

Rolltronics-enabled MoS₂ nanoscroll for efficient hydrogen evolution

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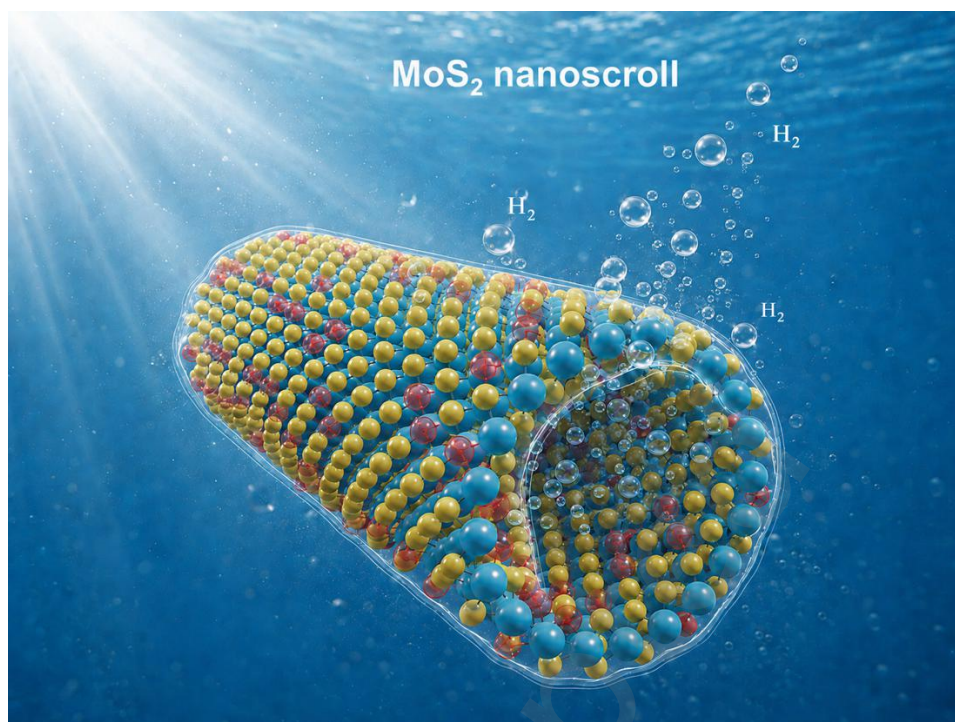
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This paper reports a rolltronics strategy which converts defect-engineered MoS₂ nanosheets into stable metallic 1T-MoS₂ nanoscrolls through sulfur-vacancy-induced asymmetric stress. The resulting curved, hollow architecture integrates abundant active sites, rapid charge transfer, optimized hydrogen adsorption, and enhanced mass transport, enabling efficient and durable dye-sensitized photocatalytic hydrogen evolution with markedly improved activity.

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ABSTRACT: Rolltronics represents an emerging curvature engineering paradigm utilizing van der Waals nanoscrolls to tailor material properties beyond planar twistrionic systems. Rolltronics modulation holds great promise as a new platform to advance the hydrogen evolution reaction (HER) activity of MoS₂. Here, we designed 1T-MoS₂ nanoscrolls via asymmetric stress induced by the different defect densities on opposite surfaces of MoS₂ sheets to serve as highly efficient HER catalyst. In the dye-sensitized photocatalytic system with Eosin Y (EY) serving as the photosensitizer, and triethanolamine (TEOA) acts as the sacrificial electron donor to regenerate EY and sustain the photocatalytic cycle, the 1T-MoS₂ nanoscrolls exhibited extraordinary HER activity with 24.1 mmol g⁻¹ h⁻¹ under UV-visible light, which is nearly 25 times higher than that of the semiconducting pristine 2H-MoS₂ nanosheets. This superior activity originates from the synergistic effects of abundant active sites exposed by S-vacancies, enhanced charge transfer from the metallic 1T phase, and optimized hydrogen adsorption free energy (ΔG_{H^+}). The improved HER activity of 1T-MoS₂ nanoscrolls can be mainly attributed to the metallic conductive characteristic of the 1T phase, abundant S-vacancy-derived active sites, curvature-modulated hydrogen adsorption energy, as well as facilitated mass transfer within the tubular architecture. This rolltronics engineering strategy provides a viable pathway for designing low-cost and highly efficient transition metal dichalcogenide for large-scale energy applications.

KEYWORDS: rolltronics; MoS₂ nanoscrolls; S vacancy; 1T phase; hydrogen evolution

1 Introduction

Two-dimensional (2D) molybdenum disulfide (MoS₂) has emerged as a highly promising, non-precious co-catalyst for the photocatalytic hydrogen evolution reaction (HER). Despite its favorable thermodynamic properties, the practical catalytic efficiency of pristine 2H-MoS₂ is intrinsically limited by its semiconducting nature and the resulting poor electrical conductivity[1, 2]. Furthermore, the catalytic activity of pristine MoS₂ is severely limited by the scarcity of surface active sites, as the basal plane is largely inert and hydrogen adsorption primarily occurs at the sparsely distributed active sites. Therefore, to accelerate the overall catalytic HER kinetics, it is imperative to develop synergistic strategies that maximize the density of accessible active sites while promoting the efficient spatial separation and transfer efficiency of electrons.

To address these bottlenecks, extensive efforts have been dedicated to phase and defect engineering[3, 4]. Inducing a structural phase transition from the semiconducting phase to the metallic phase dramatically enhances the intrinsic electrical conductivity, thereby facilitating rapid electron transfer. Conventional strategies for inducing the 2H to 1T phase transition mainly rely on chemical synthesis, intercalation-assisted exfoliation, electrochemical reduction, microwave irradiation, plasma treatment etc. However, a critical dilemma persists in practical catalytic systems, 1T-MoS₂ nanosheets are thermodynamically metastable and highly susceptible to severe face-to-face van der Waals

restacking[5-7]. Moreover, the resulting planar nanosheets readily restack under interlayer van der Waals interactions and revert to the thermodynamically stable 2H phase[8-10].

Recently, twistrionics, which manipulates interlayer twist angles, has demonstrated remarkable capability in tailoring the physicochemical properties of two-dimensional (2D) materials[11-13]. As a complementary and emerging paradigm beyond twistrionics, rolltronics has opened a unique curvature-engineering avenue for property modulation of van der Waals materials[14-17]. By constructing tubular nanoscroll architectures, rolltronic engineering integrates geometric curvature, intrinsic strain, and confined hollow channels, offering unprecedented opportunities to simultaneously regulate electronic conductivity, expose abundant active sites, optimize hydrogen adsorption free energy, and accelerate mass transport[18-21]. Various approaches have been employed to construct 1D MoS₂ nanoscrolls from 2D nanosheets, such as strain-induced scrolling, hydrothermal synthesis, alkaline-assisted processing, organic-solvent-induced scrolling, etc.[22-24]. Nevertheless, the rational fabrication of high-quality phase-controlled MoS₂ nanoscrolls via a facile defect-induced rolling strategy remains underexplored for high-efficiency photocatalytic HER.

In this work, we propose a facile asymmetric stress strategy derived from discrepant surface defect densities to fabricate metallic 1T-phase MoS₂ nanoscrolls. Benefiting from the synergistic advantages of metallic 1T conductivity, sulfur vacancy-enriched active sites, curvature-optimized

hydrogen adsorption thermodynamics, and tubular channel-facilitated mass transfer, the as-prepared 1T-MoS₂ nanoscrolls deliver exceptional catalytic HER performance. Specifically, the optimized catalyst achieves a prominent hydrogen evolution rate of 24.1 mmol g⁻¹ h⁻¹ under UV-visible light irradiation, which is nearly 25-fold higher than that of pristine 2H-MoS₂ nanosheets. This work demonstrates that rolltronic curvature engineering serves as a reliable and versatile platform for designing high-efficiency TMD-based catalysts, providing a feasible pathway for the development of low-cost and scalable energy conversion materials.

2 Experimental section

2.1 Chemicals

All reagents and materials were obtained commercially from the Beijing Chemical Reagent (Beijing, China) and used as received without further purification.

2.2 Synthesis of MoS₂ nanosheets

Typically, hexaammonium heptamolybdate tetrahydrate (1mmol, (NH₄)₆Mo₇O₂₄·4H₂O, i.e. 7 mmol Mo) and thiourea (30 mmol) were dissolved in deionized water (35 mL) under vigorous stirring to form a homogeneous solution. Then, the solution was transferred into a 45 mL Teflon-lined stainless steel autoclave and maintained at 220 °C. After 18 h, the reaction system was allowed to cool to room temperature. The final product was washed with water and absolute ethanol for several times to remove any possible ions, and dried at 60 °C under vacuum. All the materials were purchased from SinoPharm and used without further purification.

