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A brief summary of recent developments of cathode electrode materials in lithium-ion batteries

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Abstract— Lithium-ion batteries have played an important role in large energy storage applications such as electric vehicles and portable devices due to their high energy density. Due to growing demands for better performance batteries with superior cycling stability and higher energy density, various investigations were performed on finding new cathode materials to satisfy those requirements. This review will first go over the current challenges of lithium-ion batteries and briefly outline some recent developments in carbon-based nanomaterials and low-Co Ni-based layered oxide cathodes. Research on various carbon materials coating such as single/multi-walled carbon nanotubes, reduced graphene oxide will be presented. Furthermore, this review will summarize the advancements in low-Co layered oxide and their reported performances. Lastly, the perspectives of future advancements in cathode materials will be presented.

Keywords— lithium-ion batteries, carbon nanomaterials, high-Ni layered oxide, cathodes, specific capacity

I. Introduction

The high consumption in fossil fuels has not only quickly depleted this natural resource but also created multiple concerns for the environment. One of the most promising methods to solve the above-mentioned issues is electrochemical energy storage (EES) technique, which can reduce the disproportion between energy demand and energy production. Today, with the development in portable devices and their high demand in specific energy, many parameters are needed to be considered in the research of EES systems, namely longer cycle life, high power and energy density, and environmental friendliness [1–4]. Among several energy storage systems, rechargeable lithium-based batteries such as Li-ion (LIBs) received significant attention due to their safety and fast charging capacity. LIBs are known to have high energy density, low weight, long cycle and shelf life [5–7]. However, the excellent performance of lithium-ion batteries is highly dependent on the electrode materials, in which electrode materials with high electrical, ionic conductivity, and superior electrochemical performance are required.

Currently, different electrode materials are being implemented in LIBs, namely carbon-based materials, transition metal oxides (TMOs) and mixed transition metal oxide (MTMOs). Carbon-based materials such as graphene, graphene oxide, and carbon nanotubes (CNTs) are suitable materials for LIBs owing to their high conductivity, porosity, and chemical stability [8]. As for TMOs and MTMOs, their high theoretical capacity can greatly improve the performance of LIBs [9].

Since 2020, various novel nanostructured cathode materials have been developed and used as electrode material in lithium-based batteries. Specifically, about 1470 research articles were published, in which 944 of them are related to LIBs while the second most common lithium-based batteries (lithium-sulfur batteries - LSBs) are accounted for 311 articles (Fig. 1). For example, Sanchez et.al. electrophoretic coated $LiFePO_4$/graphene oxide onto carbon fibers as cathode in lithium-ion batteries and achieved a high-capacity retention of 88.1% at 1 C over 300 cycles [10]. Molybdenum disulfide compound nanoflakes co-doped with nitrogen and carbon also shows potentials in LIBs, in which a high retention percentage of 90% was achieved after 3000 charge/discharge cycles [11]. With all the recent developments in lithium-ion batteries cathode material and the plethora of recent studies on LIBs, we consequently believe that a review article to go over the main research breakthroughs in lithium-ion batteries cathode materials is necessary.

This review will first summarize the working principle of LIBs, including their major drawbacks of using traditional cathodes. Subsequently, we will summarize the latest developments in various carbon materials and nanostructured TMOs and MTMOs and their performance in LIBs. Lastly, we summarize the current research developments in flexible electrode materials and its future potential challenges.

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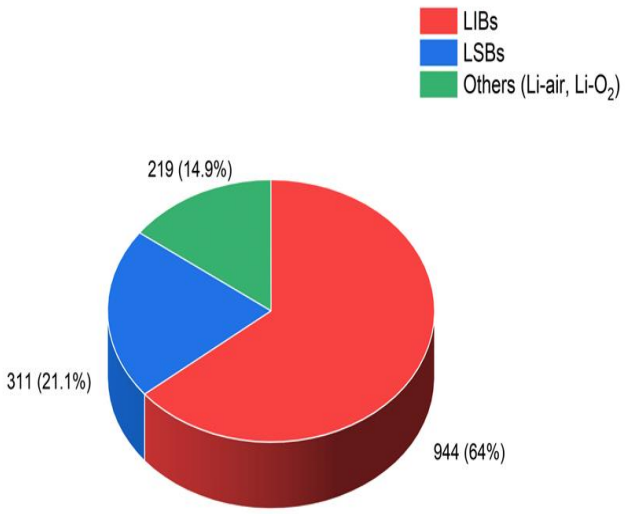

Fig. 1: Publication of cathode nanomaterials in lithium-based batteries since 2020 (indexed by EBSCO Information Services Database)

II. Principles of Lithium-Ion Batteries

After their first introduction by Sony in 1991, LIBs have emerged as the major energy storage system in portable devices due to their high energy density and rechargeable capabilities [12–14]. Typically, LIBs consist of graphite anode (theoretical capacity of 372 mAh g^{-1} [15,16]) and lithium oxides or phosphate ($LiCoO_2$, $LiFePO_4$) as cathode [17,18]. The devices also comprised of organic carbonate electrolyte and a permeable separator, which allow free ionic movement between anode and cathode in the batteries. During the charging process of LIBs, Li^+ ion is separated from the cathode and move through the electrolyte to be inserted into the porous structure of the anode [19]. Conversely, during the discharge process, LIBs release their stored energy from the anode back to the cathode. The chemical mechanism in LIBs is represented in Equations 1 and 2 [20]. A schematic representation of this process can be seen in Fig. 2.

$$Anode: 6C + xLi^+ + xe^- \leftrightarrow LiC_6 \qquad (1)$$

$$Cathode: LiCoO_2 \leftrightarrow Li_{1-x}CoO_2 + xe^- + xLi^+ \qquad (2)$$

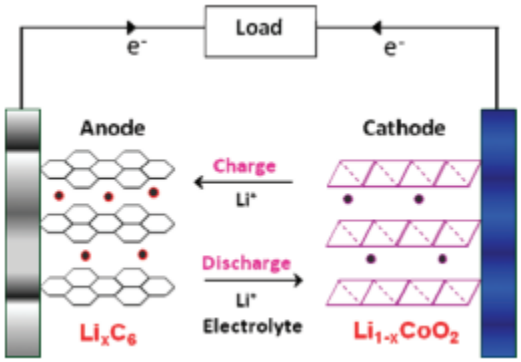

Fig. 2: Schematic representation of lithium-ion battery. Reprinted with permission from [21]. Copyright 2011 American Chemical Society

III. Current Limitations of Lithium-Ion Batteries and Roles of Nanomaterials to Resolve These Limitations

Electrode materials issues are one of the main problems in LIBs. For traditional $LiCoO_2$ cathode, although it has a high ionic and electrical conductivity, the overlapping of $Co^{3+/4+}$:3d band with top of the O^{2-} :2p band limits the theoretical capacity of $LiCoO_2$ to 50% [21]. Moreover, the morphology changes of electrode materials during lithium ion insertion and extraction can cause

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reduction in cycle life and destruction of cell operation. Furthermore, the change in morphology can cause exposure of fresh electrode material directly with the electrolyte, which generate solid electrolyte interface film (SEI), resulting in loss of capacity [22,23]. Beside the change in morphological properties, LIBs electrode materials have been reported to form crystal structures during the cycling process, resulting in poor electrical conductivity and capacity [24,25]. The other problem of LIBs is the sudden volume increase during the alloying and dealloying of Li^+, causing electrode strain and isolation of electrode materials from current collector (short circuit, which result in capacity fading and poor cycle life. Lastly, since LIBs performance is strongly depended on the heat generated during operation and internal impedance, there is a risk of thermal runaway during electrochemical reactions [26].

A. Potential solutions from nanomaterials

Various characteristics of nanomaterials such as high surface area, good electrical and thermal conductivities can positively impact the performance of LIBs. The high surface area in nanomaterials can improve the exposed area between electrode materials and electrolyte, resulting in more redox site and higher power and energy density [27]. In lithium-ion batteries specifically, more exposed area can result in more sites for Li insertion channels and shortened path for Li^+ diffusion [28]. The superior thermal and electrical conductivities of nanomaterials can facilitate ion transport between the redox sites, which can enhance the cycle life and capacitance [29,30]. Moreover, since nanomaterials can evolve in different structure (i.e., 0 D, 1 D, 2 D, 3 D), they can be utilized appropriately based on the advantages and disadvantages of each dimension. Lastly, nanomaterials are known for their mechanical stiffness, which can withstand structural damage during the cycling process [31].

