

Supplement

Effect of a Pre-dispersed GO:PCE Suspension on Compressive Strength and Density of Cement Paste

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1 Hardened-state characterization of GO-modified cement paste

1.1 Mercury intrusion porosimetry of GO-modified cement paste at 28 days

At 28 days, randomly selected fragments were obtained from hardened cement-paste cubes. The fragments were dried at 120 °C to constant mass using a KERN DBS60-3 moisture balance. Their density was determined using an Anton Paar Ultrapyc 5000 helium pycnometer. Mercury intrusion porosimetry

(MIP) was then performed using a Quantachrome PoreMaster33 instrument. The low- and high-pressure measurement stages were combined to obtain the MIP-accessible pore-entry distributions and normalized intrusion-volume metrics for pore-entry diameters below and above 10 μm (Fig. S1). The corresponding normalized MIP-accessible intrusion-volume metrics are summarized in Table S1.

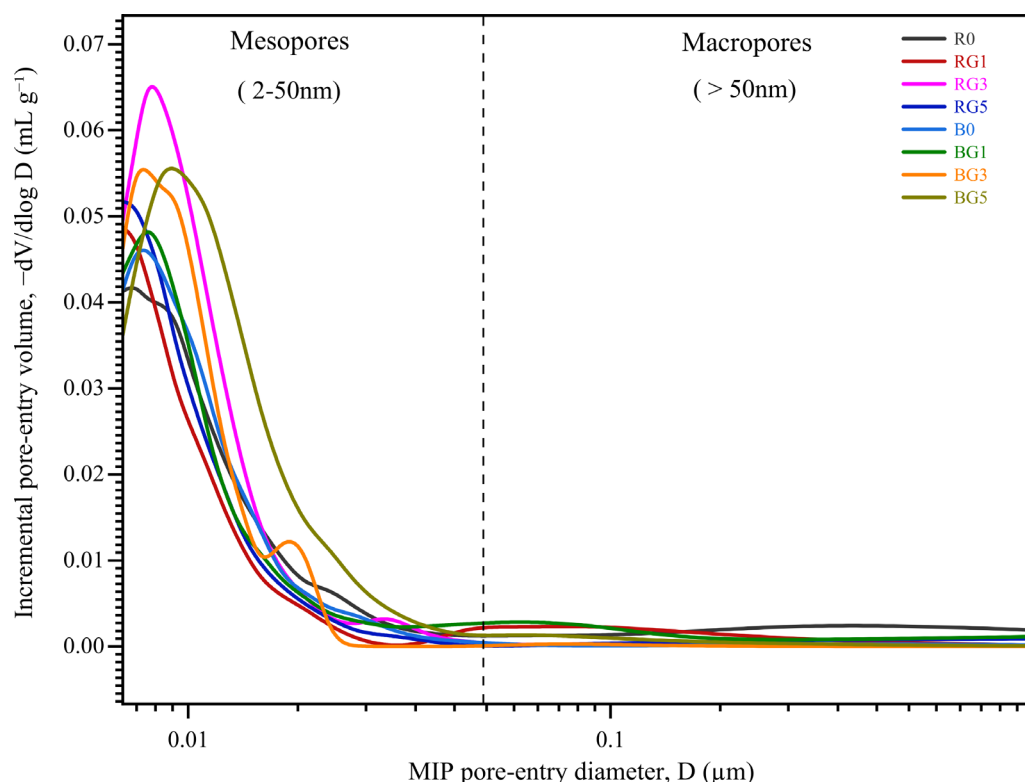


Fig. S1 Incremental MIP pore-entry distributions of 28-day R-series and B-series cement pastes containing 0, 0.01, 0.03, and 0.05% GO. Fig. S1 presents the displayed fine pore-entry diameter range to facilitate comparison of the differential intrusion profiles. Curves were plotted from instrument-exported incremental intrusion data without line smoothing. Integrated MIP-accessible intrusion-volume metrics for the broader measurement range are reported separately in Table S1.

