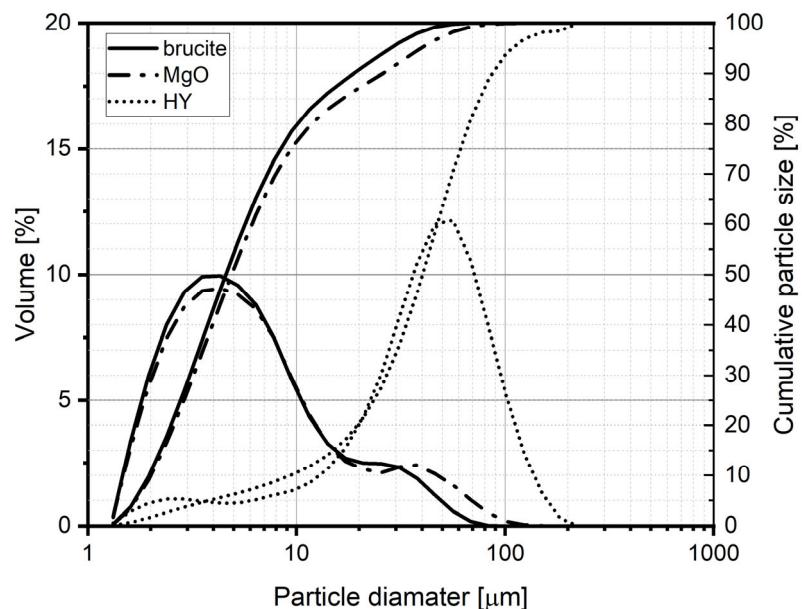


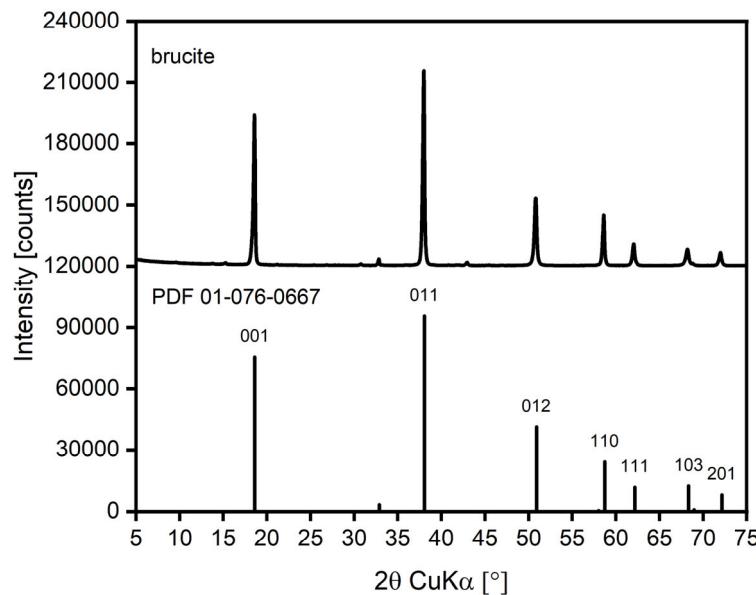
1    **1 Electronic supplementary materials (ESM)**

2    **1.1 Particle size distribution of raw materials**



3  
4    **Figure S1:** Differential and cumulative particle size distribution of raw materials.  
5

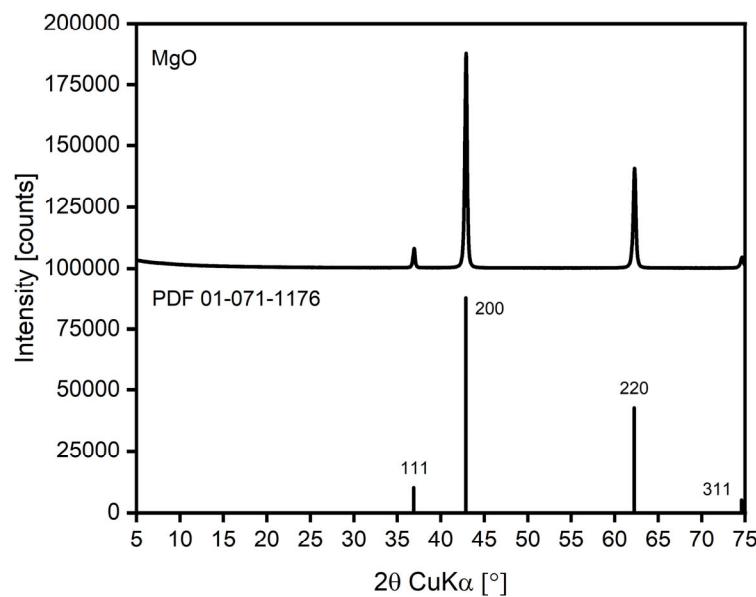
6    **1.2 X-ray diffraction patterns of raw materials**



7

8    **Figure S2:** X-ray diffraction patterns of brucite (above) and reference pattern PDF 01-076-0667 [61] (below). Only  
9    reflections with relative intensity  $\geq 5\%$  are indexed. Minor reflections not visible due to the low resolution of this  
10    image were allocated to HY, most probably a trace mineral from brucite manufacture.

