**SUPPLEMENTARY INFORMATION**

**Molecular dynamic simulations of cementitious systems using a newly developed force field suite ERICA FF**

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**S1. Total energy calculations**

To calculate the total energy in molecular mechanics, we have to evaluate the appropriate energy term for every atom-atom interaction in the system. Generally, the total energy is expected to have contributions from the Coulombic (electrostatic) interactions, the short-range interactions (often referred to as the Van der Waals (VdW) term), the bonded interactions and angle interactions. The equations used and charges assigned to the different atoms are given in equations E1-E8 and Table S1.

|  |  |
| --- | --- |
|  | (E1) |

In the SPC/Fw water model [1], the bonded term includes bond stretch and angle bend energy and is represented by the harmonic term. For all bonded pairs, Coulombic and VdW terms are considered except for OSi\_Core-OSi\_Shell that only Coulombic and bond terms were considered (We refer the reader to the LAMMPS documentation [2] on the implementation of the adiabatic core-shell model). In our model, there are four types of bonds: OOH-HOH in hydroxyl is represented by Morse bond potential, Ow-Hw bond in water is represented by harmonic term, as are the OSi\_Core­-O­Si\_Shell bond and the sulfate bonds between S-OS.

For Morse (E2) and harmonic bonds (E3) we used the below equations:

(E2)

(E3)

Where and are bond coefficients is the equilibrium bond length, and is stiffness parameter.

For the angles we used the harmonic angle description (E4):

(E4)

For VdW we have used three sets of potentials: 12-6 Lennard-Jones (E5), N-M Potential (E6), and Buckingham potential (E7):

|  |  |
| --- | --- |
|  | (E5) |
|  | (E6) |
|  | (E7) |

The last contribution is a Coulombic or electrostatic potential which is shown in equation 8:

|  |  |
| --- | --- |
|  | (E8) |

Where the numerical value of charges Qi and Qj (multiplied by the electronic charge e), are as given in table S1. Please note that is the Coulombic constant and is the vacuum permittivity.

Table S1. Charges of atom species in different force fields discussed in the main text.

|  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- |
|  | **Cement FF1** | **Cement FF2** | **Erica FF** | **Byrne** | **SPC/Fw** | **CLAYFF** | **Predota** |
| **Ref** | [3] | [4] |  | [5] | [1] | [6] | [7] |
| **Ca** | + 2.00 | + 2.00 | + 2.00 | + 2.00 | - | + 1.36 | - |
| **OOH** | - 1.40 | - 1.40 | - 1.40 | - | - | - 0.95 | - |
| **HOH** | + 0.40 | + 0.40 | + 0.40 | - | - | + 0.425 | - |
| **Ow** | - 1.1128\* | -1.1128\* | - 0.82 | - 0.82 | - 0.82 | - 0.82 | - |
| **Hw** | + 0.5564 | 0.5564 | + 0.41 | + 0.41 | + 0.41 | + 0.41 | - |
| **Si** | + 4.00 | + 4.00 | + 4.00 | - | - | - | - |
| **Osi\_Shell** | - | - 2.85 | - 2.85 | - | - | - | - |
| **OSi\_Core** | - 2.00 | + 0.85 | - 2.00 | - | - | - | - |
| **OS** | - | - | - 0.84 | - 0.84 | - | - | - |
| **Al** | - | + 3.00 | + 3.00 | - | - | + 1.575 | - |
| **S** | - | - | + 1.36 | + 1.36 | - | - | - |
| **Na** | - | - | + 1.00 | - | - | + 1.00 | + 1.00 |
| **Cl** | - | - | - 1.00 | - | - | - 1.00 | - 1.00 |
| **K** | - | - | + 1.00 | - | - | + 1.00 | + 1.00 |

\* On the site of the TIP4P water model [8]

