Evaluation of Cross Section for Electron Impact with hydrogen and Their Combination Molecules in Fusion Plasma

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Data Center for Plasma Properties

Supported by
- Ministry of Science and Technology
- Ministry of Industry
- Ministry of Information and Technology

Work with
- SAMSUNG, Hyundai, LG
- 3 National Institute
- 4 National University
- Japan, Australia, India, Russia, Germany

Vienna, Austria

Daejeon, KOREA
What we are doing?

DCPP: 12 man/year
Outside: 15 man/year

- Sputter Simulator
- ICP/CCP Source Simulator
- Plasma Simulator
- Surface DB
- Feature Profile Sim/Exp
- Gas Chemistry DB
- BEB model Ionization/Excitation
- Data Validation
- SWARM analysis
- Virtual Laboratory
- Web based DB Service
- Web based Simulation
- Ion-Beam
- Positron Beam
- e-Impact Total CS -ELECS1-
Why we need data evaluation?

- Commercial simulation code: CFD-ACE+
- Using C4F8 data instead of C4F6
- Not evaluated Ar data

![Graph showing etch rate vs bias power for C4F6/Ar 10/50 sccm at 20mTorr and 30mTorr.]

Simulation result for 20mTorr
Simulation result for 30mTorr
Why we need data evaluation?

- Using same commercial code: CFD-ACE+

- Update C4F6 data (NOT C4F8)
- Evaluated Ar data
We need evaluated Data more than Data itself….

But HOW??
Following the classical way

Atomic and Molecular Physics Society

- Collection of data without quality judgment is not useful
  - only BETTER data should replace existing ones
- Quality judgment (=critical evaluation) of data is crucial for users
- Evaluation must be done by scientists with appropriate experience
- Critical evaluation requires a wide network of competent and dedicated scientists
  - This is the most expensive part of maintaining a database

Three data evaluation principles

- How well is the data generation described
- How do the data follow the known physical laws
- How do the data compare to other measurements or calculations of the same phenomena
Making Data Evaluation System

Supported by National Standard Reference Data (SRD) Project
Data Evaluation

Certified Data with Uncertainty
\rightarrow NSRD (National Standard Reference Data)

Certified by Ministry of Industry
Standard Reference Data

Definition

“Reference Data”
→ is data related to a property of a phenomenon, body, or substance, or to a system of components of known composition or structure, obtained from identified source, critically evaluated, and verified for accuracy

“Standard Reference Data”
→ is reference data issued by a recognized authority

The SRD is classified into three types according to the criteria of technical evaluation which are provided by the governmental gazette regarding ‘the Guidelines for the Establishment and Distribution of Standard Reference Data’
Standard Reference Data

[Classification of SRD]

**Qualified SRD**
- Data which satisfies **minimum technical requirements** of SRD
  
  Minimum technical requirements include clear description of measurand; rationale of measurement methods and theoretical calculation; uncertainty estimate; traceability of measuring equipment to the national measurement standards; control of variables which may impact on measurement results; description of measurement errors and its accuracy; and description of measurement methods and procedures for reproducibility by the third parties.

**Validated SRD**
- SRD derived from the qualified SRD which has consistency with the measurement results obtained by other methods.

**Certified SRD**
- SRD which is critically reviewed by relevant scientists for its reliability and accuracy.
Uncertainty??
Uncertainty
Uncertainty - ISO-GUM

ISO (International Organization for Standardization) : GUM (Guide to the Expression of Uncertainty in Measurement.)

- published by the International Organization for Standardization in 1993 in the name of 7 international organizations

<table>
<thead>
<tr>
<th>Stated Purpose</th>
</tr>
</thead>
<tbody>
<tr>
<td>Promote full information on how uncertainty statements are arrived at</td>
</tr>
<tr>
<td>Provide a basis for the international comparison of measurement results</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Benefits</th>
</tr>
</thead>
<tbody>
<tr>
<td>Much flexibility in the guidance</td>
</tr>
<tr>
<td>Provides a conceptual framework for evaluating and expressing uncertainty</td>
</tr>
<tr>
<td>Promotes the use of standard terminology and notation</td>
</tr>
</tbody>
</table>

Point

All of us can speak and write the same language when we discuss uncertainty
Uncertainty - ISO-GUM

