Read me for CASpy

February 3, 2023

Here, we explain the input file for CASpy in detail. For this purpose, we used the CO_2/MDEA/water system as a case study. The reactions (Reactions R1–R4 of the main text of the CASpy article) and the mass balance equations (Eqs. 11–14 of the main text of the CASpy article) involved in this system are shown in the Methods section of the main text. Note that the input file should be in the same directory with **main.py** and **functions.py** for solver to perform properly. An example input file for our solver is:

```
Temperature (K)
313.15
Number of Species
8
C0 (initial guess) / [mol/dm3]
1.0e-10 3.0e-10 1.0e-10 55.0638 2.500E-05 1.0e-10 1.0e-10 2.5
Names of species
HCO3- H3O+  CO3--  H2O  CO2  OH-  MDEAH+  MDEA
Charges
-1 1 -2 0 0 -1 1 0
Name of the solvent
H2O
Pure Density of the Solvent / [mol/dm3]
55.0638
mu^0 species / [kJ/mol] (only for the calculation of desired equilibrium constants)
0.0 -171.986 0.0 -858.154 0.0 0.0 -6272.15 -6661.2076
mu^ex species / [kJ/mol] (only for the calculation of desired equilibrium constants)
0.0 -779.679 0.0 -26.51 0.0 0.0 -553.792 -31.145
Impose Ptotal and gas composition? (T=True or F=False)
F
Ptotal / [kPa] (only used if Ptotal and gas composition is imposed)
0.0
Gas phase species
CO2
Gas phase composition (only used if Ptotal and gas composition is imposed)
1
mu^ex gases / [kJ/mol]
0.41013
Ctotal,gas / [mol/dm3] (Total concentration of the gases in liquid phase)
2.500E-05 4.059E-05 6.592E-05 1.070E-04 1.738E-04 2.822E-04 4.582E-04 7.441E-04
Number of Reactions
4
Stoichiometry
1 1 0 -2 -1 0 0 0
-1 1 1 -1 0 0 0 0
0 1 0 -1 0 0 -1 1
0 1 0 -2 0 1 0 0
ln(K) for reactions (if QMMC then computed using K_des expression)
```

```
-18.34 -27.55 QMMC -39.21
Number of mass balance equations (excluding charge neutrality)
3
Balances
1 3 5
7 8
2 4 6
```

The lines in the input file represent the following:

- `Temperature`: The absolute temperature.
- `Number of Species`: Number of species in the liquid phase.
- `C0 (initial guess)`: Initial guess of the composition in the liquid phase. This is a list of initial concentrations of species in $\mathrm{mol\,dm^{-3}}$. In our example, we used a lean solvent (only MDEA and water in the solution) as our initial guess. Note that none of the concentrations in the initial guess should be zero (due to the boundaries we use in our solver), instead, one can input a very low concentration. If any concentration in the initial guess is inputted zero or lower than zero (≤ 0), then it is changed by 10^{-10} $\mathrm{mol\,dm^{-3}}$. Although the solver works if the initial guess does not satisfy charge neutrality, we recommend an initial guess that satisfies charge neutrality for quicker results. The solver does not print a warning if charge neutrality is not satisfied by the initial guess and will continue to run.
- `Names of species`: Names of the species in the liquid phase.
- `Charges`: Net charges of the molecules/ions in the liquid phase.
- `Name of the solvent`: The name of the solvent.
- `Pure Density of the Solvent`: Density of the pure solvent in $\mathrm{mol\,dm^{-3}}$. The name of the solvent and the pure density of the solvent are used to compute the desired equilibrium constants of reactions using **??**.
- `mu^0 species`: A list of the values of μ_i^0 in $\mathrm{kJ\,mol^{-1}}$ for the species involved in reactions. Only used if the equilibrium constants are computed using the expression we derived for the mole fraction based equilibrium constants.
- `mu^ex species`: A list of the values of μ_i^{ex} in $\mathrm{kJ\,mol^{-1}}$ for the species involved in reactions. Only used if the equilibrium constants are computed using the expression we derived for the mole fraction based equilibrium constants.
- `Impose Ptotal and gas composition? (T=True or F=False)`: Are the total gas pressure and the gas composition imposed in the calculation? If True, the solver assumes an infinite gas phase and the speciations are

computed for all Ptotal (in kPa) listed in the next line. If the total gas pressure and gas composition are imposed, no mass balance equation is used for the species in the gas phase since there is mass transfer from the infinite gas phase to the liquid phase.

- `Ptotal / [kPa] (only used if Ptotal and gas composition is imposed)`: The list of total gas pressures. Used only if the total gas pressure and gas composition are imposed to compute the partial pressures of the species in the gas phase.
- `Gas phase species`: The names of the species in the gas phase.
- `Gas phase composition (only used if Ptotal and gas composition is imposed)`: The composition of the gas phase. The values in this list are normalized so the values sum up to 1.
- `mu^ex gases / [kJ/mol]`: The values of μ_i^{ex} for the gas phase species in the solvent.
- `Ctotal,gas / [mol/dm3] (Total concentration of the gases in liquid phase)`: A list of concentrations on the gas phase species in the liquid phase in $\mathrm{mol\,dm^{-3}}$. Only used if the total gas pressure and gas composition are not imposed.
- `Number of Reactions`: Number of reactions in the liquid phase.
- `Stoichiometry`: The stoichiometric coefficients of all species for each reaction.
- `ln(K) for reactions`: The mole fraction-based equilibrium constants for each reaction in the liquid phase. In case the desired equilibrium constant should be computed using **??**, the input should be "QMMC".
- `Number of mass balance equations`: Number of mass balance equations.
- `Balances`: A list of species involved in each mass balance equation. For example, the line "1 3 5" shows that the species at the first, third and fifth place in the names line ($\mathrm{HCO_3^-}$, $\mathrm{CO_3^{2-}}$, and $\mathrm{CO_2}$) are included in the first mass balance equation ($\mathrm{CO_2}$ balance).