Total cross sections for electron scattering by 1-propanol at impact energies in the range 40-500 eV

Citation: The Journal of Chemical Physics 147, 194307 (2017); doi: 10.1063/1.5008621
View online: https://doi.org/10.1063/1.5008621
View Table of Contents: http://aip.scitation.org/toc/jcp/147/19
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Total cross sections for electron scattering by 1-propanol at impact energies in the range 40-500 eV

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(Received 8 October 2017; accepted 1 November 2017; published online 21 November 2017)

I. INTRODUCTION

Precise values of the Total Cross Section (TCS) for electron scattering by primary alcohols such as methanol, ethanol, propanol, and butanol are very important in many applied areas, given that the TCS reflects the reactivity of a molecular target in the collision processes as a function of the electron energy. These values are very useful, for instance, when modeling alcohol combustion through a spark ignition process within an internal combustion engine. Through this modeling, we can improve the performance of motor vehicles and all technologies currently utilizing fossil fuels, when using alternative fuels sustainably produced from renewable sources.¹ Nowadays, ethanol is the best known and most used alternative fuel, used as a flex fuel in modern vehicles as well as being added in small amounts to many gasolines in order to oxygenate the fuel and to reduce the air pollution through the lower emissions of greenhouse gases when compared to fossil fuel combustion.²

Ethanol has a higher octane number than gasoline (99.5 AKI, anti-knock octane rating, for ethanol and an average of 85-96 AKI for gasoline), and so it represents a high standard quality fuel.³ A higher octane number fuel resists the higher pressure and temperature in compression ignition engines such that the air-fuel blend burns slower and allows the motor to reach optimum performance. The use of lower octane fuel, however, may produce a spontaneous combustion, leading to an undesirable auto ignition of the fuel before the spark is emitted by the plug, thereby resulting in an inefficient performance of the motor. The situation of ethanol versus gasoline as the fuel of choice is, however, complicated by its lower energy density (~20 MJ/l) compared to that of gasoline (~33 MJ/l). This property effectively mitigates the advantage of ethanol’s higher octane values, so that the net result entails in a loss of fuel economy. All these considerations bring about the question for the possibility of efficiently using larger alcohols, with a longer carbon chain, as fuel. One clear example for this role is propanol which has an octane rating of 108 AKI and an energy density of 24 MJ/l).³ Currently, the main problem of using propanol as a fuel is that it is difficult and expensive to produce on a large scale. However, better knowledge of its properties and mastering new technologies for its application as a fuel could justify the intensive research to find an effective methodology for its production in large scale at lower cost.⁴

There has been a significant volume of study for electron scattering from methanol and ethanol, as was noted in Ref. 5 and references therein, but not for propanol. Experimental and theoretical investigations for methanol, including total cross sections,⁶–¹³ elastic differential cross sections,¹³–¹⁷ from which integral cross sections and momentum transfer cross sections can also be derived, electronic excitation¹⁸ providing a pathway to neutral dissociation, as well as experimental partial ionization cross sections (PICS) and total ionization cross sections (TICS)¹⁹–²² and corresponding theoretical results.⁵,²³–²⁵ Less information is available for electron interactions with ethanol, including experimental and theoretical data for total⁶,¹⁰ and elastic differential, integral, and momentum transfer cross sections¹¹,¹⁴,¹⁵ and also total ionization cross sections.⁵,¹⁹,²¹,²² All these results were recently considered

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by Brunger,17 who looked at the available database in the context of simulating charged-particle transport in those systems.

There are relatively just a few studies about electron interactions with 1-propanol. 19,21,22,25–30 Khakoo et al.26 reported the only experimental and calculated differential cross sections for elastic scattering, for selected impact energies ranging from 1 to 100 eV, that are available in the literature. The fragmentation of 1-propanol is a little more studied; Hudson et al.22 reported experimental and calculated absolute TICSs for electron impact energies ranging from 16 to 200 eV; Bull et al.28 reported theoretical absolute TICS data and polarizability parameters, while Takeuchi et al.29 investigated the fragmentation mechanisms for energies in the range 8–25 eV and also generated potential energy curves calculated using ab initio molecular orbital methods. Rejoub et al.21 measured absolute PICS for 1-propanol from threshold to 1000 eV, for groups of cations with similar mass and determined from these data the TICS. We also recently published a study of electron impact ionization and ionic fragmentation of 1-propanol, using mass spectroscopy, where we measured for the first time the PICS for individual ionic fragments, over the energy range of 10–100 eV.30

