

Supporting Information

MgAl-Layered Double Hydroxide Nanoparticles as Smart Nanofillers to Control the Rheological Properties and the Residual Porosity of Cement-Based Materials

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EXPERIMENTAL

Table S1. Chemical composition of the cement (wt.%).

Component	wt. %
CaO	59.7
SiO ₂	19.1
Al ₂ O ₃	4.5
Fe ₂ O ₃	2.6
SO ₃	3.0
MgO	1.3
Na ₂ O	0.3
K ₂ O	0.9
TiO ₂	0.2

Table S2. Ionic species composition of the cement pore solution (determined by ion chromatography).

Component	Concentration (g/L)
Carbonate	0.5
Chloride	1.45
Calcium	0.66
Sodium	0.38
Magnesium	0.0002
Potassium	2.34
Sulfate	4.20

RESULTS AND DISCUSSION

Table S3. Basal distances and average crystallite sizes for the different samples.

Sample	d (nm)	Average crystallite size (nm)
LDH	0.761	30.8
RLDH-W	0.766	10.7
RLDH-C	0.766	10.9

Table S4. Texture parameters obtained by mercury porosimetry and N₂ adsorption-desorption isotherms for the cement specimens prepared with different contents of MO (0, 0.5, 1.0, and 2.0 wt.%).

MO content (wt. %)	Capillary pores volume (cm³ g⁻¹)	Gel pores volume (cm³ g⁻¹)	Average capillary pore size (μm)	Average gel pore size (μm)	Pores area (m² g⁻¹)	S_{BET} (m² g⁻¹)
0	0.0138	0.0482	0.140	0.0113	21	60
0.5	0.0124	0.0418	0.140	0.0114	19	45
1.0	0.0120	0.0452	0.125	0.0133	19	36
2.0	0.0086	0.0408	0.113	0.0105	18	39

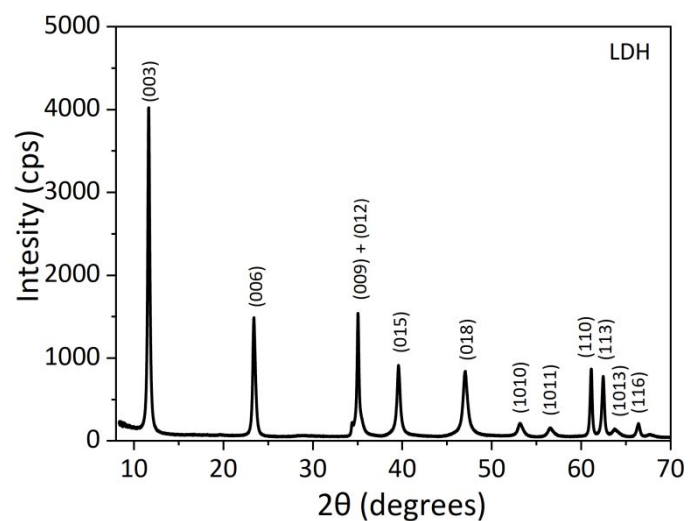


Fig. S1. XRD pattern of the layered double hydroxide (LDH).

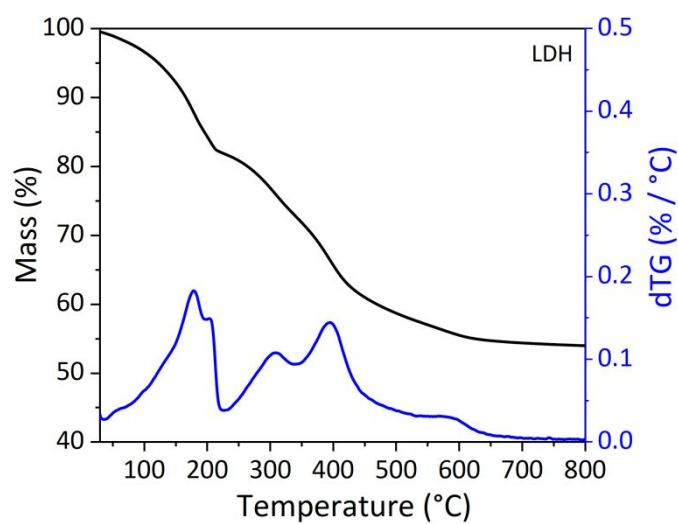


Fig. S2. Thermogravimetric analysis of the LDH sample.

