EXPERIMENTAL SECTION

Materials

Commercially available MoO_3 Powder was purchased from Sigma Aldrich (Product Number: 267856). According to the description of products, the starting MoO_3 powder has representative lateral particle sizes in the range of 6-40 μm. Nickel(II) nitrate hexahydrate ($Ni(NO_3)_2 \cdot 6H_2O$) and ethanol were purchased from Sinopharm Chemical Reagent Co., Ltd. (China) and used without further purification. The butterfly wings were supplied by Hunan Dingcheng Butterfly Art Gift Shop. CO_2 with purity of 99.99% was provided by the Zhengzhou Shuangyang Gas Co. and used as received. Aqueous solution was prepared with deionized water.

Exfoliation of MoO_3 nanosheets:

Typically, 200 mg MoO_3 powder was added into a 50 mL beaker using 10 mL ethanol/water mixture(5:5/v:v). The mixture was then sonicated in an ice bath continuously for 24h to exfoliate the bulk MoO_3 into few layers or single layer nanosheets. Finally, the dispersion was centrifuged at 3000 rpm for 45 minutes to remove aggregates and the supernatant was taken out by a pipet. The concentration of as-prepared MoO_3 nanosheets was determined by high-speed centrifugation and followed by weighing the aggregates. The concentration of as-prepared MoO_3 is 2mg/mL.

Synthesis of Ni_xMoO_3 nanodots

Firstly, 10mL MoO_3 dispersion and 5.7mg $Ni(NO_3)_2 \cdot 6H_2O$ were added into a beaker and sonicated for 1 hour. Then the dispersion was quickly transferred into the supercritical CO_2 apparatus composed mainly of a stainless-steel autoclave (25 mL) with a heating

jacket and a temperature controller. The reactor was charged with CO_2 up to 10MPa at 100 °C. Subsequently, the temperature was slowly elevated to 200°C and maintained for 6 hours with magnetic stirring. Then the reactor was cooled naturally to room temperature. After depressurization, the product was collected and centrifuged at 5000 rpm for 10 minutes, then the supernatant was taken out by a pipet and dried in vacuum at 100 °C for 1 hour.

Preparation of Ni_xMoO_3 nanodots-butterfly wing composite.

Firstly, the butterfly wings were washed with distilled water and ethanol. After drying under the natural conditions at room temperature, the butterfly wings were modified by applying the Ni_xMoO_3 nanodots dispersion (10mL, 1 mg mL^{-1}) onto the photonic dorsal side of the wing and allowing them to air-dry for several minutes.

Characterization

TEM (JEOL JEM-2100) were used to study the morphology of the nanomaterials. The Raman measurements were carried out on a Renishaw Microscope System RM2000 with a 50mW Ar+ laser at 514.5 nm. An X-ray photoelectron spectroscopy was performed using a Thermo ESCALAB 280 system with Al/K (photon energy =1486.6 eV) anode mono X-ray source. The X-ray diffraction (XRD) patterns were recorded on a Netherlands X′Pert PRO X-ray diffractometer. The IR camera (FLIR-E6390) was used to test the temperatures under the light irradiation.

Computational Details

The first-principles calculations based on density functional theory were performed by using the CASTEP plane-wave pseudopotential package with Perdew-Burke-Ernzerhof exchange-correlation function. The MoO_3 simulation supercell (Mo_8O_{24}) consists of 2*1*1 MoO_3 unit cells of 8 Mo atoms and 24 O atoms. The cutoff energy of the plane-wave basis was 570 eV, for MoO_3 unit cell, 5*2*5 k-points meshes was used for the Brillouin zone sampling to ensure the accuracy of the calculation results. Moreover, the electron-ion interactions were described by the ultrasoft pseudopotentials (USPPs), and the DFT-D2 method was used for dispersion corrections.

The self-consistent field (SCF) calculation was kept within the energy convergence criterion of 1×10^{-6} eV/atom. In the process of geometric optimization, the total energy converged to 1×10^{-5} eV/atom, and the maximum force converged to 0.03 eV/Å, while the maximum stress converged to 0.05 GPa, and the maximum atom displacement converged to 0.001 Å. All structures including the lattice parameters and the atomic positions were fully optimized by using the BroydenFletcher-Goldfarb-Shanno (BFGS) minimization scheme. The lattice constants of the optimized pristine unit cells of MoO_3 are a=3.91 Å, b=14.2 Å, c=3.69 Å.

