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Non-Metal Doped VTe_2 Monolayer: Theoretical Insights into the Enhanced Mechanism for Hydrogen Evolution Reaction

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Description of SISSO Method Settings and Definition of R^2

The Sure Independence Screening and Sparsifying Operator (SISSO) method was employed to develop an effective composite descriptor for predicting the hydrogen evolution reaction (HER) activity of NM-VTe_2 structures. In this study, the SISSO algorithm was configured with a feature space constructed from primary features such as geometric parameters (e.g., bond lengths $L_{\text{NM-M}}$) and electronic properties (e.g., pz band centers ε_{pz}). The descriptor dimension was set to 1, and the maximum complexity of the descriptor was limited to 4 to maintain interpretability. Algebraic operations including addition, subtraction, multiplication, and division were used to combine the primary features into composite descriptors.

The performance of the developed descriptor was evaluated using the coefficient of determination, R^2, which quantifies the proportion of variance in the dependent variable (ΔG_{H^*}) that is predictable from the independent variable (the composite descriptor j). R^2 is defined as:

$$R^2 = 1 - \frac{\sum_{i=1}^{n} (y_i - y_i')^2}{\sum_{i=1}^{n} (y_i - \bar{y}_i)^2}$$

where y_i is the actual value of ΔG_{H^*}, y_i' is the predicted value from the SISSO model, $\bar{y}_i$ is the mean of the actual values, and n is the number of data points. An R^2 value close to 1 indicates a strong correlation between the descriptor and the target property, confirming the effectiveness of the descriptor in predicting HER activity.

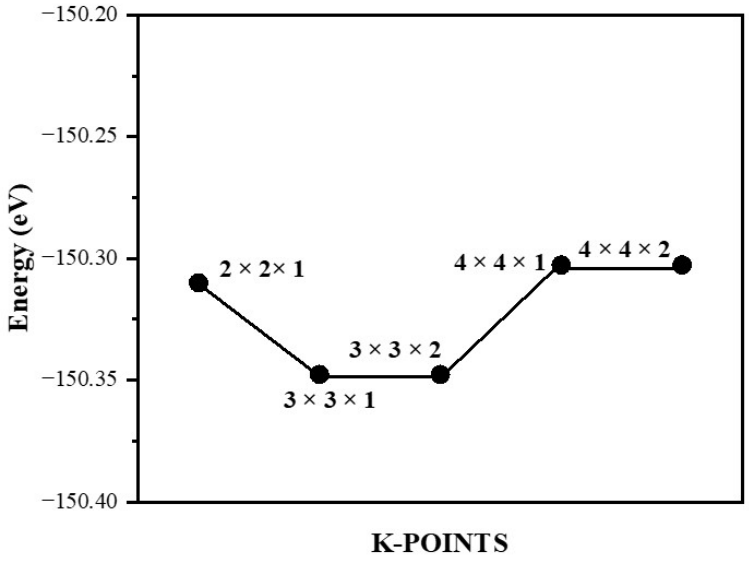

Fig. S1. The energies of VTe_2 monolayer calculated with different k-points.