Table S1 MIP-accessible intrusion-volume metrics of 28-day cement pastes, partitioned by pore-entry diameter below and above 10 μm

Mixture	Sample mass, g	<10 μm , ml	>10 μm , ml	Density, g/ml	<10 μm normalized intrusion volume, %	>10 μm normalized intrusion volume, %	Total normalized intrusion volume, %
R0	2.3110	0.0404	0.0092	2.1650	3.8	0.9	4.6
RG1	2.4066	0.0447	0.0268	2.1716	4.0	2.4	6.5
RG3	2.0020	0.0437	0.0168	2.1810	4.8	1.8	6.6
RG5	2.2602	0.0383	0.0139	2.1786	3.7	1.3	5.0
B0	1.9549	0.0290	0.0269	2.1541	3.2	3.0	6.2
BG1	2.2349	0.0469	0.0315	2.1368	4.5	3.0	7.5
BG3	1.9300	0.0395	0.014	2.1286	4.4	1.5	5.9
BG5	1.9225	0.0445	0.0183	2.1557	5.0	2.1	7.0

Note: The values below and above 10 μm were obtained from the combined low- and high-pressure MIP stages. Helium pycnometric density was determined before MIP testing. The percentage values were calculated as intruded pore volume divided by helium-pycnometric solid volume, multiplied by 100. They represent normalized MIP-accessible intrusion-volume metrics and are not conventional bulk-volume porosity values.

1.2 X-ray diffraction characterization of GO-modified cement paste at 28 days

X-ray diffraction (XRD) was used to identify the main crystalline phases in all eight investigated cement paste mixtures at 28 days (Fig. S2). The analyses were conducted at the Advanced Materials Characterization Facility, University of Dundee, using a laboratory diffractometer with Cu K α radiation. Powder samples were obtained from the inner part of the hardened specimens to reduce the influence of surface carbonation. The XRD patterns were interpreted qualitatively to identify the principal

crystalline hydration products and residual phases, including portlandite, ettringite, carbonate-related phases, and residual clinker-related phases.

1.3 Scanning electron microscopy

Selected scanning electron microscopy (SEM) micrographs of 28-day hardened cement pastes are presented in Figs. S3 and S4. The comparison includes the GO-free reference pastes R0 and B0 and the corresponding 0.03% GO pastes RG3 and BG3 prepared through the fixed GO:PCE suspension route. Within each cement series, the 0.03% GO mixture

