Supplementary Materials: Structural and Electronic Properties of Different Terminations for Quartz (001) Surfaces as Well as Water Molecule Adsorption on It: A First-Principles Study

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Geometry optimization of bulk cell

The first and most important thing is to build a rational quartz bulk cell, which is the first guarantee that all the simulation work is carried out correctly. In this part, convergence tests were conducted to evaluate the dependences of the quartz crystal rationality on the exchange-correlation functionals selection, cutoff energy and the k-point set mesh.

Various exchange-correlation functionals have been raised to describe the exchange and correlation potential, but only one can be the best for a certain structure. The most common four generalised gradient approximation (GGA) functionals and one local-density approximation (LDA) were used to optimize the quartz unit-cell. Based on the suitable exchange-correlation functional, the cutoff energy and k-point set mesh were tested by geometry optimization calculations. The computed lattice constants were contrasted with those experiment observed in X-ray diffraction to determine the suitable functional. We employed the cell parameter deviation as the evaluation criteria, that is, the smaller the cell parameter deviation is, the more suitable the calculation parameter is. And the total energy of calculation was also taken into consideration, that is, the lower the total energy is, the more stable the bulk cell is. The results of these optimizations were shown in table S1, S2 and S3.

Table S1. Lattice constant and total energy of quartz conventional cell by using different exchange correlation (the cutoff energy and k-point were set as 340 eV and 5 × 5 × 4, respectively).

Functions	Lattice parameter/Å				Total energy/eV
	a = b	Δa,b/%	c	Δc/%	
GGA-PBE	5.057	1.69	5.5977	2.35	−2955.57
GGA-RPBE	4.997	0.48	5.486	0.31	−2958.59
GGA-PW91	4.962	0.22	5.473	0.07	−2959.08
GGA-WC	5.029	1.13	5.520	0.93	−2950.71
LDA-CA-PZ	4.886	1.75	5.375	1.72	−2958.56
Text value of Ref.	4.973	-	5.469	-	-

Table S2. Lattice constant of quartz conventional cell by using different cutoff energy (the exchange correlation function and k-point were set as GGA-PW91 and 5 × 5 × 4, respectively).

Cutoff energy/eV	Lattice parameter/Å				Total energy/eV
	a = b	Δa,b/%	c	Δc/%	
300	4.932	0.82	5.530	1.12	−2958.86
320	4.956	0.34	5.486	0.31	−2959.08
340	4.962	0.22	5.473	0.07	−2959.08
360	4.964	0.14	5.470	0.02	−2959.23
380	4.964	0.14	5.470	0.02	−2959.23
400	4.962	0.22	5.475	0.11	−2959.19

Table S3. Lattice constant and band gap of quartz conventional cell by using different k-point (the exchange correlation function and cutoff energy were set as GGA-PW91 and 360 eV, respectively).

k-point	Lattice parameter/Å				Total energy/eV
	a = b	Δa,b/%	c	Δc/%	
3 × 3 × 4	4.962	0.22	5.473	0.07	−2959.23
5 × 5 × 4	4.964	0.14	5.470	0.02	−2959.23
6 × 6 × 6	4.964	0.14	5.471	0.04	−2959.23

It can be seen from table S1 that, the exchange-correlation functional of GGA-PW91 shown to be the most suitable one with the smallest cell parameter deviation as 0.22% and 0.07%, and the lowest total energy as −2959.08 eV at the same time. In table S2, the cell parameter deviation further reaches the lowest when the cutoff energy increases from 340 eV to 360 eV, and keeps invariant along with the cutoff energy further increasing to 380 and 400 eV, and the total energy is also reaches the lowest as −2959.23 eV. Therefore, the cutoff energy of 360 eV is suggested to be sufficient for this system. From table S3, for the same assessment with cutoff energy, the k-point of 5 × 5 × 4 is also suggested to be sufficient.

Above all, after geometry optimization, the exchange correlation functional chosen was GGA-PW91, and cutoff energy was set at 360 eV, and the special of a 5 × 5 × 4 grid was chosen for all structure calculations. The quartz cell parameters obtained after final minimization were a = b = 4.964 Å, c = 5.470 Å, with the deviation of 0.14% and 0.02%, respectively, compared with experiment values. Which all shows that the calculation conditions chosen are sufficient for the system, and the method is credible for the subsequent calculations.