Quantemol Global Model Documentation

This document describes the Quantemol Global Model and explicitly the equations which are being solved for inside the framework.

It is our intention, that this document gives the most complete and transparent view into the model and that anyone could recode the model based on this documentation and, with the choice of the same ODE solver, obtain exactly the same solution for any problem.

Contents

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Symbols

Symbol	Unit	Description
i, k		Index, running over species in a kinetic scheme
i_0, i_+, i_-		Indices i running over neutral, positive and negative species respectively
j		Index, running over reactions in a kinetic scheme
N_S		number of species in a kinetic scheme
n_i	$[\mathrm{m}^{-3}]$	Number density of the i-th species in a kinetic scheme
n_e	$[\mathrm{m}^{-3}]$	Electron number density
ϱ_e	$[\mathrm{eV{\cdot}m}^{-3}]$	Electron energy density
P	[W]	Absorbed power
p_0	[Pa]	Desired pressure
p	[Pa]	Instantaneous pressure
Q_i	[sccm]	Feed flow for i-th species in a kinetic scheme
$R_\mathrm{p}, Z_\mathrm{p}$	[m]	Plasma dimensions: radius and length
V	$[\mathrm{m}^3]$	Plasma volume
$\mathrm{T_g}$	[K]	Neutral temperature
$\mathrm{T_i}$	[K]	Positive ion temperature
$\mathrm{T_e}$	[K]	Electron temperature
T_g	[eV]	Neutral temperature
T_i	[eV]	Positive ion temperature
T_e	[eV]	Electron temperature
k_j	$[\mathrm{m}^{-3+3m}\mathrm{s}^{-1}]$	Reaction rate coefficient of the j-th reaction of an order m in a kinetic scheme
R_j	$[\mathrm{m}^{-3}\mathrm{s}^{-1}]$	Reaction rate of the j-th reaction in a kinetic scheme
$M_{\mathrm{cp},j}$	[kg]	Mass of the collision partner in j-th electron reaction
M_i	[kg]	Mass of the i-th species
m_e	[kg]	Electron mass
q_i	[e]	Charge of the i-th species
e	[C]	Electron charge
σ_i^LJ	[m]	σ parameter of the Lennard-Jones potential for i-th species
$\Delta E_{\mathrm{e},j}^\mathrm{inel}$	[eV]	Electron energy loss due to an inelastic collision j
a_{ij}^L		Stochiometric coefficient of i-th distinct species on left hand side in j-th reaction
a_{ij}^R		Stochiometric coefficient of i-th distinct species on right hand side in j-th reaction

Symbol	Unit	Description
A_j	$[\mathrm{m}^{-3+3m}\mathrm{s}^{-1}]$	Arrhenius parameter – pre-exponential factor
n_j		Arrhenius parameter – exponent
$E_{\mathrm{a},j}$	[eV]	Arrhenius parameter – activation energy
$\mathrm{E}_{\mathrm{a},j}$	[K]	Arrhenius parameter – activation energy
k_B	$[\mathrm{JK}^{-1}]$	Boltzmann constant
s_i		Sticking coefficient – probability of i-th species sticking to a plasma boundary ($s_i \in [0,1]$)
r_{ik}		Return coefficient – number of i-th species returned for each *one* of *stuck* k-th species ($r_{ik} \in \mathbb{R}_0^+$)
D_i	$[\mathrm{m}^2\mathrm{s}^{-1}]$	Diffusion coefficient of i-th species
D_a	$[\mathrm{m}^2\mathrm{s}^{-1}]$	Ambipolar diffusion coefficient
Λ	[m]	Characteristic diffusion length
λ_i	[m]	Mean free path of i-th species
$\overline{v}_i$	$[\mathrm{ms}^{-1}]$	Mean speed of i-th species
σ_{ik}^m	$[\mathrm{m}^2]$	Mommentum transfer cross section for i-th species scattering on k-th species
$\overline{V_\mathrm{s}}$	[V]	Mean sheath voltage
n_min	$[\mathrm{m}^{-3}]$	Minimal allowed particle density
$\varrho_{\mathrm{e,min}}$	$[\mathrm{eV}\cdot\mathrm{m}^{-3}]$	Minimal allowed electron energy density

Model Description

The model solves the set of following ordinary differential equations (ODE):

- **Particle density balance equation for heavy species**, including contributions from volumetric reactions, flow and from diffusion sinks and surface sources of n_i.

