

Supporting Information for “Formation, electronic properties, and enhanced hydrogen evolution catalytic performance of substitutional defects in 1T PdTe₂”

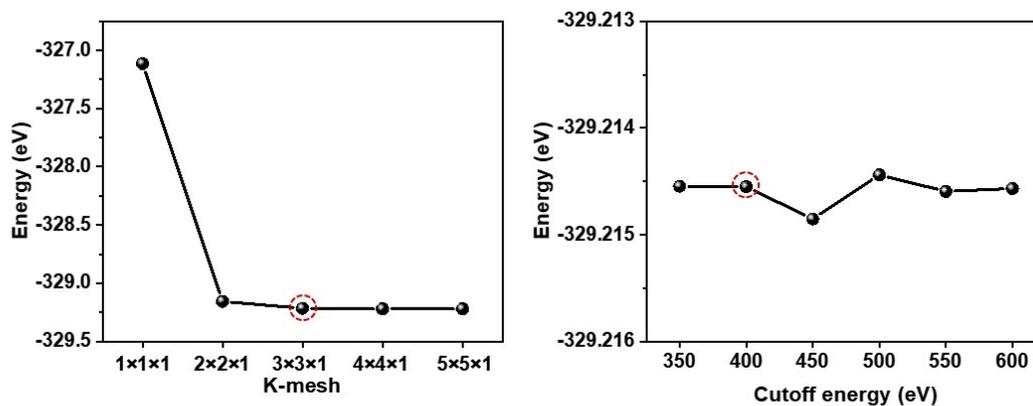


Figure S1. Convergence test on k-points density and cutoff energy.

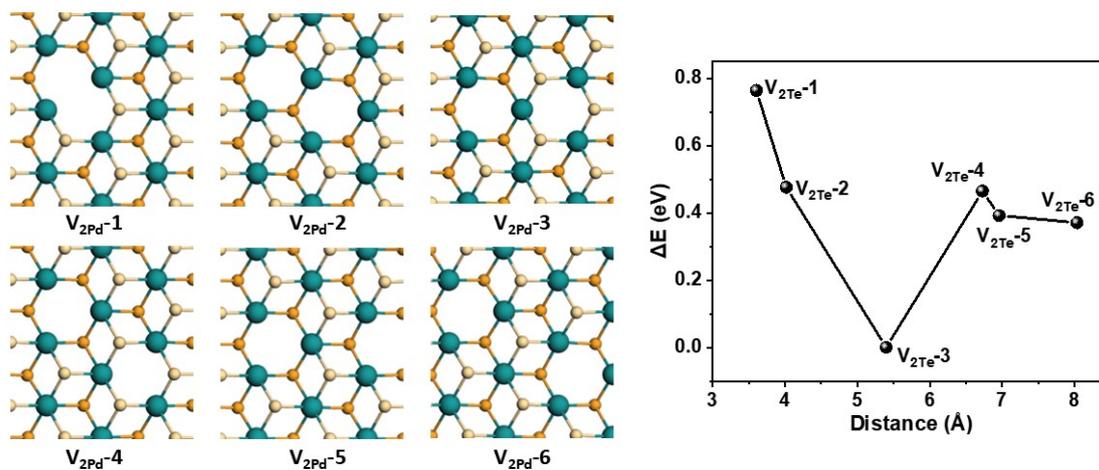


Figure S2. Atomic structures of different Te double vacancies and the relationship between relative energy and distance.