Tanaka et al.31 recently discussed why it is necessary to have complete and comprehensive cross section databases, in order to accurately model or simulate the behavior of collision driven processes found across a wide variety of important applications. Those applications include atmospheric phenomena,32,33 low temperature plasma,34 and charged-particle track behavior.35–38 The present investigation is an extension of our earlier studies on electron scattering from the primary alcohols,5,10 where we have produced new TCS data for the interaction of electrons with 1-propanol in the energy range from 40 to 500 eV. As far as we know, these data are reported for the first time in the literature. We have also calculated theoretical values of these TCS using the Independent-Atom Model with Screening Corrected Additivity Rule and Interference (IAM-SCAR+I) effects.39,40 All this work fits squarely within our aim of contributing to the 1-propanol database for auto ignition of fuel simulations.

The remainder of this paper is organized as follows. In Sec. II, we present the experimental details and discuss the configuration of our apparatus, the linear attenuation technique, including a discussion on our uncertainties, and also the IAM-SCAR+I theoretical method whose results are used to compare with our measured TCS data. Section III briefly details our TCS results and a discussion of those results, while finally in Sec. IV we provide some conclusions from this investigation.

II. EXPERIMENTAL AND THEORY DETAILS

We have developed a new TCS apparatus in the Physics Department of Juiz de Fora Federal University, Brazil, which, in principle, covers the impact energy region 5-500 eV. This equipment is an enhanced version of an earlier apparatus.10 Nonetheless, this new apparatus employs essentially the same techniques and methodology as the old equipment, consisting of an electron gun, a gas cell, an electron energy analyser composed of an array of decelerating electrostatic lenses, a cylindrical dispersive 127° analyser (CDA 127°), and a Faraday cup, as shown schematically in Fig. 1. The apparatus is housed inside a vacuum chamber covered by 5 layers of mu-metal, for shielding against spurious magnetic fields, and differentially pumped through two turbomolecular pumps; one with a pumping speed of 80 l/s, for pumping the electron gun chamber in order to avoid changes in the electron back-streaming.

This new apparatus utilizes an in-house designed electron gun and its associated control electronics, covering the energy range from 5 to 500 eV. The performance of this new gun was excellent compared to that of the gun used in the old apparatus, mainly in the energy region below 70 eV, with very good conditions of focus and collimation in a reasonable range of length (40 mm) being achieved. This characteristic is a fundamental one for the successful application of the linear transmission technique.10 While in the old apparatus, the intensity of the 60 eV electron beam was around 0.05 nA, the new gun routinely provides beams of 200 nA intensity, for that same impact energy. The present apparatus also contains a new scattering cell, consisting of a 40.00 mm long tube bounded by two apertures of diameters 2.00 mm. The collimated electron beam passes through the scattering cell, and those electrons that traverse the cell are discriminated with the electrostatic lens and the CDA 127°, which prevents electrons inelastically scattered in the forward direction being collected by the Faraday cup. The angular acceptance of the analyzer, defined by the solid angle formed by the analyzer aperture as seen from the center of the scattering cell, is $6.4 \times 10^{-4}$ sr. The temperature of the sample is measured using a mercury thermometer, with an accuracy of 0.1 °C, fixed on the external wall of the vacuum chamber, given that it was found that the temperature of the gas sample in the cell is the same as that of the vacuum chamber.10 The gas is introduced into the cell using a leak valve, while its pressure $P$ is monitored by a MKS Baratron connected by a tube to the gas cell.

All measurements were performed for an incident beam current of around $10^{-10}$ A, where no dependence of the TCS...
upon current was found. The TCS, $\sigma(E)$, obtained through the linear attenuation technique, based on the measurement of the attenuation of the collimated electron beam at a given energy through a gas cell containing the gaseous target to be studied at a given pressure and temperature, is given by the Beer–Lambert law,

$$I(E) = I_0(E) e^{-n L \sigma(E)},$$

(1)

where $I_0(E)$ and $I(E)$ are the intensity of the electron beam before and after the gas cell, respectively, $n$ is the average particle number density of the target gas, and $L$ is the path length of the electron beam through the gas cell. The TCS data, $\sigma(E)$, are in practice determined by measuring the transmitted intensities with and without gas in the cell; $n$ is taken to be equal to $P/kT$, using the ideal gas law, where $P$ is the pressure of the target measured with Baratron in a molecular flow regime, $k$ is the Boltzmann constant, and $T$ is the absolute gas temperature (i.e., in K). The pressure range of the target in the scattering cell was maintained between 1 and 4 mTorr, so that double scattering was negligible. The variation of $\ln(I/I_0)$ with the pressure $P$ is a straight line, whose slope is proportional to the TCS. Our measurements were carried out for each incident electron energy in at least five runs, each run taking at least seven pairs of values of $P$, $I_0$, and $I$. An averaging procedure was subsequently applied to determine the total cross section at a particular energy.

