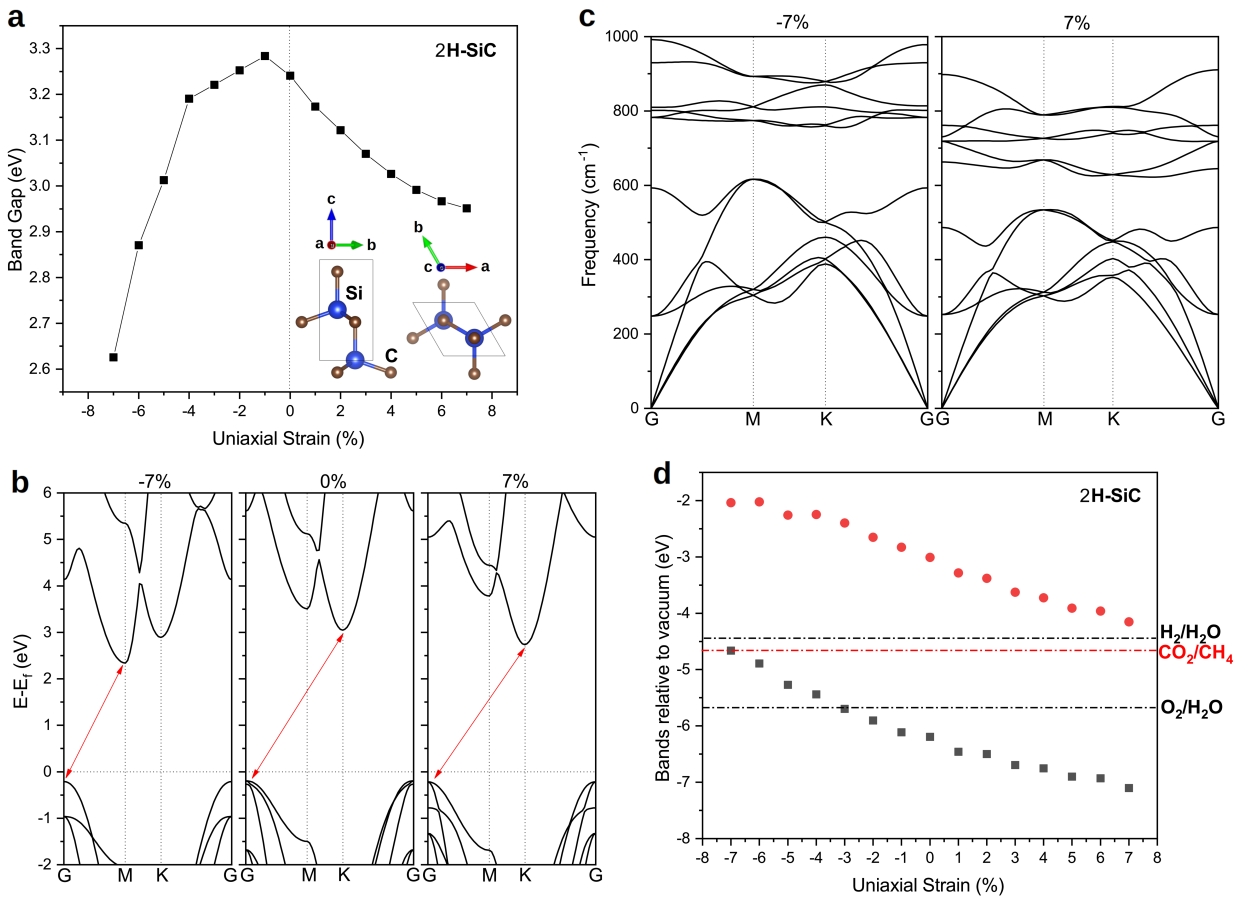
Formula	mp − ID	α_η (eV)	Formula	mp − ID	α_η (eV)
$KNbO_3$	4342	11.6	BN	604884	5.70
$BaTiO_3$	5986	8.26	PNF_2	560008	5.24
$NaNbO_3$	4681	7.85	HCl	632326	5.18
KIO_3	552729	7.72	$KBrO_3$	22958	4.97
LaN	567290	4.69	BrF_5	27987	4.53
PtF_4	8943	4.49	**ZnO**	**2133**	**4.07**
BaI_2	568536	4.14	**PdF_4**	**13868**	**3.90**
BiTeCl	28944	3.38	**CoO**	**19128**	**3.82**
BrF_3	23297	3.31	SiO_2	554089	2.91
SeBr	570589	2.69	Si_2N_2O	4497	2.39
KNO_3	6920	2.62	SbF_3	1880	1.99
Na_2O_2	2340	2.28	**$ZnCl_2$**	**22889**	**1.88**
TlF	558134	2.14	**CdS**	**672**	**1.83**
Ge_2N_2O	4187	1.87	GeO_2	223	1.55
BiTeI	22965	1.84	**$ScCuS_2$**	**6980**	**1.52**
