Supporting material

Synchrotron Radiation Pair Distribution Function Analysis of gels in cements

Ana Cuesta [1], Jesus D. Zea-Garcia [2], Diana Londono-Zuluaga [2], Angeles G. De la Torre [2], Isabel Santacruz [2], Oriol Vallcorba [1]and Miguel A.G. Aranda [1]*

[1] ALBA Synchrotron radiation facility, Carrer de la Llum 2-26. 08290 Cerdanyola del Vallès, Barcelona, Spain;
[2] Departamento de Química Inorgánica, Cristalografía y Mineralogía. Universidad de Málaga, 29071 Málaga, Spain
* Correspondence: migarcia@cells.es; Tel.: (+34) 935924305

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Description of every total scattering raw data set deposited open access.

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Figure S3. Experimental (blue circles) and fitted (red solid line) PDF patterns for the as-received Ca_3SiO_5. Difference curve as grey line.

Figure S4. Thermogravimetric data for $CaAl_2O_4$ pastes hydrated for 30 days: (a) w/s=0.55 sample hydrated at 35ºC; (b) w/s=1.20 sample hydrated at 35ºC; (c) w/s=0.55 sample hydrated at 45ºC.

Figure S5. Thermogravimetric data for (a) ye'elimite–gypsum paste hydrated with w/s=1.2 for 21 days at room temperature and (b) ye'elimite–bassanite paste hydrated with w/s=1.2 for 14 days at room temperature

Figure S6. Experimental (blue circles) and fitted (red solid line) PDF patterns for the ye'elimite–bassanite paste hydrated with w/s=1.20 for 21 days at room temperature (a) high r-range: 30–50 Å, (b) low r-range: 1.6–35 Å, (c) enlarged view of: 1.6–10 Å. Difference curve as grey lines.

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All of the total scattering raw data underlying this article, including the nickel and Ca_3SiO_5 data sets employed as standards and the empty capillary utilized for data processing, can be accessed on Zenodo at https://doi.org/10.5281/zenodo.890585, and used under the Creative Commons Attribution license.

Files:
Ni0p7_ALL.dat: Nickel sample employed as standard.
C3S_2016_anh_ALL.dat: Anhydrous alite employed as standard
empty0p7_ALL.dat: empty capillary
C3S-046_ALL.dat: alite paste hydrated with a w/s mass ratio of 0.46.
C3S-055_ALL.dat: alite paste hydrated with a w/s mass ratio of 0.55.
C3S-065_ALL.dat: alite paste hydrated with a w/s mass ratio of 0.65.
C3S-080_ALL.dat: alite paste hydrated with a w/s mass ratio of 0.80.
CA_35C_055_ALL.dat: calcium aluminate paste hydrated with a w/s mass ratio of 0.55 at 35ºC
CA_35C_120_ALL.dat: calcium aluminate paste hydrated with a w/s mass ratio of 1.20 at 35ºC
CA_45C_055_ALL.dat: calcium aluminate paste hydrated with a w/s mass ratio of 0.55 at 45ºC
C4A3s_G_120_ALL.dat: ye'elimite with gypsum paste hydrated with a w/s mass ratio of 1.20.
C4A3s_B_RT_120_ALL.dat: ye'elimite with bassanite paste hydrated with a w/s mass ratio of 1.20.

Table S1. Rietveld quantitative phase analysis results for the alite pastes after 34 hydration days.

sample	Alite (wt%)	$CaCO_3$ (wt%)	Portlandite (wt%)	ACn[#] (wt%)
alite_046	13.2	1.4	18.2	67.2
alite_055	11.2	1.2	21.8	65.8
alite_065	10.8	1.2	21.6	66.4
alite_080	10.1	1.5	21.8	66.7

[#] ACn accounts for the amorphous phase plus any crystalline not-quantified content.

Table S2. Summary of the weight losses from the TGA study for the alite pastes.

Weight loss (wt%)	RT–250ºC	250ºC–400ºC	400ºC–600ºC	600ºC–1000ºC	Full range
alite_046	13.9	2.0	6.2	2.2	24.3
alite_055	14.1	1.7	6.5	3.1	25.4
alite_065	15.1	1.6	6.6	3.3	26.6
alite_080	15.0	1.6	7.1	3.2	26.9

Table S3. Anisotropic atomic displacement parameters (ADPs) for portlandite in w/s=0.80 paste obtained in the PDF analysis. Note that the parameters for the hydrogen were not refined.

