

Supporting Information

Portland and Belite cement hydration acceleration by C-S-H seeds with variable w/c ratios

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Figure S2. Laboratory X-ray powder diffraction, MoK α 1 ($\lambda=0.7093$ Å), for pastes with w/c=0.40 at 28d: (a) BC-Buz-wc04-Ref and (b) BC-Buz-wc04-STE53.

Figure S3. Laboratory X-ray powder diffraction, MoK α 1 ($\lambda=0.7093$ Å), for pastes with w/c=0.40 at 28d: (a) BC-n.a.-wc04-Ref and (b) BC-n.a.-wc04-STE53.

Table S1. Elemental composition (wt%) from XRF for the three anhydrous cements.

	CaO	SiO ₂	SO ₃	Al ₂ O ₃	Fe ₂ O ₃	MgO	K ₂ O	Na ₂ O	Others	LoI
PC-42.5	62.9	19.7	3.4	5.0	3.4	1.5	1.1	0.3	0.3	2.7
BC-Buz	59.3	21.4	4.8	4.3	2.8	2.7	0.9	0.2	0.9	2.9
BC-n.a.	60.1	24.4	2.8	2.9	4.8	1.2	0.5	0.1	1.3	2.0

Table S2. Mineralogical composition (wt%) by Rietveld quantitative phase analysis from LXRPD, MoK α 1 ($\lambda=0.7093$ Å), for the three anhydrous cements.

	C ₃ S*	β -C ₂ S	C ₄ AF	C ₃ A [#]	C ₄ A ₃ \bar{S}	MgO	C \bar{C}	CM \bar{C}_2	C $\bar{S}H_2$	C $\bar{S}H_{0.5}$	C \bar{S}	ACn
PC-42.5	54.2	9.3	9.8	6.8	-	-	4.2	0.7	1.0	1.1	-	13.0
BC-Buz	24.8	44.1	10.1	2.0	1.9	1.4	1.3	-	-	-	1.6	12.8
BC-n.a.	28.6	48.0	15.2	-	-	-	-	-	2.0	-	-	6.2

*PC-42.5: C₃S-M3; BC-Buz: 2.9 wt% C₃S-M3 and 21.9 wt% C₃S-M1; BC-n.a.: 1.8 wt% C₃S-M3 and 26.8 wt% C₃S-M1#PC-42.5: o-C₃A; BC-Buz: c-C₃A**Table S3.** Textural analysis for the three anhydrous cements.

	Density (g/cm ³)	BET (m ² /g)	Blaine (m ² /Kg)	D _{v,10} (μm)	D _{v,50} (μm)	D _{v,90} (μm)
PC-42.5	3.13	1.2	370	3.6	18.1	50.3
BC-Buz	3.17	1.4	502	1.5	12.8	55.2
BC-n.a.	3.23	1.5	466	1.1	7.1	38.4

Table S4. RQPA results for PC-42.5-wc05 pastes. The cement also contained at t_0 0.7 wt% of CSH_2 and 0.7 wt% of $\text{CSH}_{0.5}$.

Phases	t_0	PC-42.5-wc05-Ref			PC-42.5-wc05-XS130			PC-42.5-wc05-STE53		
		1d	7d	28d	1d	7d	28d	1d	7d	28d
C_3S	36.1	16.7	8.1	3.8	16.4	8.4	4.2	14.7	8.7	2.7
C_2S	6.2	6.1	5.4	5.8	6.2	6.0	5.7	6.5	7.0	5.8
C_4AF	6.6	6.1	4.6	3.3	5.4	3.6	2.1	5.4	3.8	2.3
C_3A	4.5	4.2	2.0	0.5	3.5	1.8	0.6	3.3	1.6	0.5
$\text{C}\bar{\text{C}}$	2.8	4.9	5.2	3.1	6.6	4.3	2.4	5.9	2.4	2.2
$\text{CM}\bar{\text{C}}_2$	0.5	0.4	0.7	0.9	0.4	0.7	0.7	0.4	0.8	0.6
CH	-	6.5	10.2	12.3	5.7	8.9	11.1	7.1	10.0	10.8
AFt	-	5.0	8.4	8.0	6.1	9.0	8.5	5.8	7.5	7.0
AFm-Hc	-	-	-	-	-	1.1	0.9	-	0.7	0.7
AFm-Mc	-	-	-	1.2	-	-	1.1	-	-	1.8
ACn	8.7	24.4	33.8	42.9	25.5	36.8	45.4	25.4	37.8	49.9
$\text{FW}_{\text{TA}}^{\#}$	33.3	25.5	21.4	18.1	24.0	19.4	17.3	25.3	19.8	15.8
$\text{C-S-H}^{\$}$	-	19.9	29.6	33.5	20.1	28.5	33.2	21.4	26.9	34.6
<i>Other ACn</i> ^{\$}	8.7	5.4	9.2	15.9	5.9	11.7	18.1	7.2	14.5	20.4
$\text{FW}^{\$}$	33.3	24.6	16.3	11.6	23.5	15.9	11.4	22.1	16.2	10.7