IV. Recent Developments of Nanomaterials As Cathode In Lithium-Ion Batteries

A. Carbon nanocomposites

Researchers have focused on the incorporation single-walled and multi-walled carbon nanotubes (SWCNTs and MWCNTs) as conductive additive for cathode in LIBs due to their unique one-dimension tubular structure, superior surface area, good chemical stability, and mechanical reliability [32–34]. For instance, Aliahmad et.al. utilized pure V_2O_5 as electrode materials in LIBs and reported a 175 mAh g^{-1} . However, when V_2O_5 was anchored with SWCNT, a superior specific capacity of 390 mAh g^{-1} was reported. As for rate test performance, V_2O_5-SWCNT composite exhibits a higher specific capacity at high current densities as compared to pure V_2O_5 (158 vs. 31 mAh g^{-1} at 10 C) (Fig. 3e). Moreover, the electronic and ionic conductivities of V_2O_5-SWCNT composite is recorded to be higher than pure V_2O_5. Internal resistance for V_2O_5-SWCNT also dropped from 414.8 to 120.7 Ω (Fig. 3f). These results show that the conductive network of the V_2O_5-anchored SWCNT composite can resist cycling strain, improving Li^+ diffusion rate and strain relaxation of the electrode [35].

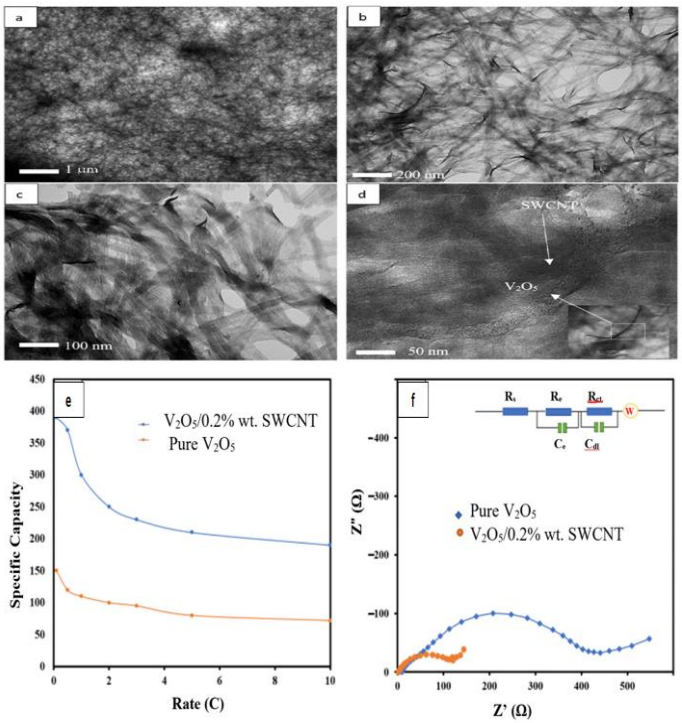

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Fig. 3: Transmission electron microscopy (TEM) image of V_2O_5-SWCNT structure at a) low magnification, b) high magnification, c) and d) V_2O_5 sandwiched structure between SWCNT; e) Rate-performance at different C rates, f) Nyquist plot of pristine V_2O_5 and V_2O_5/0.2% wt. SWCNT cells. Reproduced with permission from [35]. Copyright 2022 Multidisciplinary Digital Publishing Institute.

Furthermore, in 2021, Perumal et.al. investigated the wrapping of MWCNT on $LiMn_2O_4$ as an attempt to resist the capacity fading over multiple cycles of $LiMn_2O_4$. To prepare the $LiMn_2O_4$/MWCNT nanocomposite, predetermined ratios of lithium acetate, manganese acetate and citric acid were dissolved in 50 mL distilled water. The mixture was then evaporated and preheated for the gelation process. Following that, it was heated to 800°C to form spinel $LiMn_2O_4$ nanopowder. The nanopowder was then mixed with N-methyl-2-pyrrolidone and MWCNT, in which the mixture was stirred for several hours. Finally, the homogeneous solution was thermally treated at 400°C and $LiMn_2O_4$/MWCNT nanocomposite can be obtained after this step. When this material was subjected to field emission scanning electron microscopy (FESEM) analysis, particles with uniform sizes are distributed uniformly over the surface as compared to the non-uniform distribution in spinel $LiMn_2O_4$. The above materials are used as cathode electrode materials for the coin cell fabrication of LIB, in which their electrochemical performance was tested. Although both pure spinel $LiMn_2O_4$ and $LiMn_2O_4$/MWCNT shows the same Li^+ insertion and extraction at potential window of 3.4 to 4.2 V, the MWCNT wrapped $LiMn_2O_4$ coin cell exhibited a higher discharge capacity of 122 mAh g^{-1} as compared to the pure $LiMn_2O_4$ (108 mAh g^{-1}). It is hypothesized that the enhanced performance of $LiMn_2O_4$/MWCNT is due to the large number of Li^+ storage site on the surface of MWCNT and structural disorganization [36]. $LiMn_2O_4$ carbon composite also observed to have a great cyclic retention of 93% after 100 cycles, which is due to the smaller grain impedance as compared to spinel $LiMn_2O_4$. Overall, $LiMn_2O_4$/MWCNT nanocomposite is a suitable cathode electrode material in LIBs [37].

Other than SWCNT/MWCNT, other carbonaceous materials such as graphene also attract researchers' attention. With its honeycomb 2-D lattice structure consists of carbon atoms in sp^2 hybridization, graphene possesses a great mechanical strength, high theoretical surface area (2600 m^2/g), and great mechanical strength (~1060 GPa). These properties help graphene to form an electrically active surface that can properly be in contact with the electrolyte [38].

Although possessing numerous benefits, graphene is not suitable to be used in conjunction with metal oxides due to the inert surface of graphene can prevent uniform composites formation with the surface of metal oxides. To solve this, reduced graphene oxide (rGO) is introduced since the oxygen-containing functional groups can bind with the oxide interfaces. Moreover, rGO possesses a high conductivity, which enhance the long-term cyclic stability of LIBs [39,40]. In 2021, Zhang et.al. synthesized rGO/Cu-V_2O_5 composites and compared its performance with bare Cu-V_2O_5 material. The composites were synthesized via a simple solid-state chemical route followed by a heated reflux process with very low fraction of rGO (less than 3%) and a high content of Cu-V_2O_5. Using X-ray photoelectron spectroscopy (XPS), the analysis of O 1s and C 1s bands have confirmed the formation of rGO in rGO/Cu-V_2O_5 composites. Upon investigation under SEM imaging, the composites is observed to exhibit an irregular lamellar structure while TEM imaging showed multiple superimposed thin layers, with the crystal lattice spacing is 0.218 nm in the composites layer. The electrochemical performance of the material was studied to analyze the effect of rGO on Cu-V_2O_5, in which the specific discharge capacity of rGO/Cu-V_2O_5 composites observed a value of 203.6 mAh g^{-1} and a capacity retention rate of 82.5% as compared to 168.2 mAh g^{-1} specific discharge capacity and 59% capacity retention rate for the Cu-V_2O_5 sample [41]. rGO was also used to form phosphorus carbon composites, in which Chen et.al. used in situ phosphorization of Cu and carbonization of rGO to synthesize Cu_3P@rGO composite. This electrode material displayed a specific capacity of 2330 mAh g^{-1} after 183 cycles at 500 mA g^{-1} [42]. These results have shown that by the combination of rGO as electrode material can have positive effects on the performance of LIBs.

Other graphene-like structure such as graphene nanosheets were also reported to improve the specific capacity of metal oxides in LIBs. Gu et.al. synthesized 2-D layered lithium lanthanum titanium oxide (LLTO)/graphene nanosheet (C) composite via oxidative polymerization and carbonization processes. This composite exhibited a high specific capacity of 350 mAh g^{-1} for de-lithiation at 20 mA g^{-1} and high cycling stability with a capacity retention of 70% at 1 A g^{-1} after 800 cycles. As a comparison, the raw metal oxide material only observed a specific discharge capacity of 200 mAh g^{-1} at 20 mA g^{-1}. These results proved that LLTO/C composite not only exhibited perovskite behavior of LLTO but it also benefited from the improved electrical conductivity of graphene nanosheet [43].