<table>
<thead>
<tr>
<th>Period 1</th>
<th>~1984</th>
</tr>
</thead>
<tbody>
<tr>
<td>▪ A measure of the <strong>possible error</strong> in the estimated value of the measurand as provided by the result of a measurement</td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Period 2</th>
<th>1984 ~ 1992</th>
</tr>
</thead>
<tbody>
<tr>
<td>▪ An estimate characterizing the <strong>range of values</strong> with which the true value of a measurand lies</td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Period 3</th>
<th>1992 ~ 2007</th>
</tr>
</thead>
<tbody>
<tr>
<td>▪ Parameter associated with the result of a measurement, that characterizes the <strong>dispersion of the values</strong> that could reasonably be attributed to the measurand</td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Period 4</th>
<th>2007 ~ present</th>
</tr>
</thead>
<tbody>
<tr>
<td>▪ Parameter that characterizes the <strong>dispersion of the quantity value</strong> that are being attributed to a measurand, based on the information used.</td>
<td></td>
</tr>
</tbody>
</table>

In any measurement, the **measurand** is defined as the “particular quantity subject to measurement”
Methods for Uncertainty Evaluation

The GUM classifies methods of uncertainty evaluation (for input estimates) as either **Type A** or **Type B**

**Type A**: method of evaluation by statistical analysis of series of observations

- Let \( x_i \) be the arithmetic mean and let \( u(x_i) \) be the experimental standard deviation of the mean (the “standard error” of the mean)
- Least-squares regression can also be a Type A method
- If there is a well-defined number of “degrees of freedom” (number of observations minus number of parameters estimated), it’s probably a Type A method of evaluation

**Type B**: method of evaluation by any means other than statistical analysis of series of observations

- Often a Type B evaluation involves estimating a bound, \( a \), for the largest possible error in the estimate, \( x_i \), and dividing by an appropriate constant based on an assumed distribution for the error
- For example, if you believe the true value lies within \( \pm a \) of the estimated value, \( x_i \), but you know nothing more than that, assume a rectangular distribution, and divide \( a \) by \( \sqrt{3} \) to obtain \( u(x_i) \)
example

Modeling the measurement \( y = f(x_1, x_2, \ldots, x_n) \)

Identifying uncertainty components for each input quantity

Evaluating standard uncertainty Type A, Type B

Combining standard uncertainties of input quantities

\( u = \frac{s}{\sqrt{n}} \)

Sensitivity coefficient \( c_i = \frac{\partial f}{\partial x_i} \)

Expanded uncertainty

\( U = k u_c(y) \)

\( u(x_i) \left\{ u(x_{i,1}), u(x_{i,2}), u(x_{i,3}), \ldots \right\} \)

\( u \approx s \) or \( \frac{s}{\sqrt{n}} \)

Coverage factor \( k = 2 \)
Methods for Uncertainty Evaluation

Type A: method of evaluation by statistical analysis of series of observations

\[ x_1, x_2, x_3 \ldots \ldots x_n \] (independent value)

\[ \bar{x} = \frac{x_1 + x_2 + \ldots + x_n}{n} \] (average)

\[ s(x) = \sqrt{\sum_{i=1}^{n} \frac{(x_i - \bar{x})^2}{n-1}} \] (standard deviation)

\[ s_p^2(x) = \frac{\sum_{i=1}^{n} v_i s_i^2}{\sum_{i=1}^{n} v_i} \] (combined standard deviation, \( v_i \) : degree of freedom)

\[ u_s = \frac{a}{\sqrt{3}} \] (systematic effects)

\[ u_m = \sqrt{\frac{s_p^2(x)}{n} + u_s} \] (combined standard uncertainty)

\[ U = k \times u_m \] (additional uncertainty)
Standard Reference Data Evaluation Process

[Low data evaluation]
- Start Articles, Reports
  - reject
  - Definition
  - Method
  - Procedure
  - Traceability
  - Uncertainty
  - Comparison
  - Evaluated Journal
    - Repeatability
      - Valid Data
      - Reference Data
    - Reproducibility
      - Valid Data
  - Valid Data

[Data comparison]
- Valid Data
  - Valid Data
  - Graph TEST
  - Accuracy TEST
  - Consistency TEST
    - Consistency TEST
      - Candidate DATA
      - Uncertainty TEST
        - small
        - Dispersion
        - Big
        - DATA analysis
          - Best Value
          - Best Value
          - DATA Fitting
            - Candidate DATA

[Expert TEST]
- Candidate DATA
  - Candidate DATA
  - Validated DATA
  - Certified DATA
  - Expert Evaluation
    - Consistency
Evaluation of Cross Section for Electron Impact with Hydrogen and Their Combination Molecules in Fusion Plasma
(1) Cross sections for electron collisions with hydrogen molecules


(2) Electron impact cross sections for deuterated hydrogen and deuterium molecules

Report on Progress in Physics, 73, 116401 (2010)
(1) Cross sections for electron collisions with hydrogen molecules


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FIG. 20. Summary of the cross sections for electron collisions with $\text{H}_2$. 
After reviewing available cross section data, we have determined a set of recommended values of cross section, as far as possible. The general criteria for the selection of preferred data are as follows:

(i) In principle, experimental data are preferred to theoretical ones.

