

Hydration reactions and mechanical strength developments of iron-rich sulfobelite eco-cements

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Laboratory X-ray powder diffraction (LXRPD). Patterns were recorded in Bragg-Brentano reflection geometry ($\theta/2\theta$) on an X'Pert PRO MPD diffractometer (PANalytical B.V.) using strictly monochromatic $\text{CuK}\alpha_1$ radiation ($\lambda=1.54059\text{\AA}$) [Ge (111) primary monochromator]. The X-ray tube worked at 45 kV and 40 mA. The optics configuration was a fixed divergence slit ($1/2^\circ$), a fixed incident antiscatter slit (1°), a fixed diffracted anti-scatter slit ($1/2^\circ$) and X'Celerator RTMS (Real Time Multiple Strip) detector, working in scanning mode with maximum active length. Data were collected from 5° to 70° (2θ) for ~ 2 hours. The samples were rotated during data collection at 16 rpm in order to enhance particle statistics.

The external standard pattern was collected in identical diffractometer configuration/conditions (except for the spinning rate of the sample) and as close in time as possible to the pastes measurements to determine a diffractometer constant (G-factor). A polished polycrystalline quartz rock was used as secondary standard placed on the diffractometer in the very same orientation each measurement^{1,2}. The suitability of this quartz-rock was tested against NIST standard reference material SRM-676a ($\alpha\text{-Al}_2\text{O}_3$)³.

Table S1. Elemental composition of two clinkers and gypsum (determined by XRF)

	BCSAF_B0	BCSAF_B2	Gypsum
CaO	51.70	50.54	39.95
Al_2O_3	18.80	16.88	0.47
Fe_2O_3	6.72	6.22	0.15
SiO_2	16.70	16.38	0.93
SO_3	3.68	3.67	54.42
B_2O_3	0.23	2.70	
Na_2O	<0.08	1.55	0.26
K_2O	0.34	0.33	0.07
MgO	0.99	0.96	0.13
P_2O_5	0.055	0.059	0.56
TiO_2	0.65	0.61	
SrO	0.028	0.03	3.05
Cr_2O_3	0.028	0.023	
MnO	0.036	0.034	

ZrO

0.021

0.019

Table S2. Loss on ignition (LoI) at 800°C and weight loss obtained from DTA-TGA measurements for all pastes

Sample	Hydration time (d)	Theoretical total weight loss (%)	LoI (%)	Weight loss RT-600 °C (%)	Weight loss 600-1000 °C (%)
C5B0	3	36.2	33.1	23.9	1.8
	7		32.8	24.9	1.8
	28		32.7	26.6	1.6
	120		32.9	26.3	2.2
C10B0	3	36.8	33.9	23.9	1.7
	7		33.6	25.1	2.2
	28		33.6	27.8	1.9
	120		35.9	28.4	2.3
C15B0	3	37.5	35.7	23.7	1.3
	7		35.5	24.6	2.2
	28		36.0	29.6	1.7
	120		37.2	29.1	3.1
C5B2	3	36.2	29.6	19.0	1.4
	7		29.7	18.8	1.3
	28		32.5	25.6	4.1
	120		36.0	27.2	2.2
C10B2	3	36.8	31.5	21.2	1.7
	7		29.9	20.3	1.1
	28		34.1	25.8	3.2
	120		37.7	29.2	1.8
C15B2	3	37.5	30.7	23.3	1.3
	7		28.9	24.4	0.6
	28		34.8	25.2	3.4
	120		37.6	28.6	2.1

Table S3. Values of the cell parameter a for katoite and values of x (moles Fe_2O_3 in formula $\text{C}_3\text{A}_{1-x}\text{F}_x\text{SH}_4$).

