

Transition Metal Oxide Electrode Materials

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The rising use of nonrenewable fossil fuels in recent decades has put human existence in grave danger. As a result, it is imperative to design environmentally friendly and cost-effective energy storage devices.

Supercapacitors are a promising energy device because of their high power density, outstanding cycle stability, and quick charge/discharge process. However, supercapacitors' energy density is still lower than that of conventional batteries'. Supercapacitors' electrochemical performance is heavily influenced by the electrode materials, as is well-known to everyone.

metal oxides

Transition Metal oxides

Electrode Materials

Energy materials

1. Co_3O_4

The Cobalt oxide belongs to the spinel family and theoretical capacity of Co_3O_4 is found to be 3560 F/g [1] and moreover it is cheap and environment friendly compound along with excellent durability and stability. However, the capacitance is varied lot in many applications from theoretical capacitance value. The less conductivity, high volume expansion and contraction, slow kinetics and particle aggregation are the reasons behind this variation in capacitance [2][3].

2. Co_3O_4 Nanoparticles

The big surface area and large active sites are the two major advantages of nanoparticles/Naonoaterials. The ion diffusion path is one of the favourable conditions as diffusion is easy due to very short pathway. Morphology and dimension are important factors behind the maximum capacitance of nanomaterials. A different type of Co_3O_4 materials were reported so far such as nano fibres,sheets and wires and nanoparticles. Yue et al synthesisd Co_3O_4 / rGO composite by simple environment friendly hydrothermal method [4]. A 3-D structure of small Co_3O_4 material is allowed to disperse on rGO flakes to show excellent electrochemical activity than Co_3O_4 nanowires. Indira-priyadarshini et al prepared Co_3O_4 / rGO-120-12 and which achieved capacitance of 1152 F/g at 1 A/g of current rate [5]. By using PVDF (polyvinylidene fluoride), carbon and Co_3O_4 nanoparticle in the rao of 1:1:8, slurry was made and coated on nickel foam to get capacitance of 761 F/g at 11 mA/cm² [6]. Here adding PVDF hinders the capacitance of material because it causes decrease in the interaction between the active material and collector, which tend to result in the less number of active sites and hence charge transfer rate. On the other hand, growing Co_3O_4 NPs directly on the metal foam, carbon sheets and carbon cloth can eliminate the need for binders and conducting materials [7][8]. Moreover, Ag- Co_3O_4 nanosheets grown on Ni foam exhibits the excellent capacitance of 1323 F/g at 10A /g current rate and which shows retention of 104% of initial capacitance at 2000 cycles.

3. Synthesis of Co_3O_4 Nanomaterials

Metal organic frameworks / MOFs type of materials have gained popularity in recent years in applications such as electro catalysis, adsorption of gas, degradation of pollutants, energy devices and so on. MOFs are also regarded to be an excellent template for the creation of Co_3O_4 nanoparticles because of their tunable porosity structure, variable pore size distribution, and large surface area [9][10]. The Zeolitic Imidazolate Frameworks 67 is acts as a precursor to synthesise Co_3O_4 NPs and Co_3O_4 material is converted by calcinations method to gain a good capacity of 190 F/g at 5 A/g. α -Co/Ni(OH)₂ nanocages. The composite α -Co/Ni(OH)₂@ Co_3O_4 -70 prepared by Bao et al exhibits a large number reactive sites including good charge diffusion channels, because of this reason it shows excellent capacity of 1000 F/g at 1A/g current rate [1]. The addition of active carbon to α -Co/Ni(OH)₂ increases the capacity retention of 72.3 % at current density of 10 A/g. In addition it delivers 0.075 kW/kg and 23.88 Wh/kg of power and energy density respectively [11]. In another way, Wei et al developed a process where thermal treatment converts ZIF-67 into ultrathin Co_3O_4 nanoparticles. A very good results in oxygen evolution reactions of 2D- Co_3O_4 ultrathin nanomaterials is because of its Tafel slope value of 74 mV/dec and potential of 230 mV. The 3D porous carbon developed by Li et al shows low specific capacitance of 423 F/g at current rate of 1 A/g. The low capacitance of material is due to the usage of 3D graphene / Co-metal organic framework (MOF) as a precursor, which slows down the transportation of electrons between electrolyte and active material [11].

4. MnO_2

MnO_2 has been thoroughly investigated as the high efficient TMO due to its abundant natural occurrence, lack of environmental pollution, and higher theoretical specific capacitance (1380 F/g) [12]. The MnO_2 material is limited in supercapacitor applications; this is because of very less charge transfer rate [13].