Atom	Ca1	O1	H
x	0	0.3333	0.3333
y	0	0.6667	0.6667
z	0	0.2161	0.4256
u11	0.0037	0.0084	0.0264
u22	0.0037	0.0084	0.0264
u33	0.0118	0.0212	0.0264
u12	0.0019	0.0042	0.0132
u13	0	0	0
u23	0	0	0

Table S4. Quantitative phase analysis results obtained by pair distribution function (PDF) using two crystal structures for the nanocrystalline CSH gel in the alite w/s=0.46, 0.55, and 0.65 pastes. Rw values are also included.

sample	Rw (%)	Ca_3SiO_5 (wt%)	$CaCO_3$ (wt%)	Cryst-$Ca(OH)_2$ (wt%)	C-S-H (wt%)
Alite_046					
clinotobermorite T3_14sc	28.5	32.4	0.3	23.1	44.1
Jennite, ICSD #151413	30.0	36.8	0.4	26.3	36.5
Alite_055					
clinotobermorite, T3_14sc	29.5	23.4	0.9	33.6	42.2
Jennite, ICSD #151413	32.5	24.9	0.9	35.9	38.3
Alite_065					
clinotobermorite, T3_14sc	23.2	14.1	0.8	30.2	54.9
Jennite, ICSD #151413	27.4	13.2	0.8	28.5	57.5

Table S5. Refined unit cell parameters for portlandite and clinotobermorite T3_14sc in the alite w/s=0.46, 0.55, and 0.65 pastes by PDF analysis.

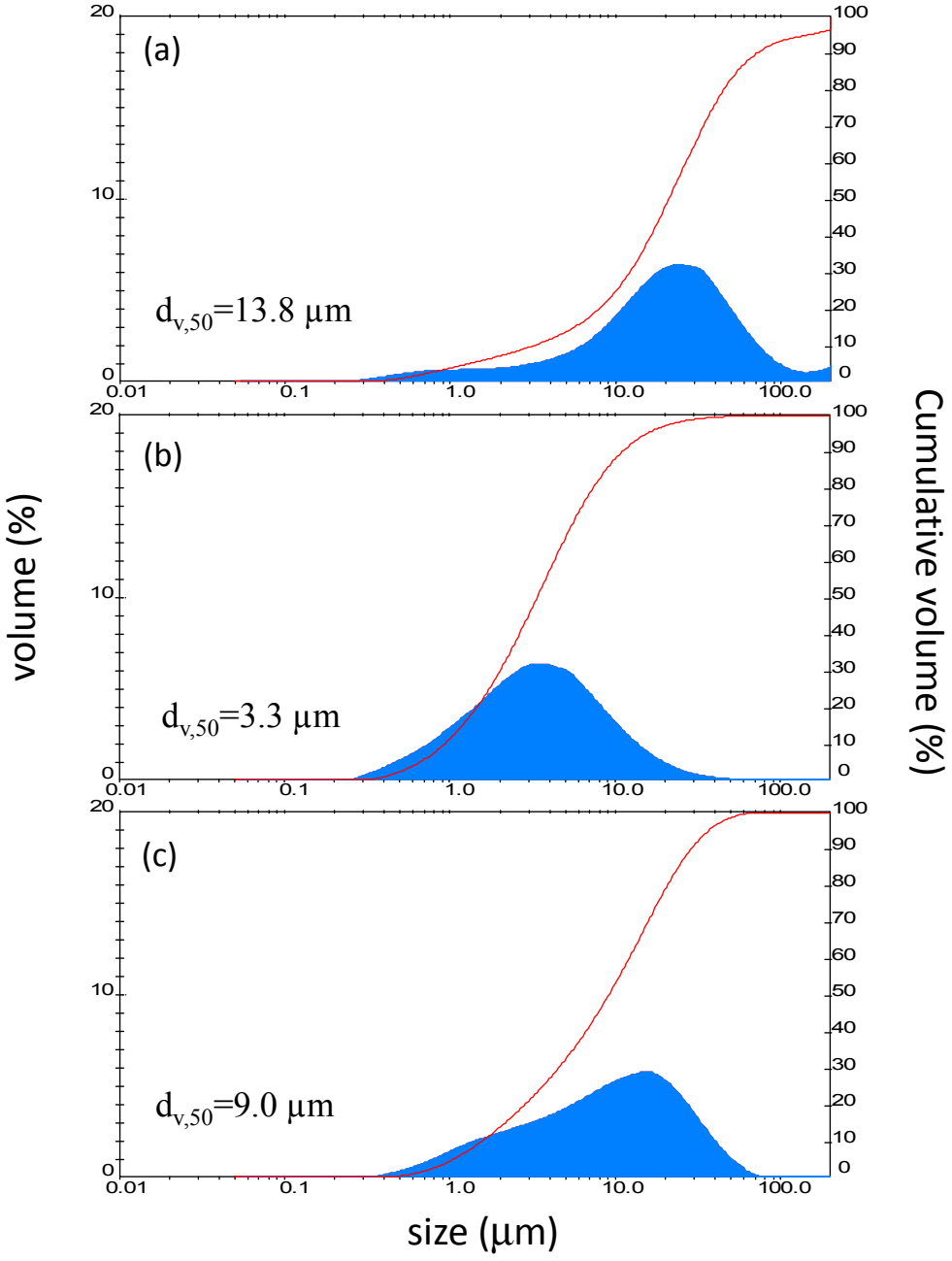
Sample	Portlandite		Clinotobermorite_T3_14sc			
	a (Å)	c (Å)	a (Å)	b (Å)	c (Å)	β (º)
alite_046	3.593	4.914	11.304	7.307	42.375	94.5
alite_055	3.594	4.914	11.180	7.356	42.003	93.0
alite_065	3.594	4.916	11.238	7.302	42.621	94.2