(ii) The reliability of the experimental methods employed is critically assessed. Agreement between independent measurements of the same cross section is generally taken as an endorsement of the accuracy of the measured data. A strong emphasis is placed on the consistency of the results determined by different techniques.

(iii) In cases where only a single set of data is available for a given cross section, those data are simply shown here (i.e., not designated as recommended), unless there is a strong reason to reject them. Even when multiple sets of data are available, no recommendation is made if there is a significant disagreement among them or they are fragmentary (i.e., only a few data points being reported).

In this way, the present paper aims to provide a more complete data set for electron collisions with H$_2$ than those published before. A survey of literature has been made
Further studies are still needed to make the cross section data more comprehensive and more accurate.

Following problems should be addressed:

1. The rotational cross section has a large value at the energies up to a few tens of eV. However, no experimental data are available at the energies of about 1 eV and higher.
2. Only one old set of measured values is available for the elastic cross sections at the energies of 100-1,000 eV. They are not completely consistent with the recommended cross sections in the energy region below 100 eV.
3. Although several sets of experimental cross sections are available for the excitation of electronic states, more detailed measurement should be done for the excitation of triplet states.
4. The total dissociation cross sections are available with fair certainty. Further information is necessary for the details of the dissociation process.
5. Finally, cross section data for the target molecules in their excited states are of practical importance. Any experiment of the electron collision with excited hydrogen molecules would be very valuable.
(2) Electron impact cross sections for deuterated hydrogen and deuterium molecules

Report on Progress in Physics, 73, 116401 (2010)

Abstract
The demand for electron-impact cross sections has increased tremendously in recent years.
There is, however, a special interest in such cross sections for hydrogen molecules and its isotopomers, HD and D₂, because of their presence in tokamak edge plasmas, planetary atmospheres and at different astrophysical sites. This explains the need for having well validated sets of electron-impact cross sections for different processes. This work reviews the electron-scattering cross sections for elastic and inelastic processes at different electron energies for both these molecules. The elastic momentum transfer cross sections and inelastic cross sections for electron-impact rotational, vibrational and electronic excitation, emission, dissociation, ionization and dissociative electron attachment have been evaluated and well validated in this work wherever and whenever possible.

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Table 1. Molecular constants of H₂, HD and D₂ molecules.

<table>
<thead>
<tr>
<th>Molecule</th>
<th>H₂</th>
<th>HD</th>
<th>D₂</th>
</tr>
</thead>
<tbody>
<tr>
<td>Reduced mass (u)</td>
<td>0.504</td>
<td>0.672</td>
<td>1.067</td>
</tr>
<tr>
<td>Equilibrium internuclear distance (Å)</td>
<td>0.7414</td>
<td>0.7414</td>
<td>0.7415</td>
</tr>
<tr>
<td>Ionization potential (eV)</td>
<td>15.4259</td>
<td>15.445</td>
<td>15.467</td>
</tr>
<tr>
<td>Polarizability (10⁻²⁵ cm³)</td>
<td>0.8023</td>
<td>0.7975</td>
<td>0.7921</td>
</tr>
<tr>
<td>Vibration frequency (cm⁻¹)</td>
<td>401.21</td>
<td>381.3</td>
<td>311.5</td>
</tr>
<tr>
<td>Vibrational energy (eV)</td>
<td>0.5456</td>
<td>0.7273</td>
<td>0.38628</td>
</tr>
<tr>
<td>Dissociation energy</td>
<td>4.478</td>
<td>4.414</td>
<td>4.556</td>
</tr>
</tbody>
</table>

molecules if more than adequate data, both experimental as well as theoretical, have been reported over the years. Problems only arise when the data available are sufficiently inadequate. That is why selecting well-validated data for D₂ and HD molecules has become an uphill task. Irrespective of data being adequate or inadequate, the general criteria for selection remain, more or less, the same though the special criteria may change from process to process. Some of the points leading to general criteria are discussed below.

(i) It is difficult and tedious to obtain absolute measurements of electron-impact cross sections for a certain process; but instead relative measurements can be obtained as a function of the incident impact energy with greater ease. These measurements are, sometimes, normalized at one or two energy points using other standard data from some other experiment. The relative values thus made absolute at one/two energies would be good only at these energies and there would, invariably, be a mismatch of cross sections at other electron energies. Such data should be avoided and not be considered for evaluation.

