Supplementary Materials: Kinetics of the direct DME synthesis: state of the art and comprehensive comparison of semi-mechanistic, data-based and hybrid modeling approaches

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1. Model specific parameters of the ANN-HM

The model specific parameters of the ANN-HMs discussed in the manuscript (ANN-HM with 5, 26 and 28 HNs) are given in this section. Especially, these are the connection weights between the input and the hidden layer ($W_{i,h}$) and between the hidden and the output layer ($W_{h,o}$), as well as the biases of the hidden and outputs neurons (b_h and b_o) as shown in Figure S1.

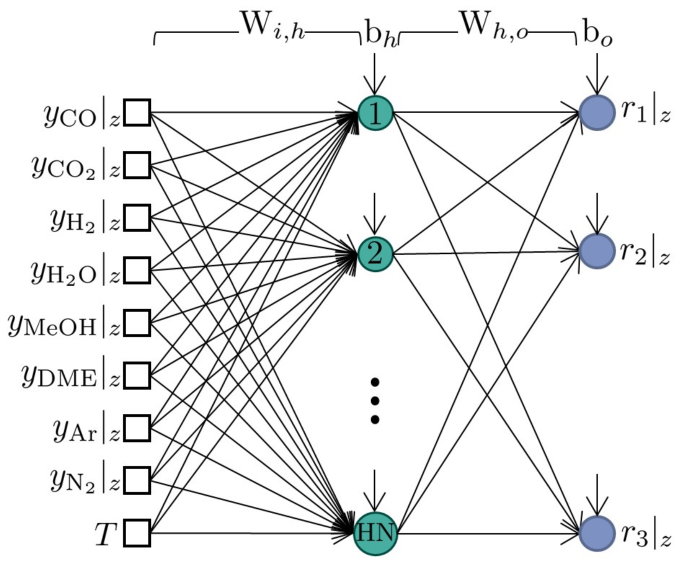

Figure S1. Schematic representation of the ANN-HM.

Table 1: Model specific parameters of the ANN-HM with 5 HNs. Connection weights of the input and hidden layer, biases of the hidden layer.

				$\mathbf{W}_{i,h}$					$\mathbf{b}_h$
-0.60114	0.14025	-0.63213	0.80736	0.08968	-0.04125	-0.27108	0.00641	-0.43090	3.10519
-1.14673	0.65526	1.31330	1.05803	0.01463	-0.00635	-0.26234	0.05774	-0.44970	3.95600
-3.02931	-0.02462	-3.53521	0.79813	0.04093	19.72959	-3.39553	-0.23304	-1.22452	20.09301
3.98021	-5.89989	0.18136	-1.04395	-0.77207	-0.14084	0.37947	0.21915	0.88733	-7.69831
2.60925	0.85926	2.62343	0.47530	-1.60951	-18.55797	3.14491	0.16885	1.05177	-19.56385

Table 2: Model specific parameters of the ANN-HM with 5 HNs. Connection weights of the hidden and output layer, biases of the output layer.

	$\mathbf{W}_{h,o}^{-1}$		$\mathbf{b}_o$
-14.986096	-16.498396	-14.484296	2.8317
4.86557018	5.96789325	4.68863279	10.1015
6.23236401	-0.5675959	-0.8749624	10.0811
-11.948762	-13.012473	-11.014278	
8.14194045	0.55032097	-1.1746633	

Table 3: Model specific parameters of the chosen ANN-HM with 26 HNs. Connection weights of the input and hidden layer, biases of the hidden layer.

