Electronic Supplementary Information

Utilizing Machine Learning to Optimize Metal-Organic Framework-Derived Polymer Membranes for Gas Separation

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1 Synthesis

1.1 Diverse Set

Table S1 All parameter combinations and their corresponding fitness value of the Diverse Set (DS).

Number	c(Linker) [mM/l]	c(Metal) [mM/l]	Modulator (Water) [ml]	DABCO [eq of Linker]	EtOH [%]	Fitness
1	0.01	0.05	0	0.10	0	0.33
2	1.00	5.00	40	2.00	100	0.48
3	0.01	0.05	20	2.00	100	0.37
4	1.00	0.05	40	0.10	50	0.63
5	1.00	2.52	0	2.00	0	0.00
6	0.01	5.00	40	1.05	0	0.63
7	0.51	5.00	0	0.10	100	0.00
8	0.51	0.05	40	2.00	0	0.00
9	1.00	0.05	0	1.05	100	0.00
10	0.01	2.52	40	0.10	100	0.36

1.2 Genetic Algorithm 1

Table S2 All parameter combinations and their corresponding fitness value of the first genetic algorithm (GA1).

Number	c(Linker) [mM/l]	c(Metal) [mM/l]	Modulator (Water) [ml]	DABCO [eq of Linker]	EtOH [%]	Fitness
11	0.01	0.05	0	0.25	87	0.00
12	0.85	0.05	2	0.91	16	0.00
13	0.49	1.20	40	0.85	23	0.00
14	1.00	5.00	40	2.00	100	0.89
15	0.97	4.39	36	0.61	66	0.00
16	0.85	4.70	40	0.99	100	0.00
17	0.84	0.05	40	1.43	26	0.93
18	0.23	0.00	12	0.85	48	0.00
19	0.33	1.64	40	1.78	0	0.00
20	0.78	5.00	36	1.87	100	0.72

1.3 Genetic Algorithm 2

Table S3 All parameter combinations and their corresponding fitness value of the second genetic algorithm (GA2).

Number	c(Linker) [mM/l]	c(Metal) [mM/l]	Modulator (Water) [ml]	DABCO [eq of Linker]	EtOH [%]	Fitness
21	0.51	5.00	19	1.26	100	0.88
22	0.31	0.33	0	1.36	25	0.00
23	0.70	0.56	40	1.36	24	0.94
24	0.37	0.83	38	1.23	52	0.00
25	0.60	2.35	22	0.74	49	0.92
26	0.01	0.05	0	0.25	87	0.00
27	0.87	5.00	40	2.00	100	0.83
28	0.78	5.00	36	1.87	100	0.91
29	0.58	0.05	7	0.32	78	0.00
30	0.92	4.89	15	1.47	91	0.87

2 Fitness

2.1 Diverse Set

Table S4 Fitness criteria and overall fitness for the Diverse Set

Number	Phase Identity	Crystallinity	Fitness
1	1	32.7	0.33
2	1	47.6	0.48
3	1	37.3	0.37
4	1	63.4	0.63
5	0	0.0	0.00
6	1	62.7	0.63
7	0	0.0	0.00
8	0	0.0	0.00
9	0	0.0	0.00
10	1	35.8	0.36

2.2 Genetic Algorithm 1

Table S5 Fitness criteria and overall fitness for the Genetic Algorithm 1

Number	Phase Identity	Crystallinity	Fitness
11	0	0.0	0.00
12	0	0.0	0.00
13	0	0.0	0.00
14	1	88.9	0.89
15	0	0.0	0.00
16	0	0.0	0.00
17	1	93.0	0.93
18	0	0.0	0.00
19	0	0.0	0.00
20	1	72.1	0.72

2.3 Genetic Algorithm 2

Table S6 Fitness criteria and overall fitness for the Genetic Algorithm 2

Number	Phase Identity	Crystallinity	Fitness
21	1	87.6	0.88
22	0	0.0	0.00
23	1	93.8	0.94
24	0	0.0	0.00
25	1	91.6	0.92
26	0	0.0	0.00
27	1	82.7	0.83
28	1	91.0	0.91
29	0	0.0	0.00
30	1	86.5	0.87

