Supplementary Material for

An analytical Method to Estimate Supersaturation in Gas-liquid Systems as a Function of Pressure-reduction Step and Waiting Time

Sushobhan Pradhan, Prem Bikkina*

School of Chemical Engineering, Oklahoma State University, Stillwater, OK 74078, USA

*Corresponding Author

Phone: +1-405-744-9112. Fax: +1-405-744-6338. Email: prem.bikkina@okstate.edu

Derivation of Supersaturation Equation

The Fick's second law of diffusion given below describes the one-dimensional diffusion equation for binary mixtures of species A and B under transient condition [14, 15]:

$$\frac{\partial C}{\partial t} = D_{AB}\frac{\partial^2 C}{\partial x^2} \tag{S1}$$

where D_{AB} is the diffusion coefficient of species A (gas) in species B (liquid), C is the solute concentration, t is the time, and x is the position.

$y = f(x,t)$ defined by the following dimensionless characteristic equation:

$$y = \frac{x}{2\sqrt{D_{AB}t}} \tag{S2}$$

$$\frac{\partial y}{\partial x} = \frac{1}{2\sqrt{D_{AB}t}}, \quad \text{and } \frac{\partial y}{\partial t} = \frac{-x}{4\sqrt{D_{AB}t^3}} \tag{S3}$$

$$\frac{\partial C}{\partial t} = \frac{\partial C}{\partial y} \times \frac{\partial y}{\partial t} \tag{S4}$$

From Equations (S3) and (S4):

$$\frac{\partial C}{\partial t} = \frac{-x}{4\sqrt{D_{AB}t^3}} \times \frac{\partial C}{\partial y} \tag{S5}$$

The $\frac{\partial^2 C}{\partial x^2}$ from Equation (S1) is determined by the following steps:

$$\frac{\partial^2 C}{\partial x^2} = \frac{\partial}{\partial x}\left(\frac{\partial C}{\partial x}\right) = \frac{\partial}{\partial x}\left[\frac{\partial C}{\partial y} \times \left(\frac{\partial y}{\partial x}\right)\right] = \frac{\partial}{\partial x}\left[\frac{\partial C}{\partial y} \times \frac{1}{2\sqrt{D_{AB}t}}\right] \tag{S6}$$

Assuming $\frac{\partial C}{\partial y} \times \frac{1}{2\sqrt{D_{AB}t}} = m$ (S7)

Therefore, $\frac{\partial^2 C}{\partial x^2} = \frac{\partial m}{\partial x}$ (S8)

Also, $\frac{\partial m}{\partial x} = \frac{\partial m}{\partial y} \times \frac{\partial y}{\partial x}$ (S9)

Now, $\frac{\partial m}{\partial y} = \left[\frac{\partial}{\partial y}\left\{\frac{\partial C}{\partial y} \times \frac{1}{2\sqrt{D_{AB}t}}\right\}\right] = \frac{1}{2\sqrt{D_{AB}t}} \times \frac{\partial^2 C}{\partial y^2}$ (S10)

From Eqn.(S3), $\frac{\partial y}{\partial x} = \frac{1}{2\sqrt{D_{AB}t}}$ (S11)

From Equations (S9)–(S11), we get:

$$\frac{\partial m}{\partial x} = \frac{\partial m}{\partial y} \times \frac{\partial y}{\partial x} = \left(\frac{1}{2\sqrt{D_{AB}t}} \times \frac{\partial^2 C}{\partial y^2}\right)\left(\frac{1}{2\sqrt{D_{AB}t}}\right) = \frac{1}{4D_{AB}t} \times \frac{\partial^2 C}{\partial y^2}$$

From eqn. (S8):

$$\frac{\partial^2 C}{\partial x^2} = \frac{\partial m}{\partial x} = \frac{\partial}{\partial x}\left[\frac{\partial C}{\partial y}\left(\frac{\partial y}{\partial x}\right)\right] = \frac{1}{4D_{AB}t} \times \frac{\partial^2 C}{\partial y^2}$$ (S12)

Substituting eqns. (S5) & (S12) in eqn. (S1) gives:

$$\frac{-x}{4\sqrt{D_{AB}t^3}} \times \frac{\partial C}{\partial y} = D_{AB} \times \frac{1}{4D_{AB}t} \times \frac{\partial^2 C}{\partial y^2}$$

$$\frac{\partial C}{\partial y} = \frac{-\sqrt{D_{AB}t}}{x} \times \frac{\partial^2 C}{\partial y^2}$$ (S13)

From eqn. (S2), we have:

$$y = \frac{x}{2\sqrt{D_{AB}t}}$$

$$\frac{\partial C}{\partial y} = \frac{-1}{2y} \times \frac{\partial^2 C}{\partial y^2}$$ (S14)

Let us assume: $n = \frac{\partial C}{\partial y}$ (S15)

Putting eqn. (S15) in eqn. (S14):

$$n = \frac{-1}{2y} \times \frac{\partial n}{\partial y}$$

$$-2\int y\,dy = \int \frac{1}{n}dn$$

$-y^2 = \ln n - \ln A$, where A is an integration constant

$$n = A\exp\,(-y^2) \tag{S16}$$

Applying eqn. (S15) in eqn. (S16):

$$\frac{dC}{dy} = A\exp\,(-y^2)$$

$$dC = A\exp\,(-y^2)\,dy \tag{S17}$$

The boundary conditions to solve the integral are given by:

$C_A = C_{Aeq}\ at\ y = 0$ (i. e., x = 0)

$C_A = C_{AS}\ at\ y = \infty$ (i. e., t = 0)