2.3 Preparation of MoS₂ nanoscrolls

MoS₂ nanosheets were dispersed in 120 mL of a mixed solvent containing N-methyl-2-pyrrolidone (NMP) and water at a volume ratio of 2:1. The suspension was transferred into a titanium vessel, sealed under ambient atmosphere, and maintained at 20 ± 5°C using a circulating water bath. High-intensity ultrasonication was then applied for 6 h. The resulting black precipitate was collected and sequentially washed several times with deionized water and absolute ethanol. Finally, the product was vacuum-dried at 40°C for 10 h.

2.4 Characterization

The morphology and micro-structure of the as-prepared materials were characterized via field emission scanning electron microscopy (SEM, Hitachi S-4800), transmission electron microscopy (TEM, JEM-3010), high angle annular dark field-scanning TEM (HAADF-STEM, FEI Tecnai G2 F20 S-TWIN). X-ray photoemission spectroscopy (XPS) was measured on a Thermo Scientific K-Alpha instrument (USA) system with a monochromatic Al K α source. Raman spectroscopy was characterized on a Thermo Fischer DXR.

2.5 Photocatalytic HER

Photocatalytic hydrogen evolution activities were evaluated under UV-visible light irradiation using Eosin Y (EY) as the photosensitizer and triethanolamine (TEOA) as the sacrificial electron donor. All measurements were conducted

in a sealed quartz reactor under continuous magnetic stirring at ambient temperature. Typically, 10 mg of the as-prepared MoS₂ catalyst was dispersed in 100 mL of aqueous solution containing 10 vol% TEOA and 20 mg EY. Prior to irradiation, the reaction system was degassed with argon for 30 min to thoroughly eliminate dissolved oxygen. A 300 W xenon lamp (PLS-SXE300+/UV, Beijing Perfectlight Technology Co., Ltd.) was employed as the irradiation source. The generated H₂ was quantified in real time using a LabSolar- III AG online photocatalytic analysis system (Beijing Perfectlight Technology Co., Ltd.) coupled with a GC9790 gas chromatograph (Zhejiang Fuli Analytical Instruments Co., Ltd., China) equipped with a thermal conductivity detector (TCD) with argon as the carrier gas.

2.6 Computational details

All MD calculations were carried out using the large-scale atomic-molecular massively parallel simulator (LAMMPS) software package. To investigate the structural stability of MoS₂ nanoscrolls in aqueous environment, all MD simulations were performed using LAMMPS under an NVT ensemble at 300 K with a hybrid REBO-Lennard-Jones force field, and then production simulations were conducted with a time step of 1 fs for a total of 5×10⁵ steps, with atomic trajectories recorded for structural stability and water dynamics analysis. Periodic boundary conditions (PBCs) were applied in all three orthogonal directions to eliminate finite-size effects.

All electronic structure calculations were performed within the framework of density functional theory (DFT) as implemented in the Vienna Ab initio Simulation Package (VASP). The exchange-correlation interactions were treated by generalized gradient approximation (GGA) parameterized by Perdew, Burke, and Ernzerhof (PBE). We describe the interaction between ions and electrons using the projected augmented wave (PAW). The periodic unit was optimized by the conjugate gradient algorithm with an energy cutoff of 500 eV until the force on each atom was less than 0.01 eV/Å. A vacuum region of 20 Å was added to avoid the interactions between adjacent images. The Brillouin zone was sampled by Monkhorst-Pack method with 5 × 5 × 1 k-point grid. The adsorption energy is defined as

$$\Delta E_H = E(\text{MoS}_2 + \text{H}) - E(\text{MoS}_2) - \frac{1}{2}E(\text{H}_2)$$

(1)

where (MoS₂+H) refers to hydrogen adsorbed on the MoS₂ surface, (MoS₂) refers to a clean MoS₂ surface, and H₂ refers to gas phase hydrogen molecule. The hydrogen adsorption free energy was calculated at zero potential and pH = 0 as

$$\Delta G_H^* = \Delta E_H + \Delta E_{\text{ZPE}} - T\Delta S \quad (2)$$

where ΔE_H is the hydrogen adsorption energy, ΔE_{ZPE} is the difference in zero point energy, T is the temperature (300 K) and ΔS is the difference in entropy between H that is adsorbed and in the gas phase at 101325 Pa. A normal mode analysis was used to determine the vibrational frequencies of the adsorbed species, which were used to determine the zero point energy correction and the entropy. The adsorption is too strong if ΔG_H^{*} is very negative or too weak if ΔG_H^{*} is very positive.

Finite element analysis (FEA) was performed using MSC

software to characterize the stress evolution and morphological transition during the defect-driven scrolling of monolayer MoS₂ nanosheets. The MoS₂ monolayer was modelled as a linearly elastic thin film with a thickness of 0.65 nm, a Young's modulus of 270 GPa and a Poisson's ratio of 0.22. To reproduce the asymmetric lattice distortion induced by disparate S-vacancy densities on opposing surfaces of the sheet, a through-thickness gradient of equivalent in-plane strain was prescribed across the film. The computational domain was discretized with four-node shell elements, and periodic boundary conditions were enforced along the axial direction to mimic the extended geometry of the nanoribbon. The von Mises equivalent stress field and the structural evolution from a planar configuration to a tubular scroll were numerically solved to corroborate the strain-energy-release mechanism governing nanoscroll formation.

2.7 Reproducibility and cycling stability tests

Reproducibility and cycling stability of 1T-MoS₂ nanoscrolls for photocatalytic H₂ evolution were assessed through repeated tests under identical conditions. Each run employed 10 mg catalyst, 20 mg EY, and 10 vol% TEOA in 100 mL solution. After Ar purging (30 min), H₂ production was measured under 300 W Xe lamp irradiation using online

GC-TCD.

After each run, the irradiation was stopped and the suspension was centrifuged at 10,000 rpm for 10 min to separate the catalyst from the supernatant. The precipitate was washed with deionized water (three times) and absolute ethanol (once) via centrifugation, with each washing step involving re-dispersion by ultrasonication for 3 min. The washing process was repeated until the supernatant was virtually colorless.

To avoid aggregation during drying, the washed wet catalyst was directly reintroduced into 100 mL of fresh reaction solution and dispersed by bath ultrasonication for 5 min, without using probe-type high-intensity ultrasonication to prevent structural damage. Fresh EY (20 mg) and TEOA (10 vol%) were then added, followed by Ar purging for 30 min. All experimental parameters were maintained consistently across cycles. The recovery–washing–redispersion process was repeated for multiple cycles, with each cycle tested in triplicate; results are presented as mean $\pm$ SD.

