令和４年度
原子力規制庁技術基盤グループ–原子力機構安全研究・防災支援部門
合同研究成果報告会

Analysis of mass transfer effect on chemically produced iodine release from aqueous phase

令和４年１１月２２日

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Iodine behavior under severe accident conditions

Due to its volatility and radiological impact, radioactive iodine is one of the major contributors to the source term during a severe accident in the nuclear power plant

CsI —(Dissolve in aqueous phase)→ I^- —(Radiation induced chemical reactions)→ I_2

Low volatility → High volatility

Risk of release to the environment

Fukushima Daiichi accident: aerosol and gaseous iodine species had spread throughout the vast area[1,2]

Proper modeling of iodine behavior under severe accident conditions is essential for the source term evaluation

Chemical processes + **Mass transfer**

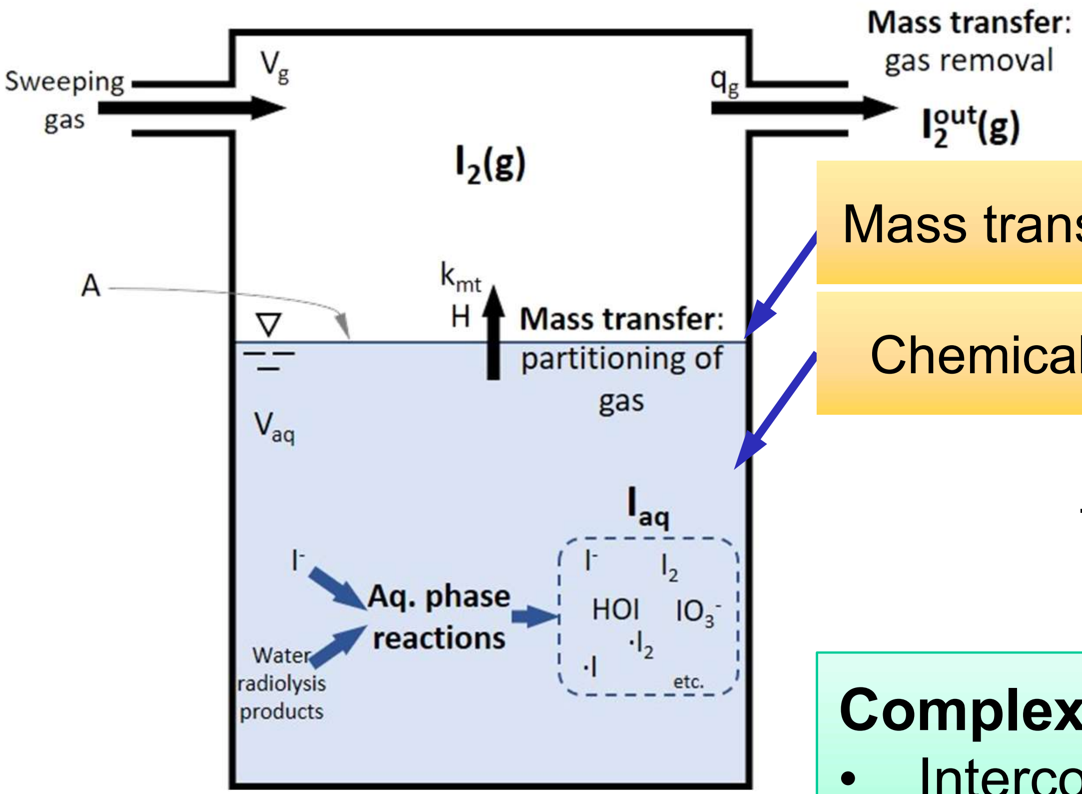
Chemical processes:
- Chemical kinetics model

Mass transfer:
- Re-volatilization through the gas-liquid interface
- Release to the environment

[1] L. S. Lebel, R. S. Dickson, and G. A. Glowa, J. Environ. Radioact. **151(**82) (2016).
[2] T. Ohkura *et al*, *JAEA-Data/Code 2012-010* (JAEA, 2012).