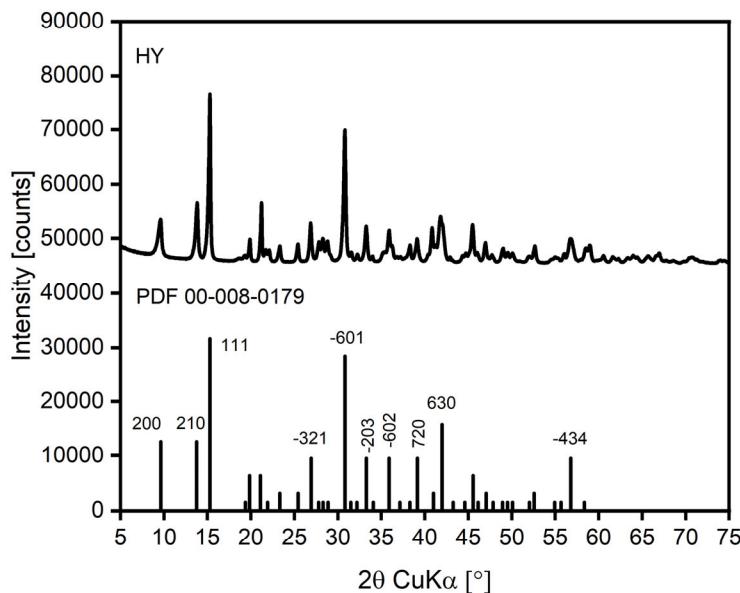
11    [61] F. Zigan, R. Rothbauer, Neutronenbeugungsmessungen am Brucit, Neues Jahrb. für Mineral. Abhandlungen,  
12    (1967) 137-143.



13

14    **Figure S3:** X-ray diffraction patterns of MgO (above) and reference pattern PDF 01-071-1176 [64] (below).

15    [64] S. Sasaki, K. Fujino, Y. Takeuchi, X-Ray Determination of Electron-Density Distributions in Oxides, MgO, MnO,  
16    CoO, and NiO, and Atomic Scattering Factors of their Constituent Atoms, Proceedings of the Japan Academy, 55  
17    (1979) 43.



18

19 **Figure S4:** X-ray diffraction patterns of HY (above) and reference pattern PDF 00-008-0179 [65] (below). Only re-  
20 flections with relative intensity  $\geq 30\%$  are indexed.

21 [65] J. Murdoch, Unit Cell of Hydromagnesite, Am. Mineral. 39 (1954) 24-29.  
22

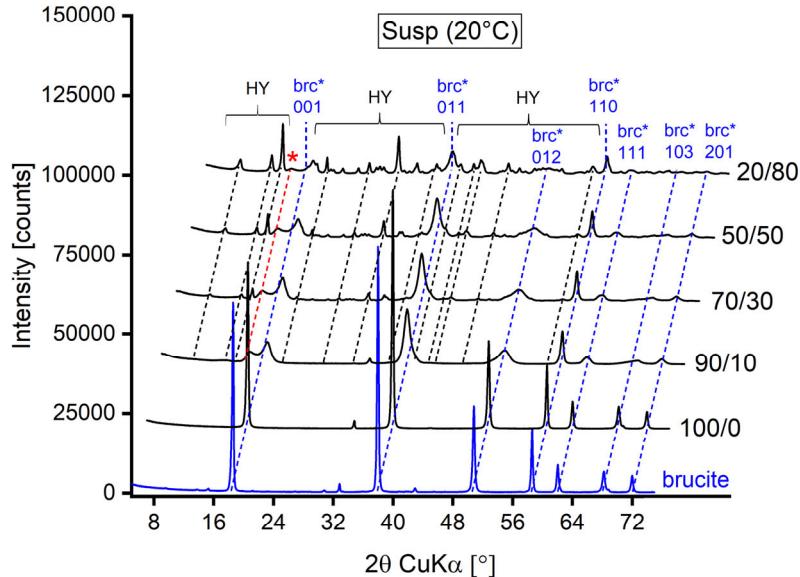
### 23 1.3 Quantification of HY content in unhydrated binder by $^{13}\text{C}$ CP-MAS NMR

24 **Table S1:** HY content of unhydrated binder determined by  $^{13}\text{C}$  CP-MAS NMR.

	theoretical HY content [%]	number of scans	sample mass [mg]	absolute signal intensity	rel. amount of carboxylic species = HY content in samples [%]
<b>HY reference</b>	100	3072	232.7	100.3	100
<b>90/10</b>	10	15537	223.0	65.8	9.3
<b>70/30</b>	30	19081	252.4	232.4	31.1
<b>50/50</b>	50	2239	249.6	41.0	50.0
<b>20/80</b>	80	3072	235.1	81.4	79.4