				$\mathbf{W}_{i,h}$					$\mathbf{b}_h$
0.26106	0.12391	0.82471	0.33482	-0.72017	-0.16497	-0.19633	-0.32429	0.86960	-0.36237
0.19960	0.28507	-1.86320	0.22985	-0.67681	-1.02954	0.16398	0.11994	0.36954	-1.43625
-1.16703	-1.23930	1.94422	0.56587	0.49974	0.16683	1.36101	0.10762	0.45637	-1.66672
0.20960	0.84537	0.24244	0.16916	2.68825	-2.82029	0.66726	-0.00679	2.02909	-4.48702
0.09335	0.43959	-0.34136	0.30138	-0.16581	-0.58593	-0.26262	-0.07968	-1.58115	0.47128
0.00205	0.06033	-0.14307	-0.04459	0.11140	-0.40748	0.06257	-0.22526	0.28571	-0.02085
0.16260	0.45512	2.20344	-0.94117	-0.25011	-0.46153	0.34534	-0.03916	0.32684	-2.61076
-0.24824	-0.39428	0.83872	-0.18088	0.37006	-0.00575	0.06836	0.20577	-1.22799	1.04678
0.06642	0.20981	0.64012	0.33408	-0.77574	-1.37976	0.24136	-0.16015	0.97653	-1.36315
-0.22450	-0.46566	-0.82017	-0.21527	1.06934	0.04703	-0.35663	0.05209	-1.94161	3.53783
0.35660	0.70139	-0.16036	-0.35555	0.14775	0.37536	0.00984	0.08682	1.31660	-2.49721
-0.56388	-0.14535	0.24360	0.31073	-0.98601	-0.13431	0.77766	0.02603	0.25057	-1.83903
-0.79324	1.26824	1.96136	-1.87042	0.29754	0.27066	0.87333	0.10029	1.36528	-2.69636
0.85912	1.50609	-2.13724	-0.40449	-0.68632	-0.33117	-1.34989	-0.21578	-0.16249	2.31635
1.18093	0.13571	-2.43287	-0.26083	0.13894	-0.98120	-0.62123	-0.01411	0.16473	-0.56915
0.13604	0.99348	-0.45484	-0.78550	-0.55885	-1.27551	0.17066	0.31817	0.22216	-1.61553
0.21975	0.25746	-1.96152	0.22746	-0.90822	0.80587	-0.28249	-0.00652	-1.18703	0.06408
0.46520	0.10702	0.07642	0.22147	0.77300	0.61035	-0.50695	-0.01416	0.11848	0.16956
0.25148	0.94812	-0.01635	-0.31923	-0.37376	-0.26056	-0.16762	0.11647	-0.40989	-1.07456
0.65769	0.69521	-0.14171	-0.00214	0.16558	0.92229	-0.69206	-0.32748	-0.59464	-0.22123
0.47396	-0.10615	0.40137	0.06801	0.67045	0.12422	-0.72975	-0.20161	-1.00531	2.23222
0.27067	-0.45266	-1.49560	-0.23085	0.80500	-0.47034	0.42334	0.07572	0.62688	-2.28619
-0.30719	-0.34633	0.06663	1.04917	0.81328	0.10817	0.47108	-0.04857	1.04581	-1.57837
0.57060	0.64921	-1.08245	-0.15155	-0.39120	0.14038	-0.56275	-0.03471	-0.73006	0.16251
0.19670	-0.23625	-0.58807	0.57185	-0.11151	0.01358	-0.15972	0.04672	0.66813	0.32134
0.12129	-0.23764	-2.28032	1.35823	-0.28722	-0.20917	-0.11084	-0.00218	-0.25148	0.02584

Table 4: Model specific parameters of the chosen ANN-HM with 26 HNs. Connection weights of the hidden and output layer, biases of the output layer.

$\mathbf{W}_{h,o}^{-1}$			$\mathbf{b}_o$
0.6009	-0.1806	0.2787	-0.2325
-1.5689	0.0367	1.3037	-0.8352
0.5055	1.0450	1.8062	-0.4410
-0.2549	4.9671	0.2776	
-0.9679	0.4391	0.1689	
-0.5245	-0.2991	-0.3791	
1.4918	-0.1403	1.6442	
-0.5030	-1.7649	-0.2364	
-0.4736	-2.2558	-0.4178	
-1.6924	2.0570	0.3809	
2.9428	1.3525	0.0294	
0.7372	0.6209	-1.2581	
-2.6214	-1.4086	0.8564	
0.2751	1.0030	2.0603	
1.5861	2.0752	0.7404	
2.0191	0.6807	-0.0775	
0.3512	-1.8184	-0.0947	
-0.0987	0.2139	1.2012	
-2.3025	-1.8966	-1.4302	
0.4858	0.0648	-0.4688	
1.6234	0.2751	-0.5682	
-1.1198	-2.5540	-2.5515	
-1.5483	-1.0474	-1.0012	
-1.1105	-1.1005	-0.8157	
0.1349	-0.3539	-1.0865	
1.1528	-0.0161	-0.9238	

Table 5: Model specific parameters of the ANN-HM with 28 HNs. Connection weights of the input and hidden layer, biases of the hidden layer.

