Enhanced Photocatalytic Activity of SiC-Based Ternary Graphene Materials: A DFT Study and the Photocatalytic Mechanism

Yuyan Zhang,† Yan Zhang,*‡† Xue Li,† Jianhong Dai,† Fengjuan Song,† Xiaoqiang Cao,† Xianjun Lyu,† and John C. Crittenden†§

†School of Chemical and Environmental Engineering, Shandong University of Science and Technology, Qingdao 266000, China
‡School of Materials Science and Engineering, Harbin Institute of Technology at Weihai, 2 West Wenhua Road, Weihai 264209, China
§School of Civil and Environmental Engineering, Georgia Institute and Technology, 828 W. Peachtree Street, Suite 320, Atlanta, Georgia 30332-0595, United States

ABSTRACT: A graphene-like semiconductor composite is one of the most promising photocatalyst that does not use noble metals. These composites have excellent photocatalytic properties and have attracted great attention for water splitting. Here, a facile method called the hydrothermal method was used to prepare graphene oxide (GO)/SiC/MoS2 composites. Under visible-light irradiation, the GO/SiC/MoS2 composite had excellent photocatalytic production of hydrogen from water splitting. In particular, the catalyst added 8 wt % of Mo weight yielded the highest quantum of 20.45% at 400–700 nm of wavelength. A positive synergistic effect between the layered GO and MoS2 components contributed to the enhanced photoactivity of the SiC particles. The synergistic effect reduced the recombination of photogenerated holes and electrons, enhanced the rate of electron transfer, and provided more reaction active sites for water splitting. The interactions among SiC, GO, and MoS2 were investigated using a density functional theory. The calculations showed that the relative positions between graphene only slightly affect the stability of the interface, and the MoS2 layers have a great influence. The photocatalytic mechanism was also discussed, and electron transfer was predicted.

INTRODUCTION

The use of sunlight for water splitting is a method that can replace fossil fuels and protect the environment.1–4 Presently, a number of semiconductor materials, such as TiO2,5–7 and ZnO,8,9 can be used for photocatalysis using ultraviolet light. Other semiconductor materials, such as BiVO4,10,11 and CdS,12,13 can split water using visible light. However, these semiconductors do not have high photocatalytic activities without a cocatalyst even in the presence of the sacrificial chemical. The low activity is caused by the recombination of photogenerated electrons and holes before they migrate to the surface for photocatalytic reaction. To retard the recombination of photogenerated electrons and holes, metals, especially noble metals, such as Pt14 and Au,15 are incorporated on the surface of semiconductors to reduce hole–electron recombination. Semiconductor-based photocatalysis is regarded as a green technology for environmental remediation and solar energy conversion.16

Silicon carbide (SiC) belongs to the third generation of semiconductor materials and has a suitable band gap (2.4–3.2 eV), large thermal conductivity, and good chemical stability.17–19 SiC itself does not produce hydrogen under visible light due to its high surface energy. However, the position of the valence band and the conduction band completely meet the requirements of photolysis water, that is, the bottom position of the conduction band is negative compared with the redox potential of H+/H2 and the position of the top of the valence band is positive compared with the redox potential of O2/H2O. In the field of photocatalysis, SiC is regarded as the most promising photocatalytic material due to its high photo reduction of conduction band electrons. Early in 2012, Hao et al.20 reported that the β-SiC nanowires can absorb visible light and exhibit excellent photocatalytic hydrogen evolution performance from pure water under visible-light irradiation. Peng et al.21 further studied the importance of heterojunctions in SiC/CdS composite photocatalytic materials and found that lattice matching can increase hydrogen production. Improving the photocatalytic efficiency of SiC remains a challenge. Most recently, the use of nanosized noble metals, such as Au,22 to decorate SiC has been used to form a new Fermi level equilibrium that inhibits the photogenerated electrons and holes recombination. The adoption of noble metals and other substances, such as Pt/SiC18 and SiC–CdLa2S4,23 significantly improves the photocatalytic efficiency of SiC. However, the scarcity of precious metals, high cost, and subsequent environmental pollution hinder their widespread...
use. Therefore, the development of nonprecious metals as efficient cocatalysts to promote the effective separation of photogenerated electrons and holes is urgently needed.

