Homogeneity of Metal-Organic Frameworks with mixed linkers

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Table S1. Crystal data for A (Al-MIL-53)

	Structure lt, np, monoclinic Cc SG 9					Structure ht, lp, orthorhombic			
Temperature	a (Å)	b(Å)	c(Å)	Bêta	V(Å)3	a (Å)	b(Å)	c(Å)	V(Å)3
°C	19,5130	7,6120	6,5760	104,24	946,8	6,6085	16,6750	12,8130	1412,0
80						6,6240	16,7871	12,7463	1417,4
100						6,6180	16,7728	12,7525	1415,6
120						6,6171	16,7538	12,7708	1415,8
140						6,6155	16,7477	12,7725	1415,1
160						6,6142	16,7393	12,7719	1414,1
180						6,6117	16,7359	12,7702	1413,0
200						6,6099	16,7292	12,7722	1412,3
220						6,6089	16,7283	12,7727	1412,1
240						6,6070	16,7258	12,7739	1411,6
260						6,6051	16,7227	12,7737	1410,9
280						6,6039	16,7189	12,7764	1410,6
300						6,6032	16,7242	12,7764	1410,9
320						6,6031	16,7237	12,7702	1410,2
340						6,6021	16,7180	12,7721	1409,7
360						6,6003	16,7128	12,7740	1409,1
380						6,6001	16,7109	12,7769	1409,2
400						6,5995	16,7026	12,7821	1409,0
420						6,5975	16,6817	12,8018	1408,9
440						6,5932	16,6178	12,8604	1409,0
460						6,5869	16,5647	12,9199	1409,7
480						6,5798	16,5216	12,9680	1409,7

Table S2. Crystal data for B (Al-MIL-53-11.1%NH2)

	Structure lt, np, monoclinic Cc SG 9					Structure ht, lp, orthorhombic			
Temperature	a (Å)	b(Å)	c(Å)	Bêta	V(Å)3	a (Å)	b(Å)	c(Å)	V(Å)3
°C	19,5130	7,6120	6,5760	104,24	946,8	6,6085	16,6750	12,8130	1412,0
25	19,590	7,628	6,597	104,67	953,7				
40	19,559	7,345	6,594	104,72	916,2				
60	19,684	7,295	6,619	104,95	918,3				
80	19,705	7,337	6,623	105,09	924,5				
100	19,751	7,395	6,629	105,26	934,0	6,653	16,821	12,818	1434,4
120	19,779	7,443	6,633	105,41	941,4	6,656	16,837	12,843	1439,4
140	19,767	7,488	6,633	105,51	946,1	6,654	16,837	12,836	1438,0
160	19,753	7,532	6,632	105,63	950,3	6,650	16,835	12,825	1435,8
180	19,768	7,572	6,629	105,85	954,6	6,648	16,815	12,815	1432,6
200	19,761	7,618	6,631	106,13	958,9	6,644	16,823	12,804	1431,1
220	19,778	7,651	6,633	106,07	964,5	6,636	16,808	12,789	1426,5
240	19,775	7,691	6,630	105,97	969,6	6,628	16,795	12,771	1421,6
260	19,761	7,741	6,620	105,92	973,8	6,623	16,786	12,764	1418,9
280	19,963	7,776	6,608	106,37	984,1	6,619	16,779	12,754	1416,4
300	19,956	7,830	6,585	106,36	987,2	6,614	16,773	12,754	1414,8
320	19,987	7,886	6,547	106,49	989,4	6,613	16,765	12,758	1414,4
340	20,038	7,929	6,521	106,55	993,2	6,613	16,766	12,759	1414,5
360	20,123	7,935	6,514	106,82	995,6	6,612	16,769	12,751	1413,8
380						6,617	16,778	12,748	1415,3
400						6,621	16,779	12,760	1417,6
420						6,624	16,773	12,776	1419,4
440						6,634	16,785	12,802	1425,5
460						6,645	16,818	12,838	1434,8
480						6,655	16,870	12,857	1443,4

Table S3. Crystal data for C (Al-MIL-53-20%NH2)