25

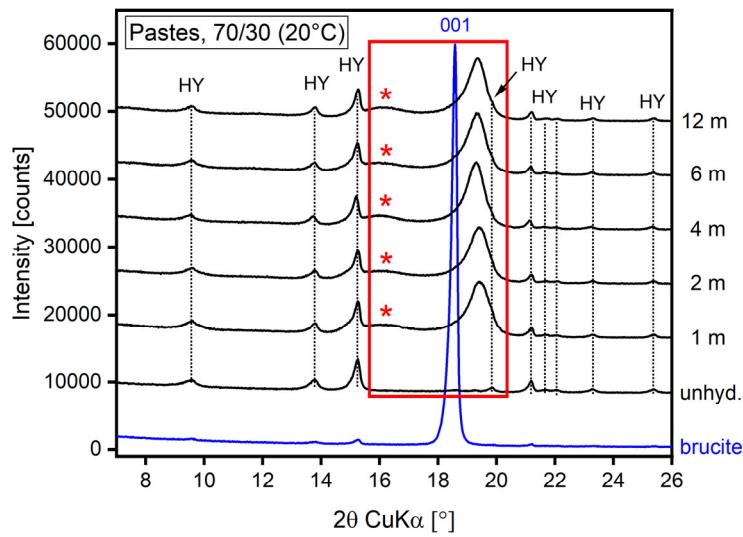
26 **1.4 X-ray diffraction patterns of suspensions**



27

28 **Figure S5:** X-ray diffraction patterns of brucite (blue) and hydrated 100/0, 90/10, 70/30, 50/50, and 20/80 suspen-  
29 sions cured for 12 months at 20°C. Red asterisk marks hump of unknown origin.

30

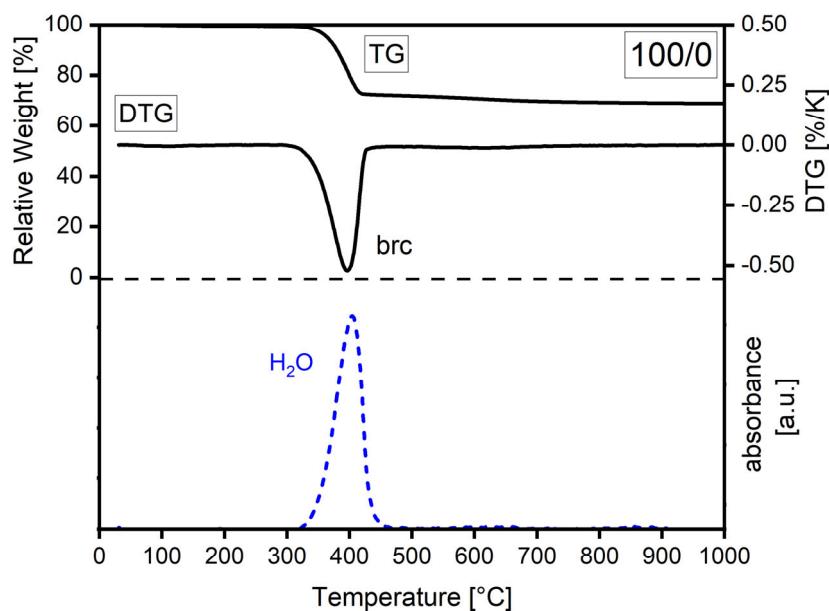


31

32 **Figure S6:** Diffraction patterns of brucite (blue), unhydrated 70/30 binder, and hydrated 70/30 pastes cured at  
33 20°C for 1, 2, 4, 6, and 12 months. Red asterisks mark the position of the hump of unknown origin. Red square  
34 marks area of 001 brucite reflection and hump shifts.

