Supporting Information

Modulating the electronic structure of Ru *via* VS_2 decoration for efficient pH-universal electrocatalytic hydrogen evolution reaction

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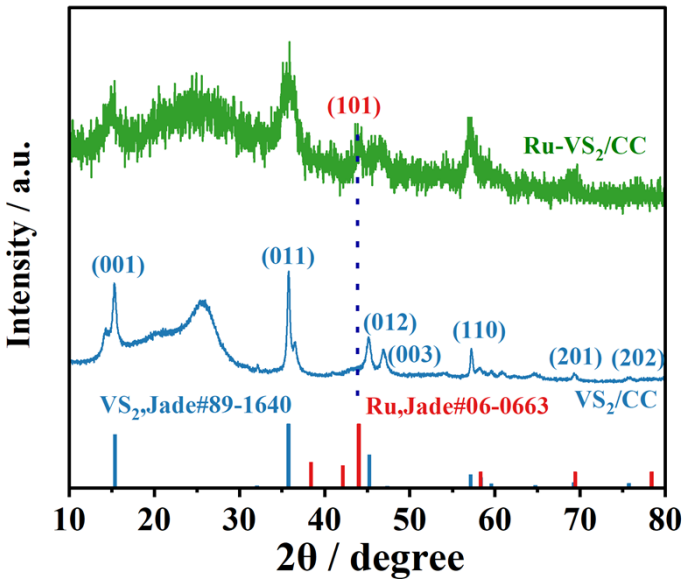

Fig. S1. XRD patterns of the prepared VS_2 and Ru-VS_2.

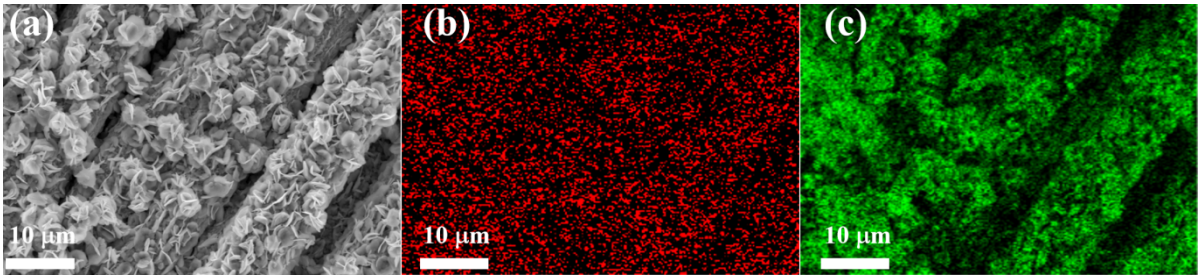

Fig. S2. (a) SEM image of VS_2. EDS-mapping of (b) V and (c) S in VS_2.

