Supporting Information

Revolutionizing Water Splitting Performance by Probing the Influence of Electron Transfer in NiCr-LDH/VS₂/NF Heterostructure

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This file contains 31 pages in which the specification of reagents, methods of synthesis, instruments used in the study, electrochemical characterization, electrochemical results, characterizations like XRD, HR-TEM images, EDS, XPS, BET result analysis, contact angle of NF, Band structures, Mott Schottky plot, probable reaction mechanisms, post-FESEM, post-HR-TEM images, post-XPS, comparison table, computational methodology, references are given.

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Experimental Section

Synthesis of Bare VS₂/NF:

Before synthesis, with the purpose of removing the oxide coating on the surface, two pieces of $(4.5\times0.5 \text{ cm}^2)$ Ni foam (NF) were dipped in diluted hydrochloric acid following sonication for 5 minutes. Afterward, the NFs were washed with ethanol and deionized water respectively several times. VS₂ were synthesized by hydrothermal process. For a typical synthesis, 2 mmol of NH₄VO₃ was dissolved in a 30 mL of DI water. Further, 15 mmol of C₂H₅NS and 2 ml of NH₃ were added to the same solution and stirred for 20 minutes on a magnetic stirrer. The resulting dispersed solution then transferred to Teflon-lined autoclave. To this dissolved precursor, surface cleaned 2 × 2 nickel foam (NF) was kept inside and sealed properly. The autoclave kept for 14 h at a temperature of 160 °C in a hot air oven. After the completion of reaction, the in-situ grown VS₂ was cleaned several times with DI water and dried at 60 °C. Then the obtained bare VS₂/NF was used for formation of heterostructure with the NiCr-LDH. Also, the suspension was centrifugated, washed with DI water and ethanol respectively, and dried at 60 °C, and then Bare VS₂/NF powder was obtained which is further used for characterization purposes.

Synthesis of NiCr-LDH/NF:

Before synthesis, with the purpose of removing the oxide coating on the surface, two pieces of $(4.5 \times 0.5 \text{ cm}^2)$ Ni foam (NF) were dipped in diluted hydrochloric acid following sonication for 5 minutes. Afterward, the NFs were washed with ethanol and deionized water respectively several times. Typically, for the synthesis of NiCr-LDH/NF 0.05 M Ni(NO₃)₂.6H₂O, 0.05 M Cr(NO₃)₃.9H₂O, 0.35 mM CO(NH₂)₂ (Urea), and NH₄F were dissolved in a 1:1 mixture of 30 mL of deionized water with continuous stirring for 10 minutes. Henceforth, the resulting solution was relocated into a 100 mL Teflon-lined stainless-steel autoclave and the washed pre-prepared NFs

were immersed into the solution. Subsequently, the sealed autoclave was transferred to an electric oven for hydrothermal treatment at 160 °C for 14 hours and henceforward cooled down to room temperature.^{1,2} The as-synthesized NiCr-LDH sample coated on NF was collected and rinsed with deionized water quite a few times and then dried overnight. Also, the suspension was centrifugated, washed with DI water and ethanol respectively, and dried at 60 °C, and then NiCr LDH powder was obtained which is further used for characterization purposes.

Synthesis of heterostructured NiCr-LDH/VS₂/NF

The heterostructured NiCr-LDH/VS₂/NF was synthesized through same hydrothermal treatment for 14 h. Here instead of bare NF, in-situ grown Bare VS₂/NF was immersed in the disperse solution of 0.05 M Ni(NO₃)₂.6H₂O, 0.05 M Cr(NO₃)₃.9H₂O, 0.35 mM CO(NH₂)₂ (Urea), and NH₄F in a Teflon-lined stainless-steel autoclave and kept at 160 °C for 14 h.

Similarly, for the synthesis of $VS_2/NiCr-LDH/NF$ heterostructure, vice-versa procedure is used, where in-situ grown NiCr-LDH/NF was immersed in the disperse solution of VS_2 and hydrothermal method was carried out.

The obtained NiCr-LDH/VS₂/NF and VS₂/NiCr-LDH/NF was washed several times with water-ethanol mixture and dried in hot air oven at 60 °C for 12 h. Finally, all the obtained product was used as electrocatalyst for OER, HER and overall water splitting reaction.

