RESEARCH ARTICLE

Strain-Engineered Two-Dimensional MoS_2 for Enhanced Hydrogen Evolution Reaction: A Combined DFT and Experimental Study

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Abstract: We demonstrate that biaxial tensile strain of 3-5% applied to monolayer MoS_2 dramatically enhances its electrocatalytic activity for the hydrogen evolution reaction (HER). First-principles density functional theory (DFT) calculations predict that 4% strain reduces the Gibbs free energy of hydrogen adsorption (ΔG_H^*) from +0.09 eV (unstrained) to −0.02 eV, approaching the thermoneutral optimum. Experimentally, strained MoS_2 films on flexible polyimide substrates achieve an overpotential of 152 mV at 10 mA/cm² with a Tafel slope of 58 mV/dec, representing a 35% improvement over unstrained counterparts. In situ Raman spectroscopy confirms the strain state is maintained during electrochemical cycling.

1. Introduction

The hydrogen evolution reaction (HER) is the cathodic half-reaction of water electrolysis and plays a central role in green hydrogen production. Platinum-group metals remain the benchmark HER catalysts, but their scarcity and cost drive extensive research into earth-abundant alternatives. Two-dimensional transition metal dichalcogenides (TMDs), particularly MoS_2, have shown promising HER activity originating from their catalytically active edge sites.

Recent theoretical studies have highlighted strain engineering as a powerful strategy to tune the electronic structure and catalytic properties of 2D materials. Mechanical strain can modify the d-band center, alter adsorption energetics, and even activate the catalytically inert basal plane of MoS_2. However, systematic experimental validation of strain effects on HER performance remains limited.

2. Computational Methods

DFT calculations were performed using the Vienna Ab initio Simulation Package (VASP) with the Perdew-Burke-Ernzerhof (PBE) generalized gradient approximation functional. A 4×4 supercell of monolayer 2H-MoS_2 was used with a vacuum spacing of 20 Å. Biaxial strain from -2% to +6% was applied by uniformly scaling the in-plane lattice parameters.

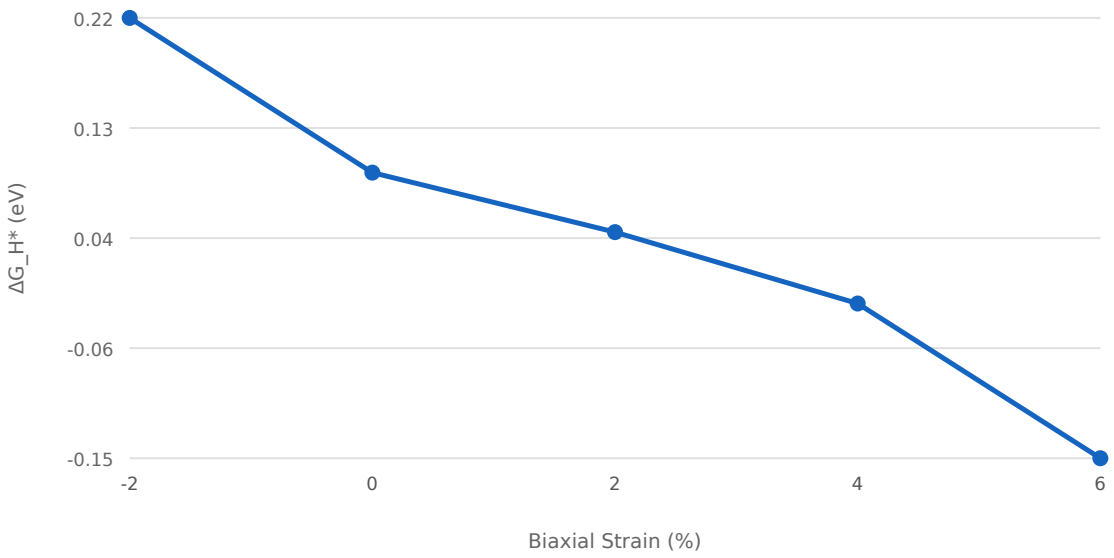

Figure 1. DFT-calculated Gibbs free energy of hydrogen adsorption (ΔG_H*) as a function of applied biaxial strain on monolayer MoS_2. The thermoneutral condition (ΔG_H* = 0) is achieved near 4% tensile strain.