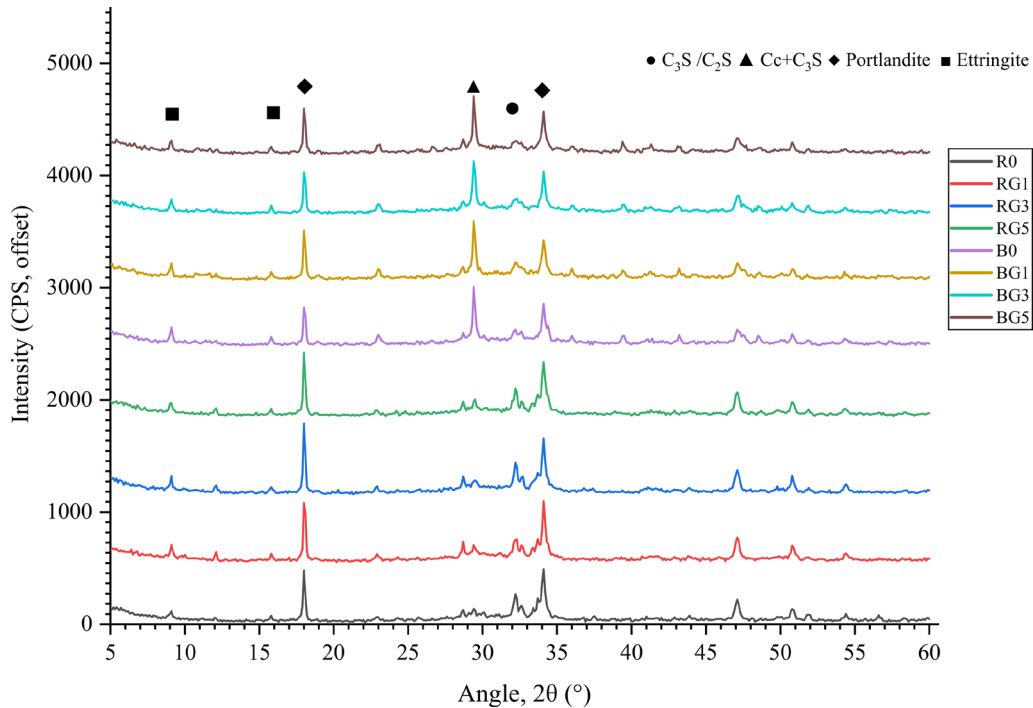


Fig. S2 Qualitative X-ray diffraction patterns of 28-day R-series and B-series cement pastes containing 0, 0.01, 0.03, and 0.05% GO. The patterns are vertically offset for readability. The indicated reflections correspond to $\text{C}_3\text{S}/\text{C}_2\text{S}$, calcite (Cc) with overlapping C_3S contribution, portlandite, and ettringite. Fig. S2 is intended for qualitative phase identification only and is not used for quantitative phase comparison.

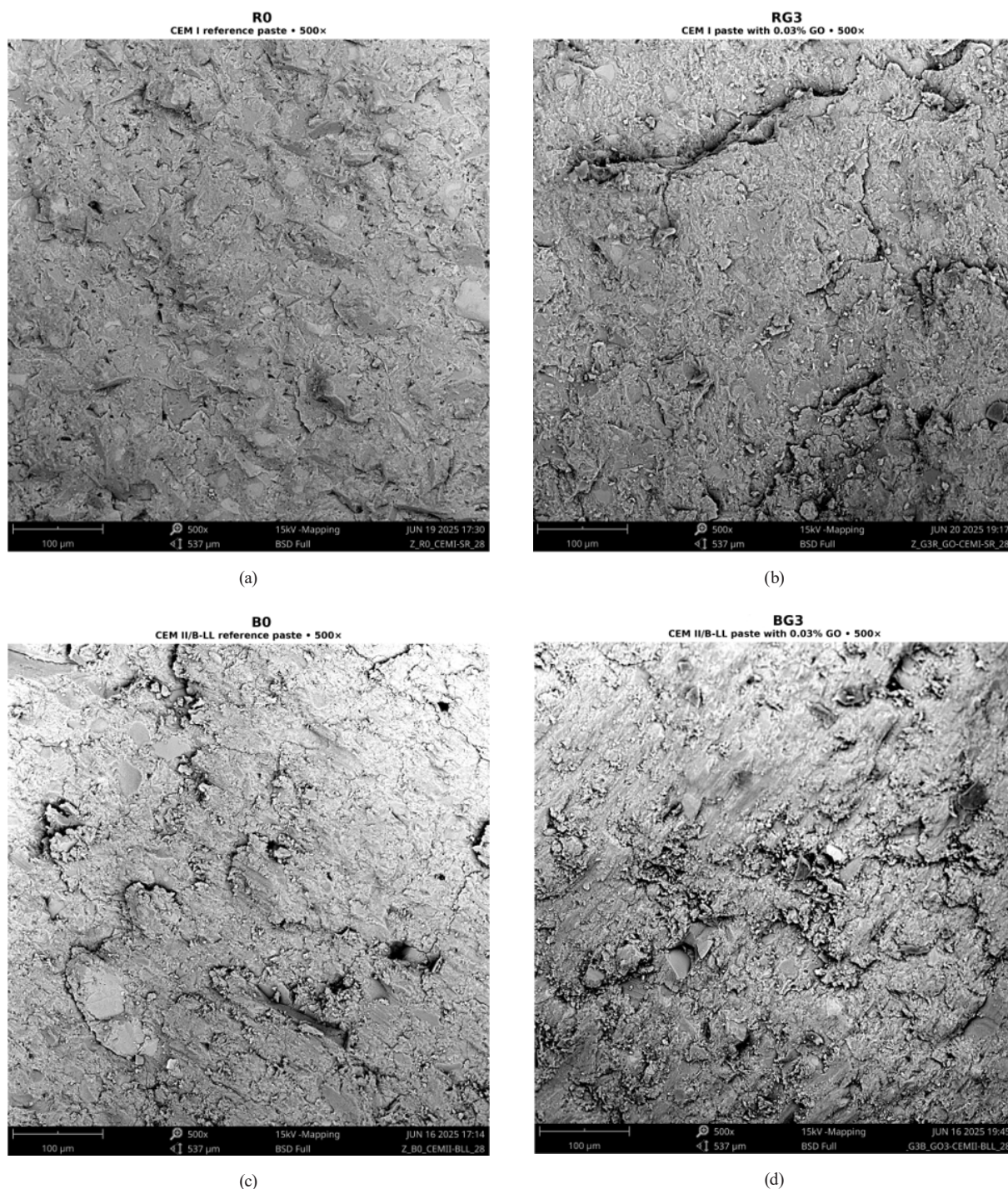
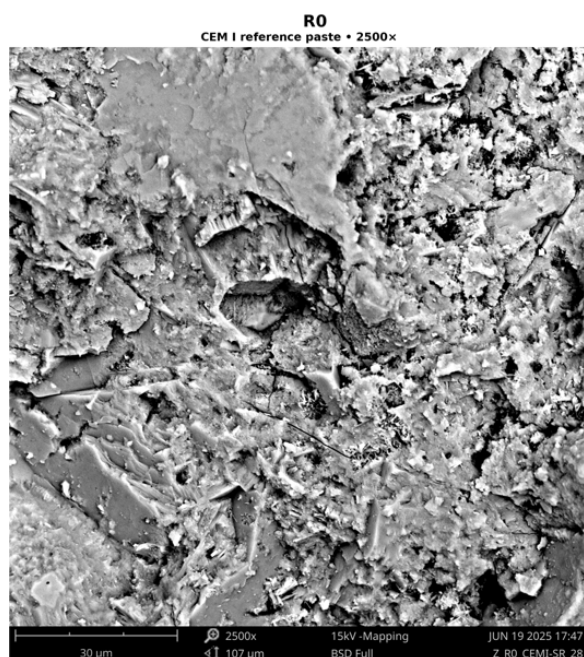