Integrating eqn. (S17) and applying the above boundary conditions will get:

$$\int_{C_{Aeq}}^{C_{AS}} dC = A\int_0^{\infty} \exp\,(-y^2)\,dy \tag{S18}$$

$$C_{AS} - C_{Aeq} = A\int_0^{\infty} \exp\,(-y^2)\,dy \tag{S19}$$

From the Gaussian integral [16], we have

$$\int_0^{\infty} \exp\,(-y^2)\,dy = \frac{\sqrt{\pi}}{2} \tag{S20}$$

From eqns. (S19) and (S20):

$$A = \frac{2}{\sqrt{\pi}} \times \left(C_{AS} - C_{Aeq}\right) \tag{S21}$$

The following error function and the boundary conditions were used to solve Fick's second law of diffusion [14]:

$$\text{erf}\ (y) = \frac{2}{\sqrt{\pi}} \int_0^y \exp\ (-y^2)\ dy \tag{S22}$$

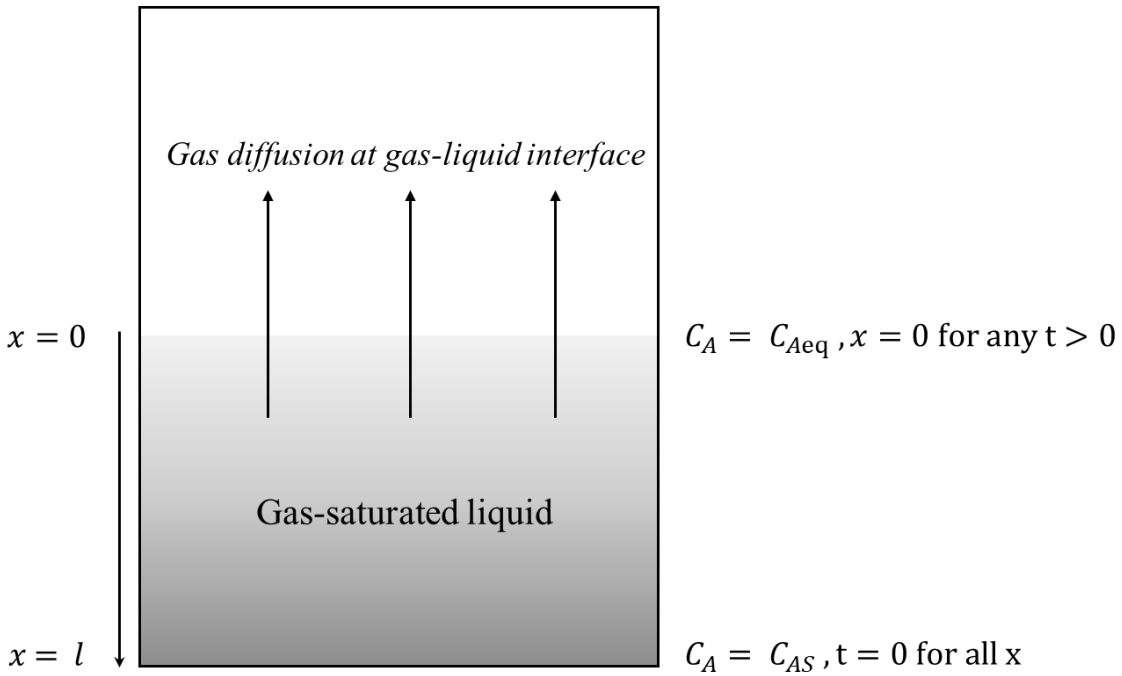

Figure S1: Schematic of bubble nucleation system used to study gas diffusion and saturation in a liquid

The boundary conditions for the gas-liquid system shown in Figure 1 are given below:

$C_A = C_{Aeq}$, at x = 0 and for any t

$C_A = C_{AS}$, at t = 0 and for all x

where, C_{AS} is the saturated gas concentration in mol/l (after complete saturation), C_A is the gas concentration after any time t, C_{Aeq} is the gas concentration at the gas-liquid interface at equilibrium (with the current gas pressure) in mol/l. As mentioned earlier, $y = f(x, t)$; therefore it is required to redefine x and y in Figure S1. In Figure S1, x is an independent variable and it is a positon in the liquid column, which is taken as 5 mm (total height of the liquid column) for estimating C_A in Tables S2 to S3 and S2 to S5. y is a dependent variable that varies with position in the liquid column, and waiting time after each pressure reduction step.

From Equation (S18), we have: $dC = A \exp(-y^2)\, dy$

Integrating the above equation and applying the boundary conditions, we get the following:

$$\int_{C_{Aeq}}^{C_A} dC = A \int_0^y \exp(-y^2)\, dy \tag{S23}$$

Applying Equations (S20) and (S21) in Equation (S19):

$$C_A - C_{Aeq} = \frac{2}{\sqrt{\pi}} \times \left(C_{AS} - C_{Aeq}\right) \times \frac{\sqrt{\pi}}{2} \times \mathrm{erf}(y)$$

$$\frac{C_A - C_{Aeq}}{C_{AS} - C_{Aeq}} = \mathrm{erf}\left(\frac{x}{2\sqrt{D_{AB}t}}\right) \tag{S24}$$

Equation (S24) is the solution to the problem for time-dependent diffusion assuming semi-infinite system.

