Supporting Information

Great Improvement in Pseudocapacitor Properties of Nickel Hydroxide via Simple Gold Deposition

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Chemicals used

Nickel Nitrate hexahydrate (sigma-Aldrich, >99%), Hexamethylenetetramine (HMTA) (Sigma-Aldrich,>99%), Hydrogen tetrachloroaurate (Alfa-aesar, >99.999%). All chemicals were used without any further purification.

Synthesis of Ni(OH)₂ structures

To synthesize 3D-Ni(OH)₂ structures, a 300 mM nickel nitrate solution (40 ml), and 300 mM HMTA (40 ml) were mixed well before transferring them to a 120 ml Teflon lined stainless steel autoclave. The autoclave was then heated at 100 °C for 4 hours. The final product was washed and filtered thoroughly using DI water and ethanol. The synthesized powder was dried at 80 °C for 12 hours under vacuum, yielding 3D-Ni(OH)₂ structure powders.

Synthesis of Au/Ni(OH)₂

The gold nanoparticles were coated by using a colloidal deposition method. Briefly, 0.3 g of Ni(OH)₂ was mixed with 30 mM of chloroauric acid and dissolved in ethanol, to which 1 wt%

of sodium citrate dissolved in water was added. The mixture was heated to 100 °C for 30 min and then allowed to cool naturally along with the substrate. The resulting solution was dried at 80 °C for 12 hours.

Characterization

The synthesized structures were characterized using FE-SEM (SEM, FEI/USA Nanonova 230) and high-resolution transmission electron microscopy (FETEM, JEOL TEM 2100) at an accelerating voltage of 200 kV. The crystallinity of the samples were checked using a X-ray diffraction system (Bruker D8 Advance system) with Cu Ka radiation ($\lambda = 1.5406$ A) at a scan rate of 2 °/min the 20 range from 20° to 80°. The specific surface area, pore size, and pore volume were analyzed using the Brunauer–Emmett–Teller (BET) method with a Belsorp max system (Bel Japan).



Figure S1. XRD patterns of $Ni(OH)_2$ (in black) and $Au/Ni(OH)_2$ (in red). An intentionally increased amount of Au NPs were deposited in the Au/Ni(OH)₂ sample in order to confirm the presence of Au NPs.

Electrochemical Measurements.

The working electrode was prepared by mixing the electro-active material Ni(OH)₂ or Au/Ni(OH)₂, 85 wt%), acetylene black (10 wt%), and polyvinylidene difluoride (PVDF, 5 wt%) as a binder. The mixture was then pasted onto a Ni foam and dried at 150 °C in an air atmosphere for 2 hr. The electrochemical performance of the nanostructured Au/Ni(OH)₂ was evaluated using Pt foil as a counter electrode and a KOH solution (1 M) as an electrolyte. Galvanostatic charge–discharge and cyclic voltammetry were measured using a computer controlled electrochemical interface (VMP3 biologic) from 0.2 V to 0.6 V at room temperature. Electrochemical impedance spectroscopy (EIS) was analyzed at a frequency range from 100 kHz to 0.1 Hz using a potentiostat (Versa STAT 3, AMETEK).

Optimization of the amount of Au NPs

The amount of Au NPs was controlled by varying the concentration of the gold precursor solutions at a fixed concentration of metal hydroxide and it was measured by inductively coupled plasma-mass spectrometer (ICP/MS). The optimized ratio of Au NPs to NiO was 0.3 wt% which shows the capacitance value of 1,927 F/g at a scan rate of 1 A/g.



Figure S2. SEM image of Au/Ni(OH)₂ showing the variation in coverage rather than the size of Au NPs by varying the content of Au (by varying the feed ratio of Au precursor to Ni(OH)₂.

	Au amount (mg/kg)	Au amount (wt%)	Capacitance (F/g)
Sample 0	0	0	1,483
Sample 1	4,736	0.47	1,781
Sample 2	6,204	0.62	1,927
Sample 3	7,343	0.73	1,697
Sample 4	8,559	0.86	1,550
Sample 5	11,130	1.11	1,049
Sample 6	17,773	1.77	852



Figure S3. Comparison of capacitance with the various contents of Au NPs.