As a result, low-cost and highly efficient cocatalysts are needed for photocatalytic hydrogen production. A typical layered transition-metal sulfide, molybdenum disulfide (MoS2), with a structure of three stacked atom layers (S–Mo–S) held together by van der Waals forces, has attracted much attention for photocatalytic hydrogen evolution.24,25 In addition, a layered transition material such as MoS2 has proven to be an encouraging promoter due to its rich active edge sites. Shi et al.26 reported that MoS2 as a cocatalyst of photocatalytic H2 generation activity of g-C3N4 under visible-light irradiation. They concluded that the emission of g-C3N4 is quenched by MoS2 nanodots more effectively than by MoS2 monolayers. Graphene oxide (GO) exhibits superior electron mobility and excellent electronic behavior. Additionally, GO is an efficient cocatalyst for photocatalytic hydrogen production reactions because of its high specific surface area and good electron-transfer ability.27,28

However, to the best of our knowledge, SiC powder photocatalyst with layered GO/MoS2 has not been reported. Herein, we report the fabrication of SiC particles loaded with a nanosized layer-structured GO/MoS2 nanosheet. We used a facile hydrothermal method and optimized MoS2 content in the GO/SiC/MoS2 photocatalyst. This catalyst provides abundant reactive sites for hydrogen evolution and can increase the transport of charges and reduce the recombination of photogenerated electrons and holes. As a result, our GO/SiC/MoS2 photocatalyst exhibited significantly improved photocatalytic activity for photocatalytic water splitting.

## RESULTS AND DISCUSSION

### Structures and Morphologies of the Samples.

The morphology and microstructure of SGM-8 were characterized by field emission scanning electron microscopy (FESEM) and transmission electron microscopy (TEM), and the relevant results are shown in Figure 1. Figure 1a,b shows the FESEM images of pure SiC and SGM-8, respectively. Figure S2a–c (in the Supporting Information S4) shows the FESEM images of SGM-2, SGM-5, and SGM-10, respectively. The GO and MoS2 modification effect on the pure SiC structure is shown in Figures 1b and S2a–c. Sharper nanosheets were obtained in all GO/SiC/MoS2 samples and seem to be more disorganized than pure SiC. Sharper nanosheets can lead to more exposed surfaces and active edges, which may contribute to the enhanced photocatalytic hydrogen production performance.29

Energy-dispersive spectrometry (EDS), as shown in Figure 1c, determined the elemental composition at (c) in Figure 1b. As depicted in Figure 1c, the presence of the elements of Si, C, Mo, and S is validated for the SGM-8 photocatalyst. High-resolution TEM (HRTEM) images of SGM-8 samples are shown in Figure 1d. Obviously, the lattice fringes correspond to the interplanar distance of 0.25 and 0.27 nm, which can be attributed to the (006) plane of the SiC phase and the (101) plane of the MoS2 phase, respectively. This tight contact enables stability and promotes the separation and transfer of photogenerated carriers between SiC, GO, and MoS2. This is definitely beneficial for the photocatalytic hydrogen evolution reaction.30 Further, the distribution of the three substances in SGM-8, a TEM image, and the corresponding energy-dispersive X-ray (EDX) spectrum were taken and are shown in Figure 3f. There are notable distributions of Si, C, Mo, and S elements in the SGM-8 sample, indicating that the SGM-8 hybrid structure was successfully constructed. There was also a contact surface between GO and MoS2. In the following density functional theory (DFT) calculations, specific research will be conducted on the influence of GO and MoS2 on the photocatalytic activity.