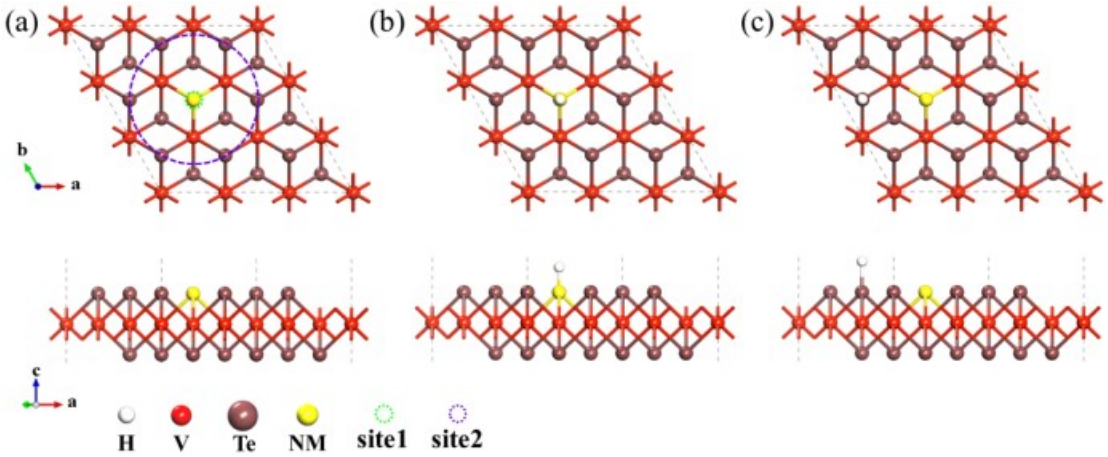

Figure S2. Schematic diagrams of different hydrogen adsorption sites on NM-VTe_2. (a) Illustration of adsorption sites site1 and site2 on the NM-VTe_2 surface. Site1 corresponds to the location of the non-metal dopant (green dashed line), and site2 corresponds to the adjacent Te atom near the dopant (purple dashed box). (b) Structure depicting hydrogen adsorption at site1. (c) Structure depicting hydrogen adsorption at site2.

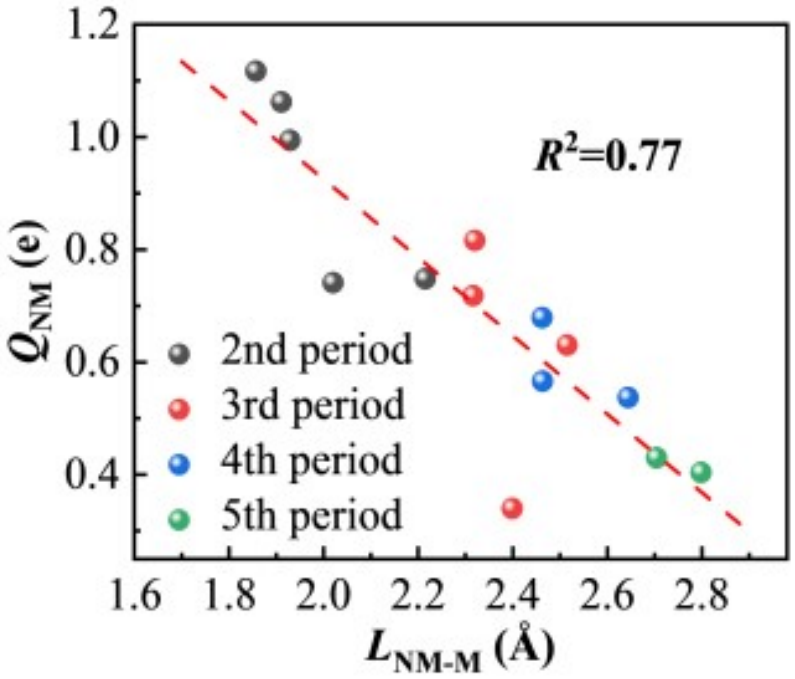

Figure S3. Scatter plot of Q_{NM} versus L_{NM-M} for NM-VTe_2 structures. Q_{NM} represents the net electron gain of the doped non-metal atom, and L_{NM-M} is the bond length between the doped atom and neighboring vanadium atoms.