Sample	Hydration time (d)	a (Å)	a_{average} (Å)	x
C5B0	7	12.435(2)	12.440	0.94
	28	12.444(1)		
	120	12.441(1)		
C10B0	7	12.437(3)	12.435	0.91
	28	12.435(1)		
	120	12.433(1)		
C15B0	7	12.426(4)	12.418	0.80
	28	12.414(1)		
	120	12.415(1)		
C5B2	28	12.393(1)	12.397	0.66
	120	12.400(1)		
C10B2	3	12.397(7)	12.405	0.72
	7	12.413(9)		
	28	12.387(1)	12.388	0.61
	120	12.389(1)		
C15B2	3	12.415(6)	12.415	0.78
	7	12.415(9)		
	28	12.416(6)	12.396	0.66
	120	12.396(1)		

References

- (1) Jansen, D.; Goetz-Neunhoeffer, F.; Stabler, C.H.; Neubauer, J. A remastered external standard method applied to the quantification of early OPC hydration. *Cem. Concr. Res.* **2011**, *41*, 602–608.
- (2) O'Connor, B. H.; Raven, M. D. Application of the Rietveld refinement procedure in assaying powdered mixtures. *Powder Diffr.* **1998**, *3*, 2–6.
- (3) Cline, J. P.; Von Dreele, R. B.; Winburn, R.; Stephens, P. W.; Filliben, J. J. Addressing the amorphous content issue in quantitative phase analysis: the certification of NIST standard reference material 676a. *Acta Crystallogr. A.* **2011**, *67*, 357–367.

Figure Captions

Figure S1. Differential thermal analysis and thermogravimetric curves for two stopped-hydration cements C10B0 and C10B2 at 7 days if hydration time.

Figure S2. Hydration and crystallization rates of (a) and (b) sulfate groups; (c) and (d) silicate groups for C5B0 and C5B2 pastes. Open square and open rhombus stand for silicate from katoite and stratlingite, respectively. Dashed lines represent the maximum values of dissolved groups.

Figure S3. Hydration and crystallization rates of (a) and (b) sulfate groups; (c) and (d) silicate groups for C15B0 and C15B2 pastes. Open square and open rhombus stand for silicate from katoite and stratlingite, respectively. Dashed lines represent the maximum values of dissolved groups.

Figure S4. Hydration and crystallization rates of (a) and (b) iron-bearing groups; (c) and (d) aluminate groups for C5B0 and C5B2 pastes. Dashed lines represent the maximum value of dissolved groups.

Figure S5. Hydration and crystallization rates of (a) and (b) iron-bearing groups; (c) and (d) aluminate groups for C15B0 and C15B2 pastes. Dashed lines represent the maximum value of dissolved groups.

