

## Electronic Supplementary Information (ESI)

### Synthesis of sulphur-modified bifunctional hydrotalcites and study of their surface characteristics by inverse gas chromatography

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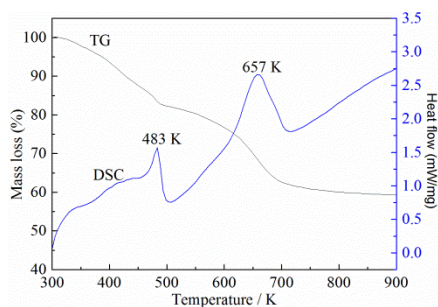


Fig.S1 The TG-DSC analysis of S/LDH-823 catalyst

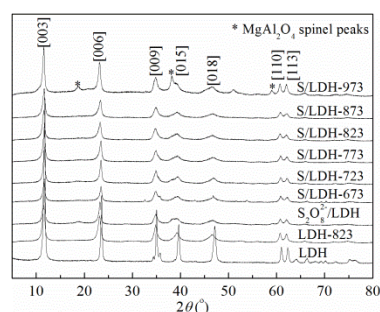


Fig.S2 The XRD patterns of the seven samples

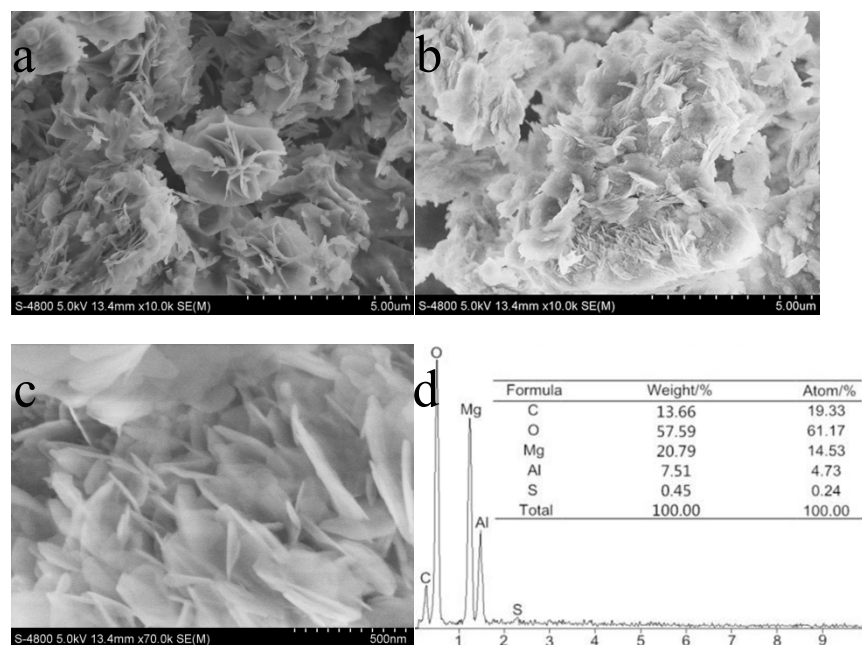


Fig.S3 (a) SEM images of LDH (b) SEM images of S/LDH-823 (c) Enlarged SEM images of S/LDH-823 (d) SEM-EDS spectrum of S/LDH-823

**Table S1** The properties of probes used for IGC Measurements

Probe	Character	$a \times 10^{20} / \text{m}^2$	$\gamma_l^d / \text{mJ} \cdot \text{m}^{-2}$	AN* / $\text{KJ} \cdot \text{mol}^{-1}$	DN / $\text{KJ} \cdot \text{mol}^{-1}$	$2aN(\gamma_l^d)^{0.5}$
n-hexane	apolar	0.515	18.4	--	--	2.6598
n-heptane	apolar	0.570	20.3	--	--	3.0921
n-octane	apolar	0.630	21.3	--	--	3.4896
n-nonane	apolar	0.690	22.7	--	--	3.9581
DCM	acidic	0.315	27.6	16.32	0.00	1.9925
TCM	acidic	0.440	25.9	22.60	0.00	2.6960
DEE	amphoterich	0.470	47.0	5.86	80.37	2.1916
THF	basic	0.450	22.5	2.09	83.72	2.5700
Acet	amphoterich	0.480	19.6	6.28	71.58	2.5586

\*DCM-Dichloromethane; TCM-Chloroform; THF-Tetrahydrofuran; DEE-Diethylether; Acet-Ethylacetate.