Powder X-ray diffraction (XRD) measurements were employed to further confirm the crystal structure of the GO/SiC/MoS2 composites. Figure 2 shows the XRD pattern. The diffraction peaks at 2θ = 34.097, 35.658, 38.136, 41.392, 45.294, 54.630, 60.010, 65.626, 70.856, 71.765, 73.358, and 75.520° are attributed to the (101), (006), (103), (104), (105), (107), (110), (109), (201), (116), (203), and (204) planes of SiC, respectively.
planes of a hexagonal structure SiC and match well with those of 6H-SiC (No.72-0018). The XRD patterns of the GO/SiC/MoS₂ composites show that it has a similar structure to pure SiC, suggesting that the hydrothermal process has no significant influence on the crystal structure of SiC. The GO peaks were not observed since the content of graphene in the catalysts is low. SiC combined with GO and MoS₂ has stronger diffraction peaks than SiC. The addition of GO and MoS₂ promoted the growth of SiC crystals. Good crystallization will reduce defects of semiconductor photocatalysis, which is beneficial to the transfer of photogenerated electrons and holes in and between the lattices, thus improving the activity of photocatalyst. In comparison, as the MoS₂ loading increased, the intensity of the characteristic peaks in the sample is strong first and then weaker. Additionally, the intensity of the characteristic peaks of SGM-8 is the strongest. The XRD result is in good agreement with the SEM and TEM results, suggesting a new photocatalytic alternative to SiC.

The textural properties of the prepared samples were analyzed by nitrogen adsorption–desorption experiments. Figure 3 presents the adsorption–desorption isotherms of nitrogen and the corresponding pore size distribution curves (the inset) for pure SiC, SGM-2, SGM-5, SGM-8, and SGM-10. These isotherms show a typical IV adsorption–desorption isotherm according to the IUPAC classification, indicating the presence of mesopores. According to the BDDT adsorption isotherm classification, the composite material has a small change in SGM-2, as an H3 type-shape hysteresis loop in the P/P₀ range of 500–750 mmHg with no saturated adsorption till P/P₀ = 800 mmHg, suggesting the presence of mesopores. This can be attributed to the interaction of SiC with MoS₂ and GO. The pore-size distributions of the samples (inset of Figure 3) indicate a large number of pores centered at approximately 45.2, 38.3, 33.5, 32.6, and 37.1 nm for the pure SiC, SGM-2, SGM-5, SGM-8, and SGM-10 samples, respectively. The Brunauer–Emmett–Teller (BET) specific surface areas obtained from the desorption of branches are presented in Table S1 (in the Supporting Information S5) as the average results of two repeated measurements with an error bar for each case. The average specific surface areas were 0.40, 0.97, 2.57, 3.73, and 2.67 m²·g⁻¹ for the pure SiC, SGM-2, SGM-5, SGM-8, and SGM-10 samples, respectively. Combined with the SEM results, GO, and MoS₂ could change the morphology of SiC. Notably, a large surface area contributes more reaction sites during the photocatalytic reaction process, which is beneficial for increasing the activity of the photocatalytic reaction.

**Optical Absorptive Performance.** The optical properties of the GO/SiC/MoS₂ composites were investigated using UV–vis diffuse reflectance spectroscopy (DRS). Figure 4a shows the UV–vis absorption spectra of the pure SiC and GO/SiC/MoS₂ composites. Compared with the pure SiC UV–vis DRS, the other samples show enhanced photo-absorption in the wavelength range from 370 to 770 nm caused by the strong photoabsorption of the MoS₂/GO, and the degree of red shift is similar in SGM-8 and SGM-10. This was mainly because of their narrow band gap and deep color, which is beneficial for the photocatalytic activity. The light absorption edge in the visible-light region indicates that the GO/SiC/MoS₂ composites have a better response to the visible light. The band gap energy of synthesized composites plays an important role in deciding the photocatalytic activity of the photocatalyst. The band gaps of pure SiC and GO/SiC/MoS₂ composites were calculated by extrapolating the linear part of the (Aν)² versus energy (νh) function (where A = absorbance, h = Planck’s constant, and ν = frequency) as shown in the inset of Figure 4b. The so-called Tauc plots are derived by using the Kubelka–Munk method. The corresponding band gap energies calculated based on Tauc plots are 2.91, 2.73, and 2.61 eV for SGM-2, SGM-5, and SGM-8, respectively. The obtained band gap of SGM-10 is similar to that of SGM-8. For the composites, the obtained band gap of pure SiC (≈3.02 eV) is greater than those of the photocatalytic samples.
Interaction among SiC, GO, and MoS₂. X-ray photoelectron spectroscopy (XPS) results of SGM-8 and SGM-10 are shown in Figure 5. Generally, the Si 2p peaks at the binding energy (BE) of 100.8 and 102.1 eV are attributed to the Si—C and Si—O bonds, respectively, of which the C 1s peak BE values are 282.9 and 284.7 eV for the C—Si and C—C bonds.

