## Supplementary information

Table S1 The enthalpy ($H$) for (CaO)$_4$, CaO(bulk) and nano-CaO

| T     | (1/4)(CaO)$_4$ | 2 nm  | 3 nm  | 4 nm  | 5 nm  | 6 nm  | 7 nm  | 8 nm  | 9 nm  |
|-------|----------------|-------|-------|-------|-------|-------|-------|-------|-------|
| 1000  | 46.669         | 47.271| 48.050| 48.589| 48.961| 49.228| 49.429| 49.584| 49.708|
| 1100  | 51.158         | 51.817| 52.672| 53.263| 53.671| 53.964| 54.184| 54.354| 54.490|
| 1200  | 55.660         | 56.378| 57.308| 57.952| 58.395| 58.715| 58.954| 59.139| 59.287|
| 1300  | 60.173         | 60.950| 61.955| 62.651| 63.131| 63.477| 63.736| 63.936| 64.096|
| 1400  | 64.694         | 65.529| 66.100| 67.359| 67.876| 68.247| 68.526| 68.741| 68.913|
| 1500  | 69.223         | 70.117| 71.274| 72.075| 72.628| 73.025| 73.323| 73.554| 73.738|
| 1600  | 73.757         | 74.709| 75.942| 76.796| 77.385| 77.809| 78.126| 78.372| 78.568|
| 1700  | 78.295         | 79.306| 80.615| 81.522| 82.147| 82.597| 82.934| 83.195| 83.403|
| 1800  | 82.837         | 83.907| 85.292| 86.252| 86.913| 87.389| 87.746| 88.022| 88.242|
| 1873  | 86.473         | 87.590| 89.036| 90.038| 90.728| 91.225| 91.598| 91.886| 92.116|
| 1900  | 87.383         | 88.512| 89.973| 90.985| 91.683| 92.185| 92.562| 92.853| 93.085|
| 2000  | 91.931         | 93.119| 94.656| 95.721| 96.455| 96.984| 97.380| 97.686| 97.930|

| 10 nm | 15 nm | 20 nm | 30 nm | 40 nm | 50 nm | 100 nm | 200 nm | CaO(bulk) |
|-------|-------|-------|-------|-------|-------|--------|--------|----------|
| 1000  | 49.809| 50.120| 50.281| 50.444| 50.527| 50.578 | 50.679 | 50.730 | 50.781 |
| 1100  | 54.601| 54.942| 55.118| 55.298| 55.389| 55.444 | 55.555 | 55.611 | 55.667 |
| 1200  | 59.407| 59.779| 59.971| 60.166| 60.265| 60.325 | 60.446 | 60.507 | 60.568 |
| 1300  | 64.226| 64.628| 64.835| 65.046| 65.154| 65.218 | 65.349 | 65.415 | 65.481 |
| 1400  | 69.053| 69.485| 69.708| 69.935| 70.051| 70.120 | 70.261 | 70.331 | 70.403 |
| 1500  | 73.887| 74.350| 74.588| 74.832| 74.955| 75.029 | 75.180 | 75.256 | 75.332 |
|   T   | \(1/4)(\text{CaO})_4\) | 2 nm  | 3 nm  | 4 nm  | 5 nm  | 6 nm  | 7 nm  | 8 nm  | 9 nm  |
|------|-------------------------|-------|-------|-------|-------|-------|-------|-------|-------|
| 1000 | 148.768                 | 140.355 | 129.464 | 121.920 | 116.721 | 112.978 | 110.171 | 108.003 | 106.273 |
| 1100 | 156.964                 | 148.657 | 137.903 | 130.454 | 125.321 | 121.625 | 118.854 | 116.713 | 115.004 |
| 1200 | 160.576                 | 152.316 | 141.623 | 134.215 | 129.111 | 125.436 | 122.681 | 120.552 | 118.853 |
| 1300 | 163.927                 | 155.710 | 145.073 | 137.705 | 132.628 | 128.972 | 126.231 | 124.114 | 122.423 |
| 1400 | 167.051                 | 158.875 | 148.291 | 140.958 | 135.906 | 132.268 | 129.541 | 127.434 | 125.752 |
| 1500 | 169.977                 | 161.839 | 151.303 | 144.005 | 138.976 | 135.355 | 132.640 | 130.543 | 128.869 |
| 1600 | 172.728                 | 164.626 | 154.136 | 146.870 | 141.863 | 138.258 | 135.555 | 133.467 | 131.800 |
| 1700 | 175.324                 | 167.255 | 156.809 | 149.573 | 144.587 | 140.997 | 138.306 | 136.226 | 134.566 |
| 1800 | 177.301                 | 169.258 | 158.845 | 151.632 | 146.662 | 143.082 | 140.400 | 138.327 | 136.672 |
| 1873 | 177.782                 | 169.745 | 159.340 | 152.133 | 147.166 | 143.590 | 140.909 | 138.838 | 137.185 |
| 1900 | 180.115                 | 172.108 | 161.742 | 154.562 | 149.614 | 146.051 | 143.381 | 141.317 | 139.670 |
| 2000 | 148.768                 | 140.355 | 129.464 | 121.920 | 116.721 | 112.978 | 110.171 | 108.003 | 106.273 |
| 10 nm |                       |       |       |       |       |       |       |       |       |
| 1000 | 104.864                 | 100.511 | 98.262 | 95.973 | 94.812 | 94.110 | 92.696 | 91.983 | 91.264 |
| 1100 | 113.613                 | 109.315 | 107.095 | 104.835 | 103.688 | 102.996 | 101.599 | 100.895 | 100.185 |
| 1200 | 117.470                 | 113.196 | 110.988 | 108.741 | 107.601 | 106.912 | 105.523 | 104.823 | 104.117 |