$\mathbf{W}_{i,h}$									$\mathbf{b}_h$
0.46664	0.28557	0.43946	-0.00902	-0.16684	0.54786	-0.43955	0.21328	-0.19793	-0.16975
0.60392	0.25781	-0.59811	-1.10325	-0.45429	0.44810	-0.66461	-0.11598	0.98309	1.48690
0.07021	-0.02014	0.00078	0.01422	0.72290	-0.30191	-0.02811	0.14087	0.04316	-0.13027
-0.65256	-0.60885	-0.01501	-0.47318	0.50558	-0.77336	0.62161	0.16996	0.63660	0.93549
-0.19340	0.37544	-1.33623	0.05289	-0.24590	0.51843	-0.46170	-0.06380	-1.95280	2.33584
-0.12152	-0.18302	-1.03475	0.82643	0.44755	0.75689	0.47722	-0.03124	1.65909	-2.46539
0.04288	0.29545	0.68057	-0.10564	0.35736	-0.67963	-0.00200	-0.39468	0.95627	-0.38664
-0.56356	0.67832	2.15088	-0.97097	2.05435	-0.92779	0.52143	0.00043	1.51639	-0.88890
-0.57398	-0.33225	-0.48717	0.55994	-0.00700	1.28148	0.26237	-0.00604	-0.04937	0.32635
0.48020	-0.09632	1.15148	0.49759	-0.08547	1.44197	-0.99544	-0.29592	-0.29415	2.49784
-0.49102	-0.47841	0.52580	-0.25073	0.55789	-0.56585	0.53654	0.08371	0.09385	-0.09893
-0.18325	-0.13461	0.47380	-0.18808	0.43415	-0.08744	0.13086	0.05678	0.17174	0.11998
-0.06853	0.28728	0.73074	-0.53532	0.24692	-0.38353	-0.01040	-0.14186	-0.69265	0.32885
-0.24823	-0.48929	0.60280	-0.27136	1.18187	-1.33482	-0.17447	-0.01340	-1.50596	3.43600
-0.75584	-0.31503	1.63727	0.08533	1.34722	-0.20588	0.42242	-0.18111	0.90560	0.94341
-1.28804	-0.83502	-1.31090	-0.65176	-0.91017	-1.24448	1.05639	0.28885	-0.59850	1.50718
-0.44566	-0.36471	1.78173	-0.59253	0.53608	0.07252	0.27134	-0.01676	0.84184	0.93740
0.73352	-0.90533	-1.51640	2.18375	0.16292	-1.12499	-0.88997	-0.03892	-1.02122	2.41060
1.49650	1.85872	-2.08273	-2.02300	0.28420	-0.19405	-2.46870	-0.23513	-0.77545	6.15865
-0.44727	-0.36909	-0.31667	-0.18004	-0.21224	1.43467	0.19270	-0.01945	0.35410	0.54217
-0.17363	-0.40172	0.07947	0.27894	-0.19348	-0.23665	0.17962	0.07693	0.10611	0.02526
-0.07722	0.60267	-1.06913	-0.95967	0.15029	-0.05226	0.62780	0.02814	0.57058	-3.35825
-0.86457	0.74219	3.77944	-1.28672	0.87443	-0.04660	0.54564	0.00464	0.77024	-0.37002
-0.57021	-0.43266	-1.12872	0.23752	-0.51371	-0.83880	0.80082	0.27356	0.13021	-1.04363
-0.22845	0.49498	1.06860	-0.88429	0.87760	0.09871	0.36785	-0.10285	1.83416	-1.25260
0.09510	-0.11401	0.15330	0.24541	0.33723	-0.14926	-0.07379	0.13185	-0.16622	0.02456
0.49600	0.60806	-1.44494	-0.91344	-0.63084	-0.42098	-0.15879	0.11993	0.01247	-0.91623
0.25459	-0.19486	-0.00958	0.53563	0.49777	-0.08896	-0.23045	-0.21662	-0.94394	0.13169

Table 6: Model specific parameters of the ANN-HM with 28 HNs. Connection weights of the hidden and output layer, biases of the output layer.