Reagents and Instruments:

Nickel Nitrate $[Ni(NO_3)_2.6H_2O]$, Chromium Nitrate $[Cr(NO_3)_3.9H_2O]$, Urea $CO(NH_2)_2$, Ammonium Vanadate (NH_4VO_3) , Thioacetamide (C_2H_5NS) , Ammonia $[NH_3]$ were purchased from Sigma-Aldrich and used as received. Ni foam was procured from Sigma-Aldrich and used after surface cleaning. The electrochemical analyzer AURT-M204 was used for all electrochemical characterizations. Hg/HgO reference electrode (in 1 M KOH) was purchased from CH instruments and platinum (Pt) and graphite as counter electrodes from Alfa-Aesar were used throughout the electrochemical studies along with the Ni foam with materials grown as working electrodes. DI water was used throughout the entire experiment. The as-prepared catalysts with different stoichiometric ratios were characterized with HR-TEM, (TecnaiTM G2 TF20) working at an accelerating voltage of 200 kV and by Talos F-200-S with HAADF elemental mapping. Color mapping and Energy Dispersive X-ray Spectroscopy (EDS) analysis were carried out with the FESEM instrument with the images (SUPRA 55VP Carl zeiss) with a separate EDS detector connected to that instrument. The XRD analysis carried out with a scanning rate of 5° min-1 in the 2θ range 10-90° using a Rigaku X-ray powder diffractometer (XRD) with Cu Ka radiation ($\lambda =$ 0.154 nm). X-ray photoelectron spectroscopic (XPS) analysis was performed using a Theta Probe AR-XPS system (Thermo Fisher Scientific, UK). The Bruker Tensor 27 (Optik GmbH, Germany) with an RT DLaTGS (Varian) detector was used for FT-IR analysis. LASER Raman spectroscopic measurements were carried out by green emitting semiconductor as a laser source of 532 nm. For Raman experiment the excitation light intensity is about approximately 10 mW with a spectral collection of time of 1 sec. The integration time for our measurement was set to be 10 s.

Electrochemical Characterizations:

The electrochemical workstation AURT-M204 had been used for the entire total water splitting studies. Electrocatalytic studies were done with the conventional three-electrode system. For OER, the commercial Hg/HgO, and Pt ring were used as a reference and counter electrodes respectively, and for HER commercial Hg/HgO, and graphite were used as a reference and counter electrodes respectively in 1 M KOH solution. For total water splitting, NiCr-LDH/VS₂/NF were used as both

cathode and anode. The polarization studies were carried out at a slow scan rate of 5 mV/sec. 50% iR compensation was done manually from the Rs from EIS. Continuous rapid sweeping through accelerated degradation (AD) studies at a very high sweep rate of 200 ms⁻¹ for 500 Cycles were Carried out in 1 M KOH for OER and also for HER. For handling the chemicals and glassware for the synthesis process as well as the application part, safety gloves, lab coats, and safety glass were mandatory and used accordingly.

All the resulting potential data that were collected by taking Hg/HgO as a reference electrode were later converted with respect to the reversal hydrogen electrode (E_{RHE}) by considering the Nernst equation of

$$E_{RHE} = E_{ref} + 0.059 \times 14 + 0.098...$$
Equation S1

Over potential (η) values of all the catalysts at benchmarking current density of 10 mA/cm² calculation has been done by following this equation

 $\eta = E_{RHE}$ - 1.23 V.....Equation S2

Tafel slope was calculated by fitting η vs log(j) using the Tafel equation

 $\eta = b \cdot \log(j/j_0)$ Equation S3

where b represents the Tafel slope, j signifies the current density and j_0 is the exchange current density. Electrochemical impedance spectroscopy (EIS) measurements were done on the frequency ranges from 10^5 to 1 Hz at 300 mV *vs* RHE. The value of electrochemical active surface areas (ECSA) can be measured by determining the electrochemical double layer capacitance (C_{dl}) as follows:

$$i_c = v \times C_{dl}$$
.....Equation S4

 $ECSA = \frac{C_{dl}}{C_s}....Equation S5$

Where i_c indicates the double-layer charging current resulting from scan-rates (v) dependent CVs at non-faradic potential, C_s denotes a specific capacitance value of 0.040 mF/cm² depending on the typical reported values.

Determination of Surface concentration of various materials from the redox features of CV:

• Calculated area associated with the reduction of Ni^{3+} to Ni^{2+} of NiCr-LDH/VS₂/NF = 0.034 VA

Hence, the associated charge is = $0.034 \text{ VA} / 0.005 \text{ Vs}^{-1}$

= 6.8 As = 6.8 C

Now, the number of electrons transferred is = $6.8 \text{ C} / 1.602 \times 10^{-19}$

$$= 4.24 \times 10^{19}$$

Since the reduction of Ni³⁺ to Ni²⁺ is a single electron transfer reaction, the number of electrons calculated above is exactly the same as the number of surface-active sites.

