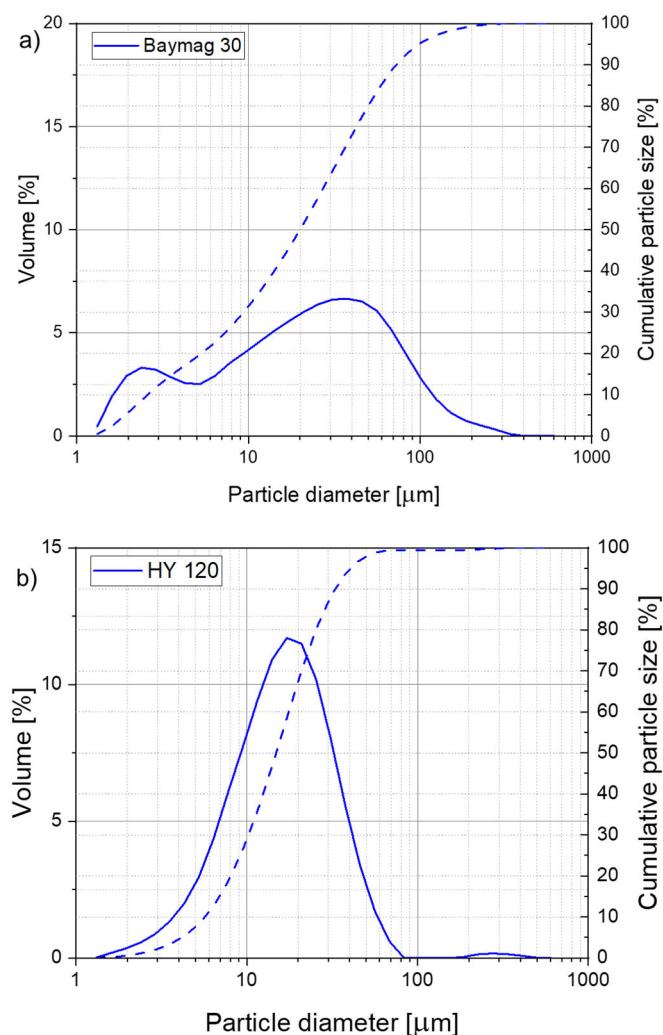


1 **Supplementary information**



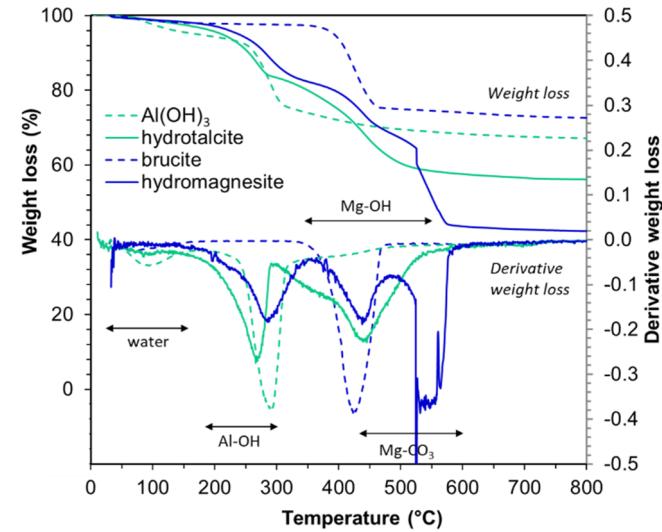
3

4 **Figure S 1: Particle size distribution (PSD) for a) industrial magnesium oxide and b)**
5 **hydromagnesite.**

6

7 **Figure S 2: Thermogravimetric analyses of the reference materials aluminium hydroxide,**
 8 **hydrotalcite ($Mg/Al=2$) from [20], brucite and hydromagnesite [30].**

9



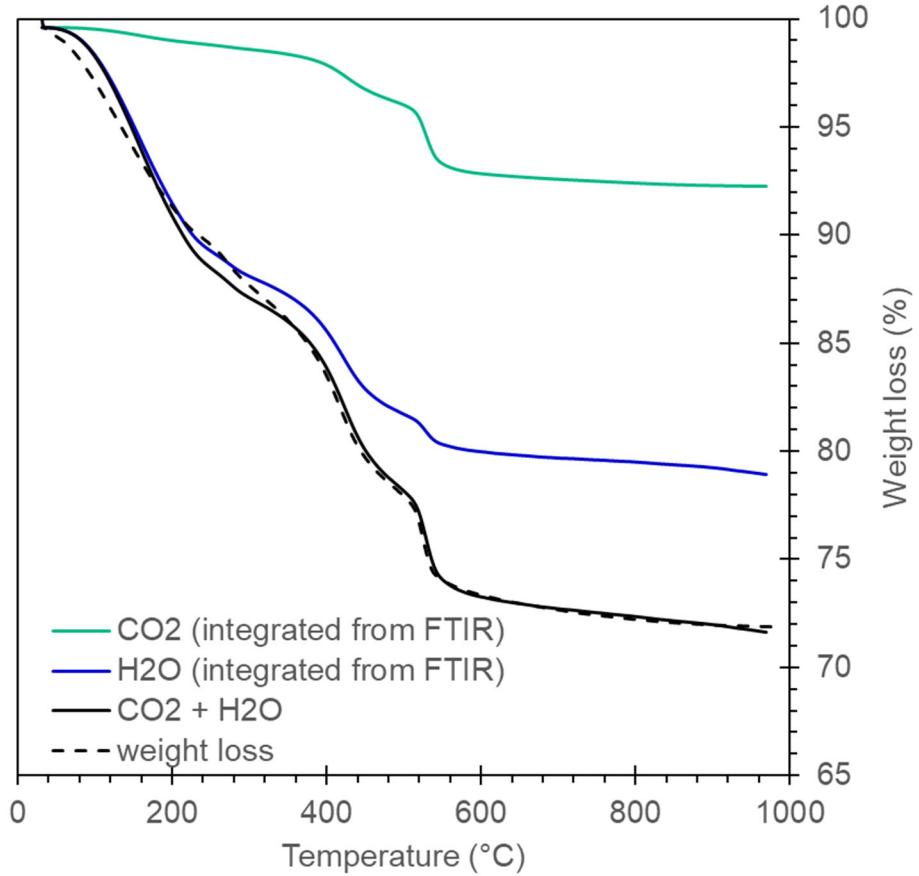
10

11 **Figure S 3: Integrated gas traces determined by FTIR and the fitted total weight loss (sum**
 12 **of the traces) compared to the weight loss for the H-3AMS sample obtained by TGA (182**
 13 **days).**

