Supplementary Information

Theoretical Roadmap for Best Oxygen Reduction Activity in Two-dimensional Transition Metal Tellurides

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This file includes:
- Methods
- Geometric and Electronic Properties (Figure S1-2)
- Origin of $O_2$ activation (Figure S3-11)
- ORR catalytic performance and linear scaling (Figure S12-18)
- Rational screening of activity descriptor (Figure S19-27)
- Supplementary Tables (Table S1-7)
- References
Methods

Computational details

Density functional theory computations were performed by using the Vienna \textit{Ab initio} simulation package (VASP) within the projected augmented wave method.\textsuperscript{1, 2} The Brillouin zone was sampled using k-points of $4 \times 4 \times 1$ for the supercell with $a \times b$ of around 10 Å × 10 Å.\textsuperscript{3} For some monolayers with $a \times b$ of around 20 Å × 10 Å, such as TaPd3Te16, the k-points were sampled using $2 \times 4 \times 1$. The $p$-band center ($\epsilon_p$) of the Te sites was calculated by

$$\epsilon_p = \frac{\int_{-\infty}^{0} \epsilon \rho_p(\epsilon) d\epsilon}{\int_{-\infty}^{0} \rho_p(\epsilon) d\epsilon}$$

(1)

where $\rho_p(\epsilon)$ is the $p$-band density of states of a Te atom and $\epsilon$ is the energy of electrons.

ORR Reactions

The 4e$^-$ dissociative and associative mechanism pathways for ORR are generally the following equations (2) and (3), respectively.\textsuperscript{4} Here, * denotes an adsorption site of the catalyst.

$$
\begin{align*}
O_2 & \rightarrow O_2^\ddagger \rightarrow 2O^\ddagger \rightarrow O^\ddagger + OH^\ddagger \rightarrow O^\ddagger + H_2O \rightarrow OH^\ddagger + H_2O \rightarrow 2H_2O \\
O_2 & \rightarrow O_2^\ddagger \rightarrow OOH^\ddagger \rightarrow O^\ddagger + H_2O \rightarrow OH^\ddagger + H_2O \rightarrow 2H_2O
\end{align*}
$$

(2)

(3)

The free energy of each species can be expressed using the equation:

$$
G = E_{DFT} + E_{ZPE} + \int C_p dT - TS
$$

(4)

where $E_{DFT}$, $E_{ZPE}$ and $S$ are the ground state electronic energy, zero-point vibrational energy and entropy, respectively. The values of $E_{ZPE}$ and $S$ of intermediates are obtained from frequency calculations and the values for molecules are taken from the NIST database (Computational Chemistry Comparison and Benchmark Database), which are listed in Table S6. $T$ is the temperature of 298.15 K. Solvent effects have been considered by using the Poisson-Boltzmann implicit solvation model.\textsuperscript{5} The dielectric constant $\varepsilon$ is taken as 80 for water. The absorption free energy for the adsorbates can be calculated using the equation:

$$
\Delta G_{ads} = \Delta E_{ads} + \Delta E_{ZPE} - T\Delta S
$$

(5)

where $\Delta G_{ads}$ is the adsorption free energy of adsorbates, and $\Delta E_{ZPE}$ and $\Delta S$ are the difference of $E_{ZPE}$ and $S$, respectively. The free energy of $O_2$ was computed by the equation:

$$
G_{O_2} = 2G_{H_2O} - 2G_{H_2} - 4.92 \text{ eV}
$$

(6)

The calculation of energy in volcano plots

The free energies for the four steps in dissociative mechanism can be defined as:

$$
\Delta G_1 = G_{(O+OH)^\ddagger} - G_{(O+O)^\ddagger} - G_{(H^\ddagger+e^-)} = \Delta G_{(O+OH)^\ddagger} - \Delta G_{(O+O)^\ddagger},
$$