Hence, the number of Ni participating in OER is = 4.24×10^{19}

In our study, the determination of Turnover Frequency (TOF) from OER Current Density TOF was calculated assuming that the surface-active Ni atoms that had undergone the redox reaction just before the onset of OER only participated in OER electrocatalysis. The corresponding expression is,

$$TOF = j \times N_A / F \times n \times \Gamma$$

Where, j = current density, N_A = Avogadro number, F = Faraday constant, n = Number of electrons, Γ = Surface concentration.

Hence, for NiCr-LDH/VS₂/NF at 1.71 V, we have the TOF value of

$$\text{TOF}_{1.71 \text{ V}} = [(551.7 \times 10^{-3}) (6.023 \times 10^{23})] / [(96485) (4) (4.24 \times 10^{19})]$$

$= 0.02028 \text{ sec}^{-1}$

- Similarly, we calculated the TOF value for Bare VS₂/NF, NiCr-LDH/VS₂/NF and VS₂/NiCr-LDH/NF at 1.71 V and also at different potential value of 1.65 V and 1.76 V, where in every potential value the TOF value increases from Bare VS₂/NF → NiCr-LDH/NF → VS₂/NiCr-LDH/NF → NiCr-LDH/VS₂/NF, suggesting optimized NiCr-LDH/VS₂/NF is the best active synthesized catalyst.
- In the case of HER reaction TOF was calculated by considering the area of the C_{dl} curve to calculate surface concentration value and the obtained TOF values at -0.161 V, -0.213V, -0.261V for NiCr-LDH/NF, Bare VS₂/NF, NiCr-LDH/VS₂/NF and VS₂/NiCr-LDH/NF are portrayed in Figure 3a.



catalysts for visualization of peak shift of reflection planes.



Figure S2: (*a*), (*b*) Low to high magnified FE-SEM images of NiCr-LDH/NF; (*c*), (*d*) Low to high magnified FE-SEM images of Bare VS_2/NF ; and (*e*), (*f*)) Low to high magnified FE-SEM images of $VS_2/NiCr$ -LDH/NF.



Figure S3: (*a*), (*b*), (*c*), and (*d*) are EDAX spectrum of of NiCr-LDH/NF, Bare VS₂/NF, NiCr-LDH/VS₂/NF, and VS₂/NiCr-LDH/NF respectively.

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Figure S4: (*a*), (*b*) Low to high magnified HR-TEM images of NiCr-LDH/NF; (*c*) SAED pattern of NiCr-LDH/NF; inset: Lattice fringes of NiCr-LDH/NF; (*d*) HAADF area of NiCr-LDH/NF chosen for color mapping and (*e*-*i*) portray the colour mapping of the mix, Ni, Cr, C, and O respectively; (*j*), (*k*) Low to high magnified HR-TEM images of Bare VS₂/NF; (*l*) SAED pattern of Bare VS₂/NF; inset: Lattice fringes of Bare VS₂/NF; (*m*) HAADF area of Bare VS₂/NF chosen for color mapping and (*n*-r) portray the colour mapping of the mix, V, S, C, and O respectively.



Figure S5: (*a*), (*b*) Low to high magnified HR-TEM images of VS₂/NiCr-LDH/NF; (*c*) Lattice fringes of VS₂/NiCr-LDH/NF; (*c*) SAED pattern of NiCr-LDH/VS₂/NF; (*e*) HAADF area of VS₂/NiCr-LDH/NF chosen for color mapping and (f-l) portray the colour mapping of the mix, Ni, Cr, V, S, C, and O respectively.