14

15 **Table S 1: Total weight loss, H_2O and CO_2 weight losses deconvoluted from TGA-FTIR**
 16 **data (3d samples were measured with classical TGA instrument).**

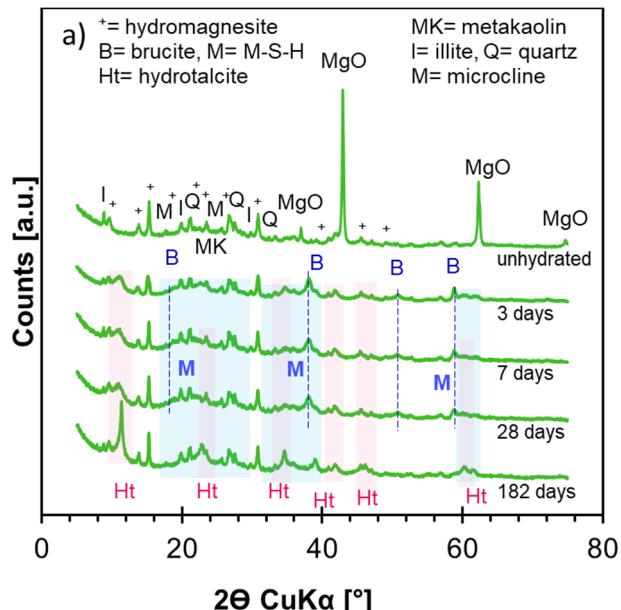
	total	H_2O	CO_2
--	-------	--------	--------



		wt% hydrated sample		
MS	3d	27.6		
	7d	23.3	21.1	2.2
	28d	30.4	28.5	2.0
	182d	28.8	26.7	2.1
H-MS	3d	32.9		
	7d	32.8	23.6	9.2
	28d	27.9	21.1	6.7
	182d	31.1	25.3	5.8
AMS	3d	24.5		
	7d	25.0	23.3	1.7
	28d	30.2	26.8	3.4
	182d	28.9	25.9	2.9
H-AMS	3d	26.4		
	7d	26.4	19.3	7.1
	28d	26.1	18.3	7.9
	182d	28.5	20.1	8.4
3AMS	3d	20.2		
	7d	21.5	20.0	1.5
	28d	20.2	19.0	1.2
	182d	24.4	22.0	2.4
H-3AMS	3d	25.2		
	7d	26.6	19.9	6.7
	28d	26.4	18.0	8.4
	182d	28.1	20.7	7.3