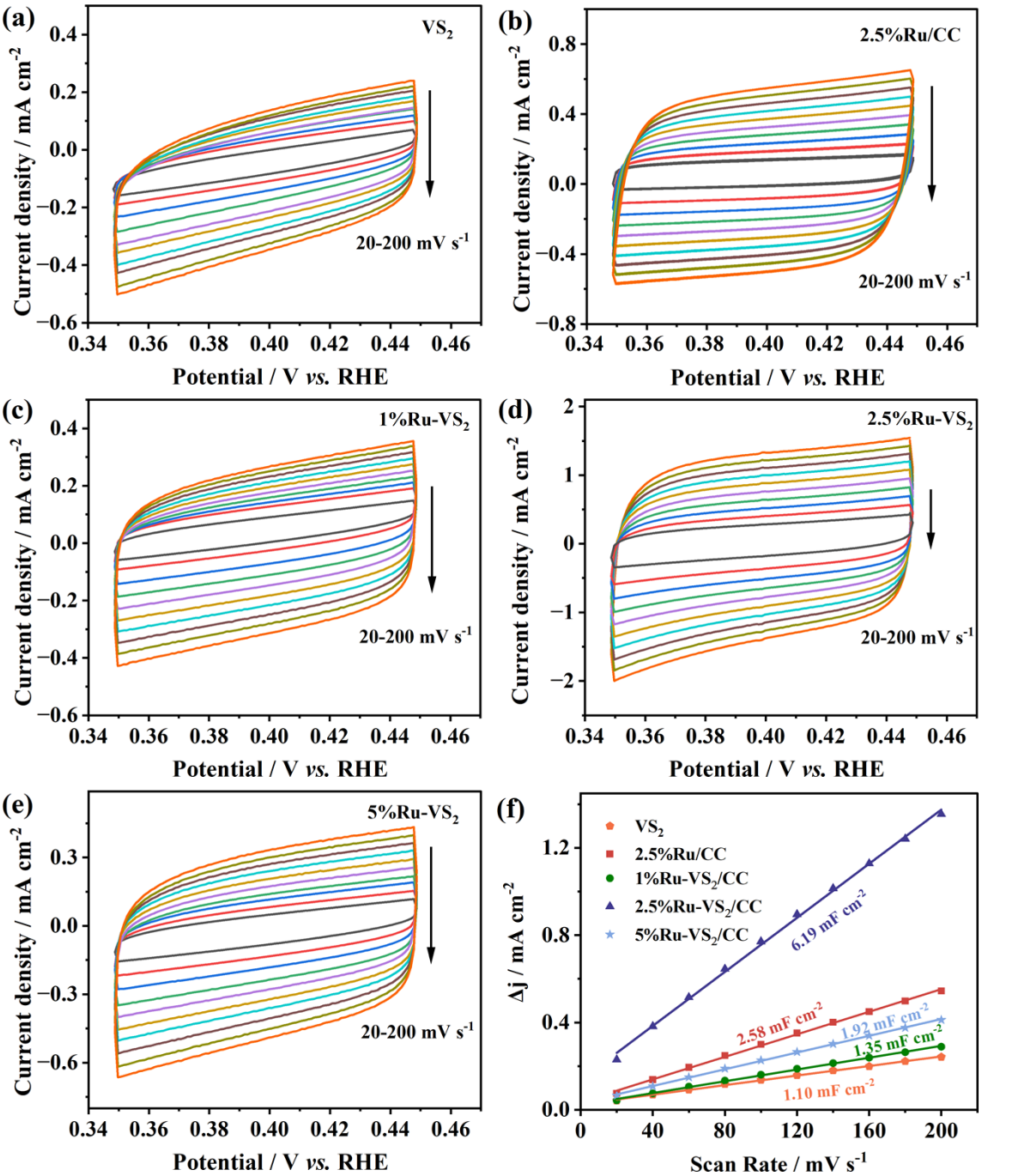

Fig. S3. CV curves of (a) VS_2/CC, (b) 2.5% Ru/CC, (c) 1% Ru-VS_2/CC, (d) 2.5% Ru-VS_2/CC and (e) 5% Ru-VS_2/CC samples at scanning rates of 20-200 mV s^{-1} in a potential

window without faradaic process and (f) the corresponding C_{dl}.

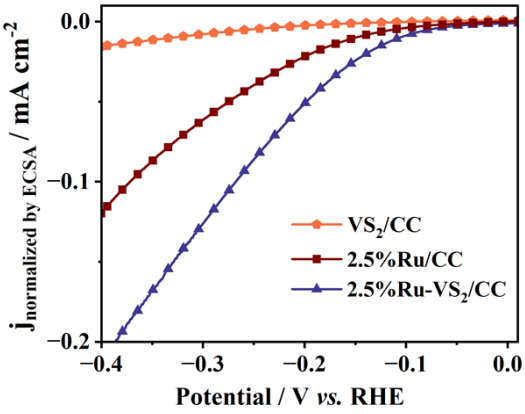

Fig. S4. ECSA-normalized LSV curves of VS_2/CC, 2.5%Ru/CC and 2.5%Ru-VS_2/CC for HER.