Iodine behavior modeling

The mass transfer coupling with chemical reaction kinetics



Mass transfer (two-film theory) — Described by k_{mt}

Chemical processes — Large **pH** impact[3,4]

$$\frac{dc_Z}{dt} = Dg_Z c_Z + \sum_n k_n c_X c_Y$$

~200 chemical reactions (LIRIC 3.2 [5])

Complex problem:
- Interconnected chemical reactions
- Large number of parameters

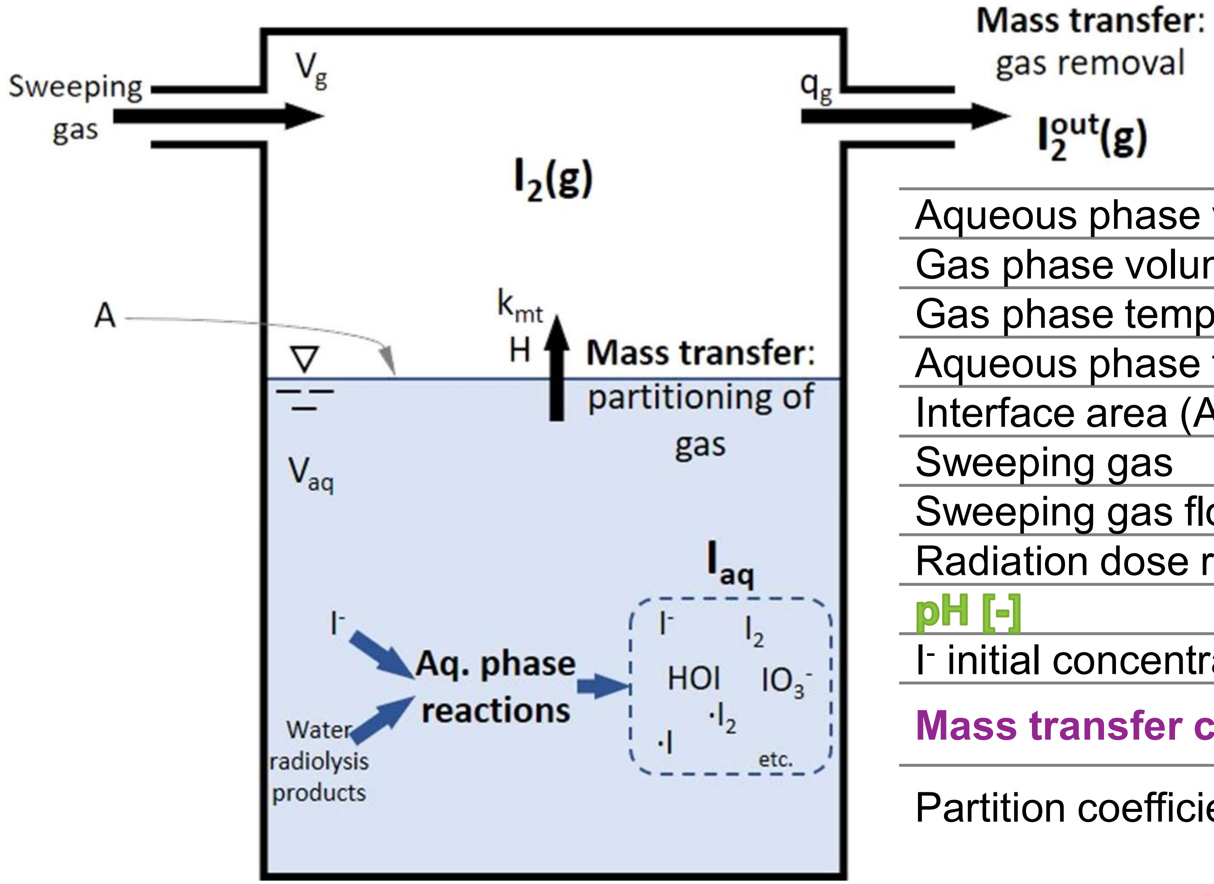
How significant are the separate contributions of chemical and mass transfer processes?

[3] C. B. Ashmore, J. R. Gwyther, and H. E. Sims, Nucl. Eng. Des. **166**(347) (1996).
[4] B. Clément, et al, NEA/CSNI/R(2007)1 (NEA, 2007).
[5] J. C. Wren and J. M. Ball, Radiat. Phys. Chem. **60**(577) (2001).