	Structure lt, np, monoclinic Cc SG 9					Structure ht, lp, orthorhombic			
Temperature	a (Å)	b(Å)	c(Å)	Bêta	V(Å)3	a (Å)	b(Å)	c(Å)	V(Å)3
°C	19,5130	7,6120	6,5760	104,24	946,8	6,6085	16,6750	12,8130	1412,0
25	19,602	7,661	6,593	104,77	957,3				
40	19,719	7,555	6,606	105,31	949,2				
60	19,729	7,364	6,617	105,20	927,8				
80	19,760	7,392	6,623	105,31	933,1	6,650	16,697	12,845	1426,26
100	19,776	7,427	6,625	105,41	938,1	6,650	16,870	12,799	1435,86
120	19,792	7,455	6,631	105,51	942,7	6,653	16,836	12,823	1436,24
140	19,789	7,491	6,631	105,62	946,8	6,651	16,846	12,798	1433,95
160	19,795	7,535	6,633	105,72	952,3	6,652	16,844	12,793	1433,47
180	19,796	7,576	6,631	105,81	956,8	6,653	16,837	12,783	1431,84
200	19,808	7,617	6,629	105,95	961,7	6,648	16,835	12,775	1429,81
220	19,799	7,657	6,628	105,98	966,1	6,639	16,822	12,768	1425,93
240	19,848	7,694	6,618	106,06	971,2	6,631	16,823	12,732	1420,21
260	19,956	7,731	6,611	106,30	979,0	6,626	16,818	12,714	1416,76
280	19,976	7,779	6,601	106,35	984,2	6,622	16,802	12,722	1415,48
300	19,990	7,832	6,582	106,38	988,7	6,617	16,797	12,723	1414,27
320	20,007	7,887	6,546	106,49	990,4	6,616	16,791	12,739	1415,23
340	20,087	7,919	6,525	106,64	994,5	6,615	16,792	12,740	1415,24
360						6,614	16,804	12,709	1412,47
380						6,619	16,814	12,699	1413,44
400						6,624	16,812	12,713	1415,76
420						6,624	16,802	12,733	1417,27
440						6,634	16,819	12,771	1424,87
450						6,657	16,878	12,838	1442,32
460						6,661	16,968	12,832	1450,35

Table S4. Crystal data for D (Al-MIL-53-50%NH2)

	Structure lt, np, monoclinic Cc SG 9					Structure ht, lp, orthorhombic			
Temperature	a (Å)	b(Å)	c(Å)	Bêta	V(Å)3	a (Å)	b(Å)	c(Å)	V(Å)3
°C	19,5130	7,6120	6,5760	104,24	946,8	6,6085	16,6750	12,8130	1412,0
25	19,675	7,669	6,588	105,07	959,8				
40	19,724	7,593	6,599	105,40	952,9				
60	19,779	7,421	6,619	105,45	936,5				
80	19,810	7,428	6,630	105,54	939,9				
100	19,809	7,454	6,632	105,61	943,1				
120	19,811	7,476	6,633	105,69	945,8				
140	19,805	7,508	6,634	105,76	949,3	6,632	16,551	13,063	1433,8
160	19,795	7,540	6,633	105,81	952,6	6,636	16,606	12,948	1427,0
180	19,796	7,572	6,633	105,89	956,2	6,642	16,739	12,800	1423,0
200	19,804	7,609	6,634	106,00	961,0	6,653	17,044	12,637	1433,0
220	19,807	7,648	6,634	106,07	965,7	6,636	16,807	12,721	1418,8
240	19,809	7,687	6,633	106,16	970,2	6,634	16,811	12,687	1414,9
260	19,817	7,726	6,632	106,27	974,7	6,639	16,912	12,607	1415,5
280	19,822	7,768	6,642	106,49	980,6	6,637	16,772	12,620	1404,7
300	19,863	7,841	6,654	106,65	992,8	6,639	16,834	12,680	1417,2
320	19,876	7,881	6,649	106,79	997,0	6,647	16,953	12,597	1419,6
340	19,913	7,929	6,653	106,92	1005,0	6,650	16,966	12,620	1423,7
360	19,979	7,962	6,658	107,07	1012,6	6,650	16,963	12,657	1427,7
380						6,650	16,951	12,814	1444,6
400						6,650	16,907	12,788	1437,9
420						6,641	16,870	12,827	1437,2

440						6,617	16,615	13,044	1434,1
460						6,590	16,464	13,200	1432,2
480						6,581	16,404	13,252	1430,6

Table S5. Crystal data for F (Al-MIL-53-NH2)

