Data needed for modeling Low Temperature Plasmas (LTPs)

Leanne Pitchford
Laboratory des Plasma et Conversion d'Energie (LAPLACE)
CNRS & University of Toulouse III, France

pitchford@laplace.univ-tlse.fr
This talk will focus low-temperature, non-equilibrium, collision dominated plasmas created in electric discharges operating at pd (pressure x dimensions) such that volume, not surface, processes dominate.

1) Context
2) Modeling LTPs - identification of data needs
3) Presentation of LXCat
4) Other data needs for modeling & actions
5) Conclusions
Technologies based on LTPs – some examples

Electrical energy

Radiant energy
Source of photons
- Lamps
  - Lighting
  - Water purification
  - Sterilization
- Visualisation
  - Plasma display panels
- Lasers

Kinetic energy
Source of charged particles
- Ion sources
  - Ion implantation
  - Propulsion
  - Ion sources for ITER
  - Elemental analysis of solids
- Electron beams
  - X-ray sources
- Switches, current interrupters

Chemical energy
Source of reactive species
- Surface processing
  - Microelectronics
  - Functionalisation of surfaces
- Volume processes
  - Pollution control
  - Chemical conversion
- Biological applications
Generation of LTPs

LTPs can be generated most simply by applying a voltage ($> V_b$) between two electrodes, separated by a gas gap.

For a range of conditions: $T_e >> T_i = T_g$ => the electrons are the vector through which electrical energy is deposited in the gas through collisions, leading to excitation, dissociation, & ionization.
Overview of discharge models

**INPUT:** gas composition and pressure; geometry; circuit,…

**OUTPUT:** $E, n_e, n_+, T_g$, neutral species densities,… as functions of $x, t$. 
Overview of discharge models

**INPUT:** gas composition and pressure; geometry; circuit,…

**OUTPUT:** $E$, $n_e$, $n_+$, $T_g$, neutral species densities,… as functions of $x,t$. 

Discharge model

- Charged particle transport and generation
- $E$ field
- + boundary conditions
Overview of discharge models

**INPUT:** gas composition and pressure; geometry; circuit,…

**OUTPUT:** \( \mathbf{E}, n_e, n_+, T_g \), neutral species densities,… as functions of \( \mathbf{x}, t \).
# Modeling charged particle transport & generation

<table>
<thead>
<tr>
<th>LEVEL OF DESCRIPTION</th>
<th>DATA NEEDS</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>PARTICLE MODELS</strong></td>
<td><strong>Electron- and ion-neutral scattering cross sections</strong>&lt;br&gt;“Complete” sets of electron/ion-neutral cross sections</td>
</tr>
<tr>
<td>(Boltzmann or Monte Carlo simulations)</td>
<td>Certain aspects of ion or electron behavior are treated with particle models; other aspects with fluid models.</td>
</tr>
<tr>
<td><strong>HYBRID MODELS</strong></td>
<td><strong>Electron and ion transport coefficients</strong> – mobility and diffusion coefficients - and ionization rates.</td>
</tr>
<tr>
<td><strong>FLUID MODELS</strong></td>
<td></td>
</tr>
<tr>
<td>(multi-fluid)</td>
<td></td>
</tr>
</tbody>
</table>
Measurements of transport coefficients (one example)

- **E = E_z =** constant, except near radial boundaries.
- Neutral gas number density, N.
- Electrons, generated at t=0, z=0, are accelerated to the anode \((a = -eE_z/m)\), undergoing collisions with neutrals along the way.
- **E/N** is a good parameter.
- "Swarm"

Analysis of current waveforms at the anode yields drift velocity, longitudinal diffusion coef, and ionization rate coef.