3 X-Ray Diffraction

3.1 Diverse Set

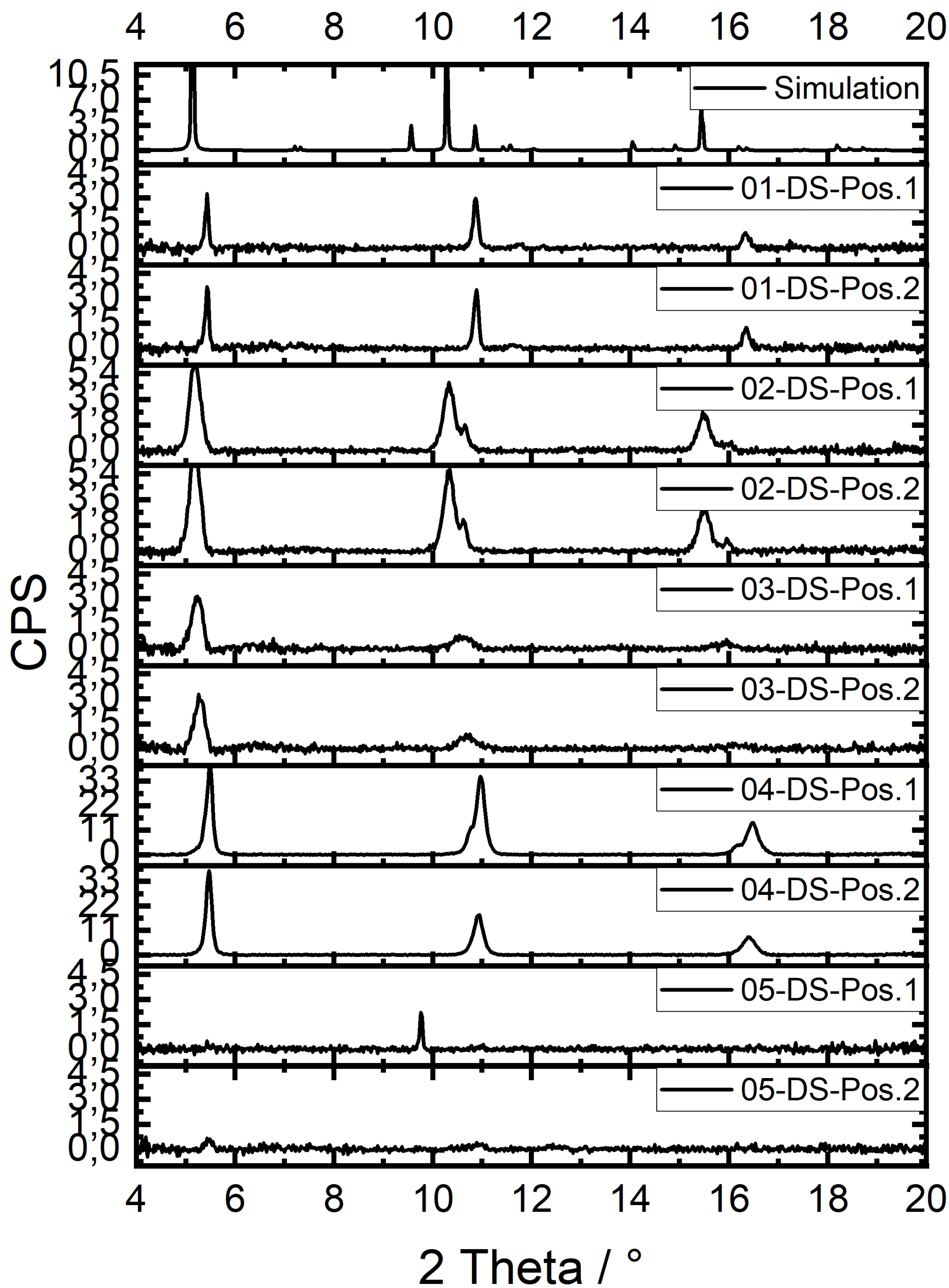


Fig. S1 X-Ray Diffractograms of the Diverse Set for Parametercombinations 1 to 5.