(ii) In some experiments, to achieve better energy resolution and a cleaner vacuum environment, one has to sacrifice some other characteristic. For example, total electron scattering cross sections were measured at low electron energies for noble gases and some diatomic and triatomic molecules using a photoelectron source ([67] and references therein). Measurements in this otherwise technically new and innovative experiment had to be carried out at discrete electron energies. Such cross section data, though scientifically more accurate, should, by all means, be avoided and instead data obtained at continuous electron energies should be preferred.

(iii) Measurements of cross sections if made by two very different techniques can be used as a cross check on one another and on theory. Agreement of cross section data in such a case could be taken as an endorsement of the accuracy of the data. Also, experiments using the latest technologies should be preferred over others. Repeatability in the same experiment within the stipulated error is always desired.

(iv) Sometimes, only a single set of data is available for a given cross section. Such data may not be preferred for data evaluation and possible recommendation. However, such data may only be considered if repeatability is ensured by the authors, experiments have been carried out using new technology and the experimentalists in question have proven credentials.

(v) It is strongly believed that the experimental data should be preferred to those obtained by theoretical computation. This is probably true provided the targets are polyatomic and sometimes diatomic molecules. Such molecular systems undergo loss of symmetry and also, in such cases, the target wave function cannot be represented unambiguously. As a result, the theoretical cross sections obtained are much cruder. As against this, H₂, D₂ and HD are the simplest two-electron systems and are, therefore, of interest for quantum mechanical calculations beyond the Born-Oppenheimer approximation. Therefore, the theoretical data in the case of these molecules could at least be considered for evaluation.

Some specific comments are in order; some supporting the arguments put forward above and others regarding the technology and methodology used in the experiments.

(i) Cross checking experimental data could, sometimes, be useful if carried out by choosing some standard values provided by theory. The total electron-helium scattering cross section (electron energy 0–10 eV) calculated to an accuracy of 0.5% using variational principle [68] has been considered to be the best so far. So, all experiments on total electron scattering cross section measurement should be calibrated against Nesbet’s results for helium before taking up another target.

(ii) In case the measurement has to be carried out to an accuracy of 50 mev at say 15 eV, then the energy resolution (FWHM) of the projectile beam, in our case electron beam, should be much better than 50 mev at 15 eV. The energy resolution of 50 mev could be tolerable but anything above that value would lead to larger error bars in the measurement. In such a case, claiming accuracy to the third decimal place would practically be meaningless.

(iii) New technology can sometimes bring drastic improvements in the cross section measurement. All cross sections are, in general, measured at room temperature. Vibrational levels up to ν > 3 would be populated in the ground electronic state of any diatomic molecule at this temperature. There would also be rotational population in a few lowest vibrational states. Even though ν = 0 level is the most populated, the cross sections for higher vibrational states have been found to be much larger. In other words, the cross section values obtained are not exactly the desired ones. To counter this effect, the target molecules should be introduced in the reaction chamber through supersonic jets. The nearly adiabatic expansion cools them rapidly to temperatures of a few degree kelvin, while still leaving them in gas phase. This leads to a narrow velocity distribution of the target molecules and a highly resolvable and simplified spectrum. A still better device would be a time resolved molecular beam obtained using a pulsed supersonic jet [69].

5. Total and elastic scattering and momentum transfer cross sections

The well-validated integrated cross sections for elastic and inelastic processes obtained using different techniques are
(2) Electron impact cross sections for deuterated hydrogen and deuterium molecules

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The present paper should serve as a complete update for e+D₂, e+HD at different electron impact energy

But unfortunately there is a paucity of data for both HD and D₂

(1) Total electron impact scattering cross sections have not been measured so far for both HD and D₂ molecules
(2) The rotational and vibrational excitation cross sections for D₂ reported in the literature are the old sets of measured values. Also, such cross sections are not available for HD molecules at all.
(3) No measurements for the appearance energies of HD⁺, H⁺/HD and D⁺/HD are available. Also, total electron impact ionization cross sections for HD molecules have not been reported in the literature
(4) The absolute dissociative electron attachment cross sections for HD and D₂ molecules at 300K and at higher temperatures in the case of the 4eV process have been measured earlier but have been found in error. So, these need to be measured afresh. Also, no effort has been made to measure these cross sections at higher temperatures for the 8 and 14 eV processes
Summary and discussions

- Briefly introduce how to evaluate the cross section data from literatures
- Briefly introduce how to evaluate the uncertainty

- For the CRP (Evaluation of cross section for electron impact with hydrogen and their combination molecules in fusion plasma) we are going to start with our previous research for $H_2$, $D_2$, HD (+theoretical data ???)