Photothermal experiments

The photothermal effect was measured by monitoring the temperature of the Ni_xMoO_3 nanodots dispersions in DI water at various concentrations (0, 25, 50, 100, 200, 300, 400, 500 $\mu g \cdot mL^{-1}$). Briefly, 1.0 mL of the sample dispersion in a glass cuvette (total volume of 2 mL) was irradiated by the 808 nm laser (1.0 $W \cdot cm^{-2}$), and the temperature of the solution was measured every 30 s by a thermocouple microprobe submerged in the solution. The temperature of 1.0 mL of DI water upon the same irradiation was measured as a control

experiment. To evaluate the single wavelength photothermal conversion efficiency, the dispersion (500 μg·mL^{-1}, 1mL) was irradiated under the 808 nm laser (1.0 W·cm^{-2}) for 480 s. After switching off the laser, the temperature was further measured for another 720 s. Single wavelength photothermal conversion efficiency (η) was then calculated according to the reported method. To investigate the photothermal stability of the sample, the sample dispersion (500 μg·mL^{-1}, 1.0 mL) was irradiated by the 808 nm laser (1.0 W·cm^{-2}) for 480 s, and then the laser was off for another 720 s. Such cycle was repeated for six times.

Calculation of the photothermal conversion efficiency

The photothermal conversion efficiency of Ni_xMoO_3 nanodots was calculated according to previous reports. The detailed calculation was carried out as following equations:

The total energy balance of this system as following equation:

$$\sum_i m_i C_{p,i} \frac{dT}{dt} = Q_{NDs} + Q_s - Q_{loss} \quad (3)$$

where m and C_p are the mass and heat capacity, respectively. The suffix "i" of m and C_p refers to solvent(water) or dispersed matter (nanodots). T is the solution temperature. Q_{NDs} is the photothermal energy absorbed by Ni_xMoO_3 nanodots per second:

$$Q_{NDs}=I(1-10^{-A\lambda})\,\eta \quad (4)$$

where I is the laser power, A_λ is the absorbance of Ni_xMoO_3 nanodots at the wavelength of 808 nm in aqueous solution, and η is the photothermal conversion efficiency of Ni_xMoO_3 nanodots which means the ratio of absorbed light energy converting to thermal energy. Q_{loss} is thermal energy lost to the surroundings:

$$Q_{loss} = hA\Delta T \quad (5)$$

Where h is the heat transfer coefficient, A is the surface area of the container, and ΔT is the changed temperature.

Q_s is the heat associated with the light absorbed by solvent per second. In the situation of heating pure water, the heat input is equal to the heat output at the maximum steady-statue temperature, so the equation can be:

$$Q_s = Q_{loss} = hA\Delta T_{max,H2O} \quad (6)$$

Where $\Delta T_{max,H2O}$ is the temperature change of water at the maximum steady-state temperature.

As to the experiment of Ni_xMoO_3 nanodots dispersion, the heat inputs are the heat generated by nanodots (Q_{NDs}) and the heat generated by water (Q_s), which is equal to the heat output at the maximum steady-statue temperature, so the equation can be:

$$Q_{NDs} + Q_s = Q_{loss} = hA\Delta T_{max,mix} \quad (7)$$

Where $\Delta T_{max,mix}$ is the temperature change of the Ni_xMoO_3 nanodots dispersion at the maximum steady-state temperature.

According to the equation (4), (6) and (7), the photothermal conversion efficiency (η)

can be expressed as following:

$$\eta = \frac{hA(\Delta T_{max,mix} - \Delta T_{max,H_2O})}{I(1 - 10^{-A_\lambda})} \quad (8)$$

In this equation, only hA is unknown. In order to get the hA, we introduce θ, which is defined as the ratio of ΔT to ΔT_{max}

$$\theta = \frac{\Delta T}{\Delta T_{max}} \quad (9)$$

Substituting equation (7) into equation (1). When the laser was shut off, the $Q_{NDs} + Q_s = 0$. Meantime, compared with solvent (water, 1g), mass of NPs (500μg) was too little. Generally, the specific heat of water is much higher than other materials. Consequently, the m_{NPs} and $C_{p,NPs}$ of nanodots were neglected. So, hA can be expressed as following:

$$hA = \frac{mC_p}{\tau_s} \quad (10)$$

$$t = -\tau_s \ln\theta \quad (11)$$

Thus, the photothermal conversion efficiency η can be obtained from equation 8.

In this work, m and C_p are the mass (1.0 g) and the thermal capacity (pure water data of 4.2 J • g^{-1} • °C $^{-1}$ was used). By linear fitting cooling time (t) to negative natural logarithm of temperature (- ln θ), τ_s was determined to be 224.5s for Ni_xMoO_3 nanodots in the concentration of 500 μg mL^{-1}(Figure 4c and Figure S8). Thus, the η value was calculated to be 87.4% for Ni_xMoO_3 nanodots

Solar water evaporation.

