Development of a surrogate artificial neural network for microkinetic modeling: case study with the methanol synthesis

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Electronic Supplementary Information (ESI)

S1. Microkinetic model

Table S1. Three-site field extended reaction mechanism for the methanol synthesis and the water-gas shift reaction over Cu (211) and Cu/Zn (211), thermodynamically consistent. (a): Cu (211), (b): Cu/Zn (211), (c): special Cu (211) site for hydrogen and water adsorption. Reproduced with permission from Campos et al.[1] Copyright 2021 Royal Society of Chemistry.

No.	Reaction	Forward reaction			Reverse reaction		
		$\Delta S^{\neq,+} \cdot 10^3$ [kJ·(mol·K)$^{-1}$]	E_A^+ [kJ·mol^{-1}]	β^+ [-]	$\Delta S^{\neq,-} \cdot 10^3$ [kJ·(mol·K)$^{-1}$]	E_A^- [kJ·mol^{-1}]	β^- [-]
R1	$H_{2(g)} + 2 \cdot (c) \rightleftarrows 2 \cdot H_{(c)}$	-119.24	69.57	0.000	1.72	93.01	0.000
R2	$CO_{(g)} + (a) \rightleftarrows CO_{(a)}$	-158.23	0.00	0.000	0.00	58.37	0.000
R3	$CO_{(g)} + (b) \rightleftarrows CO_{(b)}$	-151.60	8.90	-0.119	-6.64	0.00	0.119
R4	$CO_{2(g)} + (a) \rightleftarrows CO_{\mathbf{2}(a)}$	-144.74	0.00	-0.129	-7.22	52.29	0.129
R5	$CO_{2(g)} + (b) \rightleftarrows CO_{\mathbf{2}(b)}$	-138.11	0.00	-0.249	-13.86	51.59	0.249
R6	$H_3CO_{(a)} + H_{(c)} \rightleftarrows CH_3OH_{(g)} + (a) + (c)$	56.99	81.22	-0.547	-181.26	32.93	0.547
R7	$H_3CO_{(b)} + H_{(c)} \rightleftarrows CH_3OH_{(g)} + (b) + (c)$	47.03	92.56	-0.368	-171.30	23.61	0.368
R8	$H_2O_{(g)} + (c) \rightleftarrows H_2O_{(c)}$	-177.19	0.00	0.378	21.08	31.21	-0.378
R9	$H_2O_{(c)} + (c) \rightleftarrows OH_{(c)} + H_{(c)}$	5.99	79.84	0.140	21.60	105.17	-0.140
R10	$CO_{(a)} + H_{(c)} \rightleftarrows HCO_{(a)} + (c)$	25.38	84.62	-0.299	-21.90	20.55	0.299
R11	$HCO_{(a)} + H_{(c)} \rightleftarrows H_2CO_{(a)} + (c)$	15.73	56.26	-0.299	-8.10	92.81	0.299
R12	$CO_{\mathbf{2}(a)} + H_{(c)} \rightleftarrows HCOO_{(a)} + (c)$	36.44	77.74	-0.249	8.72	109.64	0.249
R13	$CO_{\mathbf{2}(b)} + H_{(c)} \rightleftarrows HCOO_{(b)} + (c)$	43.07	60.24	-0.368	2.08	105.43	0.368
R14	$HCOO_{(a)} + H_{(c)} \rightleftarrows HCOOH_{(a)} + (c)$	10.42	128.23	0.000	-153.70	29.81	0.000
R15	$HCOO_{(b)} + H_{(c)} \rightleftarrows HCOOH_{(b)} + (c)$	10.42	136.53	0.000	-153.70	7.72	0.000
R16	$HCOOH_{(a)} + H_{(c)} \rightleftarrows H_2COOH_{(a)} + (c)$	-150.42	46.89	0.000	-25.96	62.43	0.000
R17	$HCOOH_{(b)} + H_{(c)} \rightleftarrows H_2COOH_{(b)} + (c)$	-150.42	1.54	0.000	-25.96	48.92	0.000
R18	$H_2COOH_{(a)} + (c) \rightleftarrows H_2CO_{(a)} + OH_{(c)}$	-20.59	20.52	-0.249	-15.21	38.77	0.249
R19	$H_2COOH_{(b)} + (c) \rightleftarrows H_2CO_{(b)} + OH_{(c)}$	-13.95	11.22	-0.368	-21.85	9.96	0.368
R20	$H_2CO_{(a)} + H_{(c)} \rightleftarrows H_3CO_{(a)} + (c)$	42.42	52.66	-0.547	-13.47	107.12	0.547
R21	$H_2CO_{(b)} + H_{(c)} \rightleftarrows H_3CO_{(b)} + (c)$	32.46	51.46	-0.368	-3.51	132.06	0.368
R22	$CO_{(a)} + OH_{(c)} \rightleftarrows COOH_{(a)} + (c)$	-11.31	79.90	-0.119	-3.74	16.05	0.119
R23	$CO_{(b)} + OH_{(c)} \rightleftarrows COOH_{(b)} + (c)$	-11.31	13.33	-0.119	-3.74	16.05	0.119
R24	$COOH_{(a)} + OH_{(c)} \rightleftarrows CO_{\mathbf{2}(a)} + H_2O_{(c)}$	9.92	17.09	-0.119	-22.36	60.82	0.119
R25	$COOH_{(b)} + OH_{(c)} \rightleftarrows CO_{\mathbf{2}(b)} + H_2O_{(c)}$	9.92	17.09	-0.119	-22.37	60.82	0.119
	If the CO_2/CO_X ratio in feed is higher than 0.65, the activation energies of reactions 24 and 25 are adjusted:						
R24	$COOH_{(a)} + OH_{(c)} \rightleftarrows CO_{\mathbf{2}(a)} + H_2O_{(c)}$	9.92	**32.53**	-0.119	-22.36	**76.22**	0.119
R25	$COOH_{(b)} + OH_{(c)} \rightleftarrows CO_{\mathbf{2}(b)} + H_2O_{(c)}$	9.92	**32.53**	-0.119	-22.37	**76.22**	0.119