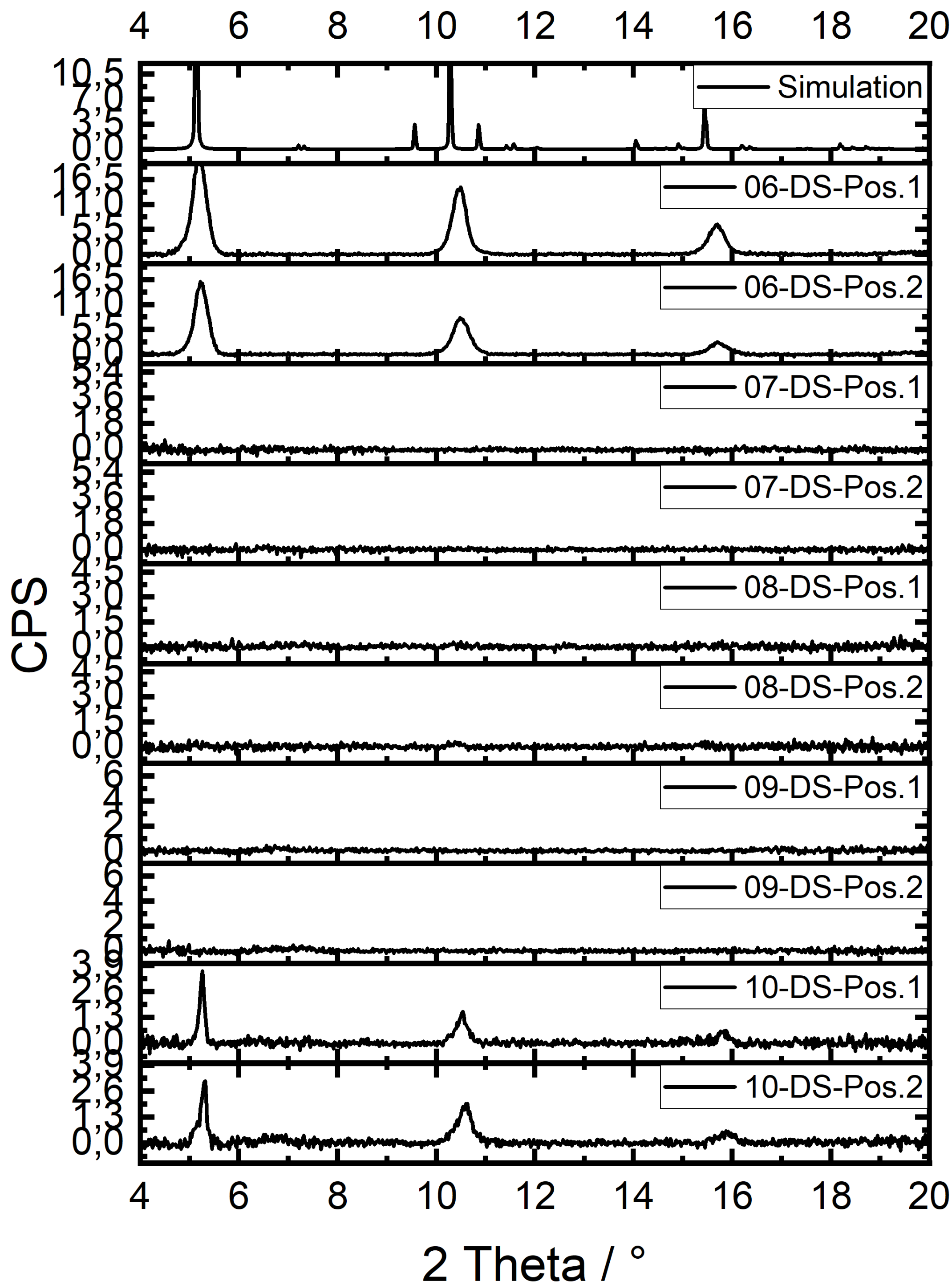


Fig. S2 X-Ray Diffractograms of the Diverse Set for Parametercombinations 6 to 10.