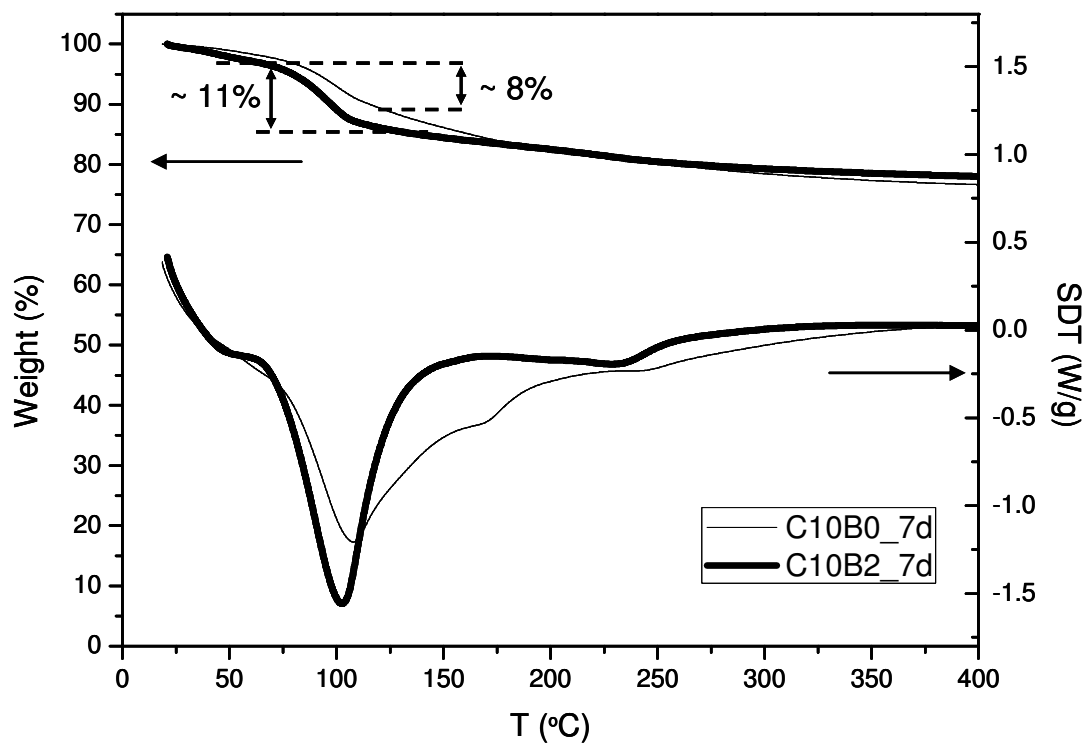


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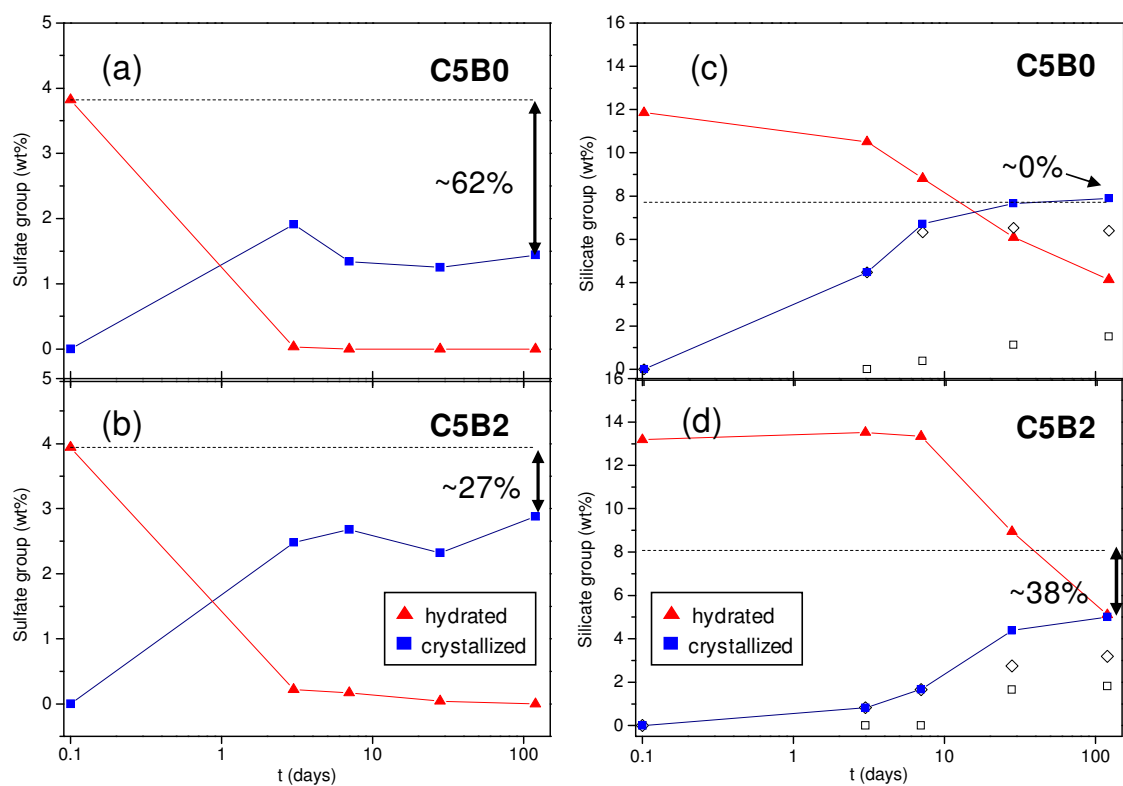