Solar water evaporation by the Ni_xMoO_3 nanodot-Butterfly wings film was tested in a 50 mL glass beaker. For each run, the beaker was filled with 40 mL of distilled water, and the Ni_xMoO_3 nanodot-Butterfly wings film floated on the water surface. A 300 W xenon lamp was used with an AM 1.5 filter to simulate the solar irradiation. The light intensity was 0.3 kW·m^{-2}. After the given durations of the irradiation, the weights of water in the beaker were recorded. Butterfly wing was measured as the control group. The

evaporation rate (v) of Ni_xMoO_3 nanodot-Butterfly wings film was calculated by following equation:

$$v = \frac{dm}{s \times dt} \tag{12}$$

Where s is the evaporation surface, $\frac{dm}{dt}$ is the mass change rate.

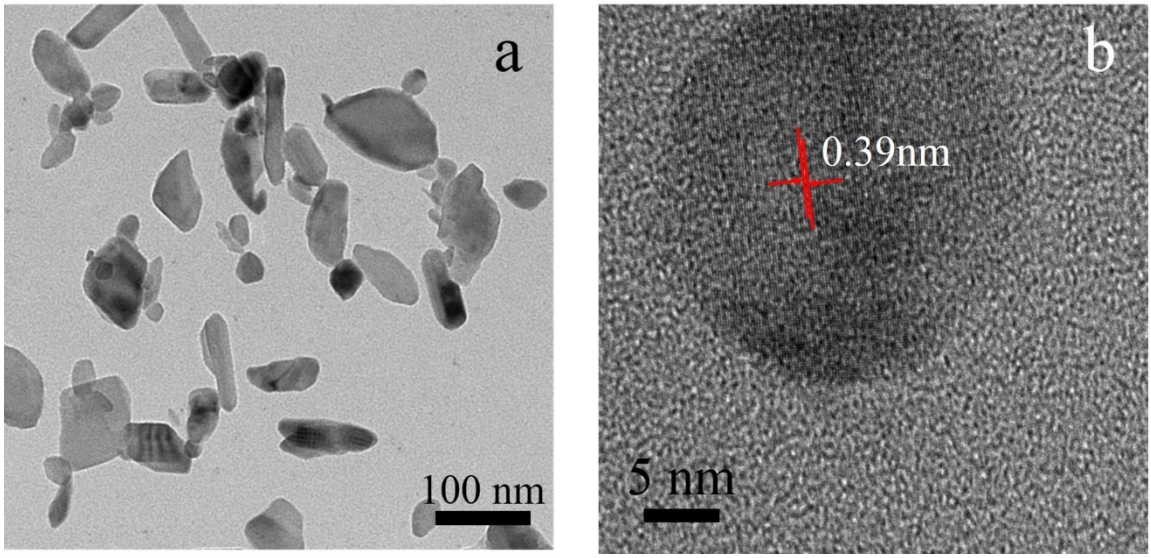

Figure S1. TEM and HRTEM images of as-prepared MoO_3 nanosheets.