Fig. S3 Selected SEM micrographs of internal fractured surfaces of 28-day hardened cement pastes at 500× magnification: (a) R0, CEM I 42.5 N-SR0 GO-free reference paste; (b) RG3, CEM I 42.5 N-SR0 paste prepared through the fixed GO:PCE suspension route at 0.03% GO; (c) B0, CEM II/B-LL 42.5 N GO-free reference paste; and (d) BG3, CEM II/B-LL 42.5 N paste prepared through the fixed GO:PCE suspension route at 0.03% GO. Specimens were obtained from internal fractured surfaces after compressive-strength testing, gold-coated for 30 s, and imaged using a Phenom XL desktop SEM at 15 kV with the BSD Full setting.

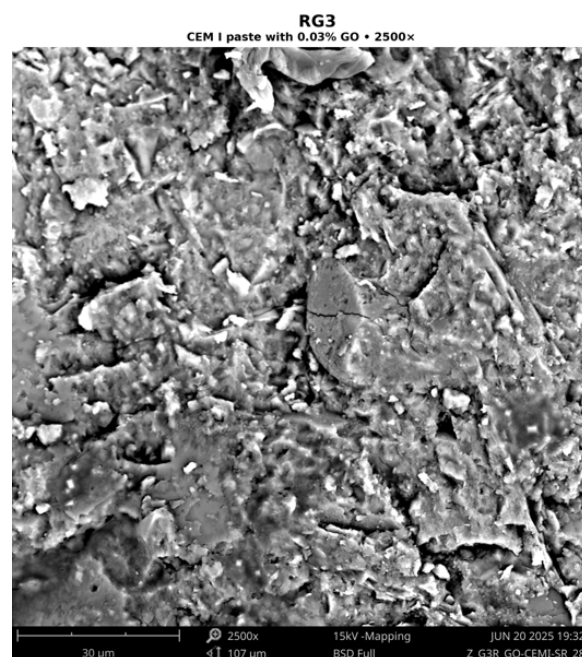
was selected because it showed the highest measured mean compressive strength at 28 days.

