



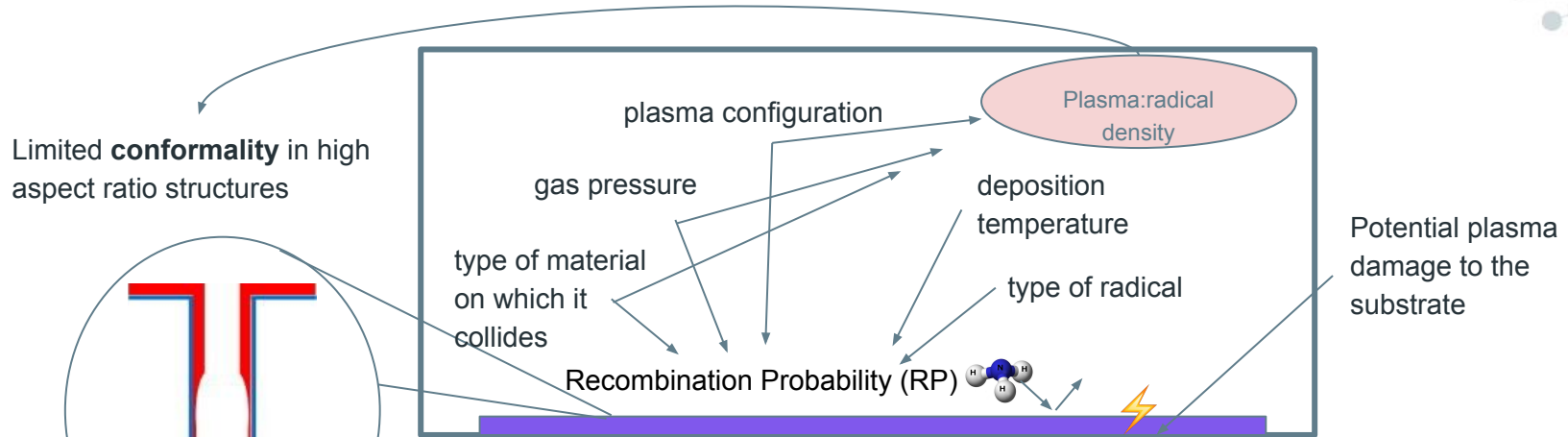
Assembling and optimizing plasma chemistry sets using QDB

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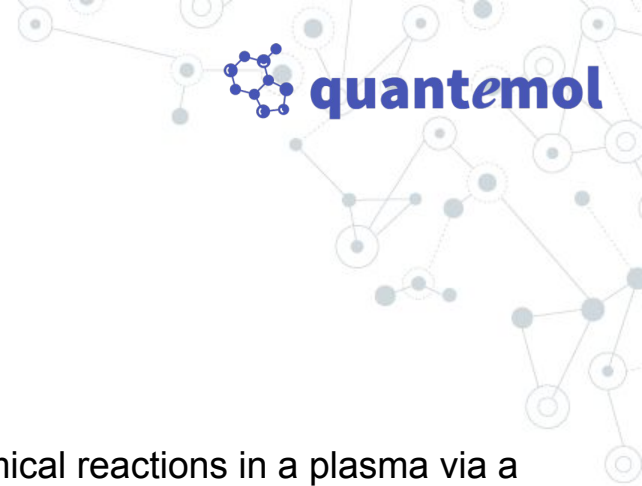


Potential issues which can be addressed with simulations



["Conformality in atomic layer deposition: Current status overview of analysis and modelling featured."](#)
 V.Cremers, R.L. Puurunen and J.Dendooven, Applied Physics Reviews 6, 021302 (2019)

Higher radical densities lead to higher radical fluxes deeper in the trench, enhancing the conformality



Problem statement

Every plasma simulation consists of two parts:

Physical Model:

Describes particle transport, electromagnetic fields etc.