	Structure lt, np, monoclinic Cc SG 9					Structure ht, lp, orthorhombic			
Temperature	a (Å)	b(Å)	c(Å)	Bêta	V(Å)3	a (Å)	b(Å)	c(Å)	V(Å)3
°C	19,5130	7,6120	6,5760	104,24	946,8	6,6085	16,6750	12,8130	1412,0
25	19,7677	7,6680	6,5982	105,56	963,5				
40	19,7674	7,6077	6,5994	105,73	955,3				
60	19,7888	7,5030	6,6094	105,83	944,1				
80	19,8380	7,4924	6,6282	105,93	947,3				
100	19,8398	7,5121	6,6290	105,97	949,9				
120	19,8537	7,5267	6,6294	106,06	952,0				
140	19,8506	7,5538	6,6298	106,12	955,0				
160	19,8516	7,5807	6,6315	106,19	958,4				
180	19,8556	7,6084	6,6338	106,26	962,1				
200	19,8583	7,6370	6,6352	106,32	965,7				
220	19,8594	7,6661	6,6361	106,37	969,4				
240	19,8652	7,6979	6,6378	106,46	973,4				
260	19,8661	7,7298	6,6394	106,53	977,4				
280	19,8653	7,7597	6,6396	106,57	981,0				
300	19,8667	7,7892	6,6392	106,63	984,4				
320	19,8727	7,8118	6,6410	106,68	987,6				

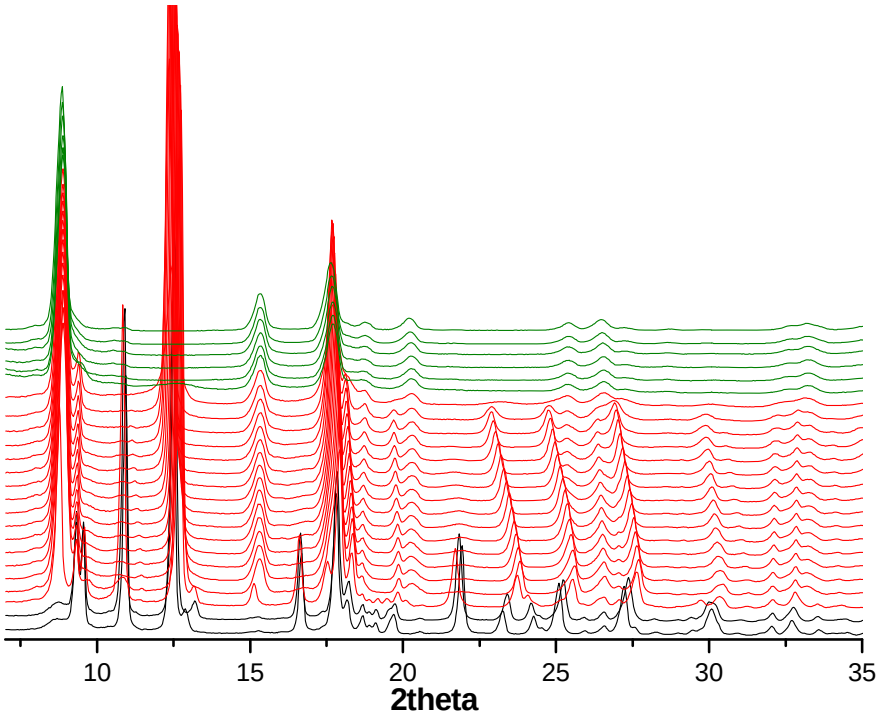

Fig. S1. Calculated thermodiffractograms of a stoichiometric mixture of Al-MIL-53 and Al-MIL-53-NH_2