Table S6. Laboratory x-ray powder diffraction (LXRPD) Rietveld quantitative phase analysis results for the calcium aluminate pastes.

sample	Hydrogarnet (wt%)	Hemicarbonate (wt%)	ACn (wt%)
CA_055_35ºC	42.0	1.6	56.4
CA_120_35ºC	42.7	1.7	55.6
CA_055_45ºC	45.0	2.2	52.8

Table S7. Refined unit cell parameters and ADPs for hydrogarnet and gibbsite in the calcium aluminate pastes obtained by the PDF analysis.

Sample	Hydrogarnet				Gibbsite					
	a (Å)	ADPs (Å^2)			a (Å)	b (Å)	c (Å)	β (º)	ADPs (Å^2)	
		Ca	Al	O					Al	O
CA_055_35ºC	12.579	0.0063	0.0072	0.0182	8.693	5.046	9.713	94.6	0.0050	0.0153
CA_120_35ºC	12.578	0.0054	0.0063	0.0152	8.668	5.075	9.699	94.6	0.0012	0.0062
CA_055_45ºC	12.573	0.0063	0.0071	0.0167	8.671	5.064	9.712	94.5	0.0021	0.0093



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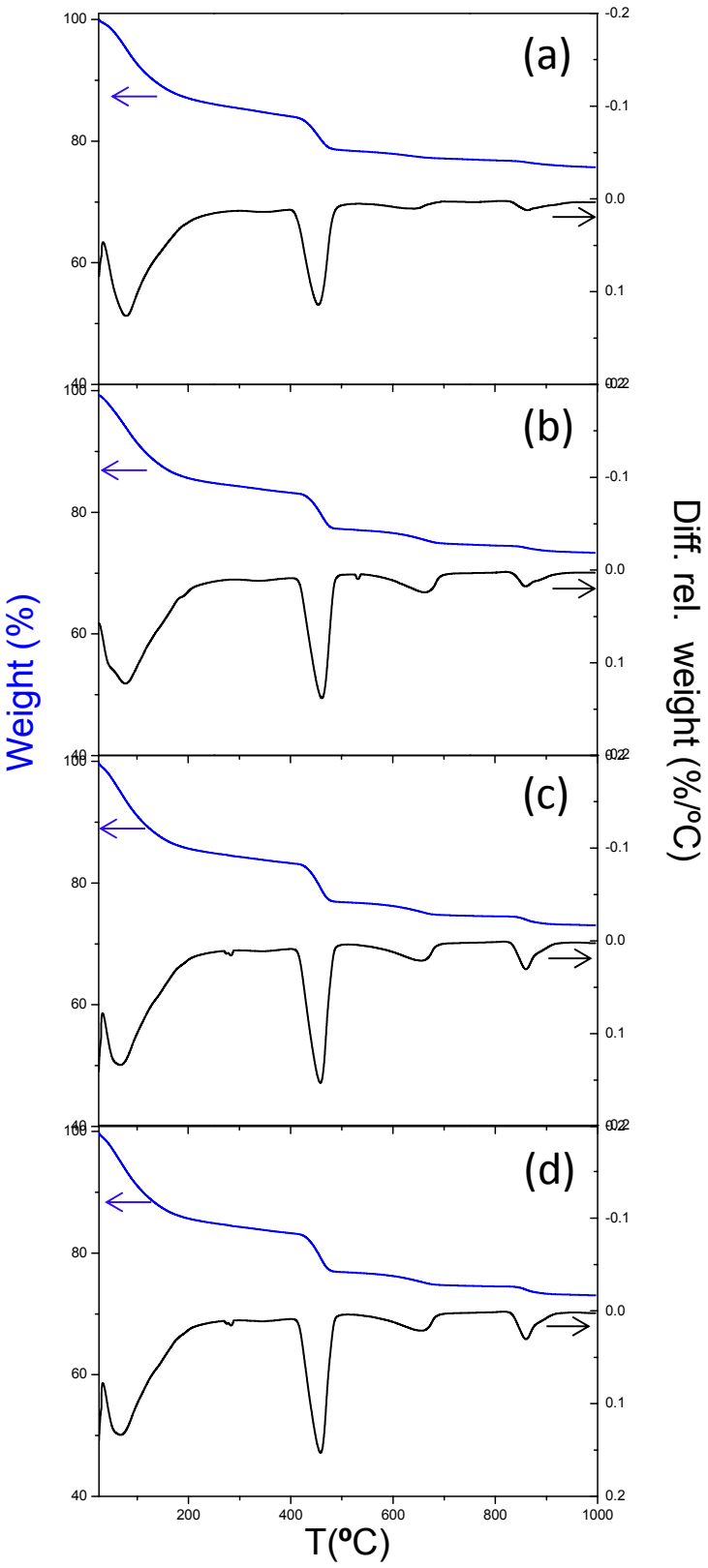

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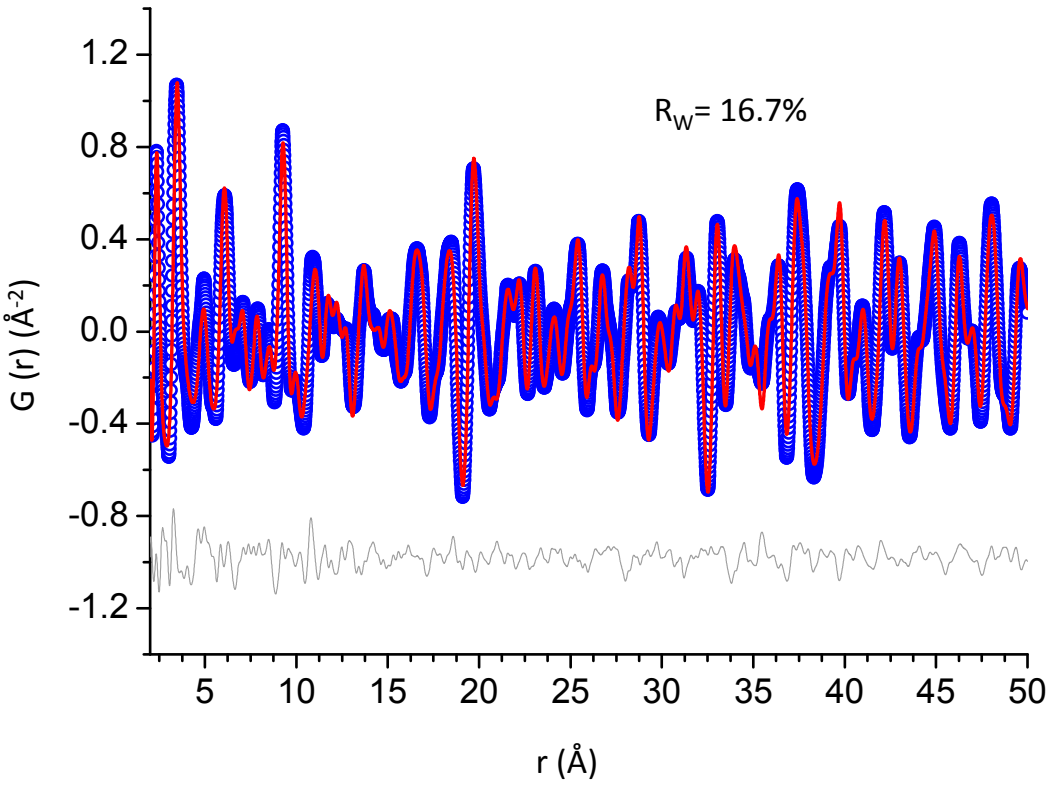