(7)

$$
\Delta G_2 = G_{O^\ddagger} + G_{H_2O} - G_{(O+OH)^\ddagger} - G_{(H^\ddagger+e^-)} = \Delta G_{O^\ddagger} - \Delta G_{(O+OH)^\ddagger},
$$

(8)

$$
\Delta G_3 = G_{OH^\ddagger} - G_{O^\ddagger} - G_{(H^\ddagger+e^-)} = \Delta G_{OH^\ddagger} - \Delta G_{O^\ddagger},
$$

(9)

$$
\Delta G_4 = G^\ddagger + G_{H_2O} - G_{O^\ddagger} - G_{(H^\ddagger+e^-)} = -\Delta G_{OH^\ddagger},
$$

(10)

The relations between OH and other adsorbates can be used to define the limiting potentials in dissociative mechanism as a function of the OH adsorption energy as:

$$
U_{L1} = -\Delta G_1 = -0.2 \Delta G_{OH^\ddagger} + 1.6
$$

(11)

$$
U_{L2} = -\Delta G_2 = 1.7 \Delta G_{OH^\ddagger} - 0.6
$$

(12)

$$
U_{L3} = -\Delta G_3 = -1.2 \Delta G_{OH^\ddagger} + 2.1
$$

(13)

$$
U_{L4} = -\Delta G_4 = \Delta G_{OH^\ddagger}
$$

(14)

The free energies for the four steps in associative mechanism can be defined as:
The relations between OH and other adsorbates can be used to define the limiting potentials in an associative mechanism as a function of the OH adsorption energy as:

\[
\Delta G_1 = G_{OOH} - G_O - G_{(H^+e^-)} = \Delta G_{OOH} - 4.92 \\
\Delta G_2 = G_{O^*} + G_{H_2O} - G_{OOH} - G_{(H^+e^-)} = \Delta G_{O^*} - \Delta G_{OOH} \\
\Delta G_3 = G_{OH^*} - G_O - G_{(H^+e^-)} = \Delta G_{OH^*} - \Delta G_O \\
\Delta G_4 = G_e + G_{H_2O} - G_{OH^*} - G_{(H^+e^-)} = -\Delta G_{OH^*}
\]

The above equations result in four limiting potential lines for the four elementary steps, which could form the volcano plot.

\[
U_{L1} = -\Delta G_1 = - \Delta G_{OH^*} + 1.6 \\
U_{L2} = -\Delta G_2 = 1.2 \Delta G_{OH^*} + 1.2 \\
U_{L3} = -\Delta G_3 = -1.2 \Delta G_{OH^*} + 2.1 \\
U_{L4} = -\Delta G_4 = \Delta G_{OH^*}
\]
Geometric and Electronic Properties

Figure S1. The calculated cleavage energies of studied 9 monolayer tellurides.

Figure S2. Band structure and density of states (DOS) of monolayer (a) NbTe₂, (b) WTe₂, (c) NbNiTe₂, (d) TaCo₂Te₂, (e) TaNi₂Te₃, (f) NbIrTe₄, (g) NbPdTe₅, (h) TaPd₃Te₅, and (i) Ta₃Pd₃Te₁₄. NbIrTe₄ is semi-metallic with zero band-gap and others are metallic.
**Origin of O$_2$ activation**

The O$_2$ adsorption configurations, energy barriers of O$_2$ dissociation, and the analysis of underlying O$_2$ adsorption mechanism are shown in this part.

**Figure S3.** The O$_2$ adsorption configurations of (a) NbTe$_2$, (b) WTe$_2$, (c) NbNiTe$_2$, (d) TaCo$_2$Te$_2$, (e) TaNi$_2$Te$_3$, (f) NbIrTe$_4$, (g) NbPdTe$_5$, (h) Ta$_2$Pd$_3$Te$_5$, and (i) Ta$_3$Pd$_3$Te$_{14}$.

**Figure S4.** The energy barriers of O$_2$ dissociation in monolayer (a) NbTe$_2$ and (b) NbIrTe$_4$ from NEB calculations.