**Experimental set-up**

**Results**

![Current waveform graph](image)

*FIG. 4. Measurement with a typical best SNR in N_2 at p = 10 kPa, d = 25 mm, and \((E/N) = 110\) Td. (blue solid line) Single waveform and (black solid line) average of 200 single waveforms. (+) \(T_0\) and \(T_e\) of average.*

Haefliger & Franck, Rev. Sci. Instrum. 2018
Examples of measured electron transport coefficients

- The SI Unit for E/N is Townsend (Td), $10^{-21}$ V m$^2$.
- The relation between drift velocity and reduced mobility: $v_d = \mu N \times E/N$.
- The relation ionization rate coefficient and Townsend coeff.: $\alpha/N = k_i/v_d$.
- In Ar at 1 Td, the thermal energy is $\sim 2.4$ eV, whereas the directed energy is $\sim 2.10^{-2}$ eV.
- Other measureable transport data include $D_T N$, $D_L N$, $D_T/\mu$.

See Dutton database on www.lxcat.net for data with references
LTPs – a closer look

cathode

electron

anode

\[ V \]
Boltzmann transport equation

\[ f(r,v,t) = \text{electron velocity probability distribution function} \]

\[
\frac{\partial f}{\partial t} + \vec{v} \cdot \nabla f + \vec{a} \cdot \nabla f = NC \left[ f \right]
\]

\( \vec{a} = -eE/m \)

\[
\text{transport in } r
\]

\[
\text{transport in } v
\]

Collisions

where \( \int f(r,\nu, t) d\nu = n_e(r, t) \)

Electron transport & rate coefficients are integrals over \( f(r,\nu, t) \).
Input data required for Boltzmann equation solutions

**Types of collisions:**

- **Elastic**
  Recoil energy loss, momentum transfer ($Q_{m,el}$)

- **Inelastic**
  Discrete energy losses due to excitation of rotational, vibrational and electronic states ($Q_{k,T}$)

- **Ionization**
  Two electrons exit the collision event ($Q_{i,T}$, energy sharing)

Pitchford & Phelps, 1982
Solutions of the Boltzmann equation

**Analytical solutions** exist only for simplified cases, elastic scattering only, power law $Q_{m,el}$ (eg Maxwellian, Druyvesteyn).

An important research activity has been the development of **numerical solution techniques** which enable direct connection of fundamental AMO data & plasma transport.
The LXCat project (initiated at LAPLACE in 2010)

LXCat is a web-based, community-wide project for the curation of data needed in modeling low-temperature plasmas

Electron + neutral cross sections / oscillator strengths / swarm parameters

Ion + neutral cross sections / interaction potentials / swarm parameters

Plasma-surface interactions

Plasma chemistry

Radiation

LXCat

ELECtron (and ion) SCATtering

About the project
The Plasma Data Exchange Project is a community-based project which was initiated as a result of a public discussion held at the 2010 Ossucio Electronic Conference (OEC), a leading international meeting for the Low-Temperature Plasma community. The project aims to address, at least in part, the well-recognized need of the community to organize the masses of collating, validating and sharing data both for modeling and for interpretation of experiments. The heart of the Plasma Data Exchange Project is LXCat (pronounced ‘Marxcat’), an open-access website for collecting, cataloging, and downloading electron and ion scattering cross sections, swarm parameters (mobility, diffusion coefficient), reaction rates, energy distribution functions, etc., and other data required for modeling low-temperature plasmas. The electronic database has been contributed by members of the community and is maintained by the contributors' choice titles.

This is a dynamic website, evolving as contributors add or upgrade data. Check back again frequently.

Supporting organizations
Contributors to the LXCat project (to Sept 2018)

**Website conception:**  S Pancheshnyi, (France /Switzerland)

**Scattering cross sections (compilations, quantum calculations, measurements) :**  MC Bordage, V. Puech, LC Pitchford (France); SF Biagi, D Brown, J Tennyson (UK); K Bartschat, WL Morgan, AV Phelps, J. Stephens, L Viehland, MC Zammit, O Zatsarinny (USA); LL Alves, C Ferreira, V Guerra (Portugal); NA Dyatko, IV Kochetov, AP Napartovich (Russia); Y Itikawa (Japan); I Bray, S Buckman, M Brunger, L Campbell, D Fursa, McEachran (Australia); A Stauffer (Canada); RK Gangwar, L Sharma, R Srivastava (India)