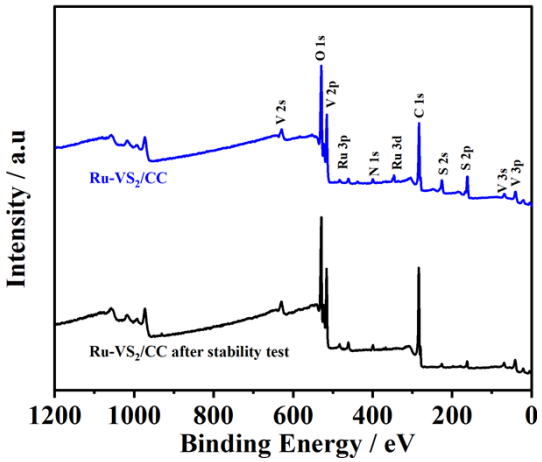

Fig. S5. XPS survey of the Ru-VS_2 before and after stability test.

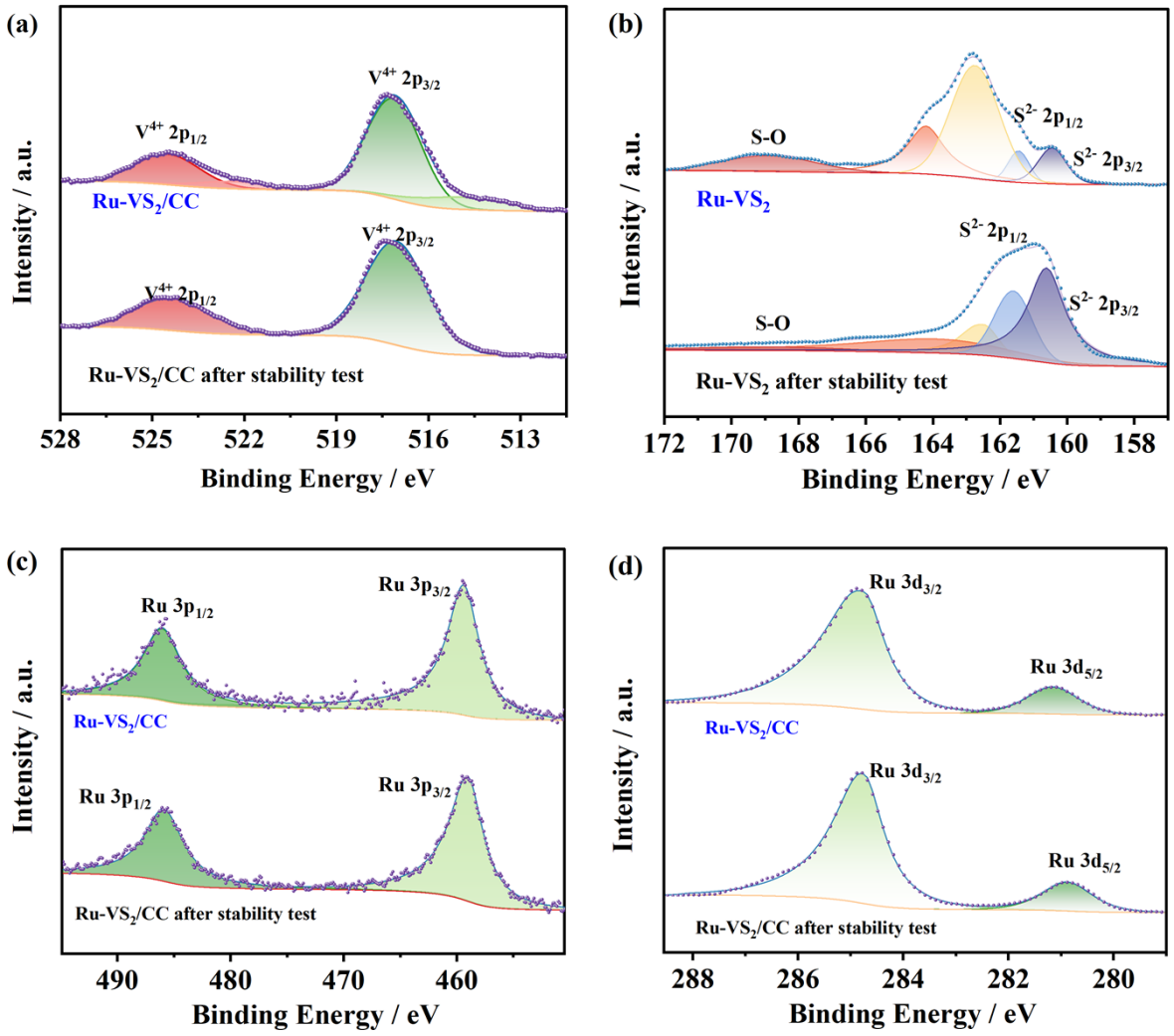

Fig. S6. High-resolution XPS spectra of (a) V, (b) S, (c) Ru 3p and (d) Ru 