For a given x, D_{AB}, and t, assuming $erf\left(\frac{x}{2\sqrt{D_{AB}t}}\right)$ as η, Equation (S24) reduces to:

$$\frac{C_A - C_{Aeq}}{C_{AS} - C_{Aeq}} = \eta \tag{S25}$$

Table S1: Saturation time calculations for carbon dioxide, methane and nitrogen gases in water

Type of gas	τ	D **(mm²/sec)**	l **(mm)**	t **(sec)**
Carbon dioxide	4	0.0016	5	62,500 (17.4 h)
Methane	4	0.00177	5	56,497 (15.7 h)
Nitrogen	4	0.00189	5	52,910 (14.7 h)

Table S2. Supersaturation and diffusion calculations for carbon dioxide with 500 mbar (50000 Pa) step-down pressure

Pressure (Pa)	**Equilibrium Concentration (C_{Aeq}) (mol/l)**	**Supersaturation ($C_{AS} - C_{Aeq}$) (mol/l)**	**Gas concentration (C_{A}) (mol/l)**	**Amount of gas diffused (ΔC) (mol/l)**
600,000	0.20133		0.20133	
550,000	0.18455	0.01678	0.20128	0.00005
500,000	0.16778	0.03350	0.20117	0.00011
450,000	0.15100	0.05017	0.20101	0.00016
400,000	0.13422	0.06679	0.20079	0.00021
350,000	0.11744	0.08335	0.20053	0.00027
300,000	0.10067	0.09986	0.20021	0.00032
250,000	0.08389	0.11632	0.19983	0.00037
200,000	0.06711	0.13272	0.19940	0.00043
150,000	0.05033	0.14907	0.19892	0.00048
100,000	0.03356	0.16537	0.19839	0.00053
50,000	0.01678	0.18162	0.19781	0.00058
0	0.00000	0.19781	0.19717	0.00064

Table S3. Supersaturation and diffusion calculations for carbon dioxide with 100 mbar (10000 Pa) step-down pressure

Pressure (Pa)	**Equilibrium Concentration (C_{Aeq}) (mol/l)**	**Supersaturation ($C_{AS} - C_{Aeq}$) (mol/l)**	**Gas concentration ($C_{A)}$ (mol/l)**	**Amount of gas diffused (ΔC) (mol/l)**
600,000	0.20133		0.20133	
590,000	0.19798	0.00336	0.20132	0.00001
580,000	0.19462	0.00670	0.20130	0.00002
570,000	0.19127	0.01003	0.20127	0.00003
560,000	0.18791	0.01336	0.20122	0.00004
550,000	0.18455	0.01667	0.20117	0.00005
540,000	0.18120	0.01997	0.20111	0.00006
530,000	0.17784	0.02326	0.20103	0.00007
520,000	0.17449	0.02654	0.20095	0.00009
510,000	0.17113	0.02981	0.20085	0.00010
500,000	0.16778	0.03307	0.20074	0.00011
490,000	0.16442	0.03632	0.20063	0.00012
480,000	0.16107	0.03956	0.20050	0.00013
470,000	0.15771	0.04279	0.20036	0.00014
460,000	0.15435	0.04601	0.20021	0.00015
450,000	0.15100	0.04922	0.20006	0.00016
440,000	0.14764	0.05241	0.19989	0.00017
430,000	0.14429	0.05560	0.19971	0.00018

Pressure (Pa)	**Equilibrium Concentration (C_{Aeq}) (mol/l)**	**Supersaturation ($C_{AS} - C_{Aeq}$) (mol/l)**	**Gas concentration ($C_{A)}$ (mol/l)**	**Amount of gas diffused (ΔC) (mol/l)**
420,000	0.14093	0.05878	0.19952	0.00019
410,000	0.13758	0.06194	0.19932	0.00020
400,000	0.13422	0.06510	0.19911	0.00021
390,000	0.13087	0.06825	0.19889	0.00022
380,000	0.12751	0.07138	0.19866	0.00023
370,000	0.12415	0.07451	0.19842	0.00024
360,000	0.12080	0.07762	0.19817	0.00025
350,000	0.11744	0.08073	0.19791	0.00026
340,000	0.11409	0.08383	0.19764	0.00027
330,000	0.11073	0.08691	0.19736	0.00028
320,000	0.10738	0.08999	0.19707	0.00029
310,000	0.10402	0.09305	0.19678	0.00030
300,000	0.10067	0.09611	0.19647	0.00031
290,000	0.09731	0.09916	0.19615	0.00032
280,000	0.09396	0.10219	0.19582	0.00033
270,000	0.09060	0.10522	0.19548	0.00034
260,000	0.08724	0.10824	0.19513	0.00035
250,000	0.08389	0.11124	0.19477	0.00036
240,000	0.08053	0.11424	0.19441	0.00037
230,000	0.07718	0.11723	0.19403	0.00038

Pressure (Pa)	**Equilibrium Concentration (C_{Aeq}) (mol/l)**	**Supersaturation ($C_{AS} - C_{Aeq}$) (mol/l)**	**Gas concentration ($C_{A)}$ (mol/l)**	**Amount of gas diffused (ΔC) (mol/l)**
220,000	0.07382	0.12021	0.19364	0.00039
210,000	0.07047	0.12318	0.19325	0.00040
200,000	0.06711	0.12614	0.19284	0.00041
190,000	0.06376	0.12909	0.19243	0.00042
180,000	0.06040	0.13203	0.19200	0.00042
170,000	0.05704	0.13496	0.19157	0.00043
160,000	0.05369	0.13788	0.19112	0.00044
150,000	0.05033	0.14079	0.19067	0.00045
140,000	0.04698	0.14369	0.19021	0.00046
130,000	0.04362	0.14659	0.18974	0.00047
120,000	0.04027	0.14947	0.18926	0.00048
110,000	0.03691	0.15235	0.18877	0.00049
100,000	0.03356	0.15521	0.18827	0.00050
90,000	0.03020	0.15807	0.18776	0.00051
80,000	0.02684	0.16092	0.18724	0.00052
70,000	0.02349	0.16375	0.18672	0.00053
60,000	0.02013	0.16658	0.18618	0.00054
50,000	0.01678	0.16940	0.18563	0.00054
40,000	0.01342	0.17221	0.18508	0.00055
30,000	0.01007	0.17501	0.18452	0.00056