[#] FW (free water) content determined from thermal data; ^{\$} In italics, calculated data of the chemical reaction

Table S5. RQPA results for PC-42.5-wc04 pastes. The cement also contained at t_0 0.7 wt% of CSH_2 and 0.8 wt% of $\text{CSH}_{0.5}$.

Phases	t_0	PC-42.5-wc04-Ref			PC-42.5-wc04-XS130			PC-42.5-wc04-STE53		
		1d	7d	28d	1d	7d	28d	1d	7d	28d
C_3S	38.7	18.2	8.6	4.5	17.6	9.8	6.7	16.8	9.5	4.9
C_2S	6.6	6.9	6.7	7.2	7.0	7.1	7.1	7.1	7.8	7.5
C_4AF	7.0	6.4	5.0	4.3	5.6	3.9	3.5	5.9	3.8	2.6
C_3A	4.8	4.5	2.0	0.8	3.6	1.4	0.8	3.7	1.2	0.3
$\text{C}\bar{\text{C}}$	3.0	5.4	5.7	4.5	5.7	3.6	3.1	5.6	3.6	2.3
$\text{CM}\bar{\text{C}}_2$	0.5	0.5	0.3	0.4	0.6	0.6	0.5	0.6	0.8	0.9
CH	-	6.7	9.9	11.3	6.2	7.9	9.5	7.2	9.2	10.3
AFt	-	5.1	8.2	8.2	6.2	7.0	6.3	5.5	6.3	5.8
AFm-Hc	-	-	-	0.3	-	0.7	0.3	-	1.0	0.4
AFm-Mc	-	-	-	1.2	-	1.3	1.5	-	-	2.0
ACn	9.3	26.4	38.9	45.1	28.5	42.0	49.4	28.7	42.8	52.9
$\text{FW}_{\text{TA}}^{\#}$	28.6	19.7	14.8	12.3	19.1	14.7	11.4	18.8	14.0	10.2
$\text{C-S-H}^{\$}$	-	20.9	30.7	34.9	21.5	29.5	32.6	22.3	29.8	34.5
<i>Other ACn</i> ^{\$}	9.3	5.8	12.4	16.0	8.8	17.7	21.1	7.9	18.0	23.6
$\text{FW}^{\$}$	28.6	19.4	10.6	6.5	17.3	9.5	7.1	17.2	9.0	5.1

[#] FW (free water) content determined from thermal data; ^{\$} In italics, calculated data of the chemical reaction

Table S6. RQPA results for BC-Buz-wc05 pastes. The cement also contained at t_0 1.0 wt% of $\text{CS}\bar{\text{S}}$.

Phases	t_0	BC-Buz-wc05-Ref			BC-Buz-wc05-XS130			BC-Buz-wc05-STE53		
		1d	7d	28d	1d	7d	28d	1d	7d	28d
C_3S	16.6	5.4	4.2	3.6	5.3	4.2	2.8	5.0	3.4	2.6
C_2S	29.4	33.5	28.0	18.7	32.0	23.8	16.0	31.8	27.7	18.1
C_4AF	6.7	5.5	4.0	1.7	3.3	1.7	1.1	3.1	1.6	0.9
C_3A	1.3	0.4	-	-	-	-	-	-	-	-
$\text{C}_4\text{A}_3\bar{\text{S}}$	1.2	0.7	-	-	-	-	-	0.3	-	-
$\text{C}\bar{\text{C}}$	0.9	3.1	0.9	1.2	2.6	1.3	0.9	2.7	0.9	0.5
MgO	0.9	1.1	0.9	0.8	1.0	0.9	0.9	1.0	0.9	0.7
CH	-	2.1	3.5	4.0	1.5	2.9	3.5	1.7	3.1	3.7
AFt	-	7.1	8.9	13.1	9.6	12.9	12.5	9.4	9.0	11.4
AFm-Hc	-	-	0.5	0.7	1.4	2.4	1.7	1.0	0.9	1.7
AFm-Mc	-	-	-	1.2	-	-	-	-	1.9	2.3
Katoite	-	-	0.5	0.8	-	0.5	0.5	-	0.5	0.5
ACn	8.6	15.1	26.5	34.1	18.5	28.5	42.1	19.1	29.2	40.4
FW_{TA}^*	33.3	26.0	22.1	20.1	24.9	21.0	18.0	25.0	21.1	17.1
$\text{C-S-H}^{\$}$	-	11.4	14.5	27.7	11.5	20.2	32.2	11.8	15.8	29.5
<i>Other ACn</i> ^{\$}	8.6	3.6	11.2	9.0	8.3	9.3	12.2	8.5	13.2	11.5
$\text{FW}^{\$}$	33.3	26.1	22.8	17.5	23.6	20.0	15.8	23.7	21.4	16.5