3d of Ru-VS_2 before and after stability test.

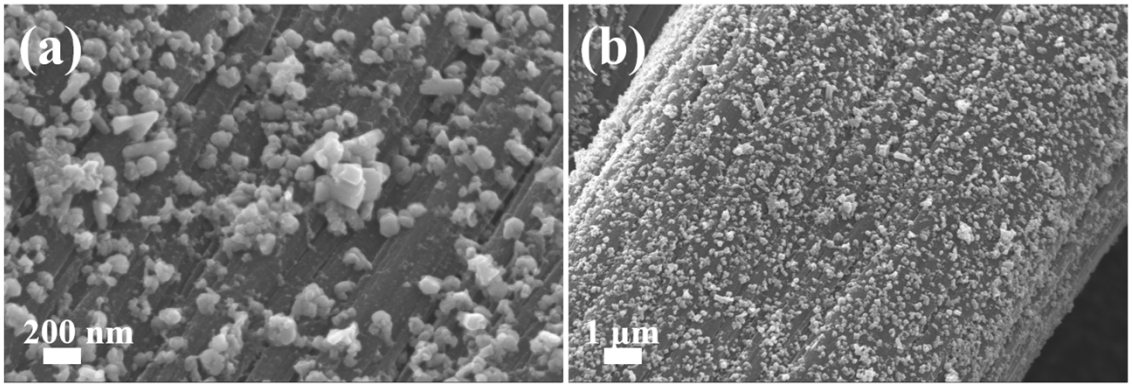

Fig. S7. SEM images of Ru-VS_2 after stability test.

Table S1. Comparisons of HER activity of 2.5%Ru-VS_2/CC with other electrocatalysts.