Pressure (Pa)	**Equilibrium Concentration (C_{Aeq}) (mol/l)**	**Supersaturation ($C_{AS} - C_{Aeq}$) (mol/l)**	**Gas concentration ($C_{A)}$ (mol/l)**	**Amount of gas diffused (ΔC) (mol/l)**
20,000	0.00671	0.17781	0.18395	0.00057
10,000	0.00336	0.18059	0.18337	0.00058
0	0.00000	0.18337	0.18278	0.00059

Table S4. Supersaturation and diffusion calculations for methane gas with 500 mbar (50000 Pa) step-down pressure

Pressure (Pa)	**Equilibrium Concentration** (C_{Aeq}) **(mol/l)**	**Supersaturation** $(C_{AS} - C_{Aeq})$ **(mol/l)**	**Gas concentration** (C_{A}) **(mol/l)**	**Amount of gas diffused** (ΔC) **(mol/l)**
600000	0.00817		0.00817	
550000	0.00749	0.00068	0.00817	0.00000
500000	0.00681	0.00136	0.00816	0.00001
450000	0.00613	0.00203	0.00815	0.00001
400000	0.00545	0.00270	0.00814	0.00001
350000	0.00477	0.00337	0.00812	0.00002
300000	0.00409	0.00403	0.00810	0.00002
250000	0.00340	0.00469	0.00808	0.00002
200000	0.00272	0.00535	0.00805	0.00003
150000	0.00204	0.00601	0.00802	0.00003
100000	0.00136	0.00666	0.00798	0.00003
50000	0.00068	0.00730	0.00795	0.00004
0	0.00000	0.00795	0.00791	0.00004

Table S5. Supersaturation and diffusion calculations for methane with 100 mbar (10000 Pa) step-down pressure

Pressure (Pa)	Equilibrium Concentration (C_{Aeq}) (mol/l)	Supersaturation ($C_{AS} - C_{Aeq}$) (mol/l)	Gas concentration ($C_{A)}$ (mol/l)	Amount of gas diffused (ΔC) (mol/l)
600000	0.00817		0.00817	
590000	0.00804	0.00014	0.00817	0.00000
580000	0.00790	0.00027	0.00817	0.00000
570000	0.00776	0.00041	0.00817	0.00000
560000	0.00763	0.00054	0.00816	0.00000
550000	0.00749	0.00067	0.00816	0.00000
540000	0.00735	0.00081	0.00816	0.00000
530000	0.00722	0.00094	0.00815	0.00000
520000	0.00708	0.00107	0.00815	0.00001
510000	0.00695	0.00120	0.00814	0.00001
500000	0.00681	0.00133	0.00813	0.00001
490000	0.00667	0.00146	0.00813	0.00001
480000	0.00654	0.00159	0.00812	0.00001
470000	0.00640	0.00172	0.00811	0.00001
460000	0.00626	0.00184	0.00810	0.00001
450000	0.00613	0.00197	0.00809	0.00001

Pressure (Pa)	Equilibrium Concentration (C_{Aeq}) (mol/l)	Supersaturation ($C_{AS} - C_{Aeq}$) (mol/l)	Gas concentration ($C_{A)}$ (mol/l)	Amount of gas diffused (ΔC) (mol/l)
440000	0.00599	0.00210	0.00808	0.00001
430000	0.00586	0.00222	0.00807	0.00001
420000	0.00572	0.00235	0.00806	0.00001
410000	0.00558	0.00247	0.00804	0.00001
400000	0.00545	0.00260	0.00803	0.00001
390000	0.00531	0.00272	0.00802	0.00001
380000	0.00518	0.00284	0.00800	0.00001
370000	0.00504	0.00296	0.00799	0.00002
360000	0.00490	0.00308	0.00797	0.00002
350000	0.00477	0.00320	0.00796	0.00002
340000	0.00463	0.00332	0.00794	0.00002
330000	0.00449	0.00344	0.00792	0.00002
320000	0.00436	0.00356	0.00790	0.00002
310000	0.00422	0.00368	0.00788	0.00002
300000	0.00409	0.00380	0.00786	0.00002
290000	0.00395	0.00391	0.00784	0.00002
280000	0.00381	0.00403	0.00782	0.00002
270000	0.00368	0.00415	0.00780	0.00002

Pressure (Pa)	Equilibrium Concentration (C_{Aeq}) (mol/l)	Supersaturation ($C_{AS} - C_{Aeq}$) (mol/l)	Gas concentration ($C_{A)}$ (mol/l)	Amount of gas diffused (ΔC) (mol/l)
260000	0.00354	0.00426	0.00778	0.00002
250000	0.00340	0.00438	0.00776	0.00002
240000	0.00327	0.00449	0.00774	0.00002
230000	0.00313	0.00460	0.00771	0.00002
220000	0.00300	0.00472	0.00769	0.00002
210000	0.00286	0.00483	0.00766	0.00002
200000	0.00272	0.00494	0.00764	0.00003
190000	0.00259	0.00505	0.00761	0.00003
180000	0.00245	0.00516	0.00759	0.00003
170000	0.00232	0.00527	0.00756	0.00003
160000	0.00218	0.00538	0.00753	0.00003
150000	0.00204	0.00549	0.00750	0.00003
140000	0.00191	0.00560	0.00748	0.00003
130000	0.00177	0.00571	0.00745	0.00003
120000	0.00163	0.00581	0.00742	0.00003
110000	0.00150	0.00592	0.00739	0.00003
100000	0.00136	0.00603	0.00736	0.00003
90000	0.00123	0.00613	0.00733	0.00003