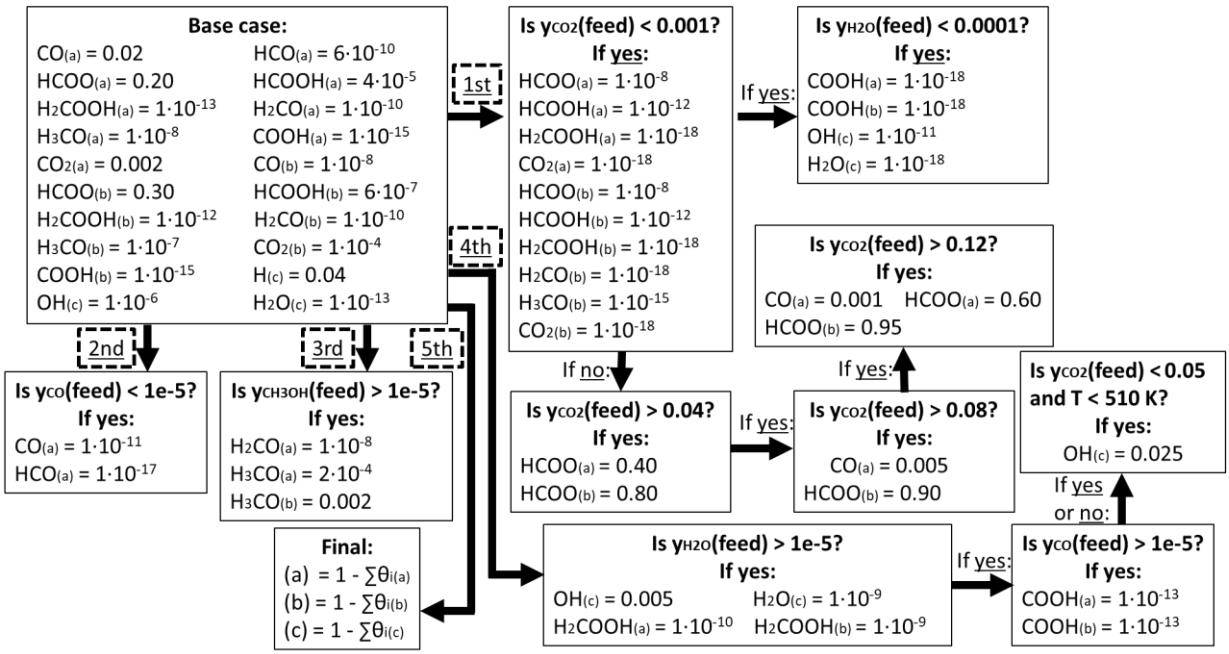

Figure S1. Initial guess method: Finding an educated initial guess to calculate the coverages. Reproduced with permission from Campos et al.[1] Copyright 2021 Royal Society of Chemistry.

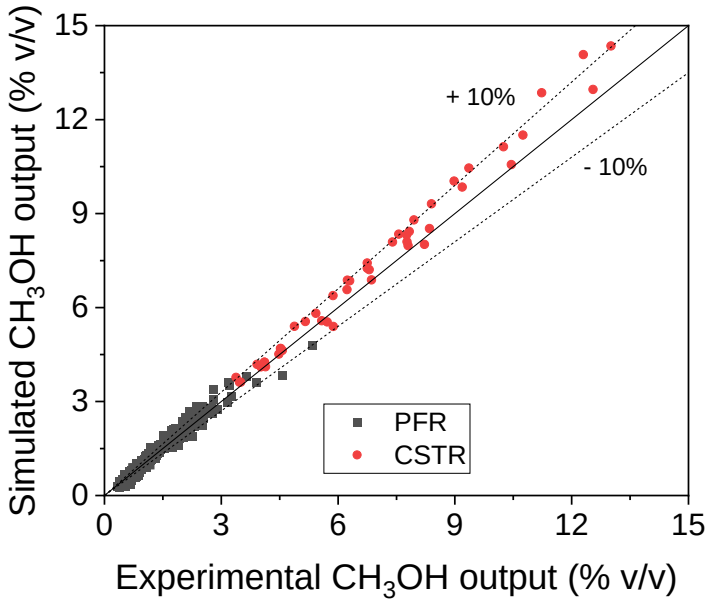

Figure S2. Parity plot of the methanol output concentration (% v/v) from steady-state experiments of the methanol synthesis performed in a PFR[1] and in a CSTR.[2] MKM simulations vs. experimental data.