3.2 Genetic Algorithm 1

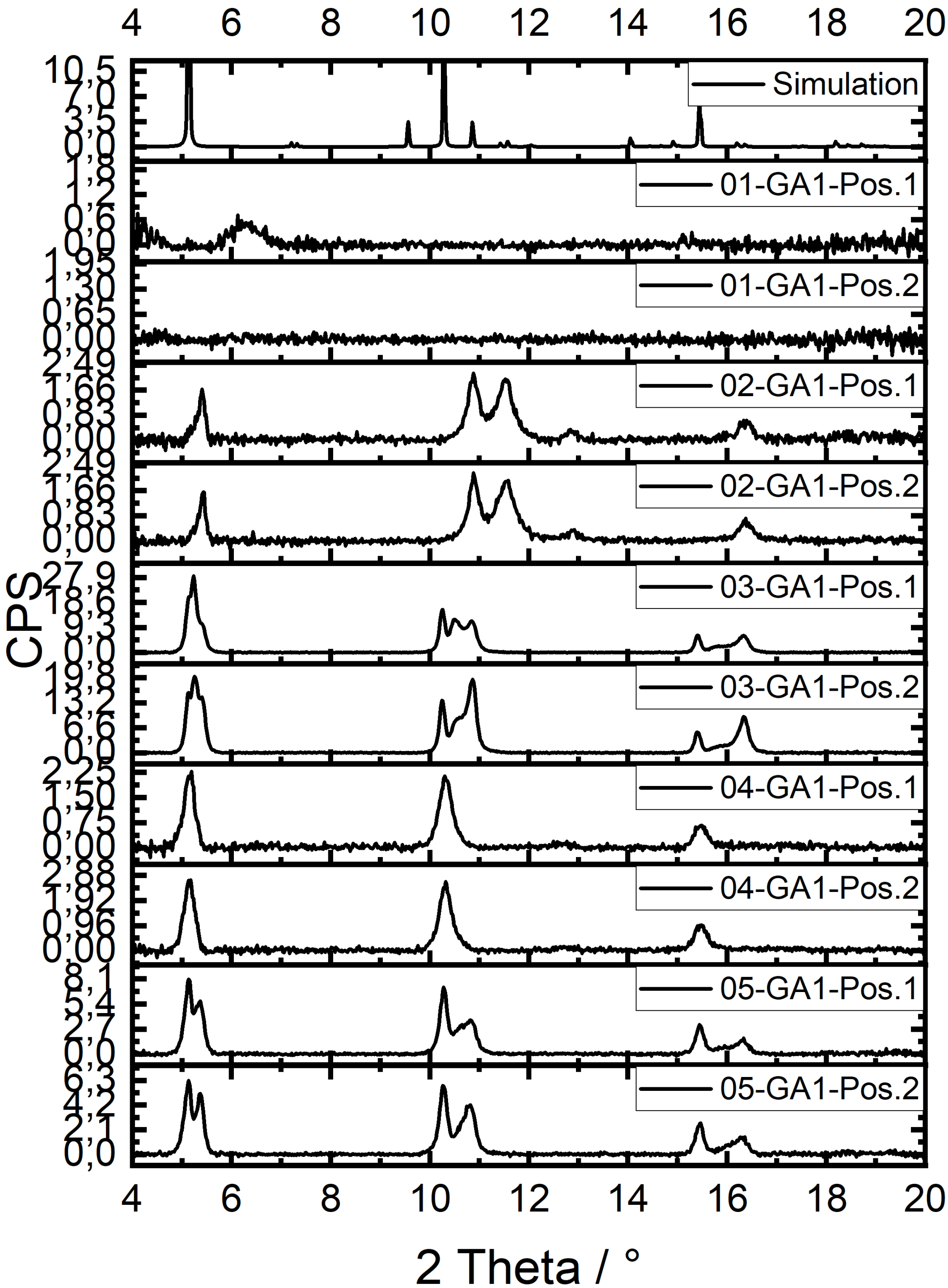


Fig. S3 X-Ray Diffractograms of the Genetic Algorithm 1 for Parametercombinations 1 to 5.