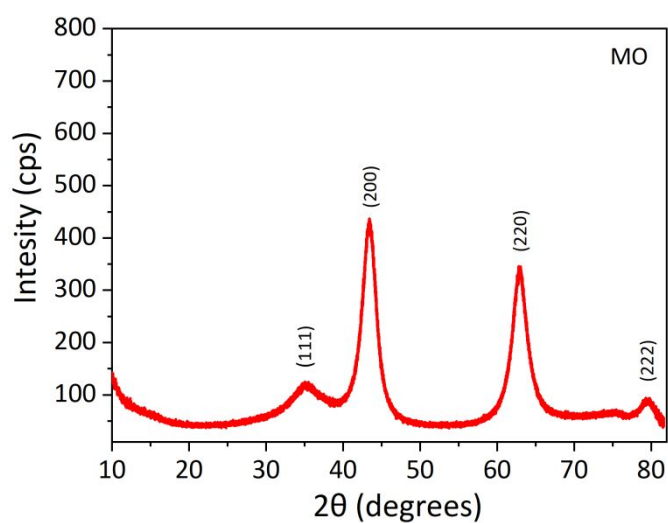


Fig. S3. X-ray diffractogram of the MO sample obtained by calcination of LDH at 500 °C, under an air atmosphere.

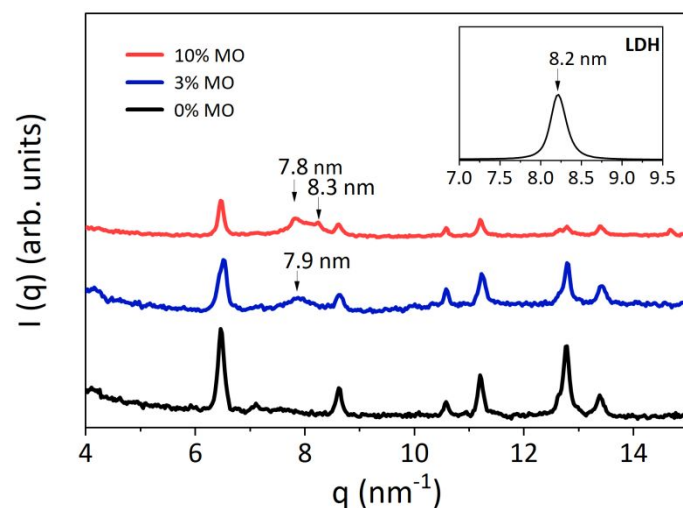


Fig. S4. WAXS pattern of the cement samples containing 0, 3, and 10 wt.% of MO, after hydration for 3 days. The inset shows the peak for the pure LDH.

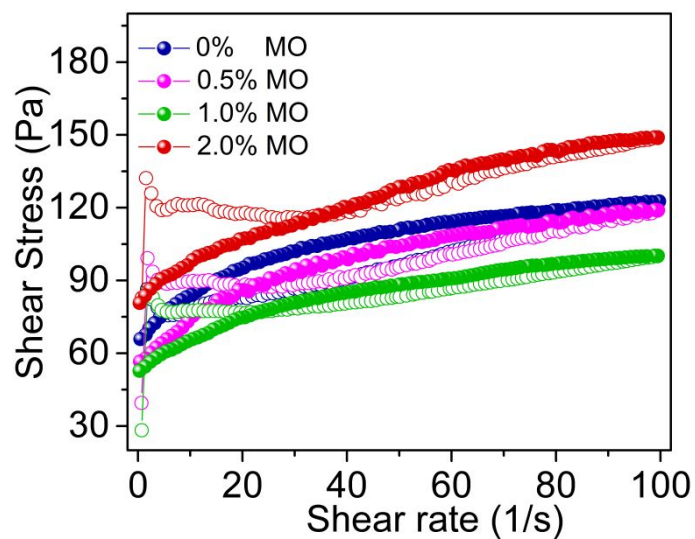


Fig. S5: Second cycle of flow test for the different cement paste formulations, with acceleration (open symbols) and deceleration (closed symbols) curves.

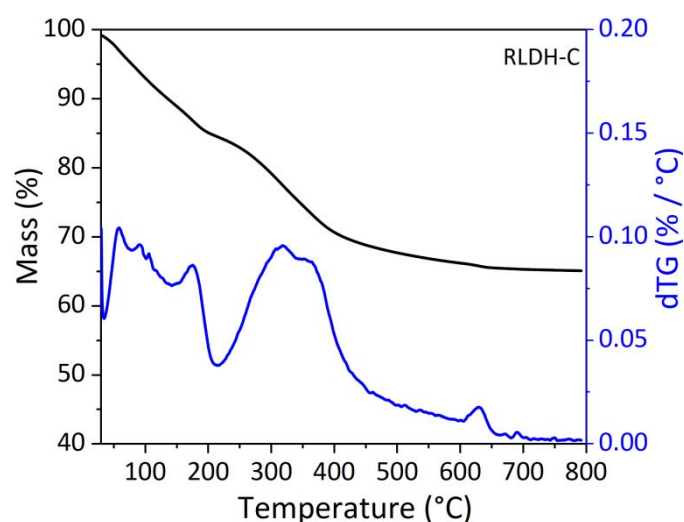


Fig. S6. Thermogravimetric analysis of the RLDH-C sample after 3 h of regeneration.