SEM specimens were taken from internal fractured surfaces obtained after compressive-strength testing. The fractured surfaces were gold-coated for 30 s and examined using a Phenom XL desktop

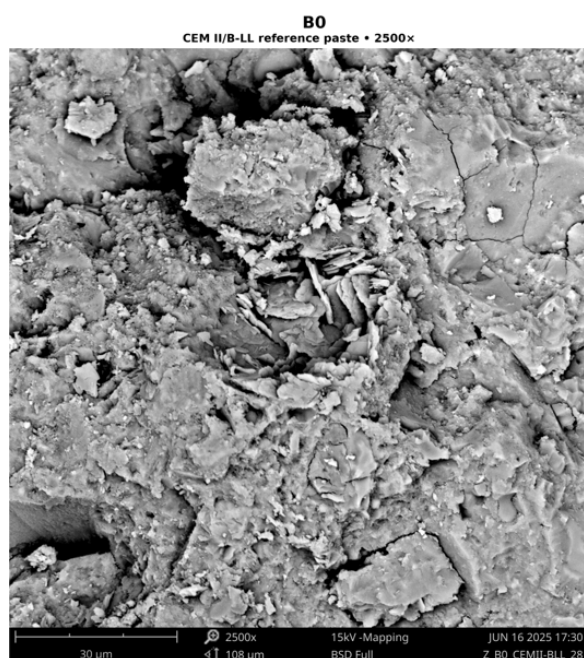
SEM at an accelerating voltage of 15 kV with the manufacturer-designated BSD Full imaging setting. Fig. S3 presents selected fields at 500× magnification, whereas Fig. S4 provides selected local views at 2500× magnification. The original scale bars and instrument footers are retained in all panels. At both



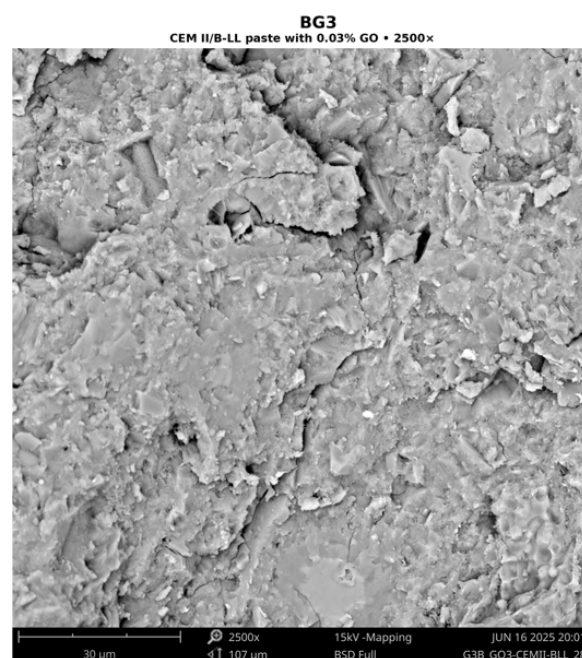
(a)



(b)



(c)



(d)

Fig. S4 Selected SEM micrographs of internal fractured surfaces of 28-day hardened cement pastes at 2500× magnification: (a) R0, CEM I 42.5 N-SR0 GO-free reference paste; (b) RG3, CEM I 42.5 N-SR0 paste prepared through the fixed GO:PCE suspension route at 0.03% GO; (c) B0, CEM II/B-LL 42.5 N GO-free reference paste; and (d) BG3, CEM II/B-LL 42.5 N paste prepared through the fixed GO:PCE suspension route at 0.03% GO. These selected local views are presented as qualitative illustrations of fractured-surface morphology only and are not used for phase identification or quantitative microstructural comparison.