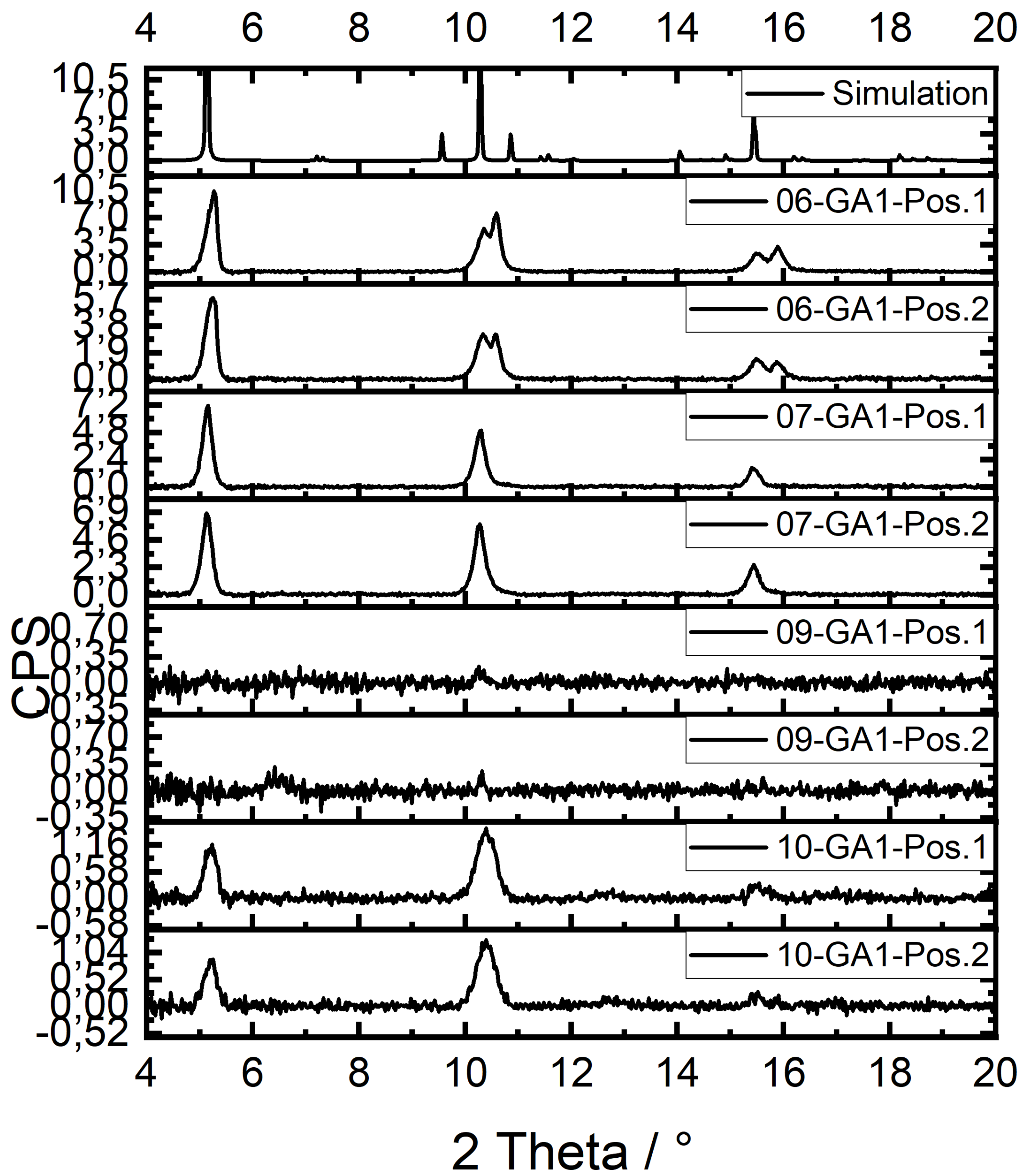


Fig. S4 X-Ray Diffractograms of the Genetic Algorithm 1 for Parametercombinations 6 to 10. It is to be noted, that experiment number 8 is missing, since the amount of metal-salt was suggested to zero by the genetic algorithm.