Table S2 The entropy (S) for (CaO)$_4$, CaO(bulk) and nano-CaO
| T     | (1/4)(CaO)$_{4}$   | 2 nm     | 3 nm     | 4 nm     | 5 nm     | 6 nm     | 7 nm     | 8 nm     | 9 nm     |
|-------|-------------------|----------|----------|----------|----------|----------|----------|----------|----------|
| 1000  | -1976218.206      | -1976258.794 | -1976311.340 | -1976347.739 | -1976372.819 | -1976390.879 | -1976404.418 | -1976414.877 | -1976423.228 |
| 1100  | -1976233.300      | -1976273.050 | -1976324.510 | -1976360.157 | -1976384.719 | -1976402.406 | -1976415.665 | -1976425.908 | -1976434.086 |
| 1200  | -1976248.804      | -1976287.720 | -1976338.102 | -1976373.001 | -1976397.048 | -1976414.365 | -1976427.346 | -1976437.375 | -1976445.381 |
| 1300  | -1976264.683      | -1976302.771 | -1976352.080 | -1976386.237 | -1976409.772 | -1976426.721 | -1976439.425 | -1976449.240 | -1976457.077 |
| 1400  | -1976280.910      | -1976318.174 | -1976366.417 | -1976399.835 | -1976422.861 | -1976439.443 | -1976451.873 | -1976461.476 | -1976469.142 |
| 1500  | -1976297.461      | -1976333.905 | -1976381.087 | -1976413.770 | -1976436.290 | -1976452.507 | -1976464.663 | -1976474.055 | -1976481.553 |
| 1600  | -1976314.314      | -1976349.943 | -1976396.068 | -1976428.020 | -1976450.036 | -1976465.890 | -1976477.774 | -1976486.956 | -1976494.286 |
| 1700  | -1976331.450      | -1976366.267 | -1976411.342 | -1976442.565 | -1976464.079 | -1976479.572 | -1976491.185 | -1976500.157 | -1976507.321 |
| 1800  | -1976348.855      | -1976382.863 | -1976426.891 | -1976457.389 | -1976478.403 | -1976493.536 | -1976504.880 | -1976513.644 | -1976520.641 |
| 1873  | -1976362.960      | -1976396.324 | -1976439.517 | -1976469.438 | -1976490.054 | -1976504.900 | -1976516.029 | -1976524.626 | -1976531.491 |
| 1900  | -1976366.511      | -1976399.714 | -1976442.699 | -1976472.475 | -1976492.992 | -1976507.767 | -1976518.842 | -1976527.398 | -1976534.229 |
| 2000  | -1976384.406      | -1976416.807 | -1976458.754 | -1976487.811 | -1976507.832 | -1976522.249 | -1976533.057 | -1976541.407 | -1976548.073 |

Table S3: The Gibbs free energy (G) for (CaO)$_{4}$, CaO(bulk) and nano-CaO.
|    |      |      |      |      |      |      |      |      |      |      |
|----|------|------|------|------|------|------|------|------|------|------|
|1000| -1976430.025 | -1976451.026 | -1976461.874 | -1976472.916 | -1976478.520 | -1976481.905 | -1976488.729 | -1976492.170 | -1976495.637 |
|1100| -1976440.743 | -1976461.311 | -1976471.934 | -1976482.748 | -1976488.236 | -1976491.551 | -1976498.235 | -1976501.604 | -1976505.000 |
|1200| -1976451.898 | -1976472.035 | -1976482.436 | -1976493.023 | -1976498.396 | -1976501.641 | -1976508.185 | -1976511.483 | -1976514.809 |
|1300| -1976463.455 | -1976483.163 | -1976493.342 | -1976503.704 | -1976508.963 | -1976512.139 | -1976518.544 | -1976521.772 | -1976525.026 |
|1400| -1976475.383 | -1976494.665 | -1976504.624 | -1976514.762 | -1976519.907 | -1976523.014 | -1976529.280 | -1976532.439 | -1976535.623 |
|1500| -1976487.656 | -1976506.514 | -1976516.254 | -1976526.169 | -1976531.201 | -1976534.240 | -1976540.368 | -1976543.457 | -1976546.571 |
|1600| -1976500.253 | -1976518.688 | -1976528.210 | -1976537.903 | -1976542.822 | -1976545.793 | -1976551.784 | -1976554.804 | -1976557.848 |
|1700| -1976513.151 | -1976531.167 | -1976540.472 | -1976549.944 | -1976554.751 | -1976557.654 | -1976563.509 | -1976566.460 | -1976569.435 |
|1800| -1976526.336 | -1976543.933 | -1976553.022 | -1976562.274 | -1976566.969 | -1976569.805 | -1976575.524 | -1976578.406 | -1976581.312 |
|1873| -1976537.078 | -1976554.341 | -1976563.258 | -1976572.335 | -1976576.941 | -1976579.724 | -1976585.334 | -1976588.162 | -1976591.012 |
|1900| -1976539.790 | -1976556.970 | -1976565.844 | -1976574.877 | -1976579.461 | -1976582.230 | -1976587.813 | -1976590.627 | -1976593.464 |
|2000| -1976553.499 | -1976570.264 | -1976578.924 | -1976587.738 | -1976592.212 | -1976594.914 | -1976600.362 | -1976603.108 | -1976605.877 |