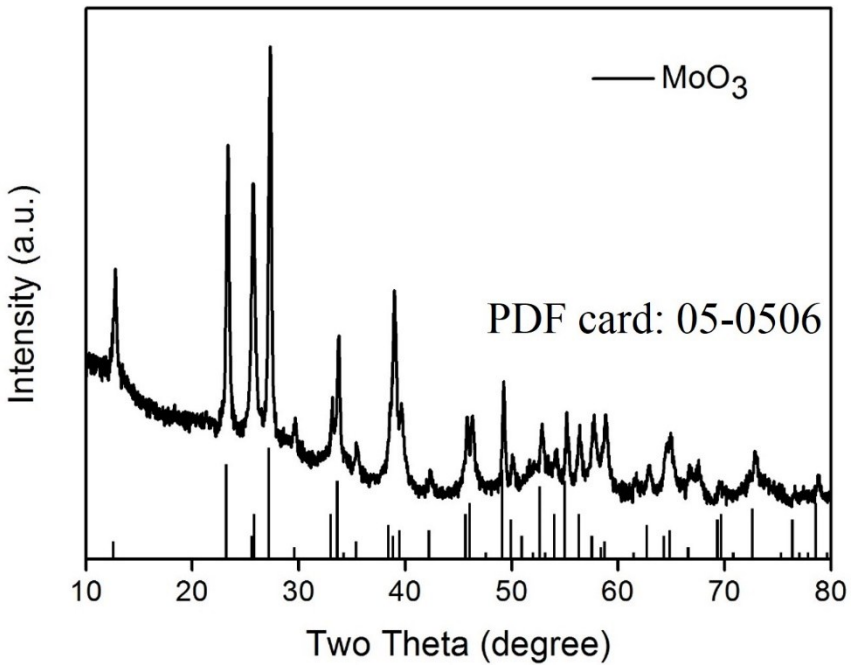

Figure S2 XRD pattern of sonication-assisted exfoliated MoO_3 nanosheets

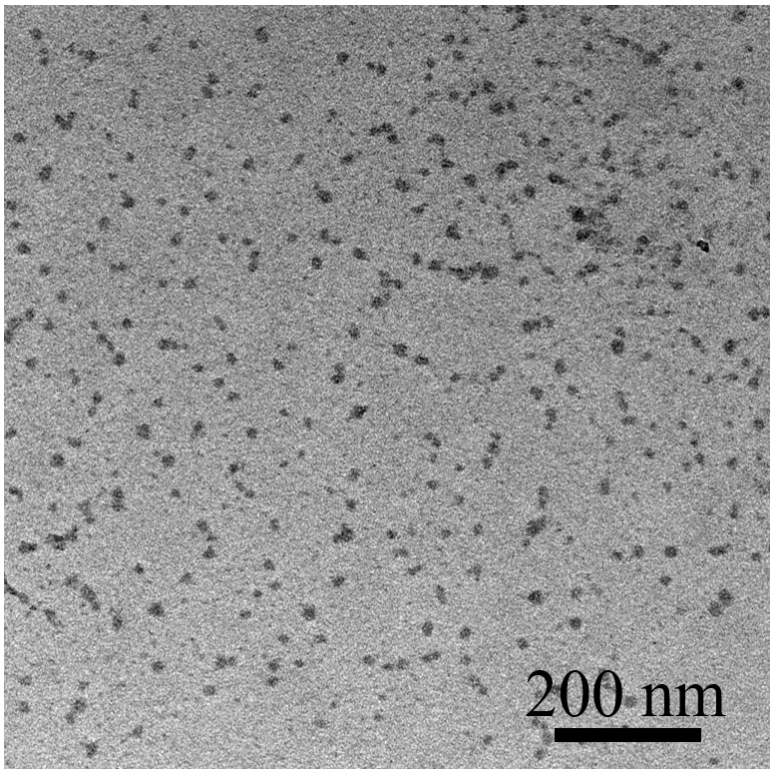

Figure S3. TEM image of the as-prepared Ni_xMoO_3 nanodots.

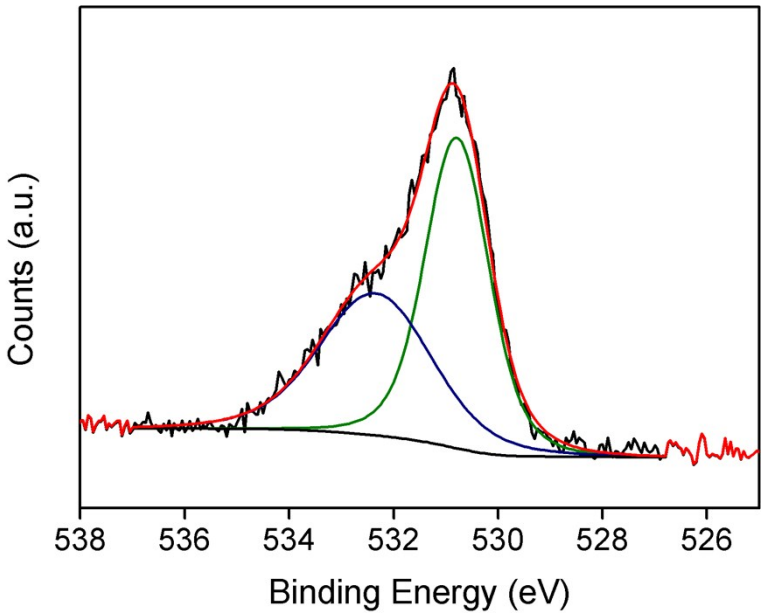

Figure S4. XPS of O 1s spectra of the Ni_xMoO_3 sample

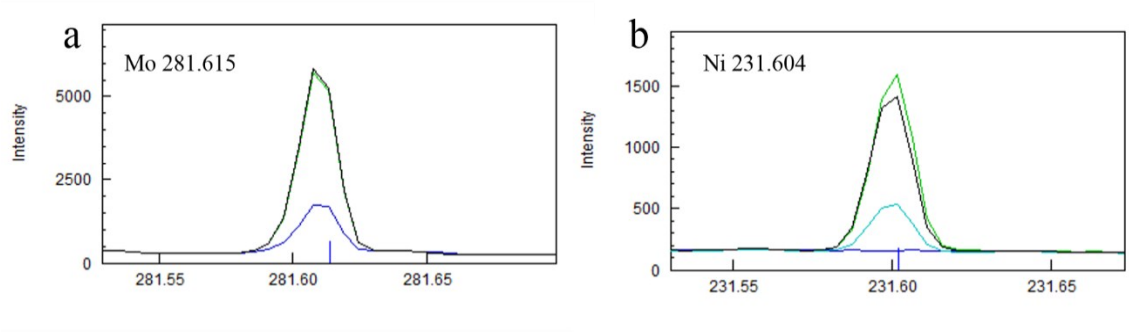

Figure S5. The ICP-AES spectra of Mo(a) and Ni(b) in Ni_xMoO_3 nanodots.

