu(E) = \int d(\eta \omega) \tau(E, \eta \omega).
\]

The rate of energy loss of the electron, or the stopping power of the medium, is given by

\[
S(E) = -dE/dx = \int d(\eta \omega) \eta \omega \tau(E, \eta \omega),
\]

and the mean square energy loss per unit path length by

\[
\langle \Delta E \rangle^2 (E) = \int d(\eta \omega)(\eta \omega)^2 \tau(E, \eta \omega).
\]
With these results we may calculate the range of an electron in the continuous-slowing-down approximation (csda range) by

$$R_0(E) = \int_{E_0}^{E} dE'/S(E')$$

(6)

and the mean square fluctuation in the range or "range straggling" will be calculated from Eqs. (4) and (5) as

$$\langle (R - R_0)^2 \rangle_{AV} = \int_{E_0}^{E} dE' \frac{\omega^2(E')}{[S(E')]^3}$$

(7)

For our tabulations we take the lower limit in the integrations of Eqs. (6) and (7) as $E_0 = 10$ eV.

### III. DIMFP FOR THE VALENCE BAND

The model insulator theory used to derive the DIMFP for interaction of an electron with the valence band electrons has been described and employed in several previous calculations. Instead of repeating the detailed formulae here, we present graphically some of the steps required to obtain the energy loss function, $\text{Im}[-1/\varepsilon(k,\omega)]$, which is the key ingredient in the calculation of the DIMFP.

The first step in applying the model insulator theory is to fix the adjustable parameters by fitting the theoretical expression for the imaginary part of the dielectric response function in the optical limit ($k\rightarrow 0$) to experimentally determined values of this quantity. In Figure 1 we
Fig. 1 The imaginary part of the optical dielectric function for polystyrene as measured (solid line) and as calculated from a model insulator theory.

Fig. 2 The energy loss function (in the optical limit) for polystyrene as calculated from experimental data (solid curve) and from a model insulator theory (dashed curve).
show the results of the fit using experimental values obtained by Inagaki et al.\textsuperscript{12} for polystyrene. The valence band is assumed to result from a combination of three ground state orbitals and the fit shown in Figure 1 gives the following set of parameters (as defined in References 3, 6, and 7):

<table>
<thead>
<tr>
<th>i</th>
<th>( \alpha a_0 )</th>
<th>( \eta \omega_{B_i} ) (eV)</th>
<th>( n_i )</th>
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<td>3</td>
<td>1.90</td>
<td>25.0</td>
<td>7.5</td>
</tr>
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</table>

where \( n_i \) is the number of valence electrons per monomeric unit accounted for by each level and \( \beta = 1/2 \) for each of the three levels. Note that we account for 41.5 electrons per monomeric unit in the valence band instead of the expected 40. This redistribution of electron numbers between the core and valence electrons is due to oscillator strength coupling between the core and valence levels.\textsuperscript{12} Since there are 56 electrons per monomeric unit, we will account for an effective number of 14.5 carbon K-shell electrons (or 7.25 K shells) per monomeric unit in our calculations of DIMFP's in Section IV. The density of polystyrene for these calculations is taken to be 1.05 g/cm\(^3\). With the molecular weight of 104.14 g/mole for polystyrene, this density corresponds to \( 6.07 \times 10^{-3} \) monomeric units/A\(^3\).

As a further comparison we show in Figure 2 the energy loss function in the optical limit calculated from the experimental data\textsuperscript{12} and calculated from the model insulator theory using the parameters determined above. Quite reasonable agreement is seen in both Figures 1 and 2.

The extension of the energy loss function to arbitrary values of momentum transfer as determined by the model insulator theory is illustrated in Figure 3 where energy and momentum transfer are given in atomic units.
Fig. 3 Extension of the energy loss function into the momentum transfer plane as prescribed by a model insulator theory.
Calculations of DIMFP from Eq. (2) are illustrated in Figure 4 where we plot $E_t$ as a function of energy transfer for several values of electron energy, with all quantities expressed in atomic units.

IV. DIMFP FOR CARBON K SHELL

From a general expression for the dielectric function of a homogeneous, isotropic system, we may show that for values of $\omega$ which correspond to ionization of a given inner shell in a solid that

$$\text{Im} \left[ -1/\epsilon(k,\omega) \right] \approx \text{Im} \epsilon(k,\omega) \approx \frac{2\pi ne^2}{\hbar \omega} \frac{df(k,\omega)}{d\omega},$$

where $df/d\omega$ is the GOS and $n$ is the number of those inner shells per unit volume in the solid. Equation (2) thus leads to

$$\tau(E,\hbar\omega) = \frac{8\pi a_0^2 n}{(E/R)(\hbar\omega/R)} \int_{k^-}^{k^+} \frac{dk}{k} \frac{df(k,\omega)}{d(\hbar\omega)},$$

where $R = e^2/2a_0 = 13.6$ eV.