S2. Expliciting the kinetic and thermodynamic terms in a microkinetic model

To develop the artificial neural network (ANN) in this work, one of the main assumptions was that the rate of a global reaction (r) consists in a kinetic term (r^K) and a thermodynamic term (r^T) which are multiplying each other. That is:

$$r = r^K \cdot r^T \tag{S1}$$

While in formal kinetic models these terms are easily identified in the equations, they can also be separated in microkinetic models containing a single global reaction. Let's take a hypothetical system as an example: A adsorbs (R1), forms B (R2), which forms C (R3), and desorbs (R4).

$$A_{(g)} + * \overset{\text{R1}}{\rightleftarrows} A^* \overset{\text{R2}}{\rightleftarrows} B^* \overset{\text{R3}}{\rightleftarrows} C^* \overset{\text{R4}}{\rightleftarrows} C_{(g)} + * \tag{S2}$$

Where * denotes a free active site.

The following rate equations can be written:

$$r_1 = k_1^+ \cdot f_A \cdot \theta_* - k_1^- \cdot \theta_{A*} \tag{S3}$$

$$r_2 = k_2^+ \cdot \theta_{A*} - k_2^- \cdot \theta_{B*} \tag{S4}$$

$$r_3 = k_3^+ \cdot \theta_{B*} - k_3^- \cdot \theta_{C*} \tag{S5}$$

$$r_4 = k_4^+ \cdot \theta_{C*} - k_4^- \cdot f_C \cdot \theta_* \tag{S6}$$

Here, k is the reaction constant, f_j is the fugacity of gas species j (in bar), θ_i is the coverage of surface species i, and the superscripts + and – refer to forward and reverse reaction, respectively.

The equilibrium constant of an elementary step (K) is defined as follows:

$$K = \frac{k^+}{k^-} \qquad \rightarrow \qquad k^- = \frac{k^+}{K} \tag{S7}$$

Considering that the catalyst surface is in steady-state, all four elementary steps must have the same rate. That is:

$$r_1 = r_2 = r_3 = r_4 = r \tag{S8}$$

Substituting Eq. (S7) and (S8) into Eqs. (S3-S6):

$$r = k_1^+ \cdot f_A \cdot \theta_* - \frac{k_1^+}{K_1} \cdot \theta_{A*} \tag{S9}$$

$$r = k_2^+ \cdot \theta_{A*} - \frac{k_2^+}{K_2} \cdot \theta_{B*} \tag{S10}$$

$$r = k_3^+ \cdot \theta_{B*} - \frac{k_3^+}{K_3} \cdot \theta_{C*} \tag{S11}$$

$$r = k_4^+ \cdot \theta_{C*} - \frac{k_4^+}{K_4} \cdot f_C \cdot \theta_* \tag{S12}$$

Isolating θ_{A*} from Eq. (S9):

$$\theta_{A*} = K_1 \cdot f_A \cdot \theta_* - \frac{K_1}{k_1^+} \cdot r \tag{S13}$$

Isolating θ_{B*} from Eq. (S10) and substituting θ_{A*} from Eq. (S13):

$$\theta_{B*} = K_1 \cdot K_2 \cdot f_A \cdot \theta_* - r \cdot \left(\frac{K_1 \cdot K_2}{k_1^+} + \frac{K_2}{k_2^+} \right) \tag{S14}$$

Isolating θ_{C*} from Eq. (S11) and substituting θ_{B*} from Eq. (S14):

$$\theta_{C*} = K_1 \cdot K_2 \cdot K_3 \cdot f_A \cdot \theta_* - r \cdot \left(\frac{K_1 \cdot K_2 \cdot K_3}{k_1^+} + \frac{K_2 \cdot K_3}{k_2^+} + \frac{K_3}{k_3^+} \right) \tag{S15}$$

Substituting Eq. (S15) in Eq. (S12):

$$r = k_4^+ \cdot \left[K_1 \cdot K_2 \cdot K_3 \cdot f_A \cdot \theta_* - r \cdot \left(\frac{K_1 \cdot K_2 \cdot K_3}{k_1^+} + \frac{K_2 \cdot K_3}{k_2^+} + \frac{K_3}{k_3^+} \right) \right] - \frac{k_4^+}{K_4} \cdot f_C \cdot \theta_* \tag{S16}$$

Multiplying both sides by K_4/k_4^+ and grouping the terms with r on the left side:

$$r \cdot \left(\frac{K_1 \cdot K_2 \cdot K_3 \cdot K_4}{k_1^+} + \frac{K_2 \cdot K_3 \cdot K_4}{k_2^+} + \frac{K_3 \cdot K_4}{k_3^+} + \frac{K_4}{k_4^+} \right) = K_1 \cdot K_2 \cdot K_3 \cdot K_4 \cdot f_A \cdot \theta_* - f_C \cdot \theta_* \tag{S17}$$

The equilibrium constant of the global reaction $A \rightleftarrows C$ (K_P^0) is the product of the elementary equilibrium constants:

$$K_P^0 = K_1 \cdot K_2 \cdot K_3 \cdot K_4 \tag{S18}$$

Substituting Eq. (S18) in Eq. (S17):

$$r \cdot \left(\frac{K_P^0}{k_1^+} + \frac{K_2 \cdot K_3 \cdot K_4}{k_2^+} + \frac{K_3 \cdot K_4}{k_3^+} + \frac{K_4}{k_4^+} \right) = K_P^0 \cdot f_A \cdot \theta_* - f_C \cdot \theta_* \tag{S19}$$

Isolating r and reorganizing:

$$r = \frac{K_P^0 \cdot f_A \cdot \theta_*}{\left(\frac{K_P^0}{k_1^+} + \frac{K_2 \cdot K_3 \cdot K_4}{k_2^+} + \frac{K_3 \cdot K_4}{k_3^+} + \frac{K_4}{k_4^+} \right)} \cdot \left(1 - \frac{f_C}{f_A \cdot K_P^0} \right) \tag{S20}$$