5. Carbon@ MnO_2

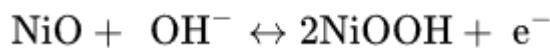
The huge surface area and great electrical conductivity is the key factor behind the large use of carbonaceous materials like graphene, carbon nanofibres, carbon nanotubes and carbon nanowires. In order to achieve a good capacity of material, there should be very less path difference between the electrolyte and electrode surface, which can be seen in carbon materials [14]. The N-doped hollow HNC@ MnO_2 3D cores shell synthesised by Cai and co workers exhibits specific capacitance of 247 F/g at 0.5 A/g current rate [15]. Long and co workers prepared δ - MnO_2 on carbon cloth and exhibited excellent power and energy density(Asymmetric device) of 1198 W/kg and 49 Wh/kg respectively. Lei et al developed MnO_2 nanosheet@CNT framework through chemical vapour deposition method.

Commercial carbon compounds, on the other hand, are prohibitively expensive and difficult to prepare on a big scale due to their high cost and complicated preparation process. As a result, developing low-cost and renewable materials is critical in order to meet rising demand [16]. Biomass is a renewable resource with a large value of usage. Yang et al prepared MnO_2 /biomass-based porous carbon via hydrothermal approach by using banana peel

as a carbon source, and which shows 139 F/g of specific capacitance at 300 mAh/g of current density and 70 F/g at current rate of 10 A/g [17][18].

6. Nickel Oxide

Nickel oxide (NiO) has got a huge amount of value in latest years because of its unique properties in terms of heat, light, electricity, sound, catalysis and magnetism properties [19]. As a result of their environmental friendliness and huge availability, they are often employed in the areas of supercapacitors. Because it has two or more oxidation states, it promotes rapid redox reactions, which contributes in storage techniques that are in charge. At 0.5 V potential window, Nickel oxide has exhibits theoretical capacitance of 2584 F/g [20]. Unfortunately, due to NiO₂'s low electrical conductivity of 0.01 to 0.32 Sm⁻¹ [21], the experimental findings never achieve the theoretical capacitance because it can expand which leads to destroying the active materials and causing electrical contact damage [22]. As of now, the SC values for NiO-type electrodes including a nanostructure and SSA have been 50 to 1776 F/g [23]. The link between NiO and NiOOH is described using two primary hypotheses. The energy storage mechanism occurs between NiO and NiOOH in one model, whereas the other, NiO converted to Ni(OH)₂ in the influence of an alkaline medium, resulting from Ni(OH)₂ and NiOOH reactions like given below [24][25].



The existence of Gr produces the core shell semicoated NiO/Ni structure, as seen in **Figure 1**. Furthermore, it has an unusually high Csp (2048.3 F/g at 1.0 A/g), as well as exceptional cyclic stability (77.8%) and retention in capacity after 10000 cycles at a current rate of 50 A/g [26].