In the comparison of SGM-10, the peak intensity, Si—C and Si—O bonds area of SGM-8 decrease. However, from the C 1s peak, there are no C—Si bonds in the SGM-10 sample because of inorganic C pollution. As shown in Figure 5e,f, the fitted S peaks for the S 2p3/2 and 2p1/2 orbitals are located at 161.3 and 163.3 eV and are S²⁻. The Mo 3d₅/₂ and Mo 3d₃/₂ peaks are...
located at 229.3 and 232.5 eV, respectively,\textsuperscript{24,35} and these two fitted peaks represent the presence of Mo\textsuperscript{4+}. The fitted peak at 226.3 represents the 2s orbital interaction of Mo with S. The fitted peaks at 233.1 and 230.6 eV represent the presence of Mo\textsuperscript{5+}. The fitted peak at 235.7 eV represents the presence of Mo\textsuperscript{6+} (Figure 5g,h).\textsuperscript{36} Compared with the SGM-8 sample, too many Mo chemical states appeared in the SGM-10 sample. The presence of MoS\textsubscript{2} and other new substances caused chemical changes during the hydrothermal reaction.

**Photocatalytic Activity and Stability.** Photocatalytic hydrogen evolution activities of SGM-2, SGM-5, SGM-8, and SGM-10 were evaluated under visible-light irradiation (400−700 nm) in a mixed aqueous solution containing 0.1 M Na\textsubscript{2}S\textcdot9H\textsubscript{2}O and 0.1 M Na\textsubscript{2}SO\textsubscript{3}. The photocatalytic hydrogen production activities of the samples were measured and compared in Figure 6. Figure 6a shows the hydrogen evolution result of 0.5 g of SGM-2, SGM-5, SGM-8, and SGM-10 under 4 h illumination of simulated sunlight, with pure SiC for comparison. Pure SiC has no potential for hydrogen production due to its high surface potential. The samples of SGM-2, SGM-5, SGM-8, and SGM-10 showed the hydrogen evolution of 2.7054, 2.7538, 4.2721, 2.8265, 3.3673, and 2.2922 mL, respectively. The sample of MG has no photocatalytic hydrogen production. This is because the position of the conduction band does not meet the requirements of photocatalytic hydrogen production. Figure 6b shows the rate of H\textsubscript{2} evolution of samples with GO/SiC/MoS\textsubscript{2} composites. Only low hydrogen was detected when MoS\textsubscript{2} alone was employed as a photocatalyst (47 μmol·h\textsuperscript{-1}·g\textsuperscript{-1}) under the same experimental conditions.\textsuperscript{37} This indicates that SiC is the predominant active component for photocatalytic H\textsubscript{2} evolution in the nanocomposites. The catalyst of SGM-2, SGM-5, SGM-8, SGM-10, SM, and SG gave a rate of 27.60, 28.10, 43.59, 28.84, 34.36, and 23.38 μmol·h\textsuperscript{-1}, respectively. The QY\textsuperscript{400−700} on the SGM-8 catalyst could reach up to 20.45%. The result shows that the introduction of MoS\textsubscript{2} led to a significant enhancement in photocatalytic hydrogen production. We speculate that the ratio of MoS\textsubscript{2} and
GO is very important for the hydrogen generation rate. A small amount of MoS$_2$ and GO is not enough to improve the charge-carrier separation and transfer, while much MoS$_2$ may lead to the aggregation of SiC, and this is responsible for a lower photocatalytic performance.