Here, we can define k as the effective global reaction constant:

$$k = \frac{K_P^0}{\left(\frac{K_P^0}{k_1^+} + \frac{K_2 \cdot K_3 \cdot K_4}{k_2^+} + \frac{K_3 \cdot K_4}{k_3^+} + \frac{K_4}{k_4^+} \right)} \tag{S21}$$

Finally, substituting Eq. (S21) in Eq. (S20):

$$r = [k \cdot f_A \cdot \theta_*] \cdot \left(1 - \frac{f_C}{f_A \cdot K_P^0}\right) \tag{S22}$$

In Eq. (S22), the kinetic term (r^K) and thermodynamic term (r^T) are now identifiable:

$$r^K = \left[k \cdot f_A \cdot \theta_*\right] \tag{S23}$$

$$r^T = \left(1 - \frac{f_C}{f_A \cdot K_P^0}\right) \tag{S24}$$

The majority of the reactive systems have multiple global reactions, in which this explicit separation of r^K and r^T is not possible, as an algebraic system with multiple variables would be reached. Still, the concept of Eq. (S1) should still be true, and one can calculate the kinetic term indirectly by dividing the reaction rate with the thermodynamic part, i.e.:

$$r^K = \frac{r}{r^T} \tag{S25}$$

S3. Calculation of the hydrogen equilibrium conversion

At chemical equilibrium in the methanol synthesis, the following equations must hold:[1, 3-4]

$$K^0_{P,CO\ hyd.} = T^{-3.384} \cdot \exp(10092.4 \cdot T^{-1} - 4.200) = \frac{f_{CH_3OH}}{f^2_{H_2} \cdot f_{CO}} \quad \text{(S26)}$$

$$K^0_{P,CO_2\ hyd.} = T^{-4.481} \cdot \exp(4755.7 \cdot T^{-1} + 8.369) = \frac{f_{CH_3OH} \cdot f_{H_2O}}{f^3_{H_2} \cdot f_{CO_2}} \quad \text{(S27)}$$

$$K^0_{P,rWGSR} = T^{-1.097} \cdot \exp(-5337.4 \cdot T^{-1} + 12.569) = \frac{f_{CO} \cdot f_{H_2O}}{f_{H_2} \cdot f_{CO_2}} \quad \text{(S28)}$$

Since this system of reactions has only two degrees of freedom, two reactions are sufficient to describe it. To calculate H_2 equilibrium conversion, we chose the hydrogenation of CO and the hydrogenation of CO_2. The reaction extensions are defined as follows, with CO and CO_2 as references (X_{CO} and X_{CO_2}):

$$X_{CO} = \frac{\dot{n}^{in}_{CO} - \dot{n}_{CO}}{\dot{n}^{in}_{CO}} \quad \text{(S29)}$$

$$X_{CO_2} = \frac{\dot{n}^{in}_{CO_2} - \dot{n}_{CO_2}}{\dot{n}^{in}_{CO_2}} \quad \text{(S30)}$$

Where $\dot{n}^{in}_{CO}$ and $\dot{n}^{in}_{CO_2}$ are the inlet mole flow of CO and CO_2, respectively, and $\dot{n}_{CO}$ and $\dot{n}_{CO_2}$ are the mole flow of CO and CO_2 after the reaction.

The mole flow of each component after the reaction is calculated according to its respective stoichiometric coefficients and the reaction extensions, as follows:

$$\dot{n}_{H_2} = \dot{n}^{in}_{H_2} - 2 \cdot X_{CO} \cdot \dot{n}^{in}_{CO} - 3 \cdot X_{CO_2} \cdot \dot{n}^{in}_{CO_2} \quad \text{(S31)}$$

$$\dot{n}_{CO} = \dot{n}^{in}_{CO} \cdot (1 - X_{CO}) \quad \text{(S32)}$$

$$\dot{n}_{CO_2} = \dot{n}^{in}_{CO_2} \cdot (1 - X_{CO_2}) \quad \text{(S33)}$$

$$\dot{n}_{CH_3OH} = \dot{n}^{in}_{CH_3OH} + X_{CO} \cdot \dot{n}^{in}_{CO} + X_{CO_2} \cdot \dot{n}^{in}_{CO_2} \quad \text{(S34)}$$

$$\dot{n}_{H_2O} = \dot{n}^{in}_{H_2O} + X_{CO_2} \cdot \dot{n}^{in}_{CO_2} \quad \text{(S35)}$$

The mole fraction of each component (y_j) after the reaction is calculated as follows:

$$y_j = \frac{\dot{n}_j}{\dot{n}} \quad \text{(S36)}$$

$$\dot{n} = \sum_{j=1}^{5} \dot{n}_j \tag{S37}$$

Here, $\dot{n}$ is the total mole flow after the reaction.