Analyzed system and used parameters

Mechanistic simulation code for kinetics of iodine chemistry KICHE[6] with Library of Iodine Reactions in Containment LIRIC 3.2 [5] reaction database was utilized to evaluate the time-dependent concentrations of iodine species

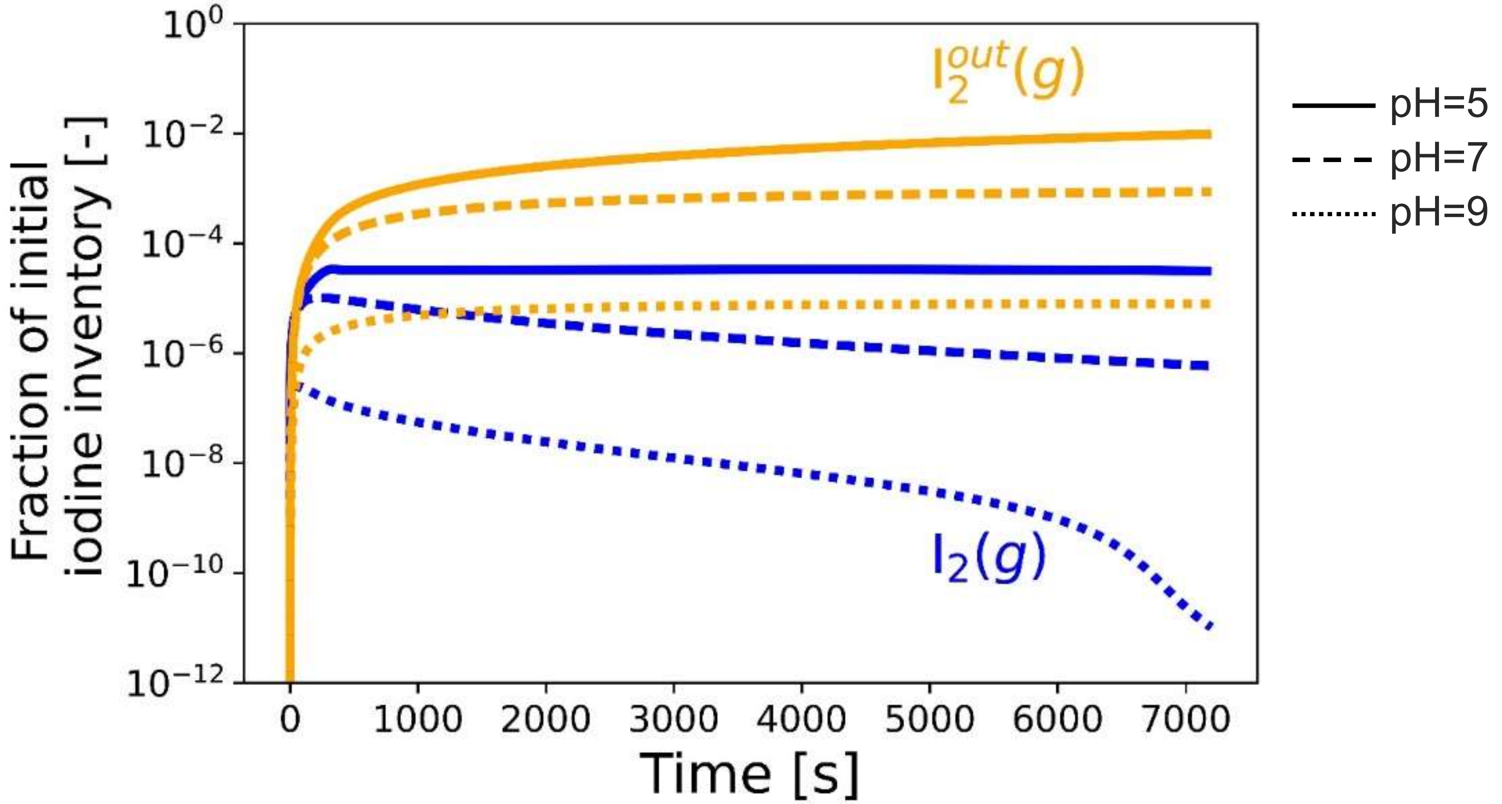
- Independent variables: **pH**, k_{mt}



Parameter	Value
Aqueous phase volume (V_{aq}) [m^3][7]	2980
Gas phase volume (V_g) [m^3][7]	3567.3
Gas phase temperature [K]	298.15
Aqueous phase temperature [K]	298.15
Interface area (A) [m^2][7]	934.5
Sweeping gas	N_2
Sweeping gas flow rate (q_g) [m^3/s]	300
Radiation dose rate (D) [kGy/h][8]	3
pH [-]	**3–14**
I^- initial concentration [mol/m^3][9]	$1.034\cdot10^{-2}$
Mass transfer coefficient for I_2 (k_{mt}) [m/s]	**10^{-7}–10^{-1}**
Partition coefficient for I_2 (H) at 298.15K [-]	71.318

[5] J. C. Wren and J. M. Ball, Radiat. Phys. Chem. **60**(577) (2001).
[6] K. Moriyama, et al, JAEA-Data/Code 2021-034 (JAEA, 2007).