35

36    **1.5 TGA/FTIR diagrams**

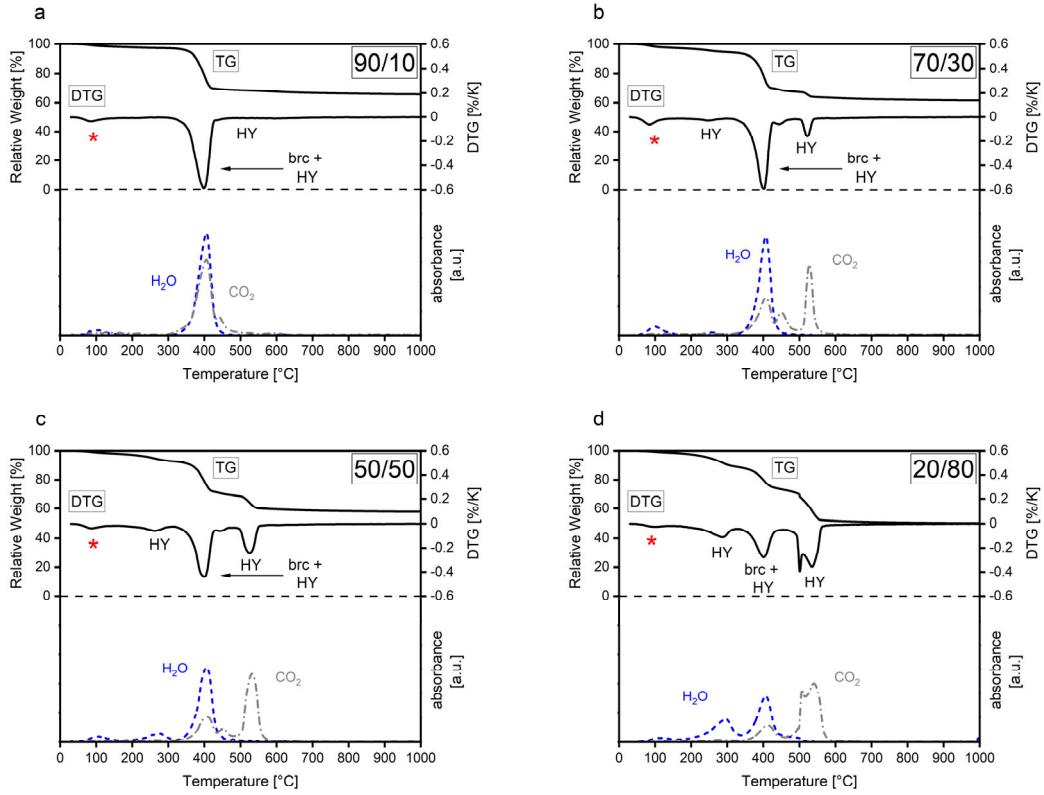


37

38    **Figure S7:** TGA/FTIR data of a hydrated 100/0 sample cured at 20°C for 12 months. Mass loss and differential mass  
39 loss curve are displayed in the upper half of the diagram. Exhaust gas analysis of the FTIR spectra recorded during  
40 TGA measurement is shown in the lower half of the diagram.

41

42

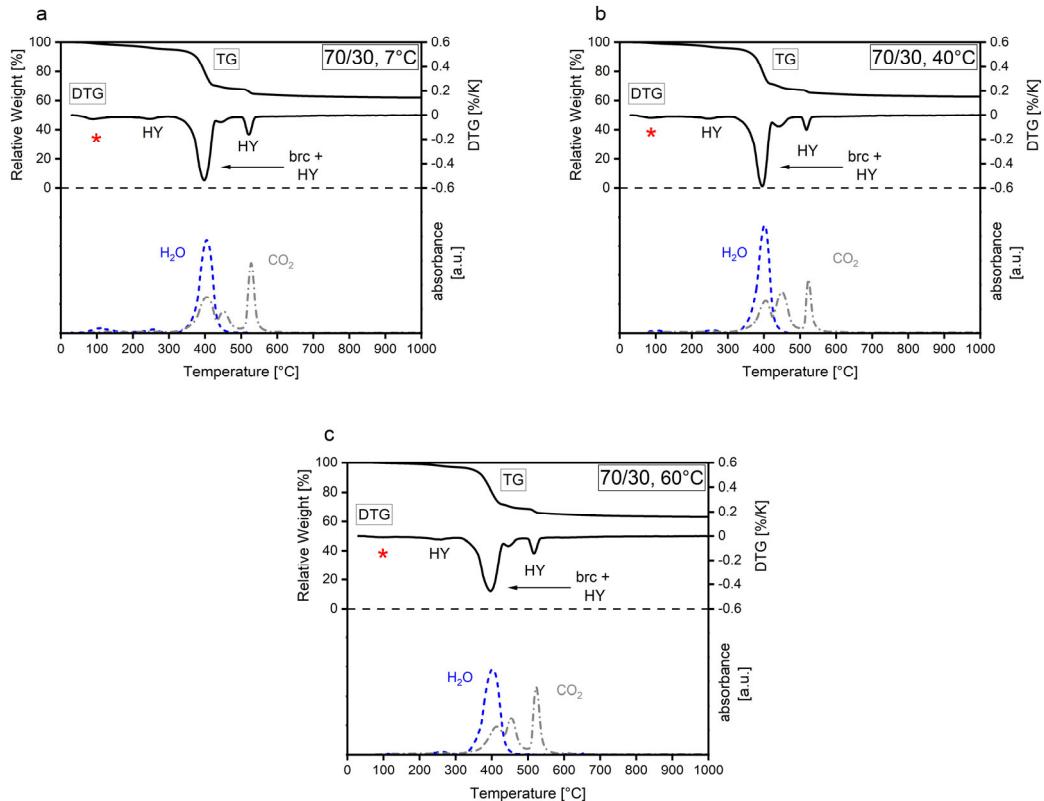


43

44 **Figure S8:** TGA/FTIR data of hydrated (a) 90/10, (b) 70/30, (c) 50/50, and (d) 20/80 suspensions. Samples were cured  
 45 at 20°C for 12 months. Mass loss and differential mass loss curves are displayed in the upper half of each diagram.  
 46 Exhaust gas analysis of FTIR spectra recorded during TGA measurement are shown in the lower halves of the dia-  
 47 grams.