magnifications, the selected fractured-surface fields show local morphological heterogeneity, including rough granular regions, locally smoother plate-like regions, fracture separations, and surface depressions. These features are directly visible only at the selected locations and vary among the individual

fractured-surface fields. The 2500× images provide closer visualization of local surface relief and fracture morphology but do not provide a separate quantitative comparison among mixtures. The SEM observations are included as qualitative visual documentation only. No phase identification, elemental

assignment, quantitative pore analysis, direct observation of graphene oxide sheets, or conclusion regarding pore refinement, matrix densification, hydration degree, or the mechanism responsible for the measured compressive-strength differences is derived from these images. In particular, differences in gray-scale brightness or local contrast among panels are not interpreted as material-property differences.

2 Individual compressive-strength data

Table S2 provides the individual cube compressive-strength results at 7 and 28 days for all investigated mixtures. The data supplement the summary values reported in the main manuscript.

Table S2 Individual compressive-strength values underlying the mean $\pm$ SD results reported in the main manuscript

Mixture	Age, d	Cube 1, MPa	Cube 2, MPa	Cube 3, MPa	Cube 4, MPa	Cube 5, MPa	Mean $\pm$ standard deviation, MPa	Coefficient of variation, %	Range, MPa
R0	7	78.43	79.76	80.14	83.88	80.65	80.57 $\pm$ 2.02	2.51	78.43–83.88
RG1	7	82.24	88.43	89.47	87.83	84.12	86.42 $\pm$ 3.09	3.57	82.24–89.47
RG3	7	94.83	92.39	92.91	96.51	94.63	94.25 $\pm$ 1.65	1.75	92.39–96.51
RG5	7	83.55	87.10	90.71	76.51	79.39	83.45 $\pm$ 5.72	6.85	76.51–90.71
R0	28	89.19	99.37	94.92	92.38	86.05	92.38 $\pm$ 5.14	5.56	86.05–99.37
RG1	28	110.49	114.04	102.14	110.76	98.94	107.27 $\pm$ 6.40	5.97	98.94–114.04
RG3	28	106.53	117.50	109.85	115.58	116.35	113.16 $\pm$ 4.74	4.19	106.53–117.50
RG5	28	108.48	102.74	98.82	98.91	98.88	101.57 $\pm$ 4.21	4.15	98.82–108.48
B0	7	67.99	70.95	71.76	62.50	66.93	68.03 $\pm$ 3.68	5.41	62.50–71.76
BG1	7	77.40	74.42	75.73	73.96	76.26	75.55 $\pm$ 1.39	1.84	73.96–77.40
BG3	7	75.15	78.33	77.04	78.89	79.68	77.82 $\pm$ 1.77	2.28	75.15–79.68
BG5	7	78.04	67.97	79.19	76.99	76.35	75.71 $\pm$ 4.46	5.89	67.97–79.19
B0	28	77.64	90.35	96.90	83.63	101.39	89.98 $\pm$ 9.63	10.70	77.64–101.39
BG1	28	108.25	103.32	102.40	107.73	100.44	104.43 $\pm$ 3.42	3.27	100.44–108.25
BG3	28	107.27	107.81	109.45	100.79	107.48	106.56 $\pm$ 3.34	3.13	100.79–109.45
BG5	28	95.06	94.82	90.29	100.69	95.27	95.23 $\pm$ 3.69	3.87	90.29–100.69

Note: Each mixture was prepared as one paste batch. At each test age, five cube specimens (30 $\times$ 30 $\times$ 30 mm) were cast from that batch and destructively tested. Mean $\pm$ SD, coefficient of variation (CV), and range describe specimen-level variability within the tested batch. The cube specimens do not represent independently prepared paste batches. The statistical comparison of the compressive-strength results is reported in Table 4 in the main manuscript.