3 Results and discussion

3.1 Stable 1T-MoS₂ nanoscrolls

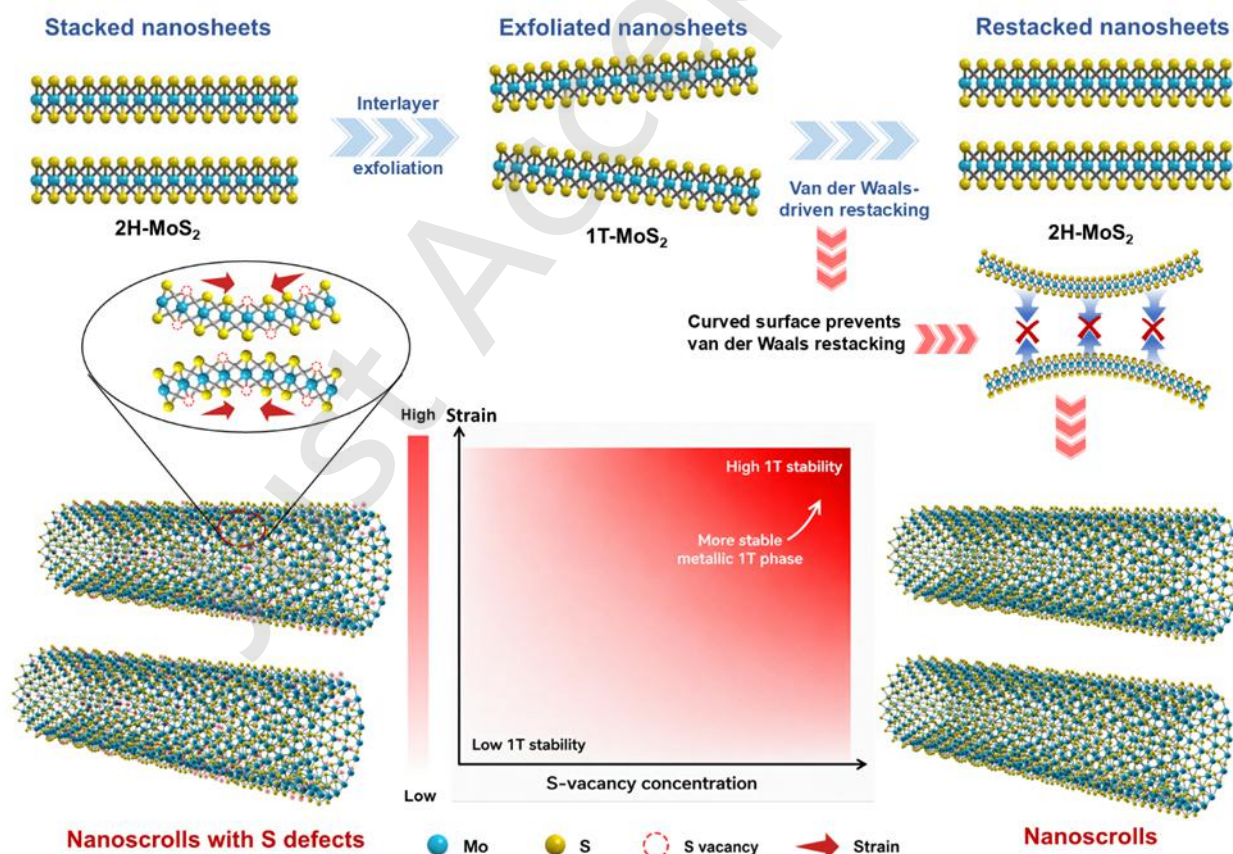


Figure 1 Stabilization mechanism of 1T-MoS₂ nanoscrolls.

Extensive efforts have been devoted to converting 2H-MoS₂ into its metallic 1T phase[25-27]. However, the 1T-MoS₂ is easy to revert to 2H-MoS₂ due to S-S van der Waals interactions[28-30]. Therefore, there is an urgent need to develop an efficient and scalable strategy for producing 1T-MoS₂ with structural stability. As shown in Fig. 1, the

reversion of metastable 1T-MoS₂ to the thermodynamically stable 2H phase is often associated with nanosheet restacking, whereas the curved nanoscroll geometry can restrict extensive face-to-face contact and thereby mitigate compact restacking[24]. The nanoscroll geometry provides an effective structural route to stabilize 1T-MoS₂. Moreover,

both S vacancies and strain diminish the surface energy disparity between 1T and 2H-MoS₂, with increasing strain and S-vacancy concentrations lowering the 1T-phase surface energy and thereby suppressing phase transition[31-33]. This curved topology simultaneously mitigates van der Waals-mediated restacking via reduced face-to-face contact and lowers the surface energy gap between 1T and 2H-MoS₂, yielding stable 1T-MoS₂.

3.2 The formation mechanism of MoS₂ nanoscrolls

To describe the asymmetric force-induced deformation and subsequent scrolling of the MoS₂ monolayers, the interatomic interactions were represented by a hybrid force field combining a many-body reactive empirical bond-order (REBO) potential with a Lennard-Jones (LJ) potential[34, 35]. The total potential energy of the system is expressed as

$$E_{total}=E_b+E_{LJ} \quad (3)$$

where E_{total} is the total potential energy, E_b is the short-range binding energy described by the REBO potential, and E_{LJ} denotes the long-range van der Waals contribution. The short-range interatomic energy is written as

$$E_b=\frac{1}{2}\sum_i\sum_{j(>i)}[V^R(r_{ij})-b_{ij}V^A(r_{ij})] \quad (4)$$

Here, r_{ij} is the distance between atoms i and j ; $V^R(r_{ij})$ and $V^A(r_{ij})$ are the pairwise repulsive and attractive terms, respectively; and b_{ij} is an environment-dependent bond-order function. Because b_{ij} varies with the local coordination, bond angles, and neighboring atomic configuration, the REBO formalism can capture the changes in Mo-S bonding induced by sulfur vacancies, including bond

weakening, breaking, reforming, and local structural relaxation. The repulsive and attractive terms are given by

$$V^R(r)=f^C(r)(1+Q/r)Ae^{-\alpha r} \quad (5)$$

$$V^A(r)=f^C(r)\sum_{n=1}^3B_ne^{-\beta_n r} \quad (6)$$

where $f^C(r)$ is a smooth cutoff function that restricts the REBO interaction to the short-range bonding region. A , Q , α , and B_n are fitted potential parameters that determine the magnitude and distance dependence of the repulsive and attractive interactions[36]. The long-range van der Waals interaction between nonbonded atoms, particularly between different regions of the curved MoS₂ sheet, is described using the LJ potential:

$$E_{LJ}(r)=4\varepsilon \left[\left(\frac{\sigma}{r} \right)^{12} - \left(\frac{\sigma}{r} \right)^6 \right] \quad (7)$$

where r is the interatomic distance, ε is the depth of the potential well, and σ is the distance at which the LJ potential becomes zero. In the present simulation, σ and ε were set to 3.13 Å and 0.00693 eV, respectively. The r^{-12} term represents short-range repulsion arising from electron-cloud overlap, whereas the r^{-6} term accounts for long-range dispersion attraction. Accordingly, the REBO component captures vacancy-induced changes in local bonding and asymmetric stress, while the LJ component describes the van der Waals attraction that develops as separated regions of the nanosheet approach each other during scrolling. The final nanoscroll morphology is therefore governed by the competition among defect-induced stress relaxation, bending-energy cost, and van der Waals adhesion.

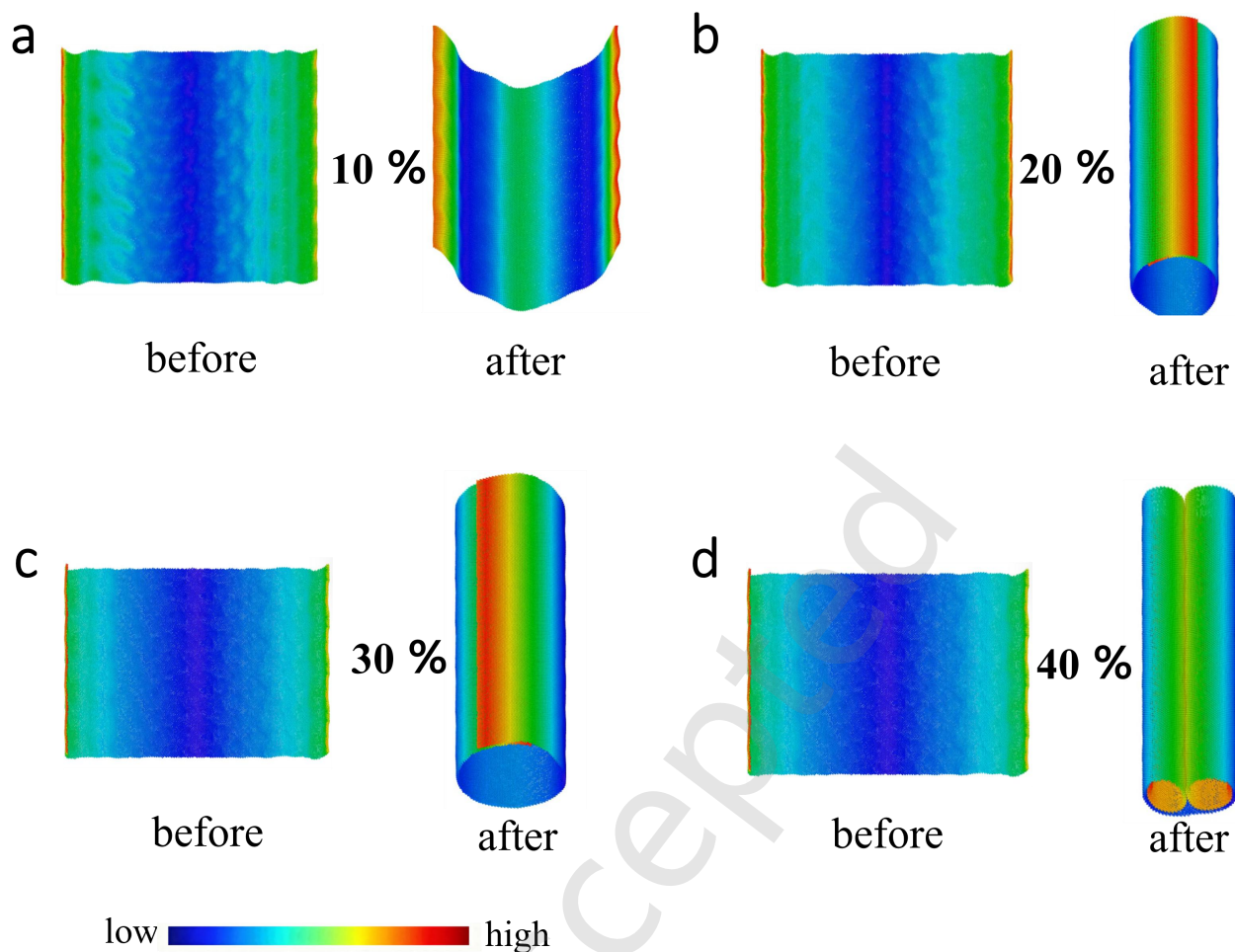


Figure 2 Structural developments of MoS₂ nanosheets before and after MD simulations. Structural morphology at a desulfurization content of (a) 10%, (b) 20%, (c) 30%, (d) 40% before and after MD simulations. All sulfur vacancies are generated on one surface of MoS₂ monolayer sheets by randomly deleting sulfur atoms on one surface of the MoS₂ monolayer. Snapshots of free-standing MoS₂ monolayer sheets are colored on the basis of the relative displacements in the out-of-plane (z) direction.