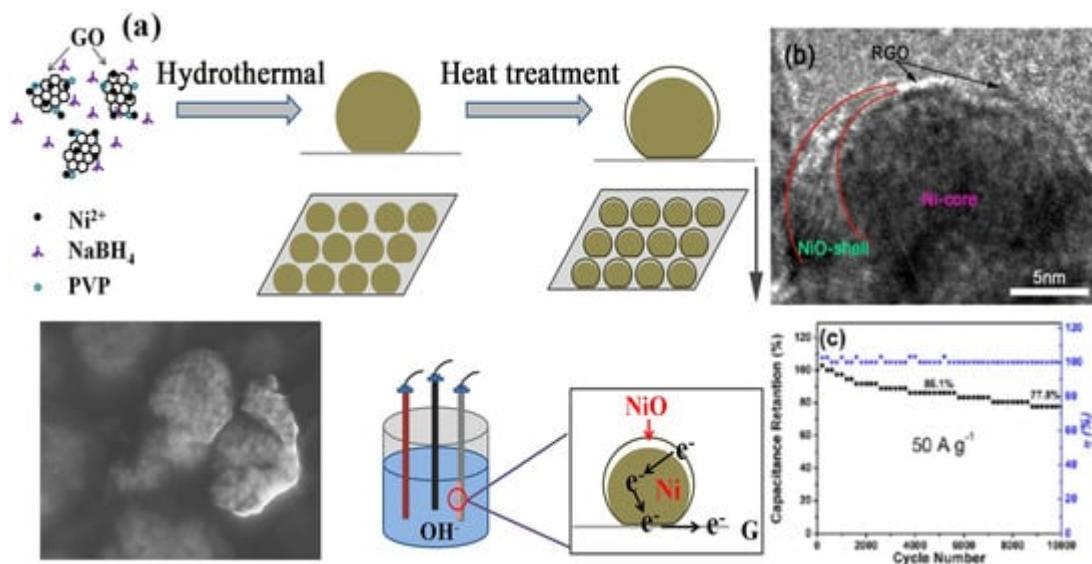


Figure 1. (a) Schematic representation of Conducive mechanism of material. (b) HRTEM micrographs of NiO/Ni/RGO. (c) Columbic efficiency at current rate of 50 A/g. Figure reprinted from ref. . The Author(s) licensed under CC BY 4.0.

7. Copper Oxide

Copper oxides, like CuO and Cu₂O-based SCs, have gained a lot of attention because of their abundance, low cost, nontoxicity, and ease of synthesis of diverse nanostructures [27]. Further, the capacity (loading storage) was diminished by very less electrical and cyclic abilities [28]. For example, Zhang et al reported the fabrication of flower-like CuO in a KOH electrolyte, yielding a specific capacitance of 133.6 F/g [29][30], whereas Li and co workers focused on developing CuO nanostructures immediately onto Cu foam surfaces, yielding a capacitance of 212 F/g in the same electrolyte [4][31]. To achieve a higher C_{sp} of 569 F g⁻¹, Wang et al constructed CuO nanosheet arrays on Ni foam surface; yet, the synthesis approach is a pretty tricky approach with a very low yield [32][33][34].

8. Battery Type MOs (Metal Oxides)

Incorporating battery-type MO's with MnO₂-type of electrodes has been regarded to be an important technique to boost the energy density and capacity of supercapacitors [35][36], according to Nie's work. Synergistic effect, redox reactions and battery metal oxides are the main reasons to improve the capacitance of supercapacitors. In addition to that MnO₂@NiO exhibits very high capacitance of 1277 F/g at 10 A/g with retention of 76% even after 10,000 cycles [37][38]. The Co₃O₄ on MnO₂ shows extraordinary performance by exhibiting 616 F/g at 2 A/g current rates and moreover it was achieved 83% of capacity retention after 10,000 cycles. In other hand, AS electrode of Co₃O₄@ MnO₂CC90 exhibits energy density and power density of 54 Wh/kg 1 kW/kg respectively [39].

9. ZnO

The specific capacity, stability and all other aspects of supercapacitors are controlled by choosing the good and capable material as an electrode. As a result, researchers have been investigating electrode materials that perform well electrochemically [40]. Because of their good theoretical capacitance, strong redox activity and affordable prices, TMOs have earned a lot of interest. ZnO has the features of environment friendly, wide availability, and constant capacitance [41]. Dhivya Angelin and co workers modify the ZnO by doping it with Zirconium, an appreciable capacitance of 518 F/g at 1 A/g was achieved in 9 Wt% Zr-ZnO nanoparticle and capacity retention of 94% even after 5000 cycles is achieved. ZnO nanomembranes exhibit different capacities in different electrolytes, like 846 F/g in 6 M KOH, 465 F/g in 1 M KCl, 65 F/g in 6 M Na₂SO₄ each at 1 A/g of current densities [42].

10. ZnO Composites

Various types of ZnO composites are synthesised as supercapacitor electrodes, such as metal oxide-Zinc oxide, polymer-Zinc oxide, carbon-Zinc oxide to find out the most suitable material for electrochemical studies [43]. Graphene nanocapsules (GNCs) shows excellent capacitance of 194 F/g at 20 A/g current rate and moreover only 2.6% of capacity loss is found even after 15,000 cycles [44]. Chebrolu et al synthesised ZnO/NiO electrode and which exhibits extraordinary capacitance of 1248 F/g at 8 mA/cm² than ZnO/PbO, ZnO/FeO, ZnO/CuO electrode materials. The reason behind this is, uniform surface area of nanosheets including good electrical conductivity. To

avoid the distraction of ZnO framework, Di's team synthesised ZnO with little quantity of Al_2O_3 . The specific capacitance of 463 F/g with excellent stability of 96% is achieved. Later all studies proves that multicomponent compounds along with ZnO could increase the capacity and stability. The $\text{CoO}_3\text{-CuO-ZnO} @ \text{GO}$ nanocomposite prepared by Obodo et al and which delivered excellent Csp of 1950 F/g at 10 mV/s current density.