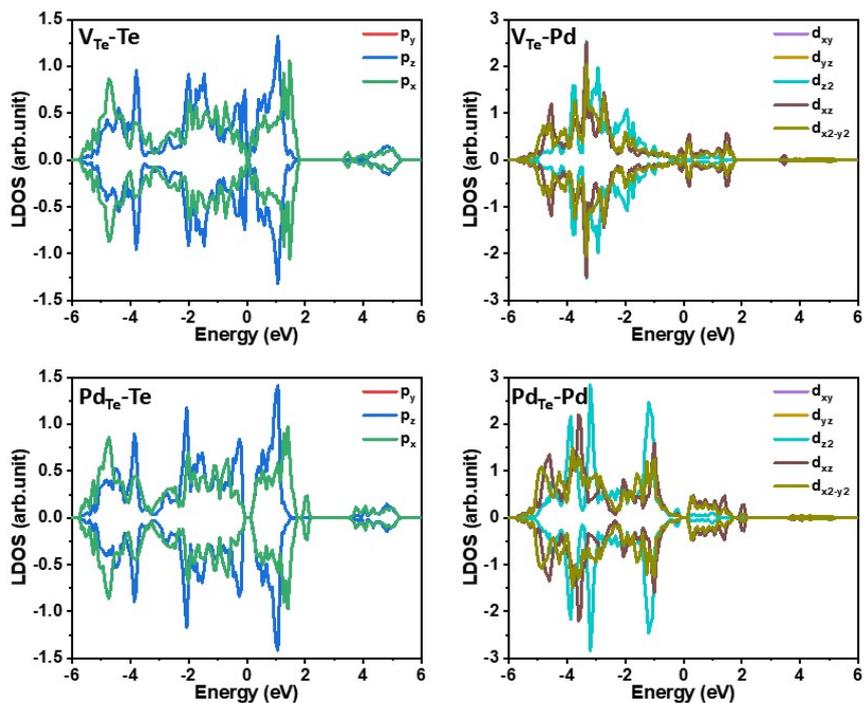


Figure S3. LDOS of the nearest-neighbor atoms around V_{Te} and Pd_{Te} defects.

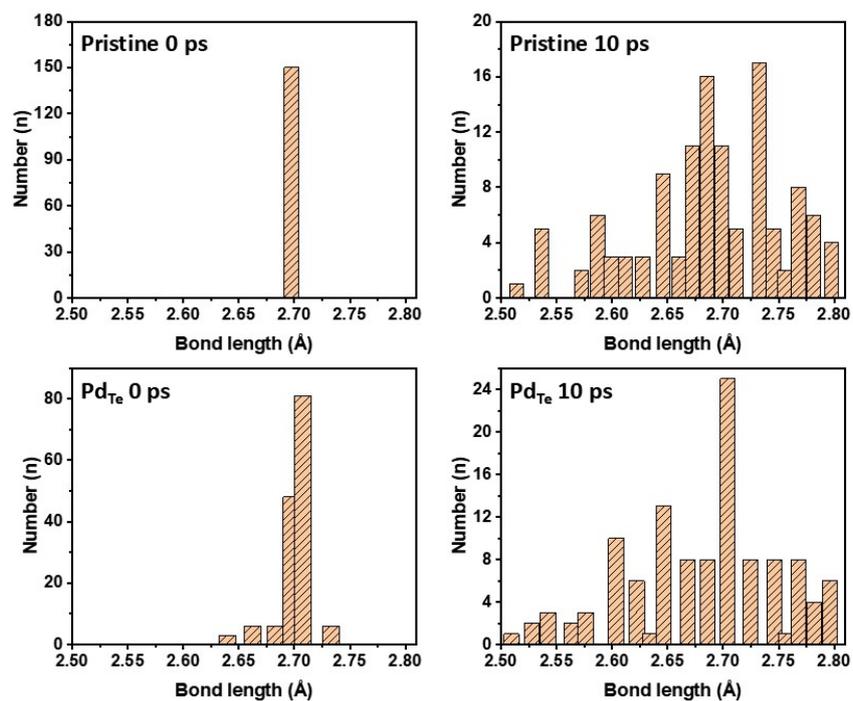


Figure S4. The bond-length evolution of pristine $PdTe_2$ and Pd_{Te} defect at 0 ps and 10 ps of AIMD process.