Furthermore, to evaluate the stability of photocatalysts, we conducted the cycling tests of the photocatalytic hydrogen evolution by repeatedly using the same catalyst three times (as shown in Figure 6c–f). After three consecutive experiments, the SGM-8 composite still showed high photocatalytic activity, indicating better stability. The average hydrogen generation evolution of SGM-8 was approximately 4.2721 mL, which is greater than that of other composites, indicating that the appropriate addition of MoS$_2$ is expected to make the charge separation suppress the electron–hole recombination, as well as enhance the photocatalytic performance. Therefore, this SGM-8 photocatalytic material is a promising catalyst with high efficiency and low cost for the H$_2$ evolution reaction.

Interfacial Interactions of DFT Calculations for the Formation of the GO/SiC/MoS$_2$ Composite. The density functional theory (DFT) method was used to predict the possible active sites of GO–SiC–MoS$_2$ to understand the formation mechanisms. After relaxation, all of the atomic layers were only slightly distorted due to the weak bonding interactions between them. To evaluate the adhesion strength between films in the interface systems, the adhesion work ($W_{ad}$) was calculated according to eq 1

$$W_{ad} = (E_{all} - E_{graphene} - E_{SiC} - E_{MoS_2})/A$$

$E_{all}$ is the total energy of the interface and $A$ is the area of the interface. $E_{graphene}$, $E_{SiC}$, and $E_{MoS_2}$ denote the total energy of the isolated graphene, SiC slab, and the MoS$_2$ slab, respectively. Based on the calculated work of adhesion shown in Figure 7, all of the studied systems show a weak adhesion between the films. The I-a and I-b interface systems show the smallest work of adhesion, indicating that graphene and SiC both prefer to interact with MoS$_2$. The I-a and I-b interface systems show a nearly similar work of adhesion, indicating that the relative positions between graphene only slightly affects the stability of the interface system, while the MoS$_2$ layers have great influence.

Electronic Structures. The densities of states (DOS) of photocatalytic material systems are calculated to study the interaction mechanisms between films of the interface systems. According to the work of adhesion, the relative positions of graphene show a weak influence on the stability of the interface systems. Therefore, only the DOSs of the I-a system are studied. Figure 8 shows the DOSs of the I-a system.

For the total DOSs of interface systems, a small band gap is observed below the Fermi energy level, which is mainly caused by an interaction between films. There are few overlaps between the C1 and S1 orbitals, indicating the weak interactions between them. S2 and S1l show strong interactions in the energy region of $-7.5$ and $-2.0$ eV. Therefore, the interactions between SiC and MoS$_2$ are much stronger than those between graphene and MoS$_2$.

The plane-averaged charge (PAC) distributions of the interface systems are evaluated to investigate the charge transfer near the interface. The PAC describes the charge distributions on a specific plane along a particular direction. The difference of PACs between the interface and separated graphene, MoS$_2$, and SiC are plotted in Figure 9, which will help to describe the charge-transfer mechanisms between the films. The positive and negative values denote the increase and decrease in charge after the formation of the interface, respectively. The PAC difference is mainly distributed in the region of 7.5 and 12.5 Å. The peak zone of approximately 11 Å
is the MoS₂ and SiC interfacial zone of the interface. The PAC distributions between the graphene and MoS₂ interface changed only slightly, indicating weak interactions between them, which is consistent with the above results from the work of adhesion and DOSs.

Possible Photocatalytic Mechanism of the GO/SiC/MoS₂ Composite. Active components, such as h⁺, e⁻, and H⁺, produced in the photocatalytic water splitting to hydrogen, were closely related to the photocatalytic activity of the GO/SiC/MoS₂ composite. As a good two-dimensional graphene-like material, such as GO and MoS₂, the composite can enhance the conductivity of the photocatalytic materials and facilitate good charge transfer. A tentative mechanism proposed for the high H₂ production activity of the GO/SiC/MoS₂ composite is illustrated in Figure 10. Under visible light, the photogenerated electrons of SiC are excited and transferred to MoS₂ through graphene, and hydrogen is generated by reaction with H⁺ under visible-light irradiation.