The electron energy resolution in all our measurements was typically 0.7 eV (FWHM). The uncertainties on the TCS data were estimated by taking into account the statistical uncertainty, estimated at around 5.7%; the fluctuations of the primary electron beam of $\sim 0.5\%$; the accuracy of the pressure reading from MKS Baratron of $\sim 2.0\%$; and knowledge of the effective scattering length of the gas cell of 0.3\%. The primary electron beam current at any energy was always stable to better than 1.5\%, while the stability of the flow of the gas entering the gas cell varied by less than 1\%. The temperature of gas in the cell was measured to an accuracy of 0.5\%. Considering all these factors, the root sum square of the overall uncertainties was a total of around 9.4\%. The bona fides of our new apparatus were tested in the energy range from 14 eV to 500 eV using argon as a reference target. The results of our argon TCS measurements are shown in Fig. 2, which demonstrates the reliability of our technique and methodology when compared with other TCS from the literature.

Propanol vapor was obtained from the volatile propanol liquid, sourced from Sigma-Aldrich, with a stated purity of 99%. In addition, we degassed the sample using at least six freeze–pump–thaw cycles before it was admitted into the chamber using a needle valve (Granville–Phillips-203 variable leak). The vapor pressure of 1-propanol in its storage container was calculated to be 17.39 Torr, using the Antoine equation, where the constants employed were $A = 5.313 \, 84$, $B = 1690.864$, and $C = -51.804$. The propanol liquid was housed in a glass flask, vacuum coupled to the relevant leak valve by a glass-to-metal seal, with a 1/4 in. covar tube being used to send the vapor into the gas feed line. Although we expect some condensation of propanol in our gas lines, gas cell, and on other elements of the spectrometer, the experiment was conducted without any baking and consequently conducted at the laboratory temperature of around 22 °C as maintained using conditioned air. This decision not to bake, however, meant that we observed that after a data acquisition period of approximately 6 h, condensation of propanol in our experiment caused the electron beam to become somewhat unstable. A waiting period, $\sim 12$ h of pumping, was then necessary to remove the condensed propanol in order to continue the measurements.

Finally, we note that the present results are not corrected for the so-called “forward angle scattering effect.” However, this effect, which results from very forward elastically scattered electrons not being discriminated against in our detector system, is expected to be small in this case. This follows as such corrections are known to be typically smaller at the intermediate energies of this work, even for strongly polar molecules, and given the very good angular resolution of our spectrometer.

The IAM-SCAR+I method has been described many times in the past, most recently by Traoré Dubuis et al., and so only a brief précis is needed here. The fundamental premise of this method is that the molecule of interest is not considered as a single multi-center target but as an aggregate of individual atoms. As a consequence, this approximation effectively assumes that the molecular binding does not affect the electronic distribution of the atom, so that each atom scatters independently from the others. Put simply, the atoms of the molecule are viewed as isolated entities. Thus, the initial task in any IAM-SCAR computation is to describe the electron scattering from the atoms constituting the molecular target (here carbon, hydrogen, and oxygen). The electron–atom interaction is represented by an ab initio complex optical potential, where the real part accounts for elastic scattering, while the imaginary part represents the inelastic processes. Note that those inelastic processes are considered as “absorption”
channels from the incident beam. The complex potential for each atom in the molecule is represented as

\[ V_{\text{opt}}(r) = V_R(r) + iV_{\text{abs}}(r) = V_S(r) + V_{\text{ex}}(r) + V_{\text{pol}}(r) + iV_{\text{abs}}(r). \]  

(2)

In Eq. (2), the real part \( (R) \) consists of three terms: (i) a static term \( (V_S) \) derived from a Hartree–Fock calculation of the atomic charge distributions,\(^{52}\) (ii) an exchange term \( (V_{\text{ex}}) \) which accounts for the indistinguishability of the incident and target electrons,\(^{53}\) and (iii) a polarization potential \( (V_{\text{pol}}) \) that describes the long-range interactions.\(^{54}\) Finally, the absorption potential \( (V_{\text{abs}}) \) is based on the quasi-free model developed by Staszewska et al.\(^{55}\) Initially, quite significant discrepancies between experimental data and the optical potential model were found, although these were subsequently corrected.\(^{56,57}\)

With those changes, the Spanish team have developed an approach capable of representing electron-atom scattering over a broad range of energies and targets (e.g., Ref. 58).