Generalized oscillator strengths for ionization of electrons from the K shell of carbon have been calculated by McGuire. These GOS values have been used in Eq. (9) to obtain the differential cross section $d\sigma/d(\hbar\omega) = \tau/n$. Some typical results are shown in Figure 5 for several values of electron energy. The binding energy of the K-shell electrons in carbon is $\sim 282$ eV. As discussed in Section III, we account for an effective 7.25 K shells per monomeric unit in calculating $\tau$. 
Fig. 4  DIMFP's for excitation of electrons from the valence band of polystyrene as determined by a model insulator theory for several values of incident electron energy $E$. 
Fig. 5. Differential cross section for ionization of the K shell in carbon as derived from generalized oscillator strengths. $E$ is the Rydberg energy $e^2/2a_0 = 13.6$ eV.
V. EXCHANGE CORRECTED DIMFP's AND FORMULAE FOR THE TABULATIONS

We have included the effect of electron exchange in our calculations in a simple manner based on the form of the Mott formula (nonrelativistic Møller formula) for scattering of an incident electron with a free electron. The cross section for finding a scattered electron with energy \( W \) per unit energy interval is given by

\[
\frac{d\sigma}{dW} = \frac{4\pi e^4}{E} \left[ \frac{1}{W^2} + \frac{1}{(E-W)^2} - \frac{1}{W(E-W)} \right]
\]

for an incident electron of energy \( E \), except for energies close to \( W = 0 \) and \( W = E \). Near \( W = 0 \) and \( W = E \) the interference term (third term on the right side of Eq. (10)) is small compared with the first or second term, respectively.

The DIMFP for excitation of an electron from a particular state \( i \) may be written in the form

\[
\tau_i(E, \omega) = \frac{1}{E} F_i(E, \omega).
\]

If we assume that the width of the level from which an electron is excited is quite narrow, we obtain from Eq. (11) the DIMFP for production of a secondary electron with energy \( E_s \) as

\[
\tau_i^S(E, E_s) = \frac{1}{E} F_i(E, E_s^B + E_s).
\]
where $E_i^B$ is the binding energy of the $i^{th}$ level (a positive quantity).

The exchange corrected DIMFP is taken as

$$\tau_{i}^{\text{exc}}(E, \mathcal{H}_0) = \frac{1}{E} \left\{ F_i(E, \mathcal{H}_0) + F_i(E, E + E_i^B - \mathcal{H}_0) \right\}$$

$$- \left[ 1 - \sqrt{\frac{E_i^B}{E}} \right] \left[ F_i(E, \mathcal{H}_0) F_i(E, E + E_i^B - \mathcal{H}_0) \right]^{1/2}. \quad (13)$$

Since $E_{i} = 1/(\mathcal{H}_0)^2$ for large $E$ and $\mathcal{H}_0$, Eq. (13) reduces in this limit to the form given by Eq. (10). The factor $1 - \sqrt{E_i^B/E}$ reduces the contribution of the third term in Eq. (13) as $E \to E_i^B$. This form for the exchange corrected DIMFP has been used in our calculations for the inner shell and for the valence bands (since our model assumes the width of these levels to be quite narrow).

If we now define the more energetic of the two electrons after collision to be the primary and account for exchange through Eq. (13), Eq. (3) gives the contribution to the inverse mean free path due to excitation of an electron from the $i^{th}$ level as

$$\mu_i(E) = \int_{E_i^B}^{(E+E_i^B)/2} d(\mathcal{H}_0) \tau_i^{\text{exc}}(E, \mathcal{H}_0). \quad (14)$$

Similarly, for the stopping power and mean square energy loss per unit path length, we have from Eq. (4) and Eq. (5)

$$S_i(E) = \int_{E_i^B}^{(E+E_i^B)/2} d(\mathcal{H}_0) \mathcal{H}_0 \tau_i^{\text{exc}}(E, \mathcal{H}_0) \quad (15).$$
and

$$\omega_i^2(E) = \int_{E_i^B}^{(E+E_i^B)/2} \frac{d(\Omega_{\omega})}{(\Omega_{\omega})^2} \tau_i^{\text{exc}}(E,\Omega_{\omega}).$$  \hfill (16)