GaN	804	1.75	**KZnSb**	**7438**	**1.50**
AgI	22894	1.73	NaHS	36582	1.44
MgTe	1039	1.67	**KZnAs**	**15687**	**1.29**
PI_3	27529	1.49	H_2S	33024	1.26
GaP	8882	1.11	**Rb_2Se_3**	**7447**	**1.05**
SiC	7140	1.05	$LiMgH_3$	23736	1.01

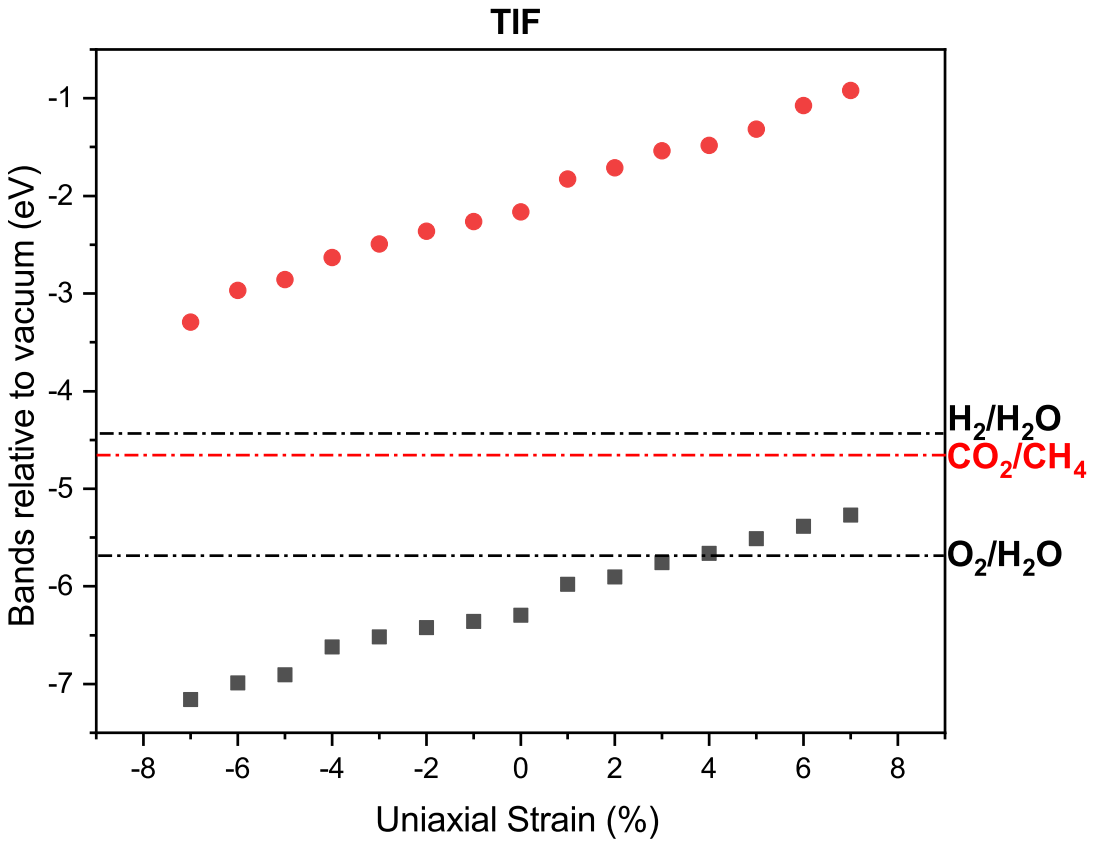
Supplementary Table I: Materials that according to the Materials Project (MP) database posses $\alpha_\eta \equiv \frac{c}{\epsilon} e_{33}$ values larger than 1 V. Compounds listed in the left (right) column fulfill (do not fulfill) the condition of $1.23 \lesssim E_g \lesssim 3.0$ eV (according to the MP database). Materials with $E_g \lesssim 1.23$ eV are marked with bold letters in the right column.