Should be chosen to according to parameter space and specific balance of speed and accuracy:

PIC, Fluid, DSMC, Hybrid

Global, 1D, 2D, 3D

Chemical Model:

Describes the chemical reactions in a plasma via a set of species :

- Neutrals
- Excited states
- Positive and negative ions
- Electrons

And reactions:

- Electron impact collisions
- Heavy particle collisions

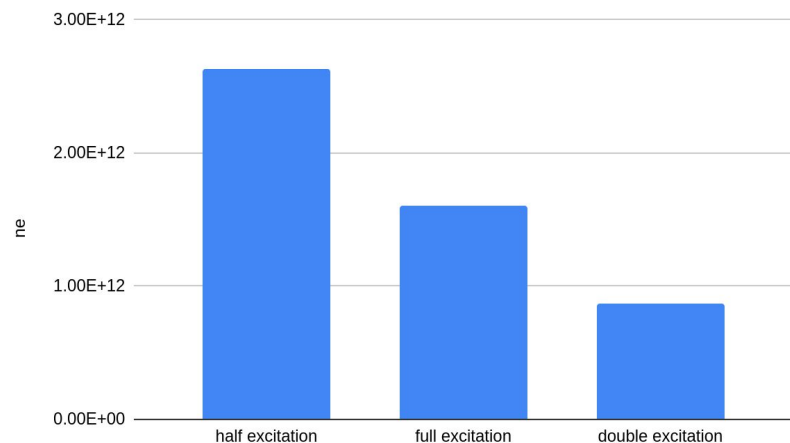


Overview of Electron impact collisions

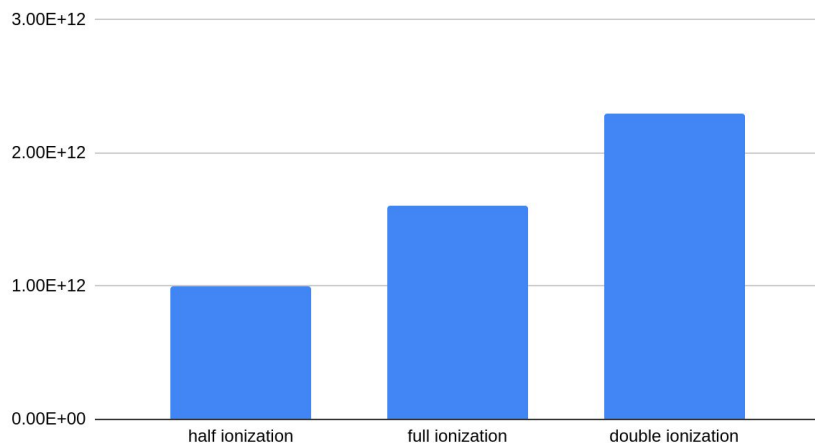
- Elastic collision/momentum transfer: electron transport, gas heating
- Excitation: Determines density of excited states.
- Dissociation: Determines density of dissociation products.
- Dissociative Attachment: Determines electronegativity
- Electron detachment: Determines electronegativity
- Ionization: Determines ionization rate
- Recombination: Determines recombination rate

Overview of Electron impact collisions

Electron density - Excitation Variation



Electron density - Ionization Variation

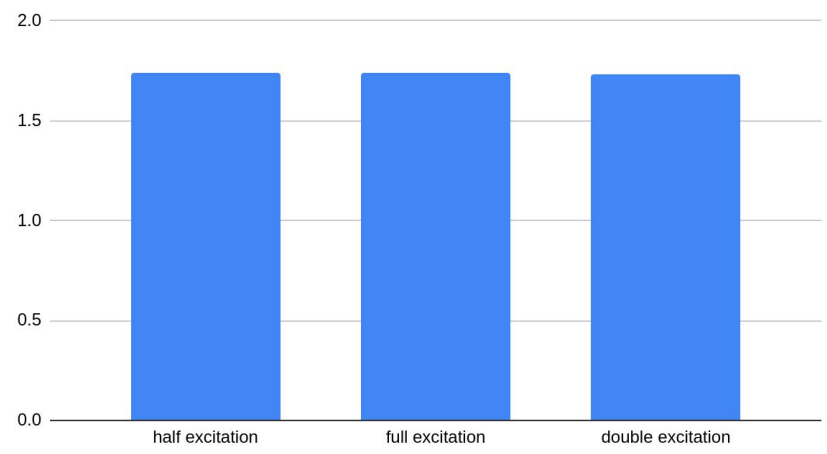


Excitation processes have a larger influence on electron density than the actual ionization processes.