With the information of the total pressure, the reaction temperature, and the mole fractions, the fugacity of the components (f_j) were estimated with the Peng-Robinson equation of state.[5] Binary interaction parameters and other necessary information were taken from literature,[6-7] including an effective hydrogen acentric factor of −0.05.[8]

The aforementioned equations (Eqs. S26-S27, S29-S37, and the Peng-Robinson equations) form a non-linear algebraic system with two variables (X_{CO}, X_{CO_2}). This system is solved with the Matlab function *fsolve*, with the function and step tolerances set to 10^{-6}. With the final $X_{CO,eq}$ and $X_{CO_2,eq}$, the equilibrium H_2 conversion ($X_{H_2,eq}$) is calculated as follows:

$$X_{H_2,eq} = \frac{\left(\dot{n}_{H_2}^{in} - 2 \cdot X_{CO,eq} \cdot \dot{n}_{CO}^{in} - 3 \cdot X_{CO_2,eq} \cdot \dot{n}_{CO_2}^{in}\right)}{\dot{n}_{H_2}^{in}} \tag{S38}$$

S4. Parameters of the developed ANNs

Table S2. ANN_{10} parameters of the hidden layer (p_{wj}). w refers to the cell index while j refers to the input index.

p_{wj}	$j = 0$	$j = 1$	$j = 2$	$j = 3$	$j = 4$	$j = 5$	$j = 6$
$w = 1$	-0.9716898	0.0041664	-0.0061756	0.0100854	-0.0101530	0.0003118	-0.2135094
$w = 2$	-0.9959330	0.0240174	0.0051838	-0.0024755	-0.3549400	0.0002915	-0.0028441
$w = 3$	-0.9748353	0.0192092	0.0140785	-0.0291657	-0.0107911	0.0007449	-0.0138335
$w = 4$	-0.9605484	0.0259080	0.0053966	0.0075902	-0.0970261	0.0068462	-0.0081281
$w = 5$	0.9384540	-0.0468252	-0.0046727	-0.0112876	0.1626274	-0.0126964	0.0115955
$w = 6$	0.8449822	-0.0413623	0.0264285	-0.0291187	0.0202620	0.0017597	-0.3306908
$w = 7$	-1.0459629	0.0638154	0.0261562	-0.0030565	0.0170041	-0.0035466	-0.0047106
$w = 8$	0.8054555	-0.0534534	0.0582256	0.5045525	-0.0113396	-0.0154450	-0.9923904
$w = 9$	0.9736794	-0.0229409	0.0358447	-0.0001882	-0.0047470	0.0027775	-0.0288284
$w = 10$	0.9566228	-0.0345442	-0.0065016	-0.0005030	0.0005019	0.0001845	0.0061815

Table S3. ANN_{10} parameters of the output layer ($\hat{p}_{nw}$). n refers to the output index while w refers to the cell index. The table was transposed because of space.

$\hat{p}_{nw}{}'$	$n = 1$	$n = 2$	$n = 3$
$w = 0$	11068.3626	-94142.7103	-29624.1371
$w = 1$	709.9192	-282.5021	-56164.3427
$w = 2$	649.2352	-101175.0807	4744.4682
$w = 3$	-15995.9726	-3.6427	1353.2367
$w = 4$	34472.1413	5405.3482	18589.0573
$w = 5$	5102.2267	998.5848	2790.5874
$w = 6$	-4.3013	2.0209	-152.1544
$w = 7$	577.4606	11890.9206	1277.1276
$w = 8$	0.7117	-0.2048	13.0161
$w = 9$	7507.0901	12992.4854	-5848.5115
$w = 10$	-3262.2452	-4016.0001	2619.7030

Table S4. ANN_{20} parameters of the hidden layer (p_{wj}). w refers to the cell index while j refers to the input index.

p_{wj}	$j = 0$	$j = 1$	$j = 2$	$j = 3$	$j = 4$	$j = 5$	$j = 6$
$w = 1$	0.4107634	-0.3476371	-0.0343069	-0.4910167	0.3421876	0.0587555	0.3425156
$w = 2$	0.4788869	-0.5265364	-0.0705651	0.0468037	-0.0430378	0.0112581	0.0226196
$w = 3$	0.5235582	-0.0012607	0.1937560	0.4779385	0.1020426	-0.0414237	-1.3779806
$w = 4$	-0.4101937	0.0879208	-0.1416665	-0.0958177	1.1418159	-0.0091950	0.0577283
$w = 5$	1.0299699	-0.0610526	-0.0215066	0.0006823	-0.0074083	-0.0004268	0.0119220
$w = 6$	-0.0667126	-0.0922277	-0.1715939	-0.0862231	0.5347287	-0.0446638	0.9247602
$w = 7$	0.4684067	-0.0953478	0.1615514	0.1012457	-1.3064510	0.0100775	-0.0753646
$w = 8$	-0.9865289	0.0214754	0.0044386	-0.0040830	-0.8306544	-0.0012574	0.0120926
$w = 9$	0.1672123	-0.1619341	0.0224200	0.0914151	-0.1196893	0.0058689	0.0063903
$w = 10$	0.1316965	-0.5871169	0.6962471	0.1545037	-1.0756671	0.1769251	-0.2042572
$w = 11$	-0.9663492	0.0023345	-0.0048524	0.0006847	-0.0033376	0.0003150	-0.6501090
$w = 12$	0.2377170	-0.1813779	0.0092137	-0.3371499	0.2234223	0.0387758	0.1925004
$w = 13$	-0.9496994	0.0345588	-0.0588820	-0.0009962	-0.0107111	0.0057568	0.0347929
$w = 14$	-0.0548285	-0.0008430	0.1472877	-0.1680355	0.0992056	0.0230020	-0.6062469
$w = 15$	-0.2002659	0.0226395	0.0513328	0.2348420	0.0364116	-0.0330605	-1.0627367
$w = 16$	-1.0033942	0.0376572	0.0057459	0.0014855	-0.1792533	0.0078282	-0.0097843
$w = 17$	-0.1562959	0.0498811	0.0054537	0.0517360	0.0080279	-0.0135540	-0.9118531
$w = 18$	0.0752436	0.0109804	0.0557756	-0.2336342	0.0568813	0.0205622	-0.1238210
$w = 19$	0.9272197	0.0160861	0.0387003	-0.0102710	0.0799524	-0.0009957	-0.1313046
$w = 20$	0.2602767	-0.0104805	-0.0757907	-0.3215158	-0.0402635	0.0363001	1.0610396

Table S5. ANN_{20} parameters of the output layer ($\hat{p}_{nw}$). n refers to the output index while w refers to the cell index. The table was transposed because of space.