**Figure S5.** The O$_2$ adsorption energy of studied tellurides versus charge transfer. The Te active sites are Te1 atoms that mainly interact with O$_2$ with stronger bonding strength. The previous studied tellurides PtTe, NiTe, TaTe$_2$, and Ni$_2$SbTe$_2$ have also been considered for comparison. Notably, the O$_2$ adsorption energy of NiTe was recalculated in this study and the details of the calculation of NiTe are shown in Table S7.
Figure S6. pDOS of Te1 and Te2 atoms in NbIrTe₄ (a) from -7.2 to -6.2 eV and (b) from -0.2 to 0 eV. (c) DOS of Te and O elements with orbitals of O₂ marked on the top and (d) integrated electrons of the difference of pDOS of Te1 and Te2 between NbIrTe₄ and O₂-adsorpted NbIrTe₄. Notably, the orbitals here are O₂ that has adsorbed on NbIrTe₄. (e) Differential charge density distributions of O₂ adsorbed NbIrTe₄.
Figure S7. (a) pDOS of Te1 and O atoms in O₂ adsorbed PtTe and (b) Te atoms in PtTe. (c) DOS of Te and O elements with orbitals of O₂ marked on the top (d) integrated electrons of the difference of pDOS between PtTe and O₂ adsorbed PtTe. Notably, the orbitals here are O₂ that has adsorbed on PtTe. (e) Differential Charge density distributions of O₂ on PtTe monolayer.
Figure S8. (a) pDOS of Te1 and O atoms in O$_2$ adsorbed NbTe$_2$ and (b) Te atoms in NbTe$_2$. (c) DOS of Te and O elements with orbitals of O$_2$ marked on the top (d) integrated electrons of the difference of pDOS between NbTe$_2$ and O$_2$ adsorbed NbTe$_2$. Notably, the orbitals here are O$_2$ that has adsorbed on NbTe$_2$. (e) Differential Charge density distributions of O$_2$ on NbTe$_2$ monolayer.
Figure S9. (a) pDOS of Te1, Te2 and O atoms in O2-adsorbed Ta3Pd3Te14 and (b) Ta3Pd3Te14. (c) DOS of Te and O elements with orbitals of O2 marked on the top (d) integrated electrons of the difference of pDOS between Ta3Pd3Te14 and O2-adsorbed Ta3Pd3Te14. Notably, the orbitals here are O2 that has adsorbed on Ta3Pd3Te14. (e) Differential Charge density distributions of O2 on Ta3Pd3Te14 monolayer.
Figure S10. Schematic representation for O$_2$ molecule and calculated DOS and pCOHP curves.

Figure S11. (a) Schematic representation of π interactions and (b) the $5\tilde{\sigma}$, $1\tilde{\pi}_p$, $\tilde{\rho}_{1\pi}$, $\tilde{\rho}_{2\pi^*}$, and $2\tilde{\pi}_p^*$ orbitals for O$_2$ adsorbed on NbIrTe$_4$. Notably, $5\tilde{\sigma}$ contributes to the “donation” and $\tilde{\rho}_{2\pi^*}$ and $2\tilde{\pi}_p^*$ contributes to “back-donation” process.
**ORR catalytic performance and linear scaling**

The configurations and the free energy diagrams for monolayer NbTe$_2$, NbIrTe$_4$, and Ta$_3$Pd$_3$Te$_{14}$, the structural, electronic, and catalytic properties of NbIrTe$_4$ multilayers, and the relationship between $\Delta G_{O^*}$ and $\Delta G_{OH^*}$ are shown in this part.

**Figure S12.** (a) Scheme of the possible two ORR reaction pathways and (b) free energy diagram for the associative ORR pathway on monolayer NbTe$_2$.

**Figure S13.** (a) Scheme of the possible two ORR reaction pathways monolayer Ta$_3$Pd$_3$Te$_{14}$, and free energy diagrams for the associative ORR pathway on monolayer (b) NbIrTe$_4$ and (c) Ta$_3$Pd$_3$Te$_{14}$ at $U=0$ V.
Figure S14. The structures of (a) bilayer and (b) trilayer NbIrTe₄.