**S2. ERICA FF1 parameters**

**Van der Waal interactions**

Oxygens: OS – Sulfur Oxygen, Ow ­– Water Oxygen, OOH – Hydroxyl Oxygen

Hydrogens: Hw – Water Hydrogen, HOH ­– Hydroxyl Hydrogen

1. **Buckingham Potential – Equation E7**

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| **Atom 1** | **Atom 2** | **Force Field** | **A [eV]** | **ρ [Å]** | **C [eV Å6]** | **Ref** |
| Ca | OS | Byrne et al. | 1815.7 | 0.2834 | 0.0 | [5] |
| Ca | OOH | Cement FF2 | 2251.05 | 0.297 | 0.0 | [4] |
| Ow | OS | Byrne et al. | 12534.5 | 0.246 | 0.0 | [5] |
| Ow | HOH | Erica FF | 21562.5 | 0.1065 | 0.0 |  |
| OOH | OOH | Cement FF2 | 22764.0 | 0.149 | 6.97 | [4] |
| OS | OS | Byrne et al. | 103585.02 | 0.2 | 0.0 | [5] |

1. **N-M Potential – Equation 6**

|  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- |
| **Atom 1** | **Atom 2** | **Force Field** | **E0 [eV]** | **r0 [Å]** | **n [-]** | **m [-]** | **cutoff [Å]** | **Ref** |
| OOH | HOH | Cement FF2 | 0.0073 | 2.71 | 8.5 | 9 | 6 | [4] |

1. **L-J Potential – Equation 5**

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| **Atom 1** | **Atom 2** | **Force Field** | **ε [eV]** | **σ [Å]** | **Ref** |
| Ow | Ow | SPC/Fw | 0.0067 | 3.16 | [1] |
| Ow | OOH | Erica FF | 0.00747 | 3.18 |  |
| OS | OOH |  |
| Ow | Ca | Byrne et al. | 0.00095 | 3.35 | [5] |

**Bonds**

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| **Atom 1** | **Atom 2** | **Force Field** | **Bond type** | **Equation** | **Parameters** | **Ref** |
| Ow | Hw | SPC/Fw | Harmonic | E2 | K = 22.96 eV  r0 = 1.012 Å | [1] |
| OS | S | Byrne et al. | Morse | E3 | D = 5.0 eV  α = 1.2 Å-1  r0 = 1.505 Å | [5] |

**Angles**

1. **Harmonic angle, equation E4**

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| **Centre Atom** | **Atom 1** | **Atom 2** | **Force Field** | **E [eV]** | **ϴ0 [deg]** | **Ref** |
| Ow | Hw | Hw | SPC/Fw | 1.64567 | 113.24 | [1] |
| S | OS | OS | Byrne et al. | 7.50 | 109.47 | [5] |

**S3. ERICA FF2 parameters**

**Van der Waal interactions**

Oxygens: OSi – Silicon Oxygen (Shell), Ow ­– Water Oxygen, OOH – Hydroxyl Oxygen

Hydrogens: Hw – Water Hydrogen, HOH ­– Hydroxyl Hydrogen

1. **Buckingham Potential – Equation E7**

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| **Atom 1** | **Atom 2** | **Force Field** | **A [eV]** | **ρ [Å]** | **C [eV Å6]** | **Ref** |
| Ca | Osi | Cement FF2 | 2152.36 | 0.309 | 0.09944 | [4] |
| Ca | Ow | Cement FF2 | 1286.60 | 0.297 | 0.00 |
| Ca | OOH | Cement FF2 | 2251.05 | 0.297 | 0.00 |
| Si | OSi | Cement FF2 | 1283.91 | 0.321 | 10.66 |
| Si | Ow | Cement FF2 | 1283.56 | 0.321 | 10.66 |
| Si | OOH | Cement FF2 | 983.56 | 0.321 | 10.66 |
| Osi | OSi | Cement FF2 | 22764.30 | 0.149 | 27.88 |
| OSi | Ow | Cement FF2 | 22764.30 | 0.149 | 28.92 |
| OSi | OOH | Cement FF2 | 22764.00 | 0.149 | 13.94 |
| OSi | Hw | Cement FF2 | 512.00 | 0.250 | 0.00 |
| OOH | OOH | Cement FF2 | 22764.30 | 0.149 | 6.97 |
| Al | OSi | Cement FF2 | 1474.40 | 0.301 | 0.00 |
| Al | OOH | Cement FF2 | 1032.00 | 0.301 | 0.00 |
| Al | Ow | Cement FF2 | 590.04 | 0.301 | 0.00 |