Formula	c (Å)	ϵ (ϵ_0)	$\lvert e_{33}\rvert$ ($\mathrm{C}\cdot\mathrm{m}^{-2}$)	$\alpha_\eta^{\mathrm{rev}}$ (V)
KIO_3	6.71	10.85	1.93	13.46
$KNbO_3$	4.04	11.84	2.80	10.81
LaN	4.68	17.45	3.19	9.67
PtF_4	8.21	5.94	0.56	8.80
$BaTiO_3$	4.00	27.08	4.81	8.03
CdS	5.00	1.06	0.14	7.69
SeBr	8.75	1.81	0.13	7.00
$ScCuS_2$	4.43	4.92	0.40	4.10
ZnO	3.90	6.45	0.59	4.07
PI_3	7.02	8.91	0.41	3.56
BiTeCl	7.06	7.61	0.34	3.52
GaN	3.89	9.65	0.74	3.35
MgTe	5.51	7.75	0.28	2.22
AgI	5.52	6.81	0.22	1.99
BiTeI	5.19	10.42	0.35	1.96
$2H-SiC$	3.73	10.19	0.38	1.57

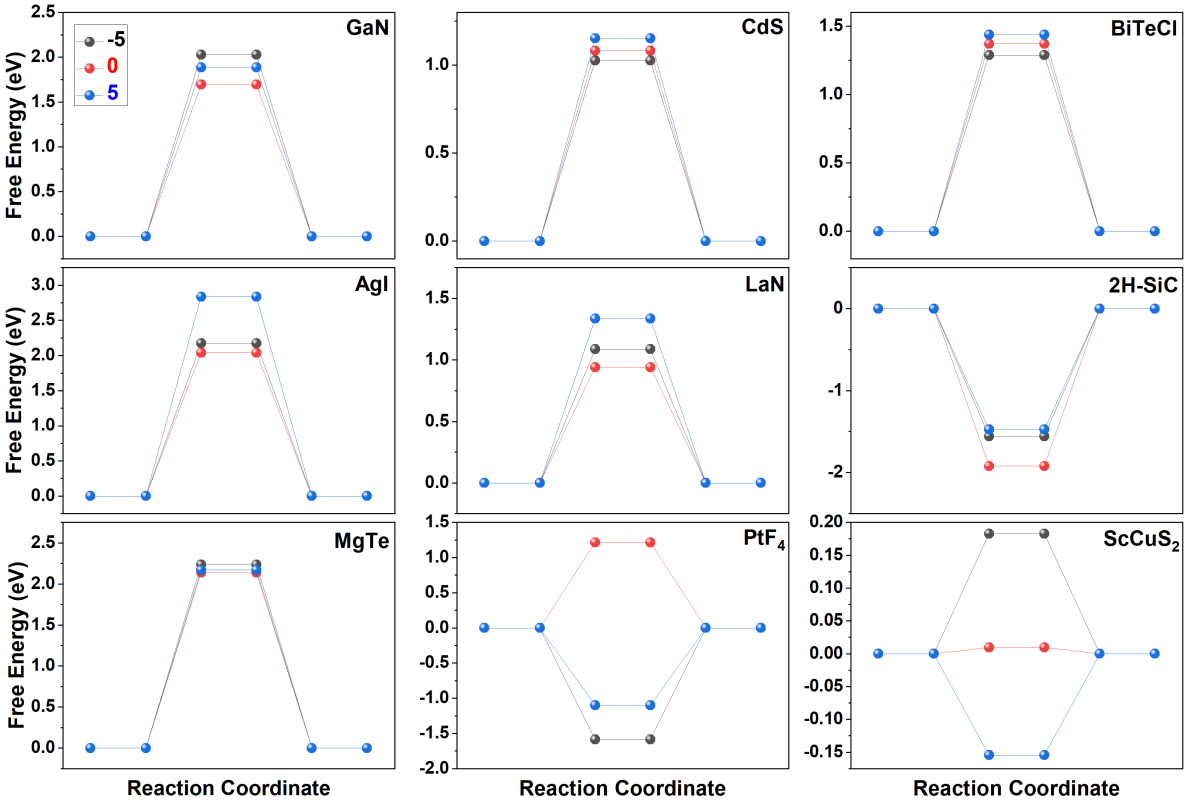
Supplementary Table II: Additional properties of the potential piezo-photocatalysts for water splitting under visible light determined based on the refinement of the DFT data employed in our initial first-principles high-throughput screening. Compounds are ranked according to their revised piezoelectric voltage coefficient, $\alpha_\eta^{\mathrm{rev}}$.