EXPERIMENTAL SECTION

Chemicals. All chemicals except SiC were analytical grade and used as received without further purification. See S1 (in the Supporting Information) for more information on all chemicals.

Synthesis of SiC/GO/MoS₂ Composites. See S2 (in the Supporting Information) for the preparation of pure SiC. The process for preparing SiC/GO/MoS₂ composites is illustrated in Figure 11. The loading weight percent of Mo was 2.0 wt % based on the total cocatalyst and SiC mass. The GO/SiC/MoS₂ composites were prepared by a simple hydrothermal synthesis. The GO/SiC/MoS₂ composites were prepared as follows: 0.1741 g of MoS₂ was dissolved in 100 mL of deionized water too. Then, 0.05 g of GO and 5.0 g of pure SiC particles were added to the solution and then ultrasonicated for 4 h (25 W L⁻¹ and a frequency of 40 KHz at room temperature). Two drops of ionic liquid ([BMIM]PF₆), was added to the suspension and stirred for 6 h. Then, the mixed suspension was transferred into a 300 mL Teflon-lined stainless steel autoclave and stored in an oven at 200 °C for 20 h. After cooling to room temperature, the precipitate was washed and centrifuged with deionized water until the pH reached 7. The samples were then dried (vacuum) at 60 °C overnight. The resulting products are denoted as SGM-2, indicating an added weight percent of 2 wt % Mo. SiC and GO with MoS₂ (wt %) alone are respectively denoted as SGM-x (x = 2, 5, 8, 10). Based on the best composite photocatalyst, we prepared comparative samples. SiC/MoS₂, SiC/GO, and MoS₂/GO were fabricated in the same condition. These samples are denoted as SM, SG, and MG, respectively.

Characterization of the Prepared Photocatalysts. The morphology of the products was examined using a field-emission scanning electron microscopy (FESEM, TESCAN VEGA II). Transmission electron microscopy (TEM) and high-resolution TEM (HRTEM) images were acquired on a JEOL-2100 electron microscope at an acceleration voltage of 200 kV. The crystal phases of the samples were determined using an X-ray powder diffractometer (XRD) with a Ultima IV diffraction meter (Rigaku, Japan) and Cu Kα radiation source. We scanned the sample for the range of 30–80° at a scanning rate of 4° min⁻¹. The BET surface area was determined from nitrogen adsorption–desorption isotherms using a Micromeritics ASAP 2460. The optical absorption property of the samples was measured by a UV–vis diffuse reflectance spectroscopy (DRS, UH4150, Hitachi) with BaSO₄ as the reflectance standard. Further evidence for the composition of the product was recorded from X-ray photoelectron spectroscopy (XPS) using an ESCALAB 250Xi X-ray photoelectron spectrometer (Thermo Scientific) using an Al Kα radiation.

Photocatalytic Hydrogen Production Activity. The water-splitting reactor is shown in Figure S1 (in the Supporting Information S3). Prior to the photocatalytic reaction, 0.5 g of the photocatalyst was dispersed in 100 mL of distilled water with 0.1 M Na₂S·9H₂O and Na₂SO₄, which have been proved to be one of the most effective electron donors for water-splitting reactions. The solution was maintained at pH 7. After passing N₂ for 30 min to remove the dissolved oxygen, the N₂ valve was closed. Under continuous stirring of 400 rpm, the system was irradiated with a Xe lamp (400–700 nm). A specific amount of the produced gas was collected, and the...
hydrogen content was analyzed using gas chromatography (GC-9790 II, TCD) with argon as the carrier gas. The photocatalytic activity of the samples was evaluated by the rate of H₂ evolution in 4 h.

The photocatalytic H₂ production rate is calculated via eq 2

\[ r_m = \frac{n}{t \times m} \]  

where \( r_m \) is the H₂ production rate (\( \mu \text{mol-g}^{-1}\cdot\text{h}^{-1} \)) and \( n \) is moles of H₂ evolved in 4 h (\( \mu \text{mol} \)). The calculated H₂ concentration was measured with GC through a standard curve; \( t \) is the duration of the photocatalytic reaction (h); and \( m \) is the mass dose of photocatalyst (g).