Electrocatalyst	η_{10} (mV)	Tafel slope (mV dec^{-1})	Electrolyte	iR	Reference
2.5%Ru-VS_2/CC	89	63	0.5 M H_2SO_4	no	This Work
	87	71	1.0 M KOH		
	220	137	1.0 M PBS		
1Pt/VS_2/CP	77	44.13	0.5 M H_2SO_4	-	[1]
10:MoCo-VS_2/CC	63	50	1.0 M KOH	yes	[2]
MoS_2/VS_2	199.6	95.2	0.5 M H_2SO_4	yes	[3]
VS_2-Mo-10	243	52.6	0.5 M H_2SO_4	yes	[4]
$CoMnS_2$@1T-Fe-VS_2@NF	89	61	1.0 M KOH	yes	[5]
2H-VS_2-Pd	157 (20 mA cm^{-2})	75	0.5 M H_2SO_4	yes	[6]
VS_2 NDs annealed	350	79	0.3 M H_2SO_4	-	[7]
Bio-templated VS_2	160	50	0.5 M H_2SO_4	-	[8]
VS_2	68	34	0.5 M H_2SO_4	yes	[9]
TS-Co_3O_4@VS_2	175.29	57	0.5 M H_2SO_4	-	[10]
VS_2	58	34	0.5 M H_2SO_4	-	[11]
self-assembled VS_2	197	134.39	1.0 M KOH	yes	[12]
Ru-MoS_2/CC	169	95	0.5 M H_2SO_4	85% iR	[13]
	90	130	1.0 M KOH		
Ru-MoS_2	110	78	0.5 M H_2SO_4	no	[14]
	98	65	1.0 M KOH		
Ru-MoS_2/CC	61	114	1.0 M KOH	yes	[15]
Ru/np-MoS_2	30	31	1.0 M KOH	-	[16]
SA-Ru-MoS_2	76	21	1.0 M KOH	-	[17]
Ru/Ni-MoS_2	32	41	1.0 M KOH	no	[18]
Ru_1@D-MoS_2	107	96	1.0 M KOH	-	[19]
Ru-$MoSe_2$/CMT	70	39	1.0 M KOH	yes	[20]
Ru@Ni_3S_2	19.8	33.2	1.0 M KOH	90% iR	[21]
Ru/Mo_2CT_x	64	66	0.5 M H_2SO_4	85% iR	[22]
	78	49	1.0 M KOH		
	73	57	1.0 M PBS		
Ru-MoP-P_V	100	49	0.5 M H_2SO_4	95% iR	[23]
	79	49	1.0 M KOH		
	161	70	1.0 M PBS		
N, Ru Co-doped Sb_2S_3	72	193	1.0 M KOH	yes	[24]
Ru@WNO-C	172	38.9	0.5 M H_2SO_4	100% iR	[25]

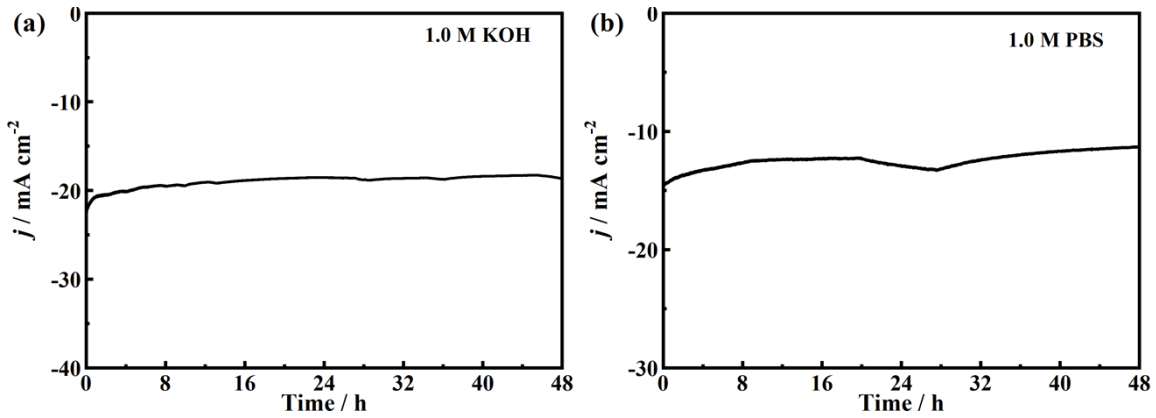

Fig. S8. chronoamperometry curves of Ru-VS_2/CC in (a) 1.0 M KOH and (b) 1.0 M PBS.