NiCr-LDH/NF	Bare VS ₂ /NF	NiCr-LDH/VS ₂ /NF		
a	b	С		
BET summary Slope = 139.675 Intercept = 1.0976 Correlation coefficient, r = 0.8425 C constant= 13.731	BET summary Slope = 537.363 1 Intercept = 2.113e+(Correlation coefficient, r = 0.991923 0.991923 C constant= 26.436 0.001100	BET summary Slope = 40.328 Intercept = 6.417e+00 Correlation coefficient, r = 0.781183 C constant= 7.284		
Surface Area = 23.117	Surface Area = 6.236 m ²	g Surface Area = 74.500 m²/g		
Total Pore Volume summ Total Pore Volume	Y Total Pore Volume summ. Total Pore Volume	Total Pore Volume summary Total Pore Volume		
Total pore volume = 3.020e-02 cc/g for pores smaller than 1027.0 Å (Radii at P/Po = 0.99058	Total pore volume = 6.974e-03 cc/g for pores smaller than 958.9 Å (Radius at P/Po = 0.98990	Total pore volume = 5.326e-02 cc/g for pores smaller than 930.5 Å (Radius) at P/Po = 0.98959		
Average Pore Size sum	Average Pore Size summary	Average Pore Size summary		
Average pore Radius = 2.61276e+01 Å	Average pore Radius = 2.23672e+01 Å	Average pore Radius = 1.42983e+01 Å		
Average Pore Width of NiCr-LDH/NF	Average Pore Width of Bare VS2/NF	Average Pore Width of NiCr-LDH/ VS ₂ / NF		
= 2 × Pore Radius	= 2 × Pore Radius	= 2 × Pore Radius		
$= 2 \times 2.613 \text{ nm}$	$= 2 \times 2.237 \text{ nm}$	= 2 × 1.429 nm		
= 5.226 nm	= 4.474 nm	= 2.858 nm		
= mesoporous	= mesoporous	= mesoporous		
Figure S7: BET results s	nmary.			



Figure S8: Contact Angle measurement of Nickel Foam used as substrate of working electrodes.





Figure S10: (*a*),(*b*), (*c*),(*d*) shows the CVs recorded for NiCr-LDH/NF, Bare VS₂/NF, NiCr-LDH/VS₂/NF and VS₂/NiCr-LDH/NF respectively in a non-faradaic region at various scan rate for the determination of ECSA from its double layer capacitance in 1M KOH solution.



VS₂/NF, NiCr-LDH/VS₂/NF and VS₂/NiCr-LDH/NF respectively.



*VS*₂/*NiCr-LDH*/*NF*, *NiCr-LDH*/*NF*, and Bare *VS*₂/*NF* respectively.





Figure S14: (*a*), and (*b*) Mott–Schottky plot for NiCr-LDH and Bare VS₂ with respect to RHE.





Figure S16: Partial charge density distribution of the NiCr-LDH/VS₂ heterostructure in the Energy interval of -1eV to 0eV; (a) side view and (b) top view. Dotted black lines denote the boundary of simulation cell. white, red, green, light blue, grey and yellow colored balls represent H, O, Cr, Ni, V, and S atoms respectively.



Figure S17: (*a*), (*b*) Low- to high-magnified Post-FE-SEM images of NiCr-LDH/VS₂/NF; (*c*) EDAX spectrum of post-NiCr-LDH/VS₂/NF; (*d*) HR-TEM image of post-NiCr-LDH/VS₂/NF; (*e*) SAED pattern of post-NiCr-LDH/VS₂/NF; Inset- Lattice fringes of post-NiCr-LDH/VS₂/NF; (*f*) HAADF area of post-NiCr-LDH/VS₂/NF chosen for color mapping and (*g*-*m*) portray the colour mapping of the mix, Ni, Cr, V, S, C, and O respectively; (*n*-*p*) are deconvoluted XPS curve of Ni 2p, Cr 2p, O 1s of post-NiCr-LDH/VS₂/NF respectively.



SI.		Catalyst		Potential	Electrolyte	Current	Reference
No	SI. No	Catalyst	Ov	erpotential (mV)	Current density	Taf erstojs e (m (val A/stale)	Reference
1		CeCO ₃ OH/Ce-CoFe LDH		1.61	(matting)H	10	3
2	1	CeCO ₂ OH/Ce-CoFe LDH) CeCO ₂ OH/Ce-CoFe N1V-LDH(@Mn ₂ O ₃		216	1.0 M KOH	40.1	3 4
2	$\frac{2}{3}$	NIV-LDHCGMncQ3 NFVMorNitEg LDHKNiSt		298 249 60	50 10100KOH	89 44170	5 5
	4	NFMD-HEELDINIS,		209	50	65.4	6
4		ICo-LDH@NiCoS@NiFe-LDH VICoNIDSI & ORFOSED H& PEDH		1.55 330	1.0 M KOH 100	<u>10</u> 90.9	6
5	NP3S	ZrCoFe-159142189F #RH3S2/ZrCoI Fe-dopadr Goe DDH/20052-	Fe-	24 3 .57 220	1.0 ₩ KOH	59. <u>50</u> 47	8 7 9
6	, 	VSe2Nine-IDH		1.66	1.0 M KOH	10	8
7	− Pe-	USE/WHELLIDH doped Co-LDHSeWS3Ni2S2/NF		227.54	1.0 M KOH	59.90	11 9
	_1₽e	doped Corther Mos 2-Wi 3S2 /NF	11	209	50	70	This work
8		Ni_2Cr_1 -LDH Ni_2Cr_1 -LDH		1.55	1.0 M KOH	10	10
9		$Ni_2S_2/VS_2 \parallel N-Ni_2S_2/VS_2$		1 648	10 M KOH	10	11

Table S1: Comparison table for OER activity of NiCr-LDH/VS₂/NF with similar type of catalyst.