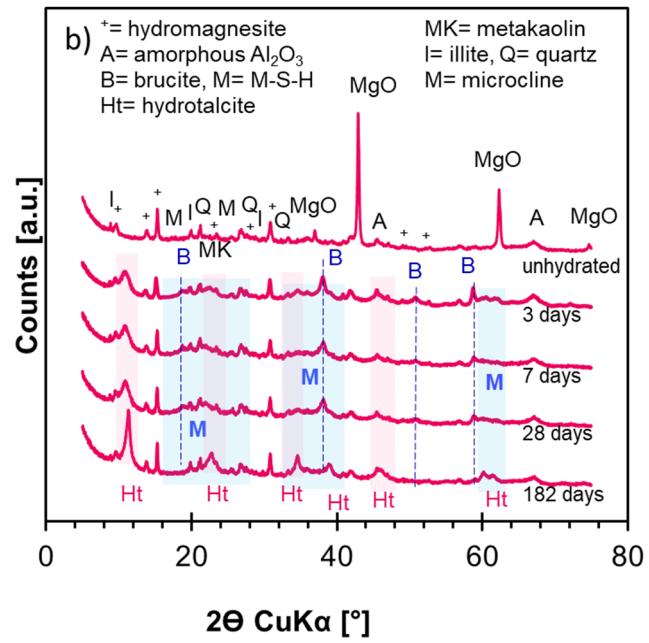
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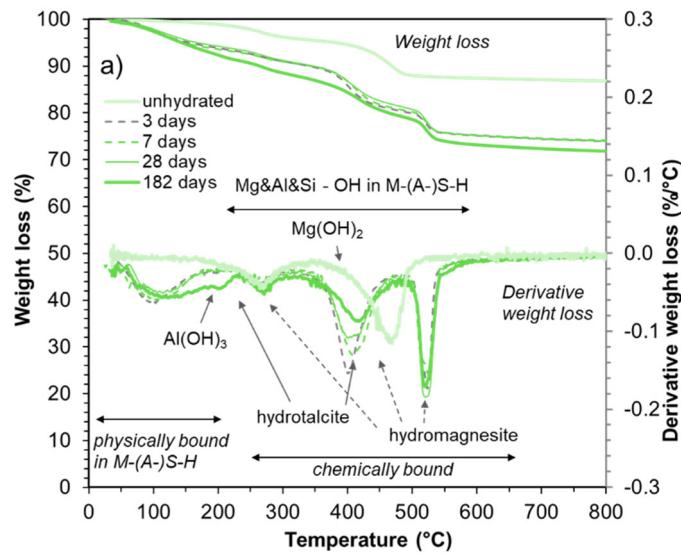
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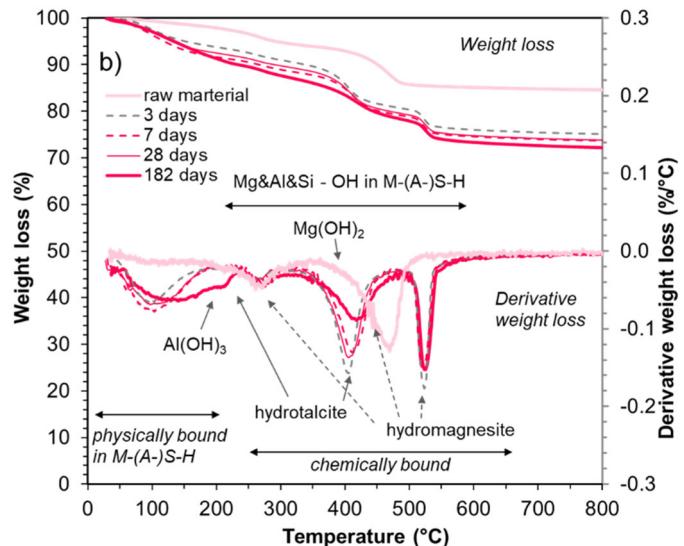
21 **Figure S 4:** X-ray diffraction (XRD) patterns of a) the H-AMS pastes (green) and b) the
 22 H-3AMS pastes (pink) after different hydration times. Light blue areas correspond to the
 23 M-S-H broad reflections, light pink to the region of the reflection related to hydrotalcite.
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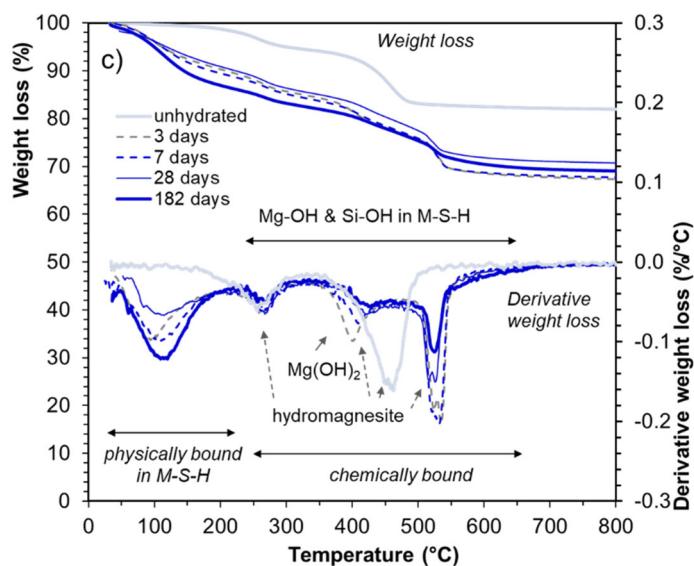
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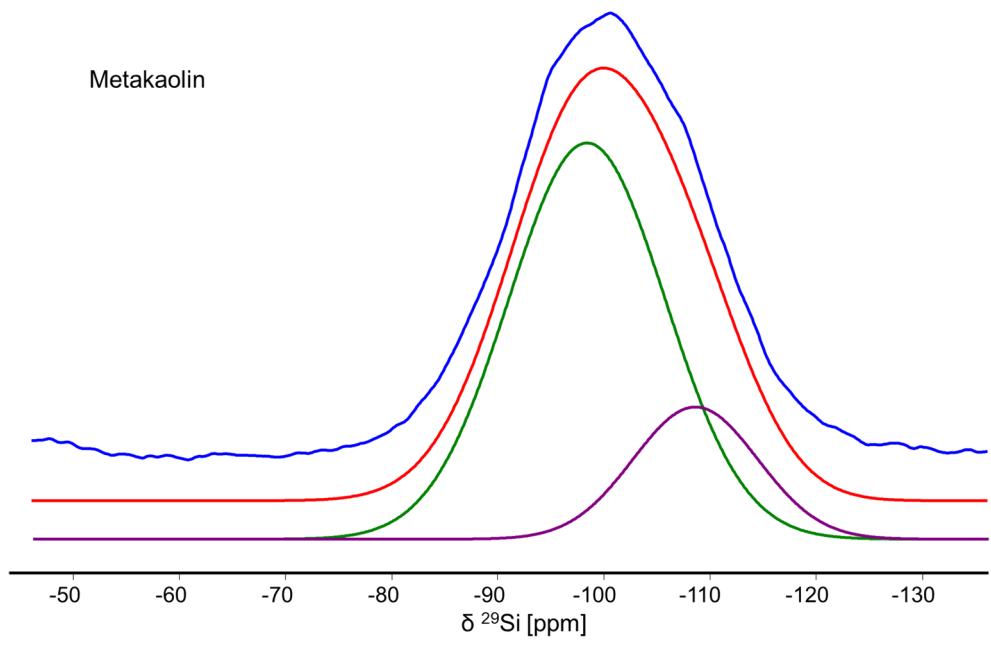
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Figure S 5: Thermogravimetric analysis (TGA) of a) H-AMS pastes (green) and b) H-3AMS pastes (pink), c) H-MS pastes (blue) (reproduced from [30]) after different hydration times.

