Electronic Supplementary Information

Experimental section

Materials

Sodium hypophosphite (NaH_2PO_2) was purchased from Aladdin Ltd. (Shanghai, China). $Ni(NO_3)_2.6H_2O$, urea, and NH_4F were bought from Beijing Chemical Corporation. NF was provided by Hongshan District, Wuhan Instrument Surgical Instruments business. All the reagents were used as received. The water used throughout all experiments was purified through a Millipore system.

Preparation of $Ni(OH)_2$ NA/NF:

Typically, a piece of NF (2 × 3 cm^2) was washed with HCl, ethanol and deionized water several times to ensure the surface of the NF was well cleaned before use. The cleaned NF was immersed into a 40 mL aqueous solution containing 4 mmol $Ni(NO_3)_2.6H_2O$, 20 mmol urea, and 8 mmol NH_4F at room temperature. The aqueous solution and the NF were transferred to a 50 mL Teflon-lined stainless-steel autoclave and maintained at 120 °C for 6 h, and then allowed to cool down. Then the NF with precursor was washed with water several times and dried in oven.

Preparation of Ni_2P NA/NF:

$Ni(OH)_2$ NA/NF and NaH_2PO_2 were placed at two separate positions in one closed porcelain crucible with NaH_2PO_2 at the upstream side of the furnace. Subsequently, the samples were heated at 280 °C for 2 h with a heating speed of 2 °C min^{-1} in Ar atmosphere. Ni_2P NA/NF was collected after cooled to ambient temperature under Ar.

Characterizations

XRD data were acquired on a RigakuD/MAX 2550 diffractometer with Cu Kα radiation (λ=1.5418 Å). XPS measurements were performed on an ESCALABMK II X-ray photoelectron spectrometer using Mg as the exciting source. The C 1s photoelectron line at 284.6 eV was used as an internal standard for the correction of the charging effect in all samples. The signal was filtered with a XPSPEAK41 software. SEM measurements were carried out on a Hitachi S-4800 field emission scanning electron microscope at an accelerating voltage of 20 kV. HRTEM image was collected on a Hitachi H-8100 electron microscope with an accelerating voltage of 200 kV. Surface areas and pore sizes for Ni_2P nanosheets scratched down from NF were measured by the Brunauer–Emmett–Teller (BET) method and Barrett-Joyner-Hallender (BJH) method at 77.0 K using a Quantachrome NOVA 1000 system. The horizontal axis was normalized with the vapor pressure of nitrogen at 77.0 K (= 0.101 MPa). The sample were heated at 300°C under vacuum for 6 hours before the measurements. Inductively coupled plasma mass spectrometry (ICP-MS) analysis was performed on ThermoScientific iCAP6300.

Hydrogen generation measurements

The volumes of hydrogen were measured using water-displacement method. Typically, the hydrolytic dehydrogenation experiments were performed in a 25 mL two-necked round-bottom flask containing 2 ml of AB solution with one neck connected to a gas burette and the other sealed with a rubber cap. The temperature of the reaction was kept at the desired value by using a constant temperature bath to

achieve thermal equilibrium. Then Ni_2P NA/NF monolithic catalyst was transferred into reaction flask. The mass of Ni_2P was accurately determined by ICP-MS as follows. The amount of P, m(P), was measured to be 0.086 mg cm^{-2}. Thus, the mass of Ni_2P grown on the NF (1*1 cm^2), m(Ni_2P), was calculated as 0.4 mg according to the equation: m(Ni_2P) = m(P) * M(Ni_2P)/M(P) = 0.086*148.4/32.

TOF calculation

The TOF($mol_{(H2)}$ $mol_{(Ni2P)}^{-1}$ min^{-1}) was calculated using the following equation:

$$TOF = P*(HGR10^{-6})* M / RTm$$

where HGR is initial hydrogen generation rate (2.8 ml g^{-1} min^{-1}), P is gas pressure ($1.01*10^5$ Pa), M is molar mass (148.4 g mol^{-1}), R is ideal gas constant (8.314 J mol^{-1} K^{-1}), T is temperature (298 K) and m is the mass of Ni_2P (0.4 mg).