Pressure (Pa)	**Equilibrium Concentration (C_{Aeq}) (mol/l)**	**Supersaturation ($C_{AS} - C_{Aeq}$) (mol/l)**	**Gas concentration ($C_{A)}$) (mol/l)**	**Amount of gas diffused (ΔC) (mol/l)**
80000	0.00109	0.00624	0.00729	0.00003
70000	0.00095	0.00634	0.00726	0.00003
60000	0.00082	0.00644	0.00723	0.00003
50000	0.00068	0.00655	0.00720	0.00003
40000	0.00054	0.00665	0.00716	0.00003
30000	0.00041	0.00675	0.00713	0.00003
20000	0.00027	0.00685	0.00709	0.00003
10000	0.00014	0.00696	0.00706	0.00004
0	0.00000	0.00706	0.00702	0.00004

Table S6. Supersaturation and diffusion calculations for nitrogen with 500 mbar (50000 Pa) step-down pressure

Pressure (Pa)	**Equilibrium Concentration** (C_{Aeq}) **(mol/l)**	**Supersaturation** $(C_{AS} - C_{Aeq})$ **(mol/l)**	**Gas concentration** $(C_{A)}$ **(mol/l)**	**Amount of gas diffused** (ΔC) **(mol/l)**
600000	0.00361		0.00361	
550000	0.00331	0.00030	0.00361	0.00000
500000	0.00301	0.00060	0.00361	0.00000
450000	0.00271	0.00090	0.00360	0.00001
400000	0.00241	0.00119	0.00359	0.00001
350000	0.00211	0.00148	0.00358	0.00001
300000	0.00181	0.00178	0.00357	0.00001
250000	0.00151	0.00207	0.00356	0.00001
200000	0.00120	0.00235	0.00354	0.00002
150000	0.00090	0.00264	0.00352	0.00002
100000	0.00060	0.00292	0.00350	0.00002
50000	0.00030	0.00320	0.00348	0.00002
0	0.00000	0.00348	0.00346	0.00002

Table S7. Supersaturation and diffusion calculations for nitrogen with 100 mbar (10000 Pa) step-down pressure

Pressure (Pa)	**Equilibrium Concentration** (C_{Aeq}) **(mol/l)**	**Supersaturation** $(C_{AS} - C_{Aeq})$ **(mol/l)**	**Gas concentration** $(C_{A)}$ **(mol/l)**	**Amount of gas diffused** (ΔC) **(mol/l)**
600000	0.00361		0.00361	
590000	0.00355	0.00006	0.00361	0.00000
580000	0.00349	0.00012	0.00361	0.00000
570000	0.00343	0.00018	0.00361	0.00000
560000	0.00337	0.00024	0.00361	0.00000
550000	0.00331	0.00030	0.00361	0.00000
540000	0.00325	0.00036	0.00360	0.00000
530000	0.00319	0.00041	0.00360	0.00000
520000	0.00313	0.00047	0.00360	0.00000
510000	0.00307	0.00053	0.00359	0.00000
500000	0.00301	0.00058	0.00359	0.00000
490000	0.00295	0.00064	0.00359	0.00000
480000	0.00289	0.00070	0.00358	0.00000
470000	0.00283	0.00075	0.00358	0.00001
460000	0.00277	0.00081	0.00357	0.00001
450000	0.00271	0.00086	0.00357	0.00001

Pressure (Pa)	Equilibrium Concentration (C_{Aeq}) (mol/l)	Supersaturation ($C_{AS} - C_{Aeq}$) (mol/l)	Gas concentration ($C_{A)}$ (mol/l)	Amount of gas diffused (ΔC) (mol/l)
440000	0.00265	0.00092	0.00356	0.00001
430000	0.00259	0.00097	0.00355	0.00001
420000	0.00253	0.00102	0.00355	0.00001
410000	0.00247	0.00108	0.00354	0.00001
400000	0.00241	0.00113	0.00353	0.00001
390000	0.00235	0.00118	0.00352	0.00001
380000	0.00229	0.00124	0.00351	0.00001
370000	0.00223	0.00129	0.00351	0.00001
360000	0.00217	0.00134	0.00350	0.00001
350000	0.00211	0.00139	0.00349	0.00001
340000	0.00205	0.00144	0.00348	0.00001
330000	0.00199	0.00149	0.00347	0.00001
320000	0.00193	0.00154	0.00346	0.00001
310000	0.00187	0.00159	0.00345	0.00001
300000	0.00181	0.00164	0.00344	0.00001
290000	0.00175	0.00169	0.00342	0.00001
280000	0.00169	0.00174	0.00341	0.00001
270000	0.00163	0.00179	0.00340	0.00001