$\mathbf{W}_{h,o}^{-1}$			$\mathbf{b}_o$
0.04726	-0.79667	0.43343	0.1063
-1.19233	-0.88727	0.03829	-0.6298
-0.69891	-0.61537	-0.24272	-0.4188
1.02313	1.75151	0.62214	
-1.01982	-1.59516	-0.84936	
-1.76369	-1.66547	-1.24119	
-0.25505	-1.44239	0.32571	
-1.59085	2.50186	0.09647	
-0.49080	0.93114	1.42095	
1.28131	0.54790	-0.37088	
0.28152	-1.14893	-0.84685	
0.13544	-0.88629	0.25524	
-1.00940	-0.10411	1.01269	
-3.10077	1.21421	0.07743	
1.35216	1.63563	-0.66500	
-1.00380	-1.34031	0.03127	
-0.13046	1.07453	1.51489	
2.51110	0.87342	-2.01953	
0.93582	1.24484	2.61213	
-0.03322	-1.04785	-1.38058	
0.18938	-0.40356	-0.61202	
0.13238	-1.13961	-2.50424	
0.15077	-3.01093	-0.01719	
-0.51431	-1.25468	-0.09573	
1.28006	-0.24813	-0.14723	
-0.35293	-0.46369	-0.09210	
2.32963	1.98265	-0.13629	
-0.50319	-0.76767	-0.46772	

2. Supplementary figures

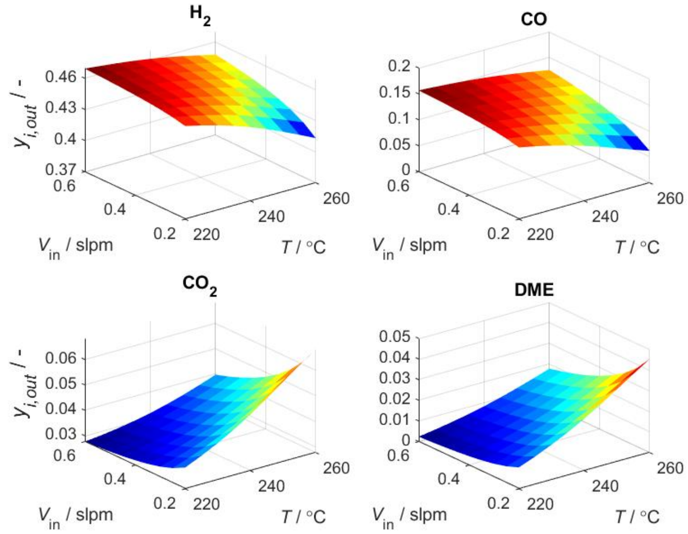

Figure S2. Surface response for the hybrid model predictions of the mole fractions of H_2, CO, CO_2 and DME within the validity range of the temperature and total gas flow. Feed composition: 48.0 % H_2, 16.11 % CO, 2.88 % CO_2. Pressure 50 bar. CZA-to-γ-Al_2O_3-ratio $\mu = 1$. ANN-HM with 26 HNs.

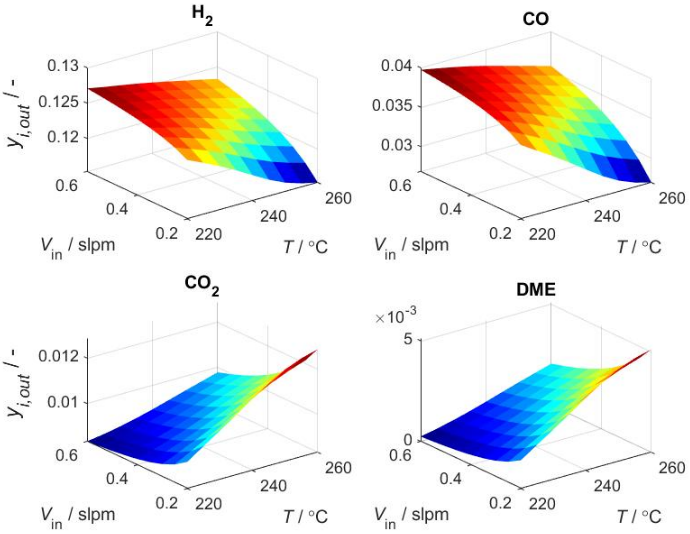

Figure S3. Surface response for the hybrid model predictions of the mole fractions of H_2, CO, CO_2 and DME within the validity range of the temperature and total gas flow. Feed composition: 13.05 % H_2, 4.10 % CO, 0.86 % CO_2. Pressure 50 bar. CZA-to-γ-Al_2O_3-ratio $\mu = 1$. ANN-HM with 26 HNs.