3.3 Genetic Algorithm 2

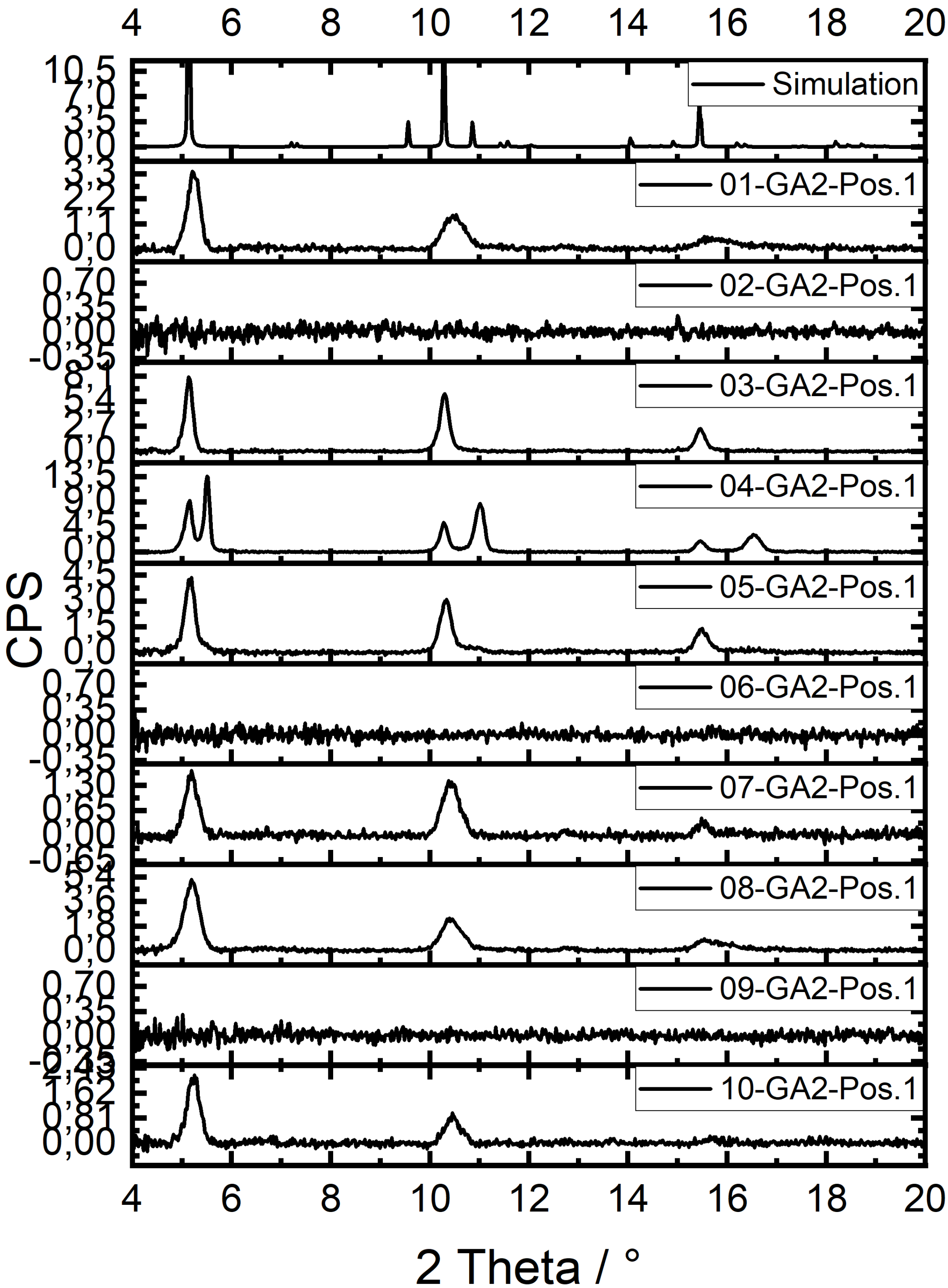


Fig. S5 X-Ray Diffractograms of the Genetic Algorithm 2 for Parametercombinations 1 to 10.

4 Gas permeance and selectivity of the substrate

Table S7 Gas permeance of the gold-coated alumina substrate

	Permeance (x 10^{-7} mol m^{-2} s^{1} Pa^{-1})				Selectivity		
	H_2	CH_4	N_2	CO_2	H_2/CH_4	H_2/N_2	H_2/CO_2
Gold-coated alumina substrate	31.1	24.6	25.5	22.4	1.2	1.2	1.4

5 Synthesis Set-up

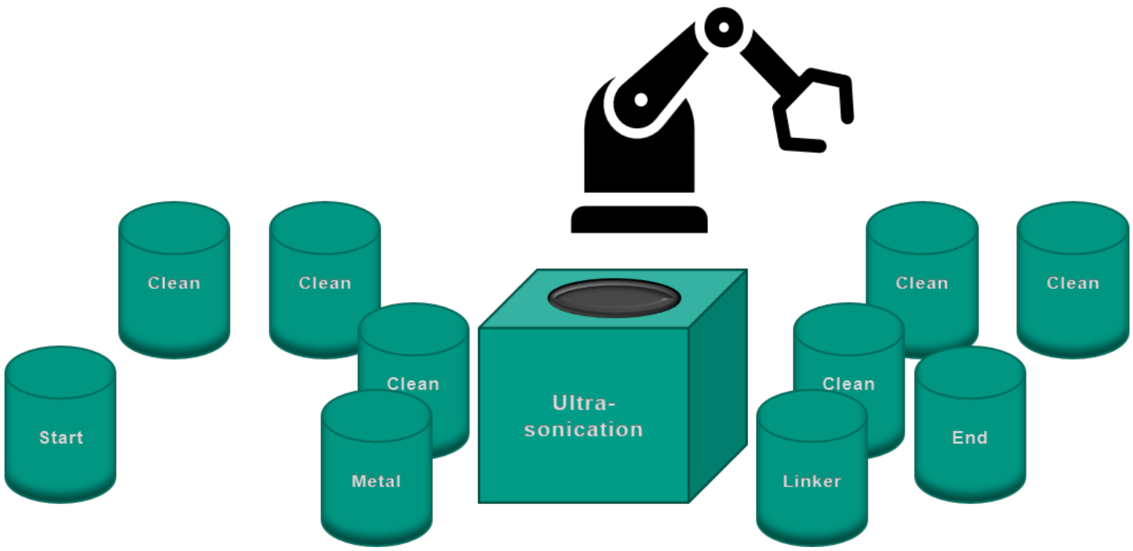
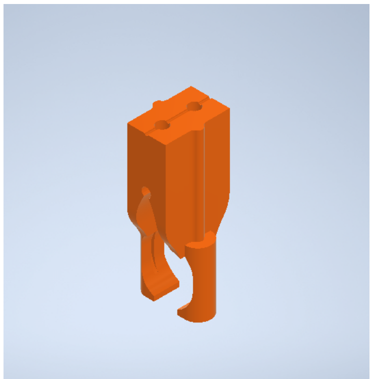


Fig. S6 Set-up Robot in Glovebox



(a) CAD sketch of the newly designed gripper for the gold coated alumina substrate for the robotic sampleholder.

(b) Picture of the robotic sampleholder with four applied grippers for the gold coated alumina substrate.

Fig. S7 Design and executed version of the sample holder for the robotic setup.