For the original interaction we refer to the paper JACS …

1. **N-M Potential, equation E6**

|  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- |
| **Atom 1** | **Atom 2** | **Force Field** | **E0 [eV]** | **r0 [Å]** | **n [-]** | **m [-]** | **Ref** |
| OSi | HOH | Cement FF2 | 0.0073 | 2.71 | 9 | 6 | [4] |
| Ow | OOH | Cement FF2 | 0.0013 | 4.63 | 9 | 6 |
| Ow | HOH | Cement FF2 | 0.0556 | 2.00 | 9 | 6 |
| OOH | HOH | Cement FF2 | 0.0073 | 2.71 | 9 | 6 |

1. **L-J Potential, equation E5**

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| **Atom 1** | **Atom 2** | **Force Field** | **ε [eV]** | **σ [Å]** | **Ref** |
| Ow | Ow | SPC/Fw | 0.0067 | 3.16 | [1] |

**Bonds**

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| **Atom 1** | **Atom 2** | **Force Field** | **Bond type** | **Equation** | **Parameters** | **Ref** |
| Ow | Hw | SPC/Fw | Harmonic | E2 | K = 22.96 eV  r0 = 1.012 Å | [1] |
| OSi\_Core | OSi\_Shell | Cement FF2 | Harmonic | E2 | K = 37.5 eV  r0 = 0 Å | [4] |
| OOH | HOH | Cement FF2 | Morse | E3 | D = 7.0525 eV  α = 3.1749 Å-1  r0 = 0.94285 Å | [4] |

**Angles**

1. **Harmonic Angle, equation E4**

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| **Centre Atom** | **Atom 1** | **Atom 2** | **Force Field** | **E [eV]** | **ϴ0 [deg]** | **Ref** |
| Ow | Hw | Hw | SPC/Fw | 1.64567 | 113.24 | [1] |
| Si | OOH | OOH | Cement FF2 | 7.7482 | 109.47 | [4] |
| Si | OOH | OSi |
| Si | OSi | OSi |
| OOH | HOH | Si | Cement FF2 | 7.7482 | 141.50 | [4] |

**S4. ERICA FF3 parameters**

Erica FF3 includes the parameters from Erica FF2.

**Van der Waal interactions**

Oxygens: OSi – Silicon Oxygen (Shell), OS – Sulphur Oxygen, Ow ­– Water Oxygen, OOH – Hydroxyl Oxygen

Hydrogens: Hw – Water Hydrogen, HOH ­– Hydroxyl Hydrogen

1. **Buckingham Potential – Equation E7**

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| **Atom 1** | **Atom 2** | **Force Field** | **A [eV]** | **ρ [Å]** | **C [eV Å6]** | **Ref** |
| Na | OSi | Tilocca et al. | 56465.34 | 0.194 | 0 | [9] |
| K | OSi | Tilocca et al. | 56465.34 | 0.194 | 0 | [9] |

1. **L-J Potential, equation E5**

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| **Atom 1** | **Atom 2** | **Force Field \*** | **ε [eV]** | **σ [Å]** | **Ref** |
| Na | Ow | Erica FF | 0.0058 | 2.85 |  |
| Na | OOH | Erica FF | 0.0055 | 2.87 |  |
| Na | Cl | Predota et al. | 0.0044 | 3.41 | [10] |
| Cl | Ca | Dang | 0.003 | 4.11 | [11] |
| Cl | Ow | Erika FF | 0.0058 | 3.25 |  |
| Cl | K | Dang | 0.0047 | 3.52 | [11] |
| K | Ow | Erica FF | 0.0058 | 3.81 |  |
| K | OOH | Erica FF | 0.0063 | 3.12 |  |
| OS | Na | Cannon et al. | 0.006071 | 3.12 | [12] |
| OS | Cl | Cannon et al. | 0.0052 | 4.25 | [12] |
| OS | K | Cannon et al. | 0.006071 | 3.12 | [12] |

**S5: Water correction term**

The water dissociation enthalpy can be calculated according to this reaction

H2O + O 2- ⇌ 2 OH-

As shown by de Leeuw et al. [13], this can be roughly estimated by calculating the heats of formation of the following reaction type:

MOx+ y.H2O→ M(OH)x+y

Where M is Ca, Al, and Si.