Figure S15. Band structure and density of states (DOS) of (a) bilayer and (b) trilayer NbIrTe₄.

Figure S16. Free energy diagrams for the dissociative ORR pathway on (a) bilayer and (b) trilayer NbIrTe₄.
Figure S17. (a) The Variation of $\Delta G_{0^*}$ versus $\Delta G_{OH^*}$ in studied 9 tellurides. (b) Variation of $\Delta G_{OOH^*}$ as a function of $\Delta G_{OH^*}$ in newly found promising telluride catalysts. We here determined the limiting potentials: $U_L = \min\{-\Delta G_1, -\Delta G_2, -\Delta G_3, -\Delta G_4\}$, where $\Delta G_1, \Delta G_2, \Delta G_3$, and $\Delta G_4$ are the free energy of four reaction steps. With the scaling relations between OH* and other adsorbates, the limiting potential of dissociation pathway can be given by equation:

$$U_L = \min\{-0.2 \Delta G_{OH^*} + 1.2, 1.9 \Delta G_{OH^*} - 0.9, -1.3 \Delta G_{OH^*} + 2.2\}$$

Thus the estimated highest limiting potentials for dissociation pathway is 0.94 V.

Figure S18. O$_2$ adsorption energy versus other considered descriptors of (a) $p$ band center, (b) $\epsilon_{px} / 2 + \epsilon_{py} / 2 + \epsilon_{pz} / 2$, (c) $\epsilon_{px} / 2 + \epsilon_{px} / 2$, and (d) $\epsilon_{py} / 2 + \epsilon_{pz} / 2$, where $\epsilon_{px}$, $\epsilon_{py}$, and $\epsilon_{pz}$ defines centroid of the projected density of states of the $p_x$, $p_y$, and $p_z$ orbitals relative to the Fermi level. The correlation coefficient $R^2$ of other descriptors are smaller than that of $\epsilon_{pz}$. 
Rational screening of activity descriptor
The calculated cleavage energies and ORR pathways of newly studied monolayer tellurides, and the analysis of the adsorption energies of oxygen-containing groups are shown here.

Figure S19. (a) The calculated cleavage energies of newly studied monolayer tellurides. (b) The calculated phase diagram of Nb-Rh-Te system.

Figure S20. Free energy diagrams for the dissociative ORR pathway on monolayer (a) NbRhTe₄, (b) TaRhTe₄, and (c) TaIrTe₄.

Figure S21. (a) Scheme of the dissociation reaction pathway and (b) free energy diagrams for the dissociative ORR pathway on monolayer TaPd₃Te₁₆.
Figure S22. Free energy diagrams for the associative ORR pathway on monolayer (a) NbRhTe₄, (b) TaRhTe₄, (c) TaIrTe₄, and (d) Ta₄Pd₃Te₁₆.

Figure S23. Free energy diagrams for the dissociative ORR pathway on (a) bilayer and (b) trilayer NbRhTe₄.
Figure S24. The structures of (a) M$_3$XTe$_8$(I) and (b) M$_3$XTe$_8$(II) and the phase diagrams of (c) Nb-Ir-Te, (d) Ta-Rh-Te, and (e) Ta-Ir-Te systems. The calculated formation energies of the bulk phases of hypothetic Nb$_3$RhTe$_8$, Nb$_3$IrTe$_8$, Ta$_3$RhTe$_8$, Ta$_3$IrTe$_8$ were 20, 24, 20, and 25 meV/atom, respectively. The phase diagram of Nb-Rh-Te system was shown in Figure S19.