- **Electron energy density balance equation**, including contributions of power absorbed by the plasma, elastic and inelastic collisions between electrons and heavy species, generation and loss of electrons in volumetric reactions and power lost to walls by electrons and ions.

The electron density n_e is not solved for explicitly but rather implicitly by enforcing the charge neutrality. Also, the heavy species temperature is not resolved in the model, but rather treated as a constant input parameter. The collisional kinetics is not described by cross sections, but rather parametrized for each reaction with the Arrhenius formula. The model was developed mainly for the purpose of plasma chemistry reduction, which justifies it's simplicity and the degree of approximation. For those reasons, the model should only be used with a great care to obtain any sort of quantitative results.

Input parameters for the model:

- Plasma parameters: P, p_0, Q_i, R_p, Z_p, $\mathrm{T_g}$

- Kinetic scheme parameters: M_i, q_i, σ_i^LJ, k_j, $\Delta E_{\mathrm{e},j}^\mathrm{inel}$

Model outputs:

- Densities of heavy species and electrons: n_i, n_e

- Electron temperature T_e

The rest of this document describes all the solved equations in great detail.

Particle Density Balance

$$\frac{\mathrm{d}n_i}{\mathrm{d}t} = \left(\frac{\delta n_i}{\delta t}\right)_{\mathrm{vol}} + \left(\frac{\delta n_i}{\delta t}\right)_{\mathrm{flow}} + \left(\frac{\delta n_i}{\delta t}\right)_{\mathrm{diff}}, \tag{1}$$

Volumetric Reactions Contribution

$$\left(\frac{\delta n_i}{\delta t}\right)_{\mathrm{vol}} = \sum_j G_{ij} - \sum_j L_{ij}, \tag{2}$$

where G_{ij} and L_{ij} are contributions of generation and loss of n_i due to inelastic reaction j. In greater detail, it can be written as

$$\left(\frac{\delta n_i}{\delta t}\right)_{\mathrm{vol}} = \sum_j (a_{ij}^R - a_{ij}^L) R_j, \tag{3}$$

$$R_j = k_j \prod_l n_{lj}^L, \tag{4}$$

where n_{lj}^L is the density of l^{th} species on the left hand side of reaction j. The reaction rate coefficient k_j in this model takes form of the Arrhenius equation

$$k_j = A_j \left(\frac{\mathrm{T_g}}{300\mathrm{K}}\right)^{n_j} \exp\left(-\frac{\mathrm{T_g}}{\mathrm{E}_{\mathrm{a},j}}\right) \tag{5}$$

for heavy species reactions j (reactions where all the reactants are heavy species) and

$$k_j = A_j \left(\frac{T_\mathrm{e}}{1\mathrm{eV}}\right)^{n_j} \exp\left(-\frac{T_\mathrm{e}}{E_{\mathrm{a},j}}\right) \tag{6}$$

for electron processes j (reactions where at least one reactant is an electron). The Arrhenius parameters A_j, n_j and $E_{\mathrm{a},j}$ (or $\mathrm{E}_{\mathrm{a},j}$) describe the collisional kinetics of the model.

Flow Contribution

The contribution of flow to the time evolution of heavy species densities will consist of inflow and outflow terms as well as a term regulating the pressure.

$$\left(\frac{\delta n_i}{\delta t}\right)_{\text{flow}} = \left(\frac{\delta n_i}{\delta t}\right)_{\text{flow}}^{\text{in}} + \left(\frac{\delta n_i}{\delta t}\right)_{\text{flow}}^{\text{out}} + \left(\frac{\delta n_i}{\delta t}\right)_{\text{flow}}^{\text{reg}} \tag{7}$$

Inflow

$$\left(\frac{\delta n_i}{\delta t}\right)_{\text{flow}}^{\text{in}} = \frac{Q'_i}{V}, \tag{8}$$

where $Q'_i = 4.485 \times 10^{17} \cdot Q_i$ is the inflow expressed in [particles/sec] rather than in [sccm].