48

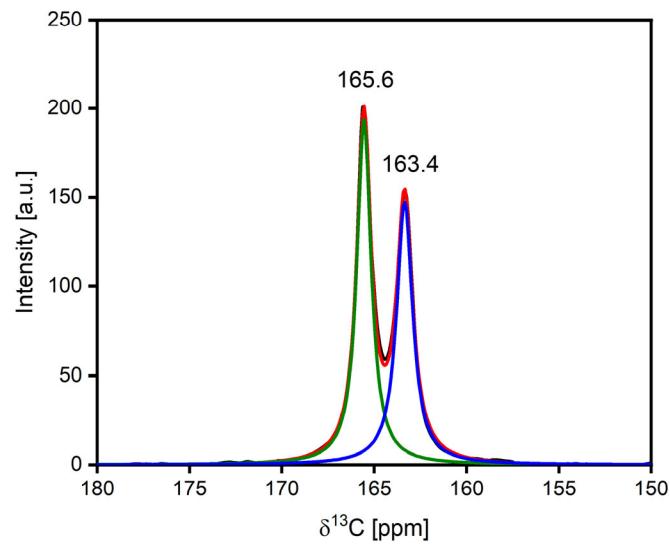
49



50

51 **Figure S9:** TGA/FTIR data of hydrated 70/30 suspensions cured at (a) 7, (b) 40, and (c) 60°C for 12 months.

52

53 **1.1  $^{13}\text{C}$  CP-MAS NMR spectra and results from line shape analysis of HY and of  
54 hydrated MgO/HY suspensions**

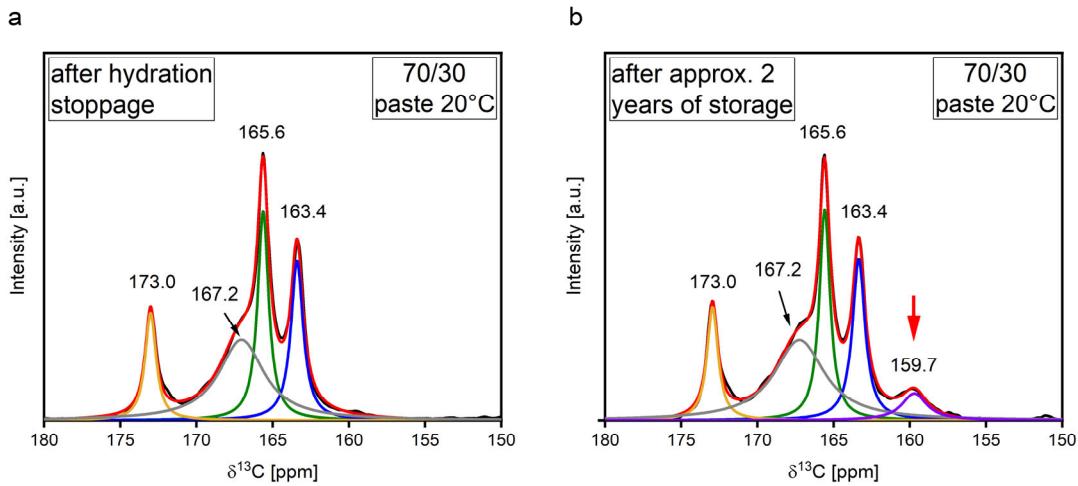
55

56 **Figure S10:**  $^{13}\text{C}$  CP-MAS NMR spectrum of HY (reference).