\*The corresponding donor and corrected acceptor numbers were taken from reference [1].

[1] F. L. Riddle and F. M. Fowkes, *J. Am. Chem. Soc.*, 1990, 112, 3259-3264.

**LDH catalyst****Table S2** The retention time,  $V_n$ , and  $RT \ln V_n$  of LDH catalyst at various temperatures

Temperature	Probe	Retention time/min	$V_n$	$RT \ln V_n$
333.15 K	n-hexane	0.076	0.1017	-6.3318
	n-heptane	0.324	0.4334	-2.3155
	n-octane	1.096	1.4662	1.0600
	n-nonane	4.082	5.4609	4.7020
	Dichloromethane(DCM)	0.020	0.0268	-10.0295
	Chloroform(TCM)	0.135	0.1806	-4.7404
	Diethylether(DEE)	0.012	0.0160	-11.4444
	Tetrahydrofuran(THF)	0.045	0.0602	-7.7834
Ethylacetate(Acet)	0.057	0.0762	-7.1286	
343.15 K	n-hexane	0.056	0.0772	-7.3087
	n-heptane	0.193	0.2659	-3.7786
	n-octane	0.610	0.8405	-0.4956
	n-nonane	2.498	3.4421	3.5265
	DCM	0.014	0.0193	-11.2637
	TCM	0.086	0.1185	-6.0848
	DEE	0.008	0.0110	-12.8603
	THF	0.030	0.0413	-9.0894
Acet	0.038	0.0524	-8.4150	
353.15 K	n-hexane	0.037	0.0525	-8.6542
	n-heptane	0.140	0.1985	-4.7470
	n-octane	0.451	0.6396	-1.3123
	n-nonane	1.369	1.9414	1.9478
	DCM	0.010	0.0142	-12.4956
	TCM	0.055	0.0780	-7.4902
	DEE	0.006	0.0085	-13.9954
	THF	0.021	0.0298	-10.3172
Acet	0.026	0.0369	-9.6901	
363.15 K	n-hexane	0.023	0.0335	-10.2503
	n-heptane	0.091	0.1327	-6.0978
	n-octane	0.275	0.4010	-2.7588
	n-nonane	0.674	0.9829	-0.0522
	DCM	0.007	0.0102	-13.8420
	TCM	0.033	0.0481	-9.1604
	DEE	0.004	0.0058	-15.5316
	THF	0.014	0.0204	-11.7492
Acet	0.017	0.0248	-11.1630	

333.15 K  $y=8.4914x-28.7420$   $R^2=0.9974$ 343.15 K  $y=8.3401x-29.5357$   $R^2=0.9998$ 353.15 K  $y=8.1975x-30.2423$   $R^2=0.9942$ 363.15 K  $y=7.8847x-30.8084$   $R^2=0.9801$ **Table S3**  $\gamma_s^d$  values of LDH catalyst versus temperature

	$\gamma_s^d / \text{mJ}\cdot\text{m}^{-2}$			
Temperature / K	333.15 K	343.15 K	353.15 K	363.15 K
LDH	72.10	69.56	67.20	62.17

**Table S4** Adsorption free energy and adsorption enthalpy of polar probes on the surface of LDH catalyst

Temperature / K	$-\Delta G^{SP} / \text{KJ}\cdot\text{mol}^{-1}$				$-\Delta H_a^S / \text{KJ}\cdot\text{mol}^{-1}$
	333.15 K	343.15 K	353.15 K	363.15 K	
CH <sub>2</sub> Cl <sub>2</sub>	1.7934	1.6544	1.4132	1.2561	7.9726(R <sup>2</sup> =0.9895)
CHCl <sub>3</sub>	1.1088	0.9660	0.6516	0.3908	9.3247(R <sup>2</sup> =0.9744)
Diethyl ether	-1.3122	-1.6028	-1.7187	-2.0033	2.9666(R <sup>2</sup> =0.9420)
THF	-0.8643	-0.9878	-1.1425	-1.2045	3.0660(R <sup>2</sup> =0.9391)
Ethyl acetate	-0.1127	-0.2183	-0.4219	-0.5284	4.7289(R <sup>2</sup> =0.9697)