Pressure (Pa)	Equilibrium Concentration (C_{Aeq}) (mol/l)	Supersaturation ($C_{AS} - C_{Aeq}$) (mol/l)	Gas concentration ($C_{A)}$ (mol/l)	Amount of gas diffused (ΔC) (mol/l)
260000	0.00157	0.00184	0.00339	0.00001
250000	0.00151	0.00188	0.00338	0.00001
240000	0.00144	0.00193	0.00336	0.00001
230000	0.00138	0.00198	0.00335	0.00001
220000	0.00132	0.00203	0.00334	0.00001
210000	0.00126	0.00207	0.00332	0.00001
200000	0.00120	0.00212	0.00331	0.00001
190000	0.00114	0.00216	0.00329	0.00001
180000	0.00108	0.00221	0.00328	0.00001
170000	0.00102	0.00226	0.00326	0.00002
160000	0.00096	0.00230	0.00325	0.00002
150000	0.00090	0.00235	0.00323	0.00002
140000	0.00084	0.00239	0.00322	0.00002
130000	0.00078	0.00243	0.00320	0.00002
120000	0.00072	0.00248	0.00318	0.00002
110000	0.00066	0.00252	0.00317	0.00002
100000	0.00060	0.00256	0.00315	0.00002
90000	0.00054	0.00261	0.00313	0.00002

Pressure (Pa)	**Equilibrium Concentration** (C_{Aeq}) **(mol/l)**	**Supersaturation** $(C_{AS} - C_{Aeq})$ **(mol/l)**	**Gas concentration** $(C_{A)}$ **(mol/l)**	**Amount of gas diffused** (ΔC) **(mol/l)**
80000	0.00048	0.00265	0.00311	0.00002
70000	0.00042	0.00269	0.00310	0.00002
60000	0.00036	0.00273	0.00308	0.00002
50000	0.00030	0.00278	0.00306	0.00002
40000	0.00024	0.00282	0.00304	0.00002
30000	0.00018	0.00286	0.00302	0.00002
20000	0.00012	0.00290	0.00300	0.00002
10000	0.00006	0.00294	0.00298	0.00002
0	0.00000	0.00298	0.00296	0.00002

Table S8. CO_2 supersaturations generated in water with 500 mbar (50000 Pa) step-down pressure and different waiting times (saturation pressure: 6000 mbar or 600000 Pa)

Pressure (Pa)	Supersaturation after each waiting time (mol/l)			
	15 min	**30 min**	**45 min**	**60 min**
550000	0.01678	0.01678	0.01678	0.01678
500000	0.03350	0.03293	0.03206	0.03119
450000	0.05017	0.04848	0.04599	0.04358
400000	0.06679	0.06346	0.05868	0.05423
350000	0.08335	0.07787	0.07024	0.06337
300000	0.09986	0.09175	0.08077	0.07123
250000	0.11632	0.10511	0.09036	0.07799
200000	0.13272	0.11798	0.09910	0.08379
150000	0.14907	0.13037	0.10707	0.08878
100000	0.16537	0.14229	0.11432	0.09306
50000	0.18162	0.15377	0.12093	0.09675
0	0.19781	0.16483	0.12696	0.09991

Table S9. CO_2 supersaturations generated in water with 100 mbar (10000 Pa) step-down pressure and different waiting times (saturation pressure: 6000 mbar or 600000 Pa)

Pressure (Pa)	Supersaturation after each waiting time (mol/l)			
	15 min	30 min	45 min	60 min
590000	0.00336	0.00336	0.00336	0.00336
580000	0.00670	0.00659	0.00641	0.00624
570000	0.01003	0.00970	0.00920	0.00872
560000	0.01336	0.01269	0.01174	0.01085
550000	0.01667	0.01557	0.01405	0.01267
540000	0.01997	0.01835	0.01615	0.01425
530000	0.02326	0.02102	0.01807	0.01560
520000	0.02654	0.02360	0.01982	0.01676
510000	0.02981	0.02607	0.02141	0.01776
500000	0.03307	0.02846	0.02286	0.01861
490000	0.03632	0.03075	0.02419	0.01935
480000	0.03956	0.03297	0.02539	0.01998
470000	0.04279	0.03509	0.02649	0.02053
460000	0.04601	0.03714	0.02749	0.02099
450000	0.04922	0.03912	0.02840	0.02139
440000	0.05241	0.04102	0.02923	0.02174
430000	0.05560	0.04284	0.02998	0.02204
420000	0.05878	0.04461	0.03067	0.02229

Pressure (Pa)	Supersaturation after each waiting time (mol/l)			
	15 min	30 min	45 min	60 min
410000	0.06194	0.04630	0.03130	0.02251
400000	0.06510	0.04793	0.03187	0.02270
390000	0.06825	0.04950	0.03239	0.02286
380000	0.07138	0.05102	0.03287	0.02300
370000	0.07451	0.05247	0.03330	0.02312
360000	0.07762	0.05388	0.03369	0.02322
350000	0.08073	0.05523	0.03405	0.02331
340000	0.08383	0.05653	0.03438	0.02338
330000	0.08691	0.05778	0.03468	0.02345
320000	0.08999	0.05898	0.03495	0.02351
310000	0.09305	0.06014	0.03520	0.02355
300000	0.09611	0.06126	0.03542	0.02359
290000	0.09916	0.06234	0.03563	0.02363
280000	0.10219	0.06337	0.03581	0.02366
270000	0.10522	0.06437	0.03598	0.02369
260000	0.10824	0.06533	0.03614	0.02371
250000	0.11124	0.06625	0.03628	0.02373
240000	0.11424	0.06714	0.03641	0.02375
230000	0.11723	0.06800	0.03653	0.02376
220000	0.12021	0.06882	0.03663	0.02377