[7] OECD/NEA, https://fdada.info/en/home2 (2016)
[8] J. Ishikawa, K. Kawaguchi, and Y. Maruyama, J Nucl Sci Technol **52**(308) (2015).
[9] A. L. Wright, NUREG-6193 (U.S. NRC, 1994).

Time-dependent concentrations of gaseous iodine



The amount of gaseous iodine that escapes from the aqueous phase decreases with increasing pH

- Clear pH effect on the chemical production of volatile iodine in the aqueous phase

Effective mass transfer coefficient $\tilde{k}_{mt}$

An analytical approach to perform the decomposition of contributions by the chemical and mass transfer processes by revising the two-film theory

Mass transfer contribution

$\tilde{k}_{mt}(\mathrm{pH}, k_{mt})$

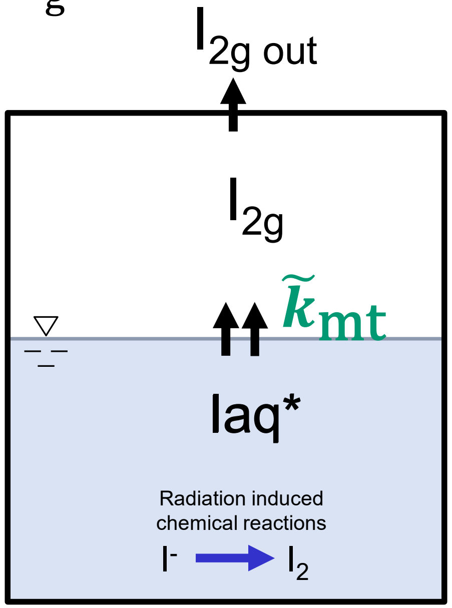
Chemical process contribution

KICHE results

$$\frac{d}{dt} f_{I_2(g)} = A\tilde{k}_{mt}\left[\frac{f_{I(aq)}}{V_{aq}} - \frac{H}{V_g} f_{I_2(g)}\right] - q_g \frac{f_{I_2(g)}}{V_g}$$

$$\frac{d}{dt} f_{I(aq)} = A\tilde{k}_{mt}\left[\frac{H}{V_g} f_{I_2(g)} - \frac{f_{I(aq)}}{V_{aq}}\right]$$