(a) Blue line represents the standard solution of Mo ion, the black and the green line represent the Mo ion in solution (I) and solution (II) of Ni_xMoO_3 nanodots, respectively.

(b) Black line represents the standard solution of Ni ion, the cyan line and the green line represent the Ni ion in solution (I) and solution (II) of Ni_xMoO_3 nanodots, respectively.

Table S1 The ICP-AES results calculated from Figure S3

	Solution（I）	Solution（II）
Mo	176 μg mL^{-1}	180 μg mL^{-1}
Ni	13.5μg mL^{-1}	46.1μg mL^{-1}

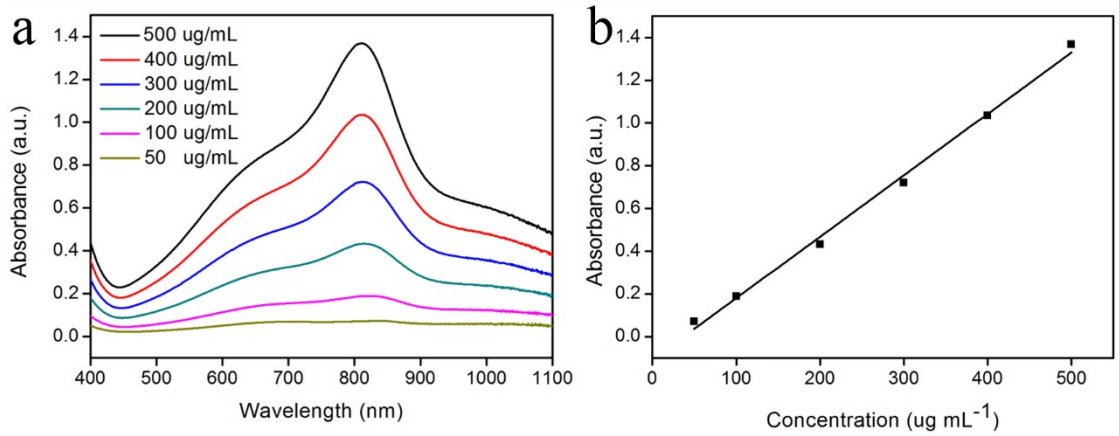

Figure S6 a) UV−Vis−NIR absorption spectra and b) fitting curve of the absorption values at 808 nm of the $Ni_{0.125}MoO_3$ nanodots of dispersions at different concentrations in water.

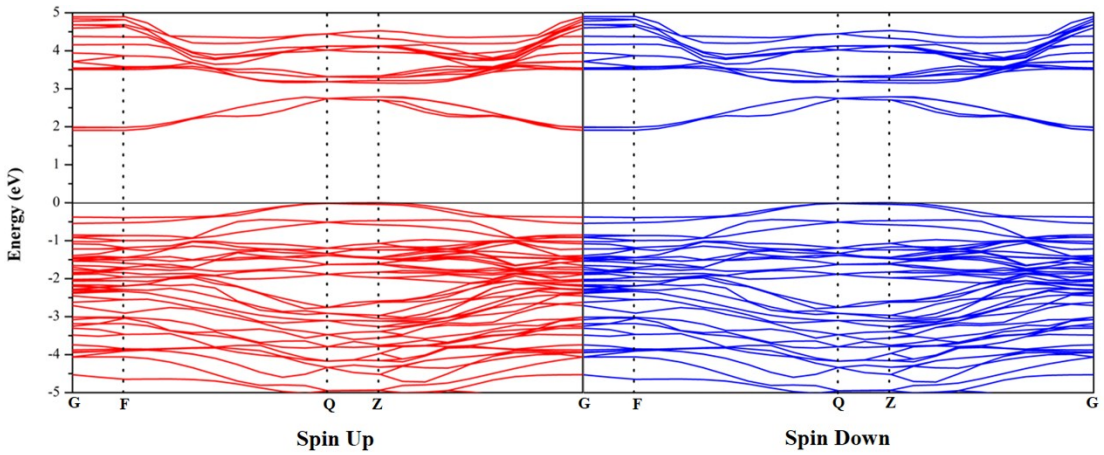

Figure S7 The band structure of bulk MoO_3. The band gap value is 1.9eV.