Pressure (Pa)	Supersaturation after each waiting time (mol/l)			
	15 min	30 min	45 min	60 min
210000	0.12318	0.06962	0.03673	0.02378
200000	0.12614	0.07038	0.03682	0.02379
190000	0.12909	0.07112	0.03690	0.02380
180000	0.13203	0.07182	0.03697	0.02381
170000	0.13496	0.07251	0.03704	0.02381
160000	0.13788	0.07316	0.03710	0.02382
150000	0.14079	0.07380	0.03716	0.02382
140000	0.14369	0.07440	0.03721	0.02382
130000	0.14659	0.07499	0.03726	0.02383
120000	0.14947	0.07556	0.03730	0.02383
110000	0.15235	0.07610	0.03734	0.02383
100000	0.15521	0.07662	0.03737	0.02383
90000	0.15807	0.07713	0.03740	0.02384
80000	0.16092	0.07761	0.03743	0.02384
70000	0.16375	0.07808	0.03746	0.02384
60000	0.16658	0.07853	0.03748	0.02384
50000	0.16940	0.07896	0.03750	0.02384
40000	0.17221	0.07938	0.03752	0.02384
30000	0.17501	0.07978	0.03754	0.02384
20000	0.17781	0.08016	0.03756	0.02384

Pressure (Pa)	Supersaturation after each waiting time (mol/l)			
	15 min	30 min	45 min	60 min
10000	0.18059	0.08053	0.03757	0.02384
0	0.18337	0.08089	0.03759	0.02384

Table S10. CH_4 supersaturations generated in water with 500 mbar (50000 Pa) step-down pressure and different waiting times (saturation pressure: 6000 mbar or 600000 Pa)

Pressure (Pa)	Supersaturation after each waiting time (mol/l)			
	15 min	**30 min**	**45 min**	**60 min**
550000	0.00068	0.00068	0.00068	0.00068
500000	0.00136	0.00133	0.00129	0.00125
450000	0.00203	0.00195	0.00183	0.00173
400000	0.00270	0.00254	0.00232	0.00213
350000	0.00337	0.00310	0.00276	0.00247
300000	0.00403	0.00363	0.00315	0.00275
250000	0.00469	0.00414	0.00349	0.00299
200000	0.00535	0.00462	0.00381	0.00319
150000	0.00601	0.00508	0.00408	0.00335
100000	0.00666	0.00552	0.00433	0.00349
50000	0.00730	0.00594	0.00455	0.00361
0	0.00795	0.00634	0.00475	0.00371

Table S11. CH_4 supersaturations generated in water with 100 mbar (10000 Pa) step-down pressure and different waiting times (saturation pressure: 6000 mbar or 600000 Pa)

Pressure (Pa)	Supersaturation after each waiting time (mol/l)			
	15 min	30 min	45 min	60 min
590000	0.00014	0.00014	0.00014	0.00014
580000	0.00027	0.00027	0.00026	0.00025
570000	0.00041	0.00039	0.00037	0.00035
560000	0.00054	0.00051	0.00046	0.00043
550000	0.00067	0.00062	0.00055	0.00049
540000	0.00081	0.00073	0.00063	0.00055
530000	0.00094	0.00083	0.00070	0.00060
520000	0.00107	0.00092	0.00076	0.00064
510000	0.00120	0.00102	0.00082	0.00067
500000	0.00133	0.00110	0.00087	0.00070
490000	0.00146	0.00119	0.00091	0.00072
480000	0.00159	0.00127	0.00095	0.00074
470000	0.00172	0.00134	0.00099	0.00076
460000	0.00184	0.00142	0.00102	0.00077
450000	0.00197	0.00148	0.00105	0.00078
440000	0.00210	0.00155	0.00107	0.00079
430000	0.00222	0.00161	0.00109	0.00080
420000	0.00235	0.00167	0.00112	0.00081

Pressure (Pa)	Supersaturation after each waiting time (mol/l)			
	15 min	30 min	45 min	60 min
410000	0.00247	0.00173	0.00113	0.00081
400000	0.00260	0.00178	0.00115	0.00082
390000	0.00272	0.00183	0.00116	0.00082
380000	0.00284	0.00188	0.00118	0.00083
370000	0.00296	0.00193	0.00119	0.00083
360000	0.00308	0.00197	0.00120	0.00083
350000	0.00320	0.00202	0.00121	0.00083
340000	0.00332	0.00206	0.00122	0.00084
330000	0.00344	0.00209	0.00122	0.00084
320000	0.00356	0.00213	0.00123	0.00084
310000	0.00368	0.00217	0.00124	0.00084
300000	0.00380	0.00220	0.00124	0.00084
290000	0.00391	0.00223	0.00125	0.00084
280000	0.00403	0.00226	0.00125	0.00084
270000	0.00415	0.00229	0.00125	0.00084
260000	0.00426	0.00232	0.00126	0.00084
250000	0.00438	0.00234	0.00126	0.00084
240000	0.00449	0.00237	0.00126	0.00084
230000	0.00460	0.00239	0.00127	0.00084
220000	0.00472	0.00241	0.00127	0.00084

Pressure (Pa)	Supersaturation after each waiting time (mol/l)			
	15 min	30 min	45 min	60 min
210000	0.00483	0.00243	0.00127	0.00084
200000	0.00494	0.00245	0.00127	0.00084
190000	0.00505	0.00247	0.00127	0.00084
180000	0.00516	0.00249	0.00128	0.00084
170000	0.00527	0.00251	0.00128	0.00084
160000	0.00538	0.00253	0.00128	0.00084
150000	0.00549	0.00254	0.00128	0.00084
140000	0.00560	0.00256	0.00128	0.00084
130000	0.00571	0.00257	0.00128	0.00084
120000	0.00581	0.00259	0.00128	0.00084
110000	0.00592	0.00260	0.00128	0.00084
100000	0.00603	0.00261	0.00128	0.00084
90000	0.00613	0.00262	0.00128	0.00084
80000	0.00624	0.00263	0.00128	0.00084
70000	0.00634	0.00264	0.00128	0.00084
60000	0.00644	0.00265	0.00128	0.00084
50000	0.00655	0.00266	0.00128	0.00084
40000	0.00665	0.00267	0.00128	0.00084
30000	0.00675	0.00268	0.00128	0.00084
20000	0.00685	0.00269	0.00129	0.00084