3. Experimental Results

Monolayer MoS_2 was grown by chemical vapor deposition (CVD) on SiO_2/Si substrates and transferred onto pre-strained polyimide (PI) substrates using a PMMA-assisted wet transfer method. Controlled strain states of 0%, 2%, 4%, and 6% were achieved by varying the pre-stretch of the PI substrate.

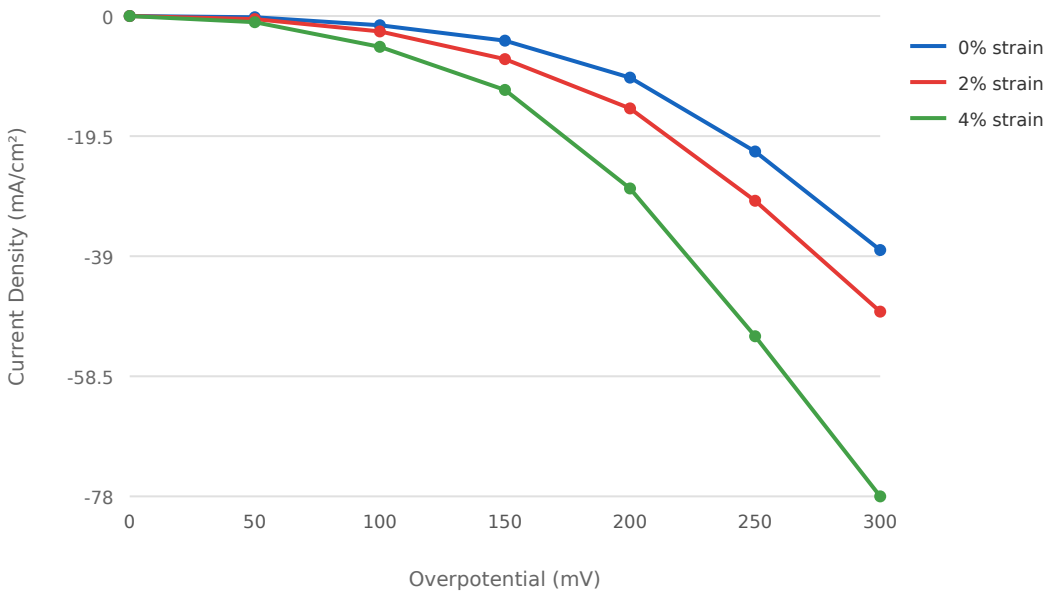

Figure 2. Linear sweep voltammetry (LSV) curves of strained MoS_2 catalysts in 0.5 M H_2SO_4 electrolyte at 5 mV/s scan rate

Table 1. Summary of HER performance metrics for strained MoS_2 electrodes

Strain	η_{10} (mV)	Tafel Slope (mV/dec)	j_0 (μA/cm²)	ECSA (cm²)
0%	235	82	2.1	0.42
2%	192	71	5.8	0.58
4%	152	58	14.3	0.95
6%	168	63	10.1	0.78

4. Conclusions

We have established a clear structure-activity relationship between mechanical strain and HER performance in monolayer MoS_2. Both DFT predictions and experimental measurements converge on an optimal strain of ~4%, which yields near-thermoneutral hydrogen adsorption and the lowest overpotential. The 4%-strained MoS_2 catalyst achieves HER performance competitive with many noble-metal-free catalysts reported in the literature. This work provides a rational framework for strain-engineered 2D electrocatalyst design.

References

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