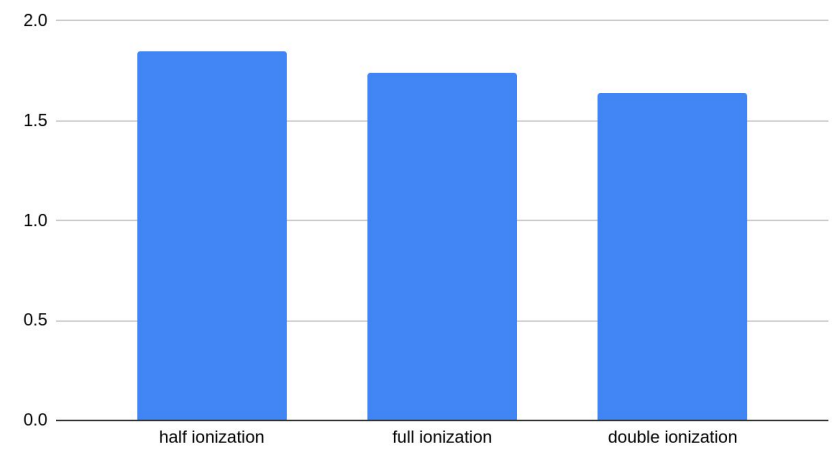
→ If the electron density looks off, turn your attention to excitation and dissociation reactions!

Overview of Electron impact collisions

Electron temperature - Ionization Variation

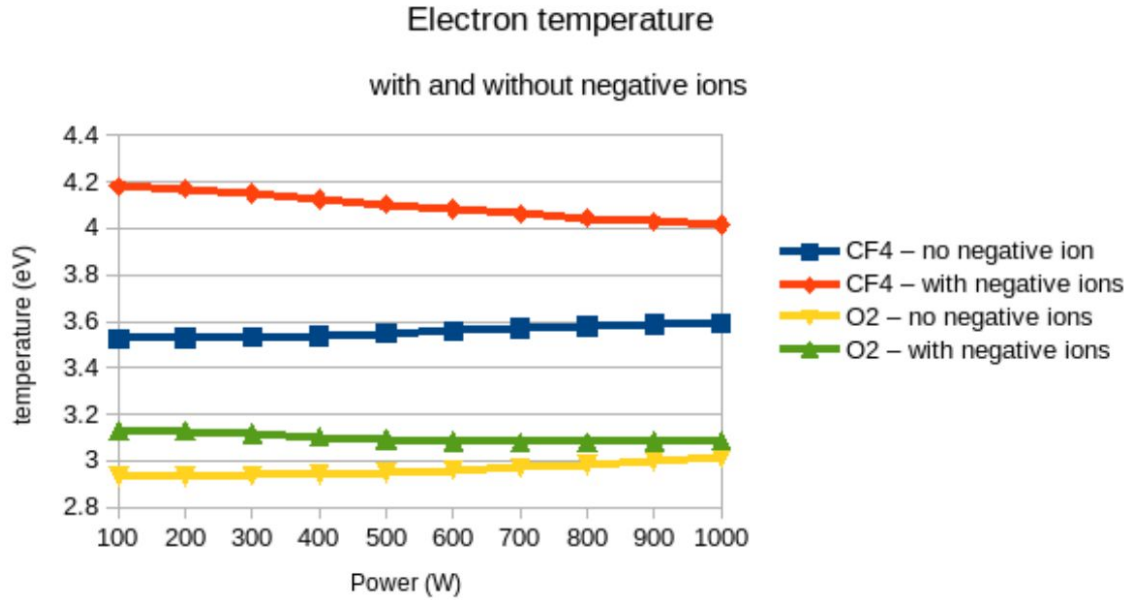


Electron temperature - Ionization Variation



Only ionization processes significantly affect the electron temperature in a simple argon model.

Overview of Electron impact collisions



The presence of negative ions generally increases the electron temperature.
 → If the electron temperature looks off, turn your attention to electron production and losses!

Overview of Heavy Particle Collisions

- **Ion-Ion Recombination**
- Symmetric Charge Exchange
- Asymmetric Charge Exchange
- Neutral conversion
- Electron detachment
- Penning Ionization



Vital for electronegative plasmas.

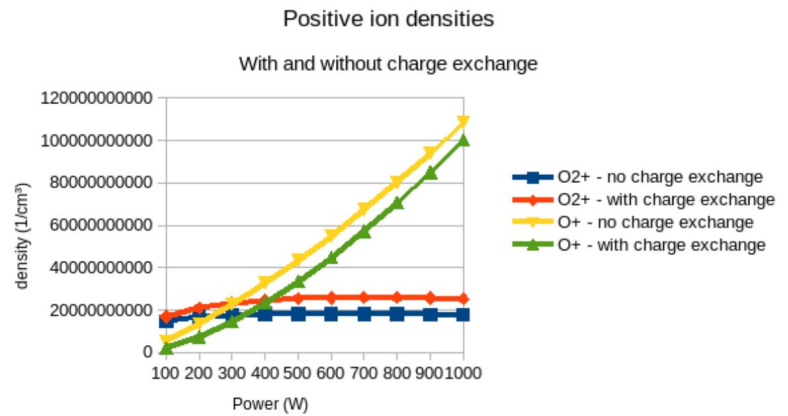
Typically represents the only efficient loss channel for negative ions.