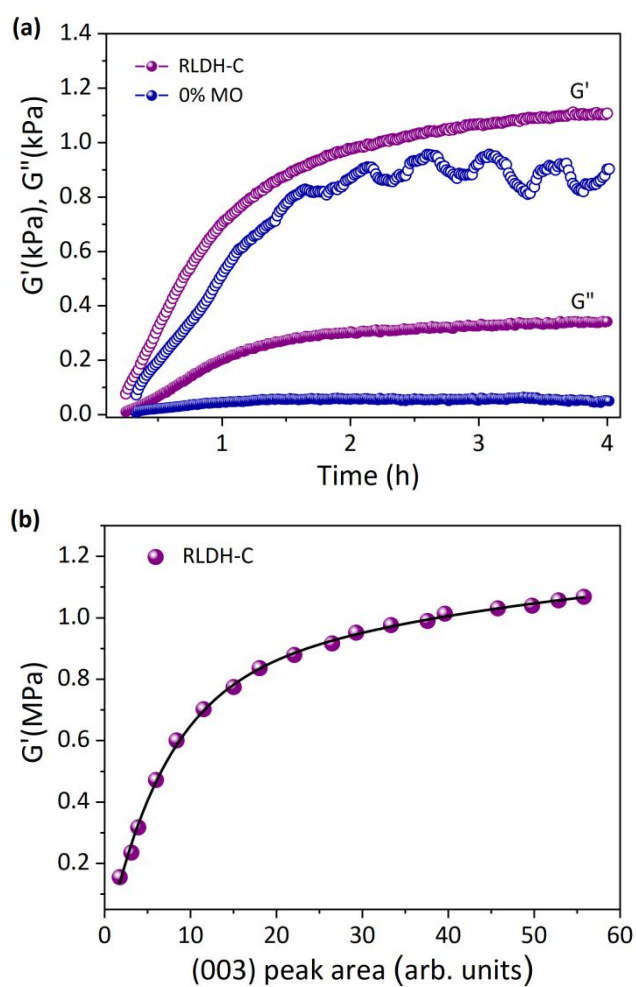


Fig. S7. (a) Elastic (G') and viscous (G'') components, as a function of time, for the MO during regeneration, and for the cement paste without MO. (b) Elastic component (G') for the RLDH-C, as a function of the (003) peak diffraction area obtained by WAXS analysis.

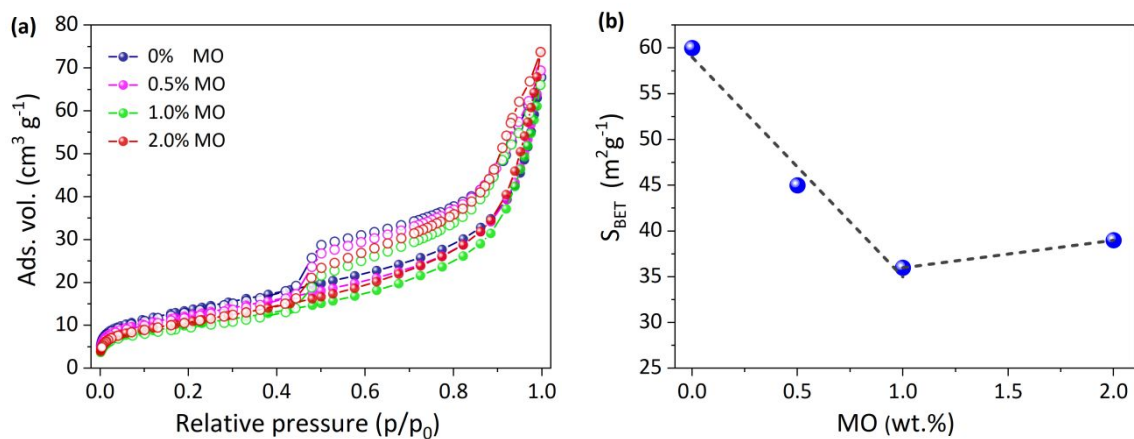


Fig. S8. (a) N₂ adsorption-desorption isotherms and (b) BET area for the hydrated cement specimens prepared with different contents of MO (0, 0.5, 1.0, and 2.0 wt.%).