*FW (free water) content determined from thermal data; ^{\$}In italics, calculated data of the chemical reaction

Table S7. RQPA results for BC-Buz-wc04 pastes. The cement also contained at t_0 1.1 wt% of $\text{CS}\bar{\text{S}}$.

Phases	t_0	BC-Buz-wc04-Ref			BC-Buz-wc04-XS130			BC-Buz-wc04-STE53		
		1d	7d	28d	1d	7d	28d	1d	7d	28d
C_3S	17.7	6.1	3.7	2.7	6.3	3.4	3.0	5.9	3.4	3.2
C_2S	31.5	35.9	28.9	19.9	35.9	30.4	21.2	36.0	34.7	21.1
C_4AF	7.2	6.1	3.5	1.9	2.9	2.1	1.5	2.9	1.8	0.8
C_3A	1.4	0.6	-	-	0.3	-	-	-	-	-
$\text{C}_4\text{A}_3\bar{\text{S}}$	1.3	0.8	-	-	-	-	-	0.3	-	-
$\text{C}\bar{\text{C}}$	0.9	3.3	5.7	3.8	2.4	0.6	0.3	2.4	0.3	-
MgO	1.0	1.1	0.9	0.9	1.2	1.2	1.0	1.2	1.2	1.0
CH	-	2.0	3.3	4.2	1.0	1.5	2.7	1.5	1.6	3.2
AFt	-	6.8	10.0	11.0	9.2	12.3	8.9	8.2	11.2	8.8
AFm-Hc	-	-	-	0.2	1.3	2.8	1.2	1.1	2.5	1.3
AFm-Mc	-	-	1.1	2.1	-	-	-	-	-	-
Katoite	-	-	0.5	0.7	-	0.5	0.7	-	0.4	0.5
ACn	9.2	15.7	24.7	38.0	19.8	29.8	47.3	21.5	26.5	47.9
FW_{TA}^*	28.6	21.5	17.8	14.6	19.7	15.5	12.2	19.0	16.4	12.2
$\text{C-S-H}^{\$}$	-	11.8	17.8	31.0	11.6	16.1	28.9	12.0	14.6	28.8
<i>Other ACn</i> ^{\$}	9.2	3.9	8.5	10.4	9.2	13.0	18.9	10.1	11.8	19.8
$\text{FW}^{\$}$	28.6	21.4	16.2	11.2	18.7	16.2	11.7	18.4	16.5	11.5

*FW (free water) content determined from thermal data; ^{\$}In italics, calculated data of the chemical reaction

Table S8. RQPA results for BC-n.a.-wc05 pastes.

Phases	t ₀	BC-n.a.-wc05-Ref			BC-n.a.-wc05-XS130			BC-n.a.-wc05-STE53		
		1d	7d	28d	1d	7d	28d	1d	7d	28d
C ₃ S	19.0	6.9	2.4	1.7	8.1	2.3	1.2	6.9	3.0	1.3
C ₂ S	32.1	33.1	34.5	24.9	34.2	33.7	17.9	31.8	34.4	21.9
C ₄ AF	10.1	9.0	6.4	6.7	6.5	5.3	2.9	7.7	5.9	3.1
C ₃ H ₂	1.4	0.4	0.3	0.4	0.4	0.3	0.5	0.5	0.5	0.6
C ₂ C	-	2.2	-	-	1.8	-	-	1.2	-	-
CH	-	3.0	5.0	4.9	2.2	4.2	5.1	3.1	4.9	4.7
AFt	-	4.0	5.2	3.0	4.0	4.8	4.3	4.5	4.9	3.9
AFm-Hc	-	-	0.3	0.4	-	0.4	0.6	-	0.3	0.7
ACn	4.1	14.0	21.6	37.6	16.2	24.9	47.6	17.3	21.6	43.6
FW _{TA} [#]	33.3	27.5	24.3	20.4	26.6	24.1	19.9	27.0	24.4	20.2
C-S-H ^{\$}	-	12.3	16.9	27.4	11.1	17.0	37.3	12.3	16.3	31.8
Other ACn ^{\$}	4.1	1.3	4.1	9.0	4.4	7.7	13.5	4.7	4.7	13.4
FW ^{\$}	33.3	27.9	24.9	21.6	27.3	24.4	16.7	27.3	24.9	18.6