57

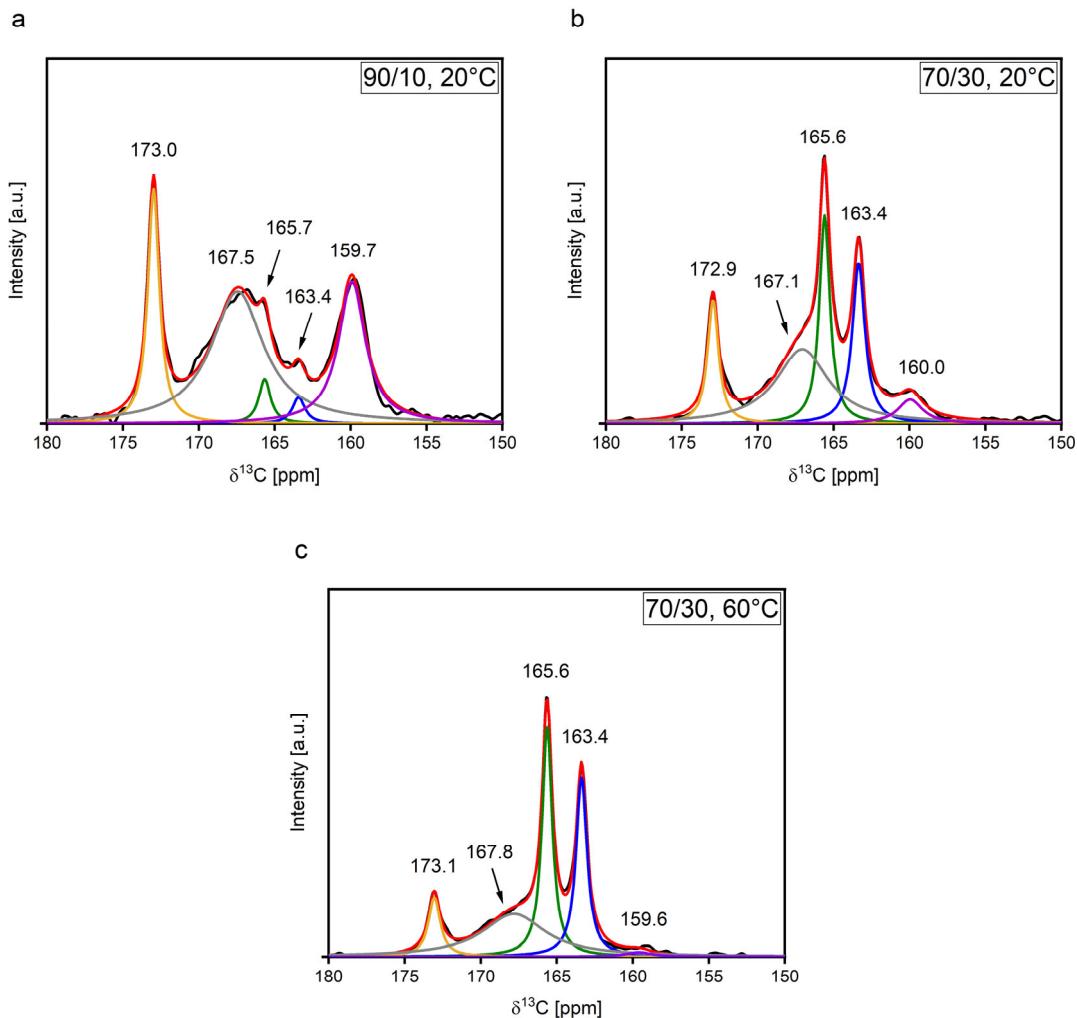
**Table S2:**  $^{13}\text{C}$  CP-MAS NMR line shape analysis results of HY (reference).

	$\delta^{13}\text{C}$ [ppm]	rel. signal intensity HY [%]
1 <sup>st</sup> HY signal	163.4	46.2
2 <sup>nd</sup> HY signal	165.6	53.8



61 **Figure S11:**  $^{13}\text{C}$  CP-MAS NMR spectra of 70/30 paste cured at 20°C for 12 months. (a) Spectra recorded directly  
62 after hydration stoppage and (b) after approximately 2 years of storage in a desiccator (spectrum shown in **Figure**  
63 **S11b** is presented in the main text as **Figure 13b**). Resonance at 159.7 ppm (marked by red arrow) is judged to be  
64 an ageing effect, not being related to the phase assemblage of a freshly stopped sample.

66



67

68 **Figure S12:** <sup>13</sup>C CP-MAS NMR spectra with simulated shapes of individual resonances (a) 90/10 suspension cured  
69 at 20°C for 12 months and 70/30 suspension cured at (b) 20°C and (c) 60°C for 12 months.

70

71 **Table S3:** <sup>13</sup>C CP-MAS NMR chemical shifts and relative amounts of carbonate species determined by line shape  
72 analysis of a hydrated 90/10 suspension cured at 20°C and of 70/30 suspensions cured at 20°C and 60°C.

suspension, 90/10 (20°C)			
	δ¹³C [ppm]	rel. amount [mol/mol-%]	
<b>carboxylic resonance A</b>	159.7	26.6	
<b>1<sup>st</sup> HY signal</b>	163.4	2.3	38.9
<b>2<sup>nd</sup> HY signal</b>	165.7	3.6	61.1
<b>sorbed CO<sub>3</sub><sup>2-</sup>/HCO<sub>3</sub><sup>-</sup></b>	167.5	48.7	
<b>carboxylic resonance B</b>	173.0	18.8	

suspension, 70/30 (20°C)			
	δ¹³C [ppm]	rel. amount [mol/mol-%]	
<b>carboxylic resonance A</b>	160.0	5.9	