For the remaining calculations we form the sums

$$S_{\text{exc}}(E) = \sum_i S_i(E)$$  \hfill (17)

and

$$\omega_{\text{exc}}^2(E) = \sum_i \omega_i^2(E).$$  \hfill (18)

where the index $i$ includes the terms appropriate for a given solid, including exchange corrections as indicated above. The csda range is calculated from

$$R_0(E) = \int_{10 \text{eV}}^{E} \frac{dE'}{S_{\text{exc}}(E')}$$  \hfill (19)

corresponding to an electron slowing down in a continuous manner from an energy $E$ to $10 \text{eV}$. The mean square fluctuation in the csda range based on Eq. (7) is calculated as

$$\langle (R-R_0)^2 \rangle_{\text{AV}} = \int_{10 \text{eV}}^{E} \frac{dE'}{S_{\text{exc}}(E')} \frac{\omega_{\text{exc}}^2(E')}{[S_{\text{exc}}(E')]^3}. \hfill (20)$$
VI. RESULTS

Before presenting the tabulations for polystyrene, we discuss briefly the results for IMFP and stopping power. In Figure 6 the IMFP's are shown for interactions with the valence band (sum of contributions from the three levels, Section III) and with the carbon K shell. The K-shell contribution is quite small compared to the valence band contribution and amounts to \( \approx 1\% \) of the total IMFP in the energy range covered here. We have found no measurements of electron mean free paths in polystyrene for \( E \leq 10 \text{ keV} \). However, Swanson and Powell\textsuperscript{14} performed characteristic energy loss measurements using 20 keV electrons on polystyrene films and determined mean free paths for the 7 eV and 21 eV losses. These losses correspond to the peaks seen in the energy loss function derived from optical data shown in Figure 2. They determine a mean free path \( \lambda \), in Å, of 17,400 ± 5,500 for the 7 eV loss and 410 ± 80 for the 21 eV loss. If we assume these represent the dominant inelastic loss processes, then the total mean free path (\( 1/\lambda_{\text{TOTAL}} = 1/\lambda_{7\text{eV}} + 1/\lambda_{21\text{eV}} \)) is 400 Å (± 20%) at 20 keV. Extrapolating our IMFP values to 20 keV yields \( \lambda \approx 2.5 \times 10^{-3} \text{ Å}^{-1} \) or \( \lambda = 1/\mu = 400 \text{ Å} \) in excellent agreement, possibly fortuitous, with the experimental result.

In Figure 7 we show the contributions to the stopping power of polystyrene for electrons. Also shown is the stopping power derived from Bethe-Bloch theory\textsuperscript{1,2} for \( E \geq 10 \text{ keV} \). At 10 keV, the Bethe-Bloch result is \( S = 0.237 \text{ eV/Å} \) for polystyrene of density \( \rho = 1.05 \text{ g/cm}^3 \). Our value for \( S = S \) (valence) + \( S \) (K-shell) is \( S = 0.238 \text{ eV/Å} \) which
Fig. 6. IMFP for inelastic interaction of an electron of energy \( E \) with the valence-band electrons and K-shell electrons in polystyrene.

Fig. 7. Contributions from valence-band electrons and K-shell electrons to the stopping power of polystyrene for an electron of energy \( E \).
agrees remarkably well with the previously tabulated results\textsuperscript{1,2} at this energy. Our tabulated results for $\mu$, $S$, range, and straggling are presented in Section VIII.
VII. REFERENCES


VIII. TABLES

The results of the calculations for polystyrene, at a density of 1.05 g/cm$^3$, are given in the following tables:

Table 1 presents total inverse mean free path and contributions to IMFP due to interactions with valence band electrons or carbon K-shell electrons, Eq. (14), in units of $\text{Å}^{-1}$ for incident electrons with energies $5 \text{ eV} \leq E \leq 10 \text{ keV}$.

Table 2 presents total stopping power and contributions to the stopping power due to interactions with valence band electrons or carbon K-shell electrons, Eq. (15), in units of $\text{eV}/\text{Å}$ for electron energies $5 \text{ eV} \leq E \leq 10 \text{ keV}$.

Table 3 presents the csda range, Eq. (19), in units of $\text{Å}$, the mean square energy loss, Eq. (16), in $(\text{eV})^2/\text{Å}$, the mean square range fluctuation, Eq. (20), in $\text{Å}^2$, and the relative range straggling given by $\left[\frac{(R-R_0)_{AV}^2}{R_0}\right]^{\frac{1}{2}}$, for electron energies $15 \text{ eV} \leq E \leq 10 \text{ keV}$.
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<th>VALENCE BAND</th>
<th>INNER SHELL (C-K SHELL)</th>
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