To further investigate the formation mechanism of MoS₂ nanoscrolls, we employed MD simulations to study the structural evolution of MoS₂ monolayers under different sulfur vacancy densities[37-39]. Movie S1 and Movie S2 illustrate the dynamic scrolling behaviour of MoS₂ nanosheets at different sulfur-vacancy concentrations, and depicts that vacancy-induced asymmetric strain contributes to scrolling[40, 41]. Fig. 2 displays snapshots of MoS₂ nanosheets with desulfurization contents of 10%, 20%, 30%, and 40% before and after MD simulations, where atoms are colored based on their relative atomic displacements along the out-of-plane (z) direction. Evidently, the desulfurization nanosheet to undergo complete self-scrolling behavior. The simulation results demonstrate that the initially flat two-dimensional (2D) nanosheet successfully transformed into a structurally intact one-dimensional (1D) nanoscroll. At this stage, a distinct displacement gradient in the z-direction (transitioning from blue at the edges to red at the top) indicates the formation of a regular cylindrical topological structure. Compared to the 20% case, the nanoscroll structure at 30% desulfurization appears more compact, suggesting that the defect-induced internal stress significantly increases with concentration. At a high

content plays a crucial role in determining the structural morphology of the MoS₂ nanosheets. At a desulfurization content of 10%, the MoS₂ nanosheet did not form a closed tubular structure after MD simulation. Since the lattice mismatch and local stress introduced by sulfur vacancies were insufficient to overcome the bending rigidity, the nanosheet exhibited only significant arched bending, representing an intermediate metastable state transitioning toward a U-shape (Fig. 2(a)). When the desulfurization content was increased to 20% (Fig. 2(b)) and 30% (Fig. 2(c)), the surface energy and strain energy within the system reached a specific equilibrium, driving the desulfurization content of 40% (Fig. 2(d)), the excessive S-vacancy concentration causes severe rearrangement of the lattice structure. The simulated structure is no longer a single cylindrical nanoscroll but instead presents a complex double-scroll or coupled tubular structure. To minimize the total potential energy of system, the nanosheet releases the lattice tension caused by defects through out-of-plane bending and scrolling, ultimately achieving self-assembly. As a complement to the atomistic MD simulations, finite element analysis (Movie S3) further validates the scrolling mechanism from a continuum mechanics viewpoint. By

imposing a gradient in-plane strain to mimic the lattice asymmetry caused by uneven S-vacancy distribution, the simulation reproduces the spontaneous scrolling process and confirms that tubular rolling is an effective pathway to release accumulated in-plane strain energy.

3.3 Fabrication of 1T-MoS₂ nanoscrolls

To elucidate the growth evolution of MoS₂ nanoscrolls, the schematic illustration of their formation mechanism is presented in Fig. 3(a). During the ultrasound-assisted exfoliation process, the bulk layered MoS₂ is initially dissociated into few-layer nanosheets, accompanied by the introduction of defects (indicated by red spots). The

S-vacancy disparity between two sides disrupts the centrosymmetry of the lattice, inducing asymmetric stress on the nanosheet surface. The asymmetric stress induced by unevenly distributed sulfur vacancies serves as the primary intrinsic driving force for scrolling, whereas the NMP/H₂O solvent system and ultrasonication mainly facilitate exfoliation, defect formation, and initial bending, thereby acting as kinetic promoters. Driven by the minimization of the system's total surface energy, the metastable nanosheets overcome their bending rigidity and spontaneously scroll inward from the edges, eventually evolving into structurally stable one-dimensional tubular structures.

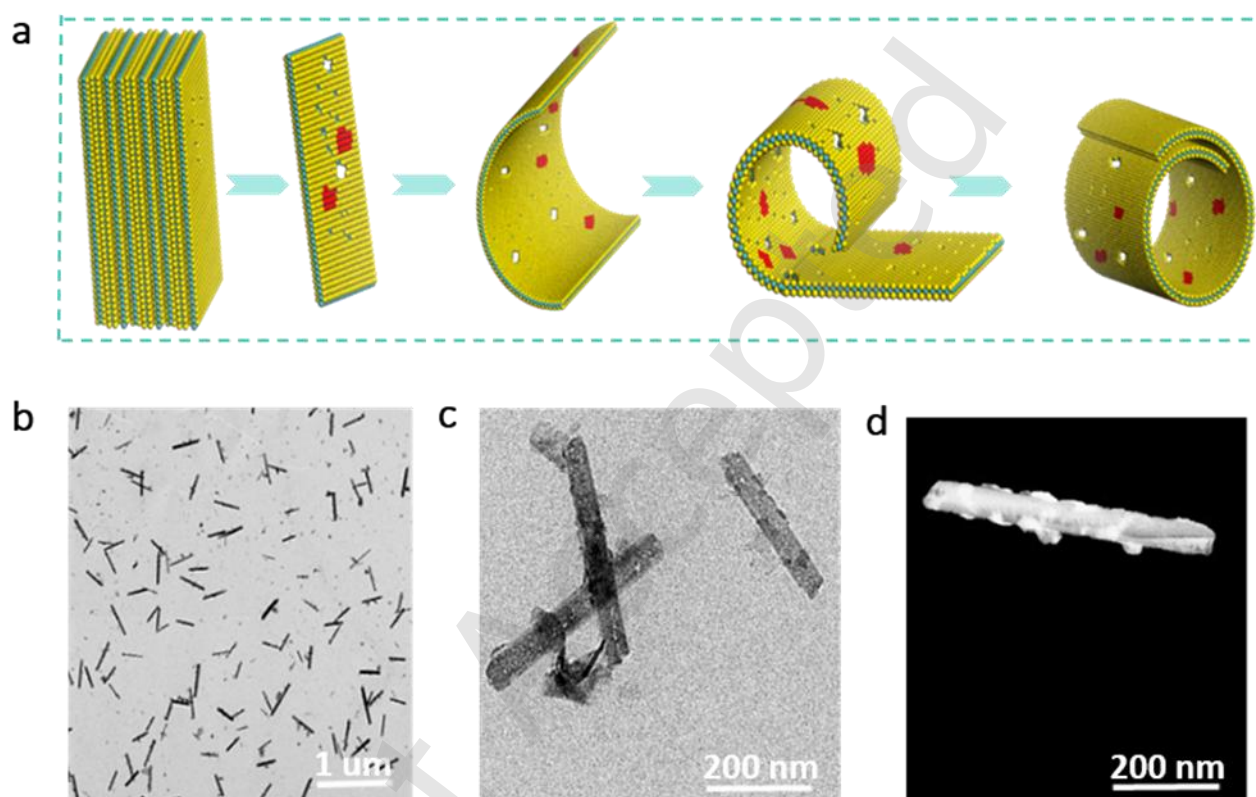


Figure 3 (a) Schematic illustration of the time-dependent formation of MoS₂ nanoscrolls through the ultrasound-assisted cracking process. SEM image of as-prepared MoS₂ with different morphology. (b) TEM image of MoS₂ nanoscrolls with scale bars of 1 μm. (c) TEM image of MoS₂ nanoscrolls with scale bars of 200 nm. (d) HAADF-STEM image of MoS₂ nanoscrolls.