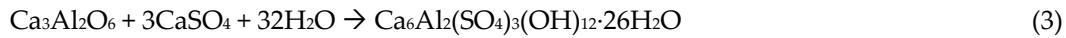
[#] FW (free water) content determined from thermal data; ^{\$} In italics, calculated data of the chemical reaction**Table S9.** RQPA results for BC-n.a.-wc04 pastes.

Phases	t ₀	BC-n.a.-wc04-Ref			BC-n.a.-wc04-XS130			BC-n.a.-wc04-STE53		
		1d	7d	28d	1d	7d	28d	1d	7d	28d
C ₃ S	20.4	8.5	3.6	1.5	10.4	2.9	3.1	7.5	2.8	2.5
C ₂ S	34.2	35.9	35.4	27.9	35.5	35.4	20.6	35.4	37.2	24.9
C ₄ AF	10.9	9.2	7.3	6.7	8.2	5.6	2.9	8.6	6.0	2.6
C ₃ H ₂	1.5	0.5	0.7	0.5	0.6	0.6	0.6	0.4	0.5	0.7
C ₂ C	-	1.8	-	-	0.6	-	-	0.4	-	-
CH	-	3.9	4.4	4.5	2.1	5.3	5.0	3.2	5.5	6.5
AFt	-	3.8	5.0	3.8	5.1	5.0	3.6	5.7	4.9	3.5
AFm-Hc	-	-	0.4	0.6	-	-	0.4	-	-	0.6
ACn	4.4	14.0	23.8	39.3	15.2	26.2	50.5	17.3	23.9	44.7
FW _{TA} [#]	28.6	22.5	19.4	15.2	22.4	19.0	13.3	21.6	19.2	14.1
C-S-H ^{\$}	-	12.1	17.1	27.8	10.2	17.9	36.0	13.2	18.0	30.8
Other ACn ^{\$}	4.4	1.4	5.9	10.4	4.1	8.2	15.7	2.4	5.9	14.4
FW ^{\$}	28.6	23.0	20.2	16.3	23.3	19.1	12.1	23.3	19.3	13.6

[#] FW (free water) content determined from thermal data; ^{\$} In italics, calculated data of the chemical reaction

◆ **Theoretical calculations of the free water, the C-S-H gel, and the other ACn contents based on chemical reactions and the RQPA results.**

For the theoretical calculation of free water, equations (1) to (5) were used; equations (1) and (2) were been used for the calculation of C-S-H gel



Detailed example for free water (FW) calculation. The FW at a given hydration time was calculated for each sample and age, using the initial free water and considering the bounded water according to the stoichiometry of reactions (1) to (5).

The calculations for BC-Buz-wc04-Ref at 28d are detailed next as an example; see Table S7.

I. C₃S: There is 2.7 wt% of unreacted C₃S. This means that 15.0 wt% (C₃S_{initial} – C₃S_{final}) has reacted. According to reaction (1), 6.1 wt% of free water has been consumed.

II. C₂S: There is 19.9 wt% of unreacted C₂S. This means that 11.6 wt% (C₂S_{initial} – C₂S_{final}) has reacted. According to reaction (2), 5.1 wt% of free water has been consumed.

III. C₃A: All C₃A has been dissolved at this age. This means that 1.4 wt% (C₃A_{initial} – C₃A_{final}) has reacted. According to reaction (3), 2.4 wt% of free water has been consumed.

IV. C₄A₃̄S: All C₄A₃̄S has been dissolved at this age. This means that 1.3 wt% (C₄A₃̄S_{initial} – C₄A₃̄S_{final}) has reacted. According to reaction (4), 1.5 wt% of free water has been consumed.

V. C₄AF: There is 1.9 wt% of unreacted C₄AF. This means that 5.3 wt% (C₄AF_{initial} – C₄AF_{final}) has reacted. According to reaction (5), 2.3 wt% of free water has been consumed.

The calculated free water content for BC-Buz-wc04-Ref at 28d is **11.2 wt%** (28.6 wt% - 6.1 wt% - 5.1 wt% - 2.4 wt% - 1.5 wt% - 2.3 wt%)

Detailed example for C-S-H gel calculation. C-S-H gel was calculated according to reactions (1) and (2). The calculations for BC-Buz-wc04-Ref at 28d are detailed as an example; see Table S7.