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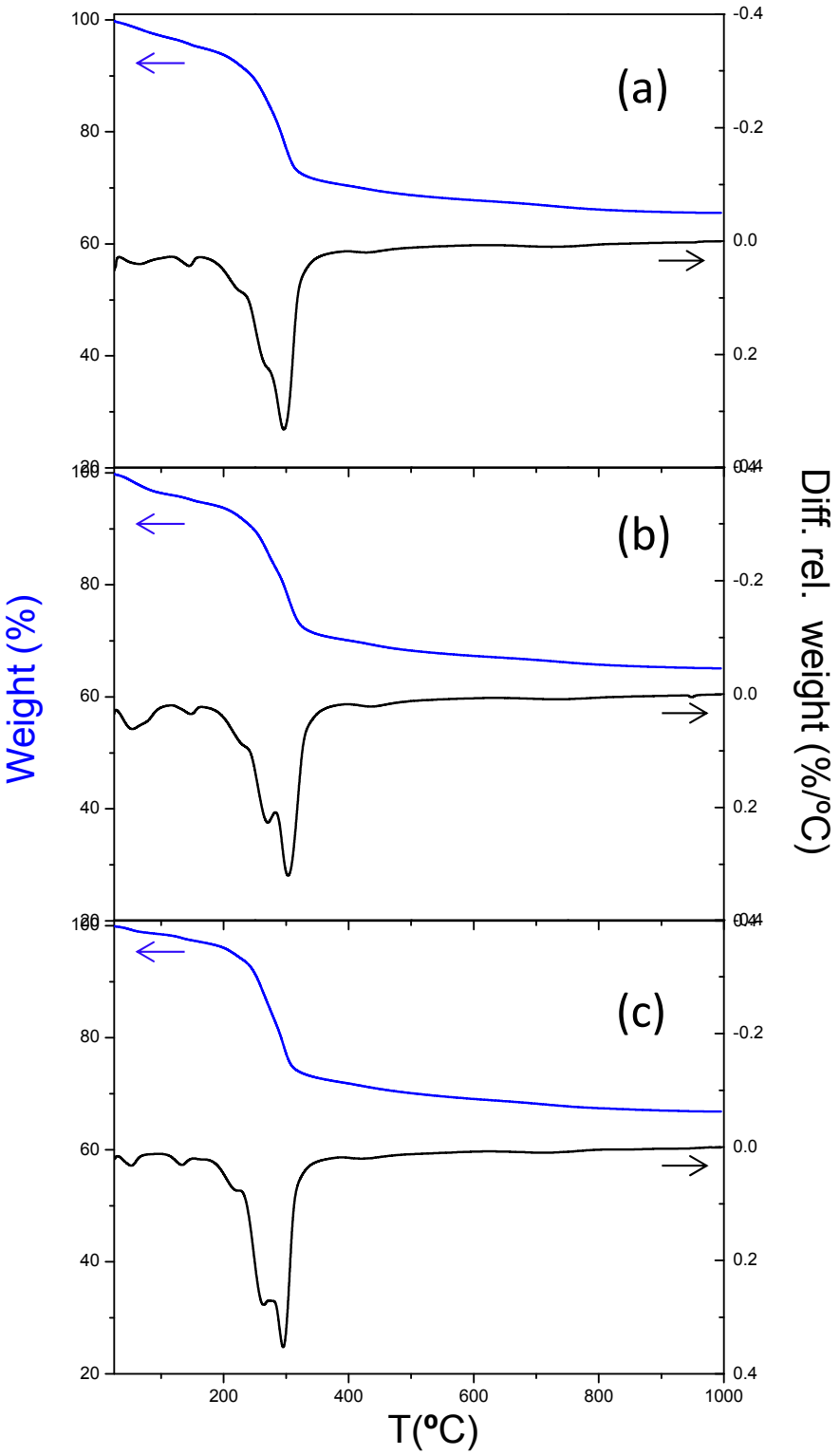