$$y=0.0255x+0.4955(R^2=0.9270)$$

$$K_a=0.0255$$

$$K_b=0.4955$$

$$K_a+K_b=0.5210$$

$$K_a/K_b=0.0515$$

S/LDH-673 catalyst

**Table S5** The retention time,  $V_n$ , and  $RT\ln V_n$  of S/LDH-673 catalyst at various temperatures

Temperature	Probe	Retention time/min	$V_n$	$RT\ln V_n$
333.15 K	n-hexane	0.079	0.1516	-5.2246
	n-heptane	0.234	0.6622	-1.1417
	n-octane	1.100	2.1114	2.0700
	n-nonane	3.747	7.1921	5.4648
	DCM	0.097	0.1862	-4.6561
	TCM	0.316	0.6065	-1.3849
	DEE	0.030	0.0576	-7.9065
	THF	0.138	0.2649	-3.6796
	Acet	0.158	0.3033	-3.3048
343.15 K	n-hexane	0.056	0.1107	-6.2788
	n-heptane	0.286	0.4626	-3.1991
	n-octane	0.752	1.4334	1.0271
	n-nonane	2.219	4.3870	4.2185
	DCM	0.059	0.1166	-6.1299
	TCM	0.174	0.3440	-3.0444
	DEE	0.020	0.0395	-9.2162
	THF	0.090	0.1779	-4.9252
	Acet	0.092	0.1819	-4.8625
353.15 K	n-hexane	0.042	0.0854	-7.2221
	n-heptane	0.154	0.3133	-3.4072
	n-octane	0.411	0.8362	-0.5251
	n-nonane	1.375	2.7976	3.0206
	DCM	0.039	0.0794	-7.4396
	TCM	0.094	0.1913	-4.8567
	DEE	0.013	0.0264	-10.6653
	THF	0.057	0.1160	-6.3254
	Acet	0.056	0.1139	-6.3774
363.15 K	n-hexane	0.025	0.0523	-8.9086
	n-heptane	0.083	0.1736	-5.2856
	n-octane	0.241	0.5042	-2.0673
	n-nonane	0.675	1.4123	1.0423
	DCM	0.020	0.0418	-9.5823
	TCM	0.041	0.0858	-7.4150
	DEE	0.007	0.1465	-12.7520
	THF	0.030	0.0628	-8.3581
	Acet	0.029	0.0607	-8.4605

333.15 K  $y=8.2105x-26.8018$   $R^2=0.9941$ 343.15 K  $y=8.0765x-27.4596$   $R^2=0.9914$ 353.15 K  $y=7.8291x-27.8688$   $R^2=0.9972$ 363.15 K  $y=7.6938x-29.1935$   $R^2=0.9953$ **Table S6**  $\gamma_s^d$  values of S/LDH-673 catalyst versus temperature $\gamma_s^d / \text{mJ}\cdot\text{m}^{-2}$

Temperature / K	333.15 K	343.15 K	353.15 K	363.15 K
LDH	67.41	65.23	61.29	59.19

**Table S7** Adsorption free energy and adsorption enthalpy of polar probes on the surface of S/LDH-673 catalyst

Temperature / K	$-\Delta G^{SP} / \text{KJ}\cdot\text{mol}^{-1}$				$-\Delta H_a^S / \text{KJ}\cdot\text{mol}^{-1}$
	333.15 K	343.15 K	353.15 K	363.15 K	
CH <sub>2</sub> Cl <sub>2</sub>	5.7862	5.2373	4.8297	4.2813	22.1723 (R <sup>2</sup> =0.9971)
CHCl <sub>3</sub>	3.2814	2.6410	1.9048	0.0054	28.1398 (R <sup>2</sup> =0.9941)
Diethyl ether	0.9012	0.5429	0.0452	-0.4202	15.7577 (R <sup>2</sup> =0.9935)
THF	2.0212	1.7778	1.4226	1.0623	12.7755 (R <sup>2</sup> =0.9910)
Ethyl acetate	2.4896	1.9326	1.4599	1.0476	18.4971 (R <sup>2</sup> =0.9944)

