*Supplementary Materials*

**Table 1.** Lattice parameters of optimized structures of cassiterite (SnO2). a

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| Functional | Basis sets | GTH pseudopotentials | a (Å) / Error (%) b | b (Å) / Error (%) b | c (Å) / Error (%) b | V (Å3) / Error (%) b |
| LDA | Sn: DZVP-MOLOPT-SR-GTH-q4O: DZVP-MOLOPT-SR-GTH-q6 | Sn: GTH-PADE-q4O: GTH-PADE-q6 | 4.695 / -0.89 | 4.695 / -0.89 | 3.186 / -0.02 | 70.233 / -1.79 |
| PBE | Sn: GTH-PBE-q4O: GTH-PBE-q6 | 4.847 / 2.31 | 4.847 / 2.31 | 3.271 / 2.66 | 76.846 / 7.46 |
| SCAN | Sn: GTH-SCAN-q4O: GTH-SCAN-q6 | 4.616 / -2.57 | 4.616 / -2.57 | 3.104 / -2.60 | 66.120 / -7.54 |
| SCAN(1800Ry) | Sn: GTH-SCAN-q4O: GTH-SCAN-q6 | 4.615 / -2.57 | 4.615 / -2.57 | 3.103 / -2.60 | 66.110 / -7.55 |
| SCAN | Sn: GTH-PBE-q4O: GTH-PBE-q6 | 4.794 / 1.19 | 4.794 / 1.19 | 3.256 / 2.17 | 74.816 / 4.62 |
| TPSS | Sn: GTH-PBE-q4O: GTH-PBE-q6 | 4.810 / 1.53 | 4.810 / 1.53 | 3.262 / 2.37 | 75.474 / 5.54 |
| SCAN | Sn: DZVP-MOLOPT-SCAN-GTH-q4O: DZVP-MOLOPT-SCAN-GTH-q6 | Sn: GTH-SCAN-q4O: GTH-SCAN-q6 | 4.619 / -2.50 | 4.619 / -2.50 | 3.106 / -2.53 | 66.256 / -7.35 |
| SCAN(1800 Ry) | Sn: GTH-SCAN-q4O: GTH-SCAN-q6 | 4.619 / -2.50 | 4.619 / -2.50 | 3.105 / -2.54 | 66.248 / -7.36 |
| SCAN | Sn: GTH-PBE-q4O: GTH-PBE-q6 | 4.796 / 1.23 | 4.796 / 1.23 | 3.263 / 2.39 | 75.042 / 4.94 |
|  |  |  |  |  |  |  |
| LDA | Sn: TZVP-MOLOPT-SR-GTHO: TZVP-MOLOPT-GTH | Sn: GTH-PADE-q4O: GTH-PADE-q6 | 4.696 / -0.88 | 4.696 / -0.88 | 3.188 / 0.06 | 70.301 / -1.69 |
| PBE | Sn: GTH-PBE-q4O: GTH-PBE-q6 | 4.850 / 2.38 | 4.850 / 2.38 | 3.270 / 2.63 | 76.925 / 7.57 |
| SCAN | Sn: GTH-SCAN-q4O: GTH-SCAN-q6 | 4.620 / -2.48 | 4.620 / -2.48 | 3.096 / -2.82 | 66.095 / -7.58 |
| SCAN | Sn: GTH-PBE-q4O: GTH-PBE-q6 | 4.798 / 1.27 | 4.798 / 1.27 | 3.252 / 2.06 | 74.858 / 4.68 |
| TPSS | Sn: GTH-PBE-q4O: GTH-PBE-q6 | 4.810 / 4.55 | 4.810 / 1.55 | 3.262 / 2.37 | 75.495 / 5.57 |
| SCAN | Sn: TZVP-MOLOPT-SCAN-GTH-q4O: TZVP-MOLOPT-SCAN-GTH-q6 | Sn: GTH-SCAN-q4O: GTH-SCAN-q6 | 4.612 / -2.65 | 4.612 / -2.65 | 3.094 / -2.89 | 65.814 / -7.97 |
| SCAN | Sn: GTH-PBE-q4O: GTH-PBE-q6 | 4.790 / 1.12 | 4.790 / 1.12 | 3.253 / 2.10 | 74.655 / 4.39 |
|  |  |  |  |  |  |  |
|  | Experiment c |  | 4.737 | 4.737 | 3.186 | 71.512 |

1. For all structures in this table, α=β=γ=90°.

Except the two calculations marked “1800 Ry”, the cutoff of the electronic density was set to be 1200 Ry.