Figure S25. Free energy diagrams for the dissociative ORR pathway on monolayer (a) Nb$_3$RhTe$_8$(I), (c) Ta$_3$RhTe$_8$(I) and (d) Ta$_3$IrTe$_8$(I) and the associative ORR pathway on monolayer (b) Nb$_3$IrTe$_8$(II).
Figure S26. Free energy diagrams for the associative ORR pathway on monolayer (a) Nb$_3$RhTe$_8$(I), (c) Ta$_3$RhTe$_8$(I) and (d) Ta$_3$IrTe$_8$(I) and the dissociative pathway on monolayer (b) Nb$_3$IrTe$_8$(II).

Figure S27. (a) $O_2$ adsorption energy versus the average $p_z$-band center of Te atoms for initially studied tellurides. Notably, the $\Delta G_{O_2}$ of NiTe was recalculated in this study and the details of the calculation are shown in Table S7. The $\Delta G_{O_2}$ of NiTe can’t accord with the linear relationship, which might attract more attention in subsequent study. (b) The $\Delta G_O$ versus the $p_z$-band center of active Te sites for all studied tellurides. (c) Variation of $\Delta G_{\text{OOH}^*}$ as a function of $\Delta G_{\text{OH}^*}$ for monolayer tellurides. (d) Catalytic activity of the materials as a function of intermediates formation free energies $\Delta G_{\text{O}^*} - \Delta G_{\text{OH}^*}$ and $\Delta G_{\text{OH}^*}$. The circle, star, triangle, and square points represent initially-studied tellurides, newly-studied tellurides, M-doped variants, and known catalysts in other studies, respectively.
Table S1. Bader charge calculation results of charge transfer in monolayer tellurides.

|       | M (e) | X (e) | Te1 (e) | Te2 (e) | Te3 (e) | Te4 (e) | Te5 (e) | Te6 (e) | Te7 (e) | Te8 (e) |
|-------|-------|-------|---------|---------|---------|---------|---------|---------|---------|---------|
| NbTe2 | -0.95 | 0.48  | 0.5     | 0.5     |         |         |         |         |         |         |
| WTe2  | -0.48 | 0.28  | 0.19    |         |         |         |         |         |         |         |
| NbNiTe2 | -1.03 | 0.29  | 0.36    | 0.37    |         |         |         |         |         |         |
| TaCo2Te3 | -1.12 | 0.4   | 0.24    | 0.02    | 0.12    |         |         |         |         |         |
| TaNi2Te3 | -1.15 | X1 0.1| 0.32    | 0.24    | 0.21    | X2 0.26 |         |         |         |         |
| NbIrTe4 | -1.15 | 0.58  | 0.5     | 0.07    | -0.29   | 0.28    |         |         |         |         |
| NbPdTe5 | -1.14 | 0.27  | 0.28    | 0.21    | 0.02    | 0.32    | 0.02    |         |         |         |
| Ta3Pd3Te14 | -1.22 | 0.43  | 0.24    | 0.23    | 0.23    | 0.26    | 0.10    | 0.37    | 0.01    | 0.31    |
| TaIrTe4 | -1.15 | 0.26  | 0.10    | 0.37    | 0.01    | 0.31    | 0.07    | 0.23    | 0.25    |         |
| MoTe2  | -0.44 | 0.25  | 0.17    |         |         |         |         |         |         |         |
| TaNiTe2 | -1.01 | 0.31  | 0.34    | 0.35    |         |         |         |         |         |         |
| Ta4Pd3Te16 | -1.15 | 0.29  | 0.47    | 0.18    | 0.21    | 0.28    | 0.05    | 0.31    | -0.03   | 0.35    |
| NbRhTe4 | -1.13 | 0.38  | 0.49    | 0.14    | -0.24   | 0.33    |         |         |         |         |
| TaRhTe4 | -1.12 | 0.38  | 0.47    | 0.15    | -0.24   | 0.32    |         |         |         |         |
| TaIrTe4 | -1.14 | 0.60  | 0.48    | 0.07    | -0.30   | 0.27    |         |         |         |         |
| Ta2Ni3Te5 | -1.15 | 0.22  | 0.38    | 0.26    | 0.33    |         |         |         |         |         |