Table S2 The supercell lattice constants of MoO_3 and $Ni_{0.125}MoO_3$.

	a (Å)	b (Å)	c (Å)	alpha (°)	beta (°)	gamma (°)
MoO_3	7.83	14.23	3.69	90.00	90.00	90.00
$Ni_{0.125}MoO_3$ (Model 3)	7.61	15.10	3.72	88.38	90.00	90.13

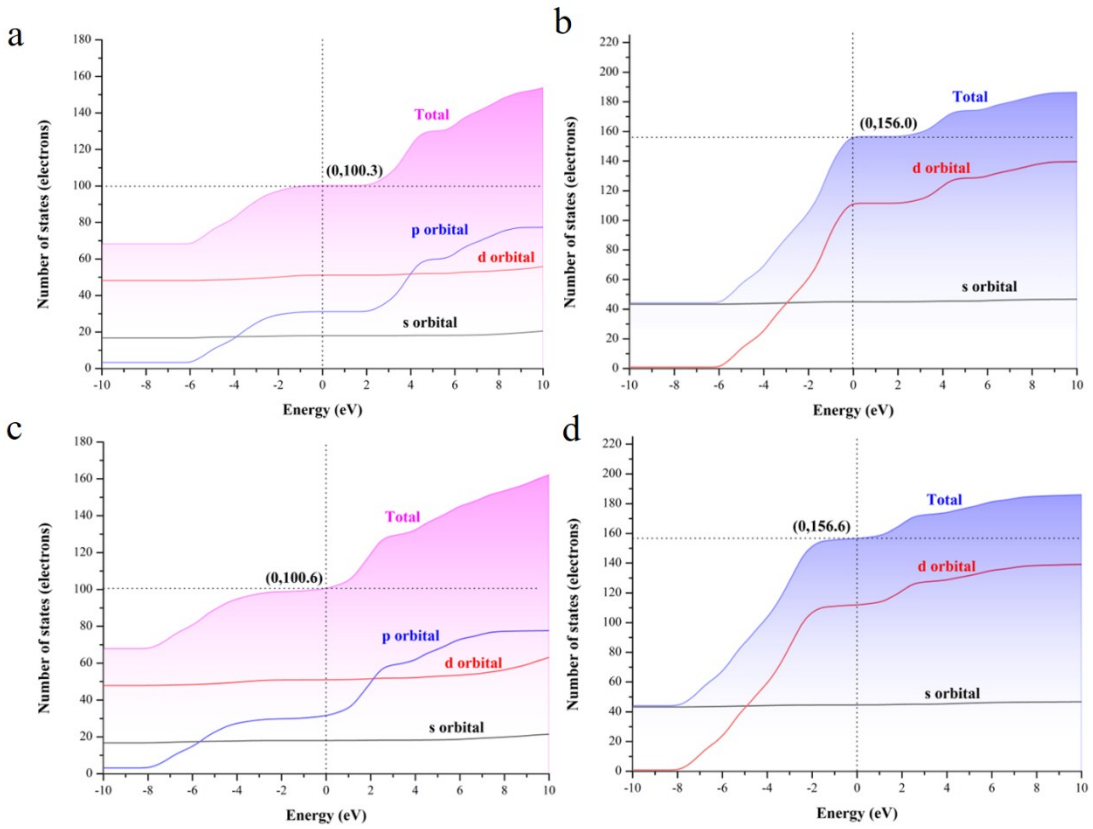

Figure S8 The integrated densities of states of MoO_3 and $Ni_{0.125}MoO_3$. Mo atom (a) and O atom (b) in MoO_3, and Mo atom (c) and O atom (d) in $Ni_{0.125}MoO_3$. The integrated densities of states indicates that the inserted Ni atoms transfer electrons to MoO_3, and all Mo atoms and O atoms have obtained some electrons.