$\hat{p}_{nw}{}'$	$n = 1$	$n = 2$	$n = 3$
$w = 0$	-3192.6495	-48814.6635	-23609.9722
$w = 1$	19.1794	0.3520	5.1972
$w = 2$	-5.9923	-1.8207	-1.2544
$w = 3$	-9.7435	-0.3226	-37.5713
$w = 4$	64.1003	3.1109	20.3288
$w = 5$	-4512.9794	-11740.4489	1189.2378
$w = 6$	0.2324	0.0888	-5.9175
$w = 7$	42.6292	2.2433	12.4537
$w = 8$	236.6448	-35073.4100	1479.1315
$w = 9$	80.5643	5.2245	6.9051
$w = 10$	0.2542	-0.0543	0.3699
$w = 11$	275.8564	206.0867	-19976.7228
$w = 12$	-80.8444	-1.8040	-22.7775
$w = 13$	-1658.9701	-1800.4704	-1332.2755
$w = 14$	0.6853	-0.0314	129.9186
$w = 15$	102.0798	1.5156	460.0508
$w = 16$	-4947.7715	-23877.7532	2314.5123
$w = 17$	-24.5357	0.5622	-133.4187
$w = 18$	61.3209	1.8288	-202.0073
$w = 19$	1608.1333	9.3080	4925.4788
$w = 20$	96.7751	2.1706	411.3621

Table S6. ANN_{30} parameters of the hidden layer (p_{wj}). w refers to the cell index while j refers to the input index.

p_{wj}	$j = 0$	$j = 1$	$j = 2$	$j = 3$	$j = 4$	$j = 5$	$j = 6$
$w = 1$	0.9210260	0.0144556	-0.0055219	-0.0254152	0.0265035	0.0012437	0.7429898
$w = 2$	-0.9764701	0.0370388	-0.0669420	-0.0983673	0.0187385	0.0166996	0.1093835
$w = 3$	-0.9514059	0.0003606	-0.0140501	-0.1407724	-0.0163624	0.0081432	0.0657403
$w = 4$	-0.9676759	0.0011831	-0.0200063	0.0013056	-0.0232506	0.0025903	-1.0061496
$w = 5$	-0.9860803	0.0153332	0.0020657	-0.0040916	-0.7942445	-0.0006715	0.0210091
$w = 6$	-1.0314882	0.0454929	0.0179973	-0.0002921	0.0078623	-0.0008390	-0.0048101
$w = 7$	0.9484849	0.0042774	-0.0191207	-0.0020081	0.0181528	0.0001254	-0.0308138
$w = 8$	-0.9820565	-0.0081976	0.0098745	0.0063298	-0.1588040	-0.0018979	0.0043109
$w = 9$	0.9800557	-0.0200801	0.0256699	-0.0011864	0.0120047	-0.0036895	-0.0071408
$w = 10$	-1.0237492	0.1225330	0.0721624	0.0169683	-0.8349192	0.0280220	-0.1447876
$w = 11$	-0.2962458	-0.0197919	0.1882503	-0.0428655	-0.0017384	0.0123888	-1.0239027
$w = 12$	-0.8529406	-0.0309410	0.0117282	0.0510897	-0.0522445	-0.0022420	-1.3906473
$w = 13$	-0.0412349	-0.0239703	-0.0166151	-0.0425768	0.2393774	-0.0089242	0.0314645
$w = 14$	-1.0136615	0.0805500	0.0448732	0.0104937	-0.5435348	0.0179463	-0.0907573
$w = 15$	0.0630118	0.1521816	0.1063422	0.0638993	-0.1034713	0.0127883	-0.2630422
$w = 16$	-0.0208821	-0.0524536	0.0550701	0.0802684	0.0609775	0.0010868	0.0847652
$w = 17$	-0.1048916	-0.0497711	-0.0234816	0.1069471	0.0761395	0.0004229	0.1142789
$w = 18$	0.3777372	-0.0152203	-0.0987326	-0.2421954	-0.0604080	-0.0216252	0.8987170
$w = 19$	0.8877308	-0.5220655	0.1144222	0.1379238	-0.2481600	0.0105050	0.5708149
$w = 20$	0.0315558	0.0517333	-0.0794345	0.0396991	-0.1372819	0.0049044	-0.0271347
$w = 21$	0.5344184	-0.5093209	-0.0224921	0.3218643	0.5777068	-0.0424688	-0.1385745
$w = 22$	0.9566903	-0.2122409	-0.2471810	-0.4637741	-0.1823084	-0.0257392	-0.3850709
$w = 23$	-0.7326411	-0.0041719	-0.0927623	-0.7648524	-0.0931389	0.0395052	0.3862078
$w = 24$	0.8701789	-0.0895655	-0.2855420	-0.5321962	-0.1703353	-0.0436570	0.9563571
$w = 25$	0.9039345	0.1360256	-0.8335025	-0.0212982	0.2684881	0.0329665	0.8461650
$w = 26$	-0.9189061	-0.1482228	0.8775237	0.0217204	-0.2768904	-0.0450017	-0.7943776
$w = 27$	0.3367264	0.6245309	-0.1381385	-0.2949816	-0.3098113	0.0680996	-1.1056290
$w = 28$	0.9425932	-0.1788266	-0.2369277	-0.4742727	-0.1979102	-0.0250730	-0.3491069
$w = 29$	-0.0055630	0.2531751	-0.1584149	-0.0627589	0.2618731	-0.0104989	-0.1385452
$w = 30$	-0.9658869	0.0534001	-0.0954103	-0.1401058	0.0273488	-0.0006017	0.1378509

Table S7. ANN_{30} parameters of the output layer ($\hat{p}_{nw}$). n refers to the output index while w refers to the cell index. The table was transposed because of space.