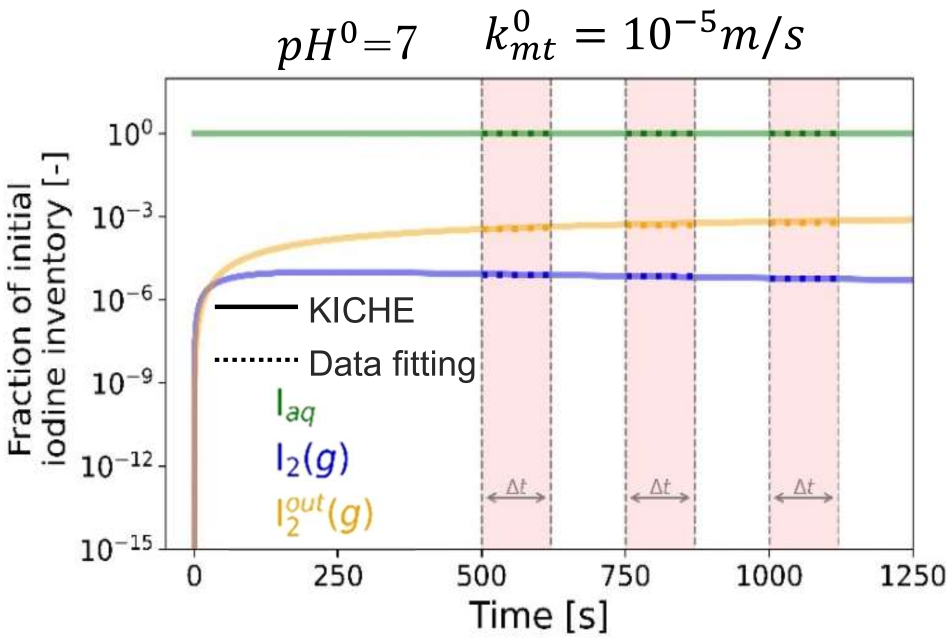
$$\frac{d}{dt} f_{I_2^{out}(g)} = q_g \frac{f_{I_2(g)}}{V_g}$$

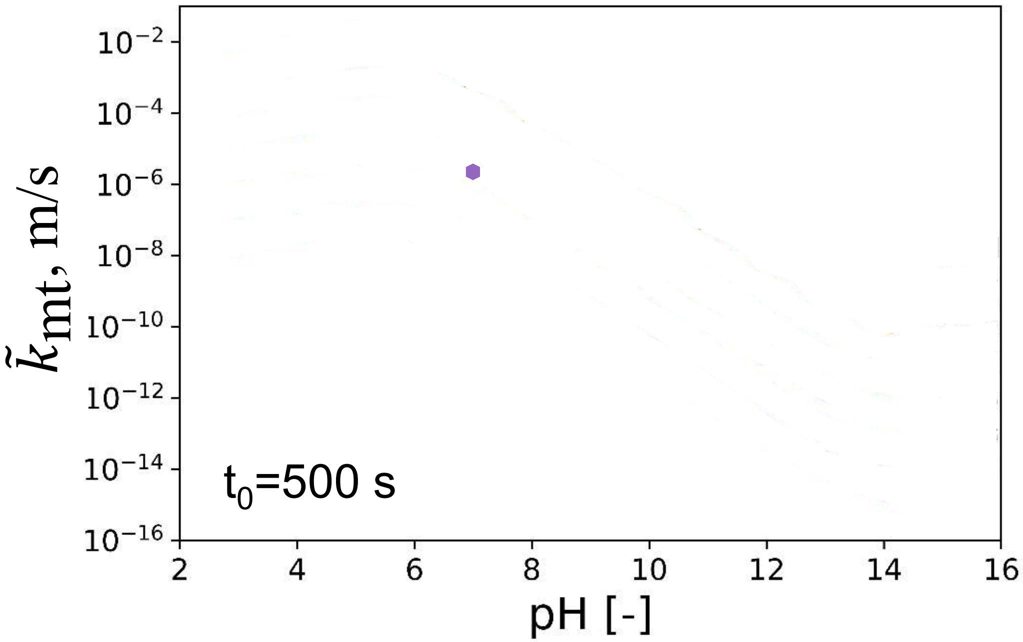


- ✓ Comprises contributions of both iodine chemistry in the aqueous phase and mass transfer
- ✓ Does not depend on iodine chemical state in aqueous phase