Subsequent electron microscopy characterization further corroborates this structural transformation. Fig. 3(b) reveals that the obtained product consists of a large quantity of uniform tubular structures, indicating the excellent yield and dispersity of this synthesis strategy. Fig. 3(c) further unveils the topological details of an individual nanoscroll; its characteristic hollow structure and the distinct contrast difference at the tube walls provide strong evidence of its multi-walled nature formed by the scrolling of 2D nanosheets. Furthermore, the HAADF-STEM image (Fig. 3(d)), utilizing Z-contrast imaging, further visualizes the three-dimensional morphology of the nanoscroll. The bright contrast corresponds to the atomic accumulation of MoS₂, clearly exhibiting the structural integrity and surface topographical details along the axis. MoS₂ nanoscrolls suffer structural degradation under high-dose electron beam irradiation in TEM, accompanied by continuous atomic sputtering and loss from the scroll surface. After 100 s of

irradiation, only the outer contour of the nanoscroll can be clearly distinguished. (Figure S1) This phenomenon further provides indirect evidence for the hollow internal structure of the MoS₂ nanoscrolls. To characterize the chemical composition of the as-prepared nanoscrolls, point EDS measurements under HAADF-STEM mode were performed (Figure S2). The corresponding results only exhibit characteristic signals of Mo and S. This observation sufficiently verifies the successful fabrication of MoS₂ nanoscrolls. To fully characterize the length and diameter of MoS₂ nanoscrolls, we conducted statistical analysis on a large number of samples. Statistical statistics reveal that the average length of the nanoscrolls is 300 nm, with an average diameter of 35 nm (Figure S3).

3.4 Interfacial interaction and activation of water molecules on helical MoS₂ nanoscrolls

To evaluate the motion of water molecules surrounding the nanoscrolls in water, we performed MD simulation. The

blue-to-red scale denotes increasing instantaneous water velocity. The six snapshots reveal spatially heterogeneous and temporally fluctuating dynamics inside and outside the nanoscroll (Fig. 4(a)-(f)). Most molecules exhibit intermediate velocities, while sparse yellow-red and dark blue regions indicate faster and slower motion, respectively. Their continuous redistribution reflects rapid fluctuations driven by molecular collisions, hydrogen-bond rearrangement and water-wall interactions, while the overall velocity distribution remains stable at thermal equilibrium. Water molecules near the inner nanoscroll wall show greater velocities, as indicated by the prevalence of yellow and red regions. This enhanced mobility likely arises

from water-wall interactions and geometric confinement, which boost translational motion, particularly in the radial direction. A fast interfacial water layer therefore forms near the wall. Continuous switching between fast and slow states highlights the dynamic heterogeneity of nanoconfined water.

Both confined and bulk water are dominated by intermediate molecular velocities, indicating that nanoscroll confinement does not freeze thermal motion. Outside the nanoscroll, velocities are more uniformly and randomly distributed. Inside, they show clear radial dependence, with faster motion near the wall. Further details are available in the supporting video (Movie S4).

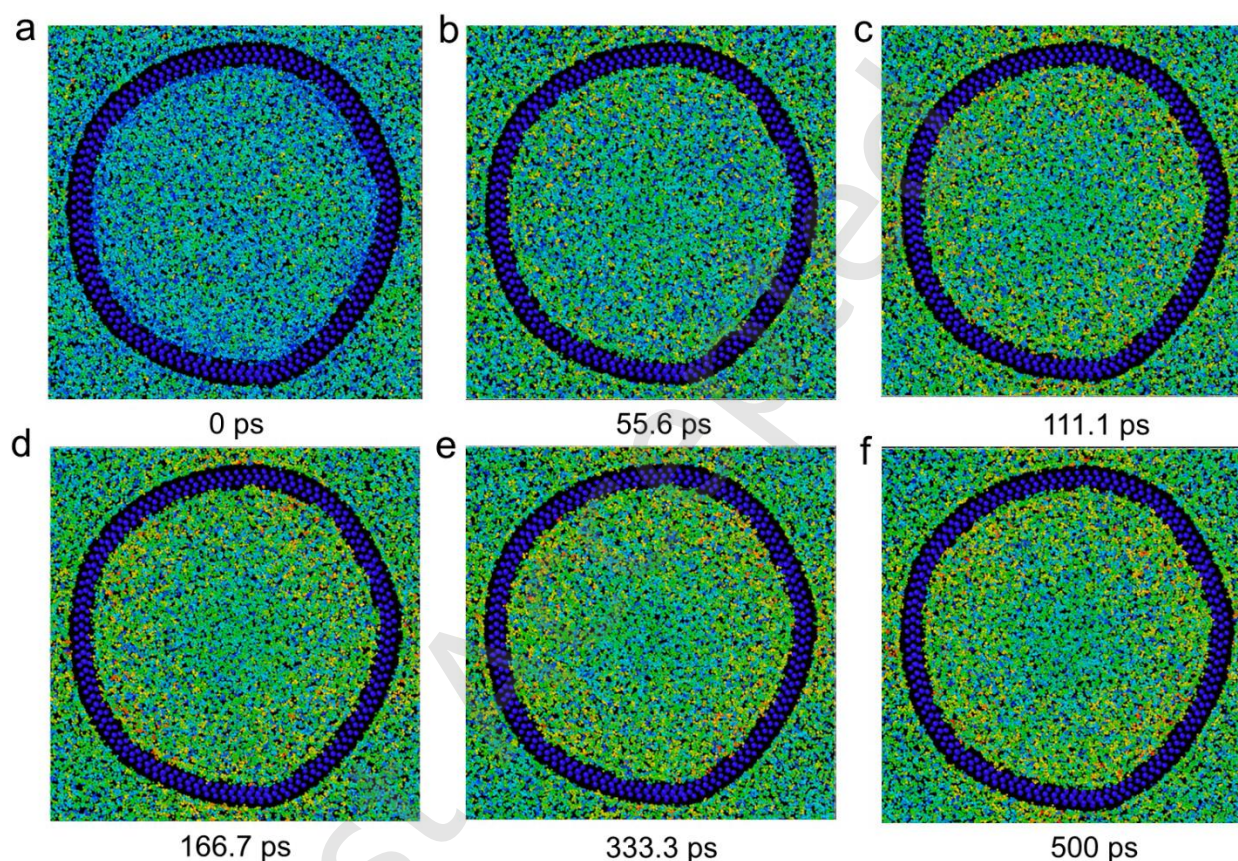


Figure 4 MD simulation on interfacial water behaviors induced by MoS₂ nanoscroll structures. Specifically, the colors represent water-molecule velocities ranging from 0 to 1200 m s⁻¹, with the color changing progressively from dark blue at the lowest velocity to red at the highest velocity.

3.5 Dye-sensitized HER performance

To comprehensively elucidate the origin of the exceptional catalytic performance and validate the practical applicability of the engineered catalysts, a dye-sensitized photocatalytic hydrogen evolution system was constructed (Fig. 5). As schematically illustrated in Fig. 5(a), the highly efficient HER process is driven by a synergistic interplay of rapid charge transfer and enhanced mass transport. Under light irradiation, the Eosin Y (EY) photosensitizer undergoes excitation and intersystem crossing (ISC) to generate the long-lived triplet state (EY^{3*}), which is subsequently reduced by the triethanolamine (TEOA) sacrificial donor. The resulting highly reducing EY^{3*} species efficiently injects

electrons (e⁻) into the highly conductive 1T-MoS₂ nanoscroll network. Crucially, the unique hollow tubular architecture functions as an open microfluidic channel, enabling water molecules to rapidly penetrate the inner cavity. This capillary-like fluidic transport guarantees dual-surface accessibility for a continuous proton supply while facilitating the immediate detachment and rapid diffusion of the generated H₂ bubbles. Consequently, this structural advantage effectively prevents the active sites from being blocked by gas accumulation, sustaining the interfacial reaction kinetics.