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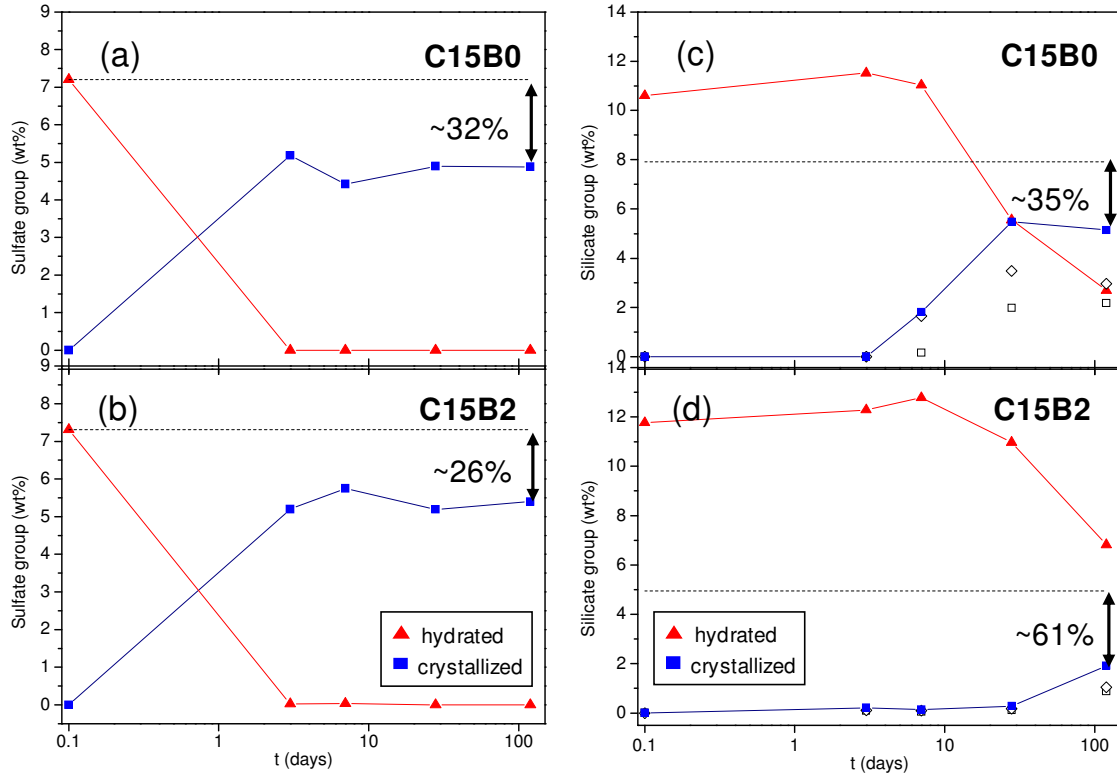


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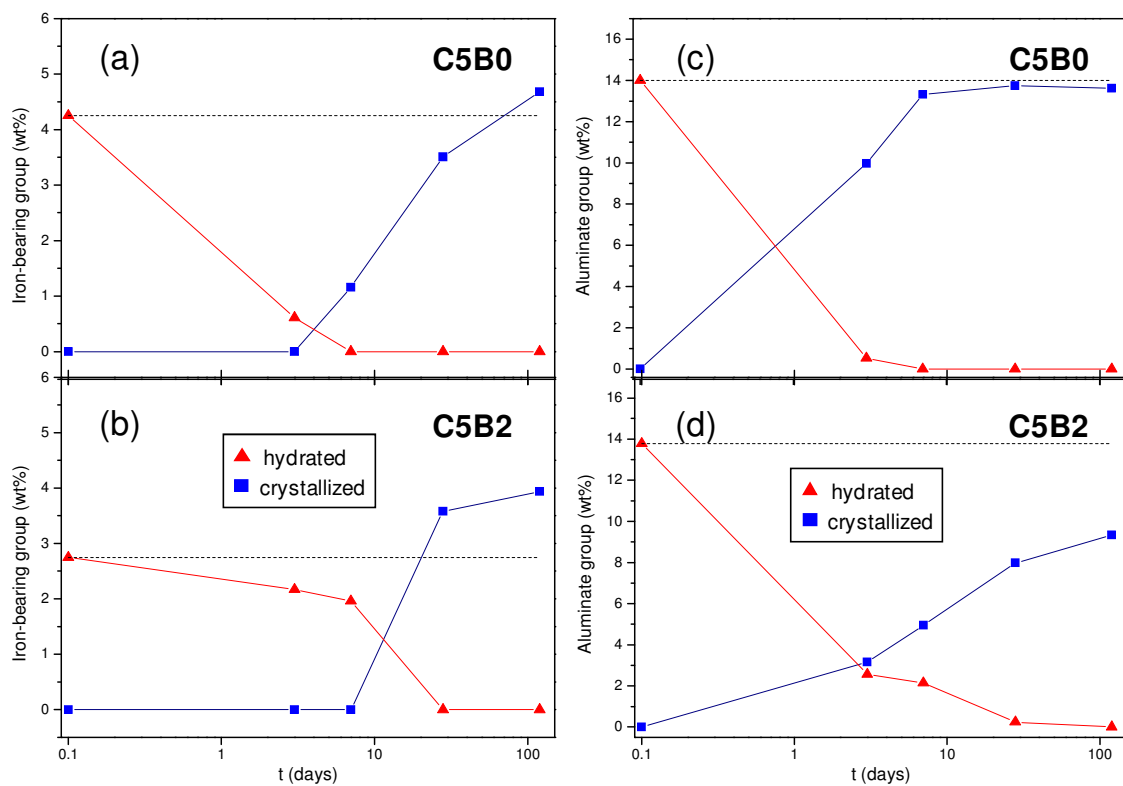