Outflow

The outflow term is set in such a way that only neutrals are leaving the plasma region due to the flow, the neutral species flow rate is proportional to the species density and the total flow rate out of the plasma region is the same as total inflow rate:

$$\left(\frac{\delta n_i}{\delta t}\right)_{\text{flow}}^{\text{out}} = \begin{cases} -\frac{\sum Q'_i}{\sum n_{i_0}} \cdot \frac{n_i}{V} & \text{neutrals,} \\ 0 & \text{ions,} \end{cases} \tag{9}$$

where the index i_0 runs only over neutral species.

Pressure Regulation

A term regulating the plasma pressure is added to the particle balance equation, accounting for changes in p due to dissociation/association processes and to diffusion losses and surface sources. This term, similarly to the outflow term, only acts upon the neutral species and can be viewed as an addition to the outflow term, or physically as adjusting a pressure-regulation valve between a plasma chamber and a pump, based on the instantaneous pressure.

$$\left(\frac{\delta n_i}{\delta t}\right)_{\text{flow}}^{\text{reg}} = \begin{cases} -\frac{p - p_0}{p_0} \frac{n_i}{\tau_p} & \text{neutrals,} \\ 0 & \text{ions.} \end{cases} \tag{10}$$

Here, p is the instantaneous pressure from the state equation for an ideal gas

$$p = k_{\mathrm{B}} T_{\mathrm{g}} \cdot \sum_i n_i, \tag{11}$$

and τ_p is a pressure recovery time scale, in the model set to $\tau_p = 10^{-3}$ s.

Diffusion Contribution

The diffusion contribution towards the particle balance equation is ultimately controlled by vector of sticking coefficients s_i and matrix of return coefficients r_{ik} and the diffusion model:

$$\left(\frac{\delta n_i}{\delta t}\right)_{\mathrm{diff}} = \left(\frac{\delta n_i}{\delta t}\right)_{\mathrm{diff}}^{\mathrm{out}} + \left(\frac{\delta n_i}{\delta t}\right)_{\mathrm{diff}}^{\mathrm{in}}. \tag{12}$$

Diffusion losses

As proposed (among others) in [1], the rate of species loss to the plasma boundaries due to diffusion is expressed as

$$\left(\frac{\delta n_i}{\delta t}\right)_{\mathrm{diff}}^{\mathrm{out}} = -\frac{D_i}{\Lambda^2} n_i s_i, \tag{13}$$

where

$$\Lambda = \left[\left(\frac{\pi}{Z_{\mathrm{p}}}\right)^2 + \left(\frac{2.405}{R_{\mathrm{p}}}\right)^2\right]^{-1/2}. \tag{14}$$

The diffusion coefficient is calculated separately for neutrals and ions. For positive and negative ions, the diffusion coefficient is the coefficient of ambipolar diffusion in electronegative plasma, as proposed in [2].

$$D_i = \begin{cases} D_i^{\mathrm{free}} & \text{neutrals,} \\ D_+^{\mathrm{free}} \dfrac{1 + \gamma(1 + 2\alpha)}{1 + \alpha\gamma} & +\text{ions,} \\ 0 & -\text{ions.} \end{cases} \tag{15}$$

Here, $\gamma = T_{\mathrm{e}}/T_{\mathrm{i}}$ and $\alpha = \sum n_{\mathrm{i}^-}/n_{\mathrm{e}}$. $D_i = 0$ for negative ions implies that no negative ions are reaching the plasma boundaries and therefore there are no negative ion diffusion losses. It should be noted that the stated ambipolar diffusion coefficients are only valid for the case of $\alpha << \mu_\mathrm{e}/\mu_\mathrm{i}$, where μ are mobilities of electrons and ions respectively. The free diffusion coefficient for heavy species is calculated as

$$D_i^{\mathrm{free}} = \frac{\pi}{8}\lambda_i \overline{v}_i. \tag{16}$$

The mean free path λ_i for all heavy species is

$$\frac{1}{\lambda_i} = \sum_k n_k \sigma_{ik}^{\mathrm{m}}(1-\delta_{ik}), \tag{17}$$

where σ_{ik}^{m} is the momentum transfer cross section, and the mean speed $\overline{v}_i$ is the mean thermal speed