Pressure (Pa)	Supersaturation after each waiting time (mol/l)			
	15 min	**30 min**	**45 min**	**60 min**
10000	0.00696	0.00270	0.00129	0.00084
0	0.00706	0.00271	0.00129	0.00084

Table S12. N_2 supersaturations generated in water with 500 mbar (50000 Pa) step-down pressure and different waiting times (saturation pressure: 6000 mbar or 600000 Pa)

Pressure (Pa)	Supersaturation after each waiting time (mol/l)			
	15 min	30 min	45 min	60 min
550000	0.00030	0.00030	0.00030	0.00030
500000	0.00060	0.00059	0.00057	0.00055
450000	0.00090	0.00085	0.00080	0.00075
400000	0.00119	0.00111	0.00101	0.00092
350000	0.00148	0.00135	0.00119	0.00106
300000	0.00178	0.00157	0.00135	0.00118
250000	0.00207	0.00179	0.00149	0.00127
200000	0.00235	0.00199	0.00162	0.00135
150000	0.00264	0.00218	0.00173	0.00141
100000	0.00292	0.00236	0.00183	0.00147
50000	0.00320	0.00253	0.00191	0.00151
0	0.00348	0.00269	0.00199	0.00155

Table S13. N_2 supersaturations generated in water with 100 mbar (10000 Pa) step-down pressure and different waiting times (saturation pressure: 6000 mbar or 600000 Pa)

Pressure (Pa)	Supersaturation after each waiting time (mol/l)			
	15 min	30 min	45 min	60 min
590000	0.00006	0.00006	0.00006	0.00006
580000	0.00012	0.00012	0.00011	0.00011
570000	0.00018	0.00017	0.00016	0.00015
560000	0.00024	0.00022	0.00020	0.00018
550000	0.00030	0.00027	0.00024	0.00021
540000	0.00036	0.00031	0.00027	0.00024
530000	0.00041	0.00036	0.00030	0.00025
520000	0.00047	0.00040	0.00032	0.00027
510000	0.00053	0.00044	0.00035	0.00028
500000	0.00058	0.00047	0.00037	0.00029
490000	0.00064	0.00051	0.00038	0.00030
480000	0.00070	0.00054	0.00040	0.00031
470000	0.00075	0.00057	0.00041	0.00032
460000	0.00081	0.00060	0.00042	0.00032
450000	0.00086	0.00063	0.00043	0.00032
440000	0.00092	0.00065	0.00044	0.00033
430000	0.00097	0.00067	0.00045	0.00033
420000	0.00102	0.00070	0.00046	0.00033

Pressure (Pa)	Supersaturation after each waiting time (mol/l)			
	15 min	30 min	45 min	60 min
410000	0.00108	0.00072	0.00046	0.00033
400000	0.00113	0.00074	0.00047	0.00034
390000	0.00118	0.00076	0.00048	0.00034
380000	0.00124	0.00078	0.00048	0.00034
370000	0.00129	0.00079	0.00048	0.00034
360000	0.00134	0.00081	0.00049	0.00034
350000	0.00139	0.00083	0.00049	0.00034
340000	0.00144	0.00084	0.00049	0.00034
330000	0.00149	0.00085	0.00049	0.00034
320000	0.00154	0.00087	0.00050	0.00034
310000	0.00159	0.00088	0.00050	0.00034
300000	0.00164	0.00089	0.00050	0.00034
290000	0.00169	0.00090	0.00050	0.00034
280000	0.00174	0.00091	0.00050	0.00034
270000	0.00179	0.00092	0.00050	0.00034
260000	0.00184	0.00093	0.00050	0.00034
250000	0.00188	0.00094	0.00051	0.00034
240000	0.00193	0.00095	0.00051	0.00034
230000	0.00198	0.00096	0.00051	0.00034
220000	0.00203	0.00096	0.00051	0.00034

Pressure (Pa)	Supersaturation after each waiting time (mol/l)			
	15 min	30 min	45 min	60 min
210000	0.00207	0.00097	0.00051	0.00034
200000	0.00212	0.00098	0.00051	0.00034
190000	0.00216	0.00098	0.00051	0.00034
180000	0.00221	0.00099	0.00051	0.00034
170000	0.00226	0.00099	0.00051	0.00034
160000	0.00230	0.00100	0.00051	0.00034
150000	0.00235	0.00101	0.00051	0.00034
140000	0.00239	0.00101	0.00051	0.00034
130000	0.00243	0.00101	0.00051	0.00034
120000	0.00248	0.00102	0.00051	0.00034
110000	0.00252	0.00102	0.00051	0.00034
100000	0.00256	0.00103	0.00051	0.00034
90000	0.00261	0.00103	0.00051	0.00034
80000	0.00265	0.00103	0.00051	0.00034
70000	0.00269	0.00104	0.00051	0.00034
60000	0.00273	0.00104	0.00051	0.00034
50000	0.00278	0.00104	0.00051	0.00034
40000	0.00282	0.00104	0.00051	0.00034
30000	0.00286	0.00105	0.00051	0.00034
20000	0.00290	0.00105	0.00051	0.00034

Pressure (Pa)	Supersaturation after each waiting time (mol/l)			
	15 min	**30 min**	**45 min**	**60 min**
10000	0.00294	0.00105	0.00051	0.00034
0	0.00298	0.00105	0.00051	0.00034