$$y=0.1195x+1.3123(0.9871)$$

$$K_a=0.1195$$

$$K_b=1.3123$$

$$K_a+K_b=1.4318$$

$$K_a/K_b=0.0911$$

### S/LDH-723 catalyst

**Table S8** The retention time,  $V_n$ , and  $RT\ln V_n$  of S/LDH-723 catalyst at various temperatures

Temperature	Probe	Retention time/min	$V_n$	$RT \ln V_n$
333.15 K	n-hexane	0.107	0.2054	-4.3843
	n-heptane	0.301	0.5777	-1.5196
	n-octane	0.819	1.5720	1.2529
	n-nonane	2.864	5.4627	4.7030
	DCM	0.203	0.3896	-2.6106
	TCM	0.583	1.1190	0.3115
	DEE	0.060	0.1152	-5.9866
	THF	0.219	0.4204	-2.4005
	Acet	0.274	0.5259	-1.7799
343.15 K	n-hexane	0.090	0.1779	-4.9252
	n-heptane	0.239	0.4725	-2.1388
	n-octane	0.618	1.2218	0.5716
	n-nonane	2.044	4.0411	3.9842
	DCM	0.137	0.2708	-3.7264
	TCM	0.403	0.7968	-0.6482
	DEE	0.042	0.0830	-7.0995
	THF	0.143	0.2827	-3.6042
	Acet	0.169	0.3341	-3.1276
353.15 K	n-hexane	0.067	0.1363	-5.8508
	n-heptane	0.164	0.3337	-3.2225
	n-octane	0.425	0.8647	-0.4267
	n-nonane	1.225	2.4924	2.6814
	DCM	0.089	0.1811	-5.0172
	TCM	0.257	0.5229	-1.9036
	DEE	0.029	0.0590	-8.3095
	THF	0.094	0.1913	-4.8567
	Acet	0.105	0.2136	-4.5318
363.15 K	n-hexane	0.053	0.1109	-6.6399
	n-heptane	0.126	0.2636	-4.0253
	n-octane	0.280	0.5858	-1.6144
	n-nonane	0.842	1.7617	1.7097
	DCM	0.059	0.1234	-6.3161
	TCM	0.167	0.3494	-3.1748
	DEE	0.019	0.0398	-9.7372
	THF	0.066	0.1381	-5.9776
	Acet	0.067	0.1402	-5.9322

333.15 K  $y=7.0005x-23.0881$   $R^2=0.9991$

343.15 K  $y=6.8623x-23.2720$   $R^2=0.9988$

353.15 K  $y=6.6138x-23.5294$   $R^2=0.9989$

363.15 K  $y=6.4045x-23.7767$   $R^2=0.9974$

**Table S9**  $\gamma_s^d$  values of S/LDH-723 catalyst versus temperature

Temperature / K	$\gamma_s^d / \text{mJ} \cdot \text{m}^{-2}$			
	333.15 K	343.15 K	353.15 K	363.15K

S/LDH-450	49.01	47.09	43.74	41.02
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**Table S10** Adsorption free energy and adsorption enthalpy of polar probes on the surface of S/LDH-723 catalyst

Temperature / K	$-\Delta G^{SP} / \text{KJ}\cdot\text{mol}^{-1}$				$-\Delta H_a^S / \text{KJ}\cdot\text{mol}^{-1}$
	333.15 K	343.15 K	353.15 K	363.15 K	
CH <sub>2</sub> Cl <sub>2</sub>	6.5290	5.8725	5.3342	4.6996	26.6000 (R <sup>2</sup> =0.9987)
CHCl <sub>3</sub>	4.8572	4.1159	3.1597	2.4121	32.4984 (R <sup>2</sup> =0.9969)
Diethyl ether	1.7592	1.1331	0.7251	0.0034	20.6286 (R <sup>2</sup> =0.9865)
THF	2.6963	2.0317	1.6752	1.3395	17.4809 (R <sup>2</sup> =0.9630)
Ethyl acetate	3.3967	2.5865	2.0755	1.4579	24.4871 (R <sup>2</sup> =0.9891)