$$\overline{v}_i = \begin{cases} \left(\dfrac{8k_\mathrm{B}\mathrm{T_g}}{\pi M_i}\right)^{1/2} & \text{neutrals,} \\ \left(\dfrac{8k_\mathrm{B}\mathrm{T_i}}{\pi M_i}\right)^{1/2} & \text{ions,} \end{cases} \tag{18}$$

where, as proposed in [3],

$$\mathrm{T_i} = \begin{cases} (5800 - \mathrm{T_g})\,\dfrac{0.133}{p} + \mathrm{T_g} & p > 0.133 \text{ Pa}, \\ 5800 & p \leq 0.133 \text{ Pa}. \end{cases} \tag{19}$$

The momentum transfer cross section σ_{ik}^{m} is for the purpose of this model crudely approximated with hard sphere model for neutral–neutral and ion–neutral collisions, and with momentum transfer for Rutherford scattering for the case of ion–ion collisions:

$$\sigma_{ik}^{\mathrm{m}} = \begin{cases} \left(\sigma_i^{\mathrm{LJ}} + \sigma_k^{\mathrm{LJ}}\right)^2 & i = i_+, i_-, i_0, \text{ and } k = k_0, \\ & i = i_0, \text{ and } k = k_+, k_-, \\ \pi b_0^2 \ln\left(\dfrac{2\lambda_{\mathrm{De}}}{b_0}\right) [4] & i = i_+, i_-, \text{ and } k = k_+, k_-, \end{cases} \tag{20}$$

with Debye length

$$\lambda_{\mathrm{De}} = \left(\frac{\epsilon_0 T_\mathrm{e}}{\mathrm{e} n_\mathrm{e}}\right)^{1/2}, \tag{21}$$

classical distance of closest approach

$$b_0 = \frac{q_i q_k \mathrm{e}^2}{2\pi\epsilon_0 m_\mathrm{R} v_\mathrm{R}^2}, \tag{22}$$

reduced mass

$$m_{\mathrm{R}} = \frac{m_i m_k}{m_i + m_k}, \tag{23}$$

and the relative speed being approximated by the mean thermal speed

$$v_{\mathrm{R}} = \overline{v}_i. \tag{24}$$

The δ_{ik} term filters out self–collisions, which do not contribute to species diffusion

$$\delta_{ik} = \begin{cases} 1 & \text{for } i = k, \\ 0 & \text{for } i \neq k. \end{cases} \tag{25}$$

Finally, the free diffusion coefficient for positive ions is approximated by

$$D_{+}^{\mathrm{free}} = \overline{D_{i_+}^{\mathrm{free}}}. \tag{26}$$

Boundary sources

Each k-th species which is lost to the plasma boundary can get returned as k-th species, introducing the boundary sources

$$\left(\frac{\delta n_i}{\delta t}\right)_{\mathrm{diff}}^{\mathrm{in}} = -\sum_k r_{ik} \left(\frac{\delta n_k}{\delta t}\right)_{\mathrm{diff}}^{\mathrm{out}} = \sum_k \frac{D_k}{\Lambda^2} n_k s_k r_{ik} \tag{27}$$

Minimal Allowed Species Density

To prevent the ODE solver from *overshooting* into unphysical negative densities, an additional *artificial* term is added to the right-hand side of (1), ensuring a finite minimal value of particle densities (which can be considered zero). This correction term takes form of

$$\left(\frac{\delta n_i}{\delta t}\right)_{n_{\mathrm{min}}} = \begin{cases} \dfrac{n_{\mathrm{min}} - n_i}{\tau} & n_i < n_{\mathrm{min}}, \\ 0 & n_i \geq n_{\mathrm{min}}, \end{cases} \tag{28}$$

where n_{min} was set to 1 particle/m^3 and τ to 1.0×10^{-10} s.