The electron-molecule cross sections are now computed from the atomic data by the additivity rule (AR).\(^{59}\) Within that approach, the molecular scattering amplitude is derived from the sum of all the relevant atomic amplitudes, including the phase coefficients, from which the molecular differential cross sections (DCSs) can be generated. Integral cross sections (ICSs) are then derived by integrating those DCS. The TCSs are finally determined from the sum of the elastic ICS from the atomic data by the additivity rule (AR), which effectively “see” the target molecule as a sum of individual atoms. For lower incident energies, the atomic cross sections are sufficiently large to overlap with one another, leading to an overestimation of the AR molecular cross sections. To solve that limitation, at least in part, Blanco and Garcia\(^{60,61}\) developed the SCAR code, which takes into account the geometric features of the molecule by introducing some screening coefficients. More recently, they incorporated some interference (I) term corrections.\(^{40}\) Within these SCAR and SCAR+I approaches, the range of validity of the model might be extended down in energies of 20–30 eV. Certainly there is some evidence to suggest that either the IAM-SCAR or IAM-SCAR+I approaches are quite powerful tools for calculating cross sections at intermediate to high energies.\(^{62–64}\)

**III. RESULTS AND DISCUSSION**

The present experimental results of TCS for electron scattering by 1-propanol in the energy range from 40 to 500 eV are shown in Fig. 3 and numerically displayed in Table I, where they are compared with data from our IAM-SCAR+I method calculated for the energy range from 1 to 500 eV. It is clear from Fig. 3 that a very good overall level of accord is found between our measured and calculated TCS, across the entire common energy range. It is also possible to derive an estimate of the TCS by summing the elastic ICS of Khakoo et al.\(^{26}\) and the TICS of Pires et al.,\(^{30}\) at a given electron impact energy. Those data are also plotted in Fig. 3 and listed in Table I and

**FIG. 3.** Absolute total cross section (TCS) for electron scattering by 1-propanol covering the energy range 40-500 eV. The error bars correspond to the one standard deviation absolute uncertainties. See legend on figure and text for further details. Note that (filled red squares) present data; (filled blue circles) “Hybrid” TCS.

**TABLE I.** Absolute TCS for electron scattering on 1-propanol. The total errors obtained as described in the article are given in absolute numbers. “Hybrid” denotes the TCS derived as the sum of the elastic ICS from the study of Khakoo et al.\(^{26}\) and the recent TICS from the study of Pires et al.\(^{30}\)

<table>
<thead>
<tr>
<th>Energy (eV)</th>
<th>Present data ((10^{-16} \text{ cm}^2))</th>
<th>IAM-SCAR+I ((10^{-16} \text{ cm}^2))</th>
<th>“Hybrid” TCS ((10^{-16} \text{ cm}^2))</th>
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<tr>
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<tr>
<td>400</td>
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as shown by Pires et al.,\textsuperscript{30} in the range 10–100 eV agreement between the study of Pires et al. and Rejoub et al. TICS, where a direct comparison is possible, is excellent so that the choice of TICS to generate our “hybrid” TCS does not impact upon the discussion that follows. Further note that technical issues, in respect to the PICS and TICS of Pires et al.,\textsuperscript{30} such as the performance of their mass spectrometer in dealing with cationic fragments of differing kinetic energy release, are addressed in detail in the study of Nixon et al.\textsuperscript{5} to whom the interested reader is referred.” Typically the “Hybrid” TCS is somewhat lower in magnitude than what we found in the present measurements and IAM-SCAR+I calculations. This observation is, however, not particularly surprising for the following reasons. First, the TICS of Pires et al.\textsuperscript{30} account for \textasciitilde97\% of the cations generated by electron impact and so slightly underestimates its true value. Second, the “Hybrid” TCS does not account for all the open channels at energies above 30 eV. In particular, the cross sections for vibrational excitation, discrete electronic-state excitation, dissociative electron attachment, and neutral dissociation are in principle omitted. While it might be argued that the ICSs for those processes are likely to be small at intermediate energies,\textsuperscript{65–67} their absence does contribute to why the “Hybrid” TCS is lower in magnitude compared to that which we find from our direct measurements. Finally, we note that the elastic ICS of Khakoo et al.\textsuperscript{26} was derived from their DCS that were measured over the angular range 10°–130°. This means that an extrapolation of those DCS to 0° and 180° was required before the elastic ICS could be obtained.\textsuperscript{26} Such an extrapolation can well be problematic and might have caused the elastic ICS to be somewhat underestimated, leading to a “Hybrid” TCS that is too low in magnitude (as seen in Fig. 3).