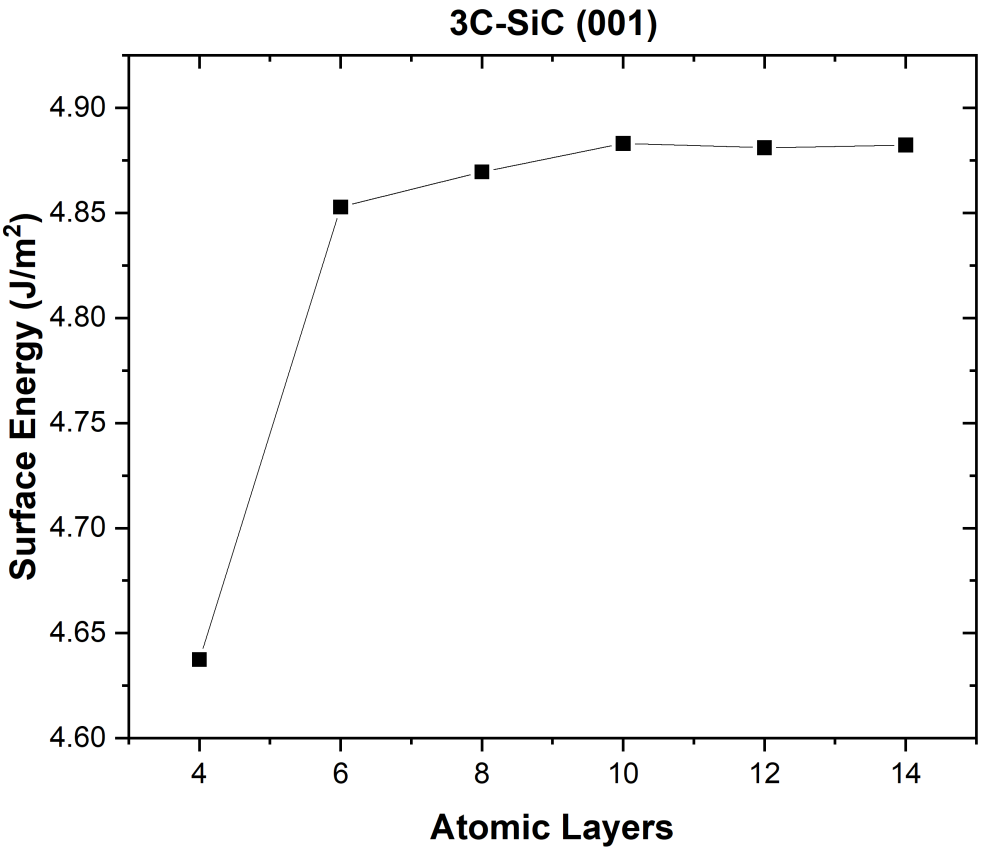
Supplementary Figure 1: First-principles analysis of the optoelectronic, vibrational, and band alignment properties of piezoelectric 2H-SiC. **a** Band-gap variation induced by uniaxial strain. **b** Dependence of the electronic band structure on uniaxial strain and reciprocal space point. **c** Phonon frequencies estimated as a function of uniaxial strain and reciprocal space point. **d** Effect of uniaxial strain on the band alignments of the crystal as referred to the vacuum level. The VBT and CBB energy levels are represented with solid grey squares and red circles, respectively.