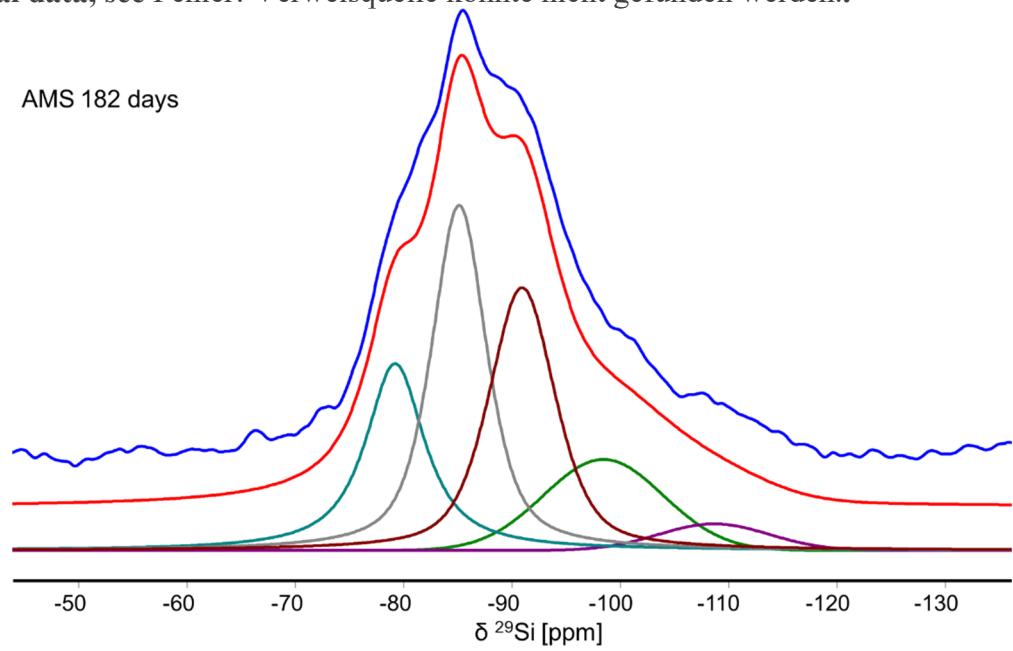




34

35 **Figure S 6: Deconvolution of the ^{29}Si MAS NMR spectrum of the Metakaolin. For**

36 numerical data, see Fehler! Verweisquelle konnte nicht gefunden werden..



37

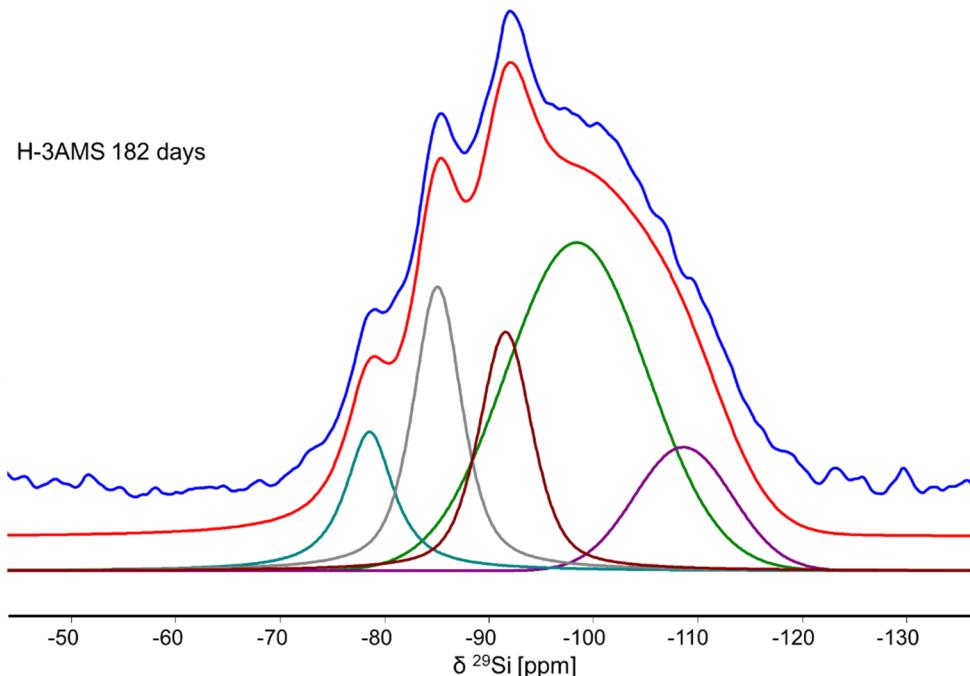
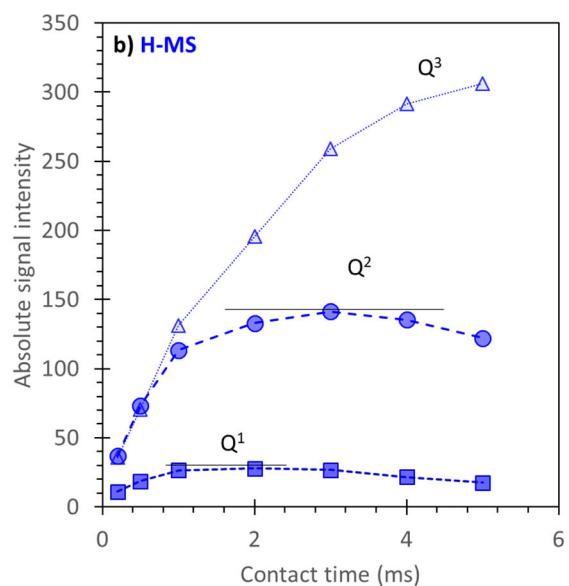
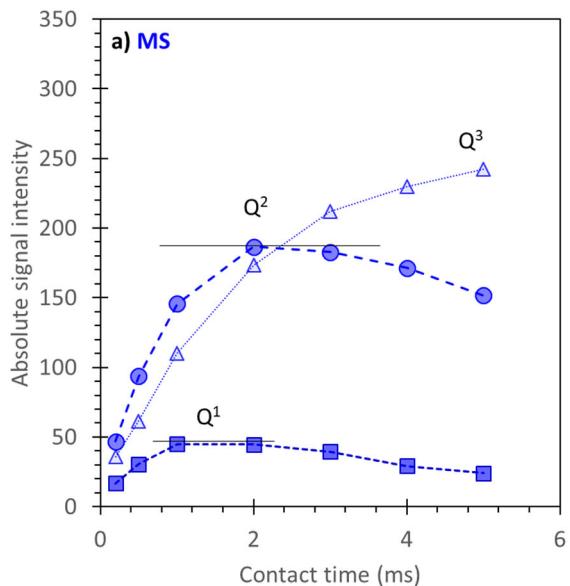


Figure S 7: Deconvolutions of the ^{29}Si MAS NMR spectra of AMS and H-3AMS pastes samples (at 182 days). For numerical data, see Fehler! Verweisquelle konnte nicht gefunden werden..