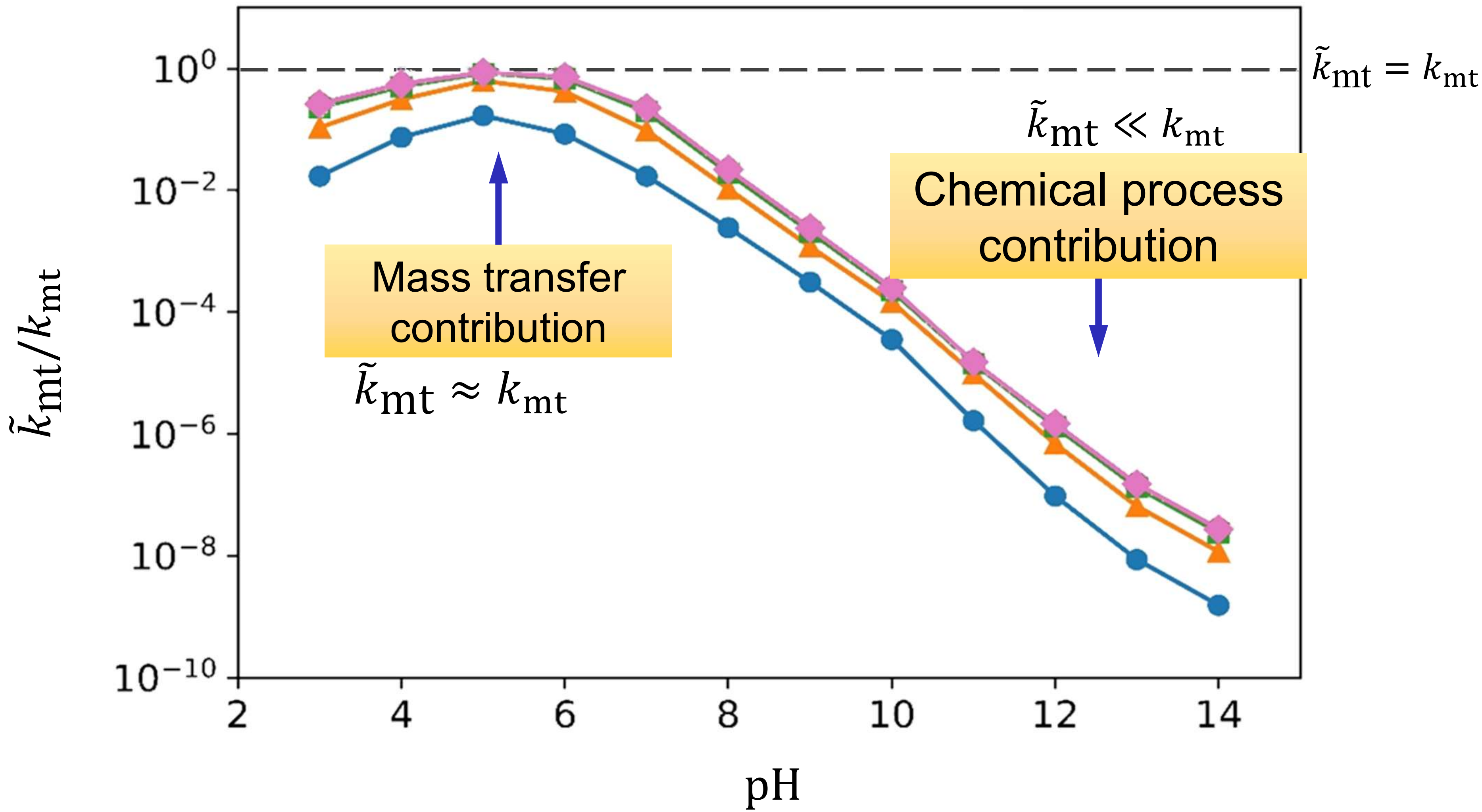
$\tilde{k}_{\mathrm{mt}}$ determination procedure

For obtaining $\tilde{k}_{\mathrm{mt}}$ least square fitting was applied on time-dependent iodine concentrations obtained by KICHE