$\hat{p}_{nw}{}'$	$n = 1$	$n = 2$	$n = 3$
$w = 0$	5014.6684	-67218.7139	-24640.1621
$w = 1$	-141.8928	-356.0612	12208.2237
$w = 2$	-4537.3650	-1618.7976	-33857.4346
$w = 3$	-15669.8679	-2035.7802	-34751.8883
$w = 4$	-1698.7260	74.0943	-23834.3334
$w = 5$	-2897.8441	-67366.3029	2648.7725
$w = 6$	-4253.7984	51194.8479	1622.2464
$w = 7$	1738.9680	3965.0607	-25216.0578
$w = 8$	47509.0643	-3933.6909	21278.2679
$w = 9$	11449.9315	31428.3834	-17823.7016
$w = 10$	316.2938	2112.2209	-587.5362
$w = 11$	2.5557	-0.6271	54.5921
$w = 12$	-10.0019	-33.2223	1099.8649
$w = 13$	105.3360	-24.9122	54.2074
$w = 14$	-1602.1186	-10959.1007	2929.0181
$w = 15$	-40.8261	-6.9090	5.7855
$w = 16$	314.5280	-1.1459	126.6244
$w = 17$	-255.9859	5.5620	6.4420
$w = 18$	-4.4710	-2.1267	62.6596
$w = 19$	-1.5361	-1.6503	-1.2003
$w = 20$	217.8041	-22.7268	131.8603
$w = 21$	-1.2949	-0.3805	-0.9806
$w = 22$	32.6347	-3.2432	-3.7043
$w = 23$	23.2024	3.1948	59.0229
$w = 24$	3.6723	0.6332	-11.5088
$w = 25$	4.3456	2.7947	24.1452
$w = 26$	3.9460	2.6198	19.2900
$w = 27$	1.2927	0.3668	1.5258
$w = 28$	-36.7011	2.8556	9.0313
$w = 29$	2.1071	6.4978	5.4788
$w = 30$	917.3079	377.9798	7931.9928

Table S8. ANN_{40} parameters of the hidden layer (p_{wj}). w refers to the cell index while j refers to the input index.

p_{wj}	$j = 0$	$j = 1$	$j = 2$	$j = 3$	$j = 4$	$j = 5$	$j = 6$
$w = 1$	0.9200367	0.0076764	-0.0009857	-0.0165760	0.0175379	0.0036085	1.1262685
$w = 2$	-0.9827898	0.0496525	-0.1002516	-0.0668408	0.0047046	0.0415199	0.1402700
$w = 3$	-0.9308891	-0.0001035	-0.0269745	-0.4010498	-0.0368052	0.0032319	0.1173029
$w = 4$	-0.9687155	-0.0009855	-0.0243023	0.0029107	-0.0203690	0.0033600	-1.3451289
$w = 5$	-0.9852336	0.0142400	0.0014441	-0.0031647	-0.8427927	0.0003946	0.0189562
$w = 6$	-1.0577440	0.0645960	0.0225978	-0.0035011	0.0133353	-0.0033700	-0.0141970
$w = 7$	0.9672956	-0.0006928	-0.0020129	0.0020031	0.0492340	0.0005421	-0.0786190
$w = 8$	-0.9820317	-0.0094197	0.0094059	0.0063725	-0.1994111	-0.0020577	0.0111338
$w = 9$	0.9335614	-0.0199485	0.0194790	-0.0012524	0.0097728	-0.0032763	-0.0050180
$w = 10$	-1.0206142	0.1171118	0.0694301	0.0106954	-0.9272709	0.0216159	-0.0984454
$w = 11$	-0.2021453	-0.0663709	0.3816407	-0.0399833	-0.0261569	-0.0198502	-1.1507226
$w = 12$	-0.8642980	-0.0142196	0.0024998	0.0284852	-0.0300141	-0.0066122	-1.9375957
$w = 13$	-0.3665186	-0.0618812	-0.0245794	-0.0813862	0.2434990	-0.0155301	0.0221331
$w = 14$	-1.0105644	0.0696289	0.0388431	0.0058528	-0.5436944	0.0125731	-0.0555272
$w = 15$	0.1173194	0.1951941	-0.1354525	0.0246064	-0.1158351	0.0259153	0.0048485
$w = 16$	-0.0292299	-0.0397383	0.0363245	0.0751064	0.0140031	0.0035132	0.0213155
$w = 17$	-0.1151793	-0.0272537	-0.0379548	0.1299361	0.0129362	0.0044727	0.0567924
$w = 18$	0.2381513	0.0069968	-0.0690950	-0.3235856	0.0025524	-0.0123455	0.9082660
$w = 19$	0.6764443	-0.3708194	0.0610742	-0.0138102	-0.0098356	-0.0233576	0.0042944
$w = 20$	0.1776015	0.0447696	-0.0705152	0.0920699	-0.1000310	0.0024711	0.0430449
$w = 21$	0.4755637	-0.3271537	-0.0750362	0.3348838	0.6578049	-0.0353007	-0.0507947
$w = 22$	0.8001454	-0.2364633	-0.0887444	-0.3893658	0.0442344	0.0264606	-0.2680961
$w = 23$	-0.8155336	-0.0009335	-0.0734646	-1.0857503	-0.1055481	0.0064367	0.3174450
$w = 24$	0.7063370	-0.0240587	-0.2451499	-0.9691192	0.0643380	-0.0601396	2.2032006
$w = 25$	0.6289627	0.3954138	-1.2483782	0.0063962	0.1141701	0.1673217	1.5710404
$w = 26$	-0.6302366	-0.3982644	1.2502565	-0.0051650	-0.1157481	-0.1688652	-1.5453826
$w = 27$	0.4735275	0.2978224	-0.1135825	-0.0780181	-0.4178516	0.0576710	-0.2308728
$w = 28$	0.8221592	-0.1029569	0.0060872	-0.5483422	-0.0782219	0.0694359	-1.4717670
$w = 29$	0.1352859	0.1493739	-0.1644704	-0.0052121	0.1976783	0.0194770	0.0758168
$w = 30$	-0.9755096	0.0670431	-0.1349110	-0.0906471	0.0062291	0.0325506	0.1668614
$w = 31$	1.6862808	-0.6694639	-1.2833303	-1.0903861	2.3292406	-0.0465565	1.2805244