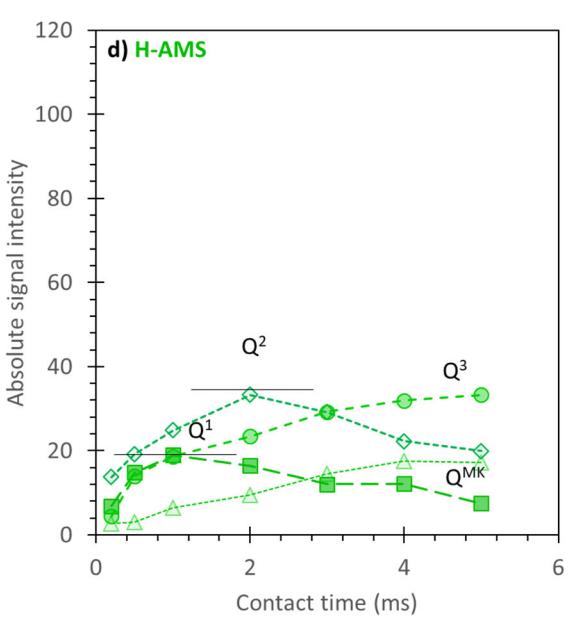
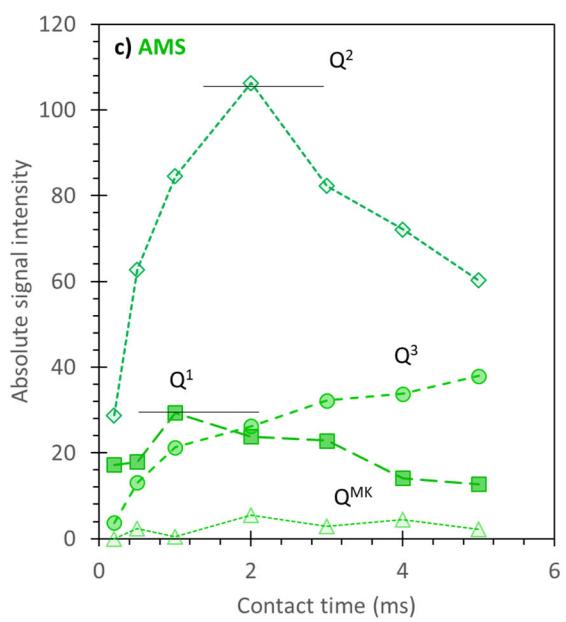
Table S 2: Normalization to amount of material and number of scans of the absolute ^{29}Si NMR signal intensities of the CP MAS NMR data recorded with 0.2 ms contact times. The absolute and normalized signal intensities of sample MS were set to 100%.

sample (182 days)	number of scans (NS)	amount of material [mg]	absolute ^{29}Si NMR signal intensity	normalized ^{29}Si NMR signal intensity
MS	7168	150.0	100.00	100.0%
H-MS	7168	127.5	71.23	83.8%
AMS	6144	154.0	43.72	49.7%
H-AMS	6144	176.2	27.89	27.7%
3AMS	7168	208.8	43.96	31.6%
H-3AMS	8192	199.5	25.85	17.0%

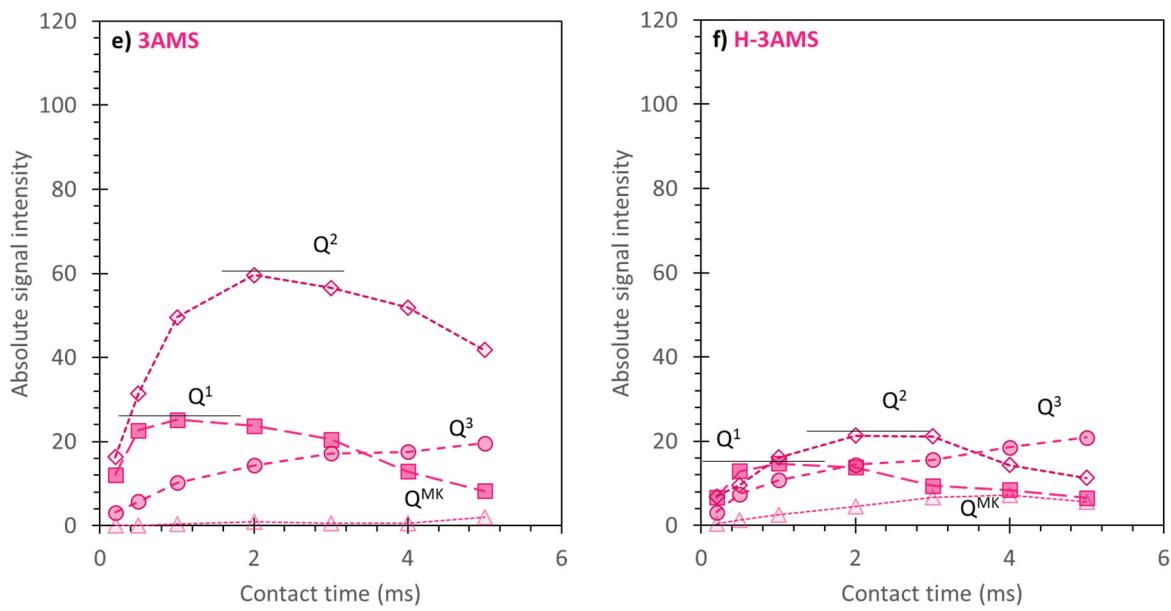
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Figure S 8: ^{1}H - ^{29}Si CP MAS-NMR data of a) MS, b) H-MS, c) AMS, d) H-AMS, e) 3AMS and f) H-3AMS pastes (182 days of hydration) recorded with contact times of 0.2 - 5 ms. The absolute signal intensities were normalized by number of scans and sample weights with respect to the data of sample MS (Table S2); error of the fitting = 5%, horizontal lines indicate a local maximum.