In addition to the above-mentioned materials, Table 1 summarizes the electrochemical results of more carbon-based cathode materials in lithium-ion batteries. As shown in Table 1, carbon materials can be intercalated with different metal oxides such as $LiMnO_2$ [44], FeF_3 [45], and LFMP [46]. It can be observed that all materials show superior specific discharge capacity and cyclic stability, which proves the importance of carbon nanocomposites in the advancements of lithium-ion batteries. Moreover, an electrode material can contain more than one carbon structure such as rGO/CNT-CC (carbon cloth) to incorporate both structures' advantages such as high structural ability and maintain a moderate lithium-ion transmission distance [47].

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Table 1: Summary of recently reported carbon nanomaterials cathode for LIBs

Cathode materials	Capacity	Capacity retention	Reference
$LiMnO_2$/rGO	185.6 mAh g^{-1} at 100 mA g^{-1}	80.97% after 100 cycles at 100 mA g^{-1}	[44]
FeF_3/C/rGO	268 mAh g^{-1} at 0.04 A g^{-1}	96% after 50 cycles at 0.04 A g^{-1}	[45]
$LiFe_{0.2}Mn_{0.8}PO_4$/rGO	120 mAh g^{-1} at 0.2 C	93% after 200 cycles at 2 C	[46]
Boron-doped $LiFePO_4$@C	162.2 mAh g^{-1} at 0.1 C	95.2% after 200 cycles at 20 C	[48]
V_2O_5 microspheres/graphene	313.6 mAh g^{-1} at 0.5 C	68% after 200 cycles at 0.5 C	[49]
$V_2O_5.nH_2O$/rGO	268 mAh g^{-1} at 100 mA g^{-1}	55% after 500 cycles at 1000 mA g^{-1}	[50]
$WO_3.0.33H_2O$@rGO/CNT-CC	1504 mAh g^{-1} at 200 mA g^{-1}	90% after 400 cycles at 1000 mA g^{-1}	[47]
Perylene diimide (PI)/SWCNT	80 mAh g^{-1} at 1000 mA g^{-1}	76% after 2000 cycles at 500 mA g^{-1}	[51]
$LiMn_{0.8}Fe_{0.2}PO_4$/rGO/C	156.4 mAh g^{-1} at 0.05 C	95.5% after 300 cycles at 1 C	[52]

B. High nickel layered oxide cathodes

Since the introduction of $LiCoO_2$ in the 1980s by Goodenough's group, it has been a dominant cathode material in commercialized LIBs due to its high volumetric energy density and reliability [53–55]. With the successful implementation of $LiCoO_2$ cathode in portable electronic devices, it is highly expected that $LiCoO_2$ can lead to the mass production of electric vehicles (EVs). However, it is widely agreed that $LiCoO_2$ has yet to meet this expectation due to the heavy dependence on cobalt metal, which is an expensive rare earth metal that drive the cost of LIBs up significantly [56,57]. Nickel-rich and low cobalt layered ternary cathodes ($LiNi_xCo_yM_zO_2$, or NCM, where M is Mn or Al, $x + y + z = 1$) are promising alternative electrode materials in LIBs due to their high theoretical capacity and moderate cost of production [58]. It is worth noting that when the Ni content is limited ($x<0.5$), the cycling stability of the material increase but a weaker thermal-chemical stability is observed [59–62]. Hence, using the correct ratio of Ni and Co is important to maximize cyclic, thermal, and chemical stability while reducing the cost of production as low as possible. In 2021, Zhang et.al. developed single crystalline $LiNi_{0.6}Co_{0.1}Mn_{0.3}O_2$ layered cathodes (SC-NCM) by one-step calcination method, in which the reduced Ni/Co ratio coupling with the manganese Mn^{4+} ion substitution could reduce the cost of production and stabilize the NCM layered structure. SC-NCM also possesses several advantages as compared to conventional polycrystalline $LiNi_{0.6}Co_{0.1}Mn_{0.3}O_2$ layered cathodes (PC-NCM) such as the micron size particle can reduce the micron cracks and polarized overpotential for Li^+ diffusion on the surface [63,64]. The morphologies of SC-NCM and PC-NCM are confirmed under SEM, whereas PC-NCM exhibited micron spheres with 10-15 µm diameter while SC-NCM shows much smaller micron spheres of 2-5 µm diameter with no nanoparticle aggregation. When these cathode materials were fabricated into LIBs by coin cell apparatus, the capacity retention of SC-NCM is 90% after 150 cycles at 1 C as compared to 72.8% capacity retention in PC-NCM within voltage range of 2.75-4.3 V. Moreover, a polarization voltage value after 150 cycles of 265 mV was recorded in SC-NCM while PC-NCM showed a value of 652 mV. These results signify a smaller electrode polarization for SC-NCM cathode as compared

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to PC-NCM. Lastly, a full cell with SC-NCM cathode showed a high-capacity retention of 73.9% after 900 cycles at 45C, which shows its opportunity for applications at high temperature. It can be concluded that due to the crack-free structure of SC-NCM, the side reaction products and unexpected interfacial effects can be reduced, which is beneficial for practical applications in EVs [65].

Another nickel-rich layer oxide is reported by Shen et.al. in 2021. Using two-step synthesis method of hydroxide co-precipitation and lithiation calcination, the researchers generated $LiNi_{0.6}Co_{0.05}Mn_{0.35}O_2$ (NCM60535) material as cathode material in LIBs. This material possesses numerous benefits such as low cost due to low cobalt content, high capacity resulted from the nickel content, and improved stability with a relatively high amount of manganese. Upon XRD characterization of NCM60535, it was discovered that NCM60535 exhibited a low degree of cation mixing, which greatly affect the lithium diffusion. The material also shown spherical morphology, dense structures, and homogenous size distribution under SEM imaging while also exhibited an interplanar distance of 0.474 nm under high resolution TEM. Upon electrochemical evaluation, NCM60535 was found to have a high specific capacity of 156.1 mAh g^{-1} and a high-capacity retention of 95.8% for 100 cycles at 0.5 C. When this material was fabricated in conjunction with graphite as the anode material in LIB, the capacity of the battery can maintain at 161.2 mAh g^{-1} after 100 cycles at 0.5 C. Using an empirical model, its energy density is calculated to be 249 Wh kg^{-1}. These results have shown a great potential for NCM60535 as a cathode material for applications in high energy density LIBs [66].

In another study, Cui et.al. developed an ultra-high Ni cathode material with zero Co and Mn contents ($LiNi_{0.93}Al_{0.05}Ti_{0.01}Mg_{0.01}O_2$ – NATM) by metal hydroxide coprecipitation and calcination steps. The elimination of Co and Mn is due to the scarcity and toxicity of Co and the high dissolution tendency of Mn. The performance of NATM was compared with Co-containing (NC) and Mn-containing (NMC) cathode materials. Under SEM imaging, all NC, NMC, and NATM display spherical shapes with secondary particle size of 12 µm. Upon lowering the SEM magnification, all particles show uniform size distribution with no cracking by calcination (Fig. 4a and 4b). The materials were then fabricated in a coin half-cell configuration with a Li-metal anode to evaluate their electrochemical performance (Fig**.** 5a-d). At C/10 rate, NC, NMC, and NATM delivered a specific capacity of 236, 225, and 221 mAh g^{-1} , respectively. For the cycling stability at C/3 rate, NATM exhibited the highest capacity retention of 94% after 100 cycles as compared to 79% for NC and 90% for NMC. When the materials are paired with graphite anode and evaluated as a pouch full cell configuration (Fig**.** 5e-f), NATM can deliver 82% of its original capacity after 800 cycles between 2.5 and 4.25 V, which is higher than that of NC (52%) and NMC (60%). When evaluate the discharge curve at 25°C from the 1st to the 800th cycle at C/2 rate, the voltage plateaus of NATM are observed to remained intact after the 800th cycle, which prove a superior structure and surface stability of NATM as compared to NC and NMC. These results have shown that cheap and abundant metals such as Al, Mg, and Ti can be incorporated into ultra-high Ni-cathode materials, which can be a promising cathode material for LIBs [67].