It is well known\textsuperscript{68–70} that the electronic properties of a molecule, such as its permanent dipole moment and dipole polarizability, affect the scattering dynamics and therefore the scattering cross sections. In this case, 1-propanol has a reasonably large dipole moment of 1.55 D\textsuperscript{71} and a quite significant dipole polarizability of 6.67 Å\textsuperscript{3}\textsuperscript{71} with the observed behavior of the TCS as a function of energy, namely, the TCS increasing quite a lot in magnitude as the incident electron energy is lowered, being entirely consistent with those values of its target electronic properties. It would certainly be interesting to extend the present measurements to lower energies, to both check that this observed energy dependence persists in going to those lower energies and to investigate if any shape resonances are observed. However, we should note here that the average rotational excitation energy of 1-propanol is of the order of a few meV, much lower than the present energy resolution (0.7 eV) and therefore any electron reaching the analyzer after a rotational excitation collision would be considered as “unscattered.” Nonetheless, a simple rotational excitation calculation, based on the Born approximation, as that described in Ref. 50, showed that the possible contribution of this effect to the present experimental results ranges from 2.9\% at 500 eV up to 8.7\% at 40 eV.

Finally, we note that the very good level of agreement we find between our measured and calculated intermediate energy TCS gives us confidence that these data could be incorporated into a database like LXCat,\textsuperscript{72} along with other relevant cross sections, in order to model the behavior of 1-propanol under low temperature plasma conditions.

\section{Conclusions}

We presented new measurements and IAM-SCAR+I calculations of TCSs for electron scattering by 1-propanol in the intermediate and low electron energy ranges. Propanol was studied in the region of impact energy region of 40–500 eV experimentally, and from 1 to 500 eV theoretically. Our measured TCS data for 1-propanol are found to be in very good agreement with our theoretical data. This suggests, at least for intermediate energies, that our results might be gainfully employed in a cross section database for modeling studies. Unfortunately, at this time, there appeared to be no other experimental or theoretical data against which we could compare our results. This is a situation that we believe should be addressed by colleagues in the future.

Total cross sections have over the years been somewhat undervalued,\textsuperscript{73} as in the so-called hierarchy of cross sections (so named in terms of how much physical information they provide about the scattering dynamics in question), they sit at the bottom of the table. Nonetheless, even though they do not discriminate between the various open scattering channels at a given energy and nor do they provide any angular information, they still fulfill several vital roles. Namely, they represent an upper limit for the sum of the various ICSs for all processes open at a given energy and thus represent an important self-consistency check for scientists in the modeling and simulation communities whom are constructing databases. In addition and perhaps this is not so well appreciated in the scattering community, they provide a direct link to quantities like the stopping power [see Eq. (19) of Ref. 74] which are fundamental in describing charge-particle track behavior in atoms and molecules. As a consequence, we believe that measurements and calculations of TCSs, such as we have presented here, remain topical and important in many areas of science and in understanding and characterising various applications including technology.

\section*{Acknowledgments}

M.C.A.L. acknowledges financial support from Brazilian Conselho Nacional de Desenvolvimento Científico e Tecnológico (CNPq), Fundação de Amparo à Pesquisa do Estado de Minas Gerais (FAPEMIG), and FINEP, while M.J.B. thanks CNPq for his “Special Visiting Professor” award. Some financial assistance from the Australian Research Council through Grant No. DP160102787 is also noted. Finally, S. Ghosh acknowledges his grant for his project Grant No. FIS 2016-80440 and the EU Project No. FP7–ITN–ARGENT-608163.

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