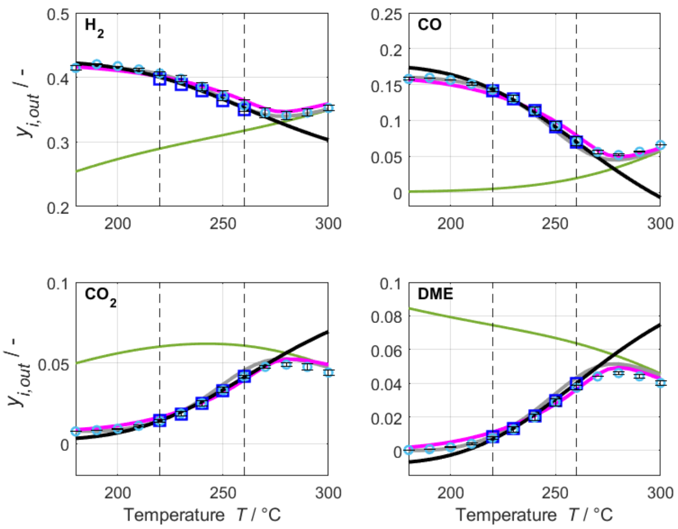

Figure S4. Range extrapolation of the temperature. Nominal feed composition: 42.3 % H_2, 16.1 % CO, 0.82 % CO_2. Total gas flow 0.2 slpm. Pressure 50 bar. CZA-to-γ-Al_2O_3-ratio μ = 1. ANN-HM with 5 HNs.

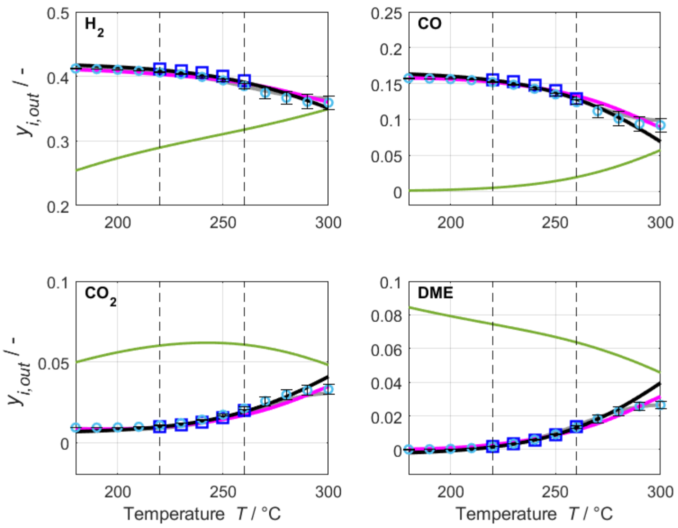

Figure S5. Range extrapolation of the temperature. Nominal feed composition: 42.3 % H_2, 16.1 % CO, 0.82 % CO_2. Total gas flow 0.6 slpm. Pressure 50 bar. CZA-to-γ-Al_2O_3-ratio μ = 1. ANN-HM with 5 HNs.

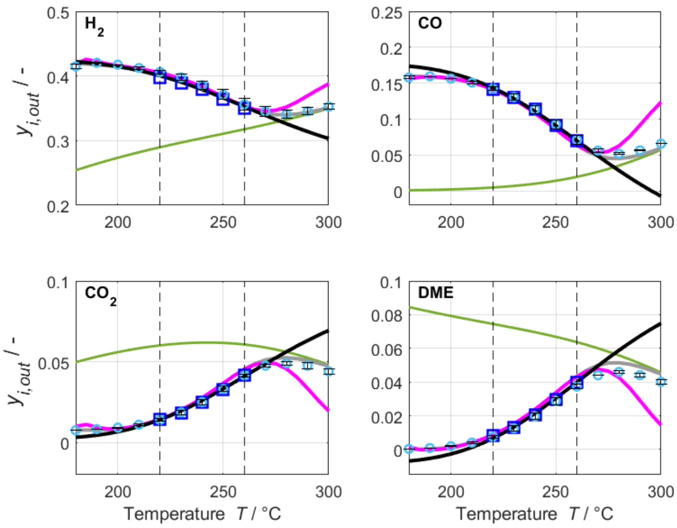

Figure S6. Range extrapolation of the temperature. Nominal feed composition: 42.3 % H_2, 16.1 % CO, 0.82 % CO_2. Total gas flow 0.2 slpm. Pressure 50 bar. CZA-to-γ-Al_2O_3-ratio μ = 1. ANN-HM with 28 HNs.

