Supporting information for article:

Quantitative X-ray pair distribution function analysis of nanocrystalline calcium silicate hydrates: a contribution to the understanding of cement chemistry

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Table S1  All parameters refined during PDF data simulation but those reported in the main text. The corresponding input file is given in Figure S1.

| Target Ca/Si ratio | Scale factor | Delta2 (Å²) | Spdiameter (Å) | Ca and Si atoms | O atoms |
|-------------------|--------------|-------------|----------------|-----------------|---------|
|                   |              |             |                | u₁₁ and u₂₂ (Å²) | u₃₃ (Å²) | u₁₁ and u₂₂ (Å²) | u₃₃ (Å²) |
| 0.6               | 1.08         | 2.52        | 28             | 0.010           | 0.002   | 0.004            | 0.04    |
| 0.8               | 1.17         | 2.54        | 37             | 0.006           | 0.008   | 0.007            | 0.07    |
| 1                 | 1.17         | 2.52        | 41             | 0.006           | 0.009   | 0.007            | 0.06    |
| 1.2               | 1.18         | 2.52        | 30             | 0.005           | 0.008   | 0.008            | 0.06    |

Notes: delta2 is a quadratic atomic correlation factor. U₃₃ associated to layer oxygen atoms are higher than the corresponding U₁₁ and U₂₂ values, certainly because the c lattice parameter was fixed during the modelling procedure so as to reduce the number of free parameters.
Table S2  Detail of the four PDF simulations performed on C-S-H samples having a target Ca/Si of 1.2.

| Parameter | Constraints on occupancy of Si sites |
|-----------|-------------------------------------|
|           | Bridging sites refined | Paired sites refined | Equal occupancy of bridging and paired sites | Bridging and paired sites refined independently |
| \( a (\text{Å}) \) | 6.685(8) | 6.678(8) | 6.680(8) | 6.683(8) |
| Scale factor | 1.16(6) | 1.25(7) | 1.23(7) | 1.18(7) |
| Delta2 (Å²) | 2.5(1) | 2.5(1) | 2.5(1) | 2.5(1) |
| Spdiameter (Å) | 29(4) | 31(4) | 31(4) | 30(4) |
| \( z \)-coordinate of bridging Si | 0.065(4) | 0.067(1) | 0.0678(2) | 0.066(4) |
| Ca and Si \( U_{11} \) and \( U_{22} \) (Å²) | 0.005(1) | 0.006(1) | 0.005(1) | 0.005(1) |
| Ca and Si \( U_{33} \) (Å²) | 0.007(3) | 0.011(4) | 0.011(4) | 0.008(3) |
| O \( U_{11} \) and \( U_{22} \) (Å²) | 0.007(5) | 0.007(2) | 0.008(2) | 0.008(2) |
| O \( U_{33} \) (Å²) | 0.06(1) | 0.06(1) | 0.06(1) | 0.06(1) |
| Occupancy of bridging Si | 0.37(9) | 1* | 0.78(4)§ | 0.45(11) |
| Occupancy of paired Si | 1* | 0.75(5) | 0.78(4)§ | 0.91(6) |
| \( R_{WP} \) | 32.70% | 34.66% | 35.74% | 32.41% |

Notes: §Relative to the model given in Figure S1; *fixed; covaried
Figure S1: Typical PdfGUI input file used to model PDF patterns. Greyed parameters are those which were possibly refined (see main text and Table SI1).