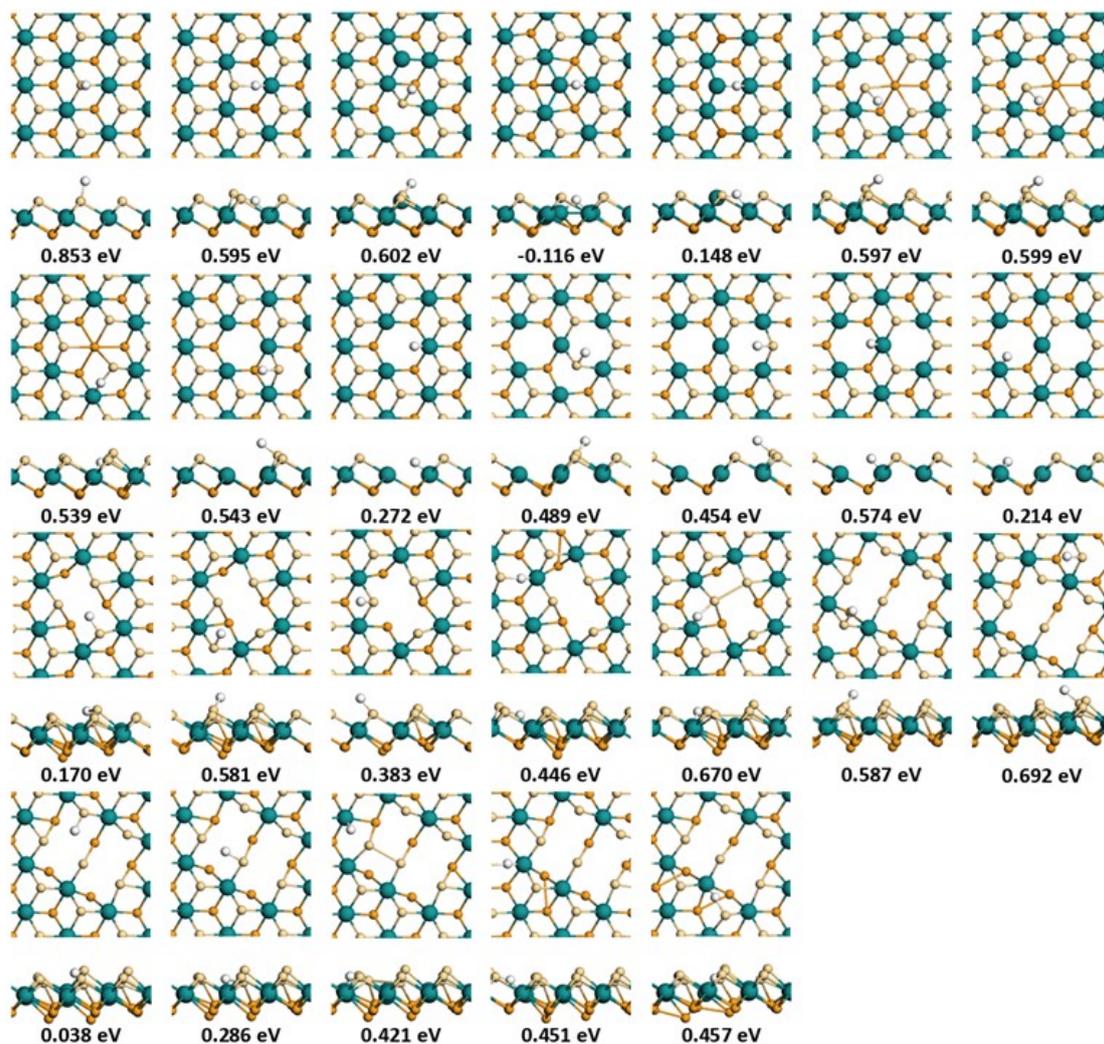


Figure S5. Atomic structures of hydrogen adsorption on various sites and corresponding adsorption energy.

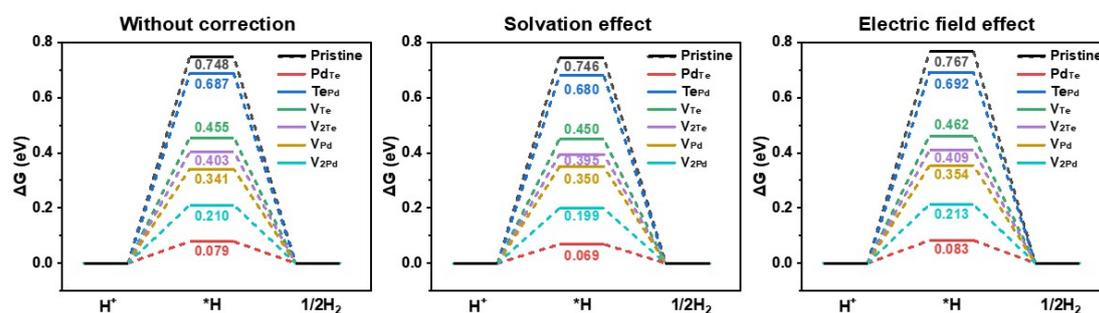


Figure S6. HER results without correction and with solvation effect and electric field effect correction.