**Oscillator strengths:**  C Brion (Canada)

**Transport/rate coefficients (compilations, measurements) :**  L Viehland, AV Phelps (USA); S Chowdhury (France), J de Urquijo (Mexico); LL Alves, V Guerra (Portugal); Christophorou (Greece); A Chachereau, CM Franck, P Haefliger, A Hoesl, M M Hildebrandt (Germany); L Rabie (Switzerland); X-M Zhu (China); I Jogi (Estonia)

**Ion-neutral interaction potentials:**  L Viehland (USA)

**Initial website development:**  S Pancheshnyi, (France /Switzerland); B Chaudhury (India),

**Tech support:**  W Graef, D Mihailova, J van Dijk (The Netherlands); M Hopkins, B Yee (USA), Pancheshnyi

**Outreach:**  K Bartschat (USA), E Carbone (Germany), LC Pitchford (France), Y-K Pu (China)

**On-line Bolsig+ :**  GJM Hagelaar (France); S Pancheshnyi (Switzerland)

**Servers:**  Eindhoven Technical Univ. & Univ Toulouse
Complete sets of e/neutral cross sections available on LXCat

....for electron scattering in COLD gases, $Q_{k,T}$ for inelastics, $Q_{m,el}$ for elastic

**Atomic gases**
Ar, C, Cu, H, He, Hg, Kr, Mg, N, Na, O, Xe

**Diatomcic gases**
CH, CO, Cl$_2$, D$_2$, F$_2$, H$_2$, HCl, N$_2$, NO, O$_2$

**Polyatomic gases**
C$_2$H$_2$, C$_2$H$_4$, C$_2$H$_6$, C$_3$H$_6$, C$_3$H$_8$, CCl$_2$F$_2$, CCl$_4$, CF$_4$, CH$_4$, CH$_3$, CH$_4$, CHF$_3$, CO$_2$, H$_2$O, N$_2$O, SF$_6$, SO$_2$, Si$_2$H$_6$, Si(CH$_3$)$_4$, SiH$_4$

**ATTENTION:** LXCat does not recommend data.

On-line: **BOLSIG+ Lite** => Boltzmann solver for quick on-line calculations of transport and rate coefficients in gas mixtures, comparisons with measurements
Other data needs include plasma chemistry

The consensus in the Low Temperature Plasma Community is that there is a need to define a strategy to develop, validate (to the extent possible) and distribute reaction mechanisms for some common gas mixtures, analogous to the effort already made in the combustion community (GRE-Mech).

Example: a basic mechanism for helium, suitable for ionization balance calculations.

"Mechanism" – kinetic scheme with associated rate coefficients, validated over a range of conditions by comparison with expts. The detail required depends on the questions being addressed by the model.

LXCat policy (1)

1) Anyone willing to contribute data to the site can request a password and set up a database. => Data for the same processes can be listed in multiple databases. **LXCat does not recommend data.**

Example: complete sets of electron-Argon cross sections

---

**Phelps database**

![Graph of cross section vs energy for Phelps database]

**Hayashi database**

![Graph of cross section vs energy for Hayashi database]
2. The site is open access and data can be downloaded without registering or paying a fee, but proper referencing is essential for the survival of LXCat.

Downloaded data should not become anonymous!

Required reference format:
[database name], www.lxcat.net, [retrieved date]

+ List all references given in the database for the species

3. **Databases are dynamic.** Contributors make changes as new data become available or when corrections are needed. **Data as they existed at a specific data in the past can be recovered online.**
Conclusions

This talk focused on the data related to the electron component in LTPs. Data needs are still far from being satisfied.

It is becoming very important for the LTP community to establish and distribute recommended reaction mechanisms, at least for common gas mixtures. Actions underway towards this end include a round-robin exercise and a proposed COST action (EU) coordinated by Miles Turner.

Plasma surface/liquid interactions are areas of continuing/emerging interest for which data are largely unavailable.
END
What’s been learned from the LXCat experience

Reasons why LXCat is a success:
- responds to a well-recognized need
- well-defined scope
- easy to use, open-access, on-line tools....
- a community-wide effort
- responsive but not beholden to commercial activities
- compatible with existing software
- nurtured by the GEC

Outstanding issues:
- long-term survivability (non-profit association)
- efficient use of limited resources
- maintain visibility of LXCat and its contributors
- recommended data?