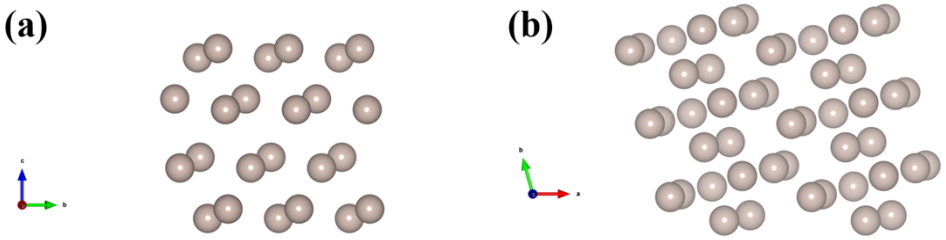

Fig. S9. Ball-stick models of VS_2 (a) side view and (b) top view.

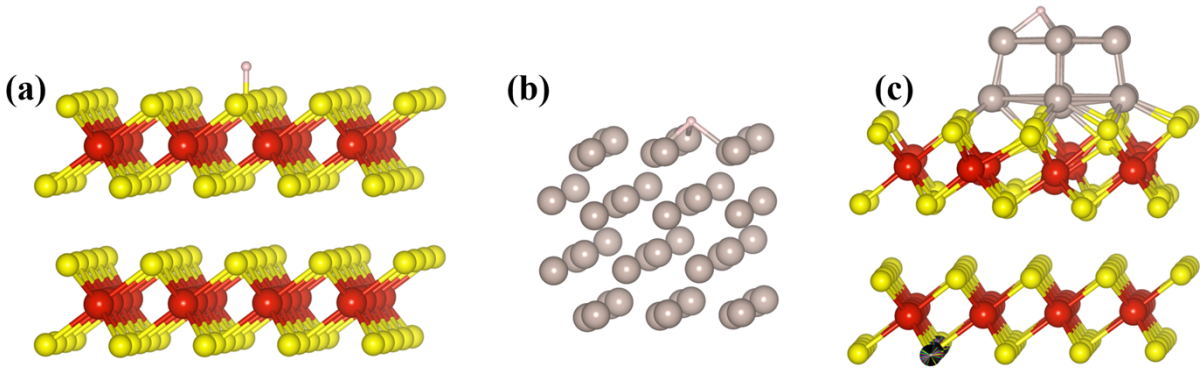

Fig. S10. Optimized structure models of H* adsorbed on the (a) VS_2, (b) Ru and (c) Ru/VS_2.