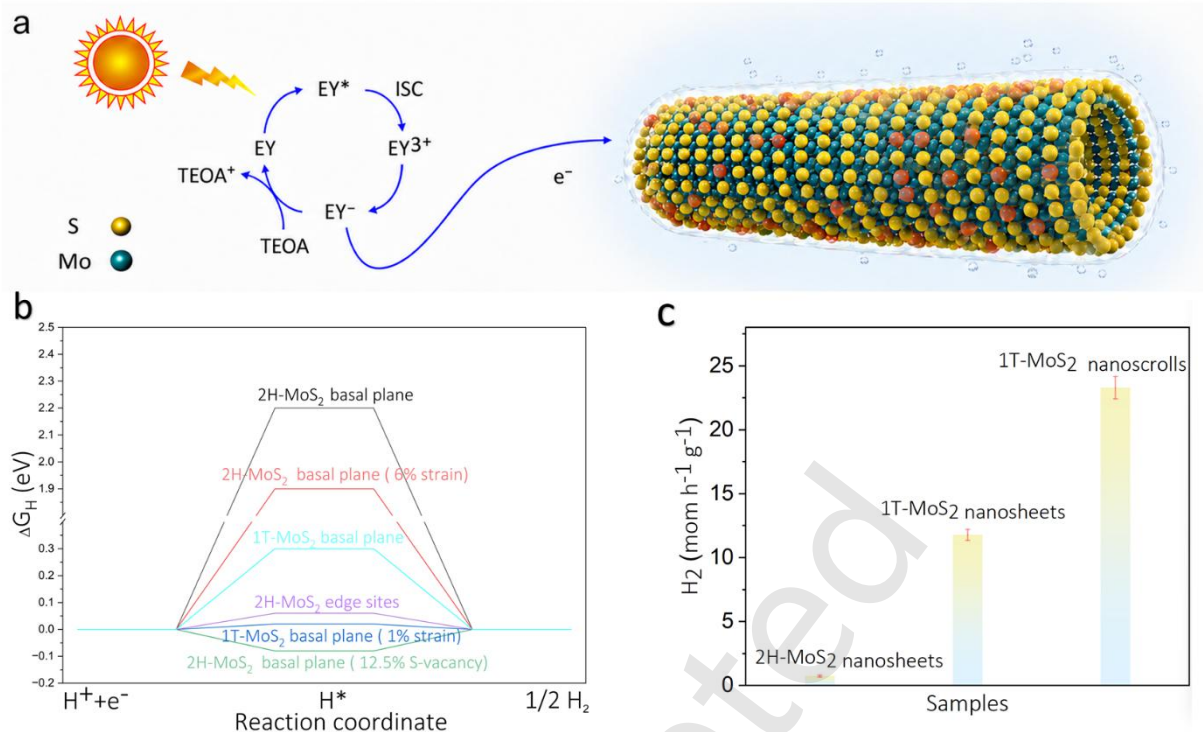


Figure 5 (a) High-efficiency HER mechanism of 1T-MoS₂ nanoscrolls in practical photocatalytic reaction. (b) ΔG_H^* diagram of hydrogen evolution in the 2H-MoS₂ basal plane, 2H-MoS₂ basal plane (6% strain), 1T-MoS₂ basal plane, 2H-MoS₂ edge sites, 1T-MoS₂ basal plane (1% strain), and 2H-MoS₂ basal plane (12.5% S-vacancy) (c) H₂ generation activity values of 2H-MoS₂ nanosheets, 1T-MoS₂ nanosheets and 1T-MoS₂ nanoscrolls in practical photocatalytic reaction.

Fig. 5(b) shows that strain and S vacancies markedly influence the hydrogen adsorption free energy (ΔG_H^*) on MoS₂. Pristine MoS₂ without strain or vacancies exhibits a ΔG_H^* of 2.20 eV. This value decreases to approximately 1.84 eV under 6% strain and to -0.08 eV in the presence of 12.5% S-vacancies concentration. Notably, 1T-MoS₂ basal plane with 1% strain combining strain further reduces ΔG_H^* from 0.30 eV to only 0.02 eV, approaching the thermoneutral value required for efficient hydrogen evolution. The resulting electronic-structure modulation therefore substantially lowers ΔG_H^* and activates the otherwise inert MoS₂ basal plane for the hydrogen evolution reaction[42]. The synergistic coupling of the abundance of sulfur vacancies, and the inherent lattice strain induced by the highly curved tubular topology shifts the ΔG_H^* remarkably close to the ideal thermoneutral state $\Delta G_H^* \approx 0$. Driven by this optimized thermodynamic profile and the highly efficient mass transport network, the macroscopic catalytic performance exhibits a dramatic enhancement (Fig. 5(c)). In the practical dye-sensitized HER, the 1T-MoS₂ nanoscrolls with appropriate strain deliver a robust H₂ generation rate of 24.1 mmol g⁻¹ h⁻¹. This performance substantially outperforms the conventional 1T-MoS₂ nanosheets (approximately 12 mmol g⁻¹ h⁻¹) and exhibits an activity nearly 25 times higher than that of the pristine 2H phase. This pronounced enhancement highlights rolltronics as an effective structural-design paradigm for achieving efficient and stable HER by synergistically integrating curvature engineering, defect activation and metallic-phase stabilization within a single MoS₂ nanoscroll architecture. Overall, the enhanced HER performance of the MoS₂ nanoscrolls arises from the cooperative regulation of electronic structure, active-site chemistry, adsorption thermodynamics, and mass transport. The metallic 1T phase facilitates rapid electron transfer, while S vacancies provide abundant catalytically

accessible sites. Meanwhile, curvature-induced strain further optimizes ΔG_H^* toward the thermoneutral value, thereby promoting hydrogen adsorption and evolution. In parallel, the open tubular architecture exposes both inner and outer surfaces and facilitates the transport of reactants and the release of generated H₂, collectively accounting for the superior HER activity of the 1T-MoS₂ nanoscrolls.

3.6 Stability of 1T-MoS₂

The long-term structural and chemical stability of 1T-MoS₂ nanoscrolls was evaluated by comparing as-prepared samples with counterparts stored under ambient conditions for 6 months, using high-resolution X-ray photoelectron spectroscopy (XPS, Fig. 6(a-d)) and Raman spectroscopy (Fig. 6(e,f)). The high-resolution Mo 3d XPS spectra can all be deconvolled into the Mo 3d_{5/2} and Mo 3d_{3/2} doublet of Mo⁴⁺ in 1T-MoS₂, a minor Mo 3d doublet corresponding to 2H-MoS₂, and a weak Mo⁶⁺ component located in the higher binding energy region, the peak at approximately 226 eV is assigned to the S 2s signal [43, 44]. The Mo 3d_{5/2} component located at approximately 229.4 eV is assigned to 2H-MoS₂, while the component at approximately 228.6 eV corresponds to the metallic 1T-MoS₂ phase. A Shirley-type background was adopted, and Gaussian-Lorentzian mixed functions were employed for peak deconvolution, with constrained spin-orbit splitting and area ratio between the Mo 3d_{5/2} and Mo 3d_{3/2} doublets. No unphysical peak positions were allowed during iteration. The 1T phase fraction was calculated from the integrated area ratio of the 1T-related Mo 3d_{5/2} peak to the total area of both the 1T and 2H Mo 3d_{5/2} peaks. The 1T-MoS₂ content remains stable at approximately 70%, with the remaining 30% assigned to 2H-MoS₂. Despite the presence of this 2H component, the

sample retains abundant defects and substantial lattice strain. We therefore employed density functional theory (DFT) calculations to examine the influence of defects on the electronic band structure of MoS₂ (Figure S4). The sulfur vacancy concentration can also be quantified by XPS, yielding an estimated S-vacancy concentration of approximately 14.5%. As shown in Figure S5, the bandgap progressively narrows with increasing defect concentration. This defect-induced electronic modulation may facilitate charge transport and thereby contribute to the enhanced HER performance.

Compared with the initial sample, the binding energy positions, peak profiles, and relative distributions of all

components in the sample stored for 6 months remain generally consistent, with no discernible new chemical state peaks observed. Meanwhile, the Mo⁶⁺-related signal shows no significant enhancement, indicating that only limited surface oxidation occurs during long-term storage, without substantial deep oxidation or bulk chemical state transformation. The S 2p spectra all consist of S 2p_{3/2} and S 2p_{1/2} spin-orbit doublets, with the peak positions and line shapes of the two sets of samples essentially overlapping, and no prominent high-binding-energy oxidized sulfur species signals appear, suggesting that the primary chemical environment of the sulfur species remains stable over the 6 month period.