<b>1<sup>st</sup> HY signal</b>	163.4	20.0	47.8
<b>2<sup>nd</sup> HY signal</b>	165.6	21.9	52.2
<b>sorbed CO<sub>3</sub><sup>2-</sup>/HCO<sub>3</sub><sup>-</sup></b>	167.1	38.8	
<b>carboxylic resonance B</b>	172.9	13.4	
<b>suspension, 70/30 (60°C)</b>			
	$\delta^{13}\text{C}$ [ppm]	rel. amount [mol/mol-%]	rel. signal intensity HY [%]
<b>carboxylic resonance A</b>	159.6	1.4	
<b>1<sup>st</sup> HY signal</b>	163.4	25.2	45.5
<b>2<sup>nd</sup> HY signal</b>	165.6	30.1	54.5
<b>sorbed CO<sub>3</sub><sup>2-</sup>/HCO<sub>3</sub><sup>-</sup></b>	167.8	34.4	
<b>carboxylic resonance B</b>	173.1	8.9	

73

74

75    **1.2 Mass balance calculations of hydrated MgO/HY suspensions**76              **Table S4:** Results of mass balance calculations of hydrated 90/10, 70/30, 50/50, 20/80 suspensions cured for 12 months.

binder composition	90/10		70/30			50/50		20/80		
curing temperature	20°C	7°C	20°C	40°C	60°C	20°C	7°C	20°C	40°C	60°C
<b>initial HY content [mass-%]</b>	10	30	30	30	30	50	80	80	80	80
<b>HY content calculated by TGA [mass-%]</b>	0.4	12.3	12.4	8.2	10.7	30.2	67.9	67.5	66.9	66.4
<b>HY content calculated by <sup>13</sup>C-NMR [mass-%]</b>	1.6	n.d.	16.2	n.d.	14.7	n.d.	n.d.	n.d.	n.d.	n.d.
<b>residual MgO [mass-%]</b>	65.1	56.7	56.2	59.1	58.6	44.8	20.9	21.2	21.8	22.3
<b>residual CO<sub>2</sub> [mass-%]</b>	2.0	3.8	3.7	5.4	4.6	3.8	2.2	2.4	2.8	3.1
<b>residual H<sub>2</sub>O [mass-%]</b>	31.3	27.2	27.7	27.2	26.1	21.1	9.0	8.9	8.5	8.2
<b>mol norm MgO [-]</b>	1	1	1	1	1	1	1	1	1	1
<b>mol norm CO<sub>2</sub> [-]</b>	0.029	0.062	0.061	0.084	0.072	0.079	0.096	0.103	0.117	0.129
<b>mol norm H<sub>2</sub>O [-]</b>	1.075	1.075	1.104	1.031	0.998	1.054	0.961	0.944	0.878	0.826

77

78 **1.3 Ion concentrations**

79 **Table S5:** Aqueous ion (Na, K, Mg, Ca, S, Cl) concentrations, inorganic carbon concentration, and pH of suspension  
 80 samples hydrated for 1, 2, 4, 6, and 12 months at 20°C. Detection limits for IC and TOC were 0.1-50 ppm and 1-50  
 81 ppm, respectively. BDL = below detection limit.

hydration time [months]	binder composition [wt%]	Na [mM]	K [mM]	Mg [mM]	Ca [mM]	S [mM]	Cl [mM]	inorganic carbon [mM]	OH <sup>-</sup> [mM]	pH <sup>1</sup>
1	100/0	0.179	0.036	0.034	0.462	0.148	0.13	BDL	0.32	10.7
1	90/10	0.39	0.031	0.994	0.04	0.329	0.093	1.088	0.12	10.2
1	70/30	0.605	0.024	1.156	0.037	0.419	0.082	1.321	0.13	10.3
1	50/50	0.797	0.018	1.225	0.04	0.501	0.067	1.412	0.11	10.2
1	20/80	1.042	0.008	1.304	0.031	0.606	0.042	1.595	0.13	10.3
2	100/0	0.178	0.032	0.06	0.309	0.155	0.124	BDL	0.26	10.6
2	90/10	0.413	0.029	1.062	0.042	0.361	0.087	1.088	0.12	10.3
2	70/30	0.633	0.023	1.231	0.039	0.463	0.075	1.337	0.14	10.3
2	50/50	0.811	0.016	1.283	0.041	0.558	0.064	1.462	0.15	10.3
2	20/80	1.055	0.008	1.374	0.041	0.7	0.042	1.586	0.15	10.3
4	100/0	0.136	0.030	0.101	0.261	0.180	0.139	BDL	0.12	10.2
4	90/10	0.356	0.027	1.239	0.036	0.391	0.105	1.272	0.08	10.1
4	70/30	0.608	0.021	1.255	0.044	0.464	0.085	1.363	0.12	10.2
4	50/50	0.782	0.016	1.286	0.045	0.542	0.074	1.454	0.13	10.3
4	20/80	1.051	0.008	1.374	0.054	0.705	0.049	1.313	0.13	10.3
6	100/0	0.107	0.017	0.167	0.251	0.209	0.154	BDL	0.07	10.0
6	90/10	0.319	0.016	1.199	0.037	0.339	0.113	1.288	0.07	10.0
6	70/30	0.573	0.012	1.219	0.038	0.411	0.108	1.388	0.11	10.2
6	50/50	0.758	0.009	1.258	0.036	0.497	0.084	1.521	0.06	9.9
6	20/80	1.028	0.005	1.364	0.036	0.675	0.056	1.604	0.08	10.1
12	100/0	0.1	0.018	0.293	0.227	0.266	0.141	BDL	0.07	10.0
12	90/10	0.314	0.019	1.371	0.048	0.359	0.102	1.424	0.11	10.2
12	70/30	0.571	0.016	1.33	0.049	0.443	0.085	1.39	0.11	10.2
12	50/50	0.767	0.012	1.357	0.048	0.523	0.073	1.49	0.12	10.3
12	20/80	1.059	0.008	1.451	0.05	0.71	0.053	1.556	0.13	10.3