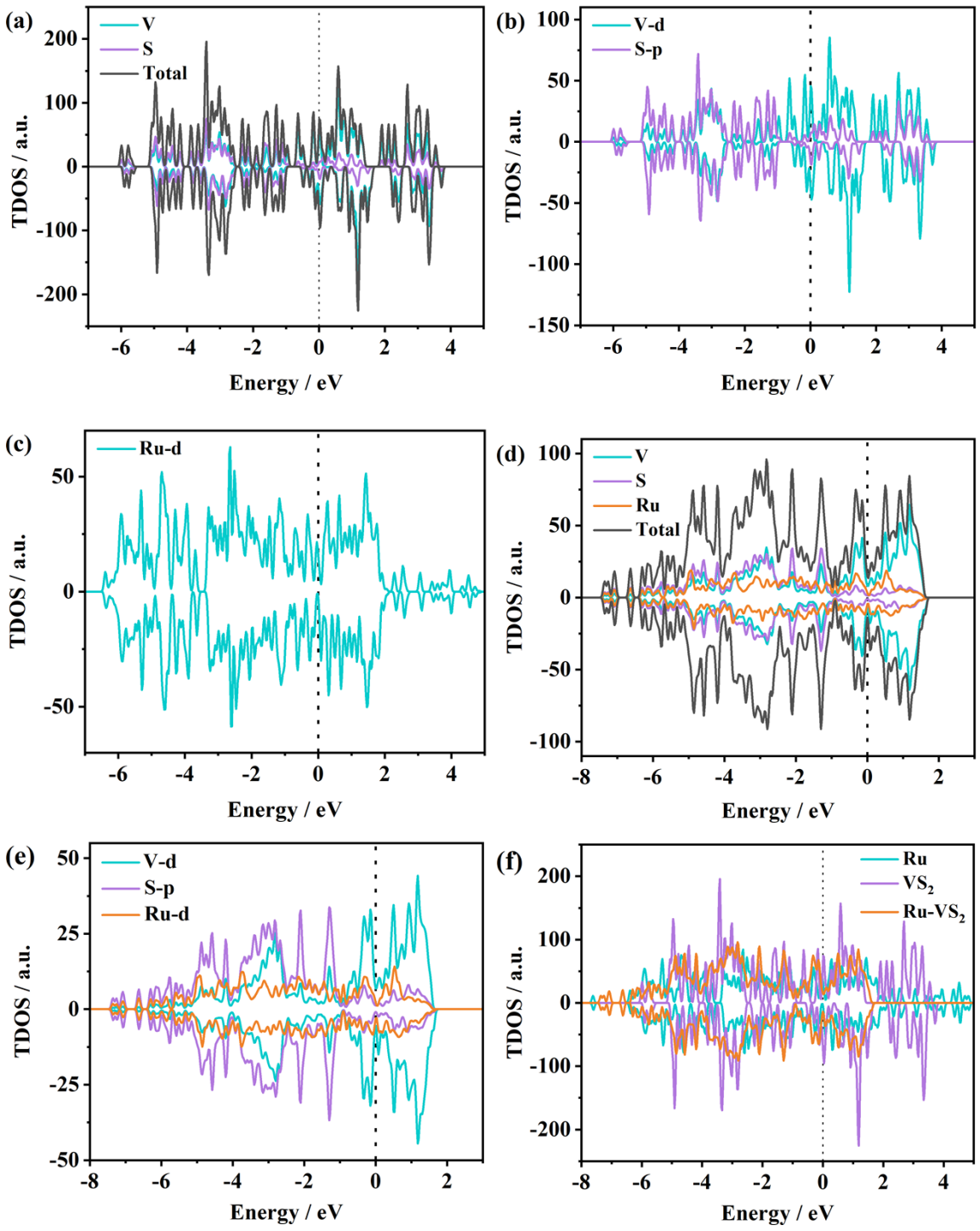

Fig. S11. Calculated density of electronic states of (a-b) VS_2 , (c) Ru, (d-e)Ru/VS_2 and the comparison of the VS_2, Ru and Ru/VS_2.

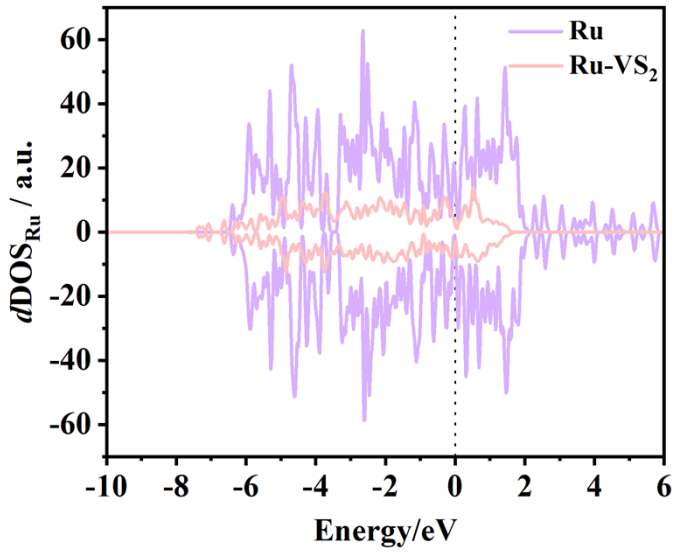

Fig. S12. Density of electronic *d* states of Ru and Ru/VS_2.

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