11. XCo_2O_4 (X-Cu, Ni, Mn)

The spinel structure exhibits good electrochemical activity and conductivity so these ternary transition materials used extensively for supercapacitor studies [45][46]. The MnCo_2O_4 electrode synthesised by shanmugavadivel et al by combustion method shows an excellent capacitance of 270 F/g. Moreover, electrodeposition method followed to prepare the same material shows increased Csp of 585 F/g at 0.2 mA/cm² current rate [47]. Later, by electrodeposition method NiCo_2O_4 is coated on nickel wire and which exhibits good capacitance of 315 C/g at 1 A/g with loss of 8.4% capacity after 50,000 cycles [48][49]. CuCo_2O_4 developed by Pawar et al by electrodeposition and annealing process [50]. An appreciable capacity of 1473 F/g even after 5,000 cycles is achieved at 1A/g current density [51]. The Csp of 1933 F/g at current rate of 1 A/g was achieved by Wang's group by synthesising MnCo_2O_4 electrode material [52]. The $\text{NiCo}_2\text{O}_4@ \text{MnO}_2$ synthesised by Xu and co-workers exhibits a good specific capacitance of 1.23 F/cm⁻² at 50 mA/cm⁻² after 8000 cycles.

12. Transition Metal Molybdates

Due to a redox mechanism on materials surface, pseudocapacitive materials such as organic conductive materials and metal oxides show large specific capacitances than carbon materials [53]. Furthermore, due to large number of active sites, quick redox reactions ternary metal oxide could be the potential material to replace RuO_2 . The transition metal molybdates have got a lot of interest as main spot of mixed transition metal oxides because of their features such as abundant availability, higher specific, theoretical capacitance, and low prices [54][55]. The morphology and structure of supercapacitor electrode materials play a big role in their performance. As a result, it's critical to create electrode materials with distinct spatial structural features. The large specific surface area leads to the improved interfacial conductivity, increase in the number of active sites and porous structure, the Ag Quantum Dots/ NiMoO_4 nanopartilcles shows excellent specific capacitance of 3342 F/g at voltage of 1 mV/s and 2074 F/g at current density of 1 A/g as shown in Figure-2 [56].

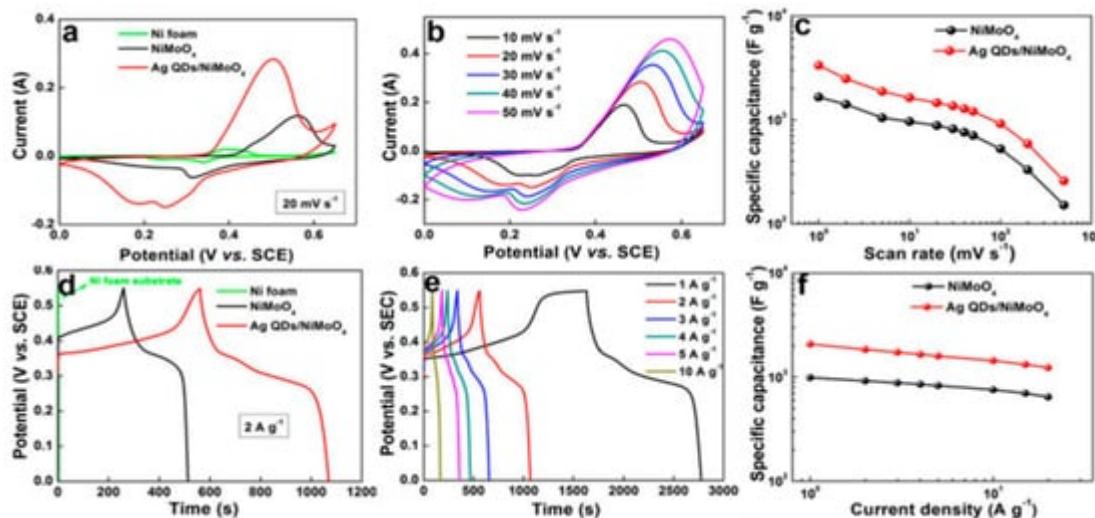


Figure 2. The three-electrode system showing Ag QDs/NiMoO₄ and NiMoO₄ materials CV and GCD measurements (a) CV curves at 20 mV/s; (b) CV curves of Ag Quantum Dots/NiMoO₄ electrode; (c) specific capacitances Vs scan rate measurements; (d) GCD curves at 2 A/g; (e) rate capabilities of electrodes and (f) specific capacitances Vs current densities. Reproduced with permission from ref. . Copyright 2020 Elsevier.