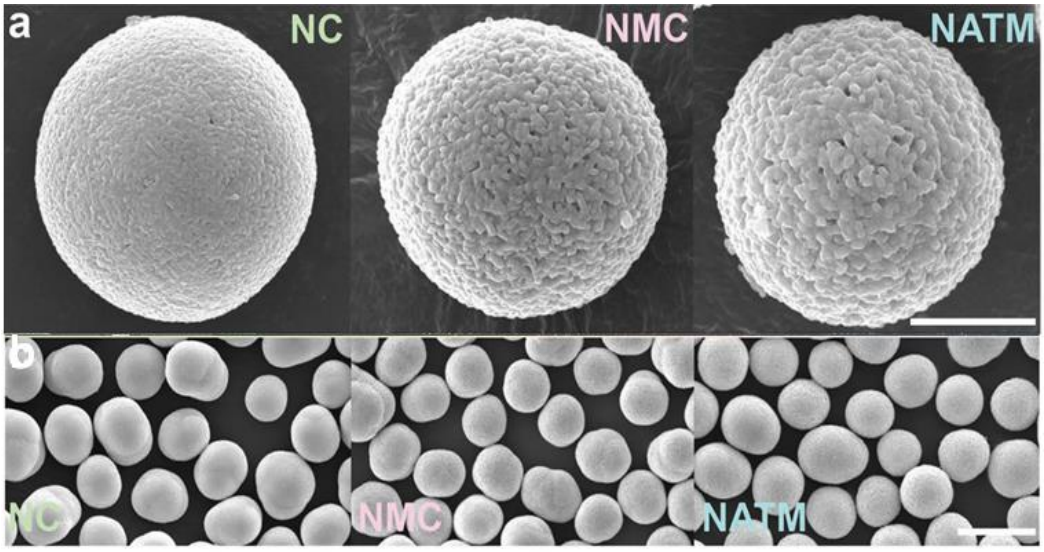

Fig. 4: Morphological and structure characterizations of NC, NMC, and NATM cathode materials: a) SEM images of a single secondary particle, b) SEM images at a low magnification. Reproduced with permission from [67]. Copyright 2021 John Wiley & Sons.

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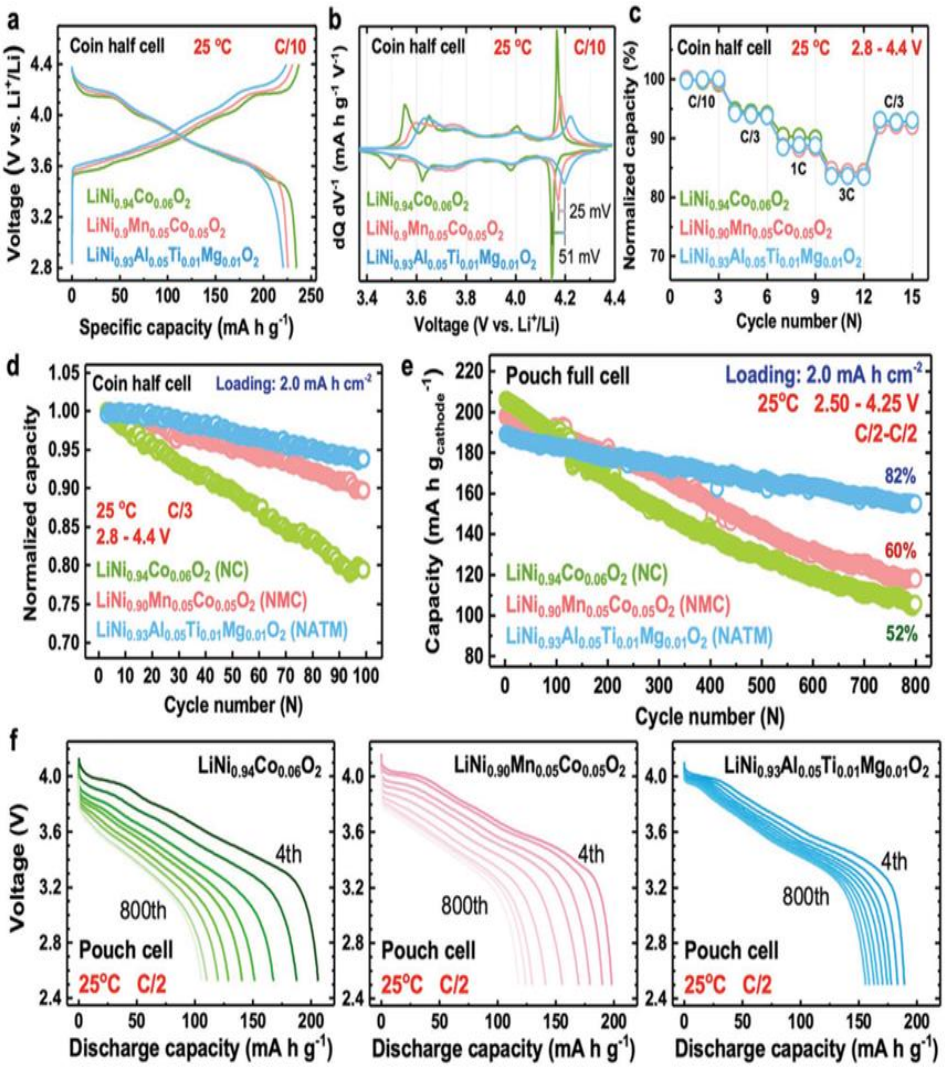

Fig. 5: Electrochemical performances of NC, NMC, and NATM cathodes in coin half cells paired with Li-metal anode and in pouch full cells. a) Charge and discharge curves and b) $dQ\ dV^{-1}$ curves of the 3rd C/10 formation cycle. c) Rate performance tests with a constant C/5 charge rate. Cyclability evaluation in d) coin half cells for 100 cycles at a C/3 rate between 2.8 and 4.4 V and in e) pouch full cells for 800 cycles at a C/2 rate between 2.5 and 4.25 V. f) Evolution of discharge curves every 100 cycles (i.e., 4th cycle, 100th cycle, 200th cycle, etc.) at a C/2 rate. 1C equals 180 mA g^{-1}. Reproduced with permission from [67]. Copyright 2021 John Wiley & Sons.

Furthermore, Xi et.al. also reported a development cobalt-free and high-nickel layered oxide cathode ($LiNi_{0.90}Mn_{0.06}Al_{0.04}O_2$ – NMA) by organic amine coprecipitation method. The electrochemical evaluation of the material showed that it can provide a discharge capacity of 223.1 mAh g^{-1} at 0.1 C in voltage window of 2.5-4.3 V. NMA also exhibited a higher discharge voltage of 47 and 17 mV as compared to NC and NMC, respectively. Moreover, when increasing the voltage window from 2.5-4.5 V, its discharge capacity increased to 232.1 mAh g^{-1} with a capacity retention of 93.3 % at 0.5 C after 100 cycles, which is superior to the 66.9% capacity retention in NC. These promising results showed that NMA is a great candidate for high-Ni and Co-free cathode material for next-generation LIBs [68].

Table 2 summarizes more recently developed layered oxide cathode materials for LIBs. In general, researchers are currently limiting the amount of cobalt in LIBs cathode while maintaining a competitive performance as compared to NC. Researchers also utilized surface stabilizing agent such as 1,2-propyleneglycol sulfite (PGS) in Ni-rich layered oxide, which exhibited a superior performance for the material as compared PGS-absence layered oxide (Fig. 6) [69]. Overall, these studies are guided towards materials with lower cost and higher energy density for next-generation lithium-ion batteries.

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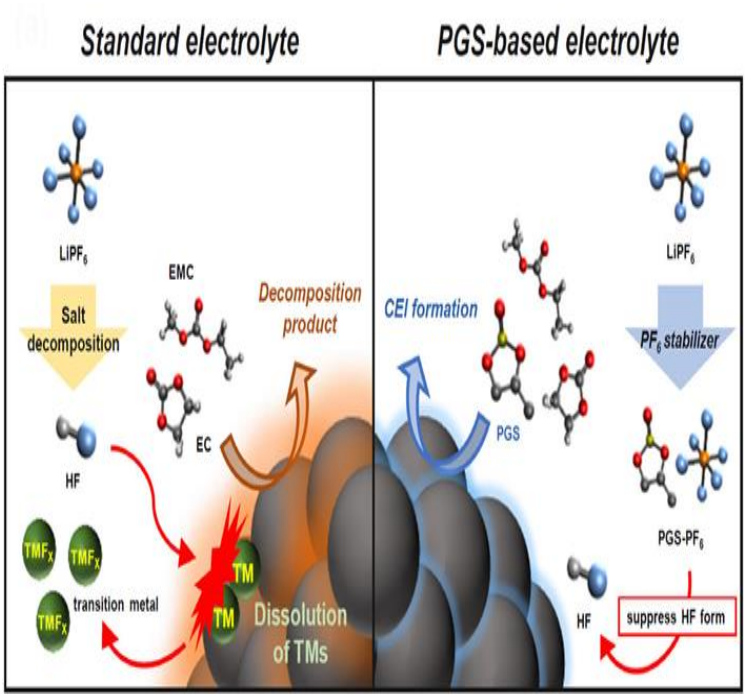

Fig. 6: The role of PGS additive in LIB cells. Reproduced with permission from [69]. Copyright 2022 John Wiley & Sons.