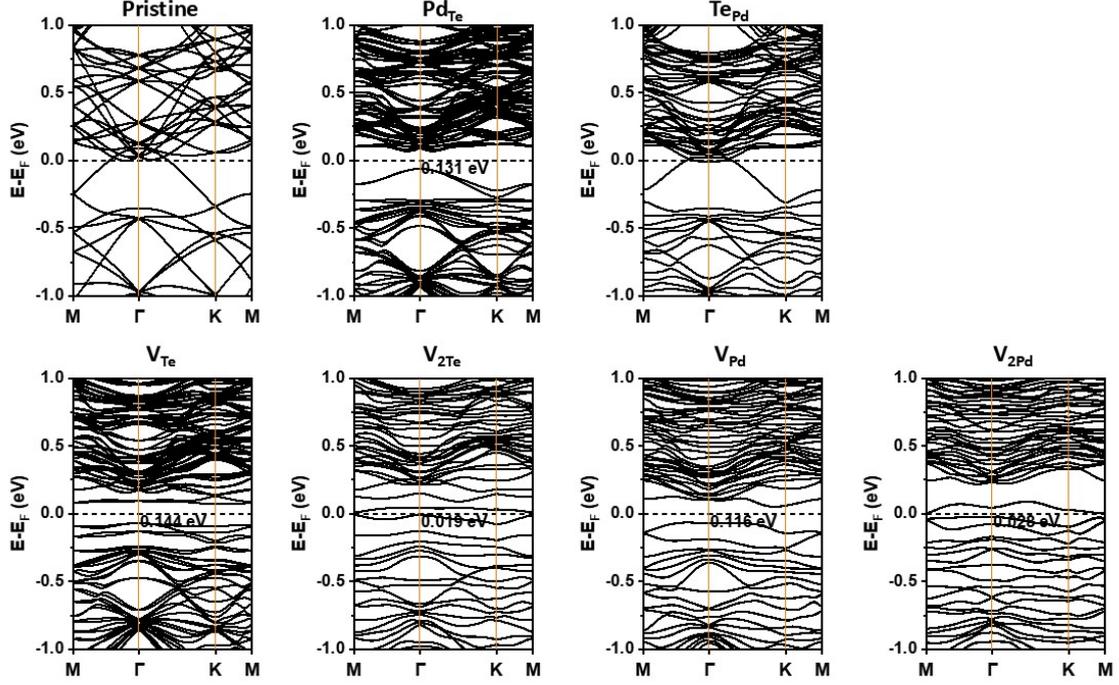


Figure S7. Band structures of pristine and defective PdTe₂ with consideration of spin-orbit coupling.

Table S1. Testing of hydrogen adsorption in different supercells (Unit: eV).

Defect type	Supercell	E_{total}	E_{sub}	E_{ads}
Pristine	4×4×1	-213.448	-210.692	0.626
	5×5×1	-331.991	-329.204	0.595
	6×6×1	-476.152	-473.362	0.592
V_{Te}	4×4×1	-209.189	-206.098	0.291
	5×5×1	-327.642	-324.532	0.272
	6×6×1	-471.907	-468.788	0.263
Pd_{Te}	4×4×1	-214.551	-211.174	0.005
	5×5×1	-333.457	-329.960	-
	6×6×1	-477.722	-474.224	0.117

Table S2. Detailed energy in HER without correction (Unit: eV). The energy of H₂ molecule is -6.763 eV.

Structures	E_{total}	E_{sub}	E_{ads}	ΔE_{ZPE}	$T\Delta S$	ΔG
Pristine	-331.991	-329.204	0.595	0.161	0.008	0.748
V_{Te}	-327.642	-324.532	0.272	0.194	0.011	0.455
$V_{2\text{Te}}$	-323.362	-320.194	0.214	0.199	0.010	0.403
V_{Pd}	-324.799	-321.587	0.170	0.186	0.015	0.341
$V_{2\text{Pd}}$	-317.564	-314.220	0.038	0.186	0.014	0.210

Pd _{Te}	-333.457	-329.960	-	0.202	0.007	0.079
			0.116			
Te _{Pd}	-328.801	-325.958	0.539	0.158	0.010	0.687

Table S3. Detailed energy in HER with correction of solvation effect (Unit: eV). The energy of H₂ molecule is -6.756 eV.

Structures	E _{total}	E _{sub}	E _{ads}	ΔE _{ZPE}	TΔS	ΔG
Pristine	-331.821	-329.036	0.593	0.161	0.008	0.746
V _{Te}	-327.473	-324.362	0.267	0.194	0.011	0.450
V _{2Te}	-323.202	-320.030	0.206	0.199	0.010	0.395
V _{Pd}	-324.659	-321.460	0.179	0.186	0.015	0.350
V _{2Pd}	-317.453	-314.102	0.027	0.186	0.014	0.199
Pd _{Te}	-333.291	-329.787	-0.126	0.202	0.007	0.069
Te _{Pd}	-328.632	-325.786	0.532	0.158	0.010	0.680

Table S4 Detailed energy in HER with correction of electric-field effect (Unit: eV). The energy of H₂ molecule is -6.774 eV.

Structures	E _{total}	E _{sub}	E _{ads}	ΔE _{ZPE}	TΔS	ΔG
Pristine	-332.025	-329.252	0.614	0.161	0.008	0.767
V _{Te}	-327.680	-324.572	0.279	0.194	0.011	0.462
V _{2Te}	-323.395	-320.228	0.220	0.199	0.010	0.409
V _{Pd}	-324.834	-321.630	0.183	0.186	0.015	0.354
V _{2Pd}	-317.603	-314.257	0.041	0.186	0.014	0.213
Pd _{Te}	-333.503	-330.004	-	0.202	0.007	0.083
			0.112			
Te _{Pd}	-328.839	-325.996	0.544	0.158	0.010	0.692