We have used the five reactions shown in the table below to estimate such a correction for our cementitious systems. Where we have the simulated enthalpy of reaction without correction (Hsim), the experimental value (Hexp), the correction needed for that reaction (Hcorr) and in the final column the absolute value for the correction for one water molecule (Habs). For the energetic validation of the force field, for the reactions containing water splitting we used a correction term of 2.9 eV, with standard deviation of 0.6 eV. (see main text Table 3, for source of experimental value).

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| Reaction | Hsim (eV) | Hexp (eV) | Hdiff (eV) | Hcorr (eV) |
| 1-    Ca(OH)2→  CaO + H2O | 4.07 | 0.66 | 3.14 | 3.14 |
| 2-    Si(OH)4 → SiO2 + 2 H2O | -2.23 | -6.27 | 4.04 | 2.02 |
| 3-    AlOOH +  H2O -→ Al(OH)3. | -2.75 | -0.11 | 2.64 | 2.64 |
| 4-   Al2O3+3H2O → 2Al(OH)3 | -7.80 | -0.46 | 7.34 | 2.45 |
| 5-   3CaO+2Al(OH)3 +3H2O → Ca3Al2(OH)12 | -14.36 | -5.00 | 9.36 | 3.12 |
| 6-   3CaO+2Al(OH)3 → Ca3Al2O6+3H2O | 10.67 | -0.67 | 11.34 | 3.78 |
| Water splitting correction (average) |  |  |  | 2.9 (0.60) \* |

\*The standard deviation for the average correction value.

**S6 Error estimation for reaction enthalpies.**

The error on calculated reaction enthalpies comes from the errors on the calculated energies on the individual structures estimated. With this in mind, we are using the empirical estimation of the force field error developed by Galmarini et al. [3,14]. The exact calculation of the empirical estimation of the force field error, εestFF for a reaction enthalpy ΔH used here can be found in equations E9-11 where Si are the stoichiometry coefficients, Ri the reactants, Pi the products and Hi the enthalpies.

|  |  |
| --- | --- |
|  | (E9) |
|  | (E10) |
|  | (E11) |
|  |  |

In the case of aqueous species, the energy of the aqueous species is estimated based on the energy difference between two boxes of water with the same number of water molecules but with and without the aqueous species. As long as the number of water molecules is large enough, the size of the water box should not have any influence on neither the calculated energy of the aqueous species nor on the error on the calculated energy.

**S7. Details of simulations for structural validation**

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| No | Name | Box size | Ensemble | Equilibration time (ps) | Production time (ps) |
| (unit cells) |
| 1 | Gypsum (Ca.SO4.2H2O) | 3x1x3 | NPT | 260 | 800 |
| 2 | Anhydrate (CaSO4) | 3x3x3 | NPT | 250 | 900 |
| 3 | Lime (CaO) | 3x3x3 | NPT | 500 | 250 |
| 4 | Portlandite (Ca(OH)2) | 8x8x5 | NPT | 500 | 250 |
| 5 | 14 Å tobermorite (Ca5Si6O16(OH)2.26H2O) | 4x4x2 | NPT | 1000 | 500 |
| 6 | Corundum (Al2O3) | 5x5x2 | NPT | 500 | 250 |
| 7 | Alite ((Ca3SiO5)) | 2x3x2 | NPT | 200 | 800 |
| 8 | Gibbsite (Al(OH)3) | 3x4x3 | NPT | 500 | 250 |
| 9 | Millosevichite (Al2(SO4)3) | 3x3x1 | NPT | 210 | 840 |
| 10 | Böhmite (AlO(OH)) | 8x2x8 | NPT | 500 | 250 |
| 11 | Tricalcium aluminate  (Ca3Al2O6) | 2x2x2 | NPT | 500 | 250 |
| 12 | Hydrogarnet (Ca3 Al2 O12 H12) | 2x2x2 | NPT | 500 | 250 |
| 13 | Ettringite (Ca6Al2(SO4)3(OH)12.26H2O) | 2x2x1 | NPT | 220 | 750 |
| 14 | Quartz (SiO2) | 3x3x3 | NPT | 500 | 250 |
| 15 | Sodium Chloride (NaCl) | 4x4x4 | NPT | 220 | 790 |
| 16 | Potassium Chloride (KCl) | 4x4x4 | NPT | 200 | 800 |
| 17 | Sodium Hydroxide (NaOH) | 4x1x4 | NPT | 300 | 1200 |
| 18 | Potassium hydroxide (KOH) | 4x4x3 | NPT | 300 | 1200 |
| 19 | Calcium chloride (CaCl2) | 2x2x3 | NPT | 300 | 1000 |

|  |
| --- |
|  |
| Figure S1. Coordination number versus Ca-O distance for Bonaccorsi’s 14Å tobermorite [15] and the resulting structure form the ERICA FF shown in Figure 6 (d) in the main text. |