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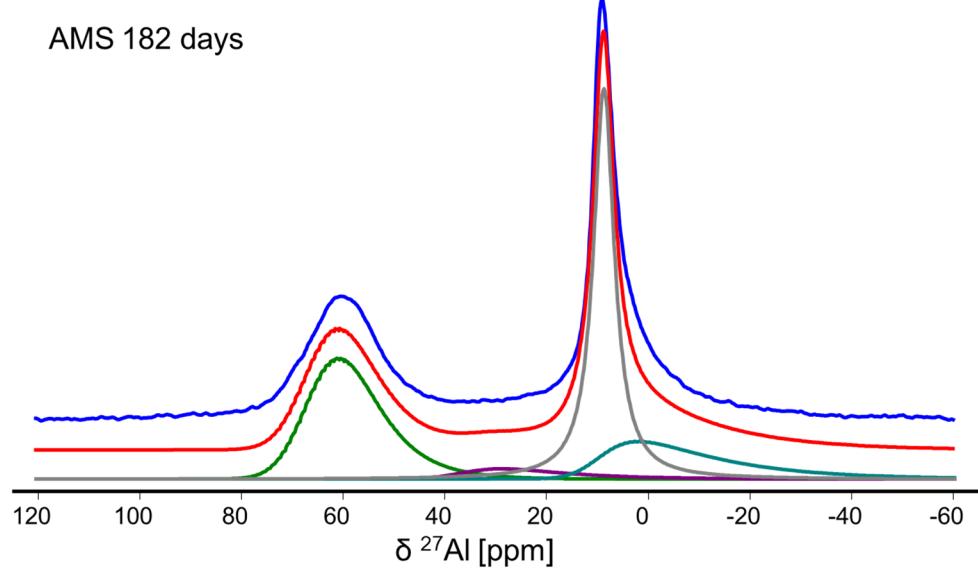
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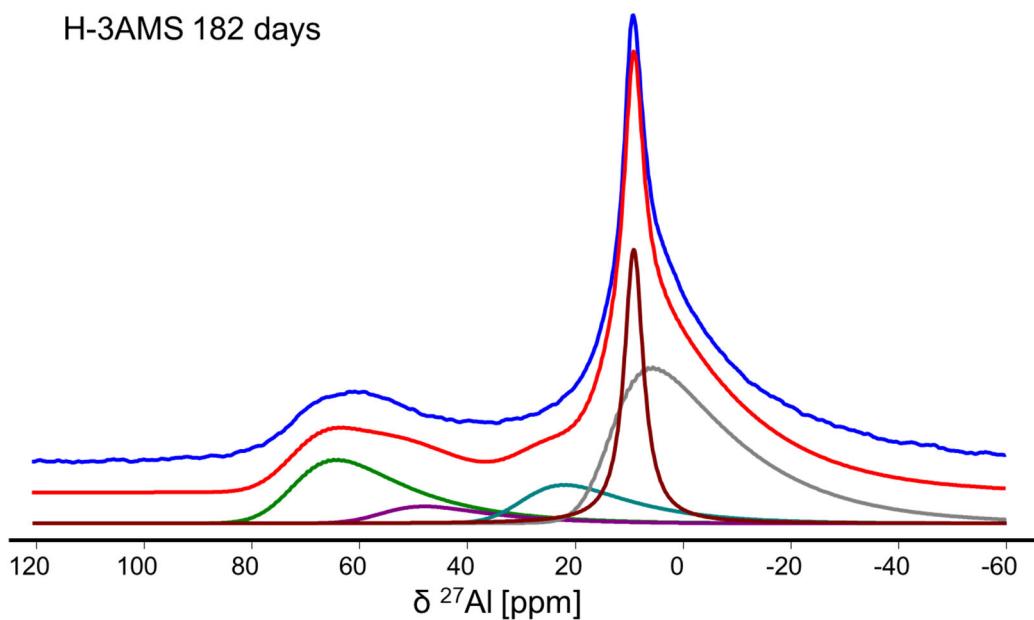
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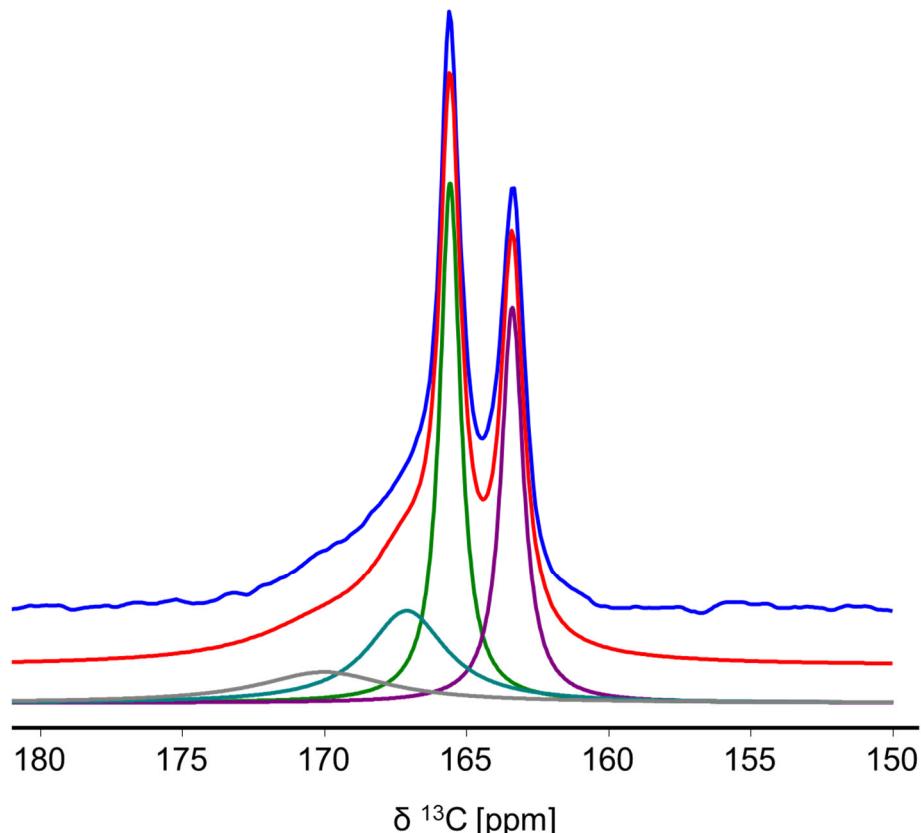
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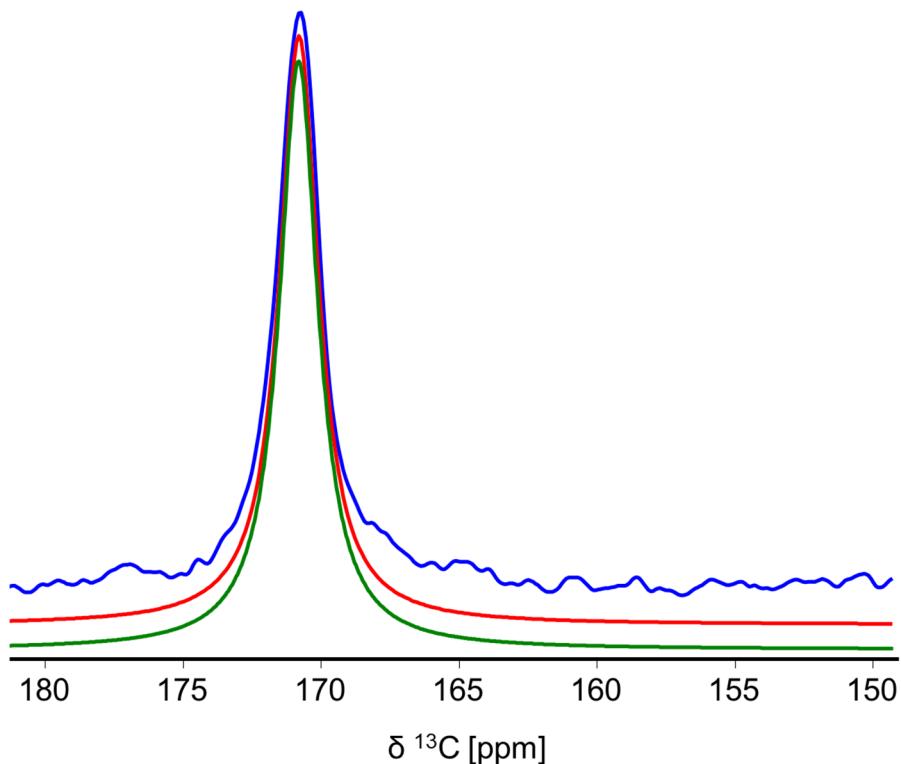