 $\Phi_{Au} > \Phi_{Ni(OH)2}$



Figure S4. Ohmic contact between Au NP metals and $Ni(OH)_2$ semiconductor: (a) Band alignment of Au and $Ni(OH)_2$. (b) Current–Voltage characteristics of Au/Ni(OH)₂ and $Ni(OH)_2$ pellets.



Figure S5. Schematic diagram of the electron transfer pathways in Ni(OH)₂ and Au/Ni(OH)₂.: Au/Ni(OH)₂ has a shorter and faster pathway for the electron transfer via highly conductive metal/semiconductor contact, resulting in the enhanced conductivity.



Figure S6. (a) SEM image and (b) XRD patterns of $Au/Ni(OH)_2$ after 5000 cycles at 50 mV/s.

Table 1 Specific capacitance, power density, and energy density of the Ni(OH)₂ electrode at

 different current densities

Current density (A/g)	1	2	5	10	15	20
Specific capacitance (F/g)	1,363	1,130	912	851	777	655
Power density (W/kg)	200	400	1,000	2,000	3,000	4,000
Energy density (Wh/kg)	30	25	20	19	17	14

Table 2 Specific capacitance, power density, and energy density of the Au/Ni(OH)₂ electrode

 at different current densities

Current density (A/g)	1	2	5	10	15	20
Specific capacitance (F/g)	1,927	1,765	1,506	1,450	1,391	1,274
Power density (W/kg)	200	400	1,000	2,000	3,000	4,000
Energy density (Wh/kg)	42	39	33	32	31	28



Figure S7. Ragone plot of Ni(OH)₂ (Black line) and Au/Ni(OH)₂ (Red line)

For other metal oxide systems

Synthesis of metal oxides

To synthesize 3D NiO, CuO, MnO₂ structures, a solution A, B, and C, respectively, was transferred to the 120 ml teflon-lined stainless steel autoclave. The autoclave was then heated at 120°C for 4 hours. The final product was washed and filtered thoroughly using D.I water and ethanol. The synthesized powder was dried at 80°C for 12 hours. Finally, the dried sample was annealed at 500°C for 2 hours.

МО	Solution	Compositions
NiO A	٨	300 mM of nickel nitrate solution (40 ml), 300 mM of HMTA
	11	(20 ml), and 10 mM of lithium hydroxide (20ml)
CuO B	в	100 mM of copper sulfate solution (40 ml) and 1 M of sodium
	D	hydroxide solution (40ml)
MnO ₂	С	200 mM of manganese sulfate solution (40 ml) and 5 ml of
		ammonium hydroxide solution

Au NPs on metal oxides (Au/MOs)

The Au NP coating was done using the colloidal method. Briefly 0.3 g of metal oxides (NiO, CuO, and MnO₂) was mixed with 30 mM of chloroauric acid, dissolved in ethanol, to which 1 wt% of sodium citrate dissolved in water was added. The mixture was heated to 100° C for 30 min and then allowed to cool naturally. The resulting solution was dried at 80° C for 6 hours.



Figure S8. (a, c, e) SEM images of metal oxide nanostructures: (a) NiO, (c) CuO, and (e) MnO₂. (b, d, f) SEM images of Au NPs-coated metal oxide nanostructures: (b) Au/NiO, (d) Au/CuO, and (f) Au/MnO₂.



Figure S9. XRD patterns of metal oxide (in black) and Au/metal oxide (in red): (a) NiO and Au/NiO, (b) CuO and Au/CuO, (c) MnO and Au/MnO₂



Figure S10. Cyclic voltammograms at a scan rate of 50 mV/s for (a) NiO and Au/NiO, (b) CuO and Au/CuO, and (c) MnO₂, Au/ MnO₂.



Figure S11. SEM image and X-ray elemental mapping of Au/Ni(OH)₂ composite; (a) SEM image and elemental mapping of (b) nickel (Ni), (c) oxygen (O), and (d) gold (Au).