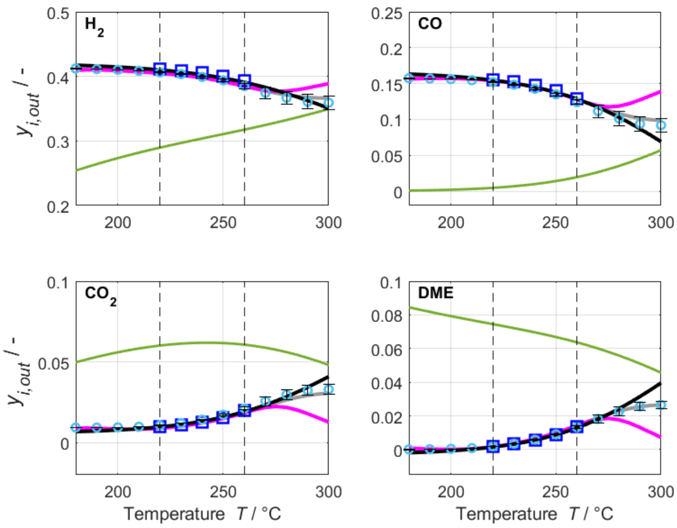

Figure S7. Range extrapolation of the temperature. Nominal feed composition: 42.3 % H_2, 16.1 % CO, 0.82 % CO_2. Total gas flow 0.6 slpm. Pressure 50 bar. CZA-to-γ-Al_2O_3-ratio μ = 1. ANN-HM with 28 HNs.

3. A priori criteria for the determination of mass and heat transport limitations

A priori criteria were employed for ruling out mass and heat transport limitations. These criteria were calculated for temperatures between 180-300 °C and total gas flows between 0.15-0.8 slpm.The values calculated for the worst case scenarios are give in Table 7 for each reaction, and show that mass and heat transport limitations do not play a significant role at the evaluated operating conditions.

Table 7: Calculated a priori criteria for determination of transport limitations.

Phenomenon	Equation [1]	Max. Calculated Value
Outer mass transfer	$\frac{r_{j,eff}\lvert n\rvert}{a' k_f c_b} < 0.05$	For reaction 1: $3.29 \cdot 10^{-5}$ For reaction 2: $2.09 \cdot 10^{-5}$ For reaction 3: $5.74 \cdot 10^{-5}$
Outer heat transfer	$\frac{E_A}{RT_b}\left\lvert\frac{-\Delta H_R}{hT_b}\right\rvert\frac{r_{j,eff}}{a'} < 0.05$	For reaction 1: $4.75 \cdot 10^{-4}$ For reaction 2: $3.09 \cdot 10^{-4}$ For reaction 3: $4.97 \cdot 10^{-4}$
Inner mass transfer	$\frac{r_{j,eff}L^2}{D_{eff}c_s}\frac{(n+1)}{2} < 0.15$	For reaction 1: $4.62 \cdot 10^{-4}$ For reaction 2: $2.93 \cdot 10^{-4}$ For reaction 3: $8.06 \cdot 10^{-4}$
Inner heat transfer	$\frac{E_A}{RT_b}\left\lvert\frac{-\Delta H_R}{\lambda_{eff,p}T_b}\right\rvert r_{j,eff}L^2 < 0.1$	For reaction 1: $4.28 \cdot 10^{-5}$ For reaction 2: $2.78 \cdot 10^{-5}$ For reaction 3: $4.48 \cdot 10^{-5}$

4. Experimental values

The experimental values measured for validation of the simulation results at extrapolated conditions are given in Table 8.

5. Catalyst conditioning and deactivation

Figure S8 shows the COx-conversion (X_{COx}) as a function of the Time-On-Stream (ToS) for a reference operating point measured at 513 K, 50 bar and 7599.5 h^{-1} and a feed of 38.2 % H_2, 15.2 % CO, 1.0 % CO_2, 45.7 % N_2. Within the first 25 hours the reaction conditions were left constant at 513 K, 50 bar and 7599.5 h^{-1}. After that, the conditions were varied dynamically between 453-573 K, 40-60 bar and 2849.8-15199.0 h^{-1}. After 129 hours the experiments for model validation at 453-573 K, 40-60 bar as well as 2849.8-15199.0 h^{-1} were taken. Between the ToS of 129 and 160 hours, where the experiments were conducted, the X_{COx} remains stable with a relative deviation of 10 % between the minimal and maximal measured X_{COx}.