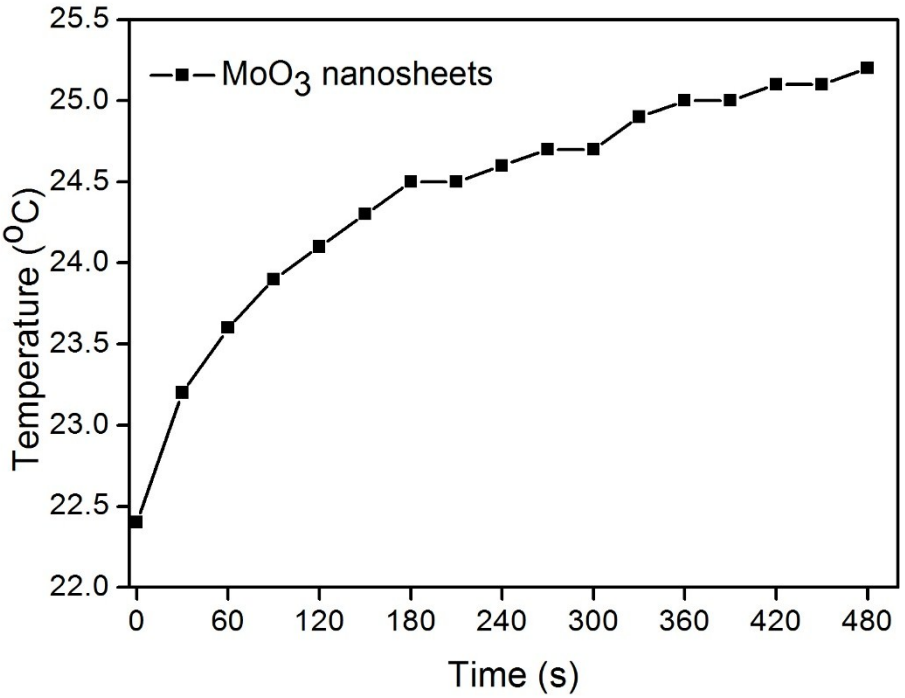

Figure S9. Temperature change of MoO_3 nanosheets suspension at 500ug/mL.

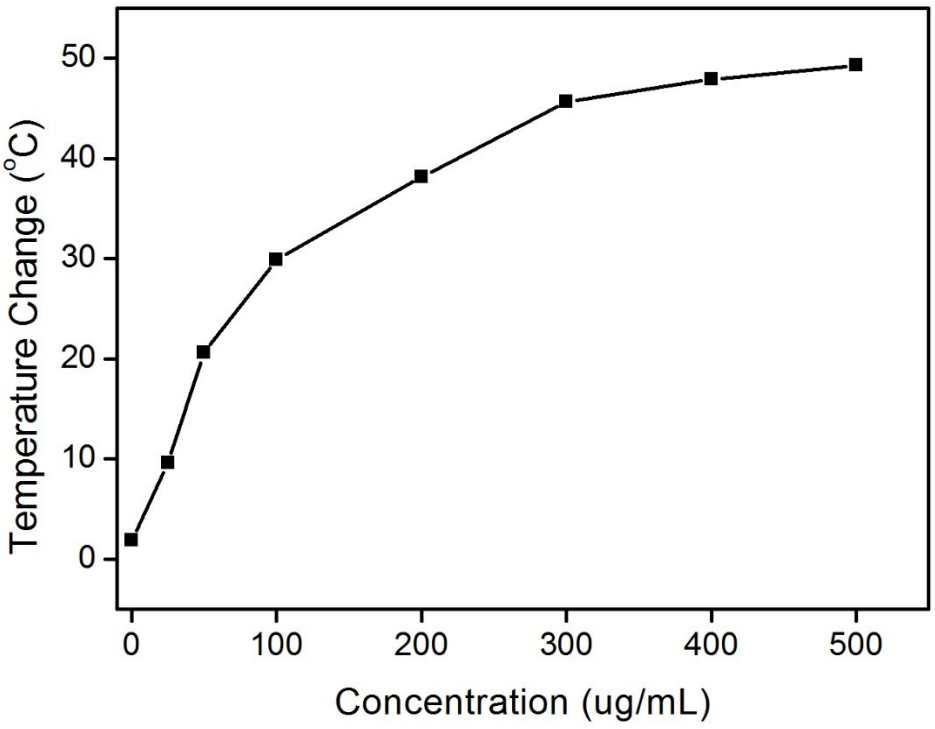

Figure S10. Plot of temperature variation upon the 8-min irradiation *versus* the different concentration suspension of $Ni_{0.125}MoO_3$ nanodots.