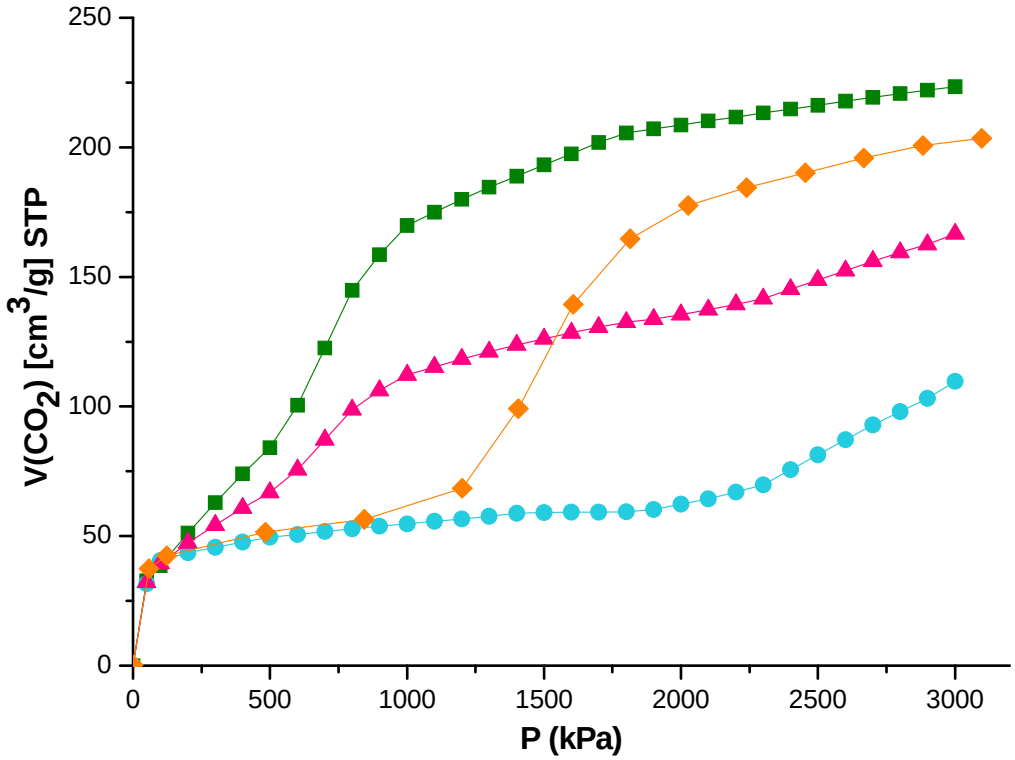

Fig. S2. Calculated CO2 isotherm of a stoichiometric mixture of Al-MIL-53 and Al-MIL-53-NH_2 (pink triangle).
Al-MIL-53 (green square), Al-MIL-53-NH2 (Blue round) and Al-MIL-53-50%NH2 (orange losange) are also showed for comparison.

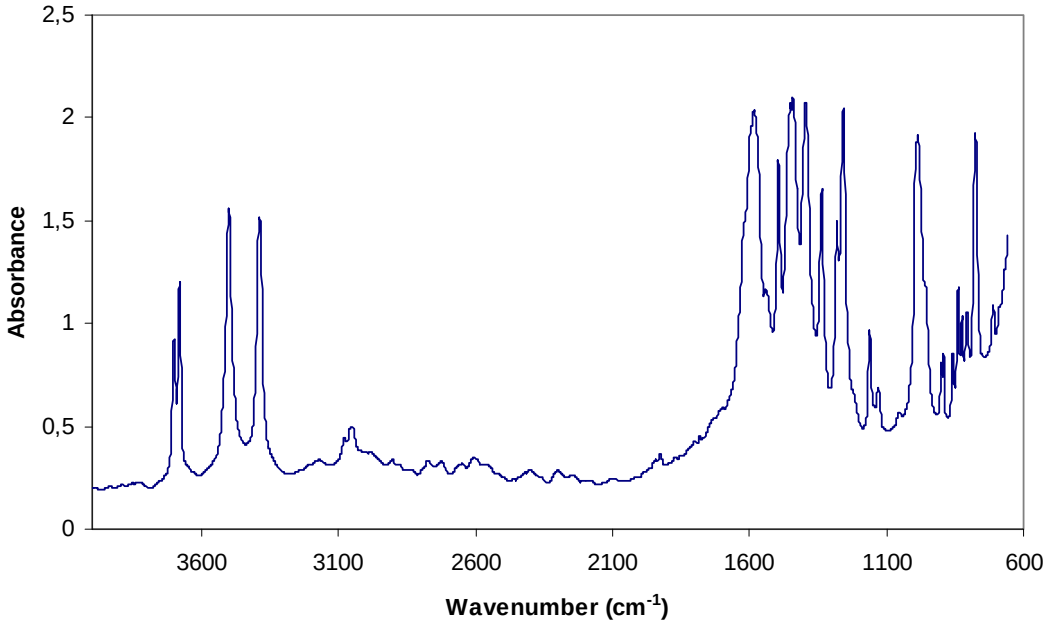

Fig. S3. Drift spectrum of Al-MIL-53-NH2 (F) after one hour desorption at 100°C.
The bands at 3500 and 3390 cm^{-1} correspond to the symmetric and asymmetric stretching of the primary amine .the doublet at 3660 and 3700cm-1 is due to the bridging OH in the chains of trans corner-sharing $AlO_4(OH)_2$ octahedra[1].The broad signals between 3100 and 2500cm^{-1} are due to the hydroxyl group in the presence of NH_2 group. Large signals between 1600 and 1300cm^{-1} are carboxylic acid function. The C=O band of free acid and DMF are not observed[2].