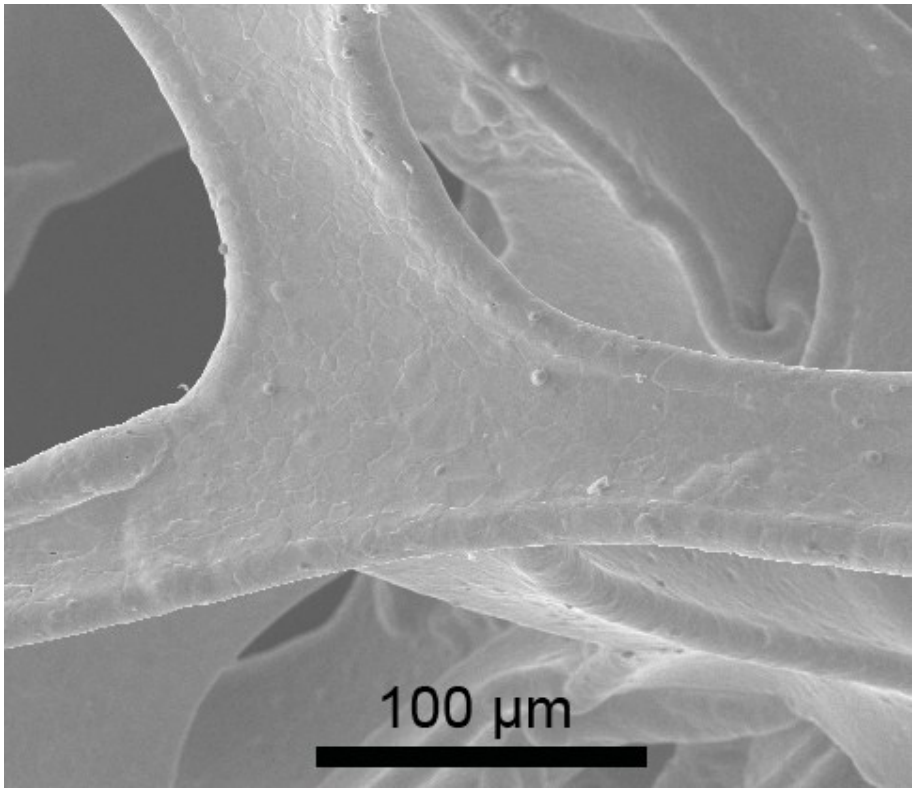

Fig. S1. SEM image of bare NF.

Fig. S2. Optical photograph of hydrogen evolution measurement system.

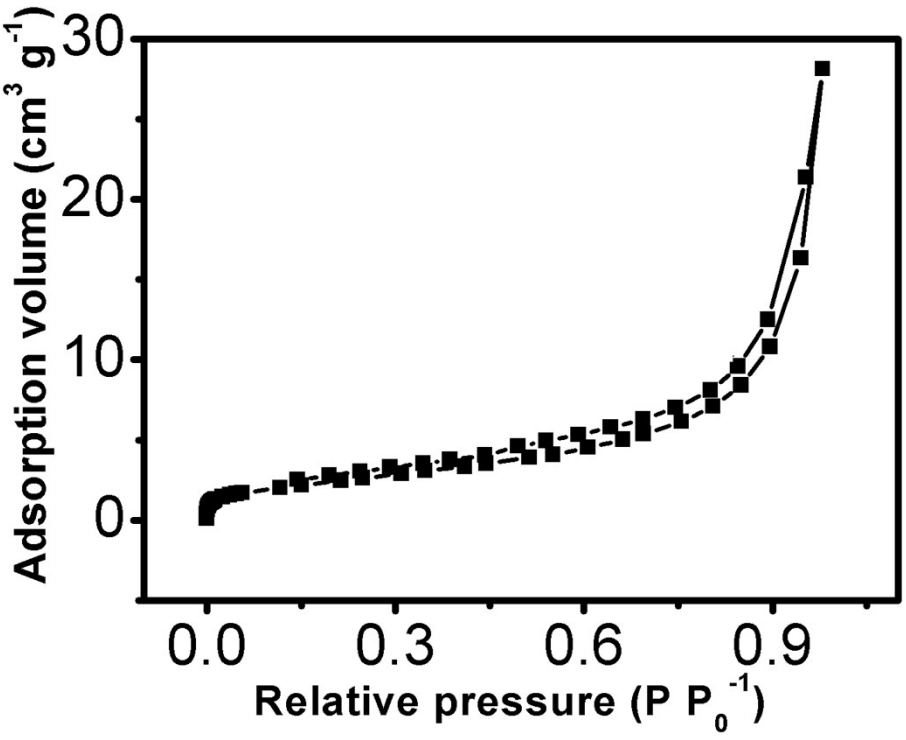

Fig. S3. Nitrogen adsorption/desorption isotherm curve for Ni_2P nanosheets after recycling usage.

Movie S1. This movie shows hydrogen evolution on Ni_2P NA/NF for on-demand hydrolytic dehydrogenation of AB (1 wt% AB under ambient conditions).