I. C₃S: There is 2.7 wt% of unreacted C₃S. This means that 15.0 wt% (C₃S_{initial} – C₃S_{final}) has reacted. According to reaction (1), 15.3 wt% of C-S-H gel has been formed.

II. C₂S: There is 19.9 wt% of unreacted C₂S. This means that 11.6 wt% (C₂S_{initial} – C₂S_{final}) has reacted. According to reaction (2), 15.7 wt% of C-S-H gel has been formed.

A total of **31.0 wt%** of C-S-H gel has been formed.

Detailed example for other-ACn calculation. The calculated FW and C-S-H gel should be close to the total ACn content. Differences between total ACn and calculated C-S-H and FW are considered to be other-ACn (such as AFm-type, iron-siliceous hydrogarnet, and the remaining fraction of amorphous phase(s) within the unreacted clinker components).

BC-Buz-wc04-Ref at 28d is used as an example; see Table S7.

The difference of total ACn obtained by internal standard (52.6 wt%) and free water (11.2 wt%) and the calculated C-S-H gel (31.0 wt%) give the other-ACn contents (52.6 wt% - 11.2 wt% - 31.0 wt% = **10.4 wt%** other-ACn)

◆ Raw data availability.

Raw data from the following techniques have been deposited: Calorimetry (excel file), LXRPD (txt file) MIP (excel file), TA (TA Universal Analysis software file). Data can be accessed at: <https://doi.org/10.5281/zenodo.6335735>

Calorimetry

PC-42.5-wc05-Ref (1 file)
PC-42.5-wc05-XS130 (1 file)
PC-42.5-wc05-STE53 (1 file)
PC-42.5-wc04-Ref (1 file)
PC-42.5-wc04-XS130 (1 file)
PC-42.5-wc04-STE53 (1 file)
BC-Buz-wc05-Ref (1 file)
BC-Buz-wc05-XS130 (1 file)
BC-Buz-wc05-STE53 (1 file)
BC-Buz-wc04-Ref (1 file)
BC-Buz-wc04-XS130 (1 file)
BC-Buz-wc04-STE53 (1 file)
BC-n.a.-wc05-Ref (1 file)
BC-n.a.-wc05-XS130 (1 file)
BC-n.a.-wc05-STE53 (1 file)
BC-n.a.-wc04-Ref (1 file)
BC-n.a.-wc04-XS130 (1 file)
BC-n.a.-wc04-STE53 (1 file)

LXRPD

PC-42.5-Anh (1 file)
PC-42.5-wc05-Ref-Serie (3 files)
PC-42.5-wc05-XS130-Serie (3 files)
PC-42.5-wc05-STE53-Serie (3 files)
PC-42.5-wc04-Ref-Serie (3 files)
PC-42.5-wc04-XS130-Serie (3 files)
PC-42.5-wc04-STE53-Serie (3 files)
BC-Buz-Anh (1 file)
BC-Buz-wc05-Ref-Serie (3 files)
BC-Buz-wc05-XS130-Serie (3 files)
BC-Buz-wc05-STE53-Serie (3 files)
BC-Buz-wc04-Ref-Serie (3 files)
BC-Buz-wc04-XS130-Serie (3 files)
BC-Buz-wc04-STE53-Serie (3 files)
BC-n.a.-Anh (1 file)
BC-n.a.-wc05-Ref-Serie (3 files)
BC-n.a.-wc05-XS130-Serie (3 files)
BC-n.a.-wc05-STE53-Serie (3 files)
BC-n.a.-wc04-Ref-Serie (3 files)
BC-n.a.-wc04-XS130-Serie (3 files)
BC-n.a.-wc04-STE53-Serie (3 files)

MIP

PC-42.5-wc05-Ref-Serie (3 files)
PC-42.5-wc05-XS130-Serie (3 files)
PC-42.5-wc05-STE53-Serie (3 files)
PC-42.5-wc04-Ref-Serie (3 files)
PC-42.5-wc04-XS130-Serie (3 files)
PC-42.5-wc04-STE53-Serie (3 files)
BC-Buz-wc05-Ref-Serie (3 files)
BC-Buz-wc05-XS130-Serie (3 files)
BC-Buz-wc05-STE53-Serie (3 files)
BC-Buz-wc04-Ref-Serie (3 files)
BC-Buz-wc04-XS130-Serie (3 files)
BC-Buz-wc04-STE53-Serie (3 files)
BC-n.a.-wc05-Ref-Serie (3 files)
BC-n.a.-wc05-XS130-Serie (3 files)
BC-n.a.-wc05-STE53-Serie (3 files)
BC-n.a.-wc04-Ref-Serie (3 files)
BC-n.a.-wc04-XS130-Serie (3 files)
BC-n.a.-wc04-STE53-Serie (3 files)