Table 2: Summary of recently reported high-Ni layered oxide cathode for LIBs

Materials	Specific capacity	Capacity retention	References
PGS-stabilizing agent NCM	199.3 mAh g^{-1} from 3.5 to 4.3 V for 0.5 wt.% PGS additive	76.9 % for 2.0 wt.% PGS additive	[69]
Ni-rich Mg-substitute NMC	187.2 mAh g^{-1} at 0.1 C	80% after 350-600 cycles, depends on Mg content	[70]
Li-rich $Li_2Ni_{0.2}Ru_{0.4}O_3$	210 mAh g^{-1} at 100 mA g^{-1}	82% after 20 cycles	[71]

V. Conclusions and Perspectives

In summary, this review briefly summarizes various recent developments of carbon-based and nickel layered oxide cathode nanomaterials in lithium-ion batteries. Carbon-based nanomaterials are widely used due to their high electronic conductivity, superior stability, which can greatly improve the performance of LIBs. When carbon was coated, it created a protective layer which is capable of preventing the dissolution of cathode and electrolyte degradation. Moreover, carbon coating can also restrict the conductive ion/electron transfer path between the active cathode materials and electrolyte [72]. When coating with different cathode materials, carbon can positively affect their performance in different ways. For example, the addition of carbon in $LiMn_xFe_{1-x}PO_4$ formed a 3-D conductive network to enhance the rapid electron migration, which improves the electrochemical performance of the material [73–75]. As for V_2O_5, graphene oxide can comfortably bind with the oxide interface, so that the vanadium composites' long-term cycling stability can be enhanced due to the high conductivity of the carbon material [76]. Lastly, carbon can reduce the phase transition of $LiMnO_2$ in charge-discharging cycles, which prevent the irreversible loss of circulation capability [77,78].

Furthermore, a growing number of researchers are currently focusing on the development of Ni-rich cathode materials. Conventional NC or NMC have proved to be one of the most suitable materials for electric vehicles due to their high specific capacity of 200 mAh g^{-1} at high working voltage (3.8 V) [79–81]. In these materials, cobalt metal has an important role in improving the rate performance while limiting the Li^+/Ni^{2+} mixing in NMC and NC. However, due to the rising prices and limited resources, coupling with the fact that cobalt is extremely toxic, numerous studies have been concentrated on low-Co or Co-free high Ni-based materials such as $LiNi_{0.90}Mn_{0.06}Al_{0.04}O_2$ [68], $LiNi_{0.96}Mg_{0.02}Ti_{0.02}O_2$ [82], $LiNi_{0.93}Al_{0.05}Ti_{0.01}Mg_{0.01}O_2$ [67], $LiNi_{0.91}Co_{0.05}Mn_{0.05}O_2$

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[83], and $LiNi_{0.94}Co_{0.04}Zn_{0.02}O_{1.99}$ [84]. These materials are hoped to play an important role in the development of high-energy density lithium-ion batteries.

Although various breakthroughs have been achieved in the production of cathode materials with superior specific capacity, more work needs to be done to further seeking batteries with higher energy density, longer cycle life and better thermal stability. As for carbon-based nanomaterials, more analysis needs to be done to incorporate more renewable carbon from a variety of feedstock such as agriculture residue or industrial wastes. The impact of low-Co or Co-free cathode materials and their trade-off also needed to be studied better. Lastly, it is important to analyze the affinity between the newly developed cathodes with different anodes and electrolyte in LIBs. In the future, studies on next-generation LIBs materials should focus on low energy-intensive processes to satisfy large scale production with affordable cost.

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References

1. Phan, T.N.; Gong, M.K.; Thangavel, R.; Lee, Y.S.; Ko, C.H. Ordered Mesoporous Carbon CMK-8 Cathodes for High-Power and Long-Cycle Life Sodium Hybrid Capacitors. *J Alloys Compd* **2018**, *743*, 639–645, doi:10.1016/j.jallcom.2018.02.007.
2. Wang, L.; Guo, M.; Zhan, J.; Jiao, X.; Chen, D.; Wang, T. A New Design of an Electrochromic Energy Storage Device with High Capacity, Long Cycle Lifetime and Multicolor Display. *J Mater Chem A Mater* **2020**, *8*, 17098–17105, doi:10.1039/D0TA04824K.
3. Akinwolemiwa, B.; Chen, G. Fundamental Consideration for Electrochemical Engineering of Supercapattery. *J Braz Chem Soc* **2018**, doi:10.21577/0103-5053.20180010.
4. Hasan, M.W.; Huynh, K.; Soha, T.; Rubayet, J.; Wasi, S. Investigation of a Cow Urine-Based Power Generation Technology Model for a Cost-Effective and Sustainable Energy Source. *Int J Innov Appl Stud* **2023**, *38*, 779–793.
5. Xie, J.; Lu, Y.-C. A Retrospective on Lithium-Ion Batteries. *Nat Commun* **2020**, *11*, 2499, doi:10.1038/s41467-020-16259-9.
6. Chen, Y.; Kang, Y.; Zhao, Y.; Wang, L.; Liu, J.; Li, Y.; Liang, Z.; He, X.; Li, X.; Tavajohi, N.; et al. A Review of Lithium-Ion Battery Safety Concerns: The Issues, Strategies, and Testing Standards. *Journal of Energy Chemistry* **2021**, *59*, 83–99, doi:10.1016/j.jechem.2020.10.017.
7. Xiong, R.; Pan, Y.; Shen, W.; Li, H.; Sun, F. Lithium-Ion Battery Aging Mechanisms and Diagnosis Method for Automotive Applications: Recent Advances and Perspectives. *Renewable and Sustainable Energy Reviews* **2020**, *131*, 110048, doi:10.1016/j.rser.2020.110048.
8. Kumar, R.; Sahoo, S.; Joanni, E.; Singh, R.K.; Tan, W.K.; Kar, K.K.; Matsuda, A. Recent Progress in the Synthesis of Graphene and Derived Materials for next Generation Electrodes of High Performance Lithium Ion Batteries. *Prog Energy Combust Sci* **2019**, *75*, 100786, doi:10.1016/j.pecs.2019.100786.
9. Chikkannanavar, S.B.; Kim, J.H.; Jung, W. Degradation and Life Performance of Transition Metal Oxide Cathodes Used in Lithium- <scp>I</Scp> on Batteries. In *Transition Metal Oxides for Electrochemical Energy Storage*; Wiley, 2022; pp. 257–272.
10. Sanchez, J.S.; Xu, J.; Xia, Z.; Sun, J.; Asp, L.E.; Palermo, V. Electrophoretic Coating of LiFePO4/Graphene Oxide on Carbon Fibers as Cathode Electrodes for Structural Lithium Ion Batteries. *Compos Sci Technol* **2021**, *208*, 108768, doi:10.1016/j.compscitech.2021.108768.
11. Fayed, M.G.; Attia, S.Y.; Barakat, Y.F.; El-Shereafy, E.E.; Rashad, M.M.; Mohamed, S.G. Carbon and Nitrogen Co-Doped MoS2 Nanoflakes as an Electrode Material for Lithium-Ion Batteries and Supercapacitors. *Sustainable Materials and Technologies* **2021**, *29*, e00306, doi:10.1016/j.susmat.2021.e00306.
12. Blomgren, G.E. The Development and Future of Lithium Ion Batteries. *J Electrochem Soc* **2017**, *164*, A5019–A5025, doi:10.1149/2.0251701jes.
13. Schipper, F.; Erickson, E.M.; Erk, C.; Shin, J.-Y.; Chesneau, F.F.; Aurbach, D. Review—Recent Advances and Remaining Challenges for Lithium Ion Battery Cathodes. *J Electrochem Soc* **2017**, *164*, A6220–A6228, doi:10.1149/2.0351701jes.
14. Jin, Y.; Zhu, B.; Lu, Z.; Liu, N.; Zhu, J. Challenges and Recent Progress in the Development of Si Anodes for Lithium-Ion Battery. *Adv Energy Mater* **2017**, *7*, 1700715, doi:10.1002/aenm.201700715.