$\tilde{k}_{mt}$ dependency on pH and k_{mt}

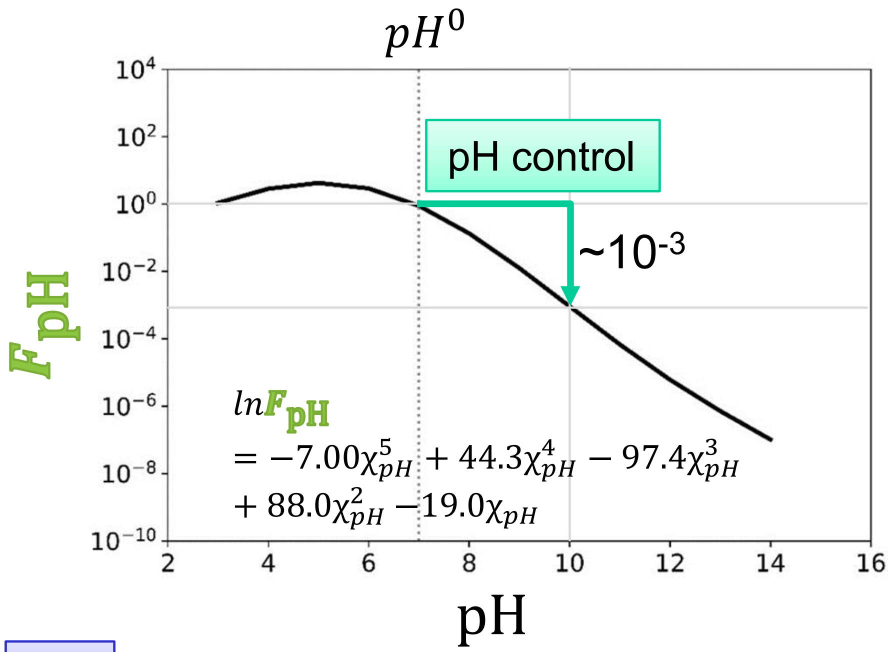


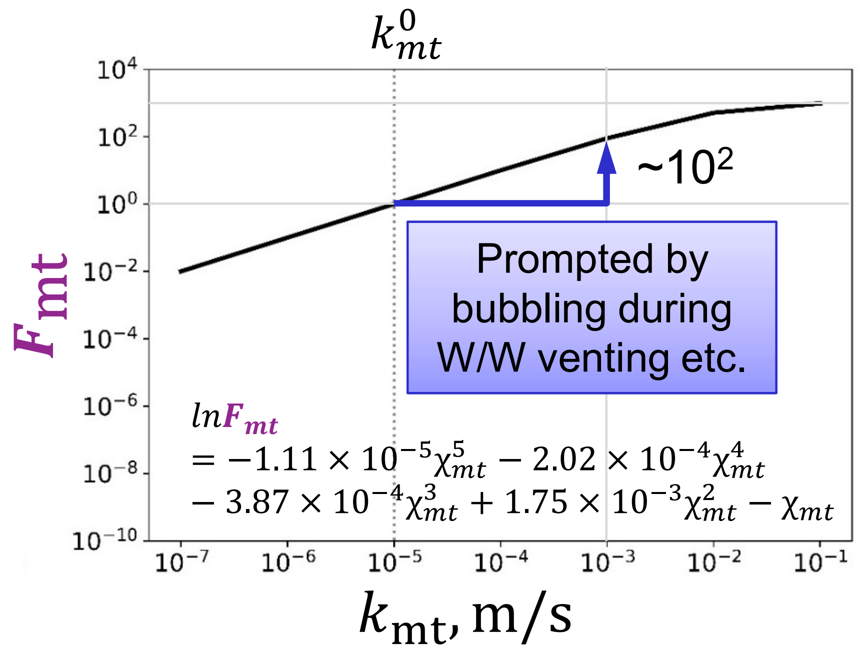
$\tilde{k}_{mt}$ decomposition into pH and k_{mt} contributions

Assumed as: $\tilde{k}_{mt}(\mathrm{pH}, k_{mt}) = F_{pH} F_{mt} \tilde{k}^0_{mt}$, where $\tilde{k}^0_{mt} = 2.256 \times 10^{-6} m/s$

$\ln(\tilde{k}_{mt}): R^2 = 0.999$ Expanded as 5th order polynomials

$\chi_{pH} = pH/pH^0 \qquad \chi_{mt} = ln(k_{mt}/k^0_{mt})$





! Concerning the source term evaluations, if the mass transfer is prompted, the amount of released iodine from the aqueous phase may increase even under achieved alkaline pH

Summary and conclusion

- Effective mass transfer coefficient $\tilde{k}_{mt}$ was introduced by revising the two-film theory for evaluating the contributions of chemical processes and mass transfer on iodine release from aqueous phase
- $\tilde{k}_{mt}$ was decomposed into contributions of chemical processes and mass transfer as functions of pH and k_{mt}

  - $\tilde{k}_{mt}$ enables to assess the significance of each process

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Concerning the source term evaluations, if the mass transfer is prompted, the amount of released iodine from the aqueous phase may increase even under achieved alkaline pH