TA

PC-42.5-wc05-Ref-Serie (3 files)
PC-42.5-wc05-XS130-Serie (3 files)
PC-42.5-wc05-STE53-Serie (3 files)
PC-42.5-wc04-Ref-Serie (3 files)
PC-42.5-wc04-XS130-Serie (3 files)
PC-42.5-wc04-STE53-Serie (3 files)
BC-Buz-wc05-Ref-Serie (3 files)
BC-Buz-wc05-XS130-Serie (3 files)
BC-Buz-wc05-STE53-Serie (3 files)
BC-Buz-wc04-Ref-Serie (3 files)
BC-Buz-wc04-XS130-Serie (3 files)
BC-Buz-wc04-STE53-Serie (3 files)
BC-n.a.-wc05-Ref-Serie (3 files)
BC-n.a.-wc05-XS130-Serie (3 files)
BC-n.a.-wc05-STE53-Serie (3 files)
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BC-n.a.-wc04-XS130-Serie (3 files)
BC-n.a.-wc04-STE53-Serie (3 files)

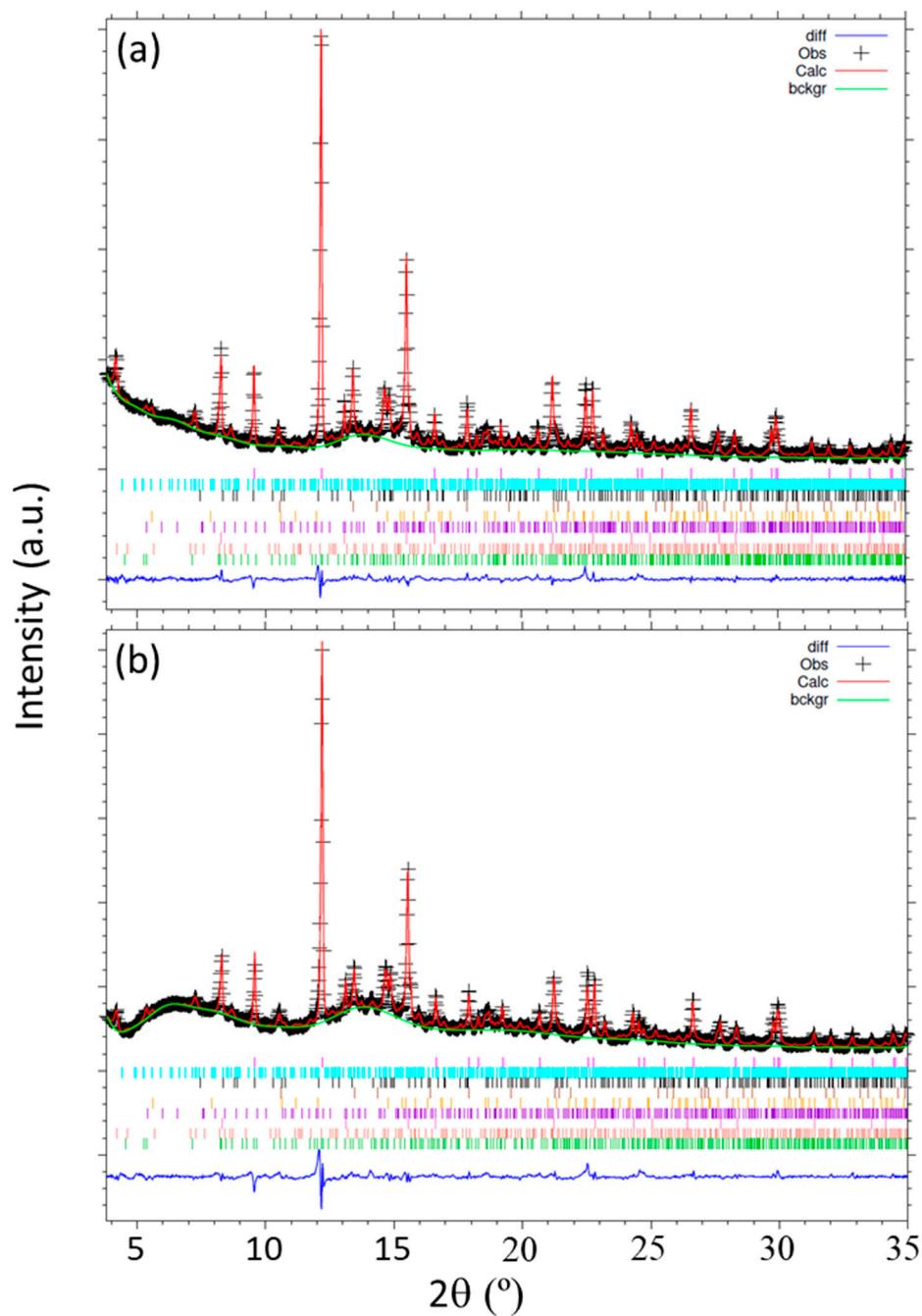


Figure S1. Laboratory X-ray powder diffraction, MoK α_1 ($\lambda=0.7093$ Å), for pastes with w/c=0.40 at 28d: (a) PC-42.5-wc04-Ref and (b) PC-42.5-wc04-STE53.

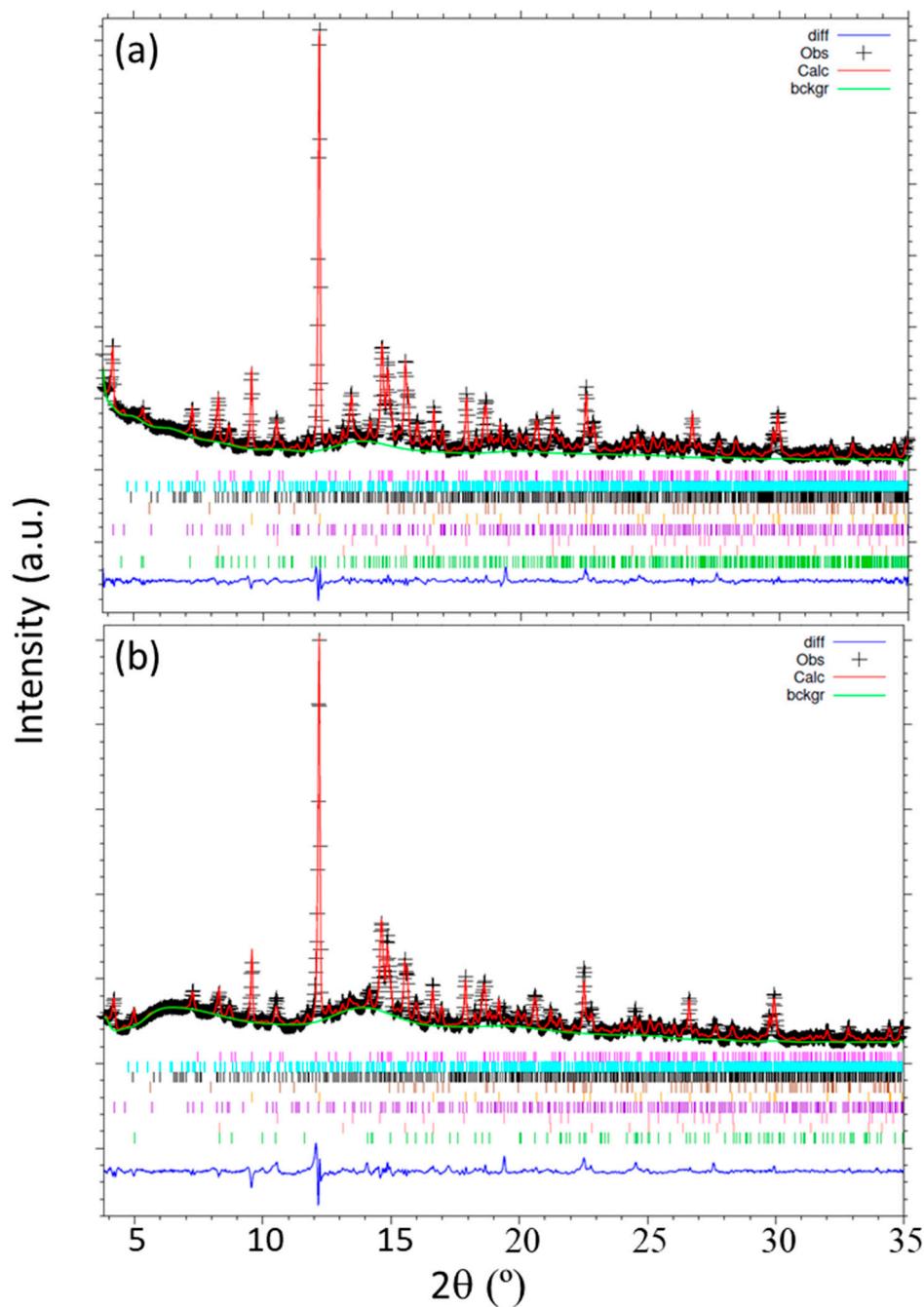


Figure S2. Laboratory X-ray powder diffraction, MoK α_1 ($\lambda=0.7093$ Å), for pastes with w/c=0.40 at 28d: (a) BC-Buz-wc04-Ref and (b) BC-Buz-wc04-STE53.