$$y=0.1696x+1.5590(0.9864)$$

$$K_a=0.1696$$

$$K_b=1.5590$$

$$K_a+K_b=1.7286$$

$$K_a/K_b=0.1088$$

### S/LDH-773 catalyst

**Table S11** The retention time,  $V_n$ , and  $RT\ln V_n$  of S/LDH-773 catalyst at various temperatures

Temperature	Probe	Retention time/min	$V_n$	$RT\ln V_n$
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333.15 K	n-hexane	0.084	0.1472	-5.3071
	n-heptane	0.234	0.4100	-2.4694
	n-octane	0.609	1.0671	0.1799
	n-nonane	1.575	2.7598	2.8118
	DCM	0.291	0.5099	-1.8656
	TCM	0.684	1.1985	0.5016
	DEE	0.090	0.1577	-5.1160
	THF	0.317	0.5555	-1.6286
343.15 K	Acet	0.429	0.7517	-0.7905
	n-hexane	0.059	0.1065	-6.3899
	n-heptane	0.150	0.2707	-3.7278
	n-octane	0.359	0.6479	-1.2381
	n-nonane	0.938	1.6929	1.5020
	DCM	0.163	0.2942	-3.4907
	TCM	0.298	0.5378	-1.7694
	DEE	0.050	0.0902	-6.8621
353.15 K	THF	0.169	0.3050	-3.3876
	Acet	0.191	0.3447	-3.0384
	n-hexane	0.043	0.07987	-7.4206
	n-heptane	0.100	0.1857	-4.9426
	n-octane	0.242	0.4495	-2.3478
	n-nonane	0.567	1.0532	0.1521
	DCM	0.094	0.1746	-5.1243
	TCM	0.150	0.2786	-3.7521
363.15 K	DEE	0.028	0.0520	-8.6801
	THF	0.090	0.1672	-5.2520
	Acet	0.101	0.1876	-4.9134
	n-hexane	0.039	0.0745	-7.8412
	n-heptane	0.084	0.1604	-5.5247
	n-octane	0.192	0.3667	-3.0287
	n-nonane	0.450	0.8595	-0.4571
	DCM	0.068	0.1299	-6.1627
363.15 K	TCM	0.091	0.1738	-5.2830
	DEE	0.019	0.0363	-10.0124
	THF	0.062	0.1184	-6.4416
	Acet	0.064	0.1222	-6.3457

333.15 K  $y=6.2845x-21.9344$   $R^2=0.9976$

343.15 K  $y=6.0931x-22.5700$   $R^2=0.9997$

353.15 K  $y=5.8905x-23.0777$   $R^2=0.9979$

363.15 K  $y=5.7387x-23.1500$   $R^2=0.9987$

**Table S12**  $\gamma_s^d$  values of S/LDH-773 catalyst versus temperature

	$\gamma_s^d / \text{mJ} \cdot \text{m}^{-2}$			
Temperature / K	333.15 K	343.15 K	353.15 K	363.15 K
S/LDH-450	39.49	37.12	34.70	32.93

**Table S13** Adsorption free energy and adsorption enthalpy of polar probes on the surface of S/LDH-773 catalyst

Temperature / K	$-\Delta G^{SP} / \text{KJ}\cdot\text{mol}^{-1}$				$-\Delta H_a^S / \text{KJ}\cdot\text{mol}^{-1}$
	333.15K	343.15K	353.15K	363.15K	
CH <sub>2</sub> Cl <sub>2</sub>	7.5469	6.9388	6.2167	5.5529	29.8810 (R <sup>2</sup> =0.9990)
CHCl <sub>3</sub>	5.4930	4.3736	3.4448	2.3955	39.5418 (R <sup>2</sup> =0.9986)
Diethyl ether	3.0453	2.3543	1.4880	0.5607	30.7334 (R <sup>2</sup> =0.9943)
THF	4.1546	3.5231	2.6871	1.9599	28.8744 (R <sup>2</sup> =0.9968)
Ethyl acetate	5.0644	3.9418	3.0929	2.1213	37.3122 (R <sup>2</sup> =0.9964)

$$y=0.3107x+1.7619(0.9869)$$

$$K_a=0.3107$$

$$K_b=1.7619$$

$$K_a+K_b=2.0726$$

$$K_a/K_b=0.1734$$

**S/LDH-823 catalyst****Table S14** The retention time,  $V_n$ , and  $RT\ln V_n$  of S/LDH-823 catalyst at various temperatures