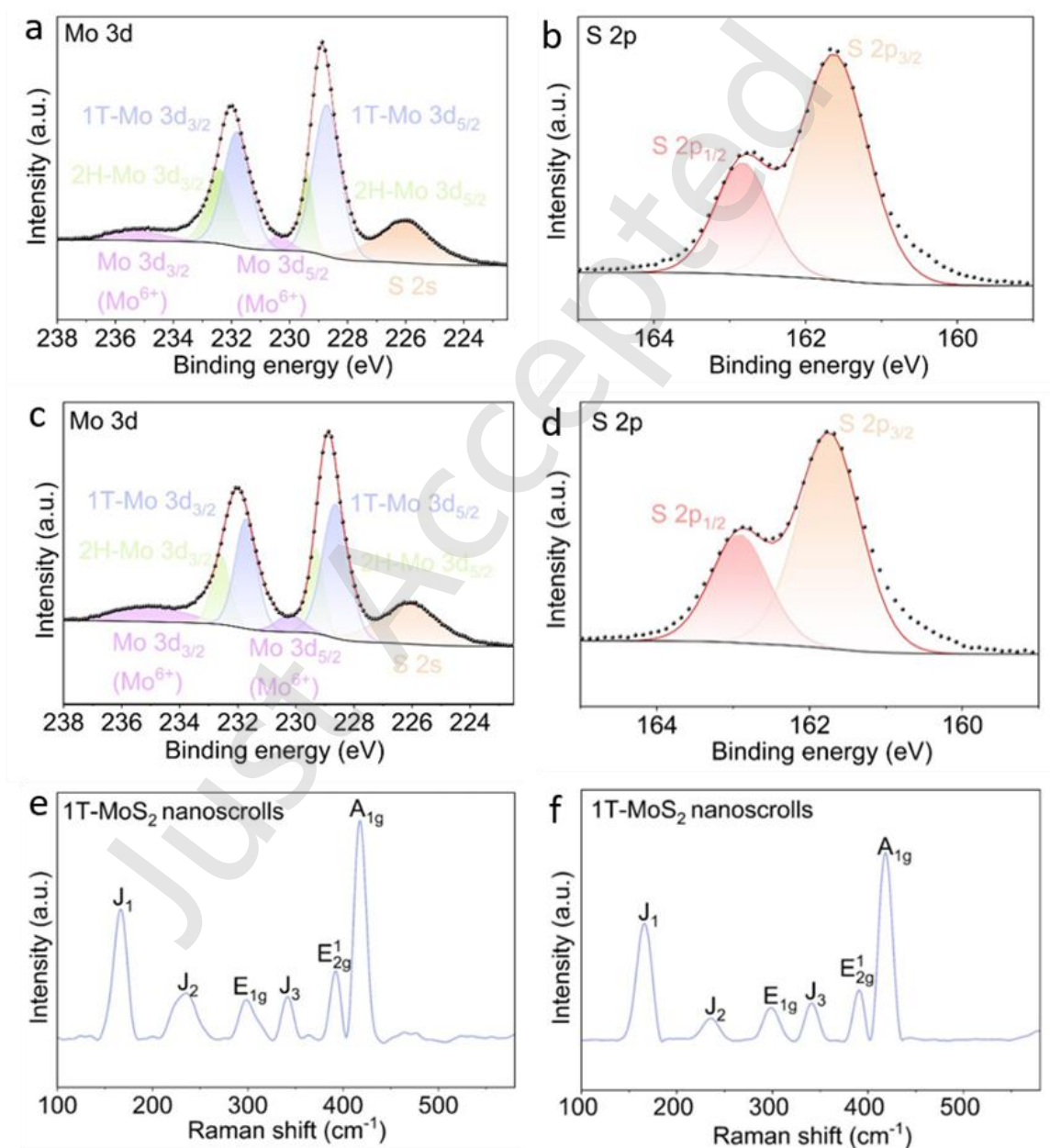


Figure 6 XPS spectra and Raman spectra of 1T-MoS₂ nanoscrolls before and after 6 months of storage. (a,b) XPS spectra before 6 months of storage. (c,d) XPS spectra after 6 months of storage. (e) Raman spectra before 6 months of storage. (f) Raman spectra after 6 months of storage.

Raman spectroscopy further confirms the long-term stability of the 1T phase. In the initial sample (Fig. 6(e)), the characteristic vibrational modes J₁, J₂, and J₃ are clearly observed, accompanied by the E_{1g}, E_{2g}¹, and A_{1g} vibrations.

After 6 months of storage (Fig. 6(f)), these characteristic peaks are fully retained, with no significant changes in their peak positions or spectral profiles, exhibiting only limited differences in relative intensity, which may be attributable to

the testing region, sample orientation, and signal normalization.

To summarize, the combined XPS and Raman results demonstrate that after 6 months of storage, the 1T-MoS₂ nanoscrolls maintain elemental chemical states and crystalline phase compositions comparable to those of the initial sample, without significant oxidation, reconstruction of the sulfur chemical environment, or large-scale 1T→2H phase transformation. This indicates that the as-prepared sulfur-vacancy-containing 1T-MoS₂ nanoscrolls possess excellent long-term stability.

4 Conclusion

In summary, we introduce a rolltronics strategy in which a disparity in sulfur-vacancy concentration between the two surfaces of MoS₂ sheets generates asymmetric stress, thereby driving their spontaneous transformation into nanoscrolls. The resulting nanoscrolls deliver a hydrogen evolution rate of 24.1 mmol g⁻¹ h⁻¹ in a dye-sensitized photocatalytic system, representing a 25-fold enhancement over 2H-MoS₂ nanosheets. This pronounced improvement arises from the cooperative regulation of catalytic and transport processes within the curved architecture. Sulfur vacancies increase the density of accessible active sites, the metallic phase promotes interfacial charge transfer, and curvature modulates the hydrogen adsorption free energy towards more favourable reaction kinetics. Meanwhile, the open tubular geometry exposes both inner and outer surfaces and facilitates reactant diffusion and hydrogen release. These results establish MoS₂ nanoscrolls as an effective structural platform for simultaneously regulating phase, defects, adsorption energetics and mass transport. More broadly, rolltronics provides a general route towards low-cost and efficient transition-metal dichalcogenide catalysts for sustainable hydrogen production.

Electronic Supplementary Material: Supplementary material (photograph of the TEM images, HAADF-STEM image, point EDS spectra, electronic structures of MoS₂ with S-vacancies, evolution of the band structure with increasing S-vacancy concentration, and video of MD simulation and FEA) is available in the online version of this article at <https://doi.org/10.26599/NR.2026.94909162>.

Data availability

All data needed to support the conclusions in the paper are presented in the manuscript and/or the Electronic Supplementary Material. Additional data related to this paper may be requested from the corresponding author upon request.

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Declaration of competing interest

All the contributing authors report no conflict of interests in this work.

Author contribution statement

Y. C.L.: Data curation, project administration, conceptualization, validation, writing – original draft, experimental design. J. Y. X.:

Data curation, formal analysis, writing review & editing. D. Y. Y.: Writing review & editing, visualization. N. S.: Visualization. X.L.: Investigation, software, formal analysis. L. L. W.: Resources, funding, acquisition, supervision, project

administration. Q. Z.: Resources, supervision, methodology.

Use of AI statement

None.

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Electronic Supplementary Material

Rolltronics-enabled MoS₂ nanoscroll for efficient hydrogen evolution

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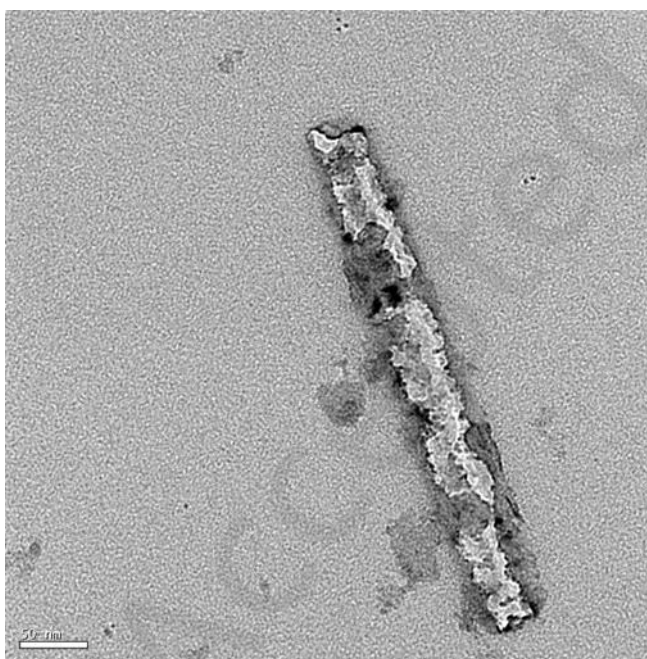


Figure S1 TEM image of MoS₂ nanoscrolls upon the electron beam exposure over 100 s.

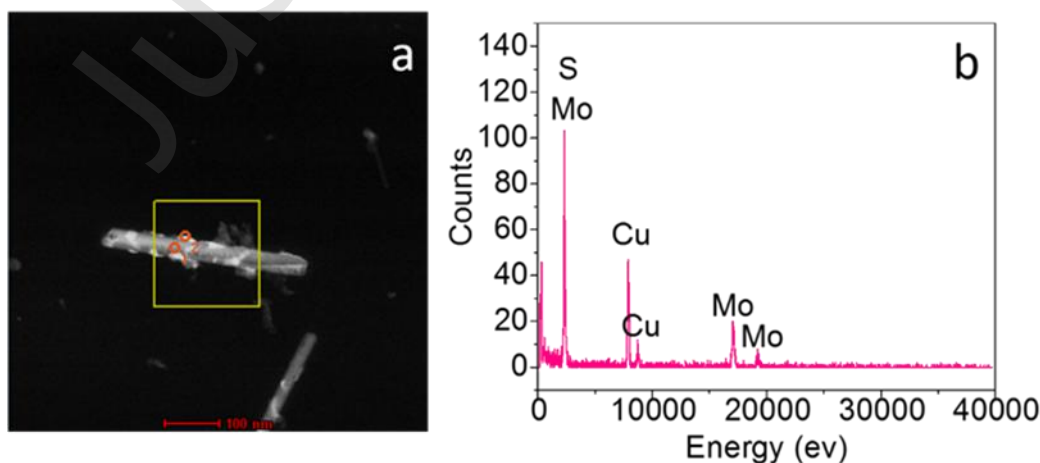


Figure S2. (a) HAADF-STEM image and (b) point EDS spectra of MoS₂ nanoscrolls.