ISSN No. 2454-6194 | DOI: 10.51584/IJRIAS | Volume VIII Issue II February 2023

15. Lung-Hao Hu, B.; Wu, F.-Y.; Lin, C.-T.; Khlobystov, A.N.; Li, L.-J. Graphene-Modified LiFePO4 Cathode for Lithium Ion Battery beyond Theoretical Capacity. *Nat Commun* **2013**, *4*, 1687, doi:10.1038/ncomms2705.
16. Eftekhari, A. On the Theoretical Capacity/Energy of Lithium Batteries and Their Counterparts. *ACS Sustain Chem Eng* **2019**, *7*, 3684–3687, doi:10.1021/acssuschemeng.7b04330.
17. Jiang, Y.; Zhao, H.; Yue, L.; Liang, J.; Li, T.; Liu, Q.; Luo, Y.; Kong, X.; Lu, S.; Shi, X.; et al. Recent Advances in Lithium-Based Batteries Using Metal Organic Frameworks as Electrode Materials. *Electrochem commun* **2021**, *122*, 106881, doi:10.1016/j.elecom.2020.106881.
18. Lyu, Y.; Wu, X.; Wang, K.; Feng, Z.; Cheng, T.; Liu, Y.; Wang, M.; Chen, R.; Xu, L.; Zhou, J.; et al. An Overview on the Advances of $LiCoO_2$ Cathodes for Lithium-Ion Batteries. *Adv Energy Mater* **2021**, *11*, 2000982, doi:10.1002/aenm.202000982.
19. Mekonnen, Y.; Sundararajan, A.; Sarwat, A.I. A Review of Cathode and Anode Materials for Lithium-Ion Batteries. In Proceedings of the SoutheastCon 2016; IEEE, March 2016; pp. 1–6.
20. Mahmood, N.; Hou, Y. Electrode Nanostructures in Lithium-Based Batteries. *Advanced Science* **2014**, *1*, 1400012, doi:10.1002/advs.201400012.
21. Manthiram, A. Materials Challenges and Opportunities of Lithium Ion Batteries. *J Phys Chem Lett* **2011**, *2*, 176–184, doi:10.1021/jz1015422.
22. Shin, J.; Ryu, W.-H.; Park, K.-S.; Kim, I.-D. Morphological Evolution of Carbon Nanofibers Encapsulating SnCo Alloys and Its Effect on Growth of the Solid Electrolyte Interphase Layer. *ACS Nano* **2013**, *7*, 7330–7341, doi:10.1021/nn403003b.
23. Hong, Y.J.; Son, M.Y.; Kang, Y.C. One-Pot Facile Synthesis of Double-Shelled SnO_2 Yolk-Shell-Structured Powders by Continuous Process as Anode Materials for Li-Ion Batteries. *Advanced Materials* **2013**, *25*, 2279–2283, doi:10.1002/adma.201204506.
24. Zhou, S.; Mei, T.; Wang, X.; Qian, Y. Crystal Structural Design of Exposed Planes: Express Channels, High-Rate Capability Cathodes for Lithium-Ion Batteries. *Nanoscale* **2018**, *10*, 17435–17455, doi:10.1039/C8NR04842H.
25. Shi, F.; Song, Z.; Ross, P.N.; Somorjai, G.A.; Ritchie, R.O.; Komvopoulos, K. Failure Mechanisms of Single-Crystal Silicon Electrodes in Lithium-Ion Batteries. *Nat Commun* **2016**, *7*, 11886, doi:10.1038/ncomms11886.
26. Feng, X.; Ren, D.; He, X.; Ouyang, M. Mitigating Thermal Runaway of Lithium-Ion Batteries. *Joule* **2020**, *4*, 743–770, doi:10.1016/j.joule.2020.02.010.
27. Su, X.; Wu, Q.; Li, J.; Xiao, X.; Lott, A.; Lu, W.; Sheldon, B.W.; Wu, J. Silicon-Based Nanomaterials for Lithium-Ion Batteries: A Review. *Adv Energy Mater* **2014**, *4*, 1300882, doi:10.1002/aenm.201300882.
28. Liu, B.; Zhang, J.-G.; Shen, G. Pursuing Two-Dimensional Nanomaterials for Flexible Lithium-Ion Batteries. *Nano Today* **2016**, *11*, 82–97, doi:10.1016/j.nantod.2016.02.003.
29. Higgins, T.M.; Park, S.-H.; King, P.J.; Zhang, C. (John); McEvoy, N.; Berner, N.C.; Daly, D.; Shmeliov, A.; Khan, U.; Duesberg, G.; et al. A Commercial Conducting Polymer as Both Binder and Conductive Additive for Silicon Nanoparticle-Based Lithium-Ion Battery Negative Electrodes. *ACS Nano* **2016**, *10*, 3702–3713, doi:10.1021/acsnano.6b00218.
30. Zhang, J.; Zhao, N.; Zhang, M.; Li, Y.; Chu, P.K.; Guo, X.; Di, Z.; Wang, X.; Li, H. Flexible and Ion-Conducting Membrane Electrolytes for Solid-State Lithium Batteries: Dispersion of Garnet Nanoparticles in Insulating Polyethylene Oxide. *Nano Energy* **2016**, *28*, 447–454, doi:10.1016/j.nanoen.2016.09.002.
31. Cheng, D.; Wu, P.; Wang, J.; Tang, X.; An, T.; Zhou, H.; Zhang, D.; Fan, T. Synergetic Pore Structure Optimization and Nitrogen Doping of 3D Porous Graphene for High Performance Lithium Sulfur Battery. *Carbon N Y* **2019**, *143*, 869–877, doi:10.1016/j.carbon.2018.11.032.
32. Liang, J.; Wang, S.; Yu, H.; Zhao, X.; Wang, H.; Tong, Y.; Tang, Q.; Liu, Y. Solution-Processed PDMS/SWCNT Porous Electrodes with High Mass Loading: Toward High Performance All-Stretchable-Component Lithium Ion Batteries. *Sustain Energy Fuels* **2020**, *4*, 2718–2726, doi:10.1039/C9SE01120J.
33. Mujahid, M.; Ullah Khan, R.; Mumtaz, M.; Mubasher; Soomro, S.A.; Ullah, S. NiFe2O4 Nanoparticles/MWCNTs Nanohybrid as Anode Material for Lithium-Ion Battery. *Ceram Int* **2019**, *45*, 8486–8493, doi:10.1016/j.ceramint.2019.01.160.
34. Nulu, A.; Nulu, V.; Sohn, K.Y. Silicon and Porous MWCNT Composite as High Capacity Anode for Lithium-Ion Batteries. *Korean Journal of Chemical Engineering* **2020**, *37*, 1795–1802, doi:10.1007/s11814-020-0559-5.
35. Aliahmad, N.; Biswas, P.K.; Dalir, H.; Agarwal, M. Synthesis of V2O5/Single-Walled Carbon Nanotubes Integrated into Nanostructured Composites as Cathode Materials in High Performance Lithium-Ion Batteries. *Energies (Basel)* **2022**, *15*, 552, doi:10.3390/en15020552.