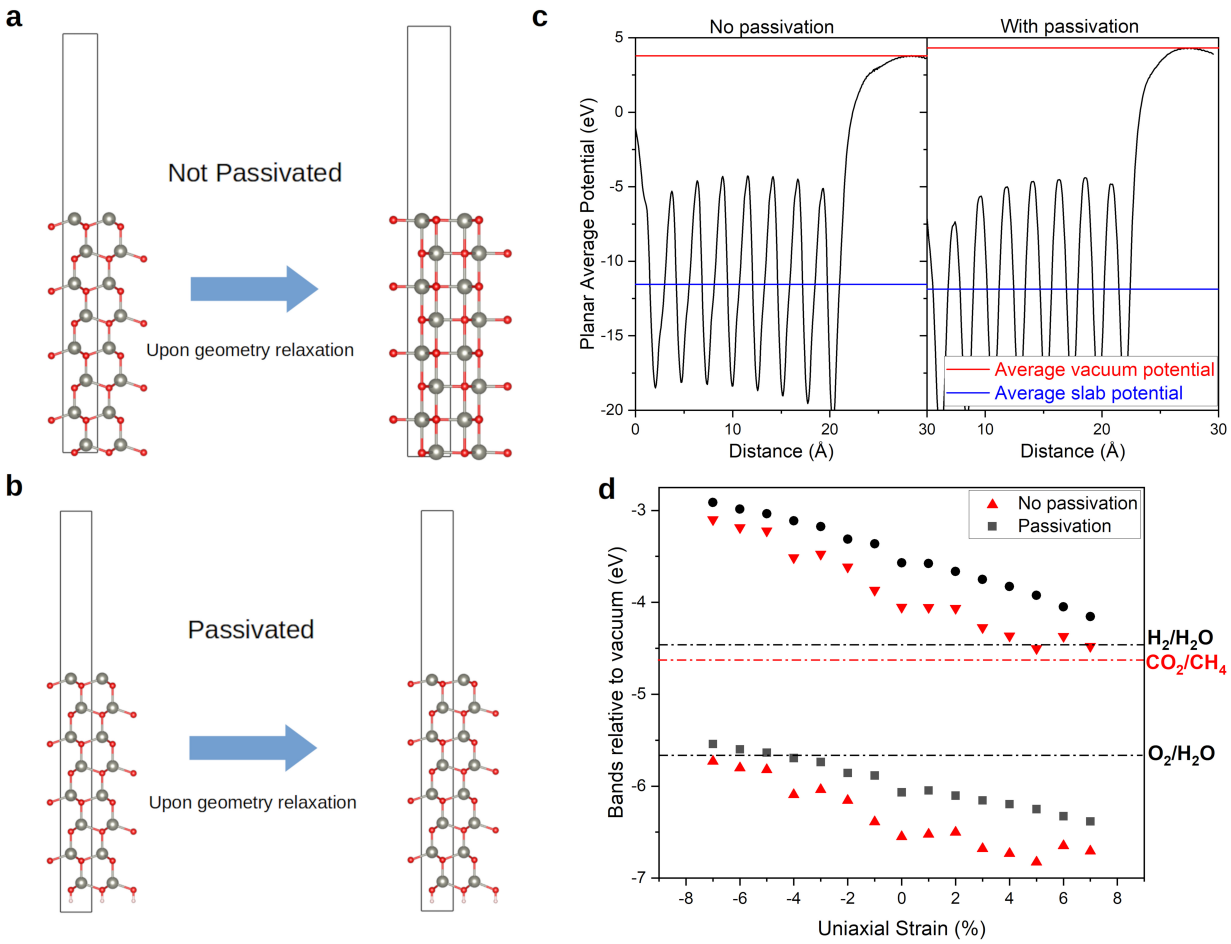
Supplementary Figure 2: Effect of uniaxial strain on the band alignments of TlF as referred to the vacuum level. TlF is an anomalous piezoelectric in the sense that it possesses negative piezoelectric coefficient values (i.e., $e_{33} < 0$). The VBT and CBB energy levels are represented with solid grey squares and red circles, respectively. The η-induced VBT and CBB variations are opposite in sign to those found in standard piezoelectrics exhibiting positive piezoelectric coefficient values (e.g., SiC and ZnO in the main text).



Supplementary Figure 3: Strain engineering of the hydrogen evolution reaction (HER) activity of the screened photocatalytic materials. First-principles estimation of the adsorption free energy of hydrogen atoms on the materials surface.



Supplementary Figure 4: First-principles estimation of the surface energy of a semiconductor SiC slab as a function of its thickness. Sufficiently well converged surface energy results, that is, accurate within 0.01 J/m^2, are already obtained for 8 atomic layers, which approximately are equal to a lenght of 2.0 nm.



Supplementary Figure 5: H-passivation scheme used for the wurtzite ZnO slabs. **a** ZnO slab without H passivation; after geometry optimization, the structural traits change drastically. **b** ZnO slab with H passivation; after geometry optimization, the original structural traits are conserved. **c** Comparison of the planar average potential calculated for the ZnO slabs with and without H passivation. **d** Comparison of the band alignments calculated for the ZnO slabs with and without H passivation.