Table S2. Bader charge results of charge transfer and bond length of O2-adsorbed tellurides.

| Charge       | Te1 (e) | Te2 (e) | O1 (e) | O2 (e) |
|--------------|---------|---------|--------|--------|
| NbTe2        | -0.42   | -0.41   | 0.40   | 0.39   |
| NbIrTe4      | -0.56   | Te2 (A) -0.15 | 0.44 | 0.43   |
| Te2 (B) -0.14 |         |         |        |        |
| Ta3Pd3Te14   | Te1 (A) -0.35 | -0.33 | 0.44   | 0.43   |
| Te1 (B) -0.28 |         |         |        |        |
| Ta4Pd3Te16   | -0.53   | Te2 (A) -0.28 | 0.52 | 0.31   |
| Te2 (B) -0.12 |         |         |        |        |
| NbRhTe4      | -0.54   | Te2 (A) -0.25 | 0.46 | 0.38   |
| Te2 (B) -0.18 |         |         |        |        |
| TaRhTe4      | -0.55   | Te2 (A) -0.20 | 0.38 | 0.47   |
| Te2 (B) -0.18 |         |         |        |        |
| TaIrTe4      | -0.57   | Te2 (A) -0.20 | 0.38 | 0.49   |
| Bond length  | Te1-O (Å) | Te2-O (Å) | O1-O2 (Å) |
|--------------|-----------|-----------|------------|
| NbTe2        | 2.22      | 2.28      | 1.38       |
| NbIrTe4      | 2.13      | 2.56      | 1.39       |
| Ta3Pd3Te14   | 2.35      | 2.25      | 1.37       |
| Ta4Pd3Te16   | 2.15      | 2.32      | 1.39       |
| NbRhTe4      | 2.14      | 2.43      | 1.38       |
| TaRhTe4      | 2.14      | 2.49      | 1.39       |
| TaIrTe4      | 2.13      | 2.51      | 1.39       |
| Monolayer        | ΔG1 (eV) | ΔG2 (eV) | ΔG3 (eV) | ΔG4 (eV) | Limiting potential (V) | Overpotential (V) |
|------------------|----------|----------|----------|----------|-------------------------|-------------------|
| NbTe2            | -1.37    | -0.63    | -1.11    | -0.80    | 0.63                    | 0.60              |
| NbIrTe4          | -1.25    | -1.24    | -0.88    | -1.04    | 0.88                    | 0.35              |
| Ta3Pd3Te14       | -1.38    | -1.17    | -0.80    | -1.08    | 0.80                    | 0.43              |
| NbRhTe4          | -1.21    | -1.17    | -0.98    | -0.96    | 0.96                    | 0.27              |
| TaRhTe4          | -1.26    | -1.22    | -0.84    | -1.12    | 0.84                    | 0.39              |
| TaIrTe4          | -1.32    | -1.25    | -0.78    | -1.16    | 0.78                    | 0.45              |
| Ta4Pd3Te16       | -1.48    | -0.78    | -1.00    | -0.99    | 0.78                    | 0.45              |
| Nb3RhTe8(I)      | -1.49    | -0.86    | -1.22    | -0.82    | 0.82                    | 0.41              |
| Nb3RhTe8(II)     | -0.96    | -0.78    | -0.92    | -0.83    | 0.78                    | 0.45              |
| Nb3IrTe8(I)      | -1.55    | -0.79    | -1.18    | -0.79    | 0.79                    | 0.44              |
| Nb3IrTe8(II)     | -1.37    | -0.64    | -1.15    | -0.81    | 0.64                    | 0.59              |
| Ta3RhTe8(I)      | -1.52    | -0.96    | -1.13    | -0.90    | 0.90                    | 0.33              |
| Ta3RhTe8(II)     | -1.47    | -0.78    | -1.25    | -0.81    | 0.78                    | 0.45              |
| Ta3IrTe8(I)      | -1.53    | -0.90    | -1.05    | -0.93    | 0.90                    | 0.33              |
| Ta3IrTe8(II)     | -1.28    | -0.77    | -1.07    | -0.88    | 0.77                    | 0.46              |
Table S5. The free energy of four reaction steps, the limiting potential and corresponding overpotential in association pathway