Overview of Heavy Particle Collisions

- Ion-Ion Recombination
- Symmetric Charge Exchange
- **Asymmetric Charge Exchange**
- Neutral conversion
- Electron detachment
- Penning Ionization



Asymmetric charge exchange affect the relative ion densities apart from heating neutrals.

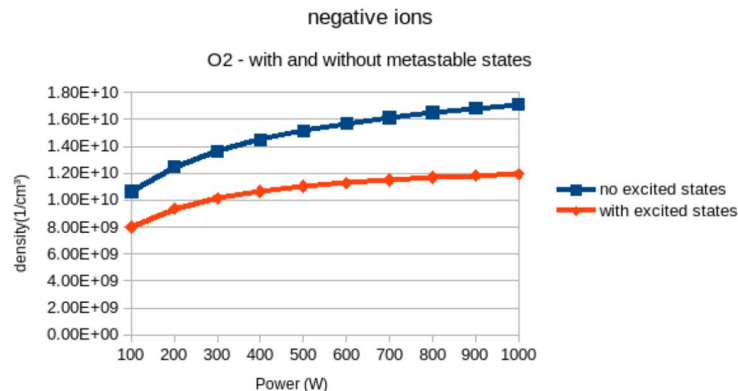


Overview of Heavy Particle Collisions

- Ion-Ion Recombination
- Symmetric Charge Exchange
- Asymmetric Charge Exchange
- Penning Ionization
- Neutral conversion
- **Electron detachment**



Electron detachment can significantly influence the electronegativity of a plasma.



Approaches to chemistry set design

All-In

Simply collect all reactions and species found relevant for process parameters

Pro: Fast

Contra: Hard to identify problematic reactions, e.g. missing ones, duplicates etc.

Well suited for established sets with sufficient available data

Step-by-Step

Start with one feed gas and add reactions. Identify “new” species, e.g. ions, excited states, dissociation products and reactions one by one.

Repeat for other feed gases.

Add cross-reactions and reactions for resulting species.

Pro: More control, easy to identify missing reactions etc.

