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Fabrication of Polypyrrole Conductive Matrix covered MnNi₂O₄ Nanocomposite as the Positive Electrode Material in Asymmetric Supercapacitors

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1. Materials used

Pyrrole monomer, cetyltrimethyl ammonium bromide (CTAB) and potassium peroxodisulphate (PDS) were received from Merck. $Ni(NO_3)_2.6H_2O$, Manganese acetate and ethanol (C₂H₅OH) were purchased from SRL (India). Acetylene black, polyvinylidene fluoride (PVDF) and N-methyl-2-pyrrolidone (NMP) were received from Merck.

2. Synthesis of MnNi₂O₄

Manganese nickel oxide was synthesized via a simple co-precipitation method. First, 0.4 M of Ni(NO₃)₂.6H₂O and 0.2 M of manganese acetate were dissolved separately in 50 ml of water. After that, 0.2 M of manganese acetate was added drop by drop to the 0.4 M Ni(NO₃)₂.6H₂O solution at 60 °C for 15 min., then 0.5 M NH₄OH was added drop by drop for an hour under stirred conditions. The precipitate was filtered and washed several times with

water and ethanol, and dried at 80 °C for 12 h. Finally, the precipitate was calcined at 350 °C for 3 h. to get a dark-brown colored MnNi₂O₄.

3. Synthesis of Polypyrrole (PPY)

 $25 \text{ ml} \text{ of } 0.5 \text{ MH}_2\text{SO}_4$, 200 ml of double distilled water, and 0.01 g of CTAB were taken in a beaker and stirred for 1 h in a magnetic stirrer. Then, the beaker was kept in an ice bath, and 5 ml of pyrrole was added, and stirring was continued. To this mixture, 20 ml of 0.8 M of $K_2S_2O_8$ was added dropwise till black coloration developed. Then, the reaction mixture was kept in a stirrer for 6 h. under cold conditions. Further, the product was neutralized with water and filtered. The dried precipitate was powdered and used for further analysis.

4. Materials characterization

The phase configuration and surface functionalities of the as-prepared samples were performed using Bruker (XRD, Rigaku D/maxB, DMX-2200) and Jasco FT-IR-4600 spectroscopy, respectively. The microscopic analysis of the as-prepared samples was done by using Hitachi S-2400 scanning electron microscope equipped with an energy dispersive spectrometer (EDS), and the surface chemistry of the MnNi₂O₄/PPY composite was performed using ESCA/Auger Laboratory X-ray photoelectron spectroscopy. Supercapacitor measurements were performed using the Autolab (AUT51770) workstation.



Fig. S1: CV curves of MnNi₂O₄, PPY and MnNi₂O₄/PPY composite at a scan rate of 5 mV/s in 1 M KOH.



Fig. S2: (A) N₂ adsorption-desorption isotherm curve and (B) corresponding pore size distribution curve of MnNi₂O₄/PPY composite.



Fig. S3: Nyquist plot of (A)MnNi₂O₄, (B)PPY and (C) MnNi₂O₄/PPY composite with fittings.