Electron Energy Density Balance

$$\frac{\mathrm{d}\varrho_\mathrm{e}}{\mathrm{d}t} = \frac{P}{V\mathrm{e}} - \left(\frac{\delta\varrho_\mathrm{e}}{\delta t}\right)_\mathrm{el/inel} - \left(\frac{\delta\varrho_\mathrm{e}}{\delta t}\right)_\mathrm{gen/loss} - \left(\frac{\delta\varrho_\mathrm{e}}{\delta t}\right)_\mathrm{el\rightarrow walls} - \left(\frac{\delta\varrho_\mathrm{e}}{\delta t}\right)_\mathrm{ion\rightarrow walls} \tag{29}$$

Contribution of Elastic and Inelastic Collisions

This term describes energy loss due to electron collisions.

$$\left(\frac{\delta\varrho_\mathrm{e}}{\delta t}\right)_\mathrm{el/inel} = \sum_j R_j \Delta E_{\mathrm{e},j}, \tag{30}$$

with the electron energy loss for j-th reaction $\Delta E_{\mathrm{e},j}$ being

$$\Delta E_{\mathrm{e},j} = \begin{cases} \Delta E_{\mathrm{e},j}^\mathrm{inel} & \text{inelastic collisions,} \\ 3\dfrac{m_\mathrm{e}}{M_{\mathrm{cp},j}}(T_\mathrm{e} - T_\mathrm{g}) & \text{elastic collisions,} \\ 0 & \text{heavy species collisions or } a_{\mathrm{e}j}^R = 0, \end{cases} \tag{31}$$

and

$$T_\mathrm{e} = \frac{2}{3}\frac{\varrho_\mathrm{e}}{n_\mathrm{e}}. \tag{32}$$

The electron density n_e is not resolved explicitly, but rather calculated from plasma charge neutrality

$$n_\mathrm{e} = \sum_i n_i q_i. \tag{33}$$

For that reason, T_e might reach unphysically low values, when ϱ_e governed directly by (29) is much greater than $\sum_i n_i q_i$. This happens for example in a simulation of an afterglow, when after P is turned off, ϱ_e falls to zero value drastically fast, while n_e decrease is governed only by much slower diffusion and recombination processes. A correction is therefore introduced to T_e in the form of

$$T_\mathrm{e} = \max\left\{T_\mathrm{i}, \frac{2}{3}\frac{\varrho_\mathrm{e}}{n_\mathrm{e}}\right\} \tag{34}$$

Electron Generation and Loss Contribution

This term describes power dissipation associated with generation and loss of electrons.

$$\left(\frac{\delta\varrho_\mathrm{e}}{\delta t}\right)_\mathrm{gen/loss} = \frac{3}{2}T_\mathrm{e}\sum_j (a_{\mathrm{e}j}^R - a_{\mathrm{e}j}^L)R_j \tag{35}$$

Energy Loss by Electron Transport

Under a Maxwellian energy distribution assumption, each electron lost through the plasma boundary sheath takes away $2k_\mathrm{B}\mathrm{T}_\mathrm{e}$ of energy with it [4], which gives

$$\left(\frac{\delta\varrho_\mathrm{e}}{\delta t}\right)_{\mathrm{el}\to\mathrm{walls}} = -2T_\mathrm{e}\left(\frac{\delta n_\mathrm{e}}{\delta t}\right)_{\mathrm{walls}}, \tag{36}$$

while the total charge flux needs to be zero, yielding

$$\left(\frac{\delta n_\mathrm{e}}{\delta t}\right)_{\mathrm{walls}} = \sum_i q_i\left(\frac{\delta n_i}{\delta t}\right)_{\mathrm{diff}}. \tag{37}$$

Energy Loss by Ion Transport

If it is assumed, that ions leave the plasma boundary sheath with the Bohm velocity, each positive ion removed from the plasma takes away $\frac{1}{2}k_\mathrm{B}\mathrm{T}_\mathrm{e}$ of kinetic energy, as well as sheath voltage acceleration energy [4]

$$\left(\frac{\delta\varrho_\mathrm{e}}{\delta t}\right)_{\mathrm{ion}\to\mathrm{walls}} = -\frac{1}{2}T_\mathrm{e}\sum_{i_+}\left(\frac{\delta n_{i_+}}{\delta t}\right)_{\mathrm{diff}} - \overline{V_\mathrm{s}}\sum_{i_+} q_{i_+}\left(\frac{\delta n_{i_+}}{\delta t}\right)_{\mathrm{diff}}. \tag{38}$$

The last open parameter in the system is the mean sheath voltage $\overline{V_\mathrm{s}}$, which, according to [4], can be approximated by

$$\overline{V_\mathrm{s}} = T_\mathrm{e}\cdot\ln\left(\frac{\overline{M_{i_+}}}{2\pi m_\mathrm{e}}\right)^{1/2}. \tag{39}$$

This value of $\overline{V_\mathrm{s}}$ is only consistent with ICP plasma sources.