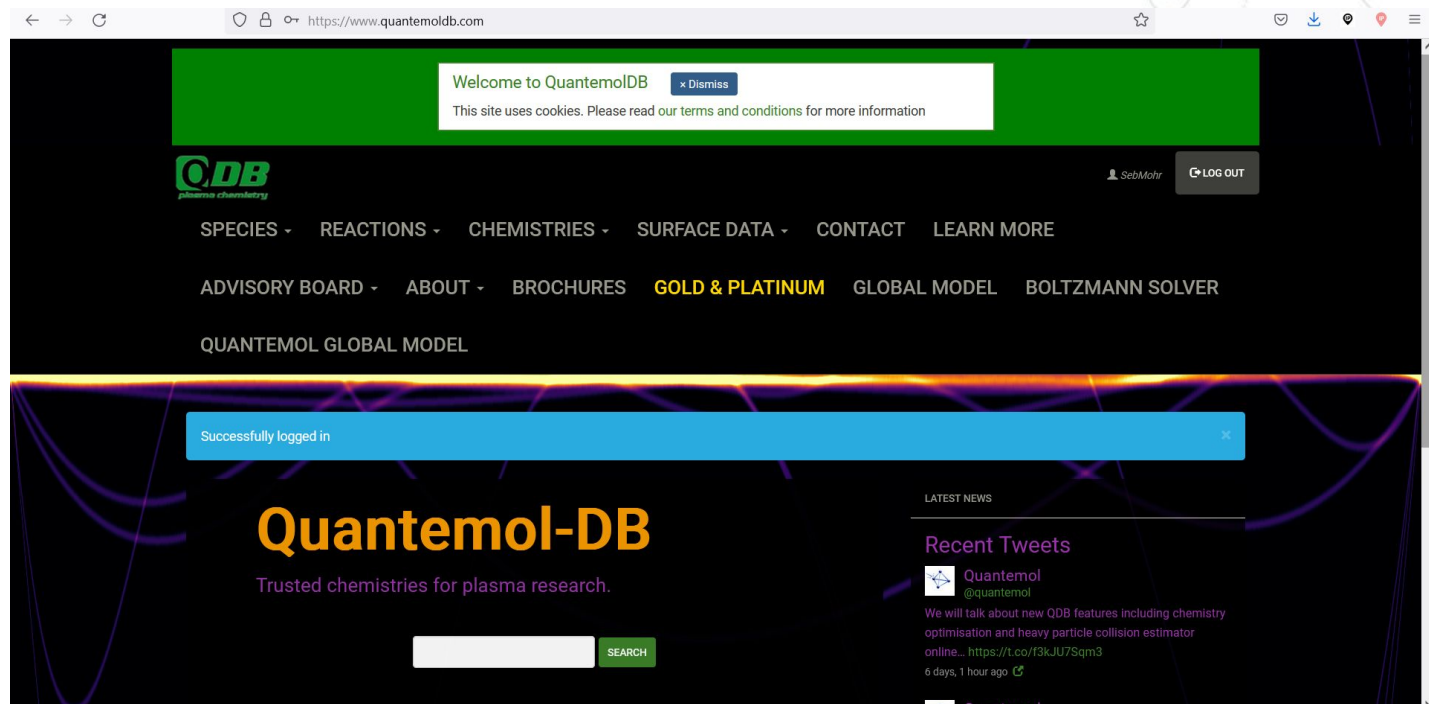
Contra: Needs more time

Suited for sets with “new” gases or data gaps.

Where to get relevant data?

QDB offers access to cross-section and rate coefficients data for almost 30 000 reactions.

It also gives access to pre-assembled chemistry sets, species data such as enthalpies of formation, and some surface coefficients.



The screenshot shows the homepage of the Quantemol-DB website. At the top, there is a green banner with a "Welcome to QuantemolDB" message and a "Dismiss" button. Below this is a navigation menu with links for SPECIES, REACTIONS, CHEMISTRIES, SURFACE DATA, CONTACT, LEARN MORE, ADVISORY BOARD, ABOUT, BROCHURES, GOLD & PLATINUM, GLOBAL MODEL, and BOLTZMANN SOLVER. The main content area features the Quantemol-DB logo and the tagline "Trusted chemistries for plasma research." There is a search bar with a "SEARCH" button. On the right side, there is a "Recent Tweets" section with a tweet from @quantemol about new QDB features.

Desktop app

Chemistry sets can be quickly created and tested via 0D simulations in the QDB desktop app.

QDB Global Model

Set Generation | Chemistry Set | Boltzmann Solver | Model Settings | Results | Analysis | Data Estimation

25 species are selected.

	Name	Sticking Coefficient	Return Coefficient	Return Species
<input checked="" type="checkbox"/>	Delete Properties H	0.0000	0.0000	H
<input checked="" type="checkbox"/>	Delete Properties H+	1.0000	1.0000	H
<input checked="" type="checkbox"/>	Delete Properties H-	1.0000	1.0000	H
<input checked="" type="checkbox"/>	Delete Properties H2	0.0000	0.0000	H2
<input checked="" type="checkbox"/>	Delete Properties H2+	1.0000	1.0000	H2
<input checked="" type="checkbox"/>	Delete Properties H3+	1.0000	1.0000	H
<input checked="" type="checkbox"/>	Delete Properties H[n=2]	1.0000	1.0000	H
<input checked="" type="checkbox"/>	Delete Properties N	0.0000	0.0000	N
<input checked="" type="checkbox"/>	Delete Properties N+	1.0000	1.0000	N
<input checked="" type="checkbox"/>	Delete Properties N2	0.0000	0.0000	N2
<input checked="" type="checkbox"/>	Delete Properties N2+	1.0000	1.0000	N2
<input checked="" type="checkbox"/>	Delete Properties N2H	0.0000	0.0000	N2H
<input checked="" type="checkbox"/>	Delete Properties N2H2	0.0000	0.0000	N2H2

Auto-Add Reactions

Filtering

Show only selected species

Ground State Neutrals

Excited States

Positive Ions

Negative Ions

Excited States

Use pooled states

Use distinct states

Enable Consistency Checks

Some symmetric charge exchange reactions are missing!

149 reactions are selected.