6 Machine Learning Workflow

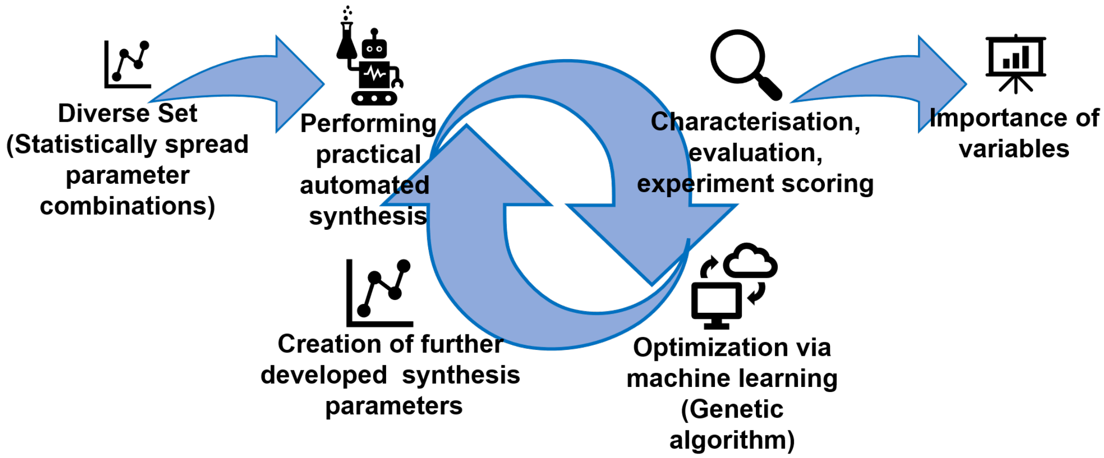
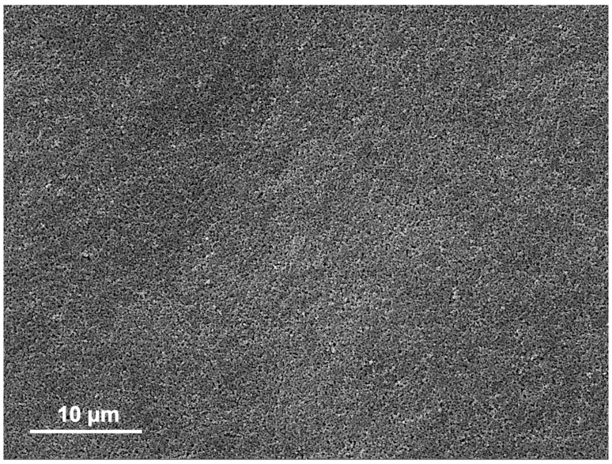
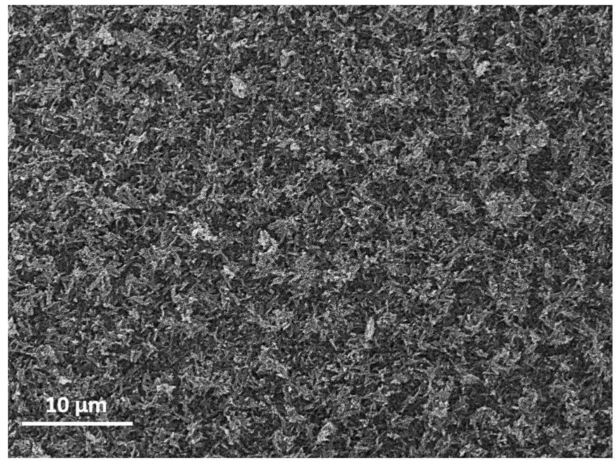


Fig. S8 Chart of the practical workflow when performing the machine learning optimization.