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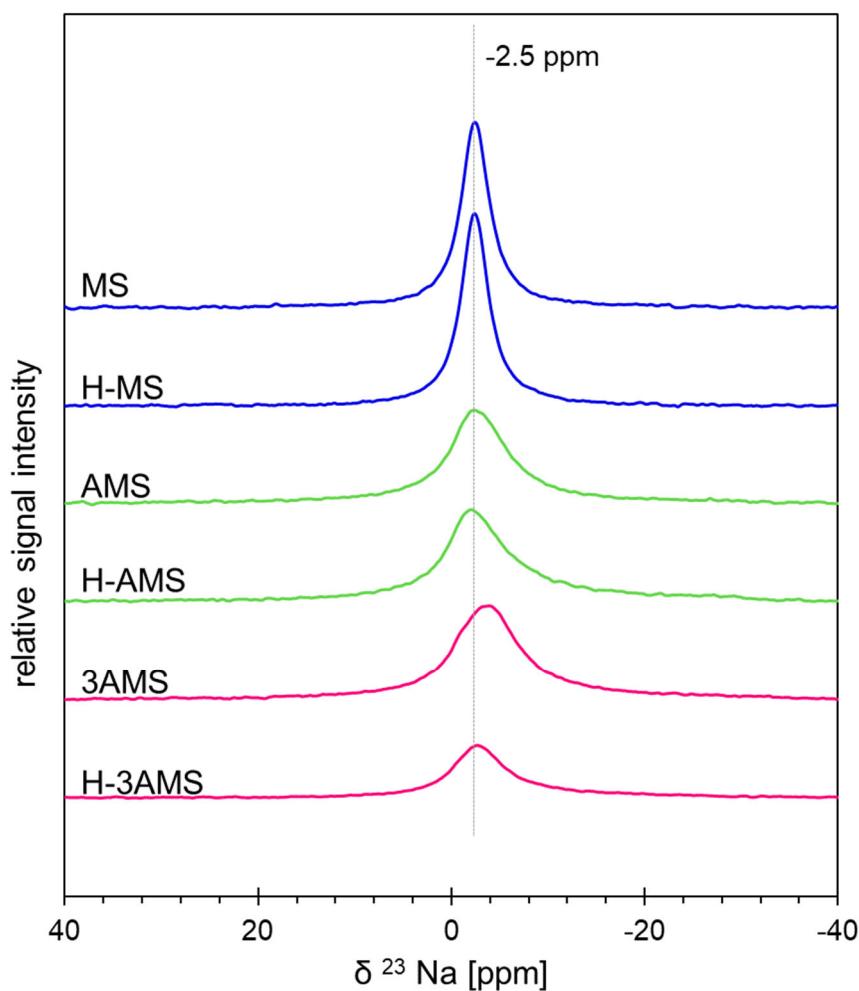
62
63 **Figure S 9:** Deconvolutions of the ^{27}Al MAS NMR spectra of AMS and H-3AMS paste
64 samples (at 182 days). Numerical data are given in Fehler! Verweisquelle konnte nicht
65 gefunden werden..
66