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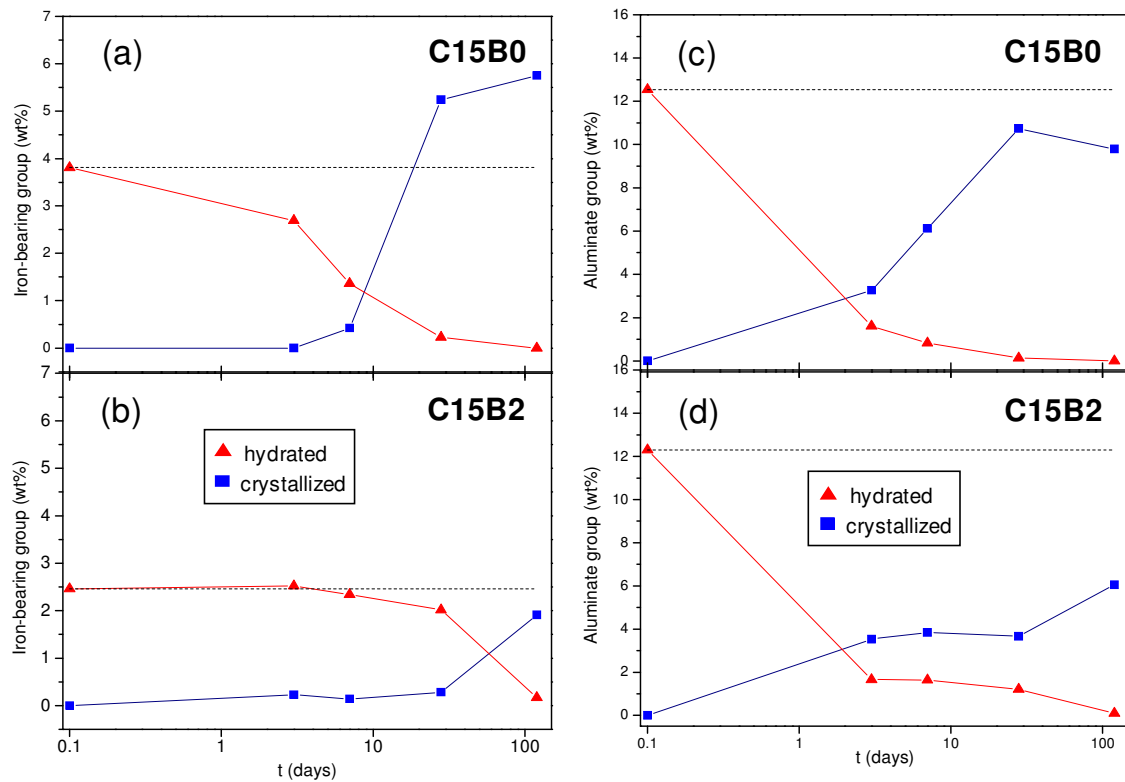


Figure S5. Hydration and crystallization rates of (a) and (b) iron-bearing groups; (c) and (d) aluminate groups for C15B0 and C15B2 pastes. Dashed lines represent the maximum value of dissolved groups.