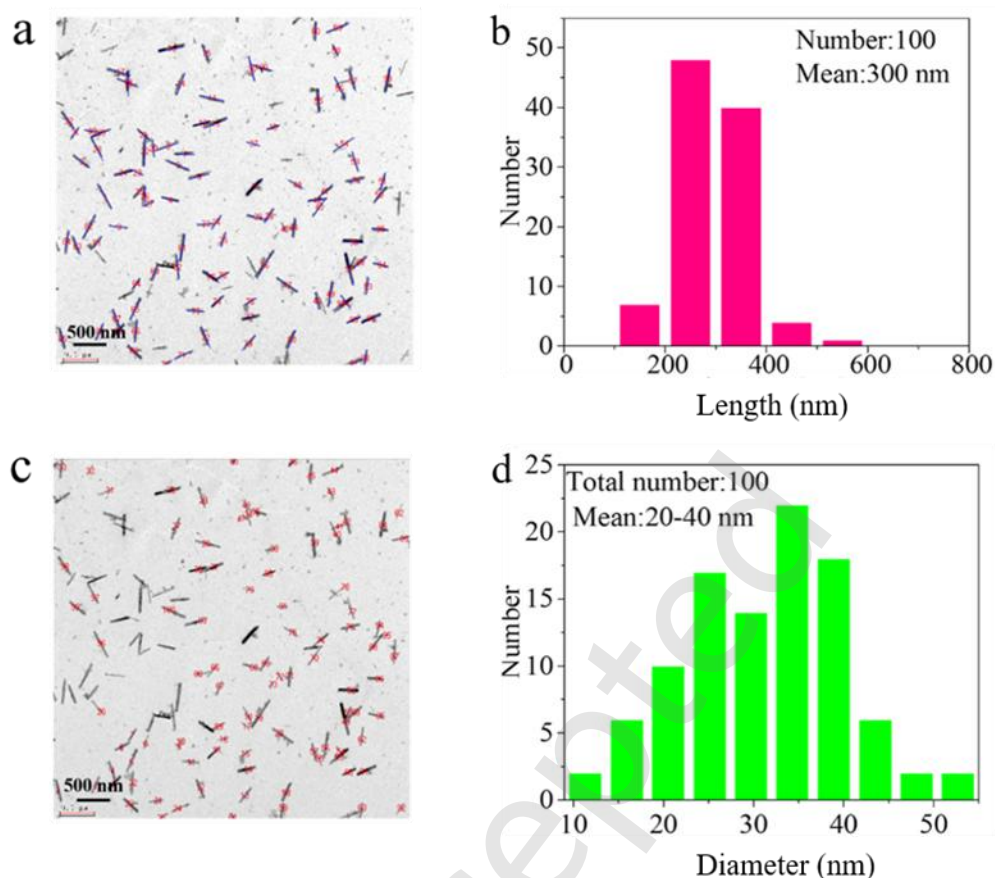


Figure S3 TEM images (a,c) and the statistical analysis of length distribution (b) and diameter distribution(d) of MoS₂ nanoscrolls from TEM images.

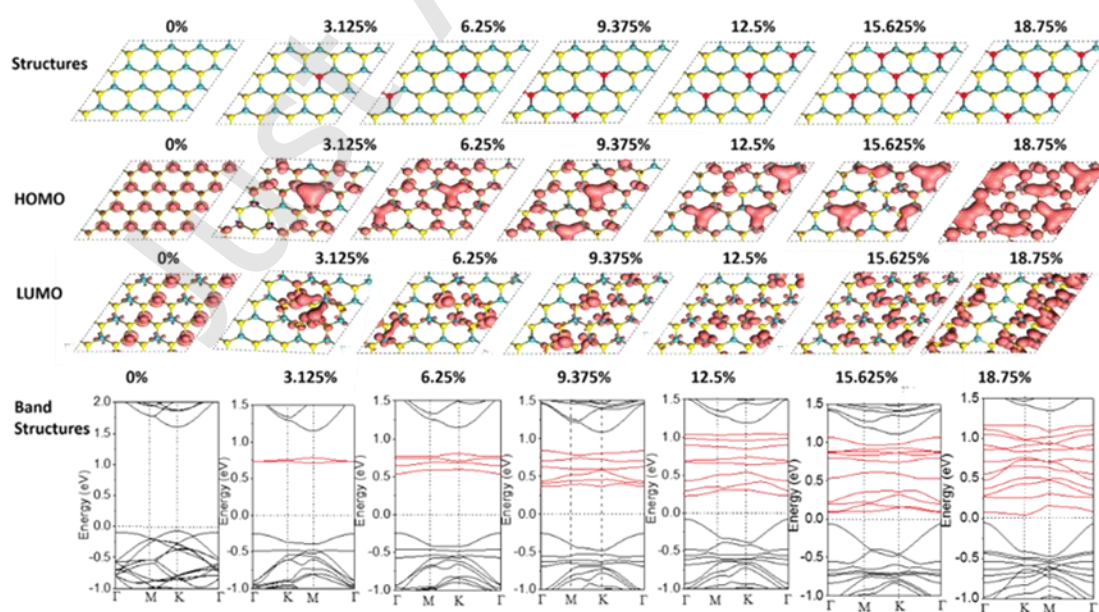


Figure S4 Electronic structures of MoS₂ with S-vacancies. Evolution of the LUMO and HOMO states of MoS₂ with increasing S-vacancy concentration. The electron densities are the red surfaces. Evolution of the band structure with increasing S-vacancy concentration (indicated by %).

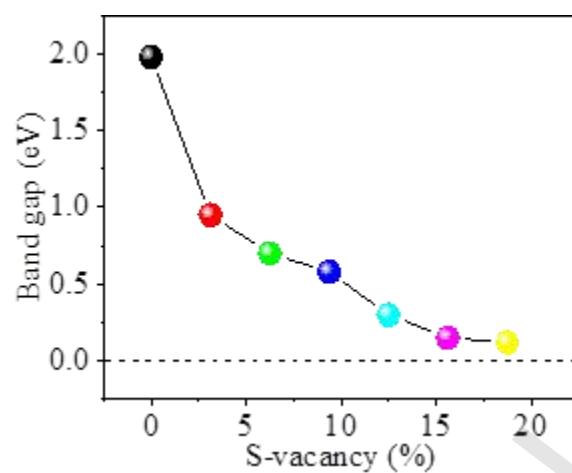


Figure S5 Evolution of the band structure with increasing S-vacancy concentration.

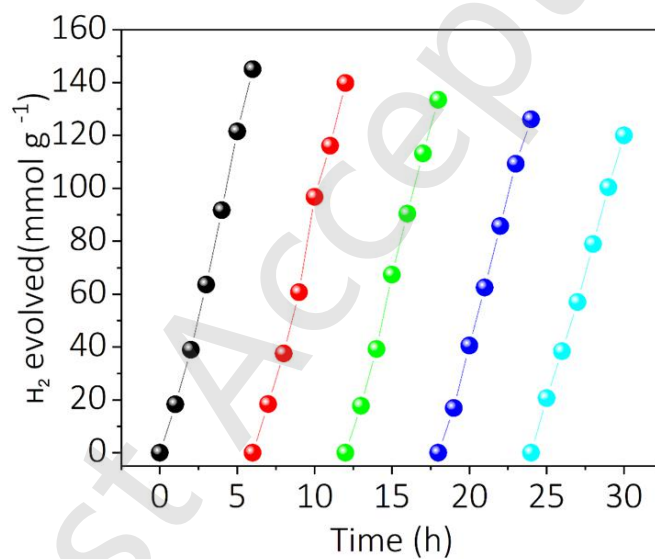


Figure S6 H₂ evolved by 1T-MoS₂ nanoscrolls over a period of 30 hrs in cycles of 6 hrs each showing the stability of the catalyst for H₂ evolution.