Sources of parameters:

DZVP-MOLOPT-SR-GTH basis sets (for Sn and O): the *BASIS\_MOLOPT* file;

TZVP-MOLOPT-SR-GTH basis sets (for Sn): the *BASIS\_MOLOPT\_UCL* file;

TZVP-MOLOPT-GTH basis sets (for O): the *BASIS\_MOLOPT* file;

DZVP-/TZVP-MOLOPT-SCAN-GTH basis sets (for Sn and O): the GitHub project page of Prof. Jürg Hutter;

GTH-PADE/-PBE pseudopotentials: the *GTH\_POTENTIALS* file;

GTH-SCAN pseudopotentials: the GitHub project page of Prof. Jürg Hutter.

1. Relative error compared with experimental value.
2. Bolzan et al. (1997, *Acta Cryst. B*), powder neutron diffraction.

**Table 2.** Analytic stress tensors (in bar) derived from the geometry optimizations for cassiterite (SnO2). a

|  |  |  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| Functional | Basis sets | GTH pseudopotentials | xx | xy | xz | yx | yy | yz | zx | zy | zz |
| LDA | Sn: DZVP-MOLOPT-SR-GTH-q4O: DZVP-MOLOPT-SR-GTH-q6 | Sn: GTH-PADE-q4O: GTH-PADE-q6 | -35035.269 | 0.186 | -0.001 | 0.186 | -35035.108 | 0.002 | -0.000 | 0.002 | -24918.377 |
| PBE | Sn: GTH-PBE-q4O: GTH-PBE-q6 | 122346.320 | -0.061 | -0.003 | -0.061 | 122346.345 | 0.005 | -0.003 | 0.005 | 168989.064 |
| SCAN | Sn: GTH-SCAN-q4O: GTH-SCAN-q6 | -127362.451 | -0.001 | 0.003 | -0.001 | -127362.440 | 0.000 | 0.003 | 0.000 | -168448.424 |
| SCAN | Sn: GTH-PBE-q4O: GTH-PBE-q6 | 75602.095 | -10.834 | -0.002 | -10.834 | 75431.819 | 0.004 | -0.002 | 0.004 | 121665.198 |
| SCAN | Sn: DZVP-MOLOPT-SCAN-GTH-q4O: DZVP-MOLOPT-SCAN-GTH-q6 | Sn: GTH-SCAN-q4O: GTH-SCAN-q6 | -124795.927 | 0.002 | 0.000 | 0.002 | -124795.923 | -0.001 | 0.000 | -0.001 | -165665.625 |
| SCAN | Sn: GTH-PBE-q4O: GTH-PBE-q6 | 81162.432 | -8.010 | -0.002 | -8.010 | 81112.937 | -0.000 | -0.002 | -0.000 | 133126.166 |
|  |  |  |  |  |  |  |  |  |  |  |  |
| LDA | Sn: TZVP-MOLOPT-SR-GTHO: TZVP-MOLOPT-GTH | Sn: GTH-PADE-q4O: GTH-PADE-q6 | -32775.545 | -0.010 | -0.000 | -0.010 | -33578.461 | 0.001 | -0.000 | 0.001 | -21029.739 |
| PBE | Sn: GTH-PBE-q4O: GTH-PBE-q6 | 122847.356 | -0.098 | 0.000 | -0.098 | 122847.332 | 0.001 | 0.000 | 0.001 | 167445.239 |
| SCAN | Sn: GTH-SCAN-q4O: GTH-SCAN-q6 | -125514.347 | -0.024 | -0.003 | -0.024 | -125514.346 | -0.001 | -0.003 | -0.001 | -174318.484 |
| SCAN | Sn: GTH-PBE-q4O: GTH-PBE-q6 | 76293.191 | -8.463 | 0.002 | -8.463 | 76204.974 | 0.002 | 0.002 | 0.002 | 117464.709 |
| SCAN | Sn: TZVP-MOLOPT-SCAN-GTH-q4O: TZVP-MOLOPT-SCAN-GTH-q6 | Sn: GTH-SCAN-q4O: GTH-SCAN-q6 | -132783.239 | 28.104 | -0.001 | 28.104 | -132783.235 | 0.000 | -0.001 | 0.000 | -181772.061 |
| SCAN | Sn: GTH-PBE-q4O: GTH-PBE-q6 | 70720.213 | -16.463 | 0.015 | -16.463 | 70720.166 | 0.005 | 0.015 | 0.005 | 114976.384 |