	Reaction	n	E
<input checked="" type="checkbox"/>	Delete Info e- + H2 > e- + H2	1.73e-07	-0.35
<input checked="" type="checkbox"/>	Delete Info e- + H2 > e- + H + H	1.15e-07	-0.58
<input checked="" type="checkbox"/>	Delete Info e- + H2 > e- + e- + H2+	1.09e-08	0.46
<input checked="" type="checkbox"/>	Delete Info e- + H2 > e- + H + H[n=2]	1.96e-10	0.27
<input checked="" type="checkbox"/>	Delete Info e- + H2+ > e- + H2+	1.44e-04	-1.19
<input checked="" type="checkbox"/>	Delete Info e- + H2+ > e- + H+ + H	2.72e-07	-0.24
<input checked="" type="checkbox"/>	Delete Info e- + H2+ > H + H	4.94e-08	-0.50
<input checked="" type="checkbox"/>	Delete Info e- + H > e- + H	1.97e-07	-0.29
<input checked="" type="checkbox"/>	Delete Info e- + H > e- + H[n=2]	2.03e-08	0.37
<input checked="" type="checkbox"/>	Delete Info e- + H > e- + e- + H+	4.54e-09	0.72

Auto-Update Species

Show only selected reactions

Filter by Type

show electron collisions

show heavy particle collisions

Filter by Reactants

Has Reactants

H
H+
H-
H2
H2+
H3+
H[n=2]
N
N+
N2
N2+
N2H
N2H2
N2H3
N2[*]

Filter by Products

Has Products

H[n=2]
N
N+
N2
N2+
N2H
N2H2
N2H3
N2[*]
N3+
N4+
NH
NH+
NH2
NH2+

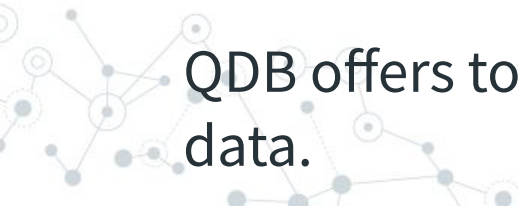
How to optimize sets for given process parameters

The logo for 'quantemol' is located in the top right corner. It features a stylized molecular structure icon consisting of several blue spheres connected by lines, followed by the word 'quantemol' in a blue, sans-serif font.

On the one hand, comprehensive sets usually contain more species/reactions than necessary to sufficiently predict important quantities such as the density of neutrals reacting with the surfaces.

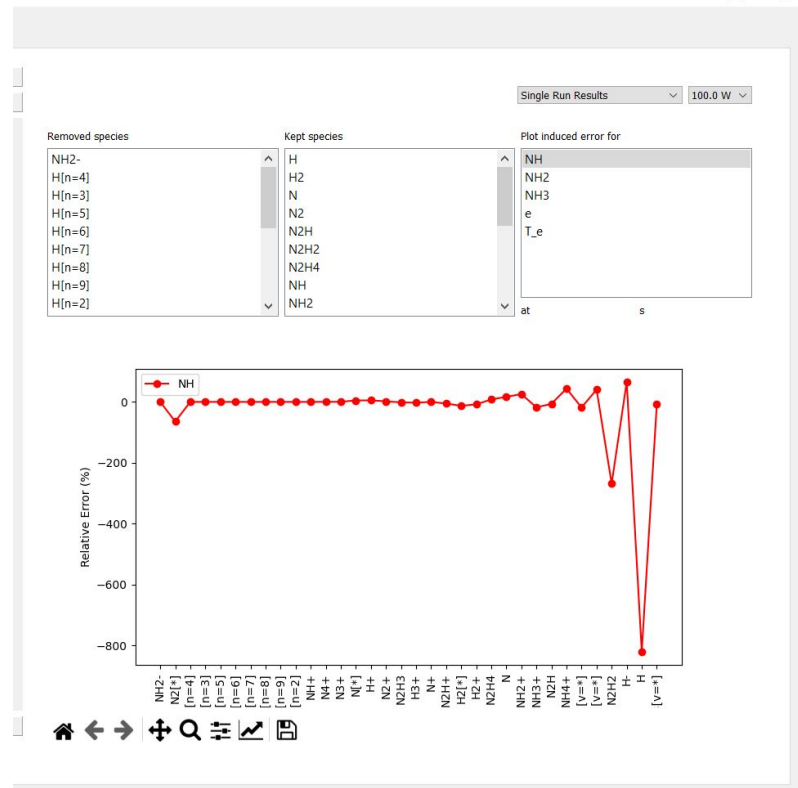
On the other hand, important data such as coefficients describing surface processes might be missing/unknown/imprecise.

QDB offers tools to optimize sets and fill/calibrate missing data.

A decorative graphic in the bottom left corner shows a network of grey nodes connected by thin lines, resembling a molecular or computational graph.