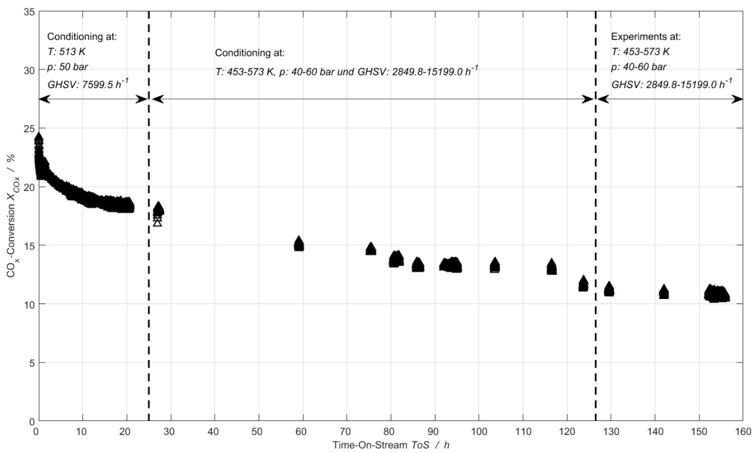

Figure S8. The COx-conversion (X_{COx}) as a function of the Time-On-Stream (ToS) for a reference operating point measured at 513 K, 50 bar and 7599.5 h^{-1} and a feed of 38.2 % H_2, 15.2 % CO, 1.0 % CO_2, 45.7 % N_2.

Table 8: Experimental values measured for validation of simulation results at extrapolated conditions. The catalyst bed consisted of 1.007g CZA, 0.9996 g γ-Al_2O_3, 9.98 g SiC, and it was 7.8 cm long.