Table S2: Comparison table for HER activity of NiCr-LDH/VS₂/NF with similar type of catalyst.

SI. No	Catalyst	Overpotential (mV)	Current density (mA/cm ²)	Tafel slope (mV/decade)	Reference
1	CeCO ₃ OH/Ce-CoFe	165	10	89.8	3
2	NiV-LDH@Mn ₂ O ₃	185	50	98	4
3	NF/Mo-NiFe LDH/NiS _x	169	100	42.4	5
4	NiCo-LDH@NiCoS@NiFe-	93	10	93.3	6
	LDH				
5	Ni ₃ S ₂ /ZrCoFe-LDH@NF	159.2	10	96	7
6	VSe ₂ /NiFe-LDH	168	10	98	8
7	Fe-doped Co-LDH@MoS ₂ -	95	10	44	9
	Ni ₃ S ₂ /NF				
8	Ni ₂ Cr ₁ -LDH	138	100	61.5	10
9	N-Ni ₃ S ₂ /VS ₂	151	10	107.5	11
10	NiCr-LDH/VS ₂ /NF	116	50	110	This work

Table S3: Comparison table for TWS activity of NiCr-LDH/VS₂/NF with similar type of

Computational Methodology:

All calculations were performed using spin-polarized density functional theory (DFT) with the Vienna *Ab initio* Simulation Package (VASP). ¹² The unit cell was optimized using Perdew–Burke–Ernzerhof (PBE) functional ¹³ of the generalized gradient approximation (GGA) with the addition of on-site Hubbard U term for Ni, Co, and V. Based on previous literature, ¹⁴ U value of 6.2, 3.0, and 3.0 eV were chosen for Ni, Co, and V, respectively. On the other hand, J value of 0.75 eV was taken invariably for all three elements. We also used a cut-off energy value of 500 eV and the Brillioun zone was sampled using a $9 \times 9 \times 1$ k-points mesh. To analyse the charge distribution, Bader charge analyses¹⁵ were performed to understand the charge density distribution of the systems and VESTA ¹⁶ software were used for visualize the atomic models as well as charge density distribution. Vacuum thickness of ~15 Å is maintained along the z-axis of the structures to nullity the interaction between the real and periodic images of the system.

Reference

- Wang, Z.; Liu, W.; Hu, Y.; Xu, L.; Guan, M.; Qiu, J.; Huang, Y.; Bao, J.; Li, H. An Fe-Doped NiV LDH Ultrathin Nanosheet as a Highly Efficient Electrocatalyst for Efficient Water Oxidation. *Inorg Chem Front* 2019, 6 (7), 1890–1896. https://doi.org/10.1039/C9QI00404A.
- (2) Li, G.; Xu, Y.; Pan, H.; Xie, X.; Chen, R.; Wu, D.; Wang, L. A Bimetallic Synergistic Effect on the Atomic Scale of Defect-Enriched NiV-Layered Double Hydroxide Nanosheets for Electrochemical Phenol Hydroxylation. *J Mater Chem A Mater* 2022, *10* (12), 6748–6761. https://doi.org/10.1039/D1TA09412B.