36. Zhao, X.; Hayner, C.M.; Kung, H.H. Self-Assembled Lithium Manganese Oxide Nanoparticles on Carbon Nanotube or Graphene as High-Performance Cathode Material for Lithium-Ion Batteries. *J Mater Chem* **2011**, *21*, 17297, doi:10.1039/c1jm12373d.
37. Perumal, P.; Abhilash, K.P.; Selvin, P.C.; Sofer, Z. A Short Investigation on LiMn2O4 Wrapped with MWCNT as Composite Cathode for Lithium-Ion Batteries. *Bulletin of Materials Science* **2021**, *44*, 243, doi:10.1007/s12034-021-02532-0.
38. Geng, D.; Yang, H.Y. Recent Advances in Growth of Novel 2D Materials: Beyond Graphene and Transition Metal Dichalcogenides. *Advanced Materials* **2018**, *30*, 1800865, doi:10.1002/adma.201800865.
39. Qiu, B.; Zhao, X.; Xia, D. In Situ Synthesis of CoS2/RGO Nanocomposites with Enhanced Electrode Performance for Lithium-Ion Batteries. *J Alloys Compd* **2013**, *579*, 372–376, doi:10.1016/j.jallcom.2013.05.148.
40. Li, J.; Huang, J.; Li, J.; Cao, L.; Qi, H.; Cheng, Y.; Xi, Q.; Dang, H. Improved Li-Ion Diffusion Process in TiO2/RGO Anode for Lithium-Ion Battery. *J Alloys Compd* **2017**, *727*, 998–1005, doi:10.1016/j.jallcom.2017.08.121.
41. Zhang, H.; Rong, Y.; Cao, Y. Enhanced Cycling Stability of Cu-V2O5 Composited with Low-Fraction RGO as Cathodes for Lithium Ion Batteries. *J Alloys Compd* **2021**, *863*, 158761, doi:10.1016/j.jallcom.2021.158761.
42. Chen, H.; Gao, S.; Lan, X.; Wang, W.; Ke, B.; Lu, S.; Gu, C.; Li, G. Plasma Phosphorization of Self-Adaptive Electrode Cu3P@RGO for Lithium Ion Storage. *Vacuum* **2021**, *193*, 110537, doi:10.1016/j.vacuum.2021.110537.
43. Gu, B.; Zhan, C.; Liu, B.H.; Wang, G.; Zhang, Q.; Zhang, M.; Shen, Z. Two-Dimensional Layered Lithium Lanthanum Titanium Oxide/Graphene-like Composites as Electrodes for Lithium-Ion Batteries. *Dalton Transactions* **2022**, *51*, 7076–7083, doi:10.1039/D2DT00751G.
44. Tian, Y.; Qiu, Y.; Liu, Z.; Wei, X.; Cao, H. LiMnO $_2$ @rGO Nanocomposites for High-performance Lithium-ion Battery Cathodes. *Nanotechnology* **2021**, *32*, 015402, doi:10.1088/1361-6528/abb48c.
45. He, D.; Zhang, Y.; Cao, D.; Sun, M.; Xia, J.; Yang, Y.; Ding, Y.; Chen, H. A Flexible Free-Standing FeF3/Reduced Graphene Oxide Film as Cathode for Advanced Lithium-Ion Battery. *J Alloys Compd* **2022**, *909*, 164702, doi:10.1016/j.jallcom.2022.164702.
46. Zoller, F.; Böhm, D.; Luxa, J.; Döblinger, M.; Sofer, Z.; Semenenko, D.; Bein, T.; Fattakhova-Rohlfing, D. Freestanding LiFe0.2Mn0.8PO4/RGO Nanocomposites as High Energy Density Fast Charging Cathodes for Lithium-Ion Batteries. *Mater Today Energy* **2020**, *16*, 100416, doi:10.1016/j.mtener.2020.100416.
47. Lin, Y.; Sun, L.; Wang, J.; Shi, X.; Zhang, Y.; Gong, Y.; Sun, C.; Zhang, Y. RGO Wrapped Tungsten Trioxide Hydrate on CNT-Modified Carbon Cloth as Self-Supported High-Rate Lithium-Ion Battery Electrode. *Electrochim Acta* **2021**, *394*, 139162, doi:10.1016/j.electacta.2021.139162.
48. Li, C.; Yuan, H.; Yang, Z. Enhanced Electrochemical Performance of Boron-Doped Graphene-Decorated LiFePO4@C Cathode Material for Lithium-Ion Batteries. *Solid State Ion* **2020**, *352*, 115366, doi:10.1016/j.ssi.2020.115366.
49. Chen, A.; Li, C.; Zhang, C.; Li, W.; Yang, Q. The Mechanical Hybrid of V2O5 Microspheres/Graphene as an Excellent Cathode for Lithium-Ion Batteries. *Journal of Solid State Electrochemistry* **2022**, *26*, 729–738, doi:10.1007/s10008-021-05108-4.
50. Zhang, Y.; Yuan, X.; Lu, T.; Gong, Z.; Pan, L.; Guo, S. Hydrated Vanadium Pentoxide/Reduced Graphene Oxide Composite Cathode Material for High-Rate Lithium Ion Batteries. *J Colloid Interface Sci* **2021**, *585*, 347–354, doi:10.1016/j.jcis.2020.11.074.
51. Yang, P.; Xi, X.; Huang, T.; Zhong, Q.; Jiang, B.; Liu, R.; Wu, D. An Acid-Assisted Vacuum Filtration Approach towards Flexible PDI/SWCNT Cathodes for Highly Stable Organic Lithium Ion Batteries. *Electrochim Acta* **2020**, *338*, 135771, doi:10.1016/j.electacta.2020.135771.
52. Wang, Y.; Hu, G.; Cao, Y.; Peng, Z.; Lai, X.; Xie, X.; Du, K. Highly Atom-Economical and Environmentally Friendly Synthesis of LiMn0.8Fe0.2PO4/RGO/C Cathode Material for Lithium-Ion Batteries. *Electrochim Acta* **2020**, *354*, 136743, doi:10.1016/j.electacta.2020.136743.
53. Wang, B.; Lin, X.-Y.; Tang, Y.; Wang, Q.; Leung, M.K.H.; Lu, X.-Y. Recycling LiCoO2 with Methanesulfonic Acid for Regeneration of Lithium-Ion Battery Electrode Materials. *J Power Sources* **2019**, *436*, 226828, doi:10.1016/j.jpowsour.2019.226828.
54. Liu, Z.; Scott Cronin, J.; Chen-Wiegart, Y.K.; Wilson, J.R.; Yakal-Kremski, K.J.; Wang, J.; Faber, K.T.; Barnett, S.A. Three-Dimensional Morphological Measurements of LiCoO2 and LiCoO2/Li(Ni1/3Mn1/3Co1/3)O2 Lithium-Ion Battery Cathodes. *J Power Sources* **2013**, *227*, 267–274, doi:10.1016/j.jpowsour.2012.11.043.