Optimization - Set reduction

The desktop app now allows the user to reduce chemistry sets for given process parameters including variations of one parameter to the smallest set which reproduces chosen plasma parameters within a defined error margin compared to the full set. This allows to optimize sets before exporting them to multi-dimensional models.



Optimization - Set reduction

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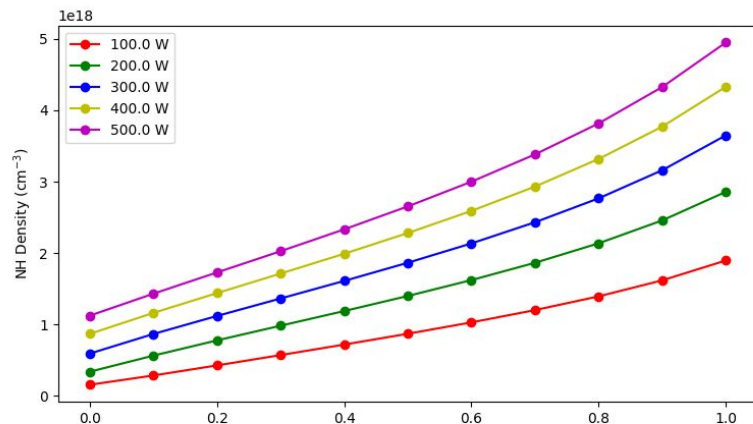
Parameter Variation Overview | 100.0 W

	100.0 W	200.0 W	300.0 W	400.0 W	500.0 W
H[n=5]	0.42 % in e	-0.14 % in NH3	-5.01 % in NH2	-2.14 % in NH2	-2.44 % in NH3
H[n=6]	0.44 % in e	-0.42 % in NH3	-4.91 % in NH2	-2.43 % in NH2	-1.21 % in NH2
H[n=7]	0.45 % in e	-0.85 % in NH3	-4.55 % in NH2	-2.23 % in NH3	-2.89 % in NH3
H[n=8]	0.45 % in e	-0.86 % in NH3	-3.90 % in NH2	-3.48 % in NH3	-4.69 % in NH3
H[n=9]	0.46 % in e	-0.87 % in NH3	-3.89 % in NH2	-6.07 % in NH3	-4.88 % in NH3
H[n=2]	0.93 % in e	-1.98 % in NH3	-6.63 % in NH3	-12.03 % in NH3	-22.74 % in NH3
NH+	0.96 % in e	-2.87 % in NH	-3.32 % in NH2	5.20 % in NH3	-2.52 % in NH3
N4+	0.84 % in e	-2.08 % in NH	-5.76 % in NH3	-2.99 % in NH3	-4.95 % in NH3
N3+	0.86 % in e	2.06 % in e	-6.88 % in NH3	-5.22 % in NH3	-8.75 % in NH3
N[*]	4.81 % in NH	9.55 % in NH2	32.21 % in NH2	47.23 % in NH2	56.28 % in NH2
H+	5.08 % in NH	9.98 % in e	11.18 % in e	-14.38 % in NH3	-31.25 % in NH3
N2+	6.71 % in e	7.20 % in e	8.28 % in e	17.76 % in NH3	27.00 % in NH3
N2H3	6.73 % in e	-2.76 % in NH	-1.82 % in NH	-1.32 % in NH	-1.30 % in NH2
H3+	6.76 % in e	10.09 % in e	8.33 % in e	2.02 % in e	2.59 % in NH2
N+	12.40 % in e	-13.20 % in NH2	-11.13 % in NH3	-10.62 % in NH3	-17.47 % in NH3
N2H+	6.93 % in e	10.13 % in e	27.73 % in NH3	68.75 % in NH3	68.36 % in NH3
H2[*]	12.91 % in NH	-11.64 % in NH	11.63 % in NH3	5.72 % in NH2	6.92 % in NH2
H2+	15.56 % in e	21.85 % in NH2	19.81 % in e	10.66 % in NH2	11.16 % in NH2
N2H4	17.37 % in NH2	9.83 % in NH2	-5.20 % in NH2	-2.50 % in NH2	-2.52 % in NH2

Optimization - Surface Coefficient Variation

You can now run automated variations of surface coefficient for one absorbed/produced species pair, e.g. H producing NH at surfaces. Options are:

- a) Vary only sticking coefficient
- b) Vary only return coefficient
- c) Vary both