$w = 32$	-0.4655295	-0.1729073	0.0404600	-0.3131722	0.2125989	-0.0497864	1.9471693
$w = 33$	0.5020775	-0.2096806	0.0905325	-0.0383210	0.5208753	-0.0528558	0.2703349
$w = 34$	0.2849129	0.0954594	-0.0380159	0.5053490	0.1045337	-0.0474973	0.0986259
$w = 35$	0.8222148	-0.7026919	-0.1287898	-0.0123962	-0.0646418	-0.0159605	0.0724903
$w = 36$	-0.0989402	-0.0397708	-0.5586013	-0.0453288	0.0873285	0.0487099	0.2711086
$w = 37$	0.2411174	0.0070856	0.0771951	-0.0647974	0.0479239	-0.0473074	1.5814832
$w = 38$	-0.5713111	0.1007126	0.1058824	-0.0806702	-0.4140267	0.0331325	-0.4111302
$w = 39$	-0.3920248	-0.1036485	-0.0980156	0.1339449	0.2666204	-0.0394928	1.0521331
$w = 40$	0.4839421	-0.1284074	-0.0375403	-0.0579197	0.5687728	-0.0458338	0.2178815

Table S9. ANN_{40} parameters of the output layer ($\hat{p}_{nw}$). n refers to the output index while w refers to the cell index. The table was transposed because of space.

$\hat{p}_{nw}{}'$	$n = 1$	$n = 2$	$n = 3$
$w = 0$	9507.3185	-66056.0886	-26159.9740
$w = 1$	1522.8483	-103.7176	14027.6114
$w = 2$	-8147.1965	-1610.6743	-33520.0646
$w = 3$	-8364.9046	110.7577	-35273.7378
$w = 4$	-432.1208	328.3294	-25438.0037
$w = 5$	-2605.5484	-67054.3050	3217.6946
$w = 6$	-6243.7633	48481.3382	1503.8741
$w = 7$	-2631.4782	1147.4086	-24189.5422
$w = 8$	43947.8750	-4943.5291	17180.0172
$w = 9$	12730.2588	26530.2238	-20943.9385
$w = 10$	-55.7499	2112.2702	-391.8940
$w = 11$	6.4570	1.9100	57.3262
$w = 12$	207.1185	-14.2433	1791.6267
$w = 13$	146.7510	-22.4728	70.4588
$w = 14$	246.3041	-16426.9455	2902.5883
$w = 15$	-40.3999	-14.6354	2.5303
$w = 16$	353.2149	-30.2512	159.6581
$w = 17$	-257.6934	15.8314	3.7484
$w = 18$	4.6366	-0.2838	67.7121
$w = 19$	-15.4434	-17.1775	-0.1232
$w = 20$	283.2293	-11.2045	161.0353
$w = 21$	-3.5059	-0.8316	-2.6006
$w = 22$	12.4337	0.1920	-0.1678
$w = 23$	185.8430	-3.1862	824.5165
$w = 24$	-0.1302	0.0448	-2.4149
$w = 25$	15.7102	10.2271	101.9132
$w = 26$	15.5747	10.2622	99.8888
$w = 27$	35.0183	1.8037	18.6600
$w = 28$	-1.8163	-0.3499	7.8325
$w = 29$	14.1689	10.2108	15.5896
$w = 30$	2433.1696	529.2693	9965.6195

$w = 31$	0.0477	-0.0012	0.0022
$w = 32$	-0.8329	0.1420	-7.9189
$w = 33$	-50.2144	-3.2071	-15.8676
$w = 34$	-9.2777	0.4421	-12.1313
$w = 35$	0.9162	2.7283	-0.3383
$w = 36$	2.9042	2.4056	-1.2588
$w = 37$	0.9017	0.4573	-11.5757
$w = 38$	4.5984	-8.1404	-30.9170
$w = 39$	1.8114	1.1330	28.8090
$w = 40$	48.5507	-5.4538	4.9950

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