- (3) Tang, J.; Hu, J.; Chen, X.; Yang, B.; Zhang, K.; Li, Y.; Yao, Y.; Zhang, S. Boosting Activity on CoFe Layered Double Hydroxide by Doping and Heterostructure as a Bifunctional Electrocatalyst for Efficient Overall Water Splitting. *Fuel* 2024, 365. https://doi.org/10.1016/j.fuel.2024.131128.
- Madhu, R.; Kundu, S. Unlocking the Potential of NiV-LDH@Mn2O3 Heterostructure via Band Gap Modulation for Enhanced Water Splitting. *J Mater Chem A Mater* 2024, *12* (32), 21385–21397. https://doi.org/10.1039/d4ta03779k.
- (5) Peng, W.; Zhang, W.; Lu, Y.; Li, W.; He, J.; Zhou, D.; Hu, W.; Zhong, X. Mo-Doping and Construction of the Heterostructure between NiFe LDH and NiSx Co-Trigger the Activity Enhancement for Overall Water Splitting. *J Colloid Interface Sci* 2024, 664, 980–991. https://doi.org/10.1016/j.jcis.2024.03.062.
- Jiang, H.; Qin, H.; Zhou, P.; Kong, L.; Wang, C.; Ji, Z.; Shen, X.; Zhu, G.; Yuan, A. Partial Sulfidation Strategy to NiCo-LDH@NiCoS Coupled with NiFe-LDH for Highly Efficient Overall Water Splitting. *Int J Hydrogen Energy* 2024, 58, 892–901. https://doi.org/10.1016/j.ijhydene.2024.01.292.
- (7) Guo, Y.; Zou, X.; Wei, X.; Bao, W.; Zhang, J.; Han, J.; Jia, F. Fe Regulating Ni3S2/ZrCoFe-LDH@NF Heterojunction Catalysts for Overall Water Splitting. *Chinese Journal of Structural Chemistry* 2024, 43 (2), 100206. https://doi.org/10.1016/j.cjsc.2023.100206.
- (8) Hegde, A. P.; Mukesh, P.; G., L. S.; Kumar, A.; Nagaraja, H. S. Nano-Composites of NiFe-LDH/V Se2 Heterostructures for Effective Water Splitting Electrocatalyst. *Int J Hydrogen Energy* 2024, *71*, 1456–1467. https://doi.org/10.1016/j.ijhydene.2024.05.232.

- (9) Phadikar, U.; Sanyal, G.; Das, S.; Kundu, A.; Kuila, C.; Murmu, N. C.; Chakraborty, B.; Kuila, T. Unique Multi-Hetero-Interface Engineering of Fe-Doped Co-LDH@MoS2-Ni3S2 Nanoflower-Based Electrocatalyst for Overall Water-Splitting: An Experimental and Theoretical Investigation. *ChemSusChem* 2024. https://doi.org/10.1002/cssc.202400821.
- Ye, W.; Fang, X.; Chen, X.; Yan, D. A Three-Dimensional Nickel-Chromium Layered Double Hydroxide Micro/Nanosheet Array as an Efficient and Stable Bifunctional Electrocatalyst for Overall Water Splitting. *Nanoscale* 2018, 10 (41), 19484–19491. https://doi.org/10.1039/c8nr05974h.
- (11) Zhong, X.; Tang, J.; Wang, J.; Shao, M.; Chai, J.; Wang, S.; Yang, M.; Yang, Y.; Wang, N.;
 Wang, S.; Xu, B.; Pan, H. 3D Heterostructured Pure and N-Doped Ni3S2/VS2 Nanosheets for High Efficient Overall Water Splitting. *Electrochim Acta* 2018, 269, 55–61. https://doi.org/10.1016/j.electacta.2018.02.131.
- (12) Kresse, G.; Furthmüller, J. Efficient Iterative Schemes for Ab Initio Total-Energy Calculations Using a Plane-Wave Basis Set. *Phys Rev B Condens Matter Mater Phys* 1996, 54 (16), 11169–11186. https://doi.org/10.1103/PhysRevB.54.11169.
- (13)Perdew, J. P.; Chevary, J. A.; Vosko, S. H.; Jackson, K. A.; Pederson, M. R.; Singh, D. J.; Fiolhais, C. Erratum: Atoms, Molecules, Solids, and Surfaces: Applications of the Generalized Gradient Approximation for Exchange and Correlation (Physical Review B (1993)48, 7, (4978)). В 1993, 48 4978. Phys Rev (7),https://doi.org/10.1103/PhysRevB.48.4978.2.
- (14) Hubbard U Values | MP Public Docs. (n.d.). Retrieved September 18, 2024,. MP Public Docs.

- (15) Henkelman, G.; Arnaldsson, A.; Jónsson, H. A Fast and Robust Algorithm for Bader Decomposition of Charge Density. *Comput Mater Sci* 2006, *36* (3), 354–360. https://doi.org/10.1016/j.commatsci.2005.04.010.
- Momma, K.; Izumi, F. VESTA 3 for Three-Dimensional Visualization of Crystal, Volumetric and Morphology Data. J Appl Crystallogr 2011, 44 (6), 1272–1276. https://doi.org/10.1107/S0021889811038970.