Supporting Information

Robust Photocatalyst of Monoclinic Ga₂O₃ (100) Surface for Water Splitting

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On the basis of Lewis Acid-Base theory, seven initial fundamental models were given in Fig. 4. To take into account the damaged symmetry of the deficient surfaces, there are sixteen asymmetric positions were labeled in **Fig. S1**. The defective position is marked with (1). In the defective surface, the initial adsorption structures were modeled according to **Fig. 4** and the superficial atomic types shown in Fig S1. The name of the modeled adsorption structure depends on not only the lab of **Fig. S1** but also the subhead in **Fig. 4**. For instance, the name of water molecules adsorbed at the top position of the defect location (marked as position (1)) of S-doped surface with a structure of Fig. 4 (g) can be called as **(1)g**. Meanwhile, the **(1)(4)a** means water adsorbed between the S atom (position (1)) and Ga₂ atom (position (4)) modeled as **Fig. (4)** a. The initial structures near the deficient position are listed as **(1)g**, **(1)(2)b**, **(1)(4)a**, **(1)(7)a**, **(1)(2)(4)d** and **(1)(4)(6)d**, respectively.



Fig. S1. The top view of defective β -Ga₂O₃ (100) surface marked with asymmetric positions. The defective site is represented by yellow sphere in the figure.

The calculated adsorption energy (in eV) for water adsorption near the deficient site and the most stable case are symmetrized in Table S1. Beyond S-doped surface, the most stable structures are away from defective position as compared in Table S1.

Table S1. The data of adsorption energy (in eV) for water adsorption near the deficient site and the most stable case (marked by red front).

| V ₀₃ | Туре | (1)c | 1(g) | (1)(4)(6)d | (1)(2)(4)d | (2)g | (4)f | (13)(11)(14)d |
|-----------------|--------------------------|--------|---------|------------|------------|------------|------------|---------------|
| | $\Delta E_{ads.}^{H_2O}$ | -0.167 | -0.157 | -0.262 | -0.372 | -0.066 | -0.387 | -0.459 |
| S-doped | Туре | (1)g | (1)(2)b | (1)(4)a | (1)(7)a | (1)(2)(4)d | (1)(4)(6)d | (3)(4)(6)d |
| | $\Delta E_{ads.}^{H_2O}$ | -0.250 | -0.069 | -0.341 | -0.187 | -0.240 | -0.349 | -0.484 |
| Se-doped | Туре | (1)g | (1)(2)b | (1)(4)a | (1)(7)a | (1)(2)(4)d | (1)(4)(6)d | (10)(11)(14)d |
| | $\Delta E_{ads.}^{H_2O}$ | -0.223 | -0.104 | -0.327 | -0.146 | -0.278 | -0.310 | -0.470 |
| Te-doped | Туре | (1)g | (1)(2)b | (1)(4)a | (1)(7)a | (1)(2)(4)d | (1)(4)(6)d | (13)(11)(14)d |
| | $\Delta E_{ads.}^{H_2O}$ | -0.214 | -0.055 | -0.341 | -0.168 | -0.244 | -0.133 | -0.502 |

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- § Electronic supplementary information (ESI) available.

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