67
68 **Figure S 10:** Deconvolution of the ^{13}C CP MAS NMR spectrum of AMS paste (at 182
69 days). Numerical data are given in Fehler! Verweisquelle konnte nicht gefunden werden..
70



71
72 **Figure S 11: Deconvolution of the ^{13}C CP MAS NMR spectrum of the CO_3 -hydrotalcite**
73 **sample synthesised in [20].**
74

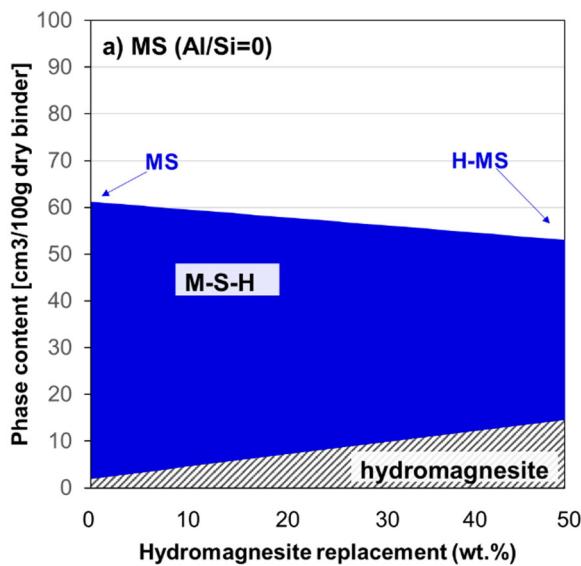


75
76 **Figure S 12:** Region of interest of ^{23}Na MAS NMR spectra of 182 days old samples. The
77 relative signal intensity has been corrected by the weight of the samples.
78

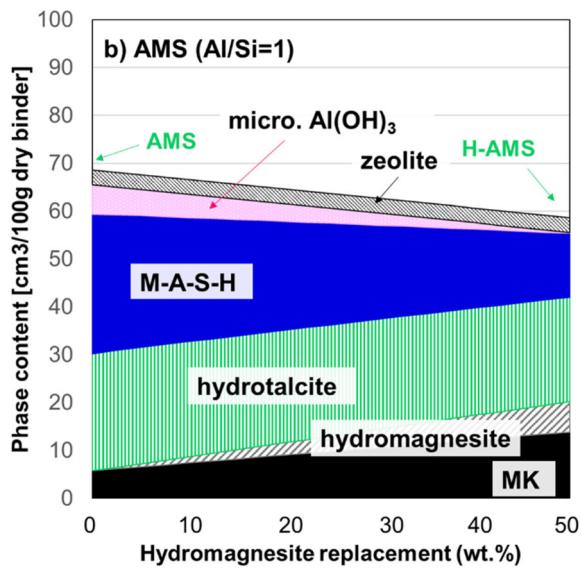
79 **Table S 3:** Relative amount of sodium in 182 days old samples determined by ^{23}Na NMR
80 data recorded
81

	Relative amount of sodium species ^a	position
M-S-H [41]	---	-5.7
MS	96	-2.5
H-MS	95	-2.5
AMS	85	-2.5
H-AMS	95	-2.1
3AMS	100	-4.0
H-3AMS	48	-2.8

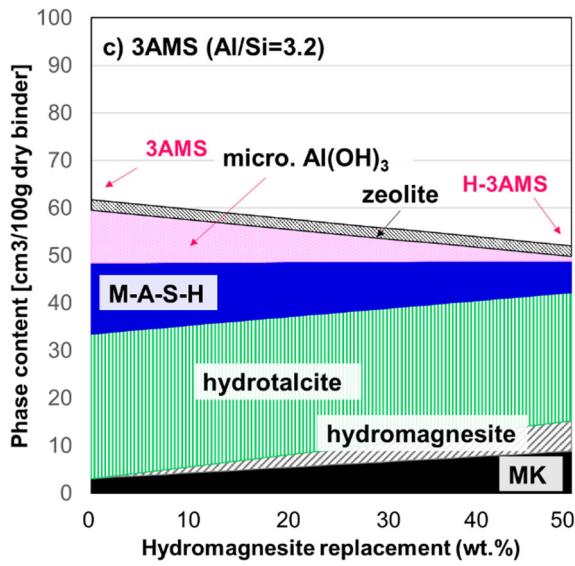
82 ^a relative amount of sodium species = absolute signal intensity per amount of material (^{23}Na MAS NMR
83 experiments recorded with 1024 scans)



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87 Figure S 13: Thermodynamically modelled changes in the a) $\text{MgO}-\text{SiO}_2$ system and in the
 88 b) & c) $\text{MgO}-\text{Al}_2\text{O}_3-\text{SiO}_2$ systems upon replacement of MgO by hydromagnesite after
 89 182 days. No constraint was applied to the $\text{MgO}-\text{SiO}_2$ system. The reactivity of the
 90 hydromagnesite was restrained to the reaction degree calculated from ^{13}C CP MAS NMR
 91 data (Fehler! Verweisquelle konnte nicht gefunden werden.) and the reactivity of MK to the
 92 reaction degree calculated from ^{29}Si MAS NMR data (Fehler! Verweisquelle konnte nicht
 93 gefunden werden.).

94

95 **Table S4:** Modelled phase assemblage volume compared to the modelled solution volume
 96 for the calculation of the porosity and modelled Mg/Si and Al/Si in the M-A-S-H phases.

phase assemblage	cm³/100g binder	solution	porosity %	in M-A-S-H	
				Mg/Si	Al/Si
MS	61.1	65.5	51.7	1.4	
H-MS	53.0	80.5	60.3	1.0	
AMS	68.6	51.9	43.1	0.9	0.2
H-AMS	58.7	72.9	55.4	0.9	0.2
3AMS	61.7	63.7	50.8	0.9	0.2
H-3AMS	50.0	78.0	60.9	0.9	0.2

97