**S8. Surface energy calculations and morphology predictions**

The Time step used is 0.7 fs. A cutoff of 8.5 Å was used between short-range interaction and long-range interaction like the work of Galmarini et al. [3]. Like previous parts the total time will be given in equilibration time plus production time. We have used periodic boundary conditions in our simulations. For bulk portlandite, bulk water and portlandite-water box, the NPT ensemble was used. The final simulations were carried out for 200+800 ps where the first 200 ps were used for equilibration of the system and the following 800 ps were used for all calculations.

For the morphology prediction we used Wulffman software [3]. This software uses surface energy per each surface to calculate the final equilibrium morphology. Below figure S2 shows the predicted Wulffman shape of portlandite in water and vacuum.

|  |  |
| --- | --- |
|  |  |
| Predicted morphology of portlandite in vacuum. Color code: yellow: 001, red: 100 green: 101 | Predicted morphology of portlandite in water. Color code: yellow: 001, red: 100 green: 101 |

Figure S2. Predicted Wulffman equilibrium morphologies from ERICA FF1 for portlandite in vacuum and water.

**S9. Elastic Constants**

**Comparison of calculated elastic tensor coefficients for tricalcium silicate from different force fields**

|  |  |  |  |
| --- | --- | --- | --- |
|  | Erica FF2 | IFF [16] | CLAYFF [16] |
| C11 [GPa] | 228.91 | 219.6 | 118.6 |
| C22 [GPa] | 228.45 | 216.0 | 34.81 |
| C33 [GPa] | 206.98 | 189.6 | 95.4 |
| C12 [GPa] | 89.61 | 77.54 | 34.81 |
| C13 [GPa] | 60.14 | 52.79 | 35.98 |
| C23 [GPa] | 58.61 | 52.72 | 27.95 |
| C44 [GPa] | 57.65 | 37.0 | 33.01 |
| C55 [GPa] | 63.79 | 43.4 | 38.22 |
| C66 [GPa] | 66.58 | 67.65 | 32.16 |
| C14 [GPa] | 4.43 | - | - |
| C15 [GPa] | 8.95 | 4.0 | 17.69 |
| C16 [GPa] | 0.87 | - | - |
| C24 [GPa] | -1.32 | - | - |
| C25 [GPa] | 24.0 | -21.7 | 1.39 |
| C26 [GPa] | 2.09 | - | - |
| C34 [GPa] | -0.56 | - | - |
| C35 [GPa] | 1.73 | -34.04 | 6.65 |
| C36 [GPa] | -4.81 | - | - |
| C45 [GPa] | -1.91 | - | - |
| C46 [GPa] | -13.14 | -6.39 | 3.28 |
| C56 [GPa] | -0.64 | - | - |
| Bulk Modulus [GPa] | 120.12 | 104.7 | 53.9 |
| Shear Modulus 1 [GPa] | 62.67 | 54.3 | 33.5 |
| Shear Modulus 2 [GPa] | 75.99 | 54.3 | 33.5 |
| Poisson Ratio [-] | 0.239 | 0.279 | 0.243 |
| Young's Modulus [GPa] | 188-155 | 139 | 83.2 |

**Calculated elastic tensor coefficients for tobermorite 14Å with Erica FF2**

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| C11 [GPa] | C22 [GPa] | C33 [GPa] | C12 [GPa] | C13 [GPa] | C23 [GPa] | C44 [GPa] |
| 99.44 | 130.70 | 60.61 | 46.38 | 13.97 | 19.01 | 26.15 |
|  |  |  |  |  |  |  |
| C55 [GPa] | C66 [GPa] | C14 [GPa] | C15 [GPa] | C16 [GPa] | C24 [GPa] | C25 [GPa] |
| 6.46 | 39.43 | -3.29 | 0.36 | -3.81 | -4.23 | 1.44 |

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