$y_{i,in}$				Conditions			$y_{i,out}$							Standard Deviation $y_{i,out}$						
H_2 %	CO %	CO_2 %	N_2 %	T °C	$\dot{V}_{in}$ slpm	p bar	H_2 %	CO %	H_2O %	CO_2 %	MeOH %	DME %	N_2 %	H_2 %	CO %	H_2O %	CO_2 %	MeOH %	DME %	N_2 %
37.7	15.2	0.8	46.2	180	0.20	50	41.47	15.79	0.09	0.75	0.16	0.04	41.71	0.35	0.19	0.01	0.01	0.01	0.00	0.53
37.7	15.2	0.8	46.2	190	0.20	50	42.04	15.95	0.10	0.83	0.23	0.08	40.77	0.07	0.00	0.01	0.02	0.03	0.01	0.14
37.7	15.2	0.8	46.2	200	0.20	50	41.75	15.63	0.10	0.92	0.32	0.20	41.09	0.02	0.05	0.01	0.03	0.06	0.02	0.05
37.7	15.2	0.8	46.2	210	0.20	50	41.23	15.07	0.11	1.10	0.38	0.41	41.70	0.18	0.15	0.01	0.04	0.08	0.05	0.14
37.7	15.2	0.8	46.2	220	0.20	50	40.62	14.25	0.12	1.40	0.43	0.75	42.43	0.31	0.27	0.02	0.08	0.09	0.09	0.31
37.7	15.2	0.8	46.2	230	0.20	50	39.78	13.05	0.17	1.86	0.47	1.26	43.42	0.47	0.42	0.03	0.12	0.08	0.14	0.51
37.7	15.2	0.8	46.2	240	0.20	50	38.56	11.35	0.23	2.50	0.52	1.97	44.86	0.69	0.59	0.04	0.16	0.06	0.21	0.81
37.7	15.2	0.8	46.2	250	0.20	50	37.11	9.26	0.28	3.30	0.57	2.88	46.61	0.76	0.63	0.01	0.17	0.03	0.22	0.97
37.7	15.2	0.8	46.2	260	0.20	50	35.63	7.09	0.28	4.15	0.59	3.73	48.53	0.85	0.58	0.01	0.10	0.01	0.14	1.16
37.7	15.2	0.8	46.2	270	0.20	50	34.51	5.61	0.27	4.75	0.58	4.40	49.89	0.79	0.30	0.00	0.07	0.00	0.00	1.16
37.7	15.2	0.8	46.2	280	0.20	50	34.11	5.17	0.26	4.89	0.56	4.60	50.40	0.66	0.13	0.01	0.14	0.00	0.09	1.01
37.7	15.2	0.8	46.2	290	0.20	50	34.53	5.63	0.27	4.75	0.54	4.41	49.86	0.57	0.03	0.02	0.17	0.00	0.12	0.89
37.7	15.2	0.8	46.2	300	0.20	50	35.26	6.58	0.29	4.40	0.51	4.01	48.96	0.45	0.02	0.00	0.17	0.00	0.14	0.72
37.5	14.9	1.0	46.7	180	0.60	50	41.22	15.74	0.07	0.93	0.09	0.01	41.93	0.07	0.04	0.03	0.00	0.07	0.03	0.02
37.5	14.9	1.0	46.7	190	0.60	50	41.13	15.70	0.09	0.93	0.13	0.02	42.00	0.02	0.01	0.01	0.00	0.01	0.00	0.04
37.5	14.9	1.0	46.7	200	0.60	50	41.03	15.62	0.11	0.94	0.18	0.04	42.08	0.02	0.01	0.00	0.00	0.01	0.00	0.03
37.5	14.9	1.0	46.7	210	0.60	50	40.86	15.47	0.11	0.98	0.23	0.09	42.26	0.01	0.00	0.00	0.00	0.02	0.00	0.00
37.5	14.9	1.0	46.7	220	0.60	50	40.62	15.20	0.12	1.07	0.26	0.19	42.55	0.07	0.04	0.00	0.01	0.02	0.00	0.08
37.5	14.9	1.0	46.7	230	0.60	50	40.35	14.84	0.13	1.20	0.28	0.35	42.85	0.09	0.07	0.00	0.02	0.03	0.01	0.09
37.5	14.9	1.0	46.7	240	0.60	50	39.93	14.27	0.13	1.42	0.27	0.58	43.40	0.14	0.12	0.00	0.03	0.01	0.03	0.18
37.5	14.9	1.0	46.7	250	0.60	50	39.38	13.49	0.16	1.72	0.28	0.91	44.06	0.18	0.18	0.01	0.06	0.01	0.05	0.25
37.5	14.9	1.0	46.7	260	0.60	50	38.49	12.39	0.22	2.11	0.31	1.32	45.15	0.37	0.30	0.03	0.08	0.02	0.07	0.52
37.5	14.9	1.0	46.7	270	0.60	50	37.45	11.15	0.27	2.57	0.34	1.82	46.40	0.91	0.88	0.03	0.29	0.04	0.24	1.19
37.5	14.9	1.0	46.7	280	0.60	50	36.68	10.12	0.36	2.97	0.38	2.27	47.23	1.01	0.95	0.03	0.30	0.04	0.25	1.33
37.5	14.9	1.0	46.7	290	0.60	50	36.06	9.38	0.38	3.23	0.40	2.57	47.97	1.11	1.01	0.01	0.31	0.04	0.26	1.51
37.5	14.9	1.0	46.7	300	0.60	50	35.92	9.20	0.39	3.30	0.40	2.65	48.14	1.10	0.98	0.01	0.29	0.04	0.23	1.51
38.2	15.2	1.0	45.7	260	0.40	40	38.04	12.07	0.23	2.15	0.25	1.54	44.33	0.53	0.56	0.03	0.20	0.05	0.06	1.79
38.2	15.2	1.0	45.7	260	0.40	45	37.38	11.41	0.24	2.33	0.31	1.79	45.00	0.35	0.46	0.02	0.19	0.06	0.02	1.64
38.2	15.2	1.0	45.7	260	0.40	50	36.72	10.70	0.29	2.55	0.39	2.07	45.65	0.49	0.68	0.01	0.29	0.09	0.12	1.81
38.2	15.2	1.0	45.7	260	0.40	55	36.07	10.06	0.31	2.73	0.47	2.32	46.25	0.39	0.73	0.03	0.36	0.14	0.17	1.71
38.2	15.2	1.0	45.7	260	0.40	60	35.31	9.35	0.29	2.92	0.55	2.59	46.78	0.27	0.74	0.01	0.40	0.15	0.17	1.82

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