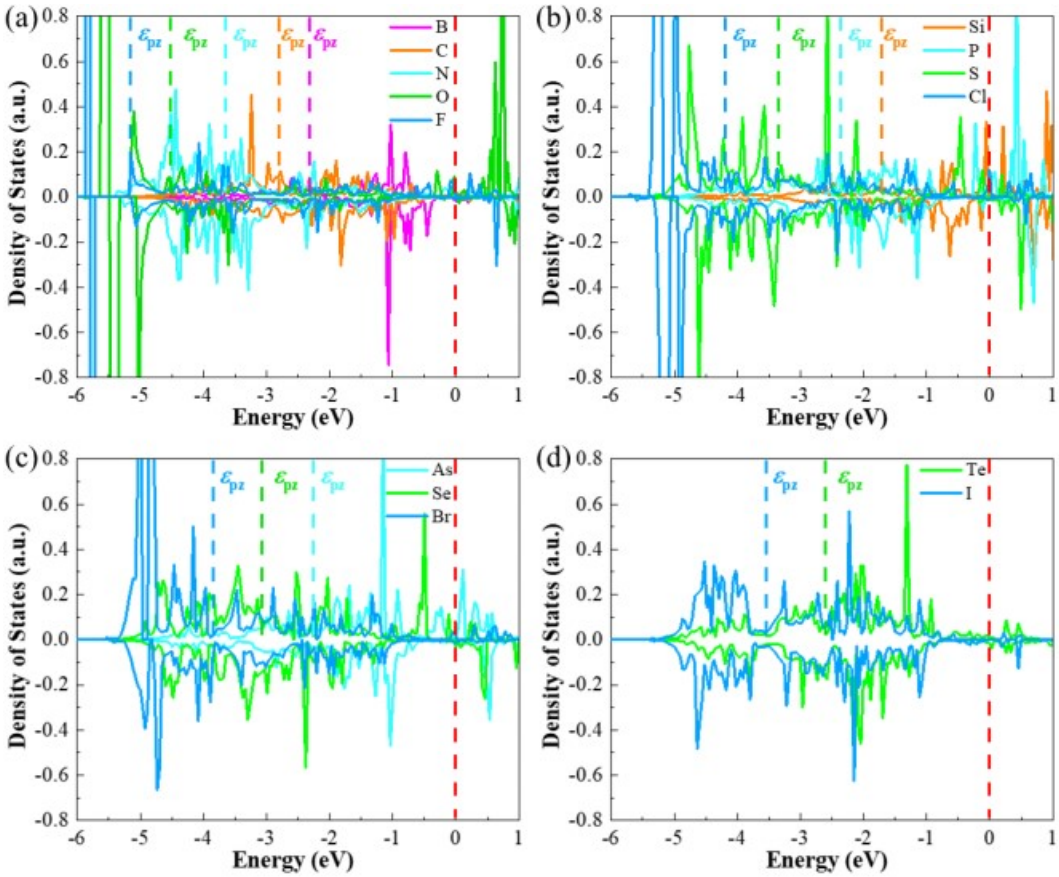

Figure S4. Density of states (DOS) of the pz orbitals of non-metal dopants from different periods in NM-VTe_2, highlighting the relative positions of their pz band centers. (a) Second-period elements; (b) Third-period elements; (c) Fourth-period elements; (d) Fifth-period elements.

Table S1. Binding energies (E_b), bond lengths (L_{NM-M}), net electron gain (Q_{NM}), pz band centers (ε_{pz}), and Gibbs free energies of hydrogen adsorption (ΔG_{H^*}) for various NM-VTe_2 structures.

Structure	E_b (eV)	L_{NM-M} (Å)	Q_{NM} (e)	ε_{pz} (eV)	ΔG_{H^*} (eV)	
					site1	site2
B-VTe_2	-1.798	2.02	0.741	-2.328	-0.17	0.507
C-VTe_2	-2.656	1.911	1.062	-2.794	-0.161	0.605
N-VTe_2	-1.783	1.857	1.117	-3.641	0.031	0.552
O-VTe_2	-4.301	1.929	0.994	-4.54	0.476	0.523
F-VTe_2	-4.13	2.215	0.748	-5.172	1.524	0.555
Si-VTe_2	0.108	2.399	0.34	-1.737	-0.532	0.462
P-VTe_2	-1.451	2.316	0.718	-2.355	-0.032	0.548
S-VTe_2	-2.867	2.32	0.816	-3.363	0.846	0.467
Cl-VTe_2	0.243	2.515	0.63	-4.188	2.115	0.672
As-VTe_2	-1.77	2.463	0.566	-2.196	0.024	0.476
Se-VTe_2	-2.232	2.462	0.679	-3.079	1.265	0.439
Br-VTe_2	-3.974	2.644	0.537	-3.817	1.906	0.66
Te-VTe_2	/	2.704	0.43	-2.584	0.413	0.413
I-VTe_2	-1.912	2.798	0.404	-3.505	1.724	0.566