Temperature	Probe	Retention time/min	$V_n$	$RT\ln V_n$
333.15 K	n-hexane	0.100	0.1841	-4.6870

	n-heptane	0.250	0.4603	-2.1490
	n-octane	0.567	1.0440	0.1191
	n-nonane	1.389	2.5574	2.6008
	DCM	0.615	1.1323	0.3442
	TCM	1.017	1.8725	1.7374
	DEE	0.229	0.4216	-2.3921
	THF	0.524	0.9648	-0.0993
	Acet	0.701	1.2907	0.7068
343.15 K	n-hexane	0.091	0.1726	-5.0124
	n-heptane	0.196	0.3717	-2.8234
	n-octane	0.416	0.7889	-0.6764
	n-nonane	1.000	1.8964	1.8258
	DCM	0.450	0.8534	-0.4523
	TCM	0.585	1.1094	0.2962
	DEE	0.152	0.2883	-3.5488
	THF	0.318	0.6031	-1.4428
	Acet	0.384	0.7282	-0.9048
353.15 K	n-hexane	0.076	0.1483	-5.6030
	n-heptane	0.143	0.2791	-3.7470
	n-octane	0.286	0.5582	-1.7119
	n-nonane	0.618	1.2062	0.5503
	DCM	0.309	0.6031	-1.4848
	TCM	0.321	0.6265	-1.3729
	DEE	0.098	0.1913	-4.8565
	THF	0.190	0.3708	-2.9127
	Acet	0.210	0.4099	-2.6188
363.15 K	n-hexane	0.068	0.1365	-6.0131
	n-heptane	0.118	0.2368	-4.3490
	n-octane	0.207	0.4154	-2.6521
	n-nonane	0.422	0.8470	-0.5016
	DCM	0.230	0.4616	-2.3340
	TCM	0.192	0.3853	-2.8792
	DEE	0.067	0.1345	-6.0579
	THF	0.123	0.2469	-4.2237
	Acet	0.131	0.2629	-4.0335

333.15 K  $y=5.6187x-19.5702$   $R^2=0.9991$

343.15 K  $y=5.2797x-19.0942$   $R^2=0.9997$

353.15 K  $y=4.7744x-18.3829$   $R^2=0.9983$

363.15 K  $y=4.2505x-17.4051$   $R^2=0.9975$

**Table S15**  $\gamma_s^d$  values of S/LDH-823 catalyst versus temperature

Temperature / K	$\gamma_s^d / \text{mJ} \cdot \text{m}^{-2}$			
	333.15 K	343.15 K	353.15 K	363.15 K
S/LDH-450	31.57	27.88	22.79	18.07

**Table S16** Adsorption free energy and adsorption enthalpy of polar probes on the surface of S/LDH-823 catalyst

Temperature / K	$-\Delta G^{SP} / \text{KJ}\cdot\text{mol}^{-1}$				$-\Delta H_a^S / \text{KJ}\cdot\text{mol}^{-1}$
	333.15K	343.15K	353.15K	363.15K	
CH <sub>2</sub> Cl <sub>2</sub>	8.7191	8.1221	7.3851	6.6020	32.3101 (R <sup>2</sup> =0.9968)
CHCl <sub>3</sub>	6.1596	5.1563	4.1382	3.0666	40.4524 (R <sup>2</sup> =0.9997)
Diethyl ether	4.8601	3.9744	3.0628	2.0318	36.1386 (R <sup>2</sup> =0.9984)
THF	5.0308	4.0826	3.2000	2.2576	35.6832 (R <sup>2</sup> =0.9998)
Ethyl acetate	5.9010	4.6808	3.5483	2.4963	37.7271 (R <sup>2</sup> =0.9987)

$$y=0.3774x+1.8747(0.9877)$$

$$K_a=0.3774$$

$$K_b=1.8747$$

$$K_a+K_b=2.2521$$

$$K_a/K_b=0.2013$$

**S/LDH-873 catalyst****Table S17** The retention time,  $V_n$ , and  $RT\ln V_n$  of S/LDH-873 catalyst at various temperatures