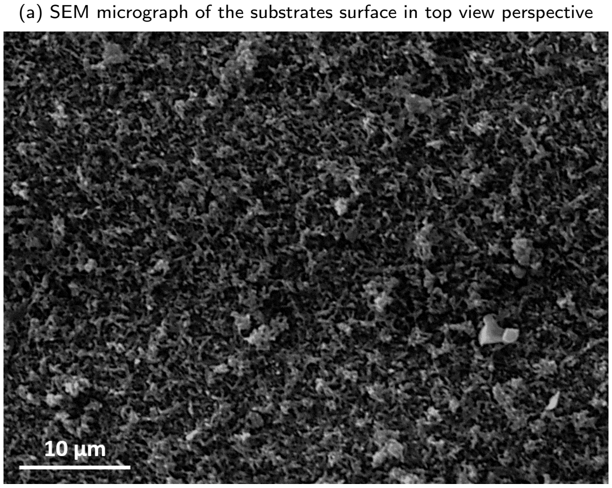
7 Scanning Electron Microscopy



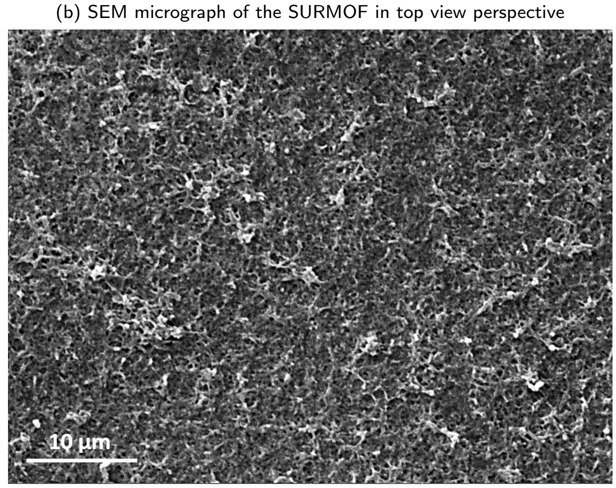
(a) SEM micrograph of the substrates surface in top view perspective



(b) SEM micrograph of the SURMOF in top view perspective

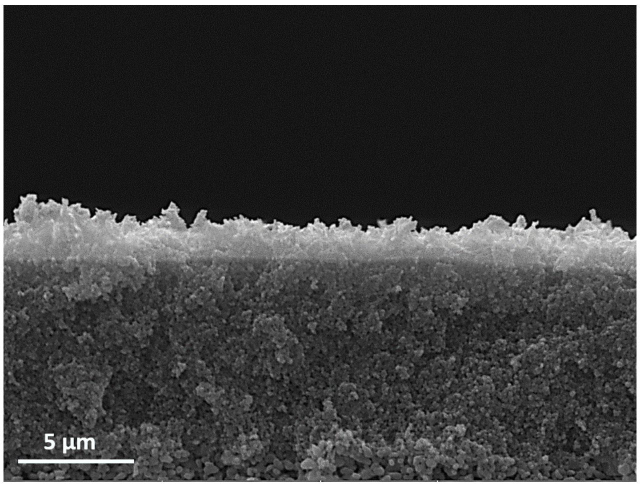


(c) SEM micrograph of the SURMOF-CL in top view perspective

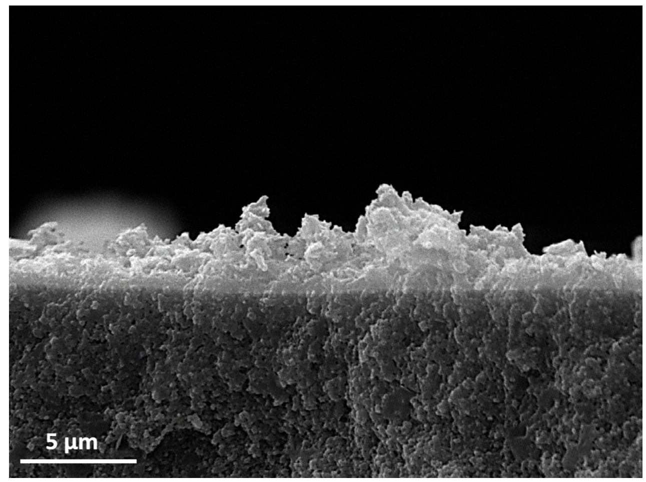


(d) SEM micrograph of the SURGEL in top view perspective

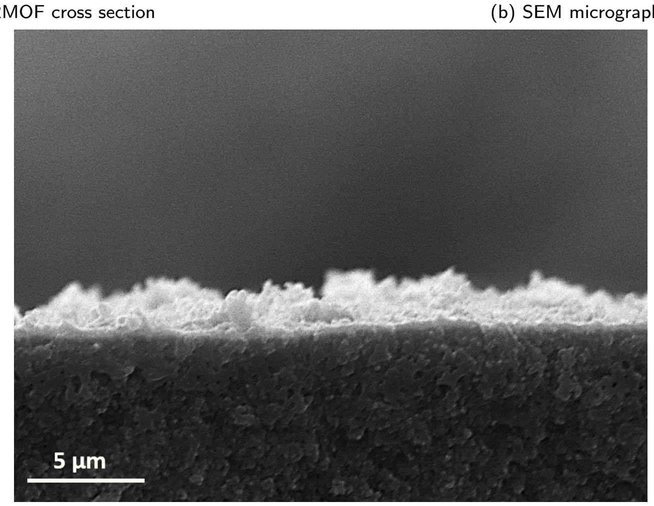
Fig. S9 SEM micrographs of the bare gold coated alumina substrate and all three synthesis stages: SURMOF, SURMOF-CL and SURGEL on a gold coated porous alumina substrate.



(a) SEM micrograph of the SURMOF cross section



(b) SEM micrograph of the SURMOF-CL cross section



(c) SEM micrograph of the SURGEL cross section

Fig. S10 SEM micrographs of the SURMOF, SURMOF-CL and SURGEL cross section on a gold coated porous alumina substrate.