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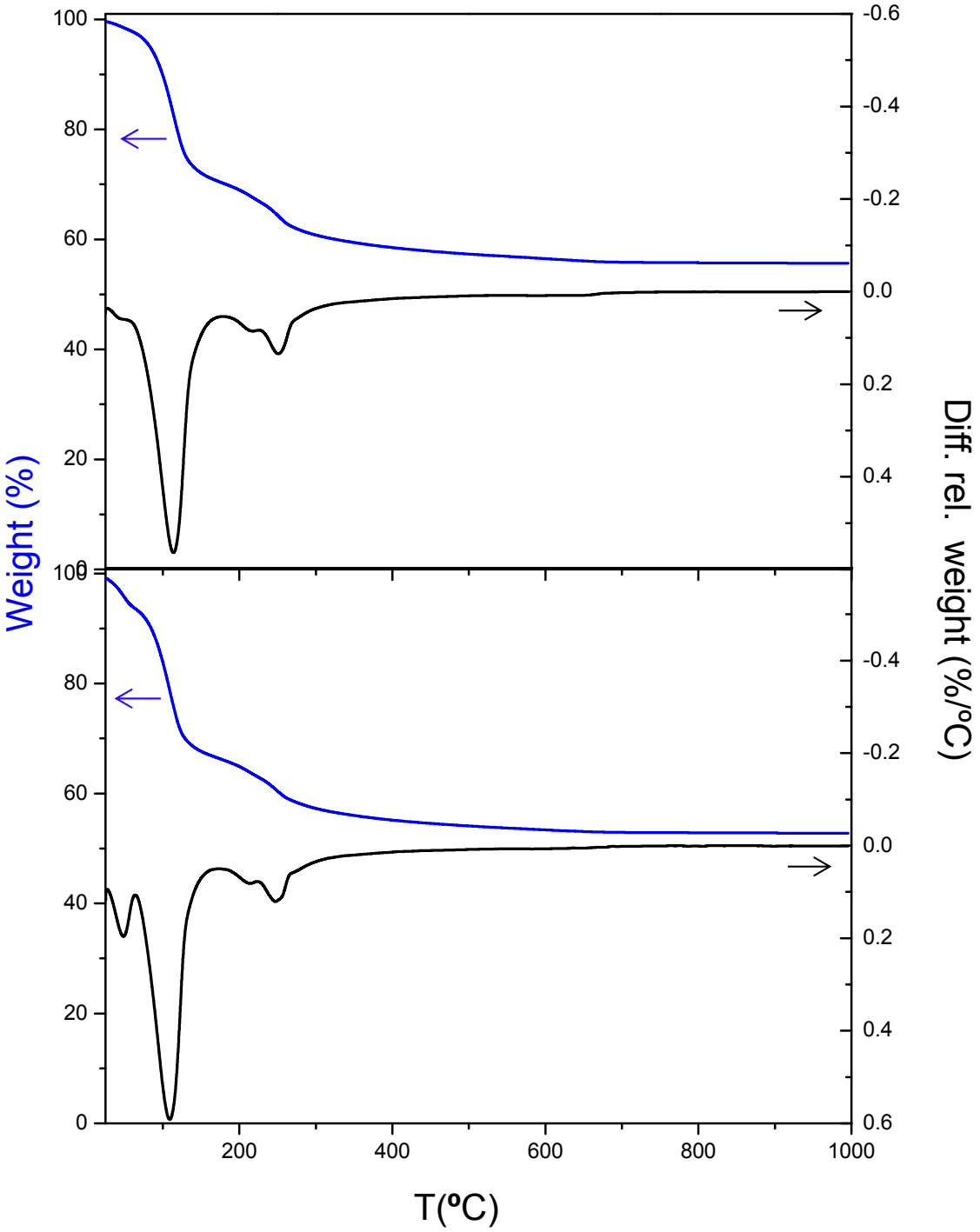