Temperature	Probe	Retention time/min	$V_n$	$RT\ln V_n$
333.15 K	n-hexane	0.153	0.2611	-3.7193
	n-heptane	0.391	0.6673	-1.1205

	n-octane	0.875	1.4933	1.1106
	n-nonane	1.957	3.3398	3.3402
	DCM	0.819	1.3977	0.9274
	TCM	1.442	2.4609	2.4943
	DEE	0.225	0.3840	-2.6511
	THF	0.712	1.2151	0.5396
	Acet	0.917	1.5650	1.2405
343.15 K	n-hexane	0.094	0.1652	-5.1364
	n-heptane	0.224	0.3938	-2.6590
	n-octane	0.444	0.7805	-0.7071
	n-nonane	0.908	1.5961	1.3340
	DCM	0.463	0.8139	-0.5876
	TCM	0.568	0.9984	-0.0044
	DEE	0.115	0.2022	-4.5611
	THF	0.315	0.5537	-1.6864
	Acet	0.381	0.6697	-1.1437
353.15 K	n-hexane	0.052	0.0941	-6.9400
	n-heptane	0.113	0.2044	-4.6612
	n-octane	0.225	0.4070	-2.6391
	n-nonane	0.410	0.7417	-0.8773
	DCM	0.189	0.3419	-3.1510
	TCM	0.212	0.3835	-2.8138
	DEE	0.047	0.0850	-7.2369
	THF	0.129	0.2334	-4.2724
	Acet	0.138	0.2496	-4.0744
363.15 K	n-hexane	0.030	0.0558	-8.7130
	n-heptane	0.055	0.1023	-6.8829
	n-octane	0.095	0.1767	-5.2328
	n-nonane	0.175	0.3256	-3.3883
	DCM	0.095	0.1767	-5.2328
	TCM	0.081	0.1509	-5.7141
	DEE	0.021	0.0391	-9.7898
	THF	0.054	0.1004	-6.9383
	Acet	0.055	0.1023	-6.8829

333.15 K  $y=5.4470x-18.0717$   $R^2=0.9955$

343.15 K  $y=4.9714x-18.1973$   $R^2=0.9938$

353.15 K  $y=4.6987x-19.2848$   $R^2=0.9904$

363.15 K  $y=4.1042x-19.5978$   $R^2=0.9996$

**Table S18**  $\gamma_s^d$  values of S/LDH-873 catalyst versus temperature

	$\gamma_s^d / \text{mJ}\cdot\text{m}^{-2}$			
Temperature / K	333.15 K	343.15 K	353.15 K	363.15 K
S/LDH-450	29.67	24.71	22.08	16.84

**Table S19** Adsorption free energy and adsorption enthalpy of polar probes on the surface of S/LDH-873 catalyst

Temperature / K	$-\Delta G^{SP} / \text{KJ}\cdot\text{mol}^{-1}$				$-\Delta H_a^S / \text{KJ}\cdot\text{mol}^{-1}$
	333.15K	343.15K	353.15K	363.15K	
CH <sub>2</sub> Cl <sub>2</sub>	8.1460	7.7042	6.7716	6.1874	30.8445 (R <sup>2</sup> =0.9844)
CHCl <sub>3</sub>	5.8809	4.7900	3.8033	2.8188	39.7836 (R <sup>2</sup> =0.9992)
Diethyl ether	3.4830	2.7409	1.7502	0.8132	33.4512 (R <sup>2</sup> =0.9953)
THF	4.6125	3.7344	2.9367	2.1117	32.2674 (R <sup>2</sup> =0.9995)
Ethyl acetate	5.3755	4.3338	3.1883	2.2139	40.8128 (R <sup>2</sup> =0.9990)

$$y=0.3381x+1.8519(0.9865)$$

$$K_a=0.3381$$

$$K_b=1.8519$$

$$K_a+K_b=2.1900$$

$$K_a/K_b=0.1826$$

### LDH-823 catalyst

**Table S20** The retention time,  $V_n$ , and  $RT\ln V_n$  of LDH-823 catalyst at various temperatures

Temperature	Probe	Retention time/min	$V_n$	$RT\ln V_n$
333.15 K	n-hexane	0.147	0.2604	-3.7262
	n-heptane	0.428	0.7583	-0.7662
	n-octane	1.025	1.8161	1.6528