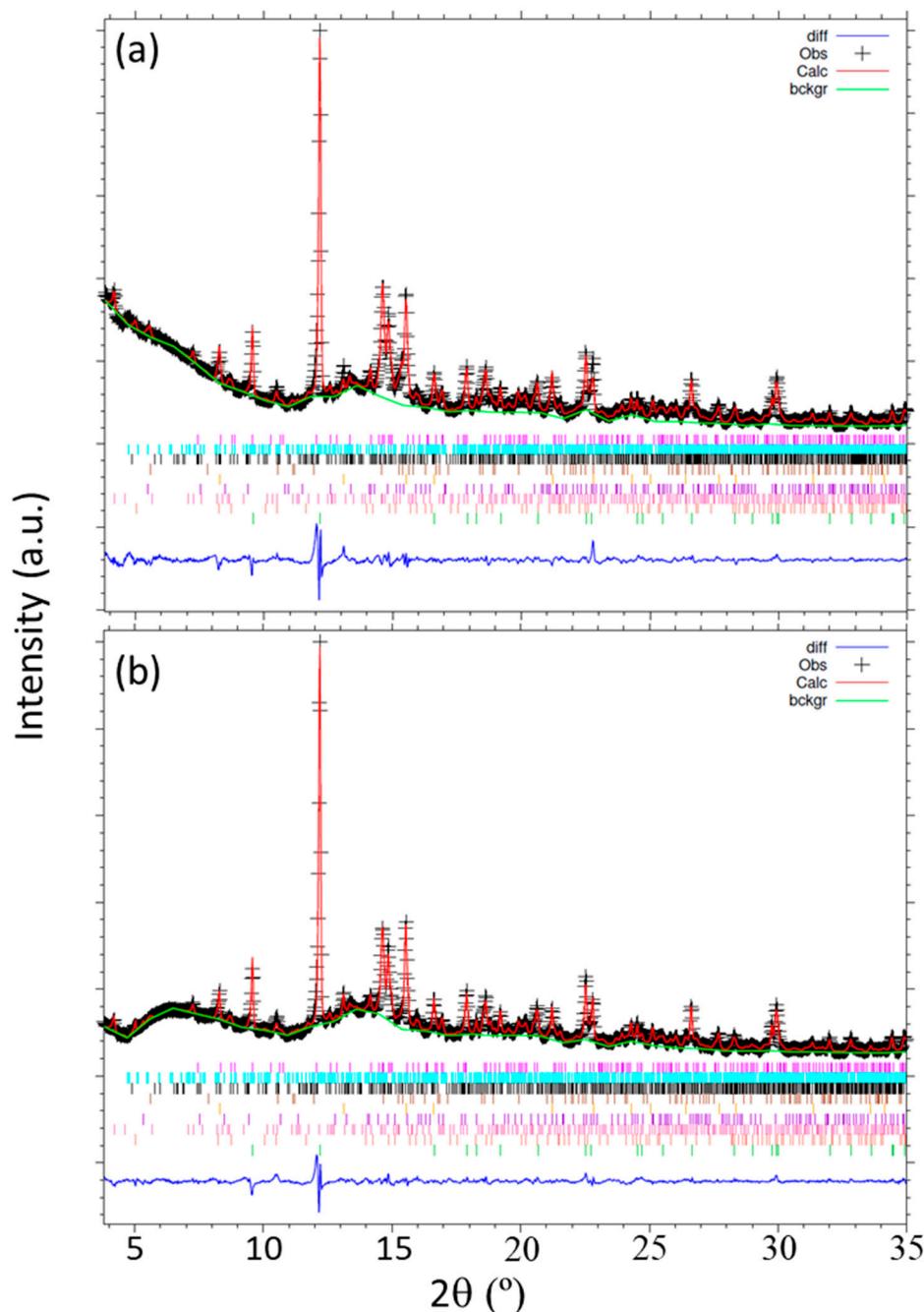


Figure S3. Laboratory X-ray powder diffraction, MoK α_1 ($\lambda=0.7093$ Å), for pastes with w/c=0.40 at 28d: (a) BC-n.a.-wc04-Ref and (b) BC-n.a.-wc04-STE53.