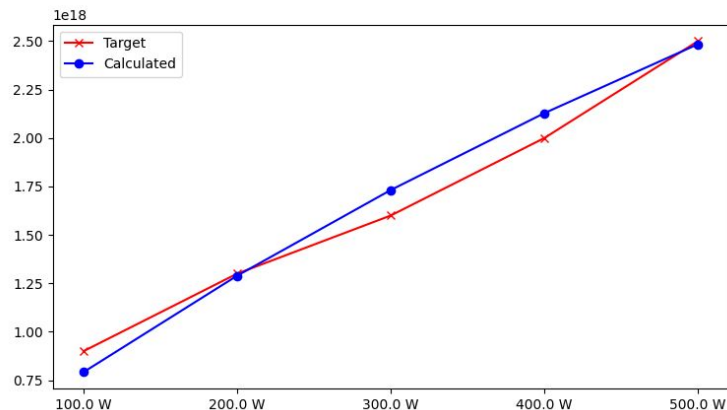
Here the return coefficient for NH produced by H with a constant sticking coefficient of H was varied.

Optimization - Surface Coefficient Calibration

You can also input target densities for the absorbed/produced species and find the coefficient (pair) which best reproduces these densities.

Options are:

- a) Optimize only sticking coefficient for absorbed species
- b) Optimize only return coefficient for produced species
- c) Optimize both for either or both densities.



The example shows again NH produced by H with a constant sticking coefficient for H; plotted are the calculated NH densities for the optimum coefficient as a function of pressure compared to the target densities.

Estimating rate coefficients by Machine Learning



Martin Hanicinec has developed an ML model as part of his PhD to estimate rate coefficients for binary heavy particle collisions $A + B > C + D$. This algorithm is embedded into the website and available via the QDB desktop app.

The screenshot shows the 'QDB Global Model' desktop application. The main window displays a table titled 'Rate Coefficient Estimator by Machine Learning'. The table has columns for 'Formula', 'Enthalpy of Formation (kJ/mol)', 'Neutral Enthalpy of Formation (kJ/mol)', 'Dipole Moment (Debye)', and 'Polarisability (Å³)'. The data is organized into rows for Reactant 1, Reactant 2, Product 1, Product 2, and Product 3 (optional).

	Formula	Enthalpy of Formation (kJ/mol)	Neutral Enthalpy of Formation (kJ/mol)	Dipole Moment (Debye)	Polarisability (Å³)
Reactant 1	CF	255.28266101278808	255.28266101278808	-	-
Reactant 2	H	218.03572309032182	218.03572309032182	0.0	0.667
Product 1	C	716.714662560772	716.714662560772	0.0	1.76
Product 2	HF	-272.49733587268776	-272.49733587268776	1.82	-
Product 3 (optional)					

At the bottom of the window, a status bar indicates: 'The rate coefficient is estimated to be 5.43e-11 cm³/s' and provides a link: 'Get Species Data from QDB: Estimate Rate Coefficient'.

The user inputs the desired reaction; used species parameters are displayed and can be changed manually.

Radiative Process Database LIDB

A separate database for radiative processes, LIDB, has been developed. It currently contains mostly information about molecular transitions; once atomic data has been added, an API will be added to the desktop app to quickly add radiative processes to the chemistry sets.

LIDB [/fetimes/database](#) [Data](#) [About](#) [API](#) [Contact](#)

Transitions of HF

Search:

Initial state	Final state	ΔE (eV)	Partial Lifetime (s)
v=1	v=0	-0.491	5.16e-03
v=2	v=0	-0.961	4.44e-02
v=2	v=1	-0.470	3.02e-03
v=3	v=0	-1.410	8.21e-01
v=3	v=1	-0.919	1.52e-02
v=3	v=2	-0.449	2.40e-03
v=4	v=0	-1.839	8.05e+00
v=4	v=1	-1.348	2.25e-01
v=4	v=2	-0.878	7.77e-03
v=4	v=3	-0.429	2.21e-03
v=5	v=0	-2.248	7.12e+01
v=5	v=1	-1.757	1.75e+00
v=5	v=2	-1.287	9.88e-02
v=5	v=3	-0.838	4.71e-03
v=5	v=4	-0.409	2.25e-03
v=6	v=1	-2.146	1.20e+01

Initial state: Final state: ΔE (eV): Partial Lifetime (s):

Showing 1 to 17 of 35 entries

LIDB is funded by STFC project ST/W005504/1.
It uses ExoMol data supported by ERC Advanced Investigator Projects 267219 and 883830.