ISSN No. 2454-6194 | DOI: 10.51584/IJRIAS | Volume VIII Issue II February 2023

55. Lyu, Y.; Wu, X.; Wang, K.; Feng, Z.; Cheng, T.; Liu, Y.; Wang, M.; Chen, R.; Xu, L.; Zhou, J.; et al. An Overview on the Advances of LiCoO 2 Cathodes for Lithium-Ion Batteries. *Adv Energy Mater* **2021**, *11*, 2000982, doi:10.1002/aenm.202000982.
56. Li, M.; Lu, J.; Chen, Z.; Amine, K. 30 Years of Lithium-Ion Batteries. *Advanced Materials* **2018**, *30*, 1800561, doi:10.1002/adma.201800561.
57. Kim, Y.; Seong, W.M.; Manthiram, A. Cobalt-Free, High-Nickel Layered Oxide Cathodes for Lithium-Ion Batteries: Progress, Challenges, and Perspectives. *Energy Storage Mater* **2021**, *34*, 250–259, doi:10.1016/j.ensm.2020.09.020.
58. Xiao, B.; Sun, X. Surface and Subsurface Reactions of Lithium Transition Metal Oxide Cathode Materials: An Overview of the Fundamental Origins and Remedying Approaches. *Adv Energy Mater* **2018**, *8*, 1802057, doi:10.1002/aenm.201802057.
59. Xu, G.; Liu, X.; Daali, A.; Amine, R.; Chen, Z.; Amine, K. Challenges and Strategies to Advance High-Energy Nickel-Rich Layered Lithium Transition Metal Oxide Cathodes for Harsh Operation. *Adv Funct Mater* **2020**, *30*, 2004748, doi:10.1002/adfm.202004748.
60. Wang, X.; Ding, Y.; Deng, Y.; Chen, Z. Ni-Rich/Co-Poor Layered Cathode for Automotive Li-Ion Batteries: Promises and Challenges. *Adv Energy Mater* **2020**, *10*, 1903864, doi:10.1002/aenm.201903864.
61. Voronina, N.; Sun, Y.-K.; Myung, S.-T. Co-Free Layered Cathode Materials for High Energy Density Lithium-Ion Batteries. *ACS Energy Lett* **2020**, *5*, 1814–1824, doi:10.1021/acsenergylett.0c00742.
62. Liao, B.; Li, H.; Xu, M.; Xing, L.; Liao, Y.; Ren, X.; Fan, W.; Yu, L.; Xu, K.; Li, W. Designing Low Impedance Interface Films Simultaneously on Anode and Cathode for High Energy Batteries. *Adv Energy Mater* **2018**, *8*, 1800802, doi:10.1002/aenm.201800802.
63. Bi, Y.; Tao, J.; Wu, Y.; Li, L.; Xu, Y.; Hu, E.; Wu, B.; Hu, J.; Wang, C.; Zhang, J.-G.; et al. Reversible Planar Gliding and Microcracking in a Single-Crystalline Ni-Rich Cathode. *Science (1979)* **2020**, *370*, 1313–1317, doi:10.1126/science.abc3167.
64. Xu, Q.; Li, X.; Kheimeh Sari, H.M.; Li, W.; Liu, W.; Hao, Y.; Qin, J.; Cao, B.; Xiao, W.; Xu, Y.; et al. Surface Engineering of LiNi0.8Mn0.1Co0.1O2 towards Boosting Lithium Storage: Bimetallic Oxides versus Monometallic Oxides. *Nano Energy* **2020**, *77*, 105034, doi:10.1016/j.nanoen.2020.105034.
65. Zhang, Z.; Bai, M.; Fan, X.; Yi, M.; Zhao, Y.; Zhang, J.; Hong, B.; Zhang, Z.; Hu, G.; Lai, Y. A Low Cost Single-Crystalline LiNi0.60Co0.10Mn0.30O2 Layered Cathode Enables Remarkable Cycling Performance of Lithium-Ion Batteries at Elevated Temperature. *J Power Sources* **2021**, *503*, 230028, doi:10.1016/j.jpowsour.2021.230028.
66. Shen, Y.; Xue, H.; Wang, S.; Wang, Z.; Zhang, D.; Yin, D.; Wang, L.; Cheng, Y. A Highly Promising High-Nickel Low-Cobalt Lithium Layered Oxide Cathode Material for High-Performance Lithium-Ion Batteries. *J Colloid Interface Sci* **2021**, *597*, 334–344, doi:10.1016/j.jcis.2021.04.008.
67. Cui, Z.; Xie, Q.; Manthiram, A. A Cobalt- and Manganese-Free High-Nickel Layered Oxide Cathode for Long-Life, Safer Lithium-Ion Batteries. *Adv Energy Mater* **2021**, *11*, 2102421, doi:10.1002/aenm.202102421.
68. Xi, R.; Zhang, J.; Lan, Z.; Yuan, Y.; Kang, J.; Li, Y.; Wang, J.; Zhang, C.; Hou, X. High-Nickel and Cobalt-Free Layered LiNi0.90Mn0.06Al0.04O2 Cathode for Lithium-Ion Batteries. *Ceram Int* **2022**, *48*, 36690–36696, doi:10.1016/j.ceramint.2022.08.228.
69. Jeon, Y.J.; Lee, J.; Han, Y.; Yim, T. 1, <scp>2-Propyleneglycol</Scp> Sulfite as a Surface Stabilizing Agent for <scp>Ni-rich</Scp> Layered Oxide Cathodes of <scp>lithium-ion</Scp> Batteries. *Int J Energy Res* **2022**, *46*, 19402–19413, doi:10.1002/er.8511.
70. Gomez-Martin, A.; Reissig, F.; Frankenstein, L.; Heidbüchel, M.; Winter, M.; Placke, T.; Schmuch, R. Magnesium Substitution in Ni-Rich NMC Layered Cathodes for High-Energy Lithium Ion Batteries. *Adv Energy Mater* **2022**, *12*, 2103045, doi:10.1002/aenm.202103045.
71. Ke, B.; Chu, S.; Li, J.-C.; Xu, X.; Yao, H.; Guo, S.; Zhou, H. A New Li-Rich Layered Cathode with Low Lattice Strain for Lithium-Ion Batteries. *Chemical Communications* **2022**, *58*, 10488–10491, doi:10.1039/D2CC04044A.
72. Zhou, L.; Yang, H.; Han, T.; Song, Y.; Yang, G.; Li, L. Carbon-Based Modification Materials for Lithium-Ion Battery Cathodes: Advances and Perspectives. *Front Chem* **2022**, *10*, doi:10.3389/fchem.2022.914930.
73. Guo, L.; Ren, L.; Wan, L.; Li, J. Heterogeneous Carbon/N-Doped Reduced Graphene Oxide Wrapping LiMn0.8Fe0.2PO4 Composite for Higher Performance of Lithium Ion Batteries. *Appl Surf Sci* **2019**, *476*, 513–520, doi:10.1016/j.apsusc.2018.12.227.

74. Ma, F.; Zhang, X.; He, P.; Zhang, X.; Wang, P.; Zhou, H. Synthesis of Hierarchical and Bridging Carbon-Coated LiMn 0.9 Fe 0.1 PO 4 Nanostructure as Cathode Material with Improved Performance for Lithium Ion Battery. *J Power Sources* **2017**, *359*, 408–414, doi:10.1016/j.jpowsour.2017.05.067.
75. Fu, Y.; Wei, Q.; Zhang, G.; Zhong, Y.; Moghimian, N.; Tong, X.; Sun, S. LiFePO4-Graphene Composites as High-Performance Cathodes for Lithium-Ion Batteries: The Impact of Size and Morphology of Graphene. *Materials* **2019**, *12*, 842, doi:10.3390/ma12060842.
76. Chen, J.; Li, W.; Jiang, J.; Wu, C.; Liu, Y. Facile and Creative Design of Hierarchical Vanadium Oxides@graphene Nanosheet Patterns. *RSC Adv* **2016**, *6*, 13323–13327, doi:10.1039/C5RA25521J.
77. Deng, Y.; Wu, Z.; Liang, R.; Jiang, Y.; Luo, D.; Yu, A.; Chen, Z. Layer-Based Heterostructured Cathodes for Lithium-Ion and Sodium-Ion Batteries. *Adv Funct Mater* **2019**, *29*, 1808522, doi:10.1002/adfm.201808522.
78. Tian, Y.; Qiu, Y.; Liu, Z.; Wei, X.; Cao, H. LiMnO $_2$ @rGO Nanocomposites for High-performance Lithium-ion Battery Cathodes. *Nanotechnology* **2021**, *32*, 015402, doi:10.1088/1361-6528/abb48c.
79. Xi, Y.; Wang, M.; Xu, L.; Kheimeh Sari, H.M.; Li, W.; Hu, J.; Cao, Y.; Chen, L.; Wang, L.; Pu, X.; et al. A New Co-Free Ni-Rich LiNi $_{0.8}$ Fe $_{0.1}$ Mn $_{0.1}$ O $_2$ Cathode for Low-Cost Li-Ion Batteries. *ACS Appl Mater Interfaces* **2021**, *13*, 57341–57349, doi:10.1021/acsami.1c18303.
80. Xu, H.; Cao, G.; Shen, Y.; Yu, Y.; Hu, J.; Wang, Z.; Shao, G. Enabling Argyrodite Sulfides as Superb Solid-State Electrolyte with Remarkable Interfacial Stability Against Electrodes. *ENERGY & ENVIRONMENTAL MATERIALS* **2022**, *5*, 852–864, doi:10.1002/eem2.12282.
81. Li, Y.; Zhang, Y.; Liu, J.; Bei, Y.; Che, J.; Song, Y.; Liu, L.; Ma, J. Inducing Favorable Dual-Substitution Reactivity by Doping Mo6+ and F− to Enhance Electrochemical Performance of LiNi0.6Co0.2Mn0.2O2 Cathode Materials. *Ceram Int* **2022**, *48*, 23016–23023, doi:10.1016/j.ceramint.2022.04.279.
82. Zhang, X.; Jia, H.; Zou, L.; Xu, Y.; Mu, L.; Yang, Z.; Engelhard, M.H.; Kim, J.-M.; Hu, J.; Matthews, B.E.; et al. Electrolyte Regulating toward Stabilization of Cobalt-Free Ultrahigh-Nickel Layered Oxide Cathode in Lithium-Ion Batteries. *ACS Energy Lett* **2021**, *6*, 1324–1332, doi:10.1021/acsenergylett.1c00374.
83. Qiu, Q.-Q.; Yuan, S.-S.; Bao, J.; Wang, Q.-C.; Yue, X.-Y.; Li, X.-L.; Wu, X.-J.; Zhou, Y.-N. Suppressing Irreversible Phase Transition and Enhancing Electrochemical Performance of Ni-Rich Layered Cathode LiNi0.9Co0.05Mn0.05O2 by Fluorine Substitution. *Journal of Energy Chemistry* **2021**, *61*, 574–581, doi:10.1016/j.jechem.2021.02.012.
84. Cui, Z.; Xie, Q.; Manthiram, A. Zinc-Doped High-Nickel, Low-Cobalt Layered Oxide Cathodes for High-Energy-Density Lithium-Ion Batteries. *ACS Appl Mater Interfaces* **2021**, *13*, 15324–15332, doi:10.1021/acsami.1c01824.