1. The cutoff of the electronic density was set to be 1200 Ry. Sources of parameters are the same with Table 1.

**Table 3.** Lattice parameters of optimized structures of rutile (TiO2). a

|  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- |
| Functional | Basis sets | GTH pseudopotentials | a (Å) / Error (%) b | b (Å) / Error (%) b | c (Å) / Error (%) b | V (Å3) / Error (%) b | Note |
| LDA | Ti: DZVP-MOLOPT-SR-GTH-q12O: DZVP-MOLOPT-SR-GTH-q6 | Ti: GTH-PADE-q12O: GTH-PADE-q6 | 4.549 / -0.95 | 4.549 / -0.95 | 2.935 / -0.75 | 60.732 / -2.62 |  |
| PBE | Ti: GTH-PBE-q12O: GTH-PBE-q6 | 4.650 / 1.26 | 4.650 / 1.26 | 2.997 / 1.33 | 64.797 / 3.90 |  |
| SCAN | Ti: GTH-SCAN-q12O: GTH-SCAN-q6 | 4.594 / 0.05 | 5.594 / 0.05 | 2.968 / 0.37 | 62.658 / 0.47 | γ = 89.8° |
| SCAN(1800Ry) | Ti: GTH-SCAN-q12O: GTH-SCAN-q6 | 4.597 / 0.10 | 4.597 / 0.10 | 2.976 / 0.64 | 62.889 / 0.84 |  |
| SCAN | Ti: GTH-PBE-q12O: GTH-PBE-q6 | 4.587 / -0.10 | 4.587 / -0.10 | 2.965 / 0.27 | 62.406 / 0.06 | γ = 89.9° |
| SCAN | Ti: DZVP-MOLOPT-SCAN-GTH-q12O: DZVP-MOLOPT-SCAN-GTH-q6 | Ti: GTH-SCAN-q12O: GTH-SCAN-q6 | 4.600 / 0.18 | 4.600 / 0.18 | 2.975 / 0.61 | 62.968 / 0.96 | γ = 89.9° |
| SCAN(1800 Ry) | Ti: GTH-SCAN-q12O: GTH-SCAN-q6 | 4.600 / 0.18 | 4.600 / 0.18 | 2.976 / 0.62 | 62.978 / 0.98 |  |
| SCAN | Ti: GTH-PBE-q12O: GTH-PBE-q6 | 4.580 / -0.26 | 4.580 / -0.26 | 2.959 / 0.05 | 62.068 / -0.48 | γ = 89.9° |
|  |  |  |  |  |  |  |  |
| LDA | Ti: TZVP-MOLOPT-SR-GTHO: TZVP-MOLOPT-GTH | Ti: GTH-PADE-q12O: GTH-PADE-q6 | 4.554 / -0.84 | 4.554 / -0.84 | 2.933 / -0.83 | 60.817 / -2.49 |  |
| PBE | Ti: GTH-PBE-q12O: GTH-PBE-q6 | 4.654 / 1.36 | 4.654 / 1.36 | 2.991 / 1.15 | 64.803 / 3.91 |  |
| SCAN | Ti: GTH-SCAN-q12O: GTH-SCAN-q6 | 4.598 / 0.12 | 4.598 / 0.12 | 2.969 / 0.38 | 62.757 / 0.63 | α = 89.9°, γ = 89.9° |
| SCAN | Ti: GTH-PBE-q12O: GTH-PBE-q6 | 4.593 / 0.02 | 4.593 / 0.02 | 2.962 / 0.14 | 62.484 / 0.19 | γ = 89.9° |
| SCAN | Ti: TZVP-MOLOPT-SCAN-GTH-q12O: TZVP-MOLOPT-SCAN-GTH-q6 | Ti: GTH-SCAN-q12O: GTH-SCAN-q6 | 4.599 / 0.14 | 4.599 / 0.14 | 2.965 / 0.24 | 62.694 / 0.52 | γ = 89.8° |
| SCAN | Ti: GTH-PBE-q12O: GTH-PBE-q6 | 4.589 / -0.08 | 4.589 / -0.08 | 2.958 / 0.03 | 62.285 / -0.13 | γ = 89.9° |
|  |  |  |  |  |  |  |  |
|  | Experiment c |  | 4.5922 | 4.5922 | 2.9574 | 62.367 |  |

1. For all structures in this table, if there is no note, α=β=γ=90°.

Except the two calculations marked “1800 Ry”, the cutoff of the electronic density was set to be 1200 Ry.

Sources of parameters:

DZVP-MOLOPT-SR-GTH basis sets (for Ti and O): the *BASIS\_MOLOPT* file;

TZVP-MOLOPT-SR-GTH basis sets (for Ti): the *BASIS\_MOLOPT\_UCL* file;

TZVP-MOLOPT-GTH basis sets (for O): the *BASIS\_MOLOPT* file;

DZVP-/TZVP-MOLOPT-SCAN-GTH basis sets (for Ti and O): the GitHub project page of Prof. Jürg Hutter;

GTH-PADE/-PBE pseudopotentials: the *GTH\_POTENTIALS* file;

GTH-SCAN pseudopotentials: the GitHub project page of Prof. Jürg Hutter.

1. Relative error compared with experimental value.
2. Swope et al. (1995, *Am. Mineral.*), single-crystal X-ray diffraction.