Minimal Allowed Electron Energy Density

To prevent the ODE solver from *overshooting* into unphysical negative ϱ_e, an additional *artificial* term is added to the right-hand side of (29), ensuring a finite minimal value of electron energy density (which can be considered zero). This correction term takes form of

$$\left(\frac{\delta\varrho_\mathrm{e}}{\delta t}\right)_{\varrho_\mathrm{min}} = \begin{cases}\dfrac{\varrho_\mathrm{e,min}-\varrho_\mathrm{e}}{\tau} & \varrho_\mathrm{e} < \varrho_\mathrm{e,min},\\ 0 & \varrho_\mathrm{e} \geq \varrho_\mathrm{e,min},\end{cases} \tag{40}$$

where $\varrho_\mathrm{e,min}$ was set to 1 eV/m^3 and τ to 1.0×10^{-10} s.

Full Equations

Particle Density Equation

$$\frac{\mathrm{d}n_i}{\mathrm{d}t} = \sum_j \left[(a_{ij}^R - a_{ij}^L) k_j \prod_l n_{lj}^L \right] + \frac{Q'_i}{V} - \frac{\sum Q'_i}{\sum n_{i_0}} \frac{n_{i_0}}{V} - \frac{p - p_0}{p_0} \frac{n_{i_0}}{\tau_p}$$
$$- \frac{D_i}{\Lambda^2} n_i s_i + \sum_k \frac{D_k}{\Lambda^2} n_k s_k r_{ik} + \frac{n_{\min} - n_{i_{n<n_{\min}}}}{\tau} \tag{41}$$

Electron Energy Density Equation

$$\frac{\mathrm{d}\varrho_\mathrm{e}}{\mathrm{d}t} = \frac{P}{V\mathrm{e}} - \sum_{j_\mathrm{inel}} R_j \Delta E_{\mathrm{e},j}^{\mathrm{inel}} - \sum_{j_\mathrm{el}} 3 \frac{m_\mathrm{e}}{M_{\mathrm{cp},j}} (T_\mathrm{e} - T_\mathrm{g}) R_j - \frac{3}{2} T_\mathrm{e} \sum_j (a_{\mathrm{e}j}^R - a_{\mathrm{e}j}^L) R_j$$
$$- 2T_\mathrm{e} \sum_i q_i \left(\frac{D_i}{\Lambda^2} n_i s_i - \sum_k \frac{D_k}{\Lambda^2} n_k s_k r_{ik} \right) - \frac{1}{2} T_\mathrm{e} \sum_{i_+} \left(\frac{D_i}{\Lambda^2} n_i s_i - \sum_k \frac{D_k}{\Lambda^2} n_k s_k r_{ik} \right)$$
$$- \overline{V_\mathrm{s}} \sum_{i_+} q_i \left(\frac{D_i}{\Lambda^2} n_i s_i - \sum_k \frac{D_k}{\Lambda^2} n_k s_k r_{ik} \right) + \frac{\varrho_{\mathrm{e,min}} - \varrho_{\mathrm{e},\varrho_\mathrm{e}<\varrho_{\mathrm{e,min}}}}{\tau} \tag{42}$$

References

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[3] C. Lee and M. A. Lieberman, "Global model of Ar, O2, Cl2, and Ar/O2 highdensity plasma discharges," *Journal of Vacuum Science & Technology A: Vacuum, Surfaces, and Films*, vol. 13, pp. 368–380, Mar. 1995.

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Quantemol Global Model is developed for people working with complex chemistries at the Quantemol Database (QDB).
QDB has been developed by Quantemol Ltd; to find out more please visit the Quantemol website at www.quantemol.com.

QDB supports and compares multiple data sets as well as providing services aiding plasma research.
Chemistry reaction sets can be used in several different plasma modelling software packages and optimised specifically for customer process conditions and requirements.
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