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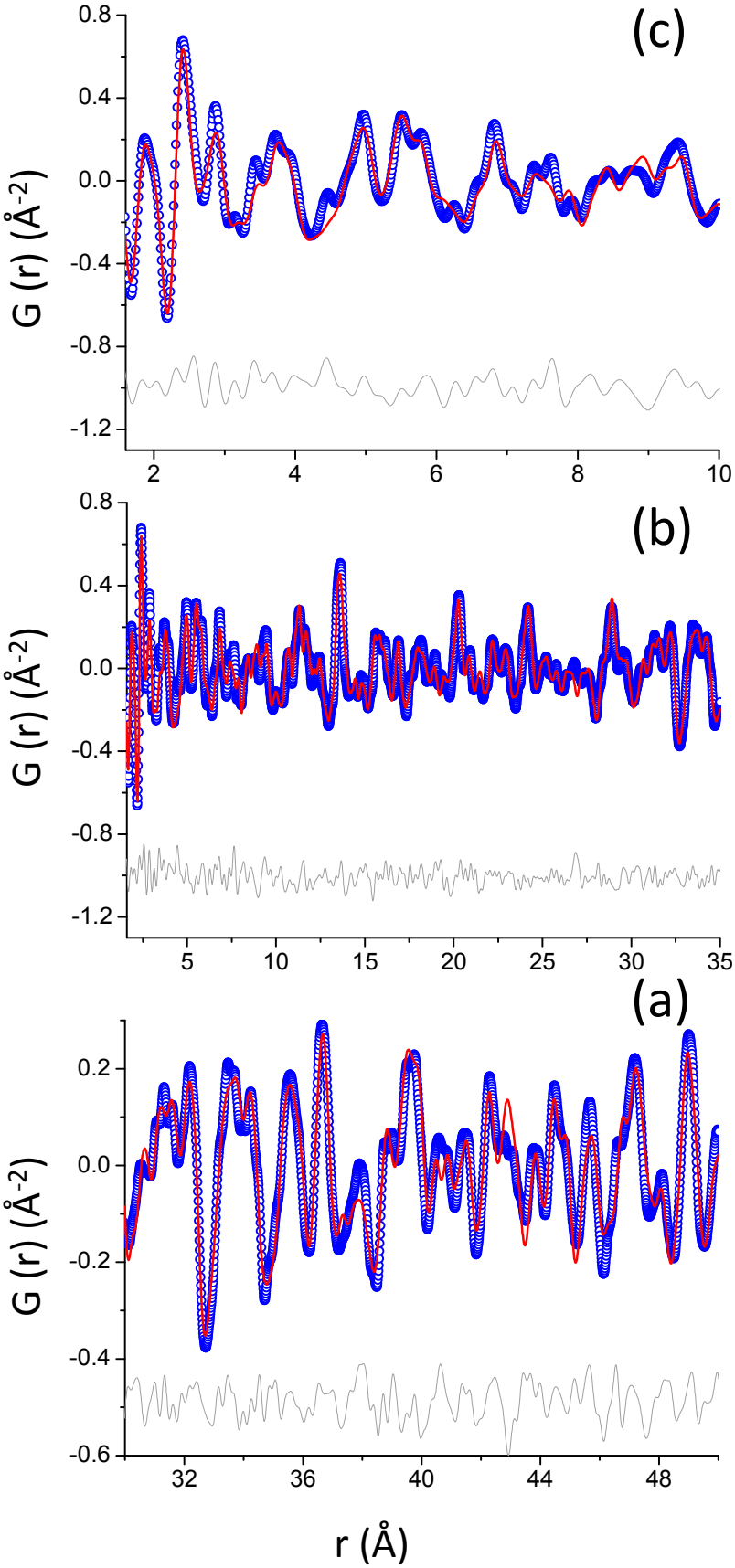

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