The QY defined with the following eqs 3–6, was measured using a band-pass interference filter (\( \lambda = 400–700 \text{ nm}, \text{Edmund Optics} \)) and the spectroradiometer

\[ \text{QY} = \frac{r \times 2}{N} \times 100\% \]  

\[ N = S \times I \]  

\[ I = \int_{\lambda_1}^{\lambda_2} R \, d\lambda \]  

\[ r = \frac{n}{t} \]  

where \( S \) is the cross-sectional area of the reactor (\( \text{cm}^2 \)); \( N \) is the number of incident photons (einsteins-s⁻¹); \( R \) is the irradiance at a given wavelength (einsteins cm⁻²-s⁻¹-nm⁻¹), when the wavelength is 400–700 nm, \( R = 2 \times 10^2 \); \( I \) is the light intensity (einsteins cm⁻²-s⁻¹); \( \lambda \) is the irradiation wavelength (nm), and \( \lambda_1 \) and \( \lambda_2 \) are the lower and upper limits of the irradiation wavelengths, respectively; \( r \) is the H₂ production rate (\( \mu \text{mol-h}^{-1} \)); and \( n \) is the moles of H₂ evolved per 4 h time (\( \mu \text{mol} \)).

**Calculation Methods.** The total energy and electronic structure of all catalysts were calculated using DFT via VASP code with projector-augmented wave Perdew–Burke–Ernzerhof pseudopotential. Various interfacial systems were constructed. The energy cutoff of the plane-wave basis was set to 450 eV, and only the \( k \) point was used for the relaxations of the interfacial systems because we had a large supercell. The DFT-D3 method was used to consider the van der Waals interactions between films. To simulate the interactions between graphene, MoS₂, and SiC, all the atoms in graphene and MoS₂ slabs were relaxed, and the two bottom Si–C layers were fixed to simulate the bulk SiC systems. The simulation models are shown in Figure 12. Single graphene and MoS₂ layers, four SiC layers, and a 1.5 nm vacuum layer comprise the models. Eight interface models with different terminations were considered to study the interactions between graphene, MoS₂, and SiC.

**CONCLUSIONS**

A novel GO/MoS₂ hybrid structure decorated with SiC particles was synthesized by a facile one-step soft hydrothermal process using a green photocatalyst for solar energy production of hydrogen. The as-prepared GO/SiC/MoS₂ composite photocatalysts show a highly enhanced photocatalytic activity for the solar energy production of hydrogen as compared to that of the pure SiC photocatalyst. By optimizing the loadings of MoS₂, the GO/SiC/MoS₂ composite showed the highest photocatalytic H₂ production activity when the content of the MoS₂ catalyst was 8.0 wt % with a small amount of graphene. The calculation results of DFT and the experimental results indicated that the interactions between SiC and MoS₂ are much stronger than those between graphene and MoS₂. It is believed that the interfacial photocatalytic activity between GO and MoS₂ in the GO/SiC/MoS₂ composite is believed to be small. The incorporation of graphene enhances the charge-transfer abilities and retards the recombination of the photogenerated carrier, improving the photocatalytic H₂ production activities. Therefore, noble-metal-free GO/SiC/MoS₂ composite photocatalyst has the potential for H₂ production under visible-light irradiation.

**ASSOCIATED CONTENT**

**Supporting Information**

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.9b01832.

Chemical (S1); preparation of pure SiC (S2); water-splitting reactor (S3); scanning electron microscopy (SEM) of SGM-2, SGM-5, and SGM-10 (S4); and specific surface area and pore volume of composites (S5) (PDF)

**AUTHOR INFORMATION**

*Corresponding Author*
E-mail: tougaoktz@163.com.

**ORCID**

Yan Zhang: 0000-0002-7267-3269
John C. Crittenden: 0000-0002-9048-7208

**Notes**

The authors declare no competing financial interest.

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