13. Design of Transition Metal Oxides

The construction of hetero type of composites has proven to be a promising technique to execute the electrochemical characteristics of TMOs, as per Yi's research [55]. Heterostructures include core–shell structures. The core–shell configuration can provide a lot of surface area with a lot of porosity. Likewise, core materials enhance electron transmission, while shell materials provide electrochemical redox active sites. Furthermore, each material's synergistic effect is used to boost the electrochemical behaviour of the electrode. 3D hierarchical core–shell (CoMoO₄@CoS) was successively developed by Xuan and team, here they took rGO/Ni foam for the preparation. **Figure-3** displays different magnifications of SEM images of CoS, CoMoO₄ and CoMoO₄@CoS materials [57].

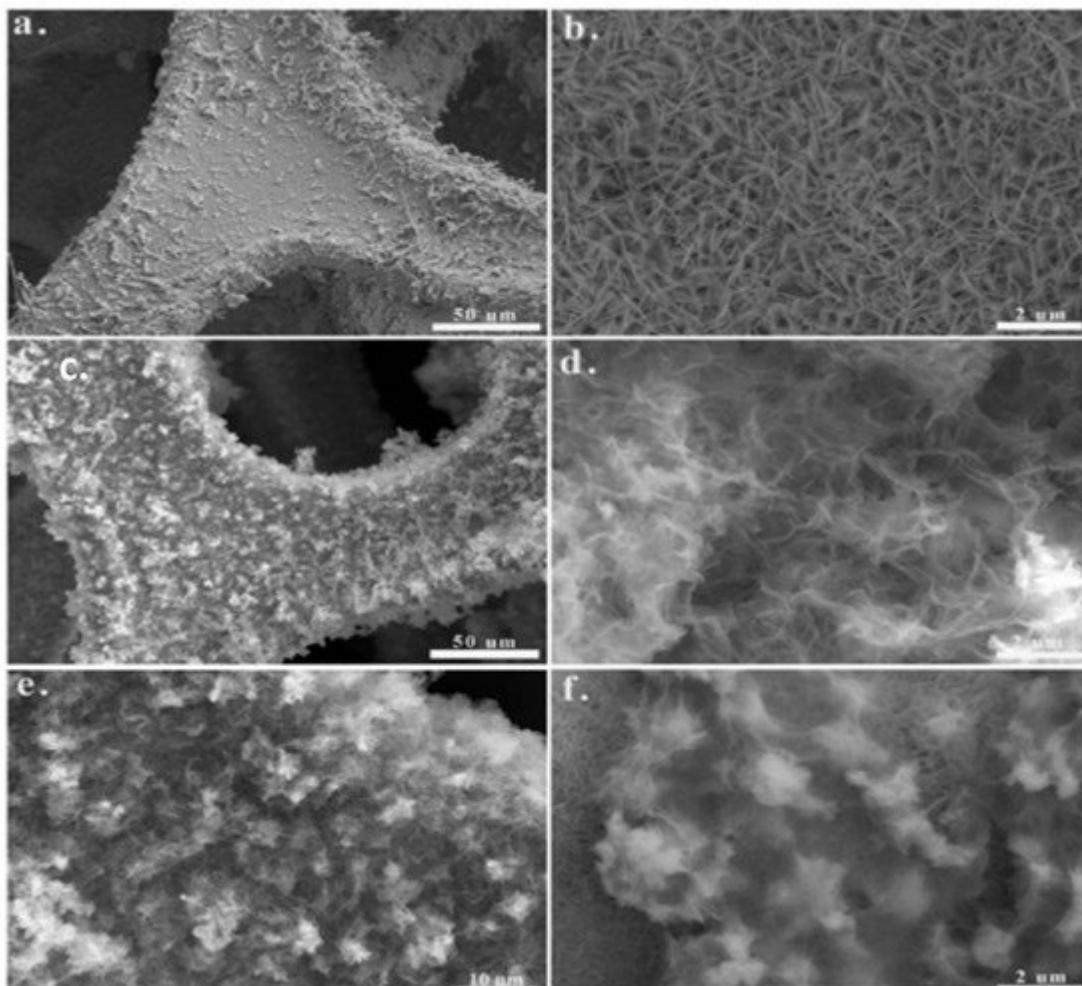


Figure 3. (a,b) Different magnification micrographs of CoMoO_4 ; (c,d) SEM micrographs at two magnifications of CoS ; (e,f) SEM micrographs of $\text{CoMoO}_4@\text{CoS}$ with different magnification. Reproduces with permission from ref. . Copyright 2020 Elsevier.

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