	n-nonane	3.077	5.4519	4.6975
	DCM	0.951	1.6850	1.4452
	TCM	1.612	2.8562	2.9069
	DEE	0.043	0.0762	-7.1310
	THF	0.154	0.2729	-3.5974
	Acet	0.190	0.3366	-3.0156
343.15 K	n-hexane	0.090	0.1642	-5.1534
	n-heptane	0.228	0.4161	-2.5015
	n-octane	0.556	1.0147	0.0416
	n-nonane	1.478	2.6974	2.8309
	DCM	0.470	0.8578	-0.4378
	TCM	0.794	1.4490	1.0582
	DEE	0.024	0.0438	-8.9244
	THF	0.090	0.1642	-5.1534
	Acet	0.097	0.1770	-4.9398
353.15 K	n-hexane	0.054	0.1014	-6.7191
	n-heptane	0.124	0.2329	-4.2784
	n-octane	0.274	0.5146	-1.9505
	n-nonane	0.717	1.3467	0.8739
	DCM	0.217	0.4076	-2.6353
	TCM	0.268	0.5034	-2.0155
	DEE	0.012	0.02254	-11.4352
	THF	0.048	0.0902	-7.0649
	Acet	0.050	0.0939	-6.9451
363.15 K	n-hexane	0.035	0.0676	-8.1343
	n-heptane	0.076	0.1468	-5.7933
	n-octane	0.162	0.3129	-3.5081
	n-nonane	0.379	0.7320	-0.9420
	DCM	0.099	0.1912	-4.9950
	TCM	0.102	0.1970	-4.9049
	DEE	0.008	0.0154	-12.5904
	THF	0.029	0.0560	-8.7021
	Acet	0.027	0.0521	-8.9178

333.15 K  $y=6.4523x-20.8274$   $R^2=0.9993$

343.15 K  $y=6.1702x-21.5567$   $R^2=0.9997$

353.15 K  $y=5.8508x-22.3255$   $R^2=0.9996$

363.15 K  $y=5.5579x-22.9351$   $R^2=0.9998$

**Table S21**  $\gamma_s^d$  values of LDH-823 catalyst versus temperature

	$\gamma_s^d / \text{mJ}\cdot\text{m}^{-2}$			
Temperature / K	333.15 K	343.15 K	353.15 K	363.15 K
S/LDH-450	41.63	38.07	34.23	30.89

**Table S22** Adsorption free energy and adsorption enthalpy of polar probes on the surface of LDH-823 catalyst

	$-\Delta G^{SP} / \text{KJ}\cdot\text{mol}^{-1}$	$-\Delta H_a^S / \text{KJ}\cdot\text{mol}^{-1}$
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Temperature / K	333.15K	343.15K	353.15K	363.15K	
CH <sub>2</sub> Cl <sub>2</sub>	9.4164	8.8248	8.0325	6.8660	37.4520 (R <sup>2</sup> =0.9788)
CHCl <sub>3</sub>	6.3389	5.9800	4.5362	3.0461	43.9359 (R <sup>2</sup> =0.9312)
Diethyl ether	-0.4445	-0.8903	-1.6323	-1.8360	16.0085 (R <sup>2</sup> =0.9354)
THF	0.6476	0.5459	0.2240	-0.0508	8.6862 (R <sup>2</sup> =0.9469)
Ethyl acetate	1.3030	0.8298	0.4105	-0.2031	17.7204 (R <sup>2</sup> =0.9650)

$$y=0.0508x+2.1227(0.9558)$$

$$K_a=0.0508$$

$$K_b=2.1227$$

$$K_a+K_b=2.1735$$

$$K_a/K_b=0.0239$$

**Table S23**  $\gamma_s^d$  values of these catalysts versus temperature

Temperature / K	$\gamma_s^d / \text{mJ}\cdot\text{m}^{-2}$			
	333.15 K	343.15 K	353.15 K	363.15 K
LDH	72.10	69.56	67.20	62.17
LDH-823	42.06	38.07	34.13	30.95
S/LDH-673	67.41	65.23	61.29	59.19
S/LDH-723	49.01	47.09	43.74	41.02
S/LDH-773	39.49	37.12	34.70	32.93
S/LDH-823	31.57	27.88	22.79	18.07
S/LDH-873	29.67	24.71	22.08	16.84