82 <sup>1</sup> pH measured at 23±1°C, recalculated to 20°C

84 **Table S6:** Liquid phase analysis results of 70/30 and 20/80 suspensions hydrated for 1, 2, 4, 6, and 12 months at 7,  
 85 40, and 60°C. Detection limits for IC and TOC were 0.1-50 ppm and 1-50 ppm, respectively.

hydration time [months]	binder composition [wt%]	curing temp. [°C]	Na [mM]	K [mM]	Mg [mM]	Ca [mM]	S [mM]	Cl [mM]	inorganic carbon [mM]	OH <sup>-</sup> [mM]	pH <sup>1</sup>
1	70/30	7	0.564	0.023	1.145	0.058	0.378	0.082	1.312	0.15	10.8
2	70/30	7	0.582	0.022	1.332	0.053	0.451	0.072	1.362	0.19	10.9
4	70/30	7	0.563	0.020	1.282	0.044	0.450	0.088	1.371	0.10	10.6
6	70/30	7	0.529	0.013	1.416	0.053	0.425	0.187	1.488	0.09	10.6
12	70/30	7	0.533	0.013	1.479	0.048	0.438	0.087	1.507	0.15	10.8
1	20/80	7	0.911	0.006	1.318	0.040	0.464	0.035	1.578	0.15	10.8
2	20/80	7	0.951	0.007	1.524	0.042	0.568	0.038	1.686	0.18	10.9
4	20/80	7	0.971	0.007	1.538	0.038	0.597	0.045	1.695	0.13	10.8
6	20/80	7	0.944	0.005	1.555	0.035	0.555	0.057	1.778	0.16	10.9
12	20/80	7	0.990	0.010	1.572	0.045	0.593	0.047	1.706	0.15	10.8
1	70/30	40	0.649	0.022	1.080	0.033	0.397	0.082	1.321	0.10	9.5
2	70/30	40	0.655	0.024	1.123	0.037	0.450	0.078	1.337	0.09	9.5
4	70/30	40	0.625	0.020	1.163	0.039	0.455	0.090	1.363	0.03	9.1
6	70/30	40	0.581	0.012	1.166	0.034	0.424	0.099	1.471	0.08	9.5
12	70/30	40	0.599	0.015	1.481	0.047	0.533	0.096	1.573	0.08	9.5
1	20/80	40	1.081	0.007	1.214	0.033	0.674	0.040	1.445	0.10	9.5
2	20/80	40	1.113	0.008	1.275	0.038	0.778	0.045	1.462	0.11	9.6
4	20/80	40	1.098	0.008	1.285	0.039	0.766	0.048	1.421	0.05	9.3
6	20/80	40	1.098	0.004	1.238	0.037	0.739	0.060	1.512	0.08	9.4
12	20/80	40	1.136	0.010	1.438	0.048	0.820	0.054	1.532	0.08	9.5
1	70/30	60	0.680	0.021	1.098	0.035	0.421	0.090	1.570	0.05	8.8
2	70/30	60	0.666	0.024	1.626	0.044	0.618	0.096	1.819	0.09	9.0
4	70/30	60	0.620	0.022	1.851	0.036	0.683	0.113	1.927	0.06	8.8
6	70/30	60	0.666	0.012	1.629	0.036	0.630	0.134	2.110	0.04	8.6
12	70/30	60	0.634	0.017	1.911	0.053	0.715	0.118	1.947	0.06	8.8
1	20/80	60	1.189	0.007	1.315	0.040	0.778	0.047	1.470	0.09	9.0
2	20/80	60	1.192	0.009	1.506	0.042	0.948	0.053	1.628	0.09	9.0
4	20/80	60	1.209	0.008	1.568	0.044	0.981	0.062	1.612	0.09	9.0
6	20/80	60	1.197	0.006	1.498	0.044	0.961	0.065	1.703	0.07	8.9
12	20/80	60	1.281	0.010	1.645	0.052	1.033	0.057	1.639	0.08	8.9

86 <sup>1</sup> pH measured at 23±1°C, recalculated to 7, 40 or 60°C