Supplementary Information

Transparent *p*-Type Epitaxial Thin Films of Nickel Oxide

Pengfei Zhai,[a] Qinghua Yi,[a] Jie Jian,[b] Haiyan Wang,[b] Pingyuan Song,[a] Chao Dong,[c] Xin Lu,[a] Yinghui Sun,[a] Jie Zhao,[a] Xiao Dai,[a] Yanhui Lou[a], Hao Yang[a] and Guifu Zou*[a]

[a]*School of Physical Science and Technology, School of Energy, Soochow University, Suzhou 215000, PRC*
[b]*Department of Electrical and Computer Engineering, Texas A & M University, College Station, TX 77843, USA*
[c]*Department of Chemistry and Chemical Biology, The University of New Mexico, Albuquerque, NM 87131, USA*
*Corresponding author
E-mail: zouguifu@suda.edu.cn

EXPERIMENTAL DETAILS

Preparation of epitaxial NiO thin films

Epitaxial NiO thin films were grown on (0001) Al_2O_3 substrates by PAD. The precursor used nickel chloride hexahydrate ($NiCl_2 \cdot 6H_2O$, Aldrich) as a source of Ni. In a typical experiment: 3.10 g of polyethylenimine (PEI, average Mw~25,000 by LS, average Mn~10,000 by GPC, branched, Aldrich) was dissolved in 50 ml deionized water. Then, 1.54 g $NiCl_2 \cdot 6H_2O$ was added into the solution under stirring vigorously at room temperature for 6 h. The precursor solution was then diluted to 180 ml with deionized water and filtered using an Amicon ultrafiltration unit containing an ultrafiltration membrane designed to pass materials having a molecular weight of less than about 3000 g mol^{-1} while retaining the desired materials of a larger size. After ultrafiltration, the solution was concentrated to 25 ml in volume. Inductively coupled

plasma-atomic emission spectroscopy (ICP-AES, PerkinElmer Optima 8000) showed that the final solution had 14.5 mg ml^{-1} of Ni. The precursor was spin-coated onto (0001) Al_2O_3 substrates at 3500 revolutions per minute for 30 s. Then, the precursor films were heated at 500 °C for 4 h in an ambient air. The thicker film of NiO film can be grown by coating layer by layer. Although it is hard to gain the thicker epitaxial film due to growing layer by layer, the crystallinity of the thick NiO film is still high. Meanwhile, the reproducibility of PAD is good to grow the epitaxial thin films on the base of large quantities of experience. In addition, we have operated PAD to design and synthesize a wide range of materials (including metals, metal-oxides, metal-nitrides, metal-carbides, and their derived composites) due to nature of the solution workup.

Characterization

The crystal structures of resultant NiO thin films were characterized by X-ray diffraction (XRD, Riguku D/MAX-2000PC). High resolution transmission electron microscopy analysis (HRTEM, FEI TECNAI G2 F20) was also performed to study the microstructures. The TEM samples were prepared using a standard cross-section sample preparation procedure, including manual grinding, polishing, dimpling and a final ion milling step (PIPS 691 precision ion polishing system, 3.7 keV). Field-emission scanning electron microscopy (FESEM, HTTACHI SU-8010) and atomic force microscopy (AFM, Asylum Research MFP-3D-BIO) were performed to investigate the surface morphology. The optical transmission was measured by ultraviolet-visible (UV-Vis) spectrometer (Shimadzu UV-2450). Hall and resistivity were measured by physical property measurement system (PPMS, Quantum Design PPMS-9T). NiO standards were characterized by X-ray photoemission spectroscopy (XPS, Kratos AXIS ULTRA HSA). XPS data were taken with a Al Kα source . Peak deconvolution and analysis was performed in XPSPEAK 4.1 software. The adventitious C 1s peak was referenced to 284.6 eV.

Electronic Supplementary Material (ESI) for Chemical Communications

XPS measurement

(a) and (b) show the XPS spectrum for the Ni $2p_{3/2}$ state and O 1s states, respectively.