| Monolayer        | ΔG1 (eV) | ΔG2 (eV) | ΔG3 (eV) | ΔG4 (eV) | Limiting potential (V) | Overpotential (V) |
|------------------|----------|----------|----------|----------|-------------------------|------------------|
| NbTe2            | -0.80    | -2.21    | -1.11    | -0.80    | 0.80                    | 0.43             |
| NbIrTe4          | -0.65    | -2.35    | -0.88    | -1.04    | 0.65                    | 0.58             |
| Ta3Pd3Te14       | -0.45    | -2.59    | -0.80    | -1.08    | 0.45                    | 0.78             |
| NbRhTe4          | -0.67    | -2.31    | -0.98    | -0.96    | 0.67                    | 0.56             |
| TaRhTe4          | -0.45    | -2.51    | -0.84    | -1.12    | 0.45                    | 0.78             |
| TaIrTe4          | -0.52    | -2.46    | -0.78    | -1.16    | 0.52                    | 0.71             |
| Ta3Pd3Te16       | -0.49    | -2.44    | -1.00    | -0.99    | 0.49                    | 0.74             |
| Nb3RhTe8(I)      | -0.77    | -2.10    | -1.22    | -0.82    | 0.77                    | 0.46             |
| Nb3RhTe8(II)     | -0.73    | -2.44    | -0.92    | -0.83    | 0.73                    | 0.50             |
| Nb3IrTe8(I)      | -0.98    | -1.97    | -1.18    | -0.79    | 0.79                    | 0.44             |
| Nb3IrTe8(II)     | -0.83    | -2.13    | -1.15    | -0.81    | 0.81                    | 0.42             |
| Ta3RhTe8(I)      | -0.70    | -2.19    | -1.13    | -0.90    | 0.70                    | 0.53             |
| Ta3RhTe8(II)     | -0.79    | -2.07    | -1.25    | -0.81    | 0.79                    | 0.44             |
| Ta3IrTe8(II)     | -0.74    | -2.20    | -1.05    | -0.93    | 0.74                    | 0.49             |
| Ta3IrTe8(II)     | -0.73    | -2.24    | -1.07    | -0.88    | 0.73                    | 0.50             |

Table S6. Zero-point energy correction (ΔZPE), entropy contribution (TAS), and total free energy correction (ΔE_{ZPE} - TAS) used in the calculations.

| Species   | ΔE_{ZPE} (eV) | TAS (eV) | ΔE_{ZPE} - TAS (eV) |
|-----------|---------------|----------|--------------------|
| H2        | 0.27          | 0.40     | -0.13              |
| H2O       | 0.56          | 0.67     | -0.11              |
| O2*       | 0.12          | 0.16     | -0.04              |
| O*        | 0.06          | 0.08     | -0.02              |
| OH*       | 0.33          | 0.13     | 0.21               |
| OOH*      | 0.43          | 0.21     | 0.22               |

Table S7. The lattice parameters of bulk NiTe in R-3m phase and the adsorption energy of O2 on monolayer NiTe in different calculations. The results of calculation in solvent model have been used in the above analysis.

| Lattice parameters (Å) |
|------------------------|
| Experimental result    | a=b=c=7.04 |
| Theoretical result     | a=b=c=7.06 |
| Theoretical result (U=6 eV) | a=b=c=6.93 |

| Adsorption energy of O2 (eV) |
|-----------------------------|
| Monolayer NiTe              | 0.13         |
| Monolayer NiTe ( solvent model) | 0.01         |
| Monolayer NiTe ( solvent model & vdW & U=6 eV ) | -0.3         |
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