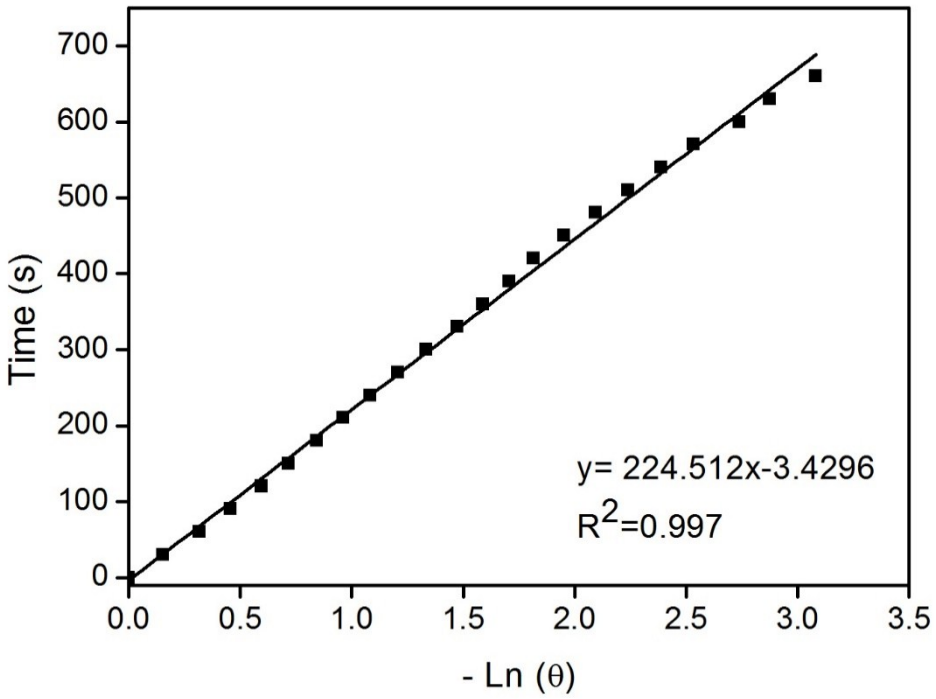

Figure S11. The determination of the time constant for heat transfer of the system using linear regression of the cooling profile of $Ni_{0.125}MoO_3$ nanodots.

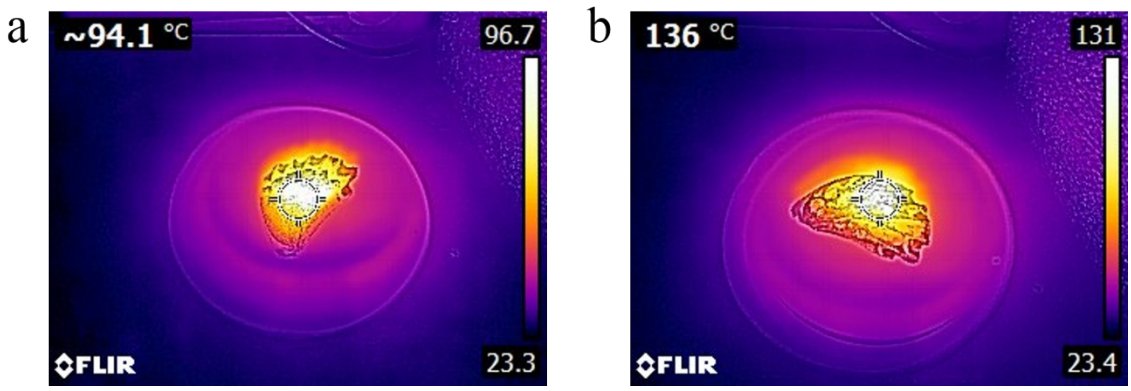

Figure S12. The IR photographs of natural butterfly wing(a) and (b) the $Ni_{0.125}MoO_3$ nanodot- butterfly wing composite under simulated solar irradiation (0.3kW m^{-2})

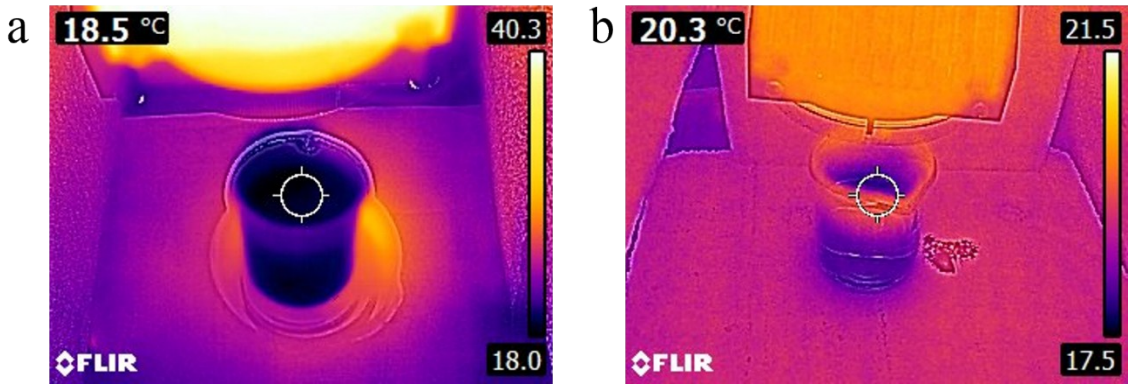

Figure S13. IR photographs of 50mL beakers without the $Ni_{0.125}MoO_3$ nanodot- butterfly wing composite on top of the water surface before the solar irradiation(a) and after the solar irradiation for 3 hours(b).