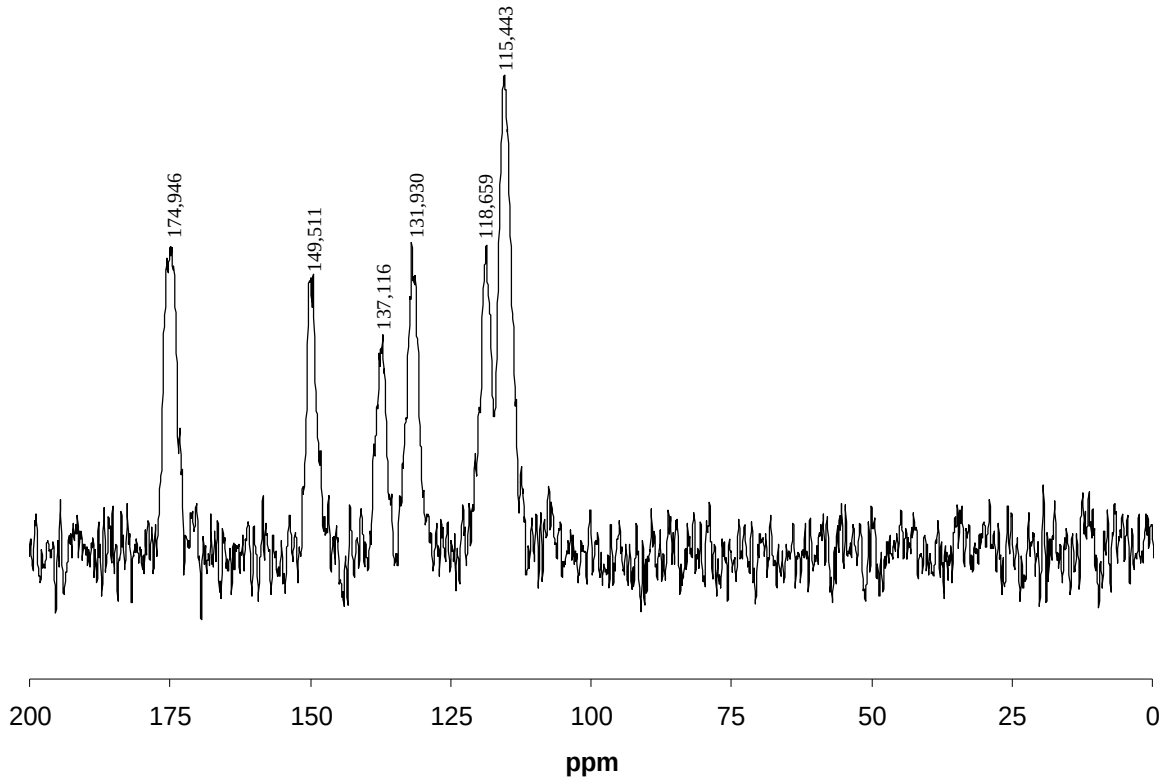

Fig. S4. 13C MAS NMR spectrum of Al-MIL-53-NH2
The 13C spectrum of the Al-MIL-53-NH2 exhibits five broad signals between 149,5 and 115,4 ppm due to the C atoms of the phenyl ring and one signal at 174,9 ppm which match with the carboxylate groups. No signals attribuable to DMF molecules are observed and the whole spectrum is consistent with the Al-MIL-53-NH2 lt one described in the literature[2].

1. S. Couck, J. F. M. Denayer, G. V. Baron, T. Rémy, J. Gascon and F. Kapteijn, *J. Am. Chem. Soc.*, 2009, **131**, 6326-6327.
2. T. Ahnfeldt, D. Gunzelmann, T. Loiseau, D. Hirsemann, J. r. Senker, G. Férey and N. Stock, *Inorg. Chem.*, 2009, **48**, 3057-3064.